Turk J Chem
28 (2004) , 149 – 156.

Synthesis and Some Reactions of 4-Benzoyl-5-Phenyl-1-(2,4,6-Trichloro Phenyl)-1*H*-Pyrazole-3-Carboxylic Acid

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Received 19.06.2003

Furandione **1** was reacted with 2,4,6-trichlorophenylhydrazine to give the 1*H*-pyrazole-3-carboxylic acid **2** together with a new pyridazinone derivative **3**. Then **2** was converted into the corresponding ester **5** or amide **6** derivatives, respectively, via reactions of its acid chloride **4** with various alcohols or *N*-nucleophiles. Nitrile derivative **7** was obtained by dehydration of **6a** in a mixture of $SOCl_2$ and DMF. Condensation reactions of **2** with some hydrazines led to the formation of pyrazolo [3,4−*d*] pyridazinone **8** derivatives.

Key Words: Cyclic oxalyl compounds, Pyrazole, Pyridazine, Pyrazolo [3,4-d] Pyridazinone.

Introduction

It has long been known that pyrazole and fused pyrazole compounds possess numerous chemical, biological, medicinal and agricultural applications because of their versatile biological activities appearing as antimicrobial[1], antiviral[2], antitumor[3], anti-inflammatory[4], antihistaminic[5], pesticidal[6], antifungal[7] and antipyretic[8] agents. On the other hand, cyclic oxalyl compounds of type **1** have been used successfully as starting materials to synthesize various 1,4,5-trisubstitute pyrazol-3-carboxylic acids and their derivatives via reactions of them with various hydrazines or hydrazones for some 2 decades[9−11]. Here, we report the chemical behavior of furandione **1** toward a different arylhydrazine compound, the ortho- positions of which are occupied by chlorine atoms, and present the reactions of cyclic oxalyl compounds with various hydrazines or hydrazones to prepare new pyrazole and pyridazine derivatives.

Experimental

Solvents were dried by refluxing with the appropriate drying agents (metallic sodium for ether, $CaCl_2$ or $NaSO_4$ for benzene, toluene etc.) and distilled. Likewise, 2,4,6-trichlorophenylhydrazine was also crystallized from methanol for purification before use. Melting points were determined on an electrothermal Gallenkamp apparatus and are uncorrected. Microanalyses were performed on a Carlo Erba elemental analyzer Model

1108. The IR spectra were obtained as potassium bromide pellets using a Mattson 1000 FTIR spectrometer. The ^{1}H and ^{13}C–NMR spectra were recorded on Varian XL-200 (200 MHz) and (50 MHz) spectrometers, respectively, using TMS as an internal standard. The mass spectra of **2** and **3** were measured on a Varian Mat III at 80 e.V. All experiments were followed by TLC using a DC Alufolien Kieselgel 60 F 254 Merck and Camag TLC lamp (254/366 nm).

4-Benzoyl-5-Phenyl-1-(2,4,6-Trichlorophenyl)-1*H*-Pyrazole-3-Carboxylic Acid 2

Furandione (0.278 g, 1 mmol) and 2,4,6-trichlorophenylhydrazine (0.211 g, 1 mmol) were refluxed in dry benzene (30 mL) for about 5 h. After cooling, the precipitate was filtered off and washed with cold benzene to give a crude solid that was recrystallized from carbon-tetrachloride. The yield was 0.165 g (35%), mp 198 °C (Decomposition); IR: 3400-2750 cm^{-1} (br, OH, COOH), 3130-3050 cm^{-1} (Ar-H), 1727 cm^{-1}(C=O, COOH), 1676 cm^{-1} (C=O, benzoyl); ^{1}H-NMR (DMSO-d$_6$): δ = 8.1-7.3 ppm (Ar-H); ^{13}C-NMR (DMSO-d$_6$): δ = 192.98 (C=O, benzoyl), 160.05 (C=O, COOH), 151.83 (C-3), 138.97 (C-5), 137.51, 137.02, 136.64, 135.94, 132.41, 130.93, 130.69, 130.50, 130.27, 130.21, 128.64, 128.53, 124.15 ppm (C–4); Mass (80 e.V.): m/e = 471.0, 435.2, 427.1, 393.1, 357.1, 349, 247.0, 207.9, 178.9, 144.0.

Anal. Calcd. for $C_{23}H_{13}Cl_3N_2O_3$: C, 58.56; H, 2.78; N, 5.94; Cl, 22.55. Found: C, 58.69; H, 2.77; N, 5.96; Cl, 22.40.

5-Benzoyl-4-Hydroxy-6-Phenyl-2-(2,4,6-Trichloro-Phenyl)-2H-Pyridazin-3-one 3

After the solvent of the filtrate of **2** was removed by evaporation, the oily residue was treated with ether and the crude product formed was recrystallized from isoamyl alcohol to give 0.142 g (30%) of **3**, mp 243 °C; IR: 3313 cm^{-1} (OH), 3080 cm^{-1} (Ar-H), 1687 cm^{-1}(C=O, benzoyl), 1635 cm^{-1} (C=O); ^{1}H-NMR (DMSO-d$_6$): δ = 10.41 (b, H, OH), 7.7-6.7 ppm (Ar-H); ^{13}C-NMR (DMSO-d$_6$): δ = 196.00 (C=O, benzoyl), 158.99 (C=O), 155.73 (C-4), 141.20 (C-6), 139.82 (C-5), 138.88, 138.02, 136.43, 136.01, 132.96, 132.22, 130.77, 130.62, 130.54, 130.31, 130.24, 128.05 ppm; Mass (80 e.V.): m/e = 471.1, 429.1, 401.1, 375.1, 339.1, 303.0, 263.0, 271.1, 255.0, 239.0, 224.0, 210.0, 189.0.

Anal. Calcd. for $C_{23}H_{13}Cl_3N_2O_3$: C, 58.56; H, 2.78; N, 5.94; Cl, 22.55. Found: C, 58.42; H, 2.79; N, 5.42; Cl, 22.59.

Methyl 4-Benzoyl-5-Phenyl-1-(2,4,6-Trichlorophenyl)-1*H*-Pyrazole-3-Carboxylate 5a

General Procedure

Compound **2** (0.472 g, 1 mmol) and thionylchloride (1 mL, 13.8 mmol) were refluxed on a steam bath for 5 h. After cooling, the crude precipitate was triturated with dry diethyl ether. The resultant crude acyl chloride **4** was directly reacted as below. The acid chloride **4** (0.49 g) and a large excess of methanol were refluxed together with a catalytic amount of pyridine for 2 h. After cooling, the solution was acidified by adding diluted hydrochloric acid (12%) to give a crude solid that recrystallized from the same alcohol. The yield was 0.316 g (65%), mp 169 °C; IR: 3082 cm^{-1} (Ar-H), 2949 cm^{-1} (R-H), 1732 cm^{-1} (C=O, ester), 1668 cm^{-1} (C=O, benzoyl); ^{1}H-NMR ($CDCl_3$-d$_6$): δ = 7.94-7.29 (m, Ar-H), 3.50 ppm (s, OCH_3); ^{13}C-NMR

(DMSO-d_6): δ = 193.53 (C=O, benzoyl), 159.99 (C=O, ester), 153.57 (C-3), 139.82 (C-5), 138.45, 137.07, 136.91, 135.71, 132.90, 131.27, 130.99, 130.77, 130.64, 130.56, 129.68, 125.01 (C–4), 54.29 ppm (OCH_3).

Anal. Calcd. for $C_{24}H_{15}Cl_3N_2O_3$: C, 59.34; H, 3.11; N, 5.77; Cl, 21.90. Found: C, 59.41; H, 3.13; N, 5.75; Cl, 21.84.

n-Propyl 4-Benzoyl-5-Phenyl-1-(2,4,6-Trichlorophenyl)-1*H*-Pyrazole-3-Carboxylate 5b

The yield was 60% (0.308 g); mp 160 °C; IR: 3074 cm^{-1} (Ar-H), 2970, 2927, 2854 cm^{-1} (R-H), 1720 cm^{-1} (C=O, ester), 1678 cm^{-1} (C=O, benzoyl); ^{1}H-NMR ($CDCl_3$-d_6): δ = 7.95-7.90 (m, 2H, Ar-H), 7.53 (s, 2H, Ar-H), 7.68-7.27 (m, 8H, Ar-H), 3.90 (t, 2H, OCH_2), 1.21 (sextet, 2H, CH_2), 0.60 ppm (t, 3H, CH_3).

Anal. Calcd. for $C_{26}H_{19}Cl_3N_2O_3$: C, 60.78; H, 3.73; N, 5.45; Cl, 20.70. Found: C, 60.90; H, 3.75; N, 5.47; Cl, 20.64.

Isobutyl 4-Benzoyl-5-Phenyl-1-(2,4,6-Trichlorophenyl)-1*H*-Pyrazole-3-Carboxylate 5c

The yield was 70% (0.37 g); mp 144 °C; IR: 3076 cm^{-1} (Ar-H), 2966, 2935, 2885 cm^{-1} (R-H), 1718 cm^{-1} (C=O, ester), 1681 cm^{-1} (C=O, benzoyl).

Anal. Calcd. for $C_{27}H_{21}Cl_3N_2O_3$: C, 61.44; H, 4.01; N, 5.31; Cl, 20.15. Found: C, 61.25; H, 4.04; N, 5.33; Cl, 20.20.

4-Benzoyl-5-Phenyl-1-(2,4,6-Trichlorophenyl)-1*H*-Pyrazole-3-Carboxamide 6a

To the solution of compound **4** (0.49 g, 1 mmol) in carbontetrachloride (20 mL) in an ice-bath was added 0.5 mL of concentrated aqueous ammonia (25%). Then the reaction mixture was left stirring for 30 min. The precipitate formed was filtered and recrystallized from methanol to give 0.165 g (35%) of **6a**, mp 243 °C; IR: 3398 cm^{-1} (NH_2), 3062 cm^{-1} (Ar-H), 1683 cm^{-1} (C=O, benzoyl), 1649 cm^{-1} (C=O, amide).

Anal. Calcd. for $C_{23}H_{14}Cl_3N_3O_2$: C, 58.68; H, 3.00; N, 8.93; Cl, 22.59. Found: C, 58.57; H, 3.03; N, 8.89; Cl, 22.65.

N-Ethyl 4-Benzoyl-5-Phenyl-1-(2,4,6-Trichlorophenyl)-1*H*-Pyrazole-3-Carboxamide 6b

General Procedure

Acid chloride **4** (0.49 g, 1 mmol) and ethyl amine (0.1 g, 2 mmol) were refluxed in xylene for about 3 h. After evaporation, the oily residue was treated with dry ether and the crude product formed was crystallized from ethyl alcohol to give 0.225 g (45%) of **6b**; mp 233 °C; IR: 3286 cm^{-1} (NH-Et), 3082 cm^{-1} (Ar-H), 2981, 2933 cm^{-1} (R-H), 1670 cm^{-1} (C=O, benzoyl), 1637 cm^{-1} (C=O, amide);^{1}H-NMR ($CDCl_3$-d_6): δ = 8.54 (br, 1H, NH), 7.68-7.63 (m, 2H, Ar-H), 7.50 (s, 2H, Ar-H), 7.48-7.12 (m, 8H, Ar-H), 3.35 (dt, 2H, CH_2), 1.19 ppm (t, 3H, CH_3).

Anal. Calcd. for $C_{25}H_{18}Cl_3N_3O_2$: C, 60.20; H, 3.64; N, 8.42; Cl, 21.32. Found: C, 60.09; H, 3.67; N, 8.35; Cl, 21.37.

N,N-Diethyl 4-Benzoyl-5-Phenyl-1-(2,4,6-Trichlorophenyl)-1*H*-Pyrazole-3-Carboxamide 6c

Compound **6c** was prepared according to the general procedure above with a reflux time of 3 h resulting in 50% yield (0.263 g); mp 198 °C (isopropyl alcohol); IR: 3031 cm^{-1} (Ar-H), 2983, 2939, 2850, 2830 cm^{-1} (R-H), 1672 cm^{-1} (C=O, benzoyl), 1635 cm^{-1} (C=O, amide).

Anal. Calcd. for $C_{27}H_{22}Cl_3N_3O_2$: C, 61.55; H, 4.21; N, 7.98; Cl, 20.19. Found: C, 61.63; H, 4.18; N, 7.95; Cl, 20.24.

N-[4-Benzoyl-5-Phenyl-1-(2,4,6-Trichlorophenyl)-1*H*-Pyrazole-3-Carbonyl]-*N'*-Methyl Urea 6d

Compound **6d** was prepared according to the general procedure above with a reflux time of 3 h resulting in 45% yield (0.238 g); mp 230 °C (Methanol); IR: 3321 cm^{-1} (NH), 3076 cm^{-1} (Ar-H), 2935 cm^{-1} (R-H), 1741, 1674, 1643 cm^{-1} (C=O); ^{1}H-NMR (DMSO-d_6): δ = 8.53 (b, NH), 7.67-7.12 (m, Ar-H), 2.90 ppm (d, CH_3).

Anal. Calcd. for $C_{25}H_{17}Cl_3N_4O_3$: C, 56.89; H, 3.25; N, 10.62; Cl, 20.15. Found: C, 57.12; H, 3.23; N, 10.67; Cl, 20.19.

N-[4-Benzoyl-5-Phenyl-1-(2,4,6-Trichlorophenyl)-1*H*-Pyrazole-3-Carbonyl]-*N'*-Phenyl Urea 6e

Method A. From Acid Chloride

Compound **6e** was prepared according to the general procedure above with a reflux time of 3 h resulting in 35% yield (0.206 g); mp 244 °C (n-butanol); IR: 3281, 3267 cm^{-1} (NH), 3086 cm^{-1} (Ar-H), 1736, 1682, 1620 cm^{-1} (C=O); ^{13}C-NMR ($CDCl_3$-d_6): δ =196.95 (C=O, benzoyl), 157.30 (C=O), 156.39 (C=O), 142.29 (C-3), 139.41 (C-5), 139.16, 138.00, 137.71, 136.49, 135.85, 133.22, 132.28, 130.97, 130.70, 130.52, 130.22, 128.63, 126.95, 122.49, 119.73 ppm (C–4).

Anal. Calcd. for $C_{30}H_{19}Cl_3N_4O_3$: C, 61.09; H, 3.25; N, 9.50; Cl, 18.03. Found: C, 61.17; H, 3.27; N, 9.46; Cl, 18.07.

Method B. From Acid Amide

The acid amide **6a** (0.471 g, 1 mmol) and phenylisocyanate (0.2 mL, 1.8 mmol) were refluxed in xylene for 3 h. Then the solvent was evaporated and the residue was recrystallized from n-butanol to give 0.354 g (60%) of **6e**, identical in mp and IR spectrum with that obtained in method A.

4-Benzoyl-5-Phenyl-1-(2,4,6-Trichlorophenyl)-1*H*-Pyrazole-3-Carbonitrile 7

A cold solution of the acid amide **6a** (0.471 g, 1 mmol) in a mixture of DMF (0.7 mL) and $SOCl_2$ (0.15 mL) was stirred at 0-5 °C for 2 h, and the solution was left stirring overnight. Then the reaction mixture was poured over crushed ice and the solid formed was isolated by filtration, washed with water and recrystallized from methanol to give 0.249 g (55%) of **7** mp 172 °C; IR: 3081 cm^{-1} (Ar-H), 2240 cm^{-1} (CN), 1649 cm^{-1} (C=O, benzoyl); ^{13}C-NMR ($CDCl_3$-d_6): δ = 190.28 (C=O, benzoyl), 155.59 (C-3), 140.26 (C-5), 138.43 (C-4), 137.40, 136.22, 134.07, 131.85, 131.50, 131.20, 131.08, 130.74, 130.57, 128.13, 121.79 (C-3a), 110.13 ppm (C≡N).

Anal. Calcd. for $C_{23}H_{12}Cl_3N_3O$: C, 61.02; H, 2.67; N, 9.28; Cl, 23.49. Found: C, 60.59; H, 2.64; N, 9.26; Cl, 23.53.

3,4,6-Triphenyl-2-(2,4,6-Trichlorophenyl)-2,6-Dihydro-Pyrazolo [3,4-*d*] Pyridazin-7-one 8a

General Procedure

A milliequimolar mixture of **2** (0.47 g) and phenylhydrazine (0.1 mL) was refluxed in xylene for 3 h. The solvent was evaporated, then the oily residue was treated with ether and the crude product formed was crystallized from n-propanol. Compound **8a** was obtained in 65% yield (0.353 g); mp 242 °C; IR: 3060, 3028 cm^{-1} (Ar-H), 1674 cm^{-1} (C=O); ^{13}C-NMR ($CDCl_3$-d_6): δ = 155.72 (C=O), 151.66 (C-7a), 146.07 (C-4), 142.89 (C-3), 138.77, 137.31, 136.26, 132.72, 131.69, 131.12, 130.95, 130.69, 130.52, 130.19, 130.00, 129.86, 129.70, 127.87, 119.10, 114.61 ppm (C-3a).

Anal. Calcd. for $C_{29}H_{17}Cl_3N_4O$: C, 64.05; H, 3.15; N, 10.30; Cl, 19.56. Found: C, 64.71; H, 3.11; N, 10.35; Cl, 19.59.

6-(4-Methoxyphenyl)-3,4-Diphenyl-2-(2,4,6-Trichlorophenyl)-2,6-Dihydro-Pyrazolo [3,4-*d*] Pyridazin-7-one 8b

Compound **8b** was prepared according to the general procedure above with a reflux time of 3 h resulting in 70% yield (0.402 g); mp 232 °C (ethanol); IR: 3068 cm^{-1} (Ar-H), 2929 cm^{-1} (R-H), 1680 cm^{-1} (C=O); ^{13}C-NMR ($CDCl_3$-d_6): δ = 161.13 (C=O), 154.01 (C-4', Ph-OCH_3), 151.61 (C-7a), 145.81 (C-4), 138.70 (C-3), 137.31, 136.31, 135.96, 132.76, 131.68, 131.11, 130.68, 130.49, 129.96, 129.82, 129.03, 121.40, 119.05, 116.12, 115.94 (C-3a), 57.56 ppm (OCH_3).

Anal. Calcd. for $C_{30}H_{19}Cl_3N_4O_2$: C, 62.79; H, 3.34; N, 9.76; Cl, 18.53. Found: C, 62.51; H, 3.36; N, 9.81; Cl, 18.35.

3,4-Diphenyl-2-(2,4,6-Trichlorophenyl)-2,6-Dihydro-Pyrazolo [3,4-*d*] Pyridazin-7-one 8c

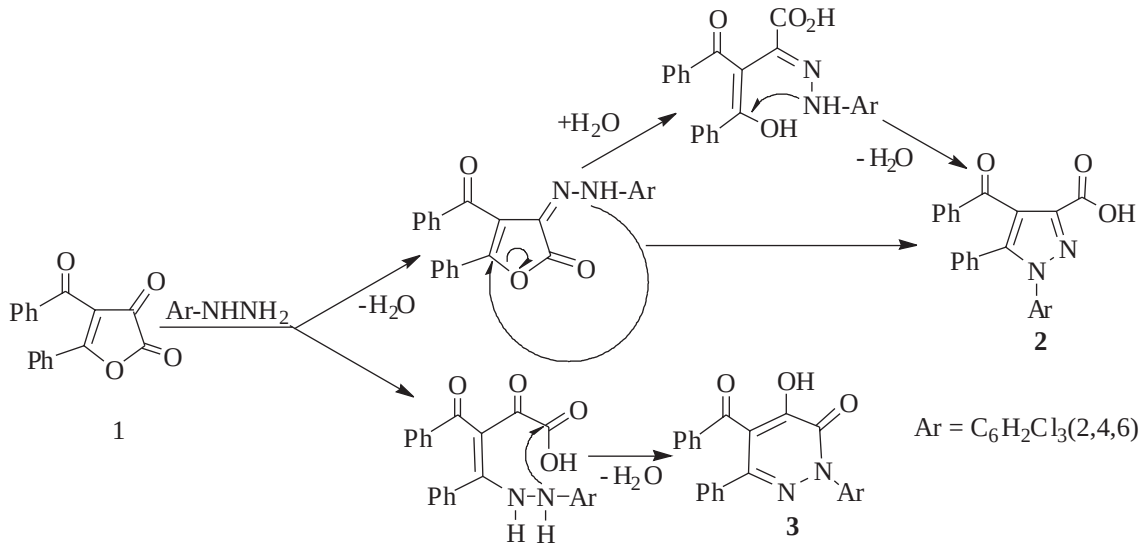
Compound **8c** was prepared according to the general procedure above with a reflux time of 3 h resulting in 75% yield (0.351 g); mp 268 °C (n-butanol); IR: 3150-2900 cm^{-1} (b, HN-C=OΔN=C-OH), 3076 cm^{-1}

(Ar-H), 1668 cm^{-1} (C=O); ^{13}C-NMR ($CDCl_3$-d_6): δ = 154.94 (C=O), 146.91 (C-7a), 138.86 (C-4), 137.99 (C-3), 136.18, 132.63, 131.64, 131.38, 131.07, 130.91, 130.78, 130.63, 129.99, 129.77, 119.59, 114.63 (C-3a).

Anal. Calcd. for $C_{23}H_{13}Cl_3N_4O$: C, 59.06; H, 2.80; N, 11.98; Cl, 22.74. Found: C, 59.29; H, 2.82; N, 11.93; Cl, 22.65.

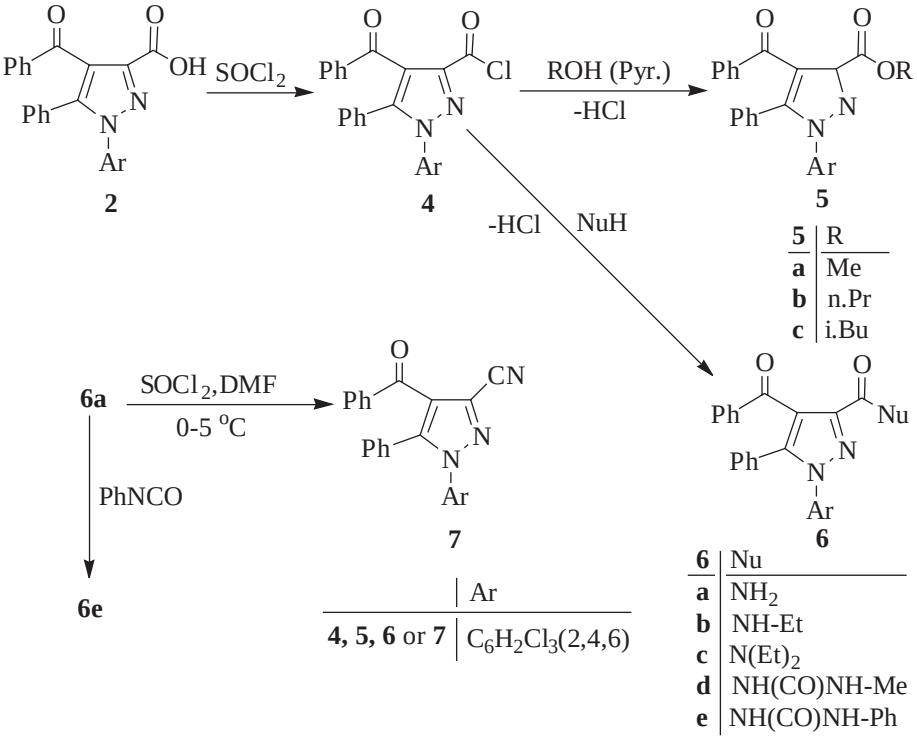
Results and Discussion

Our experiment resulted in the formation of the title compound **2** together with a new pyridazinone derivative **3,** which is a by-product, from furandione **1** and 2,4,6-trichlorophenyl- hydrazine, in approximately 35% and 30% yields, respectively. It can be seen that the yield of pyrazole acid **2** decreases, while the yield of pyridazinone derivative **3** increases after a prolonged reaction (5 h), compared with the reaction between furandione **1** and phenylhydrazine (45 min)[9] in which pyrazole acid and pyridazinone derivatives are obtained in yields of 55% and 10%, respectively. This shows that nucleophilic attacks of the NH group adjacent to the phenyl ring in the hydrazine compound at C-2, C-3 and C-5 positions of the furan ring in **1**, for the starting of reactions leading to intermediates, were either lost or decreased considerably due to the steric and inductive effects of chlorine atoms on the phenyl group. Owing to these effects, the reactions for the formation of both **2** and **3** should mainly be started by nucleophilic attacks of the only NH_2 group of 2,4,6-trichlorophenylhydrazine at C-3 and C-5 positions of furandione **1**, respectively. In addition, the reaction speeds of the final steps of these reactions should probably be much slower than the reaction speeds of their first steps. Reasonable proposals for the reaction pathways are outlined briefly in Scheme 1.



Scheme 1

The structures of **2** and **3** were confirmed by analytical, IR, ^{1}H-NMR and ^{13}C-NMR spectroscopic data, based on the structural analogy of similar compounds[9]. Mass spectra of compounds **2** and **3** show the presence of molecular ions at 471.0 for **2** and 471.1 for **3**. In addition, acid **2** could easily be converted into the corresponding derivatives such as ester **5**, amide **6** and nitrile **7** via reactions of acid chloride **4** with various O- and N- nucleophiles (Scheme 2).

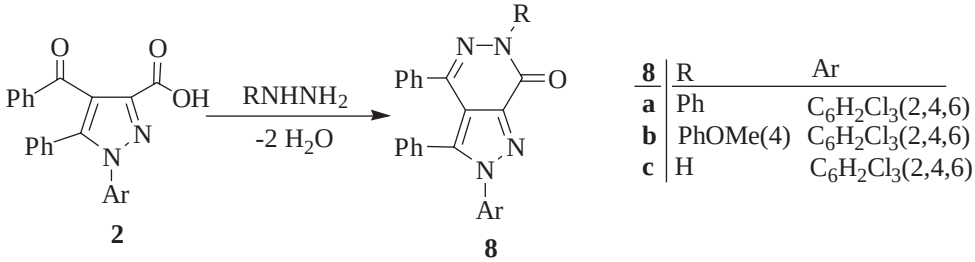


Scheme 2

The structural analogy of all the ester **5** or amide **6** derivatives can easily be seen from their IR spectra. Absorption bands of ester or amide groups at about 1724 cm^{-1} and 1630 cm^{-1}, respectively, are structural characteristics. ^{13}C-NMR and IR absorptions of the nitrile group in **7** are found at 110.13 ppm and 2240 cm^{-1}, respectively.

The correct structures of unsymmetrically substituted urea derivatives **6d-e** were established by another chemical procedure consisting of the reaction of the primary amide **6a** with phenyl isocyanate, which resulted from the formation of phenyl urea derivative **6e**, originally prepared from the acid chloride **4** in the usual way. The ^{13}C-NMR and ^{1}H-NMR spectra of **6d-e** are also in full agreement with the proposed structures (see Experimental).

Reactions of pyrazole derivatives having the dicarbonyl groups in the ortho- positions with hydrazines may be a convenient method to build the pyrazolo [3,4−*d*] pyridazine systems[10]. Thus, the pyrazole acid **2** and hydrazines were cyclized to the pyrazolo [3,4−*d*] pyridazinones **8a-c**, in approximately 55-75% yields (Scheme 3).



Scheme 3

While the formation of **8c** clearly takes place via reactions of the amino groups of hydrazine with the benzoyl carbonyl and the carboxyl groups affording the pyridazine nucleus, the priority of attacks of amino or imino groups on the side chain of aryl hydrazines in benzoyl carbonyl or carboxyl groups is important for determining the reaction pathway in the formation of pyrazolo [3,4−*d*] pyridazinone derivatives **8a-b**. However, according to their elemental analyses, the elimination of 2 mol of water during the reaction indicates that it is possible for only one reaction pathway, outlined briefly in Scheme 3, to build the compounds **8a-b**.

Structure elucidations of **8a-c** are mainly based on their ^{13}C-NMR spectral data. Signals at 157.0, 149.5, 143.5, 140.0 and 185.5 ppm are assigned to the hetero-ring carbon atoms.

Acknowledgment

The authors thank the research fund of Yüzüncü Yıl University for its financial support.

References

1. R.N. Mahajan, F.H. Havaldar and P.S. Fernandes, **J. Indian Chem. Soc. 68**, 245 (1991).
2. S.L. Janus, A.Z. Magdif, B.P. Erik and N. Claus, **Monat. Chem. 130**, 1167 (1999).
3. H. Katayama and T. Oshiyama, **Can. J. Chem. 75**, 913 (1997).
4. E. Badawey and I.M. El-Ashmawey, **Eur. J. Med. Chem. 33**, 349, (1998).
5. P.D. Mishra, S. Wahidullah and S.Y. Kamat, **Indian J. Chem. Soc. Sec. B**, **37**, 199 (1998).
6. W.T. Thomson, "**Agricultural chemicals - Book II, Herbicides**", 13^{th} ed, pp. 268-269-278, Thomson Publications, California, 1997.
7. V.V. Badiger and S.C. Bennur, **Indian J. Heterocyclic Chem. 6**, 63 (1996).
8. R.H. Wiley and P. Wiley, "**Pyrazolones, Pyrazolidones and Derivatives**", pp. 102, John Wiley and Sons, New York, 1964.
9. Y. Akçamur, G. Penn, E. Ziegler, H. Sterk, G. Kollenz, K. Peters, E.M. Peters and H.G. Von Schnering, **Monat. Chem. 117**, 231, (1986)
10. Y. Akçamur, A. Şener, A.M. İpekoğlu and G. Kollenz, **J. Heterocyclic Chem. 34**, 221 (1997).
11. A. Şener, R. Kasımoğulları, M.K. Şener, İ. Bildirici and Y. Akçamur, **J. Heterocyclic Chem. 39**, 869, (2002).