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HUISGEN REACTION OF NITRILE OXIDES AND NITRILE IMINES LEADING TO ISOXAZOLINE AND PYRAZOLE-4,6-DIONES

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Huisgen reaction of nitrile oxides and nitrile imines generated in situ in the presence of N-benzylmaleimide afforded regiospecifically the corresponding cis-3-aryl-5-benzyl-3a,4,6,6a-tetrahydro-4H,6H-pyrrolo[3,4-d]isoxazoline-4,6-diones and cis-3-aryl-5-benzyl-1-(2',4'-dibromophenyl)-3a,4,6,6a-tetrahydro-1H,5H-pyrrolo[3,4-c]pyrazole-4,6-diones in good yield.

Keywords: N-benzylmaleimide, nitrile imines, nitrile oxide, Huisgen reaction, regiospecific.

The exploitation of a simple dipolarophile with different functionalities for the synthesis of heterocycles is a worthwhile contribution to cycloadditions [1-7]. Among the plethora of dipolarophiles, maleimides have taken an important place in organic chemistry. This is an important class of substrates for biological and chemical applications. In biological applications they are used as chemical probes of protein structure [8] and for the production of antibiotics [9], herbicides [9], and pesticides [10]. The maleimide moiety, due to its dienophilic nature [11-13], can be used in Michael addition as well as a dipolarophile in 1,3-dipolar cycloadditions [14-19].

Earlier we reported cycloadditions of a number of 1,3-dipoles with various dipolarophiles [20-41]. In this communication as a part of our continuing studies we are reporting new isoxazoles and pyrazoles from Huisgen reaction of variously substituted nitrile oxides and nitrile imines with N-benzylmaleimide. The reaction of various nitrones with N-benzylmaleimide has already been reported [40]. These reactions have been taken up to study the effect of various substituents on the reaction, especially when the phenyl ring moves one atom away from the nitrogen atom of the imide moiety.

The nitrile oxides **1a-h** were prepared *in situ via* dehydrohalogenation of the corresponding aldoxime [42-44] in boiling ethanol using chloramine-T [45]. The nitrile imines **4a-e** were prepared *in situ via* dehydrobromination of the corresponding N-arylbenzohydrazonoyl bromide in ice-cold THF in the presence of triethylamine [33]. N-Benzylmaleimide was prepared as reported earlier [40]. The Huisgen reaction of N-benzylmaleimide with nitrile oxides (Scheme 1) and nitrile imines (Scheme 2) produced *cis*-3-aryl-5-benzyl-3a,4,6,6a-tetrahydro-4H,6H-pyrrolo[3,4-*d*]isoxazoline-4,6-diones and *cis*-3-aryl-5-benzyl-1-(2',4'-dibromophenyl)-3a,4,6,6a-tetrahydro-1H,5H-pyrrolo[3,4-*c*]pyrazole-4,6-diones, respectively. The products have been characterized by their melting points, elemental analysis, and spectral (IR, ^{1}H NMR and mass-spectra) data. In these reactions only a single stereoisomer corresponding to the *cis*-configuration has been obtained, showing that the reaction is regiospecific.

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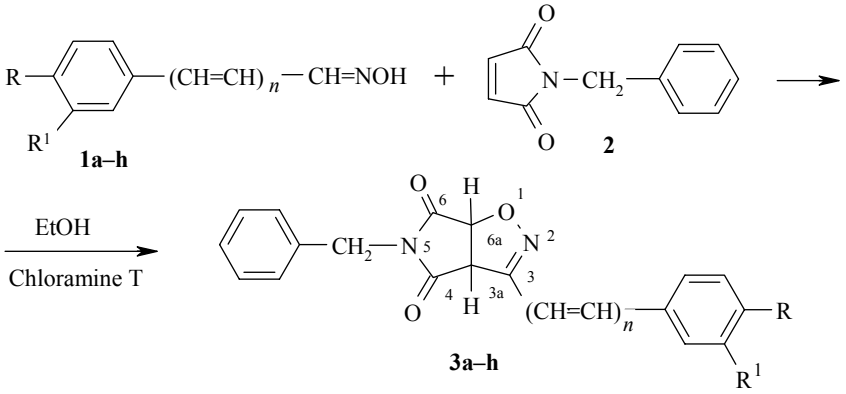
Scheme 1



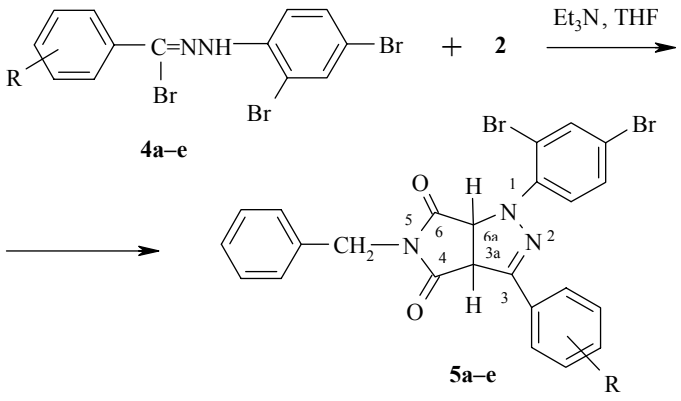
1,3 a R = R^1 = H, **b** R = OMe, R^1 = H, **c** R = Cl, R^1 = H, **d** R = R^1 = OMe, **e** R = R^1 = H, **f** R = NMe_2, R^1 = H, **g** R = OH, R^1 = OMe, **h** R = NO_2, R^1 = H; **a-d**, **f-h** *n* = 0, **e** *n* = 1

The infrared spectra of all these products showed two characteristic absorption bands for two carbonyl functions of the imide moiety at 1770-1798 (medium to weak intensity), at 1714-1728 (sharp to strong) for isoxazolines, at 1771-1785 (medium to weak), and at 1701-1708 cm^{-1} (sharp and strong) for pyrazole derivatives. The five-membered imide ring behaved as a cross-conjugated system where delocalization of the nitrogen lone pair of electrons was possible only with one carbonyl group.

This delocalization lowered the frequency of one carbonyl, but the other carbonyl behaved like in a five-membered ketone, demonstrating a more intense second absorption band at a higher frequency.

In the ^1H NMR spectra of these products two doublets were observed at δ 4.80-5.06 (H-3a) and δ 5.61-5.78 ppm (H-6a) in the case of isoxazolines, and at δ 4.73-4.85 (H-3a) and δ 5.63-5.78 ppm (H-6a) in the case of pyrazoles. Both doublets had a coupling constant typical of the *cis*-product. Thus only a single diastereoisomer corresponding to the *cis*-configuration was obtained, showing that the reaction is regiospecific.

Scheme 2



4,5 a R = H, **b** R = 4-Cl, **c** R = 4-OMe, **d** R = 3-NO_2, **e** R = 4-OH

In conclusion, we have prepared new isoxazole and pyrazole derivatives in one step by the regiospecific Huisgen reaction of nitrile oxides and nitrile imines with N-benzylmaleimide. The one atom spacer between the nitrogen and aryl ring of maleimide increased the reaction rate.

EXPERIMENTAL

Melting points were determined by a Gallen Kamp apparatus and are uncorrected. IR spectra were recorded in nujol on a Perkin-Elmer spectrum RX-I series FT IR spectrophotometer at the Department of Chemistry, Punjabi University, Patiala. ^{1}H NMR spectra were recorded on a Bruker AC-300F (300 MHz) spectrometer using TMS as an internal standard. The mass spectra were recorded on a VG-70-S mass spectrometer by SAIF, Punjab University, Chandigarh.

Preparation of Compounds 3a-h (General Procedure). In a 250 ml round bottom flask fitted with a reflux condenser, a mixture of aldoxime (0.01 mol), N-benzylmaleimide (0.01 mol), and chloramine-T (0.01 mol) in ethanol (100 ml) was heated to reflux with continuous stirring for the appropriate time. The sodium chloride formed in the reaction was filtered off and washed with ethanol. The filtrate and washings were concentrated under reduced pressure, and the residue was extracted with CH_2Cl_2. The extract was first washed with distilled water and then with 1N aqueous NaOH and dried over anhydrous Na_2SO_4. The solvent was removed under vacuum, and the viscous residue was crystallized.

5-Benzyl-3-phenyl-3a,4,6,6a-tetrahydro-4H,6H-pyrrolo[3,4-*d*]isoxazoline-4,6-dione (3a). Mp 155-157°C. IR spectrum, ν, cm^{-1}: 1780, 1720 (>C=O). ^{1}H NMR spectrum, δ, ppm (*J*, Hz): 4.59 (2H s, >CH_2); 4.81 (1H d, $J_{3a\text{-}6a}$ = 9.2, H-3a); 5.67 (1H d, $J_{6a\text{-}3a}$ = 9.4, H-6a); 7.21-7.45 (10H, aromatic). Mass spectrum, *m/z*: 306. Found, %: C 70.63; H 4.42; N 9.31. $C_{18}H_{14}N_2O_3$. Calculated, %: C 70.58; H 4.57; N 9.15.

5-Benzyl-3-(4-methoxyphenyl)-3a,4,6,6a-tetrahydro-4H,6H-pyrrolo[3,4-*d*]isoxazoline-4,6-dione (3b). Mp 160-162°C. IR spectrum, ν, cm^{-1}: 1772, 1715 (>C=O). ^{1}H NMR spectrum, δ, ppm (*J*, Hz): 3.84 (3H, s, OCH_3); 4.57 (2H, s, >CH_2); 4.83 (1H, d, $J_{3a\text{-}6a}$ = 9.1, H-3a); 5.66 (1H, d, $J_{6a\text{-}3a}$ = 9.2, H-6a); 7.28-8.12 (9H, aromatic). Mass spectrum, *m/z*: 336. Found, %: C 67.14; H 4.83; N 8.21. $C_{19}H_{14}N_2O_4$. Calculated, %: C 67.85; H 4.76; N 8.33.

5-Benzyl-3-(4-chlorophenyl)-3a,4,6,6a-tetrahydro-4H,6H-pyrrolo[3,4-*d*]isoxazoline-4,6-dione (3c). Mp 198-199°C. IR spectrum, ν, cm^{-1}: 1798, 1714 (>C=O). ^{1}H NMR spectrum, δ, ppm (*J*, Hz): 4.59 (2H, s, >CH_2); 5.01 (1H, d, $J_{3a\text{-}6a}$ = 9.4, H-3a); 5.61 (1H, d, $J_{6a\text{-}3a}$ = 9.5, H-6a); 7.28-8.27 (9H, aromatic). Mass spectrum, *m/z*: 340.5. Found, %: C 63.20; H 3.93; N 8.16. $C_{18}H_{13}ClN_2O_3$. Calculated, %: C 63.43; H 3.81; N 8.22.

5-Benzyl-3-(3,4-dimethoxyphenyl)-3a,4,6,6a-tetrahydro-4H,6H-pyrrolo[3,4-*d*]isoxazoline-4,6-dione (3d). Mp 183-186°C. IR spectrum, ν, cm^{-1}: 1780, 1728 (>C=O). ^{1}H NMR spectrum, δ, ppm (*J*, Hz): 3.98 (6H, s, 2 × OCH_3); 4.59 (2H, s, >CH_2); 4.91 (1H, d, $J_{3a\text{-}6a}$ = 9.8, H-3a); 5.56 (1H, d, $J_{6a\text{-}3a}$ = 9.7, H-6a); 7.04-7.58 (8H, aromatic). Mass spectrum, *m/z*: 366. Found, %: C 65.43; H 4.75; N 7.51. $C_{20}H_{18}N_2O_5$. Calculated, %: C 65.57; H 4.91; N 7.65.

5-Benzyl-3-styryl-3a,4,6,6a-tetrahydro-4H,6H-pyrrolo[3,4-*d*]isoxazoline-4,6-dione (3e). Mp 220-221°C. IR spectrum, ν, cm^{-1}: 1770, 1715 (>C=O). ^{1}H NMR spectrum, δ, ppm (*J*, Hz): 4.58 (2H, s, >CH_2); 4.80 (1H, d, $J_{3a\text{-}6a}$ = 9.2, H-3a); 5.78 (1H, d, $J_{6a\text{-}3a}$ = 9.4, H-6a); 5.82 (1H, q, $J_{\alpha\text{-}\beta}$ = 16.2, H_α styryl); 6.42 (1H, d, $J_{\beta\text{-}\alpha}$ = 16.3, H_β styryl); 7.30-7.95 (10H, aromatic). Mass spectrum, *m/z*: 332. Found, %: C 71.90; H 4.92; N 8.21. $C_{20}H_{16}N_2O_3$. Calculated, %: C 72.18; H 4.84; N 8.43.

5-Benzyl-3-(4-N,N-dimethylaminophenyl)-3a,4,6,6a-tetrahydro-4H,6H-pyrrolo[3,4-*d*]isoxazoline-4,6-dione (3f). Mp 174-175°C. IR spectrum, ν, cm^{-1}: 1782, 1720 (>C=O). ^{1}H NMR spectrum, δ, ppm (*J*, Hz): 2.94 (6H, s, $N(CH_3)_2$); 4.59 (2H, s, >CH_2); 4.87 (1H, d, $J_{3a\text{-}6a}$= 9.1, H-3a); 5.71 (1H, d, $J_{6a\text{-}3a}$ = 9.1, H-6a); 7.11-8.23 (9H, aromatic). Mass spectrum, *m/z*: 349. Found, %: C 68.52; H 5.61; N 11.91. $C_{20}H_{19}N_3O_3$. Calculated, %: C 68.76; H 5.44; N 12.03.

5-Benzyl-3-(4-hydroxy-3-methoxyphenyl)-3a,4,6,6a-tetrahydro-4H,6H-pyrrolo[3,4-*d*]isoxazoline-4,6-dione (3g). Mp 189-191°C. IR spectrum, ν, cm^{-1}: 1781, 1722 (>C=O). ^{1}H NMR spectrum, δ, ppm (*J*, Hz): 3.62 (3H, s, OCH_3); 4.58 (2H, s, >CH_2); 4.91 (1H, d, $J_{3a\text{-}6a}$ = 9.2, H-3a); 5.52 (1H, s, OH); 5.69 (1H, d, $J_{6a\text{-}3a}$ = 9.3, H-6a); 7.01-8.41 (8H, aromatic). Mass spectrum, *m/z*: 352. Found, %: C 64.31; H 4.32; N 7.82. $C_{19}H_{16}N_2O_5$. Calculated, %: C 64.77; H 4.54; N 7.95.

5-Benzyl-3-(4-nitrophenyl)-3a,4,6,6a-tetrahydro-4H,6H-pyrrolo[3,4-*d*]isoxazoline-4,6-dione (3h). Mp 180-183°C. IR spectrum, ν, cm^{-1}: 1780, 1721 (>C=O). ^1H NMR spectrum, δ, ppm (*J*, Hz): 4.59 (2H, s, $>CH_2$); 4.91 (1H, d, $J_{3a\text{-}6a}$ = 9.6, H-3a); 5.72 (1H, d, $J_{6a\text{-}3a}$ = 9.5, H-6a); 7.14-8.34 (9H, aromatic). Mass spectrum, *m/z*: 306. Found, %: C 60.43; H 3.79; N 11.83. $C_{18}H_{14}N_2O_3$. Calculated, %: C 61.53; H 3.70; N 11.96.

Preparation of 5a-e (General Procedure). In a 250 ml conical flask fitted with a two-way head having a dropping funnel and a guard tube N-(α-bromobenzylidene)-N'-(2',4'-dibromophenyl)hydrazine (0.01 mol) and N-benzylmaleimide (0.01 mol) were taken in 50 ml THF. This mixture was magnetically stirred and cooled in an ice-bath. To purify the solution so obtained, 0.01 mol of triethylamine was added dropwise. The solution was allowed to stand overnight and then filtered off to remove triethylamine hydrogen bromide. Then the filtrate was washed with water and dried over anhydrous Na_2SO_4. After that the solvent was distilled off in vacuum, and the crude product obtained was crystallized.

5-Benzyl-1-(2',4'-dibromophenyl)-3-phenyl-3a,4,6,6a-tetrahydro-1H,5H-pyrrolo[3,4-*c*]pyrazole-4,6-dione (5a). Mp 188-190°C. IR spectrum, ν, cm^{-1}: 1771, 1701 (>C=O). ^1H NMR spectrum, δ, ppm (*J*, Hz): 4.57 (2H, s, $>CH_2$); 4.80 (1H, d, $J_{3a\text{-}6a}$ = 10.7, H-3a); 5.74 (1H, d, $J_{6a\text{-}3a}$ = 10.7, H-6a); 7.13-8.04 (13H, aromatic). Mass spectrum, *m/z*: 539. Found, %: C 53.2; H 3.13; N 7.71. $C_{24}H_{17}Br_2N_3O_2$. Calculated, %: C 53.4; H 3.15; N 7.79.

5-Benzyl-3-(4-chlorophenyl)-1-(2',4'-dibromophenyl)-3a,4,6,6a-tetrahydro-1H,5H-pyrrolo[3,4-*c*]-pyrazole-4,6-dione (5b). Mp 161-164°C. IR spectrum, ν, cm^{-1}: 1785, 1708 (>C=O). ^1H NMR spectrum, δ, ppm (*J*, Hz): 4.61 (2H, s, $>CH_2$); 4.85 (1H, d, $J_{3a\text{-}6a}$ = 10.7, H-3a); 5.63 (1H, d, $J_{6a\text{-}3a}$ = 10.7, H-6a); 7.19-8.28 (12H, aromatic). Mass spectrum, *m/z*: 573.5. Found, %: C 50.4; H 2.83; N 7.31. $C_{24}H_{16}Br_2ClN_3O_3$. Calculated, %: C 50.2; H 2.78; N 7.32.

5-Benzyl-1-(2',4'-dibromophenyl)-3-(4-methoxyphenyl)-3a,4,6,6a-tetrahydro-1H,5H-pyrrolo[3,4-*c*]-pyrazole-4,6-dione (5c). Mp 182-185°C. IR spectrum, ν, cm^{-1}: 1782, 1706 (>C=O). ^1H NMR spectrum, δ, ppm (*J*, Hz): 3.89 (3H, s, OCH_3); 4.58 (2H, s, $>CH_2$); 4.83 (1H, d, $J_{3a\text{-}6a}$ = 10.8, H-3a); 5.76 (1H, d, $J_{6a\text{-}3a}$ = 10.7, H-6a); 6.95-7.96 (12H, aromatic). Mass spectrum, *m/z*: 569. Found, %: C 53.2; H 3.13; N 7.71. $C_{25}H_{19}Br_2N_3O_3$. Calculated, %: C 52.7; H 3.33; N 7.38.

5-Benzyl-1-(2',4'-dibromophenyl)-3-(3-nitrophenyl)-3a,4,6,6a-tetrahydro-1H,5H-pyrrolo[3,4-*c*]-pyrazole-4,6-dione (5d). Mp 207-210°C. IR spectrum, ν, cm^{-1}: 1780, 1708 (>C=O). ^1H NMR spectrum, δ, ppm (*J*, Hz): 4.58 (2H, s, $>CH_2$); 4.73 (1H, d, $J_{3a\text{-}6a}$ = 10.7, H-3a); 5.72 (1H, d, $J_{6a\text{-}3a}$ = 10.7, H-6a); 7.25-8.73 (12H, aromatic). Mass spectrum, *m/z* (*I*, %): 584, 148 (100). Found, %: C 49.76; H 2.65; N 9.71. $C_{24}H_{16}Br_2N_4O_4$. Calculated, %: C 49.31; H 2.73; N 9.58.

5-Benzyl-1-(2',4'-dibromophenyl)-3-(4-hydroxyphenyl)-3a,4,6,6a-tetrahydro-1H,5H-pyrrolo[3,4-*c*]-pyrazole-4,6-dione (5e). Mp 186-190°C. IR spectrum, ν, cm^{-1}: 1780, 1701 (>C=O). ^1H NMR spectrum, δ, ppm (*J*, Hz): 4.59 (2H, s, $>CH_2$); 4.75 (1H, d, $J_{3a\text{-}6a}$ = 10.7, H-3a); 5.78 (1H, d, $J_{6a\text{-}3a}$ = 10.7, H-6a); 7.12-8.19 (12H, aromatic). Mass spectrum, *m/z*: 555. Found, %: C 51.92; H 3.01; N 7.61. $C_{24}H_{17}Br_2N_3O_2$. Calculated, %: C 51.89; H 3.06; N 7.56.

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