ration of 9 and 34–36. The styrylchromone was dissolved in dichloromethane (5 mL/mmol), and 1.1 equiv of 1.0 M boron trichloride in dichloromethane was added dropwise at 0 °C. The reaction mixture was stirred for 1 h at 0 °C and was then warmed to ambient temperature. The reaction mixture was poured into water, and the products were extracted with either dichloromethane or ethyl acetate. The combined organic extracts were dried over sodium sulfate and concentrated. The products were purified via chromatography on silica gel with 20% methanol–dichloromethane as eluent. The styrylchromones were recrystallized from methanol.

9 (hormothamnione): 89%; mp 268–270 °C (lit.[11] mp 271 °C); ^{1}H NMR ($CDCl_3$, acetone-d_6) δ 12.70 (s, 1 H), 8.04 (s, 2 H), 7.6 (d, 1 H, J = 15.8 Hz), 7.10 (d, 1 H, J = 15.8 Hz), 6.75 (d, 2 H, J = 2 Hz), 6.51 (t, 1 H, J = 2 Hz), 4.11 (s, 3 H), 4.00 (s, 3 H), 3.94 (s, 3 H), 2.17 (s, 3 H) [lit.[11] ^{1}H NMR (THF-d_8) δ 12.8 (s, 1 H), 11.00 (s, 1 H), 8.70 (s, 1 H), 7.55 (d, 1 H, J = 15.7 Hz), 7.17 (d, 1 H, J = 15.7 Hz), 6.58 (d, 2 H, J = 2 Hz), 6.29 (t, 1 H, J = 2 Hz), 4.03 (s, 3 H), 3.93 (s, 3 H), 3.85 (s, 3 H), 2.16 (s, 3 H)]; FDMS, m^+/z 400 ($C_{21}H_{20}O_8$).

6,7-Dimethoxy-5-hydroxy-2-[2-(3,5-dihydroxyphenyl)ethenyl]chromone (31): mp 233–237 °C; ^{1}H NMR ($CDCl_3$, acetone-d_6) δ 12.44 (s, 1 H), 7.92 (s, 1 H), 6.8–7.2 (m, 3 H), 5.8–6.5 (m, 5 H), 3.64 (s, 3 H), 3.51 (s, 3 H); IR (KBr) 3400, 1600, 1490, 1460, 1370, 1290 cm^{-1}; FDMS, m/z 356 ($C_{19}H_{16}O_7$).

6,7-Dimethoxy-5-hydroxy-2-[2-(3,4-dihydroxyphenyl)ethenyl]chromone (32): mp 188–191 °C; ^{1}H NMR ($CDCl_3$, acetone-d_6) δ 12.54 (s, 1 H), 7.8 (br s, 1 H), 7.5 (br s, 1 H), 7.20 (d, 1 H, J = 16 Hz), 6.80 (d, 1 H, J = 2 Hz), 6.68 (dd, 1 H, J = 2, 10 Hz), 6.30 (d, 1 H, J = 16 Hz), 6.297 (s, 1 H), 5.83 (s, 1 H), 3.66 (s, 3 H), 3.54 (s, 3 H); IR (KBr) 3400, 1650, 1600, 1490, 1460, 1370, 1290 cm^{-1}; FDMS, m/z 356 ($C_{19}H_{16}O_7$).

6,7-Dimethoxy-5-hydroxy-2-[2-(3,5-dihydroxyphenyl)ethenyl]-3-methylchromone (33): mp 244–247 °C; ^{1}H NMR ($CDCl_3$, acetone-d_6) δ 12.51 (s, 1 H), 7.9 (br s, 1 H), 7.40 (d, 1 H, J = 16 Hz), 7.00 (d, 1 H, J = 16 Hz), 6.5–6.9 (m, 3 H), 6.15 (s, 1 H), 3.47 (s, 3 H), 3.33 (s, 3 H), 1.65 (s, 3 H); IR (KBr) 3400, 1650, 1600, 1490, 1460, 1350, 1300 cm^{-1}; FDMS, m/z 370 ($C_{20}H_{18}O_7$).

Anal. Calcd for $C_{20}H_{18}O_7$: C, 64.9; H, 4.9. Found: C, 64.8; H, 4.9.

Diels–Alder Cycloaddition Reactions of Isobenzofuran and *o*-Quinodimethane with 1,2-Diheteroethylenes

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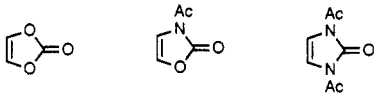
2,3-Diheterotetrahydronaphthalenes are produced efficiently and stereoselectively by the [4 + 2] cycloaddition of *o*-quinodimethane and isobenzofuran with various 1,2-diheteroethylenes.

Isobenzofuran (IBF)[1] and *o*-quinodimethane (QDM)[2] have found extensive use as reactive dienes in intermolecular and intramolecular Diels–Alder reactions for the rapid construction of polycyclic ring systems. The obvious synthetic utility of these highly reactive substrates has prompted research into new methods for their generation and novel usage in [4 + 2] cycloadditions.[3] Although examples of [4 + 2] cycloadditions of these dienes (IBF and QDM) with "electron-poor" and "electron-neutral" dienophiles are abundant, we have uncovered few examples employing "electron-rich" dienophiles, specifically 1,2-diheteroethylenes.[4]

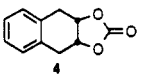
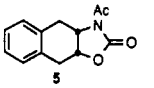
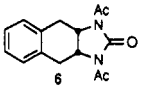
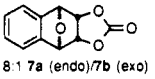
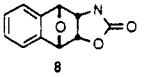
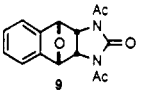
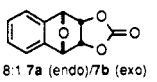
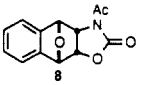
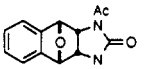
1,2-Diheteroethylenes such as vinylene carbonate (**1**),[5] 4-oxazolin-2-one (**2**),[6] and 1,3-diacetylimidazolin-2-one (**3**)[7]



1 **2** **3**

have been shown to undergo Diels–Alder cycloadditions with substrates such as cyclopentadiene (endo), hexa-

Table I

entry	dienophile	diene or precursor	conditions	product	yield
1			48h, 230°C, Toluene	 **4**	70%
2			48h, 230°C, Toluene	 **5**	90%
3			48h, 230°C, Toluene	 **6**	74%
4			60°C, PhH, 23h	 8:1 **7a** (endo)/**7b** (exo)	79%
5			60°C, PhH, 23h	 **8**	98%
6			100°C, PhH, 18h	 **9**	100%
7			xylenes, 135°, Ac_2O, 24h	 8:1 **7a** (endo)/**7b** (exo)	89%
8			xylenes, 135°, AcOH, 48h	 **8**	93%
9			xylenes, 135°, AcOH, 36h	 **9**	81%

(1) (a) Rodrigo, R. *Tetrahedron* **1988**, *44*, 2093. (b) Rickborn, B. *Advances in Theoretically Interesting Molecules*; Thummel, R. P., ed.; JAI Press Inc.: Greenwich, CN, 1989; Vol. I, pp 1–134. (c) Friedrichsen, W. *Adv. Heterocycl. Chem.* **1980**, *26*, 135. (d) Haddadin, M. J. *Heterocycles* **1978**, *9*, 865.

(2) For recent reviews, see: (a) Charlton, J. L.; Alaudin, M. M. *Tetrahedron* **1987**, *43*, 2873. (b) Wong, H. N. C.; Lau, K.-L.; Tam, K.-F. *Top. Curr. Chem.* **1986**, *133*, 85.

(3) (a) Naito, K.; Rickborn, B. *J. Org. Chem.* **1980**, *45*, 4061. (b) Jung, M. E.; Lam, Y.-S. P.; Mansuri, M.; Speltz, L. M. *J. Org. Chem.* **1985**, *50*, 1087. (c) Carre, M.-C.; Gregoire, B.; Caubere, P. *J. Org. Chem.* **1984**, *49*, 2050. (d) Meegalla, S. K.; Rodrigo, R. *Synthesis* **1989**, 942. (e) Choy, W.; Yang, H. *J. Org. Chem.* **1988**, *53*, 5796.

(4) For one isolated example, see: Newman, M. S. *J. Org. Chem.* **1961**, *26*, 2630.

(5) (a) Scharf, H.; Seidler, H. *Chem. Ber.* **1971**, *104*, 3030. (b) Scharf, H.; Kusters, W. *Chem. Ber.* **1972**, *105*, 564. (c) Russell, G.; Schmitt, K.; Mattox, J. *J. Am. Chem. Soc.* **1975**, *95*, 1882. (d) Hartmann, W. *Chem. Ber.* **1968**, *101*, 1643.

0022-3263/90/1955-4356$02.50/0

chloropentadiene (endo), 2,3-dimethylbutadiene, and anthracene, although high temperatures (110–180 °C) and an excess of diene are often required. Scholz et al. have demonstrated that in [4 + 2] cycloadditions with 1,3-cyclohexadiene, **1**, **2**, and **3** exhibit comparable dienophilicity.[6a,8] In contrast, Whitney has noted that in some cases **3** shows a greater reluctance to participate in normal (1,3-nonadiene) and inverse demand (3,6-di(2-pyridyl)-1,2,4,5-tetrazine) Diels–Alder reactions than either **1** or **2**.[7a] Our interest in the preparation of *cis*-2,3-diheterotetrahydronaphthalenes as pharmaceutical templates prompted our investigation into the cycloaddition chemistry of these dienophiles with IBF and QDM.

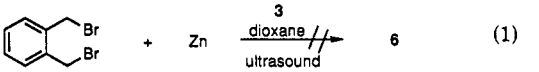
Results and Discussion

The results obtained in a series of Diels–Alder reactions utilizing dienophiles **1**, **2**, **3**, and the reactive dienes isobenzofuran and *o*-quinodimethane are shown in Table I. Isobenzofuran was either generated in situ from 1-ethoxy-1,3-dihydroisobenzofuran[9] (entries 7–9) or prepared in advance as a solution[10] (entries 4–6). These reactions involving IBF were typically run at concentrations of 0.05–0.10 M. *o*-Quinodimethane was generated in situ by heating benzocyclobutane as a 0.48 M solution in toluene at ca. 230 °C.[2] The cycloadducts were isolated in good to excellent yield without optimizing reaction conditions or isolation procedures. In all cases the diene and dienophile were used in a 1:1 ratio.

Compounds **7a** and **7b** (entries 4 and 7, Table I) were obtained in a ratio of ca. 8:1, respectively. The major isomer was determined to be the endo adduct **7a** (J = 2 Hz) and the minor isomer the exo adduct **7b** (J = 0 Hz) by comparison of the methine coupling constants with related cycloadducts.[9b,11] The stereoselectivity of this reaction seemed to be relatively insensitive to the reaction temperature (entry 4 vs entry 7). Interestingly, reactions of dienophiles **2** and **3** with isobenzofuran gave only one detectable (200-MHz ^{1}H NMR) stereoisomer under all reaction conditions.[12] Compounds **8** (bridgehead methines appear as doublets with J = 4 Hz) and **9** (bridgehead methines appear as an apparent triplet with J = 2 Hz) were assigned as endo adducts by comparison of the coupling constants observed in compounds **7a** and **7b**. The endo selectivity demonstrated in the [4 + 2] cycloaddition reactions of **1**, **2**, and **3** is in accord with previous results employing cyclopentadiene and hexachlorocyclopentadiene.[5,6,7a] The use of acetic acid in place of acetic anhydride as the "catalyst" in the reaction shown in entry 7 resulted in a slower conversion of starting materials to product as well as an appreciable amount of decomposition.

The temperatures employed in entries 4–6 were chosen on the basis that they produced the cycloadducts in high yield in a reasonable amount of time, but they do not represent the minimum temperature required to induce the [4 + 2] cycloaddition between these substrates. In fact, dienophiles **1**, **2**, and **3** will react with isobenzofuran (0.033 M) at room temperature albeit at a reduced rate (3 days vs 18–23 h) to give the corresponding cycloadducts accompanied by some decomposition of IBF (a 0.15 M solution of isobenzofuran has an approximate half-life of 6 days at room temperature[3a]). In a competition experiment run at a concentration of 0.033 M (23 °C, 72 h) employing 1 equiv of isobenzofuran and 1 equiv each of dienophiles **1**, **2**, and **3**, cycloadducts **8** and **9** were detected in a ratio of 8:1 by ^{1}H NMR. There was no indication of compounds **7a** or **7b**. Thus, in the case of isobenzofuran, the dienophilicity of these 1,2-diheteroethylenes is **3** > **2** > **1**.

One might expect that *o*-quinodimethane would also react at ambient temperatures with dienophiles **1**, **2**, and **3** if it was generated at this temperature by one of the presently available methods.[13] One such procedure involves in situ generation of *o*-quinodimethane at near ambient temperature by treatment of α,α'-dibromo-*o*-xylene with activated zinc under ultrasonic irradiation. It has been reported that in the presence of electron-deficient olefins the corresponding cycloadducts are obtained.[14] However, in the presence of the electron-rich dienophile **3**, our preliminary attempts to produce cycloadduct **6** were unsuccessful (eq 1).



(1)

In conclusion, it has been shown that isobenzofuran and *o*-quinodimethane undergo efficient and stereoselective [4 + 2] cycloadditions with 1,2-diheteroethylenes such as compounds **1**, **2**, and **3** to give 2,3-diheterotetrahydronaphthalenes. In light of the new methods for the preparation of variously substituted isobenzofurans and benzocyclobutanes, the work presented herein should enhance the breadth of reactions and target molecules available via the Diels–Alder chemistry of these dienes.

Experimental Section

General Procedures. ^{1}H NMR spectra were obtained on a 200-MHz Magnachem instrument and were recorded in $CDCl_3$ unless otherwise noted. Proton chemical shifts are given in ppm relative to Me_4Si (=0 ppm, ^{1}H). IR spectra were recorded on a Perkin-Elmer infrared spectrometer. Melting points are uncorrected. Mass spectra were obtained on MAT CH-5-DF (FAB) and Finnigan 8230 B (EI) mass spectrometers.

Elemental analyses were performed by The Upjohn Company and M-H W Laboratories, Phoenix, AZ.

The following chemicals were obtained from commercial sources and were used without further purification: vinylene carbonate (**1**), α,α'-dibromo-*o*-xylene (Aldrich), toluene, xylene, acetic anhydride (Fisher Scientific, certified A.C.S.), diethyl ether (Fisher, reagent A.C.S., anhydrous).

The following chemicals were prepared according to literature procedures: benzocyclobutane,[15] isobenzofuran,[3a] 3-acetyl-4-oxazolin-2-one (**2**),[16] 1-ethoxy-1,3-dihydroisobenzofuran,[17] 1,3-di-

(6) (a) Scholz, K.-H.; Heine, H.-G.; Hartmann, W. *Liebigs Ann. Chem.* **1977**, 2027. (b) Scholz, K.-H.; Heine, H.-G.; Hartmann, W. *Org. Synth.* **1984**, *62*, 149. (c) Deyrup, J. A.; Gingrich, H. L. *Tetrahedron Lett.* **1977**, *36*, 3115.

(7) (a) Whitney, R. *Tetrahedron Lett.* **1981**, *22*, 2063. (b) Szmuszkovicz, J., The Upjohn Company, unpublished results.

(8) In a competition experiment employing 1 equiv of 1,3-cyclohexadiene and 1 equiv each of dienophiles **1**, **2**, and **3** the corresponding cycloadducts were obtained in a ratio of 10:14.9:6.8, respectively, by Scholz et al.[6a]

(9) (a) For the use of acetic acid as catalyst, see: Makhlouf, M.; Rickborn, B. *J. Org. Chem.* **1981**, *46*, 2734. (b) For the use of acetic anhydride as catalyst, see: Fier, S.; Sullivan, R. W.; Rickborn, B. *J. Org. Chem.* **1988**, *53*, 2353. See ref 16 for the preparation of 1-ethoxy-1,3-dihydroisobenzofuran.

(10) Estimated molarity based on 70% conversion from starting material. See ref 3a for preparation of isobenzofuran.

(11) Rickborn, B.; Crump, S. L. *J. Org. Chem.* **1984**, *49*, 304.

(12) Isobenzofuran often gives mixtures of endo and exo adducts with typical dienophiles, see refs 1 and 3a.

(13) For the in situ generation of *o*-quinodimethane at near room temperature, see ref 2.

(14) Han, B. H.; Boudjouk, P. *J. Org. Chem.* **1982**, *47*, 751.

(15) Markgraf, J. H.; Basta, S. J.; Wege, P. M. *J. Org. Chem.* **1972**, *37*, 2361. Bubb, W. A.; Sternhell, S. *Aust. J. Chem.* **1976**, *29*, 1685.

(16) Rickborn, B.; Moss, R. *J. Org. Chem.* **1982**, *47*, 5391.

(17) (a) We thank Professor J. A. Deyrup at the University of Florida for his private communication describing the preparation of 3-acetyl-4-oxazolin-2-one (June, 1989). (b) For an alternate preparation, see ref 6b.

acetyl-4-imidazolin-2-one (3).[18]

General Procedure for the Reaction of Benzocyclobutane with Dienophiles 1, 2, and 3. Synthesis of Compound 4. A solution of benzocyclobutane (50 mg, 0.48 mmol) and vinylene carbonate (41 mg, 0.48 mmol) in 1 mL of toluene was heated in a pressure vessel at ca. 230 °C (external) for 48 h. The solvent was removed by rotary evaporation to give 64 mg (70%) of 4 as a yellow solid: mp (toluene/hexane) 150 °C; IR (film) 1770 (CO) cm^{-1}; ^1H NMR δ 2.88 (d, J = 17 Hz, 2 H), 3.13 (d, J = 17 Hz, 2 H), 5.18 (s, 2 H, OCH), 7.25 (m, 4 H, Ar H); MS (EI) m/e 190 [M^+]. Anal. Calcd for $C_{11}H_{10}O_3$: C, 69.46; H, 5.30. Found: C, 69.26; H, 5.45.

Reaction of Benzocyclobutane with 3-Acetyl-4-oxazolin-2-one (2). Synthesis of Compound 5. According to the procedure described above, benzocyclobutane (50 mg, 0.48 mmol) and 3-acetyl-4-oxazolin-2-one (55 mg, 0.48 mmol) gave 103 mg of a yellow oil. Recrystallization from toluene/hexane gave 94 mg (90%) of 5 as a white solid: mp 123–124 °C; ^1H NMR δ 2.45 (s, 3 H, CH_3), 2.99 (m, 2 H), 3.09 (m, 2 H), 4.79 (m, 1 H), 5.06 (m, 1 H), 7.22 (m, 4 H, Ar H); IR (film) 1775 (acetyl CO), 1700 (carbamate CO) cm^{-1}; MS (EI) m/e 231 [M^+]. Anal. Calcd for $C_{13}H_{13}NO_3$: C, 67.52; H, 5.67; N, 6.06. Found: C, 67.62; H, 5.71; N, 6.25.

Synthesis of Cycloadduct 6. As described above, benzocyclobutane (1.0 g, 9.6 mmol) and 1,3-diacetyl-4-imidazolin-2-one (3) (1.6 g, 9.6 mmol) in 20 mL of toluene gave 2.3 g (89%) of 6 as a yellow solid. The crude solid was recrystallized from hexane to give 1.93 g (74%) of 6 as a yellow crystalline solid: mp 186 °C; ^1H NMR δ 2.46 (s, 6 H, 2 × CH_3), 3.07 (m, 4 H, 2 × CH_2), 4.62 (m, 2 H, 2 × CH), 7.20 (m, 4 H, Ar H); IR (KBr) 1740 (acetyl CO), 1690 (urea CO) cm^{-1}; MS (EI) m/e 272 [M^+]. Anal. Calcd for $C_{15}H_{16}N_2O_3$: C, 66.16; H, 5.92; N, 10.29. Found: C, 66.11; H, 6.13; N, 10.39.

General Procedure for the Synthesis of 7 and 8 via Isobenzofuran. Synthesis of Cycloadduct 8. Isobenzofuran (1.50 mmol), generated as a solution in 15 mL of 4:1 benzene/cyclohexane, was combined with 3-acetyl-4-oxazolin-2-one (2) (169 mg, 1.50 mmol) and heated under a nitrogen atmosphere at 60 °C for 23 h. The cooled reaction mixture was concentrated in vacuo to give 359 mg (98%) of 8 as a tan solid: mp (toluene) 149 °C; ^1H NMR δ 2.18 (s, 3, CH_3), 4.76 (dd, J = 4, 9 Hz, 1, CH), 5.08 (dd, J = 4, 9 Hz, 1, CH), 5.58 (d, J = 4 Hz, 1, CH), 5.82 (d, J = 4 Hz, 1, CH), 7.35 (m, 4, Ar H); IR (film) 1780 (CO), 1700 (CO) cm^{-1}; MS (FAB) [M + H]$^+$ at m/z 246. Anal. Calcd for $C_{13}H_{11}NO_4$: C, 63.67; H, 4.52; N, 5.71. Found: C, 63.87; H, 4.35; N, 5.41.

Synthesis of 7a and 7b via Isobenzofuran. Following the procedure described below for the synthesis of 8 via isobenzofuran, vinylene carbonate (129 mg, 1.50 mmol) and isobenzofuran (1.50 mmol) gave 346 mg of a yellow solid (ca 8:1 **7a**/**7b** by ^1H NMR). Recrystallization of the crude product from toluene/pentane yielded 242 mg (79%) of **7a** and **7b** (8:1) as white needles: mp 182 °C; ^1H NMR (**7a**, endo) δ 5.16 (dd, J = 2, 3 Hz, 2, 2 × CH), 5.59 (dd, J = 2, 3 Hz, 2, 2 × CH), 7.45 (m, 4, Ar H); (**7b**, exo) δ 4.70 (s, 2, 2 × CH), 5.43 (s, 2, 2 × CH), 7.45 (m, 4, Ar H); IR (KBr) 1800 (CO) cm^{-1}; ^{13}C NMR (75 MHz, acetone-d_6) (**7a**) δ 154.67, 141.11, 128.94, 123.37, 81.11, 75.68; high resolution MS calcd for $C_{11}H_9O_4$ 205.0501, found 205.0502.

Preparation of 9 via Isobenzofuran. A 0.14 M solution of isobenzofuran in 4:1 benzene/cyclohexane (5 mL, 0.71 mmol) was combined with a solution of 1,3-diacetyl-4-imidazolin-2-one (3) (132 mg, 0.78 mmol) in 10 mL of toluene, and the mixture was heated at 100 °C for 18 h. The cooled reaction was diluted with 50 mL of ether and washed with saturated aqueous Na_2CO_3 (2 × 25 mL). The organic phase was dried ($MgSO_4$), filtered, and concentrated in vacuo to afford 241 mg (ca. 100%) of 9 as a brown oil. Crystals were obtained from hexane: mp 115 °C; ^1H NMR δ 2.26 (s, 6 H, 2 × CH_3), 4.64 (m, 2 H, 2 × CH), 5.86 (m, 2 H, 2 × OCH), 7.24 (m, 4 H, Ar H); MS (FAB) [M + H]$^+$ at m/e 287. Anal. Calcd for $C_{15}H_{14}N_2O_4$: C, 62.93; H, 4.93; N, 9.79. Found: C, 63.07; H, 5.07; N, 9.69.

Competition Experiment. Isobenzofuran (0.50 mmol), generated as a solution (0.033 M) in 15 mL of 4:1 benzene/cyclohexane, was combined with dienophiles 1 (43 mg, 0.50 mmol), 2 (57 mg, 0.50 mmol), and 3 (84 mg, 0.50 mmol) and stirred at room temperature under a nitrogen atmosphere for 72 h. The solvent was removed and the residue was analyzed by 200-MHz ^1H NMR. In addition to recovered starting materials 2 and 3, cycloadducts 8 and 9 were apparent in a ratio of 1:8, respectively; there was no indication of compounds **7a** or **7b**. In an analogous reaction employing only dienophiles 1 and 2, cycloadducts 7 (**7a** + **7b**) and 8 were produced in a ratio of ca. 1:2 according to ^1H NMR analysis.

General Procedure for the Synthesis of 7 and 8 via 1-Ethoxy-1,3-dihydroisobenzofuran. Synthesis of Compounds 7a and 7b. A solution of 1-ethoxy-1,3-dihydroisobenzofuran (251 mg, 1.53 mmol), vinylene carbonate (132 mg, 1.53 mmol), and acetic anhydride (156 mg, 1.53 mmol) in 15 mL of xylene was heated at reflux under nitrogen for 24 h. The solvent was removed in vacuo to give 277 mg (89%) of **7a** and **7b** as a white solid (ca 8:1). This material was 95% pure by ^1H NMR analysis containing 1-ethoxy-1,3-dihydroisobenzofuran as the only contaminate.

Synthesis of 8 via 1-Ethoxy-1,3-dihydroisobenzofuran. According to the procedure described above, substituting acetic acid (1 drop) for acetic anhydride and heating at reflux for 48 h, 349 mg (93%) of 8 was obtained as a yellow solid. This material was identical with that prepared above and contained ca. 5% of 1-ethoxy-1,3-dihydroisobenzofuran as the only contaminate.

Preparation of 9 via 1-Ethoxy-1,3-dihydroisobenzofuran. A solution of 1-ethoxy-1,3-dihydroisobenzofuran (656 mg, 4.00 mmol) and 1,3-diacetyl-4-imidazolin-2-one (3) (672 mg, 4.00 mmol) in 40 mL of xylenes was treated with 4 drops of glacial acetic acid and heated at 135 °C for 36 h. The solvent was removed in vacuo. The residue was dissolved in 60 mL of CH_2Cl_2 and washed with saturated aqueous $NaHCO_3$ (30 mL). The layers were separated, and the aqueous layer was back-extracted with 30 mL of CH_2Cl_2. The combined organic layers were dried ($MgSO_4$), filtered, and concentrated in vacuo to give 925 mg (81%) of 9 as a golden oil. This compound was identical (^1H NMR, mp) with that reported above.

(18) (a) Prepared by. R. Pedersen at The Upjohn Company. (b) For literature procedures, see: Haines, D. R.; Leonard, N. J.; Wiener, D. F. *J. Org. Chem.* **1982**, *47*, 474. Hilbert, G. E. *J. Am. Chem. Soc.* **1932**, *54*, 3413.

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