Acylation of spiro(1-pyrazoline-3,1′-cyclopropanes) to form 1-acyl-3-(2-chloroethyl)-2-pyrazolines and transformation of bicyclic 2-pyrazolines into 1,4,5,6-tetrahydropyridazines

Yu. V. Tomilov,* G. P. Okonnishnikova, E. V. Shulishov, and O. M. Nefedov*

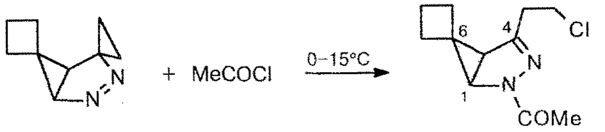
N. D. Zelinsky Institute of Organic Chemistry, Russian Academy of Sciences,
47 Leninsky prosp., 117913 Moscow, Russian Federation.
Fax: +7 (095) 135 5328

Strained polycyclic spiro(1-pyrazoline-3,1′-cyclopropanes) react with acetyl or benzoyl chlorides at 0—15 °C regioselectively to give in high yields corresponding 1-acyl-3-(2-chloroethyl)-2-pyrazolines. Under the same conditions 6-ethenyl-4,5-diazaspiro[2,4]hept-4-ene gives a mixture of two pyrazolines resulting from the acyl group attack directed at different nitrogen atoms. Bicyclic pyrazolines-2 obtained by acylation of the cycloaddition products of diazocyclopropane with 3,3-disubstituted cyclopropenes transform under the action of hydrogen chloride to 1,4,5,6-tetrahydropyridazines in high yields.

Key words: spiro(1-pyrazolines-1,3′-cyclopropane), 2,3-diazabicyclo[3.1.0]hex-3-enes, 1,4,5,6-tetrahydropyridazines, acylation reaction, opening of cyclopropane ring.

Recently we have found[1,2] that 1-pyrazolines having a spirocoupled cyclopropane fragment adjacent to the azo-group interact with acyl chlorides to form in almost quantitative yield corresponding 1-acyl-3-(2-chloroethyl)-2-pyrazolines, which are the products of 1,5-addition of acyl chlorides to the conjugated azocyclopropane system. In the present work the interactions of acetyl and benzoyl chlorides with strained polycyclic 1-pyrazolines (**1**—**3**) as well as with 6-ethenyl-4,5-diazaspiro[2,4]hept-4-ene (**4**) were investigated. The starting 1-pyrazolines **1**—**3** were obtained by the addition of *in situ* generated diazocyclopropane to spiro[2,4]hex-1-ene, benzvalene, and the dimer of spiro[2,4]heptadiene,[3] **4** was prepared by the addition of diazocyclopropane to butadiene.[3] The study of the related transformations, when a molecule has fragments potentially capable of addition of electrophilic reagents present in a molecule, is of interest both for evaluation of the regioselectivity of electrophilic addition reactions and for the search for new ways of synthesis of functionally substituted heterocyclic compounds.

We have determined that the interaction of equimolar amount of acetyl chloride with dispiropyrazoline **1** bearing two different cyclopropane fragments takes place exclusively at the conjugated (N=N)—spirocoupled cyclopropane ring system to form spiro{2-acetyl-4-(2-chloroethyl)-2,3-diazabicyclo[3.1.0]hex-3-ene-6,1′-cyclobutane} (**5**) with high selectivity. The reaction readily proceeds at 0—15 °C and in accordance with ^{1}H NMR data leads to adduct **5** in almoust quantitative yield. Analytically pure sample was obtained both by a microdistillation *in vacuo* (<1 Torr) and by passing **5** in benzene solution through a small silica gel layer, drying over $MgSO_4$ and removal of the solvent *in vacuo.*



1

1 : 1

5, >95%

Addition of one more equivalent of acetyl chloride to 2-pyrazoline **5** does not cause essential changes of the reaction mixture for several hours as evident from ^{1}H NMR spectra, but keeping it for 3—5 days at 10—15 °C leads to the partial transformation of **5**. Dichloride **6** formed by HCl (but not AcCl) addition to bicyclic pyrazoline **5** was identified as one of the products of this reaction. In this connection we have studied the direct interaction of gaseous HCl with 2-pyrazoline **5** and established that in CH_2Cl_2 or $CHCl_3$ solution at 0—15 °C the addition of HCl to the C(1)—C(5)-bond of the diazabicyclohexane fragment to produce 1,4,5,6-tetrahydropyridazine derivative **6** proceeds with high regioselectivity. According to NMR spectra the content of the major component is no less than 93 % in obtained practically colorless reaction mixture. The appearance of the proton resonance in very low field (δ_H = 6.48 ppm) and the presence of two non-equivalent geminal protons (δ_H = 2.58 and 2.28 ppm, J_{gem} = 17.2 Hz) indicate that the HCl addition proceeds with intracyclic opening of cyclopropane ring so that

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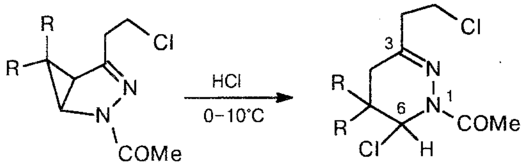
CHCl-fragment is arranged at amide N-atom. Although the proton is likely to attack initially imine N-atom.



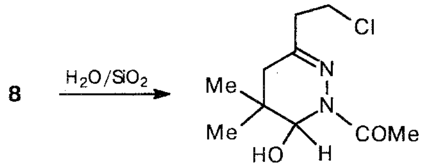
5: R + R = $(CH_2)_3$

6: R = Me

7, ≥93%

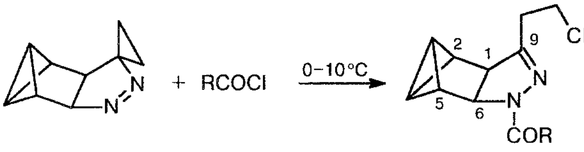
8, ≥85%

Analogous reaction is observed also when dry HCl is passed at 0—5 °C through solution of previously synthesized 2-acetyl-6,6-dimethyl-4-(2-chloroethyl)-2,3-diazabicyclo[3.1.0]hex-3-ene (**7**) (Ref. 2), whose structure is similar to that of 2-pyrazoline **5**. In this case in accordance with ^1H and ^{13}C NMR spectra 1,4,5,6-tetrahydropyridazine is formed in 85—88 % yield. The chromatography of the reaction mixture on silica gel gives instead of chloroderivative **8** 6-hydroxytetrahydropyridazine (**9**) in ~85 % yield. The structure of the latter was confirmed by NMR and mass spectra. When solution of **9** in $CDCl_3$ was shaken with D_2O the resonance of OH-group in the region of 4.2 ppm in ^1H NMR spectra disappered.



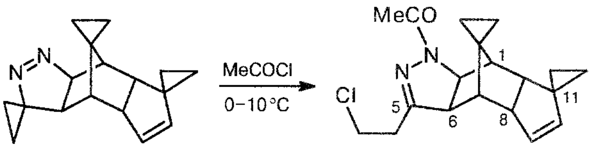
9, 85%

In contrast to N-substituted 2-pyrazolines **5** and **7** structures like 2,3-diazabicyclo[3.1.0]hex-2-enes, which are derivatives of 1-pyrazoline, open the three-membered ring in the presence of a catalytic amount of acid to transform into isomeric 1,4-dihydropyridazines.[4,5] Acylation of pyrazolines **2** and **3** proceeds exclusively at the conjugate azocyclopropane fragment, like **1**. Thus, the interaction of **2** with acetyl or benzoyl chloride gives in high yield 7-acetyl or 7-benzoyl-9-(2-chloroethyl)-7,8-diazatetracyclo[4.3.0.0^{2,4}.0^{3,5}]non-8-enes (**10**) with completely retained bicyclobutane fragment. Both compounds were purified by preparative TLC on silica gel; benzoyl derivative **10b** being obtained as the colorless crystalls with m.p. 82—83 °C.



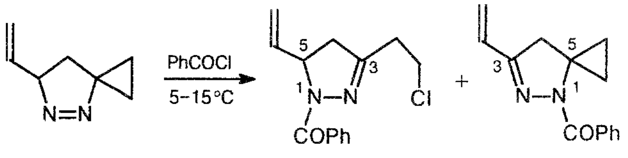
2 R = Me (**a**), Ph (**b**) **10**, >93%

Polycyclic pyrazoline **3** involved in the reaction as the isomer with anti-oriented cyclopropane fragments[3] reacts in like manner. Even with excess of acylating reagent at 0—10 °C the reaction is stopped on the stage of formation of polycyclic *N*-acetyl-(2-chloroethyl)-2-pyrazoline (**11**) which is produced in high yield as a result of selective 1,5-addition of AcCl to azocyclopropane fragment. Further addition of acetyl chloride to conjugated vinylcyclopropane system as opposed to azocyclopropane system proceeds when catalyst (for example, $AlCl_3$, $SnCl_4$) is added. However, in this case the reaction results in the mixture of difficultly identified compounds and resins.



3 **11**, ~95%

Acylation of 6-ethenyl-4,5-diazaspiro[2,4]hept-4-ene **4** by an equimolar amount of benzoyl chloride (CH_2Cl_2, 5—15 °C) occurs less selectively and gives rise to a mixture (ratio ~1 : 2) of two compounds, *i.e.*, the expected 1-benzoyl-3-(2-chloroethyl)-5-ethenyl-2-pyrazoline (**12**) and 1-benzoyl-3-ethenyl-2-pyrazoline (**13**), which is the product of formal substitution of a H-atom. The formation of pyrazoline **13** in this case demonstrates that the electrophilic attack at N-atom adjacent to cyclopropane ring is favoured. In this case the reaction is analogous to a transformation of usual monocyclic 3-substituted 1-pyrazolines into 2-pyrazolines.[6] However, the presence of vinyl substituent and the formation of conjugated double bonds system in compound **13** are likely to be driving force of this reaction path, because unsubstituted spiro{1-pyrazolin-3,1'-cyclopropane} is acylated almoust completely according to the 1,5-addition pathway[2] as it takes place in the course of pyrazoline **12** formation.



4 **12**, ~32% **13**, ~60%

Thus, azocyclopropane system in contrast to conjugated vinylcyclopropane group or strained carbocyclic fragments reacts smoothly with acyl chlorides under mild conditions and in the absence of a catalyst to form products of 1,5-addition, corresponding 1-acyl-3-(2-chloroethyl)-2-pyrazolines. The analogs of pyrazolines synthesized can be used as perspective starting compounds for the preparation of functionally substituted nitrogen containing heterocyclic compounds, in particular hydrogenated pyridazine derivatives.

Experimental

^{1}H and ^{13}C NMR spectra were registered on Bruker WM-250 (250 and 62.90 MHz) and Bruker AM-300 (300 and 75.47 MHz) spectrometers for solutions in $CDCl_3$ with 0.1 % TMS as the internal standard. IR spectra were obtained on a Bruker IFS-113v spectrometer in a thin layer. Mass spectra were registered on a Finnigan MAT INCOS-50 (70 eV) instrument with a capillary column (RSL-200, 30 m) or with direct injection of samples into the ion source. The starting spiro{1-pyrazoline-3,1′-cyclopropanes} were synthesized using the procedure previously discribed.^{3}

General procedure of acylation of spiro(1-pyrazoline-3,1′-cyclopropanes). A cooled solution of 5 mmol of acetyl or benzoyl chloride in 3 ml of solvent was added to a stirred solution of 5 mmol of 1-pyrazoline **1—4** in 3—4 ml of CH_2Cl_2 or $CHCl_3$ at 0—10 °C over 3—4 min. The mixture was stirred for 20—30 min, the solvent was evaporated *in vacuo,* and the practically colorless residue was analysed by NMR spectroscopy. The samples for analysis were isolated by microdistillation *in vacuo* or by preparative TLC (silica gel L 40—100 μm, ether—hexane, 3 : 1) and were identified by spectral techniques.

Spiro{2-acetyl-4-(2-chloroethyl)-2,3-diazabicyclo[3.1.0]-hex-3-en-6,1′-cyclobutane} (5). Yield ~95 %. B.p. 93—96 °C (0.6 Torr). IR (ν/cm^{-1}): 1670 (C=O), 1590 (C=N). ^{1}H NMR (δ, ppm, *J*/Hz): 4.39 (d, 1 H, $J_{1,5}$ = 5.5, H(1)), 3.72 (t, 2 H, *J* = 6.4, CH_2Cl), 2.78 (m, 2 H, $=CCH_2$), 2.48 (d, 1 H, $J_{1,5}$ = 5.5, H(5)), 2.23 (s, 3 H, Me), 2.17, 1.98 and 1.61 (three m, 3×2 H, 3 CH_2 in cyclobutane). ^{13}C NMR (δ, ppm): 169.4 (CO), 155.5 (C-4), 47.5 (C-1), 40.7 (CH_2Cl), 37.1 (C-5), 33.8 (CH_2 at C-4), 27.8, 23.0 and 15.5 (3 CH_2), 23.0 (C-6), 21.1 (Me). MS, *m/z* (*I*(%)): 228 (2) and 226 (5) M^+, 185 (4), 183 (11), 158 (24), 156 (63), 43 (100). Found (%): C, 57.89; H, 6.44; N, 12.31; Cl, 16.02. $C_{11}H_{15}ClN_2O$. Calculated (%): C, 58.28; H, 6.67; N, 12.36; Cl, 15.64.

7-Acetyl-9-(2-chloroethyl)-7,8-diazatetracyclo[4.3.0.0^{2,4}.0^{3,5}]non-8-ene (10a). Yield >93 %. B.p. 95—98 °C (0.6 Torr). IR (ν/cm^{-1}): 1652 (C=O), 1620 (C=N). ^{1}H NMR (δ, ppm, *J*/Hz): 4.45 (dt, 1 H, $J_{1,6}$ = 8.3, *J* ~1.4, H(6)), 3.67 (br t, 2 H, *J* ~7.0, CH_2Cl), 3.24 (br d, $J_{1,6}$ = 8.3, H(1)), 2.5—2.75 (m, 3 H, $=CCH_2$ and H(5)), 2.32 (d q, 1 H, $J_{2,5}$ = 4.9, *J* ~1.5, H(2)), 2.12 and 1.87 (two m, 2×1 H, $J_{3,4}$ = 8.7, *J* ~1.5, H(3) and H(4)), 2.10 (c, 3 H, Me). ^{13}C NMR (δ, ppm): 168.3 (CO), 156.5 (C-9), 64.1 (C-6), 56.0 (C-1), 40.1 (CH_2Cl), 38.8 (C-5), 37.3 (C-2), 32.5 (CH_2), 21.0 (Me), 7.6 and 0.4 (C-3, C-4). MS, *m/z* (*I*(%)): 226 (3) and 224 (9) M^+, 183 (4), 181 (11), 147 (48), 52 (96), 43 (100). Found (%): C, 58.37; H, 5.64; N, 12.41; Cl, 16.00. $C_{11}H_{13}ClN_2O$. Calculated (%): C, 58.80; H, 5.83; N, 12.47; Cl, 15.78.

7-Benzoyl-9-(2-chloroethyl)-7,8-diazatetracyclo[4.3.0.0^{2,4}.0^{3,5}]non-8-ene (10b). Yield ~95 %. M.p. 82—83 °C. ^{1}H NMR (δ, ppm, *J*/Hz): 7.83 and 7.35 (two m, 2 H, 3 H, Ph), 4.76 (d t, 1 H, $J_{1,6}$ = 8.3, *J* ~1.4, H(6)), 3.69 (br t, 2 H, *J* ~7.0, CH_2Cl), 3.30 (br d, $J_{1,6}$ = 8.3, H(1)), 2.86 (dq, 1 H, $J_{2,5}$ = 4.9, *J* ~1.5, H(5)), 2.69 (m, 2 H, ^2J = 16.3, ^3J ~7.0, $=CCH_2$), 2.38 (dq, 1 H, $J_{2,5}$ = 4.9, *J* ~1.5, H(2)), 2.22 and 1.96 (two m, 2×1 H, $J_{3,4}$ = 8.6, *J* ~1.5, H(3) and H(4)). ^{13}C NMR (δ, ppm): 165.5 (CO), 157.2 (C-9), 134.0 (C_{ipso} in Ph), 130.2 (C_p in Ph), 129.5 (C_o in Ph), 127.3 (C_m in Ph), 65.3 (C-6), 54.9 (C-1), 39.9 (CH_2Cl), 38.7 (C-5), 37.2 (C-2), 32.3 (CH_2), 7.0 and 0.2 (C-3, C-4). Found (%): C, 66.79; H, 5.42; N, 9.71; Cl, 12.92. $C_{16}H_{15}ClN_2O$. Calculated (%): C, 67.02; H, 5.27; N, 9.77; Cl, 12.36.

Dispiro{3-acetyl-5-(2-chloroethyl)-3,4-diazatetracyclo-[5.5.1.0^{2,6}.0^{8,12}]tridec-4-en-11,1′:13,1″-biscyclopropane} (11). Yield ~95 %. ^{1}H NMR (δ, ppm, *J*/Hz): 5.40 (br dd, 1 H, $J_{9,10}$ = 5.5, $J_{8,9}$ = 2.0, H(9)), 5.17 (dd, 1 H, $J_{9,10}$ = 5.5, $J_{8,10}$ = 1.7, H(10)), 4.62 (br d, 1 H, $J_{2,6}$ = 8.7, H(2)), 3.67 (t, 2 H, *J* = 7.0, CH_2Cl), 3.42 (m, 1 H, $J_{8,12}$ = 9.7, $J_{7,8}$ = 5.0, H(8)), 3.11 (br d, 1 H, $J_{2,6}$ = 8.7, H(6)), 2.62 (m, 2 H, ^2J = 16.1, ^3J = 7.0, CH_2 at C(5)), 2.45 (br dd, 1 H, $J_{8,12}$ = 9.7, $J_{1,12}$ = 4.8, H(12)), 2.12 (s, 3 H, Me), 1.79 (m, 2 H, H(1) and H(7)), 1.04, 0.77, 0.22—0.66, and 0.13 (four m, 1 H, 1 H, 5 H, 1 H, protons of cyclopropane rings). ^{13}C NMR (δ, ppm): 168.1 (CO), 158.5 (C-5), 140.5 (C-9), 128.3 (C-10), 60.3 (C-2), 54.4 (C-6), 52.3 (C-8), 48.5, 48.3, 48.1 (C-1, C-7, C-12), 40.1 (CH_2Cl), 32.5 (C-13), 32.2 (CH_2), 27.5 (C-11), 21.4 (Me), 15.6, 11.5 (CH_2CH_2 at C-11), 6.2 and 0.9 (CH_2CH_2 at C-13). Found (%): C, 68.74; H, 7.12; N, 8.37; Cl, 10.93. $C_{16}H_{15}ClN_2O$. Calculated (%): C, 68.98; H, 7.01; N, 8.47; Cl, 10.72.

1-Benzoyl-3-(2-chloroethyl)-5-ethenyl-2-pyrazoline (12) was characterized in the mixture with compound **13** by GLC-MS and NMR spectra. Yield ~32 %. ^{1}H NMR (δ, ppm, *J*/Hz): 7.80 and 7.30 (two m, Ph), 5.84 (ddd, J_{trans} = 16.5, J_{cis} = 9.6, *J* = 6.0, =CH), 5.18 (br d, J_{trans} = 16.5) and 5.11 (br d, J_{cis} = 9.6, $=CH_2$), 5.06 (m, H(5)), 3.60 (t, *J* = 6.6, CH_2Cl), 3.02 (br dd, J_{gem} = 17.3, $J_{4a,5}$ = 10.7, H(4a)), 2.69 (m, $=CCH_2$), 2.56 (br dd, J_{gem} = 17.3, $J_{4b,5}$ = 4.6, H(4b)). ^{13}C NMR (δ, ppm): 166.1 (CO), 156.7 (C-3), 134.1, 130.5, 128.7, 127.2 (Ph), 129.4 (=CH), 115.6 ($=CH_2$), 58.5 (C-5), 40.5 (C-4), 40.2 (CH_2Cl), 33.0 (CH_2). MS, *m/z* (*I*(%)): 264 (0.2) and 262 (0.6) M^+, 233 (0.9), 173 (19), 105 (100), 77 (78), 51 (40).

4-Benzoyl-6-ethenyl-4,5-diazaspiro[2,4]hept-5-ene (13) was characterized in the mixture with compound **13** by GLC-MS and NMR spectra. Yield ~60 %. ^{1}H NMR (δ, ppm, *J*/Hz): 7.63 and 7.28 (two m, Ph), 6.50 (br dd, J_{trans} = 16.8, J_{cis} = 10.1, =CH), 5.49 (br d, J_{cis} = 10.1,) and 5.38 (br d, J_{trans} = 16.8, $=CH_2$), 3.01 (s, 2 H(4)), 2.23 and 0.67 (two m, 2 CH_2 in cyclopropane). ^{13}C NMR (δ, ppm): 166.9 (CO), 154.2 (C-3), 135.3, 130.0, 129.0, 127.1 (Ph), 135.1 (=CH), 122.4 ($=CH_2$), 44.7 (C-5), 40.6 (C-4), 11.0 $(CH_2)_2$. MS, *m/z* (*I*(%)): 228 (1) and 226 (3) M^+, 173 (27), 105 (100), 77 (86), 51 (44).

Spiro{1-acetyl-6-chloro-3-(2-chloroethyl)-1,4,5,6-tetrahydropyridazin-5,1′-cyclobutane} (6). Two-fold molar excess of dry HCl was bubbled with a flow of Ar into a solution of 0.136 g (0.6 mmol) of *N*-acetylpyrazoline **5** in 2 ml of $CHCl_3$ for 5—7 min at 0—10 °C; then the mixture was flushed with Ar for 5 min. The solvent was evaporated *in vacuo,* and the residue was analyzed by spectral techniques (content of major compound no less than 93 %). 0.14 g of lightly colored viscous liquid was obtained by microdistillation *in vacuo* (0.8 Torr, bath temperature 95—100 °C). IR (ν/cm^{-1}): 1678 (C=O), 1635 (C=N). ^{1}H NMR (δ, ppm, *J*/Hz): 6.48 (d, 1 H, $J_{4,6}$ = 1.6, H(6)), 3.76 (m, 2 H, CH_2Cl), 2.70 (m, 2 H, ($=CCH_2$)), 2.58 (d, 1 H, ^2J = 17.2, H(4)), 2.28 (dd, 1 H, ^2J = 17.2, $J_{4,6}$ = 1.6, H(4)), 2.26 (s, 3 H, Me), 2.12, 1.86, 1.72 (three m, 1 H, 3 H, 2 H, cyclobutane). ^{13}C NMR (δ, ppm): 172.4 (CO), 150.1 (C-3), 68.9 (C-6), 40.5 and 40.2 (C-4 and CH_2Cl), 38.6 (C-5), 34.5 (CH_2 at C-3), 30.9, 30.0 and 13.5 $(CH_2)_3$), 21.5 (Me). MS (direct injection), *m/z* (*I*(%)): 266 (1), 264 (4) and 262 (8) M^+, 229 (5), 227 (16), 222 (11), 220 (20), 187 (11), 185 (48), 96 (29), 43 (100).

1-Acetyl-6-chloro-3-(2-chloroethyl)-5,5-dimethyl-1,4,5,6-tetrahydropyridazine (8) was obtained by the procedure discribed above for **7** by bubbling of HCl into a solution of 0.193 g (0.9 mmol) of 2-acetyl-4-(2-chloroethyl)-6,6-dimethyl-

2,3-diazabicyclo[3.1.0]hex-3-ene **(7)** (Ref. 2). After microdistillation *in vacuo* (0.8 Torr, bath temperature up to 90 °C) we obtained 0.18 g of almost colorless liquid containing according to ^{1}H NMR spectra up to 12 % of unidentified impurities together with tetrahydropyridazine **8**. IR (v/cm^{-1}): 1685 (C=O), 1635 (C=N). ^{1}H NMR (δ, ppm, J/Hz): 6.24 (d, 1 H, $J_{4,6}$ = 1.7, H(6)), 3.81 (m, 2 H, CH_2Cl), 2.76 (m, 2 H, (=CCH_2)), 2.47 (d, 1 H, ^2J = 17.4, H(4)), 2.31 (s, 3 H, COMe), 1.91 (dd, 1 H, ^2J = 17.4, $J_{4,6}$ = 1.7, H(4)), 1.20 и 0.92 (two s, 2×3 H, 2 Me). ^{13}C NMR (δ, ppm): 172.3 (CO), 150.2 (C(3)), 70.2 (C(6)), 40.5 and 39.9 (C(4) and CH_2Cl), 36.2 (CH_2 at C(3)), 32.4 (C(5)), 26.6, 25.1 (2 Me), 21.5 (COMe).

1-Acetyl-3-(2-chloroethyl)-5,5-dimethyl-6-hydroxy-1,4,5,6-tetrahydropyridazine (9). After chromatography on silica gel (ether—hexane, 2.5 : 1, R_f 0.3) 0.085 g of hydroxyderivative **9** as slightly colored liquid was isolated from 0.11 g of the reaction mixture obtained in the preceding experiment. Yield ~85 %. IR (v/cm^{-1}): 3365 (O—H), 1663 (C=O), 1635 (C=N). ^{1}H NMR (δ, ppm, J/Hz): 5.47 (d.d, 1 H, J = 3.5, $J_{4,6}$ = 1.0, H(6)), 4.22 (d, 1 H, J = 3.5, OH (disappears after shaking with D_2O)), 3.77 (m, 2 H, CH_2Cl), 2.69 (m, 2 H, (=CCH_2)), 2.30 (d, 1 H, ^2J = 17.0, H(4)), 2.27 (s, 3 H, COMe), 1.76 (dd, 1 H, ^2J = 17.4, $J_{4,6}$ = 1.0, H(4)), 1.10 and 0.74 (two s, 2×3 H, 2 Me). ^{13}C NMR (δ, ppm): 173.4 (CO), 149.1 (C-3), 75.6 (C-6), 40.2 and 39.7 (C-4 and CH_2Cl), 34.7 (CH_2 at C-3), 29.8 (C-5), 24.5, 24.0 (2 Me), 21.1 (COMe). MS, m/z (I(%)): 234 (4) and 232 (14) M^+, 215 (2), 197 (5), 175 (12), 173 (15), 127 (16), 105 (34), 43 (100). Found (%): C, 51.64; H, 7.52; N, 11.83; Cl, 15.04. $C_{10}H_{17}ClN_2O_2$. Calculated (%): C, 51.61; H, 7.36; N, 12.04; Cl, 15.23.

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