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The Effect of Antimony Substitution on the Magnetic and Structural Properties of $Fe_{0.75-x}Si_{0.25}Sb_x$ Alloys

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ABSTRACT

The results of the investigation of the magnetic and structural properties of the alloy system $Fe_{0.75-x}Si_{0.25}Sb_x$, where $x = 0$, 0.05, 0.1, 0.15, 0.2, and 0.25 synthesized by mechanical alloying followed by heat treatment are described. The *x*-ray diffraction reveals that all samples crystallize in the DO_3-type cubic phase structure. Substituting Fe by Sb led to a decrease in the lattice constant and the unit cell volume. The magnetic properties are investigated by vibrating sample magnetometer and show that all the samples are ferromagnetically ordered at room temperature. The Curie temperature is found to decrease linearly from (850 ± 5) K for the parent alloy to (620 ± 5) K for the alloyith $x = 0.25$. The saturation magnetizations at room temperature and at 100 K are found to decrease with increasing the antimony concentration. The above results indicate that Sb dissolves in the cubic structure of this alloy system.

Keywords: Lattice Constant; Curie Temperature; Saturation Magnetization

1. Introduction

Intermetallic compounds Fe_3X (X = Al, Si, Ge) are fascinating group of materials, which have recently attracted much attention from the viewpoint of fundamentals as well as application. This class of materials with the ordered phase at elevated temperatures (T > 800 K) has promising mechanical and corrosion properties both in the microcrystalline and nanocrystalline states [1-4]. Ternary element additions to these systems have been used to improve the high temperature strength and other physical properties. At present, numerous experimental data are available on the ordered and disordered Fe_3Si and Fe_3Al alloy systems, because of their high temperature strength, excellent oxidation and corrosion resistance. Substitution of Fe by a transition metal element affects the magnetic properties, the lattice parameter, and the structural ordering of these alloys [5-12]. Waliszewski *et al.* [12] studied the $Fe_{3-x}Cr_xSi$ system and found that the Curie temperature decreases from 840 K for $x = 0$ to 712 K for $x = 0.4$, and the magnetic moment of iron at the B site to be 2.44 μ_B, and about 1.18 μ_B for (A, C) sites, while the magnetic moments of Cr were determined to be (2.03 ± 1.3) μ_B for Cr at the B site and (0.41 ± 0.63) μ_B for the (A, C) sites with orientation antiparallel to the magnetic moments of Fe.

The structural and magnetic properties of the intermetallic Fe-Si alloys depend on the Si concentration. $FeSi_2$ was found to form the tetragonal type structure, FeSi forms the B20 type structure, while Fe_3Si forms the DO_3 type cubic structure. Fe_3Si is a well ordered ferromagnetic alloy with four sites: 2-equivanlt sites (A, C) occupied by Fe and the other two sites are occupied by Fe (B site) and Si (D site) [13]. $Fe_{1-x}Si_x$ forms a continuous rage of solid solution with a bcc structure between $x = 0$ and 0.265 [14]. Mössbauer and NMR [15,16] investigations for this system have yielded information about the hyperfine field and Si site occupation. These studies showed that the hyperfine field and the magnetic moment at the Fe sites strongly depend on the Si occupancy and the number of Fe nearest neighbors and they decrease with increasing the Si concentration.

A recent study by Mantovan *et al.* [4] showed that Fe_3Si thin film is a good candidate as ferromagnetic electrode in spintronics devices due to its high spin polarization and Curie temperature. Furthermore, the use of Fe_3Si has been proposed for spin injection in semiconductors [17]. Antimony is widely used in alloying to increase hardness and mechanical strength. Many of Antimony compounds are also used in superconductors, Hall-effect devices, diodes, batteries cable sheathing, tartar emetic which has been used in medicine. Prakash *et al.* [18] studied the effect of antimony doping on the transport and magnetic properties of $CeO_{0.9}F_{0.1}FeAs_{1-x}Sb_x$ Super-

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conductors and found that Sb increases the superconducting transition temperature (T_c) from 26 K for the parent compound with $x = 0$ to 43.17 K for $x = 0.1$. Antimony is the most effective additive to inhibit diffusional growth of particles. Dixon *et al.* [19] have shown that the addition of antimony to iron-in-mercury magnetic liquids forms a layer around the iron and reduces the growth of the nanoparticles which reduces the magnetization. The effect of antimony doping on magnetic properties of Ni-Zn ferrites was studied by Purnanandam *et al.* [20]. This study confirmed that the magnetic quality factor increases with increasing Sb percentage while the saturation magnetization and the Curie temperature decrease with increasing antimony doping in Ni-Zn ferrites.

In this paper we focused our attention on the synthesizes of the $Fe_{0.75-x}Si_{0.25}Sb_x$ alloy system by mechanical alloying followed by heat treatment for six different samples which allows us to investigate the effects of substituting ferromagnetic iron be diamagnetic antimony on the structural and magnetic properties of the $Fe_{0.75-x}Si_{0.25}Sb_x$ alloy system.

2. Samples Preparation and Experimental Procedures

$Fe_{0.75-x}Si_{0.25}Sb_x$ alloys (x = 0, 0.05, 0.10, 0.15, 0.2 and 0.25) were synthesized by mechanical alloying of high purity elemental Fe, Si, and Sb in a SPEX 8000 mixer/mill for 15 hours using tungsten-carbide (WC) balls and WC vial. The ball to powder mass ratio was 3:1, and the milling was carried out under argon atmosphere. After the milling, the powder samples were annealed under vacuum in quartz ampoules for 24 hours at 800˚C, to ensure homogeneity. The ampoules were then rapidly quenched into cold water. Subsequent the samples were characterized by powder x-ray diffraction on a Philips diffractometer by using Cu-K_α radiation. The magnetic measurements were carried out in the temperature range 100 K ≤ T ≤ 975 K, using a Vibrating Sample Magnetometer (VSM) in magnetic fields up to 13.5 kOe.

3. Results and Discussion

Figure 1 shows the powder x-ray diffraction patterns for two representative samples of the alloy system $Fe_{0.75-x}Si_{0.25}Sb_x$ with x = 0.00, and 0.15. All the observed reflection lines obtained at room temperature for all investigated samples exhibit lines characteristic for the DO_3-type structure. The indices for the various peaks are shown in the diffraction patterns of **Figure 1**. All the other samples studied have similar behavior with a shift in the peaks' position. The patterns for each of the samples with x between 0.00 and 0.25 are consistent with a single phase of the cubic type with lattice parameter a = 5.652 Å for x = 0. The composition dependence of the lattice constant a on the Sb concentration are listed in **Table 1**. The lattice parameter is found to decrease linearly with increasing the Sb concentration from 5.652 Å for x = 0 to 5.630 Å for x = 0.25. The lattice parameter a = 5.652 Å for the parent alloy, with x = 0, is in good agreement with previous reported values of 5.653 Å by Cowdery and Kayser [21], and 5.650 Å by Yoon and Booth [11]. The decrease in the lattice parameter and hence the unit cell volume with increasing the Sb concentration is due to the volume contraction because of the smaller atomic radius of Sb (1.53 Å) compared with the atomic radius of Fe (1.72 Å). This behavior is in good agreement with previous reported observations for similar alloys by Shobaki *et al.* [22] for $Fe_{0.7-x}V_xAl_{0.3}$ alloys and Al-Omari and Hamdeh for $Fe_{0.7-x}V_xSi_{0.3}$ [23]. The linear decrease in the lattice parameter for these alloys as we increase the Sb concentration suggests a simple dilution process.

The magnetic field (H) dependence of the magnetization (M) for the different samples at room temperature and at low temperature, T = 100 K, is shown in **Figures 2** and **3**. It is clear from these figures that all the samples exhibit ferromagnetic behavior with soft magnetic properties, as no hysteresis observed in any of the samples. The saturation magnetization (M_s) for $Fe_{0.75-x}Si_{0.25}Sb_x$ alloys as a function of the antimony concentration (x), for

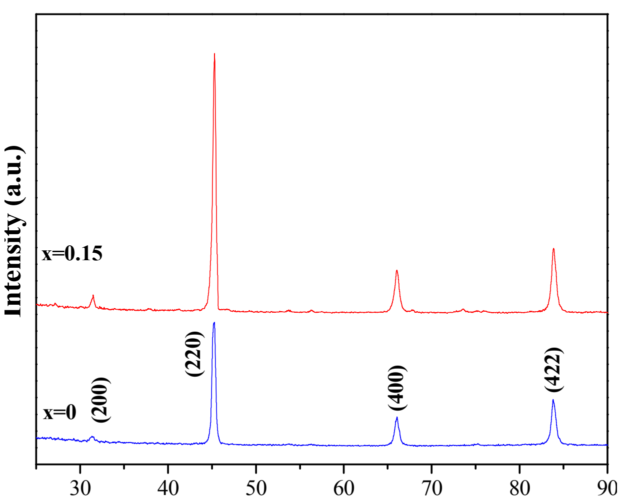

Figure 1. Typical x-ray diffraction pattern for two representative samples of the alloy system $Fe_{0.75-x}Si_{0.25}Sb_x$.

Table 1. Dependence of the lattice constant (a) and the saturation magnetization at T = 0 K (found from the extrapolation) on the antimony concentration (x), for $Fe_{0.75-x}Si_{0.25}Sb_x$ alloys.

x	0.00	0.05	0.10	0.15	0.20	0.25
a (Å)	5.652	5.649	5.644	5.639	5.635	5.630
M_s (at T = 0 K) (emu/g)	140.0	132.8	122.0	114.0	102.0	94.0

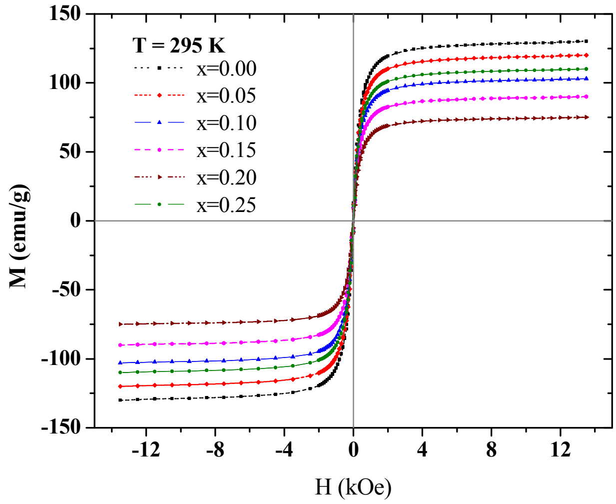

Figure 2. Magnetization curves for $Fe_{0.75-x}Si_{0.25}Sb_x$ alloys, at room temperature for different antimony concentrations (x).

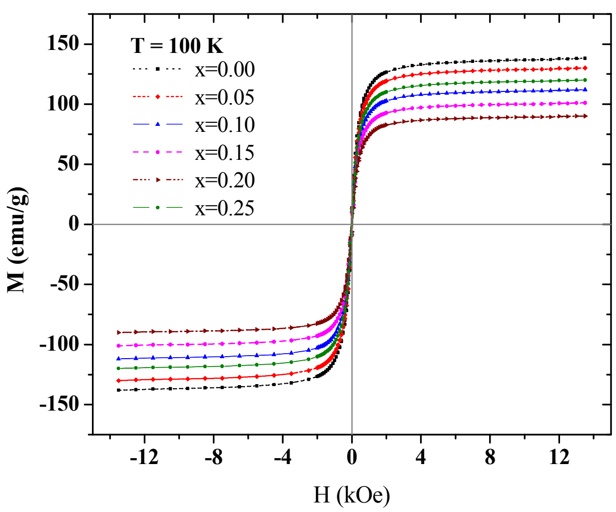

Figure 3. Magnetization curves for $Fe_{0.75-x}Si_{0.25}Sb_x$ alloys, at low temperature (T = 100 K) for different antimony concentrations (x).

the two temperatures T = 295 K and T = 100 K, is presented in **Figure 4**. As shown in this figure, M_s decreases with increasing the Sb concentration (x). This decrease is due to the replacement of ferromagnetic Fe by diamagnetic Sb in these alloys.

The temperature dependence of the magnetization for the different samples of the alloy system $Fe_{0.75-x}Si_{0.25}Sb_x$ under an applied magnetic field of 13.5 kOe is shown in **Figure 5**. It can be seen from this figure that all the samples studied have ferromagnetic ordering below Curie temperature (T_c) while the spontaneous magnetization decreases with increasing the Sb concentration (x). It is clear from **Figure 5** that the magnetization increases with decreasing the temperature, as would be expected from a system with ferromagnetic coupling below T_c. This figure

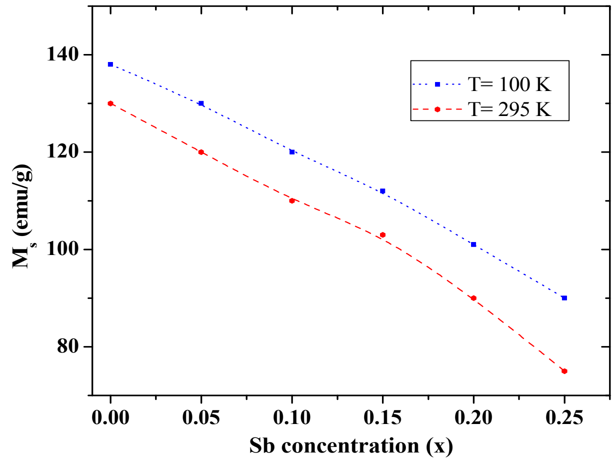

Figure 4. The saturation magnetization (M_s) as a function of the antimony concentration (x), at T = 295 K and T = 100 K, for $Fe_{0.75-x}Si_{0.25}Sb_x$ alloys.

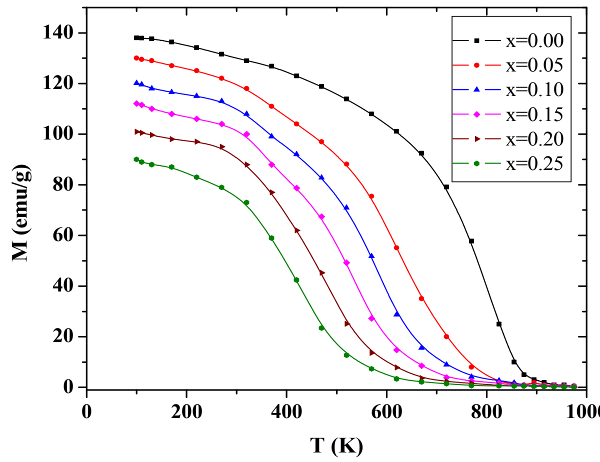

Figure 5. Temperature dependence of the magnetization for the different samples of the alloy system $Fe_{0.75-x}Si_{0.25}Sb_x$ under an applied magnetic field of 13.5 kOe.

also shows that M_s tends to decrease linearly with decreasing the temperature far below T_c. This decrease is expected for a system with ferromagnetic coupling and in qualitative agreement with our observation of the magnetic hyperfine field from the Mössbauer measurements on a similar $Fe_{0.7-x}Si_{0.3}Mn_x$ alloy system [24].

Using the method of intersecting tangents for M versus T curves in **Figure 5**, we found that the Curie temperature T_c decreases linearly with increasing the antimony concentration and the results are shown in **Figure 6**. The Curie temperature of (850 ± 5) K for $x = 0$ is in agreement with other previously reported value of 840 K by Pugaczowa-Michalska *et al.* [25]. The decrease in T_c with increasing x can be attributed to the reduction of the exchange field interaction between the spins of the magnetic Fe atoms, which can be a result of the decrease of the number of magnetic atoms surrounding each iron atom upon increasing the Sb concentration.

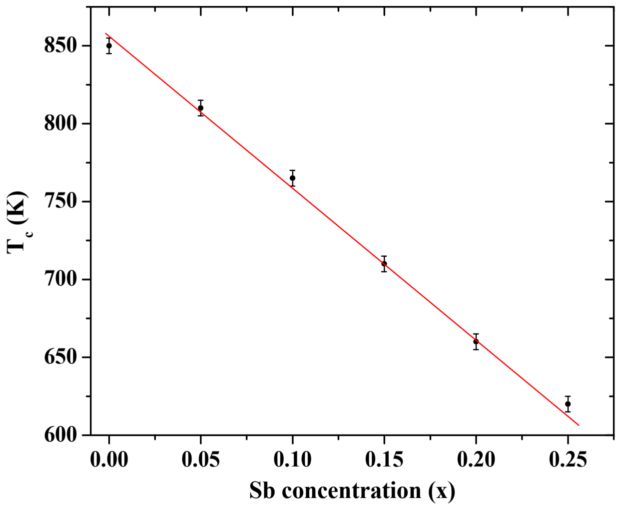

Figure 6. The Currie temperature (T_c) as a function of on the antimony concentration (x), for $Fe_{0.75-x}Si_{0.25}Sb_x$ alloys. The solid line represents a guide to the eye.

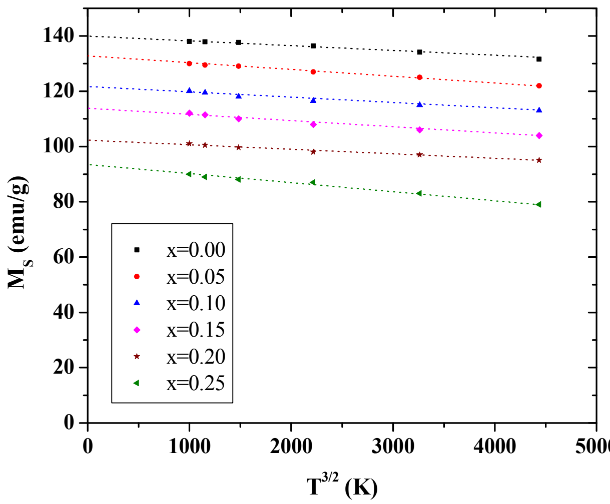

Figure 7. The saturation magnetization (M_s) as a function of $T^{3/2}$ (at low temperatures, $T \le 300$ K), for $Fe_{0.75-x}Si_{0.25}Sb_x$ alloys. The solid lines represents a straight line extrapolated to T = 0.

Since the magnetization of all the samples saturates for temperatures at room temperature and below we plotted in **Figure 7** the saturation magnetization as a function of $T^{3/2}$ at low temperatures ($T \le 300$ K), where T is the temperature in Kelvin. This figure shows that M_s as a function of $T^{3/2}$ obeys Bloch law at low temperatures, where by extrapolating M_s to T = 0 K we can estimate the saturation magnetization at that temperature, and the results are presented in table I for the different Sb concentrations. The decrease in M_s with increasing the antimony concentration, x, is due to the decrease in the iron magnetic moment due to replacing magnetic Fe by diamagnetic Sb. The extrapolated saturation magnetization for Fe_3Si at T = 0 K of (140 emu/g = 4.91 μ_B/f.u.) is in agreement with the calculated values of 5.1 μ_B/f.u. by Pugaczowa-Michalska *et al.* [25], 4.9 μ_B/f.u. by Kulikov *et al.* [26] and the experimental value of 4.7 μ_B/f.u. by Waliszewski *et al.* [12].

4. Conclusion

We have investigated the magnetic and structural properties of the alloy system $Fe_{0.75-x}Si_{0.25}Sb_x$. Structural studies revealed the formation of the DO_3-type cubic phase structure, with a decrease in the lattice constant and the unit cell volume upon substituting Fe by Sb. Magnetic measurements showed that all the samples have ferromagnetic behavior up to the Curie temperature, T_c. The obtained linear dependence of the lattice constant, Curie temperature, and the saturation magnetization at low temperatures on the antimony concentration indicate a simple dilute solution when Fe is substituted by Sb. All the results of the present work strongly suggest the reduction of the saturation magnetization with increasing x, which is due to the replacement of ferromagnetic Fe by diamagnetic Sb.

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