3D Printing of Thermoplastic-Bonded Soft- and Hard-Magnetic Composites: Magnetically Tuneable Architectures and Functional Devices

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Magnetic materials are key players for the development and implementation of novel technologies. However, the integration and tunability of these materials in complex designs remain challenging due to constraints in currently available manufacturing approaches. Herein, the fabrication of 3D printing (3DP) thermoplastic-bonded magnetic composites is investigated with both tailored geometries and magnetic properties using a customized fused deposition modeling 3D printer. First, the level of complexity that can be achieved by printing magnetic architectures with different geometries is shown. Next, it is shown that architectures with tailored magnetic properties (saturation magnetization and coercivity) can be achieved by combining prints with different magnetic properties (i.e., hard- and soft, or hard- and semi-hard). Additionally, to demonstrate the versatility and powerfulness of fused deposition, self-contained mechanisms, which comprise multiple parts (magnetic and non-magnetic) without the need for assembly, are successfully fabricated. As an example, planetary magnetic gearboxes are printed with different configurations and their potential applicability as magnetic rotary encoders is demonstrated.

1. Introduction

Additive manufacturing, also known as 3D printing (3DP), has recently sparked a revolution in material processing technologies. Although the origins of 3DP can be traced back to the 70s,[1] we have recently seen a significant increase in the number of publications dedicated to 3DP, as the technique has become more mature and accessible to the research community. Chemical reactors,[2] bionic organs,[3] tissue scaffolds,[4] soft-mechanical fasteners,[5] electrodes,[6] batteries,[7] strain sensors,[8] and pneumatic actuators[9] are a few examples of 3D-printed tools and setups. The advantages of 3DP include the fabrication of architectures with complex hollow parts,[10] the use of a wide pallet of materials (single or composite),[11] rapid and inexpensive prototyping, and a high level of customization. Recently, there has been an interest in printing hard magnets, owing to the significance of these materials in a wide range of applications.[12] Hard-magnetic components are ubiquitous in industrial equipment and consumer goods.[13]

However, the processability and integration of these materials have hampered further development of advanced magnetic technologies, which are crucial in the fields of robotics, renewable energy, aeronautics, or automotive engineering. To date, neodymium–iron–boron and samarium–cobalt are the magnetic compounds with the highest energy products, followed by aluminum–nickel–cobalt (AINCO) alloys, and barium and strontium ferrites.[14] Current manufacturing methods for these materials such as injection molding and sintering face several obstacles, particularly in the production of complex geometries.[15] Injection molding offers significant design freedom, but the technique cannot be used to form parts with a complex combination of materials. Sintered magnets require the use of pressing tools, which limit the complexity of the parts. Postprocessing machining is also challenging, as hard-magnetic materials are usually brittle, pyrophoric, and prone to cracking.[16] By exploiting the capabilities of 3DP, high-performance composite magnets can be precisely patterned in arbitrary shapes and architectures. For example, the incorporation of custom-made, complex-shaped magnets in electric motors for electric
vehicles and robots would increase performance and reduce the overall weight.\textsuperscript{[1,3]} In addition, using 3DP, custom magnets can be tailor-made for specific applications and directly printed on the desired structure, minimizing the additional costs of further assembly of parts and components.

In this work, we investigated the manufacturing of 3DP thermoplastic-bonded magnetic composites (TBMC), with both tailored geometries and magnetic properties, using a customized fused deposition modeling (FDM) 3D printer. The composites are made of polyamide (PA) and contain magnetic particles of different sizes and magnetic behavior. Three TBMC filaments featuring different magnetic properties were produced and tested. The first stage was to show that the properties of the feedstock magnetic materials are preserved in the resulting magnetic structure. We then demonstrated how to successfully adjust the magnetic behavior of printed structures by printing parts with different magnetic attributes using a double nozzle in our customized printer. To demonstrate the versatility of our double-nozzle–customized FDM printer, we also fabricated 3D structures made of nonmagnetic and magnetic parts. To this end, we printed planetary magnetic gearboxes with different configurations, and we demonstrated their potential applicability as magnetic rotary encoders (MRE). MREs are key components in industrial robotics and are used for positioning and controlling the motion of robotic tools. MREs consist of a magnetic ring or disc, on which a magnetic pattern of alternating poles is written. The absolute and/or incremental rotation of these rings or discs can be measured using a magnetic field sensor. By 3DP the magnetic material directly onto the rotary shaft, the need for additional sensor components is eliminated, and a wide selection of materials can then be used. By using a customized FDM 3D printer with a double nozzle, the planetary gearbox combining nonmagnetic and magnetic gears was successfully manufactured, without the need for a postassembly procedure.

2. Results and Discussion

2.1. Magnetic Filament Fabrication and Characterization

The 3DP was carried out using a Felix 3.0 FDM 3D printer (FELIXprinters, The Netherlands). In an FDM printer, a continuous filament of a thermoplastic material is fed through a heated nozzle; the material melts and is deposited on the working piece. The nozzle moves first along the x and y planes, depositing the material on the printing plane, before moving the printing table downward to deposit material in the z plane to form a new layer. Figure 1a shows a schematic drawing of the production sequence for the fabrication of the 3D-printed TBMC devices. First, TBMC pellets were fed to an extruder to create printing filaments. These pellets are commonly used in the production of molded magnetic objects and are cast by the pressurized injection of TBMCs pellets into a heated mold. In our study, the pellets were forced through a heated chamber by an extrusion screw. The polymer matrix of the composite pellets was melted, and the liquid passed through the nozzle of the extruder forming the filament. The nozzle had to be sufficiently cooled to ensure that the forming filament was below the glass transition temperature before leaving the end of the nozzle. This procedure was necessary to avoid any warping of the filament caused by internal thermal stresses from the extrusion heat. More details on the extruder and the filament process can be found in the Supporting Information. Three different commercially available PA-based magnetic composite pellets (MATE CO LTD) were used in this work: Nd\textsubscript{2}Fe\textsubscript{14}B (NFB/PA12), Fe\textsubscript{6.72}Si\textsubscript{1.27}Al (FSA)/PA12, and SrFe\textsubscript{12}O\textsubscript{19}/PA12 (SFO/PA12). The extrusion parameters for the production of the filaments (speed of the extruder screw and the temperature of the heating element) were optimized for each individual material. Table S1, Supporting Information, shows the extrusion temperatures used to achieve the optimal filament quality with three different composite materials. The extruded filaments displayed a diameter of 1.75 mm, and their quality was assessed by visual inspection. Figure 1b shows the resulting TBMC filaments. The scanning electron microscopy (SEM) micrographs reveal that filaments made of SFO/PA12 exhibited a smooth surface, in contrast with NFB/PA12 and SFO/PA12, which displayed a rougher surface morphology. This is related to the size of the particles, which correspond approximately to 1.5, 50, and 70 μm for SFO, FSA, and NFB respectively. The FSA/PA12 composite exhibited the roughest surface and was also the most brittle among the three materials tested in this work. Figure 1c shows the hysteresis loops of cross-sections of the filaments, with an approximate volume of 4.8 mm\(^3\). The magnetic properties of structures were characterized using a Vibrating Sample Magnetometer (VSM, MicroSense EZ9). As expected, FSA/PA12 showed a soft-magnetic behavior with a saturation magnetization of \(M_s = 95.71\, \text{emu}\, \text{g}^{-1}\), a remanence of \(M_r = 0.19\, \text{emu}\, \text{g}^{-1}\), and a coercivity of \(H_c = 24\, \text{Oe}\). With SFO/PA12, we observed a moderately hard-magnetic behavior, with \(M_s = 44.95\, \text{emu}\, \text{g}^{-1}\), \(M_r = 24.31\, \text{emu}\, \text{g}^{-1}\), and \(H_c = 2910\, \text{Oe}\), respectively. NFB/PA12 exhibited distinct hard-magnetic features with \(M_s = 93.76\, \text{emu}\, \text{g}^{-1}\), \(M_r = 66.86\, \text{emu}\, \text{g}^{-1}\), and \(H_c = 8010\, \text{Oe}\). The magnetic characterization showed that the results were in agreement with the magnetic data provided by the pellet supplier.

2.2. 3D Printing of Magnetic Architectures

The printing parameters were optimized for each filament material by printing lines and measuring their thickness reproducibility. The governing parameters were the nozzle temperature, the flowrate of the filament, and the printing speed. Table S2, Supporting Information, summarizes the optimized printing temperature values for each material. Similar to the filament production, the NFB/PA12 required the least amount of thermal energy. This could be explained by the larger particle size as they retain heat much longer than smaller ferrite particles. The printing parameters also depend on the complexity of the structure and the volume of the material printed. We found that the printing temperature for smaller structures (<1 cm\(^3\)) should be set to a lower value from that reported in Table S2, Supporting Information, for two reasons: first, each layer of a smaller object has less time to cool, as the next layer will be printed on top much faster; second, the nozzle is longer in the vicinity of the already printed part of the object, so the printed part is overheated and precision and resolution is lost. However, with bigger printed parts, the layers have time to cool.
down before subsequent layers are deposited, so the available thermal energy decreases, resulting in an inferior adhesion. A critical parameter in the printing process is the flowrate, which is defined as the percentage of material that exits the nozzle at a given position and is related to the printing speed. Figure S3, Supporting Information, shows the main differences among 1) low (80%); 2) optimal (110%); and 3) high (130%) flowrates. For low flowrates, gaps formed in the structure caused by lack of material, whereas high flowrates led to oversized structures. More information on the optimization of the printing parameters can be found in the Supporting Information. Several magnetic architectures made of different types of composites are shown in Figure 2a to show the capabilities and complexity that can be achieved—from the Eiffel Tower at a scale of 1:1200 printed with NFB/PA12, to a complex helical structure combined with a conical body printed with the SFO/PA12. Figure 2b shows a disc and a rectangular 3DP structure and their corresponding SEM images. From the SEM image, each individual printed layer can be clearly observed. The thickness of each layer was ≈100 μm.
Taking advantage from the double nozzle (or double extruder) provided by the Felix 3.0 FDM printer, we also fabricated complex structures consisting of magnetic and nonmagnetic materials. Double nozzle refers to two units positioned parallel to each other as a unit depicted in Figure 2Sb, Supporting Information. Figure 2c shows the ETH logo printed with
SFO/PA12 TBMC on a polylactic acid (PLA) surface (Figure 2c(i)) and three magnetic blocks connected with nonmagnetic PLA hinges (Figure 2c(ii)). The structure was subjected to manual bending to assess the adhesion of the magnetic material on the substrate (Figure 2c(iii)). The SFO/PA12 TBMC bonded strongly with PLA, creating a seamless connection, which was resistant to bending and shear stress.

We then evaluated the magnetic field flux densities $B$ that simple 3D-printed structures are able to generate after magnetization. For this, cylindrical magnets were printed using hard-magnetic NFB/PA12 and SFO/PA12 composites and magnetized using the VSM with a magnetic field of 2.2 T, which saturated both composite materials. The magnetic flux densities generated by these cylinders were measured with a hall sensor attached to a precision positioner. The sensor was moved along the central axis above the cylinder magnets in 100 $\mu$m steps, and the measurements were recorded over a distance of 5 mm. Figure 2d shows the 3D-printed cylindrical shape and the measurement procedure. The measured flux densities $B$ generated by the printed cylindrical magnets were compared with the theoretical flux densities $B$ obtained from the following analytical equation

$$B = B_r \left( \frac{D + z}{\sqrt{R^2 + (D + z)^2}} - \frac{z}{\sqrt{R^2 + z^2}} \right) \quad (1)$$

where $B_r$ corresponds to the remanent magnetic flux density. $D$ corresponds to the height of the cylinder, $R$ to the radius and $z$ to the distance from the pole face. The permeability and the remanence were provided by the manufacturer (MATE CO LTD) and can be found in Table S4, Supporting Information, together with the printed cylinder dimensions. The results of the measured cylinders and the analytical equation are plotted in Figure 2d(iii). We can see that the measured values are in good agreement with the analytical model.

![Figure 2c](image1.png)

![Figure 2d](image2.png)

**Figure 2.** 3D-printed structures made of magnetically dissimilar TBMC parts. a, i) Illustration of a disc composed of two layers, a, ii) 3D-printed disc where the bottom layer is made of a hard-magnetic TBMC and the top layer of a soft-magnetic TBMC. This strategy can be used to increase the saturation magnetization of a hard-magnetic component without excessively compromising its coercivity and remanence. a, iii) The resulting hysteresis loop of the 3D-printed disc composed of two layers. b, i) Illustration of a crisscrossed-patterned cuboid, b, ii) 3D-printed crisscrossed-patterned cuboid consisting of hard-magnetic TBMC stripes alternating with soft-magnetic TBMC ones. b, iii) The resulting hysteresis loop of the crisscrossed structure made of two different magnetic composites.
2.3. 3D Printing of Magnetic Structures with Customized Magnetic Properties

A powerful feature that can be exploited using a dual-nozzle process is shown in Figure 3a, which shows 3D-printed structures made of magnetically dissimilar TBMC parts. By combining TBMCs with different magnetic properties, the overall magnetic behavior can be customized. For example, it is possible to adjust the coercivity, the saturation magnetization, or the remanence of the final printed structure. Two different architectures were printed using this approach. The first is a disc composed of two layers, where the bottom layer is made of a hard-magnetic TBMC and the top layer of a soft-magnetic TBMC (Figure 3a). This strategy can be used to increase the saturation magnetization of a hard-magnetic component without excessively compromising its coercivity and remanence. Figure 3a(iii) shows the resulting hysteresis loop of the 3DP disc composed of two layers. By increasing the relative volume of the soft-magnetic layer (x\% FSA) with respect to that of the hard-magnetic part ([(1 - x)\% SFO], the $M_s$ of the bilayer composite increases, while the coercivity decreases. By tuning the relative amounts, magnetic materials with virtually any intermediate values of coercivity and saturation magnetization could be obtained. More complex arrangements with customized magnetic properties are also possible, such as the crisscrossed-patterned cuboid shown in Figure 3b. The structure consists of hard-magnetic TBMC stripes alternated with soft-magnetic TBMC ones. Figure 3b(iii) shows the resulting hysteresis loop of the crisscrossed structure made of two different magnetic composites. By

![Image](https://via.placeholder.com/150)

**Figure 3.** (a) 3D-printed gearbox with a helical teeth profile. (b) Gearbox arranged inside the magnetic manipulation setup. (c) FEM simulation of the magnetic field density of the magnetic corona. (d) Magnetic encoder arrangement used to measure the rotations of the gear. (e) The applied magnetic rotational field and the measured field as a function of time at a magnetic rotational frequency of 3 Hz. (f) Rotating speed of the gear as a function of the applied rotational magnetic frequency.
2.4. 3D Printing of Magnetic Gear Systems

The versatility of the dual-nozzle extruder enables 3DP of self-contained mechanisms, which can contain multiple moving parts, without the need for separate assembly. Gear systems are particularly interesting devices that can be produced using our customized double-nozzle 3D printer. The ability to 3D print gears, or even the entire gear system with mechanically stable magnetic composite components, brings unprecedented opportunities in the field of gear technology such as vibration sensing, torque control, and absolute and incremental encoding, among others. An epicyclic, or planetary, gear system was fully 3D printed using magnetic and nonmagnetic materials, based on the printer and printing parameters explained previously. It consists of five nonmagnetic planet gears, revolving around the central nonmagnetic sun gear and an outer magnetic ring gear (also known as the corona), which meshes with the planet gears. The magnetic parts were printed with hard-magnetic NFB/PA12 TBMC and the nonmagnetic parts with PLA. The gearing system consists of gears with a helical teeth profile, which are perfectly interlocked to block the axial movement of any of the components. Figure 4a shows the 3DP gearbox and the helical teeth profile. Additional results showing a gear system with a central magnetic gear are provided in Figure S4, Supporting Information. The actuation of the gear system was tested in a five-degree-of-freedom electromagnetic system Octomag. Applied fields of up to 40 mT and magnetic field rotation frequencies of up to 5 Hz were tested. The sun gear was fixed with the Octomag frame, and the corona was actuated with the external magnetic field. Figure 4b shows the gearbox in the magnetic manipulation setup. The magnetic corona gear was designed to separate into two half-moons to fit in the VSM space, and both halves were magnetized perpendicular to the rotational axis with a magnetic field of 2.2 T.

Finite element method (FEM) simulations were performed with COMSOL MultiPhysics to analyze the magnetization behavior. Figure 4c shows the FEM of the magnetic flux density $B$ of the magnetic corona. As we can see, the magnetic corona behaves in the same manner as that of a cylinder with two distinct poles. Positions (i) and (ii) in the figure represent the points with the highest field, i.e., the poles of the magnetic corona. Figure 4d shows a concept image of a possible encoder example. As the corona gear rotates, a sinusoidal signal is registered. For our experiment, the rotation of the gear was provided via an external magnetic field as opposed to a mechanical system. A magnetic field sensor was placed next to the magnetic corona to measure the field strength and magnetic field direction. In this experiment, an Infineon 3D hall sensor was placed next to the magnetic corona at a distance of 1 mm. Figure 4e shows the applied magnetic rotational field and the measured field for an applied rotational frequency of 3 Hz. Figure 4f shows the linear behavior between the two rotational speeds of the applied field and the rotation of the gear.

For our experiment, the maximal applied rotational frequency (cutoff frequency) was measured at 28 Hz. Above this frequency, the gearbox starts to stutter and the motion changes from a revolving to a shaking motion. The cutoff frequency and the shaking motion are related to the interplay between rotational drag and the alignment of the magnetization of the gear with the external magnetic field. Above 28 Hz, the drag generated by the gears is sufficient to make the magnetic gear lag back. The induced lag allows the external magnetic field to overtake the gear itself, causing the torque on the magnetic gear to switch direction back-and-forth, producing the rocking motion of the gearbox.

3. Conclusions

In this work, we produced three different TBMCs with distinctive magnetic properties and characterized their magnetic properties via magnetic hysteresis loop measurements, FEM simulation, and analytical calculation. The produced filaments were then used in conjunction with a dual-head FDM printer to 3D print different smart magnetic structures. The printed structures demonstrated that the produced materials transfer their magnetic properties to the printed structure without evidence of deterioration. A variety of TBMC complex shapes and/or tunable customized magnetic properties were produced showing the high complexity that can be achieved by 3DP. Finally, we successfully fabricated and actuated an epicyclic magnetic gear system consisting of magnetic and nonmagnetic parts. The use of 3DP for magnetic applications represents a leap toward on-site versatile fabrication of complex magnetic devices, which would otherwise be unfeasible with conventional manufacturing techniques.

Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

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Conflict of Interest

The authors declare no conflict of interest.

Keywords

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[1] Spie, SPIE Professional 2013, https://spie.org/news/spie-professional-magazine/2013-january/chuck-hull?SSO=1.

[2] M. D. Symes, P. J. Kitson, J. Yan, C. J. Richmond, G. J. Cooper, R. W. Bowman, T. Vilbrandt, L. Cronin, Nat. Chem. 2012, 4, 349.

[3] M. S. Mannoor, Z. Jiang, T. James, Y. L. Kong, K. A. Malatesta, W. O. Soboyejo, N. Verma, D. H. Gracias, M. C. McAlpine, Nano Lett. 2013, 13, 2634.

[4] B. Leukers, H. Guerboukha, M. Girard, A. D. Squires, R. A. Lewis, M. Skorobogatiy, Adv. Opt. Mater. 2016, 4, 2085.

[5] A. L. Rutz, K. E. Hyland, A. E. Jakus, W. R. Burghardt, R. N. Shah, Adv. Mater. 2015, 27, 1607; b) N. Maldonado, V. G. Vegas, O. Halevi, J. I. Martinez, P. S. Lee, S. Magdassi, M. T. Wharmby, A. E. Platero-Prats, C. Moreno, F. Zamora, P. Amo-Ochoa, Adv. Funct. Mater. 2019, 29, 1808424.

[6] C. Huber, C. Abert, F. Bruckner, M. Groenefeld, S. Schuschnigg, I. Teliban, C. Vogler, G. Wautischer, R. Windl, D. Suess, Sci. Rep. 2017, 7, 9419; b) C. Huber, C. Abert, F. Bruckner, M. Groenefeld, O. Muthsam, S. Schuschnigg, K. Sirak, R. Thanhoff, I. Teliban, C. Vogler, R. Windl, D. Suess, Appl. Phys. Lett. 2016, 109, 162401; c) E. Peng, X. Wei, T. S. Herrn, U. Garbe, D. Yu, J. Ding, RSC Adv. 2017, 7, 27128; d) L. Li, B. Post, V. Kunc, A. M. Elliott, M. P. Paranthaman, Scr. Mater. 2017, 135, 100; e) M. Ortner, C. Huber, N. Vollert, J. Pilz, D. Suss, in IEEE Sensors, Glasgow, United Kingdom, 29 October–1 November 2017, 190; f) A. Shen, C. P. Bailey, A. W. K. Ma, S. Dardona, J. Magn. Magn. Mater. 2018, 462, 220; g) X.-Z. Chen, Z.-X. Cheng, L. Liu, X. Yang, Q.-D. Shen, W.-B. Hu, H. Li, Colloid Polym. Sci. 2013, 291, 1989.

[7] O. Gutleisch, M. A. Willard, E. Bruck, C. H. Chen, S. G. Sankar, J. P. Liu, Adv. Mater. 2011, 23, 821.

[8] M. Ortner, C. Huber, N. Vollert, J. Pilz, D. Suss, in IEEE Sensors, Glasgow, United Kingdom, 29 October–1 November 2017, 190; f) A. Shen, C. P. Bailey, A. W. K. Ma, S. Dardona, J. Magn. Magn. Mater. 2018, 462, 220; g) X.-Z. Chen, Z.-X. Cheng, L. Liu, X. Yang, Q.-D. Shen, W.-B. Hu, H. Li, Colloid Polym. Sci. 2013, 291, 1989.

[9] J. M. D. Coey, Scr. Mater. 2012, 67, 524.

[10] J. Jacimovic, F. Binda, L. G. Herrmann, F. Greuter, J. Genta, M. Calvo, T. Tomse, R. A. Simon, Adv. Eng. Mater. 2017, 19, 1700098.

[11] S. Pane, E. Pellicer, K. M. Sivaraman, S. Surinach, M. D. Baro, B. J. Nelson, J. Sort, Electrochim. Acta 2011, 56, 8979.