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Synthesis and Polycondensation of Some New Organic Tellurium Compounds Containing Mono- and Di-amino Groups

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Several new and known organic tellurium compounds containing amino groups (i.e. $ArTeBr_3$, Ar_2Te_2 and Ar_2Te, where Ar = 4-$NH_2C_6H_4$, 2-$NH_2C_6H_4$, 4-$CH_3CONHC_6H_4$ or 2-NH_2-5-NO_2-C_6H_3) were prepared by reacting aminoarylmercury chlorides with tellurium tetrabromide in glacial acetic acid. Bis(4-aminophenyl) telluride and bis(2-amino-5-nitrophenyl) telluride were polymerized with aromatic and aliphatic diacid chlorides (i.e. terephthaloyl chloride and sebacoyl chloride), as well as with toluene diisocyanate, leading to new organic tellurium polyamides and polyurea. All organic tellurium compounds and their condensation polymers were characterized by elemental analyses, IR, ^1H and ^{13}C NMR, and mass spectroscopy. The thermal stabilities of the resulting polymers were determined by thermogravimetric and derivative thermogravimetric techniques. © 1997 John Wiley & Sons, Ltd.

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INTRODUCTION

Few organic tellurium compounds containing an amino group have been reported in the literature. McWhinnie and co-workers[1, 2] prepared bis(*o*-aminophenyl)ditelluride by reduction of (2-phenylazo)phenyltellurium trichloride with sodium tetrahydroborate ($NaBH_4$). 2-Aminophenyl ethyl telluride was obtained by reduction of 2-nitrophenyl ethyl telluride.[3] Engman *et al.*[4, 5] synthesized amino-substituted diphenyl tellurides via the reduction of 2:1 complexes of anilines and telurium tetrachloride by sodium disulfite. Our recent work described the synthesis of novel organic tellurium compounds, with an amino group *ortho* to the tellurium atom,[6] from 2-aminoarylmercury chloride with telurium tetrabromide.

To the best of our knowledge, polymers containing organic tellurium monomers have not been previously described.[7, 8] The objective of this study was to synthesize amino-substituted organic tellurium compounds and to prepare from them polymers via solution polycondensation techniques.

EXPERIMENTAL

Synthesis

2-Aminophenylmercury chloride[9] and 4-aminophenylmercury chloride[9] were prepared by published procedures.

4-Aminophenyltellurium tribromide (1)
A solution of 4-aminophenylmercury chloride (2.60 g; 8 mmol) and tellurium tetrabromide (1.97 g; 8 mmol) in glacial acetic acid (60 cm^3) was refluxed for 14 h. The solution was filtered hot. After cooling to room temperature, the yellow precipitate that had formed was collected by filtration. Crystallization from ethanol/hexane (9:1) gave yellow crystals, m.p. 105–106 °C. Yield: 52% (1.90 g).

Analysis: Calcd for $C_6H_6Br_3NTe$: C, 15.69; H, 1.32; N, 3.05. Found: C, 15.41; H, 1.21; N, 2.78%. ^1H NMR (DMSO-d_6) (ppm): 3.42 (s, br,

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2H, NH_2); 6.50 (d, J = 6.5 Hz, 4H, Ar–H); 7.24 (d, J = 6.3 Hz, 2H, Ar–H).

Bis(4-aminophenyl) telluride (2)
This compound was prepared by two methods, as follows.

(a) A solution of 4-aminophenylmercury chloride (1.97 g; 6 mmol) and tellurium tetrabromide (1.34 g; 3 mmol) in dry dioxane (50 cm^3) was refluxed for 4 h. The hot mixture was filtered. On cooling, the HgClBr·2diox complex separated as white crystals and was removed by filtration. The filtrate was evaporated in a rotary evaporator on a water bath (60 °C) under 4 Torr vacuum to leave a dark-brown residue. This residue was dissolved in toluene (30 cm^3). To the refluxing solution, hydrazine hydrate (1.60 g) in methanol (30 cm^3) was added dropwise until evolution of nitrogen had ceased. The resulting solution was poured into water (1 dm^3). The brown solid that had formed was recrystallized from petroleum ether (60–80 °C) to give brown crystals, m.p. 108–110 °C. Yield: 70% (0.38 g).

(b) To bis(4-aminophenyl) ditelluride (0.88 g; 2 mmol) dissolved in dioxane (15 cm^3), activated copper powder 0.38 g; 6 mmol) was added. The mixture was refluxed for 8 h and then filtered hot. The solvent was removed in a rotary evaporator at 40 °C under 4 Torr vacuum to leave a brown residue, which was recrystallized from petroleum ether (60–80 °C) to give brown crystals, m.p. 109–110 °C. Yield: 79% (0.44 g).

Analysis: Calcd for $C_{12}H_{12}N_2Te$: C, 46.22; H, 3.88; N, 8.98. Found: C, 46.07; H, 3.87; N, 8.98%. ^{1}H NMR (DMSO-d_6) (ppm): 3.66 (sb, 4H, NH_2); 6.81 (d, J = 6.7 Hz, 4H, Ar–H); 7.48 (d, J = 6.5 Hz, 4H, Ar–H).

Bis(4-aminophenyl) ditelluride (3)
To a suspension of 4-aminophenyltellurium tribromide (2.76 g; 6 mmol) in water (50 cm^3) was added sodium disulfite (5.70 g; 55 mmol) at 0 °C. The resulting mixture was stirred for 30 min at 0 °C and then extracted with chloroform (3 × 100 cm^3). The combined extracts were dried over anhydrous calcium chloride for 4 h and filtered. The filtrate was evaporated to dryness on a rotary evaporator at 40 °C under 4 Torr vacuum. The residue was dissolved in hot ethanol and the solution filtered. Hexane was dropped onto the filtrate until the solution became turbid. The solution was left in a refrigerator at −4 °C for 16 h. Golden-yellow crystals were obtained, m.p. 80–82 °C (Lit.[4, 5] 79–81 °C).

Analysis: Calcd for $C_{12}H_{12}N_2Te_2$: C, 32.80; H, 2.75; N, 6.37. Found: C, 32.50; H, 2.97; N, 5.95%.

Bis(4-acetamidophenyl) ditelluride (4)
Acetic anhydride (15 cm^3) was added to bis(4-aminophenyl) ditelluride (**3**) (0.35 g; 0.8 mmol). The reaction vessel was covered and set aside for 5 h. Ice–water was added to the solution. A deep red precipitate formed and was collected by filtration. The precipitate was washed several times with water (4 × approx. 15 cm^3) and recrystallized from ethanol–hexane to give red crystals of bis(4-acetamidophenyl) ditelluride (**4**) in 79% yield, m.p. 180–181 °C (Lit.[10] 172 °C and Lit.[11] 196–198 °C).

Analysis: Calcd for $C_{16}H_{16}N_2O_2Te_2$: C, 36.71; H, 3.08; N, 5.35. Found: C, 36.54; H, 3.03; N, 5.26%. ^{1}H NMR (DMSO-d_6) (ppm): 2.08 (s, 6H, CH_3CO); 7.43 (d, J = 7.11 Hz, 4H, Ar–H); 7.67 (d, J = 6.9 Hz, 4H, Ar–H); 9.99 (sb, 2H, NH).

2-Aminophenyltellurium tribromide (5)
2-Aminophenylmercury chloride (1.32 g; 4 mmol) and tellurium tetrabromide (1.80 g; 4 mmol) were refluxed in glacial acetic acid (60 cm^3) for 14 h. The reaction mixture was treated as described for the preparation of 4-aminophenyltellurium tribromide (**1**), to give yellow crystals, m.p. 261–262 °C (dec). Yield: 60% (1.11 g).

Analysis: Calcd for $C_6H_6Br_3NTe$: C, 15.69; H, 1.32; N, 3.05. Found: C, 15.37; H, 1.18; N, 2.83%.

Bis(2-aminophenyl) telluride (6)
This compound was prepared by following the same procedures as for bis(4-aminophenyl) telluride (**2**).

(a) 2-Aminophenylmercury chloride (1.32 g; 4 mmol) and tellurium tetrabromide (0.90 g; 2 mmol) were refluxed in dioxane (50 cm^3) for 4 h. The reaction mixture was treated as described for compound **2**. M.p. 117–118 °C. Yield: 70% (0.43 g).

(b) Bis(2-aminophenyl) ditelluride (0.26 g; 0.6 mmol) was treated with activated copper (0.12 g; 1.8 mmol) in dry dioxane (30 cm^3) and as described for the preparation of bis(4-aminophenyl) telluride (**2**). Brown crystals, m.p.

117–118 °C. Yield: 84% (0.16 g).

Analysis: Calcd for $C_{12}H_{12}N_2Te$: C, 46.22; H, 3.88; N, 8.98. Found: C, 45.91; H, 3.73; N, 8.84%.

Bis(2-aminophenyl) ditelluride (7)
2-Aminophenyltellurium tribromide (1.38 g; 3 mmol) was reduced with sodium disulfite (2.83 g; 15 mmol) at 0 °C. The mixture was worked up as described for bis(4-aminophenyl) ditelluride (**3**) to give red crystals, m.p. 104–105 °C (Lit.[1] 100 °C and Lit.[4, 5] 104–105 °C). Yield: 70% (0.93 g).

Analysis: Calcd for $C_{12}H_{12}N_2Te_2$: C, 32.80; H, 2.75; N, 6.37. Found: C, 32.63; H, 2.48; N, 6.12%.

Bis(2-amino-5-nitrophenyl) telluride (8)
A solution of 2-amino-5-nitrophenylmercury chloride (4.3 g; 12.5 mmol) and 2-amino-5-nitrophenyltellurium tribromide (6.3 g; 12.5 mmol) in dioxane (100 cm^3) was refluxed for 4 h. The hot mixture was filtered. The filtrate was cooled to room temperature. The HgClBr·2diox complex was removed by filtration. The filtrate was evaporated to dryness on a rotary evaporator at 40 °C under 4 Torr vacuum. A brown precipitate of bis(2-amino-5-nitrophenyl)tellurium dibromide was obtained. The product was dissolved in hot methanol (150 cm^3). To the refluxing solution, a solution of hydrazin hydrate (3.6 g) in methanol (100 cm^3) was added slowly until the evolution of nitrogen had ceased. The solution was then filtred and the filtrate cooled to room temperature to afford brown crystal that were recrystallized from benzene, m.p. 226–228 °C (Lit.[6] 228 °C).

Analysis: Calcd for $C_{12}H_{10}N_4O_4Te$: C, 35.87; H, 2.51; N, 13.94. Found: C, 35.66; H, 2.42; N, 13.81%.

Polycondensation of bis(4-aminophenyl) telluride and terephthaloyl chloride (9)
A solution of bis(4-aminophenyl) telluride (**2**) (0.05 g; 0.5 mmol) in dry dichloromethane (20 cm^3) was added to a stirred solution of terephthaloyl chloride (0.04 g; 0.2 mmol) in dry dichloromethane (20 cm^3). The mixture was vigorously stirred for 30 min. A precipitate formed. The contents of the flask were poured into dry hexane (300 cm^3). The resulting white precipitate was collected by filtration, washed with water then with acetone, and dried *in vacuo* at 60 °C to give a yellowish-brown solid in 50% yield (0.05 g), m.p. >340 °C (dec.).

Analysis: Calcd for $(C_{20}H_{14}N_2O_2Te)$: C, 54.36; H, 3.19; N, 6.34. Found: C, 48.95; H, 3.60; N, 5.63%.

Polycondensation of bis(4-aminophenyl) telluride and sebacoyl chloride (10)
Bis(4-aminophenyl) telluride (**2**) (0.06 g; 0.2 mmol) was treated with sebacoyl chloride (0.05 g; 0.2 mmol) as described for the preparation of compound **9**. A white solid was obtained in 55% yield, m.p. 249–252 °C.

Analysis: Calcd for $(C_{22}H_{26}N_2O_2Te)_n$: C, 55.27; H, 5.48; N, 5.86. Found: C, 52.28; H, 5.76; N, 5.81%.

Polycondensation of bis(2-amino-5-nitrophenyl) telluride and terephthaloyl chloride (11)
Bis(2-amino-5-nitrophenyl) telluride (**8**) (0.62 g; 1.55 mmol) was treated with terephthaloyl chloride (0.32 g; 1.55 mmol) in the presence of triethylamine(0.40 g; 3.88 mmol) as described above. A yellow solid was obtained in 51% yield (0.48 g), m.p. 281–284 °C (dec.).

Analysis: Calcd for $(C_{20}H_{12}N_4O_6Te)_n$: C, 45.16; H, 2.27; N, 10.53. Found: C, 44.62; H, 2.43; N, 9.78%.

Polycondensation of bis(2-amino-5-nitrophenyl) telluride and sebacoyl chloride (12)
Bis(2-amino-5-nitrophenyl) telluride (0.62 g; 1.55 g) was treated with sebacoyl chloride (0.37 g; 1.55 mmol) in the presence of triethylamine (0.40 g; 3.88 mmol) as described above. A yellow solid was obtained in 54% yield (0.53 g), m.p. 233–236 °C.

Analysis: Calcd for $(C_{22}H_{24}N_4O_6Te)_n$: C, 46.52; H, 4.26; N, 9.86. Found: C, 46.96; H, 3.96; N, 10.42%.

Polycondensation of bis(2-amino-5-nitrophenyl) telluride and toluene di-isocyanate (TDI) (13)
Into a three-necked, round-bottomed flask was placed 0.27 g (1.55 mmol) of toluene di-isocyanate (80% 2,4- and 20% 2,6-isomer) in dry chloroform (60 cm^3). Bis(2-amino-5-nitrophenyl) telluride (0.62 g; 1.55 mmol) in dry chloroform (40 cm^3) was added with stirring at

room temperature. Immediately, a yellow precipitate formed. Stirring was continued for 30 min. The precipitate was collected by filtration, washed with dry chloroform, and dried. The yellow product was washed several times with hot dry toluene and then with dry diethyl ether and dried *in vacuo* at 60 °C. The resulting yellow solid 92% yield (0.82 g) melted at 285–288 °C.

Analysis: Calcd for $(C_{21}H_{16}N_6O_6Te)_n$: C, 43.79; H, 2.80; N, 14.59. Found: C, 43.27; H, 2.51; N, 14.95%.

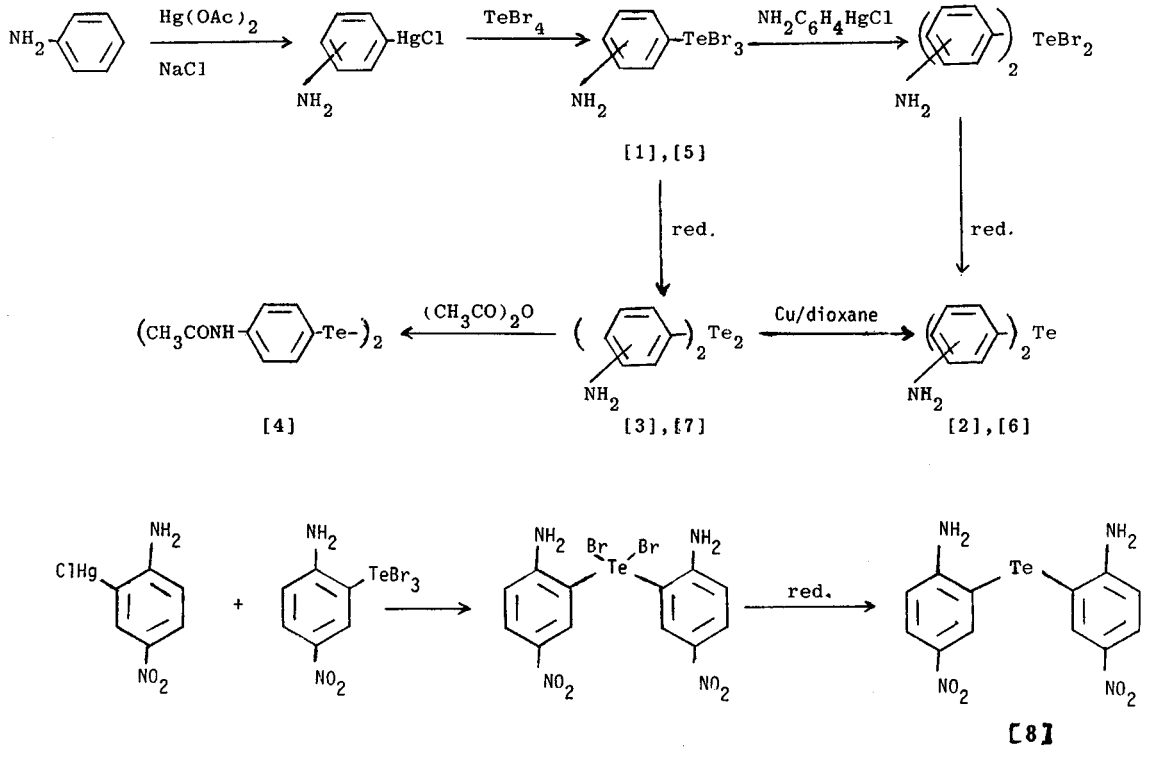
Physical measurements

IR spectra were recorded as KBr discs in the range 4000–200 cm^{-1} on a Pye–Unicam SP3–300s IR spectrophotometer. NMR spectra were obtained in a JEOL GSX–270 (270 MHz) and Brucker–250 (250 MHz) as solutions in DMSO-d_6 or $CDCl_3$ with tetramethylsilane (TMS) as internal standard. Microanalyses for carbon, hydrogen and nitrogen were carried out with a Hearaus HP–85 instrument. Melting points were determined on a Gallenkamp melting point apparatus and are uncorrected. The thermogravimetric (TG) and derivative thermogravimetric (DTG) analyses were obtained on a Du Pont 1090 thermal analyzer. All measurements were carried out under nitrogen (flow rate 50 $cm^3\ min^{-1}$) and against standard α-Al_2O_3. Mass spectra were obtained with a Finnigan instrument (Universität Konstanz, Germany) at 70 eV.

RESULTS AND DISCUSSION

The present work describes the synthesis of some organic telurium compounds containing amino groups by reacting arylmercury chloride with tellurium tetrabromide in glacial acetic acid to produce the required telurium-containing materials. Thus, tellurium tetrabromide and 4- or 2-aminophenylmercury chloride are capable of producing 4- or 2-aminophenyltellurium tribromide when brought together in a 1:1 molar ratio in glacial acetic acid (Scheme 1).

Bis(4-aminophenyl) telluride (**2**) and bis(2-minophenyl) telluride (**6**) were prepared either



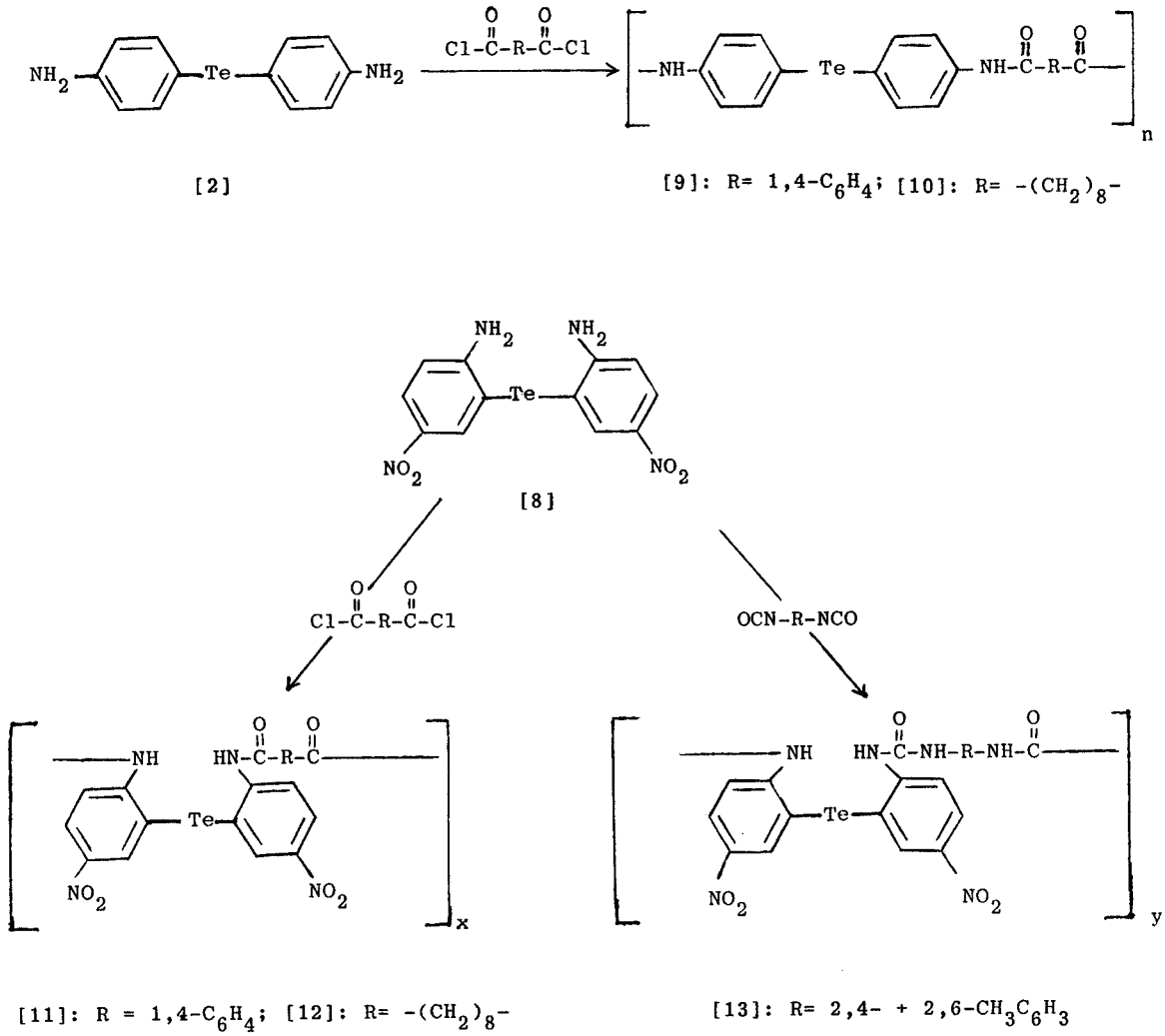
Scheme 1

by reduction of the corresponding bis(4-aminophenyl)tellurium dibromide or bis(2-aminophenyl)tellurium dibromide (not isolated) by potassium disulfite in aqueous medium at 0 °C, or by refluxing the corresponding ditellurides (i.e. **3** and **7**) in dioxane for 8 h in the presence of a three-fold molar excess of activated copper powder to give the tellurides **2** and **6** in 70% and 75% yield, respectively (Scheme 1).

Reduction of 4-aminophenyltellurium tribromide (**1**) and 2-aminophenyltellurium tribromide (**5**) with potassium disulfite in aqueous medium at 0 °C yielded the ditellurides **3** and **7** in 60% and 70% yield, respectively.

Bis(4-acetamidophenyl) ditelluride (**4**) is a known compound and had been prepared in 7% yield by reduction of 4-acetamidophenyltellurium trichloride by $K_2S_2O_5$.[10, 11] In the present work compound **4** was prepared in 79% yield by reacting compound **3** with acetic anhydride (Scheme 1).

The IR spectra of **1**–**3** and **5**–**8** gave characteristic bands that are quite similar to those of organomercury chloride derivaties, confirming that telluration has occurred at the position initially occupied by HgCl. These compounds showed medium bands due to NH_2 asymmetric and symmetric stretching vibrations (ν_{as} and ν_s) in the range 3290–3485 cm^{-1} and 3180–3380 cm^{-1}, respectively.[12–15] The spectra showed strong bands in the range



Scheme 2

1610–1690 cm^{-1} due to the internal deformation mode of N–H. In general, the NH_2 bands were shifted to lower frequencies compared with aniline.[14, 15] This is probably due to donation of the lone pair of electrons by nitrogen to tellurium through intra- or/and inter-molecular interactions.

The ^1H NMR spectra of **1**, **2** and **4** were recorded in DMSO-d_6. In the spectra of **1** and **2**, broad resonances at 3.42 and 3.66 ppm are attributed to NH_2 protons, whereas compound **4** shows a broad singlet at 9.99 ppm due to NH protons. The aryl protons appeared as doublets, as expected for 1,4-disubstituted phenyl rings. The ^{13}C NMR spectrum of bis(4-aminophenyl) telluride (**2**) is in agreement with its structure.

The mass spectrum of bis(4-acetamidophenyl) ditelluride (**4**) shows a molecular ion at *m/z* 528 with a relative intensity of 15.4%, together with peaks at the following *m/z* values: 398, 268, 226, 184, 156, 92 and 65.

In general, compounds **2**, **3**, **4** and **7** were prepared previously by indirect methods.[1, 4, 5] Our method gave tellurated anilines by a direct method and in good yields.

Solution polycondensation of **2** and **8** with acid chlorides and toluene di-isocyanate, with stirring, was the general procedure[16–18] used for the preparation of the new tellurium-containing polyamides and polyurea (Scheme 2). The polymerization reactions were carried out at ambient temperature, in contrast to the polycondensation of organometallic compounds.[16, 17] The results of CHN analyses of polymers **11**, **12** and **13** are in reasonable agreement with calculated values (see the Experimental section). The discrepancies in the results for polymers **9** and **10** could be attributed to the fact that some branching can occur by a secondary reaction of the acid chloride with the amide group to yield an imide structure.[19, 20]

The IR spectra of the new polyamides and polyurea (i.e. polymers **9**, **10**, **11** and **12** and polymer **13**) showed broad intense N–H stretching vibrations in the range 3300–3380 cm^{-1} and a strong carnbonyl stretching in the range 1640–1720 cm^{-1}. The amide II band was evident near 1540 cm^{-1}. In addition, aromatic C–H stretching occurred around 3080 cm^{-1} and asymmetric and symmetric aliphatic C–H stretches around 2930 cm^{-1} and 2880 cm^{-1}, respectively, were also observed.

The ^1H NMR spectrum of polyurea **13** showed a singlet at 1.97 ppm due to methyl protons of TDI, and multiplets at 6.40–9.10 ppm due to aromatic protons. The signal of the N–H proton and phenyl protons overlapped. The ^1H NMR spectrum of **13** showed signals due to the presence of ether and toluene molecules.

The ^{13}C NMR spectrum of **13** in DMSO-d_6 solution showed signals due to ether and toluene residuals. In general, the spectrum showed the aromatic carbon atoms within the expected range. The $\delta(C=O)$ signal appeared at the range 152.4–153.3 ppm. The ^1H and ^{13}C spectral data are in good agreement with the values reported for similar compounds.[12–15]

The low solubilities of these polymers in any common solvents made conventional viscometric, osmotic and light-scattering techniques inapplicable for molecular mass determinations.

The thermal stability characteristics of polymers **11**, **12** and **13** are presented in Table 1. The percentage of char content was determined at 700 °C. In general these polymers show good thermal stability in comparison with the corresponding aliphatic and aromatic polymers.[16, 18–20]

To evaluate the ability of these new polymers as conducting polymers, the solid conductivity of **11** was measured at room temperature ($1.07\times10^{-13}\ \Omega^{-1}\ cm^{-1}$). This polymer is an insulator at room temperature.

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Table 1 Thermal stability characteristics of new polymers containing tellurium

Polymer	Decomposition temperature, DT (°C)[a] T_i	T_{max}	T_f	Char content at 700 °C (%)
11	270	305	365	58.75
12	165[b]	210[b]	245[b]	33.75
	315[c]	350[c]	365[c]	
	570[d]	620[d]	660[d]	
13	310[b]	325[b]	340[b]	33.75
	365[c]	395[c]	445[c]	

[a] T_i = initial DT, the temperature at which the polymers start to decompose and intimate weight loss occurs; T_{max} = maximum DT, the temperature at which the decomposition reaction reaches optimum; T_f = final DT, the temperature at which the decomposition reactions are complete. [b] First DT. [c] Second DT. [d] Third DT.

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