

"Journal of Economics and Social Sciences"

Mode selection of deuterium flux supply through palladium-silver filter in experiments with Pb15.7Li eutectic at reactor irradiation

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Abstract

The paper presents preliminary results of experiments on permeability of deuterium through a palladium-silver filter. For conduction of the present study a differential method was chosen. The method consists in following: during reactor experiment into ampoule device with eutectic sample a spectrally pure deuterium flux was supplied constantly through palladium-silver filter. The description of the experimental setup and palladium-silver filter is presented. In this paper, the results of diffusion coefficients calculations, solubility, permeability of deuterium through a palladium-silver filter, activation energies of these processes and Arrhenius dependence are presented. Based on the calculations the modes supply of deuterium flux into ampoule device for experiments with Pb-15.7Li eutectic is determined.

Keywords: Pb15.7Li eutectic, deuterium flux, diffusion, solubility, permeability, palladium-silver filter;

1. Introduction

To demonstrate the commercial highlights of thermonuclear power, a project on building a DEMO reactor is being implemented in the world. A lead-lithium eutectic was selected as a functional tritium-generating material for use in the fuel cycle of the new reactor [2]. Currently several concepts of Test Blanket Modules (TBM) are developed: a) a helium-cooled lead-lithium blanket (European union) [3]; b) water-cooled lead-lithium blanket (USA, European Union) [1]; c) double-circuit lead-lithium blanket (USA) [4]. One of the terms for the successful application of the lithium-lead eutectic in fusion reactor is that tritium breeding should be maximally efficient, and the amount of it being leaked into the environment must not exceed the maximum allowable values.

Thus, there is a need to research the processes of tritium generation and breeding from lead-lithium in experiments on fission reactors. Currently, there is a limited number of survey works intended for the interaction of hydrogen isotopes with lead-lithium eutectics under the conditions of reactor irradiation. Within these studies, however, it has been found that tritium breeding from the eutectic is of complex nature and its breeding is significantly affected by the hydrogen isotopes in a purge gas [5]. To study this finding, the experiments on tritium generation and segregation from lead-lithium eutectics at a dynamic mode of supply of deuterium to the irradiation device with a Pb15.7Li eutectic will be performed at the IVG.1M reactor in the near future.

The experiments above are needed to implement adjustable deuterium supply into the irradiation device with a Pb15.7 Li eutectic. In turn, to do that it is required experimentally to

get temperature dependence of deuterium flux through the palladium-silver filter and to determine the main parameters of the hydrogen permeability using the data obtained.

2. Permeation technique

To achieve the goals and determine the parameters of deuterium permeability through the palladium-silver filter, the well-known permeability technique was used. The basic equations describing the mass transfer of hydrogen in metals are the Fick equations. For the one-dimensional case, they are as follows:

$$J(x,t) = -D(t)\frac{\partial C(x,t)}{\partial x};$$

$$\frac{\partial C(x,t)}{\partial t} = D(t)\frac{\partial^2 C(x,t)}{\partial x^2};$$
(1)

The first equation describes the rate of gas penetration through a unit on the surface of the medium. The second equation defines the hydrogen accumulation at a certain point of the medium as a function of time t.

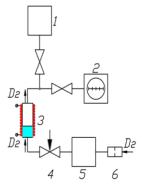
This paper recorded the flux J(t) and the amount of diffused material by mass spectrometry

$$Q(t) = \int_0^t J(t) dt. \tag{2}$$

Further, the main parameters of hydrogen permeability were calculated using the values obtained by an experimental approach.

3. Design and conditions of experiments

Design (figure 1) and conditions for experiments to determine the main parameters of deuterium penetration through palladium-silver filter (figure 2) depending on the temperature are given below.



1 – quadrupole mass spectrometer, 2 – turbomolecular pump, 3 – ampoule with lithium eutectic sample, 4 – leak valve, 5 – buffer tank, 6 – palladium-silver filter

1 – input, 2 – palladium-silver tube, 3 – case, 4 – dipper, 5 – isolation, 6 – output

Figure 2– Palladium-silver filter

Figure 1 – Design of experiment

Experimental conditions were as follows:

- the residual pressure in the measuring vacuum path -10^{-6} torr;
- the temperature of the experimental cell of AD from 25 to 30 °C;

- deuterium pressure supplied to the filter input 750 torr
- investigated temperatures range of the filter from 400 to 160 °C.

4. Experimental procedure

The experimental procedure of selecting the modes for supplying deuterium fluxes into the irradiation ampoule device was the mass spectrometric recording of deuterium partial pressure on the output of a palladium-silver filter. The parameters to be recorded are: temperature of palladium-silver tube, the flux of gas passing through the diffusion filter.

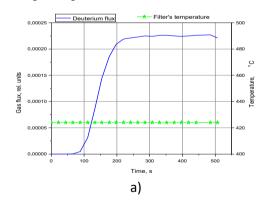
As a deuterium was delivered to the filter at pressure P_0 when measuring with the Deuterium permeability rate for filter, it turned out that there was a certain time called the delay time, after expiration of which the deuterium started to emerge from the filter at a constant rate. It is a quasi-steady state of the flux of deuterium that is required to record during these experiments.

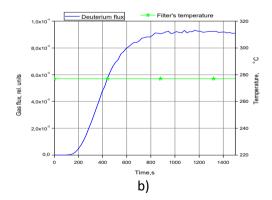
The algorithm for the experiments was as follows:

- at the beginning of the experiment, the high-vacuum path of the experimental LIANA bench, along with the ampoule device, was pumped to a pressure of around 10⁻⁶torr. The palladium-silver filter was also pumped from the output by a high-vacuum pump to a pressure of 10⁻⁶torr, and from the input side by the fore pump to a pressure of 10⁻⁴torr;
- the palladium-silver filter was heated to the first set temperature and deuterium was fed to the filter input from the cylinder, with the dripper completely open;
- at the output of the filter, the partial pressure of spectral pure deuterium passed through the experimental cell was recorded using the quadrupole mass spectrometer RGA- 100.
- installation of the deuterium flux at a quasi-stationary level (by mass spectrum) is followed by the filter temperature change, and the following measurements were made.

5. Experiments results

The experiments resulted in obtaining time dependences of the change in the deuterium flux on the output of palladium-silver filter and at different temperatures (see Figure Figure 3).





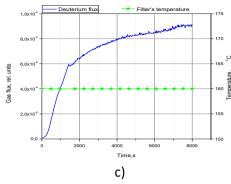


Figure 3 – Time dependences of the change in the deuterium flux on the output of palladium-silver filter at temperature of: a) 424 °C; b) 278 °C; c) 160 °C

The charts above show that as the temperature of the diffusion palladium-silver filter increases, the level of the deuterium flux increases proportionally and the rate of flux output to the quasistationary level rises.

6. Outcome analysis

Based on the obtained experimental data, the coefficients of diffusion, solubility, and permeability of deuterium for a palladium-silver filter were calculated and the temperature dependences of the variation of these constants were built (see Figures 4 and 5).

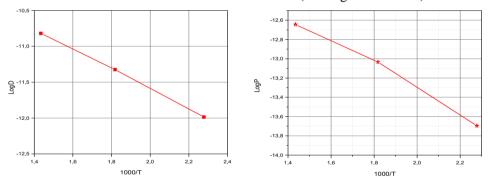


Figure 4 – Temperature dependence of the change in deuterium diffusion and permeability coefficients for a palladium-silver filter

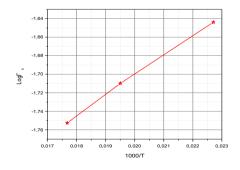


Figure 5 – Temperature dependence of the change in the deuterium solubility coefficient for palladium-silver filter

Arrhenius dependences of the deuterium diffusion, permeability and solubility coefficients for the palladium-silver filter, which were determined from the charts above (Figures 4, 5) are as follows:

$$D = 1 \cdot 10^{-9} \exp\left(\frac{-27 \frac{kJ}{mol}}{RT}\right) \tag{3}$$

$$P = 1.6 \cdot 10^{-11} \exp\left(\frac{-23 \frac{kJ}{mol}}{RT}\right) \tag{4}$$

$$S = 0.01 \cdot \exp\left(\frac{-2.4 \frac{kJ}{mol}}{RT}\right)^{\prime} \tag{5}$$

Based on the obtained results, the necessary temperature modes of diffusion palladium-silver filter were determined for implementing a controlled inflow of spectral pure deuterium into the irradiation device at the experiments with the Pb-15.7 Li eutectic at the IVG.1M reactor.

7. Conclusion

Thus, in the course of this work, the experiments were carried out, the results of which were as follows: the deuterium diffusion, solubility, and permeability coefficients calculated for palladium-silver filter were determined. Based on the calculations, the temperature modes of the diffusion filter and the deuterium fluxes were determined that corresponded to the following parameters: at filter temperatures of 424 °C , 278 °C and 160 °C, the deuterium fluxes through the experimental cell with a sample of Pb-15.7Li will be about $2.5 \cdot 10^{-10}$, $9 \cdot 10^{-11}$ and $3 \cdot 10^{-11}$ mol/s, respectively.

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