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NEW POLYMER SYNTHESES. XIII. SYNTHESIS AND CHARACTERIZATION OF NOVEL POLYKETONES VIA FRIEDEL-CRAFTS REACTION AND BASED ON DIARYLIDENE DERIVATIVES

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NEW POLYMER SYNTHESES. XIII. SYNTHESIS AND CHARACTERIZATION OF NOVEL POLYKETONES VIA FRIEDEL-CRAFTS REACTION AND BASED ON DIARYLIDENE DERIVATIVES

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ABSTRACT

An interesting new class of polyketones based on diarylidene derivatives were synthesized by Friedel-Crafts reaction of 2,7-dibenzylidenecycloheptanone (**I**) and dibenzylideneacetone (**II**) monomers, with different diacid chlorids including: isophthalic, terephthalic, 3,3′-azodibenzoic, 4,4′-azodibenzoic, biphenic, adipic, sebacic and oxalic diacids in dry dichloromethane solvent and $AlCl_3$ catalyst. The model compounds were synthesized by reacting **I** and **II** with benzoyl chloride and characterized by ^{1}H-NMR, IR and elemental analyses. The polyketones were insoluble in most organic solvents but dissolved easily in concentrated sulfuric acid. The thermal properties of these polymers were evaluated and correlated to their structural units by TGA, DTG measurements and had inherent viscosity up to 0.36–0.84 dl/g. The crystallinity of some polymers were tested by X-ray analysis. The electronic spectra of selected examples were measured in DMSO solution and the morphological properties were detected by SEM.

Key Words: Polyketones; Arylidene polymers, Synthesis; Characterization; Friedel-Crafts

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INTRODUCTION

Polyketones represent a new class of potential engineering thermoplastics. Their property set is positioned in the area of polyamides and polyacetales. They are characterized by their stiffness, high melting temperature and resistance against water and moisture. In particular, one should also mention chemical resistance, stability at hydrolysis conditions, impact toughness, flame resistance without additives containing halides or red phosphor, and short cycle times in injection molding [1–3].

Friedel-Crafts polymers formed on polycondensation of 4,4′-dichloromethyldiphenyl ether (DDE) with benzen, toluene, isomeric chlorotoluenes and xylenes, phenol and isomeric cresols have been reported [4–6].

Recently, a considerable amount of research effort has concentrated on the synthesis of polyketones via Friedel-Crafts reaction [7–12].

In continuation of our interest in the synthesis and characterization of some new polyketones based on diarylidenecycloalkanones [13], the work reported here is on the synthesis and characterization of some new polyketones based on diarylidenecycloheptanone or diarylideneacetone. The major aim of this work has been to investigate the effect of inclusion of cycloheptanone or acetone moieties, as well as the aromatic and aliphatic chains on the polymer properties. In addition, other characteristics, of these new polymers such as thermostability, solubility, morphology and crystallinity, are discussed.

EXPERIMENTAL

Measurements

Melting points were determined on a Perkin-Elmer 240°C electrothermal melting point apparatus and are uncorrected. Infrared spectra were recorded on a Schimadzu 2110 PC spectrophotometer with KBr pellets. The ^{1}H-NMR spectra were recorded on a GNM-LA 400 MHz NMR spectrophotometer at room temperature in DMSO or $CHCl_3$ using TMS as the internal reference. Viscosity measurements were made with 0.5% (w/v) solution of polymers in H_2SO_4 at 25°C using an Ubbelhode suspended level viscometer. The X-ray diffractograms of the polymers were obtained with a Philips X-ray unit (Philips generator Pw-1710) and Ni-Filtered CuK_α radiations. TGA and DTG measurements were performed on V 5.1A Du Pont 2000 thermal analyzer at a heating rate of 10°C/min in air. The solubility of the polymers were determined using 0.02 g of polymer in 3–5 mL of solvent. The morphology of the polymers were examined by scanning electronic microscopy (SEM) using a Jeol JSM-5400 LV-ESM.

REAGENTS AND SOLVENTS

Benzaldehyde from (Aldrich) was used as purchased. Terephthaloyl chloride (Aldrich) was recrystallized from n-hexane (m.p 83–84°C), isophthaloyl chloride (Aldrich) was recrystallized from n-hexane (m.p 44°C). Adipoyl and sebacoyl dichlorides [14] freshly distilled at 125°C/11 Torr, and at 182°C/16 Torr, respectively were used. Oxaloyl chloride was obtained from Aldrich and used immediately without purification. 4,4′-Diphenyl dichloride was prepared according to the literature [15]. 4,4′-Azobenzenedibenzoyl chloride and 3,3′-azobenzenedibenzoyl chloride were prepared according to the method of Tomlinson [16]. Acetone, cycloheptanone from (Merck) and benzoyl chloride from (BDH) were used as purchased. Anhydrous aluminum chloride (Merck) and all other solvents were of high purity and were further purified by standard methods [15].

Monomer Synthesis

2,7-Dibenzylidenecycloheptanone I

A mixture of 0.1 mol cycloheptanone and 0.2 mol of benzaldehyde in warm ethanol was stirred, and treated with few milliliters of a 10% solution of alcoholic sodium hydroxide. The reaction mixture was stirred at room temperature for 1 hour, and the resulting solid was collected by filtration, washed with cold water, dried and recrystallized from ethanol, as yellow needles; yield 70%, m.p 105°C (lit 17 m.p 106°C). Calculated for $C_{21}H_{20}O$: C, 87.50; H, 6.94%. Found: C, 87.98; H, 6.12%. IR (KBr, cm^{-1}): at 1600 (s, C=C), at 1660 (s, C=O). ^{1}H-NMR (DMSO-d_6, ppm), 7.0–8.0 (m, 10H of Ar-H and 2H of 2CH=C); 2.5 (s, 4H of $2CH_2$ cycloheptanone); 2.0 (s, 4H of middle $2CH_2$ of cycloheptanone).

Dibenzylideneacetone II

This compound was prepared from the condensation of 0.2 mol of benzaldehyde with 0.1 mol of acetone in ethanol as described in previous method. The final product was recrystallized from ethanol as yellow needles, yield 95%, m.p 112°C (lit 18, m.p 111°C). Calculated for $C_{17}H_{14}O$: C, 87.17; H, 5.98%. Found: C, 87.34; H, 5.21%. IR (KBr, cm^{-1}): at 1580 (s, C=C), at 1640 (s, C=O). ^{1}H-NMR (DMSO-d_6, ppm), 7.0–8.0 (m, 10H of Ar-H and 4H of 2CH=CH).

Preparation of Model Compounds III, IV

General Procedure

In a 100 mL round-bottomed flask equipped with a magnetic stirrer, and a nitrogen inlet and outlet, was placed a solution of 1 mmol 2,7-dibenzylidenecycloheptanone **I** or dibenzylideneacetone **II** and 2 mmol of benzoyl chloride in 40 mL of dry dichloromethane. The flask was purged with nitrogen while stirring and 20 mmol of anhydrous aluminum chloride was added portionwise to the solution. The solution was stirred for 24 hours at 25°C, and the separated solid product was filtered off washed with water dried *in vacuo* at room temperature.

The individual model compounds were as follows:

2,7-*bis*(4-Benzoylbenzylidene)cycloheptanone III

An analytical sample was obtained by recrystallization from benzene in yellow needles, yield 75%, m.p 220°C. Calculated for $C_{35}H_{28}O_3$: C, 84.67; H, 5.64%. Found: C, 84.13; H, 5.04%. IR (KBr, cm^{-1}): at 1665 (C=O of cycloheptanone), at 1700 (C=O of benzoyl group), at 1600 (C=C group). ^{1}H-NMR (DMSO-d_6, ppm), 7.2–8.5 (m, 18H, Ar-H and 2H of 2CH=C), 2.5 (s, 4H of $2CH_2$ of cycloheptanone), 2.0 (s, 4H of middle $2CH_2$ of cycloheptanone).

bis(4-Benzoylbenzylidene)acetone IV

A pure sample was obtained by recrystallization from benzene in yellow needles, yield 70%, m.p 240°C. Calculated for $C_{31}H_{22}O_3$: C, 84.16; H, 4.97%. Found: C, 84.97; H, 4.09%. IR (KBr, cm^{-1}): at 1650 (C=O of acetone), at 1700 (C=O of ketone group), 1600 (C=C group), ^{1}H-NMR (DMSO-d_6), 7.4–8.5 (m, 18H, Ar-H and 4H of 2CH=CH).

Polymer Synthesis

Friedel-Crafts method was applied for the preparation of all the polymers. Typically, in a 100-mL round-bottomed flask equipped with a magnetic stirrer and, nitrogen inlet and outlet, was placed a solution of 1.728 (6 mmol), 2,7-dibenzyl-idenecy-cloheptanone I and 1.218 g (6 mmol) of isophthaloyl chloride in 25 mL of dry dichloromethane. The flask was purged with nitrogen while stirring, and 2.66 g (20 mmol) of aluminum chloride was added during the nitrogen flash. The reddish brown reaction mixture was stirred for 24 hours, then it was filtered off and the separated product was titrated three times with a large excess of methanol. The powdery material was again filtered off, washing with water, methanol, acetone and dried under reduced

Table 1. Elemental Analysis, Inherent Viscosity, Yield, and Color of Polymers $\mathbf{V_{a-h}}$

Polymer Code	C%		H%		N%		η Inh (dl/g)	Yield %	Appearance of Polymer
	Calcd.	Found	Calcd.	Found	Calcd.	Found			
$\mathbf{V_a}$	83.25	82.16	5.26	5.09	–	–	0.78	85	Powder yellowish
$\mathbf{V_b}$	83.25	81.34	5.26	4.13	–	–	–	77	Powder yellowish
$\mathbf{V_c}$	85.02	84.76	5.26	4.65	–	–	0.84	84	Powder brown
$\mathbf{V_d}$	80.45	80.01	4.98	4.11	5.36	4.32	–	88	Powder brown
$\mathbf{V_e}$	80.45	82.21	4.98	3.53	5.36	5.98	0.36	86	Powder brown
$\mathbf{V_f}$	80.70	81.24	5.26	4.28	–	–	–	81	Powder yellowish
$\mathbf{V_g}$	81.40	82.14	6.53	7.35	–	–	0.70	87	Powder yellowish
$\mathbf{V_h}$	81.93	80.71	7.48	7.29	–	–	–	76	Powder yellowish

η Inherent viscosity measured in H_2SO_4 at 25°C.

Table 2. Elemental Analysis, Inherent Viscosity, Yield, and Color of Polymers $\mathbf{VI_{a-h}}$

Polymer Code	C%		H%		N%		η Inh (dl/g)	Yield %	Appearance of Polymer
	Calcd.	Found	Calcd.	Found	Calcd.	Found			
$\mathbf{VI_a}$	82.41	81.87	4.39	5.24	–	–	–	90	Powder yellowish
$\mathbf{VI_b}$	82.41	83.25	4.39	4.93	–	–	0.63	90	Powder yellowish
$\mathbf{VI_c}$	84.54	84.84	4.54	3.07	–	–	–	78	Powder yellowish
$\mathbf{VI_d}$	79.48	81.02	4.27	3.41	5.98	4.13	0.59	83	Powder brown
$\mathbf{VI_e}$	79.48	80.45	4.27	5.16	5.98	4.78	–	84	Powder brown
$\mathbf{VI_f}$	79.16	79.88	4.16	4.79	–	–	0.48	89	Powder yellowish
$\mathbf{VI_g}$	80.23	81.49	5.81	5.23	–	–	–	78	Powder yellowish
$\mathbf{VI_h}$	81.00	82.05	7.00	7.63	–	–	0.52	80	Powder yellowish

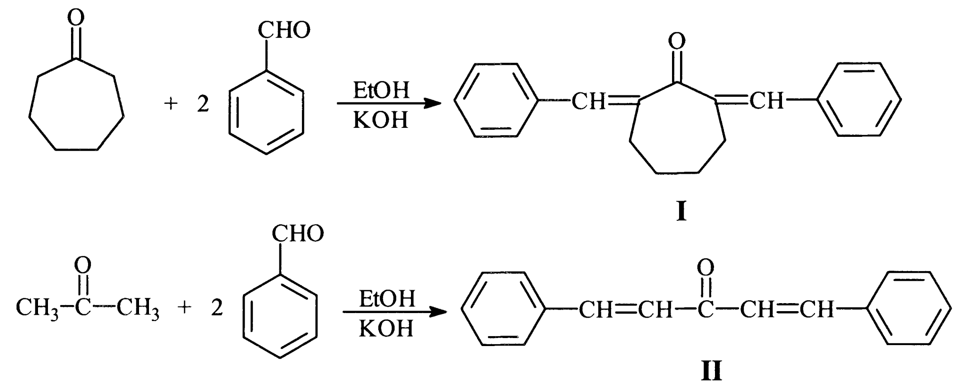
η Inherent viscosity measured in H_2SO_4 at 25°C.

pressure (1 mmHg) at 70°C for 2 days. This method was applied for the preparation of other polyketones; whose yields, elemental analyses, viscosity and colors are listed in Tables 1 and 2.

RESULTS AND DISCUSSION

Synthesis of Monomers I and II

Dibenzylidenecycloheptanone (**I**) and dibenzylideneacetone (**II**) monomers are synthesized in good yields by the base catalyzed condensation of two mols of benzaldehyde with one mol of cycloheptanone or acetone respectively, as reported in the literature [17, 18], (Sch. 1).



Scheme 1. Synthesis of monomers **I, II**.

Synthesis of Model Compounds

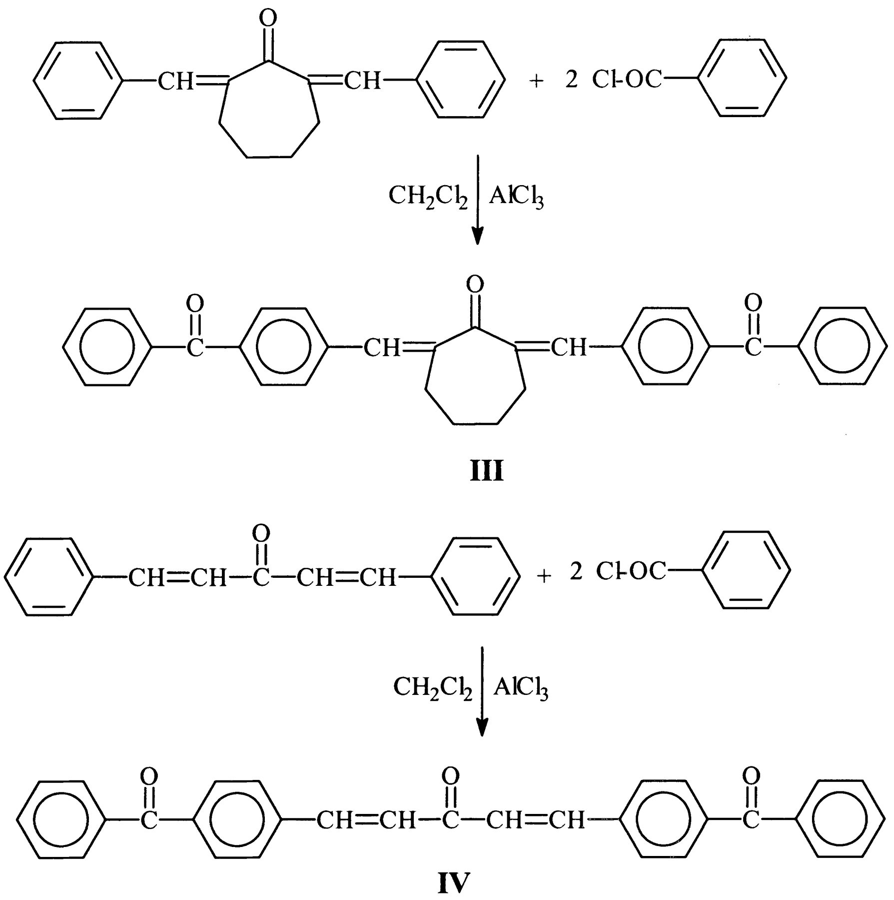
Before attempting the polymerization, model compounds were prepared by the reaction of dibenzylidene derivatives (**I, II**) with two equivalents benzoyl chloride. A typical reaction is depicted in Sch. 2.

The identities of these model compounds were confirmed by both elemental and spectral IR and ^{1}H-NMR data (Figs. 1, 2).

Synthesis of Polyketones

Two novel series of polyketones based on dibenzylidenecycloalkanone or dibenzylideneacetone in the polymer main chain, were prepared from reactions **I** and **II** with diacid chlorides via Friedel-Crafts reaction as shown in Sch. 3.

The polymerization reactions were carried out at room temperature in dichloromethane as a solvent and anhydrous aluminum chloride as a catalyst. The reaction times varied from 20–24 hours, and the polymers were immediately isolated when the reaction mixtures were poured into methanol/water mixture, with a yield in the range of 76–90%. These polymers were characterized by elemental analysis, IR spectroscopy, solubility, viscometry, thermal analysis and morphological properties. The elemental data of all the polymers coincided with the characteristic repeating units (Tables 1 and 2). Spectral analyses in (KBr disks) showed the appearance of new carbonyl absorption at 1690–1720 cm^{-1}, characteristic for the C=O of ketonic group besides the original C=O of cycloheptanone or acetone at 1680–1640 cm^{-1}, and at C=C at 1595–1610 cm^{-1}. Other characteristic absorption peaks for the rest of the groups in the molecules were also present (Fig. 3).



III

IV

Scheme 2. Synthesis of model compounds **III, IV**.

The solubility characteristics of polyketones ($\mathbf{V}_{a-h}$, $\mathbf{VI}_{a-h}$) were tested in various solvents (Table 4) including dimethylsulfoxide (DMSO), N, N-dimethylformamide (DMF), N-methylpyrrolidone (NMP), tetrahydrofuran (THF), methylene chloride, $CHCl_3$-acetone (1:1) and concentrated sulfuric acid. It was found that all the polymers dissolved readily in concentrated H_2SO_4 at room temperature giving a deep red color due to the unstability of the polymers. It can be clarified from Table 4, that the polyketones which are based on the cycloheptanone moiety ($\mathbf{V}_{a-h}$) soluble more than those which contain the acetone moiety ($\mathbf{VI}_{a-h}$). For halogenated solvents like CH_2Cl_2 and $CHCl_3$-acetone mixture, polymers $\mathbf{V}_{f-h}$ and $\mathbf{VI}_{f-h}$ were partially soluble. Comparing the solubility of polyketones containing 3,3-azo-linkage ($\mathbf{V}_e$, $\mathbf{VI}_e$) with those containing 4,4′-azo-linkage ($\mathbf{V}_d$, $\mathbf{VI}_d$), we found that the former possess better solubility than the later. This may be attributed to the close packing of the polymer chain in the latter polymer [19, 20]. More particu-

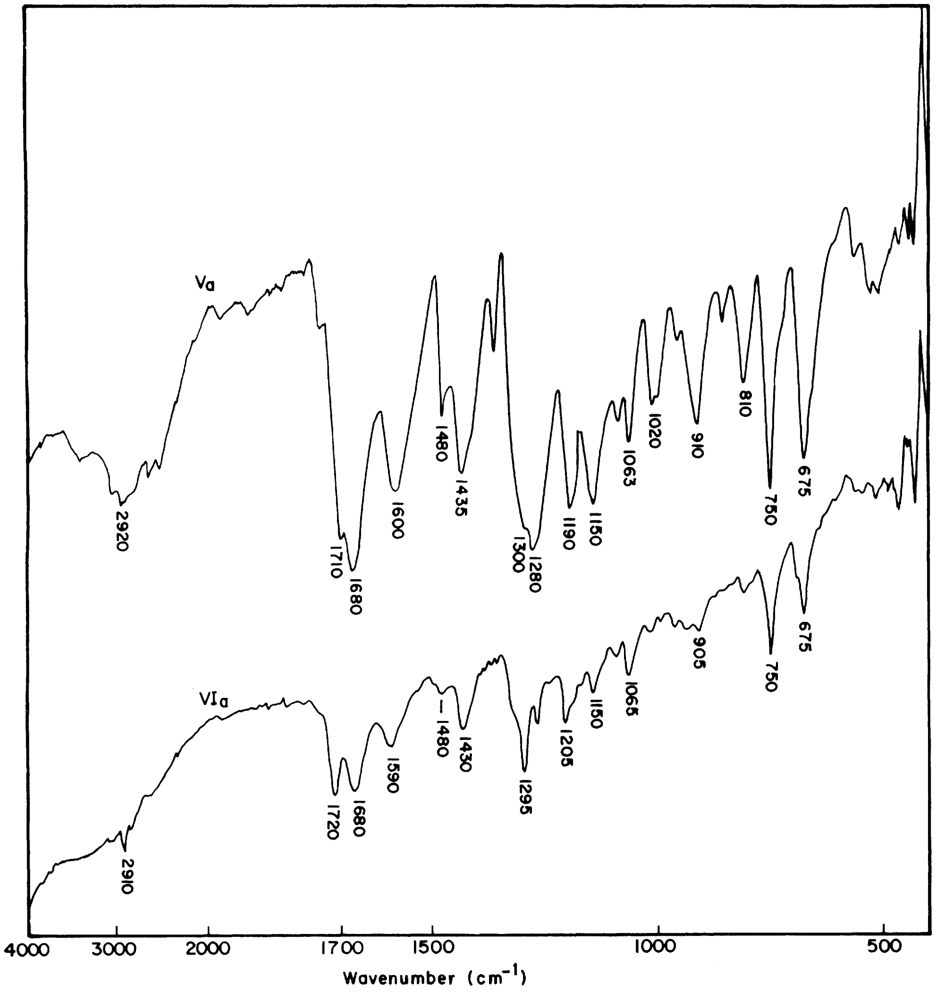


Figure 1. IR spectra of monomer II and model IV.

larly, polymers **V**$_h$ and **VI**$_h$, (with $n = 8$) showed good solubility in most organic solvents, due the high flexibility of these polymers through the polymethylene spacers.

X-ray diffractograms of selected examples of polyketones **V**$_b$, **V**$_g$, **VI**$_b$ and **VI**$_g$ in Fig. 4 showed few sharpness peaks with an amorphous background, in the region $2\theta = 5 - 60°$, this indicating that there is a large class of structure, in polymer main chains, are intermediate in the ordered states between crystals (with pronounced long-range order) in the arrangement of their atoms and molecules. Moreover, the presence of C=O as polar group in addition to high C=C band levels chains in the polymer, leading to some extent of crystallinity [21]. Also, the diffractogram, indicated that

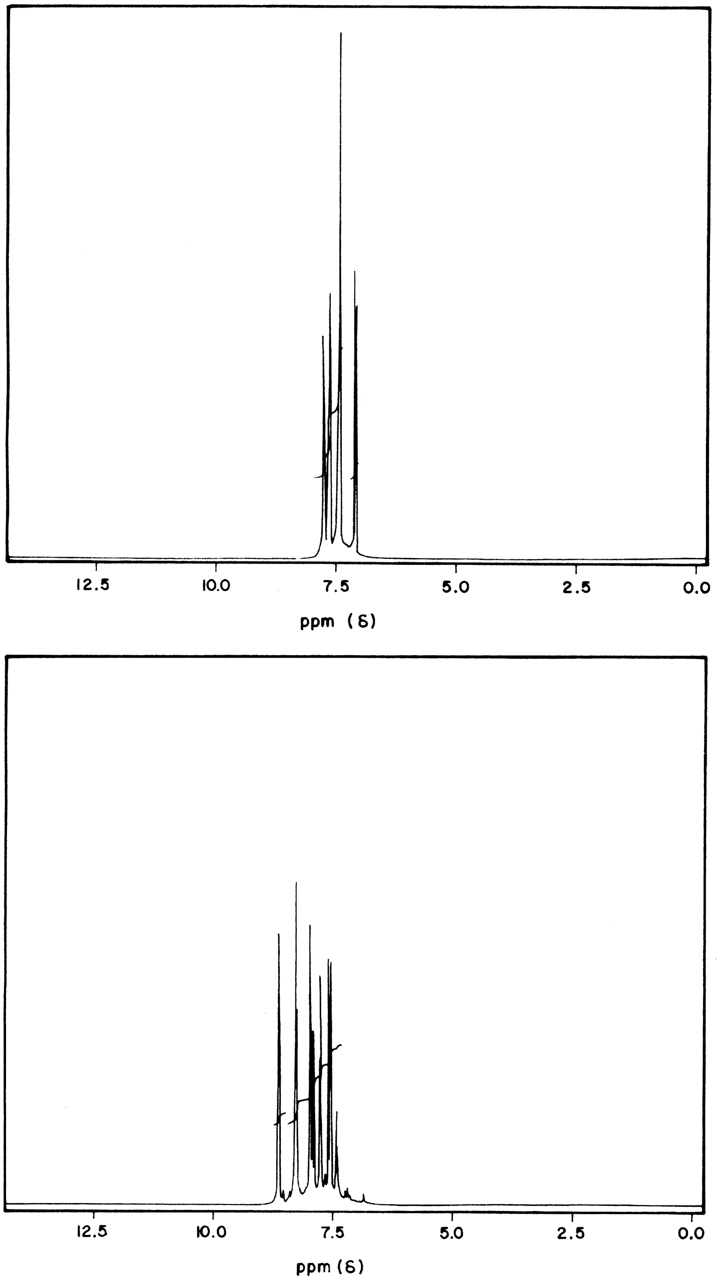
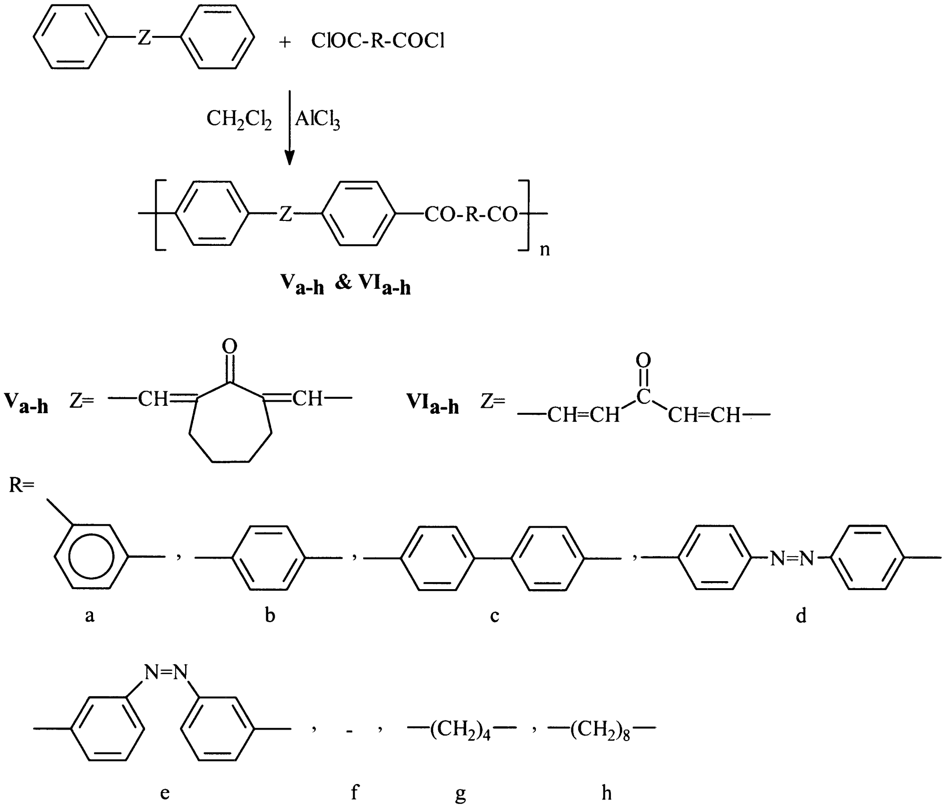


Figure 2. ^{1}H-NMR spectra of monomer II and model IV.



Scheme 3. Synthesis of polyketones $\mathbf{V}_{a-h}$, $\mathbf{VI}_{a-h}$.

Table 3. Solubility Characteristics of Polyketones $\mathbf{V}_{a-h}$

Polymer Code	DMSO	DMF	NMP	Chloroform Acetone	THF	Methylene Chloride	H_2SO_4
$\mathbf{V}_a$	±	±	±	±	±	±	+
$\mathbf{V}_b$	±	±	±	±	±	−	+
$\mathbf{V}_c$	±	±	±	±	±	−	+
$\mathbf{V}_d$	+	+	±	+	±	±	+
$\mathbf{V}_e$	±	±	±	±	±	±	+
$\mathbf{V}_f$	+	±	±	±	±	±	+
$\mathbf{V}_g$	+	+	±	+	±	+	+
$\mathbf{V}_h$	+	+	+	+	+	±	+

(+) Soluble at room temperature RT.
(±) Partially soluble at RT.
(−) Insoluble.

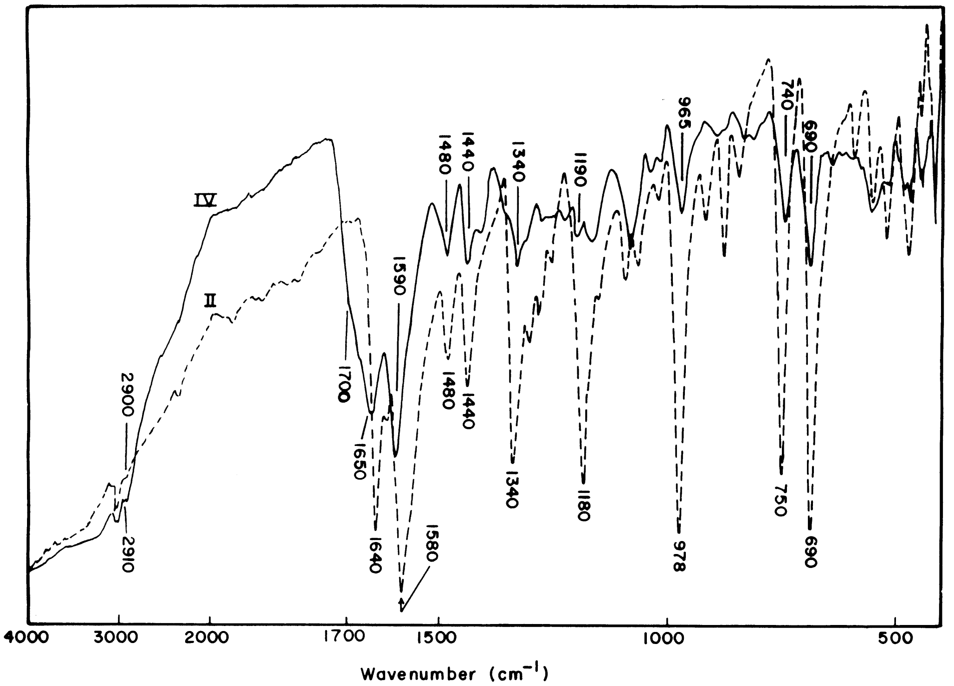


Figure 3. IR spectra of polyketones V_a, VI_a.

polyketones $\mathbf{V_g}$, $\mathbf{VI_g}$ has a high degree of crystallinity in comparison with polyketones $\mathbf{V_b}$, $\mathbf{VI_b}$.

The morphology of the synthesized polyketone $\mathbf{VI_e}$ in Fig. 5(a,b,c) was examined by SEM (Jeol-SM-5400 LV instrument). The SEM sample was

Table 4. Solubility Characteristics of Polyketones $\mathbf{VI_{a-h}}$

Polymer Code	DMSO	DMF	NMP	Chloroform Acetone	THF	Methylene Chloride	H_2SO_4
$\mathbf{VI_a}$	±	±	−	±	−	±	+
$\mathbf{VI_b}$	±	±	±	±	±	±	+
$\mathbf{VI_c}$	±	±	−	−	±	−	+
$\mathbf{VI_d}$	+	+	±	±	±	±	+
$\mathbf{VI_e}$	±	±	±	±	±	±	+
$\mathbf{VI_f}$	±	±	±	±	±	±	+
$\mathbf{VI_g}$	+	±	±	±	±	+	+
$\mathbf{VI_h}$	+	+	+	±	±	+	+

(+) Soluble at room temperature RT.
(±) Partially soluble at RT.
(−) Insoluble.

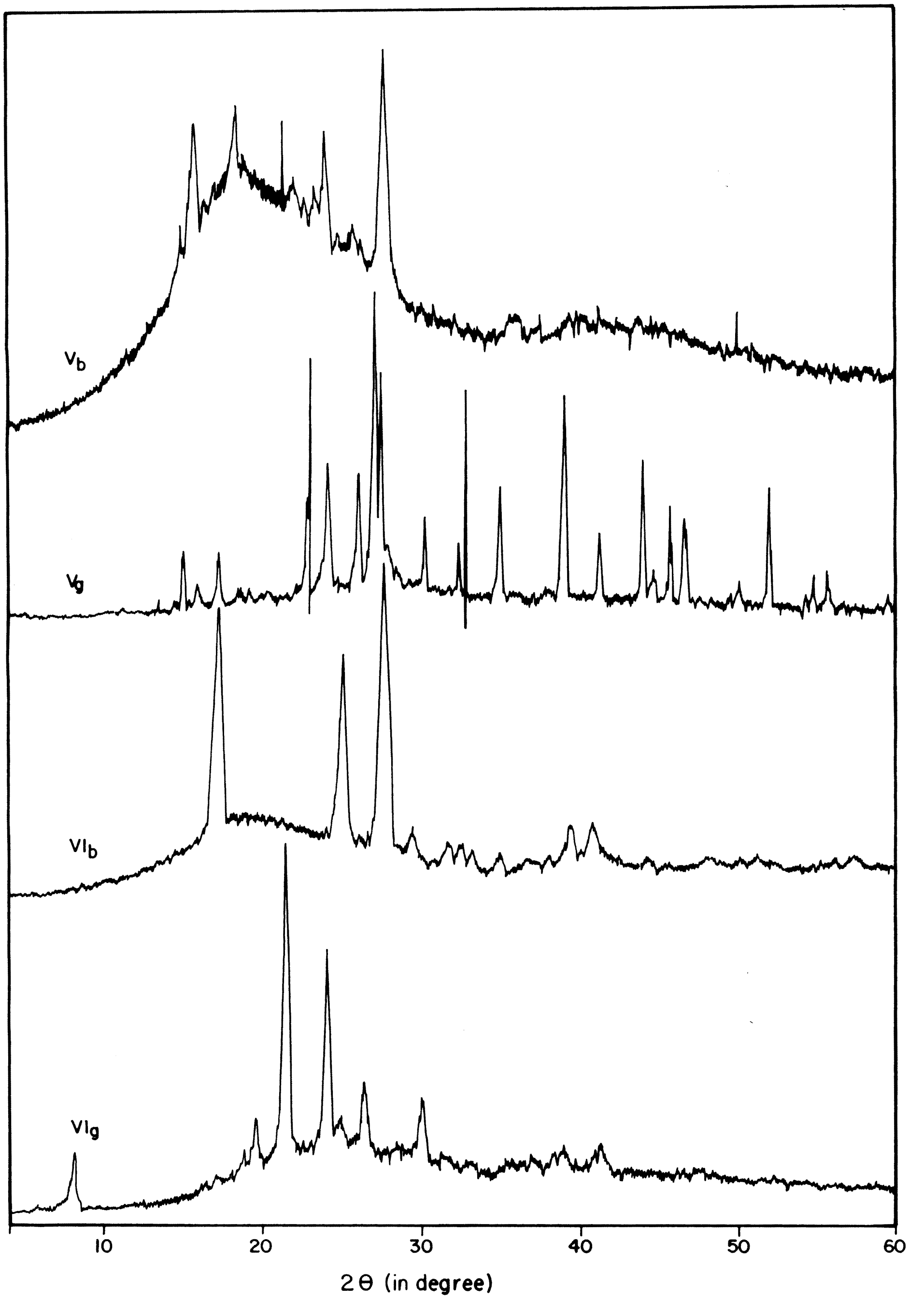
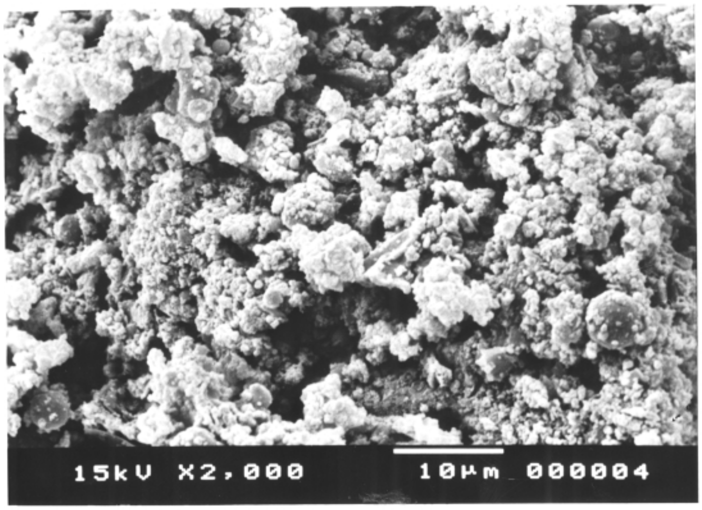
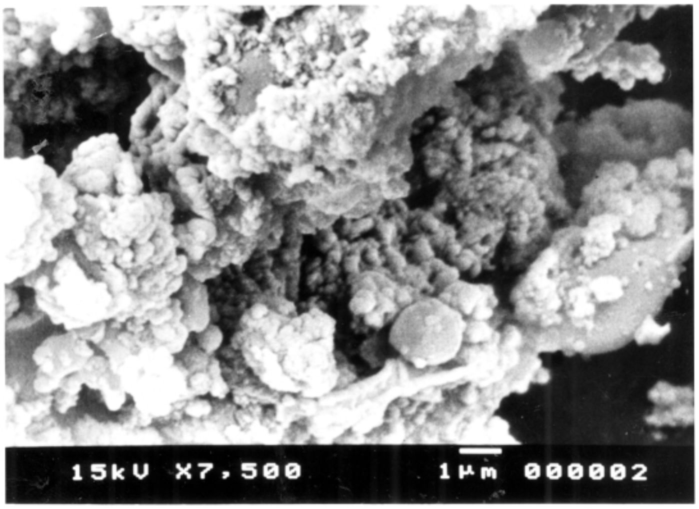


Figure 4. X-ray diffraction patterns of polyketones V_b, V_g, and VI_b, VI_g.



(a)

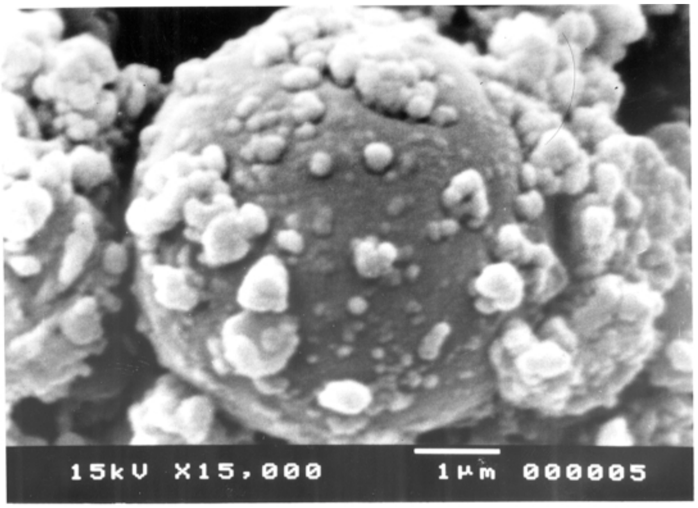


(b)

Figure 5. SEM images of polyketone VI_e.

(continued)

prepared by evaporating a dilute solution of polyketones, a smooth surface of aluminum foil and coating it with gold-palladium alloy. The SEM (Camera) with Ilford film at an accelerating voltage of 15 kv using a low dose technique [22]. The SEM study of polymer $\mathbf{VI_e}$ in Fig. 5(a,b,c) showed that



(c)

Figure 5. Continued.

Table 5. Thermal Properties of Polyketone $\mathbf{V}_{\mathbf{a,e,g}}$ and $\mathbf{VI}_{\mathbf{a,e,g}}$

	Temperature (°C) for Various Decompostion Levels				
Polymer	10%	20%	30%	40%	50%
$\mathbf{V}_{\mathbf{a}}$	300	350	385	445	470
$\mathbf{V}_{\mathbf{e}}$	200	235	260	273	300
$\mathbf{V}_{\mathbf{g}}$	190	235	252	260	270
$\mathbf{VI}_{\mathbf{a}}$	250	360	450	500	540
$\mathbf{VI}_{\mathbf{e}}$	195	225	260	285	305
$\mathbf{VI}_{\mathbf{g}}$	150	200	265	355	440

*Heating rate: 10°C min^{-1}.

the polymer has a polymorph globular and subglobular structures which appeared in a continues chain with some coalescence present.

The thermal behavior of polyketones $\mathbf{V}_{\mathbf{a,e,g}}$ and $\mathbf{VI}_{\mathbf{a,e,g}}$, as selected examples, were evaluated by thermogravimetric analysis (TGA) in air at a heating rate of 10°C min. The thermographs of these polymers are given in Figs. 6 and 7, also Table 5 gives the temperature of various percentages of weight loss. In Fig. 6, TGA curves show a small weight loss in the range 2–4% starting at 160°C until 200°C, which may be attributed to loss of observed moisture and entrapped solvents. The thermographs also indicate, the polymers decompose in two stages. The first stage between 200°C and

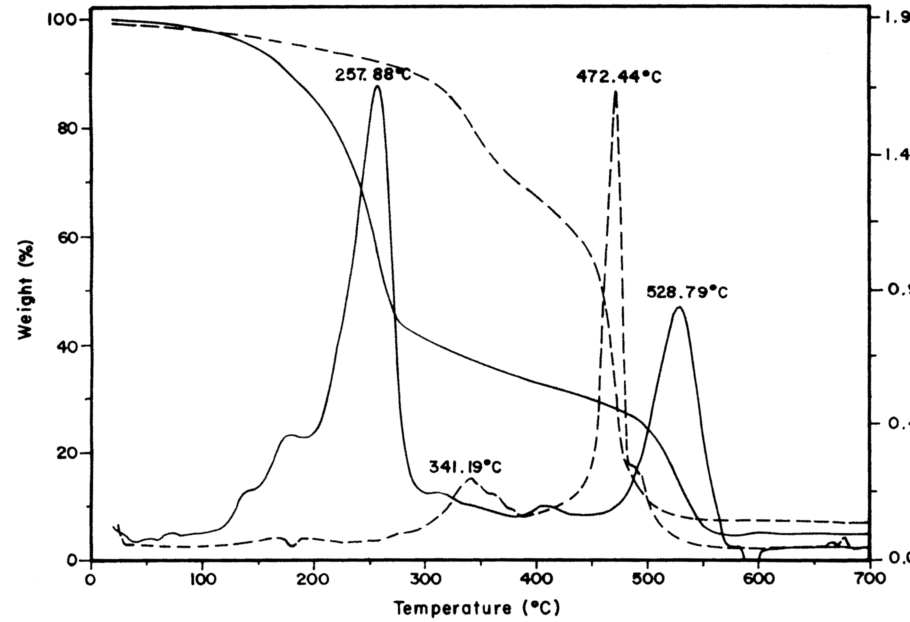


Figure 6. The TGA and DTG traces of polyketones V_a (– – –), V_g (——).

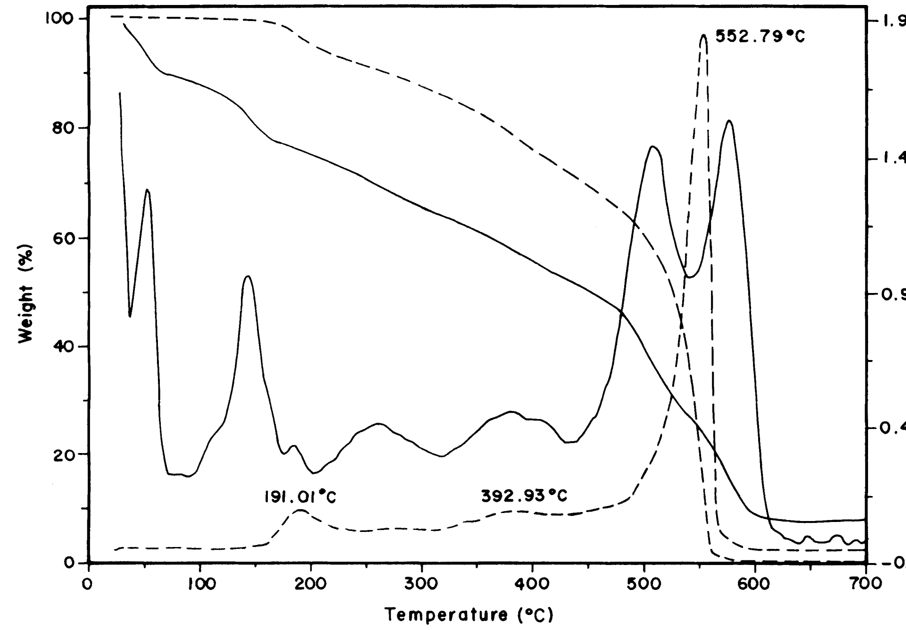


Figure 7. The TGA and DTG traces of polyketones V_a (– – –), V_g (——) in air at heating rate 10°C/min.

300°C depends upon the nature of the polyketones. This result is in good agreement with decomposition of ketone-linkage observed by Swedo and Marvel [23]. The second stage of degradation of polyketones occured between 300°C and 540°C. The rate of degradation in the first stage is somewhat faster than in the second stage. A comparison of the T_{10} values of polymer $\mathbf{V_a}$ containing aromatic moiety showed better thermal stability than others based on aliphatic or azo groups ($\mathbf{V_{e,g}}$). This may be attributed to the molecular architecture that causes rigidity in para-linked aromatic rings (Polymers $\mathbf{V_{b,c,d}}$,$\mathbf{VI_{b,c,d}}$). Meta-Linked aromatics are also thermally stable, but not rigid (Polymers $\mathbf{V_{a,e}}$, $\mathbf{VI_{a,e}}$). More particularly, polyketones $\mathbf{VI_{a,e,g}}$ which are based on the acetone moiety, were slightly more thermally stable than the polyketones $\mathbf{V_{a,e,g}}$ that contain the cycloheptanone ring. This was attributed to the flexibility of the latter moiety [24].

The electronic spectra of selected examples of ($\mathbf{V_{a,e}}$ and $\mathbf{VI_{a,e}}$) were measured in DMSO solution. The UV spectra of polyketones in Fig. 8 showed a broad absorption band at λ_{max} 376 nm for polymer $\mathbf{V_a}$, at λ_{max} 419 nm for polymer $\mathbf{V_e}$, at λ_{max} 387 nm for polymer $\mathbf{VI_a}$, and at λ_{max} 389 nm for polymer $\mathbf{VI_e}$. The appearance of broad absorption band in the UV spectra

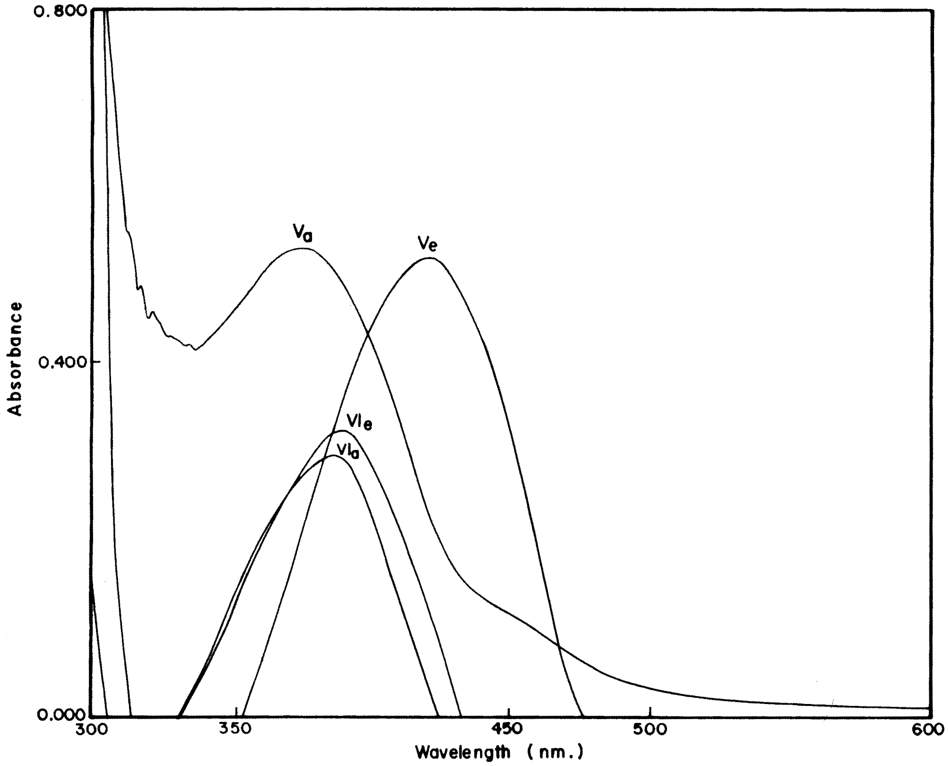


Figure 8. Ultraviolet visible spectra of polymers V_a, V_e and VI_a, VI_e.

is attributed to the excitation of (π-π* and n-π*) for C=C groups, and -N=N- groups (especially in polymer $\mathbf{V_e}$ and $\mathbf{VI_e}$).

CONCLUSION

Two novel series of polyketones based on diarylidenecycloheptanone and diarylideneacetone moieties were synthesized via Friedel-Crafts reaction. All the polyketones were soluble in concentrated H_2SO_4. Thermogravimetric analyses showed that the cycloheptanone based polyketones were somewhat less thermally stable than their acetone counterparts. X-ray diffraction analyses indicated that most of the polymers are semicrystalline. SEM images of polymer $\mathbf{VI_e}$ indicated that their surfaces possess globular and aggregates structure.

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