

The structural and magnetic properties of Fe-Si and Fe-C solid solutions as a function of milling times

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We have studied the formation of metal-metalloid alloys for Fe-Si and Fe-C as a function of milling times. These alloys were produced using mechanical alloying. The effect of milling time on local structural changes of Fe-Si and Fe-C has been investigated by means of Mössbauer spectrometry, EXAFS study and XRD. Saturation magnetization was also measured by VSM. XRD pattern from mechanically alloyed Fe-Si and Fe-C powders indicates the formation of solid solution. The different variation of lattice parameters could be analysed from the different substitution of Si and C atoms into the Fe structure. The Mössbauer spectrum showed typical sextets in the 1 h milled sample corresponding to alpha-Fe spectrum. Increasing the milling time, the sextets became broader due to appearance of disordered Fe atoms in both solid solutions. The hyperfine field distributions were decreased as increasing milling time, which is similar trend with magnetization distribution. © 2004 Kluwer Academic Publishers

1. Introduction

Metal-metalloid systems such as Fe-Si and Fe-C have been extensively studied for application to magnetic and electronic devices, recently [1, 2]. It is important to study the evolution of alloying process in this system to understand the formation of these alloys. Mechanical alloying (MA) has been used widely to prepare metastable phases such as supersaturated solid solution, amorphous phases and nanostructured powders, starting from a mixture of elemental components or intermetallic compounds, in many alloy systems [3–5].

In this work, Fe₈₀C₂₀ and Fe₈₀Si₂₀ alloys were produced by MA. The structural evolution of these alloys was examined by X-ray diffraction (XRD) and Extended X-ray Absorption Fine Structure (EXAFS) as a function of milling time. The magnetic properties were measured by using a vibrating sample magnetometer (VSM) and ⁵⁷Fe Mössbauer spectrometry for comparison of structural variation and magnetic properties. Special attention is paid to model the local structure and local ordering around Fe atoms in these alloy systems using by EXAFS technique and Mössbauer spectrometry.

2. Experiments

Mixtures of appropriate amounts of Fe, C and Si (–80 mesh, 99.9%) powders were mechanically alloyed by using a SPEX 8000 mixer and mill with stainless balls and vial. MA was performed under an Ar atmosphere to prevent oxidation of the alloy. Chosen compositions were Fe₈₀C₂₀ and Fe₈₀Si₂₀ in order to study the influence of metalloid in Fe metal based system on the structural and magnetic properties. The balls to powder weight ratio was 6:1. The milling time varied from 1 to 24 h.

Structural change for prepared samples was examined by X-ray diffraction (XRD) and EXAFS spectroscopy. XRD traces were obtained with a monochromatic Cu-K_α radiation. EXAFS experiments were carried out at the beam line 3C1 EXAFS of the Pohang Light Source (PLS) in the Pohang Acceleratory Laboratory (PAL) in Korea. The PLS was operated with an electron energy of 2.5 GeV and the maximum current of 220 mA. The magnetic properties of the powdered alloys milled for different times were studied by vibrating sample magnetometer (VSM) and Mössbauer spectrometry at room temperature.

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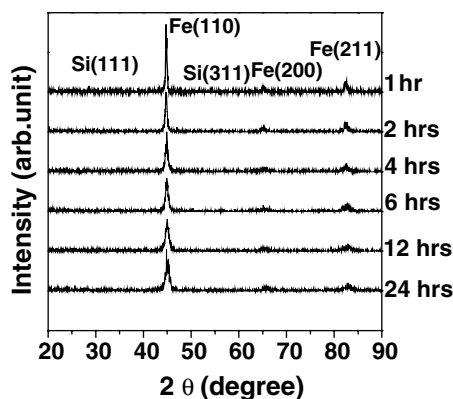


Figure 1 XRD patterns of mechanically alloyed $\text{Fe}_{80}\text{Si}_{20}$ for increasing milling time.

3. Results and discussion

Fig. 1 shows XRD patterns obtained from mechanically alloyed $\text{Fe}_{80}\text{Si}_{20}$ powders. The diffraction peaks corresponding to silicon disappeared after 1 h milling while the remaining Bragg peaks from the bcc Fe phase became weaker and broader. It could be assumed that the amorphization of the silicon occurs and deformation of Fe structure is started as undergoing fracture and welding, repeatedly. As increasing milling time, bcc Fe peaks further become weaker and broader and then (200) and (211) peaks are disappeared after 6 h milled sample. This means that the silicon atoms diffuse into the Fe structure and then form a bcc solid solution [6]. After 12 h milling time, XRD pattern has only the (110) peak and no more changes in the intensity and width, which indicate the formation of bcc solid solution. The mechanically alloyed $\text{Fe}_{80}\text{C}_{20}$ powder also has shown similar results with $\text{Fe}_{80}\text{Si}_{20}$ powder.

The two theta value of Fe peaks are shifted with increasing milling time. We used, both for Fe-Si and for Fe-C, the shift of the dominant (110) peak to determine the variation of the lattice parameter (see Fig. 2). In case of the $\text{Fe}_{80}\text{Si}_{20}$, the lattice parameter increased up to 1 h of the milling time and then decreased steeply. After 6 h, the lattice parameter was not much changed by further milling. From this result, we can assume that diffusion of Si atoms into the Fe structure occurs, say, from 1 h up to 6 h of the milling

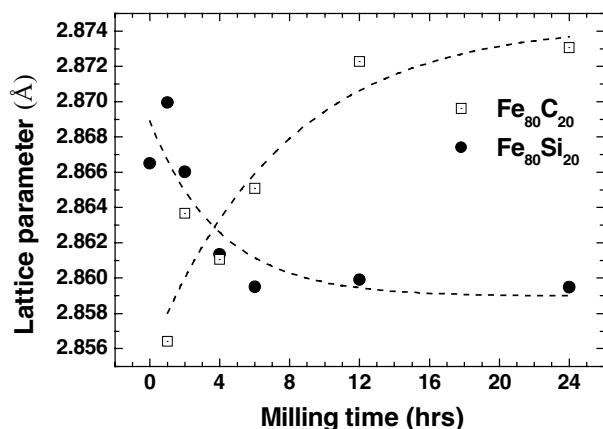


Figure 2 Variations of the lattice parameter of mechanically alloyed $\text{Fe}_{80}\text{Si}_{20}$ and $\text{Fe}_{80}\text{C}_{20}$ with milling time.

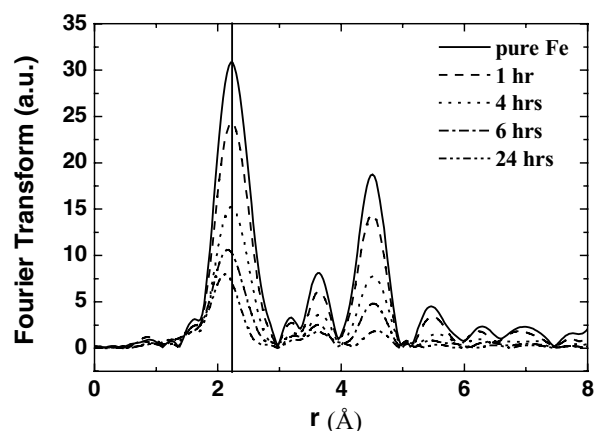


Figure 3 Fourier transform of EXAFS spectra of mechanically alloyed $\text{Fe}_{80}\text{C}_{20}$ for different milling times. The vertical solid line indicates the location of Fe first shell.

time, and the formation of a substitutional solid solution takes place after 6 h of the milling. On the other hand, $\text{Fe}_{80}\text{C}_{20}$ shows an increase of the lattice parameter with increasing milling time. After 24 h, the lattice parameter is increased about 0.23% in comparison to the pure Fe value. Because the carbon atoms interstitially diffuse into the bcc Fe structure, the lattice parameter increase indicates the formation of a solid solution.

Fig. 3 shows the variations of the Fourier transforms of the EXAFS spectra obtained at the Fe K-edge absorption as a function of milling time. The vertical solid line indicates the location of Fe first shell. Also the Fourier transforms of the EXAFS spectra obtained from pure Fe foils at the Fe K-edge absorption was included for comparison. The Fourier transform EXAFS spectrum of the 1 h milled powder is quite the same as that of the pure-Fe spectrum, indicating that the local structure surrounding Fe atoms has a behavior similar to that of pure Fe. However, the peaks in the Fourier transformed spectra decreased as increasing the milling time. In 6 h milling time, the position of the first peak starts to shift to a lower distance which means that the dominant atoms around the Fe central atom start to change. It is good agreement with XRD results due to carbon atoms diffusing into Fe structure with milling times. In 12 h milling time, the shift of first peak position increased and the peak intensity of higher shells above the third shell disappeared. There is no more change from the 12 h processing time indicating that the alloying process has been completed.

Fig. 4 shows the magnetization variation of $\text{Fe}_{80}\text{Si}_{20}$ and $\text{Fe}_{80}\text{C}_{20}$ alloys with milling time. Both alloys exhibit a reduction of the magnetization with increasing milling time. The magnetization of the as mixed $\text{Fe}_{80}\text{Si}_{20}$ and $\text{Fe}_{80}\text{C}_{20}$ powders was about 174 emu/g. For these alloys, the magnetization decreased rapidly in the initial stage of mechanical alloying. Only slight changes occur after 6 and 12 h in case of $\text{Fe}_{80}\text{Si}_{20}$ and $\text{Fe}_{80}\text{C}_{20}$ powders, respectively. With increasing milling time, welding and fracture of the powder particles occurs repeatedly. Thus, interdiffusion of the Fe and the metalloid (Si or C) increases, resulting in magnetic dilution, i.e., decrease of the magnetization. The steady

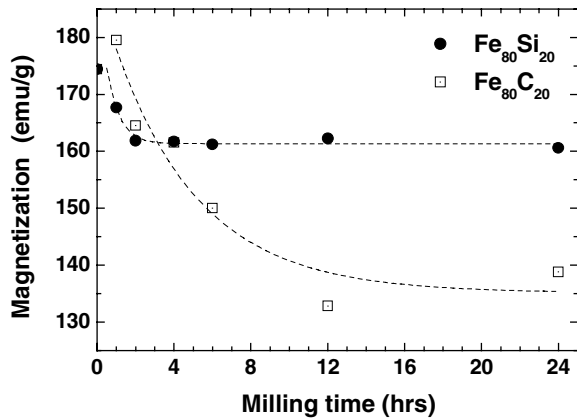


Figure 4 Variation of the magnetization of mechanically alloyed Fe₈₀Si₂₀ and Fe₈₀C₂₀ powders with milling time.

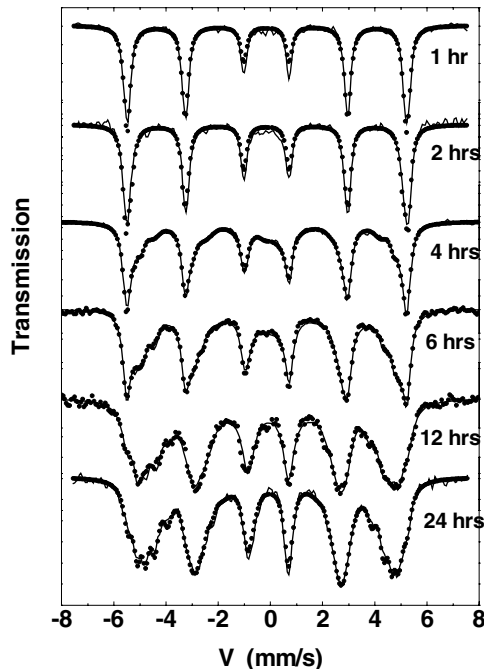


Figure 5 Mössbauer spectra of mechanically alloyed Fe₈₀Si₂₀ powders for different milling times.

state, as shown in Fig. 4, indicates that the solid solution formed. The variation of magnetization corresponds to the results of the XRD and EXAFS structural analysis.

Fig. 5 shows Mössbauer spectra obtained on Fe₈₀Si₂₀ powders, processed for different milling times ranged from 1 to 24 h. The hyperfine structure consists of magnetic sextets composed of lines which progressively broaden when the milling time increases, suggesting the occurrence of an increasing atomic disorder around Fe nuclei. Mössbauer spectra obtained from Fe₈₀C₂₀ powder show similar trend as Fe₈₀Si₂₀ powder. This result indicates that the milling time favours the incorporation of Si or C atoms into the α -Fe phase to form solid solution. We can confirm the formation of solid solution as hyperfine field distribution with milling time (see Fig. 6). The reduction of the average value of hyperfine

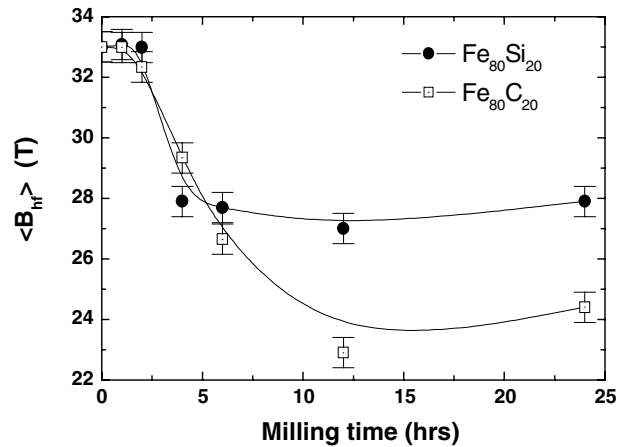


Figure 6 Average value of hyperfine field of mechanically alloyed Fe₈₀Si₂₀ and Fe₈₀C₂₀ powders as a function of milling time.

field with increasing milling time exhibits similar trend with variation of the magnetization in both alloys.

4. Conclusion

Solid solutions of Fe₈₀Si₂₀ and Fe₈₀C₂₀ alloys have been prepared by MA starting from elemental components. Structural and magnetic properties have been determined as a function of the milling time. XRD profiles and lattice parameter indicate the formation of solid solution with increasing milling time. The EXAFS and Mössbauer studies revealed local structural properties of the solid solution. According to these results, we could assume that Si or C atoms diffuse into Fe structure with increasing milling time which gives rise to formation of solid solution. The hyperfine field distributions were decreased as increasing milling time, which is similar trend with magnetization distribution.

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