Investigations for the build-up of trans-uranics in the fuel of modern pressurized water reactors

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1 Introduction

The determination of the build-up of trans-uranic heavy materials in the fuel of commonly used modern pressurized water reactors (PWR) is of high interest for a number of issues of the backend of the nuclear fuel cycle. Examples are consequences for the reactivity state of the reactor during burnup, for the cooling conditions after discharge of the fuel and for the handling during disposal or reprocessing. In a closed fuel cycle with reprocessing of the spent fuel and re-use of the recycled material in nuclear reactors, the prediction of the composition of the recycled material is of high importance. Especially the fissile characteristics are decisive; both in view of use in nuclear reactors and with respect to proliferation aspects [1].

Since the beginning of commercial nuclear energy production considerable efforts were spent to gather experimental information about the build-up of trans-uranics and of fission products in irradiated nuclear reactor fuel. However, much of this experimental information is not available in open literature, but handled as confidential know-how of the companies involved. A comprehensive well evaluated experimental program was performed in the first german commercial nuclear light water reactor in Obrigheim (KWO) in the early seventies. The evaluation was carefully carried out in an international collaboration program (Isotope Correlation Experiment, ICE [2]). The results are published in open literature. These experiments are the basis of the experimental validation of the corresponding simulation tools at Forschungszentrum Karlsruhe (e.g [3, 4]). A main objective of the work in reference [5] was to validate, after 25 years of developments of computer hardware, software and of nuclear data, the current versions of the programs and libraries realized during the investigations in reference [4] in the code system KAPROS [6]. In section 2 the successful re-validation of the current version of the codesystem and libraries, is decribed. After this validation work in reference [5] high burnup analysis have been performed. Section 3 analyses the impact of different constant and variable boron concentrations as well as lattice configurations on trans-uranics build-up. In particular, considerable differences are observed for concentrations of minor actinides (neptunium, americium, curium). Extention of the open literature data base for build-up of these elements in high PWR burnup experiments is of high interest for further analysis.

2 Validation of PWR fuel pin burnup with experimental data from ICE KWO

More detailed descriptions of the validation investigations may be found in the references [3, 4, 5]. A short summary is given in the next sections.

2.1 Applied calculation models

The evaluations in reference [2] enable simulation on the basis of pin-cell burnup analysis. In our investigations a one-dimensional three zone Wigner-Seitz cell was applied with the specifications of table 1. Further, mean time dependant boron concentrations are available, as listed in table 2. The irradiation power history of the PWR fuel in the commercial power producing reactor KWO is resolved in some detail, as shown in figure 1. The applied simulation codes (section 2.2) can handle these boron concentrations and irradiation variations.

2.2 Applied codes and libraries

The main objective of this part of the investigations was the re-validation of the burnup capabilities of the current version of the code system KAPROS, developed since the late sixties for the simulation of nuclear reactor systems at Forschungszentrum Karlsruhe [6]. This code system is developed over a long time period in a fast emerging computer environment, starting from an IBM mainframe computer with fast memory space less than 1 megabyte to modern workstations with fast memory on gigabyte level. Moreover, the database for the generation of nuclear constants for the simulations is extended and improved significantly. In this study the standard procedures for pin-cell burnup calculations were applied, using 69 group libraries, varying from the final versions developed in the framework of reference [4] to libraries based on the state of the art evaluated nuclear data files ENDFB/6.5 and JEFF3.0. These newer libraries contain only data directly calculated from the corresponding evaluated nuclear data libraries, without any manual adaptations. It should be noted here that during the original evaluation work the applied multi-group libraries contained few manual adaptations to available experimental information (see reference [4] for details).

2.3 Selected results of the calculations

In reference [5] the validation work of reference [4] is completely covered. Detailed comparisons are presented for all experimental results. Generally, the agreement is very satisfactory. However, in some cases the new results obtained with the standard method for resonance shielding treatment show somewhat larger discrepancies to the experimental data, compared to the original results obtained with improved resonance shielding treatments. A typical result is presented in figure 2 for the overall time behaviour of the isotope Pu^{241}. This isotope has several interesting

characteristics: it has very good fissile properties, has a relative short decay time ($\approx$ 14 years) and the timedependant changes under irradiation contain significant production and losses components. Although other results indicate compensating effects by partial reactions, the overall comparison for the important isotope Pu^{241} is very satisfactory.

2.4 Conclusions from the validation investigations

The re-validation of the code system KAPROS [6] for nuclear reactor simulations in reference [5] shows that, despite the large changes in computer environments and availability of improved evaluated nuclear data libraries, the results remain in accordance with the conclusions in [4]. The application of the code system for analysis of high burnup fuel characteristics is justified, keeping in mind that the ICE KWO project only gives information for burnups up to 31 GWd/tHM.

3 Influence of boron concentration treatment on trans-uranics build-up

After re-validation of the calculation tools, several interesting aspects of high burnup of discharged nuclear fuel from PWR's were investigated in reference [5]. In the following sections selected topics from these investigations are presented.

3.1 Applied calculation models, codes and libraries

The high burnup investigations are performed with the same procedures as applied to the validation work, described in sections 2.1,2.2. For the multi-group calculations the 69 group library based on ENDF/B-6.5 was applied. Input modifications were necessary to account for the intended increase of discharged fuel burnup and for improvements in PWR fuel assembly design. The main characteristics are summarized in table 3. Main changes compared to the KWO fuel are decrease of fuel-radius and canning thickness, and increase of U^{235} enrichment. The moderator-to-fuel volumetric ratio V_m/V_f and the soluble B^{10} concentration is varied over a wide range to analyse the spectral effects.

3.2 Selected results of the calculations

In reference [5] various aspects of high burnup in PWR's are investigated. A very interesting issue is the treatment during the burnup simulation of the reactivity control by variable soluble boron in the coolant. The boron isotope B^{10} is a strong neutron absorber with $1/v$ energy dependency. Especially in well thermalized reactor lattices, the boron has significant impact on the burnup dependant neutron energy spectrum in the system. In the benchmark investigations of section 2 the boron concentration is taken into account by stepwise variations with quite large time intervals. In reference [5] the size of the boron steps is systematically analysed. The capabilities of

the calculation tools [6] allowed to apply very small timesteps for the simulation of a quasi-linear decrease in time of the B^{10} values. With this option, the investigations in [5] evaluated a constant value approximation for the linear boron letdown with comparable characteristics for the isotopic buildup. In the following, a few examples of the results are presented. Figure 3 shows the typical behavior of calculated K_∞ values for variable and constant boron modelling during fuel burnup. The linearly decreasing boron influence between the "begin" and "end" concentrations is clearly visible. In view of the work in reference [1] the characteristics of Pu^{238} build-up had special attention. Figure 4 shows the build-up of Pu^{238} as a function of fuel burnup for three boron treatment cases. The upper curve is calculated with the constant value of 80 ppm B^{10} concentration. The lower, close together, curves are obtained with a quasi-continous B^{10} concentration decrease from 500/376 to zero ppm. The difference between the limit cases is considerable ($\approx$ 30% at 80 GWd/tHM). Figure 5 shows the build-up of the minor actinide isotope Np^{237}, having good fissile properties and being a precursor of Pu^{238}, as a function of fuel burnup for two constant B^{10} concentrations and one linear letdown. It was found that for most isotopes in this lattice a constant value of 80 ppm B^{10} gives a reasonable representation for continuous letdown. The difference between the calculated limit cases is for most isotopes around 5% at 80 GWd/tHM, for Np^{237} about 25%. However, conclusions about the absolute fit of calculations can hardly be made in this extrapolated burnup range without additional relevant experimental data. Finally, a typical result of lattice parameter variation on burnup characteristics is shown in figure 4, again for Pu^{238}. Here variable boron letdown is applied. The Pu^{238} build-up during burnup clearly shows a strong increase for better moderated lattices (larger V_m/V_s values).

3.3 Conclusions from high burnup investigations.

The results in section 3.2, representing typical findings from reference [5], show high prediction uncertainties at high burnups for important man-made isotopes in the nuclear fuel cycle. These results have been obtained with simulation tools, reasonably validated up to 30 GWd/tHM. It is evident that relevant experimental validation up to higher burnups (e.g. 80 GWd/tHM) will improve significantly the prediction quality.

4 Summary and recommendations

The accurate prediction of transuranic heavy material inventories in irradiated spent nuclear fuel is of high interest, especially in a closed nuclear fuel cycle. The validation of the simulation tools for these predictions only can be done on the basis of experimental results. Only few information related to experimental results from relevant integral or sample irradiations is available in open literature. In this sense, the results of the Isotope Correlation Experiment ICE in the PWR Obrigheim (KWO) are well documented in open literature. In a longterm experiment with irradiation over several reactor cycles the composition of irradiated PWR fuel up to 31 GWd/tHM is analysed in detail by various methods and in various institutions in Europe. This

experimental data is applied for successful re-validation of the current versions of our simulation codes and libraries. These validated tools were applied for detailed investigations for trans-uranics build-up up till 140 GWd/tHM. Significant influence of the treatment of the boron control in the PWR coolant is observed for standard and wider lattices. An extention of the experimental data base in open literature to higher burnups is very desirable.

5 References

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	Fuel		Canning		Moderator	
Material	UO_2 3.1% U^{235}		zirconium		borated H_2O	
Temperature [K]	1028		605		572	
Outer radius [cm]	0.465		0.535		0.8449	
Numberdensity	U^{235}	7.120E-04	Zr	4.325E-02	H	4.806E-02
[1/(cm barn)]	U^{238}	2.197E-02			O	2.403E-02
	O	4.538E-02			B^{10}	7.738E-06

Table 1: KWO ICE lattice specifications [4]

Time-step	B^{10}-Conc. $[10^{-6}]$	B^{10}-Conc. [ppm]	Time-step	B^{10}-Conc. $[10^{-6}]$	B^{10}-Conc. [ppm]
1	7,738E-06	322,1	15	4,686E-06	195,1
2	7,567E-06	315,0	16	4,249E-06	176,9
3	6,836E-06	284,5	17	3,414E-06	142,1
4	5,831E-06	242,7	18	2,891E-06	120,3
5	5,395E-06	224,6	19	2,651E-06	110,4
6	5,031E-06	209,4	20	2,338E-06	97,3
7	4,495E-06	187,1	21	1,711E-06	71,2
8	3,726E-06	155,1	22	1,404E-06	58,4
9	2,027E-06	84,4	23	6,705E-06	279,1
10	7,605E-07	31,7	24	5,733E-06	238,7
11	2,558E-07	10,6	25	3,978E-06	165,6
12	7,494E-06	312,0	26	3,000E-06	124,9
13	6,663E-06	277,3	27	2,248E-06	93,6
14	5,447E-06	226,7	28	6,976E-07	29,0

Table 2: Soluble B^{10} concentration during the ICE-KWO experiment [5].

Parameter	Value
Fuel Pin Radius	0.411 [cm]
Canning Thickness	0.064 [cm]
Vm/Vs	1.283 - 2.565
Parameter	Value
Moderator Density	0.74730 [g/cm^3]
Fuel Density	0.89683 [g/cm^3]
Temperature of Fuel	773 [K]
Temperature of Canning	605.8 [K]
Temperature of Moderator	583 [K]
Initial Enrichment U^{235}	5.0 [%]
Initial Enrichment Pu_{tot}	0.0 [%]
Initial B^{10} Concentrations in the Moderator	80 - 500 [ppm]

Table 3: Characteristics for high burnup boron investigations [5].

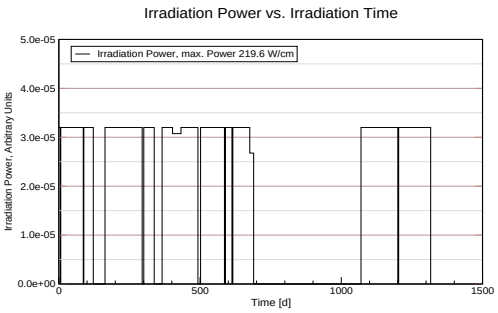

Figure 1: Irradiation power-rating history of the KWO ICE project [5]

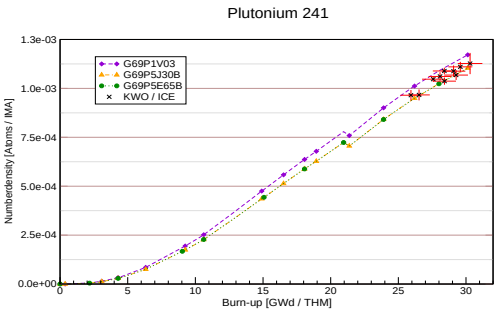

Figure 2: Comparison of number densities as a function of burnup for Pu^{241} [5]

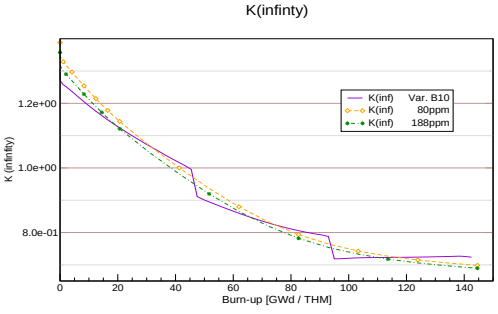

Figure 3: Influence of soluble boron treatment in PWR on k_∞ [5]

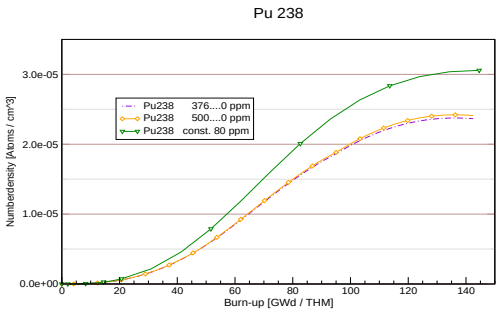

Figure 4: Impact of boron modeling on Pu^{238} build-up [5]

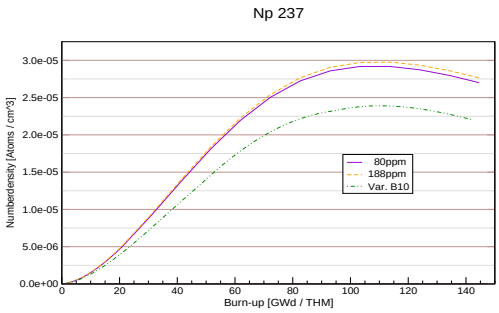

Figure 5: Influence of soluble boron concentrations on Np^{237} build-up in PWR [5]

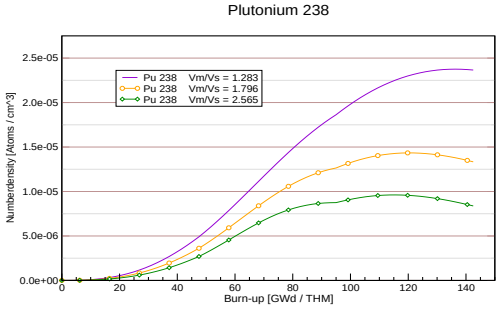

Figure 6: Influence of moderator-to-fuel ratio V_m/V_s on Pu^{238} build-up in PWR [5]