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Electrochemically induced synthesis of quinazolinones *via* cathode hydration of *o*-aminobenzonitriles in aqueous solutions†

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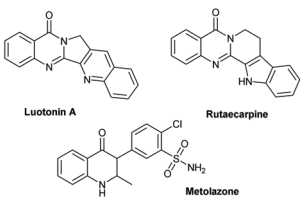
An efficient and practical electrochemically catalyzed transition metal-free process for the synthesis of substituted quinazolinones from simple and readily available *o*-aminobenzonitriles and aldehydes in water has been accomplished. I_2/base and water play an unprecedented and vital role in the reaction. By electrochemically catalysed hydrolysis of *o*-aminobenzonitriles, the synthesis of quinazolinones with benzaldehyde was first proposed. The synthetic utility of this method was demonstrated by gram-scale operation, as well as the preparation of bioactive *N*-(2,5-dichlorophenyl)-6-(2,2,2-trifluoroethoxy) pteridin-4-amine, which enables straightforward, practical and environmentally benign quinazolinone formation.

Quinazolinones are significant structural motifs due to their widespread occurrence in natural products and synthetic pharmaceuticals.[1] In particular, they are used as privileged scaffolds in drug discovery and are well known for their broad range of biological activities such as anti-malarial, anti-hypertensive, diuretic, anti-inflammatory, anticonvulsant and anticancer activities.[2] In recent years, numerous quinazolinone embedded natural products have been identified.[3] The cytotoxic alkaloid luotonin A and its derivatives infused with a quinazolinone moiety are clinically proven as anti-cancer agents (Scheme 1).[4] Based on the wide range of applications of quinazolinones and their analogues, many pathways for the synthesis of this similar structured compound have been described.[5]

The most classical and general protocols for the synthesis of quinazolinones are still performed through condensation between *o*-aminobenzamides and aldehydes followed by the oxidation of the resulting aminal intermediates (Scheme 2 path **a**).[6] Other strategies include the carbonylative annulation of *o*-aminoarylamides with a CO source and aryl halides (Scheme 2 path **b**).[7] In addition, the coupling reactions of transition-metal-catalyzed *o*-haloarylamides with a nitrogen source of amides or aldehydes as well as other annulation reactions have also received much attention recently (Scheme 2, path **c**).[8] Similarly, the reactions of alcohols with *o*-animoarylamides are also described (Scheme 2, path **d**).[9] Recently, Huang and coworkers have reported a series of synthesis strategies of N-heterocyclic compounds from benzyl ethers and *o*-aminobenzamides[9d] or alcohols and *o*-substituted anilines by electrochemical oxidative cyclization.[10] Although various N-heterocyclic compounds, especially quinazolinones and derivatives, can be obtained by the above methods, many still have inherent drawbacks that are difficult to overcome. The disadvantages of most methods include the use of volatile organic solvents, harsh reaction conditions, toxic catalysts ($KMnO_4$, DDQ, MnO_2, *etc.*), difficulty in product isolation, and low product yields.[11] Furthermore, some require transition metal catalysts/ligands that can lead to metal residue contaminants in the products and limit their applications in drug synthesis and can lead to the production of large amounts of waste.[1b,c,12] Therefore, developing direct, efficient, atom-economical and transition metal- and waste-free methods that can employ greener substrates is still of great importance in the field.

On the other hand, there is a large number of *o*-aryl benzamides on the market that are typically obtained from the



Scheme 1 Selected examples of biologically active quinazolinone derivatives.

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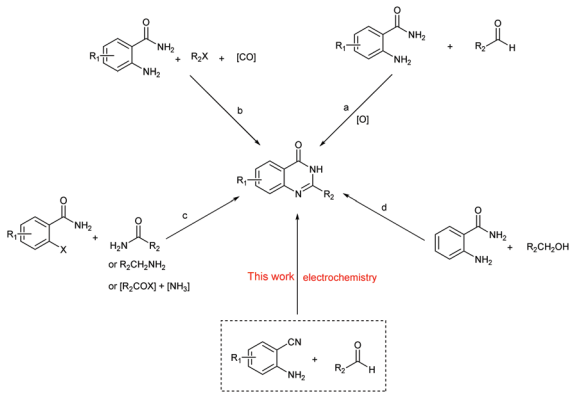


Scheme 2 General methods for the synthesis of quinazolinones.

hydrolysis of *o*-aryl benzonitriles. However, the direct synthesis of quinazolinones from *o*-aryl benzonitrile has been rarely reported.[13] Hence, the construction of the quinazolinone skeleton has aroused much interest employing *o*-aminobenzonitriles as the starting substrates, and several catalytic methods have been reported.[14–16] Recently, Li[16] has described a $[Cp^*IrCl_2]_2$-catalyzed one-pot two-step sequential selective hydration/condensation/acceptorless dehydrogenation of *o*-aminobenzonitriles and aldehydes to quinazolinones. However, excess amounts of *n*-butylaldoxime and aldehydes were required. We were therefore inspired to develop a more direct and environmentally friendly method for the preparation of quinazolinones from *o*-aryl benzonitrile.

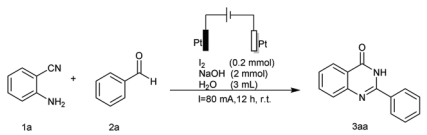
From the perspective of green chemistry, electrochemically induced catalysis has gained significant interest in organic synthesis due to its environment-friendliness, mild conditions, and low-energy consumption.[17] Due to the atom economy of electrochemical catalysis, we applied electrochemistry to the synthesis of quinazolinones from *o*-aminobenzonitriles as starting materials. We are pleased that the attempt at the electrocatalysis of *o*-aminobenzonitrile and benzaldehyde has yielded a satisfactory combination.

Moreover, owing to the increasing environmental concerns, water as reaction medium is generally considered as a cheap, safe, and environmentally benign alternative to non-natural solvents.[18] From both synthetic and environmental points of views, the development of efficient homogeneous catalytic systems for the acceptorless hydration of nitriles to the corresponding primary amides to nitrogen-containing heterocycles in water is apparently highly desirable.

As part of our ongoing research program toward the development of economically sustainable green synthetic methodologies,[19] herein, we demonstrate that I_2/base could be used as an efficient catalytic system for one-pot novel electrochemically induced tandem reaction leading to quinazolinones from *o*-aminobenzonitriles as starting materials. Iodine has been shown to be more low-cost and more in line with the concept of green chemistry than several metal catalysts.[20] Recently, some strategies have shown good activity with a heterogeneous catalyst for the synthesis of quinazolinones in the presence of I_2 with a low dose.[21] For example, recently, the Chang group has developed a one-pot I_2-mediated annulation reaction of substrates containing diamino groups and aldehydes *via* oxidative C–N bond formation.[21a] Thus, the present method represents substantial improvements over the earlier reported methods from the green and sustainable chemistry point of view.

We started the research by investigating the model reaction of *o*-aminobenzonitrile (**1a**) and benzaldehyde (**2a**) (Table 1). Various parameters, such as the nature of the bases, the type of solvents, reaction time and so on, were screened in order to optimise the reaction conditions. The reaction was initially carried out in a mixture of acetonitrile and water with different oxidants using NaOH (2 mmol) as the base and Pt as the electrode under an 80 mA current (Table 1, entries 1–3). The results showed that, with the change of the oxidant action, the yield of product **3aa** could be decreased accordingly, with I_2 giving the highest yield of **3aa** (Table 1, entry 1). The nature of the base used for the reaction was found to have much influence on the yield (Table 1, entries 4 and 5). Further evaluation of solvents revealed that water as the solvent was the best and environment friendly choice (Table 1, entries 6 and 7). In addition, when the time of reaction was halved, the yield of **3aa** was reduced to 73% (Table 1, entry 8). The increase in time did not have much effect on the product (Table 1, entry 9). Subsequently, the investigation of different electrodes showed that platinum plate electrodes were the best choice for both anode and cathode (Table 1, entries 10 and 11). Lower reaction yields were obtained when the current was halved or in its absence (Table 1, entries 12 and 13). It is worth noting that changing the constant current has a significant impact on the reaction.

Table 1 Optimization of the reaction conditions[a]

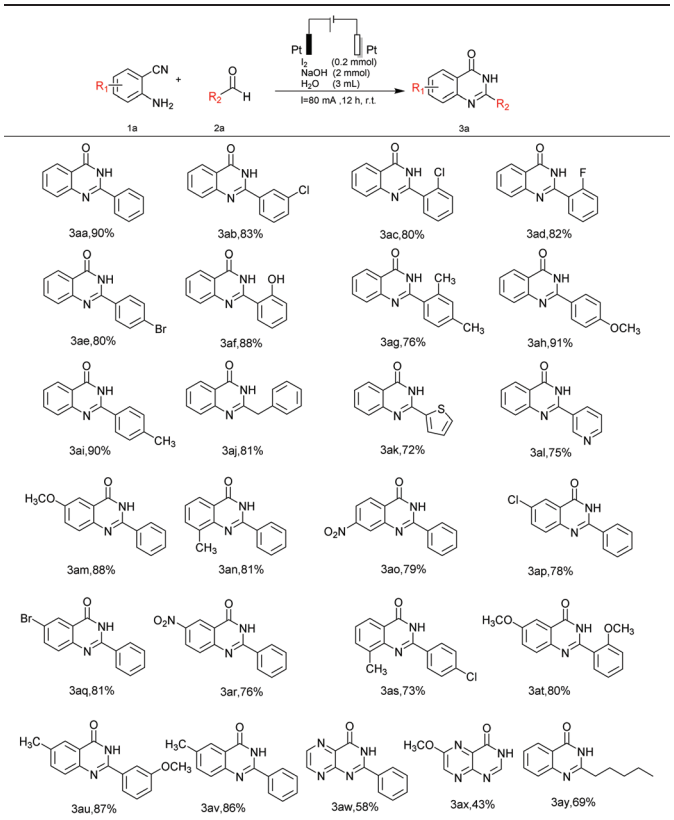


Entry	Variations from the standard conditions	Yield[b] (%)
1	None	90
2	CuI instead of I_2	23
3	KI instead of I_2	68
4	KOH instead of NaOH	75
5	Cs_2CO_3 instead of NaOH	72
6[c]	DMSO/H_2O (1/2) instead of H_2O 3 mL	86
7[d]	THF/H_2O (1/2) instead of H_2O 3 mL	79
8	6 h instead of 12 h	73
9	24 h instead of 12 h	90
10	C (−) instead of Pt (−)	54
11	C (+) instead of Pt (+)	77
12	40 mA instead of 80 mA	67
13	No electric current	Trace

[a] Standard conditions: **1a** (0.7 mmol), **2a** (0.5 mmol), I_2 (0.2 mmol), NaOH (2 mmol), undivided cell, Pt anode (1 cm × 1 cm), Pt cathode (1 cm × 1 cm), constant current (80 mA), H_2O (3 mL), r.t., 12 h. [b] Isolated yield. [c] DMSO/H_2O (1/2) = 3 mL. [d] THF/H_2O (1/2) = 3 mL.

Having the optimized reaction conditions in hand, we evaluated the substrate scope of the reaction. Various benzaldehydes (**2a**) were used as substrates to react with *o*-aminobenzonitriles (**1a**), and the results are summarized. The results listed in Table 2 demonstrate that several of the benzaldehydes tested underwent smooth transformation to afford the corresponding quinazolinones in high yields. In general, aldehydes bearing electron-donating groups on the benzene ring were more reactive than those with electron-withdrawing groups (Table 2, **3aa–3al**). This may be related to the fact that the presence of electron-donor groups makes the intermediate more stable. Benzaldehydes bearing electronically activating substituents such as –OMe, –OH and –Me could be coupled with *o*-aminobenzonitriles to give excellent yields of 91%, 88% and 90%, respectively (Table 2, **3ad**, **3af** and **3ai**). Halogen-substituted (–F, –Cl, and –Br) benzaldehydes (Table 2, **3ab–3ae**) were well tolerated. Moreover, thiophene-2-formaldehyde (Table 2, **3ak**) and pyridine-3-formaldehyde (Table 2, **3al**) were also suitable for the construction of quinazolines in 72% and 75% yields, respectively. Furthermore, inert aliphatic aldehydes such as benzene acetaldehyde and hexanal were converted with reasonably good yields of 81% and 69%, respectively (Table 2, **3aj** and **3ay**).

Table 2 Substrate scope[a,b]



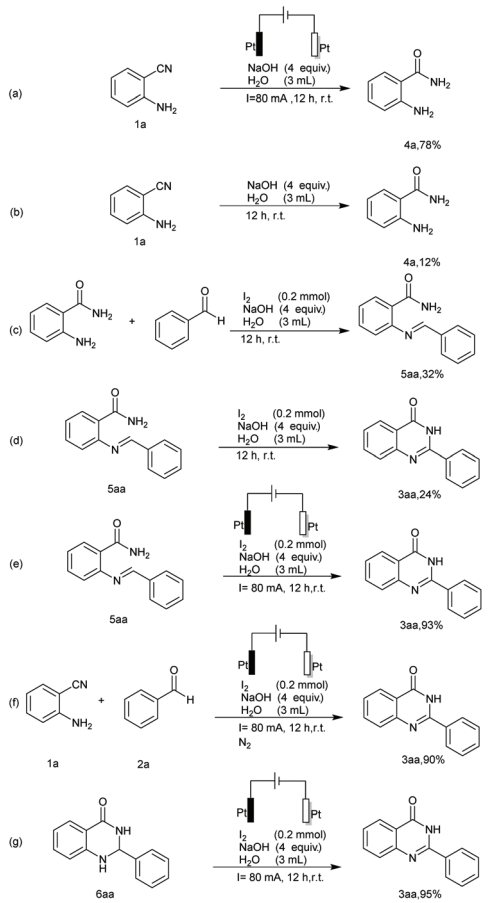
[a] Standard conditions: **1a** (0.7 mmol), **2a** (0.5 mmol), I_2 (0.2 mmol), NaOH (2 mmol), undivided cell, Pt anode (1 cm × 1 cm), Pt cathode (1 cm × 1 cm), constant current (80 mA), H_2O (3 mL), r.t., 12 h. [b] Isolated yield.

To explore the generality of this protocol, different *o*-aminobenzonitrile derivatives were tested, and the results are summarized in Table 2. Alkyl-substituted and halogenated aminobenzamides gave the corresponding quinazolinones with moderate to high yields (Table 2, **3am–3ax**). Similarly, electron-donating group-substituted *o*-aminobenzonitriles showed relatively higher catalytic activity compared with electron-withdrawing group substituted ones. As shown in Table 2, the benzonitriles **1a** with electron-donating substituents such as –OMe and –Me reacted with benzaldehyde smoothly to convert the corresponding quinazolinones in 88% and 86% yields, respectively (Table 2, **3am** and **3av**). Under the action of halogen electron-withdrawing groups such as –Cl and –Br, yields of 78% and 81% were obtained, respectively (Table 2, **3ap** and **3aq**). In addition, the strong electron-withdrawing group $-NO_2$ caused a large decrease in the yield of 79% and 76% (Table 2, **3ao** and **3ar**). Gratifyingly, pteridine compounds were also proven compatible with this method, providing **3aw** and **3ax** in decent yields.

We then scaled up the reaction, and the scalability of this amination of benzaldehyde and *o*-aminobenzonitrile was evaluated by performing a gram-scale synthesis of 2-phenylquinazolin-4(3*H*)-one. *O*-Aminobenzonitriles (7 mmol) and benzaldehydes (5 mmol) smoothly furnished the desired products in 81% yield (0.90 g).

To shed light on the reaction mechanism, some preliminary mechanistic studies were carried out. Control reactions were then performed to probe the reaction mechanism and the role of current in the reaction. Firstly, *o*-aminobenzonitrile **1a** effectively afforded *o*-aminobenzamide **4a** by hydration in the presence of NaOH (Scheme 3a). In the absence of current, the amount of **4a** produced by nitrile hydrolysis would decrease (Scheme 3b). In order to confirm the involvement of current in the reaction, we removed the current from the reaction system. The amount of either **1a** and **2a** generated or the amount of **3aa** generated by the direct reaction of **5aa** decreased dramatically (Scheme 3c and d). Subsequently, **5aa** under the 80 mA circuit conditions provided the expected product **3aa** (Scheme 3e). This suggested that the condensation of **1a** and **2a** or **5aa** can readily take place to give the product **3aa**, but current is the key to obtain high yield. Under the protection of nitrogen, the yield of product **3aa** did not change greatly, indicating that oxygen did not play a big role in this reaction (Scheme 3f). The intermediate **6aa** was oxidized under the standard reaction conditions to afford the desired product **3aa** after 12 h which supported the involvement of dihydroquinazolinone in the proposed mechanistic cycle (Scheme 3g).

The reaction was then examined by cyclic voltammetry (CV) (Fig. 1). Using a glass electrode as the working electrode, platinum wire as the opposite electrode, and a SCE as the reference electrode, the scanning rate is 0.1 V s^{-1}. An oxidation peak was observed for the reaction of a mixture of **1a** and NaOH at E_p = +1.23 V, in the region of 0.0–2.0 V. In addition, a mixture of **1a**, **2a** and NaOH showed an oxidation peak at E_p = +1.23 V with a slight increase of current (Fig. 1, curve **b**). These results indicate that **1a** reacts easily with **2a** when it interacts with NaOH.



Scheme 3 Control reactions.

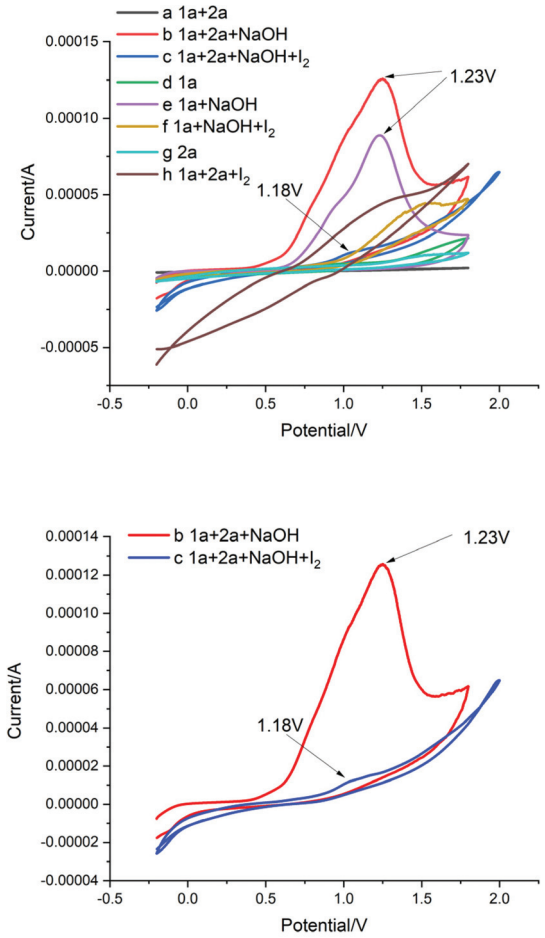


Fig. 1 Cyclic voltammetry experiments were performed at room temperature in a standard three electrode system.

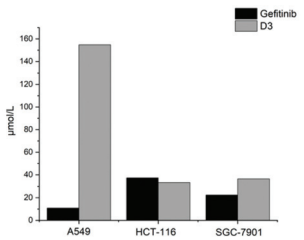
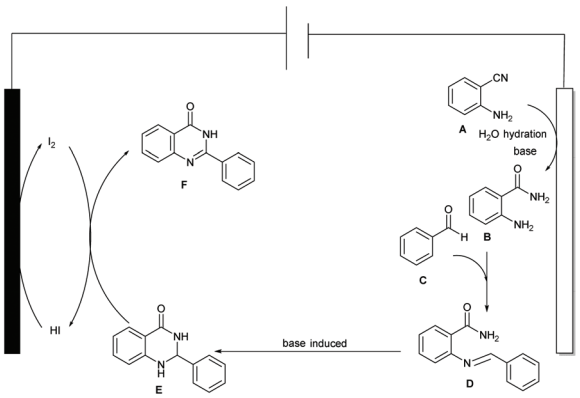


Fig. 2 *In vitro* inhibitory data of target compounds against A549, HCT-116 and SGC-7901 cell lines.

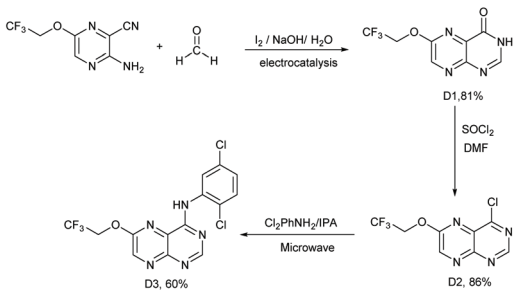
It is noteworthy that the oxidation potential of the mixture of **1a**, **2a** and NaOH decreased from 1.23 V to 1.18 V in the presence of iodine (Fig. 2, curves b and c). It turns out that iodine promotes further reactions of **1a**, **2a**, and NaOH.

On the basis of the above results and control experiments, a plausible mechanism is proposed in Scheme 4. Firstly, *o*-aminobenzonitriles **A** converted into **B** on the cathode *via* hydration and NaOH.[21] Then, **B** can readily react with **C** to yield the imine **D**, which is converted to **E** in the presence of the base, and then **E** oxidizes and dehydrogenates to afford the desired product **F**, and iodide ions, under the action of the anode along with the regeneration of iodine anions, to complete the catalytic I^- cycle.[22a,23,24]

A pteridine compound is a kind of widely existing aromatic compound, similar to quinazoline, which acts as an inhibitor of tyrosine kinase and shows good specificity and inhibitory activity on tumour cells. Secondly, the introduction of fluorine-containing substituents into drugs has been shown to increase the anti-tumoural activity. Based on this, we applied this electrocatalytic system to the synthesis of Gefitinib analogues. *N*-(2,5-Dichlorophenyl)-6-(2,2,2-trifluoroethoxy)pteridin-4-amine (**D3**) was synthesized from 3-amino-6-(2,2,2-trifluoroethoxy)pyrazine-2-carbonitrile in a total yield of 60% (Scheme 5). The compounds were then used in MTT assay, and the results are shown in Fig. 2. **D3** inhibited human non-small-cell Lung Carcinoma Cells A549, human colon cancer cells HCT-116 and human gastric cancer cells SGC-7901 (which were all obtained from the Cell Bank of the Chinese Academy of Sciences) to different degrees. As shown in Fig. 2, the results showed that **D3** displayed inhibitory effects against HCT-116 cell lines with IC_{50} values ranging from 33.48 μmol L^{-1} to 154.8 μmol L^{-1}, and the IC_{50} values of **D3** in A549 and SGC-7901 cell lines were less than those of gefitinib.



Scheme 4 Mechanistic pathway.



Scheme 5 Synthesis of *N*-(2,5-dichlorophenyl)-6-(2,2,2-trifluoroethoxy)pteridin-4-amine (**D3**).

Conclusions

In summary, our team has developed a method for the one-pot synthesis of quinazolinones by the electrocatalytic hydrolysis of *o*-aminobenzonitrile and benzaldehyde for the first time. Water not only promoted the hydrolysis of nitrile but also was the solvent of the reaction system. Iodine-mediated non-metal catalytic reaction has the advantages of high efficiency and low energy consumption. A variety of substituted quinoline and quinazolin-4(3*H*)-ones were synthesized using a low dose of iodine as the catalyst from a variety of cheap and readily available starting precursors. Since a wide range of substrates can be used to prepare substituted quinazolinones without contamination by transition metal residues, this method may be of potential application in pharmaceutical synthesis. Further application of the electrochemical synthesis strategy in organic synthesis is underway in our laboratory.

Conflicts of interest

There are no conflicts to declare.

Acknowledgements

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