# International comparison of a depletion calculation benchmark devoted to fuel cycle issues
# Results from the Phase 1 dedicated to PWR-UOx fuels

B. Roque*[1], R. Gregg[2], R. Kilger[3], F. Laugier[4], P. Marimbeau[1], A. Ranta-Aho[5], C. Riffard[1], K. Suyama[6], J.F. Thro[7], M. Yudkevich[8], K. Hesketh[2], E. Sartori[9]

[1] *French Atomic Energy Commission DEN/DER/SPRC, CE Cadarache, F-13108 St Paul-Lez-Durance Cedex, France*
[2] *Nexia Solutions Ltd, B709 Springfields, Preston, UK PR4 OXJ,*
[3] *Gesellschaft für Anlagen- und Reaktorsicherheit (GRS) mbH, Abt. Brennstoffkreislauf, Forschungsgelände, 85748 Garching b. München*
[4] *EDF, R&D, 1 av. Général de Gaulle, F-92141 Clamart Cedex, France*
[5] *VTT Processes, Nuclear Energy, POB 1604,FI-02044 VTT, Finland*
[6] *Japan Atomic Energy Research Institute, Tokai-Mura, Ibaraki-ken, Japan*
[7] *AREVA-NC, 2 Rue P. Dautier, BP 4, 78141 Velizy, Cedex*
[8] *Russian Research Center "Kurchatov Institute", Kurchatov sq. 1, 123182, Moscow, Russia*
[9] *Organization for Economic Cooperation and Development/NEA, Paris, France*

## Abstract

This paper presents the results from the first phase of an international depletion calculations comparison devoted to PWR-UOx fuel cycle issues. This 'benchmark' has been defined within the NEA/OECD Working Party on Scientific Issues in Reactors Systems (WPRS). The aim is to investigate a large range of isotopes, physics quantities and fuel types applied to fuel and back-end cycle configurations.

The results analyses have shown that there is a good agreement between participants for the mass calculation of many isotopes. However, it is interesting to observe that better agreement is obtained for isotopes which benefit from experimental validation. In this benchmark, the poorest agreement is obtained in calculating activation products originating from fuel impurities. Some discrepancies on neutron emission rates were also observed, mainly due to the discrepancies on masses calculations. Good agreement was obtained for the total decay heat calculation.

***KEYWORDS : International Benchmark, Depletion calculation, Fuel cycle, PWR-UOx fuel***

## 1. Introduction

Although there are many reactor system benchmarks in the literature, they mostly concentrate on the reactor system in isolation and only a few consider the fuel cycle. However, there is currently increased emphasis on the performance of reactor systems linked with their associated fuel cycle (Generation IV for example).

Published international benchmark studies related to burnup depletion calculations were restricted to specific fuel cycle questions:

* Benedicte Roque, Tel. 33-4-42-25-36-48, Fax. 33-4-42-25-79-79, E-mail: benedicte.roque@cea.fr

• Burnup credit benchmark (working party on nuclear criticality safety (WPNCS) [1]: the nuclide density calculations focused mainly on the 15 most poisoning fission products and for a short cooling time (5 years)
• Benchmark on decay heat calculation: this benchmark focused on decay heat calculation due to 235 uranium fissions [2].

The NEA/OECD Working Party on Scientific Issues in Reactors Systems (WPRS) has defined a new benchmark investigating a broader range of isotopes, physics quantities and fuel types [3]. The objective is to compare existing burnup depletion calculations obtained with various codes and data libraries, applied to fuel and back-end cycle configurations: transport, reprocessing, interim storage and waste repository. The benchmark concentrates on nuclide densities for the most important nuclides in the fuel cycle: actinides, fission products and activation products and also to calculate the associated fuel cycle quantities - masses, neutron emission rate and decay heat. Additional but optional calculations were also specified; they relate to sensitivity calculations linked to the assumptions made on irradiation parameters: the width of the half water gaps, the boron content, the fuel and moderator-coolant temperature and the irradiation history.

This paper presents the results from the first phase of the benchmark devoted to UOx fuel. The aim of this phase is to constitute a reference case on a standard fuel. The benchmark used experimental information from the Japanese Post Irradiation Experiment in the TAKAHAMA-3 PWR [4], [5]; thus it is possible to compare the calculational and experimental results for some major actinides and fission products and verify for well-known isotopes how accurate the predictions are. For other isotopes, a code-to-code comparison is made.

## 2. Description of the benchmark

The benchmark consists of two calculations: a simple cell calculation and/or an assembly calculation where the analysed sample is explicitly described. For the assembly calculation, the studied sample originates from a $UO_2$ spent fuel pin located in the peripheral row of the assembly. This assembly is loaded with 248 $UO_2$ fuel pins, 4.1%wt U235 enriched, 16 $UO_2$-$Gd_2O_3$ pins and 25 water holes.

All the geometry data for the pincell and the assembly calculations, the fuel composition data, the clad and moderator composition data are explicitly given in the benchmark specification. Furthermore, the activation products masses from fuel impurities are compared; values for the initial impurities content are also specified.

The calculated quantities for the benchmark are as follows:

**Masses**

*Actinides (ACT)*:
$^{232}$U, $^{233}$U, $^{234}$U, $^{235}$U, $^{236}$U, $^{238}$U, $^{236}$Np, $^{237}$Np, $^{236}$Pu, $^{238}$Pu, $^{239}$Pu, $^{240}$Pu, $^{241}$Pu, $^{242}$Pu, $^{243}$Pu, $^{244}$Pu, $^{241}$Am, $^{242m}$Am, $^{243}$Am, $^{242}$Cm, $^{243}$Cm, $^{244}$Cm, $^{245}$Cm, $^{246}$Cm, $^{247}$Cm, $^{248}$Cm, $^{226}$Ra, $^{228}$Ra, $^{227}$Ac, $^{229}$Th, $^{230}$Th, $^{232}$Th, $^{252}$Cf

*Fission Products (FP)*:
$^{79}$Se, $^{85}$Kr, $^{85}$Rb, $^{87}$Rb, $^{88}$Sr, $^{90}$Sr, $^{93m}$Nb, $^{95}$Mo, $^{97}$Mo, $^{99}$Tc, $^{101}$Ru, $^{106}$Ru, $^{103}$Rh, $^{107}$Pd, $^{108m}$Ag, $^{109}$Ag, $^{110m}$Ag, $^{127}$I, $^{129}$I, $^{130}$Xe, $^{131}$Xe, $^{132}$Xe, $^{134}$Xe, $^{136}$Xe, $^{133}$Cs, $^{134}$Cs, $^{135}$Cs, $^{137}$Cs, $^{136}$Ba, $^{138}$Ba, $^{139}$La, $^{140}$Ce, $^{144}$Ce, $^{142}$Nd, $^{143}$Nd, $^{144}$Nd, $^{145}$Nd, $^{146}$Nd, $^{148}$Nd, $^{150}$Nd, $^{147}$Pm, $^{146}$Sm, $^{147}$Sm, $^{148}$Sm, $^{149}$Sm, $^{150}$Sm, $^{151}$Sm, $^{152}$Sm, $^{154}$Sm, $^{153}$Eu, $^{154}$Eu, $^{155}$Eu, $^{154}$Gd, $^{155}$Gd, $^{156}$Gd, $^{166m}$Ho

*Activation Products (AP)*
$^{36}$Cl, $^{41}$Ca, $^{53}$Mn, $^{54}$Mn, $^{55}$Fe, $^{60}$Fe, $^{60}$Co, $^{59}$Ni, $^{63}$Ni, $^{93}$Mo
Some activation products are produced both by fissions and activation reactions and these two contributions are evaluated separately for $^{3}$H, $^{10}$Be, $^{14}$C, $^{93}$Zr, $^{94}$Nb, $^{119m}$Sn, $^{121m}$Sn, $^{126}$Sn, $^{125}$Sb.
**Neutron emission**: (alpha,n) emission, spontaneous fission and total emission
**Decay heat**: alpha, beta and gamma decay heat and total decay heat.
All these results are compared at discharge (zero cooling), 5, 50, 100 and 300 years cooling.

## 3. Participants and codes used

In total, seven contributions were submitted to this benchmark exercise, from six different organisations and six countries. Six calculations were submitted for both the cell geometry representation and the assembly calculation. Both deterministic and Monte-Carlo methods are represented in the contributed solutions. Table 1 summarises the participants and methods used in their analyses. The organisation label given in this Table represents the abbreviation used to identify each set of calculations throughout the remainder of the paper.

**Table 1**: List of participants and codes used

| Organisation Label | Organisation | Codes used | Nuclear data library used | Calculations made |
|---|---|---|---|---|
| CEA-D | Commissariat à l'Energie Atomique, France | DARWIN (APOLLO2-PEPIN2) | JEF2.2 (ACT,FP) EAF01 (AP) | Cell Assembly |
| CEA-C | Commissariat à l'Energie Atomique, France | CESAR | JEF2.2 (ACT,FP) EAF99 (AP) | Cell Assembly |
| GRS | Gesellschaft für Anlagen-und-Reaktorsicherheit, Germany | OREST-V03t01 (HAMMER + ORIGEN) KENOREST-V03t01 (KENO + OREST) | JEF2.2 | Cell Assembly |
| JAEA | Japan Atomic Energy Agency Japan | ORIGEN2.2 | JENDL3.2 | Cell |
| JAEA | Japan Atomic Energy Agency Japan | ORIGEN2.2 | JENDL3.3 | Cell |
| NEXIA | Nexia Solutions United Kingdom | CASMO-4 FISPIN | JEF2.2 | Assembly |
| RRC KI | Russian Research Center Kurchatov Institue Russian federation | MCU-REA /BURNUP | DLC/MCUDAT-2.2 | Cell Assembly |
| VTT | VTT Processes, Nuclear Energy Finland | MONTEBURNS 1.0 (MCNP+ORIGEN-2) | JENDL3.2 | Cell Assembly |

## 4 Method of results analysis

For each calculation, cell or assembly calculation, each isotope and each cooling time an average value was calculated; the associated Standard Deviation (SD) and the Relative Standard Deviation (RSD) of the **calculated** concentrations were also determined. The Relative Standard Deviation indicates the degree of consistency between the results provided by participants: a small RSD for a given isotope indicates consistency between the various codes and data used when a large RSD indicates a poor agreement.

For this benchmark, it was assumed that a good agreement is obtained when the RSD is below 10%. One can notice that a low RSD doesn't mean that all participants calculate the correct value but just that they calculate the same value.

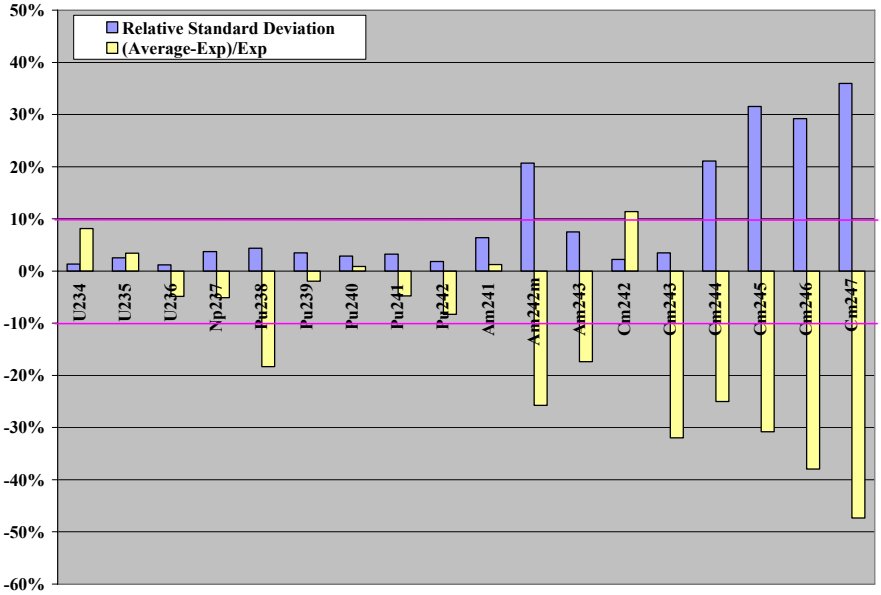
In the case of comparison with the experimental measurements, the reference used is the experimental value. To be rigorous enough in comparison between experimental results and calculations, we have to use the actual configuration of the sample pin during irradiation, i.e. the assembly calculation proposed in the benchmark; however the benchmark results analysis has shown that cell and assembly calculations give nearly the same values; consequently for the comparison with experimental values, the results presented below are including the JAEA 'cell' isotopic concentrations in the average and RSD calculations.

## 5. Results and discussion

### 5.1 Results and comparison of calculated isotopic concentrations with experimental values for some actinides and fission products

The RSD and the comparison between the average calculated value and the experimental values are plotted on Figure 1 for the actinides and on Figure 2 for the fission products. These comparisons are given at discharge (no cooling time).

**Figure 1:** Results for actinides; comparison of calculated average values to measured values



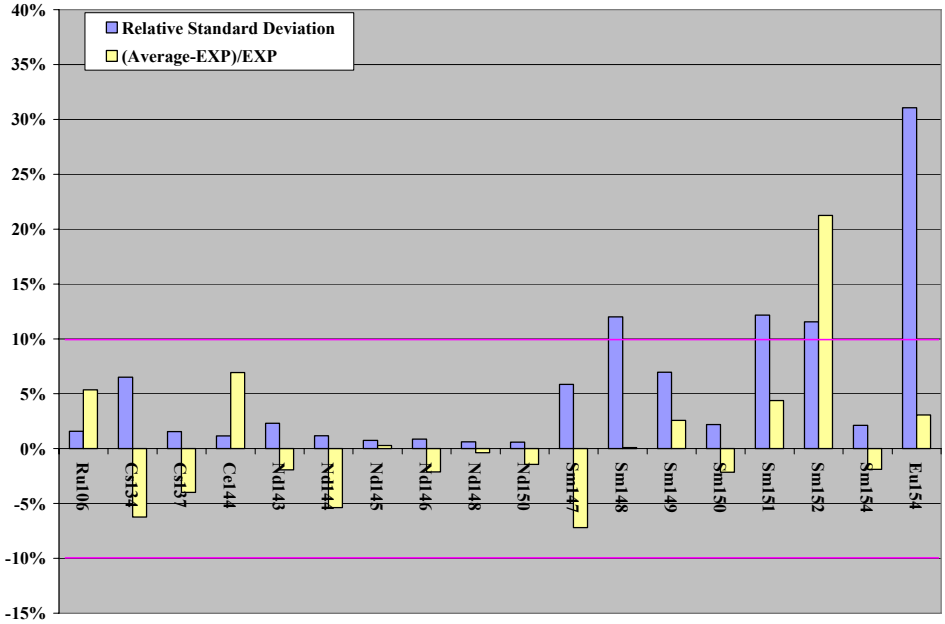

The Figure 1 shows that there is a good agreement between experimental values and calculations on one hand, and between all participants on the other hand, for the following isotopes: $^{234}$U, $^{235}$U, $^{236}$U, $^{237}$Np, $^{239}$Pu, $^{240}$Pu, $^{241}$Pu, $^{242}$Pu, $^{241}$Am and $^{242}$Cm. One can notice a slight underestimation by all participants for $^{236}$U, $^{241}$Pu and $^{242}$Pu.

The RSD and the (Average-Exp/Exp) values indicate that there is a strong underestimation by all participants for $^{238}$Pu, $^{243}$Am and $^{243}$Cm, by about -20%.

The large RSD observed on $^{242m}$Am, $^{244}$Cm, $^{245}$Cm, $^{246}$Cm and $^{247}$Cm are linked to the VTT values. However, these VTT calculated values are at the same time in closest agreement with the measured concentrations

The results presented on Figure 2 show very good agreements between calculation and experimental values for $^{106}$Ru, $^{137}$Cs, $^{144}$Ce, $^{143}$Nd, $^{144}$Nd, $^{145}$Nd, $^{146}$Nd, $^{148}$Nd, $^{150}$Nd, $^{147}$Sm, $^{149}$Sm, $^{150}$Sm, $^{154}$Sm. Higher discrepancies are observed for $^{154}$Eu, $^{134}$Cs, $^{148}$Sm, $^{151}$Sm and $^{152}$Sm.

**Figure 2:** Results for fission products; comparison of calculated average values to measured values




Possible reasons for the discrepancies obtained on these isotopes have been identified and the analyses of corresponding nuclear data are in progress. For $^{154}$Eu, the large RSD is due to CEA and NEXIA calculations using the JEF2.2 evaluation including an inaccurate $^{154}$Eu (n,γ) cross-section.

### 5.2 Results of code to code comparison for isotopic concentrations

The comparison between the computed results for the actinides, the fission products and the activation products requested in the benchmark are presented in the next paragraphs. For these isotopes there is no experimental measurement with which one can compare the calculated results.

*5.2.1 Actinides results*

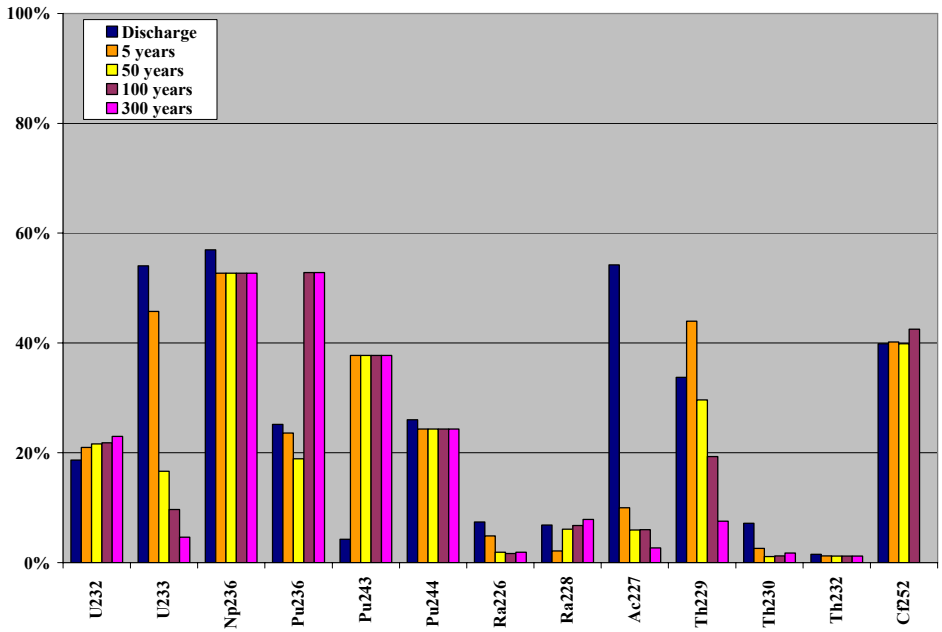
Figure 3 shows that there is very good agreement for $^{226}$Ra, $^{228}$Ra, $^{230}$Th and $^{232}$Th. Large discrepancies can be noticed for $^{232}$U, $^{233}$U, $^{236}$Np, $^{236}$Pu, $^{243}$Pu, $^{244}$Pu, $^{227}$Ac, $^{229}$Th, $^{252}$Cf.

The three isotopes $^{232}$U, $^{236}$Np and $^{236}$Pu are connected via their depletion chain. Studies have shown that the deviation observed on these isotopes is mainly coming from difference in $^{236}$Np (n,γ) cross-section values. The $^{237}$Np (n,2n) branching ratios values are also responsible for a part of the deviation.

The difference between participants for $^{233}$U calculation is due to the difference in $^{234}$U(n,2n) and $^{235}$U (n,3n) cross-sections. The RSD is decreasing with cooling due to the feeding of $^{233}$U by $^{237}$Np decay.

For $^{243}$Pu, there is quite a good agreement at discharge. After that, the RSD is increasing due to the feeding of $^{243}$Pu by $^{247}$Cm. The differences obtained on $^{244}$Pu are linked to $^{243}$Pu(n,γ) or $^{244}$Pu(n,γ) cross-sections values.

**Figure 3:** Results of Relative Standard Deviation versus cooling time for actinides




The large RSD for $^{227}$Ac at discharge is caused by the difference in $^{227}$Ac(n,γ) cross-section used in the codes libraries.

All the participants have obtained a higher value for the mass calculation of $^{229}$Th than the CEA one. This is due to the lack of $^{228}$Th (n,γ) cross-section in the JEF2.2 library.

A large disagreement among participants' results can be notice for $^{252}$Cf. The discrepancy on $^{248}$Cm can explain a part of this discrepancy because the $^{252}$Cf is always produced via the $^{248}$Cm. However, the cross-sections used for $^{249}$Bk, $^{250}$Cf and $^{251}$Cf may also explain some part. The spread of the results can also be the reflect of a small discrepancy on the flux, increasing as we go on in the depletion chain.

*5.2.2 Fission Products results*

Over the 56 calculated fission products, only nine have a RSD above 10%. It concerns silver isotopes, $^{108m}$Ag, $^{109}$Ag, $^{110m}$Ag with RSD respectively of 40%, 15% and 18%, $^{130}$Xe with a 20% relative standard deviation, $^{127}$I with a 12% RSD, $^{166m}$Ho with a 80% RSD. For the studied Europium and Gadolinium isotopes, discrepancies between participants have been only observed for $^{154}$Eu and its daughter $^{154}$Gd.

Comparisons between participants for fission yields, decay constant, branching ratios and capture cross-sections have to be performed in order to explain these large RSD, particularly for silver isotopes.

*5.2.3 Activation Products results*

The code-to-code comparisons for activation products, originating mainly from fuel initial impurities, are presented on Figures 4 and 5 for respectively the assembly and cell calculations. It's interesting here to compare these two calculations because this enable us to point out the origins of discrepancies coming from one participant. The difference (in term of participant) is the absence of NEXIA results in the cell calculation and the absence of JAEA results in the assembly calculation.

**Figure 4**: RSD results for Activation Products in assembly calculations

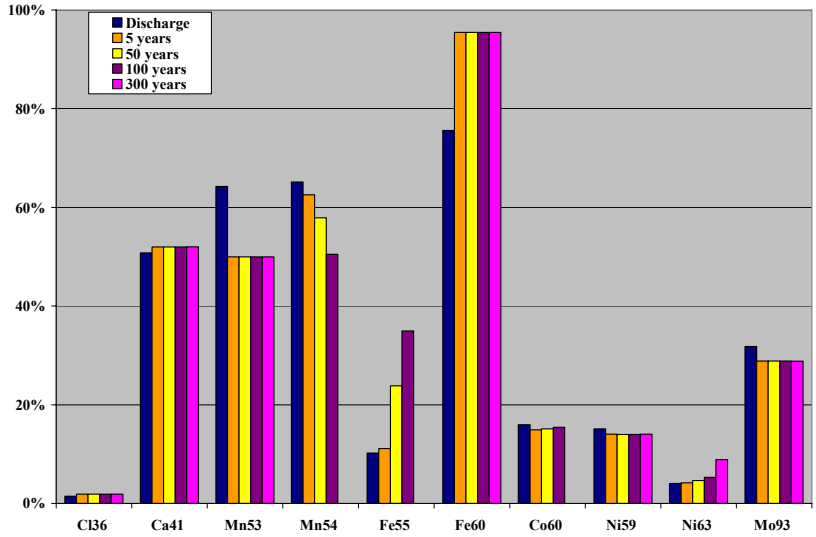
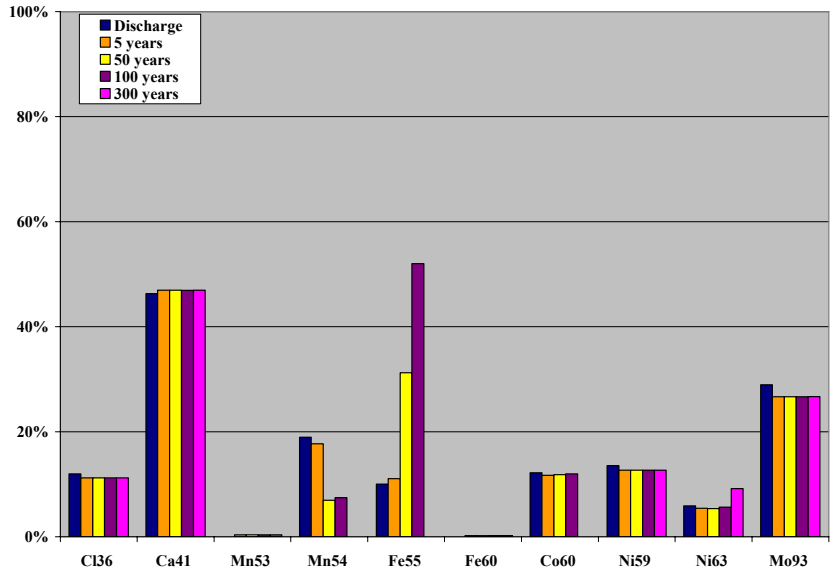



**Figure 5:** RSD results for Activation Products in cell calculations




A good agreement between participants' results is only obtained for $^{63}$Ni.

The RSD for $^{36}$Cl is correct from assembly contributions but a bit higher in cell calculations due to the JAEA value with the JENDL3.3 library.

The same trends are obtained in cell and assembly calculations for $^{41}$Ca; the large RSD is due to KI and GRS calculations giving nearly the same values but higher by a factor 2.5 from other participants' values.

Only the CEA has calculated $^{53}$Mn and $^{60}$Fe masses in cell calculation; in the assembly representation, the NEXIA values are also available but as it can be seen on Fig.4 and Fig.5 the CEA and NEXIA calculated masses are very different.

For $^{54}$Mn, the RSD from cell contributions is of 20% while the RSD from assembly contributions is higher (60%) due to the KI value which is very different from the cell value.

For $^{55}$Fe a trend appears with cooling, showing that the decay constant of $^{55}$Fe have to be compared.

For $^{60}$Co, there is a relatively good agreement between computed results, with the exception of the VTT value which is higher. However the RSD value, on the order of 15%, is not very large.

The RSD relative to $^{59}$Ni are almost the same in the cell and assembly representations. There is a good agreement between computed results for CEA, JAERI, NEXIA and GRS whereas KI values are higher and VTT values lower than the average.

The deviation found on $^{93}$Mo is due to the VTT predicted value which is low relative to the other participants' values.

More investigations are needed in order to explain the quite large differences observed on these activation products.

*5.2.3 'Activation-Fission' Products results*

Some activation products are produced both by fissions and activation reactions and these two contributions were evaluated separately in the benchmark. This is the case for $^{3}$H, $^{10}$Be, $^{14}$C, $^{93}$Zr, $^{94}$Nb, $^{119m}$Sn, $^{121m}$Sn, $^{126}$Sn and $^{125}$Sb. The comparisons for assembly and cell representations are presented on Figures 6 and 7.

A good agreement is obtained for $^{3}$H and $^{93}$Zr both in the cell and assembly calculations. One can notice that the CEA/CESAR results using the EAF99 evaluation has been removed from the average calculated value due to a wrong (n,t) reaction on $^{124}$Sn.

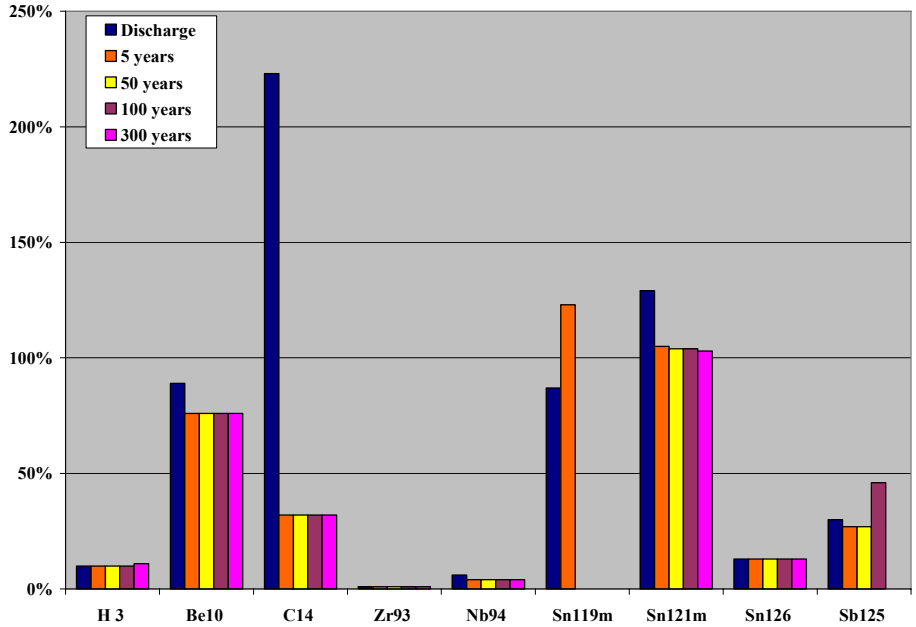
The relative standard deviation is very large for $^{10}$Be. There is a good agreement between CEA, NEXIA and KI, however the other values are lower. In order to explain this disagreement the ternary fission yields and (n,p) reaction on $^{10}$B need to be compared.

For $^{125}$Sb and $^{126}$Sn the CEA, NEXIA and VTT results are higher than all the other. For the $^{121m}$Sn, the CEA and BNFL results are again very high compared to the other ones. The JEF2 fission yields seem to be at the origin of the discrepancies.

There is two groups of calculated mass for $^{14}$C: CEA-NEXIA-GRS-KI on one hand, JAERI-VTT on the other hand; the (n,$\alpha$) reaction on $^{17}$O, (n,p) reaction on $^{14}$N and ternary fission yields have to be compared.

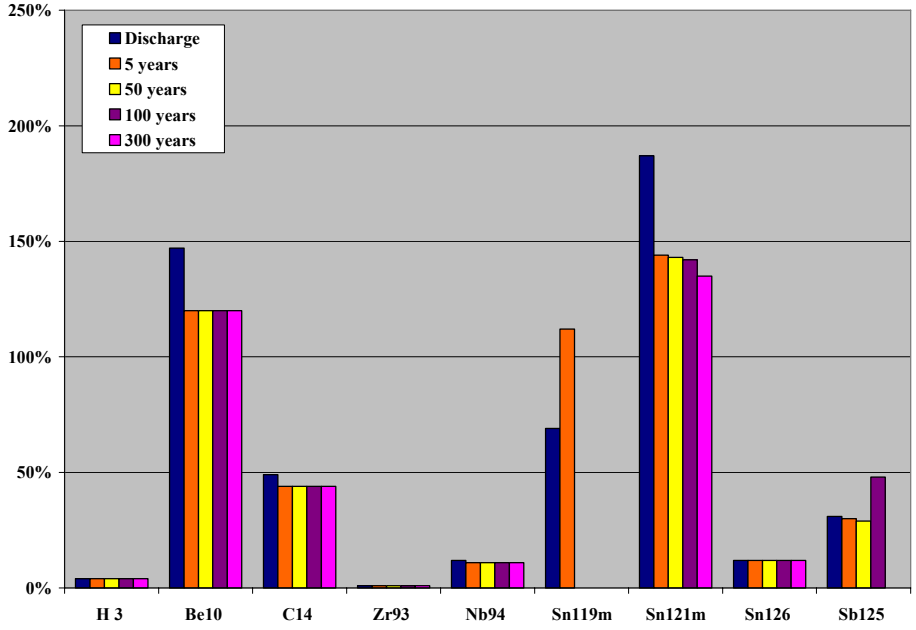
JAEA calculated masses with JENDL3.2 and JENDL3.3 for $^{94}$Nb are lower compared to all the other predicted values.

**Figure 6**: RSD results for Activation-Fission Products in assembly calculations



For $^{119m}$Sn, lower values are obtained by CEA-GRS-NEXIA-JAERI compare to the ones obtained by VTT and KI; this is particularly the case for the activation way of production and indicate probably a difference for $^{118}$Sn (n,γ) cross-section.

**Figure 7**: RSD results for Activation-Fission Products in cell calculations




### 5.3 Results of code to code comparison for calculated decay heat

Table 2 shows that a good agreement is obtained for the total decay heat calculation. However, because of the lack of measurement for this quantity, a conclusion about how accurate the calculation is cannot be drawn.

**Table 2**: RSD results for assembly and cell calculations of decay heat

| | Discharge | 5 years | 50 years | 100 years | 300 years |
|---|---|---|---|---|---|
| Assembly calculation | 3% | 2% | 1% | 2% | 2% |
| Cell calculation | 3% | 1% | 1% | 3% | 5% |

### 5.4 Results of code to code comparison for calculated neutron emission

The tables 3 and 4 indicate a high RSD for the total calculated neutron emission, about 20%; we can notice that the cell and assembly calculations are consistent. The total emission rate is mainly due to emission by spontaneous fission (SF), that's why the RSD on total neutron sources is the RSD on emission by SF.

**Table 3**: RSD results for assembly calculations of neutron emission rate

| | Discharge | 5 years | 50 years | 100 years | 300 years |
|---|---|---|---|---|---|
| (α,n) emission rate | 2% | 5% | 5% | 5% | 5% |
| Spontaneous fission (SF) | 19% | 29% | 29% | 26% | 20% |
| Total emission rate | 18% | 29% | 27% | 21% | 15% |

**Table 4**: RSD results for cell calculations of neutron emission rate

| | Discharge | 5 years | 50 years | 100 years | 300 years |
|---|---|---|---|---|---|
| (α,n) emission rate | 7% | 14% | 8% | 8% | 9% |
| Spontaneous fission (SF) | 17% | 26% | 26% | 23% | 17% |
| Total emission rate | 16% | 26% | 24% | 18% | 12% |

The discrepancies obtained on SF are in fact mainly due to the discrepancies on masses calculations, particularly on $^{244}$Cm mass calculation. Studies have shown that the spontaneous fission yields used by CEA, NEXIA, GRS, KI and VTT are nearly the same. The JAEA values seem to be different, particularly at discharge where the most contributing isotope is $^{242}$Cm.

For the (α,n) emission rate calculation, higher values for VTT and JAERI are observed. However study of individual nuclide contributions to the total (α,n) neutron emission and discrepancies on masses can't explain the discrepancies between participants: the (α,n) yields need to be compared.

## 6. Conclusion

The main results of this international depletion calculation comparison have been presented in this paper.

The following more significant conclusions are noted: there is a good agreement between participants for the mass calculation of many isotopes. However, it is interesting to observe that better agreement is obtained for isotopes which benefit from experimental validation. In this benchmark, the poorest agreement is obtained in calculating activation products originating from fuel impurities. The cell and assembly calculations are consistent and the method used for reaction rate calculation (deterministic or Monte-Carlo) does not seem to have a large impact on results obtained in the depletion calculation of this UOx Phase. Good agreement was also obtained for the total decay heat calculation. Some discrepancies on neutron emission rates were observed, mainly due to the discrepancies on masses calculations. Comparisons of some nuclear data (branching ratio, capture cross-section, half-life) have helped us to understand some discrepancies but it is necessary to go on with comparisons, particularly for activations products.

All the results and analyses related to this first phase will be published in an OECD/NEA report. The second phase of the benchmark devoted to MOx fuels has been defined in 2006 and is in progress.

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