LOCAL STRUCTURE AND MAGNETIC PROPERTIES OF Fe-Al NANOCRYSTALLINE ALLOYS FABRICATED BY MECHANICAL ALLOYING TECHNIQUE AS A FUNCTION OF ALUMINIUM CONTENT

by Kontan Tarigan

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ABSTRAK

Struktur dan sifat magnetik dari pada sampel-sampel paduan nanokristal $Fe_{100-x}Al_x$ (x = 10, 30, 50, 70 dan 90 at. %) yang disiapkan dengan teknik mechanical alloying (MA) dengan waktu milling masing-masing sampel 24 jam, telah dilakukan dengan rinci. Struktur diperiksa dengan XRD dan EXAFS dan mengungkapkan semua sampel merupakan paduan nanokristalin Fe-Al dengan ukuran Kristal rata-rata 10 nm. Untuk sifat maknetik, VSM menunjukkan hasil baik saturasi magnetic (Ms) dan kursivitas (Hc) sangat bergantung pada kandungan Al, yang terkait dengan perubahan struktur dan ukuran Kristal. Berdasarkan penelitian ini, kandungan Al dapat dimanipulasi untuk mendapatkan harga-harga Ms dan Hc.

Kata kunci: $Fe_{100-x}Al_x$ nanocrystalline, XRD, VSM, and EXAFS.

ABSTRACT

Structural and magnetic characterizations of $Fe_{100-x}Al_x$ (x = 10, 30, 50, 70, and 90 at. %) nanocrystalline alloys, prepared by mechanical alloying (MA) with 24 hrs milling time, have been carried out in details. The structures were examined by using XRD and EXAFS. Our XRD and EXAFS studies revealed that the Fe-Al alloy nanocrystalline with an average crystallite size around 10 nm was formed by milling of Fe and Al powders. For the magnetic properties, the data obtained from a vibrating sample magnetometer (VSM) exhibited both magnetic saturation (M_s) and coercivity (H_c) depending strongly on the Al content, which are related to the changes in structure and crystallite size. Based on these results, one can manipulate the Al content to obtain the appropriate M_s and H_c values.

***Key words**: $Fe_{100-x}Al_x$ nanocrystalline, XRD, VSM, and EXAFS.*

INTRODUCTION

It is known that ferromagnetic (F) and paramagnetic (P) phases observed at room temperature in the Fe-Al series have been exhibited spin-glass (SG) and superparamagnetic (SP) behaviors. The properties of the F phases present in the Fe-Al alloys are strongly dependent both on their composition and on their fabrication details [1]. The fabrication of nanocrystalline alloys can be carried out by mechanical milling (MA), including two important effects: (i) the disordering of the structure, and (ii) the decrease in the grain size. Basically, $Fe_{100-x}Al_x$ alloys prepared by MA exhibit magnetic behaviors depends on Al content and the milling conditions [1],[2].

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It s shown that Fe-Al intermetallics alloys with the increasing of Al concentration, the oxidation and sulphidization resistances of FeAl alloys also increased, while their densities decreased [3],[4]. Particularly, the Fe-Al intermetallic compounds offer a combination of attractive properties, such as high specific strength, good strength at intermediate temperatures under oxidizing, carburizing and sulfidizing atmosphere [5],[6].

The degree of order in Fe-Al intermetallic alloys has an important influence on their magnetic properties. Moreover, the deformation of ordered alloys causes a dramatic increase in magnetization [7]. In Fe-Al alloys with the bcc structure, the magnetic moment of Fe atoms depends on the local structure. As a rule, Fe atoms with less than four nearest Fe neighbors possess no localized magnetic moments, and Fe atoms become magnetic only when they have four or more Fe nearest neighbors [8]. The magnetic properties of an assembly of small grains depend on the counter play between the local magnetic anisotropy energy and the ferromagnetic exchange energy [9].

The Fe-Al equilibrium phase diagram after Kubaschewski is shown in Figure 1. The system is characterized with an iron-based solid solution and six non stoichiometric intermetallic compounds of Fe_3Al, FeAl (α_2), $FeAl_2$, Fe_2Al_3 (ε), Fe_2Al_5 and $FeAl_3$. Table 1 indicates crystal structure and stability range for this phase diagram with special emphasis on the intermetallic Phases (IMPs)[10],[11].

Table 1: Crystal structure and stability range of the phases formed in Fe-Al binary System at room temperature [7].

Phases	Crystal structure	Stability range (at.%)
Fe solid solution	BCC	0-45
γ-Fe	FCC	0-1.3
FeAl	BCC (Order)	23-55
Fe_3Al	DO3	23-34
Fe_2Al_3	Cubic (complex)	58-65
$FeAl_2$	Triclinic	66-66.9
Fe_2Al_5	Orthorhombic	70-73
$FeAl_3$	Monoclinic	74.5-76.5
Al solid solution	FCC	99.998-100

In this work, we studied $Fe_{100-x}Al_x$ alloys with by changing aluminum content. Their structural and magnetic properties were then studied by means of X-ray diffraction (XRD), extended X-ray absorption fine structure spectroscopy (EXAFS), and vibrating sample magnetometer (VSM).

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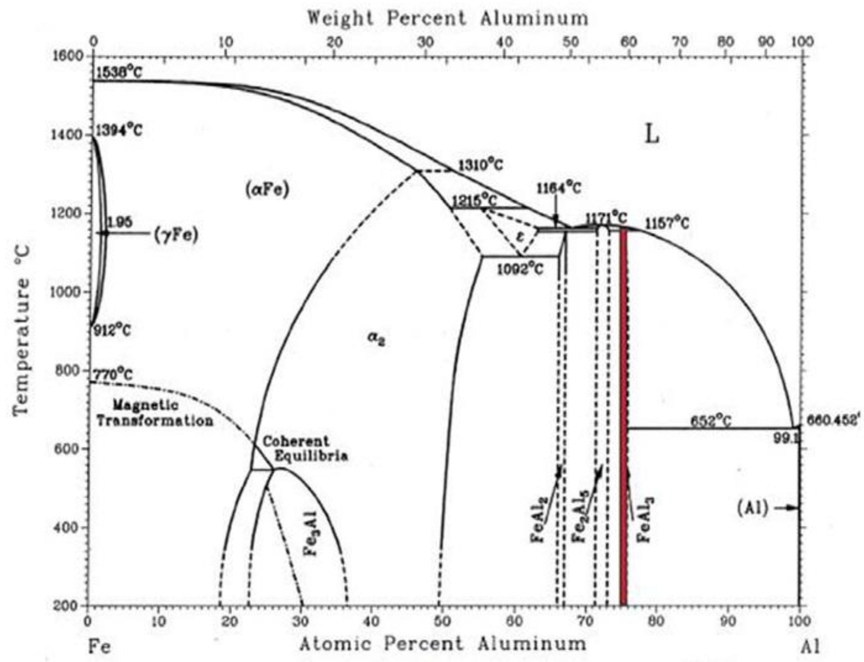

Fig. 1: Fe-Al equilibrium phase diagram [8].

EXPERIMENTAL

$Fe_{100-x}Al_x$ (x = 10, 30, 50, 70 and 90 at. %) nanocrystalline metastable alloys were prepared via mechanical alloying (MA) by utilizing a high-energy planetary ball mill SPEX 8000 mixer. A starting mixture was made by combining pure Fe (53 µm, 99.9 %) and Al (53-106 µm, 99.9 %) powders. The weight ratio of balls-to-powder mixture was 5:1. $Fe_{100-x}Al_x$ alloys were mixed and grounded for 24 hrs. This process was performed in Ar ambience to avoid oxidation. The sizes of the particles and their morphologies were checked preliminarily by using scanning electron microscopy (SEM). Magnetic properties were measured by utilizing a vibrating sample magnetometer (VSM). XRD data were obtained by using the Cu-K_α radiation. EXAFS data were collected from the 3C1 EXAFS beam line of the Pohang Light Source (PLS). The PLS was operated with an energy of 2.5 GeV, and a maximum current of 200 mA. EXAFS spectra were obtained at Fe K-edge (7112 eV) in the transmission mode at room temperature. The sample chamber was filled with pure nitrogen gas. Furthermore, the EXAFS data were analyzed by making use of IFEFFIT software. The magnetic properties were discussed in connection with structural characterizations.

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RESULT AND DISCUSSION

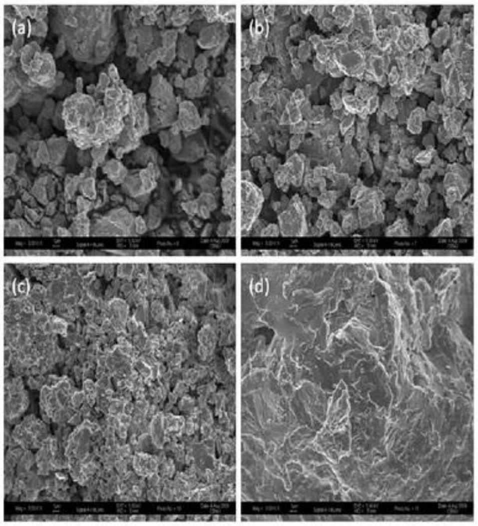

Fig. 2: Typical SEM images of $Fe_{100-x}Al_x$ nanocrystalline with (a) x = 10, (b) x = 50, (c) x = 70, and (d) x = 90 at. %.

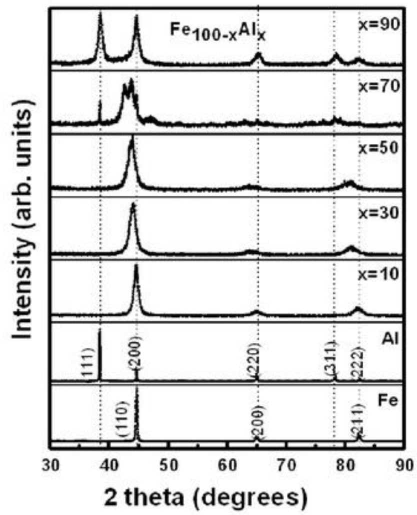

Fig. 3: XRD pattern of $Fe_{100-x}Al_x$ with x is different Al contents.

Figure 2 shows SEM images of four representative $Fe_{100-x}Al_x$ samples with various content of x = 10, 50, 70, and 90 at. % Al. The morphology and size of particles changed when the composition is changed. This is due to the impaction of high energy generated from the ball-to-ball and ball-to-vial wall collisions. The average particle size estimated from the SEM images is found to increase with increase of the aluminum content. To examine structural characterization, we have based on an XRD and EXAFS. In the Figure 3 we can see the XRD patterns of pure Fe, pure Al, x =10, x = 30, x =50, x = 70, and x =90_0, respectively. The peaks have checked by using PDFWIN software. The peaks positions of $Fe_{90}Al_{10}$ are the same and broaden compare to that of the pure Fe and the peaks of Al have disappeared. It means that the atoms Al have filled the interstitial position of Fe structure. So, the host with A2 structure has non-uniform strain. For the $Fe_{70}Al_{30}$ patterns are a little bit shifted to smaller angel and broaden compare to that of the pure Fe. Some Al atoms filled the body center and interstitial position and the new structure B_2 is formed. For $Fe_{50}Al_{50}$, the composition of the structure is fixed in B_2, but the intensity is decreased because of Al atoms filled body center and interstitial position is also increased. The $Fe_{30}Al_{70}$ alloy has new peaks. It means that the sample has a new structure, based on the phase diagram it may be orthorhombic lattice. The last, peaks position of $Fe_{10}Al_{90}$ are same and broaden compare to that of the pure Al. It means

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that some atoms Fe have filled the interstitial position of host Al lattice and the structure has non-uniform strain.

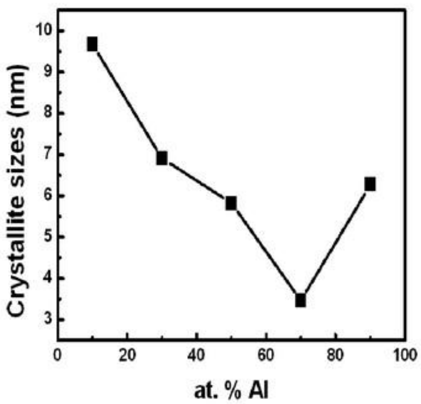

Fig. 4: Variation of the crystallite size of the samples as a function of aluminum content.

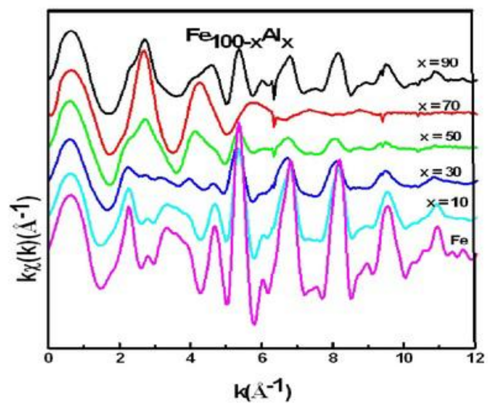

Fig. 5: *k*-weighted EXAFS spectra for mechanically-alloyed $Fe_{100-x}Al_x$ (x=10, 30, 50, 70, and 90 at. % Al).

The variation of crystallite size with aluminum concentrations is shown in Fig.4. The crystallite size is calculated from XRD data by using Debye Scherrer formula. The crystallite size is decreased with increasing aluminum content up to 70 % and then increased with increasing of aluminum content. The average crystallite size of all samples varied in the range of 3.5 to 10 nm.

The local structure and the atomic ordering were examined also by using EXAFS experiment. Variations of EXAFS spectra are related to the information on the structural changes of alloys at atomic scale. Mostly, the reduction of the amplitude of EXAFS spectrum is caused by the disorder in local structure. The phase shift of EXAFS spectrum is related to the change of chemical order [12]. In Figure 5, we can see many patterns are changing just like those of the XRD results. The radial atomic density in real space can be seen in the Fourier transformed spectrum. The peaks of the Fourier transformed spectra have the local structural information, such as the coordination number and the bonding distance, and the information on the vibration of neighboring atoms [13].

Figure 6 shows the Fourier transform of the EXAFS spectra measured at the Fe K-edge for different composition. The vertical dot lines indicate the first, second, third, and fourth shell of pure Fe which served as the guide lines (for alloyed samples to be compared). Based on the spectra at first shell shown that the patterns of x = 10 to 50 relative in a good agreement with Fe spectrum (x = 0) even thought the amplitudes are decreased and phases are shifted. It means the lattices are same but there are some variations of atoms in the structure. This indicates that the numbers of Fe–

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Fe direct bonds were decreased due to the diffusion of Al atoms into the BCC Fe shells. For x = 70 the patterns are quite different from the Fe signal. It means that the lattice has changed to new one. Also, for x = 90 we can say that Fe atoms are placed out of FCC lattices are belong to Al as the host.

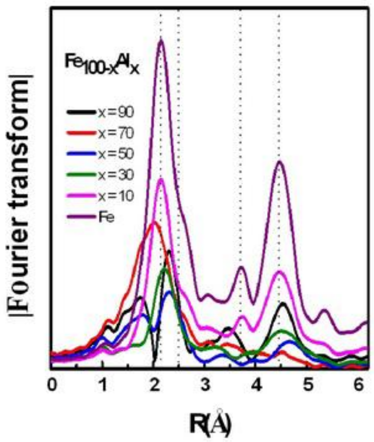

Fig. 6: Fourier transform measured at the Fe K-edge EXAFS spectra for mechanically-alloyed $Fe_{100-x}Al_x$ (x=10, 30, 50, 70, and 90 at. % Al).

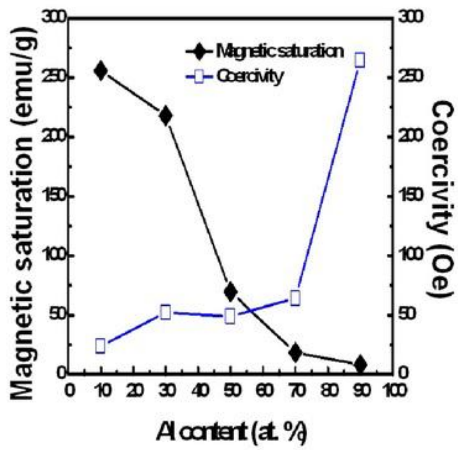

Fig. 7: The magnetization and coercivity versus a variation of aluminum content.

The magnetic properties were being characterized using VSM, exhibited both magnetic saturation (M_s) and coercivity (H_c) strongly dependent on its composition. In Figure 7, the magnetization and the coercivity are shown as a function of aluminum content. The M_s is decreased because there are a decrease in its particle size and a change in the composition of a mixed powder. This variation could come from the dilution of magnetic lattice of Fe which is caused by the increasing of aluminum content. Furthermore, coercivity (H_c) is found to increase as the aluminum content also increased, which is assigned to a reduction in the crystallite size in single domain regime.

Especially of x = 90, the coercivity remain increased even though the crystallite size increase because of the high lattice strains. The coercivity increases due to the magnetoelastic density stored in the sample, which is higher than the structural anisotropy, and thus, Hc behavior is governed mainly by the strains present in the sample [14]. One we can see from the Fig. 7 also that magnetic saturation and coecivity is conflicting, the magnetic saturation decreased as the coercivity increased.

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CONCLUSION

In this work, we found that the structure of $Fe_{90}Al_{10}$ is A2, $Fe_{70}Al_{30}$ is B2, $Fe_{50}Al_{50}$ is B2, $Fe_{30}Al_{70}$ is orthorhombic, and $Fe_{10}Al_{90}$ is fcc. It was explicitly shown in the XRD patterns by the peaks of $Fe_{100-x}Al_x$, which are reduced, broadened and shifted to small angle. The difference of them can be shown clearly by EXAFS. In EXAFS, the structure change is manifested by the variation of amplitude and phase spectra. Both of magnetic saturation (M_s) and coercivity (H_c) were significantly changed when the composition were changed in principle. We may control the Al content to obtain appropriate M_s and H_c values.

REFERENCES

1. Ligia E. Zamora, et al., *Physical Review B* 79, 094418 (2009).
2. Laszlo F. Kiss, et al., *Reviews on Advanced Materials Science* 18 (2008) 505-508.
3. Varkey Sebastian, N. Lakshmi, K. Venugopalan, *Materials Letters 61* (2007) 4635-4638.
4. R.A. Varin, J. Bystrzycki, A. Calka, *Intrmetallics* 7 (1999) 917-930.
5. Hongwei Shi, Debo Guo, Yifang Ouyang, *Journal of Alloys and Compounds*. 455 (2008).
6. D. P. Dutta, et al., *Journal of Materials Chemistry* 19, 6 (2007).
7. Martin Rodriguez, et al., *Phyical Review* B 71, 212408 (2005).
8. Laszlo F. Kiss, et al., *Reviews on Advanced Materials Science* 18 (2008).
9. G. Herzer, *IEEE Transactions on Magnetics* 26, 5 (1990).
10. M. Potesser, et al., TMS, *EPD Congress* 2006 (167-176).
11. M. Palm, *Intermetallics* 13 (2005) 1286–1295.
12. D.S. Yang, et al., *Journal of the Korean Physical Society* 50 (2007)1078–1083.
13. D. S. Yang, et al. *Journal of the Physical Society of Japan* 71 (2002) 487–490.
14. M. Lopez, et al., *Journal of Magnetism and Magnetic Matererials* 290–291 (2005) 171–174.

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