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Research

EFFECT OF OPERATING PARAMETERS ON FORMATION PROCESS OF DEFECTS IN MODERATOR BLOCK IN GRAPHITE REACTORS

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ABSTRACT

Analytical scheme of defect formation process was designed, describing temporal dynamics of number of atoms in lattice nodes as well as point and complex defects. According to this scheme, a system of differential equations was made. Analysis of solution of the equation system and experimental data on stored energy (Wigner energy) for industrial graphite reactors allowed to determine the dependence of constant recombination of point defects on irradiation temperature. Calculated and experimental asymptotes of Wigner energy dependence on irradiation time of graphite were compared.

INTRODUCTION

Extension of lifetime of operating graphite reactors and reactor decommissioning are of great importance nowadays. Solving these problems requires correct estimations of reactor graphite life-time as well as energy stored in it (Wigner energy).

Neutron irradiation of graphite alters its properties because of damaging its lattice structure. During moderation process neutron energy is transferred to atoms of carbon which can be dislocated in the lattice relative to the initial location. Many of these shifted (primary knocked out) atoms having high kinetic energy can induce displacement of other atoms, slow down, etc. For example, neutron with 1 MeV energy can induce up to 900 atom displacements losing its energy till the energy of thermal neutron. To displace an atom in graphite lattice it is required about 25 eV of energy. Many displaced atoms immediately return on vacant places. A large number of atoms occupy intermediate position. This can be individual atoms as well as groups of atoms. They have significant impact on numerous properties of the material and this significance depends on dose and temperature of irradiation (Virgilev, 2001).

Temperature – one of the main factors affecting the degree of radiation damage in materials' structure (Glushkov *et al.*, 1985; Baybakov *et al.*, 2015; Nesterov *et al.*, 2013; Avdohin *et al.*, 2013). Neutron bombardment leads to formation of point defects, the fate of which is determined by temperature conditions. Migration of defects to sinks, annihilation of Frankel pairs, formation of complexes and other diffusion processes are related with temperature. The number of initially knocked out atoms at the moment of interaction of radiation with the material at low and high temperatures is practically the same, however, mobility of these atoms at high temperature is higher, i.e. they annihilate faster. This leads to decrease of defects' concentration and therefore to lesser change of properties during irradiation.

Decrease of irradiation temperature and attendant gamma-radiation flux density in the range 100–300°C due to decrease of thermal and radiation gamma-annealing leads to increase in concentration of defects and consequently to decrease of critical neutron fluence (Avdohin *et al.*, 2013; Karpuhin *et al.*, 1997). Critical fluence – is the fast neutron fluence that enables polycrystalline graphite to compensate shrinkage. The value of this fluence defines the lifetime of reactor graphite and limit concentration of defects because during further growth of fast neutron

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fluence thermal-physical and strength properties of graphite deteriorate abruptly.

In higher temperature range (higher 300°C) prevail much more complex defects than point defects. These complex defects have little effect on lattice parameters but take part in formation of additional basal planes in the graphite irreversibly changing form of graphite crystallites (Karpuhin *et al.*, 1997; Golovatsky *et al.*, 2011; Baybakov *et al.*, 2015; Golovatsky *et al.*, 2011; Nesterov, 2013).

Nowadays research in the field of radiation damage of reactor graphite is mainly experimental.

MATERIALS AND METHODS

This paper sets the goal of analytical description of damaging and recovery process of the crystal structure of reactor graphite to determine the dependence of Wigner energy on the fluence of damaging neutrons.

Formulation of the problem requires selecting 3 types of atoms in graphite structure:

- Atoms present in the lattice. Its concentration is denoted *N*.
- Atoms relating to point defects – N_t.
- Atoms creating complex defects – N_c.

The sum of these atoms represents the concentration of all atom types and is defined by the relation:

$$N_{\text{Ô}} = \frac{N_a \cdot \rho}{M} = N + N_t + N_c \qquad (1)$$

where M – molar mass of carbon; ρ – density of reactor graphite.

The problem is formulated using the following approximations:

- Process of damaging of the graphite does not lead to significant change of specific volume.
- Number of point defects formed during the interaction of neutron with nuclei of carbon is equal for atoms present in the lattice and for atoms forming complex defects. The number of defects formed per neutron with energy of 1 MeV is value of 10^3 order.
- Recombination constant of point defects λ is a sum of constant fraction of point defects returning to the initial state (recombining) λ_a and constant fraction of defects that are transformed into complex defects λ_c. The recombination constant of point defects is defined by the relation:

$$\lambda\left(T,\text{Ö}_{\gamma}\right) = \lambda_a\left(T,\text{Ö}_{\gamma}\right) + \lambda_c\left(T,\text{Ö}_{\gamma}\right) \qquad (2)$$

where all the values depend on equivalent irradiation temperature (T) and on attendant gamma-radiation flux density (Φ_γ).

The scheme of defect formation process for graphite is shown in Fig. 1 and is described as follows:

Change of the number of atoms in the lattice (dN / dT) is affected by two competing processes: the first one – increase due to recombination process of point defects (transition of atoms of point defects to the lattice nodes – $\lambda_a N_t$); the second one – decrease due to interaction of neutron flux with the lattice (knocking out of atoms from the lattice and formation of point defects – $\text{Ô}\sigma_s nN$).

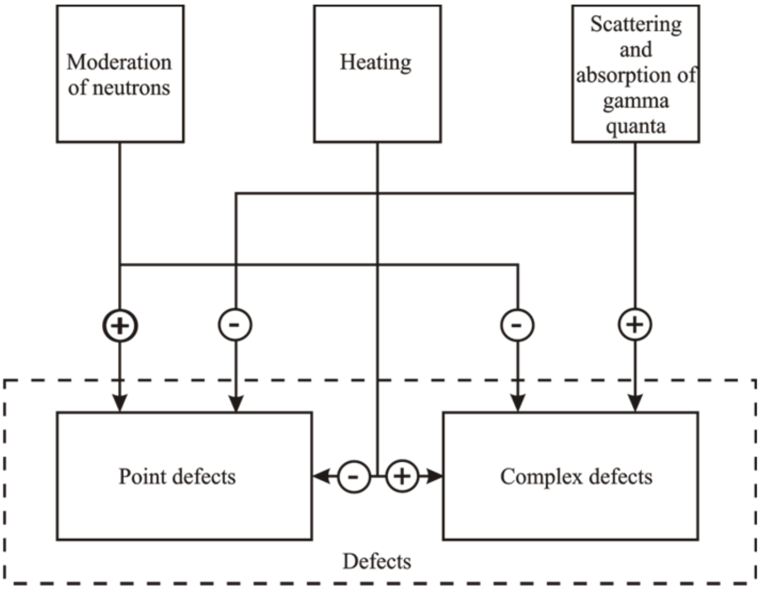

Fig. 1 Scheme of defect formation.

Change of the number of atoms forming complex defects ($d\mathrm{N}_c / \mathrm{dT}$) is affected by two competing processes: the first one – increase due to transition of atoms of point defects into complex defect ($\lambda_c \mathrm{N}_t$); the second one – decrease due to effect of neutron flux on complex defects (transition of atoms of complex defects into point defects – $\text{Ô}\,\sigma_s n N_c$).

Change of the number of atoms of point defects ($d\mathrm{N}_t / \mathrm{dT}$) is affected by two competing processes: the first one – decrease due to recombination process of point defects and its transition into complex defects (λN_t); the second one – increase due to effect of neutron flux on the lattice and complex defects (transition of atoms in the lattice and complex defects into point defects – $\text{Ô}\,\sigma_s \mathrm{n}(\mathrm{N}+\mathrm{N_c})$.

Thus, the system of differential equations describing the change of the number of atoms in the lattice as well as change of complex and point defects is as follows:

$$\begin{cases} \dfrac{dN_t}{dt} = -\dfrac{dN}{dt} - \dfrac{dN_c}{dt} = \Phi \sigma_s n \left(N + N_c \right) - \lambda_t; \\ \dfrac{dN}{dt} = \lambda_a N_t - \Phi \sigma_s n N; \\ \dfrac{dN_c}{dt} = \lambda_c N_t - \Phi \sigma_s n N_c \end{cases} \tag{3}$$

where n – the number of formed point defect per one act of neutron scattering from nucleus of carbon; t – time; Φ – damaging neutrons flux density; σ_s– scattering microscopic cross-section for the damaging neutrons.

The solution of this system are the following expressions:

$$\begin{cases} N_t = k - m e^{-(\text{Ö}\sigma_s n + \lambda)t}; \\ N_c = N_{0c} e_s^{-\text{Ö}\sigma n t} + \dfrac{\lambda_c k}{\text{Ö}\sigma_s n}\left(1 - e^{-\text{Ö}\sigma n t}\right) + \dfrac{\lambda_c m}{\lambda}\left(e^{-(\text{Ö}\sigma_s n + \lambda)t} - e^{-\text{Ö}\sigma n t}\right); \\ N = N_0 e^{-\text{Ö}\sigma n t} + \dfrac{\lambda_a k}{\text{Ö}\sigma_s n}\left(1 - e^{-\text{Ö}\sigma n t}\right) + \dfrac{\lambda_a m}{\lambda}\left(e^{-(\text{Ö}\sigma_s n + \lambda)t} - e^{-\text{Ö}\sigma n t}\right) \end{cases} \tag{4}$$

where $k = \dfrac{\text{Ö}\sigma_s n N_{\text{Ó}}}{\text{Ö}\sigma_s n + \lambda}$; $m = \dfrac{\text{Ö}\sigma_s n \left(N_{\text{Ó}} - N_{0t}\right) - \lambda N_{0t}}{\text{Ö}\sigma_s n + \lambda} = k - N_{0t}$; N_{0t}, N_{0c}, N_0 – concentrations of atoms of point defects, complex defects and atoms in the lattice at initial time, respectively.

RESULTS

The amount of energy stored in graphite is in direct proportion to the amount of point defects. This allows determining the dependence of recombination constant of point defects on irradiation temperature. Experimental data on Wigner energy for the reactors of Siberian Chemical Combine allowed determining this dependence.

To displace one atom in the lattice it is required around 25 eV of energy (E_d). It can be assumed that the same amount of energy is released when atom returns to vacant place in the lattice. Solution of the system of differential equations (3) defines the ratio for the amount of point defects:

$$N_t = \frac{\text{Ö}\sigma_s n N_{\text{Ó}}}{\text{Ö}\sigma_s n + \lambda}\left(1 - e^{-(\text{Ö}\sigma_s n + \lambda)t}\right) + N_{0t} e^{-(\text{Ö}\sigma_s n + \lambda)t} \tag{5}$$

In this ratio the first term describes increase of the number of point defects during irradiation process; the second term describes decrease of the number of point defects present in graphite at the beginning of irradiation. Thus, for sufficiently long time (about one year) this ratio tends asymptotically. The dependence of Wigner energy per weight unit of graphite on irradiation temperature for $\text{Ö} = 10^{13}\, cm^{-2} s^{-1}$, $\sigma_s = 3.1 \cdot 10^{-24}\, cm^2$, $n = 500 \cdot N_{\text{Ó}} = 5 \cdot 10^{22}\, g^{-1}$ is defined by the relation:

$$E = E_d \cdot N_t = \left(25\ eV\right) \cdot \frac{\text{Ö}\sigma_s n N_{\text{Ó}}}{\text{Ö}\sigma_s n + \lambda(T)} = \frac{7.4 \cdot 10^{-4}}{15.5 \cdot 10^{-9} + \lambda(T)},\ cal./g. \tag{6}$$

Then the problem is reduced to determining the function, which approximates the dependence of recombination constant on temperature $\lambda(T)$. Where in values *E* and *T* are known (experimental data), the end result is shown in Fig. 2.

Exponential dependence $\lambda(T)$ can be excluded from consideration. The dynamics of concentration of point defects vs. irradiation time for different irradiation temperatures and, accordingly, Wigner energy vs. fluence at constant flux density of damaging neutrons can be described by using polynomial and power dependencies of recombination constant on

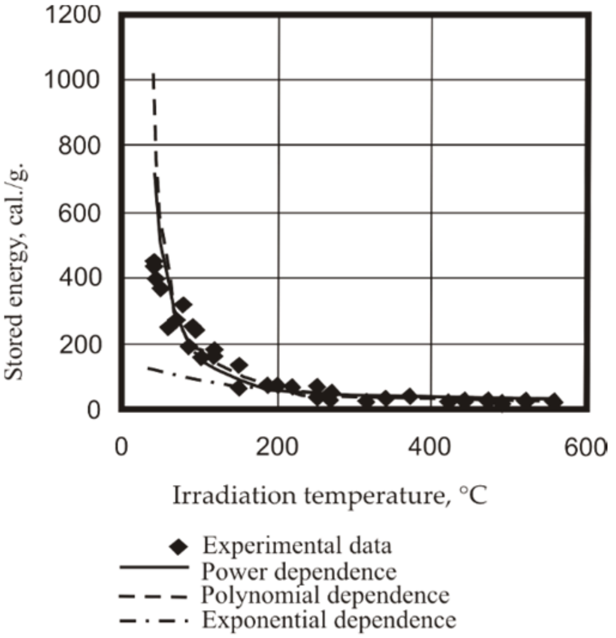

Fig. 2 Dependence of stored energy on irradiation temperature for different approximation functions λ(T).

irradiation temperature. Fig. 3 shows calculated dependence of Wigner energy on the fluence. The dependence is obtained assuming that all neutrons are damaging and irradiation conditions are constant. Experimental dependence of store energy on neutron fluence is shown in Fig. 4.

As seen from results, maximum deviation of the calculated value from the experimental one corresponds to temperature of 260°C which is quite close to the transition region (around 300°C). In this region, one would expect a significant deviation due to uncertainty of the experimental data on critical fluence vs. irradiation temperature dependence.

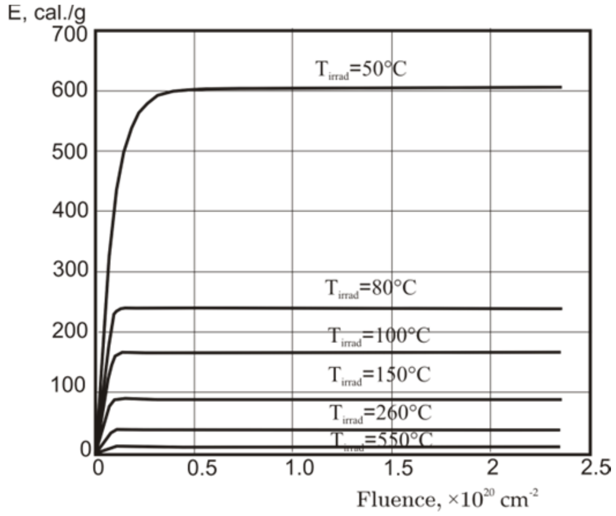

Fig. 3 Calculated dependency of stored energy on the neutron fluence ($\Phi=10^{13}\ cm^{-2}\ s^{-1}$).

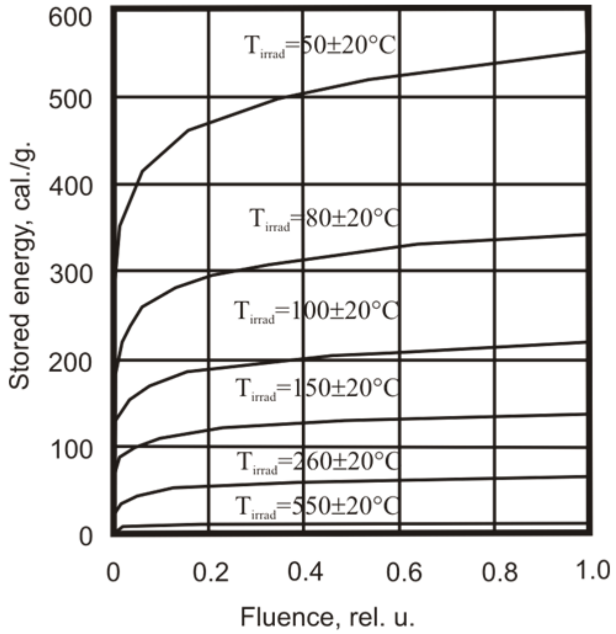

Fig. 4 Experimental dependence of stored energy on the neutron fluence (Tsiganov *et al.*, 2008). The dependence.

CONCLUSION

Thus, the dependence of recombination constant on graphite irradiation temperature at the conditions present in industrial graphite reactors is determined in this paper. To specify the type of functional dependence of critical fluence on irradiation temperature and attendant gamma-radiation flux density it is required to carry out further analysis to select specific function type describing recombination of point defects $(\lambda_a(T,\Phi_\gamma))$ and transition of point defects into complex defects $(\lambda_c(T,\Phi_\gamma))$.

of lattice nodes' concentration on irradiation temperature is in satisfactory agreement with the experimental data obtained by defining the value of stored energy in graphite blocks and reactor core bushings of graphite reactors as a function of irradiation temperature.

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