

In Situ Reactive Compatibilized Noryl/LCP Blends

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SYNOPSIS

This article reveals that the already known improved properties of the thermoplastic-liquid crystalline polymer (LCP) blends can be further improved substantially over the corresponding noncompatibilized counterparts by using a reactive *in situ* type compatibilizer, the styrene-glycidyl methacrylate (SG) copolymer. This SG copolymer has been demonstrated in this article to be an effective reactive compatibilizer to improve the processability, heat deflection temperature, and mechanical properties of Noryl/LCP blends. The epoxy functional groups of the SG copolymer can react with the end groups of PPO (in Noryl) and LCP. The *in situ*-formed SG-g-LCP copolymer tends to reside along the interface of Noryl-LCP and reduces the interfacial tension during melt processing. The resultant LCP fibers in the Noryl matrix of the compatibilized blends have a higher aspect ratio because the fibers become finer, longer, and tend to form lamellate domains with a greater interphase contact area than those from the noncompatibilized blends. The compatibilized blends also improve the interphase adhesion between Noryl and LCP. The presence of ethyl triphenylphosphonium bromide catalyst promotes the grafting reaction to improve blend compatibilization. © 1995 John Wiley & Sons, Inc.

INTRODUCTION

Polymer blends between thermotropic liquid crystalline polymers with other thermoplastics have been the subject of great interest recently from both academia and industries.¹⁻³⁰ The LCP phase, due to its inherent molecular structure and lower viscosity in general, with respect to the thermoplastic matrix, tends to form fibrils in the matrix under normal melt processing conditions. Among the numerous published literature, two major achievements by the addition of a small amount of LCP in thermoplastic are processability improvement and mechanical properties enhancement, especially for modulus and tensile strength. Other than certain blending pairs with structural similarity, most LCP/thermoplastic blends are considered to be immiscible, incompatible, and with poor interfacial adhesion. However, reducing viscosity alone does not solve all the processing problems such as die swelling and melt fracture during extruder blending usually encountered

in most incompatible blends. Most literature tends to emphasize the processability improvement due to viscosity reduction in essentially all the LCP/thermoplastic blends but fails to address these processing difficulties due to the incompatibility of the blends.

The morphology and properties of the *in situ* blends should be affected by the interaction between LCP and the matrix. A prerequisite for good interfacial adhesion and mechanical strength in the solid state is a reduced interfacial tension in the melt state.³¹ Interfacial tension determines the wettability and compatibility of the blend components. Factors such as miscibility or compatibility could contribute significantly to the interaction between the matrix and the LCP and, hence, the resultant physical properties of the blends. Various types of compatibilizers have been extensively used to reduce the interfacial tension in many immiscible and incompatible thermoplastic blends. However, this approach has rarely been extended to the thermoplastic/LCP blends. Transesterification reactions can increase the miscibility between liquid crystalline copolyesters and isotropic polyesters.⁵⁻⁷ Amendola et al. studied the morphologies of the compatibilized PC/LCP blends using the ester interchanged products of the blend constituents but did

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Table I Extrusion Processing Conditions

Twin Screw Extruder: Temperature °C											
Stage:	1.	2.	3.	4.	5.	6.	7.	8.	9.	Die	
Temp.:	220	260	270	280	285	295	290	295	290	290	
Motor Rate:	250 rpm										
Feeder Rate:	165 g/min										
Composition										Extruder Current (Amps)	
NORYL (N)										23 ~ 25	
N/LCP				95/5							22 ~ 23
N/LCP/SG5				95/5							23 ~ 24
N/LCP/SG5/CAT				95/5/0.02							23 ~ 24
N/LCP				90/10							21 ~ 22
N/LCP/SG2				90/10/5							22 ~ 23
N/LCP/SG2/CAT				90/10/5/0.02							22 ~ 23
N/LCP/SG5				90/10/5							22 ~ 24
N/LCP/SG5/CAT				90/10/5/0.02							23 ~ 24
N/LCP/SG10				90/10/5							23 ~ 24
N/LCP/SG10/CAT				90/10/5/0.02							23 ~ 24
N/LCP				85/15							20 ~ 22
N/LCP/SG5				85/15/5							22 ~ 24
N/LCP/SG5/CAT				85/15/5/0.02							22 ~ 24
N/LCP				80/20							20 ~ 22
N/LCP/SG5				80/20/5							21 ~ 23
N/LCP/SG5/CAT				80/20/5/0.02							22 ~ 24

not give the resultant mechanical properties.³⁰ The conventional compatibilizer, a block or graft nonreactive copolymer with segments identical to or miscible with the blend components, is the common approach to compatibilize an incompatible blend. A reactive compatibilizer is a copolymer C-X, which is able to react with component A to form a C-X-A block or graft copolymer in an A/B blend, provided that C is either identical or miscible (or at least partial miscible) with component B.³¹⁻³⁶ This same reactive copolymer is now used to compatibilize the LCP/thermoplastic blend in this article.

MATERIALS AND EXPERIMENTAL

Unmodified poly(2,6-dimethyl-1,4-phenylene oxide) (PPO), UPO2, is the pilot plant product from CTCI of Taiwan. Polystyrene (PS), was obtained from Taita Chemical Co., Ltd. of Taiwan. Noryl, a miscible PPO/PS = 50/50 blend, was prepared in our lab with a twin screw extruder. We will treat this Noryl mixture as a single matrix in this article because full miscibility between PS and PPO has been well recognized.³⁹⁻⁴¹ The liquid crystalline polymer, Vectra A950, which was kindly donated by Hoechst Celanese

Corp., is a wholly aromatic LCP composed of 70 mol % 6-hydroxy-2-naphthonic acid and 30 mol % *p*-hydroxybenzoic acid. Reactive compatibilizers, styrene-glycidyl methacrylate copolymers (SG) with various monomer ratios, were synthesized by suspension polymerization.³⁴ Compatibilizers SG2, SG5, and SG10 represent 2, 5, and 10 wt % of glycidyl methacrylate monomer in the SG copolymers. The catalyst employed in this study, ethyl triphenylphosphonium bromide, was purchased from Merck.

Fourier transform infrared spectroscopic (FTIR) analysis, to detect epoxy groups, was carried out using a Nicolet 500 Infrared Spectrophotometer.

Melt blending was carried out using a 30 mm co-rotating twin screw extruder, and the detailed processing conditions with corresponding extruder current input are given in Table I. The extruded pellets were dried and injection molded into standard $\frac{1}{8}$ inch ASTM testing specimens using an Arburg 3 oz injection-molding machine.

To verify the reaction between SG with LCP and PPO based on viscosity increase, 40 g of sample were tested at 290°C and 30 rpm in a Brabender Plastic-Corder.

Capillary rheological measurements of the blends and matrices were carried out using a capillary

rheometer ($L/D = 40$, orifice radius = 0.02 inch, and orifice length = 0.8 inch) from Kayeness Co. Model Galaxy X at 290°C.

Heat deflection temperature measurements (HDT) were carried out according to ASTM-D648, standard using 66 psi loading at a heating rate of 2.0°C/min.

Morphologies of the cryogenically fractured surfaces (in liquid nitrogen) were examined in a scanning electron microscope (Model S-570, Hitachi Co. of Japan). Unless specially mentioned, all the SEM morphologies were taken at a region midpoint between the central line and skin of the injection-molded specimens.

Notched and unnotched (10 mil) Izod impact tests were carried out at ambient conditions according to ASTM-D256 standard. Standard tensile tests (ASTM-D638), using an extensometer with a cross-head speed of 50 mm/min, were carried out also at ambient conditions.

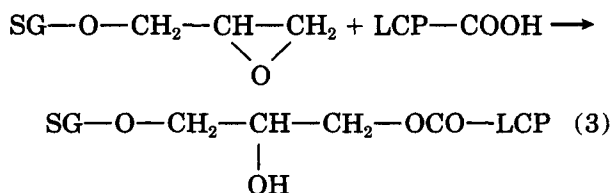
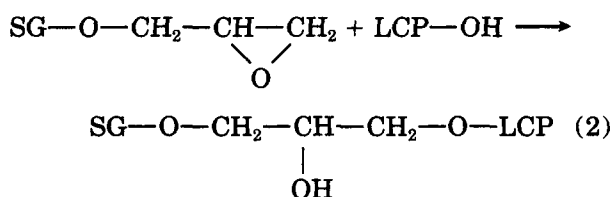
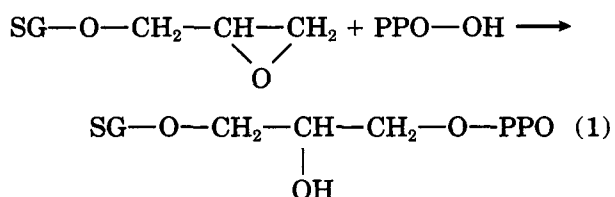
RESULTS AND DISCUSSION

Chemistry and Fundamental of *In Situ* Compatibilization

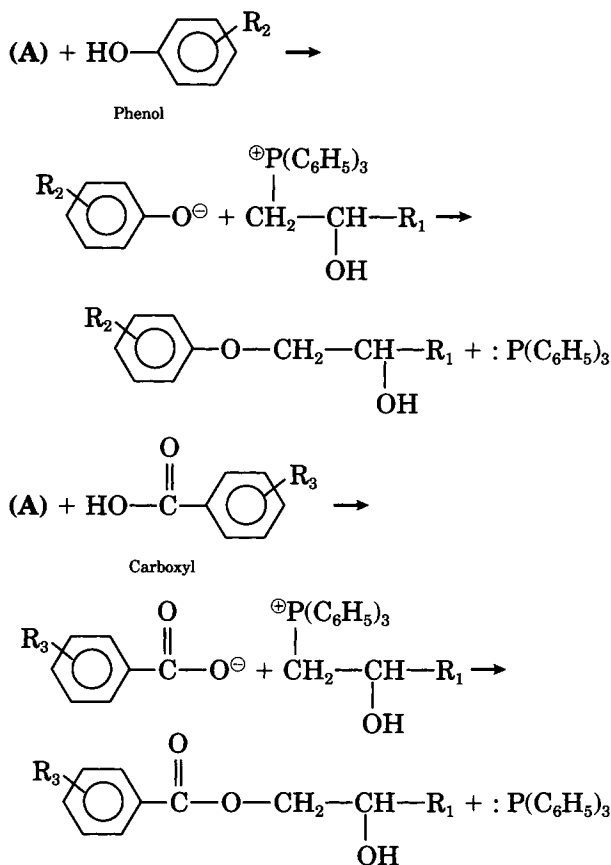
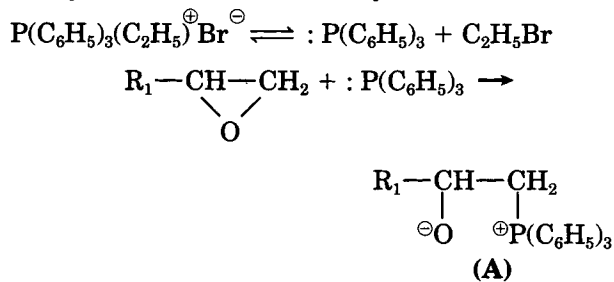
A nonreactive block or graft copolymer C-D may compatibilize the incompatible blend pair A + B, provided that C and D are either structurally identical or miscible with blend components A and B, respectively. This nonreactive compatibilizer is considered as a specific type compatibilizer because its chemical structure and quantity remain unchanged and are independent of blending conditions. Contrarily, a reactive *in situ*-formed compatibilizer is considered as a nonspecific type compatibilizer because the chemical structure and quantity of the eventually formed copolymer will vary with the content of the reactive group in the compatibilizer, temperature, time, catalyst, mixing efficiency, and blending sequence. A reactive compatibilizer is a selected reactive copolymer C-X which has C segments identical or miscible with component A, and X is able to react with component B in the melt to form nonspecific C-X-B copolymers in a binary A + B blend. This reactive C-X copolymer itself is not considered as a compatibilizer for the A + B blend, while those *in situ*-formed C-X-B copolymers tend to anchor along the interface and reduce the interfacial tension. Such a reactive compatibilization approach is applicable only to polymers containing certain functional groups (as chain ends or within main chain) that can be reacted with the reactive compatibilizer. Usually a blend component pos-

sessing chain-end functional groups is particularly suitable for such *in situ* compatibilization. Typical examples are the $-NH_2$ groups of polyamides and $-COOH$ (and/or $-OH$) of polyesters. The reactive compatibilizer (usually a copolymer) normally contains functional monomer units and the *in situ*-formed copolymer is the nonspecific graft-type compatibilizer. Excessive grafting will result in highly branched comb-like graft copolymer or even a crosslinked network, which is considered less effective as a compatibilizer. A lightly grafted copolymer, with one or a few grafts per chain, is believed to be a more efficient compatibilizer than the excessively grafted one.³⁴ Therefore, proper control of the graft reaction by using optimized content of the reactive groups, selection of a suitable catalyst, correct blending sequence, and processing conditions is essential to achieve the optimum performance of the resulted blend.

In this study, the reactions between the reactive compatibilizer SG and the blend constituents (LCP and PPO) are very complex. It involves at least three known competitive reactions, epoxy with phenolic-OH, epoxy with $-OH$ of LCP, and epoxy with $-COOH$ of LCP. The grafted copolymers resulting from the reaction between SG and PPO cannot function as a phase compatibilizers, and, certainly, they are undesirable but unavoidable. The copolymers from the reaction between SG and LCP have chemical structures suitable to act as compatibilizers. However, a SG chain containing both PPO and LCP graft chains can still be considered as a phase compatibilizer. The reactions of SG copolymer with end groups of PPO and LCP (phenolic-OH and carboxylic acid) can be expressed by the following equations:



Ethyltriphenyl phosphonium salts have been used commercially as catalysts to manufacture solid epoxy resins^{42,43} and epoxy ester resins,⁴⁴ which involve the reactions of epoxy with phenol and carboxyl. The possible reaction mechanisms of these catalyzed reactions in the study are as follows:



Nucleophilic attack by triphenylphosphine opens the epoxy ring and abstracts a reactive hydrogen from phenol or carboxyl to yield the phenolate or the carboxylate anion.^{45,46} The phenolate or the carboxylate reacts with the electrophilic carbon attached to the positive phosphorus to regenerate the catalyst.⁴⁵

Fourier Transform Infrared Spectroscopy (FTIR)

The IR peak at 913.2 cm⁻¹, a characteristic response of the epoxide group, has been used to monitor qual-

itatively the reactions between SG copolymer and end groups of PPO, LCP, or others. Figure 1 gives the IR spectra of pure PPO, pure SG5, melt-blended, and powder-blended PPO/SG5 = 90/10 mixtures. The area of this epoxide peak of the melt-blended mixture is significantly smaller than the powder-blended one. Figure 2 shows the IR spectra of pure LCP, SG5, melt-blended, and powder-blended LCP/SG5 = 90/10 mixtures. Again, the epoxide characteristic peak of the melt-blended mixture is significantly smaller than the powder-blended counterpart. The reduction of the epoxide peak observed does not account for all the epoxide consumed in the reactions between SG5 and PPO, and LCP, epoxide hydrolysis, or other unknown reactions may also be involved.³⁴

Processability

The addition of LCP reduces the viscosity of Noryl, as indicated by the recorded extruder input current (Table I). However, the melt extruder blending of the noncompatibilized blends of Noryl and LCP did experience various processing difficulties such as die swelling and melt fracture. The presence of 5 phr SG2 copolymer in the blend significantly reduced these processing problems. The presence of 5 phr of SG5 or SG10 (with or without catalyst) essentially solved all these processing problems and resulted in a smooth extrusion blend. Figure 3 compares the die swelling ratio between the straight Noryl/LCP blends and the corresponding compatibilized Noryl/LCP/SG5 blends. The compatibilized blends slightly increase the resultant viscosity over the corresponding noncompatibilized blends (Table I), but they did not encounter any noticeable viscosity-induced processing problem. Therefore, this *in situ* compatibilizer (SG, with or without catalyst) employed in this study is able to convert these incompatible Noryl/LCP blends into compatible blends.

Torque vs. Time

Figure 4 illustrates the comparative torque vs. time curves for Noryl, SG5, Noryl/SG5 = 50/50, and Noryl/SG5/Cat = 50/50/0.02 mixtures. The torque of the pure Noryl shows a continuous decrease with time, an indication of thermal degradation. The SG5 curve also decreases continuously with time, again an indication of thermal degradation rather than self-curing. This batch of SG5 appears less stable thermally than the one we used previously.³⁴ The mixture of Noryl and SG5, with or without catalyst, shows a gradual torque increase with time that pro-

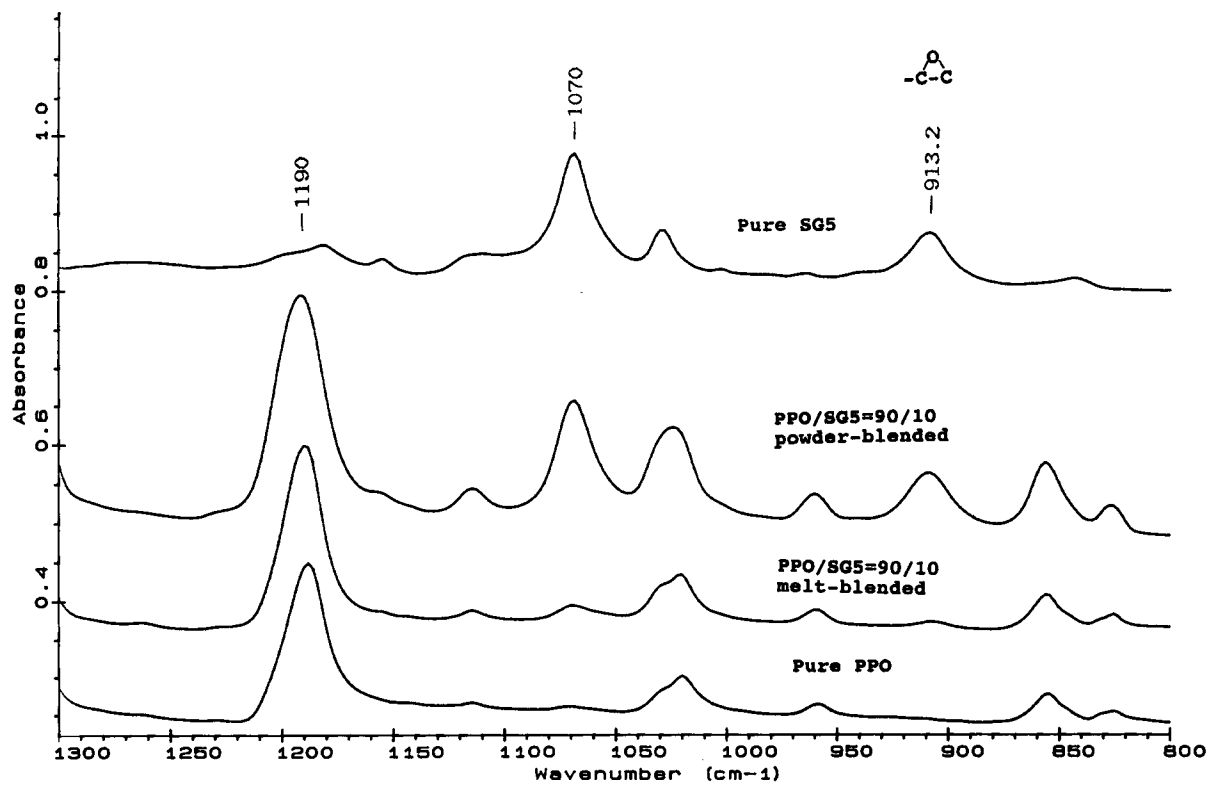


Figure 1 IR spectra of SG5, PPO, melt-blended, and powder-blended PPO/SG5 blends.

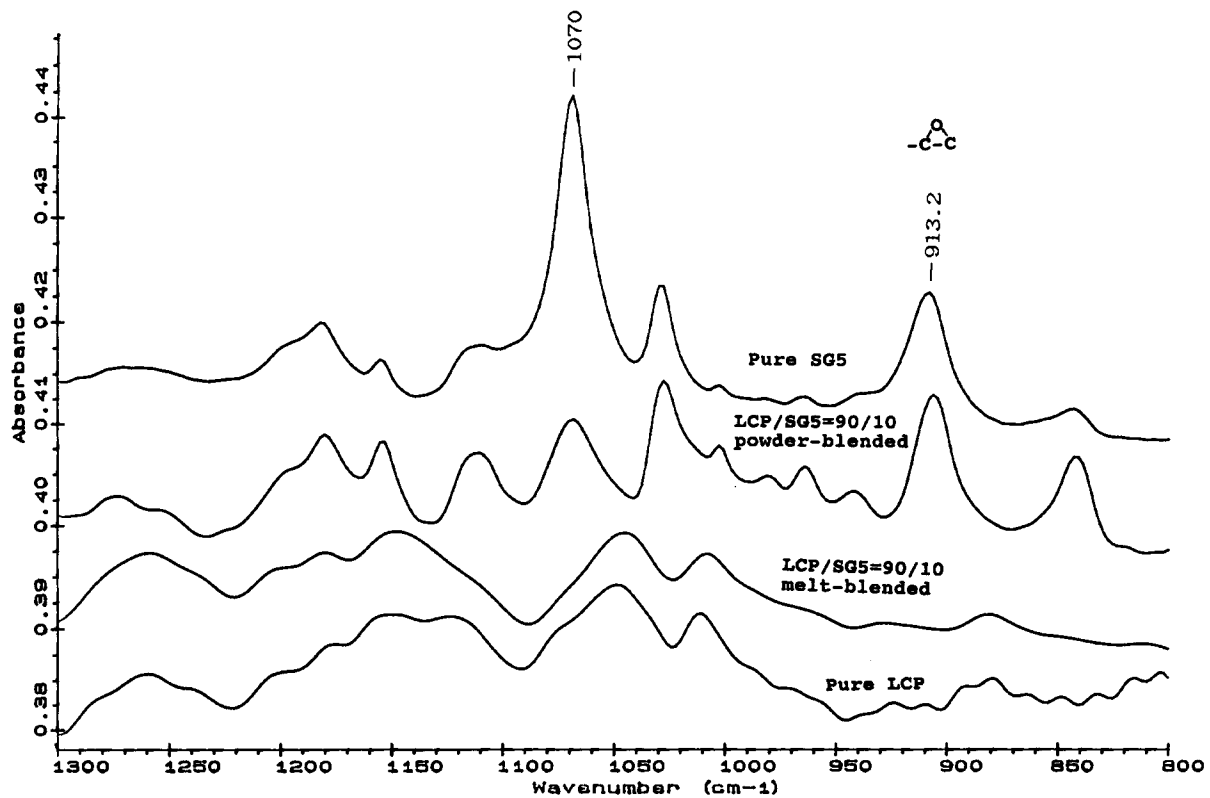


Figure 2 IR spectra of SG5, LCP, melt-blended, and powder-blended LCP/SG5 blends.

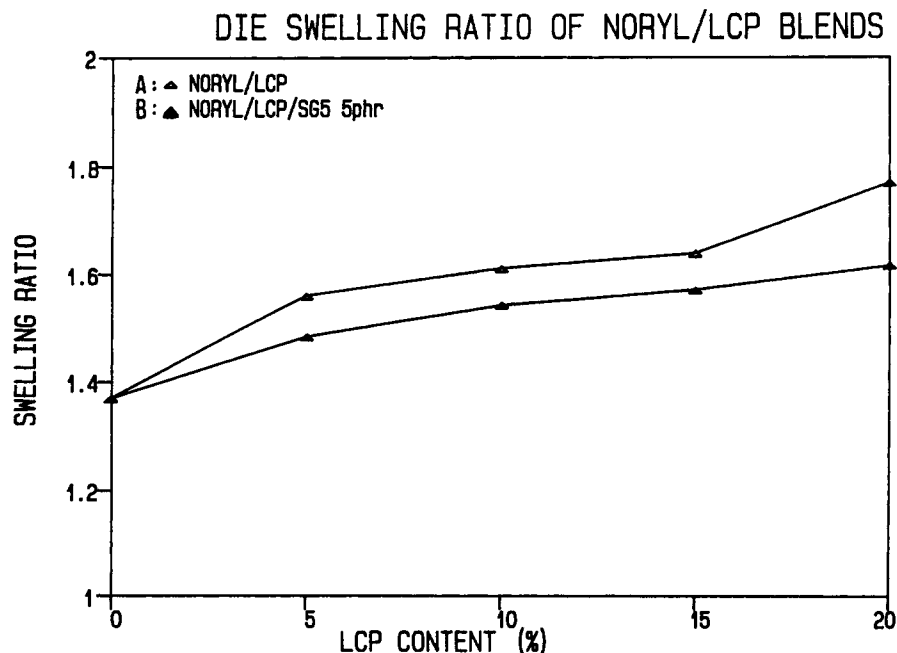


Figure 3 Effect of SG5 compatibilizer on die swelling ratio of the extrudate of the Noryl/LCP blends.

vides indirect evidence of molecular weight increase due to the anticipated graft reaction between SG5 and PPO in Noryl. Figure 5 illustrates similar plots based on LCP where the LCP also shows signs of a continuous torque decrease with time. The mixtures, LCP/SG5 and LCP/SG5/Cat, all show gradual in-

creases of the resulting torques after about 140 s. The presence of 200 ppm catalyst in the mixture increases the rate of torque increase. Again, this viscosity increase also can be attributed to the molecular weight increase from the graft reaction between LCP and SG5. The observed slopes of the Noryl/

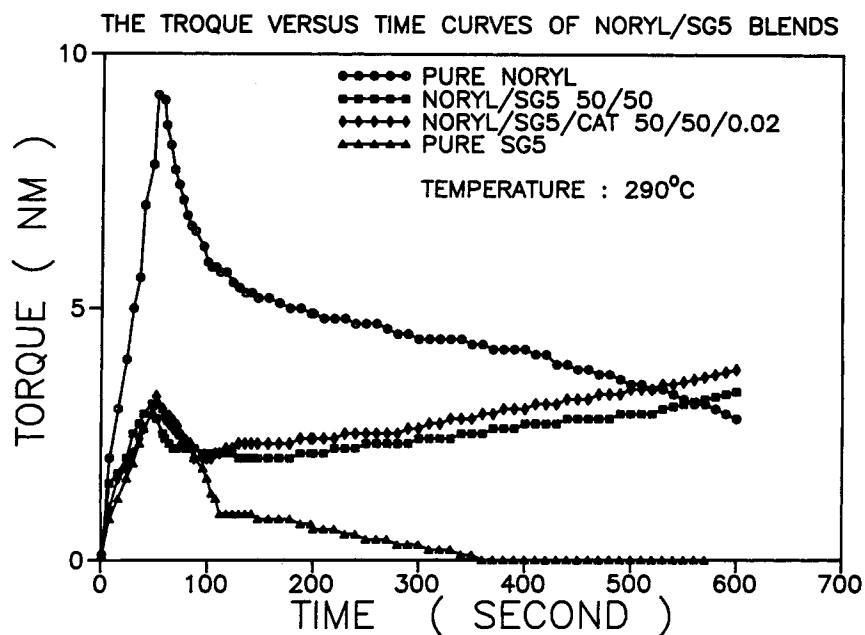


Figure 4 Plots of torque vs. time for Noryl, SG5, Noryl/SG5, and Noryl/SG5/catalyst blends.

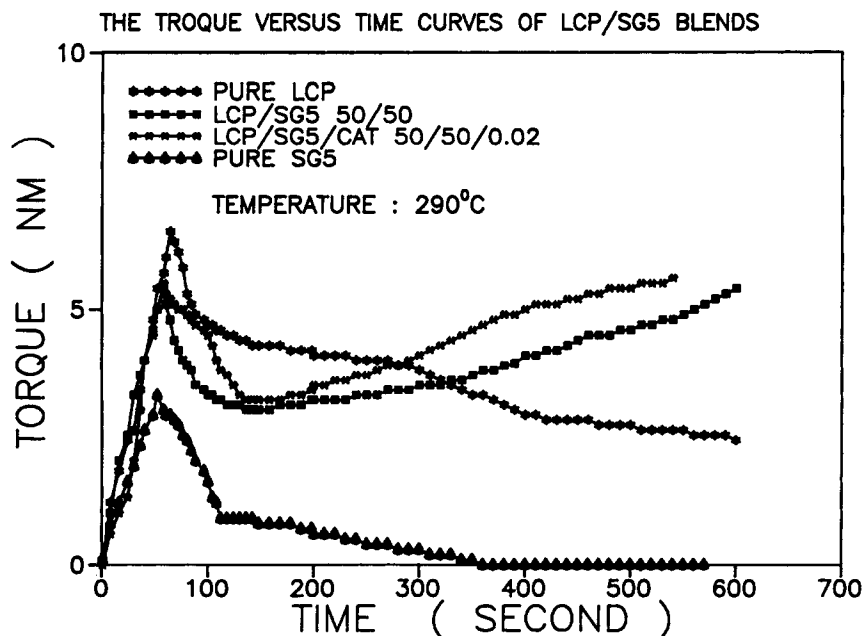


Figure 5 Plots of torque vs. time for LCP, SG5, LCP/SG5, and LCP/SG5/catalyst blends.

SG5 and the LCP/SG5 curves are comparable. This means that a significant portion of the reactive compatibilizer SG may be consumed in the reaction with PPO to form the SG-PPO graft copolymer, which does not function as a phase compatibilizer. However, the presence of an SG reactive copolymer may provide the advantage of making up the PPO molecular weight loss due to thermal degradation and maintains the intrinsic mechanical properties of the high molecular weight PPO.

Capillary Rheometry

Figure 6 gives the viscosity vs. shear rate plots at 290°C for LCP, Noryl/LCP blends, and Noryl. All the polymers in the molten phase in the studied range of shear rates exhibit a non-Newtonian flow behavior. The viscosities shear thin continuously at about the same rate over the shear rates investigated. Limtasiri and Isayev²³ reported that the dependence of viscosity on shear rate is much steeper for LCP (same type LCP as this article) and PPO/LCP blends containing a larger amount of LCP, than for PPO and lower amounts of LCP in PPO blends. At 290°C, the viscosity ratio of these two pure components, Noryl/LCP, stays nearly constant at approximately 2.5, throughout the whole range of shear rates investigated. This component viscosity ratio is relatively lower than other previously reported LCP/thermoplastic blend systems.^{15,20,21,23,26,28} The measured viscosities of the LCP/Noryl blends in this

study are about the average of the pure components. Valenza and La Mantia²⁸ reported that the viscosity of the blend decreases between those of the two parent components when the viscosity ratio, η_{TP}/η_{LCP} , is much larger than 1. There is no intersection between the LCP and Noryl flow curves under the shear rate range and temperature investigated. Tsebrenko³⁷ suggested that maximum fibrillation occurs during flow at a shear rate corresponding to the intersection point, where the viscosity ratio of the original components is approximately equal to unity.

Figure 7 shows the viscosity vs. shear rate plots of the noncompatibilized and compatibilized Noryl/LCP = 90/10 blends. All molten polymers again show non-Newtonian flow behavior. Like previous torque vs. time results, the compatibilized blends have higher viscosity than the corresponding noncompatibilized counterparts.

Heat Deflection Temperatures (HDT)

The HDTs of Noryl (PS/PPO = 50/50) and Noryl/LCP blends, with and without the presence of 5 phr SG5, are shown in Figure 8. The HDT of Noryl increases with the increase of LCP quantity. The presence of 5 phr SG5 reactive compatibilizer gains an additional HDT increase of approximately 6–10°C from all the blends. The HDT increases progressively with the increase of GMA content in the SG copolymer (data not shown here). The oriented

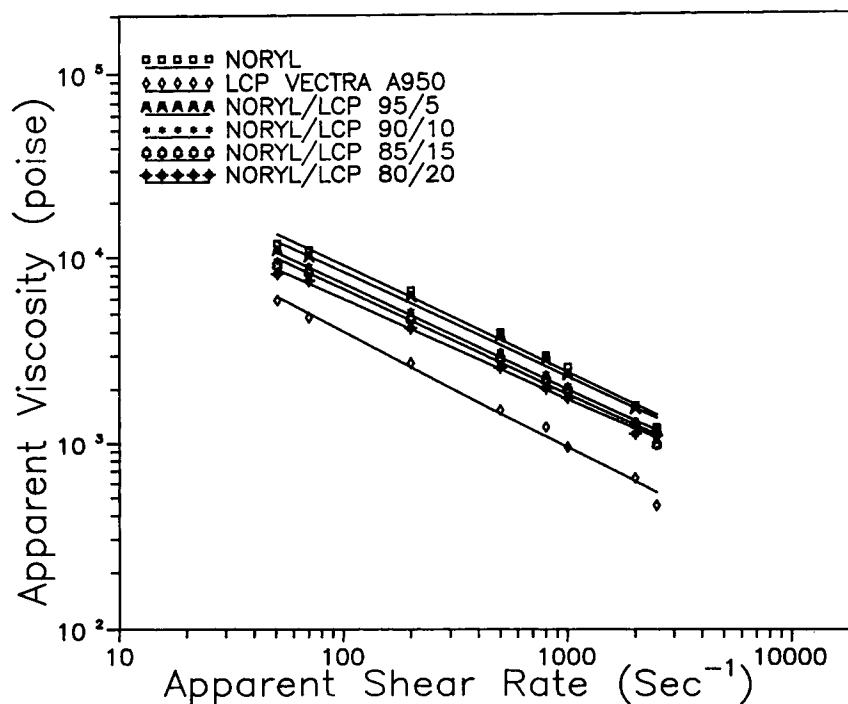


Figure 6 Plots of apparent viscosity vs. shear rate for Noryl and various Noryl/LCP blends.

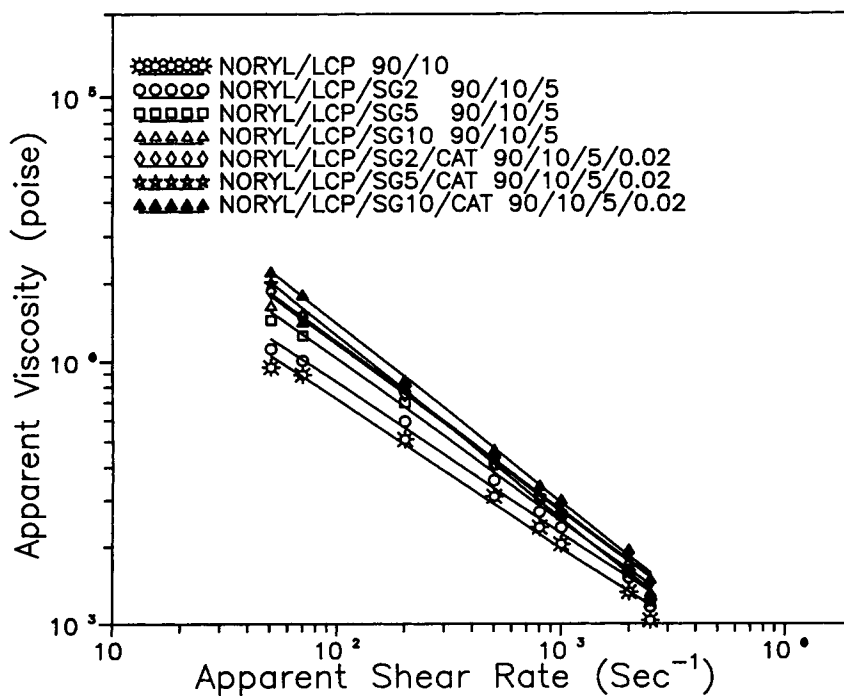


Figure 7 Plots of apparent viscosity vs. shear rate for the noncompatibilized and compatibilize Noryl/LCP = 90/10 blends.

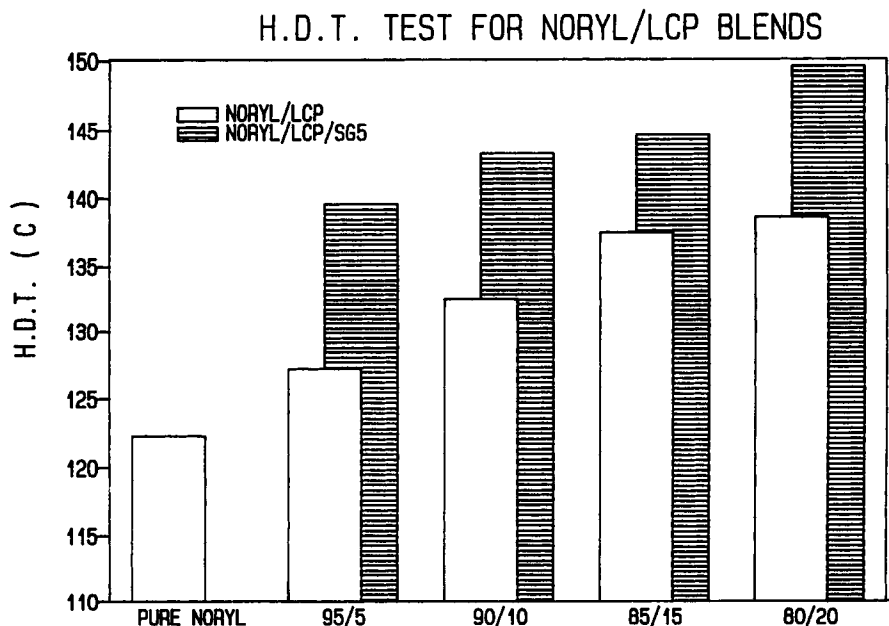


Figure 8 Effect of SG5 compatibilizer on heat deflection temperature of various Noryl/LCP blends.

LCP fibers in Noryl matrix act as a reinforcement agent to increase the material strength and HDT, similar to glass or carbon fiber/polymer composites. As mentioned earlier, most thermoplastic/LCP blends are immiscible and incompatible with poor interfacial adhesion, and the presence of a compatibilizer in the blends is expected to improve the interfacial adhesion between Noryl and LCP and, therefore, give higher HDT.

Tensile and Impact Properties

One of the major objectives of blending LCPs with thermoplastic polymers has been to use the LCP as a reinforcement for the flexible thermoplastic polymers. This thermotropic LCP (Vectra 950) is long-chain fully aromatic molecules that has sufficient chain stiffness and strength to give high stiffness and strength, yet has sufficient flexibility to melt.¹³ Addition of a small amount of LCP in thermoplastics, in general, is able to increase their tensile strength and modulus but decreases their tensile elongation.^{10,16,20,24-26,29} Most of these researchers have attempted to explain the changes in mechanical properties in terms of the morphology of the LCP domains in the blends. The two component polymers are immiscible in most of the studies previously reported on LCP/thermoplastic blends.

In this study, we emphasize the changes in mechanical properties due to the presence of the reactive compatibilizer. Table II summarizes all the

tensile and Izod impact properties, and the observed property improvements due to the presence of compatibilizer and catalyst are very substantial and consistent.

Figure 9 compares the tensile strength of the pure Noryl with compatibilized and noncompatibilized Noryl/LCP blends. The observed tensile strength increases with increasing LCP content. The presence of 5 phr SG5 gains an additional improvement from all the blends. The combination of SG5 and 200 ppm catalyst further increases the resultant tensile strength. Figure 9 demonstrates the effect of SG5 content (with or without catalyst) on the tensile strength. Table II also shows that higher GMA content in the SG compatibilizer results in higher tensile strength. Similar trends also occur in tensile modulus and elongation to break, as shown in Table II. The only exception observed is that the optimized quantity of SG5 is at 5 phr. The blend containing 10 phr SG5 (with or without catalyst) actually decreases its tensile elongation. Figures 10 and 11 show the notched and unnotched Izod impact strength of the compatibilized (with and without catalyst) and noncompatibilized blends, and have an identical trend to that for the tensile properties.

The improvements of the compatibilized blends over the noncompatibilized counterparts in tensile and Izod impact properties are substantial and consistent. Polymer-polymer adhesion plays a significant role in determining the ductility-related properties in any immiscible blend. The extent of local

Table II Summarized Mechanical Properties

Composition	Tensile			Izod Impact	
	Elong. %	Str. MPa	Modulus MPa	Notch J/M	Unnotch J/M
N (Noryl)	15.2	71.6	2940	11.9	160
N/LCP = 95/5	12.3	74.3	3200	13.5	185
N/LCP/SG5 = 95/5/5	13.7	77.6	3390	14.5	194
N/LCP/SG5/Cat = 95/5/5/0.02	14.5	81.6	3590	16.2	210
N/LCP = 90/10	12.1	77.0	3550	14.1	173
N/LCP/SG2 = 90/10/5	12.4	80.4	3650	14.9	189
N/LCP/SG2/Cat = 90/10/5/0.02	13.2	82.8	3740	16.5	215
N/LCP/SG5 = 90/10/2	12.3	78.1	3690	15.1	186
N/LCP/SG5/Cat = 90/10/2/0.02	13.2	81.9	3823	16.5	217
N/LCP/SG5 = 90/10/5	12.6	81.3	3740	16.0	199
N/LCP/SG5/Cat = 90/10/5/0.02	13.9	84.6	4000	17.1	226
N/LCP/SG5 = 90/10/10	13.1	83.5	3780	15.7	223
N/LCP/SG5/Cat = 90/10/10/0.02	13.4	85.1	4020	16.0	222
N/LCP/SG10 = 90/10/5	13.2	82.7	3850	18.5	213
N/LCP/SG10/Cat = 90/10/5/0.02	13.9	86.2	4190	21.0	235
N/LCP = 85/15	11.8	80.7	3990	16.2	170
N/LCP/SG5 = 85/15/5	12.4	82.5	4060	18.0	212
N/LCP/SG5/Cat = 85/15/5/0.02	13.4	91.7	4165	19.2	236
N/LCP = 80/20	9.7	82.5	4110	18.7	164
N/LCP/SG5 = 80/20/5	11.4	84.6	4460	21.2	220
N/LCP/SG5/Cat = 80/20/5/0.02	12.7	92.3	5020	22.3	242

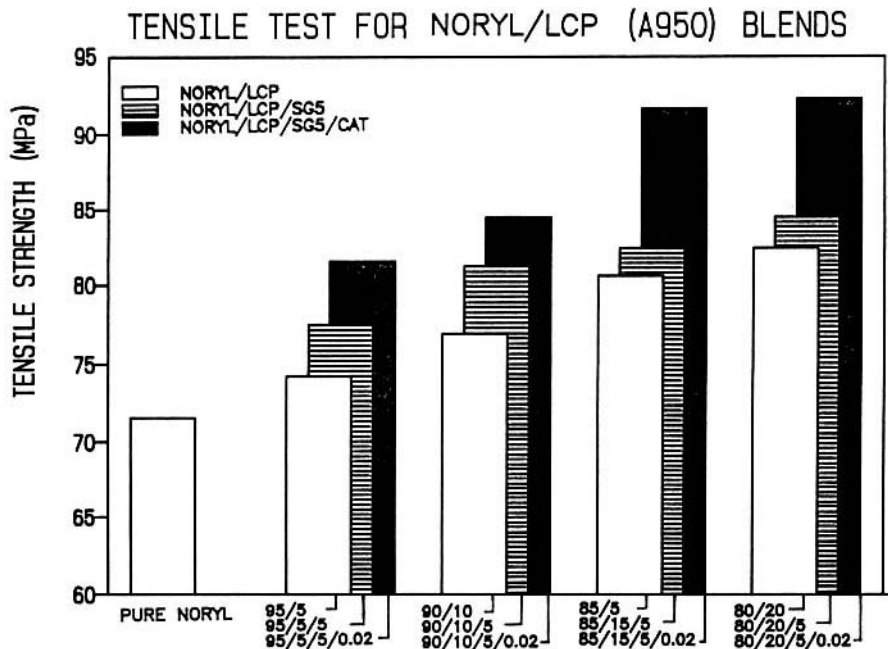


Figure 9 Tensile strength of Noryl, and various noncompatibilized and compatibilized Noryl/LCP blends.

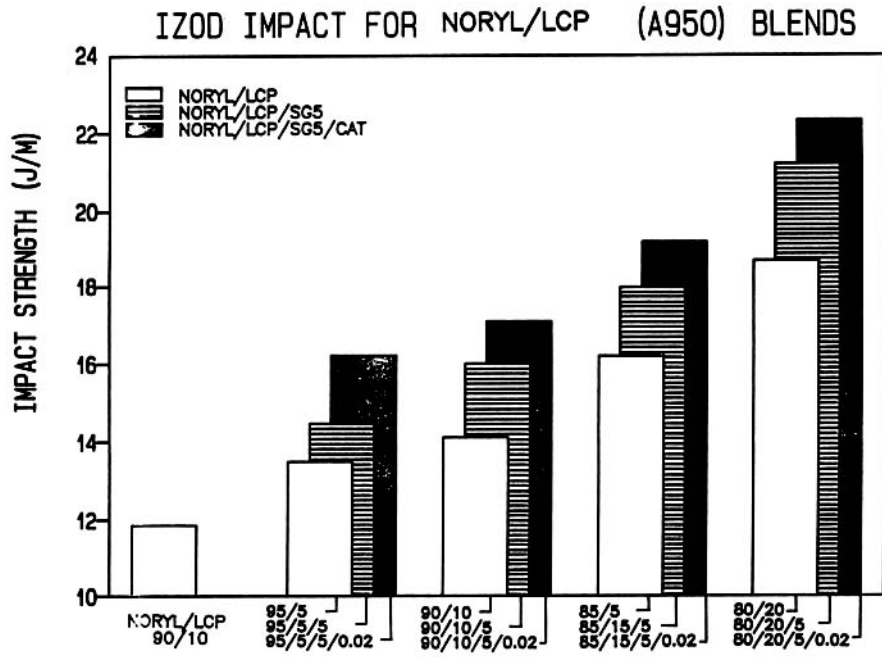


Figure 10 Notched Izod impact strength of various noncompatibilized and compatibilized Noryl/LCP blends.

segment mutual diffusion across the interface between the blend constituents critically affects the mechanical strength of the adhesive bond.³⁸ The improved properties consistently observed for the compatibilized blends over the noncompatibilized

counterparts in this study can be partially attributed to better interfacial adhesion. The LCP fiber structural difference between the noncompatibilized and compatibilized blends (details later) is also believed to be responsible for the observed difference. The

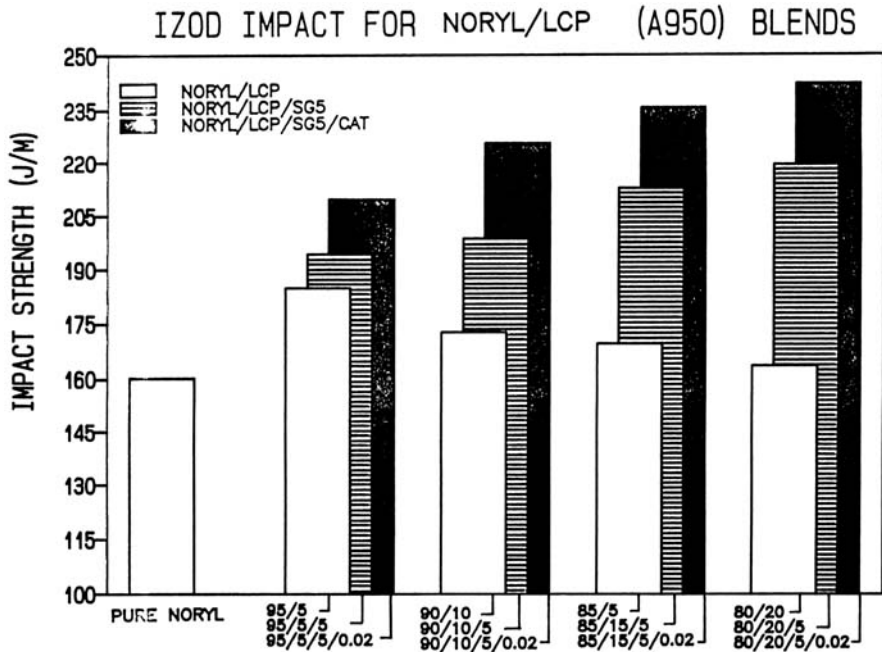


Figure 11 Unnotched Izod impact strength of various noncompatibilized and compatibilized Noryl/LCP blends.

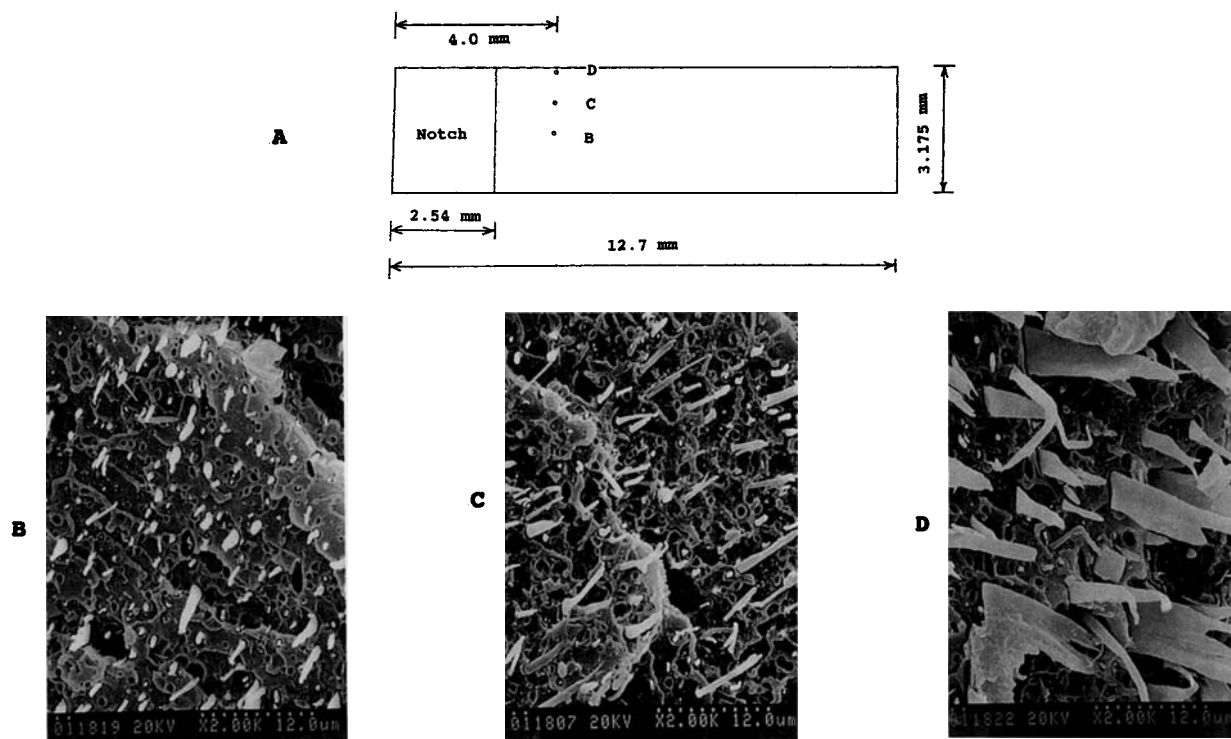


Figure 12 Core-skin SEM morphologies of the fractured surfaces of the noncompatibilized Noryl/LCP = 90/10 blend. (A) Specimen diagram showing core to skin locations where the SEM morphologies were taken. (B) SEM morphology at core region, at point B of (A). (C) SEM morphology at midpoint between core and skin, at point C of (A). (D) SEM morphology close to the skin region, at point D of (A).

LCP fibers of the compatibilized blends are relatively finer and longer (greater orientation) and tend to be in the form of lamellae. The stiffness and strength of the formed LCP fibers increase with the increase of orientation. Therefore, the LCP fibers in the compatibilized blends have relatively higher strength and greater interfacial contact area (higher aspect ratio) with the Noryl matrix than the corresponding noncompatibilized blends.

Scanning Electron Microscopy (SEM)

The size, shape, and distribution of the LCP domains depend on many factors such as composition, viscosity ratio of the component polymers, interfacial tension, the rheological characteristics of the matrix polymer, and the processing conditions. Because most of the previous studies on LCP/thermoplastic involved only noncompatibilized systems, interfacial tension was not treated as a factor in dictating the morphology of the LCP domains.

In this study, our emphasis is the morphological changes resulting from reducing the melt interfacial

tension due to the presence of the reactive compatibilizer. Undoubtedly the presence of the compatibilizer can also change the viscosity ratio of the component polymers and their individual rheological characteristics. Here, morphological studies on Noryl/LCP blends were undertaken to correlate the mechanical properties with the morphologies of these blends and to study the dimension and distribution of the LCP fibers formed in the Noryl matrix.

Figure 12 gives micrographs of the Noryl/LCP = 90/10 blend at different locations, from core to skin of the injection-molded specimen. The skin-core morphologies of the LCP dispersed in thermoplastic polymers have been reported previously.^{2,4,8,19,22,23,25} Point C in Figure 12(A), the midpoint between central line and skin of the specimen, is the region where all the SEM micrographs were taken in this study. At the core region [Point B in Fig. 12(A)], the LCP fibers are in the form of relatively larger ellipsoidal droplets or rod-like structures, as shown in Figure 12(B). The LCP fibers at point C region [Fig. 12(A)] are relatively finer and

longer, as shown in Figure 12(C), because of relatively higher shear at this region than in the core region during injection molding. Near the skin region, with its highest expected shear [Point D, Fig. 12(A)], most of the LCP fibers are in the form of elongated and wide lamellate domains, as shown in Figure 12(D). Such a lamellate structure provides greater interfacial contact area between LCP and Noryl matrix than a typical cigar-type structure and, therefore, superior fiber reinforcement is expected. Such lamellate LCP domains near the skin region should also possess better barrier properties. The LCP has the tendency to concentrate heavily near the skin region [compare Figs. 12(D) with (B) and (C)] because the LCP has relatively lower viscosity than that of the Noryl matrix. Similar results, wherein LCP is concentrated near skin, have also been observed previously.^{2,4}

Figure 13 shows the morphologies of the compatibilized (with and without catalyst) and noncompatibilized Noryl/LCP=85/15 blends. The noncompatibilized blend [Fig. 13(A)] shows the relatively coarser cigar-like LCP fibers and the interfacial adhesion with Noryl matrix appears poor. The LCP fibers become finer and wider when the blend contains 5 phr of SG5 compatibilizer [Fig. 13(B)]. In addition, the gaps between the fibers and the matrix are significantly reduced, an indication of improved interfacial adhesion. The LCP sheet-like fibers now become wider and thinner in the blend containing 5 phr SG5 and 200 ppm catalyst, which are similar to the lamellate fibers occurring near skin region of the noncompatibilized blend

[compare Figs. 13(C) vs. 12(D)]. The gaps between the lamellate fibers and the matrix essentially disappear, an indication of further improvement of the interphase adhesion. The same trend was also observed for other blend series with other Noryl/LCP ratios (data not shown here). It is interesting to notice that a very similar trend of the LCP fiber morphology in Noryl matrix was obtained from the higher shear region of the specimen and from the better compatibilized blend. The *in situ*-formed grafted copolymer can reduce the interfacial tension of the blend at the melt, and the dispersed LCP phase can better respond to the applied shear force to deform into lamellate domains for a better compatibilized blend.

CONCLUSIONS

An *in situ* reactive compatibilizer has been successfully used to turn many otherwise incompatible thermoplastic blend pairs into compatible blends. Copolymers possessing monomer units with epoxy group have been applied in several selected polymer pairs with different degrees of success. The reactive copolymer, styrene-glycidyl methacrylate (SG), has demonstrated to be effective to improve the processability, heat deflection temperature, and various mechanical properties in this thermoplastic (Noryl)/LCP blend. This SG copolymer can react with LCP end groups to form SG-g-LCP copolymers, which will function as a phase compatibilizer for the Noryl/LCP blends. The compatibilized blends slightly in-



Figure 13 SEM morphologies of the fractured surfaces of the noncompatibilized and compatibilized Noryl/LCP = 85/15 blends. (A) Noryl/LCP = 85/15. (B) Noryl/LCP/SG5 = 85/15/5. (C) Noryl/LCP/SG5/Cat = 85/15/5/0.02.

crease the viscosity but do solve extrusion problems such as die swelling and melt fracture. The LCP fibers in the Noryl matrix of the compatibilized blends become finer and longer and tend to form lamellate structure domains, which can provide greater interphase contacting area. The interfacial adhesion is also improved after compatibilization. The presence of ethyl triphenylphosphonium bromide catalyst promotes the grafting reaction to make the resultant blend even more compatibilized. The change of the LCP morphology and the increase of the interphase adhesion of the compatibilized Noryl/LCP blend are believed to be responsible for the observed property improvement

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