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# EFFECT OF LCP ADDITION ON THE PROPERTIES OF HYBRID COMPOSITES

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## Abstract

A hybrid composite consisting of rubber toughened nylon 6,6, glass fiber and LCP was investigated by varying LCP content. The hybrid system exhibited better processability than the glass fiber reinforced composite. A decrease in the total torque was observed with the LCP content indicating the reduction in the energy consumed during the processing of the hybrid composites. Thermal stability of the glass fiber reinforced composites improved with LCP addition. SEM observation of the tensile fracture surface revealed LCP fibrillation in the toughened matrix.

## Introduction

Reinforcing toughened polymer matrices with short glass fibers is an attractive route to minimize the reduction in strength and stiffness arising from the compliant elastomeric phase [1,2]. Elastomers toughen the polymer matrix by promoting matrix plasticity through rubber cavitation, which first relieve the triaxial tension at the crack tip [3]. The advantage of fiber reinforcement is two-fold: (1) enhance the mechanical properties such as strength, stiffness and heat deflection temperature and (2) promote fiber-induced toughening in the matrix [4]. Presence of inorganic fillers raises the melt viscosity of the polymer, resulting in higher energy consumption and lower processability. Fiber reinforcement often catalyzes the tear and wear of the processing equipment. The shear forces during the compounding process lead to fiber breakage affecting the reinforcement effect of glass fibers. Blending of a reinforced matrix with LCP offers the potential to reduce the viscosity thereby improving the processability. The mechanical performance of such a hybrid composite is also comparable to that of glass fiber reinforced polymers [5]. He and coworkers [6,7] reported synergistic properties for LCP/glass fiber/thermoplastic hybrid composites. However, little is understood on materials containing an engineered combination of short glass fibers, LCP and elastomeric phase. Through a series of

studies we seek to investigate the desirable properties of such a complex hybrid system. This paper addresses the effect of addition of LCP on the processability, thermal stability and mechanical properties of glass fiber reinforced toughened thermoplastics.

## Experimental work

Rubber-toughened nylon 6,6 (Zytel ST801 from Du Pont) was dry blended with 20 wt% of short E-glass fibers (length=12mm; diameter=17 $\mu$ m) and independently with 5, 10, 15 and 20 wt% of LCP (Vectra A950 from Hoechst-Celanese). The LCP comprised 27-mol% of 2-hydroxy-6-naphthoic acid (HNA) and 73-mol% of p-hydroxy benzoic acid (HBA). Melt blending was carried out using a high shear rate, inter-meshing, co-rotating twin-screw extruder (Leistritz Micro 18; with a screw diameter of 18 mm and L/D ratio = 30). The temperature profile in the extruder was 260-280-285-285-292 $^{\circ}$ C. Screw speed was kept at 200 rpm. The extruded pellets were injection molded into 3.5 mm thick dog-bone specimens (ASTM D638 type I) using a Battenfeld BA 300 CD<sup>plus</sup> injection-molding machine. The temperatures at zone 1 and zone 2 were kept at 285 $^{\circ}$ C and 292 $^{\circ}$ C respectively. The nozzle temperature was kept at 275 $^{\circ}$ C and the mold temperature at 30 $^{\circ}$ C. An injection pressure of 70 bar and holding time of 50 s were used. The screw speed was kept at 140 rpm. Rubber toughened nylon 6,6 containing 20 and 30 wt% glass fibers were processed and compared to the properties pertaining to the hybrid composites. All the materials were dried at 80 $^{\circ}$ C for at least 72 h in a vacuum oven before processing.

Torque measurements were carried out in a Haake Rheocord-90 intermeshing, counter-rotating, twin-screw extruder. 100 g each of 20 wt% reinforced rubber-toughened nylon 6,6 and its independent blends with 5, 10, 15, 20 wt% LCP were extruded at a temperature of 290 $^{\circ}$ C while maintaining a screw speed of 100 rpm. Accumulated torque for a time of 2 min and the instantaneous torque were measured.

Thermal stability of the blends was assessed by thermogravimetric analysis (TGA) using a TA TGA 2950

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equipped with TA thermal analysis software. TGA measurements were done at a scan rate of 20°C/min purged with a stream of nitrogen.

Dynamic mechanical properties were assessed by dynamic mechanical thermal analysis (DMTA) using a TA DMA 2980 equipped with TA thermal analysis software. The measurements were done on the injection molded specimens at a scan rate of 3°C/min with a frequency of 1Hz. A double cantilever clamp was used in a bending mode

Scanning Electron Microscopy (SEM) observations of the tensile fracture surfaces were performed on a JEOL 5410 LV model SEM. The fracture surface was coated with gold in an SPI sputter coater. The tensile fracture surface was also studied using SEM. Morphological observation of the extrudates was conducted after cryogenic fracture in liquid nitrogen.

Tensile tests were conducted according to ASTM D638M using an Instron Model 5565 computer-controlled testing machine and the tensile strength, tensile modulus and tensile strain were simultaneously recorded. The crosshead speed was 5 mm/min at room temperature. Five specimens of each blend composition were tested.

## Results and Discussion

### Processability

Torque rheometry measures the work accumulated in the studied sample during melt mixing under controlled temperature and shear rate. Torque is directly proportional to the melt viscosity at a given time. The information generated from the total torque could be rather complex. It represents the resistance of the solids in the conveying zone of and the melt resistance throughout the extruder; this torque is integrated with respect to time. The total torque is indicative of the energy consumed for blending. Figure 1 illustrates the variation in torque with time for the 20 wt% glass fiber reinforced toughened nylon 6,6 and its hybrid composition containing LCP. The torque during extrusion for all hybrid compositions with varied content of LCP is lower than that experienced during the processing of rubber toughened nylon 6,6 reinforced with 20 wt% glass fiber. The accumulated torque for the LCP-containing hybrid composites integrated for 2 min decreases with increasing LCP content (Fig. 2). Clearly, the addition of LCP to glass fibre reinforced toughened nylon slashes the energy consumption during processing and enhances the processability of hybrid composites. The low energy consumption can be envisaged in light of viscosity reduction owing to the addition of LCP. It is understood that LCP could serve as a processing aid [8]. Previous studies [9,10,11] reported viscosity reduction on addition of LCP to thermoplastics. The viscosity reduction was attributed to the characteristic melt orientation of LCP

domains. But little work has been done on the effect of LCP on glass fiber reinforced thermoplastics. Based on these studies we conjecture that the LCP domains orient in the direction of the flow facilitating the viscosity reduction of glass fiber reinforced matrix.

### Extrudate Morphology

Figures 3a and b compares the cryogenically fractured extrudate morphologies containing 5 wt% (Fig. 3a) and 20 wt% LCP (Fig. 3b). The micrograph shows two main features. The continuous phase is the glass fiber reinforced toughened nylon and the LCP fibrils appear sparsely drawn out from the surface. At 5 wt% LCP, the LCP fibrils seem to be sparsely distributed over the surface, but as the LCP content increases to 20 wt% (Fig.3b) the LCP phase forms sheet like structures. We believe that this orientation and fibril formation of the LCP phase effectively enhanced the processability of LCP hybrid composites.

### Thermal Stability

Thermal stability results are summarized in Table.1. Blending of glass fiber reinforced toughened nylon 6,6 with Vectra A950 has improved the thermal stability of the blends. Since LCP possesses better thermal stability than flexible chain polymers, good thermal stability is expected for the hybrid matrix. Campoy and coworkers [12] studied the thermal stability of blends of Vectra A950 and nylon 6. They found that the thermal stability of Vectra A950 was much better than nylon 6 and it steadily improved with the addition of LCP in the blends.

### Dynamic mechanical properties

Figure 4 illustrates the variation of storage modulus with temperature for the 20 wt% glass fiber reinforced toughened nylon 6,6 and the hybrid composites. The hybrid composition containing 20 wt% LCP content exhibits the best storage modulus. It is interesting to note that inclusion of 5 and 10 wt% LCP into glass fiber reinforced nylon has a negative effect on the stiffness of hybrid composites. These variations in the storage modulus can be ascribed to the changes in the morphological features with increasing LCP content to be discussed later.

### Mechanical Properties

Figures 5a and b show the plots of tensile strength and modulus of the hybrid composite vs. LCP content. The trends in strength and stiffness vs. LCP content appear to be similar. With an addition of 5 wt% LCP there is a slight drop in the strength and stiffness. At 20 wt%LCP a rapid rise in the tensile strength and modulus is noted. Comparing the tensile strengths of the unreinforced, glass fiber reinforced and LCP containing hybrid material (Fig. 6) the tensile strength of 20 wt%

LCP hybrid composite (70 MPa) is higher than that of a rubber toughened nylon 6,6 reinforced with 30 wt% glass fiber (67 MPa). Clearly presence of LCP not only provided good processability but also a strengthening role in the hybrid system.

Figure 7 shows the skin regions of tensile fracture surface of injection molded LCP hybrid composites containing 10 and 20 wt% LCP. The size and nature of the LCP domains change with the LCP content. In the 10 wt% composition (Fig 7a) LCP phase appears in the form of spherical aggregates. The skin region of 20 wt% hybrid composite (Fig. 7b) displays some thicker fibrils and predominantly oriented sheet like structures. These oriented structures contribute to the enhancement in strength and stiffness by means of their higher effective aspect ratio. It is believed that the lack of fibrillation or orientation curtails the improvement in strength and stiffness until 20 wt% LCP is reached. The highest stiffness exhibited by the 20 wt% LCP composition in DMA can also be ascribed to this oriented sheet like morphological feature of the LCP phase. The lowest stiffness of the 10 wt% LCP composition is due to ineffective stress transfer and the existence of the LCP as spherical domains. Thus DMA results reflect the evolution of the morphology of hybrid composites with increasing LCP content.

Future work will focus on the role of elastomer and micromorphology on the fracture toughness and synergistic performance in the hybrid composite.

### Conclusions

Based on the results presented, it was concluded that inclusion of a small amount of LCP into glass fiber reinforced toughened nylon 6,6 significantly improved the processability of fiber reinforced toughened thermoplastics. The hybrid composites so obtained exhibited better thermal stability than the glass fiber reinforced polymers alone. Other than enhanced processability, LCP addition also played a strengthening role. The strength and stiffness were primarily derived from the oriented sheet like structures in the load bearing direction. At 20 wt% LCP, the hybrid composite exhibited the best combination of processability and mechanical properties.

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KEYWORDS: Hybrid, LCP, composite, processability

Composition (PA6,6/GF/LCP)	Degradation temperature T <sub>d</sub> (°C)	Residue at 450°C (%)
80/20/0	382.59	42.8
65/20/15	397.06	63.8
60/20/20	400	64.5

Table 1 TGA results of hybrid composites

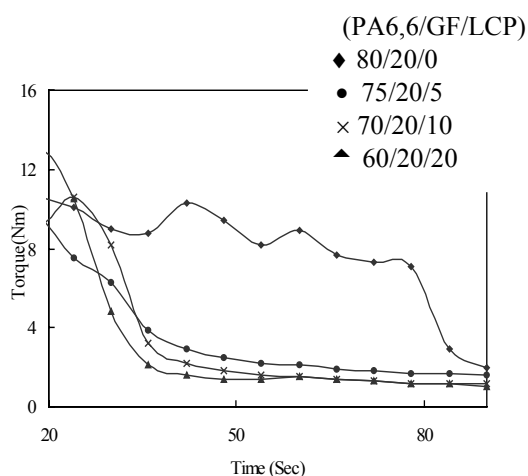


Figure 1 Torque versus time profiles for hybrid composites

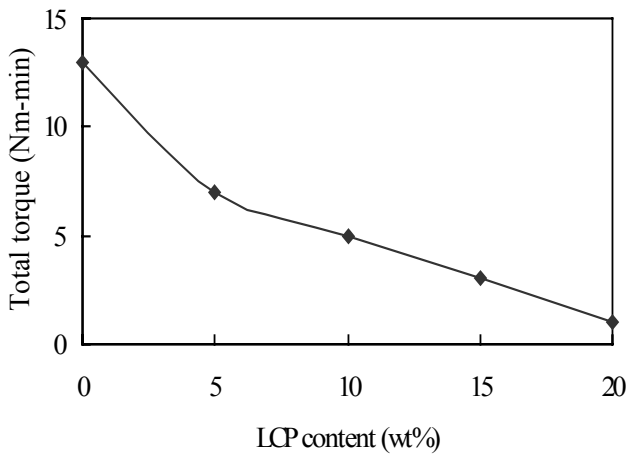
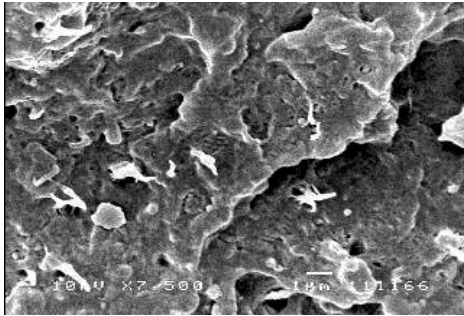
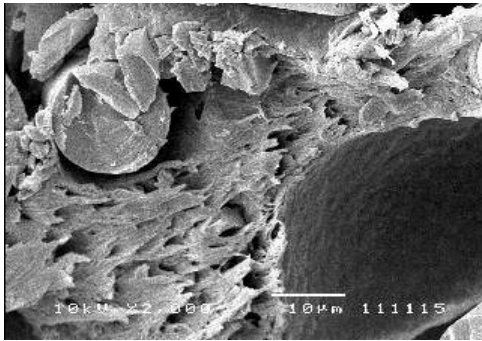


Figure 2 Total torque versus LCP content



(a) PA6,6/GF/LCP(75/20/5)



(b) PA6,6/GF/LCP(80/20/20)

Figure 3 Extrudate morphologies of nylon/glass fiber/LCP

hybrid composites

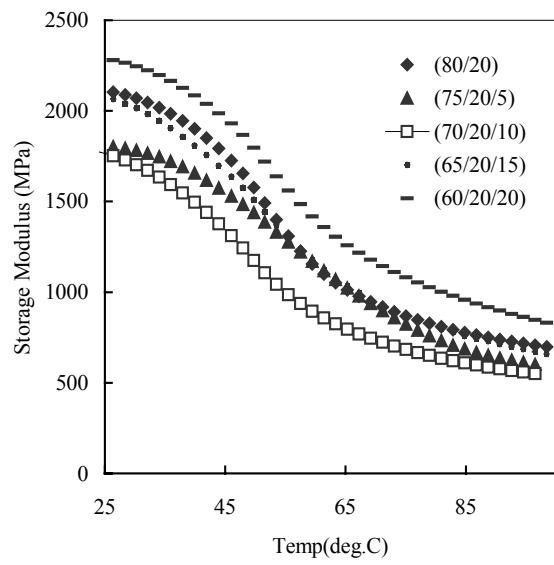


Figure 4 Storage modulus versus temperature for hybrid composites

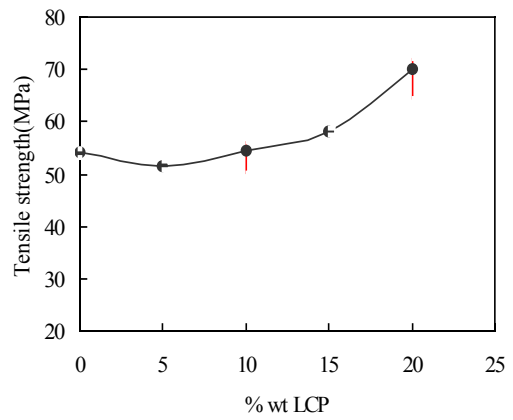


Figure 5a Tensile Strength variation with LCP content

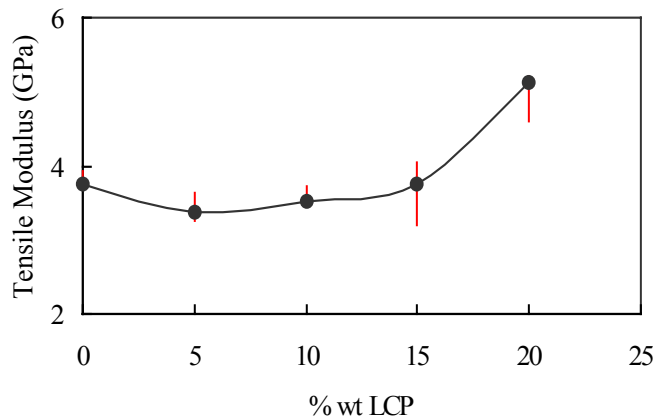
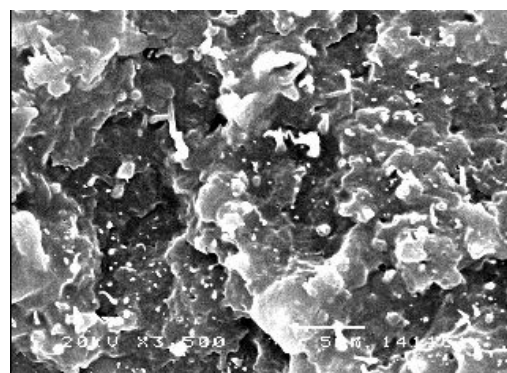
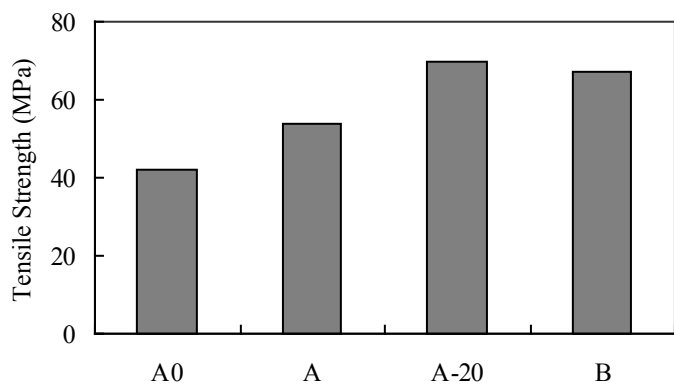


Figure 5b Tensile Modulus variation with LCP content



(a) PA 6,6/GF/LCP (70/20/10) (Skin)



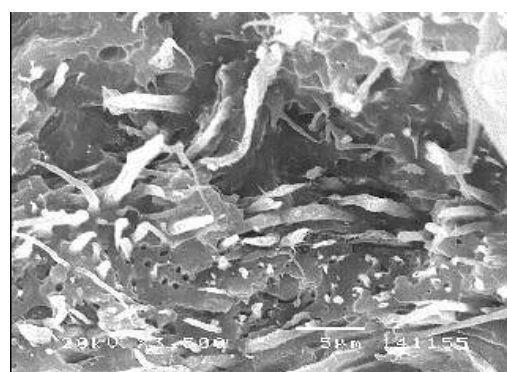
A0: PA6, 6/GF/LCP (100/0/0)

A: PA6, 6/GF/LCP (80/20)

A-20: PA6, 6/GF/LCP (60/20/20)

B: PA6, 6/GF/LCP (70/30)

Figure 6 Comparison of tensile strength for PA6,6/GF/LCP composites



(b) PA 6,6/GF/LCP (60/20/20) (Skin)

Figure 7 Tensile fracture surfaces of hybrid composites