THE INSTITUTE OF PHYSICS
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EVOLUTION OF COMPOSITION OF RADIONUCLIDES IN THE GRAPHITE OF THE RBMK-1500 REACTOR

Summary of the Doctor's Thesis

Physical sciences, Physics 02P

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FIZIKOS INSTITUTAS
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RBMK-1500 REAKTORIAUS GRAFITO RADIONUKLIDINĖS SUDĖTIES EVOLIUCIJA

Daktaro disertacijos santrauka

Fiziniai mokslai, fizika 02P

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INTRODUCTION

Two RBMK-1500 reactors of Ignalina nuclear power plant (NPP) are entering the final stage of their service life. The Unit 1 was already shut down in 2004. The shut down of the Unit 2 is foreseen in the year 2009. The decommissioning and dismantling activities of the reactors will involve handling and disposing of a big amount of operational waste and decommissioning residues. In particular, the graphite used as moderator and reflector in both reactors representing some 3800 tones will contribute largely to the total volume of radioactive waste. Nuclear safety and environmental standards require that the radioactive waste management strategies are based on minimization of amounts of radioactive waste. Although only limited practical experience of decommissioning of graphite reactors is available today, modern radioactive waste processing techniques and recent research on activated graphite treatment offer a number of interesting possibilities. The options of graphite treatment vary from direct disposal of conditioned graphite blocks to separation of radioactive impurities from bulk carbon. The selection of relevant option focused on real radiotoxicity of graphite may result in considerable savings of decommissioning funds. Therefore the determination of concentrations of radioactive impurities in activated graphite has a primary importance for its management strategy. However, representative measurement campaign first of all may not be performed earlier than a certain period after the shut down of reactor due to limited accessibility and dose rates. Secondly, experimental measurements are costly and do not always permit to measure all radionuclides. This leads to new methodologies of radiological characterization of radioactive waste based on easy-measured key radionuclides and establishing of radionuclide vectors calculated by various computer codes.

The principal aim of the Doctor's Thesis

The principal aim of this Thesis is the creation of the model of formation of radioactive impurities in the graphite of Ignalina NPP, evaluation of radiological characteristics of graphite and formation of scientific grounds for the selection of appropriate option for its further management.

The principal tasks

The principal aim of this Thesis is achieved through following tasks:

1. Creation of the model for evaluation of activation of various graphite structures of the RBMK-1500 reactor.
2. Identification of the main factors determining the balance of activation radionuclides.
3. Investigation of composition of impurities of fresh sample of the Ignalina NPP graphite.

4. Estimation of specific activities of radionuclides based on experimental results of investigation of composition of impurities of fresh sample of the Ignalina NPP graphite.
5. Evaluation of radiotoxicity of RBMK-1500 graphite in various locations of moderator and reflector, prediction of evolution of the radiotoxicity due to the long-term storage of graphite.

The scientific novelty

The axial and radial non-homogeneity of concentrations of radioactive impurities in graphite moderator and reflectors of RBMK-1500 reactor have been modelled for the first time. The model takes into account both the density of the flux of neutrons and their spectral variations in graphite. The new data on radiotoxicity of graphite are presented in this Thesis. The impurities affecting the classification of radioactive waste resulting from the processing of spent graphite are identified

Structure and the scope of the Thesis

The Thesis consists of the following parts: introduction, the list of scientific publications and contributions at the conferences, review of scientific literature, numerical methods of calculation, the scheme of calculation and methodology of impurity investigation, results, conclusions and references. The scope of the Thesis is: 110 pages, 27 figures, 16 tables and 117 references.

Publications on the subject of the Thesis and approbation

Articles

A 1. Maceika E., Remeikis V., Ancius D., Ridikas D., Evaluation of the Radiological consequences of ^{14}C due to technogenic graphite incineration, Lithuanian Journal of Physics, Vol 5, 2005.

A 2. Cometto M., Ridikas D., Aubert M.C., Damoy F., Ancius D. Activation analysis of concrete and graphite in the experimental reactor RUS, Radiation Protection Dosimetry, Vol 115, No 1-4, 2005, pp. 104-109.

A 3. Ancius D., Ridikas D., Remeikis V., Plukis A., Plukienė R., Cometto M. Radiological characteristics of the irradiated graphite from RBMK-1500 reactor, Environmental and Chemical Physics, p.p. 140-147, vol 26, No 4, 2005.

A 4. Ancius D., Ridikas D., Remeikis V., Plukis A., Plukienė R., Cometto M. Evaluation of the activity of irradiated graphite in the Ignalina Nuclear Power Plant RBMK-1500 reactor, Nukleonika, p.p. 113-120, vol 50, No 3, 2004. http://www.ichtj.waw.pl/ichtj/nukleon/back/full/vol50_2005/v50n3p113f.pdf

A 5. Cometto M., Ancius D., Ridikas D. Testing of Different Data Libraries in Activation Analysis of Graphite and Concrete from Nuclear Installations. Proceedings of Workshop on Nuclear Data for the Transmutation of Nuclear Waste, GSI-Darmstadt, Germany, 2-5 September, 2003. http://www-wnt.gsi.de/tramu/proceedings/cometto.pdf

Approbation of the results:

B 1. Cometto M., Ridikas D., Aubert M. C., Damoy F., Ancius D. Activation analysis of concrete and graphite in the experimental reactor RUS, International Conference "21st Century Challenges in Radiation Protection and Shielding" ICRS10/RPS2004, Madeira, Portugal, 9-14 May 2004.

B 2. D. Ancius, M. Cometto**,** D. Ridikas. Estimation d'activation du graphite dans le cœur du réacteur RBMK par une méthode MC, Journées Jeunes Chercheurs, Aussois, France, 1-5 Décembre, 2003.

B 3. D. Ancius, M. Cometto, D. Ridikas Activation Analysis of Graphite for Ignalina RBMK-1500 power reactor and Strasbourg research reactor (RUS), International Conference "Decommissioning Challenges: an Industrial Reality", Avignon, France, 23-25 November, 2003.

B 4. Cometto M., Ancius D., Ridikas D. Testing of Different Data Libraries in Activation Analysis of Graphite and Concrete from Nuclear Installations. Workshop on Nuclear Data for the Transmutation of Nuclear Waste, GSI-Darmstadt, Germany, 2-5 September, 2003.

Chapter 1. REVIEW OF SCIENTIFIC LITERATURE

The graphite reactors are often associated with the beginning of the atomic energy era. A number of countries being the pioneers of atomic energy like France, Great Britain and Russia started their nuclear programmes with this branch of reactors. The nuclear safety standards and especially the safety issues concerning the decommissioning and management of dismantled reactor residues were not a priority at that time. This caused a major problem of their decommissioning and some 50 power reactors worldwide using graphite are in permanent shutdown status waiting for their decommissioning and graphite management strategies.

The most problematic issues of used graphite waste management are its considerable mass and the presence of a long lived ^{14}C isotope due to the following nuclear reactions: $^{13}C(n,\gamma)$ ^{14}C and $^{14}N(n,p)$ ^{14}C. ^{13}C is present as a natural carbon isotope in graphite (~1.11%) and ^{14}N is the impurity in graphite ranging from a few ppm to 200 ppm (part per million of the mass) [1, A5]. The deep geological disposal of big volumes of graphite would significantly affect the cost of radioactive waste management. On the other hand the concentrations of ^{14}C isotope as well as the isotopes of ^{10}Be, ^{36}Cl, ^{59}Ni, ^{63}Ni and the others will constitute the main acceptance criteria limiting the placement of the used graphite to the surface disposal. A number of graphite treatment techniques are under investigation [2]. These are focusing mainly on the reduction of the mass, immobilisation or separation of long-lived nuclides from the bulk carbon or both combinations at once. Cementation and impregnation of used graphite blocks permit to prevent the leakage of nuclides at a reasonably low price, however the mass of waste is eventually increasing. Incineration significantly reduces the volume of used graphite, the ^{14}C being released in the form of CO_2 to the atmosphere. Although the doses are negligible to the public, as the calculations show, this method was not well accepted by the public [3]. This raised the elaboration of new methods relying on steam pyrolisis and carbonization of ^{14}C, separation of ^{14}C from ^{nat}C by the methods of cryogenic fractional distillation, ^{14}CO and ^{12}CO gas flow excitation by electrical charge and separation based on different excitation energy of ^{14}C and ^{12}C, laser enrichment technologies and chemical non-aqueous CO_2 reactions with amines, where liquid phase of reaction is enriched by heavier $^{14}CO_2$ gas component [4]. Development of new used graphite treatment methods as well as elaboration of design requirements of graphite as a structure material for the reactor is also promoted by research on the generation IV reactors. Recent feasibility studies confirmed 6 new reactor concepts and even 2 of them are based on the use of graphite (namely, Molten Salt Reactor and Very High Temperature Reactor) [5].

The first step in the decision-making process of choosing the optimal technique for the used graphite management is the determination of concentrations of key radionuclides.

Experimental investigations are far from being the easiest way in drawing the detailed radiological characterisation because the sampling is usually limited to the accessibility to different reactor construction, I addition, the experimental analysis does not allow measuring all radionuclides concerned and, finally, such investigations are costly. Modern activation analysis of reactor components includes the use of numerical calculation techniques. Thus, the cost of investigation is significantly reduced, and the computer methods, on the contrary, are not limited to a certain activation radionuclides.

Preliminary numerical evaluation by the code ORIGEN of the activated radionuclide composition in the RBMK-1500 type reactor graphite was performed by the Lithuanian Energy Institute [6]. The uninterrupted activation of graphite for 15 years by the thermal neutron flux of $3 \cdot 10^{13}$ neutrons per cm^2 per second ($n/cm^2 \cdot s$) was used to evaluate the activity of graphite. The concentrations of impurities were used as given in scientific literature for this type of graphite. Another example is the evaluation of the induced radioactivity in Chernobyl and Leningrad NPPs with RBMK-1000 type reactors. The activity was calculated by combination of 2 computer programs: MCNP and CHAIN [7, 8]. The first code simulates the fluxes of neutrons in graphite and the other uses them for activation calculations. In all above calculations the total activity of graphite differs more than 10 times, and the concentrations of ^{14}C, ^{3}H and ^{55}Fe also varies significantly. The impurity rates, as reported in these works, considerably influence the calculation results. The bibliographic investigation of impurities in different types of graphite resumes that they may easily differ up to 100 times and more even for the same type of graphite. Consequently, the activation calculations are so much dispersed that may not be directly used for other type of graphite. The differences of operational parameters among reactors contribute to the uncertainty as well. In addition, special attention should be paid to the spectral variations of neutron flux because of the use of different fuel elements and its burn-up as investigations of evolution of neutron sensor characteristics in RBMK-1500 show [9]. Finally, the activation estimations of some individual radionuclides may be uncertain due to different neutron effective cross-section libraries used by the computer codes [A5].

Considerable experience of the use of computer codes for the neutron flux and activation calculations in reactors shows their effectiveness and credibility. For the modeling of simple reactor elements, like separate fuel assemblies, fuel channels, etc the deterministic computer codes are preferred versus Monte Carlo codes, whereas for the complex systems, containing many elements and materials, like reactor and its external structures, the calculations using Monte Carlo method give better results.

Chapter 2. METHODOLOGY

Mathematical model for the activation calculations involves solution of two equations: Bolzmann equation of neutron transport and Bateman equation of evolution of atomic densities of material in the flux of neutrons. The Bolzmann equation may be presented in integral-differential form:

$$\frac{1}{v}\frac{\partial}{\partial t}\Phi(\vec{r},v,\vec{\Omega},t) = -div\,|\vec{\Omega}\;\;\Phi(\vec{r},v,\vec{\Omega},t)\,| - \sum_T(\vec{r},v,\vec{\Omega},t)\;\;\Phi(\vec{r},v,\vec{\Omega},t) +$$

$$+\int_0^\infty dv' \int_{(4\pi)} d^2\vec{\Omega}' \sum_S(\vec{r},v'\to v,\vec{\Omega}'\to\vec{\Omega},t)\;\Phi(\vec{r},v',\vec{\Omega}',t) + S(\vec{r},v,\vec{\Omega},t)$$

Where $\Phi(\vec{r},v,\vec{\Omega},t)$ is the density of the neutron flux, $\vec{r}$ and $\vec{\Omega}$ are the radius and the angle vectors, t and v are the time and the scalar velocity. $\sum_T$ and $\sum_S$ are the total and the scatering macroscopic cross-sections. v' and Ω' are accordingly the velocity and the angle of the neutron before collision. S is the source of neutrons.

The Bateman equation representing the change of the concentration N_i for a nuclide i can be written as:

$$\frac{dN_i}{dt} = \sum_j \gamma_{ji}\sigma_{f,j}N_j\Phi + \sigma_{c,i-1}N_{i-1}\Phi + \lambda'_i\,N'_i - \sigma_{f,i}N_i\Phi - \sigma_{c,i}N_i\Phi - \lambda_i N_i$$

Where $\sum_j \gamma_{ji}\sigma_{f,j}N_j\Phi$ is the yield rate of N_i due to fission of j nuclides in the neutron flux, $\sigma_{c,i-1}N_{i-1}\Phi$ is the rate of transmutation of a nuclide $i-1$ to i due to neutron capture, $\lambda'_i\,N'_i$ is the rate of formation of nuclide i from nuclide i' due to radioactive decay, $\sigma_{f,i}N_i\Phi$ is the rate of destruction of nuclide i due to fission, $\sigma_{c,i}N_i\Phi$ is the rate of destruction of nuclide i due to all forms of neutron capture, $\lambda_i N_i$ is the rate of radioactive decay of the nuclide i itself.

Solution of the first equation for RBMK-1500 reactor in this Thesis is performed by using the Monte Carlo method and solution of the second equation is performed by using the deterministic method.

The overal calculation scheme is given in Fig. 1. The calculations are carried out in two stages. In the first stage the calculations of the spatial neutron flux distribution and energy spectra in the graphite are performed assuming desired reactor's thermal power. In the second stage material activation and radioactive decay during irradiation and after reactor shut down are simulated. For the input data the Monte Carlo code MCNPX [10] requires nuclear data, materials

and detailed geometry description. The obtained continuous neutron energy spectrum is merged into 63 energy groups and normalized to a nominal power of the reactor (4200 MW). The spatial neutron flux is further used as the entry data for the calculations in the second (activation) phase. To obtain the irradiation scenario which is close to realistic, the history of the power load of Unit 1 during 21 years was entered for the transmutation code CINDER'90 [11] by averaging in the steps of one year.

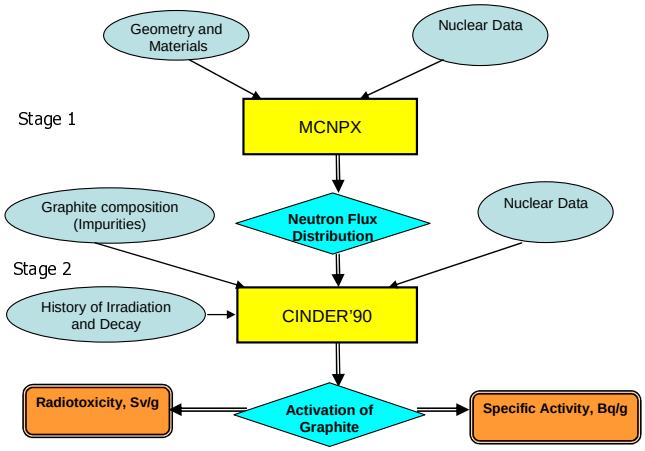

Fig. 1. The overall scheme of activation calculations.

The activation code also needs the chemical composition and impurities of graphite. The specimen of the fresh RBMK-1500 graphite taken from the fuel channel sleeve has been analyzed in Saclay Research Centre of French Atomic Energy Commission (CEA) by 2 independent methods: gamma spectroscopy based on activation by neutrons and glow discharge mass spectroscopy (GDMS).

Analysis of the first method consists of irradiation of the specimens by the thermal and fast neutron flux in the CEA Saclay research reactors namely ORPHEE and OSIRIS (Φ_{th} = (1.2 – 2.5)·10^{13}n/cm^2·s for ORPHEE and Φ_{fast} = 2·10^{13}n/cm^2·s for OSIRIS). After irradiation the specimens are processed and analyzed by gamma spectrometry. This method permits identifying the majority of chemical elements starting with the Z > 11 at the precision of the order of 10^{-13} - 10^{-9} g/(g of graphite). However, some important impurity elements such as Li, N, S, Nb, Pb may not be quantified by the gamma spectrometry method after irradiation.

GDMS is based on sputtering to plasma of the atoms of cathode which is made from the material to be analyzed. The atoms of material are subject to ionization by electrons ejected to argon gas plasma due to a potential difference of 500 – 1500 V or Penning ionization due to the ionization by argon atoms. The ionized atoms of material are then analyzed with the mass spectrometer. Although the method is rather sensitive (the limit of detection is below ((0.05 – 2)· 10^{-7} g /(g of graphite)) some elements as Li, N, O, Cl, F may not be quantified because of the absence of Penning ionization in argon.

Both impurity analysis methods allowed obtaining the maximum of data on impurities present in graphite. However, for more complete impurity composition, some important elements as Li, N and a few others, which could not be quantified by the above methods, have been taken from scientific literature, mainly from the "typical" RBMK graphite composition.

The code CINDER'90 uses 63 neutron energy groups and has its own nuclear data library originating from the merge of ENDF, JEF and JENDL data libraries including some model calculations. The output of the code is the isotopic composition and radionuclide activity in the irradiated material. The radiotoxicity of radionuclides is calculated conservatively using coefficients of committed effective dose per unit intake for adult person via inhalation or ingestion [12].

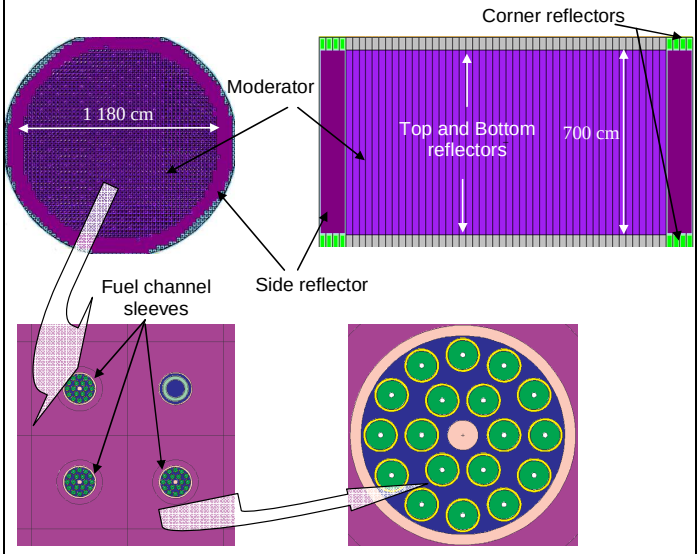

Fig. 2. The MCNPX model of RBMK-1500 reactor with horizontal and vertical sections (top), 4 lattice elements containing (bottom): moderator prisms, fuel channel sleeves, 3 fuel channels with 18 fuel rods, and a control rod. A single fuel channel is also presented separately.

The horizontal and vertical sections of the RBMK-1500 reactor as modeled by MCNPX are given in Fig. 2. The model also allows taking into account the smallest (pellets, cladding, air gaps between cladding and pellets, etc.) and the biggest details of reactor (whole reflector, reflectors and reactor external parts: metal constructions, water tanks, serpentine structures – they are not illustrated in Fig. 2).

Five graphite constructional zones (structures) were identified in the model and investigated separately: the **moderator** (graphite prisms in active zone), the **side** (circular) **reflector**, the **top** and the **bottom** reflectors, the part of reflectors, where the flux of neutrons is the lowest and nearly fully thermalized (hereinafter referred to as the **corner reflector**) and the **fuel channel graphite sleeves** which surround the fuel channel and are designed to fill the space between the fuel channel and moderator (in Fig. 2. - circle around the fuel channel).

Chapter 3. RESULTS

The flux of neutrons in the graphite of RBMK-1500

The MCNPX model for the Ignalina NPP reactors mainly uses the nuclear data from the ENDF/B-VI nuclear data library. The model takes into account the detailed geometrical parameters of the RBMK-1500 reactor. The cell of fuel assembly, the graphite prism and the control rods have been modeled as precisely as possible, taking into account the data given in the technical description [13]. The external reactor constructions (do not represented in **Fig. 2**) such as upper and bottom metal constructions of reactor, outer steel liner, serpentine constructions and circular water tank have been also included in the model. The temperatures of graphite and water are chosen from the available list in the nuclear data libraries and are correspondingly equal to 800 ^{o}K and 600 ^{o}K, which are close to the realistic operational temperatures. The density of water in model is 0.5 g/cm^3 and of graphite – 1.675 g/cm^3. The neutron flux and its energy spectrum are calculated in each of the above mentioned graphite constructions. Fig. 3 represents normalized distribution of neutron fluxes per unit lethargy in the RBMK-1500 reactor graphite constructions. As expected the "fastest" neutron flux is in the fuel channel sleeves (the

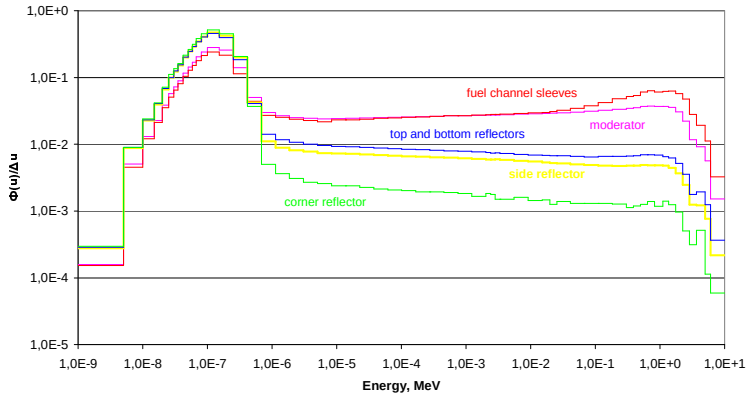

Fig. 3. Normalized distribution of neutron fluxes per unit lethargy in the RBMK-1500 reactor graphite constructions.

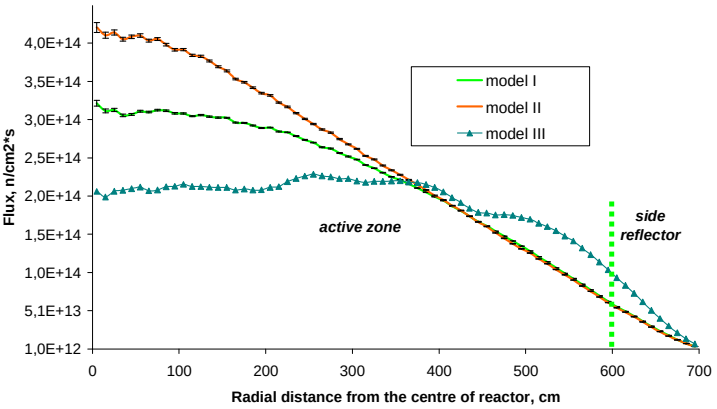

Fig. 4. Radial distribution of the neutron flux in moderator and side reflector of the RBMK-1500 reactor.

share of thermal neutrons is about 48 %) as they are the closest graphite constructional parts to the fast neutrons source – the fuel. The neutron flux is less fast in the moderator (with ~56 % thermalization) and in the reflectors the flux of thermal neutrons is more than 90 %.

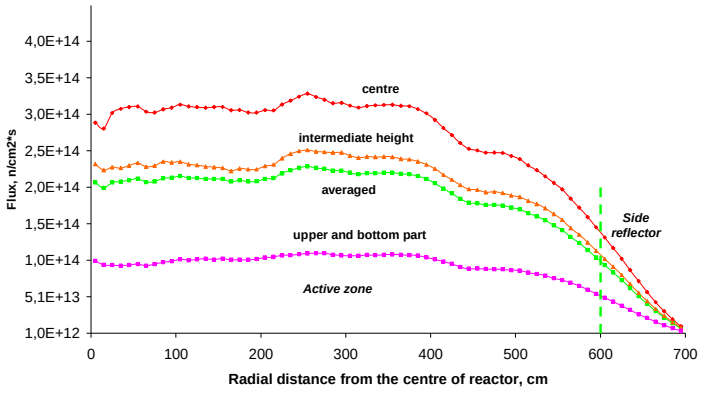

Fig. 5. Axial distribution of the neutron flux in moderator and in reflector of the RBMK-1500 reactor.

To evaluate how the neutron flux and its energy distribution in the moderator are sensible to the fuel burn-up and the presence of control rods, three models having the same basic geometry but a few important differences have been employed. Model I uses fresh fuel and has no control rods. The fresh fuel is replaced with the fuel of 10 MWd/kgU burn-up in model II (composition of irradiated fuel is obtained by calculations carried out with the ORIGEN code). Model III has the same burn-up as in the model II but has both fully and partially inserted control rods in the core. In all cases the initial enrichment of fuel was 2.0% for ^{235}U. The maximal values of the fluxes obtained in models I and II are in the center of the reactor and the flux regularly decreases at the boundaries of the core (**Fig. 4**). Such flux distribution is due to the fastest fuel burning in the center of the core and slower burning at periphery. However, they do not represent the realistic distribution of the flux which is uniform in the plateau of the core due to the presence of control rods. Model III allows “decreasing” the power in the center of the reactor and “pushing” it to the periphery, which constitutes its principal advantage compared to models I and II and increases the flux in the side reflector up to 85% (see **Fig. 4**). The axial distribution of the neutron flux in moderator and in reflector is shown in **Fig 5**. Comparing the calculated curve of axial neutron flux with the curve of the axial power in the reactor, which is proportional to the neutron flux, the variations are less than 30%, for radial flux – less than 15%. The possibility to improve the model further faces some difficulties. Indeed, in further optimization even smaller fragments of control rods, including the axial fuel burn-up profile, should be used, which makes the task too sophisticated and costly in terms of computer time. It will be shown later that 30 % variation in the neutron flux is not the most important contribution to the final activation analysis.

Fig. 6 shows the effects the different models have on the energy spectra of the neutrons. The relative differences of neutron spectra in moderator are less than 1% in thermal energy part and less than 9% in fast neutron energy part for the same geometry model but different fuel burn-up and presence or absence of the control rods. This result ensures that deviations of neutron flux spectrum in graphite structures because of the fuel burn-up and the changing positions of control rods may not lead to the significant error in activation calculations.

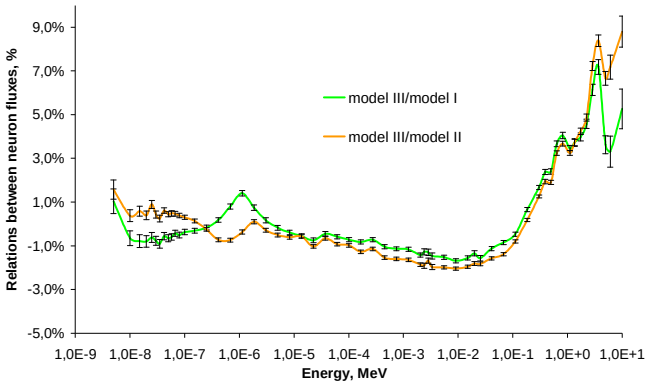

Fig. 6. Relations between neutron fluxes of moderator in models I - III.

Other parameters, which may influence the flux calculations, are the variable density of the water-steam mixture in the fuel channel and the use of higher fuel enrichment (and poisoned with erbium). It has been evaluated that if the realistic water-steam mixture distribution in the fuel channel were used, the averaged neutron energy spectrum in graphite would remain almost the same (deviations are less than 1% in thermal and epithermal region and less than 3% in fast neutron region). The share of thermal neutrons would differ about 5% in the bottom and upper part of the core from the share of averaged neutron flux in graphite. However, such differences have no much influence to the real axial distribution of the flux in the reactor where the flux of thermal neutrons is regulated by the means of the control rods and may be neglected in modeling.

As long as the erbium poison is concerned, the fuel in model has been replaced with the erbium poisoned fuel of 13MWd/kgU burn-up. In this case the difference in neutron energy spectrum in graphite is less than 4%. Taking into account the fact that the erbium fuel campaign in the Ignalina NPP Unit 1 constitutes less than 1/3 of the whole operation period, this difference may be regarded as insignificant.

Unfortunately, there are no representative experimental measurements to compare the calculated total flux of neutrons in graphite structures of RBMK-1500. However, some calculations done by different computer codes are available. Such is the estimation of the flux of fast neutrons in the Ignalina NPP graphite by the code WIMS-D4 within the framework of the studies on the gap problem between the fuel channel and graphite stack [14]. The flux of fast neutrons calculated in this Thesis agrees with the flux of the study within 10%. The flux of

thermal neutrons of RBMK-1500 may be also compared with the same flux of RBMK-1000 reactor providing the thermal power of both reactors is normalized accordingly. Calculated average flux of thermal neutrons in the graphite of RBMK-1000 reactor [15] is 16% lower than the same flux calculated in this work. Finally, the Monte Carlo calculations carried out by Kurchatov Institute give good agreement of the proportionality of the flux of thermal neutrons in the graphite sleeves and graphite moderator prisms [16]. The agreement is within 3% margin. These comparisons confirm that the calculations of neutron fluxes in this work are credible and realistic for the activity calculations in graphite.

Investigation of impurities in the graphite sample of the Ignalina NPP.

The results of the investigation of impurities of Ignalina NPP graphite are presented in **Table 1**. The results of irradiation-activation analysis are in **black** and the results of GDMS – in **green**. The results of two different techniques are in good agreement for Na, K, Cr, and Mn. But they differ in case of Mg, Al, Ca and Fe. In general, the concentrations measured by irradiation technique are higher. Therefore for the conservativeness of final results they are used for the activation analysis together with the impurity concentrations of B, Si, P, and Cu measured only by GDMS technique.

Note that some important impurities were not quantified by irradiation, or by GDMS technique. The average value of such impurity concentrations were taken as follows: Li – 0.027 ppm, Be – 0.02 ppm, N – 35.25 ppm, O – 119 ppm, S – 28.5 ppm. The bibliographic study was performed to compare the impurity concentrations, measured by irradiation and GDMS techniques in RBMK-1500 graphite and the impurities of RBMK and other graphite types as found in the scientific literature The comparison is presented in **Fig. 7**.

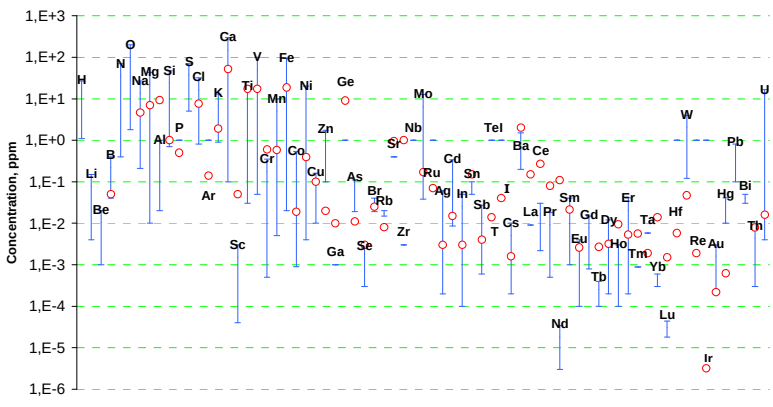

Fig. 7. Comparison of measured impurities of RBMK-1500 graphite by irradiation and GDMS techniques (red circles) with bibliographic data (in blue - presented as the margins of maximal and minimal concentration value).

Table 1. Investigation of impurities of Ignalina NPP graphite. In **black** – irradiation analysis; in **green** – GDMS analysis.

Impurity	concentration, ppm	3 σ , %	Impurity	concentration, ppm	3 σ , %
B	**0.05**		*Cd*	0.015	-
Na	4.64; **5**	19	*In*	0.003	-
Mg	7; **0.5**	-	*Sn*	0.15	-
Al	9.2; **1.0**	4	*Sb*	0.004	6
Si	**1.0**		*Te*	0.014	35
P	**0.5**		*I*	0.04	-
Cl	7.6	7	*Cs*	0.0016	28
Ar	0.14	-	*Ba*	2.0	7
K	1.9; **1.5**	36	*La*	0.15	19
Ca	51.9; **2.0**	13	*Ce*	0.27	2
Sc	0.05	1	*Pr*	0.08	-
Ti	17.4	8	*Nd*	0.11	6
V	17.4	4	*Sm*	0.021	1
Cr	0.60; **0.3**	5	*Eu*	0.0026	6
Mn	0.58; **0.2**	5	*Tb*	0.0027	6
Fe	18.7; **1.0**	16	*Dy*	0.0032	45
Co	0.019	2	*Ho*	0.0094	20
Ni	0.39	25	*Er*	0.0053	-
Cu	**0.1**		*Tm*	0.0056	4
Zn	0.02	-	*Yb*	0.014	5
Ga	0.01	-	*Lu*	0.0015	4
Ge	9	-	*Hf*	0.0058	5
As	0.011	20	*Ta*	0.0019	10
Se	0.003	-	*W*	0.047	11
Br	0.025	41	*Re*	0.0019	14
Rb	0.008	-	*Ir*	0.000003	-
Sr	0.96	17	*Au*	0.0002	5
Zr	1.0	21	*Hg*	0.0006	-
Ru	0.07	-	*Th*	0.0079	5
Mo	0.17	2	*U*	0.016	8
Ag	0.003	60			

This comparison (**Fig. 7**) permits to evaluate how conservative the measurements performed in this work are and to estimate expected trends of activated radionuclides in graphite. It also shows the big disperse of impurity concentrations among the nuclear grade graphites. It is worth mentioning that in this work the concentrations of Ga, Rb, Sr, Zr, Mo, In, Te, Ba, La, Ce, Pr, Nd, Sm, Eu, Tb, Dy, Ho, Er, Tm, Ta, Yb, Lu impurities in RBMK type graphite were estimated for the first time. The impurities of P, Ar, Ge, Ru, Te, I, Hf, Re, Ir were not measured at all in any type of graphite before.

Calculation of specific activities and radiotoxicities of Ignalina NNP used graphite

The result of the calculations of activity of graphite under realistic irradiation conditions and impurity concentrations as described above is the list of more than 1300 radionuclides with the range of their half-lives from 10^{-7}s to 10^{20}y. Only the radionuclides which specific activity or radiotoxicity in graphite 10 years after the shut down of the reactor is higher than 0.1 Bq/g and 10^{-6} Sv/g accordingly are discussed in this work for detailed evolution analysis. However the estimation of the total specific activity and radiotoxicity takes into account all radionuclides. The total specific activity of isotopes in moderator and side reflector as a function of radial distance and the time after the shut down of the reactor is presented in the **Fig. 8**. The activity is constant in the zone of reactor power plateau and it decreases to the periphery of the active zone. However, in the side reflector at the edge of the core the total activity slightly increases. It is due to the higher flux of thermal neutrons in reflector whose contribution to the total activity is essential. Total activity decreases sharply after the shut down of the reactor because of decay of short-lived nuclides and after 30 years of cooling it changes very slowly for a long time. Axial distribution of specific activity in moderator, top and bottom reflector, side reflector and corner reflectors is shown in **Fig. 9**. The increase of specific activity in corner reflector zone is much more important as in case of radial distribution. Isotopic analysis shows that tritium is responsible

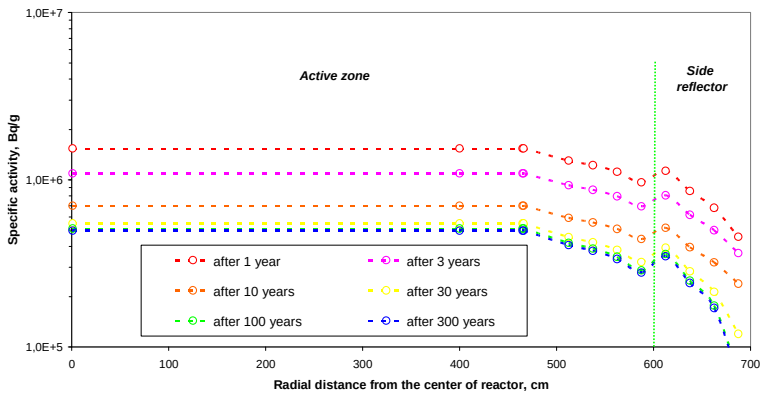

Fig. 8. Radial distribution of specific activity of RBMK-1500 graphite as a function of cooling time.

for such increase. The production rate of tritium from ^{6}Li is very intensive in the active zone. Practically all ^{6}Li is burnt in a high neutron flux of moderator during the first 2 years of operation and afterwards the balance of this isotope is described by the law of radioactive decay only. But the rate of this reaction is less intensive in reflectors and tritium is being produced during the whole operation time. Because of this fact the tritium activity proportion in graphite constructions is very different.

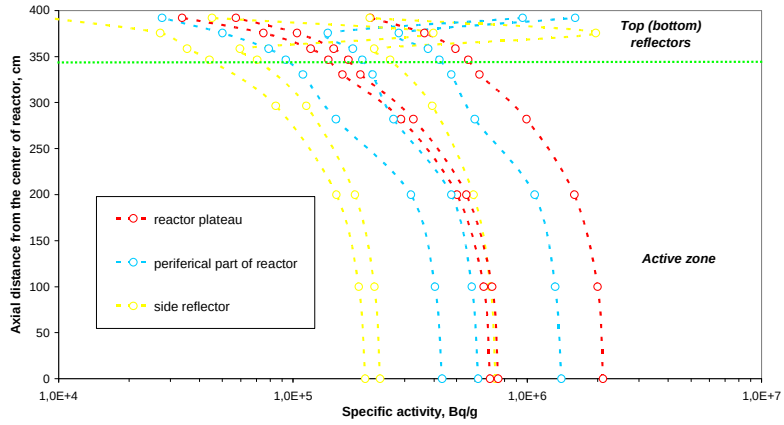

Fig. 9. Axial distribution of specific activity of RBMK-1500 graphite as a function of cooling time.

The same effect is observed for other mother-nucleus with big neutron capture cross-sections and the half-lives of resulted radionuclides shorter or comparable with the operation time of the reactor, e.g. ^{59}Co, ^{133}Cs, ^{151}Eu, ^{153}Eu. The distribution of activities of resulting radionuclides as ^{60}Co, ^{134}Cs, ^{152}Eu, ^{154}Eu, ^{155}Eu is not linearly proportional to the density of flux of neutrons.

The specific activity in the plateau of moderator of RBMK-1500 reactor is shown in **Fig. 9**. In the right part of the graph the clearance levels of individual radionuclides are defined. The activity of graphite in the corner reflector is given in **Fig. 10** where the elevated activity of tritium may be observed. In other graphite constructions the specific activities are intermediate with some exception concerning above mentioned nuclides

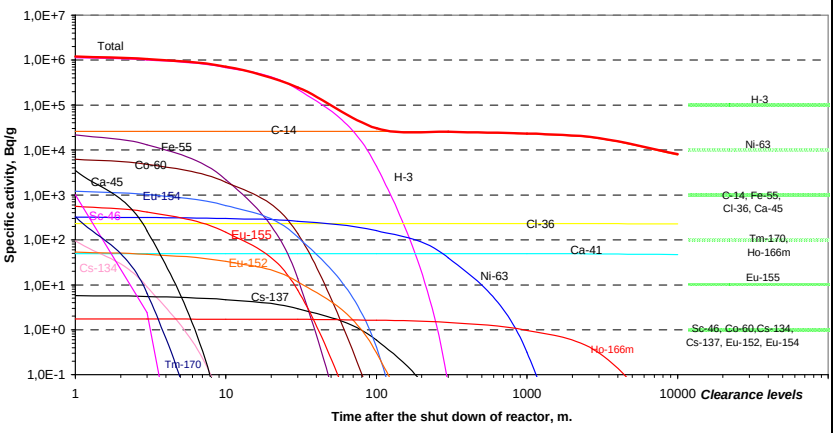

Fig. 10. Specific activity of radionuclides in the corner reflector of RBMK-1500 as a function of cooling time.

which are burnt intensively in high fluxes. From the specific activity point of view ^{14}C and tritium contribute mostly to the total activity of all the irradiated graphite constructions respectively $3.91 \cdot 10^{14}$ Bq and $2.19 \cdot 10^{14}$ Bq. The activity of other radionuclides such as ^{55}Fe ($4.45 \cdot 10^{14}$ Bq) and ^{60}Co ($9.85 \cdot 10^{13}$ Bq) have to be also taken into account, especially for dismantling and on-site radioactive waste management purposes. However, radioisotopes as ^{3}H (12.33 y), ^{55}Fe (2.73 y) and ^{60}Co (5.27 y) having the short half-lives do not represent especial concerns for long term management. Their specific activities are expected to reach their clearance levels in all graphite constructions after 40, 30 and 90 years respectively. ^{14}C (5730 y) activity remains above clearance levels more than 50000 years and constitutes the main concerns for the long term management.

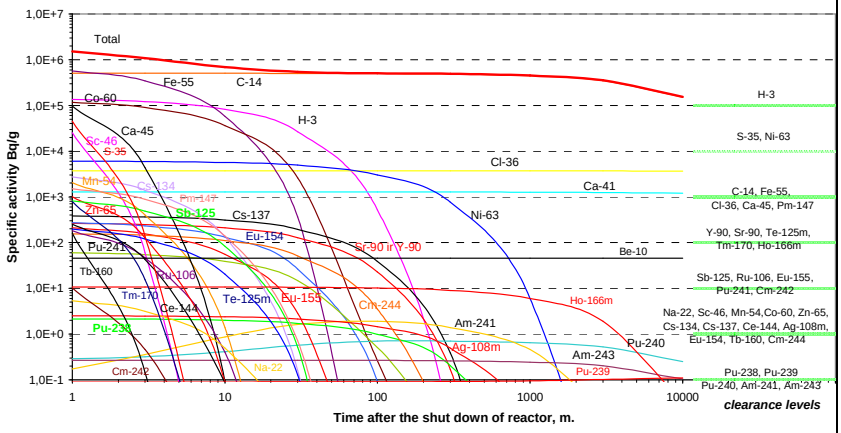

Fig. 9. Specific activity of radionuclides in the plateau of RBMK-1500 moderator as a function of cooling time.

The following nuclear reactions contribute mostly to the total activity of irradiated graphite:

^{6}Li (n, α) → Tritium;
^{13}C (n, γ) → ^{14}C;
^{14}N (n, p) → ^{14}C;
^{35}Cl (n, γ) → ^{36}Cl;
^{40}Ca (n, γ) → ^{41}Ca;
^{54}Fe (n, γ) → ^{55}Fe;
^{59}Co (n, γ) → ^{60}Co;
^{62}Ni (n, γ) → ^{63}Ni;
^{133}Cs (n, γ) → ^{134}Cs;
^{153}Eu (n, γ) → ^{154}Eu;
^{154}Eu (n, γ) → ^{155}Eu;

The reactions: ^{151}Eu (n, γ) → ^{152}Eu; ^{107}Ag (n, γ) → ^{108m}Ag; ^{9}Be (n, γ) → ^{10}Be; ^{165}Ho (n, γ) → ^{166m}H are not very important in the context of total activity and radiotoxicity as calculated in this work for the RBMK-1500 reactor, although they have been reported to be as the critical ones in the literature. Other important radionuclides, like ^{137}Cs, ^{90}Sr are the fission products of uranium and thorium impurity naturally present as impurities in graphite. Moreover, the presence of

uranium in graphite results in creation of transuranium isotopes, the concentrations of which in most irradiated graphite constructions are much higher than their decommissioning clearance levels.

The radiotoxicity in (Sv/g) has been obtained simply by multiplying the specific activity (Bq/g) of the radionuclide in graphite with the conservative value of committed effective dose per unit intake for an adult via inhalation or ingestion (Sv/Bq) [17]. The results of radio-toxicity calculations are presented in **Fig. 11**. The total radiotoxicity of graphite reaches 6.6 mSv/g in the moderator. It is evident, that transuranium elements due to their radio-toxicity (^{244}Cm, ^{238}Pu, ^{241}Am, ^{240}Pu) must be taken into account and may cause particular limitations for short and long time waste handling and packaging. After approximately 50 years, the radiotoxicity of ^{14}C becomes mostly important. ^{60}Co is also important from the radiotoxicity point of view. In reflectors the radiotoxicity of each ^{154}Eu and ^{3}H constitutes up to 20% of the total radiotoxicity.

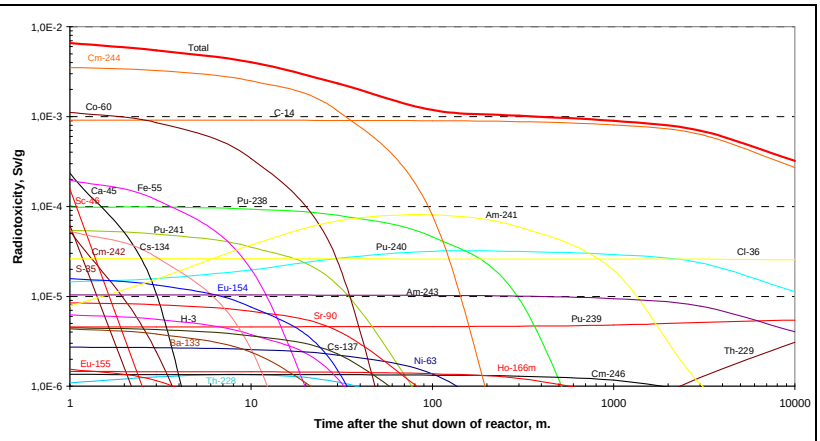

Fig. 11. Radiotoxicity of radionuclides in the plateau of RBMK-1500 moderator as a function of cooling time.

Uncertainty estimations

The accuracy of such activation calculations is mainly determined by the uncertainties of the data on impurity concentrations and uncertainties of the effective neutron cross-section data libraries used. The presence of impurities is due to the source and other materials involved in fabrication of nuclear graphite. Therefore their concentrations are sensitive to technological process and sometimes vary even for the same grade of graphite. The results of investigation of variance of impurity concentrations (**Fig. 7**) were used to estimate probable uncertainty of activity calculations. In **Fig 12** and **Fig. 13** such uncertainty estimations for the specific activity and radiotoxicity are shown. Uncertainty of a given radionuclide is within 2 curves corresponding to minimal and maximal impurity concentration of a mother-nuclide. Dotted curve in between of these curves corresponds to the measured (average – for Li and N impurities) concentrations.

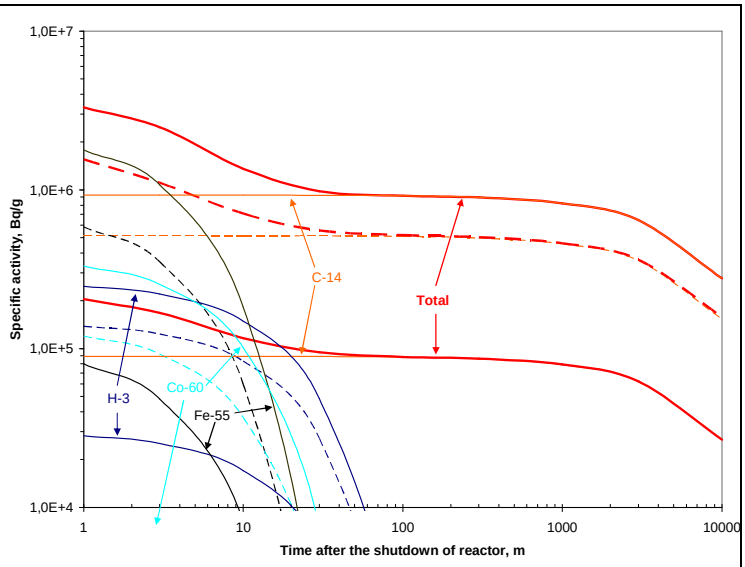

Fig. 12. Specific activity of radionuclides in the plateau of RBMK-1500 moderator in case of minimal, maximal and measured impurity concentrations of mother-nuclides.

Following impurity concentrations were taken for this estimation (ppm): Li (0.004; 0.027; 0.05), N (0.5; 32.25; 70), Fe (2.6; 18.7; 58), Co (0.0009; 0.019; 0.053) U (0.01; 0.016; 0.13).

In the context of the total activity and radiotoxicity the uncertainties in predicting the production of ^{14}C and ^{244}Cm are mostly significant. In the case of the U impurity, its measured concentration (0.016) is very close to a minimal value (0.01), therefore the activities of ^{244}Cm is likely to be higher than evaluated in this work.

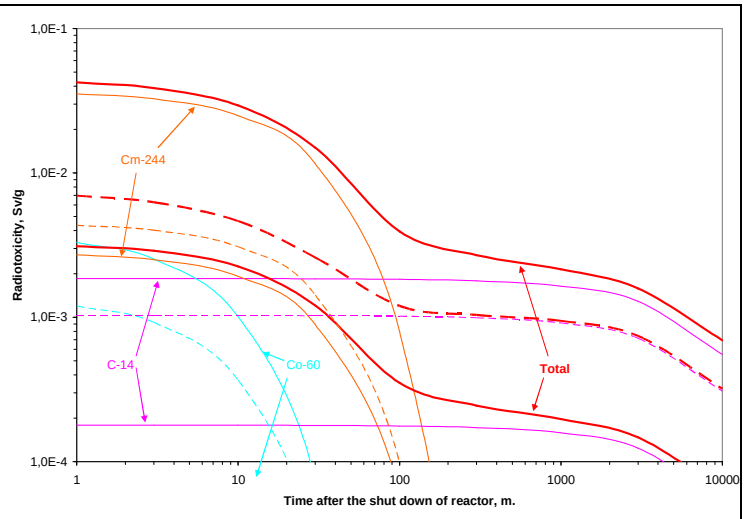

Fig. 13. Radiotoxicity of radionuclides in the plateau of RBMK-1500 moderator in case of minimal, maximal and measured impurity concentrations of mother-nuclides.

In fact, the production rate of a radionuclide and the neutron reaction rate per unit of flux may differ from one graphite construction to another due to the difference in spectrum of the neutron flux in these constructions (**Fig. 3**). Although the major activity is a result of reactions from thermal neutrons, some radionuclides in graphite in comparable quantities are produced by reactions with epithermal neutrons e.g.: ^{60}Co - 15%, ^{108m}Ag - 22%, ^{134}Cs -

52%, ^{154}Eu – 35%. Fast neutrons produce about 9% of ^{14}C from ^{13}C in moderator. Therefore, activity calculations of these radionuclides based only on thermal neutrons may lead to a considerable underestimation of their total activity.

The uncertainties which are the result of the use of different libraries of neutron reactions effective cross-section data are less important than those caused by uncertainties of impurity concentrations. Nevertheless, for some individual radionuclides they are not negligible. The best known nuclear data libraries, namely ENDF/B-VI, JEF-2, and JENDL-3 were compared with the original CINDER'90 library.

Table 2 gives relations of these libraries calculated for the neutron flux in RBMK-1500 moderator for thermal as well as both epithermal and fast neutrons. It is evident that the significant variation of the results for some individual radionuclides (^{14}C, ^{134}Cs, ^{154}Eu) may take place because of the use of a certain cross-section data library and negligible influence of epithermal and fast neutrons. The following neutron energy intervals were taken: thermal (10^{-3} eV – 0.7 eV), epithermal (0.7 eV – 0.1 MeV), fast (0.1 MeV – 25 MeV).

Table 2. Differences of neutron effective cross-sections using different libraries (related to ENDF/B-VI or to the library with the note "Ref.").

Reaction	*Thermal region*			*Epithermal and fast region*		
	JEF-2	JENDL-3	CINDER'90	JEF-2	JENDL-3	CINDER'90
^{6}Li (n, α) → ^{3}H	-0.164%	0.106%	-0.580%	-0.064%	0.119%	-0.757%
^{13}C (n, γ) → ^{14}C	-	-	-32.00%	-	-	-74.99%
^{14}N (n, p) → ^{14}C	-0.558%	-2.995%	95.58%	-1.689%	-4.822%	92.78%
^{35}Cl (n, γ) → ^{36}Cl	-	Ref.	-0.170%	-	Ref.	2.674%
^{40}Ca (n, γ) → ^{41}Ca	-	Ref.	0.057%	-	Ref.	0.000%
^{54}Fe (n, γ) → ^{55}Fe	14.80%	-4.17%	0.126%	15.81%	12.54%	0.117%
^{59}Co (n, γ) → ^{60}Co	-0.154%	0.111%	0.430%	-0.475%	-0.335%	-1.868%
^{62}Ni (n, γ) → ^{63}Ni	-0.178%	-1.218%	0.583%	-0.090%	16.89%	0.325%
^{133}Cs (n, γ) → ^{134}Cs	-1.726%	-1.822%	2.323%	14.41%	3.347%	2.155%
^{151}Eu (n, γ) → ^{152}Eu	-0.186%	0.319%	-31.21%	-0.313%	-10.56%	-34.53%
^{153}Eu (n, γ) → ^{154}Eu	-15.85%	Ref.	4.659%	3.036%	Ref.	20.631%
^{154}Eu (n, γ) → ^{155}Eu	-79.13%	Ref.	-16.99%	119.62%	Ref.	11.05%

CONCLUSIONS

1. The model for the evaluation of spatial and spectral distribution of neutrons as well as formation of radioactive impurities in RBMK-1500 reactor's moderator and reflectors is created and is based on 2 computer codes (MCNP and CINDER'90).
2. The specific activity of irradiated RBMK-1500 graphite is mainly determined by the specific activities of ^{14}C, ^{3}H, ^{60}Co, and ^{55}Fe. The most important contributor to the radiotoxicity of graphite is ^{244}Cm, ^{60}Co, ^{14}C and ^{154}Eu. The specific activities and radiotoxicities of these and other radionuclides in various graphite structures differ significantly.
3. Activated graphite of Ignalina NPP due to the high activity, up to $5 \cdot 10^5$ Bq/g, of the long-lived ^{14}C is classified as the long-lived radioactive waste. The presence of other short-lived radionuclides and especially actinides may cause certain short or long term limitations for the packaging of spent graphite waste.
4. Modelling of axial and radial distributions of the flux of neutrons in moderator and reflectors by Monte Carlo technique and estimation of their activation has revealed the non-homogeneous distribution of some radionuclides. ^{6}Li, ^{59}Co, Eu and some other isotopes are intensively transmuted ("burned") in the active zone of moderator. However their burn up in reflectors where the flux of neutrons is significantly lower is less intensive and ^{3}H, ^{60}Co and Eu radioisotopes are produced during all operational time.
5. Differences of estimation of radioactive impurities in irradiated graphite of the Ignalina NPP are possible due to the big variation of impurity concentrations in graphite. These differences influence the radiological characteristics of graphite significantly. Analysis of the modelling results has shown that better estimation of radioactive impurities requires update of nuclear libraries.

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RBMK-1500 REAKTORIAUS GRAFITO RADIONUKLIDINĖS SUDĖTIES EVOLIUCIJA

Reziumė

Šio darbo tikslas – sukurti radioaktyviųjų priemaišų Ignalinos atominės elektrinės reaktorių grafite susidarymo modelį, įvertinti grafito radiacines charakteristikas bei pateikti mokslines prielaidas tolesniems panaudoto grafito tvarkymo būdams pasirinkti. Tikslams įgyvendinti suformuluoti šie uždaviniai: Ignalinos atominės elektrinės reaktorių grafito konstrukcijų neutronų aktyvacijos modelio sukūrimas; pagrindinių veiksnių, lemiančių aktyvacijos radionuklidų balansą nustatymas; aktyvacinių radionuklidų kiekio grafite, remiantis eksperimentiniais Ignalinos AE naudojamo neapšvitinto grafito priemaišų tyrimo rezultatais skaičiavimas ir RBMK-1500 reaktoriaus grafito radiotoksiškumo įvairiose lėtiklio ir reflektoriaus zonose įvertinimas bei radiotoksiškumo kaitos ilgalaikio saugojimo metu prognozavimas.

Įvertinus neutronų srauto tankio pasiskirstymą bei spektrines charakteristikas pirmą kartą sumodeliuotas RBMK reaktoriui būdingas aksialinis ir radialinis radioaktyviųjų priemaišų aktyvumo nehomogeniškumas reaktoriaus lėtiklyje ir reflektoriuose. Darbe pateikiami nauji duomenys apie reaktoriaus grafito radiotoksiškumą ir identifikuojamos priemaišos, lemiančios radioaktyviųjų atliekų klasifikacijos rezultatus.

Šiame darbe atlikti aktyvacijos tyrimai rodo, kad Ignalinos AE aktyvuotas grafitas dėl didelio, iki $5 \cdot 10^5$ Bq/g, ilgaamžio ^{14}C anglies izotopo savitojo aktyvumo yra priskiriamas ilgaamžėms radioaktyviosioms atliekoms. Savitąjį grafito aktyvumą lemia šie radionuklidai: ^{14}C, ^{3}H, ^{60}Co, ^{55}Fe ir ^{154}Eu. Daugiausia įtakos grafito radiotoksiškumui turi ^{244}Cm, ^{14}C ir ^{60}Co. Monte Karlo metodu sumodeliavus neutronų srauto pasiskirstymą reaktoriaus aktyviojoje zonoje ir reflektoriuose ir atlikus aktyvacijos skaičiavimus, gautas aksialinis ir radialinis savitojo aktyvumo pasiskirstymas Ignalinos AE grafito konstrukcijose. Nustatyta, kad kai kurie radionuklidai yra neproporcingai pasiskirstę neutronų srauto tankio atžvilgiu. ^{6}Li, ^{59}Co, Eu ir kai kurie kiti izotopai sparčiai transmutuoja ("išdega") klojinio aktyviojoje zonoje, tačiau jų išdegimas reflektoriuose, kur neutronų srauto tankis daug mažesnis, yra lėtesnis. Tai sąlygoja dideles ^{3}H, ^{60}Co, Eu radiozotopų koncentracijas reflektoriuose, nes juose minėtų radionuklidų formavimasis vyksta visą eksploatavimo laiką.

Dėl didelio grafito priemaišų duomenų skirtumų, galimi radioaktyviųjų priemaišų vertinimo skirtumai Ignalinos AE reaktoriuose naudojamame grafite. Šių skirtumų įtaka radiacinėms grafito charakteristikoms yra esminė. Skaitinio modeliavimo rezultatų analizė parodė, kad tikslesniam radioaktyviųjų priemaišų grafite įvertinimui reikalingos tikslesnės branduolio duomenų bibliotekos.

Apie 3800 tonų masės panaudoto grafito išmontavimas ir galutinis jo sutvarkymas yra vienas iš pagrindinių Ignalinos AE išmontavimo darbų. Šioje disertacijoje gauti rezultatai ir išvados bus naudingi detaliems grafito aktyvacijos skaičiavimams ir eksperimentiniams tyrimams bei sudarys prielaidas saugios ir ekonomiškai pagrįstos grafito tvarkymo technologijos pasirinkimui.

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