Grain-Boundary Diffusion of Pr-Fe-B Magnets for Cryogenic Applications

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Abstract: Conventional Pr-Fe-B magnets for low temperature applications frequently suffer the risk of partial demagnetization when handled at room temperature (RT). To increase the coercivity of Pr-Fe-B magnets at RT without sacrificing the remanent magnetic polarization, a grain-boundary-diffusion (GBD) process using Tb can be applied which allows the manufacturing of Pr-Fe-B magnets with $H_{cJ} = 1800$ kA/m and $B_r = 1.41$ T at 293 K and $H_{cJ} = 6300$ kA/m and $B_r = 1.62$ T at 77 K. The temperature dependence of the coercivity increase induced by the GBD correlates well with the different anisotropy fields of Pr$_2$Fe$_{14}$B and Tb$_2$Fe$_{14}$B supporting the model of higher nucleation fields due to Tb-rich high anisotropy $(Pr,Tb)_2Fe_{14}B$ layers along the grain-boundaries. The coercitivity gradients induced by the GBD process have been determined and an increase in $H_{cJ}$ has been observed up to a depth of ~4 mm.

Keywords: Pr-Fe-B, grain-boundary diffusion; spin-reorientation transition (SRT); coercivity; anisotropy

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1. Introduction

It is well known that the usage of Nd-Fe-B magnets at cryogenic temperatures is limited to temperatures $T > \sim 140$ K due to the spin-reorientation transition (SRT) [1,2]. It was recently demonstrated that this transition temperature can be decreased by replacing Nd with Pr which allows the fabrication of magnets with a maximum energy density of $(BH)_{max} = 520$ kJ/m³ at $T = 85$ K [3]. These Pr-Fe-B based magnet qualities have excellent properties for low temperature applications like cryogenic permanent magnet undulators [4–6]. However, due to the limited coercivity at room temperature (RT) and the corresponding risk of partial demagnetization when no cooling is applied, only a few prototypes using this class of material have been built. This issue can be solved by applying a grain-boundary-diffusion (GBD) process, a technology first established by Park et al. [7]. We will provide actual data on the performance of commercially available Pr-Fe-B based magnet grades after implementation of GBD. The temperature dependence of the magnetic properties as well as the thickness limitations associated with the application of the GBD process will be discussed.

2. Magnetic Properties

Typical demagnetization curves of Pr-Fe-B magnets fabricated by conventional pressing and sintering technology (VACODYM® 131 TP) as well as by state-of-the-art GBD technology using Tb as the diffusing heavy rare earth (HRE) element (VACODYM 131 DTP) are shown in Fig. 1. A minimum increase in coercivity by GBD of $\Delta H_{cJ} = 400$ kA/m without a significant loss in remanent magnetic polarization can be observed.

<table>
<thead>
<tr>
<th>Characteristic</th>
<th>VD 131 TP</th>
<th>VD 131 DTP</th>
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</thead>
<tbody>
<tr>
<td>Remanence $B_r$ at 293 K (T)</td>
<td>$\geq 1.38$</td>
<td>$\geq 1.38$</td>
</tr>
<tr>
<td>Coercivity $H_{cJ}$ at 293 K (kA/m)</td>
<td>$\geq 1230$</td>
<td>$\geq 1640$</td>
</tr>
<tr>
<td>Remanence $B_r$ at 77 K (T)</td>
<td>$\geq 1.58$</td>
<td>$\geq 1.58$</td>
</tr>
<tr>
<td>Coercivity $H_{cJ}$ at 77 K (kA/m)</td>
<td>$&gt; 3185$</td>
<td>$&gt; 3185$</td>
</tr>
</tbody>
</table>

Fig. 1. Typical demagnetization curves of VACODYM 131 TP and VACODYM 131 DTP magnets at RT. The field was applied parallel to the direction of orientation.
A summary of the minimum remanence and coercivity values of these new material grades are given in Table I. Please note that the magnetic specification of the GBD processed magnet quality is restricted regarding the dimensions of the magnet.

While the reversible permeability $\mu_{\text{rev}}$ along the direction of orientation is around 1.03 at RT and corresponds to typical values of Nd-Fe-B based magnet grades, the reversible permeability $\mu_{\text{rev}}^\perp$ perpendicular to the direction of orientation is $1.2 \pm 0.05$ (Fig. 2). To minimize the occurrence of domain wall motion the sample was saturated along the direction of orientation followed by a reverse field pulse with $H \sim H_c$ prior to the measurement of $\mu_{\text{rev}}^\perp$. By extrapolating the $J$-$H$ loops to higher fields the anisotropy field was estimated to $H_a \sim 6000$ kA/m which corresponds well with literature data \cite{2}. As most of the magnet volume is not affected by the diffusion process, no difference in $\mu_{\text{rev}}^\perp$ can be observed for conventional and GBD processed Pr-Fe-B magnet grades.

**3. Temperature Dependence**

The temperature dependence of the remanent magnetic polarization shows no indication of SRT down to temperatures of $\sim 4$ K and no significant differences between the two magnet grades can be detected [Fig. 3(a)].

Additionally, $B_r$ shows only a small increase for temperatures below $T = 77$ K and consequently cooling with liquid helium does not lead to a superior magnetic performance.

Both magnet qualities show a pronounced increase in coercivity upon cooling to cryogenic temperatures and values of up to $\sim 7000$ kA/m can be achieved [Fig. 3(b)]. The difference in coercivity $\Delta H_{cJ}$ between the conventional Pr-Fe-B magnet and the GBD processed quality is around $470$ kA/m $- 540$ kA/m for temperatures below RT.

**Fig. 2.** J-H loop of VACODYM 131 DTP at RT with the field applied perpendicular to the direction of orientation.

**Fig. 3.** Temperature dependence of (a) $B_r$ and (b) $H_{cJ}$ of Pr-Fe-B magnets after conventional fabrication without GBD and after fabrication involving a GBD process.

No scaling of the absolute value of $\Delta H_{cJ}$ can be observed in this temperature regime. The relative increase of $\Delta H_{cJ}$ with respect to the conventional magnet quality is, however, systematically decreasing towards lower temperatures (Fig. 4).

This trend correlates with the relative increase of $\Delta H_a = (H_{a,Tb_2Fe_{14}B} - H_{a,Pr_2Fe_{14}B}) / H_{a,Pr_2Fe_{14}B}$ indicating that the anisotropy differences of $\text{Pr}_2\text{Fe}_{14}B$ and $\text{Tb}_2\text{Fe}_{14}B$ have the major impact on $\Delta H_{cJ}$. Thus, for sintered magnets with magnetically decoupled 2-14-1 grains, the origin for the coercivity increase is most likely related to the formation of Tb-enriched high anisotropy $(\text{Pr,Tb})_2\text{Fe}_{14}B$ layers along the grain boundaries. No indication that the GBD process is
leading to a better decoupling of the grains, as for example reported for fine grained HDDR magnets [8], was observed.

4. Coercivity Gradient

The GBD process does not lead to homogenous local coercivities when it is applied to magnets with thicknesses \( t \) exceeding 5 mm. Figure 5 shows the distribution of the typical local coercivities at RT and 100°C of magnets with thicknesses >> 5 mm which have been processed with the GBD technology. It can be clearly seen that in the proximity of the Tb – coated surface, the local coercivity of the material can easily exceed 1800 kA/m at RT. With increasing distance \( d \) to the surface, the local coercivity drops and for \( d > 4 \) mm no increase in coercivity can be observed which is typically limiting the application of the GBD process to magnets with \( t < 5 \) mm. The application of the GBD process on thicker magnets is only beneficial if the strongest demagnetization fields occur close to the surface of the magnet.

5. Summary

With the development of two new magnet qualities based on Pr-Fe-B, magnetic material for the construction of cryogenic permanent magnet assemblies is commercially available. The implementation of the GBD process leads to an increased coercivity and sufficient resistivity against thermal demagnetization even at slightly elevated temperatures. The analysis of the temperature dependence of \( H_{cJ} \) reveals a good correlation with the anisotropy fields.

For thick magnets, the penetration depth of the diffusing element is limited to ~4 mm leading to a systematic drop of the local coercivity with increasing distance \( d \) to the coated surface.

![Fig. 5. Coercivity distribution of Pr-Fe-B magnets after GBD at RT and 100°C in dependence of the distance to the Tb - coated surface. The homogenous coercivity of Pr-Fe-B without GBD is indicated for comparison.](image)

Acknowledgement

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References