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## SHORT COMMUNICATIONS

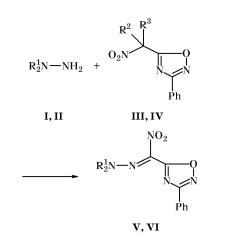
## Reaction of Unsymmetrically Substituted Hydrazines with 5-Nitromethyl-3-phenyl-1,2,4-oxadiazoles

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 $\alpha$ -C-Nitro hydrazones attract interest as compounds exhibiting a wide spectrum of fungicide and antimicrobial activity [1, 2]. It was also found that the antimicrobial effect against Staphilococcus aureus appreciably increases when the double bond (or a nitro or carbonyl group) is conjugated with a heteroring [3]. With the goal of obtaining new azole derivatives possessing antimicrobial activity and studying concurrent substituent effects in the nitromethyl fragment of 5-nitromethyl-3-phenyl-1,2,4-oxadiazoles we examined the reactions of N,N-diphenyl- and N,N-dimethylhydrazines I and II with 3-phenyl-5-trinitromethyl-1,2,4-oxadiazole (III) and ethyl chloro(nitro)-3-phenyl-1,2,4-oxadiazol-5-ylacetate (IV). These reactions occurred under mild conditions and afforded hitherto unknown substituted 1,2,4-oxadiazol-5-yl-(nitro)methylene hydrazines V and VI:



I, V,  $R^1 = Ph$ ; II, VI,  $R^1 = Me$ ; III,  $R^2 = R^3 = NO_2$ ; IV,  $R^2 = Cl$ ,  $R^3 = COOEt$ .

Presumably, the process follows the same pattern as in the reaction of unsymmetrically substituted hydrazines with tetranitro- or halotrinitromethanes [4]. The structure of compounds **V** and **VI** was determined on the basis of the IR, <sup>1</sup>H NMR, and UV spectra.

Hydrazines **I** and **II** were prepared by treatment of the corresponding hydrochlorides with a solution of alkali, followed by extraction with diethyl ether.

A solution of 5 mmol of compound **III** [5] or **IV** [6] in 10 ml of dry diethyl ether was added at 0°C to a solution of 5 mmol of substituted hydrazine **I** or **II** in 50 ml of dry ether. The mixture was kept for 2 h at 25°C and evaporated, and the residue was subjected to chromatography in a  $250 \times 10$ -mm column charged with activated silica gel (Silicagel 100/400 µm) using benzene as eluent.

*N*,*N*-Diphenyl-*N'*-[nitro(3-phenyl-1,2,4-oxadiazol-5-yl)methylene]hydrazine (V). Yield 41%, mp 175–176°C (from ethanol). IR spectrum, v, cm<sup>-1</sup>: 1545, 1290 (NO<sub>2</sub>); 1645 (C=N). <sup>1</sup>H NMR spectrum, δ, ppm: 7.20–7.61 m. UV spectrum,  $\lambda_{max}$  (log ε), nm: 240 (4.11), 375 (3.98). Found, %: C 65.39; H 3.84; N 18.10. C<sub>21</sub>H<sub>15</sub>N<sub>5</sub>O<sub>3</sub>. Calculated, %: C 65.46; H 3.90; N 18.18.

*N*,*N*-Dimethyl-*N'*-[nitro(3-phenyl-1,2,4-oxadiazol-5-yl)methylene]hydrazine (VI). Yield 45%, mp 63–64°C (from ethanol). IR spectrum, v, cm<sup>-1</sup>: 1545, 1290 (NO<sub>2</sub>); 1645 (C=N). <sup>1</sup>H NMR spectrum, δ, ppm: 7.55 m (5H, H<sub>arom</sub>), 2.83 s (6H, CH<sub>3</sub>). UV spectrum,  $\lambda_{max}$ , nm (log ε): 240 nm (4.11), 377 (3.99). Found, %: C 50.51; H 4.17; N 26.75. C<sub>11</sub>H<sub>11</sub>N<sub>5</sub>O<sub>3</sub>. Calculated, %: C 50.58; H 4.22; N 26.82.

The IR spectra were recorded on an IKS-29 spectrometer in chloroform. The <sup>1</sup>H NMR spectra were obtained on a Tesla BS-487C instrument operating at 80 MHz; acetone- $d_6$  was used as solvent, and hexamethyldisiloxane, as internal reference. The electron absorption spectra were measured on an SF-8 spectrophotometer in ethanol.

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