4-DIMETHYLAMINOMETHYLENE-2-PYRAZINYL-5(4*H*)-OXAZOLONE AS A SYNTHON FOR THE SYNTHESIS OF VARIOUS PYRAZOLES AND FUSED PYRIMIDINES[#]

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Abstract – 4-Dimethylaminomethylene-2-pyrazinyl-5(4*H*)-oxazolone (**2**) has been synthesized and transformed into various 1,4-disubstituted 1,2-dihydro-5*H*-pyrazol-5-ones (**6**) and fused pyrimidinones (**8**) by the reaction with selected hydrazines (**5**) and aminoheterocycles (**7**). Products (**6**) and (**8**) have also been prepared from methyl 3-dimethylamino-2-(pyrazinylcarbonylamino)propenoate (**3**) and nitrogen-containing nucleophiles (**5**) or (**7**).

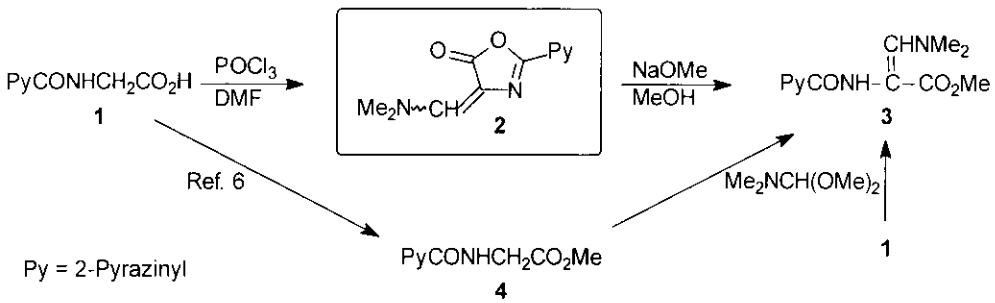
5(4*H*)-Oxazolones have been often used as starting material for the synthesis of various heterocyclic systems.[1] Among them, 4-ethoxymethylene-2-phenyl-5(4*H*)-oxazolone has found wide use as a synthon for the synthesis of different pyrazolones,[1,2] fused pyrimidinones,[3] 2*H*-pyran-2-ones and fused pyran-2-ones,[4] *etc*. When reacted with appropriate hydrazines it yielded the corresponding hydrazinomethylene-5(4*H*)-oxazolones and after heating also the corresponding pyrazolone derivatives or their tautomers.[2] Similarly, different aminoheterocycles yielded 4-arylaminomethylene-2-phenyl-5(4*H*)-oxazolones and fused pyrimidinones of various types.[3] In both cases, the substitution of the ethoxy group by the nitrogen-containing nucleophiles was the first reaction step. In contrast, the dimethylamino group of 4-dimethylaminomethylene-2-phenyl-5(4*H*)-oxazolone[5] is much more stable towards substitution by nucleophiles since the reaction with potassium methoxide yielded only the product of ring opening reaction, namely, methyl 2-benzoylamino-3-dimethylaminopropenoate.[5a]

Here we report the synthesis of 4-dimethylaminomethylene-2-pyrazinyl-5(4*H*)-oxazolone (**2**) and its further transformations into various 1,4-disubstituted 1,2-dihydro-5*H*-pyrazol-5-ones (**6**) and fused pyrimidinones (**8**). In comparison with 2-phenyl analogue,[5] the reactivity of the compound (**2**) towards

[#] This paper is a small contribution to the 50th Volume of HETEROCYCLES.

nucleophiles might be influenced by the pyrazinyl residue, especially under acidic conditions, where a protonation of the pyrazine moiety is possible. The synthesis of the oxazolone (**2**) was carried out by the method described for the preparation of 2-phenyl analogue[5] from *N*-(pyrazinylcarbonyl)glycine (**1**),[6] phosphorus oxychloride and *N*,*N*-dimethylformamide (Scheme 1). The compound (**2**) was transformed into its synthetic equivalent, methyl 3-dimethylamino-2-(pyrazinylcarbonylamino)propenoate (**3**), with sodium methoxide in methanolic solution. Compound (**3**) was also prepared by alternative methods from the acid (**1**) or its methyl ester (**4**)[6] and *N*,*N*-dimethylformamide dimethyl acetal, which is known to react with activated methylene compounds.[7]

Scheme 1



We wanted to test the applicability of compounds (**2**) and (**3**) for the preparation of some pyrazole derivatives as well as fused pyrimidines. For the preparation of the pyrazole derivatives (**6**) the reactions of compounds (**2**) and (**3**) with selected hydrazines (**5**) (hydrazine hydrate, phenylhydrazines and heterocyclic hydrazines) were performed (Scheme 2, Table 1). The reactions of oxazolone (**2**) with hydrazines took place in boiling 1-butanol and products (**6a-d**) were synthesized (Method A). Products (**6a-b**) were also prepared from propenoate (**3**) and appropriate hydrazines (hydrazine hydrate and phenylhydrazine) in boiling ethanol. Reactions of compound (**3**) with hydrazines (**5c-d**) were carried out in boiling acetic acid to give products (**6c-d**). For the synthesis of pyrazoles (**6e-g**) from propenoate (**3**) the reactions were performed with hydrazines (**5e-g**) hydrochlorides in butanolic solution (Method B).

These reactions demonstrated that the dimethylamino group in compounds (**2**) and (**3**) could be relatively easily substituted by hydrazines under various conditions.

Scheme 2

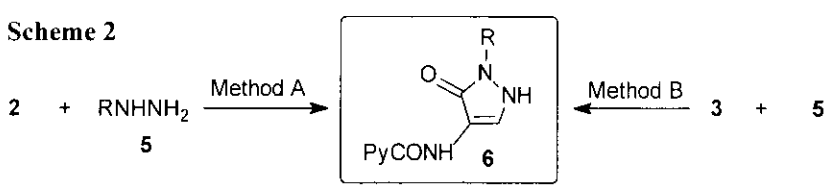
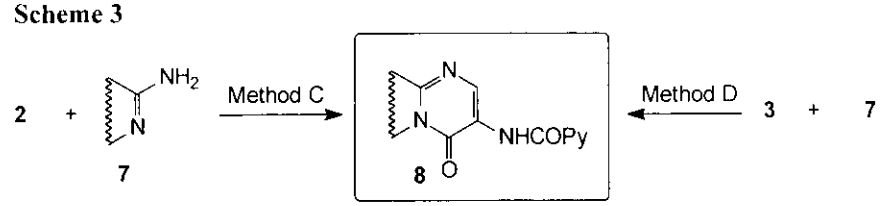


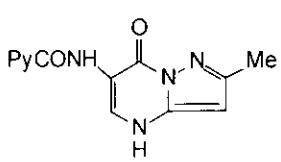
Table 1. The Synthesis of Pyrazolones (**6a-g**):

Starting Hydrazine (5, R=)	Product (6)	Method A (BuOH, Δ, h)	Yield (%)	Method B (solvent, Δ, h)	Yield (%)
H	**6a**	2	83	EtOH, 6	73
Ph	**6b**	4	70	EtOH, 6	14
6-chloropyridazin-3-yl	**6c**	4	58	AcOH, 3	85
6-chloropyrazin-2-yl	**6d**	4	57	AcOH, 4	85
4-nitrophenyl	**6e**			BuOH, 5	77
4-methoxyphenyl	**6f**			BuOH, 4	84
2,4-difluorophenyl	**6g**			BuOH, 8	73

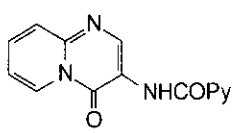
The described results stimulated us to realize similar reactions of synthetic equivalents (**2**) and (**3**) with certain less nucleophilic reagents than hydrazines. For this reason we performed reactions of both derivatives (**2** and **3**) with 1,3-binucleophiles of type (**7**), such as 3-amino-5-methyl-1*H*-pyrazole, 2-aminopyridine, 2-aminobenzothiazole and 2-aminobenzimidazole. Initially we treated oxazolone derivative (**2**) with 1,3-binucleophiles (**7**) in boiling acetic acid or *N*,*N*-dimethylformamide, but the reactions were unsuccessful, since we recovered the starting material. Then, reactions were carried out in boiling dimethyl sulfoxide and we isolated the expected products (**8a-d**) in moderate yields (Scheme 3, Table 2, Method C). On the other hand, the propenoate (**3**) and aminoheterocycles (**7**) reacted in boiling acetic acid to give the corresponding products (**8a-d**) in high yields (Method D).

Scheme 3

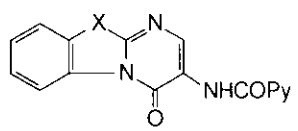




8a



8b



8c: X=S

8d: X=NH

Py = 2-Pyrazinyl

Table 2. The Synthesis of Fused Pyrimidines (**8a-d**):

Starting Amine (7)	Product (8)	Method C (DMSO, Δ, h)	Yield (%)	Method D (AcOH, Δ, h)	Yield (%)
3-amino-5-methyl-1*H*-pyrazole	**8a**	10	35	4	84
2-aminopyridine	**8b**	4	17	4	89
2-aminobenzothiazole	**8c**	10	44	4	40
2-aminobenzimidazole	**8d**	10	61	4	86

In conclusion, we synthesized the oxazolone derivative (**2**) and applied it for the preparation of various types of heterocyclic compounds. It reacts with different nucleophiles either directly in a one-step process, or it can be converted to its synthetic equivalent, propenoate (**3**), that is in some cases even more reactive than oxazolone (**2**) itself, as shown in transformations with 1,3-binucleophiles.

EXPERIMENTAL

Melting points were determined on a Kofler micro hot stage and are uncorrected. ^{1}H NMR spectra were recorded on a Bruker Avance DPX 300, JEOL JNM FX90Q and Varian EM 360 L spectrometers in DMSO-d_6 (if not stated differently) using TMS as an internal standard. MS spectra were obtained with a VG-Analytical AutospecQ spectrometer. Elemental analyses (C, H, N) were performed on a Perkin-Elmer 2400 CHN Analyzer. TLC was carried out on Fluka silica gel TLC-cards.

4-Dimethylaminomethylene-2-pyrazinyl-5(4*H*)-oxazolone (2). To the cooled (ice bath) and stirred mixture of 362 mg (2 mmol) of *N*-(pyrazinylcarbonyl)glycine (**1**) and 880 mg (12 mmol) of *N*,*N*-dimethylformamide $POCl_3$ (920 mg, 6 mmol) was slowly added. After stirring for additional 15 min, the mixture was heated for 6 h at 50-60 °C. Upon evaporation, ice (15 g) was added and the mixture was neutralized with solid $NaHCO_3$ to pH 6. By the extraction with chloroform (3 × 10 mL) product (**2**) was obtained (362 mg, 83%) and crystallized from methanol, mp 206-208 °C. ^{1}H NMR (90 MHz) δ 3.33 (s, 3H, Me), 3.35 (s, 3H, Me), 7.54 (s, 1H, CH), 8.68 (d, J = 2.6 Hz, 1H, 5-H), 8.72 (dd, J = 2.6 and 1.2 Hz, 1H, 6-H), 9.15 (d, J = 1.2 Hz, 1H, 3-H). Anal. Calcd for $C_{10}H_{10}N_4O_2$: C, 55.04; H, 4.62; N, 25.68. Found: C, 54.74; H, 4.68; N, 26.02.

Methyl 3-dimethylamino-2-(pyrazinylcarbonylamino)propenoate (3).

A. A mixture of 218 mg (1 mmol) of the oxazolone derivative (**2**) and 3 mL of a methanolic solution of NaOMe, prepared from 23 mg (1 mmol) of sodium and 3 mL of methanol, was shortly heated at 60 °C,

(until a clear solution was obtained), then it was stirred at rt for 1 h and, upon cooling and filtering off, product (**3**) was obtained (183 mg, 73%) and crystallized from the mixture MeOH/AcOEt, mp 165-166 °C. ^{1}H NMR (300 MHz) δ 2.95 (s, 6H, two Me), 3.52 (s, 3H, Me), 7.38 (s, 1H, CH), 8.76 (dd, J = 2.6 and 1.5 Hz, 1H, 6-H), 8.88 (d, J = 2.6 and 1.2 Hz, 1H, 5-H), 9.19 (d, J = 1.5 Hz, 1H, 3-H), 9.45 (br s, 1H, NH). Anal. Calcd for $C_{11}H_{14}N_4O_3$: C, 52.79; H, 5.64; N, 22.39. Found: C, 53.09; H, 5.64; N, 22.28.

B. A mixture of 181 mg (1 mmol) of the glycine derivative (**1**) and 550 mg (90%, 4.2 mmol) of *N,N*-dimethylformamide dimethyl acetal (DMFDMA) in 7 mL of toluene was refluxed for 4.5 h, than an additional amount of DMFDMA (280 mg, 2.1 mmol) was added and the mixture was refluxed for next 4 h. Upon cooling, the separated product (**3**) was filtered off (155 mg, 62%).

C. A mixture of the ester (**4**) (3.79 g, 19.4 mmol) and DMFDMA (5.80 mL of 90%, 39 mmol) in 19.5 mL of acetonitrile was refluxed for 12 h. Upon evaporation and addition of acetonitrile (15 mL), the separated product (**3**) was filtered off (3.14 g, 65%).

General procedure for the preparation of pyrazolones (6).

Method A: A mixture of the oxazolone derivative (**2**) (109 mg, 0.5 mmol) and a hydrazino derivative (**5**) *(0.5 mmol; in the case of* **5a** *0.68 mmol of hydrazine hydrate was used) in 2 mL of 1-butanol was refluxed* for 4 h (2 h in the case of **6a**). After evaporation 1 mL of ethanol was added and, upon cooling, the separated product was filtered off and washed with a small amount of ethanol.

Method B: A mixture of the propenoate (**3**) (500 mg, 2 mmol) and a hydrazino derivative (**5**) (2 mmol) in ethanol (4-5 mL, for products **6a-b**) or acetic acid (2-4 mL, for products **6c-d**) or 1-butanol (4 mL for products **6e-g**), where we started from hidrazinium hydrochlorides **5e-g**) was refluxed for 3-8 h. After evaporation a small amount of ethanol (0.5-1 mL) was added and, upon cooling, the separated product was filtered off and washed with ethanol (for products **6a-d**). To obtain pyrazolones (**6e-g**), the butanolic solution was cooled and the separated product was filtered off and washed with 1 mL of 1-butanol. Yields of TLC-pure compounds are given in Table 1.

The following products were obtained by these methods:

***N*-(1,2-Dihydro-3-oxo-3*H*-pyrazol-4-yl)pyrazine-2-carboxamide (6a):** mp 302-305 °C (decomp, DMF/MeOH); ^{1}H NMR (90 MHz) δ 7.91 (s, 1H, 5'-H), 8.79 (dd, J = 1.5 and 2.4 Hz, 1H, 6-H), 8.93 (d, J = 2.4 Hz, 1H, 5-H), 9.27 (d, J = 1.5 Hz, 1H, 3-H), 10.12 (br s, 1H, NH). Anal. Calcd for $C_8H_7N_5O_2$: C, 46.83; H, 3.44; N, 34.13. Found: C, 46.95; H, 3.56; N, 33.80.

***N*-(1,2-Dihydro-1-phenyl-5-oxo-5*H*-pyrazol-4-yl)pyrazine-2-carboxamide (6b):** mp 235-239 °C (decomp, DMF/EtOH); ^{1}H NMR (300 MHz) δ 7.30 (m, 1H, Ph), 7.49 (m, 2H, Ph), 7.77 (m, 2H, Ph), 7.95

(br s, 1H, 3'-H), 8.81 (dd, J = 1.5 and 2.6 Hz, 1H, 6-H), 8.94 (d, J = 2.6 Hz, 1H, 5-H), 9.29 (d, J = 1.5 Hz, 1H, 3-H), 10.48 (br s, 1H, NH), 11.68 (br s, 1H, NH); MS (m/z, %) 281 (M+, 100). Anal. Calcd for $C_{14}H_{11}N_5O_2$: C, 59.78; H, 3.94; N, 24.90. Found: C, 59.78; H, 4.05; N, 24.84.

***N*-[1-(6-Chloropyridazin-3-yl)-1,2-dihydro-5-oxo-5*H*-pyrazol-4-yl]pyrazine-2-carboxamide (6c):** mp above 250 °C (decomp, DMF); ^{1}H NMR (60 MHz, CF_3CO_2D) δ 7.75 (d, J = 10 Hz, 1H, 5''-H), 8.30 (s, 1H, 3'-H), 8.70 (d, J = 10 Hz, 1H, 4''-H), 8.80 (d, J = 3 Hz, 1H, 5-H), 9.10 (d, J = 3.0 Hz, 1H, 6-H), 9.32 (br s, 1H, 3-H); MS (m/z, %) 317 (M^+, 100). Anal. Calcd for $C_{12}H_8N_7O_2Cl$: C, 45.37; H, 2.54; N, 30.86. Found: C, 45.52; H, 2.49; N, 30.64.

***N*-[1-(6-Chloropyrazin-2-yl)-1,2-dihydro-5-oxo-5*H*-pyrazol-4-yl]pyrazine-2-carboxamide (6d):** mp 276-279 °C (decomp, DMF); ^{1}H NMR (60 MHz, CF_3CO_2D) δ 8.33 (s, 1H) and 8.37 (s, 1H, 3'-H, 5''-H), 8.78 (d, J = 3 Hz, 1H, 5-H), 9.11 (m, 1H, 6-H), 9.32 (s, 1H, 3-H), 9.40 (s, 1H, 3''-H); MS (m/z, %) 317 (M^+, 100). Anal. Calcd for $C_{12}H_8N_7O_2Cl$: C, 45.37; H, 2.54; N, 30.86. Found: C, 45.40; H, 2.43; N, 30.62.

***N*-[1,2-Dihydro-1-(4-nitrophenyl)-5-oxo-5*H*-pyrazol-4-yl]pyrazine-2-carboxamide (6e):** mp 269-272 °C (DMF/MeOH); ^{1}H NMR (300 MHz) δ 8.08 (br s, 1H, 3'-H), 8.13 (m, 2H, Ph), 8.37 (m, 2H, Ph), 8.82 (dd, J = 2.6 and 1.5 Hz, 1H, 6-H), 8.95 (d, J = 2.6 Hz, 1H, 5-H), 9.29 (d, J = 1.5 Hz, 1H, 3-H), 10.36 (br s, 1H, NH), 12.24 (br s, 1H, NH); MS (m/z, %) 326 (M^+, 100). Anal. Calcd for $C_{14}H_{10}N_6O_4$: C, 51.54; H, 3.09; N, 25.76. Found: C, 51.44; H, 3.16; N, 25.61.

***N*-[1,2-Dihydro-1-(4-methoxyphenyl)-5-oxo-5*H*-pyrazol-4-yl]pyrazine-2-carboxamide (6f):** mp 255-257 °C (DMF/MeOH); ^{1}H NMR (300 MHz) δ 3.80 (s, 3H, Me), 7.04 (m, 2H, Ph), 7.62 (m, 2H, Ph), 7.88 (br s, 1H, 3'-H), 8.81 (dd, J = 2.6 and 1.5 Hz, 1H, 6-H), 8.94 (d, J = 2.6 Hz, 1H, 5-H), 9.28 (d, J = 1.5 Hz, 1H, 3-H), 10.39 (br s, 1H, NH), 11.53 (br s, 1H, NH); MS (m/z, %) 311 (M^+, 100). Anal. Calcd for $C_{15}H_{13}N_5O_3$: C, 57.88; H, 4.21; N, 22.50. Found: C, 57.84; H, 4.36; N, 22.45.

***N*-[1,2-Dihydro-1-(2,4-difluorophenyl)-5-oxo-5*H*-pyrazol-4-yl]pyrazine-2-carboxamide (6g):** mp 193-195 °C (DMF/MeOH); ^{1}H NMR (300 MHz) δ 7.25 (m, 1H, Ph), 7.56 (m, 2H, Ph), 7.95 (br s, 1H, 3'-H), 8.81 (dd, J = 2.3 and 1.5 Hz, 1H, 6-H), 8.94 (d, J = 2.3 Hz, 1H, 5-H), 9.28 (d, J = 1.5 Hz, 1H, 3-H), 10.42 (br s, 1H, NH), 11.54 (br s, 1H, NH); MS (m/z, %) 317 (M^+, 100). Anal. Calcd for $C_{14}H_9N_5O_2F_2$: C, 53.00; H, 2.86; N, 22.07. Found: C, 52.87; H, 2.92; N, 22.25.

General procedure for the preparation of fused pyrimidinones (8).

Method C: A mixture of the oxazolone derivative (**2**) (85 mg, 0.39 mmol) and an aminoheterocycle (**7**) (73 mg, 0.75 mmol of **7a**; 48 mg, 0.50 mmol of **7b**; 154 mg, 1.02 mmol of **7c**; 100 mg, 0.75 mmol of **7d**) in 2 mL of DMSO was refluxed for 10 h (4 h for **8b**). Upon cooling, the separated product was filtered off and washed with a small amount of ethanol.

Method D: A mixture of the propenoate (**3**) (250 mg, 1 mmol) and an aminoheterocycle (**7**) (1 mmol) in 2 mL of acetic acid was refluxed for 4 h. After evaporation a small amount of ethanol (1 mL) was added and, upon cooling, the separated product was filtered off and washed with ethanol. Yields of TLC-pure compounds are given in Table 2.

The following products were obtained by these methods:

***N*-(2-Methyl-4,7-dihydro-7-oxopyrazolo[1,5-*a*]pyrimidin-6-yl)pyrazine-2-carboxamide (8a):** mp above 360 °C (DMF); ^{1}H NMR (60 MHz, CF_3CO_2D) δ 2.33 (s, 3H, Me), 6.28 (s, 1H, 3'-H), 8.84 (d, J = 2.5 Hz, 1H, 5-H), 9.02 (s, 1H, 5'-H), 9.18 (m, 1H, 6-H), 9.35 (s, 1H, 3-H); MS (m/z, %) 270 (M^+, 100). Anal. Calcd for $C_{12}H_{10}N_6O_2$: C, 53.33; H, 3.73; N, 31.10. Found: C, 53.25; H, 3.66; N, 30.73.

***N*-(4-Oxo-4*H*-pyrido[1,2-*a*]pyrimidin-3-yl)pyrazine-2-carboxamide (8b):** mp above 270-271 °C (DMF); ^{1}H NMR (60 MHz, CF_3CO_2D) δ 7.25-8.33 (m, 3H, 7'-H, 8'-H, 9'-H), 8.77-9.37 (m, 5H, 2'-H, 6'-H, 3-H, 5-H, 6-H); MS (m/z, %) 267 (M^+, 100). Anal. Calcd for $C_{13}H_9N_5O_2$: C, 58.43; H, 3.39; N, 26.21. Found: C, 58.28; H, 3.28; N, 25.94.

***N*-(4-Oxo-4*H*-pyrimido[2,1-*b*]benzothiazol-3-yl)pyrazine-2-carboxamide (8c):** mp 285-286.5 °C (DMF); ^{1}H NMR (60 MHz, CF_3CO_2D) δ 7.38-7.68 (m, 3H, 7'-H, 8'-H, 9'-H), 8.73-8.93 (m, 2H, 5-H, 6'-H), 9.10-9.22 (m, 2H, 6-H, 2'-H), 9.33 (s, 1H, 3-H); MS (m/z, %) 323 (M^+, 100). Anal. Calcd for $C_{15}H_9N_5O_2S$: C, 55.72; H, 2.81; N, 21.66. Found: C, 55.59; H, 2.56; N, 21.46.

***N*-(4-Oxo-4,10-dihydropyrimido[1,2-*a*]benzimidazol-3-yl)pyrazine-2-carboxamide (8d):** mp above 350 °C (DMF); ^{1}H NMR (60 MHz, CF_3CO_2D) δ 7.22-7.52 (m, 3H, 7'-H, 8'-H, 9'-H), 8.30 (m, 1H, 6'-H), 8.87 (d, J = 2.5 Hz, 5-H), 9.02 (s, 1H, 2'-H), 9.23 (m, 1H, 6-H), 9.42 (s, 1H, 3-H); MS (m/z, %) 306 (M^+, 100). Anal. Calcd for $C_{15}H_{10}N_6O_2$: C, 58.82; H, 3.29; N, 27.44. Found: C, 58.78; H, 3.04; N, 27.40.

ACKNOWLEDGMENT

The financial support from the Ministry of Science and Technology of Slovenia is gratefully acknowledged. We also thank Dr. B. Kralj and Dr. D. Žigon (Mass Spectrometry Center, Jožef Stefan Institute) for mass measurements.

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Received, 2nd July, 1998