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Technical Communication

Microstructure and hydrogen permeability of $Nb_{40}Hf_{30}Ni_{30}$ ternary alloy

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ABSTRACT

The microstructure and hydrogen permeability of the $Nb_{40}Hf_{30}Ni_{30}$ ternary alloys were investigated in particular. The as-cast $Nb_{40}Hf_{30}Ni_{30}$ alloy consists of the bcc- (Nb,Hf) solid solution, the B_f-HfNi compound and the fine eutectic phase of {(Nb,Hf) + HfNi}. The annealed $Nb_{40}Hf_{30}Ni_{30}$ alloy consists of the bcc-(Nb, Hf) solid solution and the fine eutectic phase of {(Nb,Hf) + HfNi}. The as-cast $Nb_{40}Hf_{30}Ni_{30}$ alloy shows a high Φ value of 4.3×10^{-8} [mol H_2 m^{-1} s^{-1} $Pa^{-0.5}$] at 673 K, and for the annealed $Nb_{40}Hf_{30}Ni_{30}$ alloy, the Φ value is 3.8×10^{-8} [mol H_2 m^{-1} s^{-1} $Pa^{-0.5}$] at 673 K. Annealing can't enhance the hydrogen permeability of the $Nb_{40}Hf_{30}Ni_{30}$ ternary alloy.

1. Introduction

The Pd–Ag alloy membrances are mainly used to purify hydrogen gas, because of the high hydrogen permeability and the high resistance to the hydrogen embrittlement of Pd [1–4]. However, since palladium is too expensive and scarce in resources, not suitable for large-scale industrial applications as hydrogen permeation membranes for separation and purification of hydrogen gas, it is eagerly desired to develop low cost and high performance hydrogen permeation alloys other than Pd alloys [1–3,5]. The hydrogen permeability (Φ) is defined as the product of hydrogen solubility coefficient (K) and hydrogen diffusivity coefficient (D). Group 5A metals, such as V, Nb and Ta, which show large hydrogen solubility and high hydrogen diffusivity [6], are promising for hydrogen permeation membranes. Of these metals, more attention is paid to niobium because of the highest predicted hydrogen permeability. In recent years, niobium-based hydrogen permeation membranes for hydrogen purification have actively been investigated, such as the $Nb_{40}Ti_{30}Ni_{30}$ [2] and the $Nb_{40}Zr_{30}Ni_{30}$ alloys [6], i.e., non-palladium alloys, which have good results of the hydrogen permeability and high resistance to the hydrogen embrittlement. The $Nb_{40}Ti_{30}Ni_{30}$ alloy consists of the B2–TiNi intermetallic compound and the bcc-(Nb, Ti) solid solution, having the Φ value of 1.8×10^{-8} [mol H_2 m^{-1} s^{-1} $Pa^{-0.5}$] at 673 K [2]; while the $Nb_{40}Zr_{30}Ni_{30}$ alloy also consists of the B_f–type ZrNi intermetallic compound and the bcc-(Nb, Zr) solid solution, its Φ value being 4.64×10^{-8} [mol H_2 m^{-1} s^{-1} $Pa^{-0.5}$] at 673 K [6]. As we all know, Ti, Zr, and Hf are the same transition elements in the Periodic Table, which have the approximate chemical properties. In this letter, $Nb_{40}Hf_{30}Ni_{30}$ ternary alloy is studied in particular to analyze the microstructure and hydrogen permeability of this new hydrogen permeation alloy.

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2. Experimental

About 20 g ingots of $Nb_{40}Hf_{30}Ni_{30}$ alloys (mol%) were prepared by arc melting in a purified argon atmosphere using Nb (99.9% purity), Hf (99.99% purity) and Ni (99.9% purity) as raw materials. Disk samples of 12 mm in diameter and 0.5–0.7 mm in thickness were cut from the ingots by the spark erosion method. The surface of the disks was polished using buff and alumina particle (0.5 μm). The as-cast samples were annealed at 1100 °C for 72 h in the argon atmosphere. Microstructural observation and measurement of chemical composition of the samples were carried out with a scanning electron microscope (SEM) and an energy dispersion X-ray spectroscopy (EDS). Determination of the volume percentage of a phase was carried out on a Macintosh (model) computer using the public domain NIH Image program from SEM data. Structure of the sample was identified by an X-ray diffractometer (XRD) using Cu K-radiation monochromated by graphite. Both sides of the disks were coated with pure Pd in the thickness of 190 nm by the magnetron sputtering machine. The disk sample sealed with copper gaskets was set into the hydrogen permeation measuring apparatus, that is, a conventional gas-permeation technique, and then hydrogen permeability Φ of these alloys were measured in the temperature range of 523–673 K, at the hydrogen pressure up to 0.5 MPa. The experimental procedure has been described in detail in the previous papers [7–9].

3. Results and discussion

Fig. 1 and Fig. 2 show the XRD patterns of the as-cast and the annealed $Nb_{40}Hf_{30}Ni_{30}$ alloy, respectively.

As shown in Fig. 1, the $Nb_{40}Hf_{30}Ni_{30}$ alloy consists of the bcc-(Nb, Hf) solid solution and the B_f-HfNi compound [10], including some new compounds of Hf_xNi_y ($1 \le x \le 8$; $3 \le y \le 21$): $HfNi_3$, Hf_2Ni_7, and Hf_8Ni_{21}, as compared with JCPDS (Joint Committee on Powder Diffraction Standards) files of No.32-0475, No.26-1129 and No.32-0476 (International Center for Diffraction Data, 2002). As shown in Fig. 2, after annealing, the diffraction peaks sharpen and the main phases also include the bcc-(Nb, Hf) solid solution and the B_f-HfNi compound with more quantity of new phases of Hf_xNi_y ($1 \le x \le 8$; $3 \le y \le 21$): Hf_8Ni_{21} (No.32-0476 and No.32-0425), Hf_3Ni_7 (No.71-0477), Hf_2Ni_7 (No.26-1129), and $HfNi_3$ (No.32-0475). The intensity of the (021) and (040) planes of the HfNi phase in the annealed alloy decreases sharply than that of the as-cast alloy, i.e., the quantity of the HfNi phase decreases after annealing.

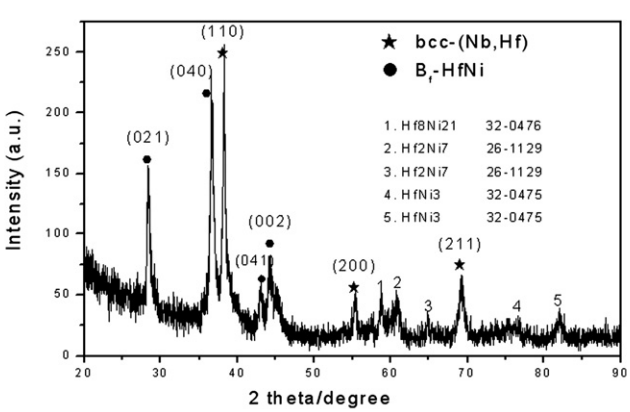

Fig. 1 – The XRD patterns of the as-cast $Nb_{40}Hf_{30}Ni_{30}$ alloy.

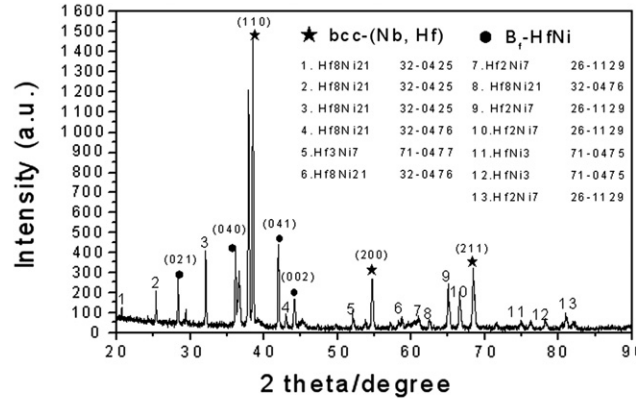

Fig. 2 – The XRD patterns of the annealed $Nb_{40}Hf_{30}Ni_{30}$ alloy.

Fig. 3 shows the SEM photographs of the $Nb_{40}Hf_{30}Ni_{30}$ alloy in the as-cast and the annealed state.

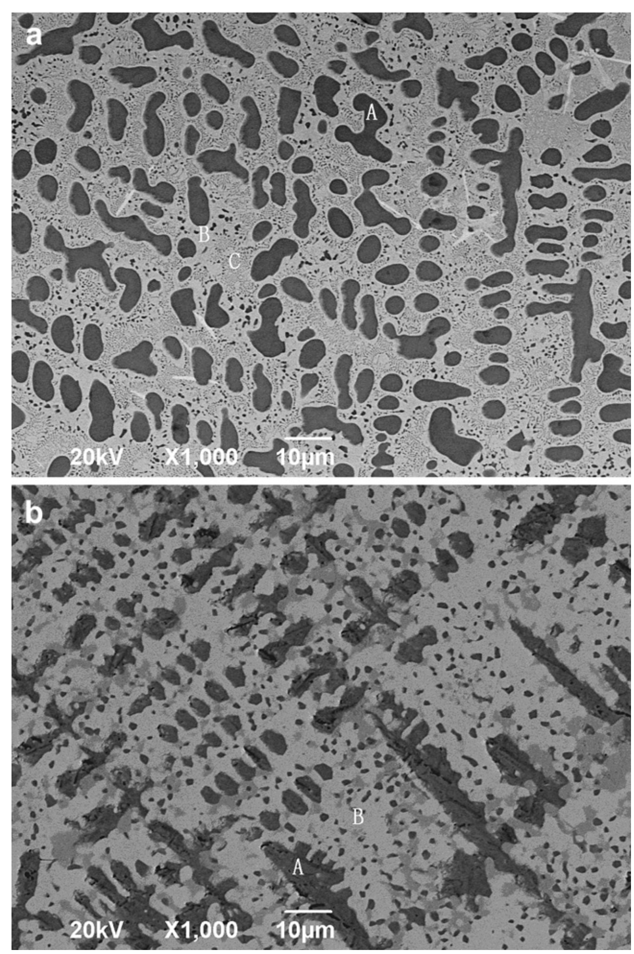

Fig. 3 – The SEM images of the $Nb_{40}Hf_{30}Ni_{30}$ alloys in the as-cast state (a) and the annealed state (b).