Chemistry of Heterocyclic Compounds, Vol. 37, No. 9, 2001

RECYCLIZATION OF 4,5-DISUBSTITUTED 2,3-DIHYDRO-2,3-FURANDIONES BY THE ACTION OF AZOMETHINES TO GIVE TETRAHYDRO-2,3-PYRROLEDIONES

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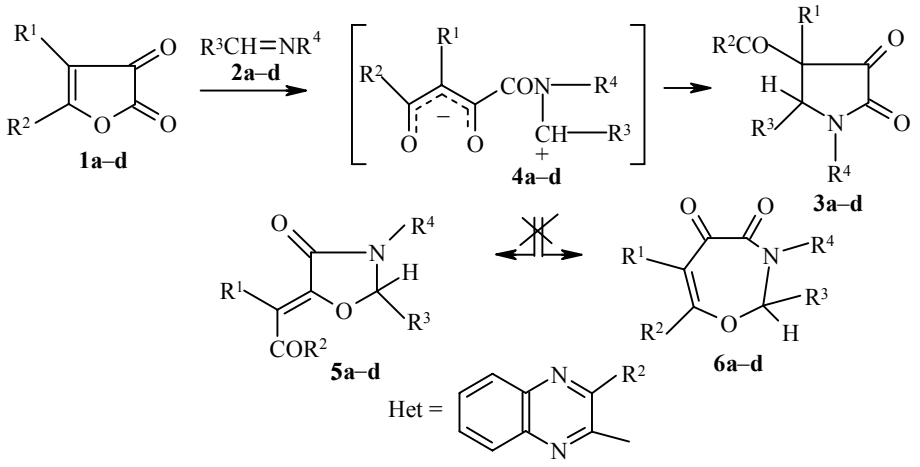
Keywords: azomethines, 2,3-dihydro-2,3-furandiones, tetrahydro-2,3-pyrrolediones, recyclization.

Unactivated azomethines react with aroylketenes generated by the thermal decarbonylation of 5-aryl-2,3-dihydro-2,3-furandiones to give 6-aryl-3,4-dihydro-2H-1,3-oxazin-4-ones [1]. Azomethines containing electron-donating groups react with 5-aryl- and 5-aryl-4-halo-2,3-dihydro-2,3-furandiones under conditions excluding decarbonylation of the furandiones to give 4-aroyl-3-hydroxy-2,5-dihydro-1H-2-pyrrolones [2]. 4-Benzoyl-5-phenyl-2,3-dihydro-2,3-furandione reacts with azomethines to give 4,7a-diphenyl-2,3,5,6-tetrahydrofuro[3,2-*e*][1,3]oxazine-2,3-diones, which recyclize upon heating to 4-benzoyl-5-phenyl-2,3-dihydro-2,3-pyrrolediones [3].



1 a $R^1 = R^2 =$ Ph; **b** $R^1 =$ Ph, $R^2 = 2,5\text{-}Me_2C_6H_3$; **c** $R^1 =$ Het, $R^2 =$ Ph; **d** $R^1 =$ Het, $R^2 = p\text{-}MeC_6H_4$.
2 a $R^3 = p\text{-}Me_2NC_6H_4$, $R^4 =$ Bn; **b** $R^3 = p\text{-}Et_2NC_6H_4$, $R^4 =$ Bn; **c** $R^3 = p\text{-}Me_2NC_6H_4$, $R^4 =$ Ph;
d $R^3 = p\text{-}Me_2NC_6H_4$, $R^4 = p\text{-}MeOC_6H_4$. **3–6 a** $R^1 = R^2 =$ Ph, $R^3 = p\text{-}Me_2NC_6H_4$, $R^4 =$ Bn;
b $R^1 =$ Ph, $R^2 = 2,5\text{-}Me_2C_6H_3$, $R^3 = p\text{-}Et_2NC_6H_4$, $R^4 =$ Bn; **c** $R^1 =$ Het, $R^2 = R^4 =$ Ph,
$R^3 = p\text{-}Me_2NC_6H_4$; **d** $R^1 =$ Het, $R^2 = p\text{-}MeC_6H_4$, $R^3 = p\text{-}Me_2NC_6H_4$, $R^4 = p\text{-}MeOC_6H_4$

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0009-3122/01/3709-1177$25.00

The reaction of 4,5-diaryl- and 5-aryl-4-(3-aryl-2-quinoxalinyl)-2,3-dihydro-2,3-furandiones **1a-d** with azomethines **2a-d** gives substituted 4,5-diaryl-4-aroyl- and 5-aryl-4-(3-aryl-2-quinoxalyl)-4-aroyl-2,3,4,5-tetrahydro-1H-2,3-pyrrolediones **3a-d**.

Opening of the ring in furandiones **1a-d** probably occurs in the first step by means of the nitrogen atom in azomethines **2a-d** to give zwitter-ions **4a-d**, stabilized through intramolecular cyclization. Isomeric structures 2,3,4,5-tetrahydro-1,3-oxazol-4-ones **5a-d** and 2,3,4,5-tetrahydro-1,3-oxazepine-4,5-diones could be eliminated for the products formed using the spectral data.

4-Benzoyl-1-benzyl-5-*p*-dimethylaminophenyl-4-phenyl-2,3,4,5-tetrahydro-1H-2,3-pyrroledione (3a). A solution of furandione **1a** (1.00 g, 3.8 mmol) and azomethine **2a** (0.90 g, 3.8 mmol) in absolute chloroform (10 ml) was heated at reflux for 2 h and cooled. The precipitate was filtered off to give 1.49 g (80%) of compound **3a**; mp 197-198°C (2-propanol). IR spectrum (vaseline mull), ν, cm^{-1}: 1770 ($C_{(2)}$=O), 1725 ($C_{(3)}$=O), 1680 ($C_{(4)}$–C=O). ^{1}H NMR spectrum (400 MHz, DMSO-d_6), δ, ppm, *J* (Hz): 2.78 (6H, s, Me_2N); 3.80 (1H, d, *J* = 14.3, CH in CH_2, part of AB system); 5.04 (1H, d, *J* = 14.3, CH in CH_2, part of AB system); 5.98 (1H, s, $C_{(5)}$H); 6.44-7.47 (19H, m, 3Ph + C_6H_4). ^{13}C NMR spectrum (100 MHz, DMSO-d_6), δ, ppm: 39.53 (Me_2N), 45.84 (CH_2), 62.49 ($C_{(4)}$), 67.74 ($C_{(5)}$), 119.58-150.08 (Ar), 156.86 ($C_{(2)}$=O), 192.15 (Ph–C=O), 192.31 ($C_{(3)}$=O). Found, %: C 78.72; H 5.78; N 5.70. $C_{32}H_{28}N_2O_3$. Calculated, %: C 78.67; H 5.78; N 5.73.

1-Benzyl-4-(2,5-dimethylbenzoyl)-5-*p*-diethylaminophenyl-4-phenyl-2,3,4,5-tetrahydro-1H-2,3-pyrroledione (3b). Yield of **3b** 1.47 g (71%); mp 182-183°C (2-propanol). IR spectrum in vaseline mull, ν, cm^{-1}: 1776 ($C_{(2)}$=O), 1719 ($C_{(3)}$=O), 1695 ($C_{(4)}$–C=O). ^{1}H NMR spectrum (400 MHz, DMSO-d_6), δ, ppm, *J* (Hz): 1.01 (6H, t, *J* = 7.0, 2MeCH$_2$); 1.92 (3H, s, Me); 1.96 (3H, s, Me); 3.24 (4H, q, *J* = 7.0, 2CH$_2$Me); 3.94 (1H, d, *J* = 14.5, CH in CH_2, part of AB system); 4.93 (1H, d, *J* = 14.5, CH in CH_2, part of AB system); 5.78 (1H, s, $C_{(5)}$H); 6.44-7.28 (17H, m, 2Ph + C_6H_4 + C_6H_3). Found, %: C 79.42; H 6.70; N 5.20. $C_{36}H_{36}N_2O_3$. Calculated, %: C 79.38; H 6.66; N 5.14.

2-(3-Benzoyl-2-*p*-dimethylaminophenyl-4,5-dioxo-1-phenyl-2,3,4,5-tetrahydro-1H-3-pyrrolyl)-3-phenylquinoxaline (3c). Yield of **3c** 1.97 g (86%); mp 146-148°C (1:2 ethyl acetate–hexane). IR spectrum in vaseline mull, ν, cm^{-1}: 1770 ($C_{(5)}$=O_{pyrrol}), 1700 ($C_{(4)}$=O_{pyrrol}), 1680 ($C_{(3)}$–C=O_{pyrrol}). ^{1}H NMR spectrum (400 MHz, DMSO-d_6), δ, ppm: 2.65 (6H, s, Me_2N); 6.23 (1H, s, $C_{(2)}H_{pyrrol}$); 6.80-7.98 (23H, m, 3Ph + 2C_6H_4). Found, %: C 77.77; H 4.98; N 9.27. $C_{39}H_{30}N_4O_3$. Calculated, %: C 77.72; H 5.02; N 9.30.

2-(2-*p*-Dimethylaminophenyl-1-*p*-methoxyphenyl-4,5-dioxo-3-*p*-toluoyl-2,3,4,5-tetrahydro-1H-3-pyrrolyl)-3-*p*-tolylquinoxaline (3d). Yield of **3d** 1.81 g (72%); mp 176-178°C (1:2 ethyl acetate–hexane). IR spectrum in vaseline mull, ν, cm^{-1}: 1770 ($C_{(5)}$=O_{pyrrol}), 1715 ($C_{(4)}$=O_{pyrrol}), 1679 ($C_{(3)}$–C=O_{pyrrol}). ^{1}H NMR spectrum (250 MHz, DMSO-d_6), δ, ppm: 2.19 (3H, s, Me); 2.29 (3H, s, Me); 2.66 (6H, s, Me_2N); 3.77 (3H, s, MeO); 6.22 (1H, s, $C_{(2)}H_{pyrrol}$); 6.80-8.11 (20H, m, 5C_6H_4). Found, %: C 76.37; H 5.50; N 8.46. $C_{42}H_{36}N_4O_4$. Calculated, %: C 76.34; H 5.49; N 8.48.

This work was carried out with the financial support of the Russian Basic Research Fund (Project No. 01-03-32641).

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