Radioactive Waste Management

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Stability and Buffering Capacity of the Geosphere for Long-term Isolation of Radioactive Waste

Application to Argillaceous Media

"Clay Club" Workshop Proceedings
Braunschweig, Germany
9-11 December 2003

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Hosted by
Gesellschaft für Anlagen- und Reaktorischerheit (GRS)mbH, Braunschweig

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NUCLEAR ENERGY AGENCY
ORGANISATION FOR ECONOMIC CO-OPERATION AND DEVELOPMENT

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NUCLEAR ENERGY AGENCY

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FOREWORD

A safety case for a geological repository for high-level and/or long-lived radioactive waste aims at conveying reasoned and complementary arguments to illustrate and instill confidence in the performances of the disposal system.

Potential geological host formations and their surroundings are chosen in particular for their long-term stability, their ability to accommodate the waste disposal facility, their ability to prevent or attenuate potential release of radioactivity (e.g. through retention capacities), and their buffering capacity vis-à-vis external and internal perturbations. Siting of potential disposal facilities is also carried out with an awareness of natural hazards. Honest recognition that no natural system is in equilibrium is required; the concept of “geosphere stability” does not therefore imply that steady state conditions are prevailing in the geosphere over (very) long periods of time. However, changes occur in many systems to an extent and at a rate such that their effects would not compromise deep disposal safety.

In building a safety case, it is therefore important to assess:

- the features, events and processes that could impact the evolution of the geosphere;
- the long-term stability of the favorable conditions displayed by the host formation;
- the buffering capacity of the formation vis-à-vis perturbations.

In this framework, a key issue is to evaluate the resilience of the main safety functions of the geosphere (including flow and transport properties) to natural perturbations. Thus, phenomenological evidence of persistence of those functions in past episodes of, for example, climatic changes, seismic activity, diagenetic evolution and burial/uplift, should enhance confidence in geosphere stability.

The NEA Integration Group for the Safety Case (IGSC) developed an initiative to establish the scientific basis for such stability and buffering capacity of deep geological waste management systems. Under this initiative, a first workshop was organised on 9-11 December 2003 in Braunschweig, Germany, under the auspices of the IGSC Working Group on the Characterisation, the Understanding and the Performance of Argillaceous Rocks as Repository Host Formations (usually named the "Clay Club") and devoted specifically to argillaceous settings. The present report synthesises the main outcomes of that workshop and presents a compilation of the related abstracts.

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INTRODUCTION

Disposal of high-level and/or long-lived radioactive waste in engineered facilities, or repositories, located underground in suitable geological formations, is being widely investigated world wide as a long-term management solution. This is in order to protect humans and the environment both now and in the future. From a quantitative point of view, a repository is said to be safe if it meets the relevant safety standards, such as internationally recommended or specified by the responsible national regulatory authorities. In recent years the scope of the safety assessment has broadened to include the collation of a broader range of evidence and arguments that complement and support the reliability of the results of quantitative analyses. The broader term "post-closure safety case", or simply "safety case",[1] is used to refer to these studies. It has also become evident that repository development will involve a number of step by step stages punctuated by interdependent decisions-making on whether and how to move from one stage to the subsequent one. These decisions require a clear and traceable presentation of technical and robust arguments that will help in giving confidence in the feasibility and safety of a proposed concept. The depth of understanding and technical information available to support decisions will increase from step to step. The safety case is a key input to support that decision to move to the next stage in repository development. It reflects the state and results of the research and development (R&D) undertaken at a certain stage, and supports decisions concerning future R&D efforts.

Potential geological host formations (and their surroundings) are chosen in particular for their long-term stability, their ability to accommodate the waste disposal facility, to prevent or to attenuate potential release of radioactivity, their buffering capacity vis-à-vis external and internal perturbations. Natural hazards are also considered in the site location choice for a potential disposal facility. It is recognised that no natural system is in equilibrium; the concept of "geosphere stability" does not therefore imply that steady state conditions are prevailing over very long periods of time. However, changes occur in many systems to an extent and at such rate that their effects would not compromise deep disposal safety.

Site characterisation and evaluation are important for determining the site suitability and the long-term safety of geological repositories of long-lived radioactive waste. Two previous NEA workshops already took into consideration "geosphere stability". The first one was held in Helsinki, September 1991 on "Long-term observation of the geological environment". It dealt particularly with the needs and techniques for long-term observations of the geological environment. At the workshop, it was considered that long-term observation programmes form an integral and crucial part of site qualification and confirmation for deep repositories in view of building confidence in geological performance models. The second workshop was held in Paris in 1994, and highlighted the "Characterisation of long-term geological changes for disposal sites". This workshop clearly noticed that scientific information concerning the long-term geological evolution is needed for several purposes such as disposal designs, safety assessments, confidence building and siting programmes.

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[1] "The safety case is an integration of arguments and evidence that describe, quantify and substantiate the safety, and the level of confidence in the safety, of the geological disposal facility" [see NEA, (2004), *Post-closure Safety Case for Geological Repositories – Nature and Purpose*].

Since then, through various initiatives, national safety assessments have been compared and studies have evolved towards providing an important basis for decision making related to geological disposal. That evolution has taken place through heightened incorporation of site characterisation data made possible by a real progress in site understanding and by an increasing depth of understanding of both technical and non-technical issues.

With regards to the long timescales involved in the concept of geological disposal, the relevance of various natural processes and events depends on the time frame to be considered. With respect to the lessons learnt from a previous NEA workshop,[2] the main concern is on features, events and processes (FEPs) over a period of about one million years – the order of magnitude of the time needed for radioactivity to decay to levels comparable to uranium ores is about a few hundred thousands years. Repositories are typically sited in stable geological environments in which key characteristics that provide safety are unlikely to change significantly in the course of time. However, over long enough timescales, even the most stable geological environments are subject to perturbing events and changes. Arguments for safety can be developed to build confidence in the overall safety case and an acknowledgement of the limits of predictability of the system will be important for credibility.

With respect to the geosphere stability, it is necessary to develop arguments for the reliance that can be placed on key safety functions. This applies in particular to the maintenance of the long-term containment capability of the geosphere and/or the maintenance of favourable mechanical or chemical conditions in and around the engineered barrier system. In building in an overall safety case, it is therefore important to assess:

- the features, events and processes that could impact the evolution of the geosphere;
- the long-term stability of the favourable conditions displayed by the host formation;
- the buffering capacity of the formation vis-à-vis perturbations.

The key issue is to evaluate the resilience of the main safety functions of the geosphere (often relying upon flow and transport properties) to natural perturbations. Thus phenomenological evidence of persistence of those functions in past episodes of climatic changes, seismic activity, diagenetic evolution, burial/uplift, etc should enhance confidence in geosphere stability.

To provide national waste management organisations and the scientific community at large with an overview on that subject of geosphere stability, the NEA Integration Group for the Safety Case (IGSC) launched a series of workshop dealing with this issue for various host rock types (i.e. crystalline rocks, argillaceous media and evaporites). The first workshop of the series dealt with argillaceous formations. It was hosted by GRS, (Gesellschaft für Anlagen und Reaktorsicherheit) in Braunschweig, Germany, on 9-11 December 2003. The workshop was organised under the auspices of the IGSC Working Group on the Characterisation, the Understanding and the Performance of Argillaceous Rocks as Repository Host Formations (referred to as "Clay Club").

Due to the multidisciplinary aspect of the IGSC initiative, the Earth Science community was solicited to give its views about the scientific and operational bases for the assessment of geosphere stability. Therefore the workshop brought together scientists from academic institutions, engineers from various research institutions or companies, consultants, regulatory authorities and national waste management organisations. One should note that the workshop did not aim to present details about all relevant disciplines but rather establish bridges between these disciplines and between the various communities.

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[2] NEA (2002), *The Handling of Timescales in Assessing Post-closure Safety of Deep Geological Repositories*, Workshop Proceedings, Paris, 16-18 April 2001.

SCOPE AND OBJECTIVE OF THE WORKSHOP

Among the favourable properties often quoted to support the choice of argillaceous media as a host formation for disposing of high-level and/or long-life radioactive waste are:

- Ability to self-seal fractures and discontinuities.
- Diffusion-controlled transport of solute.
- High sorption capacity.
- Geochemical conditions that favour low radionuclide solubilities, high sorption of migrating species and low degradation of engineered barrier system (EBS).
- High geochemical and mechanical buffering capacity, which is often considered as a means to maintain favourable properties over a long period of time.

The workshop focused on clay specific issues and considered the whole spectrum of argillaceous media envisaged as host formations, i.e. from poorly indurated, soft clays to hard, potentially fractured clays. In particular it addressed:

- The multiple lines of evidences to support the stability/buffering/robustness of the clays over long timescales.
- The resilience of the favourable properties of clays to natural perturbations; tested against, experiences on changes or alteration in response to diagenesis and geological history (tectonic, thermal loading, water expulsion, organic matter degradation, etc.), and may be quite reactive to external and internal perturbations (such as oxidation or thermal load).

A further important objective was to evaluate the extent to which there is confidence about the required level of stability, whether it is known what evidence to look for, and if there are adequate tools to carry out long-term investigations.

The workshop focused mostly on natural events. As a consequence, some repository-induced effects (e.g. radiolysis, and migration of alkaline plumes) were excluded from the remit of this workshop.

With respect to the order of magnitude of the time needed for HLW radioactivity to decay to levels comparable to uranium ores, the workshop main focus is on features, events and processes over a period of about one million years. This is in line with the conclusions of a previous NEA initiative related to the handling of timescale in assessing a post-closure safety.[3]

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[3] NEA (2002), *The Handling of Timescales in Assessing Post-closure Safety of Deep Geological Repositories*, Workshop Proceedings, Paris, 16-18 April 2001.

The synthesis aims at summarising the oral and poster presentations given at the workshop as well as presenting the outcomes of the workshop discussions. It does not aim at providing a detailed overview of the state of the art in all the geoscientific disciplines that were touched upon during the workshop. Rather it focuses on providing, to the extent possible, some insights for supporting arguments to build confidence in the stability of the geosphere for deep repositories, notably through answering the key questions that were set up in advance by the workshop Scientific Programme Committee.

The synthesis is completed by a compilation of extended abstracts (Appendix A). The list of participants is given in Appendix B.

Fifty-eight persons from the academic community, research and development institutions, national waste management organisations and regulatory authorities from eleven NEA member countries attended the workshop.

SYNTHESIS OF THE WORKSHOP

The workshop was divided into four sessions and a poster session. The first and second plenary sessions concerned respectively the fundamental requirements for the deep disposal and the scientific understanding about natural endogenic and exogenic processes that could affect the geosphere properties. The third and fourth sessions focused on more specific processes with regards to the stability and buffering capacity of argillaceous formations.

Session I – General Framework: Argillaceous Media as Host Formations

The main issues that were discussed were:

- *"What are the main functions/roles of the geosphere for disposal at different timescales (especially for argillaceous media)?"*
- *"What kind of assumptions relating to argillaceous geosphere are commonly made in safety cases?"*
- *"What are* the regulatory expectations concerning the confidence in geosphere stability?"

Ph. Lalieux, ONDRAF/NIRAS, Belgium, gave an overview of the expected roles of argillaceous media as host formations and the important aspects to consider in geological disposal safety cases. Disposal in a geological formation aims at protecting humans and the environment from the radionuclides in high-level and/or long-lived radioactive waste by isolating the waste by means of a multi-barrier system, that consists of an engineered barrier system (EBS) and a natural barrier (host formation and its surrounding geological setting).

The long-term safety is achieved through the fulfilment of a series of safety functions by multi components of the disposal system and among various time frames. The main safety functions of the host formation are: (i) to isolate the repository from human activities and from external phenomena; (ii) to provide a favourable chemical and physical environment to the engineered barriers; (iii) to act as a barrier to radionuclide release and migration toward the biosphere; and thus (iv) to attenuate the impacts of the repository. The most challenging aspect is the long timescale involved which is much greater than applies to any common human engineered structures. Quantitative assessments are made typically over one million years compared with the fifty years for civil engineering. One million years corresponds to the time at which the radio-toxicity index declines to be about five times the natural radioactivity of a typical host formation.

In case of disposal of HLW in clay, the host formation can be considered as the main barrier and so stability is one of the key characteristics of the site selection. Some favourable characteristics of the host formations to be investigated are: low hydraulic conductivity and gradients, extent and thickness continuity, high sorption capacity and generally long-term stability of those characteristics. It must be remarked that the long-term performance of the clay formation may overshadow the performances of other components of the disposal system like the EBS.

The geosphere stability issue is managed in performance assessment through the construction of one or more scenarios reflecting the likely or possible evolution of relevant features, events and processes. However, it makes an important contribution to the overall safety case if a convincing argument can be made for the long-term retention of safety functions derived from the geosphere characteristics. To improve the quality of assessments and to strengthen the confidence of stakeholders including regulators, approaches that are considered in those assessments should be in agreement with most recent developments and findings of Earth scientists.

As an illustration, the Boom Clay case was presented in particular as a practical application of what "safety functions" and "favourable characteristics" mean. In the Belgian HLW reference case, the quantitative safety assessment assumes that the EBS (with the exception of the waste matrices) does not fulfil any safety function after the thermal period. The latter has duration of a few hundreds of years for vitrified HLW and a few thousands of years for spent fuel. After this thermal period, performances of the waste forms (i.e. their resistance to lixiviation) barely influence the estimated doses as the latter are mostly controlled by the performances of the host formation. Therefore, the maintenance through time of diffusion-controlled transport (i.e. low hydraulic conductivity and low hydraulic gradients), reducing geochemical conditions and high sorption capacity is of key significance in building confidence in the safety case. The normal evolution scenario used for performance assessment considers a steady state geosphere regarding the diffusion and sorption processes. Additionally, various separate altered geosphere scenarios are or will be considered such as periglacial conditions, denudation, faulting. These scenarios are based on current geological knowledge but do not consider probabilities of occurrence of those events. They cannot be considered as representative of an evolving geosphere but rather as illustrations of various end stages of geological events.

J.-Y. Boisson, IRSN, France provided the French regulator's point of view concerning the handling of geosphere stability, which is addressed through qualitative requirements regarding site properties. He emphasised the safety recommendations as depending on the stage of the development of the disposal project. In particular, considerations of the geosphere stability might be different for the site selection stage and for subsequent stages. The French Basic Safety Rule III.2f (BFS III.2.f) distinguishes the essential criteria from important technical criteria to be fulfilled by selected sites. The long-term stability of the site and its hydrogeology characteristics are considered in the BFS III.2.f as essential criteria whereas important criteria concern thermal, mechanical and geochemical properties, and minimum depth requirement. In addition, no natural resource should be of interest in the area. After the site selection phase, the subsequent steps should confirm the favourable characteristics of a selected site and assess the potential disturbances by the engineered barrier and site development to the host formation capabilities. The overall objective of that step by step approach, which consists of site selection, feasibility studies, and design options, is to propose at least one technically conceivable disposal design for the protection of human health and the environment.

Session II – Examples of Key Exogenic and Endogenic Processes Affecting the Geosphere

This Session aimed to give an overview of the state of the art of the knowledge on key processes and events that could affect the geosphere properties. A wide range of events and processes were presented: climate, geomorphology, seismicity and faulting, basin geological history and diagenesis.

The key questions to be addressed were:

- *"What are the predominant processes for natural and anthropogenic evolutions?"*

- *"What kinds of predictions over long timescales are recommended and what are the related uncertainties?"*
- *"What predominant predictions could be provided for crustal movement and seismicity?"*
- *"Are the diagenetic processes in a subsiding basin continuous or not in particular regarding THMC mechanisms? What are the potential consequences on the geosphere at repository depths of these processes e.g. ice loading, saline water intrusion, oxygenated water intrusion, crustal movement, and seismicity?"*

V. Masson-Delmotte, CEA, France gave an overview of the understanding of past, present and future climate processes. Although short-term climate (40-1 500 years) behaviour seems rather chaotic, it is possible to understand and predict long-term climate characteristics and their general evolutions. Direct records of atmospheric parameters are very recent, but ice cores provide records for up to the last 800 000 years. Climate forcing and feedback have diverse time and spatial scales and volcanism may affect climate over a few years. Earth's orbital parameters induces climate cycles of 100 000 years. The natural variability is mainly characterised by glacial-interglacial cycles that are governed by astronomical parameters. Rapid events may be of importance such as growth or decay of ice sheets modifying the ocean circulation over tens to hundreds of years at a hemispheric scale level. Anthropogenic greenhouse effects and aerosols undoubtedly influenced the climate/over the last century.

For the future, climate models integrate the general atmospheric and ocean circulation models, land, biosphere and ice models as well as CO_2 cycle model and scenarios of human consumption for the next century. The model computations show that, due to anthropogenic releases in the atmosphere, the present interglacial, in the worst case, would span over 200 000 years instead of 50 000 years for an unperturbed natural system. During this longer warm and wet period, the temperature rise is expected to induce the complete melting of ice sheets, a significant sea level rise, variations in land cover and use. Beyond 200 000 years climate variations should be controlled again by orbital forcing, in the absence of any further anthropogenic perturbation.

Progress on modelling capacities are observed and will improve the understanding of climate processes in the future. One of the key parameters that influence the potential consequences of climate changes for repository safety is the water availability for dilution (recharge).

It was noticed during the discussion the potential helpfulness of feedback of the EC BIOCLIM project (report planned in 2004).

F. Schlunegger, University of Bern, Switzerland presented an overview of surface processes shaping the topography of mountainous systems. Three main processes were described and illustrated: hillslope creep, hillslope failure and fluvial processes (incision and sediment transport). Landscape parameters such as roughness or length scale depend on the local dominant erosion process. The fluvial and debris flow dominated systems have rough, highly incised surfaces with dense channel network, whereas areas dominated by hillslope processes have smooth topographies, low relief and greater channel spacing. Geomorphologic processes depend on two main external forcing: climate and tectonics. Climate mainly influences process rates and thus timescale and topographic roughness and length scales. A theoretical approach predicted that warm and humid climate enhances hillslope processes. Further studies are needed to validate this theory in the field. Tectonic uplift increases slopes and generates upward propagating erosion with a recognisable knick-point in the river profiles. In an Alpine study case, erosion rates were determined by measuring sediment load in the river as a proxy to total basin erosion. It illustrated the correlation between erosion and bedrock lithology as well as with former glacial zones. However, this approach has a strong bias since erosion can be much localized in the basin. Measuring changes in channels or studying aerial photographs may be some alternative tools to determine erosion rates. The discussion raised the representativeness of Alpine features (e.g. “instantaneous” erosion rate) and the way to extend such data and observations outside orogenic areas. Indeed, most – but not all – considered repository sites are in rather flat areas where an overall accumulation of sediments should occur. As many data related to erosion rates are based on observations in orogenic areas, they can only be considered as an upper limit for performances assessment purposes.

The USEPA (United States Environmental Protection Agency) standard definition of 1981 defines an active fault as “any fault that can be shown to have produced an earthquake in the last 10 000 years”. The inherent difficulty with respect to this definition might be the issue to associate an earthquake to a given “active” fault. Thus, **R.M.W. Musson, BGS, United Kingdom** argued in favour of a new classification of faults as “inert”, “reactivatable” and “controlling” faults. Inert faults are defined as faults that cannot be reactivated, at least in present stress regime. Reactivatable faults can be, or have been reactivated. Controlling faults are the preferential loci for current deformation. The assessment of faulting matters for two types of hazards: rupture and shaking hazards. The rupture hazard is clearly important as it can disrupt the waste area. Rupture features may induce secondary induced hazards such as changes in groundwater flow. The first defence against rupture hazard is obviously siting as a vast majority of earthquakes takes place along pre-existing structures (reactivatable faults). Rupture hazards in argillaceous media have been understudied. In such environments, earthquakes usually occur in the underlying crystalline rock and do not reach the surface. In some cases, argillaceous sediment may inhibit the rupture propagation but may not prevent deformation after the event.

The importance of the shaking hazard is less clear, in particular since it was shown that ground motion decreases rapidly with depth. In addition, the issue of long-term representativity of the current recognised seismicity was also discussed as tectonic processes may change over the periods considered for safety assessment and may exhibit significant heterogeneities. However, significant tectonic changes are most likely to take several million years to develop and are thus not of prime importance. Climate changes and particularly the deglaciation rebound may also affect the seismicity of a given area. Just after ice retreat, deglaciation faulting may be more severe than the one currently registered in Northern latitudes.

Current approaches for estimating seismic parameters and their limits in low seismic areas, such as most of the clay host formations for deep geological repositories, were also presented. Considering the few experiences that exist on the rare seismic events in such settings, both probabilistic and deterministic methods have strong limitations when assessing seismicity over long periods of time. Balance between adequate conservatism and a sense of physical realism is to be sought ideally.

T. Nohara, JNC, Japan presented results on research on fault activity in Japan which represents a high seismic area. Japanese islands are at the margin of the Pacific plate which collides with the Eurasian plate with a convergence velocity of 8 cm/year. It is reasonable to assume that the current tectonic setting will continue for the next hundred thousands of years and so will the associated seismic patterns: interplate earthquakes and inland shallow ones. The latter could be considered as the most dangerous for the underground repository stability. In Japan, scientific studies started in the thirties with active fault mapping. Associated distinctive landscape features can be identified by field studies, aerial maps and geophysical studies. Collected data highlighted regularity of active fault movement over time. Both laboratory and field studies investigated a relationship between the cumulative displacement and the width of the crushed zone which typically ranges from several meters to several hundred meters at most. Deformation occurs on a wider area. The associated effects of major active faults on a deep geological repository might be insignificant at distances greater than 10 km from the active major fault. This safety distance may be lowered with field study. Measurements at the Kamaishi mine have also clearly demonstrated strong attenuation of ground motion with depth.

F. Schneider, IFP, France introduced the point of view of the petroleum exploration industry which is relevant to geological basin studies. Basin modelling couples several numerical models such as kinematic, deposition and erosion processes, compaction, heat transfer, organic matter maturation and fluid transfer models. Present data and paleodata are crucial to constrain these models; they are used to estimate rock physical properties and geometrical information on the basin. Deposition history is determined from seismic and geological data and then translated into sediment supply rates and subsidence maps in order to be used as input to the 3-D stratigraphic model. This stratigraphic model simulates sediment deposits at the basin scale. A loading path is simulated since present sediment behaviour and properties depend on the burial history and tectonic setting. Constitutive laws of rheology and compaction are then computed to account for the evolution of physical properties through time. Furthermore, the model is able to simulate diagenetic processes and induced changes in hydraulic parameters. As climate and tectonic settings change with time, transient phenomena could occur over long timescales, and, therefore no steady-state should be considered. The major difficulty is to estimate the boundary conditions to apply to the modelled system. At the end, it was recognised that even if sediment history can be characterised in the laboratory, long timescale processes must be addressed by basin modelling. Some natural analogues could be used for a better understanding of the involved processes (e.g. Niger analogues for shale deposits).

L. Warr, CGS-EOST, France made an overview on diagenetic processes in clays and their forcings. First he noticed that the prediction of diagenetic evolution of argillite is based on the study of natural and experimental system. Processes could be classified in three main categories: the Good, the Bad and the Ugly, respectively precipitation and crystallisation, dissolution, and fracturing and anisotropy. Diagenesis in argillite is controlled by fluid chemistry, rock composition, hydraulic conductivity, temperature, time and stress. However, much more is known on thermodynamic processes than on kinetics. Rates of diagenesis vary greatly in nature. Fast diagenesis occurs in environments subject to weathering, fast hydrothermal, or fracture/fault controlled and spans from years to ten thousand years. Diagenesis due to burial and metamorphism is a slow process occurring over longer timescale (10 000 years to millions of years). Fine-grained mineral reactions are quite well known by applying thermodynamics, but very little is known on the kinetics of reactions in argillaceous media. Standard laboratory methods for monitoring mineral reactions use batch reactors where minerals grow. The main issues of these methods are that they concern closed systems and that sampling induces disturbances of the natural media. A new method was presented using flow-through reactors (wet-cell) in a hot chamber. This open-system method allows better reproduction of rock evolution.

The general discussion closing this session on external processes and their impact on the geosphere pointed out that there is a need to further study the impact of low-level earthquake propagation in clay media. More generally, there seems to be a discrepancy of interests between the academic world and the waste management industry. The first is studying regions where processes are active (mountainous, hydrothermal or fractured areas). In contrast, deep geological repository sites are located in areas that have been selected for their high stability and where such processes are quite slow or neglectable. However, it was stated that the waste management industry should open its facilities and data to academia because they characterise unique sites in very specific media and stable areas.

Session III – Arguments to Support Confidence in the Stability of Clays Considered as Potential Host Formations

This Session focused on arguments in support of the confidence in geosphere stability such as:

- Large-scale geo-tectonic environment and its evolution.
- Burial and temperature history.
- Scientific basis in particular on methods and arguments.
- Paleohydrogeological aspects.
- Future geological evolution, predictability, effects analysis.

P. Hoth, BGR, Germany presented the screening methodology used in Germany for site selection. Characterisation of clay sites started only recently since Germany previously had focused its research on the use of thick salt formations. The first phase involves a mapping of shale formations, an analysis of well files, and an establishment of seismic profiles in order to identify the shale formations that meet the following three thresholds: minimum thickness of formation equal to 100 m, a burial depth of the shale formation below 1 500 m and an upper level of the disposal at more than 300 m deep. Three formations meet these specifications with shale content above 80%: Tertiary, lower Cretaceous and middle to lower Jurassic in Northern and Southern Germany. The second phase comprises a regional characterisation of these sites with well-log analysis. Burial, temperature history, hydrocarbon production, compaction and diagenesis are reconstructed and detailed maps created. Moreover, shale homogeneity and predictability are major issues in the assessment of the host formation. There are addressed in this screening phase with a numerical method developed for well-log correlation. The future third screening phase will select dominant processes and criteria of importance for the site evaluation.

A. Gautschi, Nagra, Switzerland presented lessons learnt from the past studies in order to predict the future evolution and stability of the Opalinus Clay site in the Zürcher Weinland area in NE Switzerland. He focused on the multiple arguments that support the stability of this host formation. The Opalinus Clay formation is 100-125 m thick in 300 m of confining units, surrounded by two aquifers. In order to get the strongest argumentation in the prediction of the future evolution of the Opalinus Clay, the long-term history of the site was reconstructed through the application of a large number of techniques such as apatite fission track analysis, and stratigraphic evidence. In particular, the burial history showed that the site was affected by a maximum temperature of 85°C which corresponds closely to the expected temperature at the vicinity of the waste storage facility. The host formation is a diffusion dominated system and fault activity is low. Furthermore, the facility would be constructed at a sufficient distance from a major fault (Neuhausen fault) which has been inactive for millions years. The erosion rate of the fluvial system was estimated from the difference between the current topography and the reconstructed pre-glacial peneplain. Calculated erosion rates of 0.07 to 0.15mm/year are in line with the Danube terraces dating. In conclusion, predicting the future evolution and stability of a geological system for time-scales up to 1 million year using information from past evolution is an acceptable scientific approach not witchcraft.

J. Brulhet, Andra, France presented a status of the French study on the evolution of the Callovo-Oxfordian Argillite Site in Eastern France. The Callovo-Oxfordian site is a 130 m thick hard argillite layer at 420 m depth and is surrounded by non productive aquifers. The region is tectonically stable, away from ice-sheet development areas. However, the region is subject to moderate erosion:

erosion of the circular sedimentary outcrops and associated incision of valleys. Andra is following a two fold approach. Firstly, the past evolution of the basin is reconstructed in order to better understand the present conditions and the properties of the environmental settings and of the host rock. Secondly, potential external changing conditions are determined and their impacts on the future evolution of the site are assessed. There are three complementary elements to the analysis: (i) field work with a series of conceptual models at various temporal and spatial scales and using acquired data and knowledge; (ii) numerical simulations of the driving forces such as climate or geomorphologic processes; (iii) syntheses work via one conceptual model integrating the whole evolution to ensure the overall consistency of the multidisciplinary studies. An interdisciplinary research group (Bio-geoprospective Group) ensures the overall consistency of these studies. At the scale of interest, climate and geomorphology are considered as the two most important factors. Cold climate has direct effects on the geosphere characteristics through the development of permafrost and of glacio-eustatic fluctuations. Climate change was modelled for the next million years under the BIOCLIM European project. There is little indication of direct changes in the host formation. But, one can expect due to the geomorphologic context, changes in the river network and thus long-term modification of groundwater flow in the surrounding aquifers.

T. Torres, Madrid School of Mines, Spain gave an overview of the Spanish program regarding the evaluation of long-term geological and climatic changes. Two sites were presented: the Priego area and the Baza Basin. Regarding the Tufa deposits in the Priego area of Central Spain, mineralogical and chemical properties were first studied. Secondly, a geochemical model was utilised to better understand the evolution of both mineralogical and chemical properties. A dating method (aminostratigraphy) using amino acid racemisation was applied to ostracod shells of the Tufa at different terrace levels. The estimated incision rate was as high as 100 m/400 Ky. Several analytical analyses were performed on the lacustrine deposits of the Guadix-Baza Basin (AARD dating, paleomagnetism, paleobiology, isotopic analysis, fluid inclusions, and trace elements.) Based on that analysis, it was possible to establish the paleoclimatic and paleoenvironmental reconstruction of the area. T. Torres strengthened the particularity of the Mediterranean settings that are quite different from the North European settings. In particular, glacial and interglacial periods could be described respectively by “cold and humid” and “warm and arid” conditions. In addition, the Padul peat bog in Southern Spain was introduced as a paleoenvironmental record which covers 900 000 to 3 000 years before present.

Session IV: Reaction of Argillaceous Media vis-à-vis Natural Perturbations and Geosphere Evolution (Buffering)

The key questions to be addressed were:

- *“What is the stability of key transport processes and parameters (diffusion, ...)?”*
- *“What kind of analogues should be used to support confidence (e.g. other types of formations and coming from other industry, e.g. the oil industry)?”*
- *“What kind of arguments could support the THMC buffering or absence of buffering? (NB repository-induced effects are excluded)”.*

Four main topics were considered:

- Hydro-mechanical (HM) aspects.

- Long-term efficiency of diffusion.

- Chemical buffering/mineralogical processes.

- Organic matter evolution.

S.T. Horseman, BGS, United Kingdom made a state-of-the-art presentation on the understanding of self-healing properties of clay rich rocks. The term “self-healing” is regularly cited as the primary mechanism which should reduce mass transfer (water, radionuclides and gas) in natural fractures and in the excavation damage zone (EDZ). Self-healing could result from mechanical processes (change in the stress tensor), hydro-mechanical processes (change in the water content leading to swelling), geochemical processes (alteration, mineralisation) or combined processes. These processes tend to reduce the fracture openings, or fracture porosity. S. Horseman developed his presentation on the basis of: (i) the experiments performed on EDZ in Underground Research Laboratories such as Mol (Belgium) and Mont Terri (Switzerland) and on core samples in surface laboratories; (ii) self-healing observations on natural analogues; (iii) soil and rock mechanical concepts and theories (dilatancy, brittle-ductile transition, viscoplasticity, …) explaining the self-healing process. He concluded that the self-healing should lead to a gradual decrease of mass transfer. Nevertheless, the long-term geochemical evolution of the damaged rock and its effect on the transport properties of the EDZ remain to be elucidated. Clay properties favouring self-healing are usually drawbacks for engineering purposes since it is considered to be difficult to drill in such media.

P. Marshall, Nagra, Switzerland reported results on studies regarding the impact of geological evolution on long-term safety of a potential HLW repository in the Zürcher Weinland (NE Switzerland). The geological evolution comprises erosion, uplift and climate changes. The main perspective was the impact of these processes on the hydrogeology of the site: vertical gradient modifications induced by changes in recharge and discharge areas, permeability enhancement of Opalinus Clay due to uplift, expulsion of contaminated pore water from the disposal area as a result of ice loading. With regards to the latter, the speaker presented a complete modelling of the ice-loading effect. In this study, the host rock and the repository architecture were taken into account. Overpressure created at the repository level by the ice loading and the pore water expulsion towards the surface was computed. Based on this modelling and on geological observations, erosion and other geomorphologic processes will not enhance vertical gradients between layers because topography is flat. Uplift is not expected to enhance permeability of the Opalinus Clay over the next million years. Ice cover does not change the diffusion dominated transport regime. P. Marshall concluded that safety functions are met for the Opalinus Clay sites.

M. Mazurek, University of Bern, Switzerland analysed natural conservative tracer (Cl) concentrations in several clay formations (Belgium, France, Switzerland, United Kingdom, and Canada). As the speaker said: “these profiles help to understand the long-term evolution of this type of formation because they represent long-term and large-scale natural experiments that can be used for the up scaling of laboratory data in space and time”. Considering such diverse formations as the silt layer at Marcoule (France), the Opalinus Clay at Mont Terri and Benken (Switzerland) and the Boom Clay at Mol (Belgium) he interpreted most of the tracer profiles (Cl, ^{3}H and ^{18}O) as diffusion profiles. Diffusion process took place generally for periods spanning from 100 000 years up to several millions of years. The lack of information in paleo-hydrogeology is the limiting factor in interpretation. Nevertheless, these profiles tend to show the efficiency of diffusion over a very long period of time.

Martin Mazurek’s presentation was followed by a series of four presentations dedicated to the buffering capacity of clay rocks. As C. Beaucaire said: “*The long-term performance of nuclear waste repository is strongly dependant on the chemical properties of the host rock. The host rock establishes* *the chemical environment that determines such important performance attributes as radionuclide solubilities from the waste and the transport rates from the repository to the accessible environment. Clay-rich rocks are especially favourable host rocks because they provide a strong buffering capacity to resist chemical changes prompted internally either by reactions of the waste itself and emplacement materials, or externally by changes in the hydrologic system surrounding the host rock.*"

C. Beaucaire,[4] **IRSN, France** focused her presentation on the ability of a clay rock to provide pCO_2 and redox buffering and to resist external chemical changes. C. Beaucaire pointed out the buffering capacity as a measure of the system ability to resist to changes in its intensive properties which are chemical potentials of system components such as pH, pCO_2, pE, solute concentrations. Temperature and pressure can be considered as external boundary conditions. CO_2 and O_2 may be supplied externally directly from the atmosphere during the operational phase or after its closing. According to the existing literature, C. Beaucaire showed that clay rocks have a high buffering capacity, due to mineralogical buffers, for CO_2, mainly carbonate and alumino-silicate minerals, and for O_2, reduced phases such as pyrite, siderite and organic C. Thus, they are not changed rapidly in response to external chemical changes.

Then, **É. Gaucher, BRGM, France** brought some key elements that support the understanding of the argillaceous media stability. As an example, he reminded that argillaceous formations can be stable over very long periods of time, from 150 million years, regarding the Callovo-Oxfordian formation, up to 420 million years for the Silurian formation of the Arabic platform. É. Gaucher highlighted three key arguments that support a better understanding of the geochemical stability of this type of rocks: (i) favourable liquid/solid ratio (very fine granulometry) allowing very strong buffering effects through low porosity, low permeability and long water residence time); (ii) progress in the understanding of clay thermodynamics and clay stability (illite and smectite), auto-regulation of carbonate system by mineralogy (cf. Beaucaire's presentation). He concluded that clays appear to be geochemically stable if temperature and pressure do not evolve significantly over a long time period.

The two following presentations were given by **J.-C. Parneix, ERM, France**. The first one focused on natural analogues studies as arguments to support the understanding of the impact of temperature on clay geochemistry. Studies of natural analogues allowed measurement of the mineralogical transformations (smectite illitisation) with temperature and provide therefore the possibility to develop and validate coupled thermo-geochemical models. At the present time the main mineral reactions involving clays are understood and evaluated and the corresponding kinetics have been partly determined. A time vs. temperature diagram of clay mineral stability was proposed. However, the impact on the rock physical properties is poorly known and should be studied in the future. As an echo to this recommendation, the second presentation showed how to determine two- and three-dimensional micro-structures of a clay matrix. The Scanning Electron Microscope (SEM) and the Electron Probe Micro Analyser (EPMA) techniques provide the means to experimentally establish the pore and mineral spatial distribution. Based on that distribution, the inference and modelling of transport properties are on going and first, interesting results have been obtained.

J. Samper, University of Coruña, Spain gave an oral overview on the Spanish "reference" clay and the associated research programme with an emphasis on clay geochemical characterisation and determination of the buffering capacity. This "reference" formation is located in a lacustrine-continental sedimentary basin deposited during the Oligo-Miocene period. The clay layer is surrounded by two aquifers with a marly gypsum semi-impervious layer between the clay and the upper aquifer. The potential burial depth lies between 82 metres and 600 metres. The clay mineralogy

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[4] C. Beaucaire is currently working for the CEA, France.

is a mixture of smectite and illite. J. Samper presented the geochemical modelling of hydrochemical experiments performed on drill core samples. Then, he introduced some results of the preliminary paleogeochemical modelling to assess the geochemical stability of the host rock. This modelling was performed with the geochemical code CORE. A preliminary sensitivity analysis has been performed with respect to the direction and magnitude of the vertical water fluxes across the clay layer in order to fit the tracer concentrations, mainly the chloride. Results show a good agreement with tracer data with an upward flow of 2 metres per million years. Extensive future work is planned in particular on the automatic calibration of tracer profiles taking into account reactive chemical species, isotopic data, etc.

R. Michels, ERM, France summarised the results on the nature and reactivity of organic matter in the Callovo-Oxfordian of the Bure site. As the speaker quoted: “Especially in the case of argillaceous sediments, known for their proneness to organic matter, the study of fossil organic matter is able to unravel a large amount of information concerning the geological history (depositional conditions and preservation, paleoenvironmental, and burial, thermal history) as well as the future evolution (effects of induced thermal perturbation, oxidative alteration, biodegradation)”. The results of the extensive characterisation of the organic matter in the site (chemistry, thermal maturation, etc.) were presented. R. Michels concluded that “the organic matter will not react further in terms of geological thermal reactivity within the next million years unless the basin subsides or the heat flux increases significantly”.

B. Beaudoin, École des Mines, France presented a summary of the work in progress on the fracturing and associated carbonate transfer in shaly-marly blocks. This work has been performed within the MINANDRA partnership between Paris School of Mines and Andra. At the present time; two main processes are studied in the field on two large outcrops located in the South East of France. B. Beaudoin showed some examples of early fracturation at different scales and explained the evidence of presence of carbonate nodules as an indicator of carbonate transfer. The validated methods will be applied to the Andra URL at the Bure site in France. B. Beaudoin discussed the difficulty to link and extrapolated results from experiments on samples, localised field observations to formation behaviour at depth. Field and samples work are complementary in view of bringing multiple arguments to build confidence in the performance of a formation.

A. Bauer, FZK, Germany focused his oral presentation on the preliminary results on the evolution of clays in basic solutions. Basic solutions may be generated by interaction of pore-water with concrete. The potential negative impact of such solutions on clay mineralogy is one of the main issues to be considered. Smectite in an alkaline solution may lead to illitisation and collapsed layers. A. Bauer presented results of observations that were carried out on the geochemical evolution of clay samples from the Bure site and the Opalinus Clay. Clays have been in contact with highly alkaline synthetic solutions in laboratory batch experiments. No visible effect on the Bure and the Opalinus Clays was observed even after one-year experiment. Natural organic substances in potential clay host were characterised using a Scanning Transmission X-ray Microscopy (STXM). Natural organic matter still covered clay edge Si, Al functional groups after 1 year in SYF (closed system), indicating the passivation of the reactive sites. Organic matters from various origins are connected to different clay phases. This result was not expected for diagenetically overprinted sediments. The released humic and fulvic acid is related to the sources and type of argillites respectively smectite and illite/calcite.

The issue of linking the microscopic tests and observations to the formation level was debated at the end of this session. It was noted that the diffusion coefficients that are measured in samples and at the formation level at the Mont Terri site were of the same order of magnitude. Moreover, microscale studies and in particular the ones on organic matter were recognised as being useful in validating the macroscale (formation scale) approach as it was shown during the experiments made on the Boom Clay site.

Gas migration was considered as a key issue and so studies have to be continued. Natural analogue studies were viewed as potential arguments to support the process understanding and it was so suggested to go further on that topic when developing multiple lines of evidence on the performance of a site in a safety case.

No agreement was achieved on the pertinence of coupling geomechanical and geochemical models. In a Japanese unlined mudrock tunnel, oxidation by diffusion of O_2 leads to softening of the clays. These effects are rock dependent and might be negligible for other sites. Furthermore, very few studies have been conducted on the permafrost effects on clay rocks.

POSTER SESSION

The Poster Session dealt with additional case studies and with more specific technical details.

G. Kosakowski, PSI, Switzerland, presented some numerical investigations on the influence of glacial loading on radionuclide transport in the Opalinus Clay. His poster illustrated the more general presentation of P. Marshall (Session IV). Kosakowski presented a rather complete modelling study of the influence of consolidation flow due to successive glaciations on radionuclide transport in the repository architecture and in the Opalinus Clay. He highlighted that the overpressure created at the repository level by the ice loading increases the release of pore water towards the surface and therefore enhances radionuclide transport.

A. Dierckx, ONDRAF/NIRAS, Belgium, showed the burial histories of the Boom Clay and the Ypresian Clay, which is an alternative potential host rock. They concluded that the area of the HADES Underground Research Facility was relatively tectonically stable for the past 50 millions years.

M. Van Geet, SCK•CEN, Belgium, presented the work performed on the characterisation of organic matter in the Boom Clay. The organic matter contained in the Boom Clay (5% of its weight) may act as complexant for radionuclides. Van Geet concluded that the organic matter is very immature and could evolve in the future, and therefore it should be taken in consideration for the performance assessment.

J. Brulhet, Andra, France, presented a deterministic method to compute seismic ground motion. This method simultaneously treats both epistemic and random uncertainties. The first ones, being associated with the present knowledge about the geological structures and their deformation modes, are propagated using a logic tree. A Monte Carlo technique is used to simulate the latter, which are associated with the input parameters of spectra (magnitude, depth and attenuation law) in the context of deterministic seismic hazard assessment.

J. Ďúran, VUJE, Slovak Republic, gave a summary of the on-going site selection process in Slovakia for nuclear spent fuel disposal. The process which started in 1996 selected initially 15 areas. At the present time, four areas in granite and two in clay remain. The final selection is expected in 2010.

F. Claret, FZK – INE, Germany reported some experiments on the characterisation and quantification of humic and fulvic acids released by the Bure and Opalinus Clays in highly alkaline solutions. This poster related to part of the work programme presented by A. Bauer (Session IV). Colloids may play an important role in radionuclide transport due to their high complexation characteristics. The author observed the complexation of these acids with Cm^{3+} ions. They conclude that radionuclide migration in clay under cement dissolution conditions may be enhanced.

I. Szűcs, MECSEKÉRC, Hungary, presented the results of the investigation programme – the so-called Short-term Programme, till 1999 – of Boda Claystone Formation, which is the potential host rock for HLW and SF disposal in Hungary. The poster illustrated the geological properties of BCF, the explorations in the former underground research laboratory, and listed the mineralogical and hydrogeological properties of the Boda Claystone. Furthermore, long term safety issues and the geotechnical suitability were highlighted. In the final conclusion it was mentioned, that after 1999 there were not carried out further explorations, but an overall and countrywide site screening was completed. Based on the results of the screening and the SPT a new site characterisation process started in 2003.

H.-J. Herbert, GRS mbH, Germany, investigated mineralogical and chemical changes of bentonites in a natural analogue study and in laboratory experiments where bentonite samples were in contact with high saline solutions. Their purpose was to study the impact of the solid/liquid ratio on the stability of bentonites. The idea was that open geological media are characterised by low ratio (flow of large water volume over geological times) and that bentonites used as technical barriers in salt formations are characterised by high ratio (closed media with very little water volume available). Bentonites are not stable within a high saline environment, neither in an open system nor in a closed system. Important mineralogical transformations were observed.

C.L. Zhang, GRS mbH, Germany, presented their experimental results on the hydromechanical behaviour of the Callovo-Oxfordian argillite core samples. These experiments have been performed in the framework of the MODEX-REP project which aims to investigate the hydro-mechanical response of the argillite to shaft sinking. The short-term mechanical behaviour of the argillite was investigated by means of uniaxial and triaxial compression tests. The long-term behaviour was studied by uniaxial creep and relaxation tests. Some influence factors such as material anisotropy, scale effect, water content and sample origin were also examined. With respect to the short-term mechanical behaviour scale and anisotropy effects were observed. Concerning the long-term behaviour the authors concluded that no lower creep limit, no significant scale effect and no significant anisotropy effect on the pure creep behaviour for the argillite were found. The long-term mechanical behaviour of the investigated region of the argillaceous formation was relatively homogeneous.

I. Kolaříková, Faculty of Sciences, Charles University, Czech Republic, reported experimental work performed regarding the impact of heating on the physical and chemical properties of bentonite from the Rokle deposit which may be used as a backfill material. Their results showed that thermal effects strongly affect the bentonite material, i.e. decrease in rehydration ability, decrease in cation exchange capacity and increase of specific surface area with temperature increase.

CONCLUSION

The key findings as summarised below were drawn during the five sessions.

Impact of exogenic perturbations (climate change and erosion)

One thousand years seems to be the predictability limitation of current climate models. It is however possible to understand and predict general long-term climate evolutions. Worst case modelling of climate change due to anthropogenic releases shows that the present interglacial would span over 200 000 years (with complete melting of ice sheets). Beyond 200 000 years climate variations should be controlled again by orbital forcing in the absence of any further anthropogenic perturbation. The key parameter that influences the prediction outcome is the water availability for dilution (recharge).

Most – but not all – considered repository sites are in rather flat areas where an overall accumulation of sediments should occur. As many data related to erosion rates are based on observations in orogenic areas, they can only be considered as upper limit for performance assessment purposes.

Impact of endogenic perturbations (seismicity)

Uncertainties exist in defining active faults and predicting magnitude and lifetime. Current concepts in hazard analysis originating in the context of active tectonic environments may not be readily applicable to low and moderate seismicity areas where most repositories are planned. Novel methods are being developed for assessing hazards at surface and at depth. Nevertheless, a consensus exists that hazard is low in low seismic areas and that a strong attenuation of ground motion occurs with depth. For high seismic areas, as in Japan, the attenuation related to depth and to distance in relation to the fault locations needs to be considered in siting.

One needs to differentiate shaking hazard to repository in operation (i.e. open or partially open underground infrastructures – with operational lifetime rather similar to classical nuclear facilities) and to disposal facilities after backfilling and closure. There is also a need to provide perspectives to the seismicity studies by a better definition of the period over which the engineered barrier system (EBS) integrity has to be maintained.

Geological evolution of clay formations

Basin modelling for predictive purpose is still in its infancy. Clay diagenesis is very slow and might not be relevant over periods smaller than one million years.

Self-sealing

Field observations provide evidence on the positive influence of this key process that should reduce the mass transfer of a potential opened fracture. Self sealing is reasonably understood from the hydro-mechanical point of view, but the influence of geochemical mechanisms less well.

Efficiency of transport processes over geological times

Diffusion in clay formations should be the main transport mechanism over very long timescales. That is supported by the current results of the analysis of tracer profiles in various argillaceous formations which relate to the last 10^5-10^6 years.

Geochemical buffering and organic matter

Given the generally long residence times, the chemical composition of pore waters in clay-rich formations is controlled by equilibria with mineral phases, such as carbonates and silicates, and ion-exchange reactions with clay minerals. The generally reducing conditions are strongly buffered by pyrite, siderite and/or organic matter. Therefore, clay-rich rocks are especially favourable host rocks because they provide a strong buffering capacity to resist chemical changes prompted either internally or externally. Expected natural geosphere evolution at repository sites should not induce drastic changes in organic matter behaviour. However, chemical (oxidation) and thermal perturbations induced by the construction and operation of the disposal facilities will affect organic matters (e.g. CO_2 production).

Studies on potential host formations and/or sites for HLW repositories have led to the following general conclusions:

a) It is expected that the ground surface of repository areas may experience some important changes due to climate and erosion, but that changes will decrease with depth. More generally, any hydraulic, mechanical or chemical perturbations should be buffered by the clay formation properties. There is little indication of direct impact of these surface changes on the host clay transport properties at depth. Outside the argillaceous host formations, radionuclide pathways towards the biosphere may however be affected by surface changes notably through a modification of the hydrogeological and geochemical boundary conditions in the surroundings.

b) Predicting the overall geological evolution of a given site and its environment over periods of time of relevance for long-term safety assessment (i.e. few hundred thousands years to one million years) seems feasible based on the current understanding and a combination of geocientific-arguments.

c) A large spectrum of geoscientific arguments in support of the long-term stability of host formations is being used by waste management agencies when building their safety cases for long-lived geological repositories in argillaceous formations. In most case, multiple lines of arguments covering various spatial (site to basin) and temporal scales are utilised. Furthermore, comparison with much less favourable geological situations and scoping calculations notably help testing the robustness of the proposed repository systems.

d) There is a need for waste management agencies to better harmonise and justify the potential multiple arguments in support to the geosphere stability. Indeed, there is an apparent lack of consistency when comparing the various sites and therefore the corresponding sets of arguments. One should foster a better justification of why an argument is used or is valid for a site or a host formation and not in another case.

e) The need to provide a comprehensive "history" of the host formation was emphasised in view of presenting multiple line of evidence of the capacity of the host formation to fulfill the safety functions. This contributes to building confidence when presenting a safety case. The petroleum industry already has important experiences on this matter, and this should be further used.

f) An additional important issue concerns the self-sealing process, as the EDZ remains, in stable conditions, the only potential pathway for significant water, gas and radionuclide transfer. It was confirmed that the coupling between hydraulics, mechanics and geochemistry need to be further investigated, both experimentally and theoretically. The two ongoing projects EC SELFRAC and the Clay Club self healing initiative are considered as relevant steps to better understand the driving processes in EDZ generation and evolution.

More generally, there seems to be a discrepancy of interests between the academic world and the waste management industry. The first is studying regions where processes are active. In contrast, deep geological repository sites are located in areas that have been selected for their high stability and where such processes are quite slow or negligible. The challenge that has to be faced is thus bilateral, i.e. from academic to waste management, transferring knowledge from active areas towards more stable settings, and conversely, generating interest in the academic world for further confirming stability arguments. In this framework, bridging the gaps between the academic community and the waste management community should be set as a key objective of the IGSC initiative on geosphere stability.

The workshop undoubtedly helped to build confidence in the overall long-term stability of argillaceous formations that are considered as potential host for long-lived deep disposal facilities.

COMPILATION OF PAPERS

SESSION I

GENERAL FRAMEWORK: ARGILLACEOUS MEDIA AS HOST FORMATIONS

FUNCTIONS OF ARGILLACEOUS MEDIA IN DEEP GEOLOGICAL DISPOSAL AND THEIR HANDLING IN A SAFETY CASE

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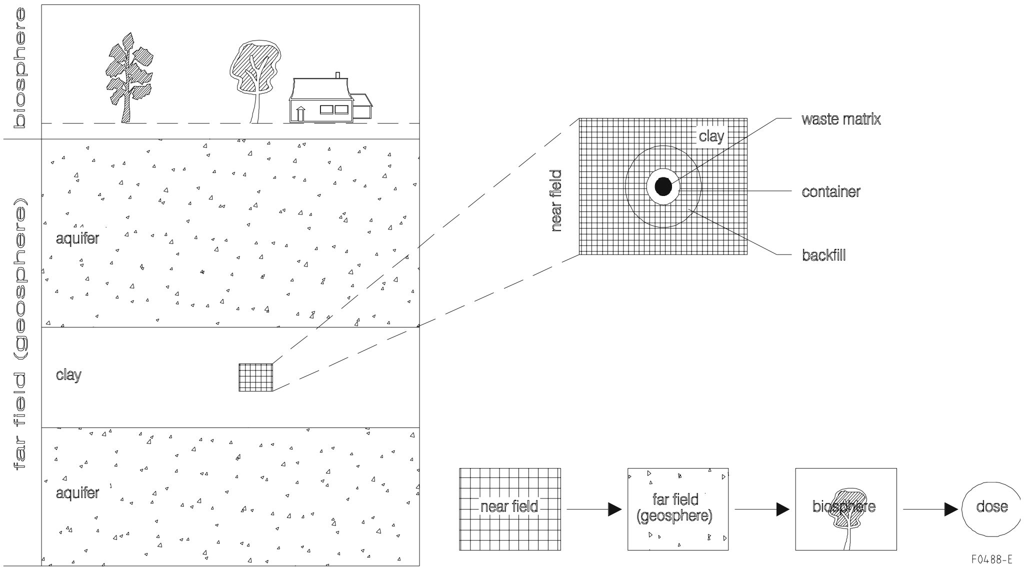
1. Introduction

Geological repositories are designed to protect humans and the environment from the hazards associated with long-lived radioactive waste. The repositories for disposal of high-level radioactive waste (HLW) generally rely on a multi-barrier system to isolate waste from the biosphere. The multi-barrier system typically comprises an engineered barrier system and the geological barrier provided by the repository host rock and its surroundings [1]. A scheme of a multi-barrier disposal system located in a clay formation is shown in Figure 1.

Figure 1. Scheme of a multi-barrier disposal system



Since more then 25 years the possibility to dispose of high level radioactive waste in argillaceous formations is investigated within the European Union [2]. Potential host formations cover a large spectrum of argillaceous media, i.e. from plastic, soft, poorly indurated clays to brittle, hard mudstones. Among the favourable characteristics that are considered for identifying possible host formations are:

- thickness, extent and continuity of the formation;
- hydrodynamic barrier: very low permeability and low hydraulic gradients, resulting in diffusion controlled transport;
- geochemical barrier: high sorption capacity and reducing chemical conditions, favouring low radionuclide solubilities and slow degradation of waste matrices and other engineered barriers;
- low seismic and tectonic activity;
- ability to self-heal fractures and other discontinuities;
- ability to maintain the above mentioned favourable characteristics over a long period of time, i.e. robustness against disturbances due to geological events or the waste disposal.

Meanwhile, detailed site investigation programmes on possible argillaceous host formations were started in various countries, e.g. Belgium, France and Switzerland, and iterative performance assessments, and more recently safety cases [3, 4 and 5] have been elaborated.

2. Time scales to be considered

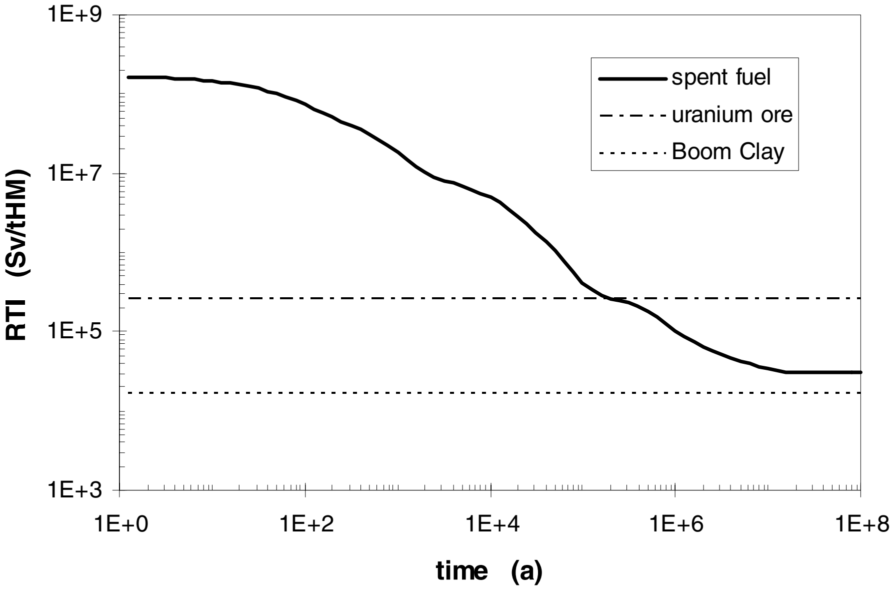
A challenging aspect of geological disposal is that the repository system has to provide protection over timescales that are considerably in excess of those commonly considered in most engineering projects [6]. The evolution of the radiotoxicity index (RTI) of the disposed waste (see Figure 2) can give an indication (orders of magnitude) of the timescales that have to be considered in a safety case. Figure 2 shows that after about 200 000 years the RTI of spent fuel has dropped to the level of the uranium needed to fabricate the fresh fuel ore and after about 1 million years the RTI is comparable to about five times the RTI of the natural radionuclides contained in the host clay formation under the repository site.

A geological repository consists of various engineered and natural barriers, what makes that the repository system is strongly redundant. During its evolution with time some barriers will degrade. However, other more stable barriers, especially the geological barrier provided by the host formation, will remain intact and will continue to fulfil their function.

Starting from the functioning of a repository the following time-scales can be distinguished [7]:

- a thermal phase during which the physical containment of the waste is assured by the intact waste container or overpack;
- an isolation phase during which the slow release from the waste matrix, the limited solubility and the slow transport through the host formation make that most radionuclides remain in the waste package and the host formation, and no release of radionuclides into the surrounding formations occur;
- a geological phase during which the host formation is the main barrier; radionuclides migrate slowly through the host formation and small amounts of radionuclides are released into the surrounding aquifers; dilution in the aquifer and surface waters further reduces the radionuclide concentrations.

Figure 2. Evolution of the radiotoxicity index (RTI) of spent fuel together with some reference values (uranium ore: RTI in uranium needed to fabricate 1 tHM fresh fuel; Boom Clay: RTI in 415 m^2 of Boom Clay, i.e. area needed to dispose of 1 tHM spent fuel)



The interpretation of the results of the long-term radiological safety assessments for a deep disposal system must be based not only on quantitative indicators but also on qualitative arguments. The relative importance of the indicators, e.g. doses and fluxes released into the accessible environment, and the bases of reasoning which accompany them may vary, depending on which phase in the evolution of the disposal system is being considered [3]. For example, in the Finnish regulations time frames are defined on the basis of the predictability of the repository system [8]:

- the environmentally predictable future (several thousands of years) during which estimates of dose can be made;
- the era of extreme climate changes (beyond about 10 000 years) when periods of permafrost and glaciations are expected for which estimates of the fluxes at the geosphere/biosphere interface can be made;
- the farthest future (beyond about 200 000 years) for which statements regarding safety can be based on more qualitative considerations.

3. The role of the geosphere in safety cases

The role of the geosphere and the main safety functions that the geosphere can provide are [8]:

- *Isolation of the waste from the human environment and from various external phenomena:* Placing the waste in a repository located deep underground, with all access routes backfilled and sealed, isolates it from the human environment and thus reduces the likelihood of any undesirable intrusion and misapplication of the materials. The waste is also isolated from various surface phenomena such as climatic events and climate change, erosion and other geomorphological processes.

- *Providing a barrier to radionuclide release and migration:* The very slow groundwater movement makes that diffusion is the main transport process in most argillaceous host rocks. Geochemical retardation and immobilisation further delays the radionuclide transport in the host rock resulting in long travel times and consequent radioactive decay for most radionuclides released from a repository.
- *Providing a favourable chemical and physical environment* that allows other barriers to fulfil their functional requirements.
- *Attenuation of releases into the environment:* Although complete confinement cannot be provided over all relevant times for all radionuclides, release rates of radionuclides from the waste forms are low, particularly from the stable SF and HLW waste forms. Furthermore, a number of processes attenuate releases during transport towards the surface environment, and limit the concentrations of radionuclides in that environment. These include radioactive decay during slow transport through the barrier provided by the host rock and the spreading of released radionuclides in time and space. In the surrounding aquifers radionuclide concentrations are further strongly reduced by dilution and hydrodynamic dispersion.

For long term safety, an appropriate functioning of the host formation is a key element. It is thus essential to limit the risk of reducing the geosphere safety functions by considering "geosphere stability" within the siting process and the site characterisation programme and by limiting as far as possible perturbations that might be induced by the presence of the repository and the waste, e.g. excavation disturbed zone, chemical fronts, thermal effects *(the perturbations induced by the repository are out of the scope of this workshop and will not further be considered in this paper).*

4. Geological events and processes and their handling in a safety case

Safety or performance assessment (PA) is an essential part of a safety case. In general, an early step of such an assessment is the scenario development [9]. Its main objectives are to try to ensure completeness or sufficiency in the scope of PA by identifying a list of relevant features, events and processes (FEPs), and to decide which FEPs to include in PA and how to treat them. This includes screening of less important FEPs, deciding which FEPs are to be treated in quantitative models, which FEPs can be handled by scoping calculations and which FEPs should be regarded as the key defining element or initiating event of separate scenarios. Geological events and processes occur in each category of the before mentioned FEPs.

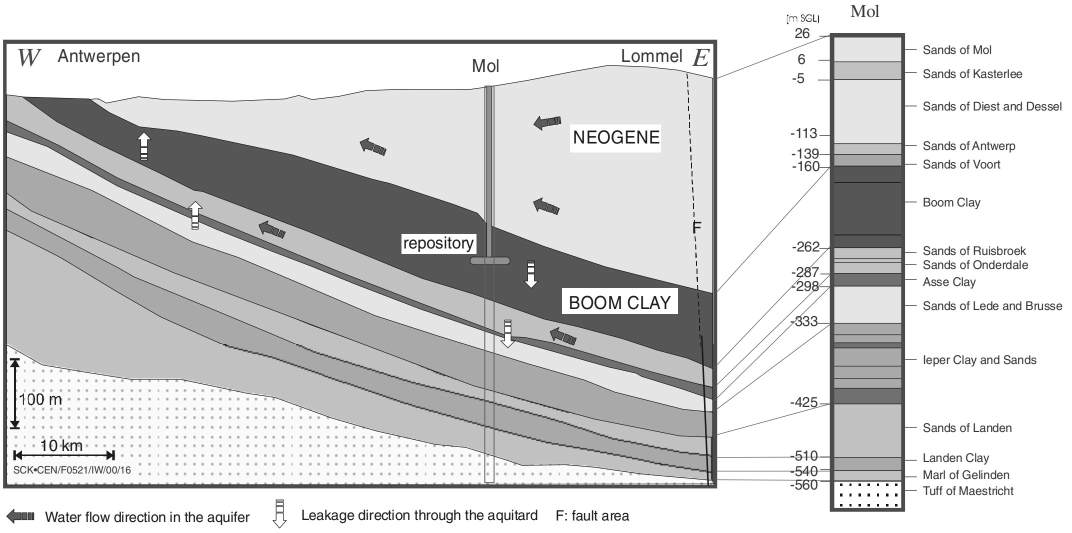
Potential geological host formations, and their surroundings, are chosen in particular for their long-term stability, their ability to accommodate the waste disposal facility, their ability to prevent or attenuate potential release of radioactivity (e.g. through retention capacities), their buffering capacity vis-à-vis external and internal perturbations. Factors that need to be considered when addressing the long-term stability and predictability of a potential repository site include, e.g. tectonics, seismicity, uplift and subsidence history, climate change, formation of glaciers and ice caps, permafrost, erosion and deposition, volcanism, the groundwater cycle, geochemistry, diagenesis, metamorphism and geomechanics.

The likelihood or probability of occurrence of a number of possible perturbing factors can already be reduced during the repository siting process by considering rate of uplift, proximity to major fault zones, proximity to active or inactive volcanoes, etc. During site characterisation, the emphasis may be on determining which phenomena are most relevant to safety, and on determining which scenarios need to be considered in safety assessment and which can be excluded.

5. An example: the Boom Clay at the Mol site

The Boom Clay has been deposited between 33 and 26 million years ago. Between 25 and 20 million years ago deposition and erosion of sand layers took place. Since 20 million years ago progressive burial occurred. The maximum burial depth of the Boom Clay (185 to 285 m at Mol) was reached 2 Ma ago, what is still the present depth of the Boom Clay [10]. A geological cross-section of the Mol site is given in Figure 3.

Figure 3. Geological cross-section of the Mol site



In the following paragraphs a brief discussion of various factors that might affect the stability of the geological barrier and components of the repository system is given.

5.1 Tectonics

During Tertiary north-west Europe has undergone continual subsidence. Over the last 40 years the Mol site has undergone a subsidence of about 30 mm. Although it is difficult to extrapolate observations over short time periods, it was concluded that the Mol area is subject to a general subsidence that is likely to continue into the future.

At a distance of about 30 km from the Mol site occurs a complex fault system that is known as the Roer Valley Graben. The number and the throw of faults in the area decreases with the distance from the Graben. A fault located at about 5 km from the considered site was active during Quaternary and the mean movement was estimated to be about 1 m every 100 000 years. The results of a recent seismic campaign are expected to provide more information on the location, depth, throw and shape of faults in the Mol area. The repository should not be located above an existing fault.

5.2 Climate change

Until a few years ago simulations of future climate were essentially based on Milankovitch's orbital theory [11]. A moderate glaciation is expected to occur after 24 000 years and a more severe one after 56 000 years. However, a more recent simulation that takes greenhouse effects into account concluded that the occurrence of these glaciations is questionable [12].

During Quaternary no glaciers or ice caps have occurred at the Mol site. Consequently their probability of occurrence during the next 200 000 years is extremely low. On the other hand the lowering of the sea level and the occurrence of permafrost can have a considerable impact on the regional hydrogeology. Simulations of the impact of future glaciations on the behaviour of the repository system were carried out by applying paleogeographical information from the Weichselian glaciation [13]. The main impact for dose calculations was a considerable reduction of the dilution of the radionuclide concentrations in the aquifers, because permafrost strongly hinders the infiltration into the aquifers.

A combination of the above mentioned subsidence with a rise of the sea level due to greenhouse effects could result in a transgression of the sea above the Mol site. This can lead to deposition of new sedimentary layers but also to the occurrence of marine erosion channels [14]. However, for the evaluation of the possible radiological consequences an important aspect is that men will not live at the site as long as it is covered by the sea. During the first period after regression of the sea the salt concentration of the groundwater will limit the use of water from the aquifers surrounding the host formation.

5.3 Geochemical evolution

Since more than 1 million years infiltrating fresh water is washing out saline water from the overlying Neogene aquifer and from the underlying Under-Rupelian aquifer, in the latter at a much lower rate because of the low hydraulic conductivity and the complex geological structure of this aquifer. For the present day conditions geochemical studies have been started to study the geochemical equilibrium between the clay minerals and the fresh water, the presence and evolution of organic matter [15] and of the high bicarbonate concentration of the interstitial Boom Clay water. The possible impact of the return of saline interstitial water, in case of a sea transgression, on the clay minerals and on migration parameters will also be evaluated.

6. Conclusions

In case of geological disposal of HLW in argillaceous formations the geological barrier provided by the host formation is the main barrier of the disposal system. It is also the only barrier for which it is expected that it will continue to function over the timescales required for geological disposal of HLW. However, clay formations can be subject to alteration processes and geological events that can drastically modify the characteristics of the formation.

Potential geological host formations and repository sites are chosen for their long-term stability, i.e. natural phenomena that might perturb the geological barrier provided by the host formation should have a very low likelihood of occurrence. Also during the design and construction phases of a repository precautions are taken to limit as far as possible the various perturbations of the host formation.

Safety cases already treat, at least partially, the expected as well as the less probable, but possible, evolutions of the geological components of the repository system. However, it is of paramount importance for strengthening the confidence of authorities and stakeholders in a safety case that the approaches applied in the evaluations are in agreement with the most recent developments and findings of Earth scientists.

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REGULATORY EXPECTATIONS CONCERNING THE GEOSPHERE CHARACTERISATION FOR DISPOSAL IN ARGILLACEOUS FORMATIONS

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Safety recommendations on geosphere stability depend on the phase considered in the development of a disposal project.

As far as deep disposal is concerned, safety generally relies on a multiple safety functions or multiple barriers concept for which each component of the repository i.e. waste package, engineered barriers and geological barrier, play complementary roles with regard to global safety. Consequently, depending on the disposal design, different types of favourable geological media can be envisaged for siting a repository. Thus, requirements on geosphere stability may be diverse when selecting a site for disposal since different options can be considered, and regulatory expectations in this field are more based on implementing a global and sound safety approach than on conforming to quantitative criteria.

It is however obvious that geological formations presenting good containment properties and evidences of very long term stability should preferably be selected since it facilitates the safety demonstration helping building confidence in it. It also offers flexibility in the choice of design options and on the performances required from the different components of the disposal facility. Such an approach was recommended in France and led, for the site selection phase, to focus on general requirements on the quality of the geological formations to select. Based on these requirements, an evaluation is made according to an incremental procedure, to state whether committing resources for further development of the project is appropriate.

The French Basic Safety Rule III.2.f (BSR III.2.f) provides some qualitative requirements regarding preferred site properties. It recommends in particular that the geological barrier must provide in the long-term adequate radionuclide isolation capability and should play a key role over the long term.

The BSR III.2.f defines technical criteria i.e. essential and important criteria to help selecting a potential site for disposal. The essential criteria relate to both the stability of the site and its hydrogeology whereas important criteria concerns properties of the host rock, depth requirements and the eventual presence of underground resources.

BSR III.2.f states that:

> "The stability of the site must be such that any change in the initial conditions due to geological phenomena that may occur (glaciation, seismicity and neotectonic movement) will be acceptable in terms of the safety of the repository. In particular, for a period of not less than 10 000 years, stability (covering limited and foreseeable evolution) must be demonstrated. When a site is investigated these phenomena must be assessed qualitatively

> and quantitatively with reference to the present situation, the recent past (historic) and above all the more distant past.
>
> The hydrogeology of the site must be characterised by a very low permeability of the host formation and a low hydraulic head gradient. Moreover, for the formations surrounding the host formation, a low regional hydraulic gradient will also be preferable. Hydrogeological characterisation must be made over an area far wider than that of the repository site so as to be able to construct flow models allowing for movements from the supply zones to the discharge zones. These regional patterns should make it possible to simulate the intensity and the direction of the groundwater movements.
>
> Finally, concerning hydrogeology, it will be necessary to make allowance for any instances of discontinuities or heterogeneity of which nature or geometry might significantly reduce the effectiveness of the geological barrier. Such features must be identified and characterised with the greatest care so as to ensure that they are avoided at the site if necessary.
>
> Important criteria involve host rock properties that are important to characterise due to their role in the feasibility of the repository. Mechanical and thermal properties determine the possibility of designing a repository which does not significantly deteriorate the geological barrier. Studies, particularly with the assistance of modelling of the combined effects of the thermal and mechanical phenomena, must be carried out to investigate the influence of the mode and sequence of the emplacement of the waste on mechanical effects in the repository.
>
> Geochemical properties play an important role in the long-term safety of a radioactive waste repository as they can affect the deterioration of the artificial barriers and they govern the phenomena of retention of any radionuclides released. Mineralogical properties of the host formation materials must be carried out and their geochemical evolution assessed.
>
> Compliance with a minimum depth requirement implies that the site must be chosen so that the planned depth of the repository guarantees that the isolation performance of the geological barrier is not significantly affected by the erosion phenomena (particularly after a glaciation), by the effect of an earthquake or by intrusion. It may be assumed that the thickness of the superficial zone liable to be disturbed in this manner is in the order of 150 to 200 meters. Finally, for sub-surface management purposes, the site must be chosen to avoid areas where the known or suspected importance is of an exceptional nature."

Once the site is selected, the next step of the approach corresponds to a phase of confirmation of the potential favourable characteristics of this site through an enhanced characterisation of the geological barrier, which is strongly connected to the disposal design.

The objective is to highlight potential weaknesses and assets of the geological barrier in terms of containment properties and then to adapt disposal design options in order to reach the best level of safety that can be achievable.

The approach is driven by an iterative process between disposal design studies and host rock characterisation through field site data acquisition.

At the stage of these feasibility studies for disposal, the objective of the approach is to gain a better knowledge of the possible radionuclide migration pathways in the geological formation and to assess the capacity to restore the initial favourable properties of the host rock, that are inevitably disturbed by the presence of a disposal.

To that respect, and in a first step, focus is given on the detection of initial weaknesses of the host formation, especially fracturing and its potential hydraulic role as well as excavations-induced effects (EDZ). More generally, the aim is to understand fluid pathways, not only past ones but also those associated with a given disposal design (possibility of internal connections).

In a second step, the approach consists in being able to progressively integrate such knowledge in safety assessments i.e. to suggest what may be the various fluid circulation patterns in future for the global system especially in the discontinuities, taking into account their possible solicitations (external events such as seism, glaciations and internal effects such as mechanical arrangements).

Finally, in a third step, the objective of the approach is to propose and elaborate disposal design options taking account of the possible weaknesses of the formation and the remaining uncertainties associated to its characterisation. Such design options should minimise the effects of these weaknesses and uncertainties on the global safety of the facility. As an example, BSR III.2.f indicates that “the evaluation of the disturbance caused by the creation of the repository must be realised and one should check that these disturbances remain acceptable in term of the safety level chosen for each of the barrier, particularly the geological barrier”.

The overall objective of the approach is to propose at least one disposal design, technically conceivable, which presumably allows reaching the objective of protection of human health and the environment.

SESSION II

EXOGENIC AND ENDOGENIC PROCESSES

AN OVERVIEW OF CLIMATE CHANGE

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We describe briefly here the main mechanisms and time scales involved in natural and anthropogenic climate variability, based on quantitative paleoclimatic reconstructions from natural archives and climate model simulations: the large glacial-interglacial cycles of the last million years (the Quaternary), lasting typically a hundred thousand years, triggered by changes in the solar radiation received by the Earth due to its position around the Sun; the century-long climatic changes occurring during last glacial period and triggered by recurrent iceberg discharges of the large northern hemisphere ice caps, massive freshwater flux to the north Atlantic, and changes in the ocean heat transport. We show the strong coupling between past climatic changes and global biogeochemical cycles, namely here atmospheric greenhouse gases. We also discuss the decadal climatic fluctuations during the last thousand years, showing an unprecedented warming attributed to the anthropogenic greenhouse gas emissions. We show the range of atmospheric greenhouse concentrations forecasted for the end of the 21st century and the climate model predictions for global temperature changes during the 21st century. We also discuss the possible climatic changes at longer time scales involving the possibility of north Atlantic heat transport collapse (possibility of abrupt climate change), and the duration of the current interglacial period.

1. Natural climate variability: glacial-interglacial cycles of the Quaternary

About 21 000 years ago, the sea-level was ~120 m lower than for present-day, due to an intensive build-up of ice on the northern hemisphere continents with huge North American and North European ice sheets [1]. During this period called the Last Glacial Maximum, Greenland temperatures were 20°C colder [2], Antarctic temperatures 9°C colder [3, 4] and European temperatures about 15°C colder than modern annual means [5]. Huge reorganisations of the oceanic circulation (with larger sea-ice cover) [1] and biosphere types (such as steppe replacing forest over large areas of the mid latitudes) [6] resulted in reduced atmospheric greenhouse gas concentrations (200 ppm of CO_2 versus 280 ppm for interglacial periods; 400 ppb of CH_4 versus 700 ppbv for interglacial periods) [7]. Drier and windier conditions induced huge levels of dust in the atmosphere [7, 8]. Climate models estimate that this dramatic cold climate corresponds to a global temperature cooling of 5°C [9].

Typically warm interglacial conditions occur during ~10 ka and their end corresponds to a progressive glacial inception lasting about ~90 ka and culminating to "glacial maximum" conditions. The forcing (factor external to the climate system) triggering changes between glacial and interglacial "modes" of the climate system is the solar radiation received by the Earth, resulting from changes in its orbit around the Sun [10, 11]. Eccentricity, obliquity and equinox precession result in insolation periodicities of 100, 41, and 19-23 ka. The current understanding of glacial cycles lies in sequences of events involving the insolation forcing (Milankovitch theory) and climate system feedbacks involving icy surfaces albedo and greenhouse gases. At the end of an interglacial period, a marked decrease of summer solar radiation at 65°N induces a summer cooling, amplified by the persistence of snow cover all year round. The resulting change of vegetation (tundra replacing boreal forest) and ocean circulation (more sea-ice) enhances this albedo feedback [12]. The decrease of wetlands both at polar and tropical locations induces a decrease of CH_4 atmospheric concentrations [13]. Both the cooling and the slowdown of the oceanic circulation reduce the atmospheric CO_2 levels [14]. The reduced greenhouse gases (less CO_2 and CH_4, less water vapour in a cold atmosphere) amplify the cooling leading to the buildup of ice sheets and the glacial climatic conditions. By contrast, the deglaciation processes start in the southern hemisphere that warms up, followed after a few centuries by increased atmospheric CO_2 levels [15], destabilisation of the northern hemisphere ice caps and positive greenhouse and albedo feedbacks.

2. Natural climate variability: abrupt climatic changes

During glacial periods, high resolution paleoclimatic records measured on Greenland ice cores have revealed the existence of a succession of 25 abrupt climatic changes (called Dansgaard-Oeschger events) [16]. They correspond to Greenland temperature changes of typically 10°C to 15°C [17] occurring during several decades to centuries and some of them have recently been identified in high resolution speleothem records of European and Chinese climate [18, 19]. In parallel, North Atlantic deep sea records show changes in deep ocean circulation and reveal among the deep sea sediments the presence of ice rafted debris (rocks and mineral fraction) [20, 21]. The simultaneous changes in atmospheric CH_4 concentration [22, 23] reveals the hemispheric scale of the associated climatic changes. The current understanding of these rapid events is an instability of major ice caps in the northern hemisphere, massive iceberg discharges to the north Atlantic, bringing light freshwater and changing the density of north Atlantic surface waters. Reduced deep water convection and reduced heat transport by the ocean induce large-scale cooling in the northern hemisphere and changes in atmospheric circulation including the tropical areas, and at the same time a warming of the southern hemisphere, in response to an oceanic “see saw” [24]. After a while, the ocean circulation starts again, warming the north Atlantic and cooling the southern hemisphere [25, 26]. These rapid changes directly influence European climate as testified by simultaneous changes in pollen distributions [27]. The largest coolings could have been associated with persistent permafrost in south western France as indicated by hiatus in well-dated speleothem growth [18]. The last of these events took place about 8.2 ka BP (during our interglacial period) and corresponds to a cooling of about 2-5°C and lasting about 40 years [28, 29].

3. Natural and anthropic climate variability: last millennium

The instrumental records of meteorological observations enable hemispheric temperature change estimates for about 150 years. Due to a huge level of interannual variability in the climatic parameters, such records remain short to detect slow centennial scale fluctuations or trends. Reconstructions of northern and southern hemisphere temperatures have been compiled for the last thousand years on an annual basis using multiple proxies of climate change calibrated against meteorological records, including specifically long tree ring chronologies [30]. Historical records of environmental conditions are also heavily used for climate reconstruction purposes [31]. The Little Ice Age, from about A.D. 1350 to A.D. 1860, is the last of the centennial scale climatic fluctuations which is well known from historical documentation. Such a slightly colder time period (about 0.6°C colder in the northern hemisphere) corresponds to dramatic environmental changes in vulnerable areas such as Alpine high altitude places, where glaciers advanced significantly [32]. These small temperature changes can be associated with large changes in the hydrological cycle. Several studies have revealed a dramatic change in the seasonal precipitation cycle at the end of the Little Ice Age in France. A study conducted on tree ring cellulose isotopes in western France has shown that during the past four centuries, the frequency of summer droughts doubles when mean summer temperatures increase by half a degree [33]. At these time scales, the external forcings of climate are mainly solar irradiance, stratospheric volcanic eruptions, land-use and anthropogenic greenhouse gas and sulfate aerosol emissions [34]. Only since about 1985 does the radiative forcing of greenhouse gases overwhelm the impacts of natural forcings.

Human activities modify the land surface (for instance through deforestation and agriculture, building of large cities, …), the land-amosphere moisture and energy exchanges, which may have large scale impacts on climate [34]. The intensive use of fossil fuels results in dramatic increases of atmospheric greenhouse gas concentrations (with long residence times: ~10 years for CH_4, ~100 years for CO_2), and in sulfate aerosols. Sulfate aerosols have short residence times in the atmosphere, a direct radiative effect (back diffusion of solar radiation) and an indirect radiative effect (they act as cloud condensation nuclei). Reconstructions for the last millennium and climate simulations show that the ongoing global warming cannot be explained without taking into account both natural and anthropogenic forcings [34].

4. Future climate changes: next century

About half of the anthropogenic CO_2 emissions accumulate each year in the atmosphere, leading to the observed exponential increase; the other half of the human emissions is stored in the biosphere and the ocean, with decadal to centennial time constants. Even though all greenhouse gas emissions are stopped tomorrow, the atmospheric CO_2 level will continue to rise during several centuries to concentrations of typically 500 ppm [34]. However economists have calculated scenarios of greenhouse gas emissions taking into account fossil fuel availabilities and under different international frames ranging from "virtuous" scenarii where alternative technologies and strong emission reduction policies induce a stabilisation of the emissions; to "business as usual" scenarii where fossil fuels (fuel, gas, coal) are used at the rate of modern economical growth and development. The atmospheric concentration of CO_2 resulting from these calculations lies between 500 and 800 ppm at the end of the 21st century. All climate models therefore forecast a rise of the Earth temperature ranging between 2 to 6°C at this horizon [34]. This change in temperature will be about at the global mean in western Europe (typically 2-3°C in France) and will be associated with changes in precipitation seasonality in Europe (more rainfall in winter and spring and significantly less in summer). This could result in an increased frequency of winter floods and summer droughts during the 21st century. Sea-level should also increase by several tens of centimeters due to thermal dilatation and melt of small glaciers [34].

5. Future climate changes: next thousands of years

During the next millennium, abrupt changes in north Atlantic thermohaline circulation may take place in response to increased freshwater supply through intense cyclogenesis and rainfall on this key area; or due to melting of the Greenland ice cap [35], which would result in a 6 m increase in sea level. Within a fast global warming context, local abrupt events such as the collapse of the thermohaline circulation may induce a local cooling affecting the North Atlantic area and therefore Europe.

At longer time scales, the Earth orbital parameters will again play a key role in the pacing of glacial inception. The summer insolation at 65°N does not exhibit clear minima during the next 50 000 years. If the mechanisms operating during the Quaternary remain the same for the next thousands of years, it is not expected that any glacial period starts within the next 50 thousand years [36]. Taking into account anthropogenic emissions of CO_2 and its fate in the climate system (ocean pumping active at 10^2 years, carbonate processes active at 10^3 years and geological silicate erosion processes active at 10^4 years), simplified climate models have shown that the human induced greenhouse gases levels should prevent the onset of the next glaciation until 200 000 years in the future, resulting in the longest warm interglacial periods ever known during the last million years.

Figure 1. The climate system

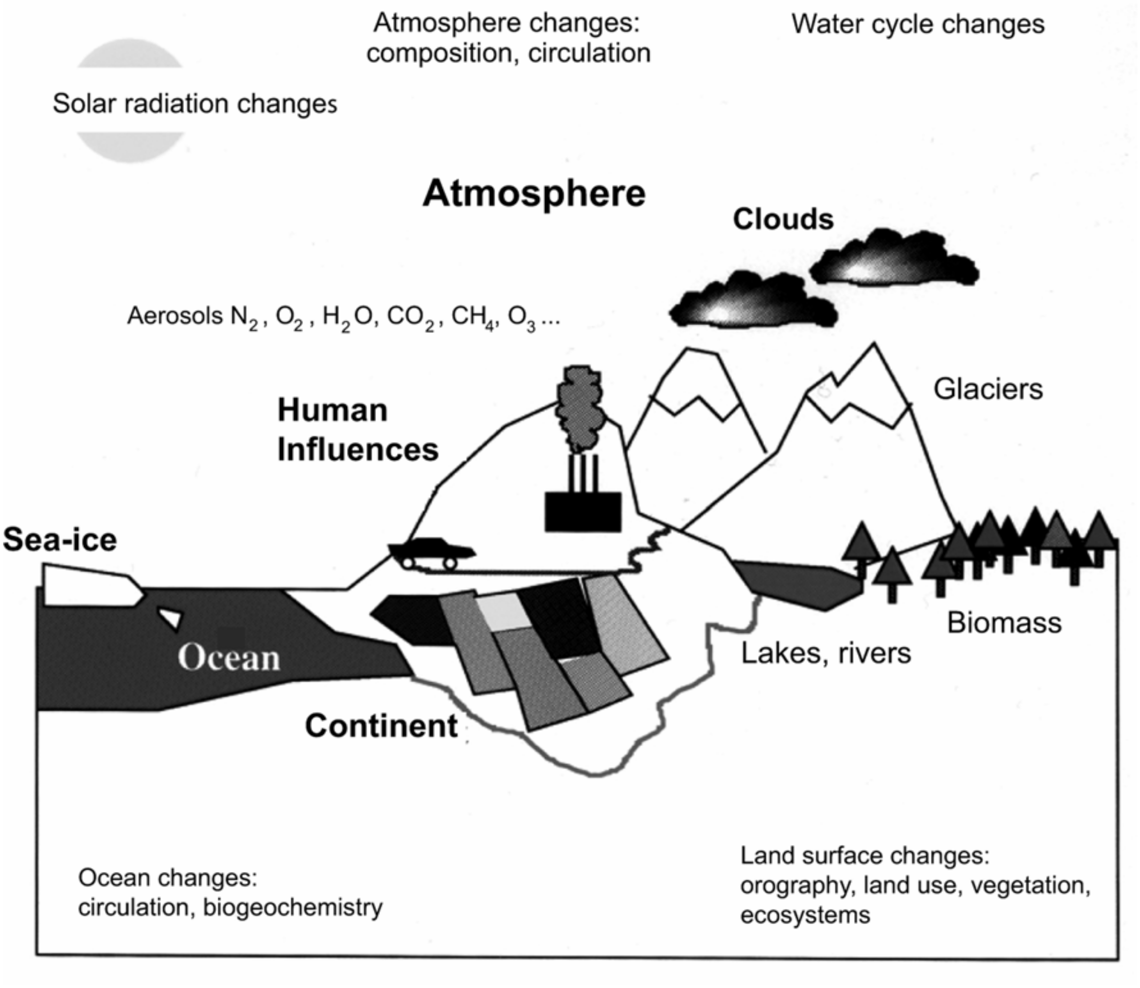
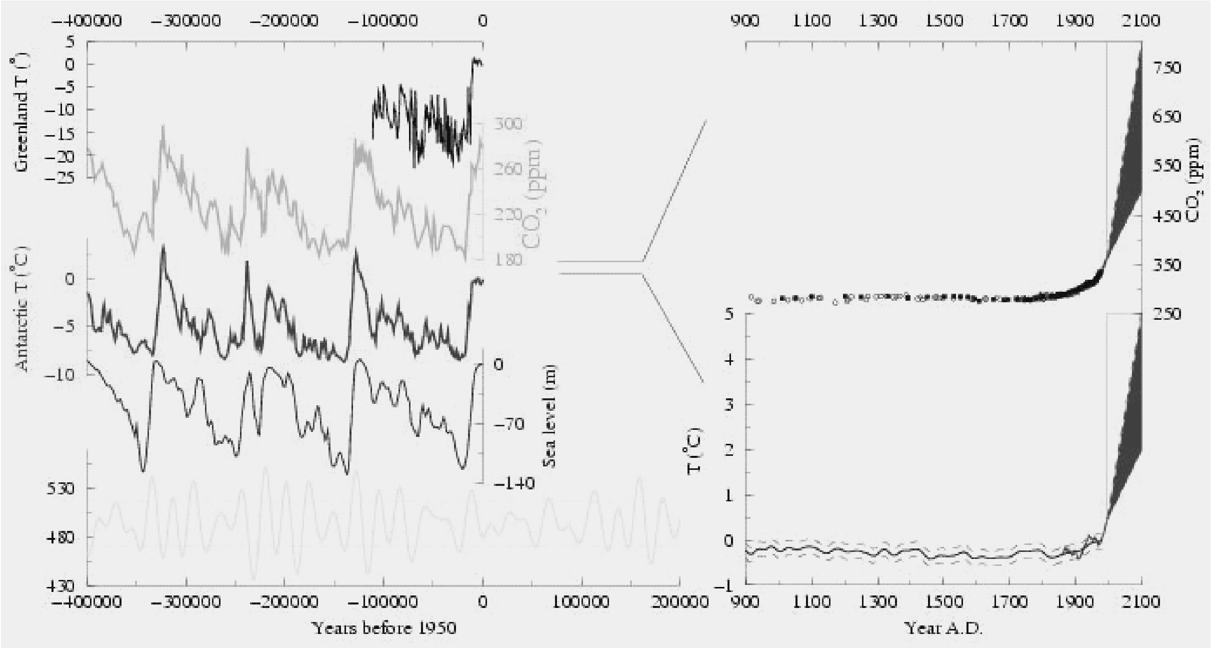


Figure 2. Four glacial-interglacial cycles



(top left panel)	Greenland temperature derived from GRIP ice core $\delta^{18}O$ corrected for changes in snowfall seasonality [37].
(second left panel)	Atmospheric CO_2 fluctuations from Vostok ice cores [7].
(third left panel)	Antarctic temperature derived from Vostok δD [4].
(fourth left panel)	Ice volume recorded by the $\delta^{18}O$ of benthic foraminifera from deep sea sediments [38].
(fifth left panel)	Past and future changes in incoming summer (June 21st) solar radiation at 65°N [39].
(top right panel)	CO_2 fluctuations during the last millennium [40] and projections for the ongoing century [34].
(bottom right panel)	Climatic changes during the last millennium [30] and projections for the ongoing century [34].

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SURFACE EROSION IN OROGENIC SYSTEMS

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1. Introduction

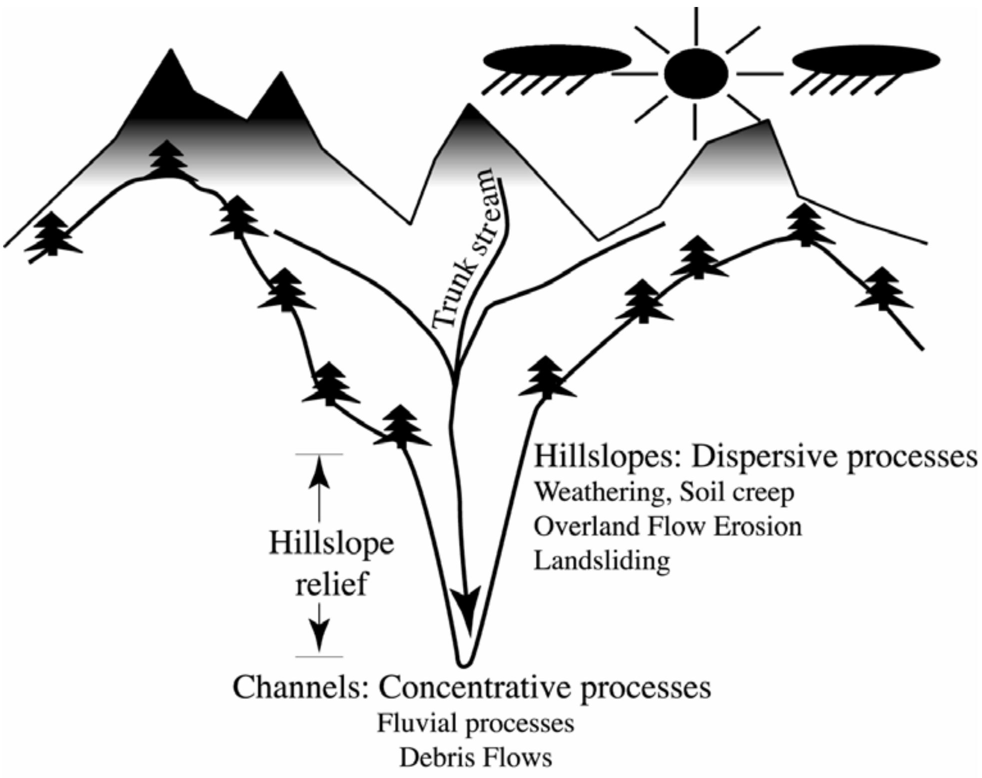
Understanding the effects of climate and tectonics on the geomorphic evolution of mountain belts has become the major research objective in surface processes. In addition, studies have been directed towards exploring possible controls on the local and the global surface mass flux. These objectives can only be addressed if the different components of sediment routing systems are known, and if possible feedbacks between the various erosional mechanisms have been analysed. Accordingly, in a first step, this paper illustrates the nature of surface erosion for different spatial and temporal length scales. In addition, it explores how the landscape evolves from initially simple conditions to more mature stages, and how and to what extent the different components of surface processes influence the topographic development. If this issue has been discussed from a more general point of view, the paper will illustrate how and to what extent climate and crustal processes influence different erosional components and topographic length scales. Finally, it will present quantitative data on current rates of surface erosion for the situation of the Swiss Alps.

2. The nature of landscape evolution

It is generally accepted that except from glacial erosion, long-term (>1 000 years) erosional processes consist of a concentrative and a dispersive component (Figure 1). The concentrative nature of erosion and sediment transport comprises fluvial processes and debris flows in channels. Erosion and sediment transport by fluvial processes then occurs if critical flow strengths are exceeded. In contrast, sediment is transported as debris flows if slopes and pore pressures surpass specific thresholds. The dispersive component of erosion and sediment transport comprises processes on hillslopes such as soil creep that is often referred to as hillslope diffusion in modeling studies (e.g. Tucker & Slingerland, 1997), overland flow erosion and landsliding if critical values for slopes and/or pore pressures are exceeded (e.g. Anderson, 1994).

Despite these general agreements, there are differences in interpretations of how and to what extent these components drive the evolution of landscapes. Some researchers consider fluvial erosion to exert the first-order control because fluvial processes are anticipated to set the lower boundary conditions for geomorphic processes on the adjacent slopes. Accordingly, increasing (or decreasing) rates of fluvial erosion result in equivalent modifications of the local relief and hence in enhanced (or reduced) rates of erosion on the adjacent hillslopes. Alternatively, it was proposed that rates of hillslope erosion do not necessarily have to be controlled by erosion in channels. For instance, Hovius *et al.* (1998) found that hillslope mass wasting at the channel heads govern the rates and the mode of drainage basin modification in the Finisterre Mountains, Papua New Guinea.

Figure 1. Overview of erosional processes



Numerical experiments and field studies carried out in humid (Central Alps, Switzerland) and arid/hyperarid climatic settings (western escarpment of the Andes of northern Chile and Peru) reveal that the controls on surface processes significantly depend on the ratio D_e between the concentrative (fluvial and debris flows) and dispersive (hillslope) characteristic time scales, i.e. on the relative strength of concentrative and dispersive processes (Simpson & Schlunegger, 2003). These studies suggest that fluvial and debris flow dominated systems (high D_e) are characterised by rough, high-relief, highly incised surfaces which contain a dense and hence a closely-spaced channel network, a thin regolith cover and highly sinuous valleys. A classical example is the Napf area of Central Switzerland. Geomorphologies resulting from low D_e-values generally have a thick regolith cover. They display smooth topographies, low reliefs, greater channel spacing and less sinuous valleys. Examples are the densely vegetated regions of the western escarpment of the northern Peruvian Andes.

3. Topographic evolution

The topographic evolution of landscapes tends to occur in two major phases reflecting formation of new channels and valleys, and formation of relief, and a later decay phase related to the gradual destruction of channels and valleys. During this initial growth phase the location of fluvial incision rapidly propagates headward thereby increasing the local relief and the topographic roughness. Besides the modification of relief the phase of backward erosion is characterised by the formation of well-recognisable knick-points in the longitudinal stream profiles, separating landscapes that still record the initial (usually smooth) topography in the headwaters from a geomorphology where the surface area and the topographic roughness grows as the erosional front passes. Examples can be found in the Andes of Chile and Peru, and in the Swiss Alps. Specifically, in the Andes, enhanced rates of rock uplift at ca. 8 Ma initiated a phase of backward erosion and relief rejuvenation. Similarly, in the Swiss Alps, the base level drop of >80 m that was caused by the retreat of the Alpine glaciers at ca. 15 Ka initiated a phase of downcutting and backward erosion of the Alpine rivers, causing the local relief to grow (Schlunegger & Hinderer, 2003). At these knick-points effective erosion rates are up to 50 times higher than the overall denudation rates in the Alpine and Andean drainage basins.

Theoretical models imply that the relative strength of concentrative and dispersive processes, i.e. D_e, also exerts a significant control on the topographic evolution of mountain belts (Simpson & Schlunegger, 2003). An increase in D_e (which is equivalent to an increase in rates of fluvial erosion and sediment transport) results in a decrease in the timescale within which the topography develops. In this case, the topographic development is characterised by increasing rates of backward erosion and vertical incision, and by an augmentation of the topographic roughness and the local relief. In contrast, an increase in the importance of hillslope processes on the geomorphic development (decrease in D_e) results in a smoothing of the topography and in a decrease in the local relief, the overall denudation rates and the associated surface mass flux.

4. The influence of climate on surface erosion

Climate influences the geomorphic development by production of unconsolidated material through weathering and landsliding, and by transport of sediment through linear processes (such as debris flows and fluvial processes). Observations in the Andes imply that a warm and humid climate enhances weathering rates, which, in turn, results in an increase in the relative importance of dispersive surface processes and hence in a decrease in the topographic roughness, the magnitudes of the local relief and the density of channels and valleys. In contrast, it has been reported that the nature and rates of surface erosion changes significantly if climate becomes more arid. Specifically, a shift towards an arid climate tends to increase the ratio between large- and small-magnitude floods (Turcotte & Green, 1993). In a conceptual publication about climate change and surface eroison, Molnar (2001) thought that an increase in this ratio results in accelerated erosion. The rational behind this argumentation lies in the assumption that a river needs to transport its bedload before the bedrock becomes eroded, and that critical flow strengths must be exceeded to remove this bedload. It appears then that an increase in the frequency of large floods (caused by a shift towards a more arid climate) tends to increase the magnitudes and the frequencies at which these critical thresholds are surpassed. Molnar (2001) then concluded that such a scenario tends to increase the surface mass flux.

Field observations suggest that an increase in aridity tends to decrease weathering rates and hence production rates of regolith. According to the theoretical concepts outlined above, such a climatic shift is anticipated to result in an increase in the relative importance of concentrative (debris flows, fluvial erosion and sediment transport) versus dispersive processes (hillslope processes) and hence in an increase in the channel density, the topographic roughness and the local relief. Such a shift in the erosional regime will also decrease the topographic length scale of the system and hence the rate at which the topography responds to changes in external perturbations. The conclusion of Molnar that an enhanced aridity will increase the erosional mass flux is thus consistent with the results of numerical models even if they are based on different conceptual approaches. In contrast, however, preliminary geochemical and sedimentological data from the western escarpment of the Andes of Chile imply that denudation rates increase with precipitation rates. This increase, however, is currently being documented at a hillslope scale. This does not mean that rates of sediment also increase at the scale of whole drainage basins. Further research is needed to explore this open question.

5. The influence of rock uplift on surface erosion

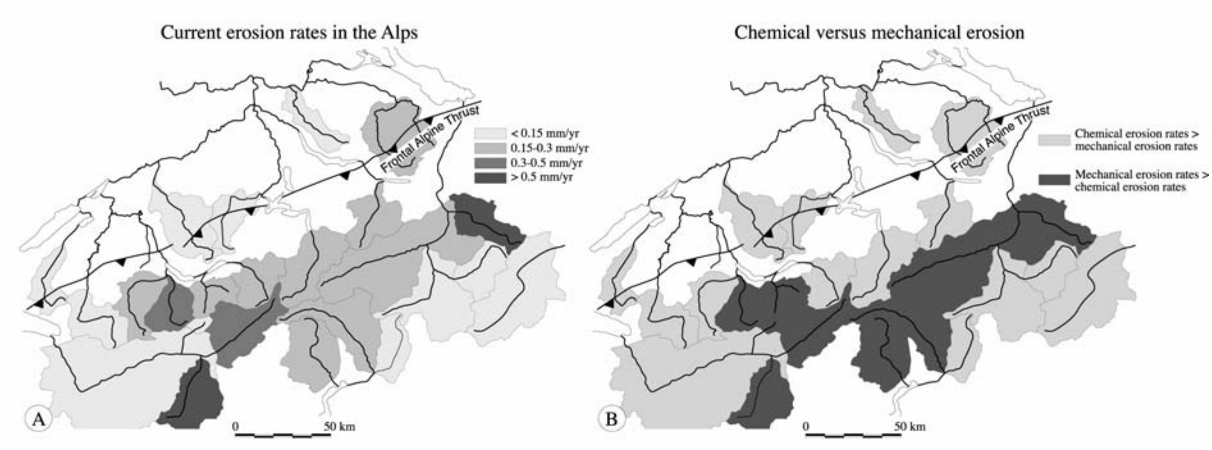
Rock uplift influences erosional processes by an increase in the gradients. Observations from the Andes and from the Central Alps reveal that the erosional systems respond to an increase in rock uplift rates (which is equivalent to a lowering of the base level, or by an overall steepening of the topography) by headward erosion. This phase of backward erosion is characterised by the presence of well-identifiable knick-points in the stream profiles. Beneath the knick-points, the drainage basins have adjusted to the modified pattern of rock uplift. The areas surrounding the knick-points currently experience a phase of relief-rejuvenation as incision has resulted in growth of the local relief. The geomorphologies in the headwaters, in contrast, still record the situation prior to the enhancement of rock uplift. It appears, therefore, that any increase in rock uplift rates (or in base level lowering) will ultimately increase the local relief especially in the segment of active backward erosion. As backward erosion proceeds the location of relief-rejuvenation shifts towards the headwaters. In contrast a reduction in the rates of rock uplift will result in a decrease in fluvial process rates, which, in turn, reduces the ratio between the concentrative and dispersive processes. The result is an overall decrease in the topographic roughness and the local relief.

6. Modern erosion rates in the Central Alps and the importance of bedrock lithology

Modern denudation rates are determined by measurements of sediment loads (mechanical and chemical) of rivers in relation to the size of the whole drainage basins. The data show that total denudation rates increase from ca. 0.15 mm/yr in the distal foreland to ca. 0.5 mm/yr in the Central Alps. Exceptionally high denudation rates of >0.5 mm/yr are observed for drainage basins that are heavily glaciated, and for systems that contain source rocks with low erosional resistance (e.g. Flysch units) (Figure 2A). The tendency towards enhanced average surface erosion from the foreland to the Alpine core is mainly due to the enhanced contribution of mechanical denudation. In contrast, total denudation rates calculated for the drainage basins in the foreland are dominated by chemical denudation reaching up to 90% of the total. Similarly, erosion rates by chemical denudation increase with the relative abundance of carbonates in the source area. The sediment loads imply mean chemical denudation rates of ca. 0.085 mm/yr for systems with predominantly crystalline sources, and rates of ca. 0.125 mm/yr, and 0.105 mm/yr, with catchments made up of carbonates and mixed lithologies, respectively (Figure 2B).

The sediment yields presented in Schlunegger & Hinderer (2003) (Figure 2A) have been calculated based on the prerequisite that erosion operates on the whole surface. However, detailed mapping in Central Switzerland revealed that post-glacial and modern erosion have only modified 30-50% of the surface that formed during glacial times (i.e. 50-70% of the glacial landscape has not been affected by erosion). Specifically, Schlunegger & Hinderer (2003) revealed that modern erosion appears to operate predominantly in V-shaped scars and on the associated hillslopes. In addition, the analysis of digital elevation models in combination with mapping suggest that the ca. 50-70% estimate of non-eroded glacial surface also applies to other portions of the Swiss Alps. Consequently, considerations that chemical erosion affects the whole drainage basin, and mechanical erosion occurs by rivers in the V-shaped scars will result in effective modern incision rates that are for some basins >50% higher than the average yields of Figure 2A.

Figure 2. (A) Total modern erosion rates for selected drainage basins, and (B) the relative importance of chemical dissolution on the erosional mass flux (Modified, Schlunegger & Hinderer, 2003)



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FAULTING AND HAZARD IN LOW SEISMICITY AREAS

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Extended abstract

The relationship between faulting and seismicity in areas of low to moderate seismicity, such as Northern Europe, tends to be obscure. Thus, concepts in hazard analysis originating in the context of active tectonic environments such as California may not be readily applicable. A classic example is the search for "active" faults, where the standard definition of an active fault follows from that laid down by USEPA (1981) along the lines that any fault that can be shown to have produced an earthquake in the last 10 000 years is so categorised. Although one can, and in some circumstances must, evaluate a list of local mapped faults to assess their seismic capability, there is a danger of overlooking the fact that all earthquakes must occur on some fault. Thus, in a country like the UK where around 300 events are detected every year, the number of faults that are "active" by the USEPA definition must also be in the hundreds, although one is in the position of not being able to name a single one of them with complete confidence.

As a result, focussing on a few cases where one can, with some reasonably high probability, suppose individual faults to be associated with certain earthquakes, doing so runs the risk of presenting a skewed picture of the seismicity, by treating some earthquakes differently from others because they can be connected to a mapped fault, whereas in seismological terms, an earthquake that cannot be so connected nevertheless has the same physical properties.

There is some merit in abandoning the idea of active and inactive faults in favour of a three-way classification, more suited to environments in which most fault movement occurs as reactivation of extremely old structures in a tectonic environment different from that in which they originally formed.

Inert faults are those that appear to have no possibility of reactivation in the present stress regime. Such faults have been classed as "extinct" by Muir Wood and Mallard (1992), but the term "inert" avoids inapt comparisons with volcanology.

Reactivatable faults are those that could be reactivated or have been reactivated. Reactivation is seen as a diffuse and largely disorganised process within a particular crustal volume.

Controlling faults are those that are the main expressions of continuing deformational activity, e.g. graben-bounding faults in areas of present extensional tectonics.

Both of the last two categories may be "active" in the sense of hosting modern earthquakes, but a fault in the latter category can be seen as the preferential locus for future movement, and thus future seismic activity along it is certain to occur. Thus, in the UK, the Lake District Boundary Fault, was, during the Permian and Early Triassic, a controlling fault for the subsidence of the East Irish Sea Basin (Akhurst *et al.,* 1998). Today, it may be subject to reactivation, given the occurrence of some small

earthquakes within its traces (Musson, 1987; 1998), but it does not seem to occupy a role more significant than any other fault in the North of England, where many earthquake epicentres may be found.

The assessment of faulting and seismicity has importance in two respects to hazard for nuclear waste disposal, depending on whether one is concerned with rupture (displacement) hazard or vibratory hazard. Both issues need to be addressed; they also share some common problems. Fault displacement hazard refers to the danger of physical movement along a fault plane disrupting the waste emplacement; one can distinguish between principal fault hazard (movement along the fault plane of the earthquake) and distributed fault hazard (secondary movement at some distance from the principal fault plane) (Stepp *et al.*, 2001). Vibratory hazard concerns the possibility of damage due to strong shaking. Secondary hazard due to seismic disruption of ground water patterns is also possible. These topics are covered in a general way in this discussion, which is relevant to a range of geological media. Considerations particularly relevant to argillaceous media are noted where they occur.

The first defence against rupture hazard is to avoid sites that are intersected by potentially dangerous faults, and it is here that categorisation of faults becomes important. Sufficient setback of the waste emplacement from fault planes avoids the obvious problem of principal fault hazard. In the case of faults that are not controlling faults in low seismicity areas, the likelihood of fault rupture is probably rather low: in the UK, for instance, the larger earthquakes typically have depths between 9 and 15 km and rupture dimensions of a few km at the very most; thus the extension of the rupture plane to near-surface is not likely, and has never occurred in historical times.

The question of whether the geological medium (and in particular, argillaceous media) influences the fault displacement hazard, has so far been little studied. It is undoubtedly the case that the nucleation of earthquakes is unlikely within clay or other surficial deposits; according to Marone and Scholz (1988), it is often possible to define an upper stability layer where, apparently, the velocity strengthening character of the upper sediments limits seismogenesis. Counter-examples where a very shallow hypocentral profile is observed tend to be from hard rock intraplate areas without clear faulting, such as the Adirondacks. Even in the United Kingdom, depth profiles show few hypocentres in the upper 5 km, other than small events.

Nevertheless, large enough earthquakes can start in the crystalline basement and rupture through to the surface. The question now has to be posed, whether argillaceous sediments may inhibit this. Evidence that it may do comes from the 1979 Imperial Valley CA earthquake. Quin (1990) showed that a thick upper sedimentary layer, through velocity strengthening, resulted in a zone of negative stress drop above the fault rupture, where rupture propagation was inhibited. Such a process cannot altogether prevent deformation, and a “catching-up” followed the earthquake in which displacement of the sediments occurred aseismically, a process known as afterslip (Crook, 1984).

However, this is a case from a high-seismicity interplate area, and, given the generally higher stress drops from intraplate areas, the effect may be reduced for large earthquakes in lower seismicity areas. Heavy clay deposits in the Mississippi Valley did not prevent rupture propagation either to, or very close to, the surface in the case of the New Madrid earthquakes (Guccione and Marple, 2002). Other examples of faulting to the surface in an intraplate, clay-rich environment could certainly be adduced from the literature.

Assessing the probability of an earthquake occurring of sufficient size to rupture the full width of the crust leads to consideration of the issue of maximum magnitude (Mmax), a fraught topic. For the United Kingdom, one suggestion is that the maximum possible event may be as low as 5.5 Mw (Ambraseys and Jackson, 1985). If this is true, the implications for the possibility of rupture hazard are significant, and are made explicitly by Ambraseys and Jackson (1985). However, there is no consensus as to how Mmax should be evaluated. Statistical methods (e.g. Kijko and Graham, 1998) tend to produce evaluations not much higher than the historically observed value; arguments from paleoseismicity and "comparable" locations elsewhere in the world can produce higher values but are generally open to debate. There are also disputes as to the magnitude of some key observed historical earthquakes elsewhere, such as the 1356 Basel earthquake.

Turning to the disruption of ground water, it is well known that when earthquake intensity reaches around 7, effects on ground water, such as changes in the flow of springs, become observable (Vogt *et al.*, 1994). Many instances can be advanced even in the relatively low seismicity environment of the United Kingdom, for example the 1884 Colchester earthquake (4.6 ML) (Musson *et al.*, 1990). Fluctuations in the water table can either be short term (due to dynamic response to the passage of earthquake waves) or long term, which may be due to change in pore fluid pressure or an increase in permeability. Expected rises in the water table due to a regional drop in shear stress following an earthquake are of the order of some tens of metres, and are sensitive to the amount of stress change, the compressibility of the whole rock compared to the compressibility of its constituent minerals, and the moisture content above the water table (Cook and Kemeny, 1991). Clay media are likely to have a low ratio of mineral to rock compressibility (K/K_s) and this increases the expected rise in water table.

The issue of shaking hazard is more familiar to seismologists, but its relevance to deep disposal repositories is less clear. Smistad *et al.,* (2001) consider that direct damage from earthquake shaking to the waste packages or engineered barriers of an underground repository is an insignificant chance, but damage to cladding is a possibility and should be evaluated.

Apart from engineering issues, there are two important seismological differences between hazard assessment for a waste repository and for a normal NPP site. The first is the simple fact that a repository is buried at a depth of a few hundred metres. The second is its long operational life.

The depth of burial is significant because strong ground motion decreases with depth. Below 70 m, peak acceleration has been shown to be one-third to one-fifth of its equivalent value at the surface (Sykora and Bastani, 1998). The reduction is greatest for stiff profiles.

The long period of time causes some considerable uncertainties for any seismic hazard assessment. Even when looking at long return period events for normal hazard studies, seismologists are still only concerned with the next 50 years or so of actual time, and can make the assumption that the gross properties of the regional seismicity over that period will be the same as for the last 50 years.

When looking at long periods into the future, this assumption no longer applies, for three possible reasons. Firstly, tectonic processes may change. Just as in the geological past the configuration of plates was different from what it is now, so in the future it will be different again. Many of these developments are more or less predictable from knowledge of current plate velocities; however, significant changes are most likely to take several million years to develop, so this is not really an issue.

Secondly, some tectonic processes have irregularities related to plate geometry. For example, in cases of continental collision, if the impacting plate has a pointed shape, it is likely that two seismic zones will develop on each flank. For mechanical reasons, the brunt of the collision will be taken up by the two flanks alternately rather than both simultaneously. This can result in seismic zones appearing to switch "on" and "off" over periods of several hundred years, and this can be seen with the Arabian Plate and in Iran. If one is dealing with a site that has been "off" over recorded history, it may be difficult to estimate what will happen when it switches "on" again.

Thirdly, climate change can be expected to play a significant role in changing seismicity patterns, at least in the northern hemisphere, a topic that has been the subject of considerable recent research. The possibility of renewed glaciation within the lifetime of a repository has to be considered. There is spectacular evidence from Scandinavia and less clear evidence from Scotland for increased seismicity due to rapid isostatic rebound immediately after deglaciation (Muir Wood, 1989, Firth and Stewart, 2000). Current understanding of the influence of glaciation on seismicity is summed up by Stewart *et al.* (2000). It is expected that the onset of glaciation is liable to suppress earthquake activity, at least for a while, followed by much higher levels of activity after the ice has retreated again, leading in the extreme case to the event (or possibly events) that produced the 150 km long, 13 m high fault scarp of the Pårve Fault in Sweden (Lagerbäck, 1979).

Consequently, estimates of seismicity based on current rates may be quite misleading as to the possible levels of activity that could occur in the more distant future. Modern earthquakes do not much exceed magnitude 4 in those parts of Scandinavia that have evidently seen events in the range 7 to 8 in immediate postglacial times.

Nor is this exclusively a high-latitude problem. Muir Wood (2000) suggests that the large historical earthquakes of the New Madrid sequence (1811-1812) and Charleston (1886) in North America may have been due to an outward moving wave of forebulge collapse from the Laurentian ice sheet, and that a future occurrence of large earthquakes in the presently quiet regions of Georgia, northern Louisiana and Arkansas is perhaps to be expected.

Although it is generally considered that probabilistic methods (PSHA) give a better assessment of seismic hazard for nuclear facilities (Stepp, 2001), some writers are at pains to point out that probabilism and determinism in seismic hazard should not be viewed as competing camps (McGuire, 1999, Bommer, 2002). In particular, McGuire (1999) suggests the principle that, when the worst possible case is reasonably likely to occur, one may as well face up to it and model it deterministically. In other cases, probabilistic methods are a better strategy to combat the problem of over-conservatism.

This principle can be extended by noting that the worst case is unlikely to be observed in cases that are low hazard-short lifetime, but likely to be observed in cases that are high hazard-short lifetime *and* cases that are low hazard-long lifetime.

Also, in cases where conventional PSHA methods are extended to very low occurrence probabilities (such as 10^{-7}), there is a strong danger of obtaining results that may be physically unrealistic, if no account is taken of the physical limits to earthquake ground motion, a subject seldom tackled, and on which there is currently no consensus. Also, even if peak ground accelerations of many g are credible at all, one can reasonably ask what engineering significance they have, given the persistently poor correlation in experience between very high pga values and actual damage to structures.

Whatever procedure one adopts, when working with seismic hazard over long periods of time, inevitably one finds that the critical issues are the limits to the tails of the distributions of parameters – the largest magnitude that can occur; the largest ground motion that can occur for a given magnitude; and the greatest distributed displacement that can occur. Unfortunately, these limits are liable to be poorly constrained; it is the nature of very rare events that we are likely to have little experience of them, even in high seismicity areas. Consequently one must try to walk a fine line between adequate conservatism, and maintaining a sense of physical realism.

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RESEARCH OF FAULT ACTIVITY IN JAPAN

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1. Background

Six hundreds and eighty earthquakes causing significant damage have been recorded since the 7th century in Japan. It is important to recognise faults that will or are expected to be active in future in order to help reduce earthquake damage, estimate earthquake damage insurance and siting of nuclear facilities. Such faults are called "active faults" in Japan, the definition of which is a fault that has moved intermittently for at least several hundred thousand years and is expected to continue to do so in future. Scientific research of active faults has been ongoing since the 1930's. Many results indicated that major earthquakes and fault movements in shallow crustal regions in Japan occurred repeatedly at existing active fault zones during the past. After the 1995 Southern Hyogo Prefecture Earthquake, 98 active fault zones were selected for fundamental survey, with the purpose of efficiently conducting an active fault survey in "Plans for Fundamental Seismic Survey and Observation" by the headquarters for earthquake research promotion, which was attached to the Prime Minister's office of Japan. Forthy two administrative divisions for earthquake disaster prevention have investigated the distribution and history of fault activity of 80 active fault zones. Although earthquake prediction is difficult, the behaviour of major active faults in Japan is being recognised.

Japan Nuclear Cycle Development Institute (JNC) submitted a report titled "H12: Project to Establish the Scientific and Technical Basis for HLW Disposal in Japan" to the Atomic Energy Commission (AEC) of Japan for official review [1]. The Guidelines, which were defined by AEC, require the H12 Project to confirm the basic technical feasibility of safe HLW disposal in Japan. In this report the important issues relating to fault activity were described that are to understand the characteristics of current fault movements and the spatial extent and magnitude of the effects caused by these movements, and to estimate the spatial extent of future fault movements and their effects on the geological environment. One conclusion from the report is that present active faults in Japan have moved repeatedly for at least the last several hundred thousand years and are likely to continue to do so under the same stress field in the next a hundred thousand years. The latest knowledge relating to active faults and features of active faults to be considered for the stability of geological environments are described.

2. Characteristics of fault activity in Japan

Tectonics of the Japan islands and faults activity

The Japanese islands and the adjacent sea areas are located near the margins of the Pacific Ocean, Philippine Sea, Eurasian and North American plates (Figure 1). The Pacific Ocean plate and the Philippine Sea plate subduct beneath the continental plates on the west side. The current general plate movements were established by the end of the Miocene to the end of the Pliocene [2]. Consequently, when estimating the spatial distribution of natural phenomena in Japan for the next hundred thousand years, it is reasonable to consider that the patterns and trends in tectonic activity over the past several hundred thousand years will continue. It is also justifiable to assume that geological phenomena as associated with the tectonic setting of Japan will progress with predictable trends for at least the next hundred thousand years. Figure 2 shows the result of calculating the deformation rate and its orientation in the Japanese archipelago based on the measurement value obtained by Global Positioning System (GPS). It has been shown that strong compression is now occurring in an east-west direction underneath the Japanese archipelago. A similar conclusion has been obtained from an analysis of seismic observation data. The results of various topographical and geological surveys form the basis for the hypothesis that a similar compression force has been exerted underground for quite some time, at least for several hundred thousand years [3].

Figure 1. Plates around Japan

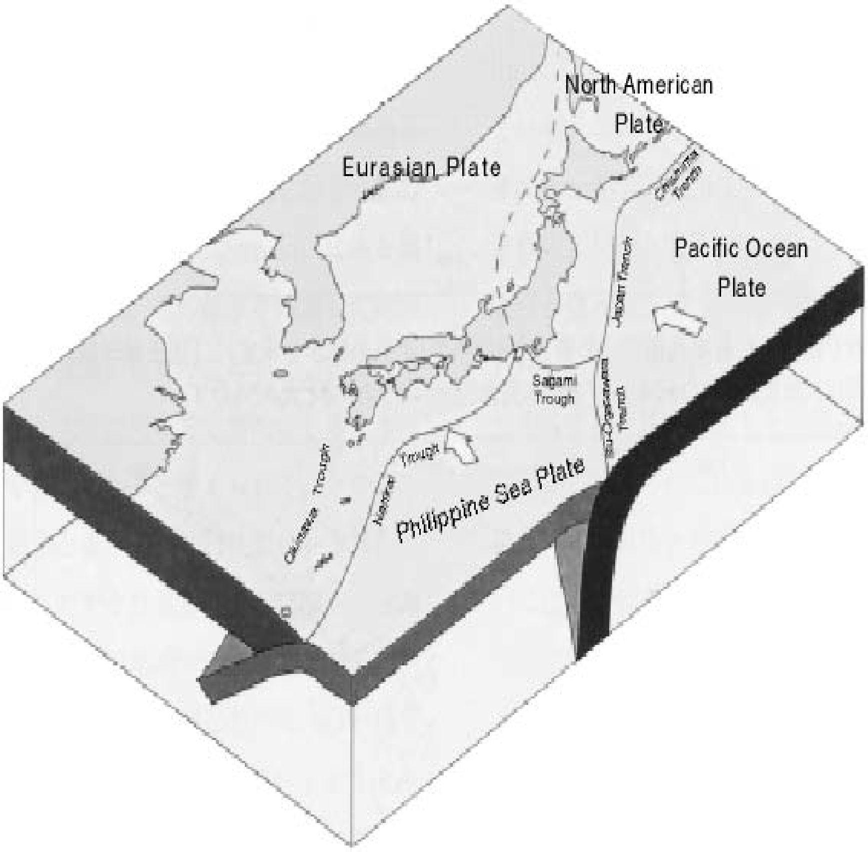


Figure 2. Crustal deformation

The history of fault activity, such as displacement, latest active time and recurrence interval, has been estimated from investigation of the development and frequency of occurrence of tectonic landforms and key beds (e.g. tephra), often by observation of outcrop. We compiled the results of relevant previous investigations. In Japan, most active faults are located in a compressive stress field caused mainly by the relative movements of the tectonic plates. For example, northeast Japan has been located within an east-west compressive stress field for several million years and many reverse faults have developed.

The situation in southwest Japan is slightly more complex [4], but the current fault movement was maintained across much of the area by 0.5 million years ago. No significant changes of orientations and of the rates of displacement have been observed in major active faults in the past several hundred thousand years. Thus, present major active faults have been active for at least several hundred thousand years and are likely to remain active.

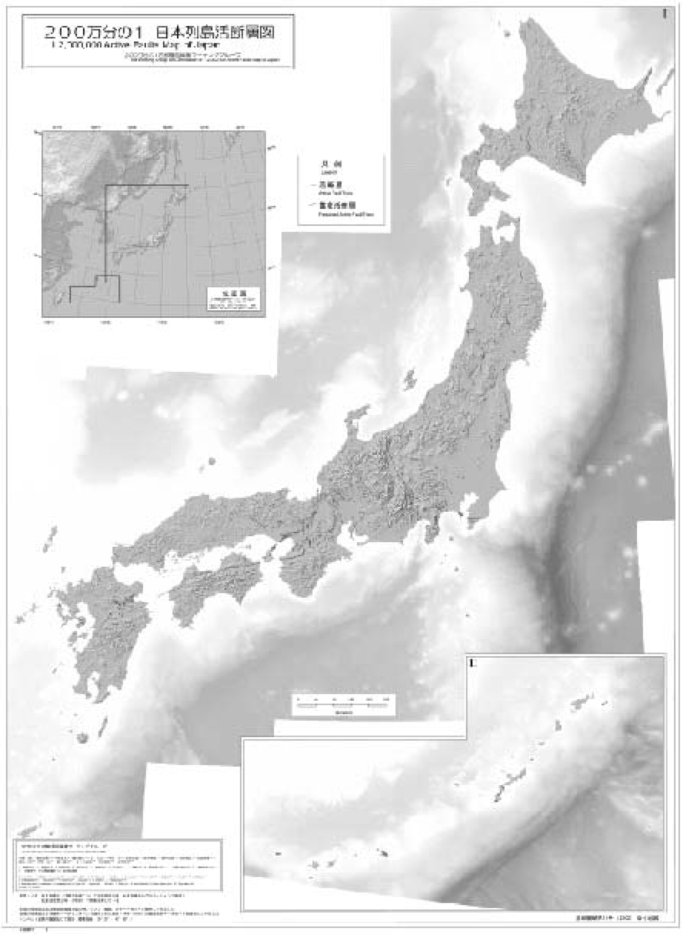
Distribution and types of active faults in new active faults map

We compiled new active faults map of Japan (Figure 3) based on the digitised Geographic Information System (GIS) data of active faults with a scale of 1:25 000 [5]. Active faults were defined in the new map as faults that have repeatedly moved during the late Quaternary with intervals of one thousand to ten thousand years forming distinctive fault-related tectonic features on the earth's surface. Previous studies [6] commonly defined active faults as faults that repeatedly moved during the Quaternary and are the potential source of future earthquakes. Broadly speaking, the localisation of active faults that have prevailed for the last several hundred thousand years can be seen throughout the Quaternary period. The polarisation of the distribution of active faults and the localisation of their degree of activity and types are also described in previous studies. Active faults clearly show localism of distribution densities and types, which reflect differences in geological structures and physical properties of the upper crust. Reverse faults with a north- south strike are concentrated in northeast Honshu and in the Kinki area. In contrast, left-lateral faults with a northeast-southwest strike are concentrated in the west of central Japan. The Izu Peninsula and its northern adjacent district differ from the other districts in trends and types of active faults.

Extent of the range of active fault movement and active fault zone

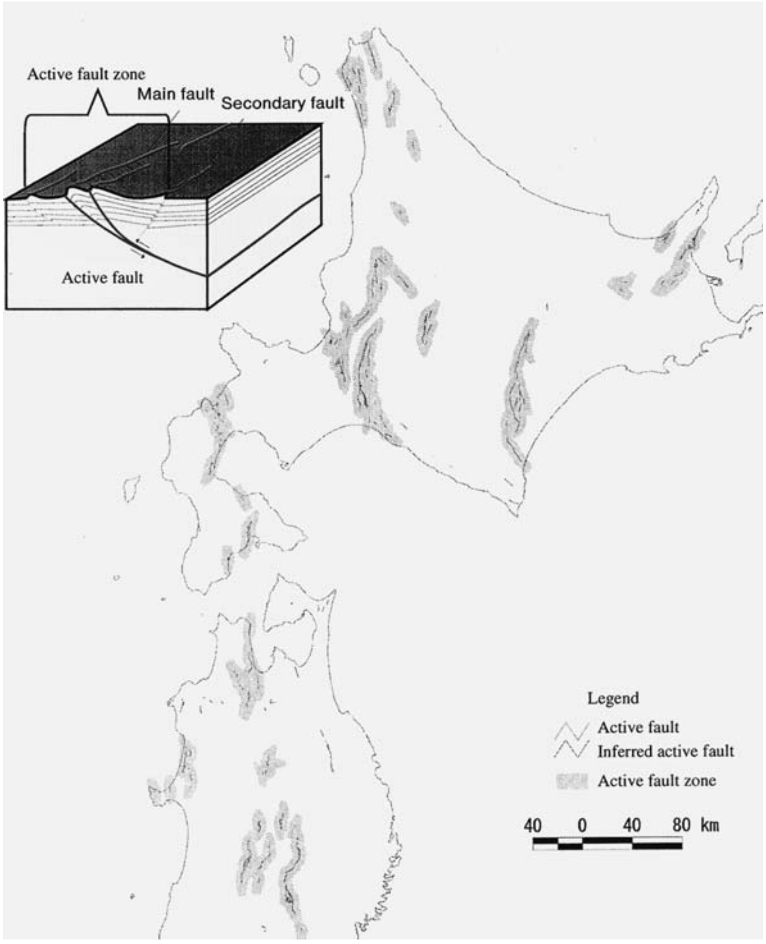
An evaluation was made of whether or not a reverse fault zone, which consists of several reverse faults running nearly parallel to one another, has extended. Geophysical exploration was conducted across a reverse fault zone at the eastern edge of the Yokote basin. This study showed that, during the past several hundred thousand years, the fault zone has propagated over a range of about 2 km near the surface. All findings indicate that possible future propagation of any reverse fault zone is unlikely to exceed 8 km and occurs in the direction of the basin.

Figure 3. New active fault map of Japan



Many large earthquakes in the past were often caused by the movement of several active faults that distribute in the vicinity of each other. Matsuda [7] grouped a series of faults, designating them as "seismogenic faults" with the potential to independently cause earthquakes. Such a grouping of faults is important for estimating the behaviour of individual active faults. On the basis of the latest map of active faults [5], the division of active fault zones exemplifies the fields of future faulting (Figure 4). A fault zone is defined by the envelope surrounding individual active faults that have: i) similar orientations; ii) separations parallel to the fault planes up to 5 km; or iii) separations perpendicular to the fault planes of up to 10 km. By comparing the overall properties of such a fault zone with the properties of individual active faults within the zone, it should be possible to infer general properties of faults within the zone that have not been identified explicitly. The identification of active fault zones could lead to a broad understanding of the extent of their expansion in future. There have been no cases of new active fault zones appearing at least during the last several hundred thousand years. The geological information mentioned above indicates that, in an area where significant changes in the crustal stress field are unlikely, there is little possibility that new fault zones might be generated independently of movements along existing active fault zones.

Figure 4. Example of active fault zone



3. Effects of fault movement

The mechanical influence of fault activity on the geological environment is limited mainly to fault crushed zones. Otsuki [8] points out that field and laboratory data on faults, both in Japan and abroad, indicate a relationship between the width of a crushed zone (mainly fault gouge and fault breccia) and the cumulative displacement. This relationship can be used to estimate the widths of fault-crushed zones from the scale (length and cumulative displacement) of the associated faults. Based on this relationship, the crushed zone of a fault with a cumulative displacement of 10 km would be roughly 200 m in width. The extent of this influence is several meters to several hundred meters at most. At the time of the 1930 Kita-Izu Earthquake, a displacement of about 2 m was observed in the fault-crushed zone in the Tanna Fault (length: 30 km). Deformation was observed to have occurred in an area extending for about 1.5 km to the west of the fault crushed zone [9]. The area where fractures are distributed in a rock mass formed by fault movement, is reported to be in the order of 10^{-2} relative to the length of the fault [10]. The range over which associated small faults are distributed in the surrounding rock mass is considered to be within a few kilometres of large active faults.

The location and scale of potentially perturbing faulting have been traced back over the past several hundred thousand years. Based on the results of these studies, it can be shown that it is possible to select a sufficiently stable geological environment in Japan, such that a repository will not be influenced by disruptive faulting for at least the next hundred thousands years. A detailed investigation of the closest active faults to any future site proposed for HLW disposal should be performed. This investigation should allow an appropriate distance between the disposal facilities and active faults to be defined. This is necessary because the size of regions affected by fault activity depend on the sizes of the active faults. In addition, an in-depth investigation should be performed at the site to identify whether or not there are small active faults that were not recognised during regional surveys. This approach will avoid the influence of such faults during detailed siting. The effects of earthquakes also should be considered during the design of the geological disposal system. However, the influence of seismic vibrations several hundred meters underground or deeper is generally regarded to be small.

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GEOLOGICAL EVOLUTION OF CLAY SEDIMENTS: THE PETROLEUM EXPLORATION VISION

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Introduction

The radioactive waste isolation capacity assessment for a clay sediment host rock is link: (1) to the understanding of their present state properties and 3-D repartition (from basin evolution, including sedimentary and diagenetic process); and (2) to the prediction of their future evolution during the next million years.

For petroleum exploration, basin modelling aims at reconstructing the accumulation of hydrocarbons at basin scale, and at geological timescale, taking into account the effects of kinematics displacements, sedimentation, erosion, compaction, temperatures history, overpressures and fluids flows (water and hydrocarbons). Furthermore, explorationists wish to address overpressure reconstruction in order to estimate the risks of drilling.

Clay sediments are of interest for petroleum exploration because source rocks and seal are generally composed of them. Nevertheless, in spite of their occurrence in nature their evolution at geological timescale is not well understood. And, most of the knowledge has been achieved by those working in the realms of soils mechanics and civil engineering until the present geological investigations for long term radioactive waste repositories. Application of this knowledge to clay sediment is considered to be valid within the first hundreds of meters at the top of the sedimentary pile, according to a repository depth.

This paper is dedicated to the sedimentary rocks behaviour at geological timescale. This behaviour is characterised by: (1) the deposition of the sediment; (2) the loading path at geological timescale; (3) the constitutive law which includes the consolidation process and the rupture criteria; and (4) the parameters evolution related to consolidation.

Deposition

Clay sediments are generally deposited in low energy environments. Geometry, composition and texture of the bodies are also controlled by the depositional environment. More generally, sediments architecture is the response to complex interaction between three processes: (1) the creation of available space of accommodation in the basin; (2) the supply of sediment brought into or produced in the basin; and (3) transport of sediment within this basin (e.g. Grangeon, 2002). These processes can now be simulated with dynamic-slope forward computer model such as Dionisos (Granjeon & Joseph, 1999). This kind of model is able to simulate the stratigraphy in 3-D on a prospect and on a basin-wide scale. And, if they are often used to predict the distribution of the reservoir bodies, their results can be interpreted in term of distribution of muds. Furthermore, the intercalation of turbidites or other geological bodies such as sedimentary sills or dykes (e.g. Parize *et al.*, in prep.) can modify drastically the permeability tensor: in some case the macroscopic permeability along the stratification may be up to five order of magnitude greater than the permeability across the stratification.

Loading path

One key point for simulating the behaviour of sediments at basin scale is to be able to compute the stress tensor. This is generally not addressed in basin model because of the complexity of this problem. (Luo *et al.*, 1998). In order to estimate the stress tensor all along the geological history, it is generally assumed that the total overburden weight (σ_v) is one of the main stresses. The second assumption is that the two other main stresses are horizontal and equal ($\sigma_H=\sigma_h$). Furthermore, we assume that the total horizontal stress can be deduced from the total vertical stress by the following formula: $\sigma_h=Ko(z).\sigma_v$. According to the tectonical environment, Ko may varies from values lower than 1 in extensional area to values greater than 1 in compressive area (e.g. Grauls, 1996).

Constitutive laws

From the top to the bottom of the sedimentary column we distinguish four major horizons. The first horizon at the top of the sedimentary column is made up of mud. Beneath this horizon we consider that the sediment forms a soil having a thickness of some meters. The appearance of cemented grains marks the start of the third horizon. It is made of the sedimentary rock *sensu stricto*. The beginning of the metamorphism marks the end of this third horizon and the beginning of the fourth one.

The previous geometrical description of the sedimentary column could be considered as an historical evolution of sediments that are mud when they are young, then they become soils, sedimentary rocks and metamorphic rocks. Determining all the mechanisms that cause this evolution would appear to be impossible. However, attempting to determine the major physical phenomena in the history of the sediment is feasible. Hedberg (1936) has already tackled this operation. Here we will only consider the sedimentary rock *sensu stricto*, and will use the word consolidation to represent all the physical processes that act on the sediment during its geological history.

Figure 1. Porosity effective stress curves for natural and lab-compacted

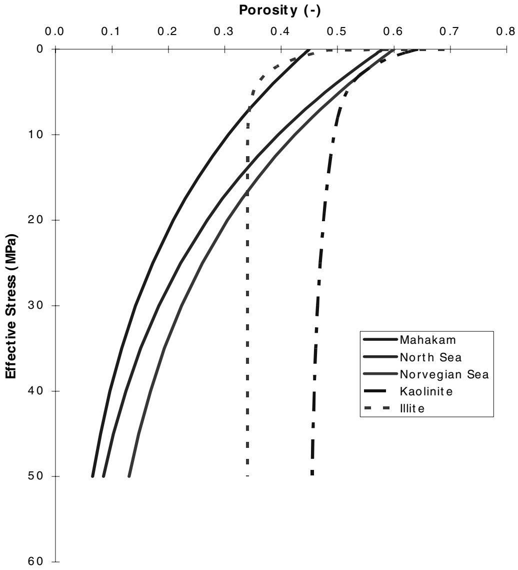
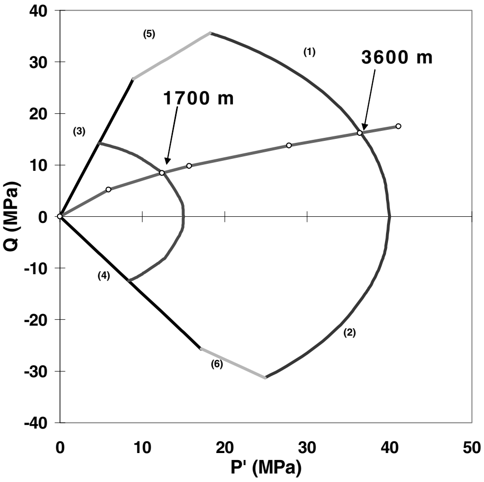


Figure 2. Example of normal hydrostatic stress path ans corresponding elastoplastic yield



Consolidation is considered here as the combined effect of the mechanical compaction and the chemical compaction (Schneider *et al.*, 1996). The mechanical compaction is mainly caused by the rearrangement of grains during burial. This mechanism is efficient near the surface. The mechanical compaction could be represented at the macroscopical scale by an elasto-plastic rheology. The chemical compaction is considered here as resulting from dissolution-precipitation mechanisms, generally induced by stress (pressure-solution). This mechanism generally becomes to be active at a depth varying from few hundred to few thousands meters. The chemical compaction could be represented at the macroscopical scale by a visco-plastic rheology. The resulting elasto-visco-plastic rheology could be written as follows:

$$\frac{d\phi}{dt} = -\beta(P')\frac{dP'}{dt} - \alpha(\phi, T)P'$$

The visco-plastic rheology has been introduced to account for pressure solution in limestones and sandstones. Nevertheless it is also used for shales because a viscous component exists as shown in Figure 1 where porosity lower than 30% cannot be reached by mechanical compaction only.

At a given time, the consolidation elastoplastic yield is supposed to be given in the (Q, P') space by the classical following equation:

$$Q^2 + P'^2 - P'^2_c = 0$$

where P'_c is the consolidation mean effective stress, which evolves with the geological history of the sediment. P'_c is not only a function of the maximum mean effective stress reached by the sediment, as assumed in soils mechanics, but also a function of time and temperature due to chemical reactions. This is what we call the chemical hardening of the sediments.

The complete elastoplastic yield (Figure 2) is defined by the union of the consolidation elastoplastic yield and of the different failure criteria that could be seen as elastobrittle yields (Schneider *et al.*, 1999). Thus, the elastoplastic yield is composed of six elementary elastoplastic yields which define the onset of: (1) vertical compaction; (2) horizontal compaction; (3) vertical tensile fracturing; (4) horizontal tensile fracturing; (5) subvertical shear fracturing; (6) subhorizontal shear fracturing.

Parameters evolution with consolidation

Due to the consolidation, most of the parameters that describe the physical properties of the sediments evolve with the geological times. One difficulty is to quantify the degree of evolution of the porous medium during its geological history. We have chosen to measure the evolution of the sediments by their porosity because this is one of the physical properties that can be measured easily. This approach is only valid when there is no strong diagenesis with creation of high secondary porosity.

Mechanical properties

The elasto-plastic coefficient is given by (Schneider *et al.*, 1996):

$$\beta(P') = \frac{\phi_a}{E_a} e^{-P'/E_a} + \frac{\phi_b}{E_b} e^{-P'/E_b}$$

where ϕ_a, E_a, ϕ_b, E_b are coefficients that characterise the sediment. These coefficients may be calibrated by using observed porosity-depth data from sediment that have not suffer diagenesis or using experimental laboratory measurements (Schneider *et al.*, 1996).

The visco-plastic coefficient is given by (Schneider *et al.*, 1996):

$$\alpha(\phi,T) = (1-\phi) / \mu_s^o e^{\frac{E}{R}\left(\frac{1}{T} - \frac{1}{To}\right)}$$

where μ_s^o is the macroscopical sediment viscosity at T^o, and E is the activation energy. These coefficients may be calibrated from set of observed data (Schneider *et al.*, 1996; Schneider & Hay, 2001).

The Young's modulus and The Poisson's coefficient could be expressed as a function of the porosity. For example, for chalk, Engstrom (1992) proposed the following relations:

$$E = 224.8\, e^{-11.2\,\phi} \quad (\mathrm{GPa}) \text{ and } \nu = 0.117 + 0.21\,\phi$$

In the plastic domain we suppose that the effective stress coefficient is equal to 1. In the elastic domain, the Biot coefficient could be expressed as a function of the porosity (Schneider *et al.*, 1993):

$$b(\phi) = \frac{\phi}{\phi + a(\phi^0 - \phi)}$$

The tensile strength (T) of sediment is generally weak (e.g. Lama & Vutukuri, 1978). If we assume that the sediment is fractured, the tensile strength could decrease to 0. Indeed, the criterion for the opening of existing fractures is the same we wrote for a tensile fracture but with T = 0.

The cohesion and the friction angle of the sediment are generally known to increase with depth or to increase as the porosity decreases. They could be expressed as functions of the porosity and mineral content. For example, for chalk we can use the empirical relations (Rhett *et al.*, 1992):

$$c = 20.9 - 41.1\,\phi \quad (\mathrm{MPa}) \text{ and } \varphi = 56.1 - 105.6\,\phi \quad (°)$$

Once the expression of the coefficients as functions of the porosity are given, the evolution of the different elastoplastic yield surfaces can be drawn in the (Q, P') space. This has been done in Figure 2 in which the yield surfaces have been drawn for chalk at 1.7 km and 3.6 km. In this figure, it should be noticed that at 1.7 km depth, the elastic domain is defined by the tensile failure criteria and the consolidation yield surface while the elastic domain needs all the rupture criteria to be defined at 3.6 km.

Hydraulic properties

Sediment permeability is given as the sum of two terms. The first one which only depends on the porosity represents the matrix porosity. The second one which is both a function of the pore pressure and the porosity represents the fracture permeability. The matrix permeability is given by an empirical formula, which is an extension of the Koseny Carman formula: $K_m(\phi) = 0.2\phi^n / S^2(1-\phi)^2$. The general permeability model, which includes the fracture permeability is given, when Ko is lower than one, by the following empirical formula:

$$\begin{array}{ll} p \le \sigma_h & K_f(\phi, p) = K_m(\phi) \\ p > \sigma_h & K_f(\phi, p) = K_m(\phi)\left(1 + \alpha_f\left(\dfrac{p}{\sigma_v} - Ko\right)^{af}\right) \end{array}$$

Conclusions

Prediction of the behaviour of clay sediments in the future needs to reach a good knowledge of their present state. On one hand, the geometry, the texture and the composition of clays sediments bodies are function of the geological environment during deposition. On the other hand, the present day physical properties of sediments are the result of a geological history characterised by a loading history (P, σ, T) and related mechanical and chemical modifications.

One of the properties of the sediment is its elastoplastic yield, which evolution is characterised by a long term chemical hardening. The relative position of the stress state at present day which respect to the elastoplastic yield is crucial for any forecasting. For example, the impact of a pore pressure modification will be different in a basin where sedimentation is active (the stress tensor is initially on the elastoplastic yield) and in a basin where erosion is active (the stress tensor is initially in the elastic domain).

If a sediment can be characterised through lab experiments, large timescale processes can only be addressed by basin modelling. Nevertheless, basin simulators needs physical models that are able to reproduce the evolution of clays sediments through geological times. These models are built accordingly to our understanding of the diagenetic evolution of shally formations.

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PREDICTING THE DIAGENETIC EVOLUTION OF ARGILLITE REPOSITORIES BASED ON THE STUDY OF NATURAL AND EXPERIMENTAL SYSTEMS

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1. Introduction

Understanding the nature of diagenetic reactions involving clays and associated mineral phases is an important aspect in predicting the long-term chemical and physical properties of host-rock argillite in underground radioactive repositories. Much of our current knowledge is based on the study of mineral phases in natural rock sequences that have formed under a range of pressure (P), temperature (T), compositional (X) and time (t) conditions (Velde, 1992). Valuable information has also been obtained by reproducing reactions in the laboratory under constrained conditions using batch or flow-through reactors (e.g. Nagy, 1995; Wogelius & Vaughan, 2000). Based on our current knowledge, six primary controls can be recognised, which drive diagenetic reactions in the upper portion of the Earth's crust.

i) Fluid chemistry, providing the essential ingredients for exchange of cations and anions.

ii) Rock composition, laying down the basic building blocks and reaction sites.

iii) Rock permeability, necessary for allowing material transport and fluid-rock interaction.

iv) Temperature, as the principle energy source for enhancing reaction rates.

v) Time, needed to allow reactions to get closer to their equilibrium conditions.

vi) Stress and strain, for mechanical mixing of reactants and products.

In order to predict the diagenetic evolution of argillite repositories, we need to know firstly, which reactions occur under a defined set of conditions, secondly, how these reactions modify the material properties of the argillite seal, and finally, how fast these chemical reactions take place. Based on the application of thermodynamics, and the construction of activity diagrams for low temperature mineral phases (e.g. Velde, 1992), fair predictions of mineral stability can be made under a given set of physical and chemical conditions. Such predictions are strengthened by examining mineral reactions that occur under natural conditions and also those that have been preserved in the geological record. Changes in the material behavior can also be reasonably assessed, as the basic physical and chemical properties of argillaceous rocks of varying mineralogy are well documented in the petrophysical and engineering literature (e.g. Bell, 1999).

One of the most difficult tasks in predicting the response of argillites to changing physical and chemical conditions is to assess the rates of the chemical reactions involved. This difficulty reflects our poor knowledge of the reaction kinetics for these low-temperature, fine-grained mineral materials. The aim of this contribution is therefore to review the timing and duration of diagenetic reactions that occur in nature and to compare them with reaction rates measured by laboratory experimentation. Some new analytical techniques are also presented, which should serve as a better analogue for investigating and predicting the outcome of radioactive repository sites under the influence of varying geological scenarios.

2. Diagenesis in the geological record

The geological record is unique in that it provides a natural laboratory that has been running since the formation of the Earth. The rock record extends back to ca. 3.9 Ga, and there are case studies of ca. 2 Ga old illitisation of argillites in contact with radioactive bodies (Bros *et al.,* 1992). Traditionally, clay mineral diagenesis of argillites has been considered to be largely a function of temperature and time (Weaver, 1960). Generally, older sedimentary sequences are often more mature or metamorphosed, with progressive mineral alteration and recrystallisation occurring in response to successive burial within the Earth's crust (Clauer & Chaudhuri, 1995). Currently, rock alteration and diagenetic reactions are more favorably viewed as occurring during discrete geological events, in response to distinct changes in physico-chemical conditions. Even the classic Gulf coast diagenetic sequence of Hower et al (1976) has been reinterpreted as resulting from number of punctuated events (Eberl, 1993). The timing of such events can be dated isotopically. For example, Rousset & Clauer (2003) determined the age of diagenesis in the Callovo-Oxfordian argillites within the Paris Basin to occur 10-15 Ma after deposition of the sediments, related to a decline in sea-level. In the case of mixed illite assemblages, the isotopic ages of the various growth events may be modeled, as by Srodon *et al.,* (2002).

The rates and duration of diagenesis are much more difficult to assess from the geological record. Whereas the rate of mineral dissolution during surface weathering can be measured (Schnoor, 1990), the kinetics of dissolution and crystal growth reactions at depth in the Earth's crust can only be extrapolated. There are, however, a number of situations where the rates of diagenesis can be approximated from the geological record. The intrusion of magmatic material into argillites provides a natural analogue for underground nuclear waste repositories, where the nature of thermally induced mineral reactions can be characterised. Techer *et al.,* (in press), for example, document the formation of illite-smectite mixed-layered clays, and the disappearance of kaolinite and chlorite, toward the contact of a 2 Ma old, 1metre thick dike. These reactions appear to occur rapidly at temperatures of <260°C and probably within a duration of a few to tens of years. The rapid growth of clay minerals is also evident from studies of seismically active fault zones, such as the San Andreas Fault System of California and the Alpine Fault of New Zealand (Chester *et al.,* 1993; Warr & Cox, 2001). Here, pore-filling low temperature mineral phases, such as clays, zeolites and carbonates, are intimately linked with the earthquake cycle and form during the inter-seismic periods that separate high velocity slip events.

3. Simulating diagenesis in the laboratory

Laboratory experiments have the distinct advantage that P-T-X conditions can be carefully controlled and measured, but the disadvantage that only short reaction times can be studied. Most quantitative studies of reaction kinetics experiments have focused on the study of mineral dissolution rather than crystal growth, and changes in mineral abundance are typically measured indirectly by monitoring changes in solution composition. Reviews of the available experimental data have been compiled by Nagy (1995) and Kloprogge *et al.* (1999).

3.1 Batch reactors

Batch reactors are the most commonly used form of reactant vessel for simulating mineral alteration and diagenesis. These reactors are closed systems, and in order to facilitate rates, a large water to rock ratio is used, along with occasional stirring to mix reactants and products.

Experiments relevant to the diagenesis of argillites under various repository conditions have been conducted. Rapid alteration of smectite has been shown to occur under low temperatures (< 60°C) and alkaline solutions (pH > 13) caused by the alteration of concrete (e.g. Rassineux *et al.,* 2001). Dissolution of smectite and formation of a random illite-smectite phase was observed in similar experiments on Callovo-Oxfordian argillites from the Meuse-Haute Marne laboratory (Claret *et al.,* 2002). However, here the organic coating on particle edges appears to reduce the reactivity of the clay minerals.

Rates for the conversion of smectite to illite at higher temperatures (250 to 350°C) have also been studied experimentally for varying K+ in solution (Huang *et al.*, 1993). Based on these experiments, a kinetic equation was presented, which can be used to predict the extent of reactions in sedimentary brines largely dependent on time, temperature and potassium concentration. The effectiveness of this, and other available kinetic models, has been evaluated by Elliot & Matisoff (1996) for various geological settings.

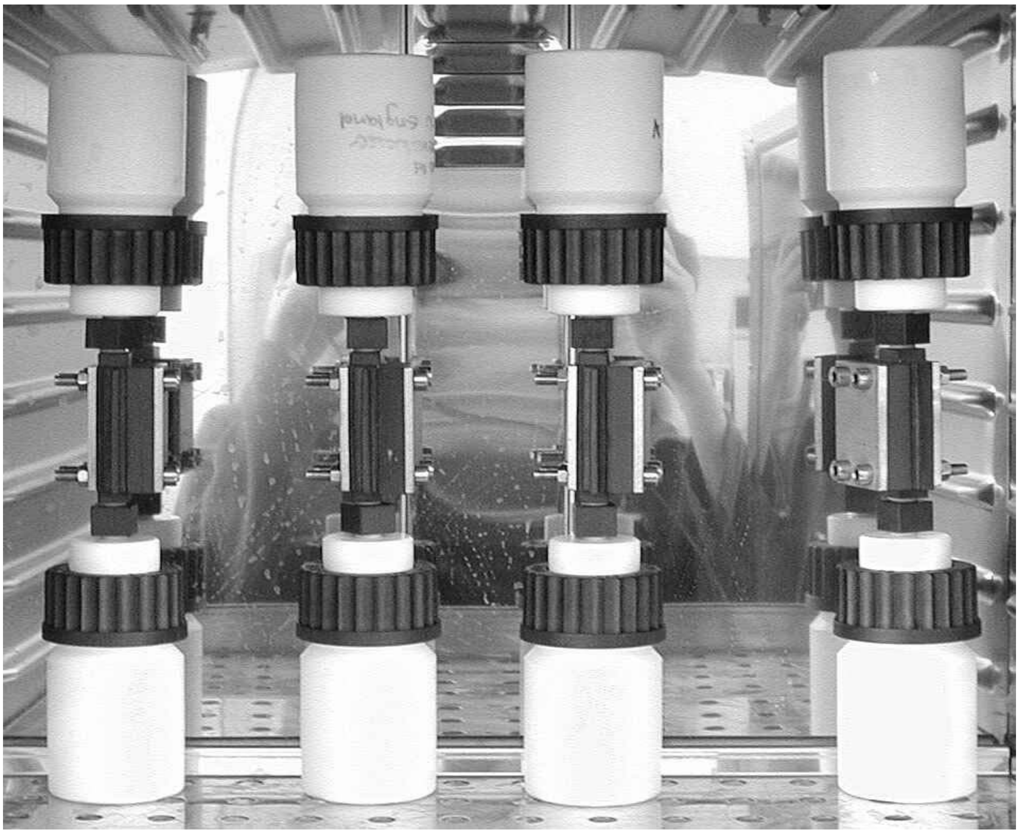
3.2 Flow-through reaction chambers and wet-cell X-ray diffraction techniques

Flow-through reactors are not commonly employed for the synthesis of low temperature mineral phases in the laboratory, probably because of their complexity. They do, however, provide better analogues for the Earth's crust, with a dynamic open system characterised by the circulation of fluids and mass transfer.

A new approach to monitoring the reaction kinetics of fine-grained minerals in solution has been presented using wet-cell X-ray diffractometry (Warr & Hofmann, 2003). This device consists of a flow-through reaction chamber, which can be routinely mounted onto the X-ray diffractometer for in-situ measurements of the sample. With the aid of a pressure cap, the device can be used under hydrothermal conditions (<150°C). Experiments are currently running in order to quantify the rates of diagenetic reactions in argillite host rocks under repository relevant conditions.

The setup in Figure 1 shows cells containing pressed powders from the Callovo-Oxfordian Formation of the Meuse-Haute Marne underground repository site. The reactive solution passes from the upper source bottle, percolates slowly through the cell, and collects in a lower catchment bottle. At 85°C the solution is driven largely by differences in vapour pressure. X-ray diffraction (XRD) data is obtained at various time intervals with minimum disturbance of the sample by measurement through a caption foil for the duration of the experiment. Time-dependent changes in the intensity of XRD reflections are used to monitor changing abundance of reactants and products and to establish kinetic relationships. The composition of the circulating solution can be monitored, and the role of permeability on reaction rates assessed.

Figure 1. Flow-through reactors (wet-cells) been used to investigate the kinetics of diagenetic reactions in Callovo-Oxfordian claystones from the HTM102 core of the Paris Basin
Reaction temperature = 85°C, solution = simple young fluid (Claret *et al.,* 2002)



4. Conclusions

Currently we know a lot more about which diagenetic reactions should occur in argillites under a fixed set of P-T-X conditions, but our knowledge of reaction rates remains limited. Therefore we can predict the type of reactions that occur in response to a particular geological scenario, but quantifying speed and duration is still of a speculative nature. Better assessment of the kinetics of diagenetic reactions is required under repository relevant conditions through the study of experimental and natural analogues.

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SESSION III

ARGUMENTS TO SUPPORT CONFIDENCE IN THE STABILITY OF CLAYS CONSIDERED AS POTENTIAL HOST FORMATIONS

SCREENING METHODOLOGY FOR SITE SELECTION OF A NUCLEAR WASTE REPOSITORY IN SHALE FORMATIONS IN GERMANY

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Introduction

The radioactive waste disposal policy in the Federal Republic of Germany is based on the principle that all types of radioactive waste must be disposed of in deep geological formations. Because of the favourable properties of rock salt and the existence of thick rock salt formations in Germany, so far most of the research in the field of radioactive waste disposal sites was focused on the study of the use of rock salt. In addition, German research organisations have also conducted generic research and development projects in alternative geological formations (Wallner & Bräuer, 2001), but a comprehensive evaluation of their utilisation has been only done for parts of the crystalline rocks in Germany. Research projects on argillaceous rocks started relatively late, so that German experience is mainly connected to German research work with the corresponding European Underground Research Laboratories and the exploration of the former Konrad iron mine as a potential repository site for radioactive waste with negligible heat generation.

The German Federal Government has signed in 2001 an agreement with national utility companies to end electricity generation by nuclear power. This decision affected the entire German radioactive waste isolation strategy and especially the repository projects. The utility companies agreed upon standstill of exploration at the Gorleben site and the Federal Ministry for the Environment tries to establish a new comprehensive procedure for the selection of a repository site, built upon well-founded criteria incorporating public participation. Step 3 of the planning includes the examination of further sites in Germany and the comparison with existing sites and concepts. Under these circumstances, argillaceous rock (clay and shale) formations are now a special area of interest in Germany and the development of a screening methodology was required for the evaluation of shales as host and barrier rocks for nuclear waste repositories.

Screening methodology and present results

Thick buried shales and clays are part of various stratigraphic sections of the different sedimentary basins in Germany. They are especially important as hydrocarbon source rocks and as flow barriers of the underground with special importance for the separation of salty formation water from drinking water, the storage of natural gases, and the existence of hydrocarbon fields. The evaluation of these shale formations started with a mapping program focused to find out about the distribution of shale formations with the following threshold values:

- Minimum of shale thickness of 100 m.
- Burial depth of the shale formation below 1 500 m.

- Upper level of the storage in the shale formation larger than 300 m deep.

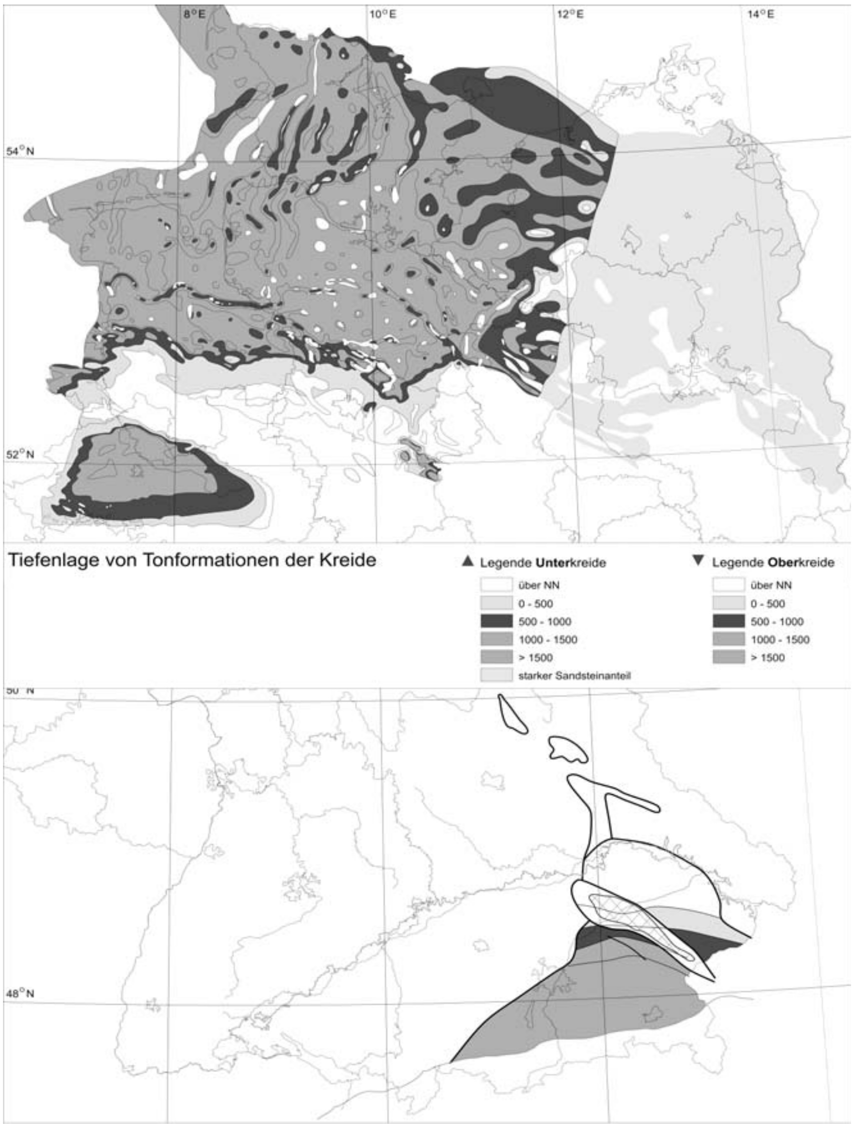
Data for the first investigation stage were primary in the form of descriptions of well files of exploration and production boreholes (mainly oil and gas, but also deep water, geothermal, ore and salt exploration, as well as research boreholes). There are about 30 000 of these boreholes in Germany (Brauner *et. al.,* 2002/2003), but only a very small percentage of those were primarily drilled for the investigation of shales. The second data sources are existing compilations which were produced under various objectives before, like the Geophysical Atlas of Eastern Germany and the Geotectonic Atlas of Northwestern Germany (Geophysik Leipzig, 1989; Kockel *et al.,* 1996/1999; Baldschuhn *et al.,* 2001). Results of the first screening stage show, that there are shale formations with a shale content above 80% meeting the above mentioned threshold values within Tertiary, Lower Cretaceous, and Middle to Lower Jurassic sections in Northern and Southern Germany. The compilation of hydraulic conductivity and transmissistivity values of shales and marlstones from Germany and the neighboring areas done by Appel & Harber (2001) indicate that for depths larger than 300 m these shales are predominantly characterised by hydraulic (k_f) values below 10^{-10} m/s and often even below 10^{-12} m/s. Although German data of well testing from shale intervals of the depth range below 300 m are rare, this observation is in agreement with other worldwide studies. For instance, the work of Bryant (2003) clearly demonstrates the dependency of clay and shale permeability from porosity, void ratio, grain size, mineralogy and pressure or depth of burial. The previous screening value of 80% shale is therefore only an approximate-boundary following the results of Appel & Harber (2001) and Bryant (2003). The latter has documented by laboratory compaction test the severe permeability difference between argillaceous sediments with 80% of clay and rocks with clay contents between 60 and 80%.

Results of screening stage 1, preliminary conclude that only Middle Jurassic and in very restricted areas Upper Cretaceous shales can be considered for future investigation for Southern Germany. In contrast, larger areas with different shale levels require additional investigation in Northern Germany. Figure 1 shows, as an example of screening stage 1, the depth of Cretaceous shales with a thickness above 100 m in Northern and Southern Germany. The combination of thickness and burial depth maps are the base for the demarcation of the areas considered for further investigation during stage 2 of the screening process.

The argillaceous rock formations of the areas passing evaluation stage 1 are in general characterised by low permeabilities, but include a great variety of clays and shales with different other petrophysical, mineralogical, and geochemical properties, as well as large variances in thickness and homogeneity. These differences are mainly caused by the different sedimentary environments during deposition, the variations in burial depth and history, and special tectonic processes. Reviewing different sedimentary environments with respect to primarily shale thickness, shale homogeneity, and shale predictability is one of the first topics of screening stage 2. A first estimation of the diagenetic stage of the shales has been done based on vitrinite reflection measurements, sonic log data evaluation (Dutta, 1986; Magara, 1986; Hansen, 1996), and 1-D modeling of burial history and maximum temperature (Tmax) following the methods of Sweeney & Burnham (1990); Welte & Yalcin (1988); Koch & Schellschmidt (2001). Although within the majority of areas Tmax and maximum burial depth of the shales are equal or close to recent ones, important parts of the investigated areas do show larger differences because of basin inversions which mainly took part during the Upper Cretaceous and the Tertiary (Baldschuhn *et al.,* 1991). Therefore, selected shales have seen a maximum temperature between 20 and 80°C. Within some of the basin areas Tmax was even higher because of special fluid flow conditions, magmatic activity, or drastic inversion movements. Although these severe higher temperatures and burial depths might give the impression of being very positive for the radioactive waste storage it has to be taken into account, that at least some of these shales underwent fracturing because of the rapid heating or the speed of the diagenetic processes.

Main part of screening stage 2 is the comparison of areas still in the screening process after stage 1 by using all available borehole, seismic, and well log data. In addition, the development of the screening methodology and the derivation of further criteria for shale suitability are part of this stage. Seismic and well log interpretation are done by using standard techniques. The studies were focused at first to areas where good digital data of seismic surveys in combination with boreholes with at least a suite of well logs (SP, gamma-ray, resistivity) were available. The shale content of the rocks is calculated by using the γ-index, the SP-index (see for instance Fricke & Schön, 1999; Doveton, 1994) and a depth dependent shale line taking into account different porosities of shales. Combining lithological descriptions of wells, the estimation of the shale content from well logs, and seismic data leads to a spatial view of the shale distribution in the subsurface of the investigated area.

Figure 1. Burial depths of Cretaceous shale formations in Northern and Southern Germany



Shale homogeneity and predictability are major issues in the assessment of shale potential. Therefore, a special method for well log correlation is part of our screening methodology. This method was developed by Olea & Sampson (2002), implemented with the computer program CORRELATOR and further developed for the screening process (Olea *et al.,* 2003). The program allows the geometry of the subsurface layering to be examined, along with some of its petrophysical attributes. It is an implementation of computer correlation that mimics the more conventional manual correlation of logs, which traditionally involves the simultaneous visual inspection of two logs per well, one of which is sensitive to the amount of shale. Mathematically, CORRELATOR solves the problem employing a form of weighted correlation coefficient, $w_{1,2,3,4}(i,k;n)$, defined as the product of a standardised shale-similarity coefficient, $\alpha_{1,3}(i,k;n)$, and the Pearsonian correlation coefficient, $r_{2,4}(i,k;n)$:

$$w_{1,2,3,4}(i,k;n)=\alpha_{1,3}(i,k;n)\cdot r_{2,4}(i,k;n),$$

where: i is the index in the depth z_i for the centre of an interval in the reference well;

k is the offset between the centre of the intervals being compared, measured in number of regular sampling intervals;

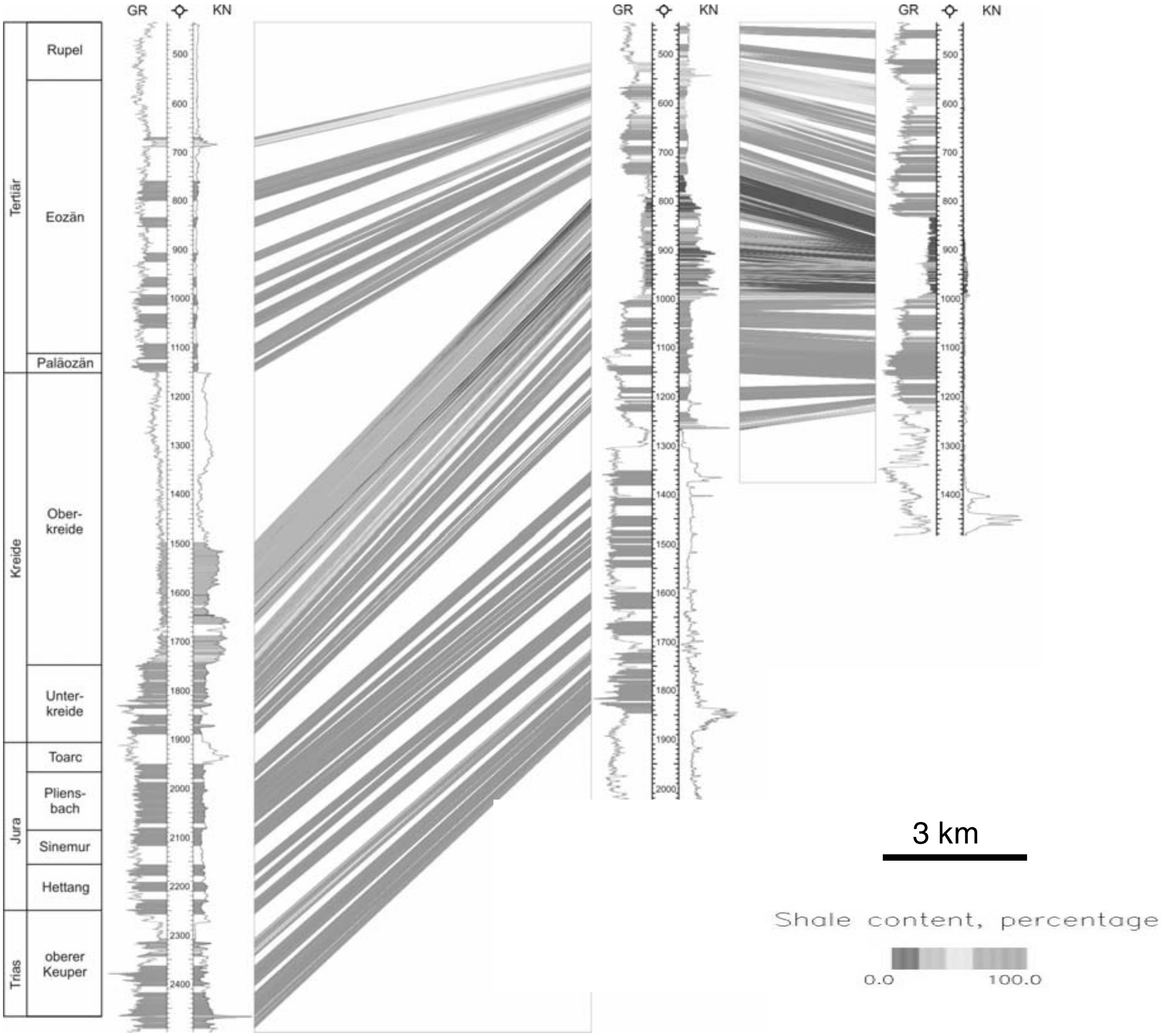
$(2n+1)$ is the number of readings in the interval.

In addition to the simplicity and efficiency of the approach, use of a weighted correlation coefficient has the advantage that the coefficient is an index for the quality of matchings. Thus, the weighted coefficient can be used in combination with a threshold to eliminate correlations of low reliability. CORRELATOR has an expert system that analyses all correlations between pairs of wells to bring inconsistencies to the attention of the interpreter, who has the power at all times to accept the advice of the expert system or to reject it if the geology is complex enough that abnormal correlations may be accepted as real matchings.

Figure 2 shows an example for one of the final result using the well log correlation method. The three wells are located in the eastern part of the North German Basin. Results document the intervals with strong correlation between the wells and clearly show that main shale intervals of the region are located in the Eocene/Paleocene, the Lower Cretaceous, and within the Lower Jurassic. It is furthermore obvious, that large differences in both shale content and homogeneity exist. Other available well logs (sonic, neutron, density) and in some cases also core measurements are used to estimate shale porosity and other shale properties. In the case of complete data sets, it seems to be even possible to come up with Clay Mineral Estimations from the Logs by Using a Calibrated Numerical Inversion Procedure (Bohling *et al.,* 1997; Hoth & Doveton, 1999). Several regions are compared with each other during screening stage 2.

The future screening stage 3 needs to start with a selection process, in order to define under the positive evaluated regions the ones which are best suitable for further investigation by new seismic measurements and exploration wells. For this selection all positive evaluated areas of stage 2 have to be ranked after selective criteria. They will be based on all former screening results and the already derived criteria but will have to consider the gained experience in the underground laboratories as well.

Figure 2. Example for the correlation of well logs (gamma, resistivity) and the determination of the shale content for 3 wells located in the eastern part of the North German Basin



Final remarks

Screening results so far show that thick and homogeneous shales exist especially in the Lower Cretaceous and the Middle and Lower Jurassic in Northern Germany and in the Middle Jurassic of Southern Germany. Although there are no question that these shales are in most cases good barrier rocks, questions with respect to the thermo-mechanical properties of these shales are the main points for the assessment of their utilisation as host rocks for radioactive waste.

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THE GEOLOGICAL EVOLUTION OF OPALINUS CLAY IN THE ZÜRCHER WEINLAND AREA (NE SWITZERLAND): LEARNING FROM THE PAST TO PREDICT FUTURE EVOLUTION AND STABILITY

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1. Introduction

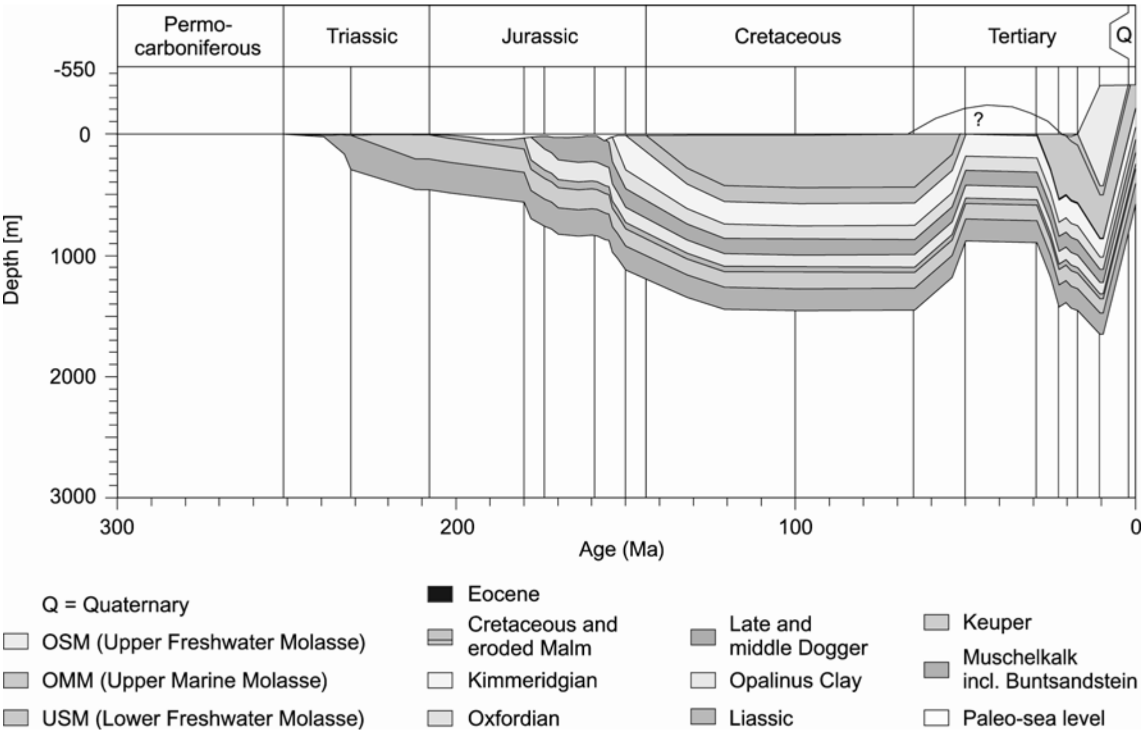
A number of safety-relevant issues need to be addressed when considering long-term evolution of a radioactive waste repository, out of which uplift/erosion, fault activity, and changes in the geochemical and hydrogeological environment are particularly important. Among the strongest arguments in the prediction of future evolution is the extrapolation of events and processes that occurred over a long period of time in the geological past (e.g. 10 Ma) to a shorter period in the future.

The future long-term evolution of Opalinus Clay in a potential siting area for a high-level waste repository in the Zürcher Weinland (NE Switzerland) is considered over a time period of around 1 Ma. The geological evolution or geological stability, respectively, can be predicted plausibly within reasonable limits over such a time period based on a detailed analysis of geological history. Predictions extending beyond this time period are feasible but contain an increasing element of uncertainty. This paper summarises the project-related conclusions, which are presented in greater detail in Nagra (2002a).

2. Burial and temperature history

A large number of techniques were applied to constrain the burial and temperature history of Opalinus Clay in northern Switzerland since its sedimentation 180 Ma b.p. These included stratigraphic evidence, apatite fission track analysis, organic maturity studies (biomarkers, vitrinite reflectance) and investigations of diagenetic cements and fluid inclusions therein. These methods were integrated in a basin model, and Figure 1 shows the resulting burial history of the basin in the Zürcher Weinland (Nagra, 2002a). The Opalinus Clay was subjected to two successive stages of burial and uplift. It reached a maximum depth (max. compaction) of ca. 1 650 m below surface some 10 Ma b.p., i.e. at the time of Molasse sedimentation (clastic sediments from the uprising Alps). A maximum temperature of about 85°C was reached during the first burial due to a higher heat flux during that time and due to a longer duration of burial. The rather high temperature to which Opalinus Clay was subjected during geological burial is only slightly lower than the 95°C that are expected at the bentonite/Opalinus Clay interface in the vicinity of MOX waste canisters over a few hundred years only (Nagra, 2002b). It could be shown that the geochemical, hydrogeological and geomechanical effects of the heat pulse on the host rock are negligible (e.g. illitisation of smectite, maturation of organic matter) or even beneficial (acceleration of self-sealing processes in the EDZ due to changed geomechanical parameters) (Nagra, 2002a).

Figure 1. Burial history of Opalinus Clay in the Zürcher Weinland based on basin modelling



3. Uplift and erosion

The reference depth for a high-level radioactive waste repository in Opalinus Clay is 650 m below surface (Nagra, 2002b). This depth may become smaller in future due to erosion, triggered either by uplift or by lowering of the erosion base level (back-erosion). Four independent methods were used to characterise uplift and erosion of the Zürcher Weinland: basin modelling, assessment of uplift of Quaternary gravel terraces and of pre-glacial peneplain, and geodetic studies (Nagra, 2002a; Müller *et al.,* 2002). The times over which these methods integrate uplift are quite different. Nevertheless, the resulting ranges are similar, and so multiple lines of evidence lead to consistent conclusions.

The region is located in the area of the foreland that underwent compressive stress as a result of Alpine crustal shortening. It is sensible, when evaluating long-term evolution, to assume that the movement recorded for the past will continue, at least over the time period of 1 Ma. Uplift will occur at a rate of about 0.1 mm/a, and through the (unlikely but possible) lowering of the base level of erosion (the Rhine river) until equilibrium is reached, an additional 100 m could be eroded away at most (singular event). This means that, in 1 Ma, the overburden of a repository constructed at a depth of 650 m will still be at least 450 m.

4. Fault activity

The potential siting area in the Zürcher Weinland is located in one of the seismically most quiet areas of Switzerland. So far, neither macro nor micro earthquake hypocentres could be correlated with faults in the investigation area. There is also no geomorphological evidence for the presence of active faults. Records of fluid flow related to earthquakes in the past, such as veins or geochemical anomalies, were not identified (Nagra, 2002a).

The last activity of most of the faults detected in the area could be dated relatively precisely based on a high-resolution 3-D seismic survey (Birkhäuser *et al.,* 2001), showing that fault activity has terminated millions of years ago. However, one fault (Neuhausen fault) which limits the potential siting area displaces Tertiary strata and – due to its orientation with respect to the regional stress field – has a propensity to be re-activated in the future (Müller *et al.,* 2002). A respect distance will be kept between a potential repository and this fault (Nagra, 2002a).

5. Magmatic activity

Magmatic activity is not expected to occur in the wider area of Zürcher Weinland within the next million years. The closest and last volcanic activity occurred about 7 Ma ago in the Hegau area in Southern Germany. There are no anomalies in the heat flux map of northern Switzerland that could provide arguments for a future hydrothermal or magmatic activity in the Zürcher Weinland (Nagra, 2002a).

6. Geochemical evolution

The long-term evolution of the Opalinus Clay pore water is complex (Nagra, 2002a). After deposition in a shallow marine environment 180 Ma ago, the connate pore water was modified by expulsion during compaction and by diagenetic rock/water interaction processes. A highly saline phase – probably related to the presence of a late Triassic evaporitic sequence below – is preserved in fluid inclusions in diagenetic carbonate cements. Today, the salinity of the Opalinus Clay pore water in the Benken borehole is about 30% of that of sea water, but it cannot be explained as a simple mixture of sea water and meteoric water. Loss of salinity may have occurred during early Tertiary, when a karstic system was active in the overlying Malm limestones for at least 20 Ma (see Figure 1), and during Quaternary by diffusive exchange with ground waters in aquifers above and below the Opalinus Clay.

The chemical composition of pore water in most clay-rich sedimentary rocks is largely determined by equilibrium reactions with minerals (clay minerals, carbonates, pyrite), which have a large buffering capacity (see Beaucaire *et al*., this volume). Mineralogy has slightly changed during diagenesis, but remained stable since and will do so even in the far future. Therefore, the same equilibria prevailing today will control pore-water chemistry in the future. Only the mobile (or conservative) elements such as chloride evolve independently of mineralogy, and for reasons of electroneutrality, transport of Cl will be accompanied by an equivalent transport of Na, predominantly by diffusion. Predictions of the future evolution of the salinity of Opalinus Clay pore water in the Zürcher Weinland have been made by Gimmi & Waber (2003) using a diffusion model, showing that the changes over 1 Ma are very small (i.e. less than 20% in the host rock formation, see also Figure 4 in Beaucaire *et al*., this volume).

7. Hydrogeological evolution

Hydrogeological measurements and observations in numerous boreholes and tunnels penetrating the Opalinus Clay throughout northern Switzerland indicate that a significant increase in the hydraulic conductivity of isolated fault zones in the Opalinus Clay will occur only if the overburden is less than 200 m (Gautschi, 2001). A drastic increase in hydraulic conductivity would be restricted to the uppermost few decametres. Because the rock overburden of a repository constructed at a depth of 650 m will still be at least 450 m after 1 Ma, the hydraulic conductivity of the surrounding host rock will be practically unaltered. The only major change from a hydrogeological point of view will be the possible dissipation of hydraulic overpressures in the Opalinus Clay and adjacent formations. This dissipation will interfere with pore-pressure changes related to erosional unloading or glacial loading. Vertical fluxes related to these pressure changes are negligible (Marschall, this volume).

8. Summary

The investigation area Zürcher Weinland is located at the edge of the zone influenced by Alpine tectonic activity, uplift and erosion and provides a stable environment for a deep repository for at least 1 Ma, most probably much longer. Due to its very low permeability and favourable mineralogical composition, the Opalinus Clay provides a stable hydrogeological and geochemical environment in which radionuclide transport is extremely restricted (Nagra, 2002b).

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THE EVOLUTION OF THE CALLOVO-OXFORDIAN ARGILLITE SITE, EASTERN FRANCE

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In France, investigations on the feasibility of a repository for long-lived radioactive waste in a deep geological formation are carried out according to a regulatory framework (Basic Safety Rule, RFS III.2.f) prescribing that the long-term geological evolution be taken into account to assess the dynamics of radionuclide transfers over the very long timescales involved.

As a matter of fact, the period in the future is in the order of 1 million years and is consistent with the timescales of geological evolutions likely to be significant, especially at ground surface, due to fast and high-amplitude climate variations. In order to achieve that goal, external and internal geodynamic phenomena that would generate the potential evolution are studied both on the surface and deep underground.

The purpose is to define a reference scenario where the evolution of the environment is induced by a series of very likely natural events in the future. In such a framework, the basis of the approach is to continue studying the geological evolution together with past situation analogies. The work also involves the description of one or several possible variations, such as potential extreme variations or disturbances of the evolution due to the impact of human activities. That aspect is particularly important with regard to the climate evolution, because of the hypotheses on the long-term effects of greenhouse gases. In that context, the goal is to forecast potential exceptional situations in the future with or without similar conditions in the past.

General presentation of the site

The clay layer being studied during current investigations on the feasibility of a geological repository through the underground laboratory on the Bure site settled during the Jurassic (Callovo-Oxfordian).

The Bure site is located in the eastern part of the Paris Basin, an emerged area the surface of which has been submitted to erosion/alteration processes since the Cretaceous.

Geologically speaking, the region is located far away from the Alpine front and any zone of tectonic deformation; it also appears as practically aseismic at the scale of historical times.

Sedimentary layers were deposited during the Mesozoic and currently show a slight extension associated with the downfold shaping of the Paris Basin, resulting from a composite Mesozoic and Cenozoic history involving only very little movements. In that context, the clay layer under review has a thickness of 130 m and a depth of 400 m on the underground laboratory site, located away from the rare faults detected in that region at the scale of 2-D seismics and of the geological map (faults of Marne and Gondrecourt rifts).

Geographically speaking, the sector being investigated is characterised by outcrops of Jurassic formations where clay and limestone layers alternate, thus allowing the development of contrasted reliefs: valleys cut out in cuesta landscapes with limestone coastlines and marl depressions laid out according to the outcrop aureoles of the large lithological blocks of the substratum.

In addition, it is located away from the extensions of the large ice sheets that develop in Northern Europe and in mountain regions during cold-climate episodes.

In such a general context where the geological evolution is both slight and slow *a priori,* the purpose is: (1) to know the past evolution of the host formation in order to understand the current state and the 3-D distribution of the characteristics of the different layers of the subsoil; and (2) to estimate the changes that might occur in the future over the lifetime of the repository and their impact on the characteristics of the host geological formation in relation to radionuclide transfers to the biosphere.

Investigation methodology

The paper will illustrate various possibilities to address that issue with examples of implemented studies that rely on the geoprospective approach developed by Andra, including its two main goals:

- To understand, reconstruct and quantify the past evolution of the region in order to reduce uncertainties on the causes and conditions of that evolution (amplitude, dynamics, dates of past events) by relying on multiproxi analyses. Those analyses are applied at different spatial scales (overall, regional and local) and timescales (climate variations combining cycles ranging from one year to about 100 000 years; the tectonic evolution operating over time periods measured in terms of tens of millions of years).
- To propose scenarios for the future evolution of the site in its regional environment, on the basis of geodynamic-evolution models serving as the driving force for that evolution: tectonics and climate. In order to achieve that goal, two complementary approaches are jointly being developed:
  - Conceptual models using acquired data and knowledge through field work: geological evolution of the clay formation and of its surroundings since their settlement, quantified analyses and reconstructions of the recent evolution of the surface conditions (erosion, geomorphology), as well as analyses of natural analogues (notably for the characteristics of biospheres during cold periods).
  - Digital simulations calibrated on past data, calling upon a series of digital models. Those simulations concern not only the possible evolution of the climate in the future, but also the processes involved in that evolution of the surface conditions on the site and in the region due to the joint actions of the climate and tectonics (erosion, permafrost and geomorphological evolution in general, as well as glacial isostasy). Those models will particularly help to understand better the uncertainties resulting from basic hypotheses on the driving forces of the evolutions and to describe possible future situations that do not have any past analogues or that are not accessible through field studies.

Those site-specific thematic studies are carried out in the framework of a research group association (known as “bio-geoprospective group”) in order to ensure the overall consistency of those studies encompassing a very large spectrum of scientific disciplines with a view to having over the long term a conceptual model integrating the evolution of the geosphere and the biosphere.

In that framework, future climate variations constitute not only an input variation for all themes, but also overall data weighed down by heavy uncertainties due to the incorporation of human activities and the potential effects of greenhouse-gas releases. More than for any other variable, it is therefore important to address it at the international level, an endeavour that remains achievable in the framework of European projects, such BIOCLIM (5th FPRD) to be completed at the end of 2003.

Evolution of surface conditions having a direct impact on transfers to the biosphere

All changes to surface conditions concern the geomorphological evolution, and especially the topography, due to the erosion processes induced by the joint effects of slow vertical tectonic movements and fast climate variations.

Field geomorphological studies help to reconstruct the evolution phases of that topographical surface during the last millions of years and of the significant restructuring of surface runoffs that accompanied them due to processes involving fluvial-capture processes. That process is characterised by strong space and time variations in eroding velocities over time, depending on the elements of the relief (valley, coast line, limestone plateau, etc.) and the period (climate phase, anterior and posterior fluvial captures, etc.) under consideration. A high differential has resulted in the average eroding velocities over the long term between valleys and limestone-plateau areas, a situation that leads to increasingly significant topographical differences of level.

That evolution will continue in the future in response to the acquired topographical situation and to regional vertical movements (glacio-eustasy and glacial isostasy regulated by climate cycles; slow regional tectonic upheaval movements) under the action of erosion phenomena controlled by climate conditions. The periodical development of a more or less thick and continuous permafrost must also be taken into account according the timescales and climate-evolution scenarios.

Any occurring changes do not affect the host formation, but have an impact on hydrogeological models in terms of:

- The limit conditions depending on the average evolution rates: topographical changes, and particularly, the displacement of outcropping zones of the aquifers (potential outlets).
- Internal conditions to hydrogeological systems, cyclic changes (evolution of the permafrost) and acquired changes (evolution of karsts).

Those changes are involved directly in the definition of the biosphere system, depending on landscape changes, physical and biological environments occurring throughout climate fluctuations and due to the effective dynamics of the processes (distribution of erosion and sedimentation episodes, especially during climate cycles).

To take in account this evolution, in order to evaluate the transfers from the Callovo-Oxfordian formation to the biosphere, Andra develops a simulation of the futur hydrogeological circulation in the different layers, at regional and local scales, aiming to define the streamlines and outlets localisation changes and transfer time for the different geological evolution scenarios.

The preliminary hydrogeological models that have already been developed during the current study level explore major potential changes over the long term (500 000 years ; 1 million years) that would occur in the framework of the hypothesis of a climate-variation scenario undisturbed by human activities. The geomorphological evolution lead to displacement of natural outlet zones of the aquifer overlying the host formation link to the apparition of new outcrops in valleys witch progressively tends to capture the streamlines.

Stability analyses of the subsoil, possible changes of the characteristics of the host formation and its surroundings

At a totally different scale, the purpose of R&D is to reconstruct, understand and date the phases of diagenetic evolution of the clay formation (see Session IV concerning the Bure site). It supports the characterisation and spatial distribution of the properties of the host formation and of its surroundings. In general, the characteristics of the clay formation seem to have been acquired in the framework of the burial phase and not very subject to changes over the next million years, especially since the site has been set in a continentalised zone for more than 65 Ma and expected to remain as such in the future.

In that framework, a specific action is carried out to assess the geochemical stability of the formations constituting the subsoil by improving our understanding of the fluid-rock interactions that occurred due to the past major continental paleosurfaces (surfaces of the Lower Cretaceous and Paleogene surfaces at the scale of the Paris Basin) and their processes from those surfaces: reconstruction of surfaces (according to their current layout) and geochemical profiles of altered layers underlying those surfaces.

In addition, is raised the issue of the seismic-hazard estimation and of its potential impact on the geological environment.

Taking into consideration the very long timescale involved with the post-closure phase of a geological repository, ranging from hundreds of thousands of years to 1 million years, leads, even in a region with very slow deformation velocities and very little seismic action at the historical scale such as the Meuse/Haute-Marne region, to the possibility that significant earthquakes may occur during that time. In fact, the timescales involved are at the scales required for the accumulation of tectonic stresses (on pre-existing faults) in order to produce exceptional earthquakes in terms of magnitude and return periods. It results in the notion of the maximum physically-possible earthquake derived from the French regulations (RFS III.2.f).

Significant uncertainties exist for the estimation of such exceptional events, becoming possible only over the very long term, with very long recurrence periods. The method applied for estimating that risk allows for taking into account most of the uncertainties in order to provide a likely spectrum, in relation notably to the respective weights attributed to the various seismo-technical models based on the current knowledge on geological accidents and their potential activity.

Lastly, the *risk* is dominated by the potential sources close by, irrespective of the seismo-technical model being considered, with due account to the possibility of activation of certain faults by the field of tectonic stresses, likely to be the source of significant seismic events with very long return periods. A first analysis reveals that if they were to occur, those events may ultimately modify the local hydrological characteristics at the level of the surrounding limestones and of the neighbouring faults around the sector, which are located beyond the potential natural discharge points of the aquifers and would therefore not have any significant impact on those transfers.

Conclusions

The case of the Meuse/Haute-Marne site illustrates the notion of geosphere stability required in the framework of the repository concept in a clay formation and the possibilities to estimate the possible changes in the future due to the natural evolution of the geosphere and of the biosphere.

Over the lifetime of a geological repository (i.e. the next million of years), the geological evolution in Eastern France will generate noticeable changes at the ground surface; those cumulative and irreversible changes will derive from erosion/sedimentation processes combined with cyclic changes associated with direct climate effects (permafrost, plant cover). There will also be, in the continuity of the past and irrespective of the climate scenario, a topographical evolution of the drainage networks, soils, ecosystems, and consequently, probable important changes for runoffs and radionuclide pathways beyond the host formation towards the biosphere, notably in terms of changes to the existing conditions at the natural outlets. The precise knowledge of those overall changes is important for conducting safety analyses (dose calculations) and justifies the development of studies on that specific subject.

However, following the detailed studies carried out to characterise the host formation (the Callovo-Oxfordian clay layer) *in situ* through the underground laboratory, the prognosis is that its characteristics should remain highly stable, thus ensuring the containment of radionuclides.

EVALUATION OF LONG-TERM GEOLOGICAL AND CLIMATIC CHANGES IN THE SPANISH PROGRAMME

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The Biomolecular Stratigraphy Laboratory of the Madrid School of Mines has been largely involved in the analysis of long-term paleoenvironmental changes in the Iberian Peninsula during the Quaternary. Some of the research projects were UE funded: Paleoclimatological Revision of Climate Evolution in Western Mediterranean Region. Evaluation of Altered Scenarios (CE-FI2W-CT91-0075), Evidence from Quaternary Infills Paleohydrogeology (F14W-CT96-00 Nb 960296), Sequential BIOSphere modelling function of CLIMate evolution models (BIOCLIM); Paleohydrogeological Data Analysis and Model Testing (FIKW-CT2001-00129). Other projects were funded by the National Company for Radioactive Waste Management (ENRESA) and the Spanish Nuclear Safety Council (CSN): “Paleoclimate reconstruction from Middle Pleistocene times through dating and isotopic analysis of tufa deposits”; “Paleoenvironmental evolution of the southern part of the Iberian Peninsula”; “Paleoclimate”. On a minor scale the laboratory was also involved in the study of some argillaceous media: “Organic Geochemistry of some deep Spanish argillaceous formations” and “Effects of climatic change on the argillaceous series of the Duero and Ebro basins”. Here we will present some of the results obtained from tufa deposits analysis and paleoenvironmental information from the Guadix-Baza Basin composite-stratigraphical-type-section study.

1. Tufa deposits of Central Spain

In the Central Part of Spain, extensive tufa deposits appear associated with the western boundary of the Iberian Range, where the rivers that go across narrow canyons reach flat Tertiary basins. In the study area (the Priego zone) three tributaries located at the head of the 1 000 km long Tagus River built terraced tufaceous deposits, Figure 1. These deposits can be explained because of degassing processes that took place after the $Ca(CO_3H)_2$-saturated karst-origin running waters lost their confinement. Tufa show both autochthonous and allochthonous facies. Extraclastic deposits made of quartzite and limestone clasts from Mesozoic-age rocks appear.

Figure 1. Geographical location of Guadix-Baza Basin and Priego area

According to the geomorphologic analysis six terraced levels have been differentiated. For dating purposes radiometric (U/Th), geophysical (paleomagnetism and ESR) and geochemical methods (amino acid racemisation) were employed. All the terraces sampled for ostracod recovery and amino acid racemisation analysis in a GC (Goofriend's, 1991 protocol). D/L ratios of aspartic acid and glutamic acid were used for dating purposes. After a cluster analysis of the obtained values six different Aminozones (AM1-AM6) were identified. The obtained aminozones matched well for each river terrace system. In order to calculate calibrated age calculation the algorithms defined by Ortiz *et al*. (in press) were used. The obtained results, Figure 2, were as follows: AM1: 407±12 ka (11th OIS); AM2: 263±14 ka (7th OIS); AM3: 181±17 ka (7th OIS); AM4: 136±13 ka(6th-5th OIS); AM5: 108±14 ka (5th OIS); AM6: 11±4 ka (1st OIS). Later the incision rates in the headwater zones of the rivers of the central part of the Iberian Peninsula were calculated (Table 1).

Figure 2. Aminochronology of the Priego area stratigraphic sections after the numerical age calculation from the D/L Asp and D/L Glu values
They are correlated with the marine Oxygen Isotope Stages (OIS)

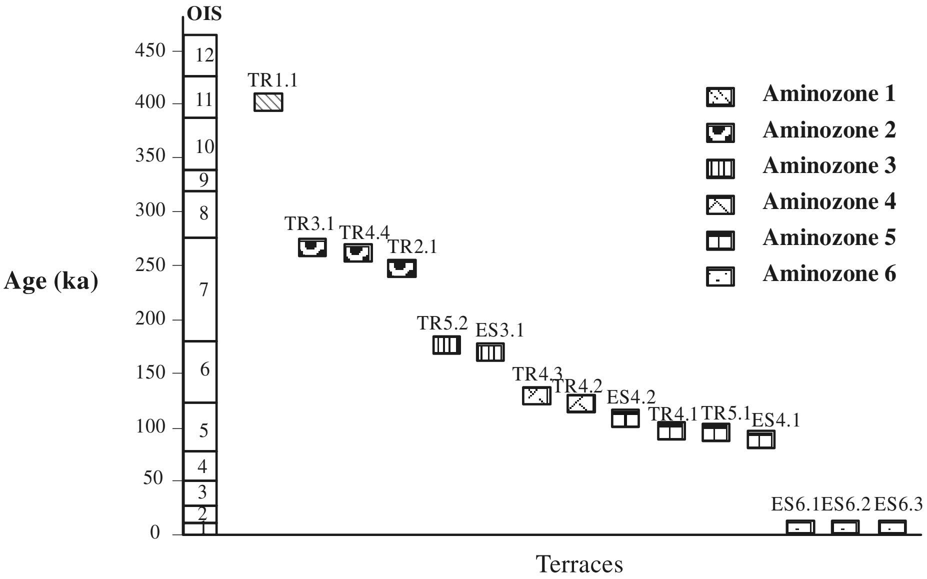


Table 1. Incision rates in the headwater zone of the Trabaque river
(central Iberian Peninsula)

Terrace	Age (yr)	Elevation above the Trabaque river talweg (m)	Incision rate (yr/m)
TR1.1	407 566 ± 12 543	100	5.131
TR2.1	253 636 ± 10 188	70	3.644
TR4.1	107 856 ± 8 685	30	3.595

2. Lacustrine deposits of the Guadix-Baza Basin

The Guadix-Baza Basin (Figure 1) is a long and narrow basin (4 500 km^2) which according to Pous *et al.* (1995) is nested between the Internal and External Zone of the Betic Range. The sediment fill of the Basin comprises gravels, sands, lutites, limestone and gypsum. Ancient evaporites affected sedimentation then as now as evidenced by the saline springs which feed rivers today. The facies distribution shows a clear centripetal pattern, with alluvial fan systems reaching the basin through still visible gorges. A complex system of sandy beaches, mud dry playa, mud flat playa and marginal lacustrine sands, connected central lacustrine environments and marginal alluvial fans was developed. In order to find a pristine lacustrine "composite-stratotype-section" containing minimal diagenesis for geochemical analyses, including amino acids and stable isotopes we selected marginal lacustrine deposits.

The composite-stratotype-section is 253 m-thick was partially described by Oms *et al.* (1994) and Calvo Sorando (in press) and Torres *et al.* (in press). For descriptive purposes we have divided this section into three different parts: Lower Lutitic Part (55 m thick), Sandy Intermediate Part (75 m thick) and Upper Heterolithic Part (123 m thick).

The section was dated using previously published palaeomagnetic analysis (Oms *et al.* 1994; Agustí *et al.* 1999) and our own data Ortiz (2000). Likewise, with the aid of the amino acid racemisation method, we have established its chronostratigraphy.

Fossils are abundant in the basin record: mammals, crustaceans, molluscs, plants, silicified tree logs and biomolecules, being scarce pollen grains. All of them were analysed for the paleoenvironmental analysis but our study was focused on ostracods because (Ortiz, 2000) they appeared along the whole composite-stratotype-section and their valves were made of low-Mg calcite which formed in isotopic equilibrium with the lacustrine environment water. The most common representative is *Cyprideis torosa* (Jones) that colonises waters with a wide range of salinity. When salinity is high, it can be the only species remaining.

Their calcitic shells were analysed in order to obtain their δ13C and δ18O values, later a cluster analysis from δ^{18}O and δ^{13}C values was made (complete linkage and euclidean distance) showing that values cluster into three groups: 1) Low δ^{18}O and δ^{13}C values; 2) High δ^{18}O and δ^{13}C values; 3) Intermediate δ^{18}O and δ^{13}C values. These groups are also distinguished in the linear regression plot between δ^{18}O and δ^{13}C and can be matched with three different climatological scenarios.

Group 1. Low δ^{18}O and δ^{13}C (Humid and Cold). The cold conditions and the amount of rain would incorporate waters with more negative δ^{18}O. Low evaporation rates should produce water bodies with low δ^{18}O values. The δ^{13}C would also be low as a result of the humid conditions, low evaporation rates, the decrease of low primary productivity and, probably, the presence of more amounts of superior plants.

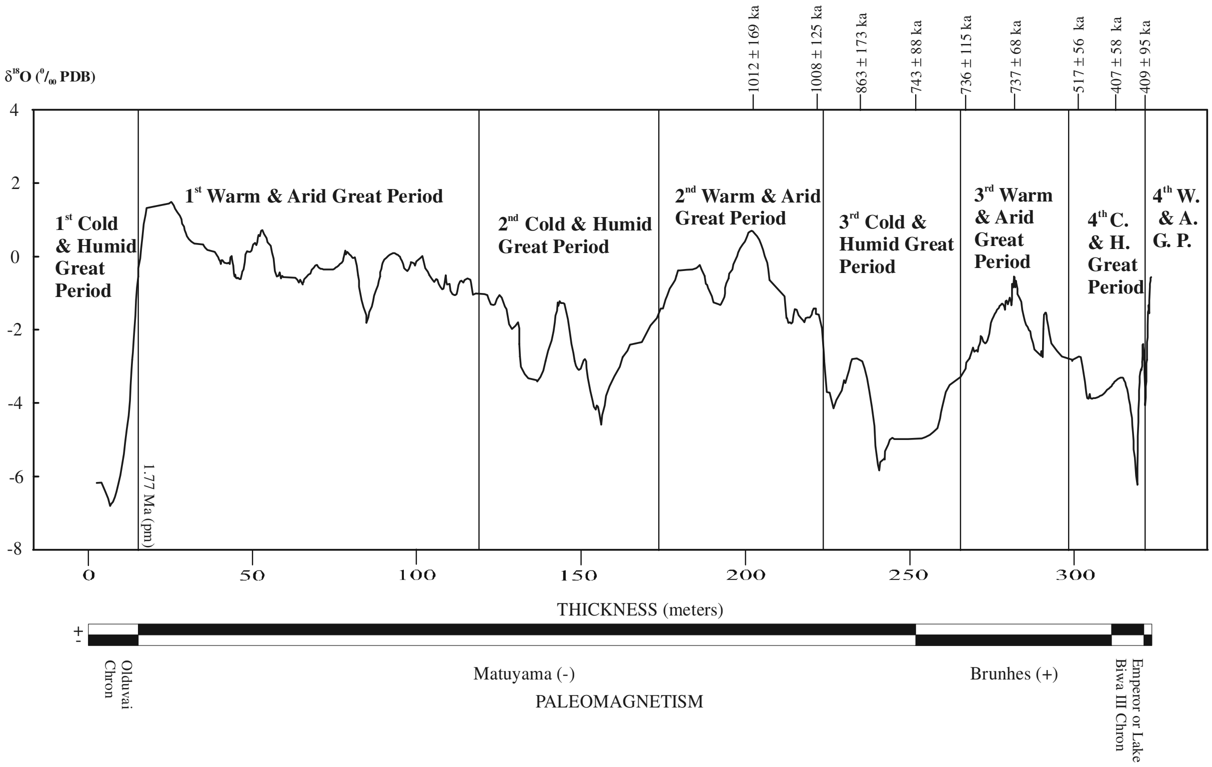
Group 2. High δ^{18}O and δ^{13}C values (Arid and Warm). The high δ^{18}O and δ^{13}C values are a consequence of higher evaporation rates and temperatures. Algae and macrophytic plant development would produce an increment of the δ^{13}C values. Vegetal cover would comprise C_4 plants, with less negative δ^{13}C.

Group 3. Intermediate δ^{18}O and δ^{13}C values (Intermediate scenario).

Paleosalinity analyses of fluid inclusions from intrasedimentary gypsum crystals confirmed this interpretation.

According to this, it has been possible to establish a paleoclimatic succession consisting of alternations of long periods of "warm and arid" and "cold and wetter" conditions (Figure 3), this being comparable to the climate evolution in other Mediterranean realms as the Hula Basin in Israel (Horowitz, 1989). In USA, basins with a similar paleoenvironmental evolution appear, such as the pluvial lakes in the "Basin and Range" zone.

Figure 3. Smoothed curve of the δ^{18}O values obtained in *Cyprideis torosa* (Jones) ostracodes from Guadix-Baza Basin with the identified paleoenvironmental periods



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SESSION IV

REACTION OF ARGILLACEOUS MEDIA VIS-À-VIS NATURAL PERTURBATIONS AND GEOSPHERE EVOLUTION (BUFFERING)

CLAY CLUB INITIATIVE: SELF-HEALING OF FRACTURES IN CLAY-RICH HOST ROCKS

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Introduction

The capacity of fractures in argillaceous rocks to self-heal (or become, with the passage of time, less conductive to groundwater) is often cited as a primary factor favouring the choice of such materials as host rocks for deep disposal. The underlying processes which contribute to self-healing can be broadly subdivided into: (a) mechanical and hydromechanical processes linked to the change in the stress field, movement of porewater, swelling, softening, plastic deformation and creep, and (b) geochemical processes linked to chemical alterations, transport in aqueous solution and the precipitation of minerals. Since chemical alteration can cause profound changes to the mechanical properties of argillaceous rocks, it is often difficult to draw a firm line between these two sub-divisions. Based on the deliberations of the recent Cluster Conference in Luxembourg, there would appear to be some support for the use of the term "self-sealing" for processes affecting fracture conductivity in argillaceous rock that are largely mechanical or hydromechanical in their origin.

There are four main areas in which the self-healing capacity of the host rock becomes relevant to repository design and performance assessment:

- potential for radionuclide transport within the excavation damage zone (EDZ);
- design and performance of repository sealing systems;
- potential impact of gas migration;
- long-term performance considering erosional unloading, seismicity and fault reactivation.

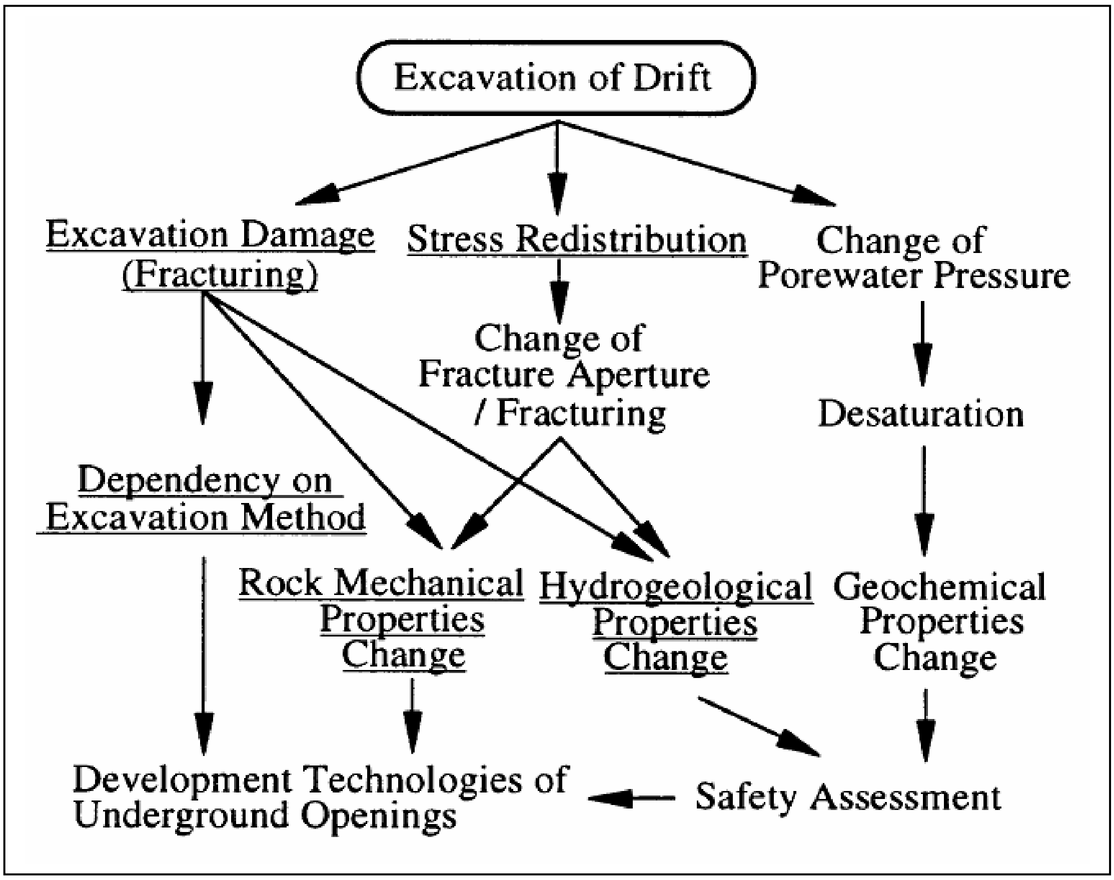
The presence of an EDZ is acknowledged to be a particularly important issue in performance assessment (Meier *et al.,* 2002). Interconnection of fractures in the EDZ could lead to the development of a preferential flow path extending along the emplacement holes, access tunnels and shafts of a repository towards overlying aquifers and the biosphere. In the preliminary French Safety Analyses, for example, the treatment of scenarios relating to early seal failure have highlighted the hydraulic role of the damaged zone as a potential radionuclide transport path (Anon, 2001).

Excavation damage zone

Figure 1 is a schematic of processes occurring in the EDZ around a test drift. The size and the properties of the EDZ depend on the excavation method, the state of stress, the pore water pressure and the hydro-mechanical properties of the rock. Bedding plane anisotropy can be an important factor.

Blasting tends to cause more damage than other methods of excavation. Damage associated with tungsten-carbide tipped tools fitted to roadheaders and similar tunneling machines tends to be much more localised. In clays and argillaceous rocks, the most pervasive forms of damage are caused by stress redistribution and unloading. Three basic forms of fracturing may be defined: (a) shear fractures, (b) tensile fractures, and (c) extensile fractures. Recent experience during development operations in several URLs clearly demonstrate that the dominant mode of fracturing can be quite different from one mudrock to another.

Figure 1. Processes occurring in the EDZ (Sato *et al.,* 2000)



Conspicuous shear fracturing was observed during recent tunneling operations in the Boom Clay. It seems clear that the shear stress ahead of the tunnel face has locally exceeded the undrained shear strength of the clay. Fracturing in more indurated mudrocks such as the Opalinus Clay would appear to be more complex. Unloading fractures are fairly ubiquitous and are suggestive of a form of extensile failure (Figure 2). The rapid reduction in radial stress around an excavation leads to a stress state that approximates to one of triaxial extension. The rock therefore responds to the stress relief by stretching in the radial direction. These rocks are incapable of sustaining large extensile strains without rupturing. Unloading cracks are therefore formed normal to the minor principal stress. This mode of failure may be similar to the "core-discing" phenomenon observed during drilling. Borehole breakout has also been observed during drilling in Opalinus Clay. There is also evidence of a bedding-plane slip mechanism which would seem to be a specific form of shear failure.

Figure 2. Extensional fracturing around the DI Niche at the Mont Terri Rock Laboratory in Switzerland

Dilatancy boundary

Horseman (2001) used the basic concept of the dilatancy boundary to interpret the temporal evolution of permeability along a stress path. Qualitative arguments were based on the Modified Cam-Clay Model (MCCM) which describes the deformation and failure of porous elastoplastic materials. The state boundary surface was assumed to comprise the Hvorslev Surface for material on the "dry side" of the critical state line and the Roscoe Surface for material on the "wet side" (Shah, 1997). Shear deformation on the "dry side" favours dilatant shear fracturing, whereas deformation on the "wet side" favours "contractant" behaviour. The MCCM, as currently implemented, does not adequately describe the effect of the intermediate principle stress in the development of fractures and, as a result, grossly overestimates strength in triaxial extension. Given the importance of extensile fracturing around tunnels driven in more-indurated mudrocks, this proves to be a significant handicap in the quantitative analysis of EDZ fractures in such rocks. Bardet (1990) demonstrates that it is feasible to modify the Cam-Clay approach to describe true-triaxial behaviour by invoking Lode-dependency in the specification of the state boundary surface.

Indurated mudrocks

Research at the Mont Terri Rock Laboratory in Switzerland has revealed the main characteristics of the EDZ in moderately indurated rock-types comparable to the Opalinus Clay (Thury and Bossart, 1999; Bossart *et al.*, 2002). Figure 3 shows a conceptual model of the fracturing around a tunnel. The inner zone, identified as f_1 extends around 1 m into the rock and consists of a network of interconnected and air-filled unloading fracture with substantially enhanced transmissivities (values varying between 10^{-5} and 10^{-9} $m^2.s^{-1}$). The highest transmissivities are found in the first 40 cm of the tunnel wall. The second or outer zone, identified f_2 in the figure extends a further 2 m into the rock and consists of partially-saturated unloading fractures. These tend to be isolated and therefore have a much lower degree of interconnectivity. The transmissivities are substantially less than in the outer zone (values varying between 10^{-9} and 10^{-12} $m^2.s^{-1}$). At Mont Terri, oxidation phenomena such as gypsum spots on fracture surfaces have been observed only in the inner f_1 zone of the EDZ. Figure 4 illustrates the radial distribution of permeability around an excavation.

Figure 3. Conceptual model of the excavation disturbed zone at the Mont Terri Rock Laboratory
(Bossart *et al.*, 2002)

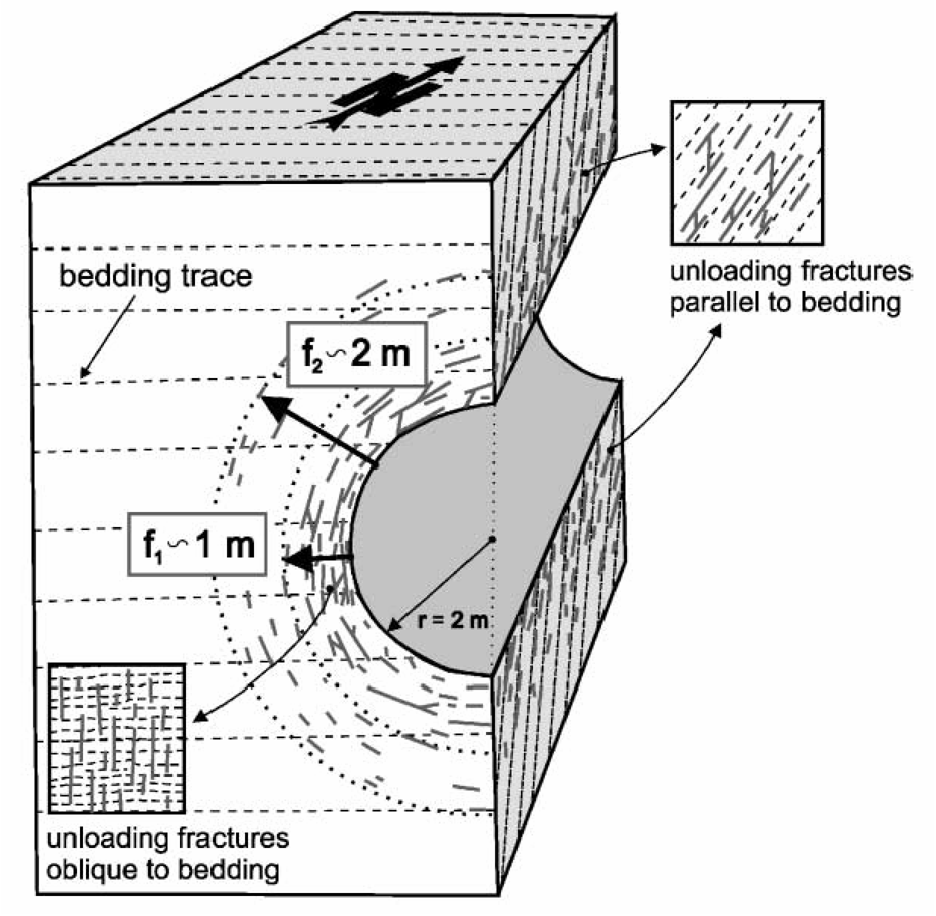
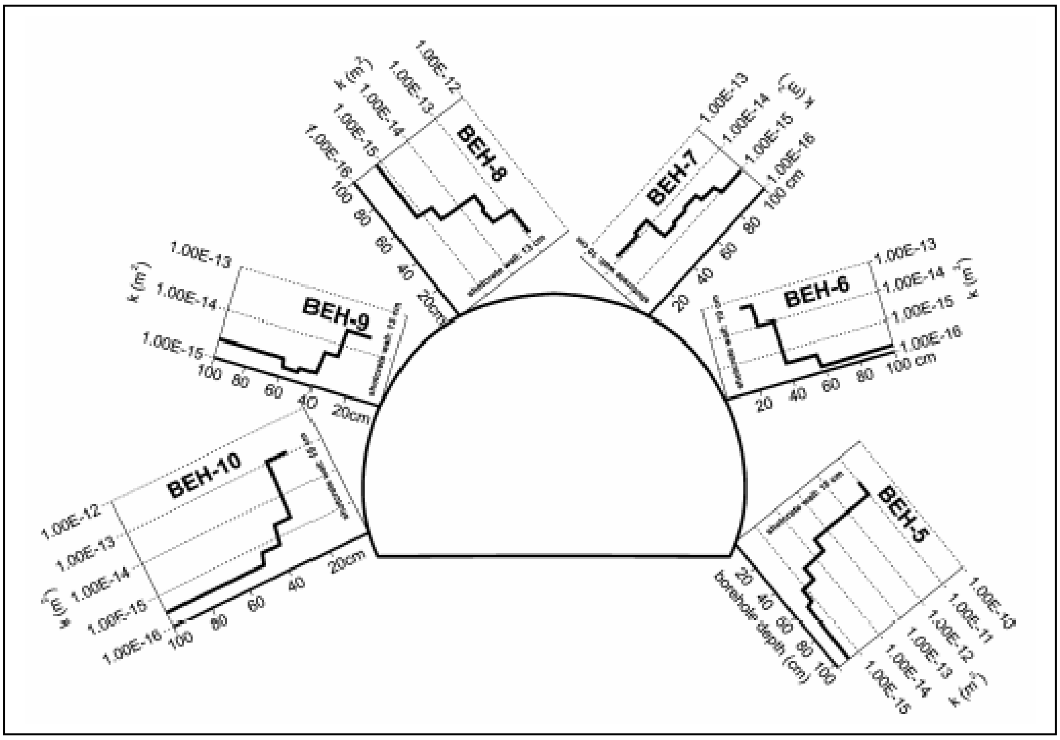


Figure 4. Permeability to air in a section of the new gallery at the Mont Terri Rock Laboratory
(Bossart *et al.*, 2002)



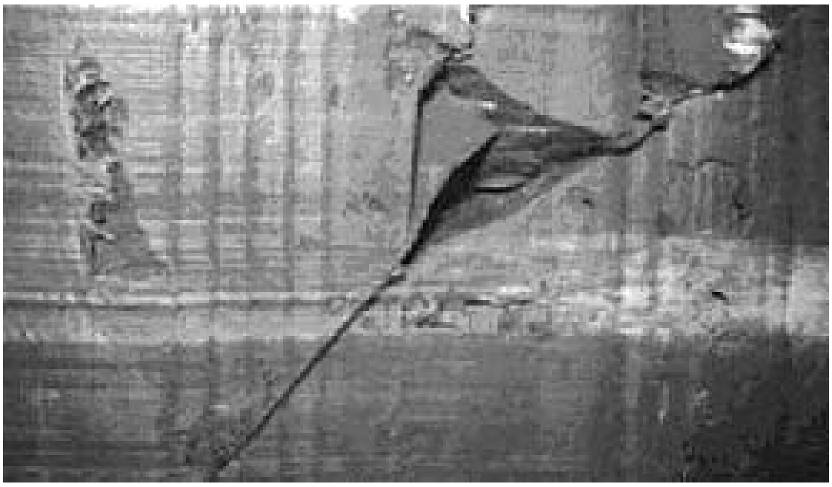
At Tournemire in Southern France, a railway tunnel built about 100 years ago is used as a laboratory for the generic scientific and geotechnical investigations related to radioactive waste disposal. The excavated disturbed zone (EDZ) around the tunnel is well expressed by a regular set of induced fractures along the vertical walls of the tunnel. The fractures are sub-vertical, parallel to the walls of the tunnel and intersect sub-horizontal fractures corresponding to open stratification beds. This results in a continuous zone adjacent to the tunnel comprising parallelepiped blocks placed side by side, generally covered by gypsum formed by oxidation (Charpentier *et al.*, 2001). The opened-fracture zone of the EDZ extends about 150 cm from the tunnel surface (Charpentier *et al.*, 2003).

Plastic clays

It is widely recognised that overconsolidated plastic clays exhibit inhomogeneous strains at failure, with deformation focussed along discrete macroscopic failure planes. Strain-softening is usually observed. Slickensides are a conspicuous feature of shear failure in such materials.

Shear fracturing was observed during underground development work at the Hades URL of the Belgian Nuclear Research Centre at Mol (SCK•CEN). A second shaft was sunk in 1998-1999. Two chambers were excavated in the Boom Clay at the bottom of this shaft to enable development of the horizontal gallery. Excavation of the starting chambers caused face instability problems resulting in significant fracturing of the clay to the excavation. Large fracture surfaces were visible at the face of each starting chamber, with fracture traces extending around the sidewalls. The slip surfaces consisted of an interconnected network of conjugate planes inclining at 35° towards the centre of the shaft. The circular shape of the slip surfaces indicated that they were symmetric around the shaft axis. Slickensides, visible on the slip surfaces, clearly indicated shear movement towards the centre of the shaft (Anon, 2002). Extensive tunnel operations were undertaken 2002 to connect the URL to the new shaft. Large fractures developed in the face of the excavation side-walls and in the sidewalls (Figure 5). Blocks of clay bounded by slickensided shear planes and tension joints became detached and caused difficulties in tunneling operations (Dehandschutter, 2002). Geometric considerations suggest that most of the fracturing was caused by the process of excavation.

Figure 5. Excavation-induced fracture in the side-wall of the connecting gallery in the Hades URL at Mol in Belgium (Dehandschutter, 2002)



Desaturated zone

The combined effect of extensile and shear-induced dilatancy in undrained situations is a significant reduction in the pore pressure. It seems probable that pore pressures can actually become negative in sign (i.e. a state of suction) close to the walls of a repository excavation, leading to localised desaturation. Ventilation can also cause localised desiccation, and increase in suction and further desaturation. The aperture of EDZ fractures is sensitive to variations in the humidity of the air. Rock properties such as shear strength will change with change in the suction.

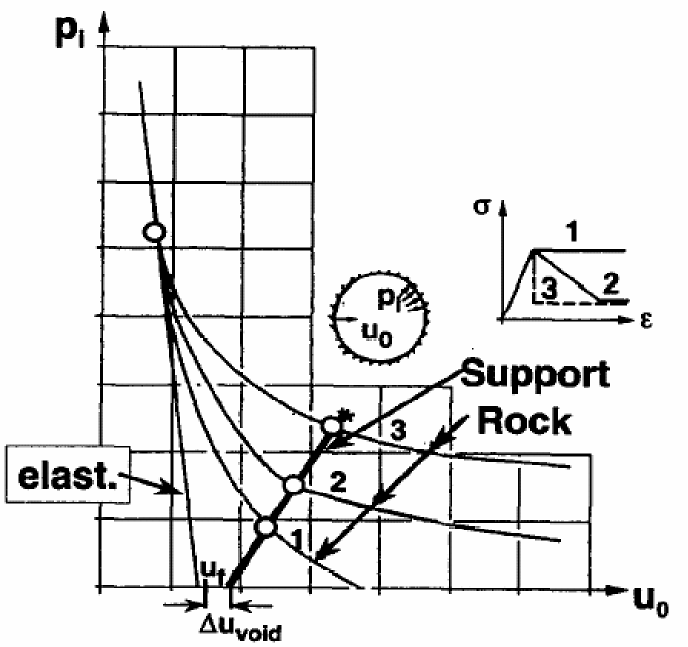
Support and backfilling

The provisions for made long-term support of repository excavations are of first order importance in assessing the capacity for EDZ self-healing. Various systems of support systems proposed including: sprayed concrete (shotcrete), steel arches with concrete lagging, concrete block linings, cast iron tubbing and cast-in-place monolithic concrete linings. Prior to repository closure, the void spaces will be backfilled with clay (e.g. bentonite) or a mixture of clay and sand. In some repository designs, temporary support will be provided using steel-fibre-reinforced shotcrete in combination with rockbolts and weldmesh. Long-term support will be assured by filling the tunnel section with a swelling clay backfill which will also serve to minimise groundwater movements. If this backfill is formulated to have a significant swelling pressure, then as it hydrates it will exert a radial stress on the shotcrete and the surrounding rock.

Force transference between the rock to the support system is a matter of central concern in tunnels engineering. One approach to the design of a tunnel lining uses the so-called "Convergence-Confinement Method". This involves the calculation of a "ground response curve" (or rock characteristic line) and a "support response curve". Convergence is expressed in terms of the radial displacement of the tunnel wall. The ground response curve is a plot of the convergence against a hypothetical pressure applied uniformly to the tunnel wall, known as the confinement. The ground response curve is calculated using a model for the strength and deformation of the rock. For concrete linings the support response curve is usually calculated assuming linear elastic stress-strain behaviour. By superimposing the response curve for the lining on that for the lining and making allowances for any overbreak (or gap) behind the lining and the rock, it is possible to estimate the maximum radial stress that will be carried by the lining.

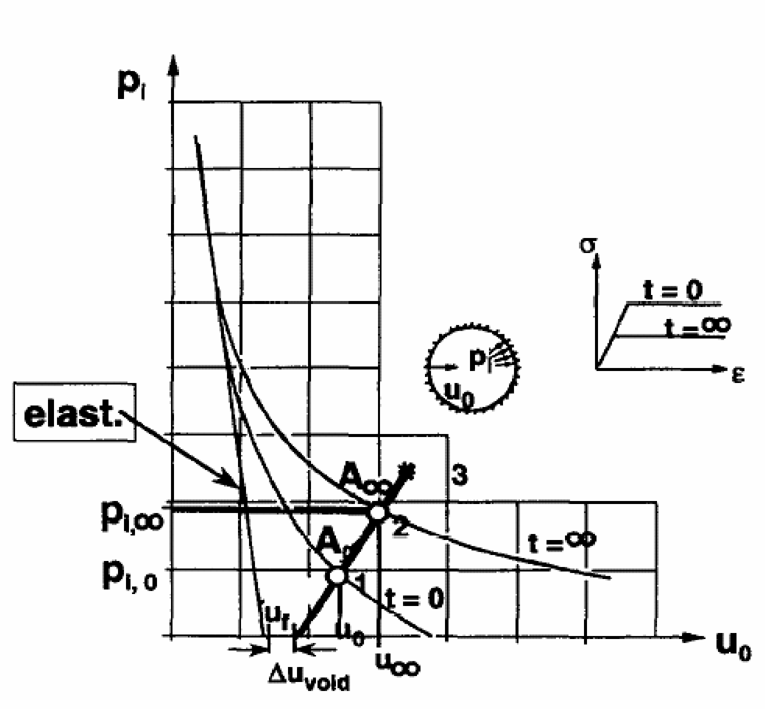
Tunnel engineers frequently assume that soft rocks exhibit either: a) perfectly elasto-plastic; b) strain-softening elasto-plastic; or c) elasto-plastic-brittle behaviour. The constitutive model is usually formulated in terms of total stresses and therefore neglects the effect of pore pressure dissipation on the redistribution of stress. Figure 6 shows a schematic of the ground response curves based on the assumption of elasto-plastic behaviour.

Figure 6. Convergence-confinement analysis of the interaction between the rock and the tunnel support system, based on: (1) elasto-perfectly-plastic rock deformation; 2) elasto-plastic strain softening deformation and 3) elasto-plastic brittle deformation



The argillaceous rocks of interest as potential host-rocks in radioactive waste disposal show a pronounced rate-dependency in their stress-strain behaviour. Creep deformation (see below) is very important in such rocks and the ground response curve therefore changes progressively over time. Figure 7 illustrates this for a rock in which creep is only significant when the deviatoric stress is larger than a specific threshold value. The stress acting on the lining is predicted to rise asymptotically with time up to some maximum limiting value.

Figure 7. Convergence-confinement analysis of the interaction between the rock and the tunnel support system based on the assumption of creep deformation above a threshold stress

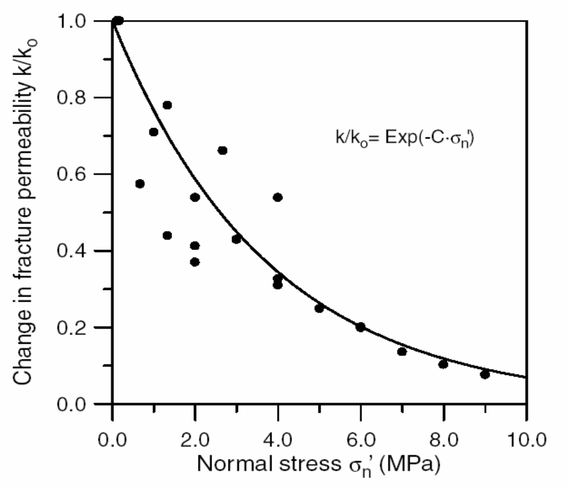


In the context of self-healing it is important to remember that any stress acting on the tunnel lining must be transmitted by the EDZ. If the rock exhibits creep then the EDZ will experience a time-dependent build up in the radial stress after installation of the support system. The magnitude of this build up will depend in part on the rigidity of the support. One possible effect of this increase in radial stress will be to cause the stress path of the rock within the EDZ to move progressively away from the failure (or yield) surface. The general movement of the stress path will be towards the "wet side" which favours contractancy over dilatancy.

Effect of normal stress

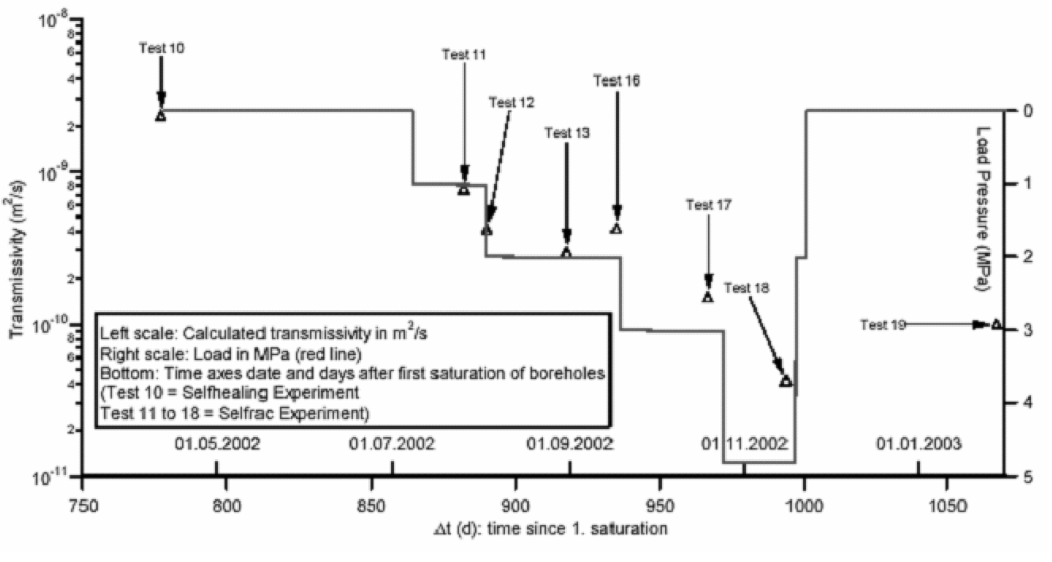
The hydro-mechanical behaviour of an extensional fracture in shale under normal and shear loading has been investigated experimentally by Gutierreza *et al.* (2000). The samples were obtained by de-mineralising cemented natural fractures in block samples of Kimmeridge Shale. These are believed to be representative analogues of hydraulic fractures in shaly formations. Increasing the normal stress across the fracture reduced the fracture permeability. This behaviour could be represented by an empirical exponential law (Figure 8). However, loading the sample to an effective normal stress twice equivalent to twice the intact rock unconfined compressive strength did not completely close the fracture. The fracture permeability was reduced by an order of magnitude after loading to an effective normal stress of 10 MPa. The measured fracture permeability was still eight orders of magnitude larger than the permeability of the intact shale.

Figure 8. Effect of normal stress on the permeability of a de-mineralised fracture in Kimmeridge Shale
(Gutierreza *et al.*, 2000)



Many of the extensile (unloading) fractures observed around openings in argillaceous rock are orientated more or less parallel to the tunnel surface. The radial component of stress around the tunnel therefore acts normal to many of these fractures. Re-application of normal stress to the EDZ should therefore lead to at least partial closure of EDZ fractures. In the SELFRAC Experiment at the Mont Terri Laboratory, the arrangements were made to reload the EDZ using hydraulic rams (Trick, 2003). Transmissivity measurements of were made before reloading and at various levels of applied stress. Figure 9 is a summary of the results. Loading the EDZ caused the transmissivity to drop from around 2 x 10^{-9} $m^2.s^{-1}$ at the start of loading to 4 x 10^{-11} $m^2.s^{-1}$ at an applied stress of 3.75 MPa. After complete removal of load, the transmissivity was 1 x 10^{-10} $m^2.s^{-1}$ showing that the effects of re-stressing the EDZ are largely irreversible.

Figure 9. Summarised results of the SELFRAC Experiment at Mont Terri Rock Laboratory showing the effect of reloading on the transmissivity of the EDZ



Viscoplastic deformation

Creep is time-dependent deformation of a material when subjected to sustained loading. In porous materials it is possible to distinguish two main types: (a) creep in shear governed by the deviatoric component of the stress tensor and (b) volumetric creep governed by the spherical component of the effective stress tensor. Consolidation of clay-rich materials is a time dependent change in volume and is considered to have a hydrodynamic component (primary consolidation) and a creep component (secondary consolidation). Most of the research in rock mechanics has focused on creep in shear. Since creep strains are largely non-recoverable on unloading, they are best described as viscoplastic strains. A variety of modelling approaches are available, including mechanical analogues assembled from basic rheological elements (e.g. the Kelvin model), empirical formulations and phenomenological approaches. Models are calibrated using laboratory data from multistage triaxial creep tests, relaxation tests or constant strain-rate tests or from field data by the back-analysis of monitored tunnel convergence or extensometer measurements. Possible scale effects have been observed in creep measurements on argillaceous rocks suggesting that over reliance on laboratory test data is inadvisable (Boidy *et al.*, 2002).

Fundamentally important issues in the specification of constitutive models for the creep are: (a) nonlinearity of the relationship between viscoplastic strain rate and deviatoric stress and the possibility of thresholds; (b) volumetric response of the material during creep deformation; (c) long-term extrapolation of creep test data and the existence or otherwise of steady-state creep under constant deviatoric stress. In contrast to evaporites such as rock salt, argillaceous rocks are only weakly nonlinear with a stress exponent only slightly greater than unity. Also in contrast to salt, argillaceous rocks can undergo substantial volumetric creep when mean effective stress is raised. Since creep models are seldom formulated in terms of effective stresses, volumetric creep behaviour is usually ignored. Coupling between shear and volumetric behaviour is likely in clay-rich rocks. Extrapolation of creep test data presents a significant problem and there are very few data sets of a quality or duration adequate to define long-term behaviour. It is necessary to compare numerical simulations with measurements obtained from monitored tunnels over long periods. Such data are very scarce.

Boidy *et al.,* (2002) present a back-analysis of the time-dependent behaviour of the reconnaissance gallery of the Mont Terri Tunnel in Switzerland. The study examined a specific section of the gallery which was excavated in Opalinus Clay and left without any support system for approximately 3 months, after which a stiff lining consisting of concrete rings was installed (Kohler, 1995; 1997). Numerical modelling was undertaken using the Lemaitre creep law. This is a total stress approach based on the assumption of isovolumetric viscoplastic behaviour. Pore pressure effects and volume-change phenomena such as consolidation and swelling are therefore ignored. Initial estimates of the parameters of the model were obtained from triaxial creep tests data supplied by the École Polytechnique Fédérale de Lausanne (EPFL) and reported in Kharchafi and Descoeudres (1995). Figure 10 shows the results of a typical laboratory test. Using the convergence measurements from the gallery, a parametric study was made on the Lemaitre parameters with particular attention to the viscosity term A. Figure 9 shows a comparison between the viscoplastic simulations and the measured convergence data. Parameter set no. 2 gives the best representation of the measured convergence. The viscosity term A is substantially smaller than that the value obtained by fitting the laboratory test results.

Figure 10. Lemaitre creep model fitted to undrained triaxial creep data for the Opalinus Clay
(test data from Kharchafi and Descoeudres, 1995; and data fits from Boidy *et al.*, 2002)

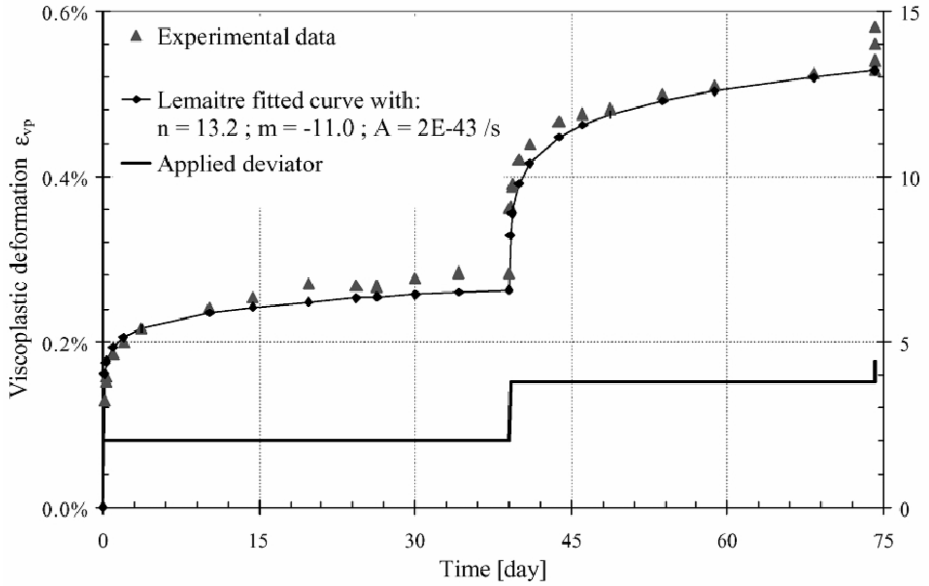
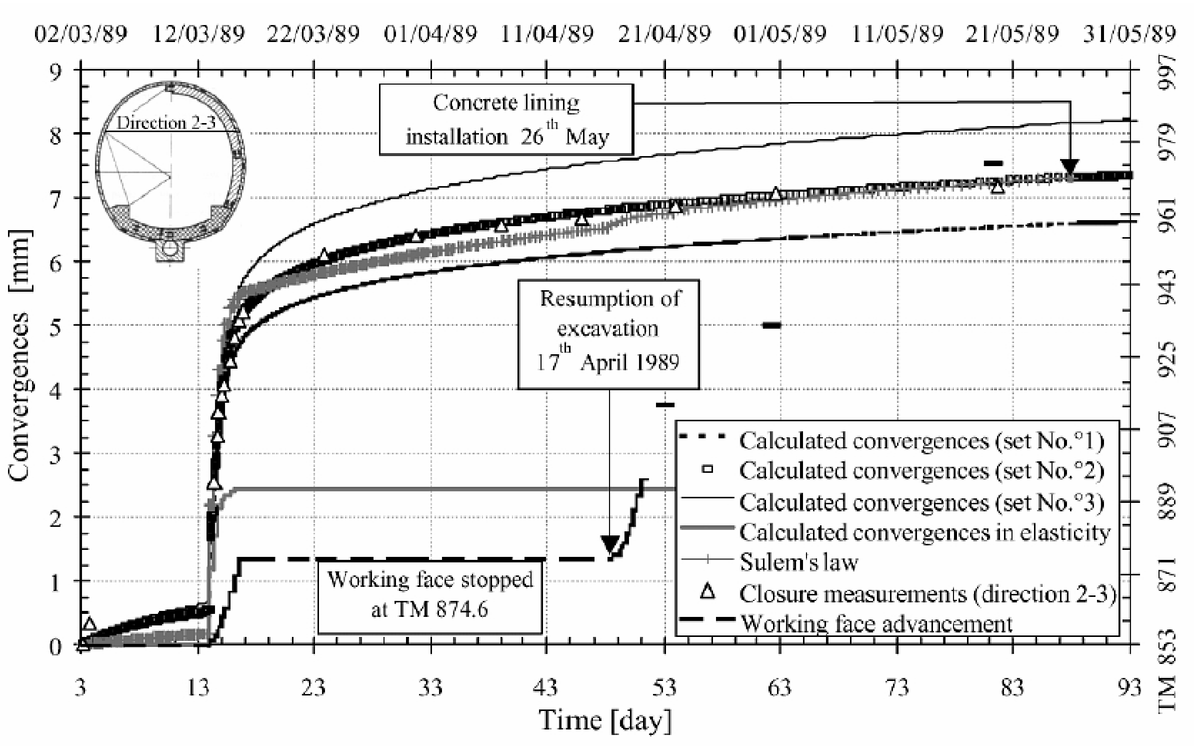
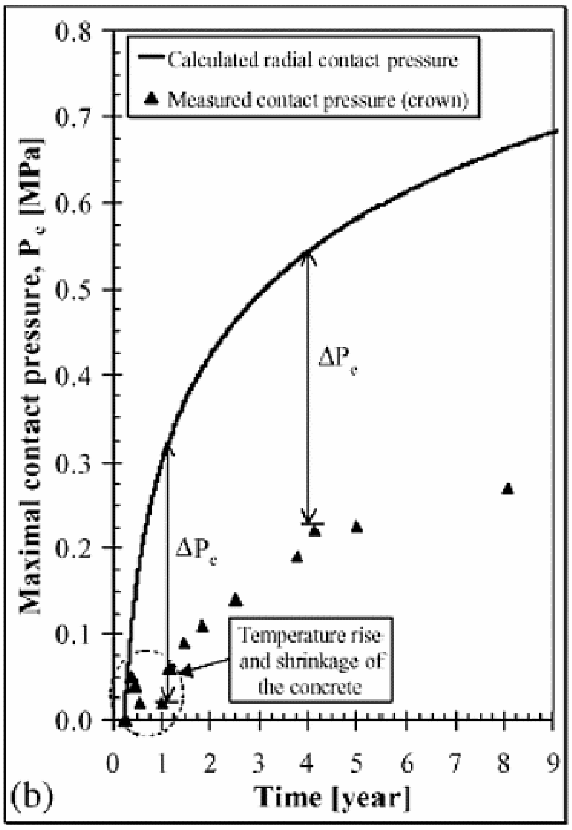


Figure 11. Measured and computed convergence for different sets of creep parameters
(measurements from Kohler, 1995; 1997; and modelling results from Boidy *et al.* 2002)



The build up of radial contact pressure is interesting from the point of view of EDZ self-healing. Figure 12 shows the measured and calculated response at the crown. The overburden stress is estimated to be 6.35 MPa. The actual contact pressure is around 0.27 MPa after 8 years showing that only a small proportion of the overburden stress is tranferred to the lining in this time period as a result of creep deformation of the formation.

Figure 12. Measured and computed build up of radial contact pressure on the concrete rings in the reconnaissance gallery at the Mont Terri Tunnel
(Boidy *et al.*, 2002)



In this respect the Opalinus Clay differs considerably from the Boom Clay. Measurements of the build-up of load on the concrete segmental lining of the connecting gallery in the Hades URL show that lining load reaches 3 to 3.5 MPa after a comparatively short period of time. The calculated vertical overburden stress is around 4.2 MPa, showing that a significant fraction of the total stress is transferred to the lining. The reapplication of this normal stress to the EDZ is likely to promote permeability decrease and self-sealing.

Swelling

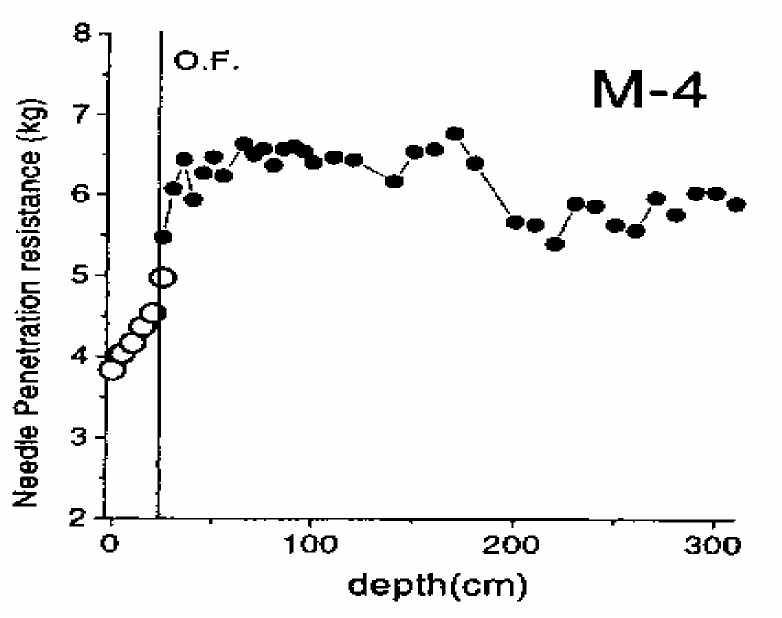
The capacity of an argillaceous rock to swell when exposed to free water depends on: (a) clay mineralogy; (b) cementation; (c) stress history and (d) water chemistry. Water is drawn into the clay fabric by physico-chemical forces and this inward movement of water molecules into the interparticle spaces between platy clay minerals causes the fabric to expand. The presence of an interparticle cement can inhibit swelling of the clay fabric. However, cementing agents such as calcite and silica are brittle and easily damaged by shear deformation and it seems likely that fabric damage is a necessary prerequisite for swelling in some of the more indurated argillaceous rocks. This form of fabric damage is sometimes referred to as bond degradation (Vaunat and Gens, 2003).

Softening

Many argillaceous rocks also exhibit a progressive process of fabric breakdown referred to as softening. This is a gradual alteration of a hard rock into a plastic soil-like clay material with substantially reduced shear strength (Botts, 1986). The water content of a softened argillaceous rock is significantly larger than that of the parent material showing that swelling and softening are closely linked. Softening usually commences around fissures and fractures. Figure 13 illustrates the process of softening around a tunnel.

Figure 13. Variation of needle penetrometer resistance with distance from a tunnel surface showing softening of the rock in the oxidised zone

(Oyama and Chigira, 1999)

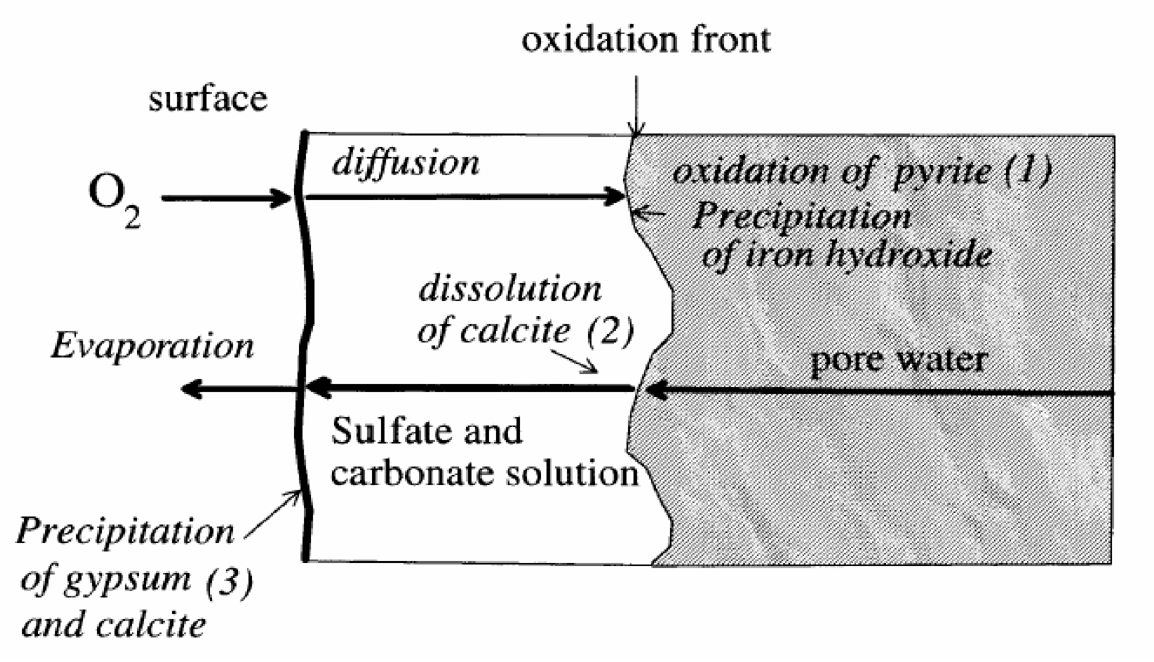


Chemical weathering

Geochemical processes play an important role in the progressive alteration of argillaceous rock in the walls of underground excavations. For example, the oxidation of pyrite can leading to acidification of the porewater. The sulphuric acid thus formed can react with calcite to form gypsum. This process is not exclusively chemical. Autotrophic bacteria obtain their energy from the oxidation of inorganic compounds and atmospheric oxygen is consumed. Proteinaceous body materials are produced from atmospheric CO_2 and nutrients such as nitrogen are usually available in pyritic shales to support bacterial growth and reproduction.

It was recently discovered that the unlined walls of road tunnels excavated 45-85 years ago in central Japan have maintained their original cut surfaces over several decades; offering suitable locations to study the long-term effects of chemical weathering on mudstones and tuffs (Oyama and Chigira, 1999). The tunnel walls are assumed to have been saturated with water before the excavation and to have become desaturated by natural ventilation. White gypsum crusts about 0.5-2 mm thick, together with powdery gypsum were found to have precipitated on the wall surfaces. Prior to tunnel excavation, the water-saturated mudrocks were in a reducing environment. After excavation, the tunnel walls became dry and desaturated by ventilation, and oxidation started from the walls and moved inward. Oxygen diffused from the surface walls to the interior through the interconnected pore-space of the rock (Figure 14). This resulted in oxidation of pyrite at the oxidation front, and the generation of sulphuric acid. FeO present in the clay minerals was oxidised. The sulphuric acid released by oxidation dissolved certain components of the rock, in particular carbonate in the mudstones and glass in the tuffaceous samples. As a consequence, rock present within the oxidised zone softened and weakened.

Figure 14. Mechanism of chemical weathering of the mudstone tunnel wall
(Oyama and Chigira, 1999)



Growth of gypsum crystals can also force shale layers apart (Figure 15). This process is similar to the formation of ice lenses in soils (Penner *et al.,* 1972). Oyama and Chigira (1999) suggest that diffusion of oxygen is the limiting factor in determining the rate of advancement of the weathering front into the rock (Figure 16). Isotopic studies at the Tournemire Tunnel in southern France indicate that oxygen isotopes are disturbed at more than 10 m indicating air diffusion around this old tunnel at a significant distance from the tunnel wall (Charpentier *et al.*, 2003).

Figure 15. The growth of gypsum crystals tends to force shale layers apart. This process is similar to the formation of ice lenses in soils.
(Penner *et al.*, 1972)

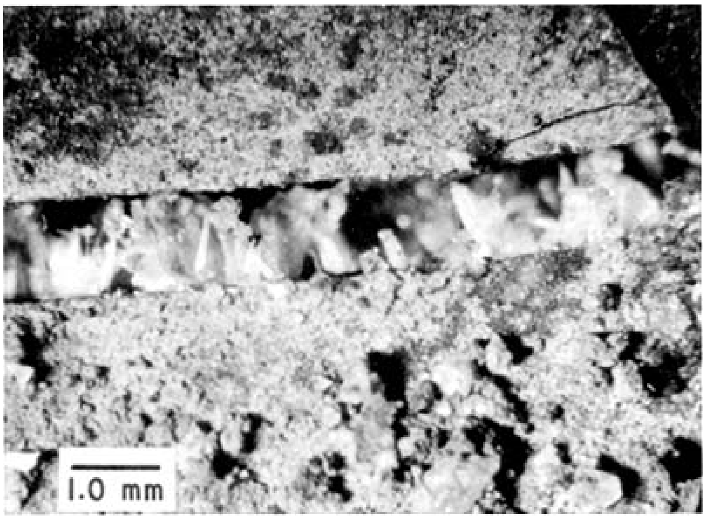
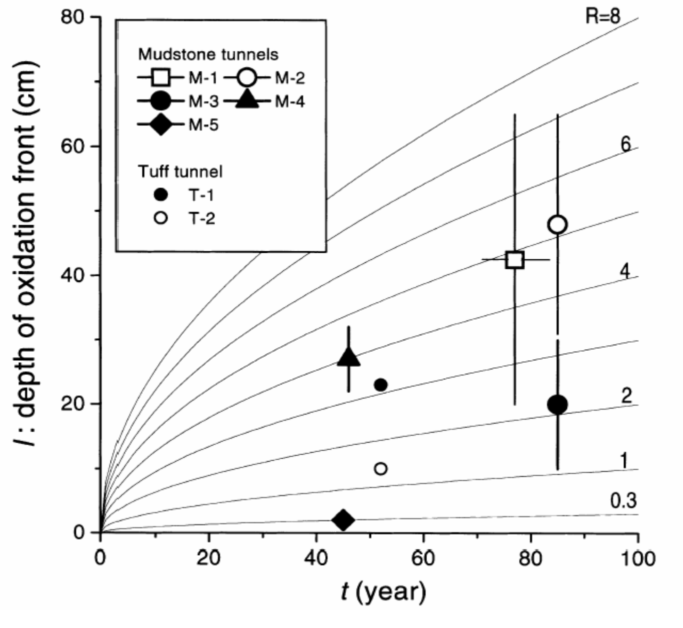


Figure 16. Advancement of the chemical weathering front into the tunnel wall. Error bars relate to various measurements of diffusivity.
(Oyama and Chigira, 1999)



It is conceivable that, under very specific circumstances, gypsum precipitation might contribute to a lowering of fracture conductivity. It is also possible that the chemical breakdown of interparticle cements could initiate the swelling and softening mechanisms discussed above. Geochemical changes might result in alteration of the retention capacity of the damaged rock.

Long-term geochemical evolution

The tunnel lining will impede the diffusion of oxygen into the wall rock. After backfilling of the excavations and closure of the repository any remaining residual air will eventually be consumed by chemical reactions and biological activity. The EDZ will then enter an anoxic phase. The long-term geochemical evolution of damaged rock and its effect on the transport properties of the EDZ remain to be elucidated. In the specific case of repositories containing large amounts of cementious materials, the precipitation of cementing minerals from a hyperalkaline plume is anticipated. Fracture sealing by this form of mineralisation has been observed at the Maqarin natural analogue site in Jordan (Alexander, 1995; Smellie, 1997).

Conclusions

The capacity of fractures in argillaceous rocks to self-heal (or become, with the passage of time, less conductive to groundwater) is often cited as a primary factor favouring the choice of such materials as host rocks for deep disposal. The underlying processes which contribute to self-healing can be broadly subdivided into: (a) mechanical and hydromechanical processes linked to the change in the stress field, movement of porewater, swelling, softening, plastic deformation and creep, and (b) geochemical processes linked to chemical alterations, transport in aqueous solution and the precipitation of minerals. Since chemical alteration can cause profound changes to the mechanical properties of argillaceous rocks, it is often difficult to draw a firm line between these two sub-divisions.

Provisions for made long-term support of repository excavations are of first order importance in assessing the capacity for EDZ self-healing. One approach to the design of a tunnel lining uses the so-called "Convergence-Confinement Method". This involves the calculation of a "ground response curve" and a "support response curve". The ground response curve is a plot of the convergence against a hypothetical pressure applied uniformly to the tunnel wall, known as the confinement. The ground response curve is calculated using a model for the strength and deformation of the rock. The argillaceous rocks of interest as potential host-rocks in radioactive waste disposal show a pronounced rate-dependency in their stress-strain behaviour. Creep deformation is very important in such rocks and the ground response curve therefore changes progressively over time. The stress acting on the lining is predicted to rise asymptotically with time up to some maximum limiting value.

If the rock exhibits creep deformation then the EDZ will experience a time-dependent build up in the radial stress after installation of the support system. The magnitude of this build up will depend in part on the rigidity of the support. Many of the extensile (unloading) fractures observed around openings in argillaceous rock are orientated more or less parallel to the tunnel surface. The radial component of stress around the tunnel therefore acts normal to many of these fractures. Re-application of normal stress to the EDZ should therefore lead to at least partial closure of EDZ fractures. The hydro-mechanical behaviour of an extensional fracture in shale under normal and shear loading has been investigated experimentally. In Kimmeridge Shale, loading to an effective normal stress twice equivalent to twice the intact rock unconfined compressive strength did not completely close the fracture. Fracture permeability was reduced by an order of magnitude after loading to an effective normal stress of 10 MPa.

Measurements of the build-up of load on the concrete segmental lining of the connecting gallery in the Hades URL show that lining load reaches 3 to 3.5 MPa after a comparatively short period of time. The calculated vertical overburden stress is around 4.2 MPa, showing that a significant fraction of the total stress is transferred to the lining in the Boom Clay. The reapplication of this normal stress to the EDZ is likely to promote permeability decrease and self-sealing.

It seems likely that the combined processes of chemical weathering, swelling and softening will lead to a gradual decrease in the amount of water flowing through interconnected fractures. The presence of an interparticle cement can inhibit swelling of the clay fabric. However, cementing agents such as calcite and silica are brittle and easily damaged by shear deformation and it seems likely that some form of mechanical damage or chemical alteration is a necessary prerequisite for swelling in some of the more indurated argillaceous rocks. It has been suggested that suggest that diffusion of oxygen is the limiting factor in determining the rate of advancement of the weathering front into the rock. It is conceivable that gypsum precipitation might contribute to a lowering of fracture conductivity.

After backfilling of the excavations and closure of the repository any remaining residual air will eventually be consumed by chemical reactions and biological activity. The EDZ will then enter an anoxic phase. The long-term geochemical evolution of the damaged rock and its effect on the transport properties of the EDZ remain to be elucidated.

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HYDRO-MECHANICAL ASPECTS: GLACIAL LOADING/EROSION – THE OPALINUS CLAY STUDY

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1. Introduction

In Nagra's high level waste programme the future geological evolution of the investigation area in the Zürcher Weinland (NE Switzerland) is considered over a time period of around 1 Ma. Uplift, erosion and climatic changes were identified as processes that may affect the long-term performance of a repository for spent fuel (SF), vitrified high-level waste (HLW) and long-lived intermediate-level waste (ILW). The possible impact of those long-term processes on the barrier function of the host rock formation (Opalinus Clay) comprises: (i) the change of the vertical hydraulic gradient caused by changing recharge/discharge conditions in the regional aquifer systems; (ii) permeability enhancement of the host rock formation due to the uplift process; and (iii) expulsion of contaminated porewater from the disposal area as a result of repeated glacial loading.

Due to the fact that the discharge level of the regional aquifer systems is defined by the Rhine valley and the regional recharge areas are characterised by moderate elevations (typically <200 m above the level of the Rhine valley) the present day *vertical hydraulic gradient* between the regional aquifers is <1; hydraulic gradients >2 are not expected within the next several million years. Hence, the topography of NE Switzerland together with the favourable hydraulic properties of the host rock formation prevent efficiently the vertical exchange of groundwater between the regional aquifer systems. Even on geological time scales of millions of years the expected vertical porewater flow through the host rock formation is in the order of 10^{-14} m/s and lower.

At present, the Opalinus Clay forms a perfect hydraulic barrier. The process of ongoing uplift, however, may give rise to embrittlement of the rock, entailed by *permeability enhancement*. Evidence for permeability enhancement of Opalinus Clay in shallow depth was observed by Hekel (1994). On the other hand, regional studies confirmed the low permeability of the Opalinus Clay formation in greater depth – even when fractured (Gautschi, 2001; Marschall *et al.,* 2003). Such evidence, together with conceptual considerations concerning the rock deformation behaviour (Horseman, 2002) leads to the conclusion that the onset of significant dilatancy phenomena in the host rock is restricted to burial depths <200 m (Nagra, 2002). Given the moderate uplift rates in the area of interest, permeability enhancement due to embrittlement of the Opalinus Clay is not expected within the next million years.

During glacial periods, ice cover and partial permafrost affect both, local and regional groundwater flow. An ice cover of several hundred metres thickness may impose a transient increase in effective stress in the host rock formation and *excite the expulsion of contaminated porewater* from the disposal systems. For such scenarios, mass fluxes and transport paths of the expulsed porewater were inferred by hydrodynamic modelling. This paper summarises key result of modelling, which are presented in greater detail in Nagra (2002).

2. Geological setting and long-term evolution of the investigation area

The Opalinus Clay was deposited some 180 million years ago in a shallow marine environment over a large area in northern Switzerland and southern Germany. In the Zürcher Weinland the Opalinus Clay is part of the sedimentary sequence of the Molasse basin with a uniform thickness of 80-120 m over many kilometres, almost flat-lying (dipping gently to the south east) and little affected by faulting. The burial and compaction history of the sedimentary formations is complex, including two major phases of subsidence during the Cretaceous and the Miocene. From about 10 million years ago, uplift processes brought the Opalinus Clay progressively up to its present burial depth of about 650 m in the region of interest. The Zürcher Weinland is characterised by a low uplift rate of about 100-200 m/Ma (Nagra, 2002).

At present, the Opalinus Clay is a slightly overconsolidated claystone formation with an overconsolidation ratio between 1.5 and 2.5. Its hydraulic conductivity is very low-recommended values for horizontal and vertical hydraulic conductivity are 2×10^{-14} and 1×10^{-13} m/s, respectively (Nagra, 2002). The major principal stress at Benken is approximately horizontal (S_H=22.6 MPa, S_h=15.1 MPa, S_v=15.9 MPa at reference depth 650 m).

Several glaciations are expected to take place during the next 1 Ma (Nagra, 2002). The typical duration of the glacial periods is 10 000-50 000 a, followed by interglacial periods of roughly the same duration. During the glacial periods an ice cover of 200-400 m is assumed to increase vertical stress within the Opalinus Clay by 2-4 MPa.

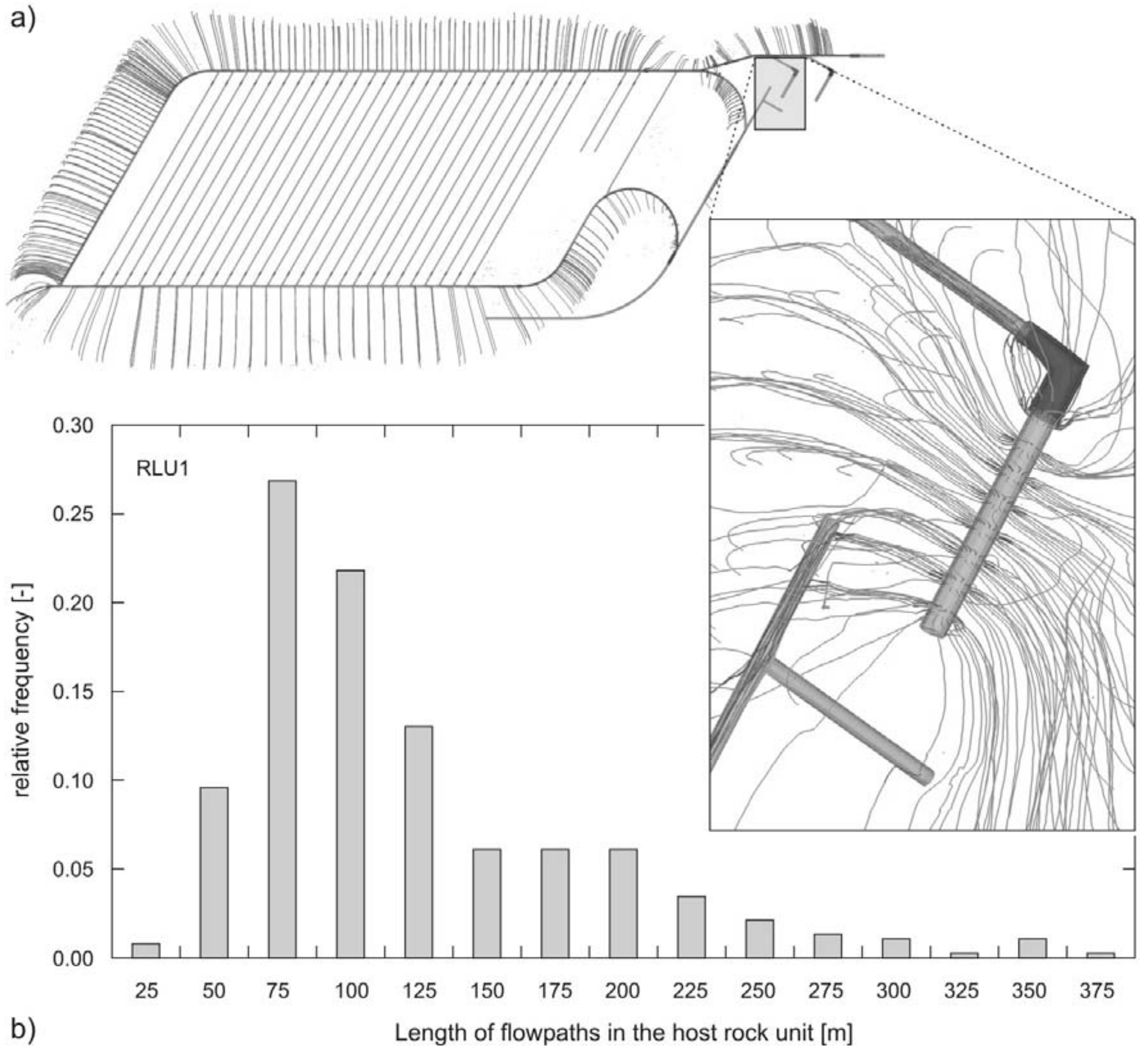
3. Results of hydrodynamic modelling

The emphasis of hydrodynamic modelling as part of the geoscientific site characterisation process has been on the understanding of both the natural, undisturbed groundwater flow in the investigation area as well as the repository-induced perturbations. The undisturbed hydrogeological system was simulated in a local scale model with a lateral extension of 10×10 km (Kuhlmann *et al.,* 2002). The repository-induced perturbations were assessed with a repository model (Kuhlmann & Marschall, 2002), which was embedded in the local model. Among the repository-induced perturbations the overpressurisation of the disposal systems (SF/HLW and ILW), entailed by the expulsion of porewater formed the most important modelling cases. Overpressurisation could be caused by the generation of repository gas or by an increase of vertical stress as a consequence of a glacial ice load. The assumed upper limits for gas-induced and ice load-induced overpressures in the disposal areas are 7.5 and 4 MPa, respectively. It is further assumed that the maximum glacial ice-load would act for a time period of about 20 000 a.

Simulations of the undisturbed hydrogeological system (local model) indicate an upwards directed vertical flow of 2×10^{-14} m/s through the host rock. Consideration of the backfilled underground excavations (repository model) does not significantly change porewater flow through the host rock formation – the flow direction is still vertical and the length of the flow paths from the disposal area through the host rock to the adjacent formation (Wedelsandstein) is about 40 m; the flow magnitude is indistinguishable with respect to the undisturbed conditions. Porewater overpressures in the disposal areas, however, give rise to marked changes of flow conditions in the vicinity of the backfilled underground excavations. As seen in Figure 1 the flow paths are no more vertical but diverge radially into the host rock formation. Furthermore, for the ILW disposal tunnels the length of the flow paths through the host rock increases substantially (histogram in Figure 1). Porewater flow into the Wedelsandstein formation is enhanced: for the case of unlimited overpressures of 7.5 MPa, the flow into the Wedelsandstein formation is 2×10^{-13} m/s and for the modelling run with overpressures of 4 MPa and limited duration (20 000 a) maximum flow rates of 1×10^{-13} m/s were determined.

As shown in Nagra (2002), diffusion is the dominating transport process in the host rock formation, when the advective flow component is in the order of 10^{-13} m/s or lower. Therefore, it is concluded that the ice load during glacial periods does not substantially affect the barrier function of the host rock formation. The unloading process during the interglacial periods may give rise to sub-hydrostatic pressures in the Opalinus Clay, resulting in a reversal of the hydraulic gradient towards the repository.

Figure 1. Expulsion of porewater from the disposal areas of an SF/HLW/ILW repository that may be caused by glacial loading (Kuhlmann & Marschall 2003)
(a) flow patterns and (b) length of flow paths in the host rock formation (ILW disposal tunnel)



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GEOCHEMICAL STABILITY OF CLAY-RICH ROCK FORMATIONS: EVIDENCE BASED ON NATURAL TRACER PROFILES

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1. Introduction

In comparison to many other rock types, clay-rich formations have high water contents and porosities. However, hydraulic conductivities measured on core samples and in boreholes are mostly very low, which is due to the extremely small apertures of individual pores (predominantly in the range of nanometers). By consequence, specific mineral-surface areas with which water molecules and solutes interact are very large. Knowledge of the microscopic characteristics of the pore space is the key to the understanding of several macroscopic properties, such as the commonly observed insignificance of advection and the large chemical buffering capacity.

Given the generally long residence times, the chemical composition of pore waters in clay-rich formations is controlled by equilibria with mineral phases, such as carbonates and silicates, and ion-exchange reactions with clay minerals (Beaucaire *et al.*, this volume). The generally reducing conditions are strongly buffered by pyrite, siderite and/or organic matter.

Some constituents of the pore water are conservative, i.e. not buffered by minerals, and can move in the interconnected pore space. These constituents include:

- the H_2O molecules of the pore water;
- halogens (e.g. Cl, Br); and
- noble gases (e.g. He, Ne, Ar).

H_2O of the pore water and dissolved noble gases can move in the entire physical porosity of a rock, provided the pore space is interconnected. The movement of water molecules can be monitored by using their stable isotopic signatures, expressed as $\delta^{18}O$ and $\delta^{2}H$. In clay-rich formations, halogens can only move in a fraction of the physical porosity due to anion exclusion effects.

The conservative constituents can be used as tracers of migration processes in clay-rich formations, such as diffusive exchange with the embedding aquifers and advection across the formation. They are archives of migration processes that occurred in the formation over geological time scales in the past. If these processes are understood quantitatively and if the evolving geological boundary conditions can be constrained, predictions of future evolution can be made.

2. Available data base and interpretation

The geochemistry of several clay-rich formations has been investigated in the recent past, and a selection is listed in Table 1. These formations were subjected to different degrees of burial and induration, reflected by the wide range of water-accessible porosity of 8-40%. Based on laboratory measurements, pore-diffusion coefficients for water in these formations are within the narrow range 8-20*10^{-11} m^2/s (direction normal to bedding), whereas for anions (Cl, I), they are smaller and scatter more widely (0.5-13*10^{-11} m^2/s).

All formations considered are marine sediments, but their current pore-water salinity is generally lower than that of sea water. Some of the formations lost most of their marine signature, others preserved it at least in the central parts of the formation. Curved profiles of tracers in pore waters, such as $\delta^{18}O$, $\delta^{2}H$, Cl, I, Br and He, were observed in several formations. Model calculations indicate that many of the observed profiles are consistent with diffusive exchange between the clay-rich formation and the surrounding aquifers, whereas advective transport in the vertical direction must be very limited or absent.

In addition to the identification of the relevant transport process, model calculations can also provide constraints on diffusion times and/or on large-scale diffusion coefficients. The most difficult part of such calculations is the definition of initial and boundary conditions, because this requires knowledge on the paleohydrogeological evolution of the clay formation and the surrounding aquifers. Available results indicate that model calculations based on laboratory-derived diffusion coefficients are consistent with the hydrogeological evolution constrained by independent evidence, suggesting that the laboratory data are applicable to the whole formation. Some cases where this is not so, or where the constraints are not sufficient, need further evaluation.

Table 1. Selection of sites where data of natural tracers are available

Formation	Site	Country	Reference
Callovo-Oxfordian	Bure (Meuse/Haute Marne)	France	Andra (1999a)
Toarcian-Domerian	Tournemire	France	Boisson *et al.* (1998), Cabrera *et al.* (2001)
Couche silteuse	Marcoule/Gard	France	Andra (1999b)
Boom Clay	Mol	Belgium	Ondraf/Niras (2002)
Opalinus Clay	Benken	Switzerland	Nagra (2002), Gimmi & Waber (2003)
Opalinus Clay	Mont Terri	Switzerland	Pearson *et al.* (2003)

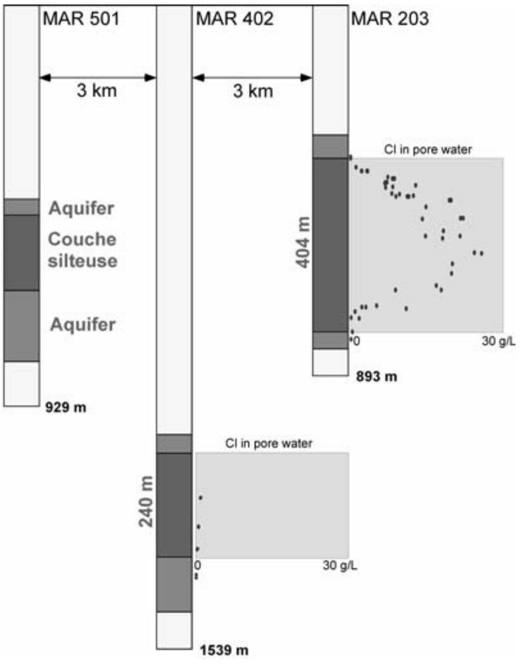
3. Example 1: Couche silteuse at Marcoule, France

The Marcoule site lies in SE France and was investigated by Andra (1999b). The target formation, the Couche silteuse, is embedded between two regional aquifers. Within a small horizontal distance (few km), three boreholes penetrating the whole sequence have been drilled, and in two of them, Cl contents in pore water have been analysed in vertical profiles (Figure 1). The interesting point is that over the short distance of 3 km, the thickness of the formation varies substantially between 150 and 400 m. In borehole MAR 203, the Cl contents show a regular, symmetric profile, with Cl contents close to the marine value in the centre of the formation but strong decrease towards the boundaries.[1] In borehole MAR 402, where the formation is only 240 m thick, Cl contents are very low throughout the profile, even though still higher in the centre than at the lower boundary. The question is whether the Cl distributions in both boreholes can be explained as diffusion profiles using the same dataset (formation properties, initial and boundary conditions), the only difference being the variable formation thickness. A simple analytical solution for time-invariant boundary conditions (Carslaw & Jaeger, 1959) was used for the calculations:

$$C(x,t) = C_0 + \left(C_1 - C_0\right)\left((1 - \frac{4}{\pi}\sum_{n=0}^{\infty} \left[\frac{(-1)^n}{2n+1} \exp\left\{ \frac{-Dp\,(2n+1)^2 \pi^2 t}{4L^2} \right\} \cos\left\{ \frac{(2n+1)\pi x}{2L} \right\} \right] \right)$$

with L = half thickness of the clay layer, [m];
Dp = pore-diffusion coefficient for Cl, [m s^{-2}];
C_0 = initial tracer concentration in the clay layer, [mg L^{-1}];
C_1 = tracer concentration at the boundaries, [mg L^{-1}];
C(x,t) = tracer concentration at time t and location x in the clay layer, [mg L^{-1}];
t = diffusion time, [s];
x = spatial coordinate (-L < x < L), [m].

Figure 1. Overview of borehole data from the Marcoule site (Gard, SE France) (Andra, 1999b)



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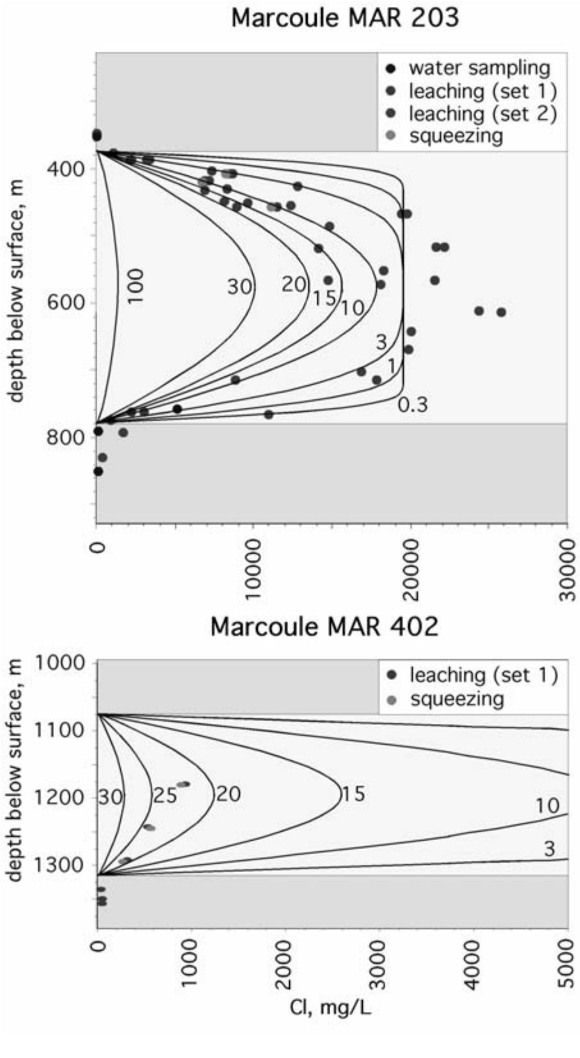
[1] Some values lie above the sea-water value and are most likely related to data uncertainties, for example in the knowledge of Cl-accessible porosity. The original data set in Andra (1999b) contains Cl concentrations mostly based on aqueous leaching experiments, and the conversion of concentrations per volume of rock to aqueous concentrations was done using porosity values measured by conventional techniques such as water loss upon heating. These techniques characterise the porosity accessible to water, while Cl-accessible (geochemical) porosity is smaller due to anion exclusion effects (Pearson, 1999). A limited number of Cl concentrations based on porewater squeezing are also given in Andra (1999b) and yield higher values. In the present paper, the leaching data were corrected in order to be consistent with the squeezing data, which are considered to best represent in situ concentrations in the Cl-accessible porosity.

Boundary conditions were assumed to be represented by the present-day Cl contents of the embedding aquifers. The marine Cl concentration of 19 350 mg/L was taken as the initial condition. Pore-diffusion coefficients Dp were derived from laboratory measurements of De of I (Andra, 1999b), recalculated to Dp using the estimated geochemical porosity and corrected for in situ temperature.

The results of the calculations are shown in Figure 2. Diffusion times that best fit the tracer distribution in borehole MAR 203 are in the range of 3-15 Ma, compared to 20-25 Ma for borehole MAR 402. These values are similar, considering the uncertainties related to the incomplete dataset for borehole MAR 402 and other data uncertainties (e.g. geochemical porosity, spatial heterogeneity). It is concluded that diffusion alone can at least roughly explain the very different tracer concentrations in both boreholes. Further studies are needed to evaluate possible 2- or 3-D effects, i.e. horizontal diffusion within the formation between the two sampling localities, and the (in)compatibility of the observed Cl concentrations with alternative conceptual models considering other processes.

Figure 2. Measured (circles) and calculated (lines) Cl contents of porewater in the Couche silteuse in boreholes MAR 203 and MAR 402 at Marcoule (Gard, SE France)

Measured data are recalculated values from Andra (1999b)
and refer to Cl concentrations in the Cl-accessible porosity



4. Example 2: Opalinus Clay at Mont Terri, Switzerland

A profile of Cl contents (and several other tracers) in pore water was acquired from boreholes drilled from the Mont Terri rock laboratory tunnel (Swiss Jura Mountains), which penetrates Opalinus Clay in an anticlinal structure (Pearson *et al.,* 2003). As shown in Figure 3, the anticline has been partially eroded since the end of the major thrusting/folding events at ca. 3.5 Ma b.p. According to a preliminary and simplified erosion scenario, the limestone aquifer overlying the Opalinus Clay was exhumed ca. 2.5 Ma b.p., while the underlying aquifer was exposed on the surface only at later times, ca. 0.35 Ma b.p. Thus in the case of Mont Terri, there is independent evidence that the onset of flushing with meteoric water occurred at different times at the two boundaries.

The distribution of Cl in pore water across the Opalinus Clay is shown in Figure 4. The striking feature is the asymmetry of the profile, with a smoother gradient towards the upper aquifer and a steeper gradient downwards. Calculations considering diffusive exchange of Cl with the aquifers were performed separately for both aquifers, and the results are also shown in Figure 4. These calculations used Dp = 3.4E-11 m^2/s, a value based on laboratory-derived De values for Cl (Nagra, 2002; Table 5.10-1), a geochemical porosity based on Pearson *et al.* (2003, Table A10.3), and a correction for in situ temperature. The best-fit diffusion time for the onset of exchange with the upper aquifer is ca. 5 Ma, the one for the lower aquifer ca. 0.3-1 Ma. The shapes of the measured and the calculated Cl distributions are remarkably consistent, while the calculated diffusion times exceed those derived from erosion scenarios by about a factor 2. Potential explanations can be the choice of the diffusion coefficient for the calculations or, more likely, the assumptions regarding the erosion scenario.

Figure 3. Cross-section through the Mont Terri anticline (Jura Mountains, Switzerland). Stippled lines and ages indicate a rough estimation of the erosion history of the anticline (Pearson *et al.* 2003)

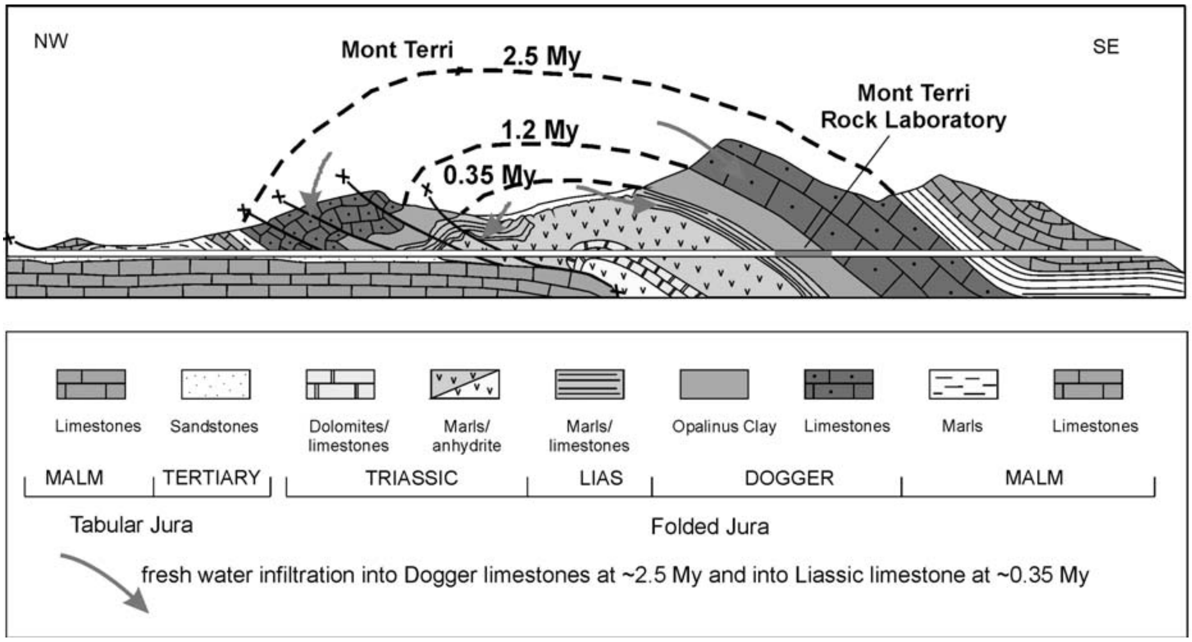
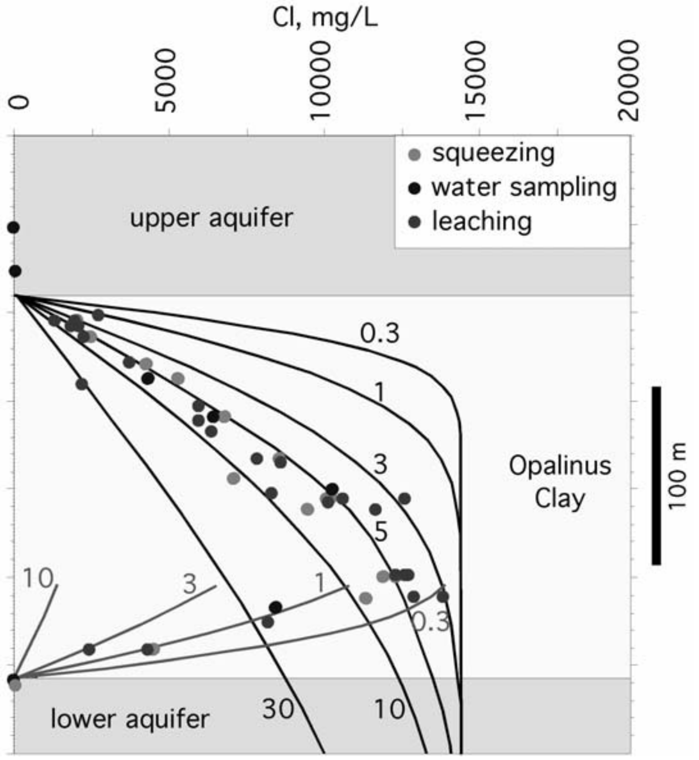


Figure 4. Measured (circles) and calculated (lines) Cl contents of porewater in the Opalinus Clay at Mont Terri (Jura Mountains, Switzerland)



5. Conclusions

In the examples above, only Cl data were discussed, even though data on other tracers are available and should be investigated in a next phase. The simultaneous consideration of all available data increases the confidence in the interpretation.

Diffusion profiles may develop in relatively thin clay-rich units over time scales that are short compared to the age of the formation (order of 0.1-1 Ma for a 100 m thick layer), provided marked chemical gradients to the aquifers are established. Very long diffusion times (tens of Ma or more) reported in the literature are not independent model results in most cases but a priori assumptions, often combined with a poorly constrained hydrogeological evolution. Overall, the study of natural tracer profiles is an important contribution to the understanding of the long-term evolution of clay-rich formations because such profiles represent long-term and large-scale natural experiments that can be used for the upscaling of laboratory data in space and time. Natural tracer profiles can contribute to the identification of the dominant transport process, to the quantification of maximum advective fluxes that are compatible with the spatial distribution of tracers. Tracer profiles crossing faults can provide additional information concerning paleofluid flow events along preferential pathways.

When the past evolution of the observed tracer profile is quantitatively understood, quantitative predictions are possible regarding the further evolution (e.g. loss of salinity to the embedding aquifer). Alternative scenarios for future development can also be explored by varying the boundary conditions.

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CHEMICAL BUFFERING CAPACITY OF CLAY ROCK

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1. Introduction

The long-term performance of a nuclear waste repository is strongly dependent on the chemical properties of the host rock. The host rock establishes the chemical environment that determines such important performance attributes as radionuclide solubilities from the waste and the transport rates from the repository to the accessible environment.

Clay-rich rocks are especially favourable host rocks because they provide a strong buffering capacity to resist chemical changes prompted either internally, by reactions of the waste itself and emplacement materials, or externally, by changes in the hydrologic systems surrounding the host rock.

This paper will focus on three aspects of the stability of clay-rich host rocks: their ability to provide pCO_2 and redox buffering, and to resist chemical changes imposed by changes in regional hydrology and hydrochemistry.

2. CO_2 buffering

It is well known that there are multiple sources for CO_2 in ground water. However, it has been demonstrated that there is a relationship between pCO_2 and temperature of formation waters in sedimentary rocks indicating a possible control of CO_2 by a mineral assemblage (Hutcheon *et al.,* 1993). This relationship seems to be independent of the sources of CO_2 and cannot exist in an open system.

A geochemical system can be defined involving a large number of constituents (H, O, C, Al, Si, Ca, Mg, K, Na) which are in equilibrium simultaneously and are representative of the local conditions in the aquifer. Mass transfer of chemical elements and the evolution of the rock porosity can be calculated using this approach to local equilibrium, as shown by Gouze and Coudrain-Ribstein (2002). In this study, a compilation of ground waters in sedimentary rocks showed that over a temperature range from 40°C to more than 200°C, pCO_2 appears to be controlled by equilibrium of the solution with calcite, dolomite and a set of hypothetical silicates including chlorite. For the chlorite-kaolinite-chalcedony-calcite-dolomite assemblage (Hutcheon *et al.*, 1993), pCO_2 can be calculated using the following relation:

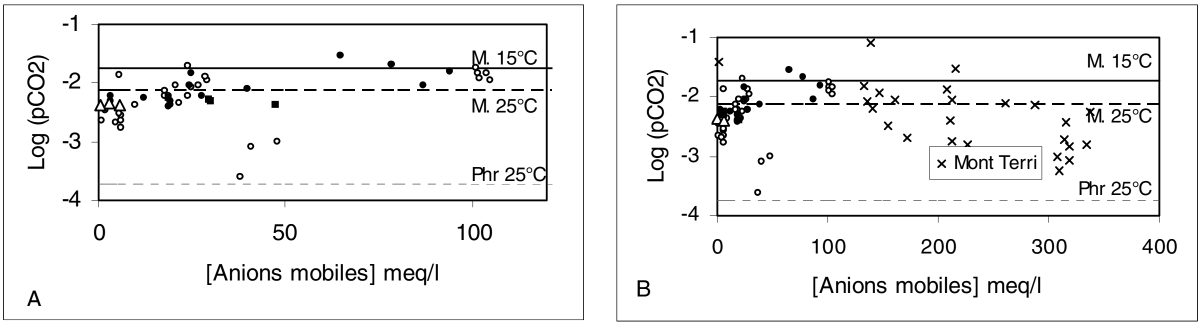
$$pCO_2 = [H_2O]_{2/5} \frac{K_{calcite} \times K_{1/5chlorite}}{K_{CO_2(g)} \times K_{1/5chalcedony} \times K_{1/5kaolinite} \times K_{dolomite}}, \qquad (1)$$

where K corresponds to the formation constants of the different minerals and CO_2.

This relation implies that some mineral assemblages can buffer CO_2 concentrations against additional CO_2 produced by bio-degradation or thermal degradation of organic matter, for example. In scenarios of waste disposal in geological formations, CO_2 production may be expected during the thermal phase. Equation (1) indicates that CO_2 concentrations can be predicted with changing temperatures regardless of the amounts of CO_2 added to the system by thermal reactions. To be generalised, the applicability of Equation (1) must be confirmed and it must be demonstrated that the masses of the reactive minerals included in Equation (1) are great enough to provide a buffer capacity sufficient to accommodate all CO_2 produced. This will be site specific, but in general the clay-rich rocks of interest here contain enough of these minerals to buffer large quantities of CO_2.

The observations supporting Equation (1) extend to temperatures as low as 40°C. While this is close to the ambient temperatures of certain proposed repositories (*e.g.* 38°C for Benken – Zürcher Weinland, Nagra, 2002a), it is also important to decide if such equilibria can be achieved in clay rocks at even lower temperatures and therefore can actually be observed in lower temperature, natural systems.

Figure 1. Evolution of Log (pCO_2) vs the sum of mobile anions in different sedimentary formations
A: Mol: interstitial fluids (triangles) and aquifers (empty and filled circles); Tournemire (squares);
B: Mol, Tournemire and Mont-Terri (crosses). Lines represent equilibrium with respect calcite-dolomite-chalcedony-chlorite-kaolinite, at 15 and 25°C for two formations constant of chlorite
(Phr: PHREEQC data; M: Michard's data)



In support of this conclusion, we summarise water chemical data obtained from several clay rocks in different sedimentary contexts: interstitial waters from the Boom Clay (Beaucaire *et al.*, 2000), ground waters from the aquifers surrounding the Boom clay (Marivoet *et al.*, 2000), fracture fluids from Tournemire (Cabrera *et al.,* 2000; Beaucaire *et al.*, in prep.) and interstitial waters from Mont-Terri drill-holes (Pearson *et al.* 2003). All these formations are characterised by a predominance of clay minerals in the rock and a low matrix permeability. pCO_2 values calculated from pH and alkalinity measurements for the different ground waters are plotted on Figure 1.

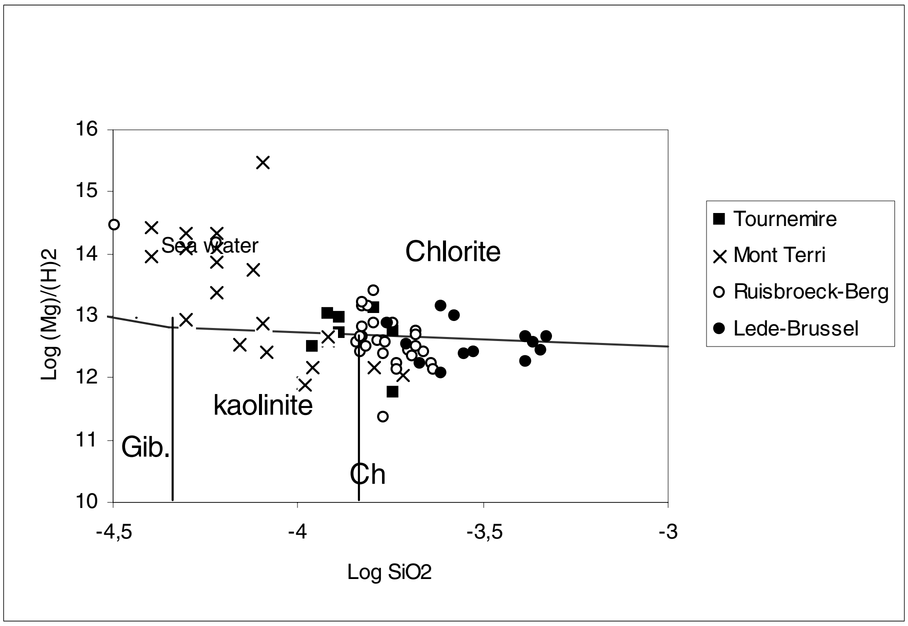
The same figure shows pCO_2 values calculated according to Equation (1) at 15 and 25°C and for two values of the chlorite formation constant. From the different chlorite dissolution constants in the literature, we have chosen two limiting values of log K, 16.16 and 23.98, from Michard and the PHREEQC data base (Parkhurst and Appelo, 1999), respectively. In these conditions, it appears that the range of variation of log (pCO_2) does not exceed a factor of 2.0 and that all the studied waters fall within this range. In order to fully validate the model, it is necessary to verify that solutions are equilibrated with respect to the involved mineral phases.

The thermodynamic behaviour of clay minerals is a continuous topic of discussion regarding the question of whether a solution can be equilibrated with respect to clay minerals. Recent laboratory studies of the dissolution of chlorite affirm that it is possible to describe the evolution of solutions at equilibrium with the kaolinite-chlorite assemblage from 25 to 175°C (Aja *et al.,* 2002). If it is the case, natural solutions must also show a relationship between $\log(Mg^{++})/(H^{+})^{2}$ and $\log(H_4SiO_4)$ corresponding to the equilibrium of the solution with the kaolinite-chlorite assemblage according to the schematic reaction:

$$\underset{\text{chlorite}}{Mg_5Al_2Si_3O_{10}(OH)_8} + 10H^{+} + 2H_2O = 5Mg^{++} + \underset{\text{kaolinite.}}{2Al_2Si_2O_5(OH)_4^{-}} + H_4SiO_4$$

Figure 2, is a plot of the data corresponding to samples from the Ruisbroeck-Berg and Lede-Brussel aquifers associated with the Boom Clay, from Boom Clay and Mont Terri interstitial waters and from Tournemire fracture fluids. The majority of samples except some of the Mont Terri fluids are close to a horizontal line limiting kaolinite and chlorite fields.

Figure 2. Isothermal (15°C) ion activity diagrams showing the variation of aqueous Mg and Si species in solution. Comparison with stability of kaolinite-chlorite assemblage. Ch: chalcedony; Gib: gibbsite



Samples from Ruisbroeck Berg aquifer, Tournemire and Boom Clay fluids can be considered as equilibrated with respect to chlorite, kaolinite and chalcedony, while Lede-Brussel fluids evolve toward silica enriched fluids. This silica enrichment was attributed to the occurrence of opal in this sedimentary horizon. It is also important to indicate that the temperatures of the studied waters are ranging from 13 to 25°C.

Mont Terri fluids present have the most variable silica concentrations and $\log(Mg^{++})/(H^{+})^{2}$ values ranging from 12.7 for the less concentrated ground waters to 14.5 for the more concentrated waters. The latter waters maintain the characteristics of the sea water originally present in the formation while the more "diluted" waters of the series seem to have evolved towards equilibrium with the mineral assemblage.

In conclusion, the residence time of interstitial waters and the very low permeability of clay rocks are in favour of the establishment of chemical equilibria. Many observations in sedimentary aquifers at low temperature and in interstitial waters within clay rocks seem to indicate that the behaviour of CO_2 continues to be constrained: pCO_2 in such formations generally ranges between 10^{-2} and $10^{-3.5}$ atm. However, the validation of such an approach must be supported by further mineralogical and thermodynamic studies: observation of the involved mineral phases and their chemical variability in the different natural contexts, and therefore evaluation of their relative stability.

Other conceptual models of controls on pCO_2 are also used. pCO_2 values in the Opalinus clay at Mont Terri and in the Callovo-Oxfordian have been modelled considering equilibria among cation-exchange reactions and carbonate minerals (Gaucher *et al.*, 2000; Motellier *et al.*, 2003, Pearson *et al.*, 2003). In this model, the predictability of CO_2 in interstitial fluids depends on the stability of the chemical properties of the clayey-exchanger with time, which is itself function of temperature, rate of diffusion of the species in the formation and other all chemical reactions induced by the presence of nuclear waste (pH perturbation, thermal or bio- degradation of organic matter, …). However, in the long term, we could expect CO_2 buffering by silicate-carbonate mineralogical assemblages.

Finally, the calculated chemical buffer capacity will differ depending on whether one considers the CO_2 regulation in interstitial fluids to be by mineral buffering, exchange reactions or external constraints. Thus, it will be important in the future to evaluate the buffer capacities of systems by all possible models.

3. Redox buffering

The redox buffering capacity of a clay rock can be illustrated by data from the Mont Terri Underground Research Laboratory. Geochemical studies at Mont Terri have been summarised by Pearson *et al.* (2003). The following discussion is based on that report and references below to Section, Figure and Table numbers refer to that report, unless otherwise indicated.

Chemical analyses of several samples of Opalinus Clay and adjacent claystones at Mont Terri show that the major redox-active elements present are C, Fe, Mn and S (Section 5.2.3.4). The formation mineralogy is such that Fe(II) would be present in pyrite and carbonates, Mn(II) in carbonates, Fe(III) in silicates, principally chlorite, and S both in SO_4^{2-} and reduced sulphur minerals, principally celestite and pyrite. Carbonate and silicate minerals are dominant in the Opalinus Clay. Celestite is present in veins in undisturbed Opalinus Clay samples and in the bulk formation as well, but only at trace levels. The formation also contains more than one percent pyrite which accounts for virtually the entire analysed S content of the formation.

The redox buffer capacity of the Opalinus Clay depends principally on the SO_4^{2-} content of the system to buffer reducing tendencies and on the reduced S, Fe(II) and reduced C contents to buffer oxidising tendencies. The water itself has virtually no capacity to resist oxidation because it has only vanishingly small concentrations of reduced substances. On the other hand, the Opalinus Clay itself is strongly resistant to oxidation.

The total reduction capacity (TRC) of a formation is a measure of its resistance to oxidation. It can be measured directly using a strong oxidising agent such as permanganate or dichromate or calculated from the concentrations of reduced materials. Calculated TRC values of Opalinus Clay samples range from 2.0 to 4.7 meq/g, in good agreement with a measured value of 4.9 meq/g (Table A9.7). Half or more of these TRC values represents the contribution of organic C in the formation.

Mäder (2002) gives a detailed description of the oxidation of Opalinus Clay. This is based on petrographic and chemical studies of samples exposed to oxidising conditions for periods from days, in the laboratory, to years, centuries and millennia, in the Mont Terri and railroad tunnels and in exposures in quarries and outcrops. From his observations, Mäder concludes that during periods of up to at least a few hundred years, pyrite oxidation will dominate according to the generalised overall reaction:

$$\mathrm{FeS_2} + \frac{15}{4}\mathrm{O_2} + \frac{15}{2}\mathrm{H_2O} + 2\mathrm{CaCO_3} \Leftrightarrow \mathrm{Fe(OH)_3} + 2\mathrm{CaSO_4}\cdot 2\mathrm{H_2O} + 2\mathrm{H_2CO_3} \qquad (2)$$

In addition, birnessite is observed associated with pyrite oxidation (Mazurek *et al.* 1996), indicating that Mn(II) oxidation also occurs. Although Mäder does not report any observations suggesting the oxidation of Fe(II) in siderite nor of organic C, Mazurek *et al. op. cit.* report siderite dissolution with oxidation to Fe(III) to form Fe-hydroxides in oxidation rims around fractures in an open clay pit.

Mäder concludes that pyrite oxidation will dominate the redox behaviour of the Opalinus Clay for periods of the order of hundreds to perhaps thousands of years. However, at full equilibrium, as might occur during prolonged exposure to O_2, additional oxidation of the organic C and siderite could also occur. The organic C of the Opalinus Clay is refractory and not easily oxidised. However, Petsch *et al.* (2001) have recently shown that even highly refractory organic carbon, such as the kerogen found in Palaeozoic shales, can be used as a substrate for growth and thereby oxidised by certain microbes during prolonged weathering. In addition, work in progress at Mont Terri (PC Experiment) shows that bacterially mediated sulphate reduction coupled to organic C oxidation takes place in anaerobic boreholes in the Opalinus Clay.

Figure 3. Masses of minerals, in mol per kg porewater, and pe for Opalinus Clay as O_2 is added to system. System is at equilibrium with calcite, dolomite and $pCO_2 = 10^{-3.5}$

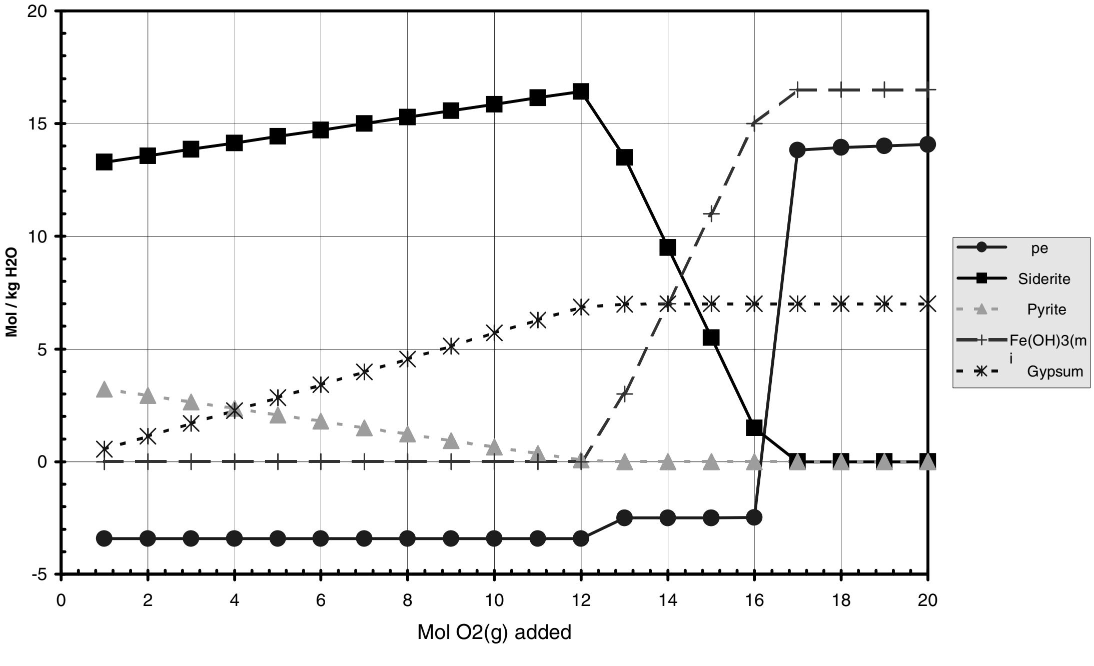
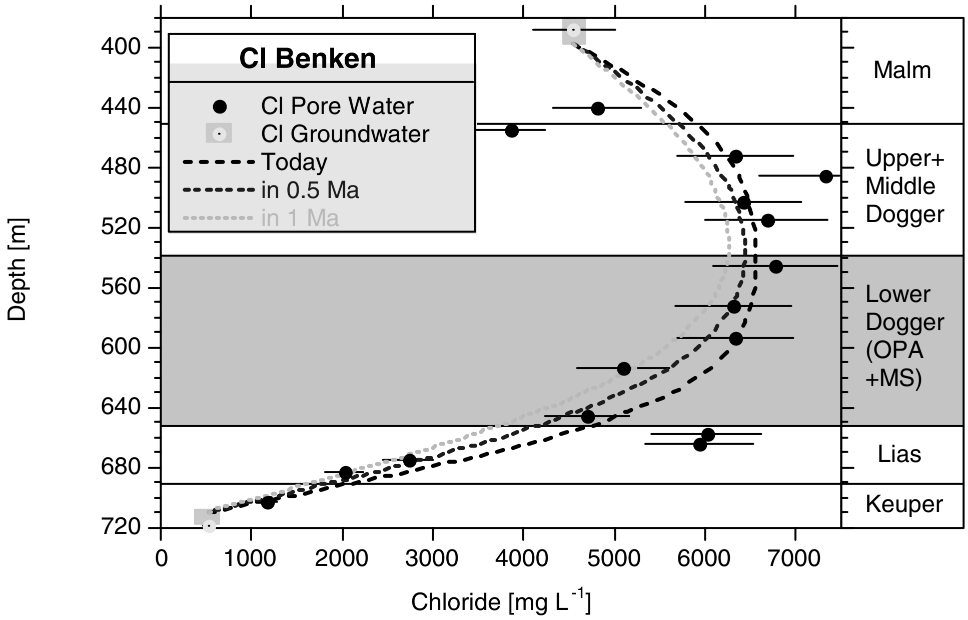


Figure 3 shows the results of equilibrium modelling of the change in mineralogy and redox potential (pe) as O_2 gas reacts with rock of the composition of the Opalinus clay. The reactions are those expected from field observations: Pyrite oxidation increases the Fe(II) concentration leading to siderite and gypsum precipitation. When pyrite disappears, there is a slight increase in the pe as siderite begins to dissolve leading to Fe(III) formation and the precipitation of $Fe(OH)_3$. Finally, as siderite is exhausted, the pe rises steeply. Note that the formation is able to absorb 16 mol O_2 per kg formation water before there is a significant increase in pe. This illustration does not include oxidation of organic C which makes up at least half the total reduction capacity of the Opalinus clay.

4. Capacity to buffer external changes

The only constituents of clay porewater that are not chemically buffered by the rock are chloride and trace components like bromide, iodide, and noble gases. Their distribution within the formation is strongly influenced by the composition of ground waters in adjacent aquifers and is therefore site-specific (see for example Mazurek *et al.*, this volume, and references cited therein). However, due to the fact that diffusion is the dominant transport process in these very low permeability rocks, long-term changes are extremely slow, even in the case of relatively large concentration gradients.

Figure 4. Evolution of the Cl profiles at Benken under the assumptions of diffusive transport and constant boundary conditions at the aquifers with values as today (Nagra, 2002a; Gimmi & Waber, 2003) The indicated times are based on a diffusion coefficient $D_p = 5\cdot10^{-11}\ m^2\ s^{-1}$. Changes within the potential host rock remain relatively small (≤ about 20%) within this time span.



Nagra has recently completed a safety analysis of a potential repository in the Opalinus Clay in the Zürcher Weinland (Nagra, 2002b). A scenario tested as part of this analysis was the effect on chemistry at the repository horizon of continuing diffusive exchange between Opalinus Clay porewater and water in the aquifers overlying and underlying the Opalinus Clays for a time of up to 1 My (Nagra, 2002a; Gimmi & Waber, 2003). Based on palaeohydrogeological considerations these assumptions are reasonable, even though future glaciation might increase the influence of advection slightly during relatively short time periods of glacial load. Direct infiltration of water during a glacial period is not expected. There is no isotopic evidence that one of the 13 Pleistocene glaciations has influenced the porewater composition of the Opalinus Clay. The results of the diffusion calculations for the long-term evolution of Opalinus Clay porewater are shown in Figure 4. They show that for times of the order of 1 My the effects of diffusive changes propagating to the repository horizon are very small and illustrate the strong buffering effect of clay rocks.

5. Conclusion

Studies of several European clay rocks illustrate their capacity to buffer changes in pH and redox potential and to resist changes in concentrations of non-reactive solutes like chloride. pCO_2 (and consequently pH) appears to be controlled by reactions among formation silicate and carbonate minerals, with the buffer capacity dependent on the abundance of these minerals. Oxidation of clay rocks are buffered by reactions with such reduced substances as pyrite, siderite and organic C. The reduction capacity depends on the concentrations of these substances. Finally, changes in non-reactive solutes in indurated clay rocks like the Opalinus Clay occur only slowly in response to external concentrations because solute transport takes place predominantly by diffusion.

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SOME ELEMENTS OF UNDERSTANDING OF ARGILLACEOUS MEDIA STABILITY

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1. Introduction

In the absence of tectonic or metamorphism influence, the argillaceous formations can be stable over very long periods of time: 150 million years for the Callovian-Oxfordian formation (COX, Bure Site, France), 180 million years for the Opalinus Clay (OPA, Mont Terri, Switzerland), 420 million years for the Silurian formation of the Arabic platform. The aim of this paper is to highlight three aspects that provide a better understanding of the stability of this type of formations in the context of their potential for the disposal of nuclear waste:

1. Favourable liquid/solid ratio, allowing very strong buffering effects.
2. Progress in the understanding of the clay stability.
3. Experimental evidence that the carbonate system is controlled internally by the mineralogy.

2. Favourable liquid/solid ratio

The very fine granulometry of the clay minerals leads to the formation of rocks with very low porosities. This porosity is typically water-saturated and therefore the water content is very low. For the COX, the water contents range between 6 and 9 ml for 100 g of dry rock (Gaucher *et al*, 2004). In other words, the masse of rock per litre of porewater ranges between 11 and 16 kg. Some major rock minerals are therefore: 18 mol/l illite, 8 mol/l montmorillonite, 34 mol/l calcite, 1.6 mol/l pyrite. The number of exchange sites is also very large (around 3 mol/l), the rock can be thus considered as a "super-exchanger". In this context, it is easy to understand that the liquid/solid ratio is extremely favourable and that infinitesimal rock dissolution leads to thermodynamic equilibrium between water and solution in static conditions.

Another consequence of this low porosity is the very low rock permeability and therefore the major transport mechanism is molecular diffusion. A low diffusion coefficient is the parametre that permit a very long water residence time. This is the ideal situation for equilibrium between solid and solution. This equilibrium is clearly imposed by the rock.

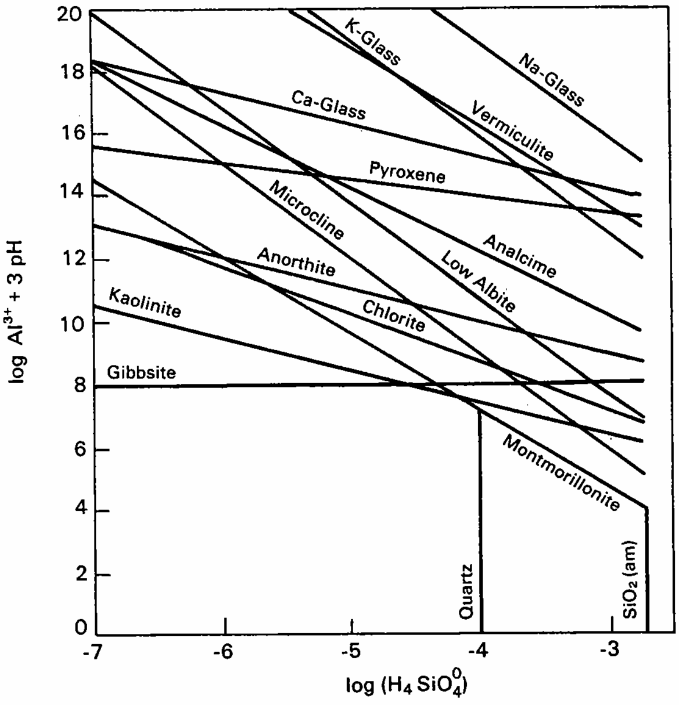
Consequently, the liquid/solid ratio is a key parameter for modelling chemical perturbations in clayey systems. It is essential that the water/rock interaction model take into account the major phases (in %) in order to be able to model the influence of buffering. The illite and smectite that are the major phases in the claystones should be considered as exchangers and as well as soluble phases.

3. Progress in the understanding of clay stability

The stability of the clay minerals is a key question is the context of nuclear waste disposal. It is a difficult question due to of the controversial nature of the clay minerals. This is illustrated, for example, by the comments of Kittrick and Peryea (1989): "For complex minerals of variable composition [clays], the present issue is not their exact stability, but whether or not their stability can be determined".

In order to compare the solubilities of clays minerals with those of other aluminosilicates, activity diagrams have been used extensively (Sass *et al,* 1987). The clays are hydrous minerals with a high exchange capacity and, in our opinion, they cannot be included in activity diagrams considering alkaline or alkaline-earth cations, for example: Log(H_4SiO_4) vs Log(K^+)/Log(H^+). In stability diagrams, the slopes of lines represent mineral-solution equilibria. However, for the 2:1 clays, the Log(K^+)/Log(H^+) axis is more related to ion exchange reactions than to clay mineral stability. Consequently, interpretations of porewater evolution made using this type of diagram are misleading. Such stability diagrams for clay minerals should consider the major ions of the mineral structure, i.e. Al and Si. Rai and Kittrick (1989) have published a very interesting diagram in the system SiO_2-Al_2O_3-H_2O at pH 7 for log(Mg^{2+})=log(Na^+)=(log K^+)=-3 mol/l, log(Ca^{2+})=-2.5 mol/l and log(Fe^{3+}) in equilibrium with hematite (Figure 1). In this diagram, the mineral that maintains the lowest log Al^{3+}+3 pH activity at a given H_4SiO_4 activity is the most stable. For example, at log(H_4SiO_4) of -3.5, the minerals in order of increasing stability are Na-glass, vermiculite, Ca-glass, K-glass, pyroxene, analcime, anorthite, low-albite, gibbsite, chlorite, microcline, kaolinite, montmorillonite. It is interesting to notice, that for the range of H_4SiO_4 activity considered for a nuclear waste disposal in argillaceous media (10^{-4} to $10^{-2.5}$ mol/l), the montmorillonite is the more stable phase of the list.

Figure 1. Relative solubility in the system SiO_2-Al_2O_3-H_2O of selected primary and secondary minerals
(Rai and Kittrick, 1989)

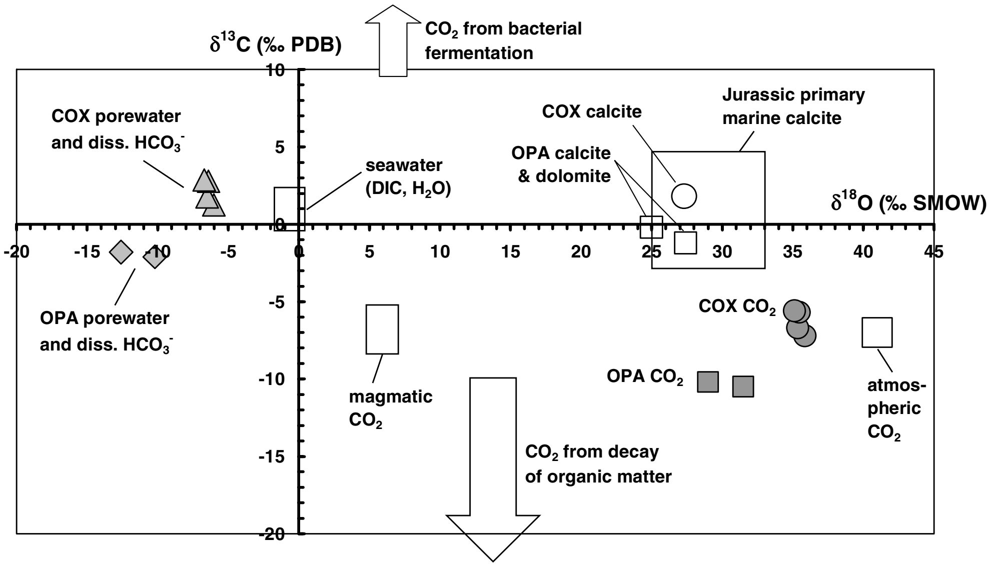


For the solubilities of 2:1 clay minerals, it is important to use a system of equations that link the solubility of the structure to the ion exchange properties and to some extent to clay mineral hydration reactions. To introduce the solubility of montmorillonite or any other mineral having an important exchange capacity in a numerical model of water/rock equilibrium, the software PHREEQC (Parkhurst and Appelo, 1999) presents a new interesting option. With this software it is possible to introduce a system of equations that accounts for the solubility of the structure of the mineral (with Si, Al, Fe, Mg, …, in tetrahedral and octahedral positions) and all the ion exchange reactions. The total solubility of the phase is defined by this system of equations and this representation is believed to be very relevant to the natural processes.

4. Evidence for carbonate system control

The argillaceous media are very often also carbonaceous media. In the COX and in the OPA formation, calcite and dolomite are present in large amounts. These minerals strongly influence the buffer capacity of the rock as a whole. It is therefore important to understand how the carbonate system functions. For this system the pCO_2 is a key parameter, which contributes to determine the pH of the porewater. In a sedimentary rock, the pCO_2 can be fixed externally by diffusion of CO_2 coming from the atmosphere, the mantle of the earth, the degradation of organic matter and the bacterial activity. Coudrain-Ribstein *et al.* (1998) have developed a model that considers an internal origin for the pCO_2. The partial pressure of CO_2 is determined by the equilibrium between the solution, calcite, dolomite and a Mg-aluminosilicate (chlorite or montmorillonite). The minerals control Ca, Mg and HCO_3 in this system and the pCO_2 is fixed by the mineralogical assemblage.

Figure 2. $\delta^{13}C$-$\delta^{18}O$ plot showing measured isotopic compositions of COX (filled circles) and OPA (filled squares) outgassed CO_2, and calculated isotopic compositions of porewater ($\delta^{18}O$ water) and dissolved bicarbonates ($\delta^{13}C$ diss-bicarb) for COX (filled triangles) and OPA (filled diamonds) interstitial water. Measured isotopic compositions of solid carbonates (calcite and/or dolomite) occurring in each of the two argillites (open circle and squares) and compositional fields of main sources of CO_2 are also shown.



At BRGM, we have developed a novel approach based on the measurement of the gas partial pressures where the gases are out-gassed from a core sample confined within a hermetically cell (Gaucher *et al.,* 2003). Isotopic analyses of CO_2 naturally released from the cores have also been performed (Girard *et al.*, 2004). The method was developed with the primary aim of avoiding any physical/chemical disturbance of the argillite sample that might affect porewater isotope composition. It provides indirect determination of the $\delta^{18}O$ and the $\delta^{13}C$ of dissolved carbon.

The log pCO_2 in the two systems (OPA -2.88 to -2.22 bar, COX -2.5 to -2.14 bar) are in the range foreseen by Coudrain-Ribstein *et al.* (1998) for sedimentary rocks. And the measure of the isotopic composition of the $CO_{2(g)}$ allows the calculation of the isotopic composition of the aqueous HCO_3 (pH between 7 and 8). The figure 2 shows clearly that the $\delta^{13}C$ of HCO_3 corresponds to the $\delta^{13}C$ of the carbonaceous minerals of the rocks. This demonstrates that the pCO_2 of the OPA and of the COX is regulated internally by the minerals of the rock and this constitutes a strong buffer for any acid-base perturbation.

5. Conclusion

The argillaceous rock appears to be a very stable media if the range of temperature and pressure do not evolved significantly during long periods of time. The characteristics of a high rock/water ratio, clay mineral stability and carbonate system auto-regulation are particularly favourable to chemical perturbation buffering in such geological formations.

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CHEMICAL BUFFERING/MINERALOGICAL ASPECTS: MINERALOGICAL STABILITY

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Part 1: Temperature effects on clay rocks in natural systems

In natural systems, the heat that leads to the conversion of clay minerals may proceed from three sources:

- The burial under sediment layers that produces diagenetic conversions and may locally reach stages of burial metamorphism; in this system, continuously heated, the duration of reactions may reach values over ten million years.
- Volcanic or igneous intrusions that induce a contact metamorphism in the surrounding rocks; the duration of reactions may vary from a hundred to a million years; heating is also progressive and continuous.
- Hydro-thermal fluid flows in fractured or porous environments (geothermal systems) are more superficial expressions of thermal transfers; the duration of reactions in that case are much shorter, ranging from a few hours to a few thousand years in thermal systems that can be pulsed. Ranges of temperatures covering the stability fields of minerals are available in the literature for hydro-thermal systems.

Clay minerals can not be considered reliable geothermometres because they are often not in chemical equilibrium with the surrounding fluids. Reactions appear to be controlled, by kinetics, and this makes it difficult to transpose temperatures from one system to another.

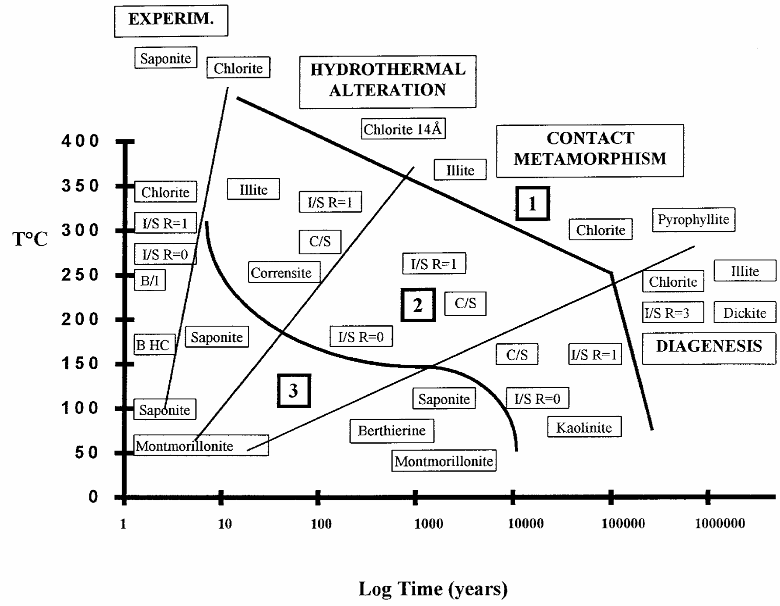
The most widespread occurrences of clay minerals are part of the dioctahedral smectite→IS →illite and trioctahedral smectite→CS→chlorite conversion series. Their spatial distribution in a given system is connected to the kinetics of conversion reactions rather than by equilibrium of thermodynamically stable phases. Nevertheless, the spatial distribution of these clay minerals may reveal a thermal gradient, but the temperature associated to a given mineral is not necessarily transposable to another system.

The most often described occurrences of clay minerals are summarised in the following time-temperature schematic diagram (Figure 1). This presentation does not take into account the fact that these minerals might appear in other zones and in specific conditions.

Overall, what emerges from the schematic diagram below is that the higher the temperature of a system, the shorter the period of time necessary to the formation of a phase. This is best illustrated by the experimental formation of saponite in two days at 800°C (Iiyama, J.T. and R. Roy (1963). It appears that smectites are a dominant reaction produced over shorter timescales, even at high temperature. So during the first thousand years smectitic minerals should be stable at temperatures near 100°C. However, with increasing time a tendency to the interstratification with illite or chlorite is seen.

It can be concluded that study of an intrusion into surrounding clays allows the detection of transformations occurring on a very short period of time (ten to thousand years). This is relevant for performance assessment of a nuclear waste storage. It seems that even with high temperature input the effect on clay is spatially limited.

Figure 1. Clay minerals stability in time-temperature diagram (Pellegrini R. *et al.,* 1999)



B	Beidellite
B/I	Beidellite/illite mixed layer
B HC	High-charge beidellite
C/S	Chlorite/smectite mixed layer
I/S	Illite/smectite mixed layer
1	Illite and chlorite preponderance area
2	Mixed layers preponderance area
3	Smectites preponderance area

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Part 2: A new approach to assess the mineralogical organisation and stability in clay rocks, construction of a tri-dimensionnal structure model at a micrometric scale

Long term evolution of the clay mineralogy and their associated chemical retention capacity is mainly controlled by the microstructure of the clay matrix. Indeed, microstructure governs possible interactions at the level of clay mineral surfaces and also is modified by them. Here, microstructure defines the complex spatial arrangement of the porosity and the minerals, from the scale of clay particles to that of laboratory samples. Characterizing microstructure allows therefore to define the sample mineralogical heterogeneity and the distribution of fluid pathways as compared to the geochemical porosity where fluid - mineral interactions occur (Pearson, 1999).

It is now commonly accepted that rock physical properties are mainly function of the microstructure (Guéguen *et al.,* 1997). Furthermore, their characterization is partly realized in laboratories in order to determine parameters used in models. Comparing experiments results with model one's often underscores discrepancies that are partly attributed to the unknown of microstructure characterictics (Gens *et al.,* 2002).

Since several years, ERM and HYDRASA have developed a new petrographic approach based on multiscale imaging techniques:

- minerals and porosity mappings of areas of several millimeter sizes with a Scanning Electron Microscope or an Electron Probe MicroAnalyser (EPMA) (Prêt *et al.,* 2003), and three-dimensional reconstruction with high resolution X-ray microtomography,
- porosity mappings by autoradiograph of sample sections of several centimeter sizes (Sammartino *et al.,* 2002; Prêt *et al.,* 2002).

Comparing these images by superimposition with the help of specicific imaging tools allows to locate quantitatively porosity related to mineral spatial distribution.

Two examples are presented: a three-dimensional reconstruction of argillite sample by X-ray microtomography and a two-dimensional mapping of bentonite minerals by combination of chemical element maps obtained with EPMA.

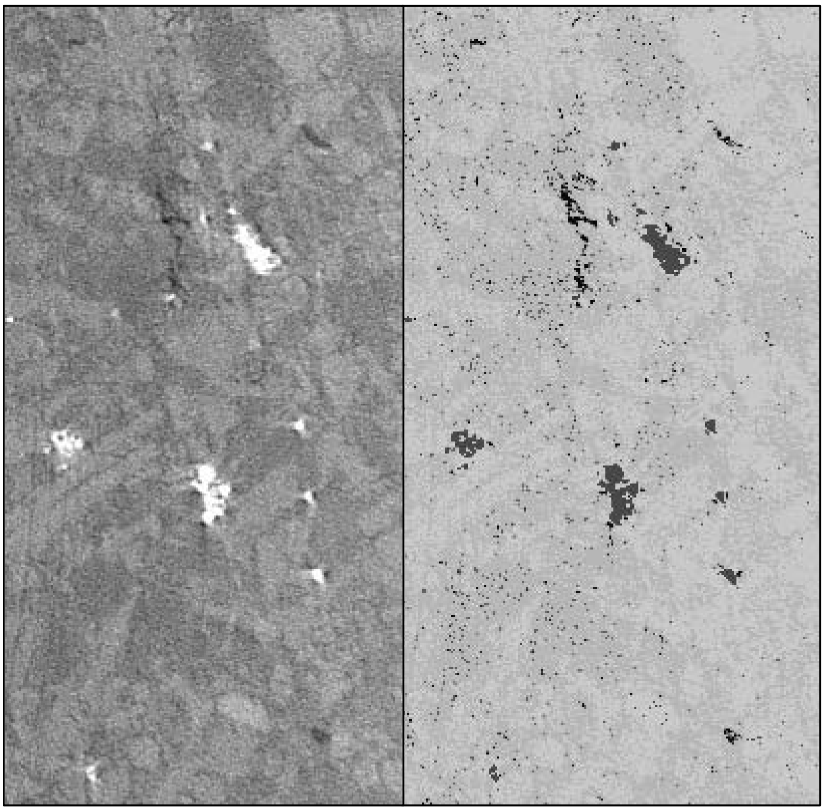
Three-dimensional reconstruction of argillite sample by X-ray microtomography

Microtomography is based on X-ray beam attenuation through a rock sample. It is similar to medical tomography but with a better contrast and an infra-micrometric resolution. X-ray attenuation coefficient is calculated for each voxel of the sample volume, by knowing initial and transmitted beam intensities regarding each section with beam multi-directional azimuths. The attenuation coefficient of each voxel is a function of the local bulk density and the average atomic number.

A three-dimensional reconstruction of an argillite sample of Bure site (ANDRA, Underground laboratory) was realized at the ESRF (European Radiation Synchrotron Facility). One section of the reconstructed volume is shown in the following figures. These images reveal the organisation of macropores and minerals in terms of bulk density variations in the sample volume.

This two-dimensional representation seems to be convenient for the reconstruction of the three-dimensional microstructure of the rock that should be suitable for the application of future modellings of transport properties (Delay *et al.*, 2002). Sample volumes will be presented during the communication.

Figure 2. Image selection (200*400 pixels) of a reconstructed slice of a compacted argillaceous sample, pixel size is 1.4 microns. X-ray beam energy was 25.6 keV and transmission ratio about 16%. On left, raw image in grey levels. On right, thresholded image to highlight the microstructure spatial distribution: light grey as carbonates, medium grey as the porous clay matrix, dark grey as pyrite and black as macroporosity.

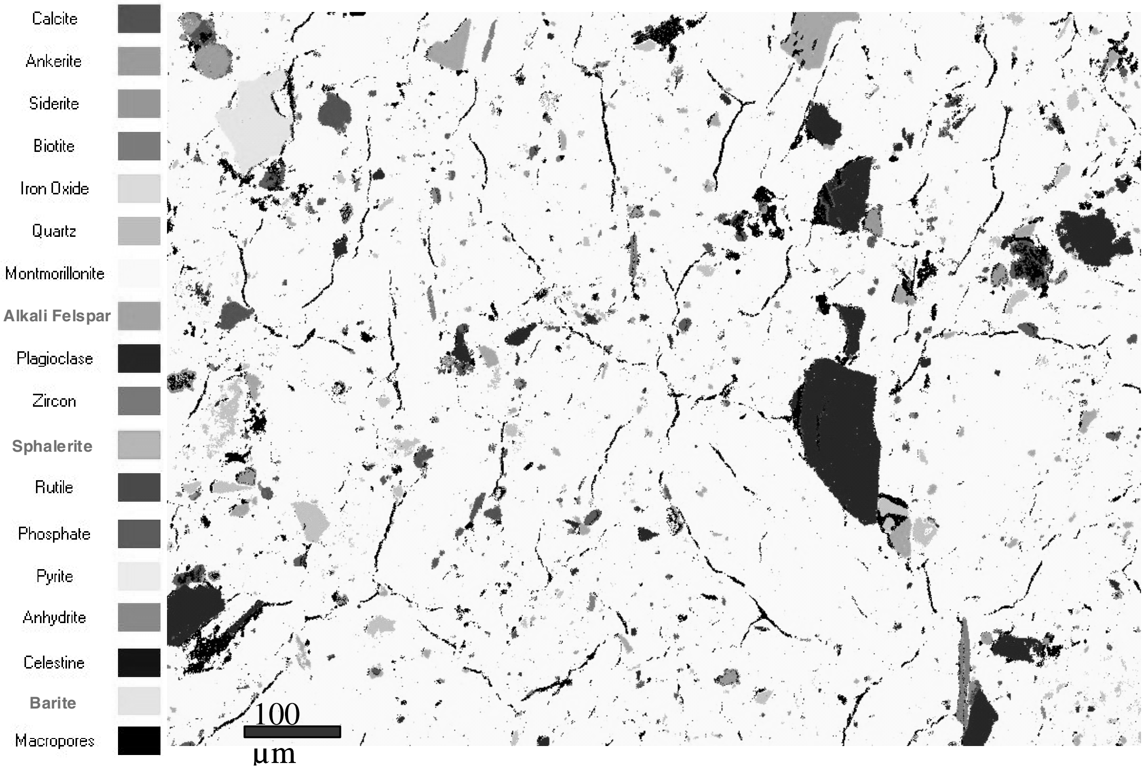


Mineral and porosity mapping using EPMA

Quantitative chemical element maps are obtained using EPMA. Until 16 elements are mapped with a spatial resolution close to one micrometer and image size of 1 024 per 768 pixels. The analysed area is thus pluri-millimetric. The identification and the spatial distribution of all mineral species is then performed thanks to a specific software, developed for the analysis of each pixel chemistry. The sum of oxide weight percentages measured for each pixel is also exploited in order to estimate the local porosity.

As an example, minerals and macropores of a compacted MX80 bentonite sample are presented in Figure 3. This imaging technique also provides the mineral contents and the mineral specific porosities. One of the main information was to quantify the mean porosity of clay aggregates and to identify the macropore network. These results are of great importance for the understanding of transport processes in this clay matrix which was long time considered as homogeneous.

Figure 3. Mineral and macropores mapping of compacted MX 80 bentonite
(density = 1.58 g/cm^3)



Conclusion

These methods are suitable to characterize textural and mineralogical transformations due to any physico-chemical impact affecting rocks constituted by swelling clay minerals.

In particular they are enough sensitive to discriminate clay mineral changes subsequent to a thermal event or a chemical perturbation (alkaline or iron plume) where mineralogical transformations inevitably induce textural modifications.

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NATURE AND REACTIVITY OF ORGANIC MATTER IN ARGILLACEOUS FORMATIONS: EXAMPLE OF THE CALLOVO-OXFORDIAN OF BURE (FRANCE)

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In carbon cycle models, it is admitted that less than 1% of produced organic carbon is transferred to the geological cycle as sedimentary organic matter (Tissot and Welte, 1984). Although, coal or petroleum source rocks are most well known, sedimentary organic matter also occurs in various concentrations throughout many different rock facies. Organic matter is therefore a witness of the record of environmental changes as well as biomass evolution through time. It is also a reliable tracer of diagenetic conditions, from sediment deposition to metamorphism and subsurface alteration.

Especially in the case of argillaceous sediments, known for their potential proneness of organic matter, the study of fossil organic matter is able to unravel a large amount of information concerning the geological past (depositional conditions and preservation, palaeoenvironment, burial, thermal history) as well as the future (effects of induced thermal perturbation, oxidative alteration, biodegradation). We are presenting here data obtained on the Callovo-Oxfordian argillaceous formations of Bure (France), which are the target layers for the installation of a future laboratory.

1. Analytical methods

Organic matter from ancient sediments is an association of kerogen (organic matter insoluble in usual solvents, Durand, 1980) and extractable organic matter (aliphatic and aromatic hydrocarbons as well as macromolecules: asphaltenes and resins). The nature of this organic material necessitates the use of specific analytical equipment. In our study we used:

I. organic petrography and palynology using visible as well as UV light, completed with SEM coupled to X-ray spectrofluorescence and imaging;
II. elemental analysis, Rock-Eval pyrolysis;
III. spectroscopic methods (micro-Fourier Transform Infrared Spectroscopy);
IV. gas chromatography coupled to mass spectrometry for molecular analysis.

Some of the information concerning the geological history of sedimentary organic matter is only revealed if its reactivity is studied. Among specific methodologies, we used artificial maturation (Landais *et al.,* 1989). This technique allows to reproduce in the laboratory the major steps of organic matter thermal alteration and leads to the determination of kinetic parameters as well as reaction pathways.

2. Petrographic, palynologic and geochemical characteristics of the organic matter from Callovo-Oxfordian argillaceous formations of Bure (France)

Rock-Eval data indicate that the sediments from the Callovo-Oxfordian of Bure contain less than 1% Total Organic Carbon. Hydrogen Index varies between 50 to 300mgHC/gTOC and Tmax from 422 to 430°C.

Petrographical analysis reveals the presence of: 1) macerals (vitrinite, inertinites) mostly identified as plant remains, often oxidized, with evidence of significant transport; 2) amorphous macerals, sometimes UVfluorescent (liptinites). Palynological analysis reveals the presence of significant amounts of acritarchs associated to amorphous organic matter. Dinoflagellates and sporomorphs are less abundant, but frequently encountered.

Dichloromethane extractible organic matter represents from 0.04 to 0.3mg/g of rock. It is composed of 60wt% polar compounds (asphaltenes and resins), 22 to 46wt% of aromatic hydrocarbons and 16 to 34wt% of aliphatic hydrocarbons. Chromatographic traces of aliphatic hydrocarbons indicate compounds in the nC_{15} to nC_{35} range, which can be roughly described in two parts: 1) The aliphatics of the nC_{15} to nC_{25} range are often composed of an unresolved hump of cyclo and iso alcanes, together with well resolved alcanes; 2) The aliphatics of the nC_{25} to nC_{35} range present n-alcanes with odd predominance as well as cyclo-alcanes typical of biomarkers. The relative proportions of the hydrocarbons of the nC_{15}-nC_{25} range compared to nC_{25}-nC_{35} range (n-alcanes and biomarkers) varies significantly throughout the sedimentary column. The pristane/phytane ratio varies from 1.5 to 3 (the stronger values corresponding to the samples with higher sulfur content). Aromatic hydrocarbons are mainly represented by series of alkylnaphthalenes, alkylphenanthrenes, alkyldibenzothiophenes and aromatic steroids.

All the above mentioned features of the organic matter from the Callovo-Oxfordian of Bure indicate a mixture between marine (acritachs, dinoflagellates, algal macerals, typical biomarkers) and continental sources (vitrinite and inertinite, recognizable plant remains with typical oxidation and transport features, sporomorphs, n-alkanes with odd predominance, biomarkers).

3. Heterogeneity of the organic matter within the sediment and throughout the argillaceous formations

3.1 Organic-mineral association at the microscale

The extremely fine texture of the claystone and macerals necessitates the use of SEM coupled with X-ray fluorescence imaging in order to visualize organic matter distribution. This technique used on polished sections of sediments shows that organic matter occurs as aggregates (local concentrations, fine layers parallel to bedding are not rare), but also as very fine particles disseminated through the rock. The distribution of the kerogen within the rock is therefore very heterogenous at the few micrometer scale. Although association between organic matter and pyrite is common, systematics and statistics of organic-mineral associations in terms of reaction micro-sites have yet to be determined.

3.2 Distribution of the organic matter in the sedimentary series: a record of environmental changes during deposition

Our data show a clear evolution of the geochemical characteristics of the organic matter from Callovian to Oxfordian. A remarkable change is noticeable during the Callovian-Oxfordian transition. The Callovian is characterized par molecular signatures dominated by marine input, while evidence of continental input significantly increases in the Oxfordian. A similar change is noticed by palynological analysis (decrease of the contribution of acritarch and increase of the spore-pollen input from Callovian to Oxfordian) (Huault *et al.,* 2003). We are currently conducting research in order to have more detail on paleoenvironmental changes during the Callovian to Oxfordian transition.

4. Maturity of the organic matter and impact on the determination of paleothermicity and paleoburial

All data collected on the organic matter of the Callovo-Oxfordian from Bure indicate a thermal immature sediment. More specifically, the presence of biomarkers (triterpanes for instance) bear biological conformation and contain double bonds which can only subsist since the Callovo-Oxfordian if the sediment never experienced a significant heating. We used artificial maturation (Landais *et al.*, 1989) in order to study the thermal behaviour of the organic matter. Experiments allowed to study the thermal stability of the triterpane assemblage and determine their maturation kinetic parameters. Modeling of the biomarker behaviour as a function of heat lead to the conclusion that the organic matter from the Callovo-Oxfordian of Bure did not experience temperatures higher than 40°C. Similar conclusions were reached by fluid inclusion and clay geochemistry studies. Moreover, the maturity modeling of the biomarkers using today's heat flux of 33°C/km and a surface temperature of 10°C allowed to reconstruct the theoretical maturity/depth relationship for the Callovo-Oxfordian. Comparison with the actual maturity of the organic matter in wells located in the east of the Paris Basin indicates a shift of 400-600 m. This shift corresponds to the thickness of eroded sediments since the beginning of uplift of this part of the basin.

5. Conclusions

The study of the organic matter from the Callovo-Oxfordian claystones of Bure brings interesting information and both the sedimentological and diagenetic history of this part of the Paris Basin. Combined techniques allowed to identify and describe the various aspects of the organic matter intimately associated to the sediment. Although present in low amount (less than 1wt% of rock), its distribution is heteregenous at all scales, from the sedimentary pile to the microscale.

The combined use of facies sedimentology, sequence stratigraphy and organic geochemistry is an efficient tool to investigate the heterogeneity of argillaceous formations (Fleck, 2001; Fleck *et al.*, 2001a, b; 2002). As a matter of fact, argillaceous series are often "monotonous" on a sedimentological perspective (rare sedimentological structures, often destroyed by bioturbation, almost constantly fine layered sediments) and remain difficult to investigate through conventional sedimentological approaches. By combining sedimentological information with Organic Geochemistry, it is possible to more precisely distinguish depositional and diagenetic changes in argillaceous formations.

The study of the potential reactivity of the organic matter associated to clays using artificial maturation and kinetic modeling of biomarker transformation allows to determine maximal paleotemperature and burial. This information is an important constrain to the reconstruction of the geological history of argillaceous formations. In regards to the past conditions of burial of the Callovo-

Oxfordian and the uplift context at which this part of the Paris Basin will evolve from today to the next 1 My, it is most likely that the organic material will thermally not evolve further.

Our experimental work on the organic matter from Bure also demonstrated a significant potential reactivity towards oxidation (Faure *et al.*, 1999; Elie *et al.*, 2000) and biodegradation. This reactivity is very much conditioned by the organic-mineral associations (catalysis of low temperature oxidation of the organic matter by clays for instance: Faure and Landais, 2000; Faure *et al.*, 2003). Organic matter in argillaceous sediments can therefore also be used as tracer of human induced perturbations.

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EARLY FRACTURATION IN ARGILLACEOUS MASSIFS AND RELATED CARBONATE TRANSFER

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To be sure of the stability in the future of underground storage in argillaceous massifs needs to understand the present-day properties of the sediment in the light of its history. It is why early fracturation and associated carbonate transfer in shaly-marly massifs is one of the major themes of the *MINANDRA Partnership* (École des Mines de Paris + Andra + some collaborators). That joint program involves tens of geoscientists, and we would like to present here the main objects we study and the methods we apply.

The base of the project is made of selected large reference-outcrops where an early fracturation is undoubtedly visible in the field. Two sets of such outcrops are presently worked in south-eastern France:

- Aptian-albian marls (*Marnes Bleues*), at *Bevons* (near 04-Sisteron) and *Saint-André de Rosans* (W. of 05-Serres).
- Domerian marls, at *Clue du Vançon* (E. of 04-Sisteron).

Both are made of thick marls and intercalated limestone beds and nodules.

Early fracturation: sandy dykes and sills

Recognized since the 80s, the sandy dykes and sills of the *Marnes Bleues* of *Bevons* and *Saint André de Rosans* record an early fracturation *per descensum* (Beaudoin et Friès, 1982; Beaudoin *et al.*, 1983; Parize, 1988; …): hydraulic fracturation by a sudden sandy turbidity current led to the development of huge vertical and lateral injections in the submarine marly massif. Present depth of the dykes below the feeder channels reaches 250 m (decompacted: more than 400 m) and some sills may be followed laterally on more than 1 km; thickness of the dykes varies from 1 m (near the paleo-seafloor) to some cm, thickness of the sills is frequently plurimetric!

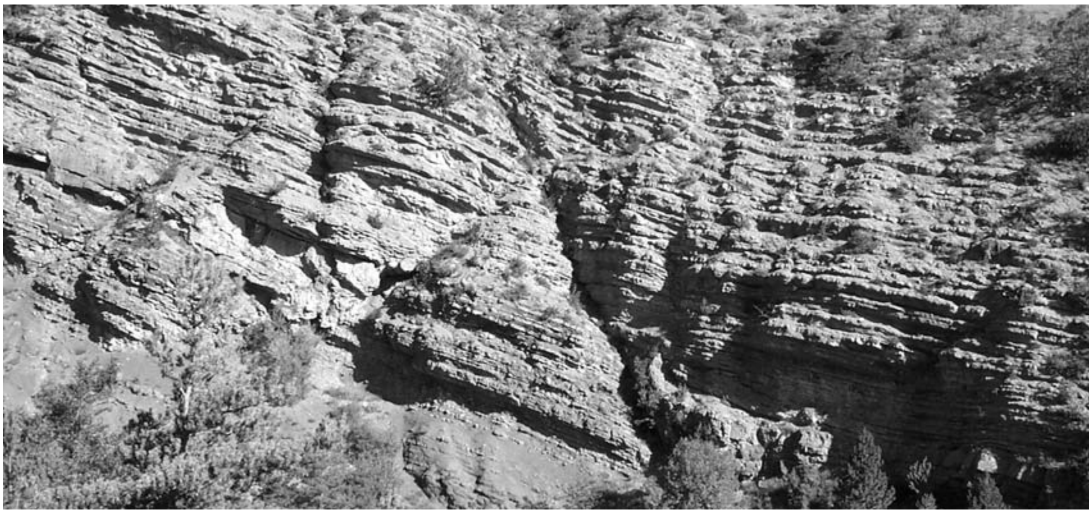
We study the present-day geometry of these fractures, their dimensions, spatial orientation and distribution (clustering? …), the relations with metric and decimetric lithology (limestone and carbonate nodules/marls), anisotropy (relations with the paleo-slope?) and heterogeneities (such as bentonites).

Figure 1. At ***Bevons***, sandy dykes (1) and sills (2) cut the upper albian marine regular alternation sills and dykes (limestone/marlstone); the sand has filled ***per descensum*** early potential fractures by sudden hydraulic fracturation

Synsedimentary faulting and double stratification

The domerian marls of *Clue du Vançon* offer a unique example of double stratification (Beaudoin *et al.*, 1989; Maillart, 1989): the outcrop shows a series of faults between tilted blocks (Figure 2), affecting domerian marls and limestones but sealed by a condensed toarcian level (Coadou & Beaudoin, 1973); these early faults are not planar, but deformed by later compaction.

Figure 2. The Clue du Vançon outcrop, with the synsedimentary faults



In such a context, the peculiarity is the presence of decimetric to metric limy nodules. Recent observations (Moreau, 2001) allowed to characterize a litho-stratigraphic succession affected by normal faults; two sets of nodules may be then recognized: one is banal, according to the general stratification; the other is made of parallel *nodular pseudo-beds*, distant of some meters, and along one family of syn-sedimentary faults (Figure 3).

Figure 3. The two sets of nodules of *Clue du Vançon*: According to the stratification (horizontal) and along syn-sedimentary faults

Early fracturation

Based on these field observations, we try to elucidate the genesis and the propagation of the fractures, the timing regarding the age of the marly massif, the (paleo)mechanical properties of the sediment at different scales, the relations with anisotropy and heterogeneities.

In such early fracturation processes, we try to decipher the role of true tectonics and of differential compaction, to observe traces of later reactivation.

A special effort is presently made on mechanical numerical simulations in diverse static and dynamic configurations.

Compaction

As we work with early fracturation, compaction has obviously to be taken in account. Not only as a cloudy concept, but as an inescapable process recorded in the sediment (Figure 4).

Figure 4. Compacted dyke in the aptian marls of Bevons
The post-injection compaction rate
($\tau = h_0/h$) may be directly measured

Bevons outcrop is a remarkable support for such studies: as the injection of sand (as deep as 400 m below the sea-floor) was instantaneous, it as in a way taken a photo of the compaction state of the massif, as a $\{\tau=f(z)\}$ law, were z is the burial depth. Later compaction led to complete compaction of the sediment. So we can estimate the compaction before and after fracturation.

The values of τ depend on the facies and the carbonate content and reflect the evolution of porosity with depth. The geometry of the dykes when cutting marls or limestones, at time of injection and after later compaction, records the geomechanical properties of the sediment during burial and related progressive compaction, but also the progressive differentiation due to carbonate transfer. Moreover fractures were deformed by later compaction.

Carbonate transfer

The presence of carbonate nodules in shales implies obviously carbonate transfer. In our cases, because of the unusual exposed processes (sand injection in the *Marnes Bleues*, double stratification in the domerian marls), we can go further: carbonate migration may be studied in time and space with the use of carbonate content in an adapted sampling.

Following Lippmann (1955) and Seibold (1962), one can imagine an instantaneous birth of the nodule (or part of such a nodule) by filling the porosity of the sediment by carbonate: if so, we can write a series of relations such as:

$$I_0 + Ű_0 + C_0 = 1 \qquad I_0 / C_0 = I / C$$

where I_0, C_0, $Ű_0$ are the insoluble and carbonate contents and the porosity at time of nodulisation, I and C the present day values.

I_0 is known (in the nodule), and then C_0 and the porosity $Ű_0$.

For example, in the *Clue du Vançon* nodules, we obtain:

(I_0)	29%	32	37	39	41	47	51	56	60
($Ű_0$)	62%	58	51	49	46	38	33	26	22

for nodules of one family or the other.

These porosity values at the moment of carbonatation, from 60% to 25%, suggest burial depths from some tens of meters to 400-500 m. These results are in agreement with the early (and shallow) timing of the faults given by de-compaction.

After carbonatation of the nodules, the marly massif has still undergone compaction. The geometry of the nodular pseudo-beds was affected, even with fault-like shifting and related re-fracturation.

Further developments

The ultimate goal of the project is to elaborate methods of investigations allowing to reveal early fractures in shaly massifs and to consider their possible reactions to future solicitations.

It is why the inter-acting processes during early diagenesis and burial are approached at different scales, from outcrop (tens, hundred of m, km …) to selected samples and sets of samples, in relation with field observations: some preliminary results will be given as examples.

The next step will obviously be to apply validated methods to samples of the Underground Laboratory.

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POSTER SESSION

NUMERICAL INVESTIGATIONS ABOUT THE INFLUENCE OF GLACIAL LOADING ON THE TRANSPORT OF RADIONUCLIDES IN THE OPALINUS CLAY

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1. Introduction

The Opalinus Clay in Northern Switzerland has been identified as a potential host rock for a repository for spent fuel (SF), vitrified high-level waste (HLW) and long-lived intermediate-level waste (ILW). The formation in the proposed siting area in the Zürcher Weinland is at least 100 m thick and is composed of highly consolidated and very low permeable claystone of Jurassic age.

In general, radionuclide transport in the Opalinus Clay is dominated by diffusion. On the one hand, fluid movements in the Opalinus Clay are hindered by the low permeability of the rock and transport by advection is normally of minor importance. On the other hand, long-term geotechnical or geological processes can locally enhance the movements of fluids.

In this study the influence of consolidation driven flow due to glaciations on the radionuclide transport in the bentonite filling of the emplacement tunnels and in the Opalinus Clay is investigated. In the past glaciers from the Alps advanced to the north and the area of the potential repository site was covered by a ice layer with a thickness of several hundred meters for certain time periods. Long term transient flow processes due to glacial loading and unloading have been investigated by Horseman *et al.* (1991). They concluded that, with respect to the pore pressure response during undrained loading the Opalinus Clay behaves more soil-like than rock-like. For such a medium the build-up of an ice sheet drives fluids out of the Opalinus Clay layer, whereas the unloading drives fluid into the clay layer.

The calculations were performed in the framework of the safety assessment for a proposed repository for SF, HLW and ILW in the Opalinus Clay of the Zürcher Weinland. The aims of the safety assessment include the following points (Nagra, 2002b):

- To determine the suitability of the host rock for a repository from the viewpoint of long-term safety.
- To enhance the understanding of the multiple safety functions that the proposed disposal system provides.
- To assess the robustness of the disposal system with respect to effects of phenomena that may adversely affect the safety functions.

All these points require the identification and evaluation of key processes influencing the transport of radionuclides in the geosphere.

2. Theory of consolidation after Terzaghi

In this study I follow the approach described e.g. by de Marsily (1986) and apply Terzaghi's theory of consolidation. Terzaghi's theory is commonly used for the description of consolidation and compaction processes in saturated low-permeability soils (e.g. clay) due to extended load acting on such a medium. The phenomenon of consolidation is associated with the outflow of interstitial water contained in the medium (soil, rock, clay). The pressure applied on the medium is absorbed by the solid phase and partly by the interstitial water. The increase in pressure starts a transient outflow of interstitial water until the fluid pressure is equilibrated again. The loss of fluid allows the consolidation.

Terzaghi's theory of consolidation assumes that:

- The outflow of the interstitial water obeys Darcy's law.
- The hydraulic conductivity of the medium does not vary during the consolidation process.
- The water and the medium (soil, rock, clay) matrix are incompressible. Incompressibility here means that the porosity change is negligible during the process.

The compressibility of the medium is elastic. There is a linear relation between the effective compression stress and the volume of water released.

3. Model setup

For the calculations the program FRAC3DVS (Therrien & Sudicky, 1996) is used. FRAC3DVS is a numerical control volume finite-element and finite-difference code for the simulation of saturated-unsaturated flow in fractures and in porous rock. Different solute transport mechanisms in both, fractures and porous media are also directly accounted for.

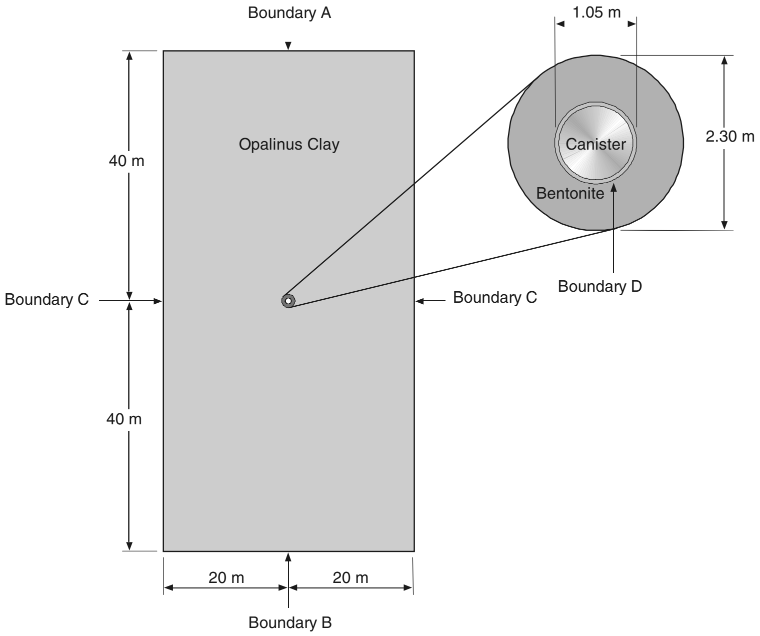
The conceptual model and its underlying assumptions are identical to those of the reference conceptualisation (Nagra, 2002b) except in the treatment of radionuclide transport through the bentonite and Opalinus Clay. Advective transport is driven by glacially-induced flow, evaluated over a one million year period. In the course of the next million years, a periodic series of 10 glaciations is assumed to occur (with an assumed frequency of one glaciation every 10^5 years), starting at 50 000 years from today. The duration of each glaciation is taken to be 20 000 years, with an assumed ice shield thickness of 200 m for eight glaciations and 400 m for two glaciations (fourth and tenth event).

As a result of these glaciations, periodic elastic compaction and rebound of the clay barrier (bentonite and Opalinus Clay) occurs, leading to spatial and temporal changes in the groundwater flow in the clay barrier. The clay barrier is assumed to remain homogeneous, i.e. no fracturing occurs before, during or after ice loading. Flow and transport modelling is based on a 2-D vertical cross-section through the repository representing a single SF emplacement tunnel and the surrounding Opalinus Clay (Figure 1). The presence of neighbouring emplacement tunnels is taken into account by requiring zero flow and transport over the vertical boundaries (boundary C). Water flows along the access tunnel system are neglected in the calculations. This is shown to be a valid assumption for a number of different situations (Nagra, 2002c), and is, therefore, also expected to be a reasonable assumption in the present context of glaciation-induced flow.

Because of the dominant contribution of SF to calculated doses, the analysis is conducted for SF only and is limited to those radionuclides that dominate the summed dose maximum of the Reference Case (organic ^{14}C, ^{36}Cl, ^{41}Ca, ^{79}Se, ^{129}I). The rate of radionuclide release from the SF canisters to the bentonite as a function of time is imposed at boundary D.

At the onset of a glaciation, the hydraulic pressure is instantaneously increased in the Opalinus Clay, but remains as before in the bounding aquifers. For modelling purposes, this situation is equivalent to an instantaneous pressure reduction at the start of a glaciation cycle at the upper (A) and the lower (B) boundary (see Figure 1). When the load is removed at the end of a glaciation, the reverse process is applied. Again, the bounding aquifers equilibrate much faster and an instantaneous increase of the reference pressure at the boundaries is implemented in the model.

Figure 1. Conceptual model for the calculation of glacially-induced flow and transport in bentonite and Opalinus Clay (Nagra, 2002c)



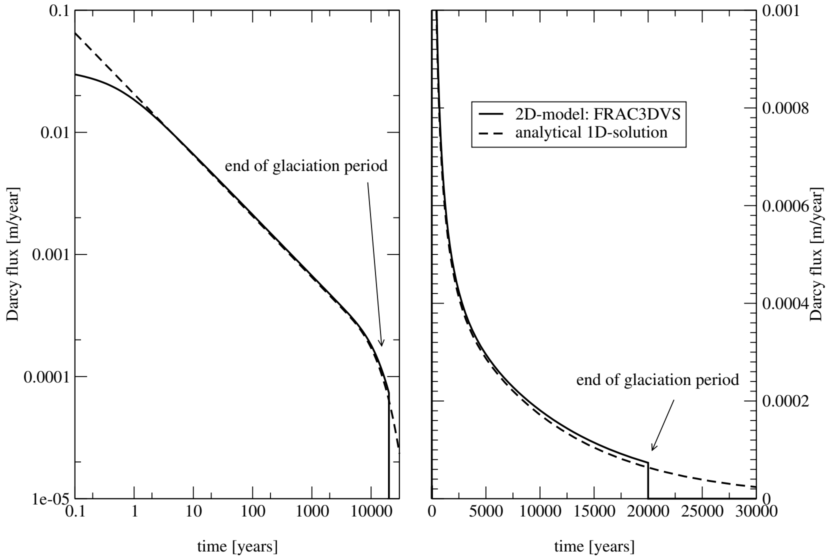
4. Results of the flow calculations

Figure 2 shows a comparison of the mean Darcy flux (i.e. Darcy flux over the upper (A) and lower (B) model boundaries) of the 2-D model, with a 1-D analytical solution (Terzaghi & Peck, 1948). The mean Darcy fluxes were extracted from the model by summing up the flow rate over all boundary elements and dividing this overall flow rate by the boundary area. The differences between the upper and lower boundary is a constant upward directed flux of $2 \cdot 10^{-14}$ m/s ($6 \cdot 10^{-7}$ m/a) due to the natural hydraulic gradient between the lower and the upper boundary.

The comparison of the 1-D solution with the results of the 2-D model shows a good approximation of the flow field. For very small times, t<1 year, the numerical deviates from the analytical solution due to limits for the spatial (and temporal) discretisation of the very high pressure gradients caused by the instantaneous (stepwise) change of the hydraulic boundary conditions.

Shortly before the end of the glaciation period, the mean Darcy flux out of the model is slightly higher than from the analytical solution expected. This can be attributed to the higher specific storage coefficient for the bentonite filling of the emplacement tunnels. This increases the amount of fluid driven out of the formation and causes slightly higher fluxes compared to the fluxes from the analytical solution (which solves the problem for a homogeneous domain).

Figure 2. Mean Darcy flux over the geosphere boundaries during a glaciation with 200 metre ice thickness in a double-logarithmic and a double-linear representation. The solid line shows the flux for the 2-D model and the dashed line is the result of a 1-D analytical solution. The glaciation starts at the time t=0 years.



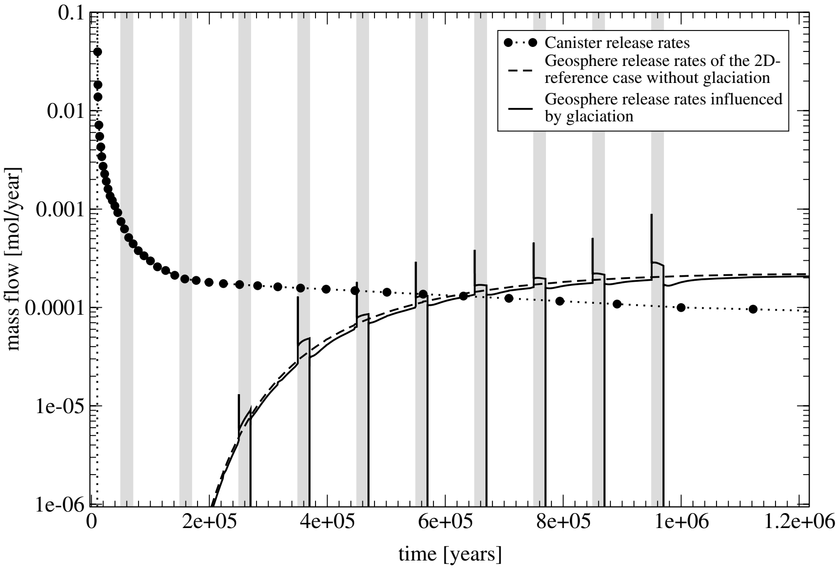
5. Increase of release rates during glaciations

In Figure 3 the breakthrough curve for ^{129}I is shown. During times of glaciation the enhanced advective transport increases the geosphere release rates. After the ice overburden is removed, the hydraulic system recovers (because of the assumed elastic material properties) and fluid flows into the formation. This fluid inflow stops the release of radionuclides for a short time period. The release of radionuclides is then increasing slowly until release rates are nearly those of a model without glaciation. Until the first glaciation the absolute release rates with and without glaciations are identical and the normalised release rate (release rate of a model with glaciation divided by the release rate of a model without glaciation) is constant and has a value of 1. For later glaciations the release rate is lower and the system is slowly reaching an undisturbed state again. Relaxation times are in the order of the length of the interglacials (80 000 years), which is much longer than the duration of the glaciations (20 000 years).

The spatial distribution of the radionuclides influences the additional release of radionuclides during the glaciations. There will be no increase in the release rate if there are no radionuclides near the Opalinus Clay boundaries. A heterogeneous distribution of the radionuclides near the boundary can change the form and the height of the release curve.

The approximate ratios of the release rates in the middle of the two different types of glaciations (approximately 10 000 years after the start of the glaciation) are compiled in Table 1. For all species (anions, cations and neutral species) geosphere transport is dominated by diffusion and advective transport contributes only during or shortly after glaciations. A first obvious effect is that doubling the ice thickness also doubles the relative increase in the release rates.

Figure 3. Example for the influence of glaciation induced flow on the geosphere release rates for ^{129}I. The shaded areas represent glaciation periods. However, for a time period of about 1 year after the beginning of the glaciations the calculated release rates should not be interpreted. This release rates are influenced by an oversimplification in the conceptual model where glaciations start and end instantaneously.



As can be seen from Table 1, the relative increase of the release rates for cations and neutral species ($^{14}C_{org}$, ^{41}Ca) during glaciation periods is much smaller than for anions (^{79}Se, ^{36}Cl, ^{129}I). This can be attributed to the higher (advective) transport velocities of anions. Advective transport is coupled to the fluid velocity, which is calculated from the Darcy flux by division with the flow porosity. As explained in Nagra (2002c) the value of the flow porosity is set to the value of the accessible porosity for the specific nuclide. According to Nagra (2002a) anions see a lower porosity (0.06 for Opalinus Clay) as cations and neutral species (0.12 in Opalinus Clay).

Another process affecting the increase of the release rates during glaciations is the retardation due to sorption. Non-sorbing radionuclides show higher release rates during glaciations (compare e.g. $^{14}C_{org}$ with ^{41}Ca, or ^{36}Cl and ^{79}Se with ^{129}I). This effect is caused by the delayed migration of sorbing radionuclides in connection with the decrease of the fluid velocities (near the geosphere boundaries) during glaciations (see Figure 2).

Table 1. Approximate relative increase of the release rates during glaciations compared to the values of the reference case without glaciations (release rate of a model with glaciation divided by the release rate of a model without glaciation)

	$^{14}C_{org}$	^{41}Ca	^{129}I	^{36}Cl	^{79}Se
200 m ice thickness	1.08	1.02	1.25	1.35	1.35
400 m ice thickness	–	1.04	1.5	1.7	1.7

6. Summary

Glaciation induced flow may enhance the transport of radionuclides in the geosphere. The release rates for non-sorbing anions during the glaciations are up to 1.7 times higher compared to the reference case not influenced by glaciation.

The influence on the transport of cations or neutral species is less than for anions, since the relative importance of the advective transport for anions is higher than for cations and neutral species.

The increase in the release rates during glaciations is lower for sorbing than for non-sorbing radionuclides. This effect is caused by the delayed migration of sorbing radionuclides, compared to the movement of the water, coupled with the reduction of the flow velocities at the geosphere boundary.

Acknowledgements

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BURIAL HISTORY OF TWO POTENTIAL CLAY HOST FORMATIONS IN BELGIUM

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Summary

When dealing with long term stability of repository host rocks, it is important to consider and learn from all past geological events since the deposition of the formations. The burial history of the Boom Clay and Ypresian Clays, both considered as potential host rocks in Belgium, illustrates that the North Belgian region was tectonically relatively stable since deposition. In Northern Belgium, where both formations are located at a few hundreds meters of depth, tectonic movements were relatively small and no significant uplifts took place. The burial history of the Boom Clay in Mol, where the HADES underground research facility is located illustrates this. On the poster, the burial history for both formations is presented at two locations each: one location in the outcrop region and one research site location, where the formation is currently buried under a few 100 metres of sediment.

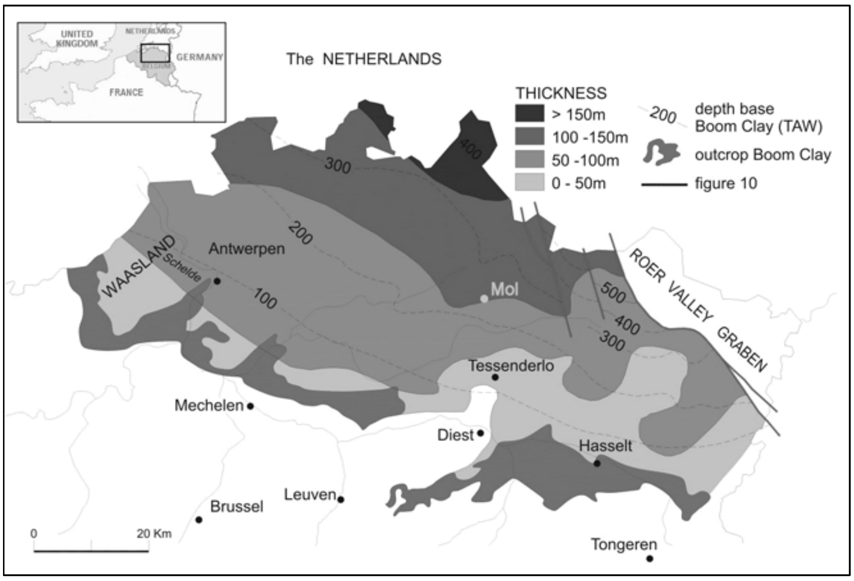
Introduction

At this moment, the Boom Clay and, as an alternative, the Ypresian Clays are investigated as a potential host rock for the disposal of high and medium level radioactive waste in Belgium (SAFIR II, 2001).

The Boom Clay is a marine Oligocene clay of several tens of metres thick. It is the unit stratotype of the Lower-Oligocene Rupelian stage and a well published example of cyclostratigraphy (Vandenberghe *et al.* 1997). The clay was deposited in the southern part of the North Sea basin. It is known in Germany, The Netherlands and Belgium as a continuous gently dipping (±1°) layer. Early diagenesis transformed the originally marly horizons in septaria beds (limestone concretions), which borrowed its name to the German clay, known as Septarienton.

In Figure 1, the present location of the Boom Clay (isopachs and depth of the base) in Belgium is shown (SAFIR II, 2001). As can be seen, the Boom Clay dips towards the North-North-East but also gains thickness in this direction. The reason for the latter is dual: the more north, the higher the sedimentation rate was during deposition, but also during the further burial history, the Boom Clay in the Southern part experienced significant erosion at the end of the Oligocene. The lateral continuity of this deposit, together with the burial history discussed further in this extended abstract will illustrate the geological stability of the region since the deposition of the formation.

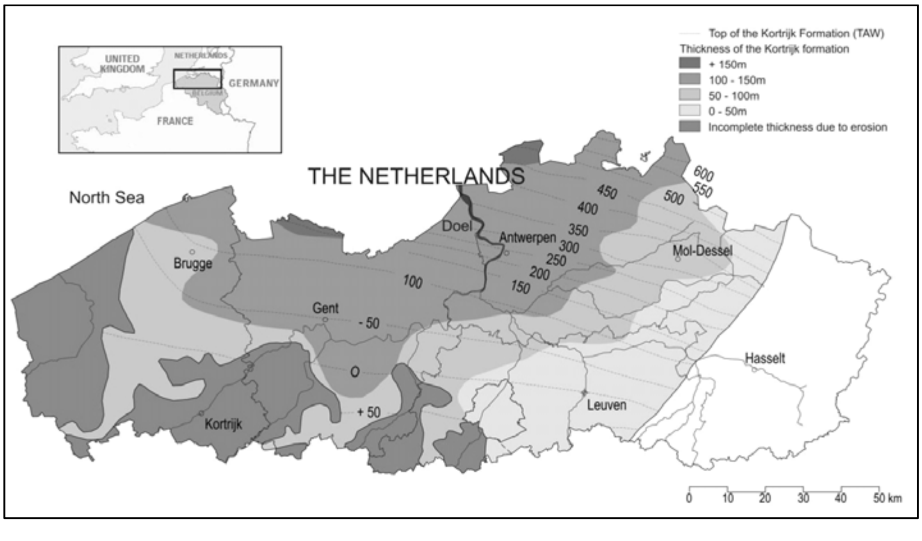
Figure 1. Location of the Boom Clay in Belgium The position of Mol is also indicated (SAFIR II, 2001)



The Ypresian Clays are, obviously, deposited during the Ypresian, which is the first stage of the Eocene and comprises the period of time between 55 and 49 million years ago when the North Sea extended past Paris in the South and London in the West. During this transgression of the North Sea more than 150 m of marine sediments, most of which are rich in clay, were deposited. The "Ypresian Clays" is a term that groups the Kortrijk Formation and the Kortemark Member of the overlying Tielt Formation, and which roughly corresponds with the London Clay (Vandenberghe *et al.*, 1998).

Figure 2 shows the isopachs and depth of the present top of the Kortrijk Formation (SAFIR II, 2001). Just as the Boom Clay, the Ypresian Clays dip gently towards the North-North-East. Towards the east from Antwerpen on, the middle part of the Kortrijk Formation becomes more sandy thus making it a more heterogeneous clay formation.

Figure 2. Location of the Ypresian Clays in Belgium (SAFIR II, 2001)

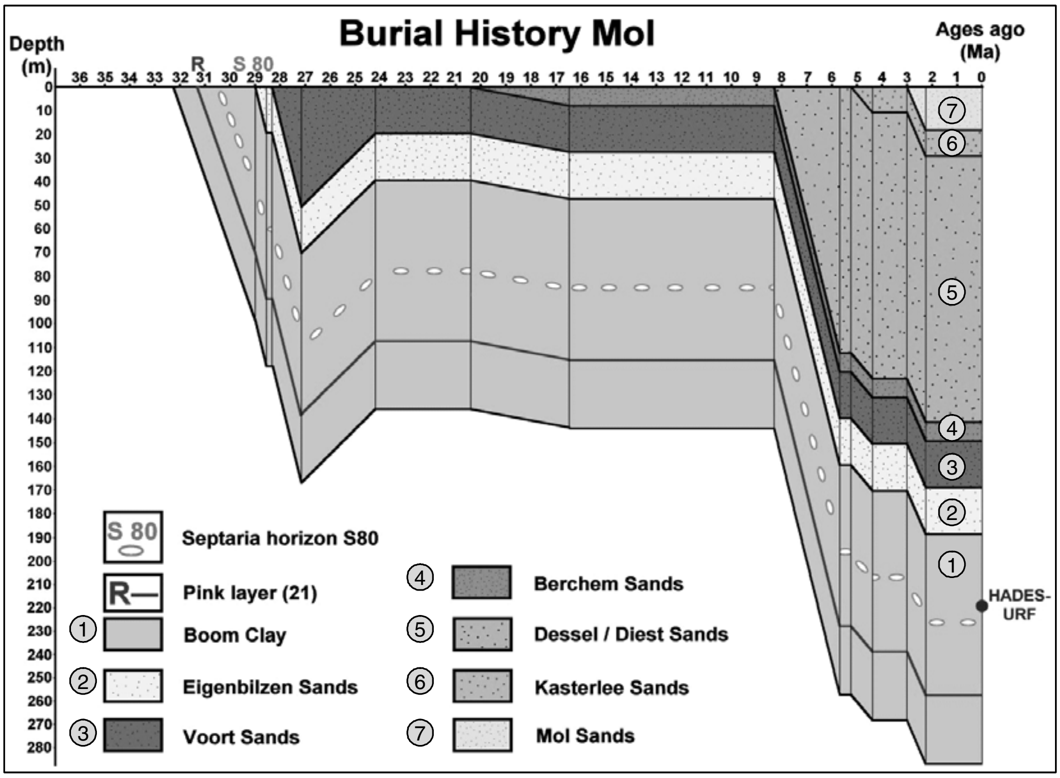


Burial history of the Boom Clay

On the poster, the burial history of the Boom Clay will be illustrated for two reference locations, one at an outcrop place, the other at Mol, where the underground research facility HADES is located.

The burial history of Mol will be briefly discussed (Figure 3). The figure is an updated version of the previously published one (Mertens *et al.*, 2003) as it takes into account the latest stratigraphic research findings from Vandenberghe *et al.* (in prep.). After deposition of the Boom Clay, a continuous sedimentation of more sandy deposits followed, referred to as "Eigenbilzen Formation". After a short interruption, at the beginning of the Chattian, more sand was deposited referred to as "Voort Formation". Later during this time period, a slight tectonic uplift/tilting was responsible for the partial erosion of these sands. The rest of the burial history consists of further depositions of sand layers, from which the most voluminous one is known as the Diest Formation (more than 100 m of sand). These Neogene sand sedimentations finally resulted in the present situation where the Boom Clay in Mol is currently at its deepest level ever, roughly in between 185 and 285 m depth.

Figure 3. Burial history of the Boom Clay at Mol. The position of the HADES underground research facility is also indicated. The history can almost be described as a tranquil burial till the present state.



During the time of the first Voort Sand deposition in Mol it was found that in the outcrop zone, the Eigenbilzen Sand and a large part of the Boom Clay were eroded. During the further burial history at the outcrop location, the clay deposit always remained close to the surface (<50 m). No thick sand layers, as for example the Diest formation in Mol, were deposited in the history of the outcrop area. We refer to the poster for a figure showing the burial history of the Boom Clay in the outcrop zone.

Burial history of the Ypresian Clays

On the presented poster, the burial history of the Ypresian Clays will be illustrated at two locations, in a similar way as is done for the Boom Clay. The Eocene deposits, overlying the Ypresian Clays show an alternation of periods of continuous sedimentation and periods where no significant sedimentation or erosion occurs. This resulted in a more than 100 m thick deposition of sand and clay layers. At the start of the Oligocene, the further burial history follows that of the Boom Clay: in the southern part few sedimentation or even erosion took place, and in the northern part deeper burial, due to the deposition of Boom Clay and Neogene sands, was observed.

Conclusions

From the burial histories of the Boom Clay and Ypresian Clays, it can be concluded that Northern Belgium, where both formations occur at depths of a few hundred meters, was tectonically relatively stable during the past 50 million years. The example of Mol, where the HADES-URF is located, is an illustration of this and shows that the Boom Clay there is at its deepest level ever since it was deposited.

Acknowledgements

This study is part of the Belgian programme concerning high-level and/or long-lived radioactive waste disposal which is under the responsibility of ONDRAF/NIRAS. The authors are very grateful to Prof. Noël Vandenberghe from the department of Historical Geology at the KULeuven for his help on the Palaeogene stratigraphy of Belgium.

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PRESENCE AND EVOLUTION OF NATURAL ORGANIC MATTER IN THE BOOM CLAY

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1. Introduction

Because of its very low hydraulic conductivity, reducing conditions, slightly alkaline pH, high specific surface, high cation exchange capacity and high plasticity, the Boom Clay is studied as a reference host formation for the deep disposal of high-level long-lived radioactive waste (NIRAS/ONDRAF, 1989). However, Boom Clay also contains up to 5% wt. of organic matter (OM). As radionuclides can form complexes with this organic matter, a detailed characterisation and knowledge of the evolution of the organic matter is necessary. An overview of the characteristics of the organic matter present in Boom Clay is given by Van Geet *et al.*, (2003). The solid phase OM can be up to 5%. The dissolved OM fraction is around 200 mg C per liter of Boom Clay porewater. Both kinds of OM will be discussed. Concerning the solid phase OM the focus will be on the past evolution and its possible future evolution due to a thermal stress. For the dissolved OM, the focus will be on its origin.

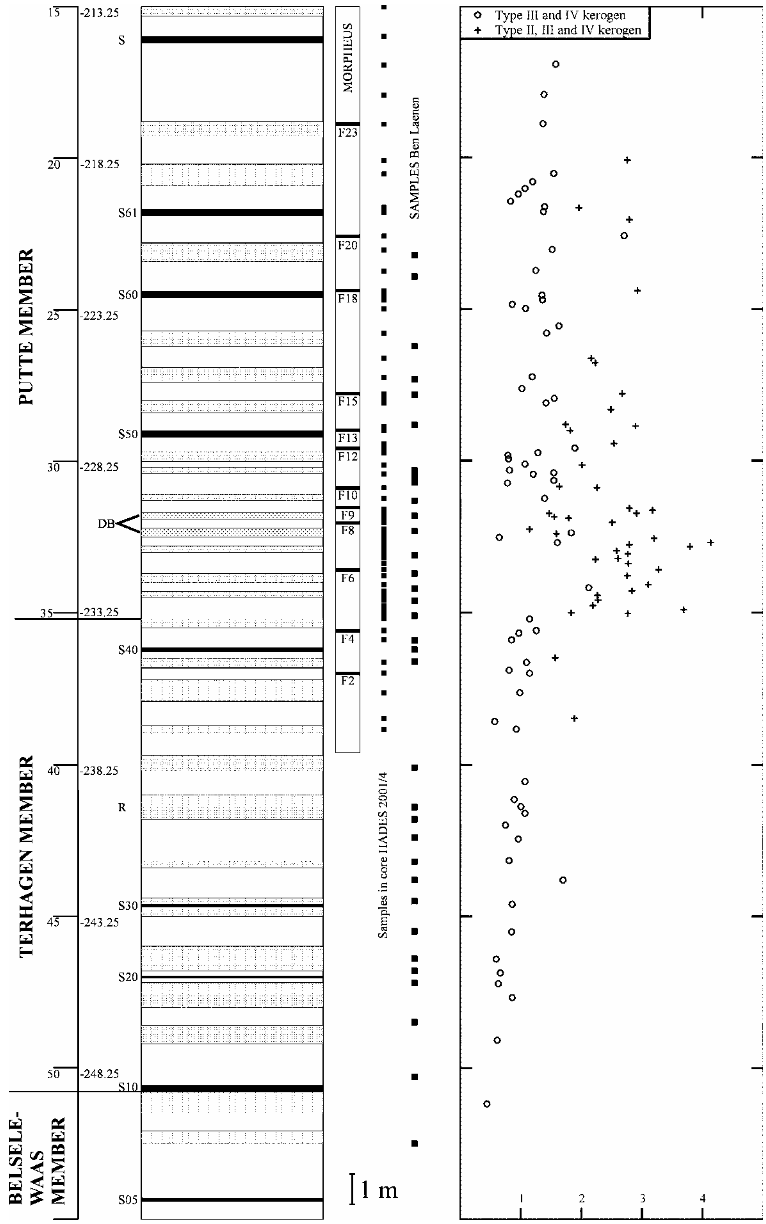
2. Geological background and sampling

Boom Clay outcropping along the river Rupel and slowly dipping to the N-NE, has been studied since 1970 (Vandenberghe, 1978). It shows a very detailed layering, continuous over the whole outcrop area and also observable in cored samples. This layering is caused by an alternation of grainsize (silt-clay), organic matter content and carbonate content. A profile of the organic matter content and characteristics has also been performed on outcrop sections (Laenen, 1997). However, organic matter is very sensitive to oxidation (Landais *et al.,* 1984), so that a comparison on fresh core material is necessary. Starting from the Underground Research Facility (URF) HADES in Mol (Belgium), a core drilling was performed 40 m downwards (core HADES, 2001/4), covering a major part of the outcrop sections. The core was stored under vacuum Al-coated PE bags, which were then kept in an argon atmosphere at 4°C to avoid any oxidation and to minimise microbial growth. The drilled borehole was then used for the emplacement of a piezometer nest (code name MORPHEUS) containing twelve filters, positioned at predefined particular levels with regards to the characteristics of the layers (organic matter, grain size and carbonate content). The dissolved organic carbon (DOC) has been sampled by such piezometers, or by squeezing or leaching of clay material.

3. Solid phase organic matter

Rock-Eval analyses of the core material were very similar to the ones obtained in outcrop (Van Geet, 2002). Figure 1 shows the stratigraphical location of the Boom Clay samples as obtained from outcrop and core HADES 2001/4 sections, together with the total organic carbon (TOC) evolution.

Figure 1. On the left the analysed Boom Clay statigraphy is given together with the depth to the URF and the depth to Second General Leveling in Belgium. On the right the TOC content in function of depth is given.



The bottom axis gives the TOC content in %wt. The markers used divide the samples based on a cut-off value of the Hydrogen Index (HI) of 130 mg HC/g TOC, including data from Laenen (1997) and data on core HADES 2001/4.

The Rock-Eval data clearly showed a bimodal distribution. The first population has TOC values below 2% and is omnipresent throughout the stratigraphically sampled part. This first population contains only Type III and Type IV kerogen (terrestrial organic matter). The second population has TOC values above 2% and is limited to the Putte member. The latter population consists of a mixture of Type II, Type III and Type IV kerogen (marine and terrestrial organic matter). Detailed biomarker analyses by Laenen (1997) on outcrop samples pointed towards the same conclusions. Type IV fragments (strongly oxidised) are believed to be reworked coal fragments from the British Isles. The Rock-Eval data have clearly indicated that the OM present in Boom Clay is immature (T_{max}: 358-429°C, mean: 420°C).

More detailed molecular organic geochemical characterisation of the kerogen has been performed as well. These studies supported the fact that a mixture of marine and terrestrial organic matter is present in the kerogen. However, an extra source of carbon of bacterial origin was recognised as well. From a diagenetic point of view, sulphate reduction seems most important, especially in organic-rich layers. Aerobic microbial biodegradation of organic matter was very limited and probably limited to the reworked organic matter. The most important formation pathways of the kerogen is expected to be degradation – recondensation and oxygen cross – linking (Deniau *et al.,* 2001). Carbonate concretions are present at specific horizons in Boom Clay and are formed by early diagenesis (De Craen, 1998). The concretions have also been studied for their organic matter content (Kiriakoulakis *et al.,* 1994). The difference between the kerogen of the clay and the kerogen of the concretions is not significant. However, the early diagenetic formation of concretions probably lead to a better preservation as higher TOC, higher extractable OM and higher concentrations of most biomarkers were noticed. Biomarkers of bacterial origin were the only with a significant higher concentration in the clay compared to those of the concretions.

During heating at 300°C of the Boom Clay kerogen a weight loss of 25% is noticed, while heating up to 400°C results in a weight loss of 35%. Consequently, a considerable amount of weight loss already occurs at a weak thermal stress. The pyrolysate of both temperatures is further analysed in more detail. Some of the products generated at 300°C come from thermal cracking of weak bonds. Some other products, however, are believed to be organic molecules trapped into the macromolecular structure of the kerogen and liberated by heating. The term kerogen used here comprises a mixture of physically trapped molecules and kerogen *sensu stricto* (macromolecular material insoluble in organic solvents). The trapped molecules might be soluble, but were not extracted during kerogen isolation. They might be trapped physically into the interior of the macromolecular kerogen structure or might be associated to this structure by weak intermolecular bonds, like hydrogen bonds. The presence of such trapped molecules is well described in coal literature (Vahrman and Watts, 1982; Marzec *et al.,* 1983). The trapped molecules in the Boom Clay kerogen correspond mainly to potentially reactive oxygenated compounds

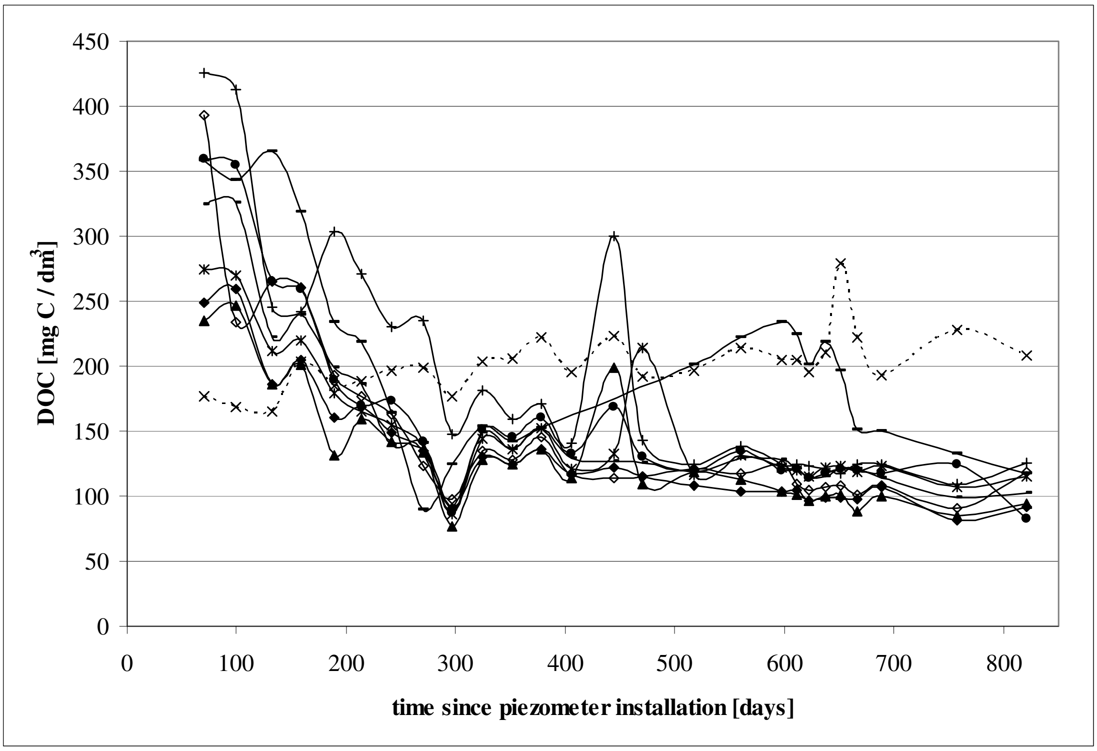
Many pyrolysis experiments in closed system were carried out on the Boom Clay kerogen to simulate the production of effluents (gas and liquid). Under a weak thermal stress there is a relatively important production of gas (up to 30 mg/g kerogen under 250°C/3h). The identification of this gas shows that it corresponds to an exclusive production of CO_2. H_2S and hydrocarbons appear under a thermal stress above 250°C/3h. However their production is definitely lower than the CO_2 production. The liquid production increases regularly with the temperature increases. The identification of the released molecules will be undertaken.

4. Dissolved organic matter

The sampling technique of the pore water (piezometer, squeezing and leaching), clearly has an effect on the dissolved organic carbon (DOC) content measured. We believe that pore collapse during squeezing and aqueous dispersion during leaching causes the variability in DOC measurements possibly due to ultrafiltration effects or modifications of OM during leaching. According to our opinion piezometer water is giving the most representative pore water concerning potentially mobile DOC. Figure 1 shows the stratigraphical distribution of the filters of the MORPHEUS piezometer, mainly used for DOC characterisation. DOC content varies between 100 and 200 mg C/dm^3 and seems to be spatially dependent. However, no correlation between DOC content in the pore water and TOC content of the neighbouring sediment is evidenced. Moreover, an important change of the DOC content in time is noticed (Figure 2). For most filters a steep decrease of the DOC with time is noticed during about 300 days. From 300 to 600 days a less pronounced decrease is noticed to finally end up with a kind of plateau. Superposed on this trend some erratic variation in time is noticed as well. The reason of this variation in time is not clear.

One clay sample was leached sequentially with Synthetic Boom Clay Water (same composition as Boom Clay water, but without dissolved organic matter). The first three extracts show a strong decrease in OM content, while the following extracts only show a small decrease in OM content.

Figure 2. DOC evolution in 10 of the 12 filters of the MORPHEUS piezometer nest



Data of two filters are not included as technical problems occurred during sampling. For one filter a more or less constant DOC is measured (dotted line). This filter is located in zone with relative high pore water fluxes, so that steep decrease might have occurred before the first sampling.

For a more detailed molecular characteristion of the DOC, the latter was extracted by liquid – liquid extraction with dichloromethane (DCM). The obtained extracts contain especially small molecules. However, these fractions showed a strong contamination with plasticisers (NBBS). These plasticisers are probably originating from the piezometer, which was non-metallic and constructed of

PVC. The latter is being checked. To avoid the artefacts of the plasticiser, a test on freeze-dried pore water has been performed as well. The obtained solids, composed of salts and bulk DOC, are then analysed with Pyrolysis GC-MS. The plasticiser can still be defined, but is no longer obscuring the whole profile. Current research focusses on the characterisation of DOC and neighbouring TOC by Py-GC-MS, to check a possible link of origin between both pools of OM.

5. Conclusion

The Boom Clay kerogen makes up to 5% wt. of the total sediment. Rock Eval analysis has shown that the kerogen is a mixture of terrestrial and marine OM and especially that it is very immature. Consequently, an evolution of the OM seems plausible, which should be taken into account for the long-term stability of the Boom Clay as major barrier. The past diagenetic evolution of OM seems to be very minor as illustrated by comparison of the molecular characteristics of the kerogen present in the clay and present in early-diagenetic concretions. A weak thermal stress of the Boom Clay kerogen also revealed that some smaller oxygenated molecules are entrapped into the 3-D macromolecular network of the kerogen. Some first results on the quality and quantity of effluents released during thermal stress will be presented as well.

The Boom Clay pore water contains around 100 to 200 mg C/dm^3, but this value might be as high as 400 mg C/dm^3. Up to now, the DOC cannot be linked to the TOC of the neighbouring sediment as no correlation is observed in their concentrations. However, a more detailed molecular analysis of the dissolved organic carbon is going on. The DOC content is clearly decreasing in time until some kind of plateau is reached. Such a rapid decrease in DOC was also noticed in sequential leaching experiments. A possible microbiological effect cannot be excluded.

Acknowledgement

This work is performed in the frame of the National Belgian Waste Management Program supervised and financed by NIRAS/ONDRAF. The European Commission (EC) is also gratefully acknowledged for its financial support in the frame of the Trancom-II Project – Transport of radionuclides in a reducing clay sediment – phaseII, contract N° FIKW-CT-2000-00008. I. Deniau thanks IRSN for financial support for her PhD thesis.

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UNCERTAINTY PROPAGATION IN A DETERMINISTIC SEISMIC HAZARD ASSESSMENT

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[1]GEO-TER SARL, and [2]Andra, France

Introduction

The objective of this study was to give to Andra an estimation of the seismic ground motion on surface and at depth in the 150 million year-old clay formation investigated on the Meuse/Haute-Marne underground laboratory site located in the eastern part of the Paris Basin (France).

The estimation of this ground motion must correspond to the different time periods involved for a long-lived radioactive waste geological repository: "exploitation" period covering the next hundreds of years (called "short time") to post closure time, covering the next 1 million years (called "long term"). The method proposed here to achieve this objective is original due to the simultaneous treatment of the epistemic uncertainties and the random uncertainties of seismic activity, very large for the estimation of maximal possible earthquakes during the long post closure time in a tectonically stable area like the Paris Basin.

The first ones are associated to present knowledge about the local and regional geological structures and their deformation modes and rates, witch allow to construct different plausible seismotectonic models. They are propagated using a technique of logic tree. The second ones are related to the input parameters (magnitude, depth and attenuation law) in a deterministic seismic hazard assessment and they are treated, in the calculation process, by using a Monte-Carlo technique.

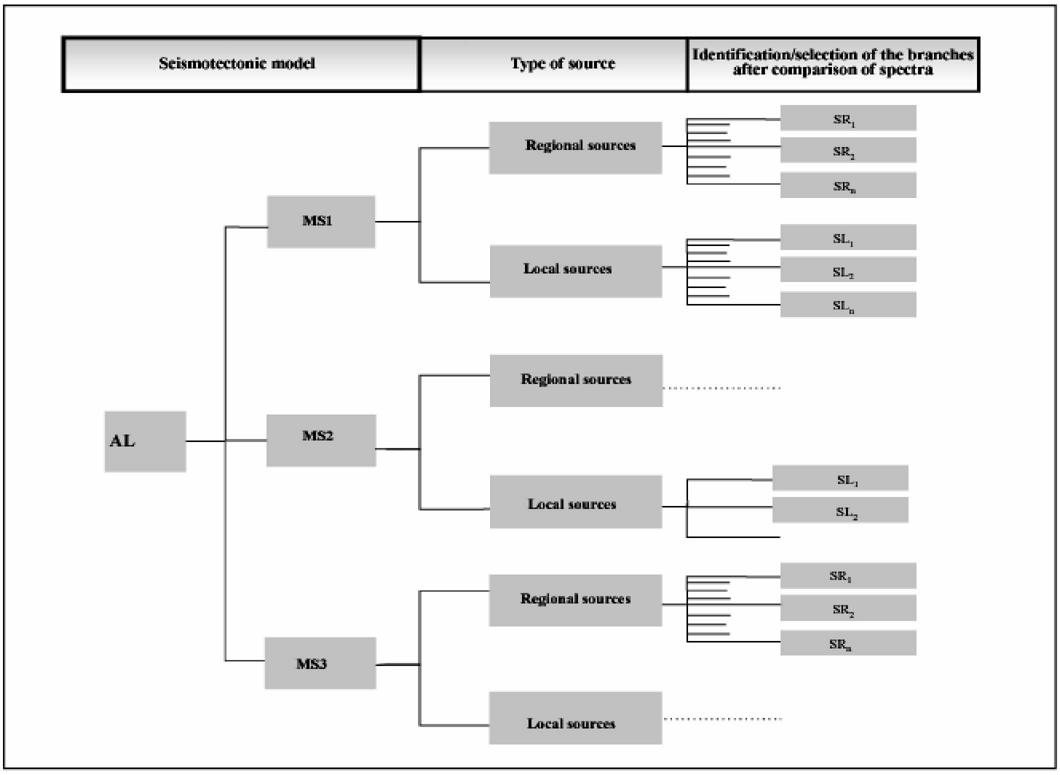
The results obtained correspond to elastic response spectra, on surface and at depth, in the Callovo-Oxfordian argillaceous layer.

Method of construction of the logic tree

The method of construction of the logic tree developed in this study is shown in the following graphic with three input seismotectonic models (MS1, MS2 and MS3). Their treatment leads us to the consideration of regional and local sources characteristics of each model. An equal logic three to that shown in the first figure is built for each level of earthquake analysed: short time (SMHV, SMS) and long time (SMPP). In each seismotectonic model considered, we identify two types of branches corresponding to:

- The local sources, which will be responsible of ground motions richer in the domain of high frequencies.
- The regional sources, contributing to ground motions richer in the domain of low frequencies.

When all branches corresponding to a type of source are identified, we carry out a first selection of those branches SRi and SLi, that we have to retain to perform the ground motion determination for the site. These sources correspond to those that lead to a dominant ground motion on the site, in the interval of frequencies 0.1 to 34 Hz.



Methodology of response spectra on surface determination

The determination of the spectra is performed following different phases:

Phase 1: Generation of 100 spectra per branch to propagate the random uncertainties using a Monte-Carlo technique. To propagate the uncertainties associated to the input parameters representing the magnitude, depth and attenuation law, a Monte-Carlo method has been used. The probability distribution used to perform the random generation of parameters has been:

- The truncated gaussian distribution (to one standard deviation) for the attenuation law parameters.
- The uniform distribution for the magnitude and for the depth. This probability distribution has its limits in the mean value of magnitude and depth augmented and reduced one standard deviation.

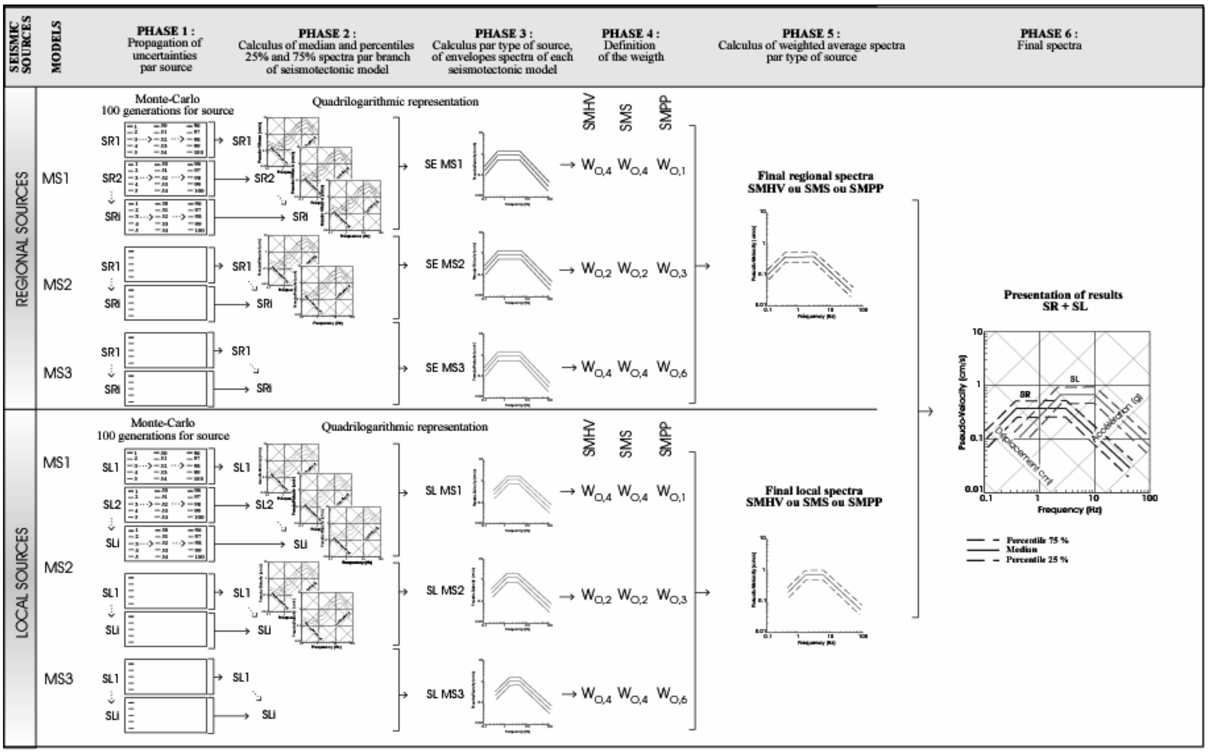
Phase 2: Calculus of median and percentiles 25% and percentiles 75% spectra per main branch of each seismotectonic model. We calculate, for all spectra periods, the median, percentile 25% and percentile 75% values of spectral acceleration. This process allows to quantify the uncertainty on each branch and justifies the use of the Monte-Carlo technique. The process described is used in each of the branches of the seismotectonic models of each logic tree.

Phase 3: Determination per type of source, of envelopes spectra of each seismotectonic model. The RFS I.2.c recommend retaining, on the site, the most penalising ground motions for each spectral period. Therefore, some sources of one type, SRi or SLi, and belonging to same model (MS1, MS2 or MS3), have to be considered and the construction of the envelope spectra is needed. For each frequency, we identify the greatest values of seismic ground motion. This process is repeated with median, percentile 25% and percentile 75% spectra. The envelope spectra and their uncertainties are calculated par type of source of each seismotectonic model (MS1 to MS3), and for each logic tree (SMHV, SMS and SMPP).

Phase 4: Definition of the weight. A weight is associated to each model sismotectonic, MS1 to MS3.

Phase 5: Determination of weighted average spectra per type of source. Here, we have to find the weighted average of median and percentiles spectra of each seismotectonic model applying the weights defined in the phase 4.

Phase 6: Determination of final spectra for the conception and design of the installations. The final spectra proposed for the conception and the design of installations are smoothed spectra. The values of acceleration, velocity and displacement are given at the characteristic frequencies of the spectra.



Methodology of spectra at depth determination

Two methods have been retained to define these movements:

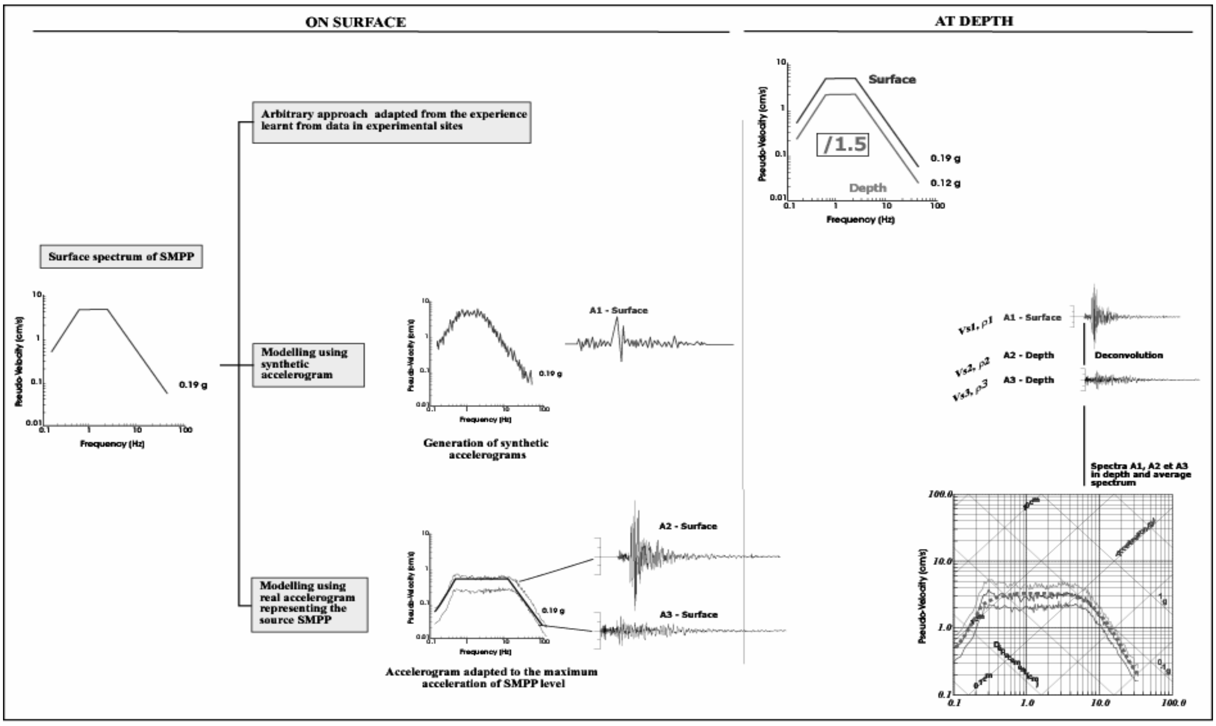
- Deconvolution of the seismic ground motion predefined on surface.
- Analyse of the knowledge obtained from data records in underground laboratories and from attenuation laws defined at depth.

The first method is based on a technique of deconvolution of the seismic ground motion defined on surface, with the transfer function corresponding to the geological profile from the surface to the substratum. This method is based on:

- The definition of some accelerograms (synthetics and naturals) representing of the response spectra on surface. It's needed to consider some accelerograms to take into account the variability of ground motion.
- The determination of the accelerograms and response spectra at depth using a deconvolution process of the accelerograms on surface, and using the transfer function of the geological column of the site. This deconvolution is made using a validated software in seismic engineering (PROSHAKE and EERA).

The second method used is based in the knowledge obtained from the simultaneous recordings on surface and at depth made in some pilot sites. It is well known that the effects at depth are, for the same earthquake, less damaging at depth than on surface. This effect comes from the fact that the ground motions at depth are weaker than on surface.

After the revision of some studies, the application of a dividing factor to all frequencies of the surface response spectra of 1.5 allows to define a first approach of the ground motion at depth. The application of this coefficient is in the sense of the security because the acceleration obtained with its use is always equal or greater than the observations available until now.



Conclusions

The approach is original due to the simultaneous treatment of epistemic uncertainties, associated to the present knowledge on the geological structures, and random uncertainties, associated to the input parameters of spectra (magnitude, depth and attenuation law) in the scope of the deterministic seismic hazard assessment. Their propagation inside the method allows to extract and to quantify the uncertainty associated to the median seismic ground motion in surface. A methodology of determination of response spectra at depth has also been proposed.

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GEOLOGICAL DISPOSAL OF SPENT NUCLEAR FUEL IN CLAY HOST FORMATION IN SLOVAKIA

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Introduction

The primary objective of a geological repository is the long-term isolation of radioactive materials from the human being and surface environment to ensure that radioactive decay will be greatly reduce to the acceptable level of activity.

Geological disposal in argillaceous media is being considered as viable option in many countries with radioactive waste management. In order to determine the suitability of this rock for deep disposal, it is necessary to evaluate migration of the radionuclides in such an environmental system.

Site selection process

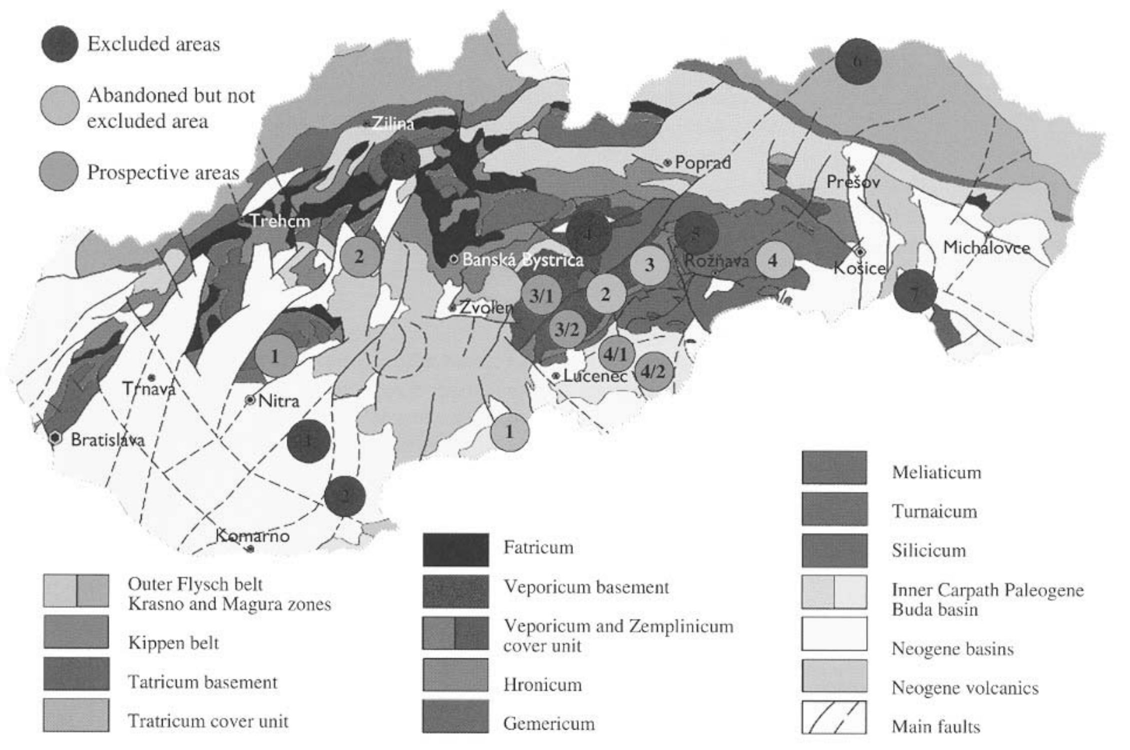
The site selection process in Slovakia started in 1996. The critical review which included a survey of published and archival regional geology, hydrogeology, engineering, and geophysical data led to identification of 15 perspective areas. In Figure 1 are these areas divided into three groups: excluded, abandoned but not excluded, and prospective areas. Based on the results, four distinct areas in six localities were determined as perspective for the construction of deep geological repository. Four localities are situated in granitoid and two in argillaceous rocks. The selection of candidate localities is expected around 2010.

Concept of geological disposal

According to the current status of the geological disposal in the Slovak Republic, the concept foresees direct disposal of spent nuclear fuel and high level waste into the repository without reprocessing. The concept is similar to that considered in other countries, being based on a system of multiple passive barriers consisting of engineered barrier system (waste form, canister, buffer and backfill) and geological formation. The disposal of spent fuel elements is assumed to be in carbon steel canisters, arranged vertically in boreholes, filled up with bentonite and surrounded by the clay layer. Cylindrical geometry of barriers is considered. The source term consists of 24 290 fuel elements, with average burnup of 45 MWd/tU, with enrichment 3.82% pull up from reactor (type VVER 440) after 4 years of cooling. Decay chains and radioactive decay are considered. The computer code ORIGEN2.1 was used to calculate the inventory of the spent fuel. [2] The repository is located at the depth of 300 m. The only way for the radionuclides to reach the aquifers is by migration and diffusion through the interstices of the clay formation.

The design of the underground facility is defined in consistency with the results of a thermo-mechanical study. [3] According to the thermal calculations, the maximum temperature at the interface container/backfill does not exceed the value of 100°C. The distance between the centres of canisters is considered to be 6.3 m.

Figure 1. Status of deep geological repository siting in 2001. Red circles mark excluded areas, orange circles mark abandoned but not excluded areas, green circles mark prospective areas [1]



Modelling and calculations

Radionuclide migration in the deep geological environment was simulated as a one-dimensional model with the computer code GoldSim for the timescale of one million years.

It is assumed that after sealing of the repository natural degradation of canisters will occur and induce the release of radionuclides. After the canister failure, the groundwater reaches the waste and radionuclide release starts. The clay is assumed to be homogeneous, porous and saturated with water. The host clay is also low conductivity and diffusive transport dominates, although low advection in aquifer layer is considered. The properties of the clay formation are given in Table 1. The processes being considered are [4]:

- diffusion;
- dissolution;
- precipitation;
- sorption;
- radioactive decay and ingrowth.

Table 1. Properties of the clay formation

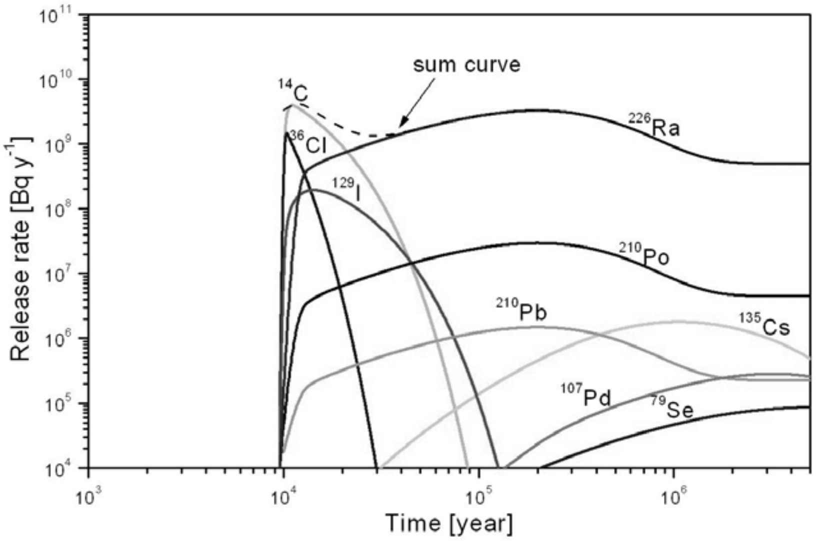
Parameter	Value
dry density of clay	1 700 kg m^{-3}
water content	12%
porosity	30%
tortuosity	0,6
diffusion coefficient	10^{-9} m^2 s^{-1}
hydraulic conductivity	10^{-10} m s^{-1}
groundwater velocity	2 cm y^{-1}

Doses are calculated for three different biosphere pathways according to the [5]: drinking water pathway, root vegetable and meat, using dose conversion factors for ingestion, specific for adults.

Results

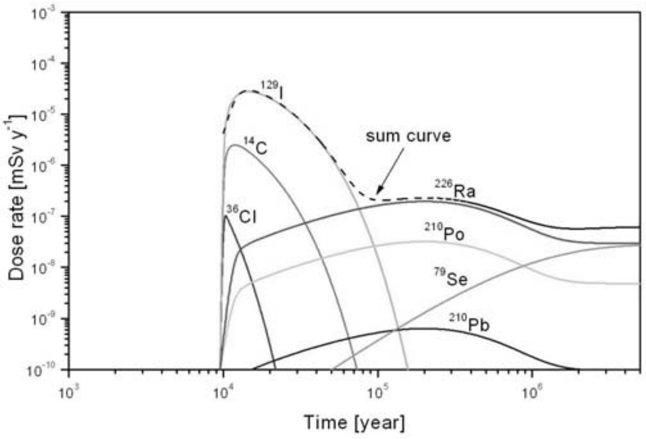
The release rates from the clay layer as a function of time are given in Figure 2. All dose curves rise at 10^4 years, at time of canister breaching. The sum curve has two maximums. The early one occurs at 1.1×10^4 years, very shortly after the canister breaching. The main contribution to the peak is due to ^{14}C, ^{36}Cl and ^{129}I. The second maximum occurs at 2×10^5 years due to ^{226}Ra.

Figure 2. Release rates from the clay layer as a function of time



The individual dose rates as a function of time are illustrated in Figure 3. At early stages, the most important radionuclides are ^{129}I, ^{14}C and ^{36}Cl. The maximum dose rate of 2.8×10^{-5} mSv y^{-1} is mainly due to ^{129}I and it occurs at 1.7×10^4 years. Later lower peak with dose rate of 2×10^{-7} mSv y^{-1} occurs at 2×10^5 years and is due to ^{226}Ra and ^{210}Po.

Figure 3. Deterministic estimate of individual dose rates



Conclusion

The results shown that the majority of radionuclides which would release from the waste packages will decay and sorb within the bentonite buffer and clay. Only a few highly soluble and low sorbing radionuclides, namely ^{129}I, ^{14}C, ^{36}Cl, ^{226}Ra, ^{210}Po, ^{79}Se and ^{210}Pb penetrate the clay barriers to an extent whereby the calculated doses exceed 10^{-10} mSv y^{-1}. The sum dose maximum is bellow 10^{-4} mSv y^{-1}.

Results of calculations indicates that the clay formation can ensure the long-term protection for a spent nuclear waste repository, provided that the clay has a sufficient thickness and care is taken in characterising the overlying aquifers.

The present results have only preliminary character. The performance assessment calculations will be extended and completed in later stages of the geological disposal process, depending on the quality of available extent of data and depth of understanding of the repository system as whole.

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CHARACTERISTIC PROPERTIES AND CM(III) COMPLEXATION OF HUMIC AND FULVIC ACIDS FROM CALLOVO-OXFORDIAN AND OPALINUS CLAY

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1. Introduction and context

Clay is foreseen both as back-fill material and host rock for nuclear waste disposal. It contains organic matter that can be released and form dissolved organic matter (DOC). Part of this DOC consists of humic and fulvic acids. Humic substances in natural water are present in the form of humic colloids, consisting of the organic entities, associated mineral structures and complexed metal ions. These humic colloids can play a major role in the radionuclide migration in natural aquifer systems. [1] Cement may be present in a nuclear waste repository as a waste form or as part of engineered structures. In case of water intrusion, cement dissolution will, amongst others, lead to high pH values (initial pH>13). On a short time scale, a minor fraction (few percent) of the clay organic matter is dissolved. With prolonged contact time with alkaline solution, the hydrophobic clay organic matter becomes chemically converted and a large portion (up to around 50% after about 1.5 years) is forming hydrophilic humic and fulvic acids. [2] In this paper, humic and fulvic acids initially released from Callovo-Oxfordian and Opalinus Clay under near-field high pH conditions are quantified and characterised. Furthermore, their complexation with Cm^{3+} is investigated. The humic and fulvic acids are characterised by asymmetrical flow field-flow fractionation (AFFFF) and near edge X-ray absorption fine structure (NEXAFS) spectroscopy. The latter technique can offer practically the same level of information as nuclear magnetic resonance (NMR) [3] but require very low amount of samples (~10^{-3} mg). The complexation with the Cm^{3+} ion is studied by time resolved laser fluorescence spectroscopy (TRFLS). It is shown that humic and fulvic acids may result in enhanced radionuclide migration in clay under cement dissolution conditions.

2. Method and results

2.1 Clay samples

Callovo-Oxfordian clay samples from three different depths from the boreholes EST 104 samples (447, 494, 516 m below surface) and one Opalinus shale sample (579.19-579.45 m) are studied. The origin of the clay organic matter, identified by biomarkers, in the Oxfordian series (447 m depth) is mainly of terrestrial origin, whereas organic carbon in the Callovian (494-516 m) series is mainly of marine origin. [4]

The total organic carbon (TOC) concentration in the Bure clay is ~1.3 wt% (independent of depth) whereas only <0.4 wt% is found in the Opalinus clay sample. [5, 6] Humic and fulvic acids are extracted using slightly modified International Humic Substances Society protocol. The different amounts extracted are given in Table 1. No humic acid was found in the Opalinus clay sample.

Table 1. Total organic carbon (TOC) in clay samples and the fraction of TOC extracted as humic acid (HA) and fulvic acid (FA)

Sample*	TOC (weight %)	HA or FA (% of TOC)	Sample*	TOC (weight %)	HA or FA (% of TOC)
447 HA/FA	1.4	5.2/1.3	516 HA/FA	1.4	7.1/0.5
494 HA/FA	1.4	5.2/1.2	OPA FA	0.4	1.1

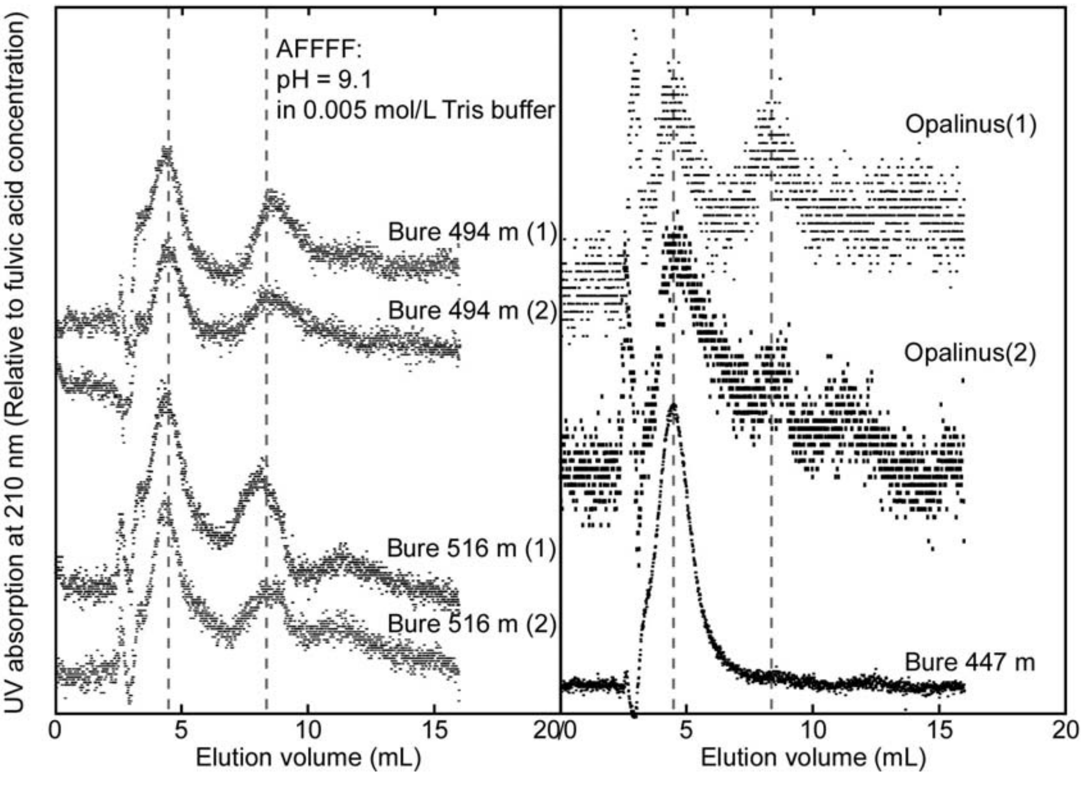
* 447, 494 and 516: Respective depths of samples from the Bure Site. "OPA FA": fulvic acid extracted from Opalinus Clay.

2.2 Characterisation of humic and fulvic acids

2.2.1 Asymmetrical flow field-flow fractionation

Fulvic acids are characterised by the size distribution at pH 9.1 in 0.005 mol/L Tris buffer. The size distribution of the sample 447 m from the Bure Site shows the typical features of fulvic acid. The other Bure samples of marine origin, contrary to the sample from 447 m depth of terrestrial origin, show a bi-modal distribution. The first peak with the smaller particles agrees with fulvic acid in general and also with the fulvic acid from the fulvic acid from 447 m depth (terrestrial origin). The second peak shows larger particles that of atypical size for fulvic acid. The same bi-modal distribution is found for the fulvic acid from the Opalinus Clay.

Figure 1. AFFFF of fulvic acid samples (parallel samples)



2.2.2 *NEXAFS*

NEXAFS spectra were obtained using scanning transmission X-ray microscopy (STXM) performed at the beam line X1A (NSLS) operated by the State University of New York at Stony Brook. The principle of the method is described in [7]. For the comparison all NEXAFS spectra were baseline corrected and normalised prior to peak fitting. The spectra were then deconvoluted following the protocol described in [3]. The resulting distribution of carbon in different functional groups is given in Table 2.

Table 2. Distribution of carbon as different functional groups determined by NEXAFS

	% carbon in functional structures					
Sample*	Quinone	Aromatic	Phenol	Aliphatic	Carboxyl	Carbonyl
447 HA	12.7	16.0	4.0	15.0	24.6	17.7
494 HA	10.3	12.7	10.8	16.9	29.4	19.9
516 HA	11.5	15.2	12.2	16.8	25.0	19.4
447 FA	9.4	16.5	12.8	17.0	27.2	17.1
494 FA	9.0	12.4	10.1	18.5	30.7	19.3
516 FA	4.1	6.4	5.0	26.8	36.3	21.3
OPA FA	0	8.7	0	30.9	36.7	23.7

* 447, 494 and 516: Respective depths of samples from the Bure Site. "OPA FA": fulvic acid extracted from Opalinus Clay.

In all samples, the largest individual peak is from carboxylic groups. This peak area is also bigger for fulvic acids compared to the humic acids. The composition of humic material is related to the originating material and history. [8] The humic acids show no strong trend with the differences in origin. For the fulvic acids, however, aliphatic, carboxyl and carbonyl functional group increase with depth, with a corresponding decrease in the other groups. This reflects differences in marine and terrestrial originating organic material. [9, 10] The Opalinus fulvic acid has a comparably low content of oxygen containing functional groups. This reflects the history of this sample, namely higher burial temperature compared to the Bure samples (80°C instead of 40°C [4, 11]) which may lead to deoxygenation processes and possibly also polymerisation.

The relative peak areas of functional groups correlate between 13C-NMR and NEXAFS. One exception are carboxylic groups. [3] Therefore, the absolute amount of carboxylic groups cannot be determined directly. The relative carboxylic peak area of typical fulvic acids of aquatic origin (from different types of Gorleben groundwater) is found to be 41.9 ± 1.3. [3] In the present samples lower numbers are found and thus the carboxylic group content is lower.

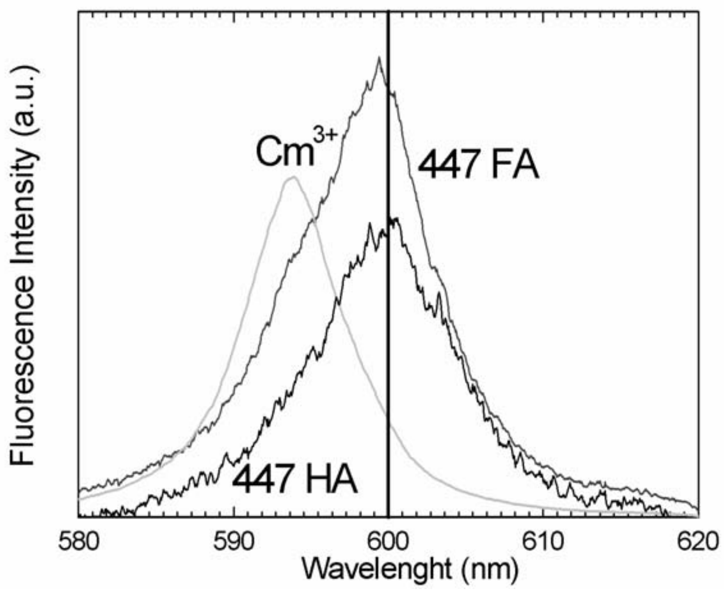
2.3 Complexation behaviour

The complexation behaviour was studied using time resolved laser fluorescence spectroscopy (TRFLS). By this method metal ion speciation can be done with respect to both emission band shape and fluorescence decay behaviour. Investigations were conducted at $5.7<pH<6$ to avoid metal ion hydrolysis and thus results can be compared to a large number of published results. 0.2 µmol/L of Cm were added for the three different humic acids, whereas 0.1 µmol/L was used for fulvic acid samples. The emission spectra show the characteristic shape of humate and fulvate curium complexes with peak maximum around 600 nm, compared to 593 nm for the non-complexed Cm^3+ ion (Figure 2). [12, 13] The fluorescence emission decay also shows the same timely behavior as other humate and fulvate complexes.

3. Summary and outlook

Humic and fulvic acids are released from Callovo-Oxfordian and Opalinus Clay under alkaline conditions. With progressing time, their amounts increase by chemical conversion of hydrophobic clay organic matter. They lead to the generation of the same curium(III) complexes as typical humic and fulvic acids. This provides the potential for enhanced radionuclide transport under near-field cement dissolution conditions. The characteristic properties of the present humic samples show somewhat different behavior with respect to the comparably low carboxylic group content, and oxygen containing functional groups in general, as well as the presence of a fraction with atypically large entities (with exception for fulvic acid from clay of terrestrial origin). This leads to the expectation that their effective ligand concentration is lower than for rather typical humic and fulvic acids. Further analysis of data and additional investigations are required in order to quantify the potential radionuclide transport enhancement.

Figure 2. Emission spectra of humic and fulvic acid samples from Bure sample at 447 m depth
For comparison, the spectrum of the non-complexed Cm^3+ ion is also shown



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CONFINEMENT PERFORMANCE OF BODA CLAYSTONE FORMATION, HUNGARY

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Preface

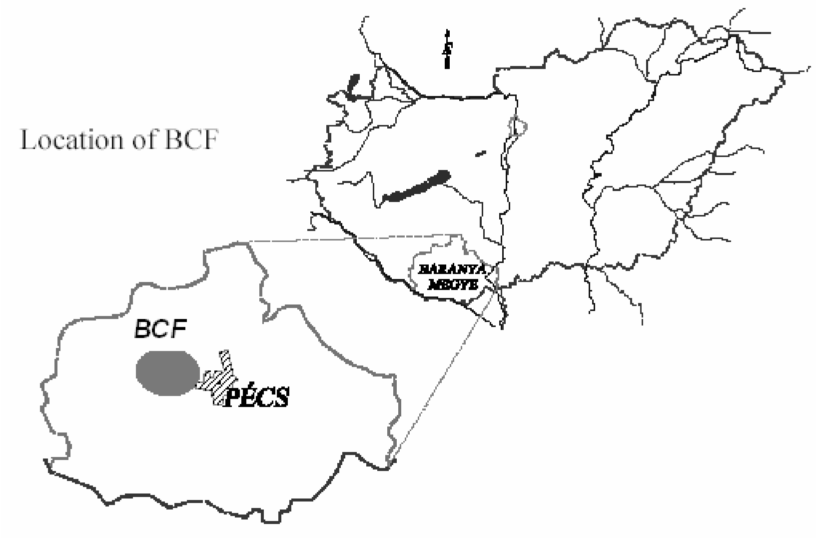
Paks Nuclear Power Plant (Paks NPP), with its four VVER-440 reactors, generates approximately half of the nation′s electricity (40% of the total consumption). Therefore it is indispensable that the country should have a secure spent nuclear fuel (SNF) and radioactive waste disposal route. In Hungary the currently available disposal capacity ensures the disposal of institutional wastes for many decades, but for low and intermediate level waste (L/ILW) produced by the nuclear power plant a new facility should be built. Disposal of operational and decommissioning LLW and short-lived ILW is planned on the same site and in the same depth. Long-lived ILW is anticipated to be disposed together with high level waste (HLW).

The SNF generated during the operation of Paks NPP were formerly transported for reprocessing to the Soviet Union and later to Russia with no waste return to Hungary. Due to the altered political situation in Russia, Hungary had to re-evaluate its back-end strategy. As an interim solution a Modular Vault Dry Storage facility (MVDS) has been commissioned on the Paks NPP site.

Final decision concerning the back-end strategy has not been made yet. The European integration efforts of Hungary require the application of internationally accepted methods in handling of these problems critical both from professional and political aspects. Almost all the professionals agreed that disposal of one part of HLWs (the type and quantity depends on the option selected) in deep geological formations will be required as the last step of each of the possible options.

Surface and Underground Research Laboratory (URL)-based siting for HLW disposal in Hungary has been focusing on a Permian mudstone called Boda Claystone Formation (BCF) since 1993. It is situated in SW Hungary to the west of the city of Pécs (see map). Detected extension of this candidate host formation exceeds 150 km^2, its maximal thickness is between 700-900 metres. The customers of this exploration were the Paks Nuclear Power Plant Ltd. and from July 1998 the Public Agency for Radioactive Waste Management (PURAM). The Mecsek Ore Mining Company (MÉV) and its successor the MECSEKÉRC Environment, a professional co-ordinating organisation, undertook the tasks of planning, documentation and, implementation together with the subcontractors.

BCF is at regional level well known due to the high quantity of information about its stratigraphic, structural and geological anamorphotic relations of the Western Mecsek Mountains as well as the geological, mineralogical, structural, hydrogeological and geotechnical features of the overlying formations.



Background

The exploration was conducted between 1989 and 1992, and the “Alfa” Project provided a great progress in learning the characteristics of BCF. In the summer of 1994 the underground excavation of the formation was completed using the favourably located tunnels of the uranium ore mine at the average depth of 1 050 m.

Prior to the short-term programme (STP) BCF was crossed at a length of 4 680 m by surface drills and 530 m by underground drills with full core sampling. The rocks of the formation were excavated at 1 050 m underground level by a 90 m long tunnel. Additionally, several previous surface geophysical profiles of the area provided useable structural-geological information. Figures 1 and 2 show the geological map and the geological profile of the BCF.

After an initial phase without considering international references, during the planning and implementation of the different phases of the exploration programme the internationally accepted investigation and assessment methods were increasingly applied taking the proposals of international projects supported by IAEA, EC and the OECD/NEA into consideration that was granted by the involvement of foreign companies having reliable experience in this field.

The basic principles of international consensus were generally taken into consideration in the Western Mecsek Programme. However, some of the exceptions shall be highlighted:

- Since there was no progression in the (at least concept level) planning of waste form and the engineering barrier system during the STP, in this phase of explorations we could exclusively investigate the geological suitability.
- Due to the lack of certain basic data the exploration programme was not controlled with safety analysis.
- Due to the financing and scheduling problems comprehensive quality-control (in the classic sense of the expression) and quality assurance programme for exploration activities have not been compiled up to now.

In the course of planning and implementation of STP we attempted the as thorough realisation of the following working methods as it was possible:

- The superposition and iterative extension of databases generated by subsequent exploration phases.
- Mutual information exchange and assessment together with the exploration sub-programmes.
- Collecting of data set suitable for statistical evaluation and their recording in electronic data files for ensuring easier and more effective data processing.
- Application of available state-of-the-art investigation, assessment and implementation technologies.
- Accurate recording of conditions of examinations and investigations.

The activities aiming at the qualification of BCF can be carried out in a very advantageous exploration situation due to the good geometry. The entire depth range between the surface outcrop and the deep level exploration tunnel can be examined simultaneously in its connection ensuring the acquisition of the maximum set of information in this way. In longer term the spatial variability of various parameters can be quantified. Through the application of different surface and drilling investigation methods the exploration was extendable towards the underlying rock of BCF.

Figure 1. Geological map of the Western-Mecsek Anticline with the location of URL and surface outcrop area of BCF

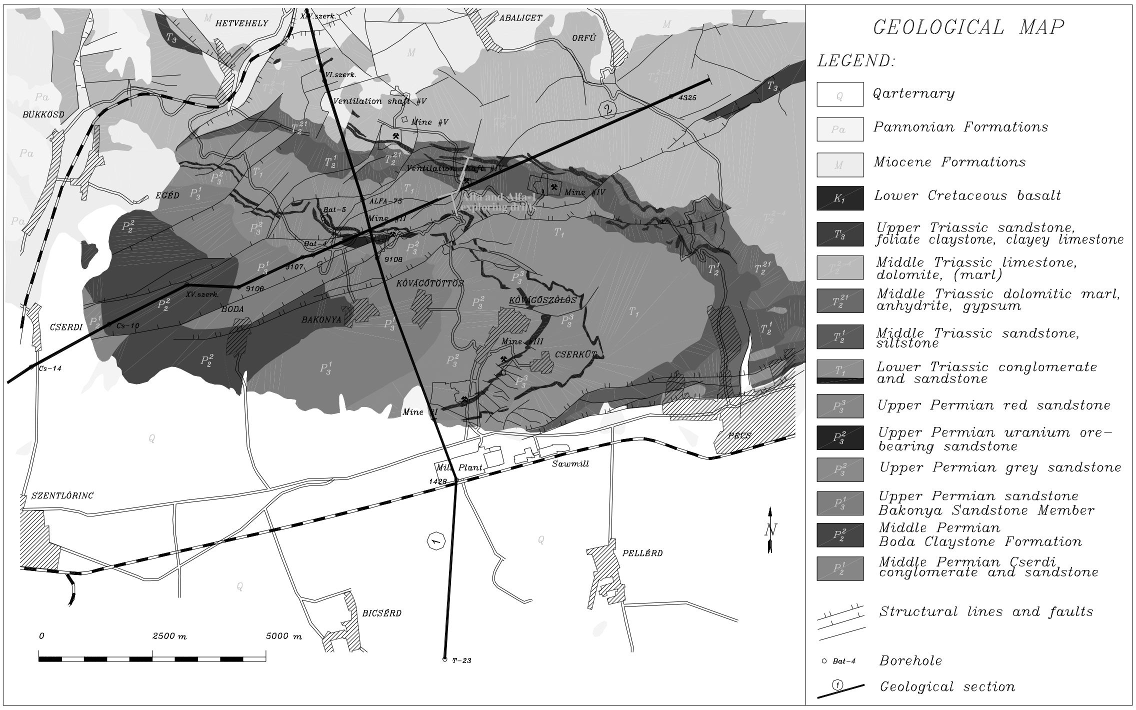
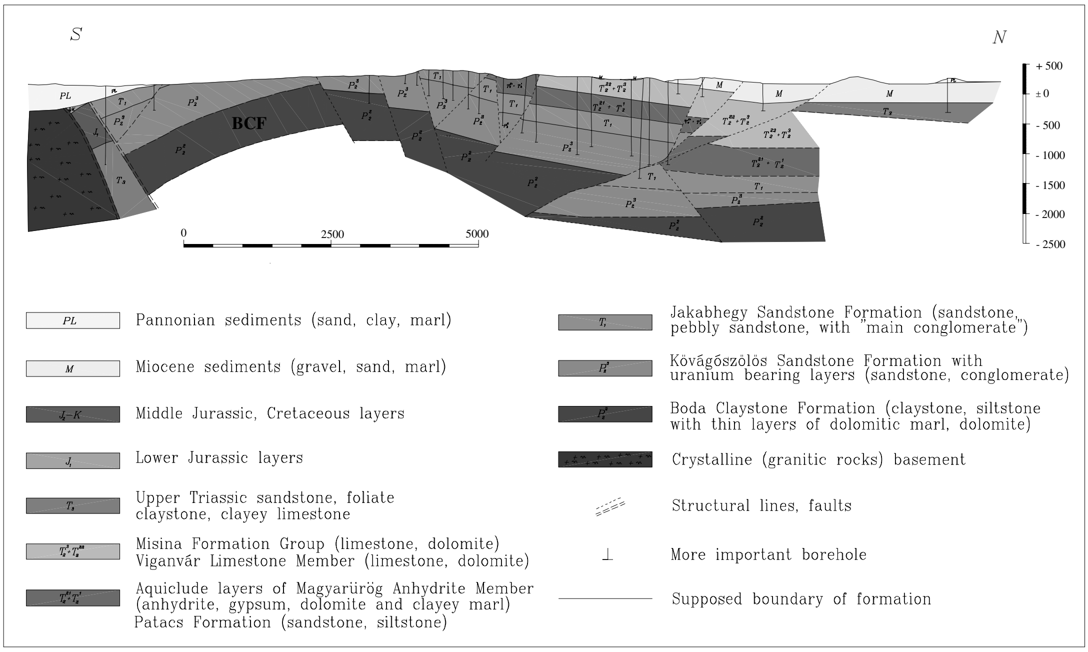


Figure 2. Geological profile of the Northern-Southern direction in the Western-Mecsek Anticline



Safety concept of BCF as a potential host rock for final disposal

The followings can be stated briefly concerning the confining performance and long-term stability of BCF.

Tectonic history

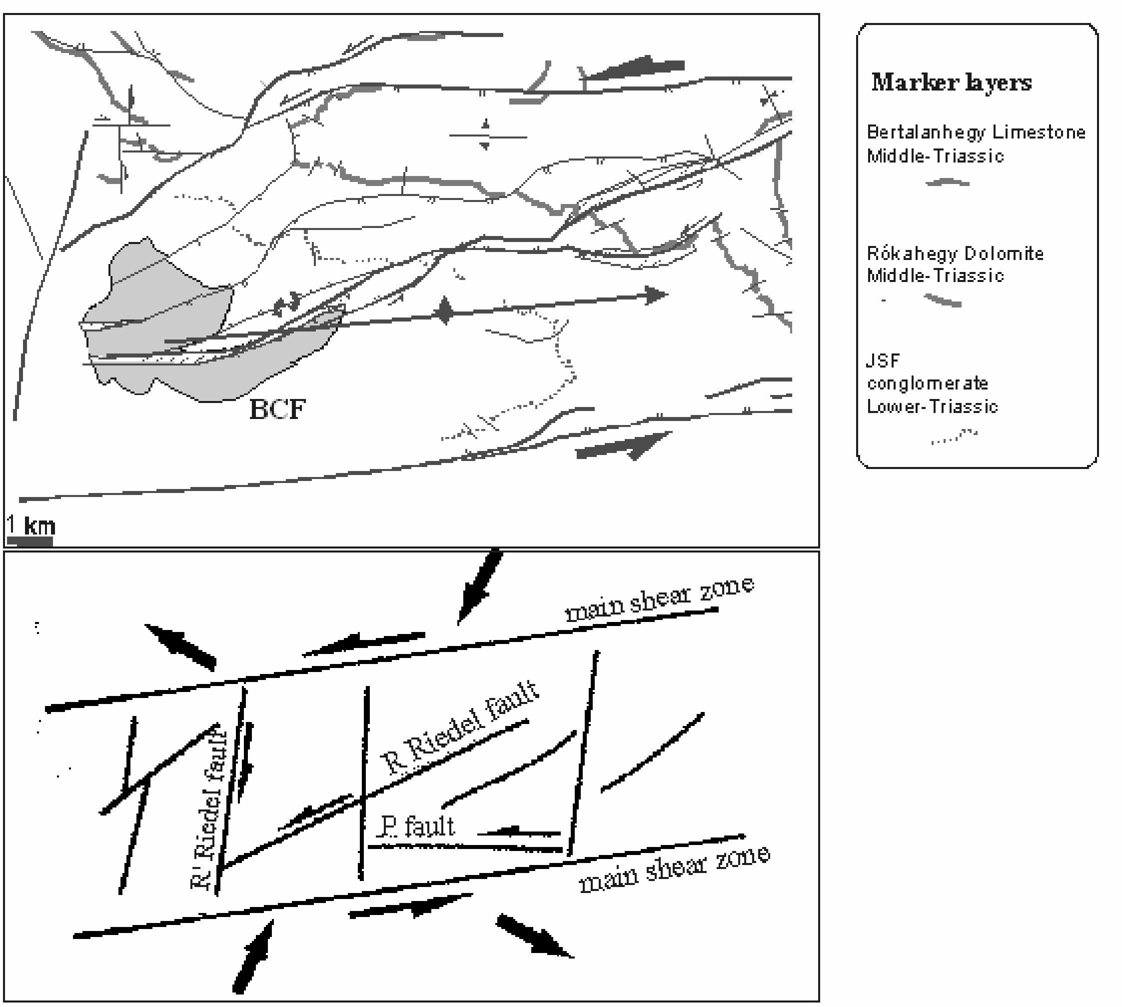
The recent 700-1 000 m thick layers of BCF were settled in an alkaline (or playa) basin under extreme climatic inflow and geochemical conditions, and later they were buried to at least 3.5 to 4.5 km depth. The diagenesis of sediments occurred at high temperature (approx. 150 to 200°C) and at high pressure (120-150 MPa). This situation resulted in the present overconsolidated, highly indurated character of BCF. Bulk-porosity and hydraulic conductivity of intact rock-matrix is very low. The typical interval of Young-modulus is between 30-40 GPa and the average unconfined strength exceeds 100 MPa. Of course, these mechanical properties are not favourable for primary, stress-controlled self-healing processes.

Based on several information Relicts of four differently featured tectonic periods have been discovered so far (see also Figure 3):

- At the beginning of the Cretaceous period (more precisely about 100 million years ago) the rock series of the Mecsek Mountains was molded by the intense N-NE direction overthrusting movement of the Villány and Dráva geological units. This resulted in the formation of flexures originally with E-W orientation, such as the Kővágószőlős anticline. Within the anticline fracture systems being codirectional with axle of flexures formed due to these movements.

- Left-cutting shearing action at the end of the Cretaceous period producing belts with NE-SW strike direction. This deformation cuts through the tectonic elements described above and partly rearrange them.
- Right-cutting shearing action along the Mid-Hungarian tectonic belt in the Oligocene. This phase did not have significant effect on the investigated areas it caused twist in NE direction and dextral dislocation only in the W, NW part of the Mecsek Mountains.
- The youngest system is the horizontal compression phase with N-S (NW-SE) axis molding the Miocene basins and sediments at the end of the Miocene epoch, in the Pliocene epoch (6 up to 7 million years ago). Some scientists mention sinistral dislocations, too, concerning this event.
- The described tectonic events will be probably suitable for the correlation at each scale with those tectonic and micro-tectonic events which were described during the former exploration phases of BCF.

Figure 3. The tectonic style and the simplified model of the W-Mecsek Mts. (Konrád, 1999)



The phenomena of self-healing (sealing) in the Boda Claystone Formation

The process of self-healing and self-sealing, observed in various soft and younger age clay formations and in over-consolidated siltstones and claystones alike is well known and widely reported. The process of self-healing (sealing) has been associated to and explained by closure of narrow aperture joints due to high horizontal stresses on one hand and with the formation of carboniferous or siliceous infillings on the other hand. The self-healing of joints as the result of primary, locked-in, stresses is explained and clearly understood. On the other hand, the process of self-healing by infill formation is a more complicated process. The components of infill forming materials could be originated either in the argillaceous host formations or could be transported in through water filtration. Figure 4 shows the lithological column of BCF.

The formation had been affected by tectonic events mostly under the self-healing depth. Owing to the paleo-self-healing processes, most of the explored discontinuities of tectonic (and lithological) origin are entirely closed and watertight. The joints and fractures are generally completely filled with various (clayey, carbonate or sulphatic) infilling materials. These findings of subsequent surface and subsurface explorations indicated, it was observed and measured during local deep mining, that the BCF carries the clear signs of self-healing (sealing) by all means (see Figures 5, 6, and 7), but in the real depth interval of final disposal there is no entire and general self-healing in BCF. On the other hand considering the water-tightness of the BFC, combined it with the self-healing capability due to regional high horizontal stresses and/or infill forming, it is expected that the BFC could be an ideal host formation for the Hungarian HLW disposal. It is assumed and believed that EDZ formed as the result of mining around future waste disposal vaults, transport tunnels and shaft is subject of continuous and progressive self-healing at a depth of 500 metres and below.

Therefore it is really a great challenge to understand the recent confinement performance of BCF and describe the future scenarios.

Figure 4. Lithological column of BCF

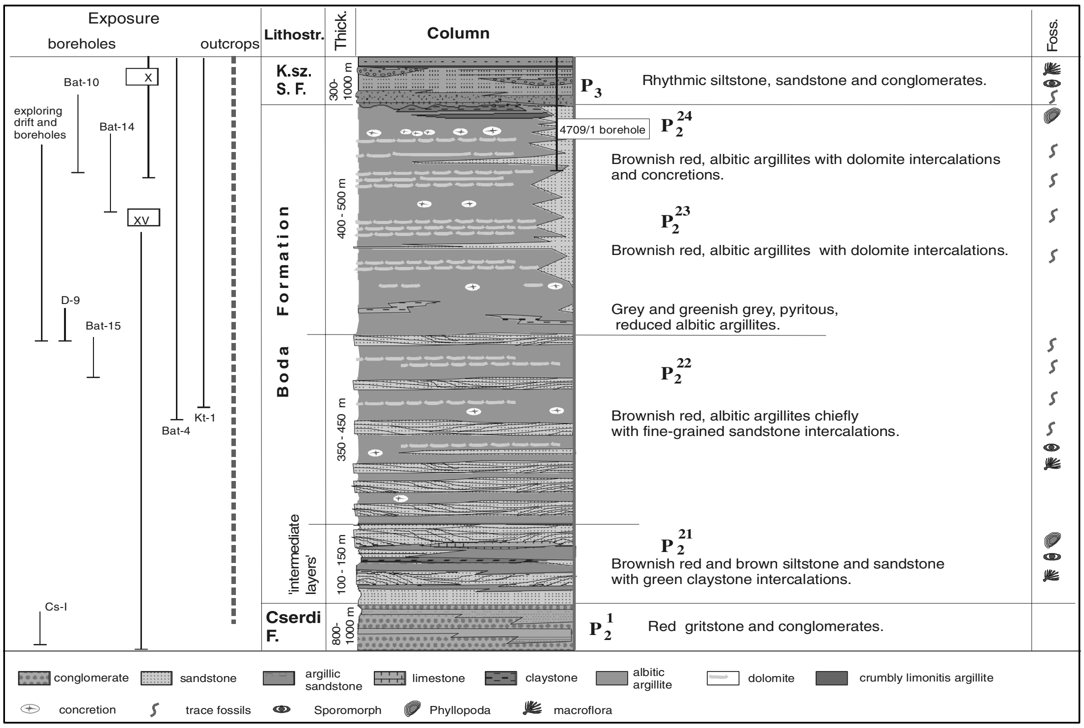


Figure 5. A typical sample gained from a Cretaceous tectonic zone, as an evidence of paleo-self-healing

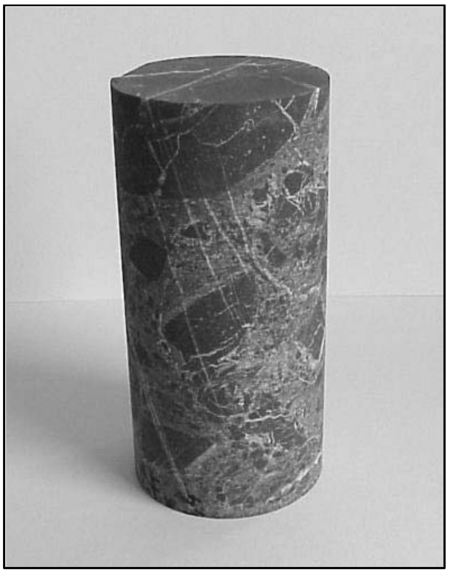


Figure 6. Increase of the proportion of swelling clay minerals I ta tectonic zone

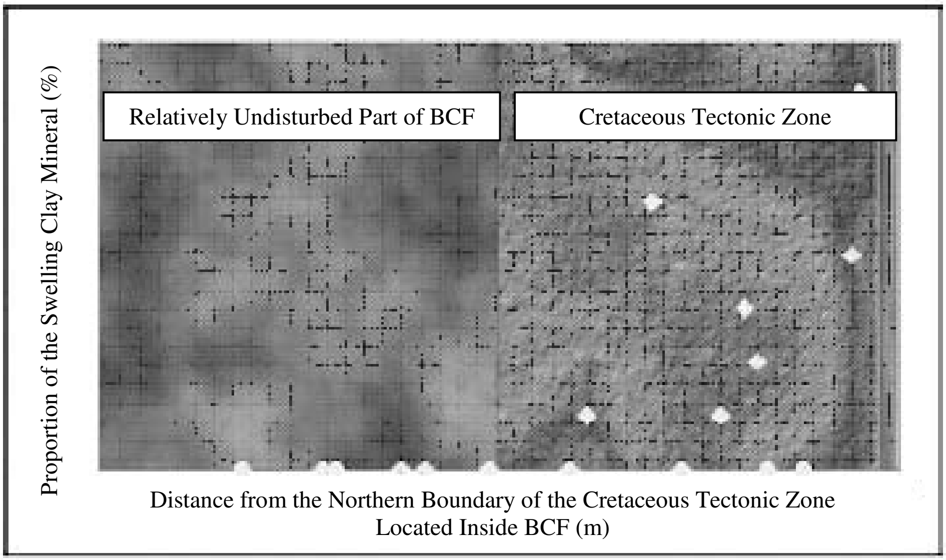
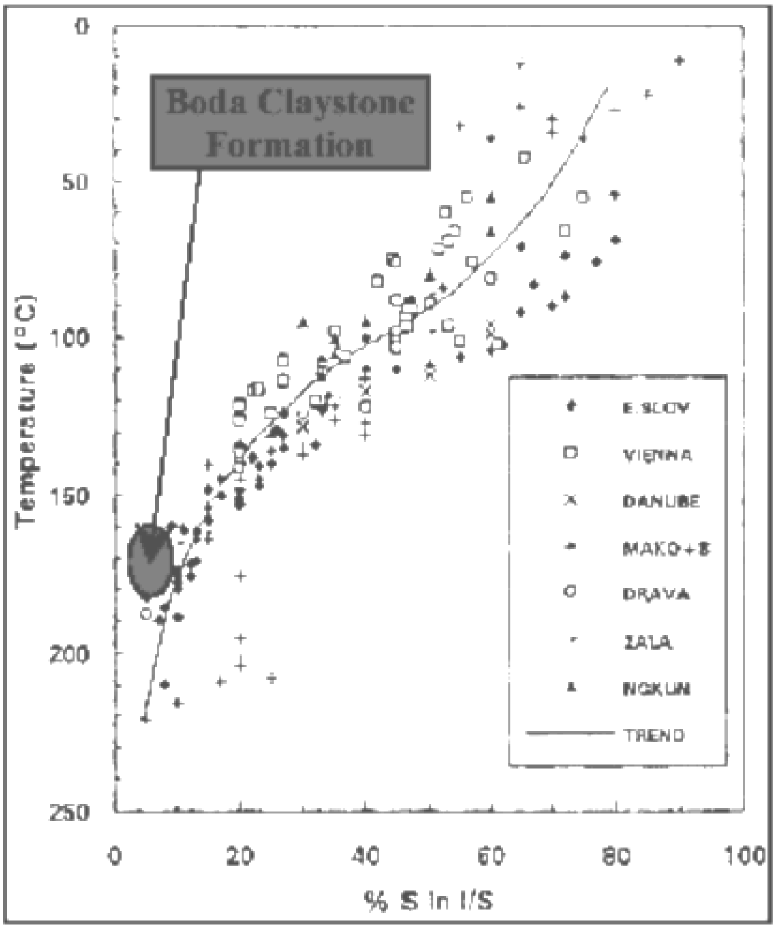


Figure 7. Variation of smectite proportion (S) versus temperature in partial depressions of the Pannonian and Vienna Basins (I. Viczián)



Extrapolation of the present confinement properties

According to the actual Hungarian prescription the possible risk factors after the closure of the repository shall be investigated for a 10 000 year long period, while the trends shall be investigated for 1 000 000 years. Within the STP (and partially even earlier) two more extensive sub-programmes were implemented dealing with the longer term risks:

- The speed and direction of slow crustal movements and the possible deterioration of efficiency of isolation due to them.
- Seismic hazard, probability of destructive earthquakes in the region.

We were able to collect relevant information about the direction of principal displacements, uplifting and subsiding during the Pliocene-Pleistocene (and even recently) through the reconstruction of late Cenozoic evolution, but still need complementary investigations. The most important implication is that the mass of Western Mecsek recently shows uniform elevation uninfluenced by the older tectonic elements.

The probability of dramatic changes in the trends of slow crustal movements is not very high, i.e. the infiltration nature of the area, its movement as an integrate block, the mutual placement of surrounding and overlaying formations are likely to remain almost steady. Thus the confinement capacity of BCF will probably not change significantly in the geological near future (a few hundred thousands of years).

GPS as an adequate method for determining the actual speed and direction of slow movement processes for each geological block are available newly. Unfortunately the first set of data is not suitable for drawing any conclusions.

Even in 1991 the preliminary analysis of seismicity risks and dynamic processes of the region was compiled (see Figures 8 and 9).

Figure 8. Morphological map of South-western part of Hungary with the block of W-Mecsek Mountains

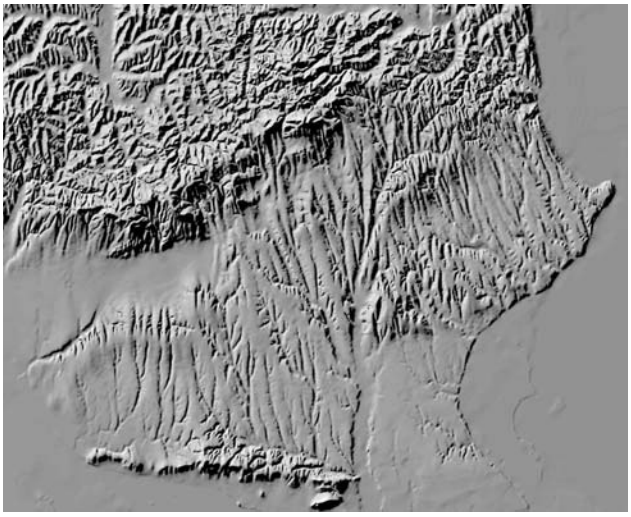
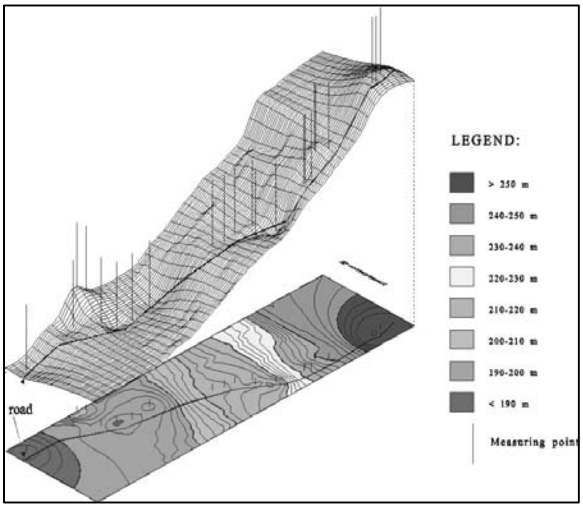


Figure 9. One of the undisturbed Pleistocene terrace deposits in Sás-valley observed by detailed geological mapping



Composition and structure

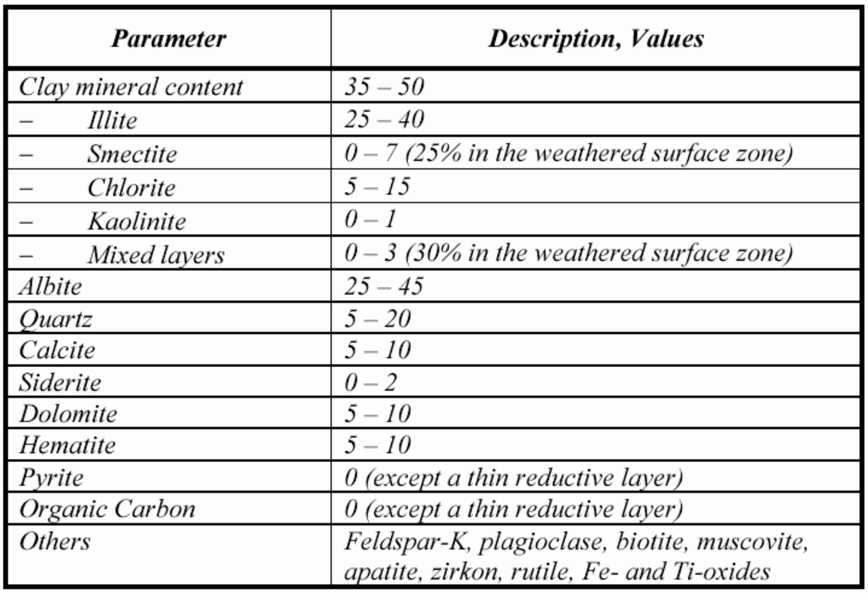
BCF belongs to the group of argillaceous potential host rocks. The confinement and geotechnical features of the formation comprise the following principal components (see Table 1 and Figure 10):

- **Clay minerals:** The total clay-mineral content of the main rock subtypes is generally 35-50% under the dominance of illite. The Chlorite figures out additional. Smectite content is generally below the detectable limit, however, smectites also occur in the weathered zone at outcrops and inside the deformation band of faults.
- **Albite** can be found in disperse form or in inclusions. Together with the quartz grains it ensures the favourable strength of BCF. The albite can be detected in the entire investigated zone (except for the lower and upper transition zones). Albite penetrates even the thin dolomite layers as well. This very rarely observed characteristic gives the unique nature of BCF.
- **Quartz.**
- **Calcite,** found as rock component and as a filling material of fractures as well. As filling material it plays a significant role in the re-closing of tectonically affected rock bodies.
- **Dolomite.**
- **Hematite** and dispersed ferric oxi-hydroxide.

However, with the exception of some special places the proportion of the swelling clay minerals is near the detectable limit (>1%). Thus self-healing caused by swelling may theoretically occur, but it does not seem to be important at first sight.

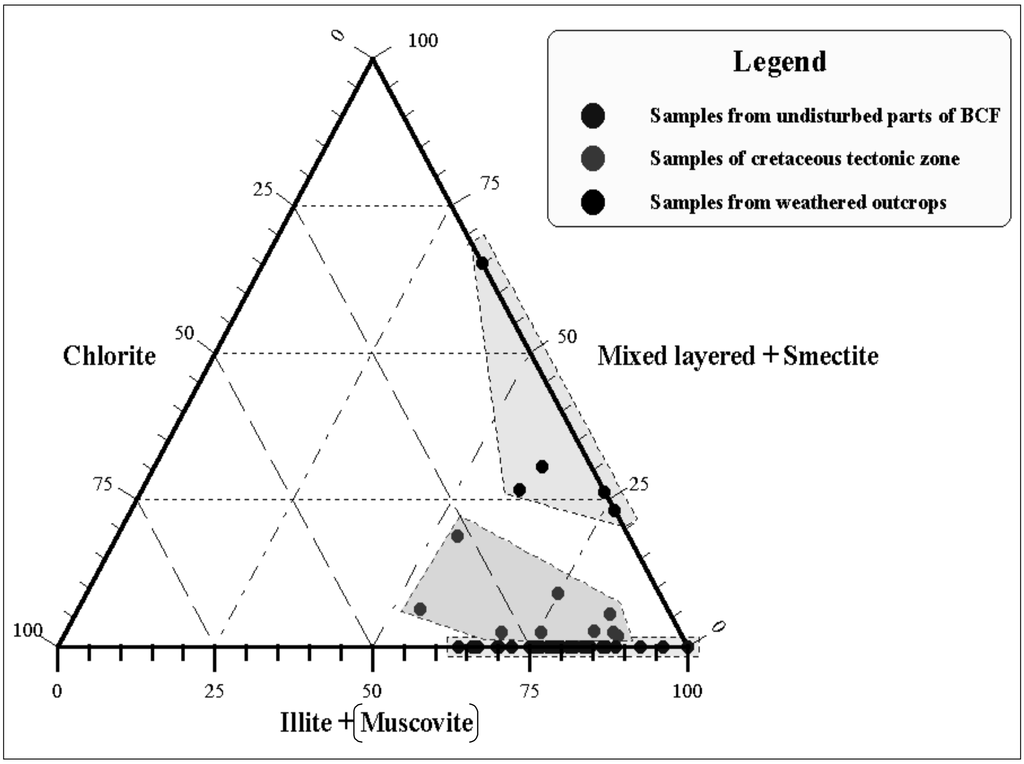
The more detailed mineralogical characterisation can be seen in the table below. (All values are given in % of total dry weight.)

Table 1. Mineralogical characterisation of BCF



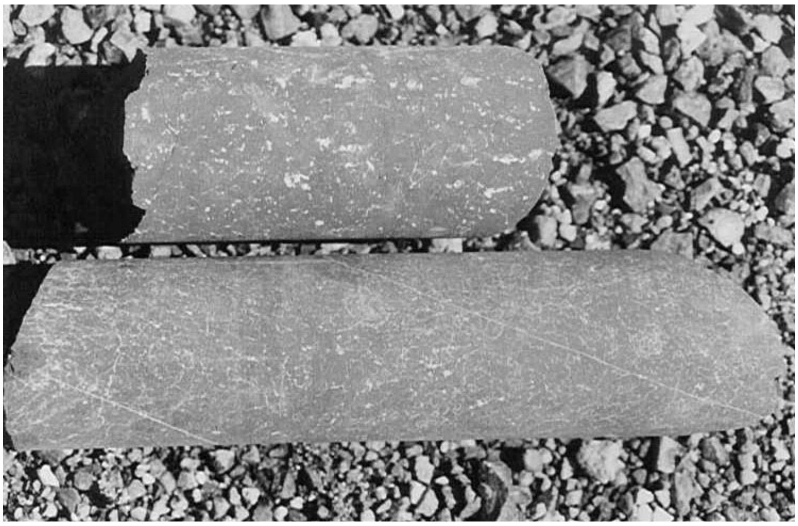
Parameter	*Description, Values*
Clay mineral content	*35 – 50*
– *Illite*	*25 – 40*
– *Smectite*	*0 – 7 (25% in the weathered surface zone)*
– *Chlorite*	*5 – 15*
– *Kaolinite*	*0 – 1*
– *Mixed layers*	*0 – 3 (30% in the weathered surface zone)*
Albite	*25 – 45*
Quartz	*5 – 20*
Calcite	*5 – 10*
Siderite	*0 – 2*
Dolomite	*5 – 10*
Hematite	*5 – 10*
Pyrite	*0 (except a thin reductive layer)*
Organic Carbon	*0 (except a thin reductive layer)*
Others	*Feldspar-K, plagioclase, biotite, muscovite, apatite, zirkon, rutile, Fe- and Ti-oxides*

Figure 10. Minerals in the various zones of BCF

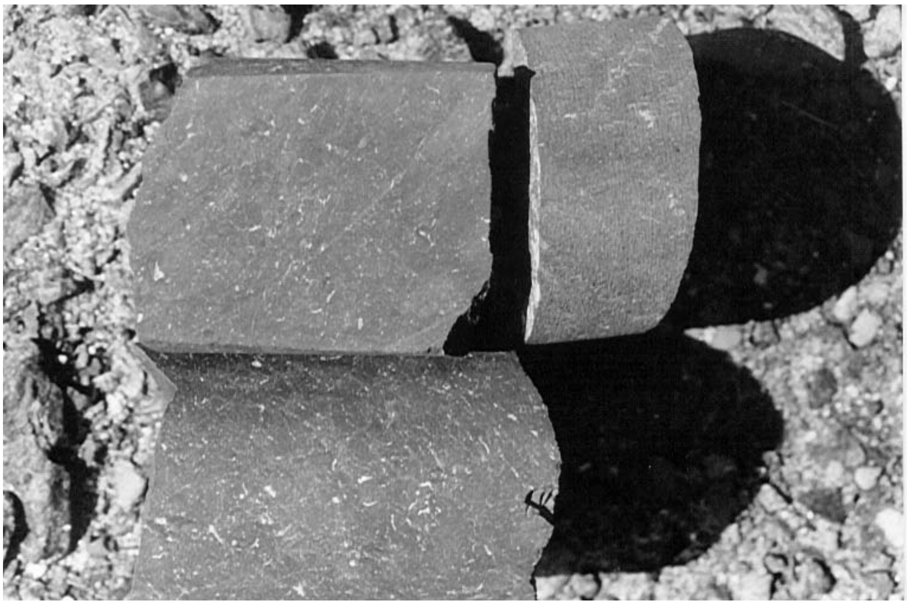


Within the examined zone of BCF (excluding the boundary and transition parts) the above listed minerals appear in four characteristic rock types in different proportions due to the cyclical formation conditions:

1. **Albitic claystones (claystones with albite clusters).** The quantity of the clay minerals and the albite varies among 35-50% (illite >> chlorite). Quartz (about 5-10%) and hematite (7-9%) are also considerable. Calcite and dolomite also.

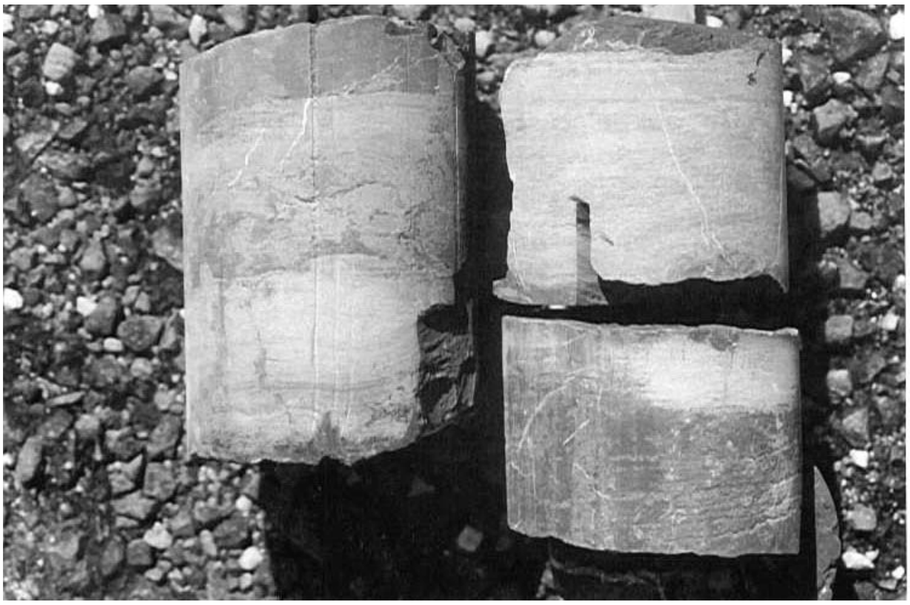


2. **“Albitolites”.** Albite content exceeding 50%, the quantity of clay mineral is low, quartz is minimal. Carbonate content is higher than at the albitic claystones. This statement is particularly true for dolomite. Hematite is similar.



3. **“True” siltstones.** They are always laminated (parallel and cross lamination), high quartz and albite content. The albite is cementing material (non-dispersed), minimal clay content, calcitic cementing material, lower proportion of hematite).

4. **Dolomite layers** (interbedding). High dolomite content, usually high albite content as penetrations, however albite-free dolomite layers can be also found. Quartz and hematite content is minimal).



It should be noted that obviously there are also rocks of transient composition (in the very thick sediment) among the four principal rock types, too. However, it can be concluded that each group consists of the same mineralogical and chemical constituents.

Due to their less favourable confinement properties, both the true siltstones and the dolomites shall be mentioned. These can be found with low clay mineral content and usually in thin layer laminated structure. This is very important as the series of strata is characterised by a small-scale cyclic feature: the thin layer bundles located with 1…3 m spacing between the non-stratified albitic claystone banks forming the main mass of the formation. The latest examinations have indicated a 20 to 50 m periodicity as well, but the importance of this has been unknown yet.

Geotechnical suitability

The detailed laboratory measurement programme, the underground explorations and the state of tunnels itself unambiguously verified that despite of 1 100 m maximum depth, BCF has unexpectedly favourable geotechnical capabilities. Comparing with the sandstone broken by the same tectonic zone and having the same fracturing level BCF shows a significantly more favourable stability status.

As it was formerly indicated, the initial slacking of the walls proved to be the only one stability problem during and after tunnelling. After the excavation the mentioned effect also ceased within 1-2 weeks. After a further 6-8 month period perfect stability was observed even at the most tectonised zones. Posterior stability problem or significant spalling was not observable in the tunnel sections drifted more than four years ago.

It can be supposed that the geotechnical conditions are similar everywhere within the 500-1 000 m depth range. However, it shall be highlighted we have not got appropriate knowledge about the exploration of tectonic areas with less favourable oriented tunnel.

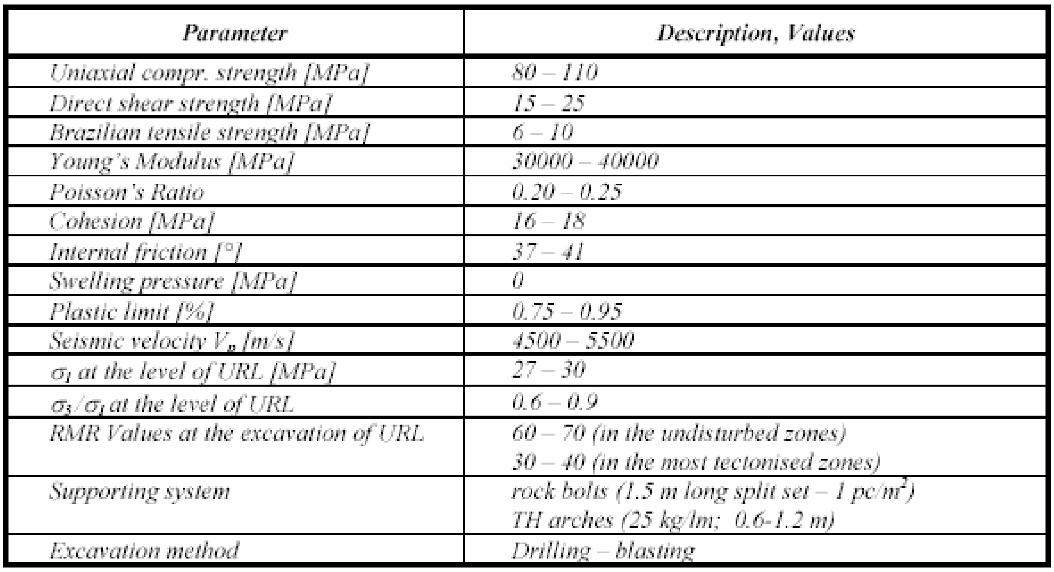
Table 2 and Figures 11 and 12 show the results of experiments in the URL.

The final judgement of geological suitability of the formation will be made via an iterative series of performance assessments. The largest problem of the Hungarian characterisation programme is that it was not controlled by PA. Several basic data sets are missing to perform a really effective assessment. The acquisition or refining of the following information would have crucial importance:

- the biosphere parameters including the present consumption habits too;
- the isotope composition of the waste to be disposed;
- at least conceptual plans of technical solutions;
- augmenting the recently incomplete geological information.

Of course, the lack or inaccuracy of these parameters does not mean that the execution of an initial or interim PA is unnecessary at the actual level of knowledge. It would be indispensable for the verification of results accomplished so far and for determination of the contents of next characterisation phases.

Table 2. Geomechanikal and geotechnical parameters of BCF



Parameter	*Description, Values*
Uniaxial compr. strength [MPa]	*80 – 110*
Direct shear strength [MPa]	*15 – 25*
Brazilian tensile strength [MPa]	*6 – 10*
Young's Modulus [MPa]	*30000 – 40000*
Poisson's Ratio	*0.20 – 0.25*
Cohesion [MPa]	*16 – 18*
Internal friction [°]	*37 – 41*
Swelling pressure [MPa]	*0*
Plastic limit [%]	*0.75 – 0.95*
Seismic velocity V_p *[m/s]*	*4500 – 5500*
σ_1 *at the level of URL [MPa]*	*27 – 30*
σ_3/σ_1 *at the level of URL*	*0.6 – 0.9*
RMR Values at the excavation of URL	*60 – 70 (in the undisturbed zones)* *30 – 40 (in the most tectonised zones)*
Supporting system	*rock bolts (1.5 m long split set – 1 pc/m*^{2}*)* *TH arches (25 kg/lm; 0.6-1.2 m)*
Excavation method	*Drilling – blasting*

Figure 11. Comparison of calculated RMR-values and the supporting techniques applied in fact at the different parts of Alfa-1 tunnel

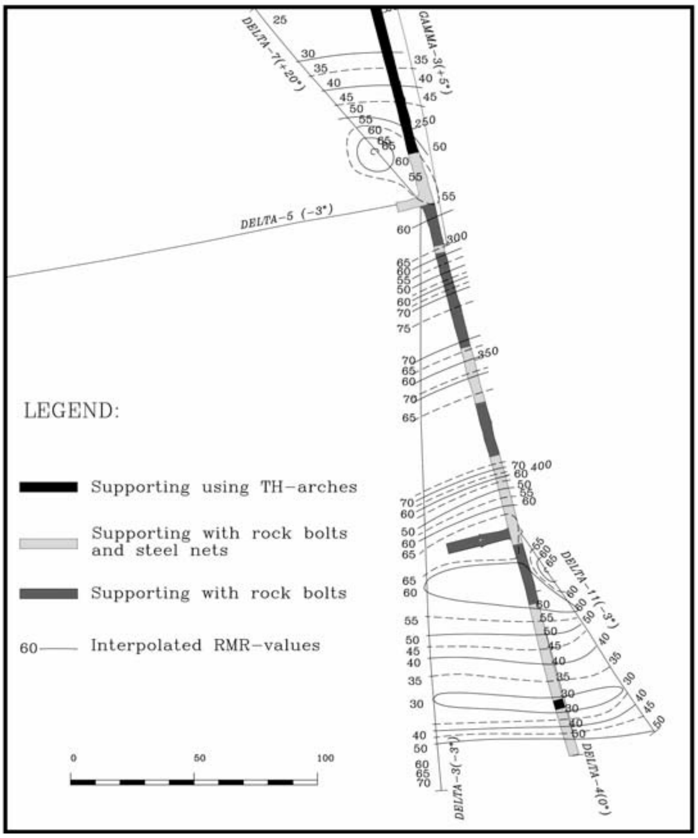
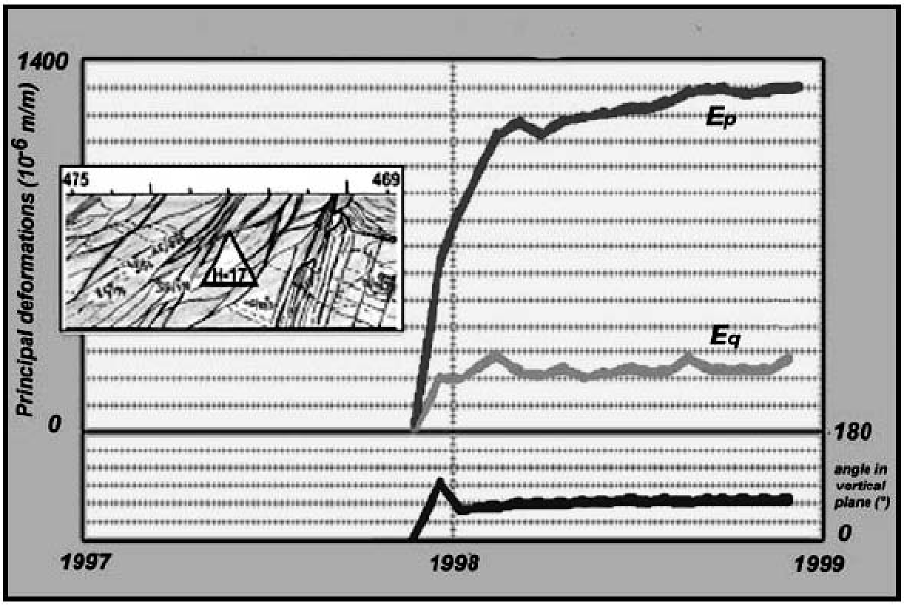


Figure 12. Long-term deformation of rock wall measured at t large tectonic zone by H-17 triangular deformation monitoring device

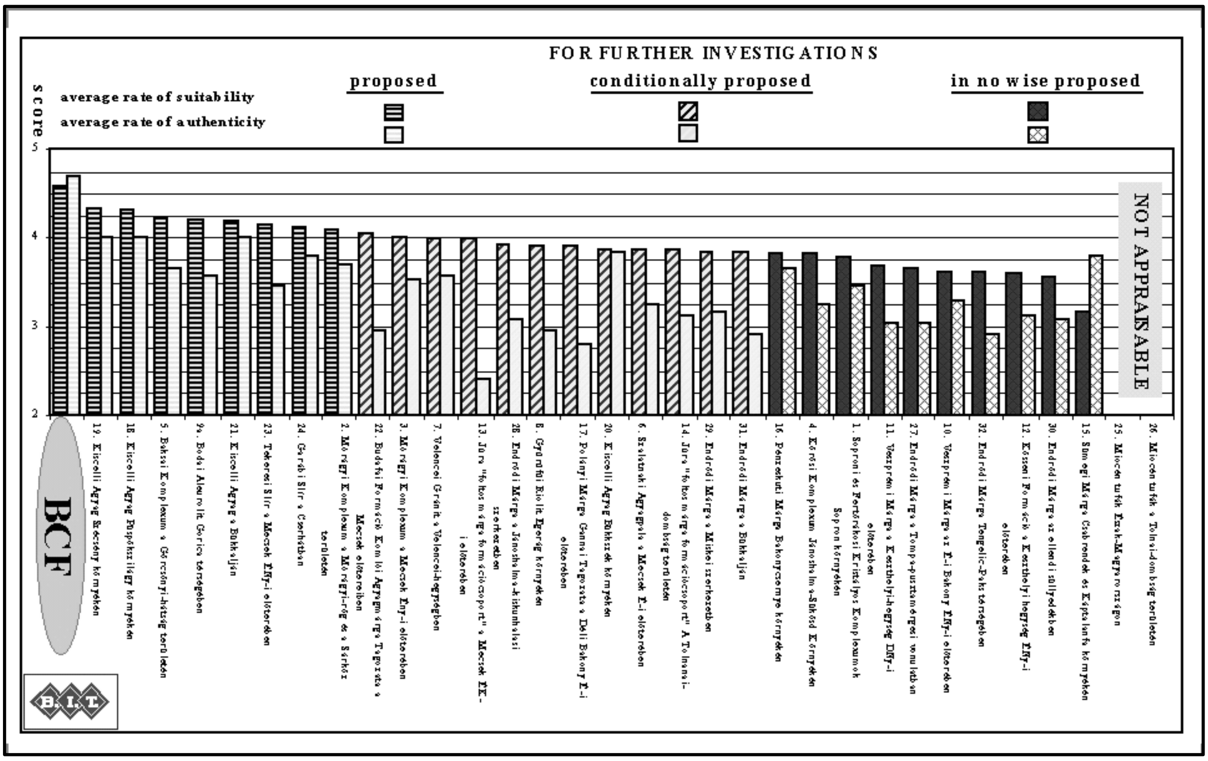


Epilogue

At the present level of knowledge there is no real alternative for resolving the disposal problems of HLW other than the final disposal in deep geological formations. It can also be assumed that Hungary will disburden of its own HLW inside its own territory.

The Short-Term Programme completed in 1999 was already the third one in the row of exploration programmes aiming the characterisation of the Boda Formation. The results of intensive investigations and national screening (see Figure 13) executed by BIT Ltd. in 2000 confirm that the Boda Claystone Formation located in the West-Mecsek, SE-Hungary still has the leading position among the potentially suitable sites. A new site selection and characterisation project based on this position and started in 2003.

Figure 13. The result of nationwide screening



MINERALOGICAL BEHAVIOUR OF BENTONITES IN OPEN AND CLOSED SYSTEMS

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Mineralogical and chemical changes of bentonites were investigated in a natural analogue study and in laboratory experiments.

As a working hypothesis we assumed that in geological, i.e. open systems, bentonites may be penetrated over geological time scales by larger water volumes than high compacted bentonites used as technical barriers in repositories in salt formations. Under this assumption open geological systems are characterised by low solid/liquid ratios and closed repository systems by high solid/liquid ratios. Consequently in laboratory experiments the mineralogical changes were investigated under different solid/liquid ratios and compared with results of a natural analogue study.

In the natural analogue study in deep boreholes in the East Slovakian Basin the expandability of montmorillonites and the degree of transformation in illite-smectite (IS) mixed layer structures was found to be dependent not only on depth and temperature but also on the salinity of the pore waters. In this open geological system with a comparatively low solid/liquid ratio the observed changes in the montmorillonites were significantly different than those observed in the laboratory study on compacted MX-80 bentonite.

Long time experiments (600 days) with uncompacted MX-80 bentonites and saline solutions at a high solid/liquid ratio (closed system)

In the laboratory experiments the MX-80 bentonite was reacted with two high saline solutions, a NaCl and a $MgCl_2$ rich brine at 25°, 90° and 150°C, at three different pH, 1, 6.5 and 13. 10 g bentonite were mixed with 10 ml solution. No further stirring or shaking of the samples was done during the experiments. The reaction products were analysed by XRD, DTA/TG and transmission electron microscopy. In all experiments montmorillonite remained at all temperatures and all pH the predominant mineral phase, with full ethyleneglycol expandibility, 16.9 Å. However, fast and significant significant changes could be detected by looking at parameters like morphology, crystallinity, particle height, particle surface, interlayer charge and chemistry of octahedral layers. After 2 days already an alteration towards Al-rich end-members of montmorillonite could be detected. This alteration increased with time. The main process was a substitution of octahedral Mg by Al. The composition of the tetrahedral layers remained unchanged (very close to $Al_0Si_4O_{10}$). This substitution reduced the interlayer charge continuously from a starting value of 0.3 to 0.2 and less. This remarkable charge reduction is interpreted as a pyrophyllitisation process. Pyrophyllite has zero interlayer charge. The charge reduction of the MX-80 montmorillonite interlayers was confirmed by independent measurements after alkylamonium treatments. As a result of the charge reduction technical parameters like the swelling capacity, the water uptake capacity and the swelling pressure were reduced significantly compared with the original bentonite.

The early appearance of high Si and Al concentrations in the solutions of all investigated systems demonstrates that fast dissolution is an important process in the investigated reactions. Decreasing concentrations in solution on the further reaction path indicate that precipitation perdominates dissolution. The results obtained so far, do nor allow however a quantification of the amount of dissolved initial material.

The appearance of Mg in the solution of the NaCl-series is interpreted as a Mg release from the octahedral layers. In accordance the TEM-EDX measurements on reaction products indicate a substitution of Mg by Al in the octahedral layers.

In the samples with $FeCl_3$ at increased temperatures K was mobilised in remarkable amounts probably from collapsed interlayers. Obviously, the activation energy necessary for the replacement of K in collapsed interlayers was high enough to demand for the combined forces of the tri-valent cation Fe^{3+} and a temperature of 150°C. Temperature dependent precipitation influences largely the concentrations of some of the aqueous species in the solution. The observed transformations of the original montmorillonites towards pyrophyllite may be expected according to Lippmann (1979) in case of steadily high concentrations of Si in the solutes at the reaction sites

Short time experiments (30 days) with compacted bentonites and NaCl-solutions simulating open and closed systems

Pusch & Kasbohm (2002) performed 30 days experiments at 100°C with compacted MX-80 bentonites with a dry density of 1 200-1 300 kg/m^3. Two sets of experiments were performed in order to simulate the open and closed system. In the closed system experiments three bentonite samples were saturated before compaction, one with destilled water and two with salt solutions. One solution contained 10 wt-% the other 20% NaCl. In the experiments simulating the open system the compacted bentonite was percolated continuously with water and the two NaCl solutions. At the end of the experiments the bentonite was characterised

Changes in the closed system

After 30 days montmorillonit was still the dominating mineral phase. Full expandibility with ethylenglycol to 16.9 Å was still present but asymmetrical peaks at 8.5 and 5.65 Å indicated the first appearance of IS mixed layers and irreversible contractions of the layer structure. TEM-EDX investigations showed for all particles a substitution of Mg by Al. The medium particle height of 80-90 Å did not change. The original bentonite compacted to a raw dry density of 1 300 kg/m^3 percolated with water had a hydraulic conductivity of 3E-12 m/s. After 30 days in contact with 10% NaCl solution this value decreased to 9.8E-12 m/s and to 2E-12 m/s with 20% NaCl solution.

Changes in the open system

Here too, montmorillonite was the dominating phase after 30 days and no reduction of the ethylenglycol expandibility was observed. However the medium particle height increased from the initial 80-90 Å to 150-200 Å. Considerable amounts of the original montmorillonite were transformed in beidellite. Traces of kaolinite were detected. In the original MX-80 bentonite the minerals beidellite and kaolinite do not exist. In the remaining montmorillonite particles a substitution of Mg by Al was observed. The reduction of the Si/Al ratio from 2.4 in the montmorillonite to 1.5 in the beidellite and 1 in kaolinite are clear indications of a removal of Si during the reaction in the open system. No important illitisation was observed, probably due to the fact that no K source existed in the system. The reduction of permeability was similar to that observed in the closed system.

Short time experiments (20 days) with other compacted bentonites and NaCl-solutions simulating an open system

In order to make sure that the observed mineralogical changes are not only particularities, linked to the special mineralogical composition of MX-80 bentonite, several other bentonites were investigated similarly in short term experiments. 1.25 g samples of the US bentonite standards Chambers/Arizona (No. 23), Belle Fourche/South Dakota (No. 27), Pioche/Nevada (No. 32) and Otay/California were reacted with 5 ml 1N NaCl solution in an overhead shaker for 20 days.

Table 1. General overview over observed mineralogical changes in MX-80 and other bentonites in open and closed system experiments

Reaction system	Tetrahedral layer		Octahedral layer		Interlayer		Type of reaction	Investigated bentonite
	Cations	Charge	Cations	Charge	Cations	Charge		
“Closed”	unchanged	unchanged	Al > Mg	reduced	Mg increased	reduced	Pyrophyllitisation	BS-MX80* Lund-MX80*
“Open”	unchanged	unchanged	unchanged	unchanged	unchanged	unchanged	un-changed	Belle Fourche
	Al > Si	higher	unchanged	unchanged	Na, K	higher	Beidellitisation/ kaolinitisat.	Lund*-MX80
	Al > Si	higher	Mg>Fe (Al)	higher	Na (K)	higher	illitisation	Chambers, Pioche, Otay

* Experiments performed in the GRS in Braunschweig/Germany.

** Experiments performed in Lund/Sweden.

The mineralogical investigations showed that most of the bentonites were transformed but to different degrees and in different directions. Whereas the Belle Fourche bentonite remained almost unchanged after 20 days the fastest changes were observed in the Chambers bentonite, were IS mixed layers became dominant, Al was substituted by Mg in the octahedral layer in remarkable amounts causing an increased octahedral layer charge, processes interpreted as illitisation. Similar changes were observed in the Pioche and Otay bentonites (see Table 1).

Conclusions

The experimental results indicate a fast alteration of the montmorillonites in most of the investigated bentonites. Significant changes were observed (with one exception) very soon after the start of the experiments. The direction and the velocity of the observed reactions were different in different bentonites. In the MX-80 bentonite in the closed system a reduction of the octaedric layer charge and interlayer charge was induced by a substitution of Mg by Al. This process is interpreted as pyrophyllitisation. In the open system the changes were different and lead via an increase of the interlayer charge and a decrease of the Al/Si ratios of the particles to a beidellitisation and kaolinitisation. In other bentonites in the open system experiments not only the interlayer charge but also the octahedral layer charge increased, leading to an increase of the IS mixed layer structures. This process is interpreted as illitisation.

We conclude that bentonites are not stable in the environment of high saline solutions neither in the short term nor in the long term. Different transformations may be expected in open geological systems and closed repository systems. Illitisation seems to be characteristic for open systems whereas pyrophyllitisation may be expected in the compacted bentonite barriers in repositories in salt formations in the long run.

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EXPERIMENTAL STUDY OF THE HYDROMECHANICAL BEHAVIOUR OF THE CALLOVO-OXFORDIAN ARGILLITES

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Various laboratory experiments were carried out in the framework of the MODEX-REP project on the Callovo-Oxfordian argillite samples taken from the Meuse/Haute-Marne Underground Research Laboratory (MHM-URL) in Eastern France, to provide basic data for modelling the hydro-mechanical response of the argillite to shaft sinking. The short-term mechanical behaviour of the argillite was investigated by means of uniaxial and triaxial compression tests, whereas the long-term behaviour was studied by uniaxial creep and relaxation tests. Some influence factors such as material anisotropy, scale effect, water content and sample origin were examined. Permeability of the argillite was determined on wet and dry specimens parallel and perpendicular to the bedding plane by using gas under different confining pressures.

The uniaxial compression tests focused on investigating the effect of water content on the mechanical behaviour. The compressive strength and the failure strain of the air-dried specimens are about two times higher than those of the saturated ones. Young's modulus measured increases from 5 500 MPa to 7 500 MPa with an increase of the axial stress from 2 MPa to 20 MPa, independent of the water content. In the multistage triaxial compression tests, dilatancy, failure and residual strength of the clay rock were examined at different confining pressures of 3 to 16 MPa. From the tests, the strength parameters were estimated. With regard to the short-term mechanical behaviour scale and anisotropy effects were observed.

The uniaxial creep tests were conducted in creep rigs at ambient temperature. Axial load was applied stepwise from 2 MPa to 15 MPa on saturated samples of different sizes with 45/100 mm diameter and 90/200 mm length, oriented perpendicular and parallel to the bedding. Most of the tests lasted for about 1 year. In the creep tests, no lower creep limit, no significant scale effect and no significant anisotropy effect on the pure creep behaviour for the argillite were found. The long-term mechanical behaviour of the investigated region of the argillaceous formation is relatively homogeneous.

Gas permeabilities of the dried specimens were measured under confining pressures up to 16 MPa. The gas permeability parallel to the bedding plane is about one order of magnitude higher than that perpendicular to the bedding and decreases with increasing confining pressure. The gas permeability at a water content of 4-5% is about three orders of magnitude lower than that of the dried specimens. In fully saturated conditions, the argillite is impermeable for gas.

CHANGES IN X-RAY PATTERNS, REHYDRATATION, ABILITY, CATION EXCHANGE CAPACITY AND SPECIFIC SURFACE AREA OF BENTONITES FROM ROKLE DUE TO THE EXPERIMENTAL HEATING

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Introduction

Bentonites from Rokle deposit (Czech Republic) were studied as a supply source of the candidate buffer and backfill material for the deep repository concept.

Important physical and chemical properties of bentonites could be significantly affected by the effects of increased temperature due to heat generation from the canister.

The aim of this paper is to determine changes in rehydratation ability, position of 001 XRD powder profiles, cation exchange capacity (CEC) and specific surface area after heating.

Material and methods

The studied bentonites were collected in the operating Rokle clay quarry (western part of the Bohemian Massif). Bentonites include a 25 m thick formation of basal pyroclastics, forming the periphery of Tertiary alcaline volcanic rocks of the Doupovské Mts. (Franče, 1992).

Natural bentonites from Rokle deposit consist mainly of montmorillonite with Ca and Mg as dominant interlayer ions. Kaolinite and quartz are subordinate. The mineralogical composition of studied bentonites was determined by X-ray diffraction. Analysis of smectites was facilitated by preparation of oriented and glycolated samples. Semiquantitative mineralogy of selected samples was determined using recalculation program CQPA and XRD and chemical analyses data.

Experiments were done using high-temperature XRD camera. X-ray patterns were taken 15 minutes after reaching the following temperatures 25°C, 50°C, 70°C, 90°C and 110°C using diffractometer Siemens D5000. The X-ray camera was not evacuated during exposure because vacuum dehydrates the samples and would cause changes similar to those inducted by heating.

Powder samples for CEC and specific surface experiments were heated in a muffle furnance to 50°C, 60°C, 70°C, 80°C, 90°C, 100°C, 150°C, 200°C, 250°C, 300°C, 350°C, 400°C, 450°C and 500°C and kept for 60 minutes at each temperature. After dehydratation at individual temperature, CEC and specific surface area was recorded.

Cation exchange capacity was determined using substitution of exchangeable cations by solutions (1 mmol/l CsCl, 50 mmol/l Ca^{2+} and 50 mmol/l Mg^{2+}) and by measurement of concentrations of sorption before and after experiments using AES and ICP-MS.

Parameters characterising the porous structure of bentonite were obtained from the sorption analyses using the SORPTOMATIC 1 800 apparatus. The samples were outgassed until they reached a constant weight, at pressure of $<10^{-6}$ Pa, at the temperature of 333 K.

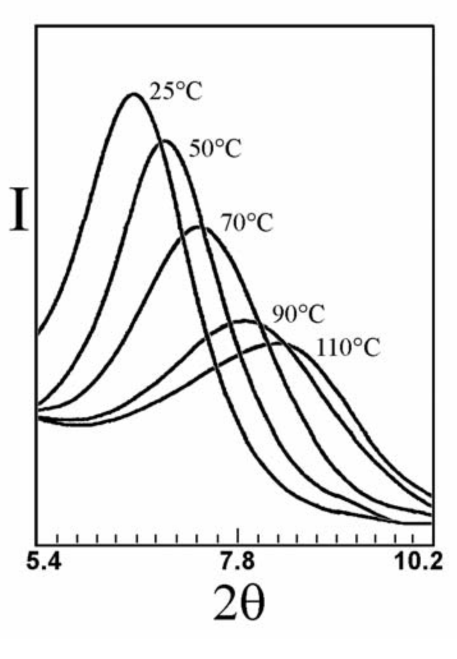
The specific surface area S_{BET} which represents predominantly the surface area of meso and macropores (>2 nm) was determined according to BET method (Brunauer *et al.*, 1938) from the N_2 adsorption isotherm at 77 K.

The microporous structure S_{micro} (<2 nm) parameters were determined from CO_2 isotherms at temperature of 298 K within a pressure range from 0 to 1 000 mbars. The isotherms were evaluated according to Dubinin (1967).

Results

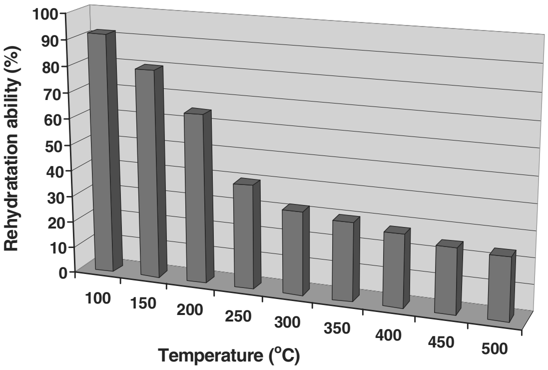
Profiles of the first basal diffraction (001) of Ca/Mg montmorillonite exhibit significant changes induced by dehydratation (Figure 1). There is a decrease of the profile intensity and a shift of the maximum from 2θ=5.7 (d=15.36 Å) to 2θ=8.6 (d=10.20 Å) the temperature increases from 25°C to 110°C.

Figure 1. XRD profiles of 001 diffraction of Ca /Mg montmorillonite from Rokle



The dehydratation process of Ca/Mg montmorillonite can be divided into two stages (Figure 2). The first corresponds to temperatures between 50°C and 200°C where the dehydratation ability drops to 61%. The second stage corresponds to temperatures between 200°C and 500°C where the dehydratation ability drops to 20%.

Figure 2. The dehydratation ability of Ca /Mg montmorillonite from Rokle



The decrease in CEC appears to be continuous with increasing temperature (see Figure 3). The S_{micro} also decreases with increasing temperature, whereas S_{BET} increases with increasing temperature (Figure 4).

Figure 3. CEC of bentonite from Rokle

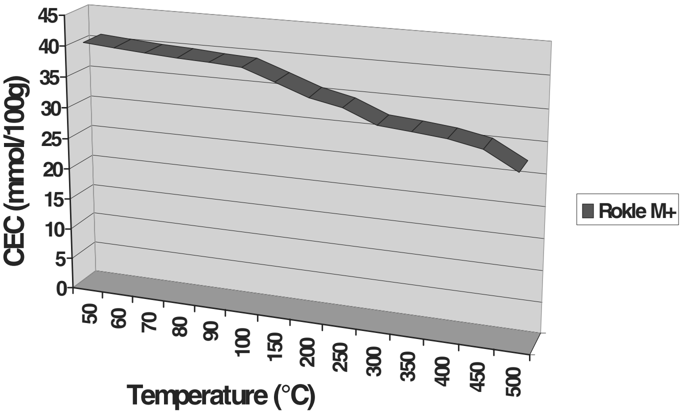
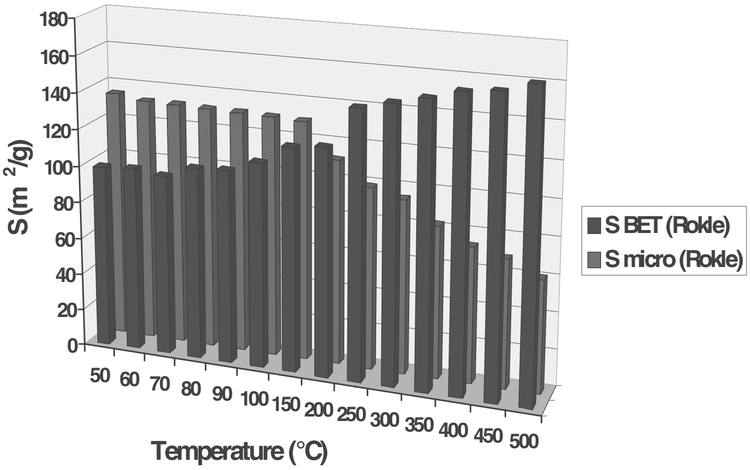


Figure 4. Specific surface area of bentonite from Rokle



Conclusions

The thermal effect strongly affected the bentonite material. The dehydratation processes of Ca/Mg montmorillonite appear at temperature around 50°C. The rehydratation ability drops to 20% at 500°C. Moreover, big changes of important physical properties (CEC, specific surface area) of bentonites were observed. The CEC drops to 25 mmol/100g and S_{micro} to 60 m^2/g. However, S_{BET} increases up to 158 m^2/g.

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