Journal of Alloys and Compounds 742 (2018) 318–324

Contents lists available at ScienceDirect

Journal of Alloys and Compounds

journal homepage: http://www.elsevier.com/locate/jalcom

Ductile FeNi-based bulk metallic glasses with high strength and excellent soft magnetic properties

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ARTICLE INFO

Article history:
Received 16 November 2017
Received in revised form
22 January 2018
Accepted 24 January 2018

Keywords:
FeNi-based BMGs
Large plasticity
Soft magnetic properties
p-d hybrid bonds

ABSTRACT

Fe-based bulk metallic glasses (BMGs) have attracted great attention due to their excellent soft magnetic properties and high fracture strength, but few applications have been materialized as structural materials because of their brittleness at room temperature. Here, we successfully synthesized an $Fe_{39}Ni_{39}B_{14.2}Si_{2.75}P_{2.75}Nb_{2.3}$ BMG which exhibits large plastic strain of 7.8%, high fracture strength of 3.35 GPa and excellent soft magnetic properties, i.e., rather high saturation flux density of 0.88 T, low coercive force of 0.7 A/m and high permeability of 20800. The results indicated that the mutual repulsion between ductility and strength could be renovated in the Fe-based BMGs by the rearrangement of atomic configurations through Ni addition. With proper combination of non-directional metal-metal bonds and directional metal-metalloid bonds, the mechanical properties of FeNi-based BMGs can be improved. Our studies provide a guideline in designing ductile FeNi-based BMGs with high strength, large GFA and excellent soft magnetic properties.

1. Introduction

Fe-based metallic glasses have attracted great research interest ever since their first synthesis in 1967 [1], because of the combination of superior anticorrosion property, magnetic and mechanical properties, and relatively low manufacturing cost [2–5]. Especially after the discovery of Fe-based bulk metallic glasses (BMGs) in 1995 [6], they were considered to possess promising applications as structural materials. Unfortunately, Fe-based BMGs usually fracture catastrophically when deformed at room temperature (RT) [7,8], e.g., undergoing only a few percent of plastic strain in compressions [9,10], which limits their widespread applications. Therefore, improving the ductility of Fe-based BMGs at RT has been an important goal over the recent decades. Tremendous efforts have been devoted in enhancing the ductility of Fe-based BMGs. A number of ductile Fe-based BMGs, such as (Fe, Co, Ni)-B-Si-Nb [11], Fe-Ni-P-B [12], Fe-Ni-Nb-B [13], Fe-Al-P-C-B [14] and Fe-Co-B-Si-Mo [15] have been already reported and their plastic strain to failure in compression was shown to be in the range of 0.5–5% [11–15]. The high plastic strain reported so far for Fe-based alloys which contain a glassy matrix is almost 40% [16]. Recently, ductile Fe-Ni-P-C BMGs with above 20% compressive plastic strain were synthesized [17,18]. However, the improvement of ductility inevitably leads to the deterioration of soft magnetic properties, fracture strength and GFA of Fe-based BMGs [17,19]. Therefore, it poses a serious challenge to develop ductile Fe-based BMGs with high strength, large GFA and good soft magnetic properties.

The aim of the present research was focused on improvement of ductility for Fe-based BMGs with high strength and soft magnetic properties. Based on previous reports on the positive effect of Ni addition on ductility and GFA of Fe-based BMGs [20], an alloy system from Fe-B-Si-P-Nb [21–23] with partial substitution of Fe by Ni was developed. The $Fe_{78-x}Ni_xB_{14.2}Si_{2.75}P_{2.75}Nb_{2.3}$ (x = 0, 7.8, 15.6, 23.4, 31.2 and 39) BMGs with large compressive strain and high strength, as well as excellent soft magnetic properties by copper mold casting were successfully synthesized. X-ray photoelectron spectroscopy (XPS) signatures have been obtained to examine the change in bonding states during brittle to ductile transition in the FeNi-based BMGs. This work provides guidance to design new

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https://doi.org/10.1016/j.jallcom.2018.01.317
0925-8388/

ductile FeNi-based BMGs with high strength and excellent soft magnetic properties.

2. Experimental

Alloy ingots were prepared by induction melting of high-purity Fe (99.99%), Ni (99.99%), B (99.99%), Si (99.999%), Nb (99.95%) and pre-alloyed Fe-P ingots that consist of 75% Fe and 25% P in a purified argon atmosphere. Ribbons with thickness of 25 μm and width of 1.5 mm were produced by single roller melt-spinning method. Cylindrical rods with diameters of 1–2 mm were produced by copper mold casting in a pure argon atmosphere. Thermal properties including glass transition temperature (T_g), crystallization temperature (T_x) and supercooled liquid region ($\triangle T_x$) were measured using differential scanning calorimetry (DSC, NETZSCH DSC404F3) at a heating rate of 0.67 K/s. The structure of rods were examined using X-ray diffraction (XRD, D8-Discover, Bruker) with Cu Kα radiation. As the magnetic properties depend on the sample sizes, in the interest of clarification the intrinsic soft magnetic properties of this metallic glass system, ribbon samples with similar size mentioned above were used for measurement. Saturation magnetization (B_s) under a maximum applied field of 800 kA/m was measured with a vibrating sample magnetometer (VSM). Coercivity (H_c) was measured with a DC B-H loop tracer under a field of 800 A/m. Effective permeability (μ_e) at 1 kHz was measured with an impedance analyzer under a field of 1 A/m. All of the ribbon samples for magnetic property measurements were annealed at the temperature of T_g-50 K for 600 s in order to reduce the influence of inner stress on soft magnetic properties through structural relaxation. Mechanical properties were measured by compression testing with a mechanical testing machine at RT using a strain rate of 5×10^{-4} s^{-1}. The specimens were cut from the as-cast glassy rods with a gauge aspect ratio of 2:1 (1 mm in diameter). The deformation behaviors and fracture surface were examined by scanning electron microscopy (SEM, Sirion 200, FEI). The bonding states of the samples with the addition of Ni element were examined by X-ray photoelectron spectrograph (XPS) using a Kratos AXIS ULTRA^{DLD} instrument with a monochromic Al Kα X-ray source ($hv = 1486.6$ eV).

3. Results and discussion

The X-ray diffraction patterns confirmed that all the melt-spun ribbons used for thermal and magnetic tests are composed of a full glassy phase without crystallization. Fig. 1 shows DSC curves of the melt-spun $Fe_{78-x}Ni_xB_{14.2}Si_{2.75}P_{2.75}Nb_{2.3}$ (x = 0, 7.8, 15.6, 23.4, 31.2 and 39) metallic glasses. The samples exhibit a glass transition followed by a large supercooled liquid region and then crystallization. T_g and T_x decrease gradually from 780 to 710 K and from 810 to 755 K, respectively, with an increase in the Ni content from x = 0 to x = 39. Additionally, the ΔT_x increases from 30 to 45 K with an increase in the Ni content to x = 39. Thus, the thermal stability of the supercooled liquid effectively increases with an increase in the Ni content to x = 39.

Based on the results of the DSC measurement and the thermal stability analyses, we expect the GFA of this system to be effectively enhanced. Therefore, we tried to form cylindrical glassy rods with different diameters up to 2 mm by the copper mold casting method. The glassy rods were produced at all compositions in Fe-Ni-B-Si-P-Nb system. The critical diameter for formation of a glassy single phase was 1 mm at x = 7.8 and 15.6, 1.5 mm at x = 23.4, 2 mm at x = 31.2 and 39. All the cast glassy rods with different diameters were characterized by XRD using Cu-Kα radiation, as shown in Fig. 2. The XRD patterns only contained broad peaks without crystalline peaks indicating the formation of a glassy phase. Their as-cast surfaces all appeared smooth and lustrous. No apparent volume reduction was recognized on their surfaces indicating that there was no crystallization during the formation of these samples. The reasons why Ni is effective in improving the GFA in the Fe-Ni-B-Si-P-Nb glassy system are based on the empirical component rules for achievement of high GFA [24]. The mixing enthalpies between Ni and Fe, Si, B, P and Nb atomic pairs are −2, −40, −9, −26 and −30 kJ/mol, respectively [25]. It is well known that large negative mixing enthalpies between the constituent elements lead to a highly stable supercooled liquid, which is consistent with the DSC results in Fig. 1.

Fig. 3 shows the hysteresis loops of melt-spun $Fe_{78-x}Ni_xB_{14.2}Si_{2.75}P_{2.75}Nb_{2.3}$ (x = 0, 7.8, 15.6, 23.4, 31.2 and 39) metallic glasses. The insert is the hysteresis curves measured by DC *B–H* loop tracer. It is clear that the B_s decreases monotonically from 1.39 to 0.88 T upon increasing the Ni content from x = 0 to x = 39, and this can be attributed to the lower magnetic moment of Ni (0.6 μ_B) compared with that of Fe (2.2 μ_B) [26]. With an increase in the Ni

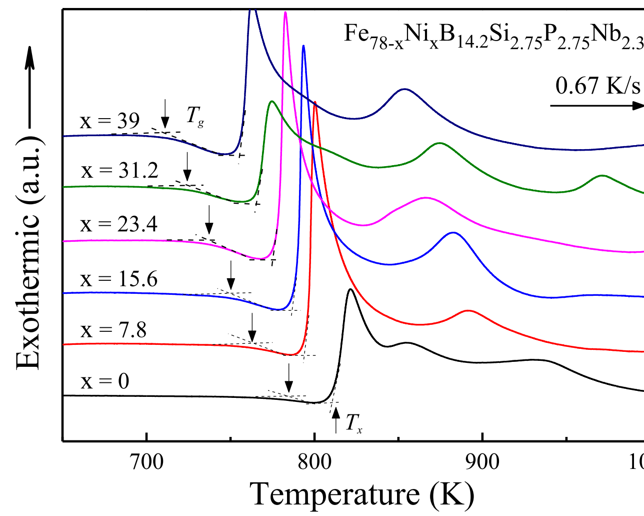

Fig. 1. The DSC curves of melt-spun $Fe_{78-x}Ni_xB_{14.2}Si_{2.75}P_{2.75}Nb_{2.3}$ (x = 0, 7.8, 15.6, 23.4, 31.2 and 39) metallic glasses.

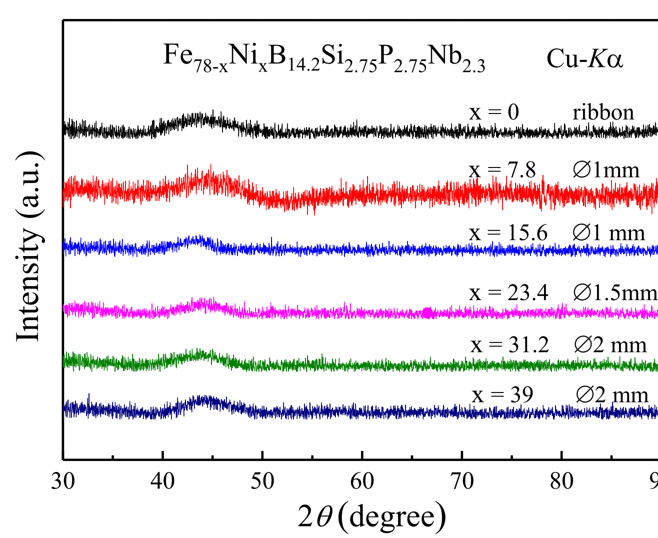

Fig. 2. XRD patterns of as-cast $Fe_{78-x}Ni_xB_{14.2}Si_{2.75}P_{2.75}Nb_{2.3}$ (x = 0, 7.8, 15.6, 23.4, 31.2 and 39) BMGs with corresponding critical maximum diameters.