SYNTHESIS OF 3-(2-OXO-2*H*-PYRANYL-3) SUBSTITUTED LACTIC ACID DERIVATIVES

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Dedicated to Professor Teruaki Mukaiyama, Science University of Tokyo, on the ocassion of his 73rd birthday

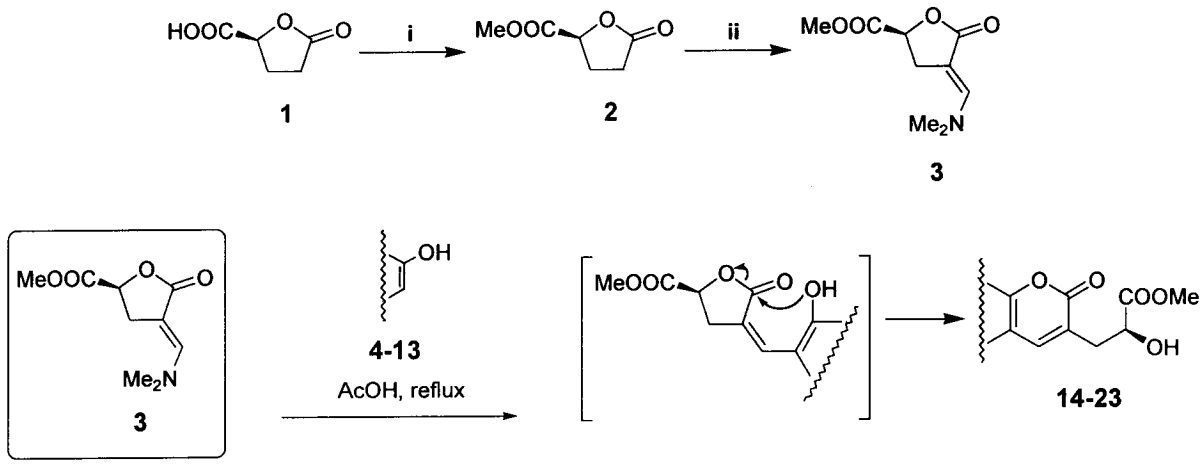
Abstract – (*S*)-3-[(*E*)-Dimethylaminomethylidene]-5-(methoxycarbonyl)tetrahydrofuran-2-one (**3**) was transformed in one step with various *C*,*O*-dinucleophiles (**4-13**) into the corresponding 3-(2-oxo-2*H*-pyranyl-3)-2-hydroxypropanoates (**14-23**).

Chiral hydroxy acids, such as lactic acid, malic acid, mandelic acid, tartaric acid, and their derivatives found a wide applicability in asymmetric synthesis, especially as chiral synthons, chiral auxiliaries, and resolving agents.[1] However, much less attention has been paid to the synthesis and utilisation of heteroaryl substituted α-hydroxy acid derivatives. Previously, we have shown that 3-dimethylaminopropenoates can serve as versatile reagents for the preparation of a variety of heterocyclic systems.[2,3] In this connection we have recently shown the utilisation of chiral 3-dimethylaminopropenoate analogs, derived from L-pyroglutamic acid, for the preparation of 3-heteroarylalanine derivatives and heterocyclic systems with an α-amino acid structural element.[4–6] In continuation of our work in this field, we now report on novel, one step synthesis of methyl 3-(2-oxo-2*H*-pyranyl-3)-2-hydroxypropanoates (**14-23**) from easily available (*S*)-3-[(*E*)-dimethylaminomethylidene]-5-(methoxycarbonyl)tetrahydrofuran-2-one (**3**).[6]

The starting compound, (*S*)-3-[(*E*)-dimethylaminomethylidene]-5-(methoxycarbonyl)tetrahydrofuran-2-one (**3**), was prepared from commercially available (*S*)-5-oxotetrahydrofuran-2-carboxylic acid (**1**).[6] When compound (**3**) was treated with carbocyclic and heterocyclic 1,3-dicarbonyl compounds and their analogs (**4-13**), the corresponding methyl 3-heteroaryl-2-hydroxypropanoates (**14-23**) with fused 2*H*-pyran-2-one heterocyclic residue were formed (Scheme 1).

The following 1,3 dicarbonyl compounds and their analogs were chosen: cyclohexane-1,3-dione (**4**), 5,5-dimethylcyclohexane-1,3-dione (**5**), 1,5-dihydroxynaphthalene (**6**), 2,3-dihydroxynaphthalene (**7**), 2,7-

Scheme 1



Reagents and conditions: **i** MeOH, $SOCl_2$; **ii** $(Me_2N)_2CHO$-*t*-Bu, toluene, 100°.

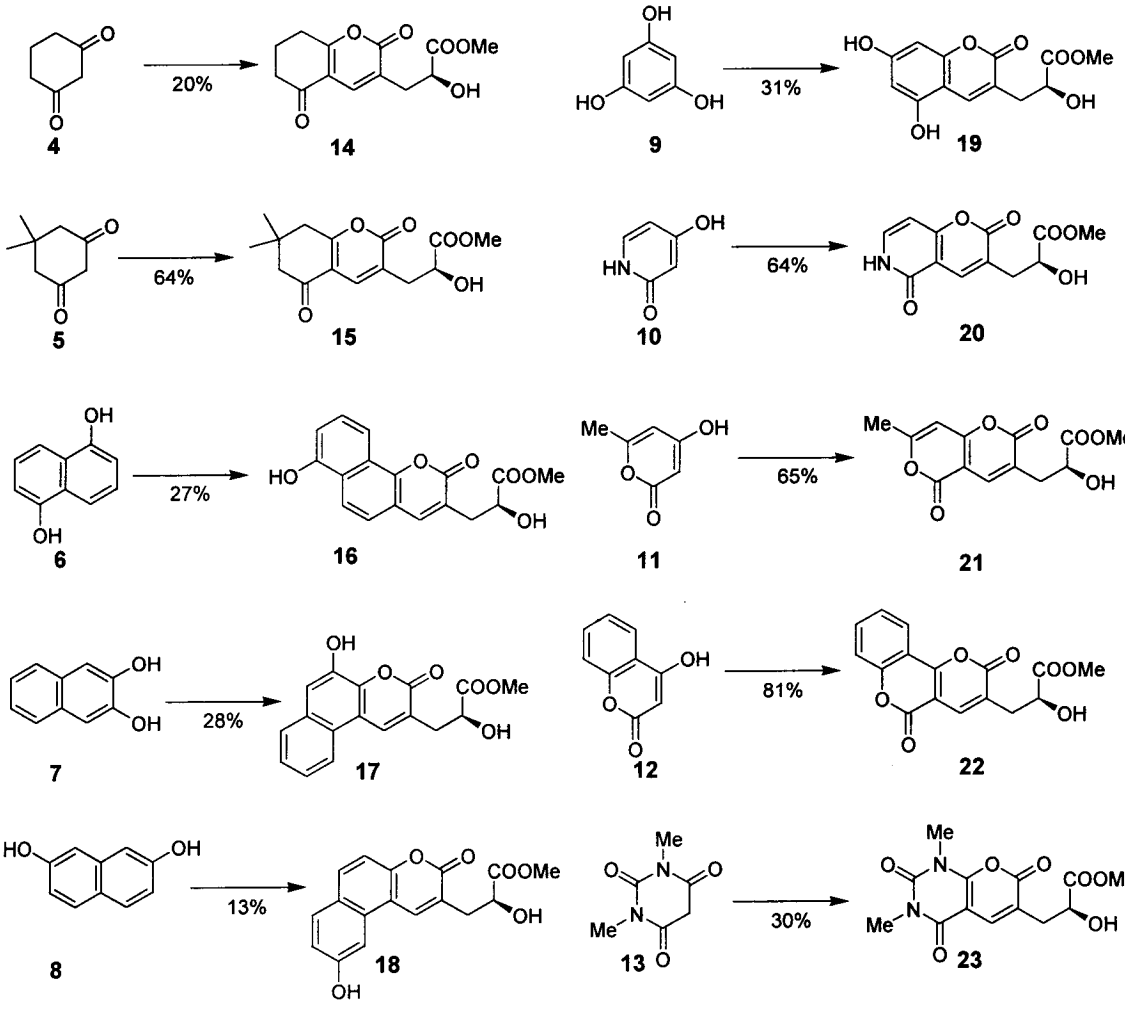
dihydroxynaphthalene (**8**), 1,3,5-trihydroxybenzene (**9**), 4-hydroxypyridin-2-one (**10**), 4-hydroxy-6-methyl-2*H*-pyran-2-one (**11**), 4-hydroxy-2*H*-benzo[*b*]pyran-2-one (**12**) and 1,3-dimethylbarbituric acid (**13**). Compounds (**4-13**) gave after treatment with **3** in refluxing acetic acid, the corresponding methyl (*S*)-3-heteroaryl-2-hydroxypropanoates with the following heteroaryl residues: 2,5-dioxo-5,6,7,8-tetrahydro-2*H*-benzo[*b*]pyranyl-3 (**14**), 7,7-dimethyl-2,5-dioxo-5,6,7,8-tetrahydro-2*H*-benzo[*b*]pyranyl-3 (**15**), 7-hydroxy-2-oxo-2*H*-naphtho[1,2-*b*]pyranyl-3 (**16**), 10-hydroxy-2-oxo-2*H*-naphtho[2,1-*b*]pyranyl-3 (**17**), 6-hydroxy-2-oxo-2*H*-naphtho[2,1-*b*]pyranyl-3 (**18**), 5,7-dihydroxy-2-oxo-2*H*-benzo[*b*]pyranyl-3 (**19**), 2,5-dioxo-5,6-dihydro-2*H*-pyrano[3,2-*c*]pyridinyl-3 (**20**), 2,5-dioxo-7-methyl-2*H*,5*H*-pyrano[4,3-*b*]pyranyl-3 (**21**), 2,5-dioxo-2*H*,5*H*-benzo[*b*]pyrano[4,3-*b*]pyranyl-3 (**22**), and 1,3-dimethyl-2,4,7-trioxo-1,2,3,4-tetrahydro-7*H*-pyrano[2,3-*d*]pyrimidinyl-6 (**23**), respectively (Scheme 2).

EXPERIMENTAL

Melting points were taken on a Kofler micro hot stage. The ^1H NMR spectra and ^{13}C NMR spectra were obtained on a Bruker Avance DPX 300 (300 MHz) spectrometer with DMSO-d_6 and $CDCl_3$ as solvents and Me_4Si as internal standard. The microanalyses for C, H, and N were obtained on a Perkin-Elmer CHN *Anal*yser 2400. The optical rotations were measured on a Perkin-Elmer 241 MC Polarimeter.

(*S*)-5-(Methoxycarbonyl)tetrahydrofuran-2-one (**2**) and (*S*)-3-[(*E*)-dimethylaminomethylidene]-5-(methoxycarbonyl)tetrahydrofuran-2-one (**1**) were prepared according to the procedures described in the literature.[6, 7]

Scheme 2



(*S*)-3-[(*E*)-Dimethylaminomethylidene]-5-(methoxycarbonyl)tetrahydrofuran-2-one (3).[6] A mixture of (*S*)-5-(methoxycarbonyl)tetrahydrofuran-2-one (**2**) (1.441 g, 10 mmol), toluene (20 ml), and bis(dimethylamino)-*tert*-butoxymethane (2.61 g, 15 mmol) was heated at 90–100°C for 2 h, volatile components were evaporated *in vacuo*, and the solid residue was crystallised from ethyl acetate. The precipitate was collected by filtration to give **3**; yield:1.154 g, 58%; mp 113–115°C (from ethyl acetate); $[\alpha]_D^{23}$ = +2.3° (*c* = 1.10, CH_2Cl_2). ^{1}H NMR (300 MHz, $CDCl_3$): δ 3.04 (6H, s, NMe_2), 3.20 (1H, ddd, *J* = 1.3, 4.7, 14.3 Hz, 4-Ha), 3.43 (1H, ddd, *J* = 1.1, 10.0, 14.1 Hz, 4-Hb), 3.79 (3H, s, OMe), 4.86 (1H, dd, *J* = 5.0, 10.0 Hz, 5-H), 7.13 (1H, t, *J* = 1.5 Hz, 3'-H). ^{13}C NMR (75.5 MHz, $CDCl_3$): δ 30.2, 42.1, 52.8, 72.6, 84.8, 148.4, 171.8, 174.2. *Anal.* Calcd for $C_9H_{13}NO_4$: C, 54.26; H, 6.58; N, 7.03. Found: C, 54.31; H, 6.79; N, 7.14.

General Procedure for the Preparation of Fused 2*H*-Pyran-2-one Hydroxypropanoates (13-24):

A mixture of (*S*)-3-[(*E*)-dimethylaminomethylidene]-5-(methoxycarbonyl)tetrahydrofuran-2-one (**3**) (0.199 g, 1 mmol), 1,3-dinucleophile (**4-13**) (1 mmol), and glacial acetic acid (4 mL) was heated at reflux temperature for 2–5 h. Volatile components were evaporated *in vacuo*, the solid residue was crystallised from the appropriate solvent and the precipitate was collected by filtration to give substituted lactic acid derivatives (**14-23**).

Methyl (*S*)-3-(2,5-Dioxo-5,6,7,8-tetrahydro-2*H*-benzo[*b*]pyranyl-3)-2-hydroxypropanoate (14). This compound was prepared from cyclohexane-1,3-dione (**4**), reflux for 2 h; the crude product was purified by column chromatography using ethyl acetate/*n*-hexane (10 : 1) as solvent to give **14**; yield 20%; mp 122–124°C (from ethyl acetate/ether = 1 : 1); $[\alpha]_D^{23}$ = +25.5° (*c* = 1.00, CH_2Cl_2). ^{1}H NMR (300 MHz, DMSO-d_6): δ 2.06 (2H, deg. tt, *J* = 6.0 Hz, 7-CH_2), 2.48 (2H, t, *J* = 6.0 Hz, 8-CH_2) 2.62 (1H, dd, *J* = 8.7,14.3 Hz, 3-Ha), 2.82 (1H, dd, *J* = 4.5, 14.0 Hz, 3-Hb), 2.85 (2H, t, *J* = 6.0 Hz, 6-CH_2), 3.64 (3H, s, OMe), 4.28 (1H, ddd, *J* = 4.7, 8.7, 6.2 Hz, 2-H), 5.59 (1H, d, *J* = 6.4 Hz, OH), 7.65 (1H, s, 4'-H). ^{13}C NMR (75.5 MHz, DMSO-d_6): δ 20.7, 28.0, 35.7, 36.9, 52.4, 68.7, 114.7, 122.6, 138.2, 161.6, 174.3, 174.4, 195.0. *Anal.* Calcd for $C_{13}H_{14}O_6$: C, 58.65; H, 5.30. Found: C, 58.47; H, 5.29.

Methyl (*S*)-3-(7,7-Dimethyl-2,5-dioxo-5,6,7,8-tetrahydro-2*H*-benzo[*b*]pyranyl-3)-2-hydroxypropanoate (15). This compound was prepared from 5,5-dimethylcyclohexane-1,3-dione (**5**), reflux for 3 h; yield 64%; mp 109–111°C (from ethyl acetate); $[\alpha]_D^{23}$ = –2.2° (*c* = 1.16, $CHCl_3$). ^{1}H NMR (300 MHz, DMSO-d_6): δ 1.05 (6H, s, 7-Me), 2.41 (2H, s, CH_2), 2.62 (1H, dd, *J* = 8.7, 13.9 Hz, 3-Ha), 2.80 (2H, s, CH_2), 2.82 (1H, dd, *J* = 4.4, 14.3 Hz, 3-Hb), 3.63 (3H, s, OMe), 4.29 (1H, ddd, *J* = 4.5, 8.7, 6.4 Hz, 2-H), 5.60 (1H, d, *J* = 6.4 Hz, OH), 7.64 (1H, s, 4'-H). ^{13}C NMR (75.5 MHz, DMSO-d_6): δ 27.5, 27.6, 32.3, 34.8, 49.7, 51.5, 67.8, 112.9, 121.7, 137.0, 161.0, 171.8, 173.5, 194.0. *Anal.* Calcd for $C_{15}H_{18}O_6$: C, 61.22; H, 6.16. Found: C, 60.92; H, 6.50.

Methyl (*S*)-3-(7-Hydroxy-2-oxo-2*H*-naphtho[1,2-*b*]pyranyl-3)-2-hydroxypropanoate (16). This compound was prepared from 1,5-dihydroxynaphthalene (**6**), reflux for 6 h; the crude product was purified by column chromatography using ether as solvent to give **16**; yield 27%; mp 171–173°C (from ethyl acetate); $[\alpha]_D^{23}$ = –39.5° (*c* = 1.16, DMF). ^{1}H NMR (300 MHz, DMSO-d_6): δ 2.77 (1H, dd, *J* = 9.0, 13.7 Hz, 3-Ha), 2.97 (1H, dd, *J* = 4.7, 13.8 Hz, 3-Hb), 3.66 (3H, s, OMe), 4.43 (1H, dt, *J* = 5.3, 7.9 Hz, 2-H), 5.68 (1H, d, *J* = 6.0 Hz, 2-OH), 7.06 (1H, d, *J* = 7.9 Hz, 8'-H), 7.50 (1H, t, *J* = 7.9 Hz, 9'-H), 7.61 (1H, d, *J* = 7.7 Hz, 10'-H), 7.79 (1H, d, *J* = 8.3 Hz, 6'-H), 8.01 (1H, s, 4'-H), 8.02 (1H, d, *J* = 8.3 Hz, 5'-H), 10.48 (1H, br

s, 7'-OH). ^{13}C NMR (75.5 MHz, DMSO-d_6): δ 36.5, 52.4, 68.9, 111.7, 112.4, 115.8, 119.3, 123.4, 124.3, 124.9, 125.9, 128.9, 143.4, 150.1, 154.4, 161.8, 174.5. *Anal.* Calcd for $C_{17}H_{14}O_6$: C, 64.97; H, 4.49. Found: C, 64.57; H, 4.79.

Methyl (*S*)-3-(10-Hydroxy-2-oxo-2*H*-naphtho[2,1-*b*]pyranyl-3)-2-hydroxypropanoate (17). This compound was prepared from 2,3-dihydroxynaphthalene (**7**), reflux for 6 h; the crude product was purified by column chromatography using ether as solvent to give **17**; yield 28%; mp 192–195°C (from ethyl acetate); $[\alpha]_D^{23}$ = –8.0° (*c* = 0.99, $CHCl_3$). ^{1}H NMR (300 MHz, DMSO-d_6): δ 2.85 (1H, dd, *J* = 8.7, 13.9 Hz, 3-Ha), 3.03 (1H, dd, *J* = 4.9, 13.6 Hz, 3-Hb), 3.65 (3H, s, OMe), 4.48 (1H, dd, *J* = 4.9, 8.7 Hz, 2-H), 5.65 (1H, br s, 2-OH), 7.44 (1H, s, 9'-H), 7.47-7.54 (2H, m, 6'-H and 7'-H), 7.80-7.84 (1H, m, 8'-H), 8.35-8.38 (1H, m, 5'-H), 8.73 (1H, s, 4'-H), 10.45 (1H, br s, 10'-OH). ^{13}C NMR (75.5 MHz, DMSO-d_6): δ 35.8, 51.5, 68.1, 113.0, 114.2, 121.9, 123.1, 123.7, 124.8, 126.0, 126.7, 130.6, 138.2, 144.0, 144.1, 160.5, 173.6. *Anal.* Calcd for $C_{17}H_{14}O_6$: C, 64.97; H, 4.49. Found: C, 64.57; H, 4.55.

Methyl (*S*)-3-(6-Hydroxy-2-oxo-2*H*-naphtho[2,1-*b*]pyranyl-3)-2-hydroxypropanoate (18). This compound was prepared from 2,7-dihydroxynaphthalene (**8**), reflux for 5 h; the crude product was purified by column chromatography using ether as solvent to give **18**, yield 13%; mp 181–183°C (from ethyl acetate); $[\alpha]_D^{23}$ = –6.5° (*c* = 0.65, DMF). ^{1}H NMR (300 MHz, DMSO-d_6): δ 2.83 (1H, dd, *J* = 8.3, 13.9 Hz, 3-Ha), 3.00 (1H, dd, *J* = 4.9, 13.9 Hz, 3-Hb), 3.65 (3H, s, OMe), 4.45 (1H, ddd, *J* = 5.3, 6.0, 8.7 Hz, 2-H), 5.67 (1H, d, *J* = 6.4 Hz, 2-OH), 7.17 (1H, dd, *J* = 2.3, 9.0 Hz, 7'-H), 7.30 (1H, d, *J* = 9.0 Hz, 10'-H), 7.65 (1H, d, *J* = 2.3 Hz, 5'-H), 7.89 (1H, d, *J* = 8.7 Hz, 8'-H), 8.00 (1H, d, *J* = 9.0 Hz, 9'-H), 8.52 (1H, s, 4'-H), 10.16 (1H, br s, 6'-OH). ^{13}C NMR (75.5 MHz, DMSO-d_6): δ 36.5, 52.4, 69.1, 105.3, 112.7, 113.7, 118.7, 123.5, 125.2, 131.5, 131.6, 133.1, 138.9, 153.7, 158.5, 161.9, 174.6. *Anal.* Calcd for $C_{17}H_{14}O_6$: C, 64.97; H, 4.49. Found: C, 65.14; H, 4.51.

Methyl (*S*)-3-(5,7-Dihydroxy-2-oxo-2*H*-benzo[*b*]pyranyl-3)-2-hydroxypropanoate (19). This compound was prepared from 1,3,5-trihydroxybenzene (**9**), reflux for 3 h; the crude product was purified by column chromatography using ethyl acetate/*n*-hexane (2 : 1) as solvent to give **19**; yield 31%; mp 205–207°C (from methanol); $[\alpha]_D^{23}$ = +1.3° (*c* = 0.90, $CHCl_3$). ^{1}H NMR (300 MHz, DMSO-d_6): δ 2.60 (1H, dd, *J* = 8.3, 13.9 Hz, 3-Ha), 2.82 (1H, dd, *J* = 4.7, 13.7 Hz, 3-Hb), 3.62 (3H, s, OMe), 4.28 (1H, ddd, *J* = 4.9, 6.4, 8.3 Hz, 2-H), 5.56 (1H, d, *J* = 6.4 Hz, OH), 6.17 (1H, d, *J* = 1.9 Hz, 6'-H), 6.26 (1H, d, *J* = 2.3 Hz, 8'-H), 7.80 (1H, s, 4'-H), 10.24 (1H, s, Het-OH), 10.56 (1H, s, Het-OH). ^{13}C NMR (75.5 MHz, DMSO-d_6): δ 35.5, 51.4, 68.4, 93.6, 98.2, 101.9, 116.6, 137.2, 155.4, 155.5, 161.2, 161.5, 173.3. *Anal.* Calcd for $C_{13}H_{12}O_7$: C, 55.72; H, 4.32. Found: C, 55.30; H, 4.30.

Methyl (*S*)-3-(2,5-Dioxo-5,6-dihydro-2*H*-pyrano[3,2-*c*]pyridinyl-3)-2-hydroxypropanoate (20). This compound was prepared from 4-dihydroxypyridin-2-one (**10**), reflux for 3 h; yield 64%; mp 227–229°C (from ethyl acetate); $[\alpha]_D^{23}$ = –2.1° (*c* = 0.78, $CHCl_3$). ^1H NMR (300 MHz, DMSO-d_6): δ 2.66 (1H, ddd, *J* = 0.8 , 8.4, 13.9 Hz, 3-Ha), 2.86 (1H, ddd, *J* = 0.8, 4.5, 13.9 Hz, 3-Hb), 3.64 (3H, s, OMe), 4.31 (1H, dt, *J* = 8.3, 5.3 Hz, 2-H), 5.62 (1H, d, *J* = 6.4 Hz, OH), 6.34 (1H, dd, *J* = 7.5, 0.8 Hz, 8'-H), 7.58 (1H, d, *J* = 7.5 Hz, 7'-H), 7.82 (1H, d, *J* = 0.8 Hz, 4'-H), 11.88 (1H, br s, 6'-H). ^{13}C NMR (75.5 MHz, DMSO-d_6): δ 36.1, 52.4, 68.9, 98.3, 109.1, 122.2, 139.0, 139.1, 160.9, 161.2, 163.3, 174.4. *Anal.* Calcd for $C_{12}H_{11}NO_6$: C, 54.34; H, 4.18; N, 5.28. Found: C, 54.12; H, 4.24; N, 5.37.

Methyl (*S*)-3-(2,5-Dioxo-7-methyl-2*H*,5*H*-pyrano[4,3-*b*]pyranyl-3)-2-hydroxypropanoate (21). This compound was prepared from 4-hydroxy-6-methyl-2*H*-pyran-2-one (**11**), reflux for 3 h; yield 65%; mp 162–164°C (from ethyl acetate); $[\alpha]_D^{23}$ = + 0.9° (*c* = 0.57, CH_2Cl_2). ^1H NMR (300 MHz, DMSO-d_6): δ 2.32 (3H, s, 7'-Me), 2.67 (1H, dd, *J* = 8.5, 14.1 Hz, 3-Ha), 2.86 (1H, dd, *J* = 4.1, 13.6 Hz, 3-Hb), 3.64 (3H, s, OMe), 4.32 (1H, dt, *J* = 5.4, 7.7 Hz, 2-H), 5.64 (1H, d, *J* = 6.4 Hz, OH), 6.64 (1H, s, 4'-H), 7.72 (1H, s, 8'-H). ^{13}C NMR (75.5 MHz, DMSO-d_6): δ 20.8, 35.9, 52.5, 68.7, 99.6, 101.8, 123.4, 138.8, 160.4, 160.8, 165.5, 166.4, 174.3. *Anal.* Calcd for $C_{13}H_{12}O_7$: C, 55.72; H, 4.32. Found: C, 55.28; H, 4.31.

Methyl (*S*)-3-(2,5-Dioxo-2*H*,5*H*-benzo[*b*]pyrano[4,3-*b*]pyranyl-3)-2-hydroxypropanoate (22). This compound was prepared from 4-hydroxy-2*H*-benzo[*b*]pyran-2-one (**12**), reflux for 2 h; yield 81%; mp 208–210°C (from ethyl acetate); $[\alpha]_D^{23}$ = –35.9° (*c* = 1.14, $CHCl_3$). ^1H NMR (300 MHz, DMSO-d_6): δ 2.77 (1H, dd, *J* = 8.4, 14.3 Hz, 3-Ha), 2.95 (1H, dd, *J* = 4.0, 14.2 Hz, 3-Hb), 3.66 (3H, s, OMe), 4.38 (1H, ddd, *J* = 4.7, 6.4, 8.5 Hz, 2-H), 5.70 (1H, d, *J* = 6.4 Hz, OH), 7.48-7.55 (2H, m, 7'-H and 9'-H), 7.79 (1H, ddd, *J* = 8.6, 7.2, 1.5 Hz, 8'-H), 7.87 (1H, s, 4'-H), 8.00 (1H, br dd, *J* = 7.9, 1.5 Hz, 10'-H). ^{13}C NMR (75.5 MHz, DMSO-d_6): δ 35.9, 52.5, 68.7, 104.4, 113.8, 118.0, 123.7, 125.5, 126.2, 135.1, 139.1, 153.6, 159.4, 160.1, 160.6, 174.3. *Anal.* Calcd for $C_{16}H_{12}O_7$: C, 60.76; H, 3.82. Found: C, 60.77; H, 3.83.

Methyl (*S*)-3-(1,3-Dimethyl-2,4,7-trioxo-1,2,3,4-tetrahydro-7*H*-pyrano[2,3-*d*]pyrimidinyl-6)-2-hydroxy-propanoate (23). This compound was prepared from 1,3-dimethylbarbituric acid (**13**), reflux for 5 h; the crude product was purified by column chromatography using ethyl acetate as solvent to give **13**; yield 30%; mp 172–174°C (from ethyl acetate); $[\alpha]_D^{23}$ = +2.0° (*c* = 0.82, CH_2Cl_2). ^1H NMR (300 MHz, DMSO-d_6): δ 2.65 (1H, ddd, *J* = 0.8, 8.7, 13.9 Hz, 3-Ha), 2.83 (1H, ddd, *J* = 0.8, 4.7, 14.1 Hz, 3-Hb), 3.23 (3H, s, Het-Me), 3.38 (3H, s, Het-Me), 3.64 (3H, s, OMe), 4.27 (1H, ddd, *J* = 4.9, 8.3, 6.4 Hz, 2-H), 5.62 (1H, d, *J* = 6.4 Hz, OH), 7.81 (1H, s, 4'-H). ^{13}C NMR (75.5 MHz, DMSO-d_6): δ 28.9, 29.7, 35.3, 52.5, 68.9, 93.0,

115.1, 141.2, 150.3, 158.3, 159.4, 159.7, 174.4. *Anal.* Calcd for $C_{13}H_{14}N_2O_7$: C, 50.33; H, 4.55; N, 9.03. Found: C, 50.16; H, 4.52; N, 8.87.

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