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Shielding and dose-rate calculations of CANDU spent fuelUsing monte-carlo methods

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Abstract: The paper aims to apply the Monte Carlo methods for spent fuel shielding and radiation dose-rate calculations. The spent fuel analysis starts at the moment of its discharge from the reactor, and after a defined period of cooling the radiation dose rates characterizing the spent fuel transport are calculated. Two types of fuel, namely natural uranium and slightly enriched uranium (SEU) fuel, in a CANDU standard fuel bundle with 37 fuel elements have been considered. The fuel burnup was simulated by means of the ORIGEN-S code in order to obtain the radioactive inventory and the photons source characterizing the spent fuel. For the spent fuel transport, a generic stainless steel shipping cask type B was considered; the radiation doses at the cask wall and in air up to 2 m distance from the shipping cask have been calculated by using Monte Carlo MORSE-SGC code. Both ORIGEN-S burnup code and MORSE-SGC shielding code are included in SCALE5.1 programs package. Considering the nuclear safety aspects, the spent fuel is necessary to be kept in temporary wet cooling storage for at least 6 months. The estimated dose rates were low, their values sustaining a safe manipulation of the spent fuel shipping cask; however, the dose rates characterizing SEU fuel were about 30% higher than those obtained for natural Uranium fuel

Keywords: Shielding calculations; Monte Carlo methods; CANDU fuel bundle; Spent fuel; Radiation dose rates

1.Introduction

Shielding calculations represent an essential element of the nuclear safety, considering the difficulties and challenges that may occur during the spent fuel manipulation, transport and storage, both for protection of human health and impact on the environment [1]. The Monte Carlo (MC) particle transport simulation has become a popular method for calculating shielding fusion reactors like in the International Thermonuclear Experimental Reactor (ITER), Chinese Fusion Engineering Testing Reactor /Demonstration Power Plants, and CANDU power reactors. Using MC code for the shielding calculations is challenging due to the complex geometry and heavy shielding of the reactor spent fuel [2,3].

The spent fuel management is targeting some challenges for the nuclear safety such as:

- criticality control and providing the spent fuel cooling; - reduction of the radioactive waste amounts produced during the reactor operation; - providing the efficient protection for the personnel, the people living near the nuclear facilities and the environment, with respect to the international criteria and regulations; reduction of the impact of the radioactive waste management and spent fuel on future generations; avoiding the transfer of responsibility over future generations in terms of control, monitoring, management and storage of the radioactive waste and spent fuel.

The spent fuel discharged from the reactor is characterized by: a high radioactivity due to the fission products disintegration (at 1 m distance in air from a CANDU spent fuel bundle just discharged, the radiation dose reaches 2000 Sv/h); releases heat, and contains plutonium and unconsumed uranium. Considering the nuclear safety aspects, the spent fuel is necessary to be kept in temporary wet cooling storage for at least 6 months. During the cooling period, the spent fuel temperature decreases, and the fission products disintegration lead to reduction of the spent fuel radioactivity at levels allowing its safe manipulation.

The main objective of the paper is to evaluate the radioactive inventory characterizing the spent fuel discharged from the reactor and to assess the radiation dose rates associated to spent fuel transport after a defined cooling period in the reactor pool. The proposed analyses were performed for a CANDU standard fuel bundle containing natural uranium (NU) or slightly enriched uranium (SEU) fuel, irradiated in CANDU reactors specific conditions, discharged from the reactor after reaching "the useful life", and cooled down up to 5 years in intermediate wet storage. The intermediate wet storage is assured by keeping the spent fuel in the reactor's pool, with concrete walls, stainless-steel reinforced, at adequate distances to avoid critical mass formation; as shielding material and cooling agent, the light water is used.

2. Theoretical model set-up

CANDU standard fuel bundle has a cylindrical shape and is composed by 37 fuel elements arranged on 3 concentric rings (of 6, 12 and 18 elements, respectively) and one central element [1], as presented in Figure 1. All the fuel elements in a CANDU standard fuel bundle are identical.

The fuel bundle elements are tubes made of zircaloy, in which the fuel pellets are loaded. The fuel rod, with an external diameter of ~ 13 mm and a length of ~ 50 cm, has a 0.4 mm thick zircaloy clad and can accommodate a column of 31 sintered fuel pellets. As it can be seen in Figure 2, the fuel elements are stiffened in the bundle by two end plates. On the fuel elements positioned on the outer ring, bearing pads are welded to allow the bundle sliding inside the pressure tube and to assure a space between the bundle and the pressure tube for the coolant circulation. The rigidity and spacing of the rods inside the bundle are ensured by the spacers brazed on the inside of the fuel elements [1].

The proposed analyses were performed for two fuels, namely natural uranium fuel and slightly enriched uranium fuel, with 1.1 wt% enrichment in ²³⁵U. NU37 and SEU37 will be further used to identify the two CANDU fuels considered for analysis.



Fig. 1. CANDU standard fuel bundle view [1]



Fig. 2. CANDU standard fuel bundle component parts [1]

In order to perform the shielding calculations, a geometrical model was developed both for the source of radiation and the shipping cask. A single CANDU spent fuel bundle was considered as source of radiation. The fuel bundle was irradiated in CANDU reactors specific conditions, being discharged from the reactor after a residence period of 231 days for NU37 fuel and 278 days for SEU37 fuel, respectively. After the discharge, a cooling period (inside the reactor's spent fuel pool) up to 5 years was considered. The shipping cask used in the paper for the proposed shielding calculations is a cask type B with stainless steel walls, recommended for the transport of fissile materials; the geometrical and material data used in this paper were taken according to the shipping cask prototype designed, manufactured and tested in RATEN ICN Pitesti [4, 5].

The cask model developed for the shielding calculations consists basically of a container region and a source region. The assumption of cask symmetry to its midplane was considered based on the following aspects: (1) symmetry improves the computing efficiency; (2) symmetry is necessary to implement the automated biasing procedure in the Monte Carlo analysis.



Fig. 3. 2-D geometrical configurations for the radiation source (left) and the cask (right) [4, 5]

The source of radiation geometrical model was obtained approximating the fuel bundle by 3 straight, concentrical cylinders (Hf height), separated by thin air gaps: a central cylinder for the center element and the inner ring of 6 fuel elements (R_1 radius), 2nd cylinder for the intermediate ring of 12 fuel elements (R_2 radius), and the 3rd cylinder for the outer ring of 18 fuel elements (R_3 radius). These three cylinders contain a homogenous mixture of fuel, clad and structure materials, mixture named in the following "fuel", with respect to

the volume preservation.

The simplified geometrical model for the shipping cask (R_{ext} radius, H_{ext} height) consists in straight, concentrical cylinders of shielding materials with a central cavity (Rin radius, Hin height) to accommodate the source region, and an air gap (R_g radius, H_g height).

Fig. 3 shows both geometrical models previously described; these models were used and tested in the shielding analyses performed in [4-9].

3. Nuclear fuel burn up simulation and shielding calculations

CANDU reactor specific conditions of irradiation have been simulated, the burnup being performed at the fuels specific burnup powers (34.644 kW/kg HE for NU, and 43.24 kW/kg HE for SEU, respectively), up to 12 MWd/kg HE (the useful life for SEU37). It should be noticed that for NU37 the considered useful life was 8 MWd/kg HE, so the radioactive inventory and photon sources for NU37 were taken for this step of irradiation. The unit MWd/kg HE stands for mega-watt day/kg for heavy element.

The fuel burnup was simulated by means of the burnup code ORIGEN-S, included in the SCALE 5.1 programs package [10] developed by Oak Ridge National Laboratory, and received with single user license under the ORNL-INR Pitesti collaboration agreement. The radioactive inventory of interest for this analysis was the inventory of the fissile isotopes ²³⁵U and ²³⁹Pu, and the inventory of isotopes interesting for neproliferation ²³⁸Pu, ²⁴⁰Pu and ²⁴²Pu, respectively. The photon characterizing the spent sources fuel intermediate storage in the reactor cooling pools were further used as input data for the shielding calculations.

The photon dose rates calculations have been performed by means of Monte Carlo MORSE-SGC shielding code also included in SCALE 5.1 programs package. In the shielding calculations, the (27n-18g) coupled nuclear data library (27 neutron and 18 gamma energy groups) and the ANSI standard flux-to-dose conversion factors (dose rates will be in rem/h) were used. As regarding the Monte Carlo simulation, 1000 bunches of 3000 particles each have been generated. The shielding analysis aims to obtain the radial photon dose rates characterizing the spent fuel after 0.5 year, 1 year and 5 years after its discharge from the reactor, considering the spent fuel transport in a shipping cask type B. The photon dose rates at the cask wall and in air at 0.5 m, 1 m and 2 m distance from the cask were evaluated.

4. Analysis of the Results

In the following, the NU and SEU fuels composition is presented (see Table 1 and **Table 1** – Initial inventory in NU37 and SEU37 fuels, [11]

Table 2). For each fuel, the computation is established first for the components weights in the fuel, then the isotopic weights of 234 U, 235 U, 236 U, 238 U, and O₂, respectively, in the fuel and, finally, the initial inventory of materials (Table 1). The fuel bundle cladding is made of Zircaloy (Zy), containing Zirconium, alloying elements (Tin, Iron, Chromium, Nickel) and impurities (Carbon, Oxygen). Table 2 presents the cladding composition for the CANDU standard fuel bundle.

Fuel project	Enrichme nt in ²³⁵ U [wt%]	²³⁴ U [kg]	²³⁵ U [kg]	²³⁶ U [kg]	²³⁸ U [kg]	O ₂ [kg]	Zy [kg]	UO ₂ [kg]
<i>NU</i> 37	0.71	0	0.138	0	19.357	2.620	2.144	22.115
SEU37	1.10	0.015	0.175	0.064	19.151	2.608	2.144	22.013

Table 3 and Table 4 contain the inventory obtained for NU37 and SEU37 at the End of Irradiation (EoI) regarding the fissile isotopes and the ones of interest for proliferation resistance, respectively.

Table 2 – CANDU standard fuel bundlecladding composition, [11]

Element	Mass [kg]	
Zirconium (Zr)	2.103	
Alloying elements		
Tin (Sn)	3.109×10^{-2}	
Iron (Fe)	3.645×10^{-3}	
Chromium (Cr)	2.144×10^{-3}	
Nickel (Ni)	1.1/9 x 10 ⁻¹	
Impurities		
Carbon (C)	4.074×10^{-4}	
Oxygen (O)	2.573 x 10 ⁻³	

The total fissile content is higher in SEU37 fuel bundle (~ 54 g/assembly) comparatively with the NU37 fuel bundle (~ 51 g/assembly), a relative difference of 6% being obtained. The fissile inventory reduces as the fuel irradiation increases, due to a higher consumption of 235 U than the accumulation of 239 Pu. The relative differences calculated at EoI against beginning of irradiation were as follows: for NU37 - 235 U inventory 70% lower, 239 Pu inventory 34% higher; for SEU37 - 235 U inventory 80% lower, ²³⁹Pu inventory 47% higher. For the total fissile content, the relative differences calculated at EoI against the beginning of irradiation were 50% for NU37 and 66% for SEU37, respectively.

Table 3 – Fissile material inventory for NU37and SEU37 at the EoI

Isotope /	Fissile inventory [g/assembly]		
Fuel	NU37	SEU37	
²³⁵ U	26.26	29.53	
²³⁹ Pu	24.64	24.41	
Total	50.90	53.94	

Table 4 – Inventory of the isotopes ofproliferation resistance interest for NU37 andSEU37 at the EoI

Isotope /	Inventory of isotopes interesting				
	for neproliferation [g/assembly]				
Fuel	NU37	SEU37			
²³⁸ Pu	0.13	0.38			
²⁴⁰ Pu	23.71	27.88			
²⁴¹ Pu	1.60	2.71			
Total	25.44	30.98			

The inventory of 238 Pu, 240 Pu and 241 Pu is higher in SEU37 fuel bundle (~ 31 g/assembly) in comparison with the NU37 fuel bundle (~ 25

g/assembly), a relative difference of 18% being obtained. The total inventory of these isotopes interesting for the resistance to proliferation become significantly higher as the fuel irradiation increases, the relative differences calculated at the EoI against the beginning of irradiation being of 87% for NU37 and 94% for SEU37, respectively.

In order to perform the assessment of photon dose rates characterizing the spent fuel discharged from the reactor and cooled down up to 5 years in the reactor's spent fuel pool, the radiation sources were extracted from the output files produced by the ORIGEN-S's simulation of fuel burnup.

The radiation sources obtained for NU37 and SEU37 spent fuels considering four cooling times (0.5 years, 1 year, 3 years and 5 years, respectively) are represented in Figure 4. The spent fuel radiation emission rates evolution with the cooling time reveals a decreasing trend as the cooling time become longer, so after 5 years of cooling the photon emission rates characterizing the considered fuels are 70% -97% lower comparatively with those obtained after 6 months of cooling. It can be noticed the similar profile for both spent fuels; however, the radiation emission rates characterizing SEU37 spent fuel are about 30% higher than the NU37 spent fuel ones.

For the shielding analysis four photon dose rates measuring points were considered, namely: the shipping cask wall and 0.5 m, 1 m and 2 m (in air) distance from the cask. The minimum (cask wall) and the maximum (2 m distance from the cask) measuring points were thus chosen to check the safety of the spent fuel transport in terms of compliance with the dose rates limits specified in the international agreed regulations for the safe transport of radioactive material [12].

To assess the radiological risk implied by the spent fuel transport using the shipping cask type B, the photon dose rates corresponding to the selected fuel bundle projects, taking into consideration the selecting cooling times after the discharge from the reactor core and the above mentioned measuring points, have been estimated and are presented in Figure 5.

The estimated photon dose rates are lower as the cooling time considered for the spent fuel inside the reactor's pool become longer, and decrease significantly with increasing of the distance from the shipping cask wall.



Fig. 4. Radiation source profile corresponding to NU37 and SEU37 spent fuels



Fig. 5. Dose rates evolution with cooling time and measuring point for NU37 and SEU37 spent fuels

Thus, the photon dose rates obtained to the cask wall after 6 months of cooling were 0.76 mSv/h for NU37 and 1.3 mSv/h for SEU37 spent fuels. After 5 years of cooling the corresponding dose rates to the cask wall were 0.024 mSv/h for NU37 and 0.035 mSv/h for SEU37 spent fuels. These values are drastically reduced as the distance from the cask external surface increases, so at 2 m distance from the cask the photon dose rates become 0.018 mSv/h (after 6 months of cooling) and 0.006 mSv/h (after 5 years of cooling) for NU37 spent fuel, and 0.027 mSv/h (after 6 months of cooling) and 0.008 mSv/h (after 5 years of cooling) for SEU37 spent fuel, respectively.

The radiation dose rates corresponding to SEU37 spent fuel are higher than the NU37 ones, the calculated relative differences being ~ 30%. Estimated photon dose rates for both NU37 and SEU37 spent fuels have safe values being lower than the internationally agreed limits for safe transport of radioactive material using the shipping cask type B. These internationally agreed transport regulations for radioactive material establish that the radiation level under routine conditions of transport shall not exceed 2 mSv/h to the cask wall and 0.1 mSv/h at 2 m from the external surface of the conveyance [12].

5. Conclusion

The present study aimed the application of Monte Carlo methods to perform shielding calculations for CANDU spent fuel. The considered source of radiation was a single CANDU standard fuel bundle with 37 fuel elements. Two fuel compositions were selected for a comparative analysis of the spent fuel, namely natural uranium fuel (NU) and slightly enriched uranium (SEU) with 1.1 wt% enrichment in ²³⁵U. The considered fuels were irradiated according to CANDU reactors specific conditions up to 8 MWd/kg HE for NU fuel, and up to 12 MWd/kg HE for SEU fuel, respectively.

The inventory of the fissile isotopes (²³⁵U and ²³⁹Pu) and the inventory of isotopes interesting for proliferation resistance (²³⁸Pu, ²⁴⁰Pu and ²⁴²Pu) were obtained after performing the simulation of fuel irradiation using ORIGEN-S burnup code. As the fuel irradiation increases, ²³⁵U consumption is higher than ²³⁹Pu accumulation leading to a reduction in the

fissile inventory; at the end of irradiation the total fissile inventory reduces by 50% for NU and 66% for SEU, respectively, in comparison with the initial fissile inventory (²³⁵U only). The total fissile content in SEU37 fuel bundle was 6% higher comparatively with the NU37 fuel bundle one.

After the irradiation, the spent fuel was discharged from the reactor and kept inside the reactor's pool for cooling up to 5 years, in order to allow the fission products disintegration and reduction of the spent fuel radioactivity and heating. The photon dose rates were estimated using MORSE-SGC Monte Carlo code both to the shipping cask wall and in air at different distances from the cask external surface to assure safe transport and manipulation of the spent fuel.

As the cooling time increases, the estimated photon dose rates became lower and were significantly reduced as the distance to the shipping cask wall increases. SEU37 spent fuel is characterized by radiation dose rates ~ 30% higher than those estimated for the NU37 spent However, the photon dose fuel. rates characterizing both considered fuel compositions have values lower than the internationally agreed limits for safe transport of radioactive material using the shipping cask type B.

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