

Performance characteristics of compatibilized ternary Nylon 6/ABS/LCP *in-situ* composites

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Ternary Nylon 6/poly(acrylonitrile-butadiene-styrene)(ABS)/liquid crystalline polymer (LCP) blends compatibilized by the maleic anhydride-grafted polypropylene (MAP) and solid epoxy resin (bisphenol type-A) were injection molded. Thermoplastic matrix consisting of Nylon 6/ABS (60/40) (wt%) was melt blended with various LCP (Vectra A950) contents. The effects of compatibilizer additions on the structure-mechanical property relationship of the *in-situ* composites reinforced with LCP were investigated. SEM observation revealed that the additions of epoxy and MAP compatibilizers to the Nylon 6/ABS/LCP blend were very effective to enhance the interfacial adhesion of its phase components. Consequently, extended long and fine LCP fibrils were formed in the skin section of *in-situ* composites. Tensile measurements revealed that uncompatibilized Nylon 6/ABS/LCP blend exhibited lower strength, stiffness and impact toughness. However, the incorporation of MAP and epoxy resin compatibilizers into Nylon 6/ABS/LCP blend improved its tensile strength, stiffness and impact toughness considerably. This was due to the compatibilizers effectively promoted stress transfer at the interfacial phase regions of ternary *in-situ* composites. The compatibilized composites exhibited mechanical anisotropy, especially for the tensile strength. Thermogravimetric measurements showed that the heat resistance and heat stability of the *in-situ* composites tended to increase with increasing LCP content.

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1. Introduction

Nylon 6 is used in a wide range of engineering applications because of its attractive combination of good mechanical properties and processability. However, Nylon 6 suffers from poor toughness and low dimensional stability. Poly(acrylonitrile-butadiene-styrene) (ABS) terpolymer consists of a two-phase system in which polybutadiene phase disperses as fine particles in continuous styrene acrylonitrile (SAN) matrix. In ABS, acrylonitrile contributes to the heat resistance, chemical resistance and surface hardness of the system. The styrene component improves the processability, rigidity and strength whilst butadiene increases the toughness resistance. The overall properties of ABS are determined by the ratio of monomers present in the system. Several classes of ABS with different impact strengths and special purposes are commercially available. ABS also exhibits certain limitations, namely low thermal stability, poor flame resistance and low mechanical properties compared to other engineering plastics. The above mentioned drawbacks can be overcome by blending ABS with Nylon 6. Blends of ABS and nylon are of technological interests because they inherit superior combination properties from their parent polymers such

as good impact strength, modulus, heat, chemical and abrasion resistances [1–9].

Polymer blending is a useful method to produce new polymeric materials due to their simplicity and comparatively low cost. Liquid crystalline polymers (LCPs) with high strength and stiffness, high chemical resistance, good dimensional stability, low melt viscosity, and low linear thermal expansion coefficient are attractive high performance materials. LCPs consist of long chain rod-like molecules that exhibit an ordered structure in the molten state. Under appropriate processing conditions, the LCP minor phase of LCP/thermoplastic blends is elongated into fine fibrils, leading to the formation of *in-situ* composites. The degree of LCP fibrillation generally depends on the LCP content, the viscosity ratio between the LCP and thermoplastics, and processing condition. There has been many studies of binary blends of LCP with thermoplastics [10–21]. Fewer studies on ternary blends containing an LCP component with their matrices consisting of two thermoplastic components are reported in the literature [22–24]. *In-situ* ternary LCP/thermoplastic blends present an attractive approach to the development of high performance LCP reinforced

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in-situ composites. Introducing a suitable third polymeric component into the binary LCP/thermoplastic blends is an effective route to improve the mechanical performance of such blends. Recently, Bretas and Baird have studied the mechanical properties of the ternary poly(ether imide)(PEI)/poly(ether ether ketone)(PEEK)/LCP blends. The matrices of this *in-situ* composite are miscible PEI/PEEK blends. They reported that high stiffness can be achieved in such ternary blends containing high LCP content, while high ultimate tensile strength can be obtained with high composition loading of PEI or PEEK [22]. Kwon and Chung have fabricated the PC/poly(ethylene terephthalate) (PET)/LCP blends, and reported that the addition of less than 10% LCP improves the mechanical properties of extruded PC/PET blends [23]. For LCP loading above 10%, the ternary blends exhibited poor mechanical properties owing to the aggregation of LCP in the PC/PET matrix. Sawhney *et al.* [24] have incorporated the LCP into a commercial blend of nylon 6 and ABS (Triax 1180; Monsanto Chemical Company). They indicated that the tensile and flexural properties of the polyblends can be increased by selecting proper processing conditions. These include a lower barrel temperature, greater injection pressure, lower injection speed, higher mold temperature, and a greater residence time in the heated mold.

In general, the properties of heterogeneous polymer blends are dependent upon their microstructural morphology, and the adhesion between the matrix and dispersed phases. Most thermoplastics are immiscible and incompatible with LCPs, leading the reinforcing effect of LCP is below than that expected from the rule of mixtures. For effective stress transferring from the polymer matrix to LCP fibrils, a strong interfacial adhesion between LCP fibrils and the matrix is needed. Compatibilization is usually adopted to enhance the interfacial adhesion in polymer blends. The compatibility of LCP/thermoplastic blends can be achieved either by the modification of the LCP structure or by the addition of compatibilizers. The former involves the incorporation of semi-flexible chain into LCPs [25–27]. Most semi-flexible LCPs are not commercially available, and are generally prepared by synthesis process in various laboratories [27]. In this case, commercially LCP with its backbones consisting of aromatic polyester, e.g., Vectra A950 is commonly used as the reinforcing phase. And the compatibilizers such as epoxy functionalized polymer [11, 17, 18] or maleic-anhydride-grafted polypropylene (MAP) [20, 21] are added to improve the compatibility of the phase components of such *in-situ* composites. More recently, Tjong and Meng have used a solid epoxy resin (bisphenol A type) to compatibilize the PC/PBT blend with LCP. They reported that the epoxy resin addition leads to a finer dispersion of PBT in the PC matrix of a binary PC/PBT blend, and to the formation of elongated LCP fibrils in the PC/PBT matrix [28]. This paper aims to study the effects of compatibilizer additions on the mechanical strength and thermal stability behavior of injection molded nylon 6/ABS blends containing LCP. The

compatibilizers used were solid epoxy resin and maleic-anhydride-grafted polypropylene (MAP).

2. Experimental

2.1. Materials

Nylon 6 (B30S), ABS (type 100) and LCP (Vectra A950) were purchased from Bayer Company (Germany), Toray Plastics (Malaysia) and Hoechst Celanese Company (USA), respectively. The LCP used consisting of 27 mol% of 2,6 hydroxynaphthoic acid (HNA) and 73 mol% of *p*-hydroxybenzoic acid (HBA). Maleic-anhydride-grafted PP (Hercoprime HG 201) was supplied by Himont Company, and abbreviated as MAP in this paper. Solid epoxy resin (NPES) was kindly supplied by Nan Ya Plastics of Taiwan.

2.2. Blending and sample preparation

Nylon 6, ABS and LCP pellets were dried in a vacuum oven at 105°C for 40 h prior to blending. The blend consisting of 95% ABS and 5% MAP (wt%) was prepared in a twin-screw Brabender Plasticorder at 240°C and 30 rpm. The extrudates exiting from Brabender were cut into pellets by a pelletizer. These pellets were designated as ABS(M) in this paper. The matrix of ternary blends was prepared by mixing 100 parts per hundreds (phr) Nylon 6/ABS(M) 60/40 pellets with 2 phr epoxy resin in Brabender at 240°C and 40 rpm. Finally, extruded matrix granules and LCP pellets were mixed thoroughly in a plastic box, and subsequent injection molded into plaques with dimensions of 200 × 80 × 3.2 mm. The LCP contents of the ternary blends were fixed at 5, 10, 15, 25 and 35%, respectively. The barrel zone temperatures of injection molder were set at 285, 290 and 285°C.

The injection molded plaques were cut into dog-bone shaped tensile bars. Notched Izod impact specimens were also prepared from the plaques. Both longitudinal and transverse specimens were used for the tensile and impact tests. For the longitudinal specimens, the length direction was parallel to the mold-filling direction, whilst it was perpendicular to the mold-filling direction for the transverse specimens. In order to study the effect of the compatibilizer additions on the properties of Nylon 6/ABS/LCP blends, uncompatibilized the Nylon 6/ABS 60/40 (wt%) blend containing 25 wt% LCP was also prepared under similar processing conditions.

2.3. Torque viscosity

The melting torques of Nylon 6/ABS(M) and pure LCP were determined with a Brabender Rheometer at a capacity of 50 cm³ and at a fixed speed of 30 rpm. The employed temperatures were 285 and 295°C, respectively. Forty grams of specimens were introduced in a Brabender batch mixer. The test time was 15 min.

2.4. Mechanical measurements

The tensile behavior of the blends was determined using an Instron tester (model 4206) at room temperature under a cross-head speed of 1 mm min^{-1} . At least five specimens of each composition was tested and the average values were reported.

Izod impact specimens with dimensions of $65 \times 13 \times 3.2 \text{ mm}$ were cut from the plaques, and were tested by a Ceast impact pendulum tester. These specimens were sharply notched with a V-shape knife. The radius of curvature for the notch is 0.025 mm . At least five specimens were tested and the average values reported.

Dynamic mechanical analysis (DMA) of the injection molded rectangular specimens with dimensions of $65 \times 13 \times 3.2 \text{ mm}$ were conducted using a Du Pont dynamic mechanical analyzer (model 983) at a fixed frequency of 1 Hz and an oscillation amplitude of 0.4 mm . The temperature studied ranged from 30 to 140°C , and the heating rate employed was 4°C min^{-1} .

2.5. Thermal analyses

Thermal analysis was carried out in a Seiko thermogravimetric analyzer (model SSC/5200). The weight loss versus temperature was measured at a rate of $10^\circ\text{C min}^{-1}$ in a helium atmosphere from 50 to 600°C .

2.6. Morphological observation

The morphologies of the surfaces of the blend specimens were observed in a scanning electron microscope (SEM, Jeol JSM 820). The specimens were cryo-fractured in liquid nitrogen. All the samples were coated with a thin layer of gold prior to SEM observations.

3. Results and discussion

3.1. Torque behavior

Fig. 1 shows the typical torque curves for the Nylon 6/ABS(M) (60/40) blend specimens mixed at 285 and 295°C , respectively. For the purposes of comparison, the torque curves for the pure LCP processed at 285 and 295°C are also shown in this figure. From Fig. 1,

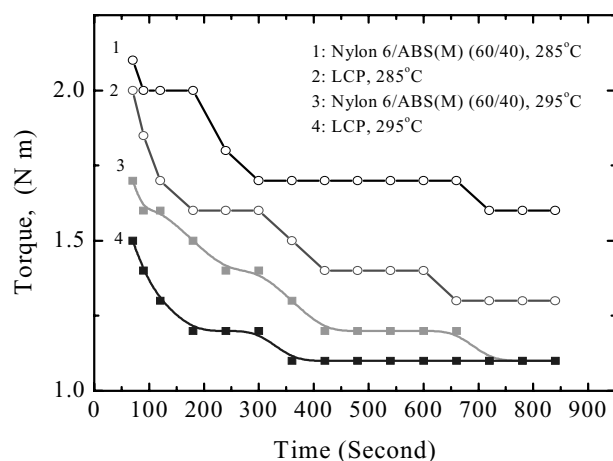


Figure 1 Torque curves of pure LCP and Nylon 6/ABS(M) blend as a function of mixing time at 285 and 295°C , respectively.

it can be seen that the mixing torque of both Nylon 6/ABS(M) blend and pure LCP decreases considerably after $\sim 400 \text{ s}$, thereafter it approaches a nearly steady state level at 285 and 295°C . Moreover, the torque values of Nylon 6/ABS(M) are significantly higher than those of the LCP at 285 and 295°C , respectively. The torque of the melt polymer is roughly related to the melt viscosity of the blend by neglecting the shear strain rates. Accordingly, the viscosity ratio of LCP to Nylon 6/ABS(M) is lower than unity. Such a ratio promotes the formation of LCP fibrils in the Nylon 6/ABS(M)/LCP blends. On the basis of torque results, the barrel zone temperatures of injection molder were set at 285 , 290 and 285°C during injection molding of the Nylon 6/ABS(M)/LCP blends in this study.

3.2. Morphology

In injection molding, the molten polymer is subjected to both elongational and shear flows. The molten polymer flows faster in the center of the mold initially. The flow pattern of the molten polymer from the center towards the surface is called the 'fountain flow'. The fountain melt front in contact with cooled mold wall will freeze immediately. Thus, the mesogen molecules of LCPs tend to align with the flow to form the skin layer of the product. Less oriented material is located in the center of the molding where shear flow predominates, and the solidification occurs more slowly. Fig. 2 is a schematic diagram showing the formation of a 'skin-core' in *in-situ* ternary composites based on the SEM observations. SEM micrographs are taken in the vicinity of skin and core regions, respectively. Fig. 3a-d are the SEM micrographs showing the morphology of the skin region of Nylon 6/ABS(M)/LCP specimens containing different LCP contents. Apparently, fine and elongated LCP fibrils are formed in the skin section, particularly for *in-situ* composites containing higher LCP content. The fibrils are aligned along the molding direction. In contrast, LCP domains are dispersed into ellipsoids in the core region of Nylon 6/ABS(M)/LCP composite (Fig. 4a). Fig. 4b shows the SEM micrograph of the core section of Nylon 6/ABS (M) (60/40) blend. Both Nylon 6 and ABS appear as continuous phases in binary Nylon

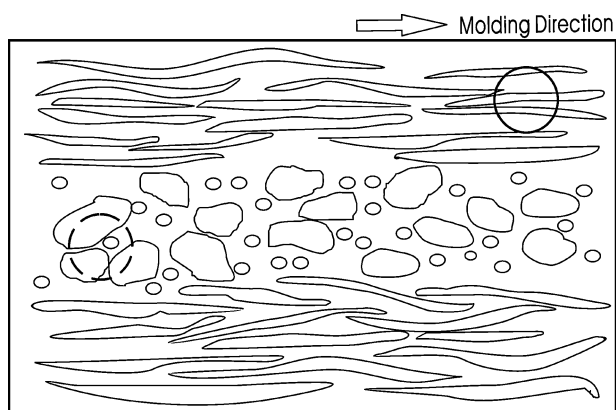
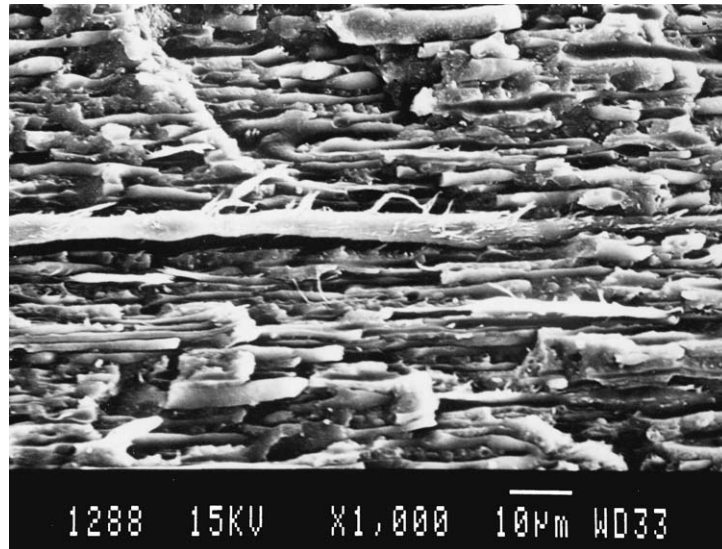
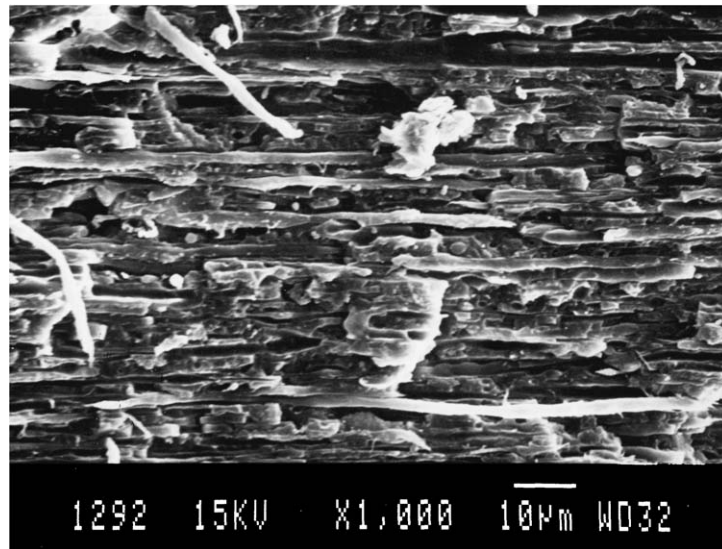


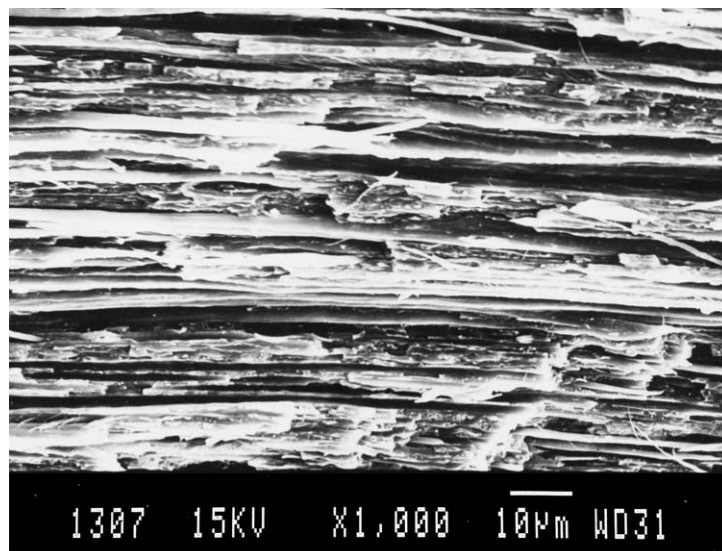
Figure 2 Schematic diagram showing the formation of a 'skin-core' morphology in *in-situ* composite specimens. SEM micrographs are taken in the skin (marked by full circle) and core (marked by dashed circle). The LCP fibrils are aligned along the molding direction in the diagram.



(a)

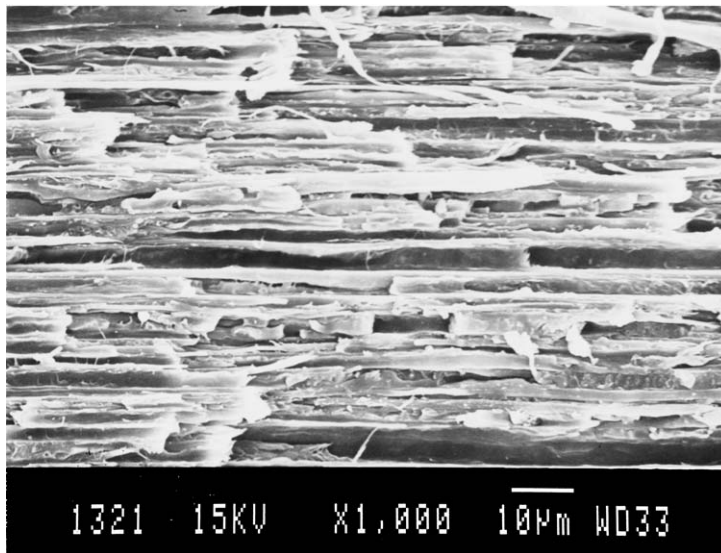


(b)



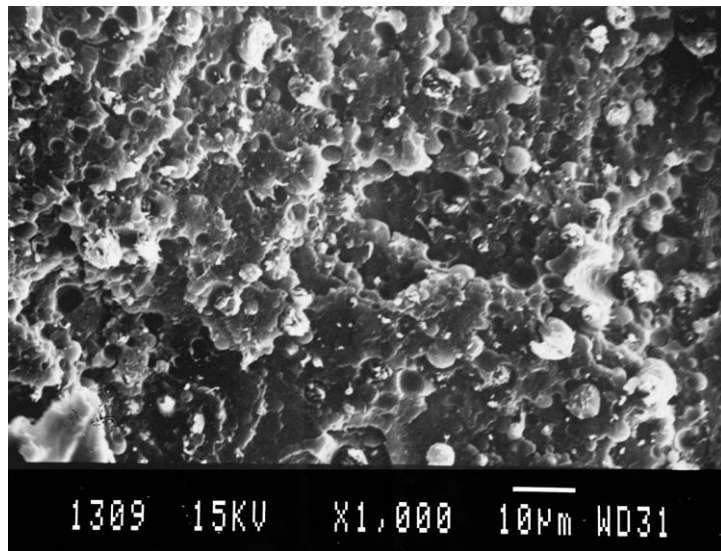
(c)

Figure 3 SEM fractographs showing the skin morphology of Nylon 6/ABS(M)/LCP blends: (a) 5 wt% LCP, (b) 15 wt% LCP, (c) 25 wt% LCP and (d) 35 wt% LCP. (Continued)

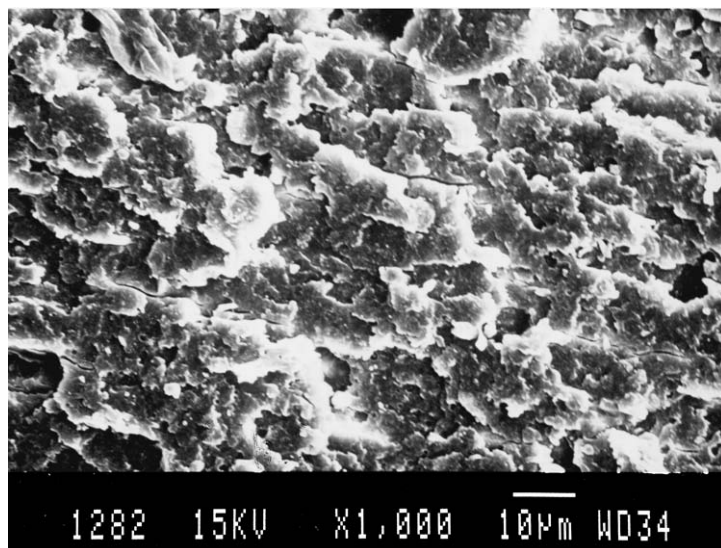


(d)

Figure 3 (Continued).



(a)



(b)

Figure 4 SEM fractographs of the core regions: (a) Nylon 6/ABS(M)/25% LCP blend and (b) Nylon 6/ABS (M) (60/40) blend.

6/ABS (M) (60/40) blends, i.e., no dispersed phase is developed in the blend. This observation confirms that the ellipsoids in Fig. 4a are derived from the LCP polymer.

3.3. Mechanical properties

It is generally known that fiber-reinforced polymer composites exhibit anisotropic mechanical properties. Similarly, ternary *in-situ* composites are believed to exhibit anisotropic mechanical properties associated with the formation of elongated LCP fibrils. Fig. 5 shows the variation of tensile strength with LCP content for Nylon 6/ABS(M)/LCP samples along longitudinal and transverse directions. Apparently, the tensile strength of the longitudinal samples increases with increasing LCP content. However, the tensile strength of the transverse sample appears to decrease with increasing LCP content. Fig. 6 shows the variation of Young's modulus with LCP content for Nylon 6/ABS(M)/LCP blends in both longitudinal and transverse directions. It can be seen that the stiffness of all longitudinal specimens tends to increase continuously with increasing LCP

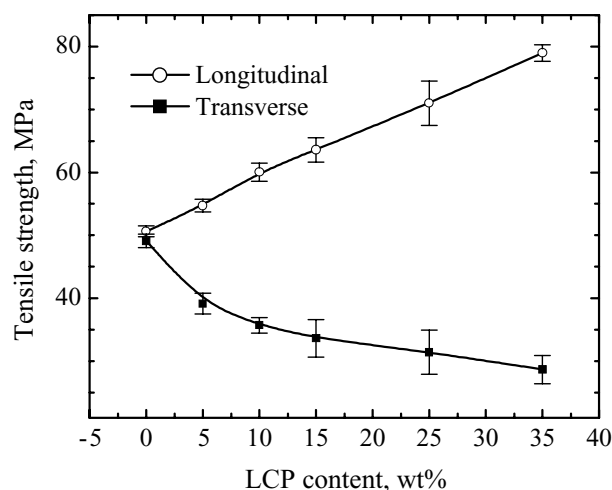


Figure 5 Variation of tensile strength with LCP content for Nylon 6/ABS(M)/LCP blends.

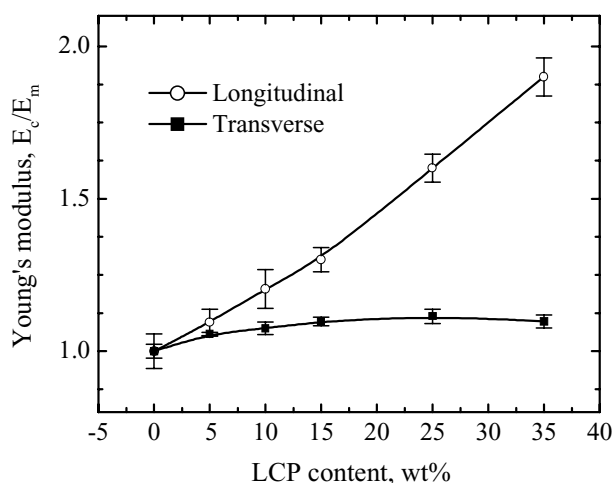


Figure 6 Variation of Young's modulus with LCP content for Nylon 6/ABS(M)/LCP blends.

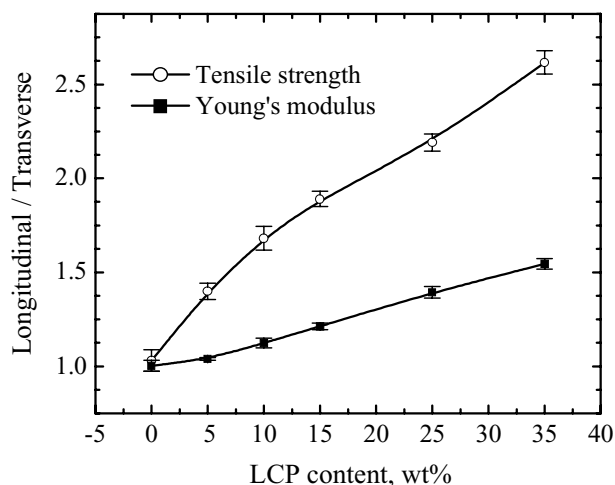


Figure 7 The ratios of tensile strength and Young's modulus of the longitudinal sample to those of the transverse sample versus LCP content for Nylon 6/ABS(M)/LCP blends.

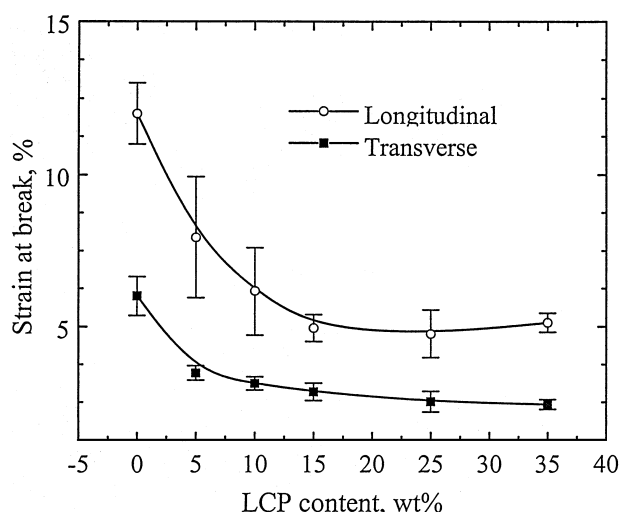


Figure 8 Variation of strain at break with LCP content for Nylon 6/ABS(M)/LCP blends.

content, whereas it remains nearly unchanged for the transverse samples. Accordingly, the tensile anisotropy can be expressed in terms of the ratio between the tensile strength (or stiffness) of the longitudinal samples to that of the transverse samples (Fig. 7). It is evident that the mechanical anisotropic properties increase markedly with increasing LCP content, especially for the tensile strength. This is a typical behavior of the polymer composites reinforced with fibrous fillers. The results imply that the LCP phase is elongated to long fibrils in *in-situ* composites. The variation of strain at break with LCP content for longitudinal and transverse Nylon 6/ABS(M)/LCP is shown in Fig. 8. This figure reveals that the strain at break for both longitudinal and transverse samples falls sharply with the addition of LCP up to 15 wt%. Thereafter, they decrease slowly with increasing LCP content. Fig. 9 shows the variation of impact strength with LCP content for Nylon 6/ABS(M)/LCP samples. Apparently, the transverse impact strength decreases with the addition of only 5 wt% LCP, thereafter it remains unchanged with increasing LCP content. The longitudinal

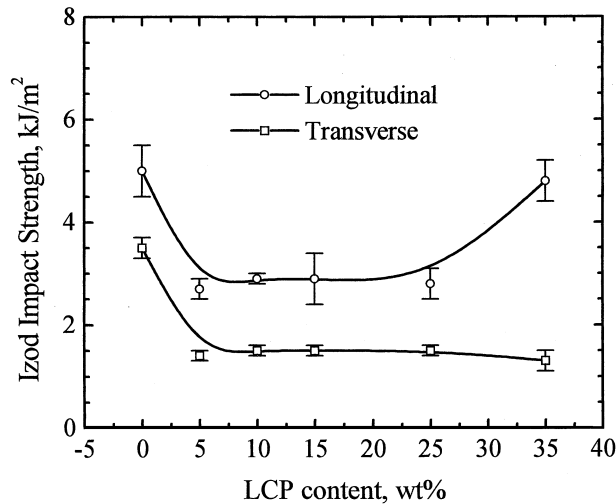


Figure 9 Variation of Izod impact strength with LCP content for Nylon 6/ABS(M)/LCP blends.

impact strength also exhibits a similar decreasing trend behavior up to 25 wt% LCP, but it shows an increase when the LCP content reaches 35 wt%. Such an improvement of impact toughness at high LCP content is commonly observed in polymer composites. It is attributed to the enhanced fibrillation of the LCP phase at higher LCP concentration [13], and to the enhanced matrix plasticity at fibril ends in the crack tip region where the fibrils are closely spaced [29].

We now attempt to correlate the longitudinal modulus (Fig. 6) with theoretical predictions. The relationships most commonly used to predict the elastic modulus of a discontinuous short fiber reinforced composite from the moduli of the individual components is the Tsai-Halpin equation [30, 31]. Crevecoeur and Groeninckx [32], and Tjong *et al.* [33] reported that this equation could be also used to evaluate the modulus of the *in-situ* blends reinforced with LCP. The Tsai-Halpin equation is given as follows,

$$\frac{E_c}{E_m} = \frac{1 + \xi \eta \varphi_f}{1 - \eta \varphi_f} \quad (1)$$

where E_c and E_m are the elastic moduli of blend and matrix, respectively, and φ_f is the volume fraction of short fibers, η is given by

$$\eta = \frac{E_f - E_m}{E_f + \xi E_m} \quad (2)$$

where E_f is the modulus of the fibers, and $\xi = 2(L/D)$. L/D is the aspect ratio L/D (length/diameter) the LCP fibers in polymer matrix. The volume fraction φ_f of the fibers is defined as,

$$\varphi_f = \frac{W_f \rho_m}{W_f \rho_m + (1 - W_f) \rho_f} \quad (3)$$

where W_f , ρ_f and ρ_m refer to the weight fraction of the fibers, density of the fibers and density of the matrix, respectively. Tsai-Halpin equation is generally used to characterize the stiffness of short-fiber rein-

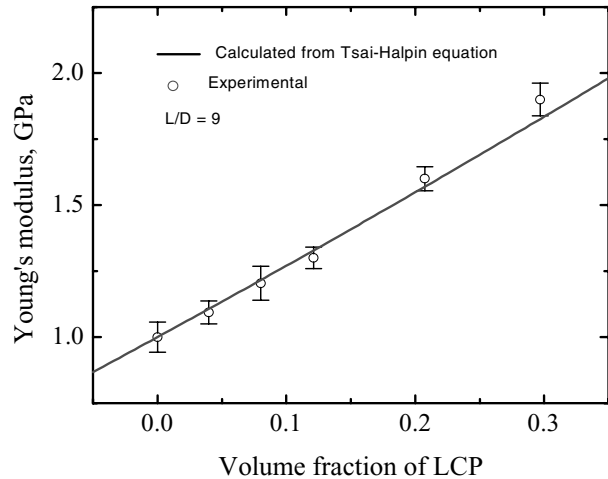


Figure 10 Experimental and theoretical plots showing the variation of longitudinal Young's modulus with LCP content.

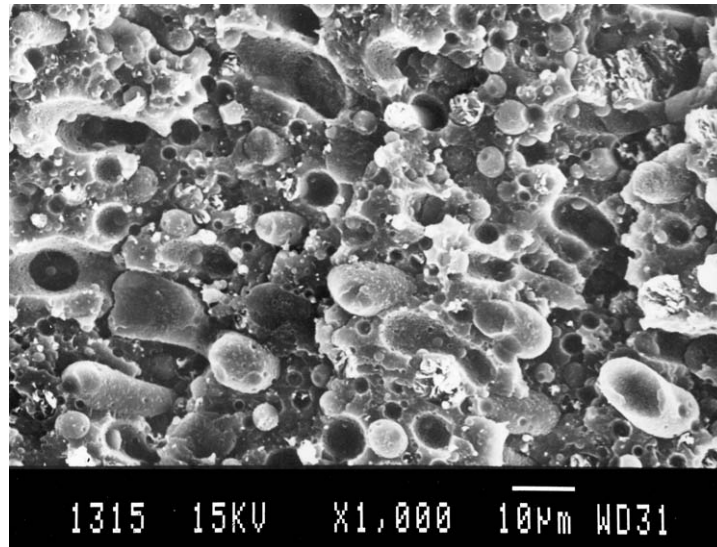
forced composites in which the matrix is a homopolymer. In this study, we consider that the matrix components of *in-situ* composites are compatibilized by the solid epoxy and MAP as discussed in next section. For Nylon 6/ABS(M)/LCP blends, the density of the matrix can be calculated from the composition and the densities of Nylon 6 (1.14 g/cm^3) and ABS (1.04 g/cm^3). In the present composition (Nylon 6/ABS 60/40), $\rho_m = 1.10 \text{ (g/cm}^3)$. For the LCP fibrils, $\rho_f = 1.40 \text{ (g/cm}^3)$, $E_f/E_m = 4.1$. The theoretical prediction based on Tsai-Halpin equation is shown in Fig. 10. It can be seen that the experimental data correlate well with the theoretical prediction using $L/D = 9$ criterion.

3.4. Effects of compatibilizer

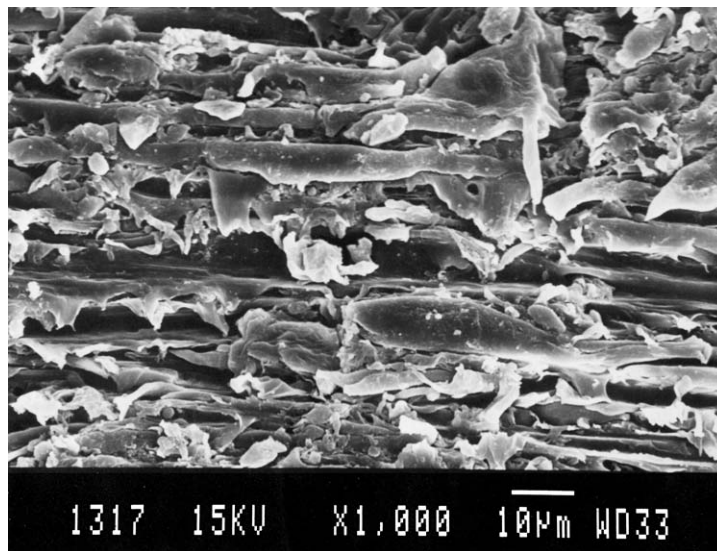
It is generally known that poor adhesion between multi-components of polyblends can result in inferior mechanical properties. In ternary Nylon 6/ABS/LCP blends, we use solid epoxy and MAP to improve the compatibility between the blend components. The effects of the compatibilizers additions on the tensile and impact properties of a selected Nylon 6/ABS/25 wt% LCP blend are listed in Table I. The incorporation of solid epoxy resin and MAP leads to an increase of 27% and 13% for the tensile strength and stiffness, respectively. This is due to the compatibilizers promote effective stress transfer at the interface, leading to considerable improvements of the tensile strength and stiffness as well as adequate enhancement of Izod impact strength.

TABLE I Effects of MAP and epoxy additions on the longitudinal mechanical properties of Nylon 6/ABS/25 wt% LCP blend

Sample	Tensile strength (MPa)	Strain at break (%)	Young's modulus (GPa)	Izod impact strength (kJ/m²)
Without compatibilizers	56 ± 3	4.1 ± 0.3	1.57 ± 0.08	2.3 ± 0.2
With MAP	60 ± 3	4.4 ± 0.4	1.60 ± 0.09	2.8 ± 0.2
With MAP and epoxy	71 ± 3	4.8 ± 0.4	1.77 ± 0.1	2.8 ± 0.3



(a)

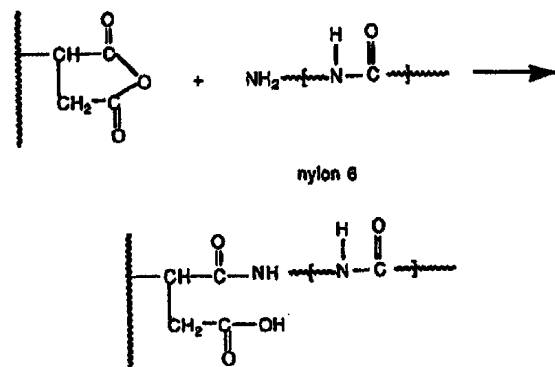


(b)

Figure 11 SEM micrographs of uncompatibilized Nylon 6/ABS/25 wt% LCP specimen: (a) core and (b) skin sections.

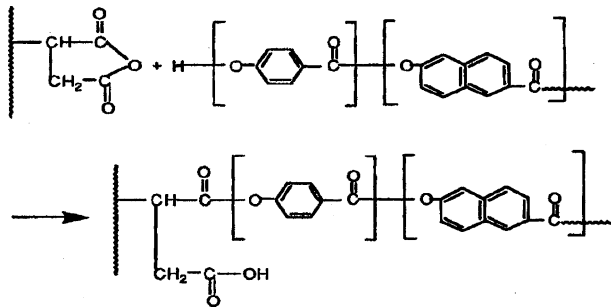
Fig 11a and b show the SEM micrographs of the core and skin sections of the uncompatibilized Nylon 6/ABS/25 wt% LCP specimen, respectively. From Fig. 11a, it is evident that the size of dispersed LCP domains in the core section is much larger than that of the Nylon 6/ABS(M)/25 wt% LCP blend (Fig. 4a). Comparing Fig. 11b with Fig. 3c, it is obvious that the LCP fibrils of the skin section for the Nylon 6/ABS(M)/25 wt% LCP blend are finer and longer than those of the uncompatibilized Nylon 6/ABS/25 wt% LCP specimen. The formation of LCP fibrils in the skin section of the Nylon 6/ABS(M)/25 wt% LCP blend is derived from the compatibilizing effects of maleic anhydride functional group and solid epoxy resin. The maleic anhydride grafted onto ABS during compounding results in the formation of MA-g-ABS that can react with polymers containing reactive end groups [34]. The MA functional group can react either with the amide group of Nylon 6 [35], or with the hydroxyl group of Vectra A-950 [36] during processing, thereby forming graft polymers at the interfaces.

Such graft polymers act as effective compatibilizers for the Nylon 6/ABS(M)/LCP blends. The reaction between MA-g-ABS with amide group of Nylon 6 is given as:



The reaction takes place between MA functional group and the hydroxyl group of Vectra A-950 during

compounding via,



Furthermore, the -NH_2 group of Nylon 6 also tends to react with the solid epoxy resin in the molten state, forming the block copolymer accordingly [37]. The aforementioned block copolymers improve the interfacial bonding of ternary phases of the Nylon 6/ABS(M)/LCP blends effectively.

3.5. Thermal properties

Fig. 12 shows the longitudinal storage moduli versus temperature for Nylon 6/ABS(M)/LCP blends. It can be seen that the storage modulus increases markedly with increasing LCP content from 30–150°C. Fig. 13

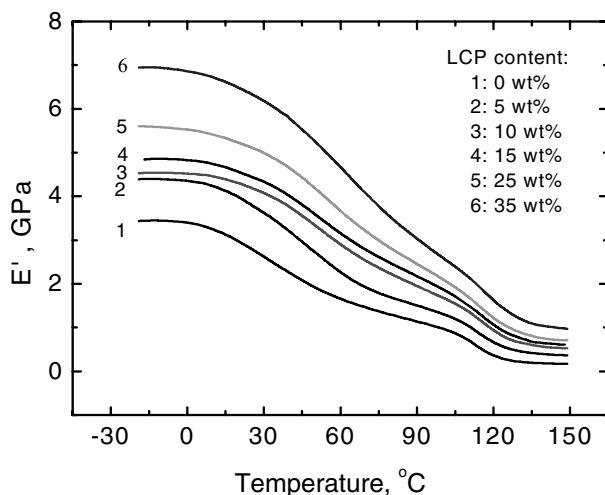


Figure 12 Storage modulus spectra of longitudinal Nylon 6/ABS(M)/LCP specimens.

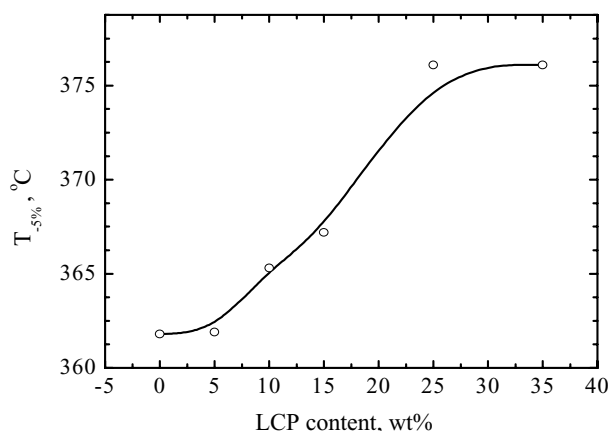


Figure 13 Variation of the 5% weight loss temperatures ($T_{-5\%}$) with LCP content for Nylon 6/ABS(M)/LCP blends from TGA measurements.

shows the variation of the 5% weight loss temperatures ($T_{-5\%}$) obtained from TGA with LCP content for Nylon 6/ABS(M)/LCP blends. The results indicate that the thermal stability of Nylon 6/ABS blend can be improved gradually with increasing LCP content.

4. Conclusions

Ternary Nylon 6/ABS(M)/LCP composites were prepared by means of injection molding. The effects of maleic anhydride functional group and epoxy resin on the structure-mechanical property relationship of such *in situ* composites were studied. Epoxy resin and MAP compatibilizers were found to improve the mechanical properties of Nylon 6/ABS(M)/LCP composites effectively. Therefore, extended long and fine LCP fibrils were formed in the skin section of *in-situ* composites as evidenced from SEM observations. Tensile tests showed that the ternary composites exhibit anisotropic mechanical properties due to the formation of elongated LCP fibrils. The longitudinal tensile strength and stiffness of ternary composites were considerably higher than those of transverse specimens. Moreover, the longitudinal tensile strength and stiffness of ternary composites tended to increase with increasing LCP content. Impact tests revealed that the transverse impact strength of these composites decreases with the addition of only 5 wt% LCP, thereafter it remains unchanged with increasing LCP content. The longitudinal impact strength also exhibited a similar decreasing trend behavior, but it showed an increase when the LCP content reached 35 wt%. DMA measurements showed that the storage modulus increases markedly with increasing LCP content from 30–150°C. Finally, TGA tests indicated that the 5% weight loss temperatures ($T_{-5\%}$) of the Nylon 6/ABS(M)/LCP composites tend to increase with increasing LCP content.

References

1. S. C. TJONG, *Mater. Sci. Eng. R: Reports* **41** (2003) 1.
2. R. A. KUDVA, H. KESKKULA and D. R. PAUL, *Polymer* **41** (2000) 239.
3. *Idem.*, *ibid.* **41** (2000) 335.
4. *Idem.*, *ibid.* **39** (1998) 2447.
5. V. J. TRIACCA, S. ZIAEE, W. BARLOW, H. KESKKULA and D. R. PAUL, *ibid.* **32** (1991) 1401.
6. Y. AOKI and M. WATANABE, *Polym. Engng. Sci.* **32** (1992) 878.
7. A. MISRA, G. SAWHNEY and R. A. KUMAR, *J. Appl. Polym. Sci.* **50** (1993) 1179.
8. B. MAJUMDAR, H. KESKKULA and D. R. PAUL, *Polymer* **35** (1994) 3164.
9. *Idem.*, *ibid.* **35** (1994) 5453.
10. T. LIMTASIRI and A. L. ISAYEV, *J. Appl. Polym. Sci.* **42** (1991) 2923.
11. R. M. HOLSTI-MIETTINEN, M. T. HEINO and J. V. SEPPALA, *ibid.* **57** (1995) 573.
12. F. P. LA MANTIA, R. SCAFFARO, P. L. MAGAGNINI, M. PACI, C. CHIEZZI, L. SEK, L. I. MINKOVA and T. MITEVA, *Polym. Eng. Sci.* **37** (1997) 1164.
13. S. C. TJONG and Y. Z. MENG, *Polymer* **38** (1997) 4609.
14. Y. Z. MENG and S. C. TJONG, *ibid.* **39** (1998) 99.
15. Y. Z. MENG, S. C. TJONG and A. S. HAY, *ibid.* **38** (1998) 1845.
16. S. C. WONG, Y. W. MAI and Y. LENG, *Polym. Eng. Sci.* **38** (1998) 156.

17. H. CHIH, K. CHIOU and F. CHANG, *J. Appl. Polym. Sci.* **60** (1996) 2503.
18. S. C. TJONG and W. JIANG, *ibid.* **74** (1999) 2274.
19. Y. SEO, B. KIM, S. KWAK, K. U. KIM and J. KIM, *Polymer* **40** (1999) 4441.
20. H. J. DONNELL and D. G. BAIRD, *ibid.* **36** (1995) 3113.
21. A. DATTA and D. G. BAIRD, *ibid.* **36** (1995) 505.
22. R. E. BRETAS and D. G. BAIRD, *ibid.* **33** (1992) 5233.
23. S. K. KWON and I. CHUNG, *Polym. Eng. Sci.* **35** (1995) 1137.
24. G. SAWHNEY, S. K. GUPTA and A. MISRA, *J. Appl. Polym. Sci.* **62** (1996) 1395.
25. S. JOSLIN, W. JACKSON and R. FARRIS, *ibid.* **54** (1994) 439.
26. L. CARPANETO, G. LESAGE, R. PISINO and V. TREFILETTI, *Polymer* **40** (1999) 1781.
27. R. K. LI, S. C. TJONG and X. L. XIE, *J. Polym. Sci. B, Phys. Ed.* **38** (2000) 403.
28. S. C. TJONG and Y. Z. MENG, *J. Appl. Polym. Sci.* **74** (1999) 1827.
29. M. L. SHIAO, S. V. NAIR, P. D. GARRETT and R. E. POLLARD, *Polymer* **35** (1994) 306.
30. J. C. HALPIN and J. L. KARDOS, *Polym. Eng. Sci.* **16** (1976) 344.
31. L. E. NIELSEN, "Mechanical Properties of Polymer and Composites" (Marcel Dekker, New York, 1974).
32. G. CREVECOEUR and G. GROENINCKX, *Polym. Eng. Sci.* **30** (1990) 532.
33. S. C. TJONG, S. L. LIU and R. K. LI, *J. Mater. Sci.* **32** (1997) 2163.
34. C. CARROT, M. JAZIRI, J. GUILLET and J. F. MAY, *Plas. Rubb. Process. Appl.* **14** (1990) 245.
35. A. CECERE, R. GRECO, G. RAGOSTA, G. SCARINZI and A. TAGLIALATELA, *Polymer* **31** (1990) 1239.
36. Y. SEO, S. S. HWANG, K. U. KIM, J. LEE and S. I. HONG, *ibid.* **34** (1993) 1667.
37. C. C. HUANG and F. C. CHANG, *ibid.* **38** (1997) 2135.

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