

# Preparation and Identification of a Soluble Copolymer from Pyrrole and *o*-Toluidine

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**ABSTRACT:** A series of copolymers were prepared by chemically oxidative polymerization of pyrrole (PY) and *ortho*-toluidine (OT) in HCl aqueous medium. The yield, intrinsic viscosity, and solubility of the copolymers were studied by changing the monomer molar ratio. The resulting PY/OT copolymers were identified by FTIR, <sup>1</sup>H-NMR, DSC, and WAXD techniques. The experimental results showed that the oxidative polymerization of pyrrole and *o*-toluidine is exothermic and the resulting polymers exhibit an enhanced solubility in most organic solvents compared with that of pyrrole homopolymer. The polymer obtained is a real and amorphous copolymer containing pyrrole and *o*-toluidine units. © 2001 John Wiley & Sons, Inc. *J Appl Polym Sci* 82: 510–518, 2001

**Key words:** pyrrole–toluidine copolymer; pyrrole-containing copolymer; oxidative copolymerization; Fourier transform infrared; proton nuclear magnetic resonance; differential scanning calorimetry

## INTRODUCTION

Conducting nitrogen heterocyclic polymers have attracted much attention recently because they exhibit high gas separation ability, high electric conductivity, and performance stability.<sup>1</sup> Polypyrrole of aromatic nitrogenous polymers is the most attractive one because of its higher oxygen/nitrogen separation factor (up to 8).<sup>2</sup> Polypyrrole has been proposed as an insoluble and nonthermo-

plastic polymer like most other conductive polymers. Poly(*o*-toluidine) has been reported to be a soluble polymer in common organic solvents.<sup>3</sup> Attempts have been made to solubilize pyrrole (PY) polymer by inducing one or two long aliphatic substituent groups on every pyrrole ring and copolymerizing pyrrole with other monomers. One example is poly(3-octylpyrrole) that can be soluble in low boiling point solvents such as ethylene chloride, tetrahydrofuran, chloroform, acetone, and *N*-methylpyrrolidone.<sup>4</sup> The copolymerizations of pyrrole with aniline,<sup>5–7</sup> *N*-methylpyrrole,<sup>8</sup> or thiophene<sup>9</sup> have been reported, but no report on the chemically oxidative copolymerization of pyrrole with *o*-toluidine (OT) was found. In this study, a soluble copolymer of pyrrole and *o*-tolu-

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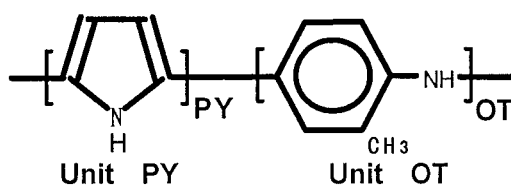
idine was prepared by oxidative polymerization for the first time. The polymerization exotherm, yield, and intrinsic viscosity of the copolymer were studied by changing the PY/OT monomer ratio. Its solubility, macromolecular and supermolecular structures, and thermal and tensile behaviors are evaluated.

## EXPERIMENTAL

Pyrrole (PY), *o*-toluidine (OT), oxidants, and solvents were commercially obtained and used as received.

### Polymerization

PY/OT copolymers were prepared by oxidative polymerization by using a previously described method.<sup>10–13</sup> A representative procedure for the preparation of the PY/OT (20/80) copolymer is as follows: 0.42 mL (6 mmol) PY and 2.58 mL (24 mmol) *o*-toluidine was added to 40 mL of a 1M HCl solution in a 150-mL glass flask in water bath and stirred vigorously for 0.5 h. Ammonium persulfate [(NH<sub>4</sub>)<sub>2</sub>S<sub>2</sub>O<sub>8</sub>, 6.84 g (30 mmol)] was dissolved separately in 20 mL 1M HCl to prepare an oxidant solution. The monomer solution was then treated with the oxidant solution added dropwise at a rate of 0.1 mL/min at 23.4°C for a period of about 115 min (the total molar ratio: monomer/oxidant = 1/1). Immediately, after the first few drops the reaction solution turned blue-violet. The reaction mixture was stirred for 24 h at ambient temperature. The copolymer hydrochloride salt was isolated from the reaction mixture by filtration and washed with an excess of distilled water to remove the oxidant and oligomers. The hydrochloride salt was subsequently neutralized in 180 mL 0.2M ammonium hydroxide and stirred for 24 h to obtain the base form of PY/OT copolymer. The copolymer base was washed with excess water. A bluish-black solid powder was left to dry in ambient air for 1 week. The copolymer of 1.691 g was obtained with the yield of about 57.4%. The PY/OT copolymers exhibit the following nominal structure:



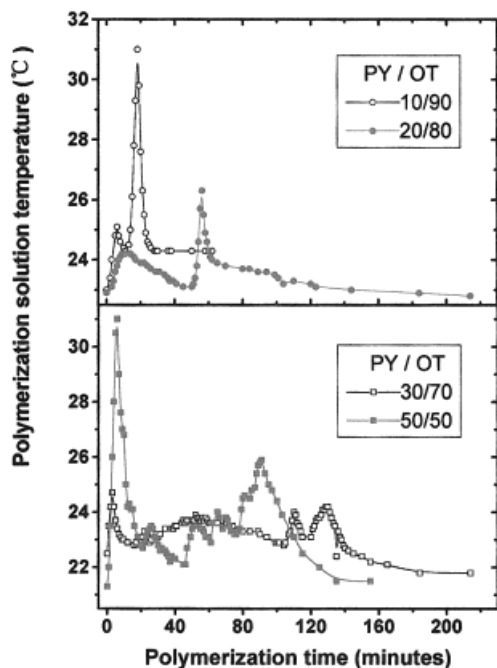
## Measurements

The intrinsic viscosity for the copolymers of pyrrole with *o*-toluidine in DMSO was measured with a Ubbelodhe viscosimeter at 25°C. To evaluate the solubility of the copolymers, a polymer powder sample of 5 mg was added to the solvent of 0.5 mL and dispersed thoroughly. After the mixture was swayed continuously for 24 h at room temperature, the solubility of the polymers was characterized. IR spectra were recorded on a Spectrum One spectrometer (Spectrum, Houston, TX) at 4 cm<sup>-1</sup> resolution on KBr pellets. <sup>1</sup>H-NMR spectra were obtained in deuterated chloroform and dimethylsulfoxide using a Bruker AC200 spectrometer (Bruker Instruments, Billerica, MA) operated at 200 MHz and a Bruker MSL-300 spectrometer operated at 300.13 MHz. DSC measurement was performed at a heating rate 10°C/min with the sample size of 6–7 mg using a Perkin-Elmer 7 Pyris thermal analyzer (Perkin Elmer Cetus, Norwalk, CT). A blend film of PY/OT (10/90) copolymer with ethyl cellulose by solution cast method was characterized by Fourier wide-angle X-ray diffraction (WAXD) in a Bragg angle range of 3–50° using the wavelength 0.154 nm of CuK<sub>α</sub> electron beam in step-by-step scanning region and recorded nickel-filtered radiation at 25°C with a Bruker Analytical X-ray Systems D8 Advance X-ray Diffractometer (made in Germany). The scanning rate was 2°/min. The stress-strain curves of the films were obtained by using an Instron 5565 tensile tester (Instron) at a stretching rate of 10 mm/min, relative humidity of 77%, and 22°C.

## RESULTS AND DISCUSSION

### Preparation of the Copolymers of Pyrrole and *o*-Toluidine

The copolymerization of pyrrole and *o*-toluidine with ammonium persulfate as an oxidant in 40 mL 1M HCl aqueous solution yielded bluish-black precipitates as products. The progress in copolymerization reaction was followed by measuring the solution temperature. The variation of polymerization solution temperature with polymerization time is shown in Figure 1. It was found that with dropping oxidant solution slowly and regularly the polymerization solution temperature shows several peaks and finally reaches a nearly constant value. Several temperature peaks result from the difference in the dropping rate of



**Figure 1** Variation of polymerization solution temperature with oxidative copolymerization time of pyrrole (PY)/*o*-toluidine (OT) molar ratios of 10/90 (—○—), 20/80 (—●—), 30/70 (—□—), 50/50 (—■—), and  $(\text{NH}_4)_2\text{S}_2\text{O}_8$  : monomer = 1 : 1 in 1M HCl.

the oxidant solution. When the dropping rate is faster the temperature must be higher. Because the dropping rate of oxidant solution for PY/OT (50/50) copolymerization is the fastest, this copolymerization exhibits the strongest exothermic capability among the four copolymerization systems. The enhancement of the solution temperature is about 9.7°C for the PY/OT (50/50) system. On the contrary, PY/OT (30/70) copolymerization exhibits the lowest exothermic effect, with an enhancement of the solution temperature of about 2.3°C. In any case, the PY/OT copolymerization is exothermic, like the copolymerization of pyrrole with *o*-anisidine or *m*-toluidine.

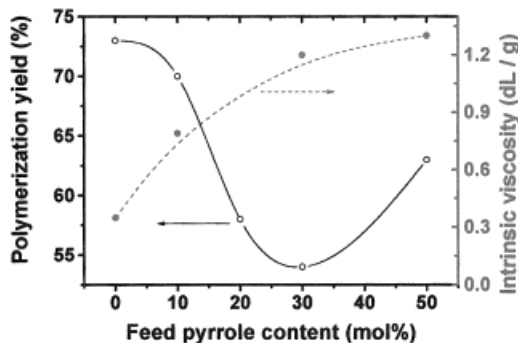
It is found from Figure 2 that the copolymerization yield of pyrrole and *o*-toluidine was dependent on the monomer ratio. The yield decreases with an increase in feed PY content from 0 to 30 mol % but increases with a further increase in feed PY content from 30 to 50 mol %. The PY/OT copolymerization shows the minimal yield at the feed PY content of about 30 mol % but the highest yield at the feed PY content of 10 mol %, except for the *o*-toluidine homopolymer. The highest yield (69%) is higher than that of polypyrrole when  $\text{FeCl}_3$  was used as oxidant and the oxidant/PY

ratio is 1/1.<sup>14</sup> A different relationship between the intrinsic viscosity and comonomer ratio (Fig. 2) is observed. The intrinsic viscosity of the PY/OT copolymers increases monotonically with increasing feed pyrrole content from 0 to 50 mol % and reached the highest value of 1.3 dL/g at the pyrrole content of 50 mol %. The enhancement of the intrinsic viscosity may be responsible not only for the enhancement of the molecular weight of the copolymers but also for the rigidity of the copolymer chains. Note that almost no report on the intrinsic viscosity of polypyrrole is found because polypyrrole is insoluble in most organic solvents. Therefore, it is impossible for us to compare the intrinsic viscosities of PY/OT copolymers with polypyrrole. It can be concluded that the yield and intrinsic viscosity of the copolymer of pyrrole with *o*-toluidine was significantly influenced by the monomer ratio. The copolymer with the pyrrole/*o*-toluidine ratio of 50/50 exhibits both the larger yield and the highest intrinsic viscosity.

The copolymerization yield of pyrrole and *o*-toluidine was affected by an addition of surfactant. When sodium dodecylbenzene sulfonate (DBS) was added into PY/OT (30/70) copolymerization system with the monomer/oxidant/DBS molar ratio of 10/2/5, the polymerization yield is lower than 23%, although the intrinsic viscosity of the copolymer obtained is higher than 1.6 dL/g. The molecular structure of the copolymer obviously changes with the addition of surfactant, as discussed below.

#### Solubility of the Copolymers of Pyrrole and *o*-Toluidine

It can be seen from Table I that with increasing feed OT content, the PY/OT copolymers exhibit an



**Figure 2** Influence of pyrrole (PY) feed content on polymerization yield and intrinsic viscosity of PY/OT copolymers with monomer :  $(\text{NH}_4)_2\text{S}_2\text{O}_8$  ratio of 1 : 1 for 24 h at 23°C in 1M HCl.

**Table I Solubility of Pyrrole (PY)/*o*-Toluidine (TO) Copolymers in Organic Solvents**

Feed PY/TO Molar Ratio	Solubility in Solvents <sup>a</sup> (%) and Solution Color <sup>b</sup>					
	NMP	DMSO	CHCl <sub>3</sub>	THF	Benzene	CH <sub>2</sub> Cl <sub>2</sub>
0/100	100	97	S (bv)	PS (bg)	MS (db)	
10/90	98 (b)	90 (b)	S (bv)	S (b)	PS (bv)	PS (v)
20/80	100 (bb)	88 (bb)	S (bv)	S (b)	PS (bv)	PS (v)
30/70	83 (b)	47 (b)	S (v)	S (b)	PS (v)	PS (v)
30/70 <sup>c</sup>	59 (rb)	47 (rb)	SS	SS	I	SS
50/50	40 (b)	64 (b)	PS (bv)	PS (b)	PS (bv)	PS (v)
100/0	I	I	I	I	I	I

<sup>a</sup> I = insoluble; MS = major soluble; PS = partially soluble; S = soluble; SS = slightly soluble.

<sup>b</sup> The solution color is indicated in parentheses: b = blue; bb = bluish black; bg = bluish green; bv = bluish violet; db = dark blue; rb = reddish brown.

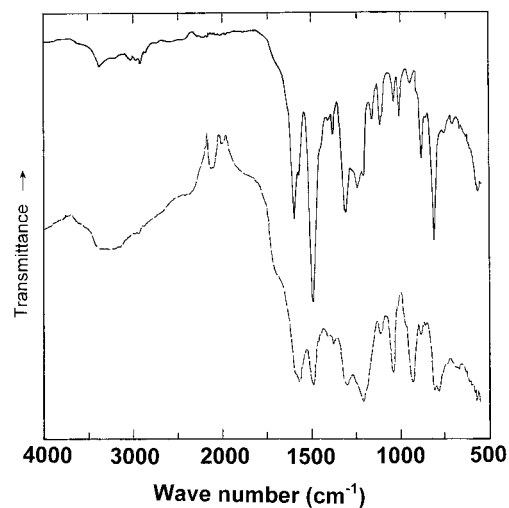
<sup>c</sup> The copolymer obtained at the monomer/ammonium persulfate/sodium dodecylbenzene sulfonate (DBS) molar ratio of 10/2/5.

enhanced solubility in the five solvents. Finally, PY/OT (0/100), (10/90), (20/80), and (30/70) polymers were basically soluble in NMP, DMSO, chloroform, and tetrahydrofuran, and partially soluble in benzene. It should be noted that the PY/OT (50/50) copolymer with the lowest feed OT content but the highest intrinsic viscosity exhibits the poorest solubility among the five PY/OT copolymers. Thus, it could be considered that the improvement of the copolymer solubility with increasing OT content may be attributed to the change of molecular structure or intrinsic viscosity of the copolymers.<sup>3</sup> These observations confirm that the oxidative copolymerization between the two monomers PY and OT has occurred. As compared with OT homopolymer, an improvement in solubility results from the incorporation of PY moieties into OT polymer chain.

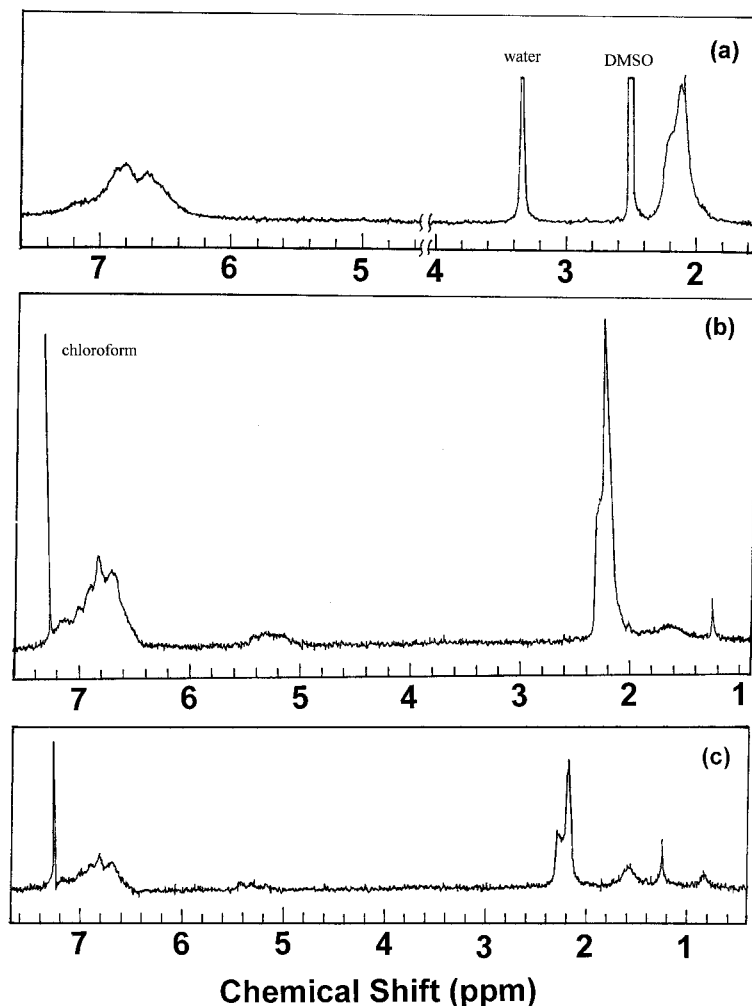
#### FTIR Spectra of the Copolymers of Pyrrole and *o*-Toluidine

Representative FTIR spectra for the copolymers with PY/OT molar ratios of 10/90 and 50/50 are shown in Figure 3. It was reported that poly(*o*-toluidine) exhibits  $\text{-NH-}$  stretching vibration at  $3372\text{ cm}^{-1}$  and polypyrrole hardly ever exhibits  $\text{-NH-}$  vibration peak.<sup>3,14,15</sup> A broad and weak band centered at  $3371\text{--}3252\text{ cm}^{-1}$ , attributed to the characteristic  $\text{N-H}$  stretching vibration, suggests the presence of  $\text{-NH-}$  groups in OT and PY units. This broad band becomes even broader and shifts to lower wavenumber with an increase in PY feed content from 0 to 50 mol %. Four weak peaks at  $3023, 2969, 2919,$  and  $2854\text{ cm}^{-1}$  should

be attributed to aromatic and aliphatic  $\text{C-H}$  stretching vibrations, respectively. With increasing feed PY content, these four peaks all become weaker because the PY unit contains fewer aromatic  $\text{C-H}$  bonds and does not contain aliphatic  $\text{C-H}$  bonds at all. Thus the characteristics of IR absorption spectra of PY/OT copolymers above  $2000\text{ cm}^{-1}$  are dominated by the OT unit. However, the IR absorption below  $2000\text{ cm}^{-1}$  is strongly influenced by the PY units. A comparison of the spectra of poly(*o*-toluidine),<sup>3</sup> polypyrrole,<sup>14,15</sup> and PY/OT copolymers shown in Figure 3 reveals some differences in the relative absorbance and wavenumber. With increasing feed PY content, the relative absorption intensity at  $1571,$



**Figure 3** FTIR absorption spectra of the copolymers with PY/OT molar ratios of 10/90 (—) and 50/50 (---).



**Figure 4**  $^1\text{H}$ -NMR spectra of the copolymers with PY/OT molar ratios of (a) 10/90 in deuterated dimethylsulfoxide ( $\text{DMSO-}d_6$ ), (b) 20/80, and (c) 30/70 in deuterated chloroform at 200 MHz.

1208, 1040, and  $928\text{ cm}^{-1}$  becomes significantly stronger. In particular, a medium absorption at 1208 for PY/OT (10/90) copolymer became the strongest absorption for PY/OT (50/50). The change of IR absorption should be attributed to the introduction of PY unit into the polymer chain because the bands at about 1540, 1170, and  $900\text{ cm}^{-1}$  are proposed to be characteristic of polypyrrole.

In addition, the FTIR spectrum of PY/OT (10/90) copolymer looks almost the same as that of PY/MT (10/90) copolymer.<sup>16</sup> The FTIR spectrum of PY/OT (50/50) copolymer is similar to that of PY/MT (50/50) copolymer. Therefore, it can be concluded that both copolymers have similar macromolecular structure.

#### $^1\text{H}$ -NMR Spectra of the Copolymers of Pyrrole and *o*-Toluidine

$^1\text{H}$ -NMR spectra of the copolymers of pyrrole and *o*-toluidine in deuterated DMSO and chloroform are not the same (Fig. 4). The  $^1\text{H}$ -NMR spectrum of the PY/OT (10/90) copolymer in deuterated DMSO exhibits an aromatic proton peak at 6.3–7.3 ppm and a methyl proton peak at 1.9–2.3 ppm.  $^1\text{H}$ -NMR spectra of the PY/OT (20/80) and (30/70) copolymers in deuterated chloroform exhibit an aromatic proton peak at 6.4–7.2 ppm, a methyl proton peak at 2.0–2.4 ppm, and an additional weak resonance peak around 5.1–5.5 ppm, which might be attributed to the end amino group. Other weak peaks at 0.82, 1.25, and 1.6 ppm in the  $^1\text{H}$ -NMR spectra of the PY/OT (20/80)

and (30/70) copolymers in deuterated chloroform might result from impurity such as water. The peak shape of aromatic proton in the  $^1\text{H-NMR}$  spectra of PY/OT copolymers in both solvents changes slightly with increasing feed PY content from 10 to 20 mol %, although the peak shape of methyl proton is substantially the same. Even though the peak shape of the aromatic proton in the  $^1\text{H-NMR}$  spectra of PY/OT copolymers in deuterated chloroform does not change with increasing feed PY content from 20 to 30 mol %, the methyl proton peak exhibits an enhanced resolution. These spectra provide little information toward the calculation of sequence distribution of the comonomer units. Because there are the same numbers of aromatic protons as those of methyl protons on every OT unit in the polymer chains, the number of aromatic protons on PY units may be calculated through the following equation:

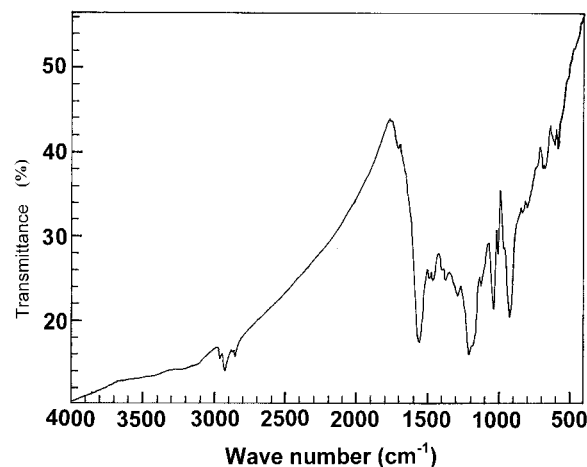
$$\text{PY proton area} = \text{Total aromatic proton area} \\ - \text{methyl proton area}$$

Therefore,

$$\text{Molar ratio of PY/OT} = (\text{PY proton area}/2) \\ \div (\text{methyl proton area}/3)$$

On the basis of a comparison of the area of the aromatic proton peak (6.3–7.3 ppm) on PY and OT units with the methyl proton peak (1.9–2.3 ppm) on the OT unit, the molar ratio of PY to OT units for PY/OT copolymer with feed molar ratio of 10/90 can be calculated to be 7/93. It seems that the actual PY content is slightly lower than the feed PY content for PY/OT (10/90) copolymer. Unfortunately, the actual PY/OT molar ratio cannot be calculated by the preceding method when deuterated chloroform is used as NMR solvent because the total area of the aromatic proton peak is smaller than that of the methyl proton peak. This appears impossible because there should be a greater, or at least the same, number of aromatic protons as methyl protons in the polymer chain, even if there is no pyrrole unit. If there is a pyrrole unit, the number of aromatic protons must be greater than that of methyl protons.

The  $^1\text{H-NMR}$  spectrum of PY/OT (10/90) copolymer in deuterated DMSO is slightly different from that of PY/MT (10/90) copolymer,<sup>16</sup> but the  $^1\text{H-NMR}$  spectrum of PY/OT (30/70) copolymer in deuterated chloroform is very similar to that of

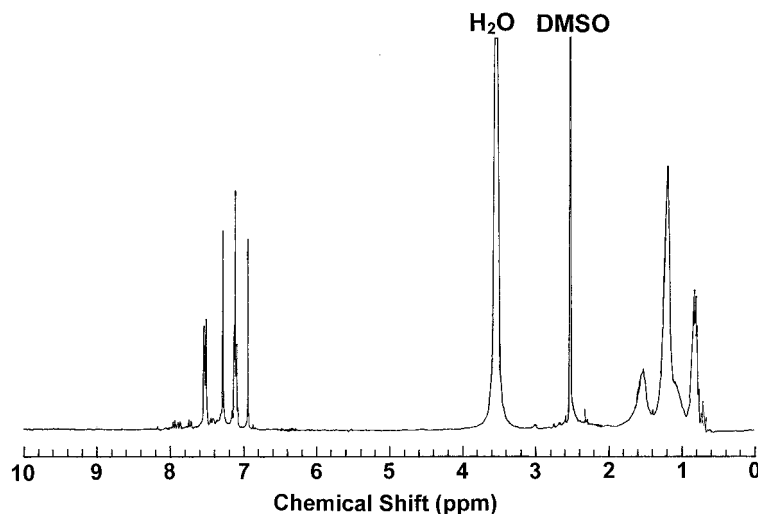


**Figure 5** FTIR absorption spectrum of PY/OT (30/70) copolymer base powder. The copolymer was prepared at the monomer/ammonium persulfate/sodium dodecylbenzene sulfonate molar ratio of 10/2/5 and the copolymer hydrochloride salt was subsequently neutralized in 180 mL 0.2M ammonium hydroxide, stirred for 24 h, and washed with excess water to obtain the base form of PY/OT (30/70) copolymer.

PY/MT (30/70) copolymer. Therefore, it can be further concluded that both copolymers have similar macromolecular structures, which is the same as the results illustrated by FTIR spectra.

#### Effect of Sodium Dodecylbenzene Sulfonate on the Copolymerization of Pyrrole and *o*-Toluidine

The PY/OT (30/70) copolymer synthesized by adding sodium dodecylbenzene sulfonate (DBS) exhibits different solubility, FTIR spectrum, and  $^1\text{H-NMR}$  spectrum, as shown in Table I and Figures 5 and 6. Surprisingly, the solubility of the copolymer containing DBS as an additive of copolymerization is lower than that of the corresponding additive-free copolymer, whereas it has been reported that the solubility of polypyrrole increases in the presence of dodecylbenzene sulfonic acid.<sup>17,18</sup> Perhaps, as compared with dodecylbenzene sulfonic acid, the copolymer obtained in the presence of sodium DBS has a different composition or molecular structure that influences the solubility. The FTIR spectrum of DBS-containing copolymer powder looks very much like the spectrum of pure polypyrrole<sup>14,15</sup> in the wavenumber range below 2000  $\text{cm}^{-1}$ , except that there are three weak absorptions of C—H vibration at 2850–2970  $\text{cm}^{-1}$  attributed to the dodecyl group on DBS. The  $^1\text{H-NMR}$  spectrum of the soluble



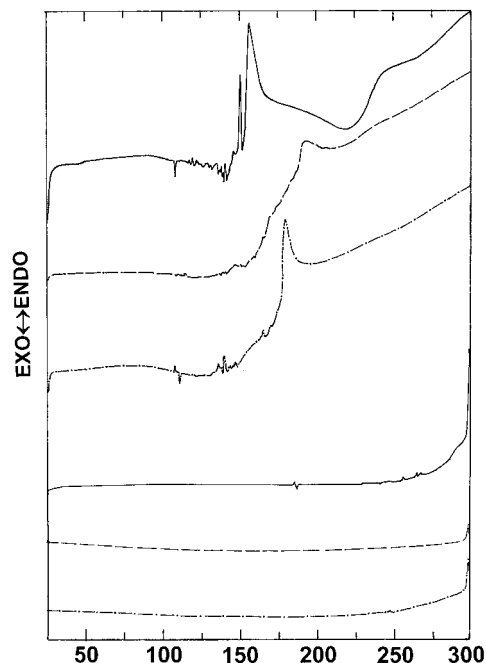
**Figure 6**  $^1\text{H}$ -NMR spectrum of the soluble part of dodecylbenzene sulfonate-containing PY/OT (30/70) copolymer base in  $\text{DMSO-}d_6$  at 300.13 MHz. The copolymer base was prepared under the same polymerization conditions as in Figure 5.

part of the DBS-containing copolymer in  $\text{DMSO-}d_6$  exhibits a very strong aliphatic proton peak at 0.7–1.5 ppm, a medium and sharp aromatic proton peak at 6.92–7.9 ppm, and a very weak methyl proton peak on the toluidine unit at about 2.3 ppm. On the basis of a comparison of the peak areas of aliphatic and aromatic protons, the resonance peaks at 6.92–7.9 ppm should be attributed to the benzene ring on DBS, and a small number of the aromatic rings on *o*-toluidine and pyrrole units. Furthermore, these results reveal that the soluble part of the DBS-containing copolymer in  $\text{DMSO}$  consists of the DBS and a small number of *o*-toluidine and pyrrole units, given that the DBS bonded to the PY/OT copolymer cannot be removed by washing with excess water. Briefly, the effect of DBS on the oxidative copolymerization of pyrrole and *o*-toluidine is apparent.

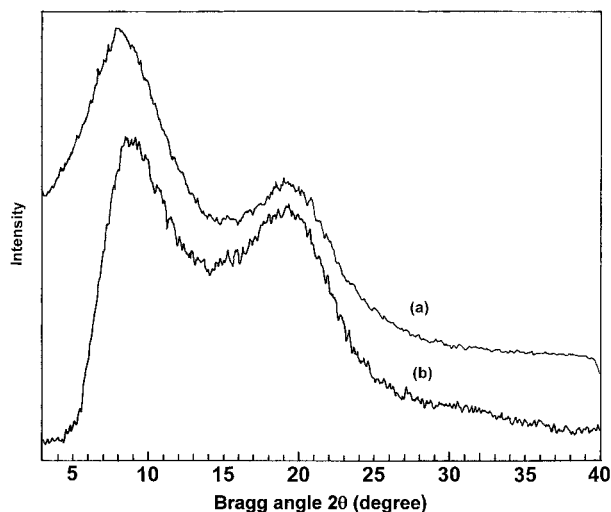
#### Thermal Behavior of the Copolymers of Pyrrole and *o*-Toluidine

The DSC scans of the PY/OT copolymers with three PY/OT ratios of 20/80, 30/70, and 50/50 are shown in Figure 7. Three scans all show a small and sharp endothermic peak at 156, 192, and 180°C for PY/OT (20/80), (30/70), and (50/50) copolymers, respectively. The respective endotherms are 38, 93, and 70 J/g. This endotherm should be attributed to the dedoping process of the copolymers. A broad exothermic peak was observed at 221°C for PY/OT (20/80) copolymer. It

was reported that poly(*o*-toluidine) exhibits a similar exothermic process at 250°C.<sup>3</sup> This exotherm could be attributed to the crosslinking reaction of



**Figure 7** Differential scanning calorimetry thermograms of the as-polymerized copolymer powders with PY/OT molar ratios of 20/80 (—), 30/70 (---), and 50/50 (—●—). The upper three lines are heating curves at a heating rate of 10°C/min and the lower three lines are cooling curves at a cooling rate of 10°C/min.



**Figure 8** Wide-angle X-ray diffractograms of (a) pure ethyl cellulose (EC) film (thickness 0.04 mm) and (b) the blend film of 10 wt % PY/OT (10/90) copolymer with 90 wt % EC (thickness 0.056 mm) made by solution cast technique in chloroform.

the polymers. These speculations are further confirmed by the fact that the cooling scans are featureless, except for very weak endothermic and exothermic peaks after the samples were allowed to maintain at 300°C for about 0.5 h. Therefore the PY/OT copolymers could not melt at a temperature below 300°C.

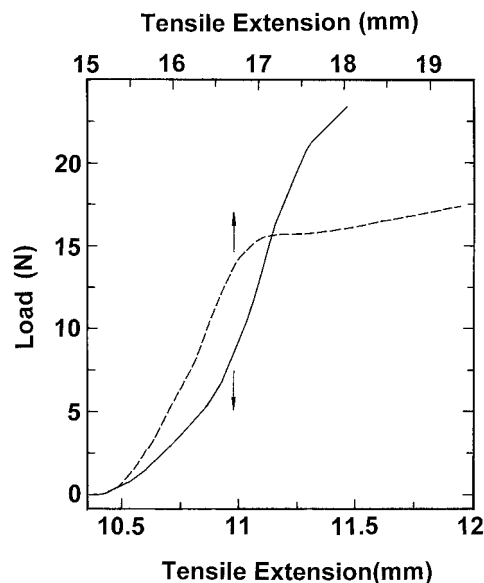
#### Wide-Angle X-ray Diffractogram and Tensile Property of the Blend Film of PY/OT Copolymer with Ethyl Cellulose

The blend film structure of the 10 wt % PY/OT (10/90) copolymer with 90 wt % ethyl cellulose prepared by a solution cast technique with chloroform as solvent was investigated by using wide-angle X-ray diffraction (WAXD), as shown in Figure 8. The WAXD diffractogram of pure ethyl cellulose film is shown in Figure 8 for an easy comparison. It can be seen that both films exhibit two broad diffraction peaks, suggesting that both of films are amorphous. With adding 10 wt % PY/OT copolymer into the ethyl cellulose film, the stronger diffraction peak at the Bragg angle of 8° becomes weaker and shifts to a larger Bragg angle (8.8°) but the weaker diffraction peak at the Bragg angle of 19° becomes stronger. These observations imply that the supermolecular structure of the films is strongly influenced by the PY/OT copolymer content. Consequently, the tensile stretching behavior of the blend film is quite

different from that of pure ethyl cellulose film, as shown in Figure 9. There is an apparent yield point with the yield strength of 45 MPa and yield elongation of 11% for pure ethyl cellulose. Upon adding 10 wt % PY/OT (10/90) copolymer into ethyl cellulose, the yield point almost disappears. The tensile strength (47 MPa) and elongation at break (11%) of the blend are lower than those (48 MPa and 28%, respectively) of pure ethyl cellulose, although the blend exhibits much higher initial modulus (1093 MPa) than that of pure ethyl cellulose (560 MPa).

#### CONCLUSIONS

The copolymers with four pyrrole to *o*-toluidine molar ratios were prepared by chemically oxidative polymerization in HCl aqueous medium at room temperature. The oxidative copolymerization of pyrrole with *o*-toluidine is highly exothermic. The copolymers exhibit an obvious dependency of polymerization yield, intrinsic viscosity, and solubility on copolymer monomer PY/OT ratio. Available highest yield and intrinsic viscosity are 69% for PY/OT (10/90) copolymer and 1.3 dL/g for PY/OT (50/50) copolymer. The polymers obtained are real copolymers, consisting of two



**Figure 9** Tensile stress-strain curves of (---) pure ethyl cellulose film (thickness 0.04 mm) and (—) its blend film (thickness 0.056 mm) with 10 wt % PY/OT (10/90) copolymer at a stretching rate of 10 mm/min, relative humidity of 77%, and 22°C.



monomer units, and do not melt at a temperature below 300°C. The blend film of PY/OT copolymer with ethyl cellulose, prepared by a solution cast technique, is amorphous and exhibits lower tensile strength and elongation at break but much higher modulus than that of pure ethyl cellulose film.

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