Journal of Nuclear Materials 511 (2018) 231–234

Contents lists available at ScienceDirect

Journal of Nuclear Materials

journal homepage: www.elsevier.com/locate/jnucmat

# Hydrogen diffusive transport parameters through CLAM steel

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ARTICLE INFO

*Article history:*
Received 19 June 2018
Received in revised form
2 September 2018
Accepted 9 September 2018
Available online 11 September 2018

*Keywords:*
CLAM
Hydrogen permeation
Diffusive transport parameters

ABSTRACT

China Low Activation Martensitic (CLAM) steel has been selected as the primary candidate structural material for Chinese Helium Cooled Ceramic Breeder ITER Test Blanket Module. In the present work, the hydrogen transport behavior in CLAM steel have been experimentally measured with gas evolution permeation technique in the temperature range of 573–823 K at hydrogen pressures of $10^2$–$10^4$ Pa. The pressure exponent n of permeation was between 0.54 and 0.55 and it showed that the permeation was approximately pure diffusion limited. The hydrogen transport parameters of CLAM were presented based on experimental results at $10^4$ Pa, and the transport parameters were $\Phi(\mathrm{mol \cdot m^{-1} \cdot Pa^{-1/2} \cdot s^{-1}}) = 3.11 \times 10^{-8}\exp(-38712/RT)$, $D(\mathrm{m^2 \cdot s^{-1}}) = 1.19 \times 10^{-7}\exp(-16404/RT)$, $Ks(\mathrm{mol \cdot m^{-3} \cdot Pa^{-1/2}}) = 0.27\exp(-22308/RT)$. All those transport parameters were compared with the available data corresponding to several other RAFM steels and the results shown the similar hydrogen transport behavior of the RAFMs.



## 1. Introduction

Reduced activation ferritic/martensitic (RAFM) steel is considered to be one of the preferred structural materials for fusion reactors due to its comprehensive advantages of high resistance to irradiation, low neutron activation, good thermal properties, high thermal conductivity and many other advantages [1,2]. China Low Activation Martensitic steel (CLAM) developed by Institute of Nuclear Energy Safety Technology (INEST), Chinese Academy of Sciences (CAS), is selected as the primary candidate structural material for Chinese helium ceramic breeder (HCCB) Test Blanket Modules (TBM) for ITER and China fusion engineering test reactor (CFETR) blanket [3,4]. After continuous development and research, CLAM steel has achieved 6.4 tons scale industrialization level of melting and processing [5,6]. A lot of efforts are being devoted on the R&D of CLAM in fabrication process, property evaluation and database establishment [7].

Hydrogen isotopes, generated from neutron transmutation, tritium breeders [8,9], and D-T plasma, will diffuse into material and lead to an abominable embrittlement effect. The permeation of hydrogen isotopes through structural walls even affect isotopes mixture balance in plasma and cause the radioactive contamination. All of these problems that affect fuel economy, radioactive safety and plasma operation of fusion reactors need to be evaluated before reactor construction [10]. Therefore, hydrogen isotope transport must be analyzed and characterized, and the deuterium diffusive transport parameters of CLAM steels have been measured [11].

In this work, hydrogen transport parameters of diffusivity, permeability and Sieverts' constant in CLAM have been measured with gas evolution permeation technique. Measurements were conducted in the temperature range of 573–823 K at hydrogen driving pressure of $10^2$–$10^4$ Pa. The experimental results were compared with those of the other candidate RAFM steels, such as EUROFER97 [12] and F82H steel [13].

## 2. Experimental

The material studied is CLAM steel (HEAT 0912), and the main chemical compositions are listed in Table 1. The ingot was hot-forged and rolled into plates with different thicknesses. The standard heat treatment was performed [7]: normalization (980 °C/

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https://doi.org/10.1016/j.jnucmat.2018.09.014
0022-3115/

**Table 1**
Main chemical compositions of RAFMs (wt%).

| Materials | Cr | W | V | Ta | Mn | Si | C | S | P | Fe | Ref. |
|---|---|---|---|---|---|---|---|---|---|---|---|
| CLAM | 8.87 | 1.48 | 0.21 | 0.13 | 0.47 | 0.05 | 0.092 | 0.0022 | 0.003 | Bal. | T.W. |
| F82H | 7.7 | 1.94 | 0.16 | 0.02 | 0.16 | 0.11 | 0.09 | 0.002 | 0.002 | Bal. | [18] |
| Eurofer 97 | 9.0 | 1.1 | 0.15–0.25 | 0.06–0.09 | 0.4 | 0.05 | 0.11 | <0.005 | <0.005 | Bal. | [20] |

30 min/air cooling) and tempering (760 °C/90 min/air cooling). Specimens with size of Φ20 × 0.5 mm were cut from the plates. Then these specimens were grinded with abrasive papers and polished with diamond lapping paste down to thicknesses of 0.45–0.5 mm. Pd-coating with thinness of about 0.3 μm was deposited on the both sides by magnetron sputtering, $Ar^+$ ion etching was done before the deposition.

The disk-shaped specimen sealed with two O-rings divided the device into two parts: the upstream (high-pressure volume) and the downstream (low-pressure volume). The pressure of the upstream was measured by Baratron capacitance manometer and a ionization gauge was installed at the downstream for checking vacuum. The analyses of the permeated gas were carried out with a quadrupole mass spectrometer. The hydrogen was filled up in the chamber of the permeation inlet side within 3 s and kept at a fixed pressure of $10^4$ Pa with an error less than 3%. An electrical resistance furnace was used to heat the sample to a temperature among 573K and 823K with a deviation of less than 1K.

## 3. Theory

Hydrogen molecule adsorbs on the material surface and dissolves to hydrogen atom, the permeation of hydrogen atom through a membrane with uniform thickness d (m) can be expressed by a theoretical expression of flux J ($mol\cdot m^{-2}\cdot s^{-1}$), in the case where the driving pressure increases instantly from 0 to p (Pa), which is very much higher than the pressure on the downstream [14,15], and where the initial concentration throughout the membrane is 0. J(t) can be expressed by the one-dimensional solution of Fick's law [16].

$$J(t) = \frac{DK_S p^{1/2}}{d}\left[1 + 2\sum_{n=1}^{\infty}(-1)^n \exp\left[-D\frac{n^2\pi^2}{d^2}t\right]\right] \tag{1}$$

where D ($m^2\cdot s^{-1}$) is the diffusivity and $K_S$ ($mol\cdot m^{-3}\cdot Pa^{1/2}$) is Sieverts' constant, t (s) is time. It is assumed that the diffusivity D is independent of concentration, and the process of limiting diffusion rate is the bulk diffusion rather than the surface reactions [14,17].

When the t tends to infinity (steady-state), Eq. (1) will become $J = \Phi \cdot P^{1/2}/d$, where $\Phi = D \cdot K_S$ ($mol\cdot m^{-1}\cdot s^{-1}\cdot Pa^{-1/2}$) and Φ is considered to be the permeability of the membrane. The total amount of permeated gas Q(t) (mol) is expressed by the following equation [18]:

$$Q(t) = \int_0^t J(t')dt' = \frac{\Phi p^{1/2}}{d}t - \frac{\Phi p^{1/2}d}{6D} - \frac{2\Phi p^{1/2}d}{\pi^2 D} \times \sum_{n=1}^{\infty}\frac{(-1)^n}{n^2}\exp\left(-D\frac{n^2\pi^2}{d^2}t\right) \tag{2}$$

Where Q(t) is obtained by using the ideal gas law from the experimental data of pressure rise.

The value of permeability Φ can be obtained from the permeation flux at steady state and diffusivity D is calculated from the time lag $t_L = d^2/6D$, where the $t_L$ (s) is the time that the line by fitting the pressure rise curve intersects with the time axis. The Sievert's constant $K_S$ can be directly derived from the quotient of Φ/D.

The process of capture and release hydrogen atoms by some sites delay the hydrogen tansport in the membrane, and is called trapping effect [19]. Then the lattice occupied flux can be described as following by Fick's first law:

$$J = -D_L\frac{\partial c_L}{\partial X} \tag{3}$$

The diffusion equation exhibiting the trapping effect in the lattice can be expressed as:

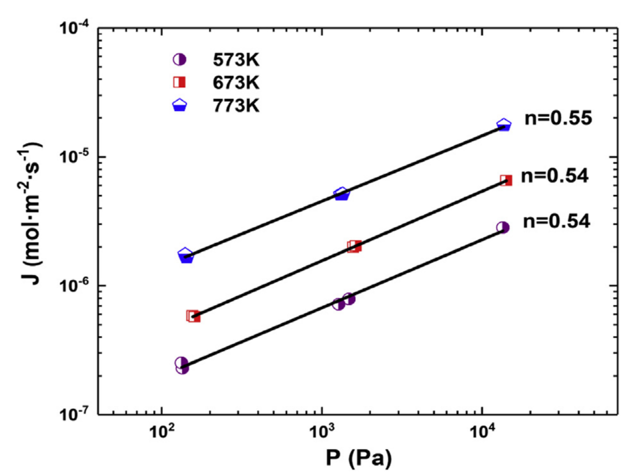


**Fig. 1.** Driving pressure dependence of hydrogen permeation flux.

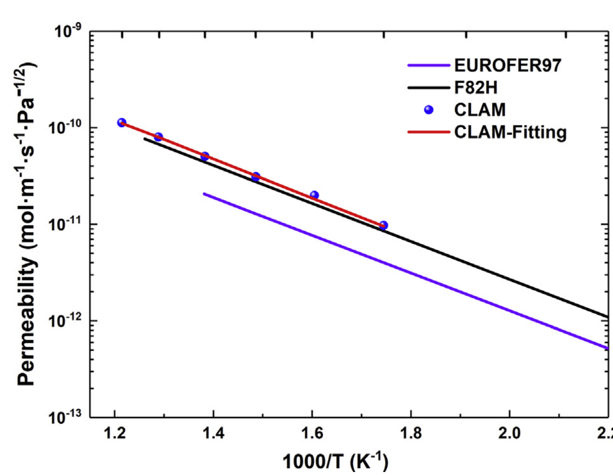


**Fig. 2.** Hydrogen permeability in CLAM and other RAFMs [18,20].