Miscibility Behavior of Blends of Poly(alkyl methacrylate)s with Poly(*p*-methylstyrene-*co*-methacrylonitrile)

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SYNOPSIS

The miscibility behavior of various poly(*p*-methylstyrene-*co*-methacrylonitrile) (pMSMAN)/poly(alkyl methacrylate)s blends was studied using differential scanning calorimetry. pMSMAN is miscible with poly(methyl methacrylate), poly(ethyl methacrylate), poly(*n*-propyl methacrylate), poly(isopropyl methacrylate), and poly(*n*-butyl methacrylate) over certain copolymer composition ranges, but is immiscible with poly(isobutyl methacrylate) and poly(*n*-amyl methacrylate). The width of the miscibility window decreases with increasing size of the pendant ester group of the poly(alkyl methacrylate), and is wider than that of the corresponding poly(*p*-methylstyrene-*co*-acrylonitrile) blend system. Various segmental interaction parameters are calculated using a binary interaction model. © 1995 John Wiley & Sons, Inc.

INTRODUCTION

The miscibility behavior of poly(alkyl methacrylate)s with copolymers of acrylonitrile has been extensively studied. Poly(methyl methacrylate) (PMMA), poly(ethyl methacrylate) (PEMA) and poly(*n*-propyl methacrylate) (PnPMA) are miscible with poly(styrene-*co*-acrylonitrile) (SAN),[1-4] poly(α-methylstyrene-*co*-acrylonitrile) (αMSAN)[5-7] and poly(*p*-methylstyrene-*co*-acrylonitrile) (pMSAN)[8] over certain copolymer composition ranges, showing "miscibility windows." The width of the miscibility window decreases with increasing size of pendant groups of the poly(alkyl methacrylate). Poly(isopropyl methacrylate) (PiPMA) is immiscible with SAN[2] and pMSAN.[8] PiPMA is immiscible with an αMSAN sample containing 30 wt % of acrylonitrile,[5] but the miscibility of PiPMA with αMSAN of other compositions has not been reported. Poly(*n*-butyl methacrylate) (PnBMA) is immiscible with SAN,[2] but it is miscible with αMSAN[7] and pMSAN[8] over very narrow copolymer composition ranges.

We have recently studied the miscibility behavior of poly(alkyl methacrylate)s with poly(styrene-*co*-methacrylonitrile) (SMAN).[9,10] PMMA, PEMA, PnPMA, and PiPMA are miscible with SMAN. The widths of the miscibility windows of SMAN blends are wider than those of the corresponding SAN blends. We now report the miscibility of poly (alkyl methacrylate)s with poly(*p*-methylstyrene-*co*-methacrylonitrile) (pMSMAN). It will be shown that PMMA, PEMA, PnPMA, PiPMA, and PnBMA are miscible with pMSMAN, and the widths of the miscibility windows are also wider than those of the corresponding pMSAN blends.

EXPERIMENTAL

Materials

Methacrylonitrile (MAN) (Fluka) was purified by passing through a column packed with hydroquinone remover (Scientific Polymer Products, Inc.). *p*-Methylstyrene (Tokyo Chemical Industry) was purified by fractional distillation at 45°C/10 mmHg. pMSMAN copolymers of varying compositions were prepared by free radical polymerization in 2-butanone at 80°C for 15 h, using 0.30 wt % azobisisobutyronitrile (AIBN) as an initiator. The copolymers were obtained by precipitation of the solutions in excess of hexane. They were then dried *in vacuo* at 65°C for a week. The MAN contents of the copolymers were determined by elemental analysis of

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the nitrogen. The copolymerization behavior of *p*-methylstyrene (pMS) with MAN and the microstructures of the copolymers were reported elsewhere.[11] PMMA, PEMA, PnPMA, PiPMA-1, PnBMA, and poly(*n*-amyl methacrylate) (PnAMA) were prepared by AIBN-initiated free radical polymerization in 2-butanone. Poly(isobutyl methacrylate) (PiBMA) and another PiPMA sample (designated PiPMA-2) were obtained from Scientific Polymer Products, Inc. The characteristics of the polymers are shown in Table I.

Preparation of Blends

The blends of each poly(alkyl methacrylate) with various pMSMAN in a weight ratio of 1 : 1 were prepared by casting from tetrahydrofuran (THF) solutions at room temperature. To examine a possible solvent effect on miscibility, PMMA/pMSMAN and PnBMA/pMSMAN blends were also prepared by precipitating from the THF solutions using excess methanol. The blend samples were dried *in vacuo*, first at 60°C for a week and then at 90°C for another week.

Characterization

The miscibility behavior of various blends was ascertained using the single glass transition temperature (T_g) criterion. T_gs of various blend samples were measured with a Perkin-Elmer DSC-4 differential scanning calorimeter at a heating rate of 20°C/min. Since the T_g values of pMSMAN, PMMA, and PiPMA are quite close to each other, the PMMA/pMSMAN and PiPMA/pMSMAN blends were subjected to an annealing process.[12] Each of the blend samples was first kept at 150°C for 5 min, and then annealed at 65°C for 2 weeks. Miscibility of the annealed samples was then ascertained using first-running DSC curves. The appearance of a single enthalpy recovery peak indicates miscibility. All clear films were examined for lower critical solution temperature (LCST) behavior following the method described previously.[13]

RESULTS

PMMA/pMSMAN Blends

Blends of PMMA with pMSMAN having ≤ 17.3 wt % of MAN and PMMA/pMSMAN 72.1 blend were hazy and two glass transitions were observed in each blend, indicating that PMMA is immiscible with these pMSMAN copolymers. The blends of PMMA with pMSMAN having MAN contents between 21.9 and 68.2 wt % were clear and each had a single glass transition, indicating miscibility in this copolymer composition range. Of these miscible blends, PMMA/pMSMAN 52.9, PMMA/pMSMAN 55.9, PMMA/pMSMAN 63.4, and PMMA/pMSMAN 68.2 blends showed LCST behavior while others remained transparent up to 300°C, the highest temperature allowed by the apparatus. The phase diagram of this system is shown in Figure 1. PMMA/pMSMAN blends obtained from the co-precipitation method have similar results to those prepared from the casting method. Therefore the miscibility window of this system is between 19 and 70 wt % of MAN in copolymers.

Table I Characteristics of Polymers

Polymer	T_g (°C)	M_n (kg/mol)	M_w/M_n	N**
pMSMAN4.4*	103	23.4	1.48	205
pMSMAN6.1	104	22.8	1.84	200
pMSMAN10.2	104	25.3	2.15	230
pMSMAN14.8	106	25.4	1.69	240
pMSMAN17.3	107	27.3	1.79	260
pMSMAN21.9	106	28.8	2.12	280
pMSMAN22.7	107	28.0	2.20	280
pMSMAN25.3	107	23.2	2.07	235
pMSMAN31.4	108	26.5	2.08	280
pMSMAN36.5	107	25.1	1.87	270
pMSMAN39.5	105	23.6	1.79	260
pMSMAN45.7	110	22.0	2.05	250
pMSMAN52.9	110	21.2	1.58	250
pMSMAN55.9	109	18.5	1.88	220
pMSMAN63.4	110	17.0	1.54	210
pMSMAN68.2	110	11.7	1.28	150
pMSMAN72.1	111	15.3	1.49	200
PMMA	119	63.6	1.70	640
PEMA	57	56.3	1.87	490
PnPMA	51	73.0	1.65	570
PiPMA-1	91	161.0	1.56	1260
PiPMA-2	82	41.6	1.80	325
PnBMA	36	61.0	1.98	430
PiBMA	56	144.5	1.68	1020
PnAMA	9	78.5	1.94	500

* Number after pMSMAN denotes weight percentage of MAN in the copolymer.
** Degree of polymerization.

PEMA/pMSMAN Blends

Blends of PEMA with pMSMAN having MAN contents ≤ 10.2 wt % and ≥ 55.9 wt % were cloudy and two T_gs were observed in each blend, indicating that

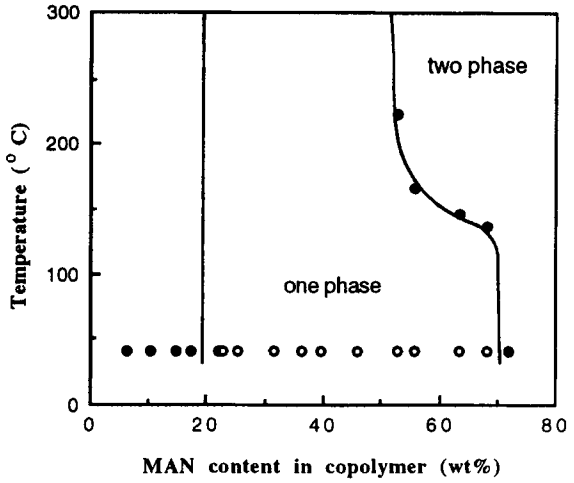

Figure 1 Phase diagram of PMMA/pMSMAN (1 : 1) blends: (O) miscible and (●) immiscible blends.

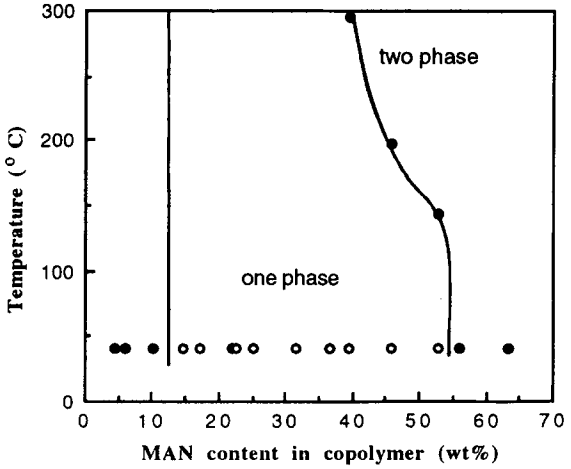

Figure 2 Phase diagram of PEMA/pMSMAN (1 : 1) blends: (O) miscible and (●) immiscible blends.

PEMA is immiscible with these pMSMAN samples. Blends of PEMA with pMSMAN having MAN contents between 14.8 and 52.9 wt % were transparent at room temperature and one glass transition was observed in each blend, indicating miscibility in this copolymer composition range. For the miscible blends, PEMA/pMSMAN 39.5, PEMA/pMSMAN 45.7, and PEMA/pMSMAN 52.9 blends showed LCST behavior and others remained clear up to 300°C. The phase diagram of PEMA/pMSMAN blends is shown in Figure 2. The miscibility range of this blend system is 12 to 55 wt % of MAN.

PnPMA/pMSMAN Blends

Blends of PnPMA with pMSMAN having MAN contents ≤ 10.2 wt % and ≥ 45.7 wt % were cloudy and two T_gs were observed in each blend, indicating that PnPMA is immiscible with these pMSMAN copolymers. Blends of PnPMA with pMSMAN having MAN contents between 14.8 and 39.5 wt % were transparent at room temperature and only one glass transition was observed in each blend, indicating miscibility. For the miscible blends, the blends of PnPMA/pMSMAN 39.5 and PnPMA/pMSMAN 36.5 showed LCST behavior and others were clear up to 300°C. The phase diagram of PnPMA/pMSMAN blends is shown in Figure 3. The miscibility range of this blend system is between 12 and 43 wt % of MAN.

PiPMA/pMSMAN Blends

The optical clarity of PiPMA/pMSMAN blends may not be taken as an indication of miscibility because of the matching in refractive indices[14] of PiPMA and several pMSMAN copolymers. The LCST behavior of the blends was studied using the isothermal annealing technique.[10] Each blend was kept at an elevated temperature for 5 min followed by annealing at 65°C for 2 weeks. The glass transition behavior of the annealed blend was then examined. For PiPMA-1/pMSMAN 21.9, PiPMA-1/pMSMAN 22.7, PiPMA-1/pMSMAN 25.3, and PiPMA-1/pMSMAN 31.4 blends, each had a single glass transition when they were treated at 150°C, while other blends showed two T_gs. At 220°C, all the PiPMA-1/pMSMAN blends separated to two phases. The miscibility range of PiPMA-1 blend system is from 19 to 34 wt % of MAN. For PiPMA-2/pMSAN 17.3, PiPMA-1/pMSMAN 21.9, PiPMA-2/pMSMAN 22.7, PiPMA-2/pMSMAN 25.3, and

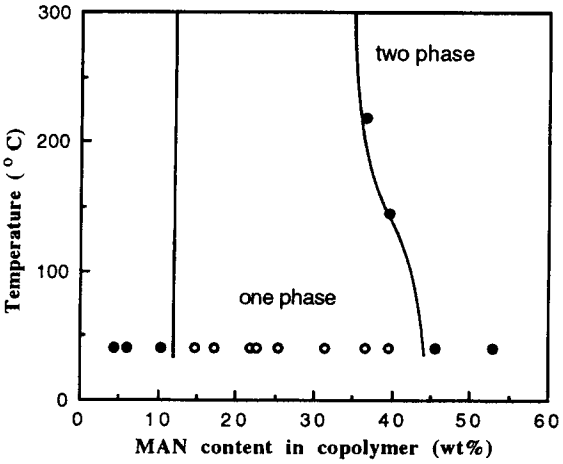

Figure 3 Phase diagram of PnPMA/pMSMAN (1 : 1) blends: (O) miscible and (●) immiscible blends.

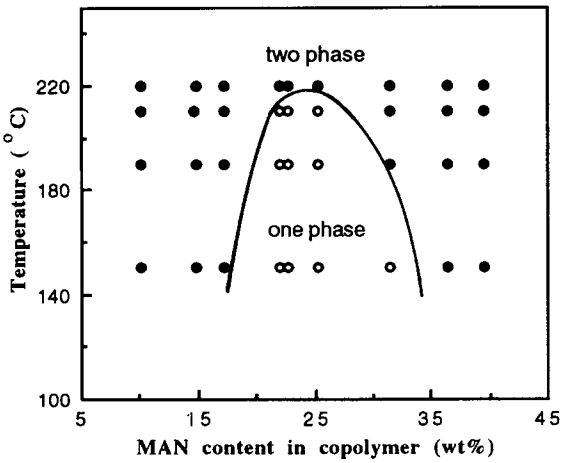

Figure 4 Phase diagram of PiPMA-1/pMSMAN (1 : 1) blends: (○) miscible and (●) immiscible blends.

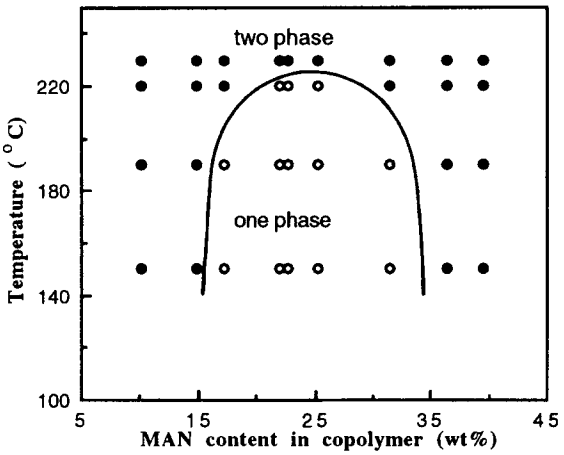

Figure 5 Phase diagram of PiPMA-2/pMSMAN (1 : 1) blends: (○) miscible and (●) immiscible blends.

PiPMA-2/pMSMAN 31.4 blends, each had a single glass transition when they were treated at 150°C, and others showed two T_gs. The miscibility range of PiPMA-2 blend system is from 16 to 34 wt % of MAN. At 230°C, all the PiPMA-2/pMSMAN blends separated to two phases. Figures 4 and 5 show the phase diagrams of PiPMA/pMSMAN blends. Blends of PiPMA with a lower molecular weight have a slightly wider miscibility window and higher LCSTs.

PnBMA/pMSMAN Blends

For the PnBMA/pMSMAN blend system, blends of PnBMA with pMSMAN having MAN contents $\leq$ 10.2 wt % and $\geq$ 31.4 wt % were cloudy and two T_gs were observed in each blend, indicating immiscibility. Blends of PnBMA with pMSMAN having MAN contents between 14.8 and 25.3 wt % were clear and only one glass transition was observed in each blend, indicating miscibility in this range. All the miscible PnBMA/pMSMAN blends showed LCST behavior. The phase diagram is shown in Figure 6. The miscibility range of this blend system is from 12 to 28 wt % of MAN. Blends prepared by co-precipitation method also showed the same miscibility range.

PiBMA/pMSMAN and PnAMA/pMSMAN Blends

All the PiBMA/pMSMAN and PnAMA/pMSMAN blends were cloudy. Each of the blends had two glass transitions, indicating that pMSMAN is immiscible with PiBMA and PnAMA.

DISCUSSION

The miscibility behavior of various poly(alkyl methacrylate)s with pMSMAN copolymers is summarized in Table II. The width of miscibility range of these blend systems decreases with increasing size of the pendant ester group of alkyl methacrylate. Similar trends were also observed for blend systems of poly(alkyl methacrylate)s with SAN,[1-4] αMSAN,[5-7] pMSAN,[8] and SMAN.[9,10] For comparison purposes, the corresponding blend systems with SMAN and pMSAN are also listed in Table II. In contrast with SMAN, pMSMAN is miscible with PnBMA when the MAN content in the copolymer is in the range of 12–28 wt %. pMSMAN systems also show wider miscibility windows compared to the pMSAN systems.

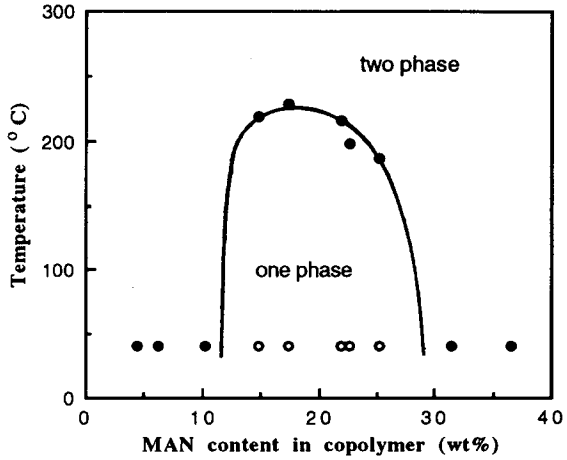

Figure 6 Phase diagram of PnBMA/pMSMAN (1 : 1) blends: (○) miscible and (●) immiscible blends.

Table II Miscibility Ranges of Binary Blends

	pMSMAN	SMAN[9,10]	pMSAN[8]
PMMA	19–70 wt % MAN	8–63 wt % MAN	12–34 wt % AN
PEMA	12–55	4.5–44	12–33
PnPMA	12–43	2–32	6–28
PiPMA-1	19–34	12–38	immiscible
PiPMA-2	16–34	8–43	
PnBMA	12–28	immiscible	12–14
PiBMA	immiscible	immiscible	not studied
PnAMA	immiscible	not studied	not studied

Segmental Interaction Parameters

For a homopolymer A/copolymer BC blend, the miscibility depends on both intramolecular and intermolecular interactions between various segments. Based on a binary interaction model,[15-17] the net interaction parameter of a blend system (χ_{blend}) is related to various segmental interaction parameters by the equation:

$$\chi_{blend} = y\chi_{AB} + (1 - y)\chi_{AC} - y(1 - y)\chi_{BC} \quad (1)$$

where y is the volume fraction of segment B in the copolymer. For pMSMAN/poly(alkyl methacrylate) blend systems, χ_{blend} is expressed by the following equation:

$$\chi_{blend} = y\chi_{MA/MAN} + (1 - y)\chi_{MA/MS} - y(1 - y)\chi_{MS/MAN} \quad (2)$$

where y represents the volume fraction of MAN in pMSMAN and subscripts MA, MAN, and MS denote alkyl methacrylate, methacrylonitrile, and *p*-methylstyrene, respectively. At the phase boundary χ_{blend} equals χ_{crit}, which is given by the following equation

$$\chi_{crit} = 0.5(N_1^{-1/2} + N_2^{-1/2})^2 \quad (3)$$

where N_1 and N_2 are the degrees of polymerization for homopolymer and copolymer, respectively. The homopolymer would be miscible with the copolymers if $\chi_{blend} < \chi_{crit}$. In the present work, an N value of 240 for pMSMAN was used to calculate χ_{crit}. For the PMMA/pMSMAN system χ_{crit} was 0.0054, based on the N values of 640 and 240 for PMMA and pMSMAN, respectively. The miscibility range was considered as 19 and 70 wt % of MAN, corresponding to y values of 0.185 and 0.685. Using the value of 0.19 for $\chi_{MMA/MAN}$,[9] $\chi_{MMA/MS}$ and $\chi_{MS/MAN}$ are calculated to be 0.096 and 0.71, respectively. Based on the miscibility range listed in Table II, various segmental interaction parameters were then calculated using the value of 0.71 for $\chi_{MS/MAN}$. The results are summarized in Table III, together with the values calculated from the corresponding SMAN and pMSAN systems. In general, the values of $\chi_{MA/MAN}$ (between MAN and various alkyl methacrylates) calculated from the pMSMAN systems agree well with those calculated from the SMAN systems, except for the systems with PiPMA. The values of $\chi_{MA/MS}$ (between MS and various MA) calculated from the pMSMAN systems are, however, all slightly larger than the corresponding values calculated from the pMSAN systems. We believe the χ values obtained from the MAN copolymer systems are more reliable. Commercial PMMA and PEMA samples were used in our previous study on the pMSAN systems.[8] The presence of a small amount of acrylate comonomer in these samples could affect the miscibility behavior and the χ values obtained from the phase boundaries. It is interesting to note that $\chi_{MA/MAN}$ and $\chi_{MA/MS}$ change with the size of pendant groups of poly(alkyl methacrylate)s in different ways. The value of $\chi_{MA/MAN}$ increases almost linearly with increasing size of the pendant group, indicating an increasing repulsive interaction between MAN and MA when MA changes from methyl to *n*-butyl. The value of $\chi_{MA/MS}$, on the other hand, decreases rather gradually with the pendant group size. The relationships between the χ values and the number of carbon atoms in the linear pendant ester group are shown in Figure 7. They can be fitted by the following equations:

$$\chi_{MA/MAN} = 0.109 + 0.089n \quad (4)$$

$$\chi_{MA/MS} = 0.125 \exp(-0.37n) \quad (5)$$

where n is the number of carbon atoms of the linear pendant ester group in the polymethacrylate. Using

Table III Segmental Interaction Parameters for Some Blend Systems

	pMSMAN System	SMAN System	pMSAN System
PMMA	$\chi_{MMA/MAN} = 0.19$	$\chi_{MMA/MAN} = 0.19$	$\chi_{MMA/AN} = 0.56$
	$\chi_{MMA/MS} = 0.096$	$\chi_{MMA/St} = 0.03$	$\chi_{MMA/MS} = 0.037$
	$\chi_{MS/MAN} = 0.71$	$\chi_{St/MAN} = 0.52$	$\chi_{MS/AN} = 0.91$
PEMA	$\chi_{EMA/MAN} = 0.30$	$\chi_{EMA/MAN} = 0.29$	$\chi_{EMA/AN} = 0.56$
	$\chi_{EMA/MS} = 0.051$	$\chi_{EMA/St} = 0.016$	$\chi_{EMA/MS} = 0.036$
PnPMA	$\chi_{nPMA/MAN} = 0.37$	$\chi_{nPMA/MAN} = 0.35$	$\chi_{nPMA/AN} = 0.64$
	$\chi_{nPMA/MS} = 0.041$	$\chi_{nPMA/St} = 0.011$	$\chi_{nPMA/MS} = 0.018$
PiPMA-1	$\chi_{iPMA/MAN} = 0.39$	$\chi_{iPMA/MAN} = 0.29$	
	$\chi_{iPMA/MS} = 0.048$	$\chi_{iPMA/St} = 0.027$	
PiPMA-2	$\chi_{iPMA/MAN} = 0.41$	$\chi_{iPMA/MAN} = 0.29$	
	$\chi_{iPMA/MS} = 0.045$	$\chi_{iPMA/St} = 0.025$	
PnBMA	$\chi_{nBMA/MAN} = 0.46$		$\chi_{nBMA/AN} = 0.70$
	$\chi_{nBMA/MS} = 0.030$		$\chi_{nBMA/MS} = 0.019$

these two equations, we obtain the values of 0.56 and 0.020, respectively, for $\chi_{nAMA/MAN}$ and $\chi_{nAMA/MS}$. Together with the $\chi_{MS/MAN}$ value of 0.71, χ_{blend} for the PnAMA/pMSMAN system is positive at all y values and larger than χ_{crit}. The PnAMA/pMSMAN system is then expected to be an immiscible one, which agrees with our experimental results.

Coleman et al.[18,19] have recently suggested that the miscibility range in homopolymer/copolymer blends can be predicted by a non-hydrogen-bonded solubility parameter (δ) approach. Since the δ value of a copolymer varies with its composition, there will be a range of copolymer compositions over which the δ values of those copolymers are closely matched to that of the homopolymer, thus favoring miscibility. We have earlier applied the δ approach to predict the miscibility behavior of various poly(alkyl methacrylate)s with SMAN copolymers[9,10] and found fairly good agreement between the predicted and the experimental results. The δ values of pMSMAN copolymers are between 19.2 and 24.3 $(J/cm^3)^{1/2}$ and those of PMMA, PEMA, PnPMA, PiPMA, PnBMA, PiBMA, and PnAMA are 18.4, 18.2, 17.9, 17.4, 17.8, 17.3, and 17.6 $(J/cm^3)^{1/2}$, respectively. Using the computer program from Coleman et al. and assuming a moderate specific interaction (as for the SMAN systems), the miscibility ranges for various poly(alkyl methacrylate)/pMSMAN systems are predicted and summarized in Table IV. Except for the PnAMA system, the predictions here again agree generally well with the experimental results.

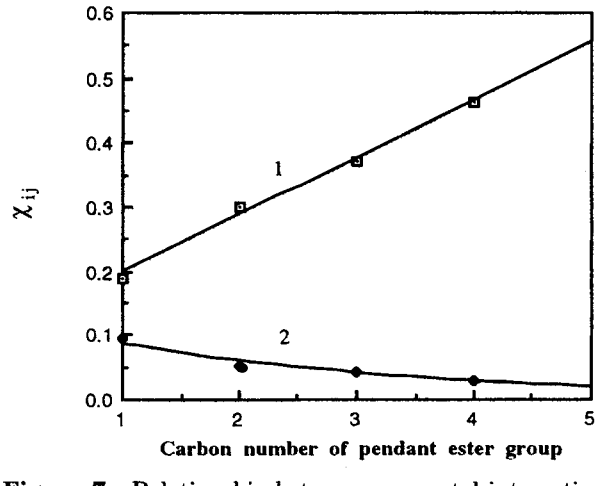

Figure 7 Relationship between segmental interaction parameters and carbon number of pendant group: (1) $\chi_{MA/MAN}$ and (2) $\chi_{MA/MS}$.

David and Sincock[20] used the individual solubility parameter components—dispersive (δ_d), polar (δ_p), and hydrogen-bonding (δ_h)—instead of the conventional total solubility parameter to estimate polymer blend miscibility. They introduced the concept of "miscibility parameter" which considered any two or all three individual solubility parameter components.

Cowie, Watson, and McEwen[21] also recently re-

Table IV Miscibility Ranges of pMSMAN Blends Predicted by δ Approach

	Predicted	Observed
PMMA	0–63 wt % of MAN	19–70 wt % of MAN
PEMA	2–54	12–55
PnPMA	3–47	12–43
PiPMA	10–30	19–34; 16–34
PnBMA	5–43	12–28
PiBMA	immiscible	immiscible
PnAMA	8–37	immiscible

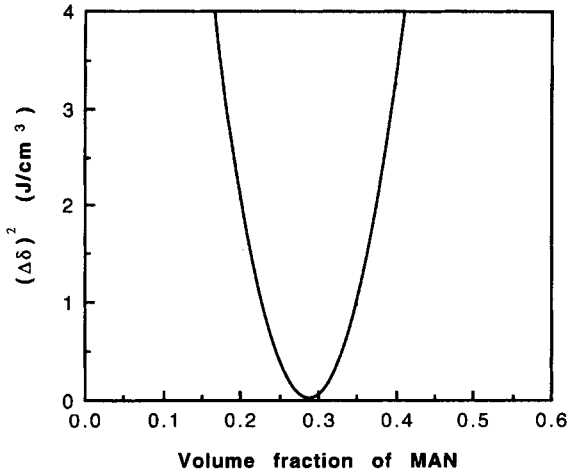

Figure 8 Variation of $(\Delta\delta)^2$ as a function of MAN content for PMMA/pMSMAN blends.

ported the miscibility of PMMA with SMAN and suggested that δ_p but not δ_d and δ_h played an important role in miscibility. Here we also analyze our systems in terms of the matching of the δ_p values of poly(alkyl methacrylate)s and pMSMAN. The δ_p values for PMMA, PEMA, PnPMA, PiPMA, PnBMA, PMAN, and PpMS are 5.66, 4.79, 4.13, 4.08, 3.57, 17.21, and 0.97 $(J/cm^3)^{1/2}$, respectively.[14] The δ_p values of pMSMAN are calculated using the equation:

$$\delta_{p(\text{pMSMAN})} = (1 - y)\delta_{p(\text{pMS})} + y\delta_{p(\text{MAN})} \qquad (6)$$

where y is the volume fraction of MAN in the copolymer. Figure 8 shows the square of the difference of the δ_p values $(\Delta\delta_p)^2$ as a function of copolymer compositions for PMMA/pMSMAN system. $(\Delta\delta_p)^2$ becomes zero at a MAN volume fraction of about 0.29, which is inside the experimentally observed miscibility range. If we arbitrarily take $(\Delta\delta_p)^2 < 1$ J/cm^3 as the criterion of miscibility, the expected miscibility range is then between $y = 0.21$ and $y = 0.35$, which is narrower than our observed miscibility range. The predicted miscibility ranges based on $(\Delta\delta_p)^2$ for other poly(alkyl methacrylate)/pMSMAN blends move toward the low MAN-content end as the alkyl group changes from methyl to *n*-butyl, and the miscibility ranges are also very narrow as compared with our experimental findings.

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