

Miscibility Behavior of Blend of Polyesteramides of Linseed Oil and Dehydrated Castor Oil with Poly(methacrylic acid)

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ABSTRACT: Blending of two polymers in solution is a simple and cost-effective technique to improve upon the physical and mechanical properties of the component polymers through synergism. To obtain maximum synergy in their properties, the component polymers should be miscible with each other on molecular scale. Polymer blends of complex physicochemical properties are being actively investigated. Poly(methacrylic acid) (PMAA), a commercial polymer, yields transparent, hard, brittle, and water-sensitive films. It has been blended with natural polymers like dextran, collagen, and gelatin to obtain films with improved physical and mechanical characteristics. Polyesteramides, which are easily synthesized from vegetable seeds oil, a sustainable resource, have found application in surface coatings. These oligomeric products do not make free standing films in the ambient condition. The polyesteramides from vegetable seeds oil can be used to obtain blend with PMMA of improved mechanical and water absorption properties. In this study, linseed oil polyesteramide (LOPEA) and dehydrated castor oil polyesteramide (DCPEA), the source oils with different unsaturation in their fatty acid chains,

were blended with PMAA through mixing in solution in the ratio DCPEA/LOPEA : PMAA as 80/20, 70/30, 60/40, 50/50, 40/60, 30/70, and 20/80. In the first instance, the miscibility of the two components was investigated in solution by viscosity and ultrasonic measurements and in solid phase through differential scanning calorimetry (DSC) and scanning electron microscopy (SEM). Moisture absorption by the blend was also studied. DCPEA and LOPEA show immiscibility with PMAA in solution phase while LOPEA with more unsaturation in the fatty acid chain of the oil was found more immiscible than DCPEA. DCPEA shows a narrow miscibility window in the solid phase while LOPEA was found immiscible with PMAA in the solid phase too. Uptake of moisture was found to be markedly reduced in the blends of DCPEA/LOPEA with PMAA. © 2006 Wiley Periodicals, Inc. *J Appl Polym Sci* 103: 1367–1374, 2007

Key words: linseed oil polyesteramide; dehydrated castor oil polyesteramide; poly(methacrylic acid); blend; miscibility

INTRODUCTION

Polymer blends are growing in importance in specific sectors of the polymer industry,^{1,2} as they can meet the performance requirements, which are not satisfied by the currently available commercial polymers. For maximum synergism in the properties in the blend, the component polymers should be miscible on molecular scale. The miscible blends have their commercial importance in the versatility they exhibit in their physical and mechanical properties, since by simply varying the composition, blends with different set of properties can be obtained. However, immiscible blends, which make heterogeneous phase, have also found commercial application.³ Considerable work has been done on blends in the last three decades.^{4–6}

Vegetable oils constitute a major resource of several polymers, viz., alkyds, polyesters, polyurethanes,

epoxies, and others.^{7,8} In addition to their other applications, vegetable-oil-based products like esters and epoxies can be used as plasticizer and additives to commercial polymers to improve upon their toughness and tensile strength.⁹ Soya bean oil epoxy has notably been used in the processing of poly(vinyl chloride).¹⁰ These additives of low molecular mass are also required to be miscible to be effective in modifying the properties of commercial polymers and also for obviating the migration of the former.

Poly(methacrylic acid) (PMAA) is a transparent, hard, and brittle polymer of high molar mass, but it is too water sensitive to serve as plastic.¹¹ Attempts have been made to improve upon its brittleness and water sensitivity through complex formation.¹² Studies on blending of PMAA with other polymers are much less. The blending of PMAA with poly(2-hydroxyethyl methacrylate) (PHEMA) has been reported.¹³ The system yields a completely miscible polymer in solid phase on a scale of 5–10 nm for all compositions. Huang and Chang¹⁴ have studied the miscibility of PMAA with poly(methyl methacrylate) by solid state ¹³C NMR and have found the complete miscibility of

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the two components. Lately, bioartificial polymers, which are obtained by blending a natural polymer with a commercial polymer, have attracted the attention of the scientists. Bioartificial polymer blends of PMAA have been studied with collagen, gelatin, starch, and dextrin.¹⁵ PMAA-gelatin blend showed good mechanical properties over a wide range of temperature. Polyacrylic acid has been found to form miscible blend with dextran.¹⁶

We have considered it profitable to study the bioartificial polymer blends of PMAA with vegetable oil polyesteramides to improve upon the physical and mechanical properties of PMAA. Polyesteramide, one of the easily synthesized product from a vegetable oil, a sustainable resource have been investigated for their surface-coating properties.^{12,13} They do not make free standing film in ambient conditions. Polyesteramides synthesized from different vegetable oils will have fatty acids chains with different unsaturation and hence will expectedly show different behavior in their interaction with a synthetic polymer with regards to the miscibility and the modification in the physical and mechanical characteristics.

In the present study, we have chosen dehydrated castor oil (DCPEA) and linseed oil polyesteramide (LOPEA), the former having 87% linoleic, 3% linolenic, and 8% oleic acid and the latter having 17% linoleic, 51% linolenic, and 22% oleic acid¹⁷ for blending with PMMA to obtain a bioartificial polymer with improved mechanical and physical properties over the original components. In the first instance, we have attempted to investigate the miscibility of the two polymers, both in solution and solid phase, using techniques of viscosity, ultrasonic velocity measurement, differential scanning calorimetry (DSC), and scanning electron microscopy (SEM).

EXPERIMENTAL

Materials

Linseed oil (LO) and dehydrated castor oil (DCO) were purchased from M/s Atul Chemicals, Delhi. The oils were dewaxed by keeping them in a refrigerator at 15°C overnight and filtering before use. Poly(methacrylic acid) (PMAA) was synthesized from methacrylic acid monomer (Aldrich, USA) in the laboratory. Molecular weight of PMAA was determined by viscosity measurement and was found to be 2.3×10^5 . The dehydrated castor oil polyesteramide (DCPEA) and linseed oil polyesteramide (LOPEA) were prepared by a reported method.¹²

Synthesis of polyesteramides

Briefly, the method consists in taking 0.1 mol of DCO/LO and heating with diethanolamine (0.3 mol) and sodium methoxide (0.007 mol) at 115°C. The reac-

tion product *N,N* bis(2-hydroxy ethyl) linseed oil fatty amide (HELA)/DCO fatty amide (HEDCA) was recovered from ethereal layer by treating the reaction product with ether, washing with NaCl solution, and removing the solvent in a rotary vacuum evaporator. HELA/HEDCA (0.05 mol) is converted into LOPEA/DCPEA by reacting it with phthalic acid (0.08 mol) in xylene at 165°C for 4 h (vide reaction Scheme 1). Final product is recovered from the solvent in a rotary vacuum evaporator and purified by solvent-nonsolvent method.

Blending

The blends of LOPEA and DCPEA with PMAA were prepared by mixing the LOPEA/DCPEA with PMAA in the weight ratios, 80/20, 70/30, 60/40, 40/60, 30/70, and 20/80, to obtain 2 and 4 wt % solutions of LOPEA : PMAA and DCPEA:PMAA blends in dimethyl sulfoxide, (Merck (AR), India). Total volume of the solution was kept 100 mL. All the solutions were thoroughly mixed by agitation on a magnetic stirrer. Selected samples were kept for over a week. Appearance of turbidity or precipitation was not noticed in any case in the above period, but beyond this period, separation of phases was noticed.

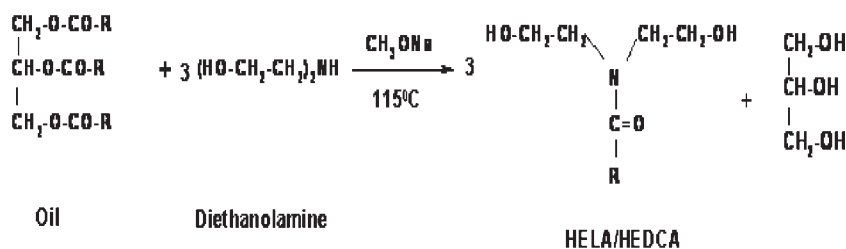
Film preparation

A few milliliters of 6 wt % solution of selected compositions of the blend in DMSO was poured on a transparency sheet and was allowed to dry under ambient conditions. In 10 days, freestanding films were obtained. The films were further dried in vacuum kept at 60°C for 8 h.

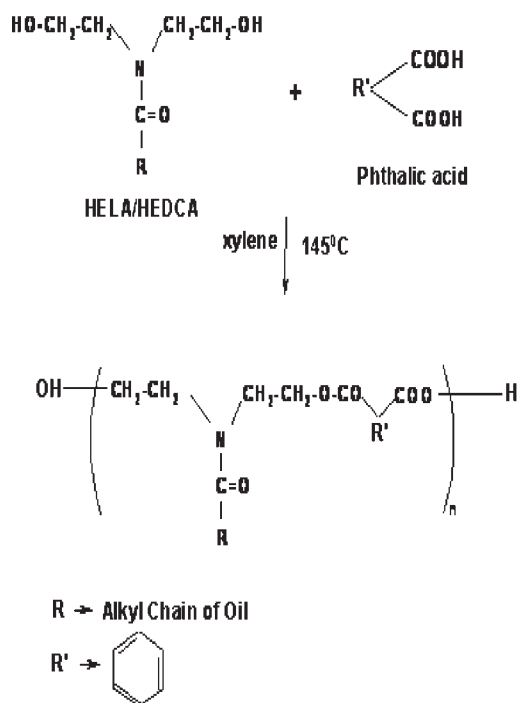
Investigations of the blends

LOPEA:PMAA blends were studied experimentally by viscosity method, ultrasonic velocity measurements, DSC, and SEM techniques. DSC thermograms were recorded on differential scanning calorimeter, Dupont 910 (TA Instruments, New Castle, USA) under N₂ atmosphere at a heating rate of 20°C min⁻¹. Viscosity of the blend solutions was measured at temperatures 20 and 30°C (accuracy = $\pm 0.05^\circ\text{C}$) in a thermostatic bath using an Ubbelohde suspended level viscometer. The ultrasonic velocity of the blend solutions was measured by an ultrasonic interferometer (Model MX-20, Mittal Enterprises, New Delhi, India) at the above-mentioned temperatures. The temperature in the sample cell was maintained by circulating water from a thermostatic bath through the outer jacket of the sample cell with a thermal stability of $\pm 0.1^\circ\text{C}$. The densities of the 2 wt % solutions of different compositions of DCPEA : PMAA and LOPEA : PMAA blends at $(30 \pm 0.2)^\circ\text{C}$ were measured by a pycnometer. The morphology of a cross section of the

a) Synthesis of HELA/HEDCA



b) Synthesis of DCPEA/LOPEA



Scheme 1

cryogenically fractured blend films was examined by scanning electron microscope (JEOL, JSM-840 scanning electron microscope) at different magnifications. SEM micrographs were taken after coating the surface with a thin layer (10–20 nm) of gold.

Moisture absorption properties of the blends were studied using a standard technique. Weighed portions of the perfectly dried samples were exposed to a dessicator containing saturated NaNO_3 solution (maintaining 65% relative humidity inside) until a constant weight was reached. From the weight gain, the moisture absorption (%) was calculated.

RESULTS AND DISCUSSION

Viscosity measurements

The relative viscosity of the blend compositions of LOPEA : PMAA and DCPEA : PMAA in the composi-

tion range of 20–80 wt % of PMAA in the blend was determined in DMSO at 2 and 4% concentrations at temperatures 20 and 30°C [Fig. 1(a,a', b,b', c,c', and d,d')]. It was observed that in both the blend systems, the relative viscosity decreases linearly up to 40 wt % PMAA in the blend. However, at the latter composition in both the blends, a sharp inflection is observed. Beyond this composition, the viscosity increases almost linearly and sluggishly in DCPEA : PMAA blend, while in LOPEA : PMAA blend relative viscosity increases sharply and to higher values with a wide inflection at the composition 70 wt % PMAA in the blend. The sharp inflection at the composition DCPEA/LOPEA : PMAA (60 : 40) indicates a phase inversion at this composition. It can be presumed that the polymer PMAA is dispersed into DCPEA/LOPEA phase in the initial composition range. DCPEA/LOPEA will tightly envelop PMAA molecules and will cause

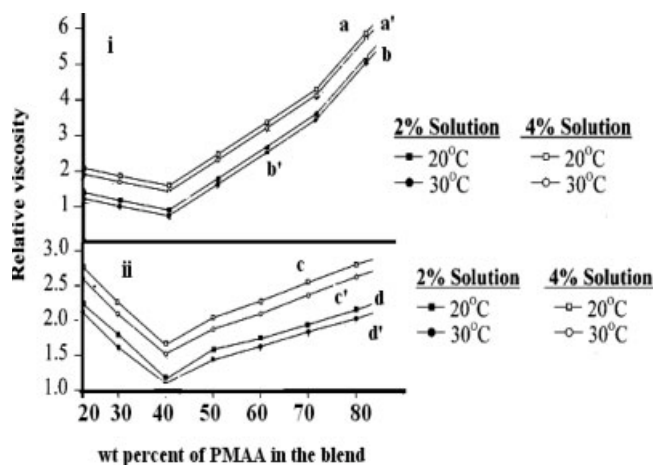


Figure 1 Effect of varying composition of (i) LOPEA : PMAA (ii) DCPEA : PMAA blend on the relative viscosity of 2 and 4% solution.

decrease in the viscosity. Beyond the composition, 40 wt % PMAA in DCPEA/LOPEA blends, as a result of the phase inversion, the DCPEA/LOPEA can now be presumed to be dispersed into PMAA phase. The DCPEA/LOPEA molecule will orient between PMAA molecules and entangle them leading to an expanded structure, which brings about increase in the viscosity as has been actually observed in this region. In case of LOPEA, the increase in viscosity has been found to be sharper and higher than the same in case of DCPEA. Because of the difference in the unsaturation in the fatty acid chains in the two polyesteramides, their interaction with PMAA and the resulting structures of blends will be different. This also causes the extent of phase separation and phase inversion in the two blend systems to be different.

The nonlinearity of the composition-relative viscosity plot with phase inversion establishes the immiscibility of the DCPEA/LOPEA with PMAA in solution. Sharma and coworkers^{18,19} and Mamza and Folaranni²⁰ have also observed that the plot of the relative viscosity of the blend solution against composition is linear at all concentrations and temperatures when the components are miscible with each other, while if the plot is nonlinear at any concentration or temperature, the components are immiscible or partially miscible depending upon the extent of nonlinearity. We find that the trend in the variation of relative viscosity with composition in 2 and 4 wt % solutions of the two blend systems at 20 and 30°C (and also at 40°C, plot at this temperature not given in the Fig. 1) is the same that shows the stability of the interaction with temperature in each case.

Reduced viscosity

We have attempted to compare the observed and calculated reduced viscosity values of the 2 wt %

DCPEA : PMAA and LOPEA : PMAA blend solutions of different compositions. The calculated reduced viscosity of the blend solutions was computed by rule of mixture.^{21,22} Paladhi and Singh²² have observed that the reduced viscosity of blend compositions follows the simple additive rule of mixture when the polymers are miscible. The calculated reduced viscosity will, therefore, follow a linear relationship against composition of the blends. Figure 2(a,a' and b,b') shows the calculated and observed reduced viscosity plots against the composition of the 2 wt % solution respectively, of DCPEA : PMAA and LOPEA : PMAA blend systems at 30°C. It is observed that reduced viscosity of the DCPEA : PMAA blends is much lower than the corresponding values of the calculated reduced viscosity. Moreover, the plot is highly nonlinear with a sharp inflection at the composition 45 wt % PMAA in the blend. The inflection shows phase inversion. These features clearly establish the overall immiscibility of DCPEA with PMAA.

The opposite trend in the variation of the observed reduced viscosity before and after phase inversion indicates the different structure of the DCPEA : PMAA composite molecule in the two regions. The broad structure of the DCPEA : PMAA composite molecules in the two regions is discussed in a preceding section. The lower values of the observed reduced viscosity than the calculated reduced viscosity values between 20 and 40 wt % PMAA in the blend shows a compact structure because of a larger interaction between DCPEA and PMAA molecules as the former envelops the latter in this composition range. It also appears that this structure of the DCPEA : PMAA composite molecules breaks the structure of the medium, which also contribute to the decrease of the observed reduced viscosity values. Beyond phase inversion, the structure will change as discussed in a proceeding

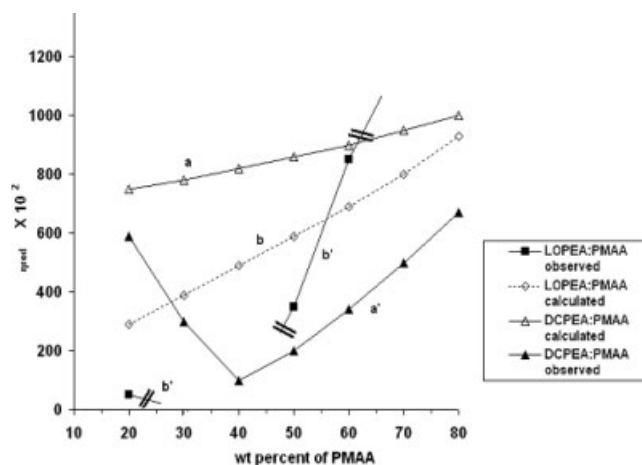


Figure 2 Effect of varying composition of DCPEA/LOPEA : PMAA blend on the reduced viscosity of 2 wt % solution at 30°C.

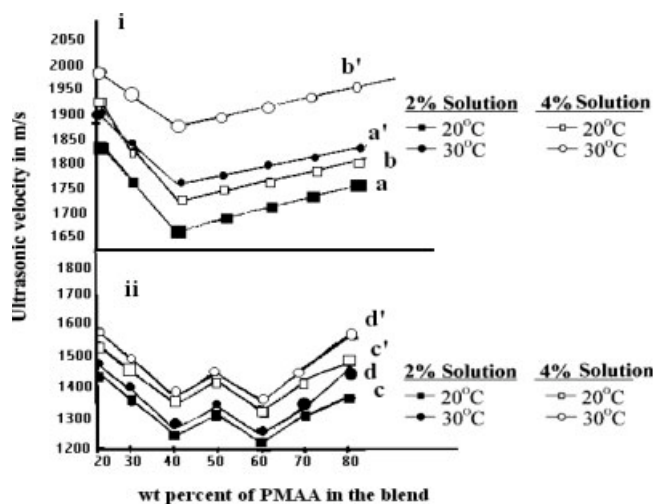


Figure 3 Effect of varying composition of (i) LOPEA : PMAA (ii) DCPEA : PMAA blend on the ultrasonic velocity of 2 and 4 wt % solution.

section causing increase in the observed reduced viscosity.

In case of LOPEA:PMAA blends [Fig. 2(b,b')], the variation of observed reduced viscosity with composition is slightly different from the same in DCPEA : PMAA system. In this case, the observed reduced viscosity values beyond 20 wt % PMAA in the blend becomes zero and dips to below zero values (this part of the plot not shown in the figure), indicating that LOPEA : PMAA composite molecules offer no resistance to flow in the medium and also break the structure of the medium considerably to reduce the viscosity of the medium below its normal value. Beyond 40 wt % PMAA in the blend, the observed reduced viscosity values increase very rapidly and crossover the calculated reduced viscosity composition plot. The marked difference in the observed reduced viscosity values of LOPEA : PMAA blends from those of DCPEA : PMAA blend highlight the marked difference in the extent of interaction between the components molecules.

Ultrasonic velocity measurements

The ultrasonic velocity studies of the DCPEA/LOPEA : PMAA blends of varying compositions have been studied at 20 and 30°C [Fig. 3(a,a', b,b', c,c', d,d')]. Figure 3(a,a' and b,b') shows that the ultrasonic velocity of DCPEA : PMAA system decreases linearly as the amount of PMAA in the blend increases till the composition 40 wt % PMAA in the blend at which a sharp inflection occurs showing phase inversion. Beyond this composition, the variation of ultrasound velocity is nonlinear for both 2 and 4 wt % blend solution at 20 and 30°C. This behavior clearly shows the immiscibility of the two components. Different authors^{22–24}

have observed that for miscible solutions, ultrasound velocity varies linearly with composition at all concentrations and temperatures, which is not found in the above-mentioned case.

The LOPEA : PMAA system shows multiple inflection in the ultrasound velocity composition curves [Fig. 3(ii) (c,c', d,d')] for 2 and 4 wt % solution at 20 and 30°C. This shows high immiscibility of the two components. We notice that because of the variation in fatty acid components in DCPEA and LOPEA, marked difference in the shapes of these curves in the two cases occurs. Ultrasound velocity like reduced viscosity curves also brings out very sharply the difference in the structures of DCPEA : PMAA and LOPEA : PMAA composite molecules because of the difference in the fatty acid chain structure and conformation in the two polyesteramides.

Adiabatic compressibility

Using ultrasound velocity, adiabatic compressibility of the system can be calculated by the following equation.^{21,25}

$$\beta = 1/v^2\rho \quad (1)$$

where β is the adiabatic compressibility of the medium, v is the velocity of the sound waves, and ρ is the density of the medium. Adiabatic compressibility is inversely proportional to the cohesive energy of the polymer molecules²⁶. In DCPEA : PMAA systems, the adiabatic compressibility plots show sharp inflection at the composition DCPEA : PMAA, 60 : 40 [Fig. 4(ii) (c,c' and d,d')], which coincides with the inflection at this composition in relative viscosity and ultrasound

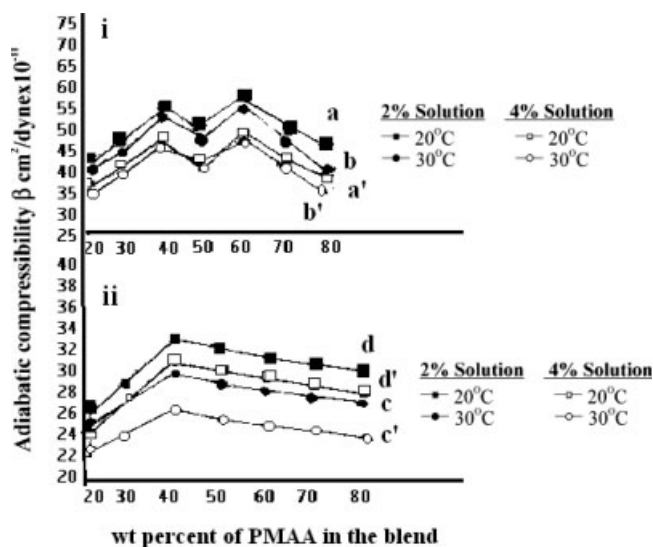


Figure 4 Effect of varying composition of DCPEA/LOPEA : PMAA blend on the adiabatic compressibility of 2 and 4 wt % solution.

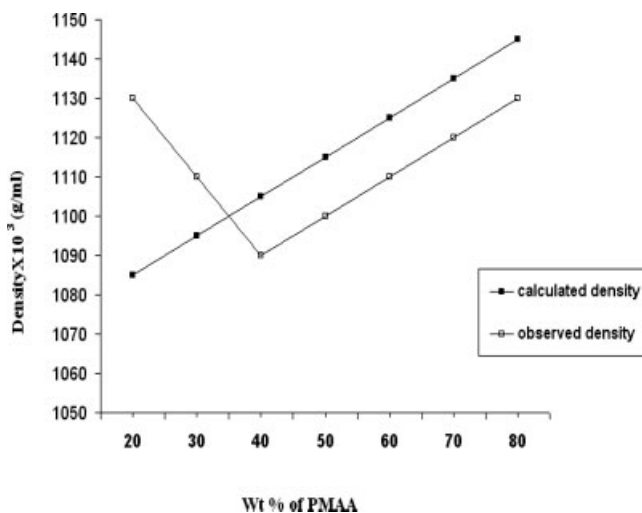


Figure 5 Effect of varying composition of DCPEA : PMAA blend on the density of 2 wt % solution at 30°C.

velocity versus composition curves. The inflection is correlated to the phase inversion in the system. Beyond the composition of phase inversion, the plots are slightly nonlinear in both 2 and 4 wt % solutions at 20 and 30°C. The overall nonlinear behavior with phase inversion shows immiscibility of the two components.

In LOPEA : PMAA systems, the adiabatic compressibility plots show multiple inflection in 2 and 4 wt % solutions at 20 and 30°C [Fig. 4(i) (a,a' and b,b')], which indicates multiple phase inversions/phase separations. In this case, conformation and structure of LOPEA : PMAA composite molecules appear to be highly dependent upon the composition of the blend. This causes gross immiscibility of the two components in the blend. Comparing the two polyesteramides, it can be inferred that in DCPEA : PMAA system there are basically two blend structures, one before and the other after phase inversion composition, while in case of LOPEA : PMAA system, more than two blend structures occur in the composition range studied.

Density measurements

The density of the 2 wt % solutions of blends of different compositions at 30°C calculated on the basis of additivity of volumes and compared with corresponding observed densities provides evidence of miscibility or immiscibility of components in the blend. In case of immiscible blend, the observed densities have been found to agree with the calculated densities or to be lower than the latter.^{25,26} Observed densities in miscible blends were found to be higher up to 5% than the calculated densities.²³ Figure 5 shows that in DCPEA : PMAA blend system, observed density varies nonlinearly with the composition of the blend.

The curve shows an inflection at the composition 40 wt % PMAA in the blend. For lower amount of PMAA in the blend, the density values of the blend compositions are slightly higher than the corresponding values of the calculated density, but beyond the point of inflection the observed density values are almost equal to the calculated density values. The inflection point in the observed density plot establishes phase inversion, which has already been observed in this system from other techniques. The overall nonlinear nature of the density composition plot along with lower values of observed density than the calculated ones in greater part of the composition range of the blend system establishes that PMAA and DCPEA are immiscible in solution phase.

Since the densities of 2 wt % solutions of LOPEA and PMAA are quite close to each other, being respectively, 1.096 and 1.076 g/cm³, no meaningful information could be derived from the observed and calculated density plots in this case and hence has not been shown.

Film study

The films of LOPEA : PMAA blend with 20 wt % PMAA were found to be free standing, dark brown, opaque, and brittle. The films of composition LOPEA : PMAA, 60:40 was dark brown, opaque, and more fragile, and brittle than the films of previous composition. The films of blend with still higher amount of PMAA were all opaque, dark brown, and highly fragile. The films of the blend with 80 wt % PMAA did not yield free standing film as it remained a gel-like mass. The above-mentioned observations also lead to the conclusion that LOPEA is immiscible with PMAA.

In case of DCPEA : PMAA blends, with 20 wt % PMAA, a transparent and highly stiff film is obtained. In the blend composition, DCPE : PMAA larger than 60 : 40, the films are highly fragile and break when they are removed. The presence of heterogeneous phase in the films is responsible for fragility of the films. It also shows the immiscibility of two components in the solid phase. It appears that when PMAA is about 20 wt % in the DCPEA, they have some mutual miscibility in solid phase as the films of this composition are transparent and very stiff.

Thermal analysis

DSC thermogram of pure PMAA Figure 6(a) exhibits glass transition temperature T_g at 175°C followed by an endothermic peak, which can be due to the decomposition of —COOH group. DSC thermogram of DCPEA : PMAA blend with 20 wt % PMAA [Fig. 6(b)] shows glass transition temperature event at 55°C. The drastic lowering of T_g of this composition from pure PMAA shows miscibility of the two components in this composition of the blend. DSC thermogram of DCPEA :

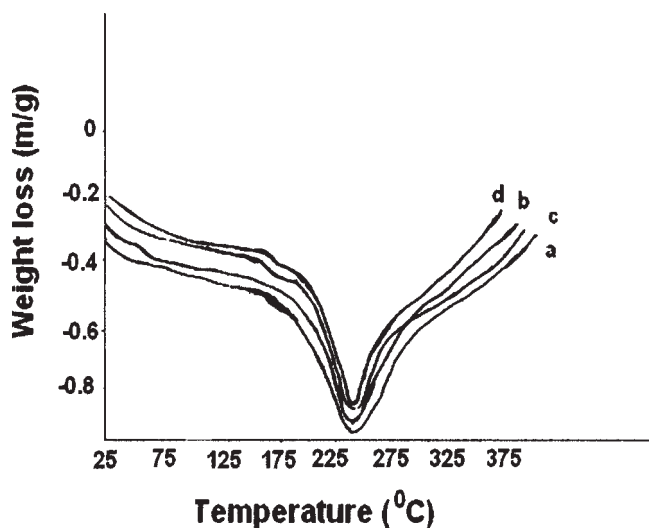


Figure 6 DSC thermogram of DCPEA/LOPEA : PMAA blends (a) pure PMAA, DCPEA : PMAA 80 : 20, (b) DCPEA : PMAA 60 : 40, and (c) LOPEA : PMAA 80 : 20.

PMAA blend with composition 40 wt % PMAA [Fig. 6(c)] shows T_g at 174°C, indicating heterogeneity in the solid phase and immiscibility of DCPEA and PMAA in solid phase at this composition. LOPEA : PMAA blend with 20 wt % PMAA [Fig. 6(d)], however, shows a T_g at 175°C, indicating immiscibility of the two components in this composition. LOPEA : PMAA blend with 40 wt % PMAA also does not show any change in T_g . DSC thermograms confirm the observation regarding the immiscibility of LOPEA and DCPEA with PMAA obtained by other techniques.

Morphological studies

The SEM micrograph of the film of DCPEA : PMAA blend with 20 wt % PMAA at 1000× [Fig. 7(a)] shows broadly a single-phase uniform sheet structure with

an occasional appearance of small domains of another phase. The microstructure predicts that the film will be tough and transparent, which has actually been observed. Since films of other compositions were highly fragile, their SEM could not be obtained. The SEM micrograph of LOPEA : PMAA blend with 20 wt % PMAA [Fig. 7(b)] shows layers of the blend lying one above the other. It appears that during film formation, the blend separates into two phases each making a separate layer. The micrograph also shows other small domains. The film, therefore, appears broadly heterogeneous. The films will, therefore, be opaque, which has been actually found. The microstructure also indicates poor stiffness of the films. It can therefore be concluded that both the LOPEA and DCPEA are immiscible with PMAA in solid phase. However, in case of DCPEA : PMAA blend, a small miscibility window below the composition 40 wt % PMAA in the blend is observed.

Moisture absorption studies

The absorption of moisture (wt %) by blends [Fig. 8] was found to be significantly lower than the pure PMAA. Pristine PMAA showed moisture absorption of about 40 wt % in about two weeks while DCPEA : PMAA and LOPEA : PMAA blends containing 80 wt % PMAA exhibited a maximum absorption of about 30 and 20 wt %, respectively, in the same period. The blend containing 80 wt % DCPEA/LOPEA and 20 wt % PMAA showed moisture absorption of 13 and 11 wt %, respectively [Fig. 8]. Hence with the increase in the DCPEA/LOPEA content in the blend, the moisture absorption was found to decrease significantly. It is interesting to note that even a lower loading of LOPEA in the blend (as low as 20 wt %) was found to reduce the moisture uptake by 10 wt %. The blending of LOPEA/DCPEA with PMAA was thus found to

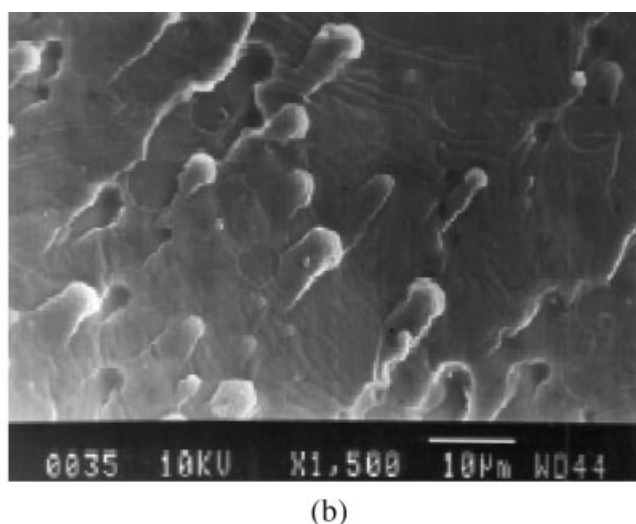
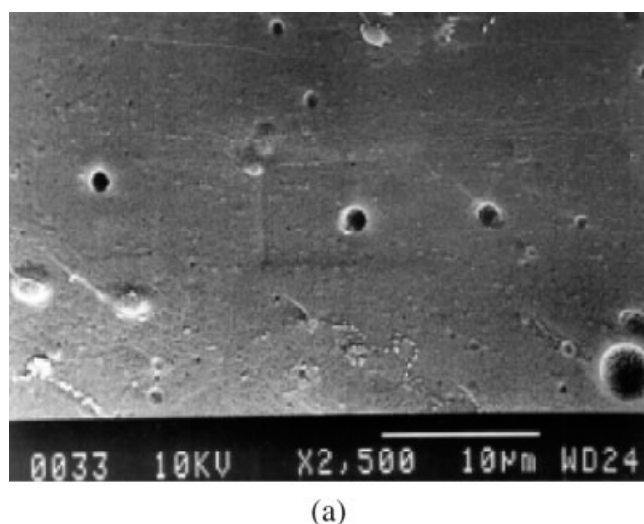


Figure 7 (a) SEM micrograph of DCPEA : PMAA 60 : 40 and (b) DCPEA : PMAA 40 : 60.

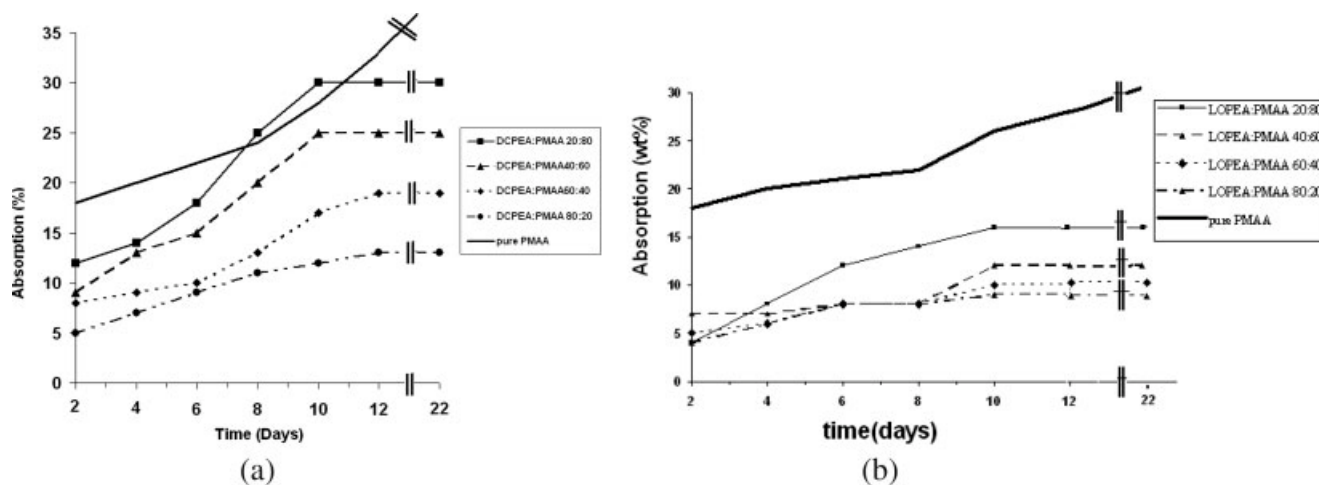


Figure 8 Moisture absorption of (A) DCPEA-PMAA and (B) LOPEA:PMAA blends.

significantly reduce the moisture absorption of the blends, which is an important aspect for the commercial utility of these blends. It is observed that while pure PMAA shows continuous increase in the moisture absorption with time in the blends of all compositions, the moisture absorption tends to level off after two weeks exposure.

CONCLUSIONS

DCPEA and LOPEA show immiscibility with PMAA in solution phase. LOPEA shows higher immiscibility than DCPEA as was found by viscosity and ultrasonic velocity measurements. LOPEA also shows higher immiscibility with PMAA in solid phase. In fact, DCPEA shows some miscibility with PMAA in solid phase below 40 wt % PMAA. The films of DCPEA : PMAA blend with 20 wt % PMAA were transparent and highly stiff. It also showed low water absorption of 13 wt % in two weeks.

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