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SYNTHESIS AND ANTIMICROBIAL PROPERTIES OF N-ORGANOSILYLMETHYLENELACTAMS

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UDC 615.281:547.466.3].012.1

Lactam derivatives, and particularly silylated lactams, have recently attracted the attention of chemists and pharmacologists as valuable physiologically active compounds. They mediate central nervous system inhibition, and they possess anticonvulsive, acaricidal, and psychoactivating properties [1, 2].

The object of this investigation was to synthesize organosilicon lactams with triorganosilylmethylene groups on the nitrogen atom, and to study their physicochemical and bactericidal properties.

The synthesis of triorganosilylmethylenepyrrolidines and triorgansilylmethylenecaprolactams was accomplished by reaction of 2-pyrrolidone or ε-caprolactam with the appropriate chloromethyltriorganosilane at 120-150°C in the presence of anhydrous Na_2CO_3 or K_2CO_3, with a molar ratio of reactants of 1:1, for 4-6 h either in toluene or in the absence of a solvent. The reaction proceeds as follows:

$$\overline{\text{CO}-(\text{CH}_2)_n-\text{NH}}| + \text{ClCH}_2\text{SiR}_2\text{R}' \longrightarrow \overline{\text{CO}-(\text{CH}_2)_n-\text{N}}|\text{CH}_2\text{SiR}_2\text{R}',$$

where $n = 3$; $R = CH_3$; $R' = C_2H_5$ (I), C_4H_9 (II), C_6H_5 (III);

$n = 5$; $R = CH_3$; $R' = C_2H_5$ (IV), C_3H_7 (V), C_4H_9 (VI), C_6H_5 (VII).

We have also carried out the reaction between 2-pyrrolidone or ε-caprolactam and 1,3-bis(chloromethyl)-1,3-tetramethyldisiloxane with a molar ratio of starting materials of 2:1 at 150°C in the presence of K_2CO_3 as follows:

$$\overline{\text{CO}-(\text{CH}_2)_n-\text{NH}}| + \text{ClCH}_2-\text{Si}(\text{CH}_3)_2-\text{O}-\text{Si}(\text{CH}_3)_2-\text{CH}_2\text{Cl} + \text{HN}|\overline{-(\text{CH}_2)_n-\text{CO}} \longrightarrow$$

$$\overline{\text{CO}-(\text{CH}_2)_n-\text{N}}|\text{CH}_2\text{Si}(\text{CH}_3)_2-\text{O}-\text{Si}(\text{CH}_3)_2\text{CH}_2\text{N}|\overline{-(\text{CH}_2)_n-\text{CO}},$$

where n = 3 (VIII) or 5 (IX).

All the compounds prepared were colorless, odorless, viscous liquids. They were readily soluble in benzene, toluene, acetone, alcohols, and other organic solvents.

The compounds were identified by their elemental analyses and determination of their molecular weights, and their structures were confirmed by IR spectroscopy.

The compounds synthesized contained no bands in their IR spectra for stretching vibrations of the C–Cl bond at 774 cm^{-1}. Instead, all the spectra showed bands characteristic of deformational vibrations of the Si–CH_3 groups at 1260-1250 cm^{-1}, and antisymmetrical and symmetrical stretching vibrations of the Si–CH_2 bond at 820-760 and 700-650 cm^{-1} respectively.

Tbilisi University. Translated from Khimiko-farmatsevticheskii Zhurnal, Vol. 16, No. 5, pp. 560-562, May, 1982. Original article submitted September 9, 1981.

TABLE 1. Effects of N-Organosilylmethylenelactams on the Growth of Some Phytopathogenic Bacteria

Microorganisms	Control	Compound			
		VI	VII	II	III
Magnitude of the zone of depression of the test organisms, mm					
Bacterium tumefaciens	0	5,0	6,0	6,0	5,0
Xanthomonas sampestris	0	4,0	5,5	8,0	1,0
Pectobacterium aroideae	0	4,0	5,5	4,0	—

Characteristic bands were observed for the CO groups bonded to nitrogen atoms at 1680 cm^{-1} (for the N-organosilylmethylene-ε-caprolactams) and 1720 cm^{-1} (for the N-organosilylmethylene-2-pyrrolidines). In addition, the spectra of (I) and (IV) displayed bands at 1235-1220 cm^{-1} (Si—C_2H_5), (II) and (VI) at 1190 cm^{-1} (Si—C_4H_9), (III) and (VII) at 1429 cm^{-1} (Si—C_6H_5), and those of (VII) and (VIII) displayed bands at 1020-1030 cm^{-1} (Si—O—Si).

EXPERIMENTAL PHARMACOLOGICAL PART

Compounds (II), (III), (VI), and (VII) were tested as growth inhibitors for some phytopathogenic bacteria.

The test organisms used were the following microorganisms: *Bacterium tumefaciens* (the causal agent of a disease of grapevines), *Xanthomonas campestris*, and *Pectobacterium aroideae*, which infect some melons. Burholter's medium (potato broth 1 liter, peptone 5 g, Na_2HPO_4 2 g, NaCl 2 g, sodium citrate 1 g, asparagine 2 g, glucose 6 g, agar 20 g, and distilled water 1 liter) was used to culture the microorganisms. Toxic properties were determined by the inhibition zone method. The magnitude of the toxic effect was determined by the zone of sterility around the spot. The control was a mixture of the solvents used (ethanol and hexane). The compounds were introduced into the depressions in a concentration of 0.1%.

The results are shown in Table 1, from which it will be seen that these compounds possess toxic properties, inhibiting the growth of *Bacterium tumefaciens*, *Pectobacterium aroideae*, and *Xanthomonas campestris*.

EXPERIMENTAL CHEMICAL PART

IR spectra were obtained on an IR-20 instrument.

N-(Dimethylethylsilylmethylene)-2-pyrrolidone (I). In a round-bottomed flask fitted with a reflux condenser and a calcium chloride tube was placed a finely ground mixture of 12.5 g of 2-pyrrolidone and 15.41 g of anhydrous potassium carbonate. To the mixture was added 15.09 g of chloromethyldimethylethylsilane, and the mixture was heated for 4 h at 100°C. The reaction mixture was then cooled, and 100 ml of water added slowly. The reaction products were salted out with sodium chloride, and extracted with ether. Distillation *in vacuo* gave 10.44 g (36%) of (I), bp 73°C (1 mm), n_D^{20} 1.4460, d_4^{20} 0.8356. Found %: C 58.38; H 10.27; N 7.57; Si 15.14. M 179. $SiC_9H_{19}NO$. Calculated, %: C 57.85; H 10.77; N 6.36; Si 15.43. M 183.

N-Dimethylbutylsilylmethylene)-2-pyrrolidone (II). The reaction was carried out as described above. The reactants were 7.32 g 2-pyrrolidone, 3.19 g of potassium carbonate, and 10.59 g of chloromethyldimethylbutylsilane. Reaction was carried out for 4 h at 130°C. Yield 6.12 g (42%) of (II), bp 107°C (3 mm), n_D^{20} 1.4640, d_4^{20} 0.8996. Found, %: C 61.59; H 7.23; N 7.03; Si 11.20. M 210. $SiC_{11}H_{23}NO$. Found, %: C 61.97; H 7.52; N 6.57; Si 10.80. M 213.

N-(Dimethylphenylsilylmethylene)-2-pyrrolidone (III). The reaction was carried out with 15 g of 2-pyrrolidone, 6.51 g of potassium carbonate, and 24.49 g of chloromethyldimethylphenylsilane, at 100°C in toluene for 5 h. Yield 9.32 g (46%) of (III), bp 30°C (2 mm), n_D^{20} 1.4849, d_4^{20} 0.9836. Found, %: C 66.95; H 8.15; N 6.01; Si 12.02. M 236. $SiC_{13}H_{19}NO$. Calculated, %: C 67.12; H 7.83; N 6.43; Si 12.52. M 230.

N-(Dimethylethylsilylmethylene)-ε-caprolactam (IV). In a flask were placed 12.52 g of ε-caprolactam and 15.41 g of anhydrous potassium carbonate. To the mixture was added 15.09 g

of chloromethyldimethylethylsilane, and the mixture heated for 6 h at 100°C. Yield of (IV), 10.44 g (45%), bp 78°C (1 mm), n_D^{20} 1.4655, d_4^{20} 0.9864. Found, %: C 62.15; H 11.00; N 6.51; Si 13.0. M 205. $SiC_{11}H_{23}NO$. Calculated, %: C 62.30; H 11.62; N 6.61; Si 13.03. M 210.

N-(Dimethylpropylsilylmethylene)-ε-caprolactam (V). The reaction was carried out with 15 g of ε-caprolactam, 6.5 g of sodium carbonate, and 19.92 g of chloromethyldimethylpropylsilane. The synthesis was carried out at 30°C for 5 h, to give 12.21 g (41%) of (V), bp 125°C (2 mm), n_D^{20} 1.4670, d_4^{20} 1.0501. Found, %: C 63.93; H 11.19; N 6.72; Si 12.01. M200. $SiC_{12}H_{25}NO$. Calculated, %: C 63.45; H 11.01; N 6.12; Si 12.33. M 221.

N-(Dimethylbutylsilylmethylene)-ε-caprolactam (VI). The reaction was carried out with 7.3 g of ε-caprolactam, 3.19 g of potassium carbonate, and 10.59 g of chloromethyldimethylbutylsilane. The synthesis was carried out at 130°C for 4 h, to give 6.12 g (40%) of (VI), bp 208°C (1 mm), n_D^{20} 1.4570, d_4^{20} 1.0540. Found, %: 65.02; H 11.94; N 5.75; Si 11.93. M 241. $SiC_{13}H_{27}NO$. Calculated, %: C 64.82; H 11.22; N 5.81; Si 11.60. M 237.

N-(Dimethylphenylsilylmethylene)-ε-caprolactam (VII). The reaction was carried out with 15 g of ε-caprolactam, 6.52 g of potassium carbonate, and 24.49 g of chloromethyldimethylphenylsilane. The synthesis was carried out at 100°C for 5 h, to give 9.32 g (46%) of (VII), bp 170°C (1 mm), n_D^{20} 1.4720, d_4^{20} 1.0548. Found, %: C 66.12; H 8.77; N 5.31; Si 10.53. M240. $SiC_{15}H_{23}NO$. Calculated, %: C 66.43; H 8.47; N 5.16; Si 10.32. M 268.

1,3-Bis-(2'-pyrrolidone-N-methylene)tetramethyldisiloxane (VIII). The reaction was carried out for 6 h at 140°C, with 10 g of 2-pyrrolidone, 13.1 g of 1,3-bis(chloromethyl)-tetramethyldisiloxane and 8.1 g of potassium carbonate, to give 13.17 g (70%) of (VIII), bp 185°C (2 mm), n_D^{20} 1.4040, d_4^{20} 1.0054. Found, %: C 51.22; H 8.54; N 8.54; Si 17.07. M 328. $Si_2C_{14}H_{28}O_3N_2$. Calculated, %: C 51.50; H 8.04; N 9.09; Si 17.10. M 320.

1,3-Bis-(ε-caprolactam-N-Methylene)tetramethyldisiloxane (IX). The reaction was carried out for 5 h at 140°C with 10 g of ε-caprolactam, 10.15 g of 1,3-bis(chloromethyl)tetramethyldisiloxane, and 4.93 g of sodium carbonate, to give 11.4 g (64%) of (IX), bp 201°C (1 mm), n_D^{20} 1.4719, d_4^{20} 1.1037. Found, %: C 55.81; H 10.02; N 7.98; Si 14.82. M 370. $Si_2C_{18}H_{36}N_2O_3$. Calculated, %: C 56.25; H 9.37; N 7.29; Si 14.58. M 384.

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