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Synthetic Methods

Electrochemical Oxidative C–H/N–H Coupling between γ-Lactams and Anilines

Ming Gong and Jing-Mei Huang*[a]

Abstract: A mild method for the direct C–H/N–H coupling between γ-lactams and anilines through electrochemical oxidation has been developed. The protocol proceeded smoothly without metal catalysts at room temperature to afford γ-substituted γ-lactams in good yields. It has been revealed that the quasi-divided cell which provided high current density on the anode was crucial for this reaction.

Introduction

Carbon–nitrogen bond formation is important in synthetic organic chemistry, due to the high prevalence of the nitrogen-containing compounds in natural products, pharmaceutical-, and materials science.[1] Nucleophilic displacement of a leaving group and reductive amination of carbonyl compounds or imine alkylation are classical transformations.[2] The newly developed amination methods include the transition-metal-catalyzed Buchwald–Hartwig C–N coupling, hydroamination, and diamination of olefins, and allylic amination.[3] C–H functionalization is an efficient and highly atom-economic method to construct complicated structures and great progress has been made in this field in recent decades.[4] Recently, alkylation of amines to construct C–N bonds through oxidative sp^3 C–H bond activations has been explored by many groups. However, most of the nitrogen containing substrates were limited to amides, sulfonamides, azoles,[5] and anilines. Alkylation by the direct use of anilines, a group of substrates susceptible to oxidization, through oxidative sp^3 C–H bond activation remains challenging and typically the substrate scope is limited.[6]

Electrochemical oxidation provides an alternative and environmentally benign method to achieve the C–H functionalization.[7] It has been revealed that an amide or carbamate is oxidized on the anode to generate an iminium cation, which is trapped in situ by a nucleophile.[8] However, only compounds with high oxidation potentials, such as methanol and cyanide ions, have been employed as nucleophiles. Introduction of an electroauxiliary, for example, a silyl group in the α-position to a nitrogen atom significantly decreases the oxidation potential of an amide or carbamate.[7b] Thereby the scope of nucleophiles is expanded. Another strategy developed by Yoshida et al,[9] described as a "cation-pool" method, involves two discreet stages and the nucleophiles are added into the pot after the power has been switched off. In addition, the laminar-flow method has been shown to enable amide oxidations in the presence of electron-rich nucleophiles such as allylsilanes.[10] However, as far as operation is concerned, the direct oxidation of an amide or carbamate with nucleophile in one-pot is highly desirable. To the best of our knowledge, the direct trap of the iminium cation of an amide by using anilines as the nucleophile has not been reported in electrochemistry owing to the relatively low oxidation potential of anilines.[11] In continuation of our interest in the application of electrochemical methods to organic synthesis,[12] we report a general method for the C–N bond formation through one-step electrochemical C–H/N–H coupling between γ-lactams and anilines in a quasi-divided cell[13] to afford the corresponding products in good yields.

Results and Discussion

Initially, we selected 4-bromoaniline (**1a**) and *N*-methyl pyrrolidone (NMP, **2a**) as the test substrates. These two compounds were treated in a solution of NH_4ClO_4 (0.24 м) in 5 mL of THF in a one-compartment cell with a Pt wire anode and a Pt foil cathode under a constant current (20 mA) at room temperature for 2.5 h. Product **3a** was obtained in a yield of 56% (Table 1, entry 1). A number of common organic solvents (Table 1, entries 2, 3, 4, and 5) were examined and we found that 81% yield of product was obtained when CH_3CN was employed as solvent (Table 1, entry 2). Studies on the effect of current density revealed that an increase or decrease of the current led to the decrease of the product yield (Table 1, entries 6 and 7). Hence, 20 mA was the optimal current for this reaction. The electrolytes, such as NH_4ClO_4, NH_4BF_4, Bu_4NClO_4, and $LiClO_4$ were examined and it was found that NH_4ClO_4 was the most efficient for this electrochemical reaction (Table 1, entries 2, 8, 9 and 10). A lower yield of the product was obtained with the addition of an acid or a base (Table 1, entries 11 and 12). When the reaction was performed under an N_2 atmosphere, the yield dropped to 66% (Table 1, entry 13). On the

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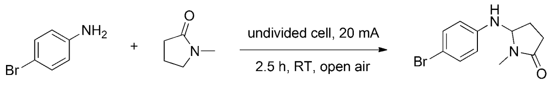
[a] M. Gong, Prof. Dr. J.-M. Huang
Key Laboratory of Functional Molecular Engineering of Guangdong Province, School of Chemistry and Chemical Engineering
South China University of Technology
Guangzhou, Guangdong 510640 (P. R. China)
E-mail: chehjm@scut.edu.cn

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

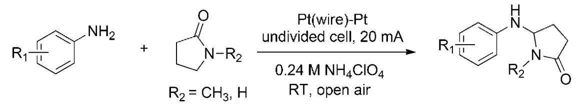


1a 2a 3a

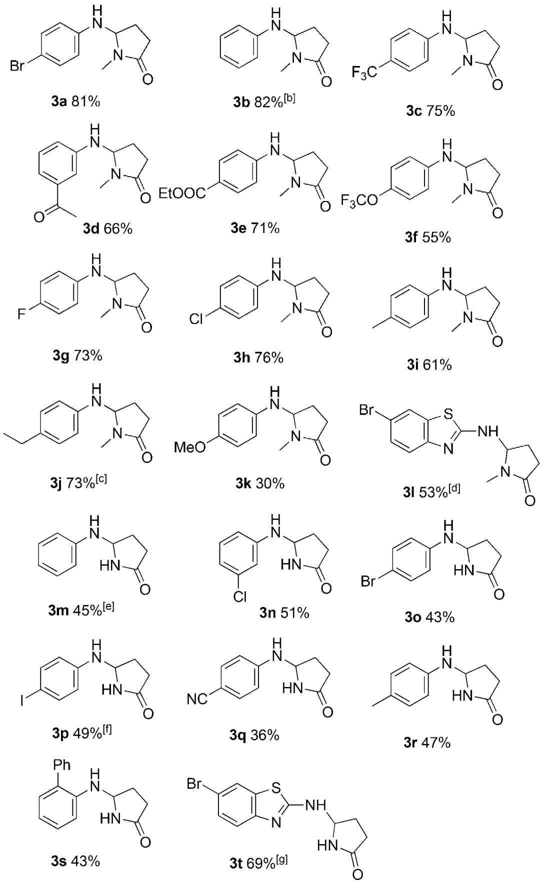
Entry	Solvent [mL]	Current [mA]	Electrolyte	Yield [%][b]
1	THF	20	NH_4ClO_4	56
2	CH_3CN	20	NH_4ClO_4	81
3	DMSO	20	NH_4ClO_4	18
4	CH_3NO_2	20	NH_4ClO_4	18
5	NMP	20	NH_4ClO_4	60
6	CH_3CN	15	NH_4ClO_4	69
7	CH_3CN	25	NH_4ClO_4	70
8	CH_3CN	20	NH_4BF_4	34
9	CH_3CN	20	$(Bu)_4NClO_4$	64
10	CH_3CN	20	$LiClO_4$	54
11[c]	CH_3CN	20	NH_4ClO_4	22
12[d]	CH_3CN	20	NH_4ClO_4	63
13[e]	CH_3CN	20	NH_4ClO_4	66
14[f]	CH_3CN	20	NH_4ClO_4	63
15[g]	CH_3CN	20	NH_4ClO_4	70
16[h]	CH_3CN	20	NH_4ClO_4	0

[a] Reaction conditions: **1a** (0.25 mmol) and **2a** (0.5 mL, 5.2 mmol) in solvent (5 mL) with 0.24 M electrolyte was electrolyzed at constant current (20 mA) in an undivided cell at RT, 2.5 h, open air. Anode: Pt (wire, diameter: 0.5 mm, height: 1.1 cm). Cathode: Pt foil (1×1.5 cm^2). [b] Yields of isolated products. [c] CH_3COOH (0.43 mmol). [d] $(Et)_3N$ (0.43 mmol). [e] Reaction performed under an N_2 atmosphere (balloon). [f] Reaction performed under an O_2 atmosphere (balloon). [g] Reaction performed under an air atmosphere (balloon). [h] No current.

Table 2. Substrate scope for synthesis of **3a–t**.[a]



1 2 3



3a 81%
3b 82%[b]
3c 75%
3d 66%
3e 71%
3f 55%
3g 73%
3h 76%
3i 61%
3j 73%[c]
3k 30%
3l 53%[d]
3m 45%[e]
3n 51%
3o 43%
3p 49%[f]
3q 36%
3r 47%
3s 43%
3t 69%[g]

[a] Reaction conditions: **1a** (0.25 mmol), **2a** (0.5 mL, 5.2 mmol), CH_3CN (5 mL), 0.24 M NH_4ClO_4 as electrolyte, 2.5 h, RT. Anode: Pt wire (diameter = 0.5 mm, height = 1.1 cm). Cathode: Pt foil (1×1.5 cm^2), constant current (20 mA), undivided cell, open air. Yield of isolated products. [b] CH_3CN:CH_3COOEt = 3:2, $(Et)_3N$ = 0.43 mmol, constant current (10 mA) and 10.5 h. [c] After 3 h. [d] After 5 h 40 min. [e] After 3 h. [f] After 1.5 h. [g] After 5 h.

other hand, the yield also decreased in the presence of O_2 (Table 1, entry 14). When the reaction was carried out under a balloon filled with air, the product was collected in a yield of 70 % (Table 1, entry 15). Finally, it was found that no reaction occurred with starting materials recovered when the reaction was performed in the absence of current (Table 1, entry 16).

With the optimized reaction conditions in hand, we then explored the substrate scope. As shown in Table 2, a wide range of anilines could afford the corresponding products in moderate to good yields. Anilines containing electron-withdrawing groups, such as CF_3, $COCH_3$, COOEt, and OCF_3, underwent the reaction to give good yields (**3c–f**). A series of halogen-containing anilines were tested. The results showed that halogen could be well tolerated in this reaction (**3a**, **3g**, and **3h**), providing a great opportunity for the further synthetic manipulations. Additionally, anilines containing electron-donating groups such as Me, Et, and OMe, afforded the corresponding products in slightly lower yields (**3i–k**).The reaction of heterocyclic aromatic primary amines, such as 6-bromobenzo[d]thiazol-2-amine also gave the respective product in good yield (**3l**). Next, to verify the utility of the methodology for pyrrolidinones, selected reactions were conducted using pyrrolidin-2-one. Pyrrolidin-2-ones could react with anilines bearing electron-deficient groups or electron-rich groups on the aromatic rings, gave corresponding products in moderate yields (**3n–r**). Sterically hindered substrates, for example, biphenyl-2-amine gave 5-([1,10-biphenyl]-2-ylamino)pyrrolidin-2-one in 43 % yield (**3s**). Heterocyclic aromatic primary amine also worked as a good partner with pyrrolidin-2-one to give the corresponding product in a yield of 69 % (**3t**). Notably, iodine substituted aniline could also give the desired products in 49 % yield (**3p**).

Cyclic voltammetry (CV) experiments (Figure 1, for more details see the Supporting Information) revealed the oxidation peak of NMP at a potential of 1.90 V versus Ag/Ag^+ and the oxidation peak of $4\text{-}MeC_6H_4NH_2$ at 0.77 V versus Ag/Ag^+.

Hence, the effects of the anode were investigated [Eq. (1), Scheme 1]. When a Pt foil (1×1.5 cm^2) anode instead of the Pt wire was employed, only a trace amount of desired product was obtained with all the anilines consumed. After reduction of the length of the Pt wire by one half, the yield decreased to 43 % with 37 % of anilines recovered. These results demonstrated that this reaction was very sensitive to current density.

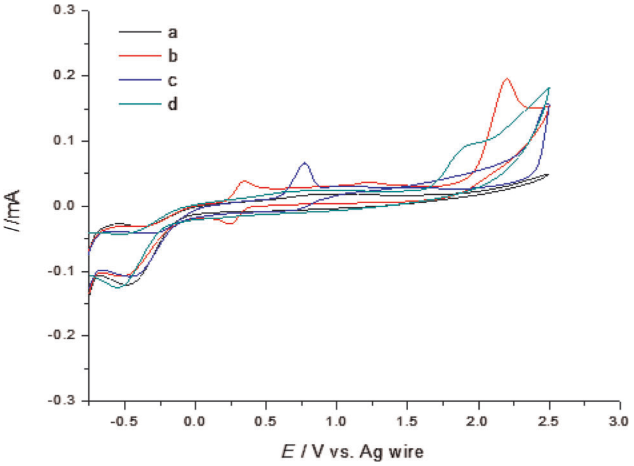
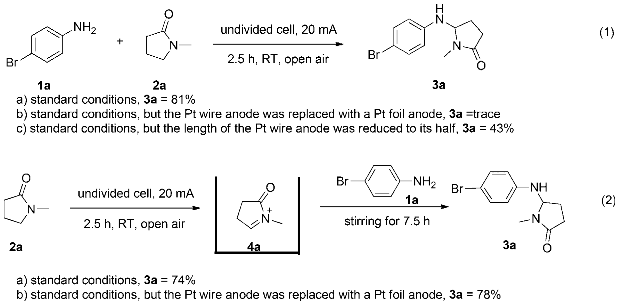


Figure 1. Cyclic voltammogram of 0.07 M NH_4ClO_4 solution in CH_3CN at RT: a) none, b) ferrocene (0.002 M), c) 4-$MeC_6H_4NH_2$ (0.009 M), d) NMP (0.004 M). The voltammogram was obtained with Pt wire as auxiliary electrode and Ag wire as pseudo-reference electrode. The scan rate was 0.1 V s^{-1} on a platinum disk electrode (d = 2 mm).



a) standard conditions, **3a** = 81%
b) standard conditions, but the Pt wire anode was replaced with a Pt foil anode, **3a** =trace
c) standard conditions, but the length of the Pt wire anode was reduced to its half, **3a** = 43%

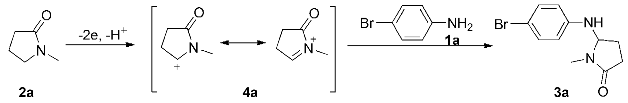
a) standard conditions, **3a** = 74%
b) standard conditions, but the Pt wire anode was replaced with a Pt foil anode, **3a** = 78%

Scheme 1. Studies on the effects of the anode.

When a certain length of Pt wire (diameter = 0.5 mm, height = 1.1 cm) was used as an anode (a cell design referred to as quasi-divided cell), the high current density on the anode might have favored the oxidation of NMP instead of anilines as the concentration of anilines was much lower than that of NMP. In addition, it is proposed that in the current reaction the acidic electrolyte, ammonium perchlorate, might have assisted to reduce the concentration of aniline in the double layer of anode by protonation of aniline.

Control reactions employing the "cation pool method" were carried out [Eq. (2), Scheme 1]. The electrochemical oxidation of NMP was initially conducted under constant current conditions (20 mA) at room temperature for 2.5 h. The current was then stopped followed by the addition of 4-bromoaniline (0.25 mmol) to the reaction mixture. After stirring the resultant mixture for 7.5 h, both the stepwise reactions using the Pt wire and Pt foil anodes gave good yields (74% and 78%, respectively).

Based on our experimental results and the precedent reports,[9b,14] a possible mechanism is depicted in Scheme 2. The NMP was oxidized on the anode and the iminum cation **4a** thereby generated. Then **4a** was trapped by 4-bromoaniline **1a** to give the target product **3a**. Reduction of NH4$^+$ to release hydrogen gas occurred on the cathode.



Scheme 2. Reaction pathway.

Conclusions

In conclusion, we have developed a mild and metal-free procedure for electrochemical C–H amination of N-methyl pyrrolidones and pyrrolidones in a quasi-divided cell. The commercially available anilines have been used directly as a nitrogen source in a one-step protocol. The present method provides an easy to operate route for the synthesis of nitrogen-containing products in good yields with a wide substrate scope. Further investigations into the mechanistic details and applications of this method are underway in our laboratory.

Experimental Section

In a round-bottomed flask (5 mL), 4-bromoaniline (0.25 mmol) and N-methylpyrrolidone (NMP, 0.5 mL,5.2 mmol) were dissolved in 5 mL CH_3CN with NH_4ClO_4 (0.24 M) as an electrolyte. The reaction flask was equipped with Pt wire as anode and Pt foil as cathode (1.5 cm^2). The solution was electrolyzed at a constant current (20 mA) for 2.5 h under open air at ambient temperature (7.5 F mol^{-1}). After electrolysis, it was quenched by water and extracted with ethyl acetate (3×15 mL). The combined organic extracts were washed with brine (5 mL), dried over $MgSO_4$, filtered and concentrated under vacuum. The crude product was purified by column chromatography on silica gel with ethyl acetate.

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Keywords: C–H/N–H coupling · electrochemistry · metal free · γ-lactams · anilines

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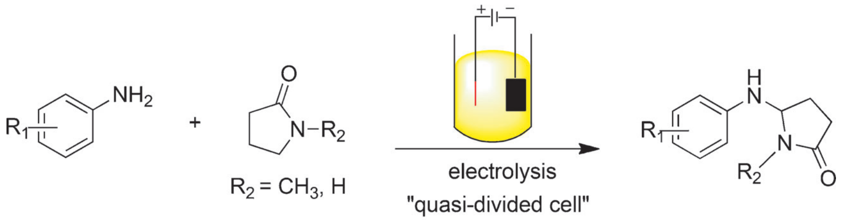

1 **2** **3**

One-pot lullaby: An easy to operate method for the electrochemical oxidative coupling of *N*-methyl pyrrolidone with anilines to synthesize *N*-aryl-γ-amino-γ-lactams was developed. Anilines were employed directly together with *N*-methyl-2-pyrrolidone in one-pot and no metal was needed. The reaction was carried out under open air at ambient temperature to afford the corresponding products in moderate to good yields with a wide range of functional groups tolerated.

Synthetic Methods

*M. Gong, J.-M. Huang**

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Electrochemical Oxidative C–H/N–H Coupling between γ-Lactams and Anilines