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LETTERS TO THE EDITOR

Reaction of *N*,*N*'-Bis(trimethylsilyl)-*N*-methyl-*N*'-arylureas with Chloro(chloromethyl)dimethylsilane

M. A. Pudovik, L. K. Kibardina, and A. N. Pudovik

Arbuzov Institute of Organic and Physical Chemistry, Kazan Research Center,
Russian Academy of Sciences, Kazan, Tatarstan, Russia

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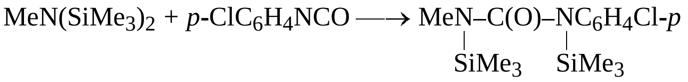
We previously showed that the reaction of chloro(chloromethyl)dimethylsilane with bis(*N*-methyl-*N*-trimethylsilylamido)methylphosphonate is accompanied by separation of 2 equiv of chlorotrimethylsilane and provides 1,2,3,4,4-pentamethyl-1,3-diaza-2-phospha-4-silolidin-2-one [1]. With the purpose of investigate the possibility of formation of new silacyclanes on the basis of chloromethyl derivatives of silicon we turned to reaction of *N*,*N*'-bis(trimethylsilyl)-*N*-methyl-*N*'-phenylurea (**Ia**) and *N*,*N*'-bis(trimethylsilyl)-*N*-methyl-*N*'-*p*-chlorophenylurea (**Ib**) with chloro(chloromethyl)dimethylsilane. Silylated urea **Ib** was prepared from heptamethyldisilazane and *p*-chlorophenyl isocyanate.

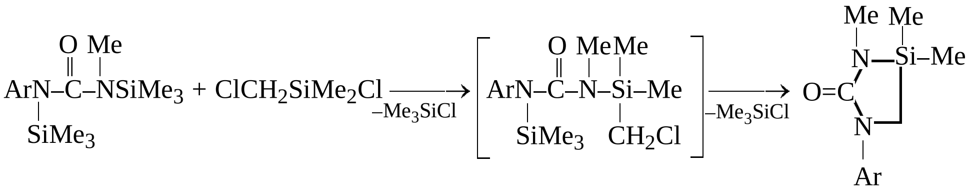


$$\mathrm{MeN(SiMe_3)_2 + \textit{p}\text{-}ClC_6H_4NCO \longrightarrow \underset{\displaystyle SiMe_3}{MeN}\text{–}C(O)\text{–}\underset{\displaystyle SiMe_3}{N}C_6H_4Cl\text{-}\textit{p}}$$

Ib

The ^{1}H NMR spectrum of compound **Ib** reveals nonequivalence of trimethylsilyl protons. They are presented by two singlets at δ 0.71 and 0.53 ppm. Methyl protons give a singlet at δ 2.73 ppm.

The reactions of ureas **Ia** and **Ib** with chloro(chloromethyl)dimethylsilane(II) involves initial *trans*silylation with separation of a molecule of chlorotrimethylsilane and intermediate formation of urea **III**. The latter undergoes cyclization involving the chloromethyl group and resulting in separation of a further molecule of trimethylchlorosilane and formation of silacyclanes **IVa** and **IVb**.



Ia, Ib **II** **III** **IVa, IVb**

The IR spectra of compounds **IVa** and **IVb** contain strong carbonyl absorption band at 1668 cm^{-1}. In the ^{1}H NMR spectrum of compound **IVa**, there are three singlets characteristic of protons of the methyl group on silicon (δ 0.40 ppm), the methyl group on nitrogen (δ 2.78 ppm), and the methylene group (δ 2.95) are observed. The intensity ratio of these three signals is 6 : 3 : 2. In addition, a multiplet of phenyl

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protons is observed at δ 7.06–7.37 ppm. Similar compounds were previously obtained by decomposition of ten-membered Si,N-containing heterocycles [2].

***N*,*N*'-Bis(trimethylsilyl)-*N*-*p*-chlorophenyl-*N*-methylurea (Ib).** *p*-Chlorophenyl isocyanate, 3.07 g, and 3.50 g of heptamethyldisilazane were dissolved in 40 ml of anhydrous ether. The reaction involved heat evolution. After 12 h, the solvent was removed to obtain 5.1 g (78%) of carbamide **I**, mp 88°C. Found, %: N 8.29; Si 8.33. $C_{14}H_{22}ClN_2OSi$. Calculated, %: N 8.51; Si 8.51.

3,4,4-Trimethyl-1-phenyl-1,3-diaza-4-silolidin-2-one (IVa). A solution of 3.0 g of urea **Ia** and 1.4 g of chlorosilane **II** in 40 ml of anhydrous ether was refluxed for 2 h. Distillation of the reaction mixture gave 1.3 g (59%) of silacyclane **IVa**, bp 145°C (0.08 mm Hg). Found, %: N 12.69; Si 12.86. $C_{11}H_{16}\cdot N_2OSi$. Calculated, %: N 12.73; Si 12.73.

1-(*p*-Chlorophenyl)-3,4,4-trimethyl-1,3-diaza-4-silolidin-2-one (IVb). A solution of 3.3 g of urea **Ib** and 1.4 g of chlorosilane **II** in 40 ml of anhydrous ether was refluxed for 1 h. Vacuum distillation of the reaction mixture gave 0.8 g (32%) of silacyclane **IVb**, bp 136–140°C (0.08 mm Hg), mp 60–62°C. Found, %: C 52.19; H 5.76; N 10.15; Si 10.17. $C_{11}H_{15}ClN_2OSi$. Calculated, %: C 52.90; H 5.88; N 10.19; Si 10.98.

The IR spectra were recorded on a UR-20 spectrometer in the range 500-3600 cm^{-1} for suspensions in mineral oil. The ^1H NMR spectra were measured on a Bruker WM-250c spectrometer (250.13 MHz) against external TMS.

ACKNOWLEDGMENTS

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