(8) Reaction of 1-methyl-2-lithioimidazole with dinitrogen tetroxide yields 1-methyl-2-nitroimidazole: B. A. Tertov, V. V. Burykin, and A. S. Morkovnik, *Chem. Abstr.*, **81**, 169 542 (1974). Iodination gives 2-iodo-1-methylimidazole: B. A. Tertov, V. V. Burykin, A. S. Morkovnik, and V. V. Bessanov, *Khim. Geterotsikl. Soedin.*, 1109 (1973): *Chem. Abstr.*, **79**, 126 395 (1973).
(9) R. Jones, *J. Am. Chem. Soc.*, **71**, 383 (1949).
(10) H. Giesemann and G. Hälsche, *Chem. Ber.*, **92**, 92 (1959). A modified synthetic procedure was used, based on that for the preparation of toluenesulfonylimidazole: G. S. Schmir, W. M. Jones, and L. A. Cohen, *Biochemistry*, **4**, 539 (1965).
(11) For entries d, e, f, and g, chromatography was omitted in the workup.
(12) B. A. Tertov, V. V. Burykin, and A. V. Koblik, *Khim. Geterotsikl. Soedin.*, 1552 (1972); *Chem. Abstr.*, **78**, 58 308 (1973).
(13) H. Gilman and S. Avakian, *J. Am. Chem. Soc.*, **68**, 1514 (1946).
(14) D. I. C. Scopes, A. F. Kluge, and J. A. Edwards, *J. Org. Chem.*, **42**, 376 (1977).
(15) T. Sheradsky, *J. Heterocycl. Chem.*, **4**, 413 (1967).
(16) The material originally identified as 2-iodoimidazole [H. Pauly and E. Arauner, *J. Prakt. Chem.*, **118**, 33 (1928)] was shown later to be 4-iodoimidazole (ref 3). Authentic 2-iodoimidazole has been obtained in 5% yield by reaction of iodine with 1-benzenesulfonyl-2-lithioimidazole [R. J. Sundberg, *J. Heterocycl. Chem.*, **14**, 517 (1977)]; this N-protecting group was found unsuitable for general use in the preparation of 2-X-imidazoles, a conclusion we had also reached from early studies.

Reactivity of Oxoindole-$\Delta^{3,\alpha}$-acrylates toward Diazoalkanes: An Unusual Ring Expansion

Gregory B. Bennett,* Robert B. Mason, and Michael J. Shapiro

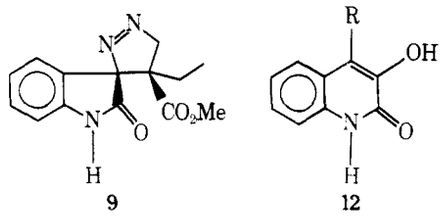
Department of Medical Chemistry Pharmaceutical Division, Sandoz, Inc., East Hanover, New Jersey 07936

Received April 4, 1978

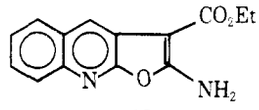
As part of our work on the regiospecific behavior of enedicarbonyl compounds[1a] we decided to examine the reaction of diazoalkanes[1b] with oxoindol-$\Delta^{3,\alpha}$-acrylates (**1**). With diazomethane acrylate **1** (X = H)[2] provided pyrazoline **3a** (X = H). The observed NMR coupling of the pyrazoline methylene and methine protons was sufficient evidence to assign structure **3a** and not **2a** to the product. Steric (tertiary vs. secondary carbon) as well as electronic (polarization of the C–C double bond) effects are such that the [1,3]-dipolar addition of diazomethane involves initial C–C bond formation α to the ester, with the reaction proceeding via a nonsynchronous intermediate such as **4** and not **5**.[3] When heated above its melting point or in refluxing xylene, pyrazoline **3a** (X = H) underwent N_2 loss giving spirocyclopropane **7a** (X = H). Reaction of acrylate **1**, X = H, with phenyldiazomethane provided the corresponding spirocyclopropane **7b** (X = H) as a single diastereomer.

Exposure of acrylate **1** (X = CN)[6] to diazomethane did not afford either a pyrazoline (**2** or **3**) or a spirocyclopropane (**7**). Instead, only quinolone **11a** (X = CN) could be isolated (92%). Rearrangement of the intermediate resulting from loss of N_2, **8**, and isomerization of the resulting exocyclic double bond out of conjugation with the cyanoester and into aromatization would account for the observed product.[7] The addition of a cyano group[8] has thus reversed the polarization of the C–C double bond while equalizing the steric effects of substitution such that the diazomethane addition now involves initial C–C bond formation β to the ester moiety (**5**). The rearrangement of isatins to quinolones has precedent in the literature.[9] Eistert and coworkers had reported that the reaction of isatin and *N*-methylisatin with diazoalkanes ($RCHN_2$) led in good yield to 4-R-substituted 3-hydroxycarbostyrils (**12**).

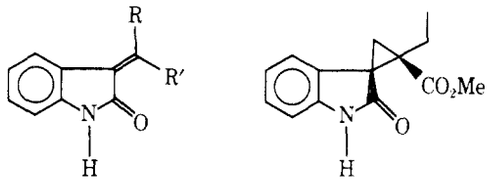
An equilibrium mixture of **11a** (X = CN) and tricyclic **13** was established after only 12 h in Me_2SO at ambient temperature. Dissolution of either **11a** or **13** in Me_2SO resulted in the same mixture. The ^{13}C chemical shifts for compounds **11a** (X = CN) and **13** were consistent with structural assignments made on the basis of other spectral data.[10] The ^{13}C NMR spectrum of **11a** displayed a single aliphatic, methine carbon which disappeared on isomerization in Me_2SO to **13**. In addition the A ring aromatic carbons β to N shifted downfield on isomerization, an indication of the imino ether tautomeric form. The rather high-field (80.2 ppm) absorption of the furan ring C α to the ester in **13** is consistent with a high-electron density resulting from mesomeric O and NH_2 participation. Furthermore, hydrolysis of **11a** (X = CN) followed by decarboxylation provided the known quinolone-3-acetic acid (**11a**, X = H), identical in all respects[11a] with the compound prepared by literature techniques.[11b]



9 **12**



13



14a, R = Et; R′ = CO_2Me
b, R = CO_2Me; R′ = Et

15

In a similar manner, reaction of **1** (X = CN) with $PhCHN_2$[12] provided the corresponding rearrangement product **11b** (X = CN) mp >325 °C in a yield of 42%. Reaction of **14a**[9c] with diazomethane, on the other hand, provided pyrazoline **9** resulting from C–C bond formation α to the ester. When heated in refluxing xylene this pyrazoline underwent smooth conversion to spirocyclopropane **15**. The double bond carbons of compound **14a** are more sterically equivalent (tertiary vs. tertiary) than in the case where X = H and yet initial C–C bond formation has still occurred α to the ester. This result lends support to the argument that only when C–C double bond polarization of the oxoindol-$\Delta^{3,\alpha}$-acrylates has been reversed (as in the case where X = CN)[13] such that dipolar species react initially at the carbon β to the ester will rearrangement to the quinolone ring system occur. Furthermore, the sequence provides an efficient procedure for the synthesis of quinolone-3-acetates. Additional work relating to this rearrangement and the regioselectivity of such dipolar addition reactions is now in process.

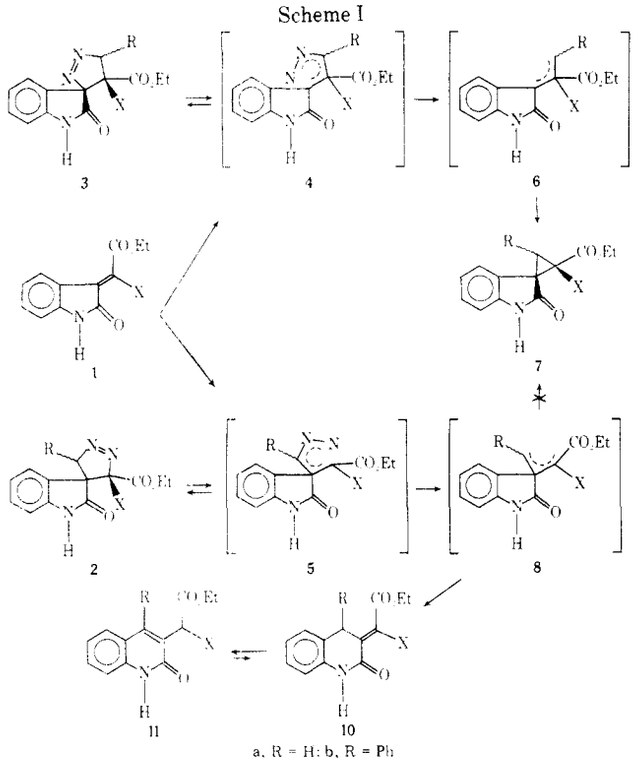
Experimental Section

The IR spectra were recorded on a Perkin-Elmer Model 257 or 457 grating spectrophotometer and NMR spectra were recorded using either a Varian T-60 or EM-360 spectrometer. ^{13}C NMR spectra were recorded using a Varian XLFT-100 spectrometer. Chemical shifts (δ) are recorded relative to Me_4Si; coupling constants (J) are given in hertz. Mass spectra were recorded using either an LKB 9000 or an AEI MS-30-D5-50 spectrometer. Melting points were obtained on a Thomas-Hoover capillary melting point apparatus and are uncorrected. In all workup procedures, the drying process involved swirling over $MgSO_4$ and filtering prior to evaporation.

Ethyl 4′,5′-Dihydro-2-oxospiro(3*H*-indole-3,3′-pyrazole)-4′-carboxylate (3a, X = H). To a solution of acrylate **1**[2] (X = H) (21.9 g, 0.1 mol) in anhydrous Et_2O (700 mL) at 0 °C was added CH_2N_2 (ca. 5.1 g, 0.12 mol) (from 36 g of Diazald).[20] After an additional 18 h at ambient temperature, the excess CH_2N_2 was quenched with HOAc and the solution was washed with aqueous $NaHCO_3$, dried, and

0022-3263/78/1943-4383$01.00/0

Scheme I



evaporated. Recrystallization of the residue from *i*-PrOH gave 17.6 g (68%) of white solid: mp 113.5–114.5 °C; NMR ($CDCl_3$) δ 1.74 (t, J = 7 Hz, 3 H), 3.45 (t, J = 8 Hz, 1 H), 3.83 (q, J = 7 Hz, 2 H), 5.12 (d, J = 8 Hz, 2 H), 6.70–7.50 (m, 4 H), and 10.83 (broad s, 1 H); IR (KBr) 3420, 1730, and 1625 cm^{-1}.

Anal. Calcd for $C_{13}H_{13}N_3O_3$: C, 60.2; H, 5.1; N, 16.2. Found: C, 60.4; H, 5.5; N, 16.3.

Ethyl 1′,2′-Dihydro-2′-oxospiro(cyclopropane-1,3′-[3*H*]-indole)carboxylate (7a, X = H). Pyrazoline **3a** (X = H) (17.6 g, 0.068 mol) was heated in refluxing toluene (300 mL) for 4 h. Evaporation of the solvent and crystallization of the residue from cold *i*-PrOH gave 9.9 g (63%) of white crystals: mp 154–6 °C; NMR ($CDCl_3$) δ 1.25 (t, J = 7 Hz, 3 H), 2.13 (d of ABq, 2 H), 2.89 (d of d, J = 7.8 Hz, 1 H), 4.17 (q, J = 7 Hz, 2 H), 6.90–7.50 (m, 4 H), and 9.82 (broad s, 1 H); IR (CH_2Cl_2) 3430, 1730, and 1625 cm^{-1}.

Anal. Calcd for $C_{13}H_{13}NO_3$: C, 67.5; H, 5.7; N, 6.1. Found: C, 67.1; H, 5.6; N, 6.2.

Ethyl 1′,2′-Dihydro-2′-oxoxpiro(2-phenylcyclopropane-1,3′-[3*H*]indole)carboxylate (7b, X = H). To a solution of $PhCHN_2$ (prepared[21] from 7.8 g of benzalhydrazone) in Et_2O (300 mL) was added acrylate **1** (X = H) (6.54 g, 30 mmol) and the mixture was allowed to stir at ambient temperature for 18 h. After quenching of the excess $PhCHN_2$ with HOAc, filtration gave a white solid. Recrystallization from *i*-PrOH provided 5.7 g (62%) of white needles: mp 175–177 °C; NMR ($CDCl_3$) δ 1.22 (t, J = 7 Hz, 3 H), 3.20 (d, J = 8 Hz, 1 H), 3.73 (d, J = 8 Hz, 1 H), 4.24 (q, J = 7 Hz, 2 H), 6.90–7.50 (m, 9 H), and 10.56 (broad s, 1 H); IR ($CHCl_3$) 1725 and 1515 cm^{-1}.

Anal. Calcd for $C_{19}H_{17}NO_3$: C, 74.2; H, 5.6; N, 4.6. Found: C, 74.1; H, 5.6; N, 4.5.

Ethyl 1,2-Dihydro-2-oxo-3-quinolinemalononitrile (11a, X = CN). To a solution of acrylate **1** (X = CN) (2.4 g, 10 mmol) in Et_2O (100 mL) and EtOH (20 mL) at 0 °C was added diazomethane (ca. 0.5 g, 12 mmol, from 3.5 g of Diazald[20]) and, after an additional 18 h at ambient temperature, the reaction mixture was quenched with HOAC, washed with aqueous $NaHCO_3$ and brine, dried, and evaporated. Recrystallization of the residue from *i*-PrOH–CH_2Cl_2 provided 2.35 g (92%) of a white solid: mp 147.5–149 °C; NMR ($CDCl_3$) δ 1.24 (t, J = 7 Hz, 3 H), 4.33 (q, J = 7 Hz, 2 H), 5.19 (s, 1 H), 7.10–7.70 (m, 4 H), 8.06 (s, 1 H), and 12.69 (broad s, 1 H); IR ($CHCl_3$) 3370, 2250, 1760, and 1670 cm^{-1}; UV (MeOH) 230 (31 910), 271 (7850), and 332 (5990) nm.

Anal. Calcd for $C_{14}H_{12}N_2O_3$: C, 65.6; H, 4.7; N, 10.9. Found: C, 65.5; H, 4.6; N, 10.8.

Upon dissolution in Me_2SO an equilibrium mixture of **11a** and **13** was established after 12 h at ambient temperature. Crystallization from *i*-PrOH–CH_2Cl_2 left **13** in the filtrate. Evaporation and recrystallization of this residue from *i*-PrOH gave **13**: mp 171.5–172.5 °C; NMR (Me_2SO) δ 1.52 (t, J = 7 Hz, 3 H), 3.26 (s, 2 H, H_2O), 4.41 (q, J = 7 Hz, 2 H), 7.40–8.00 (m, 4 H), and 8.06 (s, 1 H); IR (CH_2Cl_2) 3500, 3380, 1685, and 1640 cm^{-1}; UV (MeOH) 215 (32 700), 259 (23 810), and 343 (13 460) nm.

Anal. Calcd for $C_{14}H_{12}N_2O_3$: C, 65.6; H, 4.7; N, 10.9. Found: C, 65.3; H, 5.2; N, 10.8.

Hydrolysis of Malonitrile (11a) (X = CN). A mixture of malononitrile (**11a**, X = CN; 150 mg), H_2O (2 mL), and concentrated H_2SO_4 (1.5 mL) was heated at reflux for 18 h, cooled and poured onto ice water (30 mL). Filtration of the resulting solids gave **11a** (X = H), mp 273–275 °C; mmp 273–75 °C; lit.[11] mp 271–3 °C.

Ethyl 4-Phenyl-1,2-dihydro-2-oxo-3-quinolinemalononitrile (11b, X = CN). To a solution of $PhCNH_2$ (prepared from 2.65 g of benzalhydrazone) in Et_2O (100 mL) was added acrylate **1** (X = CN) (2.42 g, 10 mmol) in EtOH (15 mL). After 18 h at ambient temperature, the mixture was quenched with HOAc, washed with aqueous $NaHCO_3$, and extracted with 2 N NaOH. Acidification of the hydroxide extract with 2 N HCl (to pH 2) left a white solid, which after recrystallization from EtOH gave 1.39 g (42%) of a white solid: mp >325 °C; NMR ($CDCl_3$–Me_2SO) 1.21 (t, J = 7 Hz, 3 H), 4.20 (q, J = 7 Hz, 2 H), 5.39 (s, 1 H), 7.20–7.90 (m, 9 H), and 12.10 (broad s, 1 H); IR (KBr) 2240, 1750, and 1660 cm^{-1}.

Anal. Calcd for $C_{20}H_{16}N_2O_3$: C, 72.3; H, 4.9; N, 8.4. Found: C, 72.4; H, 4.4; N, 8.2.

Methyl (*Z*)-2-Oxoindole-$\Delta^{3,\alpha}$-butyrate (14a). The procedure of Mori[12c] provided a 2:1 mixture of *E* and *Z* isomers which had to be separated by chromatography over silica gel. The compound of larger R_f value was the *E* isomer, **14b**: deep yellow crystals; mp 119–20 °C; NMR ($CDCl_3$) δ 1.34 (t, J = 7 Hz, 3 H), 3.39 (q, J = 7 Hz, 2 H), 4.08 (s, 3 H), 6.88–7.50 (m, 4 H), and 9.46 (broad s, 1 H); IR ($CHCl_3$) 3450, 1710, and 1610 cm^{-1}.

Anal. Calcd for $C_{13}H_{13}NO_3$: C, 66.5; H, 5.7; N, 6.1. Found: C, 66.2; H, 5.3; N, 6.3.

The lower R_f value material, *Z* isomer (**14a**), was isolated as yellow crystals: mp 131–2 °C; NMR ($CDCl_3$) δ 1.39 (t, J = 7 Hz, 3 H), 2.87 (q, J = 7 Hz, 2 H), 4.02 (s, 3 H), 6.90–7.60 (m, 4 H), and 9.52 (broad s, 1 H); IR ($CHCl_3$) 3450, 1715, and 1615 cm^{-1}.

Anal. Found: C, 66.3; H, 5.5; N, 6.2.

Methyl 4′,5′-Dihydro-4′-ethyl 2-oxospiro(3*H*-indolo-3,3′-pyrazole)-4′-carboxylate (9). To a solution of (*Z*)-acrylate **14a** (1.30 g, 6 mmol) in Et_2O (40 mL) at 0 °C was added CH_2N_2 (ca. 0.26 g, 6.2 mmol) (from 1.82 g of Diazald[20]) and the resulting mixture was stirred at ambient temperature for 18 h. After HOAc quench, the solution was washed with $NaHCO_3$, dried, and evaporated to give 1.40 g (83%) of a light-brown oil: NMR ($CDCl_3$) δ 0.87 (t, J = 7 Hz, 3 H), 1.68 (q, J = 7 Hz, 2 H), 3.64 (s, 3 H), 4.95 (d, J = 18 Hz, 1 H), 5.41 (d, J = 18 Hz, 1 H), 6.90–7.60 (m, 4 H), and 9.28 (broad s, 1 H); IR ($CHCl_3$) 3430, 1725, and 1615 cm^{-1}.

Anal. Calcd for $C_{14}H_{15}N_3O_3$: C, 61.5; H, 5.5; N, 15.4. Found: C, 62.0; H, 6.0; N, 15.7.

Treatment of the (*E*)-acrylate **14b** in a small similar manner provided the corresponding spiropyrazole in 63% yield: mp 133–5 °C dec; NMR ($CDCl_3$) 0.90 (t, J = 7 Hz, 3 H), 1.98 (q, J = Hz, 2 H), 3.40 (s, 3 H), 4.81 (d, J = 18 Hz, 1 H), 5.46 (d, J = 18 Hz, 1 H), 6.70–7.35 (m, 4 H), and 9.12 (broad s, 1 H); IR ($CHCl_3$) 3430, 1710, and 1620 cm^{-1}.

Methyl (*Z*)-1′,2′-Dihydro-2-ethyl-2′-oxospiro(cyclopropane-1,3′-3*H*-indole)carboxylate (15). A solution of the (*Z*)-spiropyrazoline (**9**) (1.40 g, 5.1 mmol) in xylene (75 mL) was heated at reflux for 8 h. After cooling, evaporation of the solvent and distillation (180–90 (0.025 mm)) gave 0.77 g (61%) of a light yellow oil: NMR ($CDCl_3$) δ 1.06 (t, J = 7 Hz, 3 H), 2.39 (ABq, 2 H), 2.93 (q, J = 7 Hz, 2 H), 3.74 (s, 3 H), 6.90–7.40 (m, 4 H), and 10.00 (broad s, 1 H); IR (CH_2Cl_2) 3420, 1720, and 16.20 cm^{-1}.

Anal. Calcd for $C_{14}H_{15}NO_3$: C, 68.6; H, 6.2; N, 5.7. Found: C, 68.6; H, 6.6; N, 5.6.

Registry No.—**1** (X = H), 21728-28-9; **1** (X = CN), 59225-18-2; **3a** (X = H), 67487-94-9; **7a** (X = H), 67487-95-0; **7b** (X = H), 67487-96-1; **9**, 67487-97-2; **11a** (X = H), 53244-93-2; **11a** (X = CN), 67487-98-3; **11b** (X = CN), 67487-99-4; **13**, 67488-03-3; **14a**, 67488-00-0; **14b**, 67488-01-1; **15**, 67488-02-2; CH_2N_2, 334-88-3; $PhCHN_2$, 766-91-6.

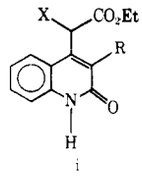
References and Notes

(1) (a) G. B. Bennett and R. B. Mason, *J. Org. Chem.*, **42**, 1919 (1977); (b) N. El-Ghandour, O. H. Rousseau, and J. Solier, *Bull. Soc. Chim. Fr.*, 2817 (1972).

(2) H. A. Brandman, *J. Heterocycl. Chem.*, **10**, 383 (1973).

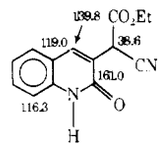
(3) While our discussion (Scheme I) makes use of a nonsynchronous concerted mechanism[4] governed by frontier orbitals, a synchronous or even a diradical mechanism[5] would have served equally well to explain both the regiospecificity of addition and the unusual ring expansion.

(4) R. Huisgen, *J. Org. Chem.*, **41**, 403 (1976).
(5) R. A. Firestone, *Tetrahedron*, **33**, 3009 (1977).
(6) G. Jones and W. J. Rae, *Tetrahedron*, **22**, 3021 (1966), and references cited therein.
(7) Structure i was dismissed on the basis of spectral data.

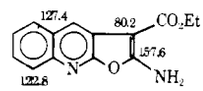


i

(8) For a discussion of the effects of an added CN moiety on pyrazoline stability see F. D. Popp and A. Catalia, *J. Org. Chem.*, **26**, 2738 (1961), and references cited therein.
(9) (a) B. Eistert and H. Selzer, *Chem. Ber.*, **96**, 1234 (1963); (b) also see P. C. Julian, H. C. Printy, R. Ketcham, and R. Doone, *J. Am. Chem. Soc.*, **75**, 5305 (1953); (c) A. Ide, Y. Mori, K. Matsumori, and H. Watanabe, *Bull. Chem. Soc. Jpn.*, **50**, 1959 (1977).
(10) Structures **11a** and **13**.



11a



13

(11) (a) mp 273–5 °C; mmp 273–5 °C; as well as comparative TLC, MS, and solution IR; (b) H. A. Lloyd, L. V. Matternas and E. C. Horning, *J. Am. Chem. Soc.*, **77**, 5932 (1955); UV (MeOH) 227 (32 700), 269 (8220), and 326 (7350).
(12) R. J. Mohrbacher and N. H. Cromwell, *J. Am. Chem. Soc.*, **79**, 401 (1957).
(13) A referee suggested that the ethyl moiety might sterically drive the ester function out of planarity with the double bond, thus destablizing any partial charge in a transition state such as **5**.

Thallium in Organic Synthesis. 53. Simple Procedures for the Replacement of a Phenolic OH Group by N=NAr, N=O, H, NH_2, and C Substituents[1,2]

Edward C. Taylor* and G. Erik Jagdmann, Jr.

Department of Chemistry, Princeton University, Princeton, New Jersey 08540

Alexander McKillop

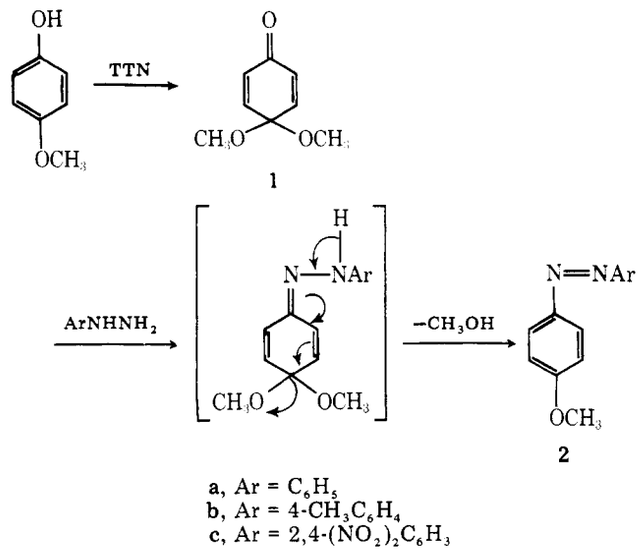
School of Chemical Sciences, University of East Anglia, Norwich NR4 7TJ, England

Received June 8, 1978

Evans et al.[3] have recently described an ingenious synthetic approach to the Amaryllidaceae alkaloid cherylline via a quinone methide prepared by a Wittig-type reaction of 4,4-dimethoxycyclohexadienones. We have recently described a general, efficient, and mild procedure for the oxidation of a variety of 4-substituted phenols to 4-substituted 4-methoxycyclohexadienones utilizing thallium(III) nitrate (TTN) in methanol or methanol/trimethyl orthoformate as solvent.[4] We now report a series of simple transformations of these cyclohexadienones which effect overall replacement of the OH group of the precursor phenol by N=NAr, N=O, H, NH_2, and C substituents.

In 1963 Hecker and Lattrell[5] reported the conversion of several 4-hydroxy-4-substituted cyclohexadienones (prepared by thallium(III) or lead(IV) acetate oxidation of the corresponding phenols) to 2,4-dinitrophenylazobenzenes by reaction with 2,4-dinitrophenylhydrazine. Because of the inaccessibility of the requisite precursor cyclohexadienones, however, there has been no subsequent synthetic exploitation

Scheme I

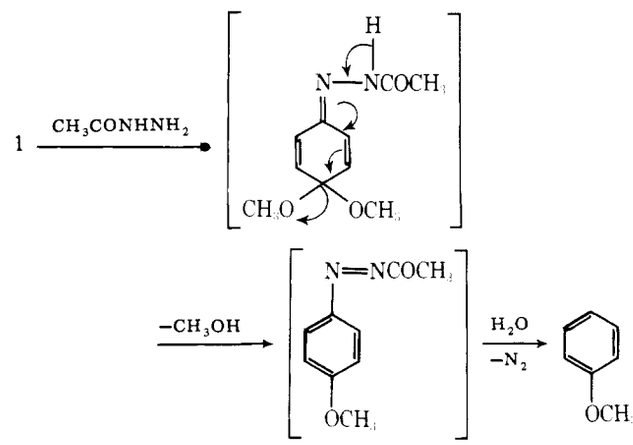


1

2

a, Ar = C_6H_5
b, Ar = 4-$CH_3C_6H_4$
c, Ar = 2,4-$(NO_2)_2C_6H_3$

Scheme II



of this type of transformation, but it appears to be general. Thus, treatment of **1** with phenylhydrazine smoothly gave 4-methoxyazobenzene **2a** in 90% yield (Scheme I). Similarly, reaction of **1** with 4-methyl- and 2,4-dinitrophenylhydrazine gave 4-methyl-4′-methoxyazobenzene (**2b**) and 2,4-dinitro-4′-methoxyazobenzene (**2c**) in 92 and 98.5% yield, respectively. This transformation can also be carried out without isolation of the intermediate cyclohexadienone; 3,4-dimethylphenol, for example, was converted to 3,4-dimethylazobenzene in 55% overall yield. Extrapolation of these results to the replacement of a phenolic OH group by H was somewhat less successful. Reaction of **1** with acethydrazide followed by addition of water resulted in the evolution of nitrogen, and anisole was isolated in 50% yield (Scheme II); 6-hydroxytetralin was similarly converted to tetralin in 31% yield.[6] Despite the moderate yields, this simple transformation could represent a mild procedure for effecting a potentially useful reduction.[7]

The principle illustrated in these transformations—conversion of **1** to an imine possessing an acidic α-hydrogen atom which can be lost in a subsequent, and spontaneous, aromatization step—appears to be capable of considerable extension. Thus, treatment of **1** with hydroxylamine led directly to 4-methoxynitrosobenzene in 91% yield (Scheme III). The overall conversion of 4-methoxyphenol to 4-methoxynitrosobenzene can also be carried out as a one-pot operation without isolation of the intermediate cyclohexadienone, although this procedure gave a somewhat lower yield (70%). Using the latter technique, 4-methylphenol, 3,4-dimethyl-

0022-3263/78/1943-4385$01.00/0