DOI 10.1007/s10593-015-1796-0

Chemistry of Heterocyclic Compounds **2015**, *51*(10), 918–922

New fluorescent 3*H*-imidazo[4,5-*e*][2,1]benzoxazoles: synthesis, spectroscopic characterization, and antibacterial activity

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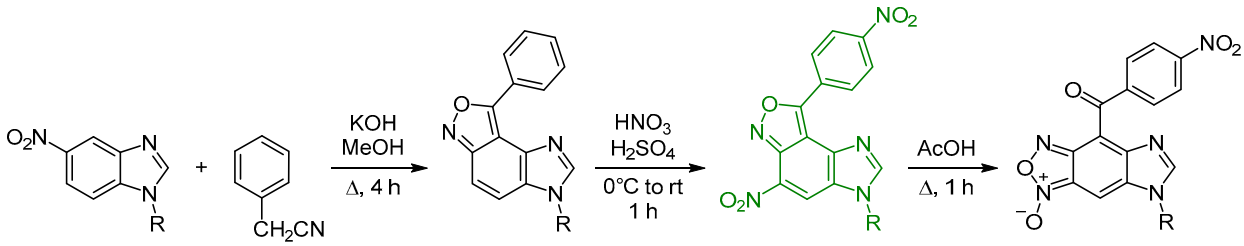
Published in Khimiya Geterotsiklicheskikh Soedinenii, 2015, *51*(10), 918–922

Submitted September 9, 2015
Accepted after revision October 17, 2015



The new fluorophores of the 3*H*-imidazo[4,5-*e*][2,1]benzoxazoles series were synthesized by the regioselective nitration of 3-alkyl-8-phenyl-3*H*-imidazo[4,5-*e*][2,1]benzoxazoles. The latter compounds were obtained from the reaction of 1-alkyl-5-nitro-1*H*-benzimidazoles with benzyl cyanide in basic MeOH solution. The structures of synthesized compounds were established using spectral (UV-vis, IR, ^{1}H NMR, ^{13}C NMR, and NOESY) and analytical data. Furthermore, it was found that these fluorophores underwent thermal rearrangement to new 5*H*-imidazo[4,5-*f*][2,1,3]benzoxadiazole 3-oxides in AcOH in moderate yields. The fluorescence properties and antibacterial activities of new compounds against Gram-positive and Gram-negative bacterial species were also studied.

Keywords: 3*H*-imidazo[4,5-*e*][2,1]benzoxazole, 5*H*-imidazo[4,5-*f*][2,1,3]benzoxadiazole 3-oxide, emission and absorption spectra, fluorescence, NOESY.

A huge number of heterocyclic systems which include mainly five- and six-membered compounds represent a diverse group of molecular scaffolds. Several of such heterocyclic scaffolds have been successfully incorporated into novel drug leads and therapeutic agents.[1–3]

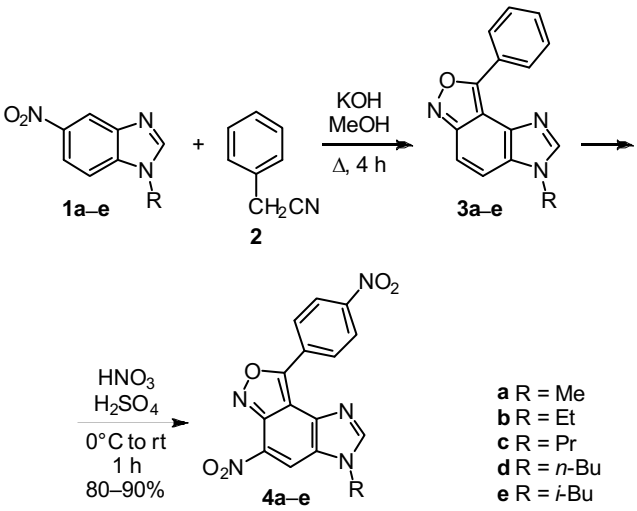
Imidazoles exhibit wide ranges of biological activities and optical properties as fluorescence compounds, dyes, and TPA (Two-photon absorption) materials[4–6] making them attractive compounds for organic chemists. Also, many commercial fluorescent brighteners for application to synthetic fibers contain an imidazole moiety.[7]

On the other hand, 2,1-benzoxazole derivatives, e.g., risperidone, are prescribed as antipsychotic drugs.[8] Compounds of this class play a key role in many organic reactions,[9] notably those leading to anthranilic acids. A combination of the 2,1-benzoxazole moiety with the imidazole nucleus may enhance optical and biological properties. Taking this fact into consideration and in continuation of our studies on the synthesis of new bioactive[10–12] and fluorescent heterocyclic compounds,[13–15] we have synthesized some new fluorescent 3*H*-imidazo-[4,5-*e*][2,1]benzoxazoles. Spectroscopic characteristics of these compounds were studied and the structure of the major products was established by Nuclear Overhauser Effect Spectroscopy (NOESY) experiment. Furthermore, it was found that these fluorophores can be converted to new 5*H*-imidazo[4,5-*f*][2,1,3]benzoxadiazole 3-oxides in AcOH. Antibacterial activities of the new compounds against Gram-positive and Gram-negative bacterial species were also studied.

The key compounds **3a**–**e** were obtained from the reaction of 1-alkyl-5-nitro-1*H*-benzimidazoles **1a**–**e** with benzyl cyanide (**2**) in basic MeOH solution.[16] Nitration of compounds **3a**–**e** was carried out using a mixture of sulfuric and nitric acids and led to the formation of new 3-alkyl-5-nitro-8-(4-nitrophenyl)-3*H*-imidazo[4,5-*e*][2,1]benzoxazoles **4a**–**e** in excellent yields (Scheme 1).

Structural assignments of the new compounds **4a–e** were based on their spectral and microanalytical (C, H, and N) data. For example, in the aromatic region of the ^{1}H NMR spectrum of compound **4e** there are two doublet signals at 9.21 and 8.51 ppm attributed to four aromatic protons of 4-nitrophenyl ring and two singlet signals at 8.76 and 8.21 ppm assignable to two aromatic protons of imidazole

Scheme 1

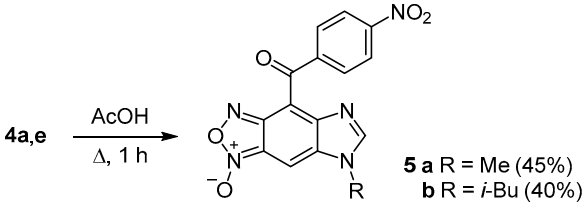


and phenyl rings, respectively. Also, there are 15 different carbon atom signals in the ^{13}C NMR spectrum of compound **4e**. Moreover, the IR spectrum of compound **4e** in KBr showed two absorption bands at 1546 and 1356 cm^{-1} corresponding to the nitro group. All this evidence taken in conjunction with molecular ion peak at *m*/*z* 381 $[M]^+$ support the structure of compound **4e**. However, these spectral characteristics are consistent with another possible structure which is isomeric with compounds **4a**–**e**. Nitration of compounds **3a**–**e** can lead to addition of the nitro group at C-4 or C-5 position. Fortunately, there are some convincing reasons that confirm the structure of compounds **4a**–**e** as the major products of the nitration of compounds **3a**–**e**. For example, electron release from the imidazole ring would make C-5 position more susceptible to electrophilic attack than C-4 position. Furthermore the latter is also hindered by its proximity to the alkyl group at N-3 position.

The data of NOESY experiment for compound **4d** showed a massive cross peak between the singlet of the H-4 proton at 8.23 ppm and the signal of NCH_2 protons of N-3 alkyl substituent at 4.41 ppm (t, J = 7.2 Hz), confirming that the nitration of compound **3d** resulted in the formation of 5-nitro-substituted compound **4d** (Fig. 1).

Compounds **4a**,**e** underwent thermal rearrangement in AcOH[17] giving new 5-alkyl-8-(4-nitrobenzoyl)-5*H*-imidazo-[4,5-*f*][2,1,3]benzoxadiazole 3-oxides **5a**,**b** in moderate yields (Scheme 2). Structural assignments of compounds **5a**,**b** were based on their spectral and microanalytical data.

Scheme 2



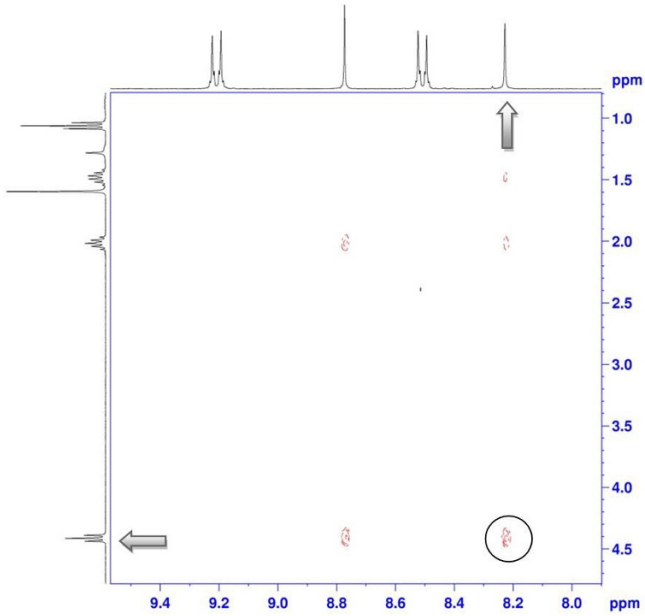


Figure 1. NOESY spectrum of compound **4d**.

Compounds **3a**–**e**, **4a**–**e**, and **5a**,**b** were spectrally characterized by UV-Vis and fluorescence spectrophotometry methods in the wavelength range of 200–1000 nm. The emission spectra of the precursors **3a**–**e** don't show any fluorescence emission peak (λ_{abs} 282–285 nm) and the emission spectra of compounds **5a**,**b** show weak emission peak at high concentration (10^{-2} mol·l^{-1}). The fluorescence absorption and emission spectra of compounds **4a**–**e** were recorded in chloroform at concentrations of $5\cdot10^{-5}$ and $1\cdot10^{-6}$ mol·l^{-1}, respectively. The absorbance and fluorescence spectral properties of compounds **4a**–**e** are similar to each other, and the numerical data are presented in Table 1.

Values of extinction coefficient (ε) were calculated as the slope of the plot of absorbance *vs* concentration. The fluorescence excitation (λ_{ex}) wavelengths at 425 and 390 nm (λ_{ex}/nm) were used for compounds **4a**–**e** and **5a**,**b**, respectively. The fluorescence quantum yields (Φ_F) of compounds **4a**–**e** and **5a**,**b** were determined *via* comparison methods, using fluorescein as a standard sample in 0.1 M NaOH and MeOH solution.[18] The used value of the

Table 1. Spectroscopic data for compounds **4a**–**e**, **5a**,**b** at 298 K

Compound	λ_{abs}, nm*	$\varepsilon\cdot10^{-4}$, $l\cdot(mol\cdot cm)^{-1}$**	λ_{ex}, nm***	λ_{flu}, nm*[4]	Φ_F*[5]
4a	420	1.30	425	515	0.39
4b	425	1.50	425	520	0.43
4c	425	1.60	425	525	0.42
4d	425	1.70	425	525	0.47
4e	425	1.60	425	525	0.45
5a	390	0.08	390	480	0.07
5b	390	0.09	390	480	0.09

* Wavelengths of maximum absorbance.
** Extinction coefficient.
*** Wavelengths of fluorescence excitation.
*[4] Wavelengths of fluorescence emission.
*[5] Fluorescence quantum yield.

fluorescein emission quantum yield is 0.79. As can be seen in Table 1, the Φ_F of compound **4d** (R = *n*-Bu) is the highest and the lowest Φ_F is attributed to compound **5a**. The fluorescence absorptions of compounds **4a**–**e** at 420–425 nm can be corresponded to π–π* transitions from N-3 atom in the imidazole cycle and oxygen atom in the isoxazole cycle to the nitro group.

The relatively high fluorescence emission of the new compounds **4a**–**e** can be explained in terms of the extended conjugation pathway.[14,15] Comparing the resonance structures of compounds **4a**–**e** and **5a**,**b** demonstrates that π-electron delocalization in compounds **4a**–**e** can occur much more easily compared to compounds **5a**,**b**.

The solvatochromic properties of compound **4a** were deduced from the fluorescence absorption and emission spectra (Fig. 2). The fluorescence absorption and emission spectra of compound **4a** in polar solvents undergo a relatively modest red shift. Increasing the solvent polarity stabilizes the molecule excited state comparative to the molecule ground state with the red shift of the absorption maximum as the experimentally observed result (Table 2). For example, in the absorption and emission spectra of compound **4a**, λ_{abs} and λ_{flu} shift from 405 to 430 nm and 475 to 555 nm, respectively, as the solvent changes from *n*-hexane to methanol (Table 2).

The antibacterial activity of compounds **3a**–**e**, **4a**–**e**, and **5a**,**b** was tested against standard strains of two Gram-positive (*Staphylococcus aureus* ATCC 29213 and *Bacillus subtilis* ATCC 6633) and two Gram-negative bacteria (*Escherichia coli* ATCC 10538 and *Salmonella typhimurium* ATCC 14028) species (Table 3), using the broth microdilution method as previously described.[19] We used amoxicillin and ciprofloxacin as reference compounds in the evaluation of antibacterial activity. The lowest concentration of the antibacterial agent that prevents growth of the test organism, as detected by lack of visual turbidity (matching the negative growth control), is assigned the minimum inhibitory concentration (MIC). Experimental details of the tests can be found in our earlier study.[12]

Table 2. Spectroscopic data for compound **4a** at 298 K in different solvents

Solvent	λ_{abs}, nm	λ_{flu}, nm
n-Hexane	405	475
1,4-Dioxane	410	495
EtOAc	415	510
Acetone	425	530
DMF	425	545
MeOH	430	555

The antimicrobial tests performed with compounds **3a**–**e**, **4a**–**e**, and **5a**,**b** confirmed that only compounds **4a**–**e** and **5a**,**b** were effective against both Gram-positive and Gram-negative bacteria. Also, the results revealed that new compound **5b** displayed greater antibacterial activity against the *Staphylococcus aureus* ATCC 29213, *Bacillus subtilis* ATCC 6633, *Escherichia coli* ATCC 10538, and *Salmonella typhimurium* ATCC 14028 species compared to the well-known antibacterial agent amoxicillin (Table 3).

In conclusion, we have synthesized some new donor–acceptor fluorescent heterocyclic compounds in excellent yields. These compounds were obtained from regio-selective nitration of 3-alkyl-8-(4-nitrophenyl)-3*H*-imidazo-[4,5-*e*][2,1]benzoxazoles at room temperature. Confirmation of the correct structure has been achieved by NOESY experiment. The fluorescence properties of the obtained nitro derivatives together with high antibacterial activity,

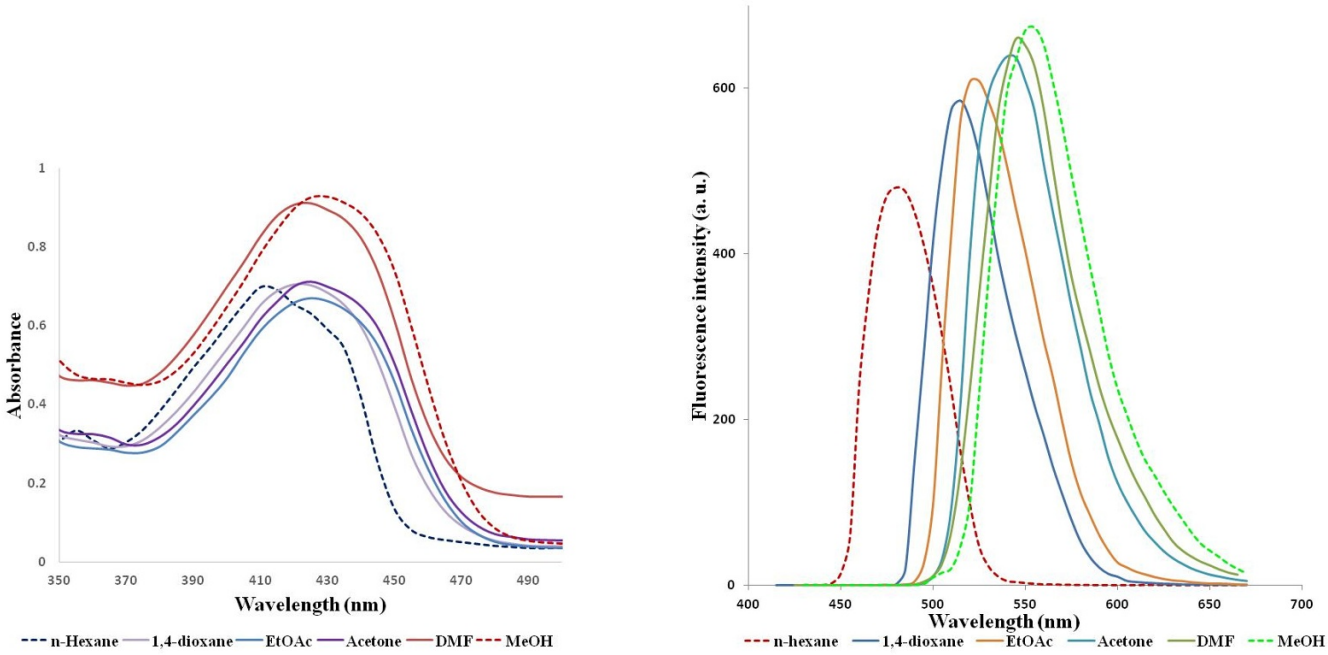


Figure 2. Visible absorption (left) ($5 \cdot 10^{-5}$ mol·l^{-1}) and emission spectra (right) ($1 \cdot 10^{-6}$ mol·l^{-1}) of compound **4a** in different solvents.

Table 3. Inhibitory activity (MIC, μg·ml^{-1}) of compounds **4a**–**e**, **5a**,**b** against bacteria

Compound	*Staphylococcus aureus* (ATCC 29213)	*Bacillus subtilis* (ATCC 6633)	*Escherichia coli* (ATCC 10538)	*Salmonella typhimurium* (ATCC 14028)
4a	125	125	150	150
4b	95	95	115	115
4c	80	80	115	115
4d	55	55	110	100
4e	75	60	90	85
5a	5	5	15	15
5b	3	3	10	5
Ciprofloxacin	16	0.05	1	>128
Amoxicillin	25	0.06	150	>128

can offer an excellent opportunity for the study of physiological functions of bacteria at single-cell level.[19] In addition, the new synthesized compounds can play a key role in many organic reactions, leading to the formation of the other significant compounds. For example, rearrangement of the nitrated 3*H*-imidazo[4,5-*e*][2,1]benzoxazoles in boiling AcOH leads to the new 5*H*-imidazo[4,5-*f*][2,1,3]-benzoxadiazole 3-oxide derivatives.

Experimental

Absorption and fluorescence spectra were recorded on a Varian 50-bio UV-Visible spectrophotometer and a Varian Cary Eclipse spectrofluorophotometer, respectively. UV-vis and fluorescence scans were recorded from 200 to 1000 nm. IR spectra were obtained on a Tensor 27 spectrometer in KBr pellets; only noteworthy absorption bands are listed. ^{1}H and ^{13}C NMR spectra were recorded on a Bruker Avance DRX-400 FT spectrometer (400 and 100 MHz, respectively) in $CDCl_3$, internal standard TMS. Mass spectra were recorded on a Varian Mat, CH-7 mass spectrometer at 70 eV. Elemental analysis was performed on a Thermo Finnigan Flash EA microanalyzer. Melting points were measured on an Electrothermaltype-9100 melting point apparatus. All measurements were carried out at room temperature.

All reagents and solvents used in this work were purchased from Merck. Amoxicillin and ciprofloxacin were purchased from Sigma–Aldrich. All solvents were dried according to standard procedures. Compounds **1a**–**e**[20] and **3a**–**e**[10] were synthesized by the literature methods. The microorganisms *Staphylococcus aureus* ATCC 29213, *Bacillus subtilis* ATCC 6633, *Escherichia coli* ATCC 10538, *Salmonella typhimurium* ATCC 14028 were purchased from Pasteur Institute of Iran.

Synthesis of compounds 4a–e by nitration of compounds 3a–e (General method). To a solution of compound **3a**–**e** (20 mmol) in concd. H_2SO_4 (4 ml), 65% HNO_3 (1 ml) was added with stirring over 0.5 h period keeping the temperature at 0–5°C. After the addition was completed, the mixture was allowed to warm to room temperature with stirring for 0.5 h. Then the mixture was poured into crushed ice and water (200 ml), the precipitated solid was filtered off, washed with water, and dried to give the crude compounds **4a–e**. More purification was achieved by recrystallization from acetone.

3-Methyl-5-nitro-8-(4-nitrophenyl)-3*H*-imidazo[4,5-*e*]-[2,1]benzoxazole (4a). Yield (85%), shiny yellow powder, mp 295–297°C. IR spectrum, ν, cm^{-1}: 1353, 1546 (NO_2). ^{1}H NMR spectrum, δ, ppm (*J*, Hz): 4.57 (3H, s, NCH_3); 8.27 (1H, s, H Ar); 8.51 (2H, d, *J* = 8.8, H Ar); 8.80 (1H, s, H Ar); 9.21 (2H, d, *J* = 8.8, H Ar). ^{13}C NMR spectrum, δ, ppm: 46.6; 112.4; 119.5; 125.5; 128.2; 129.7; 131.5; 133.8; 140.9; 146.0; 149.6; 152.2; 164.7. Mass spectrum, *m*/*z* (I_{rel}, %): 339 $[M]^+$ (7), 338 (30), 294 (35), 249 (37), 235 (43), 91 $[C_7H_7]^+$ (100). Found, %: C 52.90; H 2.64; N 20.51. $C_{15}H_9N_5O_5$. Calculated, %: C 53.10; H 2.67; N 20.64.

3-Ethyl-5-nitro-8-(4-nitrophenyl)-3*H*-imidazo[4,5-*e*]-[2,1]benzoxazole (4b). Yield (80%), shiny yellow powder, mp 287–289°C. IR spectrum, ν, cm^{-1}: 1353, 1546 (NO_2). ^{1}H NMR spectrum, δ, ppm (*J*, Hz): 1.77 (3H, t, *J* = 7.2, $NCH_2C\underline{H}_3$); 4.49 (2H, t, *J* = 7.2, $NC\underline{H}_2CH_3$); 8.25 (1H, s, H Ar); 8.52 (2H, d, *J* = 8.8, H Ar); 8.79 (1H, s, H Ar); 9.21 (2H, d, *J* = 8.8, H Ar). ^{13}C NMR spectrum, δ, ppm: 16.0; 43.2; 112.4; 119.6; 125.5; 128.1; 129.8; 131.6; 133.9; 140.7; 146.2; 149.6; 152.2; 164.8. Mass spectrum, *m*/*z* (I_{rel}, %): 353 $[M]^+$ (5), 352 (21), 308 (39), 263 (34), 235 (41), 91 $[C_7H_7]^+$ (100). Found, %: C 54.18; H 3.13; N 20.01. $C_{16}H_{11}N_5O_5$. Calculated, %: C 54.40; H 3.14; N 19.82.

5-Nitro-8-(4-nitrophenyl)-3-propyl-3*H*-imidazo[4,5-*e*]-[2,1]benzoxazole (4c). Yield (87%), shiny yellow powder, mp 283–285°C. IR spectrum, ν, cm^{-1}: 1357, 1543 (NO_2). ^{1}H NMR spectrum, δ, ppm (*J*, Hz): 1.10 (3H, t, *J* = 7.2, $NCH_2CH_2C\underline{H}_3$); 2.05–2.11 (2H, m, $NCH_2C\underline{H}_2CH_3$); 4.39 (2H, t, *J* = 7.2, $NC\underline{H}_2CH_2CH_3$); 8.24 (1H, s, H Ar); 8.51 (2H, d, *J* = 8.8, H Ar); 8.78 (1H, s, H Ar); 9.21 (2H, d, *J* = 8.8, H Ar). ^{13}C NMR spectrum, δ, ppm: 12.4; 25.1; 49.1; 112.3; 119.7; 125.5; 128.3; 129.9; 131.5; 133.6; 140.7; 146.1; 149.8; 152.3; 164.9. Mass spectrum, *m*/*z* (I_{rel}, %): 367 $[M]^+$ (3), 366 (15), 322 (30), 280 (25), 235 (37), 91 $[C_7H_7]^+$ (100). Found, %: C 55.43; H 3.54; N 19.23. $C_{17}H_{13}N_5O_5$. Calculated, %: C 55.59; H 3.57; N 19.07.

3-Butyl-5-nitro-8-(4-nitrophenyl)-3*H*-imidazo[4,5-*e*]-[2,1]benzoxazole (4d). Yield (85%), shiny yellow powder, mp 277–279°C. IR spectrum, ν, cm^{-1}: 1356, 1541 (NO_2). ^{1}H NMR spectrum, δ, ppm (*J*, Hz): 1.06 (3H, t, *J* = 7.2, $NCH_2CH_2CH_2C\underline{H}_3$); 1.42– 1.52 (2H, m, $NCH_2CH_2C\underline{H}_2CH_3$); 1.99 (2H, t, *J* = 7.2, $NCH_2C\underline{H}_2CH_2CH_3$); 4.41 (2H, t, *J* = 7.2, $NC\underline{H}_2CH_2CH_2CH_3$); 8.23 (1H, s, H Ar); 8.51 (2H,

d, J = 8.8, H Ar); 8.77 (1H, s, H Ar); 9.21 (2H, d, J = 8.8, H Ar). ^{13}C NMR spectrum, δ, ppm: 12.5; 21.4; 32.9; 54.6; 112.5; 119.8; 125.5; 128.1; 130.1; 131.6; 133.6; 140.5; 146.3; 149.7; 152.5; 164.6. Mass spectrum, *m*/*z* (I_{rel}, %): 381 $[M]^+$ (2), 380 (13), 336 (35), 291 (23), 235 (43), 91 $[C_7H_7]^+$ (100). Found, %: C 56.49; H 3.94; N 18.19. $C_{18}H_{15}N_5O_5$. Calculated, %: C 56.69; H 3.96; N 18.36.

3-Isobutyl-8-(4-nitrophenyl)-5-nitro-3*H*-imidazo[4,5-*e*]-[2,1]benzoxazole (4e). Yield (90%), shiny yellow powder, mp 265–267°C. IR spectrum, ν, cm^{-1}: 1356, 1546 (NO_2). ^{1}H NMR spectrum, δ, ppm (J, Hz): 1.07 (6H, d, J = 6.9, $NCH_2CH(C\underline{H}_3)_2$); 2.29–2.36 (1H, m, $NCH_2C\underline{H}Me_2$); 4.21 (2H, d, J = 7.2, $NC\underline{H}_2CHMe_2$); 8.21 (1H, s, H Ar); 8.51 (2H, d, J = 8.8, H Ar); 8.76 (1H, s, H Ar); 9.21 (2H, d, J = 8.8, H Ar). ^{13}C NMR spectrum, δ, ppm: 21.1; 31.0; 54.8; 112.3; 119.8; 125.5; 128.5; 129.9; 131.6; 133.6; 140.6; 146.4; 149.8; 152.2; 164.9. Mass spectrum, *m*/*z* (I_{rel}, %): 381 $[M]^+$ (5), 380 (19), 336 (31), 291 (27), 235 (43), 91 $[C_7H_7]^+$ (100). Found, %: C 56.51; H 3.95; N 18.49. $C_{18}H_{15}N_5O_5$. Calculated, %: C 56.69; H 3.96; N 18.37.

Synthesis of compounds 5a,b by thermal rearrangement of compounds 4a,e (General method). Compounds **4a**,**e** (10 mmol) were refluxed for 1 h in glacial AcOH (10 ml). The solution was cooled, and an equal volume of water was added. The precipitated solid was filtered off, washed with water, and dried to give the crude compounds **5a**,**b**. More purification was achieved by recrystallization from EtOH.

5-Methyl-8-(4-nitrobenzoyl)-5*H*-imidazo[4,5-*f*][2,1,3]-benzoxadiazole 3-oxide (5a). Yield (45%), yellow powder, mp > 300°C (decomp.). IR spectrum, ν, cm^{-1}: 1353, 1546 (NO_2), 1687 (C=O). ^{1}H NMR spectrum, δ, ppm (J, Hz): 4.59 (3H, s, NCH_3); 6.89 (1H, s, H Ar); 8.08 (2H, d, J = 8.9, H Ar); 8.27 (2H, d, J = 8.9, H Ar); 8.43 (1H, s, H Ar). ^{13}C NMR spectrum, δ, ppm: 47.3; 105.9; 117.4; 129.4; 129.9; 131.7; 132.8; 133.8; 139.2; 145.7; 150.5; 159.6; 187.5. Mass spectrum, *m*/*z* (I_{rel}, %): 339 $[M]^+$ (2), 338 (9), 325 (17), 309 (29), 264 (53), 91 $[C_7H_7]^+$ (100). Found, %: C 53.00; H 2.65; N 20.39. $C_{15}H_9N_5O_5$. Calculated, %: C 53.10; H 2.67; N 20.64.

5-Isobutyl-8-(4-nitrobenzoyl)-5*H*-imidazo[4,5-*f*][2,1,3]-benzoxadiazole 3-oxide (5b). Yield (40%), yellow powder, mp > 300°C (decomp.). IR spectrum, ν, cm^{-1}: 1353, 1546 (NO_2), 1687 (C=O). ^{1}H NMR spectrum, δ, ppm (J, Hz): 1.11 (6H, d, J = 6.9, $NCH_2CH(C\underline{H}_3)_2$); 2.33–2.40 (1H, m, $NCH_2C\underline{H}(CH_3)_2$); 4.25 (2H, d, J = 7.2, $NC\underline{H}_2CH(CH_3)_2$); 6.93 (1H, s, H Ar); 8.09 (2H, d, J = 8.9, H Ar); 8.25 (2H, d, J = 8.9, H Ar); 8.41 (1H, s, H Ar). ^{13}C NMR spectrum, δ, ppm: 21.6; 30.5; 55.9; 106.2; 117.5; 129.4; 129.9; 131.9; 132.5; 133.6; 139.3; 145.7; 150.9; 159.6; 187.8. Mass spectrum, *m*/*z* (I_{rel}, %): 381 $[M]^+$ (3), 380 (12), 325 (19), 309 (33), 264 (58), 91 $[C_7H_7]^+$ (100). Found, %: C 56.53; H 3.93; N 18.15. $C_{18}H_{15}N_5O_5$. Calculated, %: C 56.69; H 3.96; N 18.36.

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