nitrogen atmosphere at room temperature, 1.3 mmol of $CsSO_4F$ was slowly added over a peroid of 5 min. The reaction mixture was then stirred at room temperature for an additional 1 h; the reaction was slightly exothermic. Methylene chloride (20 mL) was added, the insoluble precipitate filtered off, the filtrate washed with water, the organic layer dried over anhydrous Na_2SO_4, and the solvent evaporated in vacuo. The crude reaction mixture was analyzed by ^{19}F NMR and GLC. The product distributions stated in Table I and Scheme II represent the average of at least three experiments. Pure products were isolated by gas or thin-layer chromatography.

Reaction with (*E*)- and (*Z*)-Stilbene (1, 2). The following products were isolated by preparative TLC (SiO_2, petroleum ether:CH_2Cl_2 = 9:1). ***d,l-erythro*-1-Fluoro-2-methoxy-1,2-diphenylethane (3b)**: 19.5%; mp 51–52 °C (lit.[14] mp 52–54 °C); NMR $\delta(F) = -186$ (dd, $^2J_{FH_1} = 49.5$ Hz, $^3J_{FH_2} = 15$ Hz), $\delta(H_1) = 5.5$ (dd, $^3J_{H_1H_2} = 5.5$ Hz, 1 H), $\delta(H_2)$ 4.38 (dd, 1 H), $\delta(OCH_3) = 3.25$ (s, 3 H), $\delta(H) = 7.25$ (m, 10 H); mass spectrum, *m/e* 230 (M^+, 0.5%), 210 (0.5), 122 (11), 121 (100), 109 (5), 105 (5), 91 (6), 77 (30), 51 (3). ***d,l-threo*-1-Fluoro-2-methoxy-1,2-diphenylethane (4b)**: 46.5%; oily; NMR $\delta(F) = -181.5$ (dd, $^2J_{FH_1} = 49.5$ Hz, $^3J_{FH_2} = 13$ Hz), $\delta(H_1) = 5.4$ (dd, $^3J_{H_1H_2} = 8$ Hz, 1 H), $\delta(H_2) = 4.45$ (dd, 1 H), $\delta(OCH_3) = 3.35$ (s, 3 H), $\delta(H) = 7.2$ (m, 10 H); mass spectrum, *m/e* 230 (M^+, 0.5%), 210 (0.5), 122 (11), 121 (100), 109 (5), 105 (10), 91 (5), 77 (20), 51 (3).

Reaction with Acenaphthylene (5). The following products were isolated by preparative TLC (SiO_2, petroleum ether:$CHCl_3$ = 1:1). ***cis*-1-Fluoro-2-methoxyacenaphthene (6)**: 44.5%; mp 39–39.5 °C; NMR $\delta(F) = -186.7$ (dd, $^2J_{FH_1} = 54$ Hz, $^3J_{FH_2} = 12$ Hz), $\delta(H_1) = 6.1$ (dd, $^3J_{H_1H_2} = 5$ Hz, 1 H), $\delta(H_2) = 5.1$ (dd, 1 H), $\delta(OCH_3) = 3.6$ (s, 3 H), $\delta(H) = 7.4$–7.8 (m, 6 H); mass spectrum $C_{13}H_{11}OF$, calcd *m/e* 202.0794, found *m/e* 202.0799, *m/e* 203 (M^+ + 1, 14%), 202 (M^+, 100), 201 (14), 188 (10), 187 (68), 186 (12), 172 (14), 171 (70), 170 (31), 159 (52), 158 (20), 157 (21), 139 (15), 133 (20). ***trans*-1-Fluoro-2-methoxyacenaphthene (7)**: 42%; oily; NMR $\delta(F) = -173.2$ (dd, $^2J_{FH_1} = 56$ Hz, $^3J_{FH_2} = 21$ Hz), $\delta(H_1) = 6.2$ (d, 1 H), $\delta(H_2) = 5.25$ (d, 1 H), $\delta(OCH_3) = 3.55$ (s, 3 H), $\delta(H) = 7.4$–7.8 (m, 6 H); mass spectrum for $C_{13}H_{11}OF$, calcd *m/e* 202.0794, found *m/e* 202.0790, *m/e* 203 (M^+ + 1, 14 %), 202 (M^+, 100), 201 (10), 188 (9), 187 (65), 186 (12), 172 (11), 171 (60), 170 (30), 159 (40), 158 (7), 157 (12), 139 (9), 133 (16).

Reaction with Indene (8). The following products were separated by preparative TLC (SiO_2, petroleum ether:$CHCl_3$ = 1:1). ***cis*-1-Methoxy-2-fluoroindane (9)**: 42%; mp 37–37.5 °C; NMR $\delta(F) = -203$ (dddd, $^2J_{FH_2} = 54$ Hz, $^3J_{FH_3} = 27$ Hz, 24 Hz, $^3J_{FH_1} = 14$ Hz), $\delta(H_1) = 4.6$ (dd, $^3J_{H_1H_2} = 5$ Hz, 1 H), $\delta(H_2) = 5.4$ (dddd, $^3J_{H_2H_3} = 5$ Hz, 5 Hz, 1 H), $\delta(H_3) = 3.25$ (m, 2 H), $\delta(OCH_3) = 3.57$ (s, 3 H), $\delta(H) = 7.25$ (m, 4 H); mass spectrum for $C_{10}H_{11}OF$, calcd *m/e* 166.0794, found *m/e* 166.0795, *m/e* 167 (M^+ + 1, 10 %), 166 (M^+, 80), 165 (54), 136 (12), 135 (10), 134 (22), 133 (32), 131 (31), 115 (42), 103 (38). ***trans*-1-Methoxy-2-fluoroindane (10)**: 39%; oily NMR $\delta(F) = -183.5$ (dddd, $^2J_{FH_2} = 54$ Hz, $^3J_{FH_3} = 24$ Hz, 18 Hz, $^3J_{FH_1} = 18$ Hz), $\delta(H_1) = 4.8$ (dd, $^3J_{H_1H_2} = 3$ Hz, 1 H), $\delta(H_2) = 5.22$ (dddd, $^3J_{H_2H_3} = 5$ Hz, 3 Hz, 1 H), $\delta(H^3) = 3.12$ (m, 2 H), $\delta(OCH_3) = 3.54$ (s, 3 H), $\delta(H) = 7.2$ (m, 4 H); mass spectrum for $C_{10}H_{11}OF$, calcd *m/e* 166.0794, found *m/e* 166.0790, *m/e* 167 (M^+ + 1, 8%), 166 (M^+, 72), 165 (42), 136 (10), 135 (100), 134 (18), 133 (32), 131 (30), 115 (45), 103 (45).

Reaction with 1-Phenylind-1-ene (11). The following products were isolated by preparative TLC (SiO_2, petroleum ether:$CHCl_3$ = 1:1). ***r*-1-Phenyl-1-methoxy-*t*-2-fluoroindane (12)**: 25%; oily; NMR $\delta(F) = -200.8$ (ddd, $^2J_{FH_2} = 54$ Hz, $^3J_{FH_3} = 15$ Hz, 6 Hz), $\delta(H_2) = 5.0$ (ddd, $^3J_{H_2H_3} = 7$ Hz, 1 H), $\delta(H_3) = 3.2$ (m, 2 H), $\delta(OCH_3) = 3.18$ (s, 3 H), $\delta(H) = 7.3$ (m, 9 H); mass spectrum for $C_{16}H_{15}OF$, calcd *m/e* 242.1107, found *m/e* 242.1109, *m/e* 243 (M^+ + 1, 12%), 242 (M^+, 72), 212 (20), 211 (100), 210 (46), 209 (32), 207 (21), 192 (9), 191 (35), 189 (15), 183 (11), 179 (22), 178 (31), 165 (61), 133 (32), 105 (10), 77 (23). ***r*-1-Phenyl-1-methoxy-*c*-2-fluoroindane (13)**: 51.6%; mp 102–103 °C; NMR $\delta(F) = -182.2$ (ddd, $^2J_{FH_2} = 54$ Hz, $^3J_{FH_3} = 36$ Hz, 25.5 Hz), $\delta(H_2) = 5.0$ (dd, $^3J_{H_2H_3} = 4$ Hz, 1 H), $\delta(H_3) = 3.3$ (m, 2 H), $\delta(OCH_3) = 3.0$ (s, 3 H), $\delta(H) = 7.3$ (m, 9 H); mass spectrum for $C_{16}H_{15}OF$, calcd *m/e* 242.1107, found *m/e* 242.1102, *m/e* 243 (M^+ + 1, 10 %), 242 (M^+, 60), 212 (16), 211 (100), 210 (25), 209 (21), 207 (16), 192 (7), 191 (27), 189 (11), 179 (17), 178 (25), 165 (48), 133 (26), 105 (8), 77 (18).

Registry No. 1, 103-30-0; 2, 645-49-8; 3a, 14090-31-4; (±)-3b, 52795-54-7; (±)-4a, 52855-20-6; (±)-4b, 52776-26-8; 5, 208-96-8; (±)-6, 106268-81-9; (±)-7, 106295-73-2; 8, 95-13-6; (±)-9, 106268-82-0; (±)-10, 106268-83-1; 11, 1961-97-3; (±)-12, 106268-84-2; (±)-13, 106268-85-3; HF, 7664-39-3; $CsSO_4F$, 70806-67-6.

HZSM-5-Catalyzed Dihydroxybenzene Equilibration†

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The equilibrations of disubstituted benzenes are important reactions for both practical and theoretical reasons. Billions of pounds of xylenes are equilibrated each year to provide the para isomer for polyester manufacture.[1] While not as important commercially, other disubstituted benzenes have been equilibrated by a variety of techniques.

Olah used $AlCl_3/H_2O$ to equilibrate $C_6H_4X_2$ with X = Cl[2a] and Br[2b] in liquid-phase reactions. Xylenes undergo both isomerization and disproportionation with $AlCl_3$, and the relative rate constants for the isomerization were determined.[3] Difluorobenzenes do not equilibrate with acid catalysts,[2a] but they do thermally scramble above 1000 °C.[4] The three-component equilibrium has not been experimentally achieved. Terphenyls have been equilibrated, but the analytical methods of the day were not able to quantitatively analyze all three components.[5]

Substituted benzenes with two different substituents, one of which is OH, have also been equilibrated. Cresols and xylenols probably equilibrate by CH_3 and not by OH migration;[6] hydroxybiphenyls probably equilibrate by phenyl migration.[7]

There are two competing mechanisms in methylbenzene exchange. HZSM-5 catalyzes both the intermolecular methyl exchange which disproportionates toluene to benzene and *p*-xylene and also the intramolecular exchange which equilibrates the three xylene isomers with essentially no disproportionation.

In our initial experiments looking for OH exchange, we sought to maximize our possibility for success by assuming that either mechanism might occur. Thus mixtures of dihydroxybenzene, phenol, and catalyst were studied. Phenol was introduced as a benzene solution for ease of handling.

We have now found that the zeolite[8] H-ZSM5 catalyzes the equilibration of dihydroxybenzenes. Figure 1 shows a composition diagram of the paths taken by each pure isomer toward equilibrium. When we initially found the ortho–para exchange, which is usually characteristic of an intermolecular process, we continued to leave phenol in the reaction recipe. Only toward the end of this study was the control run without phenol. The results were independent of the presence of phenol which suggests an intramolecular process for the OH exchange.

The reaction is so slow that equilibrium could not be achieved in convenient times starting from each pure component. Therefore mixtures of 1,2- and 1,4-dihydroxybenzenes were treated with HZSM-5. Each new product composition was used as the starting point for the next experiment—converging on the equilibrium of 6%

† Contribution No. 3891.

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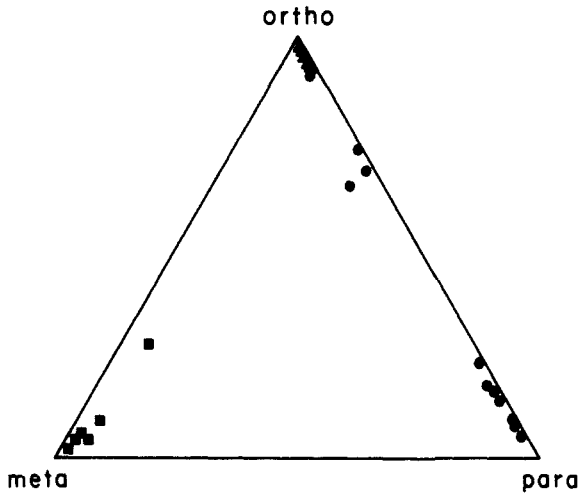


Figure 1. Dihydroxybenzene equilibration.

Table I. Kinetic Model for Dihydroxybenzene Equilibration

$1,2\text{-}C_6H_4(OH)_2 = 1,3\text{-}C_6H_4(OH)_2$ (1)
$1,2\text{-}C_6H_4(OH)_2 = 1,4\text{-}C_6H_4(OH)_2$ (2)
$1,3\text{-}C_6H_4(OH)_2 = 1,4\text{-}C_6H_4(OH)_2$ (3)

1,4- and 94% 1,2-dihydroxybenzenes.[9] The points represent separate experiments with different amounts of catalyst, temperatures, and reaction times. Other acidic zeolites such as HY do not catalyze this reaction.

We have not been able to determine the three-component equilibrium because the involvement of the 1,3-isomer is too slow. All we can say is the equilibrium 1,3-content is less than 70%.[9]

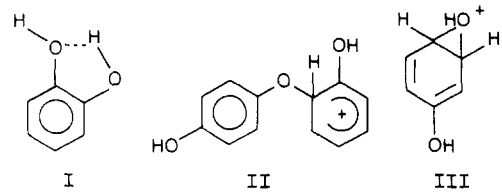
A severe side reaction is dehydroxylation, giving phenol. Other compounds seen include such acid-catalyzed phenolic condensation products as diphenyl ether, dibenzofuran, and several isomers of hydroxydiphenyl ether.

The yields of dihydroxybenzene isomers are capricious and the reaction is not synthetically useful with the catalysts identified up to now.

Although no conventional labeling studies have been done, some experiments with naphthols are relevant in this context. Solutions of either α- or β-naphthol in benzene were heated with HZSM-5 under conditions similar to those used in the dihydroxybenzene isomerizations. Although the equilibrium could not be established because this reaction is even slower than the dihydroxybenzene equilibration, each isomer produced the other and the equilibrium constant is close to one.

If the naphthalene ring remains intact during naphthol equilibration, then probably so does the benzene ring during dihydroxybenzene equilibration.

Several conclusions can be made on the basis of the facts as represented in Figure 1. The two-component equilibrium strongly favors the 1,2-isomer. This effect arises from the hydrogen-bonding stabilization in the 1,2-isomer I and



I II III

has been predicted theoretically.[10] The simplest kinetic model to equilibrate these species is three first-order, reversible reactions as shown in Table I. Equation 2 is fast compared to the other two because the 1,2- and 1,4-isomers essentially completely equilibrate while forming only small amounts of 1,3.

The slow isomerization of the 1,3-isomer initially favors the 1,4-isomer even though equilibrium highly favors the 1,2-isomer. The ratio of the relative rates of 1,4- vs. 1,2-dihydroxybenzene formation appears to be similar to those of electrophilic hydroxylation of phenol by OH^+, for example, from hydrogen peroxide in the presence of related catalysts, or under photolytic conditions.[11]

The kinetic path is not consistent with the simple 1,2-shift mechanism by which $AlCl_3$ isomerizes dichlorobenzenes[2a] or xylenes.[3] In this mechanism there is no direct path between 1,2- and 1,4-isomers and the initial directions of both are toward 1,3. As shown in Figure 1 the dominant path directly interconverts 1,2 and 1,4.

The kinetics do not allow the intermediacy of free phenol if there is a single isomerization mechanism. If free phenol were involved, the paths from each of the three vertices to the three-component equilibrium would be straight lines. The path from 1,3-dihydroxybenzene is definitely curved, but it is difficult to conclude anything definitive from the 1,2- and 1,4-isomerizations. We conclude that phenol is a byproduct formed in an irreversible reaction. Hydroxyl exchange via diphenyl ether intermediates II analogous to the diphenylmethane intermediates invoked in the intermolecular exchange of toluene to benzene and *p*-xylene[12] would seem to be unlikely because of the small pore size of HZSM-5.[13]

The NIH mechanism, in which phenols isomerize by the hydroxyl group "walking" around the ring via protonated epoxide intermediates such as III,[14] is consistent with the data in Figure 1 only if ring opening to the 1,3-isomer is slow. Hydroxylation of toluene does give mostly 1,2- and 1,4-cresol isomers via epoxide intermediates. Epoxide ring opening to 1,3-cresol is disfavored.[14]

There is no evidence for mechanisms involving zeolite peroxides as intermediates, although a concise catalyst cycle can be written.

The acid sites of HZSM-5 are among the strongest known.[15] Therefore, if this reaction requires these extremely strong acid sites, the failure of less acidic zeolites such as HY to catalyze the reaction may be understood.

Experimental Section

A mixture of 200 mg of dihydroxybenzene, 1 g of phenol, 2 mL of benzene, and 200 mg of catalyst was heated in a shaker tube, typically, 350 °C/2 h. The tube was rinsed with acetonitrile and

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the recovered liquid treated with *N,O*-bis[(trifluoromethyl)silyl]trifluoroacetamide to convert all the aromatic phenols to their TMS derivatives. The products were analyzed by GC on a 50-ft methylsilicone capillary column at 150 °C. The three dihydroxybenzenes elute in the order 1,2 < 1,3 < 1,4.

Registry No. 2-HOC_6H_4OH, 120-80-9; 4-HOC_6H_4OH, 123-31-9; 3-HOC_6H_4OH, 108-46-3; C_6H_5OH, 108-95-2; $C_6H_5OC_6H_5$, 101-84-8; $HOC_6H_4OC_6H_5$, 54774-79-7; dibenzofuran, 132-64-9; α-naphthol, 90-15-3; β-naphthol, 135-19-3.

Regioselective Alkylations of 4,6-Dialkyl-Substituted 2*H*-Pyran-2-ones

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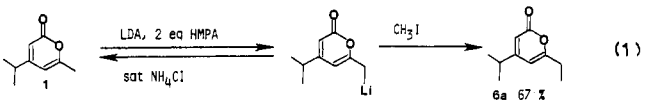
Recently, we reported[1] a versatile synthesis of α-pyrones[2] from vinylogous thiol esters which were readily prepared[3] by reaction of α-oxo ketene dithioacetals[4] with organocuprates. The alkyl substitution pattern at all four olefinic carbon atoms of the pyrone ring could be altered simply by choice of starting α-oxo ketene dithioacetal, organocuprate, or ester enolate anion. In addition, the method permitted annulation of the pyrone ring system onto an existing cycloalkanone framework. In an effort to functionalize these readily prepared alkyl-substituted α-pyrones, we examined the kinetic deprotonation of these substrates and the subsequent alkylation of the resonance stabilized carbanions. We now report that deprotonation can be effected at the 4- and 6-alkyl substituents by the use of lithium diisopropylamide and that good to excellent regioselectivity can be achieved in the alkylation reactions.

Although deprotonation of alkyl-substituted α-pyrones has been described by several groups, low yields and severe limitations have been noted.[5-7] These limitations generally involve the instability of the pyrone ring system under the reaction conditions. The general origin of the instability appears to be ring-opening reactions followed by subsequent base-catalyzed transformations.[8] Despite these limitations Harris has achieved some success with alkylations at the C6-methyl substituent of polyanions generated from 4-hydroxy-2-pyrones in yields ranging from 30% to 72%.[6] Deprotonation of the 4-hydroxy substituent in these compounds may protect the ring from nucleophilic cleavage reactions. An alternative approach involved the use of a phosphonium ylide generated from 4-methoxy-6-bromomethyl-2*H*-pyran-2-one.[9] 4-Methoxy-6-methyl-2*H*-pyran-2-one, however, has been reported to undergo deprotonation in the ring via an ortho-lithiation process, and the resulting vinyl anion has been successfully quenched with chlorotrimethylsilane.[10] 3-Bromo-2-pyrone could not be converted into the lithium derivative but did afford an organocopper species upon reaction with lithium dimethylcuprate.[11]

In an initial experiment, addition of 4-isopropyl-6-methyl-2*H*-pyran-2-one (1) to a cold (−78 °C) solution of lithium diisopropylamide (LDA) and HMPA gave the 6-ethyl derivative in 67% yield after quenching with methyl iodide (eq 1). The stability of the intermediate



(1)

carbanion was examined as a function of time and temperature by treating the carbanion containing solution with acid and measuring the yield of recovered α-pyrone. In marked contrast to vinyl anions generated from α-pyrones,[10] the resonance-stabilized anion generated from **1** was relatively stable and the α-pyrone was recovered in 83% yield upon quenching the anion after 2 h at 20 °C. Similar treatment of several alkyl-substituted α-pyrones with LDA and 2 equiv of HMPA in THF generated the resonance-stabilized carbanions, which could be alkylated with alkyl iodides, aldehydes, and acid chlorides in good to excellent yields (Table I). Alkylation could not be effected with benzyl bromide, epoxides, or enoates; very low yields of the 6-bromo derivative **6e** were achieved with molecular bromine. Interestingly, ethyl bromide did not afford the alkylation product, while ethyl iodide did, in 68% yield (entry 3). This result suggests the possibility that these alkylation reactions may proceed by an electron-transfer pathway,[12] although it may also merely reflect relative reactivities of carbanion and electrophile. The yield of alkylation product obtained with benzoyl chloride could be increased by utilizing 2 equiv of the reagent (entries 5-6). This observation is consistent with a moderately reactive, resonance-stabilized carbanion where product deprotonation by pyrone carbanion becomes competitive when 1 equiv of acid chloride is employed.

The regioselectivity of carbanion formation showed a preference for deprotonation at the C4-alkyl substituent. Deprotonation and alkylation of **4** gave **9** and **10** as a 73:27 mixture (entry 14). Analysis of the mass and 200-MHz NMR spectra of the mixture permitted structure assignments. The major isomer **9** clearly shows an isopropyl substituent [δ 1.17 (d, J = 6.8 Hz, 6 H), 2.82 (sept, J = 6.8 Hz, 1 H)] and a methyl resonance at δ 2.25 consistent with the 6-methyl resonances found in **1** and **4** (δ 2.25 and 2.23, respectively). The minor isomer **10** displays two methylene absorptions at δ 2.44 (qd, J = 1.08 Hz, J = 7.39 Hz, 2 H) and 2.56 (q, J = 7.52 Hz, 2 H); the quartet of doublets is similar to that observed for the methylene group in **4**. The mass spectrum shows an intense peak at m/e 123 for sequential loss of CO and the C6-Me for the major isomer and a small peak at m/e 109 for the similar fragmentation of the minor isomer. A small peak at m/e 137 corresponds

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