Temperature Dependence Magnetoresistance of Fe\textsubscript{73.5} –Cu\textsubscript{1} –Ta\textsubscript{3} –Si\textsubscript{13.5} –B\textsubscript{9} Magnetic Alloy

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Abstract: Fe\textsubscript{73.5}–Cu\textsubscript{1}–Ta\textsubscript{3}–Si\textsubscript{13.5}–B\textsubscript{9} magnetic alloy, in the form of ribbon, was synthesized by single roller melt-spinning technique in air. The magneto transport properties of the sample were studied as a function of temperature using conventional 4-probe technique. The sample exhibits semiconducting behavior at low temperature (35 – 295 K) and metallic behavior at high temperature (350 – 700 K). The magnetoresistance (MR) found to increase with both the temperature and field in the low temperature range due to the ordering effect of microcrystallites in the amorphous matrix. A significant dispersion of negative MR obtained at 700 K due to more disorderliness of spins in the amorphous matrix of the sample. Its Zero Field Cool (ZFC) and field cool (FC) measurement demonstrates both the positive and negative slopes of Magnetoresistance below and above a critical transition temperature, 175 K that resulted from the difference of magnetic and thermal energies. Up to this critical temperature, the system makes a transition from ferromagnetic (FM) to antiferromagnetic(AFM) and above this temperature from AFM to FM, which is an indicative of reentrant ferromagnetic phase, which leads to be used it in sensors and magnetic devices.

Keywords: Magnetic alloy, Normalized resistivity, Magnetoresistance, Microcrystallites

1. Introduction

The Fe-Si-B system exhibits superior soft magnetic properties and thus is very useful in magnetic devices such as EMI filters, current sensors, magnetic sensors, transformer core and magnetic shielding sheets as reported in various literatures. The magnetic alloy with composition of Fe\textsubscript{73.5}–Cu\textsubscript{1}–Nb\textsubscript{3}–Si\textsubscript{13.5}–B\textsubscript{9} was first developed in the form of ribbon by Yoshizawa [1]. It crystallizes in the form of ultrafine grains of α-Fe (Si) embedded in the remaining ferromagnetic amorphous matrix.

Magnetoresistance is a magnetotransport property and guiding factor for determining soft magnetic behavior of this class of magnetic alloys. It is phenomenologically known as magnetostriction, which refers

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to the change of resistance with the applied magnetic field. Its value for Fe$_{73.5}$–Cu$_1$–Ta$_3$–Si$_{13.5}$–B$_9$ magnetic alloy found to be nearly zero. This was resulted from the competition between negative magnetostriction of $\alpha$-Fe (Si) crystallites and positive magnetostriction of remaining amorphous matrix as reported in 1999 [2]. This magnetic alloy system gives rise to outstanding magnetic softness and thereby commercially fabricated under the trade name of FINEMET as reported in 1999 [3].

Fe$_{73.5}$–Cu$_1$–Ta$_3$–Si$_{13.5}$–B$_9$ is a similar magnetic alloy, prepared by rapid quenching technique in the form of ribbon, where Niobium ($Nb$) was substituted by Tantalum ($Ta$). Many investigations thus far have been performed of this magnetic alloy but significant temperature response on its magnetotransport properties are yet to be reported. As such, this paper intends to apprehend a comprehensive study on temperature dependent magnetoresistance of Fe$_{73.5}$–Cu$_1$–Ta$_3$–Si$_{13.5}$–B$_9$ magnetic alloy and effect of tantalum ($Ta$) there on.

2. Experimental

The ribbon samples with nominal composition of Fe$_{73.5}$–Cu$_1$–Ta$_3$–Si$_{13.5}$–B$_9$ were prepared by the single roller melt-spinning technique in air. The ribbons were on an average 6 mm wide and (20 – 25) $\mu$m thick. The room temperature amorphosity of the sample was verified by JDX-8P GEOL X-ray diffractometer. Temperature dependence of resistivity and magnetoresistance were calculated by resistance values obtained from current – voltage (I–V) measurement using conventional 4-probe technique. The temperatures 300 K for room temperature, (35 ~ 295) K for low temperature and (350~700) K for high temperature ranges have been taken in temperature dependent measurements. The resistances from I–V measurements have been evaluated from the slope after fitting the straight (trend) line to I–V curve. The resistivity has been calculated using formula, $\rho = R \frac{A}{l}$, where $A$ is cross-sectional area and $l$ is effective length or spacing between inner two probes. For measurements of magnetoresistance, field range has been selected starting from 1 to 4 kilo gauss (KG) and the same has been calculated using the formula, 

$$MR\% = \left(\frac{\rho_H}{\rho_0} - 1\right) \times 100$$

where $\rho_H$ is the resistivity with magnetic fields and $\rho_0$ without magnetic field. The same 4-probe technique has been used for zero field cool (ZFC) and field cool (FC) measurements in the temperature range (40 ~ 265 K), In this case, the corresponding voltages across the inner probes have been measured as a function of temperature for a constant current of 150 mA, passed through the outer probes. For ZFC, the magnetic field was kept at 0 KG and for FC, it was at 5 KG.
3. Result and Discussion

XRD

Fig. 1 illustrates the X-ray diffraction pattern. Two broad peaks found to be within scanning angular range of 5° and 65°, which conforms its bi-phase nature. It is notable here that the sample used for amorphousity verification was annealed at temperature 623K.

![X-ray diffraction pattern of magnetic alloy Fe$_{73.5}$–Cu$_1$–Ta$_3$–Si$_{13.5}$–B$_9$]

Normalized Resistivity

The low (35 – 295 K) and high (300 – 700 K) temperature dependence of normalized resistivity (NR) are shown in Fig. 2(a) and 2(b) respectively. It is observed from Fig. 2(a) that NR decreases with temperature for low temperature range, which exhibits the semiconducting behavior of the sample. This is due to presence of the more ordered phase of α-Fe (Si) crystallites in the amorphous matrix. On the other hand, NR is found to increase with temperature for the high temperature range and demonstrates its metallic behavior as shown in figure 2(b). This is expected to originate from the electron-electron and thermally generated, phonon-electron scatterings.

Figure 3 depicts the field dependence of NR for both the low and high temperature ranges. NR is found to decrease with magnetic field for the low temperature range (35 – 295 K) as shown in figure 3(a). This is expected to cause from magnetic ordering between ferromagnetic microcrystallites for the applied magnetic field. NR also found to slight decrease with applied magnetic field in the high temperature range (350 – 700 K).
K) as shown in figure 3(b). This is caused by the competition of magnetic ordering and disordering of the grains in the amorphous matrix. The competition is expected to originate from the enhanced paramagnetic effect of Ta due to thermal agitation.

**Figure. 2:** Temperature dependence of NR (a) the low temperature range (35 – 295 K) and (b) the high temperature range (300 – 700K)

**Figure. 3:** Field dependence of NR (a) the temperature range 35~ 295K and (b) the high temperature range (350 – 700 K)

**Magnetoresistance**

Figure. 4(a) illustrates the field dependence of MR for the low temperature range (35 – 295 K.). MR is found to increase with the applied magnetic field in this low temperature range. MR is also observed to increase with temperature, which is realized from its temperature dependence as depicted in figure. 4 (b). The increase of MR with both the temperature and field in the low temperature range is expected to generate from the ordering effect of microcrystallites in the amorphous matrix.

Figure. 5 (a) represents the field dependence of MR for the high temperature range (350 – 700 K). The negative MR is found to increase with the applied magnetic field. This is also observed to increase with temperature as evident from the temperature dependence of MR as depicted in figure. 5(b). The negative MR
is associated with the transport by magnetic polarons in the amorphous matrix. The increase of negative MR with both the magnetic field and temperature is caused by magnetic disordering between ultrafine grains in the imperfectly ordered ferromagnetic matrix. This imperfect order in the matrix is expected to generate from the enhanced paramagnetic effect of Ta by thermal agitation.

Despite, a significant dispersion of negative MR is obtained at 700 K for the applied magnetic field of 4 KG as shown in the figure. 5(a). This dispersion is expected to originate from the more disordered spins in the amorphous matrix. This disorderliness of spins is likely to cause ferromagnetic (FM) to paramagnetic (PM) phase transition at 700 K due to enhanced paramagnetic effect of Ta by thermal agitations.

![Figure 4](image)

**Figure 4**: (a) Field dependence of MR and (b) for temperature dependence of MR for the temperature range 35~ 295 K.

![Figure 5](image)

**Figure 5**: (a) Field dependence of MR and (b) for temperature dependence of MR for the temperature range 350~ 700 K.

**Thermal Effect for Zero Field Cool (ZFC) and Field Cool (FC) Magnetoresistance**

Figure. 6(a) presents the ZFC and FC magnetoresistance graph, where magnetoresistance exhibits both the positive and negative slopes below and above a critical transition temperature, 175 K that resulted from the difference of magnetic and thermal energies as evident from a sort of thermal hysteresis at the
temperature range (120 to 240 K) as shown in figure 6(b). Up to this critical temperature, the system makes a transition from FM to AFM and above this temperature from AFM to FM, which is an indicative of reentrant ferromagnetic phase. The negative slope of magnetoresistance corresponds to crystalline phase in \( \alpha - Fe(Si) \) crystallites, which is due to strong magnetic field that brings more regularity in the spin arrangement and reduces the scattering of \( 4s \) conduction electrons. On the other hand, the positive slope of magnetoresistance occurred in the remaining amorphous matrix owing to high content of \( B \) and \( Ta \) due to reduction of the spin wave stiffness constant as found in case of magnetostriction in FINEMET as reported in 2008 [4]. The balance struck between negative and positive slopes of magnetoresistance resulted from their competition, which provides very low or almost zero magnetostriction to this magnetic alloy. So, the sample under investigation exhibits excellent soft magnetic properties that can be used in many sensors and magnetic devices.

4. Conclusion

Temperature dependence resistivity exhibits semiconducting behavior at low temperature range and metallic behavior at high temperature range. Its semiconducting behavior is due to presence of the more ordered phase of \( \alpha - Fe (Si) \) crystallites in the ferromagnetic amorphous matrix, and metallic behavior is expected to originate from the electron-electron and thermally generated, phonon-electron scatterings. The increase of MR with both the temperature and field at the low temperature range (35 – 295 K) is expected to generate from the ordering effect of microcrystallites in the amorphous matrix. The negative MR at the high temperature range (300 – 700K) is associated with the transport by magnetic polarons in imperfectly ordered ferromagnetic matrix. This imperfect order in the matrix is expected to generate from the enhanced paramagnetic effect of \( Ta \) by thermal agitation. Beside, a significant dispersion of negative MR at temperature 700 K is expected to originate from the more disordered spins in the amorphous matrix, which
likely to cause ferromagnetic (FM) to paramagnetic (PM) phase transition due to enhanced paramagnetic effect of Ta by thermal agitations.

From ZFC and FC measurements, both the positive and negative slopes of magnetoresistance found to be at a critical transition temperature, 175 K that resulted from the difference of magnetic and thermal energies. This is an indicative of reentrant ferromagnetic phase. The negative slope in $\alpha - Fe(Si)$ crystallites is originated from more regularity of the spin arrangement and the positive slope of the same in remaining amorphous matrix is developed from reduction of spin wave stiffness constant due to high content of B and Ta. The balance struck between negative and positive slopes of magnetoresistance resulted from their competition provides very low or almost zero magnetostriction to this magnetic alloy. So, the sample under investigation exhibits excellent soft magnetic properties that can be used in many sensors and magnetic devices.

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References