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STUDY OF THE TRANSPORT PROPERTIES OF NANOCRYSTALLINE SOFT MAGNETIC ALLOY $(Fe_{0.9}Co_{0.1})_{73.5}$-$Cu_1$-$Nb_3$-$Si_{13.5}$-$B_9$

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ABSTRACT

Nanocrystalline soft magnetic materials are interesting from both the fundamental and applied viewpoints. Because of various superior mechanical, magnetic and electrical properties, in comparison with those of the crystalline state, these alloys (metallic glasses) form a class of technologically important materials. In order to study the effect of Nb on the magnetotransport properties of the nanocrystalline soft magnetic material $(Fe_{0.9}Co_{0.1})_{73.5}$-$Cu_1$-$Nb_3$-$Si_{13.5}$-$B_9$, temperature and field dependent resistivity as well as magnetoresistance have been measured using conventional four-probe technique. This alloy exhibits metallic behavior in the high temperature range due to the formation of microcrystallites around the crystallization temperature T_x since more and more conduction electrons participating in the conduction mechanism. The value of magnetoresistance is found to vary (1~15%) at different temperatures (28°C~ 240°C). The complex permeability of this magnetic alloy has been measured for both as cast and annealed samples. Permeability measurement has been taken at low frequency (1 KHz-13 MHz). The permeability is found to be in the order of 10^4. The real part of the complex permeability is found to increase while the imaginary part is found to decrease with annealing temperatures due to local segregations. The best response is obtained for the sample annealed at 450°C for 5 minutes only. The overall permeability of the sample has low value when annealed at 450°C for 1 hour. The frequency range where the sample can be useful as a soft magnetic material is found out as 10 KHz to 1MHz. The magnetization of the sample has been measured as a function of magnetic field at room temperature (28°C) using a vibrating sample magnetometer (VSM). Saturation magnetization for the sample is observed to be around 179 emu/gm.

Keywords: alloy, nanocrystalline, complex permeability, finemet.

INTRODUCTION

Materials with microstructural features of nanometric dimensions are referred to as nanocrystalline materials. Nanocrystalline materials are single or multi-phase polycrystalline solids with a grain size of a few nanometers, typically less than 100nm. Since the grain sizes are so small, a significant volume of the microstructure in nanocrystalline materials is composed of interfaces, mainly grain boundaries i.e., a large volume fraction of the atoms resides in grain boundaries. Nanocrystalline soft magnetic materials are obtained by crystallizing amorphous ribbons of specific families of (Fe, B)-based alloy chemistries. As a result of research to date, two families of nanocrystalline alloys show the best performance characteristics: Finemet family and Nanoperm family. Typical system of Finement is Fe-Cu-Si-Nb-B, which is fabricated by rapid quenching technique such as melt spinning and annealing for nanocrystallization. Soft magnetic behaviour of such materials has been studied during the last twenty years [1, 2]. These materials consistes of ultrafine bcc Fe-Si[α-Fe-Si] grains of about 10nm in grain diameter, fcc Cu grains of several nm in grain diameter and residual amorphous phase. The excellent soft magnetic properties such as extremely low coercivities, high permeabilities, low energy losses etc. noted from these materials has triggered major interest and research activity in both the academic / research community and the industrial community. Their promising applications are in transformer cores for power frequencies, saturable reactors, high-frequency transformers and magnetic heads. Additional probable applications are for data communication interface components (pulse transformers), sensors (current transformers, magnetic direction sensors), common mode choke coils and magnetic shielding. It is predicted that in the future essentially all media and heads will contain nanostructured materials.

Continuing effort has been made to improve the soft magnetic properties of the Finemet type alloys by modifying the alloy compositions. New series of Finemet type alloys including Co, $(Fe_{0.9}Co_{0.1})_{73.5}$-$Cu_1$-$Nb_3$-$Si_{13.5}$-$B_9$ are now being developed for high frequency use. The addition of Co to these alloys can enhance the values of saturation magnetization B_s and permeability μ [3]. Muller *et al.* [4] reported that the substitution of Fe by Co decreases the saturation magnetostriction in Finemet type amorphous alloy. Adding Co is also expected to be beneficial to induce in-plane magnetic anisotropy to ribbons by magnetic field annealing, because the substitution of Fe with Co is known to effective for increasing the magnetic anisotropy constant K_u, of Fe-Si-B amorphoug alloys [5]. A large K_u can increase the magnetic resonance frequency (μ-f) property in a high frequency region. Also the addition of Co to Fe- based amorphous alloys influences the crystallization process significantly [6]. The speed of heat propagation through this sample is found to be quite high and shows metallic characteristics of the sample [7]. The new nanocrystalline material fills in the gap between amorphous metals and conventional poly-crystalline alloys. Transport properties of these materials are very interesting to those who are working with the commercial applications of these materials. Transport properties of the magnetic system depend on many factors like topological disorder of grains, mechanical stress developed during the growth process, local segregation due to temperature cycling around T_G *etc.*

The aim of the present work is to analyze the effects of the partial substitution of Fe by Co for a Fe-B-Si-Cu-Nb base alloy, on the transport properties and the ac permeability. The substitution of 3d-elements by ferro or anti-ferromagnetic elements could lead to a new magnetic phase [8]. The experimental power law which indicates the phase transition has been determined from the temperature variation of magnetization in the high temperature range.

METHODOLOGY

The magnetic alloy with the composition of $(Fe_{0.9}Co_{0.1})_{73.5}$-$Cu_1$-$Nb_3$-$Si_{13.5}$-$B_9$ has been prepared in the form of ribbon with a width of 5mm and a thickness of 25μm by the rapid quenching method. Magnetotransport properties of this nanocrystalline soft magnetic alloy like temperature and field dependent resistivity as well as magnetoresistance have been calculated using conventional 4-probe technique. The samples have been annealed at 3 different temperatures for studying the annealing effect on the permeability. The ac permeability has been measured on toroidal shaped samples using LF 4192A impedance analyzer. Temperature and field dependence of magnetization has been measured by 880 DMS vibrating sample magnetometer (VSM).

RESULTS AND DISCUSSIONS

Resistivity

In order to find the temperature dependence feature of resistivity, the normalized resistivity as a function of temperature is presented in Figure-1 for the high temperature region 25°C~240°C. Below 60°C variation of resistivity is found very small. From 60°C to 90°C the resistivity increases sharply with temperature and then rises slowly, which is likely to have originated from the electron-electron and thermally generated phonon-electron scattering. The temperature dependence of resistivity exhibits metallic behaviors in the high temperature region.

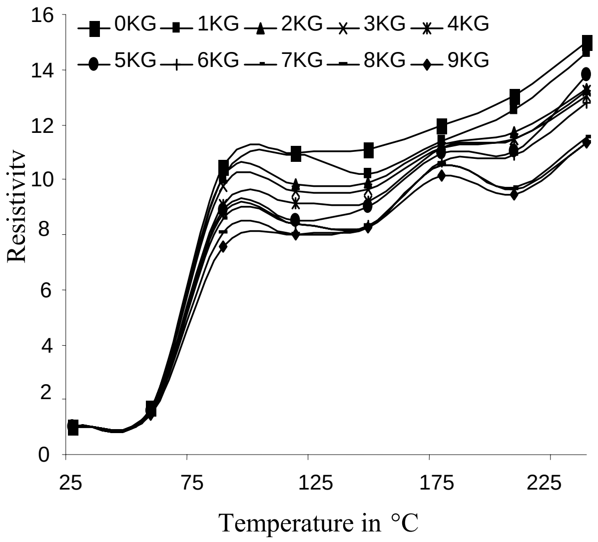

Fig.-1 Temperature dependent resistivity

To see the field dependence behaviors of resistivity, the normalized resistivity is also presented as a function of magnetic fields in Figure-2.

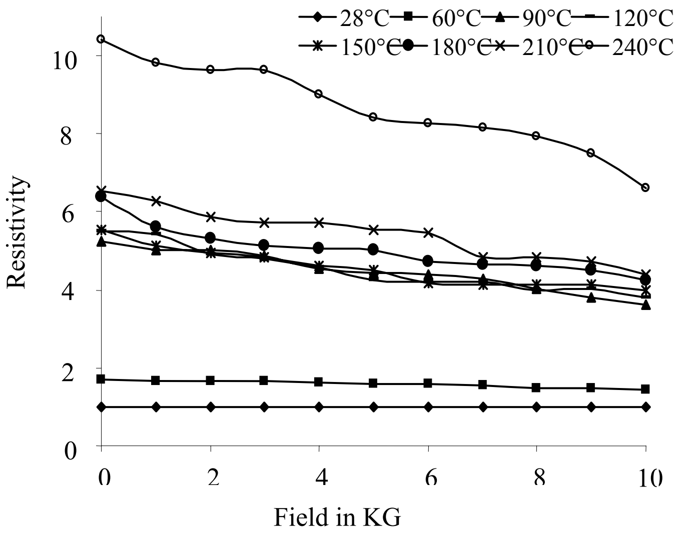

Fig.-2 Field dependent resistivity

The resistivity is found to have slightly decreased in the high temperature region. In the low temperature region, variation of resistivity is almost independent with field. The competition between the magnetic ordering and disordering process among the gains in the ferromagnetic amorphous matrix is expected to cause a slight decrease of resistivity and this competition is possibly originated from the enhanced paramagnetic effect of Nb by the thermal agitation in the high temperature region.

Magnetorsistance

Variation of magnetoresistace with temperature is depicted in Figure-3. Magnetorsesistance is found to have

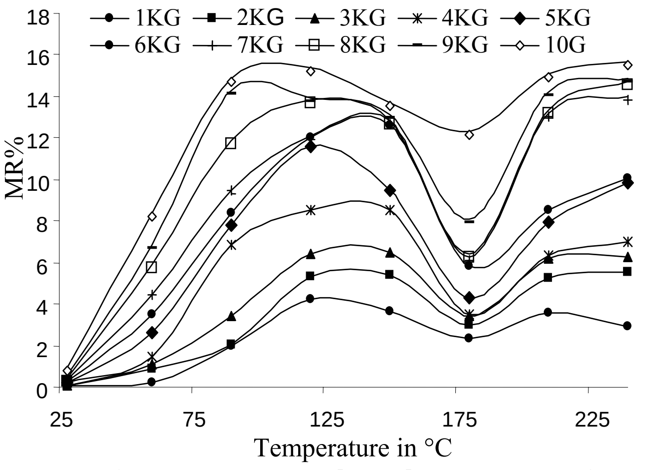

Fig.-3 Temperature dependent magnetoresistance

increased with temperature. The Magneto-resistance reaches a maximum value around the temperature 125^{0}C. From 125°C to 180°C the magnetoresistance is found to decrease and has the lowest value at 180°C. This nature of the curves may be attributed to a transition in the spin fluctuation; an increase in the magnetoresistance means an increase in the spin fluctuation in the glass phase. The spin fluctuation decrease as we increase temperature. A contrary behavior is also observed above 180°C. Above this temperature value of magnetorsesistance is again increased that is the spin fluctuation is increased at higher temperature when external magnetic fields have been applied on the sample.

Field dependence behaviors of magnetoresistance are shown in Figure-4. Magnetoresistance is found to have increased with applying field. It may be due to the magnetic disordering process and is expected to relatively weaker ferromagnetic coupling between ultrafine grains owing to the enhanced paramagnetic effect of Nb by the thermal agitation in the high temperature region.

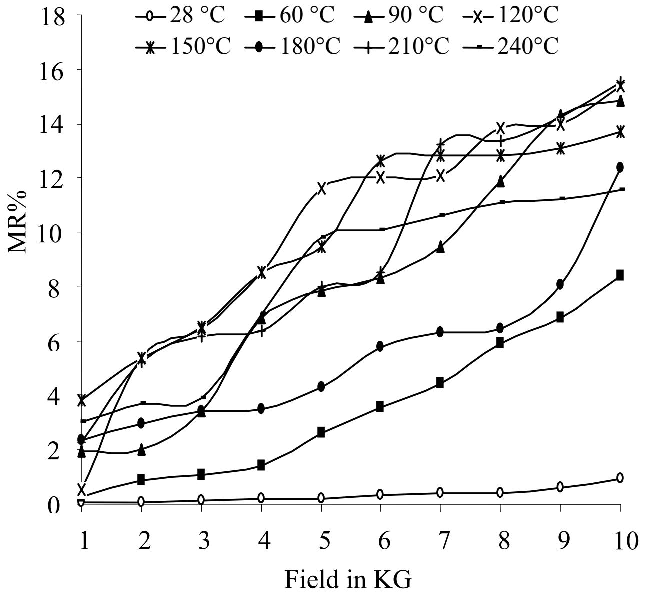

Fig.-4 Field dependent magnetoresistance

Complex permeability at low frequency

To discuss the frequency response feature, both the real and imaginary parts of complex permeability have been measured as a function of frequency in the range from 1 KHz to 13MHz. The measurement has been done on as cast specimen and also on the samples annealed at 200°C and 450°C for one hour and at 450°C for 5 minutes only. The real and imaginary parts of complex permeability for as cast and annealed samples have been presented separately as a function of frequency in Figures 5 and 6, respectively in logarithmic scale. Figure-5 representing the results that as cast specimen have high relative permeability of 24100 at 1 KHz. At 6 KHz there is a slight drop in permeability and after that the value of permeability is decreasing slowly and smoothly. For the sample annealed at 200°C for 1 hour it is found that the frequency dependent permeability becomes smoother remaining almost constant up to 8 KHz and then falling at a constant rate to a value of 7200 at 13 MHz. The overall permeability of the sample annealed at 450°C for 1 hour has a low value in the range of 100 KHz to 13MHz. The best response is obtained for the sample annealed at 450°C for 5 minutes only. The maximum permeability realized is 37730, which remains almost uniform up to 5KHz. Beyond this frequency, permeability falls more sharply and then the value becomes 9200 at 13 MHz. The frequency response of the samples annealed at different temperatures can in general be explained in terms of the growth of crystallites and their size distributions. With concentration of Nb = 3%, the grain size increases slowly with annealing temperature 450°C and annealing time 5 minutes only. The increase in permeability due to annealing at 450°C for 5 minutes indicates that this high temperature annealing removes the local defects as created during the preparation of the sample, which facilitates the domain wall movement.

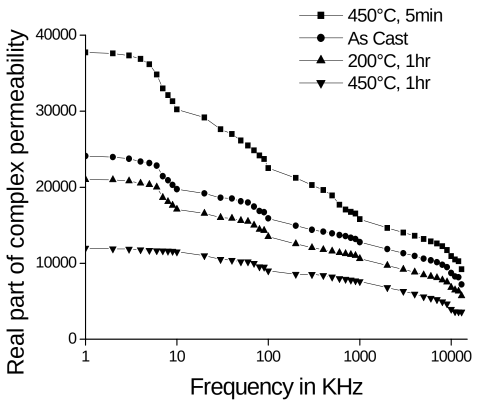

Fig-5. Frequency dependence of real part of complex permeability at different temperatures

The effect of heat treatment on the imaginary part of complex permeability and frequency dependence for the sample with composition of $(Fe_{0.9}Co_{0.1})_{73.5}$-$Cu_1$-$Nb_3$-$Si_{13.5}$-$B_9$ is shown in Figure-6.

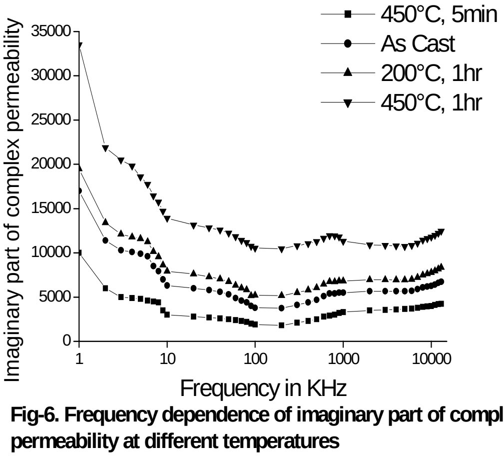

Fig-6. Frequency dependence of imaginary part of complex permeability at different temperatures

These results are quite complimentary to the results for the real part of the complex permeability of the samples. The imaginary parts of the complex permeability have shown a similar response from 1 KHz to 13MHz frequency for all the samples (as cast and annealed). However, the imaginary part has significantly decreased upon annealing the sample at 450°C for 5 min. This means that the ac permeability response has improved significantly after annealing the sample for 5 min. This enhancement in the permeability response may be attributed to the formation of nanograins after annealing the sample at 450°C. The imaginary part of the complex permeability for the sample annealed at 450°C for 1hr has increased than the sample annealed at 200°C for 1hr and than the as cast sample. Formations of nanograins resist and hence ac permeability becomes low due to anneal the sample for 1hr. The usefulness of the results of imaginary part of complex permeability lies in the determination of the quality factor of the sample. The imaginary part of complex permeability for all the samples at low frequencies has relatively high value and corresponds to high loss factor and lower quality factor as shown in Figures 7 and 8.

Relative quality factor

The frequency dependence of relative quality factor of the different annealed samples is shown in Figure-7.

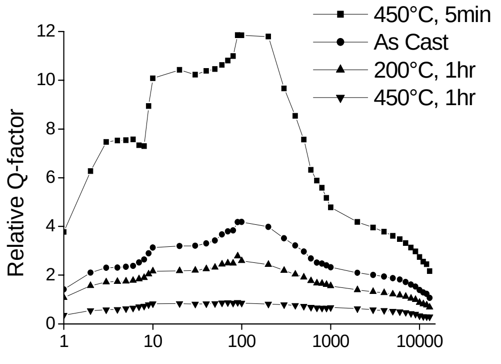

Fig-7. Relative Q-factor vs frequency curves for different annealing temperatures

The relative quality factors are controlled by the real part of the initial complex permeability have quite high value in the range 10 KHz to 1MHz. The best response is obtained for the sample annealed at 450°C for 5 minutes only. The quality factor of the sample has the height value at the frequency of 100 KHz. From the study of relative quality factor, perfect annealing temperature and frequency band can be identified at which the sample works as soft magnetic material with low loss. Thus, it is observed that 10 KHz-1MHz is the frequency range where the sample can be useful as a soft magnetic material.

Loss factor

The results for the loss factor of the samples are shown in Figure-8. The frequency range within which the loss factor has a reasonable low value is 10 KHz to 1MHz.

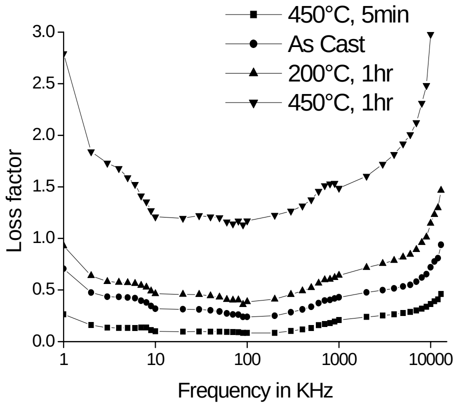

Fig-8. Loss factor vs frequency curves for different annealing temperatures

Loss factor has high value both in low frequency and high frequency. At low frequencies, the loss is controlled by hystersis losses and at high frequencies the flux penetration becomes low and loss is controlled mainly by interaction between the grains.

The origin of the loss factor can be attributed to various domain defects, which include non-uniform and non-repetitive domain wall motion, domain wall bowing, localized variation of the flux density and nucleation and annihilation of domain walls.

Magnetization

Magnetization of the nanocrystalline material with composition of $(Fe_{0.9}Co_{0.1})_{73.5}$-$Cu_1$-$Nb_3$-$Si_{13.5}$-$B_9$ has been measured as a function of magnetic field using a vibrating sample magnetometer (VSM). The magnetometer has been used as field measuring device. The proportionality constant accounting for the particular coil geometry and susceptibility is obtained by calibration with a high purity circular disk shaped nickel (Ni) sample. The sample has calibration constant of about 2.3 x10^{-3}A.m^2 with a saturation voltage of about 1.99V.
Magnetization of the as prepared sample has been studied as a function of magnetic field at room temperature (28°C) and is shown in Figure-9. Saturation magnetization for the sample is observed to be around 179 emu/gm. The saturation field for the sample is around 1.98 Kilogauss

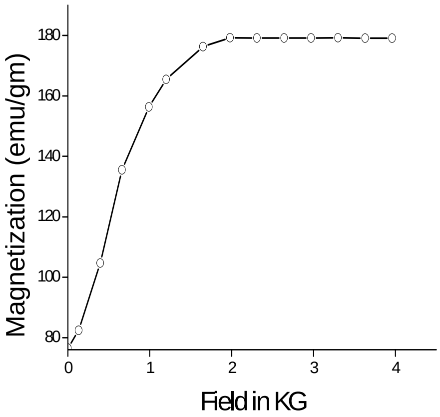

Fig-9 Field dependent magnetization

(KG). The experimental data shows that the magnetization is almost field independent above 1.98 KG.

Temperature vs. magnetization curve for as cast sample at different temperatures is shown in Figure-10. The characteristic feature of the curve is that magnetization decreases with temperature and passes through ferro-paramagnetic transition at the Curie temperature (T_c).

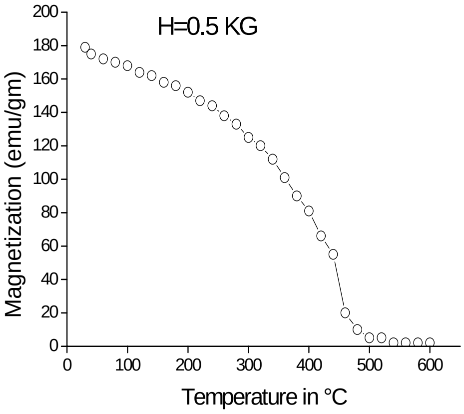

Fig-10 Temperature dependence of magnetization

The value of T_c is estimated to be around 530°C which is equivalent to the result that was found out from the anisotropy vs. temperature curve in the previous paper [7]. However, the determination of the Curie temperature is a bit uncertain because these materials are not single-phase materials. At high temperatures there is segregation of iron and cobalt and Fe-Co alloy crystallines, which have different Curie temperatures. Moreover, Curie temperature of this ribbon is lower than the ribbon containing higher percentage of iron. This is quite understandable from the consideration of higher contribution of magnetic moments in iron-rich ribbons. Addition of cobalt by a small fraction i.e. replacing iron by 7% of cobalt decreases the magnetic moment significantly.

CONCLUSIONS

The Nanocrystalline alloy with high quality soft magnetic properties have been prepared by annealing amorphous ribbons. The material has some features common with the amorphous such as magnetic softness, high electrical resistivity and low dimensionality. However, there are special advantages of the material in respect of high permeability, high Curie temperature and high operational temperature. The highest permeability for the sample is found out as 3.7 x10^4 and the maximum frequency for which the sample can be used as soft magnetic material being closed to 1.0MHz. The frequency response has been controlled by heat treatment, which in turn controlled the grain size distribution. The saturation magnetization for the sample has higher value for higher percentage of iron. There is much scope for further research on this material. The magnetic permeability, saturation magnetization, Curie temperature, magnetic anisotropy, magnetostriction and coercivity are all controllable by choosing appropriate composition, altering preparation technique, selecting the temperature and duration of the heat treatment.

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