Advanced Synthesis & Catalysis

Accepted Article

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To be cited as: *Adv. Synth. Catal.* 10.1002/adsc.201901262

Link to VoR: http://dx.doi.org/10.1002/adsc.201901262

10.1002/adsc.201901262

COMMUNICATION

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

Electrochemical Chalcogenation of *β,γ*-Unsaturated Amides and Oximes to Corresponding Oxazolines and Isoxazolines

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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. The current report represents a transition-metal-free synthesis of oxazoline and isoxazoline derivatives by a tandem electro-oxidative chalcogenation-cyclization process. Both C–Se and C–S bond-forming protocols were developed without using any external oxidant and the reaction was performed at room temperature, open to the air. Using this methodology, 29 substituted oxazoline and 16 substituted isoxazoline derivatives were synthesized with up to 91% isolated yield.

Keywords: Electrochemistry; Chalcogenation; Oxazoline; Isoxazoline; Oxidation

Oxazolines[1] and isoxazolines[2] represent two branches of privileged heterocycle motifs numerously present in several biologically active natural products (Figure 1). These are the key scaffolds in many pharmaceuticals and agronomical industry. Furthermore, oxazoline core has widespread utilization as protecting group, chiral ligand and auxiliary in asymmetric synthesis and material science.[3] Therefore, elegant methodologies for the construction of these venerable heterocycles allure many synthetic and medicinal chemists. In last few decades, an extensive endeavor has been made to develop proficient methods for the synthesis of these five-membered heterocycles.[4]

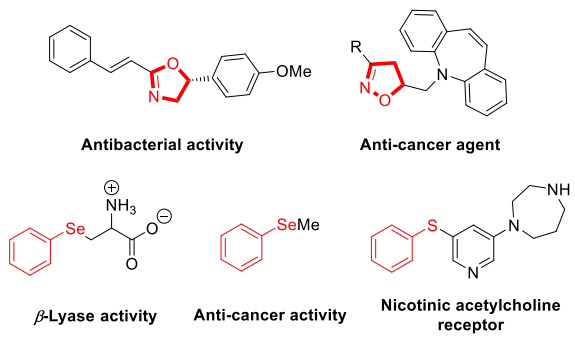
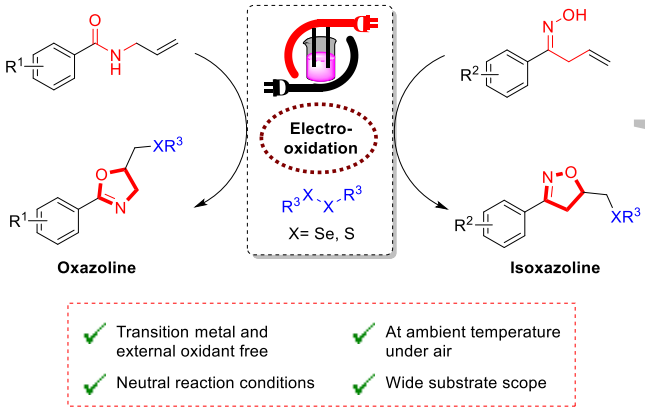


Figure 1. Biologically active a) oxazolines, isoxazolines and b) organochalcogens.

In recent years tandem oxidative cyclization reactions have been proven as an effective synthetic means for these core heterocycle skeletons.[5] *N*-Allyl amides and *β,γ*-unsaturated oximes are recognized as versatile precursors for oxidative cyclization to produce diversely functionalized oxazoline[6] and isoxazoline[7] frameworks. Regardless of the accomplished advancements, scopes of the above reactions have several shortcomings due to the use of harsh reaction conditions, stoichiometric amount of oxidants, the involvement of transition metals and prolonged reaction times. Therefore, to develop environmentally benign sustainable efficient synthetic methods are still in high demand.



Scheme 1. Electrochemical synthesis of chalcogenated oxazoline and isoxazoline scaffolds.

Organochalcogen moieties are the essential structural core found in many bioactive molecules and natural products.[8] Thus, C–S and C–Se bond forming reactions are in high demand. Furthermore, these chalcogen functionalities have potential applications in a wide number of organic transformations.[9] As

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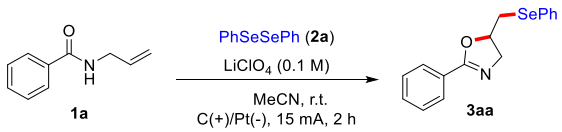
sulfur and selenium groups can be modified easily via oxidation or reduction, so incorporation of such atoms can lead to many useful conversions.[10] Due to the great potential of these ubiquitous structural motifs, tireless endeavors have been made towards the synthesis of organochalcogen compounds.[11] In the last decade various reports have been published to construct C–S and C–Se bonds using photocatalysis,[12] transition metal catalysis,[13] or by activating with different Lewis acids and Brønsted acids.[14] Despite the major progress, these methodologies have been largely restricted from some drawbacks like toxic and pungent reagents, harsh reaction conditions, excess use of oxidants, transition metals, and low scalability.[15] The majority of these reactions require super-stoichiometric amount of chalcogen source, resulting in low atom-economy and often causes undesirable byproducts. Therefore sustainable, oxidant and metal-free environment-friendly approach to synthesize organochalcogen moieties, especially attached with heteroaryl groups are still under-explored from the green chemistry perspective.

Although electro-organic synthesis was first established over 200 years ago,[16] in recent years it emerged as an attractive protocol for organoredox transformations due to its environmentally benign and economically viable features.[17] This electro-synthetic strategy is globally accepted as a smart alternative for stoichiometric redox reagents and successfully excluding chemical wastes and toxic byproducts.[18] However, fruitful electrochemical approach to synthesize C–chalcogen bond is underexplored.[19] In 2017, Aiwen Lei disclosed electrochemical thiolation of indole derivatives starting from thiol,[19e] but thiolation using disulfide is yet to be investigated. To the best of our knowledge oxidant free chalcogenation of *β,γ*-unsaturated oxime to form isoxazoline derivative under electrochemical conditions has not been reported so far. Given the importance of oxazoline and isoxazoline moieties, herein we report a sustainable method for the electro-oxidative chalcogenation of different amides[20] and oximes to synthesize chalcogen-functionalized heterocycles, in a mediator free undivided electrochemical cell (Scheme 1).

To ensure the synthetic utility of our electrochemical methodology, we commenced our study by treating *N*-allyl benzamide (**1a**) with diphenyl diselenide (**2a**) in an undivided cell equipped with graphite as electrodes. To our delight, 79% isolated yield of selenylated oxazoline **3aa** was achieved when electrolyzed in 0.1 M $LiClO_4$ containing acetonitrile solution (Table 1, entry 2). We further investigated the effect of different electrode materials. Replacing cathode as platinum plate instead of graphite increased the efficacy of the reaction to 91% isolated yield (Table 1, entry 1). The yield of the desired product was diminished slightly when platinum was employed both as anode and cathode (Table 1, entry 3). Subsequently, Ni foam cathode was found not very efficient in product formation (Table 1, entry 4). The following screening of the solvents, such as MeOH, MeCN/H_2O mixtures were investigated and found that MeOH is also equally effective while the mixture of MeCN/H_2O leads to a decrease in yield. Among the various electrolytes used, tetrabutylammonium tetrafluoroborate ends up with comparable yield to 87% with that of lithium perchlorate (Table 1, entry 7). Altering the output current to 10 mA or 20 mA proved to be less efficient (Table 1, entries 8 and 9). Different proportions of diphenyl diselenide (**2a**) were tested and a substoichiometric amount (0.8 equiv) was found to be the most effective. The desired oxazoline did not form when no electricity was run (entry 12).

Table 1. Optimization of the reaction conditions[a]



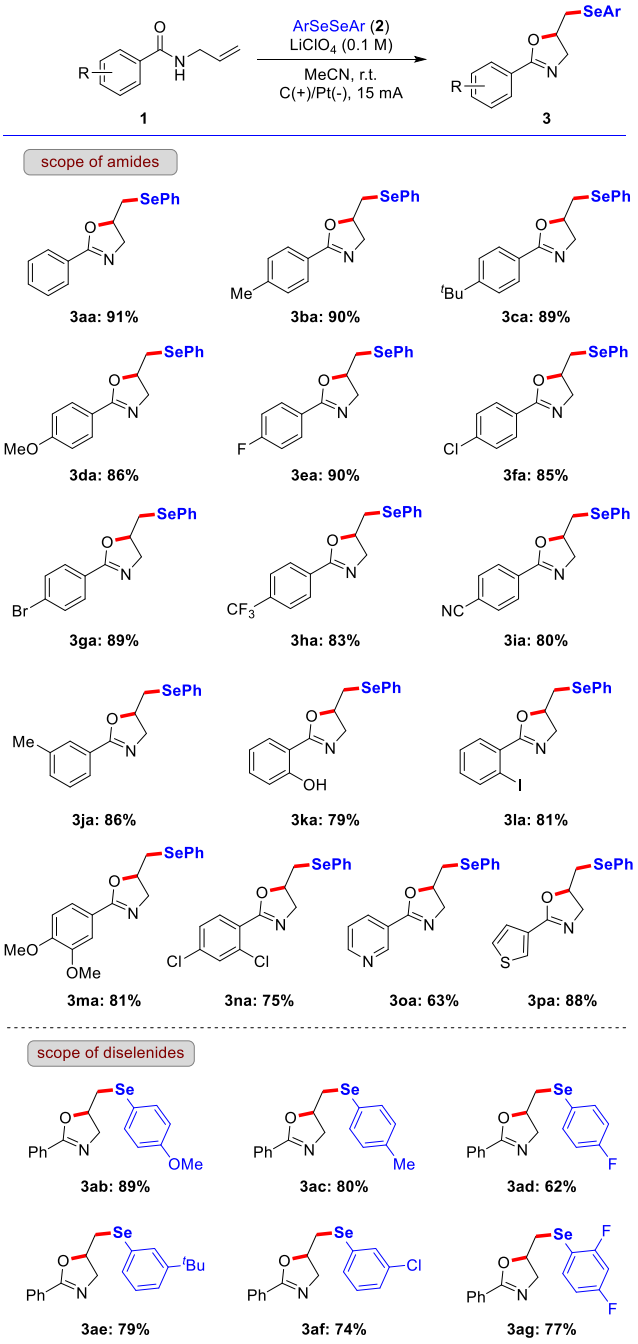
Entry	Deviation from Standard Conditions[a]	Yield [%][b]
1	None	91
2	Graphite both as anode and cathode	79
3	Pt both as anode and cathode	84
4	Ni foam as cathode	77
5	MeOH instead of MeCN	86
6	MeCN/H_2O Instead of MeCN	70
7	Bu_4NBF_4 instead of $LiClO_4$	87
8	10 mA for 3 hr	83
9	20 mA for 1.5 hr	82
10	0.25 mmol of **2a** was used	90
11	0.13 mmol of **2a** was used	72
12	Without current	NR

[a] Standard conditions: **1a** (0.25 mmol), **2a** (0.20 mmol), $LiClO_4$ (0.1 M), MeCN (5 mL), C anode, Pt cathode, undivided cell, constant current = 15 mA, at r.t. under air for 2 h. [b] Isolated yield.

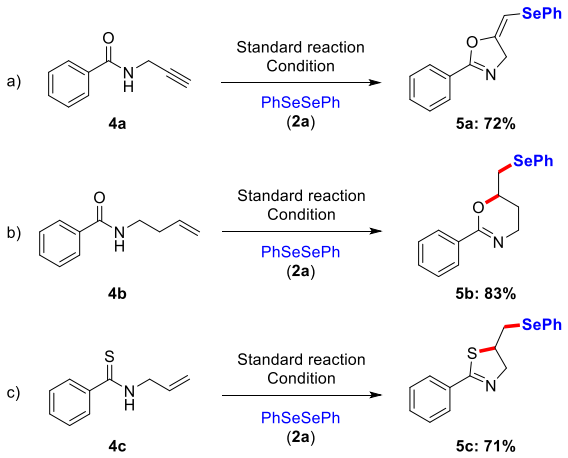
After establishing the optimized conditions, we next subjected a range of *N*-allyl amides **1** and diselenide **2** into electro-oxidative cyclization to acquire various oxazoline derivatives **3** (Table 2). We first examined the electronic and steric effects of a series of substrates and gratifyingly found that the protocol is widely applicable. Electron rich arenes containing –Me, –^{t}Bu, –OMe groups at different positions of the aryl ring reacted cleanly to furnish the corresponding selenylated oxazoline in excellent yields. Amide **1k** comprising free hydroxy group was also delivered desired oxazoline **3ka** in good yield without forming any noticeable byproduct. A series of halogen-containing arenes smoothly delivered very good isolated yield ranging from 81-90%. Electron withdrawing –CF_3, –CN groups at the *para*-position of arenes were also well tolerable. It is worth noting that heterocyclic group (pyridine, thiophene) bearing amides were tolerated well to delivered corresponding

selenylated products in satisfactory yields (**3oa** and **3pa**). The reaction scope was further reviewed with a series of *ortho*-, *meta*- and *para*-substituted diaryl diselenide derivatives. In all cases, excellent yields of the oxazolines were observed (**3ab-3ag**). The reaction was also found to be general with respect to the electronic influence, both electron-rich and electron poor diselenides displayed good to excellent efficiency.

Table 2. Substrate Scope for the Synthesis of Selenylated Oxazolines [a], [b]



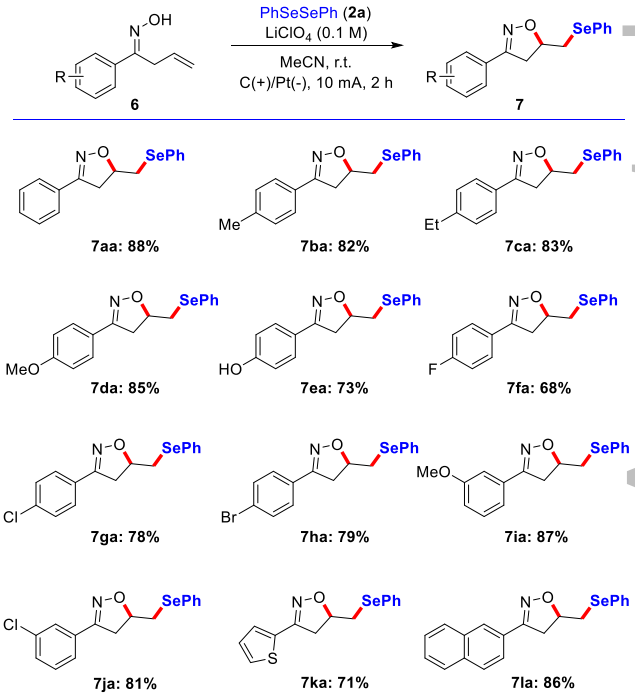
[a] Reaction conditions: **1** (0.25 mmol), **2** (0.20 mmol), $LiClO_4$ (0.1 M), MeCN (5 mL), C anode, Pt cathode, undivided cell, constant current = 15 mA, at r.t. under air for 2 h. [b] Isolated yield.



Scheme 2. Scope of Amides and Thiobenzamide.

Amides with varying chain length and unsaturation were also compatible in the oxidative cyclization process (Scheme 2). *N*-Propargyl amide **4a** delivered the oxazoline **5a** with an exocyclic double bond as a single diastereomer. Pleasingly, the six-membered heterocycle **5b** was formed efficiently with *N*-homoallyl amide **4b**. Finally, the thiazoline derivative **5c** was synthesized from corresponding *N*-allyl thiobenzamide **4c** following the standard reaction conditions.

Table 3. Substrate Scope for the Synthesis of Selenylated Isoxazoline [a], [b]



[a] Reaction conditions: **6** (0.25 mmol), **2a** (0.20 mmol), $LiClO_4$ (0.1 M), MeCN (5 mL), C anode, Pt cathode, undivided cell, constant current = 10 mA, at r.t. under air for 2 h. [b] Isolated yield.

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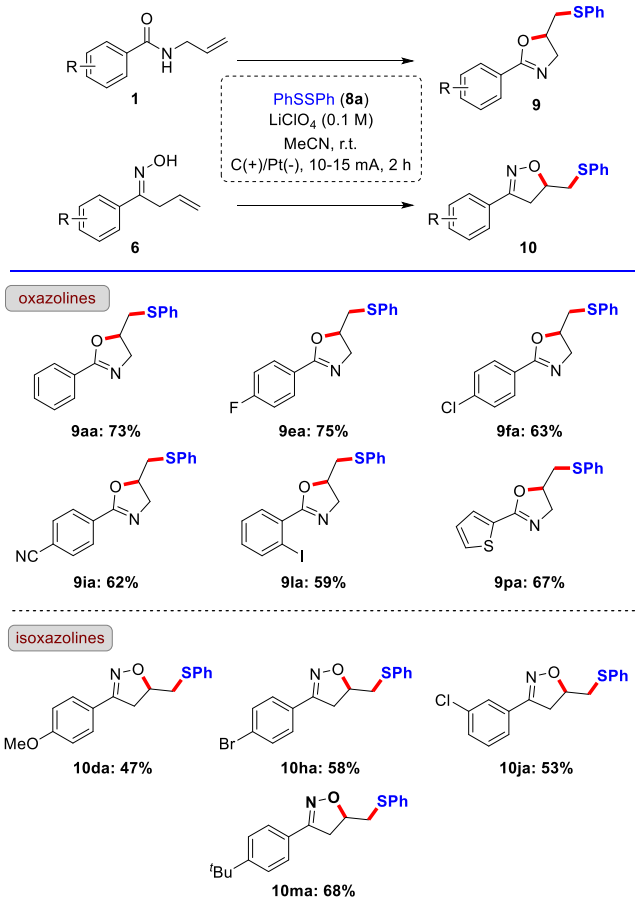
Next, we investigated the synthesis of selenylated isoxazoline moieties **7** by electrooxidation of oximes **6** and diphenyl diselenide (**2a**) with minor modification of the standard conditions (Table 3). *para*-Substituted electron-rich aryl oximes underwent smooth conversion to the isoxazoline in high yields (**7aa-da**). Oxime **6e** containing free hydroxyl group was found as a competent substrate under electrochemical conditions. A series of halo-substituted oximes were tested, in all cases, the smooth formation of selenylated isoxazolines were observed (**7fa-ha**). *meta*-Substituted aryl oximes were also yielded the corresponding isoxazolines efficiently (**7ia** and **7ja**). Finally, good to excellent yields were obtained with thiophenyl and naphthyl oximes (**6ka** and **6la**).

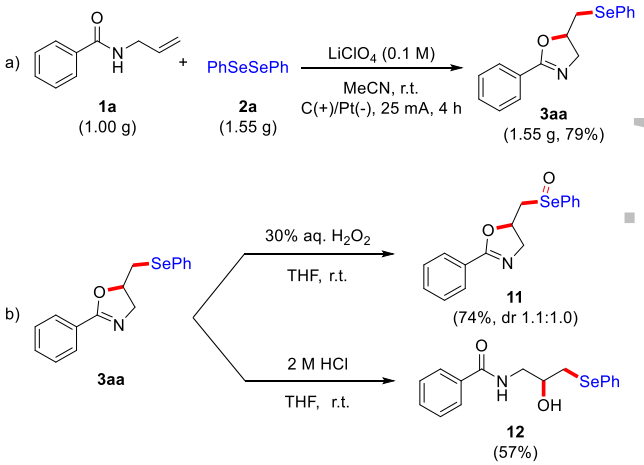
Table 4. Substrate Scope for the Synthesis of Thio Oxazolines and Isoxazolines [a], [b]



[a] Reaction conditions: **1** or **6** (0.25 mmol), **8a** (0.20 mmol), $LiClO_4$ (0.1 M), MeCN (5 mL), C anode, Pt cathode, undivided cell, constant current = 10-15 mA, at r.t. under air for 2 h. [b] Isolated yield.

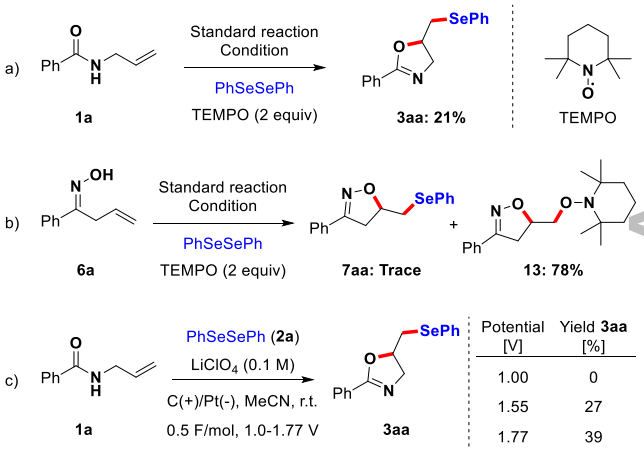
To extend the generality of the electro-synthetic protocol we explored the reaction of disulfide **8a** with various amides **1** and oximes **6** (Table 4). A series of electronically and sterically diverse amides underwent smooth cyclization to form the thiolated oxazoline **9** in good yields. In the case of oximes, comparatively lower efficiency was observed and thiolated isoxazoline **10** were formed in decent yields.

Synthetic practicability of the developed method was proved in a gram-scale synthesis of **3aa**, by maintaining the short reaction time and production efficiency (Scheme 3a). Derivatization of **3aa** was performed by oxidation to form selenoxide **11**. In another reaction, ring-opening hydrolysis of **3aa** delivered selenylated alcohol **12** (Scheme 3b).



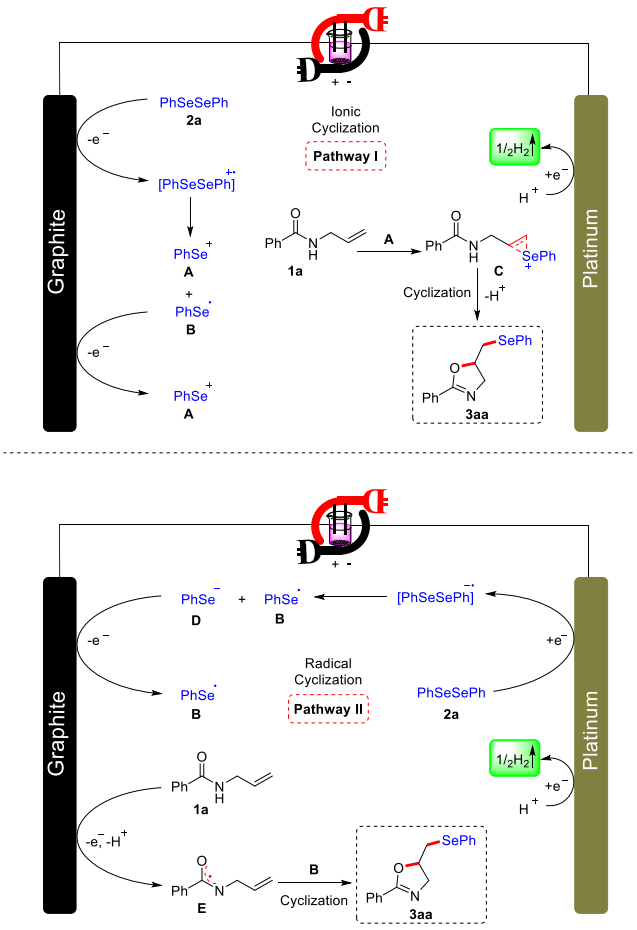
Scheme 3. Gram-scale synthesis and derivatizations.

To get the mechanistic insight, the reaction was performed in the presence of the radical quencher TEMPO. The amide **1a** underwent product formation with diminished yield (Scheme 4a), whereas shutdown in the selenylation reaction was observed for the oxime **6a** (Scheme 4b). Moreover, the adduct **13** which corresponds to the trapping of the radical intermediate by TEMPO was isolated in high yield. These observations indicate the possibility of both radical and ionic reaction pathways for the overall annulation and chalcogenation process.



Scheme 4. Mechanistic studies.

Based on the evidence gathered from the mechanistic studies, cyclic voltammetric studies (see SI for details), and literature reports, the probable mechanistic paths are depicted in Scheme 5. Oxidative activation of the diphenyl diselenide **2a** (oxidation peak at 1.66 V vs. Ag/AgCl) generates phenyl selenium cation **A** and phenyl selenium radical **B**. Further, one-electron oxidation of **B** forms another cation **A**. Addition of **A** to the double bond can generate intermediate **C** which upon nucleophilic cyclization by the amide oxygen produce oxazoline **3aa** (pathway I). Alternatively, reduction of **2a** is also feasible (reduction peak at -1.44 V vs. Ag/AgCl) at the reaction conditions to deliver radical **B** and phenyl selenium anion **D**. One electron oxidation of **D** forms another radical **B**. On the other hand, oxidation of the amide **1a** (oxidation peak at 1.32 V vs. Ag/AgCl) can produce delocalized radical **E**, follow up radical cyclization and coupling with **B** generate **3aa** (pathway II).



Scheme 5. Proposed mechanism.

The electricity demand of the above-mentioned processes is different. The ionic pathway I requires 1.0 F electricity for per mole of **3aa** production, whereas the radical pathway II needs 1.5 F per mole (see SI for details). To validate the possibility of these two pathways, **1a** and **2a** were allowed to react by applying 0.5 F/mol electricity at different potentials (Scheme 4c). At 1.0 V, no redox process is operating, thus product formation was not observed. Next, the potential was increased to 1.55 V, at this point only the reduction of **2a** and oxidation of **1a** are feasible, which implies that radical cyclization is the only viable pathway. Indeed, 27% of **3aa** was formed which is in support of the radical annulation. The potential was further increased to 1.77 V which allows both the reduction and oxidation of **2a**, thus opens up the possibility of both pathways I and II. Under this conditions, the observed **3aa** production was 39%, which is higher than the maximum possible yield (33% using 0.5 F/mol electricity) considering only pathway II. Therefore, the ionic pathway I must also be operational in the product formation. A similar mechanistic hypothesis is also applicable for the oxidative selenylation of allyl oximes to produce isoxazolines. However, the formation of TEMPO adduct **13** and comparatively low potential of **6a** (oxidation peak at 1.13 V vs. Ag/AgCl) indicate that the radical pathway may have a major contribution in isoxazoline formation.

Herein, we report the sustainable synthesis of chalcogenated oxazoline and isoxazoline derivatives under electrochemical conditions. Only sub-stoichiometric amount of chalcogen source was used and the reaction was performed at room temperature, open to air under neutral reaction conditions which results in the compatibility of a wide range of functional groups. Mechanistic studies indicate both radical and ionic reaction pathways are operative for the tandem C–O and C–chalcogen bond formations. This external oxidant and metal-free green protocol reveals synthetic benefits in terms of easily accessible amides and oximes as starting materials, short reaction time and also claims potential applications in large scale synthesis.

Experimental Section

General Procedure for Electrochemical Synthesis of Selenylated Oxazolines 3:

In an oven-dried undivided reaction flask (10 mL) equipped with a stir bar were added *N*-allylbenzamide derivatives (**1**, 0.25 mmol), diselenide (**2**, 0.2 mmol), $LiClO_4$ (0.1 M) and MeCN (5 mL). The flask was equipped with a platinum sheet (8 mm×30 mm×0.25 mm) as the cathode and a graphite plate (5.2 cm×0.8 cm×0.2 cm) as the anode. Thereafter the solution was electrolyzed at a constant current of 15 mA at room temperature open to the air for 2 h. After completion of the reaction, the solvent was evaporated in vacuum and purified by silica gel column chromatography to deliver oxazolines **3**.

General Procedure for Electrochemical Synthesis of Selenylated Isoxazolines 7:

In an oven-dried undivided reaction flask (10 mL) equipped with a stir bar were added allyl-oxime derivatives (**6**, 0.25

mmol), diphenyl diselenide (**2a**, 0.2 mmol, 62 mg), $LiClO_4$ (0.1 M) and MeCN (5 mL). The flask was equipped with a platinum sheet (8 mm×30 mm×0.25 mm) as the cathode and a graphite plate (5.2 cm×0.8 cm×0.2 cm) as the anode. Thereafter the reaction mixture was stirred and electrolyzed at a constant current of 10 mA under room temperature for 2 hrs. After the completion of the reaction the solvent was evaporated in vacuum and purified by silica gel column chromatography to deliver isoxazolines **7**.

Acknowledgements

Generous support by IISER Kolkata [Fellowship to SM and infrastructure], CSIR [Fellowship to MB and DM], UGC [Fellowship to KM] and SERB, DST, India [ECR/2016/000225 to SDS] is gratefully acknowledged.

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Electrochemical Chalcogenation of *β,γ*-Unsaturated Amides and Oximes to Corresponding Oxazolines and Isoxazolines

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