

Physical and Mechanical Properties of Thermotropic Liquid Crystalline Polymer and PBT Blends

Seong Hun Kim, Jung Gyu Lee and Han Sang Kim
*Department of Textile Engineering,
College of Engineering, Hanyang University,
Seoul, 133-791, KOREA.*

INTRODUCTION

The increasing demands for high modulus and strength polymeric materials have drawn considerable interest in industry. Thermotropic liquid crystalline polymers (TLCPs), differing from lyotropic liquid crystalline polymers (LLCPs), have excellent melt processability and mechanical property resulting from a high degree of molecular orientation under a shear flow field in the molten state with relatively low viscosity.^{1,2} This attractive mechanical properties with desirable optical and physical characteristics have made LCPs one of the promising materials for various applications in the electric, chemical and aerospace industry. From the advantages of LCP, the blends of rigid chain polymers forming TLC states with conventional thermoplastic partially flexible-chain polymers such as poly(butylene terephthalate) (PBT) could lead to the improvement of mechanical properties. In general, increment of the physical properties is dependent on the influence of TLCPs upon the crystallization of thermoplastic polymer (or matrix polymer) through a thermal process.³ In this research, the feasible chemical interaction^{4,5} between LCP and matrix polymer, and the effects of TLCPs on the crystallization and thermal behavior of matrix polymer were studied by thermal analysis and morphological observations. Relatively little activity has been directed to melt flow and processing characteristics. Viscosity-shear rate data have been reported for PHB60/PET40 and this type of system appears to exhibit unbounded viscosity increase at low shear rate. The flexural modulus and strength of injection molded aromatic polyester parts are found to be considerably greater in the machine than the transverse direction.

EXPERIMENTAL

The TLCP used in this research is a commercial LCP copolyester, RODRUN 3000 (Unitika Co., Japan) supposed to be prepared from 60 mol% of p-hydroxy-benzoic acid (HBA) and 40 mol% of poly(ethylene terephthalate) (PET). Poly(butylene terephthalate) used as thermoplastic matrix polymer was supplied from Samyang Co. All samples in form of pellets were dried *in vacuo* at 110°C for 24 hours before processing. Melt blending of HBA60/PET40 and PBT was performed in a Minimax Molder model CS-183 MMX (CSI Co.) at 240°C or 260°C with different compositions, and differential scanning calorimetric (DSC) measurements were carried out in a Perkin-

Elmer DSC7. Samples approximately 10mg were heated and cooled under nitrogen atmosphere at a rate of 10°C/min in the range from 30 to 280°C. After the first scanning, the second scan was conducted with the same conditions and adopted as experimental data. In addition, for the study of thermal decomposition and thermal stability, the thermogravimetric analysis (TGA) was carried out. The copolymer was rheologically characterized in capillary and torque rheometers between 190 and 300°C with various shear rates. Filaments were extruded and melt spun from a HAAKE torque rheometer with a die of diameter 0.1cm and L/D ratio of 10. The mechanical properties of the copolymer were investigated in Instron 4202 (Universal Testing machines) and Izod Impact test was also performed. The morphology and crystallization behavior of blends were investigated on Nikon HFX-11A Polarized Optical Microscope with hot stage during the sample was cooled from heated state at 270°C.

RESULT and DISCUSSION

Thermal gravimetric analysis shows that the thermal property of blend containing 5% LCP is more stable than that of 3,7 or 10% LCP blends, and the data obtained from DSC thermograms indicated that the 3 or 5% addition of LCP to PBT matrix processed at 260°C causes the ascent of melting temperature due to the increase of crystallinity or transition of the crystal structure, as shown in Figures 1 and 2.

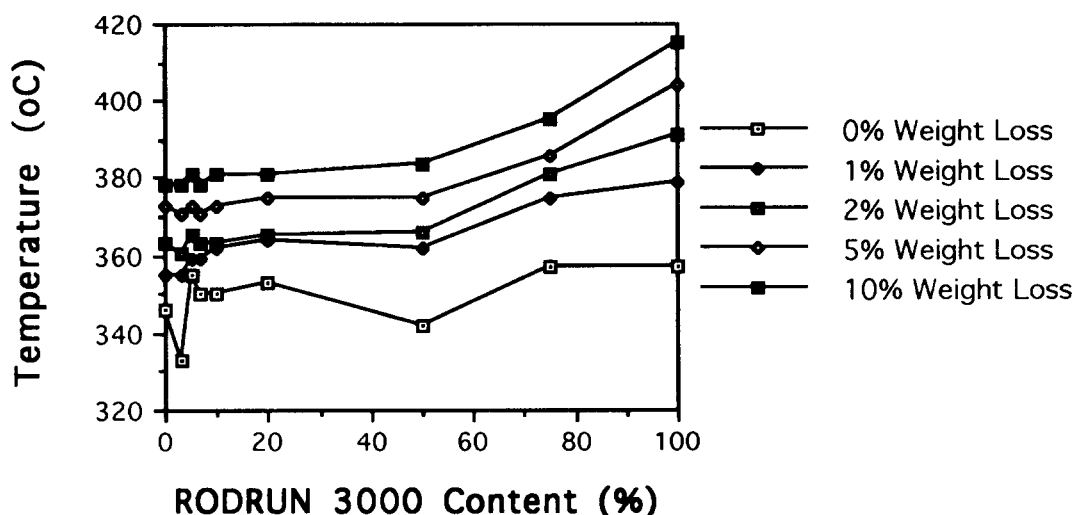


Fig 1. Degradation temperature (Td) of HBA60/PET40 and PBT blends at each weight loss with LCP content.

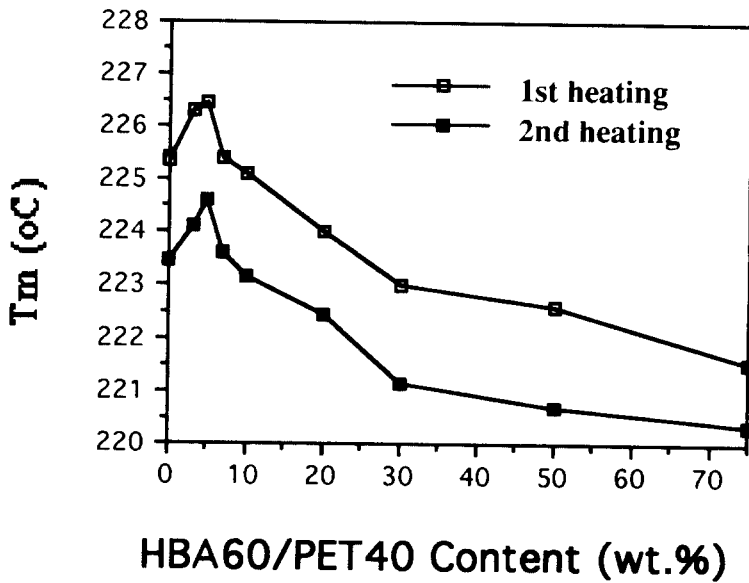


Fig 2. Melting temperature (T_m) changes of HBA60/PET40 and PBT blends compounded at 260°C for 8 min.

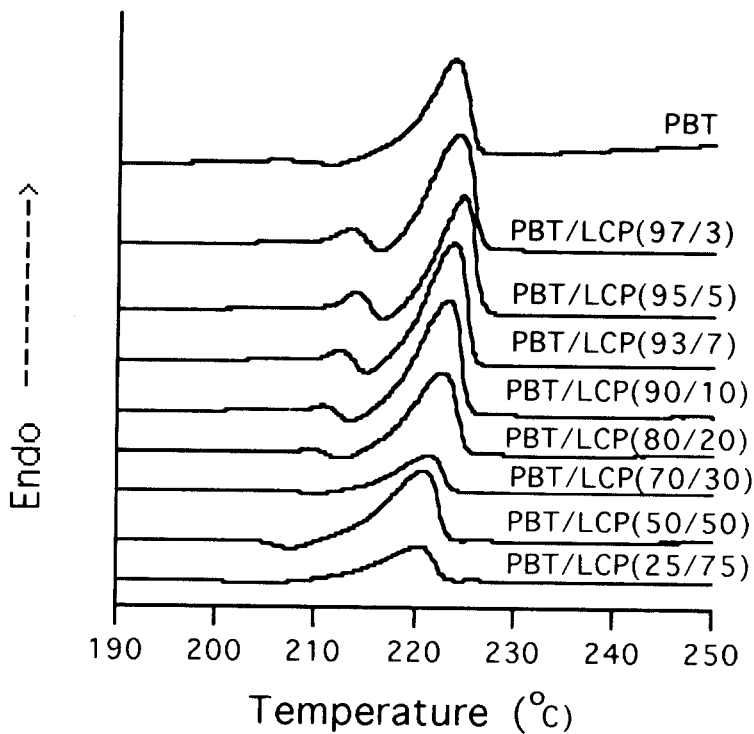


Fig 3. DSC thermograms of HBA60/PET40 and PBT blends in 2nd heat scan.

The pre-melting endothermic peaks of blends containing 3-20 wt.% LCP can be observed in Fig 3. One of the possible explanations for the phenomena is a certain crystallization induced by intermolecular reaction such as transesterification reaction or the melting of second crystallized parts occurred during heat scan. The crystallization temperature and the heat of crystallization tend to increase with LCP content from 3 up to 7 wt.%, followed by decrease thereafter. In the cooling thermogram of 75% LCP blend, a new type of exothermic transition peak appeared. The viscosity decrease with increasing with LCP content as shown in Fig 4. However, the highest value was shown at 5-7wt.% LCP. The data were unbounded at low shear rate and then "group together" in the high shear rate.

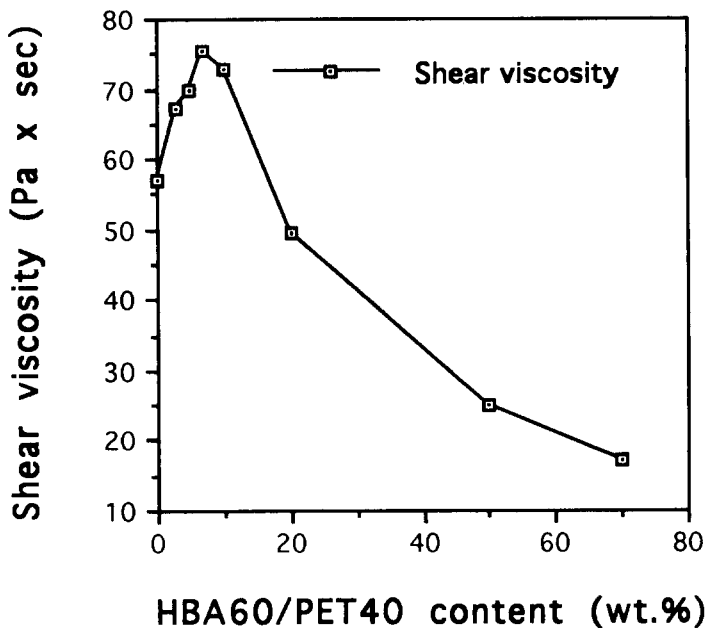


Fig 4. Shear viscosity changes of HBA60/PET40 and PBT blends at shear rate 7000 (1/sec).

The tensile strength at 5-7wt.% of LCP adding to the PBT is found to be increased approximately 25% over that of pure PBT. Beyond 10% LCP content, the ultimate tensile strength of the composite becomes dependent upon the ultimate tensile strength of the reinforcing species. Pure PBT and PBT/LCP blends containing 5% and 7% LCP exhibit highly ductile behavior while the blends with LCP content above 10% show brittle fracture. Polarized optical micrographs reveal that 3-7% addition of LCP result in a considerably well dispersed phase of LCP in matrix polymer. In 7% LCP/PBT blend, comparatively smaller and a little more crystalline domain than that of pure PBT implies

the role of LCP as nucleating agent in PBT matrix. Thus these conclusions are consistent with the results of other thermal analysis.

CONCLUSIONS

The thermal, mechanical and rheological properties, microstructural view and crystallization behavior of polymeric blends consisting of HBA60/PET40 and PBT have been studied as a function of LCP content. In tensile testing, initial modulus of blends increased with LCP content, and a synergistic effect of tensile strength in 5 or 7% LCP/PBT blends was observed approximately 26% higher than in 3 or 10% LCP/PBT blends. 5% addition of LCP to PBT matrix made the blend more stable in thermal properties than 3, 7 or 10% LCP/PBT blends. According to the differential scanning calorimetric measurement, melting temperature depression was occurred in the blends processed at 240°C, seriously with increasing LCP component, while the blends compounded at 260°C showed the ascent of melting temperature especially in the case of 3 or 5% addition of LCP. Crystallization temperature and heat of crystallization were found to increase gradually in the range from 3 up to 7% LCP/PBT and begin to decrease on further addition of LCP to PBT matrix. In the cooling thermogram of 75% LCP blend melt blended at 260°C, a new type of exothermic transition peak around 160°C seems to be overlapped with the crystallization peak of PBT appearing at 177°C. From the result of polarized optical microscopic observation, it is suggested that excessive addition of LCP to PBT matrix can generate a considerable phase separation at the interface of two materials, and comparatively excellent compatibility between dispersed LCP and PBT matrix can be achieved in 3-7% LCP/PBT blends. At high shear rate, LCP domains are oriented and aligned themselves in the flow direction. They remain in such deformed state because of high relaxation time of LCP. Inclusion of LCP results in the ease of processing and tremendous improvement in mechanical properties. Extrusion and injection molding conditions have been identified at which LCP forms *in situ* fiber reinforced composites in PBT matrix. However, the degree of anisotropy increase with an increase in LCP concentration. Fibrous LCP domains were created during processing and are present in the solidified material. This results in the formation of self-reinforced composites.

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