Research Article

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Fused deposition modeling of polypropylene-aluminium silicate dihydrate microcomposites

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Abstract: Polypropylene (PP) undergoes fast crystallization and resulting in rigorous shrinkage when it is subjected to high temperature likewise of the fused deposition modeling (FDM) process. This research study focuses on the investigation of the processing parameters and factors that decrease the warpage of PP during the FDM process. Aluminium silicate dihydrate (K) microparticles of different ratios were melt blended with PP by a twin-screw extruder, and filaments of about 1.7 mm diameter were extruded in a single screw extruder. Then, the extruded filaments were used to fabricate the dumbbells structure through the FDM process. The effects of optimizing the fused deposition temperature, coating the chamber with thick papers/fabrics, and coating a printer bed with PP material were also investigated in this study. Scanning and transmission electron microscopy, differential scanning calorimetry, melt flow, and mechanical properties testing instruments are used to analyze the microparticles dispersion, crystallization, flow, and mechanical properties of resulting samples. Uniformly dispersed filler and increased printing chamber temperature result in an increase of crystallization temperature and improve the dimensional accuracy of fused deposited specimens. The fused deposited PP-K10 wt% composite showed an improvement of up to 32% in tensile modulus compared to the neat PP.

Keywords: microcomposites, fused deposition modeling, extrusion, filament, warpage

1 Introduction

The three-dimensional (3D) printing technique is one of the recently introduced methods for the production of lightweight and complex structures of polymers and their composite. In this method, a final product is created by laying successive layers of material. The 3D printed object has the advantage of higher accuracy, flexibility on the product geometry, high-speed production, less material wastage, and no need for an extra tool. Fused deposition modeling (FDM) is among highly emerging 3D printing technologies due to its extensive prototyping capability (1–4). The 3D printing of an object by using the FDM process starts by designing an object with an appropriate software and changing it into a recognizable format for the printer. Then, the filament of appropriate size is fed to the extrusion nozzle where it melts and gets shaped through a 3D printer die. Amorphous polymers such as acrylonitrile butadiene styrene and polylactic acid are the most materials used for the FDM process due to their very low coefficient of thermal expansion and low crystallinity compared to the semicrystalline polymers (5,6).

However, these polymers have drawbacks such as very weak chemical resistance, low toughness, loss of structural integrity and deformation at low temperature particularly if they are subjected to a load, and low mechanical strengths. Thus, it is necessary to develop other FDM printable materials such as polyolefins to overcome those challenges. Polyolefins such as polypropylene (PP) are preferred due to their low cost, varied properties that can be designed for specific applications, resistance to chemicals, humidity, and elevated temperatures, and recyclability (7). This group of polymers also has limited use in high temperatures as it suffers from chain degradation and melts when exposed to direct heat. In addition, the developments of 3D printable polyolefin polymers such as PP continue to be challenging due to their rigorous shrinkage and tendency of warping (2). Fast crystallinity of PP results in the development of

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out-of-plane warpage of the parts due to relief of large thermo-mechanical stresses that lead to failure of the printing process. Limitations of printing the neat PP have been described by a few experimental studies (8–10). The limitation in properties of polymers and their additives can be enhanced by the addition of fillers or by using alternative production methods (11). Fillers such as nanoparticles (12–15) and short glass or carbon fibers (16–18) are incorporated to improve the printability of PP. The clay micro/nanocomposites introduced in PP resulted in a drastic decrease of viscosity and 60% improvement of tensile modulus in a filament for FDM compared to that of the neat PP (12). Shulga et al. (17) used aligned glass fibers as a reinforcing agent in PP and fabricated a composite through a fused filament method of 0.1 mm layer thickness and obtained a tensile strength of 57.4 MPa and modulus of 5.5 GPa. The improvement of adhesion between the printed part and bed substrate is another factor that contributes to the decrease of warping of 3D printed PP and its composites. Bachhar et al. (2) used thin layers called a “brim” at the base of the printing process and polyvinyl acetate glue to fix the printed substrate to the brim, which improves adherence and decreases stresses generated during the 3D printing of PP.

Many kinds of studies have been carried out to obtain optimal parameters of the extrusion process, including varying the inlet and die temperature, speed of the spindle, roller, and puller, which influences the diameter of the resulting filament (19–21). In addition to adding the microfillers into the PP matrix, alteration of the printing process was one of the possible ways to reduce the warping effect and improve the strength of the fused deposited PP composites. During the printing process, the material moves with a nozzle in a rapid heating and cooling system repeatedly, which means it is subjected to diverse and complex temperature conditions (22,23). This results in a growth of controlled crystal and formation of internal stresses that lead to deformations of printed samples (24). To overcome these challenges, several researchers have tried to achieve the homogeneous distribution of temperature by maintaining the temperature in the printing chamber that surrounds the printed part (25–27).

Different researchers studied the effect of single parameters such as varying the composition of fillers, controlling the extrusion temperature, adhesion between printer beds and fused deposited samples, and extrusion speed on the warpage of PP and its composites. However, there are a limited number of studies that combine several factors to optimize the extrusion process of PP and its microcomposites as FDM material. Thus, this study aims to determine the optimal parameters of utilizing PP and its microcomposites as FDM materials through a combination

of several factors. Accordingly, aluminium silicate dihydrate microparticles up to 20 wt% were introduced in the PP matrix to produce filaments suitable for FDM processes for the first time. Also, the printing parameters such as maintaining uniform temperatures in the printing chamber by covering the printer area with thick fabrics and papers, coating the printer bed with PP materials, and optimizing the extrusion temperature were considered in this study to reduce the warping effect.

2 Materials and methods

2.1 Materials

Isotactic PP of high molecular weight in the pellets form (Moplen HP500) was provided by Lyondell Basell Polyolefins (Ferrara, Italy). This material is characterized by a density of $0.9 \text{ g}\cdot\text{cm}^{-3}$ and 1.8 g per 10 min melt flow index (MFI) at 230°C and 2.16 kg. Aluminium silicate dihydrate, $\text{Al}_2\text{O}_3\cdot 2\text{SiO}_2\cdot 2\text{H}_2\text{O}$, which is also known as kaolinite in powder form was manufactured by CADAM S.A. and supplied by the trade name of Paralux (Vale, Brazil). The technical data sheet of the manufacturer reports an average of $0.9 \mu\text{m}$ diameter, $2.6 \text{ g}\cdot\text{cm}^{-3}$ density, and $12 \text{ m}^2\cdot\text{g}^{-1}$ surface areas. In addition, woven fabrics made from polyester fibers with a structure of twill 2/1 and thickness of 1.25 mm, hard papers of double wall and corrugated cardboard manufactured from kraft grade on its outside liner, and a transparent PP adhesive tape of about $30 \mu\text{m}$ thickness manufactured by ABRO Industries Inc. (Savannah, USA) with a brand name of ABRO was used in this study.

2.2 Preparations of sample

2.2.1 Melt blending of samples

Thermo-Haake PolyLab Rheomix (Karlsruhe, Germany), which has an internal mixer of counter-rotating with a speed of 50 rpm, was used to melt compound batches of about 50 g of PP and aluminium silicate dihydrate with 10 and 20 wt% signified with a code of K10 and K20, respectively, at a temperature of 200°C until a plateau of torque curve developed in the mixer. The amount of the microfillers was selected based on the preliminary study and also findings of improved properties at high filler content by Dabrowska et al. (28). The time for the torque plateau, commonly 10–15 min, suggested the filler's leveling

dispersion, and a maximum time limit of 15 min has been chosen to avoid further degradation of the matrix. To compare with the effect of the addition of aluminium silicate dihydrate, similar processing conditions were implemented to neat PP. The resulting sample was granulated using Piovani grinder Model RN 166 (S. Maria di Sala, Venice, Italy).

2.2.2 Extrusion of filaments

Thermo-Haake PTW16 (Karlsruhe, Germany) twin-screw extruder with 16 mm diameter and 3 mm die diameter was fed with ground materials to extrude the filament. During processing, the temperature gradually increased from 150°C to 200°C at a heating rate of 10°C·min⁻¹ from hopper to rod die zone (five heating compartments), respectively. A final filament diameter of 1.7 mm was obtained by optimizing the screw speed in the range of 15–20 rpm and regulating a collection rate at a pressure of 6.0 bars during the extrusion process. A collection rate of 12 m·min⁻¹ was implemented to the extruded filament, and then it wrapped around a 20 cm diameter bobbin through a close follow-up as there is a probability of fry-out due to a large diameter of filaments.

2.2.3 Fused deposition modeling of PP microcomposites

Extruded filaments of about 1.7 mm were fed to a Sharebot Next Generation Desktop 3D printer (Nibionno (LC), Italy) to manufacture 3D printed specimens. A Slic3r software developed by Alessandro Ranellucci (Rome, Italy) was used with chosen parameters such as concentric infill pattern, maximum infill percentage, 0.35 mm nozzle diameter, and 0.2 mm layer height. After a preliminary test, a nozzle temperature and bed temperatures of 240°C, 230°C, and 225°C and 90°C, 80°C, and 75°C for neat PP, K10-PP, and K20-PP, respectively, give an optimum adhesion between layers

and geometry of samples. An increment in the aluminium silicate dihydrate content in PP results in the melt flow increase, and this ensued in the decrease of working temperature, which is also described by Dul et al. (29). All samples are deposited at a constant rate of 20 mm·s⁻¹. As it was mentioned, the immediate crystallization, which results in warpage, is the main challenge of printing PP composites. To reduce this problem, the 3D printing machine was covered with hard papers and thick fabrics to maintain the temperature inside the 3D printer as shown in Figure 1, and also the printing bed was covered by PP adhesive tape to increase the adhesion between printed samples and printer bed.

2.3 Characterization of PP microcomposites

2.3.1 Morphology and microstructure analysis

PP microcomposites were submerged in liquid nitrogen for 60 min and broken apart in a brittle way, and the morphology of the samples was examined through a Carl Zeiss AG field-emission scanning electron microscope (FESEM; Oberkochen, Germany) at an acceleration voltage of 3 kV. To analyze the dispersion and orientation of aluminium silicate dihydrate particles inside the filaments and 3D printed samples, ultramicrotomed cross sections of the samples were observed by using a Philips EM400 transmission electron microscope (TEM; Amsterdam, Netherlands). TEM images were also used to investigate the interfacial adhesion of the components.

2.3.2 Differential scanning calorimetry (DSC)

The thermal properties and crystallinity characteristics of the produced PP microcomposites were measured using Mettler DSC 30 calorimetry (Columbus, OH, USA), under

Figure 1: 3D printer covered with hard paper to maintain the inside temperature.

temperatures that ranged between 0°C and 220°C, at a heating rate of 10°C·min⁻¹ and possessing a constant temperature of 220°C for 5 min under nitrogen gas purge of 100 mL·min⁻¹. Then, the cooling process of samples takes place at the same rate, that is, -10°C·min⁻¹, from 220°C to 0°C and reheated again to the same temperature and heating rate of the first heating.

PP microcomposites degree of crystallinity (χ_c) was calculated by using Eq. 1:

$$\chi_{PP}(\%) = 100 \frac{\Delta H_i}{\Delta H_{PP} \cdot (1 - f)}, \quad (1)$$

where ΔH_i – melting enthalpy, ΔH_{PP} – fully crystalline PP enthalpy, referenced as 207 J·g⁻¹, and f – microfillers weight fraction. The area under the melting peak was used to determine the crystallization enthalpy (ΔH_i).

2.3.3 MFI

LMI 4000 MFI tester (Morgantown, PA, USA) was used to measure the MFI of samples at a temperature of 230°C and 2.16 kg of applied load based on the American Society for Testing and Materials (ASTM) D1238-04c standard.

2.3.4 Tensile tests

The mechanical properties of PP microcomposites were carried out on an electromechanical testing machine, Dynamometer Instron 4502 (Norwood, MA, USA), supplied with a load cell of 100 N at room temperature. The testing of each group of samples was assessed at a crosshead speed of 50 mm·min⁻¹ and an average value of at least three times repeated tests were used to determine the ultimate strength, modulus, and strain at break. The tested samples include as-spun filaments of about 1.7 mm diameter and fused deposited dumbbell of about 2 mm thickness.

Specimen modulus of elasticity (E) was determined from a line that intersects strain values of 0.05% and 0.25% by using Eq. 2, according to ISO 527 standard:

$$E = \frac{\sigma_{0.25\%} - \sigma_{0.05\%}}{\varepsilon_{0.25\%} - \varepsilon_{0.05\%}}. \quad (2)$$

3 Results

3.1 Aluminium silicate dihydrate content optimization

To determine the effective amount of the aluminium silicate dihydrate content in the PP matrix, a preliminary study of

selecting the appropriate concentration of microparticles was carried out. The average values of MFI for melt-blended samples were 1.85 g per 10 min for neat PP and increased to 2 g per 10 min for 10 wt% and 2.95 g per 10 min for 20 wt% of the aluminium silicate dihydrate content. This increment of the MFI with the microfillers content up to 20 wt% could be due to a short microfillers network formation that results in the decrease of microcomposites viscosity. Also, SEM and TEM images show almost a uniform distribution of microfillers up to 20 wt%, whereas there is a tendency of large agglomerates formation above 20 wt% of the aluminium silicate dihydrate content. Accordingly, the mechanical properties such as modulus of elasticity and tensile strength of the aluminium silicate dihydrate filled PP were increased up to 20 wt% and decreased above this value due to the formation of high agglomerates as shown in Figure 3d. Similarly, the strain at break was highly reduced when the amount of aluminium silicate dihydrate microparticles exceeds this amount. Thus, 20 wt% was selected as an optimum content of the filler to be extruded in the 3D printing process. Therefore, the samples investigated in this study are limited to the aluminium silicate dihydrate content of 10 and 20 wt%.

3.2 Extrusion of filaments

Neat PP and PP microcomposites with 10 and 20 wt% of aluminium silicate dihydrate microparticles filaments of apparent draw ratio (ADR) of 1.76 mm were successfully extruded. ADR is calculated as a ratio of the extrusion die cross-sectional area (SD) and filament cross-sectional area (SF) as shown in Eq. 3:

$$ADR = \frac{SD}{SF} \quad (3)$$

So as to avoid the dirt and kept safe, the extruded filaments are reeled on spools of different diameters. Due to the large diameter of the extruded microcomposites, that is, about 1.7 mm, microcomposite filaments were less flexible and more brittle than the neat polymer. Thus, to avoid the fracture of filament during the FDM process, the extruded microcomposite filaments of about 5 m were reeled on the spools of 20 cm diameter relative to 10 cm diameter spools used for neat PP filaments, which were similarly used by Dul et al. (29).

The melt flow test after the filament extrusion process also reveals that the flowability values were increased as the percentage of aluminium silicate dihydrate increased in the PP matrix. The average MFI values of about 2, 2.4,

and 2.5 g per 10 min were obtained for extruded filaments of neat PP, K10-PP, and K20-PP, respectively. As the printing process of neat PP or K-PP microcomposites proceeds, successive layers of melted filaments laid on the 3D printer bed were subjected to a formation of high warping stresses. This is mainly due to the fast crystallization properties of PP and also its poor adhesion with the 3D printer bed as shown in Figure 2a. These results are due to the low surface energy and low adherence properties of PP and its composites to any materials other than itself. In addition to adding fillers, combinations of three other methods were implemented in this study to reduce the warping effect. The first method was coating a printer bed with PP adhesive tape before the commencement of the printing process. Compared to the bare bed, there was a relatively good adhesion between the PP coated printer bed and the first layer of the fused deposited filaments. Figure 2b shows that the PP tape-coated bed helps to maintain certain successive layers of the deposited layers flat on the build plate and results in a decrease of warpage compared to bare bed printed samples.

The second method executed to improve the warping effect was covering the 3D printer opening areas with thick fabrics and hard papers to increase the temperature in the printing chamber to about 65°C from about 40°C

or less temperature initially existing in the chamber. As it was stated by Spoerk et al., an increase in the printing chamber temperature above 60°C results in a significant homogeneous temperature distribution within the printing chamber and promotes annealing during the FDM process (26). Consequently, the amount of internal stresses is reduced, and also the adhesion between the deposited layers is increased, which leads to the improvement of dimensional accuracy of the fused deposited specimens as shown in Figure 2c. The third factor investigated to reduce the warping effect was the application of proper nozzle and bed temperature. During the experimental study, it was observed that an inadequate extrusion temperature leads to low layer adhesion, peeling off of layers, and even nozzle clogging. However, a large thermal gradient is generated in the excessive-high printing temperatures that alter the layer height and desirable geometry of specimens. After a preliminary test, the optimum temperatures of the nozzle were set to 240°C, 230°C, and 225°C, whereas the printer bed temperatures were set to 90°C, 80°C, and 75°C for neat PP, K10-PP, and K20-PP, respectively, owing to the PP composites melt flow increase with the aluminium silicate dihydrate content. The samples produced on a bare bed and open chamber show a tendency of high warpage formation as shown in Figure 2a and b. However, Figure 2d shows that the

Figure 2: Photos of FDM samples of $\times 1.5$: (a) neat PP on bare bed, (b) K10-PP on PP tape coated bed and open chamber, (c) K10-PP in coated chamber, and (d) K10-PP on coated printer bed, coated chamber, optimized nozzle, and printer bed temperatures.

Figure 3: FESEM micrographs of extruded filaments: (a) neat PP $\times 5,000$, (b) K10-PP $\times 20,000$, (c) K20-PP $\times 20,000$, and (d) K30-PP $\times 20,000$. FDM printed specimens of: (e) K10-PP $\times 120$ and (f) K20-PP $\times 100$.

combination of fillers inclusion, coating printer bed and chamber, and optimizing the fused deposition temperature leads to the production of the specimen with improved adherence of one layer to the next layer, compacted in all directions, and optimally reduced warpage.

3.3 Morphological and microstructural analysis

FESEM pictures of the fracture surface of filaments of neat PP, K10-PP, K20-PP, and K30-PP are shown in Figure 3a–d,

respectively, whereas Figure 3e and f shows 3D printed dumbbells of K10-PP and K20-PP, respectively.

FESEM micrographs of filaments show almost a uniform distribution of aluminium silicate dihydrate, even though there was a tendency of small agglomerates formation in K20-PP composites. This indicates that there is a relatively good dispersion of aluminium silicate dihydrate microparticles in the PP matrix. The agglomerates formed in 20 wt% of microfillers lead to the reduction of mechanical properties. Figure 3d shows that there was a formation of large agglomerates formation at 30 wt% of microfillers, which makes further processing of the composite difficult. As it can be observed from 3D printed

Figure 4: TEM micrographs of (a) K10-PP and (b) K20-PP fused deposited specimens.

samples cross sections shown in Figure 3e and f, the shape of individual filaments varied from the circular section of the nozzle. This is mainly due to the pressure applied between the bed and nozzle, which results in a slight reduction of orientation during the printing process. By taking the estimated ratio between the original sections of the filaments for the 3D printing of about 1.7 mm and section of 3D printed microfilaments of about 0.35 mm, a draw ratio of 4.86 was obtained without considering the expansion of filaments after it leaves the nozzle. As mechanical properties are directly related to the orientation of polymer chains, they will be improved along the laying direction of 3D printed specimens.

The micrographs of 3D printed dumbbells also show that there is a formation of triangular cavities between the deposition layers due to lack of continuity. Also, fast crystallization and poor adhesion between layers of PP – aluminium silicate dihydrate microparticles – may result in an occurrence of fracture surface between certain neighboring layers as shown in Figure 3e and f. However, most contiguous filaments were completely fused as single layers, apart from small cavities formed between layers. The processing temperature mainly affects the quality of contiguous filaments bonding through determining a deposition time, the interaction, and inter-joining between 3D printed layers.

TEM images (Figure 4) show that microfillers are almost uniformly dispersed inside the PP matrix with a certain formation of agglomerates mainly in 20 wt% of aluminium silicate dihydrates (Figure 4b). TEM micrographs also confirmed the existence of physical interfacial adhesion of the components.

3.4 Thermal properties

DSC thermogram, shown in Figure 5, reveals the effect of the aluminium silicate dihydrate content on the crystallization temperature and melting temperature of 3D printed samples

of PP microcomposites. As the percentage of aluminium silicate dihydrate increased in the microcomposites, the crystallization temperatures also increased, whereas the melting temperatures almost remained the same in all 3D printed samples of neat PP, K10-HP, and K20-HP. The second heating cycle of filaments displayed a higher crystallinity content due to an increase of nucleation activities during the cooling cycle after the first cycle melting. The melting temperature (T_m) of all composition on the second melting is almost the same at about 165°C. However, the crystallization temperature (T_c) varies based on the microparticles content. Accordingly, the crystallization temperature of about 112°C for neat PP increased to about 115°C for K10-PP and further improved to about 120°C for K20-PP. The degree of crystallinity, χ_c , calculated from the melting enthalpy curve according to Eq. 1 gives almost similar values of about 47% for neat PP, K10-PP, and K20-PP fused deposited specimens (Table 1).

This indicates that aluminium silicate dihydrate microparticles initiate the mobility of the molecular chains of PP by acting as a heterogeneous nucleation center through lowering the free energy barrier for crystallites formation as reported in the studies of Guessoum et al. (30) and Luyt et al. (31). As the crystallization speed of PP is faster than the extruding process, the introduced microparticles in PP increase the crystallization temperature, which contributes to the reduction of the warping effect. Comparative analysis of thermal properties of fused deposited specimens and the extruded filaments shows that there is an improvement of about 2% crystallinity content in the fused deposited specimens. It is assumed that the obtained improvements in the crystallinity content could be due to the effect of an increase in the printing chamber temperature.

3.5 Mechanical properties

The introduction of aluminium silicate dihydrate microparticles of different contents in the PP matrix affects the

Figure 5: DSC graph of fused deposited samples of neat PP, PP-K10, and PP-K20 subjected to heating–cooling–heating cycle between 0°C and 220°C under a N_2 atmosphere at a heating/cooling rate of $10^\circ\text{C}\cdot\text{min}^{-1}$.

mechanical properties such as the modulus of elasticity, ultimate strength, and strain at break of the resulted microcomposites. Three specimens' tests were used to determine the average value of mechanical properties of fused deposited dumbbells of neat PP, K10-PP, and K20-PP. Figure 6a–c shows that the introduction of aluminium silicate dihydrate microparticles in the PP matrix improves the elastic modulus, slightly decreases the tensile strength, and reduces the strain at break of the microcomposites. Figure 6a shows that the elastic modulus of extruded filaments for 3D printing is higher than that of 3D-printed dumbbells other than the case of K10-PP composites. This result is mainly due to the low adhesion between layers and relatively high compaction pressure during the filament extrusion process compared to the FDM process. The presence of voids between fused deposited layers also contributed to the reduction of elastic modulus yield.

However, maintaining a high temperature in the printing chamber helps to improve the balance between

the crystallization speed and the fused deposition process. This leads to an increase in the dimensional accuracy and compactness of the produced specimens. Increasing the printing chamber temperature and coating a printer bed with PP materials improved the tensile strength of fused deposited specimens by about 30% for K10-PP compared to neat PP (Figure 6b). However, the tensile strength of fused deposited samples of K20-PP is lower than that of K10-PP due to a relatively large aluminium silicate dihydrate agglomerates formation as it is observed in the microstructural analysis. As it is discussed in the Section 3.3, the drop in mechanical properties could also result from a low functional group interaction between aluminium silicate dihydrate and PP matrix.

Deformation at a break is decreased with the addition of aluminium silicate dihydrate in the PP matrix. Figure 6c shows that the deformation at break of 64.4% for neat PP decreased to 38.5% for K10-PP extruded filaments. Similarly, the strain at break FDM printed samples decreased

Table 1: Summary of DSC analysis: first and second melting, crystallization temperature, and crystallinity content of fused deposited neat PP, K10-PP and K20-PP samples

Sample composition	First melting temp.		Crystallization temp.		Second melting temp.		Crystallinity content (%)	
	Peak (°C)	Integral ($\text{J}\cdot\text{g}^{-1}$)	Peak (°C)	Integral ($\text{J}\cdot\text{g}^{-1}$)	Peak (°C)	Integral ($\text{J}\cdot\text{g}^{-1}$)	First melting	After cooling
Neat-PP	166.9	94.1	112.3	96.9	165.1	97.2	45.4	47.0
K10-PP	168.1	81.2	114.8	86.9	165.2	86.9	43.6	46.6
K20-PP	168.3	71.2	119.9	79.6	165.0	78.4	43.0	47.3

Figure 6: Comparison of mechanical properties of FDM printed and extruded filaments: (a) tensile modulus, (b) tensile strength, and (c) strain at break of neat PP, K10-PP, and K20-PP.

from 57% for neat PP to 35.5% for K10-PP. The reduction in the deformation with the addition of microfillers could be due to the lower bonding strength between the layers, local stress concentration caused by microparticles, and also the brittleness properties of aluminium silicate dihydrate. However, the deformation at the break for K20-PP is higher than that of K10-PP in both filaments and 3D printed samples, which needs further investigation.

4 Discussion

The rising of the surrounding temperature in the printing chamber through covering the 3D printer open spaces with thick fabrics and papers helps to prevent warpage and improve the strength of the fused deposited samples. The finding of others researchers (26,27,30) also confirms

that higher printing chamber temperature leads to fewer occurrences of temperature fluctuation and slower cool-down of the printed samples, which results in less residual stresses and a lower degree of shrinkage. The mathematical model proposed by Wang et al. (32), similarly, approves that the higher temperature of printing chambers results in a reduced amount of warpage. Thus, the higher chamber temperature maintained in this study, about 65°C, is comparable to the findings of Ferrer-Balas et al. (33). They found that a higher printing chamber temperature (above 60°C) significantly improves the morphology, crystallinity, size, and homogeneity of the crystalline regions for PP and its microcomposites.

In addition to maintaining the printing chamber temperature, the extrusion nozzle temperature has a direct impact on the produced samples. Unnecessarily high temperatures will lead to the formation of large thermal gradients that can alter the layer height, shape, and

desirable geometry of fused deposited samples (34,35). To minimize this challenge, the optimum extrusion temperature is selected to raise the average temperature between layers, which improves the interlayer adhesion. Thus, the printing temperatures have strong correlations with the mechanical properties of printed samples. An increase in the adhesion between successive layers results in an increase of the elastic modulus and tensile strength, which is also similarly described by the findings of different researchers (36–39).

Coating of the printing bed surfaces by PP tapes highly influences the adhesion of the first layer of the printed sample, which is similar to the findings described by Spoerk et al. (9). The existence of a good distribution of aluminium silicate dihydrate microparticles confirmed by FESEM micrographs, the high temperature maintained in the printing chamber by thick fabrics and papers, and printing bed coated by PP tapes highly reduces the shrinkage deformation, which in turn results in good mechanical properties of FDM printed samples of PP composites. The comparative analysis of the tensile strength of FDM printed samples of neat PP in this study is about 14% higher than the results of almost similar studies by Dong et al. (27). This could be the combined effect of the adjusted process parameters and factors such as increasing the printing chamber temperature, coating of printer bed with PP materials, and optimizing the nozzle temperature during the fused deposition process.

This study proved that PP and its composites can be one of the potential promising materials for the FDM process. The FDM printed samples of PP and its composites can be utilized for various applications that include chemically resistant laboratory equipment for semiconductors processing, an alternative for electrolysis of water, ankle-foot orthoses, and cranial bone substitution as it shows similar strength, inertness, no degradability, and nonmagnetic properties of the bone.

5 Conclusion

PP was successfully melt blended with aluminium silicate dihydrate microparticles of up to 20 wt%, and this was used to extrude filaments of about 1.7 mm diameter that was used to feed the FDM process. As the content of the microparticles increased in PP, the melt flow of the resulted microcomposites also increased, and this, in turn, resulted in the decrease of the FDM processing temperature to avoid the alteration of layer height and desirable geometry of the fused deposited specimens. The

incorporation of aluminium silicate dihydrate microparticles initiated the mobility of the molecular chains and improved the crystallization temperature to 120°C for K20-PP from 112°C of neat PP with almost similar crystallinity content of about 47% after the cooling cycle of the DSC test. The morphological study of fused deposited samples shows almost a uniform distribution and the existence of physical interfacial adhesion up to 20 wt% of fillers, whereas the high agglomerates formation preceded beyond this amount. In this study, the warpage, the main challenge encountered during the fused deposition of PP, is decreased through the combined effect of added fillers, maintained printing chamber temperature, printer bed coated with PP tape, and optimized the fused deposition temperature to increase the fused deposited part performance. The optimization of the printing chamber, crystallization, and melting temperature helps to increase the dimensional accuracy and compactness of the produced specimens by improving the difference in temperature between the crystallization speed and the fused deposition process. The mechanical properties of fused deposited specimens show an improvement of tensile modulus from about 370 to 490 Mpa and tensile strengths from about 19 to 25 MPa and reduction in the deformation at break from about 57% to 36% for neat PP and K10-PP, respectively. In general, the fused deposited samples show lower mechanical strength compared to the extruded filaments due to the vacant space and poor adhesion between fused deposited layers.

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