

MÖSSBAUER STUDY OF THE MATERIAL OF THE ELECTRIC POWER TRANSFORMERS

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Fe-Si alloy is a soft magnetic material used as the core of electric power transformers. One method of molding the core is to press the powdered alloy into the shape of the core. In our study, the X-ray diffraction measurements performed on this processed material, we observed that the peak intensity of ordered phase Fe₃Si was decreased in the pressed sample. The amount of ordered phase is important for the magnetic material. We performed ⁵⁷Fe Mössbauer spectroscopy to elucidate this decrease in the ordered phase. The obtained Mössbauer spectra do not show change with or without pressing. This result indicates that the amount of the ordered phase does not decrease in atomic micro-environments upon pressing. The decrease of the peak intensity in X-ray diffraction can be associated with the distortion or deformation of the ordered phase particles. This consideration is also consistent with the FWHM of the Mössbauer spectrum being slightly broadened on the pressed sample.

Keywords: ⁵⁷Fe Mössbauer spectroscopy, Fe-Si, soft magnetic material, powder molding process

1. Introduction

Fe-Si alloys exhibit soft magnetism and have the characteristic that their magnetostriction is reduced by adjusting the Si concentration, so they are used as materials for the cores of transformers. A transformer core may have a complicated shape to prevent the escaping of the magnetic field from the coil, and powder molding is one of the techniques for producing a core with such a shape. This technique uses powdered materials put into a mold and pressed. In studying the magnetic properties of this process, it was observed that the peak intensity of the ordered phase decreased after pressing in X-ray diffraction. Two types of ordered phases of $D0_3$ structure and B2 structure and a disordered phase of bcc structure exist in the Si concentration region used as the core material [1]. The percentages of these phases change the magnetic properties. To elucidate the reason for this decrease, the ratio of ordered and disordered phases was measured by Mössbauer spectroscopy.

Mössbauer spectroscopy of Fe-Si alloys has been performed [2-7]. It has been observed that components showing different internal magnetic fields depend on the number of Si atoms in the nearest or second-nearest neighbor sites. Thus, Mössbauer spectra can discuss ordered phases in a local and microscopic range. It provides different information from X-ray diffraction, in which peaks are caused by interference from many atoms.

2. Experimental Procedure

The Fe-Si powder samples were prepared by atomizing process with nitrogen gas. Base alloys were Fe-6.5wt%Si and Fe-5.0wt%Si. From past experiments, we know that the former includes ordered phases, and the latter does not. The powder samples were annealed at 850°C for 3 hours for homogenization. These samples were the unpressed samples. A part of the Fe-6.5wt%Si powder was pressed at 833 MPa and then annealed at 750°C for 0.5 hours preferred as the pressed sample.

^{57}Fe Mössbauer measurements were performed at room temperature using the conventional Mössbauer spectrometer. 28 ~ 34 mg of sample and about 100 mg of boron nitride were mixed and put in the sample holder with a diameter of 10 mm for Mössbauer

transmission measurement. α -Fe was used for velocity calibration. The Mössbauer spectra were fitted with a MossWinn program [8, 9].

3. Results and Discussion

Figure 1 shows scanning electron microscope (SEM) images of the powder samples. Almost all particles were spherical in the unpressed sample. The pressed sample is also almost spherical, but there are particles with a part of the surface flattened and particles with dents formed by small particles embedded. These changes observed in morphology can indicate that the particles are deformed by pressing.

Figure 2 shows the X-ray diffraction patterns of the samples, and table 1 shows the peak intensity ratio. The peak intensities are normalized by the intensity of (220) peak. In the Fe-5.0wt%Si sample, neither the (111) peak attributed to the $D0_3$ structure nor the (200) peak attributed to the B2 structure was observed, and only the (220) peak attributed to the bcc structure was observed. In the Fe-6.5wt%Si sample, the (111) and (200) peaks were observed in both unpressed and pressed samples. In addition, the peak of the ordered phase in the pressed sample is smaller than the unpressed. This result suggests that the ordered phases in the Fe-6.5wt%Si sample were reduced by pressing.

Figure 3 shows Mössbauer spectra of Fe_3Si ordered phase sample ($D0_3$), Fe-6.5wt%Si sample (unpressed, pressed), and Fe-5.0wt%Si sample, and table 2 shows the Mössbauer parameters. The spectra of the Fe-Si sample can be analyzed with four components whose internal magnetic field 34~24T (component d1 - d4) and two components with almost the same parameters as the Fe_3Si ordered phase sample (component o1, o2).

The former four components are derived from the Fe-Si disordered phase of the bcc structure [4-7]. From the previous work, it is known that the composition is divided according to how many Si atoms are present in the nearest or the second-nearest neighbor of Fe atoms. Component d1 has no Si atoms in the nearest and the second neighbors. Component d2 has one Si atom in the nearest neighbors or no Si atom in the nearest neighbors and 1-6 Si atoms in the second-nearest neighbors. Component d3 has two Si in the nearest neighbors, and component d4 has three Si atoms in the nearest neighbors.

Considering these, the area ratios of the components are roughly the same as the number of atoms ratios in each Si concentration.

Although the ratio of the D0₃ ordered phase is very small, the 2nd and 5th peaks of component o2 do not overlap with other peaks, so that the ratio can be determined based on these. The existence of the B2 structure was also confirmed by X-ray diffraction. Still, since the Mössbauer spectrum of the B2 structure overlaps with the disordered phase, it was impossible to separate the component of the B2 phase, which is expected to contain several percent.

In the Mössbauer spectra, the ratios of the ordered phase components are unchanged before and after pressing. Microscopically, the amount of ordered phase does not change. There is no deformation-induced phase transformation in this process. However, this result is not consistent with the peak intensity change of X-ray diffraction.

From the SEM images, the sample particles were distorted by the pressing. X-ray interference reduces due to the distortion and deformation of ordered phase particles due to processing. Thus, the intensities of peaks on the X-ray diffraction decrease. The linewidth on the Mössbauer spectra of the pressed sample is broader than that of the unpressed sample. This result indicates that processing distortion remains and is consistent with this consideration.

4. Conclusion

We used Mössbauer spectroscopy to investigate the reason for the decrease found in the relative intensity of peaks of the ordered phases in the X-ray diffractograms of the pressed Fe-6.5wt%Si powder samples, compared to the unpressed ones. The obtained Mössbauer spectra do not show change with or without pressing. This result indicates that the relative amount of the ordered phase does not decrease in atomic micro-environments upon the pressing. The decrease of the peak intensity in X-ray diffraction can be attributed to the distortion or deformation of the ordered phase particles. This consideration is also consistent with the slight line broadening observed in the corresponding Mössbauer subspectra of the pressed sample. Our results can be useful for powder molding processes used for producing the core of Fe-Si transformers.

Declarations

Ethical Approval

This declaration is not applicable.

Competing interests

The authors have no relevant financial or non-financial interests to disclose.

Author's contribution

All authors contributed to the conception and design of this study and approved the final manuscript.

Funding

There is no special funding for this study.

Availability of data and materials

Data sharing is not applicable to this article as no new data were created or analyzed in this study.

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Tables

Table 1 The peak intensities of the X-ray diffraction. The intensities were normalized by the intensity of (220) peak. The (111) peak is attributed to D0₃ structure, the (200) peak is attributed to B2 structure, and the (220) peak is attributed to bcc structure.

	(111) peak	(200) peak	(220) peak
Fe-6.5wt%Si Unpressed	0.40%	0.40%	100%
Fe-6.5wt%Si Pressed	0.10%	0.20%	100%
Fe-5.0wt%Si Unpressed	0%	0%	100%

Table 2 Mössbauer parameters of Fe₃Si, Fe-6.5wt%Si unpressed, Fe-6.5wt%Si pressed and Fe-5.0wt%Si unpressed sample.

		Ratio	IS(mm/s)	H _{in} (T)	QS (mm/s)	FWHM(mm/s)
Fe ₃ Si (D0 ₃)	Component o1	36.7%	0.08	31.3	0.01	0.27
	Component o2	63.3%	0.25	20.1	0.00	0.27
Fe-6.5wt%Si Unpressed	Component d1	14.1%	0.04	33.7	-0.08	0.48
	Component d2	38.2%	0.04	31.6	0.03	↑
	Component d3	27.2%	0.10	28.4	0.02	↑
	Component d4	14.2%	0.19	23.9	0.02	↑
	Component o1	1.6%	0.08*	31.3*	0.01*	0.30*
	Component o2	4.7%	0.25	19.1	0.00	↑
Fe-6.5wt%Si Pressed	Component d1	14.4%	0.04	33.7	-0.07	0.52
	Component d2	38.7%	0.04	31.6	0.03	↑
	Component d3	27.7%	0.10	28.3	0.02	↑
	Component d4	13.7%	0.19	23.9	0.02	↑
	Component o1	1.4%	0.08*	31.3*	0.01*	0.30*
	Component o2	4.1%	0.25	18.9	0.00	↑
Fe-5.0wt%Si Unpressed	Component d1	30.2%	0.03	33.4	-0.03	0.49
	Component d2	40.4%	0.05	31.0	0.03	↑
	Component d3	22.0%	0.11	27.7	0.01	↑
	Component d4	6.4%	0.17	23.6	-0.01	↑
	Component o1	0.3%	0.08*	31.3*	0.01*	0.30*
	Component o2	0.8%	0.25	18.9	0.00	↑

Figures and Figure captions

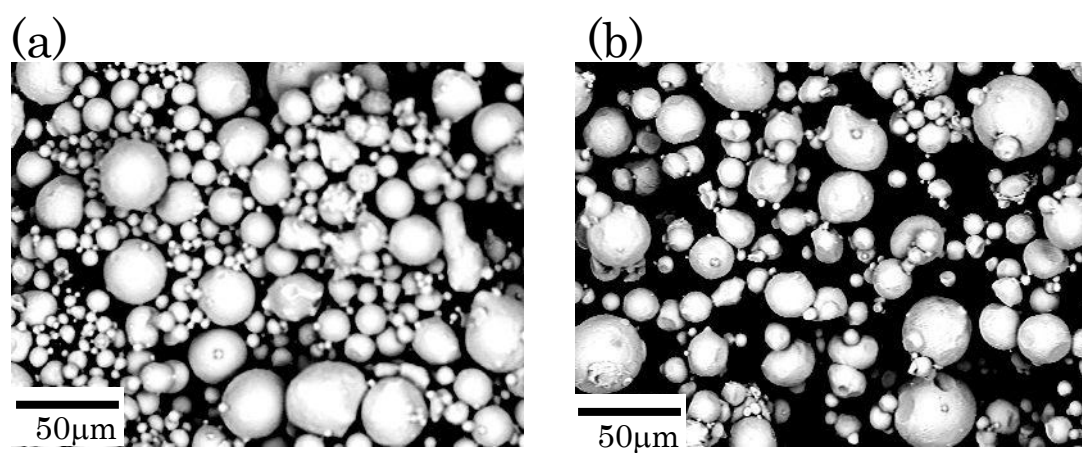


Fig. 1 Scanning Electron Microscope (SEM) images of the (a) unpresseed and (b) pressed sample.

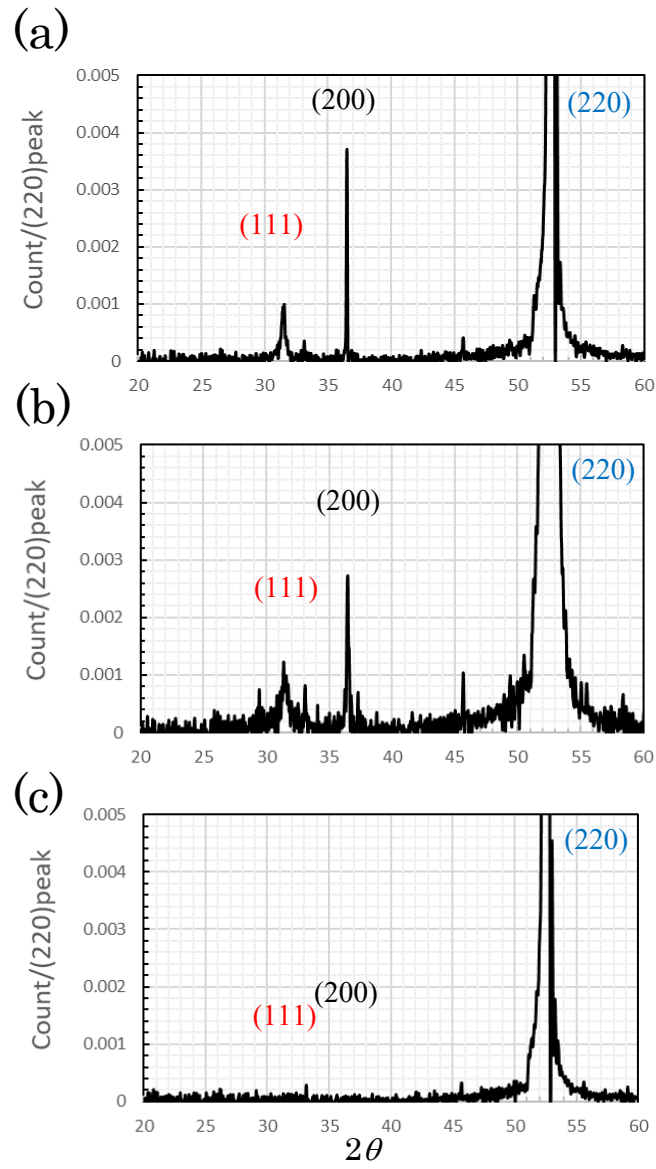


Fig. 2 X-ray diffraction patterns of (a) Fe-6.5wt%Si unpresse, (b) Fe-6.5wt%Si pressed and (c) Fe-5.0wt%Si unpresse sample.

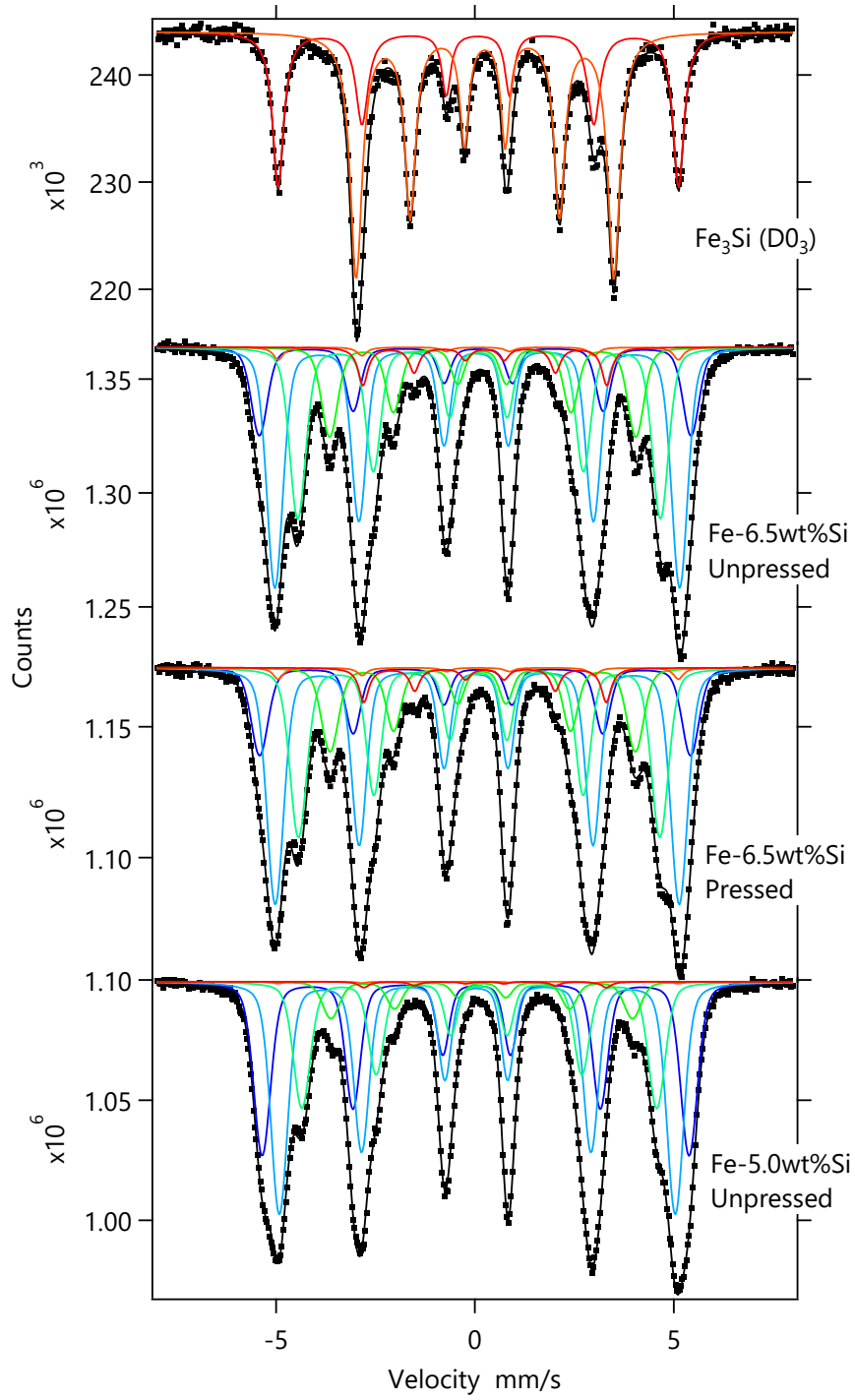


Fig. 3 Observed (closed square) and fitted ^{57}Fe Mössbauer spectra of Fe_3Si , Fe-6.5wt%Si unpressed, Fe-6.5wt%Si pressed and Fe-5.0wt%Si unpressed sample.