

Studies on Compatibilization of Blends of Polypropylene and a Thermotropic Liquid Crystalline Polymer

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SYNOPSIS

Thermotropic liquid crystalline polymers, LCPs, are frequently blended with thermoplastics to achieve an *in situ* composite structure. Significant mechanical reinforcement is obtained for the matrix polymer in the direction of the LCP fibers, but the transversal properties are often inferior because of the incompatibility of the components. Blends of LCP with polypropylene, and with three related matrix polymers, and PP/LCP blends with added potential compatibilizers were prepared and studied for their mechanical properties and morphology. A notable improvement in impact strength was achieved when a small amount of ethylene-based terpolymer was added as compatibilizer. © 1993 John Wiley & Sons, Inc.

INTRODUCTION

Thermotropic main-chain liquid crystalline polymers (LCPs) are melt blended with thermoplastics in order to achieve a so called *in situ* composite structure with thin oriented LCP fibrils in the thermoplastic matrix. In most cases the addition of LCP increases the mechanical strength and stiffness of the thermoplastic matrix polymer. Moreover, even relatively small amounts of LCP may induce a reduction in the melt viscosity, and thus improve the processability. LCP may also improve other properties of thermoplastics, such as dimensional and thermal stability, as found in our earlier studies on blends of a polyester-type aromatic LCP with PP, PET, and PPS.^{1,2} LCPs thus offer an interesting means to upgrade the properties of thermoplastics.

LCPs and thermoplastics are generally immiscible and form a two-phase structure. Often a so called skin/core morphology is found with thin fibrillar LCP phases in the skin region and spherical or ellipsoidal LCP domains in the core. The size and shape of the LCP domains depend on LCP content,

viscosity ratio of the two polymers, and processing conditions (e.g., temperature and shear forces). Orientation of the LCP phases during the processing and the formation of fine fibrils can be significantly increased by elongational drawing, which leads to improved physical properties in the fiber direction.²

The transversal properties such as impact strength are inferior, however, and the blends are stiff but brittle. This is partly caused by the anisotropic structure but also by the lack of interaction and of interfacial adhesion between the blend components. The blends are said to be incompatible. As a polymer with no functional groups, polypropylene has poor interfacial adhesion with aromatic LC copolyesters. The use of a functionalized PP grade instead of PP as the matrix polymer, or the addition of a compatibilizer, could reasonably be expected to enhance the interaction and adhesion between the blend components.

Studies on blends of LCPs and polypropylene have been reported by Ramanathan et al.³ and La Mantia et al.,⁴ and more recently by us.^{1,2}

Ramanathan et al.³ described a special mixing technique, in which the polymers were melted in two different extruders and mixed together before the die. The LCP, Vectra A 950, was preheated up to 330°C before being combined with PP. By this method the viscosity ratio (η_{LCP}/η_{PP}) could be ad-

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justed to a desired low level, a wide processing window was achieved, and PP was not overheated. Significant improvements in modulus of drawn films were reported.

La Mantia et al.⁴ used a semi-rigid LCP synthesized from sebacic acid, hydroxybenzoic acid, and 4-4'-hydroxy-biphenyl (1 : 1 : 2). This LCP had a low melting temperature similar to polypropylene. The blends were reported to be incompatible, with very poor interfacial adhesion. However, a reduction in melt viscosity and slight increase in elastic modulus was achieved.

The aim of the work reported here was to increase the toughness of PP/LCP blends. In a first set of experiments, ethylene/propylene copolymer, functionalized polypropylene, PP/EPDM compound, and polymethylpentene were used as matrix instead of PP homopolymer; in a second set, various potential compatibilizers were added to PP homopolymer/LCP blends. The mechanical properties and morphology of the resulting blends were evaluated.

The Nature of Compatibilization

Compatibilization is a process by which an incompatible blend is rendered less incompatible, so that the resulting material is useful for engineering purposes. This kind of compatibility, we would note, does not imply thermodynamical compatibility, in which the polymers exist in a single molecularly blended homogeneous phase.⁵

Compatibilization can be either physical or chemical in nature involving partial miscibility, entanglements of polymer chains, physical interactions, or chemical reactions. The usual effects are a reduction in interfacial energy, a stabilized morphology with perhaps finer dispersion, and an increase in interfacial adhesion.⁶

The compatibility of a binary blend depends upon the interaction between the two polymers. Careful selection of the components is of great importance. In particular, the ratio of the melt viscosities and the chemical nature of the components affect the blend compatibility.^{7,8} The dispersed LCP phase should have slightly lower viscosity than the matrix polymer at the processing conditions in order to obtain good dispersion and the desired fibrous morphology. Chemical similarity may lead to partial miscibility, and the presence of functional groups to a reaction, and both of these may increase compatibility. One route to a compatibilized blend is thus to functionalize either or both of the blend components.

A different type of compatibilization involves the addition of a compatibilization agent, or compatibilizer, to act as a bridge between the binary blend components at their interfaces. The action of such compatibilizers as block and graft copolymers is based on the reactivity or miscibility of their segments with at least one of the blend components. They are in fact interfacial agents, since they tend to enrich and act at the interfaces as emulsifiers enhancing the interaction between the blend components through covalent or ionic bonding. Besides block and graft copolymers, a variety of lower molecular weight reactive chemicals promote copolymer formation or cross-linking reactions and thereby improve compatibility.^{9,10}

In the case of LCP blends, it needs to be emphasized that miscible blends are not the goal but two-phase blends that combine the good properties of the LCP and thermoplastic components. The main requirements for a compatible blend of an LCP and a thermoplastic is the formation of fine oriented LCP fibers firmly attached in the thermoplastic matrix. Thus, improving the compatibility in most cases means increasing the interfacial adhesion, which may further increase the impact strength.

EXPERIMENTAL

Materials

The liquid crystalline polymer used in this work was Vectra A 950 produced by Hoechst Celanese. It is a totally aromatic polyester-type thermotropic main-chain LCP, believed to consist of *p*-hydroxybenzoic acid (HBA) and 2,6-hydroxynaphthoic acid (HNA). The following properties are given by the manufacturer: density 1.40 g/cm³, melting point 280°C, tensile strength 165 MPa, elastic modulus 9700 MPa, and elongation at break 3.0%.¹¹ The amount of LCP blended with the thermoplastic is sometimes indicated merely by number; thus PP/30 indicates 30 wt % of LCP in PP.

Polypropylene homopolymer (PP VB 19 50K from Neste) was the usual matrix polymer in the blends, and, if not otherwise stated, it is this grade that is referred to in the text.

Besides polypropylene, several polypropylene-based and polypropylene-like polymers were used as either matrix or compatibilizer, with the purpose of improving the properties of PP/LCP blends. In a first set of experiments, ethylene-propylene copol-

mer (SB 15 50K from Neste), functionalized polypropylene PP-g-MA (Novatec 196P from Mitsubishi), and polymethylpentene, PMP (TPX 18XB from Mitsui Petrochemical) were used as matrices.

In a second set of experiments, three maleic acid/anhydride grafted polypropylenes, PP-g-MA, with acid contents of 0.2 (A), 0.4 (B), and 4.0 wt % (C), and an ethylene-based reactive terpolymer (D), were tested as potential compatibilizers, to increase the interfacial adhesion and impact strength of PP homopolymer (VB 19 50K from Neste) and LCP blends.

In a third part, the influence of EPDM rubber on PP/LCP blends was studied by blending a PP/EPDM (75/25) compound (DE 2561 from Neste) with LCP, and 5 to 20 wt % of EPDM rubber (Kel-tan 778 from DSM) with PP and 30 wt % of LCP.

Blending

LCP, the matrix thermoplastic, and the potential compatibilizers in the form of dry pellets were manually mixed in a barrel. The pellets were then carefully dried before the melt blending stage in a Conair Micro-D dehumidifying dryer. The drying conditions for the blends were selected according to the matrix polymer, and were normally at least 2 h at 70°C. Unlike the pelletized materials, the compatibilizer C, which was supplied as powder, was fed separately to the extruder.

The melt blending of the materials was carried out by a Berstorff ZE 25x33D corotating twin screw extruder at temperatures ranging from 285 to 290°C, and with a screw speed of 150 rpm. The temperature of the molten polymer was typically between 290 and 295°C. The extrudate was immediately quenched in a water bath and pelletized again.

Injection Molding

The blends and the pure polymers were injection molded into the form of test specimens after thorough drying in a Conair Micro-D dehumidifying dryer. The drying conditions were the same as before blending. Injection molding was carried out with an Engel ES 200/40 injection molding machine. The pure polymers were processed at the conditions recommended by the manufacturers, polypropylenes at 210–230°C, and the blends at 280–290°C. A mold temperature of about 35°C was chosen for all materials. The processing conditions were not opti-

mized for each separate blend composition, but similar conditions were used for each blend type.

Extrusion

Some of the blends were extruded to strands of different diameter with a Brabender Plasti-Corder PLE 651 laboratory single screw extruder connected to a belt capstan. The hot extrudate was immediately quenched in a water bath, and drawn at different speeds of the take-up machine to form strands of different diameter. The speed of the belt capstan ranged from 10 to 40 m/min. The draw ratio for each strand was determined as the ratio between the die and the strand cross sections (S_o/S_s). The dimensions of the round hole capillary die were length (L) 30 mm and diameter (D) 5 mm, and thus L/D 6. The cylinder temperatures of the extruder were 280–285°C.

Characterization and Testing

Standard test bars of the injection-molded specimens were used for the tensile, bending, and impact tests.

Before testing, all samples were conditioned applying ISO 291 at test room conditions (about 23°C and 50% relative humidity) for at least 88 h.

Tensile properties were measured according to ISO 527 with an Instron 4204 testing machine equipped with a computer. The strain rate was 5 mm/min for the determination of both tensile strength and elastic modulus. The tests were performed without an extensometer with a gauge length of 115 mm. The dimensions of the test bars were length 154 mm, width of narrow part 10 mm, and thickness 4 mm.

For the extruded strands the test speed was 3 mm/min and the gauge length 100 mm. The results of the tensile tests are averages of at least five parallel samples.

Flexural properties were studied with the same equipment according to ISO 178, by the three-point-bending test. The dimensions of the test bars were length 112 mm, width 10 mm, and thickness 4 mm. The test speed was 5 mm/min.

Charpy impact strength was determined of un-notched test specimens according to ISO 179 by using a Zwick 5102 pendulum-type testing machine. The dimensions of the specimens were 50 × 6 × 4 mm. A pendulum of 40 kpcm was used.

Heat deflection temperature (HDT) was determined for some samples according to ISO 75 by using a Zwick 4204 testing machine. The measurements were performed by method A for a load of 1.82 N/mm² and with the temperature rising by 2°C/min. The dimensions of the test bars were 112 × 10 × 4 mm.

DSC measurements were carried out with Mettler DSC 30 equipment under nitrogen atmosphere. The temperature was raised by 20°C/min from 0 to 320°C, held at 320°C for 5 min, cooled to -50°C, and raised again to 320°C.

Morphology of the fractured surfaces of the blend samples was investigated with a JEOL JXA-840A scanning electron microscope (SEM). The samples were fractured after dipping in liquid nitrogen, and the fractured surfaces were coated with a layer of gold about 15 nm thick.

Melt rheology of the polymers was investigated by measuring the melt viscosities in shear flow with a Göttfert Rheograph 2002 capillary viscosimeter. The measuring temperature was 290°C. The *L/D* ratio of the die was 20/0.5 mm and the shear rate ranged from 5 to about 10 000 /s. Bagley correction was not made but the results included Rabinowitch correction.

RESULTS AND DISCUSSION

PP Homopolymer

The neat PP homopolymer used as a reference material was processed in two ways. For most samples, processing consisted of short predrying at 70°C and injection molding at 220–250°C. To a smaller num-

Table I Mechanical Properties of Injection-Molded Blends

Material	Tensile Properties		Flexural Properties		Impact Strength
	<i>E</i> (MPa)	σ (MPa)/ ϵ (%)	<i>E</i> (MPa)	σ (MPa)/ ϵ_y (mm)	Charpy (kJ/m ²)
PP	1047	24.0/> 100	954	29.8/13.2	Not broken
PP (twin-screw)	1174	26.0/> 100	1080	33.6/12.8	Not broken
PP/20	1634	29.5/6.8	1621	40.4/11.6	17.1
PP/30	2365	33.0/2.6	2467	47.6/8.2	9.2
PP/LCP/EPDM blends					
PP/30/EPDM 5	2385	30.9/3.1	2549	42.4/6.9	8.3
PP/30/EPDM 10	2151	28.2/3.2	2415	39.8/6.8	8.4
PP/30/EPDM 20	1796	22.1/6.7	1778	29.5/6.7	12.1
PP/EPDM (75/25) compound					
PP compound	627	12.4/> 110	622	17.0/11.8	Not broken
PP compound/30	2013	26.4/2.5	2157	33.4/(5.7) ^a	8.7
PP comp/PP(50/50)/30	2149	31.8/2.7	2574	41.3/(5.8) ^a	8.5
Functionalized PP					
PP-g-MA (0.2)	444	10.9/> 110	408	12.9/12.3	Not broken
PP-g-MA/10	726	13.1/> 110	753	18.0/10.5	Not broken
PP-g-MA/30	2018	25.7/6.7	1982	30.2/(5.3) ^a	9.0
PP copolymer					
PP cop./20	869	18.5/> 110	756	22.4/12.7	Not broken
PP cop./30	1699	24.1/9.7 ^b	1499	32.8/9.5	14.2
PP cop./30	2510	29.2/2.0	2162	37.8/6.6	9.8
PP cop./30/D 5	1576	24.6/3.3	1488	32.2/10.7	14.3
Without melt blending					
PP cop./10	1160	21.0/> 75	1274	29.0/10.9	(34.1) ^b
PP cop./30	2431	29.0/2.3	2390	37.4/5.4	(10.4) ^b
PMP	923	18.5/> 60	972	29.9/12.7	(17.8) ^c
PMP/30	2128	31.7/3.6	2235	46.0/8.9	6.6

E (MPa) = elastic modulus; σ (MPa) = maximal strength; ϵ (%) = strain at break; ϵ_y (mm) = displacement at yield.

Tensile test: modulus determined without extensometer with a gauge length of 115 mm.

^a Broken.

^b Not entirely broken.

^c Not all samples broken.

ber of samples the same procedure as for the blends was applied, that is, melt blending in a twin screw extruder at 290°C, pelletizing, and injection molding. The melt blending experiment confirmed that melt blending does not diminish the mechanical properties of PP (Table I). Perhaps the short residence time of the material in the extruder at temperatures higher than normal explains the lack of effect. This particular PP grade was chosen in order to get a viscosity ratio of η_{LCP}/η_{PP} slightly less than 1 over a broad range of shear rates at the processing temperature 290–295°C.

Matrices Other Than PP Homopolymer

Ethylene-propylene copolymer, functionalized polypropylene, and polymethylpentene were blended with LCP in the hope of achieving more interaction between LCP and the matrix polymer than in the PP homopolymer/LCP blend. Enhanced interaction, either chemical or physical in nature, could be expected to lead to improved interfacial adhesion

and a toughened blend. With its higher melting temperature (240°C) and greater temperature resistance, polymethylpentene was expected to be more suitable than polypropylene for blending with Vectra at high temperatures.

All materials were rather soft, so that the increases in strength and modulus due to LCP addition (Table I) were substantial. At 10 wt % LCP all blends were tough, but at 30 wt % LCP the impact properties were reduced to about the level typical of PP/LCP blends. Evidently neither the functionality (0.2%) of PP-g-MA nor the short side-chain in PMP was effective enough to improve the interaction between the blend components.

All products exhibited the characteristic skin/core morphology of LCP and thermoplastic blends, with fibril-like LCP phases in the skin layer and spherical LCP domains in the core. However, the average diameter of the LCP domains was slightly smaller (about 2–4 μm) in PP copolymer/LCP and PP-g-MA/LCP blends than in PP homopolymer/LCP blends (about 8–10 μm). Moreover, the LCP spheres or fibers seemed to be better attached in the

Table II Mechanical Properties of Injection-Molded Compatibilized Blends

Material	Tensile Strength		Flexural Properties		Impact Strength Charpy (kJ/m ²)
	<i>E</i> (MPa)	σ (MPa)/ ϵ (%)	<i>E</i> (MPa)	σ (MPa)/ ϵ_y (mm)	
PP	1047	24.0/> 100	954	29.8/13.2	Not broken
PP (twin-screw)	1174	26.0/> 100	1080	33.6/12.8	Not broken
PP/20	1634	29.5/6.8	1621	40.4/11.6	17.1
PP/30	2365	33.0/2.6	2467	47.6/8.2	9.2
PP-g-MA 0.2 (A)					
PP/30/A 2	2383	36.1/2.9	2527	51.0/8.3 ^a	9.0
PP/30/A 5	2481	36.3/2.7	2511	48.3/7.9 ^a	9.5
PP/30/A 10	2538	33.6/3.0	2331	44.4/8.0 ^a	9.8
PP-g-MA 0.4 (B)					
PP/30/B 2	2732	36.2/2.0	2595	49.5/8.0	8.9
PP/30/B 5	2898	37.9/2.0	2740	49.4/8.0	8.8
PP/30/B 10	2388	34.2/2.6	2398	48.0/9.3	8.7
PP-g-MA 4.0 (C)					
PP/20/C 3	1744	33.2/4.4	1940	48.5/10.0	9.0
PP/20/C 7	1700	33.0/3.7	2018	48.6/9.1	10.3
PP/20/D 3	1264	25.2/> 100	1302	35.4/12.6	(54.4) ^b
PP/20/D 7	962	21.0/> 100	878	27.0/13.4	Not broken
PP/30/D 2	1895	31.1/4.7	2001	46.5/11.0	11.3
PP/30/D 5	1466	26.0/8.4	1324	36.0/12.3	30.7
PP/30/D 10	1192	22.6/12.2	1044	30.5/13.1	44.6

E (MPa) = elastic modulus; σ (MPa) = maximal strength; ϵ (%) = strain at break; ϵ_y (mm) = displacement at yield.

Tensile test: modulus determined without extensometer with a gauge length of 115 mm.

^a Broken.

^b Not all samples broken.

copolymer and PP-g-MA matrices than in PP homopolymer.

Addition of compatibilizer D to a blend of PP copolymer and 30 wt % of LCP did not improve the impact strength of the blend significantly. The mechanical properties of the blend were about the same as for PP/20 wt % LCP. However, polymer D did modify the morphology just as when added to PP/LCP blends, as will be discussed below. As a result, LCP was dispersed to smaller domains of 1–2 μm in diameter.

Some of the PP copolymer/LCP blends were injection molded without prior melt blending in a twin screw extruder. The mechanical properties were virtually unchanged, which means that the melt blending stage is not necessary, at least in this case. The same was found earlier for blends of PP homopolymer and LCP.¹²

Compatibilized Blends of PP Homopolymer and LCP

Maleic anhydride grafted polypropylene, PP-g-MA at three different MA contents (A–C), and a reactive ethylene-based terpolymer (D), were studied as compatibilizers for PP/30 wt % LCP blends. Possessing chemical functionality, they were expected to have some influence on the interfacial adhesion of PP homopolymer and LCP.

PP-g-MA Compatibilizers (A–C)

The PP-g-MA compatibilizers (A–C) did not improve the impact strength of PP/LCP blends as was hoped. Nevertheless, clear enhancements in tensile strength and elastic modulus were found at all MA contents, the best combination being PP/LCP with 5 wt % of compatibilizer B (0.4 wt % MA) (Table II). The overall effects of these compatibilizers on the properties of PP/LCP blends were quite weak, however.

Polymers A–C did not substantially change the morphology of the PP/LCP blends, but allowed fiber formation, which explains the high level of strength and modulus. According to viscosity measurements made on PP/LCP/B 10 blend, the effect of polymer B on the blend viscosity was small.

Compatibilizer D

Mechanical Properties. Compatibilizer D (a reactive ethylene-based terpolymer) modified the properties of PP/LCP blends significantly more than compatibilizers A–C.

The most dramatic change was in Charpy impact strength, which increased as a function of the compatibilizer content (Table II). At the same time, however, the tensile and flexural strength and modulus of the blend drastically decreased. Nevertheless, at a proper composition an increased toughness

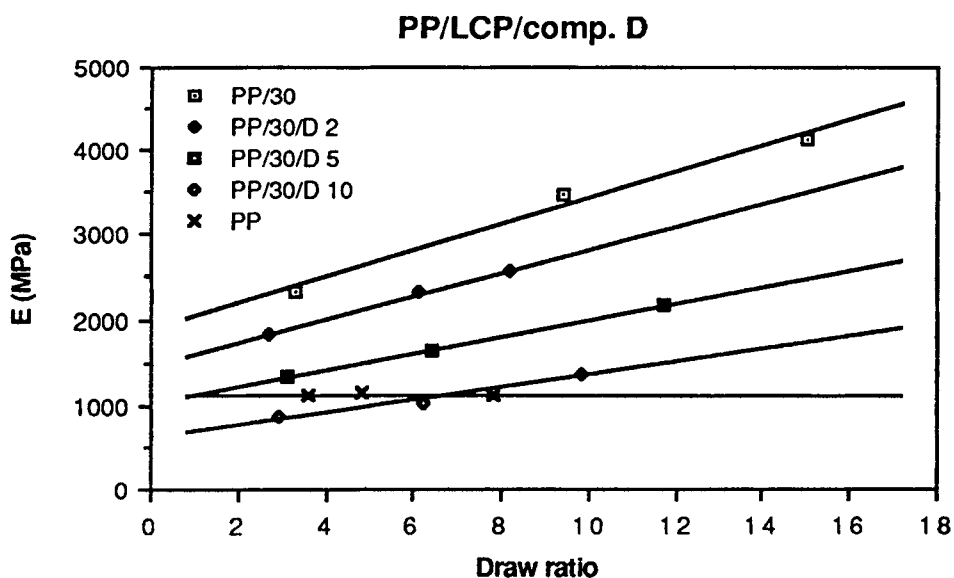


Figure 1 Elastic modulus of PP/30% LCP blends with 0, 2, 5, and 10% of compatibilizer D as a function of draw ratio.

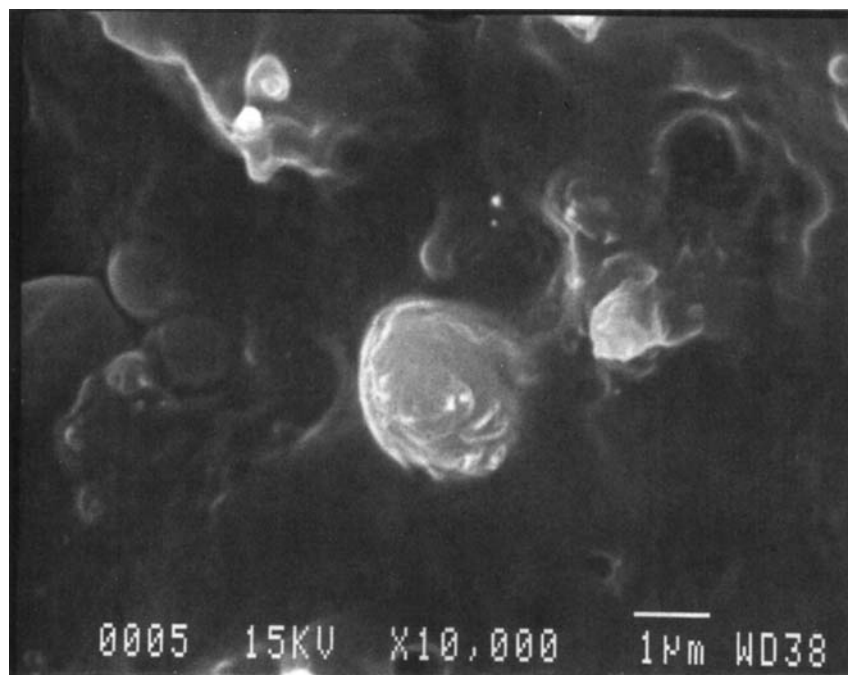


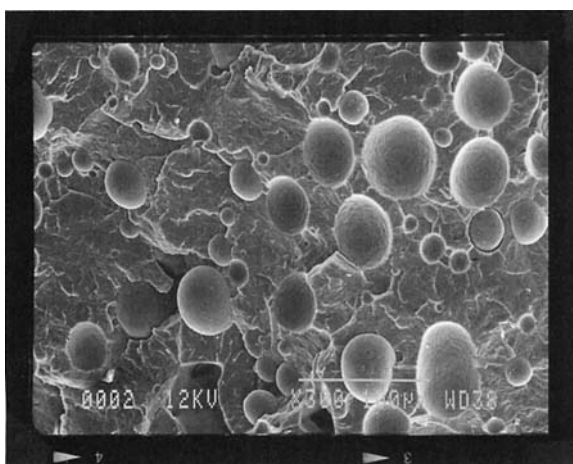
Figure 2 SEM micrograph of the fractured surface of PP/20% LCP/7% compatibilizer D blend ($\times 10,000$).

could be achieved without total loss of the improvements in tensile and flexural strength and modulus achieved through LCP addition.

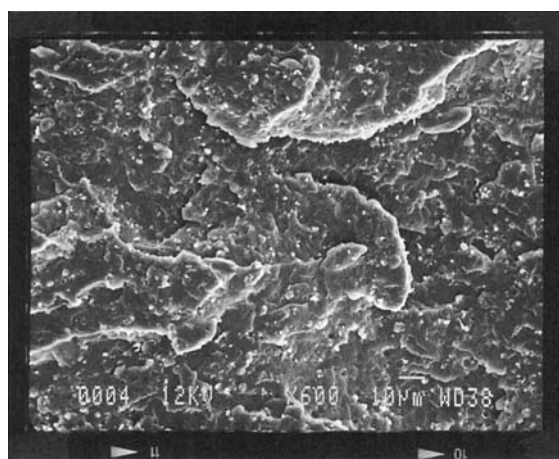
Extrusion experiments showed that higher levels of strength and modulus could be obtained by slight

drawing of the PP/LCP/D strand, as seen in Figure 1.

Morphology. The increased toughness induced by compatibilizer D can be understood from Figure 2,

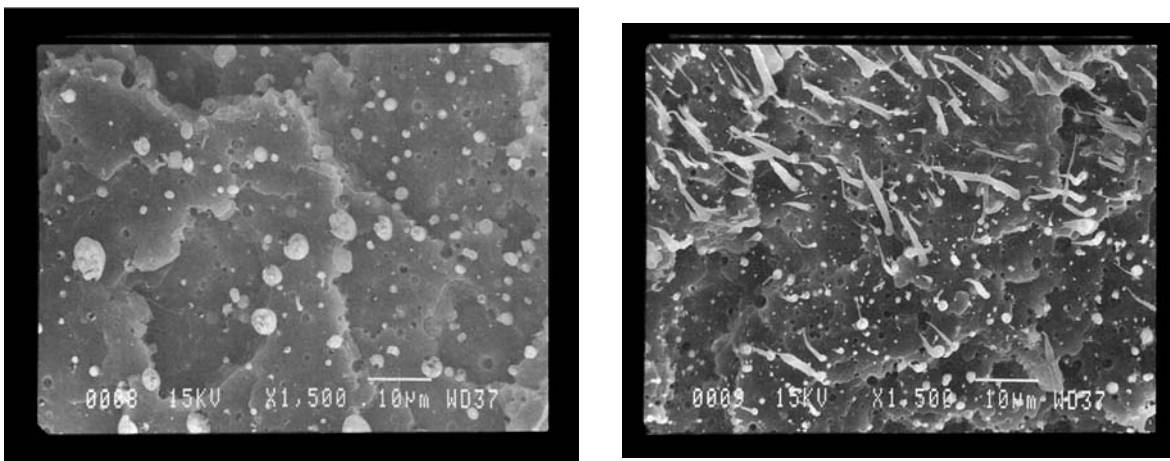


a)



b)

Figure 3 SEM micrograph of the fractured surface of a PP/30% LCP blend mixed in a Brabender Roller Mixer: (a) without compatibilizer ($\times 300$) and (b) with 7% of compatibilizer D ($\times 600$).



a)

b)

Figure 4 SEM micrographs of the fractured surfaces of blends containing PP/30% LCP/5% compatibilizer D: (a) core and (b) skin layer ($\times 1500$).

which shows the good attachment of LCP domains in the PP matrix. Polymer D also had a strong dispersive effect on LCP phases, as revealed in SEM micrographs. Figure 3 shows the huge difference in

the LCP domain size after mixing for 10 min at 290°C in a Brabender Roller Mixer W 50 EH. The differences were smaller but still noticeable after twin screw extrusion. The average diameter of the

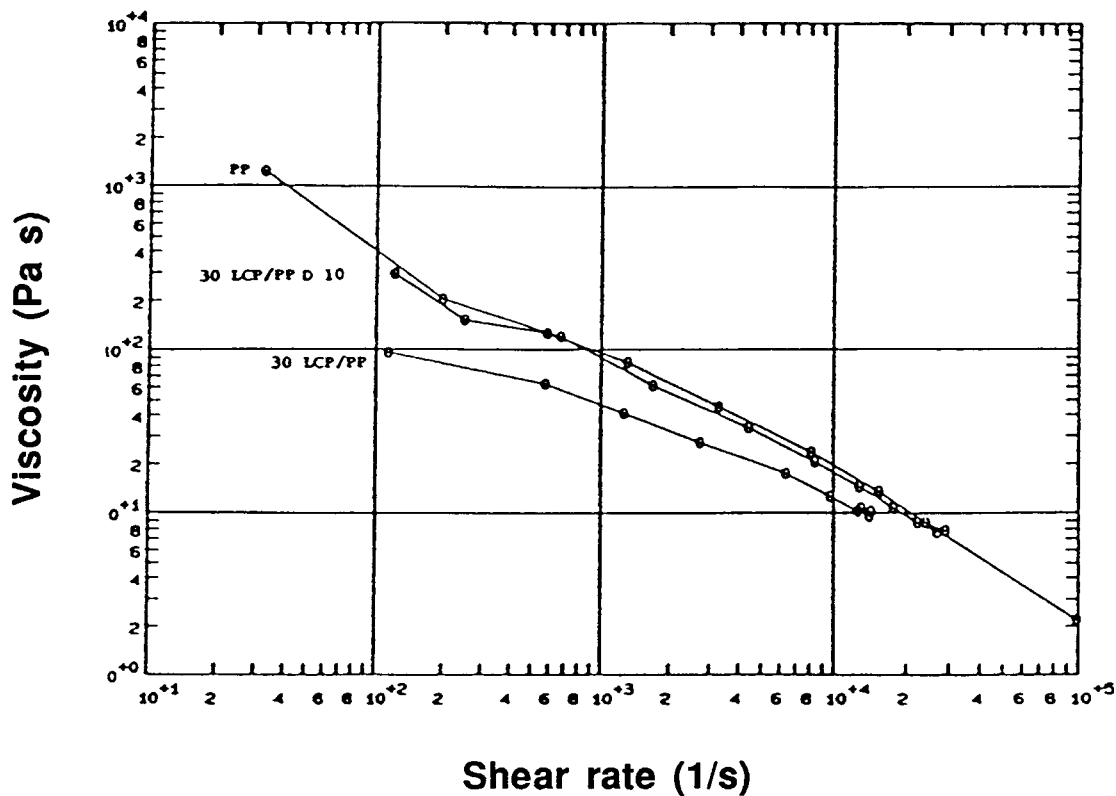


Figure 5 Melt viscosity vs. shear rate for PP, PP/30% LCP and PP/30% LCP/10% compatibilizer D measured at 290°C.

LCP domains in PP/20 wt % LCP blends was about 6–10 μm . After a small addition (3 wt %) of polymer D the thickness was reduced to 1–2 μm . The dispersion of LCP to smaller domains may have made the fiber formation more difficult, and so have contributed to the decrease in strength and modulus found for the injection-molded specimens. Extrusion experiments with PP/LCP/D blends showed, however, that fiber formation and better mechanical properties are obtained if elongational forces are imposing on the polymer melt (Fig. 4).

Rheology. Melt viscosities of PP homopolymer and of PP/30 and PP/30/D 10 blends as a function of shear rate are shown in Figure 5. The measurements were made at 290°C, which was the processing temperature for the blends. LCP decreased the melt viscosity of PP, as found earlier. Polymer D, however, increased the viscosity of the blend to about the level of neat polypropylene, although alone it was less viscous than any one of the blend components. The increase in the blend viscosity is thus evidence of enhanced interaction between the blend components, with probably a chemical reaction.

Thermal Properties. DSC measurements were made to study the possible changes in the miscibility of the blend components and crystallinity of polypropylene upon addition of polymer D (Table III). No change in the glass-transition temperature of polypropylene was found, and the decrease in the crystallinity of polypropylene from 39% for PP/30 to 33% for PP/30/D 10 was minimal. The heat deflection temperature (HDT A) of PP was increased after 30 wt % LCP addition from 51°C to 81°C. The addition of compatibilizer D, however, reduced the HDT value toward that of neat polypropylene.

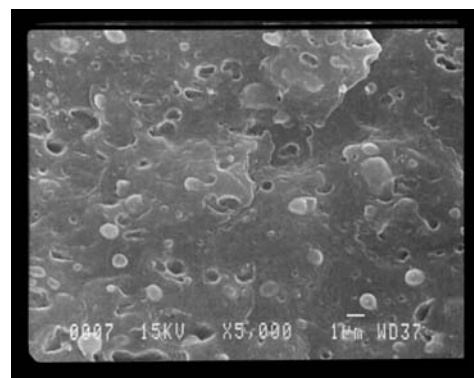
Blends Containing EPDM Rubber

A further attempt to improve the impact behavior of PP/LCP blends was made by adding 5–20 wt %

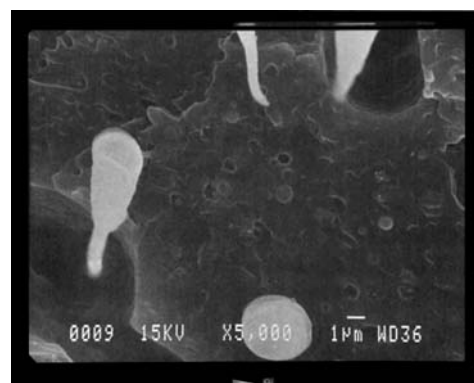
Table III Glass-Transition Temperature (T_g) and Crystallinity of PP and HDT/A for the Blends

Material	T_g (°C)	Crystallinity (%)	HDT/A (°C)
PP	-6	47	51
PP/30	-10	39	81
PP/30/D 2	-5	35	69
PP/30/D 5	-6	31	58
PP/30/D 10	-7	33	52

a)



b)



c)

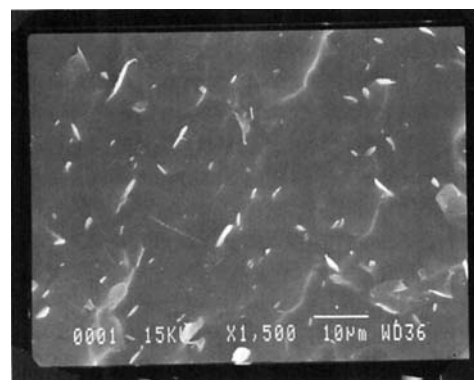


Figure 6 SEM micrographs of the cross section of (a) PP/25% EPDM compound ($\times 5000$), (b) PP/18% EPDM/30% LCP blend ($\times 5000$), and (c) EPDM/30% LCP blend ($\times 1500$).

of EPDM rubber to the blends and by using a PP/EPDM (75/25) compound as matrix. In these blends, LCP formed fibers near the skin layer, which is the basis of mechanical reinforcement. Increased addition of rubber clearly softened the material, but the hoped for improvement in impact strength was minimal. Thus the addition of EPDM rubber to the blend system did not have a similar influence on the

impact properties as the ethylene-based rubber-like functional polymer. This gives still more reason to believe that the changes induced by polymer D were based on a chemical reaction.

SEM micrographs revealed a more or less even distribution of spherical rubber particles about 0.5–1 μm in diameter over the cross section of the blend sample. Nevertheless, the rubber particles did not locate heavily at the interfaces, where they might have formed bridges between the components or acted as crack stoppers, and thus no toughening effect was found. Clearly the EPDM was well dispersed in the polypropylene matrix, but it did not interact with the LCP. The SEM micrographs in Figure 6 show the morphology of PP/LCP/EPDM blends at different compositions.

The toughening effect of rubber particles on thermoplastics strongly depends on the particle size and size distribution, which in turn depend on the viscosity ratio and molecular features of the modifier. In addition, the deformation of rubber particles is a function of the elasticity difference.¹³ The addition of a rubbery component to an incompatible blend of polypropylene and LCP makes the blend system even more complex. Further investigations are needed in order to better understand and control the morphology and properties of these ternary blends.

CONCLUSIONS

In earlier studies^{1,2} we found that an aromatic polyester-type main-chain LCP can significantly modify the properties of polypropylene. Increasing the LCP content led to considerable improvements in modulus and tensile strength, but at the same time the impact strength decreased. The blends were generally stiff but brittle.

In this work PP homopolymer, ethylene-propylene copolymer, maleic anhydride grafted polypropylene, and polymethylpentene were used as matrix polymers for the same LCP. The results were qualitatively similar and no improvement in impact strength was found.

The blends of LCP and polypropylene consisted of two discrete phases and exhibited a skin/core morphology with thin oriented LCP fibers in the skin layer and LCP spheres or ellipsoids in the core. The mechanical reinforcement and dimensional and thermal stability of the blends were based on this composite-like morphology. The anisotropic structure had an influence on the poor impact strength

of the blends, but mainly this was caused by the lack of interfacial adhesion between the components. The blends were incompatible, and compatibilization is needed to overcome the brittleness.

Compatibilizers were subsequently introduced to blends of PP and LCP with the aim of stabilizing the fibrous blend morphology and increasing the interaction and interfacial adhesion between the blend components. Maleic anhydride grafted polypropylenes A–C did not improve the impact strength of the blends, but they had a positive influence on tensile strength and stiffness. The reactive ethylene-based terpolymer, on the other hand, strongly modified the properties of the blends. At relatively small concentrations it considerably improved the impact strength of the PP/LCP blends. However, at higher concentrations it tended to soften the material, reducing the strength and stiffness toward the values typical for neat polypropylene. It also dispersed the LCP phases to smaller domains, which were able to deform to fine fibers at least in extruded blends. The action of compatibilizer D is proposed to be based on a chemical reaction, an interpretation supported by the slight increase in the melt viscosity.

Addition of EPDM rubber did not toughen the PP/LCP blend, even though the small rubber particles were evenly distributed in the matrix. The EPDM particles did not locate at the interfaces, where they might have formed bridges between the blend components.

Successful compatibilization of incompatible blends depends on the chemical suitability of the compatibilizer for the system (chemical or physical action), the amount and distribution of the modifier, and the viscosity ratios of the blend components. An effective blending, preferably in a twin screw extruder, is usually required. The morphology of ternary systems is more complex and perhaps more difficult to control. The accommodation of small amounts of compatibilizer just at the interfaces may be especially difficult to achieve.

NOMENCLATURE

DSC	differential scanning calorimetry
EPDM	ethylene propylene diene terpolymer
HBA	para-hydroxybenzoic acid
HNA	2,6-hydroxynaphthoic acid
LCP	liquid crystalline polymer
MA	maleic anhydride
PET	poly(ethylene terephthalate)
PMP	poly(4-methylpent-1-ene), polymethylpentene

PP	polypropylene
PP-g-MA	maleic anhydride grafted polypropylene
PPS	poly(phenylene sulfide)
SEM	scanning electron microscopy

SYMBOLS

D	diameter of the capillary die (mm)
E	elastic modulus (MPa)
HDT/A	heat deflection temperature (°C) measured by method A (ISO 75)
L	length of the capillary die (mm)
S_o	cross-sectional area of the capillary die (mm ²)
S_s	cross-sectional area of the extruded strand (mm ²)
T_g	glass transition temperature (°C)
T_m	melting temperature (°C)
η	melt viscosity (Pas)
σ	maximal strength (MPa)
ϵ	strain at break (%)
ϵ_y	displacement at yield (%) in 3-point-bending test

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