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Mischmetal-FeB-(Co,Ti,Al) Permanent Magnets

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Mischmetal-FeB-(Co,Ti,Al) 영구자석

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초 록 열간압축 및 다이업셋한 Mischmetal-FeB-(Co,Ti,Al) 영구자석의 자기적 특성과 미세조직을 시료진동형 자력계, 투과전자 현미경, 주사전자 현미경, X-선 회절기를 이용하여 조사하였다. $(MM)_{12.5}Fe_{71.9}Co_{5.0}Al_{2.0}B_{8.6}$ 조성의 열간압축 자석은 H_c=4.27 kOe, B_r=4.75 kG, $(BH)_{max}$=3.82 MGOe의 특성을 보였다. 다이업셋 자석은 H_c=3.10 kOe, B_r=5.58 kG, $(BH)_{max}$=5.34 MGOe의 특성을 나타냈다. $(MM)_{12.5}Fe_{77.9}Ti_{1.0}B_{8.6}$ 조성의 경우, 열간압축 자석은 H_c=3.75 kOe, B_r=4.64 kG, $(BH)_{max}$=2.78 MGOe, 다이업셋 자석은 H_c=3.29 kOe, B_r=5.01 kG, $(BH)_{max}$=3.54 MGOe의 특성을 보였다. X-선 회절 및 투과전자 현미경 조사결과에 의하면, 다이업셋 자석에서 c 축이 다이업셋 방향으로 놓이는 결정이방성이 나타나며, 이는 다이업셋 자석의 에너지적의 증가와 관련이 있는 것으로 보인다. Co의 Fe에 대한 일부 치환은 열간압축자석의 자기이방성을 증가시키는 것으로 나타났다.

Abstract Magnetic characteristics of some anisotropic mischmetal-FeB-(Co,Ti,Al) permanent magnets have been investigated. The magnets were fabricated by using hot-pressed and die-upsetting. Hot-pressed $(MM)_{12.5}Fe_{71.9}Co_{5.0}Al_{2.0}B_{8.6}$ permanent magnet showed H_c=4.27kOe, B_r=4.75kG, $(BH)_{max}$=3.82MGOe, and die-upset magnet showed H_c=3.10kOe, B_r=5.58kG, $(BH)_{max}$=5.34MGOe, respectively. Hot-pressed $(MM)_{12.5}Fe_{77.9}Ti_{1.0}B_{8.6}$ permanent magnet showed H_c=3.75kOe, B_r=4.64kG, $(BH)_{max}$=2.78MGOe, and die-upset magnet showed H_c=3.29kOe, B_r=5.01kG, $(BH)_{max}$=3.54MGOe, respectively. X-ray diffraction and transmission electron microscopy revealed that the higher energy products in the die-upset magnets results from alignment of the c-axis along the die-upsetting direction. The magnetic anisotropy in hot-pressed MM-FeB-Al magnet is increased by the substitution of Co for Fe.

1. Introduction

The Nd-Fe-B permanent magnets show the highest magnetic properties with the maximum energy product of more than 40 MGOe. However, rare earth magnets are inferior to ferrites and Alnico magnets in cost. Some hard magnetic alloys with good cost-performance have been investigated through the replacement of high price Nd element in Nd-Fe-B magnets with low price mischmetal which is the mixture of rare earth elements and transition elements.

A few papers reported magnetic properties of mischmetal-Fe-B permanent magnets[1~3] and their economic possibilities in the industry.[1~4] Nd-Co-B ribbons have coercivity as high as 10 kOe[5] although they do not achieve so high magnetic properties as Nd-Fe-B alloys. The addition of Co element in $Nd_{15}Fe_{77.5-x}B_{5.5}Co_xAl_2$ permanent magnet increased the coercivity linearly in the range of 0~10 at.% Co preventing the formation of magnetic impurity phase.[6]

In this research, the magnetic properties and microstructures of hot-pressed and die-upset mischmetal-FeB-(Co, Ti, Al) permanent magnets were investigated using vibrating sample magnetometer, transmission electron microscopy, scanning electron microscopy, optical microscopy, and X-ray diffractometer.

2. Experiments

Commercial grade mischmetal(MM), 10wt.%B ferroboron, 99.5% Co, 99.5% Ti and 99.9% Al were melted into an ingot in a vacuum arc furnace. Ingots with weight losses of less than 1.0% were taken to prepare the specimens. Actual composition of mischmetal

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Table 1. Chemical analysis of mischmetal and ferroboron compounds.

Ferroboron (wt.%)	Fe 85.5	B 9.0	C 1.0	Si 4.0	Al 0.5		
Mischmetal (wt.%)	Ce 51.90	La 26.09	Nd 14.25	Pr 5.33	Fe 1.43	Ba 0.12	Others 0.88

and ferroboron are shown in Table 1.

The nominal composition of the ingots were $(MM)_{12.5}Fe_{77.9}Ti_{1.0}B_{8.6}$ and $(MM)_{12.5}Fe_{71.9}Co_{5.0}Al_{2.0}B_{8.6}$. Ribbons were melt-spun by ejecting the molten alloy through a small orifice 0.7mmφ on a copper wheel(200mmφ) rotating with a surface velocity of about 40m/sec. The ribbons were crushed into powder with sizes less than about 150μm. Hot-pressed magnets were formed with about 8g of powder using a die with a 8mm bore. MoS_2 was used as lubricant to prevent the magnets from sticking to the punch and die. Holding times of 2~3min. were given at temperatures from 650 to 750℃, under compaction pressure of 100MPa. Hot pressing was done under initial vacuum and high purity argon gas atmosphere. Die-upset magnet was made in an over-sized die with a 200mm bore. Temperatures of 720~750℃ and pressures of 300~350MPa were applied for die-upset. Demagnetization curves of the hot-pressed and die-upset magnets were obtained using a Oxford vibrating sample magnetometer and Toei VSM B-H loop tracer at room temperature. X-ray diffraction measurement, transmission electron microscopy (Phillips 200CX, VG microscope HB601 STEM, Joel 2000 TEM) and optical microscopy were used to investigate crystal structures and microstructures.

3. Results and Discussions

X-ray diffraction peak intensities of hot-pressed and die-upset $(MM)_{12.5}Fe_{77.9}Ti_{1.0}B_{8.6}$ and $(MM)_{12.5}Fe_{71.9}Co_{5.0}Al_{2.0}B_{8.6}$ magnets are shown in Fig. 1. All but a few lines of the x-ray diffraction patterns of both the hot-pressed and die-upset magnets, could be indexed as the tetragonal $(RE)_2Fe_{14}B$ type phase with lattice parameters of a= 8.805Å and c= 12.205Å. As also shown in Fig. 1, (006) peak intensity was greatly increased through die-upset. This peak indicates the occurrence of c-axis alignment along the press direction during die-upset. The extra peaks belong to a second RFe_4B_4 phase with richer rare earth elements than matrix as suggested in Ref.[3] The density of the hot-pressed $(MM)_{12.5}Fe_{77.9}Ti_{1.0}B_{8.6}$ and $(MM)_{12.5}Fe_{71.9}Co_{5.0}Al_{2.0}B_{8.6}$ magnets were estimated as 7.17 and 7.35g/cm^3, respectively. The reduction ratios of height of die-upset magnets were about 23%. The optical and

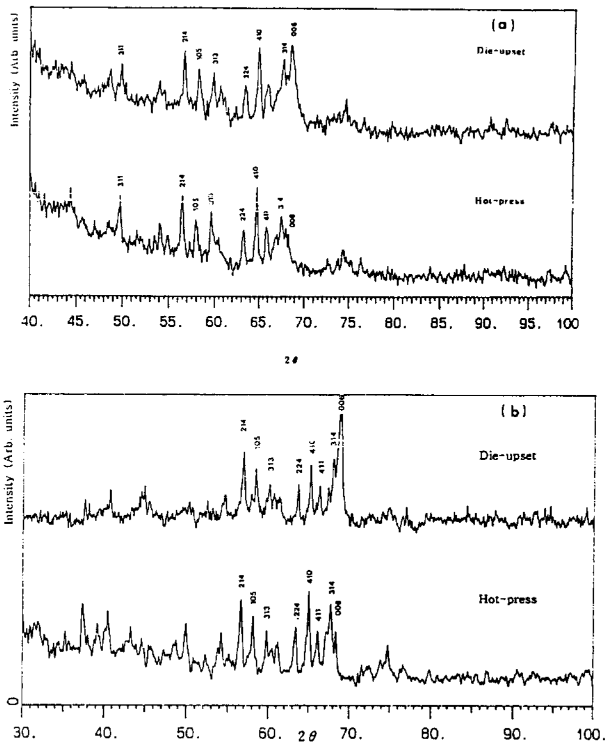

Fig. 1. X-ray diffraction patterns of hot-pressed and die-upset for $(MM)_{12.5}Fe_{77.9}Ti_{1.0}B_{8.6}$ (a) and $(MM)_{12.5}Fe_{71.9}Co_{5.0}Al_{2.0}B_{8.6}$ (b) magnets.

scanning electron microscopy of the die-upset magnets showed that the ribbons are flattened in perpendicular directions to the press direction during die-upset as shown in Fig. 2. This result supported by TEM results that parts of grains near the center showed elongated grains as shown in Fig. 3, contrast to MM-FeB-Al magnets.[4] The magnetic parameters of the hot-pressed and die-upset magnets are given in Table. 2 along with MM-FeB and MM-FeB-Al permanent magnet's results[4] for comparison. As shown in Table. 2, the remanence of the hot-pressed $(MM)_{12.5}Fe_{71.9}Co_{5.0}Al_{2.0}B_{8.6}$ magnets were 7.7% higher than those of the hot pressed MM-FeB-Al magnets. This result means that partial Co-substitution for Fe in MM-FeB-Al magnets creates an increase in magnetic anisotropy as suggested in Nd-Fe(Co,Al)-B permanent magnet systems.[6] In die-upset $(MM)_{12.5}Fe_{71.9}Co_{5.0}Al_{2.0}B_{8.6}$ magnets, the coercivity increased by nearly 33% and remanence decreased by nearly 1.4% compared with MM-FeB-Al

Table 2. Magnetic parameters of hot-pressed and die-upset $(MM)_{12.5}Fe_{77.9}Ti_{1.0}B_{8.6}$ and $(MM)_{12.5}Fe_{71.9}Co_{5.0}Al_{2.0}B_{8.6}$ magnets.

samples \ variables	B_r(KG)	$_iH_c$(kOe)	$_bH_c$(kOe)	$(BH)_{max}$(MGOe)
MM-FeB:Hot-pressed[4]	4.64	3.04	2.05	2.89
MM-FeB:Die-upset[4]	5.45	1.75	1.75	4.12
MM-FeB-Ti:Hot-pressed	4.64	3.75	2.10	2.78
MM-FeB-Ti:Die-upset	5.01	3.29	2.27	3.54
MM-FeB-Al:Hot-pressed[4]	4.41	3.03	2.07	2.83
MM-FeB-Al:Die-upset[4]	5.66	2.33	2.14	4.84
MM-FeB-Al-Co:Hot-pressed	4.75	4.27	2.73	3.82
MM-FeB-Al-Co:Die-upset	5.58	3.10	2.75	5.34

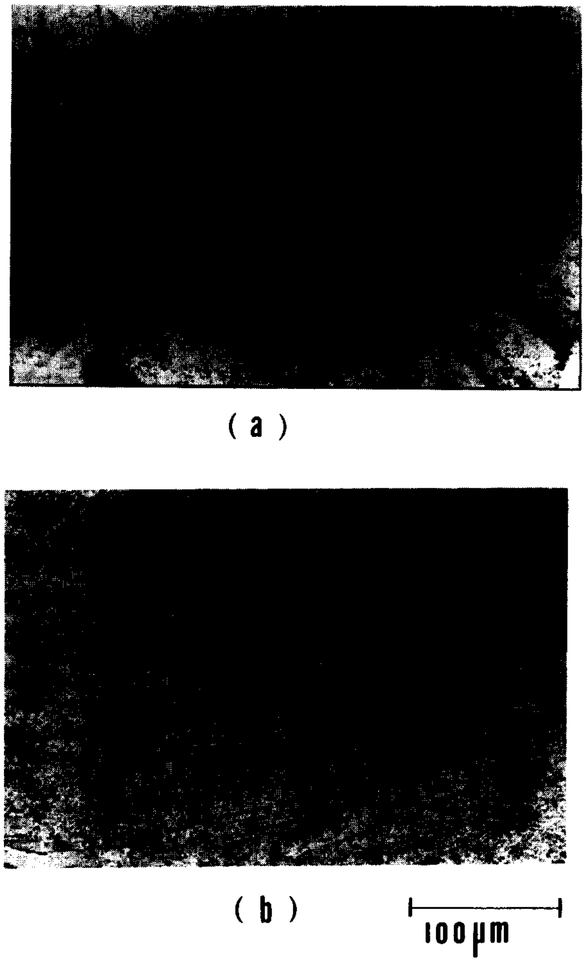

Fig. 2. Optical micrographs of hot-pressed (a) and die-upset (b) $(MM)_{12.5}Fe_{71.9}Co_{5.0}Al_{2.0}B_{8.6}$ magnets.

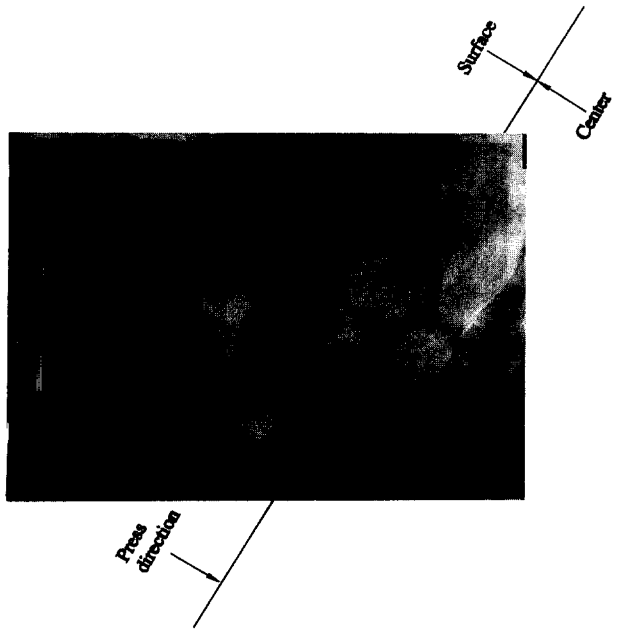

Fig. 3. TEM micrographs of die-upset $(MM)_{12.5}Fe_{71.9}Co_{5.0}Al_{2.0}B_{8.6}$ magnets.

magnets. In case of die-upset $(MM)_{12.5}Fe_{77.9}Ti_{1.0}B_{8.6}$ magnets, the coercivity increased about 88% and remanence decreased by nearly 8.1% compared with MM-FeB magnets. The remanence values of the die-upset $(MM)_{12.5}Fe_{77.9}Ti_{1.0}B_{8.6}$ and $(MM)_{12.5}Fe_{71.9}Co_{5.0}Al_{2.0}B_{8.6}$ magnets were 8.0% and 17.5% higher, respectively, than those of the hot-pressed magnets. This result means that die-upset creates an increment of magnetic anisotropy in MM-FeB-(Co, Ti, Al) permanent magnets as it does in Nd-Fe-B permanent magnets. X-ray diffraction patterns and metallurgical microanalyses suggest that this is due to the alignment of the c-axis to the die-upset direction. A small cracks existed in hot pressed $(MM)_{12.5}Fe_{77.9}Ti_{1.0}B_{8.6}$ magnets and it might result in lower $(BH)_{max}$ values than those of MM-FeB magnets. Demagnetization curves of the hot-pressed and die-upset $(MM)_{12.5}Fe_{77.9}Ti_{1.0}B_{8.6}$ and $(MM)_{12.5}Fe_{71.9}Co_{5.0}Al_{2.0}B_{8.6}$ magnets are given in Fig. 4 and 5. The magnetic properties, in general, are much lower compared with Nd-Fe-B(C) permanent magnets. This is

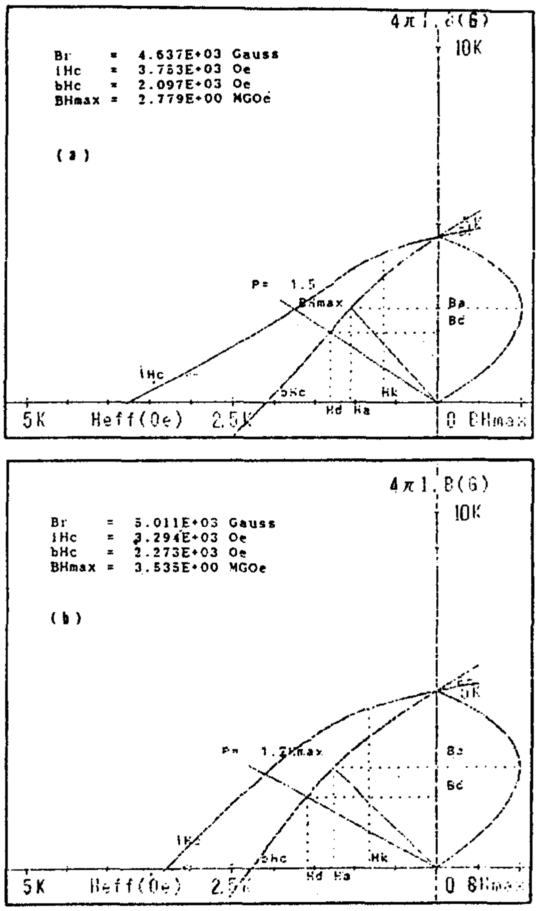

Fig. 4. Demagnetization curves of hot-pressed (a) and die-upset (b) $(MM)_{12.5}Fe_{77.9}Ti_{1.0}B_{8.6}$ magnets.

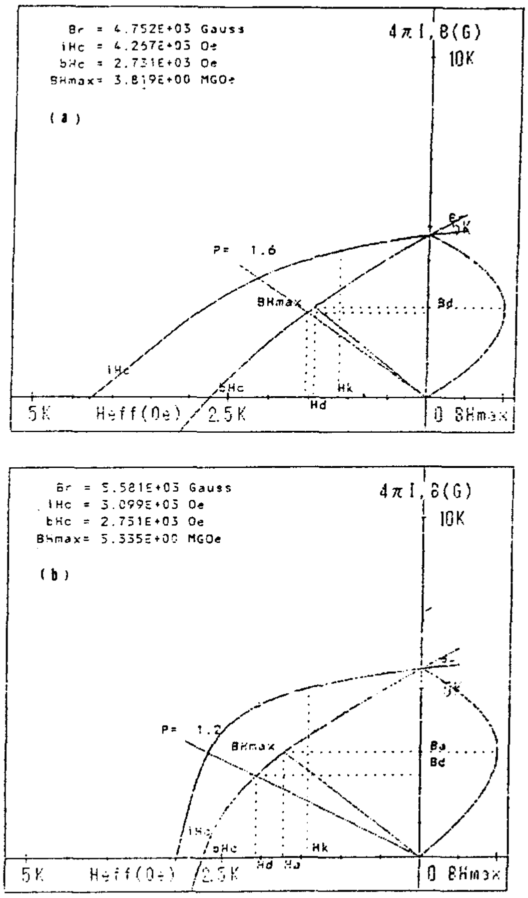

Fig. 5. Demagnetization curves of hot-pressed (a) and die-upset (b) $(MM)_{12.5}Fe_{71.9}Co_{5.0}Al_{2.0}B_{8.6}$ magnets.

mainly because of lower magnetocrystalline anisotropy of tetragonal (La,Ce)-Fe-B compounds.[5] Higher squareness in demagnetization curve of the die-upset $(MM)_{12.5}Fe_{71.9}Co_{5.0}Al_{2.0}B_{8.6}$ magnet suggests that die-upset could be a useful method for fabrication of MM-FeB-(Co,Ti,Al) permanent magnets as Nd-Fe-B(C) permanent magnets.[6]

4. Conclusions

Die-upset Mischmetal-FeB-(Co,Ti,Al) magnets showed that they are much higher $(BH)_{max}$ than that of the hot-pressed magnets in the same alloy system. This seems mainly due to the occurrence of the c-axis alignment along press direction during die-upset. The substitution of Co, Ti for Fe in MM-FeB-Al and MM-FeB magnets, respectively, attributed to increase the magnetic anisotropy in the hot-pressed and die-upset magnets.

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