



Susceptibility of minor alloying to corrosion behavior in yttrium-containing bulk amorphous steel

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ABSTRACT

Minor alloying of yttrium to bulk amorphous steels can influence their corrosion resistance significantly. The corrosion behavior was investigated by using potentiodynamic polarization measurements, Mott–Schottky analysis and X-ray photoelectron spectroscopy (XPS). It showed that, in comparison with Y-free alloy, the stability of the passive film for 0.5 at.% yttrium alloy decreased sharply, whereas enhanced with more additions of yttrium. It is noticed that the corrosion resistance was correlated to semiconductor properties of the passive film. This is understood on the basis of the yttrium-doping-induced structural defect effects in the passive films. A probable way to further enhance the corrosion resistance of amorphous alloys is suggested.

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1. Introduction

Since amorphous Fe–Cr–metalloid alloys were reported to have extraordinary high corrosion resistance firstly in 1974 [1], extensive studies over several decades have been performed on searching for highly corrosion-resistant amorphous alloys [2–6], with the presence of a certain amount of corrosion-resistant alloying elements. These alloys exhibit spontaneous passivation and hardly suffer pitting in aggressive environments, introducing them as candidates for conventional corrosion-resistant alloys. The nature of high corrosion resistance in these alloys is often directly related to the existence of uniform and protective passive films that enriched corrosion-resistant elements [3,7]. Fundamental understanding of the roles of individual corrosion-resistant elements [3,7] and their synergistic effects [8] on the formation and evolution of such passive films is beneficial for materials' design guidance.

The stability of passive film, however, is not solely attributed to the presence of corrosion-resistant elements. Passive films exhibit *per se* semiconductor properties and the electronic properties are intimately related to the universal nature of corrosion resistance of alloys [9–11]. Structural defect in the oxide film is a crucial factor in influencing its electronic properties [12,13], suggesting us that the stability of passive film could be improved by tailoring its structural defect. Recently some studies have proved that cation doping can

greatly influence the electronic properties of the oxide through changing its structural defect, such as in enhancing conductivity of solid electrolyte ceramics [13,14] and in controlling the high temperature oxidation [15]. Clearly, it is an interesting idea to elucidate the relationship between cation doping and the stability of passive films, especially in amorphous alloys.

In the study reported herein, we tried to investigate the intrinsic nature of highly corrosion-resistant amorphous alloys by changing the passive film structure via compositionally tuning the base alloy. The materials used in this work are based on the amorphous steel composition $\text{Fe}_{50}\text{Cr}_{15}\text{Mo}_{14}\text{C}_{15}\text{B}_6$, which contains sufficient amounts of corrosion-resistant elements and exhibit superior corrosion resistance. It is worthy noting that minor addition of yttrium to amorphous steel can dramatically improve the glass-forming ability [16,17] as well as effectively influence the nature of the brittle-to-plastic transition [18], but the exact roles during corrosion are less certain [19,20]. In view of thorough insight into the roles of minor yttrium alloying in passivity, we present cation (yttrium) doping controlled stabilization of passive films in corrosion of bulk amorphous steels, attempting to address a more attractive way to further enhance the corrosion resistance of alloys.

2. Experimental

The $\text{Fe}_{50-x}\text{Cr}_{15}\text{Mo}_{14}\text{C}_{15}\text{B}_6\text{Y}_x$ alloy ingots with $x = 0, 0.5, 1.0, 1.5$ and 2.0 (at.%) were prepared by melting appropriate amounts of Fe (99.9%), Cr (>98.5%), Mo (99%), Y (99.9%), C (99.9%) and B (99%) in

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an arc furnace under an argon atmosphere. Cylindrical samples with diameter of 1 mm (yttrium-free rods) or 2 mm (the others) were prepared by suction casting of the molten alloys into a copper mold. The structure of the as-quenched samples was characterized by X-ray diffraction (XRD) with Cu K α and the microstructures of the samples were examined with an FEI Tecnai F30 transmission electron microscope (TEM). The thin foil specimens were prepared by a standard twin-jet electropolishing method. Electrochemical measurements were conducted in a three-electrode cell using a platinum counter electrode and a saturated calomel reference electrode on a Potentiostat/Galvanostat (EG&G Princeton Applied Research Model 273/2273). The samples used in electrochemical measurements were finally polished with 1500 grinds sand papers. Potentiodynamic polarization curves were measured with a potential sweep rate of 0.33 mV/s in 1 mol/L HCl aqueous solution. After the samples were immersed in 1 mol/L HCl solution for 100 h, Mott–Schottky analysis of the passive films was performed at a selected frequency of 1 kHz to measure semiconductor properties of the passive film. The open-circuit potential of each passive film was recorded before Mott–Schottky measurement. For surface analysis, X-ray photoelectron spectroscopy (XPS) was employed by using an ESCALAB250 photoelectron spectrometer with Al K α excitation ($h\nu = 1486.6$ eV) to determine the depth profiles of passive film by an in situ argon sputtering gun.

3. Results and discussion

The as-cast alloy rods have been characterized as a glassy structure by XRD analysis. The microstructure of the as-prepared specimens has been carefully examined with TEM. A typical high-resolution TEM (HRTEM) image of the as-quenched 0.5 at.% yttrium alloy is shown in Fig. 1. Except for the short-range ordered clusters, no crystalline phase has been observed. The inset of Fig. 1 is the corresponding selected area electron diffraction (SAED) pattern, in which no sharp diffraction ring or spots but diffraction halos have been determined. This indicates that the as-prepared 0.5 at.% yttrium alloy possesses a fully glassy structure. Such a characteristic has also been confirmed in other yttrium-free and yttrium-containing samples (not shown).

Typical potentiodynamic polarization curves measured in 1 mol/L HCl solution indicate that the Fe_{50-x}Cr₁₅Mo₁₄B₆C₁₅Y_x

amorphous steels exhibit high corrosion resistance, as shown in Fig. 2. All these alloys are spontaneously passivated with low passive current density. Wide passive regions are recognized until the transpassive dissolution of chromium occurs beyond about 0.8 V [4,21]. It should be noted that the passive films for 0.5 and 1.0 at.% Y-containing alloys are unstable as the polarized potential is beyond 0.5 V, judged from the fact that the passive current density increases rapidly. In contrast, there exhibits only slight increase in the passive current densities for 1.5 and 2.0 at.% Y samples when polarized to more anodic potentials. Further, the passive current densities by averaging current densities in the passive region change as a function of yttrium content in the alloys, as displayed in the inset of Fig. 2. Clearly, the stability of passive films is sensitive to the presence of minor yttrium in the alloys, illuminating us to further understand the intrinsic reason for the stabilization of passive films.

Mott–Schottky analysis was used to reveal the semiconductor properties of the passive films. As depicted in Fig. 3(a), a linear region from about 0.45 to 0.75 V appears in each Mott–Schottky plot and the yttrium content in the alloys has an obvious effect on the semiconductor properties of passive films. Further, one can deduce the donor density, N_D , from the slope of linearly fitted Mott–Schottky plots using a dielectric constant $\epsilon_r = 15.6$ [21,22]. Fig. 3(b) indicates the variation of N_D values calculated as a function of yttrium. All of the N_D values are in the order of 10^{19} cm⁻³, which is much lower than those of stainless steels ($10^{20} \sim 10^{22}$ cm⁻³) [22,23]. This implies that less defective and higher protective passive films are formed on amorphous alloys. In Fig. 3(b), N_D sharply rises to 4.86×10^{19} cm⁻³ when yttrium content in the alloy is 0.5 at.%, over seven times than that in yttrium-free alloy (6.5×10^{18} cm⁻³). And then it reduces gradually to lower levels with yttrium content beyond 1.5 at.%. Meanwhile, the open-circuit potentials of the passive films for Mott–Schottky measurements were recorded as a valid method to reflect the stability of the passive films and the results were consistent with passivation behavior shown in Fig. 2. Apparently, the open-circuit potential of the passive film for 0.5 at.% yttrium alloy is less noble than any other yttrium-containing or yttrium-free alloys. Combining the polarization measurements with Mott–Schottky analysis, we can conclude that the minor addition of yttrium to Fe–Cr–Mo–B–C amorphous steel influences dramatically the electronic properties

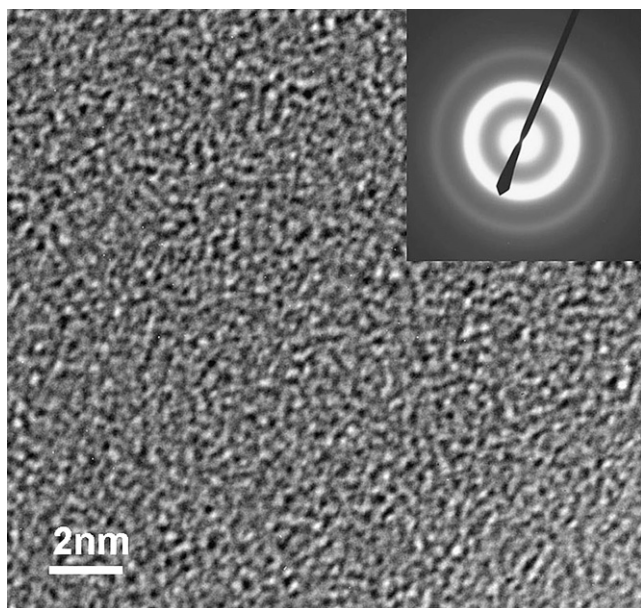


Fig. 1. Typical HRTEM image of the as-prepared 0.5 at.% yttrium amorphous alloy. The inset is the related SAED pattern.

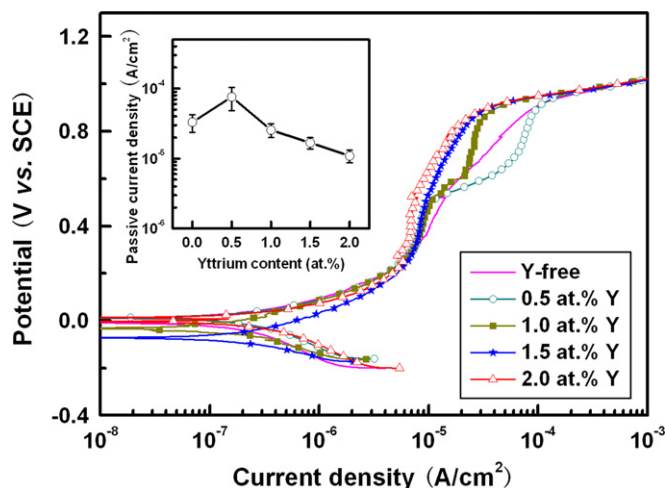


Fig. 2. Typical potentiodynamic polarization curves of amorphous steels with 0, 0.5, 1.0, 1.5 and 2.0 at.% yttrium in 1 mol/L HCl solution. The inset shows the averaged passive current density changes as a function of yttrium content, in which the averaged current density is obtained by averaging the current densities at passive region (herein, defined from 0.4 to 0.8 V (SCE)). Each polarization curve of the same yttrium-containing alloy was repeated five times.

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