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Deuterium permeation behavior of HTUPS4 steel with thermal oxidation layer

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ABSTRACT

The permeation behavior of creep-resistant, Al_2O_3-forming HTUPS austenitic stainless steels was studied using a gas driven permeation (GDP) device. The steel samples were first thermal oxidized at air condition, followed by GDP experiments. The permeability and diffusion coefficients of oxidized samples and bare 316L steels were derived and compared. In order to characterize the oxide layer, X-ray photoelectron spectroscopy was performed. An oxide layer with a thickness of 200 nm which mainly consists of Al_2O_3 was detected.

1. Introduction

For the design of fusion device, one of the most important requirements is the safe handling of tritium [1,2]. For materials working in tritium environment, an additional tritium permeation barrier (TPB) may be necessary to reduce tritium leakage. Various methods were used to prepare TPB and thermal oxidation of Al-based alloy shows its advantages over the others: (1) self-healing property of irradiation damages and (2) capability to make TPB on complicated components surface [3].

A recently developed high-temperature ultrafineprecipitation-strengthened steels (HTUPS) have such thermal oxidation ability. The HTUPS steels consist of nominally 14 wt% Cr, 2 wt% Al dissolved in a face-centered-cubic (fcc) iron lattice and nanodispersed NbC as the strengthening phase. From the previous investigation, a layer of α-Al_2O_3 with a thickness of 40–50 nm and a 60–100 nm thick intermixed layer of transition $Al_2O_3 + Cr_2O_3$ + porosity could be formed if oxide condition is proper [4]. As Cr_2O_3, Al_2O_3 are candidate materials for tritium permeation barrier [5,6], the HTUPS steels, except for good high-temperature creep strength and resistance for corrosion, could have an efficient hydrogen permeation reduction performance. These outstanding properties suggest that HTUPS type steels are feasible candidate structural materials to manufacture components of the tritium loop where receives no high dose neutrons [7].

In this paper, the deuterium permeation behavior of the HTUPS 4 steel is mainly investigated by a gas driven permeation (GDP) device. The effect of thermal oxidation is covered.

2. Experimental

2.1. Material preparation

The material chosen in the experiments is the HTUPS 4 steel, which is one type of the HTUPS steels. Disks with dimensions of Φ12 mm × 0.2 mm were cut and mechanically ground down to #2000 SiC papers. Then the disks were ultrasonically cleaned in acetone and alcohol. The oxidation was carried out at 1273 K for 150 h under air condition. The material chosen for comparison is 316L stainless steel from Goodfellow Cop. The detailed chemical composition of these two materials is shown in Table 1.

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Table 1
Detailed chemical composition of the HTUPS 4 and 316L stainless steel samples.

Elements	HTUPS 4 wt%	316L wt%
Fe	57.78	69
Ni	19.95	10
Cr	14.19	18
Al	2.48	–
Si	0.15	–
Mn	1.95	–
Mo	2.46	3
Nb	0.86	–

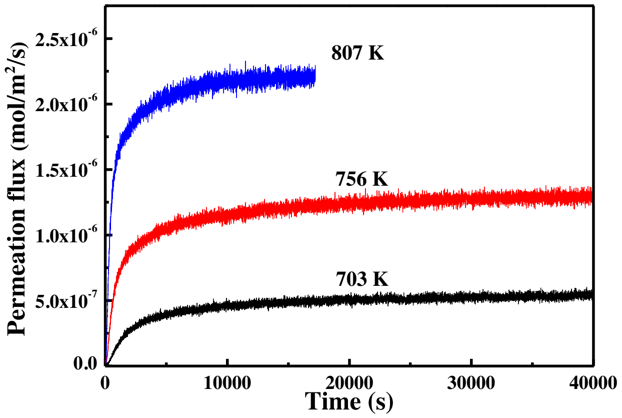

Fig. 1. Deuterium permeation flux vs. time of oxidized HTUPS 4 steel. The upstream driven pressure was 1.3 × 10^5 Pa.

2.2. Deuterium GDP experiments

Deuterium GDP experiments were done in the device constructed in our lab. We used a VCR couple produced by Swagelok Company to hold the sample. The sample can be heated by an electrical furnace with a temperature range from RT to 1473 K and a temperature stability of ±2 K. A thermocouple was placed into the leak hole of the VCR couple to monitor the temperature in real time. Before the experiments, a vacuum level of 10^{-5} Pa can be obtained by two mechanical pumps and two molecular pumps. At the beginning of the GDP experiment, deuterium gas can be introduced into the upstream by a needle valve, while a capacitance diaphragm gauge was used in the meantime to monitor the pressure of the upstream, which can help to get an fixed, known upstream pressure ranging from 100 Pa to 1.3 × 10^5 pa. The permeation signals in the downstream, mainly HD and D_2, were measured as a function of time using a quadrupole mass spectrometer (QMS). The HD and D_2 signals were calibrated using a standard D_2 calibration leak. Detailed description of the GDP device and the experiment procedure can be found in a previous paper [8]. For this study, the experimental temperature ranges from 700 K–800 K.

2.3. X-ray photoelectron spectroscopy experiments

The chemical states of the coatings were confirmed by (XPS, Quantera II Inc.) using 2 keV Ar^+ sputtering technique. 1486 eV Al Kα was used as the X-ray source. The sputtering rate was calibrated based on SiO_2 and previous XPS, TEM experiments of Al_2O_3.

3. Results and discussion

The evolution of the downstream deuterium permeation flux through oxidized HTUPS 4 steel after upstream deuterium gas introduction with a pressure of 1.3 × 10^5 Pa is shown in Fig. 1. The relationship between steady state permeation flux and the square root of upstream deuterium gas pressure is shown in Fig. 2. The steady state permeation flux (J_∞) is related to the square root of driving gas pressure ($p^{1/2}$) through:

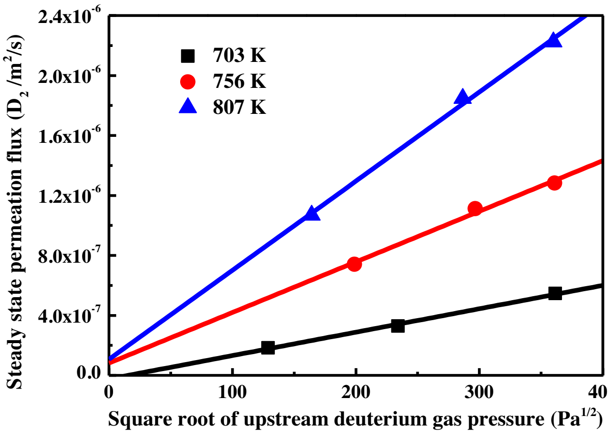

Fig. 2. Relationship between steady state permeation flux and the square root of upstream deuterium gas pressure.

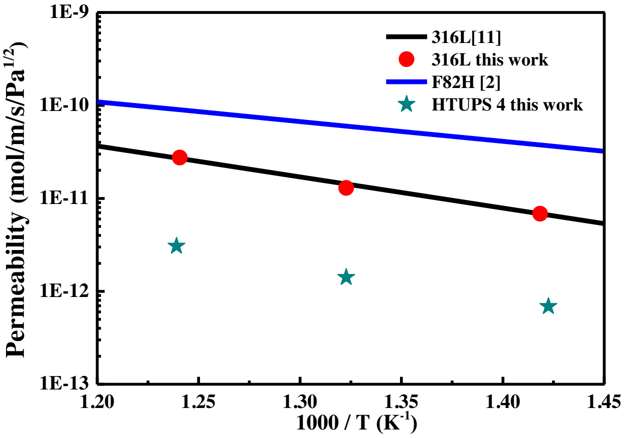

Fig. 3. Temperature dependence of permeability of deuterium in oxidized HTUPS 4 steel and bare 316L steel compared with the F82H [2] and 316L [11] steels.

$$J_\infty = \frac{P}{d} p^{1/2} \tag{1}$$

where P represents the permeability of deuterium in material; d is the thickness of the sample [9,10]. For the fact that all the lines do not go to zero, the slight change of background pressure in the GDP device owing to temperature change may be the reason.

Fig. 3 shows the temperature dependence of the permeability through the samples including oxidized HTUPS 4, bare 316L steel and F82H steel. The data of bare 316L obtained in our device and from literature [11] are shown, and no significant difference is found between these two, which means that the permeability measured in our device is reliable. RAFM steels like the F82H steel have been considered as the leading candidate structural materials for the DEMO fusion reactor and the first fusion power plant [12]. From Fig. 3, at the temperature ranging from 700 K to 800 K, the permeability of oxidized HTUPS 4 steel is 30–50 times less than that of the F82H steels and about 10 times less than that of 316L stainless steel. Data of bare HTUPS 4 steel are not given because oxide films tend to form on the surfaces of these bare aluminum-containing alloys even in low-oxygen-potential environment [13]. During the process of GDP experiments, oxide films could form and influence the results. Sun et al. [11] investigated the permeation behavior of different austenitic stainless steels, found that the

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