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Electrochemical oxidation of 2*H*-imidazole *N*-oxides

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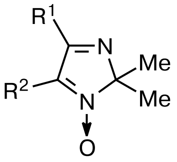
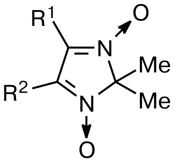
Electrochemical oxidation of 2*H*-imidazole *N*-oxides was studied using cyclic voltammetry and ESR spectroscopy. The formation of the 2,2-dimethyl-4,5-diphenyl-2*H*-imidazole 1,3-dioxide radical cation was noticed for the first time. The possibility of reaction between the 2*H*-imidazole *N*,*N*-dioxide radical cation and methanol including the detachment of a hydrogen atom from the MeOH methyl group was demonstrated.

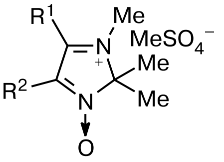
Key words: 2*H*-imidazole *N*-oxides, electrooxidation, radical cations.

We have reported previously the radical cation mechanism of the oxidative methoxylation of substituted 4*H*-imidazole *N*,*N*-dioxides giving α-methoxy-substituted iminoxyl radicals[1] and preparative electrosynthesis of these iminoxyl radicals.[2] Unlike 4*H*-imidazole *N*-oxides, 2*H*-imidazole *N*-oxides (**1**, **2**), being the structural isomers of the former, are converted into iminoxyl radicals with methoxy groups in the α-position to the radical center upon the reaction with PbO_2 in MeOH only in the case of aldonitrones, *i.e.*, *N*-oxides containing no substituent at the α-carbon atom of the nitronic group.[3,4] The formation of methoxy-substituted iminoxyl radicals from α-substituted 2*H*-imidazole *N*-oxides **1**, **2** was ascertained only on the basis of ESR data.[3,4]

We studied electrooxidation (EO) of 2*H*-imidazole *N*-oxides **1**—**3** in MeCN using cyclic voltammetry (CV). The CV curves for these compounds contain one diffusion oxidation peak ($I_p v^{-1/2}$ = const, where I_p is the peak maximum current, v is the potential sweep rate), which is irreversible up to $v = 0.33$ V s^{-1}. An exception is diphenyl-substituted *N*,*N*-dioxide **2c**, whose CV curve exhibits a reversible oxidation peak with $I_c/I_a = 1$ and the difference between the anodic and cathodic peak potentials $\Delta E_p = 0.08$ V. The I_p values for the oxidation of *N*-oxides **1**—**3** and for reversible single-electron oxidation of 2,2,5,5-tetramethyl-4-phenyl-3-imidazolin-1-oxyl 3-oxide giving the corresponding oxoammonium cation[5] are similar, which attests to a single-electron oxidation pattern of the initial 2*H*-imidazole *N*-oxides with generation of short-lived radical cations (RC).


1a—k


2a—d


3a,**b**

Note. The substituents R^1 and R^2 are defined in Table 1.

The anodic half-peak potentials ($E^a_{p/2}$) are listed in Table 1. The oxidation potentials of 2*H*-imidazole *N*,*N*-di-

Table 1. Electrooxidation[a] potentials (of the anodic half-peaks ($E^a_{p/2}$)) for 2*H*-imidazole *N*-oxides

Compound	R^1	R^2	$E^a_{p/2}$/V
1a	Me	H	1.79
1b	Me	Ph	1.64
1c	Me	*o*-$NO_2C_6H_4$	1.91
1d	Me	*p*-$NO_2C_6H_4$	1.91
1e	Ph	H	1.87
1f	Ph	OMe	1.36
1g	*p*-$NO_2C_6H_4$	H	1.96
1h	$CBrMe_2$	H	1.94
1i	CNO_2Me_2	H	2.02
1j	CH=NOH	H	1.92
1k	CN	H	2.25
2a	Ph	H	1.34 [b]
2b	Ph	Me	1.22 [c]
2c	Ph	Ph	1.29 [d]
2d	Me	Me	1.11 [e]
3a	Me	H	2.22
3b	Me	Ph	2.25

Note. For *N*,*N*-dioxides **2a,b,d**, the EO potentials for the second *N*-oxide group ($E^{2a}_{p/2}$) are also given.

[a] For a Pt electrode, *vs.* s.c.e. with 0.1 *M* Et_4NClO_4 in MeCN as the supporting electrolyte, $C = 10^{-3}$ mol L^{-1}, potential sweep rate 0.08 V s^{-1}.

[b] $E^{2a}_{p/2} = 1.75$ V.

[c] $E^{2a}_{p/2} = 1.48$ V.

[d] Reversible oxidation peak.

[e] $E^{2a}_{p/2} = 1.68$ V.

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oxides are substantially lower than those for mono-*N*-oxides. Thus, the difference between $E^{a}_{p/2}$ in the series of compounds **1b**,**e** and **2a**,**b** with the same substituents is 0.42—0.53 V. A similar effect of the *N*-oxide second O atom was observed during the EO of pyrazine, quinoxaline, and phenazine *N*-oxides and *N*,*N*-dioxides,[6] where $\Delta E^{a}_{p/2}$ was 0.40—0.60 V.

The $E^{a}_{p/2}$ values of compounds **1**—**3** depend on the nature of substituents in positions 4 and 5. Compound **2d** with two Me groups in these positions of the ring is oxidized most easily in this series of compounds ($E^{a}_{p/2}$ = 1.11 V). The oxidation potentials of 4-phenyl-substituted *N*,*N*-dioxides **2a**—**c** increase in the series of substituents R^2 = Me, Ph, H by 0.07 and 0.12 V respectively.

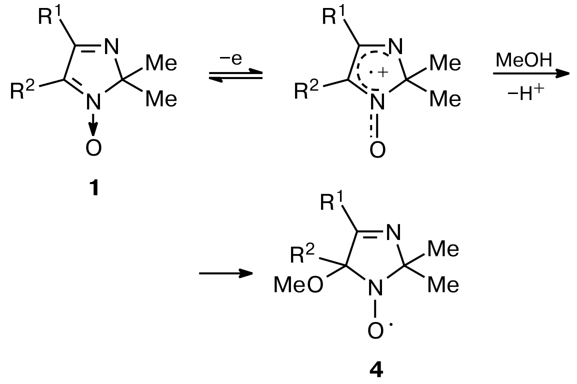
The oxidation potentials of aldonitrones **1a**,**e**,**g** increase from 1.79 V for compound **1a** (R^1 = Me) to 1.87 V for the case where a Ph group occurs in position 4 of the imidazole ring (**1e**). The presence of the *para*-nitro group in the phenyl fragment increases the oxidation potential of compound **1g** by 0.09 V ($E^{a}_{p/2}$ = 1.96 V). High $E^{a}_{p/2}$ values are also found for compounds **1h**,**i**, containing a Br atom and a nitro group in position 2 of the isopropyl fragment, respectively, and for aldonitrone **1j** with a hydroxyimine group, equal to 1.92 (**1j**, R^1 = CH=NOH), 1.94 (**1h**, R^1 = $CBrMe_2$), and 2.02 V (**1i**, R^1 = CNO_2Me_2). The introduction of the C≡N group in position 4 entails a shift of the oxidation potential to 2.25 V (**1k**, R^1 = CN), which is close to the $E^{a}_{p/2}$ values of 3-methyl-2*H*-imidazolium 1-oxides **3** and benzene (2.30 V).

The variation of R^2 also results in substantial changes in $E^{a}_{p/2}$ of compounds **1**. Thus the Ph group facilitates oxidation of *N*-oxide **1b** by 0.15 V relative to *N*-oxide **1a** unsubstituted at this position, and *ortho*- and *para*-nitro groups present in the phenyl fragment increase the oxidation potentials of **1c**,**d** by 0.27 V compared to this value for **1b**. The most pronounced effect is provided by the MeO group introduced into the α-position of the nitrone fragment, in particular, $E^{a}_{p/2}$ of compound **1f** equals 1.36 V, which is 0.51 V lower than the potential of aldonitrone **1e**. The difference between the effects of substituents in the α- and β-positions relative to the nitrone group is apparently associated with preferred localization of the electron density of the highest occupied molecular orbital of *N*-oxide **1** on the nitrone fragment.

In addition to the substituted imidazole ring containing the positively charged N(3) atom, 3-methyl-2*H*-imidazolium 1-oxides **3** contain the $MeSO_4^-$ counter-ion. In their single-electron EO, the corresponding radical dications should be the primary products, instead of the RC, which are formed in oxidation of compounds **1** and **2**. Electrooxidation of positively charged systems was observed[7] previously in the series of *C*-(*N*-methylpyridinium)-*N*-*tert*-butylnitrones, which also contain $MeSO_4^-$ as a counter-ion.

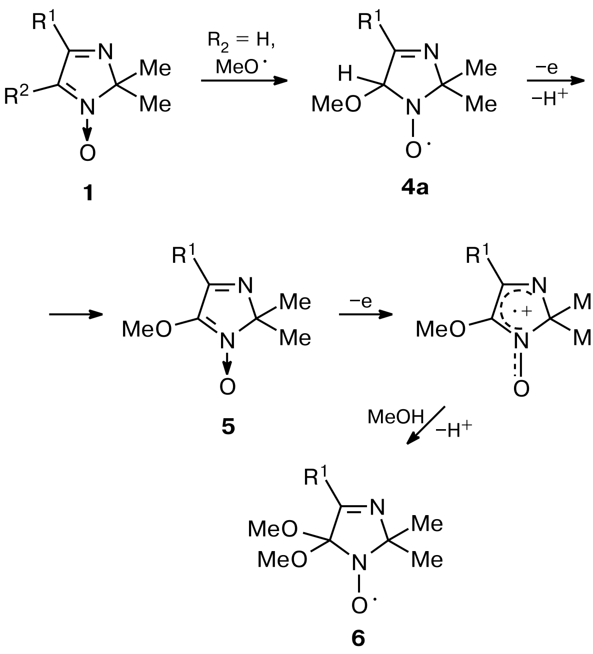
As can be seen from the data shown in Table 1, most of compounds of **1**, except for methoxynitrone **1f**, undergo EO at high potentials approaching the region of methanol oxidation (2.0—2.3 V).[8] Hence, it appears unlikely that the initial 2*H*-imidazole *N*-oxides are oxidized by lead dioxide (E^0 = 1.7 V) to the corresponding RC with subsequent addition of MeOH to give radicals **4** (Scheme 1).

Scheme 1



This consideration, along with the data[3] on the reaction of only aldonitrones **1** with PbO_2 in MeOH, allow one to assume (Scheme 2) that the formation of dimethoxy substituted iminoxyl radicals **6** from aldonitrones **1** in the PbO_2—MeOH system occurs by a spin capture mechanism *via* the intermediate formation of spin adduct **4a**,

Scheme 2



which is oxidized to methoxy nitrone **5**, which, in turn, can be oxidized to an RC; this is followed by the addition of MeOH to give finally dimethoxy product **6**.

The oxidation potentials of 2*H*-imidazole *N*,*N*-dioxides **2** are considerably lower than those of MeOH. However, the reactivity of 4,5-disubstituted compounds **2** during their oxidation by PbO_2 in MeOH is as low[4] as that of *N*-oxides **1**. To elucidate the reasons for this fact, we studied the reaction of 4,5-diphenyl-substituted *N*,*N*-dioxide **2c** with MeOH during EO using CV and ESR spectroscopy.

The ESR spectrum of the RC derived from **2c** recorded during the EO of *N*,*N*-dioxide **2c** carried out in MeOH-free MeCN in a cell placed in the ESR spectrometer resonator exhibited the following HFC constants for the N atoms: $a_N(2N) = 1.65 \cdot 10^{-4}$ T (Fig. 1). These constants are in line with the HFC constants found by INDO UHF calculations of the RC derived from the model compound, 4,5-diphenyl-2*H*-imidazole 1,3-dioxide ($a_N(2N) = -2.09 \cdot 10^{-4}$ T). The calculated HFC constants for N and C atoms of the 2*H*-imidazole ring and the O atoms of the *N*-oxide groups point to predominant localization of the RC spin density on the O atoms: $a_N(2N) = -2.09 \cdot 10^{-4}$ T, $a_C(2C) = -1.13 \cdot 10^{-4}$ T, $a_O(2O) = -10.19 \cdot 10^{-4}$ T. Previously, equally low values of the HFC constants for N atoms ($5 \cdot 10^{-5}$—$1.6 \cdot 10^{-4}$ T) were found[9] for the RC derived from pyrazine, quinoxaline and phenazine *N*,*N*-dioxides, while the highest calculated values of spin density for these RC were also noted for the *N*-oxide oxygen atoms.

In the absence of MeOH, an anodic peak *A* ($E^a{}_p$ = 1.38 V) can be seen in the CV curve of *N*,*N*-dioxide **2c** (Fig. 2, *a*) during the potential sweep in the anodic direction, and the corresponding cathodic peak *C* ($E^c{}_p$ = 1.30 V) is observed during the backward sweep. The height ratio I_c/I_a equals 1, which points to stability of the **2c** RC formed. With addition of MeOH into the cell containing the solution of **2c**, the height of peak *A* somewhat increases. Simultaneously, the current strength in the region of 1.8—2.0 V increases (Fig. 2, *b*), due to oxidation of MeOH. In turn, the height of peak *C* considerably decreases; however, in this case, the anodic peak of iminoxyl radical **7** does not appear on the CV curve. During EO of *N*,*N*-dioxide **2c** carried out under these conditions in a cell placed into the ESR spectrometer resonator, the spectrum of iminoxyl radical **7** is recorded ($a_N(1N) = 12.9 \cdot 10^{-4}$ T). This species arises,[4] most likely, according to Scheme 3 (pathway *A*). However, judging by low intensity of the signal, the concentration of radical **7**

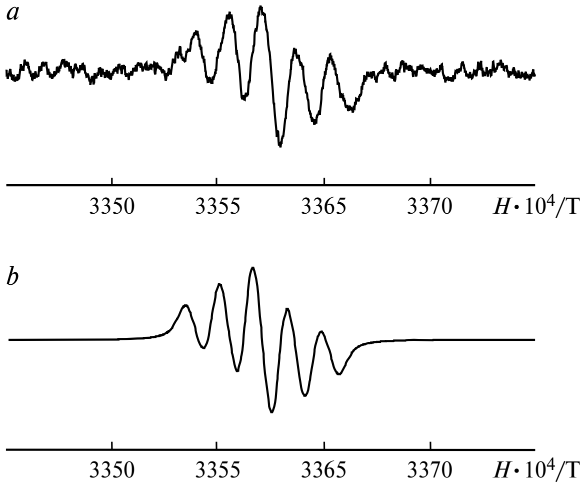


Fig. 1. Experimental (*a*) and model (*b*) ESR spectra of the radical cations generated during EO of 2*H*-imidazole 1,3-dioxide **2c** in the 0.1 *M* Et_4NClO_4—MeCN system (20 °C) (simulation parameters: $a_N(2N) = 1.65 \cdot 10^{-4}$ T, $\Gamma^0 = 1.7 \cdot 10^{-5}$ T).

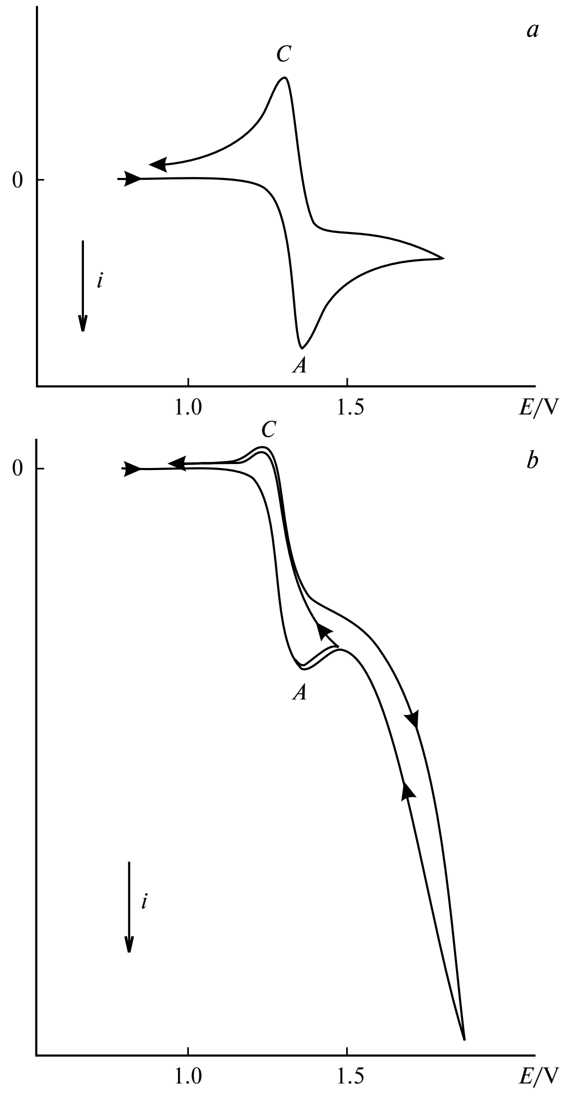
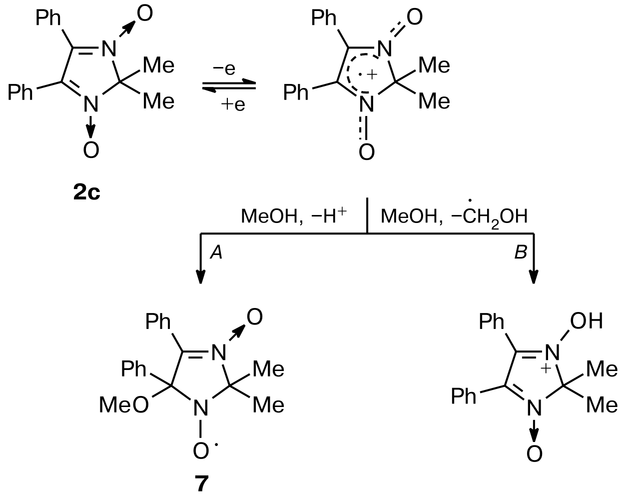


Fig. 2. Cyclic voltammograms of a $5 \cdot 10^{-3}$ *M* solution of compound **2c** in 0.1 *M* Et_4NClO_4 in MeCN without MeOH (*a*) and with 5% MeOH (*b*).

is rather low and does not increase with time. Cathodic peak *C* on the CV curve of compound **2c** does not disappear completely but is reproduced on repeated application of the potential. This may be due to the abstraction of the H atom from MeOH when it reacts with the RC derived from **2c** (see Scheme 3, pathway *B*); this was observed previously for the reaction of the pyridine *N*-oxide RC with alcohols[10—12] and is caused, apparently, by predominant localization of the spin density in the RC formed from **2c** on the *N*-oxide oxygen atoms.

Scheme 3



Thus, the EO of substituted 2*H*-imidazole *N*-oxides and *N*,*N*-dioxides, except for 4,5-diphenyl-2,2-dimethyl-2*H*-imidazole *N*,*N*-dioxide, is irreversible due to the low stability of the initial oxidation products on the CV time scale. The ease of oxidation of *N*-oxides **1—3** is determined by the number of *N*-oxide O atoms in the ring, the presence of a quaternary N atom in position 3, and the effect of substituents in positions 4 and 5 of the ring. The low reactivity of 4,5-disubstituted 2*H*-imidazole *N*-oxides **1** and **2** in oxidative methoxylation is apparently due to the high stability of *N*-oxides **1** against oxidation and to the high degree of spin density localization on the O atoms in the RC derived from 2*H*-imidazole *N*,*N*-dioxides **2**.

Experimental

The IR spectra of crystalline samples were obtained in KBr pellets (concentration 0.25%, thickness 1 mm) on a Specord M-80 spectrometer, and UV spectra were measured on a Specord UV-VIS spectrometer in EtOH. ^{1}H and ^{13}C NMR spectra were recorded on a Bruker AC-200 instrument for 1—5% solutions using the solvent signal as the standard. The ^{13}C NMR signals of compounds **1c**,**d**, and **3b** were assigned resorting to previously obtained data.[3,13,14]

Cyclic voltammograms of compounds **1a—k**, **2a—d**, and **3** were measured on a modified LP-7e polarograph using a three-electrode cell and a 0.1 *M* solution of Et_4NClO_4 as the supporting electrolyte. A stationary needle platinum electrode with an area of 8 mm^2 was used as the anode and a platinum helix was used as the auxiliary electrode. The CV curves were measured for $5 \cdot 10^{-3}$ *M* solutions of compounds in MeCN, the cell volume was 5 mL. The electrooxidation potentials were measured relative to a saturated aqueous calomel electrode (s.c.e.) with a salt bridge containing an electrolyte solution. The potentials were determined with an accuracy of 0.01 V.

The electrogeneration of the RC from **2c** was done in a three-electrode cell placed into the Bruker ESP-300 spectrometer resonator at a voltage corresponding to the diffusion current on the voltammetric curve. The cell design was similar to that described previously.[15] A $5 \cdot 10^{-3}$ *M* solution of **2c** in a 0.1 *M* solution of Et_4NClO_4 in MeCN was used to generate the RC from **2c**. Oxygen was removed by purging the solution with argon.

Acetonitrile was purified by distillation over $KMnO_4$ and P_2O_5; MeOH was distilled over $Mg(OMe)_2$. The characteristics and the synthetic procedures of compounds **1a**,**b**,**e—k**, **2a—d**, and **3a** listed in Table 1 were described previously.[4,16,17]

2,2,4-Trimethyl-5-(2-nitrophenyl)-2*H*-imidazole 1-oxide (1c) and 2,2,4-trimethyl-5-(4-nitrophenyl)-2*H*-imidazole-1-oxide (1d). A nitration mixture (10 mL) prepared by careful mixing of 100 mL of 56% HNO_3 with 200 mL of 60% oleum was added dropwise with stirring to a solution of 2,2,4-trimethyl-5-phenyl-2*H*-imidazole 1-oxide (**1b**) (1.3 g, 6 mmol) in 35 mL of concentrated H_2SO_4. The reaction mixture was stirred for 2 h and poured into 300 mL of water cooled to 10 °C. The resulting mixture was neutralized with Na_2CO_3 and extracted with $CHCl_3$ (3×50 mL). The extract was dried with $MgSO_4$, chloroform was evaporated, and the residue was chromatographed on a column with silica gel (Kieselgel 60, 60—200, Merck) using $CHCl_3$ as the eluent.

Compound **1c**. Yield 0.9 g (60%), m.p. 122—124 °C (hexane—AcOEt, 1 : 2). Found (%): C, 58.2; H, 5.3; N, 17.1. $C_{12}H_{13}N_3O_3$. Calculated (%): C, 58.4; H, 5.3; N, 17.0. IR, ν/cm^{-1}: 1520, 1370 (NO_2). UV (EtOH), λ_{max}/nm (logε): 290 (3.91). ^{1}H NMR ($CDCl_3$), δ: 1.50 (s, 6 H, *gem*-Me); 2.17 (s, 3 H, Me); 7.53 and 8.11 (both m, 3 H and 1 H, C_6H_4). ^{13}C NMR ($CDCl_3$), δ: 17.4 (Me); 23.7 (*gem*-Me); 100.4 (C(2)); 127.7 (C_i, Ph); 131.0 (C_p, Ph); 131.0 and 133.8 (C_m, Ph); 132.4 (C(5)); 125.3 and 148.1 (C_o, Ph); 164.4 (C(4)).

Compound **1d**. Yield 0.45 g (30%), m.p. 95—97 °C (hexane—AcOEt, 1 : 2). Found (%): C, 58.7; H, 5.5; N, 17.1. $C_{12}H_{13}N_3O_3$. Calculated (%): C, 58.4; H, 5.3; N, 17.0. IR, ν/cm^{-1}: 1520, 1350 (NO_2). UV (EtOH), λ_{max}/nm (logε): 337 (4.00), 265 (4.15). ^{1}H NMR ($CDCl_3$), δ: 1.53 (s, 6 H, *gem*-Me); 2.53 (s, 3 H, C(4)Me); 8.10, 8.23 (AA´BB´-system, C_6H_4, J = 9 Hz). ^{13}C NMR ($CDCl_3$), δ: 19.5 (Me); 24.4 (*gem*-Me); 100.5 (C(2)); 123.8 (C_o, Ph); 128.6 (C_m, Ph); 132.0 (C_i, Ph); 133.8 (C(5)); 147.8 (C_p, Ph); 164.4 (C(4)).

2,2,3,4-Tetramethyl-5-phenyl-2*H*-imidazolium 1-oxide methylsulfate (3b). Dimethyl sulfate (1.3 mL, 13.4 mmol) was added to a solution of 2,2,4-trimethyl-5-phenyl-2*H*-imidazole 1-oxide (**1b**) (1.6 g, 7.9 mmol) in 50 mL of anhydrous Et_2O. After 0.5 h, the solution was filtered and the solvent was removed *in vacuo*. The residue was carefully heated to 60 °C, kept at this temperature for 0.5 h, and triturated with 15 mL of anhydrous Et_2O. The precipitate was filtered off and recrystallized from propan-2-ol to give 1.3 g (50%), m.p. 172—174 °C. Found (%): C, 51.2; H, 6.1; N, 8.5; S, 9.4. $C_{14}H_{20}N_2O_5S$. Calculated (%): C, 51.2; H, 6.1; N, 8.5; S, 9.8. IR, ν/cm^{-1}: 1630, 1560, 1500 (C=N, C=C). UV (EtOH), λ_{max}/nm (logε): 350 (3.94), 232 (4.11). ^1H NMR (CD_3OD), δ: 1.63 (s, 6 H, *gem*-Me); 2.67 (s, 3 H, C(4)Me); 3.53 (s, 3 H, NMe); 3.73 (s, 3 H, OMe); 7.60 (m, 5 H, Ph). ^{13}C NMR (CD_3OD), δ: 15.3 (C(4)$\underline{\text{Me}}$); 22.9 (*gem*-Me); 33.5 (N(3)$\underline{\text{Me}}$); 54.3 (OMe); 99.6 (C(2)); 123.6 (C_i, Ph); 129.4 (C_o, Ph); 129.6 (C_m, Ph); 132.0 (C_p, Ph); 139.2 (C(5)); 176.6 (C(4)).

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