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ABSTRACT
The magnetic properties and microstructures of 2-17 type Sm-Co magnets with high Fe and low Zr content were investigated. The developed magnet achieved maximum energy product, \(BH_m\) of 34.5 MGOe, intrinsic coercivity, \(H_{cj}\) of 21.3 kOe and squareness of 73.3% at 25 \(\degree\)C. Temperature coefficients of remanent magnetic flux density, \(B_r\) and \(H_{cj}\) were 0.034%/K and 0.28%/K respectively, which values were almost as same as the conventional Sm-Co magnets. Moreover, the developed magnets had high magnetization orientation. For XRD, it was found that Zr was preferentially substituted by Co-Co pair, this made interaction between Co and Co stronger, so that heat resistance was maintained. Magnetic domain structures were observed with a Kerr effect microscope, and then it was observed that the developed magnet had strong pinning force. In the microstructures, the developed magnet had 200 ∼ 500 nm cell size with Fe and Cu separated clearly. This led to large gap of domain wall energy which produces strong pinning force. Because the developed magnet had high magnetization orientation and large gap of domain wall energy, we achieved high magnetic properties and high heat resistance on the developed magnet.

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I. INTRODUCTION
Permanent magnets with high-heat resistance have been highly demanded along with increasing electric vehicles and hybrid electric vehicles. Sm-Co magnets are well known for having high heat resistance by high Curie temperature and basically consisted of five elements, Sm, Fe, Cu, Zr and Co. As a way of enhancing magnetic properties of 2-17 type Sm-Co magnets, increasing Fe content has been actively conducted. However, increasing Fe content leads to poor squareness and deterioration of heat resistance due to decrease in Co content. To maintain heat resistance with Fe content increased, other elements such as Cu or Zr should be reduced. In this study, we tackled these problems by reducing Zr content. Although magnetic properties of Sm(Fe<sub>v</sub>, Cu<sub>0.088</sub>, Zr<sub>x</sub>, Co<sub>bal</sub>)<sub>7.0</sub> were investigated and it showed that Zr has a key roll to make Fe magnetically stable and expand Fe solubility limit due to lattice expansion by size effect, if Fe content was higher than \(v=0.25\) and Zr content was lower than \(x=0.02\), high magnetic properties have not been obtained, e.g. intrinsic coercivity \(H_{cj}\) of 7.5~12.3 kOe. Moreover, there seems to be no reports discussing heat resistance with increasing Fe and reducing Zr content. To achieve high magnetic properties on high Fe and low Zr content, obtaining the proper crystal structure, large cell structure and compositional homogenization are strongly required. Here, we employed slow cooling between sintering and solid solution treatment to obtained preferred structures and investigated magnetic properties and heat resistance from the view point of magnetization orientation, crystal structure, lattice constants, magnetic domain structures and microstructures.

II. EXPERIMENTAL PROCEDURES
Sm(Fe<sub>0.33</sub>, Cu<sub>0.055</sub>, Zr<sub>0.018</sub>, Co<sub>bal</sub>)<sub>7.55</sub> ingot for the developed magnet and Sm(Fe<sub>0.28</sub>, Cu<sub>0.055</sub>, Zr<sub>0.022</sub>, Co<sub>bal</sub>)<sub>7.55</sub> ingot for a reference
magnet (LM-32SH® which has been mass produced in our company) were prepared by using a book mold method. The ingot was heat treated at 1180 °C for 1 hr, followed by 1140 °C for 6 hrs under an inactive gas atmosphere. The heat-treated ingot was crushed and pulverized by using a ball mill with isopropyl alcohol. The obtained powder was pressed to the perpendicular direction for an applied magnetic field. The compacts were sintered at 1210 °C for 1 hr followed by 1150 °C for 25 hrs under an inactive gas atmosphere. At the time of moving between sintering and solid solution treatment, slow cooling with rate of 0.8 °C/min was employed to promote homogenization structurally and compositionally. Aging was conducted for 850 °C for 12 hrs followed by slow cooling to 350 °C. Magnetic properties were evaluated with a B-H tracer at 25 °C and 150 °C. Phase identification and lattice constants were evaluated with X-Ray Diffraction (XRD) with Cu-Kα target. Magnetization orientation was examined with Electron Back Scattering Diffraction (EBSD). Magnetic domain structures were observed with a Kerr effect microscope with a magnetic field applied from 0 to −20 kOe. Microstructure observation and its compositional line analysis were conducted with Scanning Transmission Electron Microscope (STEM) and Energy Dispersive X-ray (EDX).

III. RESULTS AND DISCUSSION

A. Magnetic properties

Fig. 1 shows demagnetization curves of the developed magnet and the reference magnet at 25 °C and 150 °C. The demagnetization curve of the developed magnet passed over the that of the reference magnet up to the knick point. With respect to magnetic properties, the developed magnet had remanent magnetic flux density, \( B_r \) of 12.10 kG, maximum energy product, \( [BH]_m \) of 34.5 MGOe and \( H_{cj} \) of 21.3 kOe respectively, these values were further higher than those the referential magnet. Moreover, squareness calculated from the following equation was 73.3%.

\[
\text{Squareness} = \frac{H_k}{H_{cj}}
\]

Where \( H_k \) is the value of reverse magnetic field when magnetic flux density becomes 90% of \( B_r \).

At 150 °C, the developed magnet had \( B_r \) of 11.59 kG, \( [BH]_m \) of 30.5 MGOe and \( H_{cj} \) of 13.8 kOe respectively. Temperature coefficients of \( B_r \) and \( H_{cj} \) were 0.034%/K and 0.28%/K respectively from the following equation. These values were almost as same as the existing magnets.

\[
\text{Temperature coefficient} = \left( \frac{B_{25} - B_{150}}{B_{25}} \right) \times 100
\]

Where \( B_{25} \) and \( B_{150} \) were the values of the \( B_r \) at 25 °C and 150 °C. This equation can be also applied to the temperature coefficient of \( H_{cj} \). Moreover, B-H line of the developed magnet was linear. This means that irreversible demagnetization doesn’t occur even at 150 °C. We believed that this was the first success which realized both high magnetic properties and high squareness on this composition condition.

Fig. 2(a) and (b) show an Inverse Polar Figure map (IPF map) and a Polar Figure map (PF map) of the developed magnet. For the IPF map, crystal grains are colored depending on magnetization orientation direction. If \( c \)-axis orientation which is favorable for Sm-Co magnets is obtained, crystal grains are supposed to be colored in red. In fact, because the developed magnet had crystal grains with red color or close to red color, \( c \)-axis orientation was obtained. Even from the PF map, it certified that high magnetization orientation was obtained.

B. Crystal structure and lattice constant

Fig. 3(a) and (b) show XRD patterns of the developed magnets with Sm(Fe₀.₃₃, Cu₀.₀₅₅, Zrₓ, Co₉₅)(x=0.018, 0.020, 0.022)
after aging process and Zr content dependence of lattice constants respectively. Here, the developed magnet shown in Fig. 1 had Zr content of $x=0.018$. From Fig. 3(a), all peaks were able to be identified Rhombohedral structure (Space group R-3m) and no impurity phases were observed. This is the one of the reasons for achieving high magnetic properties and high squareness. From Fig. 3(b), $a$-axis was increased while $c$-axis was decreased with increasing Zr content. Crystal structure of 2-17 type Sm-Co magnets consist of three sub lattices and has Co dumbbell (Co-Co pair) sites.\textsuperscript{10–12} Considering atomic radii of Zr and Co, Zr is larger than both Co and Co-Co pair for $a$-axis direction, larger than Co and smaller than Co-Co pair for $c$-axis direction. Therefore, if Zr is substituted by Co-Co pair, lattice constants of $a$-axis and $c$-axis show opposite behavior with each other. Because the result at this time was in that manner, it can be said that Zr was preferentially substituted by Co-Co pair in cobalt dumbbell sites, which led to make interaction between Co and Co stronger. By this, high heat resistance was achieved on the developed magnet.

C. Magnetic domain structures

Fig. 4 shows magnetic domain structures of the developed magnet observed with a magnetic field applied from 0 to $-20$ kOe. Observation plane is the parallel plane to the easy direction of magnetization. From Fig. 4, reverse magnetic domains were generated evenly from a grain boundary at around $-5$ kOe, inside grains at around $-11$ kOe. After that these propagated to inside grains, finally magnetic reversal finished at $-20$kOe. This magnetic reversal process was the same as the process we reported and a necessary condition for high squareness.\textsuperscript{8,13} However, a magnetic domain was generated from another grain boundary at $-9$ kOe, which means that the developed magnet had stronger pinning force than the magnets we reported.\textsuperscript{8,13} Until now, we got finding that impact to demagnetization by reverse magnetic domains is following order; generation from grain boundaries, generation from inside grains and propagation to inside grains.\textsuperscript{10} Basically, variability of generation of reverse magnetic domains is unfavorable for high squareness. However, demagnetization factor of the developed magnet almost stayed constant to the knick point. This was because sum of impact to demagnetization from any reverse magnetic domains was almost constant to the knick point.

D. Microstructures

Fig. 5(a) and (b) show a STEM image with composition mapping and results of composition line analysis of the developed magnet.
magnet respectively. STEM observation was conducted from the perpendicular direction for c-axis. From Fig. 5(a), cell size was approximately from 200 to 500 nm with Fe and Cu clearly separated to 2-17 phase and 1-5 phase. This cell size was larger than existing Sm-Co magnets. From Fig. 5(b), Fe concentration went down to 10 at%, while Cu went up to 42 at% in 1-5 phase. These composition behaviors are crucial to high magnetic properties and high squareness on Sm-Co magnets. 2-17 type Sm-Co magnets are classified into “Pinning type magnet”, which generation of reverse magnetic domains and propagation were suppressed until external magnetic field exceeds domain wall energy. Pinning force closely relates with gap of magnetic domain wall energy between 2-17 phase and 1-5 phase. This energy is determined by magnetic anisotropy and cell structure. High magnetization orientation is needed to high magnetic anisotropy. As shown in Figs. 2(a) and (b), we obtained high magnetization orientation. For cell structure, having large cell size with clear separation of Fe and Cu is essential to obtain large gap of domain wall energy. In fact, the developed magnet had this kind of structure, so that gap of domain wall energy between 2-17 phase and 1-5 phase was enlarged, and pinning force was strengthened. Moreover, the reason that generation of reverse magnetic domain occurred at −9 kOe was attributed to these things. Finally, because obtaining high magnetic anisotropy and microstructure as favored, we achieved both high magnetic properties and squareness.

IV. CONCLUSIONS

We investigated magnetic properties and heat-resistance of Sm-Co magnets with high Fe and low Zr content. The results are summarized as follows.

1. The developed magnet had $B_r$ of 12.10 kG, $[BH]_m$ of 34.5 MGOe, $H_{cj}$ of 21.3 kOe at 25 °C and $B_t$ of 11.59 kG, $[BH]_m$ of 30.5 MGOe, $H_{cj}$ of 13.8 kOe at 150 °C, moreover, squareness was 73.3%.

2. Temperature coefficients of $B_r$ and $H_{cj}$ were 0.034%/K and 0.28%/K respectively.

3. High magnetization orientation was obtained.

4. Because Zr was substituted by Co-Co pair, interaction between Co and Co become stronger, this led to high heat resistance.

5. The developed magnet had strong pinning force and sum of impact to demagnetization from any reverse magnetic domains was almost constant to the knick point.

6. Large gap of domain wall energy was obtained due to high magnetization orientation and 200–500 nm cell size with clear separation of Fe and Cu.

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