Miscibility Studies of Epoxy/Unsaturated Polyester Resin Blend in Chloroform by Viscosity, Ultrasonic Velocity, and Refractive Index Methods

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ABSTRACT: Miscibility studies of blends of epoxy with unsaturated polyester resin in chloroform were carried out by viscosity, ultrasonic velocity, and refractive methods at 30°C. By using viscosity data, the interaction parameters were computed, which indicated that epoxy/unsaturated polyester resin blends were miscible. The miscibility was further confirmed by the ultrasonic velocity and refractive index methods. © 2003 Wiley Periodicals, Inc. J Appl Polym Sci 89: 2970–2972, 2003

Key words: blends; viscosity; refractive index; miscibility

INTRODUCTION

Combining two or more polymers by mechanical or chemical infinite mixing methods forms polymer blends. These resulting polymeric systems often exhibit properties that are superior to any of the component polymers alone. There has been a great deal of interest in the studies of these polymer systems.[1] However, the manifestation of superior properties depends upon the miscibility of the component homopolymers on a molecular scale. In general, there are three different types of blends: totally miscible, partially miscible, and immiscible. The miscibility results in altogether different morphologies of the blends, ranging from single-phase systems to two- or multiphase systems. There have been various techniques of studying the miscibility of polymer blends.[2] Some of these techniques may be complicated, costly and time consuming. Hence, it is desirable to identify simple, low cost, rapid techniques to study the miscibility of polymer blends. Chee[3] and Sun et al.[4] suggested viscometry for the study of polymer-polymer miscibility. Singh et al.[5] showed that the variation of ultrasonic velocity and viscosity with blend composition is linear for miscible blends. Recently, Varada Rajulu et al.[6] used ultrasonic and refractometric techniques to study the miscibility of polymers in a blend. In the present study, the authors employed some simple and rapid techniques to measure the viscosity, ultrasonic velocity, and refractive index of the epoxy/unsaturated polyester (UP) resin blend in chloroform at 30°C in order to study the miscibility of the blend. The authors selected epoxy and unsaturated polyester resins for the present study because both of them are widely used as matrix materials in the preparation of polymer composites.

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EXPERIMENTAL

Blends of epoxy/UP resin of different compositions were prepared by mixing solutions of the polymers in chloroform. The epoxy resin, LY 556 (Ciba Geigy Specialty Chemicals, Ltd., India) with an epoxy equivalent of 6.75 kg, and the unsaturated polyester resin (ECMALON 4413) were employed in the present study. The total weight of the two components in solution was always maintained at 1 g/dL. The ultrasonic velocities of the blend solutions were measured with a 2-MHz interferometer. The measurements had accuracy better than ±0.5%. The temperature was maintained at 30°C by circulating water from a thermostat with a thermal stability of ±0.05°C through the double wall jacket of the ultrasonic experimental cell. The refractive indices of blend solutions were measured with Abbe's refractometer using a thermo-stated water circulation system at 30°C as described elsewhere.[7] The relative viscosities of blend solutions were measured at 30°C using a Ubbelohde suspended level viscometer.

RESULTS AND DISCUSSION

Figure 1 shows the Huggin's plots for epoxy, UP, and the epoxy/UP blend, in which the weight fraction of both the components was maintained at 0.5. Chee[3] gave the expression for the interaction parameter

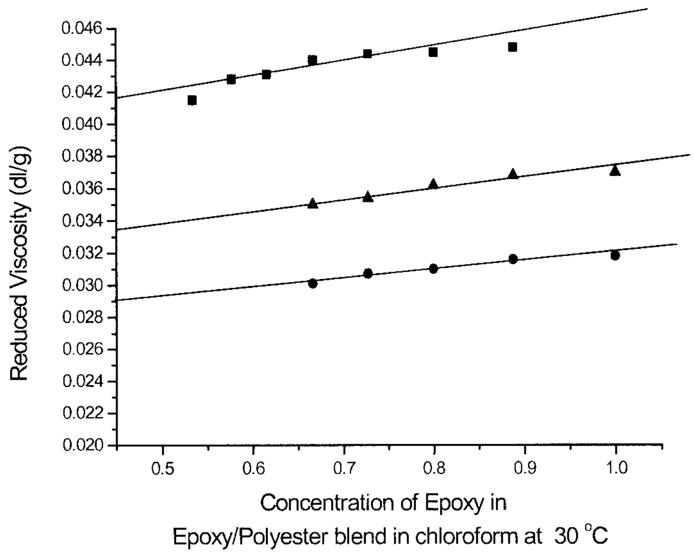

Figure 1 The variation of reduced viscosity with composition in an epoxy, UP resin, and their blend in chloroform at 30°C: (■) polyester, (▲) epoxy/polyester blend, (●) epoxy.

when the polymers are mixed in weight fractions W_1 and W_2 as

$$\Delta B = \frac{b - \bar{b}}{2W_1W_2} \quad (1)$$

Here b_{11} and b_{22} are the slopes of the reduced viscosity versus concentration curves for the epoxy and UP resin solutions respectively.

The value of b is related to Huggin's coefficient K_H as

$$b = K_H[\eta]^2 \quad (2)$$

For ternary systems, it is also given by

$$b = W_1{}^2b_{11} + W_2{}^2b_{22} + 2W_1W_2b_{12}$$

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

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

$$\mu = \frac{\Delta B}{\{[\eta]_2 - [\eta]_1\}^2} \quad (3)$$

where $[\eta]_1$ and $[\eta]_2$ are the intrinsic viscosities for the pure component solutions. The blend is miscible when $\mu \geq 0$ and immiscible[3] when $\mu < 0$. The values of μ, calculated with the above expression, are presented in Table I.

In the present study, the values of μ for epoxy/UP resin blends were found to be positive except when the weight fraction of epoxy was 0.9, 0.4, 0.2, and 0.1 in the blend. But recently Sun et al.[4] suggested a new formula for the determination of miscibility of polymers as

$$\alpha = K_m - \frac{K_1[\eta]_1^2W_1^2 + K_2[\eta]_2^2W_2^2 + 2\sqrt{K_1K_2}[\eta]_1[\eta]_2W_1W_2}{\{[\eta]_1W_1 + [\eta]_2W_2\}^2} \quad (4)$$

where K_1, K_2 and K_m are the Huggin's constants for individual components 1 and 2 and the blend respectively. While deriving this equation, the long-range hydrodynamic interactions were taken in to account. Sun et al.[4] suggested that a blend would be miscible when $\alpha \geq 0$ and immiscible when $\alpha < 0$.

In the present study, the values of α were found to be positive for all concentrations, indicating that the blend is miscible. However, for four concentrations, the μ and α values give contradictory information. A similar observation was made by Varada Rajulu et al.[8] in the case of poly(vinyl pyrrolidone)/polystyrene blends, where μ was found to be negative and α was found to be positive. They confirmed the miscibility of

TABLE I
Interaction Parameters μ and α of Epoxy/Unsaturated Polyester Resin Blends

Weight fraction of epoxy/UP resin blend	μ - Values	α - Values
0.95/0.05	9.65	7.48
0.9/0.1	−3.56	7.41
0.8/0.2	20.15	11.52
0.7/0.3	1.38	6.36
0.6/0.4	39.39	184.18
0.5/0.5	7.27	2.35
0.4/0.6	−6.87	2.95
0.3/0.7	14.93	4.85
0.2/0.8	−3.56	0.54
0.1/0.9	−3.59	3.12

Measured in chloroform at 30°C.

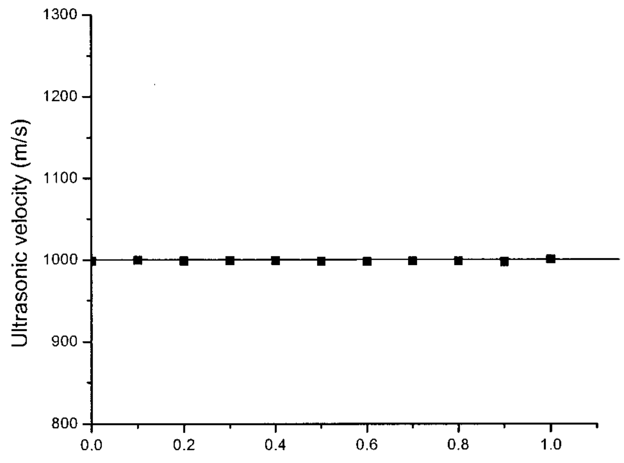

Figure 2 The variation of ultrasonic velocity with composition of an epoxy/UP resin blend in chloroform at 30°C

such blends by other methods. They further established that the equation for α is more accurate than the equation for μ, as the secondary interactions are included in it. In order to further confirm the miscible nature of the blend, the variation of the ultrasonic velocity and refractive index of the polymer blend solutions with composition are depicted in Figures 2 and 3 respectively.

From these figures, it is evident that the variation is linear, indicating a single phase in the blend. Varada Rajulu et al.[9] used these techniques for the miscibility study of poly(methyl methacrylate)/poly(vinyl chloride) blends, where a linear variation of the ultrasonic velocity and refractive index with blend composition was attributed to the miscible behavior of the blends. Similarly, the linear variation of ultrasonic velocity with blend compositions in the case of poly(methyl methacrylate)/poly(vinyl acetate) blends[10] was attributed to the miscible nature of the blends. These observations support the validity of Eq. (4) and confirm that the epoxy/UP resin blend is miscible. Furthermore, it is observed that the ultrasonic velocity and refractive index of the epoxy/UP resin blend do not vary much from those of the pure components.

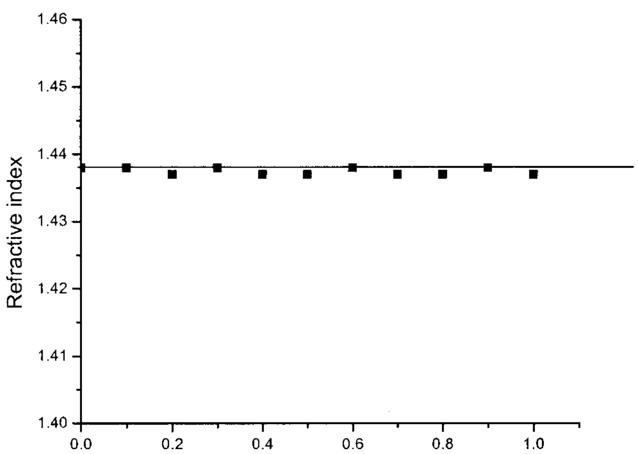

Figure 3 The variation of refractive index with composition of an epoxy/UP resin blend in chloroform at 30°C.

TABLE II
Ultrasonic Velocity and Refractive Index of Epoxy/UP Resin Blend Solutions

Weight fraction of epoxy/UP resin blend	Ultrasonic velocity (m/s)	Refractive index
Epoxy	998.8	1.438
0.9/0.1	999.5	1.438
0.8/0.2	998.6	1.437
0.7/0.3	999.0	1.438
0.6/0.4	999.1	1.437
0.5/0.5	998.1	1.437
0.4/0.6	997.8	1.438
0.3/0.7	998.5	1.437
0.2/0.8	998.3	1.437
0.1/0.9	997.6	1.438
Polyester	1000.6	1.437

Measured in chloroform at 30°C.

CONCLUSIONS

Using viscosity, ultrasonic velocity, and refractive index methods, the polymer blend of epoxy/UP resin was found to be miscible.

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