ESTIMATION OF THE GENERATION OF ^{13}C AND ^{14}C IN THE REACTOR GRAPHITE USING MCNP6 MODELLING, ISOTOPE RATIO MASS SPECTROMETRY AND ^{14}C MEASUREMENT TECHNIQUE

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ABSTRACT

Characterization of irradiated graphite in terms of ^{14}C activity is crucial for the optimization of treatment technology: geological disposal, landfill storage, recycling, etc. The main contributor to ^{14}C generation in the RBMK reactor graphite is ^{14}N(n, p)^{14}C reaction. The generation of carbon isotopes ^{13}C and ^{14}C in the virgin RBMK graphite samples irradiated at the LVR-15 research reactor (Research Centre Řež, Ltd.) were investigated in order to obtain the impurity concentration level of ^{14}N. Afterwards the modeling of graphite activation in the RBMK-1500 reactor was performed by computer code MCNP6 using obtained ^{14}N impurity concentrations and new nuclear data libraries. The irradiation parameters – neutron fluence have been checked by method based on coupling of stable isotope ratio mass spectrometry and computer modelling. The activity of ^{14}C in the different constructions of irradiated graphite of the RBMK-1500 reactor has been measured by the β spectrometry technique (LSC) and has been compared with the simulated one. Obtained results have indicated the importance of ^{14}C production from ^{14}N in the RBMK-1500 reactor and in the LVR-15 neutron spectrum. Measured ^{14}C specific activity values in the samples varied from 130-700 kBq/g in the RBMK-1500 irradiated samples and from 3-12.5 Bq/g in the LVR-15 irradiated graphite samples. This corresponds to 15±4 - 80±10 ppm impurity of ^{14}N in various graphite samples of RBMK reactor.

KEYWORDS: ^{14}C, nuclear grade graphite, RBMK-1500 reactor, LSC and stable isotope ratio mass spectrometry, MCNP6 modeling.

1. INTRODUCTION

^{14}C is one of the limiting radionuclide for long-term disposal of irradiated graphite due to half-life of 5730 years and relatively high activity as well as mobility in geological media [1]. Two RBMK-1500 reactors operated in Ignalina NPP (Lithuania): Unit 1 in 1984-2004, Unit 2 – in 1987-2009. The graphite used as a moderator and reflector in both reactors representing some 3800 tones. According to the existing radiological classification the graphite waste is attributed to the long-lived low and intermediate activity waste [2]. The timescale for geological disposal of spent nuclear fuel and operational radioactive waste of INPP is previewed by 2066 in Lithuania [3]. ^{14}C is generated in the graphite moderator-reactor due to the neutron capture by ^{13}C in the graphite matrix or by the neutron activation of ^{14}N and ^{17}O impurities. The main contributor to ^{14}C generation in the RBMK reactor is ^{14}N(n, p)^{14}C reaction due to high neutron reaction cross section. ^{14}N concentration in the graphite is determined by initial impurity concentration and because of ^{14}N abundance in the helium-nitrogen mixture used to flush the graphite stack in the core. There is an urgent need of ^{14}C activity determination for the optimization of spent graphite treatment technology (e.g. geological disposal, landfill storage, recycling, etc.). The determination of concentrations of radioactive impurities in irradiated graphite has a primary importance to its management strategy and may lead to considerable savings of decommissioning funds.

In this research importance of both ^{14}C generation channels via ^{13}C and ^{14}N was investigated with virgin and with irradiated graphite. Firstly the ^{14}N impurity concentration level was estimated in the virgin RBMK graphite samples irradiated at the LVR-15 research reactor. Afterwards the modeling of graphite activation in the RBMK-1500 reactor was performed by computer code MCNP6 using obtained ^{14}N impurity concentrations and new nuclear data libraries. The irradiation parameters – neutron fluence have been checked by method based on coupling of stable isotope ratio mass spectrometry and computer modelling.

2. ESTIMATION OF ^{14}N IMPURITY CONCENTRATIONS IN THE VIRGIN RBMK-1500 REACTOR GRAPHITE

For the impurity concentration of ^{14}N in the virgin RBMK graphite determination the samples irradiated in LVR-15 research reactor (Research Centre Řež, Ltd.) for INAA experiment – have been latter on investigated by the β spectrometry technique (LSC) at CPST (Lithuania). The formation of carbon isotopes ^{13}C and ^{14}C in INAA RBMK graphite specimens was obtained after irradiation in the LVR-15 research reactor at the thermal neutron flux of 4.9×10^{13} $cm^{-2}s^{-1}$ [4]. Neutron flux distribution is presented in Table 1.

Table I. Parameters of the neutron spectrum at LVR-15 H8 irradiation channel.

E (MeV)	Φ ($cm^{-2}s^{-1}$)	Φ (%)
$0{,}0 - 5{,}0\times10^{-8}$	$3{,}0\times10^{13}$	61.2%
$5{,}0\times10^{-8} - 0{,}1$	$1{,}0\times10^{13}$	20.4%
$0{,}1 - 20$	$9{,}0\times10^{12}$	18.4%

Modelling of sample activation in LVR-15 neutron flux was performed using MCNP6 [5]. $^{14}N(n, p)^{14}C$, $^{13}C(n, \gamma)^{14}C$ and other reaction rates have been obtained in the samples irradiated at H8 irradiation channel. Calculated reaction rates are as folows:

$$RR_i = N\int_0^\infty \sigma(E)\phi(r,E)dE \tag{1}$$

were, RR_i - rate at which reactions are occurring in the nuclide i (reactions/s); N - number of target atoms in the sample; $\sigma(E)$- energy-dependent microscopic cross-section; $\phi(E)$ - energy-dependent neutron flux in the sample (n/cm^2s). The impurity concentrations have been obtained according to measured activities. The experimental reactions rates are calculated taking into account irradiated samples activity and irradiation parameters [6]:

$$RR = \frac{A_t}{\Phi\cdot\sigma\cdot(1-e^{\lambda\cdot t_a})}, \tag{2}$$

were A_t – activity of the sample at the end of irradiation (Bq), Φ – neutron flux (n/cm^2s), σ – microscopic neutron cross section (cm^2), t_a – time of activation (s), λ – decay constant (s^{-1}). The real concentration of impurity is determined by comparing calculated and experimentally obtained activity values. The 14N reaction rate was checked by ^{60}Co activity and corresponding impurity concentration (see Table II).

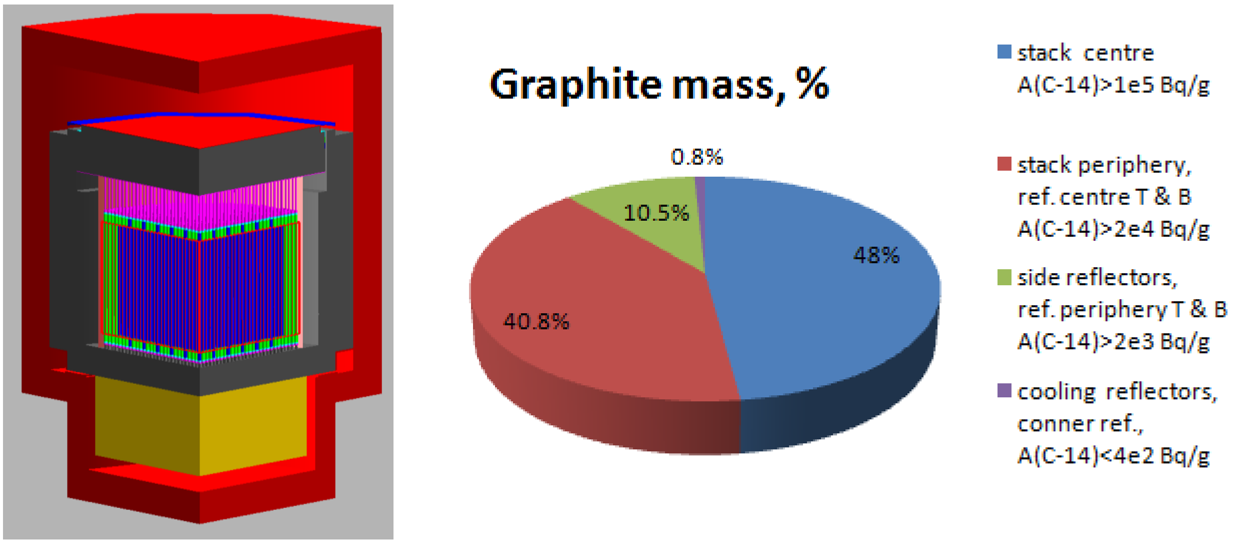
The obtained results show, that ^{14}C production from ^{13}C determines ~1,7 Bq/g graphite activity, ^{14}N input to generation of ^{14}C activity was calculated to be about ~70-90% in case of LVR-15 neutron spectrum. Measured ^{14}C specific activity values in the samples varied from 6-17 Bq/g in the LVR-15 irradiated graphite samples. This corresponds to 25±4 - 90±10 ppm impurity of ^{14}N in various graphite samples of RBMK reactor. This result is higher than maximal ^{14}N impurity concentration obtained in earlier work ^{14}N [7]. We should note what irradiation in the LVR-15 environment (irradiation in the air) corresponds to surface ^{14}N concentration of the graphite. Additional experiments are previewed for investigation of virgin samples in the RBMK-1500 reactor environment (N+He mixture flushing gas).

Table II. The neutron activation results of RBMK-1500 virgin graphite in the LVR-15 neutron flux.

No.	Graphite construction	γ measurements			β (LSC) measurements		Caculated conc. of ^{14}N (ppm)	^{14}C,% from $^{14}N(n,p)^{14}C$	^{14}C,% from $^{13}C(n, \gamma)^{14}C$
		m_{samp}, (g)	^{60}Co (Bq/kg)	Co ×10^{-3} (ppm)	$m_{LSC\ samp.}$, (g)	^{14}C, (Bq/g)			
N1	stack	0.15	2.6±0.1	4±1	0.0777	12±2	60±10	86	14
N2	bushing	0.14	4.7±0.3	14±1	0.0513	17±2	90±10	90	10
N3	sleeve	0.14	51±1	47±1	0.0891	16±2	80±10	89	11
N4	stack	0.15	2.3±0.2	4±1	0.0518	8±1	37±5	79	21
N5	stack	0.13	2.9±0.7	3.4±0.8	0.0790	6±1	25±4	72	28
N6	sleeve	0.14	49±2	45±2	0.0706	15±2	80±10	89	11

3. RBMK-1500 REACTOR MODELING AND ESTIMATION OF THE NEUTRON FLUENCE IN THE REACTOR GRAPHITE BY ISOTOPE RATIO MASS SPECTROMETRY

The activation of ^{14}C in the irradiated graphite of the RBMK-1500 reactor has been modeled previously using different simulation codes [9, 10, 11]. In this research the full scale 3D RBMK-1500 reactor model (MCNP6) with detail materials composition, power history of the reactor, operation time, the most recent graphite impurity concentrations [4] were used for MCNP6 model upgrade. The new ENDF-VIII nuclear library [12], which includes carbon isotopes, was used for detail ^{14}C production pathways estimation. The irradiation parameters – neutron fluence have been checked by estimating stable carbon isotope ratio ($\delta^{13}C$) [as in 8]. The modeling of ($\delta^{13}C$) in the RBMK-1500 spent graphite construction is presented in Table III.



(a) (b)

Figure 1. (a) Cross section of full scale 3D RBMK-1500 reactor model (MCNP6) and (b) RBMK-1500 graphite mass (%) and calculated activities in the different graphite constructions.

Table III. Stable carbon isotope ratio (δ^{13}C) calculation in the RBMK-1500 spent graphite. The ± represent 1σ uncertainties of model uncertainties due to ±10% variation of power at the graphite column.

Graphite construction	Modelled δ^{13}C	Flux, n/cm^2×s
Graphite stack	23.6±0.6	$(1.0\pm0.1)\times10^{14}$
Graph. stack top	29.1±0.1	$(1.4\pm0.2)\times10^{13}$
Graphite stack bottom	29.7±0.1	$(8.4\pm0.8)\times10^{12}$
Graphite sleeve	24.9±0.5	$(9.8\pm0.9)\times10^{13}$
Graphite sleeve top	29.1±0.1	$(1.3\pm0.1)\times10^{13}$
Graphite sleeve bottom	29.7±0.1	$(8.0\pm0.7)\times10^{12}$
Periphery graphite stack	28.6±0.2	$(3.0\pm0.3)\times10^{13}$
Periphery graphite stack top	30.2±0.1	$(4.0\pm0.3)\times10^{12}$
Periphery graphite stack bottom	30.4±0.03	$(2.5\pm0.3)\times10^{12}$
Periphery graphite sleeve	29.0±0.1	$(2.8\pm0.2)\times10^{13}$
Side reflector	29.5±0.1	$(9.5\pm0.8)\times10^{12}$
Side reflector top	30.5±0.03	$(1.4\pm0.2)\times10^{12}$
Side reflector bottom	30.6±0.02	$(1.0\pm0.1)\times10^{12}$
Side cooling reflector	30.5±0.1	$(1.1\pm0.1)\times10^{12}$
Side cooling reflector top	30.7±0.02	$(1.8\pm0.2)\times10^{11}$
Side cooling reflector bottom	30.7±0.02	$(1.4\pm0.1)\times10^{11}$
Control Rod (CR) graphite	22.7±0.8	$(8.9\pm0.9)\times10^{13}$
CR graphite top	29.3±0.1	$(1.1\pm0.1)\times10^{13}$
CR graphite bottom	29.8±0.1	$(7.4\pm0.7)\times10^{12}$
CR graphite sleeve	22.0±0.8	$(8.8\pm0.8)\times10^{13}$

The full 3D of the RBMK-1500 reactor, graphite mass (%) share and calculated ^{14}C activities in the different graphite constructions are presented Fig. 1. The simulated activity of ^{14}C was compared with the measured ^{14}C activity using β spectrometry technique (liquid scintillation counter Quantulus-1220 (*PerkinElmer, USA*) and also with the rapid method of specific activity determination [13]. Results have indicated that production of ^{14}C from ^{14}N in the RBMK-1500 reactor is considerable (60% to 90% depending on ^{14}N initial concentration) and has to be taken into account in order to make proper evaluation of ^{14}C activity in the model. Measured ^{14}C specific activity values in the samples varied from 130-700 kBq/g in the RBMK-1500 irradiated samples. This corresponds to 15±4 - 80±10 ppm impurity of ^{14}N in various graphite samples of RBMK reactor.

The estimation of the real neutron flux in different parts of RBMK-1500 reactor graphite was performed by measurement of the stable carbon isotope ratio (δ^{13}C) in the irradiated graphite samples. The measurements of RBMK spent graphite samples were performed by using the elemental analyzer FlashEA 1112 connected to the stable isotope ratio mass spectrometer ThermoFinnigan Delta Plus advantage. The measurements procedure is presented in [8]. The experimentally determined δ^{13}C values are in range from -20.0 to -25.6 in graphite core samples, while in raw graphite sample δ^{13}C value is about -30.7 (the similar result is obtained for low neutron fluence regions as side cooling reflector top and bottom parts). In the sample from the central zone of the reactor δ^{13}C value of -24.5 is obtained, while in graphite samples from the plateau zone of the reactor it differ from -20.0 to -24.4. In the sample form peripheral zone the δ^{13}C value of -24.1 was found. In summary, the results indicate clear difference between irradiated and non-irradiated raw graphite samples. However, the dependence of δ^{13}C value on the irradiated graphite sample location in reactor core is not found. This indicates that the neutron flux is quite uniform in the RBMK-1500 reactor core. The measured Δ^{13}C/^{12}C values in graphite at different positions of the reactor core indicates that according to modelling results the neutron flux in the samples varies from $9 \cdot 10^{13}$ n/cm^2·s – to $1.5 \cdot 10^{14}$ n/cm^2·s (see Table III for details).

4. CONCLUSIONS

The research on importance of ^{14}C generation channels via ^{13}C and ^{14}N in the virgin and irradiated RBMK-1500 graphite was performed. Obtained results have indicated that production of ^{14}C from ^{14}N in the RBMK-1500 reactor graphite is about ~63% in case of RBMK-1500 neutron flux, and 70-90% in case of LVR-15 neutron flux. We should note what irradiation in the LVR-15 environment (irradiation in the air) corresponds to surface ^{14}N concentration of the spent graphite. Additional experiments are needed for investigation of samples irradiated in the RBMK-1500 reactor environment (with N+He mixture flushing gas) to confirm the ^{14}N concentrations in the bulk of graphite. Measured ^{14}C specific activity values in the different irradiated graphite samples varied from 130-700 kBq/g in the RBMK-1500 irradiated samples and from 3-12.5 Bq/g in the LVR-15 irradiated graphite samples. This corresponds to 25±4 – 90±10 ppm impurity of ^{14}N in various graphite samples of RBMK reactor. Without additional proof if the high ^{14}C activity is imposed by thin surface layer, the RBMK-1500 spent graphite activation seems to be too high for additional treatment or reconsidering for lower category of waste.

ACKNOWLEDGMENTS

The authors would like to acknowledge the INPP personnel for providing the irradiated and virgin graphite samples. The virgin graphite samples were irradiated during INAA experiments performed at the Nuclear Physics Institute of CAS Řež, Czech Republic within the CANAM infrastructure supported by the Ministry of Education, Youth and Sports of the Czech Republic by project No. LM2015056.

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