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RECYCLIZATION OF CONDENSED CARBETHOXYPYRIMIDINES ACCOMPANIED BY SUBSTITUTION OF A CARBON ATOM INTO THE HETEROCYCLE

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Condensation in ethanol of ethyl ethoxymethyleneacetoacetate with systems containing an amidine fragment (substituted 3-aminopyrazoles and 3-amino-1,2,4-triazole) gave 6-carbethoxy-7-methylpyrazolo[1,5-a]pyrimidines. Addition of base to solutions of the obtained bicyclic carbethoxy derivatives in the course of several minutes caused rearrangement to 6-acetyl-7-hydroxypyrazolo-[1,5-a]pyrimidine and 6-acetyl-7-hydroxy-1,2,4-triazolo[1,5-a]pyrimidine respectively. A more prolonged refluxing in 15% aqueous alcohol solution of base caused 6-carbethoxy-7-methyl-2-phenylpyrazolo[1,5-a]pyrimidine and 6-acetyl-7-hydroxy-2-phenylpyrazolo[1,5-a]pyrimidine to recyclize to 7-methylpyrazolo[1,5-a]pyrimidine.

Keywords: pyrazolo[1,5-*a*]pyrimidine, 1,2,4-triazolo[1,5-*a*]pyrimidine, rearrangement, recyclization.

Among recyclizations of pyrimidines the most studied are Dimroth rearrangements [1] accompanied by exchange of a ring heteroatom with an exocyclic nitrogen atom (N-N recyclization conditions). We have studied a schematic relative of the Dimroth rearrangement in detail for conversion of pyrimidines to pyridine derivatives (the Kost–Sagitullin rearrangement [2-6]). Under these conditions a nitrogen atom of the pyrimidine ring is substituted by an exocyclic carbon atom occurring at the 2 position (N-C recyclization conditions). This communication relates to our study of a further recyclization conversion of pyrimidines which, according to the evidence, involves the substitution of an atom in the heterocycle and is a C-C recyclization. A similar rearrangement has previously been noted in a series of 2-substituted 5-carbethoxy-4-methylpyrimidines. Upon heating in sodium ethylate solution they are converted to the corresponding 2-substituted 5-acetyl-4-hydroxypyrimidines [7]. We have previously shown [8] that 2-substituted 4-amino-5-carbethoxypyrimidines are converted to the corresponding 5-carbamoyl-5-hydroxypyrimidines by treatment with base. It is also clear that the transformation noted above for the 5-carbethoxy-4-methylpyrimidines is, in fact, only achieved when treating the reaction mixture with water, i.e. under the action of hydroxide ion formed in solution.

In a continuation of this work we have studied the possibility of carrying out similar rearrangements for condensed bicyclic pyrimidines, in particular for 6-carbethoxy-7-methylpyrazolo[1,5-*a*]- and 6-carbethoxy-7-methyl-1,2,4-triazolo[1,5-*a*]pyrimidine derivatives. Model compounds for study were synthesized by condensation of ethyl ethoxymethyleneacetoacetate in ethanol with systems containing amidine fragments, *viz*. 3-aminopyrazoles and 3-amino-1,2,4-triazole.

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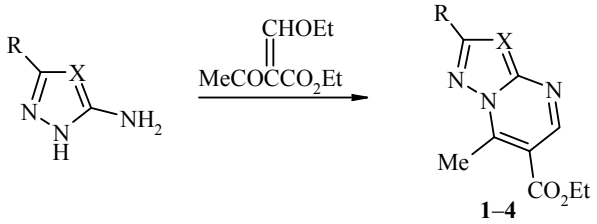
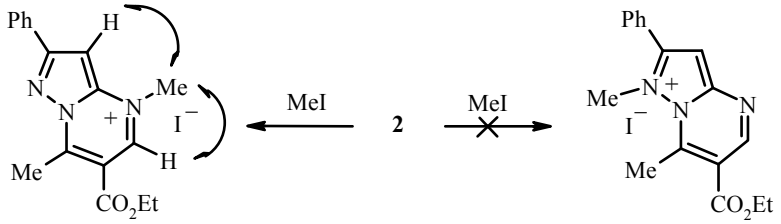


1–4

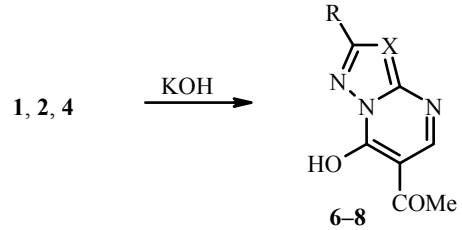
1 R = OH, X = CH; **2** R = Ph, X = CH; **3** R = CH_2CN, X = CCN; **4** R = H, X = N

The structures of all of the compounds including the presence in the pyrimidine ring of carbethoxy and methyl groups (i.e. the occurrence of a condensation *via* the acetyl and ethoxymethylene fragments of the reagent) were confirmed by spectroscopic methods and the position of the methyl group in the condensed compounds was established on the basis of the ^{1}H NMR spectra of their iodomethylates with the use of the NOESY method. In particular, the spectrum of compound **5** (the iodomethylate of compound **2**) shows a response between the protons of the N-methyl group and the CH of the pyrazole ring which points to alkylation of the $N_{(4)}$ atom. At the same time the ^{1}H NMR spectrum shows a clear cross peak between the signal of the same N-methyl group and the pyrimidine ring proton thus unambiguously proving their close positioning in the molecule. The absence in the NOESY spectrum of any kind of interaction between the protons of the two methyl groups also points to the positioning of the methyl group in compound **5**, as for the other synthesized compounds, at position 7 of the pyrazolo[1,5-*a*]pyrimidine ring, i.e. removed from the quaternary nitrogen atom.



5

The rearrangement of the pyrazolo[1,5-a]pyrimidines **1** and **2** and the triazolo[1,5-a]pyrimidine **4** to give the corresponding 6-acetyl-7-hydroxy derivatives **6-8** occurs over several minutes, even at room temperature. The rearrangement product of compound **3** could not be separated, evidently because concurrent process of transformation of the nitrile groups occurs under these conditions to form a mixture of substances.

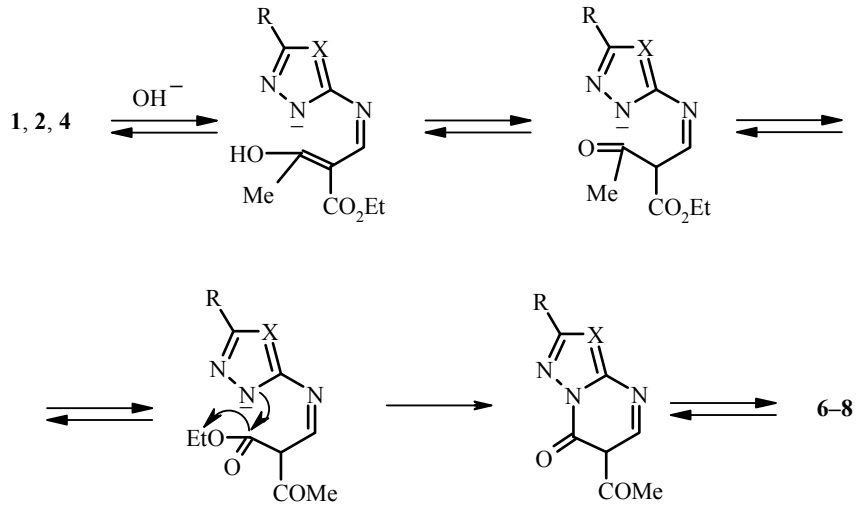


6–8

6 R = OH, X = CH; **7** R = Ph, X = CH; **8** R = H, X = N

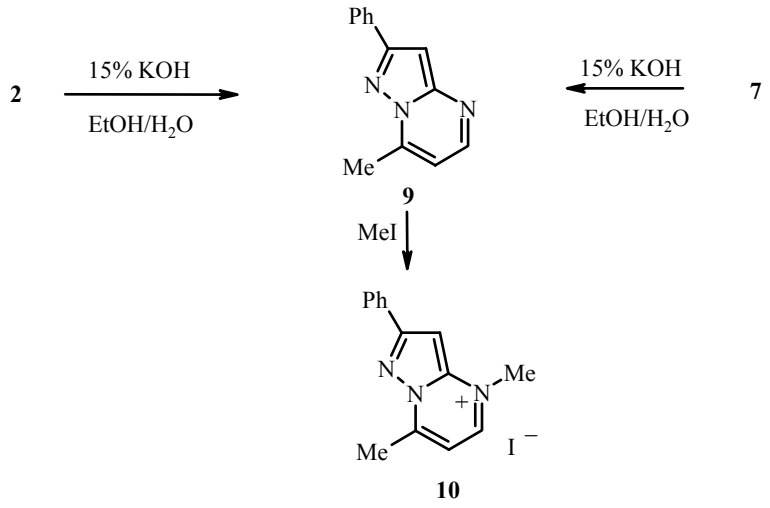
The ^{1}H NMR spectra of the recyclization products show the absence of ester proton signals characteristic of the starting materials and the appearance of a broadened signal for the hydroxyl group. The ^{13}C NMR spectra of the final transformation products differ from those of the starting materials and correspond to those expected. The structures of the rearrangement products were also confirmed by mass spectrometry.

The rearrangement probably occurs by the following scheme:

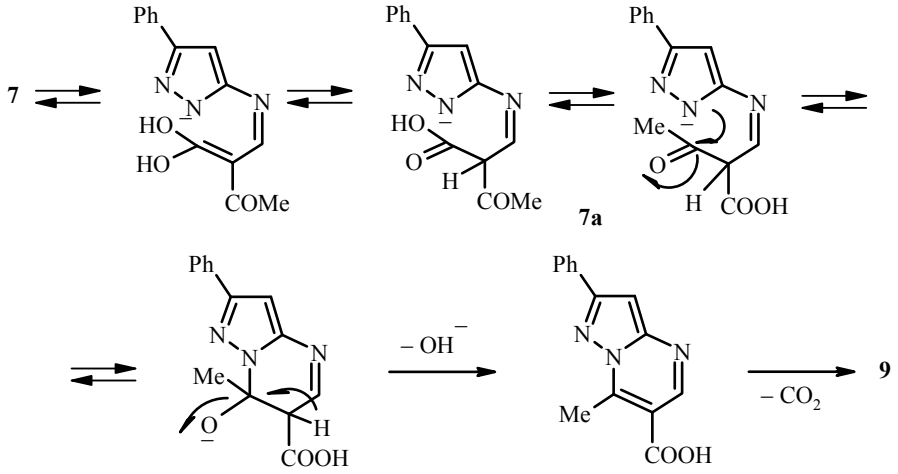


Evidently, in the recyclization process under the action of hydroxide ion there occurs a fission of the pyrimidine ring at the N–$C_{(7)}$ bond, subsequent rotation around the $C_{(5)}$–$C_{(6)}$ single bond, and repeated recyclization. As a result, the ester group C atom is included in the newly formed pyrimidine ring while the $C_{(7)}$ atom which was situated in the pyrimidine ring appears outside the heterocycle.

It should be stressed that, in the example of the transformation of the condensed systems **1**, **2**, and **4**, we have not excluded the likelihood of other possible types of recyclization occurring as mentioned at the beginning of our paper. Hence in the example of 6-carbethoxy-7-methylpyrazolo[1,5-*a*]pyrimidines a possible transformation route might be a conversion to a pyrazolo[3,4-*b*]pyridine derivative by a Kost–Sagitullin type rearrangement [9] *via* fission of the N–$C_{(7)}$ bond of the pyrimidine ring and subsequent cyclization by attack at the $C_{(4)}$ atom of the azole. However, in the conditions indicated above we were unable to separate the expected reaction products. Prolonged heating in a 15% aqueous alcohol solution of base, i.e. in the conditions reported before for the conversion of pyrazolo[1,5-*a*]pyrimidines *via* a Kost–Sagitullin rearrangement [9], the pyrazolo[1,5-*a*]pyrimidines **2** and **7** were unexpectedly for us transformed to 7-methyl-2-phenylpyrazolo[1,5-*a*]-pyrimidine (**9**). The formation of the latter can be explained by the occurrence of a series of consecutive reactions which include opening of the pyrimidine ring, cyclization, and decarboxylation. The consecutive chain of recyclizactions is also evidenced by the chromatographic formation and disappearance of compound **7** in the process of transformation of compound **2** to compound **9**.



The structure of the pyrazolopyrimidine **9** and its iodomethylate **10** were proved by the NOESY method and also using mass spectrometry and ^{1}H NMR and ^{13}C NMR spectroscopy. In the NOESY NMR spectrum of the iodomethylate **10**, as in the spectrum of compound **5**, a response was noted between the N-methyl group protons and the pyrazole H-3 and pyrimidine H-5 ring protons. Recyclization of compound **7** to compound **9** occurs by the following scheme (recalling that the conversion of the carbethoxy derivative **2** to compound **9** takes place *via* the stage of formation of the acetyl derivative **7**):



The formation of compound **9** rather than the realization of a Kost–Sagitullin rearrangement is explained by the insufficient nucleophilicity of atom $C_{(4)}$ in the pyrazole ring of intermediate **7a** when compared with atom $N_{(1)}$.

TABLE 1. Parameters for the Synthesized Compounds

Compound	Empirical formula	Found, % / Calculated, %			mp, °C	R_f*	Yield, % (method)
		C	H	N			
1	$C_{10}H_{11}N_3O_3$	54.45 / 54.30	5.12 / 5.01	18.86 / 19.00	180-182	0.66	84
2	$C_{16}H_{15}N_3O_2$	68.52 / 68.31	5.41 / 5.37	14.90 / 14.94	146-147	0.72	99
3	$C_{13}H_{11}N_5O_2$	57.78 / 57.99	4.02 / 4.12	26.18 / 26.00	126-128	0.71	77
4	$C_9H_{10}N_4O_2$	52.35 / 52.42	4.71 / 4.89	26.97 / 27.17	99-100	0.78	90
5	$C_{16}H_{15}N_3O_2 \cdot MeI$	48.51 / 48.24	4.25 / 4.29	10.12 / 9.93	250-251	—	93
6	$C_8H_7N_3O_3$	49.36 / 49.75	3.58 / 3.65	21.63 / 21.75	240-241	0.72	58
7	$C_{14}H_{11}N_3O_2$	66.32 / 66.40	4.26 / 4.38	16.43 / 16.59	262-263	0.63	67
8	$C_7H_6N_4O_2$	47.05 / 47.19	3.48 / 3.39	31.67 / 31.45	260-262	0.62	60
9	$C_{13}H_{11}N_3$	74.52 / 74.62	5.39 / 5.30	20.29 / 20.08	94-95	0.65	62 (A), 57 (B)
10	$C_{13}H_{11}N_3 \cdot MeI$	47.74 / 47.88	3.89 / 4.02	11.70 / 11.97	280-281	—	80

* Systems: toluene–acetone, 1:1 (compounds **1**, **4**); benzene–acetone, 3:1 (compounds **2**, **3**, **9**); ethanol (compounds **6-8**)

TABLE 2. ^{1}H NMR and ^{13}C NMR Spectra of the Condensed Pyrimidines **1-10**

Com-pound	^{1}H NMR spectrum, δ, ppm (*J*, Hz)	^{13}C NMR spectrum, δ, ppm
1	1.43 (3H, t, *J* = 7.1, $CH_2\underline{CH_3}$); 3.14 (3H, s, CH_3-7); 4.43 (2H, q, *J* = 7.1, OCH_2); 6.16 (1H, s, H-3); 8.95 (1H, s, H-5); 9.65-10.4 (1H, br. s OH)	14.43 ($CH_2\underline{CH_3}$); 15.29 (CH_3-7); 61.69 (CH_2); 83.39 ($C_{(3)}$); 109.97 ($C_{(6)}$); 149.85 ($C_{(7)}$); 150.34 (C_{ipso}); 151.23 ($C_{(5)}$); 164.82 (CONH); 167.94 (C=O)
2	1.43 (3H, t, *J* = 7.1, $CH_2\underline{CH_3}$); 3.29 (3H, s, CH_3-7); 4.43 (2H, q, *J* = 7.1, OCH_2); 7.05 (1H, s, H-3); 7.43 (3H, m, H-3',4',5'); 8.03 (2H, m, H-2',6'); 8.97 (1H, s, H-5)	14.39 ($CH_2\underline{CH_3}$); 15.17 (CH_3-7); 61.58 (CH_2); 94.97 ($C_{(3)}$); 110.5 ($C_{(6)}$); 126.85 ($C_{(3')}$ and $C_{(5')}$); 128.92 ($C_{(2')}$ and $C_{(6')}$); 129.57 ($C_{(4')}$); 132.5 ($C_{(1')}$); 149.8 ($C_{(7)}$); 149.95 (C_{ipso}); 151.5 ($C_{(2)}$); 157.9 ($C_{(5)}$); 164.9 (C=O)
3	1.44 (3H, t, *J* = 7.1, $CH_2\underline{CH_3}$); 3.25 (3H, s, CH_3-7); 4.17 (2H, s, CH_2CN); 4.51 (2H, q, *J* = 7.1, OCH_2); 9.20 (1H, s, H-5)	14.37 ($CH_2\underline{CH_3}$); 15.42 (CH_3-7); 17.96 (CH_2-2); 62.65 (OCH_2); 94.0 ($C_{(3)}$); 111.06 ($C_{(6)}$); 114.03 ($CH_2\underline{CN}$); 114.40 (CN); 151.16 ($C_{(7)}$); 151.52 (C_{ipso}); 153.11 ($C_{(2)}$); 154.17 ($C_{(5)}$); 163.49 (C=O)
4	1.43 (3H, t, *J* = 7.1, $CH_2\underline{CH_3}$); 3.27 (3H, s, CH_3-7); 4.45 (2H, q *J* = 7.1, OCH_2); 8.48 (1H, s, H-2); 9.32 (1H, s, H-5)	14.37 ($CH_2\underline{CH_3}$); 15.81 (CH_3-7); 62.31 (CH_2); 113.48 ($C_{(6)}$); 152.93 ($C_{(7)}$); 155.56 (C_{ipso}); 155.75 ($C_{(2)}$); 157.23 ($C_{(5)}$); 163.85 (C=O)
5	1.48 (3H, t, *J* = 7.1, $CH_2\underline{CH_3}$); 3.21 (3H, s, CH_3-7); 4.46 (3H, s, N–CH_3); 4.51 (2H, q, *J* = 7.1, OCH_2); 7.58 (3H, m, C_6H_5); 8.03 (1H, s, H-3); 8.21 (2H, m, C_6H_5); 9.74 (1H, s, H-5)	14.43 ($CH_2\underline{CH_3}$); 17.71 (CH_3-7); 43.81 (N–CH_3); 62.08 (CH_2); 90.41 ($C_{(6)}$); 107.18 ($C_{(3)}$); 126.81 ($C_{(3')}$ and $C_{(5')}$); 128.84 ($C_{(2')}$ and $C_{(6')}$); 130.31 ($C_{(1')}$); 130.43 ($C_{(4')}$); 141.29 (C_{ipso}); 148.40 ($C_{(5)}$); 156.31 ($C_{(7)}$); 157.70 ($C_{(2)}$); 164.95 (CO)
6	2.8 (3H, s, $COCH_3$); 5.81 (1H, s, H-3); 6.1-6.9 (1H, br. s OH); 8.59 (1H, s, H-5)	
7	3.22 (3H, s, CH_3); 7.03 (1H, s, H-3); 7.41 (3H, m, H-3',4',5'); 7.91 (2H, m, H-2',6'); 8.85 (1H, s, H-5); 13.18 (1H, br. s OH)	14.39 (CH_3); 93.87 ($C_{(3)}$); 110.67 ($C_{(6)}$); 126.11 ($C_{(3')}$ and $C_{(5')}$); 128.1 ($C_{(2')}$ and $C_{(6')}$); 128.58 ($C_{(4')}$); 132.06 ($C_{(1')}$); 144.26 ($C_{(7)}$); 149.63 (C_{ipso}); 149.92 ($C_{(2)}$); 156.41 ($C_{(5)}$); 165.55 (C=O)
8	2.62 (3H, s, CH_3); 4.5-5.6 (1H, br. s OH); 8.11 (1H, s, H-2); 8.51 (1H, s, H-5)	30.15 (CH_3); 110.49 ($C_{(6)}$); 145.78 ($C_{(5)}$); 149.95 ($C_{(7)}$); 151.28 ($C_{(2)}$); 154.4 (C_{ipso}); 192.89 (C=O)
9	2.85 (3H, d, *J* = 0.8, CH_3-7); 6.68 (1H, dq, J_1 = 0.8, J_2 = 4.5, H-6); 7.00 (1H, s, H-3); 7.43 (3H, m, H-3',4',5'); 8.05 (2H, m, H-2',6'); 8.36 (1H, d, *J* = 4.5, H-5)	17.40 (CH_3); 93.83 ($C_{(3)}$); 107.56 ($C_{(6)}$); 126.79 ($C_{(3')}$ and $C_{(5')}$); 128.93 ($C_{(2')}$ and $C_{(6')}$); 129.07 ($C_{(4')}$); 133.24 ($C_{(1')}$); 146.24 ($C_{(5)}$); 148.72 ($C_{(7)}$); 150.19 (C_{ipso}); 155.92 ($C_{(2)}$)
10	3.11 (3H, s, CH_3-7); 4.39 (3H, s, N–CH_3); 7.50–7.60 (3H, m, C_6H_5); 7.60 (1H, d, *J* = 6.2, H-6); 7.88 (1H, s, H-3); 8.16 (2H, dd, J_1 = 7.9, J_2 = 1.8, C_6H_5); 9.21 (1H, d, *J* = 6.2, H-5)	17.67 (CH_3-7); 43.76 (N–CH_3); 90.95 ($C_{(6)}$); 107.18 ($C_{(3)}$); 126.79 ($C_{(3')}$ and $C_{(5')}$); 128.78 ($C_{(2')}$ and $C_{(6')}$); 130.03 (*ipso*-C_6H_5); 130.39 ($C_{(4')}$); 141.33 (C_{ipso}); 148.37 ($C_{(5)}$); 156.41 ($C_{(7)}$); 157.72 ($C_{(2)}$)

EXPERIMENTAL

^{1}H NMR spectra were obtained in the Molecular Structure Research Center in the Armenian Republic National Academy of Sciences (US CRDF RESC 17-5 program) on a Varian Mercury 300 instrument (300 and 75 MHz respectively) using $CDCl_3$ (compounds **1-4**, **9**) or DMSO-d_6 (**5-8** and **10**) with TMS internal standard and a sample temperature of 303 K. Mass spectra were recorded on an MK-1321 spectrometer with direct introduction of the sample into the ion source and an ionization energy of 70 eV. TLC was performed on Silufol UV-254 plates and revealed using iodine vapor or the Ehrlich reagent.

The physicochemical and spectroscopic parameters are given in Tables 1 and 2.

6-Carbethoxy-2-hydroxy-7-methylpyrazolo[1,5-*a*]pyrimidine (1). A mixture of 3-aminopyrazol-5-one [10] (1 g, 10 mmol), ethyl ethoxymethyleneacetoacetate (1.9 g, 10 mmol) and glacial acetic acid (15 ml) was refluxed for 5-6 h. The major part of the acetic acid was removed by distillation in vacuo. The crystals formed were filtered off, washed with acetone, and recrystallized from 50% ethanol to give compound **1** (1.85 g) as light brown crystals.

6-Carbethoxy-7-methyl-2-phenylpyrazolo[1,5-*a*]pyrimidine (2). A mixture of 3-amino-5-phenylpyrazole [11] (1.44 g, 9 mmol), ethyl ethoxymethyleneacetoacetate (1.7 g, 9 mmol), and absolute ethanol (10 ml) was stirred for 10 min at about 20°C. After a completely solid mass had formed it was filtered off and washed with acetone to give compound **2** (2.5 g).

6-Carbethoxy-3-cyano-2-cyanomethyl-7-methylpyrazolo[1,5-*a*]pyrimidine (3). A mixture of 3-amino-4-cyano-5-cyanomethylpyrazole [12] (1 g, 7 mmol), ethyl ethoxymethyleneacetoacetate (1.3 g, 7 mmol), and absolute ethanol (10 m) was refluxed for 4-5 h. The crystals formed were filtered off, washed with hexane, and recrystallized from ethanol to give compound **3** (1.4 g) as orange crystals.

6-Carbethoxy-7-methyl-1,2,4-triazolo[1,5-*a*]pyrimidine (4). A mixture of 3-amino-1,2,4-triazole (0.85 g, 10 mol), ethyl ethoxymethyleneacetoacetate (1.9 g, 10 mmol), and absolute ethanol (15 ml) was refluxed for 1 h. The product was cooled and the crystals formed were recrystallized from ethanol to give compound **4** (1.85 g).

6-Acetyl-2,7-dihydroxypyrazolo[1,5-*a*]pyrimidine (6). An alcoholic solution of KOH prepared from KOH (0.56 g, 10 mmol) in absolute ethanol (10 ml) was added to a hot solution of compound **1** (1.1 g, 5 mmol) in absolute ethanol (10 ml). The crystals formed were filtered off, dissolved in a minimum amount of water, and acidified using dilute HCl solution to pH 6. The crystals were filtered off, washed with acetone, and recrystallized from ethanol to give compound **6** (0.56 g).

6-Acetyl-7-hydroxy-2-phenylpyrazolo[1,5-*a*]pyrimidine (7). An alcoholic solution of KOH prepared from KOH (0.23 g, 4 mmol) in absolute ethanol (10 ml) was added to a solution of compound **2** (0.57 g, 2 mmol) in alcohol (15 ml). The instantly formed crystals were filtered off, dissolved in a minimum amount of water, and acidified using dilute HCl solution to pH 6. The crystals formed were filtered off and recrystallized from ethanol to give compound **7** (0.34 g). Mass spectrum, *m/z* (I_{rel}, %): 253 $[M]^+$ (100), 252 (12), 236 (14), 209 (14), 208 (11), 144 (12), 142(14), 127 (7), 77 (19), 67 (9), 28 (43).

6-Acetyl-7-hydroxy-1,2,4-triazolo[1,5-*a*]pyrimidine (8). Similarly to the above, a solution of KOH prepared from KOH (0.28 g, 5 mmol) in absolute ethanol (5 ml) was poured into a hot alcoholic solution of the triazolopyrimidine **4** (0.35 g, 1.7 mmol) in absolute ethanol (5 ml). The instantly formed crystals were filtered off after 10 min, dissolved in a minimum amount of water, and acidified using dilute HCl solution to pH 6. The solution was cooled to a negative temperature. After 1 h the crystals formed were filtered off and washed with acetone to give yellowish crystals of compound **8** (0.18 g). Mass spectrum, *m/z*, (I_{rel}, %): 178 $[M]^+$ (100), 163 (13), 162 (66), 161 (22), 149 (20), 134 (12), 107 (16), 94 (34), 67 (17), 55 (16), 43 (34).

7-Methyl-2-phenylpyrazolo[1,5-*a*]pyrimidine (9). A. The acetyl derivative **7** (0.8 g, 0.4 mmol) was dissolved in an aqueous alcoholic solution (1:1) (10 ml) of KOH (1.1 g, 2 mmol) and the product was refluxed for 20 h. At the end of the reaction the solvent was evaporated to dryness and the residue was washed twice with benzene. After some time crystals of compound **9** precipitated (100 mg) with mp 94-95°C. Mass spectrum, *m/z* (I_{rel}, %): 209 $[M]^+$ (18), 208 (100), 207 (21), 194 (6), 94 (6), 84 (21), 82 (13).

B. Similarly to the above using 10 ml of an aqueous alcohol solution of KOH (1.4 g) and the carbethoxy derivative **2** (0.28 g, 1 mmol) with refluxing for 20 h. The solvent was removed and the residue was treated with benzene to give compound **9** (0.12 g). The melting point and chromatographic mobility were the same as for the sample prepared by the counter method using the acetyl derivative **7**.

Preparation of 6-Carbethoxy-4,7-dimethyl-2-phenylpyrazolo[1,5-*a*]pyrimidinium (5) and 4,7-dimethyl-2-phenylpyrazolo[1,5-*a*]pyrimidinium (10) Iodides. The corresponding pyrazolo[1,5-*a*]-pyrimidine **2** or **9** (2.5 mmol) and methyl iodide (3 ml) were heated in a sealed ampul on a water bath. After 10 h the ampul was opened, the solvent was evaporated to dryness, and the residue was washed with hexane.

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