mp 83–84 °C; ν_{max} ($CHCl_3$) 1815, 1740, 1250–1200 cm^{-1}; λ_{max} (C_2H_5OH) 214 nm (ϵ 9820), 325 (70); ^1H NMR δ 1.7 (br m, 20 H, ring H), 2.05 (s, 3 H, $OCOCH_3$), 2.10 (s, 3 H, $OCOCH_3'$); ^{19}F NMR ($CDCl_3$) −4553 Hz.

Anal. Calcd for $C_{23}H_{26}N_2O_9F_6$: C, 46.90; H, 4.42; N, 4.75; F, 19.35. Found: C, 46.99; H, 4.45; N, 4.75; F, 19.38.

2,2-Bis(trifluoromethyl)-5-[2-acetoxy-2-adamantyl)carbonyl]-1,3,4-dioxazole (27b). From 1.32 g of **25b**[7] and 16 g of hexafluoroacetone there was obtained 1.97 g of a liquid. Bulb-to-bulb distillation afforded 1.28 g [60%, 79% on the basis of **25b** consumed (vide infra)] of **27b**: bp 90–100 °C (0.1 mm); ν_{max} (liquid film) 1740, 1620, 1250–1200 cm^{-1}; ^1H NMR ($CDCl_3$) δ 1.6–2.8 (br m, 13 h, adamantyl H), 2.18 (s, 3 H, $OCOCH_3$); ^{19}F NMR (94.1 MHz, $CDCl_3$) −84885 Hz.

2,2-Bis(trifluoromethyl)-5[[(2-acetoxy-2-adamantyl)carbonyl][[[(2-acetoxy-2-adamantyl)carbonyl]oxy]imino]methyl]-1,3,4-dioxazole (26b). The residue from the distillation described in the isolation of **27b** above was chromatographed on silica gel with $CHCl_3$. In addition to 0.32 g of starting furoxan **25b**, there was obtained 0.165 g (9.5%, 13% on the basis of **25b** consumed) of **26b**: mp 184.5–186 °C (from pentane); ν_{max} (KBr) 1835, 1740, 1665, 1610, 1250–1200 cm^{-1}; λ_{max} (C_2H_5OH) 213 nm (ϵ 10 000), 335 (83); ^1H NMR ($CDCl_3$) δ 1.8–2.65 (br m, 28, adamantane ring H), 2.06 (s, 3 H, $OCOCH_3$), 2.13 (s, 3 H, $OCOCH_3'$); ^{19}F NMR ($CDCl_3$) −4500 Hz (s, CF_3).

Anal. Calcd for $C_{31}H_{34}N_2O_9F_6$: C, 53.85; H, 4.96; N, 4.05; F, 16.50. Found: C, 53.74; H, 4.94; N, 3.99; F, 16.51.

3-[[(1-Acetoxycyclohexyl)carbonyl][[[(1-acetoxycyclohexyl)carbonyl]oxy]imino]methyl]-4,5-bis(trifluoromethyl)isoxazole (28). From 4.22 g of **25a** and 5.0 g of hexafluoro-2-butyne there was obtained from fractions 4–8 of silica gel/$CHCl_3$ chromatography 2.63 g (45%) of **28**: mp 89.5–90 °C (from pentane); ν_{max} ($CHCl_3$) 1800, 1740, 1625 cm^{-1}; ν_{max} (C_2H_5OH) 220 nm (sh, ϵ 7000), 327 (55); ^1H NMR ($CDCl_3$) δ 1.4–1.8 (br m, 20 H, cyclohexane ring H), 1.94 (s, 3 H, $OCOCH_3$), 2.16 (s, 3 H, $OCOCH_3$); ^{19}F NMR ($CHCl_3$) −3230 (q, J = 6 Hz, 3 H, CF_3), −3560 Hz (q, J = 6 Hz, 3 H, CF_3).

Anal. Calcd for $C_{24}H_{26}N_2O_8F_6$: C, 49.30; H, 4.46; N, 4.79. Found: C, 49.42; H, 4.51; N, 4.70.

Registry No. 1, 75768-35-3; 3, 75768-36-4; 5, 75768-37-5; 7, 75768-38-6; 8, 75768-39-7; 10, 75768-40-0; 12, 75768-41-1; 13, 6635-54-7; 15, 75768-42-2; 21, 75768-42-3; 22, 21443-49-2; 23, 75768-44-4; 24, 75768-45-5; 25a, 75768-46-6; 25b, 75768-47-7; 26a, 75768-48-8; 26b, 75768-49-9; 27a, 75768-50-2; 27b, 75768-51-3; 28, 75768-52-4; hexafluoro-2-butyne, 692-50-2; hexafluoroacetone, 684-16-2; methyl propiolate, 922-67-8; dimethyl acetylenedicarboxylate, 762-42-5; 4-phenyl-3-butyn-2-one, 1817-57-8; phenylacetylene, 536-74-3.

Reaction of Some 1-(*p*-Tolylsulfonyl)-2,3,3-trialkyldiaziridines with Aryl Isocyanates and Benzoyl Isocyanate

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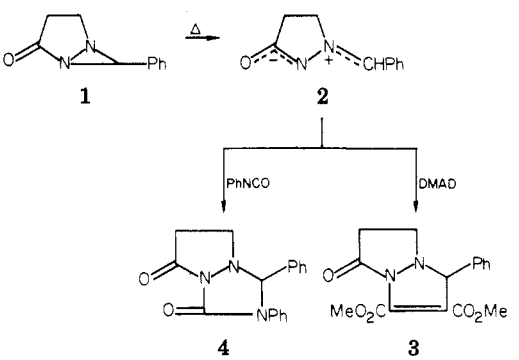
1-(*p*-Tolylsulfonyl)-2-alkyl-3,3-pentamethylenediaziridines react with aryl isocyanates and benzoyl isocyanate to form 1-alkyl-2-(*p*-toluenesulfonyl)-4-aryl-5,5-pentamethylene-1,2,4-triazolidin-3-ones. The structure of one of the latter compounds was established by a single-crystal X-ray diffraction study. 1-(*p*-Tolylsulfonyl)-2,3-dialkyldiaziridines fail to react with isocyanates.

Diaziridines substituted in the 1- or 1,2-positions with electron-withdrawing groups are prone to form azomethine imines which undergo reactions typical of 1,3-dipoles. For example, heating **1** neat affords a 78% yield of **2**.[1,2] Compound **2** reacts with dimethyl acetylenedicarboxylate (DMAD) and phenyl isocyanate to give **3** and **4**, respectively[3] (Scheme I).

In many cases the formation of the azomethine imine from the diaziridine must be inferred by trapping experiments with 1,3-dipolarophiles[4] or nitrones,[5] isomerizations to ring-expanded products,[6-8] interaction with neighboring groups,[9,10] and rearrangements to open-chain isomeric hydrazones.[9,11,12]

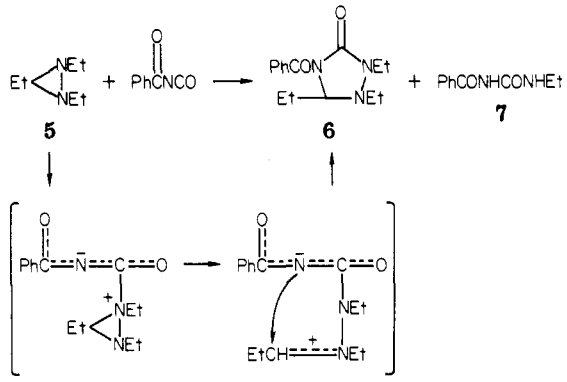
Scheme I



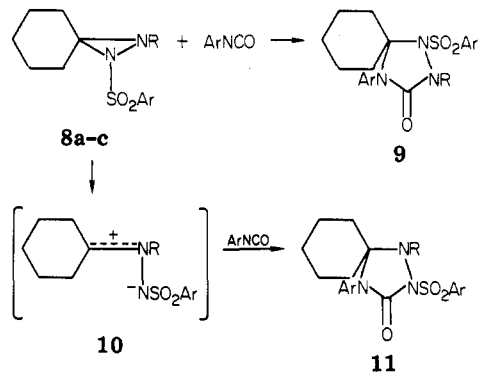
(1) Schulz, M.; West, G. *J. Prakt. Chem.* **1970**, *312*, 161.
(2) It is interesting to note that azomethine imines upon irradiation often isomerize to diaziridines. For example see: (a) Pleiss, M. G.; Moore, J. A. *J. Am. Chem. Soc.* **1968**, *90*, 4738; (b) Schulz, M.; West, G. *J. Prakt. Chem.* **1973**, *315*, 711; (c) Maki, Y.; Kawamura, M.; Okamoto, H.; Suzuki, M.; Kaji, K. *Chem. Lett.* **1977**, 1005.
(3) Dorn, H.; Otto, A. *Chem. Ber.* **1968**, *101*, 3287.
(4) Heine, H. W.; Henrie, R., II; Heitz, L.; Kovvali, S. R. *J. Org. Chem.* **1974**, *39*, 3187.
(5) Heine, H. W.; Heitz, L. *J. Org. Chem.* **1974**, *39*, 3192.
(6) Nabeya, A.; Tamura, Y.; Kodama, T.; Iwakura, Y. *J. Org. Chem.* **1973**, *38*, 3758.
(7) Nabeya, A.; Saito, J.; Koyama, H. *J. Org. Chem.* **1979**, *44*, 3935.

0022-3263/81/1946-0320$01.00/0

Scheme II



Scheme III

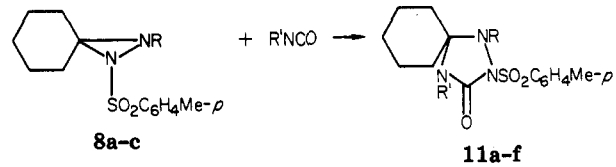


Recently Komatsu and colleagues reacted diaziridines containing no electron-withdrawing groups on the nitrogens of the ring such as 1,2,3-triethyldiaziridine (**5**) with phenyl isocyanate and benzoyl isocyanate.[13] The reaction of phenyl isocyanate with **5** gave a host of compounds—all in low yields, e.g., PhNHCONHEt (6%), PhNHCON(Et)COEt (2%), EtNHCON(Ph)CONHPh (3%), and EtCHN(Et)CON(Ph)CH_2NCONHPh (11%). Heating **5**, however, with benzoyl isocyanate afforded the 1,2,4-triazolidin-3-one (**6**) and *N*-ethyl-*N'*-benzoylurea (**7**) in 44% and 39% yields, respectively (Scheme II). A mechanism suggested for the formation of **6** involved a nucleophilic attack of the diaziridinyl nitrogen on the cumulative carbonyl carbon of the isocyanate followed by ring opening of the diaziridine ring and then ring closure to **6** (Scheme II).

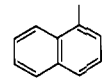
We present here a study of the reaction of some 1-(*p*-toluenesulfonyl)-2-alkyl-3,3-pentamethylenediaziridines (**8a–c**) with some aryl isocyanates and benzoyl isocyanate. It would be anticipated that if the *N*-alkyl nitrogen of **8a–c** attacked the carbonyl carbon of the isocyanate (as was the case with **5** and benzoyl isocyanate), the products would be 1-(*p*-toluenesulfonyl)-2-alkyl-4-aryl-5,5-pentamethylene-1,2,4-triazolidin-3-ones (**9**, Scheme III). If, on the other had, **8a–c** initially formed an azomethine imine (**10**) which underwent cycloaddition with the isocyanate, then the products would be **11** in which the NR and the NSO_2Ar moieties are interchanged relative to **9** (Scheme III). Diaziridines **8a–c** reacted with phenyl isocyanate or α-naphthyl isocyanate at 90 °C or with benzoyl isocyanate to give adducts that corresponded to **11** (Table I). That indeed **11** was formed rather than **9** was shown by a single-crystal, X-ray diffraction study on the adduct **11d** obtained in 79% yield from the reaction of α-naphthyl isocyanate with 1-(*p*-toluenesulfonyl)-2-ethyl-3,3-pentamethylenediaziridine. Figure 1 is a perspective drawing[14] of one of the two independent molecules of **11d** found in the crystal structure analysis. The geometry of the core of the two molecules is approximately equivalent with the triazolidinone rings having close to half-chair conformations: $\Delta C_2(3) = 6.6°$ and $\Delta C_2(3') = 2.4°$.[15] The cyclohexane rings in both molecules have standard chair conformations.

In contrast to **8a–c**, 1-(*p*-toluenesulfonyl)-2-cyclohexyl-3-methyldiaziridine failed to react when heated with phenyl isocyanate at 90 °C. When mixtures of benzoyl isocyanate and 1-(*p*-toluenesulfonyl)-2-cyclohexyl-3-ethyldiaziridine (**8d**) were heated in benzene for 3 h, no adduct was formed, but, instead, an invertomer (**8e**) was

Table I. 1-Alkyl-2-(*p*-toluenesulfonyl)-4-aryl-5,5-pentamethylene-1,2,4-triazolidin-3-ones 11a–f from Reaction of 8a–c with Isocyanates



8a–c **11a–f**

compd[a]	R	R'	yield,[b] %	mp, °C
11a	Me	Ph	68	158–159
11b	Et	Ph	85	154–155
11c	*i*-Pr	Ph	43	152–155
11d	Et		79	156–157
11e	Me	PhCO	82	136–138
11f	Et	PhCO	82	127–128

[a] All products exhibited satisfactory combustion analytical data. [b] Yields are based on recrystallized products.

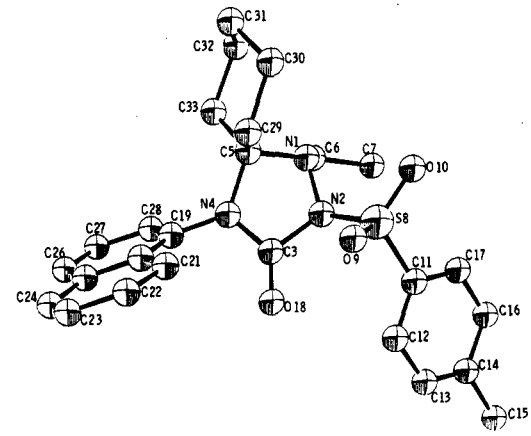


Figure 1. Perspective drawing of **11d** generated from the crystal coordinates.

(8) Heine, H. W.; Baclawski, L. M.; Bonser, S. M.; Wachob, G. *J. Org. Chem.* **1976**, *41*, 3229.
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(11) Schmitz, E.; Habisch, D.; Gründemann, C. *Chem. Ber.* **1967**, *100*, 142.
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isolated in 47% yield along with some benzamide and a high-melting (220 °C) substance which was presumably *N,N'*-dibenzoylurea. Compound **8e** was also obtained by simply heating **8d** in benzene. Mass spectroscopy revealed that **8d** and **8e** gave identical fragmentation patterns. The mass spectra of **8d** and **8e** exhibited a peak of low intensity for the molecular ion at *m/e* 308 and an intense peak at *m/e* 153 representing the loss of the *p*-toluenesulfonyl moiety. A strong peak at *m/e* 157 was identified by high-resolution mass spectroscopy to have the formula p-$MeC_6H_4SO_2H_2$. Compound **8d** melted at 79–80 °C, and **8e** melted at 75–77 °C. A mixture of **8d** and **8e** melted at 69–71 °C. The two substances had similar but different infrared spectra and ^1H NMR spectra. The methine protons of **8d** and **8e** appeared as multiplets centered at δ 3.17 and 3.57, respectively. A nuclear magnetic spectroscopic study show that pure **8d** and pure **8e** in benzene at 80 °C formed an equilibrium mixture of the invertomers within 5 min in a ratio of approximately 3:1 in favor of **8e**. The same ratio was maintained when the reaction mixture was cooled to ambient temperature. Evaporation of the solvent formed an oil which when triturated with cold methanol selectively precipitated **8e**.

The separation of invertomers of diaziridines was first achieved by Mannschreck and Seitz,[16] who isolated and characterized isomeric 1,3-dimethyl-2,3-dibenzyldiaziridines and isomeric 1,3-dimethyl-3-benzyldiaziridines. In the latter case one of the invertomers selectively crystallized from a mixture of the two forms in pentane. Nabeya and her colleagues[17] found that in the synthesis of 1-cyclohexyl-2-phenyldiaziridine and 1-benzyl-2-phenyldiaziridine invertomers were formed that were isolable by chromatography. The higher melting isomers in each instance could be transformed into their invertomeric counterparts by heating.

The failure of 1-(*p*-toluenesulfonyl)-2-cyclohexyl-3-methyldiaziridine to react with phenyl isocyanate and of **8d** to react with benzoyl isocyanate is consistent with observations that the formation of azomethine imines from diaziridines usually requires two stabilizing substituents at the diaziridinyl carbon as well as two electron-withdrawing groups at the 1- or 1,2-positions. Thus, while 1-aroyl-2,3,3-trialkyldiaziridines,[8] 1-(arylcarbamoyl)-2,3-dialkyl-3-aryldiaziridines,[6,7] and 1,1-dialkyl-1*H*-diazirino-[1,2-*b*]phthalazine-3,8-diones[4] isomerize via an azomethine imine, the same diaziridinyl systems with but one substituent on the diaziridinyl carbon do not isomerize.

Experimental Section

2-Methyl-, 2-Ethyl-, and 2-Isopropyl-1-(*p*-toluenesulfonyl)-3,3-pentamethylenediaziridines (8a–c). To a stirred mixture of 7 mmol of 1-methyl-, 1-ethyl-, or 1-isopropyl-3,3-pentamethylenediaziridine[18a] and 1.01 g (10 mmol) of triethylamine cooled in a rock salt/ice bath was added 1.37 g (7 mmol) of pulverized *p*-toluenesulfonyl chloride over a period of 30 min. After being stirred for 3 h, the mixture was washed with 200 mL of 5% $NaHCO_3$ solution, and the crude **8a–c** was filtered and washed with water. Compounds **8a–c** were recrystallized from commercial absolute ethanol. In this manner were obtained the following: **8a** (57%), mp 83–85 °C; **8b** (71%), mp 85–87 °C; **8c** (52%), mp 107–108 °C.

Anal. Calcd for $C_{14}H_{20}N_2O_2S$ (**8a**): C, 59.95; H, 7.19; N, 10.00. Found: C, 59.81; H, 7.27; N, 9.86. Calcd for $C_{15}H_{22}N_2O_2S$ (**8b**): C, 61.19; H, 7.53; N, 9.52. Found: C, 61.66; H, 7.87; N, 9.62. Calcd for $C_{16}H_{24}N_2O_2S$ (**8c**): C, 62.30; H, 7.84; N, 9.08. Found: C, 62.19; H, 7.79; N, 9.08.

2-Cyclohexyl-1-(*p*-toluenesulfonyl)-3-ethyldiaziridine (8d). This compound was prepared as described by Schmitz[19] and melted at 79–80 °C (lit.[19] 82–83 °C): IR (Nujol) 1590, 1325, 1150–1160, 1080, 970, 910, 893, 875, 820, 735, 695–680 cm^{-1}; ^1H NMR ($CDCl_3$) δ 7.92 (d, 2 H, J = 8 Hz), 7.38 (d, 2 H, J = 8 Hz), 3.17 (m, 1 H), 2.45 (s, 3 H), 2.3 (d, 1 H, J = 4 Hz), 2.02 (d, 1 H, J = 4 Hz) 1.65–1.0 (m, 14 H); mass spectrum, *m/e* 308 (molecular ion).

Anal. Calcd for $C_{16}H_{24}N_2O_2S$: C, 62.32; H, 7.84; N, 9.08. Found: C, 62.52; H, 8.01; N, 9.19.

Conversion of 8d into 8e. A solution of 991 mg of **8d** and 10 mL of C_6H_6 was refluxed for 3 h. The solvent was evaporated, and the oily residue was triturated with 0.5 mL of cold MeOH. The crude **8e** (630 mg) was filtered and recrystallized three times from methanol to give **8e** melting at 75–77 °C: IR (Nujol) 1590, 1335, 1158, 1080, 975, 897, 870, 838, 815, 758, 735–730 cm^{-1}; ^1H NMR ($CDCl_3$) δ 7.90 (d, 2 H, J = 8 Hz), 7.40 (d, 2 H, J = 8 Hz), 3.57 (m, 1 H), 2.48 (s, 3 H), 2.3–0.80 (m, 16 H); mass spectrum, *m/e* 308.

Preparation of 11a–c. In a predried, 10-mL flask equipped with a magnetic stirring bar was placed 1 mmol of **8a**, **8b**, or **8c**. The flask was stoppered with a rubber septum, and nitrogen was introduced. A 3-mL sample of phenyl isocyanate was injected through the septum and the reaction flask placed in an oil bath held at 90 °C. Reaction times were 20 min, 3 h, and 2 h for **8a–c**, respectively. The reaction mixture was cooled, 5 mL of anhydrous ether was added, and the crude compounds **11a–c** were filtered. The crude **11a,b** were recrystallized from commercial absolute ethanol. The crude **11c** was contaminated with diphenylurea. It was slurried in a small amount of chloroform, and the diphenylurea was filtered. Evaporation of the chloroform filtrate gave **11c** which was recrystallized from commercial absolute ethanol.

Anal. Calcd for $C_{21}H_{25}N_3O_3S$ (**11a**): C, 63.12; H, 6.30; N, 10.51. Found: C, 63.32; H, 6.52; N, 10.43. Calcd for $C_{22}H_{27}N_3O_3S$ (**11b**): C, 63.90; H, 6.58; N, 10.18. Found: C, 63.72; H, 6.56; N, 10.13. Calcd for $C_{23}H_{29}N_3O_3S$ (**11c**): C, 64.61; H, 6.84; N, 9.83. Found: C, 64.65; H, 6.74; N, 9.89.

Preparation of 11d. By use of the same procedure employed for making **11a–c**, a mixture of 735 mg (2.5 mmol) of **8b** and 2 mL of 1-naphthyl isocyanate was heated at 60 °C for 3 h. Crude **11d** precipitated as the reaction mixture cooled. Anhydrous ether (5 mL) was added, and the crude **11d** (mp 147–150 °C, 1.0 g, 86%) was filtered and recrystallized from commercial absolute ethanol to give 920 mg (79%) of pure **11d**.

Anal. Calcd for $C_{26}H_{29}N_3O_3S$: C, 67.36; H, 6.30; N, 9.06. Found: C, 67.85; H, 6.52; N, 9.24.

Preparation of 11e and 11f. Into a dried, round-bottomed flask equipped with a magnetic stirring bar were charged 1.12 g (4 mmol) of **8a** and 40 mL of anhydrous benzene. Nitrogen was introduced and a rubber septum fitted on the flask. Benzoyl isocyanate (600 mg, 4 mmol) was added via a syringe. The reaction flask was placed in an oil bath held at 55 °C, and the mixture was stirred for 2 h. Evaporation of the solvent afforded a yellow oil which when slurried with a small quantity of 95% EtOH caused precipitation of crude **11e**. The **11e** was filtered. Purification was effected by dissolving **11e** in hot anhydrous MeCN and then immersing the flask containing the **11e** immediately in an ice bath.

Anal. Calcd for $C_{22}H_{25}N_3O_4S$: C, 61.80; H, 5.90; N, 9.83. Found: C, 61.85; H, 5.91; N, 9.98.

Compound **11f** was formed in an analogous fashion as **11e** by reacting 294 mg (1 mmol) of **8b**, 10 mL of anhydrous benzene, and 125 mg (1 mmol) of benzoyl isocyanate for 2 h (bath temperature 70 °C). The reaction mixture was cooled, and 10 mL of anhydrous ether was added. The mixture was filtered and the filtrate evaporated. The residual solid was slurried with 95% EtOH and filtered to give crude **11f**: 389 mg (88%); mp 120–124 °C. The same method of purification as described for **11e** was followed.

Anal. Calcd for $C_{23}H_{27}N_3O_4S$: C, 62.56; H, 6.39; N, 9.52. Found: C, 62.90; H, 6.19; N, 9.32.

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(18) (a) Schmitz, E.; Ohme, R.; Schmidt, R.-D. *Chem. Ber.* **1962**, *95*, 2714. (b) Schmitz, E.; Ohme, R. *Chem. Ber.* **1962**, *95*, 795.
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Crystal Structure of 11d. Crystals of **11d** formed from 100% ethanol in the space group $P\bar{1}$ with a = 14.022 (4) Å, b = 15.719 (4) Å, c = 12.943 (3) Å, α = 97.90 (2)°, β = 115.39 (2)°, and γ = 104.91 (2)°. With Z = 4 the calculated density is 1.29 g/cm^3. Of the 6334 diffraction maxima measured with $2\theta \leq 114°$ by using Cu Kα radiation, 2996 (47%) were observed ($I \geq 3\sigma(I)$). Standard direct-method techniques[20] coupled with tangent-formula recycling[21] generated suitable starting positions. Full-matrix, least-squares refinements[22] minimizing $\sum w(|F_o| - |F_c|)^2$ with $w = (1/\sigma F_o)^2$ produced an unweighted R factor of 0.107. Because of the poor quality of the data, no attempt was made to find positions for the hydrogen atoms. The crystal lattice has two independent molecules; one is well-behaved while the naphthyl group in the second is disordered. This disordered region accounts for the small percentage of observed reflections. Bond distances and angles within this disordered naphthyl group show significant deviations from the naphthyl group on the first molecule. Table II (supplementary material) contains the final fractional coordinates and temperature parameters for **11d**.

Acknowledgment. We thank Dr. Jack Smith of Merck Sharp and Dohme Research Laboratories for obtaining the high-resolution mass spectra of compounds **8d** and **8e** and The Camille and Henry Dreyfus Foundation for financial support

Registry No. 8a, 75802-07-2; **8b**, 75802-08-3; **8c**, 75802-09-4; **8d**, 75802-10-7; **11a**, 75802-11-8; **11b**, 75802-12-9; **11c**, 75802-13-0; **11d**, 75802-14-1; **11e**, 75802-15-2; **11f**, 75802-16-3; 1-methyl-3,3-pentamethylenediaziridine, 26177-34-4; 1-ethyl-3,3-pentamethylenediaziridine, 52551-68-5; 1-isopropyl-3,3-pentamethylenediaziridine, 75802-17-4; phenyl isocyanate, 103-71-9; 1-naphthyl isocyanate, 86-84-0; benzoyl isocyanate, 4461-33-0.

Supplementary Material Available: Table II containing the final fractional coordinates and temperature parameters (2 pages). Ordering information is given on any current masthead page.

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Synthesis of Hexahydro-3,3,5,5,7-pentaalkyl-2*H*-1,4-diazepin-2-ones from 1,3-Diamines and Ketones

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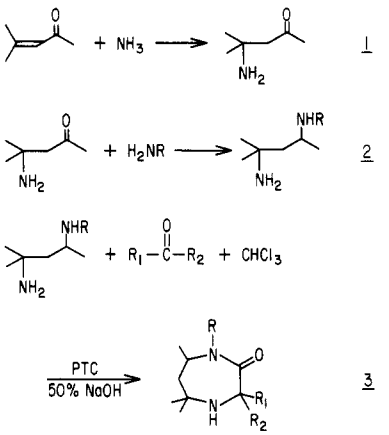
A new method for preparing mono- and bis(hexahydro-3,3,5,5,7-pentaalkyl-2*H*-1,4-diazepin-2-ones) is reported (Schemes I and II). The key step of this synthesis is the final one in which a 1,3-diamine is reacted with a ketone in the presence of sodium hydroxide, chloroform, and a phase-transfer catalyst (PTC). Bis(hexahydro-3,3,5,5,7-pentaalkyl-2*H*-1,4-diazepin-2-ones) are isolated as a mixture of diastereomers. Of these bis compounds, diastereomers of 1,1'-(1,2-ethanediyl)bis(hexahydro-3,3,5,5,7-pentamethyl-2*H*-1,4-diazepin-2-one) (**5a**) can be readily separated by a fractional recrystallization, or their diastereomeric distributions can be measured by ^{13}C NMR.

Recently we disclosed the facile preparation of hindered piperazinones.[2] The UV stabilization properties of these piperazinones and hexahydro-1,4-diazepin-2-ones which we describe in this paper are excellent in various plastics, especially in polypropylene. The synthesis of mono- and bis(hexahydro-3,3,5,5,7-pentaalkyl-2*H*-1,4-diazepin-2-ones) is presented in this paper. Emphasis is placed on the final step of the three-step synthesis as shown in Schemes I and II.

Results and Discussion

The synthesis of hexahydro-3,3,5,5,7-pentaalkyl-2*H*-1,4-diazepin-2-ones (**3**) begins with diacetonamine **1** obtained from mesityl oxide and concentrated ammonium hydroxide. This is followed by the reductive amination of **1** with an appropriate amine to give a 1,3-alkanediamine (**2**). Finally, the reaction of **2** with a ketone and trichloromethyl anion yields the desired hexahydro-3,3,5,5,7-pentaalkyl-2*H*-1,4-diazepin-2-ones (**3**). This three-step reaction is summarized in Scheme I.

Scheme I



The first step was carried out by a modified literature method.[3] When mesityl oxide and concentrated ammonium hydroxide are simply heated to 45–50 °C, the reaction time can be shortened from 3 days to several hours.

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