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Structure and properties of injection molded blends of poly(ether imide) and a thermotropic copolyester

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The phase structure and mechanical properties of blends composed of poly(ether imide) and a thermotropic liquid-crystalline copolyester, Rodrun LC-5000, processed by injection molding were studied in the PEI-rich region. The addition of Rodrun to PEI produces fully immiscible blends and strongly improves the processability of PEI. Injection molding gave rise in very poor LCP blends to fine dispersed phases; in the rest of compositions it gave rise to coarse structures. This was probably due to the very large viscosity difference between the components of the blends. An increase in the melt temperature led to a smaller viscosity difference and as a consequence dispersed phase dispersion improved. The Young's modulus clearly increased with the Rodrun content. The tensile strength was maintained up to a LCP content of 5% but decreased at higher contents because of the coarse morphologies obtained. The overall mechanical behavior was a main consequence of the lack of fibrillation and of the LCP dispersion level in the matrix.

1. Introduction

Polymer blends based on thermoplastics and liquid-crystal polymers (LCP's) are an active research area in polymer science and technology, which has been widely developed in the last years. Some of the reasons for this development are similar to those which have driven the increasing importance of blends of conventional thermoplastics [1]. Among them, cost-reduction and property tailoring are specially interesting. Cost-reduction by blending thermoplastics with LCP's enables properties comparable to those of the LCP's to be obtained at prices near those of thermoplastics. Property tailoring in these blends is even more relevant than in thermoplastics blends [2] due to the fundamental importance of the morphology obtained. It is interesting mainly with three objectives: 1) use of the LCP as a fibrous reinforcement for conventional thermoplastics, 2) addition of low thermoplastic amounts to LCP's, to reduce some LCP handicaps, such as low weld-line strength or property anisotropy, and 3) use of the LCP as a processing aid for high viscosity conventional polymers. These approaches to thermoplastic/LCP blends have been extensively studied in the literature [3–7], for blends composed of a wide variety of commodity, engineering, and high-performance thermoplastics.

Poly(ether imide), PEI, is a high performance thermoplastic, which combines favorable mechanical and electrical properties, as well as good chemical and flame resistance. PEI is commercialized in the neat state, but also in short glass fiber and carbon fiber-reinforced composites. The reinforced forms enhance mechanical properties such as stiffness and strength, but suppose high friction and wear in the processing equipment. This problem may be overcome by blending PEI with LCP's that may act, in PEI-rich compositions, both as reinforcing agents and processing aids. Blends of PEI with different LCP's have been widely studied in the literature [6–14].

Rodrun LC-5000 is a thermotropic copolyester based on *p*-hydroxy benzoate and ethylene terephthalate units in an approximate 80/20 composition [15] that has a more random structure than conventional LCP's [16]. This more random structure leads to a favorable property profile which includes high mechanical properties, chemical resistance, HDT higher than that of conventional LCP's, and extremely low diffusion and thermal expansion coefficients.

In spite of these favorable properties, blends based on thermoplastics and Rodrun LC-5000 have received little attention in the literature. The rheological behavior, morphology and mechanical properties of blends with polystyrene [17] and bisphenol-A polycarbonate (PC) [18], as well as the effect of a compatibilizer on PC/Rodrun LC-5000 blends [18], and the acid-base interactions in PEI/Rodrun LC-5000 blends [19] have been recently studied. However, no work has been carried out to our knowledge on the structure and physical properties of blends of Rodrun with PEI. These are the reasons why in this work, besides the processability and phase structure, the morphology and mechanical

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properties of directly injection molded PEI/Rodrun LC-5000 blends have been studied. The phase structure of the blends was characterized by DSC and the morphology by SEM. The mechanical properties were determined by means of tensile tests and related to the processing induced structure.

2. Experimental

The polymers used in this work were commercial products. PEI was Ultem 1000®, from General Electric Plastics, and was supplied by Polymerland Guzman, S. A. (Valencia, Spain). The molecular weights of PEI are $M_W = 30{,}000$ and $M_n = 12{,}000$. The LCP was Rodrun LC-5000®, and was supplied by Unitika Ltd. (Tokyo, Japan). It has a 80/20 *p*-hydroxy benzoate (PHB)/ethylene terephthalate (PET) composition. It has an intrinsic viscosity of 0.552 dl/g, as determined at 30 °C in a phenol/tetrachloroethane (50/50) mixture, and a Melt Flow Index (MFI) of 25 g/10 min, measured at 290 °C with a 325 g load.

PEI and Rodrun were dried following the producers' recommendations for 8 h at 135 °C before processing in order to avoid degradation reactions caused by moisture. To analyze the processability of the blends, they were mixed in a Brabender mixing head attached to a Brabender PLE 650 Plasticorder. Mixing was carried out at 330 °C and 30 rpm for a time of 12 min, when the steady torque indicated the formation of an homogeneous blend. The compositions studied were the same as those of injection molding i.e., with LCP contents of 5, 10, 20, 30 and 40%. With the aim of using the values as a reference, both the neat PEI and Rodrun were subjected to the same molding procedures as the blends.

Direct injection mixing was carried out during the plasticization step of a reciprocating screw injection molding machine Battenfeld BA230E. The screw had a diameter of 18 mm and a length/diameter relation of 17.8. No specific mixing sections were present. Three melt temperatures were used: 300, 330 and 350 °C (named I300, I330 and I350 respectively). Higher melt temperatures were not used because of the possibility of Rodrun degradation after long residence times at high temperatures. The mold provided tensile specimens according to ASTM D638, type IV. The standard injection speed and pressure were 23.0 cm^3/s and 2850 bar respectively, and the mold temperature was 85 °C.

The phase behavior of PEI/Rodrun blends was analyzed by differential scanning calorimetry (DSC), using a Perkin-Elmer DSC-7 calorimeter. A first scan was carried out from 30 to 330 °C at 20 °C/min in order to destroy the previous thermal history. After cooling at the maximum speed, a second scan was carried out. The glass transition and melting temperatures (T_g and T_m respectively) and the melting heat of Rodrun (ΔH_m) were determined in the usual way.

The morphology of the blends was studied by scanning electron microscopy (SEM) on tensile fractured surfaces, after gold coating. A Hitachi S-2700 microscope was used at an accelerating voltage of 15 kV.

Tensile tests were carried out on the injection molded specimens in an Instron 4301 tensile tester at room temperature. The mechanical properties (Young's modulus, tensile strength, and ductility, measured as the break strain) were determined from the load-elongation curves. A minimum of five specimens was tested for each reported value.

3. Results and discussion

3.1. Processability and phase structure

As stated previously, one of the advantages of the thermoplastic/LCP blends is their low viscosity with respect to that of the neat thermoplastic. This is due to the very low viscosity of LCP's which enables them to act as processing aids. In order to check this effect in PEI/Rodrun blends, the torque required to turn the Brabender mixer, which is related to melt viscosity [20], was measured at all blend compositions. The results are reported in Fig. 1. As is observed and was expected, the torque of the LCP at 330 °C (0.1 N·m) is very small compared to that of PEI. The torque reduction after the addition of only a 10% Rodrun is more than 50%, and the value after the addition of 20% Rodrun is a quarter of that of PEI. For blends with LCP contents of 30 or 40%, the torque, and consequently the blend viscosity, are of the same order as that that of pure Rodrun.

These results are in good agreement with those found for other thermoplastic/LCP systems [5] and quantify the processing aid ability of Rodrun for PEI. This effect is attributed in thermoplastic/LCP blends to phase separation and to the fact that within a flow field, the elongated and very low viscosity LCP particles enable easy melt flow, leading to melt lubrication. This effect is increased as the contact surface increases and thus, when a fibrous morphology is attained, the melt viscosity can attain values even lower than that of the LCP [3, 21]. In some cases [6], on increasing the LCP concentration, spontaneous phase coalescence will occur, and the interfacial area and slip will decrease giving rise to a rise in the blend viscosity. This effect is not seen, however, in the PEI/Rodrun blends of the present work, probably because of the LCP content that was limited to 40%.

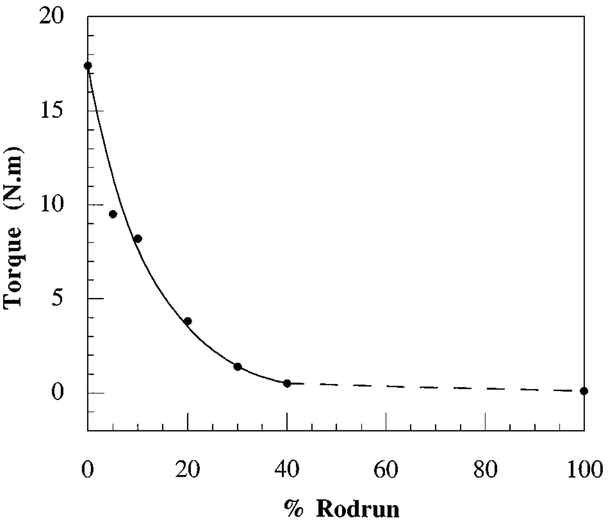


Figure 1 Torque-composition relationship for PEI/Rodrun blends at 330 °C.

TABLE I T_g's of the blends and T_m and ΔH_m of Rodrun in the blends

	I330 Blends			I350 Blends		
PEI/Rodrun	T_g (°C)	T_m (°C)	ΔH_m (J/g)	T_g (°C)	T_m (°C)	ΔH_m (J/g)
100/0	215			215		
95/05	207			210		
90/10	206			210		
80/20	206	276	0.7	211	279	2.0
70/30	208	276	0.6	211	282	0.7
60/40	209	279	0.9	206	277	1.7
0/100	59	274	3.4	58	273	3.0

The phase structure characterized by the T_g, T_m and ΔH_m was analyzed by DSC and is shown in Table I. The values corresponding to the Brabender mixed blends were similar to those of the injected blends. The I300 blends appeared not homogeneous enough for repetitive DSC scans to be obtained. For this reason only the T_g, T_m and ΔH_m values of the I330 and I350 blends are collected in Table I. The T_g of neat PEI appears at approximately 215 °C irrespective of the melt temperature. Rodrun shows a T_g at approximately 58 °C, which corresponds, according to previous results [15], to an ethylene terephthalate-rich phase of the thermotropic copolyester. A second wide transition of Rodrun, at approximately 150 °C [15], was not clearly observed.

With respect to the blends, a possible T_g of a Rodrun-rich phase close to 58 °C was not observed, probably due to the small specific heat increase associated with the transition. However, an approximately constant T_g, slightly lower than that of pure PEI (215 °C), is found for all the compositions studied. This indicates the presence of a rather pure PEI phase in the blends. A similar slight decrease in the T_g of PEI was seen in PEI/Vectra B950 blends [13] and was attributed to a more active movement of the LCP chains in the blends. The full immiscibility of the PEI/Rodrun blends agrees with results found for other PEI blends with different LCP's [8, 9, 11, 13, 14, 22], although partial miscibility has also been observed in other PEI-based systems [11, 23, 24].

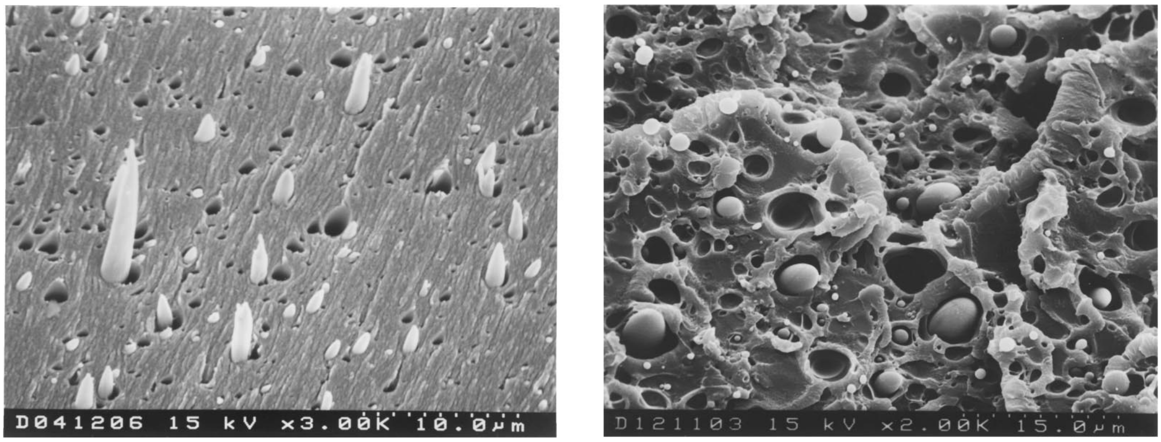
With respect to the melting endotherm of Rodrun, its very small enthalpy, near 3 J/g, indicates its very low crystallinity level. Rodrun crystallinity seems to be slightly smaller in the blends than in the neat Rodrun. However, the determination of the melting heat of Rodrun was not accurate due to the small value and the irregular shape of the endotherm, which makes the choice of baseline difficult. Thus, blending increases processability and PEI/Rodrun blends appear as fully immiscible.

3.2. Morphology

The tensile fracture surfaces of the I350 and I330 blends were studied by SEM. All the blends showed an oriented and not fibrillated skin with a coarse structure. The skin thickness was independent of composition but changed with injection temperature from roughly 150 μm in I330 blends to 80 μm in I350 blends. The presence of two phases was not clear so that its chemical nature was tested by trying to dissolve it in chloroform and by DSC of the outside layer. Its insolubility and the DSC scan demonstrated the presence of both Rodrun and PEI.

The fracture surfaces, observed at an angle from the perpendicular to the surface of 30 °, of the I330 95/5 blend close to the skin, and representative morphologies of the 90/10 blend are seen respectively in Figs 2a and b. As can be seen, blending was effective in the 95/5 blend and some elongated dispersed phases appear close to the skin where, as seen in Fig. 2a, Rodrun is present in a content apparently higher than 5%. The core was unorientated with particles smaller than 1 μm. However, in the 90/10 blend of Fig. 2b, the mixing level of the core, and of the skin that was similar, worsened with numerous particles larger than 3 μm. This also took place in the 80/20 blend. In the 70/30 blend, as in the case of the 60/40 blend, large unblended zones were present.

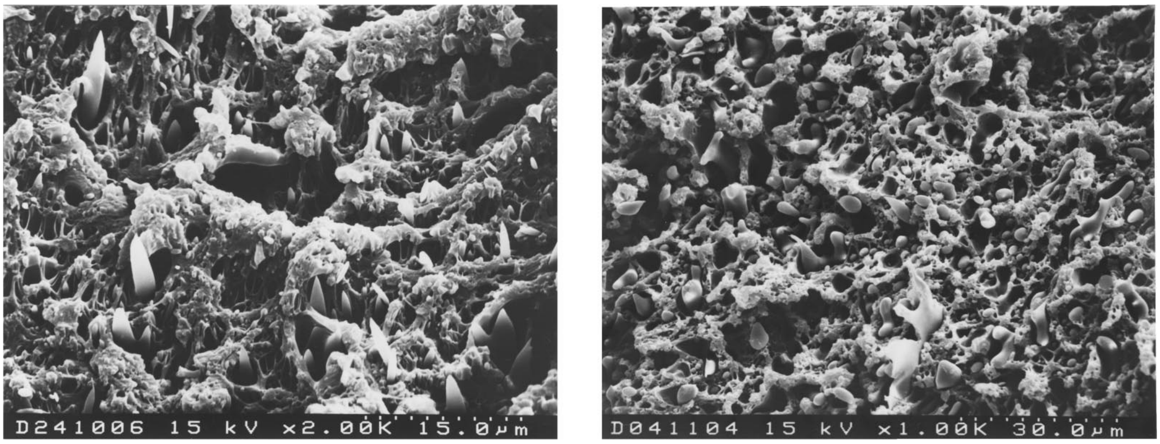
In Figs 3a and b, the zone close to the skin of the 90/10 composition and a representative zone of the 70/30 composition of the I350 blends are shown. As can be seen when Figs 2 and 3 are compared, mixing was more homogeneous in the I350 blends than in the I330 blends. Moreover, both in I330 and I350 blends, the particle size increased with the LCP content. Below



(a)

(b)

Figure 2 Fracture surfaces of the tensile I330 specimens observed by SEM, taken at 30° from the perpendicular to the surface. (a) 95/5, and (b) 90/10 compositions.



(a) (b)

Figure 3 Fracture surfaces of the tensile I350 specimens observed by SEM, taken at 30° from the perpendicular to the surface. (a) 90/10, and (b) 70/30 compositions.

and apart from the skin, the morphology of the 95/5 I350 blend did not show orientation of the dispersed phase, probably due to its too small size. A small particle size avoids the occurrence of a large enough shear strain gradient between the opposite sides of the particle to deform it. In the case of the 90/10 blend, the slight orientation of Fig. 3a is seen across a third of the specimen thickness and typical droplets of a mean size of roughly 2 μm appeared in the core. The orientation of the LCP was smaller and very slight in the case of the 80/20 blend and of the 70/30 blend of Fig. 3b. In the case of the 60/40 blend, the structure was similar to that of Fig. 3b, but with several dispersed LCP sheets (typically 20×3 μm). Thus, a melt temperature increase from 330 to 350 °C is clearly positive for blending quality. This was probably due to the fact that, when temperature increased from 330 to 350 °C and at a shear rate of 10^3 s^{-1} (of the order of that used in injection molding), the melt viscosity of Rodrun decreased only from 5 to 4 Pa s, while that of PEI decreased from 2000 to 800 Pa s. Thus, the smaller viscosity difference at higher temperatures should improve mixing.

As can be seen in Figs 2 and 3, besides the low orientation level and the lack of any fiber and even low aspect ratios, the most relevant morphological fact is the small dispersed phase size at LCP contents smaller than 5–10% and the clearly larger sizes at LCP contents higher than 10%. This fact indicates a high surface tension and the correspondent tendency to coalesce as another reason for the observed behavior. This is because droplet fracture should be easier in LCP-rich blends, where the larger droplet size will influence the Weber number [25, 26]; however, this effect does not take place.

As can also be seen, the adhesion between the matrix and the dispersed phase is low. This is since the surfaces of the dispersed phase are clear and neat, and voids surround the particles. Moreover, breaking of the dispersed phase is never seen. The deficient interfacial adhesion is probably a consequence of the immiscibility of the blend components, as observed by DSC.

Thus, adhesion, as usual in LCP blends, is low and only a slight orientation is obtained. The dispersed phase size obtained, although smaller in I350 blends, appears to be too large in high LCP contents to avoid damage of the mechanical properties of the blends.

3.3. Mechanical properties

The effects of composition and of the injection temperature on the mechanical properties of PEI/Rodrun blends were measured. Fig. 4 shows the tensile Young's modulus of the I330 and I350 blends as a function of composition. The reference lines were drawn from the correspondent moduli of Rodrun, that were, respectively, 11.2 and 9.2 GPa. The smaller modulus of the I350 Rodrun was probably due to degradation. This was proved by intrinsic viscosity measurements that showed a decrease in intrinsic viscosity from 0.52 to 0.47 dl/g as melt temperature increased from 330 to 350 °C.

With respect to the Young's moduli of the blends, they are compared with those predicted by the linear rule of mixtures in Fig. 4. The rule of mixtures is drawn only as a reference line, because it is not applicable in these blends where morphology changes with composition. The continuous and the broken reference lines correspond respectively to the continuous and broken

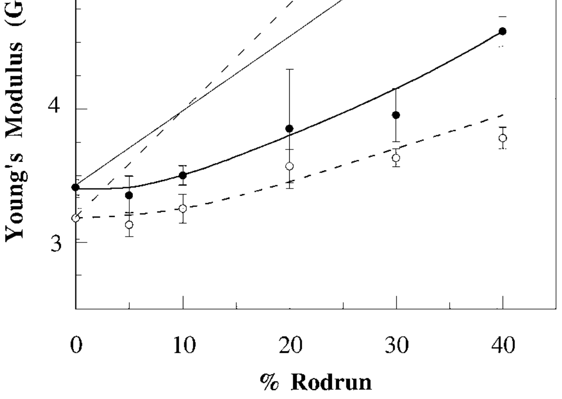


Figure 4 Modulus of Elasticity of PEI/Rodrun blends against the LCP content: (○) I330 and (●) I350.

curves. As can be seen, the modulus of the 95/5 and 90/10 compositions are very similar to that of PEI both in I330 and I350 blends. This is due to the fact that orientation is only slight both in I330 and I350 blends, and agrees with the fact [27] that the increase in modulus of LCP blends is larger when the LCP phase is more orientated. The ulterior increase at higher LCP contents is due to the higher modulus of the LCP related to that of PEI. The higher slope of the plot and the higher values of the I350 blends are probably due to the more effective mixing level and consequently higher LCP contribution to the modulus in the I350 blends. The smaller deviation from linearity of the I350 blends is partially due to the comparatively low reference value of the modulus of the I350 Rodrun.

The large particle size of LCP-rich blends does not decrease the modulus values because of the small elongations at which it is measured, that avoid debonding. The increases in modulus of elasticity indicate that, although a fibrous morphology does not appear, the large LCP dispersed phase effectively contributes to the modulus of the blends.

Generally, the effects of interfacial adhesion in blended systems are more clearly seen at high strain levels, and thus in the break stress and strain. Moreover, ductility is a parameter that, besides modulus in some cases, often determines the tensile strength. For this reason it will be discussed first. The tensile strength results will be then discussed.

Table II shows the ductility of the PEI/Rodrun systems. PEI was a ductile material that after yielding shows cold drawing and stress hardening at the end of the test. Rodrun, as usual in LCP's, was brittle and broke in the linear elastic region of the tensile curves. There is no relevant effect of the melt temperature on the ductility of the pure components. As can be seen, the ductility of PEI/Rodrun blends was clearly smaller than that of PEI, even after the addition of a LCP amount as low as 5%. Consequently, as expected [10, 28–30] there are very large negative deviations with respect to linearity. The larger ductility of the I350 blends is due to their more effective mixing and smaller particle size. The overall low ductility values are due, besides to the small adhesion between the matrix and the dispersed phase seen in the morphology section, to the low ductility of the LCP, mainly in this case when, as usual, the difference in ductility between the thermoplastic component and the LCP is high [10]. Moreover, there is a significant ductility difference between the 95/5 and the rest of the blend compositions. This takes place both in I330 and I350 blends and is due to the clear change of dispersed phase size from the 95/5 to the 90/10 blends that was seen in Figs 2 and 3. An intermediate dispersed phase size was seen in the I350 90/10 blend. As a consequence it also shows an intermediate ductility level.

TABLE II Break strain (%) of I330 and I350 PEI/Rodrun blends

PEI/Rodrun	I330	I350
100/0	50 ± 2	45 ± 6
95/5	7.0 ± 3.0	10 ± 6
90/10	2.2 ± 0.2	4.0 ± 0.5
80/20	2.0 ± 0.1	2.5 ± 0.3
70/30	1.9 ± 0.1	2.4 ± 0.2
60/40	2.0 ± 0.2	2.5 ± 0.1
0/100	2.4 ± 0.1	2.2 ± 0.2

Fig. 5 shows the tensile strength of the pure PEI and of the I330 and I350 blends compared with those of the upper limit, represented by the values of the additive rule. As can be seen, the tensile strength of pure PEI is practically independent of the melt temperature. The variation of the tensile strength of pure Rodrun is qualitatively similar to that of the modulus and can be explained similarly.

With respect to the blends, the main feature observed in Fig. 5 is a decrease in the tensile strength of the blends that takes place between 5 and 10% LCP in the blends. This decrease is clearly related to that observed in the ductility of the blends of Table II and to the increase in the LCP particle size that was seen to take place in Figs 2 and 3 in the 90/10 blends. The larger tensile strength decrease of the I330 blends is due to their higher particle size increase. The constancy in tensile strength at higher LCP contents is due to the fact that the 3–5 μm particles are large enough [31] to clearly produce fracture. The overall higher tensile strength of the I350 blends has to be a consequence of the improved LCP dispersion, in agreement [32] with the fact that smaller dispersed phases are more prone to provide compatible blends due to the increased contact surface and, thus, easier stress transmission, and also of the lesser ability of small dispersed phases to reach the critical size that would induce fracture.

In summary, although the modulus of elasticity of the blends increased above that of PEI with the LCP content, both ductility and tensile strength are clearly below linearity. With the exception of the 95/5 blends that showed small dispersed phase size, this is due to the coarse morphologies obtained. The LCP dispersion and properties improve at the highest melt temperature despite the Rodrun degradation. The overall low dispersion attained indicates that in these blends an improvement of phase dispersion by means of premixing

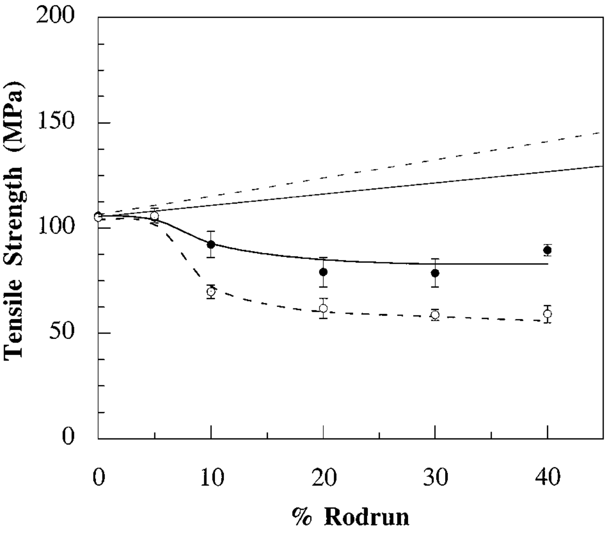


Figure 5 Tensile Strength of the blends against the LCP content: (○) I330 and (●) I350.

or compatibilization, for example, is necessary. This would reduce the possibility of degradation, should improve the dispersed phase dispersion and fibrillation, and ameliorate the mechanical performance of the blends at both low and high processing temperatures.

4. Conclusions

PEI/Rodrun LC-5000 blends are immiscible in the PEI-rich composition region. The dispersed LCP phase size increased with the LCP content, decreased with melt temperature, and generally it was large because of the very different viscosities of the components. Probably for this reason, fibrillation was not produced and the LCP orientation obtained was very slight and detected only at the skin of the specimens very poor in LCP.

The moduli of elasticity obtained were up to 50 to 80% higher than that of PEI, but smaller than those of the linear rule. The decreases in the tensile strength are attributed to the large dispersed particle size that, besides the usual low adhesion level, are the reasons for the very small ductility values. The increase in molding temperature gave rise to increased modulus and strength as a consequence of the improved LCP dispersion, but was not enough to clearly improve the ductility of the blends. A more complex mixing procedure or a compatibilization appear useful in this polymer pair to ameliorate the mechanical performance at large strains.

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