Electrooxidative Metal-Free Cyclization of 4-Arylaminocoumarins with DMF as C1-Source

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Abstract: An environmentally-benign electrochemical approach for the construction of quinoline derivatives employing *N*,*N*-dimethylformamide (DMF) as the methine source has been devised by cyclization of 4-(phenylamino)-*2H*-chromen-2-ones. In a user-friendly undivided cell, *6H*-chromeno[4,3-*b*]quinolin-6-ones were obtained under chemical oxidant-free and transition-metal-free conditions in 43–92% yields with high functional tolerance.

Keywords: Electrochemistry; Metal-free catalytic strategy; C–C bond formation; *N*,*N*-dimethylformamide; Chromene-fused quinoline

Chromene-fused quinoline derivatives are wildly distributed in natural products,[1] and display excellent performance towards pharmacological and biological activities, such as anti-cancer activity,[2] selective non-steroidal progesterone receptor modulators,[3] glucocorticoid modulators,[4] and estrogen receptor *β*-selective ligands.[5] Additionally, there are also important applications in biotechnology, such as dyes for mitochondrial imaging[6] and fluorescent pH sensor (Figure 1).[7]

In recent years, electrocatalysis has been recognized as a versatile and environmentally-friendly tool for molecular synthesis due to its effective nature, atom-efficiency, and environmentally-friendly merits.[8] Thus, electrochemistry has enabled significant developments of C–H functionalization for the formation of C–C and C–Het bonds.[9] Among them, the selective C–C formation is a crucial challenge for the modification of natural products, pharmaceutically active compounds, and ligands for catalytic transformations.

At present, the C–C bond formations under transition metal-catalyzed[10] and metal-free electrochemical conditions have been disclosed.[11–14] Especially, metal-free catalyzed electrochemistry enabled wide applications for green protocols of alkylations,[12] arylations,[13] and dehydrogenative couplings[14] (Scheme 1a). New methods for C–C bond formations by electrochemistry remain underdeveloped for an efficient access to valuable organic structural motifs.

Almost all C–C bond coupling reactions using DMF as a carbon source have the disadvantages of high temperature and harsh conditions.[15] Therefore, there is a strong demand for methods that use DMF as a carbon source to construct C–C bond under mild conditions.

We have previously reported a route using copper as catalyst and inexpensive industrial raw material DMF as the carbon source in the presence of *tert*-butyl

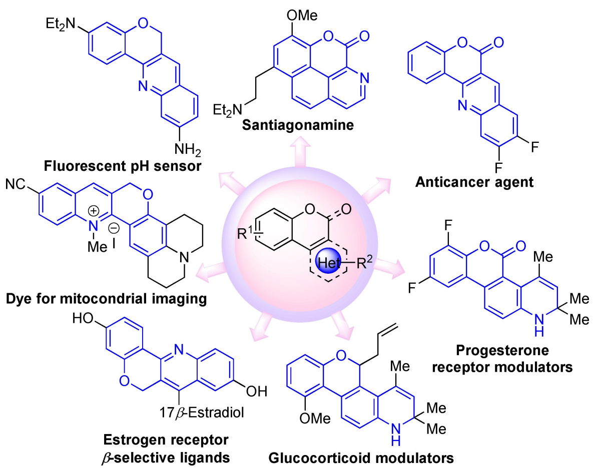
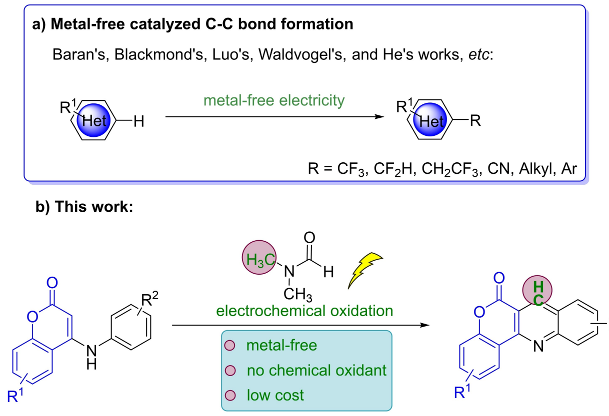


Figure 1. Some examples of chromene-fused quinolines.



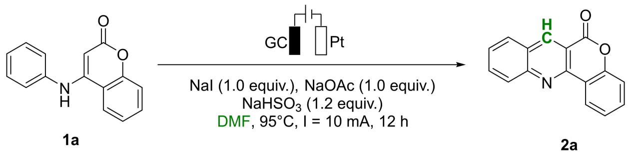
Scheme 1. The development of C–C bond formations.

perbenzoate (TBPB) as the oxidant to construct *6H*-chromeno [4,3-*b*] quinoline-6-ones.[16] Thus, we have proposed a strategy for metal-free electrocatalytic cyclizations of 4-arylaminocoumarins using DMF as the carbon source in the absence of toxic and expensive chemical oxidant in a mild, efficient, and environmentally-friendly manner (Scheme 1b).

We commenced our studies by using 4-(phenylamino)-2*H*-chromen-2-one (**1 a**) as the model substrate to identify the optimal reaction conditions (Table 1). The desired 6*H*-chromeno[4,3-*b*]quinolin-6-one **2 a** was isolated in 92% yield when substrate **1 a** was directly electrolyzed at a 10 mA constant current in a mixed solution of NaI, NaOAc, and $NaHSO_3$ in anhydrous DMF at 95 °C without additional metals (entry 1). Compared with NaI as the iodide salt, the yield dropped slightly to 89% when employing KI as the iodide additive (entry 2). However, only 45% yield of product **2 a** was obtained using $n\mathrm{Bu_4NI}$ instead of NaI (entry 3). As was expected, when switching iodide salts to other additives, such as NaBr, $n\mathrm{Bu_4NBF_4}$, and $LiClO_4$, no desired product was detected (entries 4–6). In addition, the participation of water reduced the yield, and this proved that anhydrous conditions were particularly important for this reaction (entries 7–8). Further control experiments confirmed the essential role of electricity, as this resulted in lower yields or even no desired product (entries 9–11). Interestingly, it was found that carbon rods or platinum plates were not advantageous as anodes than glassy carbon (GC) anodes (entries 12–13). The yield was decreased slightly when the reaction was performed at 100 °C (entry 14).

With the optimized conditions in hand, the substrate scope was next investigated to illustrate the robustness of the electrooxidative catalytic strategy (Scheme 2).

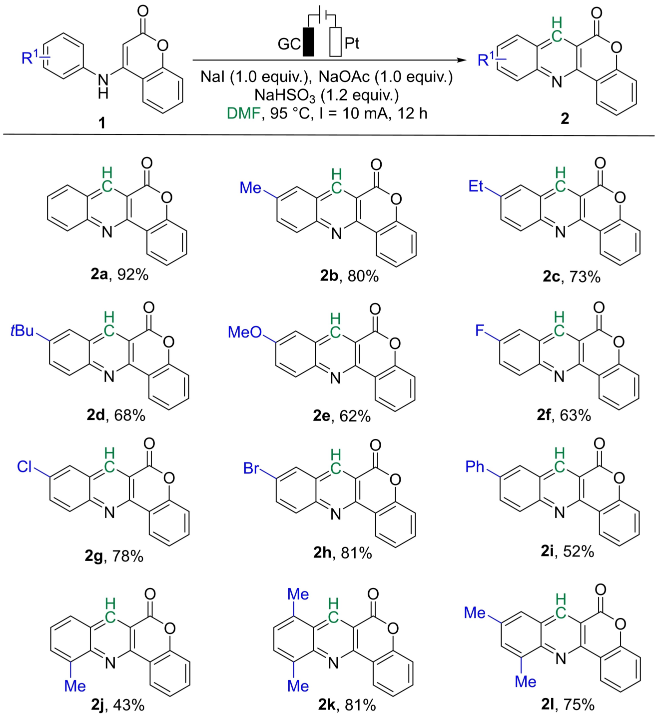
Table 1. Optimization of the reaction conditions.[a]



Entry	Deviation from standard conditions	Yield/%[b]
1	None	92
2	KI instead of NaI	89
3	$n\mathrm{Bu_4NI}$ instead of NaI	45
4	NaBr instead of NaI	0
5	$n\mathrm{Bu_4NBF_4}$ instead of NaI	0
6	$LiClO_4$ instead of NaI	0
7	DMF/H_2O (14:1) instead of DMF	77
8	DMF/H_2O (5:1) instead of DMF	55
9	I = 8 mA	78
10	I = 15 mA	82
11	No current	0
12	C as anode	0
13	Pt as anode	52
14	100 °C	80

[a] Reaction conditions: Undivided cell, Glassy carbon anode, Pt plate cathode, **1 a** (0.20 mmol), NaI (0.20 mmol), NaOAc (0.20 mmol), $NaHSO_3$ (0.24 mmol), anhydrous DMF (3.0 mL), constant current = 10 mA, 95 °C, 12 h.

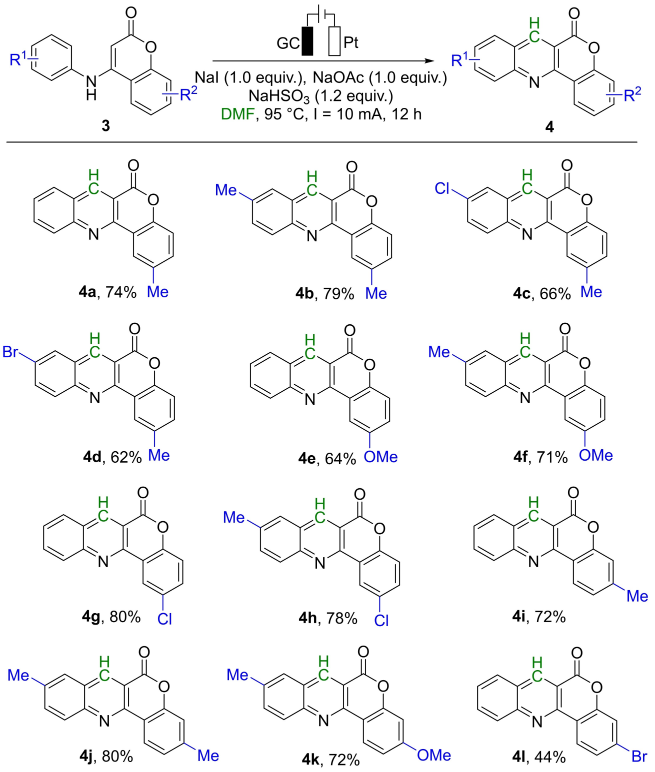
[b] Yields of isolated product.



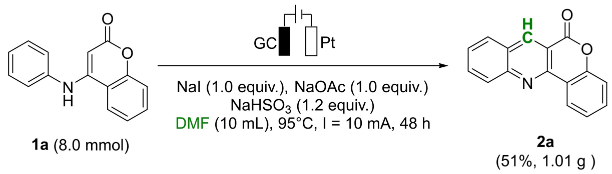
Scheme 2. Synthesis of *6H*-chromeno [4,3-*b*] quinoline-6-ones (**2 a**–**2 l**) ^{a,b}. ^{a}Reaction conditions: Undivided cell, Glassy carbon anode, Pt plate cathode, **1** (0.20 mmol), NaI (0.20 mmol), NaOAc (0.20 mmol), $NaHSO_3$ (0.24 mmol), anhydrous DMF (3.0 mL), constant current = 10 mA, 95 °C, 12 h. ^{b}Isolated yield of **2**.

As shown, electro-donating groups on the aniline ring showed moderate to excellent reactivity in the synthesis of *6H*-chromeno[4,3-*b*]quinolin-6-ones **2 a**–**2 e**, **2 j**–**2 l** (62–92%). Furthermore, halide substituents (F, Cl, Br) were well-tolerated, providing the corresponding products (**2 f**–**2 h**) in 63–81% yields. However, compared with common electron-donating groups, phenyl-substituted substrate (**2 i**, 52%) obtained lower yield, which may be due to steric hindrance.

Finally, the substitution effect on both the coumarin and the benzene ring was taken into consideration (Scheme 3). The reaction performed well with either the coumarin moiety or the aromatic ring containing electron-donating or electron-withdrawing substituents (**4 a**–**4 l**, 44–80%).



Scheme 3. Synthesis of *6H*-chromeno [4,3-*b*] quinoline-6-ones (**4 a**–**4 l**) [a,b]. [a]Reaction conditions: Undivided cell, Glassy carbon anode, Pt plate cathode, **3** (0.20 mmol), NaI (0.20 mmol), NaOAc (0.20 mmol), $NaHSO_3$ (0.24 mmol), anhydrous DMF (3.0 mL), constant current = 10 mA, 95 °C, 12 h. [b]Isolated yield of **4**.



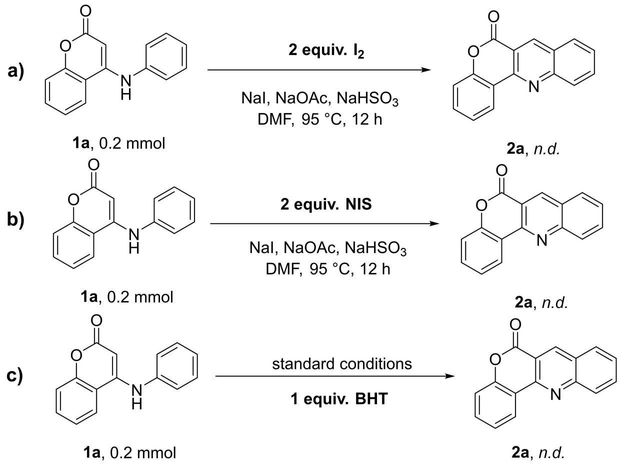
Scheme 4. Gram scale experiment.

The scalability of electrooxidative metal-free cyclization with DMF as the methine source was evaluated in 8.0 mmol scale (Scheme 4). The reaction furnished the desired product in 51% yield, which shows great potential of this electrooxidative metal-free cyclization.

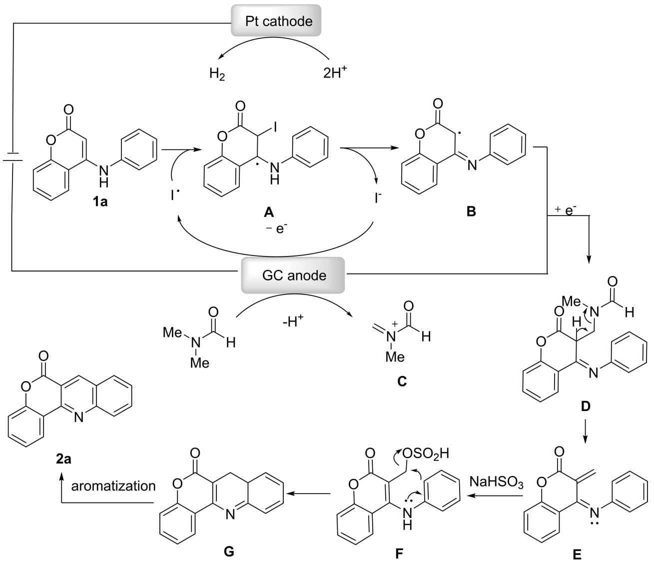
In order to understand the mechanism of the electrooxidative intramolecular C–C bond formation, a series of mechanistic studies was carried out (Scheme 5). Two equivalents of iodine (Scheme 5a) or *N*-iodosuccinimide (NIS) (Scheme 5b) were tested as the oxidant directly in the reaction of substrate **1 a**, since the iodide could be oxidized under electrochemical conditions. In the absence of electricity, neither iodine nor NIS as the oxidant can be used to obtain the desired product **2 a**. Radical inhibition experiment with butylated hydroxytoluene (BHT) was carried out to probe whether the reaction occurred by a radical reaction pathway or not. As was expected, no desired product could be observed by adding one equivalent of BHT, which indicates that the radical involves mechanism (Scheme 5c). We could infer from the results of parallel experiments that the methine group of the cyclization product should be from the methyl group on DMF (Figure S1, see SI).

Furthermore, we performed cyclic voltammetry (CV) experiments to gain further insights into this reaction mechanism (Figure S2, See SI). Substrate **1 a** was oxidized within several waves at 1.10 V_{SCE}, 1.55 V_{SCE}, and 1.90 V_{SCE}. When NaOAc, $NaHSO_3$, NaI, and DMF were added, the oxidation potential of the mixture was 1.60 V_{SCE}, which reduced the partial oxidation potential of **1 a** and promoted the reaction.

Based on our studies, a plausible mechanistic pathway was proposed in Scheme 6. In the first step, the anodically generated iodine radicals attack substrate **1 a** to afford intermediate A, which is converted to intermediate B after releasing iodine anion. At the



Scheme 5. Controlling experiments.



Scheme 6. A plausible mechanistic pathway.

same time, the iminium intermediate C, generated through anodic oxidation from DMF undergoes an electrophilic attack to form intermediate D. Then, one molecule of MeNHCHO is released to obtain intermediate **E**, which was attacked by $NaHSO_3$ to obtain intermediate **F**. Next, intramolecular cyclization enables the generation of the dihydroquinoline intermediate **G**, followed by an aromatization of anodic oxidation to furnish the desired product **2a**. Meanwhile, protons are reduced on the cathode, releasing molecular hydrogen.

In conclusion, the metal-catalyst-free electrooxidative cyclization employing DMF as the methine group was developed. This strategy provided a general and practical method to obtain chromene-fused quinolines derivatives, which was milder, more efficient, and more environmentally-friendly than previous methods.

Experimental Section

Synthesis of Chromene-Fused Quinoline

In an undivided cell (10 mL) equipped with a stirring bar, a mixture of substrates **1** (0.20 mmol), NaI (0.20 mmol, 30.0 mg), NaOAc (0.20 mmol, 16.4 mg), $NaHSO_3$ (0.24 mmol, 25.0 mg) and anhydrous DMF (3.0 mL) were added. The cell was equipped with glassy carbon as the anode and platinum plate as the cathode. The reaction mixture was stirred and electrolyzed at a constant current of 10 mA at 95 °C for 12 h. Upon completion, the solvent was removed directly under reduced pressure to afford the crude product, which was further purified by flash column chromatography to afford the desired product.

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