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INVESTIGATION ON THE FEASIBILITY OF PRODUCING POLYMER-BASED MAGNETIC COMPOSITES FROM RECYCLED PERMANENT MAGNETS USING FUSED DEPOSITION MODELLING

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Abstract

Polymer-based composites with magnetic properties represent a promising class of materials that combine conventional polymer characteristics (low density, high electrical resistance, enhanced flexibility, and processability, among others) with magnetic properties typically associated with ferromagnetic or ferrimagnetic metals and alloys. In the present study, micrometric particles of a neodymium-iron-boron (NdFeB) alloy are compounded with a flexible thermoplastic matrix comprising polybutylene adipate-co-terephthalate (PBAT). The NdFeB alloy is recovered from permanent magnets of obsolete hard drives and is demagnetised, ground to powder under an inert atmosphere, and finally sieved to a particle size below 50 μm. The obtained powder is mixed with the polymer using a twin-screw extruder. The composite material containing the NdFeB particles is then processed to obtain a

calibrated filament, which is used for the fused deposition modelling (FDM) three-dimensional (3D) printing of magnetic composites. Different routes to improve the ferromagnetic behaviour are investigated, including increasing the filler content up to 50 wt%, orientating the particles during the 3D process by printing directly onto a permanent magnet and subsequent magnetisation of the samples using a pulse magnetiser. The results of the structural, thermomechanical, and magnetic characterisations are presented and discussed.

1. Introduction

Permanent magnets are of great importance in the current green and digital transition towards a sustainable future [1]. They are currently employed in several general-purpose applications, including motors and electronic components for laptops and smartphones. Alloys comprising neodymium, iron, and boron (NdFeB) represent a significant proportion of the materials employed in the production of permanent magnets [2]. However, there are several drawbacks associated with this material, the most significant of which is the reduced availability of raw materials, which has led to an increase in the cost of permanent magnets [3, 4]. Consequently, the recovery of NdFeB components from waste electrical and electronic equipment (WEEE) represents a crucial objective that can facilitate the utilisation of these materials in innovative applications [3].

A prevalent utilisation of NdFeB magnets is as components of old hard drives (HDs), with life cycle ranging from 2 to 3 years [5]. Currently, permanent magnets recovered from waste HDs cannot be directly reused, and reshaping these components without degrading them can be quite difficult and can sgnificantly increase the overall costs, making the recycling route infeasible [6]. Common recycling routes of permanent magnets can be based on material recycling by smelting, element recycling by leaching or direct magnet recycling by hydrogen (H) decrepitation, which present many problems, namely heterogeneity, obtaining representative samples, strong magnetic forces, oxidation, and brittleness [7,8].

Several studies regarding the production of magnetic composites have been published, using different thermoplastic and thermoset polymer matrixes (such as PA, PE, PLA, epoxy resin, etc.) and magnetic materials (ferrites, SmFeN, NdFeB, etc.) [9-13]. Indeed, polymer-based composite production offers a solution to produce more complex shapes and reduce the overall costs of the process. Embedding magnetic particles into a polymer matrix leads to magnetically active composites with lower magnetic properties, but also lower specific weight and higher flexibility compared to sintered magnets [14]. Also, with these materials, a substantial reduction in costs is achieved [15]. They are intrinsically more sustainable than permanent magnets since are produced using secondary raw materials and with lower energy consumption [8]. For some applications, such as brushless motors, both the weight and cost reduction may be crucial, while the reduction of magnetic coercivity and remanence may still meet the requirements [16]. Moreover, additive manufacturing technologies may be exploited allowing for even more complex shapes and customized products for each application [17-22].

Additive manufacturing (AM) has recently emerged as a promising technology due to its great versatility, flexibility, reproducibility and high speed, , allowing its application in various fields. Polymer-based magnets are produced using a variety of AM technologies, and thermoplastic polymer-magnet composites have been fabricated using the material extrusion technique to realizenear-net shaped magnetic materials directly from 3D models. It offers several clear benefits over traditional manufacturing methods, such as the ability to produce parts with more complex geometries, the elimination of tool requirements and material waste. Additionally, by optimizing topology and adjusting the magnetic filler amount, AM makes it possible to produce permanent magnets with complex magnetic fields.

In the current work, NdFeB components from discarded HDs were ground and sieved in an inert atmosphere, to obtain particles with an average size below 50 μm. The obtained powders were mixed with a flexible thermoplastic polymer, polybutylene adipate-co-terephthalate (PBAT), using a twin-screw extruder. This composite material was pelletized and extruded into calibrated filaments, that were then used for the Fused Deposition Modeling (FDM) 3D printing process.

2. Materials and methods

2.1. Sample preparation

The NdFeB magnetic powders utilised in the present study were sourced from permanent magnets extracted from discarded hard disks (HDs) supplied by RISTA srl (Italy). The magnets were disassembled with an automated system developed by OSAI Automation System (Italy).

The magnets were demagnetised by subjecting them to a heat treatment at 350 °C in a vacuum chamber to prevent oxidation. The magnets were then ground using a homemade impact mill developed at Politecnico di Torino (Italy), which is provided with two separate chambers both kept under vacuum conditions. The NdFeB powder was sieved to obtain particles with a diameter below 50 μm, and the powder was sealed under vacuum to avoid oxidation during storage. The entire process is shown in Fig. 1.

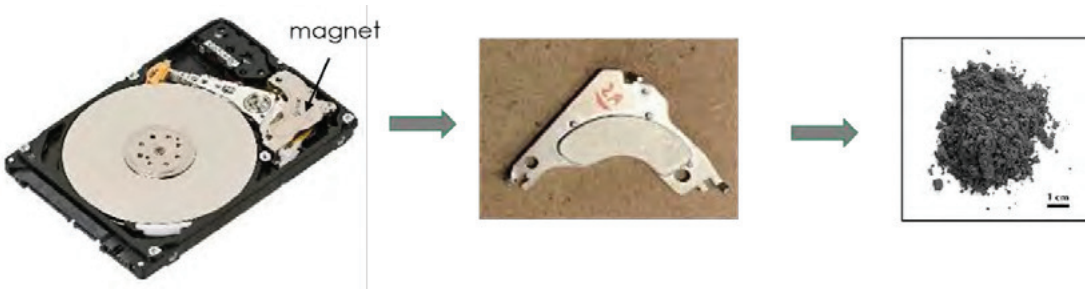

Figure 1 Steps from permanent magnets in HDs to NeFeB powder

The PBAT granules were purchased from MAgMa spa (Italy). The polymer granules were subjected to a grinding step, using an analytical mill (IKA A11 basic) under cryogenic conditions. Mechanical mixing of the ground polymer with the magnetic powder was then carried out, using two different PBAT/NdFeB weight ratios: 70/30 and 50/50. Subsequently, the composite powder was fed into a co-rotating twin-screw extruder (RES-2P/12A Explorer Extruder, Zamak Mercator, Poland) to obtain the deep mixing of the polymer and magnetic powder, thereby producing a homogeneous compound.

In order to obtain suitable filament for FDM 3D printing, the composite obtained through twin-screw extrusion was pelletised and extruded once more using dedicated equipment. This comprised a benchtop system purchased by Felfil srl (Italy), which included a single-screw extruder, an array of cooling fans, and a spooling coil equipped with a diameter control device (Fig. 2).

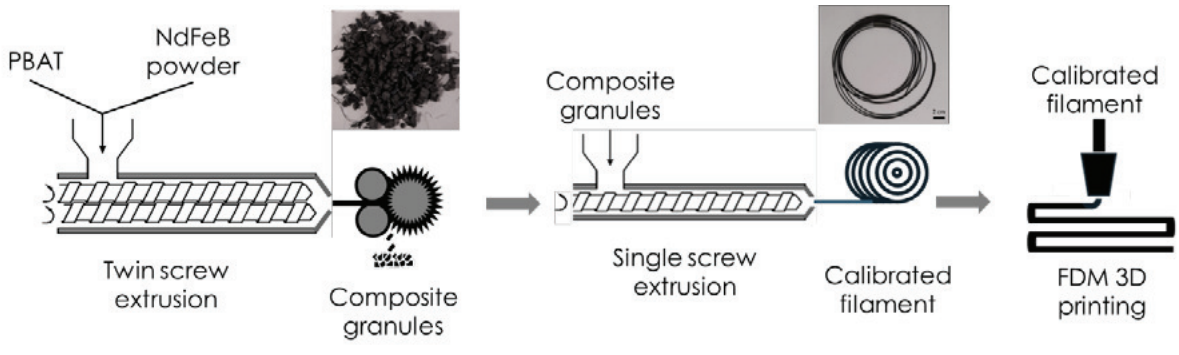

Figure 2 Experimental set-up to produce composite granules and calibrated filaments

The magnetic composite filament was then fed into a Prusa i3 MK3S+ FDM 3D printer, purchased from Prusa (Czech Republic). The printer featured a 0.4 mm nozzle and the printing parameters were set at 0.2 mm for layer height and 0.35 mm for strand width. The infill density during printing was kept at 100% in order to provide the highest density of the samples. It is possible to design samples with

basic cubic or cylindrical shapes, as well as more complex shapes, using Solidworks® or other computer aided design software, and then optimize them for printing using PrusaSlicer.

To enhance the composite magnetic strength, FDM 3D printing was also conducted in the presence of a permanent magnet, enabling the anisotropic orientation of the magnetic particles embedded in the material, as illustrated in Fig. 3.

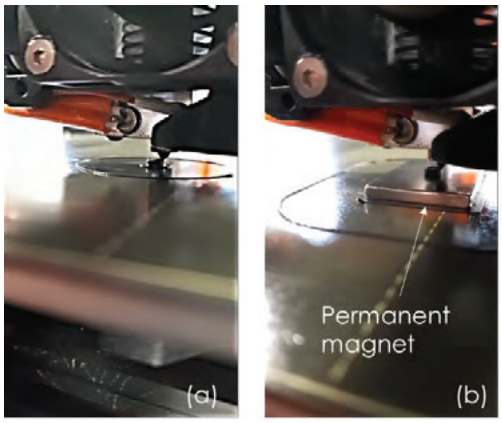

Figure 3 FDM 3D printing of magnetic composite without (a) and with the aid of a permanent magnet

2.2 Material characterization

To assess the printability of composite materials, the melt flow rate (MFR) was also evaluated according to ISO 1133, with a CEAST MF30 melt flow tester (INSTRON). Measurements were conducted at 130 °C applying a weight of 1.2 kg while maintaining constant conditions for all sample types. The skeletal density of the 3D printed samples was measured with the helium pycnometer Ultrapic 1200, purchased from Quantachrome Instruments (USA), according to ASTM 3766.

The obtained samples were characterized in terms of thermo-mechanical properties, to assess the effect of the magnetic filler on the properties of PBAT.

Dynamic Mechanical Analysis (DMA) was carried out using a TA Instruments (USA) model Q800. The measurements were carried out in the temperature range between -60 and 60 °C, with a heating rate of 3°C/min. The samples were mounted on a single cantilever configuration and kept under oscillation with a frequency of 1 Hz and an amplitude of 10 μm. At least 3 samples for each composition were analyzed.

Scanning electron microscopy (SEM) images were acquired with a field emission SEM Nova NanoSEM 450, purchased by FEI company (USA), with a voltage of 12 kV and a working distance of 6 mm. Backscattered electrons were analyzed to observe the distribution of NdFeB particles in the polymer matrix.

A vibrating sample magnetometer physical property measurement system (VSM PPMS, Quantum Design Ldt) was employed to perform measurements on the sample loaded with magnetic powder. The device operates within a temperature range of 2-300 K and can withstand an applied magnetic field of up to 9 T.

3. Results and discussion

Two compositions of flexible magnetic composite materials were prepared within the present work, either containing 30 or 50 wt.% NdFeB magnetic particles. The chosen polymer matrix was PBAT and samples of pure PBAT were also prepared, to determine the effect of the addition of the magnetic alloy to the polymer.

A summary of the weight and volume fractions of the composite matrix and filler is reported in Tab. 1. All the compositions were twin-screw extruded in the same conditions, i.e., extrusion temperature along the extruder between 160 and 180 °C and screw speed of 50 rpm. The obtained composites were

then re-granulated and fed into a single-screw extruder, that provided a filament with a regular diameter of 1.7±0.1 mm. The filament was used to produce simple rectangular specimens with dimensions of 30 x10 x 3 mm^3 suitable for thermo-mechanical analyses.

Table 1. Weight and volume fractions of the composite matrix and filler.

Specimen	*PBAT wt.%*	*NdFeB wt. %*	*PBAT vol.%*	*NdFeB vol. %*
PBAT	100	0	100	0
PBAT + 30% NdFeB	70	30	93	7
PBAT + 50% NdFeB	50	50	86	14

Also, other rectangular as well as cylindrical samples were produced with different aspect ratios, featuring thicknesses that may vary from 0.5 to 30 mm. Depending on the printing parameters more complex shapes can be easily obtained. Some of the produced samples are reported in Fig. 4.

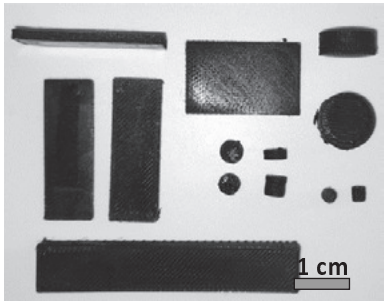

Figure 4. Details of some 3D printed samples.

The homogenous distribution of the magnetic particles in the PBAT matrix is visible in the SEM pictures, reported in Fig. 5.

In the images the various layers that are characteristic of 3D printing can be noted. These layers are perpendicular to the printing direction, which also coincides with the magnetic field generated by the support magnet which is resting on the print base. Taking this configuration as a reference, the samples printed without the magnetic support show a random distribution of the NdFeB particles within the layers, while in the samples printed on the permanent magnet, preferential orientations of the NdFeB powder are observed, perpendicular to the printing layers and aligned along the magnetic field generated by the magnet.

The density of 3D printed samples was measured according to ASTM 3766, and the results are reported in Tab. 2, along with the calculated volumetric density. The theoretical volumetric density ρ_c of the composite is the weighted average of the densities of the different volume fractions of the matrix and the filler. It can be calculated as:

$$\rho_c = \rho_m \cdot V_m + \rho_f \cdot V_f \quad (1)$$

where ρ_m (=1.26 [g/cm^3] and ρ_f (=7.8 [g/cm^3]) are the densities and V_m and V_f are the volume fractions respectively of the matrix and the filler.

The differences between the values of the calculated and measured densities for the composite material are due to the presence of closed porosity within the 3D printed samples, while the increase in the skeletal density of the pure polymer may be due to the presence of small contamination, since 0.5 vol% NdFeB, i.e., 0.08 wt.%, can justify the small increase in the measured value of the density for pure PBAT.

The density increase is accompanied by a decrease in the MFR as the content of NdFeB powder in the composite increases. This is probable since the latter causes the polymer to thicken and its viscosity

to increase, thus decreasing its fluidity and resulting in a more difficult flow.

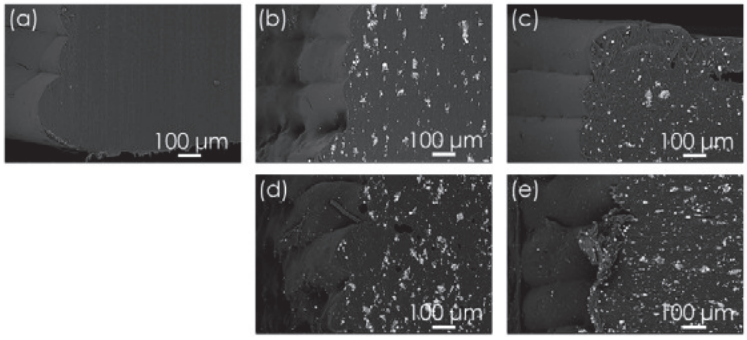

Figure 5. SEM images of pure PBAT (a), PBAT + 30% NdFeB printed respectively with (b) and without (c) the magnet support, and PBAT + 50% NdFeB printed respectively with (d) and without (e) the support magnet

Table 2 Measured skeletal density, calculated density, porosity, and MFR of the prepared samples.

Samples	*Measured skeletal density [g/cm³]*	*Calculated volumetric density [g/cm³]*	*Porosity*	*MFR [g/10 min]*
PBAT	1.2925±0.0006	1.2600*	-	19.5±0.6
PBAT + 30% NdFeB	1.6159±0.0017	1.7242	6%	16.6±1.1
PBAT + 50% NdFeB	1.9037±0.0019	2.2182	14%	11.8±1.0

* From the Technical Data Sheet provided by the polymer seller.

DMA data were used to extrapolate values of the Storage and Loss Moduli of the composite samples at different temperatures. Fig. 6 reports the values of Storage Modulus, E', of pure PBAT, PBAT + 30% NdFeB, and PBAT + 50% NdFeB extrapolated at -40, 20, and 40 °C. As expected, the addition of NdFeB to PBAT is responsible for an increase in the modulus of the material at all temperatures.

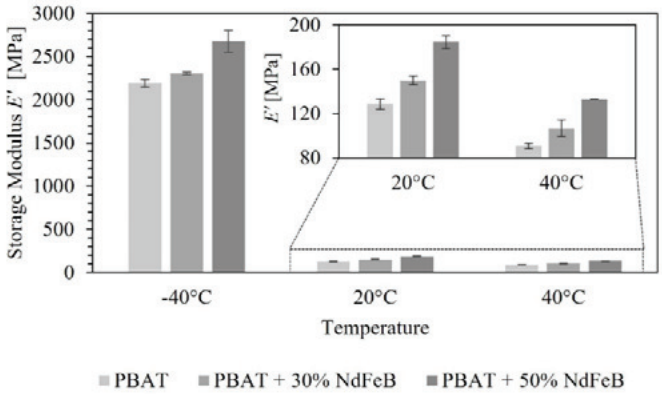

Figure 6. Plot of the Storage Modulus at different temperatures for the pure polymer and the composites. For better visualization, the inset shows a zoomed view of the 20 and 40 °C values of the Storage Modulus for the different compositions.

To quantify the enhancement of the Storage Modulus, the ratios between the composite and the matrix moduli, respectively E'_C and E'_M, were calculated for each composition at different temperatures and are reported in Table 3. At low temperatures, i.e. when the material is in the glassy state, it is possible to note an increase in the Storage Modulus of around 5% and 22%, respectively for the two samples with 30 and 50 wt.% filler concentration compared to the matrix.

On the other hand, at higher temperatures, i.e., between 20 and 40 °C, the Storage modulus of the

composite showed a 17% and 45% improvement, respectively for 30 and 50 wt.% NdFeB samples compared to the pure polymer.

The increase in both the density and Storage Modulus of the produced composite is a direct consequence of the addition of a rigid metal filler. The magnetic particles act as impediments to the polymer chain mobility thus resulting in the increased modulus. Despite these differences in the material properties, the PBAT-based composites containing magnetic particles proved to be easily processable.

The magnetic properties of composite materials are influenced by the magnetic properties of the NdFeB powder and the weight ratio of the powder. Furthermore, magnetic composites have lower magnetic properties than magnetic materials obtained by conventional methods (e.g. sintering) due to the inability to achieve the maximum density of the magnetic powder. The results of magnetic measurements with different magnetic particle contents are presented in Fig. 7.

It can be observed that an increase in saturation magnetisation (Ms), remanence (Br), coercivity (Hcb) and maximum energy product (BHmax) occurs not only with an increase in the quantity of NdFeB particles present in the PBAT matrix, but also as a consequence of the alignment of the magnetic particles within the polymer matrix.

Table 3 Calculated composite/matrix Storage Modulus ratios.

Samples	E'_C/E'_M @ -40°C	E'_C/E'_M @ 20°C	E'_C/E'_M @ 40°C
PBAT + 30% NdFeB	1,05	1,16	1,18
PBAT + 50% NdFeB	1,22	1,44	1,47

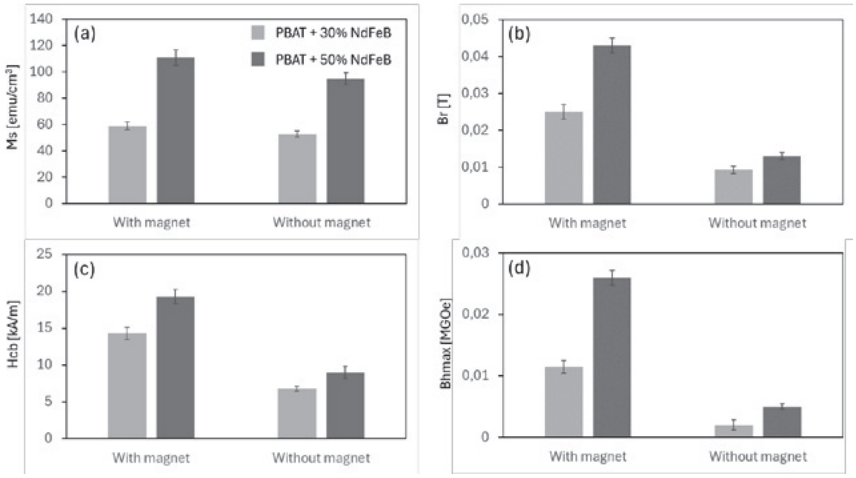

Figure 7. The values of Ms, Br, Hcb, BHmax determined by VSM measurements

4. Conclusions

The present work reports the results of the production of a polymer-based composite material containing up to 50% magnetic powder, the latter showing an average particle size lower than 50 µm.

The composite material was obtained by compounding a biodegradable and flexible polymer, PBAT, with NdFeB powders obtained by reprocessing permanent magnets extracted from discarded hard drives. The magnet extraction process has been designed on purpose to reduce both the environmental impact and costs of permanent magnet production and recycling.

Composite materials were processed as a wire, suitable for FDM 3D printing. Samples with different shapes and aspect ratios were produced. The measurement of the skeletal density revealed the presence of 6% and 14% porosity in the 3D-printed samples. The Storage Modulus of the composite materials respectively containing 30 and 50 wt.% NdFeB increased by 17% and 45% compared to the pure polymer at room temperature. As expected, the magnetic properties of the composite material exhibit a marked improvement with an elevated concentration of magnetic NdFeB particles. This improvement

is also observed when the NdFeB particles are oriented during printing. These findings offer promising indications for polymer-matrix magnetic composites, which could be of significant value in cases where the use of relatively brittle permanent metal magnets is not feasible.

Given the growing interest in polymer composites and additive manufacturing technologies, this study provides a valuable foundation for future experiments and the development of commercially important multifunctional components and polymer-based magnetic composites.

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