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Electro-oxidative cyclization: access to quinazolinones via $K_2S_2O_8$ without transition metal catalyst and base†

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A $K_2S_2O_8$ -promoted oxidative tandem cyclization of primary alcohols with 2-aminobenzamides to synthesize quinazolinones was successfully achieved under undivided electrolytic conditions without a transition metal and base. The key feature of this protocol is the utilization of $K_2S_2O_8$ as an inexpensive and easy-to-handle radical surrogate that can effectively promote the reaction *via* a simple procedure, leading to the formation of nitrogen heterocycles *via* direct oxidative cyclization at room temperature in a one-pot procedure under constant current. Owing to the use of continuous-flow electrochemical setups, this green, mild and practical electrosynthesis features high efficiency and excellent functional group tolerance and is easy to scale up.

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To date, the synthesis of nitrogen heterocycles has drawn considerable attention over the years because of their broad utilities in numerous fields particularly organic chemistry, pharmaceutical science, agrochemistry, materials science, and life science.¹ Quinazolinone derivatives, as an important class of six-membered nitrogen-containing heterocyclic skeletons, represent one of the key structural motifs in numerous natural products and are known to have a wide range of biologically activities,² including antibacterial,³ anticancer,⁴ anti-inflammatory⁵ and anticonvulsant,⁶ in molecules such as deoxyvasicinone, glycosminine and⁷ drugs such as methaqualone and piritqualone (Fig. 1).⁸

Owing to a wide range of applications, several approaches have been developed for the synthesis of quinazolinones. Among these methods, the cyclization sequence of 2-amino-benzamides with aldehydes, followed by the oxidation of the aminal intermediate in the presence of stoichiometric or large excess amounts of non-renewable oxidants such as $KMnO_4$, MnO_2 , and DDQ (Scheme 1 path a),⁹ which were the most typical methodologies. To circumvent the problem of excessive oxidation, numerous protocols using Pd, Pt and Ir, *etc.* as noble

and heavy metal catalysts connected with a base for benzyl alcohol dehydrogenate oxidative cyclization is favored by chemists (Scheme 1 path b).¹⁰ However, while a wide range of stability and substrate applicability, most of the strategies mentioned above have not been applied in the industry owing to drawbacks such as the use of expensive transition metals and harsh reaction conditions.

On one hand, oxidative tandem cyclization processes are designed to be environmentally advantageous for the dramatic reduction of waste and costs, but most of them usually need metal-catalysts or strong oxidants.¹¹ Moreover, an oxidant is usually invariably used in these transformations for the regeneration of the catalyst compared to a non-metallic catalyst. On the other hand, numerous non-metallic oxidants that have found legible use in direct oxidative cyclization reactions include *p*-benzoquinone, I_2 , oxone, persulfates and so on. Among the varieties of non-metallic oxidants, $K_2S_2O_8$ has emerged as a suitable inorganic oxidant for a wide array of oxidative transformations due to its low cost, stability, nontoxic

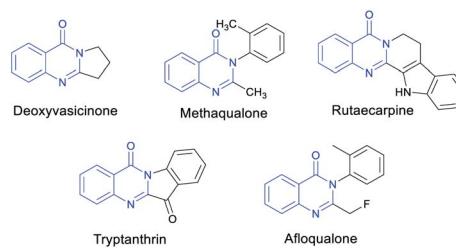


Fig. 1 Bioactive molecules containing quinazolinone skeletons.

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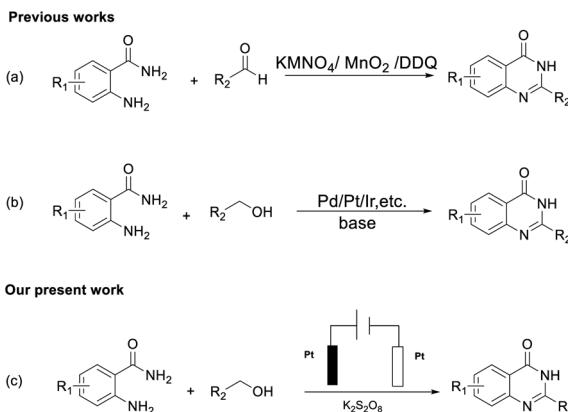
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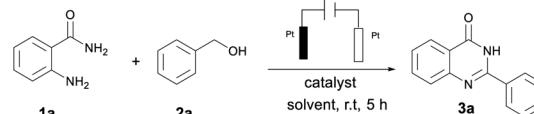


Scheme 1 Examples for synthesis of quinazolinones and our work.

nature, weakly pollutant, and easy-to-handle.¹² Recently, Yang¹³ *et al.* reported an approach for the synthesis of various phosphorated indolines *via* a copper-catalyzed radical cascade cyclization reaction with oxidant $K_2S_2O_8$. It is well known that the $K_2S_2O_8$ catalytic system has been well studied for the aerobic oxidation of alcohols and amines.¹⁴ Nonetheless, the oxidative tandem cyclization by only employing $K_2S_2O_8$ as the oxidant without any additives or base is rarely explored. Recently, electrochemical synthesis has gained considerable attention because of its major advantages of better environmental and economic acceptance compared to the traditional chemical and photochemical processes.¹⁵

Taking advantage of the highly oxidizing or reducing power of the excited state of electrogenerated ion radicals, it allows to perform thermodynamically difficult redox reactions. To the best of our knowledge alcohols are known as more available and stable and less toxic chemicals instead of the above-mentioned aldehydes.¹⁶ Furthermore, based on our previous reports on electrochemical synthesis and non-metallic oxidants, which show high reactivity towards quinazolinone synthesis or acceptorless dehydrogenation of primary alcohols to aldehydes.¹⁷ Herein, we present a high catalytic selectivity and metal co-catalyst free and base free protocol for the synthesis of quinazolinones from 2-aminobenzamides and benzyl alcohols *via* aerobic oxidative cyclization in an undivided cell (Scheme 1 path c).

At the onset of our investigation, 2-aminobenzamide **1a** with benzyl alcohol **2a** were chosen as the model substrates to optimize the reaction conditions (Table 1). Initially, different catalysts such as DTBP, $Na_2S_2O_8$, $(NH_4)_2S_2O_8$, and $K_2S_2O_8$ were examined, gave conversions of trace, and yields of 34%, 49%, and 94%, respectively (Table 1, entries 1–4). It can be found that $K_2S_2O_8$ resulted in better conversion in comparison to its congeners. One possible reason could be its better solubility in the organic medium than $Na_2S_2O_8$ and $(NH_4)_2S_2O_8$.¹² Furthermore, the solvent also plays a key role in the transformation. The product yield decreased when single and mixed ones t -BuOH, DMSO, DMF, or H_2O instead of MeCN, including that CH_3CN/H_2O was superior to the others (Table 1, entries 4–8). Further increasing the time to 6 h did not enhance the yield,

Table 1 Screening of the reaction conditions^a

Entry	Catalyst	Solvent	Current (mA)	Time (h)	Yield ^b (%)
1	DTBP	CH_3CN/H_2O	30	5	Trace
2	$(NH_4)_2S_2O_8$	CH_3CN/H_2O	30	5	34
3	$Na_2S_2O_8$	CH_3CN/H_2O	30	5	49
4	$K_2S_2O_8$	CH_3CN/H_2O	30	5	94
5	$K_2S_2O_8$	t -BuOH/ H_2O	30	5	39
6	$K_2S_2O_8$	DMSO/ H_2O	30	5	58
7	$K_2S_2O_8$	DMF/ H_2O	30	5	Trace
8	$K_2S_2O_8$	H_2O	30	5	29
9	$K_2S_2O_8$	CH_3CN/H_2O	30	6	93
10	$K_2S_2O_8$	CH_3CN/H_2O	30	4	65
11	$K_2S_2O_8$	CH_3CN/H_2O	40	5	86
12	$K_2S_2O_8$	CH_3CN/H_2O	20	5	64
13	$K_2S_2O_8$	CH_3CN/H_2O	—	5	Trace
14	—	CH_3CN/H_2O	30	5	Trace
15 ^c	$K_2S_2O_8$	CH_3CN/H_2O	30	5	82
16 ^d	$K_2S_2O_8$	CH_3CN/H_2O	30	5	67

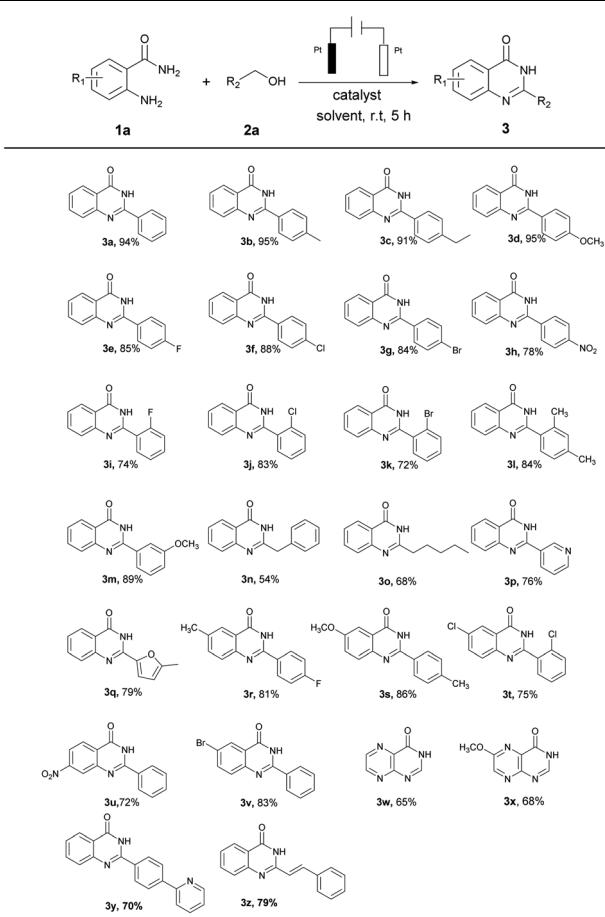
^a Reaction condition: undivided cell, Pt anode, Pt cathode, **1a** (0.5 mmol), **2a** (0.6 mmol), oxidant (0.6 mmol) and CH_3CN/H_2O (v/v = 2 : 1, 3 mL), under air at room temperature. ^b Yields were determined by HPLC. ^c $Pt(+)/C(-)$ instead of $Pt(+)/Pt(-)$. ^d $C(+)/Pt(-)$ instead of $Pt(+)/Pt(-)$.

while the yield decreased to 65% when this reaction was implemented at 4 h (entries 9 and 10). Nevertheless, the operating electric current did exert much influence on the reaction efficiency. Either increasing or decreasing the constant current led to poor yields (Table 1, entries 11 and 12). In addition, the control experiments demonstrated that $K_2S_2O_8$ and current were necessary for the reaction (Table 1, entries 13 and 14). Finally, other electrode materials such as graphite anode or graphite cathode were used and there was an obvious decrease on the yields of the reaction (Table 1, entries 15 and 16). Thus, we got the optimized reaction conditions for using $K_2S_2O_8$ (1.2 equiv.) in a mixture of 3 mL CH_3CN/H_2O (v/v = 2 : 1) with 30 mA under ambient air at room temperature for 5 h.

The substrate scope of this protocol was subsequently under the optimal conditions in Table 2. Benzyl alcohols with electron-donating groups ($-CH_3$, $-CH_2CH_3$, and $-OCH_3$) at the *p*-position afforded the corresponding products in 95%, 91%, and 95% yields, respectively (Table 2, **3b**–**3d**). Besides, electron-withdrawing groups ($-F$, $-Cl$, $-Br$, and $-NO_2$) gave the corresponding products in 85%, 88%, 84%, and 78%, yields, respectively (Table 2, **3e**–**3h**). For the *m*-position-substituted and *o*-position-substituted benzyl alcohols, both the electron-donating groups and electron-withdrawing groups led to the slightly lower yields of quinazolinones in 72–89% yields (Table 2, **3i**–**3m**). It is worth noting that aliphatic alcohol and hexanol were also compatible in the reaction to afford the corresponding products **3n** and **3o** in 54%



Table 2 Substrate scope of alcohols for electrochemical synthesis of quinazolinone derivatives.^{a,b}



^a Reaction condition: undivided cell, Pt anode, Pt cathode, **1a** (0.5 mmol), **2a** (0.6 mmol), $K_2S_2O_8$ (0.6 mmol), CH_3CN/H_2O (v/v = 2 : 1, 3 mL), constant current 30 mA, under air at room temperature for 5 h.

^b Isolated yield.

and 68% yields, respectively (Table 2, **3n** and **3o**). Interestingly, nitrogen heterocycles pyridin-3-ylmethanol and 5-methylfuran-2-ol also proceeded smoothly to afford 2-heterocyclic quinazolinones in 76% and 79% yields, respectively (Table 2, **3p** and **3q**).

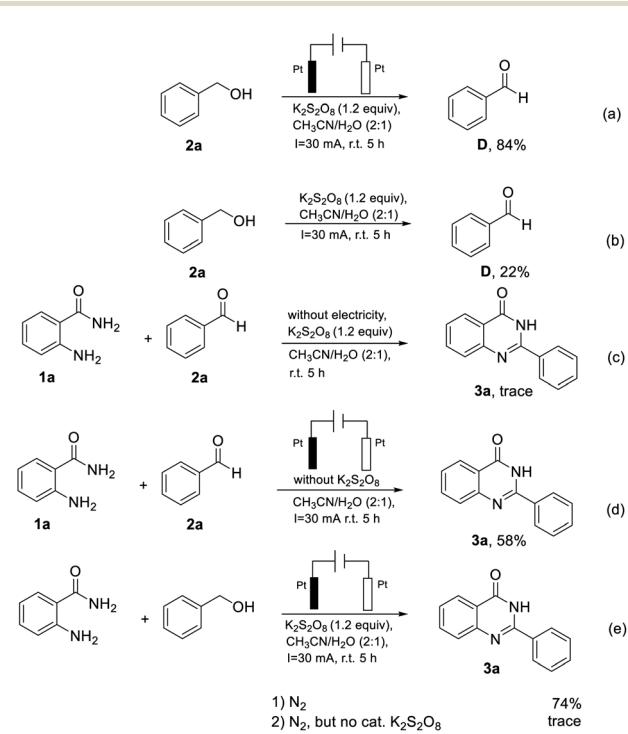
To further examine the scope of this reaction, various substituted 2-aminobenzamides were employed. Similarly, an electron-donating group-substituted 2-aminobenzamides showed relatively higher catalytic activity compared with those electron-withdrawing group substituted ones. Electron-withdrawing groups ($-NO_2$ and $-Br$) afforded the corresponding products in 72% and 83% yields, respectively (Table 2, **3u** and **3v**). Moreover, the electron-donating group ($-OCH_3$ and $-CH_3$) gave a higher yield than the withdrawing groups ($-Cl$) with 86% and 75% (Table 2, **3s**, and **3t**). Moreover, challenging substrates such as 3-aminopyrazine-2-carboxamide also afforded the desired pteridin-4(3*H*)-one in 65% and 68% yields (Table 2, **3w**, and **3x**). When 4-(pyridin-2-yl)benzaldehyde was

employed under the same reaction conditions, only 70% yield product was formed (Table 2, **3y**). Furthermore, highly catalytic activities were found in transformations of alkenyl alcohol to the corresponding products (Table 2, **3z**).

In order to demonstrate the practicality of this method, the gram-scale preparation of **1a** (8.00 mmol) from **2a** (9.6 mmol) with $MeCN/H_2O$ (v/v = 2 : 1, 25 mL) under the optimized reaction conditions was performed, which furnished the desired product **3a** in 84% yield (1.49 g). It reveals that the new procedure has significant advantages over numerous current methods for further practical applications.

In order to further understand the reaction mechanism, the following control experiments were carried out, as shown in Scheme 2. When benzyl alcohol **2a** was subjected to the standard conditions, 84% of benzaldehyde **D** was detected (Scheme 2a), while the yield of **D** was 22% in the absence of current (Scheme 2b). This result led us to confirm the presence of benzaldehyde **D** as an intermediate during the electrochemical reaction. When benzaldehyde **D** was added to the reaction system in the absence of electricity, trace amounts of **3a** was separated from the reaction mixture (Scheme 2c). Subsequently, benzaldehyde **D** was used as the starting material in the absence of $K_2S_2O_8$ and afforded **3a** with 58% yield, indicating that $K_2S_2O_8$ played a crucial role in this protocol (Scheme 2d). Furthermore, it could be quantitatively dehydrogenated into **3a** under N_2 condition with 74% yield, while a considerably lower conversion was achieved in the absence of $K_2S_2O_8$ affording trace amounts of desired product **3a** (Scheme 2, 3e).

The reaction was then examined by cyclic voltammetry (Fig. 2). The electrochemical response of 2-aminobenzamide exhibited its oxidation peak at $E_p = 0.88$ V vs. SCE and reduction



Scheme 2 Control experiments.



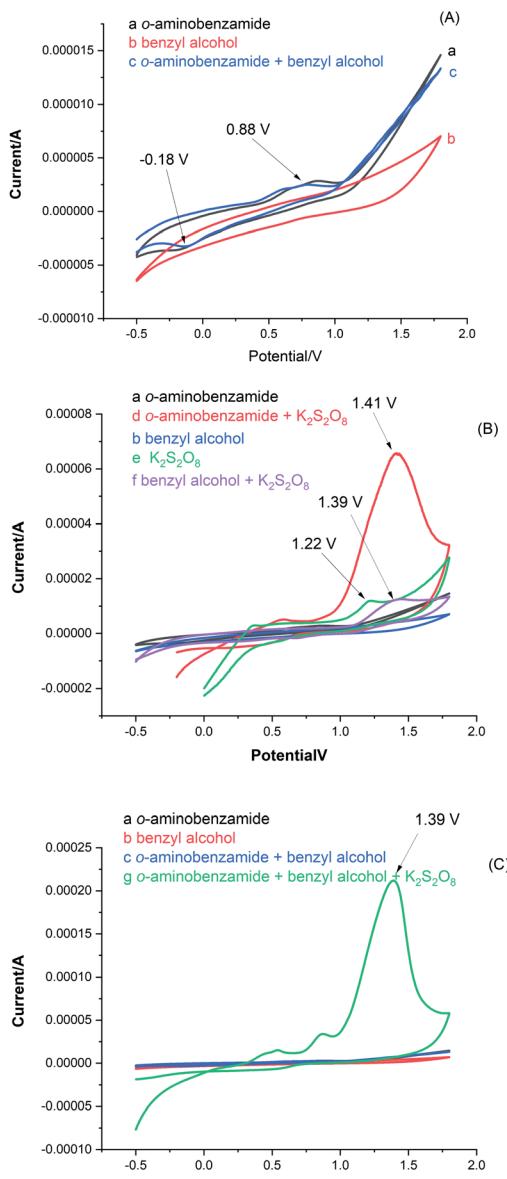


Fig. 2 Cyclic voltammograms of reactants and their mixtures in $\text{CH}_3\text{CN}/\text{H}_2\text{O}$ on a glassy carbon working electrode (diameter: 3 mm) with a Pt wire and calomel electrode as the counter and reference electrode, respectively, at a scan rate of 0.1 V s^{-1} . (A) (a) **1a**, (b) **2a**, (c) **1a** + **2a**; (B) (a) **1a**; (d) **1a** + $\text{K}_2\text{S}_2\text{O}_8$; (b) **2a**; (e) $\text{K}_2\text{S}_2\text{O}_8$; (f) **2a** + $\text{K}_2\text{S}_2\text{O}_8$; (C) (a) **1a**; (b) **2a**; (c) **1a** + **2a**; (g) **1a** + **2a** + $\text{K}_2\text{S}_2\text{O}_8$.

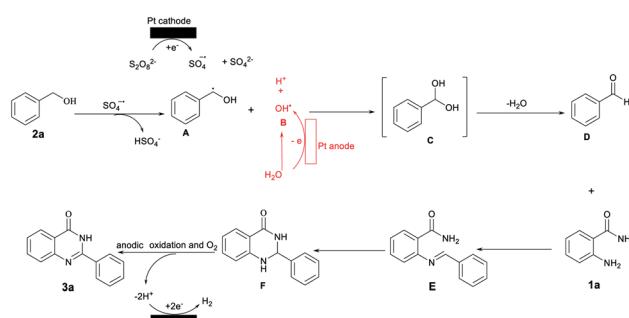
peak at $E_p = -0.18 \text{ V}$ vs. SCE (Fig. 2A, curve a). However, a mixture of **1a** and **2a** showed oxidation peak and reduction peak with a slight change in the catalytic current (Fig. 2A, curve c), which indicated that **1a** and **2a** cannot react spontaneously in the absence of $\text{K}_2\text{S}_2\text{O}_8$. When **1a** and $\text{K}_2\text{S}_2\text{O}_8$ were mixed, a new oxidation peak was observed for the reaction at $E_p = 1.41 \text{ V}$ vs. SCE, while a new oxidation peak at $E_p = 1.49 \text{ V}$ vs. SCE was observed of **2a** and $\text{K}_2\text{S}_2\text{O}_8$ mixture (Fig. 2B, curve f). These results indicate that **2a** was highly oxidizable than **1a** at constant currents in the presence of $\text{K}_2\text{S}_2\text{O}_8$. To clarify the mechanism of the reaction, a CV study on $\text{K}_2\text{S}_2\text{O}_8$ was carried out, an oxidation peak at $E_p = +1.39 \text{ V}$ with a remarkable

increase in the catalytic current (Fig. 2C, curve f and g). The above CV results indicate that $\text{K}_2\text{S}_2\text{O}_8$ as an oxidant promotes further reactions of **1a** and **2a** during the reaction.

On the basis of the above-mentioned results and the previous reports, a possible mechanism has been provided in Scheme 3.^{18–21} Initially, $\text{K}_2\text{S}_2\text{O}_8$ gains an electron at the cathode to form sulfate radical anions ($\text{SO}_4^{\cdot-}$) and anionic SO_4^{2-} ,¹⁸ which abstract hydrogens from benzyl alcohol **2a** to afford the α -hydroxyl- carbon radical **A**; this is an irreversible process.

In addition, H_2O can be oxidized to a hydroxyl radical **B** at the anode.¹⁹ Then, the radical cross coupling of hydroxyl radical **B** and carbon radical **A** gave an unstable intermediate **C** to afford benzaldehyde **D** by an elimination reaction of water.²⁰ To verify the above-proposed mechanism, DFT calculations were also performed and the results are shown in Fig. 3. First, the generated $\text{SO}_4^{\cdot-}$ can easily react with **2a** to generate **A** via a transition state (TS1) along with the release of HSO_4^- . Herein, the stability of **TS1** containing a seven-membered ring results in a very small energy barrier of $5.46 \text{ kcal mol}^{-1}$. Furthermore, the hydroxyl radical **B** will spontaneously react with **A** forming an intermediate **C**, which can further release a water molecule via a four-membered ring **TS2** to generate **D** with a moderate activation barrier of $36.34 \text{ kcal mol}^{-1}$. Subsequently, 2-amino-benzamides **1a** may react with benzaldehyde **D** to afford condensed imine intermediates **E**, which after cyclization is converted into 2,3-dihydroquinazolinones **F**.²¹ Finally, the anodic with O_2 oxy-dehydrogenation of **F** affords the desired product **3a**. On the other hand, the proton is reduced at the surface of the cathode to generate hydrogen molecules.^{21a,22}

Numerous nuclear nitrogen heterocyclic compounds similar to quinazolines also exhibit good antitumor activity. According to bioisosterism, pteridine would have similar biological activity. Based on this, the synthesis of Gefitinib analogues was taken as an example to illustrate the application of this reaction. The product *N*-(6-chloropyridin-2-yl)-6-ethoxypteridin-4-amine of Gefitinib analogues was taken as an example to illustrate the application of this reaction. The product *N*-(6-chloropyridin-2-yl)-6-ethoxypteridin-4-amine **A3** was obtained from the corresponding 3-amino-6-ethoxypyrazine-2-carboxamide (**A1**) in a total yield of 63% (Scheme 4). We also examined the anti-tumor activity of **A3** and Gefitinib on different cell lines via the MTT assay (Fig. 4). As shown in Fig. 4, the results showed that



Scheme 3 Proposed Mechanisms.

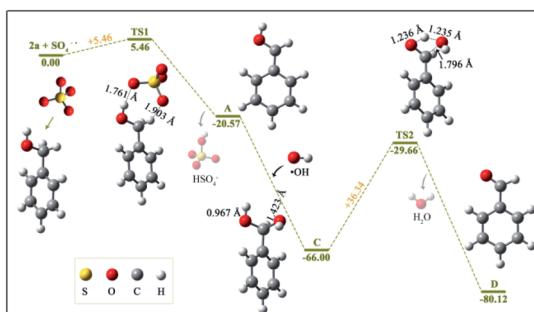
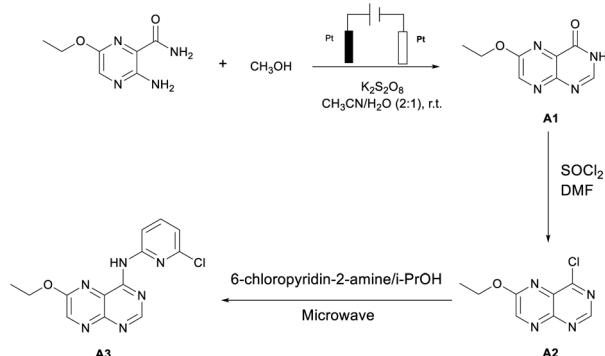
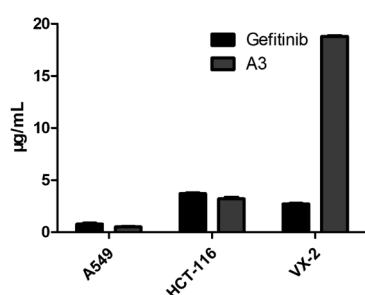


Fig. 3 DFT study during the oxidation of benzyl alcohol.

Scheme 4 Synthesis of *N*-(6-chloropyridin-2-yl)-6-ethoxypteridin-4-amine (A3).Fig. 4 *In vitro* inhibitory data of target compounds against A549, HCT116 and VX-2 cell line.

the IC₅₀ values of A3 in A549 and HCT116 cells (IC₅₀ = 0.5 µg mL⁻¹ in A549 cells, IC₅₀ = 3.2 µg mL⁻¹ in HCT116 cells, IC₅₀ = 18.8 µg mL⁻¹ in VX-2 cells) were less than Gefitinib, and the effect of A3 in A549 cell surpassed other groups.

Conclusions

In summary, an efficient electrocatalytic cascade reaction of 2-aminobenzamides and benzyl alcohols promoted by K₂S₂O₈ has been developed that affords quinazolines in moderate to excellent yields without any base. K₂S₂O₈ is better developed under electrochemical conditions by avoiding the use of other oxidizing agents. This method is of great value for the oxidative

cyclization of nitrogen-containing heterocycles from the viewpoint of green chemistry and organic synthesis. Moreover, DFT calculations were also performed to verify the mechanism that we proposed. Further synthetic application of this reaction for the efficient synthesis of various heterocyclic compounds is currently under investigation.

Conflicts of interest

There are no conflicts to declare.

Acknowledgements

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