Deuterium Permeation Though Candidates Structural Materials For a Fusion Reactor

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Abstract. In the present work gas-driven and plasma-driven permeation through reduced activated ferritic-martensitic steel RUSFER-EK-181 (Fe-12Cr-2W-V-Ta-BC), austenitic steel ChS-68 (used in fast breeder reactor BN-600 as cladding) and V-4Ti-4Cr were investigated. The experimental facility is equipped with distributed ECRH plasma source, which was used for cleaning the inlet surface of membranes by argon ions with energy 300 eV and for deuterium plasma irradiation at plasma-driven permeation measurements (ion flux density is 10 A/m^2 at accelerating potential of –300 V). Deuterium gas-driven permeation was investigated in a pressure range of $5\cdot10^{-2}\div100$ Pa and in a temperature range of 600÷900 K. At gas-driven permeation the permeability coefficient of V-4Cr-4Ti membrane is about 4 orders of magnitude higher than that of RUSFER-EK-181. The permeability coefficient of ChS-68 is higher than permeability coefficient of RUSFER-EK-181 at all pressures and temperatures. However V-Cr-Ti alloys are promising a superpermeable membranes that can be used for membrane pumping of hydrogen providing significant compression and 100% separation of fusion fuel from He and other impurities.

1. Introduction

In the next step fusion devices such as DEMO and fusion neutron sources (FNS) high neutron fluxes and fluencies are expected [1]. In this case reduce activated (RA) and heat-resistant structural materials should be used. The most promising structural materials are ferritic-martensitic steels, vanadium alloys and silicon carbides, which are being developed in several countries.

In Russia V-(4÷10) Ti-(4÷10) Cr alloys, austenitic and RA ferritic-martensitic steels for fission applications are developed and manufactured in A.A. Bochvar High-technology Research Institute of Inorganic Materials [2]. For using this materials in next step fusion devices additional studies should be done. Permeation of hydrogen isotopes through structural material is one of the key material characteristic as tritium is a component of fusion fuel available only in small quantities, expensive and radioactive [3].

The data on hydrogen permeation through structural materials of fusion reactor are necessary for estimation of tritium flows trough vacuum vessel (to coolant, blanket, etc.). Temperature application window of most structural materials for fusion application is >600 K (relatively high!) and material temperature may be higher depending on coolant. At the same time the first wall of FNS should be as thin as possible for decreasing neutrons capture by structural materials. At the same time the permeation efficiency of materials increases substantially with increasing temperature and decreasing thickness.

2. Hydrogen isotopes permeation through steels

In the present work gas-driven permeation (GDP) and plasma-driven permeation (PDP) through austenitic steel ChS-68 (used in fast breeder reactor BN-600 as cladding) and V-4Ti-

4Cr were investigated at PIM facility (NRC "Kurchatov Institute"). This facility allows investigation of gas and plasma driven permeation through metals and porous samples. The scheme of PIM is presented at the Fig. 1.

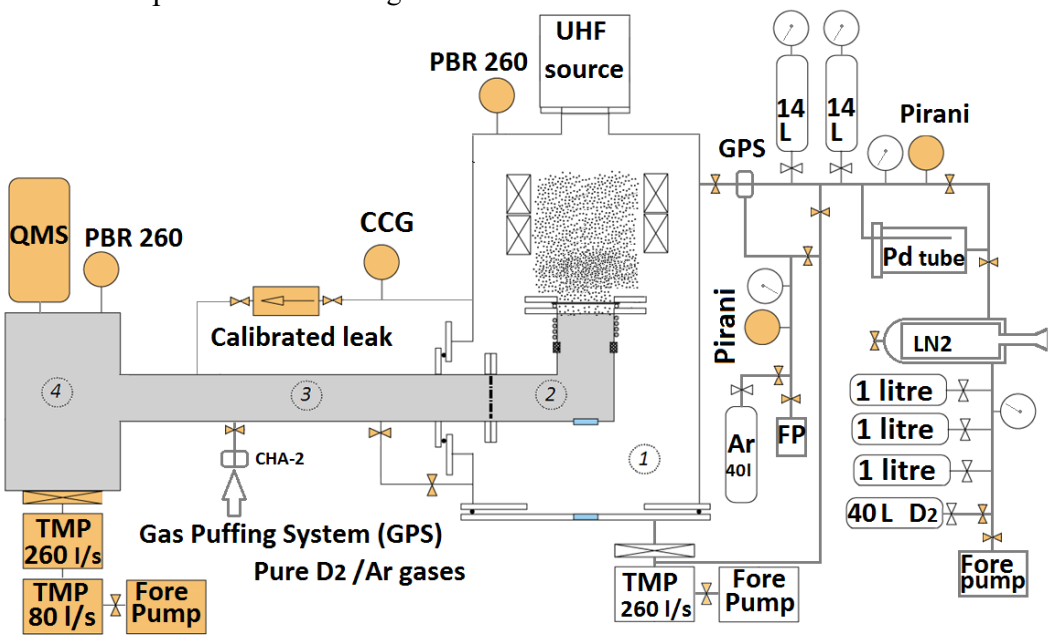


Fig. 1 –scheme of PIM facility

Installation consists of two ultra-high vacuum chambers with independent oil-free pumping systems separated by a membrane (flat or tube). Exposition chamber (1 at Fig. 1) contains a microwave plasma source, full-range (Pfeiffer PBR260), ceramic capacity gauges (CCG) (Pfeiffer CMR365, CMR364) and gas puffing system. Plasma source with electron cyclotron resonance heating allows to irradiate inlet surface of the membrane with low temperature plasma. Membrane is electrically insulated and potential (BIAS) up to -300 V can be applied. Typical deuterium plasma characteristics and background pressures are presented in Table 1. Three types of D ions present in plasma (D^+, D_2^+ and D_3^+). The concentrations of ions of different types was measured with time of flight mass-analyser.

Table 1 – PIM facility parameters

ECRH Electron temperature:	3-5 eV
Plasma composition: 71% ions D_3^+:D_2^+:D_1^+ =7:2:1 29% neutrals D_1^0	
Accelerating potential (BIAS), V	up to -300
Plasma density, cm^{-3}	up to $6 \cdot 10^{10}$
Plasma current on the sample, $A \cdot m^{-2}$	up to 20
Typical background in Registration chamber, Pa	$1 \cdot 10^{-9}$
Typical background in Exposition chamber, Pa	$2 \cdot 10^{-8}$
Calibrated leak gas conductivity, l/s	$2{,}123 \pm 0{,}012 \cdot 10^{-4}$

The permeation through ferritic-martensitic steel RUSFER-EK-181 (Fe-12Cr-2W-V-Ta-BC) was investigated early [4].

In the experiments RUSFER-EK-181 and ChS-68 tubes of 250 mm length with diameter of 6.85 mm and wall thickness of 0.4 mm (effective area 50 cm^2) were used as membranes. Photo of ChS-68 steel sample is presented on Fig. 2.

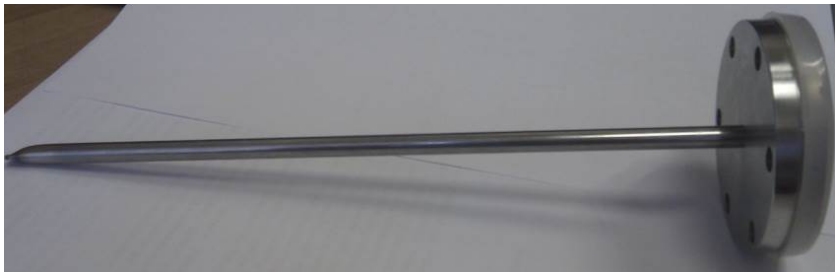


Fig. 2 – ChS-68 membrane arc-welded in a flange

Tube samples were welded with DN40CF flange by arc-welding (2 in Fig. 1) and welded the other end. Welds were tested by filling of exposition chamber with argon. No permeating argon flux was detected in registration chamber at temperature of the membrane up to 900 K.

The permeation flow in diffusion-limited regime (DLR) and surface-limited regime (SLR) can be described by the expressions:

$$J_{DLR} = \frac{DS\sqrt{p}}{L}\left(1+2\sum_{n=1}^{\infty}(-1)^n \exp(-\frac{D\pi^2 n^2}{L^2}t)\right), \quad (1)$$

$$J_{SLR} = S^2 p \frac{K_1 K_2}{K_1 + K_2}\tanh^2\left(t\frac{S}{L}\sqrt{pK_1(K_1+K_2)}\right), \quad (2)$$

where D – diffusion coefficient, S – solubility, p – the inlet pressure, L – sample thickness, K_1 and K_2 – recombination coefficients for the input and output surfaces respectively.

All measurements were carried out after sample inlet surface irradiation with argon plasma (300 V, 5–10 min). Deuterium permeation flow through ChS-68 steel Fig. 3 (a) was approximated by the expression (1), the diffusion coefficient was varied and the best approximation was obtained at $D = 3.9\cdot10^{-10}$ m^2/s. Fig. 3 (b) gives the dependence of steady-state deuterium permeation flux on the gas pressure through ChS-68 steel. As can be seen from the Fig. 3 diffusion-limited permeation regime can be achieved at pressures > 1 Pa.

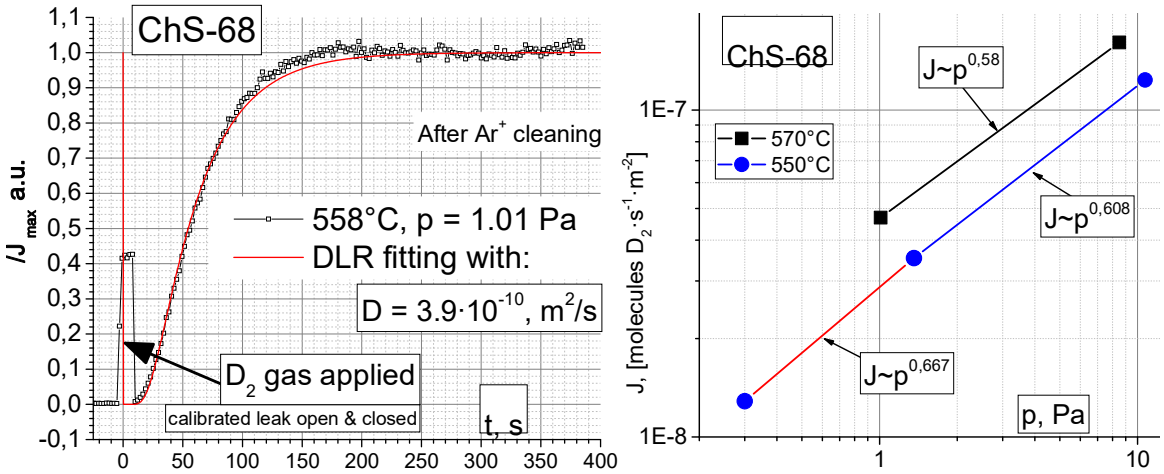


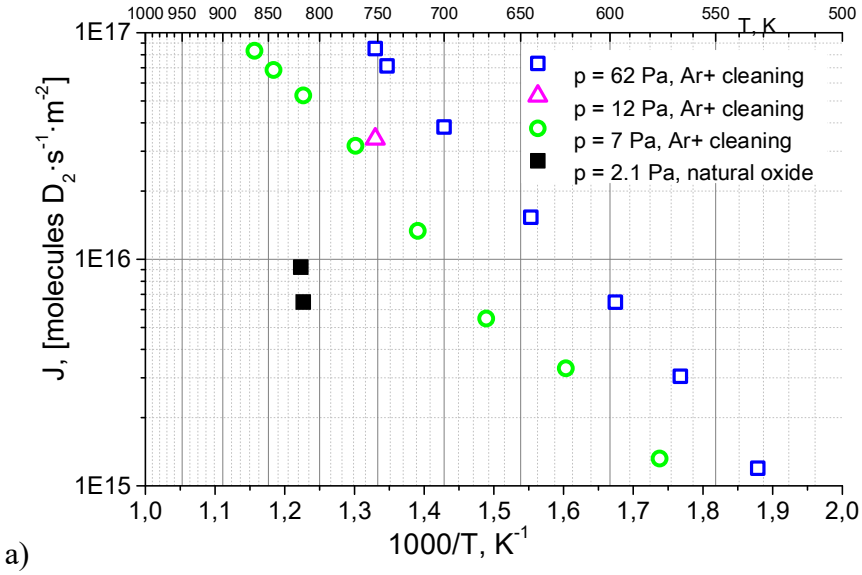
Fig. 3 – a) time dependence of deuterium permeation flow through ChS-68 steel;
b) pressure dependence of steady-state permeation flow of deuterium.
Data obtained after cleaning the inlet surface of membrane with 300 eV Ag ions. The permeation flow growth at 5-10 seconds due to QMS calibration procedure with a calibrated leak.

The temperature dependence of steady-state deuterium permeation flows through ChS-68 steel are given at Fig. 4 (a) for several pressures. The permeation constant (Fig. 4 (b)) was calculated from the Richardson equation:

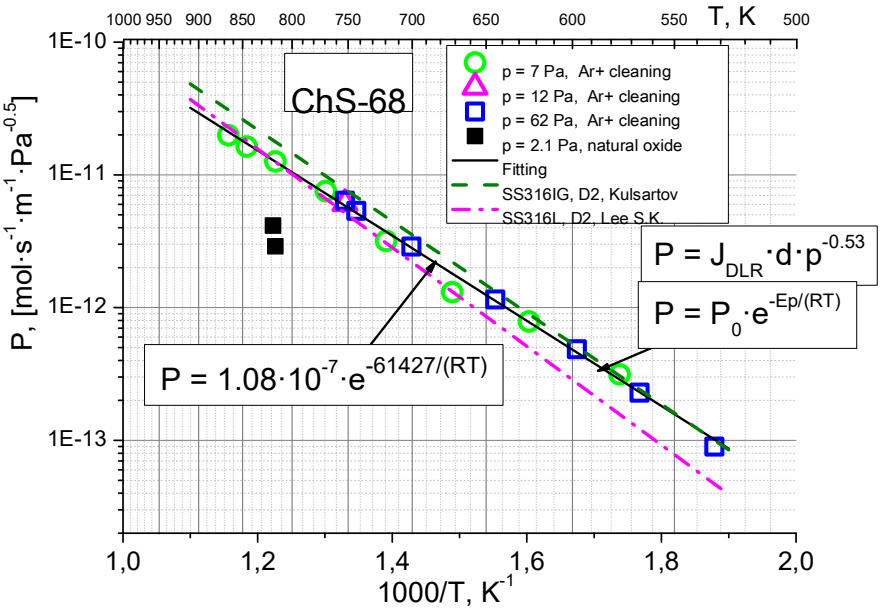
$$J_{DLR} = D \cdot S \cdot p^{0.5}/L = P \cdot p^{0.5}/L\,, \qquad (3)$$

where P = D·S – permeation constant (the product of the diffusion coefficient and solubility), p – inlet pressure, L – sample thickness.

One should mention a significant influence of an inlet surface condition on the deuterium GDP (Fig. 4 (b)). Permeation constant was increased 3-4 times after cleaning the inlet surface of the tube with Ar plasma (-300 V, 5-10 minutes). The obtained parameters of deuterium transport in the ChS-68 steel (diffusion coefficient $D = 2 \cdot 10^{-6} \cdot e^{-59382/(RT)}$ m^2/s and permeation constant $P = 1.08 \cdot 10^{-7} \cdot e^{-61427/(RT)}$, mole/(m·s·$Pa^{0,5}$)) are close to those for the austenitic steel SS316.



a)



b)

Fig. 4 – a) The temperature dependence of steady-state GDP flow of deuterium through ChS-68 steel at 7-62 Pa. b) The temperature dependence of ChS-68 steel permeation constant as compared with literature data on permeation constant of SS316L [5].

3. Gas and plasma driven permeation of hydrogen isotopes through V-4Cr-4Ti

Safety and efficient control of the DT fuel fluxes is an essential issue for the development of steady-state fusion devices like DEMO or FNS. The purpose of fuel cycle optimization is a reduction the total amount of DT fuel. For this purpose the use of the fuel technologies of fast and steady-state separation and purification of DT fuel fluxes is required [6]. For increase the efficiency of fusion fuel cycle the superpermeable metal membranes can be used that provide a significant compression effect for hydrogen and 100% separation of fuel from ash [7]. The V-group metals (V, Nb, Ta) are preferred for use due to the high hydrogen permeability. The membrane pump should be located close to the divertor and will be exposed to fusion neutrons. The V-Cr-Ti alloys are promising materials for fusion and fission applications due to their low activation by neutrons, high strength at elevated temperatures, and compatibility with liquid lithium. A prototype of the membrane pump has been developed and tested [8] with the use of a hot wire as an atomizer. However, lifetime of the atomizer is quite limited while the maintenance of a membrane pump under neutron irradiation is preferable.

The V-4Ti-4Cr membrane was a hot rolled 100 microns thick foil with a diameter of 60 mm welded between thing ring and a cup of stainless steel by arc welding in argon atmosphere. GDP experiments with deuterium were performed in temperature and pressure ranges 573÷823 K and $5\cdot10^{-2}$÷10 Pa respectively. The distributed electron-cyclotron resonance heating plasma source was used for deuterium plasma irradiation of membrane during PDP measurements at potential of -100 V. The current density of deuterium ions irradiating membrane was about $5\cdot10^{19}$ ions/($m^2\cdot s$). The deuterium pressure at PDP measurements was 0.1 Pa. PDP experiments with deuterium were carried out in the temperature range of 600÷823 K.

At GDP experiments diffusion-limited regime (DLR) was obtained. Permeation constant in DLR can be estimated from equation (3). The dependence of permeation constant from temperature is presented on Fig. 5.

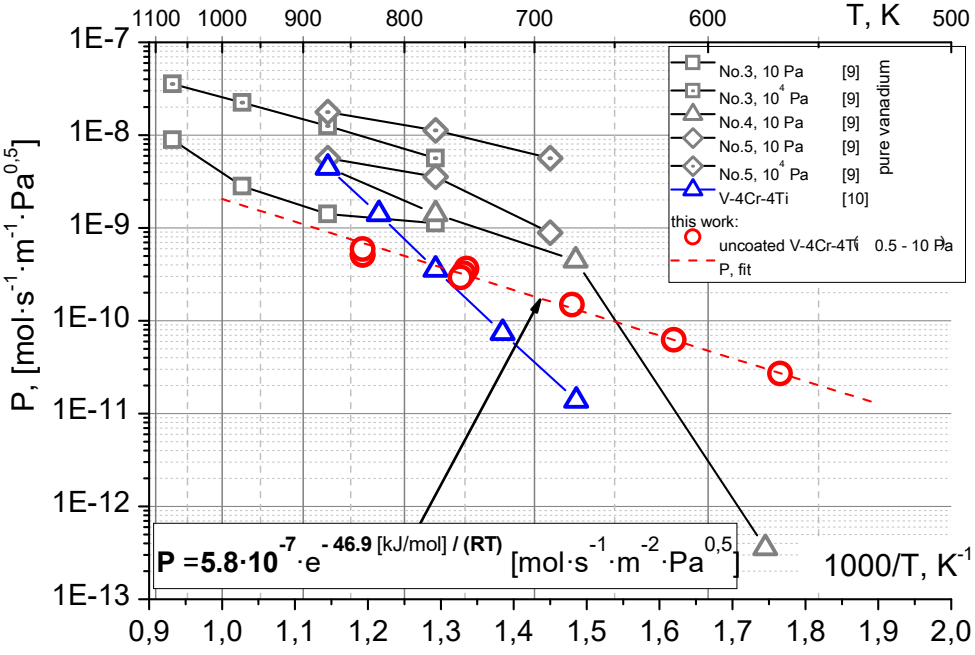


Fig. 5 Comparison of deuterium permeation constant of V-4Cr-4Ti with literature.

The measured PDP flux under deuterium plasma irradiation was an order of magnitude higher than the permeating flux from gas at the same pressure and temperature. The permeation efficiency η (the ratio of deuterium penetrating flux to the flux of plasma ions irradiating the inlet surface of the membrane) increases with the membrane temperature and reached $\eta = 0.1$ at 800 K. Two series of experiments were performed. In the first one temperature of sample was increased step by step, in the second one – decreased. In both series extracted deuterium ion fluxes from plasma were about $5 \cdot 10^{19}$ ions/($m^2 \cdot s$). Temperature dependence of permeation efficiency constant is presented on Fig. 6.

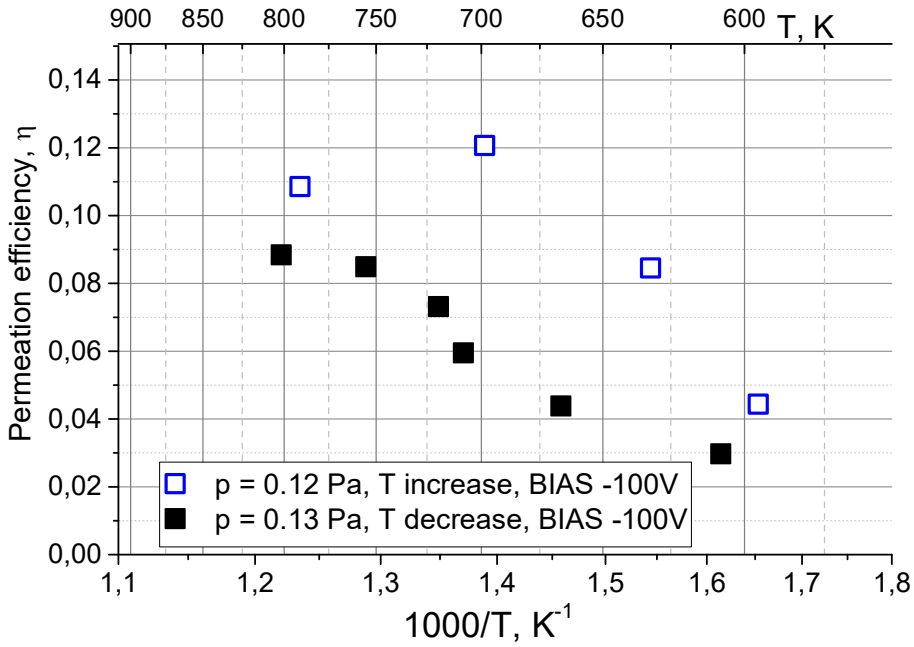


Fig. 6 Dependence of permeation efficiency constant of V-4Cr-4Ti on temperature.

4. Conclusions

Austenitic steel ChS-68 and RAFMS RUSFER-EK-181 are both acceptable as a structural material for a fusion reactor. Ferritic-martensitic steel RUSFER-EK-181 is preferable for a fusion reactors with a high neutron load to structural materials due to low-activated properties and low swelling under neutron irradiation. This steel seems only feasible structural material for a commercial hybrid fusion-fission reactor. Austenitic steel (such as ChS-68) was widely used in nuclear facilities and preferred for use as structural material in demonstration hybrid reactor (DEMO-FNS) with expected dose of radiation damage up to 20-50 dpa due to the wide experience in the use of these steel at nuclear applications.

Vanadium alloy is exceptionable to use as a structural material for a fusion reactor due to extremely high hydrogen permeability. However V-Cr-Ti alloys are promising materials for a membrane pump as a superpermeable metal membranes that can be used for providing a significant compression for hydrogen and a 100% separation of fuel from He and other impurities.

The comparison of GDP measurements are given at Fig. 7. At GDP permeability coefficient of V-4Cr-4Ti membrane is about 4 orders of magnitude higher than that of RUSFER-EK-181 while permeability coefficient of ChS-68 is higher than permeability

coefficient of RUSFER-EK-181 at all pressures and temperatures. The features of such properties are discussed.

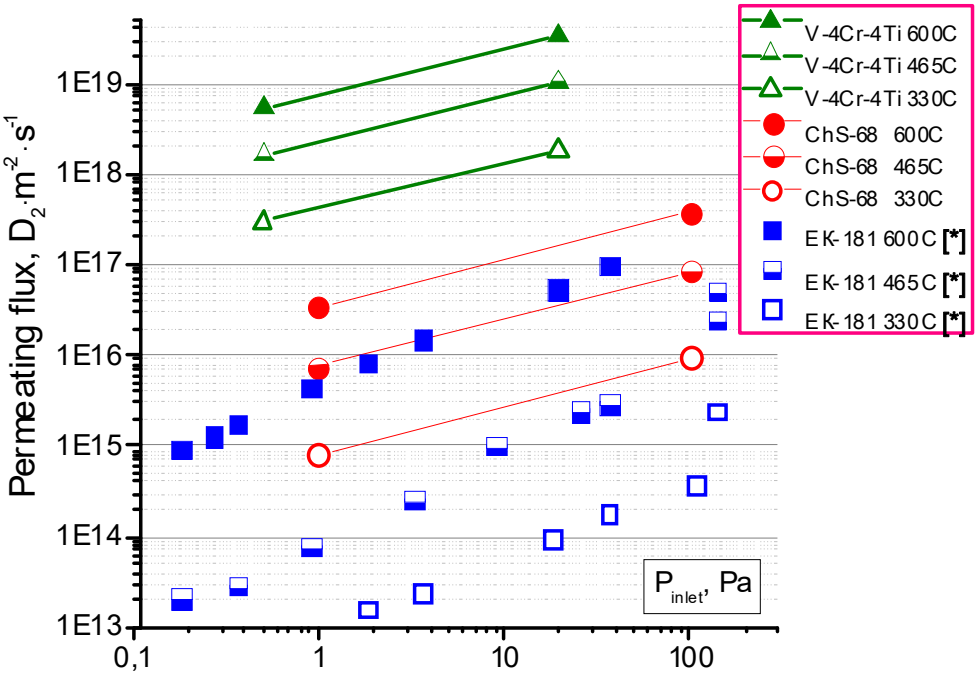


Fig.7. Permeation flux through ferritic-martensitic steel RUSFER-EK-181 (0.5 mm), austenitic steel ChS-68 (0.5 mm) and V-4Ti-4Cr alloy (0.1 mm) at different temperatures and deuterium pressure.

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