Advanced Synthesis & Catalysis

Accepted Article

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To be cited as: *Adv. Synth. Catal.* 10.1002/adsc.201801700

Link to VoR: http://dx.doi.org/10.1002/adsc.201801700

10.1002/adsc.201801700

COMMUNICATION

DOI: 10.1002/adsc.201((will be filled in by the editorial staff))

Metal- and Oxidant-free Electrosynthesis of 1,2,3-Thiadiazoles from Element Sulfur and N-tosyl Hydrazones

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Received: ((will be filled in by the editorial staff))

Supporting information for this article is available on the WWW under http://dx.doi.org/10.1002/adsc.201######.((Please delete if not appropriate))

Abstract. A metal- and oxidant-free electrochemical method for synthesizing 1,2,3-thiadiazoles by inserting element sulfur into N-tosyl hydrazones is reported. This electrochemical transformation engages electrons as reagents to achieve redox processes, and avoid excess oxidants. The cyclic voltammograms are examined to explore the mechanism of this electrolysis reaction.

Keywords: electrosynthesis; metal- and oxidant-free; NH_4I-mediated; element sulfur; 1,2,3-thiadiazoles

1,2,3-Thiadiazoles contain three heteroatoms that are common in nature and have attracted attention due to their utility in pharmaceutical agents[1] and materials.[2] Moreover, they have a series of biological activities, such as antibacterial, [3] antiviral, [4] antitumor,[5] antifungal,[6] and herbicidal growth regulating (plants)[7] activities (Figure 1).

Thus, considerable attention has been devoted to the development of many methods for synthesizing 1,2,3-thiadiazoles (Scheme 1). The traditional protocols include Hurd–Mori synthesis (Scheme 1, Path a),[8] Wolff synthesis (Scheme 1, Path b),[9] and Pechmann synthesis (Scheme 2, Path c).[10] The

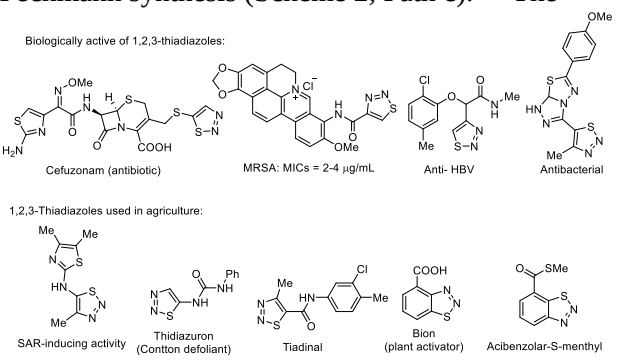
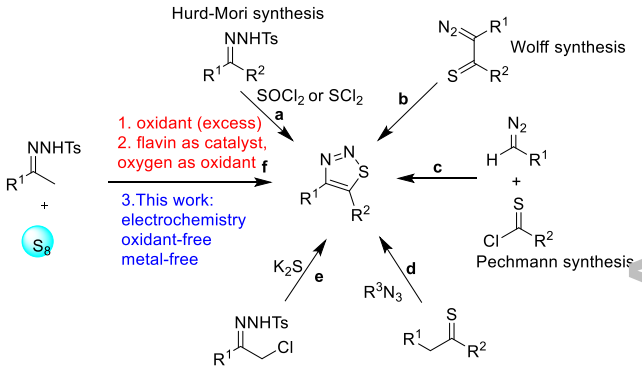


Figure 1. Bioactive molecules containing a 1,2,3-thiadiazole moiety.

diazotization of α-enolicdithioesters (Scheme 1, Path d)[11] and the reaction of α-chlorosulfonohydrazones and potassium sulfide in the presence of potassium carbonate with elevated temperatures (Scheme 1, Path e)[12] are two new methods for synthesizing 1,2,3-thiadiazoles. However, these methods require highly reactive reagents, such as SCl_2, $SOCl_2$, diazo/azide, or prefunctionalized compounds, that have safety concerns. To solve these problems, the direct oxidative cyclization of readily available N tosylhydrazones and sulfur into 1,2,3-thiadiazoles is a satisfactory strategy. Existing methods that use this strategy need excessive oxidants and complex flavins, and thus a relatively expensive process and/or generates copious amounts of waste (Scheme 1, Path f).[13] Thus, a mild, metal, oxidant-free, and sustainable synthetic method that overcomes the above limitations is needed.



Scheme 1. Different methods for the preparation of 1,2,3-thiadiazoles.

Organic electrosynthesis has recently drawn considerable attention as an environment-friendly and enabling synthetic tool because it employs mass free electrons as reagents to accomplish redox processes. [14] The replacement of metal catalysts by iodine has emerged as a green strategy in recent years.[15] In

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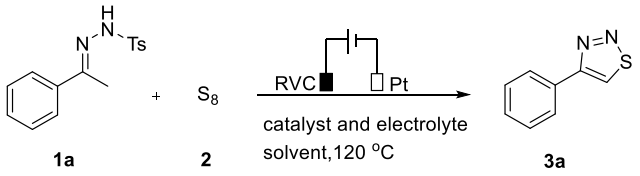
organic electrosynthesis, the anodic oxidation of iodide generates an iodine radical or cation, which can be interconverted during the reaction cycle. Thus, catalytic amounts of iodide salts are sufficient to complete the chemical transformation.[16] For example, we previously reported two dehydrogenation cross-coupling reactions with this green and sustainable catalytic system.[17] With our continued interest in the development of green methods for the synthesis of N-heterocycles,[18] we would like to develop a new method for forming 1,2,3-thiadiazoles from N-tosylhydrazones and sulfur using a metal- and oxidant-free electrochemical iodine-catalyzed strategy (Scheme 1, Path f).

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

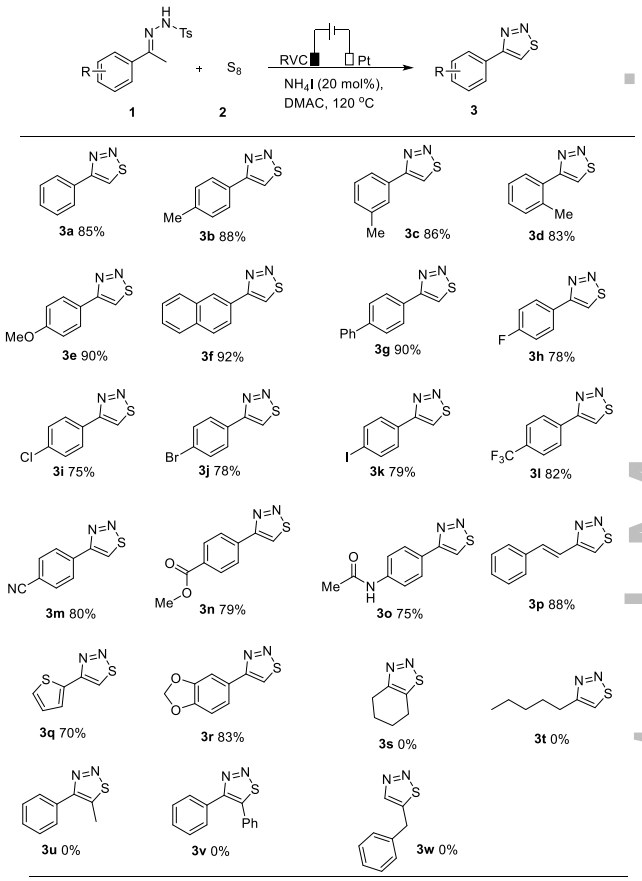


Entry	Catalyst	Solvent	Electrolyte	Yield (%)
1	TBAI	DMAC	$LiClO_4$	80
2	NaI	DMAC	$LiClO_4$	0
3	KI	DMAC	$LiClO_4$	0
4	NH_4I	DMAC	$LiClO_4$	84
5	NH_4Br	DMAC	$LiClO_4$	43
6	NH_4I	DMF	$LiClO_4$	56
7	NH_4I	1,4-Dioxane	$LiClO_4$	52
8	NH_4I	*n*-BuOH	$LiClO_4$	0
9	NH_4I	EtOH	$LiClO_4$	0
10	NH_4I	DMAC	-	85
11[c]	NH_4I	DMAC	-	0
12[d]	NH_4I	DMAC	-	0

[a]Reaction conditions: Reticulated vitreous carbon (RVC) as anode (100 PPI, 1 cm × 1 cm ×1.2 cm), Pt plate as cathode (1 cm × 1 cm), undivided cell, constant current = 10 mA, **1a** (0.5 mmol), catalyst (20 mol%), **2** (0.6 mmol), electrolyte (0.5 mmol), and solvent (6 mL) at 120 °C under air for 6 h. [b]Isolated yield. [c]Na_2S as the sulfur source (0.6 mmol), [d] $(NH_4)_2S$ as the sulfur source (0.6 mmol). DMAC = dimethylacetamide, DMF = N,N-Dimethylformamide.

We selected the tosylhydrazone derived from acetophenone **1a** and sulfur **2** as model substrates for the optimization studies (Table 1). Initially, the reaction of **1a** and **2** was carried out in an undivided cell with TBAI (20 mol%) and $LiClO_4$ (1 equiv) in DMAC at 120 °C for 6 h and offered annulation product **3a** in 80% yield (Table 1, Entry 1). NaI and KI failed to work, whereas NH_4I was optimal and provided the desired product **3a** with 84% yield (Table 1, Entry 1 versus Entries 2–4). These results were mainly due to the good solubility of NH_4I in DMAC. No progress was observed in the reaction with NH_4Br (Table 1, Entry 5). A solvent screen revealed that DMAC was the most suitable solvent for this transformation (Table 1, Entry 1 versus Entries 6–9). Changing the $LiClO_4$ to *n*-$BuNBF_4$ slightly decreased the reaction efficiency (77% yield, Table 1, Entry 9). Fortunately, the reaction could occur in the absence of electrolyte ($LiClO_4$) without reducing the yield of **3a** (85% yield, Table 1, Entry 10). However, Na_2S and $(NH_4)_2S$ could not participate in this transformation to afford the desired product **3a** (Table 1, Entries 11 and 12). Therefore, the best conditions for this transformation are constant current electrolysis at a current density of 10 mA/cm^2, undivided cell equipped with a RVC anode and a platinum plate cathode, and NH_4I (20 mol%) as the redox catalyst and electrolyte in DMAC at 120 °C for 6 h.

Scheme 2. Substrate scope.[a,b]



[a]Reaction conditions: Reticulated vitreous carbon (RVC) as anode (100 PPI, 1 cm × 1 cm ×1.2 cm), Pt plate as cathode (1 cm × 1 cm), undivided cell, constant current = 10 mA, **1** (0.5 mmol), NH_4I (20 mol%), **2** (0.6 mmol), and DMAC (6 mL) at 120 °C under air for 6 h. [b]Isolated yield.

With the optimal reaction conditions defined, we studied the substrate scope by varying the substituents of tosylhydrazones **1** (Scheme 2). Various tosylhydrazones were tolerated well. The electrolysis reaction exhibited excellent compatibility with several electron-donating groups at the phenyl ring, thereby providing a high yield of 4-aryl-1,2,3-thiadiazoles (Scheme 2, **3a**-**3g**). Tosylhydrazones **1c** and **1d** with electron-donating groups at the *meta* and

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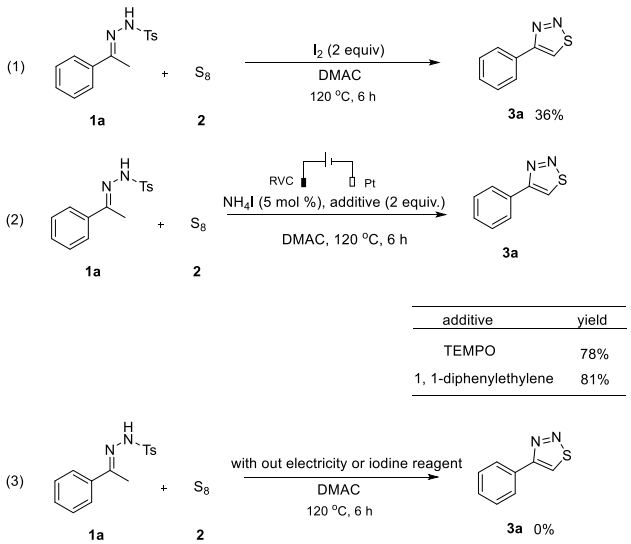
ortho aryl rings (**1c**: R = 3-Me; **1d**: R = 2-Me) reacted successfully and afforded the desired products **3c** and **3d** with yields of 86% and 83%, respectively (Scheme 2, **3c** and **3d**). Substrates bearing electron-drawing groups at the phenyl ring produced low yields (Scheme 2, **3h**-**3o**). Moreover, functional groups ester and acetamino were found suitable for procedure and gained desired products **3n** and **3o** with 79% and 75% yields, respectively. The alkene-bearing tosylhydrazone worked smoothly without transforming alkene to provide the corresponding product **3p** with 88% yield (Scheme 2, **3p**). Heteroaryl tosylhydrazones were subjected to electrolysis and formed the corresponding products **3q** and **3r** with 70% and 83% yields, respectively (Scheme 2, **3q** and **3r**). Unfortunately, this electrolysis reaction did not fit the aliphatic analogues of the N-Ts hydrazone under the standard conditions (Scheme 2, **3s** and **3t**). 4,5-Disubstituted 1,2,3-thiadiazoles and 5-benzyl-1,2,3-thiadiazole **3w** could not be synthesized with our method (Scheme 2, **3u**, **3v**, and **3w**).



Scheme 3. Controlled experiments.

According to previous reports, iodine anions can be oxidized to iodine free radicals or iodine cations at the anode.[16] To study the possible pathways, control experiments were performed. A reaction between **1a** and **2** was performed in the presence of I_2 (2 equiv.) in DMAC at 120 °C for 6 h in the absence of a current, and product **3a** was isolated with 36% yield (Scheme 3, Eq. 1). Subsequently, TEMPO or 1, 1-diphenylethylene, and substrates **1a** and **2** were performed under standard conditions, and the desired product **3a** was observed with 78%/81% isolated yield (Scheme 3, Eq. 2). The above results might exclude the presumption of a radical pathway. Control experiments without electricity or iodine reagent were also conducted, but the reactions failed to form the corresponding product **3a** (Scheme 3, Eq. 3).

To further investigate the possible reaction mechanism of this oxidant-free transformation, cyclic voltammogram was performed, and the results are shown in Figure 2. Curves b and c of Figure 2 show no apparent oxidation peak for sulfur **2** and tosylhydrazone **1a** in the 0.0−1.3 V region versus Ag/AgCl. The CV of NH_4I (curve d) exhibits two couples of reversible redox waves, with the oxidation peaks at 0.41 V (O_{x1}) and 0.75 V (O_{x2}) versus Ag/AgCl corresponding to the oxidation of iodide to form I_3^- and I_3^- to I_2, respectively.[16] Compared with curve d, no apparent change was observed for curve e, indicating that the in situ generated iodine could not oxidize sulfur **2**. By contrast, the mixture of tosylhydrazone **1a** and NH_4I exhibited an additional marked increase in catalytic current (curve f), and the peak currents of O_{x1} and O_{x2} increased dramatically from 19.22 to 58.79 μA, suggesting that NH_4I was the catalyst for this hydrazone transformation. As expected, the oxidation peaks and peak currents of the CV curve of the three components were only slightly higher than the curve of the mixture of **1a** and NH_4I (curve f versus curve g).

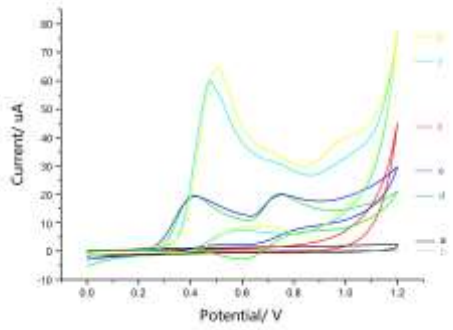
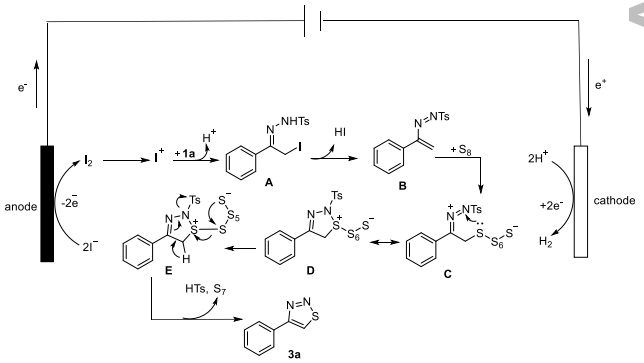


Figure 2. Cyclic voltammograms of reactants and their mixtures in 0.1 M $LiClO_4$/DMAC using a glassy carbon disk working electrode (diameter, 3 mm), Pt disk and Ag/AgCl (KCl saturated solution) as counter and reference electrode at 100 mV s^{-1} scan rate: (a) background, (b) **2** (10 mmol L^{-1}), (c) **1a** (10 mmol L^{-1}), (d) NH_4I (2 mmol L^{-1}), (e) **2** (10 mmol L^{-1}) + NH_4I (2 mmol L^{-1}), (f) **1a** (10 mmol L^{-1}) + NH_4I (2 mmol L^{-1}), and (g) **1a** (10 mmol L^{-1}) + **2** (10 mmol L^{-1}) + NH_4I (2 mmol L^{-1}).



Scheme 4. Plausible mechanism.

Based on the above results, a plausible mechanism was proposed (Scheme 4). At the anode, the electrochemical oxidation of $2I^-$ led to the formation of I_2, which easily generated I^- and I^+. The α-iodation of acetophenone tosylhydrazone **1a** formed intermediate **A**, and the elimination of HI provided azoalkene **B**. In these processes, the released iodine anions could be further oxidized to complete the cycle of the reaction. S_8 was added to azoalkene **B** to produce zwitterionic **C**, which underwent cyclization that furnished intermediate **E**. Finally, the elimination of S_7 and TsH led to the formation of the desired product **3a**. At the cathode, reduction occurred to deliver hydrogen, which completed the electrochemical cycle.

In conclusion, we developed a metal- and oxidant-free electrosynthesis of 1,2,3-thiadiazoles from sulfur and N-tosylhydrazones. Readily available raw materials, good yields, and an experimentally convenient catalytic process make this protocol practical and attractive. Importantly, the present electrochemical protocol employs mass free electrons as reagents to oxidize iodine anion to I_2 without using excess amounts of external oxidant, thereby providing an environmentally benign method for diverse iodine-catalyzed transformations that have conventionally required stoichiometric oxidants. This electrochemical transformation is being studied further in our laboratory.

Experimental Section

N- tosylhydrazones **1** (0.5 mmol, 1.0 equiv), sulfur **2** (0.6 mmol, 1.2 equiv), and NH_4I (0.1 mmol, 20 mol%) were placed in a 10-mL three-necked round-bottomed flask. The flask was equipped with a condenser, a RVC (100 PPI, 1 cm × 1 cm × 1.2 cm) anode and a platinum plate (1 cm × 1 cm) cathode. DMAC (6 mL) was added. Electrolysis was performed at 120 °C using a constant current of 10 mA until the complete consumption of the substrate (monitored by TLC, approximately 6 h). The reaction mixture was cooled to room temperature. Water (30 mL) and ethyl acetate (30 ml) were added. The phases were separated, and the aqueous phase was extracted with ethyl acetate (2 × 30 mL). The combined organic solution was dried over anhydrous $MgSO_4$, filtered, and concentrated under reduced pressure. The residue was chromatographed through silica gel elution with ethyl acetate/petroleum ether to yield product **3**.

Acknowledgements

We thank the National Natural Science Foundation of China (21861006), Guangxi Natural Science Foundation of China (2016GXNSFEA380001 and 2016GXNSFGA380005) and State Key Laboratory for Chemistry and Molecular Engineering of Medicinal Resources (CMEMR2017-A02 and CMEMR2017-A07) for financial support.

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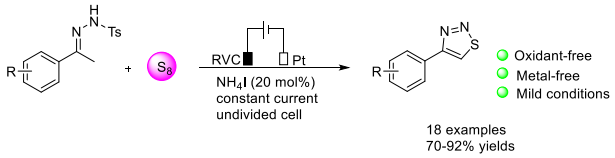

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