ABSTRACT

of the dissertation work of Tolenova Aktolkyn Userbekkyzy on the topic "Development of a model for the generation and release of tritium from lithium metatitanate under neutron irradiation", submitted for the degree of Doctor of Philosophy (PhD) in the educational program "8D05308 – Nuclear Physics"

General description of work the dissertation is devoted to the development and verification of a comprehensive mathematical model describing the processes of tritium generation, transport, and release from spherical lithium metatitanate (Li_2TiO_3) samples (pebbles) enriched in the ^6Li isotope up to 96% under neutron irradiation and deep vacuum conditions. The model is based on the results of original experimental investigations performed using the in-situ vacuum extraction method at the WWR-K research reactor of the Institute of Nuclear Physics (Almaty).

A key feature of the developed model is the first-time implementation of spatially non-uniform temperature field accounting inside an individual spherical sample. The temperature distribution was calculated using the finite element method in the COMSOL Multiphysics software environment. The physical basis of the model consists of deterministic volumetric sources of heat generation and tritium production determined through neutron-physical analysis of the reactor experiment conditions. This approach made it possible to reliably determine the kinetic parameters of tritium diffusion and desorption, estimate the tritium residence time in the material, and demonstrate the applicability of the model for predicting tritium behavior under conditions close to the operational regimes of breeder zones in fusion reactors.

The relevance of the dissertation topic. The steadily increasing global energy demand and severe environmental challenges caused by large-scale consumption of fossil fuels make the search for new, sustainable, and safe energy sources extremely relevant. One of the most promising directions is controlled thermonuclear fusion based on the deuterium-tritium (D-T) fusion reaction.

A key technological challenge hindering the implementation of fusion energy is achieving tritium fuel self-sufficiency. Since natural tritium resources are negligible, tritium must be produced directly inside the fusion reactor as a result of nuclear reactions occurring under neutron irradiation of lithium-containing materials.

In fusion reactor designs, the blanket module surrounding the plasma chamber plays a critical role. It provides neutron energy absorption, tritium generation, and tritium extraction for recirculation into the fuel cycle. The selection of an optimal breeder material combining radiation resistance, thermal stability, and favorable tritium diffusion characteristics is therefore a primary scientific and engineering task. Among candidate materials, lithium metatitanate (Li_2TiO_3) occupies a special position due to its high chemical stability and its ability to efficiently release tritium at moderate temperatures ($200 - 400\ °C$).

Despite extensive experimental research conducted within international programs such as EXOTIC, HICU, and TEQUILA, a number of fundamental issues remain unresolved. In particular, most existing computational models (DISPL1, MISTRAL, TIARA) do not account for temperature field non-uniformity inside individual granules, which significantly reduces prediction accuracy under realistic reactor conditions where local heating is inevitable. In addition, processes occurring under deep vacuum conditions characteristic of several advanced blanket concepts remain insufficiently studied.

Thus, the development of a comprehensive model capable of simultaneously accounting for heat transfer, tritium generation, diffusion, and desorption under in situ irradiation conditions with internal temperature gradients represents a relevant scientific and technological challenge.

The aim of the dissertation research. The aim of the dissertation research is to develop and verify a comprehensive mathematical model of coupled tritium generation, transport, and release processes in a single spherical lithium metatitanate (Li_2TiO_3) sample, taking into account the influence of a spatially non-uniform temperature field under neutron irradiation.

Research Tasks

To achieve the stated aim, the following research tasks were formulated and solved:

1. To analyze modern controlled thermonuclear fusion projects, experimental facilities, and research methods, as well as to generalize the results of reactor experiments on tritium generation and release from lithium ceramics and analyze existing approaches to experimental data processing and interpretation.
2. To perform neutron-physical analysis of the planned experiment conditions at the WWR-K research reactor in order to determine deterministic volumetric sources of heat generation and tritium production in the sample.
3. To develop an analytical model intended for primary processing of experimental data on tritium release kinetics and for estimating effective kinetic parameters of the process.
4. To develop a comprehensive numerical model of coupled heat and mass transfer processes in a single Li_2TiO_3 ceramic sample using the COMSOL Multiphysics software environment based on deterministic volumetric sources of heat and tritium generation.
5. To verify the developed model by comparing numerical simulation results with in situ reactor experiment data, determine tritium transport kinetic parameters (diffusion coefficient and desorption rate constant), and estimate the characteristic tritium residence time in the material.
6. To demonstrate the applicability of the verified model for predictive calculations of tritium behavior under conditions simulating operational regimes of fusion reactor breeder materials.

The objects of the study. The objects of the study are spherical lithium metatitanate Li_2TiO_3 samples (pebbles) with an average diameter of approximately $1 \pm 0.2\ mm$, enriched in the ^6Li isotope up to 96% and irradiated with neutrons under the conditions of the WWR-K research reactor.

The subject of the study. The subject of the study is the mechanisms and kinetic parameters of tritium generation, bulk diffusion, and surface desorption processes in lithium metatitanate Li_2TiO_3 under neutron irradiation, as well as their dependence on the spatially non-uniform temperature field inside a spherical sample.

Research methods

The dissertation employs a комплекс of computational and experimental research methods, including:

- in situ tritium vacuum extraction method implemented directly at the WWR-K research nuclear reactor;
- neutron-physical modeling for determining volumetric heat generation and tritium production rates;
- analytical modeling of tritium release kinetics for primary processing of experimental data;
- numerical finite element modeling of coupled heat and mass transfer problems using the COMSOL Multiphysics software package.

Provisions made for the defense

1. A comprehensive mathematical model has been developed for the processes of tritium generation, transport, and release in a single spherical lithium metatitanate (Li_2TiO_3) sample (pebble) enriched in the ^6Li isotope up to $\sim 96\%$ under vacuum and neutron irradiation conditions. Within the framework of this formulation, the spatially non-uniform temperature field formed inside the sample is taken into account for the first time. The numerical implementation of the model was carried out using the COMSOL Multiphysics software package and is based on deterministic volumetric sources of tritium generation and heat release calculated within the neutron-physical analysis of the experimental conditions at the WWR-Kresearch reactor:

– volumetric heat generation source caused by the exothermic reaction $^6Li(n,\alpha)T$: $q_{\text{gen}}(t) = 187.1 \cdot \frac{W(t)}{W_0}$ W/cm^3;

– volumetric tritium generation source from the same reaction:

$$G_T(t) = 8.64 \times 10^{-10} \cdot \frac{W(t)}{W_0} \text{ mol/(cm}^3 \cdot \text{s)}$$

where $W(t)$ is the current reactor power and $W_0 = 6\,MW$.

2. Based on the results of model verification using in situ reactor experiment data, the tritium diffusion coefficient in lithium metatitanate Li_2TiO_3 was determined and is described by an Arrhenius-type dependence:

$$D = 3 \times 10^{-8}\, exp\left(-\frac{72 \text{ kJ/mol}}{RT}\right) \text{m}^2/\text{s}$$

The obtained value determines the characteristic time scale of bulk tritium transport in the material.

3. The tritium desorption rate constant from the Li_2TiO_3 surface was determined and has the following form:

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

Together with the diffusion coefficient, this parameter allows determining the tritium residence time in the material, which is approximately $\sim 10^4$ s at $600\,°C$ and decreases $\sim 10^3$ s at $900°C$.

Scientific novelty of the work

1. An original comprehensive model of thermal and diffusion processes for a single spherical lithium metatitanate (Li_2TiO_3) sample (pebble) has been developed and implemented. The principal distinguishing feature of the model is the explicit consideration of spatial temperature field non-uniformity inside the sample. The model is based on deterministic data, namely volumetric sources of heat release and tritium generation calculated within the framework of neutron-physical analysis of specific experimental conditions at the WWR-K research reactor.

2. Within the framework of model verification using in situ experimental data, the fundamental kinetic parameters of tritium transport in Li_2TiO_3 were determined for the first time. For the diffusion process, the following parameters were established: a pre-exponential factor $D_0 = (3{,}0 \pm 0{,}15) \times 10^{-8}\ m^2/s$ and an activation energy $\text{E}_d = (72 \pm 1)\ kJ/mol$. For the desorption process, the following parameters were determined: a pre-exponential factor $K_0 = (1{,}20 \pm 0{,}06) \times 10^{-6}\ m^2/(mol \cdot s)$ and an activation energy $\text{E}_{des} = (52 \pm 1)\ kJ/mol$.

3. Based on the obtained kinetic parameters, the first quantitative evaluation of the tritium residence time in the material under conditions simulating reactor irradiation was performed. It was shown that this parameter decreases by approximately one order of magnitude with increasing temperature from $600\,°C\ (\approx 10^4\ s)\ to\ 900\,°C\ (\approx 10^3\ s)$, which is of significant importance for predicting the operation of breeder zones.

The practical significance of the work lies in the possibility of applying the developed model to predict the behavior of lithium metatitanate (Li_2TiO_3) under operating conditions of breeder blankets in advanced fusion reactors such as ITER and DEMO. The obtained kinetic parameters and modeling methodology can be used to optimize design and operational parameters of breeder zones.

The reliability and validity of the obtained results are confirmed by publications in peer-reviewed international scientific journals with high impact factors, in journals recommended by the Science and Higher Education Quality Assurance Committee of the Ministry of Science and Higher Education of the Republic of Kazakhstan, as well as in the proceedings of international scientific conferences held in both nearby and distant abroad.

Personal contribution of the author. The author independently developed and implemented a comprehensive modeling approach describing tritium

generation, transport, and release processes in lithium metatitanate Li_2TiO_3 under neutron irradiation. The author performed neutron-physical analysis, numerical finite element modeling in COMSOL Multiphysics, sensitivity analysis of the model, identification of dominant tritium transport mechanisms accounting for spatial temperature non-uniformity, and verification of the model using in situ reactor experiment data. Formulation of research tasks, discussion, and physical interpretation of the obtained results were carried out jointly with scientific advisors.