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Studies on Long Glass Fibre Reinforced Polyamide-6 Composites

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ABSTRACT

Long fibre reinforced thermoplastics (LFRT) is a recent development in newer technologies where thermoplastic material is directly compounded with glass fibre roving and then molded. There is tremendous scope of the LFRT composite in the automotive and other engineering applications. This study deals with the development and analysis of injection molded long glass fibre reinforced polyamide 6 composites. Glass fibre roving was impregnated with polyamide 6 in a single screw extruder using specially designed die. The impregnated strands were chopped into pellets of different lengths. The pellets were injection molded to get long fibre-reinforced polyamide composite and its mechanical properties were analyzed. Interfacial shear strength between glass fibres and polyamide matrix was studied. Morphological analysis was carried out to find out the fibre length distribution in the injection molded composite. As the fibre length increased, the tensile and flexural properties were improved up to 9mm initial glass fibre length followed by decrease in properties at higher initial glass fibre length. Impact strength increased with increase in glass fibre length. The average fibre length in the impregnated fibre composites was greater than the critical fibre length. The frequency of the fibres was found to be increased as the composite system traveled more distance from the gate while injection molding composite samples. The average fibre length after injection molding of the LFRT composite with 9 mm initial fibre length was found to be more than the average fibre length of composites with 3 mm, 6 mm and 12 mm initial fibre length. Moreover, the average fibre length in the injection-molded composite is greater than the critical fibre length.

Keywords : Polyamide 6, Long fibre reinforced thermoplastic composites, Injection Molding

1. INTRODUCTION

Composites are classified as short and long fibre composites in terms of aspect ratio of fibres, which is the length to diameter ratio of the fibre present in the matrix. If the aspect ratio of the fibre in the composite is greater than 100, it is considered as Long Fibre composite. This long fibre composite may be utilized to design parts that require extra strength and stiffness.

Thomason [1, 2] studied the mechanical properties and residual fibre length distributions of glass fibre reinforced polyamide 6, 6. Richad [3] investigated the methods of making continuous length reinforced plastic articles. The continuous glass fibre composites were prepared using the single screw extruder and a crosshead die which facilitated impregnation of the glass fibre to develop 2D profiles with desired cross sectional shape. Bailey et. al [4] investigated the processing and property characteristics for long glass fibre reinforced polyamide. Fibre attrition was enhanced by the backpressure during injection molding. Peogretti et. al. [5] investigated the interfacial properties between E-glass fibres with different commercial sizing on model composites with polyamide 6 matrix. Interfacial shear strength was determined using the single fibre fragmentation test. O'Regan and Akay [6] studied the distribution of fibre lengths in injection molded polyamide composite components. The fibre length distribution exhibited particular sensitivity to nozzle arrangement, mold geometry, position within the molding and fibre content. The use of hydraulic shut-off nozzle led to increased fibre breakage above a standard nozzle; this was due to a combination of nozzle design and reduced dimensions. Additional fibre attrition occurred when the gate dimensions were decreased. Hassan et al [7, 8] studied the tensile, impact and fibre length properties of injection-molded short and long glass fibre-reinforced polyamide 6, 6 composites. Extrusion and pultrusion methods were employed for the melt compounding of polymer composite feedstock for injection molding and

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produced short and long fibre composites (SFCs and LFCs), respectively. Both tensile strength and tensile modulus of LFCs improved compared to the SFCs counterpart despite reduction in fracture strain, while pultrusion compounded composites showed superior fibre characteristics, in terms of fibre length distribution compared to the extrusion compounded composite counterpart [9-12]. Detailed study of fibre length distributions in injection molded long fibre composites [13-16] decided the process parameters for injection molding.

The aim of the present work is to study the effect of the initial glass fibre length on the mechanical properties of the injection molded long fibre reinforced polyamide 6 composites. The glass fibre rovings were impregnated with the polyamide 6 melt in the specially designed radial impregnation die attached to a single screw extruder. The LFRT composite products were developed from the neat polyamide 6 and the impregnated strands were palletized at different lengths for injection molding. These long fibre pellets has the fibre length equal to the pellet length such as 3, 6, 9 and 12 mm. Mechanical and morphological properties were evaluated and correlated with the fibre length distribution. Adhesion between the glass fibre and the thermoplastic melt was studied by interfacial shear strength. This study aids in finding out the critical fibre length of composites.

2. EXPERIMENTAL

2.1 Materials

The Polyamide 6 matrix used was Gujlon M28 RC having melt flow index of 28 supplied by M/s. Gujarat State Fertilisers & Chemicals Limited, Vadodara, India and the continuous glass fibre roving (R099688) with 10 microns fibre diameter and 2400 tex having silane coupling agent was obtained from M/s. Saint Gobain, Hyderabad, India as the reinforcement for the composite specimen preparation.

2.2 Processing

The continuous glass fibre rovings were impregnated with polyamide 6 using Klockner Windsor made single screw extruder having L/D ratio of 24:1 and screw diameter of 30 mm. The process of impregnation is carried out as shown in the Figure 1. A specially designed radial impregnation die was used to better disperse the glass fibres in the thermoplastic melt. The die has been designed considering several factors like residence

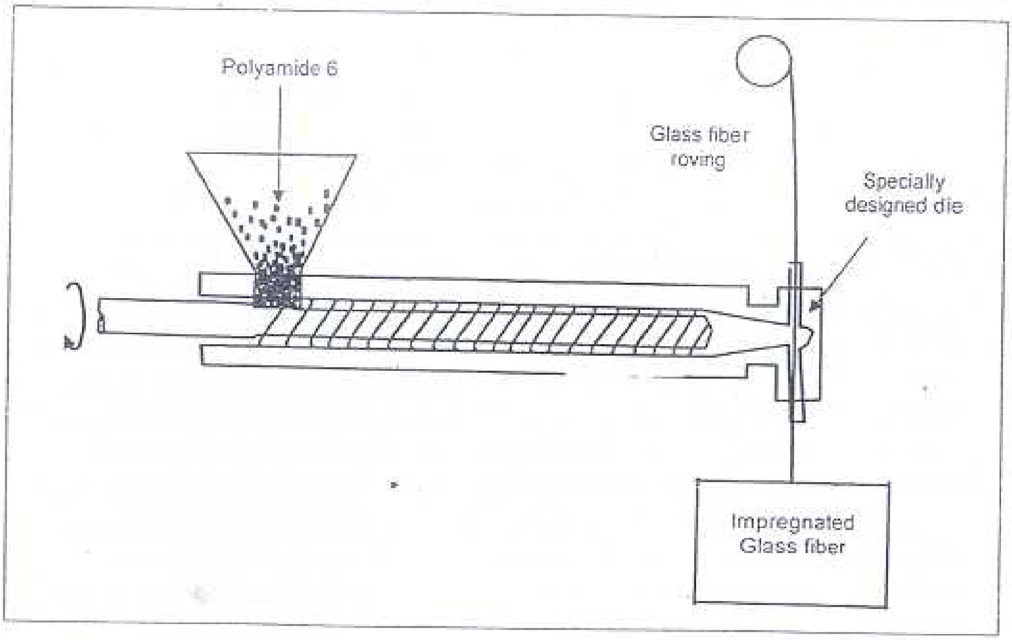


Figure 1 : Process of impregnation in Single Screw Extruder

time, fibre tension and spreadability of the glass fibres. The temperature profile of 210 – 240°C and the screw speed 20 rpm were maintained. The impregnated strands were pelletized at different lengths such as 3mm, 6 mm, 9 mm and 12 mm using vertical milling cum slotting machine. The pellet length equals the fibre length. The L&T Demag made injection-molding machine (Model LNC 4P) was used to prepare the long fibre reinforced polyamide 6 (LFPA) composite specimens. LFPA composites were prepared using all four different sizes of chopped pellets i.e. 3, 6, 9 and 12 mm at 5, 10 and 15wt% of glass fibres. The temperature profile was 210 to 260°C and the screw speed was 110 rpm.

2.3 Determination of fibre weight and fibre length

The flexural test specimen was divided in three equal portions as mentioned in Figure 2 for determining the glass fibre content in injection molded composite. The portion I selected was the nearest portion from the gate. The portion III is the farthest portion from the gate of the injection molded composite sample. These three portions were separately dissolved in the concentrated formic acid for eight hours at room temperature. Then the separated glass fibres were collected after filtration and drying. The weight of all the three portions was measured.

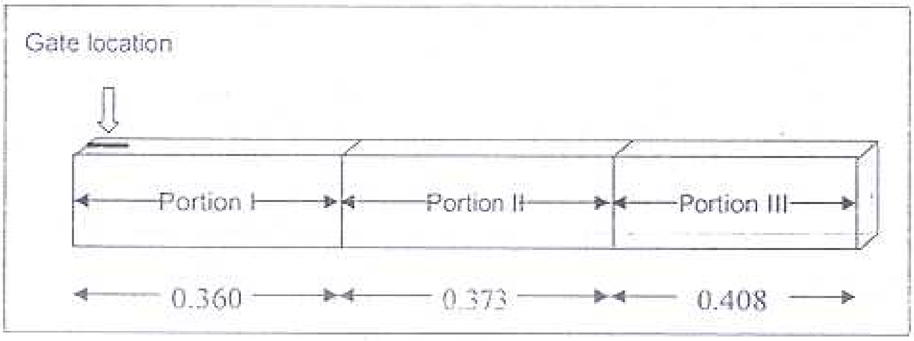


Figure 2 : Glass fiber weight fraction (%) determined from flexural test sample

Fibre length in injection molded long fibre composites are determined by dissolution technique. LFPA composite samples were cut from the injection molded flexural test specimens for dissolution technique. The specimens were dissolved in concentrated formic acid in the conical flask for eight hours at room temperature. Then the separated glass fibres were collected after filtration and drying. The fibre length distribution (FLD) was determined for various samples using optical microscope with a magnification of 10X.

2.4 Evaluation of properties

The Iosipescu test specimens were prepared by injection molding for the Interfacial Shear Strength (IFSS) testing. The universal testing machine was used to test IFSS at the crosshead rate of 5mm/min. as per ASTM D 5379. In this test double-edged notched specimen was subjected to two opposing force couples. The main advantage of this test is that a large region of uniform shear is obtained like other tests for composite materials. The stress concentration is proportional to the fibre orientation and the fibre volume fraction. The tensile properties were tested on Zwick Z010 Universal testing machine at test speed of 20 mm/min at room temperature as per ASTM D 638. The flexural properties were also tested using the same Zwick Z010 Universal testing machine at test speed of 10 mm/min as per ASTM D 790. The Izod impact testing was carried on RESIL Impactor (make: CEAST, Germany) as per ASTM D 256.

3. RESULTS AND DISCUSSION

3.1 Mechanical properties

Figures 3 and 4 show the effect of fibre length on the tensile strength and tensile modulus respectively of the injection molded samples (LFPA-3, LFPA-6, LFPA-9 and LFPA-12) containing different initial fibre lengths such as 3, 6, 9 and 12 mm at different fibre content (5, 10, 15 and 20 wt%). As fibre content increased, the tensile properties increased linearly by 20%. As fibre length was increased, the tensile strength and tensile modulus values were increased by an average of 7-9 %. The tensile strength value of neat polyamide 6 was measured as 49.4 MPa. There was very little improvement in the tensile strength value in case of 5wt% with the initial fibre length. In case of 10wt% composite, the increase in tensile strength value of the composite with 3mm initial fibre length was 40% compared to that of neat polyamide 6. Only 6% improvement in the tensile strength of LFPA-6 sample (LFPA composite having 6 mm initial fibre length) was observed compared to that of LFPA-3

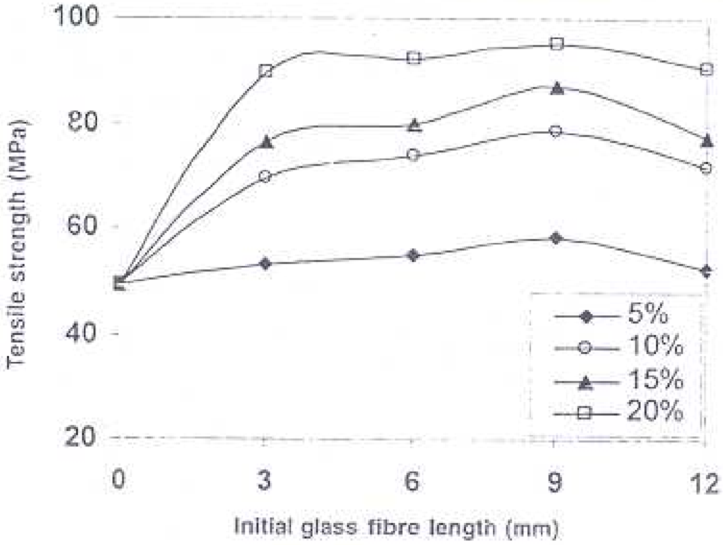


Figure 3 : Tensile strength of LFPA composites

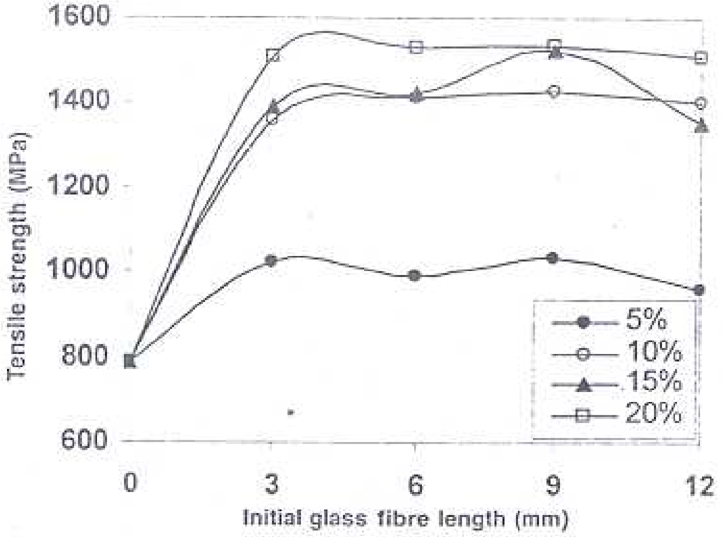


Figure 4 : Tensile modulus of LFPA composites

sample (LFPA composite having 3 mm initial fibre length). There was marginal improvement with fibre length upto 9 mm and then decreased slightly on further increase of fibre length. It is evident from the figure 4 that there has been 30% improvement in the value of tensile modulus of composite with 5 wt% glass fibres of 3mm initial length compared to that of neat polyamide 6 and then there was marginal changes in the value on further increase of the fibre length. There is good improvement in the value of tensile modulus in case of higher wt% (10, 15 and 20) of glass fibres. The tensile modulus value of the composite of 20 wt% glass fibres with 9 mm initial fibre length is 48% higher than the composite of 5 wt% glass fibres with same 9 mm initial fibre length.

Figures 5 and 6 show the effect of fibre length on the flexural strength and flexural modulus respectively of the injection molded samples (LFPA-3, LFPA-6, LFPA-9 and LFPA-12) containing different initial fibre lengths such as 3, 6, 9 and 12 mm at different fibre content (5, 10, 15 and 20 wt%). As fibre content increased, flexural strength increased linearly by 15-18%. As fibre length increased flexural strength and flexural modulus values

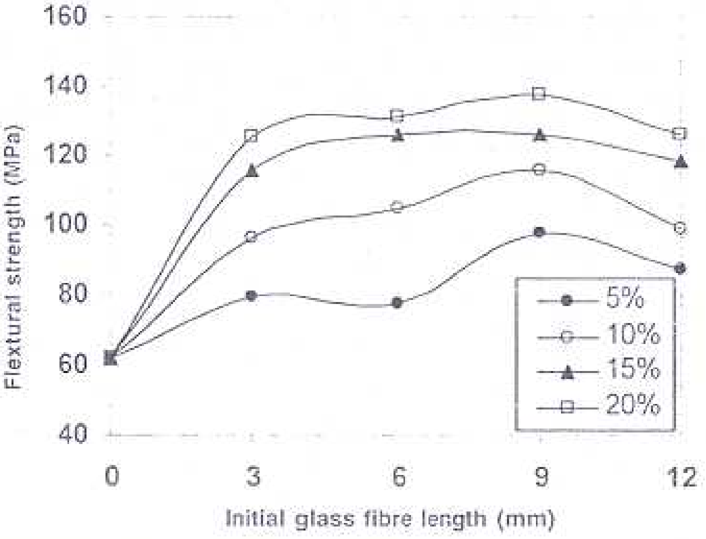


Figure 5 : Flexural strength of LFPA composites

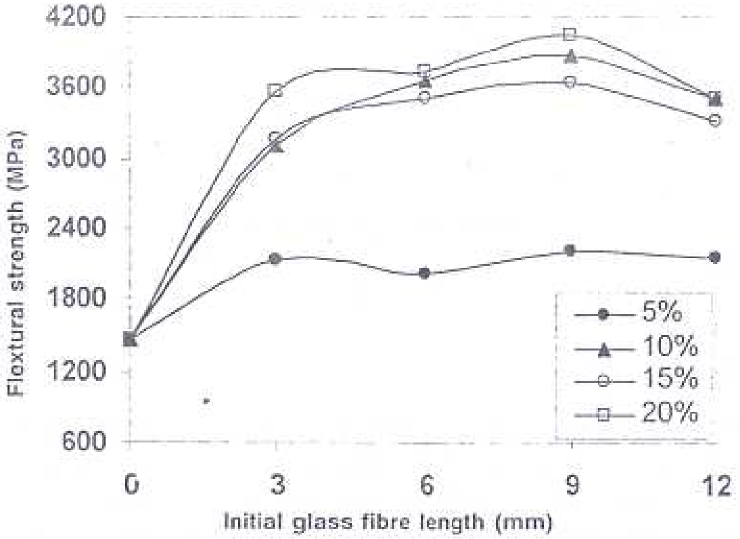


Figure 6 : Flexural modulus of LFPA composites

were increased by an average of 8%. The flexural strength value of neat polyamide 6 was measured as 61.7 MPa. There was linear improvement in the flexural strength value in case of 5 wt% upto 9 mm initial fibre length and decreased on further increase in the fibre length. In case of 10 wt% composite, the increase in flexural strength value of LFPA-6 was 10% compared to that of LFPA-3. It is evident from the figure 6 that there was 45% improvement in the value of flexural modulus of LFPA-3 composite with 5 wt% glass fibres compared to that of neat polyamide 6 and then there were marginal changes in the value on further increase of the fibre length. There was significant improvement in the value of flexural modulus in case of higher wt% (10, 15 and 20) of glass fibres. The improvement in the flexural strength and the flexural modulus value of the LFPA-9 composite with 20wt% glass fibres was found to be higher by 60% and 55% respectively than those of the LFPA-9 with 5wt% glass fibres.

Figure 7 shows the effect of fibre length on the impact strength of the injection molded samples (LFPA-3, LFPA-6, LFPA-9 and LFPA-12) containing different initial fibre lengths such as 3, 6, 9 and 12 mm at different fibre contents (5, 10, 15 and 20 wt%). As fibre content increased, the impact strength value of neat polyamide 6 is 31.9 MPa. It is evident from the Figure 7 that there was 150% improvement in the value of impact strength of LFPA-3 composite with 5wt% glass fibres compared to that of neat polyamide 6 and then there were marginal changes in the value on further increase of the fibre length. In case of 20 wt% composite, the increase in impact strength value of LFPA-6 and LFPA-9 were found to be 19% and 43% compared to that of LFPA-3. The improvement in the impact strength value of the LFPA-9 composite with 20 wt% glass fibres was found to be higher by 110% than that of the LFPA-9 with 5 wt% glass fibres.

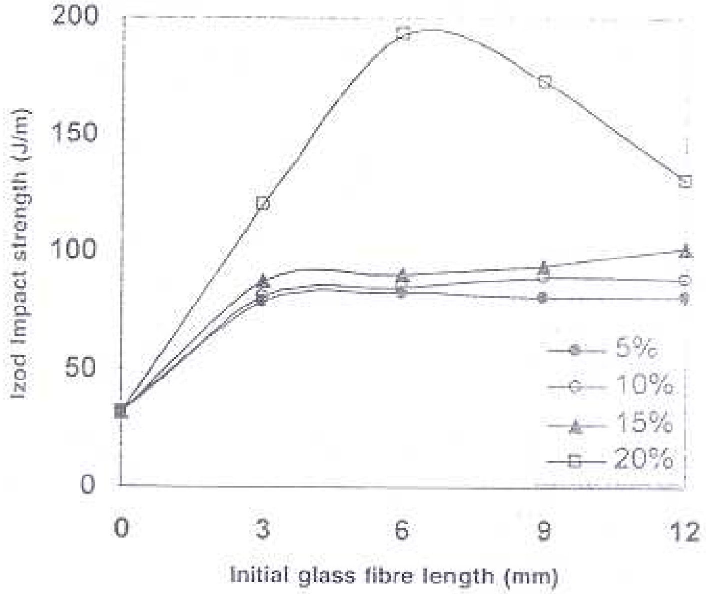


Figure 7 : Impact strength of LFPA composites

3.2 Interfacial shear strength

The interfacial bonds created by silane or other coupling agents improve interfacial shear strength so that it can allow a better shear stress transfer between fibre and matrix, which in turn improves the failure strength. If the interfacial shear strength is high, then the value of critical fibre length decreases, which is favorable for the process like injection molding where high shear is applied to the material and there are ample chances of fibre attrition. The interfacial shear strength is important to find out the critical fibre length. The mean fibre shear stress measured from the Iosipescu test was 41.82 MPa. The critical fibre length l_c can be given by

$$l_c = \frac{\sigma_{fu} d_f}{2\tau} \qquad (1)$$

where, σ_{fu} is fibre tensile strength, d_f is diameter of fibre and τ is interfacial shear stress.

Using equation (1), the critical fibre length was calculated as 0.7 mm. If mean fibre length in the composite product is greater than five times the critical fibre length, then approximately 90% of the ultimate fibre strength can be achieved in the composite product [17].

3.3 Fibre weight determination

Figure 2 also shows the weight fraction of fibres (in %) in the three portions of injection molded flexural test sample. The portion I is the nearest portion from the gate as shown in Figure 2. The portion III is the farthest portion from the gate of the injection molded composite sample. The weight of fibres in the portion II is more than the weight of fibres in the portion I and similarly the weight of fibres in the portion III is more than the weight of fibres of portion II. It was observed that the concentration of fibres increased with the distance from the gate of the injection molded samples. This may be due to the higher density of glass fibres compared to that of neat polyamide 6.

3.4 Fibre length distribution (FLD)

Fibre length distribution of composite samples (LFPA-3, LFPA-6, LFPA-9 and LFPA-12) containing different initial fibre lengths such as 3, 6, 9 and 12 mm at 15wt% fibre contents was as shown in the Figure 8. The mean fibre length in the injection molded composite increases with increase in the initial fibre length as mentioned in the Table 1. In case of 12 mm initial fibre length, the fibres are scattered in the wide range of fibre length. The average fibre length of LFPA-12 composite is almost equal to the average fibre length of LFPA-6 composite. More than 2mm of the fibre length was not measurable due to equipment constraints. However, the fibres with length more than 2mm were less than 10%.

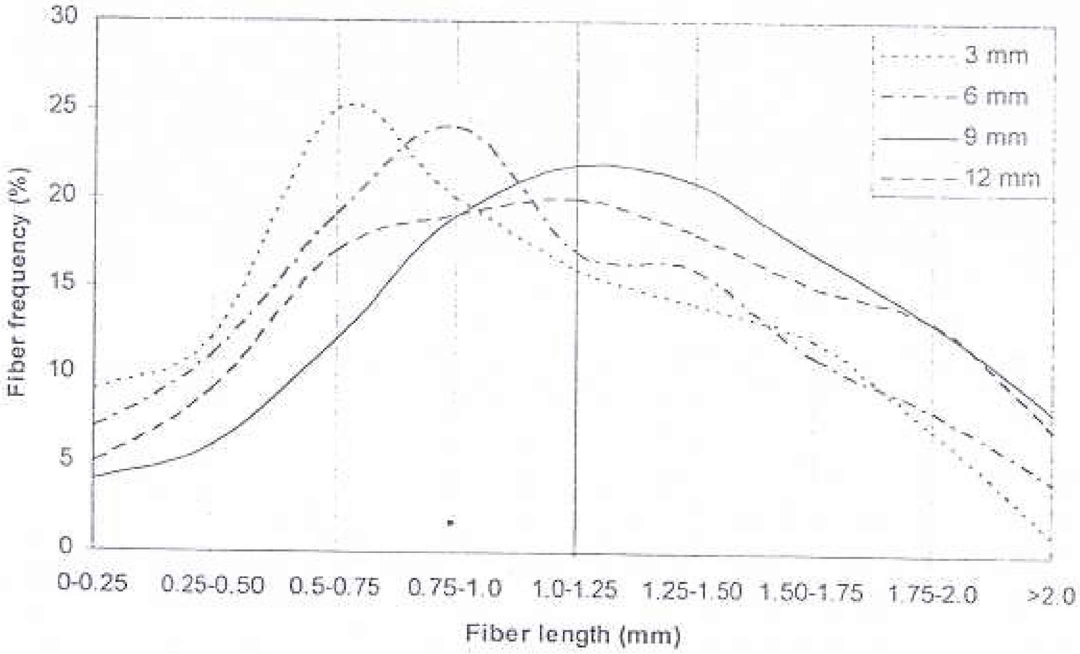


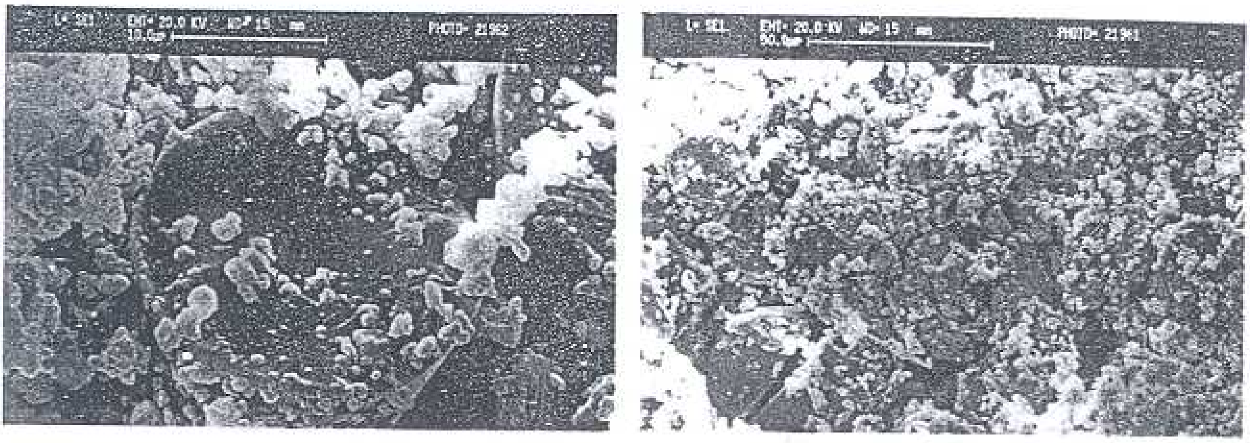
Figure 8 : Fiber length distribution of various composite samples

Table 1: Mean fiber length of injection molded LFRT composite

Initial fiber length (mm)	3	6	9	12
Mean fiber length after molding (mm)	0.743	1.026	1.131	1.010

3.5 Microscopy studies

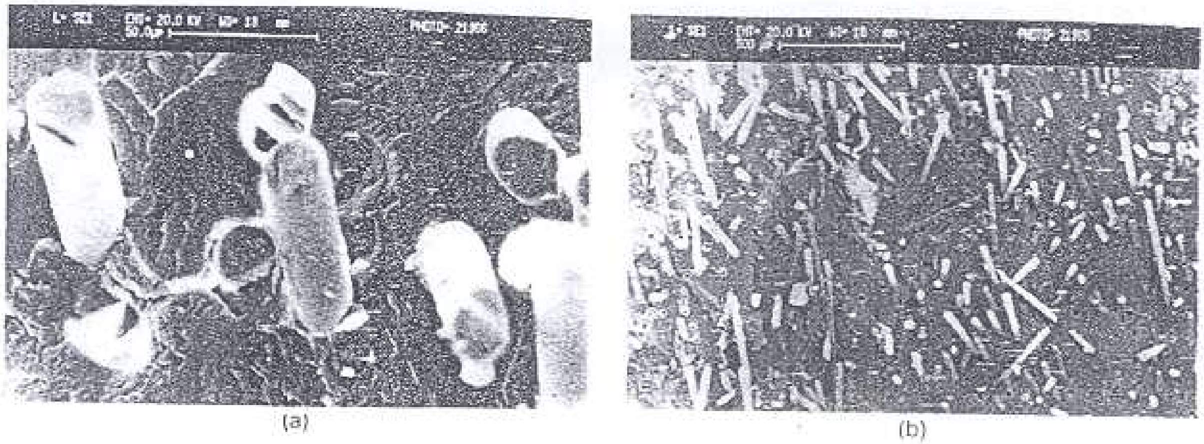
The continuous impregnated roving and the injection molded tensile test samples were chilled in the liquid nitrogen and cryogenic fracture was carried out to get the SEM test samples. The SEM was carried out on Cambridge S4-10 Stereo Scan microscope at lower as well as higher magnification to study the distribution of fibres in the product and the wetting ability of the fibres with the matrix. SEM micrographs of polyamide melt impregnated glass fibre roving at higher and lower magnification are shown in the Figure 9. These micrographs reveal that the inter-fibre spacings are found to be considerably filled with polyamide melt. The fibres remaining in the periphery of the roving are wetted completely by Polyamide melt. This may be due to the use of specially designed radial impregnation die for impregnating the fibre roving. The fibres in the central region of the roving are partially wetted by the polyamide melt because of the lesser inter diffusion of the polyamide melt from the circumferential region towards the centre.



(a) (b)

Figure 9 : SEM micrographs of extruded strands of polyamide 6 impregnated glass fiber roving at (a) higher magnification [1400 X] and (b) lower magnification [500 X]

Figure 10 show the SEM micrographs of injection molded glass fibre reinforced polyamide composite sample with 20wt% glass fibres with 9mm initial fibre length. The low magnification micrograph revealed that there was better distribution of fibres throughout the cross section of the sample. Since the fibres are coated with amino silane coupling agent on their surface, a better wetting of the glass fibres is observed. Due to this reason, very few fibres were pulled-out from the matrix during fracture. This may be due to the better mixing action and shear onto the fibres and the matrix during injection molding.



(a) (b)

Figure 10 : SEM micrographs of LFPA-9 composite with 20wt% of glass fibers at (a) higher magnification [1400 X] and (b) lower magnification [500 X]

4. CONCLUSION

The Polyamide 6 impregnated continuous glass fibre roving was developed on the single screw extruder with the specially designed die. The processing of impregnated glass fibre pellets with neat Polyamide 6 in various proportions was carried out on injection molding machine to develop long fibre reinforced thermoplastic composites. With the increase in fibre content, the tensile, flexural and impact properties of injection molded long fibre polyamide 6 composite samples improved. At a constant fibre loading, the tensile and flexural properties improve significantly with increase in fibre length upto 9 mm. The impact strength of the composite with 20 wt% glass fibre with 9 mm initial fibre length was found to be five times than that of the neat polyamide 6. The interfacial shear strength of the glass fibre reinforced polyamide 6 composite sample was measured by Iosipescu shear test to determine the critical fibre length which was 0.7 mm. It was observed that the average fibre length in injection molded LFPA composite was greater than the critical fibre length and the composite with 9 mm initial fibre length showed highest value of average fibre length after molding. The concentration of the fibres increased with the distance from the gate in the injection molded composite samples.

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