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Effect of degassing treatment on the deuterium permeability of Pd-Nb-Pd composite membranes during deuterium permeation

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ABSTRACT

Palladium coated niobium foil (Pd-Nb-Pd) is widely used as the hydrogen and deuterium-permeable materials for both the hydrogen purification and the hydrogen pumping. However, the performance of Pd-Nb-Pd membranes is usually affected by the degassing treatment (DT) that is typically implemented during the permeation test. In this paper, effects of DT on the deuterium permeability of the Pd-Nb-Pd composite membranes are investigated with varying degassing time. The deuterium permeability test is performed in the temperature range of 573–723 K at the driving pressures of 1–5 kPa. Results show that the permeation behavior of Pd-Nb-Pd membranes is affected by DT at 673 K during deuterium permeation. With the increasing DT time, the permeation performance deceases. The deuterium permeability of the sample heated at 723 K is 1.23×10^{-7} mol $m^{-1}{\cdot}s^{-1}{\cdot}Pa^{-0.5}$ at the 10 min DT time, one order of magnitude higher than 5.33×10^{-8} mol $m^{-1}{\cdot}s^{-1}{\cdot}Pa^{-0.5}$ of the sample at the 60 min DT time. The corresponding apparent activation energies are 20.11 kJ/mol and 52.55 kJ/mol for the samples at 10 min and 60 min DT time, respectively. Scanning electron microscope and Auger electron spectroscopy analysis reveal that a large number of pores are formed in the top Pd coating layer, and the inter-diffusion at the Pd-Nb interface occurs during the permeation experiment. Therefore, it is suggested that both the surface microstructure change and the inter-diffusion are attributed to the decline of deuterium permeability.

1. Introduction

As one of the most effective means for hydrogen separation and purification, membrane separation technology has advantages of low operating costs, minimum unit operations and low energy consumption [1,2]. Palladium-based (Pd-based) membranes with face-centered-cubic (fcc) lattice structure have the ability to dissociate and dissolve molecular hydrogen, and thus show excellent H_2 permeable properties. They are currently widely used for the separation and purification of hydrogen gas [3,4]. However, the high cost of Pd makes researchers constantly explore composite membranes or alternative effective membrane materials.

The group-V metals (e.g. V, Nb, Ta, etc.) with a more open body-centered - cubic (bcc) lattice are super-permeable to hydrogen. Thus, they are promising candidate materials for hydrogen permeable membranes to replace Pd-based membranes, which have been widely investigated for the purification of hydrogen and the hydrogen pumping in fusion devices [5–7]. For example, the group-V metals membranes can be used for pumping of deuterium/tritium fuel and the separation of deuterium/tritium from He ashes in ITER [8,9]. However, the group-V metals have the following limitations. First, they have little ability to catalyze the decomposition of hydrogen molecules. Second, the native oxide layer can prevent hydrogen transport through the membrane, also the oxide film can be further corroded by active gases. To overcome some of these problems, thin Pd film is usually deposited on both sides of group V metals, such as Pd-coated Nb (Pd-Nb-Pd) composite membranes. The permeability of deuterium in the Pd-Nb-Pd membrane is very important for its application in the fusion reactor fuel cycle system. Generally, the deuterium permeability is tightly related to the type of materials and the working temperature of materials as well as the operating conditions [10,11]. In recent years, it has been found that the hydrogen permeation behavior of the Pd-Nb-Pd composite membrane is greatly affected by the working temperature and pressure [12,13]. For example, Tucho et al. [14] observed an enhancement of hydrogen permeation in Pd/Ag membranes after thermal treatments in air at 300 °C and in N_2/Ar at 450 °C, which is related to the changes in

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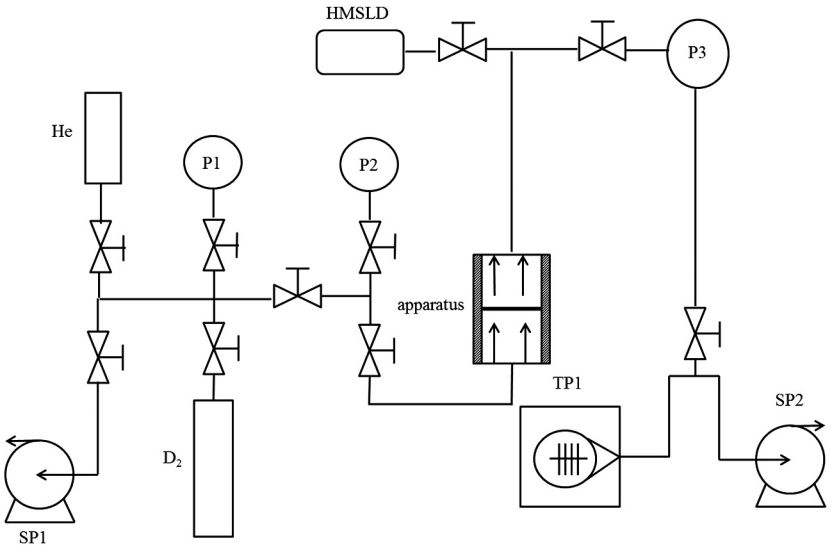

Fig. 1. Schematic of deuterium permeation apparatus. P1: pressure gauge, P2: pressure sensor, SP1: scroll pump, SP2: scroll pump, TP1: turbo molecular pump, P3: pressure gauge, HMSLD: helium mass spectrometer leak detector.

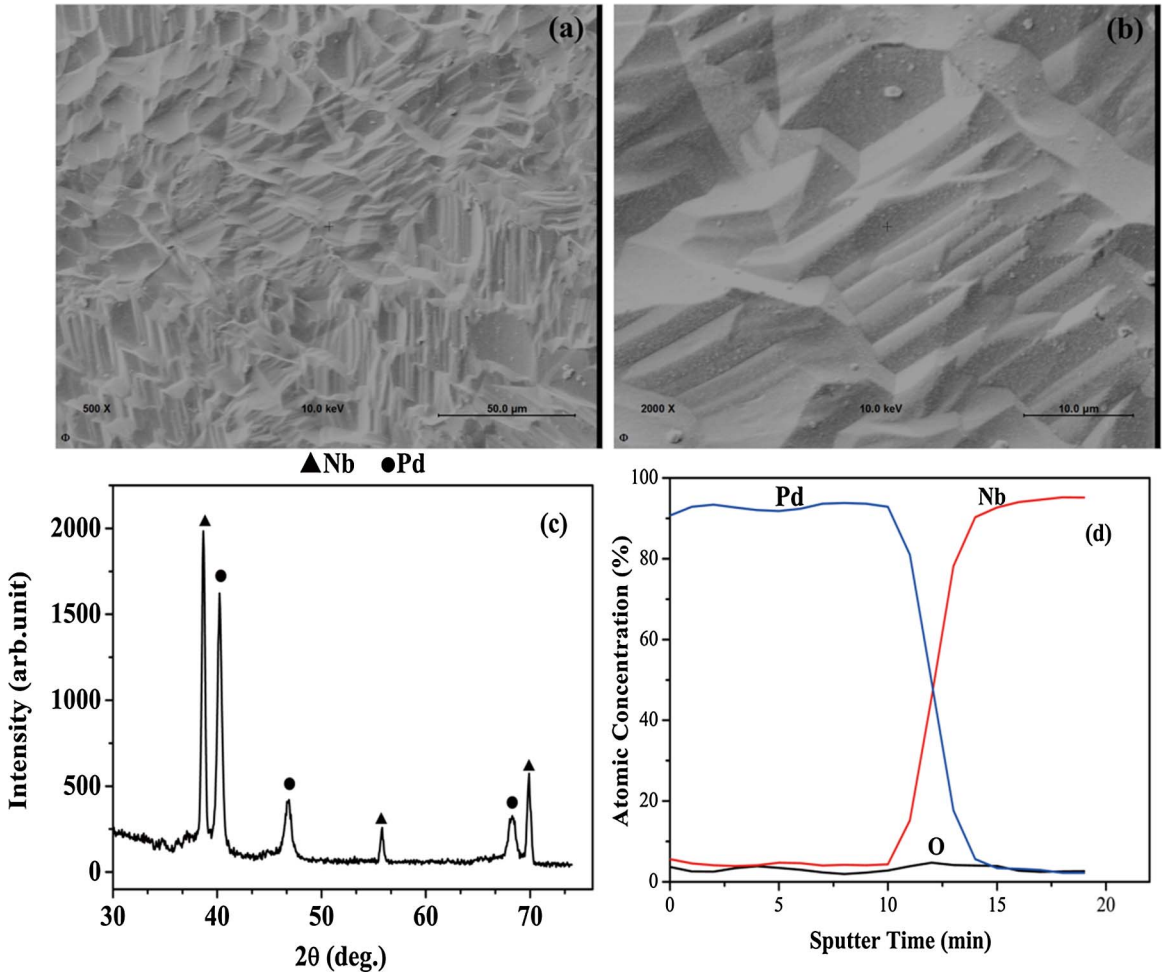

Fig. 2. SEM images (a, b) with different magnifications, XRD pattern (c) and AES depth profiles (d) of as -prepared Pd-Nb-Pd membranes.

hydrogen solubility. Hantano et al. [15] reported that the hydrogen adsorption rate for Pd/bare-Nb was sharply declined by heating at the temperature above 648 K. After introducing the Nb_2C intermediate layer, the degradation was substantially mitigated, which is ascribed to the retardation of porosity development of the Pd coating. It is suggested that the formation of Pd_3Nb due to the extended heating is responsible for the degradation of the hydrogen permeability. Zhang et al. [16] reported the significant effect of the hydrogen concentration on the hydrogen diffusivity. The hydrogen diffusivity in Nb drops sharply in the range of practical application pressures compared with that for dilute solutions. In addition, DT is usually adopted during the permeation test. In fact, the degassing process is a constant heat treatment process. Thus, the performance of Pd-Nb-Pd membranes is affected by the DT. Alimov et al. [17] examined the depth profile and surface

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