Journal of Engineering Science 01(02), 2010, 59-63

JES
an International Journal

EFFECT OF ANNEALING TEMPERATURE ON THE KINETICS OF STRUCTURAL RELAXATION AND MAGNETIZATION OF Fe-Cu-Nb-Si-B NANOCRYSTALLINE AMORPHOUS RIBBONS

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Received: 20 November 2010 Accepted: 08 December 2010

ABSTRACT

Nanocrystalline amorphous ribbons of compositions $Fe_{75.5}Cu_1Nb_1Si_{13.5}B_9$ (sample-A) and $Fe_{74}\ Cu_{0.5}Nb_3Si_{13.5}B_9$ (sample-B) prepared by rapid solidification technique have been studied in the present work. The crystallization behavior and the nanocrystal formation of the samples were performed by Differential Thermal Analysis (DTA) which in turn was supported by X-ray Diffraction (XRD) study. Magnetization measurements of the nanocrystalline samples were carried out using vibrating sample magnetometer (VSM). Throughout the study it was noticed that the crystallization onset temperature T_{x1} corresponding to primary crystallization of bcc Fe(Si) was higher for the sample-B (504°C) than that of the sample-A (470°C). This might reflects the fact that the sample-B obviously shows higher thermal stability. Above the crystallization temperature the saturation magnetization was found to be increased linearly with the increase in annealing temperature up to 525^0C for both the samples and then decreases with the increase of annealing temperature. The decrease in saturation magnetization might be attributed with the enrichment of the residual amorphous phase with Nb that weakens the coupling between the ferromagnetic nanograins.

Keywords: *Amorphous ribbons, Annealing, Structural relaxation, Magnetization*

1. INTRODUCTION

Nanocrystalline materials designate a noble type of interface-controlled solids that are characterized by a structural modulation on the length scale of several nanometers. The nanocrystalline state is achieved by subsequent heat treatment from their as cast amorphous precursor above the primary crystallization temperature. Over the past few decades, amorphous and more recently nanocrystalline materials have been investigated for applications in magnetic devices requiring magnetically soft material such as transformers, inductive devices, etc. Owing to the ductility of nanocrystalline ceramics at low temperatures, soft magnetic nanocrystalline alloys, or tailoring of new types of alloys, research interest in the soft magnetic nanostructured materials has been increasing at an accelerating rate (H. Gleiter 1989; H. Gleiter *et.al.* 1993 and R. Birringer *et. al* 1994). Recent advances in materials synthesis and characterization techniques realized that these materials exhibit many unique and interesting physical, optical, magnetic and chemical properties with a number of potential technological applications (T. Kulik *et. al.* 1994; Jing Zhi *et. al.* 1996; S. N. Kane *et. al*. 2000; M. El. Ghannami *et. al.* 1994; J. Bigot *et. al*. 1994 and M. Hasiak *et. al*. 2000). Magnetic properties of nanocrystalline materials depend upon the interaction mechanism and on the ratio between structural and magnetic correlation lengths. Exploitation of Fe-Cu-Nb-Si-B noble material in practical applications started shortly after the discovery and manufactured by Hitachi Ltd. under the trade names FINEMET (Y. Yoshizawa *et. al.* 1989) and VITROPERM (Vacuum schmelze Gmbh 1993). Amorphous ferromagnetic materials based on Fe-Si-B show good magnetic properties when they are heat-treated below their crystallization temperature. Fe-Cu-Nb-Si-B alloys exhibit exceptionally high permeability of magnitude two orders higher than their conventional Fe-Si-B alloys due to the heat treatment just above the crystallization temperature. The great scope of technical applications of nanocrystalline magnetic material with composition $Fe_{73.5}\ Cu_1\ Nb_3\ Si_{13.5}\ B_9$ aries from this freedom of tailoring the magnetic properties (Le Minh *et. al.* 1995; E. Estevez Rams *et. al.* 1996; Y. Yoshizawa *et. al.* 2005 and Sarout Noor 2005). At initial state of crystallization, saturation magnetization M_s has been reported (A. Lovas et. al. 2000 and A. E. Berkowitz *et. al.* 1981) to be increased with annealing temperature and then decreases correspond to the optimum nanocrystalline state with high volume fraction of Fe (Si) nanograins. In this study, we will focus on the nanostructure formation, crystallization behavior using differential thermal analysis method. The magnetization characteristics of two types of samples (sample A and sample B) will also be discussed in detail below and above the crystallization temperatures.

2. EXPERIMENTAL DETAILS

Generally, atomic deposition, melt-spinning technique, rapid quenching method and fast cooling melt techniques are used to produce metallic alloys in an amorphous state whose atomic arrangement have no long-range

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periodicity (J. M. D. Coey *et. al.* 1991 and K. Schnitzke *et. al*. 1990). In this work we have prepared nanocrystalline samples of $Fe_{75.5}Cu_1Nb_1Si_{13.5}B_9$ (sample-A) and $Fe_{74}Cu_{0.5}Nb_3Si_{13.5}B_9$ (sample-B) by rapid solidification method at the Centre for Materials Science, National University of Hanoi, Vietnam. In this technique, molten alloy is cooled in the range of melting temperature, T_m to the glass transition temperature, T_g very fast allowing no time for crystallization. The factors controlling T_g and crystallization are both structural and kinetic. Atomic arrangement, bonding and atomic size effect are related in the structural factors. The structural factors as discussed by Turnbull (1990) are the nucleation, crystal growth rate and diffusion rate compared to the cooling rate. DTA involves heating or cooling a test sample and inert reference under identical conditions, while recording any temperature difference between the sample and reference. The DTA technique was first suggest by Le Chatelier in 1887 and was applied to the study of clays and ceramics. DTA may be defined formally as a technique for recording the difference in temperature between a substance and a reference material against either time or temperature as the two specimens are subjected to identical temperature regimes in an environment either heated or cooled at a controlled rate. Vibrating sample magnetometer VSM is designed to measure the magnetic behaviours as a function of temperature and field. In this type of magnetometer, the sample is vibrated up and down in a region surrounded by several pickup coils. In this work, magnetization measurements have been performed by VSM at room temperature, applying a wide range of magnetic field.

3. RESULTS AND DISCUSSIONS

Calorimetric studies of amorphous alloys provide substantial information about the kinetics of crystallization and structural relaxation. Luborsky (1977) studied the kinetics of crystallization of a variety of amorphous magnetic alloys by means of calorimetric and magnetic techniques. In our present study, the experimental data have been interpreted in terms of different annealing effects on amorphous ribbons of DTA traces at constant heating rate 20°C/min. Figure-1 and Figure-2 show the DTA traces of sample-A and sample-B as cast and annealed at different temperatures for 30 min. Effect of annealing temperatures on the amorphous ribbons on their crystallization behaviors have been performed by DTA scan on both samples at a continuous heating rate 20°C/min.

According to the data presented on Figure-1, it was observed that the onset temperature for the sample-A as cast and annealed at T_a = 450°C is almost unchanged with respect to its amorphous precursor which is quite logical, since T_a is lower comparing to it's onset temperature of primary crystallization. However, when the same sample was annealed at T_a = 475°C, the primary crystallization peak has been found to be diminished to large extend and display quite diffused character which signifies that substantial amount of crystallization has been taken place. Similar kind of crystallization behavior has been noticed on sample-B as well which is displayed in Figure-2. Since the structure of the beneficial ferromagnetic nanocrystalline phase is comprised of Fe-Si, the study of primary and secondary crystallization temperatures are important for the amorphous alloys that are used as a precursor for the production of nanocrystalline FINEMET. DTA is a direct and effective technique in analyzing the kinetics of crystallization of amorphous materials. Two exothermic peaks are distinctly observed which correspond to two different crystallization events at temperatures T_{x1} and T_{x2} respectively for both samples. The soft magnetic properties correspond to the primary crystallization temperature, T_{x1} of α-Fe (Si) phase. On the other hand, secondary crystallization temperature T_{x1} corresponds to Fe-B phase which causes magnetic hardening of the nanocrystalline alloys. DTA analysis can not give us information about the phase. Phase identification of the samples was performed by XRD method. The results of DTA analysis of both samples are displayed in Table 1 and Table 2.

Table 1: Effect of annealing temperatures on the nanocrystalline amorphous ribbon with composition $Fe_{75.5}Cu_1Nb_1Si_{13.5}B_9$ at constant heating rate 20°C/min

Annealing Temperature	Primary crystallization temperature T_{x1} °C	Primary crystallization peak temperature T_{p1} °C	Secondary crystallization temperature T_{x2} °C	Secondary crystallization peak temperature T_{p2} °C
As-cast	470	482	558	569
450°C	468	480	557	569
475°C	-	-	554	566

Figures 3 and Figure 4 show magnetization measurements of sample-A and sample-B as-quenched and thermally treated samples. From the magnetization curves it is clearly observed that both samples are magnetically saturated in the amorphous and annealed states within an applied field of 2000Oe. At the same time it was also recorded that with the increase in annealing temperature magnetization increases until T_a = 525°C. Above this

temperature both samples were found to be saturated magnetically and then decreases with the raise in annealing temperature.

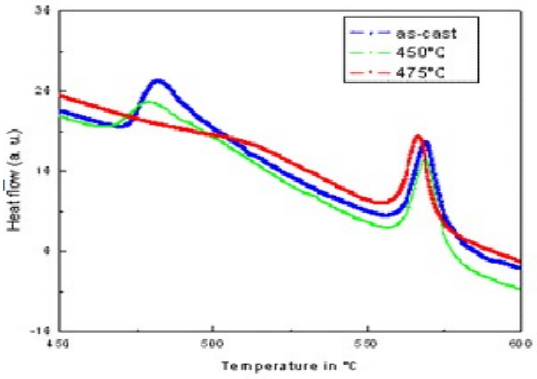

Figure 1: DTA analysis of sample-A at as-cast and different annealing temperature

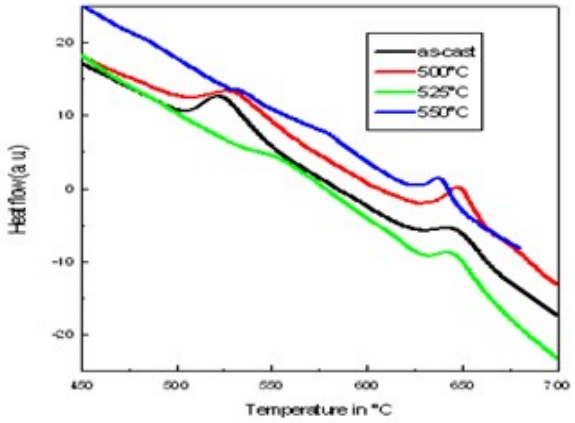

Figure 2: DTA analysis of sample-B at as-cast and different annealing temperature

Table 2: Effect of annealing temperatures on the nanocrystalline amorphous ribbon with composition $Fe_{74}Cu_{0.5}Nb_3Si_{13.5}B_9$ at constant heating rate 20°C/min

Annealing Temperature	Primary crystallization temperature T_{x1} °C	Primary crystallization peak temperature T_{p1} °C	Secondary crystallization temperature T_{x2} °C	Secondary crystallization peak temperature T_{p2} °C
As-cast	504	521	629	641
500°C	507	529	629	648
525°C	-	-	630	644
550°C	-	-	625	638

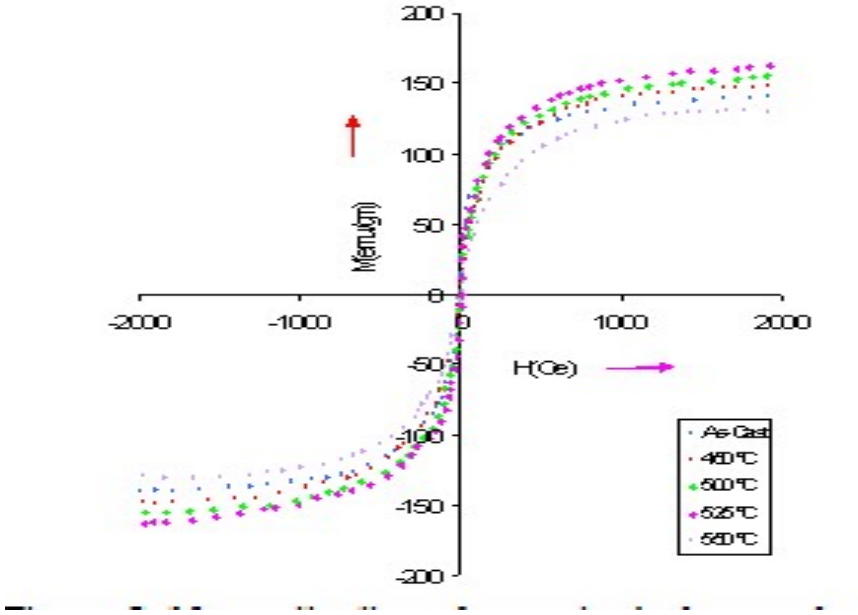

Figure 3: Magnetization of Sample-A at as-cast and different annealing temperature

Table 3 shows the variation of magnetization with annealing temperature for both samples. The increase in M_s might be related to the structural relaxation in Fe-based glasses (Berkowitz *et al.,* 1981). A rapid decrease in M_s above T_a = 525°C might be related to the fact that the enrichment of the residual amorphous phase with Nb weakens the coupling between ferromagnetic nanograins. In addition, the role of Si diffusion into Fe (Si) nanograins and local environments could also effect to decrease magnetization. The decrease in magnetization at

higher annealing temperature on ordering of Fe_3Si nanograin can not be ruled out. The same type of magnetization behaviors of nanocrystalline FINEMET materials has been reported by Aranda *et at.,* (1998).

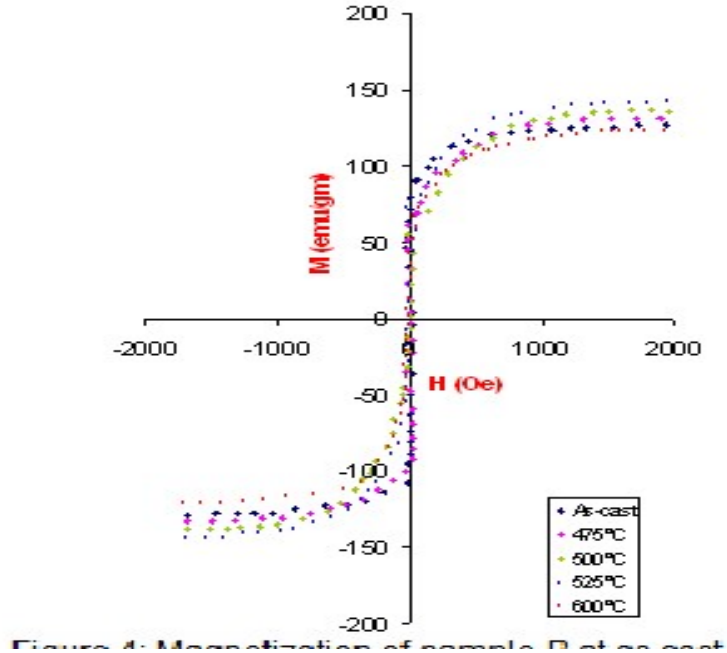

Figure 4: Magnetization of Sample-**B** at as-cast and different annealing temperature

Table 3: Magnetization data of sample-A and sample-B at various annealing temperatures annealed for 30 minutes

Samples	Annealing temperature, T_a in °C	Saturation magnetization, M_s in emu /gm
$Fe_{75.5}Cu_1Nb_1Si_{13.5}B_9$	As-cast	140
	450	148
	500	155
	525	163
	550	130
$Fe_{74}Cu_{0.5}Nb_3Si_{13.5}B_9$	As-cast	128
	475	133
	500	138
	525	141
	600	124

4. CONCLUSIONS

DTA analysis of the nanocrystalline samples reveals the fact that the primary and secondary crystallization temperatures with the manifestation of two well-defined exothermic peaks. In the case of nanocrystalline amorphous ribbons the knowledge of crystallization temperatures could be successfully used to control the magnetic properties. The primary crystallization temperatures have been found to be 470°C and 504°C for the sample-A and sample-B respectively which indicates that sample-B shows higher thermal stability against nanocrystal formations. This higher temperature is important for the stability of the primary crystallization phase Fe(Si) for the fabrication of high quality inductors. In sample-A the crystallization temperatures were found to be in between 450°C and 475°C while for sample-B in between 500°C and 525°C respectively. The saturation magnetization of nanocrystalline amorphous ribbons has slightly increased with the increase in annealing temperature around the crystallization. However, when the samples were annealed at higher temperature, at which crystallization takes place, magnetization found to be decreased. Therefore, further research could be carried out by changing composition and heat treatment in order to study the magnetic anisotropy, temperature dependence of magnetization and magnetostriction in detail for a better understanding of the characteristics.

ACKNOWLEDGEMENTS

Authors gratefully acknowledge the financial support of Dutch-Bangla Bank Foundation (DBBF), Bangladesh. DTA studies of the nanocrystalline amorphous ribbons were done in the laboratory of Industrial Physics Division (IPD), BCSIR, Dhaka, Bangladesh.

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