Magnetic Anisotropy of a Semihard Magnetic Alloy of the Co-Fe-Nb System*

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The relationship between the crystalline texture and the magnetic anisotropy in the Co-12Fe-3Nb alloy cold-rolled by 96% reduction in area was investigated through the measurements of X-ray pole figures, hysteresis-loops, and magnetic torque curves. The results obtained are summarised as follows :

(1) The magnetic anisotropy of this alloy is due to the magnetic crystal anisotropy (K_1) and the induced magnetic anisotropy (K_u). The shape of hysteresis-loop depends on which anisotropy is prevailing ; the permivar type for $|K_u| \div |K_1|$, the constricted curve or the rectangular curve for $|K_u| < |K_1|$.

(2) In the cold-rolled and annealed states, the direction of major easy magnetization is along the transverse direction.

(3) Upon annealing at temperatures above 700°C, the direction of hard magnetization changes to that of minor easy magnetization.

(4) The cold-rolling texture of this alloy is the "brass type" one, that is (110)[$\bar{1}12$]. This texture persists up to 900°C. The (111) peak intensity in the X-ray pole figure increases gradually as the annealing temperature rises. Consequently, the value of apparent crystal anisotropy constant increases gradually up to 750°C. When the annealing temperature is over 750°C, the apparent crystal anisotropy constant is -2.3×10^5 erg/cc.

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I. Introduction

Semihard magnetic materials with different magnetic characteristics are needed according to various demands. The AlNi system⁽¹⁾, the Fe-Co-V(Cr) system⁽²⁾⁽³⁾, the Fe-Ni system⁽⁴⁾, the Fe-Mn system⁽⁵⁾⁽⁶⁾, and the high-Co system⁽⁷⁾ etc. have been developed to meet various applications. We have developed a semihard magnetic alloy of the Co-Fe-Nb system⁽⁸⁾ for latching reed relays

which can effectively latch contacts magnetically without exciting coil current. With reference to the magnetic characteristic of a high Co alloy, Chin et al. have disclosed Co-10Fe alloys to which Au⁽⁹⁾ or Ti⁽¹⁰⁾ is added as the third element, and we have also disclosed an alloy containing Be⁽¹¹⁾.

Wenny et al. reported that various shapes of hysteresis loop came out depending on procedures for the prepara-

Fig. 1 Effect of annealing on the hysteresis loops of Co-12Fe-3Nb alloy ; the magnetization direction parallel to the rolling direction.

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tion of specimens in the Co-Fe alloy⁽¹²⁾.

For the Co-Fe-Nb alloy which has been developed for latching reed relays, various shapes of hysteresis loop were obtained as shown in Fig. 1 by means of cold rolling and annealing at appropriate temperatures. Since the shape of the hysteresis loop has a close relationship with the magnetic anisotropy of the materials, the magnetic anisotropy of this alloy was investigated in relation to the texture, the magnetic torque curve, and the hysteresis loops.

II. Experimental Procedure

Co, Fe, and Ferro-Nb were weighed, vacuum-melted and cast into a rod 20 mm in diameter. The chemical compositions of raw materials and alloys are shown in Tables 1 and 2, respectively. The ingot was homogenized

Table 1 Chemical composition of raw materials (wt%).

	Nb	Ta	Fe	Co	Al	Si	Mn	C
Ferro-Nb	67.12	5.43	bal.	—	0.79	0.86	—	0.07
Iron	—	—	99.9	—	—	0.003	0.003	0.002
Cobalt	—	—	0.110	99.63	Ni 0.130	0.025	0.012	0.007

Table 2 Chemical composition of specimen (wt%).

Fe	Nb	Co
11.4	3.13	bal.

at 1100°C for 4 hr in a hydrogen atmosphere and then processed into a wire 2.8 mm in diameter by a series of cold swaging and cold drawing with several intermediate annealings at 1100°C for 30 min. The wire 2.8 mm in diameter was annealed at 1100°C for 30 min and then cold-rolled to a tape 0.05 mm in thickness and 6 mm in width. In this case, the cold reduction in area was 96%. The specimens were annealed at various temperatures from 500 to 1100°C for 1 hr in vacuum.

D.C. hysteresis loops with a maximum field of 100 Oe were measured by a Cioffi-type recording fluxmeter, and in a direction parallel to the rolling direction unless otherwise specified. The texture was obtained by (111) pole figures using filtered FeK α radiation. The unit of intensity contours is 30~40 count/sec. The magnetic torque curve is measured on disks 5 mm in diameter in a rotated field of 10000 Oe with an automatic recording torque magnetometer.

The disk specimen 5 mm in diameter was cut out from the rolled strip.

III. Experimental Results

1. Variation in magnetic characteristics with annealing temperature

The relation between the hysteresis loop and the annealing temperature is shown in Fig. 1. The magnetization in the rolled state varies linearly with the

applied field (H), showing the permivar type and the hysteresis loop in hard magnetization axis. When the annealing temperature increases to 500~600°C, the hysteresis loop is constricted, and, the areas in the 1st and 3rd quadrants are expanded. The constricted hysteresis loop disappears by 650°C annealing and changes into a rectangular hysteresis loop. When the annealing temperature further increases up to 700 or 750°C, the rectangularity in the 2nd quadrant is very sharp. Figure 2 shows the variation in various magnetic properties against the annealing temperature. The magnetization at $H=100$ Oe ($4\pi I_{100}$) hardly changes with annealing temperature. The remanence (B_r) shows a very low value at an annealing temperature below 600°C, and suddenly increases at 650°C, and becomes almost constant above 650°C. The coercive force (H_c) increases with increase in annealing temperature, and

decreases after it shows the maximum value at 700~750°C. Figure 3 shows the hysteresis loop measured in a direction transverse to the rolling direction. Since the measurement was made in such a way that the test piece was cut into several slices in a direction transverse to the rolling direction and these slices were bonded with cello tape, the section area and the demagnetizing coefficient were different depending on the gap and overlapping condition on the bonded surface. Therefore, $4\pi I$ is expressed in arbitrary units. The hysteresis loop in the rolled state inclines due to the demagnetizing field, but it shows a rectangular hysteresis loop when

Fig. 2 Effect of annealing temperature on the magnetic properties of Co-12Fe-3Nb alloy.

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Fig. 3 Effect of annealing on the hysteresis loops of Co-12Fe-3Nb alloy; the magnetization direction transverse to the rolling direction.

the demagnetizing field is compensated; therefore, the direction transverse to the rolling is the direction of easy magnetization. The constricted hysteresis loop does not appear in the transverse direction even by 500 or 600°C annealing, but a rectangular hysteresis loop appears. The rectangular hysteresis loop in the transverse direction by 700°C annealing is almost the same as the hysteresis loop by 700°C annealing in the rolling direction.

2. Variation in texture with annealing temperature

The rolling and annealing textures in Fig. 4 correspond to the $(110)[\bar{1}12]$ texture similar to that found in brass. The (111) peak intensity in the X-ray pole figure increases gradually as the annealing temperature rises.

The texture $(110)[\bar{1}12]$ becomes extremely sharp by 800°C annealing. The sharpness of the texture $(110)[\bar{1}12]$ is weakened by 900°C annealing, and minor $(110)[\bar{1}13]$ component appears. The texture disappears by 1100°C annealing.

Fig. 4 (111) pole figures for Co-12Fe-3Nb alloy cold-rolled 96% and then annealed (a) as rolled, (b) 600°C, 1 hr, (c) 800°C, 1 hr, (d) 1100°C, 1 hr.

Fig. 5 Effect of annealing on the torque curves of Co-12Fe-3Nb alloy in a field strength of 10000 Oe.

Table 3 Torque coefficients of Co-Fe-Nb alloy at various annealing temperature (arbitrary unit).

Ann. temp.	As rolled	500	600	700	750	800	900	1000
L_u	52.5	40.8	50.5	39.9	37.2	33.3	34.8	45.0
L_c	-16.3	-17.2	-22.1	-27.9	-36.5	-33.5	-27.1	-31.3

3. Variation in the magnetic torque curve with annealing temperature

Figure 5 shows the change in the magnetic torque curve against the annealing temperature. The torque exerted by the unit volume of the specimen L is expressed as

$$L = -\partial E / \partial \varphi, \quad (1)$$

where E is the magnetic anisotropy energy and φ is the angle between the spontaneous magnetization and the reference axis on the measuring plane of the specimen. Accordingly, the direction of easy or hard magnetization corresponds to the direction where the torque curve intersects the line of $L=0$ with a negative or positive slope, respectively. Besides, the amplitude of the torque expresses the stability of the direction of easy magnetization.

The rolling direction is taken as the reference axis in Fig. 5. The direction transverse to the rolling direction is the direction of easy magnetization both in rolling and annealing states. On the other hand, the direction parallel to the rolling direction is the direction of hard magnetization in the processes of rolling and annealing below 600°C. However, the parallel direction changes to the direction of easy magnetization when the annealing temperature rises over 700°C. Here, it is found that the transverse direction is the direction of major easy magnetization and the parallel direction is the direction of minor easy magnetization. When the Fourier analysis is applied to these curves, the torque curve is expressed by eq.(2), separated into terms of $\sin 2\varphi$ and $\sin 4\varphi$:

$$L = L_u \sin 2\varphi + L_c \sin 4\varphi. \quad (2)$$

The changes in the coefficients of $\sin 2\varphi$, L_u , and $\sin 4\varphi$, L_c , with annealing temperature are shown in Table 3.

IV. Discussion

The origin of magnetic anisotropy of the present alloy is considered. Crystal magnetic anisotropy, magnetostrictive anisotropy, induced magnetic anisotropy and shape magnetic anisotropy due to precipitated fine particles, etc. are considered for magnetic anisotropy in ferromagnetic materials. In the torque curves of polycrystalline materials, the sum of these kinds of anisotropy is observed. In the variation in L_u and L_c with annealing temperature obtained through the analysis of the torque curve, the change in L_u is small and the change in L_c is large. This fact indicates that the crystal magnetic anisotropy makes a large contribution on the anisotropy of this alloy. Other kinds of magnetic anisotropy except for the crystal magnetic anisotropy are considered in a lump, because they yield uniaxial anisotropy. Among those, the magnetostriction anisotropy is negligibly small since the magnetostriction of this alloy shows a very small value, 5×10^{-6} (13).

Now the observed torque curve is separated into two components, of which one originates from the crystal magnetic anisotropy and the other from the uniaxial anisotropy. Let us discuss first the texture, because the crystal magnetic anisotropy component is closely related to the texture. This alloy has a $(110)[\bar{1}12]$ texture, similar to the brass type orientation. The cold rolling texture $(110)[\bar{1}12]$ does not change by annealing. However, as the annealing temperature rises, the texture $(110)[\bar{1}12]$ becomes sharper, reaching the sharpest by 800°C annealing. This is because of the fact that individual crystal particle is arranged in an ideal direction in the recovery and recrystallization processes and the width of diffraction lines becomes narrower. In other words, if β is defined as the deviation of the (112)

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direction from the rolling direction, β becomes gradually smaller as the annealing temperature rises. The result that the annealing texture is the same as that of the rolling texture is considered due to the recrystallization in situ as has been found in Nb-Ag permalloy⁽¹⁴⁾ or Co-Fe-Be alloy⁽¹⁵⁾, etc. Since the torque curve of a polycrystalline specimen is the sum of the torques on the individual grains, the torque curve based on the crystal anisotropy is calculated first for a single crystal, and then the result is applied to a polycrystalline specimens having the texture. In case of a single crystal, the torque L is expressed by eq.(3) if the reference axis is chosen at $[001]$ on the plane (110) and $|K_1| \gg |K_2|$ is presumed.

$$L = -K_1/8 (2 \sin 2\theta + 3 \sin 4\theta) \quad (3)$$

Considering that the $(110)\langle 112 \rangle$ texture has two symmetrical components $(110)[\bar{1}12]$ and $(110)[1\bar{1}2]$ and that φ is the angle between the magnetization and the rolling direction, the eq.(4) is induced from eq.(3).

$$L = -K_1/8 \left(\frac{2}{3} \sin 2\varphi - \frac{7}{3} \sin 4\varphi \right). \quad (4)$$

Fig. 6 Separation of torque curves into two parts, crystal anisotropy term L_c and uniaxial anisotropy term L_i , for Co-12Fe-3Nb alloy cold rolled 96% and then annealed.
(a) as rolled (b) 600°C, 1 hr (c) 800°C, 1 hr (d) 900°C, 1 hr

Considering deviations from a perfect texture, the coefficients of $\sin 2\varphi$ and $\sin 4\varphi$ are reduced in magnitude by the factors $\sin 2\beta/2\beta$ and $\sin 4\beta/4\beta$, respectively. Since the torque component which originates from the crystal magnetic anisotropy is given in eq.(4), the crystal magnetic anisotropy component can be separated from the observed torque curve. Among the Fourier components shown in Table 3, the quantity, $\alpha (2/3 \sin 2\varphi - 7/3 \sin 4\varphi)$ where α is positive, is the component related to the crystal magnetic anisotropy. Since the $\sin 4\varphi$ term is the one resulting only from the crystal magnetic

anisotropy, α can be decided at a single value from the measured torque curve. Furthermore, assuming that small grains of equal volume are uniformly distributed from $-\beta$ to $+\beta$, the component of the uniaxial magnetic anisotropy except for the crystal magnetic anisotropy (L_i) seems to correspond to a value obtained by subtracting $2/3\alpha$ from L_u . Torque curves thus separated into the crystal magnetic anisotropy component and the uniaxial magnetic anisotropy component are shown in Fig. 6. The values of K_1 and K_u as a function of annealing temperature, obtained from Fig. 6, are shown in Fig. 7. Since the $(110)[\bar{1}12]$ texture becomes sharper, that is, the deviation angle β becomes smaller as the annealing temperature is increased, $|K_1|$ becomes larger apparently. If the saturated value is the true value of K_1 , $K_1 = -2.3 \times 10^5$ erg/cc is obtained. On the contrary, $|K_u|$ becomes smaller, because the degree of the directional order developed during the process of plastic deformation decreases as the annealing temperature is increased. In the rolled state, K_u is equal to 0.9×10^5 erg/cc.

Fig. 7 Variation in the apparent crystal and the uniaxial anisotropy constants of Co-12Fe-3Nb alloy with annealing temperature.

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Since the values of anisotropy constants, negative K_1 and positive K_u , change with annealing temperature, the direction of easy magnetization and consequently the hysteresis loop are influenced by the emulation between the two. As K_u is positive, the direction parallel to the rolling direction becomes the hard magnetization direction, and the transverse direction the easy magnetization direction. Since K_1 is negative, the direction $\langle 111 \rangle$ has the easy magnetization axis, and $\langle 100 \rangle$ the hard magnetization axis. Having the texture $(110)[\bar{1}12]$, the transverse direction is consequently the direction $\langle 111 \rangle$, a direction of easy magnetization. Then, the rectangularity of hysteresis loop measured in the transverse direction is hardly influenced by the annealing temperature because the transverse direction is an easy magnetization direction for both K_1 and K_u . On the other hand, the rolling direction is the direction of hard magnetization according to the K_u term and is the direction of minor easy magnetization according to the K_1 term. Accordingly, the magnetic anisotropy composed of the above two depends on the values of K_u and K_1 . In the rolled state, $|K_u|$ is comparable with $|K_1|$ and then a perminvar-type hysteresis loop is observed. As the annealing temperature rises up to 700°C, $|K_1|$ increases gradually and $|K_u|$ decreases gradually. According to the change in anisotropy constants, the hysteresis loop measured in the rolling direction gradually changes from a perminvar-type to a rectangular one through a constricted hysteresis loop.

V. Summary

The magnetic anisotropy in a semihard magnetic Co-12Fe-3Nb alloy was studied through the texture, the hysteresis loop, and the magnetic torque curve.

The major results obtained are summarized as follows :

(1) The magnetic anisotropy of this alloy consists of crystal magnetic anisotropy and roll-induced magnetic anisotropy. The shape of hysteresis loop depends on which anisotropy is predominant ; the perminvar-type for $|K_u| \div |K_1|$, the constricted curve or the rectangular curve for $|K_u| < |K_1|$.

(2) The direction of major easy magnetization is the transverse direction.

(3) The direction parallel to the rolling direction changes from the direction of hard magnetization to the direction of minor easy magnetization at annealing temperatures above 700°C.

(4) The rolling texture is $(110)[\bar{1}12]$. This texture persists up to 900°C. The (111) peak intensity in the X-ray pole figure increases gradually as the annealing temperature rises. Consequently, the value of apparent crystal anisotropy constant increases gradually up to 750°C annealing.

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