Phase transition and permeability of Fe$_{73.5-x}$Cr$_x$Cu$_1$Nb$_3$Si$_{13.5}$B$_9$ alloys.

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Abstract
Magnetic phase transition and initial permeability upon nanocrystallization has been studied by controlled annealing of Fe$_{73.5-x}$Cr$_x$Cu$_1$Nb$_3$Si$_{13.5}$B$_9$ alloys with $x = 0, 1, 2, 3, 4, 5, \text{ and } 8$. Ribbons of 10 mm width and 30 $\mu$m thickness were prepared by melt-spinning technique. The amorphosity of the ribbons was checked by x-ray diffraction. Ferro-paramagnetic phase transition temperature, $T_C$ has been determined for all the alloys from temperature dependence of initial permeability with a heating rate of 5°C / min. A linear decrease of $T_C$ by about 25°C per at% Cr content is attributed to the reduction in exchange interaction between Fe atoms due to the substitution of Fe by increasing amount of non-magnetic Cr in the alloys. Similarly a dilution of magnetic moment with increasing Cr content substituted for Fe has been observed from composition dependence of saturation magnetization. Magnetic softness characterized by magnetic initial permeability, $\mu'$ increases with the increase of annealing temperature for all Cr content. Comparatively higher values of $\mu'$ corresponding to higher Cr contents are due to their low Curie temperature resulting from the decrease of anisotropy energy at room temperature. Again lower value of $\mu'$ measured at room temperature for the samples with higher Cr content annealed at higher temperature is possibly due to the decrease of $T_C$ of the residual amorphous phase below the room temperature as a result of which decoupling of the inter-granular exchange interaction takes place.

Keyword : Magnetization, Curie Temperature, Initial permeability, Amorphous, Nanocrystalline.

1 Introduction
The development of new materials and their understanding on a nanoscale is at the root of recent progress in many areas of material science [1] and in particular this is true in the development of new magnetic materials. It is well known that the properties of the isolated grains change drastically as their size is reduced to the nanometer range when such nanometric grains are consolidated to form a nanostructured material, the resulting macroscopic magnetic properties are largely determined by the grain size and the exchange interaction between the adjacent grains.

A new class of nanocrystalline materials called FINEMET with composition Fe$_{73.5}$Nb$_3$Cu$_1$Si$_{13.5}$B$_9$ developed by Yoshizawa et. al. [2] in 1988 obtained by devitrification of the Fe-based metallic glasses is characterized by a homogeneous ultrafine grain structure of bcc-FeSi nanocrystallite of around 10 nm and random texture, embedded in an amorphous matrix in its optimally annealed condition. The superior soft magnetic properties of this material are due to its extremely small effective anisotropy and simultaneously low effective saturation magnetostriiction. The theoretical understanding of the nanometric grain with magnetic softness has been developed in the light of Random Anisotropy Model (RAM) proposed by Alben et. al. [3]. Herzer, on the basis of RAM showed that extraordinary soft magnetic properties of the nanocrystalline materials are due to the suppression of effective magnetocrystalline anisotropy ($<K> = 0$) when the grain size (D) is smaller.
than ferromagnetic exchange length ($L_{ex}$), vanishing magnetostriction ($<\lambda> = 0$) which is revealed in these material after heat treatment [4] and strong intergranular magnetic coupling.

The effect of partial substitution of Fe by Co [5], Al [6], Cr-Mo[7] and Cr[8] has been investigated. Replacement of Fe by Cr or Mo enhances the thermal stability against crystallization and lowers the Curie temperature of the amorphous phase. This gives the opportunity of studying the magnetic interaction in a wider temperature range between the Curie temperature of the amorphous phase and that of nanocrystalline phase.

2 Experimental
Ribbons of composition Fe$_{73.5}$xCr$_x$Cu$_1$Nb$_3$Si$_{13.5}$B$_9$ alloys with $x = 0, 1, 2, 3, 4, 5,$ and 8 have been prepared by rapid solidification technique with a dimension of 10 mm width and 30 µm thickness. The purity of the material is Fe (99.98%), Cr (99+%), Cu (99+%), Nb (99.8%), B (99.5%) and Si (99.9%) as obtained from Johnson Mathey (Alfa Aesar Inc.). Amorphosity of the ribbon and nanocrystalline structure has been studied by Bruker D5005 x-ray diffractometer, Germany with Cu K$_\alpha$ radiation. Crystallization behavior has been performed using 2960 SDT Differential Scanning Calorimetry USA. Saturation magnetization has been measured using 880 DMS Vibrating Sample Magnetometer, USA. Curie temperature has been from temperature dependence of initial permeability using laboratory built furnace and Wayne Kerr 3255 B Impedance Analyzer.

3 Results and discussions
Amorphous state of the as-received sample has been checked by x-ray diffraction. X-ray diffraction studies have been performed for the samples isothermally annealed at 540°C for 30 minutes at which crystallization has been completed. Annealed samples clearly demonstrate the crystalline phase and has been identified as solid solution of bcc Fe(Si). Grain size determined by using Scherrer’s formula for the fundamental peak of (110) has been presented in Fig.1. Grain size decreases with increasing Cr content. It has been observed in our experiment that the thermal stability of the amorphous ribbons against crystallization is slightly enhanced due to increasing substitution of Fe by Cr, which will be discussed later.

![Graph](Image1.png)

Fig. 1. Composition dependence of grain size of Fe(Si) nanograins for Fe$_{73.5}$xCr$_x$Cu$_1$Nb$_3$Si$_{13.5}$B$_9$ developed during annealing at 540°C for 30 min.

![Graph](Image2.png)

Fig. 2. DSC traces of Fe$_{73.5}$xCr$_x$Cu$_1$Nb$_3$Si$_{13.5}$B$_9$ in the as-cast condition for different values of $x$ with the heating rate of 20°C/ min.
Fig. 2 shows the DSC traces of Fe$_{73.5-x}$Cr$_x$Cu$_1$Nb$_3$Si$_{13.5}$B$_9$ alloys with $x = 0, 1, 2, 3, 4, 5$. It is evident from the thermogram that the first crystallization temperature corresponding to the Fe(Si) phase shifts to higher temperature as the Cr content is increased. Similar trend is also observed for the second crystallization peak corresponding to the Fe-B phase. This clearly exhibits the enhancement of thermal stability of amorphous state of the materials against crystallization with the increase of Cr content.

Composition dependence of saturation magnetization, $M_s$, of Fe$_{73.5-x}$Cr$_x$Cu$_1$Nb$_3$Si$_{13.5}$B$_9$ has been presented in Fig. 3. It is observed that the saturation magnetization decreases gradually with the increase of Cr content. The decrease of $M_s$ with Cr content is attributed to the dilution of the magnetic moment of Fe atom by nonmagnetic Cr. Composition dependence of Curie temperature has been presented in Fig. 4. The Curie temperature $T_C$, which determines the strength of the exchange interaction between the magnetic atoms is found to decrease linearly with the increase of Cr content having a value of 350°C for $x = 0$ and 110°C for $x = 8$. A linear decrease of $T_C$ about 25°C per at. % Cr is attributed to the reduction in exchange interaction between Fe atoms due to the substitution of Fe by increasing amount of nonmagnetic Cr in the alloys.

Fig. 3. Composition dependence of saturation magnetization, $M_s$, of Fe$_{73.5-x}$Cr$_x$Cu$_1$Nb$_3$Si$_{13.5}$B$_9$ in the as-cast condition.

Fig. 4. Composition dependence of Curie temperature, $T_C$, of Fe$_{73.5-x}$Cr$_x$Cu$_1$Nb$_3$Si$_{13.5}$B$_9$ in the as-cast condition.
Composition dependence of optimized annealing temperature, maximum value of initial permeability, \( \mu' \), minimum value of relative loss factor, \( \tan \delta / \mu' \) has been presented in Table 1 for all the compositions. Optimized annealing temperature shifts towards lower temperature with the increase of Cr content. Maximum value of initial permeability is two orders of magnitude greater than the initial permeability in the as-gas cast condition while the minimum value of relative loss factor is of the order of \( 10^5 \). In Table 2, initial permeability of as-cast and annealed samples at different annealing temperatures measured at driving frequency of 10 kHz has been presented. It is observed that the permeability for the samples annealed at particular temperature decreases when it exceeds the optimized annealing temperature. This is possibly due to the decrease of T\(_C\) of the residual amorphous phase below the room temperature as a result of which decoupling of the inter-granular exchange interaction takes place.

**Table 2. Magnetic permeability, \( \mu \), with Cr content (x at.%) at different annealing temperatures**

<table>
<thead>
<tr>
<th>x</th>
<th>Initial permeability, ( \mu' ) at 10 kHz</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>258 6919 9075 14072 9080</td>
</tr>
<tr>
<td>1</td>
<td>270 4024 6521 12600 4744</td>
</tr>
<tr>
<td>2</td>
<td>273 3651 6347 11075 10548</td>
</tr>
<tr>
<td>3</td>
<td>337 3384 9230 17848 8746</td>
</tr>
<tr>
<td>4</td>
<td>310 5557 7823 11356 2355</td>
</tr>
<tr>
<td>5</td>
<td>258 5740 8351 4046 506</td>
</tr>
<tr>
<td>6</td>
<td>270 4282 4317 99 92</td>
</tr>
</tbody>
</table>

A linear decrease of T\(_C\) by about 25°C per at% Cr content is observed Fe is substituted by increasing amount of non-magnetic Cr in the alloys. Similarly a dilution of magnetic moment with increasing Cr content substituted for Fe has been observed from composition dependence of saturation magnetization. Magnetic softness characterized by magnetic initial permeability, \( \mu' \), increases with the increase of annealing temperature for all Cr content.
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References