

## Catalyst-free three-component synthesis of hydroxyalkyltriazolylmethylidene barbiturates

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*General procedure for the synthesis of compounds 3a-h.* A 10 ml round bottom flask was charged with water (2 ml) and 4-hydroxyalkynal **1** (1 mmol), trimethylsilyl azide (0.138 g, 1.2 mmol) was then added, and the mixture was stirred for 18 h at room temperature. Then a solution of barbituric acid **2** (1 mmol) in ethanol (4 ml) was added, and the mixture was stirred for 20 h at room temperature. The precipitate formed was collected on a filter, washed with water and cold methanol and dried at reduced pressure.

*5-[[4-(1-Hydroxy-1-methylethyl)-1H-1,2,3-triazol-5-yl]methylidene]pyrimidine-2,4,6(1H,3H,5H)-trione (3a):* yield 220 mg, 82%. Colorless powder, mp >260 °C (decomp.). <sup>1</sup>H NMR (400.13 MHz, DMSO-*d*<sub>6</sub>): δ 15.57 (br s, 1H, NH), 11.67 [br s, 1H, NH(C=O)], 11.50 [br s, 1H, NH(C=O)], 9.11 (s, 1H, HC=), 5.62 (br s, 1H, OH), 1.58 [s, 6H, (CH<sub>3</sub>)<sub>2</sub>]. <sup>13</sup>C NMR (101.62 MHz, DMSO-*d*<sub>6</sub>): δ 169.9 (C-8), 163.8 (C-12), 162.5 (CH=), 159.6 (C-4), 149.7(C-10), 136.8 (C-5), 116.3 (C-7), 69.7 (C-OH), 31.6 (CH<sub>3</sub>). IR (KBr): 3476, 3415, 3196, 3086, 2983, 2850, 2789, 1742, 1674, 1598, 1517, 1410, 1379, 1315, 1223, 1172, 1122, 1092, 1069, 994, 959, 857, 798, 753, 681, 649, 612, 564, 513, 481, 421 cm<sup>-1</sup>. Found (%): C, 45.39; H, 4.03; N, 26.34. Calcd for C<sub>10</sub>H<sub>11</sub>N<sub>5</sub>O<sub>4</sub>: C, 45.29; H, 4.18; N, 26.41.

*5-[[4-(1-Hydroxy-1-methylpropyl)-1H-1,2,3-triazol-5-yl]methylidene]pyrimidine-2,4,6(1H,3H,5H)-trione (3b):* yield 240 mg, 84%. Colorless powder, mp >260 °C (decomp.). <sup>1</sup>H NMR (400.13 MHz, DMSO-*d*<sub>6</sub>): δ 15.60 (br s, 1H, NH), 11.62 [br s, 1H, NH(C=O)], 11.50 [br s, 1H, NH(C=O)], 9.05 (s, 1H, HC=), 5.51 (br s, 1H, OH), 1.90–1.72 (m, 2H, CH<sub>2</sub>), 1.56 (s, 3H, CH<sub>3</sub>), 0.73 (t, <sup>3</sup>J = 7.1 Hz, 3H, CH<sub>3</sub>CH<sub>2</sub>). <sup>13</sup>C NMR (101.62 MHz, DMSO-*d*<sub>6</sub>): δ 165.0 (C-8), 163.2 (C-12), 162.9 (CH=), 160.0 (C-4), 150.0 (C-10), 137.1 (C-5), 117.6 (C-7), 72.5 (C-OH), 36.6 (CH<sub>2</sub>), 29.4 (CH<sub>3</sub>), 8.3 (CH<sub>3</sub>CH<sub>2</sub>). IR (KBr): 3384, 3174, 3074, 2969, 2906, 2853, 2794, 1723, 1615, 1585, 1403, 1370, 1346, 1309, 1217, 1180, 1128, 1054, 1034, 1004, 937, 909, 884, 849, 820, 724, 694, 666, 632, 595, 572, 537, 512, 411 cm<sup>-1</sup>. Found: C, 47.15; H, 4.96; N, 24.78. Calcd for C<sub>11</sub>H<sub>13</sub>N<sub>5</sub>O<sub>4</sub>: C, 47.31; H, 4.69; N, 25.08.

5-*{[4-(1-Hydroxy-1-methylbutyl)-1H-1,2,3-triazol-5-yl]methylidene}pyrimidine-2,4,6(1H,3H,5H)-trione (3c)*: yield 260 mg, 87%. Colorless powder, mp >260 °C (decomp.). <sup>1</sup>H NMR (400.13 MHz, DMSO-*d*<sub>6</sub>): δ 15.60 (br s, 1H, NH), 11.69 [br s, 1H, NH(C=O)], 11.51 [br s, 1H, NH(C=O)], 9.20 (s, 1H, HC=), 5.44 (s, 1H, OH), 1.89–1.70 (m, 2H, CH<sub>2</sub>CH<sub>2</sub>CH<sub>3</sub>), 1.59 (s, 3H, CH<sub>3</sub>), 1.39-1.03 (m, 2H, CH<sub>2</sub>CH<sub>2</sub>CH<sub>3</sub>), 0.84 (t, <sup>3</sup>J = 7.3 Hz, 3H, CH<sub>2</sub>CH<sub>2</sub>CH<sub>3</sub>). <sup>13</sup>C NMR (101.62 MHz, DMSO-*d*<sub>6</sub>): δ 170.1 (C-8), 164.1 (C-12), 162.9 (CH=), 159.8 (C-4), 150.1 (C-10), 136.7 (C-5), 116.9 (C-7), 72.4 (C-OH), 46.4 (CH<sub>3</sub>CH<sub>2</sub>CH<sub>2</sub>), 29.9 (CH<sub>3</sub>), 16.9 (CH<sub>3</sub>CH<sub>2</sub>CH<sub>2</sub>), 14.3 (CH<sub>3</sub>CH<sub>2</sub>CH<sub>2</sub>). IR (KBr): 3419, 3211, 2970, 2940, 2874, 2849, 2783, 1686, 1615, 1458, 1409, 1383, 1335, 1314, 1204, 1167, 1059, 1045, 917, 880, 808, 777, 753, 695, 684, 645, 620, 571, 534, 420 cm<sup>-1</sup>. Found: C, 49.25; H, 5.43; N, 23.57. Calcd for C<sub>12</sub>H<sub>15</sub>N<sub>5</sub>O<sub>4</sub>: C, 49.14; H, 5.16; N, 23.88.

5-*{[4-(1-Hydroxy-1-cyclohexyl)-1H-1,2,3-triazol-5-yl]methylidene}pyrimidine-2,4,6(1H,3H,5H)-trione (3d)*: yield 220 mg, 73%. Colorless powder, mp >260 °C (decomp.). <sup>1</sup>H NMR (400.13 MHz, DMSO-*d*<sub>6</sub>): δ 15.58 (br s, 1H, NH), 11.67 [br s, 1H, NH(C=O)], 11.50 [br s, 1H, NH(C=O)], 9.12 (s, 1H, CH=), 5.35 (br s, 1H, OH), 1.96–1.28 [m, 10H, (CH<sub>2</sub>)<sub>5</sub>]. <sup>13</sup>C NMR (101.62 MHz, DMSO-*d*<sub>6</sub>): δ 163.9 (C-8), 163.7 (C-12), 162.7 (CH=), 160.3 (C-4), 149.8 (C-10), 136.5 (C-5), 116.4 (C-7), 70.7 (C-OH), 38.5, 25.1, 21.2 (CH<sub>2</sub> from cyclohexyl). IR (KBr): 3419, 3211, 2970, 2940, 2874, 2849, 2783, 1686, 1615, 1458, 1409, 1383, 1335, 1314, 1229, 1167, 1059, 1045, 916, 880, 808, 777, 753, 695, 684, 620, 571, 534, 433, 420 cm<sup>-1</sup>. Found: C, 51.09; H, 4.99; N, 22.63. Calcd for C<sub>13</sub>H<sub>15</sub>N<sub>5</sub>O<sub>4</sub>: C, 51.15; H, 4.95; N, 22.94.

5-*{[4-(1-Hydroxy-1-methylethyl)-1H-1,2,3-triazol-5-yl]methylidene}-1,3-dimethylpyrimidine-2,4,6(1H,3H,5H)-trione (3e)*: yield 260 mg, 89%. Colorless powder, mp >260 °C (decomp.). <sup>1</sup>H NMR (400.13 MHz, DMSO-*d*<sub>6</sub>): δ 15.71 (br s, 1H, NH), 9.90 (s, 1H, HC=), 3.18 (s, 6H, NCH<sub>3</sub>), 1.58 (s, 6H, (CH<sub>3</sub>)<sub>2</sub>). <sup>13</sup>C NMR (101.62 MHz, DMSO-*d*<sub>6</sub>): δ 170.1 (C-8), 159.4 (C-12), 158.1 (=CH), 155.0 (C-4), 151.0 (C-10), 134.4 (C-5), 116.7 (C-7), 69.2 (C-OH), 30.1 (NCH<sub>3</sub>), 27.0 (CH<sub>3</sub>). IR (KBr): 3451, 3148, 2989, 2923, 1678, 1632, 1599, 1459, 1434, 1417, 1393, 1382, 1361, 1327, 1315, 1267, 1240, 1185, 984, 960, 951, 856, 837, 755, 632, 604, 503, 430, 413 cm<sup>-1</sup>. Found: C, 49.35; H, 4.95; N, 23.64. Calcd for C<sub>12</sub>H<sub>15</sub>N<sub>5</sub>O<sub>4</sub>: C, 49.14; H, 5.16; N, 23.88.

5-*{[4-(1-Hydroxy-1-methylpropyl)-1H-1,2,3-triazol-5-yl]methylidene}-1,3-dimethylpyrimidine-2,4,6(1H,3H,5H)-trione (4f)*: yield 260 mg, 83%. Yellow powder, mp >260 °C (decomp.). <sup>1</sup>H NMR (400.13 MHz, DMSO-*d*<sub>6</sub>): δ 9.87 (s, 1H, HC=), 3.17 (s, 6H, NCH<sub>3</sub>), 1.86-1.81 (m, 2H, CH<sub>2</sub>), 1.59 (s, 3H, CH<sub>3</sub>), 0.73 (t, <sup>3</sup>J = 7.1 Hz, 3H, CH<sub>3</sub>CH<sub>2</sub>). <sup>13</sup>C NMR (101.62 MHz, DMSO-*d*<sub>6</sub>): δ 168.1 (C-8), 158.6 (C-12), 158.1 (HC=), 151.1 (C-10), 150.0 (C-4), 135.2 (C-5), 120.1 (C-7), 72.2 (C-OH), 35.7 (CH<sub>2</sub>), 28.5 (NCH<sub>3</sub>), 27.0 (CH<sub>3</sub>), 8.1 (CH<sub>3</sub>CH<sub>2</sub>). IR (KBr): 3336, 3118, 2969, 2923, 1720, 1701, 1680, 1634, 1598, 1560, 1488, 1462, 1410, 1378, 1339, 1312, 1300, 1265,

1235, 1208, 1177, 1141, 1084, 1043, 1056, 980, 963, 950, 927, 914, 891, 858, 837, 794, 770, 763, 670, 639, 617, 597, 496, 455, 420  $\text{cm}^{-1}$ . Found: C, 51.15; H, 5.16; N, 22.36. Calcd for  $\text{C}_{13}\text{H}_{17}\text{N}_5\text{O}_4$ : C, 50.81; H, 5.58; N, 22.79.

5-[[4-(1-Hydroxy-1-methylbutyl)-1H-1,2,3-triazol-5-yl]methylidene]-1,3-dimethylpyrimidine-2,4,6(1H,3H,5H)-trione (**3g**): yield 280 mg, 86%. Yellow powder, mp >260 °C (decomp.).  $^1\text{H}$  NMR (400.13 MHz,  $\text{DMSO-}d_6$ ):  $\delta$  9.86 (s, 1H, HC=), 3.16 (s, 6H,  $\text{NCH}_3$ ), 1.80-1.76 (m, 2H,  $\text{CH}_2\text{CH}_2\text{CH}_3$ ), 1.58 (s, 3H,  $\text{CH}_3$ ), 1.31-1.30 (m, 2H,  $\text{CH}_2\text{CH}_2\text{CH}_3$ ) 0.76 (t,  $^3J = 7.3$  Hz, 3H,  $\text{CH}_3\text{CH}_2\text{CH}_2$ ).  $^{13}\text{C}$  NMR (101.62 MHz,  $\text{DMSO-}d_6$ ):  $\delta$  168.2 (C-8), 159.4 (C-12), 158.2 (HC=), 152.4 (C-10), 151.1 (C-4), 135.2 (C-5), 119.1 (C-7), 71.9 (C-OH), 45.3 ( $\text{CH}_3\text{CH}_2\text{CH}_2$ ), 28.0 ( $\text{NCH}_3$ ), 27.1 ( $\text{CH}_3$ ), 16.7 ( $\text{CH}_3\text{CH}_2\text{CH}_2$ ), 14.1 ( $\text{CH}_3\text{CH}_2\text{CH}_2$ ). IR (KBr): 3403, 3332, 3200, 3120, 2975, 2913, 1706, 1677, 1633, 1466, 1451, 1427, 1410, 1397, 1381, 1352, 1317, 1265, 1237, 1208, 1164, 1137, 1118, 1080, 1053, 1028, 980, 950, 851, 820, 779, 753, 710, 649, 602, 497, 470, 415  $\text{cm}^{-1}$ . Found: C, 52.38; H, 5.93; N, 21.54. Calcd for  $\text{C}_{14}\text{H}_{19}\text{N}_5\text{O}_4$ : C, 52.33; H, 5.96; N, 21.79.

5-[[4-(1-Hydroxy-1-cyclohexyl)-1H-1,2,3-triazol-5-yl]methylidene]-1,3-dimethylpyrimidine-2,4,6(1H,3H,5H)-trione (**4h**): yield 220 mg, 72%. Yellow powder, mp >260 °C (decomp.).  $^1\text{H}$  NMR (400.13 MHz,  $\text{DMSO-}d_6$ ):  $\delta$  9.94 (s, 1H, HC=), 3.18 (s, 6H,  $\text{NCH}_3$ ) 1.84-1.57 [m, 10H,  $(\text{CH}_2)_5$ ].  $^{13}\text{C}$  NMR (101.62 MHz,  $\text{DMSO-}d_6$ ):  $\delta$  168.1 (C-8), 162.7 (C-12), 158.1 (CH=), 151.0 (C-10), 150.1 (C-4), 135.3 (C-5), 119.2 (C-7), 69.3 (C-OH), 27.9 ( $\text{NCH}_3$ ), 27.0, 24.8, 21.0 ( $\text{CH}_2$  from cyclohexyl). IR (KBr): 3437, 3133, 2999, 2935, 1698, 1674, 1639, 1598, 1454, 1429, 1393, 1383, 1319, 1265, 1235, 1157, 1139, 1083, 1038, 982, 965, 949, 908, 846, 825, 794, 761, 751, 608, 535, 499, 473, 425, 410  $\text{cm}^{-1}$ . Found: C, 54.15; H, 5.93; N, 20.91. Calcd for  $\text{C}_{14}\text{H}_{19}\text{N}_5\text{O}_4$ : C, 54.05; H, 5.75; N, 21.01.

# $^1\text{H}$ and $^{13}\text{C}$ spectra of the synthesized products















