

Unclassified

NEA/NSC/DOC(2007)9



Organisation de Coopération et de Développement Economiques
Organisation for Economic Co-operation and Development

14-Dec-2007

English text only

**NUCLEAR ENERGY AGENCY
NUCLEAR SCIENCE COMMITTEE**

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WORKING PARTY ON SCIENTIFIC ISSUES OF REACTOR SYSTEMS

**Specification for the Phase 2 of a Depletion Calculation Benchmark
devoted to MOx Fuel Cycles**

**B. Roque, P. Marimbeau, J.P. Grouiller, L. San-Felice
CEA/DEN/DER/SPRC/LECy
CEA Cadarache**

JT03238035

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CEA CADARACHE

1 INTRODUCTION

A depletion calculation benchmark devoted to MOx fuel is an ongoing objective of the OECD/NEA WPRS following the study of depletion calculation concerning UOx fuels [1].

As in the UOx phase, the objective of the proposed benchmark is to compare existing depletion calculations obtained with various codes and data libraries applied to fuel and back-end cycle configurations: transport, reprocessing, interim storage and waste repository. We propose to focus on nuclide densities of the most important nuclides implied in fuel cycle: Actinides, Fission Products and Activation Products and also on associated fuel cycle quantities: masses, neutron emission rate and decay heat.

Detected discrepancies between participants will enable us to improve the calculation schemes (self-shielding, subdivisions in fuel or moderator, ...), to improve the knowledge of burnup chains used in depletion calculations (maybe to recommend a chain for fuel cycle applications) and to improve the knowledge of nuclear data (capture cross-sections, branching ratio, fission yields, decay constants) involved in fuel cycle studies.

The following specification is devoted to the second phase of the Benchmark and focuses on MOx fuels, more precisely on the typical plutonium vector for material derived from the reprocessing of thermal reactor UO₂ fuels.

In the absence of the possibility to have open access to results from chemical experimental analysis of well-characterised irradiated fuel, we propose to base the benchmark on a comparison between trends obtained through a French experimental validation programme and the results obtained in this benchmark. However, because the purpose of the benchmark is not the experimental validation of code systems but the determination of a degree of consistency between the various depletion schemes, codes and nuclear data, the problem specification has been simplified somewhat to provide an approximate representation of the main fuel irradiation conditions. Nevertheless the comparison between the benchmark results and the trends, which have been obtained using a reference calculation and the actual configuration of the fuel, will enable us to provide a realistic degree of confidence for some isotopes.

This French programme has been carried out on fuel cuts coming from the first French reactor using MOx assemblies, the so-called SAINT-LAURENT B1 (SLB1) reactor, [2], [3], a 900 MWe PWR, characterised by a 30% MOx fuel loading. The standard MOx assemblies, used in Saint-Laurent B1 include three zones with different plutonium enrichments to flatten the within assembly power distribution and to attenuate fission rate discontinuities at the MOx-UOx interface. The central zone is characterized by a high Pu content (5.6%) and the peripheral zone by a low Pu content (2.9%).

The measured isotopes were major and minor actinides ^{234}U , ^{235}U , ^{236}U , ^{238}Pu , ^{239}Pu , ^{240}Pu , ^{241}Pu , ^{242}Pu , ^{237}Np , ^{241}Am , $^{242\text{m}}\text{Am}$, ^{243}Am , ^{243}Cm , ^{244}Cm , ^{245}Cm , ^{246}Cm , neodymium isotopes : ^{143}Nd , ^{144}Nd , ^{145}Nd , ^{146}Nd , ^{148}Nd , ^{150}Nd , caesium isotopes : ^{133}Cs , ^{134}Cs , ^{135}Cs , ^{137}Cs and the following fission products ^{147}Sm , ^{149}Sm , ^{150}Sm , ^{151}Sm , ^{152}Sm , ^{153}Eu , ^{154}Eu , ^{155}Eu , ^{154}Gd and ^{155}Gd .

The French experimental validation based on SLB1 has already been published [3],[4] and C/E results that can be used for the comparisons are given in Annexe A.

2 SPECIFICATION

For the purpose of the benchmark, we propose to focus on fuel with high burnup, around 40 GWd/t.

Two corresponding fuel rods shall be calculated: the L14 fuel rod located in the central zone of the MOx assembly and the Q17 fuel pin located in the peripheral zone (see Figure 1). This last pin will enable us to show the importance of the UOx environment in this mixed UOx-MOx core. Studies have shown that a standard calculation based on infinite medium pattern for MOx assembly can lead to an error of 30% in Pu239 concentration at 40 GWd/t for rods located at the peripheral zone [3].

2.1 MOx fuel Compositions

The initial MOx composition selected is representative of realistic MOx fuels irradiated in a mixed UO₂-MOx PWR core, alongside UO₂ fuel assemblies. This MOx fuel consists of a typical plutonium vector for material derived from reprocessing of thermal reactor UO₂ fuels.

The plutonium isotopic composition, corresponding to the three Pu content zones, is presented in **Table 1** and the uranium isotopic composition in **Table 2**.

The initial MOx fuel enrichments for these zones and the fuel composition are given in **Table 3** and **Table 4**.

Table 1: Plutonium Isotopic Composition in Fresh MOx Fuel

Nuclide	Isotopic Composition Atom%
Pu238	0.8
Pu239	66.7
Pu240	20.6
Pu241	7.5
Pu242	2.9
Am241	1.5

Table 2: Uranium Isotopic Compositions in Fresh MOx Fuel

Nuclide	Isotopic Composition Atom %
U-234	0.002
U-235	0.22
U-236	0.004
U-238	99.77

Table 3 : Initial MOx Fuel Content

Pu content Zone	MOX Fuel Plutonium content, w/o Putotal+Am/[U+Pu+Am]
High	5.6
Medium	4.4
Low	2.9

Table 4: Initial MOx Fuel Compositions (Atoms/barn.cm)

Nuclide	High	Medium	Low
U234	4.2175E-07	4.2718E-07	4.3391E-07
U235	4.9766E-05	4.8271E-05	4.9682E-05
U236	8.4350E-07	8.5435E-07	8.6782E-07
U238	2.1037E-02	2.1309E-02	2.1644E-02
Pu238	1.0815E-05	8.1476E-06	5.4861E-06
Pu239	8.3501E-04	6.5555E-04	4.3144E-04
Pu240	2.5798E-04	2.0151E-04	1.3387E-04
Pu241	9.4430E-05	7.4065E-05	4.8185E-05
Pu242	3.6112E-05	2.7751E-05	1.8859E-05
Am241	1.7374E-05	1.4626E-05	9.1090E-06
O16	4.4678E-02	4.4681E-02	4.4685E-02

2.2 UO₂ Fuel Compositions

For the calculations, the adjacent UO₂ fuel assemblies are modelled as already burned. They have an initial enrichment of 3.25 w/o and have reached a burnup of 24 GWd/t.

The composition of this irradiated UO₂ fuels is presented in **Table 5**.

Table 5: Initial Composition for irradiated UO₂ Fuel (Atoms/barn.cm)

U235	3.0E-04
U236	8.0E-05
U238	2.0E-02
Np237	7.1E-06
Pu238	1.7E-06
Pu239	1.2E-04
Pu240	3.8E-05
Pu241	2.1E-05
Pu242	5.3E-06
Am241	4.2E-07
Xe131	1.4E-05
Xe135	8.0E-09
Eu153	2.8E-06
Sm149	9.0E-08
Rh103	1.8E-05
Nd143	2.5E-05
Cs133	3.5E-05
Gd155	8.4E-10
Tc99	3.2E-05
Mo95	3.2E-05
Pm147	6.4E-06
Sm150	7.5E-06
Sm151	4.1E-07
Sm152	3.2E-06
O16	4.51E-02

2.3 Geometry Data

We propose to represent the MOx assembly together with three UO₂ fuel assemblies, as shown in Figure 1, with **translational boundary conditions**. The assembly geometry relates to a typical 17 x 17 PWR fuel assembly, as detailed below.

Fuel Pin Pitch: **1.262 cm**
 Fuel Pin Radius: **0.474 cm**
 Fuel Pellet Radius: **0.4126 cm**

The air gap between fuel and cladding is not modelled

The assembly channel box or water buffer is 0.155 cm thick.

For both UOx and MOx assemblies, the 24 guide tubes and 1 instrument tube shall be modelled **as water filled** zircaloy tubes with the following dimensions :

Outer Radius: **0.613 cm**
 Inner Radius: **0.571 cm**

2.4 Non-Fissile Material Data

The non-fissile materials are as follows:

Cladding: Zircaloy-4
 Guide Tubes: Zircaloy-4
 Coolant/Moderator: Light Water, 550 ppm Boron

For the purpose of the benchmark exercise, these materials should be modelled as specified in **Table 6**. A reduced density zircaloy has been specified for the fuel pin cladding to take account of the air gap between the fuel and cladding; the proposed concentrations in Table 6 use this reduced density.

Table 6: Non-Fissile Material Compositions

Table 6.1 : Cladding and Guide Tubes

Nuclide	Atoms/barn.cm
Zr (natural)	3.955E-02
Fe (natural)	1.383E-04
Cr (natural)	7.072E-05
O16	2.874E-04

Table 6.2 : Coolant/Moderator (550 ppm Boron)

Nuclide	Atoms/barn.cm
H	4.724E-02
O	2.362E-02
B10	4.321E-06
B11	1.739E-05

2.5 Impurities content for MOx fuels

In this benchmark, we propose to compare the activation products masses coming from initial fuel impurities. The values of the impurities content are specified in Table 7. We propose to use analogous values to that used in the UOx Phase.

Table 7: Initial fuel impurities

isotopes	atoms/tones HM	grams/tHM
H1	2.0491E+23	3.4026E-01
H2	3.0741E+19	1.0201E-04
B10	6.2931E+21	1.0454E-01
B11	2.5331E+22	4.6268E-01
C12	5.6257E+24	1.1210E+02
C13	6.2571E+22	1.3511E+00

N14	4.8610E+23	1.1301E+01
N15	1.7857E+21	4.4471E-02
Cl35	3.6473E+23	2.1198E+01
Cl37	1.1663E+23	7.1659E+00
Ca40	4.9532E+24	3.2900E+02
Ca42	3.3058E+22	2.3055E+00
Ca43	6.8978E+21	4.9252E-01
Ca44	1.0658E+23	7.7869E+00
Ca46	2.0438E+20	1.5610E-02
Ca48	9.5547E+21	7.6153E-01
Fe54	3.5435E+23	3.1776E+01
Fe56	5.6040E+24	5.2112E+02
Fe57	1.3441E+23	1.2722E+01
Fe58	1.7106E+22	1.6476E+00
Co59	1.3896E+23	1.3614E+01
Ni58	2.3817E+24	2.2939E+02
Ni60	9.1052E+23	9.0717E+01
Ni61	3.9421E+22	3.9931E+00
Ni62	1.2524E+23	1.2894E+01
Ni64	3.1746E+22	3.3739E+00
Cu63	7.4286E+23	7.7715E+01
Cu65	3.3110E+23	3.5739E+01
Zr90	3.8493E+23	5.7467E+01
Zr91	8.3944E+22	1.2672E+01
Zr92	1.2831E+23	1.9582E+01
Zr94	1.3003E+23	2.0277E+01
Zr96	2.0949E+22	3.3363E+00
Nb93	7.3465E+23	1.1334E+02
Mo92	3.1677E+23	4.8356E+01
Mo94	1.9745E+23	3.0796E+01
Mo95	3.3982E+23	5.3568E+01
Mo96	3.5604E+23	5.6716E+01
Mo97	2.0385E+23	3.2811E+01
Mo98	5.1507E+23	8.3759E+01
Mo100	2.0556E+23	3.4111E+01
Sn112	5.5781E+21	1.0365E+00
Sn114	3.7379E+21	7.0698E-01
Sn115	2.0702E+21	3.9500E-01
Sn116	8.3557E+22	1.6081E+01
Sn117	4.4165E+22	8.5733E+00
Sn118	1.3928E+23	2.7268E+01
Sn119	4.9341E+22	9.7419E+00
Sn120	1.8741E+23	3.7314E+01
Sn122	2.6626E+22	5.3896E+00
Sn124	3.3296E+22	6.8506E+00
O16	5.0502E+27	1.3418E+05
O17	1.9237E+24	5.4321E+01
O18	1.0125E+25	3.0272E+02

2.6 Irradiation Histories

The requested calculations should be performed to attain a constant target burnup of 38 GWd/t and 42 GWd/t for the two studied MOx fuel pins. The MOx fuel irradiation history shall be represented over the three following operating cycles :

Q17 MOx fuel pin Cycle 1: **285 days full power**, EOC burnup = **12 GWd/t**
Downtime: **60 days**
Cycle 2: **300 days full power**, EOC burnup = **25 GWd/t**
Downtime: **40 days**
Cycle 3: **280 days full power**, EOC burnup = **38 GWd/t**

L14 MOx fuel pin Cycle 1: **285 days full power**, EOC burnup = **12 GWd/t**
Downtime: **60 days**
Cycle 2: **300 days full power**, EOC burnup = **25 GWd/t**
Downtime: **40 days**
Cycle 3: **280 days full power**, EOC burnup = **42 GWd/t**

2.7 Cooling: 0 years (discharge), 5, 50, 100, 300 years

2.8 Material Temperatures

Fuel Temperature: **900K**
Cladding Temperature: **620K**
Moderator Temperature: **582K**

3 NUCLIDES AND QUANTITIES REQUIRED

Please forward the results by electronic mail to Dr. Laurence SAN-FELICE at CEA/CADARACHE [**laurence.san-felice@cea.fr**](mailto:laurence.san-felice@cea.fr).

For the two studied fuel pins, the required quantities are :

- Masses of the nuclides specified in the table above in **grams/tHM**
- Neutron emission : (alpha,n) emission, spontaneous fission and total emission in **neutrons/second/tHM**
- Decay heat : alpha, beta and gamma decay heat and total decay heat in **Watts/tHM**

The required quantities should be forwarded using the following tables (you can copy the Tables in Microsoft Excel and send them as an attached file to the e-mail).

Table 8: Masses of activation products

MASSES OF ACTIVATION PRODUCTS (g/tHM)					
Nuclide	Discharge	5 years	50 years	100 years	300 years
Cl36					
Ca41					
Mn53					
Mn54					
Fe55					
Fe60					
Co60					
Ni59					
Ni63					
Mo93					

Some nuclides are produced both by fission reaction and activation reaction. We propose to evaluate separately the two contributions in the table above. *If your fuel cycle code does not separate the two contributions, please replace the two lines FP and AP in the table above by only one line with FP+AP.*

Table 9: Masses of fission and activation products

MASSES OF FISSION AND ACTIVATION PRODUCTS (g/ tHM)						
nuclide		Discharge	5 years	50 years	100 years	300 years
H3	FP (fission)					
	AP (activation)					
Be10	FP					
	AP					
C14	FP					
	AP					
Zr93	FP					
	AP					
Nb94	FP					
	AP					
Sn119m	FP					
	AP					
Sn121m	FP					
	AP					
Sn126	FP					
	AP					
Sb125	FP					
	AP					

Table 10 : Masses of fission products

MASSES OF FISSION PRODUCTS (g/ tHM)					
Nuclide	Discharge	5 years	50 years	100 years	300 years
Se79					
Kr85					
Rb85					
Rb87					
Sr88					
Sr90					
Nb93m					
Mo95					
Mo97					
Tc99					
Ru101					
Ru106					
Rh103					
Pd107					
Ag108m					
Ag109					
Ag110m					
I127					
I129					
Xe130					
Xe131					
Xe132					
Xe134					
Xe136					
Cs133					
Cs134					
Cs135					
Cs137					
Ba136					
Ba138					
La139					
Ce140					
Ce144					
Nd142					
Nd143					
Nd144					
Nd145					
Nd146					
Nd148					
Nd150					
Pm147					
Sm146					
Sm147					
Sm148					
Sm149					
Sm150					

Sm151					
Sm152					
Sm154					
Eu153					
Eu154					
Eu155					
Gd154					
Gd155					
Gd156					
Ho166m					

Table 11 : Masses of actinides

Nuclide	MASSES OF ACTINIDES (g/ tHM)				
	Discharge	5 years	50 years	100 years	300 years
U232					
U233					
U234					
U235					
U236					
U238					
Np236					
Np237					
Pu236					
Pu238					
Pu239					
Pu240					
Pu241					
Pu242					
Pu243					
Pu244					
Am241					
Am242m					
Am243					
Cm242					
Cm243					
Cm244					
Cm245					
Cm246					
Cm247					
Cm248					
Ra226					
Ra228					
Ac227					
Th229					
Th230					
Th232					
Cf252					

Table 12 : Neutron emission rate (neutrons/s/ tHM)

NEUTRON EMISSION RATE (neutrons/s/ tHM)					
	Discharge	5 years	50 years	100 years	300 years
(α,n) emission rate					
Spontaneous Fission emission rate					
Total emission rate					

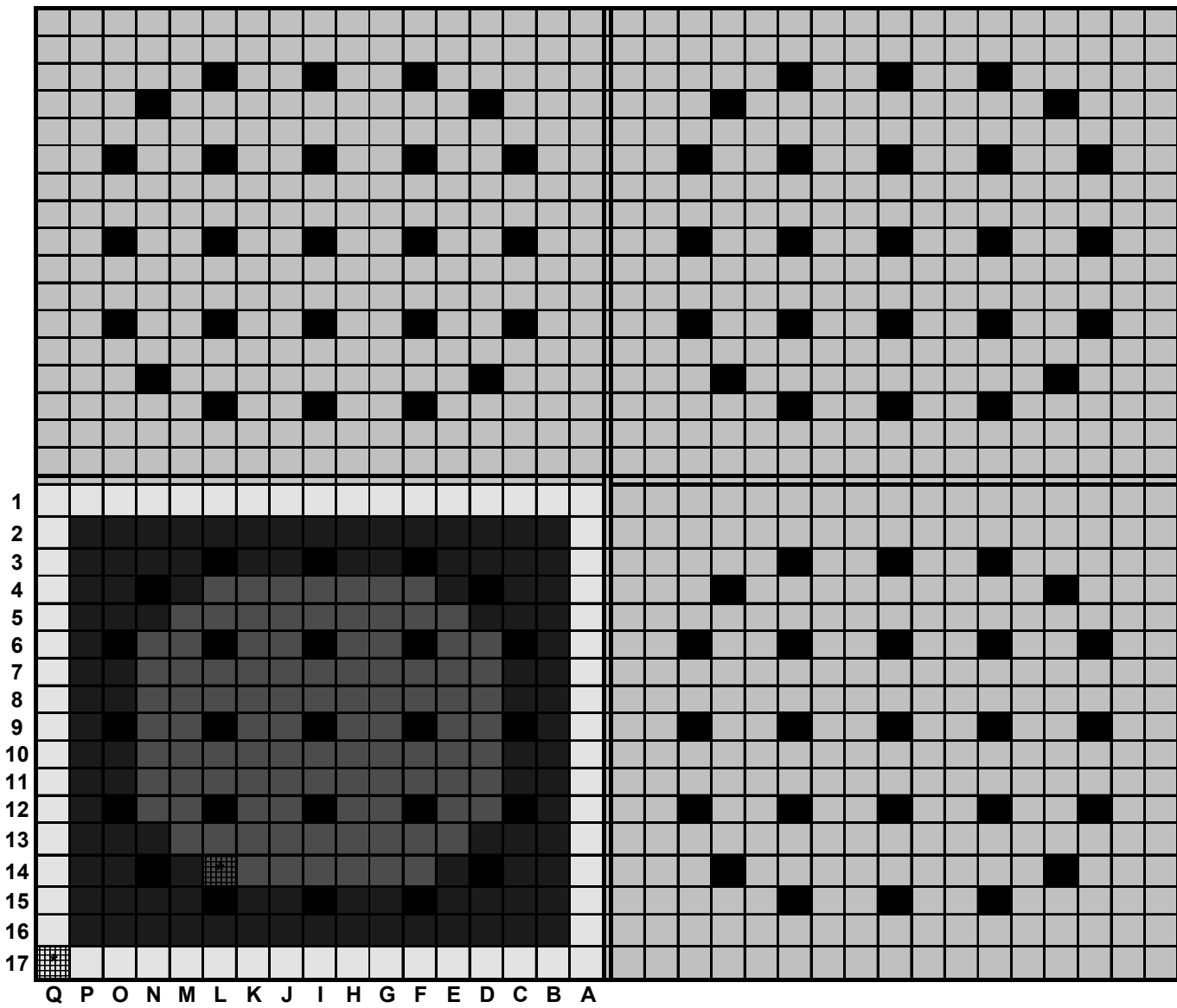
Table 13 : Decay heat (Watts/ tHM)

DECAY HEAT (Watts/ tHM)					
	Discharge	5 years	50 years	100 years	300 years
Alpha Decay Heat					
Beta Decay Heat					
Gamma Decay Heat					
Total Decay Heat					

4 OPTIONAL CALCULATION

An additional but optional calculation is suggested; it concerns a sensitivity calculation linked to the UOx environment.

A calculation considering the MOx assembly in infinite medium could be performed and a comparison on the masses obtained for the L14 fuel pin previously calculated could be done.








-  - Selected MOX pin for the Benchmark
-  - MOX pin low Pu content
-  - MOX pin intermediate Pu content
-  - MOX pin high Pu content
-  - UO₂ pin (3.25w/o U-235/U Enrichment, 24GWd/t)

Figure 1 : MOx -UO₂ geometry

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ANNEX A**C/E obtained by CEA for experimental validation of MOx fuels [4]**

The calculation-experiment comparison (C-E)/E (in %) obtained with the CEA DARWIN package is summarised below.

In the following tables, the C/E values are the mean values over several samples and the total uncertainties (2σ) correspond to the combination of uncertainties on chemical assays, determination of the burnup of the assembly derived from Nd isotopics and some assumptions on irradiation history. When results for more than one sample are available for the same burnup, the spread of results is also considered.

For the benchmark, we will use only the last line corresponding to a mean burnup of roughly 40 GWd/t. The French experimental validation have shown that there is no trend with the location of the fuel pin in the assembly, so that the same value can be use for both the L14 and Q17 pins (excepted the ^{234}U value).

Table I : (C-E)/E (%) for 'uranium' inventory

Fuel↓	BU (GWd/t)↓	$^{234}\text{U}/^{238}\text{U}$	$^{235}\text{U}/^{238}\text{U}$	$^{236}\text{U}/^{238}\text{U}$
MOx SLB1	10	4.4 ± 4.0	0.3 ± 1.0	-7.6 ± 2.0
	30	-5.0 ± 2.6	1.2 ± 2.0	-5.8 ± 2.0
	42	-3.1 ± 2.4	1.7 ± 2.8	-4.8 ± 1.2

Table II : (C-E)/E (%) for 'plutonium' inventory

Fuel↓	BU(GWd/t)↓	$^{238}\text{Pu}/^{238}\text{U}$	$^{239}\text{Pu}/^{238}\text{U}$	$^{240}\text{Pu}/^{238}\text{U}$	$^{241}\text{Pu}/^{238}\text{U}$	$^{242}\text{Pu}/^{238}\text{U}$
MOx SLB1	10	-6.3 ± 1.2	0.05 ± 1.5	0.6 ± 0.5	-5.2 ± 1.0	-1.4 ± 1.0
	30	-6.3 ± 1.0	-0.2 ± 2.0	0.9 ± 0.5	-3.0 ± 1.0	-2.7 ± 2.0
	42	-6.3 ± 1.0	-1.6 ± 3.0	0.7 ± 1.0	-3.5 ± 1.5	-3.9 ± 2.0

Table III : (C-E)/E (%) for 'neptunium' inventory

Fuel↓	BU (GWd/t)↓	$^{237}\text{Np}/^{238}\text{U}$
MOx SLB1	10	-11.3 ± 5.0
	30	-11.1 ± 4.0
	42	-7.4 ± 4.0

Table IV : (c-E)/E (%) for 'americium' inventory

Fuel↓	BU (GWd/t)↓	²⁴¹ Am/ ²³⁸ U	^{242m} Am/ ²³⁸ U	²⁴³ Am/ ²³⁸ U
MOx SLB1	10	-4.3 ± 3.0	-27.7 ± 4.0	-11.1 ± 4.0
	30	2.3 ± 2.6	-23.3 ± 4.0	-7.1 ± 4.0
	42	1.6 ± 3.5	-22.7 ± 4.0	-6.0 ± 3.0

Table V : (c-E)/E (%) for 'curium' inventory

Fuel↓	BU (GWd/t)↓	²⁴³ Cm/ ²³⁸ U	²⁴⁴ Cm/ ²³⁸ U	²⁴⁵ Cm/ ²³⁸ U	²⁴⁶ Cm/ ²³⁸ U
MOx SLB1	10	-39.8 ± 4.0	-29.5 ± 4.0	-30.6 ± 6.0	-59.4 ± 9.0
	30	-22.3 ± 4.0	-6.6 ± 4.0	-8.3 ± 6.0	-16.6 ± 9.0
	42	-16.5 ± 3.5	-5.6 ± 4.0	-7.6 ± 5.0	-14.6 ± 8.0

Table VI : (c-E)/E (%) for 'neodymium' inventory

Fuel↓	BU (GWd/t)↓	¹⁴³ Nd/ ²³⁸ U	¹⁴⁴ Nd/ ²³⁸ U	¹⁴⁸ Nd/ ²³⁸ U	¹⁵⁰ Nd/ ²³⁸ U
MOx SLB1	10	0.0 ± 4.0	-1.8 ± 3.0	0.3 ± 4.0	0.3 ± 3.0
	30	0.2 ± 3.4	-2.5 ± 3.0	0.2 ± 4.0	0.9 ± 3.0
	42	1.1 ± 2.0	-2.2 ± 3.0	0.4 ± 2.0	0.4 ± 3.0

Table VII : (c-E)/E (%) for 'caesium' inventory

Fuel↓	BU (GWd/t)↓	¹³³ Cs/ ²³⁸ U	¹³⁴ Cs/ ²³⁸ U	¹³⁵ Cs/ ²³⁸ U	¹³⁷ Cs/ ²³⁸ U
MOx SLB1	10	-2.9 ± 3.0	-10.4 ± 5.0	3.1 ± 2.0	-3.6 ± 2.8
	30	-0.2 ± 2.0	-7.2 ± 5.0	4.7 ± 1.0	-1.8 ± 2.4
	42	-0.6 ± 2.0	-6.4 ± 5.5	1.7 ± 2.0	-2.6 ± 2.4

Table VIII : (c-E)/E (%) for 'samarium' inventory

Fuel↓	BU (GWd/t)↓	¹⁴⁷ Sm/ ²³⁸ U	¹⁴⁹ Sm/ ²³⁸ U	¹⁵⁰ Sm/ ²³⁸ U	¹⁵¹ Sm/ ²³⁸ U	¹⁵² Sm/ ²³⁸ U
MOx SLB1	10	-5.7 ± 3.0	-7.1 ± 1.0	7.1 ± 4.5	-8.1 ± 1.5	-5.3 ± 4.0
	30	-3.9 ± 2.0	2.6 ± 2.0	-6.3 ± 4.5	-1.2 ± 1.5	-1.4 ± 3.0
	42	-3.8 ± 1.0	-7.7 ± 1.0	-5.7 ± 1.2	1.9 ± 1.0	2.9 ± 2.0

Table IX : (c-E)/E (%) for 'europium and gadolinium' inventory

Fuel↓	BU (GWd/t)↓	¹⁵³ Eu/ ²³⁸ U	¹⁵⁴ Eu/ ²³⁸ U	¹⁵⁵ Eu/ ²³⁸ U	¹⁵⁴ Gd/ ²³⁸ U	¹⁵⁵ Gd/ ²³⁸ U
MOx SLB1	10	-2.7 ± 10.0	-0.9 ± 15.0	83.8 ± 8.0	-0.3 ± 18.0	68.8 ± 8.0
	30	4.0 ± 10.0	13.2 ± 18.0	61.6 ± 15.0	14.2 ± 18.0	53.9 ± 10.0
	42	8.7 ± 5.5	41.5 ± 15.0	24.4 ± 8.0	38.5 ± 13.0	14.4 ± 10.0