

About chemical form and binding energy of ^{14}C in irradiated graphite of uranium-graphite nuclear reactors*

Evgeny V. Bespala¹, Alexander O. Pavliuk¹, Vladimir S. Zagumennov¹, Sergey G. Kotlyarevskiy¹

¹ JSC “Pilot and Demonstration Center for Decommissioning of Uranium-Graphite Nuclear Reactors”, 13 Avtodoroga st., bld. 179a, Seversk, 636000 Russia

Corresponding author: Evgeny V. Bespala (bespala_evgeny@mail.ru)

Academic editor: Yuri Korovin ♦ Received 18 September 2018 ♦ Accepted 19 September 2018 ♦ Published 17 October 2018

Citation: Bespala EV, Pavliuk AO, Zagumennov VS, Kotlyarevskiy SG (2018) About chemical form and binding energy of ^{14}C in irradiated graphite of uranium-graphite nuclear reactors. Nuclear Energy and Technology 4(1): 51–56. <https://doi.org/10.3897/nucet.4.29855>

Abstract

Issues associated with handling irradiated graphite of uranium-graphite nuclear reactors are examined. It is demonstrated that selection of approaches, methods and means for handling irradiated graphite are determined by the form of occurrence and binding energy of long-lived ^{14}C radionuclide with graphite crystalline lattice. The purpose of the present study is the determination of possible chemical compounds in which ^{14}C can be found and assessment of fastness of its binding in the structure of irradiated graphite. Indigent and foreign experience of handling graphite radioactive wastes was analyzed, calculations and measurements were performed. Information was provided on the channels of accumulation of ^{14}C in the structure of reactor graphite and it was demonstrated that the largest quantities of the radionuclide in question are generated according to the reaction $^{14}\text{N}(n, p)^{14}\text{C}$. Here, most part of radioactive carbon is generated on ^{14}N nuclei found in the form of impurities in non-irradiated graphite and in the composition of gas used for purging nuclear reactor in the process of operation. ^{14}C radionuclide generated according to $^{14}\text{N}(n, p)^{14}\text{C}$ nuclear reaction is localized in the near subsurface graphite layer (in the near subsurface layer of pores) at the depth of not more than 50 nm. Analysis was performed of possible chemical compounds which may incorporate radioactive carbon. It was established that the form of occurrence is determined by the operational properties of specific graphite element in the reactor core. ^{14}C binding energy in the structure of irradiated graphite was evaluated and depth of its penetration in the structure was calculated. It was established that selective extraction of this radionuclide is possible only under elevated temperatures in weakly oxidizing environment which is explained by the binding energy reaching up to 800 kJ/mole in the process of chemical sorption of ^{14}C on the surface of graphite and depth of its occurrence equal to ~ 70 nm in the course of ion implantation. It was demonstrated that radioactive carbon generated according to $^{13}\text{C}(n, \gamma)^{14}\text{C}$ nuclear reaction is uniformly distributed among graphite elements and possesses binding energy ~ 477 kJ/mole. Its selective extraction is possible only under the condition of destruction of graphite crystalline lattice and organization of the process of isotopic separation. The obtained results allow recommending the most efficient methods of handling irradiated graphite during decommissioning uranium-graphite reactors.

Keywords

Uranium-graphite reactors, irradiated graphite, binding energy, binding strength, radionuclide, radioactive carbon, processing decontamination.

* Russian text published: Izvestiya vuzov. Yadernaya Energetika (ISSN 0204-3327), 2017, n.4, pp. 127-137.

Introduction

^{14}C generated and accumulated along several independent channels under the effects of natural and technogenous factors is one of long-lived radionuclides which are the most widely spread in nature. Interaction of neutron cosmic radiation with light nuclei refers, first of all, to natural factors. The rate of generation of radioactive nuclei of ^{14}C depends on the neutron flux density and is estimated to be equal to $2.5 \text{ atom}/(\text{s}\cdot\text{cm}^2)$. In this case the largest amount of this radionuclide is generated at the height of 12 km above the sea level (Rublevskij et al. 2004).

Nuclear tests and operation of nuclear fuel cycle facilities refer to technogenous factors of generation of ^{14}C . Here, the latter factor plays the decisive role in the accumulation of this radionuclide. As much as 250 000 tons of radioactive graphite wastes (RW) were accumulated in Russia and abroad during the period of existence of nuclear power industry. Mass of irradiated graphite per one power unit reaches from 800 to 2500 t. Here, specific activity of ^{14}C in irradiated reactor graphite varies within the range of $10^4 - 10^6 \text{ Bq/g}$.

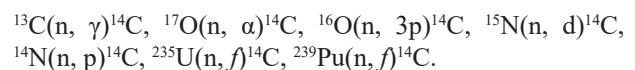
Class of graphite RW generated during decommissioning uranium-graphite nuclear reactors is determined by the quantity (activity) of the most long-lived ^{14}C radionuclide. Cost of final disposal of graphite RW is directly dependent on the RW class and, therefore, on the concentration of radioactive carbon. That is why reduction of cost of handling graphite RW is possible under the condition of successful resolution of the task of selective extraction of ^{14}C .

Selection of approaches, methods and technical means of handling irradiated graphite is determined, apart from everything else, by the form of occurrence and binding energy of ^{14}C . However, this issue is the subject of much controversy among both Russian (Kashev et al. 2013, Bushuev et al. 2015) and foreign (Dunzik-Gougar and Smith 2014) scientists. It is associated with the difference in specific activity, conditions of generation and the nature of distribution of ^{14}C in irradiated graphite which is predetermined by the difference of physical and chemical properties of non-irradiated graphite, parameters of nuclear reactor operation and history of irradiation. The purpose of the study is the analysis of possible chemical compounds in which ^{14}C can be found and evaluation of strength of its bonding in the structure of irradiated graphite.

Channels of ^{14}C accumulation in irradiated graphite

The largest quantities of ^{14}C are generated in the cores of uranium-graphite nuclear reactors of PUGR (production uranium-graphite reactors), RBMK, AM and AMB types. In this case specific features of operation of graphite stack are the determining factors from the viewpoint of accumulation of ^{14}C . For the purposes of prevention

of oxidation and cooling down of graphite internal volume of the reactor is purged with high-purity nitrogen and nitrogen-helium mixture circulated through independent closed loop under pressure close to atmospheric pressure. Besides the above, penetration of air containing nitrogen into the reactor purging system due to infiltration environment, as well as nitrogen present in the purging mixture because of the impossibility of its complete purification is possible. Because of the operational features the number of independent channels of ^{14}C accumulation in graphite elements of uranium-graphite reactor (UGR) is different. The following nuclear reactions can be attributed to the main nuclear reactions resulting in the generation of radioactive carbon:



Generation of ^{14}C from stable ^{13}C isotope is typical for all types of UGR graphite stacks but, however, the fraction of ^{14}C generated according to this channel is insignificant. Firstly, maximum possible quantity of this radionuclide is limited by the initial concentration of stable isotope which is equal in natural mixture of isotopes to $\sim 1.1\%$. Secondly, cross-section of $^{13}\text{C}(n, \gamma)^{14}\text{C}$ reaction amounts to approximately 0.9 mbarn, and, notably, absorption of thermal neutrons predominantly takes place. This results in the uniform generation of ^{14}C among graphite elements. Calculated value of ^{14}C generation rate according to the reaction channel under examination here is equal for RBMK-1000 reactor to $1.4 \cdot 10^{10} \text{ Bq/day}$ per one graphite stack (Rublevskij et al. 2004). After 43 years of continuous operation of such reactor specific activity of ^{14}C generated according to $^{13}\text{C}(n, \gamma)^{14}\text{C}$ reaction channel in the graphite stack will be equal to $\sim 8 \cdot 10^4 \text{ Bq/g}$.

The largest quantity of ^{14}C in reactor graphite is generated according to $^{14}\text{N}(n, p)^{14}\text{C}$ reaction because of high value of cross-section of neutron interactions with ^{14}N nuclei ($\sigma = 1800 \text{ mbarn}$). This nuclear reaction is possible because nitrogen is present in the non-irradiated graphite in the form of micro-impurities or is present in the composition of gases in the purging mixture used in the process of operation of nuclear reactor. In the first case microscopic impurities are present in graphite because of the impossibility of complete purification of graphite preforms in the process of their production. Despite long-term calcination in special kilns at temperatures of $2300 - 3000^\circ\text{C}$ and processing in aggressive environment (for instance, in chlorine or in difluorochloromethane) (Virgil'ev et al. 2006) concentration of nitrogen in non-irradiated graphite is estimated to be equal to 31 – 100 ppm (Sklyar 1984). In the second case ^{14}C is generated from nitrogen nuclei included in the composition of purging gas. Concentration of radionuclide accumulated according to this reaction is difficult to estimate and varies within wide range of 40 – 90% from the total activity of sample of irradiated graphite (Bushuev et al. 2015). It is first of all associated with design features, operatio-

nal modes and neutron physics parameters of a separate UGR.

One of the channels of ^{14}C accumulation in the uranium-graphite nuclear reactor core are nuclear reactions on oxygen nuclei $^{17}\text{O}(n, \alpha)^{14}\text{C}$ and $^{16}\text{O}(n, 3p)^{14}\text{C}$ for which cross-sections of neutron interactions with nucleus are equal to 240 mbarn and 2.2 mbarn, respectively. Activation of oxygen found in the purge gas, in coolant and in nuclear fuel (for nuclear reactors of RBMK type where ceramic fuel in the form of uranium dioxide UO_2 is used) takes place as a rule. Oxygen can be present in reactor graphite in the form of compounds with carbon penetrating graphite in the process of its production.

In the process of graphitization during production of nuclear graphite carbon atoms having free bonds on the discontinuity boundary can combine with O_2 which is not completely removed during purification.

It is worth mentioning that ^{14}C can be generated during ternary fission of ^{235}U and ^{239}Pu nuclei which can penetrate graphite stack in case of loss of hermiticity of cladding of fuel elements caused by drainage of water from pressure channel and disturbance of heat removal regime. Approximately $9.1 \cdot 10^{-7}$ and $3.1 \cdot 10^{-7}$ ^{14}C nuclei are generated in this case per one act of fission of ^{235}U and ^{239}Pu , respectively (Frolov et al. 2004). However, concentration of radioactive carbon accumulated according to $^{235}\text{U}(n, f)^{14}\text{C}$ and $^{239}\text{Pu}(n, f)^{14}\text{C}$ reaction channels does not exceed 0.01% of total activity of ^{14}C contained in the graphite element.

Thus, nuclear reactions $^{13}\text{C}(n, \gamma)^{14}\text{C}$, $^{14}\text{N}(n, p)^{14}\text{C}$, $^{17}\text{O}(n, \alpha)^{14}\text{C}$ are the main channels of accumulation of ^{14}C radionuclide in elements of UGR graphite stack.

Determination of form of occurrence of ^{14}C in irradiated graphite

The question of the form of occurrence of long-lived ^{14}C radionuclide in irradiated graphite is the most important from the viewpoint of selection of the methods of handling during decommissioning of UGR reactors. It is connected with the fact that carbon can form chemical bond with practically all elements of periodic table. Moreover, it is included in the composition of all organic compounds which are capable to enter biological chains thus representing hazard to human health.

^{14}C radionuclide generated according to $^{13}\text{C}(n, \gamma)^{14}\text{C}$ reaction has, probably, strong bond with other atoms of crystalline lattice and is practically immune against selective separation without destruction of graphite crystalline lattice. However, position of ^{14}C in the lattice in the case of reaction on nuclei of stable isotope ^{13}C will be determined by the energy of absorbed neutron. This is associated with the fact that fast (damaging) neutrons energy of which amounts, on the average, to 2 MeV are capable to displace atoms from the points of crystalline lattice due to elastic interactions (Trevethan et al. 2013). In this case proba-

bility of absorption of such neutron by ^{13}C nucleus with formation of ^{14}C nucleus is very low because of relatively small cross-section of the interaction ($\sim 10^{-5} - 10^{-6}$ barn). Resonance absorption resulting in the appearance of ^{14}C in the points of crystalline lattice is the exception. With moderation of neutron the value of interaction cross-section increases which also leads to the accumulation of radioactive carbon isotope. However, energy of moderating neutron may be sufficient for displacing ^{13}C from the lattice point and for subsequent interacting with it with formation of ^{14}C (Fig. 1). Here the newly formed radionuclide cannot penetrate the potential barrier and escape metastable state and positions itself in the lattice in the form of interstitions (for instance, in the form of Frenkel pair) retaining certain excess energy as compared with atom in equilibrium state. Therefore, ^{14}C generated according to $^{13}\text{C}(n, \gamma)^{14}\text{C}$ channel can be situated either in the points of crystalline lattice or in the space between lattice points migrating under the effects of external factors.

Emission of proton by excited ^{15}N nucleus and creation of ^{14}C nucleus with recoil energy $\sim 41,1$ keV take place in case of ^{14}C formation according to $^{14}\text{N}(n, p)^{14}\text{C}$ neutron reaction (Pageot et al. 2016). This energy is sufficient for breaking any chemical bonds and ionization of surrounding gaseous medium. Here, formation of chemically active centers consisting of excited atoms which can enter chemical reactions with decelerating recoil nucleus is possible. In the case of presence in the gaseous medium of molecular oxygen or water vapors (because of infiltration from the environment) oxidation reactions with formation of ^{14}CO or $^{14}\text{CO}_2$ are the most probable (Nefedov et al. 1960). However, for formation of such compounds it is necessary, first of all, that ^{14}C collides with O_2 and H_2O molecules and, secondly, particles must possess energies larger than the activation energy. Since decelerating nucleus forms compounds already at energy of ~ 20 keV then the rate of accumulation of ^{14}CO or $^{14}\text{CO}_2$ will be determined exclusively by the concentration of oxygen (because of increased frequency of molecular collisions).

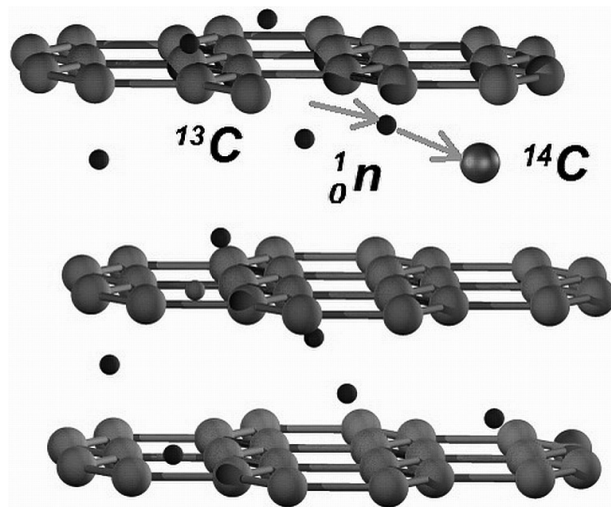


Figure 1. Layout of formation of ^{14}C positioned between graphite crystalline lattice planes.

Incipience of carbon-containing compounds more complex than carbon oxides is also possible. For example, results of studies on the determination of form of occurrence of ^{14}C by selective extraction of different functional groups from the surface of irradiated graphite of NBG-18 and NBG-25 brands are presented in (Vulpus et al. 2013). The radionuclide in question can be found in the following forms: carboxyl group, lactones (organic anhydrides), phenols, carbonyls, anhydrides, ethers, etc.

Results obtained during studies of samples of irradiated graphite using time-of-flight mass-spectrometer demonstrate that ^{14}C can also be found in the following compounds: $^{14}\text{C}(\text{N})$, $^{14}\text{CH}(\text{NH})$, $^{14}\text{CH}_2(\text{NH}_2)$, $^{14}\text{CC}(\text{CN})$, $^{14}\text{C}-^{14}\text{C}(\text{N}_2)$, $^{14}\text{C}-^{14}\text{CH}(\text{N}_2\text{H})$, $^{14}\text{CO}(\text{NO})$, ^{14}CCN , $^{14}\text{C}-^{14}\text{CC}(\text{CN}_2)$, $^{14}\text{CCO}(\text{CNO})$, $^{14}\text{CO}_2(\text{NO}_2)$ (LaBrier and Dunzik-Gougar 2015). Analysis of registered peaks is complicated because of the difficulty of identification of ^{14}C and ^{14}N in different molecules. Therefore, uncertainty emerges in the establishment of form of occurrence of radioactive carbon in different compounds consisting of oxygen, nitrogen and hydrogen.

^{14}C compounds formed during ternary fission of heavy nuclei according to (n, f) reaction are of interest. Penetrating graphite stack fragments of nuclear fuel enter as the result of effects of multiple factors in chemical interactions with graphite, water vapors and with air. Spilled fuel was irradiated during extended period in the reactor stack which facilitated accumulation of transuranic elements and fission products which contained ^{14}C . As the result of accidents radioactive carbon can be found in the following forms: UC_2 , UC , U_2C_3 , PuC_2 , PuC , Pu_2C_3 (Barbin et al. 2015). However, concentration of such compounds in the graphite stack does not exceed 0.01% of the total quantity of compounds composition of which includes ^{14}C because of low probability of the process of ternary fission of ^{235}U and ^{239}Pu .

Binding energy of ^{14}C in the structure of irradiated graphite

Question of binding energy of ^{14}C radionuclide in graphite structural elements is important from the viewpoint of selection of the method for decontamination, re-processing or utilization of irradiated nuclear graphite of UGR reactors. The character of bond of the radionuclide in question will be first of all determined by its chemical form. However, the history of exposure of the selected element of graphite stack and the channels of formation of ^{14}C in it are of not insignificant importance as well.

^{14}C radionuclide formed in $^{13}\text{C}(n, g)^{14}\text{C}$ nuclear reaction on thermal neutrons and found in the graphite crystalline lattice is rigidly bound with adjoining atoms. Here, every carbon atom forms in the crystalline lattice of irradiated graphite bond with three other atoms and is found in the state of sp^2 -hybridization. Binding energy of ^{14}C in graphite crystalline lattice is close to binding energy of stable isotopes which amounts on the average to 477 kJ/

mole (Zhou et al. 2006). However, energy of atomic oscillations in the lattice changes in the transformation of ^{13}C into ^{14}C and, as the consequence, insignificant change of binding energy of ^{14}C takes place.

Rupture of chemical bonds between nitrogen atoms because of high recoil energy of the formed radioactive carbon nucleus occurs in the formation of ^{14}C according to $^{14}\text{N}(n, p)^{14}\text{C}$ nuclear reaction. Probably the recoil energy is transformed into the energy of thermal motion of ^{14}C which is decelerated during its propagation through the purging gas. Compounds containing ^{14}C thus formed can be held on the surface of irradiated graphite under the effects of Van der Waals forces (Golkarian and Jabbarzadeh 2013). The cause of this is the adsorption of gas of compounds (Fig. 2). Surface of nuclear graphite (including surfaces of pores) is the adsorbent, purging gas or its compounds with ^{14}C are the adsorbative and any chemical compound described above is the adsorbate. Quantity of adsorbed ^{14}C is determined in the simplest case from equation of Langmuir isotherm and is dependent on thermal hydraulic parameters of reactor operation as follows:

$$a = a_v P / (K + P), \quad (1)$$

where a_v is the maximum concentration of the substance (maximum adsorption); P is the partial pressure of the gas; K is the adsorption equilibrium constant.

During physical adsorption binding energy of ^{14}C compounds on the surface of graphite amounts to (4 – 72) kJ/mole and depends on the form of occurrence of the radionuclide. During operation of graphite stack in normal operation mode specific energy of thermal movement of purging gas varies within the range of 3.6 – 10 kJ/mole depending on the type of UGR. In extreme or emergency conditions this value can reach 15 kJ/mole. Therefore, part of ^{14}C compounds adsorbed on the surface of graphite is carried away by purging gas. Only molecules located deeply inside the pores of reactor graphite (as a rule, inside closed pores) where energy of thermal movement of molecules of purging gas does not exceed the binding energy remain on the surface.

Chemisorption characterized with increased binding energy and depth of penetration of ^{14}C takes place in case

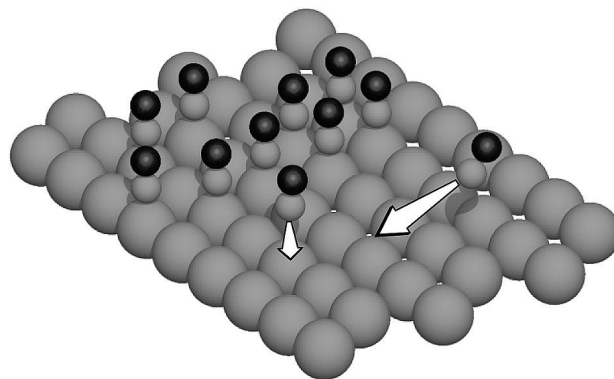


Figure 2. Adsorption of ^{14}C compounds on the surface of irradiated nuclear graphite.

of aggravation of adsorption by chemical interaction between compounds containing ^{14}C and graphite surface. This results in the irreversibility of the sorption process during operation of graphite stack of nuclear reactor and to the change of structure of adsorbate and adsorbent. Following the Zeldovich theory of chemical reaction rate on porous or powder-like substances the depth of penetration of ^{14}C radionuclide is reversely proportional to the square root of reaction rate constant (Zeldovich 1939) which, in turn, depends on the pressure and temperature. Since graphite works in the reactor at high temperatures, surface chemical reactions impossible under standard conditions take place. Energy of bond between compounds containing ^{14}C and surface of irradiated graphite varies during chemisorption from 260 to 800 kJ/mole depending on the form of occurrence of the radionuclide (Zeldovich 1939). Depth of contamination of irradiated graphite with radioactive carbon formed from nitrogen used during purging the graphite stack can be determined from the following equation:

$$D \frac{d^2c}{dx^2} = K_r S f(c), \quad (2)$$

where D is the effective diffusion coefficient depending on the number and diameter of pores in irradiated graphite; c is the concentration of contaminant at depth x ; K_r is the rate constant of chemical reaction per unit surface; S is the specific surface per unit volume of porous substance; $f(c)$ is the function of chemical reaction rate dependent on the reaction order.

Calculations were performed using equation (2). Taking into account specific features and time of exposure of different graphite elements in the UGR maximum depth of penetration of ^{14}C formed from nitrogen participating in the purging of graphite stack amounts in replaceable parts to $\sim 10 - 20$ nm and that in graphite blocks is not more than 50 nm. Estimation was performed for chemical compounds represented in the study.

Absolutely different process leading to the accumulation of the radionuclide in question in the near subsurface layer can occur on the surface of irradiated graphite along with adsorption of ^{14}C compounds formed on nitro-

gen nuclei participating in purging graphite stack (Fig. 3). Nuclear reaction with emission of proton and formation of radioactive carbon with recoil energy equal to 40 – 60 keV which is implanted in the graphite structure occurs during absorption of neutron by ^{14}N nucleus (Pageot et al. 2016).

During long time this channel of accumulation was not examined during estimation of quantity and depth of implantation of ^{14}C in irradiated graphite. However, since recent time it attracts more and more attention because it was proven that up to 80% of radioactive carbon in the near subsurface layer of graphite is formed as the result of implantation (Dunzik-Gougar and Smith 2014). This is especially important for indigenous reactors of PUGR and RBMK types where purging is performed by nitrogen of nitrogen-helium mixture. Depth of implementation of ^{14}C in the graphite crystalline lattice can be estimated using the following expression (Anischik and Uglov 2003):

$$R = \frac{1}{N} \int_0^E \frac{dE}{S_e(E) + S_n(E)}, \quad (3)$$

where N is the density of ^{14}C atoms per unit volume; E is the energy of ^{14}C atom; S_n is the cross-section of nuclear (elastic) deceleration; S_e is the cross-section of inelastic deceleration; R is the depth of implantation.

Quantity of ^{14}C along the depth of irradiated graphite can be estimated using Gaussian function knowing the depth of implantation R and assuming that distribution of implanted ions is symmetrical:

$$N(x) = \frac{D \cos \theta}{\sqrt{2\pi\Delta x}} \exp\left(-\frac{(x - R \cos \theta)^2}{2\Delta x^2}\right), \quad (4)$$

where D is the integral flux of ^{14}C per unit surface of graphite (including pores); E is the energy of ^{14}C atom; x is the distance from external surface of graphite to the place of implantation; θ is the angle between the direction of ^{14}C implantation and normal to the surface of graphite.

Maximum depth of ^{14}C implantation in the graphite crystalline lattice of UGR reactors of PUGR and RBMK types amounts to 55 – 70 nm taking into account the operational features of these reactors and with $S_e = 585 - 730$ keV/ μm , $S_n = 10 - 15$ keV/ μm . For foreign reactors of UNGG type this value amounts to ~ 0.9 nm. In this case ^{14}C is found in the form of defects and interstitions between points and planes of crystalline lattice.

Conclusions

It was demonstrated in the study that long-lived ^{14}C radionuclide is mainly formed in irradiated graphite according to the following three independent channels: $^{13}\text{C}(n, g)^{14}\text{C}$, $^{14}\text{N}(n, p)^{14}\text{C}$, $^{17}\text{O}(n, a)^{14}\text{C}$. This explains its heterogeneous distribution in the graphite structure. Here,

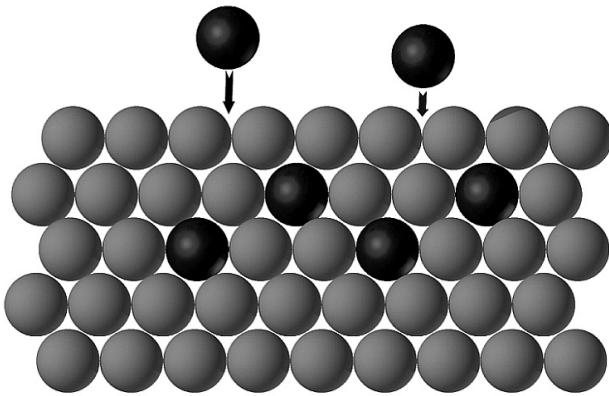


Figure 3. Process of ion implantation of ^{14}C in the crystalline structure of reactor graphite.

most part of radioactive carbon is formed on ^{14}N nuclei which are present in non-irradiated graphite in the form of microscopic impurities and in the composition of gas used for purging graphite stack in the process of reactor operation. ^{14}C radionuclide formed according to $^{14}\text{N}(n, p)^{14}\text{C}$ nuclear reaction is localized in the near subsurface layer of graphite (in the near subsurface layer of pores) at the depth of not more than 50 nm and possesses binding energy with graphite up to 800 kJ/mole which is explained by the processes of adsorption and chemisorption. However, selective extraction of this radionuclide is possible only at elevated temperatures in weakly oxidizing environment. In case of ion implantation ^{14}C is found at the depth of not more than 70 nm and complete removal of contaminated layer is required for its extraction. Besides that, radioactive carbon formed according

to $^{13}\text{C}(n, \gamma)^{14}\text{C}$ nuclear reaction is uniformly distributed over irradiated graphite elements and possesses binding energy equal to ~ 477 kJ/mole. Its selective extraction is possible only after destruction of crystalline lattice and organization of isotope separation process.

Results of experimental studies of processing the surface of irradiated graphite taken from PUGR and RBMK reactor stacks in the environment consisting of argon and oxygen concentration of which was lower than stoichiometric quantity were presented earlier (Pavliuk et al. 2017, Kashcheev et al. 2017, Bespala et al. 2016). Intensive extraction of ^{14}C from the near subsurface layer occurred without significant loss of mass of the sample at temperature equal to $\sim 850^\circ\text{C}$. This confirms the assumption on the form of occurrence and the character of ^{14}C bond in irradiated graphite.

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