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Indian
Journal of Advances in
Chemical Science

Indian Journal of Advances in Chemical Science 4(1) (2016) 68-76

Influence of Fine Particles on the Mechanical Behavior of Short Glass Fiber Reinforced Thermoplastics Blends

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Received 16th October 2015; Revised 23rd November 2015; Accepted 24th November 2015

ABSTRACT

The present investigation is on the role of fine particles on the mechanical behavior of short glass fiber (SGF) reinforced thermoplastic blends. Thermoplastic blends such as polyamide66 and polytetrafluoroethylene (PA66/PTFE) and polymethylmethacrylate and PTFE (PMMA/PTFE) in 80/20 wt. % were selected for the study. They were reinforced with SGF and filled with micro fillers such as short carbon fibers, alumina (Al_2O_3), and silicon carbide of different geometric shapes to form the mixture. These composites were prepared by melt mixing method using twin screw extruder followed by injection molding. The mechanical properties such as tensile strength, flexural strength, impact strength including hardness of the blend composites were studied as per ASTM standards. Results revealed that addition of SGF into PA66/PTFE and PMMA/PTFE blends exhibited good tensile and flexural strength obviously, but the different shape micro fillers exhibits a synergic effect on the tensile and flexure properties of PA66 and PMMA based composites, respectively, except flexural modulus. PMMA/PTFE based microcomposites showed better tensile and flexural properties than PA66/PTFE blend microcomposites. PMMA/PTFE based composites showed moderate elongation at break and better impact strength after the filler addition into PMMA/PTFE blend composites. The moderate change in hardness was observed for both microcomposites.

Key words: *Polyamide66/polytetrafluoroethylene, Short glass fibers, Fillers, Polymethylmethacrylate/polytetrafluoroethylene, Blends.*

1. INTRODUCTION

Polymer and their composites are finding ever increasing usage for numerous industrial applications such as bearing material, rollers, seals, gears, cams, wheels, and clutches. In these applications, the material having both strong mechanical strengths and tribo performance can only suits the situation. Polymer and their composites are finding ever increasing usage for numerous industrial applications such as bearing material, rollers, seals, gears, cams, wheels, and clutches. [1]. The use of polymers and polymer-based composites which are having a combination of good mechanical and tribological properties can only prove themselves as worthy. It is often found that such properties are not attainable with a homopolymer. This had led to the development of copolymers, fiber-reinforced polymer, and polymer blending. One among these is polymer blending which seems to be fascinating because it has simple processing and unfolds unlimited possibilities of producing materials with variable properties. Glass fibers and the carbon fibers are the reinforcement agents most used in the thermoplastics based composites, as they have good balancing properties.

Polyamide66 (PA66) is a semi-crystalline, thermoplastic commodity polymer that finds widespread use in applications that require considerable strength but low toughness. It is a widely used engineering thermo plastic. It possesses an outstanding combination of properties such as low density, easy processing, good strength, and solvent resistance. Polytetrafluoroethylene (PTFE) is a linear polymer with high crystallinity, strong, stiff, and tough engineering material with lower coefficient of friction. It has excellent thermal stability. It is flexible and can be used over a wide range of temperatures, 2500°C down to almost zero, and still retains its nature of being attacked by any reagent or solvent. It has been known for quite some time that PTFE polymer

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exhibits a self-lubricating behavior and its application in sliding prevents stick–slip motion instabilities. A lot of research has been made to improve the mechanical properties by means of incorporation of PTFE with various neat polymers/fillers, such as fibers, fine particles, and whiskers. Polymethylmethacrylate (PMMA) is a polymer having amorphous structure. They are stiff, strong material with outstanding weather resistance. Due to their transparency and weather resistance, they were used for light fittings, canopies and lenses of car lights, signs, and name plates.

Polymeric composites filled with inorganic fillers or reinforced with fibers are the most common engineering materials today. Incorporating fillers and/or fibers to the base polymer materials provides substantial improvement in terms of the mechanical properties [2,3]. Attempts to understand the modifications in the mechanical behavior of the polymers with the addition of fillers or fibers reinforcements have been made by many researchers. Ravikumar *et al*. [4] reported the effect of particulate fillers on the mechanical behavior of short carbon fiber (SCF) reinforced PA/polypropylene (PP) nanocomposites. They reported that mechanical properties were improved by adding nanoclay into the SCF reinforced PA66/PP blend composites. Palabiyak and Bahadur [5] studied the mechanical behavior of PA6 and high-density polyethylene (HDPE) polyblends with and without compatibilizer. Zhao *et al.* [6] studied the water absorptivity and mechanical behavior of PTFE /PA6 and PTFE/PA66 blends. Addition of PTFE results in reduction of mechanical properties and Water absorbed blends had the improved mechanical properties. It was found that the tensile strength of the polyblends increased when PA proportion was more than 20 wt.% and hardness decreased with any PA proportion. Experimental investigation on the effect of glass fibers on the mechanical properties of PP and PA6 plastics were reported by Gullu *et al*. [7]. They concluded that the PP and PA6 plastics reinforced with glass fibers exhibited better improved mechanical strength. Exploring the effect of 7.5, 15, 22.5, and 30 wt.% of PTFE on the mechanical properties of polyetheretherketone (PEEK)/PTFE in PEEK/PTFE blend was made by Bijwe *et al*. [8]. They found that addition of 30 wt.% of PTFE showed the maximum impact strength and other properties were deteriorated. Chen *et al*. [9] studied the friction and wear mechanisms of polyamide66/HDPE blends .They concluded that the blend with 70vol. % PA66 has the best mechanical properties. Chiang and Huang [10] focused their study on the various properties of the blends of polyoxymethylene (POM) with up to 20 wt.% chemically surface treated PTFE (CPTFE) and compared with those of POM/PTFE blends. They inferred that the mechanical properties of POM/PTFE blends decrease with increasing PTFE content, but the tensile strength and the young's modulus of POM/CPTFE blends are double that of POM/PTFE blends. Effect of adding red mud and silicon carbide (SiC) into SGF reinforced PEEK was reported by Naga Mahendra Babu [11]. They showed that the addition of these fillers into the short glass fiber (SGF) reinforced PEEK improves the tensile strength, flexural strength, and the flexural modulus of the composites. Recently, it has been observed that by incorporating filler particles into the fiber matrix of fiber reinforced composites, synergistic effects may be achieved in the form of higher modulus and reduced material costs, yet accompanied with decreased strength and impact toughness [12,13]. PP hybrid effects of reinforced long glass fibers (LGF) and particulate filler were studied by Hartikainen *et al*. [14]. They studied the effect of LGF reinforced PP filled with $CaCo_3$. Addition of LGF into PP improved the tensile strength and fracture toughness appreciably, but the decrease in the same properties was observed by filling $CaCo_3$ into SGF filled PP composites. Jian and Tao [15] investigated the mechanical properties of polyphenylenesulphide (PPS)/SCF composites and PA6 filled PPS/SCF composites. They showed that better flexural strength was obtained for the 25 wt.% of SCF in PPS. Furthermore, they proved that the addition of 6 wt.% of SCF into the PA66 exhibited the better flexural behavior than SCF filled PPS composites. Cao *et al*. [16] reported the effect of basalt fiber in ultra-high molecular weight polyethylene. Increase in basalt content in the composite led to decrease in toughness and increase in strength, hardness, and creep resistance. Yuan *et al*. [17] studied the effect of coupling agent on mechanical properties of glass fiber reinforced SCF filled HDPE composites. They showed that increasing coupling agent will improve the bonding strength between glass fibers and the matrix. They proved that the coupling agent will act positively in improving the mechanical behavior of SGF reinforced SCF/HDPE composites. Kumar *et al*. [18] studied the effect of banana fiber reinforced HDPE/PA66 blend composites. They revealed that treated banana fibers had good effect on the mechanical properties of HDPE/PA66 blend composites. Wacharawichananat and Siripattanasak [19] studied the mechanical properties of PP and POM blends. They showed that decrease in impact strength, tensile strength, and young's modulus for PP/POM blends with increase of PP content up to 30 wt.% and an increase in mechanical properties starts after the inclusion of PP above 30 wt.% into PP/PA66 blend composites. Fua *et al*. [20] studied the tensile properties of SGF and SCF reinforced PP composites. The results about the composite strength and modulus were interpreted using the modified rule of mixtures equations by introducing two fiber efficiency factors, respectively, for the composite strength and modulus. It was found that for both types of composites the fiber efficiency factors decreased with increasing fiber volume fraction and the more brittle fiber namely carbon fiber corresponded to the lower fiber efficiency factors than glass fiber. Chen *et al*. [21-23] systematically studied the mechanical and tribological properties of PA66/PPS blend filled with SGF/SCF fibers and inclusion of PTFE particles. They showed that 20-30 vol. % glass fiber greatly increased the mechanical properties of PA66/PPS blend. SCF reinforcement resulted in improved tensile strength, flexural strength, and hardness by 80%, 92%, and 21%, respectively. Further, addition of PTFE particles is beneficial from friction and wear behavior point of view and deteriorated the mechanical properties. The effect of PTFE filler and fiber reinforcement on the mechanical properties of 80/20 blend of PA6/HDPE was studied by Palabiyik and Bahadur [24]. They showed that the reinforcement of 5-15% of SGF to polyblend improved the tensile strength from 20% to 60%, respectively. Stuart [25] has recently published the review article on various fiber and particulate filled polyblends. Hemalatha and Ramesh [26] studied the tensile properties of natural fiber reinforced epoxy composites. They showed that the weight percentage of fiber reinforcement affects the tensile property of the material. Hybridization of fibers had the greater effect than the manofibers' behavior. Hemanth *et al*. [27] studied the effect of fibers and fillers on thermoplastic composites. They showed that POM based composites exhibited better tensile strength and flexural strength than thermoplastic copolymers. PTFE is one of the most important and promising materials to improve fracture toughness of polymer based composites. In spite of the fact that polymer composites are used in such structural applications, no data are reported on the influence of Teflon in PA as blend with SGF and other inorganic particulate fillers *viz*. SCF, SiC, and alumina (Al_2O_3). Keeping this in view, a series of PA66/Teflon and PMMA/PTFE blends with fibers and ceramic fillers were investigated for tensile, flexure, and impact properties. Efforts were also made to study the role of SGF/SCF fiber and ceramic fillers inclusion on the relevant strength properties.

2. EXPERIMENTAL

2.1. Materials

The materials used in the present investigation such as PA66, PMMA, PTFE, glass fiber, carbon fiber, SiC, and Al_2O_3 fillers are listed in Table 1. The details of materials and their source are also tabulated in the Table 1.

2.2. Fabrication of Blends and their Microcomposites

The polymers PA66 and PTFE with proper proportions were dried at 80°C for 48 h before mixing to avoid plasticization, hydrolyzing effects from humidity, and to obtain the sufficient homogeneity. The saline-coated sized SGFs were mixed in proper proportion into the thermoplastic materials. The materials are mixed and the mixture was extruded using Barbender Co-rotating twin screw Extruder, GLS Polymers, Bangalore (Make: CMEI, Model: 16CME, SPL, chamber size 70 cm^3). The extruder consists of five heating zones where the temperature maintained in these zones of the extruder barrel were Zone 1 (220°C), Zone 2 (235°C), Zone 3 (240°C), Zone 4 (265°C), and Zone 5 (270°C), and the temperature at the die was set at 220°C. The extruder screw speed was set at 100 rpm to yield a feed rate of 5 kg/h. The extrudates obtained was in the form of cylindrical rod which was quenched in cold water and then palletized using palletizing machine. During initial stage, around 1-1.5 kg of initial extrudate was removed to get the pure blend and to remove impurities of extrudate of the previous stroke of the extrusion. Before injection molding, all polymer blended composite pallets were dried at 100°C in vacuum oven for 24 h. All test specimens were injection molded from the pelletized polyblend material obtained from co-rotating extruder. The temperature maintained in the two zones of the barrel was Zone 1 (265°C) and Zone 2 (290°C) and mold temperature was maintained at 65°C. The screw speed was set at 10-15 rpm followed by 700-800 bar injection pressure. The injection time, cooling time, and ejection time maintained during injection molding were 10, 35, and 2 s, respectively. All the molded specimens as per ASTM were inspected and tested visually and those found defects were discarded for the testing (Figures 1 and 2).

2.3. Measurement of Mechanical Properties

The mechanical properties such as tensile strength, flexural strength, impact strength along with density, and hardness of the blends were measured as per

Table 1: Data and the source of the materials used in this study.

Material	Designation	Form	Size (μm)	Trade name	Manufacturer	Density (g/cc)
Polyamide 66	PA66	Granules	-	Zytel 101L NC010	DuPont Co. Ltd	1.14
Polytetrafluoroethylene	PTFE	Particles	12	MP 1000	DuPont Co. Ltd.	2.16
Polymethylmethacrylate	PMMA	Granules		-	DuPont Co. Ltd.	
Short glass fiber	SGF	Cylindrical	12-14	-	Fine Organics, Mumbai	2.5
Short carbon fiber	SCF	Cylindrical	6-8	-	Fine Organics, Mumbai	1.74
Silicon carbide	SiC	Irregular	5-10	-	Carborundum India Ltd.	3.21
Aluminum oxide	Al_2O_3	Particles	5-10	-	Aldrich, Bengaluru	3.95

Table 2: Formulations of composite blend PA66/PTFE and microcomposites in weight percentage.

Composition	Material ID	Weight percentage					
		PA66	PTFE	SGF	SCF	SiC	Al_2O_3
PA66/PTFE	1P	80	20	-	-	-	-
Blend (PA66/PTFE)/SGF	2P	80	20	20	-	-	-
Blend (PA66/PTFE)/SGF/SCF/SiC/Al_2O_3	3P	80	20	15	2.5	6.25	6.25

PA66=Polyamide 66, PTFE=Polytetrafluoroethylene, SGF=Short glass fiber, SCF=Short carbon fiber, SiC=Silicon carbide, Al_2O_3=Aluminum oxide

Table 3: Formulations of composite blend PMMA/PTFE and microcomposites in weight percentage.

Composition	Material ID	Weight percentage					
		PA66	PTFE	SGF	SCF	SiC	Al_2O_3
PMMA/PTFE	1M	80	20	-	-	-	-
Blend (PMMA/PTFE)/SGF	2M	80	20	20	-	-	-
Blend (PMMA/PTFE)/SGF/SCF/SiC/Al_2O_3	3M	80	20	15	2.5	6.25	6.25

PA66=Polyamide 66, PTFE=Polytetrafluoroethylene, SGF=Short glass fiber, SCF=Short carbon fiber, SiC=Silicon carbide, Al_2O_3=Aluminum oxide

ASTM. The tensile strength and the tensile elongation at break were measured using Universal testing machine (JJ Lloyd, London, United Kingdom, capacity 1-20 KN) in accordance with ASTM D 638. Tests were performed at constant strain rate of 5 mm/min. ASTM D 638 Type 1 standard dimensions are used. Flexural strength or three point bending were carried out on the same machine by changing the jaws of the setup and the specimen acts as simply supported beam subjected to point load at the middle. The flexural strength and flexural modulus were determined at the rate of 2 mm/min as per ASTM D790. The standard specimen dimensions for the flexural strength is 125 mm × 12.7 mm × 3.2 mm. The Izod impact strength was determined using ASTM D 256 using Izod impact testing machine at the striking rate of 3.2 mm/s. The densities of the blend composites were determined as per ASTM D756. The ASTM standards for these mechanical testing is shown in the Figure 3. All these tests were conducted at the room temperature. Minimum of three samples were tested for the data representation. On the other hand, the density and the hardness (Shore D) of the blended composites were determined as per ASTM D792 and ASTM D224, respectively (Tables 2 and 3).

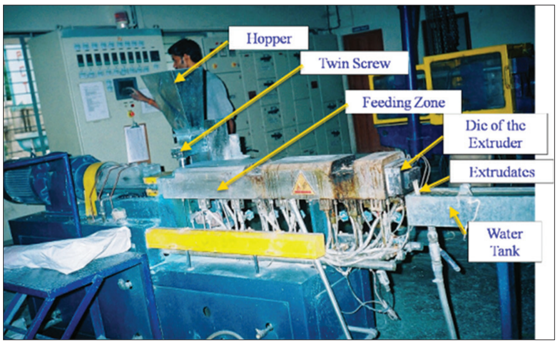

Figure 1: Brabender co-rotating twin screw extruder.

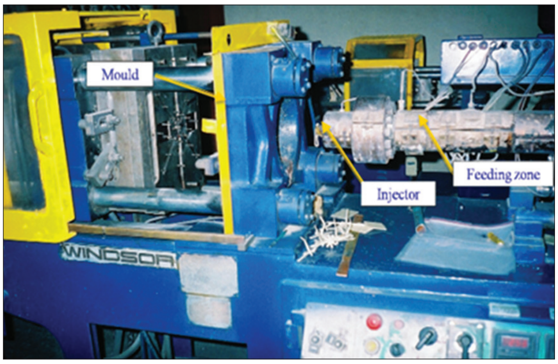

Figure 2: Injection molding machine.

3. RESULTS AND DISCUSSIONS

3.1. Density and Hardness of PA66 and PMMA Based Composites

The variation of density and hardness (Shore D) of PA66/PTFE and PMMA/PTFE blend and their microcomposites were plotted in Figure 4 a and b. The density of PA66/PTFE blend was increased after the SGF reinforcement into the blend. Further inclusion of fine particulates of SiC, Al_2O_3, and SCF into the SGF reinforced blend led to increase the density of the blend. This can be due to the presence of dense glass fibers. Addition of fine particles of SiC, Al_2O_3, and SCF into the blend improved the density of the SGF reinforced PA66/PTFE blends. This may be due to the nature of these hard particles. The similar observations were made for PMMA/PTFE blends. The effect of inclusion of PTFE into PEEK-PTFE blend increased the density of the blend PEEK/PTFE [8]. The fine particulates filled SGF reinforced blends

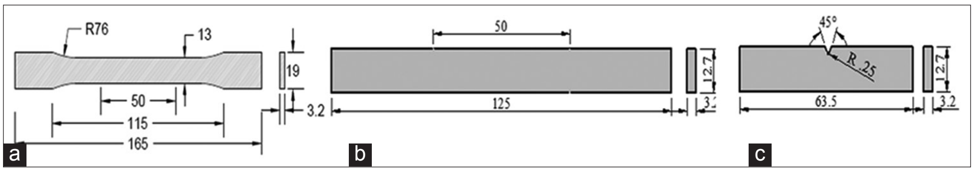

Figure 3: Specimen standards: (a) ASTM D 638 (tensile test) (b) ASTM D790 (flexure test) and (c) ASTM D256 (impact test).

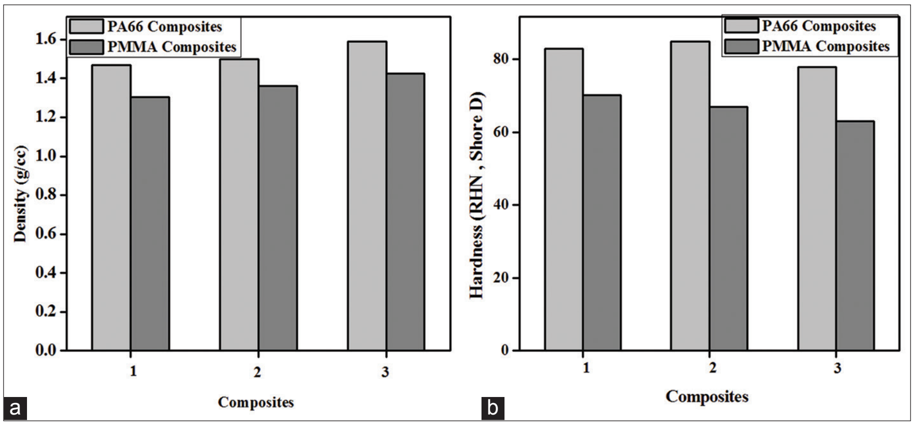

Figure 4: Variation of properties of polyamide 66/polytetrafluoroethylene (PTFE) and polymethylmethacrylate/PTFE micro composites: (a) Density and (b) hardness.

propelled toward the brittleness which may help in the development of cracks easily. Among the studied composites, PA66 based composites had appreciable density. The hardness of PA66/PTFE and PMMA/PTFE based composites was presented in the Figure 4b. The hardness of PA66/PTFE blend increased to a little extent due to the effect of fiber loading. This shows that the silane treated SGF had better interfacial bonding with the thermoplastics. However, the addition of fillers into SGF filled composites decreased the hardness of PA66/PTFE/SGF composites. However, appreciable change in the hardness was not observed for PA66/PTFE blends after SGF reinforcement, whereas decrease in hardness was observed after the inclusion of hard particles of SiC, Al_2O_3, and SCF into the blend. Similar observations were found in PMMA/PTFE based composites. This might be due to the fact that addition of fillers made the material brittle and appreciable change in the hardness value was not observed in both the blend-based composites. However, PA66/PTFE based composites had better hardness when compared with PMMA/PTFE based composites. The brittle nature of the blends acts as supporters for the stress raisers to develop affinity toward the crack growth of the composites. This could be the reason for the good impact strength of the PMMA based composites.

3.2. Effect of Fine Particles on the Tensile Behavior of SGF Reinforced Thermoplastic Blends

The variation of tensile strength and their associated properties such as tensile modulus, elongation due to tension of PA66/PTFE and PMMA/PTFE, and their microcomposites as a function of fiber and filler loading were shown in the Figure 5a-c, respectively. The tensile strength of pure PA66/PTFE blend was 46.5 N/mm^2. After the SGF reinforcement, it was 75.36 which is 62% increase over neat blend. After the addition of fine particles of SiC, Al_2O_3, and SCF, it was 44.29 which is 5% less than that of the neat blend and 41% decrease against strength of SGF filled PA66/PTFE blend composites. Similar observations were made with PMMA/PTFE blend and their microcomposites. About 51% improvement by reinforcing SGF into PMMA/PTFE blend and 29% decrease after the filler addition into SGF reinforced PMMA/PTFE blend composites. This shows that addition of SGF improved the tensile strength of both the blends and filler inclusion decreased the tensile strength of the fiber reinforced blend microcomposites. This improvement may be due to the slenderness ratio of SGF and also the improved adhesion between the fiber and matrix. The interfacial bond between these two associates, SGF, and blends were seems to be better to improve the tensile strength. After the filler addition, decreasing trend can be attributed to the brittle nature of the blends. The different shapes and angular edges of the filler had caused non-uniform adhesion between the associates of the blend microcomposites, in turn results in stress concentration in the polymer composites. Among the studied composites, PMMA/PTFE based composites seems to be more brittle when compared with that of PA66/PTFE composites. This may be due to the brittle

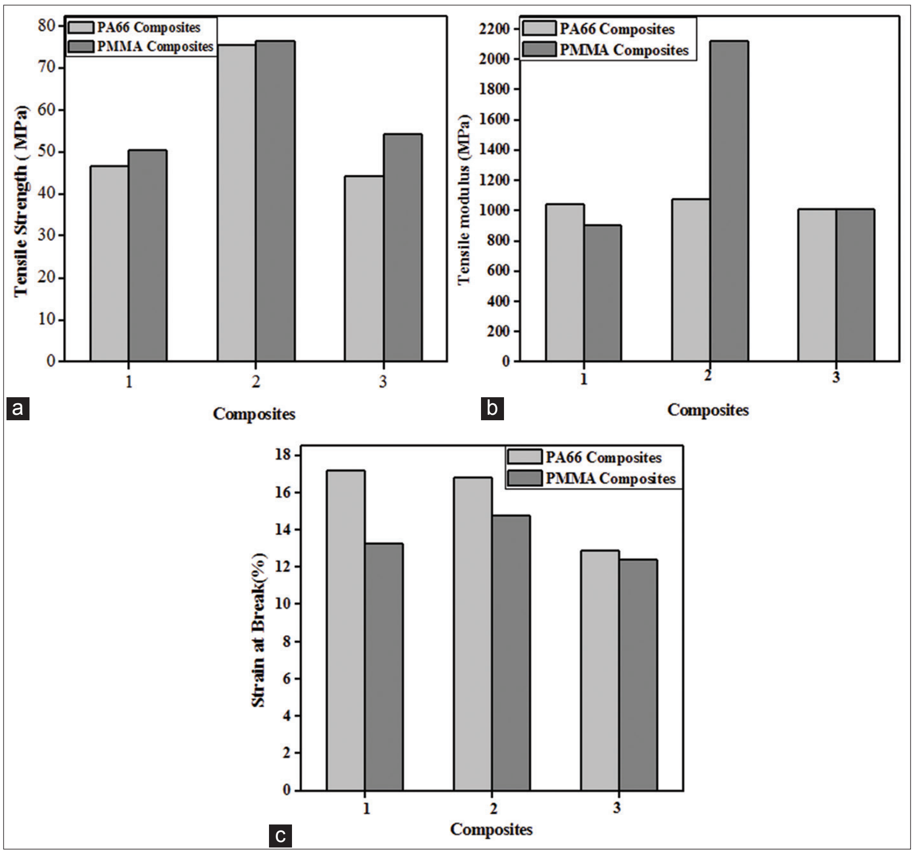

Figure 5: Variation of properties of polyamide 66/polytetrafluoroethylene (PTFE) and polymethylmethacrylate/PTFE micro composites: (a) Tensile strength, (b) tensile modulus, and (c) elongation at break.

nature of PMMA. There was no much change in the tensile modulus between the composites of PA66/PTFE blend. However, the tensile modulus of SGF reinforced PMMA/PTFE blend was almost 3 times that of neat blend. This can be attributed to the good adhesion between the SGF and neat blend when compared to PA66/PTFE blend. However, almost same tensile modulus was observed between filler added blend and the neat blend PMMA/PTFE. The elongation due to break remained almost unchanged for SGF reinforced PA66/PTFE blend, but decreased after the filler addition. SGF reinforcement into PMMA/PTFE blend improves the elongation by 11%. This can be attributed to the fiber fracture. Strain at break was decreased due to filler addition. This may due to the brittle nature of the material behavior. However, PMMA/PTFE showed better tensile properties than PA66/PTFE composites.

3.3. Effect of Fine Particles on the Flexural Behavior of SGF Reinforced Thermoplastic Blends

The flexural strength is the strength of the material during bending. The flexural behavior of PA66/PTFE and PMMA/PTFE and their microcomposites was studied. The flexural strength of PA66/PTFE blend is 78.62 N/mm^2. After reinforcing 20 wt.% SGF, it was 109.4 N/mm^2, which is 39% increase over the neat blend. However, the addition of fine particles into SGF filled PA66/PTFE decreased the flexural strength by 26%. On the other hand, 37% increase due to SGF and 28% decrease due to fine particles on flexural strength was observed for PMMA/PTFE blends, respectively. The improved properties of the studied polyblends and their microcomposites can be attributed to uniform distribution of SGF and strong adhesion of polymer blends to the SGF. The applied load penetrates the matrix and transforms it across the surface of the fibers instead of breaking it. This may cause fiber rupture. Improved flexural strength and flexural modulus of the thermoplastics were revealed by many researchers [14]. The composites PA66/PTFE and PMMA/PTFE with SGF filled and also microparticle-filled SGF reinforced polymer blends exhibited better flexural modulus. The stresses during the load transfer had been shared by these fillers to avoid more deflection. PMMA/PTFE microcomposites showed better flexural properties than PA66/PTFE microcomposites.

3.4. Impact Strength of PA66 and PMMA Based Blend Composites

The variation of impact strength of PA66/PTFE and PMMA/PTFE and their microcomposites was shown in Figure 6. The effect of SGF reinforcement on PA66/PTFE blend led the material to decrease in

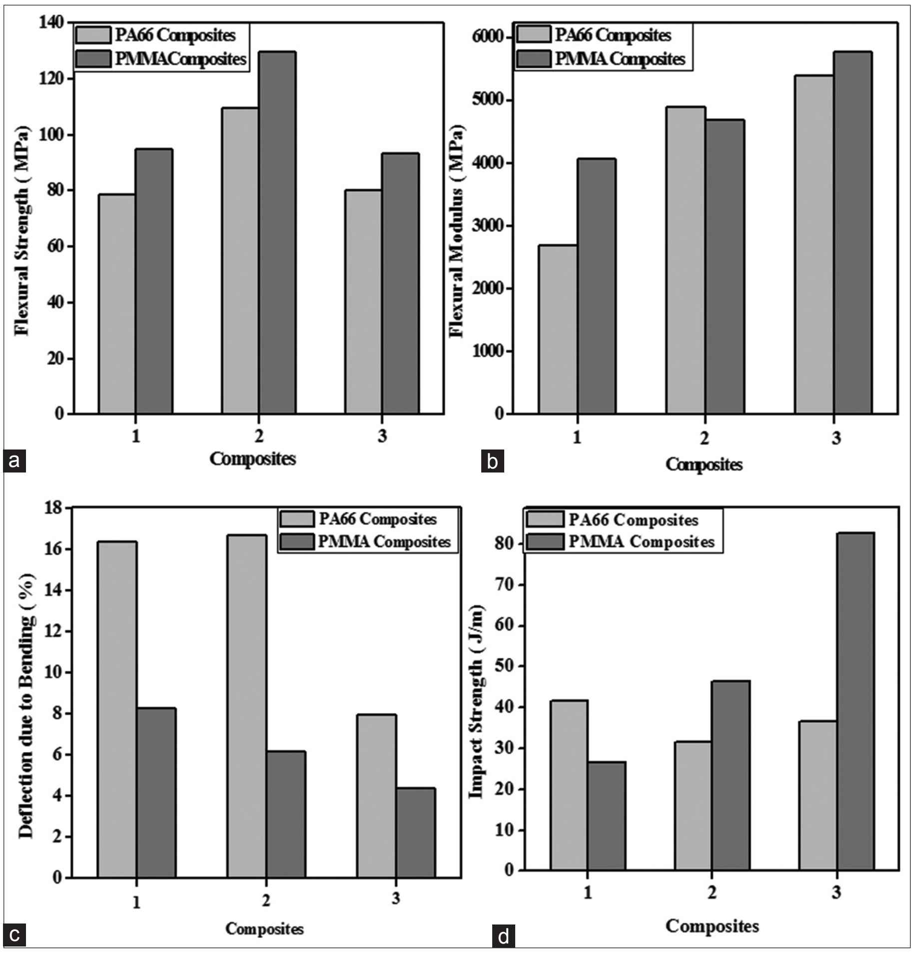

Figure 6: Variation of properties of polyamide 66/ polytetrafluoroethylene (PTFE) and polymethylmethacrylate/ PTFE micro composites: (a) Flexural strength, (b) flexural modulus, (c) deflection due to bending, and (d) impact strength.

impact strength by 23% over a neat blend. However, the effect of fine particles inclusion into the blend, improved the impact strength of the same composites. This may due to the synergic effect of micro fillers. PMMA/PTFE blend exhibited good impact strength which was 75% more than the neat blend after SGF reinforcement. The impact strength of PMMA/PTFE blend is almost three times than that of the neat blend after adding fine particles of SiC, Al_2O_3, and SCF. This may be due to the addition of fine particulates into the SGF reinforced blend led the material to become brittle. However, SGF reinforced blend further uphold the ductile nature of the composites. This may require more impact energy to break the material. However, PMMA/PTFE blends and their composites had better impact strength than PA66/PTFE blend composites.

4. CONCLUSION

1. SGF reinforcement into PA66/PTFE and PMMA/PTFE polyblend exhibited better tensile and flexural properties
2. Fine particles of SCF, Al_2O_3, and SiC decreased the tensile and flexural properties of SGF reinforced studied polymer blends except the flexural modulus
3. The impact strength of the SGF reinforced PA66/PTFE blend was less when compared with fine particles effect on the same. On the other hand, PMMA/PTFE based microcomposites exhibited better impact strength
4. The density of the studied composites increased linearly due to dense SGF and fillers
5. The hardness of PA66/PTFE blend was almost constant even after SGF reinforcement, but decrease in hardness was observed due to the effect of addition of inorganic particles. PMMA/PTFE blends and their microcomposites followed the same behavior in terms of hardness.

5. ACKNOWLEDGMENTS

The author thanks Mr. Ganesh M, Managing Director of GLS Polymers, Bengaluru, India, for providing fabrication facilities. Also expresses heartfelt thanks to Dr. K C Krishna, Principal, Government Engineering College, K.R.Pet-571426, Karnataka, India.

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