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### Metal-Ylide Complex-Initiated Radical Copolymerization of Methylmethacrylate and Styrene

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## METAL-YLIDE COMPLEX-INITIATED RADICAL COPOLYMERIZATION OF METHYLMETHACRYLATE AND STYRENE

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### ABSTRACT

Phenacyl dimethylsulfonium ylide complex of mercuric chloride (PDSY-HgCl<sub>2</sub>)-initiated radical copolymerization of styrene with methylmethacrylate (MMA) at 85 ± 0.1°C using dioxane as an inert solvent yields random copolymers as evidenced by NMR spectroscopy. The kinetic equation for the present system was  $R_p \propto [\text{PDSY-HgCl}_2]^{0.5} [\text{Sty}]^{1.0} [\text{MMA}]^{1.0}$ . The values of energy of activation ( $\Delta E$ ) and  $k_p^2/k_t$  were 48.0 kJ mol<sup>-1</sup> and 8.6 × 10<sup>-4</sup> L mol<sup>-1</sup>s<sup>-1</sup>, respectively. The mechanism of the reaction has also been proposed for the present system. The properties of copolymer were studied in the form of film. The film was highly absorptive for nitric acid but less absorptive for acetic acid. The film was water impermeable.

### INTRODUCTION

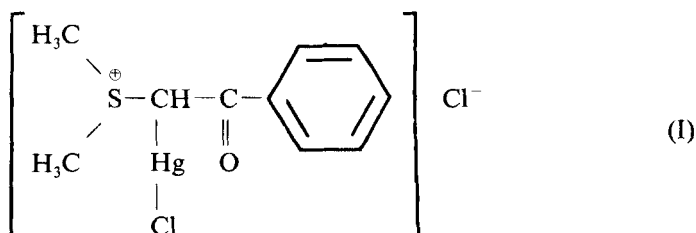
Ylides, in general, and metal ylide complexes, in particular, comprise a significant fraction of the organic chemical literature, but their application in the field of polymer chemistry is only recent. Copolymerization of methylmethacrylate with styrene has been widely studied using conventional initiators [1–4]. Recently, ylides have also been reported as a radical initiator for copolymerization of vinyl monomers [5, 6]. However, copolymerization by metal ylide complex has rarely been discussed in the literature.

During the past decades, synthesis of polymer films, because of their industrial significance, has revolutionized research in the field of polymer chemistry. The literature includes very few reports on the synthesis and characterization of copolymer films [7].

The present communication reports the kinetics of copolymerization of styrene with methylmethacrylate initiated by metal ylide complex, namely, phenacyldimethyl sulfonium ylide complex of mercuric chloride, and synthesis and properties of copolymer film.

## EXPERIMENTAL

Styrene (Sty) and methylmethacrylate (MMA) were distilled under reduced pressure and purified according to the method of Overberger [8]; solvents were purified by standard procedures [9]. Phenacyl dimethyl sulfonium ylide complex of mercuric chloride (PDSY-HgCl<sub>2</sub>) was prepared by the method reported in literature [10, 11] with the following structure:



Polymerization runs were carried out at  $85 \pm 0.1^\circ\text{C}$  for 1 h under nitrogen employing a modified dilatometric apparatus. The copolymers precipitated with acidified methanol were dried to constant weight. The copolymers were refluxed with acetonitrile and cyclohexane to remove homopolymers. There was negligible weight loss in cyclohexane but appreciable loss in acetonitrile. The copolymer was then dissolved in benzene and precipitated with acidified methanol. The reaction was carried out three times, and the coefficients for the plots varied from 0.003 to 0.008. Rates of polymerization ( $R_p$ ) were calculated from the slopes of linear conversion-time plots Fig. 1a and 1b.

The average molecular weight of various copolymer samples studied was determined with a Ubbelohde viscometer in toluene at  $32 \pm 0.1^\circ\text{C}$  using the relation

$$\eta_{\text{int}} = K\bar{M}_v^\alpha$$

The values of  $K$  and  $\alpha$  are  $13.2 \times 10^{-5}$  and 0.71, respectively [12]. The NMR spectrum of the copolymer was taken on Perkin Elmer 599-B spectrometer using CCl<sub>4</sub> as solvent and tetramethyl silane as internal reference.

### Preparation of Film Solution

In order to prepare film in bulk, varying concentrations of PDSY-HgCl<sub>2</sub> complex were dissolved in [methylmethacrylate] ( $4.5 \text{ mol L}^{-1}$ ) and [Styrene] ( $4.32 \text{ mol L}^{-1}$ ) was added. The polymerization was performed in a test tube under an inert atmosphere of nitrogen gas at  $85 \pm 0.1^\circ\text{C}$  for 4 h.

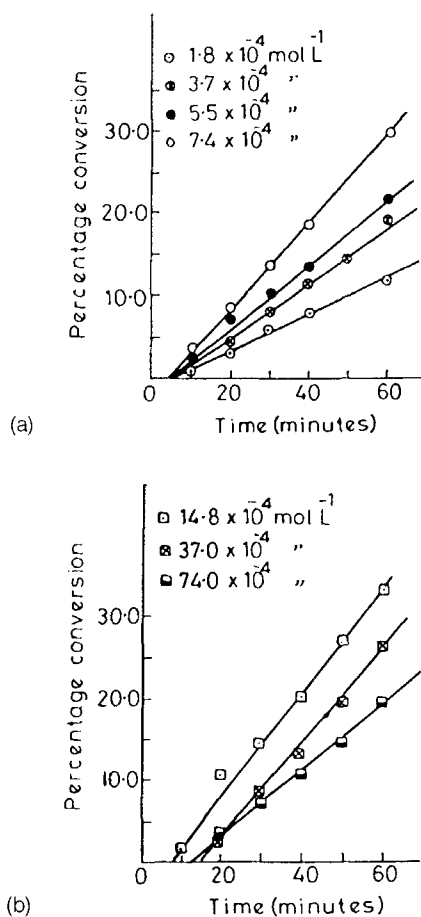


FIG. 1. Time-conversion profiles in the copolymerization of MMA with styrene initiated by [PDSY-HgCl<sub>2</sub> complex] in dioxane. (a) [PDSY-HgCl<sub>2</sub> complex] × 10<sup>4</sup> = 1.8 mol L<sup>-1</sup>; ⊗ 3.7 mol L<sup>-1</sup>; ● 5.5 mol L<sup>-1</sup>; ○ 7.4 mol L<sup>-1</sup>. (b) [PDSY-HgCl<sub>2</sub> complex] × 10<sup>4</sup> = 14.8 mol L<sup>-1</sup>; ⊗ 37.0 mol L<sup>-1</sup>; □ 74.0 mol L<sup>-1</sup>. [Sty] = 1.15 mol L<sup>-1</sup>, [MMA] = 1.2 mol L<sup>-1</sup>, temperature = 85 ± 0.1°C, time = 1 h.

### Preparation of Film

The glass plates used for casting were treated with a mixture of sulfuric acid (H<sub>2</sub>SO<sub>4</sub>) and potassium dichromate (K<sub>2</sub>Cr<sub>2</sub>O<sub>7</sub>) and then washed and rinsed with distilled water and dried in the oven at 60°C for 1 h. The film was then cast by pouring the film solution on the clean dry glass plate and tilting it back and forth to spread the solution onto the glass plate uniformly; the film was kept at room temperature.

### Permeation Experiment

In order to investigate the water permeability through the film, the end of a measuring cylinder, containing saturated sugar solution, was closed with the film.

This cylinder was then placed in a beaker containing potassium permanganate solution and this system was left for 24 h.

### Solubility and Chemical Resistance

Film strips (about 50 mg in weight) were immersed in various organic and inorganic solvents for 7 days. The change in weight was noted.

### Adsorption Experiments

In order to study the phenomenon of absorption, film strips (about 50 mg in weight) were immersed in various liquids (dimethyl sulfoxide, nitric acid, and acetic acid) at 40°C. After a relatively short time, the increase in weight due to absorption was noted.

## RESULTS AND DISCUSSION

The copolymerization of methylmethacrylate with styrene in dioxane has been carried out at 85°C by varying the [PDSY-HgCl<sub>2</sub>] from  $1.8 \times 10^{-4}$  to  $74.0 \times 10^{-4}$  mol L<sup>-1</sup>, [Styrene] 0.58 to 2.30 mol L<sup>-1</sup>, and [MMA] 0.6 to 2.4 mol L<sup>-1</sup>. The polymerization runs were associated with a short induction period of 4–15 min. The synthesis of copolymer film (bulk) was also carried out at 85°C by varying [PDSY-HgCl<sub>2</sub>] ( $1.11$ – $3.33 \times 10^{-3}$  mol L<sup>-1</sup>), [MMA] (3.0–6.0 mol L<sup>-1</sup>), and [Sty] (2.88–5.76 mol L<sup>-1</sup>). The results are compiled in Tables 1–6 and Figs. 1–10.

### Effect of [PDSY-HgCl<sub>2</sub>]

The rate of copolymerization ( $R_p$ ) increases consistently with increase in [PDSY-HgCl<sub>2</sub>] from 1.8 to  $24.8 \times 10^{-4}$  mol L<sup>-1</sup> and then decreases with increasing [PDSY-HgCl<sub>2</sub>] up to  $74.0 \times 10^{-4}$  mol L<sup>-1</sup> (Table 1).

TABLE 1. Effect of [PDSY-HgCl<sub>2</sub>] on Polymerization of Methylmethacrylate with Styrene

Run no.	[PDSY + HgCl <sub>2</sub> ] × 10 <sup>4</sup> (mol L <sup>-1</sup> )	Percentage conversion before treatment	Percentage conversion after acetonitrile treatment	Percentage conversion after cyclohexane treatment	$R_p \times 10^5$ (mol L <sup>-1</sup> s <sup>-1</sup> )	$M\bar{v}$
1	1.8	15.75	14.50	14.36	9.79	54,830
2	3.7	23.49	20.87	20.0	13.0	43,550
3	5.5	25.04	22.16	22.06	15.66	32,880
4	7.4	35.40	30.40	29.86	19.58	28,050
5	14.8	39.16	35.50	35.10	22.83	16,440
6	37.0	29.50	26.92	26.6	17.13	9,016
7	74.0	24.54	23.79	22.6	16.31	3,126

[MMA] = 1.2 mol L<sup>-1</sup>; [Sty] = 1.15 mol L<sup>-1</sup>; temperature = 85 ± 0.1°C; time = 1 h.

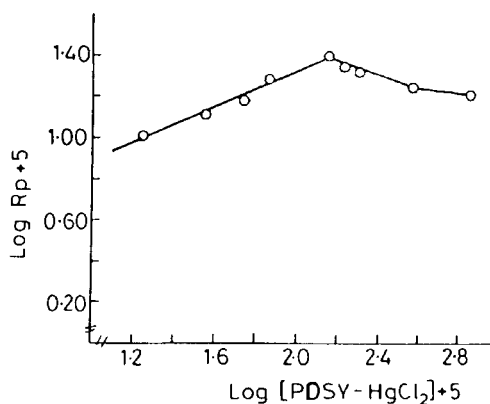


FIG. 2. Log rate of polymerization as a function of log concentration of initiator, in the copolymerization of MMA with styrene, initiated by [PDSY-HgCl<sub>2</sub> complex] at 85 ± 0.1°C in dioxane.

The initiator exponent value, calculated from the slope of the plot of log  $R_p$  against log[PDSY-HgCl<sub>2</sub>] is 0.5, as expected for radical polymerization (Fig. 2).

The average molecular weight ( $\bar{M}_v$ ) of copolymers decreases with increase in [PDSY-HgCl<sub>2</sub>].

The value of  $k_p^2/k_t$  determined from the slope of the plot of  $1/\bar{M}_v$  versus  $R_p/[M]^2$  is  $8.6 \times 10^{-4} \text{ L mol}^{-1}\text{s}^{-1}$  (Fig. 3).

### Effect of [Methylmethacrylate]

The effect of [MMA] on  $R_p$  was studied by varying MMA concentration from 0.6 to 2.4 mol L<sup>-1</sup> while [Styrene] and [PDSY-HgCl<sub>2</sub>] were kept constant. As shown in Table 2,  $R_p$  is a direct function of MMA concentration. A plot of log  $R_p$  versus log[MMA] is linear (Fig. 4) and the monomer exponent value is unity, as expected for ideal radical polymerization. The average molecular weight of the copolymer also increases with MMA concentration.

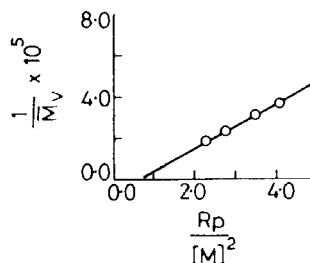


FIG. 3. Plot of average molecular weight as a function at  $R_p/[M]^2$  in the copolymerization of MMA with styrene initiated by [PDSY-HgCl<sub>2</sub> complex] at 85 ± 0.1°C in dioxane.

TABLE 2. Effect of [MMA] on Copolymerization of Methylmethacrylate with Styrene

Run no.	[MMA] (mol L <sup>-1</sup> )	Percentage conversion before treatment	Percentage conversion after acetonitrile treatment	Percentage conversion after cyclohexane treatment	$R_p \times 10^5$ (mol L <sup>-1</sup> s <sup>-1</sup> )	$M\bar{v}$
1	0.6	24.80	23.7	23.2	9.72	9,716
2	1.2	39.16	35.50	35.1	22.83	16,440
3	1.8	46.2	41.50	40.4	30.7	46,030
4	2.4	48.0	44.22	43.7	39.4	69,020

[PDSY-HgCl<sub>2</sub>] =  $14.8 \times 10^{-4}$  mol L<sup>-1</sup>; [Styrene] = 1.15 mol L<sup>-1</sup>; temperature =  $85 \pm 0.1^\circ\text{C}$ ; time = 1 h.

### Effect of [Styrene]

The effect of [Styrene] on  $R_p$  was studied by varying [Styrene] from 0.58 to 2.30 mol L<sup>-1</sup> while [MMA] (1.2 mol L<sup>-1</sup>) and ([PDSY-HgCl<sub>2</sub>] ( $14.8 \times 10^{-4}$  mol L<sup>-1</sup>) were kept constant (Table 3). It is observed that [Styrene] directly affects the  $R_p$ . The order of reaction with respect to styrene concentration is 1.0 (Fig. 5), as expected for radical polymerization. The average molecular weight increases with styrene concentration.

The monomer and initiator exponent values suggest the radical mode of polymerization, which was further confirmed by the line passing through the origin (Fig. 6).

### Effect of [Additives]

The effect of hydroquinone on copolymerization was also studied. The polymerization was retarded in the presence of hydroquinone. Addition of benzene also retarded the rate of reaction, whereas addition of dimethyl sulfoxide (DMSO)

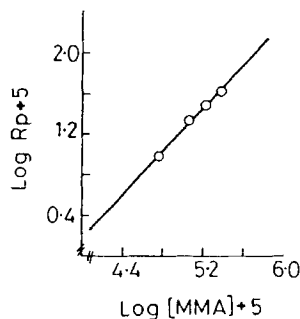


FIG. 4. Log rate of polymerization as a function of MMA in the polymerization of MMA, with styrene initiated by [PDSY-HgCl<sub>2</sub> complex] at  $85 \pm 0.1^\circ\text{C}$  in dioxane.

TABLE 3. Effect of [Styrene] on Copolymerization of Styrene with Methylmethacrylate Initiated by PDSY-HgCl<sub>2</sub> Complex

Run no.	[Sty] (mol L <sup>-1</sup> )	Percentage conversion before treatment	Percentage conversion after acetonitrile treatment	Percentage conversion after cyclohexane treatment	$R_p \times 10^5$ (mol L <sup>-1</sup> s <sup>-1</sup> )	$M\bar{v}$
1	0.58	26.35	24.97	24.62	12.13	5,846
2	1.15	39.15	35.50	35.10	22.83	16,440
3	1.73	41.20	39.26	38.20	26.63	53,330
4	2.30	42.0	41.34	40.26	35.0	58,460

[PDSY-HgCl<sub>2</sub>] =  $14.8 \times 10^{-4}$  mol L<sup>-1</sup>; [MMA] = 1.2 mol L<sup>-1</sup>; temperature =  $85 \pm 0.1^\circ\text{C}$ ; time = 1 h.

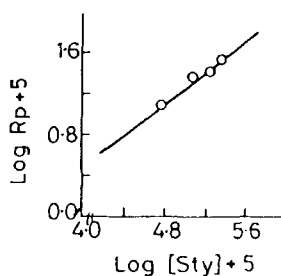


FIG. 5. Log rate of polymerization as a function of styrene in the copolymerization of MMA with styrene initiated by [PDSY-HgCl<sub>2</sub> complex] at  $85 \pm 0.1^\circ\text{C}$  in dioxane.

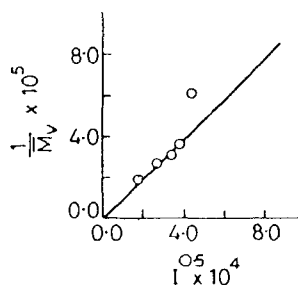


FIG. 6. Plot of average molecular weight versus square root of initiator concentration in the copolymerization of MMA with styrene initiated by [PDSY-HgCl<sub>2</sub>] at  $85 \pm 0.1^\circ\text{C}$  in dioxane.



TABLE 4. Effect of [Additives] on Copolymerization of Methylmethacrylate with Styrene

Additive	[Additives] (mol L <sup>-1</sup> )	Percentage conversion before treatment	Percentage conversion after acetonitrile treatment	Percentage conversion after cyclohexane treatment	$R_p \times 10^5$ (mol L <sup>-1</sup> s <sup>-1</sup> )
	0	39.16	35.50	35.10	22.83
Hydroquinone	$9 \times 10^{-2}$	35.86	24.79	24.23	15.29
Benzene	1.3	24.26	23.98	23.72	16.78
	2.7	22.29	20.24	19.67	14.68
DMSO	1.64	40.28	37.64	37.30	23.29
	3.23	43.54	39.87	39.26	25.84

[PDSY-HgCl<sub>2</sub>] =  $14.8 \times 10^{-4}$  mol L<sup>-1</sup>; [Styrene] = 1.15 mol L<sup>-1</sup>; [MMA] = 1.2 mol L<sup>-1</sup>; temperature =  $85 \pm 0.1^\circ\text{C}$ ; time = 1 h.

increased the rate of reaction (Table 4). DMSO, being more polar than dioxane, facilitated the dissociation of metal ylide generating a free radical; hence an accelerating effect was observed. Benzene, being nonpolar, may suppress dissociation of metal ylide; hence retardation was observed.

### Effect of Temperature

The energy of activation ( $\Delta E$ ) was determined at three different temperatures: 80, 85, and  $90^\circ\text{C}$ .  $R_p$  is a direct function of temperature, and the apparent activation energy is  $48.0 \text{ kJ mol}^{-1}$  (Fig. 7). This value is in good agreement with reported values for radical copolymerization of vinyl monomers [13, 14].

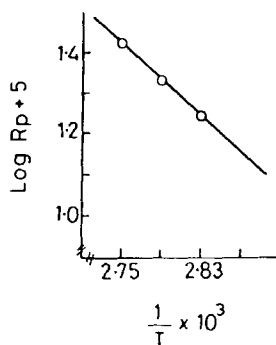


FIG. 7. Arrhenius plot of  $\log R_p$  versus  $\frac{1}{T} \times 10^3$ .

### Characterization of Copolymers

Elemental analysis of the polymers did not detect sulfur, suggesting that the metal ylide complex was not incorporated in the copolymer.

### NMR of Copolymer

Figure 8 shows the NMR spectra of copolymer. The phenyl protons appear at  $2.1\tau$ – $2.6\tau$ , the methyl protons at  $8.2\tau$ – $9.0\tau$ , and the methoxy protons at  $6.2\tau$ . There is slight shifting in the position of methoxy protons. The appearance of methoxy protons as a single peak shows that the copolymer is random in nature.

### Mechanism

Phenacyldimethyl sulfonium ylide as well as mercuric chloride failed to initiate the polymerization of MMA and styrene, but the PDSY-HgCl<sub>2</sub> complex is able to initiate it. It has been already reported [11] that the sulfur ylide has two coordination sites ( $>C=O$ ,  $-CH$ ) for complex formation, and the coordination of HgCl<sub>2</sub> involves methine carbon. Therefore, the metal ylide complex has only one coordination site remaining. Since ylide fails to initiate the polymerization of MMA, it is clear that site ( $>C=O$ ) is not capable of forming complex with monomer. It seems reasonable that the electron-deficient monomer (MMA) may form a complex with the metal ylide complex having the following structure (II).

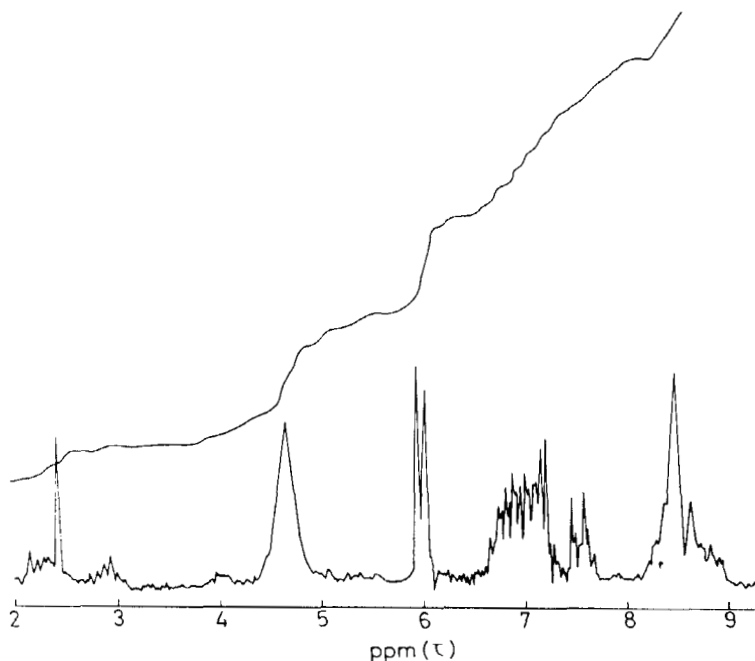
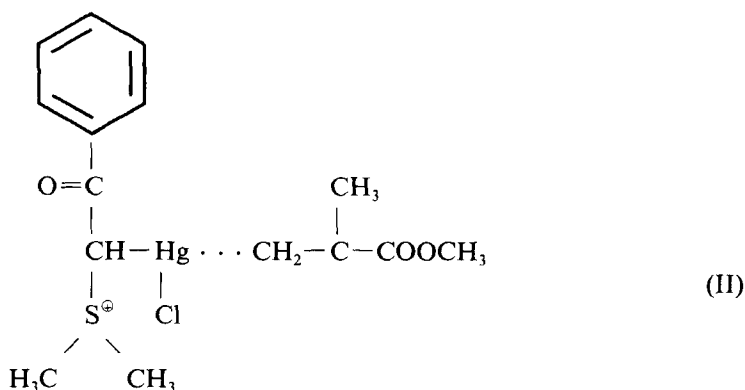


FIG. 8. NMR spectrum of copolymer of MMA with styrene initiated by [PDSY-HgCl<sub>2</sub> complex] at  $85 \pm 0.1^\circ\text{C}$  in dioxane.



The ESR spectrum (Fig. 9) of the reaction mixture suggests that  $H^{\cdot}$  is responsible for initiating activity. The gyromagnetic ratio calculated from Fig. 9 is 1.968, which is very close to the value given for  $H^{\cdot}$  ( $g = 2.0031 \pm 0.031$ ).

There are three possible ways by which the metal ylide monomer complex can dissociate to yield  $H^{\cdot}$  radical (see page 367).

An examination of the above reaction scheme reveals that there are three possible ways by which hydrogen atom is produced which bring about polymerization. The formation of  $H^{\cdot}$  atom is confirmed by ESR spectrum (Fig. 9) and the value of  $g$  is 1.968, which is close to the reported value. An examination of other radicals ( $X$ ,  $Y$ , and  $Z$ ) that may be formed by route 1, 2, and 3, respectively, reveals that the radical  $X$  (having no hydrogen atoms) attached to C is least stable and radical  $Z$  (having two  $H^{\cdot}$  atoms attached to C) is maximally stable owing to hyperconjugation.  $Y$  being between them, it is concluded that the complex is likely to dissociate via route 3 to produce  $H^{\cdot}$  radical responsible for polymerization.

### Properties of Copolymer Film

Copolymer film is colorless and transparent. Water permeability of the film was investigated at room temperature. The films prepared under different concentrations of initiator and monomer ( $PF_1$  to  $PF_5$ ) in Table 5 were impermeable to water.

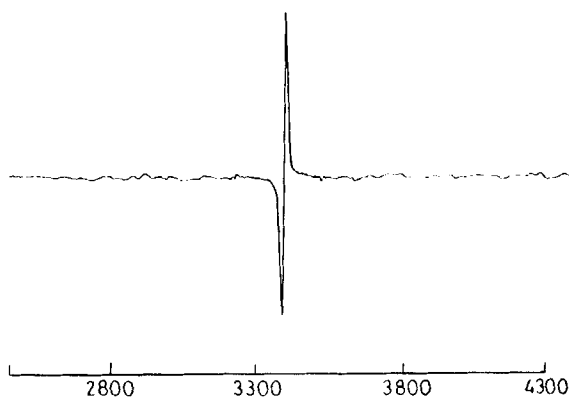


FIG. 9. ESR spectrum of reaction mixture of copolymer solution of MMA with styrene at  $85 \pm 0.1^\circ\text{C}$  initiated by [PDSY-HgCl<sub>2</sub> complex].

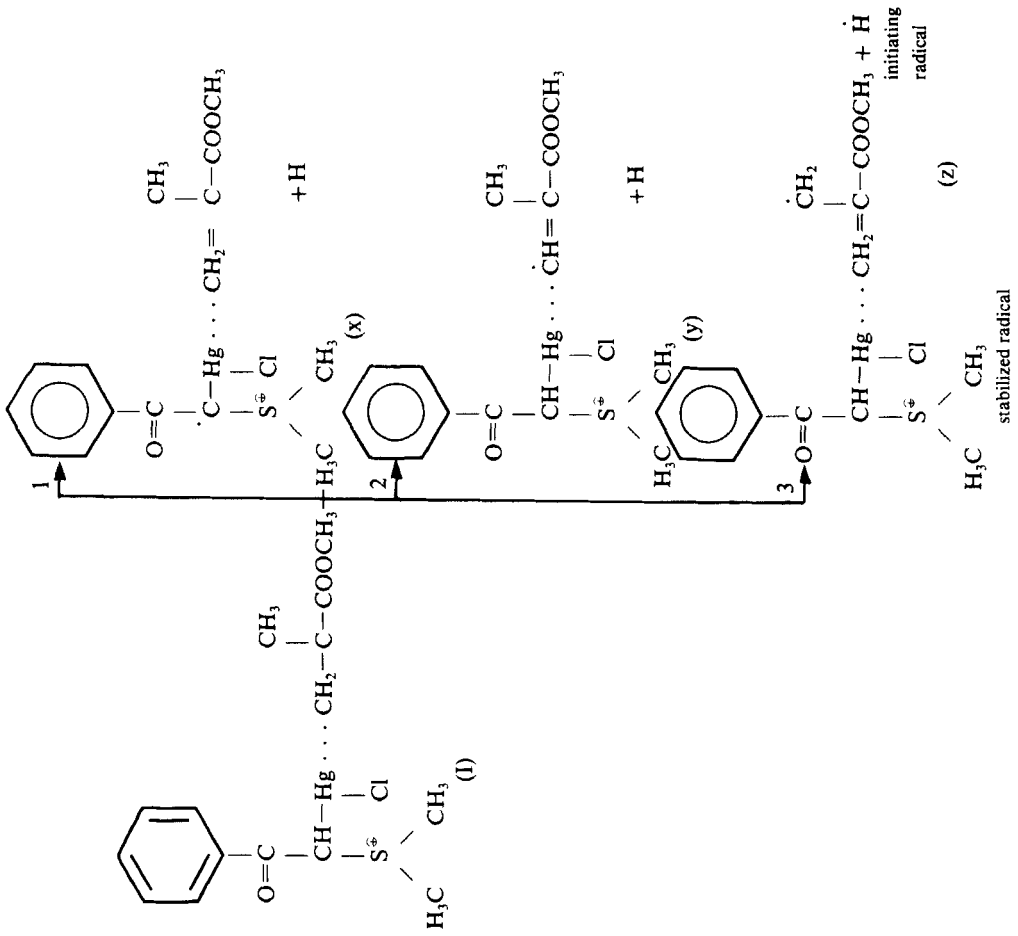


TABLE 5. Composition of Reactant Used in Preparation of Copolymer Film in Presence of PDSY-HgCl<sub>2</sub> Complex

Polymer film code	Component				
	[PDSY-HgCl <sub>2</sub> ] (mol L <sup>-1</sup> × 10 <sup>3</sup> )	[MMA] (mol L <sup>-1</sup> )	[Sty] (mol L <sup>-1</sup> )	Time (min)	Temp. (°C)
PF1	1.11	4.5	4.32	240	85 ± 0.1
PF2	2.22	4.5	4.32	240	85 ± 0.1
PF3	3.33	4.5	4.32	240	85 ± 0.1
PF4	2.22	6.0	2.88	240	85 ± 0.1
PF5	2.22	3.0	5.76	240	85 ± 0.1

The effects of various inorganic and organic solvents were used to check the solubility and chemical resistance of the film. These data showed that the film was insoluble in strong bases and distilled water but soluble in most of the organic solvents (Table 6).

Acetic acid, nitric acid, and dimethyl sulfoxide were used as test liquids. The

TABLE 6. Solubility of Copolymer Film in Presence of PDSY-HgCl<sub>2</sub> Complex

S. no.	Solvents	Polymer film		
		PF1	PF2	PF4
1	H <sub>2</sub> SO <sub>4</sub>	- +	- +	- +
2	HCl (N/10)	- +	- +	- +
3	HNO <sub>3</sub>	--	--	--
4	CH <sub>3</sub> COOH (25%)	--	--	--
5	NaOH (N/25)	--	--	--
6	NH <sub>4</sub> OH	--	--	--
7	Benzene	+ +	+ +	+ +
8	Toluene	+ +	+ +	+ +
9	DMF	+ -	+ -	+ -
10	DMSO	--	--	--
11	Dioxane	+ +	+ +	+ +
12	CH <sub>3</sub> OH	- +	- +	- +
13	Diethylether	+ -	+ -	+ -
14	Dichloromethane	+ +	+ +	+ +
15	Cyclohexane	- +	- +	- +
16	Acetone	+ +	+ +	+ +
17	THF	+ +	+ +	+ +
18	Water	- +	- +	- +

+ + = soluble at room temperature; + - = sparingly soluble at room temperature; -- = insoluble; - + = sparingly soluble on heating.

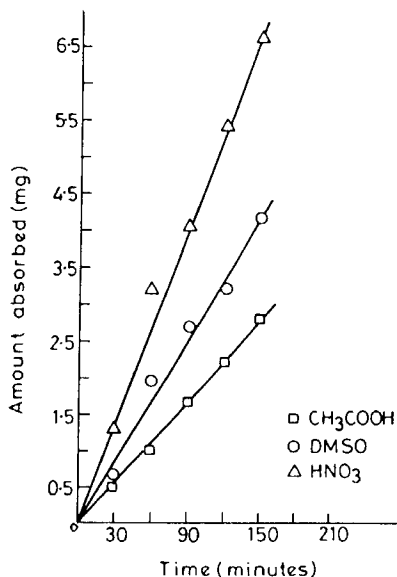


FIG. 10. Graph showing amount of different solvents absorbed in mg versus time (minutes).  $\square$   $\text{CH}_3\text{COOH}$ ;  $\circ$  DMSO;  $\triangle$   $\text{HNO}_3$ .

composition of the film is shown in Table 5. As shown in Fig. 10, the absorption of nitric acid in film was highest, while dimethyl sulfoxide also exhibited high absorption, but the absorption of acetic acid was relatively low.

### CONCLUSION

From these studies we infer that copolymerization of methylmethacrylate with styrene follows ideal kinetics. The average molecular weight of copolymer varies inversely to the [initiator] but directly to the [monomer].

The copolymer film prepared in bulk was water impermeable and had higher absorption property for nitric acid; absorption was lowest in acetic acid.

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