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Polypropylene-Based 3D Printed Skin-Core Structures With Thermal Conductivity and Balanced Mechanical Properties

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ABSTRACT

In this work, polypropylene (PP)-based filaments for Fused Filament Fabrication (FFF) containing boron nitride (BN) or talc (T) were developed and used to formulate multi-functional 3D-printed parts with enhanced thermal conductivity and mechanical properties. More specifically, skin-core structures, with BN in surface layers and talc in the core, were optimized and characterized. Results show that confining BN to surface layers improves in-plane thermal conductivity due to filler alignment during printing. Furthermore, the obtained structures maintain mechanical properties comparable to PP/T mono-material samples, despite reduced filler content and the presence of multi-material interfaces, demonstrating the potential of the proposed strategy for creating high-performance multi-material parts through FFF.

1 | Introduction

Starting from a 3D model, the Fused Filament Fabrication (FFF) process enables the construction of an object layer-by-layer through the deposition of a molten thermoplastic filament extruded through a heated nozzle. Compared to more traditional manufacturing techniques, this process offers numerous advantages, such as the ability to produce components with complex geometry, like intricate details and internal voids, easier customization of the object without additional costs associated with creating new molds, and the ability to use multiple materials simultaneously and localize them in specific areas of the sample [1]. One of the limitations of 3D printing, however, is the limited availability of materials suitable for this type of process, as these are typically amorphous or low-crystalline polymers. Otherwise, the use of polypropylene, despite its wide utilization in different industrially relevant fields, is rare. The main reasons lie in its low-dimensional stability, due to its semicrystalline nature, and its Newtonian rheological behaviour, which make both filament production and the printing process challenging [2]. The possibility of extending the use of PP to additive manufacturing techniques would lead to numerous advantages, which is why PP was chosen as the polymer matrix in this study.

The several advantages of 3D printing become even more intriguing when coupled with the utilization of composite polymers with specific properties, owing to the addition of reinforcing and functional fillers. In the present study, hexagonal boron nitride (BN) and talc (T) have been chosen as fillers. BN is an excellent electrical insulator, while simultaneously exhibiting high thermal conductivity. Conversely, talc has been employed as a reinforcing filler to enhance the mechanical properties of the polymer matrix.

The work can be divided into three parts. The first part aimed to produce PP-based filaments suitable for 3D printing, filled with BN or talc, and to evaluate their printability. In the second part, the effect of the 3D printing process on thermal conductivity values in the in-plane and through-plane directions of the samples was evaluated through thermal conductivity measurements on 3D printed specimens. Finally, in the third part, multi-material combined and skin-core 3D structures, based on the PP/BN and PP/TALC composites, were studied with the goal of obtaining specimens that simultaneously exhibit the thermal conductivity properties typical of the PP/BN composite and the superior mechanical properties of the composite containing talc.

2 | Results and Discussion

2.1 | Preliminary Characterization of PP/BN and PP/T Systems

From the DSC analysis, it emerged that, for all formulated compounds, the fillers acted as nucleating agents, leading to a slight increase in crystallinity, which, however, was found to be negligible and did not adversely affect the 3D printability of the materials. The processability of the compounds was evaluated through rheological analysis, from which it was evident that the introduction of fillers resulted in an amplification of the non-Newtonian behavior of the polymer, thus improving the 3D printability of the polymer matrix [3].

2.2 | Filament Making Optimization and 3D Printing

Filaments compatible with the 3D printer were obtained through a single-screw extruder and used to 3D print the test specimens for the following thermal conductivity and mechanical tests. Both the filaments and the 3D specimens were characterized via SEM analysis. From the observation of the interface between the 30BN and 30T layers in the multi-material 3D structures, it was evident that the adhesion between the two composites was optimal, as was further confirmed through the X-ray computed tomography analysis.

2.3 | Thermal Conductivity and Mechanical Properties of Skin-Core Structures

The results of the thermal conductivity measurements on the specimens filled with BN revealed a consistent improvement in thermal conductivity with increasing filler content across all cases. When comparing the values of conductivity in the in-plane and through-plane directions of the 3D specimens, it was found that conductivity was higher in the former case. This confirms that the FFF process helps the alignment of the BN lamellae in the printing direction, allowing for components that preferentially conduct heat along specific directions. For the production of the skin-core structures, the composites 30BN and 30T were utilized, and 3 different skin-core structures, consisting of a total of 15 layers, were investigated.

From the results reported in Table 1, it is evident that in all cases, the in-plane conductivity was higher. Furthermore, the presence of PP/TALC layers in the multi-material structures led to a decrease in conductivity only in the through-plane direction, while the in-plane conductivity appeared to be even better, demonstrating the possibility to confine the conductive filler only in the surface layers, while still achieving a component capable of heat dissipation.

TABLE 1 | In-plane and through-plane thermal conductivity values of the 3D printed specimens.

	Through-plane thermal conductivity (W/mK)	In-plane thermal conductivity (W/mK)
30BN	0.36 ± 0.02	0.39 ± 0.03
7BN+8T	0.29 ± 0.01	0.52 ± 0.03
3BN+12T	0.19 ± 0.01	0.67 ± 0.04
3BN+9T +3BN	0.22 ± 0.01	0.66 ± 0.03

TABLE 2 | Mechanical properties for the 3D printed specimens.

	Elastic modulus (MPa)	Tensile strength (MPa)
30T	1430 ± 200	19.5 ± 1.0
3BN+12T	1300 ± 50	18.7 ± 0.7
3BN+9T+3BN	1280 ± 180	20 ± 0.9
30BN	600 ± 250	13 ± 4

In relation to the mechanical properties of the 3D specimens (Table 2), the addition of boron nitride led to a noticeable deterioration of the mechanical properties, with an average elastic modulus value of ~600 MPa and maximum tensile stress of ~13 MPa. Conversely, the addition of talc resulted in modulus and stress values more than double those of 30BN. In the case of the 3BN+12T and 3BN+9T+3BN structures, it is particularly interesting to observe how the mechanical behavior of the layers containing talc predominated, thus allowing for samples with excellent mechanical performance and simultaneously improved thermal conductivity on the surface.

3 | Conclusion

This work has demonstrated the possibility of utilizing PP-based filaments functionalized with BN, to 3D print objects with enhanced thermal conductivity, especially in the in-plane direction. Moreover, it has been demonstrated that, by printing skin-core structures where only the topmost layers contain the conductive filler, while the layers within the core of the specimen contain a reinforcing filler, it is possible to simultaneously achieve good surface thermal conductivity and a significant enhancement of the mechanical properties.

4 | Experimental Section

4.1 | Materials

Polypropylene (PP) ISPLEN PB 170 G2M from Repsol Chemicals (Madrid, Spain) was used. This is a polypropylene-polyethylene random copolymer (density of 905 kg/m^3 and melt flow index of 12 g/10 min (230°C, 2.16 kg)).

As fillers, boron nitride from Sigma–Aldrich (average particle size: ~1 µm) (Darmstadt, Germany) and talc HTP1 grade (mean diameter of 1.9 µm, density of 2.8 g/cm^3), supplied by IMI Fabi Spa (Valmalenco, Italy), were used.

4.2 | Processing

The four PP-based composites, containing respectively 15 wt.% BN (15BN), 20 wt.% BN (20BN), 30 wt.% BN (30BN), and 30 wt.% talc (30T), were prepared through melt compounding using a co-rotating twin-screw extruder LEISTRITZ ZSE 18/40 D (screw rotation speed: 200 rpm, flat temperature profile at 190°C).

Compatible filaments for the 3D printer, with a diameter of 1.75 mm, were produced with a Felfil Evo filament making machine (Torino, Italy).

The 3D printer FlashForge Creator 3 Pro (Zhejiang, China), equipped with a 0.4 mm steel nozzle, was used to 3D print two different types of specimens: disks with a diameter of 25 mm and a thickness of 3 mm for thermal conductivity measurements, and tensile test specimens (ISO-527A-5A) for mechanical tensile testing. In both cases, monomaterial specimens and skin-core structures were created by combining the 30BN and 30T composites, varying the number of layers made with each material (the total number of layers for all samples is 15).

4.3 | Characterization

Frequency sweep tests were performed using an ARES (TA Instruments) rheometer under N_2 atmosphere at 190°C. The thermal properties of the produced materials were assessed using Differential Scanning Calorimetry (DSC) with a DSC Q20 by TA Instruments (New Castle, DE, USA). Scanning electron microscopy EVO 15 by Zeiss was employed for morphological characterization. The thermal conductivity in the in-plane (IP) and through-plane (TP) directions was evaluated with an anisotropic method using the transient plane heat source method (ISO 22007-2:2022) with a TPS 2500S by Hot Disk (Göteborg, Sweden). Tensile tests were carried out using a 5966 Instron dynamometer equipped with a 2 kN cell load.

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Conflicts of Interest

The authors declare no conflicts of interest.

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