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Magnetic properties and EXAFS study of nanocrystalline $Fe_2Mn_{0.5}Cu_{0.5}Al$ synthesized using mechanical alloying technique

Dwi Nanto [a,c], Dong-Seok Yang [b], Seong-Cho Yu [a,*]

[a] Department of Physics, Chungbuk National University, Cheongju 361-763, South Korea
[b] Physics Division, School of Science Education, Chungbuk National University, Cheongju 361-763, South Korea
[c] Physics Education, Syarif Hidayatullah State Islamic University, Jakarta 15412, Indonesia

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ABSTRACT

Nanocrystalline $Fe_2Mn_{0.5}Cu_{0.5}Al$ has been synthesized by the mechanical alloying technique and studied as a function of milling time. Alloy nature of $Fe_2Mn_{0.5}Cu_{0.5}Al$ was observed in a sample milled for 96 h. The magnetic saturation is 4.0 μ_B/f.u., which coincidently follows Slater–Pauling rule at 5 K. Nanocrystalline $Fe_2Mn_{0.5}Cu_{0.5}Al$ has enhanced saturate magnetization compared to any other fabrication of Fe_2MnAl reported. Cu element plays an important role in site competes with other elements and may result in the enhancement of saturate magnetization. In accordance to the magnetic results and EXAFS pattern, it was revealed that the dynamics of magnetic properties were confirmed as structural changes of nanocrystalline $Fe_2Mn_{0.5}Cu_{0.5}Al$.

1. Introduction

Freidrich Heusler discovered an alloy type well known as Heusler alloy in 1903 [1]. It was debated why an alloy consisting of copper, manganese and tin, Cu_2MnSn, that each of them has a character of nonferromagnetic, builds a ferromagnetic effect. Bradley and Rodgers suggested that a fully ordered structure of the $L2_1$ type crystal composed of copper, manganese and aluminum construct a ferromagnetism behavior [2]. A combination of all ternary intermetalic that compounds with chemical formula X_2YZ nowadays is called as a full Heusler phase [3]. Heusler alloys type materials have attracted scientists to explore more research including on environmental techniques as thermoelectrics and solar cells discovered their amenities, including tunability of their properties by substitution of atoms, nontoxicity, and the low price [4].

According to Slater–Pauling (SP) rule, there is a relation between the average valence electron concentration (*VEC*) and the magnetic moment [5,6]. SP predicts magnetic moments m for half metallic X_2YZ phases as $m = VEC - 24$. In order to achieve high magnetization, one can consider this rule. By using computational method, the SP rule has been demonstrated for the entire series of possible combination Heusler alloys. It has magnetic phases because the magnetic moments are a function of the valence electron count [3].

Mechanical alloying (MA) technique, in comparison with arc-melting, is more cost-effective and is able to process powders in quantity [7]. Therefore, MA technique is recognized as a powerful technique for the production of Heusler alloys. Some direct formations of Heusler alloys using MA technique has been reported are Co_2CrAl [7], Fe_2VAl [8], Fe_2CoGe [9] and Fe_2MnGe [10]. In this work, we use Fe_2MnAl (FMA) as a parental study due to its magnetic property is well known as polycrystalline ingot [11,12], ribbon [13] and nanosized ingot [14]. We also use Fe_2CuAl (FCA) as a parental study that has been predicted exists theoretically [3]. Recently, FCA was proven exist [15]. As best as we concern, no results reported the effect of Cu-doped in FMA or FCA on the mechanical alloying technique for studying its structure and magnetic behavior. The result shows that nanocrsytalline $Fe_2Mn_{0.5}Cu_{0.5}Al$ (FMCA) alloy has much larger coercivity than nanosized ingot FMA [14]. Interestingly, the magnetic saturation estimates more than twice higher than it. In accordance to the magnetic results, it is also interesting to discuss in detail the local structure of nanocrystalline FMCA by using extended X-ray absorption fine structure (EXAFS) investigation.

2. Experiments

We use Fe element with nominal purity 99.9% up, 5 μm average particle size and Mn with purity of 99.9%, 75 μm average particle sizes as raw material. Cu element has purity of 99.85%. Element Al powders with nominal purity of 99.5 were mixed with those elements. $Fe_2Mn_{0.5}Cu_{0.5}Al$ (FMCA) alloys were prepared by mechanical alloying technique using a SPEX 8000 mixer and stainless steel balls in Ar atmosphere to avoid oxidation during the alloying process. The starting material was a mixture of those pure Fe, Mn, Cu and Al powders originally. We varied the milling time from one

* Corresponding author. Fax: +82 42 261 2269.
E-mail addresses: scyu@chungbuk.ac.kr, scyu@cbnu.ac.kr (S.-C. Yu).

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hour up to 96 h in order to study the alloying process. The weight ratio between ball and powder was about 6:1. Magnetization and coercivity of the samples were measured by a vibrating sample magnetometer (VSM) with a maximum field of 15 kOe at room temperature. Magnetic hysteresis at low temperature was measured by superconducting quantum interference device (SQUID). X-ray diffraction (XRD) data were obtained with a monochromatic Cu–Kα radiation. The influence of milling time on local ordering around the central atom was studied by using EXAFS beamline at Pohang Accelerator Laboratory (PAL), South Korea. PAL was operated with an electron energy of 3.0 GeV and a range current of 93.3–90.4 mA. The EXAFS spectra were investigated in Fe–K edge (7112 eV) in the transmission mode at room temperature. The EXAFS data were interpreted by using Artemis and Athena software [16].

3. Results and discussions

The structure of FMCA series of milling time showed in Fig. 1. In the first hour of milling time, all elements still exist as a pure independent raw material XRD pattern of 9 h and 24 h of milling time showed similar pattern, which has AlCu peak at 2θ of 23.45. At 72 h of milling time the single broad phase has not been confirmed yet even though a broader peak has been observed. Then, after 96 h of milling time, alloy nature was confirmed as a single broaden nanocrystalline. Thus, we perform 1 h, 9 h, 24 h, 72 h and 96 h XRD pattern to show structure evolution as a function of milling time. The average crystallite size of 9, 24, 72 and 96 h of milling time estimate using Scherrer formula were about 21, 27, 38 and 14 nm, respectively. At the 96 h stage, only the 220 obviously exists while the other peaks disappears indicating *W*-type disorder [17]. It is contrast with the nanosized ingot FMA, which has *A*2-type disorder [14]. Meanwhile for FCA case, it was reported in cubic Heusler-type structure yet it does not adopt the tetragonal structures as predicted previously [15].

Magnetic saturation and coercivity characterization of FMCA as function of milling time attempted under applied magnetic external field of 15 kOe at room temperature showed in Fig. 2. At first hour of milling time magnetic saturation was 108.9 emu/g or 3.85 μ_B/f.u. In this stage the magnetic saturation suggested mainly from individual element of iron and manganese, which have been confirmed both experimental and theoretical magnetic moment of 2.35 μ_B/f.u. and 0.16 μ_B/f.u., respectively [18]. Another magnetic moment contribution may lead from Fe–Fe, Mn–Mn and Fe–Mn, which known as ferromagnetic interaction [11]. In accordance with the XRD pattern, from 24 h to 72 h of milling time the AlCu peak was continuously diffused in the Fe–Mn matrix. After 96 h, when the single broad peak has been performed, then the solid solution became stable and the magnetic saturation increased. This magnetic saturation result was similar to other reported works on $(Fe_{60}Al_{40})_{80}Cu_{20}$ [19]. Dynamics magnetic saturation indicated alloying process among those elements to construct a stable structure. As one can see in Fig. 3 the magnetic hysteresis at 5 K obtained the magnetic saturation of 113.5 emu/g or 4.02 μ_B/f.u. after 96 h which in this stage confirms in a broader peak single phase and follow SP rule.

As a parental study, the magnetic moments of FCA was 3.30 μ_B/f.u. did not follow the generalized SP rule [15]. Meanwhile, the magnetic moments nanosized ingot FMA [14] was estimated around 40 emu/g or 1.39 μ_B/f.u. which also close to 1.58 μ_B/f.u. and 1.32 μ_B/f.u. as polycrystalline ingot and ribbon, respectively [12,13]. Our magnetic saturation is about more than twice higher than nanosized ingot FMA [14] even slightly lower than that FCA [15]. In this work, it is revealed that replacing Mn concentration with Cu significantly increases magnetic saturation for any fabrication of FMA reported [12–14]. It could be understood because introducing Cu could reduce Fe–Mn and Mn–Mn coupling which has anti ferromagnetic interaction, meanwhile the ferromagnetic of Fe–Fe and Fe–Mn coupling [11] might still exist. Therefore, in current work it was revealed that Cu may enhance the saturate magnetization in Heusler alloy. Since the FMCA has *VEC* integer number of 28 then, in generalized SP point of view, at 5 K current work coincidently obeys the theoretical prediction compared to FMA and FCA.

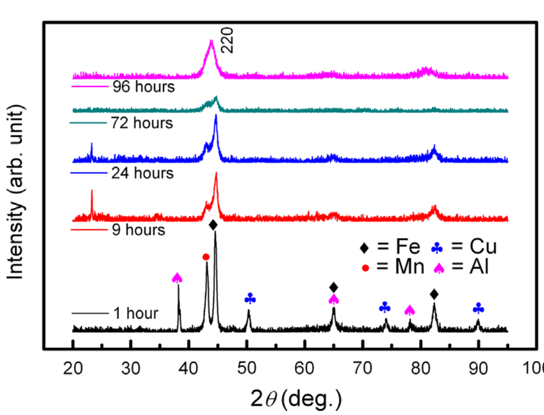

Fig. 1. XRD pattern of nanocrystalline $Fe_2Mn_{0.5}Cu_{0.5}Al$ as function of milling time.

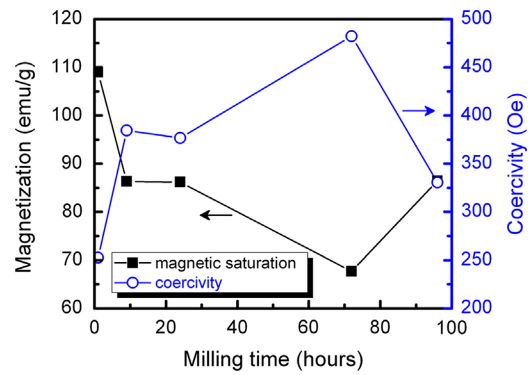

Fig. 2. Magnetic saturation and coercivity of nanocrystalline $Fe_2Mn_{0.5}Cu_{0.5}Al$ as function of milling time.

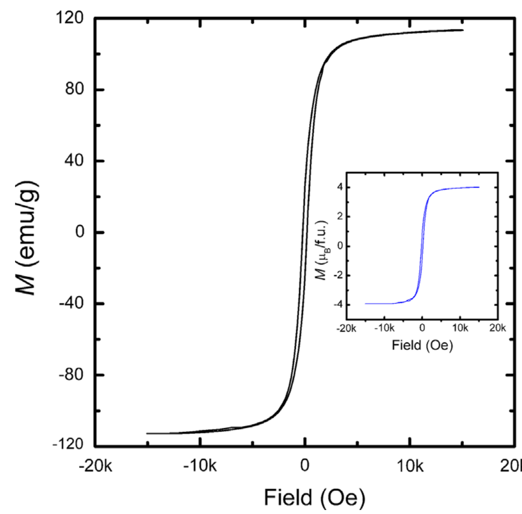

Fig. 3. Magnetic hysteresis of nanocrystalline $Fe_2Mn_{0.5}Cu_{0.5}Al$ after 96 h of milling time measured at 5 K. Inset is the same magnetic hysteresis in units of μ_B/f.u.