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INFLUENCE OF Gd ON THE STRUCTURE AND MAGNETIC PROPERTIES OF FINEMET*

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A set of $Fe_{73.5}Gd_xNb_3Cu_1Si_{13.5}B_9$ samples with Gd concentration $x = 0 - 3$ at% and the $Fe_{71.5}Gd_5Cu_1Si_{13.5}B_9$ alloy have been investigated: (i) concerning their ability to form nanocrystalline material after heat treatment and (ii) with respect to the Gd influence on the magnetic properties. All the samples showed a nanocrystalline structure after appropriate heat treatment, except the one with 5 at% of Gd. The observed decrease of the saturation magnetization with increasing Gd content indicates the ferrimagnetic exchange coupling between the Gd and Fe atoms. The Curie temperature was found to be increasing with Gd content up to 3 at% of Gd.

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1 Introduction

The nanocrystalline $Fe_{73.5}Nb_3Cu_1Si_{13.5}B_9$ alloy, generally known as FINEMET, possesses excellent soft magnetic properties and relatively high saturation magnetization, which arise from its typical multi-phase structure, composed of magnetically and structurally different phases: nanosized grains of α-FeSi solid solution or DO_3 type Fe_3Si crystallites embedded in an amorphous residual matrix.

We have replaced gradually the Nb and two Fe atoms by Gd, which has atomic diameter large enough (0.3602 nm) to retard the grain growth and has significant magnetic moment of 7.94 μ_B. Gd is the only rare-earth metal which is ferromagnetic at room temperature (T_C=292.7 K [1]), moreover, it is isotropic with no orbital momentum contribution to the total moment. We expected a considerable influence of Gd on the magnetic behaviour of the alloy through the Gd-Fe exchange interaction contribution.

2 Experimental

A set of $Fe_{73.5}Gd_xNb_{3-x}Cu_1Si_{13.5}B_9$ with $x = 0 - 3$ and the $Fe_{71.5}Gd_5Cu_1Si_{13.5}B_9$ (signed as FINEMET, Gd1, Gd2, Gd3 and Gd5) ribbon samples was prepared by the melt-spinning method

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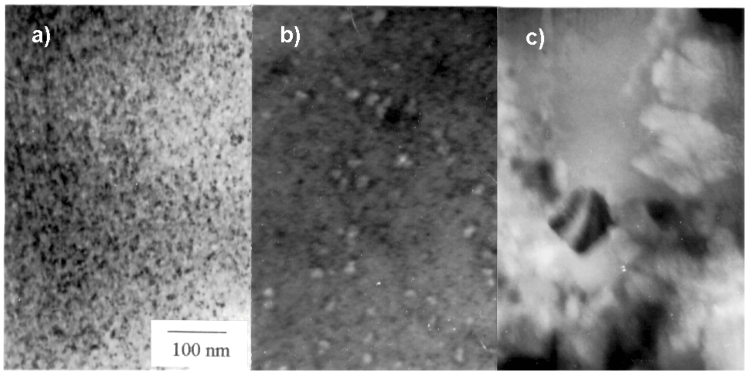

Fig.1. TEM microphotographs of the Gd3 sample annealed at a) 500^{o}C, b) 600^{o}C and c) the Gd5 sample annealed at 600^{o}C

and subsequently annealed at temperatures between 450-670^{o}C for 1 hour in a vacuum furnace. The structure was studied by TEM. The crystallization temperature of the first crystallization step T_{x1} and the Curie temperature T_C of the amorphous matrix were derived from thermomagnetic curves measured by vibrating sample magnetometer in external field of 0,4 T at the heating rate of 8 K/min from room temperature up to 800^{o}C . The temperature dependence of saturation magnetization over the temperature range of 4.2-300 K was measured in external field of 1 T. The values of saturation magnetization at 0 K, M_S, were calculated using the spin-wave theory [2]. The coercive fields were determined from the dc hysteresis loop.

3 Results and discussion

Structure: TEM investigations have revealed the nanocrystalline structure for all samples after annealing at 500-550^{o}C (Fig.1a), except the Gd5 sample. This sample crystallizes in different way. The formation of large dendritic Fe-Si-B grains (grain size more than 100 nm) was observed after annealing at 600^{o}C (Fig.1c), in spite of the fact that the sample after annealing at 550^{o}C was fully amorphous. The appearance of the dendritic Fe-Si-B grains on the regular nanocrystalline background was also detected in the Gd3 sample annealed at 600^{o}C (Fig.1b).

The influence of Gd on the crystallization temperatures is visible in Fig.2a. Gd, if together with Nb, lowers the onset temperature of the first crystallization T_{x1}, reflecting the low mutual solubility and chemical intolerance of these two elements in the alloy, that was also observed during the alloy processing. On the other hand, when Nb is fully replaced by Gd, T_{x1} increases. The second crystallization peak sharpens and shifts towards the first one with increasing Gd content, indicating the decrease of the second crystallization temperature T_{x2}. In the Gd3 sample the two peaks considerably overlap, the temperature range of existence of the nanocrystalline state narrows. The sharp and high second peak indicates besides the formation of borides the rapid transition to polycrystalline structure.

Magnetic properties: following the temperature dependence of magnetization the Curie temperature T_C of the amorphous matrix was derived from the $M^2(T)$ dependence. We found the

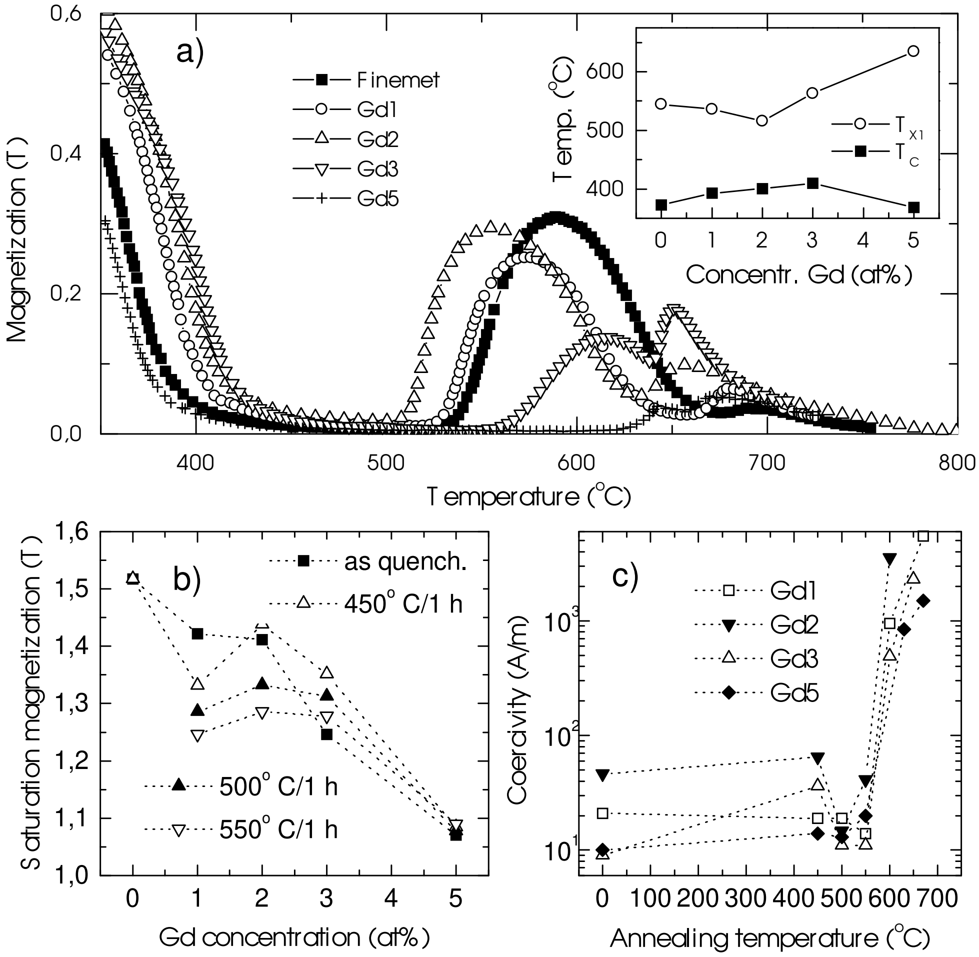

Fig.2. a) Temperature dependence of magnetization. b) Saturation magnetization as a function of Gd content. c) Coercivity as a function of annealing temperature.

T_C to be increasing as a function of Gd content - up to 3 at%. For the Gd5 sample the T_C drops off to nearly the same value as that for FINEMET (insert in Fig.2a).

Fig.2b shows the influence of Gd on the M_S of the samples. In the as-quenched amorphous state the values of the M_S are gradually decreasing with increasing Gd concentration. This fact confirmes the ferrimagnetic exchange coupling between the Gd and Fe atoms [3]. In the crystallized samples the ferrimagnetic screening effect of Gd atoms is not so effective (even a slight increase of M_S with increasing Gd concentration for samples annealed at 500 or 550°C is visible) because of the separation of the α-FeSi crystals and the Gd containing amorphous or crystalline phases.

Generally, in FINEMET-type alloys the M_S decreases after nanocrystallization [4]. This is also evident from Fig.2b as a remarkable drop in M_S after annealing at temperatures higher than 450^{o}C for the Gd1-Gd3 samples. Fig.2b corroborates the TEM results and underlines that the Gd5 sample is not nanocrystalline. We observed a noticeable increase of the M_S after annealing at 450^{o}C: the more Gd in the alloy, the more visible increase of M_S, excepting the Gd5 sample. This fact can be connected to the out of ribbon plane magnetization in the amorphous state, similarly to the case of amorphous $Fe_{77}Gd_3B_{20}$ [5]. Fig.2c shows the influence of annealing temperature on the coercivity. The dependences for the Gd1-Gd3 samples are typical for the nanocrystalline materials, the higher values of H_C measured for the Gd2 amorphous sample might be connected to the non-homogeneous chemical short-range order caused by the chemical intolerance of Gd-Nb atoms.

4 Conclusions

A set of Gd substituted FINEMET alloys with 0-5 at% of Gd was investigated with respect to the structure and magnetic properties. All the samples showed a nanocrystalline structure after appropriate heat treatment, except the one with 5 at% of Gd. This sample appeared to be not nanocrystalline. Gd, if together with Nb, lowers the onset temperature of the first crystalization T_{x1}. When Nb is fully replaced by Gd, T_{x1} increases. The Curie temperature of the amorphous matrix was found to be increasing with the raising amount of Gd in the samples - up to 3 at%. The ferrimagnetic exchange coupling between the Gd and Fe atoms was confirmed in as-quenched state, after annealing this coupling was not recognizable. The coercivities follow the structural evolution after annealing, the values were found to be higher than those for FINEMET.

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