EVALUATION OF SPENT FUEL INVENTORY AND RADIOACTIVITY FOR CANDU TYPE FUEL BUNDLES WITH INCREASED NUMBER OF ELEMENTS CONTAINING U-BASED FUEL COMPOSITIONS

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Abstract

Study of nuclear fuel behaviour during irradiation in the reactor, spent fuel characteristics evaluation after discharge from the reactor core and transfer to intermediate storage for cooling and radioactivity decreasing are topics of great interest among the nuclear safety analyses. CANDU reactors are well-known for the versatility of operation in alternative fuel cycles, other than the natural uranium one, using a variety of fuels, but a simple fuel assembly project. The paper aimed to evaluate spent fuel inventory (fissile material and isotopes of interest for proliferation resistance) and radioactivity for several CANDU type fuel bundle projects with increased number of elements (43, 52, or 61 fuel elements) containing U-based fuels (natural uranium, slightly enriched uranium or recycled uranium). The 37-element CANDU standard fuel bundle (C37) with natural uranium fuel was considered as reference. The parameters of interest, both during irradiation and at the End of Irradiation (EoI), were obtained from ORIGEN-S burn-up code (in SCALE 6 package) simulations. Comparisons for different fuel compositions in the same fuel bundle, and between different fuel bundles with the same fuel composition were carried out. Comparison against the reference CANDU fuel bundle was also performed. Spent fuel fissile inventory for C37 was greater than the ones corresponding to fuel bundle projects with increased number of elements. The inventory of isotopes interesting for proliferation resistance produced in fuel bundles with increased number of elements was greater than the one characterizing the reference. The total radioactivity in fuel for C37 fuel bundle with natural uranium fuel was greater than the ones estimated for the fuel bundle projects with increased number of elements.

Keywords: *Advanced CANDU fuel bundles, U-based fuels, Spent fuel, Fissile Inventory, Proliferation resistance, Radioactivity*

Introduction

CANDU technology is a mature, well-proven nuclear technology, CANDU reactors being characterized by a high level of safety and security, but also having an evolutionary potential in terms of proliferation resistance, versatility to use different alternative fuel cycles which ensures a superior use of nuclear fuel resources.

CANDU reactors are heavy water cooled and moderated, and operate with natural uranium fuel contained in 0.5 m long cylindrical bundles, typically composed by 37 clustered elements. Twelve fuel bundles are inserted end-to-end within a 6 m long pressure tube, through which pressurized heavy water coolant is circulated. In turn, the pressure tube is inserted into the calandria tube composing the fuel channel.

Several hundred pressure tubes compose the fissile core, contained within a horizontal tank ("calandria") filled with low-pressure heavy water moderator. The space between the pressure and calandria tubes is filled with a gas which insulates the cool moderator from the hot heat transferred to coolant by the fuel. The reactor is continuously refuelled at full power, using remotely-operated machines connected to individual fuel channels.

The alternative fuel cycles, known as advanced fuel cycles, include a variety of fissile materials: Slightly Enriched Uranium fuel (with up to 1.2 wt% enrichment in U^{235}), Recycled Uranium fuel, Natural Uranium Equivalent fuel, Mixed Oxide fuels, and Thorium-based fuels. Despite the great diversity of fuels to be used in CANDU reactors, the fuel assembly is simple and evolved in time improving the reactor operation and fuel performance, increasing the operation limits and cost efficiency, assuring adaptability to advanced fuels and fuel cycles.

CANDU type fuel assembly evolution is mainly marked by increasing the number of fuel elements. In June, 1962, started to operate NPD (Nuclear Power Demonstration) reactor (Rolphton nearby Chalk River, Canada), the first Canadian power reactor which will become later the prototype for CANDU reactors (Canada Deuterium-Uranium). NPD reactor uses 7-element fuel bundles in the outer zone and 19-element wire wrap fuel bundles in the inner zone (see Figs. 1 and 2).

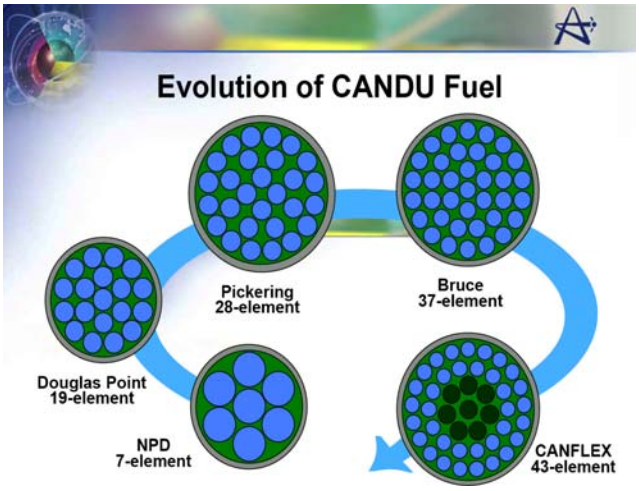


Fig. 1. CANDU type fuel assembly evolution – increasing the number of fuel elements [6]

Based on the NPD operating experience and the fabrication costs, the 19-element fuel assembly (see Fig. 2) was modified by upgrading its thermal performance and was used in Douglas Point nuclear power plant, NPP (Canada), [1-7].



Fig. 2. 7-element fuel bundle (NPD) and 19-element fuel bundles (Douglas Point) [5]



Fig. 3. 28-element fuel assembly (Pickering) and 37-element fuel assemblies (Bruce, Gentilly) [1]

The pressure tube became larger and, consequently, the fuel bundle diameter increased by adding more fuel elements and the 28-element fuel bundle (see Figs. 1 and 3) was designed to be used in Pickering NPP (Canada). Another step in CANDU type fuel assembly evolution was the 37-element fuel bundle project (see Figs. 1 and 3) developed for the Bruce NPP reactors (Canada), with a capacity of 750 MWe. After modifying the spacers, the 37-element fuel assembly will become the CANDU standard fuel bundle widely used in CANDU 600 MWe reactors operating in Canada (Gentilly NPP) and other countries (Embalse-Argentina, Qinshan-China, Cernavoda-Romania, Wolsong-South Korea), [1, 5-7].

CANDU type fuel assembly evolution continued and 43-element CANFLEX (CANDU Flexible) fuel bundle was developed [1, 8] (see Fig.1), marking a turning point by considering fuel elements with different diameters. Following to achieve higher fuel burn-up and increased fuel performance, the diameter of the central element and the fuel elements placed on the inner ring was increased comparatively with the other fuel elements in the assembly. Although the fuel bundle thermal power increased, the linear power on the elements was diminished by increasing the number of elements, which led to milder operating conditions: reduced fission gases release, lower values for total sheath deformation and central temperature in the fuel pellet than those characterizing the standard fuel bundle [8]. CANFLEX fuel assembly is associated with using of various U-based fuels (slightly enriched uranium, recycled uranium, natural uranium equivalent) and thorium based fuel for operating the CANDU reactors [1, 8].

It should mention the Argentinean efforts to develop a CANDU type fuel assembly compatible with both CANDU reactor (Embalse NPP) and PHWR Siemens (Atucha NPP) and assuring the increase of NPPs performance, especially based on the use of slightly enriched uranium fuel. This fuel assembly was named CARA (Combustible Avanzado para Reactores Argentinos) [9-11], a 52-element CANDU fuel assembly twice longer than the CANDU standard one.

The research carried out at the Institute for Nuclear Research (RATEN ICN) Pitesti on nuclear fuels, include various activities for developing/testing/characterizing of new fuel assembly projects and new fuels for higher discharge fuel burn-up, radioactive waste amount reduction, resources optimization, increasing the reliability in operation for U-based fuels, study of fuel behaviour in safe and secure operation of CANDU reactor.

Nuclear Fuel Performance group at the RATEN ICN has developed several CANDU type fuel bundles with increased number of fuel elements, as follows: C43 and CN43 (CANDU New) fuel assemblies with 43 fuel elements [12, 13], inspired by the Canadian CANFLEX project; 52-element C52 fuel assembly [14], inspired by the Argentinean CARA project, but as long as the CANDU standard fuel bundle; and 61-element C61 fuel assembly [14], integral Romanian design with selected constructive characteristics to allow its using in CANDU reactors at Cernavoda NPP (Romania).

A comparative study was carried out for the CANDU type fuel bundles with increased number of fuel elements (43, 52 or 61 fuel elements) developed at RATEN ICN Pitesti, assuming different U-based fuels and fuel irradiation in CANDU reactor conditions. The 37-element CANDU standard fuel bundle, C37, with natural uranium fuel, was considered as reference.

For the proposed analyses, the following U-based fuel compositions were selected: natural uranium (NU); slightly enriched uranium (SEU) 0.9 wt% and 1.0 wt% enriched in U^{235}; recycled uranium (RU) 0.9 wt% and 1.0 wt% enriched in U^{235}, respectively.

Spent fuel radioactive inventory (fissile isotopes U^{235} and Pu^{239}, and isotopes of interest for proliferation resistance – Pu^{238}, Pu^{240} and Pu^{242}) and radioactivity (fuel total radioactivity, radioactivity in actinides and fission products) at the End-of-Irradiation (EoI) were estimated.

Fuel irradiation was simulated by means of ORIGEN-S burn-up code included in SCALE6 programmes package (developed by Oak Ridge National Laboratory, USA) [15] assuming that all elements in the fuel bundle contain the same fuel composition.

Comparisons of different fuel compositions in the same fuel bundle and between different fuel bundles with the same fuel composition were performed. The parameters of interest were also comparatively analyzed against the CANDU standard fuel bundle ones.

Initial data for fuel burn-up simulation, computer codes used, assumptions

Fuel bundle projects considered

In present study, the 37-element CANDU fuel bundle, also known as the CANDU standard fuel bundle based on its worldwide utilization in the CANDU 600 MWe reactors (Canada, Argentina, China, Romania, South Korea), was used as reference.

C37 is a cylindrical bundle about 50 cm long, with a diameter of 10 cm, and weighs slightly over 24 kg, containing about 22 kg of NU fuel. It contains 37 identical fuel elements (rods) disposed on 3 concentric rings of 6, 12 and 18 elements, around a central element [5-7] (see Fig.4).

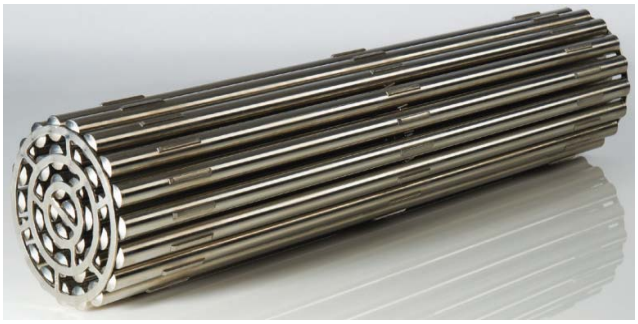


Fig. 4. CANDU standard fuel bundle (C37) view [7]

Sintered NU fuel pellets are inserted in sealed zirconium alloy tubes of about 1.5 cm in diameter and with 0.4 mm thick sheath. The clustered elements are stiffened in the assembly by two end plates, on outer fuel ring of the bundle being welded bearing pads that assure the bundle easy sliding inside the pressure tube and leave enough space for the circulation of the coolant [5-7].

Four CANDU type fuel bundle projects developed, updated and optimized by the Nuclear Fuel Performances Group at RATEN ICN have been studied, namely: C43 and CN43 fuel bundles (43 elements), C52 fuel bundle (52 elements), and C61 fuel bundle (61 elements), respectively. CANDU type fuel bundle projects geometry is similar: cylindrical shape, fuel elements disposed in annular geometry on concentric rings and one central element, except for C52 fuel bundle where the central element is replaced by a 4-element ring.

CANDU type fuel bundles projects selected for the present comparative analysis are briefly presented as follows:

▪ C43 fuel bundle was developed, updated and optimized [12] aiming compatibility with the Canadian CANFLEX fuel bundle in terms of geometry and performances. C43 fuel bundle contains 43 elements disposed on 3 concentric rings of 7, 14, and 21 elements, respectively, around a central element (see Fig.5). There are 8 thick elements (central element and 7 elements on the inner ring) and 35 thin elements (14 elements on intermediate ring and 21 elements on the outer ring); ″thick″ or ″thin″ refers to fuel element outside diameter compared to the standard CANDU one. C43 fuel bundle has the same length as the C37 fuel bundle, but it is lighter (about 23.5 kg weight) and contain about 21 kg of fuel [12].

▪ CN43 (CANDU New 43) fuel bundle [13] was developed aiming a perfect compatibility with the fuel bundles used at Cernavoda NPP for the two operating CANDU units, and accomplishing several goals: upgrading C43 fuel bundle project; higher fuel burn-up, especially by using RU slightly enriched (0.9 wt% - 1.1 wt% in U^{235}); reduction of the radioactive waste amount to be stored, increased resistance to power ramps and fuel bundles failure, increased safety operation limits. CN43 fuel bundle contains 43 identical fuel elements disposed on 3 concentric rings of 7, 14 and 21 elements around a central element; fuel element is 0.1 mm thinner than the reference one.

▪ C52 fuel bundle project was developed [14] to be used at Cernavoda NPP without any change at the fuel channel. It was inspired by the Argentinean advanced 52-element fuel bundle CARA, but the same length as C37 fuel bundle was considered. C52 fuel bundle is composed by 52 identical fuel elements disposed on 4 concentric rings of 4, 10, 16 and 22 elements, respectively (see Fig.6a); fuel element is 0.205 mm thinner than the reference one.

▪ C61 fuel bundle project [14] is an integral Romanian design with selected constructive characteristics that make it able to be used in CANDU reactors at Cernavoda NPP (Romania). C61 fuel bundle is composed by 61 identical fuel elements disposed on 4 concentric rings of 6, 12, 18 and 24 elements around a central element (see Fig.6b); fuel element is 0.262 mm thinner than the reference one.

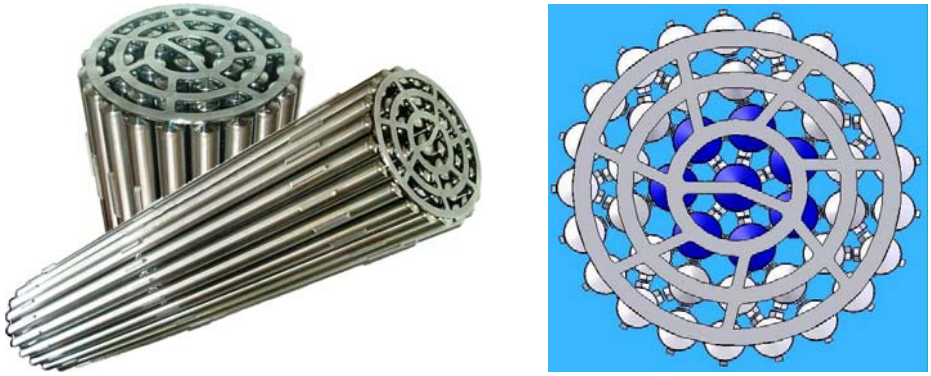


Fig. 5. 43-element CANDU type fuel bundle (C43) view [12]

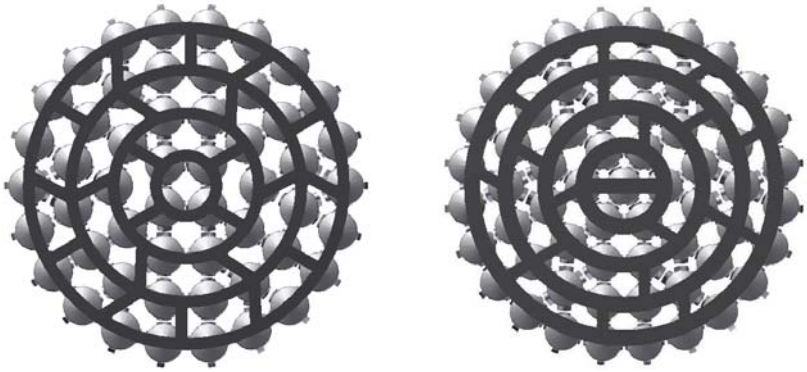


Fig. 6. C52 (a) and C61(b) CANDU type fuel bundle's view [14]

U-based fuel compositions analysed

The analyses proposed for the advanced CANDU type fuel bundles with increased number of fuel elements, were performed for the following U-based fuel compositions:
- Natural Uranium (NU) [16-20];
- Slightly Enriched Uranium (SEU) 0.9 wt% and 1.0 wt% enriched in U^{235} [17-20];
- Recycled Uranium (RU) 0.9 wt% and 1.0 wt% enriched in U^{235} [17-21].

Table 1 presents the initial materials inventory for the U-based fuel compositions selected for study (Uranium isotopes, Oxygen, Zr alloy cladding, UO_2) and the fuel bundles weight. The working cases identification, according to the fuel composition and fuel bundle project, is also given in Table 1. Zr alloy cladding contains Zirconium, alloying elements (Tin, Iron, Chromium, and Nickel) and some impurities (Carbon and Oxygen).

Table 1. Materials initial inventory in the selected U-based fuels, [16-21]

Working case ID	Enr. in U^{235} [wt%]	U^{234} [kg]	U^{235} [kg]	U^{236} [kg]	U^{238} [kg]	U [kg]	O_2 [kg]	Zy [kg]	UO_2 [kg]	Fuel bundle [kg]
C37NU	0.71	0	0.138	0	19.357	19.495	2.620	2.144	22.115	24.259
C43NU	0.71	0	0.133	0	18.512	18.645	2.506	2.253	21.150	23.403
C43SEU09	0.9	0	0.168	0	18.477					
C43SEU10	1.0	0	0.186	0	18.458					
C43RU09	0.9	0.004	0.168	0.037	18.436					
C43RU10	1.0	0.004	0.186	0.037	18.417					
CN43NU	0.71	0	0.134	0	18.657	18.791	2.525	2.237	21.316	23.553
CN43SEU09	0.9	0	0.169	0	18.622					
CN43SEU10	1.0	0	0.188	0	18.603					
CN43RU09	0.9	0.004	0.169	0.037	18.580					
CN43RU10	1.0	0.004	0.188	0.037	18.561					
C52NU	0.71	0	0.133	0	18.341	18.474	2.483	2.245	20.956	23.201
C52SEU09	0.9	0	0.166	0	18.307					
C52SEU10	1.0	0	0.185	0	18.289					
C52RU09	0.9	0.004	0.166	0.037	18.267					
C52RU10	1.0	0.004	0.185	0.037	18.248					
C61NU	0.71	0	0.139	0	19.219	19.359	2.602	2.479	21.961	24.439
C61SEU09	0.9	0	0.174	0	19.184					
C61SEU10	1.0	0	0.194	0	19.165					
C61RU09	0.9	0.004	0.174	0.039	19.142					
C61RU10	1.0	0.004	0.194	0.039	19.122					

Simulation of fuel burnup - computer codes used, data and assumptions

CANDU reactors specific conditions of irradiation have been simulated using the ORIGEN-S code included in SCALE6 programs package [15], up to 8 MWd/kgHE (NU fuel) and 12 MWd/kgHE (SEU and RU fuels), respectively. Fuel irradiation was performed at 34.644 kW/kgHE specific burn-up power, 30 burn-up steps being considered. For all the fuel bundle elements identical fuel composition was assumed. Spectral indices (thermal, epithermal and fast) characterising the fuel burn-up were previously obtained in lattice cell computations using DRAGON3.05E and WIMS programs [22-24].

The spent fuel parameters of interest for the U-based CANDU fuel simulation calculations were:
- inventory of fissile material isotopes (U^{235} and Pu^{239});
- inventory of isotopes interesting for proliferation resistance (Pu^{238}, Pu^{240} and Pu^{242});
- total radioactivity in the spent fuel;
- radioactivity of actinides and fission products..

Results and discussions

The paper proposed to investigate the behaviour to irradiation in CANDU reactor specific conditions of several CANDU type fuel bundles with increased number of elements by estimation of the spent fuel inventory (fissile isotopes and isotopes of interest for proliferation resistance) and radioactivity at EoI. For the proposed analyses, some U-based fuels were considered.

The spent fuel parameters of interest were comparatively analyzed for the different CANDU type fuel bundles with the same fuel composition, and for the same fuel bundle with different fuel compositions. Considering the CANDU standard fuel bundle (C37 with NU fuel, C37NU) as reference, a comparison of the results obtained for C43, CN43, C52 or C61 fuel bundles against the reference was also performed.

Spent fuel inventory of the interest isotopes

For the present study, the fissile isotopes (U^{235} and Pu^{239}) and the isotopes considered for the proliferation resistance (Pu^{238}, Pu^{240} and Pu^{242}) were estimated both during irradiation and at the EoI.

The fissile material amount reduces by about 15%-20% per irradiation step as the fuel burn-up increases due to the higher consumption of U^{235} (65% - 85% as enrichment in U^{235} becomes higher) comparatively to the Pu^{239} accumulation (30% - 65% for increasing U^{235} enrichment).

The amounts of isotopes interesting for proliferation resistance present a significant increasing as the fuel burn-up becomes higher, the relative differences calculated at EoI relatively to the first step of irradiation being 87% for NU fuel, and over 90% for SEU and RU fuels.

Tables 2-4 presents the fissile isotopes inventory at EoI in the considered CANDU fuel bundles for fuel compositions with natural uranium, slightly enriched uranium and recycled uranium, respectively. In Table 2, the values corresponding to C37NU (the reference) are also given.

Table 2. Inventory of fissile isotopes at EOI [g/assembly] in CANDU fuel bundles with NU fuel

Working Case ID	Inventory of fissile isotopes [g/assembly]		
	U^{235}	Pu^{239}	Total
C37NU	32.6	24.5	57.1
C43NU	28.8	23.4	52.2
CN43NU	27.6	22.6	50.2
C52NU	22.1	23.4	45.5
C61NU	21.0	22.3	43.3

Table 3. Inventory of fissile isotopes at EOI [g/assembly] in CANDU fuel bundles with SEU fuel

Working Case ID	Inventory of fissile isotopes [g/assembly]		
	U^{235}	Pu^{239}	Total
C43SEU09	27.6	23.8	51.4
C43SEU10	27.8	23.6	51.4
CN43SEU09	23.0	22.6	45.6
CN43SEU10	23.1	22.6	45.7
C52SEU09	22.6	22.3	44.9
C52SEU10	22.7	22.2	44.9
C61SEU09	20.7	23.3	44.0
C61SEU10	20.8	23.2	44.0

Table 4. Inventory of fissile isotopes at EOI [g/assembly] in CANDU fuel bundles with RU fuel

Working Case ID	Inventory of fissile isotopes [g/assembly]		
	U^{235}	Pu^{239}	Total
C43RU09	27.4	22.6	50.2
C43RU10	27.2	22.8	50.2
CN43RU09	22.3	23.0	45.3
CN43RU10	22.1	23.2	45.3
C52RU09	21.8	22.4	44.2
C52RU10	21.6	22.6	44.2
C61RU09	21.4	22.0	43.4
C61RU10	21.2	22.2	43.4

Analysing the fissile inventory, it can be noticed that C37 fuel bundle with NU fuel has the highest amount of fissile material (57.1 g/assembly), the lowest fissile material amount being estimated for C61 fuel bundle with NU fuel (43.4 g/assembly). The highest consumption of U^{235} was estimated for C61 fuel bundle with SEU fuel (0.9 wt% enr. in U^{235}); the lowest accumulation of Pu^{239} was obtained for C61 fuel bundle with RU fuel (0.9 wt% enr. in U^{235}).

Extending the analysis at the fuel bundle level for different U-based fuels, the following aspects were observed:

▪ CANDU fuel bundles with 43 fuel elements (C43 and CN43) - the highest consumption of U^{235} was obtained using RU fuel, the lowest U^{235} amounts being estimated in CN43 fuel bundle. As for Pu^{239} accumulation, the highest value was associated with C43 fuel bundle, 23.8 g/assembly (by 1.7% higher than the value obtained when using NU fuel), while lowest values were obtained for CN43 fuel bundle with SEU fuel, 22.6 g/assembly, the same values being obtained in CN43U fuel bundle.

▪ C52 fuel bundle - the highest consumption of U^{235} was obtained using RU fuel (21.6 g/assembly - C52RU10), the lowest Pu^{239} accumulation being obtained for SEU fuel (22.2 g/assembly - C52SEU10).
▪ C61 fuel bundle - the highest consumption of U^{235} was obtained using SEU fuel (20.7 g/assembly - C61SEU09), the lowest Pu^{239} accumulation being obtained for RU fuel (22.0 g/assembly - C52RU09).

Tables 5-7 presents the inventory of isotopes interesting for proliferation resistance at EoI in the considered CANDU fuel bundles for fuel compositions with natural uranium, slightly enriched uranium and recycled uranium, respectively. The values corresponding to C37NU (the reference) are also included in Table 5.

Table 5. Inventory of interest for proliferation resistance at EOI [g/assembly] in CANDU fuel bundles with NU fuel

Working case ID	Inventory of isotopes interesting for non-proliferation [g/assembly]			
	Pu^{238}	Pu^{240}	Pu^{242}	Total
C37NU	0.1	23.7	1.6	25.3
C43NU	0.05	23.8	1.8	26.4
CN43NU	0.03	23.7	1.7	25.4
C52NU	0.03	23.9	1.9	25.9
C61NU	0.04	25.1	2.0	27.1

Table 6. Inventory of interest for proliferation resistance at EOI [g/assembly] in CANDU fuel bundles with SEU fuel

Working case ID	Inventory of isotopes interesting for non-proliferation [g/assembly]			
	Pu^{238}	Pu^{240}	Pu^{242}	Total
C43SEU09	0.5	29.4	2.8	32.7
C43SEU10	0.6	30.0	3.1	33.7
CN43SEU09	0.2	26.6	2.6	29.4
CN43SEU10	0.3	27.6	2.9	30.7
C52SEU09	0.2	26.2	2.5	28.9
C52SEU10	0.3	28.0	3.2	31.4
C61SEU09	0.2	27.4	2.7	30.3
C61SEU10	0.3	29.3	3.3	32.9

Table 7. Inventory of interest for proliferation resistance at EOI [g/assembly] in CANDU fuel bundles with RU fuel

Working case ID	Inventory of isotopes interesting for non-proliferation [g/assembly]			
	Pu^{238}	Pu^{240}	Pu^{242}	Total
C43RU09	0.3	28.2	3.1	31.6
C43RU10	0.4	29.4	3.6	33.4
CN43RU09	0.1	26.7	2.8	29.6
CN43RU10	0.2	28.4	3.0	31.6
C52RU09	0.1	26.0	2.7	28.8
C52RU10	0.2	27.6	3.0	31.6
C61RU09	0.1	28.4	2.9	31.4
C61RU10	0.2	29.0	3.3	32.5

Analysing the above results, it can be observed that the highest amount of inventory interesting for proliferation resistance (33.7 g/assembly) was obtained for C43SEU10 fuel bundle, the lowest inventory being estimated for CN43 fuel bundle with NU fuel (25.4 g/assembly, with only 0.4% higher than the one corresponding to C37NU). Pu^{240} inventory represents the major contributor to the total inventory of isotopes interesting for proliferation resistance, its share being about 90% - 94%. To be mentioned that for using of NU fuel, the highest inventory interesting for non-proliferation was estimated for C61 fuel bundle (27.1 g/assembly), closely followed by C43 fuel bundle (26.4 g/assembly).

At the fuel bundle level, using considered U-based fuels led to the following observations:
- C43 fuel bundle - the highest inventory of isotopes interesting for proliferation resistance was obtained using SEU fuel, 33.7 g/assembly (C43SEU10), with 21.7% and 0.9% higher than the ones obtained for NU and RU fuels, respectively.
- CN43 fuel bundle - the highest inventory of isotopes interesting for proliferation resistance was obtained using RU fuel, 31.6 g/assembly (CN43RU10), with 20% and 2.8% higher than the ones obtained for NU and SEU fuels, respectively.
- C52 fuel bundle - similar behaviour as CN43 fuel bundle, the highest inventory of isotopes interesting for non-proliferation being obtained for RU fuel (31.6 g/assembly in C52RU10), while using of NU fuel conducted to the lowest values, 25.9 g/assembly.
- C61 fuel bundle - similar behaviour as C43 fuel bundle, the highest inventory of isotopes interesting for non-proliferation being obtained for SEU fuel (32.9 g/assembly in C6SEU10), with 17.6% and 1.2% higher than the ones obtained for NU and RU fuels, respectively.

Spent fuel radioactivity

To investigate the behaviour of the considered CANDU type fuel bundles to irradiation, the spent fuel radioactivity was estimated both during fuel irradiation and at EoI. The radioactivity characterizing the fuel bundles with different U-based fuels was estimated both as total radioactivity of the fuel and radioactivity of components (cladding, actinides and fission products). The cladding radioactivity is significantly lower than the ones of actinides or fission products, representing only up to 0.03% from the total radioactivity, while the radioactivity of actinides and fission products contribution reaches 25% and 74.7%, respectively. Therefore, the total radioactivity and the radioactivity in actinides and fission products will be further analysed.

Figs. 7-9 illustrate the evolution of the radioactivity (total, and in actinides and fission products) with the fuel burn-up for the considered fuel bundles with NU, SEU and RU fuels.

The total radioactivity values in the fuel become higher by up to 10% (for SEU fuel) as the fuel irradiation increases. The radioactivity of the actinides follows a similar trend as the total radioactivity in fuel increasing by up to 45% (for SEU fuel) as the fuel irradiation becomes higher. Meanwhile, the radioactivity of the fission products slightly reduces by up to 2% (for SEU fuel), but it has a quite insignificant effect on the total radioactivity that continues to grow further due to actinides contribution, as can be easily seen from the previous figures.

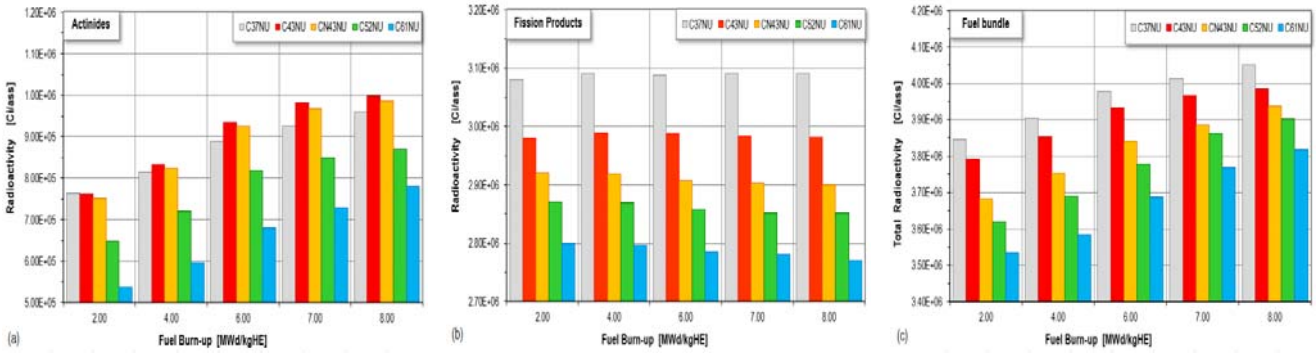


Fig. 7. Spent fuel radioactivity [Ci/assembly] evolution during fuel irradiation for NU fuel in:
(a) actinides, (b) fission products, and (c) fuel bundle

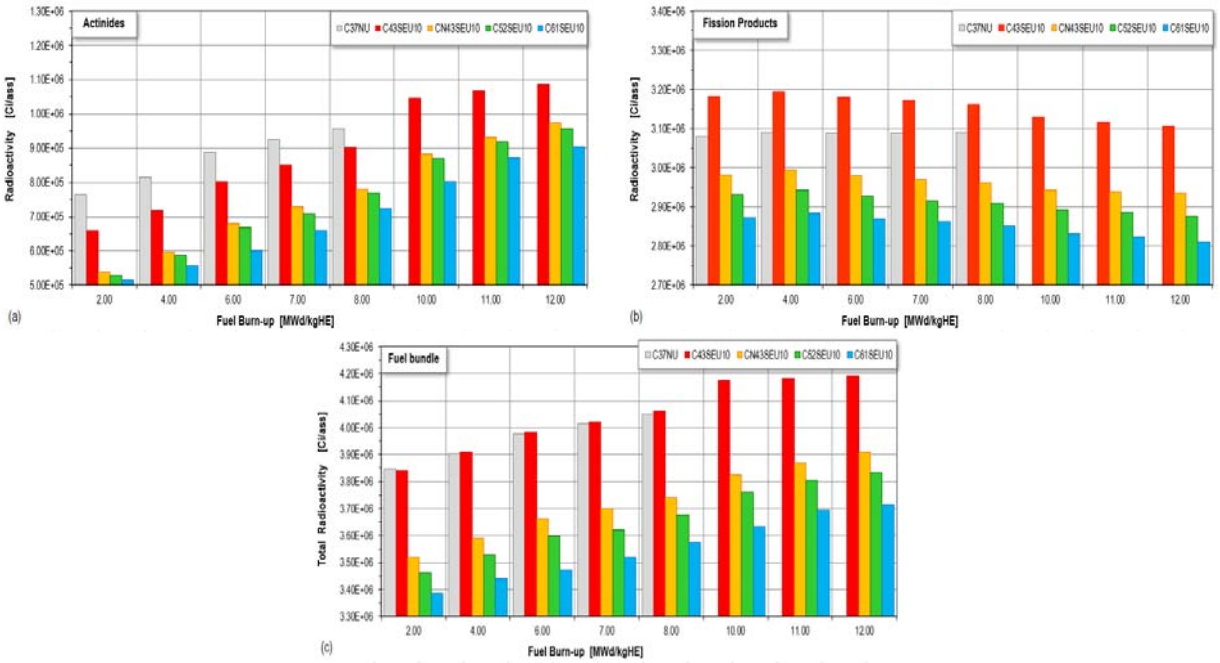


Fig. 8. Spent fuel radioactivity [Ci/assembly] evolution during fuel irradiation for SEU fuel 1.0wt% U^{235} in:
(a) actinides, (b) fission products, and (c) fuel bundle

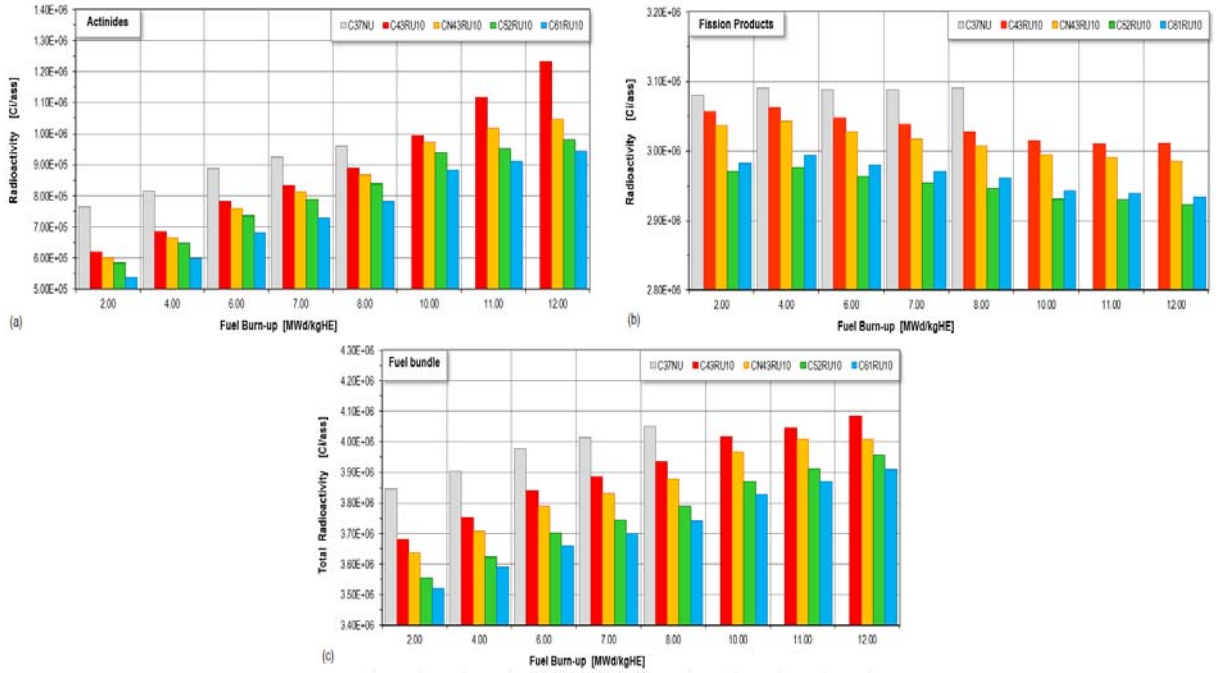


Fig. 9. Spent fuel radioactivity [Ci/assembly] evolution during fuel irradiation for RU fuel 1.0wt% U^{235} in:
(a) actinides, (b) fission products, and (c) fuel bundle

Analysing the total radioactivity evolution with burn-up, it can be observed that the highest values were estimated for C43 fuel bundle closely followed by those obtained for CN43 fuel bundle, with calculated relative differences reaching up to 6.7% (for SEU fuel). The lowest values of the total radioactivity in the spent fuel were estimated for C61 fuel bundle. For using of NU fuel, C37 fuel bundle was characterized by 1.6% higher total radioactivity comparatively to the one estimated for C43 fuel bundle.

The radioactivity estimated in actinides follows the same behaviour as the total radioactivity, with the highest values obtained for C43 fuel bundle, but the values estimated for CN43 fuel bundle are no longer so close, the calculated relative differences reaching up to 15% (for RU fuel). The lowest values of radioactivity in actinides were still those estimated for C61 fuel bundle. As for using NU fuel, the highest radioactivity values remain those estimated for the fuel bundles with 43 fuel elements (C43 and CN43), the relative differences against the values corresponding to the reference fuel bundle (C37) being 4% and 2.8%, respectively.

As it was mentioned earlier, the radioactivity of the fission products reveals a slightly decreasing behaviour during the fuel irradiation. The highest values were estimated for C43 fuel bundle, calculated relative differences reaching up to 9.5% (for SEU fuel). The lowest values of radioactivity in fission products were estimated for C61 fuel bundle, except for the case of using RU fuel when the lowest radioactivity was estimated for C52 fuel bundle (by 0.4% lower than the one obtained for C61 fuel bundle). For using of NU fuel, C37 fuel bundle was characterized by 3.5% higher radioactivity in fission products in comparison with to the one estimated for C43 fuel bundle.

Fig. 10 shows comparatively the total radioactivity estimated in the considered CANDU type fuel bundles (C43, CN43, C52, and C61) at the end of irradiation, for the selected U-based fuels. The value estimated for the reference fuel bundle with NU fuel is also shown for comparison.

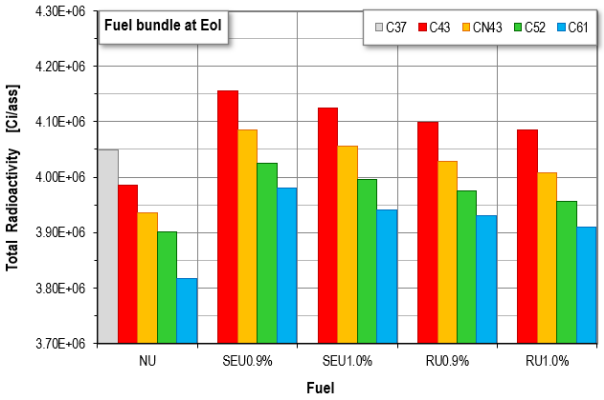


Fig. 10. Spent fuel total radioactivity in CANDU type fuel bundles at EoI [Ci/assembly] for U-based fuels

At the end of irradiation, the highest total radioactivity values were obtained in C43 fuel bundle for all the selected fuel compositions, while the lowest values were those characterizing C61 fuel bundle. It can be observed that for using of NU fuel, the reference fuel bundle, C37, is characterized by the highest total radioactivity value (4.05×10^6 Ci/assembly) in comparison with the other CANDU type fuel bundles with increased number of fuel elements.

Performing a comparison at the level of U-based fuel composition used in the fuel bundles, the highest total radioactivity values in the spent fuel was obtained for SEU fuel 0.9 wt% enriched in U^{235}, while the lowest values were estimated for NU fuel. However, it should mention that the highest total radioactivity (4.15×10^6 Ci/assembly – C43SEU09) is only by 8% greater than the lowest one (3.82×10^6 Ci/assembly – C61NU).

Conclusions

The behaviour to irradiation for advanced CANDU fuel bundles with increased number of fuel elements containing different U-based fuels was studied based on estimation of spent fuel isotopic inventory and radioactivity. The 37-element CANDU standard fuel bundle with NU fuel was considered as reference.

The following U-based fuel compositions have been considered: NU fuel, SEU fuel 0.9 wt% and 1.0 wt% enriched in U^{235}, RU fuel 0.9 wt% and 1.0 wt% enriched in U^{235}. Analyses have been carried out assuming that all fuel bundle elements contain the same U-based fuels.

Fuel irradiation was simulated by using ORIGEN-S burn-up code (SCALE6 software package), at 34.644 kW/kgHE specific burn-up power, as follows: NU fuel - up to 8 MWd/kgHE; SEU and RU fuels - up to 12 MWd/kgHE.

The parameters of interest for the study were: spent fuel inventory (fissile isotopes inventory - U^{235} and Pu^{239}), inventory of interest from proliferation resistance perspective (Pu^{238}, Pu^{240}, Pu^{242}), and radioactivity (total radioactivity and radioactivity of actinides and fission products).

As the fuel irradiation increases, U^{235} consumption is higher than Pu^{239} accumulation leading to a reduction in the fissile inventory. For the CANDU type fuel bundles with increased number of fuel elements, the total fissile inventory reduces at EoI by 48% - 55% (NU fuel) and by 63% - 68% (SEU and RU fuels), respectively. The fissile inventory reduces by 44% for the reference case, C37NU. The highest fissile content was associated to C43NU case, 52.2 g/assembly (28.8 g/assembly U^{235} and 23.4 g/assembly Pu^{239}), this value being by 8.5% lower than the one estimated for the reference case (C37NU).

Performing a comparison between different fuels used in the same fuel bundle, NU fuel brings the highest fissile inventory values, except for the C61 fuel bundle case, the calculated relative differences being as follows: 1.5% (C43SEU) and 3.8% (C43RU); 9.2% (CN43SEU) and 9.8% (CN43RU); 1.3% (C52SEU) and 2.9% (C52RU); -1.6% (C61SEU) and -0.2% (C61RU).

The spent fuel total radioactivity increases during irradiation, mainly due to a significant increase in the contribution of radioactivity in actinide by up to 25%, accompanied by a downward evolution in the contribution of the radioactivity in fission products from 84% to 75%.

The highest total radioactivity values were obtained for the C43 fuel bundle regardless of the U-based fuel used, as follows: $3.98x10^6$ Ci/assembly (NU fuel), $4.15x10^6$ Ci/assembly and $4.12x10^6$ Ci/assembly (SEU fuel 0.9 wt% and 1.0 wt% enriched in U^{235}); $4.10x10^6$ Ci/assembly and $4.08x10^6$ Ci/assembly (RU fuel 0.9 wt% and 1.0 wt% enriched in U^{235}). However, for using of NU fuel, C37 fuel bundle has a total radioactivity higher than the CANDU type fuel bundles with increased number of fuel elements. The lowest total radioactivity values were estimated for the C61 fuel bundle regardless of the U-based fuels used.

The comparison performed between different U-based fuels used in the same fuel bundle, SEU fuel 0.9 wt% enriched in U^{235} led to the highest total radioactivity in the spent fuel was obtained for SEU fuel with 0.9 wt% enrichment in U^{235}, while the lowest values were estimated for NU fuel. However, it should be mentioned that the highest total radioactivity value ($4.15x10^6$ Ci/assembly – C43SEU09) is only by 8% greater than the lowest one ($3.82x10^6$ Ci/assembly – C61NU).

The innovative CANDU type fuel bundles containing U-based fuels can be used in CANDU rectors without requiring additional radiation protection measures than the already implemented ones for the use of the CANDU standard fuel bundles with NU fuel. At the end of irradiation, the fissile inventory estimated for C43, CN43, C52 or C61 fuel bundles does not exceed the one characterizing the C37 fuel bundle with NU. However, the inventory of interest for proliferation resistance estimated for the CANDU fuel bundles with increased number of fuel elements showed an increasing by up to 30% comparatively to the reference case, C37NU.

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