chloroformate was treated with methanol in the presence of quinoline using methylene chloride as solvent. The mixture was stirred and cooled in an ice bath for about 2 h. The mixture was washed with aqueous acid and bicarbonate and water successively. The organic layer was dried by sodium sulfate, and the solvent was removed by evaporation. Vacuum distillation of the remaining liquid yielded phenyl methyl carbonate [bp 210 °C (760 mm); lit.[12] bp 212–215 °C (760 mm)]. The IR spectrum was consistent with what is expected for the pure compound (IR bands at 3020, 2960, 1765, 1600, 1500, 690, and 760 cm^{-1}). Diphenyl carbonate was obtained from Aldrich, the purity was checked by melting point determination, and the material was was used as obtained. Methanol-*d* (99% D) was purchased from MSD isotopes and used as such. The atom fraction of deuterium of individual solutions was based on the deuterium content of this stock methanol-*d*.

Kinetics. The methanolysis was followed by monitoring the release of phenoxide ion at 240 nm using a DMS-90 UV–visible spectrophotometer fitted with a thermostated cell holder. Three milliliters of the base solution was allowed to equilibrate to the required temperature in a cuvette, and 20 μL of a stock solution of the carbonate in acetonitrile was rapidly mixed with the base. The time vs absorbance data was collected using a Varian DS-15 data station interfaced to the spectrophotometer. The first-order rate constants were calculated from the time vs absorbance data using a kinetics calculation program available with the data station.

The methoxide solution was prepared by treating clean pieces of sodium metal with methanol cooled in an ice bath. The concentration of the methoxide solution was determined by titrating against potassium hydrogen phthalate. Methoxide in methanol-*d* was prepared by the same technique using pure methanol-*d*. The atom fraction of deuterium for individual solutions was calculated on the basis of the fact that the stock methanol-*d* has 99% deuterium. All operations using methanol-*d* were done in a drybox flushed with nitrogen.

Acknowledgment. Support for this research from the donors of the Petreoleum Research Fund, administered by the American Chemical Society, Research Corporation, and the TSI grants administered by the University of North Florida is gratefully acknowledged. It is a pleasure to thank Professors William P. Huskey and William J. Wilson for their help in the statistical analysis of our data. We also thank Professor Edward A. Healy for stimulating discussions and support.

Registry No. Methyl phenyl carbamate, 13509-27-8; deuterium, 7782-39-0; diphenyl carbamate, 102-09-0.

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A New High-Yielding Method for the Preparation of 2-Alkyl- and 1,2-Dialkyl-4-nitro-5-bromoimidazoles

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Received November 5, 1991

2-Alkyl-4(5)-nitro-5(4)-bromoimidazoles and 1,2-dialkyl-4-nitro-5-bromoimidazoles have considerable synthetic and pharmacological significance. They are useful as chemotherapeutic agents[1] and potential radiosensitizers.[2] These compounds are also intermediates in the synthesis of a variety of biologically important nitroimidazole derivatives because of the ease of replacement of bromine. The corresponding 5-thio derivatives have antitumor activity.[3] Azoxymethyl nitroimidazoles with antitrichomonal and antiviral properties have also been synthesized from these intermediates.[4] A series of mercaptopurine derivatives known for their immunosuppressive and cytostatic action has been prepared from 4-nitro-5-bromoimidazoles.[5] The corresponding mercaptopyrazolopyrimidine derivatives have antigout properties.[6] 4-Nitro-5-cyanoimidazole derivatives having coccidostatic activity,[7] and novel heterocyclic systems like imidazo-, dihydroimidazo-, and tetrahydroimidazotriazines have been synthesized from 4-nitro-5-bromoimidazoles.[8]

We recently reported high-yielding methods for the preparation of 1,2-dialkyl-4-nitroimidazoles.[9-11] To extend our work to the preparation of potentially useful 5-substituted 4-nitroimidazoles, a suitable method for preparation of 5-bromo compounds without affecting the sensitive functional groups of the N-alkyl side chain was required. Reaction of bromine with a DMF solution of the substrate in the presence of potassium bicarbonate has been found to be a mild brominating system which provides the required bromo compounds in nearly quantitative yields (Table I).

Application of the Br_2–DMF–$KHCO_3$ method for the bromination of a number of 4-nitroimidazoles revealed that acid- and base-sensitive functionalities like ester, nitrile, and ketone present on N'-side chain remain intact under the conditions employed. Also, no side-chain bromination in the substrates carrying acetate, propionate, and propionitrile groups (entries 9, 7, and 5 of Table I) and ring bromination of aryl groups were observed (entries 10 and 11 of Table I). The N-unsubstituted, 2-alkylimidazoles also could be brominated in high yields with no detectable N-bromination (entries 1 and 2, Table I). The yields were practically quantitative, and the products were pure enough to be employed directly in further reactions.

In the methods described in the literature, bromination of 4-nitroimidazoles was carried out by reagents like Br_2–AcOH–$KBrO_3$,[1] Br_2–NaOH,[12] Br_2–NaOAc,[13] Br_2–H_2O,[14] and Br_2–$CHCl_3$–AcOH.[15] The superiority of the

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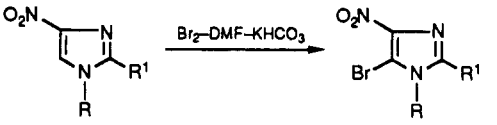
Table I



compd	R	R′	yield[a] (%)	mp (°C) (cryst solvent)	spectral data
1	H	Me	94	270–72 dec, (MeOH) (lit.[17] mp 270–71)	
2	H	Et	92	180–82 dec, (MeOH) (Lit.[18] mp 180–81)	
3[b]	Et	Me	93	136–38 (EtOAc/hexane)	^{1}H HMR (DMSO-d_6) δ 1.30 (t, 3 H), 2.45 (s, 3 H), 4.10 (q, 2 H); M$^+$ (m/z) 233
4[b]		Me	91	83–84 (EtOAc/hexane)	^{1}H NMR (DMSO-d_6) δ 0.55–0.75 (m, 4 H), 1.35 (m, 1 H), 2.55 (s, 3 H), 4.15 (d, 2 H), M$^+$ (m/z) 259
5[c]	CH_2CH_2CN	Me	95	160–61 (EtOAc)	IR (KBr) 2260 (CN); ^{1}H NMR (DMSO-d_6) δ 2.55 (2, 3 H), 3.10 (t, 3 H), 4.45 (t, 3 H); ^{13}C NMR (DMSO-d_6) δ 16.0 (q), 20.6 (t), 44.9 (t), 111.2 (s), 122.3 (s), 148.5 (s), 150.4 (s); M$^+$ (m/z) 258
6[c]	CH_2CH_2COMe	Me	93	137–38 ($CHCl_3$/hexane)	^{1}H NMR (DMSO-d_6) δ 2.15 (s, 3 H), 2.45 (s, 3 H), 3.00 (t, 2 H), 4.25 (t, 2 H); ^{13}C NMR (DMSO-d_6) δ 16.9 (q), 33.7 (t), 44.9 (t), 45.0 (q), 11.2 (s), 147.6 (s), 150.4 (s), 211.0 (s); M$^+$ (m/z) 275
7[c]	CH_2CH_2COOEt	Me	94	119–20 ($CHCl_3$/hexane)	^{1}H NMR (DMSO-d_6) δ 1.20 (t, J = 7.0 Hz, 3 H), 2.50 (s, 3 H), 2.85 (t, J = 6.0 Hz, 2 H), 3.95–4.45 (m, 4 H); M$^+$ (m/z) 305
8[d]	CH_2CH_2COOH	Me	95	224–26 (acetone)	^{1}H NMR (DMSO-d_6) δ 2.55 (s, 3 H), 2.80 (t, 2 H), 4.30 (t, 2 H); M$^+$ (m/z) 277
9[b]	CH_2COOEt	Me	93	89–90 ($CHCl_3$/hexane)	^{1}H NMR (DMSO-d_6) δ 1.25 (t, 3 H), 2.45 (s, 3 H), 4.30 (q, 2 H), 5.15 (s, 2 H); M$^+$ (m/z) 291
10[b]	$CH_2C_6H_5$	Me	91	160–61 (EtOAc) (lit.[19] mp 160–62)	^{1}H NMR (acetone-d_6) δ 2.45 (s, 3 H), 5.45 (s, 2 H), 7.10–7.60 (m, 5 H); M$^+$ (m/z) 295
11[b]	$CH_2C_6H_4$·Cl(4)	Me	92	154–56 (EtOAc)	^{1}H NMR (DMSO-d_6) δ 2.50 (s, 3 H), 5.50 (s, 2 H), 7.15–7.60 (2d, 4 H); M$^+$ (m/z) 329
12[b]	$CH_2CH_2SO_2Et$	Me	94	121–22 (EtOAc)	^{1}H NMR (DMSO-d_6) δ 1.25 (t, 3 H), 2.50 (s, 3 H), 3.25 (q, 2 H), 3.65 (t, J = 7.0 Hz, 2 H), 4.55 (t, J = 7.0 Hz, 2 H); ^{13}C NMR (DMSO-d_6) δ 9.2 (q), 17.3 (q), 42.9 (t), 50.8 (t), 53.2 (t), 111.0 (s), 148.3 (s), 151.2 (s); M$^+$ (m/z) 325

[a] Yields are for crystallized products. [b] Method employed for preparation of substrates: as in ref 10. [c] As in ref 11. [d] As in ref 9.

Table II. Bromination of 4-Nitroimidazoles by Different Methods

compd	method[a]	reaction time (h)	yield[b] (%)
3	A	10	71
	B	10	75
	E	3	93
5	A	8	64
	C	8	68
	E	2	95
6	C	8	62
	D	10	70
	E	3	93
7	A	10	61
	C	10	67
	E	3	94
10	A	10	68
	D	20	65
	E	3	91

[a] (A) Br_2–AcOH/80 °C; (B) Br_2–aq NaOH/80 °C; (C) Br_2–aq NaOAc/80 °C; (D) Br_2–$CHCl_3$ reflux; (E) Br_2–DMF–$KHCO_3$/65 °C. [b] Crystallized product.

present method over the literature methods has been demonstrated by carrying out bromination on a typical set of 4-nitroimidazoles (Table II). Br_2–DMF complex[16] could be the actual brominating species in the method described. Potassium bicarbonate serves as a mild base, scavenging the generated acid and minimizing side reactions.

Experimental Section

General. TLC was performed on silica gel plates. Melting points are uncorrected. NMR and mass spectra were recorded as reported earlier.[9,10]

General Procedure. Typically, the 4-nitroimidazole (0.1 mol) was dissolved in DMF (20 mL), and finely powdered potassium bicarbonate (15 g, 0.15 mol) was added. The stirred suspension was treated with liquid bromine (24 g, 0.15 mol), keeping the temperature below 40 °C. The mixture was warmed and held at 60–70 °C with stirring for 2–3 h. The reaction mass was diluted with ice–water (80 mL) and aqueous ammonia (10–15 mL) added until excess bromine was decolorized. The precipitated product was filtered, washed with water and dried. The yields of crystallized products along with physical and spectral data are presented in Table I.

Acknowledgment. We thank Prof. K. M. Madyastha, Indian Institute of Science, Bangalore, for useful discussions and acknowledge the technical assistance from our analytical laboratory.

Registry No. 1, 696-23-1; 1 (5-bromo), 18874-52-7; 2, 13236-63-0; 2 (5-bromo), 18874-51-6; 3, 89128-07-4; 3 (5-bromo), 139975-76-1; 4, 135009-59-5; 4 (5-bromo), 139975-77-2; 5, 89128-08-5; 5 (5-bromo), 139975-78-3; 6, 126664-28-6; 6 (5-bromo),

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Supplementary Material Available: Elemental analyses for compounds 3-9, 11, and 12 (1 page). Ordering information is given on any current masthead page.

A Practical Preparation of Functionalized Alkylbenzoquinones: Synthesis of Maesanin and Irisquinone[1]

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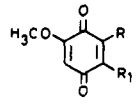
Received June 24, 1991 (Revised Manuscript Received February 10, 1992)

Natural products that contain the aromatic quinoid structure are of great medicinal importance.[2] Several highly functionalized quinones, for example, maesanin (**1a**),[3] irisquinone (**1b**),[4] irisoquin (**1c**),[5] rapanone (**1d**),[6] primine (**1e**),[7] among others,[8] which have alkyl chains of various lengths, have been isolated from natural sources and hold immense interest because of their biological activities. These molecules differ primarily in the presence



1a	Maesanin	R = $-(CH_2)_9-CH=CH-(CH_2)_3-CH_3$, R_1 = OH
1b	Irisquinone	R = $-(CH_2)_9-CH=CH-(CH_2)_5-CH_3$, R_1 = H
1c	Rapanone	R = $-(CH_2)_{12}-CH_3$, R_1 = OH
1d	Irisoquin	R = $-(CH_2)_{17}-CH_3$, R_1 = OH
1e	Primine	R = $-(CH_2)_4-CH_3$, R_1 = H

of an OH group at the C-5 position and in the type of side chain attached at the C-6 ring position, and these factors apparently determine their structure–activity relationships. The synthetic challenge of these molecules lies in the side chain attachment. This has been achieved in the past by circuitous routes, for instance, by the reaction of metallated aromatic rings with aldehydes followed by the deoxygenation of the resultant hydroxy group,[9] and by the Claisen rearrangement of a 3-phenoxy-1-alkene,[10] a method that often suffers from regiochemical problems. A conceivable convergent route to such molecules would be the direct alkylation of the aromatic nucleus with an alkyl halide. However, this method has not been utilized to synthesize any of these molecules, presumably due to the low yields of such alkylation reactions[11] with aliphatic halides. Our need for an efficient synthetic means to these alkylbenzoquinones for purposes of studying their structure–activity relationships prompted us to look for an alternative. The Heck-type reaction,[12] i.e. the palladium-mediated coupling of bromobenzene derivatives with terminal olefins, was investigated as a method for attaching a suitable side chain. The resultant molecule could then be transformed into the desired target, realizing an entry into this class of compounds. The effectiveness of this protocol has been successfully demonstrated in the syntheses of maesanin[3,13] and irisquinone.[4,14]

Maesanin is an active principle isolated from the berries of the East African *Maeses lanceolata* bush, the water extract of which is used as a preventive measure against cholera. Kubo and co-workers, who isolated this active principle on the basis of information provided by "Bwana Mganga", elucidated its structure and confirmed the structural assignment by synthesis. Maesanin is also a "host defensive stimulant" and has a direct activity against *Escherichia coli*, disrupting the enzyme KDO-transfrase, which is essential in cell-wall synthesis. In addition, maesanin has been shown to block 5-lipoxygenase and therefore may find use as an antiasthmatic drug.[15]

Results and Discussion

We began the synthesis of **1a** with the transformation of 3-bromo-4-hydroxy-5-methoxybenzaldehyde to the triacetyl compound **2** (in 54% overall yield) by a known procedure.[16] Since the acetyl groups did not survive the palladium-mediated coupling reaction, a better protecting group was sought. Methoxy methyl ether was chosen, keeping in mind that it must remain distinct from the ring methoxy group upon deprotection. Thus, the acetyl groups were removed by passing dry HCl gas through a methanolic solution of **2** at 0 °C to yield a rather unstable trihydroxy compound, which was immediately reprotected (K_2CO_3, MOM-Cl, acetone, 12 h) to obtain the tri-MOM ether **3a** in 62% yield. **3a** was subjected to a Heck-type coupling reaction with 9-decen-1-ol, mediated by palladium to furnish the alkylated product **4a**. Conversion of starting material to product was incomplete even after prolonged hours of heating, presumably because of the deposition of palladium metal. However, this problem was circumvented by the addition of excess catalyst along with Ph_3P at in-

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