Microstructure and magnetic properties of grain boundary modified recycled Nd-Fe-B sintered magnets

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Full-density recycled Nd-Fe-B sintered magnets were produced from collected waste Nd-Fe-B magnets with the coercivity, $\mu_0 H_c$, of 1.30 T and remanence, $\mu_0 M_r$, of 1.37 T. A small fraction of Nd₆Dy₂₁Co₁₉Cu_{2.5}Fe powder (Grain Boundary Modified (GBMTM) powder) was blended to the powders obtained from the Nd-Fe-B waste magnets. The addition of 5.0 wt% GBMTM powder resulted in $\mu_0 H_c=2.36$ T with $\mu_0 M_r=1.29$ T in recycled magnets. The temperature coefficient of intrinsic coercivity, β , in the recycled magnet was measured to be -0.47 %/°C. Microstructure studies showed that the addition of the GBMTM powder increases the volume fraction of a metallic Nd-rich phase in the recycled magnets. 3D tomography revealed a good interconnection of Nd-rich phases through the grain boundaries of the recycled magnet. Microstructure studies suggested that obtained high coercivity in the recycled magnet with 5.0 wt% addition of GBMTM is due to the formation of distinct grain boundary phase combined with the formation of Dy-rich shell in the Nd₂Fe₁₄B grains with ~0.8 at. % Dy.

Keywords: Nd-Fe-B sintered magnets, Recycling, Coercivity, Grain boundary, Microstructure.

1. Introduction

Anisotropic Nd-Fe-B sintered magnets are widely used in a variety of applications such as electric power generation and transportation. The typical composition of the anisotropic Nd-Fe-B sintered magnets used in the traction motors of hybrid cars is around $\text{Nd}_{22}\text{Dy}_{11}\text{Fe}_{65}\text{B}_1\text{Cu}_{0.1}$ wt% indicating that 33 wt% of these sintered magnets is comprised of precious rare earth elements [1]. Although the production and the applications of Nd-Fe-B permanent magnets are increasing year by year, the life-time of various appliances in which Nd-Fe-B based permanent magnets are used is limited [2]. Hence, it is important to recycle the Nd-Fe-B magnets after reaching to the end of the life of the devices containing Nd-Fe-B based permanent magnets.

Several methods to recycle waste Nd-Fe-B sintered magnets have been reported. Itoh *et al.* produced isotropic Nd-Fe-B bonded magnets by melt-spinning Nd-Fe-B waste sintered magnets [3]. Hydrogen decrepitation was introduced as a method to make powders out of Nd-Fe-B waste sintered magnets [4-6]. The hydrogenation decomposition desorption recombination (HDDR) process was also employed to produce powders for bonded magnets applications from Nd-Fe-B waste sintered magnets [4-6]. The recycled powders can be also used to make anisotropic Nd-Fe-B sintered magnets [7-9]. However, the low coercivity of the recycled powders is one of the drawbacks to develop high coercivity anisotropic sintered magnets. In addition, full density Nd-Fe-B sintered magnets cannot be produced only from the recycled Nd-Fe-B powders because of the oxidation of Nd-rich phases during the recycling process. Jang *et al.* and Kwon *et al.* proposed a method to produce high coercivity recycled powders using DyF_3 salt to form $(\text{Nd,Dy})_2\text{Fe}_{14}\text{B}$ shell on the surface of the powders [10,11]. On the other hand, Kawasaki *et al.* successfully produced the full density anisotropic Nd-Fe-B sintered magnets that have comparable magnetic properties to those of initial magnets by adding $\text{Nd}_{80}\text{Fe}_{20}$ binary powder to jet milled Nd-Fe-B recycled powders [7]. Zaktonik *et al.* also reported that the

addition of 1.0 at.% Nd to the recycled jet-milled Nd-Fe-B powders is sufficient to recover the magnetic properties of recycled magnets [9]. More recently, Li *et al.* reported recycled Nd-Fe-B sintered magnets with a comparable remanent magnetization, coercivity and maximum energy product by blending Nd₂₂PrFe₁₄B powder with the recycle powders before sintering [12]. These reports indicate that the composition selection of the powders to be added to the recycled powders is crucial to recover the initial hard magnetic properties. In this work, we report a new composition of additive powders to be used in two powder blending method to modify the grain boundary composition of the recycled full-density Nd-Fe-B sintered magnets. The recycled anisotropic Nd-Fe-B sintered magnet showed superior coercivity compared to those of the initial Nd-Fe-B waste magnets. The microstructure of the magnets was studied in detail to correlate the magnetic properties with the microstructures of the recycled Nd-Fe-B sintered magnet.

2. Experimental procedure

Inputs of 100 kg batches of waste magnets collected from a range of end-of-life applications such as hard disk drives, magnetic resonance imaging machines and loudspeakers were recycled via *Magnet-to-Magnet Recycling Process* [13]. These magnets are generally covered with a corrosion resistive coating such as Ni, Zn, and Al. These coatings were mechanically removed using a blasting apparatus (model 28GL) at a rate of 680 kg/min steel shot before further processing. Aiming at improving coercivity, and thermal stability overall, the optimum Nd₆Dy₂₁Co₁₉Cu_{2.5}Fe composition blend of transitional metal and lanthanides was chosen so to allow the GBE™ (Grain Boundary Engineering) of the main phase. The Nd₆Dy₂₁Co₁₉Cu_{2.5}Fe alloy powder was therefore blended to recycled powder to form a modified grain boundary phase; hereafter, this powder is called grain boundary modified (GBM™)

powder. 5.0 wt% GBM™ powder was added to waste magnet powders to produce new *recycled sintered magnets* suitable for high temperature applications.

Starting waste material with the composition of $\text{Nd}_{21.07}\text{Pr}_{7.14}\text{Dy}_{0.4}\text{Fe}_{69.1}\text{Ga}_{0.11}\text{Zr}_{0.12}\text{Al}_{0.25}\text{Co}_{0.55}\text{Cu}_{0.16}\text{B}_{0.96}\text{C}_{0.07}\text{O}_{0.07}$ wt% was used to produce recycled magnets. The waste magnets blended with the GBM™ powder were first coarsely milled and homogenized using a hydrogen-mixing reactor. This reactor was first evacuated and heated to 80°C before admitting hydrogen at a pressure of 0.9 bar. This stage is completed when the pressure gauge becomes stable, indicating that no more hydrogen is being adsorbed. The material was then transferred to a jet milling machine (model: XDK QLMR 350) to achieve a narrow powder size distribution of 3.5 μm, which was then sieved to remove large oxide particles. Powders were then compacted under a magnetic field to form anisotropic green compacts. The green body was then transferred to a hydraulic chamber and subjected to an isostatic pressure of 100 MPa. The aligned and isostatically pressed green compacts were transferred to a graphite tray under an inert atmosphere and then to a vacuum sintering and annealing furnace. The densities of the recycled Nd-Fe-B sintered magnets were determined by a density balance under a temperature controlled ionized water. Cylindrical magnets (10 mm in diameter and 10 mm in height) were cut from a recycled Nd-Fe-B sintered block using a wire-cutting machine, and the magnetic properties were then measured using a close looped permeameter after pulse magnetizing in a magnetic field of 4 T. Chemical composition is analyzed using inductive coupled plasma (ICP) before, during, and after the recycling process. Scanning electron microscopy (SEM) backscattered electron (BSE) and in-lens secondary electron (IL-SE) observations were carried out using a Carl Zeiss Cross-Beam 1540EsB. Transmission electron microscopy (TEM) was performed using a Titan G2 80-200 TEM with a

probe aberration corrector. Energy dispersive X-ray spectroscopy (EDS) was collected using a FEI Super-X EDX detector.

3. Results

3.1. Magnetic properties

Figure 1 (a) shows demagnetization curves of the waste magnet and the recycled magnet. The waste magnet shows $\mu_0 M_r = 1.38$ T and $\mu_0 H_c = 1.3$ T. The addition of 5.0 wt% GBM™ powder during the recycling process led to the density of 7.55 g/cc with $\mu_0 M_r = 1.29$ T and $\mu_0 H_c = 2.36$ T. The composition of the recycled magnet with 5.0 wt% additive was $\text{Nd}_{21.63}\text{Pr}_{6.43}\text{Dy}_{3.42}\text{Fe}_{64.75}\text{Ga}_{0.1}\text{Zr}_{0.11}\text{Al}_{0.28}\text{Co}_{1.74}\text{Cu}_{0.32}\text{B}_{0.97}\text{C}_{0.12}\text{O}_{0.13}$ wt%. Figure 1 (b) shows coercivity versus temperature for the waste magnet and the recycled magnets. Temperature coefficient of coercivity (β) of the waste magnet and recycled magnet measured from 25 °C to 200 °C were -0.50 %/°C and -0.47 %/°C, respectively. The temperature coefficient of remanence (α) measured with the same temperature range were -0.14 %/°C and -0.11 %/°C for the waste magnet and recycled magnets, respectively. Note that a more remarkable difference in the temperature coefficients of coercivity and remanence was found between the waste and recycled magnets in favor of the recycled magnets when heating up the specimens from 25 °C to 80 °C, *i.e.* α is equal to -0.12 %/°C and -0.02 %/°C, and β is equal to -0.81 %/°C and -0.61 %/°C for the waste and the recycled magnets, respectively.

3.2. Overall microstructure

Figure 2 (a) and (b) show back scattered electron (BSE) SEM images of the waste magnet and recycled magnet, respectively. In-lens secondary electron (IL-SE) SEM images obtained from the same area are shown in Fig. 2 (c) and (d). In the IL-SE images, Nd-rich oxides

can be seen with a dark contrast, and metallic Nd-rich phases are observed with a bright contrast [14]. Note that some voids are observed in the microstructure of the waste magnet as pointed out with yellow arrows in Fig. 2 (a) and (c). In addition, cracks are also observed at some grain boundary region of the waste magnet as marked in Fig. 2 (a). The areal fraction of the metallic Nd and neodymium oxides were calculated for the waste magnet and the recycled magnet based on the contrast of IL-SE images. The areal fraction of metallic neodymium and neodymium oxides in the waste magnet were estimated to be 2.0 and 3.2 %, respectively. In the case of the recycled magnet, the areal fraction of neodymium oxides slightly increases to 4.4 % while that of the metallic Nd phases were estimated to be around 6.1 %. This indicates that the addition of the GBM™ during powder processing mainly increases the areal fraction of metallic Nd that is required to form uniform grain boundary phase [15].

Figure 3 (a) and (b) shows higher magnification BSE-SEM images and SEM-EDS maps of Nd, Co, Cu, O, Dy, and Zr obtained from the waste magnet and the recycled magnet. In the microstructure of the waste magnet, rare earth (RE : Nd+Dy) oxides, one with more enrichment of oxygen and the other with less oxygen can be observed. Nd-Cu rich phase can be seen in the microstructure particularly at the triple junction regions as reported in commercial sintered magnets [16]. Zr-rich phase is found in the Nd-rich phases of the waste magnet. In the case of the recycled magnet, RE-oxides and Zr-rich phase can be seen in some of the Nd-rich grains. The distribution of Cu and Co suggests that beside Nd-Cu rich triple junction phase, Nd-Co-Cu rich phase exist in the microstructure of the recycled magnet. Metallic Nd with small concentration of Cu is also observed in some of the Nd-rich phases in the recycled magnet.

The Nd-rich phases in the recycled magnet was studied more in detail using TEM. Figure 4 (a) shows STEM-EDS maps of Nd, Fe, Cu, Zr, Dy, Co, and O obtained from the recycled

magnet. In this STEM-EDS map, 6 different Nd-rich grains can be found: Nd-Cu rich triple junction phase (grain A), Nd-Co-Cu rich phase (Grain B), RE oxides with different oxygen contents (grain C and D), metallic Nd with some content of Cu (grain E), and Zr-rich phase (region F). In order to provide some more quantitative information regarding to the composition of the constituent phases, four different EDS line profiles, i to iv as marked in Fig. 4 (a), are shown in Fig. 4 (b). Line profile (i) shows that Nd-Cu rich phase has a large Cu content of ~35 at. % without any oxygen enrichment. Line profile (ii) shows that the metallic Nd-Co-Cu rich phase contains ~15 and ~10 at. % of Cu and Co respectively. Line profile (iii) shows REO_x phase and line profile (iv) shows oxide phase with a higher oxygen content. Selected areal diffraction patterns obtained from these 6 different Nd-rich grains are shown in Fig. 5. The Nd-rich phase with large Cu content (RE-Cu rich phase) that are mainly exist in the triple junctions of the recycled magnet, such as grain A, has the fcc structure (Fig. 5 (a)). Note that Sepehri-Amin *et al.* reported NdCu precipitates distributed in metallic Nd-rich phase in the triple junction of as-sintered Nd-Fe-B magnets which changes to Nd-Cu rich triple junction phase after post-sinter annealing [16]. Sasaki et al. also reported fcc structure for Nd-Cu rich triple junction phase of optimally annealed sintered magnets [17], which is in agreement with structure of RE-Cu rich triple junction phase observed in the recycled magnet. The selected area diffraction patterns obtained from Nd-Co-Cu rich phase (grain B marked in Fig. 4 (a)) at two different zone axis of [100] and [411] shown in Fig. 5 (b) indicates that this metallic phase also has the fcc structure. Selected area diffraction patterns obtained from rare earth oxide phase with larger oxygen concentration shown in Fig. 5 (c) obtained from grain C shows that this phase is hcp-RE₂O₃, $P\bar{3}ml$, La₂O₃ type crystal structure. The selected area diffraction patterns obtained from REO_x phase, such as grain D, shows this phase is fcc-REO_x $Fm\bar{3}m$, NaCl type crystal structure. The

selected area diffraction pattern taken from the metallic (Nd+Pr)-rich phase with some minor segregation of Cu, grain E, show this phase is $Fm\bar{3}m$, Cu type structure as shown in Fig. 5 (e). The micro-beam diffraction pattern taken from the Zr-rich phase indicates this phase is $P6_3/mmc$, with the Mg-type crystal structure.

Figure 6 shows more detailed analysis of the Zr-rich phase. Figure 6 (a) shows BSE-SEM and IL-SE SEM images that can be observed from the region where Zr-rich phases exist. A bright and dark contrast of lamellae can be observed in the microstructure. A high magnification STEM-HAADF image and STEM-EDS maps of Fe, Nd, Cu, Zr, and Co obtained from such a region is shown in Fig. 6 (b). Figure 6 (c) shows EDS line profile obtained from the selected region marked in the STEM-HAADF map in Fig. 6 (b), showing the (Nd+Pr)-rich phase with a lamella structure containing 17 at.% of Cu and around 5 at. % of Co while Zr-rich region is mainly Zr with amount of 70 at. %.

Figure 7 (a) and (b) show serial sectioning of SEM-BSE and constructed 3D tomography of Nd-rich phases from recycled magnet (see the supplementary file for 3D tomographic movie). 3D tomography was constructed out of 172 SEM-BSE images with 40 nm depth step. The rare-earth oxide phases are shown with bright contrast while the metallic rare-earth phases are shown with a reddish yellow color. This result shows that Nd-rich intergranular phases are interconnected through the grain boundaries in the recycled magnet.

3.3. Composition of the grain boundary phase

Figure 8 (a) show BSE-SEM images obtained from the recycled magnets. The contrast of the image is enhanced for a better visualization of the grain boundary phase and the interfaces. Thin white contrast through the grain boundaries of the recycled magnet indicates the formation

of the Nd-rich grain boundary phase in the recycled magnet. A weak bright gray contrast can be also observed in the outer region of $\text{Nd}_2\text{Fe}_{14}\text{B}$ grains of the recycled magnet. Figure 8 (b) shows STEM-EDS maps of Dy and Nd obtained from the recycled magnet. It can be seen that Dy is not distributed uniformly in the $\text{Nd}_2\text{Fe}_{14}\text{B}$ grain; there is a Dy-rich shell in the outer region of $\text{Nd}_2\text{Fe}_{14}\text{B}$ grains. This can be found more clearly in the composition profile of Fe, Nd+Pr, Co, Cu, and Dy, shown in Fig. 8 (c) obtained from the rectangular region shown in Fig. 8 (b). The Dy content in the shell region is around 0.8 at.% while that inside of the grains reduces to 0.2 at. % in the recycled magnet.

Figure 9 (a) to (c) show high resolution STEM-HAADF images obtained from three typical grain boundary phases with different tilting angle with respect to the c-axis of $\text{Nd}_2\text{Fe}_{14}\text{B}$ grains in the recycled magnet. The grain boundary phase perpendicular to the c-axis of a $\text{Nd}_2\text{Fe}_{14}\text{B}$ grain shows crystalline structure with a thickness of ~ 3.5 nm (Fig. 9 (a)). However, the grain boundary phase located parallel to the easy axis of $\text{Nd}_2\text{Fe}_{14}\text{B}$ grains show an amorphous structure as shown in Fig. 9 (c). STEM-EDS composition profiles of the grain boundary phases are shown in Fig. 9 (d) to (f). It can be seen that the crystalline grain boundary phase located perpendicular to the easy axis of $\text{Nd}_2\text{Fe}_{14}\text{B}$ grain contains 66.5 at. % of Nd+Pr, 23.8 at. % of Fe, and 5.9 at.% Cu. Fig. 9 (e) shows STEM-EDS line profile obtained from the grain boundary phase with 33 degree tilting angle from c-axis of $\text{Nd}_2\text{Fe}_{14}\text{B}$ grain (Fig. 9 (b)) indicating that the grain boundary phase contain 45 at. % of Nd+Pr, 43 at. % of Fe, and 6.5 at. % segregation of Cu. STEM-EDS line profile obtained from the grain boundary phase located almost parallel to the easy axis of $\text{Nd}_2\text{Fe}_{14}\text{B}$ grain is shown in Fig. 9 (f) indicating that the grain boundary phase contain 27.8 at. % of Nd+Pr and 62.4 at. % of Fe with 5.8 at. % segregation of Cu in the grain boundary phase.

4. Discussion

It was shown that the addition of Nd₆Dy₂₁Co₁₉Cu_{2.5}Fe alloy powder to the recycled Nd-Fe-B powders during the powder processing is an effective way to develop full density recycled Nd-Fe-B magnet. SEM observations showed that addition of Nd₆Dy₂₁Co₁₉Cu_{2.5}Fe alloy powder provide sufficient amount of metallic Nd for the liquid sintering process, resulting in the development of full density recycled Nd-Fe-B magnets. In addition, a uniform Nd-rich phase was found at the grain boundaries of the recycled magnet, which is attributed to the recovery or improvement of the coercivity. The magnetic properties of the recycled magnet ($\mu_0 H_c = 2.36$ T and $\mu_0 M_r = 1.29$ T) are higher than those of the commercial NMX-43SH grade sintered magnet ($\mu_0 H_c = 2.21$ T and $\mu_0 M_r = 1.25$ T) that contains about 4.45 wt% Dy. The overall Dy and oxygen contents in the recycled magnet (Dy = 3.42 wt% and O = 0.13 wt%) are lower than those of the commercial NMX-43SH grade sintered magnet (Dy = 4.45 wt% and O = 0.42 wt%). This indicates that selective addition of Nd and Dy alloy powders as the grain boundary modifier results in the development of the recycled magnets with even superior magnetic properties than that of original Nd-Fe-B sintered magnets.

One question is how the individual elements of the GBM™ are distributed in the microstructure of the recycled Nd-Fe-B magnet. Microstructure analysis of the recycled magnet showed that substantial amount of the additives forms isolated Nd-rich phases containing metallic Nd+Pr, Nd-Cu rich, and Nd-Co-Cu rich phases combined with rare earth oxides. Some part of Dy was found to diffuse into Nd₂Fe₁₄B grains making a thick Dy-rich shell and Dy-lean core structure. STEM-EDS results showed that Dy content in the shell and core region of the Nd₂Fe₁₄B grains in the recycled magnet was 0.8 and 0.2 at. %, respectively. This type of

core/shell structure is commonly observed in the Nd-Fe-B sintered magnets processed from multiple alloy powders [18,19].

Composition profiles obtained from the STEM-EDS analysis of the grain boundary phases of the recycled magnet showed that the one located perpendicular to the c-axis of Nd₂Fe₁₄B grains have a crystalline structure with a higher Nd+Pr content than that of the amorphous grain boundary phase located parallel to c-axis. In fact, this irregular distribution of Nd-rich phase in the grain boundary phase was first reported in the hot-deformed Nd-Fe-B magnets [20-22]. More recently, such a feature was also reported in the commercial Nd-Fe-B sintered magnets [17]. The thickness of the grain boundary phase perpendicular to the c-axis is ~3.7-4.3 nm and that parallel to the c-axis is ~2.4 nm in the recycled magnet. These are thicker

than the exchange length of Nd₂Fe₁₄B phase, 1.97 nm, which is defined as $l_{ex} = \sqrt{\frac{A}{\mu_0 M_s^2}}$,

where A and $\mu_0 M_s$ are exchange stiffness and saturation magnetization of Nd₂Fe₁₄B phase, 8×10^{12} J/m and 1.61 T [23]. The existence of only 23.8 at. % Fe in the grain boundary phase perpendicular to c-axis suggests that the grains are exchange decoupled parallel to c-axis direction while they might be still exchange coupled perpendicular to c-axis direction since ~62.4 at.% Fe exist at the grain boundaries parallel to c-axis of the Nd₂Fe₁₄B grains. Based on these microstructure analysis, it can be concluded that formation of thick grain boundary phase, large Nd content at the grain boundary phase, good interconnection of Nd-rich phases, and the formation of the Dy-rich shell explains the high coercivity in the recycled magnet. Further modification of the microstructure, such as composition modification of the grain boundaries parallel to c-axis will lead to further enhancement of the coercivity.

5. Summary

Full-density recycled Nd-Fe-B sintered magnets were produced by the addition of a new grain boundary phase (GBM™) modifier, Nd₆Dy₂₁Co₁₉Cu_{2.5}Fe alloy powders, to the powders produced from Nd-Fe-B waste magnets. The recycled magnet showed $\mu_0 H_c$ of 2.36 T and $\mu_0 M_r$ of 1.29 T with the net Dy content of 3.42 wt% showed superior coercivity and thermal stability to those of the commercial NMX-43SH grade sintered magnet with Dy content of 4.45 wt%. Detailed microstructure characterizations revealed two main reasons for the substantial enhancement of coercivity in the recycled magnet; the formation of distinct grain boundary phase and enrichment of ~0.8 at. % Dy in the shell of Nd₂Fe₁₄B grains. The former led to the reduction of exchange coupling among Nd₂Fe₁₄B grains in the c-axis direction and the latter enhanced the local anisotropy of the main phase, which explain the high coercivity of the recycled magnet with 5.0 wt% addition of the GBM™.

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Figure 1: (a) Demagnetization curves of starting materials and recycled magnets and (b) coercivity versus temperature of the starting material and recycled magnet.

Figure 2: (a) and (b) Back scattered electron (BSE) SEM and (c) and (d) in-lens secondary electron (IL-SE) SEM images obtained from starting waste magnet and recycled magnet, respectively.

Figure 3: Back scattered electron (BSE) SEM and EDS maps of Nd, Co, Cu, O, Dy, and Zr obtained from (a) starting waste magnet and (b) recycled magnet.

Figure 4: (a) STEM-EDS maps of Nd, Fe, Cu, Zr, Dy, Co, and O obtained from the recycled magnet; and (b) Composition profiles obtained from STEM-EDS line scans i to iv, as marked in (a).

Figure 5: Selected area diffraction patterns obtained from 6 different rare earth (RE)-rich grains marked in Fig. 4a: (a) RE-Cu rich phase, (b) RE-Co-Cu rich phase, (c) RE_2O_3 , (d) REO_x , (e) metallic Nd, and (f) Zr-rich phase.

Figure 6: (a) SEM-BSE and IL images showing lamella contrast from Nd-rich grains obtained from the recycled magnet. (b) STEM-HAADF image and STEM-EDS maps of Fe, Nd, Cu, Zr, and Co obtained from the same area shown in (a). (c) EDS-line profile obtained from selected line marked in (b) showing that the lamella structure contains Zr-rich and (Nd+Pr)-Co-Cu phases.

Figure 7: (a) 3D series of BSE SEM images and (b) 3D tomography of Nd-rich phases for the recycled magnets.

Figure 8: (a) BSE SEM images obtained from the recycled magnet, (b) STEM-EDS maps of Dy and Nd obtained from the recycled magnet. (c) The composition profile of Fe, Nd+Pr, Co, Cu, and Dy obtained from selected line shown in Dy map.

Figure 9: (a) - (c) High resolution STEM-HAADF images obtained from grain boundaries of the recycled magnet with different tilting angle with respect to the easy axis of $\text{Nd}_2\text{Fe}_{14}\text{B}$ grains. (d) - (f) STEM-EDS composition profiles of constituent elements obtained across the Nd-rich grain boundary phases.

Figure 1

Figure 2

Figure 3

a) SEM-EDS maps obtained from waste magnet

b) SEM-EDS maps obtained from recycled magnet

Figure 4

Figure 5

a) RE-Cu rich phase

b) RE-Co-Cu rich phase

c) RE_2O_3

d) REO_x phase

e) Metallic Nd

f) Zr-rich

Figure 7

a)

b)

Figure 6

Figure 8

Figure 9