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Investigations on the Magnetic Properties of High-Coercivity $(Nd_{1-x}Fe_x)_{90}Al_{10}$ Bulk Amorphous Alloys

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ABSTRACT

Bulk rods with diameters up to 3 mm prepared by suction casting and respectively mould casting and melt-spun amorphous ribbons with thicknesses in the range 25 – 150 μm with compositions $Nd_{90-x}Fe_xAl_{10}$ (x = 35; 40; 45; 50) were investigated by XRD, DSC and magnetic measurements in the temperature range 5 – 600 K. The microstructure and magnetic properties are strongly dependent on the cooling rate, preparation process and Fe content. The large values of the coercive field, which amount to 320 kA/m in the as-cast state as well as the increase with decrease of the temperature and cooling rate result from the formation of very small metastable or nonequilibrium magnetic clusters.

INTRODUCTION

The interest in glass-forming alloys, which vitrify at relatively low cooling rates from the molten state, compared with conventional rapidly quenched metallic glasses has grown in the last years. Owing to their resistance to crystallization, these easy-glass forming alloys can be cast in bulk shape with dimensions of millimeters. However, no results on bulk amorphous alloys with ferromagnetic properties at room temperature have been reported earlier than 1995 [1].

Recently, it has been found that Nd-Fe-(Al,Si) ternary amorphous alloys are formed in a wide range of compositions by melt spinning and mould casting technique and exhibit large coercive fields at room temperature [2,3]. Their magnetic behavior indicates that they are structurally glasses but magnetically granular with coercive fields as high as 8.4 T at low temperatures in an applied field of 30 T [4]. These results are in contradiction to those found in conventional $Nd_2Fe_{14}B$ ternary amorphous alloys, in which the amorphous microstructure gives rise to soft magnetic characteristics with negligible coercivities, but they are in agreement with the high coercivities obtained in the past for Nd-Fe binary amorphous alloys.

We performed investigations on the glass formability, thermal stability and magnetic properties of $Nd_{90-x}Fe_xAl_{10}$ melt-spun amorphous ribbons with thicknesses between 25 and 150 μm and cast amorphous rods up to 3 mm in diameter produced by mould casting and suction casting techniques, respectively, in order to clarify the relationship between microstructure and coercivity in these bulk amorphous alloys.

EXPERIMENTAL DETAILS

$Nd_{90-x}Fe_xAl_{10}$ master alloys with x = 35; 40; 45; 50 were prepared from Fe (99.99 %), Nd (99.9 %) and Al (99.99%) in an arc furnace under Ar atmosphere, and re-melted several times for homogenisation. Ribbons with thicknesses between 25 and 150 μm and widths of 3 to 5 mm were obtained by single roller melt-spinning method in vacuum or in Ar atmosphere, at surface velocities of the Cu wheel ranging from 30 to 2.5 m/s. Bulk amorphous alloys with diameters up to 3 mm were formed in a wide composition range of 35 to 50 at. % Fe by mould casting and suction casting techniques.

The structure of the samples was checked by X-ray diffraction (XRD) using Cu-Kα radiation. For the ribbons, the structure was checked on both sides as well as on powders obtained by milling the ribbons, while structural investigations on cast rods were performed using powders obtained after crushing the rod in small pieces and then milling.

Magnetisation measurements above room temperature, at applied magnetic fields limited to 1.6 T, were carried out using a Vibrating Sample Magnetometer (VSM). The magnetisation and coercive field below room temperature (5 – 300 K) were studied using a Superconducting Quantum Interference Device (SQUID) Magnetometer in external magnetic fields not exceeding 1 T. Each sample was thermally demagnetised prior to recording each measurement.

RESULTS AND DISCUSSION

The largest diameter obtained for an amorphous rod is 3 mm for nominal composition $Nd_{55}Fe_{35}Al_{10}$, and glass forming ability decreases by increasing the Fe content. XRD patterns indicate also fully amorphous structures for ribbons with thicknesses below 100 μm and for rods with diameters less than 1 mm, regardless of their Fe content. The increase of the Fe content over 45 at. % results in the on-set of crystallization in melt-spun ribbons thicker than 100 μm.

These alloys crystallize through a single exothermic reaction and exhibit very large values for the reduced crystallization temperature (T_x/T_m) of 0.87 to 0.92, as it can be seen in figure 1 (T_x and T_m are the crystallization and eutectic melting temperatures, respectively). The amount of the crystallization energy (the area under the exothermic peak) is higher for melt-spun ribbons in comparison with the rods that contain a higher amount of Fe due to the more disordered amorphous structure developed in the first ones. It is worth to note that in contradiction with the results obtained for other amorphous alloys, neither endothermic peak nor glass transition temperatures are evidenced for Nd-Fe-based amorphous alloys. The crystallization energy doesn't change significantly as a function of the Fe content for $Nd_{90-x}Fe_xAl_{10}$ thin ribbons 30 μm thickness, whereas for cast rods it strongly decreases with the increase of the Fe content as a consequence of the decrease of the glass-forming ability. This behavior is explained by the insufficient cooling rates assured by the casting techniques for the alloys with larger contents of Fe and indicates a strong correlation between the composition, the cooling rate and the glass-forming ability for Nd-Fe-based ternary amorphous alloys.

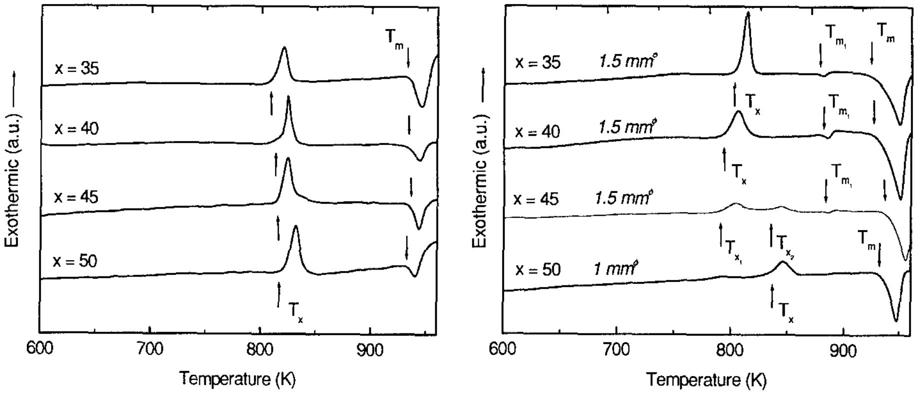

Figure 1. DSC curves plotted for $Nd_{90-x}Fe_xAl_{10}$ melt-spun ribbons and cast rods as a function of Fe content

$Nd_{90-x}Fe_xAl_{10}$ (x = 35 – 50) amorphous alloys exhibit large coercive fields at room temperature, even for low applied fields, regardless of their thickness and preparation techniques. In figure 2 are presented comparatively the hysteresis loops as a function of the Fe content for 3 different kinds of $Nd_{90-x}Fe_xAl_{10}$ samples: fully amorphous thin ribbons 25 μm in thickness (a), fully amorphous (x = 35; 40) and for comparison, partially vitrified (x = 50) thick ribbons with a thickness of about 120 μm (b) and fully amorphous cast rods 1 mm in diameter (c). The magnetization increases about 2 times by increasing the Fe content from 35 to 50 at. %, regardless of the amorphous samples shape, the same behavior being observed for high fields (up to 10 T). The coercive field presents a strong dependence on the composition. One observes that the on-set of crystallization in the $Nd_{40}Fe_{50}Al_{10}$ thick ribbon results in the decrease of the magnetization, while the coercive field has almost the same value as those obtained for fully amorphous thick ribbons with a less content of Fe because of the existence of the amorphous residual phase. Thus, the large coercive fields of Nd-Fe-based ternary bulk amorphous alloys are related to the existence of the amorphous phase, its disappearance leading to the drastically decrease of the coercive field and moreover, to the disappearance of the ferromagnetic properties [5]. The decrease of the Fe content results in the decrease of the coercive field of about 2 times for thin amorphous ribbons and in its increase of about 2 times for amorphous cast rods. For thick amorphous ribbons the coercive field is almost the same, regardless of the Fe content. For higher external fields (larger than 3 T) the coercive field doesn't change significantly with the Fe content for each different type of ribbon or for cast rods.

The large values of the coercive field can be explained by assuming the existence of very small Fe-Nd magnetic clusters dispersed in the amorphous matrix, whose size approaches a single magnetic domain, in agreement with the previous results obtained for melt spun Nd-Fe amorphous alloys [6]. The high values obtained for coercive fields of the thick amorphous ribbons comparatively with thin ones are related to the more relaxed microstructure developed in the first ones. The microstructure is more homogenous in amorphous rods due to their dimensions and lower cooling rates and consequently the values obtained for coercive field are smaller than for the thick ribbons. The difference in the absolute value of the coercive field as a function of the amorphous samples' thickness results from the volume rate between Fe-Nd magnetic clusters and the homogenous Nd-rich matrix through the cooling rate.

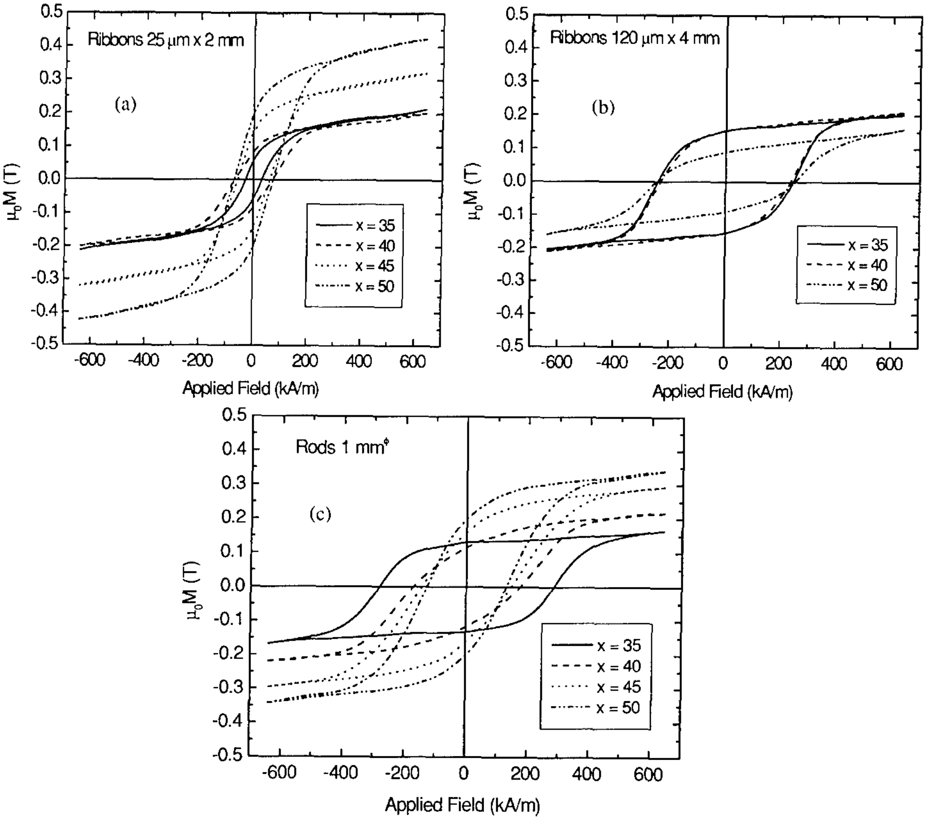

Figure 2. M-H hysteresis loops of $Nd_{90-x}Fe_xAl_{10}$ melt-spun ribbons 25 and respectively 120 μm in thickness and cast rods 1 mm in diameter, at room temperature

In order to obtain more information on the magnetic coupling in short range ordered magnetic structures existent in $Nd_{90-x}Fe_xAl_{10}$, the type of interactions as well as their influence on the coercive fields, the dependence of the magnetisation and coercive field was carried out at low temperatures (between 5 and 300 K). Figure 3 presents the dependence on temperature of the magnetization and coercive field for $Nd_{90-x}Fe_xAl_{10}$ thin amorphous ribbons, thick amorphous or partially vitrified ribbons, and amorphous cast rods. The humps appearing in the coercive field over a narrow range (150-220 K) for amorphous ribbons containing a large amount of Nd (over 50 at. %) and for cast rods in the entire range of compositions suggest the existence of one pronounced anisotropy of the magnetic clusters. The decrease of the coercive field with the temperature is similar to that obtained for similar crystalline compounds and is believed due to a change of the size of the magnetic clusters domains, which probably approach to the superparamagnetic regime.

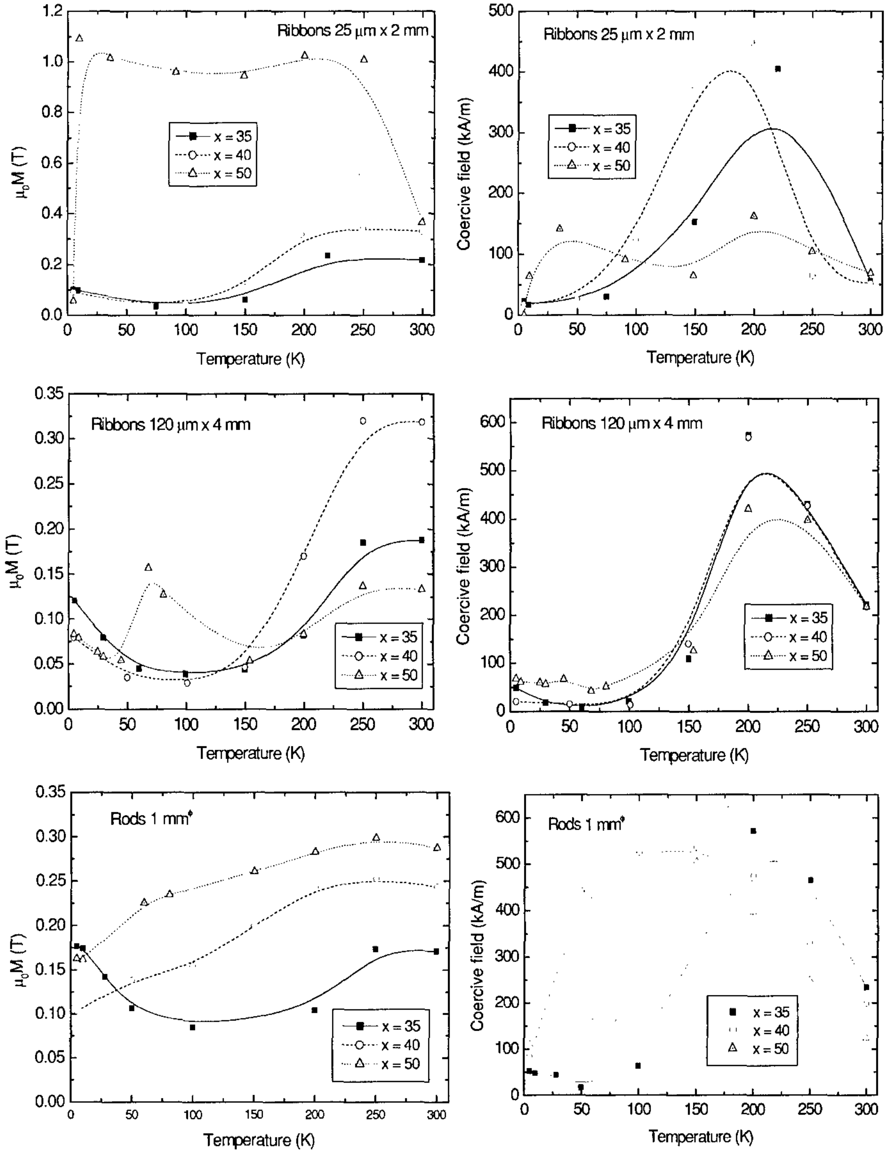

Figure 3. Temperature dependence of the magnetization at 1 T and coercive field for $Nd_{90-x}Fe_xAl_{10}$ amorphous ribbons and cast rods as a function of the Fe content

Due to the more homogenous structure existent in the thin ribbons the clusters size is smaller and consequently the interactions between them are not so stronger. The behaviour is typical for a spin-glass system with random configuration of the magnetic spins, in which thermal activation energy is enough to destroy the magnetic order. The coexistence of two types of magnetic order: long-range ferromagnetic order which gives the macroscopic behaviour, and short range ordered structures which determines the large values of the coercive fields in Nd-Fe-based bulk amorphous alloys is proved by the differences between zero-field-cooled and field-cooled magnetization curves and was reported elsewhere [7]. From the temperature dependence of the coercive field for $Nd_{40}Fe_{50}Al_{10}$ thin amorphous ribbon 25 μm in thickness one observes the existence of two maximum for the coercive field: one around 200 K which is similar to that obtained for the other bulk amorphous samples and the new one around 25 K, whose origin is unknown, but probably is caused by the larger amount of Fe which determines a more homogenous topological and magnetic structure. It is worth to note that the coercive field for this ribbons in the as cast state is the smallest one measured for $Nd_{90-x}Fe_xAl_{10}$ amorphous alloys with x=35~50. The magnetization increases with the temperature increase, this increase being more pronounced for thin ribbons 25 μm in thickness containing the largest amount of Fe in composition.

From the data presented here, it can be seen that the high coercivities obtained for $Nd_{90-x}Fe_xAl_{10}$ amorphous alloys at room temperature and their dependence on cooling rate and the preparation technique result from the formation of very small metastable or nonequilibrium magnetic clusters. The magnetic response of the Nd-rich matrix and Fe-Nd clusters phase are very different and very sensitive to any change in temperature and external field. Although these materials are currently below those considered necessary for economic viability, the mechanism that governs their unusual magnetic properties may be very interesting for basic research and consequently for finding new magnetic materials with granular structures for applications.

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