# Preparation of 3,3-Diaryloxindoles by Superacid-Induced Condensations of Isatins and Aromatics with a Combinatorial Approach

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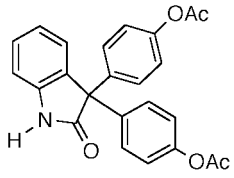
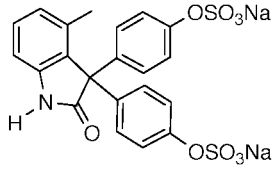
3,3-Diaryloxidoles are prepared in high yields (62−99%) by reaction of isatin or substituted isatins with aromatics in triflic acid. The reaction shows a significant dependence on acid strength which suggests the formation of diprotonated, superelectrophilic intermediates. Reaction of isatin, benzene, and acid systems of varying strength (composed of $CF_3SO_3H$ and $CF_3CO_2H$) showed that the acid strength must be more acidic than $H_o = -11.5$ for complete conversion to product. Reaction of isatins with mixtures of aromatics allows for the preparation of libraries of 3,3-diaryloxindole products by combinatorial synthesis.

## Introduction

In continuation of our studies on superelectrophilic activation in superacids, we wish report a general synthetic route to 3,3-diaryloxindoles from isatins in superacidic triflic acid ($CF_3SO_3H$, TfOH), the synthesis of modest sized libraries of 3,3-diaryloxindoles from isatins, and studies of the electrophilic chemistry of substituted isatins. Oxindole derivatives are known to possess a variety of biological activity.[1] The 3,3-diaryloxindoles have been shown to possess antibacterial, antiprotozoal, and antiinflammatory activities.[2] Compounds **1** and **2** have been used as laxatives.[3]




**1**




**2**

Oxindole derivatives have been prepared by the reaction of *N*-aryl-α-chloro-α,α-diarylacetimidoyl chlorides in sulfuric acid,[4] by the reaction of isatins with Grignard reagents,[5] by the reaction of isatin with diphenylurea and $AlCl_3$,[6] and other routes.[7] Compounds **1** and **2** are prepared by reacting isatin and 4-methylisatin, respectively, with phenol in sulfuric acid.[3] These conversions are presumably the result of protolytic activation of the isatin carbonyl group and electrophilic attack on the phenol ring.

Recently Shudo and co-workers described the conversion of some 1,2-diketones to the α,α-diphenyl ketones from benzene and TfOH.[8] For example, 2,3-butanedione was converted in 95% yield to 3,3-diphenyl-2-butanone from TfOH and benzene. It was suggested that the protonation of the 1,2-diketone generated highly reactive, or superelectrophilic, dicationic intermediates. The dicationic intermediates were sufficiently electrophilic to condense with aromatics of moderate reactivity such as benzene. Apart from these results of Shudo et al., our own studies have indicated that protosolvation of carbonyl groups in superacids may generate superelectrophilic intermediates.[9] Because the superelectrophiles often show enhanced and unique reactivity, superelectrophilic activation is a useful strategy toward the synthesis of condensed aromatic products.[10] It seemed plausible that the 1,2-dicarbonyl group of isatin might form a superelectrophilic species in superacidic solution, leading to condensation products.

## Results and Discussion

We studied the reaction of isatin in TfOH with benzene and found that 3,3-diphenyloxindole (**3a**) was produced

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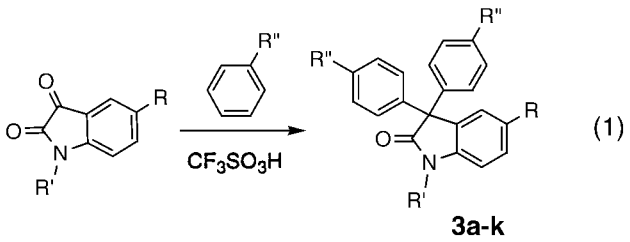
**Table 1. Reaction of Isatins with Aromatics in Triflic Acid, $CF_3SO_3H$**




(1)

| R | R′ | R″ | product | % yield[a] |
|---|---|---|---|---|
| H | H | H | **3a** | 99 |
| H | H | $CH_3$ | **3b** | 95 |
| H | H | $(CH_2)_8CH_3$ | **3c** | 62 |
| H | H | Cl | **3d** | 99 |
| H | H | F | **3e** | 71 |
| H | H | propyl-3-(4-pyridyl) | **3f** | 70 |
| F | H | H | **3g** | 87 |
| Cl | H | H | **3h** | 82 |
| $NO_2$ | H | H | **3i** | 99 |
| H | $CH_3$ | H | **3j** | 84 |
| H | Ph | H | **3k** | 95 |

[a] Isolated yields.

quantitatively (eq 1, R = R′ = R" = H). The condensation of isatin with substituted aromatics such as alkylbenzenes or chlorobenzene also proceeds in high yield (Table 1), but strongly deactivated aromatics such as nitrobenzene did not react with isatin in TfOH. *N*-Substituted and aromatic ring-substituted isatins give high yields of the 3,3-diaryloxindoles (**3g**−**k**) from TfOH. In all of the conversions, electrophilic activation occurred only at the carbonyl at the 3-position. The carbonyl at the 2-position is unreactive and this may be due to stabilization by the indole nitrogen. For alkyl- and halogen-substituted aromatics, the electrophilic attack is highly regioselective. $^1$H NMR analysis of the crude products shows only products resulting from para attack.

When isatins are reacted with mixtures of aromatic substrates, it is also possible to prepare libraries of 3,3-diaryloxindoles from combinatorial synthesis.[11] From TfOH, *N*-methylisatin, and the aromatics toluene, ethylbenzene, 1-phenylpropane, and 1-phenyl-butane, a mixture is formed containing 10 products by GC−MS analysis. The 10 peaks correspond to the 10 possible 3,3-diaryloxindoles from electrophilic attack on two aromatics, giving the products. GC−MS analysis shows ten GC peaks having respective molecular ion signals of $m/z$ = 327, 341, 355, 355, 369, 369, 383, 383, 397, 411. If one also considers the possiblity of enantiomeric products which have two different aryl groups in the 3-position of oxindole, then a total of 15 products may be produced from the combinatorial synthesis of *N*-methylisatin with TfOH and the four aromatic substrates. When the same four alkylaromatics are reacted with both *N*-methylisatin and *N*-phenylisatin in TfOH (eq 2), the library of 3,3-diaryloxindoles increases to 20 products (not including enantiomeric stereoisomers).[12] The product mixture from

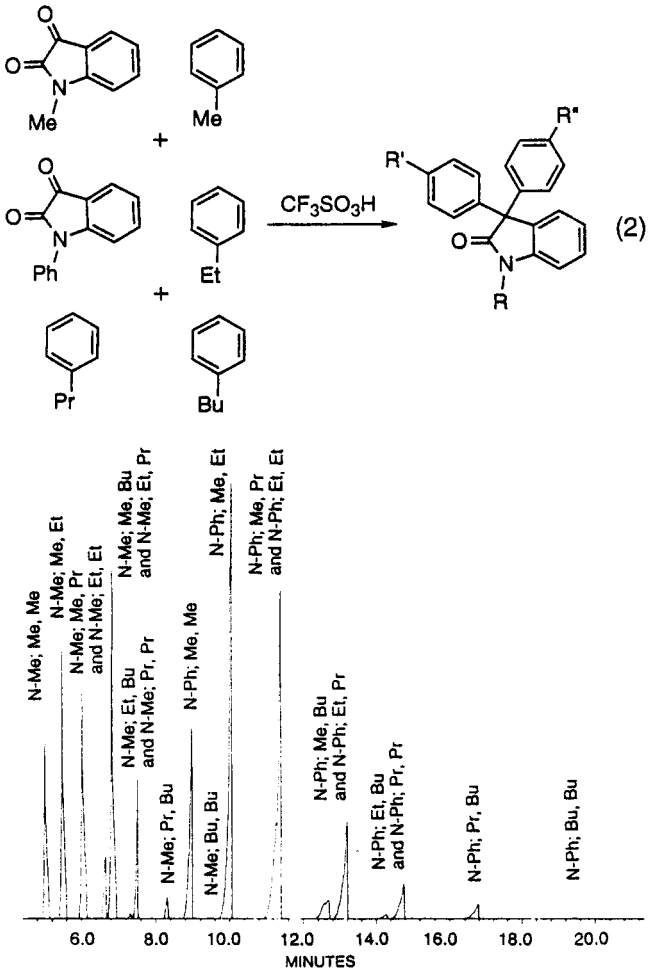



**Figure 1.** Product mixture from the reaction of *N*-methylisatin, *N*-phenylisatin, toluene, ethylbenzene, 1-phenylpropane, and 1-phenylbutane, in triflic acid (peaks are labeled as N-R, R′, and R″, from eq 2).[12]

this reaction is shown in Figure 1. All of the expected products are found in the GC−MS analysis, although the two GC peaks are unresolved which correspond to *N*-methylisatin + ethylbenzene, ethylbenzene ($m/z$ = 355) and *N*-methylisatin + toluene, 1-phenylpropane ($m/z$ = 355). GC−MS analysis shows roughly two groupings of peaks, those arising from the *N*-methylisatin and those arising from the *N*-phenylisatin.

When $H_2SO_4$ is used instead of TfOH, the condensation reaction is less effective. Isatin reacts with benzene in $H_2SO_4$, but the yield of **3a** is just 25%. In the much weaker acid $CF_3CO_2H$, isatin does not react at all with benzene. To examine the effects of acid strength on the electrophilic chemistry, isatin was reacted with benzene in varying ratios of TfOH:$CF_3CO_2H$ mixtures.[9c] Complete conversion of isatin to **3a** is seen only in media more acidic than $H_o$ = −11.5 (Table 2).[13] Incomplete conversions result in only unreacted starting material and product **3a**. No monoarylated product is observed. When 5-fluoroisatin, 5-chloroisatin, and 5-nitroisatin, are individually reacted with benzene in the TfOH:$CF_3CO_2H$ mixtures, complete conversion to the 3,3-diphenyloxindoles is seen in media of acid strengths of $H_o$ = −11.5,

(11) For a collection of reviews on combinatorial synthesis, see: *Acc. Chem. Res.* **1996**, *29*, 9 (3), 114−170.

(12) Reaction conditions: 40 mg of 1-methylisatin, 70 mg of 1-phenylisatin, and 4 mL of TfOH were combined and 0. 2 mL of toluene, 0.23 mL of ethylbenzene, 0.26 mL of 1-phenylpropane, and 0.29 mL of 1-phenylbutane were simultaneously added, and the reaction was stirred for 12 h. Products were poured into water and extracted into $CHCl_3$. Analysis was done on an HP 5890 II GC−MS with a DB-5 capillary column.

(13) The $H_o$ value is that of the starting acid. During the reaction, the acidity decreases due to the protonation of starting materials and products.

**Table 2. Results from the Reaction of Isatin, $C_6H_6$, and $CF_3SO_3H$:$CF_3CO_2H$ Acid Systems**

| $H_o$ | $CF_3SO_3H$:$CF_3CO_2H$ (w/w) | isatin:**3a** |
|---|---|---|
| -14.1 | 100:0 | 0:100 |
| -12.5 | 72.8:27.2 | 0:100 |
| -11.5 | 43.5:56.5 | 0:100 |
| -10.6 | 22.1:77.9 | 10:90 |
| -9.0 | 5.0:95.0 | 80:20 |
| -2.7 | 0:100 | 100:0 |

[a] Reaction conditions: 0.05 g of isatin, 1.0 mL of acid, and 1.0 mL of $C_6H_6$ at 25 °C for 12 h.

$H_o = -10.6$, and $H_o = -9$, respectively. Thus, substitution of isatin by electron-withdrawing groups tends to enhance the electrophilic character of the protonated isatins.

These results show that strongly acidic media is required to completely convert the isatins to 3,3-diphenyloxindoles. A significant dependence on acid strength is also seen when isatin is reacted with chlorobenzene. Isatin and TfOH with chlorobenzene produces **3d** in 99% yield, while from $H_2SO_4$ and chlorobenzene, **3d** is produced in 2% yield. The requirement for superacidic media is consistent with the formation of protosolvated, dicationic intermediates. Diprotonated isatin (or substituted isatins) would only be formed in superacidic media and would exhibit superelectrophilic chemistry. The present results obtained with isatins and Shudo's earlier studies of 1,2-diketones indicate that adjacent carbonyl groups form highly reactive electrophiles in superacidic TfOH.

## Conclusion

In summary, we have found that isatins react with aromatics in TfOH to give 3,3-diaryloxindoles in high to excellent yields. The reaction is highly regioselective in para attack on alkyl- and halogen-substituted aromatics and in condensation chemistry occurring exclusively at the 3-carbonyl group of the isatins. Modestly sized libraries of 3,3-diaryloxyindoles have been prepared from mixtures of TfOH, aromatics, and isatins, using the technique of combinatorial synthesis. Isatin is completely converted to the condensation product **5a** only in media more acidic than $H_o = -11.5$. These results are consistent with the formation of dicationic intermediates leading to products.

## Experimental Section

Triflic acid was purchased from 3M Co. and distilled under an Ar atmosphere. Isatin and substituted isatins were purchased from Aldrich and used as received. $^1H$ and $^{13}C$ NMR were recorded on a 300 MHz NMR instrument and mass spectra were recorded with a mass selective detector. High-resolution mass spectra were measured by the Southern California Mass Spectrometry Facility at the University of California at Riverside. Combustion anaylsis was done by Galbraith Laboratories, Knoxville, TN.

**General Synthetic Procedure.** Isatin (0.1 g, 0.68 mmol) is combined with 2 mL of freshly distilled TfOH, and to this mixture is added 1 mL of benzene. After stirring at 25 °C for about 8 h, the product mixture is poured over severeal grams of ice and the products are then extracted into either toluene or chloroform. The organic solution is then washed with water and then brine and dried with $MgSO_4$. Concentration in vacuo provides 3,3-diphenyloxindole as the sole product which may be further purified by recrystallization from chloroform.

**3,3-Bis(4-(1-nonyl)phenyl)oxindole, 3c**. Isatin (0.2 g, 1.36 mmol) gave **3c** (0.43 g, 0.8 mmol, 62%) after column chromatograhpy (9:1, hexanes:$Et_2O$) as a waxy solid: mp 65−68 °C ($CHCl_3$); $^1H$ NMR (300 MHz, $CDCl_3$) $\delta = 0.88$ (t, $J = 6.9$ Hz, 6H), 1.26 (m, 24H), 1.58 (m, 4H), 2.56 (t, $J = 7.5$ Hz, 4H), 6.95 (d, $J = 8.1$ Hz, 1H), 7.04 (t, $J = 7.8$ Hz, 1H), 7.10 (d, $J = 8.1$ Hz, 4H), 7.19 (t, $J = 6.9$ Hz, 4H), 7.20−7.32 (m, 2H), 9.05 (s, 1H); $^{13}C$ NMR (125 MHz, $CDCl_3$) $\delta = 14.1$, 22.6, 29.3, 29.4, 29.5, 29.5, 31.3, 31.9, 35.5, 62.5, 110.3, 122.6, 126.2, 128.0, 128.2, 128.4, 134.0, 138.8, 140.2, 142.0, 180.5; HRMS $C_{38}H_{51}NO$ calcd 537.3960, found 537.3970.

**3,3-Bis(4-chlorophenyl)oxindole, 3d**. Isatin (0.1 g, 0.68 mmol) gave **3d** (0.238 g, 0.67 mmol, 99%) as a white solid: mp 180−184 °C ($CHCl_3$); $^1H$ NMR (300 MHz, $CDCl_3$) $\delta = 6.96$ (d, $J = 7.5$ Hz, 1H), 7.10 (t, $J = 7.5$ Hz, 1H), 7.18 (m, 1H), 7.23 (d, $J = 8.7$ Hz, 14H), 7.26−7.39 (m, 6H), 9.68 (s, 1H); $^{13}C$ NMR (125 MHz, $CDCl_3$) $\delta = 62.0$, 110.9, 123.1, 125.9, 128.8, 129.7, 132.7, 133.7, 139.7, 140.2, 180.0; HRMS $C_{20}H_{13}Cl_2NO$ calcd 353.0374, found 353.0371. Anal. Calcd for $C_{20}H_{13}Cl_2NO$: C, 67.81; H, 3.70; N, 3.95. Found: C, 67.61; H, 3.74; N, 3.86.

**3,3-Bis(4-fluorophenyl)oxindole, 3e**. Isatin (0.1 g, 0.68 mmol) gave **3e** (0.153 g, 0.48 mmol, 71%) as a white solid: mp 107−110 °C ($CHCl_3$); $^1H$ NMR (300 MHz, $CDCl_3$) $\delta = 7.20$−7.63 (m, 12H), 9.35 (s, 1H); $^{13}C$ NMR (125 MHz, $CDCl_3$) $\delta = 62.1$, 111.2, 115.4 (d, $J_{c-f} = 36$ Hz), 123.4, 125.8, 128.7, 129.9 (d, $J_{c-f} = 13$ Hz), 133.3, 136.8 (d, $J_{c-f} = 5$ Hz), 139.9, 162.1 (d, $J_{c-f} = 411$ Hz), 180.9; HRMS $C_{20}H_{13}F_2NO$ calcd 321.0965, found 321.0967.

**3,3-Bis[4-(3-(4-pyridyl)propyl)phenyl]oxindole, 3f**. Isatin (0.1 g, 0.68 mmol) gave **3f** (0.247 g, 0.47 mmol, 70%) after column chromatography (9:1, hexanes:$Et_2O$) as a white solid: mp 151−156 °C ($CHCl_3$); $^1H$ NMR (300 MHz, $CDCl_3$) $\delta = 1.90$ (m, 4H), 2.60 (m, 8H), 6.92 (d, $J = 8.1$ Hz, 1H), 7.00−7.32 (m, 15H), 8.47 (M 4H), 9.98 (s, 1H); $^{13}C$ NMR (125 MHz, $CDCl_3$) $\delta = 31.3$, 34.4, 34.6, 62.3, 110.0, 122.3, 123.7, 126.0, 127.9, 128.3, 133.7, 139.4, 140.7, 141.4, 149.4, 151.0, 179.9; HRMS-CIMS $C_{36}H_{34}N_3O$ ($M + H^+$) calcd 524.2702, found 524.2695. Anal. Calcd for $C_{36}H_{33}N_3O$: C, 82.57; H, 6.35; N, 8.02. Found: C, 82.69; H, 6.34; N, 8.76.

**3,3-Diphenyl-5-fluorooxindole, 3g**. 5-Fluoroisatin (0.1 g, 0.61 mmol) gave **3g** (0.160 g, 0.53 mmol, 78%) as a white solid: mp 219−223 °C ($CHCl_3$); $^1H$ NMR (300 MHz, $CDCl_3$) $\delta = 6.84$−7.08 (m, 3H), 7.20−7.36 (m, 10H), 8.44 (s, 1H); $^{13}C$ NMR (125 MHz, $CDCl_3$) $\delta = 63.6$, 111.1 (d, $J_{c-f} = 13$ Hz), 113.9 (d, $J_{c-f} = 42$ Hz), 114.8 (d, $J_{c-f} = 39$ Hz), 127.6, 128.3, 128.6, 135.5 (d, $J_{c-f} = 125$ Hz), 141.0 (d, $J_{c-f} = 400$ Hz), 180.2; HRMS $C_{20}H_{14}FNO$ calcd 303.1059, found 303.1052.

**3,3-Diphenyl-5-chlorooxindole, 3h**. 5-Chloroisatin (0.1 g, 0.55 mmol) gave **3h** (0.145 g, 0.46 mmol, 82%) as a white solid: mp 207−210 °C ($CHCl_3$); $^1H$ NMR (300 MHz, $CDCl_3$) $\delta = 6.85$ (d, $J = 8.1$ Hz, 1H), 7.13−7.33 (m, 12H), 8.81 (s, 1H); $^{13}C$ NMR (125 MHz, $CDCl_3$) $\delta = 62.5$, 111.1, 126.7, 127.7, 128.3, 128.4, 128.7, 135.2, 136.5, 140.9, 149.0, 167.5; HRMS-CIMS $C_{20}H_{18}ClN_2O$ ($M + NH_4^+$) calcd 337.1108, found 337.1109.

**3,3-Diphenyl-5-nitrooxindole, 3i**. 5-Nitroisatin (0.1 g, 0.52 mmol) gave **3i** (0.17 g, 0.51 mmol, 99%) as a white solid: mp 203−209 °C ($CHCl_3$); $^1H$ NMR (300 MHz, $CDCl_3$) $\delta = 7.02$ (d, $J = 8.4$ Hz, 1H), 7.15−7.40 (m, 11H), 8.13 (s, 1H), 8.19 (d, $J = 8.4$ Hz, 1H), 10.18 (s, 1H); $^{13}C$ NMR (125 MHz, $CDCl_3$) $\delta = 63.1$, 110.5, 122.0, 125.4, 128.1, 128.2, 128.8, 134.4, 140.0, 143.7, 146.1, 180.8; HRMS $C_{20}H_{14}N_2O_3$ calcd 330.1004, found 330.1003.

**3,3-Diphenyl-1-methyloxindole, 3j**. 1-Methylisatin (0.1 g, 0.62 mmol) gave **3j** (0.155 g, 0.52 mmol, 84%) as a white solid: mp 169−173 °C ($CHCl_3$); $^1H$ NMR (300 MHz, $CDCl_3$) $\delta = 3.31$ (s, 3H), 6.95 (d, $J = 7.8$ Hz, 1H), 7.13 (d, $J = 7.8$ Hz, 1H), 7.20−7.35 (m, 12H); $^{13}C$ NMR (125 MHz, $CDCl_3$) $\delta = 26.5$, 62.5, 108.5, 122.8, 126.0, 127.2, 128.2, 128.3, 128.4, 132.8, 141.8, 143.0, 177.5; HRMS $C_{21}H_{17}NO$ calcd 299.1310, found 299.1315. Anal. Calcd for $C_{21}H_{17}NO$: C, 84.25; H, 5.72; N, 4.68. Found: C, 84.47; H, 5.80; N, 4.57.

**3,3-Diphenyl-1-phenyloxindole, 3k**. 1-Phenylisatin (0.1 g, 0.45 mmol) gave **3j** (0.154 g, 0.43 mmol, 95%) as a white solid: mp 161−163 °C ($CHCl_3$); $^1H$ NMR (300 MHz, $CDCl_3$) $\delta = 6.95$ (d, $J = 8.1$ Hz, 1H), 7.16−7.60 (m, 18H); $^{13}C$ NMR (125

MHz, $CDCl_3$) $\delta$ = 62.6, 109.9, 123.3, 126.4, 126.8, 127.4, 128.1, 128.2, 128.5, 129.6, 132.7, 134.5, 142.0, 177.5; HRMS $C_{26}H_{19}NO$ calcd 361.1467, found 361.1456. Anal. Calcd for $C_{26}H_{19}NO$: C, 86.40; H, 5.30; N, 3.88. Found: C, 86.27; H, 5.24; N, 3.86.

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**Supporting Information Available:** $^1H$ and $^{13}C$ NMR for products **3c**−**k** (18 pages). This material is contained in libraries on microfiche, immediately follows this article in the microfilm version of the journal, and can be ordered from the ACS; see any current masthead page for ordering information.

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