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CRYSTAL STRUCTURE TRANSFORMATION OF Ba-Sr HEXAFERRITE AND ITS EFFECT ON PARTICLE ORIENTATION IN RECYCLE PROCESS

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ABSTRACT

CRYSTAL STRUCTURE TRANSFORMATION OF Ba-Sr HEXAFERRITE AND ITS EFFECT ON PARTICLE ORIENTATION IN RECYCLE PROCESS. In general, during its fabrication, 2-5 % of total products of Ba-Sr hexaferrite permanent magnet will be rejected. Accordingly, an increase in rejected product of permanent magnet needs to be considered. It is expected that the re-utilization of rejected product can increase the production efficiency in order to achieve zero waste-production. In the present work, the recycling process of Ba-Sr hexaferrite permanent magnet was studied by applying 1 T magnetic field (anisotropic) to align the powder. The rejected products were milled using shaker mill PPF-UG for 10-40 min and then sintered at 1200 ºC for 60 min. The results show that the remanence of original powder was increased by 50% after the particle orientation. However, the recycled sample doesn't show a significant different. SEM and XRD analysis show the crystalline structure transformation from symmetrical hexagonal to asymmetrical hexagonal structure with crystalline growth in a-b axis direction. This transformation leads to lost in its magneto crystalline anisotropy. Therefore, it was difficult to align the particle.

Keywords: Ba-Sr Hexaferrite, Orientation, Magnetic Field, Anisotropy, Magneto crystalline anisotropy

ABSTRAK

TRANSFORMASI STRUKTUR KRISTAL Ba-Sr HEKSAFERIT DAN PENGARUH ORIENTASI PARTIKEL DALAM PROSES DAUR ULANG. Produksi magnet permanen Ba-Sr Hexaferrite umumnya mengalami kegagalan berkisar 2-5% dari total produksinya. Oleh karena itu, limbah magnet permanen yang terus meningkat perlu menjadi perhatian. Daur ulang magnet permanen diharapakan dapat meningkatkan efisiensi produksi untuk mencapai produksi yang ramah lingkungan. Pada penelitian ini dilakukan studi daur ulang magnet permanen Ba-Sr Hexaferrite dengan proses orientasi partikel atau anisotropy menggunakan medan magnet sebesar 1 Tesla. Ba-Sr Hexaferrite di-ball milling menggunakan shaker mill PPF-UG selama 10-40 menit dan dilanjutkan dengan sintering pada 1200ºC selama 60 menit. Hasil analisa menunjukkan bahwa sample yang menggunakan powder asli menunjukkan peningkatan remenansi sebesar 50% setelah proses orientasi partikel, sedangkan sample dari hasil daur ulang tidak menunjukkan perubahan yang signifikan. Analisa SEM dan XRD menunjukkan bahwa terjadi transformasi bentuk kristal dari hexksagonal yang simetris menjadi heksagonal non simetris, dengan pertumbuhan kristal kearah sumbu a-b. Transformasi ini menjadikan partikel kehilangan sifat magneto crystalline anisotropynya sehingga sulit diorientasikan

Kata kunci: Ba-Sr *Hexaferrite*, Orientation, *Magnetic Field*, *Anisotropy*, *Magneto crystalline anisotropy*

INTRODUCTION

The ferrite base-permanent magnet such as Barium Hexaferrite and Strontium Hexaferrite are commercial ceramic magnet which is made by sintering of the refined and compacted powder, and its characteristics strongly depend on the microstructure of the powder [1]. In a typical production line, about 2-5% of the total products will be rejected. If this large amount of rejected material can be recycled, the eeficiency of material utilization can be improved and the environmental issue may be resolved.

The typical magnetic wastes contain many additive elements, such as Zn [2], Ce, Mn[3], La, Co, Ca[4,5], Pb[6] etc. with different compositions, which makes the difficulties in controlling the properties of the recycled product. One of the possible solution is by recycling the rejected product from the fabrication line since it is more controllable. However, different synthesis method may results to the change of the optimum microstrucure of the final product. In this case, the combination of mechanical milling/mechanical alloying and proper heat treatment [7, 8] should be the solution to obtain the expected properties.

To date, the possibility of utilizing the recycled Ba-Sr Hexaferrite waste is still not intensively studied. In addition, the effect of ball milling on the characteristic of orientability is still not well understood. Thus, in this study, the crystal structure transformation of Ba-Sr hexaferrite and its effect on the orientation process will be investigated and discussed in details.

EXPERIMENTAL METHOD

The material for this experiment was the technical product, obtained from China, which are the mixture of Barium Hexaferrite and Stronsium Hexaferrite with the composition as shown in Table 1. As a reference sample, the original Ba-Sr Hexaferrite powder was dried and compacted in dry state under magnetic field of 0 – 1 T. The compaction process was conducted using magnetic field press system made in Research Center (RC) for Physics [5] to obtain the oriented magnetic particle in the direction of compaction (parallel type magnetic field press). The sample was held for 5 minute in the orientation state prior to the compaction. The pressure was set to 20kgf/cm^2 to obtain solid φ16 x 16mm sample. The as-compacted sample was sintered at 1200 ºC for 60 minute with a heating rate of 10 ºC/minute and cooled down naturally in the furnace.

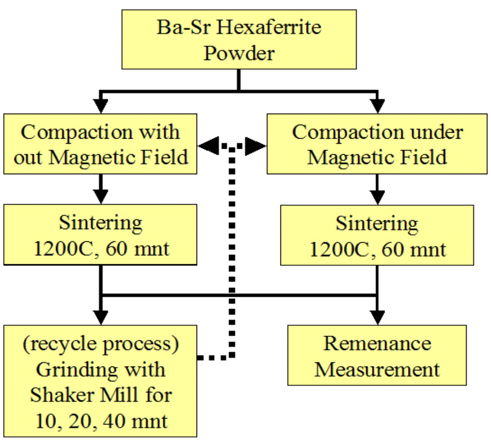

Figure 1. Schematic diagram of the experiments

Table 1. The elemental composition of Ba-Sr Hexaferrite

No	Element	Content (wt.%)
1	Fe	85.73
2	Ba	4.38
3	Sr	3.35
4	Si	3.25
5	Ti	1.63
6	Al	1.02
7	Ca	0.30
8	Mn	0.29
9	Cu	0.03
10	Ni	0.02

The recycled powder was obtained by ball milling of the sintered sample using Shaker Mill made in RC for Physics (PPF-UG) for 10, 20 and 40 min. The as-milled sample was the compacted without and with 1 T magnetic field, and then sintered at 1200 ºC for 1 hour. The schematic diagram of the experiment is shown in Fig. 1.

The as-sintered sample was then magnetized using 2 T pulse magnetization. The magnetic remanence was measured using gauss meter (Hall Effect sensor). The crystal structure features were examined using X-Ray Diffractometer (XRD) (Rigaku-SmartLab). The microstructural change was observed using Scanning Electron Microscope (SEM) (Hitachi). The SEM analyses were conducted on the surface perpendicular to the direction of given magnetic induction using fracture method.

RESULTS AND DISCUSSION

Figure 2 shows the magnetic remanence of the sintered permanent magnet. The magnetic remanence of

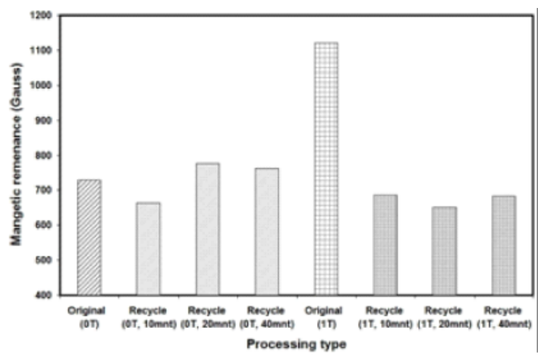

Figure 2. Figure 2. Magnetic remanence of the original and recycled powder which was processed under magnetic induction 0 and 1 T, and milling time 0, 10, 20 and 40 minutes

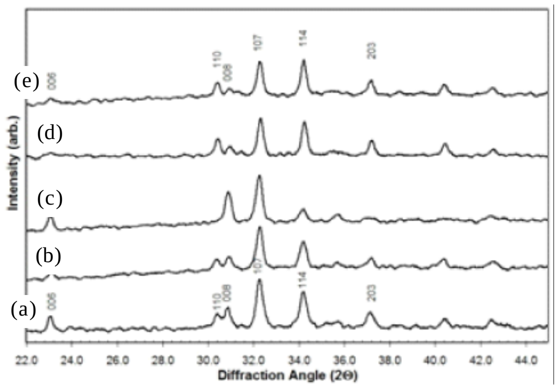

Figure 3. XRD pattern of (a). original powder, (b). sintered original powder without orientation, (c). sintered original powder original with orientation, (d). recycled powder, milled for 40 minute and (e). sintered milled recycled powder with orientation

the original powder which was oriented under 1 T magnetic induction increased 50% of initial value without orientation. The recycled powder which was milled for 10, 20, and 40 minute, and compacted under 1 T magnetic induction had magnetic remanence in the range of 663-728 Gauss, which is similar with the original powder without orientation. This implies the loss of magneto crystalline anisotropy from the recycled Ba-Sr Hexaferrite particle.

Figure 3 shows the XRD pattern of each sample. The crystal structure analysis revealed the perfect hexagonal structure of original Ba-Sr Hexaferrite powder. The sintered powder without orientation had similar XRD pattern with the decreasing intensity in some peaks, compared with the XRD pattern of the original powder. This shows that the compaction without magnetic induction did not change the crystalline structure.

Tabel 2. The intensity change and the corresponding ratio of Ba-Sr Hexaferrite original powder peaks

Peak	006		110		008	
Intensity/ratio	Int	ratio	Int	ratio	Int	ratio
Powder Original	32	1	21	1	44	1
Sintered PO. in 0T	8	0.3	15	0.7	24	0.5
Sintered PO. in 1T	30	0.9	0	0.0	69	1.6
Milled Recycle Powder for 40 mnt	4	0.1	25	1.2	12	0.3
Sintered MRP for 40mnt in 1T	6	0.2	22	1.0	8	0.2

Peak	107		114		203	
Intensity/ratio	Int	ratio	Int	ratio	Int	ratio
Powder Original	107	1	84	1	32	1
Sintered PO. in 0T	99	0.9	59	0.7	21	0.7
Sintered PO. in 1T	111	1.0	27	0.3	8	0.3
Milled Recycle Powder for 40 mnt	83	0.8	73	0.9	30	0.9
Sintered MRP for 40mnt in 1T	72	0.7	77	0.9	30	0.9

Some peaks of the compacted sample under magnetic induction showed some change in intensity, either increasing, decreasing, or disappearing. Table 2 summarize the detail of intensity change of each main peaks. When the original powder was processed under magnetic induction, the intensity of [008] peak, which is corresponded with the c-axis on <001> direction, increased 60%. Meanwhile, the [110], [114], and [203] peaks which are affiliated with a and b-axis decreased and almost disappeared. This shows that the orientation process using 1 T magnetic induction have aligned the particle of original powder in the same direction with the given magnetic field. As a result, the magnetic remanence was increased up to 50%, as can be seen in Figure 2.

On the contrary, the recycled powder showed no significant change, except for [006] and [008] peaks, where the intensity was slightly decreased. This infers that the original hexagonal structure was remained and the orientation under 1 T magnetic induction had no significant effect on the crystal alignment. This phenomenon implies the decrease of magneto crystalline anisotropy properties of Ba-Sr Hexaferrite.

The comparison of peaks intensity of original and recycled powder showed the decreasing tendency of recycled powder diffraction peaks. This shows the asymmetrical direction change of initial diffraction plane of original powder into the perpendicular asymmetrical direction of recycled powder. In addition, there is no significant change on the peak intensity of recycle powder after being oriented and sintered. This shows that the crystal of recycled powder grew to the initial direction.

Figure 4(a) depicts the surface of sintered original powder without orientation, where the grains were distributed randomly with the nearly hexagonal-shape. The grain size was ranged from 1 to 3μm and was distributed evenly, including the intra particle gaps.

Figure 4(b) shows the surface of sintered original powder with orientation which revealed the existence of relatively large hexagonal crystal at point 1, 2 and 3 aligned parallel to the normal of surface. Based on XRD analysis, the crystal was aligned along c-axis or <001> of Ba-Sr Hexaferrite. The big particles of Ba-Sr Hexaferrite aligned in the same direction as magnetic induction. The existence of many particles around bigger particles implies the broad range of original powder particle size. In that condition, the bigger particles were found easier to be oriented and formed homogeneous arrangement. The similar phenomenon was not observed for smaller particles.

Figure 4(c) and (d) (enlarged) show the microstructure of sintered recycled powder with orientation. It can be seen that the particle size distributed more homogeneously, yet there was still many pores around the particles. Figure 4(d) at point 4 and 5 shows the change of particle shape, where the asymmetric direction of hexagonal crystal changed along a and b

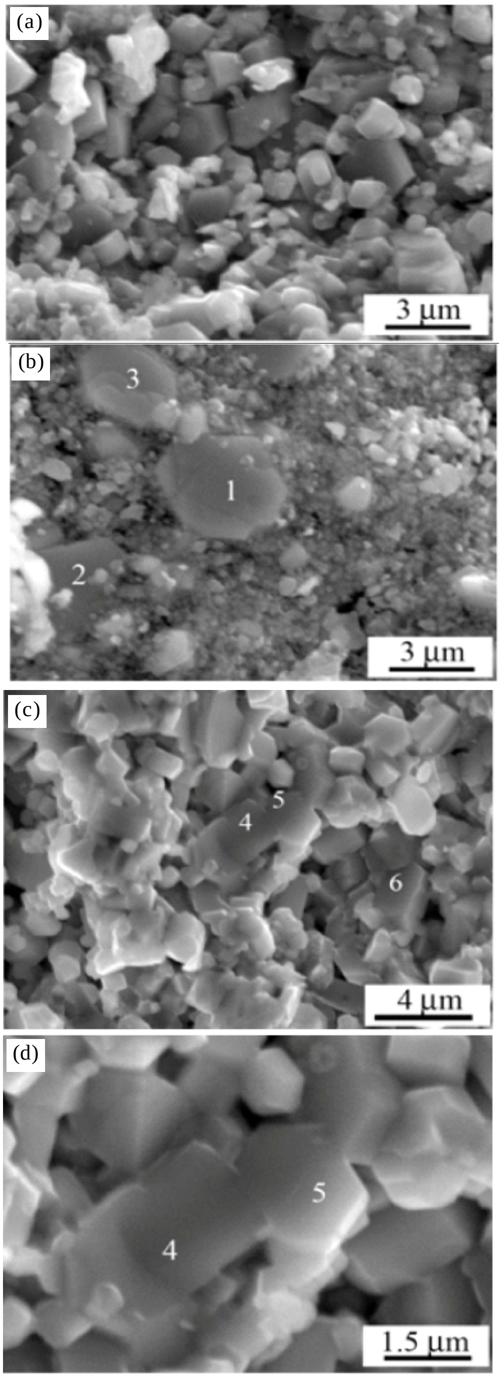

Figure 4. The microstructure of Ba-Sr Hexaferrite (a). sintered original powder without orientation, (b). sintered original powder original with orientation, the surface was in perpendicular direction to the given magnetic induction, (c). sintered recycled powder which was milled for 40 minute with orientation and (d). enlarged image of (c)

axis. In correlation with the XRD result, the intensity change of [110] and [114] peaks was associated with the crystal structure change in Figure 4(d). The change of a and b axis length, relative to c-axis length, made the change of direction possible and the particle was unable to be oriented. These occur after milling treatment, which infers the destruction of magneto crystalline anisotropy of Ba-Sr Hexaferrite due to mechanical impact from ball milling process. Thus, the recycled powder was no longer can be used as anisotropic magnet.

Though the original powder and recycle powder have the same composition, SEM result shows the abnormal crystal growing on sample of the recycle powder. Abnormal crystal growing could occur as the effect of the addition of sintering aids material such as SiO_2, CuO and B_2O_3 [9] and over sintering process. Abnormal crystal growing in hexagonal ferrite occurs as combination between grain boundary movement and Ostwald ripening, which could results in acicular platelets shape [10]. The addition of SiO2 in the original powder may reduce the properties of magneto crystalline anisotropy of recycled powder.

CONCLUSION

In the present study, the utilization possibility of recycled Ba-Sr Hexaferrite magnet powder for anisotropic magnet was investigated. The results showed the increase of magnetic remanence of original powder after being oriented (up to 50%), which was not observed for the recycled powder. Scanning Electron Microscope (SEM) and X-Ray Diffractometer (XRD) analysis showed the crystal structure transformation from symmetric hexagonal to asymmetric hexagonal, with the crystal growth direction along a and b axis. The crystal structure transformation made the recycled powder lose its magneto crystalline anisotropy property and made it unable to be oriented.

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