ELSEVIER

Journal of Alloys and Compounds 253–254 (1997) 370–373

Journal of ALLOYS AND COMPOUNDS

Hydrogen permeability and diffusivity in amorphous Metglas 2826MB under AC excitation

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Accepted 18 August 1996

Abstract

In this work the effect of magnetic field on hydrogen permeability and diffusivity in metglas 2826MB has been determined. The hydrogen permeability and hydrogen diffusivity were measured by the bi-electrode method. A magnetic field of fixed strength 20 Oe was applied parallel to the transverse direction of the ribbon, and both DC and AC excitation from 60 Hz to 400 Hz were employed. It was found that the magnetostriction constant increased linearly with the applied AC frequency, until a saturation level was reached at around 300 Hz. As the frequency increased, hydrogen permeability and diffusivity in the metglas increased accordingly. The effect can be attributed to the change in magnetoelastic strain induced by the applied excitation field. Microcracks were found on the specimens' surface after prolonged charging or charging with higher current density, showing that they were susceptible to hydrogen embrittlement. Hydrogen, however, did not affect the amorphous nature of the metglas.

Keywords: Amorphous Metglas 2826MB; Hydrogen permeability; Hydrogen diffusivity; AC excitation; Magnetostriction

1. Introduction

The nickel–iron-based amorphous alloys (e.g., Metglas 2826 MB) have a higher permeability, lower coercive field and lower magnetostriction than their iron-based counterpart. The nickel–iron-based alloys also have a better corrosion resistance, and can be used satisfactorily in field sensors, recording heads or shielding applications. In some cases, it is possible for an amorphous metal to be exposed to a hydrogen atmosphere either during the manufacturing process or in service, which renders them susceptible to hydrogen embrittlement. The research on the hydrogen embrittlement of amorphous alloys has been very active [1–6]. For examples, Nagumo and Takahaski [5] determined the hydrogen diffusivity of some Fe-based amorphous alloys, and found that the diffusivity ranged from $5{\cdot}10^{-11}$ to 10^{-10} cm^2/sec at 300 K. Lin and Perng [6] have found that the mechanical properties of Metaglas 2826MB was reversible after hydrogen charging, provided that the hydrogen can be eliminated before crack initiation. There is, however, little work relating the AC excitation field and the hydrogen environment, which are found to exist together. The objective of the present work was to reveal the effect of magnetic excitation on the hydrogen diffusivity and permeability in the metglas 2826 MB.

2. Experimental

Metallic glass ribbons of metglas 2826 MB($Fe_{40}Ni_{38}B_{18}Mo_4$) were prepared by single roller melt spinning. Melt spun ribbons of 50 mm wide were fully annealed in dry nitrogen at 355 °C for 2 h so as to relief any quench-in stress. Rectangular strips (50 mm × 25 mm) were cut from the central portion of the ribbons. The thickness of each specimen was determined by averaging two measured values near the centre, each about 10 mm apart. After ultrasonic cleaning in methanol, the matte side of the ribbon was coated with palladium by sputtering. The Pd layer was approximately 50 nm thick.

Standard permeation cell was set up following the design of Devanathan and Stachurski [7] (Fig. 1). The electrochemical cell was divided into two compartments: the hydrogen entry side (cathodic side) and the hydrogen exit side (anodic side). The solutions in these two compartments were 0.05 M H_2SO_4 and 0.1 M NaOH, respectively. The specimen was clamped between these two compartments, with the lustrous side of the strip, i.e., the side

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0925-8388/97/$17.00
PII S0925-8388(96)02906-4

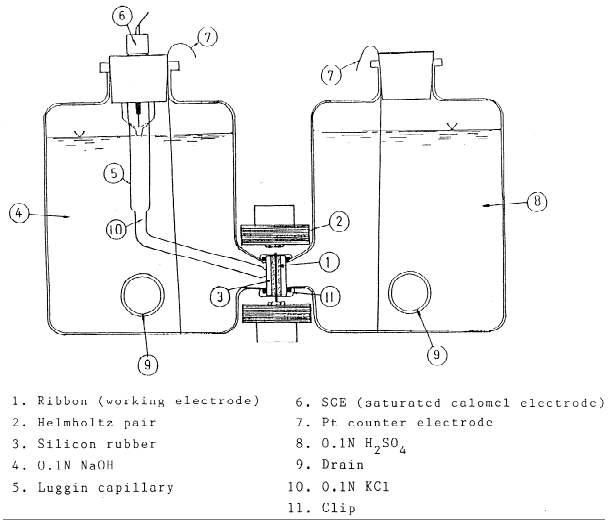

Fig. 1. Permeation cell and Helmholtz pair construction.

without Pd coating, taken as the hydrogen entry side. The thickness of the samples was in the range from 0.02 to 0.025 mm, and the exposed area to the solutions was 0.5 cm^2. On the anodic compartment, a saturated calomel electrode (SCE) was used as a reference electrode, and was connected to the compartment through a Luggin capillary tube. For both compartments Pt counter electrodes were used. The hydrogen charging current density used in the cathodic side was set at 50 mA/cm^2. However, higher values of cathodic current density (i.e., 100 mA/cm^2) have also been applied to show the degree of damage to the specimen by hydrogen. The specimen was maintained potentiostatically at 100 mV vs. S.C.E. The anodic current, which reflected the flux of hydrogen through the specimen, was recorded during the experiment. The effective hydrogen diffusivity, D_{eff}, was calculated from each permeation transient curve using the well-known time-lag method:

$D_{eff} = L^2 / 6\, t_L$ where L is the length of the specimen, and t_L is the lag time, which is defined as 0.63 times the time required to reach a steady state flux.

2.1. External applied field and magnetostriction constant

As can be seen in Fig. 1, a Helmholtz pair was used for generating the magnetic field during the hydrogen permeation experiment. A magnetic field of fixed strength 20 Oe was applied parallel to the transverse direction of the ribbon, both DC and AC excitation varied from 60 Hz to 400 Hz were used. The reason for choosing 20 Oe was that this value was very close to the saturation magnetization Hs (25 Oe), and the magnetostriction constant λ was less sensitive to the field strength within this range. Consequently, the uniformity requirement of the field was met easily. It was also considered that there was no induced current parallel to the permeation path, otherwise a complicated situation would resulted. Strain gauge extensometry was used to determine the magnetostriction constant under different excitation fields.

3. Results and discussions

The metglas 2826 MB after annealing at 355 °C has a saturation magnetostrictive constant $\lambda_s = 12 \cdot 10^{-6}$, which was low as compared with those of other Fe-based and Fe-Ni based alloys ($\sim 20–35 \times 10^{-6}$). It is thus consider favorable in core application. For 2826 MB amorphous ribbon, the magnetostrictive constant λ is known to be frequency and magnetic field strength dependent. In this study, the field strength has been kept constant at 20 Oe. The frequency response of the magnetostriction constant parallel to the field direction is shown in Fig. 2. It is clear that a linear relationship exists between λ and frequency up to 400 Hz. After 400 Hz, the magnetostriction constant became independent of the frequency. The details of these dynamics are quite complicated and are beyond the scope of this study [8].

In the hydrogen permeation test the use of a hydrogen charging current of 50 mA/cm^2 as the hydrogen charging density was simply found by trial and error. Higher values of charging current density accelerated the hydrogen permeation rate, but also led to a higher risk of cracking and the presence of pin holes. Special care has been taken

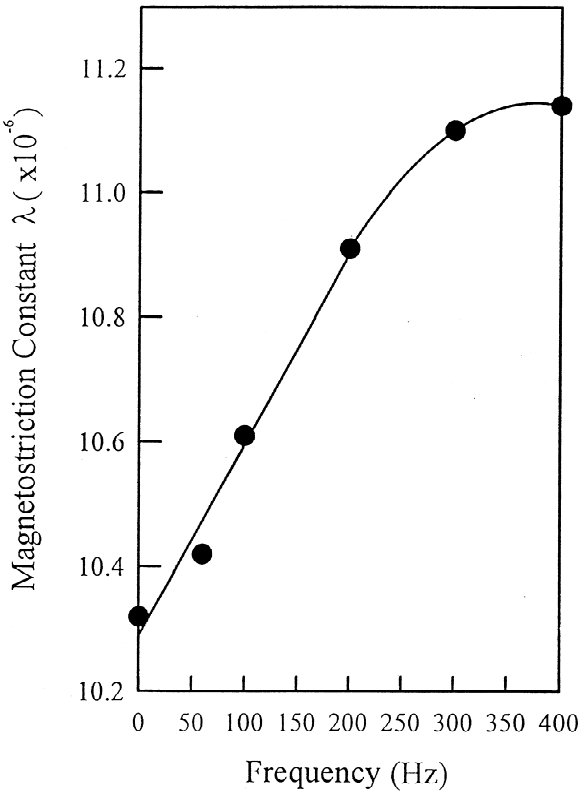

Fig. 2. Frequency response of the magnetostriction constant of metglas 2826MB.

to ensure that the specimen was free from cracking and pinholes during the measurement of diffusivity and permeability, otherwise the defects might considerably shorten the diffusion and permeation paths in an uncontrolled way. The effect of external excitation on the hydrogen permeability can be clearly seen in Fig. 3. As the frequency increased, hydrogen permeation was increased from 1.9×10^{-12} mole H/cm^{2}s in the case of DC to 3.7×10^{-12} mole H/cm^{2}s when the applied AC frequency was 400 Hz. Thus under an excitation field, the hydrogen permeation was assisted by the magnetoelastic strain or stress so induced. As the frequency was increased, so was the lattice distortion anisotropy energy, consequently increasing the hydrogen permeability. The effect of the excitation field can also be found on the hydrogen diffusivity as shown in Fig. 3. Similar to the hydrogen permeability, the diffusivity tended to increase with the frequency of the excitation field. The change in the hydrogen diffusivity, however, was not as significant as that for the permeation. Nevertheless, the diffusivity also changed from 1.90×10^{-11} to 2.73×10^{-11} cm^2/s as the frequency increased from 60 Hz to 400 Hz.

After the permeation tests, the specimens were examined under the scanning electron microscope. All specimens suffered certain degrees of corrosion, as shown on the hydrogen exit side in Fig. 4(a). With an increase in the charging current density, microcracks as well as pinholes were also found (Fig. 4(b)). No sign of induced crystallization can be observed through SEM examination nor from the X-ray diffraction data, as shown in Fig. 5, in which a typical broadened peak of amorphous material was found.

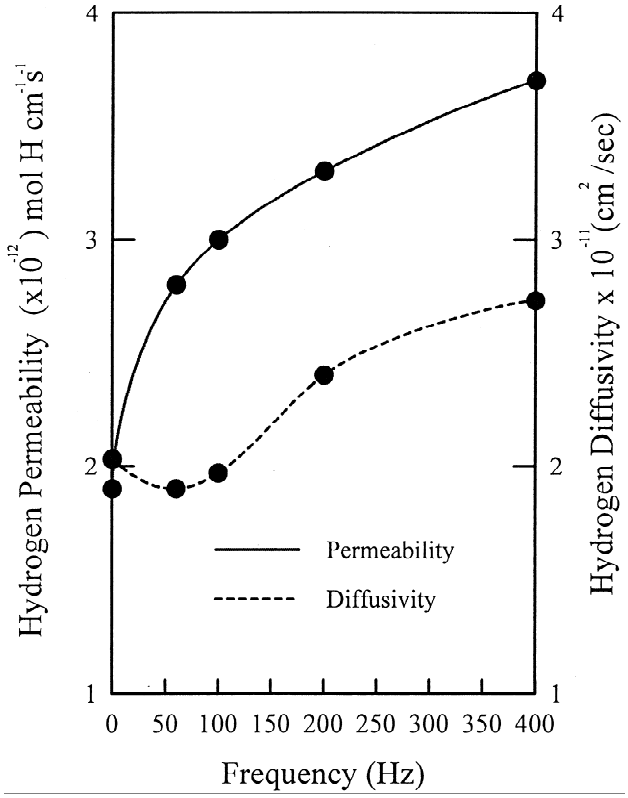

Fig. 3. Effect of excitation frequency on hydrogen permeation and diffusivity in metglas 2826MB.

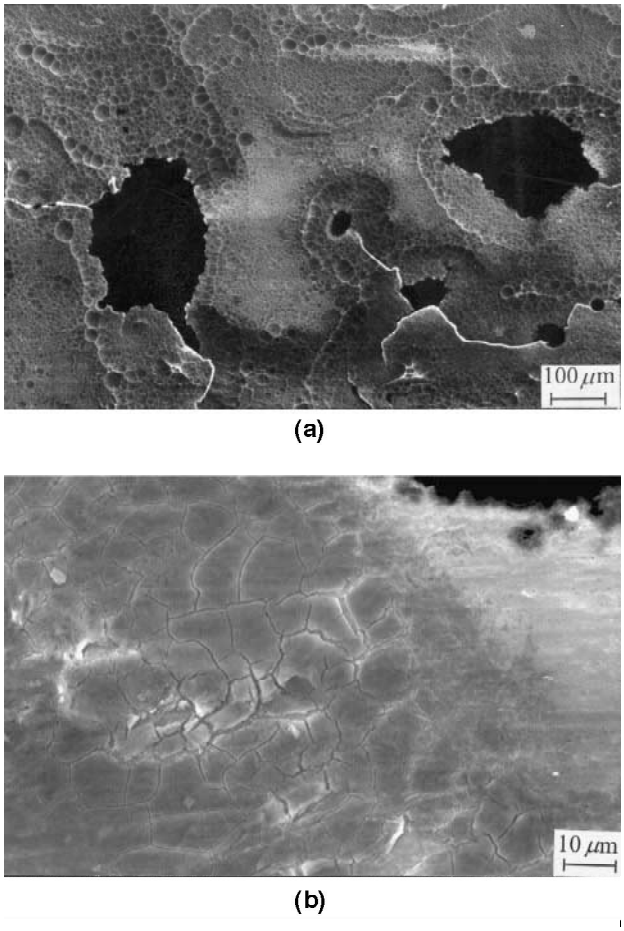

Fig. 4. Scanning electron micrographs of the specimens after permeation test showing (a) pinholes and severe corrosion pits on the exit surface, and (b) microcracks on the charging surface.

4. Conclusions

The effects of a magnetic field on hydrogen permeability and diffusivity in metglas 2826MB have been studied. For a given field strength, the magnetostriction constant in-

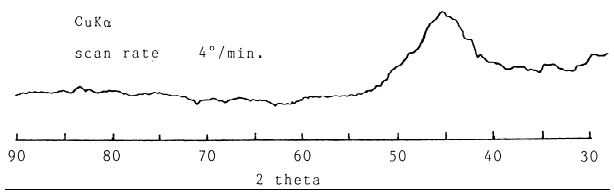

Fig. 5. X-ray diffraction pattern of Metglas 2826MB after permeation test.

creased linearly with applied AC excitation frequency until a saturation level was reached at around 300 Hz. The permeability and the diffusivity of hydrogen through the amorphous ribbon were also a function of applied frequency. As the frequency increased, hydrogen permeability was increased accordingly from 1.9×10^{-12} mole H/cm^2s to 3.7×10^{-12} mole H/cm^2s, and the hydrogen diffusivity was raised from 1.8×10^{-11} to 2.5×10^{-11} cm^2/s. These can be attributed to the change in magnetoelastic strain induced by the applied excitation field. After prolonged charging or with higher values of cathodic charging current densities applied, almost all the specimens suffered some degree of hydrogen damage. The structure of the amorphous metglas, however, remained largely unchanged.

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