Magnetostriction of heavily deformed Fe-Co binary alloys prepared by forging and cold rolling

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Abstract-

Magnetostriction of $\text{Fe}_{1-x}\text{Co}_x$ ($x=50-90$ at%) alloys prepared by forging and subsequent cold-rolling was studied as functions of alloy compositions and thermomechanical treatments. Magnetostriction of the as-forged $\text{Fe}_{25}\text{Co}_{75}$ alloy was 108 ppm and that of the as-cold rolled $\text{Fe}_{25}\text{Co}_{75}$ alloy measured parallel to the rolling direction (RD) was 128 ppm. The cold-rolled $\text{Fe}_{25}\text{Co}_{75}$ alloy possessed a nearly $\{100\}\langle 011 \rangle$ texture, leading to the maximum magnetostriction of 140 ppm when measured at an angle of 45° to RD. Moreover, the fully annealed $\text{Fe}_{25}\text{Co}_{75}$ and $\text{Fe}_{20}\text{Co}_{80}$ alloys were gradually cold rolled and magnetostriction were measured. Results showed that the magnetostriction of those cold-rolled alloys drastically increased with increasing reduction rate. According to the XRD and TEM observations, intensity of the fcc peak gradually decreased with increasing reduction rate and that the alloys became to be in a bcc single state at a reduction rate higher than 90%, leading to a drastic increase in magnetostriction.

1. Introduction

Magnetostriction is one of the functional properties of ferromagnetic materials, causing their shape/dimensions to change in an applied magnetic field. The use of magnetostrictive materials for actuators, sensors, oscillators and motors has been anticipated. In particular, as thin film of magnetostrictive alloys shows good compatibility with semiconductor manufacturing processes in the field of MEMS devices, much effort has been devoted to development of micro-sized high-sensitive magnetic sensors and energy harvesters by using magnetostrictive materials [1-5]. It is well-known that most ferromagnetic materials show a certain magnitude of magnetostriction. For example, pure iron possesses negative magnetostriction of about -9 ppm and cobalt shows relatively large negative magnetostriction of about -52 ppm [6], the largest among the pure metals.

There are some well-developed magnetostrictive materials [7-9]. Terfenol-D (Tb-Dy-Fe) is known to show giant magnetostriction larger than 2000 ppm [10-12] and TbCo₂-DyCo₂ composite shows magnetostriction of about 800 ppm [13]. Giant magnetostriction of Terfenol-D has been attributable to the magnetic domain rotation which causes internal strain [11]. Terfenol-D is commercially available, but as a single crystal in most cases, and is thus expensive. As for the origin of large magnetostriction of the TbCo₂-DyCo₂ composite, Yang et al. suggested that large magnetostriction of this material was attributable to the abnormal behavior of the magnetic and structural phase transition at the phase boundary between the TbCo₂ and the DyCo₂ phases and that this behavior was similar to the magnification behavior of piezoelectricity at the morphotropic phase boundary observed in the ferroelectric PbZrO₃-PbTiO₃ system (PZT) [13]. Some other giant magnetostrictive materials have also been reported [14]. Furthermore, Fe-Ga alloy possesses a magnitude of magnetostriction of about 400 ppm and is known as Galfenol [15-20]. As clearly seen in the phase diagram of the Fe-Ga binary alloy system, the alloy has a phase boundary between the D0₃ ordered phase (bcc) and the A2 disordered phase (bcc α -Fe) at around the composition range of 15-25 at%Ga. Fe-Ga alloy shows the largest magnetostriction in this composition range. Thus, it is conceivable that the large magnetostriction of Fe-Ga alloy is due to enhancement of the magnetic and structural phase transition such as martensitic B2 $\rightarrow$ L10 transformation [20]. The Fe-Al binary alloy system also has a phase boundary as seen in its phase diagram and it has been reported that large magnetostriction of 200 ppm was obtained at the composition around the phase boundary [21]. Fe-Pd binary alloys can also show large magnetostriction of up to

600 ppm and it has reported that the origin of this large magnetostriction is attributable to rotation of magnetic twins [22-23].

In this study, Fe-Co alloy as an alternative magnetostrictive material was focused on. According to the Slater-Pauling curve, Fe-Co alloy shows the largest number of Bohr magnetons, leading to the largest saturation magnetic flux density among the Fe-, Ni- and Co-based binary alloys [24, 25]. The Fe-Co alloy system is known to possess good magnetic properties, in particular, soft-magnetism [26-29]. Equiatomic Fe-Co alloy ($\text{Fe}_{50}\text{Co}_{50}$) is significantly brittle, so the Fe-Co-2V alloy ($\text{Fe}_{49}\text{Co}_{49}\text{V}_2$), in which a small amount of V is added to Fe-Co alloy in order to improve ductility, was developed and is called Permendur. Among commercially available soft-magnetic materials, the Fe-Co-2V alloy is known to have the most excellent soft-magnetic properties such as the highest saturation magnetic flux density, high permeability and low core loss [29-31]. Magnetostriction values of a single crystalline equiatomic Fe-Co alloy and a polycrystalline Fe-Co-2V alloy have been reported to be 150 ppm (λ_{100}) [32] and 70 ppm [33], respectively. In 1932, Masiyama studied the magnitude of magnetostriction of Fe-Co binary alloys in a wide composition range and reported that magnetostriction heavily depends on alloy composition and two distinct peaks of magnetostriction were clearly observed, the 1st peak being from around $\text{Fe}_{30}\text{Co}_{70}$ (magnetostriction: 90 ppm) and the 2nd one being from around $\text{Fe}_{60}\text{Co}_{40}$ (70 ppm) [34]. The phase boundary between the bcc region and the bcc+fcc coexisting region can be seen at a Co content of about 75 at% in the phase diagram of the Fe-Co binary system [35, 36]. According to the previous report [37], it was said that this phase boundary may enhance the magnetostriction of the Fe-Co alloy in a way similar to enhance the magnetostriction by $\text{D0}_3/\text{A2}$ phase boundary in Fe-Ga alloy, and indeed, the largest magnetostriction was obtained at a Co content of about 70 at% in that report. Although some previous studies on the phase diagram of Fe-Co binary alloy system have been conducted by experiments [35] and computer-simulation [36], an accurate phase diagram is still unclear due to very complicated phase precipitations of A1 phase (fcc), A2 phase (disordered bcc) and B2 phase (ordered bcc) in the composition range of 70-90 at%Co.

Since Hunter et al. [38] reported that thin $\text{Fe}_{34}\text{Co}_{66}$ alloy film showed large magnetostriction of 260 ppm, there has been increased interest in the Fe-Co alloy system as a good candidate for a new magnetostrictive alloy. They concluded that a mixture of the bcc phase and the fcc phase should enhance its magnetostriction. The

magnetic properties of the Fe-Co alloy thin films have been studied for application in MEMS devices [39, 40]. Moreover, if Fe-Co alloy bulk materials actually show large magnetostriction, they can be good candidates for magnetostrictive materials applicable to various actuators and power-generation devices in the future because of their abundance and lower cost as compared with Fe-Ga alloy.

Thus, in this work, Fe-Co binary alloys with Co content higher than 50 at% were prepared in bulky state by forging and subsequent cold rolling which are often adopted in mass production and then the effect of alloy composition and thermo-mechanical treatment on their microstructure and magnetic properties such as magnetostriction and magnetization were studied.

2. Experimental Procedure

Alloy ingots of $\text{Fe}_{1-x}\text{Co}_x$ ($x=50, 55, 60, 66, 70, 75, 80, 85, 90$ at%) weighing more than 7 kg were prepared by a high-frequency furnace. The ingots were heated at 1323 K for 3 h and then forged into alloy bars. The cross-sectional area of ingots was reduced by half during forging. The reduction rate of forging was 50%. Forging was finished after the temperature of the ingots decreased to 1173-1073 K, and then the ingots were air cooled to room temperature at a cooling rate of a few $\text{K} \cdot \text{s}^{-1}$.

Small specimens were cut out of each alloy ingot to study their microstructure and magnetic properties. The specimen dimensions were about $5 \text{ mm} \times 8 \text{ mm}$ and 0.3-2 mm in thickness. Some specimens were heat-treated in vacuum at 873-1073 K for 3 h and cooled in the furnace to study the effect of annealing temperature on magnetostriction.

After forging, some ingots were cold rolled at room temperature from 10 mm to 0.3 mm in thickness in order to compare magnetic properties of the forged alloys to those of the heavily cold-rolled alloys. The rolling reduction rate was 97 %. Then small specimens were cut from those cold-rolled sheets and heat treated in vacuum at 1073 K for 3 h and cooled in the furnace.

Moreover, each small piece of the forged $\text{Fe}_{25}\text{Co}_{75}$, $\text{Fe}_{20}\text{Co}_{80}$ and $\text{Fe}_{15}\text{Co}_{85}$ alloy ingots was heated at 1073 K for 12 h in vacuum and cooled in the furnace. Those fully-annealed pieces with large amount of the fcc precipitates in the bcc matrix were then cold rolled at room temperature from 10 mm to 0.3 mm in thickness. The

rolling reduction rate was gradually increased from 0 to 20, 40, 60, 80, 90, 93, 95 and 97% step by step. Specimens were cut out of the rolled ingots at each step and used for microstructure observation and magnetostriction/magnetization measurements.

Phase identification of the specimens was conducted by X-ray diffraction analysis (hereafter denoted as XRD; Cu-K α , 40 kV and 40 mA). Microstructure observation of the specimens was conducted by scanning electron microscope-electron backscattering diffraction/oriented imaging microscopy (hereafter denoted as SEM-EBSD/OIM) and transmission electron microscopy (hereafter denoted as TEM) analyses.

Non-magnetic type strain gauges (KFL series, Kyowa Electronic Instruments Co., Ltd.) and special bond (M-Bond610) to fix a strain gauge to the specimen surface were used for magnetostriction measurements. Magnetostriction and **saturation** magnetization were measured at room temperature in a magnetic field up to 1.2 T by using a vibrating sample magnetometer (VSM).

The magnetostriction of equiaxed polycrystalline materials can be described by the following equation [6]:

$$\frac{\Delta l}{l} = \frac{3}{2} \lambda_s \left(\cos^2 \theta - \frac{1}{3} \right) \quad (1)$$

l is the length of the specimen. λ_s is the measured value of saturation magnetostriction and θ is the angle between the magnetization direction and the measurement direction. When the magnetostriction is measured parallel to the magnetization direction, that is $\theta = 0^\circ$, equation (1) is simplified as shown below:

$$\frac{\Delta l}{l} = \lambda_s \quad (2)$$

Therefore, **a magnetic field was applied parallel to the measurement direction of the strain gauge in this study.**

3. Results and Discussion

3-1. Magnetostriction of the forged Fe-Co alloys

Figure 1 shows the results of (a) magnetostriction and (b) magnetization measurements of the Fe_{1-x}Co_x (x=50, 55, 60, 66, 70, 75, 80, 85, 90 at%) alloys prepared by forging and subsequent annealing at 873-1073 K for 3 h. The phase diagram of the Fe-Co binary alloy system is superimposed at the upper part of the figure [41].

Magnetostriction of the as-forged alloy specimens was around 60 ppm in the composition range of 50-66 at%Co and increased in the range of 70-75 at%Co. The as-forged Fe₂₅Co₇₅ alloy showed the largest magnetostriction up to 108 ppm. In the composition range of Co content higher than 75 at%, magnetostriction decreased drastically and showed a small negative value. In the case of the specimens annealed after forging, magnetostriction was higher than that of the as-forged specimens in the 50-70 at%Co range. Particularly, in the 60-70 at%Co range, magnetostriction increased with increasing annealing temperature. This may be because accumulated dislocations, strain and internal stress were released and grain growth occurred by annealing. Indeed, in the 50-66 at%Co range, the alloys annealed at 873 K possess the B2 ordered structure. In this range, the alloys annealed at 1073 K also possess the B2 ordered structure in this study because the cooling rate was low due to furnace cooling although the alloys were annealed at 1073 K in the A2 disordered region as seen in the phase diagram. There is no difference in crystal structure between the alloys annealed at 873 K and 1073 K in this study. So, the factors to increase magnetostriction after annealing in this range should be the microstructure change and the release of dislocations which leads to the release of strain and internal stress. The Fe₃₀Co₇₀ alloy annealed at 1073 K showed the largest magnetostriction of 101 ppm. However, the Fe₂₅Co₇₅ alloy is on the bcc/(bcc+fcc) phase boundary, leading to precipitation of the fcc phase during heat-treatment, so the magnetostriction of the annealed Fe₂₅Co₇₅ alloy was smaller than that of the as-forged alloy. From Fig. 1(a), it can be clearly concluded that magnetostriction showed a maximum in the Fe₂₅Co₇₅ alloy at around the bcc/(bcc+fcc) phase boundary and that the magnetostriction decreased significantly and then reached 0 or a small negative value in the composition range of 80-90 at%Co.

Figure 1(b) indicates the magnetization of the forged alloys. Magnetization tended to decrease with increasing Co content monotonically from about 210 emu·g⁻¹ for Fe₅₀Co₅₀ alloy to about 175 emu·g⁻¹ for Fe₁₀Co₉₀ alloy although some scatter is seen. Different from the magnetostriction behavior, magnetization did not change drastically at around the bcc/(bcc+fcc) phase boundary.

Figures 2(a) and (b) show the XRD patterns of the as-forged alloys and the alloys annealed at 1073 K for 3 h after forging, respectively. As clearly seen in Fig. 2(a), the alloys with Co content of 50-75 at% has a single bcc phase while a fcc second phase was slightly precipitated in the Fe₂₀Co₈₀ alloy. The alloys with 85-90 at%Co had a single fcc phase. According to the phase diagram, the coexisting region of the bcc phase and the fcc phase lies in

the composition range of 75-90 at%Co. It was thus clear that magnetostriction of the as-forged alloys with Co content more than 75 at% was very small because of the distinct precipitation of the fcc phase which showed a small negative magnetostriction value. In the case of the alloys annealed after forging shown in Fig. 2(b), the same tendency was obtained but the fcc second phase started to precipitate in the Fe₂₅Co₇₅ alloy. As a result, the annealed Fe₃₀Co₇₀ alloy having a single bcc phase showed the largest magnetostriction among the annealed alloys, as shown in Fig. 1(a). It was found that magnetostriction behavior of these alloy specimens corresponded to the bcc and fcc peak appearance behavior in the XRD patterns and the phase diagram.

Figure 3 shows grain mappings obtained from the forged Fe₂₅Co₇₅ alloy annealed at 1073 K for 3 h, indicating (a) a microstructure and (b) a phase mapping dividing the fcc phase from the bcc phase. In Fig. 3(b), the dark area and the gray area indicate the bcc main matrix and the fcc second phase, respectively. As seen in Fig. 3(a), the average grain size was around 10 μm , and it can also be seen in Fig. 3(b) that an fcc phase was precipitated after annealing along the grain boundaries of the bcc main matrix in the annealed Fe₂₅Co₇₅ alloy.

3-2. Effect of cold rolling on magnetostriction of Fe-Co alloys

Figure 4(a) shows the results of magnetostriction measurement of the Fe_{1-x}Co_x (x=66, 70, 75, 80, 85, 90 at%) alloys prepared by cold-rolling after forging. Magnetostriction of the as-cold rolled alloys was measured both in the rolling direction (RD) and in the direction at the angle of 45° inclined to RD. It was difficult to cold roll specimens of a near-equiatomic composition range of x=50-60 at%Co due to their severe brittleness. Fe₃₄Co₆₆ alloy was barely able to be cold rolled at room temperature, and the Fe-Co alloys with 70 at%Co or higher showed good cold rolling workability up to a reduction rate 97%. Cold-rolled specimens possessed higher magnetostriction than the as-forged specimens in the composition range of 66-80 at%Co and the as-cold rolled Fe₂₅Co₇₅ alloy specimen showed the highest magnetostriction of 128 ppm measured parallel to RD among the as-cold rolled alloys. Furthermore, the as-cold rolled Fe₂₀Co₈₀ alloy specimen showed relatively large magnetostriction of 110 ppm although the as-forged Fe₂₀Co₈₀ alloy specimen showed a very small value.

In this work, it was found that the magnetostriction measured in the direction at the angle 45° inclined to RD was slightly larger than that measured parallel to RD and that the as-cold rolled Fe₂₅Co₇₅ alloy showed the

largest magnetostriction up to 140 ppm at the angle 45° . In the similar case, Sugano et al. (1972) reported that the magnetostriction of hot-rolled Fe-Co-2V Permendur alloy was dependent on the measurement angle to RD and showed its maximum value at the angle 45° [33]. In the case of the specimens cold rolled and subsequently annealed at 1073 K for 3 h, their magnetostriction decreased with increasing Co content in the composition range of 70-90 at%Co.

Figure 4(b) shows the results of magnetization measurement of these cold-rolled alloys. Magnetization tended to decrease with increasing Co content, and detailed observation also revealed that the magnetization of the as-cold rolled alloys measured in the direction at the angle 45° inclined to RD was slightly larger than that measured parallel to RD in the composition range of 66-80 at%Co.

Figures 5(a) and (b) show the XRD patterns of the as-cold rolled alloys and the alloys annealed at 1073 K for 3 h after cold-rolling, respectively. The as-cold rolled alloys with 66-75 at%Co had a single bcc phase as did the as-forged alloys. The as-cold rolled $\text{Fe}_{20}\text{Co}_{80}$ alloy also possessed a single bcc phase while the as-forged $\text{Fe}_{20}\text{Co}_{80}$ alloy possessed both the bcc main phase and the fcc second phase. It was thus conceivable by XRD observations that the fcc phase in the $\text{Fe}_{20}\text{Co}_{80}$ alloy disappeared by severe cold rolling, which means that the bcc/(bcc+fcc) phase boundary shifts to higher Co content. In 1936, Nishiyama reported that the Stainless Invar (Co-36Fe-8.7Cr

alloy) showed the strain-induced martensite transformation from γ phase (fcc structure) to α phase (bcc structure) [42]. So, the Fe-Co alloy with relatively small amount of fcc phase precipitation may show the transformation from the fcc phase to the bcc phase by heavy deformation in a similar manner, resulting in the disappearance of the fcc phase. Therefore, the as-cold rolled $\text{Fe}_{20}\text{Co}_{80}$ alloy was in a single bcc state, with the result that the alloy showed

relatively large magnetostriction of more than 110 ppm. With higher Co content, the as-cold rolled $\text{Fe}_{15}\text{Co}_{85}$ alloy showed distinct peaks originating from the fcc single phase and its magnetostriction decreased drastically, as shown in Fig. 4(a). It seems that the strain introduced into the alloys with 85 at%Co and over was not enough to promote

the martensite transformation, the bcc distinct peaks did not appear in Fig. 5(a). In the case of the annealed specimens shown in Fig. 5(b), the largest magnetostriction of 110 ppm was obtained in the $\text{Fe}_{30}\text{Co}_{70}$ alloy having a bcc single phase. As mentioned above, the magnetostriction of the specimens annealed after cold rolling decreased with increasing Co content in the composition range of 70-90 at%Co. According to Fig. 5(b), this is because those

annealed alloys having 75-90 at%Co are around the bcc/(bcc+fcc) phase region, resulting that the amount of precipitated fcc phase in the alloys increases gradually with increasing Co content.

Besides the XRD observations, the microstructure of the as-cold rolled Fe₂₅Co₇₅ alloy was observed by using the SEM-EBSD/OIM system in order to understand the reasons for the dependence of magnetostriction on the measurement angle to RD.

Figures 6(a) and (b) show the microstructure mapping and the inversed pole figures of the as-cold rolled Fe₂₅Co₇₅ alloy having the largest magnetostriction in this work. The characteristic texture is formed by severe cold-rolling, as clearly shown in Fig. 6(a), which is a typical stripe microstructure obtained by cold rolling with an average grain size of about 2.5 μ m. Figure 6(b) clearly shows the features of the characteristic microstructure obtained in this alloy, namely, a distinct texture with crystallographic low Miller index planes scattering from (101) plane to (112) plane normal to RD, from (001) plane to (111) plane normal to surface (NS) and also from (101) plane to (111) plane normal to both RD and NS. This means that there are a large number of grains whose (101) plane is normal to RD, with the result that there are a number of grains in which the axis of easy magnetization $\langle 100 \rangle$ turns toward the direction at the angle of 45° inclined to RD. Thus, it is conceivable that the magnetization and magnetostriction measured at the angle of 45° to RD are higher than those measured parallel to RD. In previous papers, it has been reported that a heavily rolled equiatomic FeCo alloy formed the {100}<011> texture [43, 44]. In this work, a similar microstructure was obtained after severe cold rolling.

As you can see in Fig. 1(a), the magnetostriction of the forged alloys increased after annealing in the 50-70 at%Co range. However, the cold-rolled Fe₃₄Co₆₆ and Fe₃₀Co₇₀ alloys showed the decrease in magnetostriction after annealing. This may be because the distinct texture formed by heavy cold rolling became broad during heat-treatment and this leads to the decrease in magnetostriction in the cold-rolled and subsequently annealed Fe₃₄Co₆₆ and Fe₃₀Co₇₀ alloys while the accumulated dislocations, strain and internal stress were released and the grain growth occurred during heat-treatment, leading to the increase in magnetostriction in the forged and subsequently annealed alloys.

3-3. Effect of reduction rate on magnetostriction of cold-rolled Fe-Co alloys

Fe₂₅Co₇₅, Fe₂₀Co₈₀ and Fe₁₅Co₈₅ forged alloys were fully annealed at 1073 K for 12 h and then cooled in a furnace. The annealed alloy specimens were cold rolled stepwise from 10 mm (reduction rate 0%) to 8 mm (20%) → 6 mm (40%) → 4 mm (60%) → 2 mm (80%) → 1 mm (90%) → 0.7 mm (93%) → 0.5 mm (95%) → 0.3 mm (97%) in thickness. The XRD observations and the magnetostriction/magnetization measurements were conducted at each reduction rate. As for the magnetostriction measurements, a strain gauge was fixed on the specimen surface parallel to RD when the rolled alloy was thin (≤ 2 mm in thickness). When the alloy was thick (≥ 4 mm), a strain gauge was fixed to a sliced cross-section normal to RD.

Figure 7(a) shows the magnetostriction as a function of the reduction rate of cold-rolling. The magnetostriction of Fe₂₅Co₇₅ alloys with reduction rate up to 60% was almost 70 ppm. With a larger reduction rate, the magnetostriction increased significantly and reached 120 ppm at a reduction rate of 95%. In the case of the Fe₂₀Co₈₀ alloys with a reduction rate larger than 60%, the magnetostriction increased drastically and reached 105 ppm at a reduction rate of 97%. These alloys were found to show a significant increase in magnetostriction by heavy cold-rolling. On the contrary, the Fe₁₅Co₈₅ alloys showed a small negative value regardless of the reduction rate.

Figure 7(b) shows the magnetization as a function of the reduction rate. As can be seen in the figure, the magnitude of magnetization remained almost constant in all the alloys subjected to cold rolling at 0-80% although some scatter was observed. The magnetization thus increased slightly with increasing reduction rate higher than 90% as well as the magnetostriction.

Figure 8(a) shows the variations of the XRD pattern of the gradually cold-rolled Fe₂₅Co₇₅ alloys as a function of the reduction rate. The observation range was $2\theta = 43-47^\circ$. In this 2θ region, two distinct peaks from the bcc (110) plane and the fcc (111) plane were observed. It can be seen in the figure that the intensity of the fcc peak decreased with increasing reduction rate and that the fcc peak almost disappeared after heavy rolling at a reduction rate of 90% and over. Therefore, it is conceivable that the increase in magnetostriction at the higher reduction rate was attributable to both the decrease in the fraction of the fcc phase and to the formation of texture which was shown in Fig. 6.

Figures 8(b) and (c) show the variations of the XRD pattern of the gradually cold-rolled Fe₂₀Co₈₀ and

Fe₁₅Co₈₅ alloys, respectively. Figure 8(b) indicates that the intensity of the fcc peak in the Fe₂₀Co₈₀ alloys as well as that of the Fe₂₅Co₇₅ alloys gradually decreased with increasing reduction rate, leading to a significant increase in magnetostriction. In the case of the Fe₁₅Co₈₅ alloys shown in Fig. 8(c), the bcc peak which should appear at around $2\theta=45.3^\circ$ was not observed and the intensity of the fcc peak at around $2\theta=44^\circ$ did not decrease even after heavy cold-rolling up to 97%. So, it was concluded that the magnetostriction of the cold-rolled Fe₁₅Co₈₅ alloys was always a small negative value because the alloy was in a fcc single state after heavy cold rolling.

Figure 9 shows TEM observations of the Fe₂₀Co₈₀ alloys subjected to cold rolling at reduction rates of 0%, 60% and 97%, the magnetostriction values of which are shown in Fig. 7. Figures 9(a), (c) and (e) are the bright field images and (b), (d) and (f) are selected area diffraction rings with the indices. The grain size was found to decrease with increasing reduction rate and, moreover, a layered texture was formed after severe rolling at 97%. As seen in Fig. 9(b), distinct spots originating from the bcc phase and the fcc phase were observed in the diffraction rings. The intensity of those fcc spots decreased with increasing reduction rate. Finally, the spots for the fcc phase disappeared in the specimen cold rolled at 97%, as shown in Fig. 9(f), in a way similar to that in Fig. 8(b), due to the strain-induced martensite transformation [42].

It should be also noted that the grain sizes estimated by SEM-EBSD/OIM were much larger than the grain size seen directly in the TEM images. Because the measurement accuracy of the SEM-EBSD/OIM depends on the step size by which the pseudo-Kikuchi patterns are obtained from the sample surface.

In some previous papers, magnetostriction of the Fe-Co alloy was reported to be enhanced at around the bcc/(bcc+fcc) phase boundary [37, 38]. Indeed, in this work, the largest magnetostriction was obtained in the Fe₂₅Co₇₅ alloy which was located at around the bcc/(bcc+fcc) phase boundary in the phase diagram. However, the role of the fcc phase in enhancing or degrading magnetostriction is not yet clear. At least, it was found in this study that the distinct appearance of the fcc phase really caused the magnetostriction to deteriorate. Hunter et al. have reported that the volume fraction of the fcc phase was 0.3% in the Fe₃₄Co₆₆ alloy thin film which showed a large magnetostriction of 260 ppm [38]. It seems that detection of the fcc phase with its volume fraction of 0.3% may be difficult by XRD nor by conventional TEM.

At any rate, the magnetostriction of the bulky Fe-Co binary alloys prepared by forging and subsequent

heavy cold rolling was investigated in this study. As results, it was found that $\text{Fe}_{25}\text{Co}_{75}$ alloy which lies just around the two phase boundary (bcc/(bcc+fcc)) showed the largest magnetostriction up to 140 ppm by cold rolling and that the fcc phase precipitated in the alloys disappeared by heavy cold rolling, leading to being a single bcc phase in the alloys. Furthermore, a distinct texture was formed in the alloys by heavy cold rolling. In this study, it was concluded that these two factors contributed to the increase in magnetostriction in the as-cold rolled $\text{Fe}_{25}\text{Co}_{75}$ alloy.

4. Conclusions

In this work, the $\text{Fe}_{1-x}\text{Co}_x$ ($x=50, 55, 60, 66, 70, 75, 80, 85, 90$ at%) alloys were prepared by forging and subsequent cold rolling. Microstructure, magnetostriction and magnetization of the alloys in bulky state were studied as functions of alloy compositions and thermomechanical treatments. Results obtained in this work are as follows.

1. Magnetostriction of the as-forged alloys was about 60 ppm in the composition range of 50-66 at%. It increased with increasing Co content up to 75 at% and the as-forged $\text{Fe}_{25}\text{Co}_{75}$ alloy reached the largest value of about 108 ppm among the as-forged alloys. The as-forged alloy with the Co content of 80% and over showed a small negative value. In the case of the alloys annealed after forging, the $\text{Fe}_{30}\text{Co}_{70}$ alloy showed the largest magnetostriction of 101 ppm among the annealed alloys. With higher Co content, magnetostriction drastically decreased and showed a small negative value.
2. The magnetostriction of the as-cold rolled Fe-Co alloys was larger than that of the forged alloys and was dependent on the measurement angle to the rolling direction (RD). The as-cold rolled $\text{Fe}_{25}\text{Co}_{75}$ alloy showed relatively large magnetostriction of 128 ppm measured parallel to RD. With higher Co content, the magnetostriction decreased, and the as-cold rolled $\text{Fe}_{15}\text{Co}_{85}$ alloy did not possess any effective magnetostriction. The magnetostriction measured at an angle of 45° to RD was larger than that measured parallel to RD and reached 140 ppm at maximum in the as-cold rolled $\text{Fe}_{25}\text{Co}_{75}$ alloy. This is because the as-cold rolled alloy possessed a nearly $\{100\}\langle 011\rangle$ texture, which means the RD is nearly parallel to the crystallographic orientation $\langle 011\rangle$ of grains, resulting in the axis of easy magnetization $\langle 001\rangle$ being at the angle of 45° to RD. When the as-cold rolled alloys were annealed, the magnetostriction of those annealed alloys became lower than that of the as-cold rolled alloys. In

particular, the annealed alloys with 70 at%Co and over showed a monotonical decrease in magnetostriction with increasing Co content due to fcc phase precipitation.

3. Magnetization of both the Fe-Co forged and cold-rolled alloys showed a tendency to decrease with increasing Co content. From detailed comparison, it was also found that the magnetization of the cold-rolled alloys was slightly larger than that of the forged alloys.

4. The $\text{Fe}_{25}\text{Co}_{75}$ and $\text{Fe}_{20}\text{Co}_{80}$ alloys fully annealed after forging were gradually cold rolled and then magnetostriction and magnetization were measured. Results indicated that the magnetostriction of the cold-rolled $\text{Fe}_{25}\text{Co}_{75}$ and $\text{Fe}_{20}\text{Co}_{80}$ alloys at the reduction rate of 0-60% was not changed but that of the alloys significantly increased with increasing reduction rate from 60% to 97%. According to the XRD and TEM observations, the non-rolled alloys possessed both the bcc and the fcc phases, the intensity of the fcc peak decreased and disappeared with increasing reduction rate. The alloy changed to a bcc single state at the reduction rate of higher than 90%, probably leading to an increase in magnetostriction.

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Figure captions

- Figure 1 Magnetostriction (a) and **saturation** magnetization (b) of forged and subsequently annealed Fe-Co alloys as a function of Co content with the phase diagram indicated in the upper part of the figure. Solid circles: as-forged; open circles: forged and subsequently annealed at 873 K; open squares: forged and subsequently annealed at 973 K and open triangles: forged and subsequently annealed at 1073 K.
- Figure 2 XRD patterns of Fe-Co alloys forged and subsequently annealed at 1073 K. (a) as-forged and (b) annealed after forging.
- Figure 3 Microstructure of the Fe₂₅Co₇₅ alloy sample forged and subsequently annealed at 1073 K for 3h. (a) microstructure and (b) two phase mapping indicating the fcc phase precipitation maybe along grain boundaries of the bcc matrix.
- Figure 4 Magnetostriction (a) and **saturation** magnetization (b) of cold-rolled Fe-Co alloys, measured both in the directions parallel to RD and the 45° incline to RD. Solid circle: as-cold rolled, measured parallel to RD, Solid triangle: as-cold rolled, measured at the angle 45° inclined to RD and Open triangle: cold-rolled and subsequently annealed at 1073 K, measured parallel to RD.
- Figure 5 XRD patterns of Fe-Co alloys forged and then cold rolled. (a) as-rolled and (b) annealed after rolling.
- Figure 6 Microstructure (a) and inverse pole figures (b) obtained from an as-cold rolled Fe₂₅Co₇₅ alloy sheet.
- Figure 7 Magnetostriction (a) and **saturation** magnetization (b) of cold-rolled Fe-Co alloys as a function of rolling reduction rate.
- Figure 8 Variations of XRD pattern of a cold-rolled Fe₂₅Co₇₅ alloy, heavily depending on rolling reduction rate. (a) Fe₂₅Co₇₅, (b) Fe₂₀Co₈₀ and (c) Fe₁₅Co₈₅ alloys.
- Figure 9 TEM observations of the Fe₂₀Co₈₀ alloy subjected to cold rolling at 0%, 60% and 97%. (a), (c),

(e) are bright field images and (b), (d), (f) are selected area diffraction rings.

Figure

Figure 1

Figure 2

Figure 3

Figure 4

Figure 5

Figure 6(a)

(b)

Figure 6(b)

Figure 7

Figure 8(a)

Figure 8(b)

Figure 8(c)

Figures 9(a),(b)

Figures 9(c),(d)

Figures 9(e),(f)