

Review Article

Enhancement of the Mechanical and Thermal Properties of Polypropylene and Nylon 6.6 Composite Blends

Mohammad Yousef Al-Haik,¹ Saud Hamad Aldajah,¹ Mohammad Mazedul Kabir,¹ Waseem Ahmad Siddique,² Yousef Haik,³ and Md Mainul Islam⁶

¹Department of Mechanical Engineering, Higher Colleges of Technology, Abu Dhabi 25035, UAE ²Applied Engineering Department, Applied Technology High School, Abu Dhabi 111499, UAE ³Academic Affairs, University of Sharjah, Sharjah 27272, UAE ⁴Centre for Future Materials and School of Engineering, University of Southern Queensland, Toowoomba, QLD 4350, Australia

Correspondence should be addressed to Md Mainul Islam; mainul.islam@usq.edu.au

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The mechanical and thermal properties of nylon 6.6 on polypropylene (PP) blends were investigated. Five weight percentages of nylon 6.6, 1 wt%—5 wt%, were blended with PP by an injection molding process to produce PP/nylon 6.6 blends. The impact of the mixture on the mechanical properties was investigated by performing a three-point bend test to determine the flexural strength and hardness test using nanoindentation. Both differential scanning calorimetry and thermogravimetric analysis were used to study the thermal properties of the mixture. Chemical and morphological testing was performed by Fourier-transform infrared spectroscopy and scanning electron microscope tests. Mechanical testing revealed improved strength, modulus, and hardness of the PP/nylon 6.6 blends. The 2 wt% blend showed elastic modulus, tensile, and yield stress enhancement. TGA showed variable results with improved thermal resistance for the 3 wt% and 4 wt% blends, whereas there was no effect for the 2 wt% blend and reduced thermal resistance for the 1 wt% and 5 wt% blends. DSC analysis revealed no influence on the melting temperature of the blends. SEM images showed a homogeneous mixture between PP and nylon 6.6, which substantiated the interfacial adhesion between PP and nylon 6.6 and was verified by FTIR.

1. Introduction

Polypropylene (PP) is one of the most commonly used commercial polymers due to its low cost, high moisture resistance, and ease of processing. Moreover, nylon 6, also known as polyamide 6, has also attracted good attention in research due to its high mechanical properties and good oxygen transport resistance [1]. PP and nylon 6.6 blends have received much attention in the last decade due to their enhanced properties [2, 3]. The blend was selected to study the material contribution to the enhancement of the overall properties and alterations of the morphology of the blend. The major limitation of polymers and nylon 6 is high oxygen permeability and hygroscopy, respectively. The melting point of nylon 6 is higher than that of PP; therefore, the processing temperature of nylon 6 must be higher than that of polypropylene [1]. Such polymeric mixtures are considered an excellent technique for producing polymeric products that hold enhanced properties of the sourced polymer. The economic and viable methods to manufacture a multiphase polymeric system are based on the physical and chemical interaction needed in the interface between polymeric materials. The challenge in producing multiphase polymeric systems is the interfacial tension that prevents the smooth stress transfer between the polymer and matrix. High interfacial tension can be reduced by reducing the interfacial area, which is achieved by the increased size of dispersed particles. One solution to overcome this challenge is introducing a compatibilizer that creates a bond between the matrix and the dispersed phased-in polymeric material. The use of nanofillers to prepare nanocomposites opens various engineering opportunities in nanocomposite technology in terms of modifying the polymer matrix. Adding finite quantities of nanofillers increases the performance of polymers in terms of mechanical, thermal, and chemical properties. Furthermore, PP/nylon 6.6 blend morphology may be altered [4–7]. The interpenetrating polymer network structure is found in polymeric material morphology, which is closely related to block polymers [8, 9]. PP/nylon 6.6 can create an interpenetrating polymer network (IPN) by blending the two polymers in a network in which one of the systems synthesizes in the presence of another. A physically cross-linked network is formed when one of the polymer chains is entangled with another polymer chain.

In addition, recycling is another significant advantage for these multiphase polymeric materials [10]. The application of polymer blending is to dilute the polymer with a low-cost commodity polymer and focus on recycling industrial plastic waste [11, 12]. It was found that most of the polymeric materials exhibited two-phase morphology, namely, the matrix and dispersed phases. The matrix phase plays a significant role in morphology, while the dispersed phase is minor in the form of spheres, fibrils, or platelets [13]. The various types of morphological structures in polymeric materials are based on multiple factors such as interfacial adhesion, blending ratios, shear stress between the matrix phase and minor phases, processing conditions, and temperature [13, 14].

Abraham et al. [15] studied the mechanical behavior of composite materials consisting of two different sizes (large and small diameters) of waste polyamide (nylon 6.6) fibers as reinforced materials with polypropylene. They tested with two forms of PP matrix, first as pure PP and the other as maleic anhydride- (MA-) grafted PP (MA-g-PP). Grafting MA-g-PP showed great results in flexural strength [16, 17]. They were both used in the preparation of composite materials. The composite materials consisting of thin polyamide fibers showed improved strength and modulus of elasticity than pure PP. Moreover, the MA-g-PP-blended composite with polyamide fibers showed better mechanical properties than the two compositions.

The grafting phenomenon has shown promising strength and flexural properties in recent years. Monomers such as glycidyl methacrylate (GMA) were used to graft polyolefins [18, 19]. Because the nylon structure has both $-NH_2$ and -COOH groups, the epoxy group can respond with the basic or acidic groups and be a good functionalization alternative [20]. Another study by Huber et al. [21] investigated the impact and tensile strengths between two different types of compatibilized blends of nylon and polypropylene. They grafted the PP polymer with maleic anhydride (MA-g-PP) at two different grades of maleation: low 2% and high 7%. MAg-PP played the role of a stress distributor, which lowered the interfacial tension between the two immiscible phases in the polymer blend. Their results indicated an increase of up to 25% in tensile strength. Moreover, the 2% maleation compatibilized blend showed enhancements in the impact properties, whereas the higher maleation grade depicted a negative effect on PP/nylon on impact properties.

Polymer adhesion is essential for polymer processing, lamination, polymer blends, and composites, making it the subject of an investigation by many researchers [22, 23]. Polymer groups are generally thermodynamically immiscible, where fracture toughness is below the acceptable limit. To have good mechanical properties, it is necessary to have smooth stress transfer between polymers at the interface. Increasing the interfacial area by connecting polymeric chains is one of the methods to have good reinforcements that are performed by using random or block copolymers [24–26]. Polymer incompatibility can be minimized by using a reactive compatibilizer that reacts between the two polymers at the interface to improve interfacial adhesion properties [27–32].

In this work, PP/nylon 6.6 blends were prepared through an injection molding process to study the effect of different percentage concentrations of nylon 6.6 on PP. This study focuses on the impact of adding various weight percentage concentrations of nylon 6.6 on the enhancement of mechanical and thermal properties of PP. The process consisted of blends 1 wt%—5 wt% of nylon 6.6 with PP pellets, which were heated in injection molding at 190°C for 15 minutes to form composite plates. The mechanical, thermal, chemical, and morphological tests were performed on the samples to determine the characterization of polymers. The blends will be referred to as sample "#16" in the following sections.

2. Materials and Sample Preparation

Polypropylene (PP), a melt flow index (MFI) of 1.59 g-min^{-1} and density of 0.91 g·cm⁻³, was purchased from Exeed EFF Co., Oman. Nylon 6.6 was provided by local polymer industries in the United Arab Emirates. The preparation of PP/ nylon 6.6 mixtures was achieved in a twin-screw extruder at nylon 6.6 concentrations of 1, 2, 3, 4, and 5 wt%. Thereafter, the obtained compositions were processed by using an injection-molding machine (Norwood Instrument Ltd) to form the blend samples referred to as sample "#16." The cylinder and the mold temperatures of the molding machine were set at 190°C and 50°C, respectively. The compositions of the samples are listed in Table 1.

3. Experimental Procedure

3.1. Three-Point Bend Test. Three-point bending tests of the produced samples were conducted using a universal testing machine following ASTM D790 test standards. The specimens (length 100 mm × width 12 mm × thickness 3 mm) were simply supported and tested under 3-point loading with the span set at approximately 16 times the thickness of the specimen. The extensometer recorded the midspan deflection at a crosshead speed of $10 \text{ mm} \cdot \text{min}^{-1}$. Five specimens were tested for each set of the samples, and the average was considered to be the representative value.

3.2. Melt Flow Index. The melt flow index of the PP/nylon 6.6 blend thermoplastic composite was performed with a Dynisco melt flow indexer, as per ASTM D1238, to evaluate the processability of the composite. The flow rate of

TABLE 1: Designations and compositions of the studied PP/nylon6.6 blends.

Comple designation	Sample compositions (wt%)		
Sample designation	PP (%)	Nylon 6.6 (%)	
Pure PP	100	0	
1% #16 (1T)	99	1	
2% #16 (2T)	98	2	
3% #16 (3T)	97	3	
4% #16 (4T)	96	4	
5% #16 (5T)	95	5	

the composite is expressed in grams of the extruded material in 10 minutes with a load of 2.16 kg at a certain temperature [33]. MFI normally represents an inverse measurement of melt viscosity which means that composite flowability is increased with the increasing MFI values. The melt flow index (MFI) values of each category of #16 and the pure PP samples represent the average multiple measurements (three replica). The MFI ratio for each category is calculated by dividing its average MFI value by that of the pure PP MFI:

$$MFI ratio = \frac{avg. value of MFI for indi vidu al #16 sample}{avg. value of MFI for pure PP sample}.$$
(1)

3.3. Nanoindentation. A NanoTest (Micro Materials Ltd, UK) indentation instrument was used to perform the hardness test. The nanoindentation experiment consisted of performing ten indents on the sample from which the equipment software generates a load-displacement curve used to calculate the hardness of the considered specimen. The indents were conducted at a displacement rate of $0.0167 \text{ nm} \cdot \text{s}^{-1}$ until a maximum displacement of 1827 nm was attained. To avoid creep, a constant 1 mN load was held for 30 s in order not to affect the unloading behavior.

3.4. Differential Scanning Calorimetry (DSC). DSC analysis of polymeric membranes was performed using a DSC Q200 V24.4 Build 116 Model (TA Instruments, Waters LLC) thermal analyzer containing a refrigerated cooling system.

In a typical experiment, each sample (5-8 mg) was placed in a hermetically sealed aluminum pan and heated from 40°C to 200°C at a heating rate of 10°C·min⁻¹. The experiments were conducted under a nitrogen environment that allowed efficient heat transfer and removed any volatile matters from the samples. An empty hermetically sealed aluminum pan was used as a reference cell placed inside the furnace next to the filled pan. The software "TA Universal Analysis 2000 V4.5A Build 4.5.05 (TA Instruments, Waters LLC)" was used to analyze the results. The results of each sample represent the average data of three experiments.

3.5. *Thermogravimetric Analysis (TGA).* TGA analysis was carried out by using a thermal gravimetric analyzer Q50 (TA Instruments, Waters LLC) to determine the weight decomposition percentage. Similar to the DSC experiments,

samples of about 5–8 mg of PP and #16 samples were used for analyzing their thermal decompositions. The reported results are the average data obtained from three samples. The samples were heated in the temperature range of 25 to 700°C at a heating rate of 10° C·min⁻¹ under the nitrogen environment. The TA Universal Analysis 2000 V4.5A Build 4.5.05 (TA Instruments, Waters LLC) software was employed to analyze the results.

3.6. Fourier-Transform Infrared Spectroscopy (FTIR). Attenuated total reflectance FTIR spectroscopy (ATR-FTIR) analyses of pure PP and #16 samples were carried out using Nicolet Nexus 470 FTIR (Thermo Scientific Madison, WI, USA). The films of PP and #16 samples were ground into powder form and mixed with potassium bromide (KBr) powder before being placed inside the spectrophotometer. The FTIR spectra were recorded at wavenumbers from 0 to 4000 cm^{-1} . The FTIR transmission code analyzed the molecular fingerprint generated from the samples.

3.7. Scanning Electron Microscopy (SEM). Parts from pure PP and #16 samples retrieved from mechanical testing were used for morphological texture investigation. These were first dried at 50°C under vacuum for 24 h, and then, their broken surfaces were coated with gold by sputtering before being examined through a JEOL JSM-5600 scanning electron microscope (Jeol Ltd., Tokyo, Japan).

4. Results and Discussion

4.1. Mechanical Properties

4.1.1. Three-Point Bend Test. The peak and yield stresses of pure PP and the addition of nylon 6.6 on PP (#16 samples) are depicted in Figure 1. The peak and yield stresses for the pure PP samples were 31.8 MPa and 25.5 MPa [16, 17], respectively. It was noticed that only 2 wt% #16 samples showed an increase in the peak stress compared with the pure PP sample with a value of 35.5 MPa, corresponding to an 11.6% increase. However, for the yield stress, the 1 wt%, 2 wt%, and 4 wt% #16 samples showed an enhancement in mechanical properties compared to pure PP, with values of 26.7 MPa, 28.5 MPa, and 25.8 MPa, indicating 4.6%, 11.8%, and 1.3% increases, respectively. Adding nylon 6.6 to the PP polymer blend contributes to good compatibility and adhesion within the blend matrix. It was also noticed that, when 5 wt% of nylon 6.6 was added to PP, both peak stress and yield stress degraded below the pure PP sample with values of 24.3 MPa and 22.5 MPa, indicating a 23.5% and 11.8% decrease, respectively. This degradation is attributed to the weak chemical bonding between nylon 6.6 and the polypropylene matrix at high weight percentages. At a higher percentage of nylon, incompatibility of binary blends revealed the weak adhesion between nylon and pp matrix, thus resulting in weak tensile strength. The SEM micrograph also shows agglomeration at a high percentage of nylon, leading to poor mechanical properties.

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FIGURE 1: Peak and yield stresses for pure PP and #16 samples.

Figure 2 shows the elastic (flexural) modulus of pure PP and the addition of nylon 6.6 on PP (#16 samples). The elastic modulus of pure PP was found to be 684.84 MPa. The addition of nylon 6.6 on PP increased slightly for flexural modulus except for the 5 wt% sample. The 2 wt% #16 sample showed the highest value of 860.3 MPa, indicating a 25.6% increase compared with pure PP. The improved strength is attributed to the better interaction between the two dispersed phases of nylon 6.6 and PP. Mechanical entanglement and secondary bonding are the two most commonly possible interactions between the two phases. Higher entanglement creates more resistance to slide each other and thus leads to better mechanical properties. The 1 wt% #16 sample showed an increase in the modulus with a value of 797.8 MPa, resulting in a 16.5% increase compared with pure PP. The flexural modulus of 3 wt% #16 was 788.4 MPa, which similarly showed a slight increase compared to pure PP, about 15.1% higher. The 4 wt% samples had a modulus value of 804.1 MPa, giving a 17.4% increase compared with pure PP. However, the 5 wt% #16 sample showed a slight decrease in the flexural modulus with a value of 682.6 MPa, which is about 0.33% less than that of pure PP. A higher value of flexural modulus of blended materials indicates higher resistance to deformation, which increases the stiffness of the composite samples. The improved strength, especially in the 2 wt% #16 sample, is attributed to the better interaction between the two dispersed phases which can also be verified by the FTIR results.

4.1.2. Nanohardness Test. Hardness was calculated for each sample tested by performing ten indents from the loaddisplacement data obtained from nanoindentation. Figure 3 depicts the hardness values of the #16 samples compared with those of pure PP. All #16 samples showed a significant increase in hardness compared to the PP sample. The pure PP hardness value was found to be 133 MPa. However, the 1% #16 sample was enhanced by 43.6% compared to pure PP, with a hardness value of 191 MPa. It was also noticed that adding 2 wt% and 3 wt% nylon 6.6 on PP increased hardness compared to 1%. 2 wt% and 3 wt% had a hardness value of 172.4 MPa and



FIGURE 2: Elastic modulus for pure PP and #16 samples.

146.6 MPa, indicating an increase of 29.6% and 10.2%, respectively, compared to pure PP.

Moreover, 4 wt% showed turnover in hardness compared to the latter two (2 wt% and 3 wt%), with a hardness value of 187.1 MPa, resulting in a 40.7% increase compared to pure PP. The 5 wt% #16 sample reduced hardness with a value of 150.5 MPa. As nylon 6.6 is added to PP, the hardness values increase, as shown in Figure 3. Although all #16 samples increased dramatically compared to pure PP, 3 wt% and 5 wt% showed the lowest hardness compared to the other samples. The increase in hardness is attributed to the interlocking and good adhesion between the chains of the nylon 6.6 structure.

Moreover, the orientation effect where the reinforcing phase is directed towards the load indicated the bonding between PP and nylon chains. Similar results were noticed in the three-point bend tests. Table 2 summarizes the threepoint bend and hardness tests and the melt flow index ratios concerning pure PP for all samples.

The melt flow index is an important method to express the flow characteristics of the composite blend which helps know the processibility of the polymer blend. As indicated in Table 2, the MFI ratio, which is calculated by dividing the mean MFI value by that of the pure PP MFI, increases with increasing the percentage of nylon 6.6 except for the 4 wt% #16 sample. As observed, the maximum value of the MFI ratio is observed in the case of the 5 wt% #16 sample. Therefore, the high melt flow index value at 5 wt% nylon 6.6 indicated ease of processing as well as melting. However, the composite blends with a high MFI ratio can have low average molecular weight and low viscosity. It was also noticed that samples (3 wt% and 5 wt%) that have a high MFI ratio revealed poor mechanical properties due to poor intermolecular interactions. In other words, a low MFI ratio of the sample (1%, 2%, and 4%) has better mechanical properties.

4.2. Thermal Analysis

4.2.1. Differential Scanning Calorimetry (DSC). Figure 4 shows the DSC graphs obtained for the #16 samples and pure PP samples, which depict crystallization behavior and endothermic heat flow. Single peaks were exhibited for all the samples. Table 3 tabulates the melting temperature for all



FIGURE 3: Hardness values for pure PP and #16 samples.

TABLE 2: Summary of three-point bending and hardness tests for pure PP and #16 samples.

Specimen names	Peak stress (MPa)	Yield stress (MPa)	Modulus (MPa)	Hardness (MPA)	MFI ratio
Pure PP	31.8	25.5	684.8	133.0	1.0
1% #16	31.4	26.7	797.8	191.0	0.95
2% #16	35.5	28.5	860.3	172.4	0.98
3% #16	25.9	25.1	788.4	146.6	1.15
4% #16	29.4	25.8	804.1	187.1	0.83
5% #16	24.3	22.5	682.6	150.5	1.31

samples, where PP is reported to have a melting temperature (T_m) of 166.11°C. The thermal characteristics of the melting temperature (T_m) , heat of fusion (H_{Fus}) , degree of crystallinity $(D_c\%)$, and crystallinity temperature (T_c) of pure PP and #16 samples are summarized in Table 3.

Figure 4 shows no noticeable changes in the melting temperature and crystallinity. However, all #16 samples showed the heat of fusion and degree of crystallinity close to or below the pure PP sample except for the 3% #16 sample. The 3 wt% sample exhibited a higher degree of crystallinity, 47.8%, and the heat of fusion exceeding the pure PP sample. This is attributed to the nylon powder, which acts as a filler that behaves as a nucleating agent for the polymeric matrix to promote crystallization [17]. The degree of crystallinity is calculated using $D_c \% = H_{\text{Fus}}/H_{\text{Fus,100\%}} \times 100\%$, where the heat of fusion for 100% crystalline PP polymer crystals is 207 J/g [34].

4.2.2. Thermogravimetric Analysis (TGA). Figure 5 shows the TGA graphs that reveal the thermal resistance of the pure PP and #16 samples. All samples were tested under a nitrogen atmosphere. As the temperature was increased, all the samples revealed a single decomposition stage, as depicted in Figure 5. PP/nylon 6.6 blends have shown improvements in thermal resistance with an increased amount of nylon 6.6 with 2 wt%, 3 wt%, and 4 wt%. The initial decomposition temperature of pure PP was found to be 363.5°C. It is observed that the temperature corresponding to 5% weight loss of the 4 wt% PP/nylon 6.6 blend is higher than that of pure PP by about 18°C due to better interaction, and this temperature is further increased in the case of the 4 wt% nylon 6.6 blend. This implies that the thermal resistance of the blend increases with the 4 wt% nylon sample showing the highest resistance. However, in the case of 5 wt% nylon, the thermal decomposition of the blend has shown poor resistance due to strong phase separation between nylon 6.6 and PP, deteriorating the compatibility of the blend. This reduction in thermal resistance could be due to the nonuniform dispersion of nylon 6.6 on the blended samples. Furthermore, it can be noticed from Figure 5 that none of the samples showed a noticeable multistage or sharp initial decomposition as tabulated in Table 4.

4.3. Fourier-Transform Infrared Spectroscopy. The FTIR spectra of pure PP and #16 blended samples are depicted in Figure 6. The #16 samples all showed an absorption peak with less intensity than pure PP at a wavelength of 1640 cm⁻¹, which belongs to the amide group. It can be observed that the pure PP sample does not show a peak at this absorption level. However, when blended with nylon 6.6, the absorption peak was evident. The absorption peaks at 1035 cm⁻¹ and 1242 cm⁻¹ are assigned to symmetric and asymmetric C-O-C stretching, indicating the interaction between the PP skeleton and nylon 6.6. Moreover, a peak at 2928 cm⁻¹ belongs to the -CH3 group, and another peak at 2854°cm⁻¹ belongs to C-H stretching. The amide group's absorption peak has indicated the PP skeleton's functionalization, which leads to the interaction of PP and #16 samples.

4.4. Scanning Electron Microscopy (SEM). Figure 7 shows the SEM images of the pure PP and #16 samples. It was noticed that none of the #16 samples showed clusters of nylon 6.6 scattered on PP, thus indicating a homogeneous mixture between nylon 6.6 and PP. Figure 7(b) has shown better compatibility between the two blend polymers due to a significant reduction of coalescence of nylon 6.6 particle in



TABLE 3: Calorimetric parameters and the degree of crystallinity of PP and #16 samples.

Specimen designation	Melting temp. (°C)	Heat of fusion $(\Delta H_{\rm FUS})$ (J/g)	Crystallisation temp. (°C)	Degree of crystallinity (<i>D_c</i> %)
РР	166.1	94.8	118.0	45.8
1% #16	166.3	94.7	117.0	45.8
2% #16	164.3	71.7	117.0	34.6
3% #16	166.3	98.9	118.0	47.8
4% #16	165.1	75.6	118.0	36.5
5% #16	165.2	84.4	117.0	40.8



FIGURE 5: TGA graph for pure PP and #16 samples.

TABLE 4: Results for TGA analys	is.
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Specimen names	5% loss (°C)	50% loss (°C)
PP	363.5	436.5
1% #16	323.5	393.5
2% #16	363.5	443.5
3% #16	381.0	452.0
4% #16	409.0	456.0
5% #16	287.5	369.0



FIGURE 6: FTIR spectra of pure PP and #16 samples.





FIGURE 7: Continued.

(d)



FIGURE 7: SEM images of (a) pure PP, (b) 1 wt%, (c) 2 wt%, (d) 3 wt%, (e) 4 wt%, and (f) 5 wt% #16 samples.

the PP matrix and better adhesion that leads to better mechanical properties in the case of the 2 wt% # 16 sample. Figure 7(f), for the 5% #16 sample, shows a few noticeable micropores, causing the possibility of accumulation of slight agglomeration in the micropore region. This explains why the 5 wt% # 16 sample showed poor mechanical properties in flexural strength; however, it had a higher hardness level. Figures 7(c) and 7(d), for the 2 wt% and 3 wt% #16 samples, show fibrillar structures that have developed due to the addition of nylon 6.6, which could have caused the plastic deformation to occur during fracture. With all these flaws, all #16 samples showed an enhancement in the mechanical properties compared to pure PP. The images also highlight that uniform dispersion decreases when the percentage concentration of nylon 6.6 increases, specifically in the 5 wt% #16 sample. Enhancing the mechanical strength of the PP polymer is lowered due to the poor dispersion and low compatibility between the #16 samples as nylon 6.6 percentage concentrations are increased, which can be seen from the tensile strength graphs.

5. Conclusion

The mechanical and thermal properties of polypropylene were investigated with various weight percentage concentrations of nylon 6.6. PP with 2 wt% nylon 6.6 showed the highest enhancement in the mechanical properties of the blends. Introducing small percentages of nylon 6.6 onto PP alters the chemical structure, and as a result, the mechanical properties are increased. It was reported that the strength, modulus, and hardness of the PP structure increased by 11.6%, 25.6%, and 29.6%, respectively, for the 2 wt% sample. The DSC results revealed stable thermal resistance in terms of the melting temperature. TGA results showed a single decomposition phase for all samples and however showed variable weight loss degradation. FTIR results verified the absorption of amide groups that indicated interlinking between nylon 6.6 and PP. FTIR also revealed the absorption of symmetric and asymmetric C-O-C stretching consistent with the interaction of nylon 6.6 and PP. SEM images depicted a homogeneous mixture between PP and nylon 6.6. Small micropores were noticed in the 5 wt% #16 sample,

indicating that little accumulated agglomeration of nylon 6.6 is possible within the micropores. However, most samples showed good compatibility and transparency compared to pure PP. Overall, the mechanical and thermal properties were positively impacted due to the addition of nylon 6.6.

Data Availability

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

Conflicts of Interest

The authors declare that there are no conflicts of interest regarding the publication of this paper.

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