

Electrocatalytic multicomponent assembling of aldehydes, *N*-alkyl barbiturates and malononitrile: an efficient approach to pyrano[2,3-*d*]pyrimidines

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7-Amino-1,3-dimethyl-2,4-dioxo-5-phenyl-1,3,4,5-tetrahydro-2H-pyrano[2,3-d]pyrimidine-6-carbonitrile **3a**

White solid; yield 1.10 g (71%); mp 219–222 °C (lit.¹ mp 210 °C).

¹H NMR (300 MHz, [D₆]DMSO): δ = 3.08 (s, 3H, CH₃), 3.36 (s, 3H, CH₃), 4.32 (s, 1H, CH), 7.15–7.39 (m, 7H, Ar, NH₂) ppm.

¹³C NMR (75 MHz, [D₆]DMSO): δ = 27.7 (CH₃), 29.1 (CH₃), 36.6 (CH), 58.7 (C), 88.9 (C), 119.1 (CN), 126.8 (CH), 127.4 (2CH), 128.3 (2CH), 144.2 (C), 150.0 (C), 151.2 (C), 157.7 (C), 160.5 (C) ppm.

7-Amino-1,3-dimethyl-5-(4-methylphenyl)-2,4-dioxo-1,3,4,5-tetrahydro-2H-pyrano[2,3-d]pyrimidine-6-carbonitrile **3b**

White solid; yield 1.21 g (75%); mp 203 °C.

¹H NMR (300 MHz, [D₆]DMSO): δ = 2.25 (s, 3H, CH₃), 3.08 (s, 3H, CH₃), 3.35 (s, 3H, CH₃), 4.28 (s, 1H, CH), 7.05–7.15 (m, 4H, Ar), 7.28 (s, 2H, NH₂) ppm.

¹³C NMR (75 MHz, [D₆]DMSO): δ = 20.6 (CH₃), 27.6 (CH₃), 29.1 (CH₃), 36.2 (CH), 58.8 (C), 89.0 (C), 119.1 (CN), 127.3 (2CH), 128.8 (2CH), 135.9 (C), 141.2 (C), 150.0 (C), 151.0 (C), 157.6 (C), 160.5 (C) ppm.

IR (KBr): ν = 3428, 3300, 3184, 2192, 1700, 1684, 1636, 1496, 1384, 1232, 1188 cm⁻¹.

MS (*m/z*, relative intensity %): 324 (15) [M]⁺, 258 (65), 257 (100), 243 (56), 200 (51), 168 (69), 115 (65), 66 (60).

Found (%): C, 62.75; H, 4.89; N, 17.01. Calc. for C₁₇H₁₆N₄O₃ (%): C, 62.95; H, 4.97; N, 17.27.

7-Amino-5-(4-methoxyphenyl)-1,3-dimethyl-2,4-dioxo-1,3,4,5-tetrahydro-2H-pyrano[2,3-d]pyrimidine-6-carbonitrile **3c**

White solid; yield 1.24 g (73%); mp 222–225 °C (lit.² mp 225–227 °C).

¹H NMR (300 MHz, CDCl₃+ [D₆]DMSO): δ = 3.15 (s, 3H, CH₃), 3.39 (s, 3H, CH₃), 3.68 (s, 3H, OCH₃), 4.36 (s, 1H, CH), 6.32 (s, 2H, NH₂), 6.74 (d, *J* = 8.4 Hz, 2H, Ar), 7.12 (d, *J* = 8.4 Hz, 2H, Ar) ppm.

^{13}C NMR (75 MHz, $\text{CDCl}_3 + [\text{D}_6]\text{DMSO}$): $\delta = 27.6$ (CH_3), 29.0 (CH_3), 35.7 (CH), 55.0 (OCH_3), 58.9 (C), 89.1 (C), 113.6 (2CH), 119.1 (CN), 128.4 (2CH), 136.2 (C), 149.9 (C), 150.8 (C), 157.5 (C), 158.1 (C), 160.4 (C) ppm.

7-Amino-5-(4-chlorophenyl)-1,3-dimethyl-2,4-dioxo-1,3,4,5-tetrahydro-2H-pyrano[2,3-d]pyrimidine-6-carbonitrile 3d

White solid; yield 1.40 g (81%); mp 238–240 °C (lit.³ mp 241–242 °C).

^1H NMR (300 MHz, $[\text{D}_6]\text{DMSO}$): $\delta = 3.08$ (s, 3H, CH_3), 3.35 (s, 3H, CH_3), 4.35 (s, 1H, CH), 7.18–7.47 (m, 6H, Ar, NH_2) ppm.

^{13}C NMR (75 MHz, $[\text{D}_6]\text{DMSO}$): $\delta = 27.7$ (CH_3), 29.1 (CH_3), 36.0 (CH), 58.2 (C), 88.4 (C), 118.9 (CN), 128.2 (2CH), 129.3 (2CH), 131.4 (C), 143.2 (C), 150.0 (C), 151.2 (C), 157.6 (C), 160.5 (C) ppm.

7-Amino-5-(2-chlorophenyl)-1,3-dimethyl-2,4-dioxo-1,3,4,5-tetrahydro-2H-pyrano[2,3-d]pyrimidine-6-carbonitrile 3e

White solid; yield 1.36 g (79%); mp 237–238 °C.

^1H NMR (300 MHz, $[\text{D}_6]\text{DMSO}$): $\delta = 3.07$ (s, 3H, CH_3), 3.37 (s, 3H, CH_3), 4.87 (s, 1H, CH), 7.16–7.42 (m, 6H, Ar, NH_2) ppm.

^{13}C NMR (75 MHz, $[\text{D}_6]\text{DMSO}$): $\delta = 27.6$ (CH_3), 29.1 (CH_3), 33.6 (CH), 57.3 (C), 88.0 (C), 118.5 (CN), 127.5 (CH), 128.4 (CH), 129.3 (CH), 130.2 (CH), 132.2 (C), 141.1 (C), 150.0 (C), 151.5 (C), 157.8 (C), 160.3 (C) ppm.

IR (KBr): $\nu = 3395, 3313, 3193, 2195, 1708, 1688, 1642, 1490, 1390, 1223 \text{ cm}^{-1}$.

MS (m/z , relative intensity %): 346 (0.3) $[\text{M}]^+$, 344 (1) $[\text{M}]^+$, 309 (17), 243 (100), 186 (61), 136 (18), 101 (22), 75 (26), 66 (35).

Found (%): C, 55.45; H, 3.65; Cl, 10.03; N, 16.08. Calc. for $\text{C}_{16}\text{H}_{13}\text{ClN}_4\text{O}_3$ (%): C, 55.74; H, 3.80; Cl, 10.28; N, 16.25.

7-Amino-5-(3-bromophenyl)-1,3-dimethyl-2,4-dioxo-1,3,4,5-tetrahydro-2H-pyrano[2,3-d]pyrimidine-6-carbonitrile 3f

White solid; yield 1.46 g (75%); mp 217 °C.

^1H NMR (300 MHz, $[\text{D}_6]\text{DMSO}$): $\delta = 3.16$ (s, 3H, CH_3), 3.40 (s, 3H, CH_3), 4.38 (s, 1H, CH), 6.47 (s, 2H, NH_2), 7.11 (t, $J = 7.3$ Hz, 1H, Ar), 7.19 (d, $J = 7.3$ Hz, 1H, Ar), 7.27 (d, $J = 8.8$ Hz, 1H, Ar), 7.44 (s, 1H, Ar) ppm.

^{13}C NMR (75 MHz, $[\text{D}_6]\text{DMSO}$): $\delta = 27.7$ (CH_3), 29.2 (CH_3), 36.3 (CH), 58.1 (C), 88.0 (C), 118.9 (CN), 121.6 (C), 126.7 (CH), 129.8 (CH), 130.0 (CH), 130.4 (CH), 146.9 (C), 150.0 (C), 151.3 (C), 157.7 (C), 160.5 (C) ppm.

IR (KBr): $\nu = 3380, 3200, 2204, 1716, 1688, 1636, 1492, 1392, 1228, 1184 \text{ cm}^{-1}$.

MS (*m/z*, relative intensity %): 390 (3) [M]⁺, 388 (3) [M]⁺, 323 (100), 321 (100), 233 (23), 156 (41), 153 (37), 101 (56), 66 (42).

Found (%): C, 49.17; H, 3.25; Br, 20.28; N, 14.22. Calc. for C₁₆H₁₃BrN₄O₃ (%): C, 49.38; H, 3.37; Br, 20.53; N, 14.40.

7-Amino-5-(4-fluorophenyl)-1,3-dimethyl-2,4-dioxo-1,3,4,5-tetrahydro-2H-pyrano[2,3-d]pyrimidine-6-carbonitrile 3g

White solid; yield 1.26 g (77%); mp 230–232 °C.

¹H NMR (300 MHz, [D₆]DMSO): δ = 3.13 (s, 3H, CH₃), 3.39 (s, 3H, CH₃), 4.38 (s, 1H, CH), 6.69 (s, 2H, NH₂), 6.90 (t, *J* = 8.7 Hz, 2H, Ar), 7.19 (dd, *J* = 8.1, 5.1 Hz, 2H, Ar) ppm.

¹³C NMR (75 MHz, [D₆]DMSO): δ = 27.7 (CH₃), 29.1 (CH₃), 35.9 (CH), 58.5 (C), 88.6 (C), 114.9 (d, ²*J*_{C-F} = 21.1 Hz, 2CH), 119.0 (CN), 129.3 (d, ³*J*_{C-F} = 8.3 Hz, 2CH), 140.3 (d, ⁴*J*_{C-F} = 3.0 Hz, C), 150.0 (C), 151.1 (C), 157.6 (C), 160.5 (C), 161.1 (d, ¹*J*_{C-F} = 243.0 Hz, C) ppm.

IR (KBr): ν = 3380, 3308, 3192, 2200, 1708, 1684, 1640, 1492, 1388, 1224 cm⁻¹.

MS (*m/z*, relative intensity %): 328 (20) [M]⁺, 262 (85), 233 (28), 204 (22), 176 (32), 120 (40), 101 (36), 66 (100), 57 (42).

Found (%): C, 58.32; H, 3.85; F, 5.53; N, 16.92. Calc. for C₁₆H₁₃FN₄O₃ (%): C, 58.54; H, 3.99; F, 5.79; N, 17.07.

7-Amino-1,3-dimethyl-5-(4-nitrophenyl)-2,4-dioxo-1,3,4,5-tetrahydro-2H-pyrano[2,3-d]pyrimidine-6-carbonitrile 3h

White solid; yield 1.30 g (73%); mp 212–214 °C.

¹H NMR (300 MHz, [D₆]DMSO): δ = 3.06 (s, 3H, CH₃), 3.35 (s, 3H, CH₃), 4.51 (s, 1H, CH), 7.49 (s, 2H, NH₂), 7.55 (d, *J* = 8.1 Hz, 2H, Ar), 8.15 (d, *J* = 8.1 Hz, 2H, Ar) ppm.

¹³C NMR (75 MHz, [D₆]DMSO): δ = 27.4 (CH₃), 28.9 (CH₃), 36.2 (CH), 57.1 (C), 87.5 (C), 118.4 (CN), 123.3 (2CH), 128.6 (2CH), 146.2 (C), 149.7 (C), 151.2 (C), 151.5 (C), 157.5 (C), 160.2 (C) ppm.

IR (KBr): ν = 3384, 3308, 3196, 2204, 1690, 1684, 1628, 1520, 1488, 1384, 1232, 1188 cm⁻¹.

MS (*m/z*, relative intensity %): 355 (7) [M]⁺, 290 (20), 288 (32), 242 (28), 199 (35), 175 (52), 156 (60), 101 (59), 75 (67), 66 (100).

Found (%): C, 53.87; H, 3.50; N, 19.63. Calc. for C₁₆H₁₃N₅O₅ (%): C, 54.09; H, 3.69; N, 19.71.

7-Amino-5-(2-chlorophenyl)-1,3-diethyl-2,4-dioxo-1,3,4,5-tetrahydro-2H-pyrano[2,3-d]pyrimidine-6-carbonitrile 3i

White solid; yield 1.41 g (76%); mp 186–187 °C.

^1H NMR (300 MHz, $[\text{D}_6]\text{DMSO}$): δ = 1.00 (t, J = 6.6 Hz, 3H, CH_3), 1.23 (t, J = 6.6 Hz, 3H, CH_3), 3.71 (q, J = 6.6 Hz, 2H, CH_2), 3.98 (q, J = 6.6 Hz, 2H, CH_2), 4.84 (s, 1H, CH), 7.15-7.44 (m, 6H, Ar, NH_2) ppm.

^{13}C NMR (75 MHz, $[\text{D}_6]\text{DMSO}$): δ = 12.6 (CH_3), 13.9 (CH_3), 33.9 (CH), 35.8 (CH_2), 37.8 (CH_2), 57.1 (C), 88.3 (C), 118.5 (CN), 127.5 (CH), 128.4 (CH), 129.5 (CH), 130.4 (CH), 132.2 (C), 140.8 (C), 149.1 (C), 151.3 (C), 157.9 (C), 159.9 (C) ppm.

IR (KBr): ν = 3413, 3331, 3148, 2203, 1698, 1632, 1488, 1388, 1246, 1179 cm^{-1} .

MS (m/z , relative intensity %): 374 (0.5) $[\text{M}]^+$, 372 (1.5) $[\text{M}]^+$, 337 (26), 271 (58), 172 (53), 136 (28), 101 (43), 70 (86), 66 (91), 44 (100).

Found (%): C, 57.73; H, 4.42; Cl, 9.37; N, 14.82. Calc. for $\text{C}_{18}\text{H}_{17}\text{ClN}_4\text{O}_3$ (%): C, 57.99; H, 4.60; Cl, 9.51; N, 15.03.

7-Amino-1,3-diethyl-5-(4-fluorophenyl)-2,4-dioxo-1,3,4,5-tetrahydro-2H-pyrano[2,3-d]pyrimidine-6-carbonitrile 3j

White solid; yield 1.30 g (73%); mp 160–162 $^\circ\text{C}$.

^1H NMR (300 MHz, $[\text{D}_6]\text{DMSO}$): δ = 1.01 (t, J = 6.6 Hz, 3H, CH_3), 1.22 (t, J = 6.6 Hz, 3H, CH_3), 3.65-3.83 (m, 2H, CH_2), 3.88-4.05 (m, 2H, CH_2), 4.35 (s, 1H, CH), 7.10 (t, J = 8.8 Hz, 2H, Ar), 7.21-7.31 (m, 2H, Ar) 7.34 (s, 2 H, NH_2) ppm.

^{13}C NMR (75 MHz, $[\text{D}_6]\text{DMSO}$): δ = 12.7 (CH_3), 13.8 (CH_3), 35.8 (CH), 35.9 (CH_2), 58.4 (C), 89.0 (C), 115.0 (d, $^2J_{\text{C-F}}$ = 21.1 Hz, 2CH), 118.9 (CN), 129.2 (d, $^3J_{\text{C-F}}$ = 8.2 Hz, 2CH), 140.2 (d, $^4J_{\text{C-F}}$ = 3.0 Hz, C), 149.1 (C), 150.8 (C), 157.7 (C), 160.0 (C), 161.1 (d, $^1J_{\text{C-F}}$ = 241.2 Hz, C) ppm.

IR (KBr): ν = 3384, 3304, 3192, 2206, 1687, 1642, 1487, 1386, 1243, 1178 cm^{-1} .

MS (m/z , relative intensity %): 356 (25) $[\text{M}]^+$, 290 (33), 261 (82), 190 (24), 162 (32), 120 (21), 70 (47), 66 (58), 44 (100).

Found (%): C, 60.39; H, 4.68; F, 5.18; N, 15.56. Calc. for $\text{C}_{18}\text{H}_{17}\text{FN}_4\text{O}_3$ (%): C, 60.67; H, 4.81; F, 5.33; N, 15.72.

References

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