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Diffusive transport parameters of deuterium through China reduced activation ferritic-martensitic steels



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ABSTRACT

Reduced Activation Ferritic/Martensitic (RAFM) steels have been considered as the most promising candidate structure materials for a fusion reactor. In the recent decades, two new types of RAFM steels, called China Low Activation Martensitic (CLAM) steel and China Low-activation Ferritic (CLF-1) steel, have been developed. The gas evolution permeation technique has been used to investigate diffusive transport parameters of deuterium through CLAM and CLF-1 over the temperature range 623 ~ 873 K at deuterium pressure of 10⁵ Pa. The resultant transport parameters are: Φ (mol. m⁻¹ s⁻¹ Pa^{-1/2}) = 5.40 \times 10⁻⁸ exp (-46.8 (kJ. mol⁻¹)/RT), $D(m^2 s^{-1}) = 3.81 \times 10^{-7} \exp(-24.0 (kJ. mol⁻¹)/RT)$ and S (mol. m⁻³ Pa^{-1/2}) = 1.42 \times 10⁻¹ exp(-22.8 (kJ. mol⁻¹)/RT) for CLAM; while Φ (mol m⁻¹ s⁻¹ Pa^{-1/2}) = 1.76 \times 10⁻⁸ exp(-43.9 (kJ. mol⁻¹)/RT), $D(m^2 s^{-1}) = 1.02 \times 10^{-7} \exp(-16.9 (kJ. mol⁻¹)/RT)$ and S(mol. m⁻¹ Pa^{-1/2}) = 1.73 \times 10⁻¹ exp(-27.0 (kJ. mol⁻¹) /RT) for CLF-1. The results show that CLAM is more permeable than CLF-1, thus it is easier for hydrogen isotopes to transport and be removed.

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1. Introduction

China Low Activation Martensitic (CLAM) steel [1] and China Low-activation Ferritic (CLF-1) steel [2] are two types of reduced activation ferritic/martensitic (RAFM) steels considered to be the most promising candidate structure material for a fusion reactor. Their outstanding resistance to irradiation and favourable thermophysical properties have been well investigated [2–6]. They can be fabricated on an industrial scale up to tons.

In a fusion reactor blanket material, hydrogen isotopes are generated by neutron transmutation of some impurities like carbon and nitrogen, the diffusion/permeation of gaseous hydrogen isotopes from the tritium breeders, implantation by D-T plasma and chemical radiolysis of the cooling water. These alien hydrogen isotope atoms are strongly prone to be trapped by the lattice defect sites like vacancies, voids in the blanket metals [7], which may lead to an increase of hydrogen retention and an impediment of the forward migration of hydrogen atoms [8]. In order to evaluate the influence brought by the retention of hydrogen isotopes, the hydrogen transport in the material must be analysed because it determines the retention of hydrogen isotopes including the precious and radioactive tritium fuel.

In this work, the deuterium diffusive transport parameters of permeability, diffusivity and Sieverts' constant in CLAM and CLF-1 are experimentally measured by gas evolution permeation technique over the temperature range $623 \sim 873$ K at deuterium pressure of 10^5 Pa.

2. Experiment

2.1. Sample preparation

CLAM was supplied by Institute of Nuclear Energy Safety Technology, and the as-received raw material of CLAM underwent the following heat treatment: austenitizing at 1253K for 30 min, quenching in water, tempering at 1033K for 90min, slow cooling in the air [1]. The as-treated material was manufactured to thin discs with 1 mm thickness and 25 mm diameter. The both sides of specimens were mechanically polished by a sequence of fine grain silicon carbide abrasive papers and finally a 3.5 μ m, 2.5 μ m, 1 μ m diamond paste. Then the specimens were ultrasonically degreased with acetone and ethanol in sequence.

CLF-1 was obtained from Southwestern Institute of Physics, and the as-received raw material of CLF-1 had undergone the similar



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lable l	
Main elemental com	positions of CLAM and CLF-1.

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	Fe	Cr	W	V	С	Mn	Та	0	Si	Ν
CLAM	Balance	9.0	1.5	0.2	0.1	0.45	0.07	-	0.01	0.02
CLF-1	Balance	8.5	1.6	0.3	0.12	0.6	0.1	0.001	—	-

heat treatment to CLAM: austenitizing at 1253K for 45 min, quenching in the air, tempering at 1013K for 90min, slow cooling in the air [6]. Then the same disc with 1 mm thickness and 25 mm diameter had been fabricated and accompanied by the same mechanical polish process.

The main elemental compositions of two types of RAFM steels are shown in Table 1.

2.2. Permeation equipment

The gas evolution permeation technique has been used to obtain the diffusive transport parameters of deuterium through the two types of China RAFMs. The details are described as below.

The gas evolution permeation experiment has been done on the gas-solid interaction system(Fig. 1) which can measure the deuterium transport parameter through a thin membrane of material. Before the measurement, we utilize a series of standard leaks to calibrate the quadrupole mass spectrometer (PFEIFFER QMG 422I) and make the relationship of leakage and ion current. The equipment is separated by the sample, which consisting of two parts: high pressure region and low pressure region. The sample is sealed by tungsten-arc inert-gas welding(TIG). In the high pressure region (permeation inlet side) where the base pressure is about several Pa, the deuterium pressure is monitored by a capacitance diaphragm gauge (PMA Transmitter P40), while in the low pressure region (permeation outlet side) where the base pressure can be low to 10^{-5} Pa, the gas permeating through the sample is ionized in the quadrupole mass spectrometer whose ion current is recorded. The deuterium in a tank with an isotopic abundance of 99.7% is filled up in the chamber of the permeation inlet side within two or 3 s. kept at a fixed pressure of 10^5 Pa with an error under 5% through adjusting the valves and the evacuation pump. And the temperature of the sample is controlled well with an deviation of less than 1K in a set value from 623K to 873K.

3. Theory

Hydrogen isotopes adsorb and dissolve on the material surface by the process below:

$$H(D,T)_{2, gas} \Leftrightarrow H(D,T)_{2, ads} \Leftrightarrow 2H(D,T)_{sol}$$
(1)

whose concentrations are represented on the basis of Sieverts' law:

$$c = S \cdot \sqrt{p} \tag{2}$$

where, c represents the surface concentration of hydrogen isotope atoms with 0 and t as the coordinate and time, respectively; p denotes the pressure of gaseous hydrogen isotopes.

The permeation of hydrogen through a membrane in one dimension can be expressed in term of diffusion flux using the hydrogen concentration gradient with Fick's first law:

$$J = -D\frac{\partial c}{\partial x} \tag{3}$$

For the case of steady-state permeation process, taking account of the rate process limited by diffusion in the bulk rather than surface reaction [9], the diffusion flux J mentioned above, which is the amount of substance per unit area per unit time, can be expressed by Richardson's law [10], namely:

$$J = D \frac{S \cdot \left(\sqrt{p_{\rm h}} - \sqrt{p_{\rm l}}\right)}{d} \tag{4}$$

$$\underline{D \times S \text{ denoted as } \Phi} \quad \frac{\Phi}{d} (\sqrt{p}_h - \sqrt{p}_l) \tag{5}$$

$$\underline{\underline{p}_h \gg p_l \approx 0} \quad \frac{\Phi}{d} \sqrt{p_h} \tag{6}$$

where, D is the diffusion coefficient of the material with the thickness d, S is the Sieverts' constant, Φ is the product of D and S which represents permeability and p_h , p_l are the gas pressure on the high and low sides respectively. If $p_h \gg p_l \approx 0$, p_l will be omitted as usual.

D, S, Φ all abide by Arrhenius behaviour with temperature. D, Φ



Fig. 1. Schema of permeation equipment. P1: scroll pump, P2: scroll pump, P3: turbomolecular pump, P4: titanium sublimation pump, SL: standard leak, QMS: quadrupole mass spectrometer, V1: ultra high vacuum chamber, V2: analytic chamber.

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