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Permeation of deuterium and tritium through the martensitic steel F82H

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Abstract

The permeation of deuterium and tritium through a membrane of the reduced-activation martensitic steel F82H was studied by the hydrogen permeability method on an installation at the RFNC-VNIITF. The permeability and diffusion coefficients were determined at the temperature range 490–1000 K and the pressure on the inlet side of the membrane range 800–5300 Pa. It is shown that the diffusion coefficient depends, especially in the beginning of studied temperature range, on particular conditions of preliminary annealing of the samples.

1. Introduction

The reduced-activation martensitic steel F82H is one of the most promising structural materials for fusion reactors [1,2]. Therefore this steel is under close attention of researchers concerned with the interaction between hydrogen isotopes and the material of the first wall and the blanket. As a result, a database on experimental properties of the F82H steel/hydrogen/deuterium systems is being formed [3–5]. In our opinion, this database is lacking the F82H steel/tritium system, which is most significant for fusion reactors. In this work we performed direct experiments on permeation of tritium through the F82H steel using the dynamic variant of the hydrogen permeability method over a temperature range of 490–1000 K and hydrogen isotopes pressure ranges of 800–5000 Pa.

2. Experimental technique

The material under study was a reduced-activation martensitic steel F82H. A sample of this steel was a disk 25.55 mm in diameter and 0.85 mm thick, which was treated under standard thermal conditions [5]. The disk was fitted in a test chamber made of the Fe–0.12C–18Cr–10Ni–0.1Ti steel using electron beam welding. Totally five groups of experiments were performed differing in thermal treatment conditions of the sample:

1. as-delivered sample degassed in a vacuum of 1×10^{-5} Pa for 24 h at 500 K;
2. sample annealed in a vacuum of 1×10^{-5} Pa for 10 h at 1050 K;
3. sample annealed in a flow of deuterium for 48 h at 1080 K and a gas pressure of 200 Pa and 1×10^{-5} Pa on the inlet and outlet sides of the sample, respectively.

The experiments on deuterium and tritium permeation were performed using the dynamic variant of the steady-state flux method [6]. In this case it is assumed that the initial concentration of hydrogen in the sample

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is zero, the hydrogen concentration on the inlet surface of the disk becomes instantaneously equal to an equilibrium solubility after hydrogen is fed, and the hydrogen concentration on the outlet surface of the disk is kept zero throughout the experiment.

The installation and the data processing method were described elsewhere [7].

The experiments of the group 1 were performed with deuterium in the range 490–630 K (lot 1/1) and 690–1000 K (lot 1/2) when the temperature changed from low to high values at each point. Before the experiments on the lot 1/2 the sample was heated in a vacuum of 1×10^{-5} Pa for 10 h at 900 K.

In the experiments of the groups 2 and 3 the temperature changed in most cases from high to low values. The experiments were realized by the following scheme: (1) degassing of the sample in a vacuum of $\simeq 1 \times 10^{-5}$ Pa at 1000 K until liberation of hydrogen stops (10–15 min); (2) stabilization of the sample temperature, input of the gas, recording of the flux at a constant pressure on the inlet side and at a vacuum on the outlet side of the sample-membrane until the flux is stabilized; (3) recording of the dependence of the stabilized permeating flux on the pressure on the inlet side of the sample at a constant temperature; (4) evacuation of the gas on both sides of the sample, heating to 1000 K and annealing until liberation of the gas stops; (5) the experiment is performed at the next temperature as described above.

3. Results and discussion

The experimental results were used to plot the temperature dependence of the parameters of permeability P (Fig. 1), diffusivity D (Fig. 2), and solubility K_S (Fig. 3) of deuterium and tritium in the F82H steel. Constants of

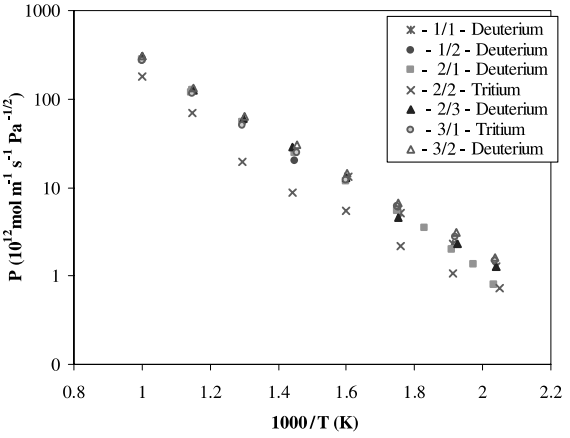

Fig. 1. Deuterium and tritium permeabilities for the F82H steel.

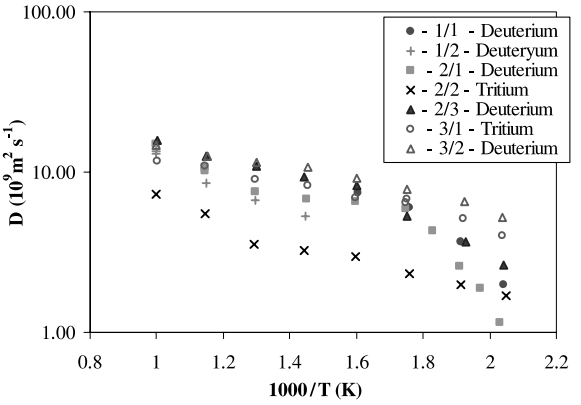

Fig. 2. Deuterium and tritium diffusivities in the F82H steel.

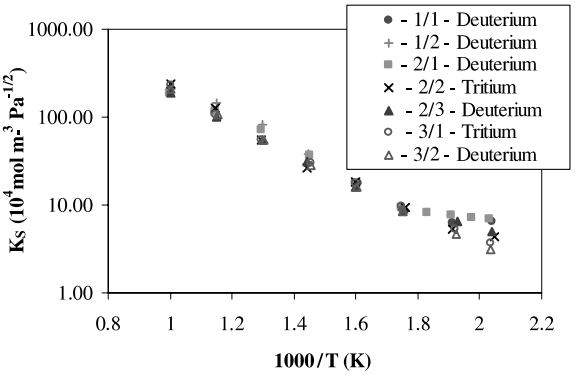

Fig. 3. Deuterium and tritium solubilities in the F82H steel.

the corresponding processes were calculated where possible (Table 1). To determine the pressure dependence of the flow of hydrogen isotopes penetrating through the membrane, permeability isotherms were plotted down to a pressure of $\simeq 50$ Pa. A characteristic shape of the isotherms is shown in Fig. 4. Since the isotherms are almost linear, then in our case the effect of surface processes is relatively small and the hydrogen diffusion represents the limiting stage of the penetration process.

From Fig. 1 it is seen that the temperature dependence of the permeability of the membrane material approaches the Arrhenius dependence at similar values of the activation energy in the experiments of each group. The temperature dependence of the permeability of the sample in the lot 2/2, which lies below the other dependences, disagrees with the general picture.

These results suggest that the structural state of the sample changed during the experiments thanks to temperature effects and possible oxidation of its surface. The structural state was stabilized by annealing the sample exposed to deuterium flow.

Table 1
Permeability, diffusion and solubility parameters

Curve no. in Figs. 1–3	Permeability parameters		Diffusion parameters		Solubility parameters	
	P_0 (mol m^{-1} s^{-1} Pa$^{-1/2}$)	E_P (kJ)	D_0 (m^2 s^{-1})	E_D (kJ)	K_S (mol m^{-3} Pa$^{-1/2}$)	E_S (kJ)
1/1, 1/2	4.7×10^{-8}	42.2				
2/1 – D_2	6.6×10^{-8}	45.4				
2/2 – T_2	2.3×10^{-8}	43.4			1.6[a]	35.6[a]
2/3 – D_2	5.6×10^{-8}	43.7			1.2[a]	34.5[a]
3/2 – T_2	3.4×10^{-8}	41.0	3.3×10^{-8}	7.8	1.13	33.0
3/1 – D_2	4.4×10^{-8}	41.6	3.9×10^{-8}	8.0	1.20	33.8

[a] In the temperature range 570–1000 K.

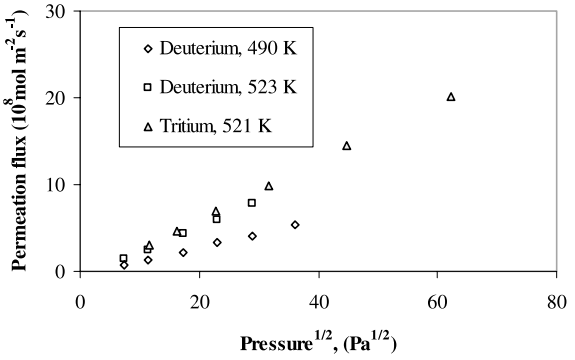

Fig. 4. Dependence of the stabilized permeating flux on the tritium pressure on the inlet side of the membrane.

A comparison of the results obtained for the lots 3/1 and 3/2 shows that the temperature dependencies of deuterium and tritium permeabilities of the F82H steel sample annealed in a deuterium flow are nearly coincident (Fig. 1 and Table 1).

An analogous picture is observed for the diffusion coefficients. One can see from Fig. 2 that a complicated temperature dependence of the logarithm of the effective diffusion coefficient (lots 1/1, 1/2 and 2/1) is simplified as the sample is heated (lots 2/2 and 2/3) and, finally, becomes almost linear (lots 3/1 and 3/2) with the same activation energy of the process and the ratio of the diffusion constants equal to 1.18 (the classical ratio, is 1.22, which is defined as the inverse ratio of the roots of the isotope masses.

The membrane material had an identical martensitic structure before and after the experiments (Fig. 5). All the effects did not cause recrystallization of the martensite. However, a large density of fine precipitates of secondary phases was observed in the final state (Fig. 5(b)). Although a detailed analysis is yet to be made, one may think that the observed precipitates represent carbides of vanadium and, probably, tungsten. Carbides, which are formed gradually at a temperature of 600–800 K, stabilize the concentration of dissolved carbon in the matrix, while a relatively long-time annealing at a higher temperature (1000–1080 K) facilitates the recovery and polygonization processes in the martensite. As a result, the density of dislocations decreases and a relatively stable subgrain structure is formed. The aforementioned processes lead to a stabilization of the microstructure and the phase composition of the steel under study. The stabilization shows up in hydrogen permeability studies as 'flattening' of the temperature dependences of the parameters of the permeability processes with the annealing time. A gradual (as the structural state is being stabilized) approach to the Arrhenius dependence is

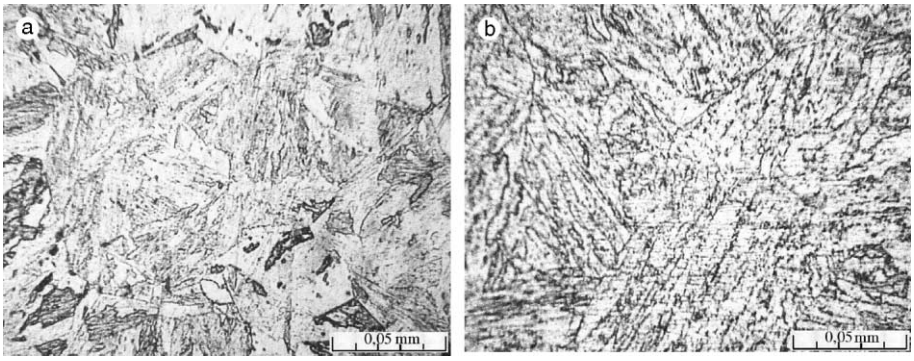

Fig. 5. Structure of the F82H steel: (a) initial condition; (b) after permeation experiments.

especially pronounced for the log D vs. $1/T$ curve, which is most sensitive to the structure (Fig. 2).

4. Conclusion

Measurements of deuterium and tritium fluxes were carried out through the sample of low-activated martensitic steel. Starting conditions of the sample for experiments were the following: as-delivered state, after vacuum annealing, and after annealing at deuterium flow. Obtained experimental results were used to determine the dependence of permeability, diffusivity, and solubility on temperature. Analysis of this dependence shows that the appearance changes on starting conditions of sample material and becomes Arrhenius-like under protracted thermal effect. It was proposed that the propellant power of the observed changes is the stabilization of the structural state (structure of dislocations, phase composition, and content of carbon in the matrix) of the investigated membrane material during annealing. A final conclusion about the reasons of changes in permeation parameters may be done after investigations of the tritium–steel interaction by the autoradiography and thermal desorption methods combined with structural control of the state of the sample material.

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References

[1] S. Mori, S. Yamazaki, J. Adachi, T. Kobayashi, S. Nishio, M. Kikuchi, M. Seki, Fus. Eng. Des. 18 (1991) 249.
[2] A. Hishimura, A. Kohyama, R.L. Klueh, D.S. Gelles, W. Dietz, K. Ehrlich, J. Nucl. Mater. 258–263 (1998) 193.
[3] E. Serra, A. Perujo, G. Benamati, J. Nucl. Mater. 245 (1997) 108.
[4] P. Jung, Fus. Technol. 33 (1998) 63.
[5] F. Schliefer, Chao Lui, P. Jung, J. Nucl. Mater. 283–287 (2000) 540.
[6] I.N. Bekman, I.E. Gabis, T.N. Kompaniets, A.A. Kurdyumov, V.N. Lyasnikov, in: Reviews of Electronics, Series 7, No. 1 (1084), CNII Elektronika, Moscow, 1985, 66 p.
[7] Yu. Dolinski, I. Lyasota, A. Shestakov, Yu. Reprintsev, Yu. Zouev, J. Nucl. Mater. 283–287 (2000) 854.