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The current and future status of rare earth permanent magnets

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ABSTRACT

With their excellent magnetic performance, Nd-Fe-B magnets have a large share of the permanent magnet market, even after the sudden rise in the price of rare earths in 2011, which have been supported by magnet manufacturers' efforts to save heavy rare earths. The demand for high performance magnets is expected to increase, and both magnet users and manufacturers therefore welcome alternative magnets containing less or no rare earths. A knowledge of thermodynamics and understanding of the mechanism of coercivity of Nd-Fe-B may become very useful to develop processes for new magnets. Furthermore, cost-performance must never be neglected in practical use.

Permanent magnets are materials used for conversion between electric and mechanical energy by generating a magnetic field without the need for electric power. They are one of a key materials in high performance, minimization, and high efficiency of appliances and electric apparatuses such as air conditioners, refrigerators, traction motors and generators of electric, fuel cells, hybrid and plug-in hybrid vehicles (referred to generally as 'xEVs'), and wind turbines.

The demand for high performance magnets is expected to increase due to increased production of permanent magnet machines and devices with the need to protect the global environment, i.e. to reduce CO_2 emissions resulting in global warming. From the environmental point of view, internal-combustion engines mounted in vehicles will be replaced promptly by electric motors, expected to exceed gasoline and diesel engines in sales by 2040 [1]. The governments of Great Britain and France have already announced a ban on traditional vehicle engines by 2040, and other European countries, India and China are following this trend. At present, sales of xEVs has reached approximately 10 million vehicles, while in 2050, the figure is expected to be 150 million [2]. Most of these xEVs will be equipped with permanent magnet motors. If 1 kg of Nd magnets is used per vehicle, 150,000 tons of the Nd magnets will be required for the traction motors alone, and the question therefore arises as to whether or not this can be achieved with Nd magnets alone. New magnets which replace or coexist with Nd magnets will be necessary.

Renewable energy such as wind power is also important in environmental conservation. Application in wind turbine is indeed growing [3]. The power output of per generator is increasing, particularly in offshore wind power generation [4]. Nd magnets are effective in reducing the size of such high-output generators. The weight of magnets used for a generator is considerable, although it depends on the generating systems (direct drive or with gearboxes to increase speed). If all of generators use permanent magnets, 1 to 2 million tons will be used for wind turbines by 2050, when cumulative installed capacity is expected to be 2500 GW [3].

In 2011, the sudden rise in prices sent a shockwave through the world market. Prices of Nd and Dy increased ten-fold, and were a deadly blow to the magnet manufacturers who were forced to raise the prices of rare earth magnets. Since then, in addition to high performance, prices of the Nd magnets have become a serious issue. Many magnet users are anxious about the sustainable supply of magnets, and in fact, their requirement for permanent magnets is basically rare earth (RE) free magnets. If this is difficult, their alternative requirement is magnets containing less RE than the $Nd_2Fe_{14}B$ based magnets, such as $ThMn_{12}$-type magnets and Ce-substituted $Nd_2Fe_{14}B$ magnets. If even these requirements prove difficult, the minimum requirement is heavy rare earth (HRE) free or less-HRE $Nd_2Fe_{14}B$ based magnets. Magnet manufacturers have only been able to meet the last requirement through investigations into enhancing coercivity without, or with less HREs. The following techniques have been developed for this purpose.

a) Grain size refinement
b) Modification of the grain boundary phase
c) The Grain Boundary Diffusion process

This paper introduces the current status of mass-produced RE magnets by focusing on the three techniques above. The necessary conditions for future magnets are also described from the point of view of industrial production.

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It is well-known that magnets with a small grain size exhibit high coercivity. Fig. 1 shows an example of the dependence of coercivity on mean grain size. The data has been obtained from the author's laboratory samples. The fine powder of Nd-Fe-B is very combustible and the most important point is to eliminate the air during the pressing process when preparing fine grained magnets. In order to avoid oxidation, a sealed system has been used for the past 20 years at least in factories in Japan. Unfortunately, magnets with smaller grains exhibit lower remanence as shown in Fig. 2, in which remanence of the same magnets shown in Fig. 1 is plotted against coercivity. The decrease in remanence occurs because alignment of fine particles decreases with decreasing particle size, due to increasing friction. This in turn results in high pressure during pressing which disrupts magnetic alignment. The torque necessary to rotate the fine particles decreases as well. Furthermore, the slope shown in Fig. 2 is almost the same as that when Dy is added to ordinary magnets. Since production of such fine powder requires considerable time for jet milling, the addition of Dy, rather than excessive grain size refinement, was chosen to enhance coercivity. After the rare earth crisis, magnet manufacturers no longer consider refinement to approximately 3 μm to be excessive, and Dy-free magnets with a coercivity of higher than 1280 kA m^{-1} are currently manufactured using grain refinement. Hot-deformed magnets show better values for coefficient of temperature dependence of coercivity β than those of sintered magnets [5]. β is defined generally as $\beta = [H_{cJ}(T) - H_{cJ}(R.T.)] / H_{cJ}(R.T.) \times 100$ (%), where $H_{cJ}(T)$ is the coercivity at an elevated temperature, T°C (e.g. T = 140), and $H_{cJ}(R.T.)$ is that at room temperature. Their grains are smaller than 1 μm, and behave as single domain particles. The reason for good β values is believed to be related to this small grain size. This may be the superior point of hot-deformed magnets and many researchers focus on fine-grained magnets to obtain high coercivity at elevated temperatures.

Magnetic properties of magnets with a modified grain boundary phase were reported in 2014 by Yamazaki et al. [6]. The significant characteristic of these magnets is the inclusion of the $R_6(Fe,Ga)_{14}$ phase as one of the grain boundary phases. In 1988, Shimoda et al. reported that Pr-Fe-B-Cu hot-pressed ingots exhibited high magnetic properties and that the addition of Cu was effective in enhancing coercivity [7]. The magnets included a new grain boundary phase and the crystal structure and intrinsic magnetic properties were firstly investigated by Kajitani et al. [8]. The new phase was a $Pr_6Fe_{13}Cu$ compound and the thick grain boundary phase was observed. $R_6(Fe,M)_{14}$ containing sintered magnets were also investigated by Velicescu [9], Knoch et al. [10]. A binary of rare earth and iron does not form a R_6Fe_{14} phase, and the third element, M is necessary to stabilize the crystal structure of $R_6(Fe,M)_{14}$. In addition to Cu, Sn [9,11], Ga [12], Al [13] and Si [14] were also reported as stabilizing elements.

It has been known for some time that $R_6(Fe,M)_{14}$-containing magnets exhibits higher coercivity, however a large amount of the

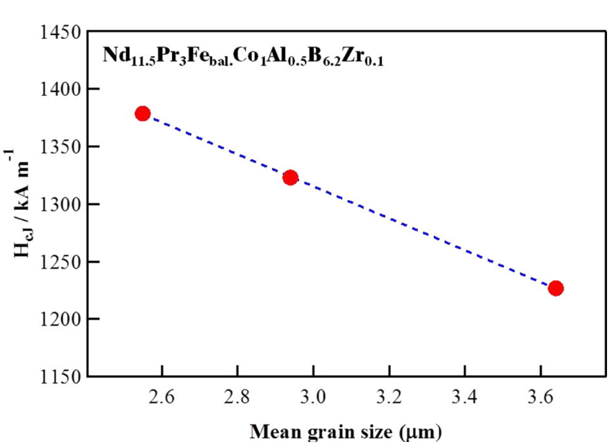

Fig. 1. Dependence of coercivity on the mean grain size for Nd-Fe-B sintered magnets.

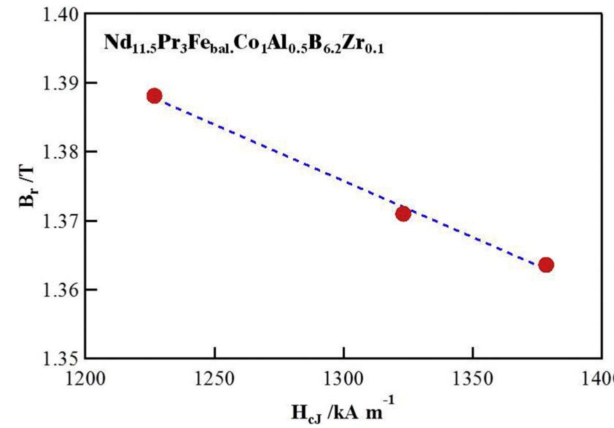

Fig. 2. Remanence plotted against coercivity of Nd-Fe-B sintered magnets with various mean grain sizes.

$R_6(Fe,M)_{14}$ phase was necessary, resulting in a lower remanence than that of ordinary magnets containing Dy. Thus, almost no researchers showed significant interest in magnets containing $R_6(Fe,M)_{14}$. After the sudden rise in the price of rare earths in 2011, we focused our attention on this type of magnet again. The use of sophisticated processes, allowed us to improve the dispersion of the $R_6(Fe,M)_{14}$ phase, and reduce the volume fraction of the $R_6(Fe,M)_{14}$ phase to obtain a high coercivity. At present, the remanence is now approaching that of conventional magnets containing Dy.

Outstanding progress has recently been made in microstructural observations. Sasaki et al. reported on the microstructures of magnets containing $R_6(Fe,M)_{14}$ in detail [15], and Sepehri-Amin et al. have reported that some grain boundary phases in ordinary magnets contain a considerable amount of Fe, and suggested that it is ferromagnetic [16]. Compared to such conventional magnets, magnets containing $R_6(Fe,M)_{14}$ have a thicker and non-ferromagnetic grain boundary phase. Judging from their observations, the high coercivity of magnets containing $R_6(Fe,M)_{14}$ would be due to the better isolation of ferromagnetic $R_2Fe_{14}B$ grains. A very high coercivity of over 1600 kA m^{-1} can be achieved for magnets containing $R_6(Fe,M)_{14}$ [17].

The third development among magnet manufacturers was the Grain Boundary Diffusion (GBD) process [18,19], in which a very small amount of HREs is used. The conventional magnets are prepared and a HRE is then supplied from the surface of the magnet. After diffusion heat treatment, Dy or Tb forms HRE-rich $R_2Fe_{14}B$ shells at the surface of $R_2Fe_{14}B$ grains. The thickness of the heavy rare earth rich shell depends on the conditions of the diffusion treatment and the portion of the magnet (i.e. near to surface, center). In practice, a shell thickness of several nanometers is sufficient to enhance the coercivity [20,21]. The absorbed HRE content after diffusion is therefore typically very low at less than 1 mass%.

Coercivity increases drastically following the GBD process as shown in Fig. 3. But the remanence remains unchanged due to the low HRE content. Fig. 4 represents remanences and coercivities of the magnets shown in Fig. 3. Remanence of the GBD processed magnet is higher by 100 mT than that of the conventional magnet with the same coercivity. The GBD process can be considered as a way to use HREs effectively, and as a result, it is possible to save approximately 4 mass% of Dy, with the primary advantage of reducing remanence by 100 mT in conventional magnets.

By applying the GBD process to the "fine-grained" magnets, Dy-free magnets with a coercivity of higher than 1800 kA m^{-1} can be manufactured. The GBD processed magnets exhibit higher remanence than that of conventional magnets with the same coercivity. This is the manifest difference of grain size refinement and modification of the grain boundary phase. The high remanence contributes high-

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