ISOTOPIC INVENTORIES OF THE DISCHARGED T37 FUEL BUNDLES

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ABSTRACT

The paper describes studies that have been undertaken to analyse the mass inventory of the thorium-uranium mixed oxide fuel removed from the standard CANDU 6 reactor. The analysed T37 fuel bundle configuration is based on thorium and low enriched uranium (LEU) oxide, considering the ThO_2 ratio of 100% in the central pin, 60% in the inner and intermediate rings and 80% in the outer ring. The ^{235}U enrichment has been varied in the radial direction from 6% to 10%.
The study focuses on identifying optimal thorium – uranium combination from the viewpoint of reducing the content of plutonium and highly radiotoxic minor actinides content the spent fuel accumulated in 30 years operating reactor.
The lattice simulations have been performed using the Continuous-energy Monte Carlo Reactor Physics Burn-up Calculation Code - SERPENT 2, version 2.1.30. In order to perform the depletion calculations, the ACE format cross section library JEFF-3.1.1 have been used.

Key words: neutronic evaluation, burn-up, CANDU, cell calculation, SERPENT

Introduction

Along with a safe, clean, CO_2 free and economically competitive source of energy, the sustainable expansion of nuclear power implies the development of technologies able to minimize the overall spent nuclear fuel (SNF) from Gen. II and III reactors.
The reduction of plutonium (Pu) and minor actinides (MA) from Light Water Reactors (LWR) fuel cycles constitutes an important objective. As known, Pu (the fissionable isotope ^{239}Pu) is the main material used in nuclear weapons manufacturing, while the high radiotoxicity and heat generation caused by MA are a major problem in the management of the SNF.
The research studies have been shown that thorium based fuel may play an important role in the reduction of Pu inventories. The capture of a neutron by an ^{238}U nucleus generates ^{239}Pu, while ^{232}Th transmutes to the fissionable ^{233}U. Althought the Significant Quantity of the ^{233}U is similar to that of ^{239}Pu (around 8 kg) [1], the contamination with ^{232}U provides better nonproliferation characteristics than ^{239}Pu [1]. The presence of ^{232}U, which decay to an intense gamma ray's emitter, ^{208}Tl, makes ^{233}U hard to handle and easy to detect [2].

Description of the problem

The selection of a suitable Th-based fuel composition to be used in CANDU 6 nuclear system requires integrated studies on neutronic, thermohydraulic and nuclear safety considerations. The neutronic evaluations for the thorium-based fuel bundle configuration (named here T37) given in Ref. [3] have been shown good neutronic results. Such that, the analyses T37 fuel bundle configuration is based on thorium and low enriched uranium (LEU) oxide, considering the ThO_2 ratio of 100% in the central pin, 60% in the inner and intermediate rings and 80% in the outer ring. For this configuration, the fissile ^{235}U enrichment has been varied in the radial direction as follows:

- 6% in the inner and in the intermediate rings and 7% in the outer ring (case T37_667),
- 7% in the inner and in the intermediate rings and 6% in the outer ring (case T37_776).

The fuel mass density is 9 g/cm^3 for ThO_2 and 9.6 g/cm^3 for $(Th,U)O_2$.
The composition of the irradiated fuel has been computed using the Continuous-energy Monte Carlo Reactor Physics Burn-up Calculation Code - SERPENT 2 [4], version 2.1.30. In order to perform the depletion calculations, the ACE format cross section library and the decay and fission yield data based on JEFF-3.1.1 [5] have been used.
SERPENT code provides the variations of the atomic densities N of the fuel components in a fuel bundle.
The atomic denstities are evaluated for burn-up or time intervals according to the nuclear reactions and radioactive transformations processes as shown in Fig. 1 [1].

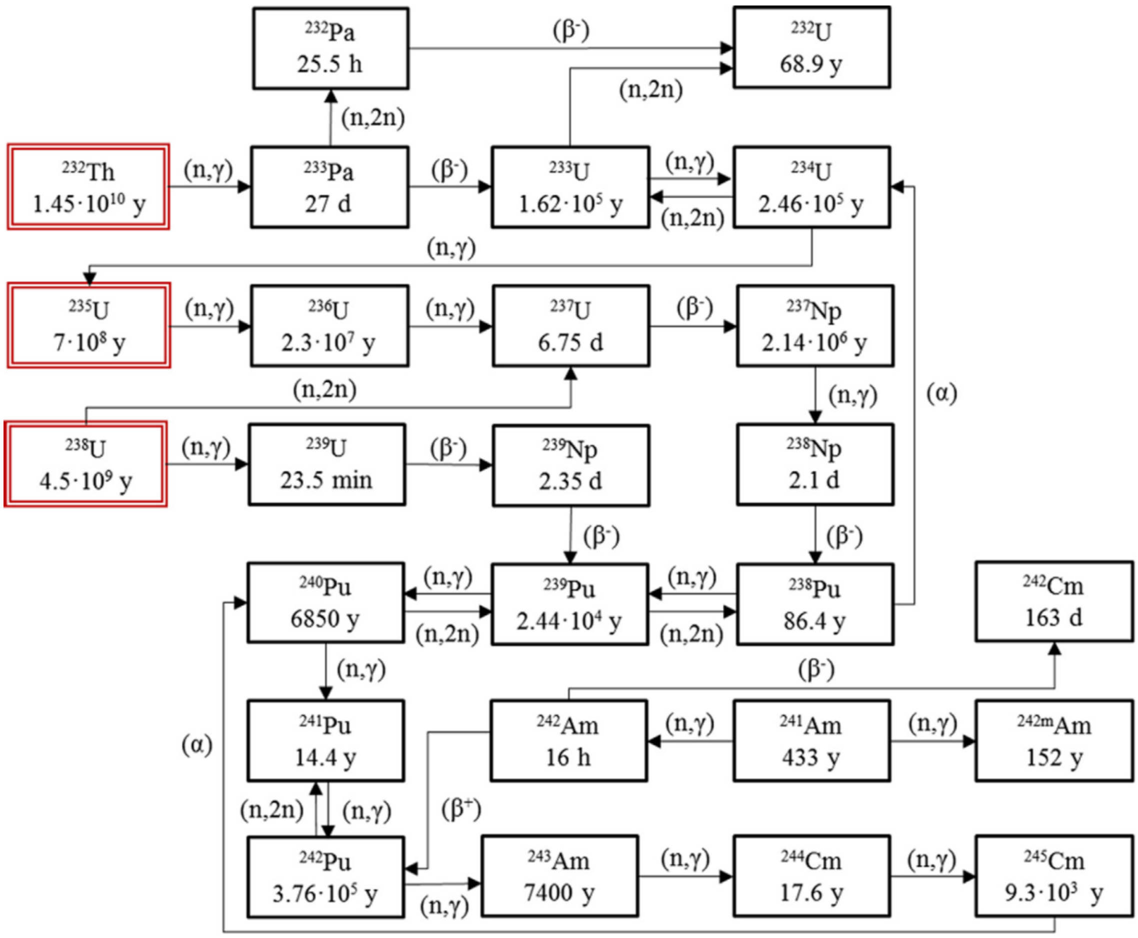

Fig. 1. *Major reactions and decay processes in CANDU reactor operation with Th-U based fuel [1]*

The simulations consist in an infinite 3D geometry defined by a cuboidal CANDU-6 lattice cell containing the fuel bundle surrounded by the moderator. Throughout the burn-up calculation, the average power of a reference CANDU 6 fuel bundle (800 kW) is used for the source rate normalization. CANDU 6 fuel bundle data (geometry, structural material, coolant, moderator), other than thorium-based fuel, are taken from reference [6].

Results and discussions

During reactor operation, the isotopic composition of the fuel changes with burn-up and some new fissionable isotopes, such as ^{233}U, ^{239}Pu, and ^{241}Pu will be produced through neutron capture in fuels followed by β^- decays, such as ^{232}Th, ^{238}U, and ^{240}Pu, respectively. Fissionable isotopes are then transmuted by nuclear fission reactions in fission products. Obviously, the composition of the discharged fuel depends on the general characteristics of the reactor (core design and performances, fuel type, fissile enrichment and energy extracted from the fuel).

The mass inventory of the discharged fuel has been evaluated as element or isotop mass to the extracted energy per fuel bundle. Let us note that the discharge burn-up obtained from SERPENT 2 calculation for U_{nat} bundle is 7.4 MWd/kg-U, 16 MWd/kg-HM, for T37_667 case and 19 MWd/kg-HM for T37_776 case.

The Pu, MA, U and the fission products (FP) masses per each extracted MWd have been shown in comparison with the reference case of the reactor fuelled with standard UO_2 fuel. The accumulations of the total Pu and main Pu isotopes are illustrated in Fig. 2.

As can be observed, T37 bundle cases produce a considerably lower quantity (over 80% less) of Pu in comparison with the reference U_{nat} fuel bundle.

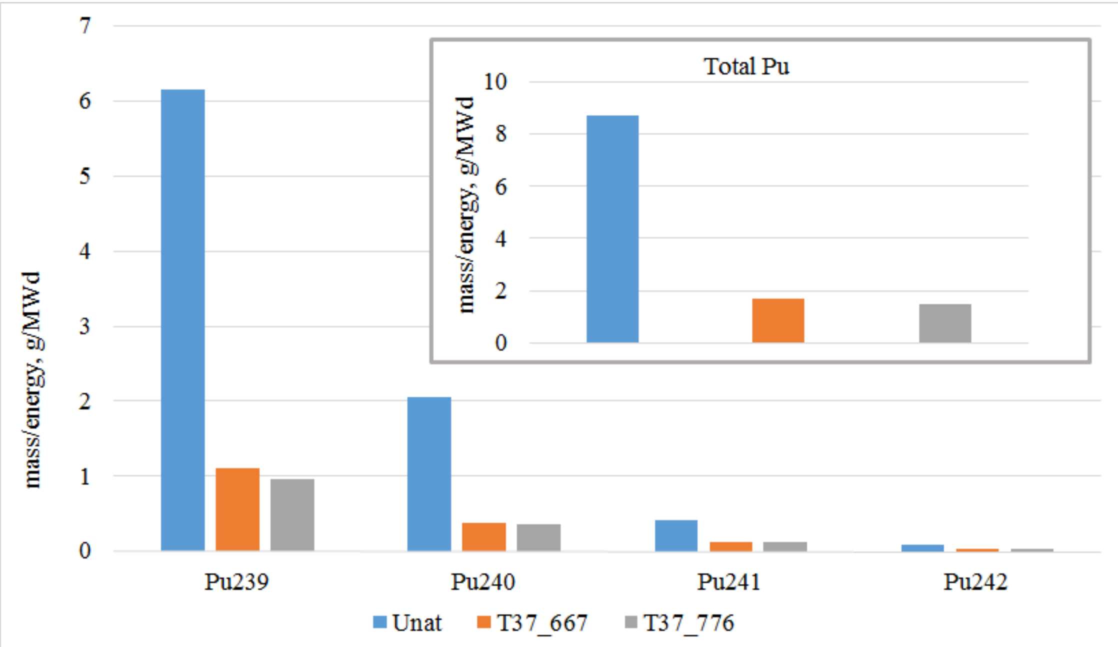

Fig. 2. *Inventory of Pu: major isotopes and total mass per discharged bundle given as mass/ extracted energy.*

The mass inventories of the main MA (Np, Am and Cm) are shown in Figs. 3 – 5. The T37 bundle cases produce lower amounts of Np, Am and Cm than the U_{nat} bundle. Quantitatively, the main isotope from the MA vector is ^{239}Np in the U_{nat} case while ^{237}Np constitues the main isotopes in T37 cases. Although ^{239}Np has a short half-life, it disintegrates to ^{239}Pu.

The total amount of MA per each extracted MWd is around 80% lower for T37 bundle cases than U_{nat} (see Fig. 6).

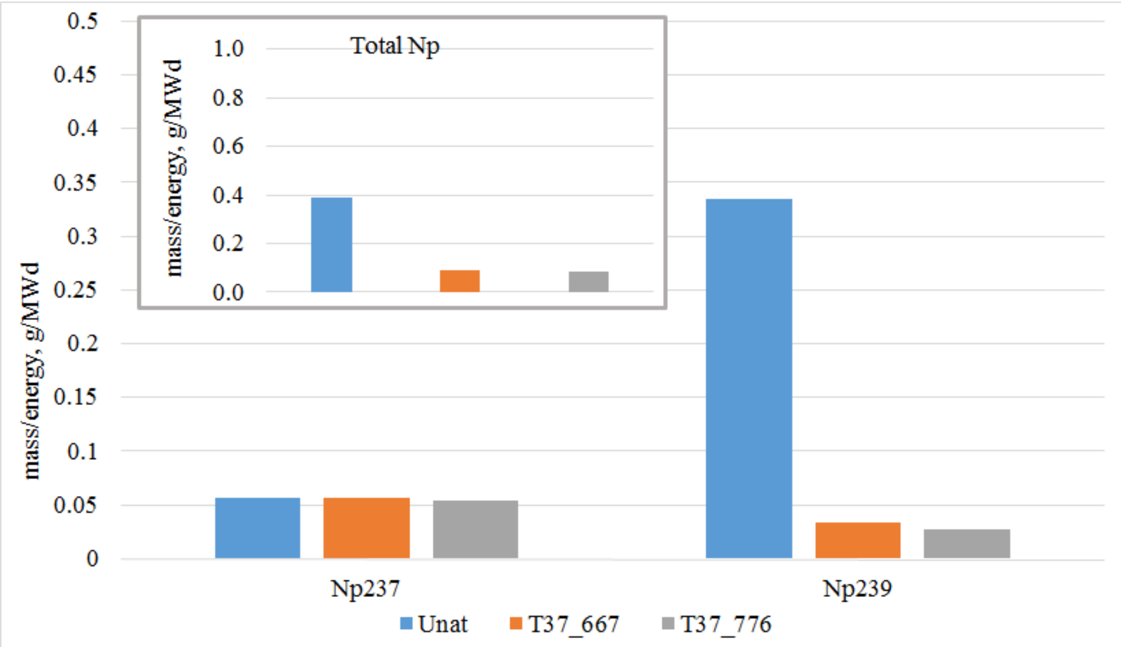

Fig. 3. *Inventory of Np: major isotopes and total mass per discharged bundle given as mass/ extracted energy.*

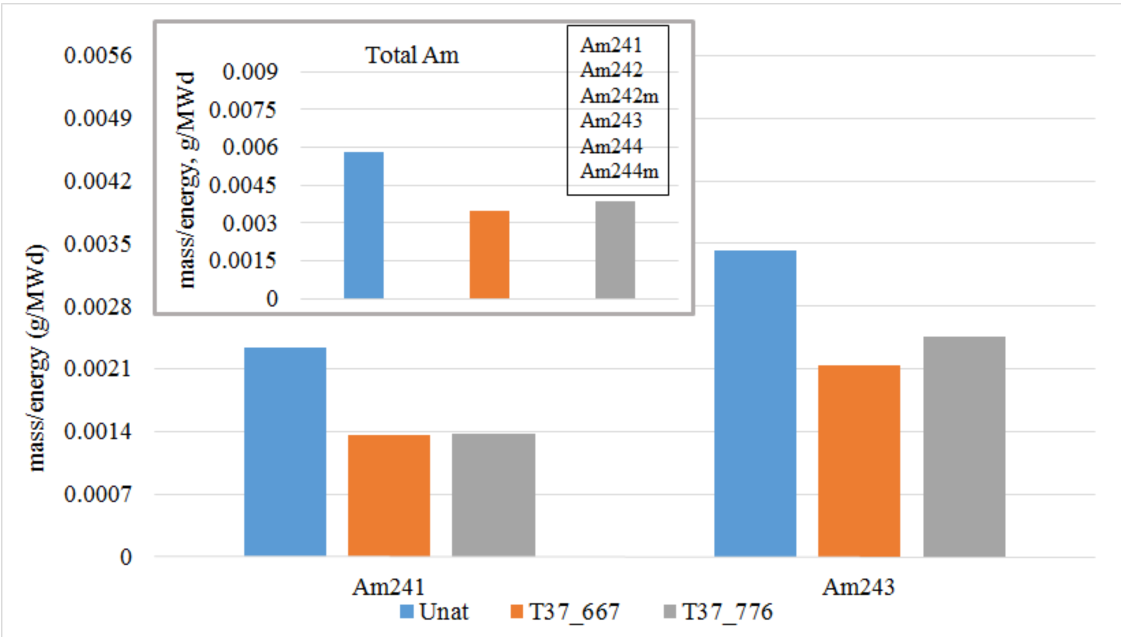

Fig. 4. *Inventory of Am: major isotopes and total mass per discharged bundle given as mass/extracted energy.*

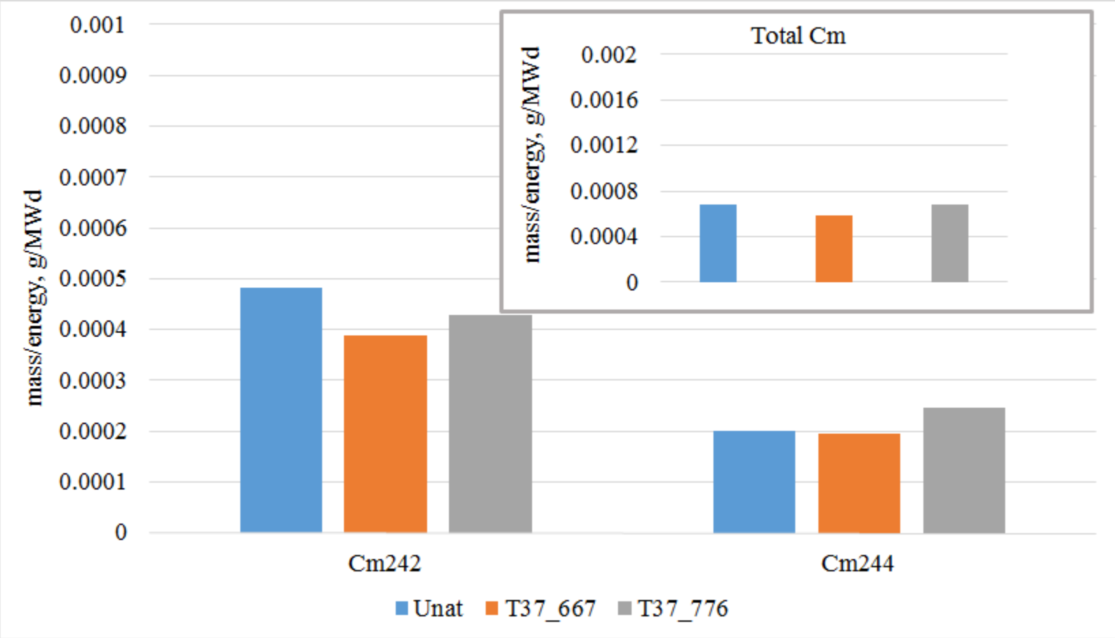

Fig. 5. *Inventory of Cm: major isotopes and total mass per discharged bundle given as mass/ extracted energy*

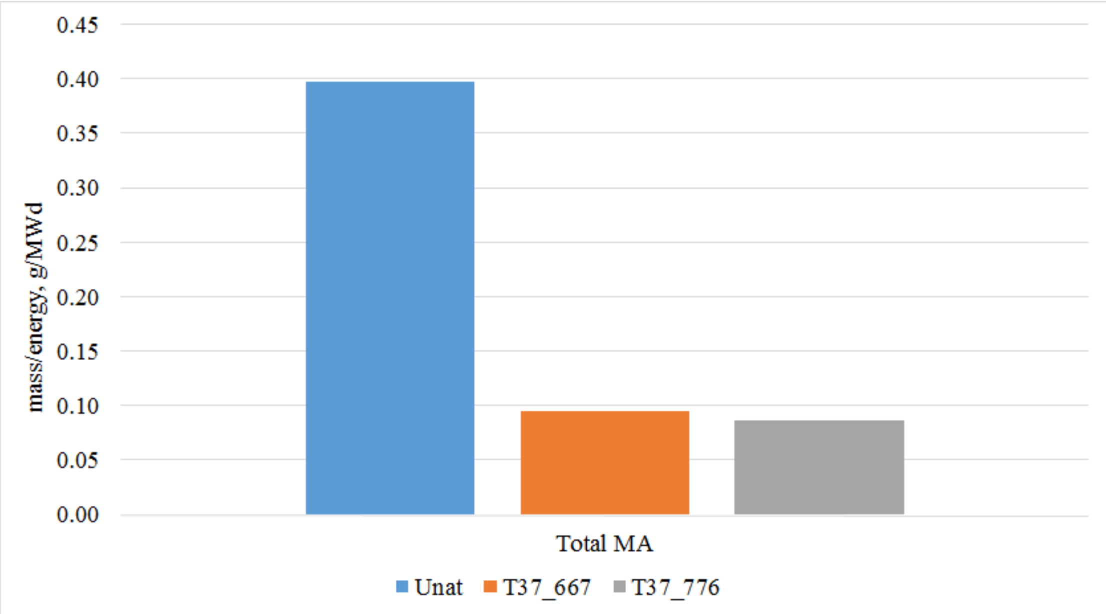

Fig. 6. *Total minor actinides mass per bundle given as mass/extracted energy*

Fig. 7 gives the masses of the main uranium isotope: the two fisionable ^{233}U and ^{235}U and the fertile ^{238}U. Unlike U_{nat} bundle case, the discharged T37 fuel contains less Pu and MA, but higher amounts of ^{233}U, thus increasing the total amount of weapon usable material. Obviously, $^{233,235}U$ can be recycled along with thorium and other actinides. On the other hand, the high radiotoxicity of ^{232}U, present in burnt $(Th,U)O_2$ fuel, can induce difficulties in the spent fuel reproccesing.
For nonproliferation considerations, the fissionable isotopes inventory per discharged fuel bundle have primary importance. Table 1 gives the estimated amounts per each extracted MWd of pertinent fissionable isotopes $^{238,241}Pu$ and $^{233,\,235}U$ from T37 bundle in comparison with U_{nat} bundle

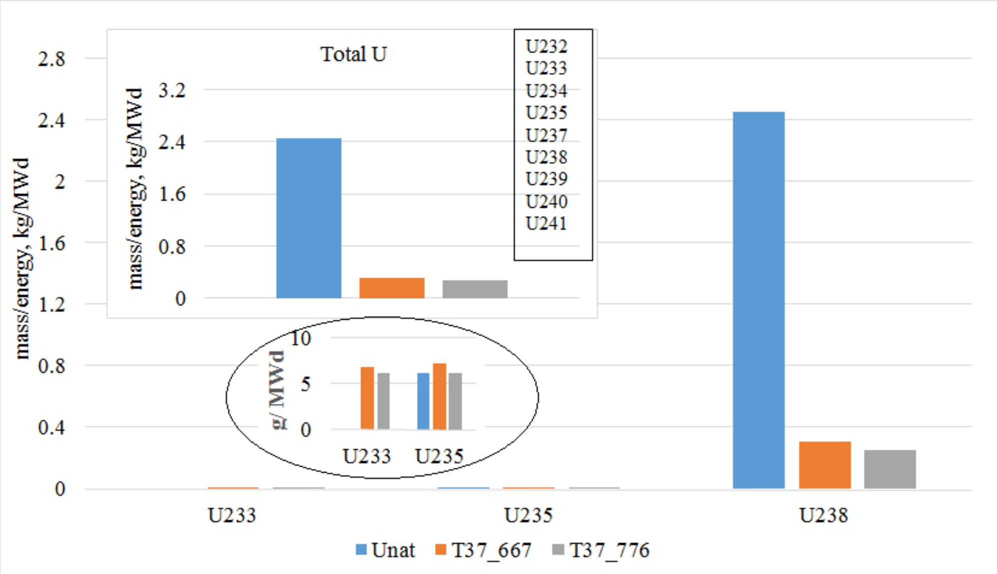

Fig. 7. *The major U isotopes and total mass of U per discharged bundle given as mass/extracted energy*

Table 1. *The fissionable isotopes mass inventory given as mass/extracted energy (g/MWd)*

Case	^{233}U	^{235}U	^{239}Pu	^{241}Pu	Total
U_{nat}	0	6	6	0.4	12.4
T37_667	7	7	1	0.1	15.1
T37_776	6	6	1	0.1	13.1

The total mass of actinides is around 18.3 kg for U_{nat} bundle and around 17.3 kg for T37 bundle. Obviously, the mass conservation law leads to a increase of the total FP mass in the case of T37 bundle from 2.6 kg in the case of U_{nat} bundle to 2.7 kg in the case of T37 bundle. The FP mass inventories per each extracted MWd are shown in Fig. 8 for T37 cases in comparison with U_{nat} bundle.

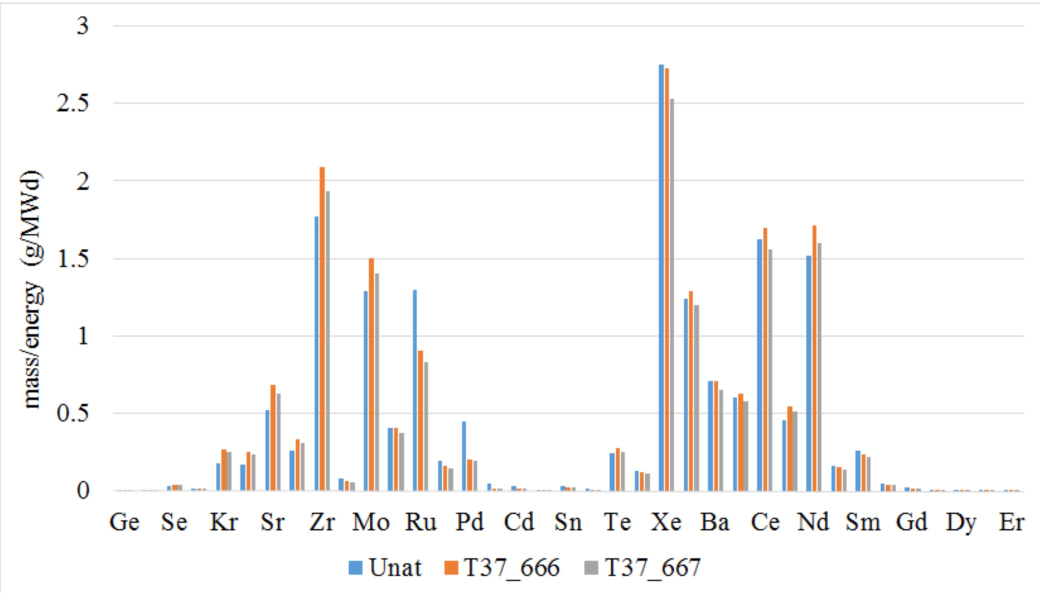

Fig. 8. *Fission products mass inventory given as mass/extracted energy*

Conclusions

The current study represents an investigation on the isotopic mass inventories of the thorium-uranium mixed oxide fuel $(Th,U)O_2$ removed from the standard CANDU 6 reactor.
The SERPENT 2 results have been shown that the partial substitution of LEU with thorium leads to the reduction of the overall Pu and MA accumulation per MWd of extracted energy in the discharged T37 bundle fuel and to the increasing of uranium fissionable isotopes (^{233}U and ^{235}U) amounts.
The overall production of fissionable materials ($^{233,235}U$, $^{239,241}Pu$) is higher in the discharged T37 bundle cases than in the reference U_{nat} bundle.
The contamination with ^{232}U of the discharged fuel could improve the nonproliferation resistance but can imply higher fuel costs and supplementary radioprotection measures.
The present results show that, due to the higher fuel burn-up grade, T37 fuel bundle has the advantage of significantly reducing the total amount of spent nuclear fuel per each MWd of extracted energy.

References

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