P12

電磁浮遊法による無容器凝固した Fe-Ga 合金の構造と磁歪特性

Structural and magnetostrictive properties of containerless solidified Fe-Ga alloys by electromagnetic levitation

村田嶺 [1], 渡邉匡人 [1]
Ryo MURATA[1], Masahito WATANABE[1]
[1] 学習院大学，Gakushuin University,

1. Introduction

Ferromagnetic materials have a property called magnetostriction, which deforms in a magnetic field. In recent years, vibration power generation using this magnetostriction has attracted attention, and the development of materials with large magnetostriction has been actively pursued. Among materials with large magnetostriction of iron-based materials, the binary system of Fe-Ga has attracted attention[1]. Substitutionally doping the body-centered cubic (bcc) phase of iron (α-Fe) with non-magnetic Ga atoms greatly enhances magnetostriction; the magnetostriction increases rapidly under a low magnetic field around 19 and 27 at.% Ga, respectively[2,3]. Fe-Ga alloys containing around 20 at.% Ga has a stable A2 (bcc) structure, therefore a single crystal can be grown for the compositions. Using the single crystals, the enhanced magnetostrictions at 19 at.% has been detailed studied and found the relation to the pseudoelasticity[4,5]. However, Fe-Ga system for the compositions from 25 to 30 at.% has a complexity phase diagram containing B2 (FeGa), $D0_3$ (Fe_3Ga), and $L1_2$, $D0_{19}$ of ordered Fe_3Ga structure phases. Therefore, for the enhanced magnetostriction of Fe-Ga at the compositions of 27 at.% it is not clearly understood the origin of large magnetostrictions. The enhanced magnetostriction behavior in Fe-Ga alloys at this composition would be associated with structural instabilities. Recently, Ikeda et al. reported the new equilibrium and metastable phase diagrams from the annealing experiment with concentration gradient ethod[6]. They show $L1_2$ structure phase existing in the compositional range from 27 to 30 at.% at the temperature around up to 690℃, which is a wide range rather than the previously reported range from 27 to 28 at.%. This was confirmed in recent another report[7]. Moreover, their metastable phase diagram shows a single $D0_3$ structure phase existing in the composition from 25 to 30 at.% at the temperature around up to 650℃. Based on the metastable phase diagram, we will directly obtain the metastable single phase of $D0_3$. Also based on the new equilibrium phase diagram the $L1_2$ phase could be obtained from the same compositions of a metastable phase of $D0_3$. If it will succeed to obtain $L1_2$ and $D0_3$ structures with same compositions, it can compare the properties of magnetostriction of Fe-Ga alloys compositions 19 and 27 at.%. From these backgrounds, we have tried to directly synthesize the

27 at.% Ga composition of Fe-Ga alloy with single $D0_3$ structure using the containerless solidification with rapid cooling by electromagnetic levitation (EML). In this research, our purpose is to confirm the effectivity of the direct synthesis method of Fe-Ga alloys of the composition of 18 and 27.5 at.% Ga by the EML and also to confirm the measurement method of the magnetostriction. In this report, our method to directly synthesize Fe-Ga alloys by a containerless method by EML is described in detail and the magnetostriction is discussed with the structure measurements.

2. Experiment

2.1 Preparation of Fe-Ga alloys by electromagnetic levitation

To achieve directly synthesize Fe-Ga alloys of 27.5 at.% Ga composition with a metastable $D0_3$ structure, the EML method shown in **Figure 1** was used. The electromagnetic levitator was operated by an alternating current of 40A with 490kHz for the generation of alternating cusp shape magnetic fields in two coils with opposite winding, respectively. The alternating magnetic fields generate the eddy current in the metallic samples, and then the Lorentz force by the magnetic field and the eddy current act to levitate the metallic samples against gravity. The coil system and sample holder and its changer were installed in the vacuum chamber filled with Ar-3%H_2 gas at 0.2MPa after the evacuation to 10^{-2} Pa. The sample temperature was monitored by a single-color pyrometer of 900nm with an emissivity one. After experiments, the temperature was calibrated using the melting temperature of Fe from the recalescence phenomena by the release of latent heat of solidification from the undercooled liquid. Using the electromagnetic levitator, we directly synthesized the Fe-Ga alloys following procedures. For EML, the sample temperature after melting rapidly increased to too high temperatures resulting in significant volatilization losses and compositional changes. Therefore, as Ga has a lower vapor pressure than Fe, Ga was added more than the targeted composition in the experiments. Also, if several pieces of metal samples were levitated at the same time, the position of each piece becomes unstable by these interactions and results in the samples being dropped off from the field of acting the Lorentz force. We, therefore, synthesized the Fe-Ga alloys by the levitation drop contacting method with the attaching Ga drops to the firstly levitating Fe drop. A Fe piece and a Ga piece were separately set on the sample holders made by BN, and at first, a Fe piece was levitated and melted at the coil's gap position stable. At the coil's gap positions, Fe drops temperature was kept constant at close to the melting temperature of Fe using He gas. Then a Ga piece was moved into the coils and melted on the sample holder. Moving Ga drop on the sample holder up to the coil's gap, Ga drop was attached to the levitated Fe drop, and the Ga drop was elevated and dissolved into Fe drops by the interfacial tension between them. The connected Fe and Ga drops were stirred uniformly by the electromagnetic force, and then they were cooled by He gas flowing directly to solidify. During cooling for solidifications, He gas flowing rate and decreasing the alternating current to the coils were controlled to the adjusted cooling rate, and solidified Fe-Ga drop was collected on the sample holder. Using the method, we succeeded to obtain the Fe-Ga alloys with the composition of 18 at.% and 27.5 at.% Ga with the different cooling rates.

2.2 Structure and magnetostriction measurement

The Fe-Ga alloy spherical specimens of a diameter of about 8mm elongated to the gravity directions small obtained by the above method were divided into three parts shaped disk with the thickness of 2mm. The disc cut out in the middle was used for magnetostriction measurements, while the other two were used for crystal structure measurement by the x-ray diffraction (XRD), metalogical structure and compositions measurements by the scanning electron microscope (SEM) and the energy dispersed x-ray analysis (EDX). The magnetostriction measurements were used a strain gauge (KFGS-02-120-C1-16, Kyowa Dengyo Corp.) attached to the surface of the disc-shaped sample by a special adhesive. The strain was measured under the applied magnetic fields of 0-1.8 T. The measurement was carried out with the strain gauge and the magnetic field parallel to each other as shown in **Figure 3**. To consider the influence of the magnetic field on the Fe-Ga alloys solidifications by the electromagnetic levitation, the magnetostriction was measured for the two different orientations of the samples in the gravity and in horizontal directions respectively during the solidifications.

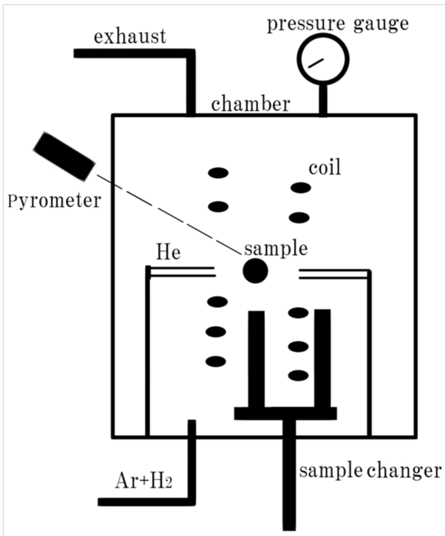

Figure 1 Schematic diagram of electromagnetic levitation system.

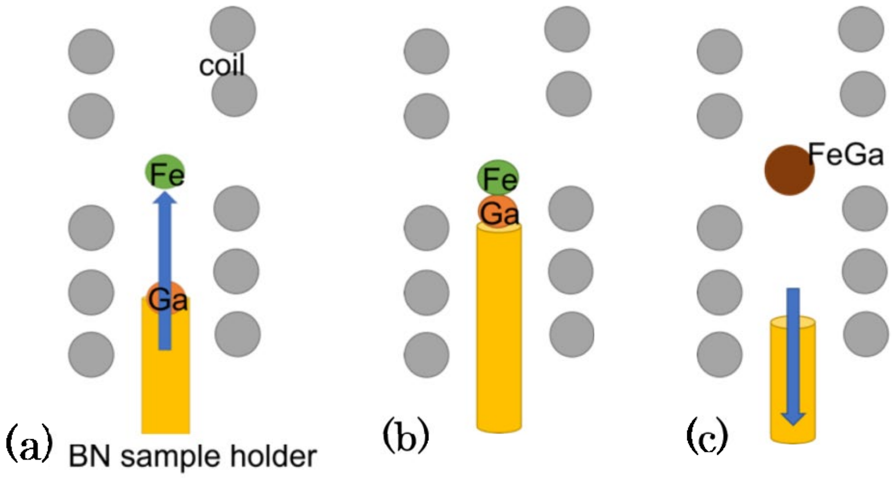

Figure.2 Movement of the specimen table during the preparation of Fe-Ga alloys.

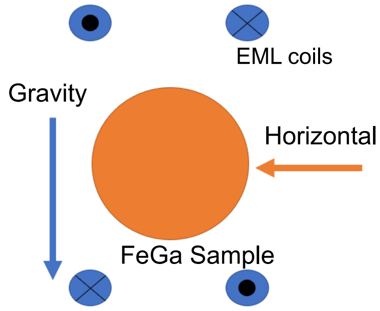

Figure.3 Definition of sample orientation for magnetostriction measurement.

3. Result and discussion

We succeeded in directly synthesizing Fe-Ga alloys of the composition of 18 and 27.5 at.% Ga using the containerless method by EML. **Figure 4** shows the SEM images of the as solidified specimens of Fe-Ga alloys for (a) 18at.% Ga and (b) 27.5 at.% Ga cut vertically the specimens to the gravity direction during solidifications. For these SEM observations, we analyzed these samples' compositions by EDX with the standard calibration method. From the EDX analysis, we found these specimens' compositions almost desired them, therefore we confirmed that our direct synthesis method using EML can be used to study the Fe-Ga alloys magnetostriction by changing the compositions. For the SEM observations, we also found the differences in microstructure of Fe-Ga alloys between 18 at.% and 27.5 at.% Ga compositions. For the composition of 18 at.% Ga, small dendrite patterns are found, on the other hand for the composition of 27.5 at.% Ga large and uniform size grains are found. Moreover, for both compositions, XRD preliminary results show the peak patterns of bcc. From these results and the phase diagram proposed by Ikeda et al., for the composition of 18 at.% Ga A2/$L1_2$ structure phase appeared and for the composition of 27.5 at.% Ga a single $L1_2$ structure phase appeared. We need a more detailed and precise structure analysis by XRD to determine the structure phases.

The magnetostriction results for both compositions are shown in **Figure 5 :** (a)18 at.% Ga and (b) 27.5 at.% Ga. The magnetostrictions were measured For the composition of 18 at.% Ga in **Figure 5 (a)**, magnetostriction has a sharp peak for a maximum of about 80ppm at around 300mT and at over 400mT magnetostriction is averaged for 60ppm. For the composition of 27.5 at.% Ga in Figure 4(b), magnetostriction has no peaks and is averaged for 60ppm at over 300mT. The sharp peaks in the 18 at.% Ga case attributes the solidification structure of two crystalline structure A2 and $L1_2$ phases. In Figure 4(a), small dendrite pattens are observed. Since the magnetostriction depends on the crystalline structure and orientation, for the two crystalline structures of A2 and $L1_2$ the magnetostriction of oriented A2 structure around at 300mT is dominated by the typically oriented crystalline and as a result, sharp peaks appear. Comparing the magnetostriction of the composition of 18 at.% and 27.5 at.% Ga, at over 300mT average magnetostriction for both compositions is the same of 60ppm, this means that for both compositions the solidifications phase would be the same. From structure analysis, for the compositions of 18 at.%, A2 structure segregated in main phase of $L1_2$ is predicted, and for the compositions of 27.5 at.%, A2/$L1_2$ or A2/$D0_3$ structure phase is predicted. Combining the magnetostriction results, we expect that for Fe-Ga alloys of the compositions of 18 at.% Ga a single A2 structure phase appears and for that of the compositions a 27.5 at.% Ga A2/$L1_2$ phase appears. The obtained samples showed the largest strain in the low magnetic field regions, which indicates that the magnetostriction in the non-Julian region is very high[9)].

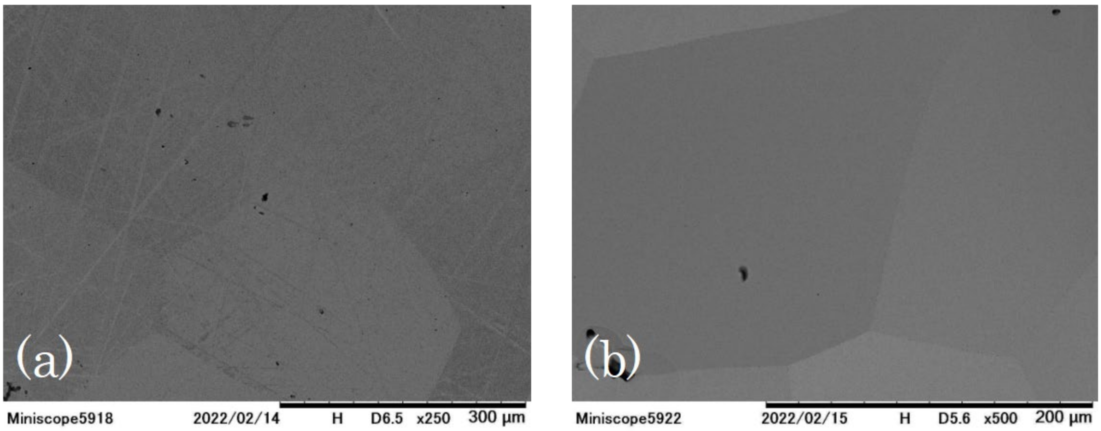

Figure.4 SEM images of direct synthesize Fe-Ga alloys by EML: (a) 18 at.% Ga compositions and (b) 27.5 at.% Ga composition, respectively.

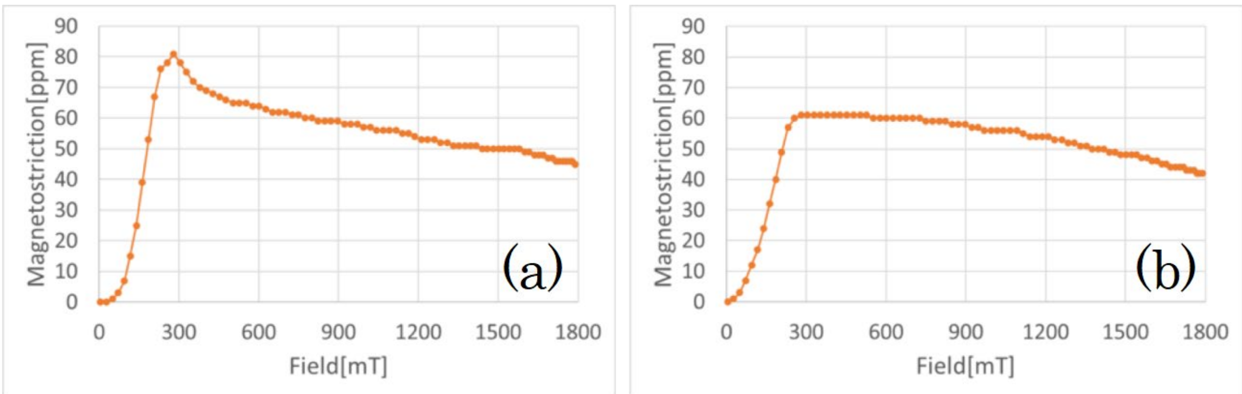

Figure.5 Magnetostriction of Fe-Ga alloys: (a) 18 at.% Ga compositions and (b) 27.5 at.% Ga composition, respectively.

4. Conclusion

We succeeded in directly synthesizing the 18 and 27.5 at.% Ga compositions of Fe-Ga alloy using containerless solidification with rapid cooling by EML. We confirmed the effectivity of the direct synthesis method of Fe-Ga alloys of the composition of 18 and 27.5 at.% Ga by the EML. We also succeeded to measure the magnetostrictions of these specimens. A single phase of bcc structure for the compositions of 18 at.% of Ga was obtained by our synthesize method and it has a sharp peak in the magnetostrictions at about 300mT. For the compositions of 27.5 at.% Ga case, since a small dendrite pattern was observed in the specimens as solidified and the magnetostriction was no sharp peak and averaged 60ppm over 300mT, we predict appearing A2/L12 structure for the compositions by present conditions. In the future, we would obtain a single phase of $D0_3$ structure by controlling the synthesize conditions. If we succeed to obtain the single phase of $D0_3$ structure, the relations between the magnetostriction and pseudoelasticity of Fe-Ga alloys and we would propose the idea of increasing the magnetostrictions of Fe-based alloys.

References

1) A. E. Clark, J. B. Restorff, M.Wun-Fogle, T. A. Lograsso and D. L. Schlagel: Magnetostrictive properties of body-centered cubic Fe-Ga and Fe-Ga-Al alloys, IEEE Trans. Magn., **36** (2000) 3238, DOI: https://doi.org/10.1109/20.908752.

2) A. E. Clark, K. B. Hathaway, M. Wun-Fogle, J. B. Restorff, T.A. Lograsso, V. M. Keppens, G. Petculescu and R. T. Taylor: Extraordinary magnetoelasticity and lattice softening in bcc Fe-Ga alloys, J. Appl. Phys., **93** (2003) 8621, DOI: https://doi.org/10.1063/1.1540130 .

3) J. B. Restorff et al.: Tetragonal magnetostriction and magnetoelastic coupling in Fe-Al, Fe-Ga, Fe-Ge, Fe-Si, Fe-Ga-Al, and Fe-Ga-Ge alloys, J. Appl. Phys, **111** (2012) 023905, DOI: https://doi.org/10.1063/1.3674318

4) H. Y. Yasuda, M. Aoki, Y. Umakoshi and A. Takaoka: Pseudoelasticity in Fe_3Ga single crystals, Scr.Mater. **53** (2005) 253, DOI: https://doi.org/10.1016/j.scriptamat.2005.03.032.

5) H. Y. Yasuda,M. Aoki and Y. Umakoshi: Effect of the ordering process on pseudoelasticity in Fe_3Ga single crystals, Aca Mater. **55** (2007) 2407, DOI: https://doi.org/10.1016/j.actamat.2006.11.033.

6) O. Ikeda, R. Kainuma*, I. Ohnuma, K. Fukamichi, K. Ishida: Phase equilibria and stability of ordered b.c.c. phases in the Fe-rich portion of the Fe–Ga system, J. Alloy. Compd., **347** (2002) 198, DOI: https://doi.org/10.1016/S0925-8388(02)00791-0.

7) A.K Mohamed, V.V. Palacheva, V.V. Cheverikin, E.N. Zanaeva, W.C. Cheng, V. Kulitckii, S. Divinski, G. Wilde, I.S. Golovin: The Fe-Ga phase diagram: Revisited, J. Alloy. Compd., **846** (2020) 156486, DOI: https://doi.org/10.1016/j.jallcom.2020.156486

8) M. Yokoyama, M. Matsumoto, K. Takahashi, K. Watanabe: Control of Preferred Orientation in Giant Magnetostriction Alloys under High Magnetic Field, Ann. Rep. High Field Labs. Superconducting Matt. Tohoku Univ., (2009) 5-12 (in Japanese), http://www.hflsm.imr.tohoku.ac.jp/files/0512-09H0083-yokoyama.pdf.

9) H. D. Chopra and M. Wuttig : Non-Joulian magnetostriction, Nature, **521** (2015) 340, DOI: https://doi.org/10.1038/nature14459.