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Review of the status of high performance permanent magnet materials and devices

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The discovery of neodymium-iron-boron magnets by Sagawa et al. and by Croat et al. in 1983 provided a large impetus for an accelerated pace of research and development in permanent magnet materials and devices. As a result, our understanding of the origin of magnetocrystalline anisotropy, spin-reorientation behavior, microstructure and coercivity mechanisms have significantly improved. Several devices such as motors, actuators and magnetic separators have been designed and constructed employing these magnets with improved performance. New materials and devices are being constantly explored.

In this article, we present an overview of the status of these materials and devices. Selected examples include permanent magnets with energy products close to 47 MGOe, thermally stable Sm_2Co_{17}-type magnets, design of a slotless motor and a few new materials.

INTRODUCTION

Rare earth - transition metal intermetallics exhibit a variety of interesting fundamental properties, such as a high Curie temperature, large magnetization and good magnetocrystalline anisotropy [1]. Efforts to fabricate permanent magnets from these intermetallics have culminated in the discovery of three families of magnets with energy products of ~28 MGOe (for $SmCo_5$), 31 MGOe (for Sm_2Co_{17}) and in excess of 40 MGOe (for $Nd_2Fe_{14}B$). These magnets have been extensively used in the design and construction of devices such as actuators, motors, generators, bearings, and magnetic separators. In these applications, the high energy magnets have helped improve energy efficiency, enhance performance characteristics and reduce the size of the devices, provided efforts are made to design these devices under optimum conditions. In this overview, we outline the recent progress made in the fabrication of very high energy magnets made from Pr-Fe-B compositions employing a dry milling technique and the fabrication of 2:17 magnets with a reasonably good thermal stability. We also discuss a few illustrative applications of these magnets such as in the design of a slotless motor. Finally, we present examples on new materials and processing.

VERY HIGH ENERGY MAGNETS

$Nd_2Fe_{14}B$-based permanent magnets have been extensively examined by a number of groups. However, not too much work has been reported on their praseodymium counterparts, except the work relating to hot pressing techniques. In some respects, the intrinsic properties of $Pr_2Fe_{14}B$ are superior to those of $Nd_2Fe_{14}B$. For example, the magnetocrystalline anisotropy energy of the former at room temperature is about 10% larger than that of $Nd_2Fe_{14}B$. There are also some subtle differences in the quantum mechanical features of the 2:14:B phase, in which the non-Kramers Pr ion and the Kramers Nd ion behave differently. This affects the crystal field interaction of the two ions, imparts a larger magnetocrystalline anisotropy to $Pr_2Fe_{14}B$ and may be related to the spin reorientation phenomenon which occurs in $Nd_2Fe_{14}B$ at ~140 K but not in $Pr_2Fe_{14}B$. Keeping all these factors in mind, we have recently fabricated magnets based on the compositions $Pr_{14.0+x}Fe_{bal}B_{6.5}$, where x is varied from 0 to 1.0. Further, the magnets were fabricated under a variety of processing techniques. Initially, the effect of long-term annealing of the cast ingots on the microstructural and magnetic properties of the alloys and finished magnets were investigated. Thermomagnetic analyses showed a reduction in the concentration of α-Fe for samples annealed for long duration. Magnets were fabricated by employing a milling technique in which comminution was effected by ball milling under argon atmosphere. Magnet properties were compared with those obtained through conventional processing techniques using toluene as a medium for milling. The results clearly showed that magnets with energy products close to 47 MGOe could be easily obtained by the dry milling techniques with the alloys which are relatively free from α-iron impurities. A typical hysteresis loop of such a magnet is shown in Figure 1. Optical micrographs clearly showed that the grains of $Pr_2Fe_{14}B$ are large, the grain boundaries are clearly discernible and that the secondary phases such as $PrFe_4B_4$, Pr_2O_3 and other lower oxides of Pr are much lower compared to the magnets obtained by conventional wet milling technique. Thus, the volume fraction of the matrix phase in these magnets is much larger and

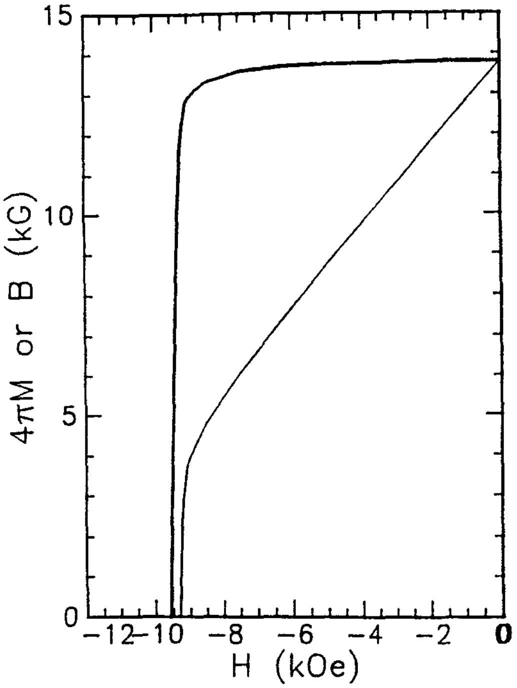

Fig. 1 Demagnetization curve of a Pr-Fe-B magnet

contributes to the higher remanence. It may also be remarked that the concentration of off-stoichiometric oxides is much smaller and that they occur as a thin boundary layer around the matrix phase. This feature undoubtedly contributes to the development of high coercivity. Oxygen content of the magnets were also carefully monitored; the finished magnets obtained by dry milling technique contain ~2200 ppm oxygen by weight while magnets obtained by the wet milling technique have an oxygen concentration of ~5500 ppm. Further details of this work will be presented by F.Z. Lian and W.E. Wallace at the forthcoming Annual Meeting of Magnetism and Magnetic Materials in Minneapolis. Additional work concerning the fabrication of magnets with a much lower concentration of oxygen and its impact on the magnetic properties is in progress.

2:17-TYPE MAGNETS

A number of researchers have been focussing their attention on the Nd-Fe-B magnets. However, Sm_2Co_{17}-type magnets fulfill a variety of demanding applications while the Nd-Fe-B magnets fail to perform in such applications. For example, Sm_2Co_{17}-based magnets exhibit good stability at elevated temperatures (up to ~150 °C). They are also reasonably stable under neutron and proton irradiation.

Ray and Liu [2] reported the fabrication of magnets with an energy product close to 30 MGOe and an intrinsic coercivity of ~15.5 kOe. Very early in the history of development of 2:17 type magnets, the need for the addition of small quantities of copper and zirconium to develop the required microstructure and provide decent coercivities was recognized. Recently, several research groups, including ours [3], have examined the possibilities of enhancing the remanence (and still retain acceptable coercivities) by partially replacing samarium and cobalt with a light rare earth element such as praseodymium and a transition metal such as iron, respectively. However, larger quantities of Pr and Fe, beyond a certain limit, seriously affect obtaining magnets with reasonable coercivities due to the change in magnetocrystalline anisotropy changing from the easy axis direction for Sm_2Co_{17} to the easy plane in Pr_2Fe_{17}. A significant amount of work has been done in optimizing the processing conditions such as sintering temperature, time and step annealing parameters in order to develop magnets with maximum energy product and reasonably good rectangular second quadrant hysteresis loops. As an illustration, the second quadrant hysteresis curve for a laboratory magnet fabricated from $Sm(Co_{bal}Fe_{0.316}Cu_{0.052}Zr_{0.02})_{7.72}$ as a starting composition is shown in Figure 2. This magnet exhibits a room temperature remanence of 12.1 kG, an intrinsic coercivity of 17 kOe and a maximum energy product of 31.7 MGOe. The variation of technical magnetic properties as a function of temperature for a magnet of nominal composition

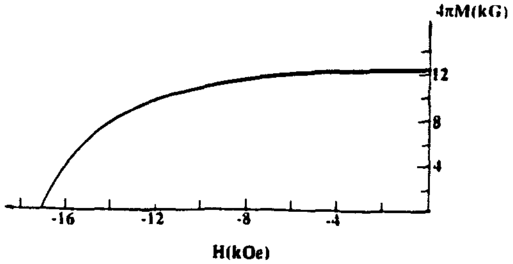

Fig. 2 Demagnetization curve of a 2:17 magnet.

$Sm(Co_{bal}Fe_{0.319}Cu_{0.052}Zr_{0.02})_{7.74}$ is given in Table 1. It may be noted that these alloys have Curie temperatures of >750 °C and hence they exhibit reasonably good thermal stabilities in technical magnetic properties up to ~150 °C.

Work carried out by Cost et al. [4], and by Krause [5] on several magnets such as 1:5, 2:14:B and 2:17-type have clearly shown that the 2:17-type magnets exhibit good stability upon exposure to large doses of neutron radiation. In this context, it may be pointed out that electromagnetic devices for aircraft, travelling wave tubes, accelerators, satellites etc. which operate at elevated temperatures and/or which are likely to be exposed to hazardous conditions of radiation will rely on the development of 2:17-type magnets. Therefore, even incremental improvements in the magnetic properties of these magnets are highly desirable.

Table 1. Magnetic properties of $Sm(Co_{bal}Fe_{0.319}Cu_{0.052}Zr_{0.02})_{7.74}$ from room temperature to 150 °C.

T (°C)	B_r (kG)	$_iH_c$ (kOe)	$(BH)_{max}$ (MGOe)
24	12.10	12.0	31.8
50	11.98	11.5	31.3
75	11.94	9.8	30.8
100	11.89	9.3	30.3
125	11.29	8.7	28.6
150	11.23	7.8	27.0

PERMANENT MAGNET DEVICES

High energy magnets have been incorporated in the construction of several devices. Examples of such devices include actuators, motors, generators, magnetic separators, accelerators, smart weapons, nuclear magnetic resonance imaging machinery and even toys. In attempting to incorporate the high energy magnets into devices, it is absolutely essential to redesign a device to obtain the best performance. Fortunately, several codes are commercially available to perform such finite element analysis. Howe and his co-workers [6] have made significant contributions in this regard. Recently, Simizu et al. [7] examined the design of a slotless brushless motor. They argued that for a certain set of parameters, a slotless motor becomes very attractive. In the example they considered, such a payoff occurs with magnets of B_r > 11 kG.

During the past three years, attention is being directed towards the development of electric vehicles. Permanent magnet-based motors and alternators are expected to play a significant role in this market due to many advantages they offer, viz., light-weight, energy-efficiency, better controllability etc.

INVESTIGATION OF MATERIALS AND PROCESSES

Recently a few intermetallics of the composition $R_2Fe_{14-x}Co_xSi_2$ were investigated employing powder x-ray diffraction pattern and bulk magnetic measurements. They may be regarded essentially as R_2Fe_{17}-based off-stoichiometric materials. X-ray patterns of these compositions were indexed on the basis of hexagonal Th_2Ni_{17}-type structure when R= Y, Dy, Ho, Tb and Er and rhombohedral Th_2Zn_{17}-type when R= Nd and Gd. The most noteworthy aspect that emerged from these studies is that a majority of the compositions investigated exhibited uniaxial anisotropy at room temperature with a reasonably large anisotropy field for some of these materials [8]. The Curie temperature increases with the addition of cobalt and is ~750 K for $Tb_2Fe_{10}Co_4Si_2$.

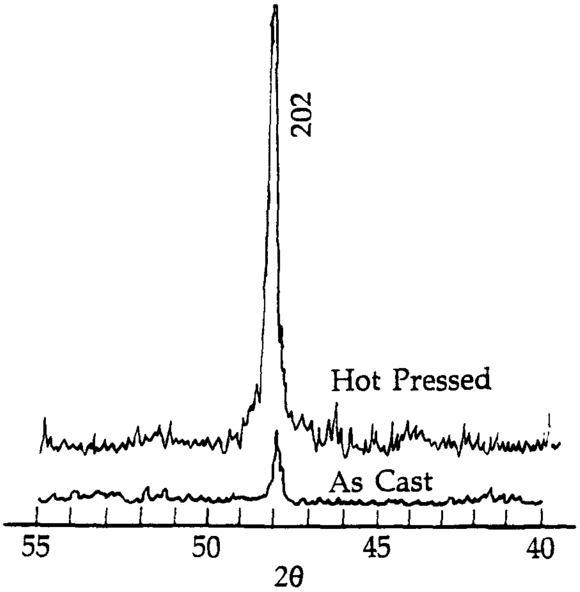

Fig. 3 X-ray diffraction pattern of as-cast and grain-aligned Dy_3Al_2 sample.

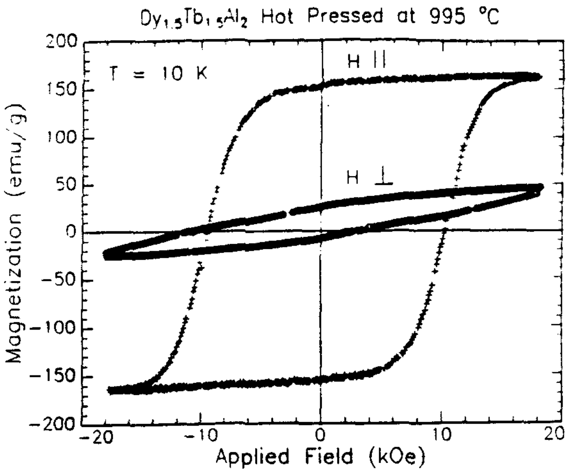

Fig. 4 Hysteresis loop of $Dy_{1.5}Tb_{1.5}Al_2$ sample at 10 K.

Several processing techniques have been employed to fabricate permanent magnets. These include powder metallurgical techniques, melt spinning followed by consolidation, mechanical alloying and hot pressing. The last approach is simple and effective. We have recently investigated the possibility of effecting good grain alignment by hot pressing Dy_3Al_2 and related materials. Single crystals of Dy_3Al_2, a tetragonal material with a Curie temperature of ~40 K, have been investigated by Barbara et al. [9], who showed that the theoretical maximum energy product of this material is nearly 70 MGOe at 4.2K. We have conducted a study of hot pressed Dy_3Al_2 and $Dy_{3-x}R_xAl_2$ (where R = Gd, Tb etc) at about 1000 °C [10]. The samples obtained exhibited considerable grain alignment (see, for example, Figure 3) and energy products of ~35 MGOe at 10K (see, for example, Figure 4). Further work is in progress.

Finally, it may be pointed out that there are two recent discoveries which are attracting world-wide attention. The first one refers to the nitrogenated magnets [11] and the second refers to the nano-composites made of a hard and a soft magnetic phase [12]. Results on these two aspects are discussed by other speakers in this symposium.

ACKNOWLEDGMENT

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