

Structures and Physical Properties of *in situ* composite based on Liquid Crystalline Polymer and Poly(ethylene 2,6-naphthalate) Blends

Hyun Oh YOO , Seong Hun KIM , Soon Man HONG* and
Seung Sang HWANG*

Dept. of Textile Engineering, Hanyang University

**Polymer Hybrid Center, Korea Institute of Science & Technology*

1. INTRODUCTION

The *in situ* composites based on poly(ethylene 2,6-naphthalate) and thermotropic liquid crystalline polymer (LCP) have been an area of increasing interest and study, since LCP exhibits high chemical stabilities and excellent thermo mechanical properties such as high strength and modulus. In a binary, however, poor compatibility and interfacial adhesions between two phases frequently results in deteriorated mechanical properties. In recent years, generating interfacial adhesion and compatibility of the *in situ* composite has been attempted by introducing new techniques such as long flexible spacer (1,2), block copolymer (3-5), functional groups having a chemical structure similar to the compounded polymers to modify the surface of binary blends (6-7), reactive extrusion (8,9), and addition of a third component having functional groups such as a maleic anhydride group (10), or incorporation of the LCP miscible with the matrix polymer (11). Of these, reactive extrusion provides the most attractive approach to the development of an *in situ* composite system. In this respect, Hong et al. (7-9) and Baird et al. (10) have investigated the effect of the third component or a compatibilizer on the physical properties of the immiscible *in situ* composite system. They observed that the key factors to property improvement are good interfacial adhesion, less phase separation, and more uniform LCP distribution. In particular, our recent interest is to expand the application area of the *in situ* composite featuring better physical properties, which can be obtained through transesterification reaction between ester groups (12) and other reactions (8,9). It has also been shown that a catalyst, such as dibutyltin dilaurate (DBTDL), can be used to induce compatibility of PEN/TLCP (13).

Many authors (12,14) report that transesterification reactions occur in blends of PET or PBT and polycarbonate at high temperatures with very long residence time.

The purpose of this work is to investigate the interrelationships among the processing structures and physical properties of PEN/LCP blends by adding a reaction catalyst and to promote the transesterification reaction in a very short residence time and to enhance the physical properties of the immiscible PEN/LCP blends

2. EXPERIMENTAL

As a matrix, poly(ethylene 2,6-naphthalate) Mitsubishi Co was used. The reinforcement was thermotropic liquid crystalline polymer LC5000 of Unitica Co, which was composed of 80 mole % of p-hydroxybenzoic acid (PHB) and 20 mole % of poly(ethylene terephthalate) (PET). The reactive catalyst dibutylitindilaurate (DBTDL) was obtained from Aldrich Co. PEN/LCP blends fiber were melt spun using a Rosand Capillary Rheometer. Measurement of tensile, thermal and morphological properties were performed by DMA, TMA and SEM, respectively.

3. RESULTS AND DISCUSSION

The *in situ* composite based on PEN/LCP blend in a twin screw extruder having a very short residence time was investigated in terms of thermal, rheological and physical properties. The dynamic mechanical thermal analysis was used to investigate different transitions occurring in PEN/LCP blends and there results are shown in Figure 1.

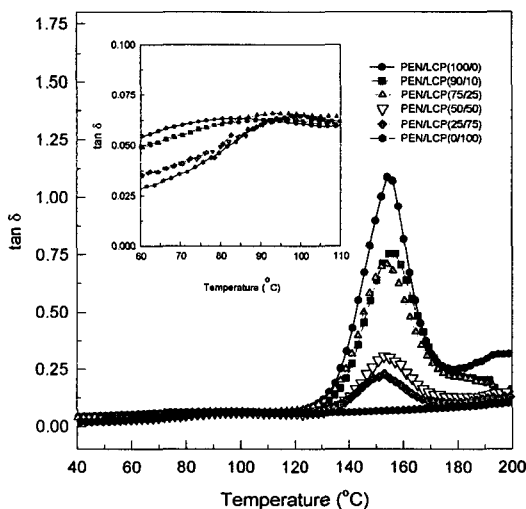


Fig. 1. Glass transition temperature of PEN/LCP blends as a function of LCP content.

Tan δ vs. temperature curve exhibits two distinct peaks, which is characteristic of an immiscible two-phase system. The high T_g (at around 154°C) comes from the PEN rich phase while the low one (at around 99°C) is due to the LCP (PHB80/PET20) rich phase respectively. This peak is related to the motion of PET segments in the PHB - rich phase, as it was also reported by Benson and Lewis (25). The height of the tan δ peak decreased with increasing LCP content. Inclusion of a small amount of LCP enhanced the crystallization rate of PEN matrix. They acted as a nucleating agent. LCP lowered the blend viscosity above T_{kn} (crystalline-nematic transition temperature), working as a processing aid. However, the addition of DBTDL as a reaction catalyst was found to increase the viscosity of the blends and enhance its adhesion with the matrix and lead to increase of mechanical properties of two immiscible phases. Hence DBTDL is helpful to produce a reactive compatibility at the interface of this polymer blend system. The optimum catalyst amount turned out to be about 500ppm when the reaction proceeds in 75/25 PEN/LCP blend system. Its effect on the mechanical properties show in Figures 2 and 3.

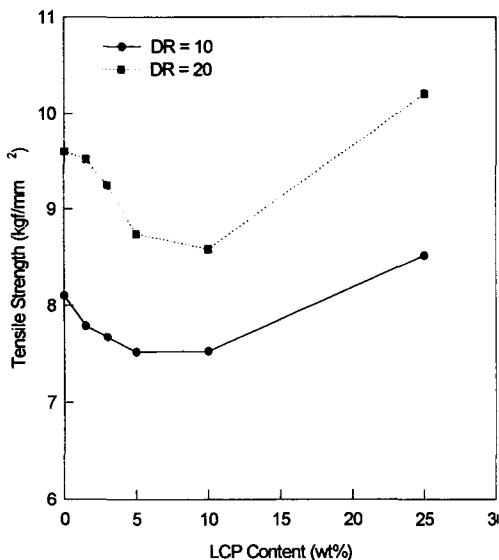


Fig. 2. Tensile strength of PEN/LCP blends as a function of LCP content at the draw ratio 10 and 20.

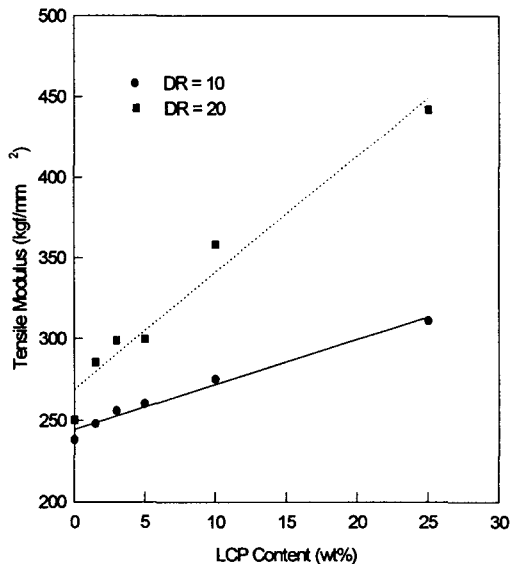


Fig. 3. Tensile modulus of PEN/LCP blends as a function of LCP content at the draw ratio 10 and 20.

4. CONCLUSIONS

Improving blend compatibility should increase the interfacial adhesion between the different components and the mechanical performance. Therefore it is important to

control the interrelationships among the processing, structure, and physical properties for the immiscible blend and to improve the interfacial adhesion and compatibility by reactive extrusion. Blends made by reactive extrusion of PEN, LCP, and DBTDL used as a transesterification reaction catalyst have advantage over general binary PEN/LCP blends in the fabrication of in situ composites in terms of desired properties. The modulus of the blends follows an additive rule with increasing the content of LCP in the PEN/LCP blends. The strength, however, shows an abrupt drop-off. Addition of DBTDL used as a reaction catalyst was found to slightly increase the viscosity of the blends, enhance its adhesion with the matrix and lead to increase of physical properties of two immiscible phases in a short residence time. DBTDL is helpful to produce a reactive compatibilizer by reactive extrusion at the interface of this polyester blend systems. Thus, to improve the physical properties between the matrix and dispersed phase, both interfacial adhesion between two polymers and the morphology of blends must be controlled.

REFERENCES

1. B. Y. Shin and I. J. Chung, *Polym. J.*, **30**, 13 (1990).
2. B. Y. Shin and I. J. Chung, *Polym. J.*, **30**, 22 (1990).
3. S. Joslin, W. Jackson, and R. Farris, *J. Appl. Polym. Sci.*, **54**, 289 (1994).
4. S. Joslin, W. Jackson, and R. Farris, *J. Appl. Polym. Sci.*, **54**, 439 (1994).
5. S. Joslin, W. Jackson, and R. Farris, *J. Appl. Polym. Sci.*, **35**, 574 (1996).
6. S. H. Kim and S. W. Kang, *J. Appl. Polym. Sci.*, Accepted **May** 14 (1998).
7. S. M. Hong and B. C. Kim, *Polym. Eng. Sci.*, **34**, 1605 (1994).
8. Y. Seo, S. M. Hong, S. S. Hwang, T. S. Park, K. U. Kim, S. Lee, and J. Lee, *Polymer*, **36**, 515 (1995).
9. Y. Seo, S. M. Hong, S. S. Hwang, T. S. Park, K. U. Kim, S. Lee, and J. Lee, *Polymer*, **36**, 525 (1995).
10. A. Datta, H. H. Chen, and D. G. Baird, *Polymer*, **34**, 759 (1993).
11. C. Ryu, Y. Seo, S. S. Hwang, S. M. Hong, T. S. Park, K. U. Kim, S. Lee, and J. Lee, and J. Lee, *Intern. Polym. Proc.*, **9**, 266 (1994).
12. Su. K. F. and Wei, K. H. *J. Appl. Polym. Sci.* **56**, 79, (1995).
13. M. J. Stachowski and A. T. DiBenedetto, *Polym. Eng. Sci.*, **37**, 262 (1997).
14. R. S. Porter and L.H. Wang, *Polymer*, **33**, 2019 (1992).
15. R. S. Benson. and D. N. Lewis., *Polym. Commun.*, **28**, 280 (1987).