Recycled Sm-Co bonded magnet filaments for 3D printing of magnets

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**Recommended Citation**

Khazdozian, Helena A.; Manzano, J. Sebastian; Gandha, Kinjal; Slowing, Igor I.; and Nlebedim, Ikenna C., "Recycled Sm-Co bonded magnet filaments for 3D printing of magnets" (2018). *Ames Laboratory Accepted Manuscripts*. 175.  
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Abstract
Recycling of rare earth elements, such as Sm and Nd, is one technique towards mitigating long-term supply and cost concerns for materials and devices that depend on these elements. In this work recycled Sm-Co powder recovered from industrial grinding swarfs, or waste material from magnet processing, was investigated for use in preparation of filament for 3D printing of bonded magnets. Recycled Sm-Co powder recovered from swarfs was blended into polylactic acid (PLA). Up to 20 vol.% of the recycled Sm-Co in PLA was extruded at 160°C to produce a filament. It was demonstrated that no degradation of magnetic properties occurred due to the preparation or extrusion of the bonded magnet material. Good uniformity of the magnetic properties is exhibited throughout the filament, with the material first extruded being the exception. The material does exhibit some magnetic anisotropy, allowing for the possibility of the development of anisotropic filaments. This work provides a path forward for producing recycled magnetic filament for 3D printing of permanent magnets.

Keywords
Microscopy, Condensed matter properties, Magnetic hysteresis, Polymers, Sintering, Magnetic anisotropy, Magnetic materials, Chemical compounds, 3D printing

Disciplines
Condensed Matter Physics | Inorganic Chemistry | Materials Chemistry

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Recycled Sm-Co bonded magnet filaments for 3D printing of magnets

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(Presented 8 November 2017; received 2 October 2017; accepted 21 November 2017; published online 11 January 2018)

Recycling of rare earth elements, such as Sm and Nd, is one technique towards mitigating long-term supply and cost concerns for materials and devices that depend on these elements. In this work recycled Sm-Co powder recovered from industrial grinding swarfs, or waste material from magnet processing, was investigated for use in preparation of filament for 3D printing of bonded magnets. Recycled Sm-Co powder recovered from swarfs was blended into polylactic acid (PLA). Up to 20 vol.% of the recycled Sm-Co in PLA was extruded at 160°C to produce a filament. It was demonstrated that no degradation of magnetic properties occurred due to the preparation or extrusion of the bonded magnet material. Good uniformity of the magnetic properties is exhibited throughout the filament, with the material first extruded being the exception. The material does exhibit some magnetic anisotropy, allowing for the possibility of the development of anisotropic filaments. This work provides a path forward for producing recycled magnetic filament for 3D printing of permanent magnets. © 2018 Author(s). All article content, except where otherwise noted, is licensed under a Creative Commons Attribution (CC BY) license (http://creativecommons.org/licenses/by/4.0/). https://doi.org/10.1063/1.5007669

I. INTRODUCTION

Rare earth elements (REEs) like Sm and Nd are used in high energy product permanent magnets necessary for many applications like hard-disk drives, electric vehicles, and military applications. Long-term supply and cost of critical REEs like Nd are potential concerns for the magnet industry. Sm supply is not currently considered critical; however, supply disruption can occur with the deployment of technologies with significant dependence on Sm such as Sm-Fe-N. Research efforts on addressing critical materials seek to provide solutions through materials conservation, recycling, and investigating alternative materials. Reuse of recycled magnets as powders in bonded magnets is one path towards materials conservation and recycling of critical REEs. By recycling magnet powder recovered from industrial grinding swarfs, the materials can be reclaimed via incorporation into bonded magnet fabrication. Swarfs are waste magnetic materials generated from post-manufacturing processing of sintered magnets such as grinding, slicing, and polishing operations.

Bonded magnets, in which magnet powder is blended in polymer binder, can be produced as near-net-shape magnets with minimal or no post-manufacturing steps.\(^1\^-3\) This conserves materials by minimizing wastes and reduces manufacturing energy. Bonded magnets are typically manufactured by injection molding, calendering, extrusion, and compression molding.\(^1,3\) These techniques allow for manufacturing of complex geometries while achieving up to 70 (injection molding and calendering), 75 (extrusion), and 80 (compression) vol.% loading of magnet powder in binder.\(^1\)

Additive manufacturing is a fast-emerging technique for bonded magnets.\(^1\^-6\) Additive manufacturing techniques like 3D printing have the potential advantage to reduce manufacturing costs by

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eliminating the need for mold fabrication. It also enables rapid prototyping of parts and reduces the time-to-market of new products. 3D printed bonded magnets with 54 to 65 vol.% loading of magnet powder in binder have been reported.\textsuperscript{5,7} Additively manufactured magnets with 65 vol.% loading of magnet powder in binder are able to outperform injection molded magnets, even at this early stage of the technique.\textsuperscript{4,7}

In this paper, we describe the preparation of permanent magnet filaments intended for 3D printing using bonded Sm-Co powder reclaimed from swarfs in polylactic acid (PLA). PLA, a thermoplastic with a melting temperature of \(\sim 178^\circ\text{C}\), was selected as the binder for this investigation since its melting temperature is well below the Curie temperature of Sm-Co and due to its standard use in 3D printing. Up to 20 vol.% loading of recycled Sm-Co powder in PLA was extruded as a filament and no degradation in magnetic properties was observed.

II. METHODOLOGY

A. Material preparation

Recycled Sm-Co powder, recovered from industrial swarfs, was loaded in polylactic acid (PLA). The PLA was first dissolved in a solvent (methylene chloride) to obtain a viscous solution, aided by a mechanical homogenization at 14,000 rpm. After complete dissolution of PLA, the recycled Sm-Co powder was then dispersed in the viscous solution by homogenizing for \(\sim 1\) min at 8,000 rpm. The solvent was evaporated at \(\sim 40^\circ\text{C}\) while stirring with the homogenizer set to between 3,500 and 8,000 rpm to ensure the Sm-Co particles remained dispersed in the binder. The recycled Sm-Co powder was loaded in PLA at 5, 10, and 20 vol.%.  

B. Extrusion

To extrude the permanent magnet filaments, the bonded Sm-Co/PLA material was cut into irregular shaped pieces \(<2\) mm in diameter and loaded into a filament extruder chamber. The chamber was inserted in the screw assembly, which was wrapped with heating tape set to \(160^\circ\text{C}\). A thermocouple was used to track the temperature of the heating tape and extruder. An allen wrench was used to push the heated material through a 0.4 mm diameter nozzle to extrude the filament.

C. Characterization

Scanning electron microscopy (SEM) was used to determine the initial particle size of the powder. \(\mu\)-X-ray fluorescence (XRF) was used to characterize the elemental composition. X-ray diffraction (XRD) was used to determine the phases present in the recycled Sm-Co powder. Differential scanning calorimetry was used to characterize the thermal properties of the PLA and bonded magnet material before and after extrusion.

Magnetic hysteresis plots were measured with a vibrating sample magnetometer (VSM) at 300 K with the applied magnetic field from -3 T to 3 T. Each prepared material was measured before and after extrusion. Magnetic field was applied along the lengths of all samples during magnetization measurements.

Uniformity of magnetic properties in the extruded filaments was determined by segmenting an extruded filament (with the 20 vol.% loading of Sm-Co in PLA) into seven parts of 3 mm length each, and measuring the magnetic hysteresis of each of the segments. For comparison, all seven segments were combined into one sample to measure the magnetic hysteresis loop.

III. RESULTS AND DISCUSSION

The SEM micrograph of the recycled Sm-Co powder (Fig. 1a) shows a range of particle size distribution. The larger particles are mostly tabular/irregular shapes of 15 – 25 \(\mu\text{m}\) in the longest dimensions, while the smaller particles range between 5 – 10 \(\mu\text{m}\) (Fig. 1a inset). The elemental composition determined by \(\mu\)-XRF was 18.4 at.% Sm, 81.4 at.% Co, and 0.2 at.% Nd. Using this composition, a theoretical density of 8.34 g/cc was determined. The XRD pattern for the recycled Sm-Co powder was compared with the powder diffraction files (pdf) for SmCo\(_5\) (pdf #659058)
FIG. 1. a) SEM micrograph of Sm-Co powder. Inset: histogram of Sm-Co particles showing their particle size distribution along the length obtained using statistical analysis of ~400 particles; b) XRD of Sm-Co powder compared to reference cases of Sm-Co 1:5 and 2:17.

and Sm$_2$Co$_{17}$ (pdf #658751) in Fig. 1b. The peaks indicate that both phases are present in the powder (Fig. 1b). It is possible that oxide phases are also present, but XRF does not have sufficient sensitivity to detect lighter elements like oxygen. XRD would also not detect small phase fractions of oxides that may be present.

The magnetic properties of the recycled Sm-Co powder (Fig. 2a) were compared to that of the un-extruded sample (Fig. 2b) and the extruded filament (Fig. 2c). The theoretical densities of the powder (8.34 g/cc) and PLA (1.25 g/cc) were used to determine the density of different loading fractions: 1.65 g/cc for 5 vol.%, 1.96 g/cc for 10 vol.%, and 2.67 g/cc for 20 vol.%. It can be seen that the hysteresis loops of the un-extruded and extruded samples have better squareness than the recycled Sm-Co powder for all loading fractions investigated. Comparable coercivity was obtained at all loading fractions. At 5, 10 and 20 vol.% loading fractions, the remanence, as estimated from linear scaling of the saturation magnetization, would be 0.23, 0.46 and 0.93 kG respectively. As seen in the insets in Fig. 2, the remanence of the recycled Sm-Co powder loaded in PLA is within 10% of these estimated values, with the exception of the 10 vol.% extrusion. One would expect the extruded

FIG. 2. Magnetic hysteresis of a) recycled Sm-Co powder, b) un-extruded Sm-Co powder loaded in PLA at 5, 10, and 20 vol.%, and c) extruded Sm-Co powder loaded in PLA at 5, 10, and 20 vol.%; SEM micrograph of d) homogenized recycled Sm-Co powder obtained after dissolving PLA and e) recycled Sm-Co powder. Insets: histogram of Sm-Co particles showing their particle size distribution along the length obtained using statistical analysis of ~400 particles.
samples to have a higher or comparable remanence with the un-extruded samples; extrusion processes are known to induce anisotropy due alignment of particles in the extrusion direction.\textsuperscript{8,9} This trend is observed for the 5 and 20 vol.% extrusions. It is nevertheless unclear why the remanence of the 10 vol.% extruded sample is reduced compared to the un-extruded sample. In general, the results show that heating and extrusion did not result in significant degradation of the magnetic properties.

The observed increase in coercivity and squareness of the hysteresis loops is likely related to particle rotation during measurement and dipolar magnetic interaction due to particle size refinement. Stronger interaction can result in distorted hysteresis loops (or wasp-waisted hysteresis loop),\textsuperscript{10} but particle rotation is most likely the dominator factor. By loading the recycled Sm-Co powder in a non-magnetic matrix (PLA), both particle-to-particle interaction and particle rotation are reduced. The homogenization process employed in dispersing the magnetic particles in the binder (as explained in the methodology section) appears to have resulted in particle size refinement, which is likely dependent on the high speed of the mechanical homogenizer. To verify the refinement, the PLA content of an un-extruded sample was dissolved away with methylene chloride and the powder derived used for SEM. Some PLA was still left on the surface of the particles, but was sufficiently removed to reveal the form of the particles. Although agglomerated, the obtained micrograph (Fig. 2d) indicated that some size refinement took place. The particles are mostly 10 \(\mu\text{m}\) in their longest dimensions with nearly equal distribution of 5 and 15 \(\mu\text{m}\) particles (Fig. 2d inset). The refinement resulted in a more uniform particle size distribution relative to the original recycled Sm-Co powder in Fig. 2e. We propose a future study on the effect of homogenizer speed on the particle size refinement and the resultant magnetic properties in filaments for 3D printing. Improved coercivity (8.91 kOe) of epoxy-cured recycled Sm-Co powders verifies the effect of particle rotation (Fig. 3).

The recycled Sm-Co powder does exhibit magnetic anisotropy. The magnetic hysteresis of the un-extruded Sm-Co powder was measured before and after applying a magnetic field of 3 T at 400 K (Fig. 3). Some particle rotation likely occurred, improving the remanence and coercivity of the recycled Sm-Co powder. This demonstrates the suitability of powders derived from sintered magnets for anisotropic magnet development, as has been suggested by others.\textsuperscript{11}

Fig. 4 shows the verification of the uniformity in the magnetic properties of the extruded filament. Both the coercivity and remanence had a standard deviation less than 2\% (Fig. 4b), indicating good uniformity of magnetic properties; this is also apparent from the magnetic hysteresis loops in Fig. 4a. The only outlier is the first piece of extruded filament. It is possible that the particle size distribution in the first part of the extruded filament is not as uniform as the subsequent part of the filament. As a result, a small level of the dipolar interaction effect observed in Fig. 2a is retained in the first part of the extruded filaments.

![Magnetic hysteresis of recycled Sm-Co powder in epoxy measured at 300 K before and after alignment at 400 K under applied magnetic field of 3 T.](image-url)
FIG. 4. a) Magnetic hysteresis of the 20 vol.% recycled Sm-Co in PLA filament: 3 mm segments and all segments combined; b) coercivity and remanence of the PLA filament segments.

IV. CONCLUSIONS

Bonded magnet filaments were prepared by extrusion of recycled Sm-Co powder in PLA with up to 20 vol.% loading of magnet powder in binder. Improvement in magnetic properties was observed, likely due to minimized particle rotation when loading recycled Sm-Co powder in PLA, compared to the original powder. Additionally at low volume fractions of magnet powder, particle size refinement resulted in changes in particle-to-particle magnetic interaction. No significant degradation of magnetic properties occurred due to the extrusion process. Furthermore, good uniformity of the magnet filament was demonstrated. In future work magnetic property dependence on particle size refinement will be investigated, filaments with higher loading fractions will be studied, and plasticizers will be added to the filament to allow for 3D printing of recycled bonded magnets.

ACKNOWLEDGMENTS

This research was supported by the Critical Materials Institute, an Energy Innovation Hub funded by the U.S. Department of Energy, Office of Energy Efficiency and Renewable Energy, Advanced Manufacturing Office. J.S.M. and I.I.S. acknowledge support of the U.S. Department of Energy, Office of Basic Energy Sciences, Division of Chemical Sciences, Geosciences and Biosciences through the Ames Laboratory. The Ames Laboratory is operated for the U.S. Department of Energy by Iowa State University under contract No. DE-AC02-07CH11358.
