ENHANCED POWDER-PROCESSED ALNICO MAGNETS BY THERMAL GRADIENT CONTROL

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INTRODUCTION

Permanent magnets are critical to several clean propulsion and green energy technologies, such as traction-drive motors for electric vehicles and generators for wind turbines. Nd-Fe-B-based permanent magnets have the highest maximum-energy product (~55 MGOe) among all types of magnets, and so are currently used in the majority of these applications.1–4 However, Nd has been identified as a critical material (Figure 1) with limited supply as a rare-earth (RE) element, and original equipment manufacturers (OEMs) are reluctant to rely on RE’s because of market and supply uncertainty.5 Also, the energy product of Nd-Fe-B falls rapidly with increasing temperature, and especially for operation above 120 °C, Nd-Fe-B requires added Dy, which is also a critical material with a greatly increased cost.6

Replacing rare-earth magnets with alternatives, such as alnico, will eliminate supply instability, increase sustainability, and decrease costs of permanent magnets for applications such as traction-drive motors. Conventional processing of alnico magnets with the best second quadrant squareness and energy product involves directionally solidified casting and final machining, which leads to higher costs and material waste. Due to their randomly oriented fine-grained microstructure, powder-processed, sintered alnico magnets have had inferior energy product and therefore have been used only for low energy-density applications. This work describes epitaxially seeded solid-state grain growth driven by thermal gradient control, leading to enhanced alnico magnets produced by powder processing. A modified alnico 8 composition was high-pressure gas atomized, compression molded, sintered to full density, and heat treated to achieve improved magnetic properties. These alnico magnets were thoroughly characterized using scanning electron microscopy (SEM), orientation image mapping (OIM), electron backscattered diffraction (EBSD), and hysteresisgraph measurements. Grain growth and some texturing control have been demonstrated through solid-state thermal processing of a fully dense, powder-processed sample of the modified alnico 8 composition. Directionally grown alnico 9 shows promise for being able to control grain orientation and epitaxially seed the [001] direction for grain growth.

Figure 1. Critical materials according to importance and supply as identified by the USDOE.

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All of these factors led to the renewed search for alternative non-RE permanent magnet materials with sufficient energy density for compact, high-performance electric motors and generators. Figure 2 shows the development of different magnet materials relative to their room temperature magnetic energy product over time. The highest energy products are from RE-containing permanent magnets, followed by alnico permanent magnets. Alnico was first developed in the 1930s and optimized during the 1960s, and has potential for further improvement due to the advent of modern processing methods, and many high-resolution characterization tools that can be directed to “tune” the self-organized nano-structure of these complex magnetic alloys. Additionally, alnico has excellent corrosion resistance and maintains its energy product at high temperatures, which is essential for electric traction-drive motors.

Conventional alnico magnet production involves directionally-solidified casting to achieve the best microstructural alignment and, after thermal-magnetic treatment, the best second quadrant squareness and energy product. Directional solidification is a complicated, costly processing method that leads to high-production costs and large quantities of excess scrap material from extensive machining of each casting. Sintered alnico maintains high coercivity, vital to motor applications, while lowering production costs through near-net shape processing and simplified manufacturing methods. If an aligned type of sintered alnico permanent magnet with an optimized composition for high coercivity and magnetic induction could be produced, it would be a competitive alternative to RE permanent magnets.

The techniques to achieve texture, or alignment, within the solid state, include hot deformation, and recrystallization or grain growth. Steinort et al. found that recrystallization and grain growth could be promoted if sufficient grain edge strains were induced through a thermal gradient and by grain boundary, γ-phase precipitation that involves a volume change. Luborsky and Aust followed up to find the γ-phase does not promote grain growth, but elements that stabilize the γ-phase may be necessary. Furthermore, they concluded it is very difficult to control the grain orientation of the recrystallized grains. This paper investigates thermal-gradient driven, solid-state recrystallization and grain growth of a modified alnico 8 composition, using a conjoined grain-oriented (alnico 9) material to seed the grain growth and control the grain orientation of the recrystallized grains.

### EXPERIMENTAL METHODS

Fine, spherical alloy powders were produced using high-pressure gas atomization at Ames Laboratory of a composition within the alnico 8 family. The gas-atomized powder was size classified and then compression molded at 160 MPa using a QPAC®-40 poly(propylene carbonate) binder (from Empower Materials Inc., New Castle, Delaware) into a 9.5 mm diameter, ~1 cm tall cylinder. The sample was debound at 300 °C in air for 2 hours and vacuum sintered for an initial 4 hours to obtain >99% relative density. Kassen et al. and Anderson et al. detail the gas-atomization process, resulting powder characteristics, and sintering procedures. Each sintered sample was then placed in a tube furnace, set to 1,260 °C, in contact with a water-cooled cold finger to produce a >20 °C/cm gradient through the height of the cylinder. To minimize oxidation, the furnace was purged with high-purity nitrogen gas and two type-K thermocouples monitored the temperature across the height of the sample. A disk of polished and oriented alnico 9, provided by Arnold Magnetic Technologies, was placed in contact with the hotter end of the sample in the furnace, to epitaxially seed the preferred grain-growth direction. EBSD of the alnico 9 disk, verifying the texture, is included in Figure 3. SEM, using an FEI Quanta FEG 250 SEM, and EBSD, using a JEOL JAMP-7830F field emission auger electron spectroscopy (FE-AES) instrument, were performed on polished, transverse and longitudinal surfaces of the samples.

The 9.5 mm diameter cylinder was cut using an electrical discharge machine (EDM) into three, 3-mm diameter rods for magnetic property measurements. The rods were heated to 1,250 °C for 30 minutes in an Oxy-Gon quench furnace (from OXY-GON Industries, Inc., Epsom, New Hampshire) with a vacuum of 0.1–0.01 mPa, followed by an oil quench to solutionize the microstructure to eliminate γ-phase precipitation, and to minimize spinodal decomposition of the high-temperature
solid-solution phase. After being wrapped in tantalum foil and sealed in a quartz vial, the rods were magnetically annealed under a ~1T field at 840 °C for 10 minutes. Finally, anneals were performed at 650 °C for 5 h, and 580 °C for 15 h to achieve a full heat treatment. The magnetic properties were measured on a closed-loop Laboratorio Elettrofisico AMH-500 hysteresigraph, under a maximum applied field of 1.2 MA/m (15 kOe).

RESULTS & DISCUSSION

SEM of the polished, transverse cross-sectional end surfaces of the cylindrical sample showed significant differences between the hot/Alnico 9 seeded side and the end in contact with the cold finger. Figure 4(a) shows a montage SEM micrograph of the Alnico 9 (seeded) end, while Figure 4(b) shows the cold-finger end. The primary goal of this work was to achieve grain growth within the sample. It is apparent this was achieved, at least at the hot end (Figure 4(a)) that was seeded with Alnico 9, as evidenced by the ~2 mm size grains in the sample. The center of the hot end retained a fine-grain size, indicating further time might be necessary at temperature in order to achieve complete recrystallization. The cold-finger end (Figure 4(b)) showed a mostly fine-grained structure with two larger grains (see arrows).

Orientations of the recrystallized grains in Figure 4 were analyzed using EBSD and are shown in Figures 5 and 6 with overlays on the montage SEM micrographs. The fine-grain sections are clearly randomly oriented...
Figure 5. EBSD analysis of grain texture overlayed on the SEM montage micrograph with corresponding inverse pole figures showing orientation. The transverse (TD) and rolling (RD) directions are as noted.

Figure 6. EBSD analysis of grain texture overlayed on the SEM montage micrograph with corresponding inverse pole figure showing orientation of the lower left rectangular area.
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(Figure 6), while a large portion of the recrystallized region of the alnico 9 (seeded) end has tended towards the preferred [001] orientation, as shown by the section labeled D in Figure 5.

The sectioned samples for magnetic property measurements were chosen from the areas shown in Figure 7 (corresponding to the sample shown in Figure 5), with three, longitudinal cross-sections from the labeled locations. Figure 8 shows an SEM micrograph of the polished “Long. 1” (longitudinal) cross-section of the cylindrical sample where the grain size transition occurs over halfway through the height of the sample. Figure 9 includes EBSD overlays on the longitudinal cross-section, with the orientations of the grains correlating well with the transverse cross-section.

The other longitudinal cross-sections showed similar results and good correspondence with the transverse cross-sections. It is evident from these results that over 50% of the sample height and over 90% of the sample area within that height exhibited abnormal grain growth. Processing defects, such as the crack extending across the middle of the sample from the left side in Figure 8, seemed to be a barrier to continued grain growth, possibly due to both oxygen contamination and grain pinning effects.

A fine-grain microstructure was evident around the circumference of the entire sample. Further examination of the edge of the sample, using SEM/EDS mapping, included in Figure 10, showed significant oxidation of both Ti and Al at the edge of the sample. Oxygen contamination promotes γ-phase formation at the grain boundaries and, apparently, stabilization of the fine grained microstructure.

Magnetic property measurements of the three cylinders machined from the bulk sample are included in Table I. Comparison with Arnold Magnetic Technologies (AMT) sintered alnico 8H and cast alnico 9, showed these samples to lie between the two sets of properties, as expected for these initial results of a sintered microstructure with some grain alignment. The values for remanence (B_r), coercivity (H_c), intrinsic coercivity (H_{ci}), and maximum energy product (BH_{max}) of the pink, green, and teal cylinders bridge the gap between the AMT sintered alnico 8H and cast alnico 9. There is potential for improvement in B_r, H_c, H_{ci}, and BH_{max} of the sintered samples by further grain growth, texture, and compositional adjustments. Future work includes improving the furnace atmosphere, and optimizing the temperature and time to achieve full grain growth and preferred texture within the sintered alnico magnet.

CONCLUSIONS

Grain growth and some texturing control have been demonstrated through solid-state thermal processing of a fully dense powder processed sample of the modified alnico 8 composition. Directionally grown alnico 9 shows promise for being able to control grain orientation and epitaxially seed the [001] direction for grain growth. The γ-phase appears to stabilize the grain size and random orientation, and must be minimized along with oxidation of the sample.
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Figure 9. EBSD analysis of grain texture overlayed on the SEM montage micrograph of longitudinal section 1 (“Long. 1”) with corresponding inverse pole figures showing orientations of large grains and randomly oriented fine grains.

Figure 10. SEM/EDS map of select elements along the edge of longitudinal section 1 (“Long. 1”).
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