

Electronic Supplementary Information

An efficient route for the synthesis of a new class of pyrido[2,3-*d*]pyrimidine derivatives

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Characterization data of selected compounds are as follows:

General procedure for the synthesis of compounds **6** with microwave irradiation

In a 10-mL reaction vial, an 4-arylidene-3-methylisoxazol-5(4*H*)-ones **4** (1 mmol), 2,6-diamino-pyrimidin-4(3*H*)-one **5** (1 mmol), DMF and HOAc (2 mL) (2:1, V/V) were mixed and then capped. The mixture was irradiated at 240 W (initial power 150 W, maximum power 240 W) at 140 °C for a given time. The reaction mixture was cooled to room temperature and poured into water (50 mL), filtered to give the crude product, which was further purified by recrystallization from mixed DMF-EtOH to give pure 2-amino-6-(1-hydroxyiminoethyl)-5,6-dihydro-5-arylpyrido[2,3-*d*]pyrimidine-4,7(3*H*,8*H*)-dione **6**.

2-amino-6-(1-hydroxyiminoethyl)-5-(4-fluorophenyl)-5,6-dihydropyrido[2,3-*d*]pyrimidine-4,7(3*H*,8*H*)-dione (**6a**)

IR (KBr ν , cm^{-1}): 3478, 3340, 3198, 2944, 1687, 1649, 1508, 1480, 1358, 1312, 816, 773, 622 cm^{-1} ;

^1H NMR (DMSO-*d*₆) (δ , ppm): δ = 10.84 (s, 1H, OH), 10.57 (s, 1H, NH), 10.23 (s, 1H, NH), 7.24–7.21 (m, 2H, ArH), 7.14–7.10 (m, 2H, ArH), 6.52 (brs, 2H, NH₂), 4.23 (s, 1H, CH), 3.30 (s, 1H, CH), 1.88 (s, 3H, CH₃);

^{13}C NMR (DMSO-*d*₆) (δ , ppm): δ 169.7, 161.7, 156.6, 155.2, 151.7, 143.7, 130.7, 129.5, 128.4, 90.2, 56.1, 35.8, 13.1;

Anal. calcd for C₁₅H₁₄FN₅O₃: C, 54.38; H, 4.26; N, 21.14. Found: C, 54.15; H, 4.07; N, 21.44.

2-amino-6-(1-hydroxyiminoethyl)-5-(4-chlorophenyl)-5,6-dihydropyrido[2,3-*d*]pyrimidine-4,7(3*H*,8*H*)-dione (**6b**)

IR (KBr ν , cm^{-1}): 3448, 3320, 3197, 2916, 1686, 1648, 1533, 1489, 1358, 1309, 813, 773, 623 cm^{-1} ; ^1H NMR (DMSO-*d*₆) (δ , ppm): δ = 10.87 (s, 1H, OH), 10.61 (s, 1H, NH), 10.28 (s, 1H, NH), 7.36 (d, 2H, ArH, *J* = 8.4 Hz), 7.21 (d, 2H, ArH, *J* = 8.4 Hz), 6.55 (brs, 2H, NH₂), 4.23 (s, 1H, CH), 3.31 (s, 1H, CH), 1.88 (s, 3H, CH₃); ^{13}C NMR (DMSO-*d*₆) (δ , ppm): δ 169.7, 161.8, 156.7, 155.3, 152.6, 141.7, 131.4, 128.8, 128.7, 89.6, 55.0, 36.4, 13.1; Anal. calcd for C₁₅H₁₄ClN₅O₃: C, 51.81; H, 4.06; N, 20.14. Found: C, 51.66; H, 4.26; N, 19.98.

2-amino-6-(1-hydroxyiminoethyl)-5-(4-bromophenyl)-5,6-dihydropyrido[2,3-*d*]pyrimidine-4,7(3*H*,8*H*)-dione (**6c**)

IR (KBr ν , cm^{-1}): 3468, 3329, 3193, 2913, 1689, 1646, 1540, 1487, 1358, 1321, 830, 787, 636 cm^{-1} ;

^1H NMR (DMSO-*d*₆) (δ , ppm): δ = 10.89 (s, 1H, OH), 10.63 (s, 1H, NH), 10.30 (s, 1H, NH), 7.49 (d, 2H, ArH, *J* = 8.4 Hz), 7.15 (d, 2H, ArH, *J* = 8.4 Hz), 6.56 (brs, 2H, NH₂), 4.20 (s, 1H, CH), 3.31 (s, 1H, CH), 1.87 (s, 3H, CH₃);

^{13}C NMR (DMSO-*d*₆) (δ , ppm): δ 169.6, 161.8, 156.8, 155.2, 152.6, 142.2, 131.6, 129.9, 119.8, 89.5, 54.9, 36.5, 13.1;

Anal. calcd for C₁₅H₁₄BrN₅O₃: C, 45.94; H, 3.60; N, 17.86. Found: C, 45.79; H, 3.46; N, 18.01.

2-amino-6-(1-hydroxyiminoethyl)-5-(2,4-dichlorophenyl)-5,6-dihydropyrido[2,3-*d*]pyrimidine-4,7(3*H*,8*H*)-dione (**6d**)

IR (KBr ν , cm^{-1}): 3451, 3318, 3193, 2909, 1697, 1654, 1541, 1469, 1354, 1326, 844, 760, 635 cm^{-1} ;

^1H NMR (DMSO-*d*₆) (δ , ppm): δ = 10.98 (s, 1H, OH), 10.71 (s, 1H, NH), 10.42 (s, 1H, NH), 7.24 (s, 1H, ArH), 7.37 (dd, 1H, ArH, *J*₁ = 8.4 Hz, *J*₂ = 2.0 Hz), 6.94 (d, 1H, ArH, *J* = 8.4 Hz), 6.62 (brs, 2H, NH₂), 4.57 (s, 1H, CH), 3.32 (s, 1H, CH), 1.91 (s, 3H, CH₃);

^{13}C NMR (DMSO-*d*₆) (δ , ppm): δ 169.5, 161.78, 155.5, 154.2, 150.7, 143.7, 132.4, 129.4, 128.3, 119.3, 115.2, 95.8, 52.1, 35.2, 13.3;

Anal. calcd for C₁₅H₁₃Cl₂N₅O₃: C, 47.14; H, 3.43; N, 18.32. Found: C, 46.98; H, 3.21; N, 18.16.

2-amino-6-(1-hydroxyiminoethyl)-5-(2-chlorophenyl)-5,6-dihydropyrido[2,3-*d*]pyrimidine-4,7(3*H*,8*H*)-dione (**6e**)

IR (KBr ν , cm^{-1}): 3458, 3324, 3193, 2908, 1699, 1652, 1540, 1468, 1353, 1328, 836, 759, 633 cm^{-1} ;

^1H NMR (DMSO-*d*₆) (δ , ppm): δ = 10.84 (s, 1H, OH), 10.58 (s, 1H, NH), 10.21 (s, 1H, NH), 6.68–6.52 (m, 4H, ArH), 5.97 (brs, 2H, NH₂), 4.15 (s, 1H, CH), 3.28 (s, 1H, CH), 1.86 (s, 3H, CH₃);

^{13}C NMR (DMSO-*d*₆) (δ , ppm): δ 170.1, 161.8, 158.2, 156.4, 155.1, 152.8, 134.6, 127.9, 123.1, 119.5, 114.1, 90.4, 55.2, 36.1, 13.1;

Anal. calcd for C₁₅H₁₄ClN₅O₃: C, 51.81; H, 4.06; N, 20.14. Found: C, 51.70; H, 3.87; N, 19.96.

2-amino-6-(1-hydroxyiminoethyl)-5-(3-nitrophenyl)-5,6-dihydropyrido[2,3-*d*]pyrimidine-4,7(3*H*,8*H*)-dione (6f)IR (KBr ν , cm^{-1}): 3479, 3321, 3192, 2908, 1697, 1649, 1529, 1490, 1352, 1315, 824, 787, 683 cm^{-1} ; ^1H NMR (DMSO- d_6) (δ , ppm): δ = 10.91 (s, 1H, OH), 10.66 (s, 1H, NH), 10.39 (s, 1H, NH), 8.09 (dd, 1H, ArH, J_1 = 8.0 Hz, J_2 = 1.2 Hz), 8.03 (s, 1H, ArH), 7.71 (d, 1H, ArH, J = 8.0 Hz), 7.63 (t, 1H, ArH, J = 8.0 Hz), 6.62 (brs, 2H, NH_2), 4.40 (s, 1H, CH), 3.44 (s, 1H, CH), 1.90 (s, 3H, CH_3); ^{13}C NMR (DMSO- d_6) (δ , ppm): δ 170.1, 161.8, 157.5, 153.2, 150.1, 139.8, 133.4, 128.8, 127.6, 120.2, 118.8, 89.6, 55.0, 36.4, 13.1;Anal. calcd for $\text{C}_{15}\text{H}_{14}\text{N}_6\text{O}_5$: C, 50.28; H, 3.94; N, 23.45. Found: C, 50.02; H, 4.09; N, 23.21.**2-amino-6-(1-hydroxyiminoethyl)-5,6-dihydro-5-phenyl-pyrido[2,3-*d*]pyrimidine-4,7(3*H*,8*H*)-dione (6g)**IR (KBr ν , cm^{-1}): 3423, 3327, 3198, 2913, 1696, 1652, 1538, 1493, 1357, 1316, 835, 762, 698 cm^{-1} ; ^1H NMR (DMSO- d_6) (δ , ppm): δ = 10.87 (s, 1H, OH), 10.60 (s, 1H, NH), 10.27 (s, 1H, NH), 7.32–7.29 (m, 2H, ArH), 7.23–7.18 (m, 3H, ArH), 6.54 (brs, 2H, NH_2), 4.22 (s, 1H, CH), 3.31 (s, 1H, CH), 1.88 (s, 3H, CH_3); ^{13}C NMR (DMSO- d_6) (δ , ppm): δ 168.8, 161.7, 155.9, 155.2, 151.7, 145.5, 135.4, 130.7, 126.5, 89.8, 57.2, 38.2, 13.0;Anal. calcd for $\text{C}_{15}\text{H}_{15}\text{N}_5\text{O}_3$: C, 57.50; H, 4.83; N, 22.35. Found: C, 57.65; H, 5.01; N, 22.50.**2-amino-6-(1-hydroxyiminoethyl)-5,6-dihydro-5-(3,4-dimethoxyphenyl)pyrido[2,3-*d*]pyrimidine-4,7(3*H*,8*H*)-dione (6h)**IR (KBr ν , cm^{-1}): 3419, 3324, 3190, 2909, 1697, 1650, 1539, 1489, 1353, 1316, 875, 770, 622 cm^{-1} ; ^1H NMR (DMSO- d_6) (δ , ppm): δ = 10.84 (s, 1H, OH), 10.59 (s, 1H, NH), 10.19 (s, 1H, NH), 6.89–6.57 (m, 3H, ArH), 6.51 (brs, 2H, NH_2), 4.18 (s, 1H, CH), 3.73 (s, 3H, OCH_3), 3.70 (s, 3H, OCH_3), 3.32 (s, 1H, CH), 1.88 (s, 3H, CH_3); ^{13}C NMR (DMSO- d_6) (δ , ppm): δ 169.7, 161.6, 156.6, 154.5, 152.4, 149.8, 139.7, 133.4, 130.8, 121.4, 118.7, 89.9, 65.6, 56.6, 36.3, 13.2;Anal. calcd for $\text{C}_{17}\text{H}_{19}\text{N}_5\text{O}_5$: C, 54.69; H, 5.13; N, 18.76. Found: C, 54.88; H, 4.96; N, 18.87.**2-amino-6-(1-hydroxyiminoethyl)-5,6-dihydro-5-(3,4,5-trimethoxyphenyl)pyrido[2,3-*d*]pyrimidine-4,7(3*H*,8*H*)-dione (6i)**IR (KBr ν , cm^{-1}): 3433, 3336, 3194, 2937, 1694, 1648, 1510, 1460, 1355, 1324, 806, 783, 636 cm^{-1} ; ^1H NMR (DMSO- d_6) (δ , ppm): δ = 10.85 (s, 1H, OH), 10.60 (s, 1H, NH), 10.24 (s, 1H, NH), 6.53 (brs, 2H, NH_2), 6.50 (s, 2H, ArH), 4.19 (s, 1H, CH), 3.71 (s, 6H, OCH_3), 3.69 (s, 1H, CH), 3.62 (s, 3H, OCH_3), 1.88 (s, 3H, CH_3); ^{13}C NMR (DMSO- d_6) (δ , ppm): δ 169.5, 161.6, 157.7, 155.5, 152.6, 138.6, 132.0, 130.3, 120.5, 91.3, 63.6, 63.6, 56.6, 36.3, 13.0;Anal. calcd for $\text{C}_{18}\text{H}_{21}\text{N}_5\text{O}_6$: C, 53.59; H, 5.25; N, 17.36. Found: C, 53.71; H, 5.38; N, 17.57.**2-amino-6-(1-hydroxyiminoethyl)-5,6-dihydro-5-(4-methoxyphenyl)pyrido[2,3-*d*]pyrimidine-4,7(3*H*,8*H*)-dione (6j)**IR (KBr ν , cm^{-1}): 3461, 3302, 3195, 2910, 1688, 1649, 1511, 1472, 1357, 1307, 815, 766, 619 cm^{-1} ; ^1H NMR (DMSO- d_6) (δ , ppm): δ = 10.84 (s, 1H, OH), 10.59 (s, 1H, NH), 10.25 (s, 1H, NH), 7.10 (d, 2H, ArH, J = 8.4 Hz), 6.85 (d, 2H, ArH, J = 8.4 Hz), 6.53 (brs, 2H, NH_2), 4.17 (s, 1H, CH), 3.71 (s, 1H, OCH_3), 3.28 (s, 1H, CH), 1.87 (s, 3H, CH_3); ^{13}C NMR (DMSO- d_6) (δ , ppm): δ 169.2, 161.6, 157.8, 155.5, 152.4, 138.9, 132.8, 127.9, 114.7, 88.7, 65.1, 54.0, 34.6, 12.8;Anal. calcd for $\text{C}_{16}\text{H}_{17}\text{N}_5\text{O}_4$: C, 55.97; H, 4.99; N, 20.40. Found: C, 56.09; H, 5.13; N, 20.54.**2-amino-6-(1-hydroxyiminoethyl)-5,6-dihydro-5-(4-hydroxy-3-nitrophenyl)pyrido[2,3-*d*]pyrimidine-4,7(3*H*,8*H*)-dione (6k)**IR (KBr ν , cm^{-1}): 3392, 3328, 3179, 2919, 1689, 1662, 1536, 1485, 1350, 1308, 826, 764, 632 cm^{-1} ; ^1H NMR (DMSO- d_6) (δ , ppm): δ = 11.97 (s, 1H, OH), 10.87 (s, 1H, OH), 10.62 (s, 1H, NH), 10.31 (s, 1H, NH), 7.64 (s, 1H, ArH), 7.41 (dd, 1H, ArH, J_1 = 8.8 Hz, J_2 = 4.0 Hz), 7.09 (d, 1H, ArH, J = 8.8 Hz), 6.58 (brs, 2H, NH_2), 4.23 (s, 1H, CH), 3.30 (s, 1H, CH), 1.92 (s, 3H, CH_3); ^{13}C NMR (DMSO- d_6) (δ , ppm): δ 169.6, 161.8, 156.8, 155.3, 152.6, 151.1, 136.6, 134.2, 133.8, 122.8, 119.7, 89.5, 54.8, 35.7, 13.0;Anal. calcd for $\text{C}_{15}\text{H}_{14}\text{N}_6\text{O}_6$: C, 48.13; H, 3.77; N, 22.45. Found: C, 48.01; H, 3.89; N, 22.66.**2-amino-6-(1-hydroxyiminoethyl)-5,6-dihydro-5-(4-methylphenyl)pyrido[2,3-*d*]pyrimidine-4,7(3*H*,8*H*)-dione (6l)**IR (KBr ν , cm^{-1}): 3436, 3291, 3161, 2914, 1696, 1638, 1541, 1488, 1353, 1306, 823, 734, 628 cm^{-1} ; ^1H NMR (DMSO- d_6) (δ , ppm): δ = 11.05 (s, 1H, OH), 10.65 (s, 1H, NH), 10.50 (s, 1H, NH), 7.10 (d, 2H, ArH, J = 8.0 Hz), 7.04 (d, 2H, ArH, J = 8.0 Hz), 6.59 (brs, 2H, NH_2), 4.12 (s, 1H, CH), 3.97 (s, 1H, CH), 2.25 (s, 3H, CH_3), 1.66 (s, 3H, CH_3); ^{13}C NMR (DMSO- d_6) (δ , ppm): δ 170.4, 161.6, 156.7, 155.4, 151.6, 140.4, 135.9, 129.3, 126.3, 90.2, 49.3, 27.3, 20.7, 17.8;Anal. calcd for $\text{C}_{16}\text{H}_{17}\text{N}_5\text{O}_3$: C, 58.71; H, 5.23; N, 21.39. Found: C, 58.92; H, 5.38; N, 21.54.**2-amino-6-(1-hydroxyiminoethyl)-5-(benzo[d][1,3]dioxol-5-yl)-5,6-dihydropyrido[2,3-*d*]pyrimidine-4,7(3*H*,8*H*)-dione (6m)**IR (KBr ν , cm^{-1}): 3467, 3315, 3192, 2906, 1692, 1652, 1538, 1442, 1348, 1315, 806, 775, 631 cm^{-1} ; ^1H NMR (DMSO- d_6) (δ , ppm): δ = 10.84 (s, 1H, OH), 10.58 (s, 1H, NH), 10.21 (s, 1H, NH), 6.83–6.78 (m, 2H, ArH), 6.62–6.59 (m, 1H, ArH), 6.52 (brs, 2H, NH_2), 5.97 (s, 2H, OCH_2O), 4.15 (s, 1H, CH), 3.28 (s, 1H, CH), 1.86 (s, 3H, CH_3); ^{13}C NMR (DMSO- d_6) (δ , ppm): δ 169.5, 160.9, 155.4, 153.2, 152.3, 143.7, 135.7, 133.4, 130.4, 123.4, 120.1, 106.9, 89.7, 54.4, 34.8, 13.3;Anal. calcd for $\text{C}_{16}\text{H}_{15}\text{N}_5\text{O}_5$: C, 53.78; H, 4.23; N, 19.60. Found: C, 53.93; H, 4.04; N, 19.75.**2-amino-6-(1-hydroxyiminoethyl)-5,6-dihydro-5-(thiophen-2-yl)pyrido[2,3-*d*]pyrimidine-4,7(3*H*,8*H*)-dione (6n)**IR (KBr ν , cm^{-1}): 3445, 3334, 3196, 2915, 1689, 1648, 1536, 1484, 1349, 1312, 816, 700, 626 cm^{-1} ;

¹H NMR (DMSO-*d*₆) (δ, ppm): δ = 10.90 (s, 1H, OH), 10.62 (s, 1H, NH), 10.27 (s, 1H, NH), 7.30–6.90 (m, 3H, ArH), 6.56 (brs, 2H, NH₂), 4.46 (s, 1H, CH), 3.49 (s, 1H, CH), 1.85 (s, 3H, CH₃);

¹³C NMR (DMSO-*d*₆) (δ, ppm): δ 169.9, 161.6, 156.2, 115.3, 152.2, 146.8, 127.2, 124.2, 123.9, 91.1, 55.2, 32.6, 13.0;

Anal. calcd for C₁₃H₁₃N₃O₃S: C, 48.89; H, 4.10; N, 21.93. Found: C, 49.05; H, 3.95; N, 21.78.

General procedure for the synthesis of compounds **8** with microwave irradiation

In a 10-mL reaction vial, an 4-arylidene-3-methylisoxazol-5(4*H*)-ones **4** (1 mmol), naphthalen-2-amine **7** (1 mmol), DMF and HOAc (2 mL) (2:1, V/V) were mixed and then capped. The mixture was irradiated at 240 W (initial power 150 W, maximum power 240 W) at 140 °C for a given time. The reaction mixture was cooled to room temperature and poured into water (50 mL), filtered to give the crude product, which was further purified by recrystallization from mixed DMF-EtOH to give pure General procedure for the synthesis of 1-aryl-2-(1-hydroxyiminoethyl)-1,2,3,4-tetrahydro-3-oxobenzof[*f*]quinolines **8**

1-(4-chlorophenyl)-2-(1-hydroxyiminoethyl)-1,2,3,4-tetrahydro-3-oxobenzof[*f*]quinolines (8a)

IR (KBr v, cm⁻¹): 3354, 3217, 3011, 2945, 2844, 1678, 1521, 1486, 815, 744, 606 cm⁻¹;

¹H NMR (DMSO-*d*₆) (δ, ppm): δ = 10.74 (s, 1H, OH), 10.60 (s, 1H, NH), 7.88–7.83 (m, 3H, ArH), 7.44 (t, 1H, ArH, *J* = 7.6 Hz), 7.37–7.32 (m, 3H, ArH), 7.22 (d, 1H, ArH, *J* = 8.8 Hz), 7.15 (d, 2H, ArH, *J* = 8.4 Hz), 5.21 (s, 1H, CH), 3.48 (s, 1H, CH), 1.90 (s, 3H, CH₃);

¹³C NMR (DMSO-*d*₆) (δ, ppm): δ 167.9, 151.6, 143.1, 134.9, 132.0, 130.3, 129.7, 129.1, 128.8, 127.6, 127.3, 126.9, 125.1, 120.3, 118.2, 115.3, 50.3, 42.4, 18.1;

Anal. calcd for C₂₁H₁₇ClN₂O₂: C, 69.14; H, 4.70; N, 7.68. Found: C, 69.27; H, 4.55; N, 7.47.

1-(4-bromophenyl)-2-(1-hydroxyiminoethyl)-1,2,3,4-tetrahydro-3-oxobenzof[*f*]quinolines (8b)

IR (KBr v, cm⁻¹): 3362, 3216, 3021, 2947, 2838, 1684, 1522, 1467, 836, 723, 660 cm⁻¹;

¹H NMR (DMSO-*d*₆) (δ, ppm): δ = 11.29 (s, 1H, OH), 10.94 (s, 1H, NH), 7.96–7.86 (m, 3H, ArH), 7.64 (d, 1H, ArH, *J* = 8.0 Hz), 7.51–7.32 (m, 4H, ArH), 7.05 (d, 2H, ArH, *J* = 8.4 Hz), 5.01 (s, 1H, CH), 4.17 (s, 1H, CH), 1.52 (s, 3H, CH₃);

¹³C NMR (DMSO-*d*₆) (δ, ppm): δ 166.4, 152.6, 142.5, 136.9, 134.1, 131.3, 130.4, 129.2, 128.7, 127.5, 127.2, 126.9, 125.4, 122.1, 119.3, 118.4, 53.4, 41.7, 17.0;

Anal. calcd for C₂₁H₁₇BrN₂O₂: C, 61.63; H, 4.19; N, 6.84. Found: C, 61.51; H, 4.30; N, 6.67.

1-(2,4-dichlorophenyl)-2-(1-hydroxyiminoethyl)-1,2,3,4-tetrahydro-3-oxobenzof[*f*]quinolines (8c)

IR (KBr): 3364, 3221, 3018, 2960, 2842, 1676, 1543, 1520, 808, 736, 654 cm⁻¹;

¹H NMR (DMSO-*d*₆) (δ, ppm): δ = 11.05 (s, 1H, OH), 10.98 (s, 1H, NH), 7.95 (d, 1H, ArH, *J* = 9.2 Hz), 7.89 (d, 1H, ArH, *J* = 8.0 Hz), 7.74 (s, 1H, ArH), 7.76–7.26 (m, 5H, ArH), 6.80 (d, 1H, ArH, *J* = 8.8 Hz), 5.26 (s, 1H, CH), 4.26 (s, 1H, CH), 1.53 (s, 3H, CH₃);

¹³C NMR (DMSO-*d*₆) (δ, ppm): δ 166.8, 149.9, 138.3, 136.3, 133.1, 132.9, 130.7, 130.4, 130.0, 129.8, 129.8, 129.1, 128.4, 127.9, 124.6, 121.7, 117.1, 115.4, 45.8, 38.2, 17.8;

Anal. calcd for C₂₁H₁₆Cl₂N₂O₂: C, 63.17; H, 4.04; N, 7.02. Found: C, 63.29; H, 4.15; N, 7.17.

1-(2-chlorophenyl)-2-(1-hydroxyiminoethyl)-1,2,3,4-tetrahydro-3-oxobenzof[*f*]quinolines (8d)

IR (KBr v, cm⁻¹): 3371, 3197, 3031, 2967, 2863, 1681, 1554, 1517, 826, 766, 677 cm⁻¹;

¹H NMR (DMSO-*d*₆) (δ, ppm): δ = 10.85 (s, 1H, OH), 10.73 (s, 1H, NH), 7.88 (t, 2H, *J* = 5.6 Hz, ArH), 7.60 (d, 1H, ArH, *J* = 8.0 Hz), 7.55 (d, 1H, ArH, *J* = 8.4 Hz), 7.46 (t, 1H, ArH, *J* = 7.6 Hz), 7.36 (d, 1H, ArH, *J* = 7.6 Hz), 7.30–7.26 (m, 2H, ArH), 6.59 (d, 1H, ArH, *J* = 8.0 Hz) 5.49 (s, 1H, CH), 4.37 (s, 1H, CH), 1.93 (s, 3H, CH₃);

¹³C NMR (DMSO-*d*₆) (δ, ppm): δ 166.9, 152.0, 138.2, 136.3, 132.4, 131.1, 130.4, 129.4, 129.3, 129.0, 128.5, 128.0, 127.7, 126.8, 124.3, 121.9, 117.1, 115.1, 53.5, 37.2, 18.7;

Anal. calcd for C₂₁H₁₇ClN₂O₂: C, 69.14; H, 4.70; N, 7.68. Found: C, 67.31; H, 4.56; N, 7.45.

1-(3-nitrophenyl)-2-(1-hydroxyiminoethyl)-1,2,3,4-tetrahydro-3-oxobenzof[*f*]quinolines (8e)

IR (KBr v, cm⁻¹): 3364, 3234, 3041, 2943, 2861, 1674, 1541, 1431, 844, 765, 647 cm⁻¹;

¹H NMR (DMSO-*d*₆) (δ, ppm): δ = 11.40 (s, 1H, OH), 11.04 (s, 1H, NH), 8.10 (d, 1H, ArH, *J* = 8.4 Hz), 8.00–7.90 (m, 3H, ArH), 7.72 (d, 1H, *J* = 8.4 Hz, ArH), 7.63 (t, 1H, ArH, *J* = 8.0 Hz), 7.54 (d, 1H, ArH, *J* = 7.6 Hz), 7.46–7.37 (m, 3H, ArH), 5.21 (s, 1H, CH), 4.22 (s, 1H, CH), 1.54 (s, 3H, CH₃);

¹³C NMR (DMSO-*d*₆) (δ, ppm): δ 166.9, 153.1, 140.2, 138.0, 134.3, 132.0, 130.3, 129.5, 129.4, 129.2, 128.5, 128.3, 127.7, 125.5, 122.7, 121.1, 119.1, 117.2, 52.8, 39.9, 17.3;

Anal. calcd for C₂₁H₁₇N₃O₄: C, 67.19; H, 4.56; N, 11.19. Found: C, 67.36; H, 4.35; N, 11.37.

1-phenyl-2-(1-hydroxyiminoethyl)-1,2,3,4-tetrahydro-3-oxobenzof[*f*]quinolines (8f)

IR (KBr v, cm⁻¹): 3357, 3230, 3014, 2975, 2856, 1684, 1536, 1521, 820, 717, 680 cm⁻¹;

¹H NMR (DMSO-*d*₆) (δ, ppm): δ = 11.26 (s, 1H, OH), 10.92 (s, 1H, NH), 7.93 (d, 1H, ArH, *J* = 8.8 Hz), 7.88 (d, 1H, ArH, *J* = 8.0 Hz), 7.66 (d, 1H, ArH, *J* = 8.4 Hz), 7.44–7.18 (m, 6H, ArH), 7.12 (d, 2H, ArH, *J* = 7.2 Hz), 5.03 (s, 1H, CH), 4.21 (s, 1H, CH), 1.53 (s, 3H, CH₃);

¹³C NMR (DMSO-*d*₆) (δ, ppm): δ 168.1, 151.7, 142.3, 135.9, 131.2, 130.3, 129.5, 129.1, 128.9, 127.6, 127.2, 126.8, 124.4, 122.5, 116.9, 115.6, 49.1, 41.6, 17.8;

Anal. calcd for C₂₁H₁₈N₂O₂: C, 76.34; H, 5.49; N, 8.48. Found: C, 76.48; H, 5.37; N, 8.37.

1-(3,4-dimethoxyphenyl)-2-(1-hydroxyiminoethyl)-1,2,3,4-tetrahydro-3-oxobenzof[*f*]quinolines (8g)

IR (KBr v, cm⁻¹): 3361, 3229, 3060, 2984, 2833, 1676, 1567, 1542, 836, 752, 621 cm⁻¹;

¹H NMR (DMSO-*d*₆) (δ, ppm): δ = 10.71 (s, 1H, OH), 10.53 (s, 1H, NH), 7.89–7.80 (m, 3H, ArH), 7.43 (t, 1H, ArH, *J* = 7.6 Hz), 7.33 (t, 1H, ArH, *J* = 7.6 Hz), 7.21 (d, 1H, ArH, *J* = 8.0 Hz), 7.03 (s, 1H, ArH), 6.73 (d, 1H, ArH, *J* = 8.4 Hz), 5.12 (s, 1H, CH), 3.73 (s, 3H, OCH₃), 3.65 (s, 3H, OCH₃), 3.49 (s, 1H, CH), 1.90 (s, 3H, CH₃);

¹³C NMR (DMSO-*d*₆) (δ, ppm): δ 167.7, 152.7, 149.1, 147.9, 135.4, 134.4, 131.5, 130.2, 128.7, 128.5, 127.1, 123.9, 122.9, 118.8, 117.1, 116.8, 112.0, 111.7, 55.7, 55.6, 40.2, 13.1;

Anal. calcd for C₂₃H₂₂N₂O₄: C, 70.75; H, 5.68; N, 7.17. Found: C, 70.56; H, 5.54; N, 7.29.

1-(4-methylphenyl)-2-(1-hydroxyiminoethyl)-1,2,3,4-tetrahydro-3-oxobenzo[*f*]quinolines (8h)

IR (KBr *v*, cm⁻¹): 3341, 3218, 3107, 3018, 2967, 2864, 1676, 1536, 1517, 838, 756, 692 cm⁻¹;

¹H NMR (DMSO-*d*₆) (δ, ppm): δ = 11.22 (s, 1H, OH), 10.87 (s, 1H, NH), 7.93–7.87 (m, 2H, ArH), 7.63 (d, 1H, ArH, *J* = 8.4 Hz), 7.44–7.31 (m, 3H, ArH), 7.08 (d, 2H, ArH, *J* = 7.6 Hz), 6.99 (d, 2H, ArH, *J* = 8.0 Hz), 4.98 (s, 1H, CH), 4.18 (s, 1H, CH), 2.21 (s, 3H, CH₃), 1.52 (s, 3H, CH₃);

¹³C NMR (DMSO-*d*₆) (δ, ppm): δ 166.1, 148.6, 143.3, 133.6, 131.2, 130.3, 129.6, 128.7, 128.4, 127.8, 127.4, 126.2, 123.2, 120.4, 118.7, 113.3, 51.2, 38.9, 20.1, 17.7;

Anal. calcd for C₂₂H₂₀N₂O₂: C, 76.72; H, 5.85; N, 8.13. Found: C, 76.58; H, 5.64; N, 8.27.

General procedure for the synthesis of compounds 10 with microwave irradiation

In a 10-mL reaction vial, an 4-arylidene-2-phenyl-5(4*H*)-oxazolones **9** (1 mmol), 2,6-diaminopyrimidin-4(3*H*)-one **5** (1 mmol), DMF and HOAc (2 mL) (2:1, V/V) were mixed and then capped. The mixture was irradiated at 240 W at 140 °C for a given time. The reaction mixture was cooled to room temperature and poured into water (50 mL), filtered to give the crude product, which was further purified by recrystallization from mixed DMF-EtOH to give pure *N*-(2-amino-4,7-dioxo-5-aryl-3,4,5,6,7,8-hexahydropyrido[2,3-*d*]pyrimidin-6-yl)benzamides **10** (Table 3).

***N*-(2-amino-5-(4-fluorophenyl)-3,4,5,6,7,8-hexahydro-4,7-dioxopyrido[2,3-*d*]pyrimidin-6-yl)benzamide (10a)**

IR (KBr *v*, cm⁻¹): 3453, 3339, 3201, 2904, 1702, 1652, 1508, 1487, 1375, 874, 794, 705 cm⁻¹;

¹H NMR (DMSO-*d*₆) (δ, ppm): δ = 10.72 (s, 1H, NH), 10.59 (s, 1H, NH), 7.83 (d, 1H, NH, *J* = 6.4 Hz), 7.75 (d, 2H, ArH, *J* = 8.0 Hz), 7.56–7.43 (m, 3H, ArH), 7.09–6.98 (m, 4H, ArH), 6.66 (brs, 2H, NH₂), 5.06 (t, 1H, CH, *J* = 7.2 Hz), 4.49 (d, 1H, CH, *J* = 7.6 Hz);

¹³C NMR (DMSO-*d*₆) (δ, ppm): δ 169.3, 166.9, 161.5, 156.2, 155.5, 143.7, 133.9, 131.7, 131.1, 130.6, 128.8, 128.6, 127.3, 92.0, 54.0, 37.6;

Anal. calcd for C₂₀H₁₆FN₃O₃: C, 61.07; H, 4.10; N, 17.80. Found: C, 61.21; H, 4.23; N, 17.95.

***N*-(2-amino-5-(4-chlorophenyl)-3,4,5,6,7,8-hexahydro-4,7-dioxopyrido[2,3-*d*]pyrimidin-6-yl)benzamide (10b)**

IR (KBr *v*, cm⁻¹): 3439, 3390, 3298, 2921, 1699, 1651, 1521, 1488, 1385, 791, 703, 650 cm⁻¹;

¹H NMR (DMSO-*d*₆) (δ, ppm): δ = 10.70 (s, 1H, NH), 10.56 (s, 1H, NH), 7.87 (d, 1H, NH, *J* = 6.4 Hz), 7.76 (d, 2H, ArH, *J* = 7.2 Hz), 7.54–7.43 (m, 3H, ArH), 7.29 (d, 2H, ArH, *J* = 8.4 Hz), 6.99 (d, 2H, ArH, *J* = 8.8 Hz), 6.68 (brs, 2H, NH₂), 5.08 (t, 1H, CH, *J* = 7.2 Hz), 4.48 (d, 1H, CH, *J* = 7.6 Hz).

¹³C NMR (DMSO-*d*₆) (δ, ppm): δ 169.3, 166.9, 161.5, 156.2, 155.5, 137.9, 133.9, 131.7, 130.0, 128.6, 128.4, 127.6, 92.0, 53.9, 37.2;

Anal. calcd for C₂₀H₁₆ClN₃O₃: C, 58.61; H, 3.94; N, 17.09. Found: C, 58.41; H, 4.12; N, 16.92.

***N*-(2-amino-5-(4-bromophenyl)-3,4,5,6,7,8-hexahydro-4,7-dioxopyrido[2,3-*d*]pyrimidin-6-yl)benzamide (10c)**

IR (KBr *v*, cm⁻¹): 3438, 3336, 3200, 2904, 1699, 1654, 1519, 1486, 1374, 792, 713, 586 cm⁻¹;

¹H NMR (DMSO-*d*₆) (δ, ppm): δ = 10.74 (s, 1H, NH), 10.59 (s, 1H, NH), 7.95 (d, 1H, NH, *J* = 6.4 Hz), 7.77 (d, 2H, ArH, *J* = 8.0 Hz), 7.54–7.42 (m, 5H, ArH), 6.92 (d, 2H, ArH, *J* = 8.0 Hz), 6.70 (brs, 2H, NH₂), 5.08 (t, 1H, CH, *J* = 7.2 Hz), 4.45 (d, 1H, CH, *J* = 7.6 Hz).

¹³C NMR (DMSO-*d*₆) (δ, ppm): δ 169.3, 166.9, 161.5, 156.2, 155.5, 138.3, 133.9, 131.7, 131.3, 130.4, 128.6, 127.6, 120.3, 92.0, 53.8, 37.3;

Anal. calcd for C₂₀H₁₆BrN₃O₃: C, 52.88; H, 3.55; N, 15.42. Found: C, 52.69; H, 3.41; N, 15.63.

***N*-(2-amino-5-(2,4-dichlorophenyl)-3,4,5,6,7,8-hexahydro-4,7-dioxopyrido[2,3-*d*]pyrimidin-6-yl)benzamide (10d)**

IR (KBr *v*, cm⁻¹): 3433, 3335, 3207, 2902, 1706, 16542, 1522, 1460, 1374, 812, 717, 591 cm⁻¹;

¹H NMR (DMSO-*d*₆) (δ, ppm): δ = 10.74 (s, 1H, NH), 10.58 (s, 1H, NH), 8.35 (d, 1H, NH, *J* = 8.0 Hz), 7.77 (d, 2H, ArH, *J* = 6.8 Hz), 7.51–7.30 (m, 5H, ArH), 7.00 (s, 1H, ArH), 6.69 (brs, 2H, NH₂), 5.32 (t, 1H, CH, *J* = 7.6 Hz), 4.87 (d, 1H, CH, *J* = 8.0 Hz);

¹³C NMR (DMSO-*d*₆) (δ, ppm): δ 169.1, 167.3, 161.3, 156.8, 155.7, 139.5, 133.5, 132.0, 131.4, 131.2, 128.8, 128.4, 128.2, 127.9, 127.7, 92.0, 53.0, 34.9;

Anal. calcd for C₂₀H₁₅Cl₂N₃O₃: C, 54.07; H, 3.40; N, 15.76. Found: C, 53.93; H, 3.61; N, 15.88.

***N*-(2-amino-5-(3,4-dichlorophenyl)-3,4,5,6,7,8-hexahydro-4,7-dioxopyrido[2,3-*d*]pyrimidin-6-yl)benzamide (10e)**

IR (KBr *v*, cm⁻¹): 3463, 3338, 3201, 2904, 1698, 1650, 1518, 1485, 1372, 794, 702, 593 cm⁻¹;

¹H NMR (DMSO-*d*₆) (δ, ppm): δ = 10.76 (s, 1H, NH), 10.61 (s, 1H, NH), 8.15 (d, 1H, NH, *J* = 6.4 Hz), 7.79 (d, 2H, ArH, *J* = 7.2 Hz), 7.56–7.43 (m, 4H, ArH), 7.11 (s, 1H, ArH), 6.96–6.93 (m, 1H, ArH), 6.70 (brs, 2H, NH₂), 5.14 (t, 1H, CH, *J* = 7.2 Hz), 4.43 (d, 1H, CH, *J* = 7.6 Hz);

¹³C NMR (DMSO-*d*₆) (δ, ppm): δ 169.1, 167.0, 161.4, 156.4, 155.6, 140.2, 134.5, 133.8, 131.0, 130.7, 130.0, 129.8, 128.7, 128.5, 127.7, 92.0, 53.6, 37.3;

Anal. calcd for C₂₀H₁₅Cl₂N₃O₃: C, 54.07; H, 3.40; N, 15.76. Found: C, 53.95; H, 3.58; N, 15.86.

***N*-(2-amino-5-(4-nitrophenyl)-3,4,5,6,7,8-hexahydro-4,7-dioxopyrido[2,3-*d*]pyrimidin-6-yl)benzamide (10f)**

IR (KBr ν , cm^{-1}): 3432, 33369, 3199, 2900, 1705, 1648, 1519, 1487, 1349, 842, 792, 694 cm^{-1} ;

^1H NMR (DMSO- d_6) (δ , ppm): δ = 10.76 (s, 1H, NH), 10.67 (s, 1H, NH), 8.12 (d, 1H, NH, J = 8.8 Hz), 8.03 (d, 2H, ArH, J = 6.4 Hz), 7.76 (d, 2H, ArH, J = 8.0 Hz), 7.55–7.42 (m, 3H, ArH), 7.25 (d, 2H, ArH, J = 8.8 Hz), 6.64 (brs, 2H, NH_2), 5.19 (t, 1H, CH, J = 7.2 Hz), 4.59 (d, 1H, CH, J = 8.0 Hz);

^{13}C NMR (DMSO- d_6) (δ , ppm): δ 169.1, 167.0, 161.5, 156.4, 155.6, 147.3, 146.8, 133.8, 131.8, 129.6, 128.5, 127.6, 123.7, 91.3, 53.5, 38.0;

Anal. calcd for $\text{C}_{20}\text{H}_{16}\text{N}_6\text{O}_5$: C, 57.14; H, 3.84; N, 19.99. Found: C, 56.98; H, 3.67; N, 19.79.

***N*-(2-amino-5-(3,4-dimethoxyphenyl)-3,4,5,6,7,8-hexahydro-4,7-dioxopyrido[2,3-*d*]pyrimidin-6-yl)benzamide (10g)**

IR (KBr ν , cm^{-1}): 3428, 3338, 3205, 2934, 1698, 1648, 1517, 1486, 1375, 792, 713, 662 cm^{-1} ;

^1H NMR (DMSO- d_6) (δ , ppm): δ = 10.70 (s, 1H, NH), 10.53 (s, 1H, NH), 7.78 (d, 1H, NH, J = 7.2 Hz), 7.72–7.50 (m, 2H, ArH), 7.45 (s, 1H, ArH), 7.40–6.80 (m, 5H, ArH), 6.52 (brs, 2H, NH_2), 5.01 (t, 1H, CH, J = 7.6 Hz), 4.47 (d, 1H, CH, J = 7.6 Hz), 3.68 (s, 3H, OCH_3), 3.48 (s, 3H, OCH_3);

^{13}C NMR (DMSO- d_6) (δ , ppm): δ 169.5, 166.4, 161.5, 158.1, 157.3, 156.2, 155.6, 134.3, 131.6, 131.4, 130.6, 129.4, 128.9, 127.1, 114.2, 93.0, 65.6, 55.4, 54.1, 37.4;

Anal. calcd for $\text{C}_{22}\text{H}_{21}\text{N}_5\text{O}_5$: C, 60.68; H, 4.86; N, 16.08. Found: C, 60.45; H, 4.71; N, 15.96.

***N*-(2-amino-5-(4-methoxyphenyl)-3,4,5,6,7,8-hexahydro-4,7-dioxopyrido[2,3-*d*]pyrimidin-6-yl)benzamide (10h)**

IR (KBr ν , cm^{-1}): 3420, 3348, 3197, 2902, 1699, 1648, 1509, 1487, 1376, 830, 762, 642 cm^{-1} ;

^1H NMR (DMSO- d_6) (δ , ppm): δ = 10.69 (s, 1H, NH), 10.54 (s, 1H, NH), 7.73 (d, 1H, NH, J = 7.2 Hz), 7.65 (d, 2H, ArH, J = 6.0 Hz), 7.54–7.43 (m, 3H, ArH), 6.89 (d, 2H, ArH, J = 4.4 Hz), 6.79 (d, 2H, ArH, J = 8.8 Hz), 6.68 (brs, 2H, NH_2), 5.00 (t, 1H, CH, J = 6.8 Hz), 4.46 (d, 1H, CH, J = 7.2 Hz), 3.68 (s, 3H, OCH_3);

^{13}C NMR (DMSO- d_6) (δ , ppm): δ 169.6, 166.7, 161.5, 158.4, 156.0, 155.3, 134.0, 131.8, 130.5, 129.2, 128.6, 127.4, 113.9, 92.8, 55.2, 54.2, 36.7;

Anal. calcd for $\text{C}_{21}\text{H}_{19}\text{N}_5\text{O}_4$: C, 62.22; H, 4.72; N, 17.27. Found: C, 62.03; H, 4.88; N, 17.48.

***N*-(2-amino-5-(4-hydroxy-3-nitrophenyl)-3,4,5,6,7,8-hexahydro-4,7-dioxopyrido[2,3-*d*]pyrimidin-6-yl)benzamide (10i)**

IR (KBr ν , cm^{-1}): 3423, 3332, 3211, 2974, 1701, 1666, 1537, 1489, 1381, 823, 798, 698 cm^{-1} ;

^1H NMR (DMSO- d_6) (δ , ppm): δ = 10.82 (s, 1H, OH), 10.73 (s, 1H, NH), 10.58 (s, 1H, NH), 8.04 (d, 1H, NH, J = 6.0 Hz), 7.78 (d, 2H, ArH, J = 8.0 Hz), 7.53–7.42 (m, 4H, ArH), 7.13 (d, 1H, ArH, J = 8.4 Hz), 7.01 (d, 1H, ArH, J = 8.4 Hz), 6.67 (brs, 2H, NH_2), 5.10 (t, 1H, CH, J = 6.8 Hz), 4.42 (d, 1H, CH, J = 7.2 Hz);

^{13}C NMR (DMSO- d_6) (δ , ppm): δ 169.2, 167.0, 161.5, 156.3, 155.6, 151.7, 136.4, 135.6, 134.0, 131.7, 130.0, 128.5, 127.6, 124.2, 119.4, 91.6, 53.8, 36.8;

Anal. calcd for $\text{C}_{20}\text{H}_{16}\text{N}_6\text{O}_6$: C, 55.05; H, 3.70; N, 19.26. Found: C, 54.92; H, 3.60; N, 19.58.