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Journal of Nuclear Materials 274 (1999) 120–126

journal of nuclear materials

www.elsevier.nl/locate/jnucmat

Validation efforts for the neutronics of a plutonium–erbium–zirconium oxide inert matrix light water reactor fuel

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Abstract

Light water reactor (LWR) neutronics codes and cross-section libraries need further qualification when used for the calculation of inert matrix fuel (IMF) cells. Three types of validation efforts have been undertaken for the PuO_2–Er_2O_3–ZrO_2 IMF concept under development at the Paul Scherrer Institute (PSI). Firstly, the PSI calculational scheme, based on the BOXER code and its data library, has been applied to the analysis of a range of LWR experiments with PuO_2–UO_2 fuel, conducted earlier at PSI's PROTEUS facility. The generally good agreement obtained between calculated and measured parameters gives confidence in the ability of the employed calculational scheme to correctly modelize Pu-containing fuel cells. Secondly, reactivity effects of various burnable poisons in a ZrO_2 matrix were measured in the CROCUS reactor of the Swiss Federal Institute of Technology at Lausanne. Modelling these experiments with BOXER resulted in satisfactory prediction of measured reactivity ratios (relative to a soluble-boron standard) for most of the experimental rods employed. This was particularly the case for experiments with erbium, as well as with mixtures of erbium and europium (the latter being used to simulate the effects of overlapping resonances, as would be expected in the case of a Pu–Er IMF). Finally, as there are no experimental results available from power reactors employing IMFs, the validation of burnup calculations (at the cell level) has been based on results obtained in the framework of an international benchmark exercise on the physics of LWRs employing IMFs. Certain discrepancies in calculated parameters have been observed in this context, several of which can be attributed to specific differences in cross-section libraries.

1. Introduction

During recent years, large plutonium inventories have accumulated and continue to grow throughout the world. This is an unwanted situation because of proliferation risks and the large costs involved for safe storage. There is thus a strong incentive to reduce the Pu-stockpiles by burning the excess quantities in power reactors. An efficient reduction method is offered by the possibility of employing an inert matrix fuel (IMF) in light water reactors (LWRs). One such fuel concept, under development at the Paul Scherrer Institute (PSI), is based on the use of a PuO_2–Er_2O_3–ZrO_2 solid solution [1].

Reactor physics calculational methods and data are currently well validated for the present-day fuels used in LWR power plants. However, they cannot be applied to the proposed IMF configurations without further qualification because new materials, such as erbium as burnable poison in the PSI concept, are incorporated into the fuel while others, in particular ^{238}U, are completely absent. The resulting fuel thus represents a mixture which is neutronically quite different from those in common use.

Till now, there has not been any major experimental programme on LWR lattices employing IMFs. Ideally, the calculational methods should be tested on experimental configurations in which the neutron spectra are as close as possible to those in the proposed power reactor situation. Certain experiments performed earlier at PSI's PROTEUS critical facility [2] meet the conditions

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0022-3115/99/$ – see front matter
PII: S0022-3115(99)00086-0

on the neutron spectrum, in spite of the fact that the PROTEUS test zone was loaded with mixed oxide (MOX) fuel. In Section 2, comparisons are made between measured results from these experiments and values obtained applying the PSI calculational scheme based on the BOXER code [3] and its cross-section library BOXRAN. There were, however, no burnable poisons in PROTEUS. Accordingly, a new series of experiments on the reactivity effects of a range of burnable poisons was conducted in the CROCUS critical facility at the Swiss Federal Institute of Technology, Lausanne (EPFL) [4]. Comparisons between measured and BOXER-calculated results from these experiments are discussed in Section 3.

Another valuable source of information about the quality of physics calculations for LWRs with IMFs is the comparison of results obtained employing different independent calculational schemes. A numerical benchmark exercise was launched recently in this context, with the participation of a number of institutions from several different countries [5]. This exercise is particularly important because it allows the testing of methods and data not only for beginning of life (BOL) conditions, as is the case with zero-power experiments, but also as function of burnup. Some results from comparisons made in this framework are presented in Section 4.

Finally, Section 5 summarizes the principal conclusions to be drawn from the current validation efforts for the PSI calculational scheme as applied to the PuO_2–Er_2O_3–ZrO_2 IMF concept.

2. Modelization of PROTEUS–LWHCR experiments

The phase II experiments on light water high conversion reactors conducted in PROTEUS, so called PROTEUS–LWHCR [2], mainly involved the investigation of undermoderated LWR lattices with PuO_2–UO_2 of about 11% Pu (total) content as fuel. The Pu-density in the fuel rods was thus ~0.94 g/cm^3 Pu (~72 wt% Pu_{fiss}), and the moderator-to-fuel volume ratio (M/F) varied from 0 (core without moderator) to 2.07. In comparison, the IMF rods considered in the PSI concept contain 0.9 g/cm^3 Pu (67.2 wt% Pu_{fiss}), together with 0.3 g/cm^3 erbium as burnable poison, the M/F being 1.90.

The comparison of the calculated neutron spectra in the IMF cell and in the PROTEUS test lattice with the highest moderation (Core 18) shows that there are very similar features in the two cases (see Fig. 1), from which it can be concluded that, from the viewpoint of plutonium, the neutronics in the PROTEUS lattices is quite representative of that of the IMF cell. Thus, these experiments can be used for validating the IMF cell calculations, as far as the treatment of Pu-isotopes is concerned.

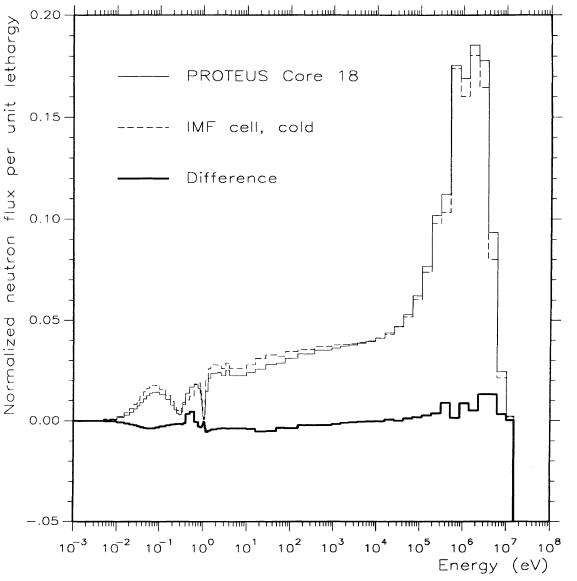

Fig. 1. Comparison of the normalized neutron spectra in a fuel rod of PROTEUS–LWHCR Core 18 and in a PuO_2–Er_2O_3–ZrO_2 IMF rod (infinite array) under cold conditions. Lethargy = $\log(E_0/E)$, with $E_0 = 10$ MeV.

For five different M/F-values, BOXER calculated results are compared with the integral parameters measured in PROTEUS (see Table 1). The Core 18 test lattice was a heterogeneous configuration which cannot be modelled exactly by BOXER. To account for the differences between the heterogeneous and simulated homogeneous lattices, correction factors for the various parameters were reported earlier [6], and these have been used in making the present comparisons.

The range of M/F-values covered by the experiments enables a testing of the BOXER calculations also for voided conditions of the IMF cell. The C/E-values in Table 1 indicate that most of the BOXER results for k_∞ and the measured reaction rate ratios are within the 1σ uncertainty of the measurements. The largest discrepancies are discussed hereafter:

- In Core 8, the BOXER result reflecting capture in ^{242}Pu is 19% higher than the measured value. It seems that ^{242}Pu capture cross-sections in the fast energy range in the data library employed are much too high. However, this discrepancy does not play any role in the k_∞ result for Core 8, because ^{242}Pu capture accounts for less than 0.5% of the total absorption in this lattice.
- It is interesting to note the variation of the C/E-value for C8/F9, the most important single reaction rate ratio from the neutron balance viewpoint in the PuO_2–UO_2 lattices. It seems that ^{238}U capture is slightly underestimated in strongly voided cases and overestimated in the better moderated cells, the more

Table 1
Ratio of BOXER-calculated (C) to experimental (E) results for the multiplication factor k_∞ and various reaction rate ratios, for five PROTEUS–LWHCR test lattices with different moderator-to-fuel volume ratios (M/F). The Δs represent the measurement uncertainties (1σ). C: capture rate; F: fission rate; $5 = {}^{235}U$; $8 = {}^{238}U$; $9 = {}^{239}Pu$; $1 = {}^{241}Pu$; $2 = {}^{242}Pu$

Core	8		9 [a]		7		13		18	
M/F	0.0		0.28		0.48		0.95		2.07	
	C/E	Δ(%)	C/E	(%)	C/E	Δ(%)	C/E	Δ(%)	C/E	Δ(%)
k_∞	1.007	±0.7	0.998	±0.4	0.999	±0.4	1.000	±0.5	1.021	±1.0
F5/F9	0.985	±1.3	1.016	±1.5	1.016	±1.5	1.024	±1.7	0.982	±2.0
C8/F9	0.955	±1.6	0.985	±1.7	1.016	±1.8	1.058	±2.0	1.052	±2.2
F8/F9	1.019	±1.8	1.023	±1.8	1.038	±1.9	1.049	±1.9	1.002	±2.2
F1/F9	0.984	±3.0	0.974	±5.0	0.989	±3.0	1.012	±2.5	nm [b]	
C2/F9	1.203	±3.0	0.988	±3.0	0.983	±3.0	0.988	±4.0	nm	

[a] In Core 9, water was replaced by Dowtherm in order to simulate H_2O-voidage.
[b] Not measured.

consistent C/E-values for F8/F9 and F5/F9 being indicative of adequate accuracy for predicting the ^{239}Pu fission rate as such. With ^{238}U completely absent in the case of the IMF cell, discrepancies in the prediction of ^{238}U capture are clearly of no consequence in the present context.

- The largest differences appear in Core 18, which should be the most representative for standard LWR lattices under cold conditions. Here, one has to remember that the test lattice was not regular and that this renders the modelization difficult. In other reported calculations for Core 18, analogous trends were found to those obtained with BOXER, overestimations of k_∞ of upto 3% being reported [6].

Based on the generally satisfactory agreement of the calculated results for k_∞ and reaction rate ratios involving the Pu-isotopes, it can be said that BOXER with its cross-section library should be able to make reasonable predictions for IMF cells, at least as far as plutonium is concerned.

3. Reactivity effects of burnable poisons

In establishing an IMF concept for LWRs, it is often necessary to incorporate a burnable poison in order to reduce the large burnup-dependent reactivity swing with such cells. While various experiments have been reported in the past for gadolinium, there has been a lack of relevant integral measurements for other suggested burnable poisons such as erbium (as employed in the PSI concept). In order to help fill this gap, a series of reactivity measurements were carried out in the CROCUS reactor at EPFL [4]. This was done for a range of burnable poisons which, in the form of oxides, had been mixed with ZrO_2 and sintered into pellets (of density ~4.6 g/cm^3) before being introduced into aluminum tubes with inner and outer diameters of 6.0 and 8.0 mm, respectively.

CROCUS is a two-zone, H_2O-moderated critical facility, the inner zone consisting of UO_2 rods of 1.806 wt% enrichment and the outer zone having metallic uranium rods of 0.947 wt% enrichment. For the measurements, the reactor was first made critical with one of the burnable poison absorber rods inserted into the centre of CROCUS (see Ref. [7] for a detailed description of the experimental configuration). The reactivity increase induced by withdrawing the absorber rod was determined by measuring the stable period and employing the inhour equation. The reactivity effect was calculated with BOXER by modelling the reactor configuration with and without the central absorber rod.

In comparing experimental and calculational results, it was found that interpretation of the 'absolute' reactivity effects entailed considerable systematic errors, principally those related to uncertainties in the CROCUS kinetic parameters used for solution of the inhour equation. Thus, differences of ~10% in the deduced reactivity values were obtained, depending upon whether one used JEF-1 or ENDF/B-6 based kinetic parameters, the former giving a much better agreement with the calculated values. With the main purpose of the experiments being to qualify the basic nuclear data employed for the individual burnable poisons, the various measured reactivity effects were considered as ratios with respect to the worth of a reference soluble-boron rod, boron being a well-known $1/v$-absorber. The consideration of such reactivity ratios largely eliminated the above type of systematic errors.

First comparisons between CROCUS measurements and BOXER-calculated values were reported earlier for absorber rods with different concentrations of B, Er, Eu and mixtures of Er and Eu [1], as well as for rods with Dy, Ho and Hf [7]. Although good agreement was obtained

[1] Used for simulating the effects of overlapping resonances, as would be expected in the case of a Pu–Er IMF.

Table 2
Comparison of BOXER-calculated (C) and experimental (E) reactivity ratios (relative to the soluble-boron standard) for various burnable-poison absorber rods measured in CROCUS

Poison	Concentration (g/cm^3)	Absorber pellet diameter (mm)	Experiment (E)	Calculation (C)	C/E [a]
B [b]	11700 (ppm)	6.00	1.000	1.000	–
Er	0.747	5.88	1.071 ± 0.008	1.086	1.014 ± 0.007
Eu	0.030	5.75	0.777 ± 0.007	0.775	0.998 ± 0.009
Er + Eu	0.295/0.029	5.87	1.123 ± 0.008	1.139	1.014 ± 0.007
Ho	1.500	5.86	0.836 ± 0.007	0.927	1.109 ± 0.009
Hf	0.484	5.77	0.598 ± 0.006	0.603	1.008 ± 0.011

[a] The uncertainties reflect the statistical errors (1σ) in the experiments.
[b] Soluble-boron standard.

for the various reactivity ratios, the statistical accuracies achieved in these initial experiments were somewhat limited (2–3%, 1σ). This was largely due to uncertainties in the reactivity measurements themselves. The experimental procedure applied at the CROCUS reactor has subsequently been refined, such as to reduce the measurement errors to below 1%. Table 2 presents the main results of the new experiments which have recently been conducted.

The agreement obtained between calculated and experimental results is seen to be excellent (within the 1σ accuracy of the measurements) for Eu and Hf. For Er, the case most relevant for the PSI IMF concept, as well as for the mixture of Er and Eu, the agreement is still satisfactory, the observed discrepancy being only 1.4% in each case. It should be mentioned that the cross-sections for Er used in the above calculations were taken from the Russian data file BROND-2 [8]. Use of JEF-1 cross-sections for the erbium isotopes ^{166}Er and ^{167}Er would yield calculated values 4–5% higher than the experimental results for the Er-rod. In the case of Ho, it is seen that the calculated reactivity effect is as much as ~10% too high, reflecting inadequacies in the cross-sections for this rather exotic absorber material. [2]

A point which needs to be borne in mind is that the neutron spectrum in CROCUS is significantly softer than that of a uranium-free LWR cell employing an IMF such as PuO_2–Er_2O_3–ZrO_2. It has been found, however, that the principal features of the absorption rates of interest (e.g. the ^{167}Er resonance at 0.46 eV in the case of the Er-rod) are adequately reflected in the experimental results [7]. The CROCUS measurements do, therefore, provide a certain confidence in the performance of BOXER as regards the prediction of burnable poison effects.

[2] ^{165}Ho being a strong resonance absorber, the discrepancy can be due either to the basic data or to an inadequate treatment of resonance self-shielding effects by the code.

4. Benchmark exercise on inert matrix fuel cells

The calculation of the characteristics of IMF cells as function of burnup is a challenge, these differing largely from standard LWR cells in that:

- they do not contain ^{238}U which represents more than 95% of the heavy nuclides in the standard case. As such, the neutron spectrum as well as the neutron flux vary much more with burnup than in a UO_2 or MOX cell;
- in many cases, relatively little studied burnable poisons are incorporated into the IMFs in order to flatten the slope of the reactivity curve.

Clearly, the variation of the infinite multiplication factor k_∞ with burnup has to be carefully checked. However, the calculational methods used for other parameters such as the fuel temperature coefficient (FTC), the moderator density coefficient (commonly called the void coefficient, VC) and the boron worth, also need to be qualified. As there are no experimental data for IMF cells under full-power conditions, an appropriate calculational benchmark exercise was launched recently with the participation of various institutions from several countries [5]. Five different fuel materials (numbered 1 to 5) have been considered, the plutonium itself being either LWR-discharged Pu with 58% ^{239}Pu (reactor-grade Pu, RG) or weapons-grade Pu (WG) with 93% ^{239}Pu. In fuel 1, the matrix is really inert, i.e. it is composed of oxides of Al, Mg and Zr, all of which have a relatively low interaction with neutrons. The matrix of fuel 2 contains 35% ThO_2. Both fuels 3 and 4 contain burnable poisons, Er in fuel 3 and ^{10}B in fuel 4, the matrix consisting of ZrO_2. In fuel 5, the matrix consists fully of ThO_2. The case most representative of PSI's IMF concept is fuel 3 (RG-3) with ZrO_2 as the inert matrix and Er_2O_3 as burnable poison. The corresponding results presented below are accordingly indicated in *bold italics*.

Table 3 gives a summary of the comparisons of the k_∞-values at BOL and at end of life (EOL) as calculated with BOXER, with the mean values of all the participants' results. The mean values indicated are

Table 3
Comparison of the multiplication factors k_∞ from BOXER calculations with the mean values of all the participants' results for the different IMF cells considered in the benchmark exercise. σ: average deviation of individual results from the mean value (%); Δ(B): deviation of the BOXER result from the mean value (%)

	k_∞ at BOL			k_∞ at EOL		
Cell	Mean	σ (%)	Δ(B) (%)	Mean	σ (%)	Δ(B) (%)
WG-1	1.6195	±0.30	+0.46	1.0240	±0.92	+0.61
RG-1	1.4505	±0.54	+0.78	0.8151	±1.78	−0.54
WG-2	1.4155	±0.43	+0.38	1.0891	±0.33	+0.19
RG-2	1.2622	±0.54	+0.62	0.9947	±0.40	+0.07
RG-3	***1.1035***	***±0.38***	***+0.31***	***0.9042***	***±1.09***	***−0.76***
RG-4	1.1058	±0.87	+0.71	0.9522	±0.45	−0.34
WG-5	1.2916	±0.59	+0.34	0.9685	±0.18	−0.32
RG-5	1.1899	±0.41	+0.39	0.9921	±0.50	+0.01

based on five different sets of calculations in each case, except RG-3 for which only four results were available.

At BOL, BOXER predictions for k_∞ are systematically higher than the mean values. This may be attributed largely to two different effects:

- Most participants use cross-section libraries based on the JEF-2 data file. It is known that ^{239}Pu from JEF-1 (as available in the BOXER library) is a little more 'reactive' than that from JEF-2 [9]. This was demonstrated for these IMF cells themselves [10], the effect accounting for 0.2–0.4% of the k_∞ overprediction with BOXER.
- In the BOXER cross-section library, the cut-off energy for the thermal energy range is at 1.3 eV. Above this energy, the scattering matrices are calculated with the assumption of scattering nuclei at rest. It has been shown [11] that a more accurate accounting of the thermal motion of the hydrogen atoms for incident neutron energies in the range between 1.3 and ~2.5 eV reduces the k_∞ of IMF cells under full-power conditions (as in the benchmark exercise) by 0.1–0.5%.

The slight k_∞ overprediction with BOXER at BOL can thus be largely explained.

The situation is more complicated at EOL. The average deviations are larger in this case, and no systematic trends can be recognized in the BOXER k_∞ results. As regards the isotopic densities at EOL (details for which are given in Ref. [5]), it has not been possible even here to recognize any systematic trends for the BOXER-predicted densities against mean values of all the participants' results. In any case, there are no undue discrepancies to be noted for the RG-3 k_∞ predictions, relative to the other cases.

In Table 4, the FTCs calculated by BOXER are compared at BOL as well as at EOL against the mean values. At both BOL and EOL, the deviations for the BOXER results are of the same order as the corresponding spread among the various participants' results. It is not possible to give a definitive judgement of the BOXER values before the average deviations, particularly the relatively high ones at EOL, are reduced to an acceptable level. Once again, there is no particular trend observable for RG-3.

The moderator density, or void, coefficient is predicted consistently by the various calculational methods as long as the degree of voidage is small. For 10% void, the results of the different participants agree within a

Table 4
Comparison of the fuel temperature coefficients (FTC, 10^{-5}/K) calculated with BOXER and the mean values over all the participants' results. σ: average deviation of individual results from the mean value (10^{-5}/K); Δ(B): deviation of the BOXER result from the mean value (10^{-5}/K)

	FTC at BOL			FTC at EOL		
Cell	Mean	σ	Δ(B)	Mean	σ	Δ(B)
WG-1	−1.22	±0.08	+0.06	−0.66	±0.18	−0.24
RG-1	−1.34	±0.15	−0.05	−0.59	±0.31	−0.37
WG-2	−3.19	±0.07	+0.05	−2.95	±0.10	−0.13
RG-2	−3.29	±0.17	−0.10	−3.13	±0.20	−0.19
RG-3	***−1.66***	***±0.14***	***−0.17***	***−1.29***	***±0.16***	***−0.20***
RG-4	−0.73	±0.17	−0.27	−1.06	±0.24	−0.31
WG-5	−3.71	±0.09	−0.10	−3.52	±0.13	−0.21
RG-5	−3.71	±0.19	−0.20	−3.63	±0.22	−0.24

few % at BOL as well as at EOL. That the agreement seems to be so good is partly due to the large negative values of the VCs for all cells at low voidage, so that the differences appear small in relative terms. When the voidage is in the range of 90% or higher, the VCs remain sufficiently negative for the cells with fuels 2 and 5 which contain Th, but they are nearly 0 or even positive for the other cells at BOL. Although absolutely not much greater, the differences become very large in relative terms. Table 5 shows a comparison of the BOXER VCs between 0 and 95% void against the mean values of all the participants' results. It can be seen that the BOXER values are systematically more negative. These differences can be explained by the following:

Cross-sections of Zr in the epithermal energy range: For Zr, the BOXER library contains cross-sections taken from ENDF/B-4. It has been shown [10] that these data result in a much higher absorption of epithermal neutrons than is the case with cross-sections from JEF-2 or ENDF/B-6. For the RG-3 cell, the ENDF/B-4 cross-sections for Zr yield a VC between 0 and 95% void which is 25×10^{-5}/% void more negative than that obtained using cross-sections from JEF-2. It can be seen from Table 5, at BOL in particular, that the differences seem to be more or less proportional to the density of Zr in the inert matrix. They are fairly small for the cells of fuels 2 and 5, which do not contain any Zr in the fuel, higher for fuel 1 with 35 mol% ZrO_2 in the matrix and highest for the RG-3 and RG-4 cells where the inert matrix consists entirely of ZrO_2.

Thermal cut-off energy: The earlier discussed effect of the low thermal cut-off energy in the BOXER library can be responsible for an underestimation of upto 5×10^{-5}/% void of the VCs. This follows from the fact that at 0% void the BOXER k_∞ is slightly higher than it would be with an improved model, while the k_∞ value at 95% void is not at all affected due to the negligible importance of thermal neutrons in this case.

5. Conclusions

Three types of validation efforts have been undertaken for the PuO_2–Er_2O_3–ZrO_2 IMF concept under development at PSI. Firstly, the applied calculational scheme based on the BOXER code and its cross-section library has been used to analyse a range of LWR lattice experiments with PuO_2–UO_2 fuel performed earlier in the PROTEUS zero-power reactor. The good agreement between the calculations and the measurements gives confidence in the modelization of Pu-containing fuel cells by BOXER.

Secondly, reactivity effects of different burnable poisons in a ZrO_2 matrix (relative to a soluble-boron standard) were measured in the CROCUS critical facility. The agreement between BOXER calculations and experimental results is in general good, especially for Er as well as for a mixture of Er and Eu. The latter served to simulate the effects of overlapping resonances as would occur in a Pu–Er IMF.

Finally, since there is little experimental evidence available on the burnup physics of IMFs, calculational results from BOXER have been compared with those obtained using a range of other methods and data in the framework of an international IMF benchmark exercise. It has been found that, relative to the other calculations, BOXER slightly overestimates the multiplication factors k_∞ at BOL. This discrepancy can be largely eliminated by:

- using new sets of ^{239}Pu cross-sections instead of the JEF-1 data currently employed;
- improving the scattering matrices for hydrogen used in the epithermal energy range.

There are no clear systematic trends observed in the k_∞ predictions at EOL. As regards reactivity coefficients, significant discrepancies are found in the void coefficients predicted for high voidages, BOXER values being more negative than the other results. For the case most relevant to the PuO_2–Er_2O_3–ZrO_2 IMF under development, the differences can be attributed mainly to

Table 5
Comparison of the void coefficients (VC, 10^{-5}/% void) between 0 and 95% void as calculated with BOXER and the mean values over all the participants' results. σ: average deviation of individual results from the mean value (10^{-5}/% void); Δ(B): deviation of the BOXER result from the mean value (10^{-5}/% void)

	VC at BOL			VC at EOL		
Cell	Mean	σ	Δ(B)	Mean	σ	Δ(B)
WG-1	−42.7	±10.2	−16.3	−405.7	±9.7	−5.9
RG-1	+0.97	±11.4	−18.6	−498.0	±6.3	−4.3
WG-2	−303.9	±5.2	−4.4	−387.2	±5.3	−4.6
RG-2	−275.9	±5.7	−6.0	−390.3	±6.1	−4.7
RG-3	***+72.6***	***±20.0***	***−30.0***	***−431.8***	***±13.1***	***−13.9***
RG-4	+106.9	±17.4	−27.1	−407.7	±12.8	−20.3
WG-5	−480.6	±6.5	−5.9	−495.1	±6.9	−1.9
RG-5	−344.0	±9.0	−8.5	−390.4	±6.6	−4.2

uncertainties in the Zr cross-sections in the epithermal energy range.

Acknowledgements

The authors are grateful to A. Stanculescu, PSI, for useful comments and discussions.

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