Permanent Magnet Materials Based on the Rare Earth-Iron-Boron Tetragonal Compounds

By Masato Sagawa, Setsuo Fujimura, Hitoshi Yamamoto, and Yutaka Matsuura; Sumitomo Special Metals Company Ltd., Egawa, Shimamoto-cho, Mishimagun, Osaka 618, Japan and Kenji Hiraga, The Research Institute for Iron, Steel and Other Metals, Tohoku University, Sendai 980, Japan

Abstract

Structural and metallographic studies were carried out on the Nd-Fe-B alloy system as well as the Nd-Fe-B tetragonal compound on which record high energy magnets have been developed using a powder metallurgical technique. The study on the new magnet has also been extended to other R-Fe-B compounds containing various rare earths (R) and to R-Fe-Co-B alloys. The results are as follows:

1. The sintered Nd-Fe-B magnet is composed of mainly three phases, the Nd$_2$Fe$_{14}$B matrix phase plus Nd-rich phase and B-rich phase (~Nd$_2$Fe$_7$B$_6$) as minor phases.

2. Nd$_2$Fe$_{14}$B has the space group of P4$_2$/mnm. The crystal structure of this phase can be described as a layer structure with alternate stacking sequence of a Nd-rich layer and a sheet formed only by Fe atoms. The sheet of Fe atoms has a structure similar to the $\sigma$ phase found in Fe-Cr and Fe-Mo systems.

3. The Nd-rich phase containing more than 95 atoms % Nd, 3-5 atoms % Fe and a trace of B has fcc structure with $a = 0.52$ nm. This phase is formed around grain boundaries of the matrix phase. Nd$_2$Fe$_{14}$B phase has an one-dimensional incommensurate structure with $d = a_0$ and $c = 8c_0$, based on a tetragonal structure with $a_0 = 0.716$ nm and $c_0 = 0.391$ nm.

4. In the as sintered Nd$_{15}$Fe$_{77}$B$_8$ alloy periodic strain contrasts are observed along grain boundaries, which disappear after annealing at 870 K. This may be related to the enhancement of the intrinsic coercivity of the sintered magnet by post sintering heat treatment.

5. Stable R$_2$Fe$_{14}$B phases are formed by various rare earths except La. Of all the R$_2$Fe$_{14}$B compounds, Nd$_2$Fe$_{14}$B has the maximum saturation magnetization as high as 1.57 T. Dy and Tb form R$_2$Fe$_{14}$B phases with the highest anisotropies. Small additions of these elements greatly enhance the coercive force of the Nd$_2$Fe$_{14}$B base magnet.

6. Partial replacement of Fe by Co raises the Curie temperature of the Nd$_2$Fe$_{14}$B compound, which improves the temperature coefficient of the remanence of the magnet. But the intrinsic coercive force is decreased by the Co addition.

Introduction

Rare earth-iron (R-Fe) intermetallics, especially those containing the abundant light rare earths (LRE), have long been studied as candidate materials for high performance permanent magnets. LRE and Fe, however, form few stable intermetallics, and none of these has a high enough Curie temperature required for a practical material and a high uniaxial anisotropy necessary for realizing a high coercivity.

We noticed that there exists a variety of stable compounds in the ternary systems composed of LRE, Fe and small quantities of third elements. Of a number of ternary compounds actually synthesized, a remarkable compound composed of Nd, Fe and B was found: This phase exists in the vicinity of 12 atoms % Nd, 82 atoms % Fe and 6 atoms % B, and has a tetragonal crystal structure. The Curie temperature of this phase is 585K, saturation magnetization is more than 1.35 T and the...
first order magnetocrystalline anisotropy constant amounts to 3.5 MJ/m³ with the easy axis parallel to the c-axis of the tetragonal unit cell. Based on this compound, a very high energy product magnet was developed using a powder metallurgical technique. This magnet, which has an approximate composition Nd₂Fe₁₄B, shows a remanence of 1.23 T, intrinsic coercivity of 960 kA/m, and maximum energy product of 290 kJ/m³. Further investigations have achieved maximum energy products as high as 360 kJ/m³.

Conventional attempts to magnetically harden the rare earth-iron alloys were carried out by forming amorphous or micro-crystalline alloys using sputtering or melt-spinning.

Initially, these magnets aimed at forming a metastable or non-equilibrium state in R-Fe alloy systems. Quite recently, however, Hadjipanayis, et al. and Croat, et al. have noticed that high coercive forces produced in melt spun R-Fe-B alloys originate from the formation of a stable compound which appears to have a tetragonal structure. This compound coincided with that which we found from a different approach.

In the previous paper, we presented the basic properties of the Nd-Fe-B new tetragonal compound, a powder metallurgical process to produce magnets and the properties of these magnets. The present paper describes the results of our investigation following the previous report, which involve the crystal structure of the Nd-Fe-B tetragonal phase, metallography and microstructure of Nd-Fe-B alloys, magnetic properties of R-Fe-B tetragonal compounds composed of various rare earths, and the effects of Co substitution on the magnetic properties of Nd-Fe-B sintered magnets.

Experimental Procedure

All alloys were prepared by induction melting commercially available constituents. The production process of the sintered magnets was reported previously.

For the analysis of the crystal structure, measurements by x-ray diffractometer on powder samples and by rotating-crystal methods on single crystal fragments were made. Powder samples under 200 mesh were prepared by crushing an ingot annealed at 1370-1420 K for 7 days. Single crystal fragments were obtained from crushed ingots which had been slowly cooled in the temperature range from just above to just below the melting point of the alloy. Transmission electron microscopy (TEM) and electron diffraction were used for the determination of the space group of the tetragonal phase as well as for the investigation of the microstructure of the sintered alloy. The samples for the TEM were prepared by combining mechanical and ion polishing. Electron probe micro-analysis (EPMA) was made on sintered samples. For the quantitative micro-analysis, melt-quenched amorphous ribbons were used as standard samples from which quantitative chemical analysis had been made. For thermal analysis, a differential scanning calorimeter (DSC) was used on sintered alloys up to 1070K.

Magnetic measurements were made by vibrating sample magnetometer (VSM) with a maximum magnetic field of 1.2 MA/m. Samples for these measurements were the powders described above. Their easy directions were aligned in a magnetic field of 1.6 MA/m in a small plastic capsule and then congealed by wax. Magnetic anisotropy fields were measured on sintered samples by using pulse magnetic field up to 9.6 MA/m. Permanent magnet properties were measured by a B-H tracer with a maximum magnetizing field of 1.6 MA/m.

Results and Discussion

1. Crystal Structure of Nd-Fe~B Tetragonal Phase

(a) Confirmation of Space Group

Electron diffraction and convergent-beam electron diffraction (CBED) were carried out to determine the space group of the tetragonal Nd-Fe-B compound. As an example, an electron diffraction pattern of the (010) incidence is shown in Figure 1. A systematic absence takes place at h + l = 2n + 1 as seen in the pattern, so that the possible space groups are P₄/n₂, P₄/nmm and P₄/mmm which belong to the point group 4/m, 4 mm and 4/mmm, respectively.

Figure 1 — Electron diffraction pattern of the [010] incidence for the Nd₂Fe₁₄B compound.
Figure 2 shows a zone-axis CBED pattern taken with the same incidence as that of Figure 1, where zero-layer reflections embedded in upper layers are enlarged so as to inspect the fine structures within the reflection disks.

Figure 2 — Zone-axis CBED pattern taken with the same incidence as that of Figure 1.

It is easily found in the pattern that there exist two mirror planes perpendicular to the \( l \) and \( h \) axes, as marked with \( m \). The result indicates that the point group is \( 4/m \) and the corresponding space is \( P4_2/mnm \).

(b) Consideration of Crystal Structure.

From the density of 7.55 Mg/m\(^3\) and an analyzed composition of \( \text{Nd}_{12}\text{Fe}_{82}\text{B}_6 \) for cast ingots and the space group of \( P4_2/mnm \), it may be concluded that the structure contains 8 Nd, 56 Fe and 4 B atoms in the unit cell. The formula for the tetragonal compound is \( \text{Nd}_2\text{Fe}_{14}\text{B} \).

\[ \text{Nd}_2\text{Fe}_{14}\text{B} \]

\[
\begin{align*}
16 \text{ Fe atoms in } 16(k) & \quad x,y,z; x \approx 7/15, y \approx 2/15, z \approx 1/6 + \Delta Z_1 \\
16 \text{ Fe atoms in } 16(k) & \quad x,y,z; x \approx 11/15, y \approx 1/15, z \approx 1/6 + \Delta Z_2 \\
8 \text{ Fe atoms in } 8(j) & \quad x,z; x \approx 2/5, z \approx 1/6 + \Delta Z_3 \\
8 \text{ Fe atoms in } 8(j) & \quad x,z; x \approx 11/60, z \approx 1/4 \\
4 \text{ Fe atoms in } 4(0) & \quad 0,0,z; z \approx 1/6 + Z_4 \\
4 \text{ Fe atoms in } 4(0) & \quad 0,1/2,0 \\
4 \text{ Nd atoms in } 4(f) & \quad 0,0,2; z \approx 1/6 + Z_4 \\
4 \text{ Nd atoms in } 4(f) & \quad x,x,0; x \approx 1/4 \\
4 \text{ B atoms in } 4(f) & \quad x,x,0; x \approx 3/8
\end{align*}
\]

where the parameters \( \Delta Z_i (i = 1 \ldots 4) \) have not yet been precisely estimated. The hexagons in the \( \sigma \)-phase structure of Figure 3 lie on the \((001) \) planes, whereas those in this structure are inclined from the \((001) \) because of considerable amounts of the parameters \( \Delta Z_i \).

During the course of the preparation of this manuscript, we became aware of a previous report on the structure of \( \text{Nd}_2\text{Fe}_{14}\text{B} \) by Herbst, Croat, Pinkerton and Yelon. Our results are in excellent agreement with this previous structure analysis by neutron diffraction.

X-ray diffraction data from powder and single crystals exhibit a characteristic intensity distribution in the \((hko)\) reciprocal plane with very strong 410, 330, 550, 720, 820 and 660 reflections. This feature is very similar to that of \( \sigma \)-phase structure found in the Fe-Cr and Fe-Mo systems. The \( \sigma \)-FeCr has a tetragonal structure with lattice parameters of \( a = 0.880 \) nm and \( c = 0.454 \) nm. Note that the value \( a = 0.880 \) nm of \( \sigma \)-FeCr is closely similar to the lattice parameter \( a = 0.882 \) nm of \( \text{Nd}_2\text{Fe}_{14}\text{B} \). The \( \sigma \)-phase structure projected down along the \( c \) direction is illustrated in Figure 3.

In this structure, two slightly distorted hexagons with a relative orientation of \( 90^\circ \) are located on the layers of different \( z \) coordinates. It is readily apparent that the atomic configuration relating to the two hexagons is similar to that of \( \text{SmCo}_5 \) structure.

The present analysis of our diffraction data predicts that the following positions in the space group \( P4_2/mnm \) are occupied:

- 16 Fe atoms in \( 16(k) \) \( x,y,z \); \( x \approx 7/15, y \approx 2/15, z \approx 1/6 + \Delta Z_1 \)
- 16 Fe atoms in \( 16(k) \) \( x,y,z \); \( x \approx 11/15, y \approx 1/15, z \approx 1/6 + \Delta Z_2 \)
- 8 Fe atoms in \( 8(j) \) \( x,z \); \( x \approx 2/5, z \approx 1/6 + \Delta Z_3 \)
- 8 Fe atoms in \( 8(j) \) \( x,z \); \( x \approx 11/60, z \approx 1/4 \)
- 4 Fe atoms in \( 4(0) \) \( 0,0,z \); \( z \approx 1/6 + Z_4 \)
- 4 Fe atoms in \( 4(0) \) \( 0,1/2,0 \)
- 4 Nd atoms in \( 4(f) \) \( 0,0,2 \); \( z \approx 1/6 + Z_4 \)
- 4 Nd atoms in \( 4(f) \) \( x,x,0 \); \( x \approx 1/4 \)
- 4 B atoms in \( 4(f) \) \( x,x,0 \); \( x \approx 3/8 \)

Figure 3 — Crystal structure of the a-phase projected on (001) plane, blue atoms lie on the first and third layers and red atoms on the second layer.

It is not possible to determine B atom positions from x-ray diffraction data, but it is supposed that 4 B atoms are located on the Nd-rich planes of \( z = 0 \) and \( 1/2 \). The structure can be described as a layer structure with alternate stacking sequence of a Nd-rich layer and a sheet of \( \sigma \)-like structure formed by all the Fe atoms except for the \( 4(0) \) positions. The detailed results will be presented elsewhere.
2. Metallography and Microstructure of Nd-Fe-B Permanent Magnets

The previous paper\(^1\) showed that high coercivities and high energy products were obtained for compositions with both Nd and B slightly rich of the stoichiometric composition of Nd-Fe-B tetragonal phase (Nd\(_2\)Fe\(_{14}\)B), and that this alloy is comprised mainly of two phases, namely, the tetragonal matrix and a Nd-rich phase containing more than 50 atoms \% Nd. Figure 4 shows an x-ray composition micrograph of a sintered alloy of Nd\(_3\)Fe\(_7\)B\(_8\), the standard composition for the permanent magnet.

The Nd-rich phase (white area) is found along grain boundaries or at triple points. This phase is easy to be lost during sample polishing due to its high chemical reactivity. The standard alloy has another minor phase which contains much B compared with the matrix. This phase is difficult to distinguish from the matrix phase by optical microscopy. Much less B-rich phase is observed than Nd-rich phase. Figure 4 shows an area where the B-rich phase concentration is exceptionally high. Quantitative analysis by EPMA shows that the Nd-rich phase consists of Nd ~ 95 atoms \%, Fe 3 ~ 5 atoms \% and a trace of B, while B-rich phase has approximately Nd\(_3\)Fe\(_7\)B\(_8\).

![Figure 4 — X-ray composition micrograph of a sintered alloy of Nd\(_3\)Fe\(_7\)B\(_8\).](image)

Typical x-ray diffraction spectrum of the standard Nd\(_3\)Fe\(_7\)B\(_8\) alloy is shown in Figure 5. In this spectrum, several reflections from the Nd-rich and B-rich phases were detected which increase in intensity with increasing Nd and B contents in the alloy, respectively. The x-ray diffraction analysis by rotating crystal method on a single crystal fragment of Nd\(_3\)Fe\(_7\)B\(_8\) has shown that this phase has an one-dimensional incommensurate structure with \(a = a_0\) and \(c = 8c_0\) based on a tetragonal structure with \(a_0 = 0.712\) nm and \(c_0 = 0.391\) nm.

![Figure 5 — X-ray diffraction spectrum of Nd\(_3\)Fe\(_7\)B\(_8\) alloy.](image)

Typical electron micrographs for the Nd-rich phase and B-rich phase in the Nd\(_3\)Fe\(_7\)B\(_8\) sintered alloy are shown in Figure 6. In most of the grains of the matrix phase, few defect contrasts were observed. The diffuse contrast in Figure 6 (a) seems to be due to the surface roughness of the sample. Energy dispersive x-ray analysis (EDXA) has shown that the area (A) in Figure 6 (b) contains more than 90 atoms \% Nd.

![Figure 6 — Electron micrographs for matrix phase(a), Nd-rich fcc phase (A in (b)) and grain boundary phase (B in (b)).](image)
As shown in Figure 7 (a), this area has fcc structure with a ~ 0.52 nm. The spacing of (111) plane of this structure is 0.3 nm, which is approximately equal to the spacing corresponding to the reflection of Nd-rich phase in the x-ray diffraction spectrum (Figure 5).

Therefore, it may be concluded that the Nd-rich phase composed of Nd and a small amount of Fe does not have dhcp but fcc structure in Nd15Fe77B8 alloy. Selected area electron diffraction pattern of the thin layer (B) in Figure 6 (b) shows diffuse rings (Figure 7 (b)) which coincide with those expected from a polycrystal of the fcc Nd-rich phase.

But the Nd concentration has not been confirmed by EDXA in this thin layer.

Differential scanning calorimeter (DSC) analysis of the standard alloy is shown in Figure 8. The cusps at 585 K and 950 K correspond to the Curie temperature of the Nd2Fe14B phase and the eutectic temperature between Nd-rich phase and matrix phase, respectively.

From the analysis described above, a phase diagram around Nd2Fe14B up to ~920 K is proposed in Figure 9. It is likely that, above ~920 K Nd-Fe-B ternary liquid is formed as a result of the eutectic reaction between Nd2Fe14B and Nd metal.

As suggested in the previous paper, the eutectic reaction between Nd2Fe14B and Nd metal phases plays an important role for enhancing coercivity of the Nd-Fe-B permanent magnets. It seems that the eutectic reaction enables liquid phase sintering, which leads to densification of the alloy without significant grain growth.

The optimum temperature for the post sintering heat treatment coincides with the eutectic temperature exactly. The optimum temperature is just below the eutectic temperature and the coercive force decreases rapidly on heating above this temperature. Below the eutectic temperature, phase separation takes place. It is considered therefore that the enhancement of coercive force by post sintering heat treatment results from the removal of defects from the grain boundary area concomitant with the phase separation.
From TEM studies, periodic strain contrasts are observed along grain boundaries for as-sintered samples, whereas they disappear after annealing at 870 K (Figure 10). Detailed TEM investigations are necessary to understand the mechanism for the effect of post sintering heat treatment.

Figure 10 — Electron micrographs for Nd$_{15}$Fe$_{77}$B$_8$ magnet as-sintered (a) and after annealed at 870K (b).

### 3. Magnetic Properties of R$_2$Fe$_{14}$B Intermetallic Compounds Containing Various Rare Earths

It has been confirmed that R$_2$Fe$_{14}$B tetragonal phases are formed for almost all rare earths except La. A La$_2$Fe$_{14}$B cast alloy has much primary iron and no tetragonal phase is detected by x-ray diffraction. Lattice parameters, density ($D$), saturation magnetization ($I_s$), magnetic moment per Nd$_2$Fe$_{14}$B formula unit ($M$), and magnetic anisotropy field ($H_a$), measured at room temperature, and Curie temperature ($T_c$) are listed in Table 1. From Pr to Tm, both $a_0$ and $c_0$ decrease with increasing atomic number because of the lanthanide contraction. The different behavior of Ce is attributed to the loss of its one 4f electron to the conduction band, resulting in a nearly tetravalent ion.

The saturation magnetization of the R$_2$Fe$_{14}$B compounds changes in the same manner as in other rare earth transition metal compounds, where the magnetization of the LRE sublattice couples ferromagnetically to the magnetization of the transition metal sublattice. The opposite is true for the heavy rare earth (HRE) elements, rendering these latter materials ferrimagnetic.

#### Table 1 — Lattice Parameter, Density and Magnetic Properties of R$_2$Fe$_{14}$B Compounds

<table>
<thead>
<tr>
<th>Compound</th>
<th>Lattice Parameter</th>
<th>$D$ (Mg/m$^3$)</th>
<th>$I_s$ (T)</th>
<th>$M$ (us/F.U.)</th>
<th>$H_a$ (MA/m)</th>
<th>$T_c$ (K)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ce$<em>2$Fe$</em>{14}$B</td>
<td>0.877 1.211</td>
<td>7.81</td>
<td>1.16</td>
<td>22.7</td>
<td>3.7</td>
<td>424</td>
</tr>
<tr>
<td>Pr$<em>2$Fe$</em>{14}$B</td>
<td>0.882 1.225</td>
<td>7.47</td>
<td>1.43</td>
<td>29.3</td>
<td>10</td>
<td>564</td>
</tr>
<tr>
<td>Nd$<em>2$Fe$</em>{14}$B</td>
<td>0.882 1.224</td>
<td>7.55</td>
<td>1.57</td>
<td>32.1</td>
<td>12</td>
<td>585</td>
</tr>
<tr>
<td>Sm$<em>2$Fe$</em>{14}$B</td>
<td>0.880 1.215</td>
<td>7.73</td>
<td>1.33</td>
<td>26.7</td>
<td>basal</td>
<td>612</td>
</tr>
<tr>
<td>Gd$<em>2$Fe$</em>{14}$B</td>
<td>0.879 1.209</td>
<td>7.85</td>
<td>0.86</td>
<td>17.3</td>
<td>6.1</td>
<td>661</td>
</tr>
<tr>
<td>Tb$<em>2$Fe$</em>{14}$B</td>
<td>0.877 1.205</td>
<td>7.93</td>
<td>0.64</td>
<td>12.7</td>
<td>28</td>
<td>639</td>
</tr>
<tr>
<td>Dy$<em>2$Fe$</em>{14}$B</td>
<td>0.875 1.200</td>
<td>8.02</td>
<td>0.65</td>
<td>12.8</td>
<td>25</td>
<td>602</td>
</tr>
<tr>
<td>Ho$<em>2$Fe$</em>{14}$B</td>
<td>0.875 1.199</td>
<td>8.05</td>
<td>0.86</td>
<td>17.0</td>
<td>20</td>
<td>576</td>
</tr>
<tr>
<td>Er$<em>2$Fe$</em>{14}$B</td>
<td>0.874 1.196</td>
<td>8.24</td>
<td>0.93</td>
<td>18.1</td>
<td>basal</td>
<td>554</td>
</tr>
<tr>
<td>Tm$<em>2$Fe$</em>{14}$B</td>
<td>0.874 1.195</td>
<td>8.13</td>
<td>1.09</td>
<td>21.6</td>
<td>basal</td>
<td>541</td>
</tr>
<tr>
<td>Y$<em>2$Fe$</em>{14}$B</td>
<td>0.874 1.204</td>
<td>6.98</td>
<td>1.28</td>
<td>25.3</td>
<td>3.1</td>
<td>565</td>
</tr>
</tbody>
</table>
According to a Mossbauer study for a Nd$_2$Fe$_{14}$B compound, the spectrum can be resolved into six non-equivalent subspectra and gives values of 21.4 - 27.6 MA/m, and a mean value of 23.6 MA/m for the magnetic hyperfine field of the Fe atom at room temperature. Applying the Gubbens' relation between the magnetic hyperfine field, $H_a$, and the magnetic moment $\mu_{Fe}$ for the Y-Fe series ($\mu_{Fe} = I_{\mu}_B \times H_a/11.7$ kA/m), the mean value of $\mu_{Fe}$ is estimated to be about 2.02 $\mu_B$ which increases to 2.12 $\mu_B$ at 0 K if the Brillouin function with $S = 1$ is applied. This value is as large as for pure iron. From $\mu_{Fe} = 2.02 \mu_B$ at room temperature, and the saturation moment, $32.1 \mu_B$ per Nd$_2$Fe$_{14}$B formula unit, we calculate $\mu_{Nd} = 1.9 \mu_B$. The huge magnetocrystalline anisotropies of the R$_2$Fe$_{14}$B compounds are attributed to the large unquenched R moments at room temperature.

Of the R$_2$Fe$_{14}$B compounds, the Curie temperature is highest for Gd and decreases on its both sides with the change of atomic number. This implies that the exchange in interaction between R and Fe moments enhances the Curie temperature of the compounds. High Curie temperature of R$_2$Fe$_{14}$B compounds compared with R-Fe binaries might result from the magnetic itinerancy as well as the changes in Fe-Fe interatomic distances or the coordination number.

The magnetocrystalline anisotropy of the R$_2$Fe$_{14}$B compounds is high for R with non-zero orbital momentum. It is therefore considered that the anisotropy of the R$_2$Fe$_{14}$B compounds mainly arises from the single-ion anisotropy of the R ion through the action of crystalline fields. But it is noted that Y$_2$Fe$_{14}$B and Gd$_2$Fe$_{14}$B have relatively strong anisotropy where no contribution from the 4f orbital momentum expected. Because these compounds have easy direction of magnetization parallel to the c-axis, the Fe-sublattice seeks the easy axis of magnetization. Sm$_2$Fe$_{14}$B shows a strong easy plane, which is in contrast to RCo$_5$ system.

The coercive force of the Nd-Fe-B magnets can be increased by HRE additions such as Tb or Dy. As an example, the demagnetization curve of a Nd-Fe-B magnet containing small amount of Dy is shown in Figure 11.

![Figure 11 — Demagnetization curve of Nd$_{13.5}$DY$_{1.5}$Fe$_{77}$B$_8$ sintered magnet.](image)

### 4. Nd$_{15}$(Fe$_{1-x}$Co$_x$)$_{77}$B$_8$ Pseudo-ternary Alloys Magnetic Properties

The temperature coefficient of the remanence of the Nd-Fe-B magnet can be improved by Co addition. This is caused by the increase in the Curie temperature of the Nd$_2$Fe$_{14}$B compound by the replacement of Fe by Co (Figure 12). In Table 2, permanent magnet properties, Curie temperature and temperature coefficient of Br are listed for Nd$_{15}$(Fe$_{1-x}$Co$_x$)$_{77}$B$_8$ sintered magnets with $x = 0$, 0.1 and 0.2. As shown in Table 2, Co addition reduces the coercivity of the magnet, which is possibly due to the diminishing of the magnetocrystalline anisotropy of the tetragonal phase and to the formation of other Co-base compounds as minor phases.

<table>
<thead>
<tr>
<th>Table 2 — Magnetic properties of sintered Nd$<em>{15}$(Fe$</em>{1-x}$Co$<em>x$)$</em>{77}$B$_8$ magnets</th>
</tr>
</thead>
<tbody>
<tr>
<td>$B_r$ (T)</td>
</tr>
<tr>
<td>Nd$<em>{15}$Fe$</em>{77}$B$_8$</td>
</tr>
<tr>
<td>Nd$<em>{15}$ (Fe$</em>{0.9}$Co$<em>{0.1}$)$</em>{77}$B$_8$</td>
</tr>
<tr>
<td>Nd$<em>{15}$ (Fe$</em>{0.8}$Co$<em>{0.2}$)$</em>{77}$B$_8$</td>
</tr>
</tbody>
</table>

Replacement of Fe by more than 30 atoms % Co in the alloy rapidly decreases the intrinsic coercivity and deteriorates the squareness of the intrinsic demagnetization curve as well.

In most of the series of R(Fe$_{1-x}$Co$_x$)$_n$ an increase of $x$ leads to an initial increase of the saturation magnetization. After passing through a maximum, further increase of $x$ brings about a continuous decrease.
Figure 12 — Curie temperature versus Co content in Nd₂(Fe₁₋ₓCox)₁₄B system.

In the series of R₂(Fe₁₋ₓCox)₁₄B, it seems that the maximum is located around x = 0.1. The temperature coefficient of the remanence of the Nd₂Fe₁₄B₈ magnet decreases by approximately one-half when 20% Fe is replaced by Co. Thus, several disadvantages of the Nd-Fe-B magnet can be improved by the addition of heavy rare earths, Co, and other elements. Compositional modifications also make it possible the realization of (BH)ₘₐₓ higher than 360 kJ/m³ or 45 MGOe. Structural and metallographic studies presented here will shed light on the origin of high Curie temperature and strong magnetic anisotropy of Nd-Fe-B intermetallic compounds as well as the mechanism of the high coercivity of the new magnet.

Acknowledgement

We would like to acknowledge the continuing support and encouragement of President N. Okada and Vice President T. Ogura and other people concerned. We also wish to thank Dr. A. Higuchi and A. Hamamura for their guidance and many helpful suggestions.

Publication History

References


9. N. C. Koon and B. N. Das; “Magnetic properties of amorphous and crystallized (Fe0.82B0.18)0.9Tb0.05La0.05,” Applied Physics Letters 39, pp. 840 (1981), doi:10.1063/1.92578

10. N. C. Koon, B. N. Das and J. A. Goehagan; “Composition dependence of the coercive force and microstructure of crystallized amorphous (Fe3Bx)0.9Tb0.05La0.05 alloys,” IEEE Transactions on Magnetics, Volume 18, pp. 1448 (1982), doi:10.1109/TMAG.1982.1061968


