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Actinides input to the dose in the irradiated graphite of RBMK-1500 reactor

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HIGHLIGHTS

- Actinides input to the dose in RBMK-1500 reactor graphite was estimated.
- SCALE 6.1 and MCNPX models were used to calculate actinides specific activities.
- ORIGEN-ARP was used for gamma power, neutron source and effective dose calculation.
- Concentrations of Pu, Am and Cm isotopes in the RBMK graphite sample were measured.
- ^{244}Cm was found to be a critical contributor to effective dose of the personnel.

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ABSTRACT

The purpose of this work is to indicate the actinides input to the total radiation dose caused by the irradiated graphite of the RBMK-1500 reactor in comparison to the dose delivered by other nuclides. We used computer codes (SCALE 6.1 and MCNPX 2.7) to estimate the dose rate delivered by actinides giving special attention to the ^{244}Cm isotope as a critical contributor to the total activity of actinides in the spent graphite for approximately up to 200 years. The concentrations of Pu, Am and Cm isotopes in the graphite sample from the Ignalina Nuclear Power Plant (NPP) Unit 1 have been measured with the inductively coupled plasma mass spectrometer and specific isotope ratios have been compared with alpha spectrometric results as well as with the values simulated by the computer codes. Good agreement of the experimental results and the simulated ratios serves as an additional confirmation of validity of the calculation models. The effective doses rates of inhalation and ingestion for personnel, gamma radiation power, and nuclides, which constitute the neutron source in the irradiated RBMK-1500 graphite constructions, have also been identified. The obtained results are important for decommissioning of the Ignalina NPP and other NPPs with graphite-moderated reactors.

1. Introduction

Graphite is widely used as a moderator, reflector, and fuel matrix in various types of nuclear reactors, such as gas-cooled reactors (e.g. AGR, MAGNOX), RBMK reactors, high temperature gas-cooled reactors (Dragon, Peach Bottom, HTR-10) and is also designed to be used in generation IV reactors (VHTR, MSR). About 250,000 t of spent graphite is already accumulated in still operating or being decommissioned nuclear reactors all over the world (Fachinger et al., 2008). During a long-lasting period of the reactor operation (20–30 years) different radionuclides are present in the irradiated graphite due to activation of impurities in the virgin graphite (usually <0.01% of weight fraction) or due to contamination transfer from the other reactor materials (for instance, nitrogen penetration due to operation of gaseous circuit) (Remeikis et al., 2010; Ancius et al., 2005; Bushuev et al., 2002; Bylkin et al., 2004; Plukienė et al., 2011). For graphite reprocessing, which is important for development of sustainable energetics, the characterization of the most significant radionuclides in the irradiated graphite and the prediction of the radiation dose rate for the personnel handling with the irradiated graphite waste are of great importance.

There are two graphite-moderated, boiling water-cooled RBMK-1500 reactors at the Ignalina site in the north-eastern part of Lithuania. Unit 1 was operated in 1984–2004, and Unit 2 – in

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1987–2009. Graphite is a one of the key elements of the RBMK reactor technology – both units contain about 3600 t of graphite (Remeikis et al., 2010). Thus, strategy of management of radioactive waste originated from the irradiated graphite is a one of the most critical issues. The strategy depends on characteristics of the irradiated graphite (Almenas et al., 1998). Graphite impurities are activated by (n, γ), (n, p), (n, α) reactions partially resulting in long-lived elements (e.g., ^{14}C, ^{36}Cl, transuranium elements). ^{14}C resulting from ^{13}C by (n, γ) and from ^{14}N by (n, p) reactions is a one of the limiting radionuclides for management of the RBMK-1500 reactor graphite itself and low- and intermediate-level radioactive waste (Remeikis et al., 2009; Narkūnas et al, 2010). The measured averaged ^{14}C specific activity in irradiated graphite samples in the graphite stack plateau region of the RBMK-1500 reactor after 11.6 years of operation was found to be 130 ± 20 kBq/g and indicated high burnup (Remeikis et al., 2010). Incineration could be used as an option to manage irradiated graphite, but all ^{14}C would be emitted to the atmosphere, then. Incineration of graphite from just a one RBMK-1500 reactor (1700 t of graphite, $\sim 7 \times 10^{14}$Bq of ^{14}C) would increase the amount of ^{14}C in the atmosphere by 0.6% (Maceika et al., 2005). Irradiated graphite contamination by ^{14}C is an issue in all graphite-moderated reactors (Magnox, UNGG and etc.) as well (Payne et al., 2015; Vende, 2012; Nijst, 2014).

Besides ^{14}C isotope, there is a wide variety of other essential radionuclides contributing to the activity of ionizing and neutron radiation in spent graphite (Ancius et al., 2005; Bushuev et al., 2002). Evaluation of concentrations of light and relatively short-lived radionuclides was performed earlier in Plukienė et al. (2011) and was also addressed in part in Šmaižys et al. (2005), where a different modelling scheme, i.e. a deterministic approach, was used. It is important to point out that long-term power operation results also in build-up of long-lived isotopes of radioactive actinides, such as Pu, Am, Cm, etc., caused by activation of U and Th impurities in the virgin nuclear graphite. The recent study on the actinides in the irradiated graphite of a RBMK-1500 reactor (Plukienė et al., 2014) has revealed that the virgin RBMK-1500 graphite contains seven times lower concentration of actinides, than it have been assumed previously (Ancius et al., 2005). Specific activities of the actinides in the irradiated RBMK-1500 graphite constructions were obtained using alpha spectrometry results of the ratios of alpha particles emitting isotopes, such as $^{238}Pu/(^{239}Pu + ^{240}Pu)$ and $^{243}Cm + ^{244}Cm/^{241}Am$, together with modelling results (MCNPX 2.7 and SCALE 6.1). Satisfactory results were obtained using both models for all radionuclides, except ^{241}Am and ^{242}Cm, which are very sensitive to the neutron energy spectra, the operational power and nuclear data library.

This paper is the continuation of the previous one (Plukienė et al., 2014). It was initiated to improve the radiological characterization of Pu and minor actinides such as Cm and Am in the RBMK-1500 graphite, and to determine an input of the actinides to the dose rate from the irradiated graphite of the RBMK-1500 reactor. Special attention is paid to show the practical value and applicability of the obtained results. For this purpose, the explicit inductively coupled plasma mass spectrometry (ICP–MS) measurement of the alpha spectrometric Pu and Am-Cm samples has been performed, the concentrations of Pu, Am, and Cm isotopes have been determined, and some limitations of applicability of the ICP–MS method discussed. Analysis of the obtained concentrations and of the model for evolution of Pu, Am and Cm isotopes in the irradiated graphite provided additional confirmation of the model for both calculation approaches. The earlier obtained specific activities of light and relatively short-lived nuclides (Plukienė et al., 2014) and experimentally confirmed specific activities of transuranium nuclides have been used for estimation of the personnel effective dose due to inhalation and ingestion of graphite particles as well as for evaluation of the gamma radiation power and the neutron source from the RBMK-1500 graphite stack.

2. Modelling

Numerical modelling is an important tool for estimation of the neutron fluence in the graphite and the radioactive waste generation in the nuclear reactors (Ancius et al., 2005; Plukienė and Ridikas, 2003; Plukienė et al., 2005; Lukauskas et al., 2006). For estimation of the neutron fluence and the amount of radionuclides in the irradiated graphite from the RBMK-1500 core, two different codes were used: MCNPX (v2.7 which includes CINDER burnup capability) and the SCALE 6.1 codes package (SCALE, 2009; Pelowitz, 2011; Wilson et al., 1999). The modelling procedure and reactor modelling details are explicitly described in our recent publication (Plukienė et al., 2014). In the previous study it was pointed out why a less sophisticated and less time-consuming computational 3D SCALE 6.1 model can be used for radiological characterization of graphite in the reactor plateau region using available standard nuclear data library ENDF/B-VII (238 energy group library).

Knowledge of the detailed radionuclide content in the radioactive waste establishes a background for assessment of the radiotoxicity and a particular dose for the personnel due to handling of the irradiated graphite waste. For radiotoxicity evaluation, there are different approaches with different intentions: the IAEA Clearance Potential Index Concept (IAEA, 2004), and the USA Radioactivity Concentration Guides (RCG) created by the International Commission on Radiological Protection (ICRP) (USCFR, 1991; ICRP, 2012).

The ICRP provides recommendations and guidance on protection against the risks associated with ionizing radiation. This approach deals with clearance levels of the isotopes and is dedicated to the radioactive waste management in case of decision making about final destination of radioactive waste (long-term storage, recycling, landfill or near surface disposal). It was used in (Plukienė et al., 2014) for determining radiological important nuclides for disposal of RBMK-1500 reactor graphite waste. The second approach (usually used in the USA) provides the RCG for continuous ingestion (from water) and inhalation (from air) in unrestricted areas, in units of Ci/m^3 (in terms of ingestion hazard factor in m^3 of water, respectively, inhalation hazard factor in m^3 of air). This approach that is implemented in the deterministic code ORIGEN-ARP is suitable for evaluation of an immediate risk for the personnel handling with the irradiated graphite waste in case of graphite reprocessing before disposal. This code was successfully used for estimation of CANDU radiotoxic inventory (Pavelescu and Gepraga, 2007). ORIGEN-ARP is an automatic rapid processing code for calculation of spent fuel depletion, decay, and source term analysis. In ORIGEN-ARP analysis scheme it is possible to use time-dependent material concentrations from ORIGEN-S (SCALE 6.1 sequence), either the input of isotopic compositions of specific activated materials (in our case—the RBMK-1500 reactor graphite) and subsequently to determine decay heat generation and radiation source terms, nuclide concentrations and activities, radionuclide toxicity and gamma radiation power, etc. The radiotoxicity characterizes alpha emitters and the gamma radiation power is directly proportional to the gamma exposure dose from gamma emitters. The most hazardous radionuclides have been identified using deterministic code ORIGEN–ARP (Gauld et al., 2009 January).

Generally, it is internationally agreed that radiotoxicity of radionuclides is expressed by the effective dose (in Sv). Thus, an effective dose for personnel from inhalation or ingestion of the irradiated graphite particles was afterwards recalculated using

committed effective dose coefficients for each radiotoxic nuclide taken from the ICRP publication No. 119 (ICRP, 2012) as followed:

$$D_{\mathrm{Inh}} = A \times E_{\mathrm{Inh}} \quad \text{and} \quad D_{\mathrm{Ing}} = A \times E_{\mathrm{Ing}}, \tag{1}$$

where A is the specific activity (Bq/kg), E_{Inh} are the radionuclide activity unit effective dose coefficients (Sv/Bq) for a reference worker in case of inhalation of 1 kg graphite dust, and E_{Ing} are the radionuclide activity unit effective dose coefficients (Sv/Bq) for a reference worker in case of ingestion of 1 kg graphite dust.

These effective dose coefficients were calculated using biokinetic models of the human respiratory and gastrointestinal tracts. These biokinetic models describe radionuclides absorption into a blood circulation system by their specific distribution, tissue retention, and excretion processes (ICRP, 2012). The calculations were applied for a reference worker with an average breathing rate of 1.2 m^3/h during an 8 h working day (daily air intake of 9.6 m^3) (ICRP, 2012). The radionuclide absorption rates into the blood circulation system were assessed considering graphite dust aerosol particles of 1 μm activity median aerodynamic diameter (AMAD). This approach was more conservative than considering aerosol particles of 5 μm AMAD. Lastly, input of the actinides to the cumulative effective dose was estimated.

3. Experimental

Concentrations of Pu, Am, and Cm isotopes in the irradiated graphite alpha spectrometric sample from the Ignalina NPP Unit 1 have been measured using alpha spectrometry and high resolution ICP–MS techniques (Plukienė et al., 2014; Lukšienė et al., 2014, 2015; Puzas et al., 2015; Remeikis et al., 2005; Ežerinskis et al., 2016; Gvozdaitė et al., 2010; Bu et al., 2013; Cizdziel et al., 2008). The sample history is reported in (Plukienė et al., 2014).

Briefly, the sample preparation for alpha spectrometry measurements comprised the graphite sample digestion using mineral acids (H_2SO_4, $HClO_4$, HNO_3), the Pu and Am-Cm isotopes preconcentration by scavenging with $Fe(OH)_2$ and $Fe(OH)_3$ and actinides separation using the UTEVA and TRU Resins (Puzas et al., 2015; Remeikis et al., 2005; Chen et al., 2001; Horwitz et al., 1993). To improve the precision and accuracy ^{242}Pu and ^{243}Am tracers were used for Pu and Am-Cm samples, respectively, to monitor chemical recoveries used for reconstruction of the real results. Pu and Am-Cm isotopes were deposited on the stainless steel disks separately. The activities of samples were measured using the OCTETE Plus-10600 spectrometer (reported in Plukienė et al., 2014). For the Pu, Am and Cm isotopic composition determination by ICP–MS the Pu and Am–Cm stainless steel discs were treated with 8 mol/dm^3 HNO_3 and heated at 50 °C for 45 min. After this chemical treatment both discs were measured by alpha spectrometry. The Pu and Am–Cm washing out efficiency from discs was evaluated according to the remained amount of ^{242}Pu and ^{243}Am tracers (4856 keV and 5276 keV alpha peaks, respectively). It was determined that the removal efficiency from discs varied in the range of 98–99%.

After the removal from stainless discs aliquots of diluted solutions were analysed with a sector field ICP–MS (Element XR; Thermo-Fisher Scientific, USA) (Gvozdaitė et al., 2010; Bu et al., 2013). Sample solutions were introduced using a PFA nebulizer inserted into an Apex Q desolvation system (ESI, USA) to boost Pu signal and reduce uranium hydride formation in ICP–MS measurements. For Am-Cm measurements, a Spiro attachment (also from ESI) was used to reduce oxide levels to <0.2%. Data acquisition consisted of 40 runs (single pass) with a fifty points per peak using a 20% mass window. Integration time was set to 10 ms for ^{238}U and 100 ms for the other isotopes. Before the samples were analysed, the system was optimized for sensitivity, stability, and oxide levels using a natural uranium solution. The instrument detection limit was previously determined using a low-level ^{239}Pu standard and was estimated to be at ~0.3 mBq/g (~0.130 fg/g) based on a 5 g sample. A correction factor for uranium hydride (~0.00003) was determined and applied to all measured ^{239}Pu signals as described elsewhere (Bu et al., 2013; Cizdziel et al., 2008). The mass discrimination effect was corrected for using a factor (~4 ‰) determined by comparing the measured ^{238}U/^{235}U ratio in a natural uranium solution to the true value (137.88). The mass spectrometry results have been compared with the results obtained by alpha spectrometry. The resolution of alpha peaks was 24 keV (FWHM) at 4–6 MeV.

Isotopes of 239–248 Da masses were assessed by the ICP–MS. A low resolution ($m/\Delta m = 300$) was used to increase the isotopic signal sensitivity, but spectral interferences appeared. $^{208}Pb^{40}Ar^+$ and $^{206}Pb^{40}Ar^+$ polyatomic ions significantly interfered with 246 Da and 248 Da masses as Pb concentration in the Am-Cm sample was found to be relatively high (in the ppb range). Because ^{242}Pu and ^{243}Am isotopes were added as tracers during the actinide separation procedure (they were used in Pu and Am-Cm alpha samples preparation), thus, 242 and 243 Da masses isotopic information was excluded in both samples, too. To sum up, reliable information of 239–241, 244–245 Da masses was collected and ^{239}Pu, ^{240}Pu, ^{241}Pu, ^{241}Am, ^{244}Cm, ^{245}Cm isotope concentrations were determined.

4. Results

Pu, Am, and Cm isotopes generation in the graphite stack and sleeves in the reactor plateau region was modelled with the MCNPX 2.7 and SCALE 6.1 code packages and was evaluated according to the known Unit 1 reactor power history during its whole operation period, namely 21 years (Plukienė et al., 2014). The modelled and experimentally determined Pu, Am, and Cm isotopic compositions in the RBMK-1500 reactor irradiated graphite sleeve are presented in Table 1.

As concluded in (Plukienė et al., 2014), satisfactory results were obtained applying both models for all Pu isotopes (the difference was in the 1–3% range). However, for Am and Cm isotopes differences were larger (6–40%), probably due to the significant sensitivity of minor actinides to the neutron energy spectra, the operational power, and nuclear data library used for calculation. The higher specific activity values (as well as concentration) were obtained by SCALE 6.1, partly due to intermediate actinide cross section recalculation (a prediction/correction step) at the given operational power and a slight simplification of a geometric model, i.e. SCALE 6.1 approach is more conservative. The greatest difference was determined in the ^{242m}Am calculation results due to the missed build-up contribution through the internal conversion channel generation of ^{242m}Am because of incorrect communication between CINDER data library (MCNPX 2.7 coupled with CINDER) (Plukienė et al., 2005), so, only SCALE 6.1 ^{242m}Am values were taken into account. However, comparing the simulated results with the experimental values, one should note that taking into account minor concentrations (on order of 10^{-12} g/g and less) both simulated values are within the uncertainty range of the experimentally determined concentrations (see Table 1 for details).

The Pu, Am, and Cm concentrations measured by ICP–MS were compared with their neutron activation calculation in the graphite using MCNPX 2.7 and SCALE 6.1 after an appropriate decay period (6 and 11 years after irradiation, respectively). In order to check the consistency of the results with Pu and Am–Cm alpha measurements and verify our calculations by both codes, the modelling results at the alpha spectrometry measurement time were compared for both decay dates. Alpha spectrometry results of ^{239}Pu, ^{240}Pu, ^{244}Cm and ^{245}Cm concentrations were recalculated from the sums of $^{239+240}$Pu and $^{244+245}$Cm activity

Table 1
Concentration of Pu, Am, and Cm isotopes in the RBMK-1500 reactor graphite sleeve after the irradiation at different decay times calculated using MCNPX 2.7, SCALE 6.1 computational codes and determined experimentally by spectrometric methods (alpha spectrometry – in 2009 and ICP–MS –in 2014).

	Concentration $\times 10^{-12}$ (g/g)							
Year	2003		2009			2014		
Nuclide	SCALE	MCNP	SCALE	MCNP	Alpha (Plukienė et al., 2014)	SCALE	MCNP	ICP–MS (2σ)
^{238}Pu	0.6	0.6	0.7	0.7	0.7 ± 0.1	0.7	0.7	–
^{239}Pu	8.9	8.6	9.0	8.6	8.6*	9.0	8.6	7.1 ± 2.1
^{240}Pu	5.9	5.8	6.6	6.3	6.0*	7.1	6.7	5.0 ± 2.7
^{241}Pu	3.5	3.6	2.6	2.7	–	2.1	2.1	2.0 ± 1.5
^{242}Pu	9.7	9.8	9.7	9.8	Tracer	9.7	9.8	Tracer
^{241}Am	0.31	0.26	1.2	1.2	0.9 ± 0.4	1.8	1.7	1.3 ± 0.4
^{242m}Am	3.3×10^{-3}	1.4×10^{-3}	3.2×10^{-3}	–	–	3.1×10^{-3}	–	–
^{243}Am	4.4	3.4	4.4	3.4	Tracer	4.4	3.4	Tracer
^{242}Cm	9.5×10^{-2}	7.4×10^{-2}	1.7×10^{-5}	1.0×10^{-5}	$(7 \pm 2) \times 10^{-4}$	8.1×10^{-6}	3.5×10^{-6}	–
^{243}Cm	2.5×10^{-3}	2.7×10^{-3}	2.2×10^{-3}	2.4×10^{-3}	$(3.2 \pm 0.4) \times 10^{-3}$	2.0×10^{-3}	2.1×10^{-3}	–
^{244}Cm	3.5	2.7	2.8	2.2	2.9*	2.3	1.8	2.4 ± 0.5
^{245}Cm	0.15	0.12	0.15	0.12	0.2*	0.15	0.12	0.2 ± 0.1
^{246}Cm	0.11	7.9×10^{-2}	0.11	0.08	–	0.11	0.08	–
^{247}Cm	2.0×10^{-3}	1.4×10^{-3}	2.0×10^{-3}	1.4×10^{-3}	–	2.0×10^{-3}	1.4×10^{-3}	–
^{248}Cm	4.2×10^{-4}	–	4.2×10^{-4}	–	–	4.2×10^{-4}	–	–
^{244}Cm/^{245}Cm	23.7	22.7	18.8	18.1	16.6*	15.5	14.9	13.7
^{239}Pu/^{240}Pu	1.5	1.5	1.4	1.4	1.4*	4.3	4.1	3.6

* ^{239}Pu and ^{240}Pu concentrations were recalculated from the sum of $^{239+240}$Pu activity measurements according to the mass-spectrometric ratios obtained for $^{239/240}$Pu. ^{244}Cm and ^{245}Cm concentrations were recalculated from the sum of $^{244+245}$Cm activity measurements according to the mass-spectrometric ratios obtained for $^{244/245}$Cm.

measurements, respectively, according to the mass-spectrometric ratios obtained for ^{239}Pu/^{240}Pu and ^{244}Cm/^{245}Cm. Good agreement of experimental results, taking into account experimental uncertainties between both spectrometry measurements (alpha spectrometry and ICP–MS), with modelling results of the graphite sleeve activation after 19 years of irradiation in the RBMK-1500 reactor core (the sample was taken in 2003) has been found for $^{239–241}$Pu, ^{244}Cm and ^{245}Cm isotopes (see Table 1 for details). Slightly lower concentration of the ^{241}Am isotope was determined by ICP–MS. The analysis of obtained concentrations and evolution of Pu, Am and Cm isotopes in the irradiated graphite provided an additional confirmation of validity of the simplified model of the RBMK-1500 reactor (core fragment of 14 fuel assemblies and 2 control protection system rods) used in both calculation approaches.

An assessment of the radionuclide toxicity, the gamma radiation power, and the neutron source intensity during a long period of time taking into account the radionuclide decay, was performed with the deterministic code ORIGN-ARP (Gauld et al., 2009) by using experimentally confirmed activated nuclide concentrations in the graphite stack calculated with SCALE 6.1 as it was already reported above. Afterwards, the radionuclide effective doses (in Sv) were recalculated from the obtained specific activities values (Bq/kg) for the personnel considering 1 μm AMAD graphite dust particle inhalation and ingestion referring to the ICRP publication No. 119 (ICRP, 2012) (see formula (1)).

The effective dose of inhalation coming from light elements and actinides of the irradiated graphite stack considering the inhalation of 1 kg graphite dust aerosol particles of 1 μm AMAD is presented in Fig. 1. After 1 year of decay the total effective dose of inhalation is of magnitude of 1.3 Sv, the actinide input is 0.5 Sv. ^{244}Cm has been found to be the most hazardous nuclide in the irradiated graphite. Its effective dose of inhalation after 1 year of decay is 0.44 Sv, after 30 years it goes down to 0.15 Sv. Another highly toxic nuclide is ^{60}Co. ^{244}Cm together with the second highly toxic nuclide ^{60}Co (0.35 Sv after 1 year of decay) constitute the 62% of the total effective dose of inhalation in the spent nuclear graphite. After 10 years of decay ^{244}Cm contributes 47% and ^{60}Co–8% of the total effective inhalation dose. ^{244}Cm dominates over the other actinides up to 200 years, afterwards ^{238}Pu and ^{241}Am contribution to the total effective inhalation dose is significant giving up to 0.01 Sv. ^{14}C, ^{36}Cl, ^{60}Co, ^{63}Ni, and ^{154,155}Eu (the computational modelling reveals dominating ^{154,155}Eu neutron activation origin because of the low U concentration in the RBMK-1500 graphite, Puzas et al., 2010) are the main light isotopes that contribute to the total effective dose of inhalation in the spent nuclear graphite as it is shown in Fig. 1.

The effective dose for the personnel from ingested graphite particles (also of diameter of 1 μm) that is caused by the nuclide activation in the irradiated graphite stack as a function of decay time is presented in Fig. 2. The total effective dose of ingestion is 0.15 Sv; the actinide input is 0.002 Sv after 1 year of decay. The comparison of the effective doses of inhalation and ingestion, which are presented on the same scale in Figs. 1 and 2, shows a difference of about one order of magnitude in the total effective dose and a difference of 2.5 orders of magnitude in the total effective dose delivered by actinides. It is obvious that the inhalation pathway of the radionuclide intake is much more dangerous than the ingestion during the nuclear graphite waste handling procedures (considering graphite dust particles of 1 μm AMAD).

The gamma radiation power delivered by the light isotopes and actinides as a function of decay time in the graphite stack was modelled using Origen-ARP and is presented in Fig. 3. The total

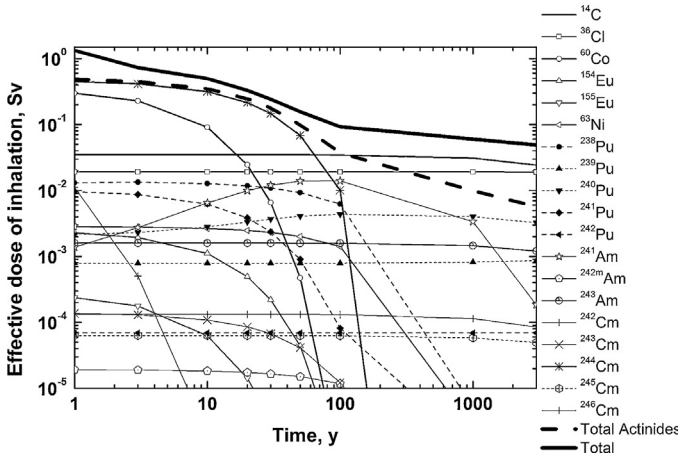

Fig. 1. The effective dose for personnel from inhaled graphite dust particles (of 1 μm AMAD) due to activation of the light elements and actinides in the irradiated graphite stack as a function of decay time.

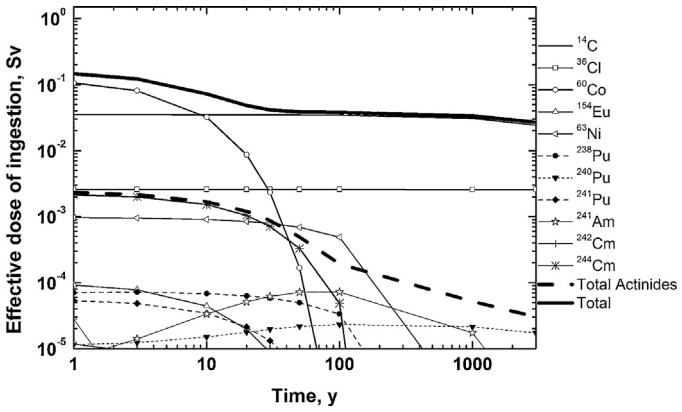

Fig. 2. The effective dose for personnel from ingested graphite dust particles (of 1 μm AMAD) due to activation of the light elements and actinides in the irradiated graphite stack as a function of decay time.

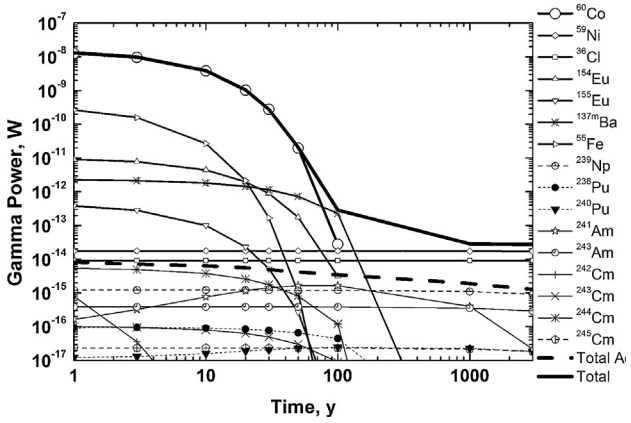

Fig. 3. The gamma radiation power (in W) caused by the light radionuclides and actinides in the irradiated graphite stack as a function of decay time.

gamma radiation power is determined by ^{60}Co (95% of total) for up to 50 years. The other gamma irradiating nuclides are neutron activation products: ^{55}Fe, ^{154}Eu, and ^{155}Eu, the influence of which is minor compared with ^{60}Co. The fission product ^{137m}Ba together with long-lived ^{36}Cl and ^{59}Ni will dominate after 50 years, while the most important actinides are ^{239}Np, ^{241}Am, and ^{243}Am.

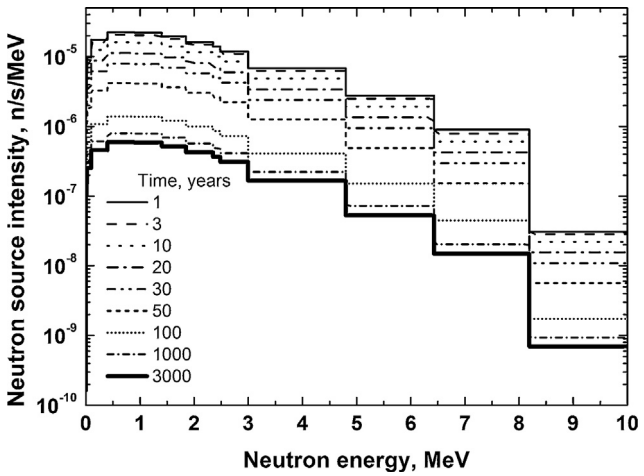

Fig. 4. The total neutron energy spectrum in the irradiated graphite stack at different decay times.

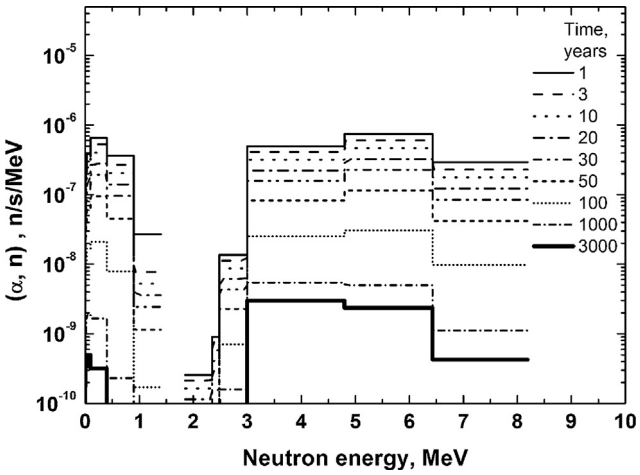

Fig. 5. The neutron spectrum from (α,n) reaction in the irradiated graphite stack at different decay times.

The total neutron source intensity in the irradiated graphite stack of the plateau region at different decay times is presented in Fig. 4. One can observe that the neutron source is mostly determined by the spontaneous fission neutrons from ^{244}Cm decay (91.93%), while ^{246}Cm (7.99%), ^{242}Pu (0.05%), and ^{240}Pu (0.03%) have a minor influence. The intensity of neutron source decreases by two orders of magnitude after the ^{244}Cm total decay. The neutrons from (α, n) reaction have a minor influence on the total neutron spectrum as it is observed from comparison of Fig. 4 with Fig. 5. The ^{13}C(α,n)^{12}C reaction is the main neutron generation reaction, while ^{244}Cm is the main alpha source (responsible for the 95% of the occurred reactions).

5. Conclusions

For assessment of radiation hazard to the personnel due to handling the graphite waste after the shutdown of the graphite-moderated RBMK-1500 reactor at the Ignalina NPP, simulation by computer codes (SCALE 6.1 and MCNPX 2.7) was used. The simulation was based on the new ICP–MS measurements of concentrations of Pu, Am, and Cm isotopes as well as earlier obtained concentrations of light and relatively short-lived isotopes in the graphite sample from the Ignalina NPP. Good agreement of experimental results and simulated ratios serves as additional confirmation of validity of both RBMK-1500 reactor computational models created with SCALE 6.1 and MCNPX 2.7 code packages.

The effective doses of inhalation and ingestion for the personnel, the gamma radiation power, and the neutron source intensity in the irradiated RBMK-1500 graphite stack have been calculated using a deterministic code ORIGN-ARP (from the SCALE 6.1 code package). The earlier obtained specific activities of light and relatively short-lived isotopes (Plukienė et al., 2014) and the experimentally confirmed specific activities of transuranium elements (Puzas et al., 2010) were used as an input to the ORIGEN-ARP calculation.

The actinide input to the dose in the irradiated graphite of the Ignalina NPP RBMK-1500 reactor was calculated and compared to the input of the other nuclides. It has been assessed that after 1 year of decay the effective dose for the personnel due to the inhalation of 1 kg of 1 μm AMAD graphite dust aerosol particles from the irradiated RBMK-1500 reactor Unit 1 graphite stack is of magnitude of 1.3 Sv. The actinide input is 0.5 Sv in case of inhalation. Due the case of ingestion the effective dose goes down to 0.15 Sv (actinides input is 0.002 Sv). After 10 years of decay ^{244}Cm is responsible for 47% and ^{60}Co 8% of the total effective

inhalation dose rate. ^{244}Cm dominates over the other actinides for up to 200 years, afterwards the contribution of ^{238}Pu and ^{241}Am appears. ^{14}C, ^{60}Co, ^{36}Cl, ^{63}Ni, and ^{154,155}Eu are the main light isotopes that contribute to the effective dose of inhalation.

The total gamma radiation power is mainly determined by ^{60}Co (95% of total) during the period of up to 50 years. The fission product ^{137m}Ba together with long-lived ^{36}Cl and ^{59}Ni will dominate after 50 years, while ^{239}Np, ^{241}Am, and ^{243}Am create the biggest actinide input to the total gamma radiation power.

The total neutron source intensity in the irradiated graphite stack is mostly determined by the spontaneous fission neutrons from the ^{244}Cm decay (91.93%) with the minor influence of ^{246}Cm (7.99%), ^{242}Pu (0.05%), and ^{240}Pu (0.03%). It decreases by two orders of magnitude at the time after the ^{244}Cm total decay. The neutrons from (α,n) reaction have a minor influence to the total neutron spectrum and the main neutron generation reaction is $^{13}C(\alpha,n)^{12}C$ (with ^{244}Cm as the main alpha source).

The analysis of effective doses of inhalation and ingestion for the personnel, the gamma radiation power, and the neutron source intensity shows that for the safety of the personnel that will handle nuclear waste during the RBMK-1500 reactor dismantlement the ^{244}Cm is a critical isotope in the spent graphite for approximately up to 200 years.

The obtained results can be used for optimization of individual effective radiation doses for the personnel dismantling the RBMK-1500 reactors and in any further decommissioning stages at the Ignalina NPP. The findings on relative importance of various radionuclides influence for the personnel effective dose in a different time scale after the shutdown of the reactor as well as the order of effective dose magnitude can be used as indications for planning nuclear waste characterization activities and graphite waste management strategies at other NPPs with graphite-moderated reactors.

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