# Thermal property and miscibility of polycarbonate/copolyester blends

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## SUMMARY

The thermal property and the miscibility of polycarbonate(PC)/copolyester blends were investigated. For the study, different copolyesters were synthesized from terephthalic acid (TPA) and various mixtures of ethylene glycol (EG) and cyclohexane dimethanol(CHDM). Various blends of PC and copolyester were prepared by melt mixing and thermal properties of the blends were studied employing differential scanning calorimeter. It was found that the blends of the PC and the copolyesters were partially miscible when the glycol in the copolyester was composed of 10, 20, or 30 mole % CHDM. However, the blends of the PC and the copolyesters were miscible in all proportions when the glycol in the copolyester was composed of 50 or 70 mole % CHDM. Miscibilities of the PC/copolyester blends depending on the composition of the copolyester are discussed based on the thermal properties of the blends.

### INTRODUCTION

blends of bisphenol-A polycarbonate(PC) and various thermoplastic Recently. polyesters have been studied by many researchers(1-5). Poly(ethylene terephthalate) (PET) and poly(butylene terephthalate) (PBT) were reported to be partially miscible with PC(1,2). But, copolyesters formed from 1,4-cyclohexane dimethanol(CHDM) and terephthalic acid(TPA) or mixture of TPA and isophthalic acid were found miscible with PC in all proportions(3). Aliphatic polyesters were also reported miscible with PC in all proportions(4). According to Paul and his coworkers, ester exchange reactions between PC and copolyesters during melt mixing are not so important and miscible phase formations are due to physical interactions between the blend components(5). It is expected that the miscibility of a copolyester and PC is largely dependent on the CHDM content of the copolyester. However, there are few papers concerning the miscibility of PC and various copolyesters from TPA, CHDM, and ethylene glycol(EG), depending on the composition of the copolyester. Thus, we synthesized copolyesters of different compositions and the blends of PC and the copolyesters were investigated. The miscibilities of PC/copolyester blends are discussed in this paper based on the thermal properties of the blends

The PC used was a commercial bisphenol A polycarbonate from Samyang Company designated TRIREX-3022 which has Mn=14,200 and Mw=42,500. Following the method described by Goodman, various copolyesters were synthesized by polycondensations of the oligomers obtained by transesterification of dimethyl terephthalate(DMT) and CHDM/EG(6). The characteristics of the copolyesters are given in Table-1. Blending of the copolyesters and the PC were carried out employing Roller Mixer of Brabender Plasticorder 331 for 3minutes at 250-270. °C. There might be ester exchange reactions between the copolyesters and the PC during melt mixing. But, it was assumed that such reactions did not affect the miscibility of the blend significantly as pointed out by Paul and his coworkers(5). The blend ratios of PC/copolyester systems were 75/25, 50/50, and 25/75 by weight. Thermal properties of the blends were studied employing differential scanning calorimeter (DSC: DSC 951 of DuPont TA-2000). The samples were heated in DSC up to 270 °C, held for 3 minutes and quenched to room temperature using liquid nitrogen. Then, the samples were scanned at 10 ° C/min to check the glass transition temperatures (T<sub>g</sub>'s), crystallizations, and melting behaviors of the blends.

#### RESULTS AND DISCUSSION

In Fig. 1, DSC thermograms of the PC/copolyester(C-10) blends are shown. Two T<sub>g</sub>'s were observed in the PC/C-10 blends. Miscible polymer blends exhibit a single Tg between the Tg 's of the pure components while, in partially miscible systems, two  $T_{e}$ 's approach each other but do not become identical(7). The two T<sub>e</sub>'s in Fig.l are due to PC-rich phase (higher T<sub>e</sub> ) and copolyester-rich phase(lower T  $_{e}$  ). It is worthwhile to note that the higher T  $_{e}$ 's are lower than the T<sub>e</sub> of PC while the lower T<sub>e</sub>'s are almost the same as the T<sub>e</sub> of C-10. It is speculated that the copolyester dissolves more in the PC-rich phase than does the PC in the copolyeste-rich phase. Li and Williams also reported two T g 's were observed in blends of PC and a copolyester the composition of which was not revealed(8). The PC/C-10(25/75) blend and the PC/C-10(50/50)blend showed crystallization exotherms and crystalline melting endotherms while the PC/C-10(75/25) blend did not. The crystallization of the copolyester seems to be depressed by the PC in the blends.

Sample Code	Molar Ratio of EG/CHDM	Mn	Mw
C-10	90/10	45, 200	77, 200
C-20	80/20	49, 600	66, 200
C-30	70/30	34, 700	69,000
C-50	50/50	39, 400	67, 100
C-70	30/70	31, 900	65, 200

Table-1. Characteristics of Copolyesters Synthesized.

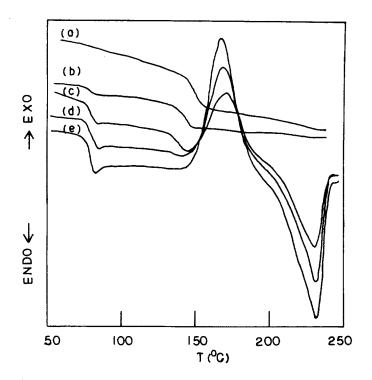


Fig.1 DSC thermograms of the PC/C-10 blends in different compositions(by wt.): (a)100/0; (b)75/25; (c)50/50; (d)25/75; (e)0/100.

In Fig.2, DSC thermograms of the PC/copolyester (C-20) blends are shown. The PC/ C-20(25/75) blend showed single T  $_{\rm g}$  and is believed to form miscible blend. However the PC/C-20(50/50) blend and the PC/C-20(75/25) blend showed two T  $_{\rm g}$  's which are due to the PC-rich phase and the copolyester-rich phase as the PC/C-10 blends. It was also found that the T  $_{\rm g}$  's of PC-rich phases were depressed more than those of the PC-rich phases in the PC/C-10 blends even though the T  $_{\rm g}$  of the C-20 was almost the same as that of the C-10. It is believed that the miscibility of the PC and the copolyester was improved as the CHDM content of the glycols in the copolyester is increased from 10 to 20 mole %. It is of interest to note that the copolyester, C-20, did not show crystallization exotherm in the DSC thermogram. The crystallization of copolyester seems to be depressed as the CHDM content in the glycols of the copolyester is increased from 10 to 20 mole %.

In Fig.3, DSC thermograms of the PC/copolyester (C-30) blends are shown. Thermal behaviors of the blends were similar with those of the PC/C-20 blends. But, it was observed that the  $T_{g}$ 's of PC-rich phases were lowered and  $T_{g}$ 's of copolyester-rich phases were increased compared with those of the PC/C-20 blends. Thus, it is believed that the miscibility of the PC and the copolyester is also improved as the CHDM content in the glycols of the copolyester is increased from 20 to 30 mole %. However, the PC/C-30 blends are still partially

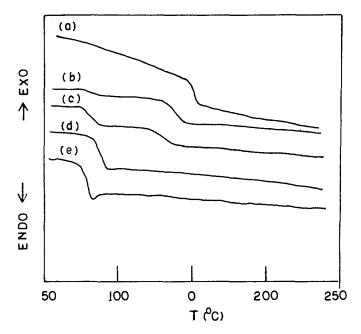


Fig.2 DSC thermograms of the PC/C-20 blends in different compositions(by wt.): (a) 100/0; (b) 75/25; (c) 50/50; (d) 25/75; (e) 0/100.

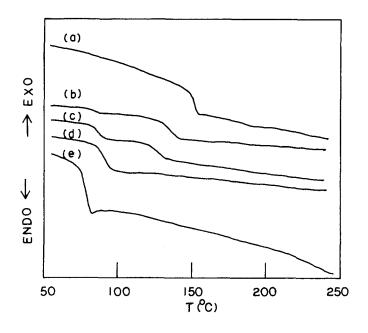


Fig.3 DSC thermograms of the PC/C-30 blends in different compositions(by wt.): (a)100/0; (b)75/25; (c)50/50; (d)25/75; (e)0/100.

miscible. It was also noted that the copolyester, C-30, did not show crystallization exotherm as the copolyester, C-20.

In Fig. 4, DSC thermograms of the PC/copolyester (C-50) blends are shown. Single T  $_{e}$  was observed between T $_{e}$ 's of the PC and the copolyester, C-50. It is believed that the PC/C-50 blends are miscible in all proportions. The copolyester C-50 did not show cystallization exotherm in the DSC thermogram as the copolyester, C-20 or C-30. In Fig.5, DSC thermograms of the PC/C-70 blends are also shown. The PC/C-70 blends also showed single T $_{e}$ 's. The PC/C-70 blends are also believed to be miscible in all proportions. It is speculated that the blends of the PC and the copolyester become miscible as the CHDM content in the glycols of the copolyester is increased from 30 to 50 mole % or more than 50 mole %. It is of interest to note that crystallization exotherm and crystallization melting endotherm were observed in C-70. But the blends did not undergo crystallization except the PC/C-70(25/75) blend.

#### CONCLUSION

Various copolyesters were synthesized and miscibilities of the PC/copolyester blends were investigated based on the thermal properties of the blends. It was found that the blends of the PC and the copolyesters (C-10, C-20, and C-30) are

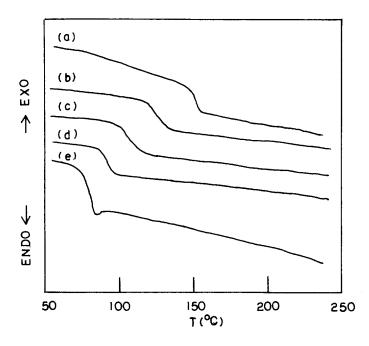


Fig.4 DSC thermograms of the PC/C-50 blends in different compositions(by wt.): (a)100/0; (b)75/25; (c)50/50; (d)25/75; (e)0/100.

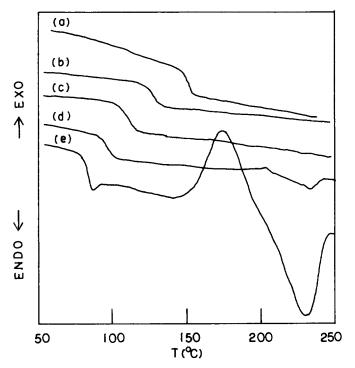


Fig.5 DSC thermograms of the PC/C-70 blends in different compositions(by wt.): (a)100/0;(b)75/25;(c)50/50;(d)25/75;(e)0/100.

partially miscible while the blends of the PC and the copolyesters (C-50 and C-70) are miscible in all proportions. It is concluded that the miscibility of the copolyester with the PC is improved as the CHDM content of the copolyester is increased.

#### REFERENCES

- 1.Xin Yue Chen and Arthur W.Birley(1985) British Polymer Journal 17:347
- 2. S. Y. Hobbs, V.L. Groshans, M.E. J. Dekkers, and A.R. Shultz(1987) Polymer Bulletin 17:335
- 3.R.N.Mohn, D.R.Paul, J.W.Barlow, and C.A.Cruz(1979) J Appl Polym Sci 23:575
- 4.C.A.Cruz, J.W.Barlow, and D.R.Paul(1979) Macromolecules 12:726
- 5.W.A.Smith, J.W.Barlow, and D.R.Paul(1981) J Appl Polym Sci 26:4233
- 6. I. Goodman(1988) Polyesters. In: J. I. Kroschwitz(ed.) Encyclopedia of Polymer Science and Engineering. John Wiley and Sons, New York(vol.12, pp1-75)
- 7.0.Olabisi, L.M.Robeson, and M.T.Shaw(1979) Polymer-Polymer Miscibility, Academic Press, New York
- 8.Yuan Li and H.Leverne Williams(1990) J Appl Polym Sci 40:1891

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