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The physicochemical characterization of an aminoester-type epoxy-resin hardener prepared from chemical degradation of poly(ethylene terephthalate)

Summary — Chemical degradation of PET scrap was performed by aminoglycolysis with triethanoloamine (TEA) at 180—220°C for 90—360 min and PET/TEA mole ratio 1:2. The reaction products were separated with 2-butanol or isopropanol as solvent into the alcohol-soluble epiphase and the alcohol-insoluble hypophase fractions. Each fraction was evaporated for 6—9 h at 60—80°C *in vacuo* to remove the solvent. Viscosity, hydroxyl number, size exclusion chromatograms, FTIR and ^{1}H and ^{13}C-NMR spectra were measured for the original PET/TEA product and for the fractions. Property differences between the products were explained in terms of side reactions of dehydration of $-N(CH_2CH_2OH)_2$ groups. Rheometric curves were ascertained at room temperature to 200°C to follow the crosslinking processes in the Epidian 6 compositions containing the PET/TEA product as hardener used with the ratios 1:4—1:8 of hardener's tertiary N to resin's epoxy groups and of the Epidian 6 (Bisphenol A) compositions containing one PET/TEA fraction (115S or 115P) or the nonfractionated PET/TEA product as hardener (N/epoxy ratio 1:5). Heat effects accompanying the crosslinking reaction run in the above compositions were followed by DSC. The thermograms disclosed that, in terms of the reaction enthalpy, the product most reactive toward epoxy resin was the alcohol-soluble PET/TEA fraction.

Key words: product of poly(ethylene terephthalate) aminoglycolysis, chemical degradation, fractionation, crosslinking of epoxy resins, rheometric test.

Recently many processes have been developed that are based on new methods of chemical recycling of waste poly(ethylene terephthalate) [1—4]. Most of these processes are intended for low-tonnage production of reactants for plastics and coatings industries [3]. Solvolysis of the ester bond in polyester resins is sometimes realized in the presence of amines, besides the main degrading agent, *e.g.*, a glycol [5]. In such a reaction system, primary and secondary amines can act as catalysts or active aminolytic agents. Mixtures of glycols and aminoalcohols have been used for the chemical degradation of PET [6].

Investigations on the chemical degradation of PET with triethanolamine (TEA) and on various possible applications of the degradation products have been performed in the authors' laboratory [3, 7—11].

The chemical degradation of PET with TEA is considered to be the glycolysis catalyzed by a tertiary amine. Therefore, the term aminoglycolysis can be used to describe this process. The resultant solvolytic activity of *tert*-alkanolamines toward PET ester bonds is a function of both the content of hydroxyl groups and of the amine basicity [9]. The degradation of PET with TEA used in excess, yields aminoester derivatives of terephthalic acid and triethanolamine. The product of aminoglycolysis of PET with TEA (denoted as PET/TEA) carried out at a temperature of about 200°C is primarily characterized by the hydroxyl number and viscosity [7, 9]. The final PET/TEA properties depend on process conditions [12]. The most promising results of PET/TEA application were obtained by using it as epoxy resin hardener. If used to crosslink epoxy resins this product exhibits the following major features:

— beneficial technological properties, *i.e.*, low viscosity in the compositions with liquid epoxy resins within the temperature range from ambient to *ca.* 100°C and a long pot life [9, 11],

— a relatively wide range of weight ratios in the epoxy resin/hardener system [11],

— high values of the mechanical properties such as flexural, tensile and impact strengths of the PET/TEA crosslinked materials [9, 11], which are similar to those prepared with a special plastifying rubber or with thermoplastic modifiers [13, 14],

— it can act as a self-emulsifying crosslinker for liquid epoxy resin/water systems and enables water-thinnable paints to be prepared [10].

A correlation between the PET/TEA characteristics and its effectiveness as the epoxy resin hardener has not yet been investigated. Therefore, physicochemical properties of the PET-derived aminoglycolysis product were evaluated. Additionally, the crosslinking process of the epoxy compositions was followed in low-*M* Bisphenol A resin (Epidian 6)—PET/TEA product systems.

EXPERIMENTAL

Materials

Post-consumer bottle PET (particles less than 6 mm long) was used for the degradation processes.

Merck's reagent-grade 98% pure TEA was used for the chemical degradation of PET.

Epoxy resin: Bisphenol A-based, low-*M* epoxy resin Epidian 6 (epoxy equivalent weight, 186 g; viscosity, 10 700 mPa · s at 25°C) a product of the Organika—Sarzyna Works (Nowa Sarzyna, Poland), was used.

PET chemical degradation products, *viz.*, the products of aminoglycolysis of PET with TEA, were investigated and used as epoxy resin hardeners. The PET/TEA fractions isolated from isopropanol or 2-butanol were also used.

Reagent-grade 99.7% isopropanol (Piekary Śląskie, Poland) and Merck's 99.5% 2-butanol, were used as solvents for fractionations of PET/TEA. Methanol was applied as a size exclusion chromatography eluent.

Methods

Chemical degradation of PET was carried out at a temperature of 180—220°C for 1.5—6 h in a glass reactor. The mole ratio of the PET recurrent unit to TEA was kept constant at 1:2. Some volatile by-products containing ethylene glycol, water and other volatiles were distilled off.

PET/TEA fractionation

The PET/TEA product from the 205°C/2 h process was mixed with a solvent (isopropanol or 2-butanol) at a weight ratio of 1:2 or 1:5 in a glass separatory funnel and set aside until the soluble epiphase and the nonsoluble hypophase separated. After the separation, each fraction was evaporated at 60—80°C for 6—9 h under reduced pressure to get solvent-free products.

^{1}H and ^{13}C-NMR spectra were recorded on a Bruker DPX 400 spectrometer equipped with 5 mm ^{1}H/BB-inverse probehead, operated at 400.13 and 100.62 MHz with of 0.12 and 0.97 Hz per point for ^{1}H and ^{13}C, respectively. For the $CDCl_3$ and CD_3OD solutions and for the D_2O solution, tetramethylsilane and sodium 3-(trimethylsilyl)tetradeuteriopropionate were used as the respective internal standards.

FTIR measurements for the fractions in 2-butanol were performed in $CHCl_3$ on a FTIR—430 "JASCO" instrument.

Size exclusion chromatography (SEC) was applied for comparative analysis of PET/TEA as well as for the fractions isolated from 2-butanol. The SEC system involving a Hewlett-Packard PL Gel 5 μm (average pore size diameter 50 nm) column operating with methanol, flow rate 0.5 cm^3/min, a DRI-Shodex RI-71 and a UV Hitachi LaChrom L-7400 detectors and a Rheodyne 7725i injection value (20 μl), was used.

Hydroxyl number determination

The PET/TEA products and their fractions were characterized in terms of hydroxyl number (HN). The HN values were determined by a modified method of the Polish Standard PN-93/C-89052/03 for samples dissolved in dimethylsulfoxide.

Rheological properties

The PET/TEA and the fractions as well as Epidian 6/hardener systems were controlled by using an ARES (Avanced Rheometric Expansion System) rheometer (Rheometric Scientific), temperature gradient 5°C/min, cone—plate, ϕ = 50 mm, spacing 1 mm.

Differential scanning calorimetry

A DSC-7 Perkin-Elmer (heating rate, 10°C/min) was used to measure the heat effects associated with the crosslinking reactions between Epidian 6 and PET/TEA or fractions.

RESULTS AND DISCUSSION

Aminoglycolysis of poly(ethylene terephthalate) with triethanolamine and characterization of products

Aminoester derivatives of terephthalic acid and TEA are formed during the aminoglycolysis of PET with the alkanolamine used in excess. The final products of the PET/TEA aminoglycolysis were found to differ in the amount of volatile substances distilled off, HN value and viscosity in relation to operating parameters (temperature and time).

Table 1 lists HN data and volumes of the volatile by-products collected in PET degradations run at 180, T a b l e 1. **Volume of volatile by-products and hydroxyl number values and viscosities of the final PET/TEA products obtained during aminoglycolysis at various experimental conditions**

No.	Reaction temperature °C	Reaction time min	Distillate*) volume cm³	Hydroxyl number, mg KOH/g	Viscosity at temperature (Pa · s)		
					ambient	35°C	80°C
1	180	90	0.8	329	49	22.0	0.52
		180	2.8	238	129	30.5	0.48
2	200	90	2.2	280	115	42.0	0.70
		180	3.0	207	132	32.5	0.52
		270	3.4	198	—	—	—
3	220	90	4.5	264	425	125.0	1.10
		180	5.0	76	180	50.7	0.67
		360	5.2	nm	—	—	—

*) 0.1 mol of PET and 0.2 mol of TEA were used, nm — nonmeasurable.

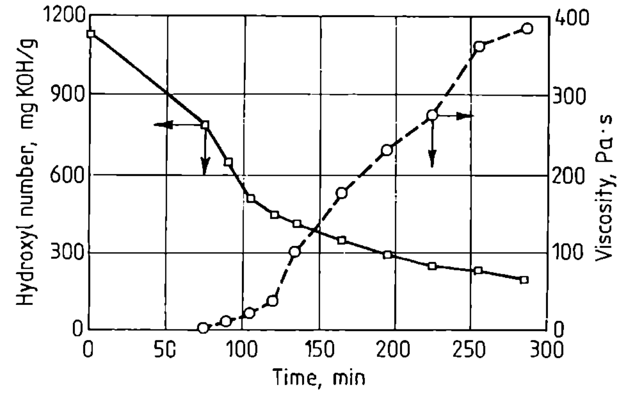

Fig. 1. Hydroxyl number and viscosity of the reactor content as a function of time during aminoglycolysis of PET with triethanolamine at temperatures of 200—210°C

200 and 220°C for 90—360 min. The HN values and the viscosities of the samples collected from the reaction vessel during the process run at 205—210°C are presented graphically in Fig. 1. Table 1 shows the amount of the volatile products distilled off at normal pressure (mainly ethylene glycol and water) and the viscosity of the aminoglycolyzate to increase and the HN values to decrease as the process temperature and time are increased. Under the extremum reaction conditions (220°C/6 h) a highly condensed product was obtained. Its HN value was inaccessible under the test conditions because of precipitation of the excess of phthalic anhydride during the titration. Figure 1 shows a continuous decrease of the hydroxyl number of the reaction mixture starting with value of 1123 mg KOH/g for TEA (theoretical value) down to 199 in 285 min of the process. The highest gradient of HN decrease is seen to occur during the first 100 min. On the other hand, the viscosity of the reaction mixture increases progressively from *ca.* 1.35 Pa · s in 75 min up to 384.5 Pa · s in 285 min of the process. The viscosity gradients are higher at reaction times exceeding 120 min. These results (Table 1, Fig. 1) show that, in addition to the aminoglycolytic solvolysis of polymer ester bonds, some other side reactions are likely to occur under the process conditions. They result in decreased hydroxyl numbers and increased molecular weights of the products. The reasons for such a behavior of the reaction system at temperatures above 200°C are probably intra and intermolecular etherification reactions of the OH-carrying primary PET/TEA degradation products, *i.e.*, morpholine ring formation [15] and condensation of OH groups from various molecules [16]. Hydrogenated pyrrol derivatives can also be formed [17] from the -N(CH_2CH_2OH)$_2$ groups. The deep brown color of the PET/TEA product can be explained by the presence of some oxidation products of pyrrol derivatives.

A scheme of the aminoglycolysis of PET with TEA and the types of possible compounds in the degradation products are presented in Fig. 2.

The PET/TEA products were fractionated with 2-butanol or isopropanol. In each case two fractions could be

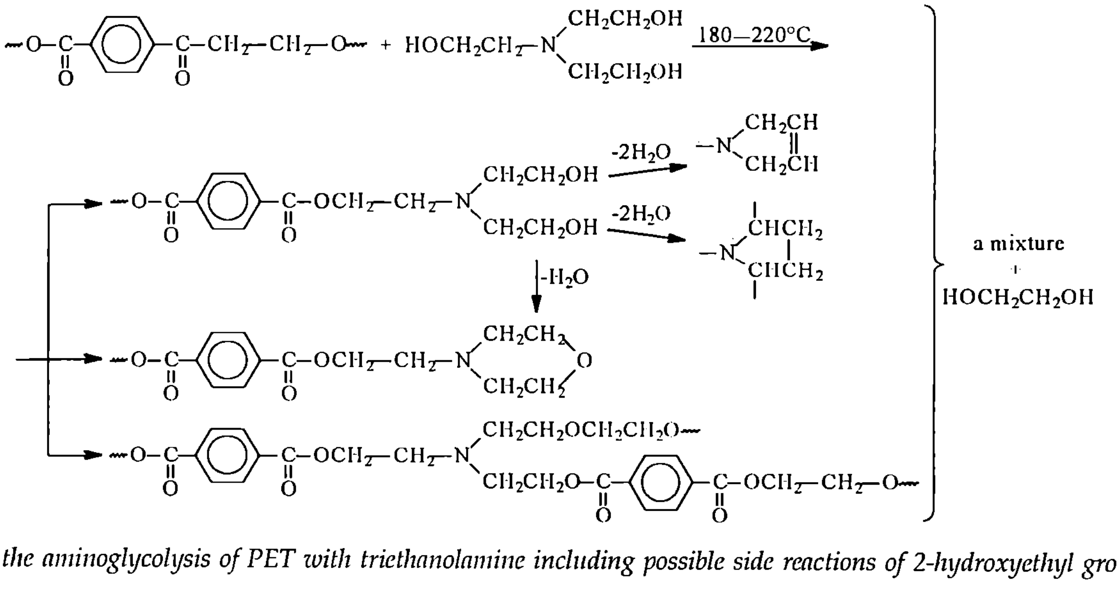

Fig. 2. Scheme of the aminoglycolysis of PET with triethanolamine including possible side reactions of 2-hydroxyethyl groups

isolated: one soluble in the alcohol and another an insoluble resinous precipitate. After the alcohol had been evaporated the fractions were examined.

The values of HN, viscosities (measured at 40°C), and weight fractions of the respective samples are compiled in Table 2. For purposes of comparison, data are also given for the reaction product taken for fractionation [PET/TEA(n)] and for the same product devolatilized [PET/TEA(d)]. The following conclusions can be drawn from Table 2: (*i*) the final PET/TEA degradation product continues to contain some volatiles; devolatilization reduced its real hydroxyl number quite significantly (by *ca.* 27%); (*ii*) the fractions soluble in 2-butanol contain more OH groups than do the insoluble fractions (the respective HN values differ by a factor of 2.4—2.6); (*iii*) viscosities of the insoluble residues measured at 40°C are considerably higher than those for the soluble fractions; (*iv*) at higher weight fractions (55—65 wt. %), a resinous residue is present.

Table 2. PET/TEA*) characteristics compared with data for fractions isolated from 2-butanol

Sample description	Sample/2-butanol weight ratio	Hydroxyl number mg KOH/g	Viscosity at 40°C, Pa · s	Weight fraction, %
PET/TEA(n)**)	1:0	273	37.0	100
PET/TEA(d)***)	1:0	198	34.5	~98
PET/TEA/112S	1:2	337	3.6	~40
PET/TEA/112P	1:2	142	81.7	~60
PET/TEA/115S	1:5	337	3.6	~45
PET/TEA/115P	1:5	126	94.5	~55

*) PET/TEA product from the process performed at 205°C for 2 h.
**) Without devolatilization.
***) After devolatilization at 80°C under lower pressure.

A similar fractionation performed for the 210—220°C/1.5 h PET/TEA degradation product with isopropanol (1:4 weight ratio) allowed to resolve it into two fractions, one with HN = 213 mg KOH/g (soluble part) and another with HN = 138 mg KOH/g (insoluble residue). These fractions differed also in nitrogen content (5.47 and 5.10 wt. % N, resp.).

The above characteristics allow to believe that the alcohol-soluble fractions contain mainly lower-*M* species with a higher number of OH groups, whereas the insoluble resinous fractions of the PET/TEA aminoglycolyzate contain higher-M (oligomeric) substances with lower amounts of OH groups.

Comparison of the SEC elution curves recorded for fractions 115S and 115P (Fig. 3) shows clearly that they are multicomponent mixtures exhibiting some differences. The first is in retention region (R_T) of 20—24 min, visible in the DRI trace (Fig. 3A) which shows a significantly higher concentration of the species eluted in 22.2 and 23.6 min present in the alcohol-soluble fraction

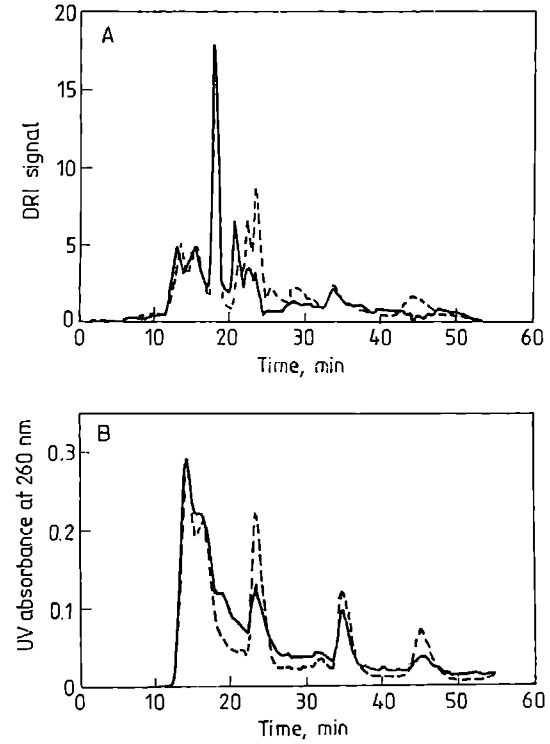

Fig. 3. Size exclusion chromatograms of the PET/TEA fractions 115S (dashed line) and 115P (continuous line) registered by differential refractometer (A) and UV detector (B)

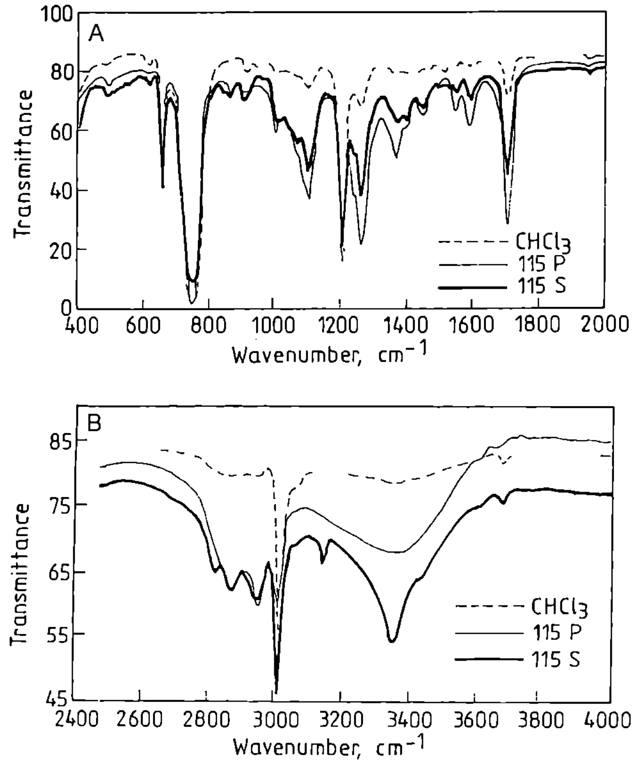

Fig. 4. FTIR spectra of the PET/TEA fractions 115S and 115P

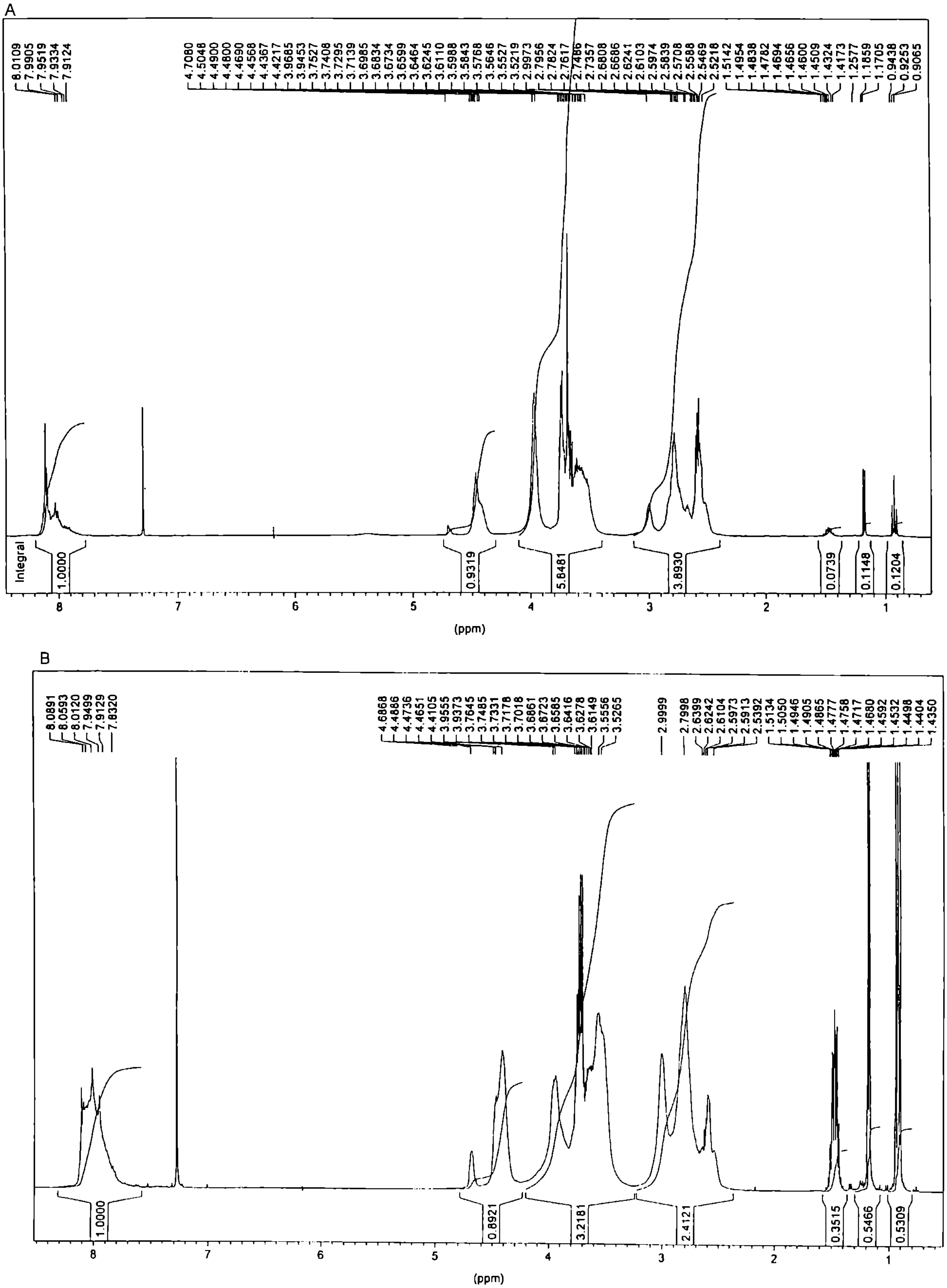

Fig. 5. Proton NMR spectra of the PET/TEA fractions 115S (A) and 115P (B)

(115S) and a less intensive peak of R_T = 20.5 min than that for the PET/TEA residue (115P). The peaks recorded by the UV detector, were different, too; the absorbances of the peaks related to sample 115S and located at 23.5, 34.9 and 45.1 min were higher, whereas the absorbances at R_T < 22 min were lower than those for the respective peaks of the fraction 115P (Fig. 3B).

The peaks with R_T higher than *ca.* 22 min are located above the region of separation based mainly on the size exclusion mechanism [18]. Above that R_T value, separation is affected by the size exclusion as well as by the adsorption of solutes in the methanol mobile phase on the styrene-divinylbenzene copolymer gel. These preliminary SEC results confirm earlier conclusions based on viscosity differences. The higher-M species were accumulated in the PET/TEA residue fraction (R_T < 22 min) as also revealed by viscosity measurements. On the other hand, the differences in the elution curves at R_T = 20—24 min may be associated, *e.g.*, with a higher content of OH groups (*i.e.*, a lower level of dehydrated by-product species, as evident from the HN values determined in the PET/TEA 115S fraction.

Taking into account the results of viscosity measurements (Fig. 1) and SEC separation (Fig. 3) it is obvious that PET/TEA product as well as its fractions are multicomponent mixtures. These findings were confirmed by FTIR and NMR spectra (Figs. 4 and 5).

The FTIR spectra (Fig. 4) show a similarity of the fraction 115S to the 115P. Stretching vibrations C-O-C at 1264 cm^{-1} and C=O at 1708 cm^{-1} are characteristic for esters of carboxylic acids. Three bands in the region of 1000—1150 cm^{-1} are responsible for the OH groups of primary and secondary alcohols, whereas 1370 cm^{-1} is due to C-O stretching vibrations in primary alcohols. Bands at 1447 cm^{-1} and 1592 cm^{-1} are due to CH_2-CH_2 and C=C stretchings in the aromatic ring, respectively. Two bands differentiate these spectra, *viz.*, the 3141 cm^{-1} that occurs only in the fraction soluble in 2-butanol and the 3351 cm^{-1} which is due to stretching vibrations of OH groups in hydrogen-bonded alcohols (significantly more intensive in fraction 115S). The first band is unknown, whereas the second may well be caused by a higher amount of the hydroxyl groups and/or residual 2-butanol in the fraction 115S. Inspection of the ^{13}C-NMR and ^{13}C (135-DEPT) NMR spectra of fractions 115S and 115P exhibited the existence of mixtures of compounds in each fraction. Chemical shifts and the multiplicity of signals, recognized by the DEPT spectra, confirmed the presence of ester carbonyl groups, substituted benzene rings and numerous CH_2N and/or CH_2O groups. To estimate the predominance of OH groups in fractions 115S and 115P, ^{1}H-NMR spectra in $CDCl_3$ solutions (Fig. 5) and, for a comparison, studies were performed in a deuterated solvent. The best solvent for this purpose was CD_3OD, which in contrast to D_2O, gave transparent solutions. Moreover, the remainder water peak much more disturbs the samples than do the remainder peaks of methanol.

In general, the ^{1}H-NMR spectra of each fraction recorded in $CDCl_3$ as well as in CD_3OD as solvent could be divided into four regions. One region is 8.2—7.6 ppm, which undoubtedly consists of unexchangeable aromatic protons of CH_2 groups attached to heteroatoms (O and/or N) and of exchangeable OH protons. The more distinct decrease of an integration area in the spectra recorded in CD_3OD in relation to the spectra measured in $CDCl_3$ as solvent was observed for the 115S (minus 21.0%) as compared with than for the 115P (minus 11.4%), indicative of the larger number of exchangeable OH groups in the former. Using the integrations area of unexchangeable aromatic protons (8.2—7.6 ppm) as the reference we concluded that the contents of OH protons in the regions 4.8—4.2 ppm, 4.2—3.3 ppm, and 3.3—2.4 ppm were 1.1%, 15.2%, 4.7% and 2.5%, 5.6%, 3.3% for the 115S and the 115P, respectively. These findings proved the contents of OH groups to be higher in the 115S fraction in keeping with the FTIR and hydroxyl number data.

Poly(ethylene terephthalate)/triethanolamine aminoglycolysis product as epoxy resin hardener

Tertiary amines can be applied as independent epoxy resin crosslinking agents, but more often they serve as accelerators when carboxylic acid anhydrides are used as hardeners. With a *tert*-amine as crosslinker, the process is based on anionic polymerization additionally accelerated by the hydroxyl groups present in the crosslinking agent molecules and/or in epoxy oligomers [19].

Non-stoichiometric amounts of *tert*-amines are used for crosslinking of epoxides; however, elevated temperatures are required to obtain materials with desirable properties. According to our experience, the recommended ratios of the tertiary nitrogen atom of the PET/TEA hardener to epoxy groups of the epoxy resin should be kept within the range 1:4 to 1:8 (Table 3) [11]. As a first approximation, the PET/TEA hardener was assumed to make epoxy resin crosslink by the anionic mechanism, similarly as tertiary amines do. However, it should be emphasized that the crosslinking mechanism in the PET/TEA/epoxy resin system has not yet been investigated.

Table 3. PET/TEA hardener/epoxy resin ratios used for the preparation of compositions

No.	Epoxy composition description	Short symbol*)	PET/TEA, phr
1	Epidian 6/PET/TEA (4)	E6/1:4	28.8
2	Epidian 6/PET/TEA (5)	E6/1:5	23.0
3	Epidian 6/PET/TEA (6)	E6/1:6	19.2
4	Epidian 6/PET/TEA (7)	E6/1:7	16.5
5	Epidian 6/PET/TEA (8)	E6/1:8	14.4

*) Numbers denote mole ratios of tertiary N atom per number of epoxy groups.

The PET/TEA aminoglycolyzate (205°C/2 h) or its fractions were used to crosslink Epidian 6. Viscosity was monitored in the Epidian 6/hardener system as the temperature was raised at 5°C/min (Fig. 6). Two diagrams (Fig. 6) present the rheometric curves [$\eta = f(T)$] for the epoxy compositions crosslinked with various

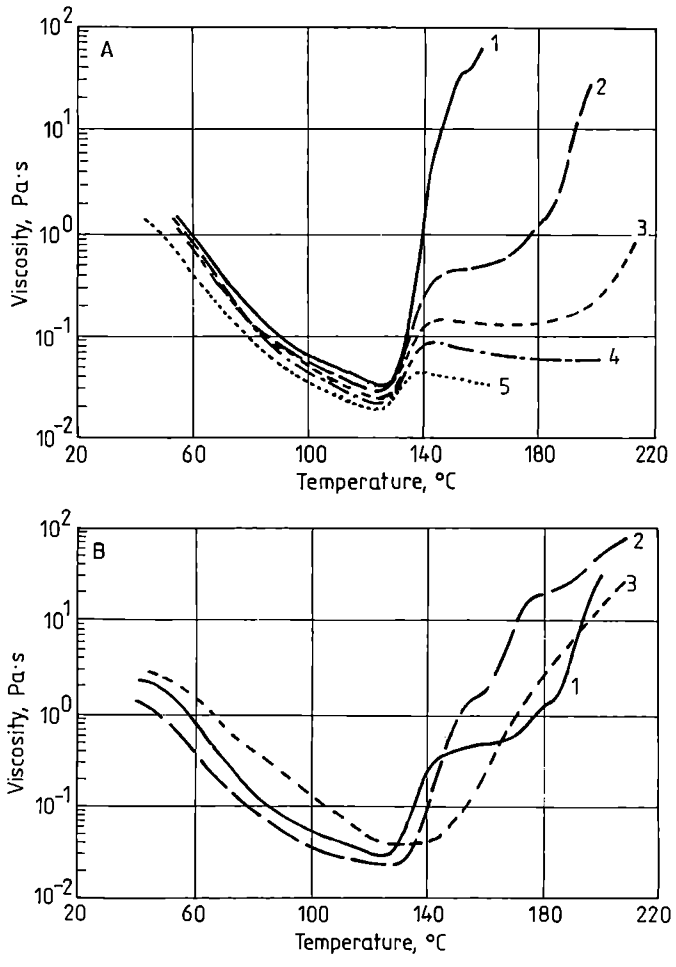

Fig. 6. Rheometric curves for the epoxy compositions crosslinked with PET/TEA hardeners: (A) Epidian 6/PET/TEA with various ratios of the tertiary nitrogen atom of the hardener per epoxy groups of the resin from 1:4 to 1:8; (B) Epidian 6/PET/TEA fractions 115S and 115P and PET/TEA before fractionation, respectively (constant N/epoxy group ratio 1:5). Part A curves: 1 — E6/1:4, 2 — E6/1:5, 3 — E6/1:6, 4 — E6/1:7, 5 — E6/1:8; Part B curves: : 1 — E6/1:5, 2 — E6/1:5 of 115S, 3 — E6/1:5 of 115P

amounts of the PET/TEA product (Fig. 6A) and with the fractions isolated from 2-butanol (Fig. 6B). The following conclusions may be drawn from Fig. 6: (*i*) within the temperature range 80°C to ~130°C (gelation point) the epoxy compositions exhibit low viscosities with a minimum not exceeding *ca.* 30 mPa · s at 120—125°C, and rather long life times at elevated temperatures; (*ii*) the curves are slightly shifted toward higher values of η as the hardener content in the composition is raised; (*iii*) a sharp and continuous increase in viscosity is observed for the Epidian 6/PET/TEA composition with the highest hardener content (E6/1:4, Fig. 6A). Other rheometric curves of the compositions with lower PET/TEA contents (the N/epoxy molar ratios 1:5 to 1:8) exhibit shoulders above 140°C. The corresponding maximum η-values (under the experimental conditions) decrease as the hardener content in the composition is diminished; (*iv*) the rheometric curves of the epoxy compositions containing PET/TEA fractions also show shifting. At temperatures below the gelation point (<130°C), the composition of Epidian 6 with the fraction 115S (soluble in 2-butanol) shows a lower viscosity. At higher temperatures (>130°C), the viscosity of the composition exceeds that for the Epidian 6/PET/TEA 115P system (Fig. 6B). These observations indicate that the former fraction is more active in the epoxy resin crosslinking reaction.

The heat of the crosslinking reaction was followed by DSC for the epoxy compositions with the PET/TEA hardener and with its fractions (Fig. 7). The thermogram recorded for the Epidian 6/PET/TEA system (mole ratio of epoxy groups to N is 4:1) is presented in Fig. 7A, whereas three heat flow curves for the compositions crosslinked with PET/TEA and with the fraction 115S and 115P are shown in Fig. 7B. The crosslinking reaction enthalpies and the temperatures at the corresponding negative peaks of the reaction exotherms are collected in Table 4. The reaction enthalpies decrease in the following order: E6/PET/TEA 115S (or 112S) > E6/PET/TEA > E6/PET/TEA 115P (or 112P) as could be expected on the basis of previously described characteristics of the hardeners. The reactivities of the PET/TEA fractions used for crosslinking of epoxy resins, evaluated from their enthalpy values, differ by a factor of 1.6—2.1 (Table 4). The temperatures at the heat flow curves peaks are 151—158°C.

Table 4. Enthalpies and temperatures of maximum heat flow for epoxy resin /PET/TEA hardener crosslinking reactions

Sample description	Short symbol*)	Enthalpy, J/g	Temperature of maximum heat flow, °C
Epidian 6/PET/TEA(n)*)	E6/1:4	-77.4	150.9
Epidian 6/PET/TEA 115S	E6/115S:4	-103.9	156.9
Epidian 6/PET/TEA 115P	E6/115P:4	-66.2	150.5

*) Epidian 6/PET/TEA(n) — Epidian 6 crosslinked with PET/TEA(n) (4 epoxy groups of the resin per tert.-N atom).

CONCLUSION

The physicochemical characterization of the poly(ethylene terephthalate) aminoglycolysis products with triethanolamine obtained within temperature range 180—220°C has shown some side reactions to proceed in addition to ester bonds solvolysis. These side reactions involve primary compounds carrying OH groups. As a consequence, the aminoglycolyzate is a multicomponent mixture of aminoester derivatives of terephthalic acid and triethanolamine as evidenced by FTIR, NMR and SEC. The dehydratation reactions involving

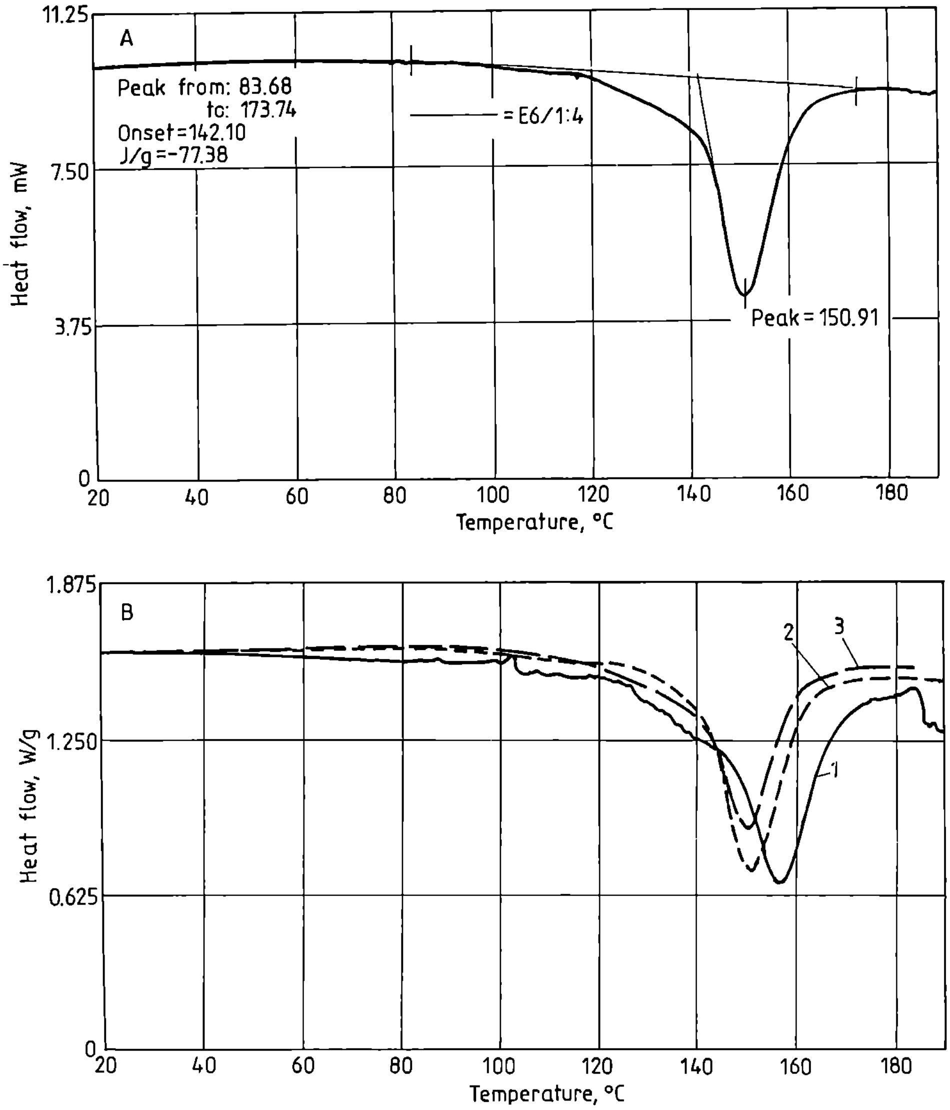

Fig. 7. Exothermic effects during epoxy compositions crosslinking with PET/TEA hardener (A) or with PET/TEA fractions 115S and 115P (B) (constant N/epoxy group ratio 1:4)

2-hydroxyethyl groups can result in the formation of intramolecular cycloaliphatic morpholine rings, hydrogenated pyrrol derivatives, or intermolecular ether bridges. These intermolecular bridges cause an increase in the molecular weight observed as viscosity increase and hydroxyl number fall.

Fractionation allows the PET/TEA aminoglycolysis product to be separated into fractions differing significantly in hydroxyl numbers and viscosities. The fractions contain almost identical components but in different weight ratios as can be seen from size exclusion chromatography patterns. .Therefore, identification of the major components of the fractions is rather difficult. Perhaps, the main component of the PET/TEA product could be identified after the product has been separated by preparative liquid chromatography into narrow fractions.

The PET/TEA fractions soluble in alcohols (2-butanol or isopropanol) proved to be epoxy resin hardeners considerably more effective than those of the residue. The former fractions exhibited higher hydroxyl numbers, higher epoxy resin crosslinking enthalpies, and lower viscosities. Rheometric data show the PET/TEA to be an epoxy resin hardener at elevated crosslinking temperatures with a rather convenient broad weight ratio of the hardener/epoxy resin (14—29 phr). The characteristic features of the epoxy compositions formed by low-M Bisphenol A resin and the PET/TEA hardener include unusually low viscosities at temperatures of 80—120°C and reasonable long life times enabling processing to be run by various techniques. To elucidate the unknown mechanism of the PET/TEA/epoxy resin crosslinking reaction, model epoxy compounds will be studied with the hardener fractions.

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