Magnetostriction and dilatation measurements of Fe-Ni-Nb-B metallic glasses

G. Vlasák1, P. Švec1, J. Turčanová2, D. Janičkovič1, I. Škorvánek2, P. Švec, Sr.1

1Institute of Physics, Slovak Academy of Sciences, Dúbravská cesta 9, 845 11 Bratislava, Slovakia
2Institute of Experimental Physics, Slovak Academy of Sciences, Watsonova 49, 040 01 Košice, Slovakia

E-mail: fyzivlas@savba.sk

Abstract. Soft magnetic metallic glasses or their corresponding nanocrystalline counterparts based on Fe and on Ni frequently exhibit different magnetic properties. This work is devoted to rapidly quenched (Fe-Ni)81Nb7B12 ribbon system with the ratio of Ni/Fe being 0, 1/6, 1/3, 1/2, 1, 2 and 3 prepared by planar flow casting. Using temperature dependencies of dilatation, the Curie temperature Tc and structural transitions (glass transition, crystallization) were determined. Direct measurement of magnetostriction was employed to determine the values of saturation magnetostriction $\lambda_S$ at room temperature. The values for as-quenched samples with low Ni-content (0, 1/6) clearly indicate the proximity of Tc to room temperature and thus the values of $\lambda_S$ are low. The highest value of $\lambda_S$ observed for Ni/Fe = 1/3 is 15.86 ppm. Increasing Ni-content leads to decrease of $\lambda_S$ down to 1 ppm for Ni/Fe = 3, in accord with its negative value observed in pure Ni.

1. Introduction

Materials research indicates important influence of the solidification process of alloys on their properties. Especially rapid solidification in diverse alloy systems with quenching rates up to $10^6$K/sec. can improve their physical properties or lead to new and unusual ones. This is the case of e.g. ferromagnetic rapidly quenched alloys with soft magnetic properties where the information about their magnetostriction is often crucial. Linear isotropic magnetostriction, which corresponds to the deformation of the material, depends on external magnetic field H, temperature T and on intrinsic properties, e. g. chemical and topological short range order, CSRO and TSRO, respectively, on homogeneity, density of internal stresses and shape. From the phenomenological description of magnetoelastic coupling in isotropic magnetic medium one can obtain $\lambda_s(H) = 1/3 \omega(H) + 3/2 \lambda_F \cos 2\Theta - 1/3 + \lambda_S$ with $\lambda_S$ being the saturation magnetostriction, $\omega(H) = \Delta V/V$ the volume magnetostriction, $\lambda_F$ the bipolar magnetostriction (form effect) and $\Theta$ is the angle between the direction of deformation measurement and the direction of the applied magnetic field $[1]$, where $\lambda_S(H) = 2/3[\lambda_{par}(H) - \lambda_{perp}(H)]$, $\omega(H) = \Delta V/V = \lambda_{par}(H) + 2\lambda_{perp}(H)$. The quantities $\lambda_{par}(H)$ and $\lambda_{perp}(H)$ in saturation as well as $\lambda_S$ can be determined as in $[2]$. Due to the quantity $\omega(H)$ being often not well defined, in technological applications the quantity $\delta\omega(H)/\delta H$ is used instead, denoted as isotropic forced volume magnetostriction, which is constant in a
certain region of the $\omega$ dependence. The coefficient of thermal dilatation $\alpha$ can, in certain ferromagnetic materials, exhibit anomalies in the vicinity of the Curie temperature and below. The coefficient $\alpha$ can range from typical positive values down to zero and can even attain negative values. This effect is called the invar effect, dependent on spontaneous volume magnetostriction $\omega_{\text{spon}}$, which, in turn, depends on magnetic moment.

The choice of alloy composition leading to properties interesting from physical and technical aspects is important, especially when preparing the systems in amorphous state by rapid quenching from the melt; combinations of suitable metals such as iron or nickel with additions of niobium and with metalloids like boron, carbon or silicon are generally used. In our work we focus our attention to the Fe-Ni-Nb-B system.

2. Experimental

Alloy systems with composition (Fe-Ni)$_{81}$Nb$_7$B$_{12}$ and the ratio Ni/Fe = 0, 1/6, 1/3, 1/2, 1, 2, and 3 were prepared in the form of master alloys. Rapidly quenched ribbons 6 mm wide and approx. 20 microns thick were prepared by planar flow casting. Chemical composition of the alloys was determined by inductionally coupled plasma spectroscopy. X-ray diffraction and transmission electron microscopy were used to check and confirm the amorphous state of the samples.

Field dependencies of linear magnetostrictions at room temperature, $\lambda_{\text{par}}(H)$ and $\lambda_{\text{perp}}(H)$ were determined by application of external in-plane magnetic field along the ribbon axis and in perpendicular direction, respectively. Samples for magnetostriction measurements were prepared from the ribbons in form of 6 mm discs by chemical etching. Measurements were performed on a special device [2] designed and constructed at the Institute of Physics SAS. Selected dependencies of $\lambda_{\text{par}}(H)$ and $\lambda_{\text{perp}}(H)$ are shown in figure 1 and figure 2, respectively. These dependencies were used to calculate the values of saturation magnetostriction $\lambda_S$ according to [2, 3] as well as the field dependencies of volume magnetostriction $\omega(H)$; the latter were used to calculate the values of forced volume magnetostriction $\partial \omega(H)/\partial H$. Compositional dependencies of $\lambda_S$ and $\partial \omega(H)/\partial H$ are shown in figure 3a as a function of the ratio of Ni/Fe.

In order to determine the value of $T_C$ and glass transition temperature $T_g$ in these alloys dilatation as a function of temperature was measured using a special dilatometer designed and adapted for measurements on rapidly quenched thin ribbons [3, 4, 5]; ribbon samples with the length of 30 mm were used.

3. Results and discussion

The measured dependencies of $\lambda_{\text{par}}(H)$ and $\lambda_{\text{perp}}(H)$ are affected, besides the parameters entering the phenomenological relation, by the position of the Curie temperature $T_C$ with respect to the temperature of measurement (room temperature). In addition, these dependencies are influenced by the concentration ratio of Ni to Fe, as shown in figures 1 and 2.

It is well known that the $\lambda_S$ values tend to vanish when the temperature of measurement approaches the Curie temperature of ferromagnetic material. The Curie temperature of the alloy with Ni/Fe=0 (355K) is only slightly above the ambient temperature therefore, its room temperature saturation magnetostriction is very low. The increase of $\lambda_S$ for the alloy with Ni/Fe=1/6 to the values close to 8 ppm is thus explained by the increase of its Curie temperature far above the room temperature.

The dependencies $\lambda_{\text{par}}(H)$ and $\lambda_{\text{perp}}(H)$ obtained on the alloy with Ni/Fe=1/3 are similar to those observed on isotropic materials with low anisotropy in the direction of the long ribbon axis. This anisotropy is observed in all Fe-Ni-Nb-B alloys with low Ni concentration. The field dependencies of magnetostriction in these alloys exhibit an initial sharp increase due to strong response of the domains and their walls to relative low applied magnetic fields followed by saturation effect. Similar considerations are valid also for the case of Ni/Fe=1/2.
Figure 1. Field dependencies of parallel (full symbols) and perpendicular (open symbols) magnetostrictions $\lambda_{\text{par}}(H)$ and $\lambda_{\text{perp}}(H)$ for Fe-Ni-Nb-B samples with ratio Ni/Fe = 0, 1/6, 1/3 and 1/2.

Figure 2. Field dependencies of parallel (full symbols) and perpendicular (open symbols) magnetostrictions $\lambda_{\text{par}}(H)$ and $\lambda_{\text{perp}}(H)$ for Fe-Ni-Nb-B samples with ratio Ni/Fe = 1/1, 2/1 and 3/1.

Figure 3. Dependencies of the saturation magnetostriction $\lambda_S$ and forced volume magnetostriction $\frac{\partial \omega(H)}{\partial H}$ (a) and of the Curie temperature $T_C$ and glass transition temperature $T_g$ (b) on the ratio Ni/Fe.

Figure 2 indicates that alloys with high Ni-content (Ni/Fe = 2, 3) do not form such types of domains – do not contain regions, which would react sharply to the presence of magnetic field. The resulting effect of lower magnetostriction without saturation can be due to the presence of higher
amount of Ni, which in non-alloyed state exhibits negative magnetostriction and thus decreases the overall magnetostriction of the alloy. Such a behaviour differs from that for sample with Ni/Fe = 1, where the sufficiently strong response of the domains and domain walls in relative low magnetic fields can still be expected, leading thus to the observed sharp increase of magnetostriction in the initial stages of magnetization.

Concentration dependence of forced magnetostriction $\partial \omega(H)/\partial H$ exhibits a local minimum at Ni/Fe = 1/2, falling down to almost zero. This indicates decreasing internal stresses and deformations. Due to this it is possible that also individual atom dipoles can contribute more easily to the magnetization and thus to the magnetostriction effect.

Dilatation measurements on as-quenched samples show a continuous change of the thermal dilatation coefficient $\alpha$. These changes are caused by diverse volume changes, which take place in the material due to structure transformations and which are superimposed on thermal vibrations of the building units. The transformations taking place are relaxation processes, volume changes related to the invar effect (in iron-containing iron-rich alloys), glass transition and crystallization stages. Relaxation processes, reversible or irreversible, are realized mainly at lower temperatures, usually in the still amorphous matrix. The invar effect is related to T_C; at temperatures below T_C a deformation of material due to spontaneous volume magnetostriction $\omega_{\text{sp}}$ takes place. This effect can also be used for determination of T_C (figure 4) using a method described in [6]. Further temperature increase leads to glass transition (figure 5). The glass transition temperature T_g determined on the investigated systems as a function of Ni/Fe ratio is shown in figure 3b.

![Figure 4](image-url)

**Figure 4.** Temperature dependencies of dilatation for Fe-Ni-Nb-B samples with varying ratio of Ni/Fe; heating rate 5K/min; curves offset for clarity. Triangles and dropped lines indicate positions of the Curie temperature.
Figure 5. Temperature dependencies of dilatation for Fe-Ni-Nb-B samples with varying ratio of Ni/Fe; heating rate 5K/min. Triangles and dropped lines indicate positions of the glass transition temperature.

The compositional dependence of $T_C$ (figure 3b) exhibits a sharp increase leading to a local maximum at Ni/Fe = 1/2 and then a slow decrease for higher Ni content while $T_g$ decreases slowly with increasing Ni/Fe. The values of $T_C$ obtained from magnetization measurements are shown for comparison and although their values are slightly lower, they exhibit a good correlation with those derived from dilatation curves.

4. Conclusions

The investigated Fe-Ni-Nb-B system in amorphous as-cast state indicates a strong influence of the ratio Ni/Fe on magnetic properties, especially on magnetoststriction and subsequently on the values of $\lambda_S$ and $\partial\omega(H)/\partial H$ due to the changes of magnetization polarization. The effect of Ni/Fe ratio is also systematically notable on the values of the Curie temperature and on the glass transition temperature.

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