

SHORT  
COMMUNICATIONSSynthesis of 6-Butyl-7-methylidene-8-phenyl-5*H*-pyrrolo-  
[3,4-*d*]tetrazolo[1,5-*a*]pyrimidin-5-oneV. L. Gein<sup>a</sup>, O. S. Panova<sup>a</sup>, E. P. Tsypliyakova<sup>a</sup>, L. I. Varkentin<sup>a</sup>,  
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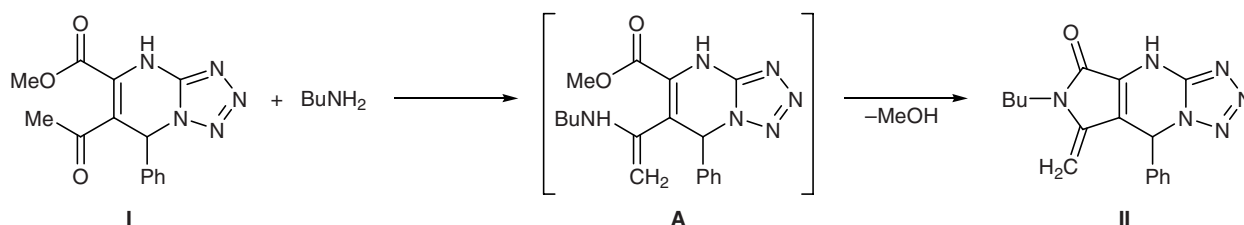
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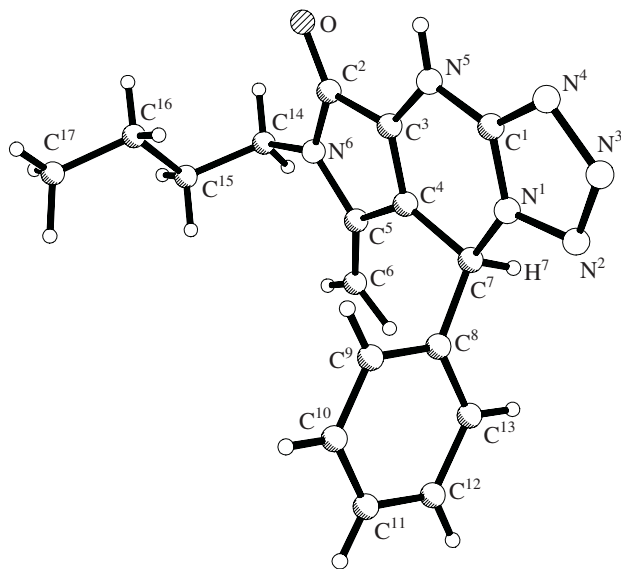
We have found that methyl 6-acetyl-4,7-dihydro-7-phenyltetrazolo[1,5-*a*]pyrimidine-5-carboxylate (**I**) reacts with an equimolar amount of butylamine at 130–150°C to give in 20–30 min 6-butyl-7-methylidene-8-phenyl-5*H*-pyrrolo[3,4-*d*]tetrazolo[1,5-*a*]pyrimidin-5-one (**II**). Presumably, in the first stage intermediate amine **A** is formed, and its enamino tautomer undergoes intramolecular cyclization to compound **II**. The latter was isolated as a yellow crystalline substance which is insoluble in water and ethanol, soluble in glacial acetic acid and acetonitrile on heating, and readily soluble in DMSO and DMF.

Compound **II** displayed in the IR spectrum absorption bands belonging to stretching vibrations of the lactam carbonyl group (1692 cm<sup>-1</sup>), secondary amino group (3144 cm<sup>-1</sup>), and double C=C bonds (1632 cm<sup>-1</sup>). Its <sup>1</sup>H NMR spectrum contained signals from aromatic protons and protons in the butyl group, doublets at δ 4.43 and 4.95 ppm (*J* = 2.5 Hz) from protons in the exocyclic methylene group, NH signal at δ 11.73 ppm, and a singlet at δ 7.02 ppm from the 8-H proton. In the mass spectrum of **II** we observed the molecular ion peak [*M*]<sup>+</sup> with *m/z* 322 and fragment ion peaks with *m/z* 294 [*M* - CO]<sup>+</sup>, 245 [*M* - Ph]<sup>+</sup>, and 77 [Ph]<sup>+</sup>, which are consistent with the assigned structure.

By slow crystallization from acetonitrile we succeeded in obtaining a single crystal of compound **II** suitable for X-ray analysis (see figure). The tricyclic fragment of molecule **II** is planar. The double bonds therein are localized with no appreciable conjugation effect. All bond lengths and bond angles have their usual values. Molecules **II** in crystal are linked through intermolecular hydrogen bonds N<sup>5</sup>-H<sup>5</sup>...N<sup>4</sup>, giving rise to centrosymmetric dimers. The distance N<sup>5</sup>...N<sup>4</sup> is 2.908 Å, indicating that the intermolecular hydrogen bond is not very strong.

**6-Butyl-7-methylidene-8-phenyl-5*H*-pyrrolo-[3,4-*d*]tetrazolo[1,5-*a*]pyrimidin-5-one (**II**).** A mixture of 5 mmol of ester **I** [1] and 5 mmol of butylamine was heated at 130–150°C until gaseous products no longer evolved (for 20–30 min). After cooling, the mixture was treated with ethanol, and the precipitate was filtered off and recrystallized from acetonitrile. Yield 0.5 g (31%), mp 196–198°C. IR spectrum, ν, cm<sup>-1</sup>: 1692 (CO), 3144 (NH), 1632 (CH<sub>2</sub>=). <sup>1</sup>H NMR spectrum, δ, ppm: 4.43 d (1H, CH<sub>2</sub>=, *J* = 2.5 Hz), 4.95 d (1H, CH<sub>2</sub>=, *J* = 2.5 Hz), 7.02 s (1H, CH), 11.73 s (1H, NH), 7.41 m (5H, C<sub>6</sub>H<sub>5</sub>), 0.87 m (3H, CH<sub>3</sub>), 1.25 m (2H, 3'-H), 1.46 m (2H, 2'-H), 3.57 m (2H, 1'-H). Found, %: C 63.22, 63.45; H 5.51, 5.74;





Structure of the molecule of 6-butyl-7-methylidene-8-phenyl-5*H*-pyrrolo[3,4-*d*]tetrazolo[1,5-*a*]pyrimidin-5-one (**II**) according to the X-ray diffraction data.

N 25.95, 26.19. C<sub>17</sub>H<sub>18</sub>N<sub>6</sub>O. Calculated, %: C 63.34; H 5.63; N 26.07.

The IR spectrum was recorded on a Specord M-80 spectrometer from a sample of **II** dispersed in mineral oil. The <sup>1</sup>H NMR spectra were measured on a Varian Mercury-300 (300.056 MHz) and Bruker DRX-500 (500.13 MHz) instruments using DMSO-*d*<sub>6</sub> as solvent and TMS as internal reference. The mass spectrum (electron impact, 70 eV) was obtained on a Finnigan MAT INCOS-50 mass spectrometer.

A set of experimental reflections was acquired on a KM-4 automatic four-circle diffractometer with  $\chi$ -geometry (monochromatized MoK $\alpha$  irradiation,  $\omega/2\theta$  scanning to  $2\theta \leq 50.14^\circ$ ). Monoclinic crystals with the following unit cell parameters:  $a = 7.815(2)$ ,  $b = 3.291(3)$ ,  $c = 15.766(3)$  Å;  $\beta = 90.96(3)^\circ$ ;  $V = 1637.4(6)$  Å<sup>3</sup>;  $M = 322.37$ ;  $d_{\text{calc}} = 1.308$  g/cm<sup>3</sup>;  $Z = 4$ ; space group  $P2(1)/c$ . Total of 2864 independent reflections ( $R_{\text{int}} 0.0208$ ) were measured. No correction for absorption was introduced ( $\mu = 0.087$  mm<sup>-1</sup>). The structure was solved by the direct method using SIR92 program [2], followed by a series of calculations of electron density maps. The positions of hydrogen atoms were set on the basis of geometry considerations. Full-matrix anisotropic refinement of the positions of non-hydrogen atoms by the least-squares procedure using SHELXL-97 program [3] was complete at  $R_1 = 0.0460$ ,  $wR_2 = 0.1163$  [from 1368 reflections with  $I \geq 2\sigma(I)$ ]; goodness of fit 0.889.

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## REFERENCES

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