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IMPACT OF IRRADIATION HISTORY ON INVENTORY PREDICTION DATA FOR PWR FUEL

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ABSTRACT

Procedures developed to allow generation of radio nuclide inventory data depend on fluxes and cross sections calculated by assembly spectrum codes. Their accuracy depends on the assembly spectrum code capacity to generate correct prediction data; otherwise corrections have to be made to eliminate the residual calculation/experiment discrepancies. In this work we present the effect of taking into account the temperature and boron history on predicted isotopic compositions (uranium and plutonium) of a 40 GWd/t UO₂ fuel.

INTRODUCTION

The overestimation of the Pu isotopic inventory due to approximations in the standard PWR calculation scheme used to set up cross section libraries and fluxes for fuel cycle predictions, is a phenomenon that has been well-identified for some years. In order to offset this drawback, a posteriori adjustments and corrections are made and the problem is circumvented, thus ensuring good quality predictive values for the isotopic masses at the end of irradiation (very little dispersion or disagreement). These corrections, however frequently mask modeling simplifications and have no physical significance; they very often exceed the uncertainties of the basic data and, moreover, they require specific validation for each type of fuel (according to the enrichment and/or the initial composition). An exhaustive study was therefore conducted to identify the causes of the discrepancies and the impact of the simplifications introduced on the final result. Among the most serious simplifications is the fact that the history of the fuel temperature and the history of the soluble boron are not taken into account.

In a first stage, the standard calculation scheme is described and a comparison with experimental results is given so as to situate the magnitude of the discrepancies obtained; we then indicate the methodology used to determine the effect of taking into account, or not, the two above-mentioned operating parameters. Finally, the real impact of the detailed modelling of irradiation history on reactivity and material balance is estimated by the comparison of a standard calculation with a detailed calculation in which this history is modelled faithfully.

THE STANDARD PWR CALCULATION SCHEME; COMPARISON OF PREDICTIONS TO EXPERIMENTAL VALUES

For several years now, CEA in collaboration with EDF and FRAMATOME has conducted a programme of irradiated fuel analysis. The investigation is generally twofold: metallurgical (control and behaviour of the fuel under irradiation, cladding corrosion, etc...) and neutronic ^{(1),(2)} (qualification of French fuel evolution and neutronics codes, assembly scheme calculations and basic data). The results of these experimental programmes are used to qualify the estimates of the isotopic masses of radionuclides knowing that the accuracy of the evolution and calculation schemes depends very much on the quality of the cross-sections and the fluxes generated by the subassembly calculation codes. So as to situate the performances of the standard calculation scheme vis à vis the experiment it is compared to the analysis results of the BUGEY 3 experimental programme on uranium and plutonium actinide isotopes (fuel pin K11).

A. DESCRIPTION OF THE CALCULATION SCHEME

The standard calculation scheme uses the APOLLO 1 subassembly calculation code to generate cross sections and fluxes; APOLLO relies on a P_{ij} transport code that has been extensively validated and qualified ^{(3),(4),(5)}. The code provides the data libraries required for cycle calculation codes. At present constant parameters are used during irradiation for the constitution of cross-section and flux data. These parameters are: a constant concentration of the soluble boron in the moderator, constant fuel temperature, constant moderator temperature and density, constant cladding temperature, constant power released by the subassembly. These values are assumed to be constant from 0 GWj/t until the end of irradiation (approximately 40 GWj/t depending on the initial enrichment of the fuel). The values for the parameters that interest us (modelled in the present study) are: a boron concentration of 500 ppm¹ and a fuel temperature of 660.3°C. All the calculations were performed with the CEA '86 nuclear data library⁽⁶⁾.

B. COMPARISON TO EXPERIMENTAL VALUES

The results of the standard calculation were compared with the experimental values obtained from chemical analyses of an irradiated 900 MWe EDF fuel pin (3.10% enrichment, 40 GWj/t burnup); the following values were obtained:

Isotopic Ratio	$\frac{^{234}\text{U}}{^{238}\text{U}}$	$\frac{^{235}\text{U}}{^{238}\text{U}}$	$\frac{^{236}\text{U}}{^{238}\text{U}}$	$\frac{^{238}\text{Pu}}{^{238}\text{U}}$	$\frac{^{239}\text{Pu}}{^{238}\text{U}}$	$\frac{^{240}\text{Pu}}{^{238}\text{U}}$	$\frac{^{241}\text{Pu}}{^{238}\text{U}}$	$\frac{^{242}\text{Pu}}{^{238}\text{U}}$
Discrepancies (C-E)/C (%)	24.62 ²	0.00	-1.14	11.67	6.68	0.82	7.63	9.64

The values presented were readjusted for the same $\frac{^{235}\text{U}}{^{238}\text{U}}$ residual ratio and do not represent the maximal accuracy of the APOLLO 1 code which would require lengthy calculation times. These discrepancies are of the same order of magnitude as those observed in the past with cycle codes such as BUDLIA-FISPIN6 with the ZORITA experiment (pin number 234)⁽⁷⁾.

DETAILED MODELLING OF IRRADIATION HISTORY

¹ppm= parts per million

²This value depends on the initial concentration which, being relatively low, is difficult to determine and therefore presents significant standard deviations.

A. SIMULATION OF SOLUBLE BORON VARIATIONS

The reactivity loss due to fuel burn-up is controlled by means of soluble boron in the moderator. The differential reactivity is of the order of -10 pcm/ppm of natural boron; this value varies from -8.8 pcm/ppm to -10 pcm/ppm with a minimum of -8.4 pcm/ppm⁽⁸⁾ during irradiation due to the variation of the boron concentration which in reality is continuous is not possible with APOLLO 1 especially if one wants to self-shield the very resonant elements; for burnup calculations computing times would become prohibitive due to the need for repetitive calculations. However it is possible to model the time dependency of boron concentrations by successive concentration plateaus.

In Figure n°1, one can see the ideal course ("REALITY") of a concentration variation for an annual cycle⁽⁸⁾. A very rapid decrease from 1250 to 912 ppm can be observed which is essentially due to the saturation of certain fission products (150 MWd/t xenon equilibrium, etc...). Beyond this and up to 14 000 MWd/t there is in reality a continuous and practically linear decrease.

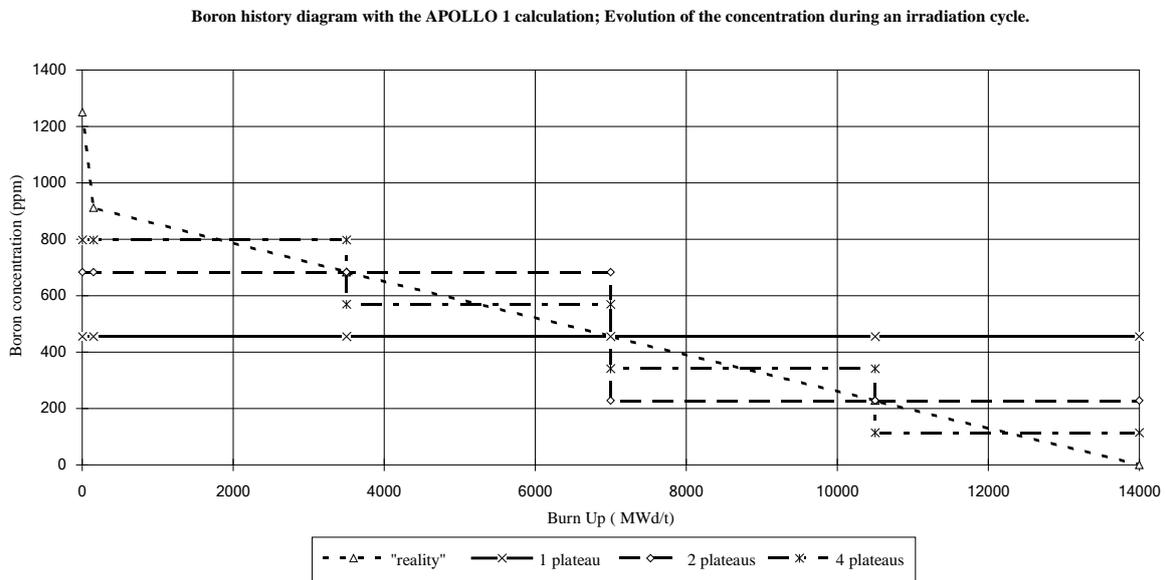


Figure n° 1

The "1 PLATEAU" straight line describes present modelling used in fuel cycle and/or qualification calculations: a constant mean concentration. The dotted lines ("2 PLATEAUS" and "4 PLATEAUS" captions) describe the hypotheses used for the present study. The exact values of the boron concentration (in ppm) for each model and for each of the irradiation cycles are given in the table below.

Burn-up MWd/t	1 PLATEAU	2 PLATEAUS	4 PLATEAUS
0-3500	456	684	798
3500-7000	456	684	570
7000-10500	456	228	342
10500-14000	456	228	114

DISCUSSION

A.1. EFFECT OF BORON CONCENTRATION VARIATIONS ON REACTIVITY³

Figure n° 3 shows the reactivities calculated for the various scenarii. It is to be noted that the reactivity jumps are proportional to the discontinuities in boron concentration (passage from one step to another, Figure 1) and that the reactivity value at the end of each cycle is higher than the "1 PLATEAU per cycle" value and of the order of magnitude of the discontinuity. By way of example, if we compare the k_{∞} of 4 PLATEAUS and 1 PLATEAU, we obtain a discrepancy of 2905 pcm for a difference of 342 ppm of boron; this can be explained by the value of the differential worth of the boron, approximately $-9 \text{ pcm}^4/\text{ppm}^{(6)}$.

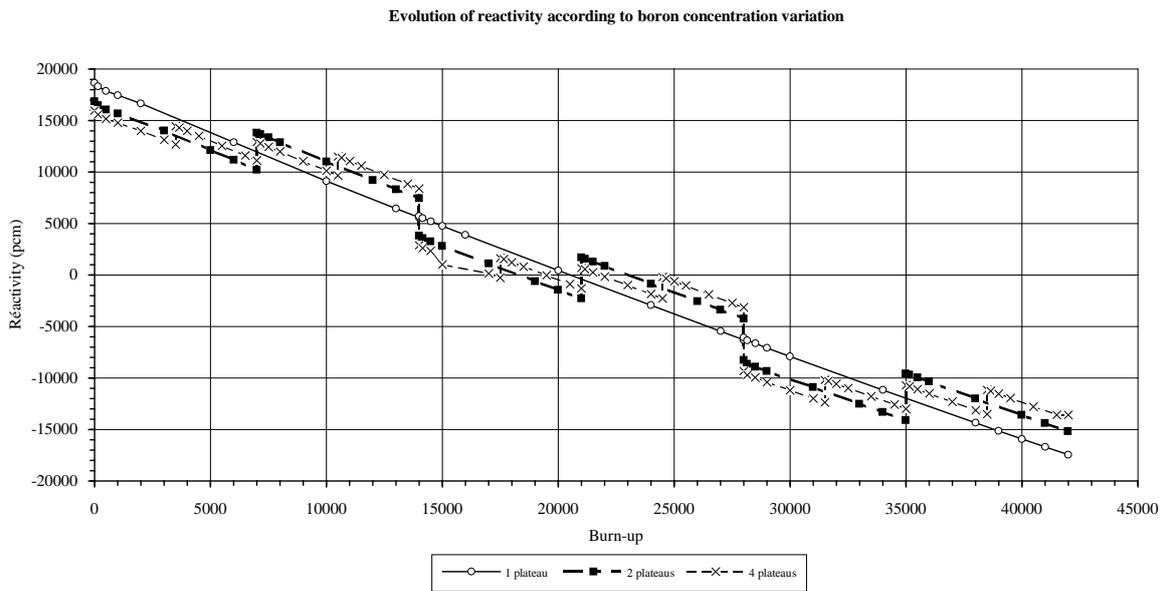


Figure n° 3

A.2. EFFECT OF BORON CONCENTRATION VARIATIONS ON ISOTOPIC RATIOS

The table below gives the discrepancies (in % and at 40 GWj/tU) in the isotopic ratios of the main U and Pu actinides.

Isotopic Ratio	$\frac{^{234}\text{U}}{^{238}\text{U}}$	$\frac{^{235}\text{U}}{^{238}\text{U}}$	$\frac{^{236}\text{U}}{^{238}\text{U}}$	$\frac{^{238}\text{Pu}}{^{238}\text{U}}$	$\frac{^{239}\text{Pu}}{^{238}\text{U}}$	$\frac{^{240}\text{Pu}}{^{238}\text{U}}$	$\frac{^{241}\text{Pu}}{^{238}\text{U}}$	$\frac{^{242}\text{Pu}}{^{238}\text{U}}$
Discrepancies (%) 2 paliers /1 palier	0.03	0.10	0.10	-0.37	-0.66	0.38	-0.73	0.28
Discrepancies (%) 4 paliers /1 palier	0.06	0.17	0.14	-0.50	-0.80	0.52	-0.90	0.35

It can be observed that in both the 2 and 4 plateau cases the effects are negligible for the uranium isotopes. For the plutonium isotopes, the effects are of unequal amplitude and have positive and negative

³The comparisons between two calculations are made after resetting on the same burnup rate.

⁴1\$=650 pcm for UO₂ fuel

signs according to the modelling and to the isotope. If these differences are slight for even isotopes, it is not the case for the fissile ^{239}Pu and ^{241}Pu isotopes. By way of example, in Figure 5 we have plotted the variation of the discrepancies between the two plateau/one plateau per cycle and four plateau/one plateau per cycle simulations for the ratio $\frac{^{239}\text{Pu}}{^{238}\text{U}}$; a notable effect can be observed here: a mainly positive discrepancy at the beginning of irradiation (+2%). Afterwards, the discrepancies remain in the +0.5%/-1% range. However, the effect is systematically negative at the end of each cycle and this discrepancy increases slightly (in absolute values) when the history of the boron is improved. The same effect can be observed and amplified on the abundance of ^{241}Pu . A similar study⁽¹¹⁾ has shown effects of the same order on the average concentrations but of opposite signs.

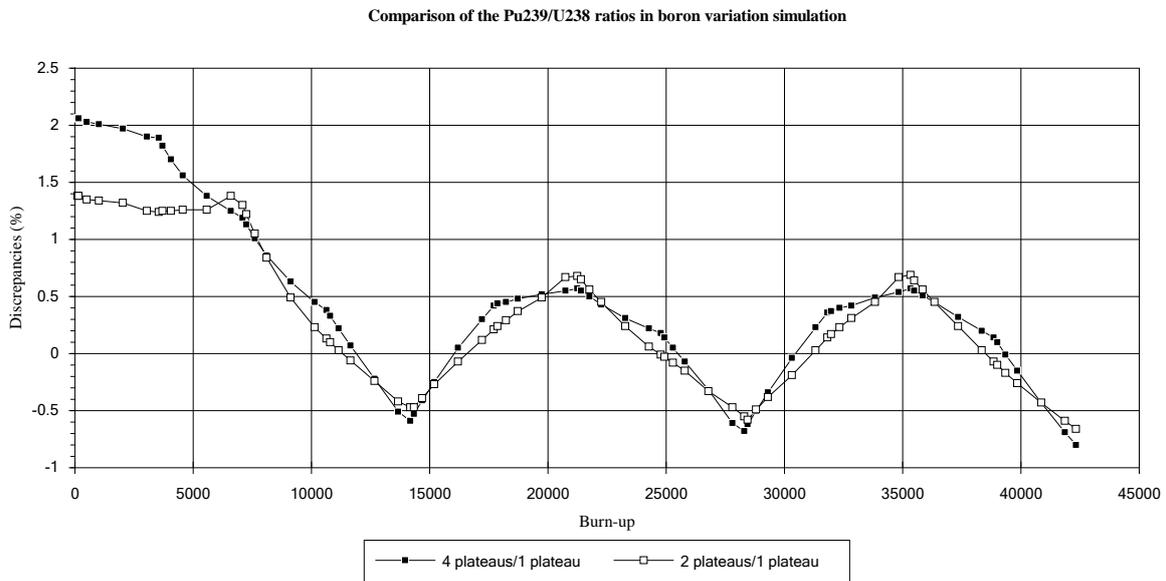


Figure n° 5

B. SIMULATION OF FUEL TEMPERATURE VARIATION

Fuel temperature has a significant effect on reaction rates and thus isotopic concentrations, due to the Doppler broadening of cross section resonances and due to variations of the mean equilibrium energy of the thermalized neutron spectrum. At low energy and far from resonances, the cross-sections are not in fact altered by the Doppler effect. On the other hand near resonances the Doppler effect is manifest through the factor $\Delta = \sqrt{\frac{4 \cdot k \cdot T \cdot E}{A}}$ (Doppler width). The area under the resonance curve being preserved

(independent of the temperature and equal to the value in the absence of the Doppler effect $I = \frac{\pi}{2} \cdot \sigma_0 \cdot \Gamma$),

a widening or a narrowing of the latter is observed according to whether there is an increase or a decrease of the temperature. A drop in the fuel temperature can be attributed to a certain number of transformations that all fuel rods undergo during irradiation in relation to their initial state, i.e.: closing of the fuel-clad gap by expansion (slight) and swelling (essentially due to intergranular F.P.) of the pellet and creep of the clad, cracking (the main cause being quenching during fast power transients such as an emergency

shutdown) with emission of fission products, modification of flux depression during irradiation and, finally, the evolution of the thermal conductivity λ .

In this complex set of physical phenomena, only the effect of closing the fuel-clad gap has been considered with the subsequent thermal evolution. This has been done using the heat exchange coefficient values h^5 supplied by EDF/SEPTEN and used industrially. The set of values cover a wide range of burnups and linear power density. It was thus possible to determine the temperature in the rods and its evolution with satisfactory accuracy despite needed simplifications: constant temperature gradient during irradiation, flat flux (no depression), constant thermal conductivity. Figure N°2 shows the effective temperature of the fuel which preserves the ^{238}U reaction rate calculated with Rowlands' (9),(10) formula during fuel burnup and the temperature currently used for project and/or simplified studies (EDF-CEA,etc...). It can thus be seen that the temperature deviation at the end of irradiation reaches 100°C. In the same figure the representation (modelling) used for our temperature history calculations (succession of plateaus) has also been plotted.

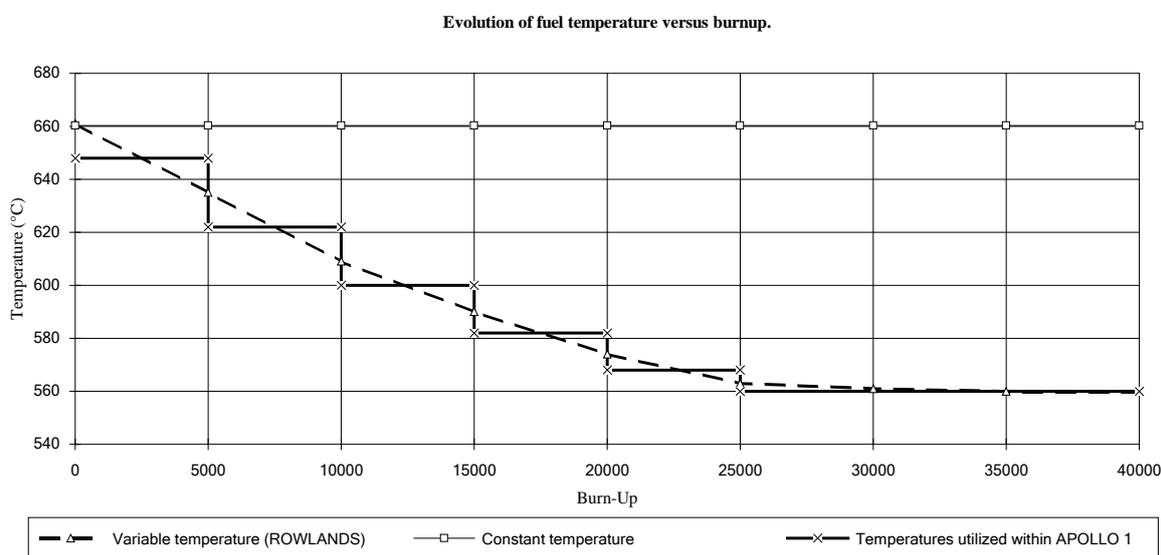


Figure n° 2

DISCUSSION

B.1. EFFECT OF TEMPERATURE VARIATION ON REACTIVITY

Figure N° 4 shows the discrepancies in pcm between the variable temperature and the constant temperature cases. The reactivity jumps due to the change of fuel temperature can be observed. These effects are slight and remain within the [-150, +150] pcm range.

⁵ $\phi = h \Delta T$ with ϕ in $\text{W cm}^{-2} \text{ } ^\circ\text{C}^{-1}$; ΔT temperature discrepancy between two media.

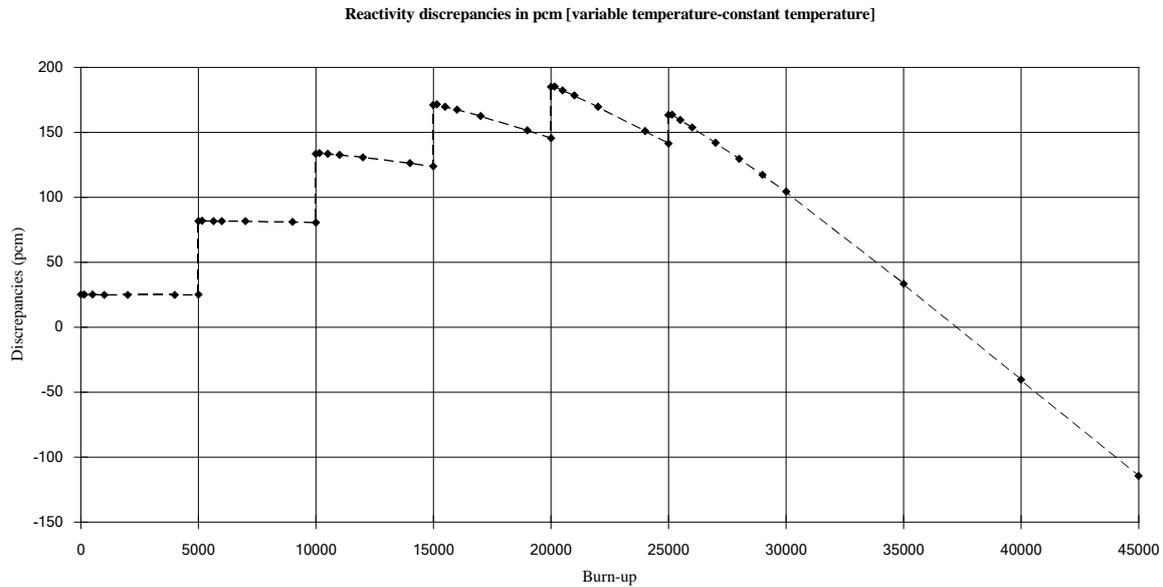


Figure n° 4

B.2. EFFECT OF TEMPERATURE VARIATION ON ISOTOPIC RATIOS

The table below gives the discrepancies (at 40 GWj/tU) related to the fuel temperature history presented above.

Isotopic Ratio	$\frac{^{234}\text{U}}{^{238}\text{U}}$	$\frac{^{235}\text{U}}{^{238}\text{U}}$	$\frac{^{236}\text{U}}{^{238}\text{U}}$	$\frac{^{238}\text{Pu}}{^{238}\text{U}}$	$\frac{^{239}\text{Pu}}{^{238}\text{U}}$	$\frac{^{240}\text{Pu}}{^{238}\text{U}}$	$\frac{^{241}\text{Pu}}{^{238}\text{U}}$	$\frac{^{242}\text{Pu}}{^{238}\text{U}}$
Discrepancies (%) Temperature variable/constante	-0.24	-1.21	0.11	0.31	-1.65	-0.18	-1.59	0.00

One can see that, as in the boron study, the discrepancies for the uranium isotopes remain low except for the $\frac{^{235}\text{U}}{^{238}\text{U}}$ ratio which presents a discrepancy that, a priori, is considerable but which concerns only small quantities (residual ratio at the end of irradiation of about 0.7%). The plutonium isotopes show a clear drop in the $\frac{^{239}\text{Pu}}{^{238}\text{U}}$ and $\frac{^{241}\text{Pu}}{^{238}\text{U}}$ quantities and very low effects (or none at all) for pair isotopes. As regards the $\frac{^{239}\text{Pu}}{^{238}\text{U}}$ ratio, a steady decreasing trend can be observed (Figure N°6) which reaches -1.60% at 40

GWj/t with an inflexion point expressing a slowing down of ^{239}Pu disappearance. Figure N°7 shows the evolution of the discrepancies for the ^{238}U capture and ^{239}Pu absorption cross-sections (formation and disappearance of ^{239}Pu). One can note that at the beginning of irradiation (up to 10 GWj/t) there is a considerable decrease in the ^{238}U capture cross-sections whereas the ^{239}Pu absorption cross-section varies very little despite the drop in temperature. Beyond 10 GWj/t the ^{238}U capture cross-section continues to decrease slightly and the discrepancy reaches a limit value (-0.7%). Similarly, the ^{239}Pu absorption cross-

section continues to increase so that the sum of effects corroborates the disappearance law observed in Figure N°6.

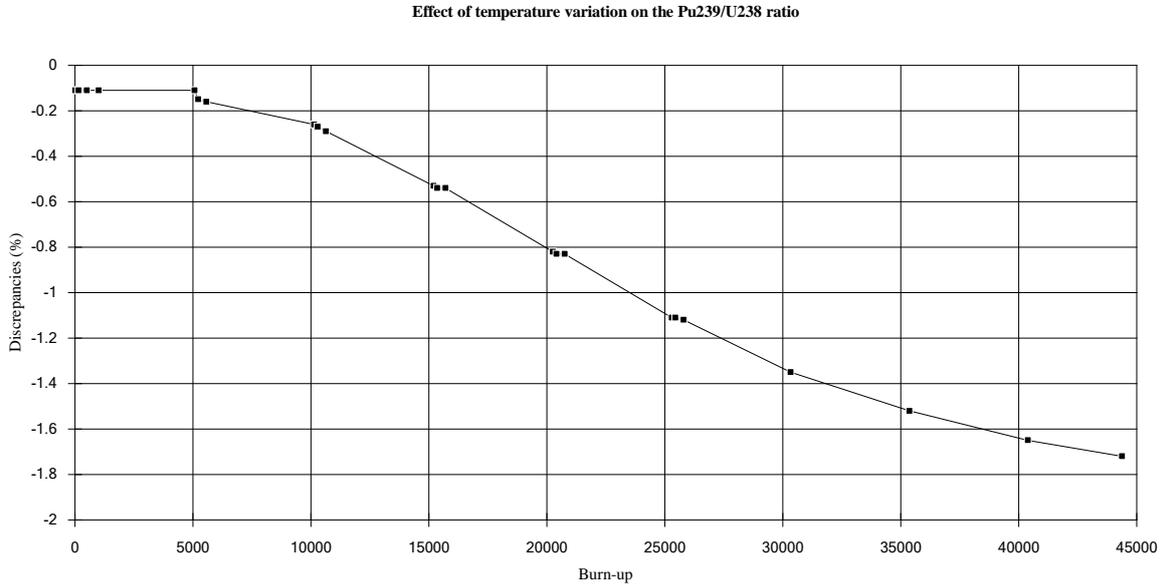


Figure n° 6

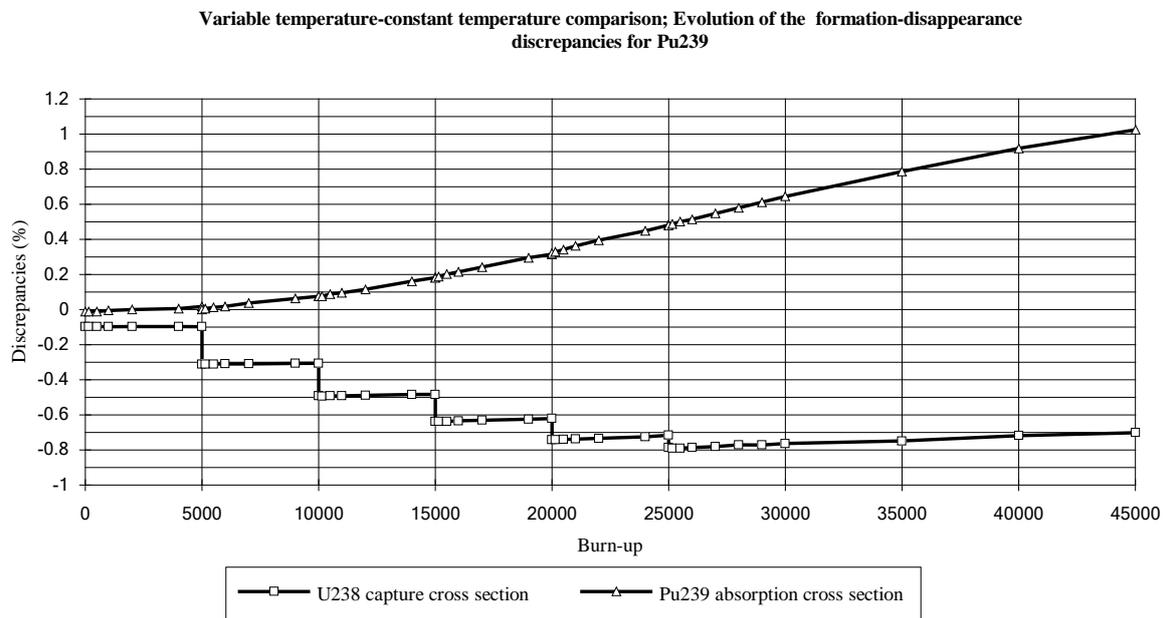


Figure n° 7

CONCLUSION

The calculations performed with the APOLLO 1 code to generate cross-section and flux libraries used by fuel cycle calculation codes lead to an overestimation of the plutonium material balance. To offset this drawback, conventionally a priori cross-section adjustments, or corrections of the estimations calculated a posteriori are made. In order to minimize this effect, what we propose here is to take into account -in a simplified way- the evolutions of the fuel temperature and the soluble boron concentration in the moderator. For the main actinides such as ^{239}Pu , these two effects result in reducing the discrepancies by -1% for variable boron and -1.6% for the fuel temperature effect. We can therefore consider that these two combined effects produce an improvement in the calculation of the material balance at the end of irradiation with positive consequences on the other elements taken into account in the evolution codes (minor actinides, fission and activation products). Moreover, the consideration of other parameters of irradiation history, and the ensuing better representativeness of reality, will significantly improve the calculation of the data required for fuel evolution codes and will remove modelling expedients. Recent work⁽¹²⁾ performed at CEA/SPRC is fully consistent with this perspective: more realistic consideration of irradiation conditions will result in a significant resorption of calculation-experiment discrepancies.

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