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Influence of particle size and particle size distribution on MICA filled nylon 6 composite

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Particulate reinforced thermoplastic composites are designed to improve the properties and to lower the overall cost of engineering plastics. In this study the effects of particle size and particle size distribution on the properties of mica filled nylon-6 was investigated. Composites of nylon-6 with varying concentrations (viz. 5 to 40 wt%) of mica were prepared by twin screw extrusion. The composite showed improved mechanical, thermal as well dielectric properties on addition of filler. © *2005 Springer Science + Business Media, Inc.*

1. Introduction

Particle filled polymer composites have become attractive because of their wide applications. Incorporating inorganic mineral fillers into plastic resin improves various physical properties of the materials such as mechanical strength, modulus and heat distortion temperature. In general the mechanical properties of particulate filled polymer composites depend strongly on size, shape and distribution of filler particles in the matrix polymer and good adhesion at the interface surface. Flakes or platelets represent a special class of reinforcing fillers for thermoplastics and thermosets [1]. Mica is one such type of filler and is particularly abundant mineral. By virtue of its crystal habit, it may be easily cleaved into thin flakes by ordinary grinding methods and with special care, may be delaminated into very thin flakes. Such flakes when suitably aligned in a plastic matrix, may impart a relatively high degree of reinforcement in terms of strength and stiffness [1]. Nylons are one of the most widely used engineering thermoplastics such as in automobile, electrical, electronic, packaging, textiles and consumer applications because of their excellent mechanical properties [2–7]. However limitations in mechanical properties, the low heat distortion temperature, high water absorption and dimension instability of pure nylons has prevented their applications to structural components. Hence numerous efforts have been undertaken to use nylons as matrix resins for composite by adding inorganic fillers such as glass beads [3, 4, 8–10], kaolin [3, 4, 11, 12], aluminatrihydrate [12], clays [12], silica [12], flyash [13], wollastonite [3, 4, 12, 14], mica [15–17], talc [3, 4, 12, 18], calcium carbonate [12, 19], whiskers [20–22] etc. In this investigation, mica of different particle size was added to nylon-6 polymer. The influence of the addition of the filler on mechanical, heat distortion temperature, dielectric, rheological and morphological behaviour was examined.

2. Materials and method

The matrix material nylon-6 (density 1160 kg/m^3) was obtained from M/s Nirlon India ltd. (Mumbai). Filler additives like dispersing agent (Finnawax SS, used 1.5 wt%) and antioxidant (Irgafos 168 and Irganox 1076, used 1 wt% each) were obtained from M/s Fine Organics (Mumbai) India and CIBA Speciality Chemicals (Mumbai) India respectively. Fillers were added to nylon-6 in 5, 10, 20, 25, 30, 35, 40% w/w ratios. The suppliers and the physical properties of the filler are listed in Table I. Nylon-6 and mica were predried at 80°C for 7 hours prior to compounding. The composite granules were prepared by using twin-screw extruder (M/s APV Baker, UK, and Model: MP19PC).

In this process, the temperature profiles in the barrel were 200°C, 220°C, 230°C, 240°C, 250°C from hopper to die. The screw length to diameter ratio (L/D) was 25 and screw rotation rate of 60 rpm was used. Tensile, flexural and Izod impact samples (according to ASTM D-638 M91, ASTM D 790 and ASTM D 256–92 respectively) were prepared using an injection molding machine (M/s Boolani Engineering, Mumbai) with a barrel temperature of 220°C, 250°C, 280°C. Uniaxial tensile tests were carried out using Universal tensile testing machine (LR 50 k) from Lloyd instruments ltd. (U.K) at a cross head speed of 50 mm per minute. The impact test was carried out at room temperature using an Avery Denison impact tester. Heat distortion temperature (according to ASTM D 648) was measured using Davenport Vicat Softening point instruments ltd. (U.K). Dielectric strength (according to ASTM D 149) was measured (using a 2 mm thick composite sheet) by

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TABLE I Suppliers and physical properties of the filler

Material used	Supplier	Avg. particle size (μm)	Particle size distribution (μm)	Specific surface area (m^2/g)
Mica	M/s BS Mica Pvt. ltd., Mumbai	75	0.9–180	0.191
	M/s Hindustan Mineral Processing, Mumbai	37	0.4–90	0.837

Zaran electrical instruments (input: 240 V, 50 Hz, 1 PH; output: 0–50 kV; capacity: 100 mA; rating: 15 min). The values reported are the average values from at least five samples.

3. Results and discussions

3.1. Effect of particle size, distribution of mica and its concentration on the mechanical properties of filled nylon-6 composites

It is observed from Fig. 1 that as the concentration of filler increases tensile strength values decreases initially and thereafter increases in case of larger particle size (75 μm) mica, whereas it gradually increases in case of smaller particle size mica (37 μm). The rate of increase of tensile strength is higher for smaller particle size mica. It is also observed from Fig. 2 that percentage elongation decreases drastically on addition of filler in case of both the particle size of mica which indicates interference by the filler to the mobility or deformability of the matrix. This interference must be created through the physical interaction, thereby immobility of the polymer matrix by imposing mechanical restraints. These variations may be due to wide particle size distribution (psd) of larger particle size mica than that of smaller particle size mica. In case of larger particle size mica at higher filler loading the interstitial volume must have been occupied by smaller particle size filler and there may be insufficient matrix available for contributing the percentage elongation. This phenomenon can also be observed from the SEM micrographs, where at lower filler loadings interstitial voids is clearly seen which are later filled up by smaller particles at higher filler loading in case of larger particle size due to wide psd. The Young's modulus values as inferred from Fig. 3 increases with increase in filler concentration in both the cases but the rate of increase of Young's modulus is higher in case of larger

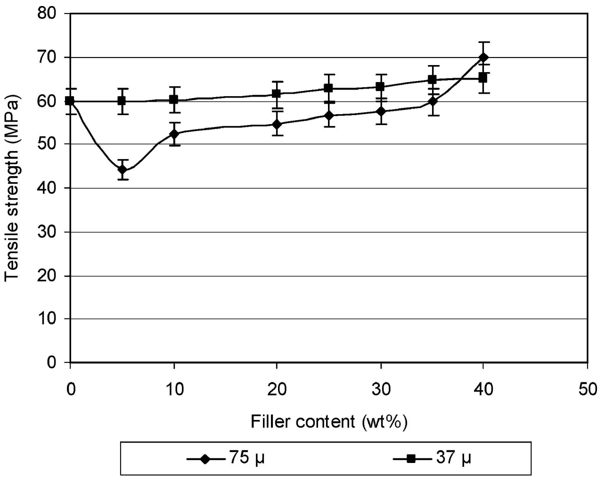

Figure 1 Variations in tensile strength of different particle size mica with varying concentration.

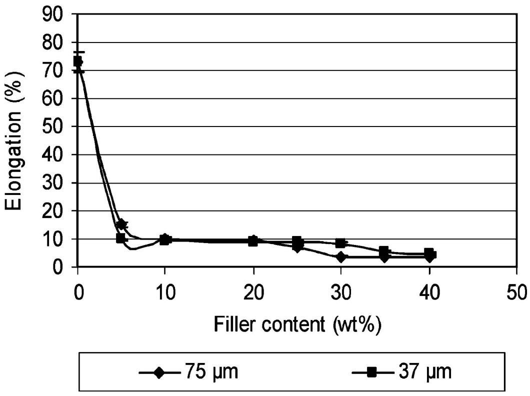

Figure 2 Variations in percent elongation of different particle size mica with varying concentration.

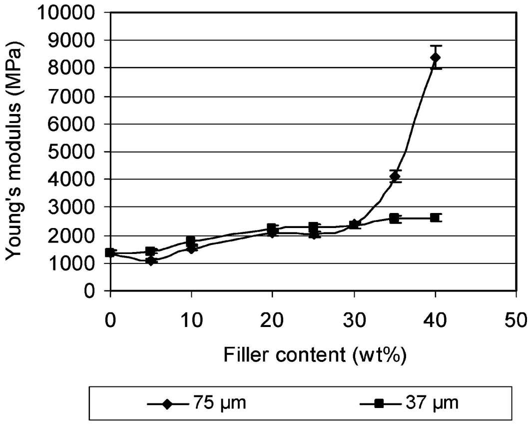

Figure 3 Variations in Young's modulus of different particle size mica with varying concentration.

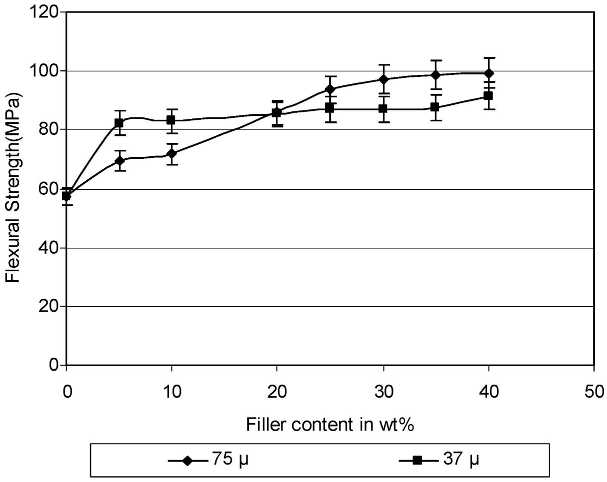

Figure 4 Variations in flexural strength of different particle size mica with varying concentration.

particle size than that of smaller particle size. At 40 wt% filler loading a six fold increase in Young's modulus is observed in case of higher particle size where as in case of lower particle size filler it is only two fold. These variations can be explained as: at higher filler loading the interstitial voids for the larger particle size mica is occupied by smaller particles and though they are agglomerated but still show some continuity where as such continuity is not observed in smaller particle size mica at higher loading which may be again due to narrow psd. These variations can be observed from the SEM micrographs. It is also reported that agglomerates or flocculated particles at higher filler loading provides higher modulli, since that portion of the matrix which is isolated in the agglomerates is less free to react to stress and strain than the continuous phase at values below the maximum packing fraction [1]. Flexural strength values as depicted from Fig. 4 increases with increase in concentration of filler loading in both the cases where as the rate of increase in flexural strength is higher in case of smaller particle size up to 20 wt%. The flexural modulus values as inferred from Fig. 5 increases continuously with increase in concentration of filler loading for both the particle size where as at higher loading for larger particle size the values of flexural modulus is higher than that of smaller particle size filler indicating the phenomenon of agglomeration of lower particle size filler. In case of higher particle size, flexural modulus increases almost with three fold with addition of 40 wt% filler loading where as in lower particle size it is only two fold.

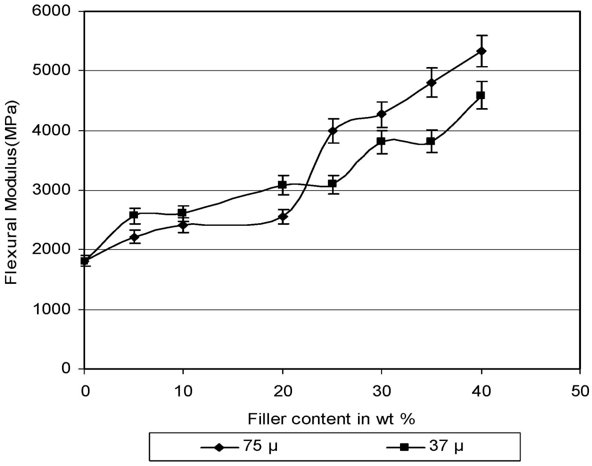

Figure 5 Variations in flexural modulus of different particle size mica with varying concentration.

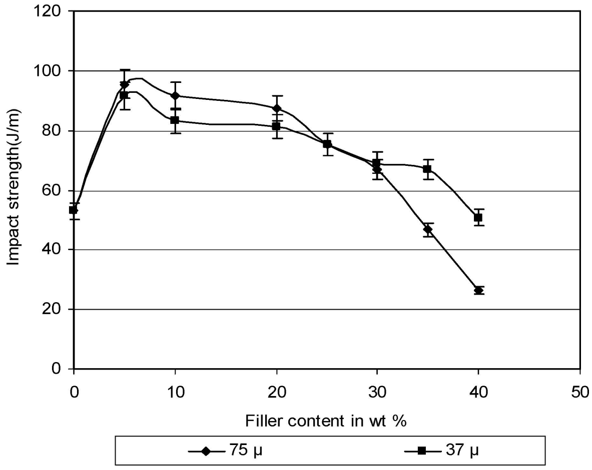

Figure 6 Variations in impact strength of different particle size mica with varying concentration.

The impact strength values as inferred from Fig. 6 attain maxima at 5 wt% in case of both the particle size. The strength increment at low weight percentage of filler may be attributed as, capacity to absorb more energy by increased portion of matrix. A further increase in weight percentage reduces the deformability of the matrix reducing in turn the ductility in the skin area so that the composite tends to form a weak structure. It is also observed that the rate of reduction of impact strength is more at higher filler concentration in case of higher particle size filler than in case of lower particle size filler. This may be due to the fact that for the same weight fraction of the filler material, the smaller the size, the larger will be their surface area, leading to increase in energy absorption before fracture.

3.2. Effect of particle size, distribution of mica and its concentration on dielectric and thermal properties of filled nylon-6 composites

It is depicted from Fig. 7 that on addition of mica the values of dielectric strength increase drastically in both the cases at lower concentration of filler. In case of

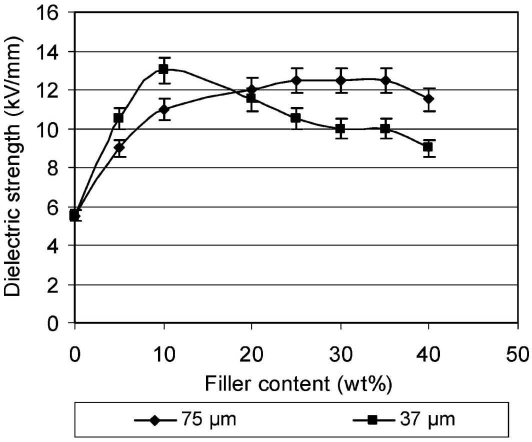

Figure 7 Variations in dielectric strength of different particle size mica with varying concentration.

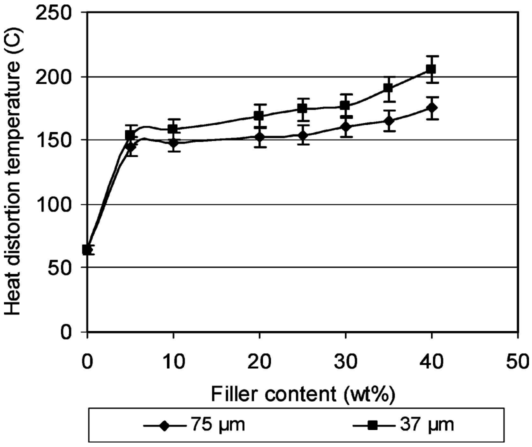

Figure 8 Variations in Heat distortion temperature of different particle size mica with varying concentration.

larger particle size mica the dielectric strength remain unchanged beyond 20 wt% filler loading, where as in case of smaller particle size it has attained maxima at 10 wt% filler loading and reduced thereafter. This trend in variation of dielectric strength in mica is attributed to the total surface area available from the filler as well as its continuity. The initial increase in dielectric strength in both the cases must be due to uniform dispersion of fillers at lower concentration, where as at higher concentration smaller particle size filler may not be dispersed properly but it agglomerates. Due to agglomeration the continuity of the filler at higher filler loading of smaller particle size may not be available but for larger particle size the platelets of fillers provide continuity, though agglomerated, in the surface area thereby maintain the constant dielectric strength. The heat distortion temperature values can be found from the Fig. 8. The values increase with increase in concentration of filler in both the cases. It is also reported that mineral fillers like mica generally rise the softening point of plastics by restricting the motion of polymer chains [23].

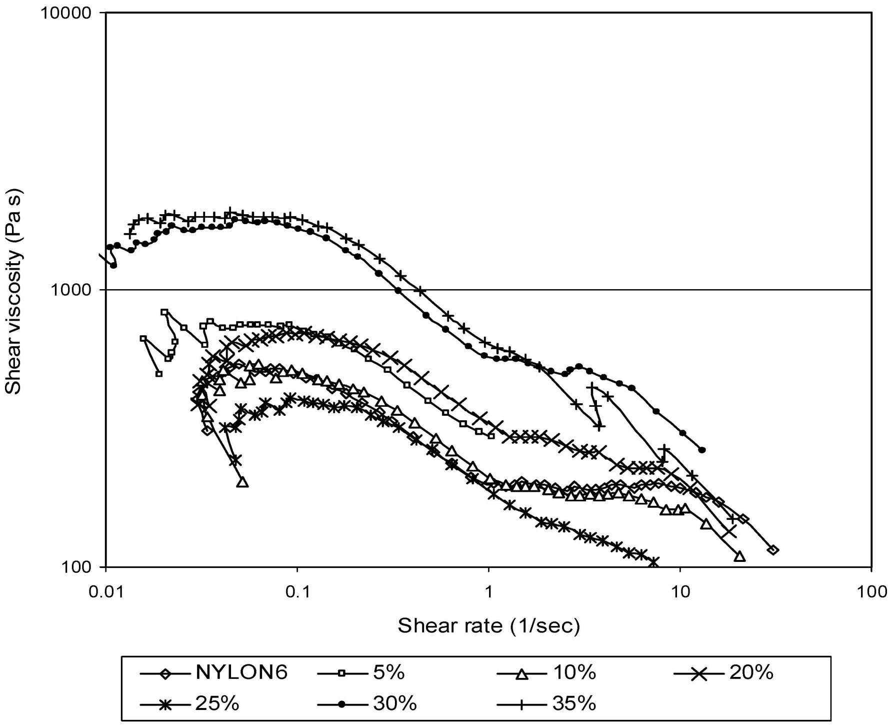

Figure 9 Variations in shear viscosity of nylon-6 with 75 μm mica at 250°C.

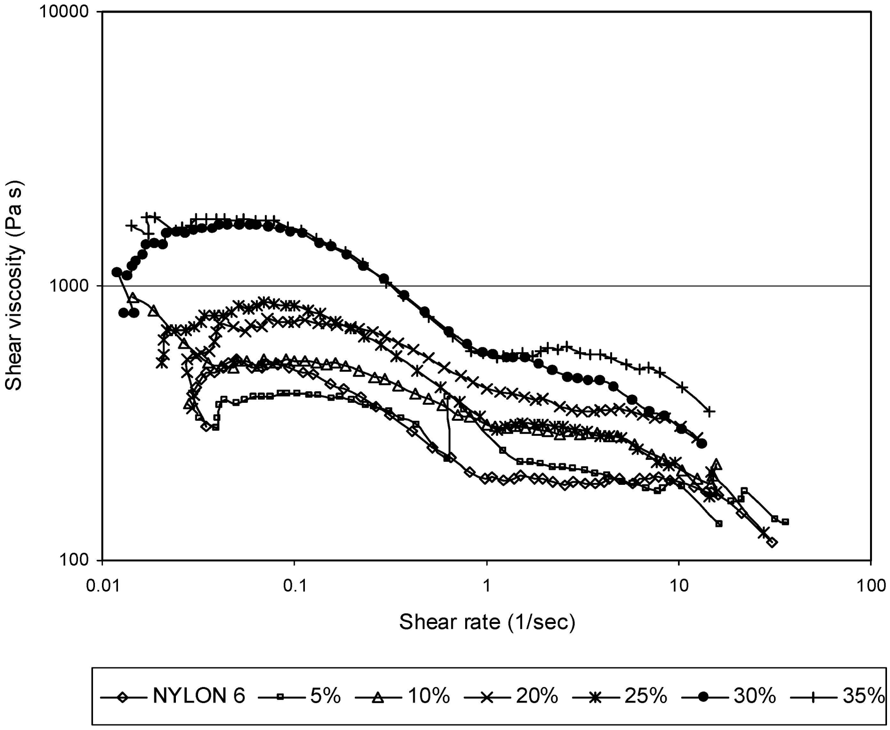

Figure 10 Variations in shear viscosity of nylon-6 with 37 μm mica at 250°C.

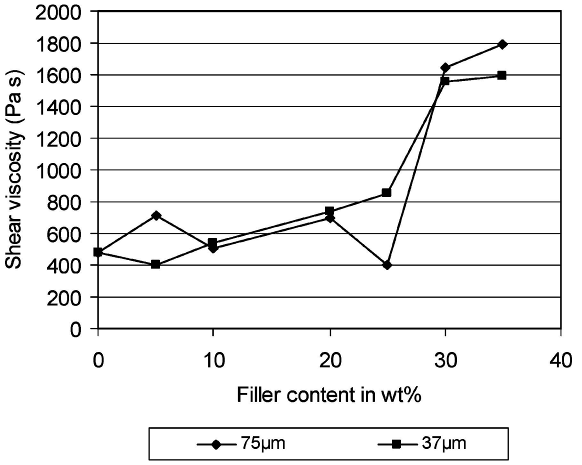

Figure 11 Variations in shear viscosity of nylon-6 with mica at 0.1 s^{-1} shear rate and 250°C.

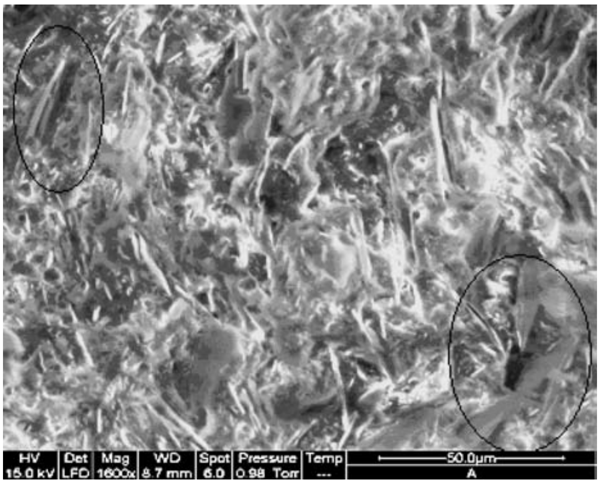

Figure 12 SEM micrograph of fractured 75μm mica composite at 30 wt% loading (agglomeration of filler particles is indicated).

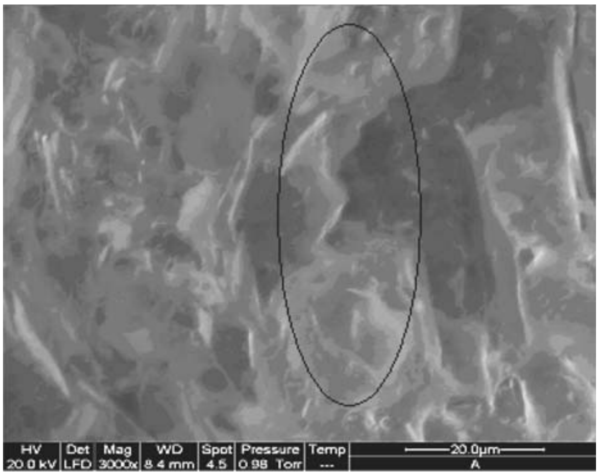

Figure 13 SEM micrograph (higher magnification) of fractured 75μm mica composite at 30 wt% loading (continuity of the agglomerates is indicated).

3.3. Effect of particle size of mica on rheological behaviour of mica filled nylon-6 composite

Figs 9 and 10 depicts the variations in shear viscosity versus shear rates at 250°C of 75 μm and 37 μm mica filled Nylon 6 composite respectively. Increase in the viscosity may be attributed to the properties of the filler such as maximum packing fraction [24]. Rate of increase in the viscosity depends upon the ratio $(ø/ ø_{\mu})$ where $ø$ = volume fraction of the filler and $ø_{\mu}$ = Maximum packing fraction. Increase in the viscosity may also be due to the ability of fine particles of fillers to form a large network that caused tighter packing. The porous and irregular shaped filler particles introduced discontinuity in the base matrix. The extent of discontinuity increased with the increase in filler content in the composite. Thus it appears that melt viscosity of base matrix increased due to the increasing obstruction to the flow caused by these irregular shaped filler particles [25, 26]. Addition of filler did not alter the pseudoplastic behavior of the polymer matrix. With an increase in filler content the viscosity of the component increased. Lower viscosity of filled compounds may indicate slip between filler particles and polymer matrix. In polymer processing, the increased viscosity of filled plastics at high shear rates is of continuing interest. It is seen that the shear viscosity increases with increase in concentration of filler except slip at 25 wt% filler loading in case of larger particle size mica. The slip in case of larger particle size mica may be due to change in packing fraction of filler beyond certain concentration of filler loading where the filler matrix interaction reduces where as it remains almost constant at higher concentration due to less variation in packing fraction. In case of smaller particle size mica the viscosity increases with increase in filler concentration indicating minimal variation in packing fraction of the filler. These variations can also be observed from Fig. 11 where the viscosities of both the particle size of mica/nylon-6 composites is compared at 0.1 s^{-1} shear rate and at 250°C.

3.4. Effect of particle size of mica on morphology of mica filled nylon-6 composite

Fig. 12–16 presents the Scanning Electron Microscope (SEM) micrograph of fractured tensile samples. The SEM micrograph of fractured 75 μm mica composite at 30 wt% loading can be found from Fig. 12. It can be clearly observed from this figure that at higher loading agglomeration of filler particles occurred but still showed some continuity (Fig. 13*)* which were not the case at lower filler loading (Fig. 14*)*. Again at lower filler loading though the filler particles are encapsulated but still there are interstitices which are to some extent filled up by smaller filler particles in case of larger particle size mica due to wide psd. The SEM micrograph of 37 μm mica composites at 30 wt% filler loading can be found from Fig. 15. Agglomeration of filler particles can be observed from this figure which is not the case at lower filler loading (Fig. 16*)* where the filler particles are encapsulated in the matrix. Thus due to agglomeration of lower particle mica and continuity in the agglomerates of filler in case of higher particle size the packing fraction at 30 wt% filler loading is same in both the cases, due to which the viscosity remained almost same at 30 wt% filler loading for both the particle size filler.

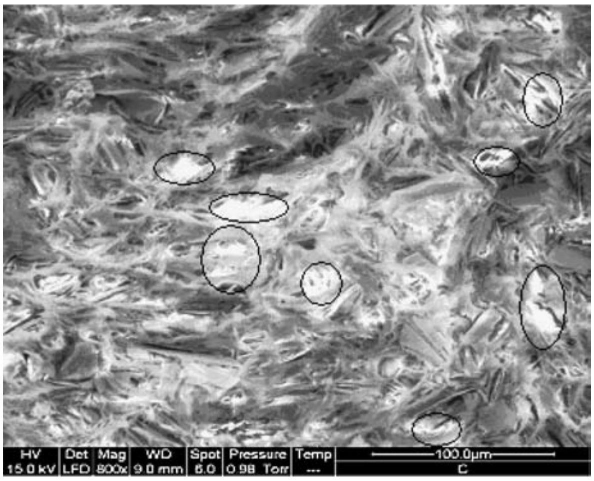

Figure 14 SEM micrograph of fractured 75 μm mica composite at 20 wt% loading (interstices are indicated).

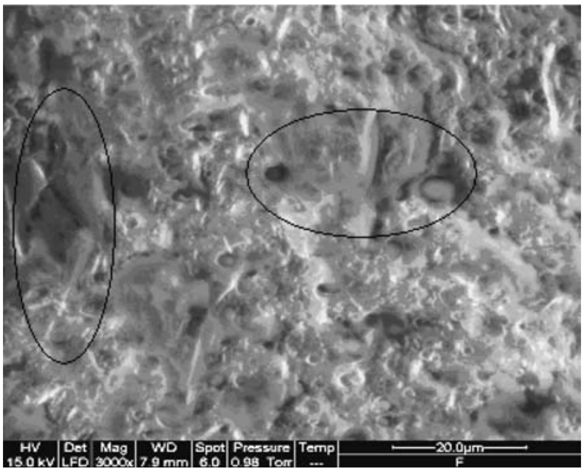

Figure 15 SEM micrograph of fractured 37 μm mica composite at 30 wt% loading (agglomeration of filler particles is indicated).

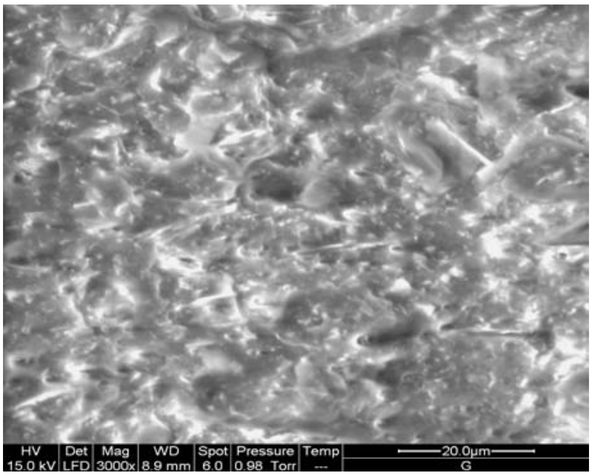

Figure 16 SEM micrograph of fractured 37 μm mica composite at 20 wt% loading.

4. Conclusions

- Addition of mica increased the mechanical properties (except percent elongation at break), dielectric as well as thermal properties of composite.
- The larger particle size mica showed better improvement as compared to smaller particle size mica, which may be due to continuity of filler particles even at higher loading.
- Inorganic fillers viz. mica added to the polymer improves their rigidity and heat resistance.
- Thus the mechanical property of composite is a function of the particle size, the dispersion and the particle size distribution.

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