Available online at www.sciencedirect.com

SCIENCE DIRECT®

Polymer 44 (2003) 5285–5291

polymer

www.elsevier.com/locate/polymer

Miscibility and interactions in blends and complexes of poly(4-methyl-5-vinylthiazole) with proton-donating polymers

X.F. Luo[a], X. Hu[a,*], X.Y Zhao[a], S.H. Goh[b], X.D. Li[b]

[a]School of Materials Engineering, Nanyang Technological University, Nanyang Avenue, Singapore, Singapore 639798
[b]Department of Chemistry, University of Singapore, 3 Science Drive, Singapore, Singapore 117543

Received 10 February 2003; received in revised form 3 June 2003; accepted 26 June 2003

Abstract

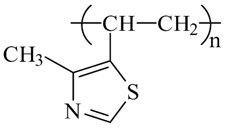
The miscibility and interactions in blends and complexes of poly(4-methyl-5-vinylthiazole) (PMVT) with poly(*p*-vinylphenol) (PVPh), poly(acrylic acid) (PAA) and poly(vinylphosphonic acid) (PVPA) were studied. PMVT formed complexes with PVPA but not with PVPh and PAA. Each of the blends of PMVT with PVPh and PAA showed a single glass transition temperature (T_g), indicating miscibility. Fourier-transform infrared spectroscopic and X-ray photoelectron spectroscopic studies provided the existence of interactions in the PMVT blends and complexes. The XPS studies indicated that the thiazole nitrogen atoms are involved in hydrogen-bonding interactions with PVPh and PAA, and ionic interactions with PVPA. The sulfur atoms of PMVT also interact with PVPh, PAA and PVPA.

Keywords: Poly(4-methyl-5-vinylthiazole); Miscibility; Interactions

1. Introduction

For the formation of a miscible blend from two dissimilar polymers, the two polymers must possess suitable interacting sites to achieve specific interactions. In many cases, a polymer or especially a copolymer contains different types of interaction sites. Therefore, different interactions may exist simultaneously in a polymer blend or complex [1–6]. The existence of specific interactions in miscible polymer blends can be detected by spectroscopic techniques such as Fourier-transform infrared (FTIR) spectroscopy [7,8] and X-ray photoelectron spectroscopy (XPS) [9–13].

Polymers containing basic groups such as pyridine, piperidine and imidazole are miscible with proton-donating polymers through hydrogen-bonding interactions or ionic interactions. For example, poly(4-vinylpyridine) (P4VPy), poly(*N*-methyl-4-piperidyl methacrylate) (PMPMA) and poly(1-vinylimidazole) (PVI) form miscible blends or even complexes with a larger number of proton-donating polymers such as poly(*p*-vinylphenol) (PVPh) [1,2,14–17], poly(acrylic acid) (PAA) [2,17–19], and poly(styrenesulfonic acid) (PSSA) [2,20,21]. However, the miscibility and complexation behavior of polymers containing basic group such as thiazole have not been studied. Thiazole (p*K*b = 11.48) is a weaker base than imidazole ($pK_b = 7.05$) and pyridine ($pK_b = 8.75$). It is envisaged that poly(4-methyl-5-vinylthiazole) (PMVT), a polymer containing thiazole group, may interact less strongly with proton-donating polymers than P4VPy and PVI do. PMVT has been used as a stationary-phase for the special selectivity of phenolic derivatives by reversed-phase liquid-chromatography [22].



Poly(4-methyl-5-vinylthiazole) (PMVT)

Miscible blends involving thioether sulfur-containing polymers have been studied [23]. Recently, Goh et al. [24] reported the existence of the sulfur atoms in the hydrogen-bonding interactions in the poly(*N*-acryloythiomorpholine)/PVPh miscible blends by XPS.

There are two possible interacting sites in each segment of PMVT, the nitrogen atom and thioether sulfur atom. Therefore, it is of interest to study whether one or both

---

[*] Corresponding author. Tel.: +65-6790-4610; fax: +65-6790-9081. *E-mail address:* asxhu@ntu.edu.sg (X. Hu).

0032-3861/03/$ - see front matter © 2003 Published by Elsevier Ltd.
doi:10.1016/S0032-3861(03)00578-0

groups are involved in the interactions with proton-donating polymers. In this paper, we report the miscibility and interpolymer complexation of PMVT with three proton-donating polymers: PVPh, PAA and PVPA. The nature of interactions between PMVT and the three proton-donating polymers are investigated by FTIR and XPS.

2. Experimental section

2.1. Materials

4-Methyl-5-vinylthiazole, PVPh ($M_w = 22,000$, $T_g = 154$ °C) and PAA ($M_v = 450,000$, $T_g = 127$ °C) were supplied by Aldrich Chemical Co. Inc. PVPA (molecular weight information not available, $T_g = 165$ °C) was obtained from Polyscience, Inc.

2.2. Synthesis of PMVT

4-Methyl-5-vinylthiazole was distilled at 66–67 °C/10 mmHg. PMVT was prepared by solution polymerization in toluene at 70 °C for 24 h. Monomer (20 ml) was dissolved in 50 ml toluene and initiated by 0.04 g of 2,2′-azobis(isobutyronitrile). The resulting polymer was purified by precipitation twice with hexane from toluene solution and dried in vacuo at 70–80 °C for 2 days. The molecular weights ($M_w = 11,000$, $M_n = 7000$) of PMVT relative to polystyrene standards were measured by gel permeation chromatography. T_g : 94 °C. The polymer shows an initial decomposition temperature of 325 °C, as shown by thermogravimetry. Partially protonated PMVT (denoted as pPMVT) was prepared by mixing water/ethanol (1:1) solutions of PMVT and HCl (3:2 mole ratio). Initial removal of solvent was done on a hot plane. pPMVT were further dried in vacuo at 60 °C for at least two weeks and stored in a desiccator to prevent absorption of moisture.

2.3. Preparation of complexes

Polymer complexes were prepared by mixing appropriate amounts of ethanol/water (1:1 volume ratio) solutions (1% w/v) of PMVT with PVPA. Precipitates formed immediately upon mixing of the two solutions. The mixture was stirred continuously for 1 h. The precipitates were then separated by centrifugation, washed with the solvent, and dried in vacuo at 90 °C for two weeks. The dried complexes were ground to fine powder and then stored in a desiccator. The bulk compositions of the complexes were determined by nitrogen analysis using a Perkin–Elmer 2400 elemental analyzer.

2.4. Preparation of blends

Polymer blends of PMVT with PVPh were obtained by solution casting from ethanol. The mixing of ethanol/water (1:1 volume ratio) solutions of PMVT and PAA did not produce precipitation. PMVT/PAA blends of various compositions were obtained by solution casting from their ethanol/water (1:1) solutions. Initial removal of solvent was done on a hot plane. The blends were further dried in vacuo at 90 °C for at least two weeks.

2.5. Differential scanning calorimetric measurements (DSC)

The glass transition temperatures (T_gs) of various samples were measured with a TA Instruments 2010 differential scanning calorimeter (DSC). The scanning rate was 20 °C $\min^{-1}$. The initial onset of the change of slope in the DSC curve was taken as the T_g.

2.6. FTIR measurements

Infrared spectra were recorded on a Bio-Rad 165 FTIR spectrophotometer; 32 scans were signal-averaged with a resolution of 2 cm^{-1}. Samples were prepared by dispersing the complexes in KBr and compressing the mixture to form discs. Spectra were acquired at 140 °C to exclude moisture, using a SPECAC high-temperature cell.

2.7. XPS measurements

XPS measurements were carried out on a VG Scientific ESCALAB MkII spectrometer equipped with a Mg K_α X-ray source (1235.6 eV photons) and a hemispherical energy analyzer. The blend or complex was mounted on a standard sample stud by means of a double-sided adhesive tape. The analyzer pass energy was set at 20 eV and several scans of the C 1s, N 1s, O 1s and S 2p spectra were obtained. All core-level spectra were referenced to the C 1s neutral carbon peak at the binding energy (BE) of 284.6 eV.

3. Results and discussion

3.1. Miscibility and complexation behavior

The mixing of the ethanol solutions of PMVT with PVPh led to the formation of a cloudy solution. However, the mixing of the ethanol solutions of P4VPy or PVI and PVPh led to instantaneous precipitation [15–17]. It is likely that the interaction between PMVT and PVPh is not as strong as that between PVPh and P4VPy or PVI.

To ascertain the miscibility of the blends, the T_gs of various blends were measured by DSC. Each blend was found to exhibit a single composition-dependent T_g, indicating miscibility. The T_g-composition curve of the blends is shown in Fig. 1. In all cases, the T_g value of the blend is larger than that calculated by a linear additivity rule. Similar positive deviation of T_g value was also observed for other blends such as P4VPy/PVPh and PVI/

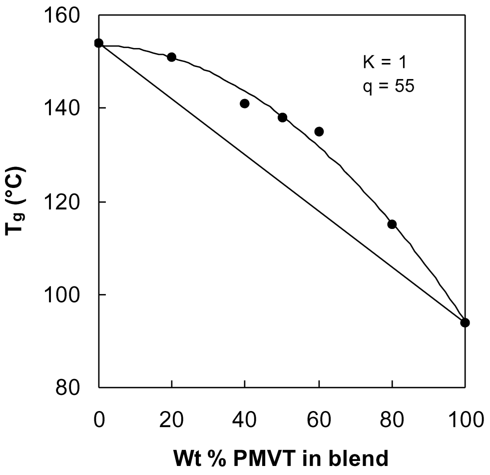


Fig. 1. T_g-composition curve of PMVT/PVPh blends.

PVPh systems [15–17]. The T_g-composition curve can be fitted by the Kwei equation

$$T_g(\text{blend}) = [(w_1T_{g1} + kw_2T_{g2})/(w_1 + kw_2)] + qw_1w_2$$

where k and q are fitting constants. The curve in Fig. 1 was drawn using k and q values of 1 and 55, respectively. In comparison, the T_g-composition curves of P4VPy/PVPh and PVI/PVPh blends can also be fitted by Kwei equation with q values of 200 and 107, respectively [15–17]. Kwei pointed out that the quadratic term qw_1w_2 is proportional to the number of specific interactions in the blends [25]. Therefore, the smallest q value suggests a weakest interaction between PMVT and PVPh.

PMVT formed complexes with PVPA but not with PAA at all feed compositions. These results suggest that intermolecular interactions in the complexes of PMVT/PVPA are stronger than those in the PMVT/PAA blends. Moreover, as PAA forms complexes with P4VPy and PVI [17,26], these results also indicate that P4VPy and PVI interact more strongly with PAA than PMVT does. The characteristics of the PMVT/PVPA complexes are shown in Table 1. As the PMVT content in the feed increases, the yield of the complex decreases.

As shown in Table 2, the T_g values of the PMVT/PAA are higher than those of the component polymers. Such a positive deviation of T_g values indicates strong interactions in the blends. No distinct T_g's could be detected for the PMVT/PVPA complexes. Similarly, the T_g's of PMPMA/PVPA and P4VPy/PVPA complexes could not be observed [2,27].

Table 1
Characteristics of PMVT/PVPA complexes

Complex sample number	1	2	3
Mole fraction of PMVT in feed	0.3	0.5	0.7
Mole fraction of PMVT in the bulk of complex	0.38	0.45	0.49
Yield (%)	64	53	30
N 1s BE (eV)	398.8, 400.9	398.8, 401.0	398.8, 400.8
Fraction of high-BE N 1s peak	0.63	0.22	0.20

Table 2
Characteristics of PMVT/PAA blends

Blend sample number	1	2	3
Mole fraction of PMVT in feed	0.3	0.5	0.7
T_g (°C)	158	154	145
N 1s BE (eV)	398.8, 399.8	398.8, 399.9	398.8, 399.9
Fraction of high-BE N 1s peak	0.68	0.29	0.21

3.2. FTIR characterization

Fig. 2 shows the infrared spectra of PVPh and PMVT/PVPh blends in the hydroxyl region at 140 °C. Pure PVPh (curve A) shows two distinct bands, a sharp band at 3531 cm^{-1} for the absorption of free hydroxyl groups and a broad band centered at 3369 cm^{-1} for the absorption of hydrogen bonded hydroxyl groups (self-association). As the PMVT content in the blend increases, the intensity of the free hydroxyl band at 3531 cm^{-1} decreases gradually and cannot be observed when the PMVT content in the blend reaches 80 wt%, indicating that more free hydroxyl groups are hydrogen bonded with thiazole groups as the concentration of thiazole groups is increased. On the other hand, the center of the hydrogen bonded hydroxyl band shifts from 3369 to 3255 cm^{-1}. Such a low-frequency shift indicates that the intermolecular hydrogen-bonding interaction between PMVT and PVPh is stronger than the self-association of PVPh. Miscible blends of PVPh with P4VPy and PVI also show similar low-frequency shifts of the hydroxyl bands. Such a frequency shift can be regarded as a measure of the average strength of the intermolecular interactions [28]. In this respect, the average strength of the hydrogen-bonding interactions in the blends of PVPh with

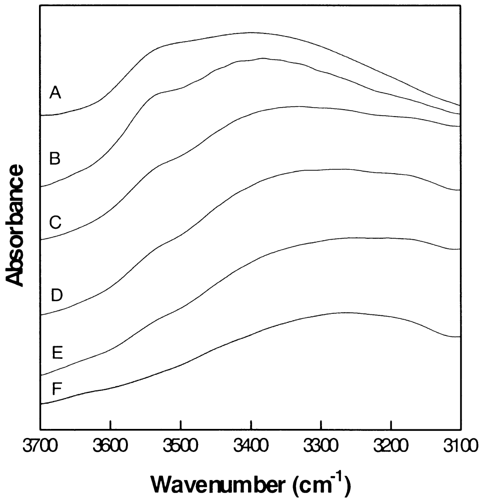


Fig. 2. FTIR spectra, recorded at 140 °C, of the hydroxyl region of PMVT/PVPh miscible blends: (A) 0, (B) 20, (C) 40, (D) 50, (E) 60 and (F) 80 wt% PMVT.

PMVT, P4VPy and PVI are in the order of PVI/PVPh ($\Delta\nu = 421\ \text{cm}^{-1}$) [16,17] >P4VPy/PVPh ($\Delta\nu = 400 \times \text{cm}^{-1}$) [15] >PMVT/PVPh ($\Delta\nu = 276\ \text{cm}^{-1}$). The result is in agreement with that from thermal analysis. Thiazole ($pK_b = 11.48$) is a weaker base than imidazole ($pK_b = 7.05$) and pyridine ($pK_b = 8.75$). Therefore, PMVT interacts less strongly with PVPh by hydrogen-bonding interaction than P4VPy and PVI do.

Fig. 3 shows the spectra in the carbonyl region of PAA and PMVT/PAA blends. PAA exhibits a rather broad carbonyl stretching band around 1716 cm^{-1} comprising two overlapping carbonyl stretching modes of the free and self-associated carboxylic acid groups in PAA. The latter is at a lower wavenumber than the former. However, for PMVT/PAA blends, the carbonyl stretching band narrows gradually as the PMVT content in blends increases. Such a change can be attributed to the interactions between thiazole groups and carboxylic acid groups. When interactions between thiazole groups and carboxylic groups occur, the intensity of carbonyl stretching band from self-associated carboxylic acid becomes decreases, leading the carbonyl stretching bands of PAA shift to high frequency.

The ionic interactions in the PMPMA/PAA and PVI/PAA complexes have been investigated by FTIR [2,17]. The transfer of protons from carboxylic groups of PAA to the nitrogen atoms of PMPMA or PVI leads to the appearance of asymmetric vibration signals of carboxylate anions that are situated around 1580 cm^{-1}. However, there is no new band developed at 1580 cm^{-1} in the PMVT/PAA blends, and therefore the thiazole nitrogen atoms are not protonated.

Evidence for interactions between the PMVT and PVPA is provided by FTIR spectroscopy. The FTIR studies focus on the P–OH deformation bands of PVPA in the complexes. The P–OH deformation band appears in the range of 864 to 1014 cm^{-1}, depending upon the structures of the compounds [29]. As shown in Fig. 4 (curve D), the 1014 cm^{-1} band of PVPA, which does not occur in the FTIR spectrum of PMVT, could be attributed to the P–OH deformation mode of PVPA. In all the PMVT/PVPA complexes, however, the P–OH band shifts to 995 cm^{-1}, suggesting changes of the deformation mode induced by complexation between PVPA and PMVT.

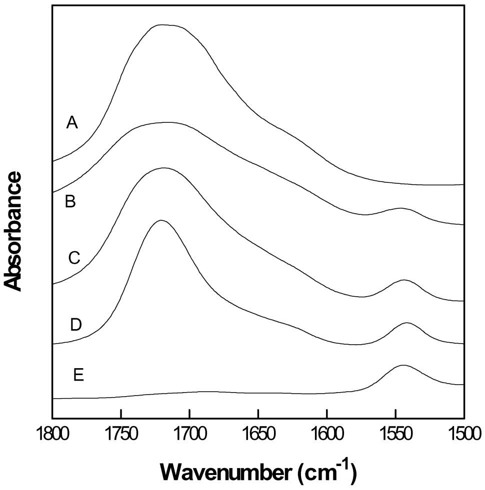


Fig. 3. FTIR spectra recorded at 140 °C, of the carbonyl region of (A) PAA, (B) blend 1, (C) blend 2, (D) blend 3 and (E) PMVT.

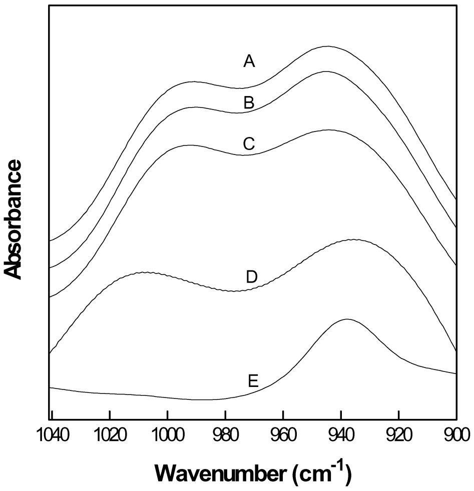


Fig. 4. FTIR spectra recorded at 140 °C, of the P–OH deformation region of (A) complex 1, (B) complex 2, (C) complex 3, (D) PVPA and (E) PMVT.

3.3. XPS characterization

3.3.1. N 1s spectra

Fig. 5 shows the N 1s core-level spectra of PMVT and the three PMVT/PVPh blends. The N 1s spectrum of PMVT shows a single nitrogen environment with a BE of 398.8 eV. However, the N 1s peaks of the blends are shifted slightly to the higher-BE ends, indicating that the nitrogen atoms in the blends become slightly more electropositive. Each N 1s peak of the blend can be deconvoluted into two component peaks: one remains at 398.8 eV and the other around

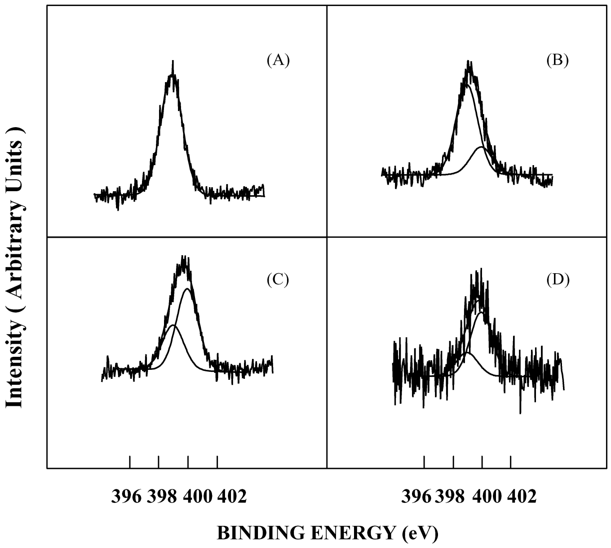


Fig. 5. N 1s spectra of PMVT and PMVT/PVPh blends: (A) PMVT, (B) blend 4, (C) blend 5 and (D) blend 6.

399.8 eV. As shown in Table 3, the fraction of hydrogen-bonding nitrogen atoms increases with increasing PVPh content in the blends.

Similarly, a high-BE peak at 399.8 eV is developed in each of the spectra of the PMVT/PAA blends (Fig. 6). Recent studies have shown that BE shift values of 1.0 and 2.0 eV indicate the involvement of the nitrogen in hydrogen-bonding interaction and ionic interaction, respectively. Therefore, XPS studies show that thiazole nitrogen atoms are involved in hydrogen-bonding interactions with PVPh and PAA. The conclusions agree with those revealed by FTIR studies as mentioned earlier. It is of interest to note that PAA interacts with P4VPy, PMPMA and PVI through ionic interactions but it interacts with PMVT through hydrogen-bonding interactions. Since thiazole is less basic than pyridine, piperidine and imidazole, it is more difficult for thiazole groups of PMVT to be protonated by PAA.

The protonation of thizaole nitrogen atoms depends on the acidity of the component polymers. As shown in Fig. 7, the BE shift values of thiazole nitrogen atoms in the PMVT/PVPA complexes about 2.0 eV which is nearly the same as those of the ionic-bonded PMPMA/PVPA and P4VPy/PVPA complexes [2,27]. This suggests that the interpolymer interaction at the site of thiazole nitrogen in the PMVT/PVPA complexes is ionic interaction. It is reasonable since the PVPA is more acidic than PAA or PVPh and thereby facilitates the proton transfer from PVPA to PMVT. The existence of ionic interactions in polymer blends depends on acidity of proton-donating polymer and basicity of proton-accepting polymer. Although PAA is a strong acidic polymer, the acidity of PAA is not strong enough for protonation of PMVT. Moreover, PMVT is less basic than PVI, P4VPy and PMPMA, it is more difficult for PMVT to be protonated by PAA. It is not surprising to note that interpolymer interactions between PVPh and PMVT are hydrogen-bonding but not ionic interactions since PVPh also forms hydrogen-bonding interactions with stronger basic polymers such as PMPMA [1], PVI [17] and P4VPy [30].

3.3.2. O 1s spectra

The hydrogen-bonding interaction in the PMVT/PVPh blends is also evidenced from the O 1s spectra of the blends. Fig. 8 shows the O 1s core-level spectra of PVPh and the three PMVT/PVPh blends. The O 1s of PVPh has a BE value of 533.2 eV. However, the O 1s peaks of the blends are shifted slightly to the low-BE ends, indicating that the oxygen atoms in the blends become more electronegative. Each O 1s peak of the blend is deconvoluted into two component peaks: one remains at 533.2 eV and the other around 532.4 eV. It is of interest to note that the BE-shift value of the oxygen in the PMVT/PVPh blend is around 0.8 eV, while that of the oxygen in the P4VPy/PVPh blend is around 1.2 eV [30]. Therefore, the larger BE-shift value of oxygen indicates stronger hydrogen-bonding interactions in P4VPy/PVPh blends. The fraction of the O 1s low-BE component peak increases with increasing PMVT content in the blend as shown in Table 3.

Table 3
Characteristics of PMVT/PVPh blends

Blend sample number	4	5	6
Mole fraction of PVPh in feed	0.21	0.51	0.81
T_g (°C)	115	138	151
N 1s BE (eV)	398.8, 399.9	398.8, 399.8	398.8, 399.9
Fraction of high-BE N 1s peak	0.24	0.64	0.73
O 1s BE (eV)	533.2, 532.4	533.2, 532.4	532.0, 532.3
Fraction of low-BE O 1s peak	0.58	0.37	0.32

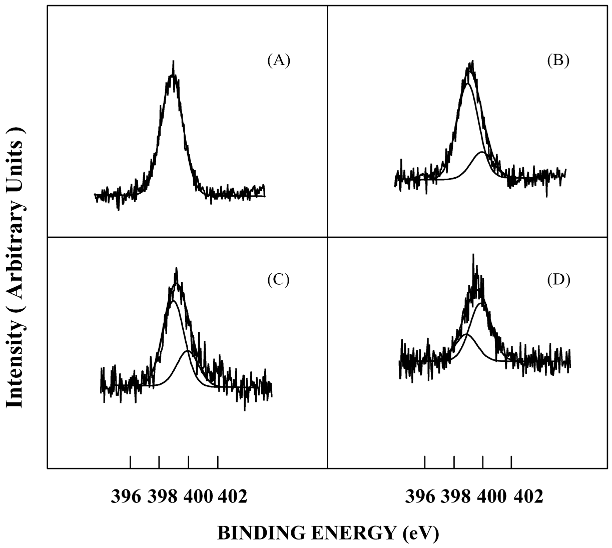


Fig. 6. N 1s spectra of PMVT and PMVT/PAA blends: (A) PMVT, (B) blend 1, (C) blend 2 and (D) blend 3.

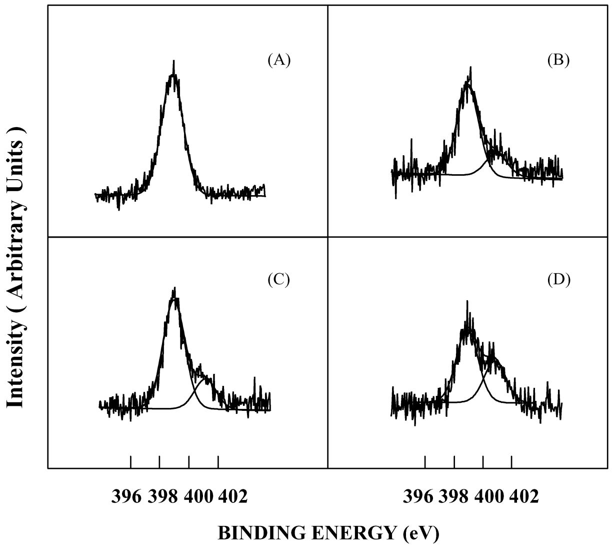


Fig. 7. N 1s spectra of PMVT and PMVT/PVPA complexes: (A) PMVT, (B) complex 3, (C) complex 2 and (D) complex 1.

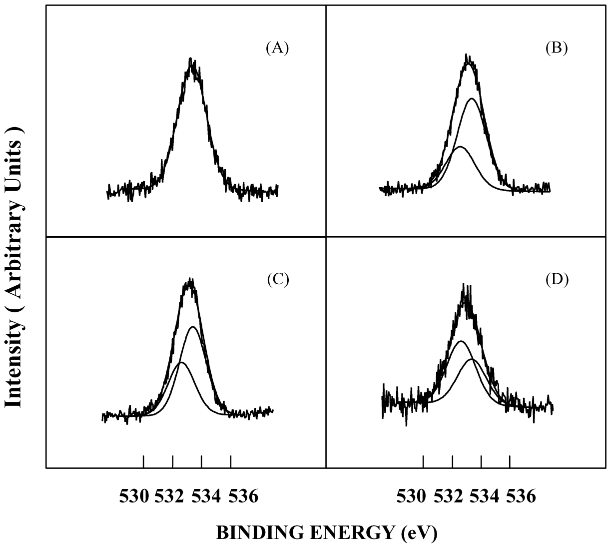


Fig. 8. O 1s spectra of PVPh and PMVT/PVPh blends: (A) PVPh, (B) blend 6, (C) blend 5 and (D) blend 4.

3.3.3. *S 2p spectra*

The S 2p spectra of the blends and complexes can provide evidence of interactions between sulfur atoms in PMVT and PVPh, PAA or PVPA. Fig. 9 shows the S 2p spectra of PMVT, blend 2, blend 5 and complex 2. For PMVT, the S 2p spin-orbit split doublet is located at 163.5 and 164.7 eV, attributed to S $2p_{3/2}$ and S $2p_{1/2}$, with an intensity of 2:1, respectively. The S 2p peaks of the blends and complex are broader, indicating a change in the environment of sulfur upon blending. Each S 2p peak of the blend and complex can be deconvoluted into two doublets, with the original doublet with BE values of 163.5 eV for S $2p_{3/2}$ and 164.7 eV for S $2p_{1/2}$ (full curve). The peak synthesis is based on the principle that the new doublet also maintains an intensity ratio of 2:1 for S $2p_{3/2}$ and S $2p_{1/2}$, and the difference in the BE values of S $2p_{3/2}$ and S $2p_{1/2}$ is also kept at 1.2 eV. In all the cases, the sum of the intensities of the two doublets agrees well with the experimental signals. The appearance of a new high-BE S 2p doublet (dashed curves) in the spectra indicates the sulfur atoms in the complex 2, blend 2 and blend 5 become more electropositive, leading to a high-BE shift which indicates the existence of interactions between sulfur atoms and the three proton-donating polymers.

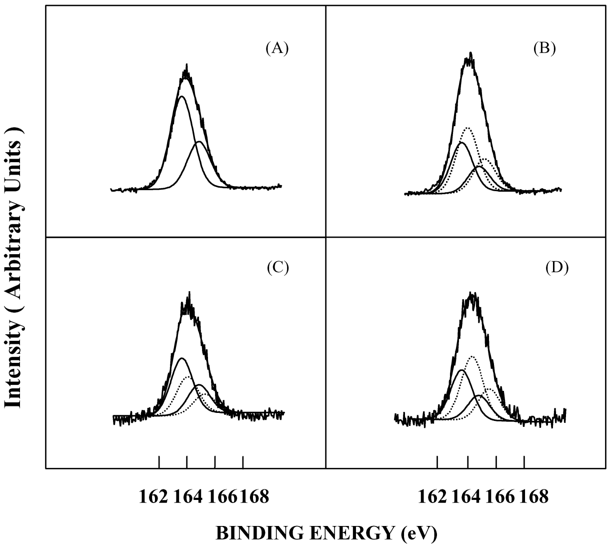


Fig. 9. S 2p spectra of (A) PMVT; (B) blend 5 (PMVT/PVPh blend); (C) blend 2 (PMVT/PAA blend) and (D) complex 2 (PMVT/PVPA complex).

The interactions between proton-donating polymers and nitrogen atoms in PMVT make the nitrogen atoms in PMVT become electropositive. Because of the conjugation effect of thiazole ring, the electronic environment of sulfur atoms also changes once nitrogen atoms interact with proton-donating polymers by hydrogen-bonding or ionic interactions. To investigate the effect, pPMVT was prepared. As shown in Fig. 10(B), two different nitrogen environments can be discerned in the spectrum of pPMVT. The appearance of a new high-BE N 1s peak at 401.2 eV indicates that some of nitrogen atoms of pPMVT are protonated. The S 2p spectrum of pPMVT is shown Fig. 11(B). No distinct changes in the S 2p spectrum of pPMVT compared with that of PMVT are observed, indicating that the electronic environment of sulfur is not appreciably affected by the protonation of nitrogen atom. Therefore, the appearance of a new high-BE S 2p doublet in the spectra of complex 2, blend 2 and blend 5 arises from interactions between sulfur atoms and the proton-donating polymers but not from conjugation effect when electronic environment of nitrogen atoms change.

4. Conclusions

PMVT forms miscible blends with PVPh and PAA in ethanol solutions and ethanol/water (1:1) solutions,

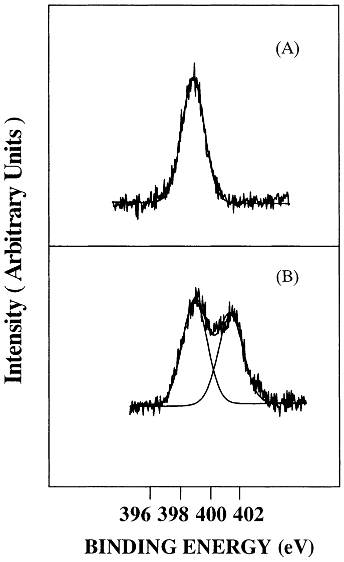


Fig. 10. N 1s spectra of (A) PMVT and (B) pPMVT.

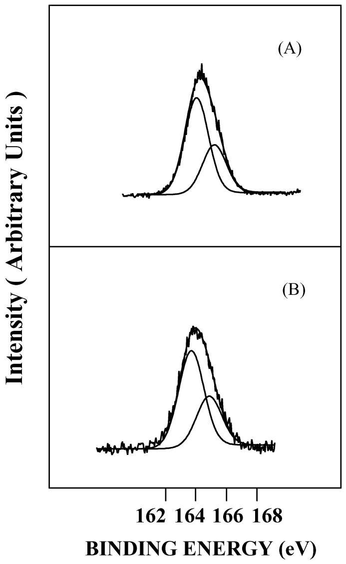


Fig. 11. S 2p spectra of (A) PMVT and (B) pPMVT.

respectively, and complexes with PVPA in ethanol/water (1:1) solutions. Thermal analysis and FTIR studies show that the strengths of hydrogen-bonding interactions in the PMVT/PVPh blends are weaker than those in the P4VPy/PVPh and PVI/PVPh blends. XPS studies show that both nitrogen atoms and sulfur atoms of PMVT are involved in the interactions with PVPh, PAA and PVPA. The nitrogen atoms of PMVT interact with PVPh and PAA by hydrogen-bonding interactions and with PVPA by ionic interactions.

References

[1] Luo XF, Goh SH, Lee SY, Tan KL. Macromolecules 1997;30:4934.
[2] Luo XF, Goh SH, Lee SY, Tan KL. Macromolecules 1998;31:3251.
[3] Liu Y, Goh SH, Lee SY, Huan CHA. Macromolecules 1999;32:1967.
[4] Goh SH, Liu Y, Lee SY, Huan CHA. Macromolecules 1999;32:8595.
[5] Liu Y, Goh SH, Lee SY, Huan CHA. J Polym Sci Polym Phys 2000; 38:501.
[6] Huang XD, Goh SH, Lee SY, Huan CHA. Macromolecules 1999;32: 4327.
[7] Coleman MM, Painter PC. Appl Spectrosc Rev 1984;20:255.
[8] Sargent M, Koenig JL. Adv Chem Ser 1993;236:191.
[9] Goh SH, Lee SY, Dai J, Tan KL. Polymer 1996;37:5305.
[10] Zhou X, Goh SH, Lee SY, Tan KL. Macromolecules 1998;31:3631.
[11] Goh SH, Lee SY, Zhou X, Tan KL. Macromolecules 1998;31:4260.
[12] Li L, Chan CM, Weng LT. Polymer 1998;39:2355.
[13] Li L, Chan CM, Weng LT, Xiang ML, Jiang M. Macromolecules 1998;31:7248.
[14] Vivas de Meftahi M, Frechet JMJ. Polymer 1988;29:477.
[15] Dai J, Goh SH, Lee SY, Siow KS. Polym J 1994;26:905.
[16] Goh SH, Lee SY, Tan LIK. Polym Bull 1996;37:253.
[17] Luo XF, Goh SH, Lee SY. Macromol Chem Phys 1999;200:399.
[18] Oyama HT, Nakajima T. J Polym Sci Polym Chem Ed 1983;21:2987.
[19] Fujimori K, Costigan MJ, Trainor GT. J Polym Sci Polym Chem Ed 1984;22:2479.
[20] Luo XF, Goh SH, Lee SY, Huan CHA. Macromol Chem Phys 1999; 200:874.
[21] Zhou X, Goh SH, Lee SY, Tan KL. Appl Surf Sci 1998;126:141.
[22] Lee SL, Den TG, Liang XM, Lintelmann J, Kettrup A. HRC-J High Resolut Chromatogr 1995;18:671.
[23] Brostow W, Seo KH, Back JB, Lim JC. Polym Eng Sci 1995;35:1016.
[24] Yi JZ, Goh SH. Macromolecules 2001;34:4662.
[25] Kwei TK. J Polym Sci Polym Lett Ed 1984;22:307.
[26] Zhou X, Goh SH, Lee SY, Tan KL. Polymer 1998;39:3631.
[27] Zhou X, Goh SH, Lee SY, Tan KL. Polymer 1997;38:5333.
[28] Moskala EJ, Varnell DF, Coleman MM. Polymer 1985;26:228.
[29] Emeleus HJ, Haszeldine RN, Paul RC. J Chem Soc 1955;563.
[30] Zhou X, Goh SH, Lee SY, Tan KL. Appl Surf Sci 1997;119:60.