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Copolyamides of Caprolactam and *m*-Xylylenediammonium Isophthalate

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Synopsis

Caprolactam was copolymerized with *m*-xylylenediammonium isophthalate (MXD-I) to form a series of partially aromatic, thermoplastic copolyamides. The MXD-I mole-% in these systems varied between 91% and 31%. Using differential scanning calorimetry (DSC), the glass transition temperature of these copolyamides was shown to decrease with decrease in their aromatic content. The T_g of a typical copolyamide with an MXD-I mole fraction of 0.34 is about 50°C higher than that of nylon 6. The DSC thermograms of MXD-I homopolymer and all the reported copolymers revealed no distinct melting endotherms or crystallization exotherms, and these polymers were considered to be essentially amorphous. Most of these copolyamides were shown by thermogravimetric analysis to be more thermally stable than nylon 6. Compression-molded films of several copolymers were prepared and were shown to be less hydrophilic than nylon 6, regardless of their suggested amorphous nature. Tensile properties of several copolyamides were measured and were shown to depend on their MXD-I content, degree of polymerization, and chain sequential arrangement. A few of the reported copolyamides exhibited some unusual solution, thermal, and tensile properties. This was ascribed to the presence of block structures and/or branching in these systems.

INTRODUCTION

The use of *m*-xylylenediamine with aliphatic comonomers for synthesis of partially aromatic copolyamides was described by several authors as a means for obtaining polymeric materials having many desirable properties.[1–21] Typical properties of these copolyamides include high glass transition temperatures,[1–4] transparency,[5,6] and light stability.[7] Although many of the reported copolyamides were based on lactams or ω-aminoalkanoic acids and *m*-xylylenediammonium carboxylates,[4,7,10–21] only a few pertained to copolymers of lactams and *m*-xylylenediammonium isophthalate (MXD-I).[14–16] This communication describes the copolymerization of MXD-I with caprolactam and the effect of composition on the thermal characteristics, hydrophilicity, and tensile properties of the resulting copolyamides.

EXPERIMENTAL

Analytical Methods

Reduced viscosities (η_{red}) were obtained for polymer solutions in concentrated sulfuric acid or *m*-cresol (0.5 g/100 ml). Infrared spectra of MXD-I in a KBr

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disk and polymer films (cast from trifluoroethanol) were recorded on a Perkin-Elmer Model 700 spectrophotometer. A Norelco x-ray diffractometer with crystal monochromatized CuK_α radiation was used to examine the morphology of typical copolymers. Differential scanning calorimetry (DSC) data were obtained (in nitrogen) on a du Pont 990-DSC, using a sample of about 10 mg and a heating rate of 10°C/min. In most cases, the polymers were heated from 0°C to 240°C in order to obtain the initial heating data. The samples were then kept at that temperature for 2 min, quenched in liquid nitrogen, and reheated to record the reheating data. Thermal stabilities of polymers (in terms of per cent weight loss in nitrogen) were determined on a Cahn RG TGA balance using a sample of about 5 mg and a 10°C/min heating rate. Selected polymers were compression molded at 250°C (I-D to I-K, II-A, II-B, II-E, and VI) or 265°C (III-A, III-B, IV, V-A, and II-B) to ⅛-in.-thick sheets or 1-mil-thick films. These were used for studying the effect of thermal history during the molding cycle on reduced viscosity, water absorption, and tensile properties. For the measurement of the polymer's tensile properties, microtensile specimens were prepared according to the ASTM specifications and examined on an Instron tensile tester, using a head speed of 0.5 in./min. The microtensile specimens were conditioned at 23°C and 50% R.H. before measuring their per cent ultimate elongation (UE), ultimate tensile strength (UTS), work to break (WB), and 2% secant modulus (SM).

Monomer and Polymer Syntheses

Preparation of m-Xylylenediammonium Isophthalate (MXD-I). Following a procedure similar to those reported earlier,[6,22] MXD-I was prepared by adding isophthalic acid (166 g, 1 mole) in small portions to a hot solution of *m*-xylylenediamine (136 g, 1 mole) in water (340 ml). The clear solution was allowed to cool, and a sufficient amount of 2-propanol was added to effect the crystallization of the salt. The crystalline product was isolated by filtration and dried in vacuo to give an almost quantitative yield of MXD-I. IR (KBr): broad absorption in the 3000 cm^{-1} region, with a distinct peak at 2170 ($-^{+}NH_3$ stretching and overtones), 1605 (N—H asymmetric bending), and 1560 cm^{-1} ($-COO^{-}$ carbonyl stretching).[23]

General Polymerization Procedure. A carefully blended mixture of MXD-I and caprolactam (or MXD-I alone) was heated in a polymerization flask under a nitrogen atmosphere at 180°C for 2 hr. The mixture was then heated to the desired temperatures during one to two hours and kept at those temperatures for the necessary period of time, as shown in Table I. The polymeric product was then cooled, isolated and ground. The polymer was then extracted with methanol in a Soxhlet extractor for 2 days, isolated and dried *in vacuo* at 70°C. The reduced viscosities of the extracted polymers are recorded in Table I.

RESULTS AND DISCUSSION

Effect of Composition and Reaction Conditions on Degree of Polymerization

The polymerization data in Table I indicate that polymerization for 2 hr at 180°C followed by another 2 hr at 245°C results in copolymers (I-A to I-K) having

TABLE I
Polymerization Conditions

Polymer no.	MXD-I,[a] mole-%	Reaction time/temp., hr/°C	η_{red} (H_2SO_4) of product	Polymer no.	MXD-I,[a] mole-%	Reaction time/temp., hr/°C	η_{red} (H_2SO_4) of product
I-A	100	2/180, 2/245	0.85				
I-B	82	2/180, 2/245	0.78	II-C	42	2/180, 5/240	1.14
I-C	76	2/180, 2/245	0.84	II-D	36	2/180, 5/240	1.06
I-D	69	2/180, 2/245	0.71	II-E	35	2/180, 5/240	1.08
I-E	65	2/180, 2/245	0.97	II-F	33	2/180, 5/240	1.17
I-F	61	2/180, 2/245	0.75	II-G	31	2/180, 5/240	0.95
I-G	58	2/180, 2/245	0.85	III-A	88	2/180, 2/240	0.55
I-H	54	2/180, 2/245	0.86	III-B	88	2/180, 3/240	0.72
I-I	44	2/180, 2/245	0.82	IV	78	2/180, 2/220	
I-J	39	2/180, 2/245	0.88			2/240	0.68
I-K	34	2/180, 2/245	0.78	V-A	91	2/180, 4/240	0.87
II-A	50	2/180, 5/240	1.60	V-B	91	2/180, 5/240	1.76
II-B	46	2/180, 5/240	1.42	V-C	91	2/180, 6/240	b

[a] MXD-I = Mole-% of *m*-xylylenediammonium isophthalate in presence or absence of caprolactam.
[b] Partially crosslinked polymer: η_{red} of soluble fraction is 1.26.

TABLE II

Effect of Molding[a] and Type of Solvent on the Reduced Viscosity of Polymers

Polym. no.	MXD-I, mole-%	Before molding in H_2SO_4	After molding in H_2SO_4	Before molding in *m*-cresol	$\frac{\eta_{red}\ (H_2SO_4)}{\eta_{red}\ (m\text{-cresol})}$
I-D	69	0.71	—	0.66	1.08
I-H	54	0.86	0.85	0.82	1.05
I-I	44	0.82	0.81	0.78	1.05
I-J	39	0.88	0.86	0.81	1.08
I-K	34	0.78	0.76	0.79	0.99
II-A	50	1.60	—	0.99	1.62
II-B	46	1.42	—	0.93	1.53
II-E	35	1.08	—	0.93	1.16
VI	0[b]	2.76		2.97	0.93

[a] Samples were molded at 250°C.

[b] Nylon 6.

moderate degrees of polymerization (D.P.), η_{red} = 0.71 to 0.95, regardless of the differences in monomer composition. The high D.P. of copolymers II-A to II-G may be attributed to the relatively long postpolymerization time (5 hr) at 240°C as compared with copolymers I-A to I-K. The decrease in η_{red} with decrease of MXD-I content in II-A to II-G (50 to 31 mole-%) may be associated with optimum melt viscosities of the II-A and II-B reaction mixtures which lead to high-D.P. polymers. On the other hand, the higher chain rigidity of II-A and II-B copolymers as compared with II-C to II-G can be partly responsible for the observed high solution viscosities. The increase of D.P. with increase in the postpolymerization time is illustrated by copolymers III-A and III-B, which have identical composition but different η_{red}. Additional heat treatment prior to the optimum temperature for postpolymerization seems to improve the D.P. of the copolymers as can be realized by comparing III-A with IV. Prolonged postpolymerization at 240°C can lead to a very high D.P. or partially crosslinked copolymers as exemplified by copolymers V-A to V-C.

Additional heat treatment of the polymers during compression molding did not cause any significant changes in their degree of polymerization, as indicated by the reduced viscosity data in Table II.

Effect of Composition and Degree of Polymerization on Polymer Properties

Mole fractions of MXD-I moieties in the initial polymerization mixtures were considered to be practically the same as those of the resulting copolyamides. This is because the copolyamides were isolated in almost quantitative yields and only negligible amounts of methanol-extractables were obtained in a few cases. Estimates of the aromatic content of the copolyamides were obtained by monitoring the relative intensities of the aromatic and aliphatic C—H stretching vibrations at about 3100 and 2950 cm^{-1}, respectively, in the infrared spectra of polymer films prepared under identical conditions.[23] The molecular weights of copolyamides were not measured directly, and their reduced viscosities were used as a measure of the degree of polymerization. The solution viscosities of all polymers were measured in concentrated sulfuric acid. Since the solvent–

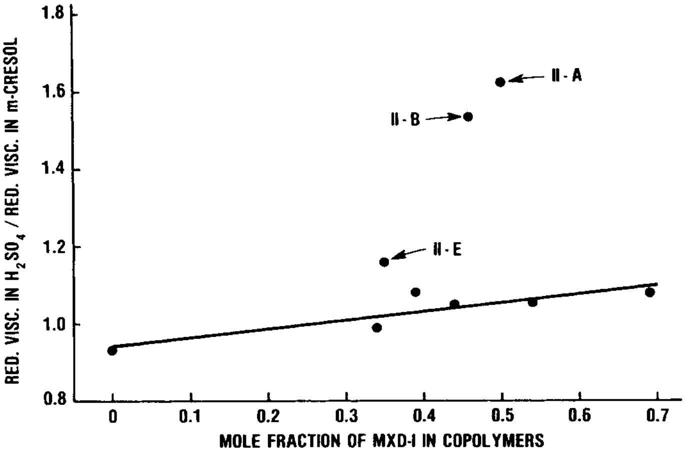

Fig. 1. Effect of copolymer composition on reduced viscosity.

polymer interactions are expected to vary with composition,[24] the reduced viscosity data are used only to compare copolymers which constitute a narrow composition range. For a series of random copolyamides, one may expect that their reduced viscosities will depend not only on composition but also on the type of solvent used.[24-26] In an attempt to ascertain the presence of such dependence in caprolactam-MXD-I copolyamides, the reduced viscosity of selected systems were determined separately in sulfuric acid and *m*-cresol, and the ratios of the two viscosities were correlated with composition. The data in Table II and Figure 1 show a general increase in the viscosity ratio values with an increase in the MXD-I mole fraction of the copolymers. Since viscosity of nylon 6 itself shows an opposite dependence on the type of solvent, the observed effect of solvent on the viscosity of the copolyamides can be ascribed to the interaction between sulfuric acid and the aromatic moieties. As one reexamines the data in Figure 1, it becomes apparent that all but three points indicate an essentially linear relation between the viscosity ratio and the MXD-I mole fraction. The deviation of these three points from the straight-line relation can be attributed to a particular sequential arrangement which is characteristic of the corresponding copolymers (II-A, II-B, and II-E). Considering some of the recent studies on the effect of sequential arrangement on the solution viscosity of copolymers,[24-26] the presence of block structures in copolymers II-A, II-B, and II-E is a strong possibility.

General dependence of the glass transition temperature of the copolyamides on their degree of polymerization and aromatic content can be illustrated using the DSC data in Table III. The T_g data in Table III correspond to T_{g_1} and T_{g_2} which were recorded in the initial heating and reheating DSC thermograms, respectively. As mentioned in the experimental section, at the end of each polymerization, the molten polymer is slow-cooled, ground, extracted, dried at 70°C *in vacuo*, and then left at room temperature for a few days before measuring its T_{g_1}. In order to measure its T_{g_2}, a polymer was heated in the DSC sample holder at 240°C for 2 min, quenched with liquid nitrogen, and then program reheated. Under these conditions, the polymers are expected to have identical thermal histories, and the measured T_g's should correspond to amorphous phases which

TABLE III

Differential Scanning Calorimetry T_g Data

Polym. no.	MXD-I, mole-%	η_{red} (H_2SO_4)	T_{g_1},[a] °C	T_{g_2},[b] °C	T_{g_2}/T_{g_1}	Polym. no.	MXD-I, mole-%	η_{red} (H_2SO_4)	T_{g_1}, °C	T_{g_2}, °C	T_{g_2}/T_{g_1}
I-A	100	0.85	130	172	1.32	I-J	39	0.88	65	98	1.50
I-B	82	0.78	131	164	1.25	I-K	34	0.78	65	90	1.38
I-C	76	0.84	118	155	1.31	II-A	50	1.60	87	118	1.36
I-D	69	0.71	107	150	1.40	II-B	46	1.42	88	120	1.36
				(75)[c]					(166)[c]		
I-E	65	0.97	102	145	1.42	II-E	35	1.08	58	110	1.90
									(160)[c]		
I-F	61	0.75	101	140	1.38						
I-G	58	0.85	95	134	1.42	II-G	31	0.95	65	118	1.82
			(58)[c]								
I-H	54	0.86	87	119	1.37	III-A	88	0.55	92	157	1.71
I-I	44	0.82	70, 74	107	1.48	III-B	88	0.72	113	154	1.36
						IV	78	0.68	115	154	1.34
						V-A	91	0.87	118	161	1.36
						V-B	91	1.76	119	167	1.40
						VI	0[d]	2.76	—	40	—

[a] Initial heating glass transition T_{g_1} (in copolyamides with high aliphatic contents, a few very weak endotherms were also recorded between 100 and 200°C).

[b] Glass transition temperature T_{g_2} of reheated polymers.

[c] Minor T_g.

[d] 0% MXD-I ≡ nylon 6.

TABLE IV
Effect of Composition on Glass Transition Temperature

Polym. no.	η_{red}	MXD-I[a]		Initial heating data[b]			Reheating data[c]		
		F	W	T_{g_1}, °C	$(T_{g_m} - T_{g_1})\left(\frac{F_m}{1 - F_m}\right)$	$(T_{g_m} - T_{g_1})\left(\frac{W_m}{1 - W_m}\right)$	T_{g_2}, °C	$(T_{g_m} - T_{g_2})\left(\frac{F_m}{1 - F_m}\right)$	$(T_{g_m} - T_{g_2})\left(\frac{W_m}{1 - W_m}\right)$
I-A	0.85	1.00	1.0	130[f]	—	—	172	—	—
V-A	0.87	0.91	0.96	118	121	288	161	111	264
I-C	0.84	0.76	0.88	118	40	88	155	56	124
I-D	0.71	0.69	0.84	107	51	121	150	49	116
							(75)[d]	(216)[d]	(510)[d]
I-F	0.75	0.61	0.78	101	46	103	140	50	113
I-G	0.85	0.58	0.76	95	49	110	134	53	120
				(58)[d]	(86)[d]	(227)[d]	—	—	—
I-H	0.86	0.54	0.73	87	50	116	119	62	143
I-I	0.82	0.44	0.65	70, 74	47, 44	112, 104	107	51	121
I-J	0.88	0.39	0.60	65	42	98	98	47	111
I-K	0.78	0.34	0.55	65	34	79	90	42	100
VI[e]	2.76	0	0	—	—	—	40	—	—

[a] F = Mole fraction; W = weight fraction.

[b] Samples were slow cooled from melt, ground, extracted, dried at 70°C *in vacuo*, and stored at room temperature. In addition to recorded T_{g_1}, a number of very weak endotherms were observed between 100 and 200°C. T_{g_m} is the glass transition temperature of MXD-I homopolymer.

[c] Samples were heated to 240°C, kept at that temperature for 2 min, quenched in liquid nitrogen, and then reheated.

[d] Minor T_g.

[e] Nylon 6.

[f] T_{g_m}.

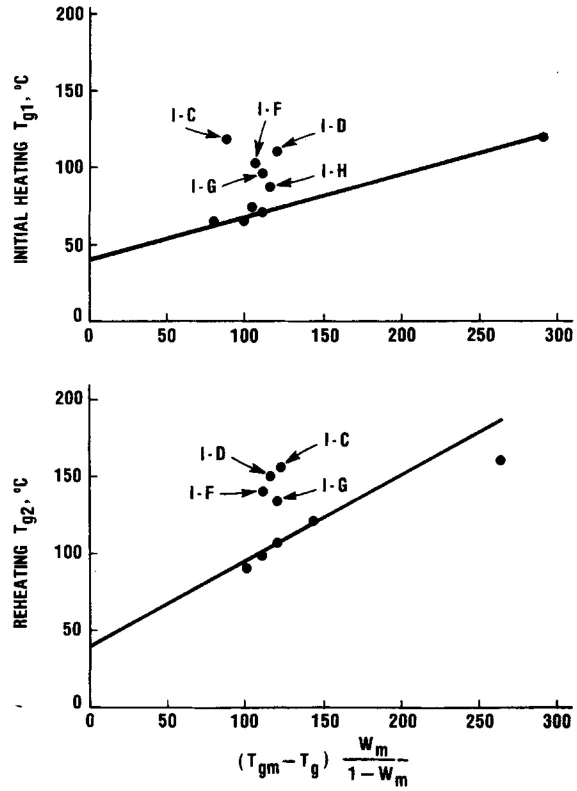

Fig. 2. Wood plot for caprolactam–MXD-I copolymer.

are free of any plasticizing agents (e.g., moisture or organic solvent) or small crystalline regions which may change their apparent values. Without exception, all polymers indicated higher T_{g2} than T_{g1} values. This can be attributed to the removal of any plasticizer upon heating the samples at 240°C. The presence of plasticizers in the polymers prior to this thermal treatment can be responsible for the low T_{g1} values. However, one should not exclude the contribution of increased molecular weight and/or branching, which could have occurred at 240°C, to the high T_{g2} values. A few of the copolyamides with high aliphatic contents (I-I to I-K and II-A to II-G) exhibited very weak, ill-defined endotherms in their initial heating DSC thermograms which may be attributed to the presence of small concentrations of crystalline regions in these systems. However, the T_{g2}/T_{g1} values of these polymers do not reflect any significant effect of the suggested crystalline regions on their transition temperatures. On the other hand, copolymers having both high and low aliphatic contents were shown by x-ray to be essentially amorphous, and, hence, the presence of crystallinity in the highly aliphatic systems becomes rather questionable. As to the effect of the degree of polymerization and composition on the T_g of these copolyamides, it can be generally stated that both T_{g1} and T_{g2} increased with increase in their aromatic content and reduced viscosities.

The effect of composition on the T_g of a variety of random copolymers was investigated by several authors.[22,27–33] Meanwhile, a number of equations based on different physical assumptions were proposed and used successfully to de-

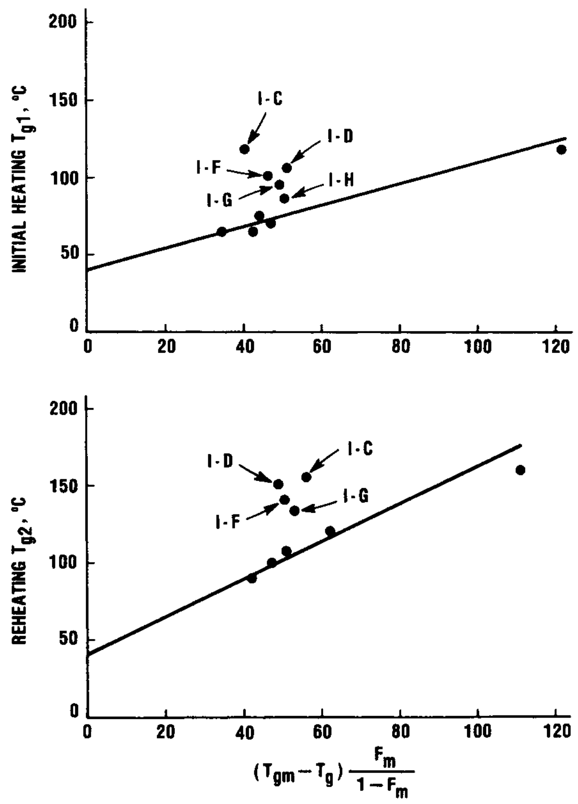

Fig. 3. Wood plot for caprolactam–MXD-I Copolymers in terms of MXD-I mole fraction.

scribe the T_g–composition relationship in amorphous, random copolymers.[29–32] The most widely used equations are those reported by Gordon and Taylor (G-T),[30] Fox,[31] and Mandelkern.[32] In his review of these equations, Wood has shown that by applying the G-T equation to amorphous, random copolymers, a linear relationship can be realized between the T_g and a composition function of these copolymers.[33] This function, $(T_{g_m} - T_g)\ (W_m/1 - W_m)$, is based on the glass temperature of the copolymer (T_g) and that of the corresponding high-T_g homopolymer (T_{g_m}) and the weight fraction of the high-T_g moieties in the copolymers (W_m). Plotting T_g versus this composition function results in the so-called Wood plots. In the present studies, the Wood plots of typical copolymers were constructed and used as a means of recognizing T_g's which cannot be associated with completely random chains.

Copolyamides having comparable reduced viscosities (I-C, I-D, I-F to I-K, and V-A) were chosen for these studies and were examined in conjunction with the MXD-I homopolymer (I-A) and nylon 6 (VI). The necessary data for the construction of the Wood plots are summarized in Table IV. In Figure 2, the composition function of selected copolymers were plotted against their initial heating (T_{g_1}) and reheating (T_{g_2}) glass temperatures. In both cases, straight lines could be drawn between most points, and their intercepts with the T_g axis coincided with the T_g of nylon 6 (~40°C). In both of the T_{g1} and T_{g2} plots, copolymers I-C, I-D, I-E, and I-G exhibited positive derivations from the straight-line relationship. These four samples have between 58 and 78 mole% of the aromatic

TABLE V
Thermogravimetric Analysis Data

Polymer no.	MXD-I,[a] mole-%	η_{red} (H_2SO_4)	Temp. of decomposition onset, °C	Temp. at 50% wt loss, °C	% Residue		
					400°C	425°C	500°C
I-A	100	0.85	370	413	75	38	30
I-B	82	0.78	370	415	80	39	30
I-C	76	0.84	370	413	70	35	29
I-D	69	0.71	365	416	79	38	27
I-E	65	0.97	365	416	79	35	26
I-F	61	0.75	365	418	82	42	28
I-G	58	0.85	360	419	87	42	25
I-H	54	0.86	355	413	72	34	23
I-I	44	0.82	350	415	80	35	18
I-J	39	0.88	355	417	82	35	16
I-K	34	0.78	350	416	78	34	14
II-A	50	1.60	370	422	84	41	23
II-B	46	1.42	360	420	83	43	21
II-E	35	1.08	360	419	84	40	19
II-G	31	0.95	355	420	86	44	19
VI	0	2.76	350	450	93	83	3

[a] MXD-I mole-% of *m*-xylylenediammonium isophthalate in presence or absence of caprolactam.

TABLE VI
Tensile Properties and Water Absorption Data of Typical Copolymers

Polymer no.	MXD-I,[a] mole-%	η_{red} (H_2SO_4)	T_{g_2}[b]	UE, %	UTS, psi $\times 10^{-3}$	SM, psi $\times 10^{-5}$	WB, in.-lb	% Water absorption[c]
I-H	54	0.86	119	2.8	11.97	4.38	0.17	3.82
I-I	44	0.82	107	3.5	12.72	4.30	0.25	3.69
I-J	39	0.88	98	4.1	14.29	4.20	0.31	4.35
I-K	34	0.78	90	4.6	14.65	3.96	0.44	4.30
III-A	88	0.55	157	2.6	11.79	4.27	0.23	—
III-B	88	0.72	154	3.2	12.40	4.42	0.40	—
IV	78	0.68	154	1.4	6.80	4.42	0.10	3.25
V-A	91	0.87	161	3.1	12.12	3.66	0.55	3.30
V-B	91	1.76	167	2.9	9.96	3.67	0.38	3.49

[a] MXD-I mole-% in copolymers based in part on caprolactam.

[b] T_{g_2}: Major transition temperature of reheated polymers (see Table III).

[c] Equilibrium values recorded in a controlled humidity atmosphere of 50% R.H. at 23°C.

moieties, and block formation could be responsible for the recorded deviations. Although the G-T equation was based on the assumption that the free volume/repeat unit is the same in the homopolymer and copolymer, it is suggested that the sequential arrangement in these copolyamides does effect their T_g's. The presence of two glass transition temperatures in the DSC thermograms of I-D, I-G, I-I, II-B, and II-E (Table III) may be associated with the presence of block structures in these copolymers. This is consistent with the proposed presence of block structures in II-B and II-E based on the viscosity data of Figure 1, as described earlier in the discussion. On the other hand, the positive deviation of copolymers I-C, I-D, I-E, and I-G may also be ascribed to a higher degree of branching in these polymers as compared with those which obey the G-T equation. The presence of branching in these copolymers becomes a strong likelihood as one recognizes the documented tendency of similar polyamides to undergo thermally induced degradation, branching, and even crosslinking.[3,34] Constructing the Wood plots using a composition function in terms of the MXD-I mole fraction, as shown in Figure 3, did not seem to improve the T_g composition function correlations as compared with those shown in Figure 2.

The DSC thermograms of MXD-I homopolymer and all the reported compolymers revealed no distinct melting endotherms upon recording the initial heating data. Reheated samples (after quenching the liquid polymer) showed no crystallization exotherms or melting endotherms. Hence, these polymers were considered to be essentially amorphous.

Thermal stability of the copolyamides can be discussed using the TGA data summarized in Table V. These data indicate the effect of composition and molecular weight (in terms of reduced viscosity) on temperature of decomposition onset, temperature at 50% weight loss, and per cent nonvolatile residues at 400, 425, and 500°C. For copolymers having comparable viscosities (I-B to I-K), the decrease in their aromatic content seems to be associated with a general decrease in the temperature of decomposition onset and a decrease in the per cent nonvolatiles at 500°C. Generally speaking, one can, therefore, conclude that the thermal stability of the copolyamides increases with increase in their aromatic content. On the other hand, no particular correlation can be established between the aromatic content of the copolyamides and the temperature at 50% weight loss or the per cent residues at 400 and 425°C. As to the effect of molecular weight on the thermal stability of the copolyamides, the data for two pairs of copolymers (I-I versus II-B and I-K versus II-E) having comparable compositions but different viscosities indicate explicitly that the thermal stability increases with increase in molecular weight.

The tensile properties and water absorption data of typical copolymers are summarized in Table VI and can be discussed as follows. The data of copolymers III-A and III-B indicate a noticeable improvement in the tensile properties with a modest increase in molecular weight. Copolymer III-B exhibited a higher UE, UTS, WB, and SM than III-A. On the other hand, an analogous effect of molecular weight on the properties of V-A and V-B was not observed. The unexpected inferior tensile properties of high-viscosity V-B as compared to V-A may be ascribed to the extensive chain branching in the former copolymer. This is consistent with the observed higher affinity of V-B to absorb water, as compared with V-A. Due to the noticeable dependence of the tensile properties on chain dimensions, a meaningful conclusion as to the effect of composition on these properties should not be expected for the copolyamides due to their variable viscosities. However, it is apparent that copolyamides IV, V-A, and V-B are less hydrophilic than nylon 6, which is reported to have a moisture content at saturation of 10.9%.[35] This can be ascribed to their high aromatic contents.

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