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Journal of Magnetism and Magnetic Materials

journal homepage: www.elsevier.com/locate/jmmm



Magnetic properties of $(Fe_{1-x}Ni_x)_{72}B_{20}Si_4Nb_4$ (x=0.0-0.5) bulk metallic glasses

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ARTICLE INFO

Article history:
Received 8 October 2012
Received in revised form
22 January 2013
Available online 11 February 2013

Keywords:
Bulk metallic glass
Saturation polarization
Curie temperature

ABSTRACT

The effects of partial substitution of Fe by Ni in $(Fe_{1-x}Ni_x)_{72}B_{20}Si_4Nb_4$ (x=0.0, 0.1, 0.2, 0.3, 0.4, and 0.5) bulk metallic glasses (BMGs) on their magnetic properties were studied. It is found that the saturation polarization decreases from 1.15 T to 0.69 T with increasing Ni content from x=0.0 to x=0.5; the Curie temperature reaches its maximum of 598 K for composition x=0.1, and is then followed by a quick decrease with higher Ni content. Moreover, the random anisotropy and mean field theory were developed to investigate the magnetic properties of these BMGs, and the variations of saturation polarization and Curie temperature were well explained by calculating the magnetic exchange stiffness constant A and the nearest neighbor transition-metal-pair exchange interactions J by using the model.

1. Introduction

Bulk metallic glasses (BMGs) have attracted great attention due to their unique properties and potential applications [1–3]. Among them, the Fe-based BMGs are more attractive for application since they not only possess many good properties, such as excellent soft magnetic properties [4,5], besides ultrahigh strength [6-8] and good anticorrosion property [9], but are also much cheaper in comparison to other BMGs [10]. Recently, Fe-based BMGs have been intensively investigated and their critical size has been increased [11-16]. It is found that partial substitution of Fe with other magnetic elements Ni or Co may remarkably enhance the GFA [17,18] and change the soft magnetic properties due to compositional variation [19,20]; nevertheless, it is also found that the effects of Ni and Co on magnetic properties are somewhat different from each other [21,22], even if they are at neighboring positions in the Element Periodic Table. However, little attention concerning how and why the magnetic properties change induced by partial substitution of Fe by Ni was paid, and even less detailed theoretical research on the mechanism of partial substitution of Fe with constituting elements in Febased BMGs was carried out.

In the present work, we investigated the effects of partial substitution of Fe by Ni on the thermal characteristics and magnetic properties of Fe-B-Si-Nb BMGs considering its high GFA and excellent magnetic properties [12,18]. On this basis, the random anisotropy and mean field theory of the magnetic properties for $(\text{Fe}_{1-x}\text{Ni}_x)_{72}\text{B}_{20}\text{Si}_4\text{Nb}_4$ (x=0.0, 0.1, 0.2, 0.3, 0.4, and 0.5) were developed to explain the variations of saturation polarization and Curie temperature.

2. Experiments

Multi-component $(Fe_{1-x}Ni_x)_{72}B_{20}Si_4Nb_4$ (x=0.0, 0.1, 0.2, 0.3, 0.4, and 0.5) ingots were prepared by arc melting the mixtures of Fe (99.99%), Ni (99.99%) and Nb (99.99%) metals, and B (99.5%) and Si (99.999%) crystals in an argon atmosphere. Cylindrical alloy rods with diameters of 1.5 mm were produced by the copper mold casting method. The structures of as-cast samples were identified by X-ray diffraction (XRD) with Cu $K\alpha$ radiation. The thermal stability of the glassy samples was examined using a NETZSCH 404 C differential scanning calorimeter (DSC) at a heating rate of 0.67 K/s under a flow of high purity argon. The Curie temperature of the as-cast alloys was measured by a physical property measurement system (PPMS) at a heating rate of 0.167 K/s. For magnetic properties investigation, M-H hysteresis loops were measured with a vibrating sample magnetometer (VSM) at ambient temperatures. Additionally, the density of the specimens was measured using Archimedes's method with the accuracy less than 1%.

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3. Results and discussion

Fig. 1 shows the XRD patterns for all six as-cast rods with diameter of 1.5 mm. It can be seen that each pattern contains only broad maxima without any additional sharp Bragg peaks, and the diffraction images did not show any traces of crystallinity. This is characteristic of fully amorphous samples. It is found that as the content of Ni increases, the diffraction angle of the principal diffuse peak in XRD pattern is almost unchanged. The position of an X-ray halo maximum is known to be directly related to the average radius of the first coordination shell, r_1 , according to the Bragg equation: $2r_1 \sin \theta = \lambda$ with λ the X-ray wavelength and 2θ the scatter angle corresponding to the halo maximum. The almost unchanged first peak might be due to the almost equal radius of Ni (0.125 nm) and Fe (0.124 nm) [23]; even if Fe was replaced by Ni, the value of r_1 remains almost unchanged, which leads to nearly invariant 2θ .

Fig. 2(a) presents the DSC traces of the as-cast $(Fe_{1-x}Ni_x)_{72}$ $B_{20}Si_4Nb_4$ (x=0.0, 0.1, 0.2, 0.3, 0.4, and 0.5) BMGs measured at a constant heating rate of 0.67 K/s. It can be seen that all DSCs exhibit one endothermic event characteristic of the glass transition from the amorphous solid to the supercooled, followed by a crystallization exothermic reaction, which further confirms the glassy nature of the rods. Although T_g and T_x decrease from 837 to 776 K and from 882 to 834 K, respectively, ΔT_x increases gradually from 45 to 58 K with increasing Ni content to x=0.5. Thus, it is suggested that the thermal stability of the supercooled liquid increases with increasing Ni. Fig. 2(b) describes DSC curves revealing the cooling behavior of the same Fe-based BMGs system, which demonstrates that the $Fe_{72}B_{20}Si_4Nb_4$ BMG exhibits the highest T_1 and T_e of 1479 and 1405 K, respectively, as well as the largest temperature interval (T_1-T_e) of 74 K, implying that this composition does not lie in the vicinity of a eutectic point. With increasing Ni content from x=0.1to x=0.5, the T_1 and T_2 decrease gradually from 1471 to 1298 K and from 1405 to 1291 K, respectively, indicating that the alloy approaches a eutectic point in this process; especially when x=0.4, T_1 is very close to T_e ; thus, it is considered to be the closest to the eutectic point in this alloy system.

3.1. Saturation polarization

As expected, the fully glassy samples display excellent magnetic properties. Table 1 summarizes the saturation magnetization M_S (given in Am²/kg and in Bohr magnetons μ_B per magnetic atom), saturation polarization J_S and Curie temperature T_C as well as the density ρ for as-cast samples. Here, the saturation

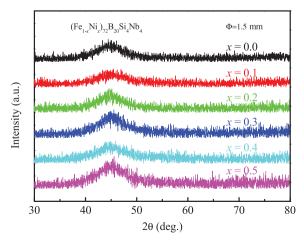
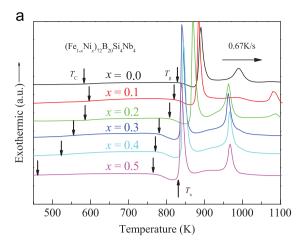


Fig. 1. XRD patterns of the cast $(Fe_{1-x}Ni_x)_{72}B_{20}Si_4Nb_4$ (x=0.0, 0.1, 0.2, 0.3, 0.4, and 0.5) alloys rods with diameters of 1.5 mm.



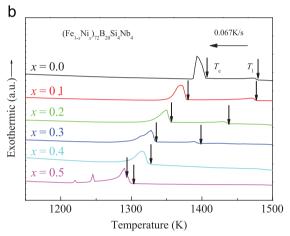


Fig. 2. (a) DSC traces measured at a constant heating rate of 0.67 K/s for cast $(Fe_{1-x}Ni_x)_{72}B_{20}Si_4Nb_4$ (x=0.0, 0.1, 0.2, 0.3, 0.4, and 0.5) alloys. (b) DSC traces measured at a constant cooling rate of 0.067 K/s for cast $(Fe_{1-x}Ni_x)_{72}B_{20}Si_4Nb_4$ (x=0.0, 0.1, 0.2, 0.3, 0.4, and 0.5) alloys.

Table 1 Magnetic properties data for $(Fe_{1-x}Ni_x)_{72}B_{20}Si_4Nb_4$ glassy rods: magnetization at saturation M_S (given in A m²/kg and in Bohr magnetons μ_B per magnetic atom), saturation polarization J_S , and Curie temperature T_C . Additionally, the density ρ is also shown.

Atomic fraction x	M_S (A m ² /kg)	$J_{S}\left(\mathrm{T}\right)$	$M_S(\mu_B)$	$T_{C}\left(K\right)$	$ ho~({ m g/cm^3})$
0.0	123.2	1.15	1.25	581	7.4377
0.1	115.4	1.08	1.14	598	7.4493
0.2	104.9	0.99	1.05	588	7.4868
0.3	95.5	0.91	0.95	565	7.5794
0.4	81.6	0.78	0.81	525	7.6559
0.5	70.8	0.69	0.71	458	7.7576

polarization was calculated by using the actual density values $(J_S=4\pi\times 10^{-7}\rho M_S)$, as presented in Fig. 3, and the saturation polarization as a function of Ni content is shown in Fig. 4. We can see that the saturation polarization decreases monotonically from 1.15 to 0.69 T upon increasing Ni content from x=0.0 to 0.5.

The variation of saturation polarization as a function of Ni content should be explained in more detail. From the random anisotropy theory [24], the correlation between the saturation polarization and the exchange stiffness constant *A* can be expressed as

$$J_S = \frac{8 \times 10^{-7} \pi \rho A}{H_{ex} R_a^2} \tag{1}$$

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