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Rheological and morphological behaviour of blends of polycarbonate with unmodified and maleic anhydride grafted ABS

S. Balakrishnan[a], N.R. Neelakantan[a,*], D. Nabi Saheb[b] and Jyoti P. Jog[b]
[a]*Department of Chemical Engineering, Indian Institute of Technology, Madras, Chennai-600 036, India*
[b]*Chemical Engineering Division II, National Chemical Laboratory, Pune 411 008, India*
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Blends of polycarbonate (PC) with ABS and polycarbonate with maleic anhydride grafted ABS (MABS) were prepared by melt blending, using a single-screw extruder with special mixing head over the whole range of compositions. The rheological behaviour was determined using a capillary rheometer. The melt viscosity–composition curves for PC/ABS blends showed negative deviation from the rule of mixture with minimum values over a wide range from 25 to 65 wt.% PC. These curves for PC/MABS also showed negative deviation, but the deviation is less than that for PC/ABS blends. Addition of ABS or MABS to PC improves the processibility. Morphological studies of these blends were done using a scanning electron microscope (SEM). Morphology of unmodified blends showed coarse dispersion, whereas that of the modified blends showed fine and lamellar dispersion. © 1998 Published by Elsevier Science Ltd. All rights reserved.

(Keywords: reactive blending; polycarbonate/ABS blends; deterioration)

INTRODUCTION

Polycarbonate (PC) is used in specialty applications due to its high toughness, higher continuous working temperature, high modulus and transparency. The drawbacks of PC are high melt viscosity (difficult processibility) and notch sensitivity. The disadvantages of PC can be overcome by blending with various thermoplastics/thermoplastics elastomers, of which ABS is the most popular[1,2]. The addition of ABS not only minimizes the drawbacks of PC but also retains the other superior mechanical properties and also generates other useful properties, such as glossiness and low-temperature toughness. PC/ABS alloys are largest selling commercial polymer alloys in the world and are replacing polyester and nylon-based alloys in engineering applications.

There are many references on mechanical, thermal, processing and morphological behaviour of PC/ABS blends. An excellent review on PC/ABS blends has been given by Greco and Sorrentino[3]. Many of the literature reports indicate that PC/ABS blends of particular compositions have a melt viscosity equal to or lower than that of virgin ABS. Lee *et al.*[4] have demonstrated that the presence of catalysts residue (metal salts) like $CaCl_2$ and $MgCl_2$ in the ABS causes deterioration of PC during melt blending which generates low-molecular weight species and volatiles. Chin and Hwang[5] also reported similar results.

PC/ABS blends are immiscible[6]. In the more popular commercial processes like reactive compatibilisation, core–shell compatibiliser is used to compatibilise the immiscible blends. For example, methyl methacrylate, styrene and maleic anhydride-based core–shell compatibiliser are being used to compatibilise PC/ABS systems[7]. In recent times plastics have been toughened by recently publicized reactive blending methods. For example, polyamide[8], polypropylene[9] and polyethylene terephthalate[10] are toughened using maleic anhydride grafted ABS. The same technique was followed for toughening polybutylene terephthalate with maleic anhydride grafted ethylene–propylene copolymer[11]. Horiuchi *et al.*[12] have demonstrated the reactive compatibilisation of maleic anhydride grafted styrene–ethylene–butadiene terblock copolymer with a polyamide/polycarbonate blend system. In the above studies, the interesting feature is that the toughness improvement/compatibilisation could be achieved throughout the whole composition range of the blends.

The present work focuses on the rheological and morphological characteristics of maleic anhydride grafted ABS (MABS) with PC through a reactive blending technique. This will be an alternate route to the present commercial PC/ABS alloy manufacturing processes. To our knowledge no similar work has been reported to date.

EXPERIMENTAL

Materials

Polycarbonate (Lexan ML 3403), extrusion grade, was obtained from GE Plastics India Ltd., Baroda, India. ABS (ABSOLAC-SP600) was obtained from ABS Industries Ltd., Baroda, India.

Procedure

PC and ABS were dried at 110 and 90°C, respectively, in a vacuum oven for 8 h. Maleic anhydride grafted ABS (MABS) was prepared by grafting maleic anhydride on ABS. PC was blended with ABS at different weight ratios of 25, 35, 50, 65, 75 and 90 using a single-screw extruder with a special mixing head. The above samples have been, respectively, coded as P25, P35, P50, P65, P75 and P90. PC

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* To whom correspondence should be addressed

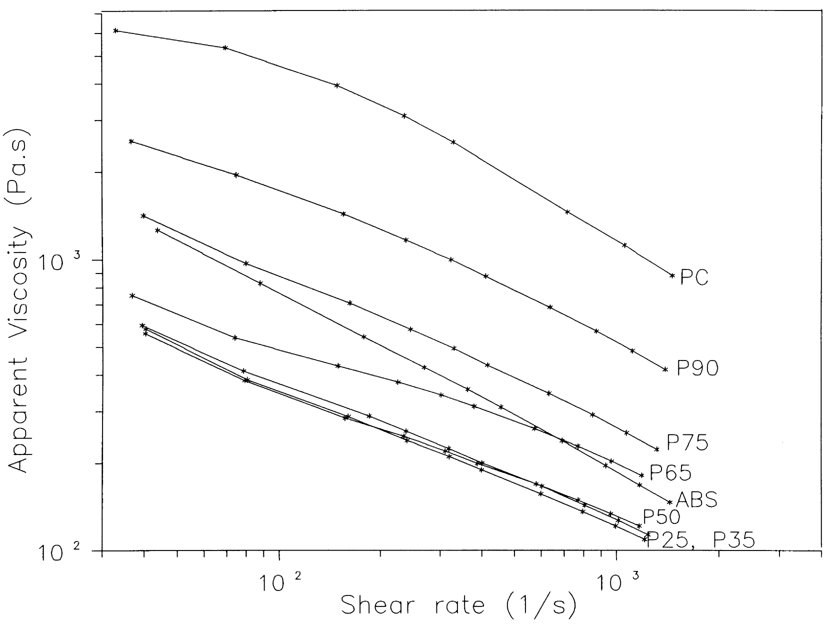

Figure 1 Apparent viscosity *versus* shear-rate curves for PC/ABS blends at different blend compositions

was also blended with MABS at the same weight ratios and similar conditions and these samples have been coded as MP25, MP35, MP50, MP65, MP75 and MP90. The process temperature was increased from 220 to 270°C and progressively as the PC weight fraction increased. The extruded pellets of these blends were vacuum dried at 90°C for 8 h then used for rheological studies and injection moulding to make samples for microstructure examination.

Characterization

The melt rheological studies were carried out using a Ceast Rheovis (Model 2100) capillary rheometer. The diameter of the die was 0.5 mm and the length was 20 mm. As the length to diameter ratio of the die was 40, there was no need for end correction. Rabinowitsch corrections were made for shear-rates. The correction factors were different for PC and ABS, but for the blends the correction factors were closer to those of PC. Between the experiments the die was removed and the barrel was cleaned with cotton cloth. Then the die was remounted and cleaned by purging twice with the material concerned.

Phase morphology has been characterized using a Jeol (JSM 5300) scanning electron microscope (SEM). Scanning electron micrographs for notched Izod impact-fractured samples of all the blends and chemically etched surfaces of selective compositions were made to determine, respectively, ductile/brittle characteristics and actual morphology. In the case of notched fractured samples, the SEM observation was made 7 mm away from the notch. This was maintained for all the samples. Alkaline etching was done to remove minor PC dispersed phase in the ABS-rich compositions (P35 and MP35). Acid etching was done to remove minor ABS dispersed phase in the PC-rich compositions (P65 and MP65). The detailed etching procedure is reported elsewhere[13].

RESULTS AND DISCUSSION

Melt rheological properties

The rheological properties give an idea about the ease of processing of PC on the addition of ABS or MABS. The apparent viscosity *versus* shear-rate plots for PC/ABS and PC/MABS blends at 240°C over the entire range of compositions are given in *Figures 1* and *2*, respectively. The plots indicate decrease in viscosity with increasing shear-rate, properties characteristic of shear thinning fluids.

From these curves, plots of viscosity *versus* wt.% of PC (*Figure 3*) are generated for PC/ABS and PC/MABS blends at two different shear-rates (100 and 1000 s^{-1}). *Figure 3* shows negative blending effect, and the viscosity of PC/ABS blends is lower than that of the PC/MABS blends. PC/ABS blends have lower viscosity than that of ABS itself, up to 65 wt.% level of PC, and any further addition of PC increases the melt viscosity. At 75 wt.% PC level, the viscosity equals that of ABS. For PC/MABS blends the melt viscosity is always more than that of MABS or ABS. The viscosity of PC/MABS blends up to 35 wt.% of PC is nearly same as or little higher than that of MABS. Further addition of PC up to 75 wt.% increases the viscosity proportionately and, beyond this, viscosity rises sharply. Both PC/ABS and PC/MABS blends have much lower viscosity than that of PC. Even 10 wt.% addition of MABS to PC reduces the viscosity (at 1000 s^{-1}) by 50% of the PC. Addition of 25 wt.% MABS to PC reduces the viscosity (at 1000 s^{-1}) to one-third of PC.

This marked low viscosity of PC/ABS blends may be because of three factors: (i) plasticisation; (ii) degradation of PC by metal salts present in ABS; and (iii) poor interfacial interaction. Plasticisation effect is caused by low-molecular weight fractions of ABS. These low-molecular weight fractions enter in between the PC molecular chains and reduce the interactive forces, thereby allowing free movement of chains[14]. The metal salts present in the ABS are known to degrade PC in the molten state[4]. This produces low-molecular weight fractions and volatiles, which reduce the melt viscosity.

The observation that the PC/MABS blends show higher melt viscosity than that of PC/ABS blends can be explained based on the same reasoning and may be because of combination of the following processes. The catalyst residues might have been denatured during the melt grafting of maleic anhydride on ABS and hence prevent attack of

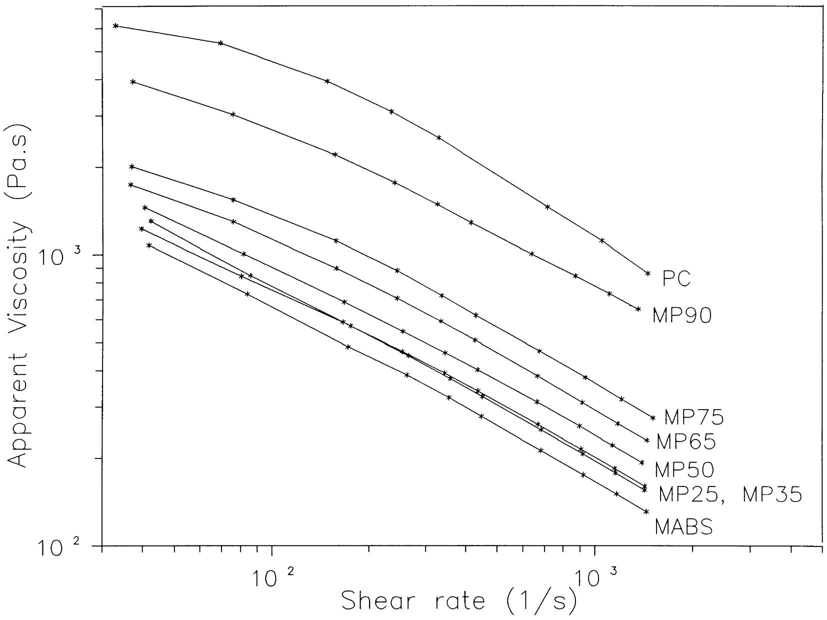

Figure 2 Apparent viscosity *versus* shear-rate curves for PC/MABS blends at different blend compositions

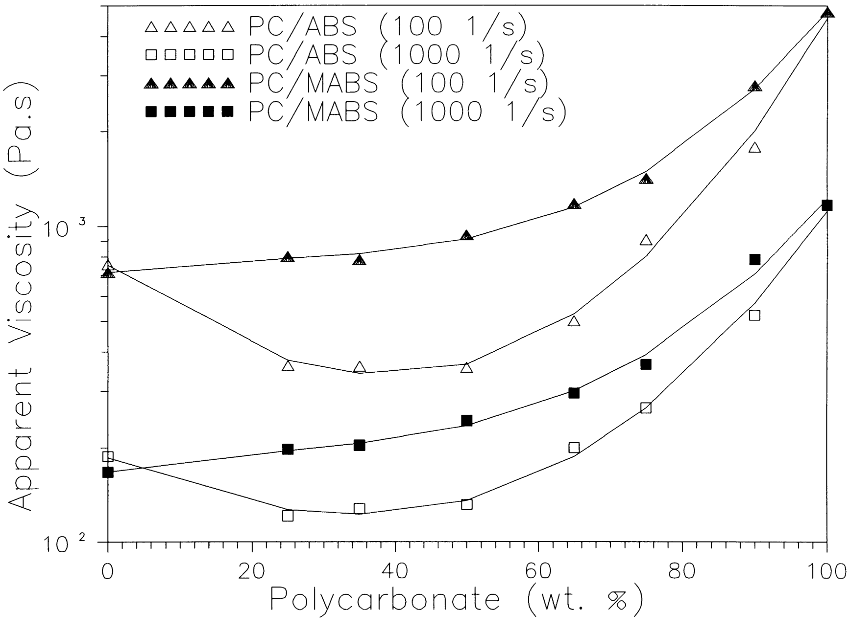

Figure 3 Apparent viscosity at shear-rates 100 and 1000 s^{-1} *versus* blend composition for PC/ABS and PC/MABS blends

coagulants on PC. Even if denaturing does not occur in the grafting stage, the deterioration initiated by catalyst residue[4] might have been chain transferred with anhydride of MABS in the compounding stage, thus preventing further propagation of deterioration. In the PC/MABS blends, the interfacial interaction is likely to be stronger than that in the PC/ABS blends. This also can be an explanation for the higher melt viscosity of PC/MABS blends.

Morphology characteristics

Morphology of PC/ABS blends is complex. It depends on the type of ABS, processing machinery, processing parameters like operating speed and temperature, viscosity ratio and volume fraction. Morphology of PC/ABS blends also depends on: (i) interfacial interaction; (ii) degradation of PC; and (iii) differential shrinkage between constituents. Lack of interfacial interaction between PC and ABS is known to produce coarse morphology. In commercial processes, the interfacial interaction is improved by the addition of a compatibiliser/copolymer[7]. As we have mentioned earlier, the melt degraded fractions which leave behind pin holes and voids cause stress concentration sites when subjected to load[4]. Shrinkage differences of polymer constituents cause interlaminar voids which can be avoided through a compatibilisation process[12].

SEM micrographs of PC/ABS blends with different weight fractions are shown in *Figure 4*. Irrespective of the composition, the micrographs of fractured surfaces show coarse dispersion and rough surfaces characteristic of brittle failure. The pin holes on fractured surfaces are due to PC deterioration. The morphology of these blends is explored in detail through selective etching studies. Micrographs of etched surfaces of PC/ABS blends (*Figure 6*) show irregular and coarse cavities, which were previously occupied by

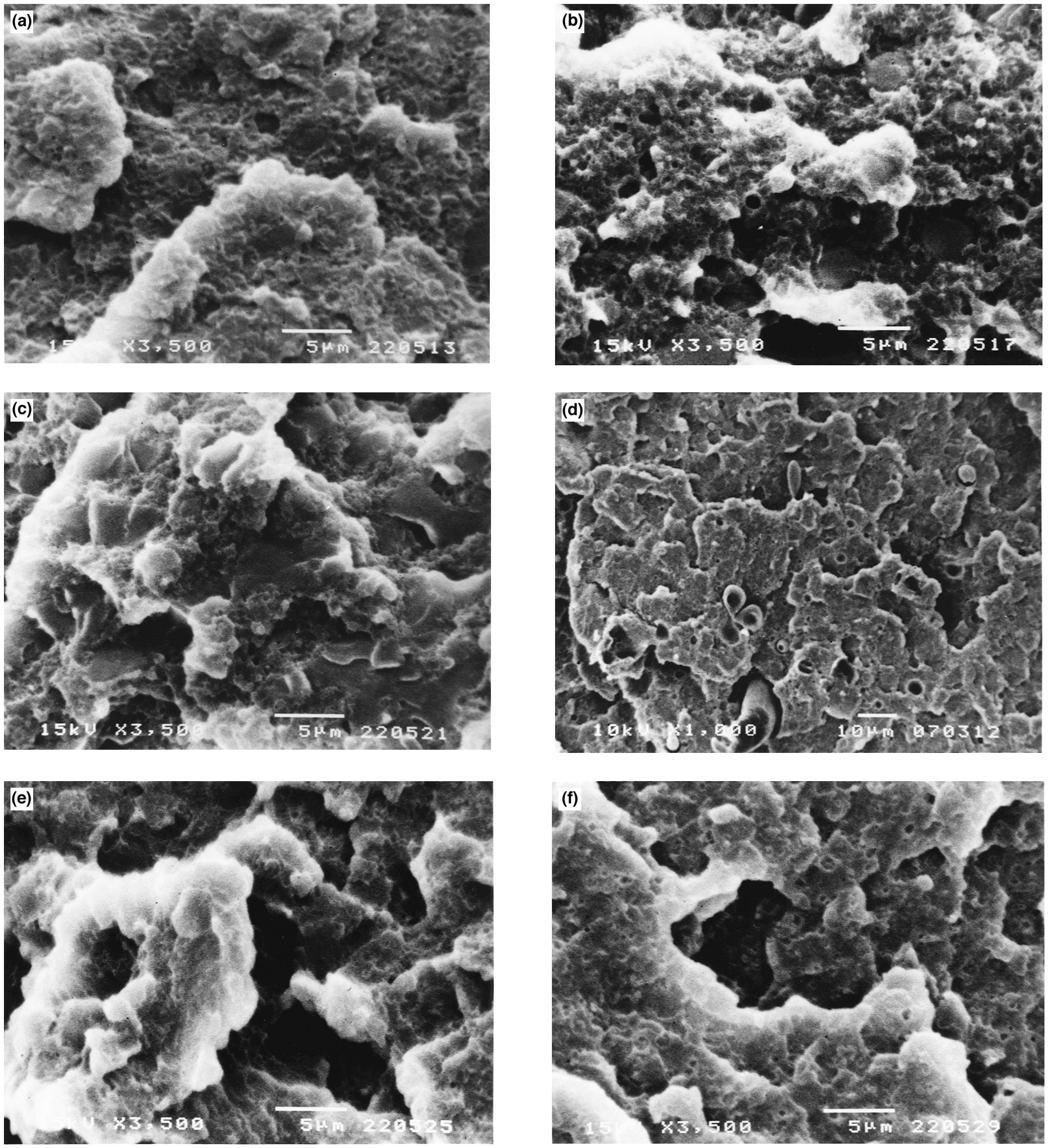

Figure 4 SEM micrographs of notched impact-fractured specimens of PC/ABS blends: (a) P25 (×3500), (b) P35 (×3500), (c) P50 (×3500), (d) P65 (×1000), (e) P75 (×3500), (f) P90 (×3500)

dispersed phase. In P35 coarse cavities were previously occupied by dispersed PC domains and in the case of P65 were due to ABS domains.

SEM micrographs of impact-fractured surfaces of PC/MABS blends are shown in *Figure 5*. Irrespective of the compositions, the micrographs of fractured surfaces of these blends show largely deformed zones characteristic of shear yielding and plastic deformation. These micrographs show that the plastic deformation increases up to 75 wt.% PC addition to MABS. Even at 90 wt.% PC, the fractured surface shows plastic deformation and shear yielding. These are characteristic of ductile failure. The ductile behaviour is explored in detail through selective etching studies for PC/MABS blends. Etched surfaces of PC/MABS blends (*Figure 6*) show fine cavities, which were previously occupied by lamellae. In MP35, the cavities/holes are finer than P35, and these are due to PC lamellae. In MP65, the cavities/holes are also finer than P65 and these are due to

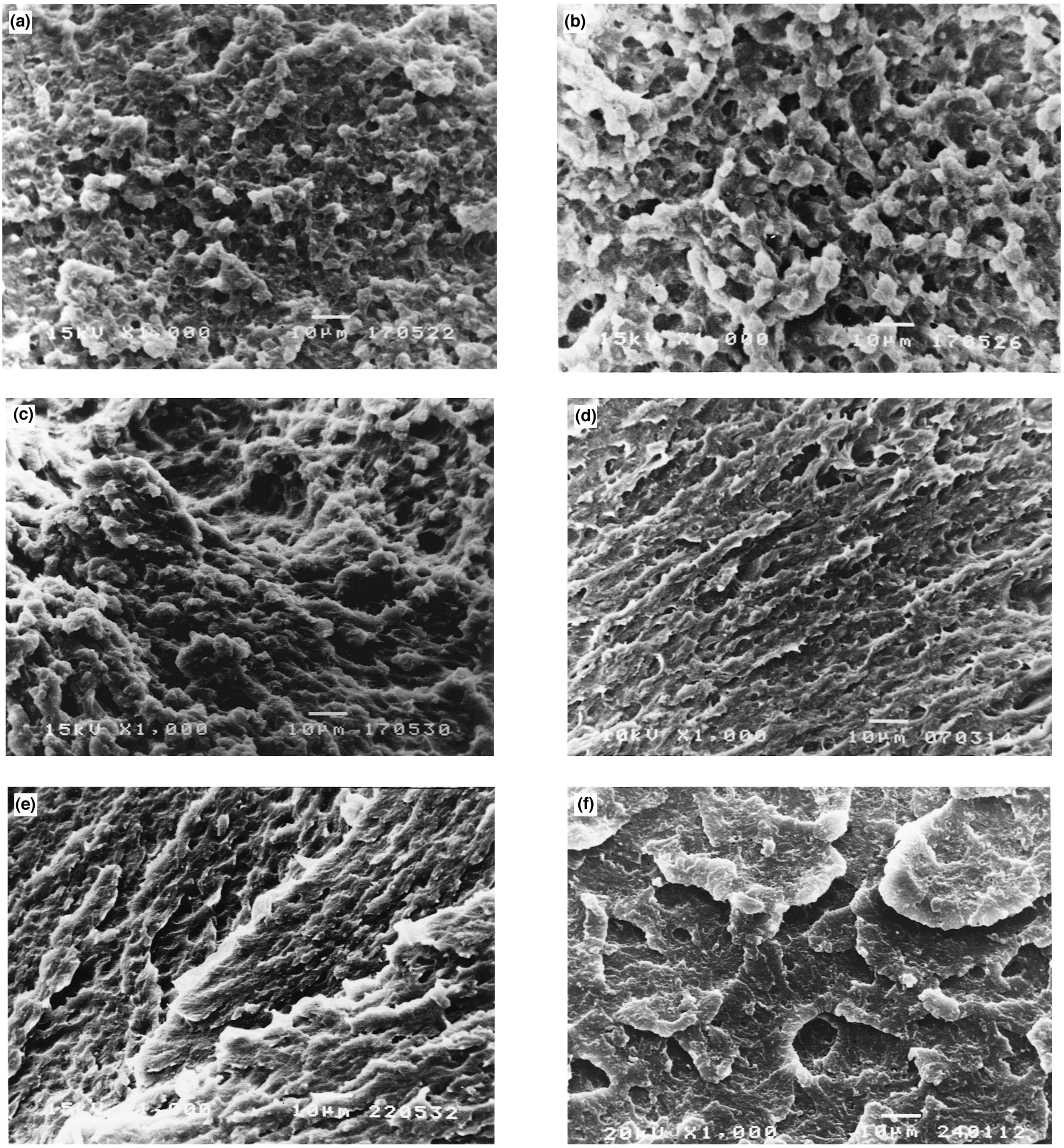

Figure 5 SEM micrographs of notched impact-fractured specimens of PC/MABS blends: (a) MP25 (×1000), (b) MP35 (×1000), (c) MP50 (×1000), (d) MP65 (×1000), (e) MP75 (×1000), (f) MP90 (×1000)

MABS lamellae. Such lamellar morphology is characteristic of better interfacial interaction and compatibilisation achieved through this reactive blending method.

There is good correspondence between the morphology of these materials and their impact behaviour. The notched Izod impact strength of the PC was 126.4 J/m and that of ABS was 320.3 J/m. The notched Izod impact performance for PC/ABS blends showed negative blending effect; it was lower than those of PC and ABS throughout whole composition range. The notched impact strength was 86.2 J/m for PC/ABS blend at 75 wt.% PC level. The notched impact strength value for MABS was almost the same as that of ABS. On the contrary, PC/MABS blends showed tremendous improvement in impact strength. The impact strength values increased progressively from 25 wt.% level of PC to 75 wt.% PC for PC/MABS blends. It was 882 J/m at 75 wt.% PC level for PC/MABS blends. But at the 90 wt.% level, this value reduced to 280.7 J/m. More details are given elsewhere[16]. Kim and Park[17] have demonstrated similar results of melt viscosity and

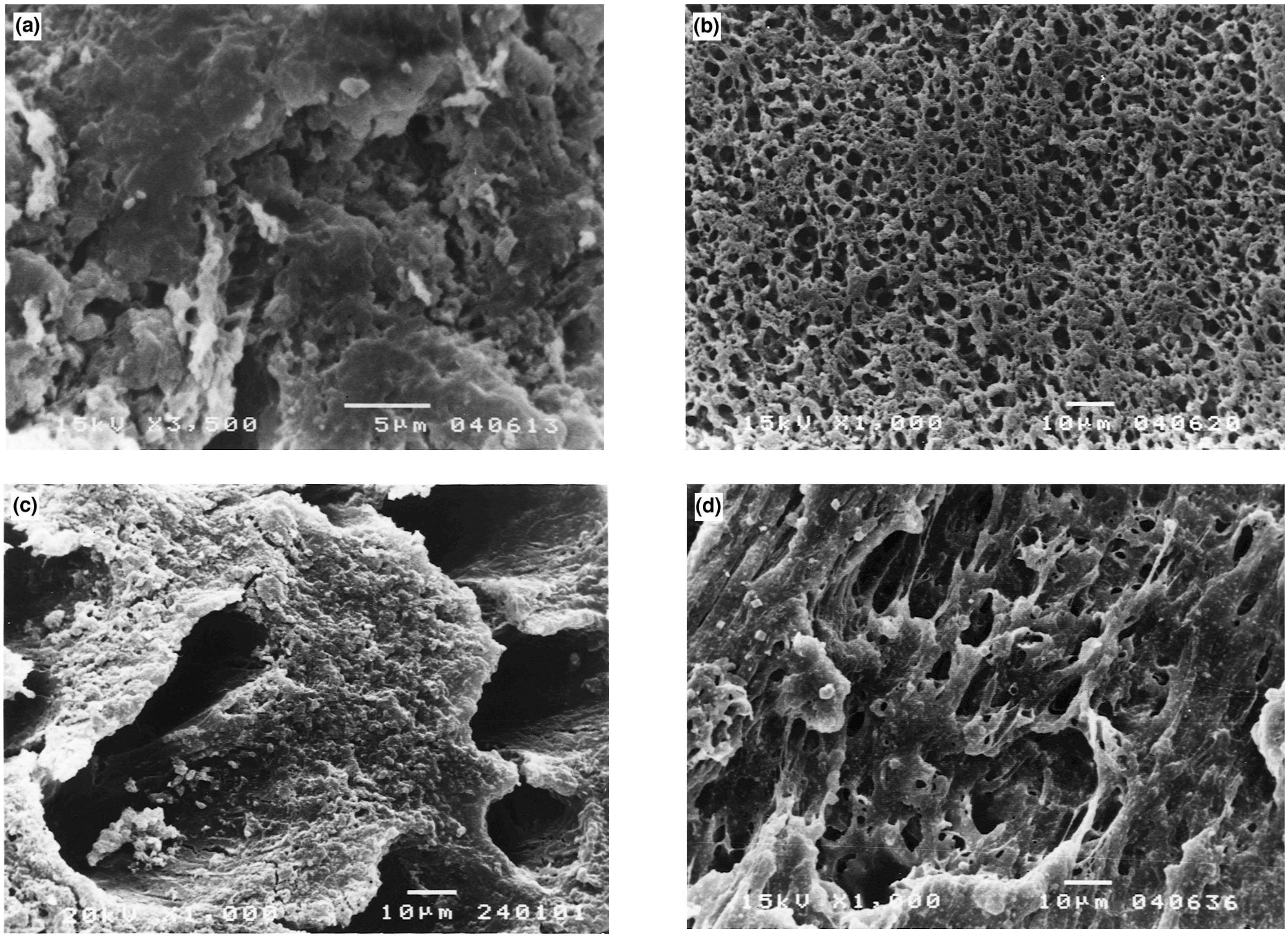

Figure 6 SEM micrographs of acid-etched surface of blends: (a) P65 (×3500), (b) MP65 (×1000). Alkaline-etched surface of blends: (c) P35 (×1000), (d) MP35 (×1000)

morphological characteristics for PC/polypropylene (PP) and PC/maleic anhydride grafted PP blends.

Morphological studies indicate the improved interfacial interaction in the PC/MABS blends. This improvement is attributed to the chemical reaction between the anhydride group of MABS and the terminal OH group of PC[11]. Such in situ formed compatibiliser stabilizes the dispersions to the fine texture during blending. Chen *et al.*[15] explained that the better interfacial interaction prevents the coalescence of the dispersed lamellae/particles and thus stabilizes to fine morphology during annealing at 200–230°C. In the PC/ABS blends, due to poor interfacial interaction, the dispersions coalesce into macro domains and the morphology is coarse irrespective of composition of PC.

CONCLUSIONS

Rheological and morphological behaviour of PC/ABS and PC/MABS blends were studied over the whole composition range. Addition of ABS or MABS to polycarbonate reduces the melt viscosity and thus improves processibility. Viscosity values of PC/ABS blends up to 65 wt.% PC are lower than those of ABS. Viscosity of PC/MABS blends are equal to or higher than that of MABS or ABS through the whole composition range. Microstructures of PC/ABS blends are coarse and irregular. Microstructures of PC/MABS blends are fine with lamellar morphology. The lamellae of dispersed phase in the PC/MABS blends are oriented along the flow direction of injection moulding. The notched impact strength values of PC/MABS blends are much higher than those of PC/ABS blends. Fractured surfaces of PC/ABS blends are rough and those of PC/MABS blends are smooth. The reactive blending process in PC/MABS blends improves interfacial interaction and thus generates fine morphology.

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