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Conversion of 2-Alkylcinnamaldehydes to 2-Alkylindanones via a Catalytic Intramolecular Friedel−Crafts Reaction

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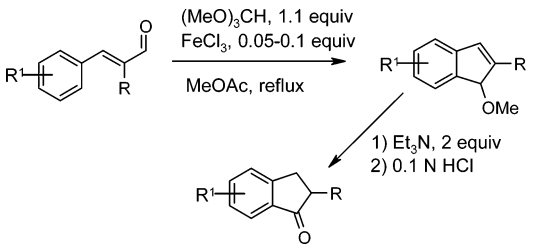

The preparation of indanones by the intramolecular acylation of 3-arylpropanoic acids or halides requires the use of non-catalytic acid promoters. In the presence of 5−10 mol % $FeCl_3$, in situ generated dimethyl acetals of (*E*)-2-alkylcinnamaldehydes cyclize to 1-methoxy-2-alkyl-1H-indenes in good-to-high yields. The 1-methoxyindenes were converted in high yield into 2-alkylindanones by treatment with triethylamine, to effect isomerization to the isomeric enol ethers, followed by acid-catalyzed hydrolysis. Thus, a catalytic, intramolecular Friedel−Crafts reaction leading to 2-alkylindanones from 2-alkylcinnamaldehydes was developed.

Intramolecular Friedel−Crafts acylation of 3-arylpropanoic acids[1] and 3-arylpropanoyl halides,[2] or intramolecular alkylation using 1-aryl-2-propen-1-ones,[3] are well-established methods for the direct preparation of 1-indanones.[4] In all cases, however, these transformations generally require the use of at least an equivalent amount of either a Lewis or Brønsted acid to be effective, and the reaction mixtures often must be heated. These methods are extremely harsh and generate large amounts of acidic waste for disposal. Because of these concerns, alternative methods for the preparation of indanones that avoid strongly acidic conditions, such as transition metal-catalyzed carbocyclizations[5a−c] and isomerization of arylpropargyl alcohols,[5d−f] continue to be of interest. Another approach is the development of a catalytic version of the intramolecular electrophilic substitution reaction. Indanones were obtained by treating 3-arylpropanoic acids with 5−20 mol % $Tb(OTf)_3$ at 250 °C.[6] Opting to modify the substrate to achieve milder reaction conditions, Fillion et al. found that catalytic quantities of some metal triflates were effective at cyclizing 5-benzyl derivatives of Meldrum's acid to yield 1-indanones.[7] The intramolecular acylation, which liberates acetone and carbon dioxide, was performed using 10 mol % $Sc(OTf)_3$ in nitromethane at 100 °C.

Recently, Yonezawa et al.[8] and we[9] have reported that the acetals of 2-alkylcinnamaldehydes will cyclize in the presence of acid catalysts to yield 1-alkoxy-2-alkyl-1H-indenes (**2**). Yonezawa reported the isolation of the indenyl ethers in moderate yields after treating (*E*)-2-alkylcinnamaldehydes (**1**) with 4 equiv of trimethyl orthoformate and 1 equiv of $BF_3{\cdot}OEt_2$ for 3 h at 25 °C (Scheme 1). The reaction also occurred when starting with the dimethyl acetal of **1a**, and a mechanism was proposed in which intramolecular electrophilic substitution proceeds through an allylic oxocarbenium ion **4**. We have found that this cyclization can be accomplished with catalytic quantities of $FeCl_3$ and that the resulting indenyl ethers can be converted in high yields into 2-alkylindanones. In addition to achieving a catalytic, intramolecular Friedel−Crafts reaction leading to indanones, the process uses the more environmentally benign reaction conditions of $FeCl_3$ as the catalyst and methyl acetate as the solvent.

Table 1 presents selected optimization experiments using **1a** as the substrate. At 40 mol % $FeCl_3$ in dichloromethane (DCM), **1a** was consumed within 1 h at rt in the presence of 1.1 equiv

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SCHEME 1. Reported Preparation of 1-Alkoxy-2-alkylindenes[8a]

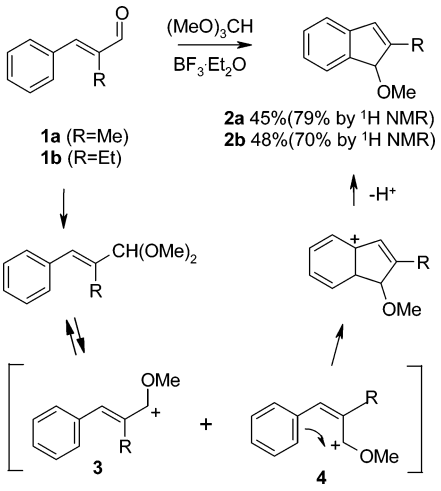
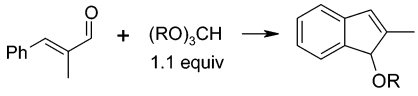


TABLE 1. $FeCl_3$-Catalyzed Formation of 2a and 5a from 1a



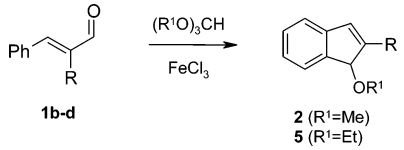
entry	R	mol % $FeCl_3$, solvent, temp, time (h)	yield (%)[a]
1	Me	40, DCM, rt, 1	78
2	Me	40, DCM, rt, 24	77
3	Me	20, DCM, rt, 24	72
4	Me	10, DCM, reflux, 4	79
5	Me	10, MeOAc, reflux, 3	89
6	Me	5, MeOAc, reflux, 8	85[b]
7	Et	40, DCM, rt, 1	45
8	Et	15, DCM, reflux, 7	40
9	Et	15, EtOAc, 70 °C, 7	69

[a] Isolated by bulb-to-bulb distillation of reaction mixture. [b] Water added prior to work up to hydrolyze remaining acetal, **2a**/**1a** = 97:3 by ^{1}H NMR.

of trimethyl orthoformate yielding **2a** in 78% (entry 1). Reducing the catalyst level to 20 mol % resulted in a slower reaction, but complete consumption of **1a** was achieved within 24 h (72% yield, entry 3). Reducing the catalyst level to 10 mol % necessitated heating the reaction mixture at reflux to consume all of **1a** and afforded **2a** in 79% yield after 4 h (entry 4). Using these conditions with MeOAc as the solvent resulted in an improved yield of 89% (entry 5). With MeOAc as solvent and a catalyst level of 5 mol %, not quite all of **1a** was consumed after 8 h at reflux, but still, **2a** (containing 3 mol % **1a**) was obtained in 85% yield after bulb-to-bulb distillation of the reaction mixture (entry 6).

Treating **1a** with triethyl orthoformate using the conditions of Table 1, entry 1 produced 1-ethoxy-2-methylindene **5a** in 45% yield (Table 1, entry 7), much lower than the 78% yield obtained for methoxy analog **2a** (entry 1). The reason for this yield difference does not appear to be due to increased sensitivity of **5a** to acid-catalyzed decomposition, since both **2a** and **5a** decomposed to similar degrees, 87 and 84% losses, respectively, after 6 h at rt when treated with 40 mol % $FeCl_3$ in the absence of trialkyl orthoformate (DCM, GC with dodecane internal standard). Stability of the alkoxyindenes under the preparative reaction conditions is probably due to catalyst deactivation with

TABLE 2. Preparation of 2b−e and 5b[a]



entry	R	enal (*E*/*Z*)	product	yield (%)
1	Et	**1b** (96:4)	**2b**	93
2	Pr	**1c** (96:4)	**2c**	92
3	Bu	**1d** (95:5)	**2d**	95
4	Pentyl	**1e** (92:8)	**2e**	90
5[b]	Bu	**1d**	**5b**	96

[a] 1.1 equiv $(MeO)_3CH$, 0.05 equiv $FeCl_3$, refluxing MeOAc, 3−4 h. [b] 1.1 equiv $(EtO)_3CH$, 0.05 equiv $FeCl_3$, refluxing DCM, 4 h.

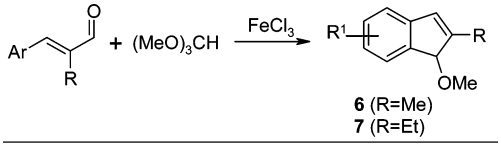
the liberation of alcohols as cyclization proceeds. A possible explanation for the yield difference is a slower rate of cyclization of the diethyl acetal, which allows for increased degradative side reactions prior to indene formation. Attempts to prepare **5a** with 10 mol % catalyst in refluxing DCM failed to achieve complete consumption of **1a** and necessitated the use of 15 mol % catalyst which afforded **5a** in 40% yield (entry 8). The yield was improved to 69% using EtOAc as the solvent (entry 9) but was still lower than the 89% yield obtained for **2a** using similar reaction conditions (entry 5).

The cyclization of other 2-alkylcinnamaldehydes (**1b**−**e**) was more efficient than that of **1a** since 5 mol % catalyst was sufficient to completely consume the enals within 4 h and resulted in high isolated yields (90−96%) of the 1-alkoxy-2-alkylindenes (Table 2). In the case of 2-butylcinnamaldehyde (**1d**), a further reduction in catalyst level to 1 mol % resulted in incomplete reaction after 8 h at reflux (ratio **2d**/**1d** = 68:32, by GC). In contrast to the results obtained with **1a**, **1d** cyclized in high yield with either trimethyl or triethyl orthoformate, resulting in 95 and 96% yields, respectively (entries 3 and 5). The higher cyclization efficiency of **1b**−**e** probably results from the larger 2-alkyl groups sterically preventing competitive decomposition reactions that would lead to lower yields and catalyst deactivation.

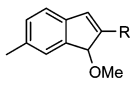
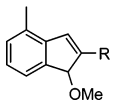
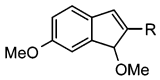
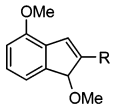
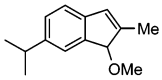
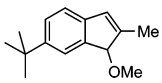
Cyclization also was achieved with either alkyl or methoxy group substituents on the aromatic ring (Table 3). As in the case of the simple 2-alkylcinnamaldehydes, when the 2-alkyl group was varied from Me to Et, higher yields of the indenyl ethers were isolated and a lower catalyst level (5 vs 10 mol %) could be employed. This was especially evident for the *para*-methoxy substituted enals **12** and **13**, in which only trace amounts of indenyl ether **6c** were detected (GC/MS) starting from **12** while the 2-ethyl analog **13** resulted in an 82% yield of **7c** (entries 5 and 6). Attempted bulb-to-bulb distillation of the **6c** reaction mixture did not yield any distillate from the black polymeric residue. Both *para* and *ortho* EDGs in enals **8**−**17** are expected to help stabilize intermediate carbenium ions **3** and **4**. Despite this, the yields of 2-methylindenyl ethers **6** were lower than the 89% yield for **2a**. Thus, inductive effects that help stabilize **3** and **4** also promote reactions leading to degradation products unless sterically restricted by the substituent at C(2).

Indene formation failed to occur using (*E*)-cinnamaldehyde as the substrate (conditions of Table 1, entry 5). GC/MS analysis indicated formation of the dimethyl acetal of benzaldehyde and higher molecular weight products but no alkoxyindene. Cyclization also failed to occur for enals **1** in the absence of trialkyl

TABLE 3. Indenyl Ethers with Substituents on the Aromatic Ring[a]



6 (R=Me)
7 (R=Et)

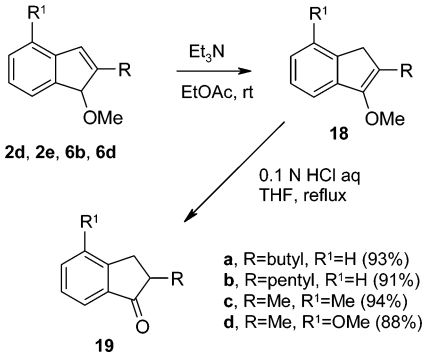
entry	enal	product		yield (%)
1	**8**		**6a**	82
2	**9**		**7a**	92
3	**10**		**6b**	83
4	**11**		**7b**	96
5	**12**		**6c**	0
6	**13**		**7c**	82
7	**14**		**6d**	56
8	**15**		**7d**	80
9	**16**		**6e**	72
10	**17**		**6f**	65

[a] Reaction conditions: 1.1 equiv $(MeO)_3CH$, 10 mol % $FeCl_3$ (R = Me) or 5 mol % $FeCl_3$ (R = Et), refluxing MeOAc, 3−4 h.

orthoformate. There was no measurable loss of **1d** after 8 h when treated with 5 mol % $FeCl_3$ in refluxing MeOAc (GC, dodecane as internal standard). These observations are consistent with those reported by Yonezawa et al.[8a] and show that a 2-alkyl group in the enal and acetal formation are both necessary for cyclization to occur. Acid-catalyzed cyclization to indenols has been reported for a few 3-aryl-2-alkyl-2-propenals with electron-rich aryl groups,[10] but there are no reports of the direct formation of indenols from **1**.[11]

The migration of the indene double bond is known to occur in the presence of basic catalysts.[12] For indenes bearing a substituent on C(1), an equilibrium mixture is established favoring the product with the more substituted double bond.

SCHEME 2. Preparation of 2-Alkylindanones



Examples include indenes with alkyl, alkoxy, acetoxy, and hydroxy subsituents. With a hydroxy substituent, the resulting enol yields the corresponding indanone.[13] For indenyl ethers **2**, **6**, and **7**, such an isomerization would yield the enol ethers **18** that could then be hydrolyzed to the 2-alkylindanones **19**. We tested this possibility by treating **2d** with 2 equiv of triethylamine at room temperature. After 3 days, **2d** was consumed and **18a** was isolated by bulb-to-bulb distillation in 98% yield. Acid-catalyzed hydrolysis of the enol ether yielded 2-butylindanone (**19a**) in 95% yield (93% yield from **2d**). In a like manner, other 2-alkylindanones also were isolated in high overall yields from alkoxyindenes (Scheme 2).

In conclusion, we have shown that (*E*)-2-alkylcinnamaldehydes can be cyclized to 1-alkoxy-2-alkylindenes in the presence of trialkyl orthoformate (1.1 equiv) and catalytic quantities of $FeCl_3$ (5−10 mol %). Activation of the aryl group by means of EDGs is not necessary for the intramolecular Friedel−Crafts reaction to proceed. The resulting 1-alkoxyindenes can be converted to 2-alkylindanones in high yield after a base-promoted isomerization to enol ethers **18** followed by acid-catalyzed hydrolysis. 2-Alkylindanone synthesis is achieved starting from 2-alkylcinnamaldehydes, readily available from aldol condensation of enolizable aldehydes with arylaldehydes.[14] In contrast, a more traditional preparation of such indanones from 2-alkylcinnamaldehydes would require oxidation and hydrogenation of the enal to the 3-arylpropanoic acids followed by intramolecular acylation using excess amounts of Brønsted acids.

Experimental Section

General Procedure for the Preparation of Indenyl Ethers. 2-Alkylcinnamaldehyde (100 mmol), trimethyl orthoformate (110 mmol), and $FeCl_3$ (5 mmol, 10 mmol for 2-methylcinnamaldehydes) were mixed in refluxing MeOAc (100−200 mL) until all enal was consumed (typically 2−6 h). The reaction mixture was diluted with diethyl ether (200 mL) and washed with water (3 × 100 mL). The organic phase was dried ($MgSO_4$), filtered, and concentrated. Indenyl ethers **2**, **6**, and **7** were isolated as pale-yellow oils (**6d** crystalline solid) by bulb-to-bulb distillation. **6f** was isolated by column chromatography on silica gel followed by bulb-to-bulb distillation.

1-Methoxy-2-butyl-1H-indene (2d). A mixture of **1d** (18.8 g, 100 mmol), trimethyl orthoformate (11.7 g, 110 mmol) and $FeCl_3$ (0.81 g, 5 mmol) in 200 mL of MeOAc was heated at reflux for 2.5 h. After work up, bulb-to-bulb distillation (bp 85−90 °C, 0.019

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mmHg) afforded 19.2 g (95 mmol, 95% yield) of **2d** as a pale-yellow oil: ^{1}H NMR (400 MHz, $CDCl_3$): δ 0.95 (t, J = 7.2 Hz, 3H), 1.36−1.46 (m, 2H), 1.51−1.70 (m, 2H), 2.29−2.43 (m, 2H), 3.02 (s, 3H), 4.93 (s, 1H), 6.43 (s, 1H), 7.11 (t, J = 7.2 Hz, 1H), 7.14 (d, J = 7.2 Hz, 1H), 7.22 (t, J = 7.2 Hz, 1H), 7.41 (d, J = 7.2 Hz, 1H); ^{13}C NMR (100 MHz, $CDCl_3$): δ 14.0 (q), 22.7 (t), 28.1 (t), 30.4 (t), 51.8 (q), 83.8 (d), 120.3 (d), 123.8 (d), 124.6 (d), 127.4 (d), 128.4 (d), 141.8 (s), 143.8 (s), 150.7 (s); IR (film): 2958, 2931, 1618, 1464, 1321, 1202, 1106, 1079, 948, 898 cm^{-1}; MS (*m*/*z*): 202 (M^+, 17), 159 (100), 145 (18), 141 (5), 129 (19), 128 (21), 115 (15), 102 (4), 91 (5); HRMS calcd for $C_{14}H_{18}O$: 202.1358, found: 202.1356.

General Procedure for the Preparation of 2-Alkylindanones 19. In a typical procedure, 3 g of indenyl ether **2** or **6** were mixed with 2 equiv of triethylamine and 25 mL of EtOAc. The mixture was monitored by gas chromatography until the ratio of the enol ether to starting material was greater than 99:1 (3−5 days). The mixture was concentrated to remove solvent and subjected to bulb-to-bulb distillation to first remove triethylamine (50−60 °C oven temperature, 2.2 mmHg) and then enol ether **18**. **18** was mixed with THF (20 mL) and 0.1 N HCl (40 mL), and the mixture was heated (75 °C) for 4 h. The mixture was diluted with ethyl ether and the aqueous phase separated. The organic phase was washed with water, dried ($MgSO_4$), filtered, and concentrated. Bulb-to-bulb distillation yielded the 2-alkylindanones **19**.

2-Butylindanone (19a).[15] From 25.3 g (0.125 m) of **2d**, 24.8 g (0.123 m, 98% yield) of **18a** were isolated by bulb-to-bulb distillation (bp 84−94 °C, 0.02 mmHg). 3 g (14.9 mmol) of **18a** were hydrolyzed to yield 2.65 g (14.1 mmol, 95% yield) of **19a** by bulb-to-bulb distillation (bp 120 °C, 0.02 mmHg) as a pale-yellow oil: ^{1}H NMR (400 MHz, $CDCl_3$): δ 0.92 (t, J = 7.2 Hz, 3H), 1.30−1.50 (m, 5H), 1.90−2.00 (m, 1H), 2.61−2.67 (m, 1H), 2.81 (dd, J = 17.1, 3.8 Hz, 1H), 3.31 (dd, J = 17.1, 7.9 Hz, 1H), 7.34 (t, J = 7.6 Hz, 1H), 7.44 (d, J = 7.6 Hz, 1H), 7.57 (t, J = 7.6 Hz, 1H), 7.74 (d, J = 7.6 Hz, 1H); ^{13}C NMR (100 MHz, $CDCl_3$): δ 14.0 (q), 22.7 (t), 29.6 (t), 31.2 (t), 32.9 (t), 47.4 (d), 123.8 (d), 126.6 (d), 127.3 (d), 134.6 (d), 136.9 (s), 153.8 (s), 209.1 (s); IR (film): 2958, 2931, 2858, 1713, 1610, 1465, 1287 cm^{-1}; MS (*m*/*z*): 188 (2), 145 (13), 133 (10), 132 (100), 131 (16), 115 (11); HRMS calcd for $C_{13}H_{16}O$: 188.1201, found: 188.1209.

Supporting Information Available: Experimental procedures, spectral data, and ^{1}H NMR and ^{13}C NMR spectra of products **2**, **5**, **6**, **7**, **18a**, and **19** are given. This material is available free of charge via the Internet at http://pubs.acs.org.

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