Magnetic and Mechanical Properties of Nanocrystalline Fe-Ni-Nb-B Alloys

J. Turčanová¹, J. Marcin¹, J. Kováč¹, D. Janíčkovici², P. Švec² and I. Škorvánek¹

¹ Institute of Experimental Physics, Slovak Academy of Sciences, SK-040 01 Košice, Slovakia
² Institute of Physics, Slovak Academy of Sciences, SK-842 53 Bratislava, Slovakia

E-mail address: turcanova@saske.sk

Abstract. The effects of Fe replacement by Ni on the formation of nanocrystalline structure and on the magnetic and mechanical properties were investigated in the series of (Fe₁₋ₓNiₓ)₈₁Nb₇B₁₂ (x = 0, 0.14, 0.25, 0.33 and 0.5) alloys. The analysis of phase evolution upon primary crystallization indicates that with an increase of Ni content in the samples the nanocrystalline grains exhibit transition from BCC-phase to a mixed state with coexistence of BCC- and FCC-phase and finally the FCC-FeNi phase is formed. The value of the Curie temperature of the amorphous phase, T_C(am), increases with an addition of Ni. We show that a replacement of Fe by Ni results in an improved magnetic softness and it has a beneficial effect on the bend ductility of amorphous and nanocrystalline samples.

1. Introduction

The nanocrystalline Fe-based alloys prepared by the crystallization of amorphous precursors represent an important class of soft magnetic materials. The discovery of the extremely soft nanocrystalline FINEMET alloys with typical composition Fe₇₃.₅Cu₁.₅Nb₁.₃Si₁.₃B₉ [1] has stimulated an enormous research activity in these materials. Subsequently, the two other families of the soft magnetic nanocrystalline alloys based on Fe-M-B-(Cu) (M=Zr, Nb and Hf), called also NANOPERM [2], and Fe-Co-M-B-(Cu) (M=Zr, Nb and Hf), called also HITPERM [3], have been developed. These three major families of nanocrystalline soft magnetic alloys are composed of BCC-nanocrystalline grains embedded in amorphous residual phase. Simple BCC-grains are present in NANOPERM, while the grains exhibiting D0₃ or B2 ordering are usually observed in FINEMET and HITPERM alloys, respectively. Considerably less attention has been devoted to other types of nanograins in the soft ferromagnets. Recently, the NiFeZrB alloys with coexistence of BCC- and FCC-FeNi grains as well as magnetically ordered cubic (FeNi)₂₃B₁₂ nanograins have been prepared [4,5]. The mixture of BCC- and FCC-FeNi grains have been observed also upon the crystallization of the FeNiMoSiB amorphous alloys [6].

The aim of the present paper is to investigate the magnetic and mechanical properties in the series of heat treated (Fe₁₋ₓNiₓ)₈₁Nb₇B₁₂ (x = 0, 0.16, 0.25, 0.33 and 0.5) alloys. It is shown that this alloy system is a suitable candidate for getting a deeper insight into the microstructure-property relationship in the FeNi-based nanocrystalline soft magnetic alloys. We report on the beneficial effects of the Ni addition on the magnetic and mechanical characteristics of these soft ferromagnetic materials.
2. Experimental

Master alloys have been prepared from elements with purity better than 99.95%. Amorphous ribbons 6 mm wide and 20-25 µm thick have been prepared by the method of planar flow casting. Pieces of these ribbons were isothermally annealed under high vacuum for 1 hour at 773 K and 873 K. The microstructure and crystallization behaviour of the samples was investigated by transmission electron microscopy (TEM). Formation of the nanocrystalline phase was studied by X-ray diffraction techniques (XRD). The temperature dependencies of magnetization were measured by the vibrating sample magnetometer (VSM). The ductile/brittle behaviour of the thermally treated samples was determined by using simple bending tests. Samples have been bent into a U-shape between two parallel plates, the separation of which decreased until fracture occurred. From the separation of the plates at failure, the relative strain at fracture can be calculated [7].

3. Results and Discussion

3.1. Microstructure

The changes in microstructure upon heat treatment of parent amorphous alloys were examined by TEM. The presence of ultrafine nanocrystalline grains is evident from the micrographs shown in Fig. 1. The micrographs on the Figs. 1 (A) – (C) correspond to the samples at different stages of the primary crystallization process. The typical grain dimensions are 5-10 nm. The grain sizes tend to decrease with an increase of Ni content. The (Fe0.5Ni0.5)81Nb7B12 sample annealed at temperature higher than optimal one (see Fig. 1 (D)) show a presence of the additional big grains of secondary crystallization phase that was identified as (Fe,Ni)23B6.

The analysis of phase evolution upon nanocrystallization has been performed by XRD. The as-quenched samples show the presence of the only amorphous phase. After nanocrystallization, the single BCC-phase in the nanograins has been observed for the samples Fe81Nb7B12 and (Fe0.80Ni0.20)81Nb7B12 annealed 1 hour at 773 K and 873 K. With a further increase of Ni content, the coexistence of the BCC- and FCC-phase has been detected for the samples (Fe0.75Ni0.25)81Nb7B12 and (Fe0.67Ni0.33)81Nb7B12. The sample (Fe0.50Ni0.50)81Nb7B12 annealed 1 hour at 773 K contains mostly the FCC-FeNi phase, while after the annealing of this sample for 1 hour at 873 K, the formation of (Fe,Ni)23B6 phase was clearly detected.

Figure 1. TEM micrographs of the samples (A, B) (Fe0.75Ni0.25)81Nb7B12 annealed at 773 K and 873 K and (C, D) (Fe0.5Ni0.5)81Nb7B12 annealed at 773 K and 873 K.

3.2. Magnetic behaviour

The temperature dependences of magnetization for the (Fe1-xNi0.5)81Nb7B12 amorphous samples (where x = 0, 0.14, 0.25, 0.33 and 0.5) are depicted in Fig. 2 (a).
The first fall of magnetization after increase of the measuring temperature corresponds to the ferro-paramagnetic transition of the amorphous phase. The values of Curie temperature of amorphous phase, $T_{C\text{(am)}}$, were determined from the corresponding derivatives of the $M(T)$ dependencies (Fig. 2 (b)) in the temperature range where the amorphous material undergoes the ferro-paramagnetic transition. The value of the Curie temperature of the amorphous phase, $T_{C\text{(am)}}$, increases with addition of Ni from 331 K (for $x=0$) to 574 K (for $x=0.33$) and then slightly decreases for the sample $(\text{Fe}_{0.5}\text{Ni}_{0.5})_{81}\text{Nb}_{7}\text{B}_{12}$ to 564 K. The increase of magnetization in the high temperature range (above 700 K) indicates the beginning of the crystallization process, which results in the formation of ferromagnetic particles with the higher value of Curie temperature than is the measuring temperature. The magnetization vanishes again in the temperature range close to the Curie temperature $T_{C\text{(cr)}}$ of the final crystalline product. The values of the $T_{C\text{(cr)}}$ show a clear tendency to a decrease with an increase of the Ni content in the samples.

Figure 3. Dependence of coercivity versus concentration of Ni for the $(\text{Fe}_{1-x}\text{Ni}_x)_{81}\text{Nb}_{7}\text{B}_{12}$ samples annealed at 773 K.

Figure 4. The relative strain of fracture as a function of annealing temperature for the amorphous and nanocrystalline $(\text{Fe}_{1-x}\text{Ni}_x)_{81}\text{Nb}_{7}\text{B}_{12}$ samples.
The dependence of the coercivity versus concentration of nickel for the \((\text{Fe}_{1-x}\text{Ni}_x)_{81}\text{Nb}_{12}\) samples annealed 1 hour at 773 K is shown in Fig. 3. The addition of nickel has a beneficial effect on the magnetic softness of the heat treated nanocrystalline samples. Here, the soft magnetic properties were found for the alloy \((\text{Fe}_{0.50}\text{Ni}_{0.50})_{81}\text{Nb}_{12}\) annealed 1 hour at 773 K. As shown in Fig. 1(d), an increase of the annealing temperature to 873 K results in the appearance of the big grains of secondary crystallization phase. The higher magnetocrystalline anisotropy of such grains consequently lead to a drastic deterioration the soft magnetic behaviour [8].

3.3. Mechanical properties

The evolution of ductile/brittle behaviour with increasing annealing temperature is shown in Fig. 4. The value of relative strain at fracture \(\varepsilon_f\) corresponds to ductile samples, which can be bent through 180° without fracture. If the ribbon fails upon bending, \(\varepsilon_f\) is smaller than unity and the more brittle the sample is the smaller the \(\varepsilon_f\). The value of relative strain at fracture of the Fe-rich amorphous samples drops abruptly with an increase of annealing temperature above 600 - 650 K and the substantial degree of embrittlement can be seen already in amorphous state. Similar behaviour was observed for other FINEMET and NANOPERM-type alloys [7, 8]. With an increase of the nickel concentration in the samples, the embrittlement level in both amorphous and nanocrystalline state has a tendency to decrease.

4. Conclusion

In this paper we have studied the microstructural, magnetic and mechanical properties of soft magnetic amorphous and nanocrystalline \((\text{Fe}_{1-x}\text{Ni}_x)_{81}\text{Nb}_{12}\) alloys (where \(x = 0, 0.16, 0.25, 0.33 \text{ and } 0.5\)). The main findings can be summarized as follows:

The TEM and XRD characterization of the samples indicates that with an increase of Ni content in the samples the nanograins of 5-10 nm in size exhibit transition from BCC-phase to a mixed state with coexistence of BCC- and FCC-phase and finally the FCC-FeNi phase is formed. The value of the Curie temperature of the amorphous phase, \(T_{C\text{am}}\), increases with addition of Ni. The replacement of Fe by Ni results in an improved magnetic softness of the nanocrystalline samples that correspond to the primary crystallization stage. The Ni-rich alloys exhibit a lower tendency towards the embrittlement in the amorphous phase as compared with the ternary FeNbB.

Acknowledgement:

This work was supported by the projects APVV-0413-06, VEGA 2/7193, APVV-0454-07 and Cex-NANOSMART.

References

[1] Y. Yoshizawa, S. Oguma, K. Yamaguchi, J. Appl. Phys. 64 (1988) 6044
[2] K. Suzuki A.Makino, A. Inoue, T. Masumoto, J. Appl. Phys. 70 (1991) 6232
[3] M.A. Willard, D.E. Laughlin, M.E. McHenry, D. Thoma, K. Sickafus, J.O. Cross, V.G. Haris, J.Appl. Phys. 84 (1998) 6773
[4] M.A.Willard, J.H.Claassen, V.G.Harris, Proc. First IEEE Conf. on Nanotechnology (2001).p. 51
[5] M. Kopcewicz, B. Idzikowski, J. Kalinowska, J. Appl. Phys. 94 (2003) 638
[6] S. Saiseng P. Winotai, S. Nilpairuch, P. Limsuwan, I.M. Tang. J. Magn. Magn. Mater. 278 (2004) 172
[7] I. Škorvánek, R. Gerling, J. Appl. Phys. 72, 3417-22 (1992)
[8] I. Škorvánek, P. Švec, J. M. Grenache, J. Kováč, J. Marcin and R. Gerling: J. Phys.: Condens. Matter 14 (2002) 4717