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Computational and experimental studies of processes of non-activation release of tritium from beryllium-containing materials of research nuclear reactors

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Abstract. Beryllium reflectors in research reactors are known sources of tritium, raising safety concerns associated with its release. While substantial outgassing occurs at elevated temperatures (>700 °C) through diffusion, measurable release has also been observed at ~50 °C, which cannot be explained by this mechanism. A non-activation release process is therefore considered, in which tritium nuclei formed near the beryllium surface escape directly as energetic ions.

A computational model was developed to quantify this process. The stopping range of MeV tritium ions in solids is limited to several tens of micrometers, defining the effective near-surface region for release. Geometric analysis allowed deriving an expression for the number of tritium ions emitted from the surface during irradiation. To verify the model, lithium metatitanate (Li_2TiO_3) pebbles with natural and 96% ^6Li enrichment were irradiated in the WWR-K reactor at ~50 °C for 516 h. Samples were wrapped in 100 μm aluminum foils serving as tritium collectors, which were subsequently dissolved and analyzed by liquid scintillation spectrometry.

Calculated tritium yields were of the order of 10^{17}-10^{18} atoms and showed good agreement with experiment. For natural Li_2TiO_3, $3.35 \cdot 10^{17}$ atoms were predicted compared to $(2.0 \pm 0.4) \cdot 10^{17}$ measured, while for enriched Li_2TiO_3, $2.14 \cdot 10^{18}$ versus $(1.1 \pm 0.2) \cdot 10^{18}$ were obtained. Lower experimental values are attributed to incomplete foil retention and model simplifications such as neglecting pebble self-shielding.

The results confirm the existence of non-activation tritium release from lithium-containing ceramics and demonstrate that the proposed model provides reliable estimates applicable to safety assessments of reactors employing beryllium components.

Keywords: lithium ceramics, neutron irradiation, tritium, helium, MCNP6.

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Introduction

Radiation safety issues of nuclear devices are of crucial importance for minimizing risks to human health and the environment, ensuring stability and efficiency of the nuclear industry. One of the key concernsin this area is the assessment of radioactive materials release into the environment during the operation of nuclear systems, including low-power nuclear reactors with research purposes. In the Republic of Kazakhstan, extensive research has been carried out over the past decades in various fieldsof nuclear and fusion energy, including radioecology andmaterial testing under reactor irradiation conditions [1-10].

This work presents a novel approach to studying non-activation tritium release from beryllium under neutron irradiation, an idea of that emerged from previous studiesof the authors of this paper on accumulation and release of tritium from beryllium-based reflector components of nuclear reactors [11-12].

Under neutron irradiation conditions, tritium is generatedin beryllium through a series of nuclear reactions. These include the formation of lithium-6 within beryllium structures, followed by neutron capture reactions that ultimately result in tritium production:

$$Be^9 + n_f \rightarrow He^4 + He^6, \quad (1)$$

$$He^6 \xrightarrow[t_{1/2} \approx 0{,}85\ s]{\beta^-} Li^6. \quad (2)$$

The resulting lithium interacts with thermal neutrons according to the following reaction:

$$Li^6 + n_{th} \rightarrow He^4 + H^3. \quad (3)$$

The main processes influencing the rate of tritium release from beryllium are diffusion processes, which become significant at high temperatures (above 700 °C) [13,14]. However, a number of studies indicate the release of tritium at temperatures below 50 °C, which cannot be explained by diffusion. A possible mechanism for such tritium release may be the release of tritium directly from the near-surface region of beryllium units of a nuclear reactor.

A detailed consideration of this problem would make it possible to more accurately assess the tritium safety of nuclear reactors containing beryllium units. This would also allow obtaining a method for estimating the amount of tritium ions released from the surface of lithium-containing materials under reactor irradiation conditions.

The study aims to develop and verify a calculation method for estimating tritium flux released from beryllium reflectors without relying on diffusion models. It seeks to experimentally confirm the existence and magnitude of this release mechanism using lithium-containing samples. Ultimately, the work contributes to more accurate assessments of tritium safety in research reactors employing beryllium components.

This work is the first to propose and experimentally confirm a non-activation mechanism of tritium release from beryllium under neutron irradiation, where energetic tritium ions escape directly from the near-surface layer. A new computational method was developed to quantify

this effect, and its predictions were validated against reactor irradiation experiments with lithium ceramics.

Calculation method for assessing the tritium ions amount released from the near-surface layer of materials

It is known that under the influence of neutron irradiation beryllium undergoes the formation and accumulation of radiation-induced defects, as well as helium and tritium nuclei generated as a result of nuclear reactions with beryllium atoms.

The occurrence of defects in beryllium under neutron flux is associated with elastic scattering of neutrons with nuclei and nuclear reactions of the types *(n,2n)* and *(n,α)*[15-17].

For the present study, particular importantance is attached to the reaction (n, α), which takes place at neutron energy E ≥ 0.71 MeV. A lithium-6 atom is formed as a result of reactions (1)-(3), which subsequently interacts with thermal neutrons, leading to the formation of tritium.

The tritium and helium ions with energies E_T and E_{He} (MeV), respectively, interact with the atoms of the material predominantly via inelastic collisions with electrons. Due to Coulomb interactions, energetic ions gradually lose their energy by ionizing or exciting atomic electrons along their trajectory. When the particle has lost all of its energy, it comes to rest. Owing to the relatively large mass of ions compared with that of electrons, individual scattering events lead only to small angular deviations, which statistically compensate each other over numerous collisions. Consequently, the trajectories of tritium and helium ions in the medium can be approximated as nearly rectilinear.

The ion path length λ (m) in the material may be estimated using the ionization loss formula (Bethe-Bloch equation) [18]:

$$-\frac{dE}{dx} = \frac{4\pi n Z^2 e^4}{mv^2} \cdot \left(ln \frac{2mv^2}{I\left(1-\frac{v^2}{c^2}\right)} - \frac{v^2}{c^2} \right) \quad (4)$$

where m is the electron mass; c is the speed of light; v is the velocity of theparticle; $\beta = v/c$; Z is the charge of a particle in units of the positron charge; n is the electron density ofthe medium; Iis the average ionization potential of the atoms in the medium through which the particle passes.

To assess the ionization losses the LISE++ program is usually used, which allows prediction of the energy losses and beam purity of radioactive ions during their transport [19]. For each specific calculation, a program module is used in which the name of the isotope being slowed down, its energy, and the target parameters are set: its density and the stoichiometric composition of atoms.

To describe the emission of tritium and helium ions from the near-surface region of beryllium, a two-dimensional geometric model is employed (Figure 1).

Suppose a number of Rtritium-producing reactions occur at point Alocatedat a distance x from the material surface. The particles formed are emitted isotropically from thispoint.The number of ions released from the material is determined by an isosceles triangle with a side of λ, and in the segment of the angle 2α all ions will leave the material. Their number of escaping ions, F, can be expressed as:

$$F = R \cdot \frac{arccos\ arccos(\alpha)}{\pi} = R \cdot \arccos(x/\lambda)/\pi \quad (5)$$

By integrating this expression over x from 0 to λ, it is easy to obtain expression for the rate N (mol/s) of ions release from the surface layer with an area of S (m^2):

$$N = \frac{R \cdot S}{\pi} \int_0^{\lambda} (arccos(x/\lambda))dx = R \cdot S \cdot \lambda/\pi. \quad (6)$$

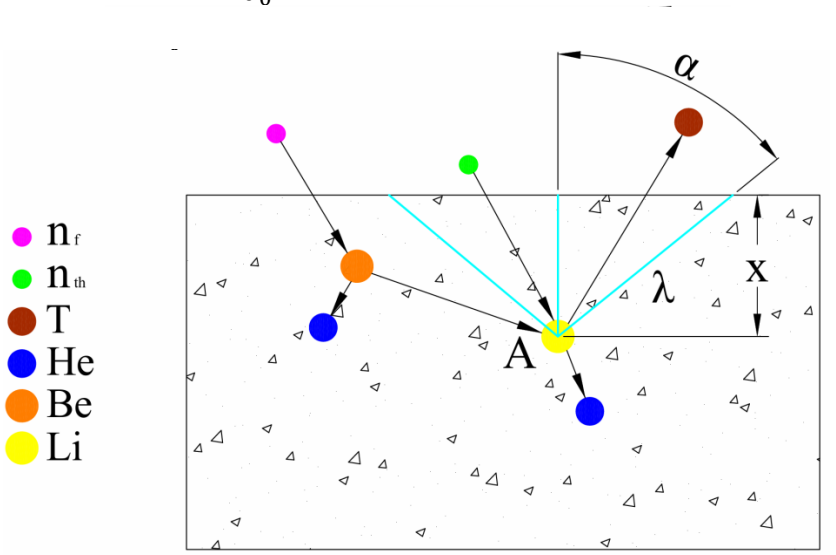

Figure 1. Two-dimensional geometry of the release of tritium and helium atoms from the near-surface layer of beryllium

Moving to integral values for estimating the amount of tritium ions released from the beryllium surface layer with an area of S during irradiation time t, the following expression is obtained:

$$N(t_{max}, S) = \frac{1}{\pi} \int_0^{t_{max}} \int_0^{S} R(S,t) \cdot \lambda\, dSdt \quad (7)$$

This methodology can also be applied to other irradiated materials containing lithium-6.To demonstrate the applicability of the delelpoed approach, the release of tritium ions from the near-surface region of lithium ceramics was experimentally investigated in a dedicated reactor experiment, described below.

Reactor experiment on irradiation of lithium ceramics

In order to obtain samples of aluminum foils that captured tritium released in a non-activating manner two types of lithium ceramics (lithium metatitanate – LMT) pebbles with natural and 96% enrichment in Li6 were exposed to neutron irradiation. It was assumed that tritium ions would be absorbed by the aluminum foil in which the ceramics were wrapped. The

thickness of the foil was about 100 μm, which significantly exceeded the free path of tritium ions in aluminum.

The irradiation consisted in the following: sealed capsules (Figure 2a) with lithium ceramics samples wrapped in aluminum foils were loaded into anampoule device, which was irradiated in the peripheral irradiation channel of the WWR-K research reactor (Figure 3) for 516 hours.

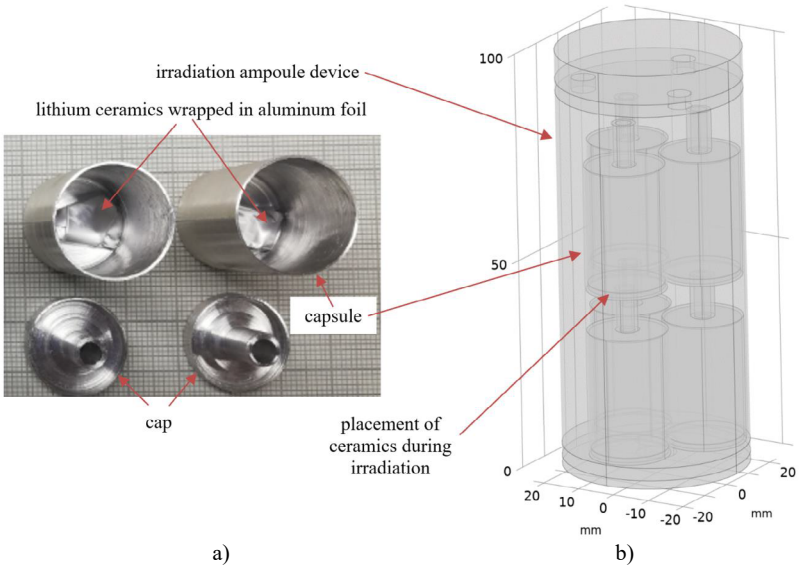

Figure 2. Photograph of capsules with lithium ceramic samples wrapped in foil (a) and irradiation diagram (b)

The capsule arrangement in the reactor ampule device is shown in Figure 2b. Three capsules with LMT (with natural enrichment in Li^6), were installed in the lower part of the ampoule device. The rest three capsules with LMT (96% of Li^6) were installed in the upper part of the ampoule device. The capsules were filled with argon. During the experiments, the ampoule device and the capsules were cooled with water of the first coolant circuit of the WWR-K reactor. Average temperature of coolant did not exceed 50°C. The flow rate of the coolant through the capsule was equal to 0.5 m/s.

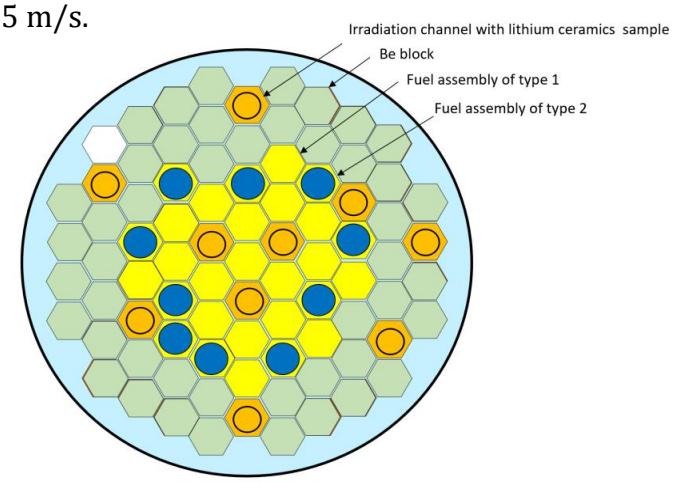

Figure 3. Configuration of the WWR-K reactor core

During irradiation experiment a diagram of the temperature change of the aluminum foil was obtained (Figure 4). As can be seen from graph, the foil's temperature in experiment was less than 50 °C. The temperature values shown on the graph correspond to the average values obtained from three independent temperature sensors measuring the coolant temperature. Since the irradiation capsule is cooled by the water of the reactor's primary circuit both from the outside and from the inside, the temperature inside the capsule is equivalent to the coolant temperature. Temperature fluctuations are associated with daily changes in the environment temperature, which affects the second cooling circuit of the reactor and, accordingly, decreases or increases heat exchange between the first and second circuits of the reactor.

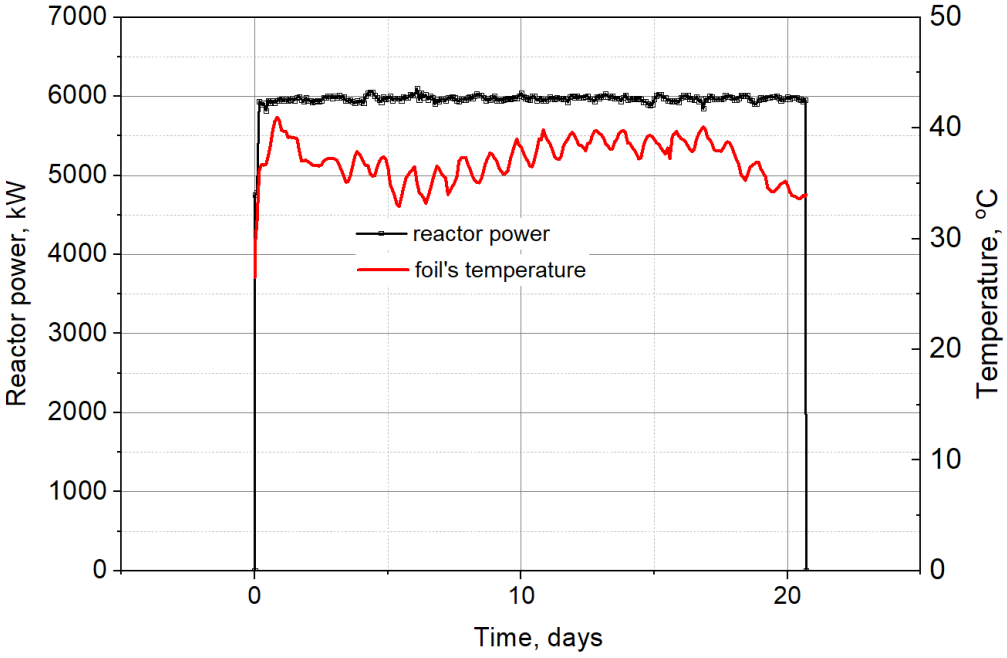

Figure 4. Diagram of the reactor experiment with lithium ceramic pebbles

Tritium amount determination in aluminum foil

After irradiation, the capsules were opened and aluminum foils were extracted. The foils were decomposed in an autoclave with 1 ml of concentrated HNO_3 and 0.5 ml of HF at 180 °C for 2.5 h. The cooled solution was neutralized with Na_2CO_3 to pH 8 and diluted to 50 ml with distilled water, followed by preliminary distillation [20]. From the second distillate fraction (≥20 ml), 1 ml was mixed with Ultima Gold LLT scintillation cocktail (1:19) and stored for 12 h before measurement. Tritium activity was determined by liquid scintillation beta spectrometry (TRICARB-3100TR) for 3 h in the 0-15 keV range with a detection efficiency of at least 60%.

The results for the amount of tritium released in non-activation way (obtained by calculation according to the above method) and the results of the experimentally determined amount of tritium in aluminum foils are given in Table 1.

Table 1. Amount of tritium released by non-activation way during irradiation of lithium ceramics (comparison of calculation and experimental results)

Type of samples wrapped in aluminum foil	Calculated value of the number of tritium atoms that entered the aluminum foil during irradiation	Experimental value of the number of tritium atoms in aluminum foil, determined by liquid scintillation beta spectrometry
Li_2TiO_3(natural enrichment)	3.35·1017	(2.0±0.4)·1017
Li_2TiO_3(96% Li6)	2.14·1018	(1.1±0.2)·1018

The comparison of calculated and experimental results demonstrates good agreement in terms of order of magnitude, with a clear proportionality between the tritium content and the degree of lithium-6 enrichment in the investigated ceramic samples.

At the same time, the experimentally measured number of tritium atoms in aluminum foils was consistently lower than the calculated estimates. This discrepancy is attributed to the incomplete retention of tritium in the foils: part of the released tritium likely penetrated only shallowly into the foil and subsequently escaped during post-irradiation handling and sample preparation. Another possible reason is the simplification adopted in the calculation procedure, where self-shielding effects of the lithium ceramic pebbles under irradiation were not considered. Specifically, formula (7) was applied to the total geometric surface area of the spherical samples, whereas in reality they formed a pebble bed with a mean pebble radius of 1 mm.

Overall, the results confirm that irradiation of lithium-containing ceramics leads to a measurable tritium release, attributable to its non-activation emission from the near-surface region of the material. The proposed computational approach provides estimates that are consistent with the experimental data within the expected accuracy.

Accounting for the discrepancy between experimental and calculated data due to the geometry of the irradiation experiment

Let us consider a configuration close to the experimental arrangement of lithium ceramic pebbles. The pebbles are sparsely arranged with a distance between them approximately equal to two of their diameters. With such an arrangement, the influence of geometry on the activation-free release of tritium becomes especially significant. In this scheme, the pebbles are arranged in one layer on a flat surface, and a sheet of aluminum foil wraps the pebble bed from above, below, and on the sides. Tritium ions formed during the reaction are emitted isotropically with an energy of about 2.73 MeV and a mean free path in lithium ceramics of about ~ 30 μm, depending on the density of the material.

For a triton to be released, it must be born in the ceramics within the range from the surface of the pebbles and have a trajectory direction that does not encounter obstacles to the foil.

In conditions of sparse packing, when there is a significant free space between the pebbles, the possibility of the tritons exiting not only in the vertical direction, but also to the sides, opens up, since the probability of a collision with a neighboring pebble is minimal.

In this case, vertical trajectories provide a contribution due to the fact that the tritons born in the upper hemisphere of the pebble and directed upward, as well as the tritons born in the lower hemisphere and directed downward, can freely reach the foil. Taking into account the isotropy of the emission and the uniform distribution of tritons by depth, the proportion of such vertical trajectories is estimated as

$$Q_1 \approx \frac{1}{2}\cdot\frac{1}{2}+\frac{1}{2}\cdot\frac{1}{2}\approx 0.5,$$

where the first term corresponds to the number of tritons born in the upper hemisphere of the pebble and flying upward, and the second term - to the number of tritons born in the lower hemisphere of the pebble and flying downward.

For lateral trajectories in sparse packing conditions, a significant space between pebbles allows most tritons flying at angles from ~30° to ~150° relative to the vertical to reach the lateral foil without obstacles. The approximate fraction of such an exit can be estimated as Q2≈0.25. Considering that the angular share of lateral directions is approximately half (50%), but only newts born in the near-surface zone and directed strictly sideways will be able to exit, the total coefficient decreases to ~25%. Thus, the total coefficient of triton exit into the foil with sparse pebble packing is calculated as the sum of the vertical and lateral components and is equal to ~0.75.

This means that with a sparse configuration of pebble placement, the efficiency of tritium detection by aluminum foil reaches about 75% of the calculated value, assuming individual wrapping of each pebble. This is significantly higher than with dense packing, where the yield coefficient is limited to 25-55% depending on the bottom coverage. In this regard, when interpreting experimental data and constructing calculation models of non-activation tritium yield, it is necessary to take into account a correction factor reflecting the influence of the geometric configuration of the pebble bed and the availability of lateral trajectories for the release of tritons into the foil.

Calculation of the rate of non-activation tritium release from beryllium blocks of the WWR-K reactor

Based on the proposed method, the rates of tritium release from beryllium blocks of the WWR-K reactor were estimated during its operation at nominal power. The beryllium block in the reactor core is a monolithic hexagon 69.5 cm high. The density of the beryllium block is 1.894 g/cm3. Blocks are located around the fuel assemblies, forming a ring around the fuel part of the core. In Figure 5, the MCNP model of a horizontal section of the WWR-K reactor core is shown, indicating the cell numbers of the beryllium blocks. Since the heating of the coolant and samples during standard irradiation is not significant, the change in the temperature of the coolant and samples was not taken into account in the calculations.

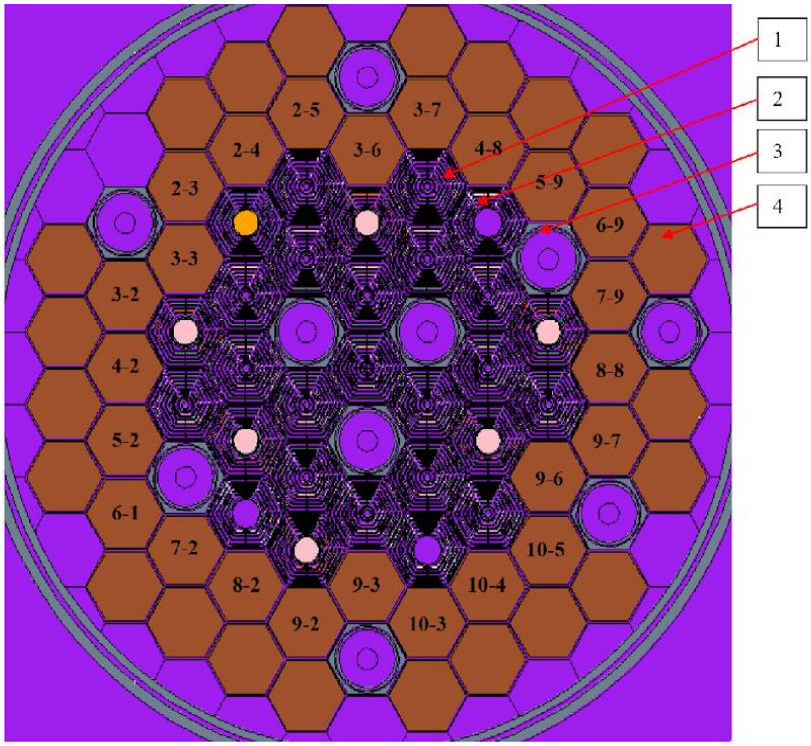

Figure 5. MCNP model of the WWR-K reactor core: 1) fuel assembly of type 1; 2) fuel assembly of type 2; 3) irradiation channel with a displacer; 4) beryllium block

During operation of the reactor at nominal power, some amount of beryllium almost immediately turns into Li^6(according to the reactions (1) and (2)). The densities of beryllium and lithium atoms are described by the following equations:

$$\frac{dN_{Be}}{dt} = -N_{Be}\{RR\}_{Be}, \tag{8}$$

$$\frac{dN_{Li}}{dt} = N_{Be}\{RR\}_{Be} - N_{Li}\{RR\}_{Li}, \tag{9}$$

where, to simplify the notation, the dependence of the numerical density N on time is omitted. The indices Be and Li denote Be^9 and Li^6, respectively. {RR} denotes the rate of isotopic reactions: (n,H^3) for Li^6 and the reaction (n,α) for Be.

{RR} is defined as:

$$\{RR\}_x = \int_0^{20} \varphi(E,t)\sigma_x(E)dE. \tag{10}$$

where x is the name of the isotope; φ(E,t) is the neutron flux in the irradiation position; σ_x (E) is the corresponding neutron-induced cross-section for isotope x. The energy interval (0-20 MeV) covers the entire range of neutron energies in the beryllium block.

To calculate the rate ofLi^6(n,H^3) reaction, a 1 mm area of the outer part of the beryllium block was selected along its entire height. The reaction rate in a unit volume (1/(cm^3s)) of the beryllium block was determined as:

$$RR_V = N_{Li}(0) \cdot N_{Be_0}\{RR\}_{Li}, \quad (11)$$

where N_{Be0})is the initial number of beryllium atoms per unit volume, (at/cm^3); N_{Li} (0) is the number of Li^6 atoms formed in 1 s as a result of the Be(n,α) reaction, (at/cm^3); $\{RR\}_{Li}$ is the rate of the $Li^6(n,H^3)$ reaction per one Li6 atom. The initial number of beryllium atoms in the block N_{Be0})=1.23·1023 at/cm^3.

The reaction rates$\{RR\}_x$ of Be(n,α) and Li6(n, H^3) per one atom of Be and Li^6, respectively, were calculated using the MCNP[6] code and the ENDF/B-VII.1 neutron cross-section library [21]. The MCNP model was built according to the real size of the reactor core with fuel assemblies and structural components. The MCNP input file included 500 cycles consisting of 450 active and 50 inactive cycles with 200,000 histories per cycle. The reaction rate values were normalized to the calculated k_{eff} value and compared with the steady-state power level of the critical system in units of fission neutrons per unit time, which allows us to move from the reaction rate to the neutron flux density. For the WWR-K reactor, for which the nominal power is 6 MW, the scaling factor is:

$$6 \cdot 10^6 MW \cdot 3.467 \cdot 10^{10} \frac{fission}{s} \cdot 2.457 \frac{neutron}{fission} \cdot 0.92 = 4.70 \cdot 10^{17} \frac{neutron}{s},$$

where 0.92 is the fraction of thermal power due to neutrons.

Figure 6 shows the calculated values of Li_6(n, H^3), (1/(cm^{3}s)) reaction rate in beryllium for the outer ring of blocks in the WWR-K reactor core. Calculations showed that the rate of tritium release into the cooling water from all beryllium blocks during reactor operation at nominal power is about ~10-14 mol/s. As can be seen, the tritium flux values determined by the proposed calculation method are quite noticeable and, in general, should be taken into account when operating research nuclear reactors containing beryllium elements in the core design.

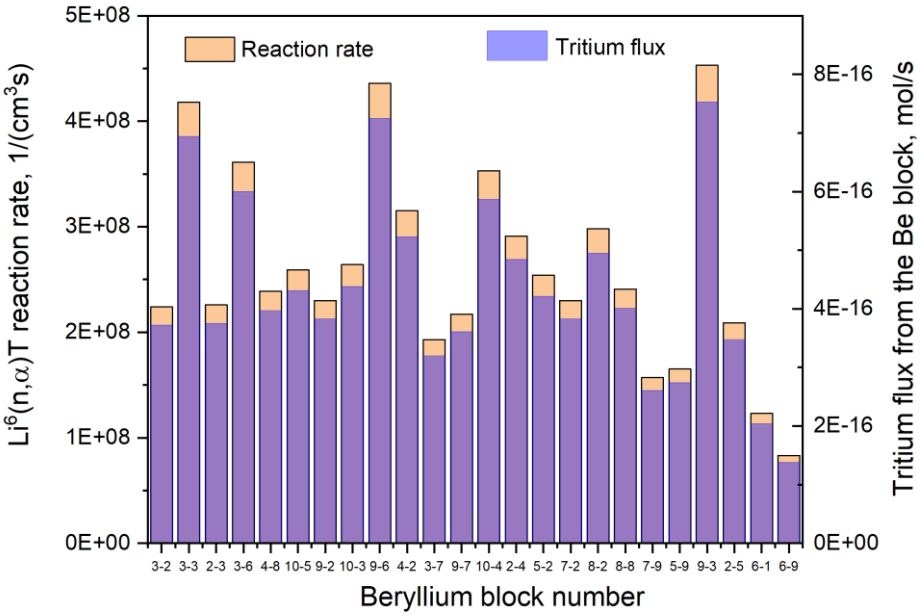

Figure 6. Li^6(n,α)t reaction rates and tritium flux from the beryllium block of the WWR-K reactor

Conclusion

Thus, as a result of the studies of non-activation release of tritium from the surface layer of beryllium under neutron irradiation, a method for calculating the tritium flux from the surface was obtained, which was confirmed in the irradiation experiments with lithium ceramics.

The results of the calculated estimates and experimental values for the amount of tritium released from the lithium ceramic samples reasonably coincided (in order of magnitude). It was shown that the tritium flux from the samples is proportional to the degree of enrichment in lithium-6, and this proportionality is fairly well observed for various ceramic samples. The observed discrepancy between the calculated data and the experiment is due to the research methodology and a simplified calculation scheme that does not take into account the actual geometry of the experiment.

Further, according to the proposed methodology, calculations were made for the non-activation release of tritium from the beryllium rods of the WWR-K reactor. Calculations showed that the rate of tritium release into the cooling water from all beryllium blocks during reactor operation at nominal power is about ~10-14 mol/s.

The obtained results can be used for a qualitative assessment of the tritium safety of nuclear research reactors containing beryllium elements in the reactor core design.

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The contribution of the authors

Kulsartov T.V. – formulated the main research concept, coordinated the design of the study, and supervised the overall scientific direction of the work. He also took part in the critical revision of the manuscript and the approval of the final version.

Temirzhanov A.A. – corresponding author, carried out data acquisition, processing, and preliminary analysis. He was responsible for organizing communication among the co-authors and ensuring the integrity of the data.

Zhanna Zaurbekova – performed experimental measurements and took part in processing and interpretation of the obtained results. Contributed to drafting selected sections of the article and critically reviewing the text.

Asset Shaimerdenov – provided methodological guidance, contributed to the design of the experimental setup, and ensured the correct interpretation of the results in the context of current research in the field.

Saulet Askerbekov – participated in the collection of experimental data, contributed to the optimization of measurement techniques, and assisted in refining the discussion section of the manuscript.

Pavel Kharkin – supervised laboratory resources and instrumentation, ensured the proper functioning of equipment during the experimental campaign, and contributed to the approval of the final version of the manuscript.

Olga Milts – carried out technical support of the experiments, participated in the calibration and control of measurement systems, and contributed to preparing figures and tables for the article.

Darkhan Sairanbayev – contributed to theoretical analysis and interpretation of the obtained spectra, took part in comparison with model calculations, and revised the discussion and conclusion sections.

Inesh Kenzhina – participated in the statistical analysis of the data, contributed to the validation of the results, and critically reviewed the manuscript for consistency and scientific soundness.

Timur Zholdybayev – provided scientific expertise in nuclear reaction mechanisms, contributed to the interpretation of the results within the framework of existing theoretical models, and revised the article with respect to its conceptual and methodological accuracy.

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Зерттеу ядролық реакторлардың құрамында бериллий бар материалдарынан тритийдің активтенбейтін бөліну процестерін эксперименттік зерттеулер және есептеу.

Андатпа. Зерттеу реакторларындағы бериллий рефлекторлары тритийдің белгілі көздерінің бірі болып табылады, бұл оның бөлінуіне байланысты қауіпсіздік мәселелерін туындатады. Тритийдің айтарлықтай бөлінуі әдетте жоғары температураларда (>700 °C) диффузия есебінен жүреді, алайда шамамен 50 °C температурада да өлшенетін мөлшердегі тритий тіркелген, бұл құбылысты диффузиялық механизммен түсіндіру мүмкін емес. Осыған байланысты белсендірусіз

бөліну процесі қарастырылады, мұнда бериллийдің беткі қабатында түзілген тритий ядролары материалдан тікелей жоғары энергиялы иондар түрінде шығады.

Бұл процесті сандық тұрғыдан бағалау үшін есептеу үлгісі жасалды. МэВ энергиялы тритий иондарының қатты денелердегі жүру ұзындығы бірнеше ондаған микрометрмен шектеледі, бұл олардың бөлінуінің тиімді беткі аймағын анықтайды. Геометриялық талдау сәулелену кезінде беттен бөлінетін тритий иондарының санын анықтауға мүмкіндік берді. Үлгіні тексеру үшін табиғи және ^{6}Li бойынша 96% байытылған литий метатитанаты (Li_2TiO_3) шариктері ВВР-К реакторында ~50 °С температурада 516 сағат бойы сәулелендірілді. Үлгілер 100 мкм алюминий фольгасына оралды, ол тритий коллекторы ретінде қызмет етті. Сәулеленуден кейін фольгалар ерітіліп, тритий мөлшері сұйық сцинтилляциялық спектрометрия әдісімен анықталды.

Есептік тритий шығымы 1017–1018 атомдар шамасында болып, тәжірибелік нәтижелермен жақсы сәйкес келді. Табиғи құрамды Li_2TiO_3 үшін есеппен 3,35·1017 атом болжанды, ал өлшенгені $(2,0\pm0,4)\cdot10^{17}$ болды; ал ^{6}Li-мен байытылған Li_2TiO_3 үшін тиісінше $2,14\cdot10^{18}$ және $(1,1\pm0,2)\cdot10^{18}$ атом алынды. Тәжірибелік мәндердің төмендеуі тритийдің фольгада толық ұсталып қалмауымен және үлгілеудегі қарапайымдандырулармен, атап айтқанда түйіршіктердің сәулелену кезіндегі өздігінен көлеңкеленуін есепке алмаумен түсіндіріледі.

Алынған нәтижелер литийқұрамды керамикалардан тритийдің белсендірусіз бөлінуінің бар екенін растайды және ұсынылған үлгінің бериллий компоненттері бар реакторлардың радиациялық қауіпсіздігін бағалауға қолдануға болатын сенімді бағалар беретінін көрсетеді.

Түйін сөздер: литий керамикасы, нейтронды сәулелену, тритий, гелий, MCNP6.

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Расчетно-экспериментальные исследования процессов без активационного выделения трития из бериллийсодержащих материалов исследовательских ядерных реакторов

Аннотация. Бериллиевые отражатели в исследовательских реакторах являются известными источниками трития, что вызывает опасения в связи с его возможным высвобождением. Хотя значительное газовыделение наблюдается при повышенных температурах (>700 °С) за счёт диффузии, измеримые количества трития фиксировались и при ~50 °С, что не может быть объяснено данным механизмом. В связи с этим рассматривается процесс безактивационного выделения, при котором ядра трития, образующиеся в приповерхностном слое бериллия, напрямую покидают материал в виде высокоэнергетических ионов.

Для количественной оценки этого процесса была разработана вычислительная модель. Длина пробега ионов трития с энергиями порядка МэВ в твёрдых телах ограничена несколькими

десятками микрометров, что определяет эффективную область приповерхностного выделения. Геометрический анализ позволил вывести выражение для числа ионов трития, покидающих поверхность в условиях облучения. Для проверки модели использовались образцы метатитаната лития (Li_2TiO_3) с естественным и 96% обогащением по ^6Li, облучённые в реакторе ВВР-К при температуре ~50 °C в течение 516 часов. Образцы были обёрнуты алюминиевой фольгой толщиной 100 мкм, служившей коллектором трития, который затем определялся методом жидкостной сцинтилляционной спектрометрии после растворения фольг.

Расчётные выходы трития составили порядок 10^{17}-10^{18} атомов и хорошо согласовались с экспериментальными данными. Для Li2TiO3 с природным изотопным составом было предсказано $3{,}35 \cdot 10^{17}$ атомов против (2,0±0,4)·1017 измеренных, а для обогащённого по ^6Li Li_2TiO_3 – $2{,}14 \cdot 10^{18}$ против (1,1±0,2)·1018. Заниженные экспериментальные значения объясняются неполным удержанием трития в фольге и упрощениями модели, в частности, отсутствием учёта самозащиты гранул при облучении.

Полученные результаты подтверждают существование безактивационного выделения трития из литийсодержащих керамик и демонстрируют, что предложенная модель обеспечивает достоверные оценки, применимые для анализа радиационной безопасности реакторов с бериллиевыми компонентами.

Ключевые слова: литиевая керамика, нейтронное облучение, тритий, гелий, MCNP6.

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