

Simulation of tritium release processes from lithium ceramics under reactor irradiation conditions with varying reactor power

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ARTICLE INFO

Keywords:

Tritium
Lithium ceramics
Blanket
WWR-K reactor
Reactor irradiation
Modelling

ABSTRACT

It is known that one of the most important parameters for lithium ceramics, promising for use in solid-state fusion blankets, includes their properties regarding tritium. Tritium generated in lithium ceramics (as a result of reaction of ${}^6\text{Li}$ atom with neutron) is released from the surface after passing through a chain of processes:

1. it is initially thermalized in the ceramic volume (it should be noted that at this stage, if the reaction of its generation has occurred in the near-surface region, it can be released from the sample in an inactivation-free manner),
2. after that it diffuses to the open surface, interacting with traps in the ceramic volume (here both reversible and non-reversible capture of tritium by traps, which are usually defects in the structure, is possible);
3. tritium atoms associate with each other or with hydrogen impurities on the surface, after which the resulting molecules are desorbed.

To determine the parameters of gas release from ceramics generated by neutron irradiation of tritium, reactor studies are usually carried out. Analysis of the results of such reactor experiments is usually complicated by the need to consider various factors and requires careful consideration of these factors while creating and selecting model parameters.

The present work presents data on modeling the results of a previously conducted reactor experiment. Modeling was carried out by the finite element method (FEM) based on a complex model that considers temperature gradients across the ceramics during the reactor experiment, as well as the processes of tritium diffusion in the volume and desorption of tritium molecules from the ceramic surface. We investigated the sections of the reactor experiment where the reactor power was varied, leading to changes in the temperature fields in the ceramic samples and the released flux of tritium molecules.

1. Introduction

In order to predict the behavior of fusion blanket materials and justify their use in a fusion reactor, it is important to know the structural, thermal, and dynamic properties of the breeder material and their changes occurring during irradiation and formation of structural defects.

For this purpose, usually reactor experiments on irradiation of the

materials under study are conducted, which are aimed at determining the parameters of the processes of tritium generation and release. In order to determine the parameters of such physical processes during irradiation, it is necessary to solve complex problems of studying the relevant mechanisms and developing models that consider the whole complex of influencing factors.

Nowadays, a set of irradiation experiments with the fusion blanket materials [1–11], specifically, lithium ceramics of various compositions,

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<https://doi.org/10.1016/j.jnucmat.2025.155979>

Received 29 January 2025; Received in revised form 30 May 2025; Accepted 10 June 2025

Available online 16 June 2025

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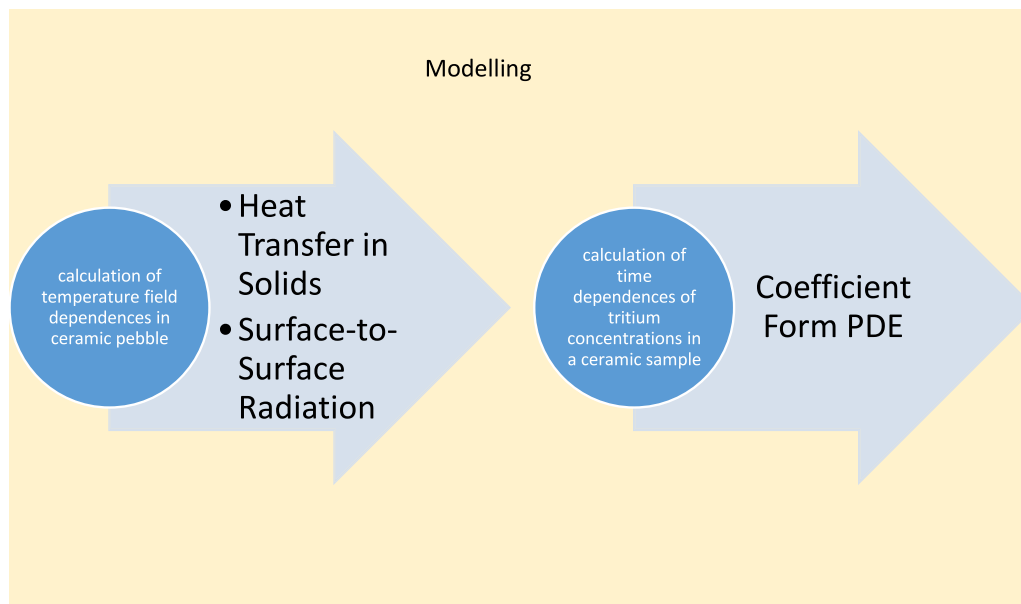


Fig. 1. Modeling module blocks.

titanium beryllides, [12–22] etc., have been performed as part of the Republic of Kazakhstan scientific programs covering a wide range of tasks devoted to materials science and radioecological issues of nuclear and fusion power engineering.

In reactor experiments with lithium ceramics based on lithium metatitanate with 95 % enrichment by ^6Li , in order to correctly analyze the results, there was a need to develop a model for estimation of tritium generation and release, which would consider changes in the temperature field in the samples investigated.

It should be noted that there are a large number of approaches to analyzing the results of reactor experiments to study the generation and release of tritium from lithium-containing fusion materials using computer modeling methods, which describe the processes of tritium transfer based on the combination of diffusion mechanisms and surface processes [23–29]. Almost all models describe reactor studies conducted under conditions of gas purging of the samples studied, and no model considers the existing temperature gradient within a single sample (pebble).

This paper presents the results of the development of a complex model of tritium generation and release from lithium ceramics Li_2TiO_3 enriched up to 96 % by ^6Li isotope, which would allow describing the results of reactor experiments under conditions of tritium generation and release in vacuum, considering the non-uniformity of the temperature field on the sample. The results of modeling of experimental dependences of tritium release from lithium metatitanate under irradiation conditions and values of certain model parameters are also given.

2. Experimental part

2.1. Description of the modeling experiment

The results of the experiment on reactor irradiation of Li_2TiO_3 lithium ceramics with 95 % enrichment by ^6Li are presented in detail in [22]. Let us briefly summarize the main points of the experiment: lithium ceramics samples having a spherical shape (sample radius 1 mm) were placed at the bottom of the irradiated capsule in a vacuum medium (residual gas pressure above the sample is $<1.3 \cdot 10^{-4}$ Pa); the samples were not in contact with each other; during irradiation, tritium was generated in the samples (as a result of interaction of neutrons with ^6Li atom), which diffused on the surface of the samples and then was released from the sample surface as a result of associative desorption

processes; tritium release and the temperature of the bottom of the capsule with samples were recorded in the experiment; some irradiation conditions were varied at different stages of the experiment: temperature of the capsule (and samples, respectively), irradiation power (which affected the parameters of energy release in the samples and the rate of tritium generation).

Based on the features of the experiment, let us distinguish the main processes that will be used as the model's basis.

Initially, the model will describe the processes in a single sample of lithium ceramics of spherical shape located in a vacuum and having physical contact with the bottom of the capsule. The area of contact between the pebble and the bottom of the capsule is determined both experimentally and as a result of calculation (by deviation from the spherical shape of the samples studied).

Next, we will describe the processes associated with the temperature change of the samples: some heat is released in the samples during irradiation due to nuclear reactions of neutrons with ceramic atoms and due to radiation heating by the gamma rays; heat transfer between the samples and the medium occurs as a result of contact between the sample and the bottom of the capsule and due to thermal exchange between the radiating surfaces of the sample and the capsule.

As for the processes of tritium generation and transfer, the main processes will be: generation of tritium as a result of nuclear reaction of neutrons with ^6Li atom (the generation rate normalized to reactor power was determined on the basis of neutron-physical calculations, and estimated from experiments analyzing the quasi-equilibrium release level of tritium molecules); diffusion of tritium in the sample; associative desorption of tritium from the sample surface into a vacuum.

It should be noted that the parameters of heat transfer, diffusion, desorption, etc., have temperature dependence, which should be considered in modeling. A closely related modeling study is given in [30]. That work focused primarily on heat transfer processes and the influence of geometry and inter-pebble spacing on temperature distribution. In contrast, the present study integrates both the thermal and tritium mass transfer domains in a single modeling framework. While both studies emphasize the need to account for non-uniform heating under vacuum, the current study uniquely addresses the kinetics of tritium generation, diffusion, and desorption processes in a self-consistent manner.

Thus, to model the change of tritium concentration in the sample during its generation/release, it is necessary to solve the coordinated

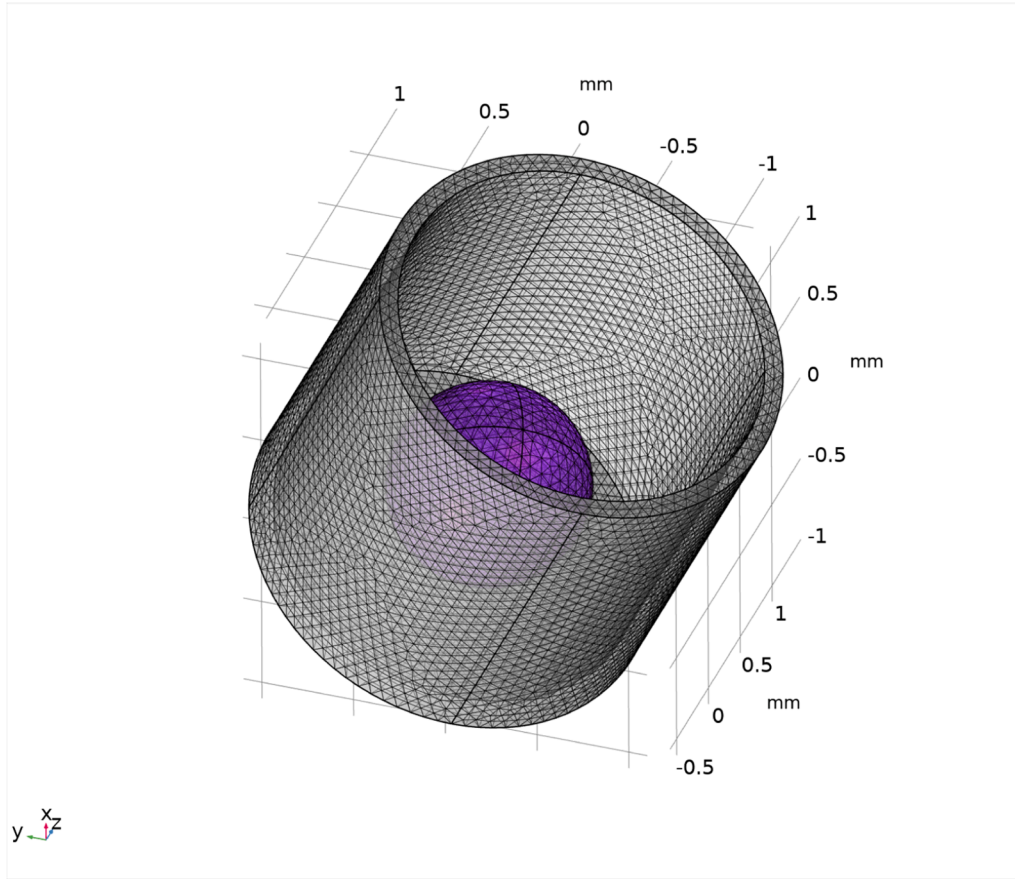


Fig. 2. Geometry of the model.

problems of transport, heat, and mass transfer (including heat transfer between radiating surfaces).

3. Results

3.1. Modelling

Modeling of the kinetics of tritium release from ceramics under reactor irradiation conditions was performed by the finite element method using the COMSOL Multiphysics software. Modeling was divided into 2 blocks (see Fig. 1): in the first block the calculation of temperature field dependences in ceramic pebble was performed, where Heat Transfer in Solids (ht) and Surface-to-Surface Radiation (rad) modules were used; in the second block, based on the obtained temperature fields in the ceramic pebble, the time dependences of tritium concentrations in the ceramic sample were calculated using the Coefficient Form PDE module.

Using the Heat Transfer in Solids module, the system of heat transfer equations (1) was solved, which allows considering the heating of the sample as a result of the interaction of radiation with the lithium ceramic material (internal heat source Q), as well as heat transfer in the pebble volume and at the boundary in the area of contact between the pebble and the bottom of the ampoule.

$$\rho C_p \frac{\partial T}{\partial t} + \rho C_p \mathbf{u} \cdot \nabla T + \nabla \cdot \mathbf{q} = Q \quad (1)$$

$$\mathbf{q} = -k \nabla T$$

T – temperature; k – thermal conductivity;

ρ – density;

C_p – heat capacity at constant pressure.

Using the Surface-to-Surface Radiation module, the system of equations (2) was solved to account for the exchange of heat fluxes between radiating surfaces based on the Stefan-Boltzmann law:

$$J = \varepsilon e_b(T) + \rho_d G \quad (2)$$

$$\varepsilon + \rho_d = 1$$

$$e_b(T) = n^2 \sigma T^4$$

$$-n \cdot \mathbf{q} = \varepsilon (G - e_b(T))$$

j – total outgoing radiation flux;

G – total incoming radiation flux;

ρ_d – reflectance;

T – temperature;

ε – radiation coefficient of the surface.

Using the Coefficient Form PDE module, we solved the system of transport equations (written in simplified form (3)), which allows considering the accumulation of diffusant in the sample volume, its diffusion in the volume, and release from the sample surface into vacuum (when boundary conditions (4) are given):

$$\frac{\partial C}{\partial t} + \nabla \cdot (-D \nabla C) = f \quad (3)$$

$$\nabla = \left[\frac{\partial}{\partial x}, \frac{\partial}{\partial y}, \frac{\partial}{\partial z} \right] \quad (4)$$

C – tritium concentration; f – tritium generation rate;

D – diffusion coefficient having Arrhenius temperature dependence:

$$D(T) = D_0 \cdot \exp\left(-\frac{E_d}{RT}\right).$$

To perform computational modeling, we build a 3D model geometry,

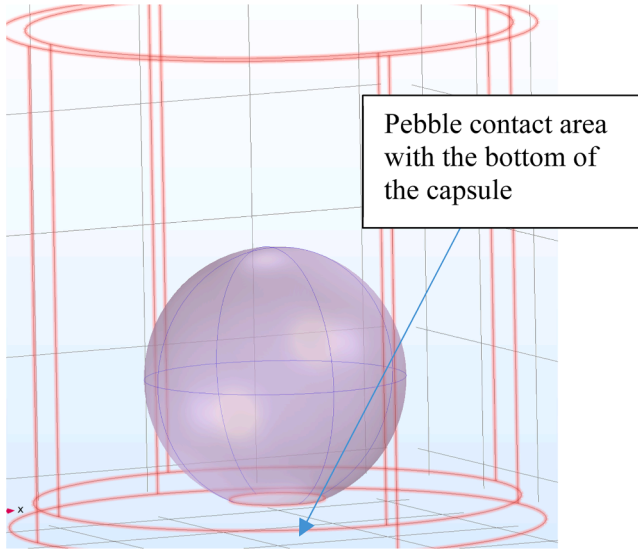


Fig. 3. Schematic of the geometric model with the area of contact between the pebble and the bottom of the capsule.

shown in Fig. 2, which consists of two domains - an external metal capsule and a ceramic pebble. Also for the calculations we assume the scheme of the geometry model with the area of contact of the pebble with the bottom of the capsule (Fig. 3).

The initial room temperature condition (293.15 K) was chosen as initial conditions for all domains of the model (Fig. 4).

The condition of equality of the capsule domain wall temperature to the measured capsule bottom temperature in the experiment ($T_{in}(t)$) was chosen as time-varying boundary conditions.

Also, the volumetric heat source (ceramic pebble) was selected and specified here, where the released heat was determined based on the time dependence of the change in reactor power $W(t)$ (from the experiment data) and the energy release in the ceramic at 6 MW (Figs. 5 and 6

and Eq. (5)).

$$Q = w_{cer} * W(t)/6 \quad (5)$$

The radiating surfaces between which the heat transfer occurs in the model were chosen as shown in Fig. 6.

Since the mass transfer of tritium was determined only in the pebble, all further calculations are given for the second domain: the ceramic pebble.

The initial concentration of tritium was assumed to be zero in the entire volume of the pebble. The conditions at its boundary were determined on the basis of the desorption equality, which relates the concentrations of desorbing particles on the surface and the concentrations of two-atomic gases released from the surface. Tritium leaves the ceramic surface and associates into T_2 and HT molecules. The HT molecule is formed by the merging of a hydrogen atom and a tritium atom. It should be noted here that the presence of hydrogen in the irradiation chamber at a pressure level of about $1.3 \cdot 10^{-4}$ Pa leads to the fact that it is always present in the form of atoms on the ceramic surface. The tritium flux from the ceramic surface is determined by the following expression:

$$\frac{dCT_R}{dt} = KT_2(T)2CS(R)_T^2 + KHT(T)CS(R)_TCS(R)_H \quad (6)$$

where $CS(R)_TCS(R)_H$ - are the concentrations of tritium and hydrogen on the pebble surface,

$KT_2(T)$, $KHT(T)$ - desorption constants of molecules having Arrhenius temperature dependence: $(K_{des}(T) = K_{0des} \cdot \exp(-\frac{E_{des}}{RT}))$.

It should be noted that the desorption constants are often assumed to be equal for isotopes of the same element: $(K_{des}(T) = KT_2(T) = KHT(T))$.

The hydrogen concentration on the pebble surface was estimated from experimental data by the ratio of HT and T_2 fluxes released: $\left(\delta(t) = \frac{J_{HT}(t)}{J_{T_2}(t)}\right)$

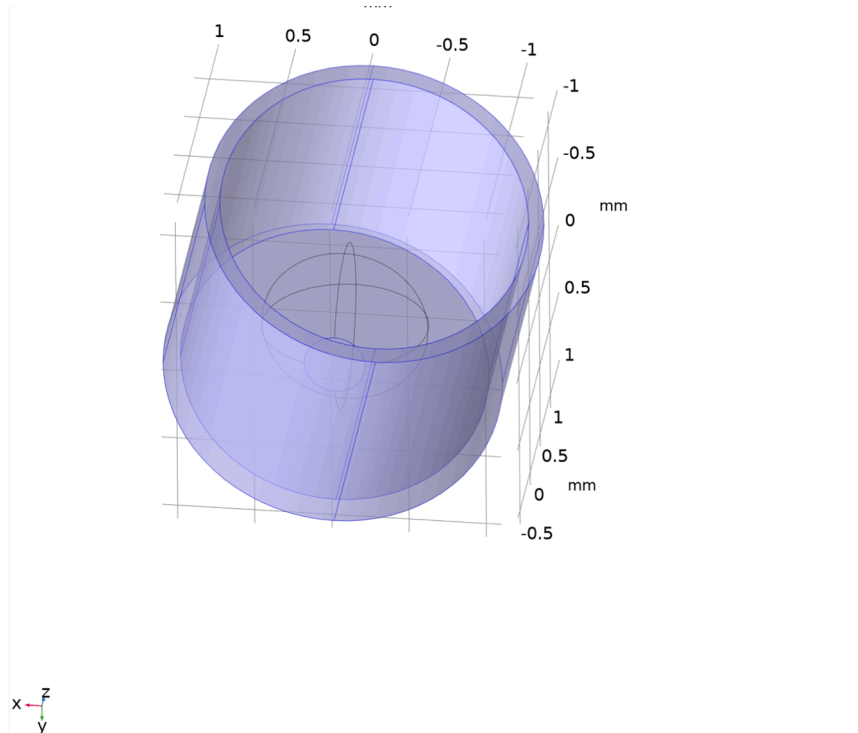


Fig. 4. Thermal model boundary conditions.

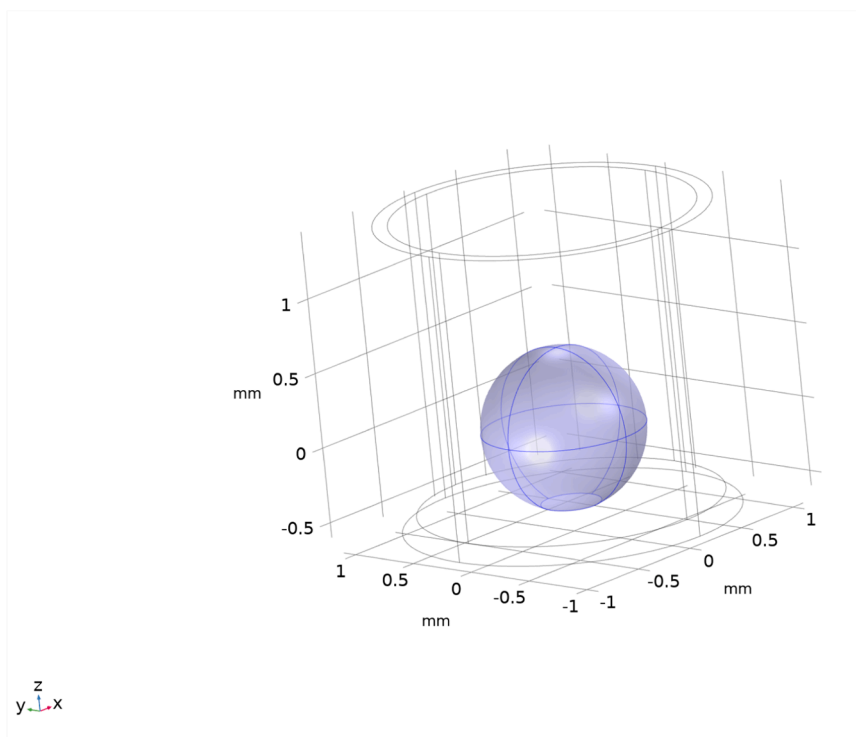


Fig. 5. Heat source in the model - ceramic pebble.

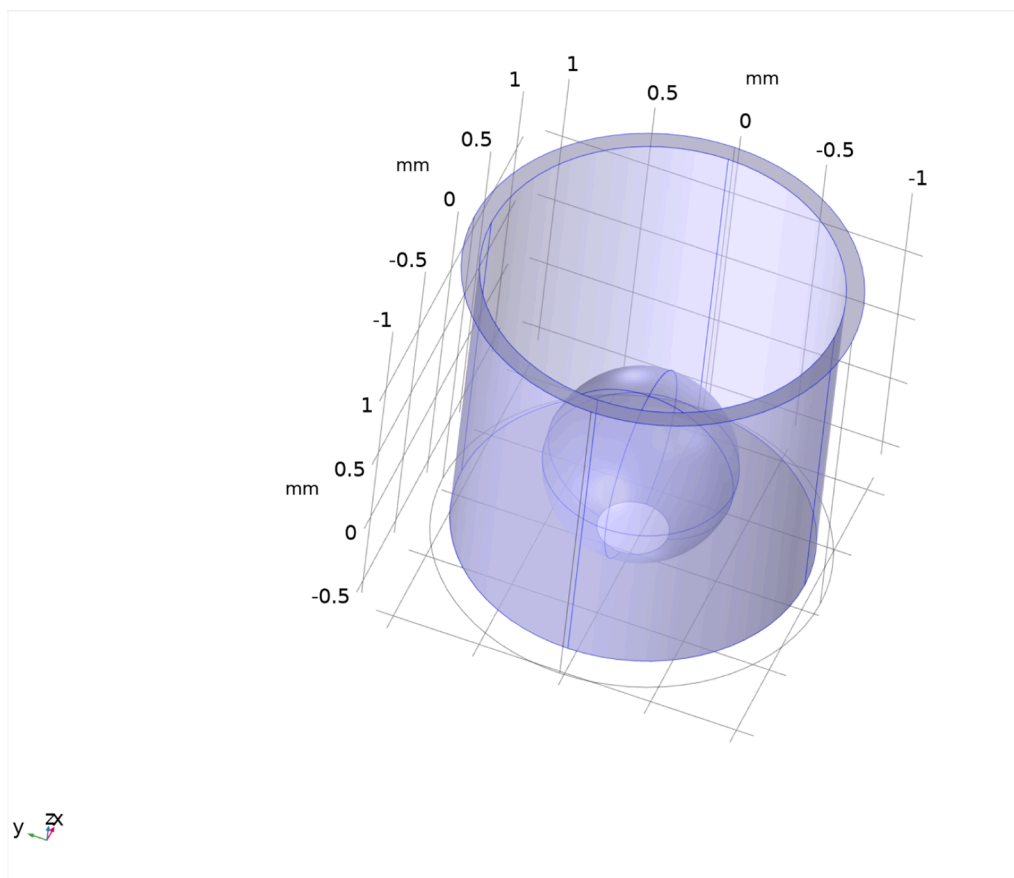


Fig. 6. Radiating surfaces between which radiation heat exchange takes place.

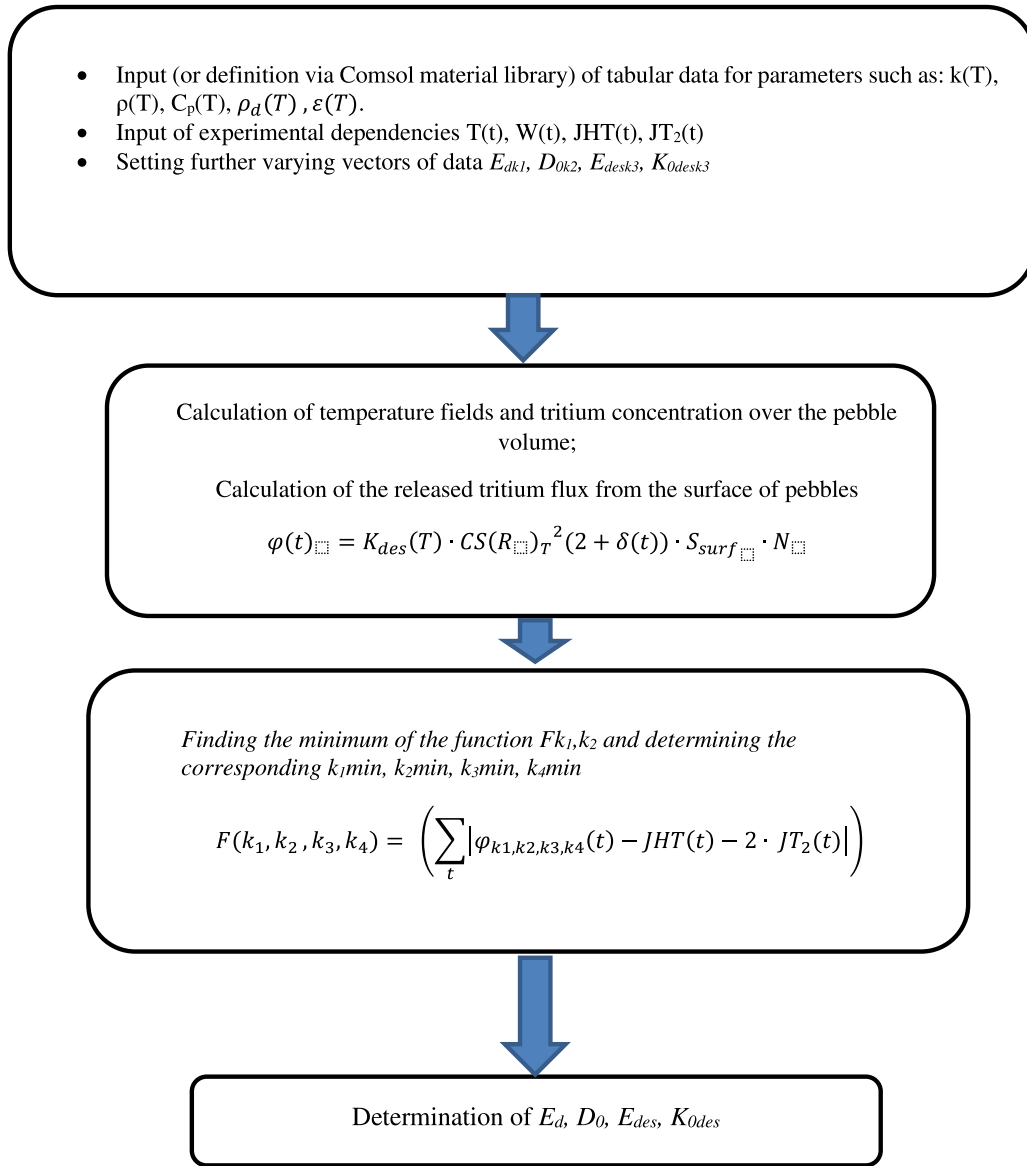


Fig. 7. Algorithm of calculations according to the developed model.

Considering the above, expression (6) took the form:

$$\frac{dCT_R}{dt} = K_{des}(T) CS(R)_T^2 (2 + \delta(t)) \quad (7)$$

Regarding the tritium concentration at the surface, we assume that it is related to the bulk tritium concentration, which is calculated within the model as follows:

$$CS(R)_T = 3\sqrt{C(R)_T^2} \cdot 1 \left(mol^{3/2} \right) \quad (8)$$

Meanwhile, the boundary condition on the pebble surface for the tritium desorption process was given in the following:

$$\frac{dCT_R}{dt} = K_{des}(T) 3\sqrt{C(R)_T^4} (2 + \delta(t)) \quad (9)$$

Thus, for the selected model, the geometry of two domains has been set: an external metallic capsule and a ceramic pebble, and for the calculations, we assume the scheme of the geometrical model with the area of contact between the pebble and the bottom of the capsule. We set initial and boundary conditions both for the thermal task and for the task of tritium mass transfer in the pebble, assuming the mechanisms of heat

exchange in the domains and tritium mass transfer in the ceramic pebble. On the basis of the developed model, we perform computational modeling by varying the parameters describing these processes.

3.2. Model algorithm

The algorithm of calculations on the developed model is presented in Fig. 7. As can be seen from the algorithm, modeling involves varying four parameters that determine the processes of desorption and diffusion, which in general is a complex task, in addition, there is uncertainty associated with the fact that it is possible to obtain a whole set of parameters that describe the experimental curve adequately. Consequently, it is necessary to set the intervals of variation of these parameters within the framework of computational criteria that have a physical justification. Therefore, one of the important stages of modeling was the estimation of some parameters by analytical method from the experimental curve of tritium release in the selected areas of the experiment, where the conditions of the irradiation process changed. We considered the areas where the temperature of the samples was changed at constant reactor power and the areas where a short-term decrease in reactor power was carried out. The problem of estimating the

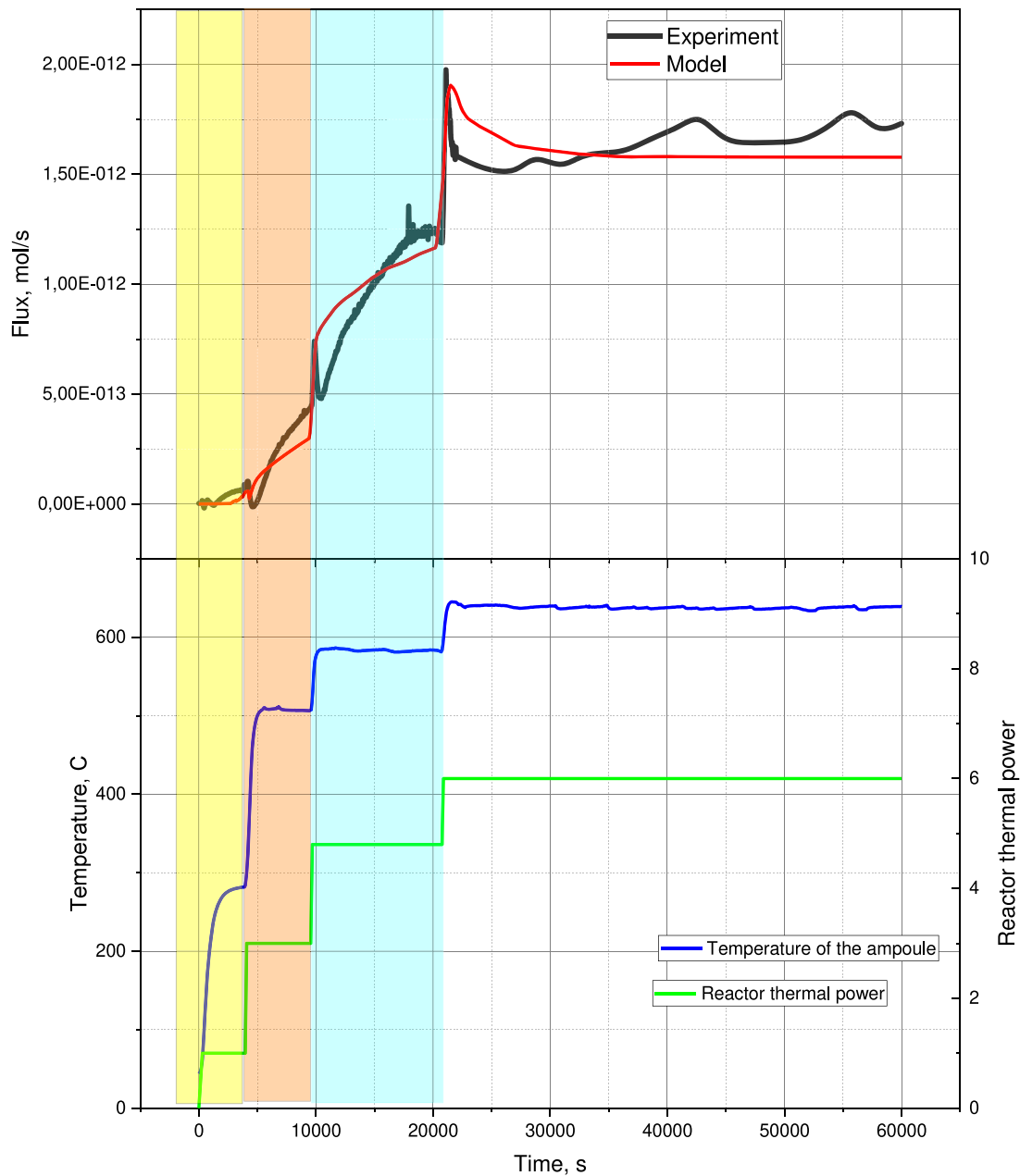


Fig. 8. Results of modeling of tritium release in comparison with the experimental curve.

parameters of the tritium diffusion coefficient in ceramics E_d , D_0 was solved for these areas within the diffusion approximation only.

4. Discussion

The obtained simulation results are shown below in Fig. 8. The best approximation was obtained for the following desorption parameters and diffusion coefficients:

$$K_{des} = 1.2 \cdot 10^{-6} (\text{m}^2 / \text{mol} \cdot \text{s}) \cdot \exp - (52 (\text{kJ} / \text{mol}) / (RT)) \quad (10)$$

$$D = 3 \cdot 10^{-8} (\text{m}^2 / \text{s}) \cdot \exp - (72 (\text{kJ} / \text{mol}) / (RT)) \quad (11)$$

As can be seen from Fig. 8, it was possible to fit the quasi-equilibrium levels of tritium release quite well, for different parts of the experiment, which varied quite significantly in irradiation conditions.

As for the thermal task, the calculations showed that there is a significant temperature gradient in the pebble (Figs. 9 and 10) and,

therefore, estimates of the real pebble temperature in the reactor experiments are necessary for a more accurate description of the experiment parameters.

A comparison of the ampoule temperature and the average pebble temperature is shown in Fig. 11. It should be noted that a possible noticeable difference between the estimated parameters of diffusion coefficients according to analytical expressions and those obtained in the course of modeling is caused, in particular, by the fact that in analytical estimates the temperature of pebbles was taken equal to the temperature of the bottom of the ampoule, where the samples were located. In the present work, the calculated temperature gradients between the average pebble temperature and the ampoule bottom temperature, performed for a reactor power of 6 MW (see Fig. 12), are presented on the basis of a complex model.

As can be seen from the calculations, the difference between the average pebble temperature and the ampoule bottom temperature is greater than 100 °C and decreases as the temperature rises. Such a

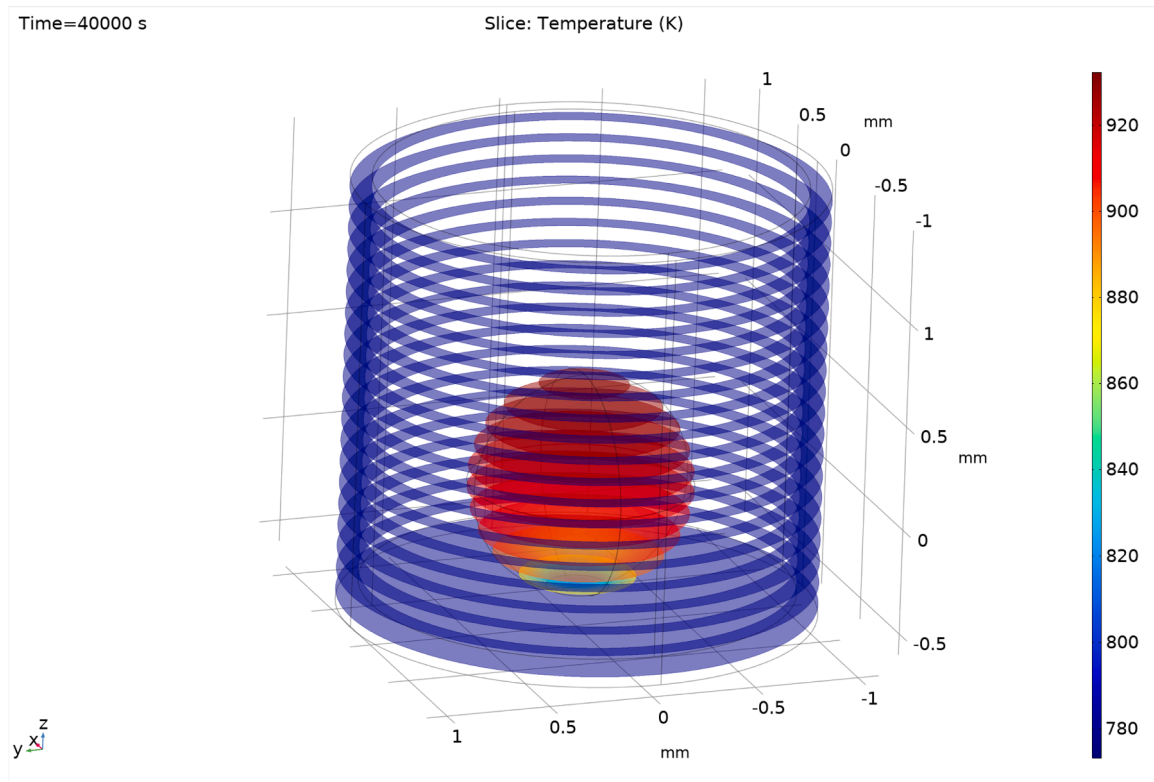


Fig. 9. Temperature field in the modeled system under quasi-equilibrium condition for a reactor power of 6 MW (40,000 s from the start of the experiment).

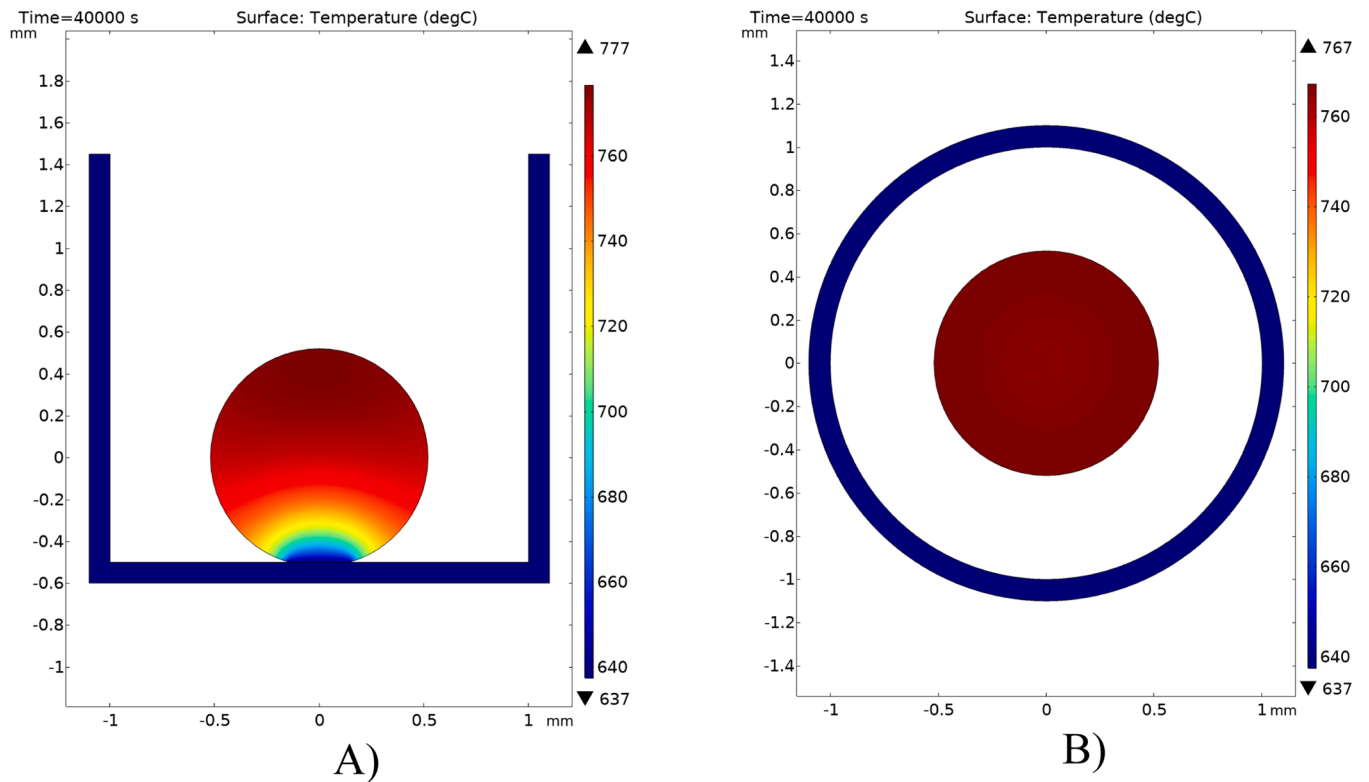


Fig. 10. Temperature distribution along the pebble cross-section (A- vertical section at the center of the pebble; B- horizontal section at the center of the pebble) for a reactor power of 6 MW (40,000 s from the start of the experiment).

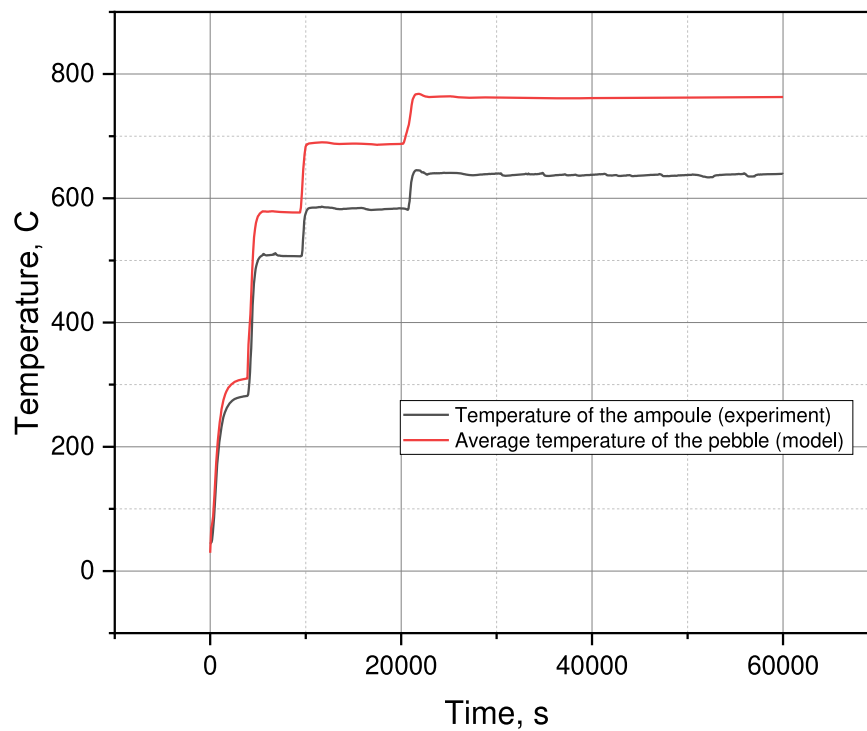


Fig. 11. Temperature of the ampoule and calculated average temperature of pebbles during the experiment.

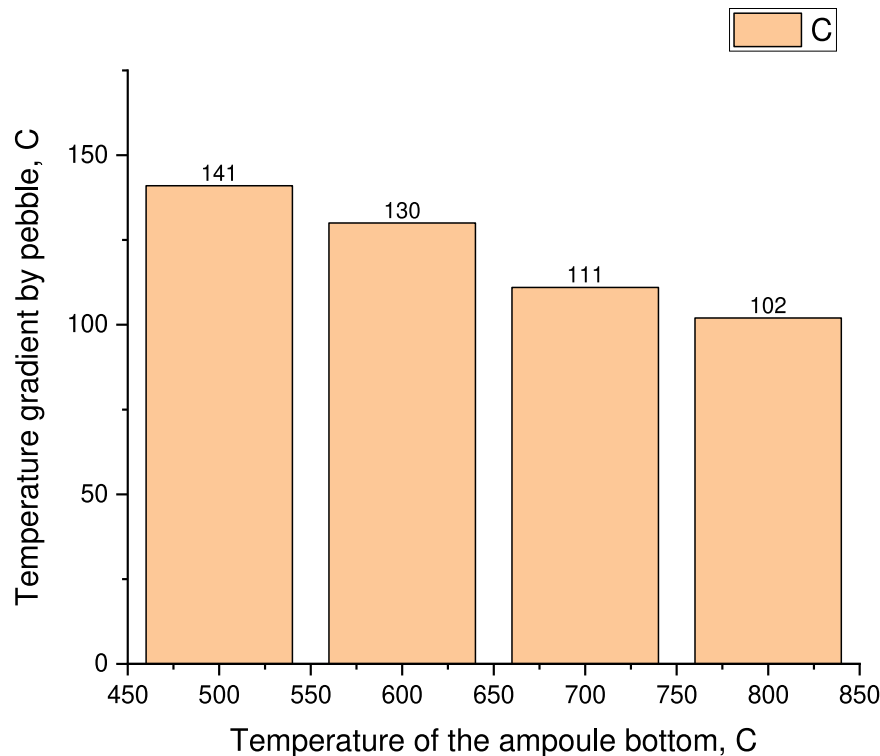


Fig. 12. Dependence of the difference between the average pebble temperature and the ampoule bottom temperature (for 6 MW reactor power).

noticeable difference between the pebble temperature and the ampoule bottom temperature is largely determined by the high values of energy release in the ceramic pebble.

Also, one of the main results of modeling was the data on the distribution of tritium concentration (mol/m^3) in the ceramic pebbles (in particular, due to the gradient in the thermal state of the pebble, tritium

concentration gradients are observed in Figs. 13 and 14). Modeling calculations have shown that there is some non-uniformity in tritium concentration along the vertical section of the pebble, specifically, an increase in tritium concentration towards the region of contact between the pebble and the bottom of the ampoule. Considering the established effects in the distribution of tritium concentration, the dependence of

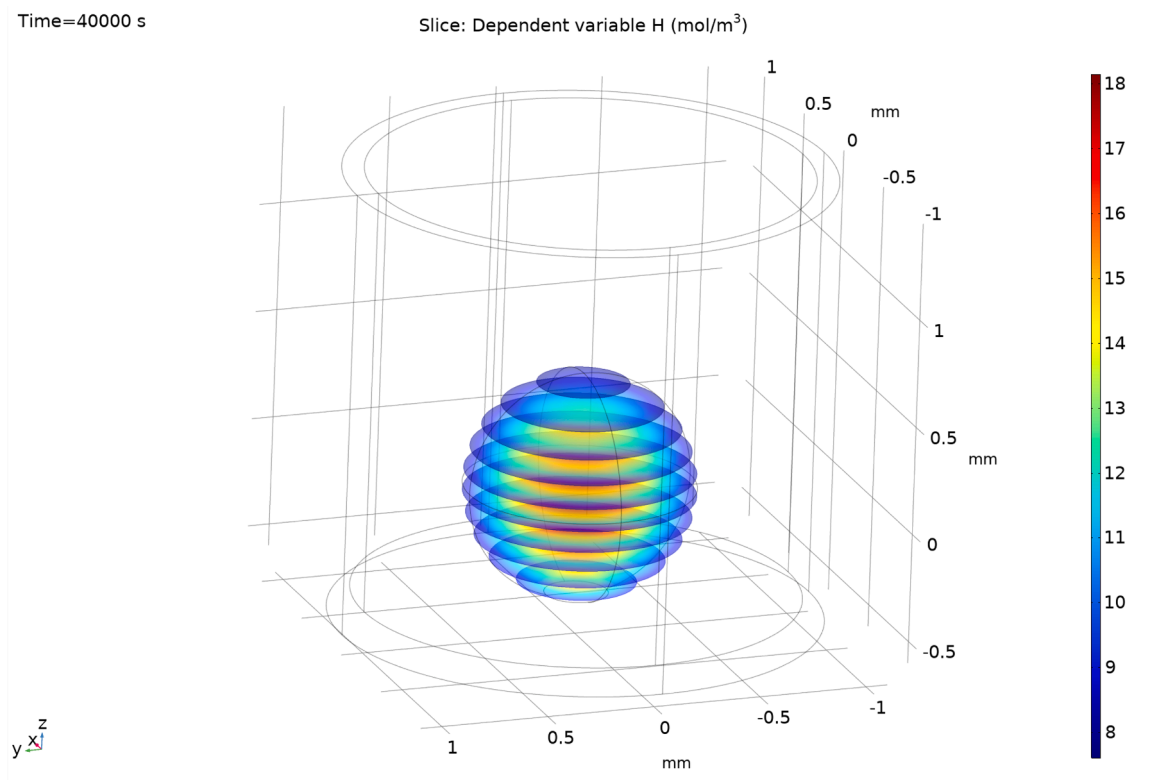


Fig. 13. Distribution of tritium concentration (mol/m^3) in pebble at quasi-equilibrium state, for a reactor power of 6 MW (40,000 s from the start of the experiment).

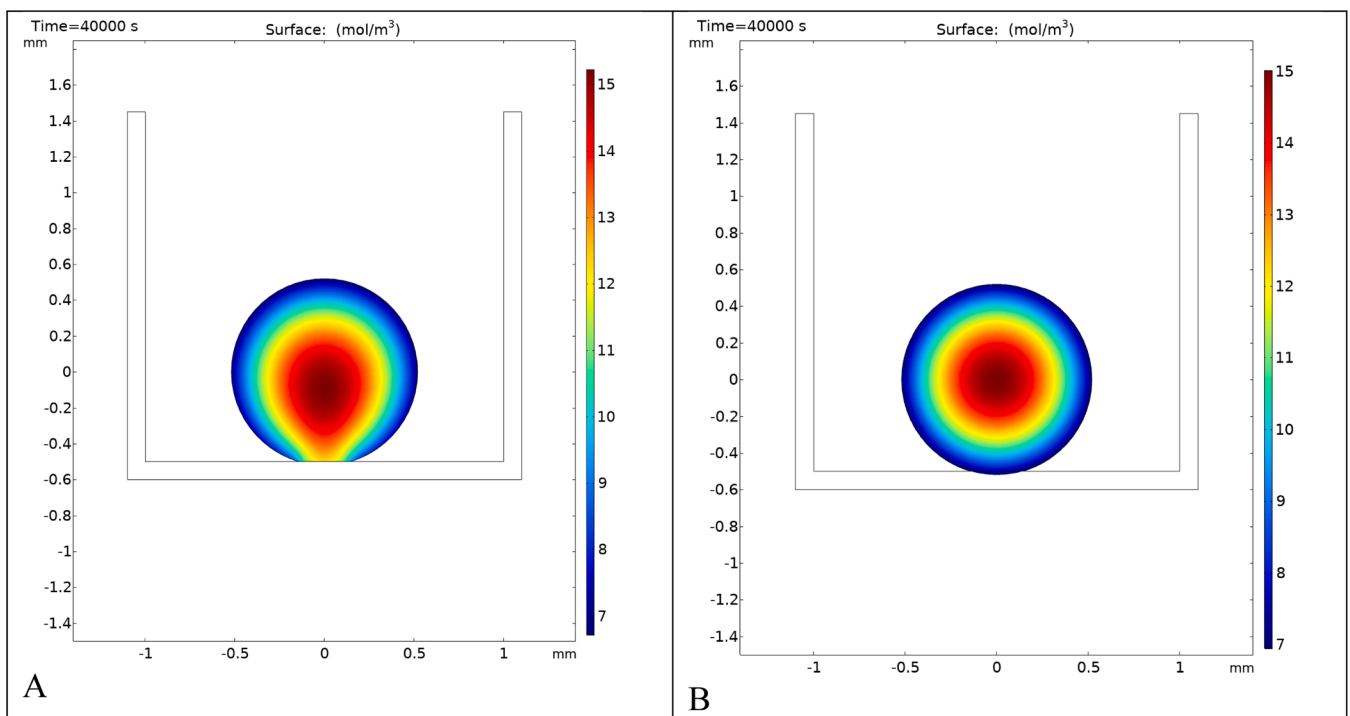


Fig. 14. Distribution of tritium concentration (mol/m^3) over the pebble cross-section (A- vertical section at the center of the pebble; B- horizontal section at the center of the pebble) for a reactor power of 6 MW (40,000 s from the start of the experiment).

the change in the amount of tritium in the ceramic pebble during the time of the experiment was plotted using the obtained data. The summarized results of modeling the kinetics of tritium release in reactor experiments considering the established dependence of the tritium

concentration in the ceramic sample on the thermal state of the sample are shown in Fig. 15.

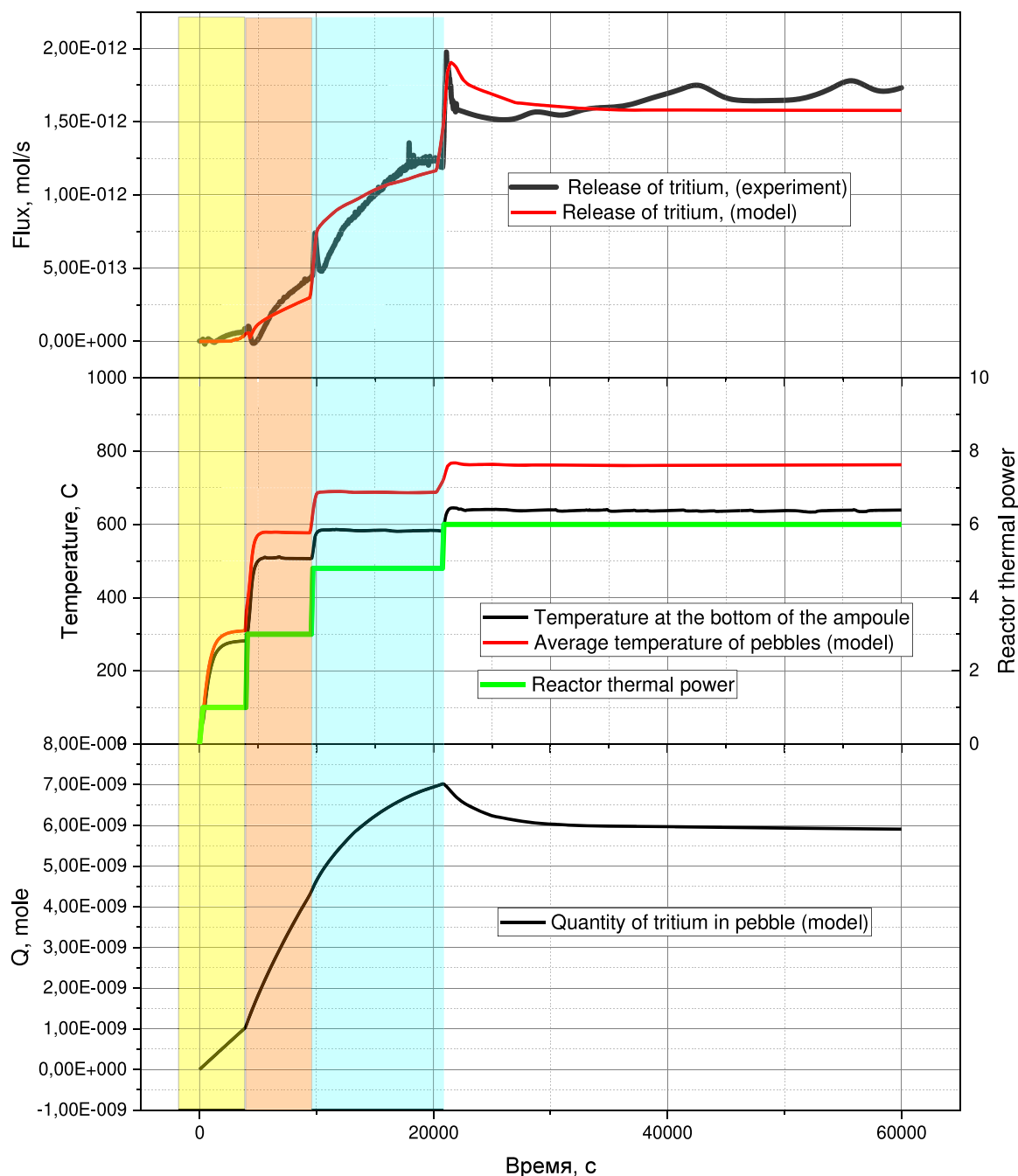


Fig. 15. Summarized results of modeling of tritium release from ceramic pebble during the reactor irradiation process.

5. Conclusion

As a result of the research, a comprehensive model describing the processes of temperature field changes and tritium generation/release in a single ceramic pebble was developed, focused on processing and analyzing the results of reactor experiments at the WWR-K research reactor. This model made it possible to describe the experimental data more accurately and to determine the key parameters of tritium mass transfer processes in Li_2TiO_3 lithium ceramics under neutron irradiation conditions, such as diffusion coefficients and tritium desorption constants.

$$K_{des} = 1.2 \cdot 10^{-6} (\text{m}^2 / \text{mol} \cdot \text{s}) \cdot \exp - (52 (\text{kJ} / \text{mol}) / (RT))$$

$$D = 3 \cdot 10^{-8} (\text{m}^2 / \text{s}) \cdot \exp - (72 (\text{kJ} / \text{mol}) / (RT))$$

It was shown that there is a noticeable difference in the temperature of the pebble and the bottom of the ampoule, which is largely determined by the high values of energy release in the ceramic pebble.

Modeling showed that there is some non-uniformity in the tritium concentration along the vertical section of the pebble, specifically, an increase in the tritium concentration toward the area of contact between the pebble and the bottom of the ampoule.

CRedit authorship contribution statement

Timur Kulsartov: Methodology, Formal analysis, Conceptualization. **Inesh Kenzhina:** Writing – review & editing, Project administration, Funding acquisition. **Yergazy Kenzhin:** Validation, Formal analysis. **Yevgeniy Chikhray:** Validation, Software. **Asset Shaimerdenov:** Resources, Investigation. **Zhanna Zaurbekova:** Writing –

original draft, Data curation. **Alexander Yelishenkov:** Visualization, Investigation. **Kuanysb Samarkhanov:** Writing – review & editing. **Meiram Begentayev:** Formal analysis. **Saulet Askerbekov:** Visualization, Investigation. **Aktolkyn Tolenova:** Investigation, Data curation.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgment

The research was funded by the Science Committee of the Ministry of Science and Higher Education of the Republic of Kazakhstan with Program number BR21882237.

Data availability

Data will be made available on request.

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