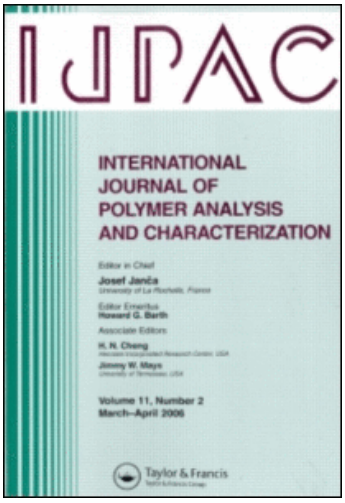

International Journal of Polymer Analysis and Characterization
Publication details, including instructions for authors and subscription information:
http://www.informaworld.com/smpp/title~content=t713646643

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To cite this Article Rajulu, A. Varada and Reddy, R. Lakshminarayana(2000) 'Miscibility Studies of Polycarbonate/ Poly(methyl methacrylate) and Polycarbonate/Polystyrene Blends as Measured by Viscosity, Ultrasonic, and Refractive Index Methods', International Journal of Polymer Analysis and Characterization, 5: 4, 467 — 473
To link to this Article: DOI: 10.1080/10236660008034639
URL: http://dx.doi.org/10.1080/10236660008034639

Int. J. Polym. Anal. Charact.,
2000 Vol. 5, pp. 467–473
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Miscibility Studies of Polycarbonate/Poly(methyl methacrylate) and Polycarbonate/Polystyrene Blends as Measured by Viscosity, Ultrasonic, and Refractive Index Methods

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(Received 7 December 1998; Revised 16 February 1999)

The ultrasonic velocity, viscosity, refractive index and density of polycarbonate/poly(methyl methacrylate) (PC/PMMA) and polycarbonate/polystyrene (PC/PS) blends in solution have been measured at 30°C. Using the viscosity data, the interaction parameters have been computed which indicate that the PC/PMMA blend is miscible, whereas the PC/PS is immiscible. This is confirmed by ultrasonic and refractive index measurements.

Keywords: Polymer blends; Polycarbonate; Poly(methyl methacrylate); Polystyrene; Ultrasonic velocity; Viscosity; Density; Refractive index; Miscibility

INTRODUCTION

The blending of polymers is one of the simplest means to obtain a variety of physical and chemical properties from the constituent polymers.[1] The gain in newer properties depends on the degree of compatibility or miscibility of the polymer at a molecular level. There have been various techniques of studying miscibility of polymer blends.[2] Some of these techniques may be complicated, costly and time consuming.

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Hence it is desirable to identify simple, low cost and rapid techniques to study the miscibility of the polymer blends. Chee[3] and Sun *et al.*[4] have suggested viscometry for the study of polymer–polymer miscibility. Hourston and Hughes[5] and Kuleznev *et al.*[6] have suggested the use of sonic and viscosity measurements for miscibility determination. Singh and Singh[7,8] and Singh *et al.*[9] have pointed out that ultrasonic velocity and viscosity measurements can be used to determine polymer–polymer miscibility. In the present investigation, miscibility studies are reported for polycarbonate/poly(methyl methacrylate) (PC/PMMA) and polycarbonate/polystyrane (PC/PS) blends by viscosity, ultrasonic velocity, density and refractive index methods.

EXPERIMENTAL

The blends of PC/PMMA and PC/PS of different compositions were prepared by mixing solutions of the polymers in chloroform. PMMA (Gujarath State Fertilizer Corporation, Vadodara, India, $M_v = 98{,}000$), PS (Gujarath State Fertilizer Corporation, Vadodara, India, $M_v = 90{,}000$) and PC (Viral Rasayan, Mumbai, $M_v = 25{,}000$) were employed in the present study. The total weight of the two components in solution always was maintained at 1 g/dL. The ultrasonic velocities of the blend solutions were measured by an ultrasonic interferometric technique.[10] The temperature was maintained at 30°C by circulating water from a thermostat with a thermal stability of ±0.05°C through a double-wall jacket of the ultrasonic experimental cell. The densities of the solutions were measured at 30°C by specific gravity bottle. The refractive indices of blend solutions were measured with an Abbe refractometer, thermostated with a water circulation system at 30°C, as described elsewhere.[11] The relative viscosities of blend solutions were measured at 30°C using a Ubbelohde suspended-level viscometer.

RESULTS AND DISCUSSION

The measured values of ultrasonic velocity, density and refractive index of blend solutions have been presented in Table I. Figure 1 shows the Huggins plots for blends of PC/PMMA and PC/PS in which

TABLE I Ultrasonic velocity, density and refractive index of PC/PMMA and PC/PS blend solutions in chloroform at 30°C

PC in blend (%)	*Ultrasonic velocity* (m/s)	*Density* (g/cc)	*Refractive index*
(A) *PC/PMMA blend*			
0	967.7	1.453	1.440
20	968.0	1.453	1.440
40	967.4	1.453	1.440
50	967.4	1.453	1.440
60	967.4	1.453	1.440
80	967.6	1.453	1.440
100	967.4	1.453	1.440
(B) *PC/PS blend*			
0	974.4	1.461	1.442
20	973.8	1.470	1.443
40	977.8	1.462	1.446
50	973.8	1.452	1.450
60	968.0	1.450	1.442
80	978.6	1.458	1.444
100	967.4	1.453	1.440

the weight fraction of both the components was maintained at 0.5. Chee[3] has given the expression for the interaction parameter when the polymers are mixed in weight fractions w_1 and w_2 as

$$\Delta B = \frac{b - \bar{b}}{2w_1 w_2}, \tag{1}$$

where $\bar{b} = w_1 b_{11} + w_2 b_{22}$, b_{11} and b_{22} are the slopes of the viscosity curves for the components and b is related to Huggins coefficient k_H as

$$b = k_H [\eta]^2. \tag{2}$$

For ternary system it is also given by

$$b = w_1^2 b_{11} + w_2^2 b_{22} + 2w_1 w_2 b_{12}, \tag{3}$$

where b_{12} is slope for the blend solution.

Using these values, Chee[3] has defined a more effective parameter:

$$\mu = \frac{\Delta B}{\{[\eta]_2 - [\eta]_1\}^2}, \tag{4}$$

where $[\eta]_1$ and $[\eta]_2$ are the intrinsic viscosities for the pure component solutions.

The blend is miscible if $\mu \geq 0$ and immiscible when $\mu < 0$.[3] In the present study for PC/PMMA, μ is 140 indicating that the

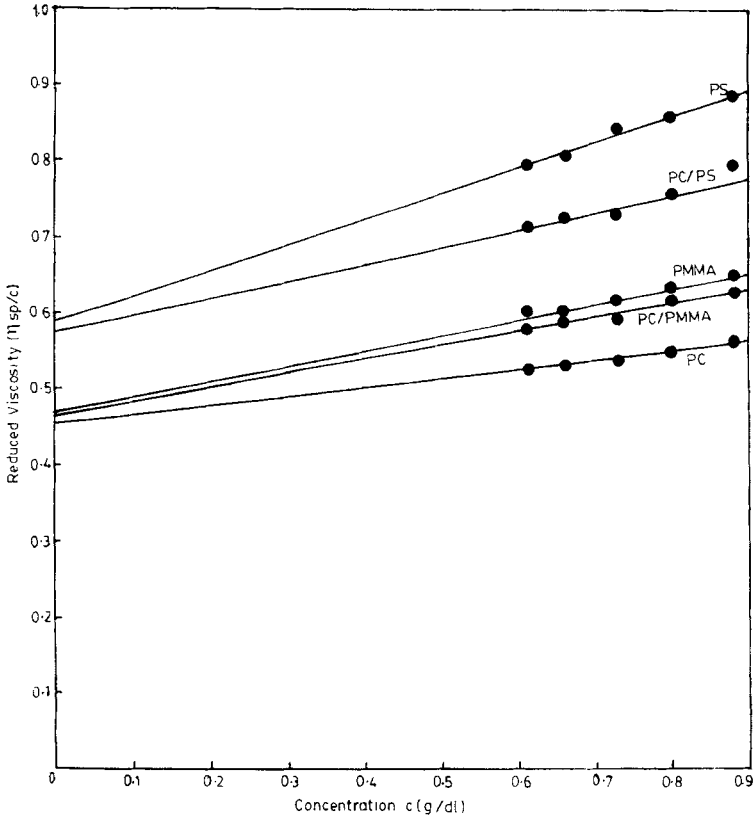

FIGURE 1 The variation of reduced viscosity with composition in PC/PMMA and PC/PS (equal weight fraction) blends in chloroform at 30°C.

PC/PMMA blend is miscible. In the case of PC/PS system, however, μ is -0.54 indicating that the PC/PS blend is immiscible.

Recently Sun *et al.*[4] have suggested a new equation for the determination of miscibility of polymers:

$$\alpha = k_m - \frac{k_1[\eta]_1^2 w_1^2 + k_2[\eta]_2^2 w_2^2 + 2\sqrt{k_1 k_2}[\eta]_1[\eta]_2 w_1 w_2}{\{[\eta]_1 w_1 + [\eta]_2 w_2\}^2}, \qquad (5)$$

where k_1, k_2 and k_m are the Huggins constants for individual components 1, 2 and blend, respectively. While deriving this equation,

the long-range hydrodynamic interactions are taken into account. Sun *et al.*[4] have suggested that a blend will be miscible if $\alpha \geq 0$ and immiscible when $\alpha < 0$. In the present study for the PC/PMMA system, the α value was found to be 0.15 indicating that it is miscible, and for the PC/PS system, the α value was found to be -0.13 indicating that this blend is immiscible.

In order to evaluate simplified techniques to investigate the miscibility of polymer blends under study, the variation of ultrasonic velocity, density and refractive index of the polymer blend solutions with composition have been depicted in Figures 2–4 respectively. From these figures it is clearly evident that the variation is linear for the PC/PMMA system showing a single phase in the blend, however, for the PC/PS system the variation is nonlinear showing a double phase in the blend.

Varada Rajulu *et al.*[12] have used these techniques for the miscibility study of cellulose acetate/PMMA blend, where nonlinear variation of the ultrasonic velocity and refractive index with blend composition was attributed to the immiscible behavior of the blend. Similarly the

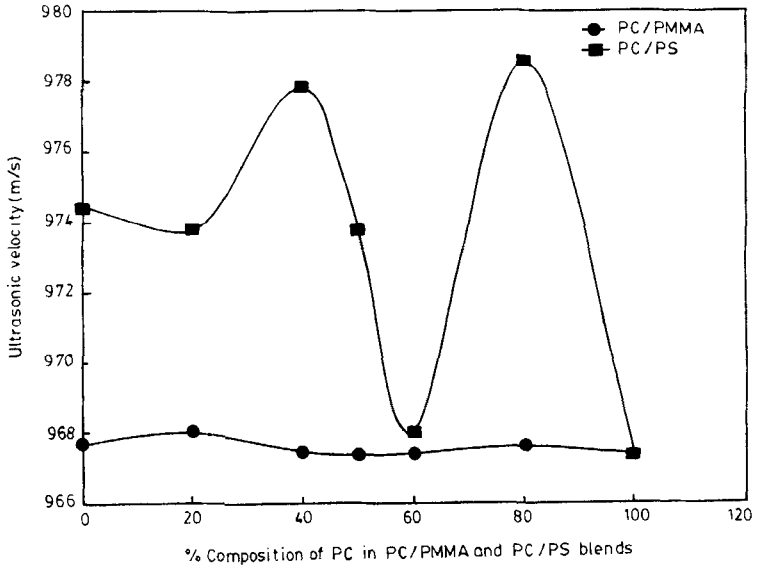

FIGURE 2 The variation of ultrasonic velocity with composition, with PC/PMMA and PC/PS blends in chloroform at 30°C.

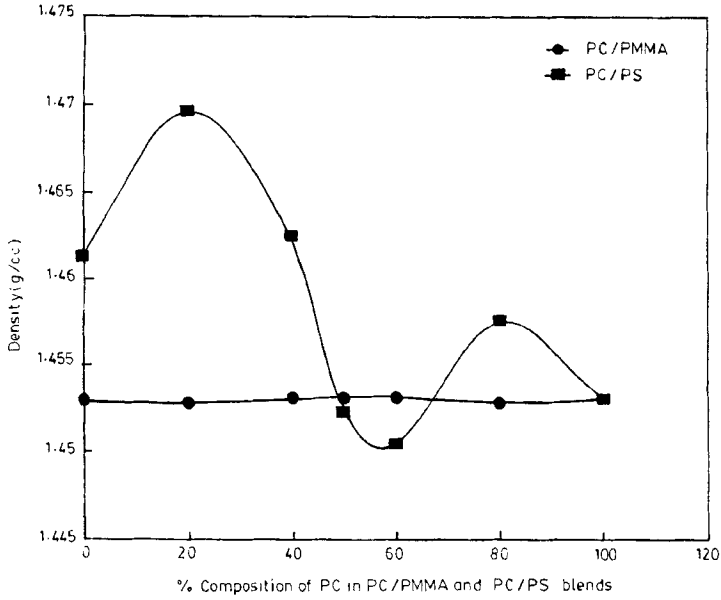

FIGURE 3 The variation of density with composition, with PC/PMMA and PC/PS blends in chloroform at 30°C.

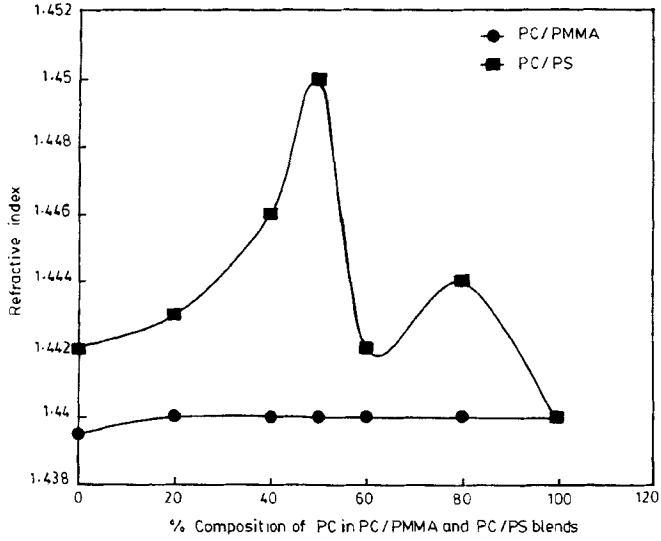

FIGURE 4 The variation of refractive index with composition, with PC/PMMA and PC/PS blends in chloroform at 30°C.

linear variation of ultrasonic velocity with blend composition in the case of PMMA/poly(vinly acetate)[7] has been attributed to the miscible nature of the blend. These observations support the validity of Equation (5) and confirm that the PC/PMMA blend is miscible and PC/PS is immiscible. Furthermore, it is also observed that the ultrasonic velocity, density and refractive index of the PC/PS blend vary more than those of the pure components. This observation further indicates that the blend is immiscible. The ultrasonic velocity, density and refractive index of PC/PMMA blend do not vary much from those of the pure components, indicating that the blend is miscible.[12]

Acknowledgments

One of the authors (R. Lakshminarayana Reddy) is thankful to Council of Scientific and Industrial Research (CSIR), India for the award of a Senior Research Fellowship.

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