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SUPPLEMENTARY MATERIAL TO

Synthesis of modified pyridine and bipyridine substituted coumarins as potent antimicrobial agents

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GENERAL PROCEDURE FOR THE SYNTHESIS OF **3a–e**

The derivatives **3a**, **3c** and **3d** were prepared according to a literature procedure.[1] The preparation of derivatives **3b** and **3e** is given below.

A mixture of 8-acetyl-7-hydroxy-4-methylcoumarin (0.01 mol) and appropriate aromatic aldehyde (0.012 mol) in ethanol (20–25 mL) containing a catalytic amount of piperidine was refluxed for half an hour to two hours. On concentrating the reaction mixture to a small volume, the chalcone separated out and was filtered and crystallized from chloroform–hexane.

*7-Hydroxy-4-methyl-8-(3-*p*-tolylacryloyl)chromen-2-one (**3b**)*. Yield: 71 %; m.p.: 156 °C; Anal. Calcd. for $C_{20}H_{16}O_4$: C, 74.99; H, 5.03 %. Found: C, 74.76; H, 5.05 %; IR (KBr, cm^{-1}): 3425 (O–H stretching), 3062 (aromatic C–H stretching), 1723 (C=O δ-lactone stretching), 1655 (C=O α,β-unsaturated carbonyl group), 1612 (aromatic C=C stretchings), 838 (C–H bending for *p*-di-substituted benzene ring); ^{1}H-NMR (400 MHz, $CDCl_3$, δ / ppm): 2.45 (6H, *s*, 2×CH_3), 6.21 (1H, *s*, C_3–H), 6.96–8.26 (8H, *m*, 6 Ar-H + 2 olefinic protons), 13.89 (1H, *s*, OH).

*7-Hydroxy-4-methyl-8-[3-(4-chlorophenyl)acryloyl]chromen-2-one (**3e**)*. Yield: 75 %; m.p.: 197–198 °C; Anal. Calcd. for $C_{19}H_{13}ClO_4$: C, 66.97; H, 3.85 %. Found: C, 66.72; H, 3.84 %; IR (KBr, cm^{-1}): 3419 (O–H stretching), 3061 (aromatic C–H stretching), 1720 (C=O δ-lactone stretching), 1657 (C=O α,β-unsaturated carbonyl group), 1610 (aromatic C=C stretchings), 836 (C–H bending for *p*-disubstituted benzene ring); ^{1}H-NMR (400 MHz, $CDCl_3$, δ / ppm): 2.46 (3H, *s*, CH_3), 6.22 (1H, *s*, C_3–H), 6.97–8.26 (8H, *m*, 6 Ar-H + 2 olefinic protons), 13.75 (1H, *s*, OH).

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ANALYTICAL AND SPECTRAL DATA FOR THE TARGET COMPOUNDS **5a**–**y**

*7-Hydroxy-4-methyl-8-(4-phenyl-6,7-dihydro-5*H*-cyclopenta[*b*]pyridin-2-yl)-chromen-2-one (**5a**)*. Yield: 66 %; m.p.: 181–182 °C; Anal. Calcd. for $C_{24}H_{19}NO_3$: C, 78.03; H, 5.18; N, 3.79 %. Found: C, 78.21; H, 5.12; N, 3.73 %; IR (KBr, cm^{-1}): 3417 (O–H stretching), 3056 (aromatic C–H stretching), 2989 (aliphatic C–H stretching), 1725 (C=O δ-lactone stretching), 1600 and 1555 (aromatic C=C and C=N stretchings), 705 and 723 (C–H out of plane bending for mono substituted benzene ring); ^{1}H-NMR (400 MHz, $CDCl_3$, δ / ppm): 2.22 (2H, *m*, C_6'–H), 2.45 (3H, *s*, CH_3), 3.17 (4H, *m*, C_5'–H and C_7'–H), 6.16 (1H, *s*, C_3–H), 6.99–7.70 (8H, *m*, 7 Ar-H + OH), 8.64 (1H, *s*, C_3'–H); ^{13}C-NMR (100 MHz, $CDCl_3$, δ / ppm): 19.23 (CH_3), 23.48 (CH_2), 31.02 (CH_2), 34.09 (CH_2), 108.42 (C), 110.37 (CH), 112.17 (C), 115.25 (CH), 123.00 (CH), 125.91 (CH), 128.43 (CH), 128.72 (CH), 128.83 (CH), 134.16 (C), 138.37 (C), 147.60 (C), 151.97 (C), 153.15 (C), 153.45 (C), 160.53 (C), 162.22 (C) and 163.90 (CO of coumarin).

*7-Hydroxy-4-methyl-8-(4-*p*-tolyl-6,7-dihydro-5*H*-cyclopenta[*b*]pyridin-2-yl)-chromen-2-one (**5b**)*. Yield: 72 %; m.p.: 201 °C; Anal. Calcd. for $C_{25}H_{21}NO_3$: C, 78.31; H, 5.52; N, 3.65 %. Found: C, 78.11; H, 5.56; N, 3.63 %; IR (KBr, cm^{-1}): 3422 (O–H stretching), 3022 (aromatic C–H stretching), 2953 (aliphatic C–H stretching), 1723 (C=O δ-lactone stretching), 1598 and 1550 (aromatic C=C and C=N stretchings), 818 (C–H bending of *p*-disubstituted benzene ring); ^{1}H-NMR (400 MHz, $CDCl_3$, δ / ppm): 2.22 (2H, *m*, C_6'–H), 2.45 (6H, *s*, 2 × CH_3), 3.16 (4H, *m*, C_5'–H and C_7'–H), 6.14 (1H, *s*, C_3–H), 6.99–7.58 (7H, *m*, 6 Ar-H + OH), 8.65 (1H, *s*, C_3'–H); ^{13}C-NMR (100 MHz, $CDCl_3$, δ / ppm): 19.26 (CH_3), 21.31 (CH_3), 23.55 (CH_2), 31.12 (CH_2), 34.10 (CH_2), 108.37 (C), 110.37 (CH), 112.17 (C), 115.31 (CH), 122.91 (CH), 125.89 (CH), 128.35 (CH), 129.58 (CH), 134.06 (C), 135.48 (C), 138.84 (C), 147.69 (C), 151.87 (C), 153.49 (C), 158.13 (C), 160.59 (C), 162.07 (C), 164.09 (CO of coumarin).

*7-Hydroxy-8-[4-(4-methoxyphenyl)-6,7-dihydro-5*H*-cyclopenta[*b*]pyridin-2--yl]-4-methyl-chromen-2-one (**5c**)*. Yield: 68 %; m.p.: 203 °C; Anal. Calcd. for $C_{25}H_{21}NO_4$: C, 75.17; H, 5.30; N, 3.51 %. Found: C, 75.32; H, 5.24; N, 3.54 %; IR (KBr, cm^{-1}): 3422 (O–H stretching), 3053 (aromatic C–H stretching), 2957 (aliphatic C–H stretching), 1716 (C=O δ-lactone stretching), 1597 and 1552 (aromatic C=C and C=N stretchings), 836 (C–H bending of *p*-disubstituted benzene ring); ^{1}H-NMR (400 MHz, $CDCl_3$, δ / ppm): 2.32 (2H, *m*, C_6'–H), 2.44 (3H, *s*, CH_3), 3.22 (2H, *t*, J = 7.2 Hz, C_5'–H), 3.50 (2H, *t*, poorly resolved triplet, C_7'–H), 3.90 (3H, *s*, OCH_3), 6.14 (1H, *s*, C_3–H), 7.06–7.86 (8H, *m*, 7 Ar-H + OH); ^{13}C-NMR (100 MHz, $CDCl_3$, δ / ppm): 19.22 (CH_3), 23.67 (CH_2), 31.23 (CH_2), 33.39 (CH_2), 55.43 (OCH_3), 110.64 (CH), 114.46 (CH), 115.41 (CH), 121.73 (C), 123.76 (CH), 126.44 (C), 127.47 (CH), 128.53 (C), 128.97

(C), 129.80 (C), 130.04 (CH), 135.11 (C), 149.06 (C), 152.92 (C), 153.34 (C), 160.46 (C), 160.62 (C), 161.43 (CO of coumarin).

*8-[4-(3,4-Dimethoxyphenyl)-6,7-dihydro-5*H*-cyclopenta[*b*]pyridine-2-yl]-7--hydroxy-4-methyl-chromen-2-one (**5d**)*. Yield: 70 %; m.p.: 164 °C; Anal. Calcd. for $C_{26}H_{23}NO_5$: C, 72.71; H, 5.40; N, 3.26 %. Found: C, 72.90; H, 5.34; N, 3.27 %; IR (KBr, cm^{-1}): 3422 (O–H stretching), 3076 (aromatic C–H stretching), 2962 (aliphatic C–H stretching), 1723 (C=O δ-lactone stretching), 1600 and 1552 (aromatic C=C and C=N stretchings); ^{1}H-NMR (400 MHz, $CDCl_3$, δ / ppm): 2.23 (2H, *m*, C_6'–H), 2.46 (3H, *s*, CH_3), 3.18 (4H, *m*, C_5'–H and C_7'–H), 3.97 and 4.07 (6H, 2 × *s*, 2 × OCH_3), 6.14 (1H, *s*, C_3–H), 6.99–7.54 (6H, *m*, 5 Ar-H + 1 OH), 8.72 (1H, *s*, C_3'–H); ^{13}C-NMR (100 MHz, $CDCl_3$, δ / ppm): 19.20 (CH_3), 23.36 (CH_2), 31.01 (CH_2), 34.15 (CH_2), 55.96 (OCH_3), 56.11 (OCH_3), 110.26 (CH), 112.21 (C), 115.35 (CH), 123.01 (CH), 125.62 (CH), 128.31 (CH), 128.72 (CH), 128.83 (CH), 134.06 (C), 138.23 (C), 147.50 (C), 151.76 (C), 153.15 (C), 153.37 (C), 154.26 (C), 158.76 (C), 160.52 (C), 162.22 (C), 163.70 (CO of coumarin).

*8-[4-(4-Chlorophenyl)-6,7-dihydro-5*H*-cyclopenta[*b*]pyridin-2-yl]-7-hyd-roxy-4-methyl-chromen-2-one (**5e**)*. Yield: 70 %; m.p.: 198–199 °C; Anal. Calcd. for $C_{24}H_{18}ClNO_3$: C, 71.38; H, 4.49; N, 3.47 %. Found: C, 71.19; H, 4.52; N, 3.51 %; IR (KBr, cm^{-1}): 3433 (O–H stretching), 3068 (aromatic C–H stretching), 2961 (aliphatic C–H stretching), 1727 (C=O δ-lactone stretching), 1599 and 1549 (aromatic C=C and C=N stretchings), 837 (C–H bending of *p*-disubstituted benzene ring); ^{1}H-NMR (400 MHz, $CDCl_3$, δ / ppm): 2.23 (2H, *m*, C_6'–H), 2.45 (3H, *s*, CH_3), 3.15 (4H, *m*, C_5'–H and C_7'–H), 6.15 (1H, *s*, C_3–H), 6.99–7.61 (7H, *m*, 6 Ar-H + OH), 8.62 (1H, *s*, C_3'–H); ^{13}C-NMR (100 MHz, $CDCl_3$, δ / ppm): 19.24 (CH_3), 23.52 (CH_2), 31.11 (CH_2), 34.15 (C_7'), 108.19 (C), 110.17 (CH), 112.10 (C), 115.15 (CH), 122.71 (CH), 125.89 (CH), 128.53 (CH), 129.85 (CH), 134.06 (C), 135.38 (C), 138.94 (C), 147.39 (C), 151.78 (C), 153.94 (C), 158.31 (C), 160.45 (C), 162.07 (C), 164.12 (CO of coumarin).

*7-Hydroxy-4-methyl-8-(4-phenyl-5,6,7,8-tetrahydroquinolin-2-yl)chromen--2-one (**5f**)*. Yield: 67 %; m.p.: 200 °C; Anal. Calcd. for $C_{25}H_{21}NO_3$: C, 78.31; H, 5.52; N, 3.65 %. Found: C, 78.20; H, 5.55; N, 3.61 %; IR (KBr, cm^{-1}): 3405 (O–H stretching), 3059 (aromatic C–H stretching), 2929 (aliphatic C–H stretching), 1717 (C=O δ-lactone stretching), 1600 and 1539 (aromatic C=C and C=N stretchings), 704 and 725 (C–H out of plane bending for mono substituted benzene ring); ^{1}H-NMR (400 MHz, $CDCl_3$, δ / ppm): 1.80 (2H, *m*, C_6'–H), 1.99 (2H, *m*, C_7'–H), 2.43 (3H, *s*, CH_3), 2.73 (2H, *m*, C_5'–H), 3.08 (2H, *t*, J = 6.4 Hz, C_8'–H), 6.10 (1H, *s*, C_3–H), 6.97–7.53 (8H, *m*, 7 Ar-H + OH), 8.50 (1H, *s*, C_3'–H); ^{13}C-NMR (100 MHz, $CDCl_3$, δ / ppm): 19.22 (CH_3), 22.55 (CH_2), 22.79 (CH_2), 27.27 (CH_2), 31.84 (CH_2), 107.44 (C), 110.15 (CH), 111.83 (C), 115.45 (CH), 124.08 (CH), 126.09 (CH), 128.23 (CH), 128.55 (CH), 128.66

(CH), 129.57 (C), 131.91 (C), 138.88 (C), 150.70 (C), 152.20 (C), 153.14 (C), 153.46 (C), 160.52 (C), 164.90 (CO of coumarin).

*7-Hydroxy-4-methyl-8-(4-*p*-tolyl-5,6,7,8-tetrahydroquinolin-2-yl)chromen--2-one (**5g**)*. Yield: 62 %; m.p.: 218 °C; Anal. Calcd. for $C_{26}H_{23}NO_3$: C, 78.57; H, 5.83; N, 3.52 %. Found: C, 78.69; H, 5.85; N, 3.48 %; IR (KBr, cm^{-1}): 3433 (O–H stretching), 3045 (aromatic C–H stretching), 2936 (aliphatic C–H stretching), 1731 (C=O δ-lactone stretching), 1603 and 1538 (aromatic C=C and C=N stretchings), 837 (C–H bending of *p*-disubstituted benzene ring); ^{1}H-NMR (400 MHz, $CDCl_3$, δ / ppm): 1.82 (2H, *m*, C_6'–H), 1.97 (2H, *m*, C_7'–H), 2.44 (6H, *s*, 2×CH_3), 2.75 (2H, *t*, *J* = 6.4 Hz, C_5'–H), 3.08 (2H, *t*, *J* = 7.2 Hz, C_8'–H), 6.10 (1H, *s*, C_3–H), 6.97–7.53 (7H, *m*, 6 Ar-H + OH), 8.50 (1H, *s*, C_3'–H); ^{13}C-NMR (100 MHz, $CDCl_3$, δ / ppm): 19.24 (CH_3), 21.25 (CH_3), 22.53 (CH_2), 22.82 (CH_2), 27.34 (CH_2), 31.79 (CH_2), 107.43 (C), 110.14 (CH), 111.82 (C), 115.48 (CH), 124.19 (CH), 126.04 (CH), 128.60 (CH), 129.24 (CH), 129.65 (C), 135.89 (C), 138.11 (C), 150.54 (C), 152.30 (C), 153.00 (C), 153.44 (C), 153.47 (C), 160.51 (C), 164.93 (CO of coumarin).

*7-Hydroxy-4-methyl-8-[4-(4-methoxyphenyl)-5,6,7,8-tetrahydroquinolin-2-yl]--4-methyl-chromen-2-one (**5h**)*. Yield: 69 %; m.p.: 192–193 °C; Anal. Calcd. for $C_{26}H_{23}NO_4$: C, 75.53; H, 5.61; N, 3.39 %. Found: C, 75.30; H, 5.63; N, 3.34 %; IR (KBr, cm^{-1}): 3420 (O–H stretching), 3050 (aromatic C–H stretching), 2927 (aliphatic C–H stretching), 1720 (C=O δ-lactone stretching), 1594 and 1538 (aromatic C=C and C=N stretchings), 835 (C–H bending of *p*-disubstituted benzene ring); ^{1}H-NMR (400 MHz, $CDCl_3$, δ / ppm): 1.81 (2H, *m*, C_6'–H), 1.98 (2H, *m*, C_7'–H), 2.44 (3H, *s*, CH_3), 2.76 (2H, *t*, *J* = 6.4 Hz, C_5'–H), 3.07 (2H, *t*, *J* = 7.2 Hz, C_8'–H), 3.89 (3H, *s*, OCH_3) 6.11 (1H, *s*, C_3–H), 6.97–7.53 (7H, *m*, 6 Ar-H + OH), 8.51 (1H, *s*, C_3'–H); ^{13}C-NMR (100 MHz, $CDCl_3$, δ / ppm): 19.29 (CH_3), 22.61 (CH_2), 22.91 (CH_2), 27.48 (CH_2), 31.84 (CH_2), 55.36 (OCH_3), 107.47 (C), 110.12 (CH), 111.79 (C), 114.05 (CH), 115.48 (CH), 124.19 (CH), 126.05 (CH), 129.62 (C), 130.11 (CH), 131.17 (C), 150.70 (C), 151.83 (C), 152.81 (C), 153.08 (C), 153.48 (C), 159.69 (C), 160.61 (C), 165.06 (CO of coumarin).

*8-[4-(3,4-Dimethoxyphenyl)-5,6,7,8-tetrahydroquinolin-2-yl]-7-hydroxy-4--methyl-chromen-2-one (**5i**)*. Yield: 70 %; m.p.: 221 °C; Anal. Calcd. for $C_{27}H_{25}NO_5$: C, 73.12; H, 5.68; N, 3.16 %. Found: C, 73.29; H, 5.62; N, 3.12 %; IR (KBr, cm^{-1}): 3415 (O–H stretching), 3068 (aromatic C–H stretching), 2932 (aliphatic C–H stretching), 1719 (C=O δ-lactone stretching), 1598 and 1539 (aromatic C=C and C=N stretchings); ^{1}H-NMR (400 MHz, $CDCl_3$, δ / ppm): 1.82 (2H, *m*, C_6'–H), 1.99 (2H, *m*, C_7'–H), 2.44 (3H, *s*, CH_3), 2.82 (2H, *t*, *J* = 6.4 Hz, C_5'–H), 3.08 (2H, *t*, *J* = 6.4 Hz, C_8'–H), 3.96 and 4.01 (6H, 2 × *s*, 2 × OCH_3), 6.10 (1H, *s*, C_3–H), 6.97–7.53 (6H, *m*, 5 Ar-H + OH), 8.54 (1H, *s*, C_3'–H); ^{13}C-NMR (100 MHz, $CDCl_3$, δ / ppm): 19.26 (CH_3), 22.58 (CH_2), 22.95 (CH_2), 27.59

(CH_2), 31.92 (CH_2), 55.99 (OCH_3), 56.14 (OCH_3), 110.12 (CH), 110.98 (CH), 111.84 (C), 112.53 (CH), 115.43 (CH), 121.32 (CH), 124.30 (CH), 126.07 (CH), 129.46 (C), 131.40 (C), 149.14 (C), 149.31 (C), 150.66 (C), 151.30 (C), 151.67 (C), 153.29 (C), 153.41 (C), 153.56 (C), 160.54 (C), 164.84 (CO of coumarin).

*8-[4-(4-Chlorophenyl)-5,6,7,8-tetrahydroquinolin-2-yl]-7-hydroxy-4-methyl--chromen-2-one (**5j**)*. Yield: 68 %; m.p.: 251–252 °C; Anal. Calcd. for $C_{25}H_{20}ClNO_3$: C, 71.85; H, 4.82; N, 3.35 %. Found: C, 71.98; H, 4.84; N, 3.30 %; IR (KBr, cm^{-1}): 3423 (O–H stretching), 3058 (aromatic C–H stretching), 2928 (aliphatic C–H stretching), 1732 (C=O δ-lactone stretching), 1602 and 1538 (aromatic C=C and C=N stretchings), 830 (C–H bending of *p*-disubstituted benzene ring); ^{1}H-NMR (400 MHz, $CDCl_3$, δ / ppm): 1.81 (2H, *m*, C_6'–H), 1.98 (2H, *m*, C_7'–H), 2.43 (3H, *s*, CH_3), 2.70 (2H, *t*, J = 6.4 Hz, C_5'–H), 3.07 (2H, *t*, J = 6.4 Hz, C_8'–H), 6.11 (1H, *s*, C_3–H), 6.96–7.53 (7H, *m*, 6 Ar-H + OH), 8.48 (1H, *s*, C_3'–H); ^{13}C-NMR (100 MHz, $CDCl_3$, δ / ppm): 19.24 (CH_3), 22.50 (CH_2), 22.74 (CH_2), 27.25 (CH_2), 31.90 (CH_2), 107.31 (C), 110.17 (CH), 111.85 (C), 115.43 (CH), 123.91 (CH), 126.18 (CH), 128.84 (CH), 129.37 (C), 130.06 (CH), 134.44 (C), 137.26 (C), 150.82 (C), 150.89 (C), 153.43 (C), 153.50 (C), 160.47 (C), 164.79 (CO of coumarin).

*7-Hydroxy-4-methyl-8-(4-phenyl-5*H*-indeno[1,2-*b*]pyridin-2-yl)chromen-2--one (**5k**)*. Yield: 72 %; m.p.: 264–265 °C; Anal. Calcd. for $C_{28}H_{19}NO_3$: C, 80.56; H, 4.59; N, 3.36 %. Found: C, 80.77; H, 4.53; N, 3.40 %; IR (KBr, cm^{-1}): 3423 (O–H stretching), 3057 (aromatic C–H stretching), 2986 (aliphatic C–H stretching), 1729 (C=O δ-lactone stretching), 1599 and 1551 (aromatic C=C and C=N stretchings), 694 and 722 (C–H out of plane bending for mono substituted benzene ring); ^{1}H-NMR (400 MHz, $CDCl_3$, δ / ppm): 2.45 (3H, *s*, CH_3), 4.09 (2H, *s*, C_5'–H), 6.14 (1H, *s*, C_3–H), 7.03–8.07 (11H, *m*, Ar-H), 8.73 (1H, *s*, C_3'–H), 15.90 (1H, *bs*, OH); ^{13}C-NMR (100 MHz, $CDCl_3$, δ / ppm): 19.25 (CH_3), 34.96 (CH_2), 108.55 (C), 110.46 (CH), 112.26 (C), 115.30 (CH), 120.97 (CH), 123.18 (CH), 125.21 (CH), 126.20 (CH), 127.51 (CH), 128.50 (CH), 129.03 (CH), 129.07 (CH), 129.48 (CH), 133.37 (C), 138.12 (C), 139.12 (C), 144.16 (C), 147.99 (C), 152.80 (C), 153.25 (C), 153.44 (C), 157.03 (C), 160.46 (C), 163.89 (CO of coumarin).

*7-Hydroxy-4-methyl-8-(4-*p*-tolyl-5*H*-indeno[1,2-*b*]pyridin-2-yl)chromen-2--one (**5l**)*. Yield: 75 %; m.p.: 280 °C; Anal. Calcd. for $C_{29}H_{21}NO_3$: C, 80.72; H, 4.91; N, 3.25 %. Found: C, 80.90; H, 4.96; N, 3.24 %; IR (KBr, cm^{-1}): 3410 (O–H stretching), 3042 (aromatic C–H stretching), 2920 (aliphatic C–H stretching), 1716 (C=O δ-lactone stretching), 1600 and 1553 (aromatic C=C and C=N stretchings), 834 (C–H bending of *p*-disubstituted benzene ring); ^{1}H-NMR (400 MHz, $CDCl_3$, δ / ppm): 2.46 and 2.47 (6H, 2×*s*, 2×CH_3), 4.11 (2H, *s*, C_5'–H), 6.16 (1H, *s*, C_3–H), 7.05–8.09 (10H, *m*, Ar-H), 8.75 (1H, *s*, C_3'–H), 15.97 (1H, *bs*, OH); ^{13}C-NMR (100 MHz, $CDCl_3$, δ /ppm): 19.28 (CH_3), 21.35 (CH_3),

35.06 (CH_2), 108.65 (C), 110.48 (CH), 112.30 (C), 115.30 (CH), 120.98 (CH), 123.13 (CH), 125.21 (CH), 126.20 (CH), 127.52 (CH), 128.40 (CH), 129.42 (CH), 129.79 (CH), 133.28 (C), 135.27 (C), 139.15 (C), 139.22 (C), 144.22 (C), 148.06 (C), 152.75 (C), 153.47 (C), 157.07 (C), 160.52 (C), 163.92 (C), 169.77 (CO of coumarin).

*7-Hydroxy-8-[4-(4-methoxyphenyl)-5*H*-indeno[1,2-*b*]pyridin-2-yl]-4-methyl--chromen-2-one (**5m**)*. Yield: 71 %; m.p.: 246 °C; Anal. Calcd. for $C_{29}H_{21}NO_4$: C, 77.84; H, 4.73; N, 3.13 %. Found: C, 77.70; H, 4.75; N, 3.16 %; IR (KBr, cm^{-1}): 3418 (O–H stretching), 3054 (aromatic C–H stretching), 2920 (aliphatic C–H stretching), 1721 (C=O δ-lactone stretching), 1598 and 1552 (aromatic C=C and C=N stretchings), 841 (C–H bending of *p*-disubstituted benzene ring); ^{1}H--NMR (400 MHz, $CDCl_3$, δ / ppm): 3.60 (3H, *s*, CH_3), 5.05 (3H, *s*, OCH_3), 5.25 (2H, *s*, C_5'–H), 7.29 (1H, *s*, C_3–H), 8.18–9.22 (10H, *m*, Ar-H), 9.88 (1H, *s*, C_3'–H), 17.15 (1H, *bs*, OH); ^{13}C-NMR (100 MHz, $CDCl_3$, δ / ppm): 19.28 (CH_3), 33.18 (CH_2), 55.42 (OCH_3), 108.59 (C), 110.44 (CH), 112.26 (C), 114.56 (CH), 115.33 (CH), 120.95 (CH), 122.85 (CH), 125.20 (CH), 126.15 (CH), 127.50 (CH), 129.40 (CH), 129.89 (CH), 130.43 (C), 132.96 (C), 139.21 (C), 144.14 (C), 147.53 (C), 152.68 (C), 153.26 (C), 153.51 (C), 156.96 (C), 160.36 (C), 160.53 (C), 163.96 (CO of coumarin).

*8-[4-(3,4-Dimethoxyphenyl)-5*H*-indeno[1,2-*b*]pyridin-2-yl]-7-hydroxy-4--methyl-chromen-2-one (**5n**)*. Yield: 73 %; m.p.: 257–258 °C; Anal. Calcd. for $C_{30}H_{23}NO_5$: C, 75.46; H, 4.85; N, 2.93 %. Found: C, 75.63; H, 4.89; N, 2.95 %; IR (KBr, cm^{-1}): 3425 (O–H stretching), 3083 (aromatic C–H stretching), 2918 (aliphatic C–H stretching), 1724 (C=O δ-lactone stretching), 1601 and 1553 (aromatic C=C and C=N stretchings); ^{1}H-NMR (400 MHz, $CDCl_3$, δ / ppm): 2.45 (3H, *s*, CH_3), 3.99 and 4.10 (6H, 2×*s*, 2×OCH_3), 4.12 (2H, *s*, C_5'–H), 6.13 (1H, *s*, C_3–H), 7.02–8.06 (9H, *m*, Ar-H), 8.78 (1H, *s*, C_3'–H), 15.98 (1H, *bs*, OH); ^{13}C-NMR (100 MHz, $CDCl_3$, δ / ppm): 19.31 (CH_3), 35.27 (CH_2), 55.92 (OCH_3), 56.34 (OCH_3), 104.01 (C), 108.47 (C), 110.31 (CH), 111.23 (CH), 111.84 (CH), 112.27 (C), 115.38 (CH), 117.98 (C), 120.86 (CH), 122.86 (CH), 125.20 (CH), 126.20 (CH), 127.47 (CH), 129.42 (CH), 130.61 (C), 132.80 (C), 139.09 (C), 144.16 (C), 147.39 (C), 149.53 (C), 149.92 (C), 152.76 (C), 153.60 (C), 157.06 (C), 160.59 (CO of coumarin).

*8-[4-(4-Chlorophenyl)-5*H*-indeno[1,2-b]pyridin-2-y])-7-hydroxy-4-methyl--chromen-2-one (**5o**)*. Yield: 70 %; m.p.: 285 °C; Anal. Calcd. for $C_{28}H_{18}ClNO_3$: C, 74.42; H, 4.01; N, 3.10 %. Found: C, 74.63; H, 4.05; N, 3.08 %; IR (KBr, cm^{-1}): 3405 (O–H stretching), 3055 (aromatic C–H stretching), 2920 (aliphatic C–H stretching), 1716 (C=O δ-lactone stretching), 1598 and 1551 (aromatic C=C and C=N stretchings), 839 (C–H bending of *p*-disubstituted benzene ring); ^{1}H--NMR (400 MHz, $CDCl_3$, δ / ppm): 2.48 (3H, *s*, CH_3), 4.10 (2H, *s*, C_5'–H), 6.18 (1H, *s*, C_3–H), 7.07–8.12 (10H, *m*, Ar-H), 8.73 (1H, *s*, C_3'–H), 15.72 (1H, *bs*,

OH); ^{13}C-NMR (100 MHz, $CDCl_3$, δ / ppm): 19.34 (CH_3), 35.28 (CH_2), 108.68 (C), 110.47 (CH), 112.33 (C), 114.66 (CH), 115.39 (CH), 121.08 (CH), 122.96 (CH), 125.36 (CH), 126.21 (CH), 127.53 (CH), 129.51 (CH), 130.06 (CH), 130.58 (C), 133.02 (C), 139.33 (C), 144.22 (C), 147.63 (C), 152.78 (C), 153.36 (C), 153.60 (C), 157.08 (C), 160.44 (C), 160.53 (C), 164.05 (CO of coumarin).

7-Hydroxy-4-methyl-8-(4-phenyl-[2,3′-bipyridin]-6′-yl)chromen-2-one ***(5p)***. Yield: 75 %; m.p.: 261 °C; Anal. Calcd. for $C_{26}H_{18}N_2O_3$: C, 76.83; H, 4.46; N, 6.89 %. Found: C, 76.62; H, 4.44; N, 6.91 %; IR (KBr, cm^{-1}): 3435 (O–H stretching), 3035 (aromatic C–H stretching), 2961 (aliphatic C–H stretching), 1732 (C=O δ-lactone stretching), 1608 and 1547 (aromatic C=C and C=N stretchings), 707 and 760 (C–H out of plane bending for mono-substituted benzene ring); ^{1}H-NMR (400 MHz, $CDCl_3$, δ / ppm): 2.48 (3H, *s*, CH_3), 6.20 (1H, *s*, C_3–H), 7.03–9.00 (12H, *m*, Ar-H), 9.25 (1H, poorly resolved doublet, C_2''–H), 15.48 (1H, *bs*, OH); ^{13}C-NMR (100 MHz, $CDCl_3$, δ / ppm): 19.29 (CH_3), 108.11 (C), 110.66 (CH), 112.42 (C), 115.24 (CH), 117.82 (CH), 123.34 (CH), 123.95 (CH), 126.92 (CH), 127.45 (CH), 129.46 (CH), 129.83 (CH), 133.70 (C), 134.33 (CH), 137.71 (C), 148.26 (CH), 150.69 (CH), 151.85 (C), 152.19 (C), 153.43 (C), 153.52(C), 154.77(C), 160.29(C) and 163.70 (CO of coumarin).

*7-Hydroxy-8-(4-*p*-tolyl-[2,3′-bipyridin]-6-yl)-4-methyl-chromen-2-one* ***(5q)***. Yield: 68 %; m.p.: 250 °C; Anal. Calcd. for $C_{27}H_{20}N_2O_3$: C, 77.13; H, 4.79; N, 6.66 %. Found: C, 77.40; H, 4.82; N, 6.62 %; IR (KBr, cm^{-1}): 3412 (O–H stretching), 3041 (aromatic C–H stretching), 2968 (aliphatic C–H stretching), 1720 (C=O δ-lactone stretching), 1607 and 1544 (aromatic C=C and C=N stretchings), 814 (C–H bending of *p*-disubstituted benzene ring; ^{1}H-NMR (400 MHz, $CDCl_3$, δ / ppm): 2.47 (6H, *s*, 2×CH_3), 6.20 (1H, *s*, C_3–H), 7.01–8.98 (11H, *m*, Ar-H), 9.23 (1H, poorly resolved *d*, C_2''–H), 15.58 (1H, *bs*, OH); ^{13}C-NMR (100 MHz, $CDCl_3$, δ / ppm): 19.30 (CH_3), 21.33 (CH_3), 108.10 (C), 110.63 (CH), 112.39 (C), 115.24 (CH), 117.51 (CH), 123.04 (CH), 123.94 (CH), 126.86 (CH), 127.29 (CH), 130.19 (CH), 133.76 (C), 134.33 (CH), 134.75 (C), 140.13 (C), 148.27 (CH), 150.64 (CH), 151.71 (C), 152.07 (C), 153.45 (C), 153.53 (C), 154.71 (C), 160.29 (C), 163.83 (CO of coumarin).

7-Hydroxy-8-[4-(4-methoxyphenyl)-[2,3′-bipyridin]-6-yl]-4-methyl-chromen--2-one ***(5r)***. Yield: 70 %; m.p.: 247–248 °C; Anal. Calcd. for $C_{27}H_{20}N_2O_4$: C, 74.30; H, 4.62; N, 6.42 %. Found: C, 74.44; H, 4.60; N, 6.48 %; IR (KBr, cm^{-1}): 3418 (O–H stretching), 3044 (aromatic C–H stretching), 2960 (aliphatic C–H stretching), 1727 (C=O δ-lactone stretching), 1605 and 1544 (aromatic C=C and C=N stretchings), 810 (C–H bending of *p*-disubstituted benzene ring; ^{1}H-NMR (400 MHz, $CDCl_3$, δ / ppm): 2.47 (3H, *s*, CH_3), 3.90 (3H, *s*, OCH_3), 6.19 (1H, *s*, C_3–H), 7.0–8.96 (11H, *m*, Ar-H), 9.22 (1H, poorly resolved *d*, C_2''–H), 15.67 (1H, *bs*, OH); ^{13}C-NMR (100 MHz, $CDCl_3$, δ / ppm): 19.27 (CH_3), 55.45 (OCH_3), 108.03 (C), 110.54 (CH), 112.31 (C), 114.90 (CH), 115.26 (CH),

117.03 (CH), 122.55 (CH), 123.91 (CH), 126.80 (CH), 128.70 (CH), 129.82 (C), 133.75 (C), 134.28 (CH), 148.22 (CH), 150.59 (CH), 151.18 (C), 151.95 (C), 153.44 (C), 153.54 (C), 154.59 (C), 160.30 (C), 161.18 (C), 163.89 (CO of coumarin).

*8-[4-(3,4-Dimethoxyphenyl)-[2,3′-bipyridin]-6-yl]-7-hydroxy-4-methyl-chromen-2-one (**5s**)*. Yield: 72 %; m.p.: 245 °C; Anal. Calcd. for $C_{28}H_{22}N_2O_5$: C, 72.09; H, 4.75; N, 6.01 %. Found: C, 71.82; H, 4.74; N, 6.03 %; IR (KBr, cm^{-1}): 3410 (O–H stretching), 3043 (aromatic C–H stretching), 2948 (aliphatic C–H stretching), 1724 (C=O δ-lactone stretching), 1604 and 1545 (aromatic C=C and C=N stretchings); ^{1}H-NMR (400 MHz, $CDCl_3$, δ / ppm): 2.48 (3H, *s*, CH_3), 3.99 and 4.11 (6H, 2×*s*, 2×OCH_3), 6.17 (1H, *s*, C_3–H), 7.01–9.06 (10H, *m*, Ar-H), 9.23 (1H, poorly resolved *d*, C_2″–H), 15.67 (1H, *bs*, OH); ^{13}C-NMR (100MHz, $CDCl_3$, δ / ppm): 19.29 (CH_3), 56.02 (OCH_3), 56.26 (OCH_3), 107.66 (C), 110.19 (CH), 110.35 (CH), 111.52 (CH), 112.21 (C), 115.29 (CH), 116.69 (CH), 119.70 (CH), 122.46 (CH), 123.88 (CH), 126.81 (CH), 129.91 (C), 133.60 (C), 134.20 (CH), 148.13 (CH), 149.78 (C), 150.53 (CH), 150.69 (C), 150.96 (C), 151.72 (C), 153.47 (C), 153.61 (C), 154.55 (C), 160.18 (C), 164.12 (CO of coumarin).

*8-[4-(4-Chlorophenyl)-[2,3′-bipyridin]-6-yl]7-hydroxy-4-methyl-chromen-2--one (**5t**)*. Yield: 60 %; m.p.: 270–271 °C; Anal. Calcd. for $C_{26}H_{17}ClN_2O_3$: C, 70.83; H, 3.89; N, 6.35 %. Found: C, 70.59; H, 3.86; N, 6.39 %; IR (KBr, cm^{-1}): 3440 (O–H stretching), 3059 (aromatic C–H stretching), 2958 (aliphatic C–H stretching), 1725 (C=O δ-lactone stretching), 1608 and 1545 (aromatic C=C and C=N stretchings), 810 (C–H bending of *p*-disubstituted benzene ring); ^{1}H-NMR (400 MHz, $CDCl_3$, δ / ppm): 2.49 (3H, *s*, CH_3), 6.21 (1H, *s*, C_3–H), 7.04–8.97 (11H, *m*, Ar-H), 9.24 (1H, poorly resolved *d*, C_2″–H), 15.34 (1H, *bs*, OH); ^{13}C-NMR (100 MHz, $CDCl_3$, δ / ppm): 19.30 (CH_3), 110.69 (CH), 112.41 (C), 115.27 (CH), 115.88 (C), 117.48 (CH), 123.14 (CH), 123.96 (CH), 124.77 (C), 125.49 (C), 126.05 (C), 127.05 (CH), 128.71 (CH), 129.71 (CH), 130.65 (C), 132.61 (C), 134.33 (CH), 136.14 (C), 136.23 (C), 147.19 (C), 148.25 (CH), 150.80 (CH), 153.55 (C), 154.95 (CO of coumarin).

*7-Hydroxy-4-methyl-8-(4-phenyl-[2,4′-bipyridin]-6-yl)chromen-2-one (**5u**)*. Yield: 73 %; m.p.: 248 °C; Anal. Calcd. for $C_{26}H_{18}N_2O_3$: C, 76.83; H, 4.46; N, 6.89 %. Found: C, 76.68; H, 4.43; N, 6.86 %; IR (KBr, cm^{-1}): 3417 (O–H stretching), 3064 (aromatic C–H stretching), 2960 (aliphatic C–H stretching), 1722 (C=O δ-lactone stretching), 1606 and 1543 (aromatic C=C and C=N stretchings), 704 and 765 (C–H out of plane bending for mono substituted benzene ring); ^{1}H-NMR (400 MHz, $CDCl_3$, δ / ppm): 2.47 (3H, *s*, CH_3), 6.19 (1H, *s*, C_3–H), 7.03–8.82 (12H, *m*, Ar-H), 9.02 (1H, poorly resolved *d*, C_3″–H), 15.40 (1H, *bs*, OH); ^{13}C-NMR (100 MHz, $CDCl_3$, δ / ppm): 19.29 (CH_3), 110.79 (CH), 112.48 (C), 115.69 (CH), 117.81 (CH), 120.86 (CH), 123.85 (CH), 128.76 (CH), 129.79 (CH), 135.91 (CH), 136.44 (CH), 144.85 (C), 150.62 (C),

150.94 (C), 150.96 (CH), 152.23 (C), 153.48 (C), 153.69 (C), 154.99 (C), 160.27 (C), 160.63 (C), 163.64 (CO of coumarin).

7-Hydroxy-4-methyl-8-(4-p-*tolyl-[2,4′-bipyridin]-6-yl)chromen-2-one* (***5v***). Yield: 65 %; m.p.: 260–261 °C; Anal. Calcd. for $C_{27}H_{20}N_2O_3$: C, 77.13; H, 4.79; N, 6.66 %. Found: C, 77.39; H, 4.81; N, 6.69 %; IR (KBr, cm^{-1}): 3415 (O–H stretching), 3030 (aromatic C–H stretching), 2961 (aliphatic C–H stretching), 1726 (C=O δ-lactone stretching), 1598 and 1542 (aromatic C=C and C=N stretchings), 812 (C–H bending of *p*-disubstituted benzene ring); ^{1}H-NMR (400 MHz, $CDCl_3$, δ / ppm): 2.47 (6H, *s*, 2×CH_3), 6.19 (1H, *s*, C_3–H), 7.02–8.82 (11H, *m*, Ar-H), 9.01 (1H, poorly resolved *d*, C_3''–H), 15.48 (1H, *bs*, OH); ^{13}C-NMR (100 MHz, $CDCl_3$, δ / ppm): 19.29 (CH_3), 21.35 (CH_3), 110.66 (CH), 112.43 (CH), 115.22 (CH), 117.68 (CH), 120.99 (CH), 123.87 (CH), 126.96 (CH), 127.25 (C), 130.23 (CH), 134.53 (C), 139.78 (C), 140.26 (C), 145.07 (C), 150.18 (C), 150.83 (CH), 151.81 (C), 151.92 (C), 153.39 (C), 153.53 (C), 154.71 (C), 160.25 (CO of coumarin).

7-Hydroxy-8-[4-(4-methoxyphenyl)-[2,4′-bipyridin]-6-yl]-4-methyl-chromen--2-one (***5w***). Yield: 78 %; m.p.: 240–241 °C; Anal. Calcd. for $C_{27}H_{20}N_2O_4$: C, 74.30; H, 4.62; N, 6.42 %. Found: C, 74.54; H, 4.58; N, 6.39 %; IR (KBr, cm^{-1}): 3417 (O–H stretching), 3032 (aromatic C–H stretching), 2963 (aliphatic C–H stretching), 1730 (C=O δ-lactone stretching), 1602 and 1542 (aromatic C=C and C=N stretchings), 818 (C–H bending of *p*-disubstituted benzene ring); ^{1}H--NMR (400 MHz, $CDCl_3$, δ / ppm): 3.08 (3H, *s*, CH_3), 3.91 (3H, *s*, OCH_3), 6.21 (1H, *s*, C_3–H), 7.04–8.83 (11H, *m*, Ar-H), 9.02 (1H, poorly resolved *d*, C_3''–H), 15.59 (1H, *bs*, OH); ^{13}C-NMR (100 MHz, $CDCl_3$, δ / ppm): 19.28 (CH_3), 55.44 (OCH_3), 107.90 (C), 110.56 (CH), 112.33 (C), 114.89 (CH), 115.25 (CH), 117.20 (CH), 120.92 (CH), 123.33 (CH), 126.91 (CH), 128.67 (CH), 129.61 (C), 145.01 (C), 150.82 (CH), 151.23 (C), 151.81 (C), 153.43 (C), 153.56 (C), 154.61 (C), 160.24 (C), 161.22 (C), 163.82 (CO of coumarin).

8-[4-(3,4-Dimethoxyphenyl)-[2,4′-bipyridin]-6-yl]-7-hydroxy-4-methyl-chromen-2-one (***5x***). Yield: 75 %; m.p.: 236 °C; Anal. Calcd. for $C_{28}H_{22}N_2O_5$: C, 72.09; H, 4.75; N, 6.01 %. Found: C, 71.80; H, 4.73; N, 6.04 %; IR (KBr, cm^{-1}): 3418 (O–H stretching), 3040 (aromatic C–H stretching), 2949 (aliphatic C–H stretching), 1736 (C=O δ-lactone stretching), 1598 and 1542 (aromatic C=C and C=N stretchings); ^{1}H-NMR (400 MHz, $CDCl_3$, δ / ppm): 2.47 (3H, *s*, CH_3), 3.98 and 4.10 (6H, 2×*s*, 2×OCH_3), 6.16 (1H, *s*, C_3–H), 7.01–8.81 (10H, *m*, Ar-H), 9.06 (1H, poorly resolved *d*, C_3''–H), 15.70 (1H, *bs*, OH); ^{13}C-NMR (100 MHz, $CDCl_3$, δ / ppm): 19.29 (CH_3), 56.00 (OCH_3), 56.25 (OCH_3), 107.51 (C), 110.09 (CH), 110.36 (CH), 111.40 (CH), 112.21 (C), 115.27 (CH), 116.86 (CH), 119.65 (CH), 120.82 (CH), 123.24 (CH), 126.92 (CH), 129.66 (C), 144.87 (C), 149.76 (C), 150.72 (CH), 150.99 (C), 151.54 (C), 153.41 (C), 153.66 (C), 154.53 (C), 160.13 (C), 163.99 (CO of coumarin).

*8-[4-(4-Chlorophenyl)-[2,4'-bipyridin]-6-yl]-7-hydroxy-4-methyl-chromen--2-one (**5y**)*. Yield: 70 %; m.p.: 265–267 °C; Anal. Calcd. for $C_{26}H_{17}ClN_2O_3$: C, 70.83; H, 3.89; N, 6.35 %. Found: C, 70.65; H, 3.87; N, 6.32 %; IR (KBr, cm^{-1}): 3422 (O–H stretching), 3030 (aromatic C–H stretching), 2960 (aliphatic C–H stretching), 1728 (C=O δ-lactone stretching), 1597 and 1542 (aromatic C=C and C=N stretchings), 819 (C–H bending of *p*-disubstituted benzene ring); ^{1}H-NMR (400 MHz, $CDCl_3$, δ / ppm): 2.47 (3H, *s*, CH_3), 6.18 (1H, *s*, C_3–H), 7.01–8.81 (11H, *m*, Ar-H), 8.95 (1H, poorly resolved *d*, C_3''–H), 15.25 (1H, *bs*, OH); ^{13}C-NMR (100 MHz, $CDCl_3$, δ / ppm): 19.29 (CH_3), 107.79 (C), 110.70 (CH), 112.40 (C), 117.72 (CH), 120.81 (CH), 123.80 (CH), 128.71 (CH), 129.69 (CH), 135.86 (CH), 136.33 (CH), 144.76 (C), 150.54 (C), 150.85 (C), 150.91 (CH), 152.19 (C), 153.38 (C), 153.62 (C), 154.94 (C), 160.21 (C), 160.63 (C), 163.54 (CO of coumarin).

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