

**An efficient synthesis of substituted 4,5-dihydroxyimidazolidin-2-ones
by oxidation of imidazolin-2-ones with HNO₃**

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General

All reagents (Acros) were used as purchased, unless otherwise indicated. HNO₃ (84%) was prepared by the literature procedure [W. L. F. Armarego and C. L. L. Chai, *Purification of Laboratory Chemicals*, 7th ed., Elsevier, 2013, 593]. ¹H and ¹³C NMR spectra were recorded at 20 to 31 °C using Bruker AM300, Bruker AV600 (¹H, 300.13 MHz; ¹³C, 75.5 MHz, 150.9 MHz). Chemical shifts are reported in the δ scale relative to Me₄Si as internal standard. High resolution mass spectra (HRMS) were recorded on a Bruker micrOTOF II instrument using the electrospray ionization method (ESI). Infrared spectra were obtained on a BrukerALPHA spectrometer using KBr pellets and reported in wavenumbers (cm⁻¹). Melting points were determined in a SMP10 instrument (Stuart). The oxidation reactions were carried out by using UV lamp VL-6.C (254 nm, 6 W).

Spectral data of compounds **1b-i**, **4b-i**, **5a,b**, **6**

rac-(4*S*,5*R*)-1,3-Diethyl-4,5-dihydroxy-4,5-diphenylimidazolidin-2-one **1b**. Yield 24%* (0.296 g), 96%** (2.190 g). White powder, mp: 161-164°C; IR (neat, cm⁻¹): 3436, 3245, 3087, 3046, 3031 (Ar), 2990, 2971, 2944, 2873 (Alk), 1673 (C=O). ¹H NMR: (300 MHz, DMSO-*d*₆) δ 1.15 (t, 6H, *J* = 7.0 Hz, (CH₂CH₃)), 2.93 (dq, 2H, *J* = 7.0 Hz, *J* = 14.0 Hz, CH₂CH₃), 3.14 (dq, 2H, *J* = 7.0 Hz, *J* = 14.0 Hz, CH₂CH₃), 6.49 (s, 2H, OH), 6.89 – 6.97 (m, 10H, Ph), 6.99 – 7.07 (m, 10H, Ph). ¹³C NMR (75 MHz, DMSO-*d*₆), δ = 15.06 (CH₂CH₃), 30.63, 35.46 (CH₂CH₃), 93.09 (Ph-C-C-Ph), 126.99, 127.07, 127.20 (CH(Ph)), 138.75 (C(Ph)), 159.49 (C=O). HRMS-ESI (*m/z*): calcd for C₁₉H₂₂N₂O₃ [M+H]⁺: 327.1703, found: 327.1713; calcd for C₁₉H₂₂N₂O₃ [M+Na]⁺: 349.1523, found: 349.1526; calcd for C₁₉H₂₂N₂O₃ [M+K]⁺: 365.1262, found: 365.1264.

*synthesized by oxidation of compound **4b** with ¹O₂

** synthesized by oxidation of compound **4b** HNO₃

The mixture of *meso*-(4*S*,5*R*)-1,3-diethyl-4,5-dihydroxy-4,5-bis(4-methoxyphenyl)imidazolidin-2-one *cis*-**1c** and *rac*-(4*R*,5*R*)-1,3-diethyl-4,5-dihydroxy-4,5-bis(4-methoxyphenyl)imidazolidin-2-one *trans*-**1c** (3:1). Yield 97% (2.621 g); white powder, mp: 152-154°C; IR (neat, cm⁻¹): 3446, 3207, 3076 (Ar), 3000, 2982, 2963, 2940, 2838 (Alk), 1677 (C=O). ¹H NMR: (300 MHz, DMSO-*d*₆) δ 1.09-1.21 (m, 6H, CH₂CH₃(**1c-cis**+**1c-trans**)), 2.92 (dq, 2H, *J* = 7.3 Hz, *J* = 14.3 Hz, CH₂CH₃(**1c-cis**)), 3.08 (dq, 2H, *J* = 7.0 Hz, *J* = 13.9 Hz, CH₂CH₃(**1c-trans**)), 3.64 (s, 3H, OMe(**1c-cis**)), 3.77 (s, 3H, OMe(**1c-trans**)), 5.69 (s, 1H, OH(**1c-cis**)), 6.33 (s, 1H, OH(**1c-trans**)), 6.62 (d, 4H, CH(PMP(**1c-cis**+**1c-trans**))), 6.85 (d, 4H, CH(PMP(**1c-cis**+**1c-trans**))), 7.17 (d, 4H, CH(PMP(**1c-cis**+**1c-trans**))). ¹³C NMR (75 MHz, DMSO-*d*₆), δ = 14.88, 15.23 (CH₂CH₃(**1c-cis**+**1c-trans**)), 35.39, 35.63 (CH₂CH₃(**1c-cis**+**1c-trans**)), 54.97 (OMe(**1c-cis**+**1c-trans**)), 93.02, 93.28 (PMP-C-C-PMP), 112.15, 112.50, 128.37, 129.87, 130.87, 158.34, 158.72, 159.58, 161.04. HRMS-ESI (*m/z*): calcd for C₂₁H₂₆N₂O₅ [M+H]⁺: 387.1914, found: 387.1923; calcd for C₂₁H₂₆N₂O₅ [M+Na]⁺: 409.1734, found: 409.1736; calcd for C₂₁H₂₆N₂O₅ [M+K]⁺: 425.1473, found: 425.1485.

The mixture of *meso*-(4*S*,5*R*)-4,5-dihydroxy-4,5-bis(4-methoxyphenyl)-1,3-dimethylimidazolidin-2-one *cis*-**1d** and *rac*-(4*R*,5*R*)-4,5-dihydroxy-4,5-bis(4-methoxyphenyl)-1,3-dimethylimidazolidin-2-one *trans*-**1d** (1:2.4). Yield 93% (2.330 g); white powder, mp: 175-176 °C; IR (neat, cm⁻¹): 3465, 3270, 3077, 3044, 3002 (Ar), 2953, 2835 (Alk), 1675 (C=O). ¹H NMR: (300 MHz, DMSO-*d*₆) δ 2.59 (s, 6H, NMe(**1d-cis** + **1d-trans**)), 3.64 (s, 6H, OMe(**1d-cis**)), 3.78 (s, 6H, OMe(**1d-trans**)), 5.81 (s, 2H, OH(**1d-trans**)), 6.40 (s, 2H, OH(**1e-cis**)), 6.62 (d, 8H, *J* = 8.6 Hz, CH(PMP(**1d-cis**))), 6.83 (d, 8H, *J* = 8.6 Hz, CH(PMP(**1d-cis**))), 6.87 (d, 8H, *J* = 8.6 Hz, CH(PMP(**1d-trans**))), 7.10 (d, 8H, *J* = 8.5 Hz, CH(PMP(**1d-trans**))). ¹³C NMR (75 MHz, DMSO-*d*₆), δ = 25.58 (NMe(**1d-cis**)), 25.81 (NMe(**1e-trans**)), 54.94 (OMe(**1d-cis**)), 54.98 (OMe(**1d-trans**)), 92.20 (PMP-C-C-PMP(**1d-cis**)), 92.27 (PMP-C-C-PMP(**1d-trans**)), 112.42 (CH(PMP(**1d-trans**))), 112.66 (CH(PMP(**1d-cis**))), 128.19, 129.50, 129.58, 130.14 (PMP(**1d-cis**+**1d-trans**)), 158.33, 158.74, 159.28, 160.58 (C=O, MeOC(PMP) (**1d-cis**+**1d-trans**)). HRMS-ESI (*m/z*): calcd for C₁₉H₂₂N₂O₅ [M+H]⁺: 359.1601, found: 359.1615; calcd for C₁₉H₂₂N₂O₅ [M+Na]⁺: 381.1421, found: 381.1432; calcd for C₁₉H₂₂N₂O₅ [M+K]⁺: 397.1160, found: 397.1173.

The mixture of meso-(4*S*,5*R*)-4,5-dihydroxy-4,5-diphenyl-1,3-dipropylimidazolidin-2-one **cis-1e** and rac-(4*R*,5*R*)-4,5-dihydroxy-4,5-diphenyl-1,3-dipropylimidazolidin-2-one **trans-1e** (1.78:1). Yield 96% (2.378 g); white powder, mp: 127-129°C; IR (neat, cm⁻¹): 3757, 3716, 3659, 3408, 3370, 3194 (OH), 3093, 3062, 3035 (Ar), 2972, 2937, 2877 (Alk), 1659 (C=O). ¹H NMR: (300 MHz, DMSO-*d*₆) δ 0.71-0.85 (m, 6H, CH₂CH₃(**1e-cis**+**1e-trans**)), 1.48-1.78 (m, 4H, CH₂CH₂CH₃(**1e-cis**+**1e-trans**)), 2.70-2.84 (m, 2H, NCH₂CH₂(**1e-cis**)); 2.85-2.97 (m, 4H, NCH₂CH₂(**1e-trans**)); 2.98-3.12 (m, 2H, NCH₂CH₂(**1e-cis**)); 5.49-6.19 (br.s, 1H, OH(**1e-trans**)), 6.20-6.70 (br.s, 1H, OH(**1e-cis**)), 6.86-7.07 (m, 10H, CH(Ph)(**1e-cis**)), 7.19-7.34 (m, 10H, CH(Ph)(**1e-trans**)). ¹³C NMR (75 MHz, DMSO-*d*₆) δ = 11.49, 11.61 (CH₂CH₃), 22.02, 22.41 (CH₂CH₂CH₃), 42.77, 43.02 (NCH₂CH₂), 92.90, 93.13 (Ph-C-C-Ph), 126.68, 126.97, 127.03, 127.24, 127.50, 128.52, 128.73, 129.44, 129.48 (CH(Ph)), 135.48, 137.80, 138.48 (C(Ph)), 159.80, 160.99 (C=O). HRMS-ESI (m/z): calcd for C₂₁H₂₆N₂O₃ [M+H]⁺: 355.2016, found: 355.2010; calcd for C₂₁H₂₆N₂O₃ [M+Na]⁺: 377.1836, found: 377.1829.

The mixture of meso-(4*S*,5*R*)-4,5-dihydroxy-4,5-bis(4-methoxyphenyl)-1,3-dipropylimidazolidin-2-one **cis-1f** and rac-(4*R*,5*R*)-4,5-dihydroxy-4,5-bis(4-methoxyphenyl)-1,3-dipropylimidazolidin-2-one **trans-1f** (1.3:1). Yield 94% (2.724 g); white powder, mp: 133-135°C; IR (neat, cm⁻¹): 3441, 3412, 3219 (OH), 3076, 3004 (Ar), 2965, 2934, 2910, 2873, 2840 (Alk), 1690, 1675 (C=O). ¹H NMR: (300 MHz, DMSO-*d*₆) δ 0.71-0.81 (m, 6H, CH₂CH₃), 1.43-1.75 (m, 4H, CH₂CH₂CH₃), 2.67-2.8 (m, 1H, NCH₂CH₂); 2.81-2.92 (m, 2H, NCH₂CH₂); 2.93-3.07 (m, 1H, NCH₂CH₂); 3.62 (s, 3H, OMe), 3.75 (s, 3H, OMe), 5.5-5.9 (br.s, 1H, OH), 6.55-6.62 (m, 2H, CH(PMP)), 6.77-6.86 (m, 4H, CH(PMP)), 7.07-7.17 (m, 2H, CH(PMP)). ¹³C NMR (75 MHz, DMSO-*d*₆) δ = 11.73, 11.86 (CH₂CH₃), 22.32, 22.70 (CH₂CH₂CH₃), 42.88, 43.18 (NCH₂CH₂), 55.11 (OMe), 93.06, 93.26, 112.29, 112.64, 128.56, 130.00, 130.76 (CH(PMP), C(PMP)), 158.56, 158.89, 160.09, 161.31 (C=O, C-OMe). HRMS-ESI (m/z): calcd for C₂₃H₃₀N₂O₅ [M+H]⁺: 415.2227, found: 415.2220; calcd for C₂₃H₃₀N₂O₅ [M+Na]⁺: 437.2047, found: 437.2043; calcd for C₂₃H₃₀N₂O₅ [M+K]⁺: 453.1786, found: 453.1779.

The mixture of meso-(4*S*,5*R*)-4,5-bis(4-fluorophenyl)-4,5-dihydroxy-1,3-dimethylimidazolidin-2-one **cis-1g** and rac-(4*R*,5*R*)-4,5-bis(4-fluorophenyl)-4,5-dihydroxy-1,3-dimethylimidazolidin-2-one **trans-1g** (25:1). Yield 96% (2.244 g); colorless crystals, mp: 201-202°C. IR (neat, cm⁻¹): 3458 (OH), 3290, 3078 (Ar), 2977, 2943, 2886 (Alk), 1677 (C=O). ¹H NMR: (300 MHz, DMSO-*d*₆) δ = 2.60 (s, 6H, NMe(**1g-cis**+**1g-trans**)), 6.11 (s, 2H, OH(**1g-trans**)), 6.66 (s, 2H, OH(**1g-cis**)), 6.86-6.93 (m, 8H, CH(C₆H₄F)(**1g-cis**)), 7.12-7.25 (m, 8H, CH(C₆H₄F)(**1g-trans**)). ¹³C NMR (75 MHz, DMSO-*d*₆) δ = 25.56 (NMe(**1g-trans**)), 25.77 (NMe(**1g-cis**)), 92.02 (FC₆H₄-C-C-C₆H₄F), 113.87 (d, C(3)(C₆H₄F(**1g-cis**)), ²J = 20.9 Hz), 114.14 (d, C(3)(C₆H₄F(**1g-trans**)), ²J = 20.6 Hz), 129.02 (d, C(2)(C₆H₄F(**1g-trans**)), ³J = 8.4 Hz), 130.37 (d, C(2)(C₆H₄F(**1g-cis**)), ³J = 8.3 Hz), 133.48 (d, C(1)(C₆H₄F(**1g-cis**)), ⁴J = 2.7 Hz), 134.40 (d, C(1)(C₆H₄F(**1g-trans**)), ⁴J = 2.7 Hz), 159.08 (C=O(**1g-trans**)), 160.35 (C=O(**1g-cis**)), 161.33 (d, C(4)(C₆H₄F(**1g-trans**)), ¹J = 244.1 Hz), 161.86 (d, C(4)(C₆H₄F(**1g-cis**)), ¹J = 243.6 Hz). ¹⁹F NMR: (282 MHz, DMSO-*d*₆) δ = -116.08 (C₆H₄F(**1g-trans**)), -115.91 (C₆H₄F(**1g-cis**)). HRMS-ESI (m/z): calcd for C₁₇H₁₆F₂N₂O₃ [M+H]⁺: 335.1202, found: 335.1191; calcd for C₁₇H₁₆F₂N₂O₃ [M+Na]⁺: 357.1021, found: 357.1011.

The mixture of *meso*-(4*S*,5*R*)-1,3-diethyl-4,5-bis(4-fluorophenyl)-4,5-dihydroxyimidazolidin-2-one *cis*-**1h** and *rac*-(4*R*,5*R*)-1,3-diethyl-4,5-bis(4-fluorophenyl)-4,5-dihydroxyimidazolidin-2-one *trans*-**1h** (9:1). Yield 87% (2.205 g); colorless crystals, mp: 151-152°C. IR (neat, cm⁻¹): 3459 (OH), 3289, 3077 (Ar), 2978, 2945, 2888 (Alk), 1678 (C=O). ¹H NMR: (300 MHz, DMSO-*d*₆) δ = 1.15 (t, 6H, *J* = 7.1 Hz, CH₂CH₃), 2.94 (dq, 2H, *J* = 7.0 Hz, *J* = 14.0 Hz, CH₂CH₃), 3.11 (dq, 2H, *J* = 6.9 Hz, *J* = 14.0 Hz, CH₂CH₃), 6.03; 6.60 (s, 2H, OH), 6.83-7.01 (m, 6H, CH(C₆H₄F)), 7.07-7.19 (m, 1H, CH(C₆H₄F)), 7.20-7.32 (m, 1H, CH(C₆H₄F)). ¹³C NMR (75 MHz, DMSO-*d*₆), δ = 14.74 (CH₂CH₃(**1h-trans**)), 15.05 (CH₂CH₃(**1h-cis**)), 35.41, 35.64 (CH₂CH₃), 92.77 (FC₆H₄-C-C-C₆H₄F(**1h-cis**)), 92.96 (FC₆H₄-C-C-C₆H₄F(**1h-trans**)), 113.56 (d, C(3)(C₆H₄F(**1h-cis**))), ²*J* = 21.2 Hz), 113.95 (d, C(3)(C₆H₄F(**1h-trans**))), ²*J* = 21.5 Hz), 113.76 (d, C(2)(C₆H₄F(**1h-trans**))), ³*J* = 8.4 Hz), 129.15 (d, C(2)(C₆H₄F(**1h-trans**))), ³*J* = 8.4 Hz), 130.63 (d, C(2)(C₆H₄F(**1h-cis**))), ³*J* = 8.3 Hz), 133.85 (d, C(1)(C₆H₄F(**1h-cis**))), ⁴*J* = 2.9 Hz), 135.17 (d, C(1)(C₆H₄F(**1h-trans**))), ⁴*J* = 2.9 Hz), 159.31 (C=O(**1h-trans**)), 160.74 (C=O(**1h-cis**)), 161.33 (d, C(4)(C₆H₄F(**1h-trans**))), ¹*J* = 244.2 Hz), 161.85 (d, C(4)(C₆H₄F(**1h-cis**))), ¹*J* = 243.6 Hz). ¹⁹F NMR: (282 MHz, DMSO-*d*₆) δ = -116.03 (d, C₆H₄F, *J* = 57.1 Hz). HRMS-ESI (*m/z*): calcd for C₁₉H₂₀F₂N₂O₃ [*M*+H]⁺: 363.1515, found: 363.1501; calcd for C₁₉H₂₀F₂N₂O₃ [*M*+Na]⁺: 385.1334, found: 385.1319.

The mixture of *meso*-(4*S*,5*R*)-1,3-diethyl-4,5-dihydroxy-4,5-di-*p*-tolylimidazolidin-2-one *cis*-**1i** and *rac*-(4*R*,5*R*)-1,3-diethyl-4,5-dihydroxy-4,5-di-*p*-tolylimidazolidin-2-one *trans*-**1i** (1:1.3). Yield 89% (2.205 g); white powder, mp: 152-154°C; IR (neat, cm⁻¹): 3455 (OH), 3292, 3074 (Ar), 2967, 2944, 2886 (Alk), 1675 (C=O). ¹H NMR: (300 MHz, DMSO-*d*₆) δ 1.09-1.23 (m, 6H, (CH₂CH₃)), 2.14 (s, 3H, Me(*p*-Tol), **1i-cis**), 2.32 (s, 3H, Me(*p*-Tol), **1i-trans**), 2.82-3.18 (m, 4H, CH₂CH₃), 5.61-5.82 (br. s, 2H, OH, **1i-trans**), 6.22-6.48 (br. s, 2H, OH, **1i-cis**), 6.79 – 6.90 (m, 8H, CH(*p*-Tol), **1i-cis**), 7.05 – 7.19 (m, 8H, CH(*p*-Tol), **1i-trans**). ¹³C NMR (75 MHz, DMSO-*d*₆), δ = 14.85 (CH₂CH₃, **1i-trans**), 15.20 (CH₂CH₃, **1i-cis**), 20.60 (CH₃(*p*-Tol), **1i-cis**), 20.76 (CH₃(*p*-Tol), **1i-trans**), 35.50 (CH₂CH₃, **1i-cis**), 35.74 (CH₂CH₃, **1i-trans**), 93.15 (*p*-Tol-C-C-*p*-Tol, **1i-cis**), 93.47 (*p*-Tol-C-C-*p*-Tol, **1i-trans**), 127.18, 127.49, 127.72, 128.63 (CH(*p*-Tol)), 135.00, 135.88, 136.35, 136.68 (C(*p*-Tol)), 159.71 (C=O, **1i-cis**), 161.11 (C=O, **1i-trans**). HRMS-ESI (*m/z*): calcd for C₂₁H₂₆N₂O₃ [*M*+H]⁺: 355.2016, found: 355.2014; calcd for C₂₁H₂₆N₂O₃ [*M*+Na]⁺: 377.1836, found: 377.1835; calcd for C₂₁H₂₆N₂O₃ [*M*+K]⁺: 393.1575, found: 393.1571.

1,3-Diethyl-4,5-diphenyl-1*H*-imidazol-2(3*H*)-one **4b**. Yield 53% (7.738 g); white powder, (R_f(CHCl₃:MeCN - 5:1) = 0.275, mp: 139-140°C; IR (neat, cm⁻¹): 3058, 3036 (Ar), 2976, 2953, 2935, 2876 (Alk), 1678 (C=O). ¹H NMR: (300 MHz, DMSO-*d*₆) δ 0.99 (t, 6H, *J* = 7.1 Hz, (CH₂CH₃)), 3.61 (q, 4H, *J* = 7.1 Hz, CH₂CH₃), 7.23 – 7.29 (m, 10H, Ph), 7.30 – 7.40 (m, 10H, Ph). ¹³C NMR (75 MHz, DMSO-*d*₆), δ = 14.45 (CH₂CH₃), 35.92 (CH₂CH₃), 120.09 (Ph-C=C-Ph), 127.99, 128.46, 129.00, 130.11 (CH(Ph), C(Ph)), 152.07 (C=O). HRMS-ESI (*m/z*): calcd for C₁₉H₂₀N₂O [*M*+H]⁺: 293.1648, found: 293.1639; calcd for C₁₉H₂₀N₂O [*M*+Na]⁺: 315.1468, found: 315.1462.

1,3-Diethyl-4,5-bis(4-methoxyphenyl)-1H-imidazol-2(3H)-one 4c. Yield 48% (8.448 g); colorless crystals; $R_f(\text{CHCl}_3:\text{MeCN} - 5:1) = 0.27$, mp: 131-133°C; IR (neat, cm^{-1}): 3071, 3041, 3000 (Ar), 2978, 2964, 2938, 2874, 2840 (Alk), 1680 (C=O); ^1H NMR: (300 MHz, $\text{DMSO}-d_6$) δ 0.99 (t, 6H, $^3J = 7.0$ Hz, $\text{Me}(\text{Et})$), 3.56 (q, 4H, $^3J = 7.0$ Hz, $\text{CH}_2(\text{Et})$), 3.75 (s, 6H, OMe), 6.91 (d, 2H, $^3J = 8.4$ Hz, $\text{CH}(\text{PMP})$), 7.19 (d, 2H, $^3J = 8.5$ Hz, $\text{CH}(\text{PMP})$). ^{13}C NMR (75 MHz, $\text{DMSO}-d_6$), δ = 14.51 ($\text{Me}(\text{Et})$), 35.74 ($\text{CH}_2(\text{Et})$), 54.97 (OMe), 113.88, 119.46, 121.19, 131.49 ($\text{CH}(\text{PMP})$), 151.88, 158.79. HRMS-ESI (m/z): calcd for $\text{C}_{21}\text{H}_{24}\text{N}_2\text{O}_3$ $[\text{M}+\text{H}]^+$: 353.1860, found: 353.1863; calcd for $\text{C}_{21}\text{H}_{24}\text{N}_2\text{O}_3$ $[\text{M}+\text{Na}]^+$: 375.1679, found: 375.1675; calcd for $\text{C}_{21}\text{H}_{24}\text{N}_2\text{O}_3$ $[\text{M}+\text{K}]^+$: 391.1419, found: 391.1415.

4,5-Bis(4-methoxyphenyl)-1,3-dimethyl-1H-imidazol-2(3H)-one 4d. Yield 67% (10.854 g); white powder, mp: 158-159°C; IR (neat, cm^{-1}): 3069, 3041, 3008 (Ar), 2980, 2956, 2834 (Alk), 1683 (C=O); ^1H NMR (300 MHz, $\text{DMSO}-d_6$) δ 3.07 (s, 6H, NMe), 3.75 (s, 6H, OMe), 6.91 (d, 2H, $^3J = 8.6$ Hz, $\text{CH}(\text{PMP})$), 7.19 (d, 2H, $^3J = 8.6$ Hz, $\text{CH}(\text{PMP})$). ^{13}C NMR (75 MHz, $\text{DMSO}-d_6$), δ = 28.26, 55.00, 113.93, 119.69, 121.08, 131.29, 152.72, 158.76. HRMS-ESI (m/z): calcd for $\text{C}_{19}\text{H}_{20}\text{N}_2\text{O}_3$ $[\text{M}+\text{H}]^+$: 325.1547, found: 325.1541; calcd for $\text{C}_{19}\text{H}_{20}\text{N}_2\text{O}_3$ $[\text{M}+\text{Na}]^+$: 347.1366, found: 347.1358; calcd for $\text{C}_{19}\text{H}_{20}\text{N}_2\text{O}_3$ $[\text{M}+\text{K}]^+$: 363.1106, found: 363.1098.

4,5-Diphenyl-1,3-dipropyl-1H-imidazol-2(3H)-one 4e. Yield 54% (8.640 g); white powder, mp: 91- 93°C; IR (neat, cm^{-1}): 3692, 3342, 3273, 3056 (Ar), 2968, 2933, 2876 (Alk), 1681 (C=O). ^1H NMR: (300 MHz, $\text{DMSO}-d_6$) δ 0.67 (t, 6H, $J = 8.4$ Hz, CH_2CH_3), 1.37 – 1.67 (m, 4H, $\text{CH}_2\text{CH}_2\text{CH}_3$), 3.55 (t, 4H, $J = 7.3$ Hz, NCH_2CH_2), 7.21 – 7.27 (m, 10H, Ph), 7.29 – 7.37 (m, 10H, Ph). ^{13}C NMR (75 MHz, $\text{DMSO}-d_6$), δ = 10.91 (CH_2CH_3), 22.08 ($\text{CH}_2\text{CH}_2\text{CH}_3$), 42.57 (NCH_2CH_2), 120.49 ($\text{Ph}-\text{C}=\text{C}-\text{Ph}$), 128.14, 128.57, 129.18, 130.26 (C(Ph), CH(Ph)), 152.81 (C=O). HRMS-ESI (m/z): calcd for $\text{C}_{21}\text{H}_{24}\text{N}_2\text{O}$ $[\text{M}+\text{H}]^+$: 321.1961, found: 321.1965; calcd for $\text{C}_{21}\text{H}_{24}\text{N}_2\text{O}$ $[\text{M}+\text{Na}]^+$: 343.1781, found: 343.1778; calcd for $\text{C}_{21}\text{H}_{24}\text{N}_2\text{O}$ $[\text{M}+\text{K}]^+$: 359.1520, found: 359.1517.

4,5-Bis(4-methoxyphenyl)-1,3-dipropyl-1H-imidazol-2(3H)-one 4f. Yield 50% (9.503 g); white powder, mp: 93-95°C; IR (neat, cm^{-1}): 3692, 3339, 3064, 3039 (Ar), 3011, 2971, 2934, 2874, 2835 (Alk), 1678 (C=O). ^1H NMR: (300 MHz, $\text{DMSO}-d_6$) δ 0.68 (t, 6H, $J = 7.3$ Hz, CH_2CH_3); 1.28 – 1.42 (m, 4H, $\text{CH}_2\text{CH}_2\text{CH}_3$); 3.50 (t, 4H, $J = 7.6$ Hz, NCH_2CH_2); 3.73 (s, 6H, OMe); 6.89 (d, 4H, $J = 8.4$ Hz, $\text{CH}(4\text{-MeOC}_6\text{H}_4)$); 7.17 (d, 4H, $J = 8.4$ Hz, $\text{CH}(\text{PMP})$). ^{13}C NMR (75 MHz, $\text{DMSO}-d_6$), δ = 10.95 (CH_2CH_3), 22.13 ($\text{CH}_2\text{CH}_2\text{CH}_3$), 48.91 (NCH_2CH_2), 55.11 (OMe), 113.98, 131.68 ($\text{CH}(\text{PMP})$), 119.90, 121.38 (C(PMP), $\text{PMP}-\text{C}=\text{C}-\text{PMP}$), 152.66 (C=O), 158.90 ($\text{C}-\text{OMe}$). HRMS-ESI (m/z): calcd for $\text{C}_{23}\text{H}_{28}\text{N}_2\text{O}_3$ $[\text{M}+\text{H}]^+$: 381.2173, found: 381.2165; calcd for $\text{C}_{23}\text{H}_{28}\text{N}_2\text{O}_3$ $[\text{M}+\text{Na}]^+$: 403.1992, found: 403.1985; calcd for $\text{C}_{23}\text{H}_{28}\text{N}_2\text{O}_3$ $[\text{M}+\text{K}]^+$: 419.1732, found: 419.1728.

4,5-Bis(4-fluorophenyl)-1,3-dimethyl-1H-imidazol-2(3H)-one 4g: Yield 54% (8.096 g); white powder, mp: 142-143°C (Me_2CO). IR (neat, cm^{-1}): 3511, 3457, 3060 (Ar), 2943, 2891 (Alk), 1689, 1673 (C=O). ^1H NMR: (300 MHz, $\text{DMSO}-d_6$) δ = 3.09 (s, 6H, NMe), 7.10-7.24 (m, 4H, CH), 7.25-7.37 (m, 4H, CH). ^{13}C NMR (75 MHz, $\text{DMSO}-d_6$), δ = 28.31, 115.56 (d, C(3)($\text{C}_6\text{H}_4\text{F}$), $^2J = 21.7$ Hz), 119.57 ($\text{FC}_6\text{H}_4-\text{C}=\text{C}-\text{C}_6\text{H}_4\text{F}$), 125.09 (d, C(1)($\text{C}_6\text{H}_4\text{F}$), $^4J = 3.2$ Hz), 132.17 (d, C(2)($\text{C}_6\text{H}_4\text{F}$), $^3J = 8.4$ Hz), 152.73 (C=O), 161.71 (d, C(4)($\text{C}_6\text{H}_4\text{F}$), $^1J = 245.9$ Hz). ^{19}F NMR: (282 MHz, $\text{DMSO}-d_6$) δ = -114.13 ($\text{C}_6\text{H}_4\text{F}$). HRMS-ESI (m/z): calcd for $\text{C}_{17}\text{H}_{14}\text{F}_2\text{N}_2\text{O}$ $[\text{M}+\text{H}]^+$: 301.1147, found: 301.1148.

1,3-Diethyl-4,5-bis(4-fluorophenyl)-1H-imidazol-2(3H)-one 4h. Yield 51% (8.363 g); colorless crystals, mp: 109-110°C (Me₂CO). IR (neat, cm⁻¹): 3504, 3440, 3062 (Ar), 2981, 2939, 2876 (Alk), 1664(C=O). ¹H NMR: (300 MHz, DMSO-*d*₆) δ = 0.99 (t, 6H, *J* = 7.0 Hz, CH₂CH₃), 3.59 (q, 4H, *J* = 7.1 Hz, CH₂CH₃), 7.10-7.24 (m, 4H, CH), 7.25-7.39 (m, 4H, CH). ¹³C NMR (75 MHz, DMSO-*d*₆), δ = 14.46 (CH₂CH₃), 35.89 (CH₂CH₃), 115.56 (d, C(3)(C₆H₄F), ²*J* = 21.6 Hz), 119.27 (FC₆H₄-C=C-C₆H₄F), 125.24 (d, C(1)(C₆H₄F), ⁴*J* = 3.2 Hz), 132.38 (d, C(2)(C₆H₄F), ³*J* = 8.4 Hz), 151.90 (C=O), 161.76 (d, C(4)(C₆H₄F), ¹*J* = 245.9 Hz) ¹⁹F NMR: (282 MHz, DMSO-*d*₆) δ = -114.01 (C₆H₄F). HRMS-ESI (m/z): calcd for C₁₉H₁₈F₂N₂O [M+H]⁺: 329.1460, found: 329.1456; calcd for C₁₉H₁₈F₂N₂O [M+Na]⁺: 351.1279, found: 351.1278.

1,3-Diethyl-4,5-di-*p*-tolyl-1H-imidazol-2(3H)-one 4i. Yield 57% (9.119 g); colorless crystals, mp: 112-114°C (Me₂CO). IR (neat, cm⁻¹): 3347, 3027, 2974, 2939, 2921, 2876, 1683(C=O). ¹H NMR (300 MHz, DMSO-*d*₆): δ (ppm) 0.98 (t, 6H, ³*J* = 7.2 Hz, CH₂CH₃), 2.29 (s, 6H, Me), 3.57 (q, 4H, ³*J* = 7.0 Hz, CH₂CH₃), 7.15-7.23 (m, 4H, CH), 7.09-7.19 (m, 8H, CH). ¹³C NMR (75 MHz, DMSO-*d*₆), δ = 14.52 (CH₂CH₃), 20.71 (Me), 35.82 (CH₂CH₃), 119.81 (p-Tol-C=C-p-Tol), 126.12, 129.08, 129.99, 137.30 (CH(p-Tol), C(p-Tol)), 152.00 (C=O). HRMS-ESI (m/z): calcd for C₂₁H₂₄N₂O [M+H]⁺: 321.1961, found: 321.1962; calcd for C₂₁H₂₄N₂O [M+Na]⁺: 343.1781, found: 343.1780.

1,3-Diethyl-1H-phenanthro[9,10-*d*]imidazol-2(3H)-one 5a. Yield 22% (0.241 g); light beige powder, R_f(CHCl₃:MeCN - 5:1) = 0.65, mp: 227-228°C (DMSO). IR (neat, cm⁻¹): 3100, 3059, 3040, 2994, 2969, 2930, 2899, 2869 (Ar), 1682(C=O). ¹H NMR (300 MHz, DMSO-*d*₆): δ (ppm) 1.39 (t, 6H, ³*J* = 7.0 Hz, CH₂CH₃), 4.47 (q, 4H, ³*J* = 7.0 Hz, CH₂CH₃), 7.59-7.69 (m, 2H, CH), 7.70-7.81 (m, 2H, CH), 8.37 (d, 2H, *J* = 8.3 Hz, CH), 8.97 (d, 2H, *J* = 8.3 Hz, CH). ¹³C NMR (75 MHz, DMSO-*d*₆), δ = 14.68 (CH₂CH₃), 37.87 (CH₂CH₃), 119.38, 120.53, 120.76, 124.06, 124.47, 126.69, 127.03, 127.38 (CH), 153.93 (C=O). HRMS-ESI (m/z): calcd for C₁₉H₁₈N₂O [M+H]⁺: 291.1492, found: 291.1498; calcd for C₁₉H₁₈N₂O [M+Na]⁺: 313.1311, found: 313.1314.

1,3-Diethyl-6,9-dimethoxy-1H-phenanthro[9,10-*d*]imidazol-2(3H)-one 5b. Yield 46% (0.610 g); light olivetone powder, R_f(CHCl₃:MeCN - 5:1) = 0.54, mp: 225-226°C (DMSO). IR (neat, cm⁻¹): 3345, 3028, 2975, 2940, 2922, 2877(Ar), 1685(C=O). ¹H NMR (300 MHz, DMSO-*d*₆): δ (ppm) 1.35 (t, 6H, ³*J* = 7.0 Hz, CH₂CH₃), 4.01 (s, 6H, OMe), 4.37 (q, 4H, ³*J* = 6.9 Hz, CH₂CH₃), 7.34-7.43 (m, 2H, CH), 8.16-8.27 (m, 2H, CH). ¹³C NMR (75 MHz, DMSO-*d*₆), δ = 14.64 (CH₂CH₃), 37.64 (CH₂CH₃), 55.41 (OMe), 106.27, 115.79, 117.12, 117.82, 122.02, 127.70, 153.77 (CH), 155.98 (C=O). HRMS-ESI (m/z): calcd for C₂₁H₂₂N₂O₃ [M+H]⁺: 351.1703, found: 351.1698; calcd for C₂₁H₂₂N₂O₃ [M+Na]⁺: 373.1523, found: 373.1524.

***N,N'*-Carbonylbis(*N*-ethylbenzamide) 6.** Yield 5% (0.061 g); colorless crystals, R_f(CHCl₃:MeCN - 5:1) = 0.92, mp: 150-151°C (CHCl₃:MeCN). IR (neat, cm⁻¹): 3105, 3087, 3059, 3037, 3008, 2949, 2977, 2934, 2873, 2854 (Ar), 1676 (C=O), 1650 (C=O). ¹H NMR (300 MHz, DMSO-*d*₆): δ (ppm) 1.08 (t, 6H, ³*J* = 7.0 Hz, CH₂CH₃), 3.30 (q, 4H, ³*J* = 7.1 Hz, CH₂CH₃), 7.29-7.42 (m, 4H, CH(Bz)), 7.43-7.55 (m, 4H, CH(Bz)), 7.56-7.66 (m, 2H, CH(Bz)). ¹³C NMR (75 MHz, DMSO-*d*₆), δ = 13.06 (CH₂CH₃), 42.20 (CH₂CH₃), 127.32, 128.62, 132.02, 135.32, (CH(Bz)), 157.88, 170.18 (C=O). HRMS-ESI (m/z): calcd for C₁₉H₂₀N₂O₃ [M+H]⁺: 325.1547, found: 325.1551; calcd for C₁₉H₂₀N₂O₃ [M+nNH₄]⁺: 342.1812, found: 342.1816; calcd for C₁₉H₂₀N₂O₃ [M+Na]⁺: 347.1366, found: 347.1370; calcd for C₁₉H₂₀N₂O₃ [M+K]⁺: 363.1106, found: 363.1106.

NMR spectra of compounds 1b-i, 4b-i, 5a,b, 6

































