Elsevier Editorial System(tm) for Materials Letters
Manuscript Draft

Manuscript Number:

Title: Effect of boron addition on structural and magnetic properties of nanostructured Fe75Al25 alloy prepared by high energy ball milling

Article Type: Short Communication

Keywords: Nanocrystalline materials; Microstructure; Magnetic materials.

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Abstract: In the present work, the effect of boron addition on microstructural and magnetic properties of (Fe75Al25)1-xBx (x=0, 0.3, 0.5 and 0.7 wt%) alloys prepared by mechanical alloying process, were investigated by X-ray diffractionand vibrating sample magnetometer and SQUID techniques. The microstructural parameters such as the crystallite size, the microstrainand the lattice parameters of the bcc- solid solution Fe(Al) were deduced from the Rietveld refinement of the X-ray diffraction patterns using the X-pert HighScore. It is found that boron additions delay the formation of the BCC- Fe(Al) solid solution by mechanical alloying. The coercivity (Hc) decreases with increasing the amount of boron added. The undoped sample showed a hard ferromagnetic behaviour (Hc~750 Oe) whereas the doped ones reached a softening ferromagnetic behaviour (Hc<100 Oe).

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Dear Professor
Editor in chief of JOURNAL OF MATERIALS Letters

We are pleased to submit our paper entitled **"Effect of boron addition on structural and magnetic properties of nanostructured $Fe_{75}Al_{25}$ alloy prepared by high energy ball milling"** in your journal Materials Letters.

This paper is original and unpublished and is not being considered for publication elsewhere.

With my kind regards

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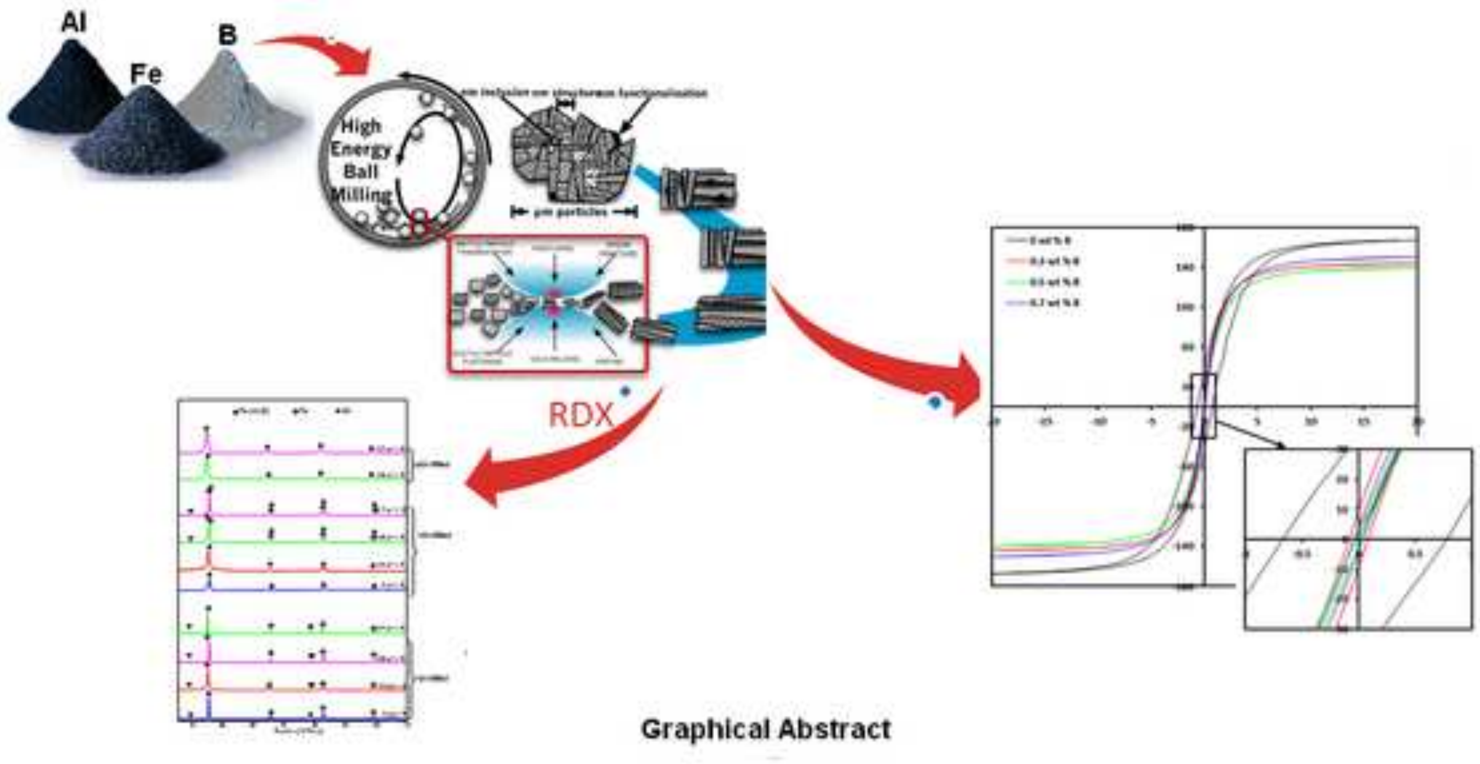
Graphical Abstract



Highlights

- The nanocrystalline $(Fe_{75}Al_{25})_{1-x}B_x$ (x= 0, 0.3, 0.5 and 0.7) powders were successfully prepared by mechanical alloying.
- The effect of boron addition on microstructural and magnetic proprieties of $Fe_{75}Al_{25}$ alloy was studied.
- The effect of boron addition on the formation of solid solution Fe (Al) and the evolution of behavior of (Hc) and (Ms) were discussed.
- Soft ferromagnetic materials were obtained with the addition of boron.

Effect of boron addition on structural and magnetic properties of nanostructured $Fe_{75}Al_{25}$ alloy prepared by high energy ball milling

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Abstract

In the present work, the effect of boron addition on microstructural and magnetic properties of $(Fe_{75}Al_{25})_{1-x}B_x$ (x=0, 0.3, 0.5 and 0.7 wt%) alloys prepared by mechanical alloying process, were investigated by X-ray diffractionand vibrating sample magnetometer and SQUID techniques. The microstructural parameters such as the crystallite size, the microstrainand the lattice parameters of the bcc- solid solution Fe(Al) were deduced from the Rietveld refinement of the X-ray diffraction patterns using the X-pert HighScore. It is found that boron additions delay the formation of the BCC- Fe(Al) solid solution by mechanical alloying. The coercivity (H_c) decreases with increasing the amount of boron added. The undoped sample showed a hard ferromagnetic behaviour (Hc~750 Oe) whereas the doped ones reached a softening ferromagnetic behaviour (Hc<100 Oe).

Keywords: Nanocrystalline materials; Microstructure; Magnetic materials; High energy ball milling.

1. Introduction

Nanomaterials have experienced a rapid development in recent years due to their existing and/or potential applications in a wide variety of technological areas such as electronics, ceramics, catalysis, structural components, magnetic data storage etc [1]. The size of the materials should be reduced to the nanometer scale to meet the technological demands in these areas. As the size reduces into the nanometer range, the materials exhibit interesting and peculiar mechanical and physical properties e.g. increased mechanical strength, higher specific heat and enhanced diffusivity, and electrical resistivity compared to conventional coarse grained counterparts [2]. Mechanical alloying (MA) is one of the most common techniques used to prepare these materials in its nanocrystalline state [3]. The mixed powders are mechanically milled to achieve nanocrystalline structure which is an effective route for the development of magnetic properties [4]. The FeAl alloy system can be selected in many studies for its rich spectrum of magnetic properties, varying from straightforward ferromagnetism (FM) in the iron-rich alloys to super-paramagnetic (SPM) behaviour [5]. They reported that alloys with compositions around 70% of Fe, present a structure composed of small ferromagnetic clusters (~25 Å) embedded in a paramagnetic matrix. Further, nanostructured Fe-based soft magnetic materials as a new class of engineering materials can be produced by MA [6]. The unusual behaviour of the clusters with temperature (e.g. increased effective magnetization) and the nature of the FM–SPM magnetic transition, to this day remains an open question. In addition, a recent study showed that the addition of a small amount of boron can change the fracture mode during milling and also produces a growth of the grain boundaries resulting to the formation of Fe_2B [7]. However, knowledge of the effect of boron addition on microstructural and magnetic properties of Fe-Al alloys remains limited [7, 8]. The purpose of the present study is to investigate the microstructure evolution and the magnetic behaviour in $Fe_{75}Al_{25}$ alloy doped with boron during mechanical alloying.

2. Experimental data

Elemental powders of Fe, Al and B with purity of 99.99% of the nominal composition $Fe_{75}Al_{25}$ powder doped with small amounts of boron (x= 0; 0.3; 0.5 and 0.7 wt.%) were mixed and mechanically alloyed by using a laboratory planetary ball mill (PM 400) under Ar atmosphere. The ball-to-powder weight ratio used was maintained as 12:1 and the milling speed was adjusted to 400 rpm. X-ray diffraction (XRD) measurements were done using a D-500 Siemens equipment with Cu K_α radiation. The magnetic characterization was carried out by Superconducting Quantum Interference Device from Quantum Design SQUID MPMS-XL at 300 K.

3. Results and discussion

Fig. 1 shows the XRD patterns obtained for the milled $(Fe_{75}Al_{25})_{1-x}$ B_x(x= 0, 0.3, 0.5 and 0.7 wt.%) powders after 10, 40 and 80 h. One can conclude that the addition of B delay the formation of the bcc-solid solution Fe(Al). However, for higher content of boron (x>0.3 wt.%), the solid solution has been accomplished after 40 h milling. The dependence of the calculated crystallite size and microstrains on B additions of the as-milled powders for 6, 20 and 40 h, is given in Fig. 2a and 2b, respectively. The refinement of the microstructure of all powders during MA is significant (less than 50 nm). Remaining at the nanoscale, the crystallite size increases and the microstrains decreases with increasing the content of B added. This behavior is similar to 6, 20 and 40 h milling results. This can be associated with a segregation of B atoms in the grain boundaries, which avoids the growth of crystallites of the bcc phase [9]. In addition, instability in the segregation process could be the justification for the increasing of the crystallite size and the decreasing of internal stress during milling. Fig. 2c shows the dependence of the lattice parameter on B contents for the as-milled powders for 6, 20 and 40 h. As shown, the lattice parameter of solid solutions should be higher than that of Fe (R_{Al}=1.43 Å is much higher than R_{Fe}=1.26 Å). In addition, when the concentration of B increases the lattice parameter decreases due to the substitution of the Al atoms by B. This substitution helps to bring the diffraction planes nearer, which leads to decrease of the lattice parameter [10]. Fig. 3 shows typical hysteresis loops of MA powders milled for 40 h at 300 K. The greatest difference observed is found in the undoped sample which has the highest saturation. It is noteworthy that all powders had similar hysteresis loops which entails that these samples have a ferromagnetic behavior as detailed in the graph inserted on Fig. 3. As suggested by Cable et al. [5], the ordered $Fe_{0.7}Al_{0.3}$ is ferromagnetic below 400 K but becomes paramagnetic on cooling below 170 K and then is mictomagnetic below 92 K. The authors used neutron-scattering methods to probe the microscopic magnetic-moment distribution of this alloy showing the presence of large ferromagnetic clusters throughout all of these ordered regions and that the bulk behavior results from inter-cluster coupling. Fig. 4 shows the influence of B additions on coercivity (Hc) and saturation magnetization (Ms). We can deduce that the doped samples show a soft magnetic behavior (Hc<100 Oe). However the undoped one has a much harder magnetic behavior (Hc~700 Oe). This can be explained by the precipitation of tetragonal phase Fe_2B in the grain boundaries. The tetragonal phase decreases Hc through the pinning of the magnetic domain walls which causes alloys to lose a part of their high magnetic anisotropy [11]. Also, this behavior can be explained by the decrease of internal stress than by the addition of B already seen in the microstructural study [12].

The dependence of (Ms) on the B additions is given in Fig. 4. This inhomogeneous evolution of Ms can be explained by the magnetostriction effective anisotropy and stress-induced during mechanical alloying. Saturation magnetization is generally regarded as independent of the microstructure and strongly dependent on the chemical composition; unlike coercivity Hc, saturation magnetization Ms is structurally insensitive.

4. Conclusion

The nanocristalline $(Fe_{75}Al_{25})_{1-x}B_x$ (x= 0, 0.3, 0.5 and 0.7 wt%) powders with good soft magnetic properties were produced by MA. By increasing boron content, the crystallite size increases with remain in nanoscale and the residual stress decreases. In addition, the hard ferromagnetic behavior becomes a soft magnetic one which can be explained by the precipitation of tetragonal phase Fe_2B in the grain boundaries. The inhomogeneous evolution of saturation magnetization can be originated from the refinement of the crystallite size below or above of the magnetic exchange length and consequently from the averaging out of the magnetocrystalline anisotropy by exchange interaction.

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Figures captions

Fig. 1. X-ray diffraction patterns obtained for the milled $(Fe_{75}Al_{25})_{1-x}$ B $_x$(x= 0, 0.3, 0.5 and 0.7 wt%) powders after (a) 10, (b) 40 and (c) 80 h.

Fig. 2. The dependence of the calculated (a) crystallite size, (b) microstrains and (c) lattice parameter on B contents of the milled powders for 6, 20 and 40 h.

Fig. 3. Typical hysteresis loops of the MA powders for 40 h at 300K.

Fig. 4. The dependence of the (a) coercivity (Hc) and (b) saturation magnetization (Ms) on boron contents of the milled powders for 40 h at 300K.

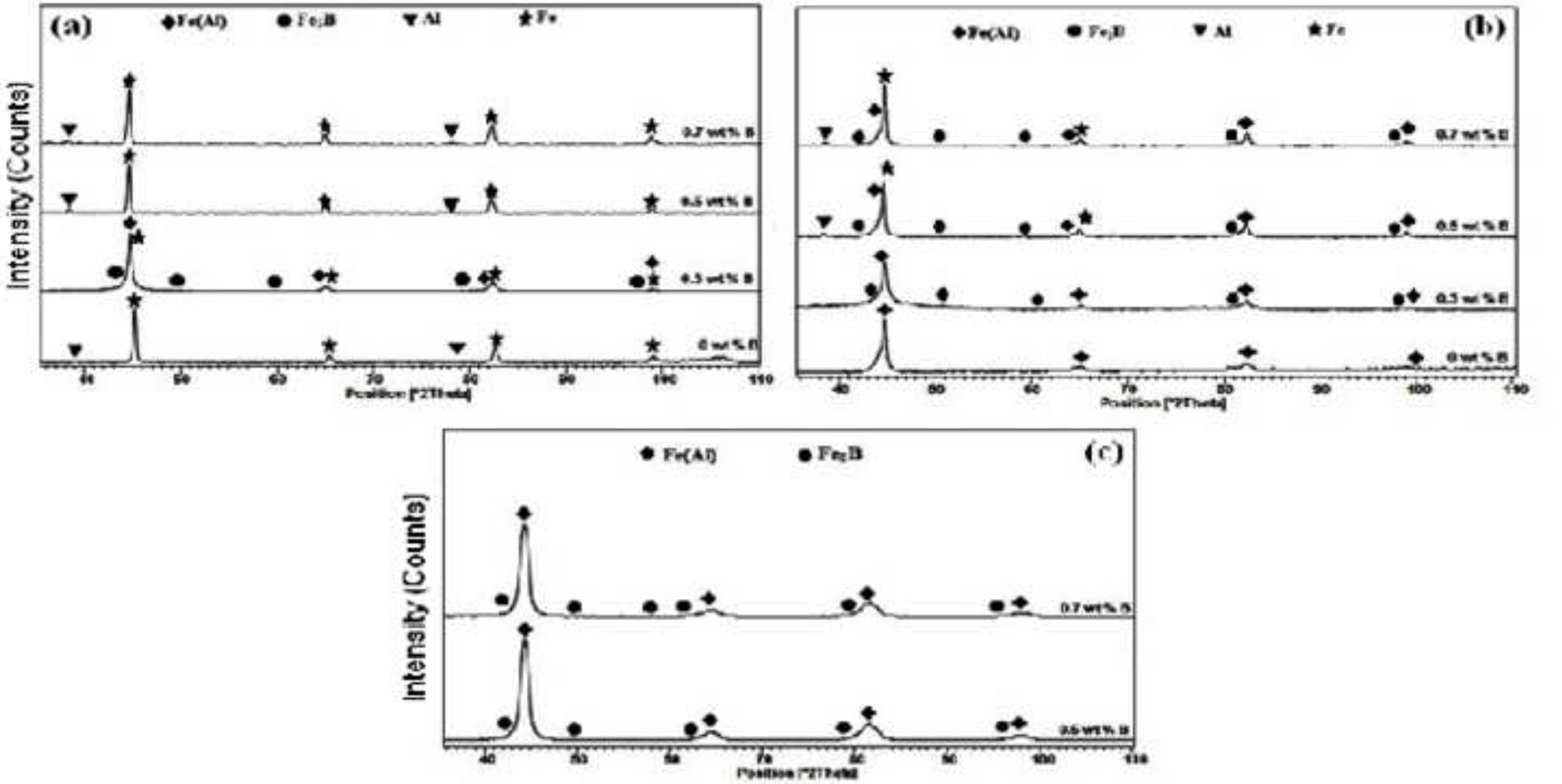
(a)
◆ Fe(Al)
● Fe2B
▼ Al
★ Fe
Intensity (Counts)
0.7 wt% B
0.5 wt% B
0.3 wt% B
0 wt% B
40
50
60
70
80
90
100
110
Position [°2Theta]
(b)
◆ Fe(Al)
● Fe2B
▼ Al
★ Fe
0.7 wt % B
0.5 wt % B
0.3 wt % B
0 wt % B
40
50
60
70
80
90
100
110
Position [°2Theta]
(c)
◆ Fe(Al)
● Fe2B
Intensity (Counts)
0.7 wt% B
0.5 wt% B
40
50
60
70
80
90
100
110
Position [°2Theta]

Fig. 1

Figure 2

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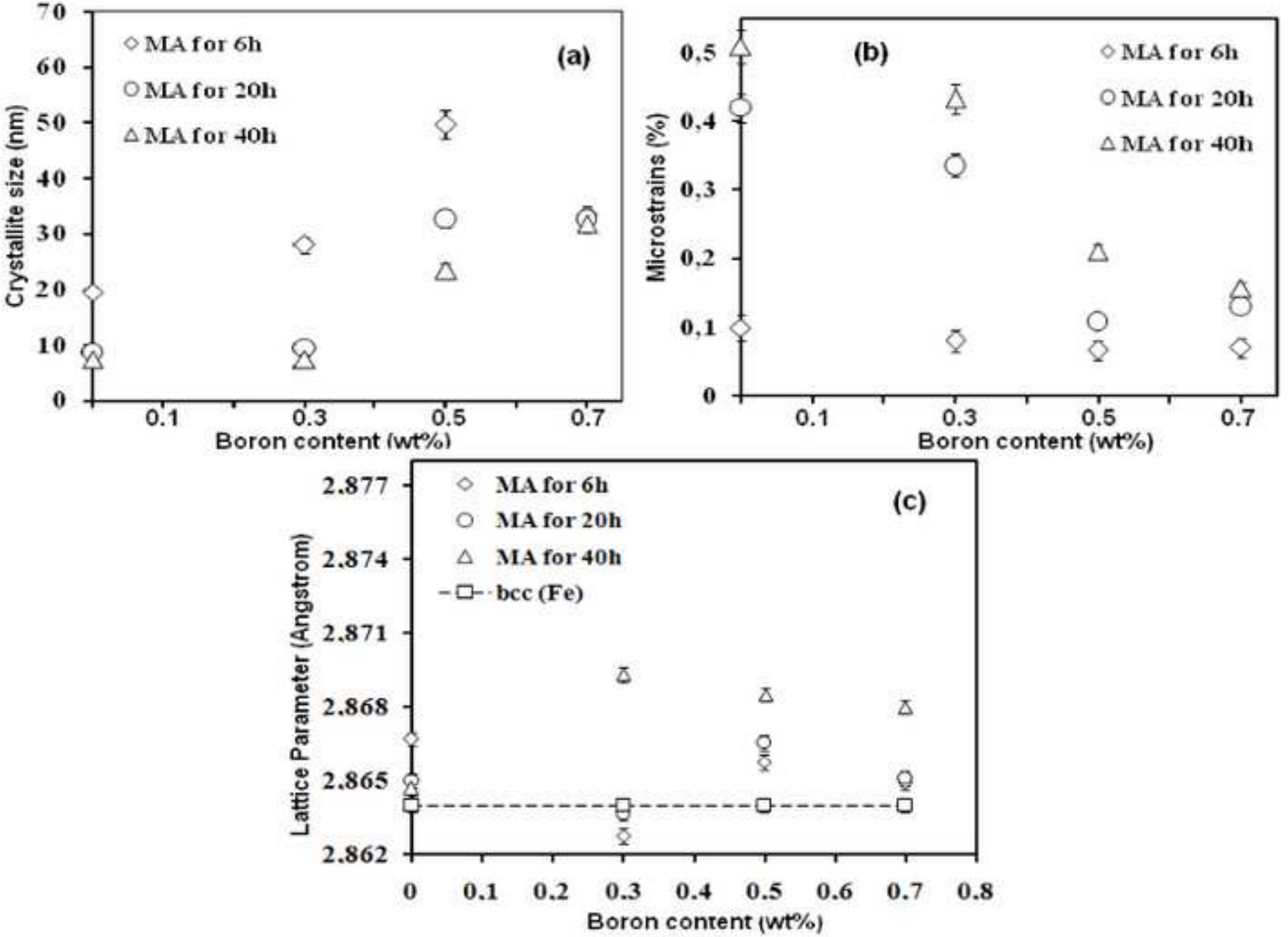

Fig. 2

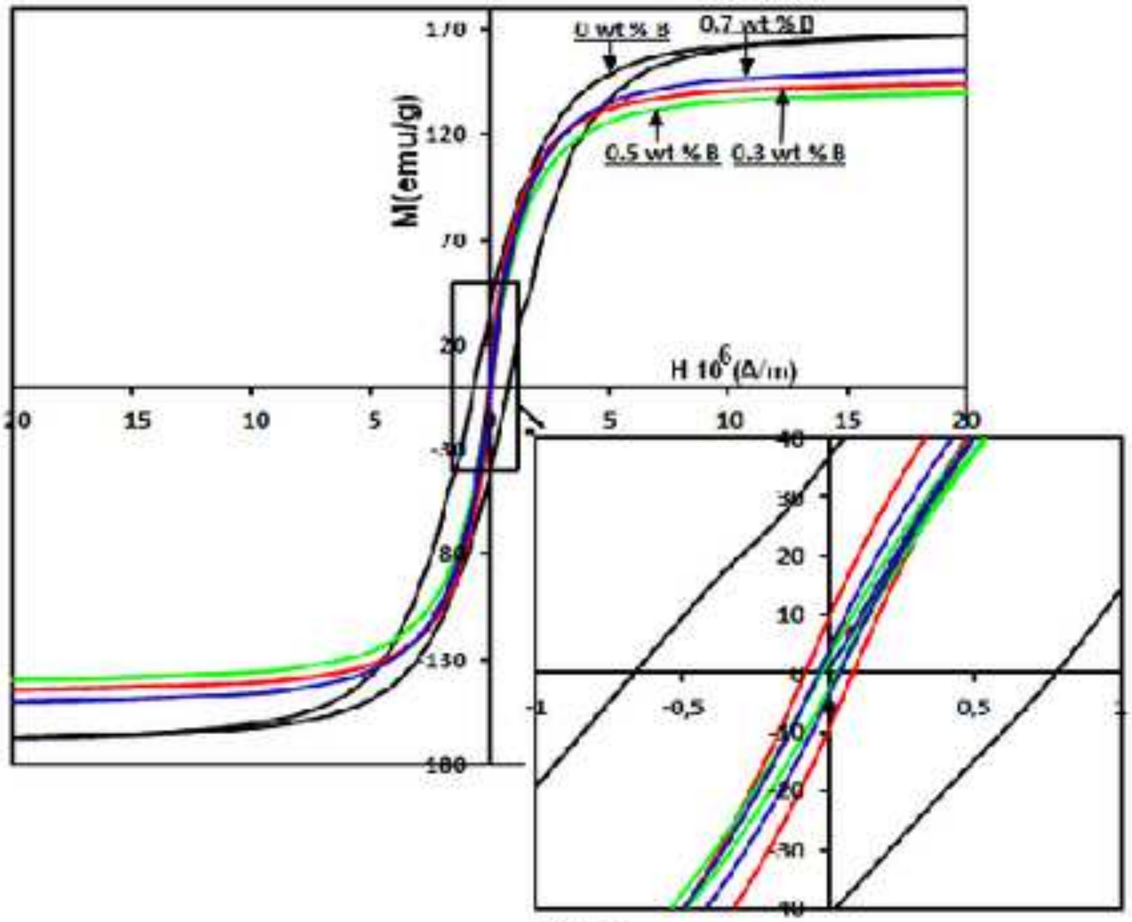
170
120
70
20
-30
-80
-130
-180
M(emu/g)
H 10^6 (A/m)
20
15
10
5
5
10
15
20
0 wt % B
0.7 wt % B
0.5 wt % B
0.3 wt % B
40
30
20
10
0
-10
-20
-30
-40
-1
-0,5
0,5
1

Fig. 3

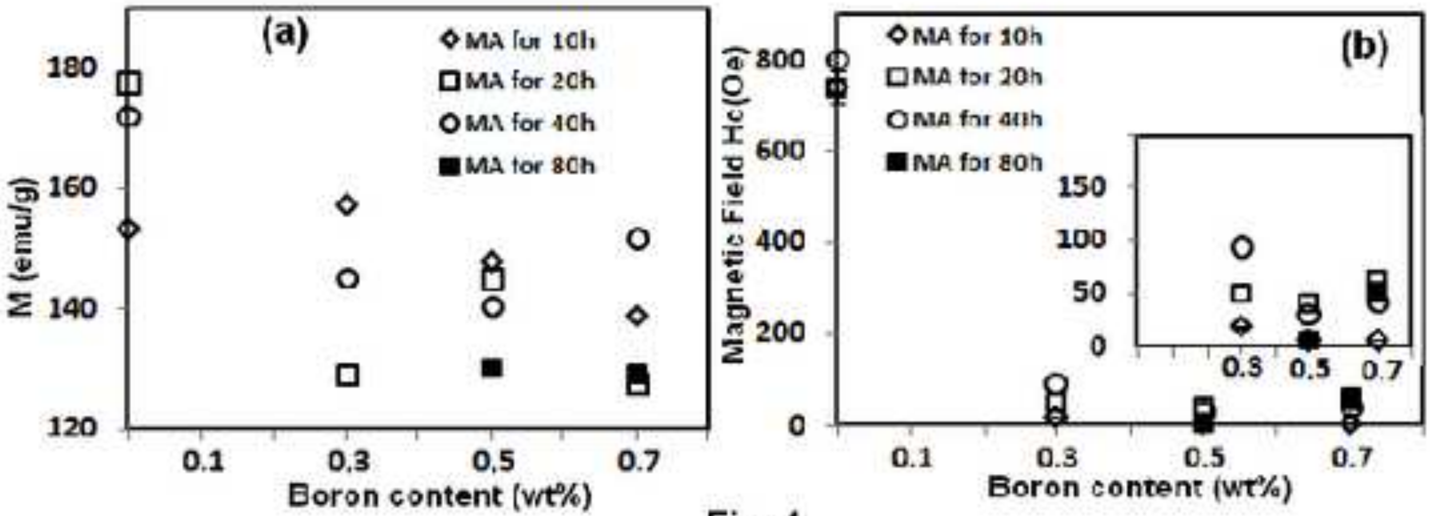
(a)
MA for 10h
MA for 20h
MA for 40h
MA for 80h
180
160
140
120
M (emu/g)
0.1
0,3
0,5
0.7
Boron content (wt%)
(b)
MA for 10h
MA for 20h
MA for 40h
MA for 80h
800
600
400
200
0
Magnetic Field Hc(Oe)
150
100
50
0
0.3
0.5
0.7
0.1
0.3
0.5
0.7
Boron content (wt%)

Fig. 4