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ABSTRACT

Magnetic Field Assisted Additive Manufacturing (MFAAM), 3D printing in a magnetic field, has the potential to fabricate high magnetic strength anisotropic bonded magnets. Here, 10, 35, and 54 wt% strontium ferrite bonded magnets using polyamide 12 binder were developed by twin screw compounding process and then printed via MFAAM samples in zero, and in 0.5 Tesla (H parallel to the print direction and print bed). The hysteresis curves were measured using a MicroSense EZ9 Vibrating Sample Magnetometer (VSM) for 3 different mount orientations of the sample on the sample holder to explore the magnetic anisotropy. The samples printed in zero field exhibited a weak anisotropy with an easy axis perpendicular to the print direction. This anisotropy is caused by the effect of shear flow on the orientation of the magnetic platelets in the 3D printer head. For the MFAAM samples, the S values are largest along the print bed normal. This anisotropy is caused by the field. The alignment of the magnetic particles happens when the molten suspension is in the extruder. When the material is printed, it is folded over on the print bed and its easy axis rotates 90° parallel to the print bed normally. Little realignment of the particles happens after it is printed, suggesting a sharp drop in temperature once the composite touches the print bed, indicating that field-induced effects in the nozzle dominate the anisotropy of MFAAM deposited samples.

I. INTRODUCTION

Bonded magnets are magnetic composites developed by binding a significant packing fraction of magnetic powders or fillers with a suitable binder matrix. The primary advantages, such as cost-effectiveness, manufacturing flexibility, corrosion resistance, and mechanical performance, have made bonded magnets considered technological alternatives to sintered magnetic material in engineering applications such as sensors, electric motors, small generators, and other magnetic devices.[1,2] The fabrication technologies widely considered for manufacturing bonded magnets are compression molding and injection molding.[2] Injection molding is suitable for fabricating flexible thermoplastic bonded magnetic composites, which can exhibit a good combination of mechanical strength and ductility, thereby enabling the structural integrity of bonded magnets for loading applications. In addition, the injection molded bonded magnets exhibit good isotropic magnetic properties. Garrell *et al.*[3] fabricated isotropic bonded neodymium magnets using a polyphenylene sulfide (PPS) binder matrix and they were able to achieve a good combination of corrosion resistance, dimensional stability, and low moisture absorption properties. The major shortcomings of the injection molding process have been identified as the cost associated with complex mold and custom tooling requirements.[3,4] The innovative development in other polymer processing technologies such as extrusion-based additive manufacturing technology has revived the research interest in fabricating bonded magnets, which is crucial in superior magnetic performance and reduction in manufacturing cost.

Additive manufacturing (AM), also known as 3D printing, is a viable process for fabricating near-net-shaped and complex 2D and 3D bonded magnetic structures, which reduces materials wastage, eliminates the cost of tooling, and minimizes manufacturing lead times. There have been concerted research efforts in employing additive manufacturing technologies to fabricate bonded magnetic composite with superior magnetic performance that can supplant conventional sintered magnets.[5–9] Such superior magnetic performance can be achieved by increasing the packing fraction of the magnetic powders in the bonded magnets. Furthermore, improvements in magnetic performance such as magnetic remanence and energy density of bonded magnets have been attributed to the ability of anisotropic magnetic powders to be post-aligned in a preferred magnetization direction via different magnetic field intensities.[10] Therefore, an integrated approach to fabricating high-performance bonded permanent magnetic materials lies in combining the benefits of high packing fraction and the use of anisotropic magnetic powders.

In this research, we have exploited anisotropic strontium ferrite magnetic powders in fabricating 3D-printed nylon strontium ferrite magnetic composites using a magnetic field-assisted fused filament fabrication AM process. The primary aim of this research focuses on evaluating the magnetic anisotropy achievable exhibited by the field-assisted and 3D-printed anisotropic strontium ferrite bonded magnets.

II. EXPERIMENTAL PROCEDURES

The magnetic filler used in this research is a low-cost strontium ferrite powder supplied by DOWA electronics. This magnetic powder exhibits a platelet shape morphology, and the average particle size diameter was estimated to be 0.99 μm with an average diameter to thickness ratio of ~3. The binder matrix for the fabricated bonded magnetic composite was polyamide 12 also known as nylon 12. The magnetic powders were compounded with the polyamide 12 binder using the twin screw extrusion technology.

10, 35, and 54 wt.% packing fractions of strontium ferrite powders were compounded with polyamide 12 binder using a process 11 twin screw extruder (S1) to produce a bonded magnetic composite monofilament for 3D printing process. By optimizing the extrusion process parameters such as extrusion temperature, screw speed, feed rate, and torque percentage, bonded magnetic monofilaments with an average diameter of 1.75 ± 0.05 mm were extruded, which is viable for fused filament fabrication (FFF) 3D printing. The extrusion temperature along the extruder barrel used in the twin screw compounding of the magnetic composite is presented in Figure 1a. The extrusion temperature baseline was set according to the melting point of the polyamide 12 binder, which has an onset value of 175 °C. The extrusion temperature along was optimized within the onset melting point + 35° C to facilitate sufficient melting of the binder for the compounding process. Unlike injection molding, it is noteworthy that excessive melting of the binder should be avoided to facilitate sufficient drawing strength in the extrusion process. According to Figure 1a, the highest temperatures along the extruder barrel existed at zone 5, which is the zone where the magnetic powders were added (side feeding), followed by zone 6. This is essential in lowering the viscosity of the binder at the point of feeding the magnetic powders to avoid potential powder buildup and enable efficient compounding. The extrusion temperatures along the barrel decrease toward the die (discharging zone). A lower temperature is essential here to facilitate the pressure build-up required for the pumping action and a continuous streamlined bonded magnetic monofilament output at the die. As shown in Figure 1a, the higher the packing fraction, the higher the temperature of the optimized extrusion temperature along the extruder barrel.

The bonded magnetic monofilaments were printed into magnetic test samples via the magnetic field-assisted additive manufacturing (MFAAM) in zero and in 0.5 Tesla. The print field was applied parallel to the print direction (see Fig. 1b) using permanent NdFeB magnets strategically situated around the nozzle creating a field of 0.5 Tesla in the nozzle and 0.3 Tesla at the print location. In order to avoid deflection of the suspension during the printing process our print-field head was designed to have a small field gradient parallel

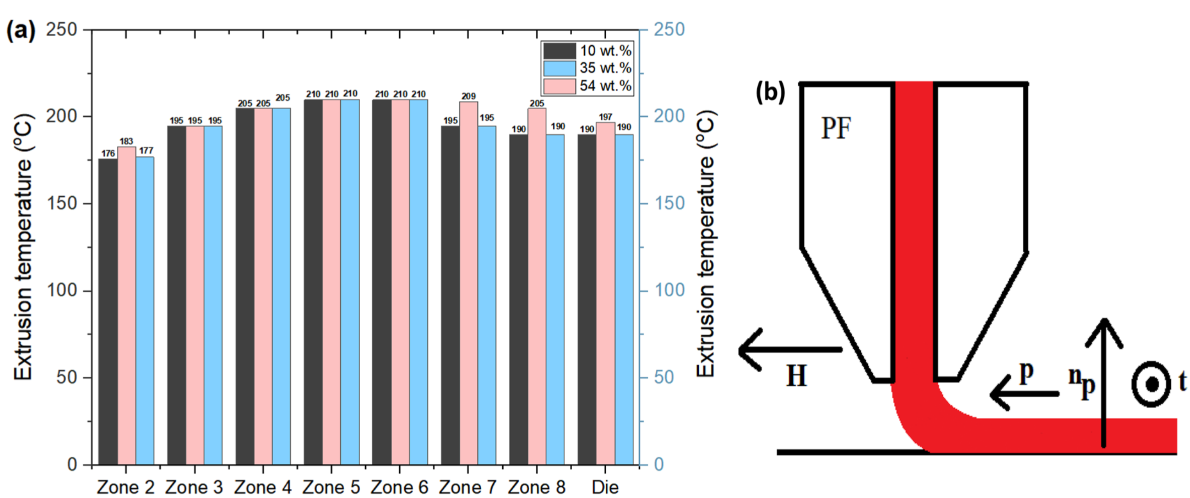

FIG. 1. (a) Extrusion temperatures along the barrel from the intake (zone 2) to the exit (die); (b) definitions of print field and various directions in the sample (parallel to print bed normal (n_p); parallel to print direction (p); parallel to transverse (t)).

to the print bed (5 Oe/mm)[11] allowing us to apply a larger magnetic field during printing than others[12–14] without distorting the printing process. The orientation of the sample during the print process is defined by three orthogonal vectors, i.e., print bed normal direction $\mathbf{n_p}$, print direction $\mathbf{p}$, and transverse direction $\mathbf{t}$. The definition of the principal axis with respect to the print bed and print field is provided in Figure 1b below. All samples were printed on a glass print bed that is kept at 135 °C using an extrusion temperature of 255°C at a print speed of 20 mm/sec. The sample was air-cooled during the print process. The MFAAM process was achieved by modifying a LulzBot FFF 3D printer.[11]

The magnetic hysteresis curves of the bonded magnetic composites were obtained using a MicroSense EZ9 Biaxial Vibrating Sample Magnetometer (VSM), which was conducted at room temperature. The magnetic hysteresis curves of the fabricated bonded magnets were measured along three different sample mount orientations (S2) to obtain magnetic signals for when the VSM driving field were parallel to print bed normal ($\mathbf{n_p}$), parallel to print direction ($\mathbf{p}$), and parallel to transverse direction ($\mathbf{t}$)).

III. RESULTS AND DISCUSSION

The magnetic hysteresis loops obtained are magnetic moment values as a function of the driving field (H) between −22000Oe to +22000 Oe. As presented in Fig. 2, the hysteresis loops of the 10 wt.% strontium ferrite bonded magnets 3D printed in no field and parallel to the print magnetic field were selected to understand the magnetic anisotropic characteristics of the field-assisted 3D printed magnets. For the zero-field printed sample, the remanence is largest along the $\mathbf{t}$ and $\mathbf{n_p}$ principal sample axes and is smaller along the print direction $\mathbf{p}$. This indicates that the zero-field printed sample has an easy plane along the radial direction of the filament and a hard axis along its cylindrical axis The 0.3 Tesla MFAAM sample has the largest remanence along the $\mathbf{n_p}$ direction, an intermediate remanence along $\mathbf{t}$, and a small remanence along the $\mathbf{p}$ axis. The large difference in M_r values along the different principal axes of the MFAAM sample indicates that the anisotropy of the field assisted 3D printed bonded magnets in Figure 2b is more defined when compared to that of the no field 3D printed bonded magnets in Figure 2a. This stronger anisotropy is attributed to the alignment of the individual magnetic particles in the direction of the print magnetic field which overshadows the effect of the flow on the particle orientation. This could be ascribed to a minimized resistance experienced by the magnetic particles to rotation by the alignment field due to the relatively smaller viscosity of the nylon 12 binder at the 3D printing extrusion temperature (255 °C). The higher remanence observed in the field-assisted strontium ferrite bonded magnets is in line with the results reported by Nlebedim *et al.* NdFeB bonded magnets[15] where it was observed that the remanence increases with alignment magnetic field, and more rapidly between unaligned state (as printed, 0kOe) up to 10kOe. Since the saturation magnetization of strontium ferrite is significantly lower, here a lower alignment field of 0.5T is sufficient.

Furthermore, the hysteresis curve's squareness (S-values) estimated from the hysteresis loops can further explain the magnetic anisotropy behavior of the strontium ferrite bonded magnets. The S-values were obtained as the ratio of the magnetic remanence to magnetic saturation, as presented in Fig. 3. The remanence-to-saturation magnetization ratio (M_r/M_s) is considered as the degree of alignment, which is a dimensionless parameter that can be used to investigate the effect of magnetic field assisted 3D printing on the alignment of the magnetic particles in the strontium ferrite bonded magnets.

As shown in Figure 3a, the S-values of the samples printed in no magnetic field appear to be smallest for the hysteresis curves measured parallel to the print direction across all loading levels. The S-values are closely similar for the other two directions measured perpendicular to the print direction (i.e., parallel to transverse and parallel to print-bed normal) and are significantly larger. This observation is similar to the results reported by Ahmed *et al.*[16] for freely extruded filaments that are not 3D printed and thus do not fold over on a print bed. These results suggest the existence of a flow-induced anisotropy with an easy axis that is perpendicular to the extruded filament's cylindrical axis. It is hypothesized here that such

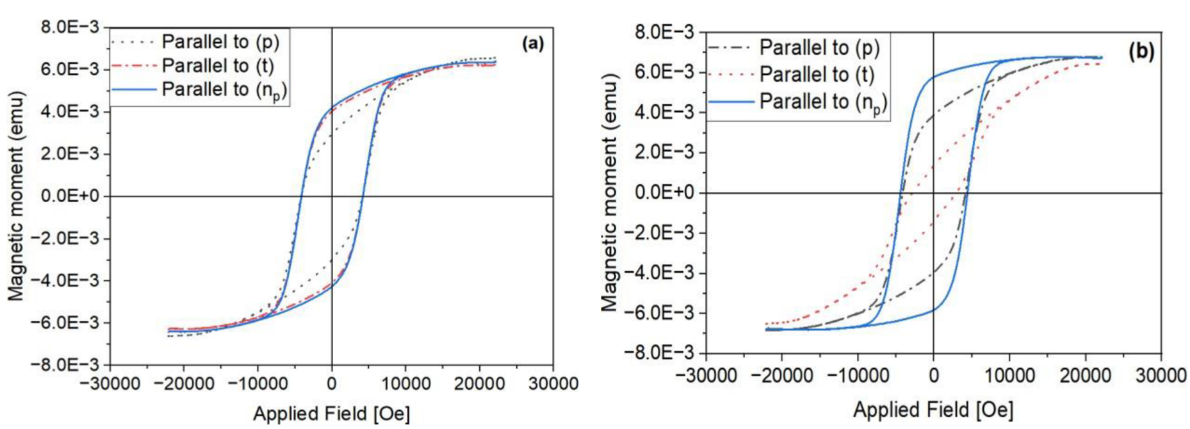

FIG. 2. Magnetic hysteresis loop of 10 wt.% strontium ferrite bonded magnet for different directions of the VSM field: (a) sample printed in no-field; (b) sample 3D printed parallel to print magnetic field.

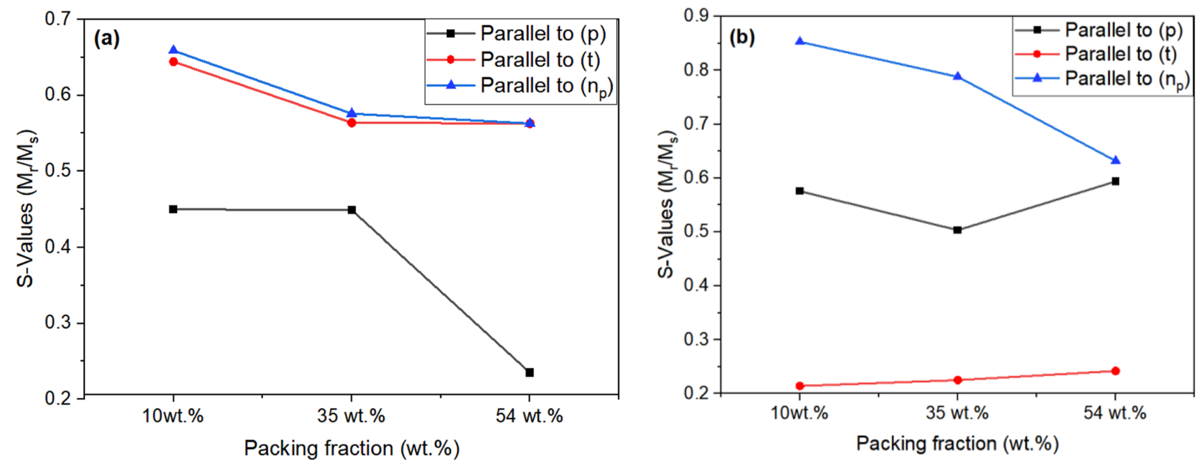

FIG. 3. S-values for the three principal axis directions of 3D printed bonded strontium in (a) no magnetic field and (b) of MFAAM sample.

anisotropy originates from the effect the shear flow has on the orientation of the strontium ferrite platelets near the wall of the extrusion nozzle.[17–20] In addition, the die swell and flow as the suspensions are squeezed between the nozzle and the print bed are expected to contribute to some sort of flow-induced anisotropy.[21–23]

Figure 3b shows the S-values of the magnetic field-assisted 3D printed strontium ferrite bonded magnets. It could be observed that the S-values of the samples when parallel to print direction (**p**) are in between the two other S-values measured parallel to transverse (**t**) and parallel to the print bed normal ($\mathbf{n_p}$). It is noteworthy that the largest S observed is parallel to the print-bed normal. As shown in the no field printed samples, the main alignment happens when the molten suspension is in the extruder nozzle although now the print field forces the particles to orient. When the extruded magnetic composite is folded over on the print-bed the whole material simply rotates with the easy axis parallel to the print-bed normal with little rotation of the individual magnetic particles by the print field outside the nozzle on the print bed. Although the magnetic field is not that different near the print-bed away from the nozzle die, the temperature apparently quickly drops once the material is deposited on the print-bed, and little additional rotation happens. It is expected that the print-field still will have some effect on the orientation of the particles,[24,25] so an easy axis that is rotated with the print-bed normal is expected. This type of canted easy axis was indeed observed by others[26,27] on 3D-printed samples. The higher S-values observed in the field-assisted 3D printed bonded magnets parallel to the print bed normal (n_p) are attributed to the appreciable alignment of the individual magnetic particles by the print field, when compared to bonded magnets printed in no magnetic field.

IV. CONCLUSIONS

Strontium ferrite bonded magnetic composites were fabricated via twin screw extrusion technology using nylon 12 binder to produce a packing fraction of 10, 35, and 54 wt.% strontium ferrites magnetic composites monofilaments, which was used to demonstrate magnetic field assisted additive manufacturing (MFAAM) process. The MFAAM was achieved by modifying a standard FFF 3D printer to 3D print the magnetic composites in no field and using 5kOe magnetic field strength to 3D print parallel to the print field direction. The magnetic hysteresis signatures of the strontium ferrite bonded magnets were obtained using a biaxial vibrating sample magnetometer (VSM) along the principal axes of the samples to evaluate the magnetic anisotropy of the fabricated strontium ferrite bonded magnets.

The S-values of the hysteresis curves, which illustrate the degree of alignment, appear to be largest along the 3D printed sample's print bed normal. This indicates that the major alignment of the no-field samples occurs in the extruder nozzle. When the extruded magnetic composite is folded over on the print-bed, the whole material simply rotates such that the easy axis rotates 90° parallel to the print-bed normal with appreciable rotation to the individual particles. This signifies that the easy axis lies in the radial direction as the field-assisted 3D printed bonded magnets folds on the print bed. As illustrated in the magnetic hysteresis loops, there was a well-defined easy and hard axis in the field assisted 3D printed magnetic composites when compared those printed in no magnetic field, which indicates that a print field is more effective in inducing an anisotropy than a flow. Note that the packing fraction of the magnetic composites studied here is too low for the sample shape to have an effect on the sample's magnetic anisotropy. The observed anisotropy of the zero-field samples is due to the partial alignment of the individual single crystalline OP56 particles. The observed anisotropy of the MFAAM samples is due to the alignment of the individual particles but also because of the agglomeration of particles in chains as previously investigated.[24,25] Summarizing both types of samples are anisotropic and have a hard axis along the print direction and have an easy plane perpendicular to the print direction for the no-field samples and an easy axis parallel to the print bed normal for the MFAAM samples. Higher remanence values and more hysteresis loop squareness, which are indications of higher magnetic energy density were observed along the print field direction in the MFAAM magnetic composites.

SUPPLEMENTARY MATERIAL

See the supplementary material for images of Process 11 twin screw extruder configurations (S1) and the sample mount orientations (S2).

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AUTHOR DECLARATIONS

Conflict of Interest

The authors have no conflicts to disclose.

Author Contributions

Mandesh Khadka: Conceptualization (equal); Data curation (equal); Formal analysis (equal); Investigation (equal); Methodology (equal); Visualization (equal). **Oluwasola K. Arigbabowo**: Formal analysis (equal); Investigation (equal); Visualization (equal); Writing – original draft (equal); Writing – review & editing (equal). **Jitendra S. Tate**: Funding acquisition (equal); Resources (equal); Supervision (equal). **Wilhelmus J. Geerts**: Formal analysis (equal); Funding acquisition (equal); Resources (equal); Supervision (equal); Writing – review & editing (equal).

DATA AVAILABILITY

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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