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Modelling the Radial Distribution of the Inventory of U and Pu Isotopes in PWR Fuel Rods at High Burnup

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Abstract

Nowadays trends on nuclear reactors are to take advantage of a more efficient use of the fuels. In this way, goals like decreasing the volume of radioactive wastes, increasing life time in the reactor, extended burnup and accident-tolerance fuels are taken as the principal guidelines for the design and construction for actual nuclear fuels.
The development of tools and the capability of prediction of the behaviour of nuclear fuels under irradiation let us design and improve the new demanding conditions without neglecting the security and economics. In this way, the study of the evolution and details of the fissile material of the fuel in particular the ^{235}U inventory evolution and the ^{239}Pu production and evolution are demanding conditions.
We implemented the PILR code, based on the RAPID model, for the radial distribution of U and Pu radioisotopes. The aim of this work is to determine production and disappearance of Pu and U isotopes during the irradiation of commercial and experimental fuel rods.

Keywords: Fuel and Fuel Channels, Code Validation.

1. Introduction

A fresh FE (Fuel Element) contains a well determined hyper-stoichiometric UO2 quantity, with an enrichment given by design for the NPP (Nuclear Power Plant) in accordance with the burnup desired and taking into account safety and security limits.

The fission products of ^{235}U, the appearance of ^{239}Pu from epi-thermal neutron capture in ^{238}U resonances and, consequently, the appearance of fertile ^{240}Pu and ^{242}Pu, and fissile ^{241}Pu determine a dynamic inside the FE that affects the burnup, the material properties (like thermal conductivity, macroscopic cross sections, rim zone, etc.). It is crucial to evaluate the inventory of each isotope mentioned before and its influence in the fuel behaviour.

In view of the raised trends, the SiM3 Division (“Laboratory of Simulation of Materials and Fuel Elements”) belonging to GCCN (Spanish acronym for “Nuclear Fuel Cycle Management”) of CNEA at CAB (“Bariloche Atomic Centre”) is studying the behaviour of LWR’s (“Light Water Reactors”) fuel elements at high burnup conditions. In this scenario,

HBS ("High Burn-up Structures") may appear because of radial variation of local power accordingly to the production of fissile Pu. The calculation of the local variations of U and Pu isotopic concentrations is essential for coupling with others phenomena (RIM effect, thermal conductivity) in a behaviour code.

The basis of the model, the problems of implementation and the solutions proposed will be presented in this study. We will also include the validation of the model using experimental data from PWR ("Pressurized Water Reactor") previously included in the CRP FUMEX II ("Co-ordinated Research Project on FUel Modelling at Extended burnup II") from IAEA ("International Atomic Energy Agency") [1].

2. PILR Code

The PILR ("Pilr Is Like RAPID") code implements simple correlations depending on the enrichment, the power density, the burnup and the radial position to calculate the cross sections ($\sigma_i(t,r)$) and flux ($\Phi(t,r)$) and therefore isotopic concentrations ($N_i(t,r)$) evolution during a fuel rod's irradiation, solving balance equations 1-6. The balance equations based on the one-group and one-dimension diffusion theory are:

$$\frac{\partial}{\partial t}N_{235}(t,r) = -N_{235}(t,r)\sigma_{235}^{a}\emptyset(t,r), \quad (1)$$

$$\frac{\partial}{\partial t}N_{238}(t,r) = -N_{238}(t,r)\sigma_{238}^{a}\emptyset(t,r), \quad (2)$$

$$\frac{\partial}{\partial t}N_{239}(t,r) = -N_{239}(t,r)(\sigma_{239}^{a}\emptyset(t,r) + \lambda_{239}) + N_{238}(t,r)\sigma_{238}^{c}\emptyset(t,r), \quad (3)$$

$$\frac{\partial}{\partial t}N_{240}(t,r) = -N_{240}(t,r)(\sigma_{240}^{a}\emptyset(t,r) + \lambda_{240}) + N_{239}(t,r)\sigma_{239}^{c}\emptyset(t,r), \quad (4)$$

$$\frac{\partial}{\partial t}N_{241}(t,r) = -N_{241}(t,r)(\sigma_{241}^{a}\emptyset(t,r) + \lambda_{241}) + N_{240}(t,r)\sigma_{240}^{c}\emptyset(t,r), \quad (5)$$

$$\frac{\partial}{\partial t}N_{242}(t,r) = -N_{242}(t,r)(\sigma_{242}^{a}\emptyset(t,r) + \lambda_{242}) + N_{241}(t,r)\sigma_{241}^{c}\emptyset(t,r), \quad (6)$$

Where λ_i is the half-life of radioactive isotope i, t the time and r the radial position.

The cross section of reaction to a group contains information on the entire energy spectrum. When is multiplied by an isotopic concentration an effective reaction rate is obtained. Even though there exists a radial dependence of both flux and cross sections in reaction rates, the PILR model considers flat flux across the radial direction.

In that way, neutron flux can be expressed as a function of the power density (POWDEN), enrichment (EN) and burnup (BU) as follows:

$$\emptyset(t,r) = ((C_1^n + C_2^n EN + C_3^n EN^2) + C_4^n BU + C_5^n BU^2) POWDEN \quad (7)$$

Where C_1^n C_2^n C_3^n C_4^n and C_5^n are constants.

The variation against BU for the isotopes with complex resonances as ^{238}U and ^{240}Pu is shown in Figure 1 and Figure 2.

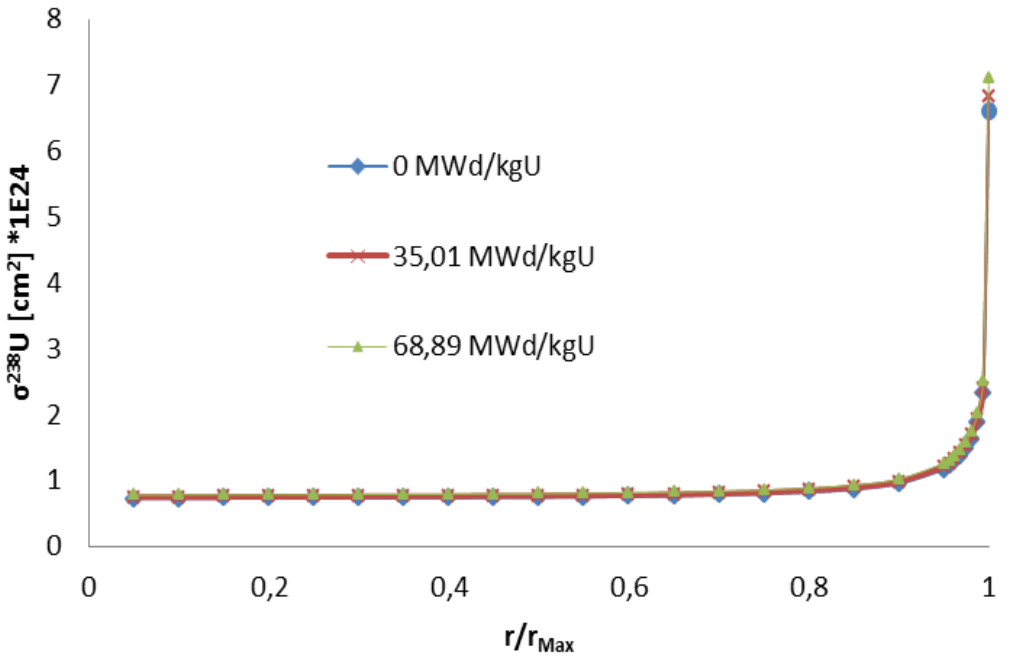

Figure 1 ^{238}U cross section for different BU vs non-dimensional radius.

In ^{238}U XS (Cross Section) correlation, the self-shielding is contemplated and Eq. 8 shows the functional form,

$$\sigma_a^{238}(t,r) = f_a^{238}(r) * \sigma_a^{238}(t) \quad (8)$$

where the temporal part depends on enrichment and BU as the parameter that evolve in time and the spatial part is a function of enrichment and radial coordinate as follows,

$$f_a^{238}(r) = C_1 + (C_2 + C_3 EN) e^{(C_4(1-r)^{C_5})} \quad (9)$$

with C1, C2, C3, C4 and C5 constants.

The radial coordinate is normalized with the outer radius of the pellet in order to cancel the effect of the exponential at the surface of it.

The burnup only presents a minor influence on the ^{238}U XS as a function of r/r_{max}.

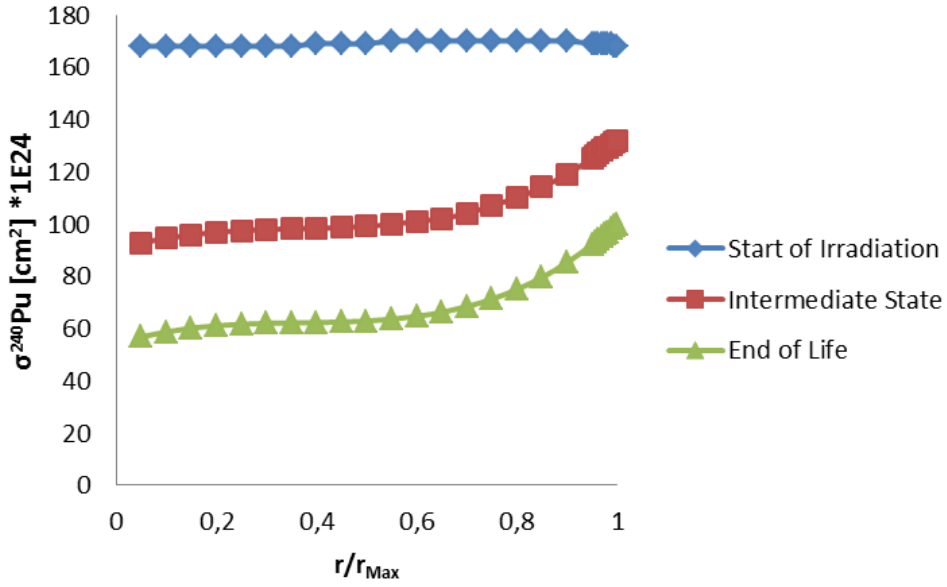

Figure 2 ^{240}Pu cross section for different BU vs non-dimensional radius.

For all the other XS except for ^{240}Pu, even fission, the functional form is the same as in Eq. 8. The radial part is shown in Eq. 10 and the temporal part is shown in Eq. 11:

$$f_i^j(r) = C_1^i + C_2^i EN + (C_3^i + C_4^i EN)r + (C_5^i + C_6^i EN)r^2 + (C_7^i + C_8^i EN)r^3 \quad (10)$$

$$\sigma_i^j(t) = (C_1^i + C_2^i EN + C_3^i EN^2)(C_4^i + (C_5^i + C_6^i EN)BU + (C_7^i + C_8^i EN)BU^2) \quad (11)$$

With i = a (absorption) or f (fission)

j = 235, 238, 239, 241 or 242; and

C_k^i are all constants.

The XS of ^{240}Pu is modelled by Eq. 12

$$\sigma_a^{240}(t,r) = f_a^{240}(r) * (C_1^{BU} + C_2^{BU} * r + C_3^{BU} * r^2 + C_4^{BU} * r^3) \quad (12)$$

Where $C_1^{BU}\ C_2^{BU} C_3^{BU}\ and\ C_4^{BU}$ are cubic functions of BU and

$$f_a^{240}(r) = (C_1^{240} + C_2^{240}BU + C_3^{240}BU^2 + C_4^{240}BU^3)(C_5^{240} + C_6^{240}BU + C_7^{240}BU^2 + C_8^{240}BU^3 + (C_9^{240} + C_{10}^{240}BU + C_{11}^{240}BU^2 + C_{12}^{240}BU^2)EN) \quad (13)$$

with C_i^{240} all constant.

3. Experimental Benchmarks

In this section we present the information of the fuel rod used to validate the code PILR.

3.1 Fuel Rods Built by FRAGEMA-CEA

These fuel rods have been designed and characterized for an irradiation program for the BR3 reactor from CEN/SCK in MOL (Belgium) [3,4,7]. There are some differences from the standard FRAGEMA design in:

- Fuel rod length and free volume (for compatibility with BR3 core)
- "Plenum" length
- Initial enrichment
- Annular pellets with open porosity
- End plugs according to the BR3 core design.

As general characteristics, it has initial enrichment of 7% 235U and Zy-4 cladding.

3.2 Fuel Rods from CEA/OSIRIS

This data set contains details of three standard PWR rods and one segmented rod irradiated in EDF commercial reactors. One full rod and one segmented rod were re-fabricated and ramp-tested in the CEA's OSIRIS reactor to investigate their PCI resistance. The data set contains details of the pre characterization of the fuel pellets, the cladding tube and the assembled fuel rod. The following is an outline of each fuel rod and the data available [5, 7].

This is rod FF0EFELX/H09/5007 irradiated for 4 cycles in the EDF Cruas 2 PWR to a final discharge burn-up of 46.060 MWd/kgU. The average powers in the 4 cycles were approximately 22, 20, 18 and 15 kW/m. The final fission gas release was measured as 0.8 %. Data available from this rod include:

- Length changes measured by neutron radiography.
- Axial diameter (excluding oxide thickness) and oxide thickness.
- Fission gas release.
- Whole pellet density measurements at 11 locations.
- Hydrogen content of the cladding at 4 locations.
- Radial distribution of Cs, Nd, Pu and Xe measured by EPMA at 2 locations.

- Metallography at 2 locations.

4. Results

4.1 Code Validation

An experimental validation was performed against three cases presented in CRP FUMEX II and III of the IAEA [7]. The results shown after were run with both RAPID [2] and PILR versions and the input power history was considered either constant or as per the original reported values.

4.1.1 Results of Modelling Fuel Rod H-09

In Figure 3 we see the power history of one of the axial segments of the fuel rod H-09 [5]. A burnup evolution for three different steps is shown in Figure 4.

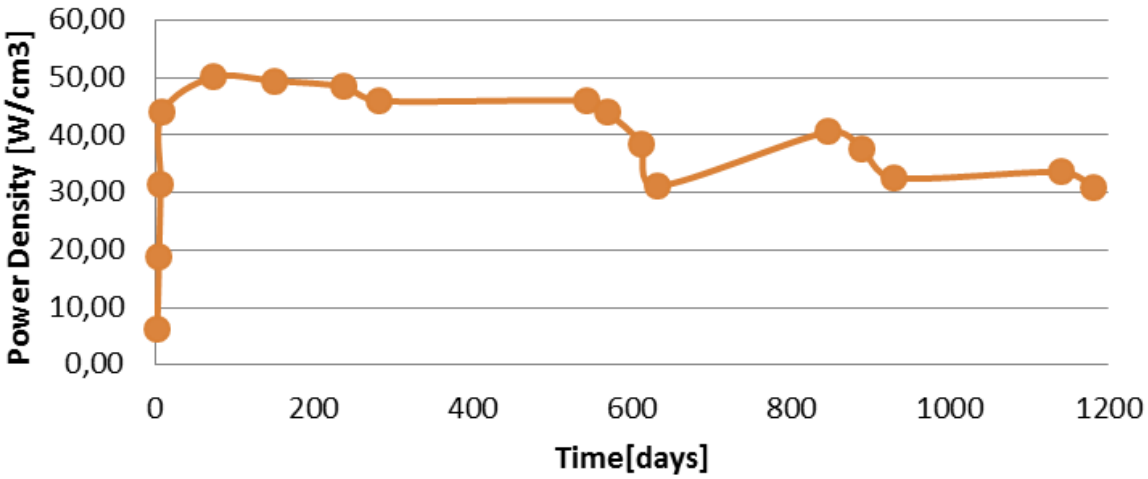

Figure 3. Power history for segment Nº12 of fuel rod H-09.

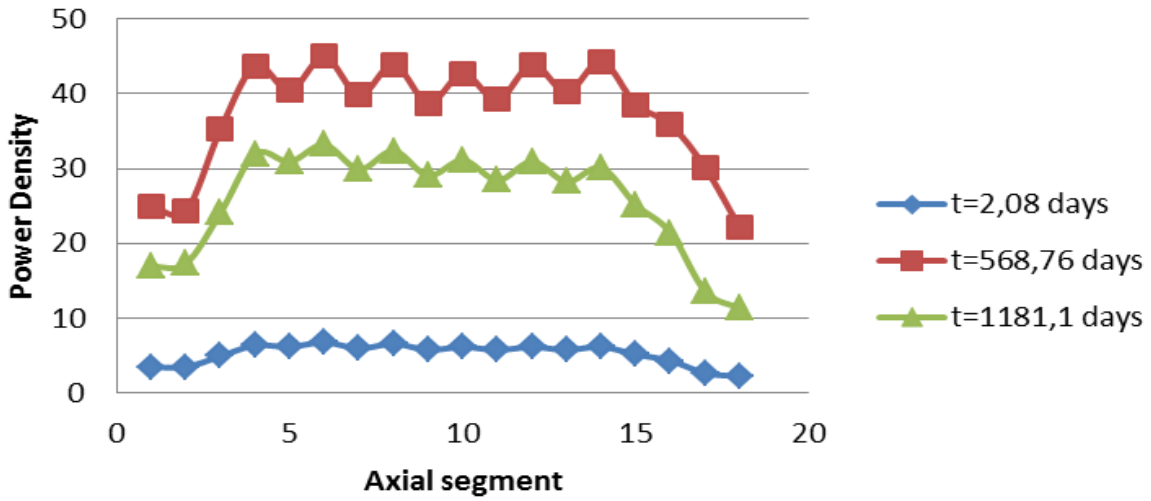

Figure 4. Burnup profile for three different steps of time for fuel rod H-09.

For fuel rod H-09 three different measurements were performed for two axial positions approximately at spatial intervals of one-third length [5,7]. The average value of the three measurements was calculated and contrasted against the calculations of PILR with an average linear power and the experimental value for the sixth segment measured. The results of Figure 5 show good agreement with experimental values.

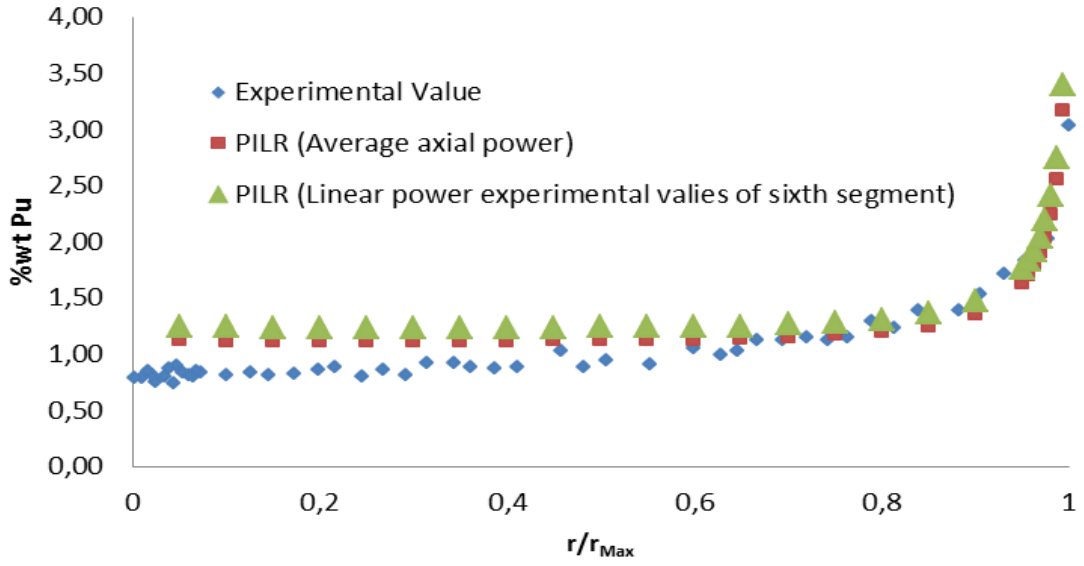

Figure 5. Pu concentration at end of irradiation.

There are no significant differences between the two input modes in PILR. In Figure 4 we showed that the burnup is very flat, so the peaking factor of the fuel rod cannot be too high. In that way the differences of the linear power are very small and accordingly the Pu build-up is almost the same.

At the inner part of the pellet is seen a small overestimation of the concentration calculated. This phenomenon is associated with a higher flux that burns more 238U and consequently produces more Pu than the measured. This happens because the enrichment is fixed and it has not changed at EOL ("End Of Life"), so what could be the appropriate flux for the outer part of the pellet could be a higher flux than the realistic for inner half.

4.1.2 Results of Modelling Fuel Rods BK365 and BK370

In Tables 1 and 2 are shown the isotopic concentrations of U and Pu isotopes calculated and measured at end of irradiation life for fuel rods BK-365 and BK-370 [3, 4, and 7]. There is good agreement between experimental data and our calculations.

The results obtained with the different ways of linear heat power inputs are consistent with the experimental values and with RAPID's results. The differences underlies, at first, in the different numerical methods implemented for the resolution of the differential equations.

5. Discussion & Conclusions

The PILR code for calculating isotopic concentrations of U and Pu as a function of the pellet radius and burnup was implemented. This kind of calculations is suitable to be added in a behaviour fuel code to repower some phenomena like thermal conductivity and the study of the RIM effect.

The results obtained with this model could never be compared strictly with a neutronic code because the XS are modelled to simplify the resonances of crucial isotopes like 238U that contribute to the Pu build-up.

The results from Table 1 and 2 have a small dispersion between them and with respect to the experimental values. The difference lies in the results obtained with the model, are an average value at the end of life of a full rod compared to the experimental value that is a sample measured of the experimental fuel rod.

The results of the model were largely according to the results of post-irradiation experiments of BK-365, BK-370 and H-09 bars belonging to the exercises included in the CRPs FUMEX II and III of the IAEA's programs [3, 4, 5 and 7].

The PILR code is suitable for inclusion in BaCo [6] and the feedback allows aspects of behaviour such as the thermal conductivity of the formation of UO_2 and "rim" structure, among others, to be studied.

Table 1: Isotopic concentrations of Pu and U at end of life fr fuel rod BK-370 calculated with an average axial linear power .

	% wt/wt Total U				% wt/wt Total Pu				
	^{234}U	^{235}U	^{236}U	^{238}U	^{238}Pu	^{239}Pu	^{240}Pu	^{241}Pu	^{242}Pu
Experimental Value	0,03	1,56	1,01	97,4	1,84	51,68	27,62	11,79	7,07
Converted Value to Isotopic Concentration [Atom/cm3]		3,89E+20		2,40E+22		1,10E+20	5,86E+19	2,49E+19	1,49E+19
PILR (pot med) [Atom/cm3]	N/C*	5,65E+20	N/C	1,96E+22	N/C	2,37E+20	7,53E+19	5,57E+19	1,37E+19
PILR (pot var) [Atom/cm3]	N/C	5,55E+20	N/C	1,96E+22	N/C	2,38E+20	7,61E+19	5,67E+19	1,44E+19
RAPID ORIGINAL (pot var) [Atom/cm3]	N/C	5,65E+20	N/C	1,90E+22	N/C	2,30E+20	6,90E+19	5,18E+19	1,29E+19

	% wt/wt Total U				% wt/wt Total Pu				
	^{234}U	^{235}U	^{236}U	^{238}U	^{238}Pu	^{239}Pu	^{240}Pu	^{241}Pu	^{242}Pu
Experimental Value	0.02	0.64	1.15	98.19	5.02	42.57	27.62	12.14	12.65
Converted Value To Isotopic Concentration [Atoms/cm3]		1.55E+20		2.34E+22		1.01E+20	6.54E+19	2.86E+19	2.97E+19
PILR (pot med) [Atoms/cm3]	N/C	2.82E+20	N/C	1.86E+22	N/C	2.21E+20	9.78E+19	7.13E+19	3.04E+19
PILR (pot var) [Atoms/cm3]	N/C	2.69E+20	N/C	1.86E+22	N/C	2.20E+20	9.85E+19	7.21E+19	3.16E+19
RAPID ORIGINAL (pot var) [Atoms/cm3]	N/C	3.21E+20	N/C	1.86E+22	N/C	2.26E+20	9.10E+19	6.77E+19	2.66E+19

Table 2: Isotopic concentrations of Pu and U at end of life for fuel rod BK-365 calculated with an average axial linear power.

6. Acknowledgements

Special thanks to Dr. Chan Bock Lee for unconditional collaboration during the development of the code and willingness.

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