Whiskers. 3.† Whiskers of Poly(ester-imide)s Derived from 4-Hydroxyphthalic Acid

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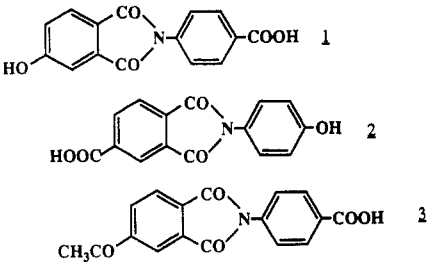
ABSTRACT: *N*-(4-Carboxyphenyl)-4-acetoxyphthalimide was prepared from 4-hydroxyphthalic acid and polycondensed under various reaction conditions. Only a highly purified monomer yields whisker-like crystals. Cocondensations with 4-acetoxybenzoic acid were conducted over a broad range of molar composition (feed ratio), and whiskers were obtained in all cases. These whiskers were characterized by elemental analyses, WAXS patterns, scanning electron micrographs, and DSC measurements. Regardless of composition they possess the same type of orthorhombic crystal lattice and a degree of crystallinity above 85%. All attempts to obtain whiskers from the isomeric monomer, i.e., *N*-(4-acetoxyphenyl)trimellitic acid, failed, even when copolyesters with acetoxybenzoic acid were prepared.

Introduction

Needlelike crystals or whiskers of organic polymers, in particular para-linked aromatic polymers, are of interest for at least two reasons. First, they are normally single crystals and, therefore, permit the characterization of a chemically and physically rather perfect form of the corresponding polymer. Second, organic whiskers may be useful as reinforcing fibers for composites. In this connection whiskers of organic polyesters possess several advantages such as lower specific gravity and easy waste treatment by complete combustion, pyrolysis, and even hydrolysis.

Poly(4-oxybenzoate) (poly(4-HBA) and poly(6-oxy-2-naphthoate) (poly(6,2-HNA)) have been found to form whiskers of variable aspect ratio when prepared under suitable reaction conditions.[1-7] To improve the adhesion of matrix polymers to the polyester whiskers, it is useful to vary the polarity and/or the functional groups on their surface. One approach to reach this goal is the synthesis of whiskers from various different monomers. However, all attempts to obtain whiskers from substituted 4-hydroxybenzoic acids such as 3-chloro-, 3-bromo-, and 3-methoxy-4-hydroxybenzoic acid or 3,5-dichloro-, 3,5-dimethyl-, and 3,5-dimethoxybenzoic acid failed. Furthermore, all polycondensations of acetylated hydroquinone, 2,6-dihydroxynaphthalene, 1,5-dihydroxynaphthalene, or 4,4′-dihydroxybiphenyl with terephthalic acid, 2,6-naphthalenedicarboxylic acid, or 4,4′-biphenyldicarboxylic acid, or their bis(trimethylsilyl) esters, failed to yield whiskers. These negative results indicate that the chances of finding polyesters which are capable of forming whiskers is extremely low.

The present work was aimed at studying the crystal growth of homo- and copolyesters derived from the isomeric imide monomers **1** and **2**. These monomers were selected because it has previously been demonstrated[8] that they fit relatively well into one of the four known crystal lattices of poly(4-HBA).



Experimental Section

Materials. Crude 4-hydroxyphthalic acid was a gift of Bayer AG (W-4150 Krefeld-Uerdingen). It was transformed into pure 4-acetoxyphthalic anhydride (mp 88–90 °C)[8] by means of acetic anhydride. Trimellitic anhydride, 4-aminophenol, 4-aminobenzoic acid, and 4-hydroxybenzoic acid (4-HBA) were gifts of Bayer AG and were used without purification. HBA was acetylated with excess acetic anhydride and pyridine in refluxing toluene. The resulting 4-acetoxybenzoic acid had a melting point of 188–190 °C after recrystallization from toluene.

Marlotherm-S, a commercial mixture of isomeric dibenzylbenzenes, was a gift of Hüls AG (W-4370 Marl). It was used without purification, because it was found that distillation in vacuo did not improve the results.

***N*-(4-Carboxyphenyl)-4-acetoxyphthalimide (3).** 4-Acetoxyphthalic anhydride (0.22 mol) and 4-aminobenzoic acid (0.2 mol) were stirred in dry dimethylformamide for 1 h at 120 °C. After addition of acetic anhydride (0.5 mol), stirring at 120 °C was continued for an additional hour. Afterward, the reaction mixture was poured into cold water (<5 °C) and isolated by filtration. It was recrystallized once from 1,4-dioxane/water and once from toluene/acetic anhydride. Yield 73%; ^{1}H-NMR (DMSO) δ 2.33 (s, 3 H), 7.59 (d, 2 H), 7.63 (dd, 1 H), 7.81 (d, 1 H), 8.03 (d, 1 H), 8.08 (d, 2 H), 12.91 (s, 1 H).

Silylated *N*-(4-Acetoxyphenyl)trimellitimide. *N*-(4-Acetoxyphenyl)trimellitimide was prepared from trimellitic anhydride, 4-aminophenol, and acetic anhydride in dry dimethylformamide as described previously.[8,9]

The dry monomer (50 mmol) and hexamethyldisilazane (100 mmol) were refluxed in dry dioxane (150 mL) until the evolution of NH_3 ceased. The reaction mixture was concentrated in vacuo, and the residue was sublimed twice in a vacuum of 10^{-3} bar at a bath temperature of 180 °C. Yield 68%; mp 186–188 °C.

Anal. Calcd for $C_{20}H_{19}NO_6Si$ (397.48): C, 60.44; H, 4.82; N, 3.52. Found: C, 60.42, H, 4.50; N, 3.40. ^{1}H NMR ($CDCl_3$) δ 0.45 (s, 9 H), 2.33 (s, 3 H) 7.25 (d, 2 H), 7.50 (d, 2 H), 8.04 (d, 1 H), 8.43–8.57 (m, 2 H).

Polycondensations. (A) Poly(ester-imide) 4b. *N*-(4-Carboxyphenyl)-4-acetoxyphthalimide (2.5 mmol) and 4-ace-

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Table I. Monomer Concentration and Results of Polycondensation of Monomer 3 and 4-Acetoxybenzoic Acid (4-ABA) in Marlotherm-S (4 h at 400 °C)

polymer (m/n)	molar concn	yield (%)	elem anal.	% C	% H	% N	morphology
4a (10/0)	0.10	81	calcd	67.93	2.66	5.28	lamellar
			found	67.58	2.51	5.19	
4a (10/0)	0.015	40	calcd	67.93	2.66	5.28	whiskers[a]
			found	66.81	2.44	5.18	
4b (5/5)	0.05	72	calcd	68.57	2.88	3.64	whiskers
			found	67.88	2.75	3.69	
4c (4/6)	0.05	72	calcd	68.77	2.94	3.15	whiskers[b]
			found	68.00	2.72	3.23	
4d (3/7)	0.05	74	calcd	68.99	3.02	2.57	whiskers
			found	68.43	2.94	2.69	
4e (2/8)	0.05	75	calcd	69.26	3.11	1.88	whiskers
			found	68.89	3.00	1.95	
4f (1/9)	0.05	78	calcd	69.59	3.22	1.04	whiskers
			found	69.21	3.05	1.10	

[a] See Figure 3A. [b] See Figure 3B.

Table II. Total Monomer Concentration and Results of Polycondensations of Monomer 5a (Method A) or 5b (Method B) and 4-Acetoxybenzoic Acid in Marlotherm-S (4 h at 400 °C)[a]

polymer (m/n)	synth method	molar concn	yield (%)	elem anal.	% C	% H	% N
				calcd	67.93	2.66	5.28
6a (10/0)	A	0.015	45	found	67.56	2.98	5.27
6a' (10/0)	B	0.05	47	found	67.96	3.06	5.27
				calcd	68.99	3.02	2.57
6b (3/7)	A	0.05	43	found	68.96	3.07	2.83
6b' (3/7)	B	0.05	25	found	68.06	3.37	2.91
				calcd	69.26	3.11	1.88
6c (2/8)	A	0.05	39	found	68.88	3.21	2.15
6c' (2/8)	B	0.10	26	found	68.82	3.29	2.44
6d (1/9)	A	0.05		calcd	69.59	3.22	1.04
				found	69.20	3.35	1.27

[a] In all cases aggregates of lamellar crystals were obtained.

toxybenzoic acid (2.5 mmol) were dissolved in warm Marlotherm-S (100 mL). This solution was then placed into a metal bath preheated to 300 °C. The temperature was then raised to 400 °C over a period of approximately 10 min, and this temperature was maintained for 4 h (under an atmosphere of nitrogen). After cooling, the reaction mixture was diluted with acetone and filtered. The isolated poly(ester–imide)s were washed several times with hot acetone and dried at 120 °C in vacuo. All other members of the series **4a–f** were prepared analogously; see Table I.

(B) Poly(ester–imide) 6a' by the "Silyl Method". Silylated *N*-(4-acetoxyphenyl)trimellitimide (1.5 mmol) was dissolved in 100 mL of Marlotherm-S, and this solution was rapidly heated to 400 °C under an atmosphere of nitrogen. After 4 h the reaction mixture was cooled, diluted with acetone, and filtered. The isolated poly(ester–imide) was washed several times with warm acetone and dried in vacuo. The copoly(ester–imide)s **6b'** and **6c'** were prepared analogously with trimethylsilyl 4-acetoxybenzoate as comonomer. The poly(ester–imide)s **6a–c** were prepared according to procedure A. The total monomer/comonomer concentrations are given in Table II.

Preparation of Composites. The whiskers were suspended in a mixture of methanol and 10 vol % of 6 N sodium hydroxide and stirred for 0.5 h. After filtration the whiskers were washed five times with methanol and dried at 65 °C in vacuo.

Composites of nylon-6 were prepared by dissolution of nylon-6 (3.5 g) in a 1:1 mixture (by volume) of formic acid and dichloromethane (50 mL). Then whiskers (1.5 g) were suspended in this solution, which was afterward dried in air and finally at 80 °C in vacuo. Poly(tetramethyl-Bisphenol-A carbonate) (3.5 g) was dissolved in chloroform, the whiskers (1.5 g) were suspended in this solution, and the resulting suspension was precipitated into methanol and finally dried at 80 °C in vacuo. Films with a thickness of 0.4 mm were pressed at 240 °C in the case of nylon-6 and at 260 °C in the case of the polycarbonate.

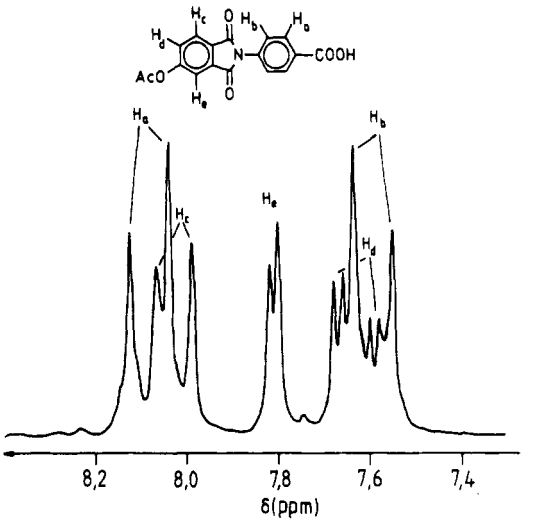


Figure 1. 100-MHz ^{1}H NMR spectrum (aromatic protons) of *N*-(4-carboxyphenyl)-4-acetoxyphthalimide (**3**, grade II) in dimethyl-d_6 sulfoxide.

Measurements. WAXS powder patterns were recorded with a Siemens D-500 diffractometer using Ni-filtered Cu Kα radiation. For this purpose the whiskers were pressed between thin aluminum foils without any pretreatment or preorientation.

DSC measurements were conducted with a Perkin-Elmer DSC-4 in aluminum pans under nitrogen at a heating rate of 20 °C/min.

SEM micrographs were obtained on a Leitz AMR 1600 T instrument with an acceleration potential of 10 kV. All samples were coated with gold on a Balzers SC-040 apparatus.

Dynamic mechanical measurements were performed with a DuPont DMA 983 equipped with a DuPont 990 computer. The resonance mode with an amplitude of 0.3 mm and a heating rate of 2 °C/min was used.

Results and Discussion

Poly(ester–imide)s of 4-Hydroxyphthalimide. For the syntheses of the copoly(ester–imide)s **4a–f** the acetyl derivative (**3**) of the hydroxy acid **1** was used. Its synthesis has previously been reported,[8] but it is described again in the experimental part of this work, because it was found that the purification procedure plays a decisive role in the crystal growth of whiskers. Two grades of purity (denoted I and II) of monomer **3** were prepared and used for polycondensations. Grade I was obtained after recrystallization from dioxane/water in analogy to the previous studies conducted with this monomer.[8] Grade II was obtained from grade I by a posttreatment with acetic anhydride and pyridine in refluxing toluene. It was observed for polycondensation of 4-acetoxybenzoic acid that contamination with 1 mol % of free 4-hydroxybenzoic acid suffices to suppress the crystal growth of whiskers.[5] Possibly the recrystallization of **3** from dioxane/water leads to a slight hydrolysis of the acetate group, albeit not detected by ^{1}H or ^{13}C NMR spectroscopy. Unfortunately, monomer **3** does not show a sharp melting point because of polycondensation at temperatures above 250 °C. For the sake of reliable identification the 100-MHz ^{1}H NMR spectrum of monomer **3** (grade II) is given in Figure 1.

All but one of the polycondensations were conducted in Marlotherm-S at 400 °C for 4 h at a total monomer concentration ≤ 0.05 mol/L. These reaction conditions were found to be optimum for the preparation of whiskers from 4-acetoxybenzoic acid.[5] A first series of homo- and copoly(ester–imide)s with the structure **4a–f** was prepared from grade I monomer **3**, at a total monomer concentration of 0.05 mol/L (not listed in the tables). Neither the

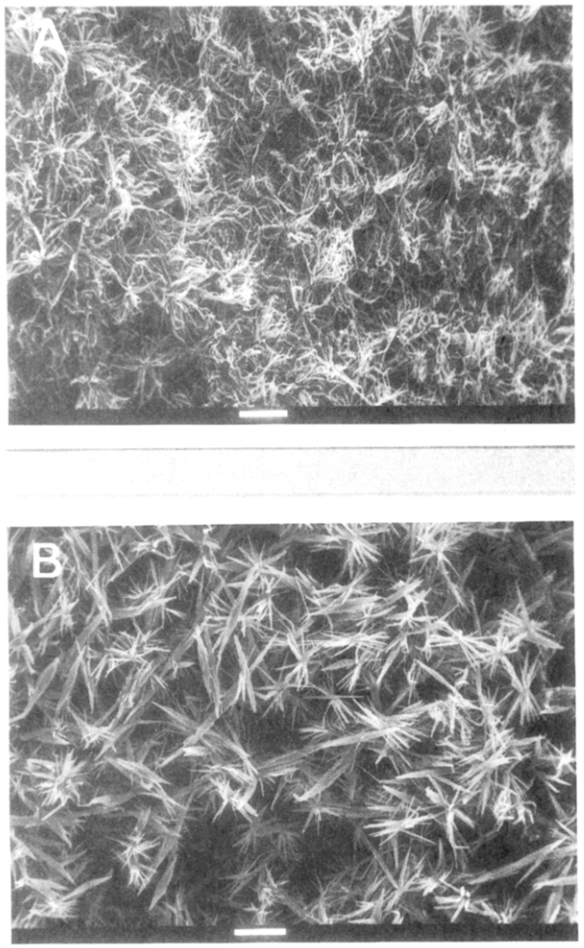


Figure 2. SEM micrographs of poly(ester–imide)s prepared from grade II monomer **3**: (A) homopolymer **4a**; (B) copolymer **4c** (4/6).

homopolyester (**4a**) nor the 5/5 copolyester **4b** yielded whiskers. However, the copolyesters formed crystals of columnar shape, and the aspect ratio increased with increasing molar fraction of 4-HBA units. At a composition of 1/9 (**4f**) whiskers similar to those of pure poly(4-HBA) were isolated (aspect ratios 20–30).

A second series of poly(ester–imide)s **4a–f** was synthesized from grade II monomer **3** (Table I). When the homopolyester was prepared at a relatively high molar concentration, no whiskers were obtained, whereas a second polycondensation of a 0.015 mol/L solution yielded fine whiskers (Figure 2A). Furthermore, copolyesters (4b–**f** formed needlelike whiskers as illustrated by Figure 2B. This result underlines that the posttreatment of monomer **3** with acetic anhydride has a tremendous effect on the crystal growth of homo- and copoly(ester–imide)s. For the sake of a complete characterization, elemental analyses were conducted which confirm that the molar composition of **4b–f** parallels the feed ratio.

To elucidate the influence of the composition of the copoly(ester–imide)s on their crystal lattice, WAXS powder patterns of all samples were recorded. The diffraction maxima in the WAXS powder patterns of samples **4b–f** may be indexed according to an orthorhombic unit cell, corresponding to modification II of pure poly(4-HBA).[10,11] Reflection indices and lattice parameters have been derived from the diffraction patterns in Figure 3A–C and are summarized in Table III. A degree of crystallinity was determined by estimating the amorphous content, subtracting a base line, and measuring the distribution of scattered intensity between the crystalline and amorphous

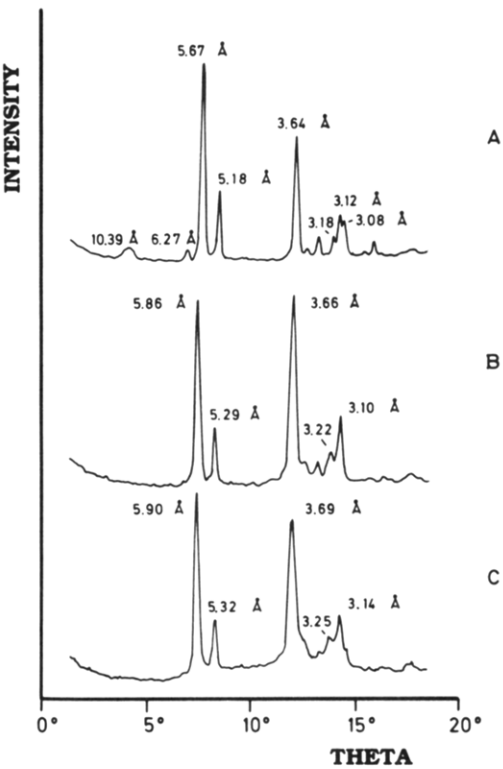


Figure 3. WAXS powder patterns of copoly(ester–imide)s of (A) **4f** (1/9), (B) **4d** (37), and (C) **4b** (5/5).

Table III. Lattice Parameters (Å), Basal Areas (Å²), and Reflection Indices for the Copoly(ester–imide)s of Monomer 3 and 4-Acetoxybenzoic Acid (4-ABA) Based upon Orthorhombic Unit Cells with the Chain Axes Parallel to *c*[a]

	sample **4f**	sample **4d**	sample **4b**
a	3.64	3.66	3.69
b	11.34	11.72	11.80
c	12.54	12.30	12.28
basal area ($a \times b$)	41.3	42.9	43.5
	d-spacings		
reflection index (*hkl*)	sample **4f**	sample **4d**	sample **4b**
010	11.34		
002	6.27		
020	5.67	5.86	5.90
021	5.18	5.29	5.32
100	3.64	3.66	3.69
112/120	3.18/3.12	3.22	3.25
121	3.08	3.10	3.14

[a] The measured *d*-spacings (Å) are taken from the diffraction patters shown in Figure 3.

parts. In all cases, the calculated degree of crystallinity for the copoly(ester–imide)s exceeded 85%.

Patterns A and B of Figure 4 compare the powder diffraction pattern of the previously synthesized homopoly(ester–imide)[8] with that obtained from the whiskers of sample **4a**. For the homopoly(ester–imide), the main reflections may be satisfactorily indexed according to an orthorhombic unit cell corresponding to modification I of pure poly(4-HBA),[10,11] and the results are listed in Table IV. This indexing scheme is based on computer modeling of the chain with a force-field program (Insight II or Biosym), which yields a length of 11.96 Å for the repeating unit. No attempt was made at this stage to index the three additional maxima which are observed in Figure 4B at 8.58, 5.50, and 3.39 Å, although it is thought that they may be attributed to the presence of another modification. It is interesting to note that the 5.98-Å reflection of the whiskers (tentatively indexed as 002) in Figure 4B is

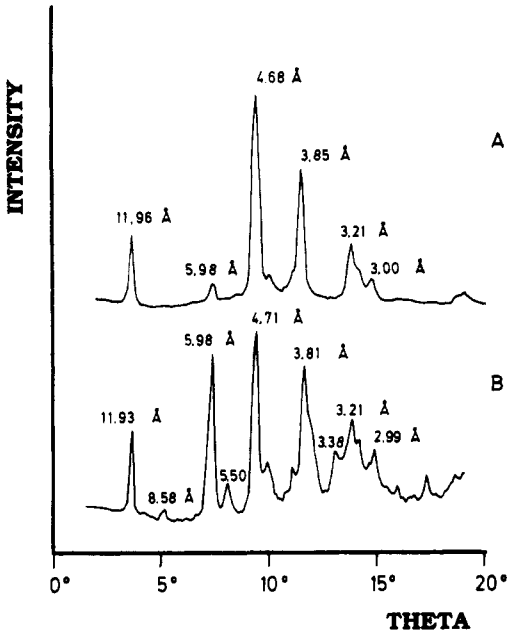


Figure 4. WAXS powder patterns of homopoly(ester–imide) **4a**: (A) prepared from grade I monomer **3** at a concentration of 0.5 mol/L at 350 °C;[8] (B) whiskers prepared from grade II monomer **3** at 400 °C/4 h (concentration = 0.015 mol/L; see Table I).

Table IV. Lattice Parameters (Å), Basal Areas (Å²), and Reflection Indices for the Homopoly(ester–imide)s of Monomer 3 Based upon Orthorhombic Unit Cells with the Chain Axes Parallel to *c*[a]

	homopoly(ester–imide) grade I monomer 3	homopoly(ester–imide) grade II monomer 3
a	7.70	7.62
b	5.89	5.99
c	11.96	11.96
basal area (*a* × *b*)	45.4	45.6

	d-spacings	
reflection index (*hkl*)	homopoly(ester–imide) grade I monomer 3	homopoly(ester–imide) grade II monomer 3
001	11.95	11.93
		8.58
002	5.98	5.98
		5.50
110	4.68	4.71
200	3.85	3.81
		3.38
210/211	3.21	3.21
004	3.00	2.99

[a] The measured *d*-spacings (Å) are taken from the diffraction patterns shown in Figure 4.

significantly stronger than that obtained from the previously synthesized material shown in Figure 4A. This may suggest that the formation of whiskers is in some way related to a higher overall degree of axial chain register.

To summarize these results, the variation of the basal areas of the unit cells and *c* lattice parameters in Tables III and IV as a function of composition has been illustrated in Figure 5. The closed circles represent the basal areas of the unit cells (*a* × *b*), and the open circles correspond to the *c* lattice parameters. The two points on the left-hand axis are for the homopoly(ester–imide), and the two points on the right-hand axis are from the work of Lieser[10] for modification II of poly(4-HBA). The findings indicate that the dimensions of the unit cells correspond to those predicted from the weighted average of the two parent homopolymer structures assuming Vegard's law, further underlining the compatibility of the hydroxy acid **1** with

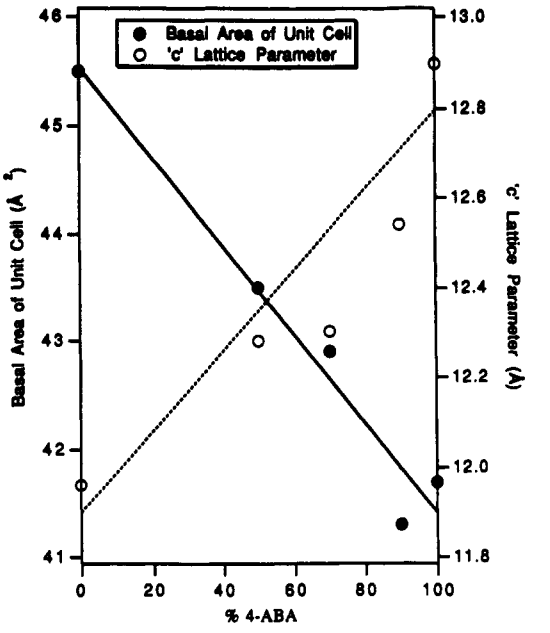
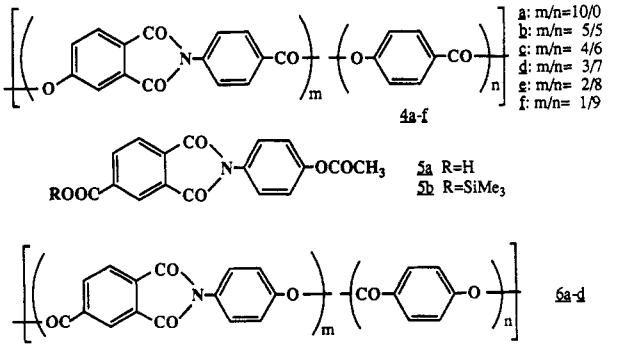


Figure 5. Variation of the basal areas of the unit cells and *c* lattice parameters in Tables III and IV as a function of composition. The closed circles represent the basal areas of the unit cells (*a* × *b*), and the open circles correspond to the *c* lattice parameters. The two points on the left-hand axis are for the homopoly(ester–imide), and the two points on the right-hand axis are from the work of Lieser[10] for modification II of poly(4-HBA). The lines are least squares fits, where the solid line runs through the solid points, and the broken line is drawn through the open circles.

the crystal lattice of poly(4-HBA). This excellent fit may be the basis for the formation of whiskers, and it should be emphasized that the copolyesters **4b–f** represent an extremely rare, if not unique, case in that two monomers with quite different chemical structures form single crystals over such a broad range of composition.

Polycondensations of *N*-(4-Acetoxyphenyl)trimellitimide. Monomer **5a** was prepared as described previously and after recrystallization from dioxane/water subjected to a second acetylation with acetic anhydride/pyridine in refluxing dioxane. Yet despite this posttreatment, the polycondensation in dilute solution at 400 °C (Table II) did not yield whiskers. Furthermore, all cocondensations with 4-acetoxybenzoic acid failed to yield whiskers. Crystals in the form of short columns were obtained at the 1/9 ratio (**6d**).



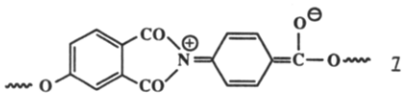
It was demonstrated in a previous paper[6] that silylated 4-acetoxybenzoic acid yields whiskers at much higher monomer concentrations, obviously because the absence of acidic protons reduces side reactions. Furthermore, free 4-HBA which might be present in small quantitites is transformed into the inactive bis(trimethylsilyl) derivative. To find out if impurities and side reactions are responsible for the failure of **5a** to yield whiskers, the

Figure 6. SEM micrograph of poly(ester–imide) **6a** prepared from monomer **4b** (silyl method) at 400 °C/4 h.

trimethylsilyl ester **5b** was prepared. However, even the polycondensation of sublimed **5b** yielded only lamellar crystals (Figure 6) and no whiskers. Lamellae or short columns were also obtained when **5b** was polycondensed with silylated 4-acetoxybenzoic acid (method B in Table II).

These results suggest that the structure of the imide monomer itself and not side reactions is responsible for the failure to yield whiskers. This hypothesis is supported by the following findings. As reported previously,[8] the hydroxy acid **2** is not compatible with one of the orthorhombic modifications of poly(4-HBA) in contrast to the isomer **1**. Incorporation of **2** into poly(4-HBA) favors the formation of a hexagonal chain packing, which in turn seems to be unfavorable for the crystal growth of whiskers. The reason why **2** favors a hexagonal or nearly hexagonal chain packing in contrast to **1** might be the result of a different torsional angle between the phenyl ring and the imide structure. Coplanarity clearly favors an orthorhombic crystal lattice. The delocalization of the nitrogen n-electrons outlined in formula **7** favors the coplanarity, even when it is weak. In the case of imide monomer **2**, such a delocalization cannot occur.

This line of argumentation is supported by the finding that all attempts to prepare whiskers by homo- or cocondensations of acetylated 4′-hydroxybiphenylcarboxylic acid (**8**) failed. WAXS powder patterns demonstrate that both the homopolyester of **8** and the copolyesters containing 4-HBA units prefer a hexagonal chain packing.[12,13] From crystal structure analyses of low molecular weight biphenyl derivatives it is known that the phenyl rings tend to adopt a torsional angle ω around 30 °C. Unfortunately, the torsional angles of the imide monomers **1** and **2** are unknown, and computer modeling with a force-field method (Insight II of Biosym) yielded ambiguous results. Thus, the above-discussed relationship between torsional angle crystal structure and crystal growth of whiskers remains a speculative hypothesis at the current state of research.



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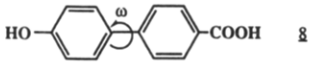

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Composites. To obtain a first impression of how whiskers of polyesters perform in composites, several composites were prepared and examined by dynamic mechanical analysis (DMA). These DMA measurements did not yield accurate absolute values of moduli or strengths, but they are reliable indicators of trends, and only comparisons and trends will be discussed in the present work. More detailed mechanical studies shall be reported in future parts of this series.

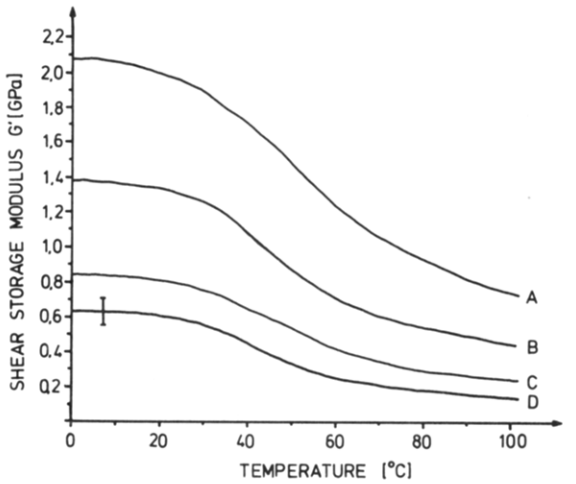

Figure 7. Dynamic mechanical measurements of nylon-6 composites conducted at a heating rate of 2 °C/min: (A) 30 wt % poly(4-HBA) whiskers; (B) 30 wt % whiskers of copoly(ester–imide) **3d**; (C) 30 wt % poly(4-HBA) globular crystals; (D) neat nylon-6.

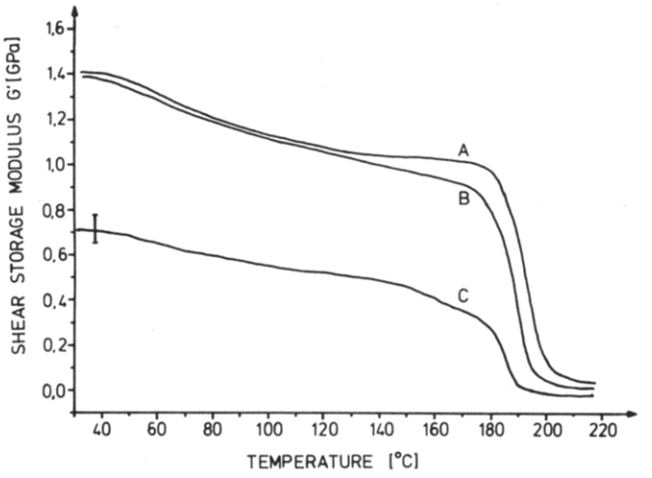

Figure 8. Dynamic mechanical measurements of poly(tetramethyl-Bisphenol-A carbonate) composites conducted at a heating rate of 2 °C/min: (A) wt % poly(4-HBA) whiskers; (B) 30 wt % whiskers of copoly(ester–imide) **3d**; (C) neat polycarbonate.

Two groups of composites were prepared from two different matrix polymers: group I was based on commercial nylon-6 and group II on poly(tetramethyl-Bisphenol-A carbonate) (PTBC). All composites were prepared from a suspension of whiskers in a stirred solution of the matrix polymer, followed by coprecipitation and pressing of the films. It was found that the "as-polymerized" whiskers possessed a feltlike consistency, and an alkaline surface treatment was required to produce a homogeneous suspension of the whiskers in the solutions of the matrix polymers. The alkaline treatment of the whiskers presumably reduces the roughness of their surface by partial hydrolysis. The homogeneity of the composites was examined and confirmed by electron microscopy of fracture surfaces.

Three kinds of nylon-6 composites were measured and compared with neat nylon-6. The results are displayed in Figure 7, where the storage modulus has been plotted

against temperature. Each plot represents the average of five measurements based on five independently prepared films. The margin of error in these five measurements is indicated by the bar perpendicular to curve D. A comparison of neat nylon-6 (curve D) with a composite containing 30 wt % of poly(HBA) whiskers (aspect ratio 20–30) indicates that the whiskers enhance the storage modulus by a factor of 3 (curve A). In contrast, the incorporation of globular crystals of poly(HBA) (similar to those of Figure 6) does not lead to a significant increase in storage modulus (curve C). These results demonstrate that the morphology of the whiskers may have a considerable influence on the mechanical properties of a composite. Curve B illustrates the effect of **7d** whiskers. The poorer performance of the poly(ester–imide) whiskers compared to those of poly(HBA) is difficult to explain without further studies, because two parameters have an influence on the results: (a) the electronic properties of the surface and (b) the roughness of the surface.

Figure 8 demonstrates that a change of the matrix has also a considerable influence on the results. The reinforcing effect of the poly(HBA) whiskers is reduced considerably and is comparable to the result obtained for the **7d** whiskers. Overall this suggests that composites of polyester whiskers are worth studying in more detail, but any further interpretation at this stage would be premature.

References and Notes

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