Impact of Nuclear Data Uncertainties on a GEN IV Thorium Reactor at Equilibrium.

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Thorium cycle

The thermal thorium cycle is much more favourable than the thermal U/Pu one. Even though the number of neutrons available is not as high as in the fast U/Pu cycle, this cycle has a very important potential. Shippingport experience has shown that regeneration could be achievable in a modified PWR. Its low higher actinide production could ease the nuclear waste management in case of a long term deployment of nuclear energy.

The high capture cross section of fission products in a thermal spectrum makes the fission chain difficult to maintain with their accumulation. That is why Molten Salt Reactors (MSR) are particularly well adapted to thorium fuels. In these concepts, the salt containing the fuel is also the coolant. Added to the good thermo-hydraulics properties of the salt as coolant, this configuration allows an almost continuous recycling of the fuel. This recycling allows the online control of the reactor criticality and fuel cycle. The removal of fission products and of other neutron poisons (such as ^{233}Pa) improves the regeneration gain and thus the deployment potential of this cycle. Any uncertainty on the regeneration gain can be interpreted as a potential increase of the reprocessing effort needed and an equivalent decrease of the economic competitiveness[1].

Nuclear Data Sensitivity studies

1. Available nuclear data uncertainties

Covariance matrices are very often said to be the limiting factor of the interpretation of this kind of analyses: the confidence that one can have on some reactor parameter uncertainty cannot exceed the one on the basic nuclear data uncertainties used to estimate this uncertainty. This can be said today, for the case of thorium fuel cycle related nuclear data. This might change very soon as the next generation of evaluated files, assembled in the scope of AIEA CRP on "on Evaluated Nuclear Data for Thorium-Uranium Fuel", will contain new covariance matrices, but they are not yet available at the time of the writing of this request.

The lack of uncertainties on the nuclear data forces Palmiotti et al. 1) to propose a quite complete set of covariance matrices based on their "expert guess" *i.e.* on the large experience of comparison of experimental results to simulated ones. In fact, it is always recommended to have a look at the uncertainty proposed there, as the uncertainties given in the evaluation tend to underestimate

[1] The Molten Salt Breeder Reactor project, developed in the 70's on the base of the successful experiences done in Oak Ridge in the 60's, was abandoned because the complexity of the fuel reprocessing was estimated to be too high when compared to the one of the more classical equivalent U/Pu fast cycle.

systematically the proposed ones, which can eventually lead to a dangerous over-confidence in the basic nuclear data.

The uncertainty information used to write this request (based on the results presented in the reference 2)) can be separated on the two sources:
First, the information available in various evaluated files allows the drawing of a reasonable portrait of the Thorium Molten Salt Reactor k_{eff} uncertainty.
Second, for the estimation of the uncertainty of the regeneration gain presented here, the uncertainties available for some key cross sections seemed too low and the ones available in reference 1) were preferred. In the spectrum of interest for this kind of epithermal reactors, the basic uncertainty comes from capture cross sections, which are “guessed” to be known with 10% accuracy. This value is consistent with what was available for ^{232}Th. It is also consistent with the spread (up to 25 % of the quite old experimental information 3)7)]) on which are based ^{233}U capture cross section.

2. Keff sensitivity and uncertainty studies

The complete nuclear sensitivity study of criticality has been presented at the Avignon conference 8). Two methodologies, both based on Perturbation Theory but using very different transport programs, where compared and showed consistent results. On the base of the available uncertainty information, the most important contribution to the global uncertainty was coming from ^{232}Th capture cross section. The uncertainty on this data raises the uncertainty on k_{eff} to more than 3000 pcm. This is quite high when compared to fast neutron breeder reactors 9), 10). A new evaluation of this nucleus, based on new experimental data taken at the n-TOF facility and in GEEL was recently completed by the IAEA Co-ordinated Research Project for the Th/U fuel cycle. The new evaluated file was tested on the available benchmarks and generally improved the C/E values. Although, it was incorporated in the ENDF/B-VII library including new covariance information, the impact on the uncertainty of k_{eff} predictions requires additional study.

3. Regeneration gain uncertainty evaluation

If the regeneration is defined as the balance between the production and the destruction of the main fissile nucleus[2], it can be calculated as a simple ratio of reaction rates (see equation 1). And the uncertainty on this ratio can be calculated using Generalized Perturbation Theory 11). Indirect terms[3] coming from Perturbation Theory would probably not change what can be seen on the definition of regeneration gain: ^{232}Th capture and ^{233}U fission cross sections must dominate the uncertainty as the sensitivity of the RG to these cross-sections must be close to 1[4]. Thus, the uncertainty will be linked to the capture cross section as its uncertainty was estimated to be of the order of 10%, while the one on the fission cross section limited to some %, and may reach 10% which is larger than the RG potential of Thorium cycle!

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[2] Equ. 1gives RG=0 for iso-generator reactor. The Thermal Molten Salt studied here has been made almost iso-generator, thus its RG is very small 200pcm.
[3] These terms take into account the uncertainty in the flux spectrum due to the uncertainty in the cross section. GPT shows that they can be written as weighted reaction rates. The “weight” is the importance function linked to the studied reaction rate ratio.
[4] A relative change in the above-mentionned 1 group-cross-sections would produce the same relative change of RG, which means exactly that the sensitivity of RG to them is 1.

$$RG = \frac{\Sigma_c^{Th} - \Sigma_c^{^{233}Pa}}{\Sigma_f^{^{233}U} + \Sigma_c^{^{233}U}} - 1$$

Reality is slightly more complex than the simple definition of RG:

- Based on this definition, it seems that one can change the RG indefinitely by changing ^{232}Th or ^{233}U densities; there could be no limit in the deployment of ^{232}Th fuel cycle! In a critical reactor this ratio is imposed by the need to maintain the reactor critical. Any increase in the ^{232}Th reaction rates, due to an increased density or by a microscopic cross section bigger than expected would increase the RG but decrease k_{eff}. In a real MSR, the decrease of k_{eff} would be compensated by a suited increase of ^{233}U density, reducing the RG. Numerical simulation 2) of this effect shows that there is not a complete compensation, the direct effect being cut by a factor 5. This shows that the main constraint of a reactor (to be kept critical !) should not be neglected in a sensitivity analysis
- Because of the previous point, regeneration is often presented as the "available neutrons", the ones that can be captured on thorium, once the neutrons needed to maintain the reaction chain[5] are subtracted from the ones produced by fission. Once again, the facts are not so simple: among the neutrons needed to maintain the reaction chain, some are captured by the ^{233}U and do not make it fission. Instead, they produce a ^{234}U, which is a fertile material. As the fuel cycle is almost closed, these atoms will accumulate until their concentration reaches the equilibrium where their production rates by capture on ^{233}U will equal their destruction rate, mostly by capture. This capture will in turn produce ^{235}U, a fissile material whose contribution to the fission rate is 10%!

Some valuable tools were developed 10 years ago in CEA Cadarache [12), 13)]. They calculate the number of neutrons produced by the destruction of one atom, taking into account all its descendants by capture and/or decay. They allow the analytical evaluation of this quantity and of some others by assuming that the nuclides have reached their equilibrium.

Criticality can be written as the balance of the neutrons produced by the only chains based on ^{233}U and ^{232}Th and the ones captured by fission products and structure materials or leaked. It has been shown [2] that the RG can be extracted from this equation.

With $\alpha = \frac{\sigma_c \Phi}{\sigma_f \Phi}$ the capture to fission ratio, $\lambda = 1/Tc$ the inverse of the characteristic time of decay[6], $\Lambda = \frac{\lambda}{\sigma_f \Phi}$ the decay to fission ratio, and $\gamma = \alpha$ or Λ depending on the disappearing mode of the nuclide k in the chain I. The number of neutrons produce by the destruction of nuclide j can be written:

$$D_j = \sum_i \left(\frac{\nu_i - (1+\alpha_i)}{1+\alpha_i+\Lambda_i} \prod_{k=0}^{i-1} \frac{\gamma_k}{1+\alpha_k+\Lambda_k} \right)$$

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[5] These neutrons can be absorbed on ^{233}U to make another fission, leak outside the fuel, or be captured by fission products of structure materials.

[6] this can be a radioactive decay and then $Tc = \tau_{1/2} / \ln 2$ or an equivalent decay due to chemical reprocessing

And the RG can be written:

$$GR=\frac{1}{1+2\frac{\sigma_c^{233Pa}\Phi}{\lambda_{233Pa}}-\frac{\lambda_{233Pa}+\sigma_c^{233Pa}\Phi}{\lambda_{233Pa}}(\frac{\nu_{Th}-1}{\alpha_{Th}}+D_{233Pa})}*$$

$$\left(\frac{\nu_{233U}-2(1+\alpha_{233U})}{1+\alpha_{233U}}+D_{233U}-2\frac{\sigma_c^{233Pa}\Phi}{\lambda_{233Pa}}+\frac{\lambda_{233Pa}+\sigma_c^{233Pa}\Phi}{\lambda_{233Pa}}(\frac{\nu_{Th}-1}{\alpha_{Th}}+D_{233Pa})-\frac{\Sigma_c^{FP}+\Sigma_c^{Loss}+\Sigma_c^{Else}}{\Sigma_f^{233U}(1+\alpha_{233U})}\right)$$

All the terms appearing at the denominator part of this equation are small when compared to one. The $\frac{\sigma_c^{233Pa}\Phi}{\lambda_{233Pa}}$ term as well as the D_{233Pa} term are negligible compared to one. Even if the ^{233}Pa capture rate is not negligible when compared to its decay rate, it is still very small (about some %). If it were not the case, the Thorium/Uranium cycle would not exist! The $\frac{\nu_{Th}-1}{\alpha_{Th}}$ term is also very small as the fission rate is very small when compared to capture rate for such a fertile material as ^{232}Th. The main term is then $\frac{\nu_{233U}-2(1+\alpha_{233U})}{1+\alpha_{233U}}$. It could have been found more easily, on the base of other strong assumptions 14)]. This methodology allows us to quantify all contributions, including that of ^{233}Pa capture for instance. It allows also estimating the sensitivity, as the “direct term” of the sensitivity can be obtained from the derivation of the previous equation.
This methodology would give very comparable results for reactors with faster spectrum. Only the most important average cross sections appear in this study and the sensitivities should be of the same order of magnitude. This means that the conclusions could be extrapolated to other reactor types and to other spectra.

Data	Uncertainty (%)	Sensitivity (%/%)	ΔRG/RG	ΔRG (pcm)
Nu U233	1	1092	11	2200
XS(cap) U233	10	-164	16,3	3260
Nu Th232	2	14,6	0,3	60
XS(cap) Th232	10	8,5	0,85	170
Nu U235	0,5	135	0,67	134
XS(cap) U235	5	-51,5	2,57	514
XS(cap) Pa233	10	-28,5	2,8	560
XS(cap) Np237	10	-1,95	0,195	39
Total			21	4200

Table**1**. Nuclear data sensitivities and uncertainties of the regeneration of a critical Molten Salt Reactor at equilibrium.

Based on this estimation of the sensitivities and of the nuclear data uncertainties[7], we can calculate the global uncertainty of the regeneration of a Thorium Molten Salt Reactor (see Tab. 1) that is kept critical and whose heavy nuclides are infinitely recycled.

Two nuclear data seem to be responsible for the major part of the total uncertainty:

- ^{233}U fission neutron yield. The information used to interpolate this yeld is quite poor when compared to ^{235}U or ^{239}Pu fission neutron yields. One can understand that the uncertainty on this data is even less well known. Thus, this result should be used with caution. Eventually, this result confirms the extraordinary importance of neutron yields for fission reactors.
- ^{233}U capture cross section. ^{233}U is a very good fissile material. This means that its capture cross section is rather low when compared to the fission one. This explains the difficulty for measuring this cross section and then the spread of the scarce data available in the EXFOR experimental data base.

The 4200 pcm of uncertainties on the calculation of RG must be compared to the sensitivity of RG to the reprocessing facility capacities. Direct calculations show that RG increases by 4000pcm if the reprocessing time of the full volume of salt is halved (i.e. decreased from 6 months to 3 months). Furthermore, stopping the extraction of Minor actinides or of Protactinium would simplify the reprocessing scheme but reduce the RG by 2000 pcm.

Even if this paper studied an epithermal reactor, the main results should be more or less true for a broad range of spectra. Recent work[15] in the field of Molten Salt Reactors shows a trend toward concepts using harder spectra without graphite at all in the core. The nucleus and the cross sections involved in the study of RG being hardly independent of the spectra, the importance of an accurate capture cross section of ^{233}U up to the fission spectrum is likely to remain constant.

Any effort done to reduce the uncertainties on neutron yields and capture cross section down to less than 1% and about 5% respectively or at least to confirm their uncertainty would be very much appreciated. If the uncertainty on RG could be reduced down to 2000pcm, the needed reprocessing scheme could be designed with more confidence, and then the research on specific goals such as minor actinide and protactinium extraction could be prioritarized.

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[7] The very small RG of the particular concept studied makes sensitivities and relative uncertainties unusually high.

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