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Gold(III)-Catalyzed Tandem Reaction of Ketones with Phenols: Efficient and Highly Selective Synthesis of Functionalized 4*H*-Chromenes

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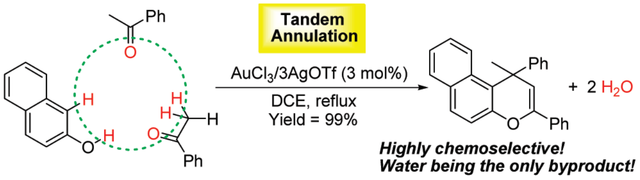

Highly chemoselective!
Water being the only byproduct!

An efficient and highly selective approach for the synthesis of functionalized 4*H*-chromenes has been developed via gold(III)-catalyzed condensation/annulation tandem reaction of ketones with phenols.

Chromenes (2*H*-chromenes and 4*H*-chromenes) constitute an important class of scaffolds found in many naturally occurring and synthetic molecules exhibiting unique biological and pharmacological activities.[1] Among many approaches for the construction of chromene structures,[2] those involving 2*H*-chromenes[3] have been well established while those for their counterpart, 4*H*-chromenes,[4] have been less developed. As catalytic processes for the synthesis of functionalized 4*H*-chromenes are concerned, they can be generally divided into two classes: (1) transition metal−catalyst involved reactions, e.g., gold(I)-catalyzed carboalkoxylation reaction of propargyl esters,[4a] $FeCl_3$-catalyzed reaction of substituted 2-(hydroxymethyl) phenols with β-ketoesters or β-diketones,[4b] ruthenium-mediated cycloaddition of propargylic alcohols with phenols,[4c] copper-catalyzed intramolecular *O*-arylation of β-ketoesters,[4d] and *O*-vinylation reaction of phenols with $Sn(vinyl)_4$ followed by a ruthenium-mediated RCM reaction,[4e] and palladium-catalyzed conjugate addition of (2-hydroxyaryl)mercury chlorides with α,β-unsaturated compounds followed by cyclization and elimination of water;[5] (2) organocatalyst involved reactions, e.g., DABCO or phosphine-catalyzed reactions of salicyclic imines with α,β-unsaturated compounds.[4f−h] Unfortunately, most of these procedures suffer from using commercially unavailable substrates as starting materials. Therefore, developing efficient and conventional catalytic processes to 4*H*-chromenes from simple and readily available substrates remains a challenging task.

On the other hand, tandem reactions,[6] compared with stepwise reactions, usually provide more efficient and environmentally benign processes to construct molecular diversity and structural complexity from readily available substrates in a single step without the separation and purification of the intermediates. Recently, gold-catalyzed tandem reactions[7] have received special attention as gold catalysts[8] generally exhibit extraordinary reactivity and show high selectivity in

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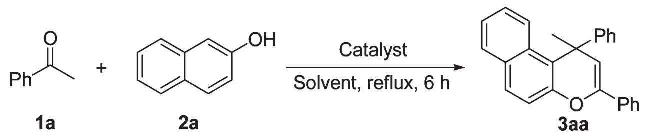
TABLE 1. **Optimization of Reaction Conditions**[a]



entry	catalyst (0.03 mmol)	solvent	yield of **3aa** (%)
1	AuCl/AgOTf	DCE[b]	75
2	PPh_3AuCl/AgOTf	DCE	93
3	$AuCl_3$/3AgOTf	DCE	99
		dioxane[c]	15
		THF	50
		toluene[c]	95
		chlorobenzene[c]	96
		MeCN	> 5
4	$AuCl_3$	DCE	0
5	AuCl	DCE	0
6	PPh_3AuCl	DCE	0
7	AgOTf[d]	DCE	0
8	$AlCl_3$[e]	DCE	0
9	$ZnCl_2$[e]	DCE	0
10	$BiCl_3$[e]	DCE	0
11	$SnCl_4$[e]	DCE	0
12	$FeCl_3$[e]	DCE	26
13	$Cu(OTf)_2$[e]	DCE	7
14	HCl[e,f]	DCE	0
15	HOTf[e]	DCE	12
16	none	DCE	0[g]

[a]Carried out on 2 mmol of **1a** and 1 mmol of **2a** in the presence of catalyst in solvent (3 mL) at reflux for 6 h. [b]DCE = 1,2-dichloroethane. [c]The reaction temperature at 90 °C. [d]The amount of catalyst is 0.09 mmol. [e]The amount of catalyst is 0.3 mmol. [f]Aqueous HCl (37 wt %) was used. [g]**1a** and **2a** were recovered quantitatively.

the reactions. In 1986, Kozlikovskii[9] reported a tandem reaction of acetophenone with phenol catalyzed by aluminum phenolate in which functionalized 4*H*-chromene could be produced, but only one example was investigated and the reaction gave very low selectivity as in this case a messy mixture of six products were formed. As part of our ongoing interest in the construction of chromene structures[10] and gold-catalyzed highly selective reactions,[11] we wish to present herein a unique gold(III)-catalyzed efficient and highly selective approach for the synthesis of functionalized 4*H*-chromenes from commercially available ketones and phenols via a one-pot tandem reaction manner.

Initially, the reaction of acetophenone **1a** and 2-naphthol **2a** was investigated to optimize the reaction conditions (Table 1). When 2 equiv of **1a** and 1 equiv of **2a** were treated with AuCl/AgOTf (3 mol % based on **2a**) in DCE at reflux for 6 h, the desired 4*H*-chromene **3aa** was isolated in 75% yield (entry 1). Under the catalysis of a PPh_3AuCl/AgOTf system, the yield of **3aa** was increased to 93% (entry 2). Gratifyingly, almost quantitative yield of **3aa** was obtained by using $AuCl_3$/3AgOTf as a promotor (entry 3). Solvent screening showed that toluene and chlorobenzene were suitable media as well while THF, dioxane, and MeCN were less effective (entry 3). The reaction could hardly take place without a catalyst (entry 16), or in the presence of AuCl, Ph_3PAuCl, $AuCl_3$, or AgOTf alone (entries 4−7), indicating that cationic gold(III) or gold(I) species were essential for the reaction due to their more electrophilic properties.[12] Catalyst screening experiments revealed that the tandem reaction of **1a** with **2a** in the presence of other conventional Lewis or Brønsted acids as catalyst was far less effective even at a catalyst loading of 30 mol % (entries 8−15), indicating gold(III) catalyst played a unique role in achieving high reactivity and selectivity for the present reaction.

Subsequently, both ketones and phenols were surveyed to investigate the scope and generality of the reaction (Table 2). First, a variety of ketones **1** were subjected to react with 2-naphthol **2a** under the optimized reaction conditions. It was found that aryl methyl ketones reacted with **2a** smoothly to give the corresponding products **3** in moderate to excellent yields (67−99%, entries 1−9). The reactions between electron-deficient aryl methyl ketones (except **1g**) and **2a** proceeded faster and gave higher yields of target products (entries 6, 8, and 9) than those reactions concerning electron-rich aryl methyl ketones with **2a** (entries 2−4). The reaction of a thiophene-tethered methyl ketone **1j** with **2a** under standard conditions furnished **3ja** in low yield (35%, entry 10); however, an enhancement of catalyst loading to 25 mol % resulted in the improvement of the yield of **3ja** to 60% (entry 10). When a more steric hindrance ketone (compared with methyl ketones), for example, **1k**, was used as a substrate, the reaction gave none of the desired product (entry 11). We assume that the initial self-condensation of ketones to chalcones might be the key step for the reaction (Scheme 2); the failed reaction of **1k** with **2a** may be ascribed to the difficult self-condensation of **1k** to chalcone **7**.[13] In fact, when preparative chalcone **7**, instead of **1k**, was subjected to the reaction conditions, a decomposition of **7** back to **1k** occurred and only a trace amount of **3ka** was detected (entry 11). To our delight, when less steric hindrance α,*β*-unsaturated ketones were used as starting materials to react with **2a**, the desired 4*H*-chromene products were obtained in moderate to excellent yields under similar reaction conditions (60−95%, Scheme 1). The reaction of cylcohexanone **1m** with **2a** resulted in the formation of a spiro 4*H*-chromene structure in moderate yield (entry 12). It is interesting that treatment of ketone **1m** with **2a** gave none of the expected product **3ma**, whereas a 2*H*-chromene isomer **3′ma**[14] was isolated in 45% yield (entry 13). An unsymmetrical aliphatic ketone **1n** was also investigated for the reaction; unfortunately, a complex mixture of products was obtained (entry 14). Next, a series of other phenols were tested for the reaction, and the results indicated that electron-rich phenols were suitable substrates for the reaction (entries 15−22). For instance, ketone **1a** or **1f** underwent the tandem reaction with electron-rich phenols **2b**, **2c**, **2d**, or **2e** smoothly to afford respective product in good to excellent yield (entries 15, 17, and 18−22). Unfortunately, the reaction

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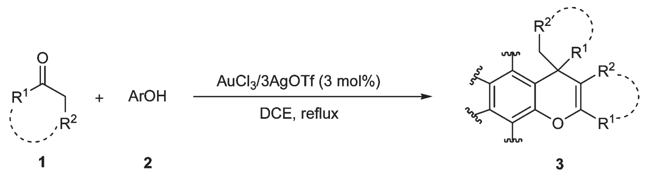
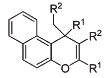
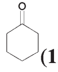
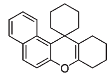
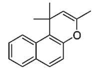
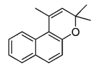
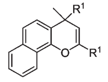
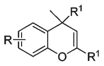
TABLE 2. Au(III)-Catalyzed Tandem Reaction of Ketones 1 with Phenols 2[a]

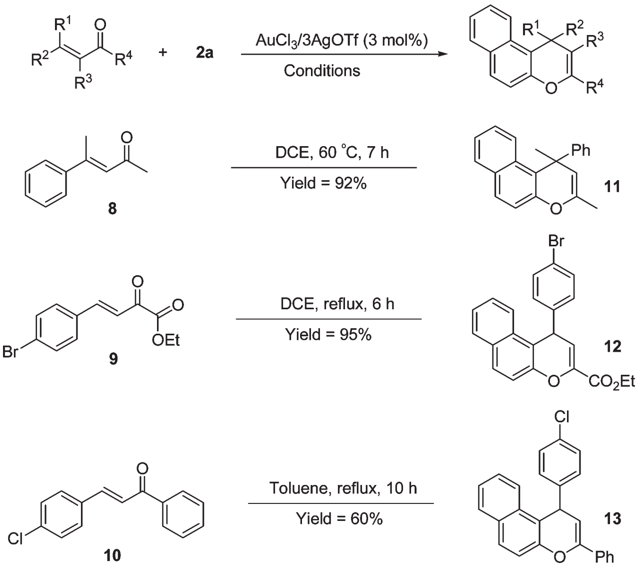


entry	R^1, R^2 (1)	ArOH (2)	time (h)	product: yield (%)
				
1	Ph, H (**1a**)	2-naphthyl (**2a**)	6	**3aa**: 99
2	4-MeC$_6$H$_4$, H (**1b**)	**2a**	8	**3ba**: 89
3	4-MeOC$_6$H$_4$, H (**1c**)	**2a**	8	**3ca**: 72
4	2-MeOC$_6$H$_4$, H (**1d**)	**2a**	8	**3da**: 67
5	4-PhC$_6$H$_4$, H (**1e**)	**2a**	10	**3ea**: 69
6	4-ClC$_6$H$_4$, H (**1f**)	**2a**	6	**3fa**: 98
7	2-ClC$_6$H$_4$, H (**1g**)	**2a**	7	**3ga**: 72
8	4-BrC$_6$H$_4$, H (**1h**)	**2a**	6	**3ha**: 98
9	4-FC$_6$H$_4$, H (**1i**)	**2a**	6	**3ia**: 92
10	2-thiophenyl, H (**1j**)	**2a**	6	**3ja**: 35; 60[b]
11	Ph, Me (**1k**)	**2a**	8	**3ka**: 0; trace[c,d]
12	 (**1l**)	**2a**	6	 **3la**: 32; 62[e]
13	Me, H (**1m**)	**2a**	8	 **3ma**: 0  **3'ma**: 45[e]
14	Et, H (**1n**)	**2a**	6	**3na**: messy
				
15	**1a**	1-naphthyl (**2b**)	6	**3ab**: 98
16	**1d**	**2b**	8	**3db**: 62
17	**1f**	**2b**	6	**3fb**: 93
				
18	**1a**	Ph (**2c**)	6	**3ac**: 86
19	**1a**	4-MeC$_6$H$_4$ (**2d**)	6	**3ad**: 91
20	**1f**	**2d**	6	**3fd**: 90
21	**1a**	4-^{t}BuC$_6$H$_4$ (**2e**)	6	**3ae**: 93
22	**1f**	**2e**	6	**3fe**: 92
23	**1a**	4-ClC$_6$H$_4$ (**2f**)	8	**3af**: trace

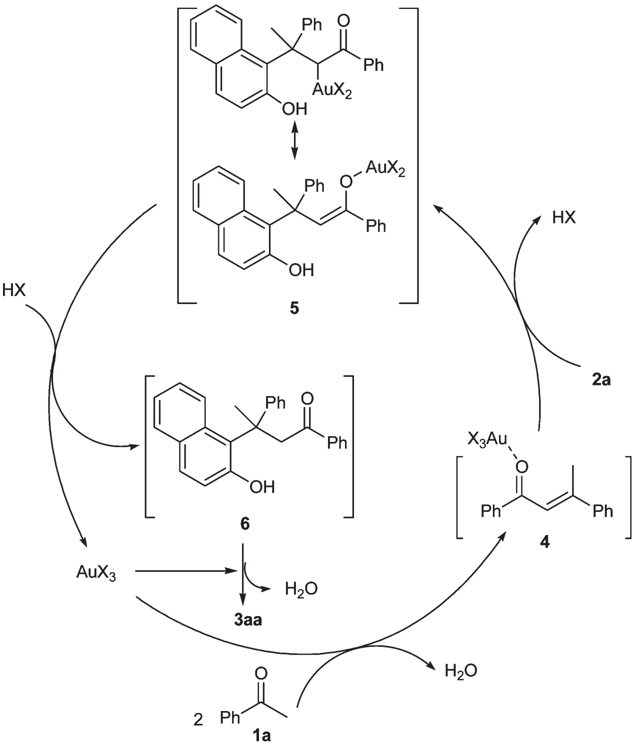
[a] Reaction conditions: **1** (2 mmol), **2** (1 mmol), AuCl$_3$/3AgOTf (0.03 mmol), DCE (3 mL), reflux. [b] The amount of catalyst is 0.25 mmol. [c] Preparative chalcone **7** was used instead of **1k** as the substrate. [d] **1k** was isolated in 96% yield. [e] The molar ratio of ketone to phenol is 4:1.

between **1a** and electron-deficient phenol **2f** was not successful (entry 20). The structure of **3** was established on the basis of the spectral analysis, which was further confirmed by X-ray diffraction analysis of a related derivative **3da** (Figure S1, Supporting Information).

SCHEME 1. Au(III)-Catalyzed Tandem Reaction of α,β-Unsaturated Ketones with 2-Naphthol 2a



SCHEME 2. Proposed Mechanism



At present stage, no clear mechanism has been elucidated. It was found that the condensation product **4** (Scheme 2) from two molecules of acetophenone **1a** could be detected in the reaction of **1a** with **2a**. When the preparative β-methyl chalcone **4**,[15] instead of acetophenone, was employed as the starting material to react with 2-naphthol, the desired product **3aa** was also obtained in quantitative yield,[16] therefore, we thought **4** might be the key intermediate for the reaction.

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[15] Muzart, J. *Synth. Commun.* **1985**, *15*, 285.

[16] The reaction between phenol and mesityl oxide in the presence of HCl has been studied, see: Dianin, A. P. *Zh. Russ. Fiz.-Khim. Ova.* **1914**, *46*, 1310.

On the basis of these facts and previous reports,[17] a possible mechanism concerning the gold(III)-catalyzed tandem reaction of **1a** with **2a** is proposed (Scheme 2). First, gold(III)-catalyzed self-condensation of **1a** formed intermediate **4**. Then the in situ generated **4** reacted with **2a** to give intermediate **5** via gold-activating the enone **4** followed by an electrophilic aromatic substitution at the 1-position of 2-naphthol and a deprotonation process.[17] Protodemetalation of **5** liberated the intermediate **6** and regenerated the gold catalyst. Intramolecular annulation of **6** with the aid of the gold catalyst eventually furnished **3aa**.

In summary, for the first time we have developed an efficient and highly selective tandem reaction of ketones with phenols catalyzed by $AuCl_3$/3AgOTf, which furnished various functionalized 4*H*-chromenes in moderate to excellent yields with water as the sole byproduct.

Experimental Section

$AuCl_3$ (9.1 mg, 0.03 mmol), AgOTf (23.1 mg, 0.09 mmol), and DCE (2 mL) were added to a 10-mL flask. The mixture was stirred at rt for 5 min before a DCE solution (1 mL) of **1a** (2.0 mmol, 0.24 g) and **2a** (1.0 mmol, 0.14 g) was added. Then the reaction mixture was stirred at reflux. Upon completion, the resulting mixture was diluted with CH_2Cl_2 (10 mL) and filtered through Celite. After evaporation of the solvent under vacuum, the residue was purified by column chromatography on silica gel (200−300 mesh) with petroleum ether−EtOAc (20:1) as eluent to give pure **3aa** (0.34 g, 99%). Pale yellow oil; R_f 0.62 (cyclohexane−EtOAc, 20:1); IR (neat) ν 1678 (C=C) cm^{-1}; ^{1}H NMR ($CDCl_3$/TMS, 500 MHz) δ 7.73−7.68 (m, 4H), 7.54 (d, J = 8.5 Hz, 1H), 7.47−7.09 (m, 11H), 5.29 (s, 1H), 2.17 (s, 3H); ^{13}C NMR ($CDCl_3$/TMS, 125 MHz) δ 150.2, 148.9, 143.0, 133.8, 131.9, 131.5, 129.3, 128.8, 128.6, 128.4, 128.3, 127.1, 126.3, 125.9, 125.6, 124.6, 123.5, 118.9, 118.3, 109.1, 40.8, 29.1; MS (EI, 70 eV) m/z (%) 348(15) [M$^+$], 333(100); HRMS (EI) calcd for $C_{26}H_{20}O$ 348.1514, found 348.1514. For more details, see the Supporting Information.

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Supporting Information Available: Experimental procedures and characterization data of all new compounds as well as X-ray structural data (CIF). This material is available free of charge via the Internet at http://pubs.acs.org.

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