Addition of *p*-Toluenesulfonyl Isocyanate to Imino Ethers. Isolation of a Stable 1,4-Dipolar Intermediate

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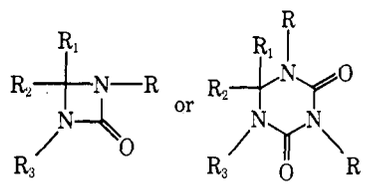
The reaction between *p*-toluenesulfonyl isocyanate and methyl *N*-*tert*-butylformimidate (1) forms a 2:1 cycloadduct (2). The azetines **3–5** form analogous 2:1 cycloadducts (**6–8**) under identical conditions. With an excess of **3**, the 1,4-dipolar intermediate **10** can be isolated. It is characterized by conversion to **6** with *p*-toluenesulfonyl isocyanate and by hydrolysis to the urea **11**.

In connection with studies on cycloaddition reactions of imino ethers,[1] we have investigated the addition reactions of isocyanates. The reactions with 2-alkoxy-1-azetines are of special interest because they have permitted the isolation of a stable 1,4-dipolar intermediate.

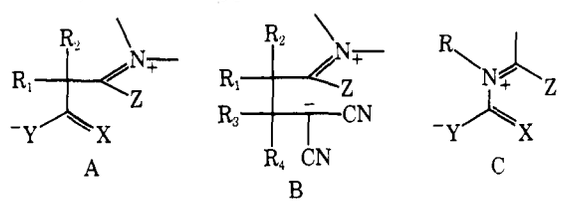
Isocyanates have been found to add to carbon–nitrogen double bonds to give 1:1 1,3-diazetidinone adducts in many

$$R_1R_2C{=}NR_3 + RN{=}C{=}O \longrightarrow$$



cases.[2-12] When there are abstractable hydrogens α to the C=N bond, 1:1 ene-type adducts are often formed.[13-21] There have also been many cases in which 2:1 adducts have been formed by addition of a second molecule of isocyanate to a hypothetical 1,4-dipolar intermediate.[3,4b,22-34] In the case of unhindered imines, 1:2 adducts incorporating two molecules of imine are sometimes found.[12,25,35-37]

The intermediacy of 1,4-dipolar ions has been suggested[2d] to account for these reactions. Such intermediates appear to be trapped as 2:1 adducts, 1:2 adducts, and, in a few cases, 1:1:1 adducts with a third component.[22,25,33,34,38] Gompper has isolated and characterized a number of stable 1,4 (or 1,5) unconjugated dipolar ions of type A from reactions of ketene *N,S*-acetals and enamines with heterocumulenes,[29-42] or of type B with electron-deficient ole-

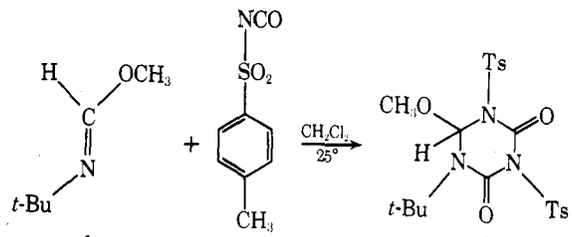


A B C

fins.[43] The cross-conjugated dipolar ions of type C, however, have not been previously isolated and characterized,[44,45] although they appear to have been trapped in additions of ketenes to imines with added water, methanol, and sulfur dioxide.[44b,46] We report here additions of *p*-toluenesulfonyl isocyanate to 2-alkoxy-1-azetines,[47] which give an isolable and characterizable cross-conjugated 1,4-dipolar ion of type C for the first time.

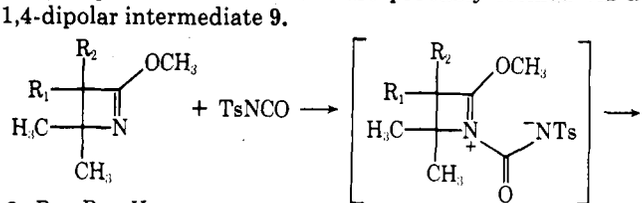
Results and Discussion

Treatment of the somewhat hindered aliphatic imino ether **1** with 2 equiv of *p*-toluenesulfonyl isocyanate results in an 86% yield of 1,3-di-*p*-toluenesulfonyl-5-*tert*-butyl-6-methoxytriazine-2,4-dione (**2**), a normal 2:1 cycloadduct. This is analogous to the reactions of other imines and isocyanates.[5-7,9,12] The 2-alkoxy-1-azetines **3–5** give similar



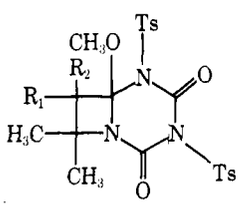
1 Ts = $-SO_2C_6H_4CH_3$ 2

2:1 cycloadducts **6–8** in high yields on treatment with 2 equiv of *p*-toluenesulfonyl isocyanate. These crystalline adducts are well characterized by their spectral data but decompose readily on melting or standing. They are typical of 1,4-dipolar additions[2d] and are probably formed via a 1,4-dipolar intermediate **9**.



3, $R_1 = R_2 = H$
4, $R_1 = R_2 = CH_3$
5, $R_1 = H$; $R_2 = CH_3$

9



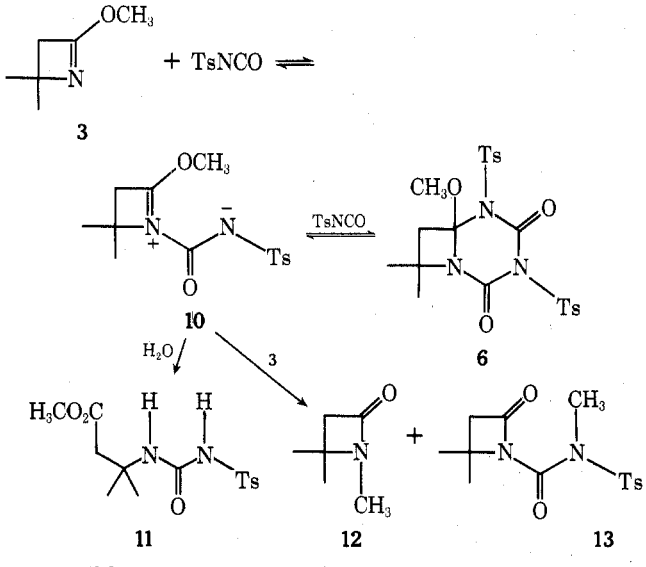
6, $R_1 = R_2 = H$
7, $R_1 = R_2 = CH_3$
8, $R_1 = H$; $R_2 = CH_3$

Upon treatment of *p*-toluenesulfonyl isocyanate with excess azetine **3** at 0°, a white solid precipitates. The white solid is an amorphous powder which decomposes on melting and is quite insoluble in organic solvents. The mass spectrum of the solid indicates only peaks from **3** and *p*-toluenesulfonyl isocyanate, to which the solid decomposes thermally. The infrared spectrum (in Nujol) shows two peaks of medium intensity at 1690 and 1645 cm^{-1}. Other 1,4-dipoles have also shown peaks in the 1600–1700-cm^{-1} range.[39-43] There were no infrared peaks in the region expected for a 1,3-diazetidinone. This white powder has been assigned the 1,4-dipolar ion structure **10** on the basis of these physical properties and the chemical interconversions that follow below.

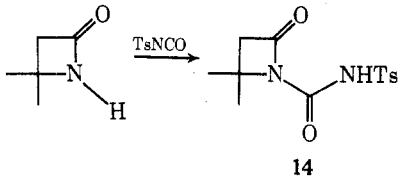
To confirm the structure of **10** and the 1,4-dipolar mechanism for the formation of **6** from **3**, *p*-toluenesulfonyl isocyanate was added to **10** and the 2:1 cycloadduct **6** isolated in 96% yield. The zwitterion **10** is moisture sensitive and readily adds water in the expected fashion. The ester urea

11 is isolated in 97% yield from addition of water to 10 or exposure of 10 to moist air, providing strong evidence for the dipolar structure 10. The azetine 3 is stable to aqueous hydrolysis under the experimental conditions and the p-toluenesulfonyl isocyanate hydrolyzes to p-toluenesulfonamide, a minor side product in these reactions.

The 2:1 adduct 6 also reacts with water to give the urea 11 and p-toluenesulfonamide. This demonstrates that 6



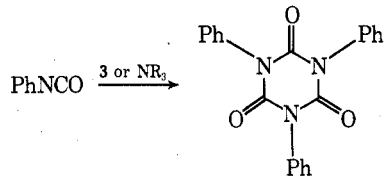
can equilibrate with the 1,4-dipolar ion 10 and isocyanate. The dipolar ion 10 decomposes on attempted dissolution in dichloromethane to a complex mixture containing 12% of 3, some p-toluenesulfonyl isocyanate, 37% of 6, 27% of 12, 12% of 13, and traces of urea 11 (8%) and p-toluenesulfonamide (~2%) from water hydrolysis. The composition of this mixture further supports the 1:1 composition assigned to 10 and clearly demonstrates that the dipolar ion 10 can readily equilibrate with imino ether 3 and isocyanate. The two amides 12 and 13 are formed under a variety of conditions in variable ratios. They appear to be formed in a Lewis acid catalyzed intermolecular alkylation mechanism.[48] The p-toluenesulfonyl isocyanate probably acts as the catalyst in these reactions via 10 as the initial alkylating agent. The structure of 13 is supported by its spectral similarities with the related urea 14 formed from addition



of p-toluenesulfonyl isocyanate to 4,4-dimethylazetidin-2-one.

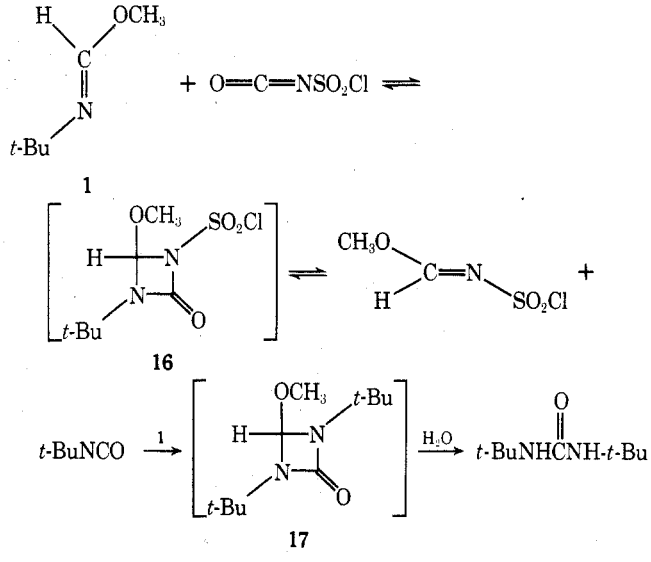
In an attempt to produce a 1:2 cycloadduct composed of two molecules of azetine 3 and one of p-toluenesulfonyl isocyanate, analogous to other imines,[12,35-37] 3 was added to 10. The only products detected and isolated were the amides 12 and 13, in about a 1:1 ratio. With the low concentration of p-toluenesulfonyl isocyanate no 2:1 adduct 6 was observed, but no 1:2 adduct was seen either. Similarly, a 2:1 mixture of imino ether 3 and isocyanate give a 1:1 mixture of 12 and 13. In an attempt to prepare a 1:1:1 cycloadduct, phenyl isocyanate was added to 10, but the only products found were the 2:1 cycloadduct 6 and amide 12.

To test the generality of these reactions with p-toluenesulfonyl isocyanate, the reaction between the less reactive phenyl isocyanate and 3 was tried. It gave only a white solid precipitate within minutes at 25°. The crystalline product was determined by NMR to be the trimer of phenyl isocyanate 15[2,3] complexed with ca. two molecules of



acetonitrile.[49,50] No adducts with 3 could be detected. From control experiments, no trimerization of phenyl isocyanate occurs in pure acetonitrile, but upon the addition of a tertiary amine, trimerization takes place overnight at 25°. The azetine 3 is therefore acting as a catalyst[22] for the trimerization process.

Finally, the very reactive chlorosulfonyl isocyanate was added to imino ether 1. Imino ether 1 was used because it most closely resembles the anils that have been added to chlorosulfonyl isocyanate to give 2:1 cycloadducts.[28] The two reagents were mixed in dichloromethane at −70°; work-up gave only a small amount (1.4%) of a solid identified as N,N'-di-tert-butylurea by comparison of spectral data with the literature.[51,52] The product can be rationalized as originating from a disproportionation sequence proceeding through a reactive 1:1 cycloadduct 16. Analogous disproportionations have been shown to occur in other systems.[2,10] Subsequent reaction with 1 and hydrolysis could give the urea. The cycloadduct 17 is postulated to give rise to the urea. The aliphatic tert-butyl isocyanate adds to 1, since it does not form dimers or trimers in base like phenyl isocyanate.



Experimental Section

All boiling points and melting points are uncorrected. Ir spectra were obtained in solution with a matched reference cell on a Perkin-Elmer 337 grating infrared spectrophotometer. NMR spectra were obtained on a 60-MHz Varian Associates T-60 spectrometer. Mass spectra were obtained on a MS-902 spectrometer.

Materials. The phenyl isocyanate and chlorosulfonyl isocyanate (CSI) were purchased from Aldrich Chemical Co. The p-toluenesulfonyl isocyanate was purchased from Eastman Chemical Co. and distilled before using. The imino ethers were made by alkylation of the corresponding amide or lactam with trimethyloxonium fluoroborate.[1,47]

Reaction of p-Toluenesulfonyl Isocyanate with 1. To a mixture of 990 mg (5.02 mmol) of p-toluenesulfonyl isocyanate, 4 ml of dichloromethane, and 50 mg of potassium carbonate at −20° was added a solution of 260 mg (2.26 mmol) of 1 in 1 ml of dichloromethane. The reaction was allowed to come to room temperature over a 2-hr period. After filtering off the potassium carbonate, the

organic solution was washed with aqueous sodium bicarbonate and dried over anhydrous potassium carbonate. Evaporation of the solvent in vacuo resulted in a solid. Recrystallization in carbon tetrachloride gave 1.07 g (86%) of 1,3-di-*p*-toluenesulfonyl-5-*tert*-butyl-6-methoxy-1,3,5-triazine-2,4-dione (**2**): mp 135–137° dec; ir (CH_2Cl_2) 2920, 1760 (sh), 1730 (s), 1605, 1380, 1340 cm^{-1}; NMR (CH_2Cl_2) δ 1.48 (s, 9 H), 2.45 (s, 6 H), 3.44 (s, 3 H), 6.51 (s, 1 H), 7.40 (m, 4 H), 8.10 (m, 4 H). The mass spectrum showed only peaks from decomposition to *p*-toluenesulfonyl isocyanate and **1** because **2** is thermally unstable and decomposes before vaporization.

Reaction of *p*-Toluenesulfonyl Isocyanate with 4. Following the procedure for **2**, treatment of 64 mg (0.45 mmol) of **4** with 172 mg (0.87 mmol) of *p*-toluenesulfonyl isocyanate gives a near quantitative yield of a 2:1 adduct by NMR analysis. The adduct decomposed upon aqueous bicarbonate treatment. The spectral data fit 3,5-di-*p*-toluenesulfonyl-6-methoxy-7,7,8,8-tetramethyl-1,3,5-triazabicyclo[4.2.0]octa-2,4-dione (**7**): ir (CH_2Cl_2) 2960, 1745, 1720, 1600, 1385 cm^{-1}; NMR (CH_2Cl_2) δ 1.18 (s, 3 H), 1.35 (s, 3 H), 1.40 (s, 3 H), 1.43 (s, 3 H), 2.44 (m, 6 H), 3.18 (s, 3 H), 7.40 (m, 4 H), 8.05 (m, 4 H).

Reaction of *p*-Toluenesulfonyl Isocyanate with 5. Following the procedure for **1**, treatment of 70 mg (0.55 mmol) of **5** and 221 mg (1.12 mmol) of *p*-toluenesulfonyl isocyanate gives a near quantitative yield of an adduct by NMR analysis. Crystallization from carbon tetrachloride gave 102 mg (36%) of an unstable adduct, 3,5-di-*p*-toluenesulfonyl-6-methoxy-7,8,8-trimethyl-1,3,5-triazabicyclo[4.2.0]octa-2,4-dione (**8**): mp 135–143° dec; ir (CCl_4) 2950, 1755 (sh), 1730 (s), 1600, 1390, 1350 cm^{-1}; NMR (CCl_4) δ 1.17 (d, J = 7 Hz, 3 H), 1.33 (s, 3 H), 1.47 (s, 3 H), 2.43 (s, 6 H), 3.18 (s, 3 H), 7.40 (m, 4 H), 8.00 (m, 4 H) (the methine hydrogen was not detected). The mass spectrum showed only peaks from decomposition to *p*-toluenesulfonyl isocyanate and **5**, again because **8** is thermally unstable and decomposes before vaporization.

Reaction of *p*-Toluenesulfonyl Isocyanate with 3. To a solution of 326 mg (1.65 mmol) of *p*-toluenesulfonyl isocyanate in 4 ml of dichloromethane was added a solution of 92 mg (0.82 mmol) of **3** in 1 ml of dichloromethane at −20°. After reaching room temperature (ca. 1 hr), the solvent was removed in vacuo. Crystallization from carbon tetrachloride–dichloromethane–hexane gave 340 mg (82%) of 3,5-di-*p*-toluenesulfonyl-6-methoxy-8,8-dimethyl-1,3,5-triazabicyclo[4.2.0]octa-2,4-dione (**6**): mp >110° dec; ir (CCl_4) 2960, 1755, 1730, 1600, 1390 cm^{-1}; NMR (CCl_4) δ 1.63 (s, 6 H), 2.50 (s, 6 H), 2.75 and 3.13 (AB, J = 13.5 Hz, 2 H), 3.27 (s, 3 H), 7.40 (m, 4 H), 8.10 (m, 4 H). The mass spectrum showed only peaks from decomposition to *p*-toluenesulfonyl isocyanate and **3**, because **6** is thermally unstable and decomposes before vaporization.

Preparation of 1,4-Dipolar Adduct 10 from 3 and *p*-Toluenesulfonyl Isocyanate. To a solution of 426 mg (3.77 mmol) of **3** in 4 ml of dichloromethane at −40° was added a solution of 312 mg (1.58 mmol) of *p*-toluenesulfonyl isocyanate in 2 ml of dichloromethane. Upon warming the reaction mixture to ca. 0°, a white precipitate slowly came out of solution. Filtration or centrifugation at room temperature in an inert atmosphere followed by a dichloromethane rinse and drying in a stream of nitrogen resulted in 147 mg (30%) of an amorphous, white powder, **10**: mp 110–113° dec; ir (Nujol) 1690, 1645, 1420, 1270, 1240, 1140, 1080, 1065, 1015, 980, 905, 850, 825, 810, 790, 775, 705, 665 cm^{-1}; NMR (CH_2Cl_2) insoluble. The mass spectrum showed only peaks from decomposition to *p*-toluenesulfonyl isocyanate (*m/e* 197, 155, 91, 42) and **3** (*m/e* 113, 84, 56). The white powder **10** decomposes to a mixture of 37% of **6**, 8% of **11**, 27% of **12**, 12% of **13**, 12% of imino ether **3**, and some *p*-toluenesulfonyl isocyanate and/or *p*-toluenesulfonamide (due to traces of moisture) upon attempted solution in dichloromethane. The NMR integration of this decomposition mixture in dichloromethane solution indicates a 1:1 molar ratio of reactants **3** and *p*-toluenesulfonyl isocyanate in the adduct **10**. The infrared spectrum of the decomposed solution shows peaks at 2250 (*p*-toluenesulfonyl isocyanate), 1740 (broad band for 2:1 adduct **6**, amide **11**, and urea **13**), 1800, 1700 (amide **12**), and 1620 cm^{-1} (from **3**).

Reaction of 10 with *p*-Toluenesulfonyl Isocyanate. To 112 mg (0.36 mmol) of **10** in a round-bottom flask in a glove box was added 73 mg (0.37 mmol) of *p*-toluenesulfonyl isocyanate in 5 ml of dichloromethane. The heterogeneous mixture was kept at room temperature overnight. The solvent was removed in vacuo, leaving a solid. Crystallization from carbon tetrachloride–dichloromethane–hexane gave 176 mg (96%) of the 2:1 adduct **6**, mp 113° dec.

Reaction of 10 with 3. To 31 mg of **10** in an NMR tube was added ca. 100 mg of **3** in dichloromethane. After several hours at room temperature, the NMR spectrum showed only amides **12** and **13** in equal proportions along with excess **3**. The amides **12** and **13** were isolated from reactions containing excess **3** after standing at room temperature for several days. The solid **13** was separated from the liquid **12**. Distillation of the oil gave 1,4,4-trimethylazetidinone (**12**): bp 100° (2 mm); ir (CCl_4) 2960, 1750, 1280 cm^{-1}; NMR (CH_2Cl_2) δ 1.37 (s, 6 H), 2.65 (s, 5 H, NCH_3 and $-CH_2-$) [lit. NMR ($CDCl_3$) 1.35 (s, 6 H), 2.65 (s, 5 H)].[47a] Recrystallization of the crude crystals above gave 1-*N*-methyl-*N*-*p*-toluenesulfonylaminocarbonyl-4,4-dimethylazetidinone (**13**): mp 133–135° (recrystallized from carbon tetrachloride–dichloromethane–hexane); ir (CH_2Cl_2) 2950, 1800, 1705, 1275 cm^{-1}; NMR (CH_2Cl_2) δ 1.53 (s, 6 H), 2.40 (s, 3 H), 2.80 (s, 2 H), 3.15 (s, 3 H), 7.62 (AA'BB', 4 H); mass spectrum (90 eV) *m/e* 246.1377 ($M^+ - SO_2$, calcd for $C_{14}H_{18}N_2O_2$, 246.1368); *m/e* (rel intensity) 246 ($M^+ - SO_2$, 34), 191 (9), 155 (46), 139 (34), 121 (11), 119 (14), 117 (14), 97 (23), 92 (14), 91 (100), 84 (83), 83 (71), 70 (11), 65 (26), 58 (26), 57 (23), 56 (69), 55 (31), 51 (11), 44 (20), 43 (86), 42 (60), 41 (60), 39 (29).

Reaction of 10 with Water. To 142 mg (0.46 mmol) of **10** was added 73 mg (4 mmol) of water in 1 ml of acetonitrile. Evaporation of the solvent gave 146 mg (97%) of methyl 3-methyl-*N*-*p*-toluenesulfonylaminecarbonyl-3-aminobutanoate (**11**): mp 116–119° (from carbon tetrachloride–dichloromethane); ir (KBr) 3400 (br d), 3370 (sh), 2970, 2820, 1740, 1695, 1550, 1450, 1350 cm^{-1}; NMR (CCl_4) δ 1.37 (s, 6 H), 2.44 (s, 3 H), 2.65 (s, 2 H), 3.66 (s, 3 H), 7.23 (m, 2 H), 7.75 (m, 2 H), ca. 8.3 (NH); mass spectrum (70 eV) *m/e* 313.0857 ($M^+ - CH_3$, calcd for $C_{13}H_{17}N_2O_5S$, 313.0858); *m/e* (rel intensity) 328 (M^+, 0.17), 313 ($M^+ - CH_3$, 2), 297 (4), 264 (5), 256 (2), 255 (13), 232 (2), 217 (2), 215 (3), 198 (2), 197 (15), 171 (3), 157 (8), 156 (3), 155 (35), 117 (3), 116 (42), 115 (4), 114 (3), 108 (12), 107 (8), 92 (6), 91 (78), 84 (14), 83 (4), 74 (3), 73 (7), 65 (13), 63 (3), 59 (6), 58 (100), 57 (4), 56 (6), 55 (4), 51 (3), 43 (6), 42 (17), 41 (7), 39 (8).

Attempted Reaction of 10 with Phenyl Isocyanate. To 41 mg (0.13 mmol) of **10** was added 15 mg (0.31 mmol) of phenyl isocyanate in 1 ml of dichloromethane in an NMR tube. After 10 hr at room temperature, the NMR spectrum indicated only the formation of the 2:1 adduct **6** (ca. 70%) and amide **12** (ca. 25%) and no apparent reaction with phenyl isocyanate.

Reaction of 2:1 Cycloadduct 6 with Water. A solution composed of 40 mg (0.08 mmol) of **6**, 14 mg (0.8 mmol) of water, 0.5 ml of dichloromethane, and enough acetone to make it homogeneous was heated for ca. 10 min in a steam cone. The NMR spectrum showed complete decomposition. Removal of volatiles left a solid behind. The solid was identified as *p*-toluenesulfonamide by comparison of NMR and ir spectra of an authentic sample. The volatiles contained the azetine **3** by comparison of NMR and ir spectra. The only other decomposed material in the NMR and ir spectra was the urea **11** (ir 1740 cm^{-1} w). Upon standing in moist air, **6** decomposes to a 50:50 mixture of urea **11** and *p*-toluenesulfonamide as analyzed by NMR and ir.

Reaction of *p*-Toluenesulfonyl Isocyanate with 4,4-Dimethylazetidinone. To a solution of 190 mg (0.96 mmol) of *p*-toluenesulfonyl isocyanate in 0.5 ml of dichloromethane was added 98 mg (0.99 mmol) of 4,4-dimethylazetidinone in 0.5 ml of dichloromethane. The addition produced an exothermic reaction. NMR analysis showed clean conversion to **14** in 24 hr. Crystallization from carbon tetrachloride–dichloromethane–hexane gave 51 mg (18%) of *N*-*p*-toluenesulfonylaminocarbonyl-4,4-dimethylazetidinone (**14**): mp 138–140°; ir (CH_2Cl_2) 3250, 1775, 1730 cm^{-1}; NMR (CH_2Cl_2) δ 1.53 (s, 6 H), 2.42 (s, 3 H), 2.87 (s, 2 H), 7.40 (m, 2 H), 8.00 (m, 2 H), ca. 9.1 (broad s, 1 H, NH); mass spectrum (70 eV) *m/e* 232.1204 (calcd for $C_{13}H_{16}N_2O_2$, 232.1212) ($M^+ - SO_2$); *m/e* (rel intensity) 296 (M^+, 0.1), 232 (15), 171 (30), 155 (35), 109 (5), 108 (55), 107 (13), 106 (4), 92 (8), 91 (100), 90 (5), 89 (8), 84 (8), 83 (6), 65 (25), 56 (20), 42 (10), 41 (12), 39 (15).

Attempted Reaction between 3 and Phenyl Isocyanate. To 165 mg (1.38 mmol) of phenyl isocyanate was added 156 mg (1.38 mmol) of **3** in 0.5 ml of acetonitrile. The reaction tube was freeze degassed and sealed under vacuum (<0.1 mm). White crystals separated out within minutes (temperature ≤25°). The crystals were filtered, washed with acetonitrile, and dried in a vacuum desiccator. This resulted in 85 mg (ca. 50%) of trimer **15**: mp 268–271° (lit. mp 285°);[2] ir (CH_2Cl_2) 3050, 2250 (from CH_3CN), 1710, 1495, 1415 cm^{-1}; NMR (CH_2Cl_2) δ 1.97 (s, variable integration in different runs from 5.4 to 7.5 H from ca. 2 CH_3CN per mole of trimer) and 7.50 (m, 15 H); mass spectrum (70 eV) *m/e* 357.1126 (calcd for $C_{21}H_{15}N_3O_3$, 357.1113); *m/e* (rel intensity) 357 (M^+, 36), 305 (1), 282 (0.5), 238 (3), 218 (1), 212 (1), 167 (1), 149 (2), 145 (3), 120 (8), 119 (100), 93 (6), 91 (15), 77 (5), 64 (7), 57 (5), 55 (5), 43 (12), 41 (13). The white crystals are composed of the trimer of phenyl isocyanate, **15**, complexed with ca. two acetonitrile molecules.[49,50]

Reaction of Phenyl Isocyanate with Base. To 210 mg (1.76 mmol) of phenyl isocyanate was added 183 mg of acetonitrile. After 4 days with no apparent change, several drops of methyldiethylamine were added. After 1 day at 25°, 200 mg (ca. 95%) of white crystals was isolated. Recrystallization from dichloromethane afforded the trimer of phenyl isocyanate, **15**, mp 271.5–272.5°. The NMR still showed a trace of acetonitrile present (δ 1.97).

Reaction of Chlorosulfonyl Isocyanate with 1. A solution of 290 mg (2.52 mmol) of **1** in 0.5 ml of dichloromethane was added to a solution of 400 mg (2.82 mmol) of chlorosulfonyl isocyanate in 0.5 ml of dichloromethane at −70°. The solution was allowed to come to room temperature, then poured into water. The organic layer was separated, washed with aqueous sodium sulfite–sodium hydroxide solution, and dried over potassium carbonate. Removal of solvent and all volatile material left 6 mg (1.4%) of a solid residue characterized as *N,N'*-di-*tert*-butylurea: mp 177–184° (sublimes) (lit. mp 245°);[51] ir (CH_2Cl_2) 3400, 2950, 1680 cm^{-1}; NMR (CH_2Cl_2) δ 1.27 (s), NH not seen; mass spectrum (70 eV) *m/e* 172.1582 (calcd for $C_9H_{20}N_2O$, 172.1575); *m/e* (rel intensity) 172 (M^+, 4), 157.1346 ($M^+ - CH_3$, 7), 84 (7), 61 (7), 58 (100), 57 (13), 56 (9), 44 (16), 41 (20).

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Registry No.—**1**, 49680-36-6; **2**, 55222-40-7; **3**, 23974-38-1; **4**, 49680-46-8; **5**, 52856-04-9; **6**, 55254-58-5; **7**, 55222-41-8; **8**, 55222-42-9; **10**, 55222-43-0; **11**, 55222-44-1; **12**, 23974-51-8; **13**, 55254-60-9; **14**, 55254-59-6; **15**, 1785-02-0; *p*-toluenesulfonyl isocyanate 4083-64-1; phenyl isocyanate, 103-71-9; 4,4-dimethylazetidinone, 4879-95-2; acetonitrile, 75-05-8; chlorosulfonyl isocyanate, 1189-71-5; *N,N'*-di-*tert*-butylurea, 5336-24-3.

References and Notes

(1) For previous work in this area see D. H. Aue and D. Thomas, *J. Org. Chem.*, **39**, 3855 (1974); **40**, 2360 (1975).
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