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Plasticity enhancement in bulk metallic glasses by electroless plating with Ni-P amorphous films



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ABSTRACT

In this study Electroless plating is used to deposit amorphous Ni-P films on the surface of the $Zr_{41.2}$ Ti_{13.8}Cu_{12.5}Ni₁₀Be_{22.5} (Vit.1) bulk metallic glasses (BMGs) with different pH values and film thicknesses. The special amorphous/amorphous structure exhibited large plasticity. The plastic strain is 10.6% for the coated pillars with a coating thickness of 86.5 μ m, which is 5.9 times of 1.8% of the bare Vit.1 pillars. The amorphous Ni-P films provide high radial confinement stress, which leads to more homogeneous formation of a high density of smaller shear bands at the Ni-P/Vit.1 interface. The technique provides an effective way to enhance the plasticity without obvious sacrifice of the yielding strength for BMGs at room temperature.

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1. Introduction

Bulk metallic glasses (BMGs) as a new class of materials possess many unique properties, including exceptionally high yielding strength, hardness, elastic limit, etc. [1–3]. In general, the deformation of BMGs involves highly localized shearing in narrow bands, which causes geometrically unconstrained BMGs with a failure catastrophically along dominant shear bands. Thus, their application is severely limited by the catastrophic fracture upon loading at room temperature.

Many methods have been developed to improve the ductility of BMGs in order to inhibit the occurrence of the catastrophic failure at room temperature [4–12]. These approaches seek to induce a more homogeneous distribution of shear bands. Qiao et al. [4] suggested that, in in-situ metallic glass matrix composites (MGMCs), the amorphous matrix provided extremely-high strength, and the dendrites possessed large plasticity, which led to distinctly plastic deformation. It is reported that the designed Zr-based BMGs, which had large Poisson's ratio, ν , and could experience superplastic deformation at room temperature [6]. And the exceptional deformability was ascribed to the homogeneous and concurrent nucleation and evolution of high-density shear bands throughout the whole materials [6]. Yang et al. [11] introduced highly

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viscous layers in sandwiched structures of adhesive/Al/BMG/Al/adhesive and achieved high degree of plasticity in brittle Zr-based BMGs. It has been demonstrated that the compressive surface stresses created by shot-peening were able to improve the plasticity of monolithic BMGs [12]. Sarac et al. [8] have reported the arrestment of shear bands via pores, with the improvement of tensile ductility in Zr₃₅Ti₃₀Cu_{7.5}Be_{27.5} metallic glass.

The surface modification is a successful approach for which the internal amorphous structure remains unchanged and the plasticity improved apparently [10,13-21]. Chen et al. [14] adopted electrodepositing method to deposit a copper coating onto Zr₅₇Al₁₀Ni₈Cu₂₀Ti₅ BMGs, and the plasticity of BMGs has an increase from ~2 to ~6.5%. Meng et al. [16] designed Zr_{52.5}Cu_{17.9}Ni_{14.6}Al₁₀Ti₅ (Vit.105) BMGs coated with Ni films, and the corresponding plasticity can reach to 9%. However, the enhancement in plasticity through electrodeposition of crystal films would lead to the decline of the yielding strength of the composites [13–17]. It is badly needed to find a new way to improve the plasticity of the BMGs without sacrifice of the yielding strength. The electroless plating of the Ni-P alloys, which has the advantage of the low cost, good-quality deposits with uniformity, and good bond adhesion, has been widely used [20-23]. In the past few years, the Ni-P amorphous alloys were usually deposited on the aluminum alloys, carbon steels, and magnesium alloys to enhance the resistant to corrosion of the substrate alloys [24-27]. In this study, the electroless plating is adopted to deposit amorphous Ni-P alloys on Vit.1 (Zr_{41.2}Ti_{13.8}Cu_{12.5}Ni₁₀Be_{22.5} BMGs) pillars to investigate the influence of the coating on the mechanical properties of BMGs.

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2. Experiment

2.1. Materials

The composition of BMG pillars was $Zr_{41.2}Ti_{13.8}Cu_{12.5}Ni_{10}Be_{22.5}$ (Vit.1), which were prepared by arc-melting the mixture of Zr, Ti, Cu, Ni, and Be of purities greater than 99.9% (wt.%) in a Ti-gettered high-purity-argon atmosphere. The chemical plating was consisted of 25 g/L NiSO₄·6H₂O, 25 g/L NaH₂PO₂, 20 g/L C₆H₈O₇·H₂O, 10 g/L NH₄HF₂, 4.5 g/L NaOH, and 1 mg/L CN₂H₄S. All these chemical materials are analytical reagent.

2.2. Methods

The BMGs pillars were produced by suction-casting into a copper mold of 2 mm in diameter. Prior to the electroless plating, the surface of the as-cast samples was polished, using #1200 emery papers. Both the bare Vit.1 pillars and plated pillars were cut into small pillars of 4 mm in height. Thus the aspect ratio of the sample is nearly 2:1. During plating, the bath temperature was maintained at 85 °C using a constant temperature water bath. The pH values of the reaction solutions are adjusted by the NH₃·H₂O. The "L" used in figures is noted as the thickness of the amorphous films.

All samples were tested under compression at room temperature at a strain rate of $5 \times 10^{-4} \, \text{s}^{-1}$. The coating thickness, lateral surfaces, and fracture morphologies of the coated BMG pillars were investigated by scanning electron microscopy (SEM), and the contents of Ni and P were detected by energy dispersive spectrum (EDS). The structure of Ni-P coating was verified by X-ray diffraction (XRD).

3. Results and discussions

During the deposition, there are some reactions in the solution to maintain the procedure of deposition, and it is a heterogeneous autocatalytic process. There are many theories about the reactions during the deposition, and the hydrogen free radical theory is universally admitted [26-28]. The experimental setup is schematically shown in Fig. 1, and the main reactive ions in the resolution are briefly indicated. The Ni from the alloys and the replacement reaction can act as the catalysis for Eq. (1). Under conditions of catalysis by heating, $H_2PO_2^-$ ions are reacted with H_2O to produce nascent hydrogen free radical, firstly. Next, the hydrogen atoms are unstable, and Ni²⁺ ions in the bath were reduced to Ni grains by these hydrogen atoms as shown in Eq. (2). As shown in Eq. (3), $H_2PO_2^-$ ions are also reacted with hydrogen free radical to produce P elementary substance. The Ni acts as the catalyst of the oxidation reaction of hypophosphite for the autocatalytic reactions of the electrochemical Ni-P plating. The coprecipitation of Ni and P is produced by Eqs. (2) and (3). The reactions that make sustainable Ni-P film deposition are as follows [22]:

$$H_2PO_2^- + H_2O \xrightarrow{Ni} HPO_3^{2-} + 2H \cdot + H^+$$
(1)

$$Ni^{2+} + 2H \cdot \rightarrow Ni + 2H^+ \tag{2}$$

$$H_2PO_2^- + H \cdot \rightarrow H_2O + OH^- + P \tag{3}$$

The surface of deposited film is shown as the inset in Fig. 2(a), and it is the typical "cauliflower-like" structure as reported previously [20–25]. The above reactive process could be used to interpret the coarse surface of Ni-P alloy by electroless plating. Firstly, Ni²⁺ ions in the bath were reduced to Ni grains and deposited on the surface of the substrate. Next, the Ni grains acted as the deposition reaction and the coprecipitation of Ni-P deposited on the surface and covered the Ni grains. Thus the obtained Ni-P presents the coarse characteristic on the surface. The pH values of the solution are 5.7, 5.9, 6.1, 6.3, and 6.5, respectively. The thickness of the coatings with different pH is controlled with the same value of about 50 µm. Then, the deposited films are controlled by the same pH of 6.5 and the different coating time of 2, 4, 6, and 8 h. The thickness of the coating tested by SEM is 27.7, 35.8, 57.5, and 86.5 µm, respectively.

The deposited Ni-P films with different pH are peeled from the substrate and tested by the X-ray diffraction (XRD). The XRD spectrums of Ni-P films are presented in Fig. 2, and a broad diffraction peak indicates that the Ni-P film is mainly amorphous, without detectable peaks corresponding to crystalline phases [17,20,21], suggesting an amorphous structure of the Ni-P coatings. The present results are consistent with the P content of the Ni-P coatings, and the formation of intermetallic compounds, such as Ni₃P, Ni₅P₂, Ni₁₂P₅, and Ni-rich microcrystalline phases is severely hindered [24]. In Fig. 2, it can be seen that the characteristic peak intensity became larger with the increasing pH values. From this point, it is obtained that the glass-forming ability of the deposited films is affected by the pH of the solution. In Fig. 2(b), the P contents are decreased with the pH of the plating solution. The coated BMGs with different pH (5.7, 5.9, 6.1, 6.3 and 6.5) were subjected to compression to investigate the influence of the pH on the plasticity of BMGs. Fig. 3 shows the engineering stress-strain curves of the bare and coated Vit.1 pillars with different pH values. The corresponding mechanical properties from Fig. 3 are summarized in Table 1. The different pH values have slight influences on the plasticity of the coated Vit.1 pillars, and it is in relation to the P contents contained in the coatings. The residual stress tends to increase slightly with the increased pH values [30]. Thus, the Ni-P film with pH = 6.5 possesses the largest residual stress, and the coating with lager residual stress can induce larger stress



Fig. 1. Schematic of the electroless plating setup (1) Vit.1 pillar, (2) reactive solution, and (3) water bath.

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