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Compatibilization of thermoplastic composites based on blends of polypropylene with two liquid crystalline polymers

Arindam Datta* and Donald G. Baird
Polymer Materials and Interfaces Laboratory and Department of Chemical Engineering, Virginia Polytechnic Institute and State University, Blacksburg, VA 24061, USA
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In an earlier paper we considered the effect of polypropylene modified with maleic anhydride (MAGPP) on the properties of *in situ* composites based on polypropylene (PP) and a thermotropic copolyesteramide (Vectra B). In this paper we consider the effect of MAGPP on the properties of *in situ* composites based on PP and two different thermotropic copolyesters. It was observed that the MAGPP was nearly as effective in improving the properties for the copolyester systems as it was for the copolyesteramide system. The addition of MAGPP leads to finer and better dispersed fibrils of liquid crystalline polymer relative to the blends with no MAGPP added. These results were somewhat unexpected based on earlier reports which suggested that compatibility would be detrimental to the formation of fibrils.

(Keywords: compatibility; liquid crystalline polymer; polypropylene)

INTRODUCTION

There has been considerable interest in recent years in composites based on blends of liquid crystalline polymers (LCPs) with thermoplastics[1–14]. During processing, drops of the LCP form oriented fibrils, leading to the *in situ* reinforcement of the matrix. The formation of the oriented fibrils depends on the presence of strong extensional flows such as those that exist at the advancing front during mould filling. The morphology of the blends and the resulting physical properties are influenced by the interaction between the matrix polymer and the LCPs. It has been claimed that compatibility between the LCP and the matrix may be detrimental to fibril formation, and hence the reinforcing effect could be lost[7,15,16]. This is due to the fact that if polymers are too compatible, then the LCP will form drops which are too finely dispersed to deform into fibrils. On the other hand, just as in the case of blends of two flexible chain polymers, the complete lack of compatibility could lead to the deterioration of properties such as strength for the LCP/thermoplastic blends, even if fibrils are present. Hence, there is a question about the role of compatibilization in the case of composites based on blends of LCPs and thermoplastics.

The focus of this investigation is on incompatible blends of polypropylene with LCPs. We[17,18], as well as other researchers[19,20], have reported reinforcement of PP with several LCPs using various processing techniques such as extrusion, injection moulding and blow moulding. The incompatibility between the matrix polymer and reinforcing LCPs gives rise to poor interfacial adhesion and non-uniform dispersion of the LCP phase. Although showing some improvements in moduli values, these composites show little or no improvement, and in some cases a decrease, in tensile strength values compared with pure PP. The PP/LCP blends also tend to have a poor surface finish.

In an earlier paper[21], we reported the effect of maleic anhydride-*grafted*-polypropylene (MAGPP) on the properties and morphology of blends of polypropylene with Vectra B950, a poly(ester-*co*-amide) based on terephthalic acid, hydroxynaphthoic acid and aminophenol. It was found that the modulus and strength of injection moulded plaques were significantly increased in both machine and transverse directions in the presence of MAGPP. For example, the modulus of the blends containing 30 and 50 wt% Vectra B950 were 37 and 27% higher, respectively, in the presence of MAGPP. On the other hand, the strength of both the blends containing 30 and 50 wt% Vectra B950 was 100% higher when MAGPP was used. Improvements were also observed for transverse strength and modulus on addition of MAGPP. The LCP fibrils formed at the advancing front during injection moulding were found to be much finer and better dispersed in the presence of MAGPP. The surface of the injection moulded samples containing MAGPP was extremely smooth relative to those containing no MAGPP. It has been reported that compatibilizing agents, such as graft copolymers, act as polymeric surfactants, thereby reducing the interfacial tension which promotes interfacial adhesion, a finer and a more uniform distribution of the dispersed phase, and improved properties in blends of other incompatible polymers[22]. Thus the improved

* To whom correspondence should be addressed. Current address: Ethicon Inc., PO Box 151, Somerville, NJ 08876, USA

properties, especially the machine and the transverse tensile strengths, and the finer fibrils which were observed when MAGPP was used, suggested that there may be a reaction between the maleic anhydride and the LCP to form graft copolymers. These serve to act as a compatibilizing agent by reducing interfacial surface tension and promoting adhesion between PP and the LCP.

Based on the knowledge of the effect of MAGPP on blends of Nylon 6 and PP and the claim that compatibility would be detrimental to the formation of LCP fibrils, it was somewhat surprising that MAGPP was so effective in promoting improved properties and finer fibrils in the case of blends of PP and Vectra B950. For example, Ide and Hasegawa[23] reported that for blends of Nylon 6 and PP in the ratio 2/8 prepared in a single-screw extruder, the drop size decreased in the presence of MAGPP from 10 μm to less than 1 μm. A reduction in drop size of this magnitude would be detrimental to the formation of fibrils in the case of LCPs, as it would be more difficult to deform the smaller drop into a fibril. In the case of flexible chain polymers the goal of compatibilization is always to disperse the minor phase into the finest drops possible[22-24]. However, as recognized by others in the case of LCPs, it may be detrimental to fibre formation if the matrix and LCP are too compatible[7,15]. Furthermore, MAGPP may degrade at the temperature required to process most LCPs, and therefore the graft may not take place. However, indications based on the improved tensile strength were that some interactions, probably a reaction, may have occurred between the maleic anhydride and the amino end-groups of Vectra B950. Although it is not certain as to whether graft copolymers actually form, it is clear that the presence of MAGPP promotes significant improvements in properties in the case of Vectra B950 and PP in spite of claims that compatibility between the LCP and matrix would be undesirable.

This paper represents a continuation of our studies concerned with the effect of MAGPP on the properties and morphology of composites based on LCPs and polypropylene. In particular, it is our goal to determine whether MAGPP has a similar effect when the LCPs are copolyesters rather than a poly(ester-*co*-amide). In other words, when LCPs with ester end-groups alone are present, will one obtain similar effects as when amide and ester end-groups are present? To fulfil this goal the physical properties and the morphology of injection moulded plaques and tensile bars will be determined for blends of two different copolyesters with PP.

EXPERIMENTAL

Materials

Two LCPs were used for this study. The first LCP is known commercially as Vectra A900 and was purchased from Hoechst Celanese. It is a copolyester of 73 mol% of hydroxybenzoic acid (HBA) and 23 mol% of hydroxynaphthoic acid (HNA). The second LCP, LC3000, is a copolyester of 60 mol% of HBA and 40 mol% of poly(ethylene terephthalate) (PET) and was supplied by Unitika Corporation. The matrix PP, Profax 6823, was purchased from Himont. A maleic anhydride-*grafted*-PP (MAGPP) was used to promote compatibility between PP and the LCPs. The matrix, PP, is pre-blended with MAGPP, the amount of MAGPP used being 10% based on the matrix PP. The exact nature of MAGPP as well as the method of sample preparation is described elsewhere[25].

Injection moulding

The LCP pellets were dried for at least 24 h in a convection oven at 120°C before processing. For making blends without MAGPP, the dried pellets were then tumbled together with pellets of PP in predetermined weight ratios for making blended injection moulded plaques and tensile bars. PP/LCP plaques and tensile bars containing MAGPP were moulded in a similar fashion except that the dried LCP pellets were tumbled together with pellets of PP containing MAGPP instead of pure PP. Sample preparation involved pre-blending of the MAGPP and the matrix PP and, as mentioned above, has been described elsewhere[25]. Blends with and without MAGPP were made containing 20, 30, 50 and 80 wt% LCP.

Rectangular plaques of approximately 75 mm by 80 mm by 1.75 mm thick and tensile bars of approximately 63 mm by 1 mm by 1.7 mm thick were injection moulded using an Arburg model 222-55-20. For processing PP/Vectra A blends with and without MAGPP, the barrel temperatures of the extruder in the injection moulder were 230°C in the first zone and 295°C in the second and third zones. The nozzle temperature was 250°C. The barrel temperatures for processing PP/LC3000 blends with and without MAGPP were 200°C, 280°C and 270°C in the first, second and third zone, respectively. For processing both types of blends, the mould was kept at room temperature.

Mechanical properties

The tensile modulus and strength of the injection moulded plaques and tensile bars were measured using an Instron mechanical tester (model 4204). For the plaques, test samples were strips approximately 80 mm long and 12.5 mm wide. The crosshead speed was 2 mm min^{-1}. The strain was measured using an extensometer (Instron model 2630-25) which can measure a maximum strain of 10%. Toughness measurements were made using the tensile bars, toughness being calculated as the area under the stress–strain curve until the sample broke. The arithmetic average and the standard deviation of the tensile properties were calculated using a minimum of five samples.

The dynamic torsional properties of the plaques were obtained using a Rheometrics Mechanical Spectrometer, RMS 800. In particular, the storage modulus (G') was obtained as a function of temperature using an angular frequency of 10 rad s^{-1} and a strain of 0.05 to 0.07%. The dynamic creep compliance was also measured as a function of frequency at a strain of 0.05 to 0.1% at 30 and 100°C. For both tests, the sample was a rectangular piece measuring approximately 50 mm long, 12.5 mm wide, and 1.1 to 1.6 mm thick.

Morphological studies

The morphology was determined by scanning electron microscopy (SEM) using a Cambridge Stereoscan S200 with an accelerating voltage of 25 kV. The samples were fractured after immersing them in liquid nitrogen for 5 min. The fractured samples were then placed on

Table 1 Tensile properties[a] of PP/Vectra A blends obtained from plaques with and without MAGPP

Material[b]	Machine direction		Transverse direction	
	Modulus (GPa)	Strength (MPa)	Modulus (GPa)	Strength (MPa)
PP	1.384 (0.075)	26.89 (1.10)	1.098 (0.027)	25.76 (0.34)
PP/Vectra A 8/2	2.555 (0.216)	27.70 (1.06)	1.178 (0.104)	16.28 (0.68)
PP/Vectra A 8/2 (MAGPP)	3.059 (0.185)	33.23 (0.70)	1.681 (0.420)	20.99 (1.50)
PP/Vectra A 7/3	2.868 (0.280)	28.26 (1.15)	1.138 (0.078)	9.90 (0.38)
PP/Vectra A 7/3 (MAGPP)	3.791 (0.299)	34.45 (1.23)	1.536 (0.084)	16.13 (0.05)
PP/Vectra A 5/5	4.523 (0.269)	26.43 (0.48)	1.084 (0.27)	8.61 (0.32)
PP/Vectra A 5/5 (MAGPP)	5.205 (0.434)	40.69 (2.50)	1.567 (0.081)	16.97 (0.75)
PP/Vectra A 2/8	5.805 (0.343)	40.50 (2.83)	1.243 (0.069)	11.77 (0.47)
PP/Vectra A 2/8 (MAGPP)	7.719 (0.341)	45.91 (2.47)	1.850 (0.041)	20.81 (1.03)
Vectra A	9.312 (0.220)	140.0 (3.50)	1.478 (0.50)	23.52 (1.67)

[a] Standard deviations are given in parentheses
[b] (MAGPP) is blend containing MAGPP

aluminium stubs and sputter-coated with gold using a sputter coater.

RESULTS AND DISCUSSION

Properties of PP/Vectra A blends

The physical properties of PP and Vectra A injection moulded plaques and tensile bars with and without MAGPP will be presented first. The difference, if any, between the physical properties of the blends with and without compatibilization will be related to any observed difference in their respective morphologies. This will be followed by a similar comparison of the physical properties of PP and LC3000 blends with and without the addition of MAGPP.

The tensile properties of plaques with and without MAGPP for different blend compositions are presented in *Table 1*. The addition of LCP enhanced the tensile moduli for the blends, with the moduli increasing with the increase in LCP content. However, on addition of MAGPP the blends showed an additional 15 to 35% increase in modulus. The tensile strength of the blends showed virtually no improvement over that of pure PP for blends containing up to 50 wt% LCP. The tensile strengths of the blends with MAGPP increased significantly over those of the blends without MAGPP and that of pure PP. Overall, the tensile strengths of the blends in the presence of MAGPP increased by 1.2 to 1.7 times that of PP, and the tensile moduli increased by 2.2 to 5.6 times that of PP, as the LCP content in the blend was increased from 20 to 80%.

The tensile properties of injection moulded PP/Vectra A tensile bars with and without the addition of MAGPP are reported in *Table 2*. The properties of the tensile bars are observed to be higher than those obtained from PP/Vectra A plaques for the same blend composition, except for the modulus of the blend containing 20 wt% LCP. The higher properties of the tensile bar specimens can be ascribed to the stronger elongational flow in the neck region of the tensile bars, which leads to higher orientation of the LCP phase. As with the plaques, addition of MAGPP enhances both the moduli and strengths of the tensile bars for all blend compositions. The levels of enhancement vary from 15 to 60% for the moduli and from 20 to 60% for the strengths. This results in 2.0 to 10.4 times increase in moduli and 1.2 to 3.9 times increase in strength over those of PP as the LCP content is increased from 20 to 80 wt% for blends containing MAGPP.

The measured modulus of the blends at different blend compositions can be compared with the predictions based on composite theory. According to this theory, the modulus of unidirectional fibre reinforced composites may be given by the weighted average of the fibre and matrix moduli, with the weighting factor being their respective volume fractions[4,7,12]. This is known as the rule of mixtures. It has been derived under certain assumptions, such as the aspect ratio of the fibre being very large, a condition that is not necessarily true for all the reinforcing LCP fibrils in the cases described above. The compositional dependence of the plaques and the tensile bars of PP/Vectra A blends with MAGPP is shown in *Figure 1*, in which the modulus is plotted against weight percentage rather than volume percentage of LCP. The lower and upper bounds of the modulus at 0 and 100 wt% LCP are the experimentally determined values of PP and Vectra A, respectively. The predictions from the rule of mixtures will lie on the straight line joining the upper and lower bounds of the modulus. It is observed that for plaques, even with addition of MAGPP, the moduli were below those values predicted by the rule of mixtures. However, for tensile bars, the blends with MAGPP containing 50 and 80 wt% LCP exhibit higher moduli than those predicted by the rule of mixtures and

Table 2 Tensile properties[a] of PP/Vectra A blends obtained from tensile bars with and without MAGPP

Material[b]	Modulus (GPa)	Strength (MPa)	Elongation at yield (%)	Toughness ($kJ\,m^{-3}$)
PP	1.369 (0.061)	31.24 (0.24)		
PP/Vectra A 8/2	2.159 (0.197)	31.06 (1.21)	2.76	618[c]
PP/Vectra A 8/2 (MAGPP)	2.695 (0.216)	37.49 (2.29)	2.77	800[c]
PP/Vectra A 7/3	3.759 (0.379)	36.61 (2.91)	1.64	742[c]
PP/Vectra A 7/3 (MAGPP)	4.255 (0.327)	49.88 (1.81)	1.92	724
PP/Vectra A 5/5	6.158 (0.312)	45.71 (3.07)	1.50	1071[c]
PP/Vectra A 5/5 (MAGPP)	7.653 (0.957)	71.86 (4.90)	1.50	706
PP/Vectra A 2/8	8.992 (0.704)	74.40 (6.66)	1.73	1483
PP/Vectra A 2/8 (MAGPP)	14.19 (1.07)	120.57 (10.22)	1.73	1437
Vectra A	10.992 (0.984)	234.00 (17.30)	5.28	3220

[a] Standard deviations are given in parentheses
[b] (MAGPP) is blend containing MAGPP
[c] toughness until 3%

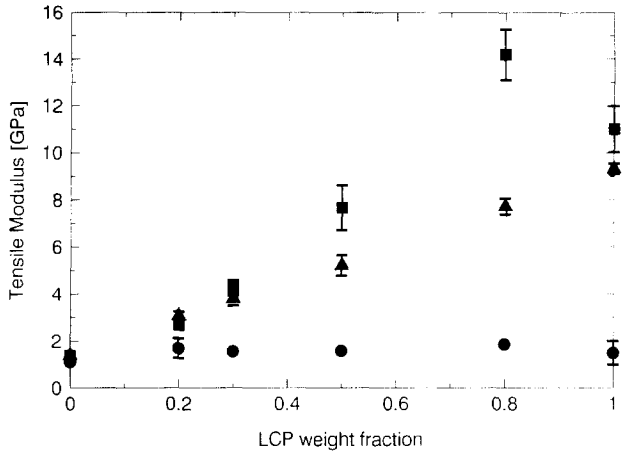

Figure 1 Tensile modulus *versus* LCP content of PP/Vectra A blends with MAGPP: ■, tensile bars; ▲, plaques (machine direction); ●, plaques (transverse direction)

equal that predicted by the rule of mixtures for a blend containing 30 wt% LCP. It is interesting to note that the modulus of the compatibilized PP/Vectra A 2/8 blend is higher than that of Vectra A.

Anisotropy in the mechanical properties of the blends is observed from the transverse direction tensile properties of the plaques reported in *Table 1*. The poor transverse properties of the blends arise from the highly anisotropic nature of Vectra A, which is typical of LCPs[7,23]. Addition of MAGPP improved the transverse direction moduli of the blends by 35 to 49% and transverse direction strengths by 29 to 97% over those of blends with the same LCP content but not containing MAGPP. Still, the transverse direction strengths of the blends with MAGPP were lower than those of PP.

Other tensile properties such as ductility and toughness of the blends were obtained from the stress–strain curves of the PP/Vectra A blend tensile bars and are also listed in *Table 2*. PP is very ductile with an elongation to break which is several orders of magnitude higher than that of the blends and has a significantly higher toughness than that of the blends. In general, the addition of LCP changes the material response from ductile to brittle. The elongation at yield decreases substantially with the increase in the LCP content but tends to go up slightly as the LCP concentration is increased from 50 to 80 wt%. The presence of MAGPP did not alter the values of elongation at yield. Although the PP/Vectra A samples containing up to 50 wt% LCP undergo elongation of over 3% before break, most PP/Vectra A samples containing MAGPP broke below 3% strain without displaying any significant ductility after the yield point. Thus, for conparison purposes, toughness was calculated until 3% strain. Addition of MAGPP enhances the toughness at 20 wt% LCP while the toughnesses of the blends at 80 wt% LCP with and without addition of MAGPP are nearly identical. For the intermediate blend compositions of 30 and 50 wt% LCP, the elongation to break drops below 3% on addition of MAGPP with consequent drops in toughness.

The variation of dynamic storage modulus (G') with temperature and the dynamic creep compliance (J') for PP and the various PP/Vectra A blends has been studied using the RMS and results are presented in *Figures 2, 3* and *4*, respectively. It was observed (although the data are not shown) that the dynamic modulus increased on addition of MAGPP at the lower temperatures for all blend compositions, but the differences were indistinguishable at higher temperatures. There were no significant differences seen in the compliances of blends on

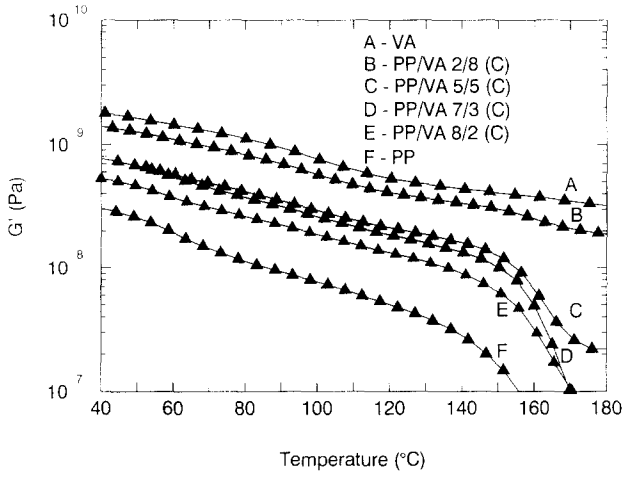

Figure 2 Torsion modulus *versus* temperature of PP, Vectra A and PP/Vectra A blends with MAGPP

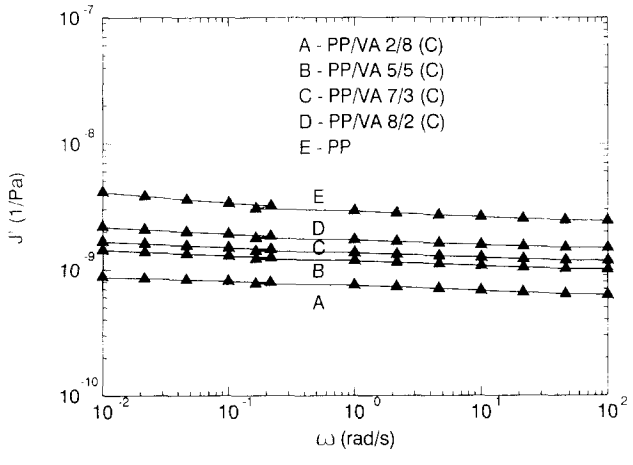

Figure 3 Dynamic creep compliance *versus* frequency of PP and PP/Vectra A blends with MAGPP at 30°C

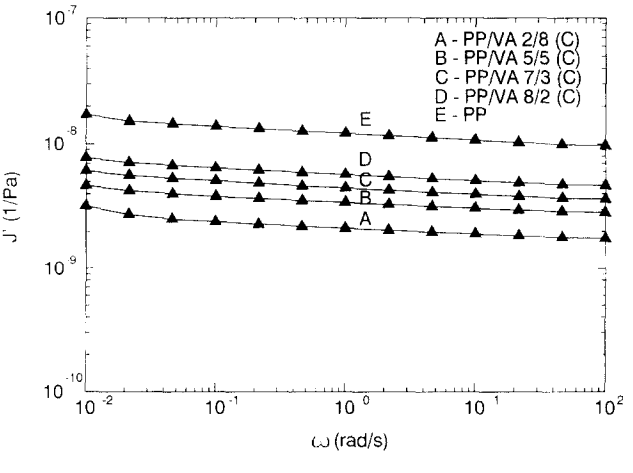

Figure 4 Dynamic creep compliance *versus* frequency of PP and PP/Vectra A blends with MAGPP at 100°C

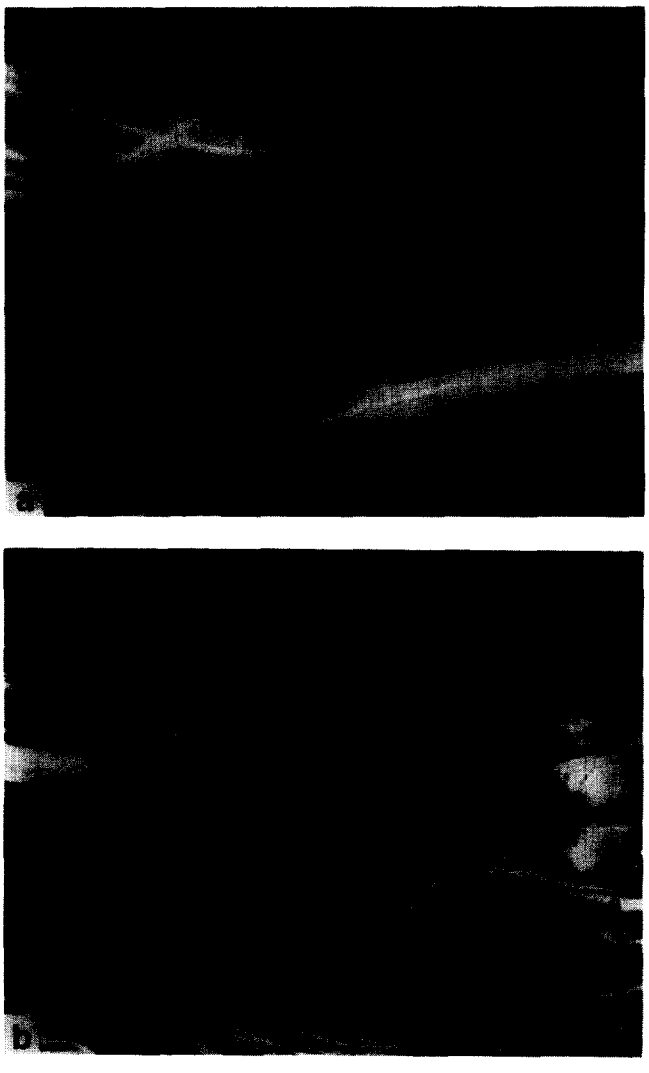

Figure 5 Scanning electron micrographs of fracture surfaces of PP/Vectra A 8/2 (a) and PP/Vectra A 8/2 with MAGPP (b) plaques. Fracture is along the flow direction

addition of MAGPP. Only G' and J' of the blends containing MAGPP (represented by (C) in the figures) are presented here to avoid putting too many results on the same plot. As observed in *Figure 2*, the torsion moduli of the PP/Vectra A blends containing MAGPP increased with increasing LCP content, and significant improvements are observed for the entire temperature range scanned. The torsion modulus of PP decreased rapidly as the temperature approached its melting point of 165°C. However, owing to the presence of the reinforcing LCP fibrils, the drop in G' of blends occurred at a slower rate than that of PP. The differences in G' between the blends and PP are greater at higher temperatures than at lower temperatures, and the temperature at which G' of the blends started to fall was shifted to higher temperatures as the LCP content was raised. These two phenomena were expected to make these blends more suitable for applications at elevated temperatures than is PP.

The dimensional stability of the blends under load was examined by monitoring the dynamic compliance (J') in frequency sweeps at different temperatures. The compliance data for the blends in the presence of MAGPP and PP are presented in *Figures 3* and *4* at 30°C and 100°C, respectively. At 30°C the compliance improved significantly with the increase in LCP content. At 100°C, the compliance of PP is almost five times higher than its compliance at 30°C. However, the compliance values at 100°C for all the blend compositions increase by approximately only three times over their values at 30°C. The results suggest the effectiveness of the LCP reinforcement in improving the dimensional stability of the blends under load at elevated temperatures.

Morphology of PP/Vectra A blends

The effect of MAGPP on the morphology of the blends was analysed via SEM in an effort to understand its effect on the properties of the composites. The fracture surfaces of PP/Vectra A 8/2 blends with and without MAGPP are shown in *Figure 5*. The samples were fractured along the flow direction which is horizontal in the figure. The presence of LCP fibrils oriented along the flow direction are clearly observed in blends irrespective of whether or not they contain MAGPP, but the morphologies differ in certain respects. The LCP fibrils are better dispersed in the presence of MAGPP while they appear to be bunched together in blends made without MAGPP. In the former case, the LCP fibrils are finer in size. Also in the case of blends with MAGPP, the LCP fibrils adhere better to the PP matrix, resulting in failure occurring within the fibrils during fracture of the samples. For the blends not containing MAGPP, whole fibrils are pulled out during fracture of the samples, indicating poor adhesion between the two phases. While the finer LCP fibrils have possibly resulted in higher moduli, the improved tensile strength must be attributed to better adhesion between the phases. The addition of MAGPP leads to different results in blends of flexible chain polymers[22–24]. Similar morphological changes have been reported by us for blends of PP and Vectra B on addition of MAGPP[21]. Although it has been claimed that greater compatibility may be detrimental to fibril formation[7,15] and consequently may affect the reinforcing capabilities of the LCP, the results presented here indicate that there is a significant increase in physical properties, especially tensile strength, owing to the presence of MAGPP. However, it is not clear whether any graft copolymers were actually formed as is the case with addition of MAGPP to blends of flexible chain polymers[22,23].

The presence or absence of MAGPP gives rise to similar differences in morphologies for the PP/Vectra A blends containing 50 wt% LCP. As seen in *Figure 6*, the LCP phase seems to be forming interconnected sheets in the PP/Vectra A 5/5 blend when MAGPP is not present. In contrast, when MAGPP is added, the compatibilized PP/Vectra A 5/5 blend exhibits more finely dispersed fibrils. It can be speculated that the tendency of the LCP to coalesce is reduced by the emulsifying effect brought about by the addition of MAGPP. Furthermore, the blends in the presence of MAGPP demonstrate better adhesion as a number of LCP fibrils are seen to have resisted being pulled out and break during the fracture process while remaining adhered to the matrix. The effect of addition of MAGPP on the morphology of the PP/Vectra A 2/8 blend is difficult to distinguish, as seen in *Figure 7*. However, it appears that the LCP is somewhat better dispersed in the presence of MAGPP. As with the PP/Vectra A 8/2 blend, the addition of MAGPP to blends with higher LCP content appears to

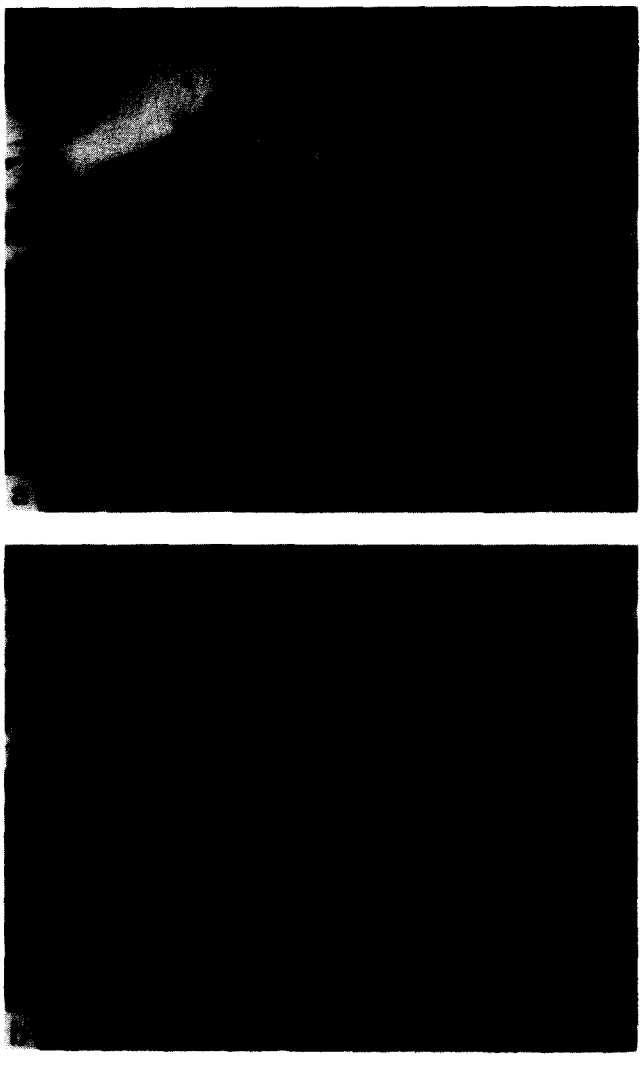

Figure 6 Scanning electron micrographs of fracture surfaces of PP/Vectra A 5/5 (a) and PP/Vectra A 5/5 with MAGPP (b) plaques. Fracture is along the flow direction

promote greater compatibility but at the same time produces enhancements in the mechanical properties which are contrary to what would have been expected based on previous work[7,15].

Properties of PP/LC3000 blends

Having established the advantages of the presence of MAGPP for blends of PP and Vectra A, we now consider its effect on blends of PP with another copolyester, LC3000. As with PP/Vectra A blends, the effect of MAGPP addition was investigated by comparing the mechanical properties of the blends with and without MAGPP.

The tensile properties of PP/LC3000 blends with and without the addition of MAGPP are summarized in *Table 3*. The reinforcing effect of the LCP on the tensile modulus was observed for both plaques and tensile bars at all of the LCP composition levels tested here. The tensile bars have higher moduli than the plaques, which can be attributed to the stronger elongational flow in the tensile bar mould. The moduli increased in the presence of MAGPP for all blend compositions for both plaques and tensile bars. With addition of MAGPP, the moduli of the plaques increased from 2.1 to 8.1 times that of PP as the level of LCP increased from 20 to 80 wt%. For the same increase in LCP content for the tensile bars containing MAGPP, the corresponding increase in moduli ranged from 2.6 to 10 times that of PP.

The changes in modulus with LCP content of plaques and tensile bars of PP/LC3000 blends in the presence of MAGPP are shown in *Figure 8*. The moduli of PP/LC3000 plaques, for all compositions, and tensile bars, with 20 wt% LCP, were below those predicted by the rule of mixtures. With MAGPP, the modulus of tensile bars at 50 wt% LCP is equal to, and at 80 wt% LCP is slightly above, the values predicted by the rule of mixtures.

As with the modulus, the tensile strengths of the PP/LC3000 blends follow trends similar to those of PP/Vectra A blends, as reported in *Table 3*. The tensile strengths of the plaques in the absence of MAGPP are below those of PP for LCP levels as high as 50 wt%, but on incorporation of MAGPP in the blend there are significant improvements in tensile strength, the values of which exceed that of the matrix at all compositions. The tensile strengths of the tensile bars follow a similar trend in that at low LCP concentration (20 wt%) the values are lower than that of PP but on addition of MAGPP there are significant improvements in the tensile

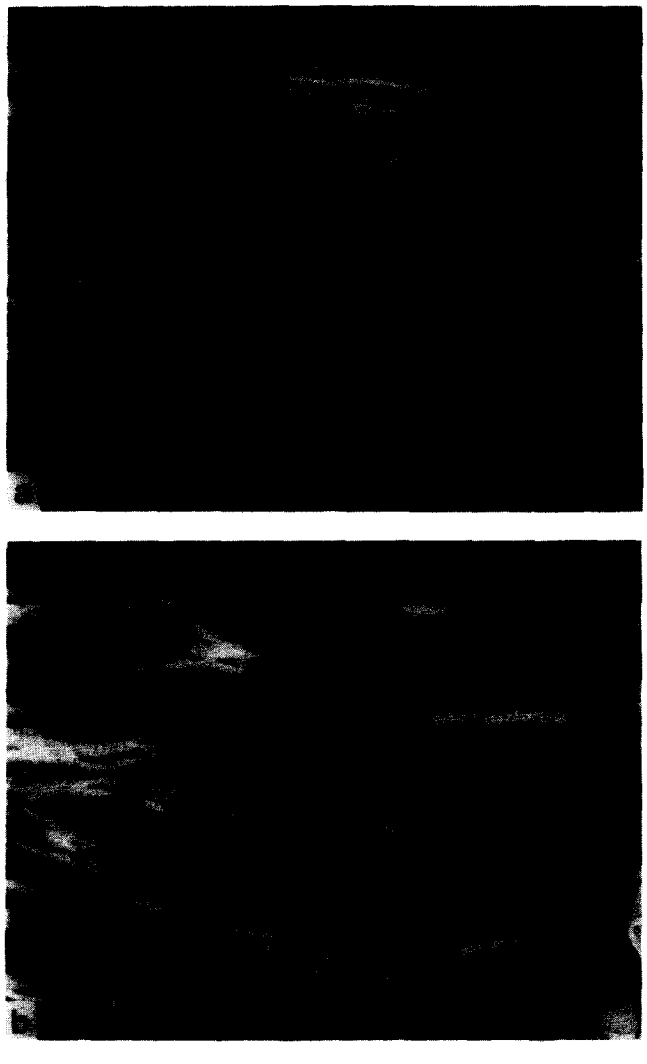

Figure 7 Scanning electron micrographs of fracture surfaces of PP/Vectra A 2/8 (a) and PP/Vectra A 2/8 with MAGPP (b) plaques. Fracture is along the flow direction

Table 3 Tensile properties[a] of PP/LC3000 blends obtained from plaques and tensile bars with and without MAGPP

Material[b]	Plaques		Tensile bars			
	Modulus (GPa)	Strength (MPa)	Modulus (GPa)	Strength (MPa)	Toughness ($kJ\,m^{-3}$)	Elongation at yield (%)
PP	1.384	26.89	1.369	31.24		
	(0.075)	(1.10)	(0.061)	(0.24)		
PP/LC3000 8/2	1.627	18.89	2.137	28.66	608[c]	2.14
	(0.047)	(0.60)	(0.094)	(1.79)		
PP/LC3000 8/2 (MAGPP)	2.896	30.21	3.589	41.19	826[c]	2.61
	(0.091)	(1.43)	(0.203)	(1.32)		
PP/LC3000 5/5	4.940	23.53	7.117	44.52	275	1.02
	(0.284)	(1.31)	(0.575)	(1.82)		
PP/LC3000 5/5 (MAGPP)	5.839	46.28	8.314	68.26	723	1.63
	(0.203)	(1.69)	(0.318)	(0.83)		
PP/LC3000 2/8	10.222	50.12	12.696	85.76	868	1.33
	(0.308)	(2.8)	(0.512)	(2.54)		
PP/LC3000 2/8 (MAGPP)	11.189	65.09	13.66	107.45	1259	1.72
	(0.440)	(1.02)	(0.406)	(4.13)		
LC3000			15.053	142.63	1838	1.82
			(0.518)	(1.21)		

[a] Standard deviations are given in parentheses
[b] (MAGPP) is blend containing MAGPP
[c] Toughness until 3%

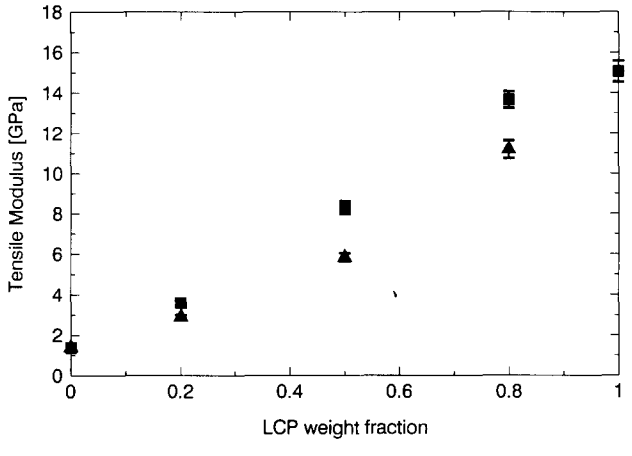

Figure 8 Tensile modulus *versus* LCP content of PP/LC3000 blends with MAGPP: ■, tensile bars; ▲, plaques (machine direction)

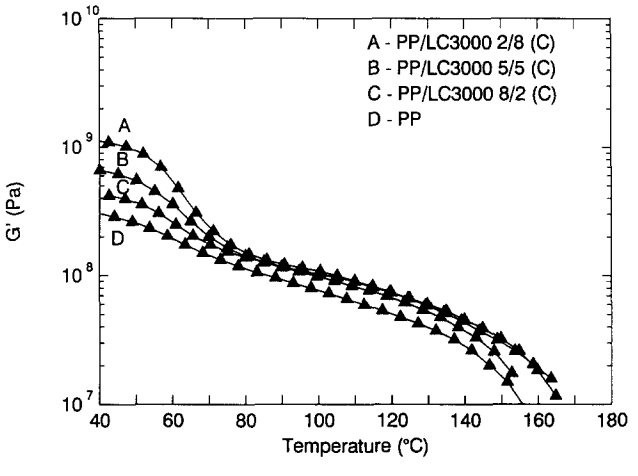

Figure 9 Torsion modulus *versus* temperature of PP and PP/LC3000 blends with MAGPP

strength values for all blend compositions. As the LCP content increases from 20 to 80 wt%, the tensile strengths of the plaques increase by 1.1 to 2.4 times that of PP. The strengths of the more oriented tensile bars are higher than those of the plaques and, for the same increase in LCP content, the corresponding increase in tensile strengths are 1.3 to 3.4 times that of PP.

Unlike PP/Vectra A blends, the toughness improved in the presence of MAGPP for all blend compositions. The toughness of the blends which do not contain MAGPP shows a minimum at 50 wt% LCP. The transition from 20 to 50 wt% LCP corresponds to elongation at break from greater than 3% to less than 3%. On addition of MAGPP, better adhesion develops and the blends approach the higher toughness of the LCP, especially at 80 wt% of LC3000.

In the light of the success in obtaining significant improvements in tensile properties of both PP/Vectra A and PP/LC3000 blends, it was interesting to investigate whether the dynamic properties also experience similar enhancements, especially at elevated temperatures. The dynamic torsional modulus *versus* temperature data of the PP/LC3000 blends in the presence of MAGPP are shown in *Figure 9*. The storage modulus increased with increased LCP level at lower temperatures. However, at about 55°C, G' starts to drop at a faster rate for blends with higher LCP content, and G' *versus* temperature curves converge to almost a single curve at approximately 80°C. Although the blends still maintain a higher G' value at elevated temperature, these PP/LC3000 composites are unlikely to have much of an additional advantage over PP when used in applications at elevated tempera-

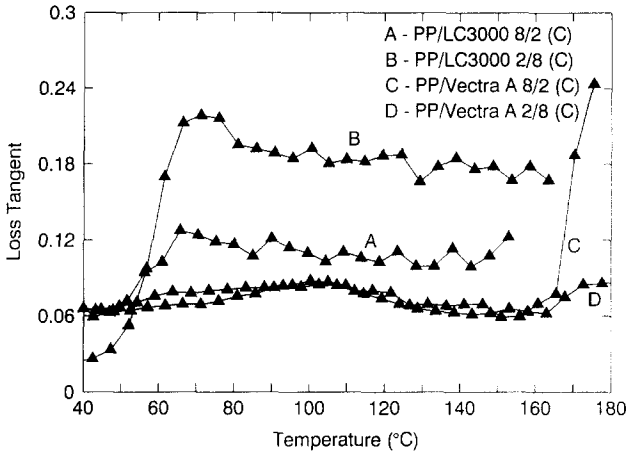

Figure 10 Loss tangent *versus* temperature of PP/LC3000 and PP/Vectra A blends with MAGPP

tures. The reason for the drop in G' for the blends at temperatures above 55°C is seen by examining the loss tangent ($\tan\delta$) *versus* temperature data in *Figure 10*. Vectra A in PP/Vectra A blends displays a transition at approximately 110°C. This transition temperature is similar to findings of other researchers[26] and is somewhat analogous to a glass transition temperature, T_g, for other traditional polymers. However, it is not associated with a large drop in elastic modulus. On the other hand, LC3000 blends clearly show a T_g with a large peak height for the loss tangent between 68 and 70°C. This T_g, corresponding to the PET-rich segment or domain, is consistent with published data[27] and causes the large drop at elevated temperatures observed for the G' *versus* temperature curves in *Figure 10*.

The dynamic compliance of PP/LC3000 blends containing MAGPP shows similar enhancements to that of PP/Vectra A blends in the presence of MAGPP at 30°C, as seen in *Figure 11*. The compliance values of the PP/LC3000 blends (not reported here) increase significantly at 100°C and are considerably higher than those of PP/Vectra A blends. This increase in compliance at 100°C can be attributed to a drop in modulus associated with the glass transition of the PET-rich domains of LC3000 at about 70°C, as mentioned before.

Comparison of blend systems

We next examine the similarities and differences between the properties of PP/Vectra A and PP/LC3000 blends with and without the addition of MAGPP. Obviously, the properties of the pure LCP will play a significant role in determining the properties of the blends. The modulus and strength of the Vectra A tensile bars are 11 GPa and 234 MPa, respectively, while the modulus and strength of LC3000 tensile bars are 15 GPa and 143 MPa, respectively. It is expected that the addition of LC3000 will lead to higher moduli and the addition of Vectra A will promote higher enhancements of strength. For blends with MAGPP this is not always true, as the effectiveness of the MAGPP at different levels of LCP seems to determine the final properties of the blends.

The properties of tensile bars of PP/Vectra A and PP/LC3000 blends at various compositions are compared to examine the effectiveness of MAGPP addition. For blends with 20 wt% LCP and containing no MAGPP, the modulus and strength are quite similar. On addition of MAGPP, the PP/LC3000 8/2 blend shows a 68% increase in modulus to 3.6 GPa and a 44% increase in strength to 41.2 MPa. However, the corresponding increase in modulus and strength for PP/Vectra A 8/2 blend in the presence of MAGPP is only 25% and 20%, respectively, to 2.7 GPa and 37.5 MPa. Clearly, at 20 wt% LCP, MAGPP is more effective in enhancing the properties of PP/LC3000 tensile bars. As the LCP level is increased to 50 wt% in blends without MAGPP, the modulus is slightly higher for PP/LC3000 blend, but the strengths are almost identical. The incorporation of MAGPP leads to a similar percentage rise in strength and therefore a slightly higher value for the PP/Vectra A blend due to the inherently higher strength of Vectra A. The modulus of the PP/LC3000 5/5 blend increased by 17% and that of PP/Vectra A 5/5 blend rose by 24% on addition of MAGPP. However, the modulus of 8.3 GPa for PP/LC3000 is equal to that predicted by the rule of mixtures while the modulus of 7.7 GPa for PP/Vectra A blend shows a positive deviation from the rule of mixtures. Unlike the blends with 20 wt% LCP, the effectiveness of MAGPP addition is slightly more pronounced for the PP/Vectra A blend at 50 wt% LCP. The effectiveness of MAGPP addition for the PP/Vectra A blend is further accentuated at even higher LCP content (e.g. 80 wt% LCP). The modulus of PP/Vectra A 2/8 blend exhibits a substantial positive deviation from the rule of mixtures so not only is the value of modulus higher than that of the PP/LC3000 2/8 blend, but it is also higher than that of Vectra A. The strength also shows a much higher increase for the PP/Vectra 2/8 blend than for the PP/LC3000 2/8 blend.

Comparison with Vectra B data

Finally, we compare the effect of MAGPP on the properties of PP and the copolyester blends relative to its effect on the properties of blends of PP and a copolyesteramide (Vectra B950). Without the addition of MAGPP, the properties of the composites are different for the three LCP systems because each LCP has different properties on injection moulding. For example, the modulus of Vectra B950 plaques is 17.2 GPa while that of Vectra A plaques is only 9.3 GPa. Hence, we can expect

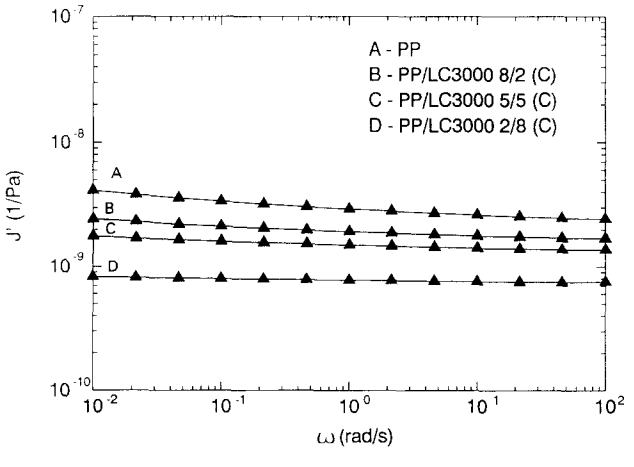

Figure 11 Dynamic creep compliance *versus* frequency of PP and PP/LC3000 blends with MAGPP at 30°C

Vectra B950 to provide more of an improvement in modulus of PP than Vectra A. For the sake of comparison, we choose compositions consisting of 70 wt% PP and 30 wt% LCP (70/30 ratio). Referring to the data in the previous paper[23] and the data in *Table 1*, we see that the machine direction modulus of 70/30 PP/Vectra B950 with and without MAGPP is higher than that of PP/Vectra A plaques. In both cases the machine direction modulus increases with addition of MAGPP, but the effect is greatest for PP/Vectra B950 plaques. In terms of machine direction strength, we observe that in the case of Vectra A it is slightly higher than that of PP and on addition of MAGPP there is a 22% increase in strength. On the other hand, in the case of Vectra B950 there is a decrease in machine direction strength relative to that of PP, but with addition of MAGPP the strength increases by 85%. We can make similar observations in terms of transverse direction properties. In this case we see that the Vectra B950 composites have the highest transverse strength and that MAGPP is more effective in improving the transverse direction tensile strength. Without going into great detail it appears that similar statements can be made with regard to the results for the PP/LC3000 composites. At this time we cannot determine whether the difference in the effect is due to a higher degree of reactivity of amide end-groups with maleic anhydride relative to ester end-groups, or whether there are more end-groups available in the case of Vectra B950, or whether there are any reactions at all. This will be dealt with in later publications.

CONCLUSIONS

There are considerable similarities among the effects of addition of MAGPP on the properties of blends of PP with two LCPs, Vectra A and LC3000, both of which are copolyesters. Similar enhancements in properties and changes in morphologies were reported earlier on addition of MAGPP to blends of PP with Vectra B950, which is a poly(ester-*co*-amide)[21]. The following conclusions can be drawn from the investigation regarding the properties of the two PP/LCP composites.

1. On addition of MAGPP, blends of PP and Vectra A exhibited significant enhancements in both tensile modulus and strength. The properties of tensile bars were higher than those of the plaques, both in the presence and absence of MAGPP, owing to the stronger elongational flow and the associated higher orientation in the tensile bar mould. For plaques containing MAGPP, as the LCP content is increased from 20 to 80 wt%, the modulus increased by 2.2 to 5.6 times whereas the strength increased by 1.2 to 1.7 times over that of PP. For tensile bars containing MAGPP, with the LCP content varying from 20 to 80 wt%, improvements in the modulus ranged from 2.0 to 10.4 times that of PP and improvements in strengths ranged from 1.2 to 3.9 times that of PP. The tensile bars with MAGPP containing 50 and 80 wt% LCP show a positive deviation from the rule of mixtures. In fact, at 80 wt% LCP, the modulus of tensile bars containing MAGPP was higher than that of Vectra A.
2. The PP/Vectra A blends are dominated by the highly anisotropic nature of Vectra A. On addition of MAGPP, there is an increase in both the transverse moduli and strength of the blends, but the transverse strengths are still below that of PP.
3. The PP/Vectra A blends are more brittle than PP and the toughness improved on addition of MAGPP for some blend compositions and not for others.
4. Over a large temperature range, the PP/Vectra A blends in the presence of MAGPP exhibited substantially improved dynamic storage moduli and creep compliances over those of PP.
5. Addition of MAGPP resulted in a finer dispersion, a more uniform distribution of the LCP phase and improved adhesion between PP and Vectra A. Greater compatibility between the components is manifested by the morphological changes brought about by the addition of MAGPP to PP/Vectra A blends. It has been previously claimed that greater compatibility between the LCPs and the thermoplastic matrix polymers may be detrimental to fibril formation and consequently may affect the reinforcing capabilities of the LCP. However, the results presented here indicate that there is a significant increase in physical properties owing to the presence of MAGPP.
6. Significant improvements in tensile properties were observed on the addition of MAGPP for blends of PP with LC3000, with tensile bars again exhibiting higher properties than plaques.
7. The PP/LC3000 blends are more brittle than PP but addition of MAGPP enhanced the toughness of the blends at all levels of LCP.
8. The final properties of the blends in the presence of MAGPP are not only determined by the amount and properties of the reinforcing LCPs but also by the effectiveness of the MAGPP, which varies with the LCP type.

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