An Efficient, One-Pot, Green Synthesis of Tetracyclic Imidazo[2,1-*b*]Thiazoles *via* Electrochemically Induced Tandem Heteroannulation Reactions

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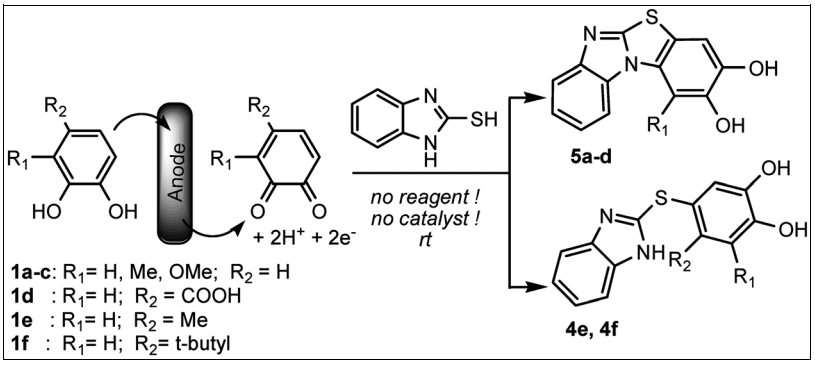

A series of novel catechol-fused tetracyclic compounds, with an imidazo[2,1-*b*]thiazole central core, were successfully synthesized through the anodic oxidation of catechols in the presence of 2-mercaptobenzimidazole in aqueous solution. The cyclic voltammetric results indicate that a one-pot four-step sequential reaction occurs between 2-mercaptobenzimidazole and the electrochemically derived *o*-benzoquinones affording fused polyheterocyclic compounds. The mechanism of this catalyst-free, domino reaction is proved as an ECEC pathway using controlled-potential coulometry. In addition, the electrosyntheses of fused compounds have been successfully performed in ambient conditions in an undivided cell using an environmentally friendly method with high atom economy. The structures of products were characterized by FT-IR, ^{1}H NMR, ^{13}C NMR, and HRMS spectrometric methods.

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INTRODUCTION

In recent years, the chemistry and synthesis of polycyclic compounds possessing an imidazo[2,1-*b*]thiazole central core (Fig. 1, **I**) has been the focus of great interest [1–5]. This is, in part, due to the broad spectrum of biological properties of these compounds. Several imidazo[2,1-*b*]thiazole derivatives have been reported in the literature as antibacterial [6], antifungal [7], antihelminthic [8,9], and antitumor [10–12] agents. For instance, the imidazo[2,1-*b*]thiazole system constitutes the main part of the well-known antihelminthic and immunomodulatory agent levamisole (**II**). A series of imidazo[2,1-*b*]thiazole guanyl hydrazones (**III**) have been proved to be active against various cancer cell lines [13]. Generally, the synthetic routes to imidazo[2,1-*b*]thiazoles are as following: (i) alkylations of cyclic thioureas by appropriate 1,2-dielectrophiles [14] or (ii) condensations of 2-aminothiazoles with 1,2-difunctionalized units, such as α,β-unsaturated carbonyl compounds [15]. Although several synthetic strategies are available, but these suffer from drawbacks including long reaction periods, toxic solvents, and/or metal additives, and therefore, the facile and green routes to these structures would be of great interest. Catechols are important species in chemistry and biology, because of their facility in undergoing electron transfer [16] and involved in the primary photoelectron donor–acceptor center in bacterial photosynthesis [17]. The most important property of catechols is the ease in which they undergo redox transformations; a very important physiological reaction. Catechol species are electrochemically oxidized to the corresponding quinines, and these can undergo a nucleophilic attack by the nucleophilic species through a 1,4-Micheal addition pathway [18,19].

Electric current is one of the cleanest tools in transformation of organic molecules [20], and accordingly, several electrochemical techniques have provided a variety of highly reactive intermediates affording useful and/or novel organic compounds without the use of any environmentally undesirable catalysts. In particular, direct electrochemical oxidation/reduction of substrates utilizes practically mass-free electrons as the only reagents. In this sense, electrochemistry is frequently referred to as one of the prototypical green procedures for synthesizing various organic molecules and structures [21].

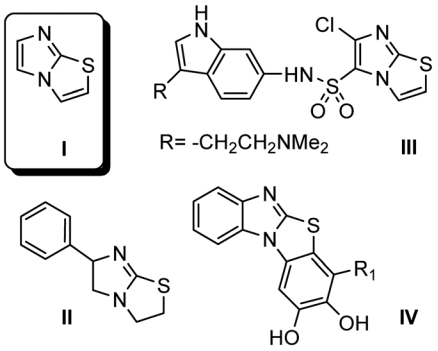
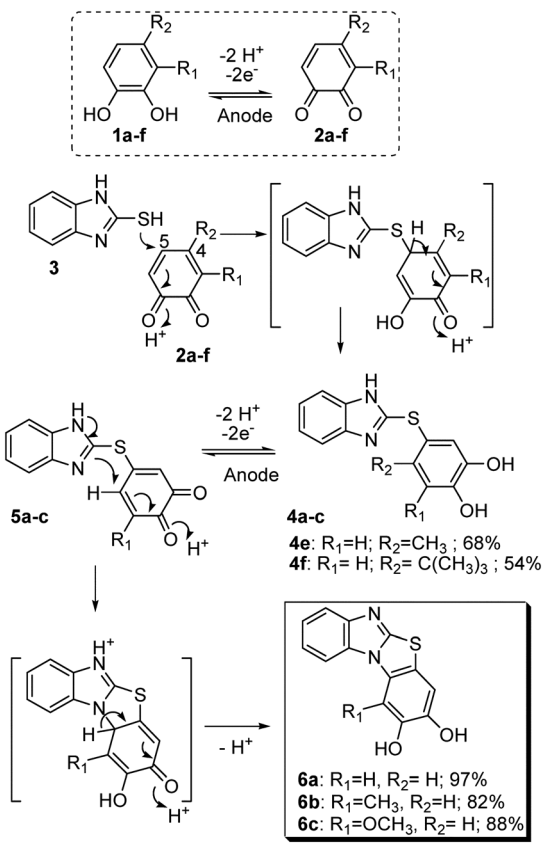


Figure 1. Some typical heterocycles with an imidazo[2,1-*b*]thiazole moiety.

In continuation of our efforts to develop more versatile and convenient electrochemical and chemical synthesis of highly functionalized heterocycles [22], here, we report the electrosynthesis of a series of tetracyclic catechol-fused benzimidazo[2,1-*b*]thiazoles (Fig. 1, **IV**) through a one-pot electrooxidative-coupling sequential fashion. The present protocol provides an efficient procedure *via* an ECEC electrochemical mechanism entry to construct nitrogen- and sulfur-containing heterocycles of type benzimidazo[2,1-*b*]thiazoles, a relatively rare fused-ring system. To the best of our knowledge, this is the first report aimed at the synthesizing of tetracyclic adducts bearing imidazo[2,1-*b*]thiazole and catechol moieties utilizing electrochemical techniques.

Scheme 1



RESULTS AND DISCUSSION

Initial characterization of the reaction was carried out using cyclic voltammetry. Cyclic voltammetric responses of a GC electrode to 1 m*M* catechol (**1a**) (Scheme 1) in water/acetonitrile (90/10) solutions containing 0.2*M* sodium acetate in the absence and presence of 2-mercaptobenzimidazole (**3**) are outlined in Figure 2(I).

Curve a in Figure 2(I) shows a well-defined oxidative peak (A_1) at 0.5 V (*vs.* SCE) consistent with the oxidation of **1a** to the *o*-benzoquinone species (**2a**). Upon reversal of the scan direction, a corresponding reduction peak (C_1) is observed at 0.05 V, which is attributed to back reduction of **2a** to the parent catechol (Scheme 1). A peak current ratio ($I_p^{C_1}/I_p^{A_1}$) of nearly unity, especially during the repetitive recycling of the potential can be observed for **1a** in the absence of **3**, and this ratio can be considered as a criterion for the stability of *o*-benzoquinone generated at the surface of GC electrode under the applied experimental conditions. In Figure 2(I), curve b shows the cyclic voltammogram obtained for a 1 m*M* solution of **1a** in the presence of 1 m*M* of **3**. The voltammogram exhibits one anodic and one decreased cathodic peaks appeared at 0.56 and –0.04 V *versus* SCE, respectively. The observed slightly shifts (positive for A_1 and negative for C_1 peaks) in the presence of **3** is probably due to the formation of a thin film of product at the surface of the electrode, inhibiting to a certain extent the performance of the electrode process [23].

In addition, similar to the oxidation of 2-mercaptobenzoxazole, which leads to the formation of bis(benzoxazolyl) disulfide [24], 2-mercaptobenzimidazole (**3**) can be oxidized to the corresponding disulfide [Fig. 2(I), curve c], and the anodic peak (A_2) appeared at 0.53 V can be assigned for the oxidation of **3**. In addition, the SH and NH groups of **3** are appropriate nucleophiles, so it seems that a Michael 1,4-addition of **3** to **2a** can proceed in a quick and simple way leading to the considerable decrease in the height of cathodic peak (C_1).

The cyclic voltammetries of **1a** in the presence of **3** in various sweep rates show that anodic peak current is increased with the addition of potential sweep rate [Fig. 2 (II)]. Also, a plot of peak current ratio ($I_p^{A_1}/I_p^{C_1}$) *versus* log v (not shown), for a mixture of **1a** and **3** confirmed the reactivity of *o*-benzoquinone **2a** toward mercaptide anion, appearing as an increase in the height of the cathodic peak C_1 at higher scan rates. A similar situation was observed when the concentration of **1a** was increased relative to **3** [Fig. 3(I)]. On the other hand, the current function for the A_1 peak, ($I_p^{A_1}/\upsilon_{1/2}$) decreased on increasing the scan rate. It is well known that such behaviors are indicative of an ECEC mechanism [25]. We also studied the effect of solution's pH on the electrochemical oxidation of **1a** in

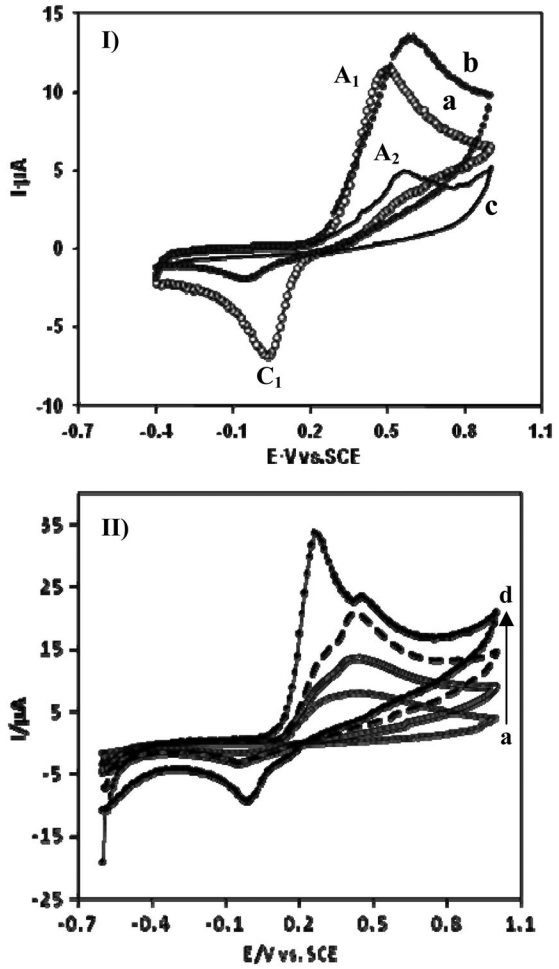


Figure 2. (I) Cyclic voltammograms of 1.0 m*M* **1a** in the absence (a) and presence (b) of 1.0 m*M* of **3** and (c) 1.0 m*M* of **3** in the absence of **1a** at glassy carbon electrode *versus* SCE in water/acetonitrile (90/10) solution containing 0.2*M* sodium acetate. Scan rate: 100 mV s^{-1}; t = 25 ± 1°C. (II) Scan rates for (a)–(d) are: 50, 100, 200, and 500 mV s^{-1}.

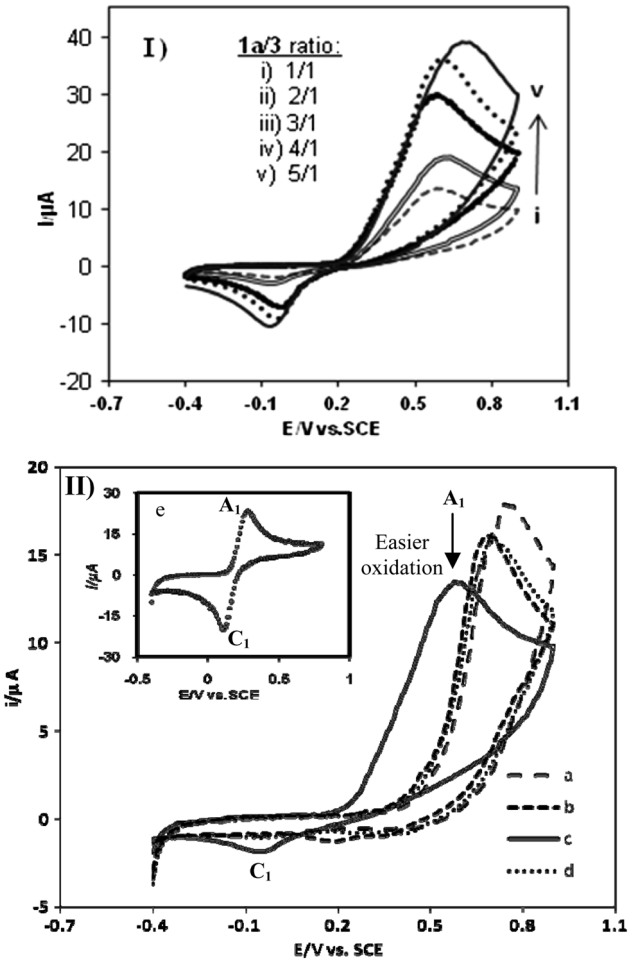


Figure 3. (I) Cyclic voltammograms obtained for the various ratio of the concentration of **1a** to **3**. (II) Cyclic voltammograms of **1a** in the presence of **3** obtained at various pHs: (a) 2, (b) 4, (c) 7, and (d) 9, at glassy carbon electrode *versus* SCE in water/acetonitrile (90/10) solution containing 0.2*M* sodium acetate. t = 25 ± 1°C. (e) CV of **1a** in the absence of **3** at pH = 7 is given for comparison.

the presence of **3**. Although, similar decreasing in the current density for the cathodic peak (indicating the reactivity of **2a** toward **3**) was observed in all pHs [Fig. 1(II)], however, it was found that the easier oxidation (less potential) of **1a** to **2a** can be occurred at pH = 7.

Considering the closeness of oxidation potential peaks of **1a** and **3** [A_1 and A_2 in Fig. 2(I), curves a and c], to minimize the oxidation of **3** and hence, achieving higher selectivity and efficiency, we applied 0.25 V potential *versus* SCE in both coulometry and preparative synthesis processes.

To determine electrochemical efficiency, controlled-potential coulometry of **1a** in the presence of **3** was performed at 0.25 V *versus* SCE. On the basis of obtained results, the electrochemical efficiency is >88%. The monitoring of electrolysis progress was carried out using cyclic voltammetry. It was shown that, proportional to the progress of coulometry, the anodic peak (A_1) decrease and disappears when the charge consumption becomes about $4e^-$ per molecule of **1a**. These results confirm the proposed ECEC mechanism for this electrochemically induced reaction and allow us to suggest the pathways shown in Scheme 1 for the electrooxidation of **1a** in the presence of **3**. According to our results, it seems that anodic oxidation process of **1a** to **2a** is followed by an intermolecular Michael-type reaction of **3** with **2a**, and this event seems to occur much faster than other side reactions, leading to the formation of intermediate **4a**.

The over-oxidation of **4a** is easier than the oxidation of the parent-starting molecule by virtue of the presence of an electron-donating group and hence, following the oxidation of **4a** to **5a**, an intramolecular Michael-type heteroannulation of **5a** occurs to give **6a**. Although the final product can be oxidized at a lower potential than **1a**, however, over-oxidation of **6a** was circumvented during the preparative reaction because of the insolubility of the products in

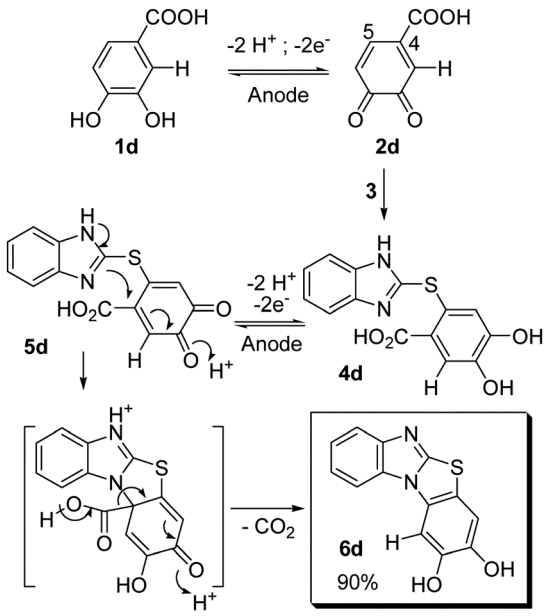
the water/acetonitrile medium. As it can be seen, there is an electrooxidative/intermolecular Michael-type addition/electrooxidative/intramolecular heteroannulation sequence (ECEC) in this one-pot four-step coupling reaction of **1a** with **3** leading to the formation of a novel catechol-fused tetracyclic compound (**6a**), with an imidazo[2,1-*b*]thiazole central core in excellent yield (92%).

In examining the scope and generality of the developed protocol as well as the influence of structural variation of catechol ring on the reactivity of *o*-benzoquinones toward **3**, we studied the electrochemically induced reaction of some other catchols bearing electron-withdrawing or electron-donating groups at the C-3 or C-4 positions (**1b–f**). The electrooxidation of 3-methylcatechol (**1b**) and 3-methoxycatechol (**1c**) in the presence of **3** proceed in a similar fashion to that of **1a**. Also notably, the existence of a methyl or methoxy group at the C-3 position of **1b** or **1c** may have subtle effects on the reactivity of their relevant *o*-bezoquinones and would probably cause these Michael acceptors (**2b** and **2c**, Scheme 1) to be attacked by **3** from the C-5 or C-4 positions. Because the methyl and methoxy groups are both electron-donating substituents, we think that *o*-benzoquinones **2b** and **2c** are more electropositive at C-5 position and therefore, can be selectively attacked in all probability only at the C-5 position by **3** leading to the formation of intermediates **4b** and **4c** (Scheme 1), respectively. As depicted in Scheme 1, similar to that of **4a**, further oxidation of these intermediates is followed by an intramolecular Michael-type heteroannulation which leads to the final products. These one-pot four-step oxidative coupling reactions (ECEC) of **1b** and **1c** with **3** lead to the formation of novel tetracyclic benzimidazo[2,1-*b*]thiazole compounds in high yields (Scheme 1: **6b**: 82%; **6c**: 88%).

On the other hand, anodic oxidation of 3,4-dihydroxybenzoic acid (**1d**) with an electron-withdrawing group, in the presence of **3** proceeds in a nearly different manner than that of **1a–c**. Oxidation of **1d** to **2d** (Scheme 2), and subsequent intermolecular Michael addition reaction of **3** with **2d** (at the more electropositive C-5 position) leads to adduct **4d** which is capable of undergoing further oxidation to afford intermediate **5d**. An intramolecular 1,4-addition (Michael) reaction in **5d** followed by an electrodecarboxylation process [26, 27] can lead to the final product. As it can be seen, this unique sequential heteroannulation reaction of **1d** with **3** leads to the formation of a novel catechol-fused tetracyclic benzimidazo[2,1-*b*]thiazole compound (**6d**) in excellent yield (90%).

The effect of an electron-donating group located at the reactive site of *o*-benzoquinone (C-4 or β-position to carbonyl group) on its reactivity was also investigated in some detail. The electrochemical oxidations of 4-methylcatechol (**1e**) and 4-*tert*-butylcatechol (**1f**; Scheme 1) were studied in the presence of **3** in water/acetonitrile (90/10) solution containing 0.20*M* sodium acetate.

Scheme 2



As mentioned earlier, any decrease observed in the current density for the cathodic peak in cyclic voltammogram of catechol relates to the reactivity of electrochemically derived *o*-benzoquinone toward nucleophile. Compared with that of **1a**, cyclic voltammograms of **1e** and **1f** in the presence of **3** show less decrease in the current density for the cathodic parts. These results prove that **2e** and **2f** are weaker Michael-acceptor than **2a** and therefore, have less reactivity in the Michael-addition reaction toward mercaptide anion of **3**, appearing as an increase in peak current ratio ($I_p^{C_1}/I_p^{A_1}$). More interestingly, contrast to the cases of **4a–d**, no further oxidations and subsequent heteroannulation reaction were observed in the cases of **4e** and **4f,** and these adducts (catechol-benzimidazole thioetheres) were obtained as the final products in moderate yields (**4e**: 68%; **4f**:54%). In addition to insolubility of **4e** and **4f** in water/acetonitrile media, one can assume that the presence of a methyl or *tert*-butyl group at the C-4 position of these adducts would probably cause a streic inhibition of the accessibility of their C-4 position to nucleophilic nitrogen atom of adjacent benzimidazole moiety and therefore, there is no possibility for the subsequent heteroannulation reaction.

CONCLUSIONS

In conclusion, we have described a one-pot, environmentally friendly and reagent-less protocol for the preparation of a series of polyfunctional tetracyclic benzimidazo [2,1-*b*]thiazoles through a domino reaction of commercially available starting materials. Besides the high efficiency and atom economy as a domino process, this reaction has the following advantages: (1) reaction proceeds smoothly under

very mild conditions without introducing any acid, base or metal catalyst and (2) it is an environmentally benign transformation because of the fact that only electrons are used as reagent instead of oxidative ones. We think that this procedure with its advantages of complementary reactivity, especially dramatically technical feasibility and because of the diversity of this method, can be adopted in organic heterocyclic chemistry to synthesize and screen libraries of related biologically important imidazo[2,1-*b*]thiazoles.

EXPERIMENTAL

Apparatus and reagents. The working electrode used in voltammetry experiments was a glassy carbon disk (1.8-mm diameter), and platinum wire was used as the counter electrode. The working electrode used in controlled-potential coulometry and bulk electrolysis (using an electronic potentiostat) was an assembly of four rods, 6-mm diameter, and ~ 10-cm length, and a large platinum gauze constituted the counter electrode. The working electrode potentials were measured *versus* SCE. ^{1}H and ^{13}C NMR spectra were recorded on a spectrometer operating at 200 MHz for proton and carbon nuclei. Chemical shifts were recorded as δ values in parts per million (ppm). Spectra were acquired in DMSO-d_6 at 25°C. For ^{1}H NMR spectra, the peak due to residual DMSO (δ 2.54) was used as the internal reference. ^{1}H NMR spectra are reported as follows: chemical shift (δ) [multiplicity (where multiplicity is defined as: br = broad; s = singlet; d = doublet; t = triplet; q = quartet; m = multiplet), coupling constant(s) *J* (Hz), relative integral, and assignment]. ^{13}C NMR spectra were conducted using the central peak (δ 39.5) of the DMSO multiplet as the internal reference. Mass spectra and exact masses were recorded on a high resolution mass spectrometer; the latter used a mass of 12.0000 for carbon, and the data are listed as follows: mass-to-charge ratio (*m*/*z*). Infrared spectra were recorded using a drop-casting technique on NaCl plates and are reported in wavenumbers (cm^{-1}). All chemicals (catechols **1a–f** and 2-mercaptobenzimidazole) were reagent-grade materials, and sodium acetate and other solvents and reagents were of proanalysis grade. These chemicals were used without further purification.

General procedure for the synthesis of tetracyclic imidazo [2,1-*b*]thiazols. In a typical procedure, an aqueous solution (*ca.* 100 mL) of water/acetonitrile (90/10), containing 0.2*M* sodium acetate, 1.0 mmol of catechols (**1a–f**), and 1.0 mmol of 2-mercaptobenzimidazole (**3**), was electrolyzed in an undivided cell equipped with a carbon anode (an assembly of four rods, 6-mm diameter, and ~ 10-cm length) and a large platinum gauze cathode at 0.20 V *versus* SCE, at 25°C. The electrolysis was terminated, when the decay of the current became more than 95%. The process was interrupted during the electrolysis, and the graphite anode was washed in acetone to reactivate it. At the end of electrolysis, a few drops of acetic acid were added to the solution, and the cell was placed in a refrigerator overnight. The precipitated solid was collected by filtration, washed copiously with distilled water, and characterized by: FT-IR, ^{1}H NMR, ^{13}C NMR, and HRMS. The final products (**6a–d** and **4e–f)** were obtained purely, and no extra purification was needed. Selected characterization data for the products are given below.

Compound 6a. Yield 97%, amorphous, pale-yellow solid, mp: 251–253°C; IR (KBr): ν (cm^{-1}) = 3376, 3056, 2993, 1622, 1529, 1446, 1257, 1196, 739; ^{1}H NMR (200 MHz, DMSO-d_6): δ = 4.27 (broad, OH), 6.94 (s, 1H, Har), 7.11 (s, 1H, Har) 7.17–7.55 (m, 4H, Har); ^{13}C NMR (50 MHz DMSO-d_6): δ = 109.8, 113.8, 121.5, 122.6, 123.6, 128.1, 128.7, 143.0, 144.2, 149.4, 156.1, 157.1, 157.6; ms (EI) *m*/*z* (relative intensity): 256 [M]$^+$ (30), 224 (33), 208 (25), 150 (100), 134 (37), 110 (75), 39 (80); HRMS (EI): *m*/*z* calcd for $C_{13}H_8N_2O_2S$: 256.0306; found: 256.0324.

Compound 6b. Yield: 82%, amorphous, beige solid, mp 240–244°C; IR (KBr): ν (cm^{-1}) = 3424, 3056, 2991, 1625, 1529, 1488, 1449, 1382, 1267, 1209, 739; ^{1}H NMR (200 MHz, DMSO-d_6): δ = 2.31 (s, 3H, CH3), 4.46 (broad, OH), 7.19 (s, 1H, Har), 7.45–7.75 (m, 4H, Har); ^{13}C NMR (50 MHz DMSO-d_6): δ = 9.01, 98.6, 110.8, 114.4, 118.2, 119.0, 121.9, 122.0, 123.3, 127.0, 131.8, 141.5, 145.6, 148.0; ms (EI) *m*/*z* (relative intensity): 270 [M]$^+$ (32), 269 (10), 155 (22), 150 (30), 110 (100), 77 (40), 43 (80); HRMS (EI): *m*/*z* calcd for $C_{14}H_{10}N_2O_2S$: 270.0463; found: 270.0471.

Compound 6c. Yield: 88%, amorphous brown solid, mp 210–213°C; IR (KBr): ν (cm^{-1}) = 3408, 3042, 2928, 1628, 1532, 1449, 1270, 1216, 1100, 742; ^{1}H NMR (200 MHz, DMSO-d_6): δ = 3.66 (s, 3H, OCH3), 4.04 (broad, OH), 6.94–7.55 (m, 5H, Har); ^{13}C NMR (50 MHz DMSO-d_6): δ = 56.4, 109.8, 114.7, 120.7, 122.2, 122.5, 136.1, 139.7, 140.1, 140.8, 146.8, 149.1, 150.1, 151.1; ms (EI) *m*/*z* (relative intensity): 286 [M]$^+$ (15), 178 (25), 164 (100), 150 (80), 140 (25), 131 (70), 39 (30); HRMS (EI) calcd. for $C_{14}H_{10}N_2O_3S$: 286.0412; found: 286.0401.

Compound 6d. Yield: 90%, amorphous, pale-yellow solid, mp: 251–253°C; IR (KBr): ν (cm^{-1}) = 3376, 3056, 2993, 1622, 1529, 1446, 1257, 1196, 739; ^{1}H NMR (200 MHz, DMSO-d_6): δ = 4.27 (broad, OH), 6.94 (s, 1H, Har), 7.11 (s, 1H, Har) 7.17–7.55 (m, 4H, Har); ^{13}C NMR (50 MHz DMSO-d_6): δ = 109.8, 113.8, 121.5, 122.6, 123.6, 128.1, 128.7, 143.0, 144.2, 149.4, 156.1, 157.1, 157.6; ms (EI) *m*/*z* (relative intensity): 256 [M]$^+$ (30), 224 (33), 208 (25), 150 (100), 134 (37), 110 (75), 39 (80); HRMS (EI): *m*/*z* calcd for $C_{13}H_8N_2O_2S$: 256.0306; found: 256.0322.

Compound 4e. Yield: 68%, amorphous brown solid, mp 161–165°C; ^{1}H NMR (200 MHz, DMSO-d_6): δ = 2.22 (s, 3H, CH3), 3.63 (broad, OH), 6.99–7.45 (m, 6H, Har), 12.58 (broad, NH); ^{13}C NMR (50 MHz DMSO-d_6): δ = 20.0, 114.4, 115.9, 118.4, 121.1, 122.8, 124.7, 133.0, 139.6, 144.3, 147.7; ms (EI) *m*/*z* (relative intensity): 272 [M]$^+$ (10), 255 (15), 150 (100), 134 (70), 119 (45), 93 (70); HRMS (EI) calcd. for $C_{14}H_{12}N_2O_2S$: 272.0619; found: 272.0607.

Compound 4f. Yield: 54%, amorphous beige solid, mp 201–204°C; ^{1}H NMR (200 MHz, DMSO-d_6): δ = 1.25 (s, 9H, 3-CH3), 3.91 (broad, OH), 7.11–7.38 (m, 4H, Har), 7.60 (s, 1H, Har), 7.85 (s, 1H, Har), 12.48 (broad, NH); ^{13}C NMR (50 MHz DMSO-d_6): δ = 29.8, 34.6, 109.8, 114.8, 121.3, 122.6, 126.1, 132.5, 139.3, 141.1, 144.1, 144.3; ms (EI) *m*/*z* (relative intensity): 314 [M]$^+$ (10), 152 (20), 150 (100), 122 (35). HRMS (EI) calcd. for $C_{17}H_{18}N_2O_2S$: 314.1089; found: 314.1028.

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REFERENCES AND NOTES

[1] Gürsoy, E.; Güzeldemirchi, N. U. Eur J Med Chem 2007, 42, 320.

[2] Andreani, A.; Granaiola, M.; Leoni, A.; Locatelli, A.; Morigi, R.; Rambaldi, M.; Giorgi, G.; Garaliene, V. Anticancer Res 2004, 24, 203.

[3] Andreani, A.; Granaiola, M.; Leoni, A.; Locatelli, A.; Morigi, R.Rambaldi, M.; Lenaz, G.; Fato, R.; Bergamini, C.; Farruggia, G. J Med Chem 2005, 48, 3085.

[4] Andreani, A.; Granaiola, M.; Leoni, A.; Locatelli, A.; Morigi, R.; Rambaldi, M. Eur J Med Chem 2001, 36, 743.

[5] Andreani, A.; Rambaldi, M.; Leoni, A.; Locatelli, A.; Andreani, F.; Gehret, J. C. Pharm Acta Helv 1996, 71, 247.

[6] Kiran, J. M. Ind J Chem 1991, 30B, 898.

[7] Gupta, G. D.; Jain, K. K.; Gupta, R. P.; Pujari, H. K. Ind J Chem 1983, 22B, 268.

[8] Robert, J. F.; Xicluna, A.; Panouse, J. J. Eur J Med Chem Chim Ther 1975, 10, 59.

[9] Amarouch, H.; Loiseau, P. R.; Bacha, C.; Caujolle, R.; Payard, M.; Loiseau, P. M.; Bories, C.; Gayral, P. Eur J Med Chem 1987, 22, 463.

[10] Andreani, A.; Rambaldi, M.; Andreani, F.; Bossa, R.; Galatulas, I. Eur J Med Chem 1988, 23, 385.

[11] Andreani, A.; Rambaldi, M.; Locatelli, A.; Bossa, R.; Fraccari, A.; Galatulas, I. Pharm Acta Helv 1993, 68, 21.

[12] Andreani, A.; Rambaldi, M.; Locatelli, A.; Bossa, R.; Fraccari, A.; Galatulas, I. J Pharm Belg 1993, 48, 378.

[13] Andreani, A.; Leoni, A.; Locatelli, A.; Morigi, R.; Rambaldi, M.; Recanatini, M.; Garaliene, V. Bioorg Med Chem 2000, 8, 2359.

[14] Jarosinski, M. A.; Reddy, P. S.; Anderson, W. K. J Med Chem 1993, 36, 3618.

[15] Gewald, K.; Heinhold, G. Monatsh Chem 1976, 107, 1413.

[16] Pierpont, C. G.; Buchanan, R. M. Coord Chem Rev 1981, 38, 45.

[17] Tompson, R. H. Naturally Occurring Quinones. III. Recent Advances; Chapman & Hall: London, 1987.

[18] Seymour, E. H.; Lawrence, N. S.; Beckett, E. L.; Davis, J.; Compton, R. G. Talanta 2002, 57, 233.

[19] Shahrokhian, S.; Amiri, M. Electrochem Commun 2005, 7, 68.

[20] Bard, A. J.; Faulkner, L. K. Electrochemical Methods: Fundamentals and Applications; John Wiley & Sons: New York, 2001; Chapter 1.

[21] Steckhan, E.; Arns, T.; Heineman, W. R.; Hilt, G.; Hoormann, D.; Jorissen, J.; Kroner, L.; Lewall, B.; Putter, H. Chemosphere 2001, 43, 63.

[22] (a) Khodaei, M. M.; Alizadeh, A.; Pakravan, N. J Org Chem 2008, 73, 2527; (b) Alizadeh, A.; Nematollahi, D.; Habibi, D.; Hesari, M. Synthesis 2007, 10, 1513; (c) Shamsipur, M.; Kazemi, S. H.; Alizadeh, A.; Mousavi, M. F.; Workentin, M. S. J Electroanal Chem 2007, 610, 218; (d) Nematollahi, D.; Habibi, D.; Alizadeh, A. Phosphorus Sulfur Silicon Relat Elem 2006, 181, 1391; (e) Habibi, D.; Nematollahi, D.; Alizadeh, A.; Hesari, M. Heterocycl Commun 2005, 11, 145; (f) Nematollahi, D.; Habibi, D.; Alizadeh, A.; Hesari, M. J Heterocycl Chem 2005, 42, 289.

[23] Nematollahi, D.; Rafiee, M. J Electroanal Chem 2004, 566, 31.

[24] Berlich, A.; Flemmig, B.; Wittstock, G. Solid State Electrochem 2001, 6, 29.

[25] Little, R. D.; Weinberg, N. L. Electroorganic Synthesis: Festschrift for Manuel M. Baizer; CRC Press: USA, 1991.

[26] Goossen, L. J.; Collet, F.; Goossen, K. Isr J Chem 2010, 50, 617.

[27] Fotouhi, L.; Mosavi, M.; Heravi, M. M.; Nematollahi, D. Tetrahedron Lett 2006, 47, 8553.