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Variation of Magnetization and Curie temperature with Isothermal Annealing of Higher Cr Content of $Fe_{73.5-x}Cr_xCuNb_3Si_{13.5}B_9$ (x = 10, 12.5, 15 & 17.5) Amorphous Alloys

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Abstract

This is well known that amorphous state is metastable. Metastability of amorphous or glassy metal alloys offers the possibility of phase separation diffusion of various species and structural relaxation even though the alloys remains amorphous when they are annealed at temperature well below the crystallization temperature. All these changes have effect on the intrinsic magnetic properties such as Curie temperature (T_c) saturation magnetization (M_s) and magnetostriction. The present paper focuses that saturation magnetization, M_s and Curie temperature (T_c) of FINEMET alloys in the amorphous state increase on annealing up to the annealing temperature (T_a) corresponding to the early stage of crystallization, beyond which M_s and T_c both decrease [1-3].

Keyword: Amorphous, Curie temperature, Magnetization, Crystallization, Finemet.

Introduction

It has been well established through extensive research work that the addition of Cu and Nb simultaneously with Fe-Si-B based amorphous alloys is the necessary condition for the extraordinary soft magnetic properties of the Finemet alloy. This addition extends the temperature range between the primary crystallization α-Fe(Si) temperature, T_{x1} and secondary crystallization Fe-B temperature T_{x2} premiere for achieving superior magnetic properties [4]. It should be stressed again that good soft magnetic properties require not only a small grain size but at the same time the absence of boron compounds. The separation between the primary crystallization of bcc Fe and the precipitation of Fe-B compounds not only is determined by the Cu and Nb additions but, decrease with increasing boron content. This puts a further constraint on the alloy composition namely that the boron content should be kept at a low or moderate level in order to obtain an optimum nanoscaled structure.

In summary, the behavior of the onset of crystallization temperature and composition shows that a basic condition for the formation of typical nanocrystalline structure is given by a primary crystallization process before stable or meta-stable intermetallic phases are formed. Obviously, this can be attained by (i) alloying additions which lead clearly separated stages of crystallization at T_{x1} and T_{x2} and (ii) by annealing at $T_{x1} < T_a < T_{x2}$ such that only the phase forming at and above T_{x1} is crystallizing. Murillo *et al.* [5] studied the influence of Cu/Nb content and annealing conditions on the microstructure and the magnetic properties of FINEMET alloys. Grain size, phase composition and transition temperatures were observed to depend on the ratio of Cu/Nb content. The magnetic properties are strongly correlated to the microstructural features.

Numerous research works have been carried out to improve the properties of $Fe_{73.5}Cu_1Nb_3Si_{13.5}B_9$ FINEMET alloy by investigating the effects of substitution and addition of various elements.

Kwapulinski *et al.*[6] have studied the effect of Cr, Mo, Zr substituted for Nb of Fe-Cu-X-$Si_{13}B_9$ (X = Cr, Mo & Zr) amorphous alloys on the optimization of annealing temperature ($T_{op,}$) corresponding to the maximum permeability and found that it varies linearly with the atomic radius ($r_{a,}$) of the substituted elements in the order of T_{op} = Zr > Mo > Cr. This result to a certain extent reflects the dependence of the crystallization temperature (T_x) on the atomic radius [7]. Niobium can be substituted by other group V or VI refractory elements, like Cr, V, Mo, W or Ta which act similarly on the crystallization process and on the magnetic properties [2.15]. Like for Nb, the atomic volumes of these refractory elements are larger than that of Fe, which reduces the diffusion coefficients and, thus stabilizes the amorphous matrix and slows down the kinetics of grain coarsening [8]. Accordingly the efficiency of these elements for grain size refinement increases in the order of their atomic volumes, i.e., Cr < V < Mo $\approx$ W < Nb $\approx$ Ta. Thus, finest grain structures and superior magnetic properties in practice require at least a certain amount of the elements Nb or Ta.

Magnetic properties and magnetoimpedence effects have been studied of Mn substitution for Fe in the Finemet alloy $Fe_{73.5-x}Mn_xCu_1Nb_3Si_{13.5}B_9$ in the amorphous and nanocrystalline state [9-10]. Gomez-Polo *et al.* [9] showed that the partial substitution of Fe by Mn in FINEMET alloys plays an important role in the evolution of magnetic properties of the alloy. This has been manifested through the enhancement of magnetic hardening of the nanocrystalline state attributed to the migration of the Mn atoms to the grain boundary region, which reduces the exchange coupling between the crystalline and residual amorphous phases. Tho *et al.* [10] found a decrease of Curie temperature (T_c) and saturation magnetization (M_s) due to the substitution of Fe by Mn in the FINEMET composition $Fe_{73.5-x}Mn_xCu_1Nb_3Si_{13.5}B_9$ (x = 1, 3 & 5). The authors found that the presence of Mn affects the shape of the magnetic hysteresis loops causing a decrease in coercivity and maximum magnetization in the amorphous and annealed samples. The frequency dependence of magnetoimpedence (MI) as a function of frequency has been measured. The correlation between MI effect and soft magnetic properties has been sought in which the giant magnetoimpedence effect corresponding to the ultrasoft magnetic properties has been well established.

Experimental

Melt-spinning is a widely used production method for rapidly solidifying materials as well as preparing amorphous metallic ribbon [11-12]. In order to prepare amorphous of $Fe_{73.5-x}Cr_xCu_1Nb_3Si_{13.5}B_9$ alloys with x = 1, 2, 3, 4, 5, 6, 7, 8, 9, 10, 12.5, 15 and 17.5, a melt spinning facilities was used at the Centre for Materials Science, National University of Hanoi, Vietnam. The arc melted master alloy was crashed into small pieces and put inside the quartz tube crucible for re-melting by induction furnace using a medium frequency generator with a maximum power of 25 kW at a nominal frequency of 10 kHz.

Results and discussions

The FINEMET is ferromagnetic at room temperature. It is observed in the present investigation that the amorphous alloys $Fe_{73.5-x}Cr_xCuNb_3Si_{13.5}B_9$ with x = 10, 12.5, 15 & 17.5, which are paramagnetic (except x = 10) at room temperature show an increase of M at room temperature when annealed below the onset of crystallization temperature. Fig. 1 (a, b) and 2 (a, b) clearly demonstrates an increase of magnetization upon annealing temperature (T_a) for all these four samples. This increase of M with T_a up to 580^0C is connected to the structural relaxation and varying degrees of chemical disorder concomitant with the previous published results on typical FINEMET alloy [2. 13]. Similar increase in M_s due to structural relaxation has been detected in Fe-based metallic glass [3]. With reference to the enhancement of magnetization of annealed samples, DSC thermograms have been taken on samples annealed at T_a = 520^0C, 540^0C, 560^0C along with as cast samples and depicted Fig. 3. It is seen in fig 3 (a, b, c, d) that the area under the first crystallization event Fe(Si) phase slightly diminishes for x = 10 and 12.5 implying that initiation of crystallization seems to take place and accordingly M vs. H curves sharply rises and looks like ferromagnetic for x = 12.5 which is paramagnetic in the amorphous condition with T_c = 247 K. This increase of magnetization for the sample x = 12.5 is due to the evolution of ferromagnetic Fe(Si) crystallites. This has also been confirmed by X-ray diffraction pattern. Note that x = 10 is ferromagnetic at room temperature. For x = 15 up to T_a = 560°C, M increases only due to structural relaxation, while for x = 17.5 at T_a = 600°C a sharp rise of M correspond to initiation of crystallization of Fe(Si) phase. The influence of partial substitution of Fe by Cr on the soft magnetic properties of $Fe_{73.5-x}Cr_xCu_1Nb_3Si_{13.5}B_9$ (x = 1, 2, 3, 4 & 5) have been studied through the measurement of magnetic hysteresis loops of amorphous and annealed samples at room temperature.

Therefore an increase in magnetization M at room temperature is expected on annealed samples according to ref. [1, 2, 13, 14]. With the increase of magnetic induction on annealing, a subsequent decrease of coercive force (H_c) is noticed for all the samples implying magnetic softening of these alloys upon annealing. Similar low field hysteresis behavior has been observed in Cr substituted FINEMET with Au instead of Cu [15-17]. The observed improvement of soft magnetic characteristic of the annealed samples at T_a = 540°C for 30 min is likely due to the formation of α-Fe(Si) phase with optimum nanometric grains, their appropriate volume fraction and strong exchange coupling among them.

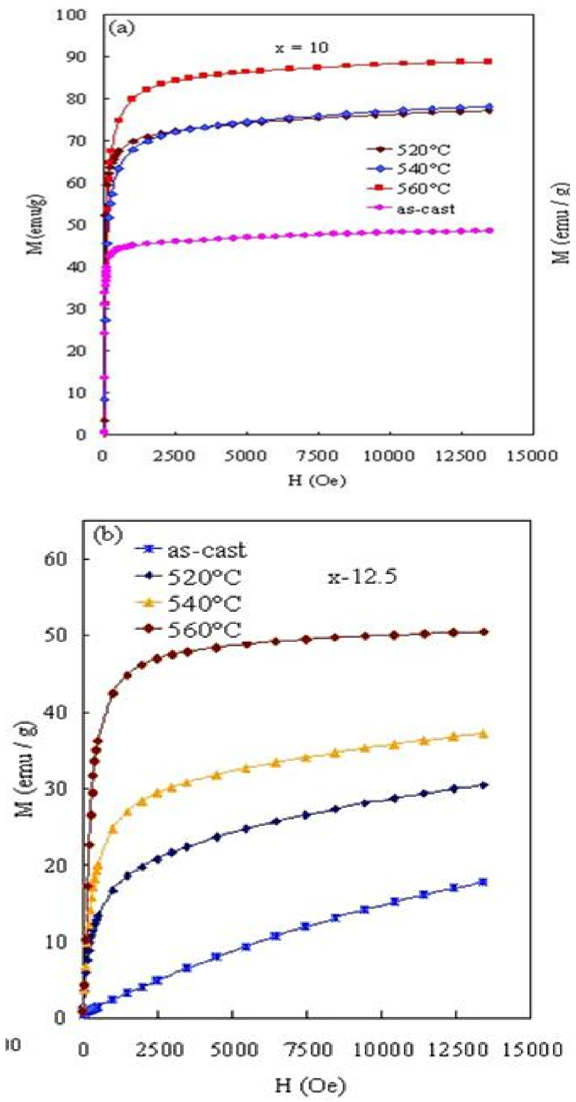

Figure 1: field dependence of magnetization curves of ribbons $Fe_{73.5-x}Cr_xCu_1Nb_3Si_{13.5}B_9$ with (a) x = 10 (b) x = 12.5 in the ascast and annealed at different frequencies

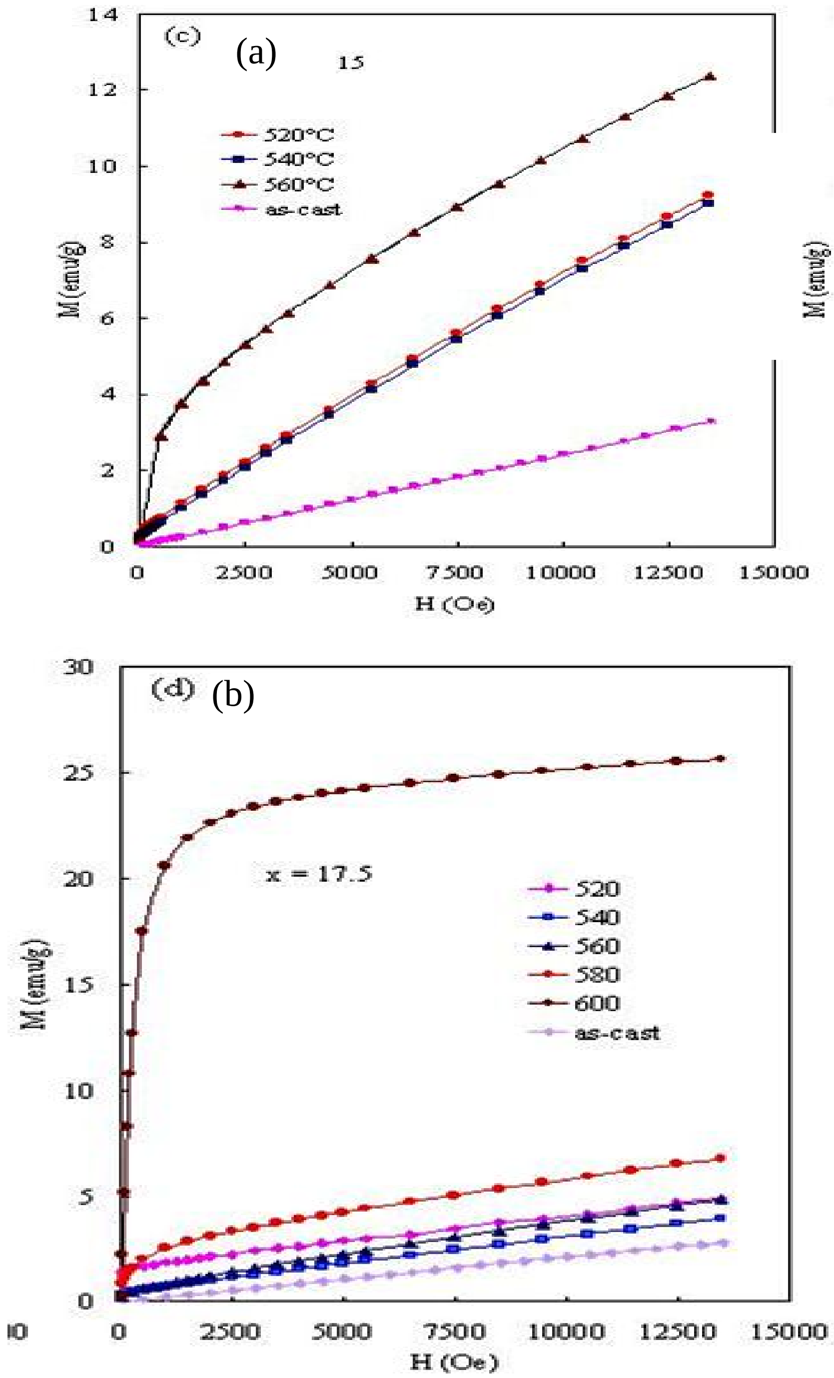

Figure 2: Field dependence of magnetization curves of ribbons $Fe_{73.5x}Cr_xCu_1Nb_3Si_{13.5}B_9$ with (a) x = 15 (b) x = 17.5 in the as cast and annealed at different temperatures

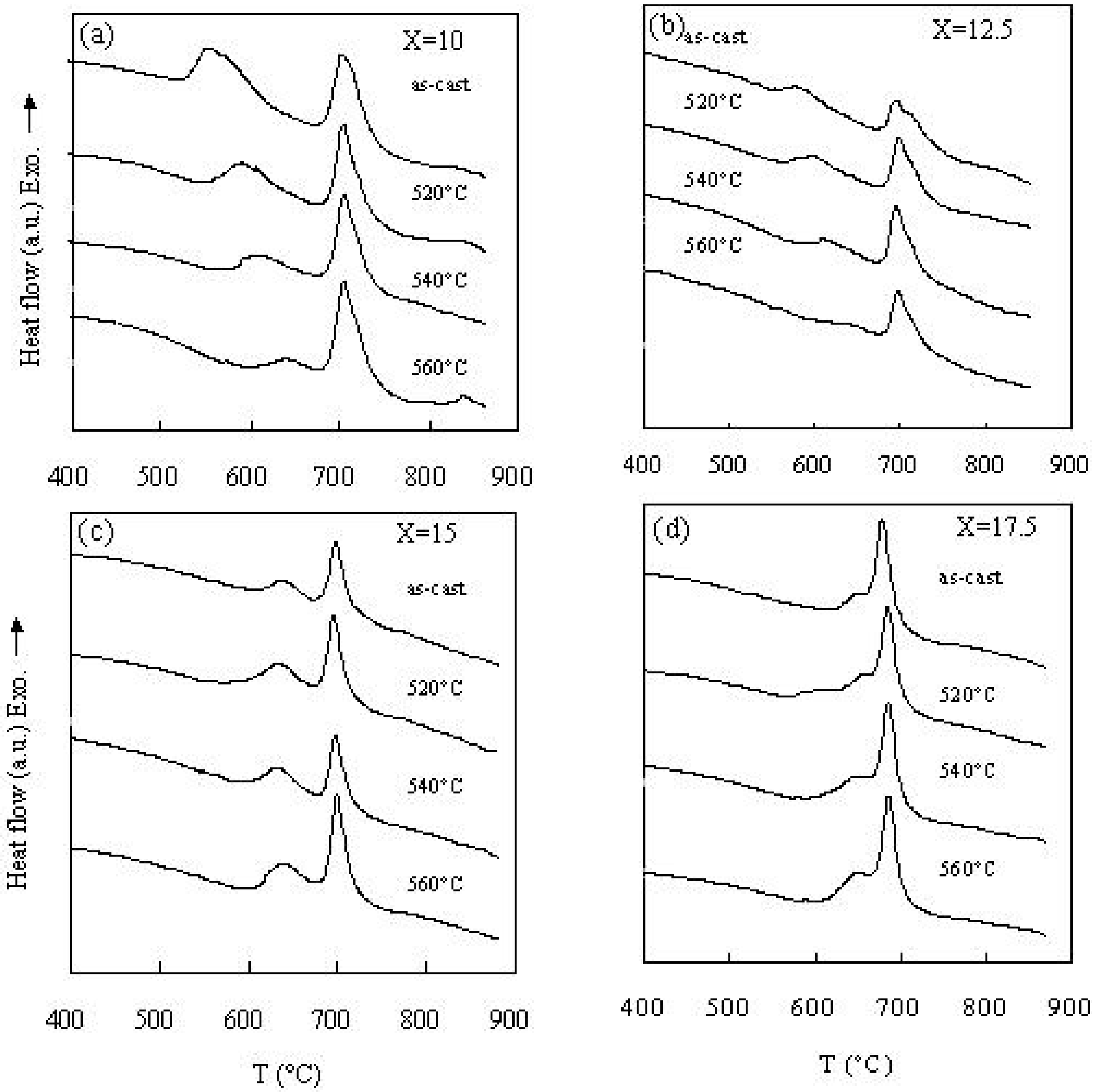

Figure 3: DSC curves of the ribbons with composition $Fe_{73.5-x}Cr_xCu_1Nb_3Si_{13.5}B_9$ (a) x = 10 (b) x = 12.5 (c) x = 15 (d) x = 17.5 in the as cast and annealed at different temperatures

Conclusion:

The effect of Cr substitution has been studied for an extended range for Fe in the $Fe_{73.5-x}Cr_xCu_1Nb_3Si_{13.5}B_9$ (x = 0, 1, 2, 3, 4, 5, 6, 7, 8, 9, 10, 12.5, 15 & 17.5) alloys on the crystallization behavior, structural and magnetic properties especially on the low temperature range. Although few reports are available on the effect of Cr substitution for Fe in a limited range, low temperature magnetization and higher Cr content

Finemet alloys have not been done in much detail. In the present work a detail study of the low temperature magnetization including the evolution of magnetic properties of higher Cr content alloys, which are normally paramagnetic in the as-cast state, was performed.

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