

THE ROMANIAN IRRADIATION TESTS PREPARED FOR IAEA/OECD DATABASE

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Abstract

The main features of two Romanian experimental fuel elements prepared for IAEA/OECD IFPE Database are described. The particular features of the time evolution of the internal fission gas pressure during the experimental fuel irradiation are analyzed. Based on the micro-structural data in regard with the Romanian route of the CANDU type fuel fabrication the discrepancies versus the empirical densification laws used to predict the CANDU type fuel behavior during irradiation will be pointed out.

Introduction

A particular characteristic of the CANDU fuel project is the small plenum (void volume) added for relaxation of the fission gases (~3% from the fuel stack volume). That means that small variations of the fuel densification thresholds give in effect high variations of the fission gas pressure inside the fuel element. To point out this expected effect two irradiation tests were performed between the years 1985-1987 in TRIGA 14 MW reactor from INR.

The effect of the fuel density on the time-evolution of the fission gas pressure has been evaluated.

Experimental fuel elements were adequately instrumented with pressure transducers to follow the time evolution of the fission gas pressure during the fuel irradiation. The first irradiation test was conducted on the experimental fuel element noted EFE 89, whose main characteristics were the nominal values of the main fuel design parameters. The second one was conducted on the experimental fuel element noted EFE 51 whose pellets density was at the upper limit of the manufacture requirement. Because of the axial flux asymmetry of the TRIGA reactor core, the experimental elements are shorter in length than the CANDU fuel design. Consequently, the axial gap was changed to a smaller value.

2. Fuel Elements Characterization

The main fabrication characteristics of EFE 51 and EFE 89 are given in the follows (see Ref.[1]).

FUEL PELLETS	EFE No.89	EFE No.51
Diameter (mm)	12.149 – 12.158	12.167 – 12.174
Length (mm)	12.846 – 13.842	12.511 – 13.520
Enrichment (wt. %)	3.92 – U ²³⁵	7.04 – U ²³⁵
Pellets density (g/cm ³)	10.539 – 10.615	10.681 – 10.714
Stoichiometry, O/U	2.0055	2.0077
Grain size average, (μm)	10.2	14.5
Roughness (μm, rms)	0.54 – 0.66	0.56 – 0.60
ROD PARAMETERS	EFE No.89	EFE No.51
Pellets Number	22	23
Length of the fuel stack (mm)	292.1	294.8
Axial gap (mm)	1.85	1.70
Diametric gap between pellet and cladding (mm)	0.084	0.080
Graphite thickness (μm)	6.8 - 9.6	5.18
Cladding material	Zy-4	Zy-4

Other differences:

- The fuel element EFE 89 has the same characteristics as an outer element of the CANDU type fuel bundle, with brazed appendices (inter-element spacers and bearing pads), while the EFE 51 is simpler without any appendices.

- The U²³⁵ enrichment was 3.92 %wt at EFE 89 and respectively, 7.04%wt. at EFE 51 to allow for the designed linear power of 550_{-20}^{+30} W/cm.

Besides, the two experimental fuel elements were shorter than the typical CANDU fuel design to allow for a constant axial distribution of the neutronic flux in TRIGA 14MW reactor. The smaller amount of fission gas expected to be released during the fuel irradiation and the particular external shape of the end caps to allow their compatibility with the irradiation device C2 and the pressure transducer, were considered in the design value of the void volume. Thus, one of the end caps is axially crossed by a hole to let the fission gasses to push on the stainless steel membrane of the pressure transducer at the both fuel elements. The detailed working scheme of the mechanism of the pressure measurement is given in Ref.[1].

The TIG technology was used to weld the pressure transducer with its corresponding fuel element end cap. The resistance welding was used for the other

end cap. As already mentioned, the EFE 89 had the same characteristics as an outer fuel element of the CANDU type fuel bundle. Its design values are close to the nominal ones of the fuel geometry, fuel density, radial gap, and brazed appendices (inter-element spacers and bearing pads), of the Romanian made CANDU type fuel element. As for example, the appendices are made of zircalloy-4 and they are attached to the cladding by brazing with beryllium. The EFE 51 is simpler without any appendices, but with a higher density of the fuel pellets.

The impurity content of the initial powder was within the admitted limits of the powder specification to the both fuel elements.

On the inner surface of the claddings there was a small graphite layer (CANLUB) whose thickness is given above.

Experimental results

The specified irradiation requirements were similar for both fuel elements EFE 89 and EFE 51, with only the precaution of a smaller pH of the cooling water (6.2-6.8 for EFE 51 versus 9.5-10.5 as was required for the previous irradiated EFE 89).

The intention of having almost identical irradiation conditions as in a CANDU reactor core, in the case of the EFE 89, conducted to the underestimation of the chemical activity of the water within the irradiation device C2 (made from low carbon stainless steel) versus the design materials of the primary coolant loop of a CANDU reactor core. A high oxidation of the EFE 89 resulted. Spots of increased oxide layers and oxide peeling are revealed in Fig.1. Analogously, in Fig. 2 where a transversal section from a cladding sample is presented, nodular corrosion of great thickness (up to 240 μm) was detected locally. The maximum thickness of the uniform oxidation layer measured on the inner surface of the cladding was about 6.5 μm .

The EFE 51 was characterized by a small oxidation. The outer cladding surface was covered almost totally with an un-adherent dark-grey oxide. In some regions the cladding was clean without any depositions.

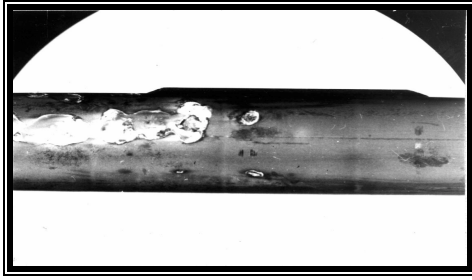


Fig.1 Oxidized regions on the cladding surface detected by visual examination at the EFE 89

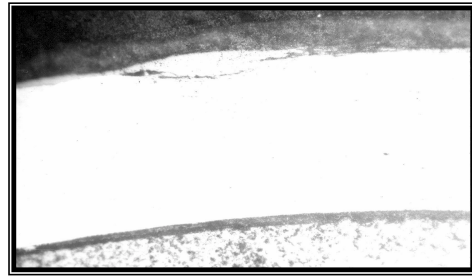


Fig.2 Nodular corrosion on the cladding outer surface at the fuel element No.89

A small oxide peeling was detected at the center of a brown-red region (Fig.3). Small hydride plates circumferentially oriented have been detected inside the zircaloy cladding at the EFE 51 (Fig.4).

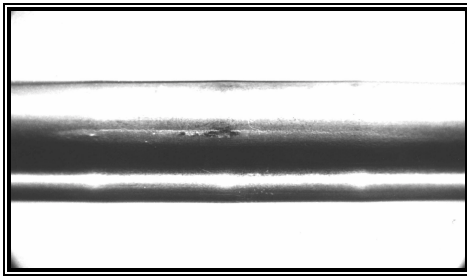


Fig.3 Small oxide peeling on the cladding surface detected by visual examination at the EFE 51

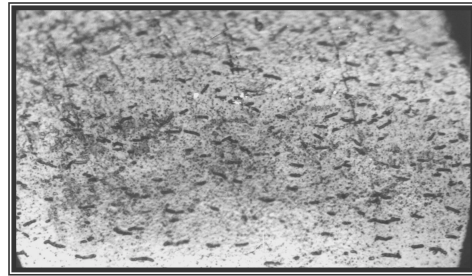


Fig.4 Small hydride plates detected in transversal section on cladding samples from the EFE 51

The inner surface of the cladding of the experimental fuel element was little affected by corrosion. On large areas from the cladding samples only small islands of ZrO_2 of very small thickness ($< 2.6\mu m$) on the outer surface were observed.

An extended area of fuel re-structuring with a large columnar region has been observed at the EFE 89 (~ 0.44 fraction from the fuel pellet radius). At the EFE 51 the presence of the columnar region is not well distinctive, but an extended equiaxed region similar (with EFE 89) was observed (~ 0.64 fraction from the fuel pellet radius).

The maximum mid pellet cladding deformation measured was $\sim 1.5\%$ at the EFE 89 and respectively $\sim 0.55\%$ at the EFE 51.

While the irradiation history was almost constant at the EFE 89 (Fig.5), with a maximum of $\sim 590 W/cm$ total linear power on the second irradiation period, the EFE 51 had a maximum linear power ($526-540 W/cm$) during the beginning of irradiation (up to $\sim 60 MWh/kgU$) and a short ramp $\sim 520 W/cm$ at the end of irradiation because of the advanced burnup of the TRIGA reactor core at that time (Fig.6).

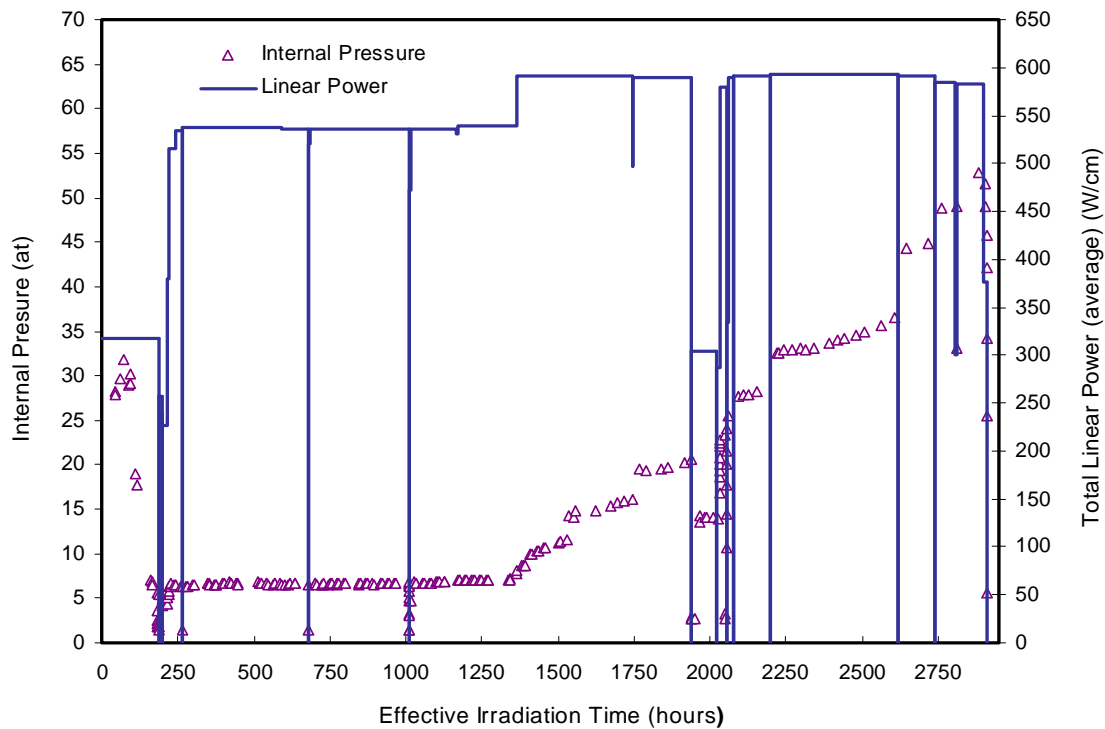
The fuel burnup at the EOL of EFE 89 was 137.6 MWh/kgU and the fuel burnup at the EOL of EFE 51 was 159.3 MWh/kgU.

From experimental measurements the fission gas pressure increased for a short period at the beginning of irradiation in both cases analyzed probably because of the initial water moisture of the internal atmosphere.

The main observations in regard with the experimental fuel element No. 89 are:

- At the beginning of the irradiation the internal pressure of the fuel element takes the highest value of 32 at. However, it slowly goes down to 5.5-6 at. up to the end of the first level of constant power.

Fig.5 Irradiation Test in C2 device, Camp.V, Fuel Element No.89.



- Over the time period between 200-1200 hours of effective irradiation the internal pressure did not change significantly.

- After ~1300 hours the internal pressure begins to increase continuously with the burnup.

- The maximum internal pressure (~ 53 at.) has been measured toward the end of the fuel irradiation (~ 2883 hours).

Some stepwise changes of the linear power were done at ~1000, 2030, 2060 and 2900 hours in order to verify the pressure transducer sensibility. It is easily to

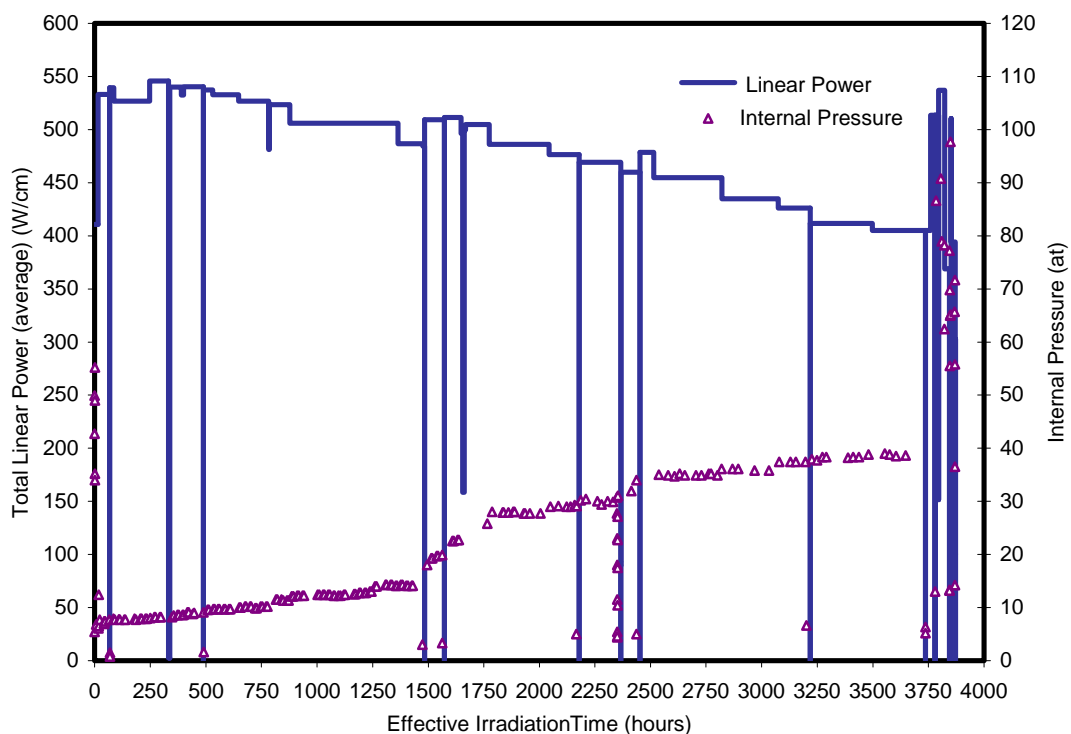
see in Fig.5 that the internal pressure followed the power changes on the all power tests.

- At the EOL, the internal pressure of the EFE 89, measured with the pressure transducer, was 5.62 at. [1].

The main observations in regard with the experimental fuel element No. 51 are:

- In the first minutes of the irradiation the internal pressure of the fuel element increased abruptly to ~50 at. and it slowly goes down to 7 at., after 1.35 hours from the beginning of irradiation.

Fig.6 Irradiation Test in C2 device,Camp VII, Fuel Element No.51



- Over the time period ~1475 hours of effective irradiation (~65MWh/kgU) the internal pressure slowly increased to ~14 at.

- After ~1475 hours the internal pressure increases continuously with the burnup and it becomes ~98 at. on the last time-step of maximum linear power of the fuel element.

- At the EOL, the internal pressure of the EFE 51, measured with the pressure transducer, was 14.2 at. [1].

As already mentioned, some stepwise changes of the linear power were done at ~104 MWh/kgU (~2400 hours) and ~158 MWh/kgU (~3840 hours). It is easily to see that the internal pressure followed the power changes on the all power tests.

The total volume of fission gas collected by fuel element puncturing was 10.79 cm³ STP at the EFE 89. The measured gas pressure was 8 at. and the resulted void volume at the end of the fuel irradiation was 1.22 cm³.

Unfortunately, the fission gas has been loosed during the fuel element puncturing and we have not similar data in the case of the fuel element EFE 51.

Discussion

Because of the fission gas losing during the puncturing of the EFE 51 and the unexpected outer cladding corrosion at the EFE 89 it is difficult to make a direct comparison between the two described experiments. However the effect of the fuel density on the fission gas internal pressure can be pointed out. Before to deal with the results comparisons, some discrepancies between the irradiation conditions will be described in the follows:

- The maximum linear power at the EFE 89 was with ~10% greater than the case of EFE 51, and the fuel burnup at the EOL in the case of EFE 51 was with ~20% greater.

- While the power history of the EFE 89 was almost constant, a small power ramp existed at the end of irradiation of EFE 51.

- The smaller pH of the cooling water during the irradiation of the EFE 51 resulted to a good aspect of the corrosion layer on its outer cladding surface.

- The measured fuel element deformation at the end of the fuel irradiation was lower at the EFE 51.

- The measured internal pressure at the EOL of EFE 51 was about 2.7 times greater than in the case of EFE 89.

- An extended columnar region was observed at the EFE 89 while this is not present at the EFE 51 as it is claimed in Ref.[2].

The greater linear powers and the increased corrosion layer, which acts as a thermal barrier at the outer surface of the cladding, sustain greater fuel temperature of the EFE 89 versus EFE 51.

This is also sustained by the as-observed activation of columnar grain growth in central region of the fuel pellets at the EFE 89. An extended equiaxed region was observed at the EFE 51 but columnar grain growth is not well distinctive. Other two arguments sustaining the greater central fuel temperatures at the EFE 89 are the

greater radial cladding deformations and the greater measured internal pressures on the second irradiation period at the EFE 89. It must be remembered that the initial density of the EFE 89 was smaller than the initial density of the EFE 51. Consequently, a greater void volume to accommodate the amount of fission gas released will result at the EFE 89 during the irradiation, because of the fuel densification.

Based on the observations above, two new comments can be given in regard with the associated void volume for fission gas accommodation. Firstly, we have to take into account that direct comparisons between the two experimental fuel elements can be made only inside the shorter effective irradiation time corresponding to the EFE 89 (2912 hours).

Now, because the initial density of the fuel pellets was greater at the EFE 51 it would be expected a lower internal void volume of this element at the end of irradiation. At a nominal fuel porosity of ~3.3% (10.6 g/cm^3) and a complete densification of the fuel pellets, about 1.1% contraction of the fuel stack length would be present inside the fuel element at the end of the fuel irradiation. Because the initial plenum length is about 1% from the fuel stack length then, if the fuel swelling is neglected, the final plenum volume would be two times greater as initial. But the initial pellets density of the EFE 51 is similar with the resulted density after 280 hours re-sintering tests at 1700°C as described in Ref.[3,8]. Thus, the fuel pellets densification at the EFE 51 can be neglected but the EFE 89 would have a greater internal volume at the EOL. That means that the greater internal pressure at the EFE 51 on the first irradiation time period (~2540 hours) could be due to its smaller void volume for fission gas accommodation. The greater internal pressure of the EFE 89 after this time could be due to the columnar grain activation and consequently a greater amount of fission gas release in void volume of EFE 89 on its last irradiation period (~2300÷2912 hours).

Another particular aspect, in regard with the last irradiation period of the EFE 51, is the fast increasing of its internal gas pressure at the end of irradiation. It must be mentioned that the low power ramps after 154MWh/kgU (3736 hours) at the EFE 51 were interrupted by two reactor scrams in only two days. Moreover, the internal gas pressure registered with the transducer increased of about two times on this short period, which means that large variations of the fuel temperatures as a result of

the reactor power changes allowed for the fuel micro-cracking and consequently a larger amount of fission gas release.

All attempts of using the same empirical densification laws as previously established on the Canadian manufactured CANDU type fuel conducted to constant under-prediction of the experimental results [2, 6, 9]. Even in the framework of the same specifications, the different technology of the fuel fabrication could give small differences of the fuel microstructure whose time evolution during the fuel irradiation must be evaluated. As an example, the different pores size distribution of the Romanian manufactured [3,4] versus the Canadian manufactured [5] CANDU type fuel pellets (see Fig.7) involve different kinetics of densification and gaseous swelling [8].

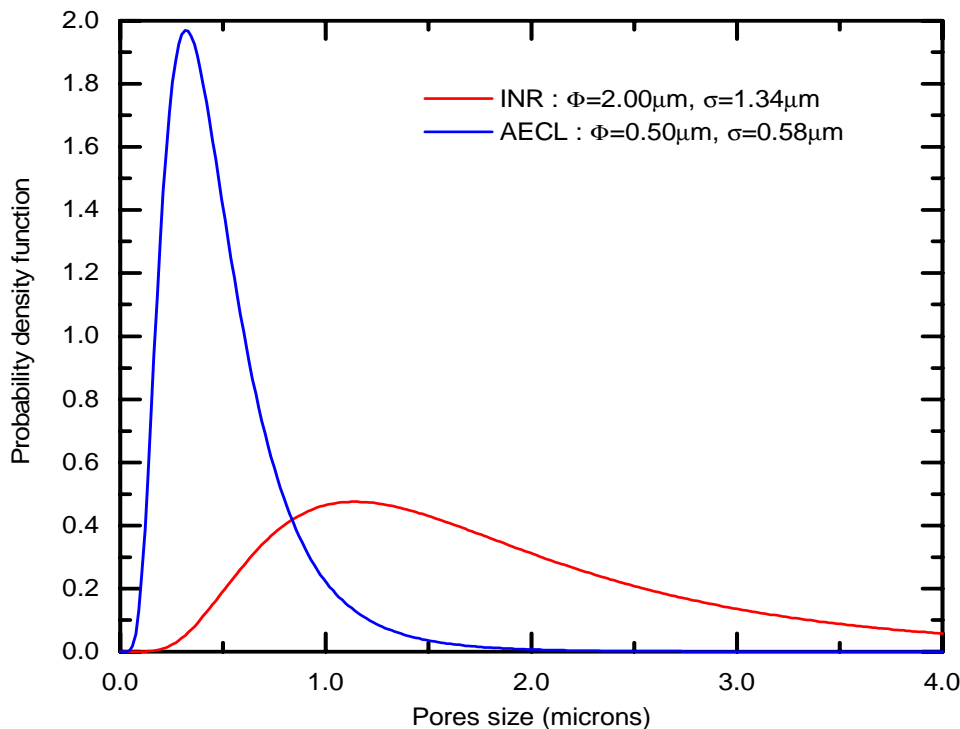


Fig.7 The pores size density probability functions of the Romanian manufactured [1] versus the Canadian manufactured CANDU type fuel [2].

Both processes will compete to the volume changes of the fuel pellets and becomes important for the case of the CANDU fuel project whose characteristic is the small plenum added for relaxation of the fission gas pressure. Thus, greater fabrication densities of the fuel pellets than the nominal value could involve larger diametrical and axial deformations and smaller internal volume to accommodate the fission gas release during the fuel irradiation.

This explain why the predicted fuel deformations using empirical densification laws previously established on Canadian manufactured CANDU type fuel were smaller than measured at the both experimental fuel elements [2,9].

References

- [1] M.C. Paraschiv, 'Romanian irradiation tests', OECD/IAEA, IFPE Database, 2001
- [2] I. Arimescu, St. Ion, ' Report regarding the density effect of the UO₂ pellets on the time evolution of the fission gas pressure during the fuel irradiation'- Internal report No.2573/05.09.1988
- [3] A. Paraschiv, M. Paraschiv, S. Ion, Internal Report INR RI-2764 (1989)
- [4] I. Popov, C. Gentea, L. Stoica, C. Hadanceanu, Internal Report INR RI-2744(1989).
- [5] I.J. Hastings, T.J. Caster, R. Dosieva et. all., CANDU Fuel Performance: Influence of Fabrication Variables, AECL, MISC,250,1983
- [6] C. Gheorghiu et. all, Internal Report INR, RI1274(1983)
- [7] St. Mehedinteanu et. all. Internal Report INR, RI-2309(1986)
- [8] M. C. Paraschiv, A. Paraschiv, V. V. Grecu, J. Nucl. Mater. 302 (2002) 109-124
- [9] C. Gheorghiu, E. Gheorghiu, A. Paraschiv, M. Paraschiv, O. Budan, I.Arimescu , ' Report regarding the density effect of the UO₂ pellets on the time evolution of the fission gas pressure during the fuel irradiation'- Internal report No.2130/20.12.1986