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Role of copper addition on properties of bulk metallic glass materials



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ABSTRACT

In this paper the role of copper addition has been studied to examine thermal, mechanical, magnetic and corrosion properties of the Fe₄₃Co₂₂Ni₇B19Si₅Nb₄ bulk metallic glass alloy material. It is found that the copper addition in alloys exhibited low supercooled liquid region as compared to copper-free alloy. It is observed that microhardness was reduced in Cu-containing alloy with generation of shear bands along faces of micro indent impression while in Cu-free alloy straight and long microcracks appeared at the indent corners indicating its brittle behavior. The magnetic properties slightly decrease with Cu addition however the corrosion resistance of Cu-containing alloy showed superior properties indicating commercial benefits in aggressive environment.

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1. Introduction

Multicomponent bulk metallic glasses (BMGs) materials gained attraction in past years due to their distinctive properties [1]. There is no long range order in BMG materials thus can be classified as amorphous materials. They are also known as novel materials due to absence of compositional and microstructural variation which usually arises by presences of grain boundaries, precipitates and microsegregation. Therefore, these materials are considered as ideal materials with complete chemical homogeneity [2]. The BMG materials exhibited good physical, chemical, magnetic and electric properties [3]. The superior corrosion properties are demonstrated because of absence of crystal defects which are responsible for localized corrosion [2]. The interesting mechanical properties such as high compressive strength, high fracture toughness and wear resistance make them attractive for many engineering and commercial applications [3].

Furthermore, recent developments in BMG materials can allow fabrication in various alloy systems such as Pd, Zr, Fe, Cu, Mg and La and retain exceptional glass forming ability (GFA), as well as casting in thickness sections ranging from a few millimeters to centimeters using cooling rates of 100 Ks⁻¹ [1].

Fe-based BMG materials have gained importance for engineering applications owing to good soft magnetic properties and lower cost. However the commercialization of these materials was

* Corresponding author. *E-mail addresses*: Sidrajilani@hotmail.com (S. Jilani), khalid@giki.edu.pk (F.A. Khalid). hindered in the past because of section size was limited due to lower packing density which ultimately lowers glass forming ability (GFA) as reported in [4]. In 1995 Inoue group was able to synthesis and cast Fe-based BMG materials in millimeters dimension [5]. Shen and Schwartz developed the concept of microalloying for fabrication of Fe-based BMGs by using fluxing technique. Its GFA was enhanced by addition of transition metals along with metalloid [6]. Since then enormous Fe-based BMGs systems have been developed using copper mold casting and water quenching techniques [5].

Lu et al. developed and casted thicker section in centimeter scale of Fe-Cr-Mo-Mn-C-B BMG alloy [6]. Later Ponnambalam et al. [7] developed $Fe_{48}Cr_{15}Mo_{14}C_{15}B_6Y_2$ BMG alloy having critical thickness of 9 mm. Later Shen et al. [6] substituted 7 at% Co against Fe contents which enhanced the GFA of the alloy to cast with section thickness up to 16 mm. There is however limitation of high cost of high purity elements. Another problem is associated with the catastrophic failure of Fe-based BMG materials at room temperature thus restricting their wider applications [8–9].

In this investigation the role of copper addition in Fe-based BMG materials is studied in order to improve ductility with better corrosion resistance. It is demonstrated that introduction of copper can lead to change in facture behavior and shear bands propagation as a result redistribution of strain throughout the section of alloy samples of BMG materials. Furthermore, Cu addition in BMG materials is optimized to study its effect on thermal, mechanical and corrosion properties of Fe₄₃Co₂₂Ni₇B19Si₅Nb₄ bulk metallic glass material.



Fig. 1. XRD patterns of alloys showing amorphous structural phases.

2. Experimental procedures

The buttons of BMG alloys compositions of having Fe₄₃Co₂₂Ni₇B₁₉Si₅Nb₄ (Cu-free allov material) and $(Fe_{0,43}Co_{0,22}Ni_{0.7}B_{0,19}Si_{0,05}Nb_{0,04})_{99}Cu_1$ (Cu-added alloy material) were fabricated by electrical arc melting furnace using elements of high purity (99.98%) in titanium gettered argon environment. For promotion of homogenous mixing of elements, the button samples were reversely re-melted three to five times. Amorphous strips having dimensions of 1 mm thickness, 5 mm width and 20 mm length were produced by suction casting using a water-cooled copper mold. The BMG alloy samples were investigated by Philips X-ray Diffraction (XRD) using Cu Kα radiation at voltage of 40 KV and applied current of 25 mA. Thermal properties such as glass transition temperature (Tg), crystallization temperatures (Tx) were examined by differential scanning calorimetry (DSC) at a heating rate of 5 K/min under argon gas. Vickers microhardness tests were carried out using diamond indenter at load of 1.96 N and 4.90 N for 15 s respectively. Prior to indentation samples were ground up to 4000 µm grit and polished to ¼ µm. Magnetic properties were determined by a vibrating sample magnetometer (VSM) with applied field 10⁴ G at room temperature. For corrosion testing Gamry Potentiostat was used and the specimens were cut to the required dimensions of 7.5 mm \times 3.5 mm and mounted in polyester resin connected with an electric wire. The sample surfaces were polished and cleaned for corrosion tests that were carried out in 3.5 wt% NaCl solution.

Table 1

Thermal stability data for alloys: glass transition temperature Tg, crystallization temperature Tx, super cooled liquid region ΔTx , liquid temperature T₁, melting temperature Tm, reduced glass transition temperature Trg and the γ parameter.

| Alloys | Tg (°K) | T _x (°K) | ΔTx (°K) | T _m (°K) | T _I (°K) | $Trg = Tg/T_l$ | $\substack{\gamma=T_x/\\(T_g+T_l)}$ |
|----------|---------|---------------------|----------|---------------------|---------------------|----------------|-------------------------------------|
| Cu-free | 802 | 843 | 41 | 1308 | 1360 | 0.589 | 0.389 |
| Cu-added | 799 | 833 | 32 | 1307 | 1372 | 0.582 | 0.384 |

3. Results

3.1. Phase analysis

XRD patterns for Cu-free and Cu-added BMGs alloy samples in the as-cast condition are shown in Fig. 1. Both alloys revealed amorphous pattern with no traces of crystalline phase. This is demonstrated that amorphous structure is present in the alloys which is a result of optimization of compositional and processing parameters.

3.2. Thermal properties

DSC curves of BMG alloys are shown in Fig. 2. The thermal stability parameters are presented in Table 1. Tg and Tx decreased simultaneously with Cu addition, and the decline occurred from 802 °K to 799 °K and from 843 °K to 833 °K respectively. The supercooled liquid region Δ Tx also decreased gradually from 41 to 32 °K. The liquid temperature T₁ and melting temperature Tm are almost identical in both alloys and the endothermic peaks corresponded to the melting occur for both alloys at 1308 °K and 1307 °K. This aspect is consistent with the reduced glass transition temperature Trg=Tg/T₁ and γ parameter [Tx/(Tg+T₁)] reported in [10]. It was noted that copper addition led to the reduction in thermal properties. The exothermal peak corresponding to crystallization occurred in multiple steps for the Cu-added alloy which indicates that some crystalline phases may have formed with addition of Cu while for Cu-free alloy two sharp peaks occur for crystallization.

3.3. Microhardness

Microhardness of both BMG alloys was measured and it was observed that the hardness value of Cu-free alloy was slightly higher as compared to Cu-added alloy (Table 2). It was interesting to note that when the load is increased up to 4.90 N then straight microcracks occurred around the four corners of indent in Cu-free alloy (Fig. 3a). Whereas a shear band morphology was found along faces of indents which eventually converted into radial microcracks (Fig. 3b). Similar behavior of microcracks is evident in the alloys as shown in Fig. 4.

The fracture toughness K_{IC} of Cu-free alloy corresponding to straight corner cracks was calculated using the following equation.

$K_{IC} = 0.018 \times (H_v \times 10^3) \times (a \times 10^{-3})^{0.5} \times (c - a/a)^{-0.5} \times (H_v/E)^{-0.4} (c/a < 2.5)$

where 'a' is half-length of the Vickers impression (mm), c is half-length of the crack (mm), E is Young's modulus (GPa) and using the values of E=210 GPa [11,12]. The data and average values of K_{1C} is 12.26 MPam^{1/2}, yield strength was calculated



Fig. 2. DSC traces for BMG (a) Cu-free alloy (b) Cu-added alloy.

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