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A NOVEL TWO STEP SYNTHESIS OF 3,4-DIHYDRO-4-OXO-5*H*-PYRANO[2,3-*d*]PYRIMIDINES

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Abstract- A novel two step synthesis of 5,7-diaryl-3,4-dihydro-4-oxo-5*H*-pyrano[2,3-*d*]pyrimidines has been reported based on condensation of 2-thiobarbiturates with chalcones followed by reductive desulfurization of 5,7-diaryl-1,2,3,4-tetrahydro-4-oxo-2-thioxo-5*H*-pyrano[2,3-*d*]pyrimidines with nickel boride.

Pyrans[1] and pyrimidines[2] individually or in combination possess significant biological activity such as antiallergic, analgesic, sedative and antiphlogistic. Some of the derivatives of pyrimidines also show herbicidal and inseticidal properties. Different routes have been described in literature for the synthesis of pyrano[2,3-*d*]pyrimidines[3] as well as 2-oxo and 2-thioxo analogues of 1,2,3,4-tetrahydro-4-oxo-5*H*-pyrano[2,3-*d*]pyrimidines.[4,5] Unlike the synthesis of 2-oxo and 2-thioxo analogues, synthesis of 3,4-dihydro-4-oxo-5*H*-pyrano[2,3-*d*]pyrimidines has not received any attention. Thus there is sufficient scope for the development of new, efficient and practical protocols for the synthesis of 3,4-dihydro- 4-oxo-5*H*-pyrano[2,3-*d*]pyrimidines. We have attempted to achieve the desired synthesis by addition of 2-thiobarbiturates to different chalcones to give 5,7-diaryl-1,2,3,4-tetrahydro-4-oxo-2-thioxo-5*H*-pyrano[2,3-*d*]pyrimidines followed by reductive desulfurization with nickel boride which has been reported as a reducing[6] and dethiating[7] agent by our group.

We report herein a convenient and efficient synthesis of 3,4-dihydro-4-oxo-5*H*-pyrano[2,3-*d*]pyrimidines **(4a-f)** and of 1,2,3,4-tetrahydro-4-oxo-5*H*-pyrano[2,3-*d*]pyrimidines **(5g-h)** in two steps. Thus 2-thioxo-5*H*-pyrano[2,3-*d*]pyrimidines **(3a-h)** were prepared by the condensation of chalcones **(1a-f)** with 2-thiobarbituric acids **(2a-b)** in high yields by a reported method.[5] Though compounds **(3a**, **3b** and **3g)** are reportedly known, a number of new compounds e.g. **3c-f** and **3h** were characterized by NMR, IR and MS spectra. These results are listed in Table 1.

Selective desulfurization of 1,2,3,4-tetrahydro-4-oxo-2-thioxo-5*H*-pyrano[2,3-*d*]pyrimidines without affecting the carbonyl group would result in the desired 3,4-dihydro-4-oxo-5*H*-pyrano[2,3-*d*]pyrimidines.

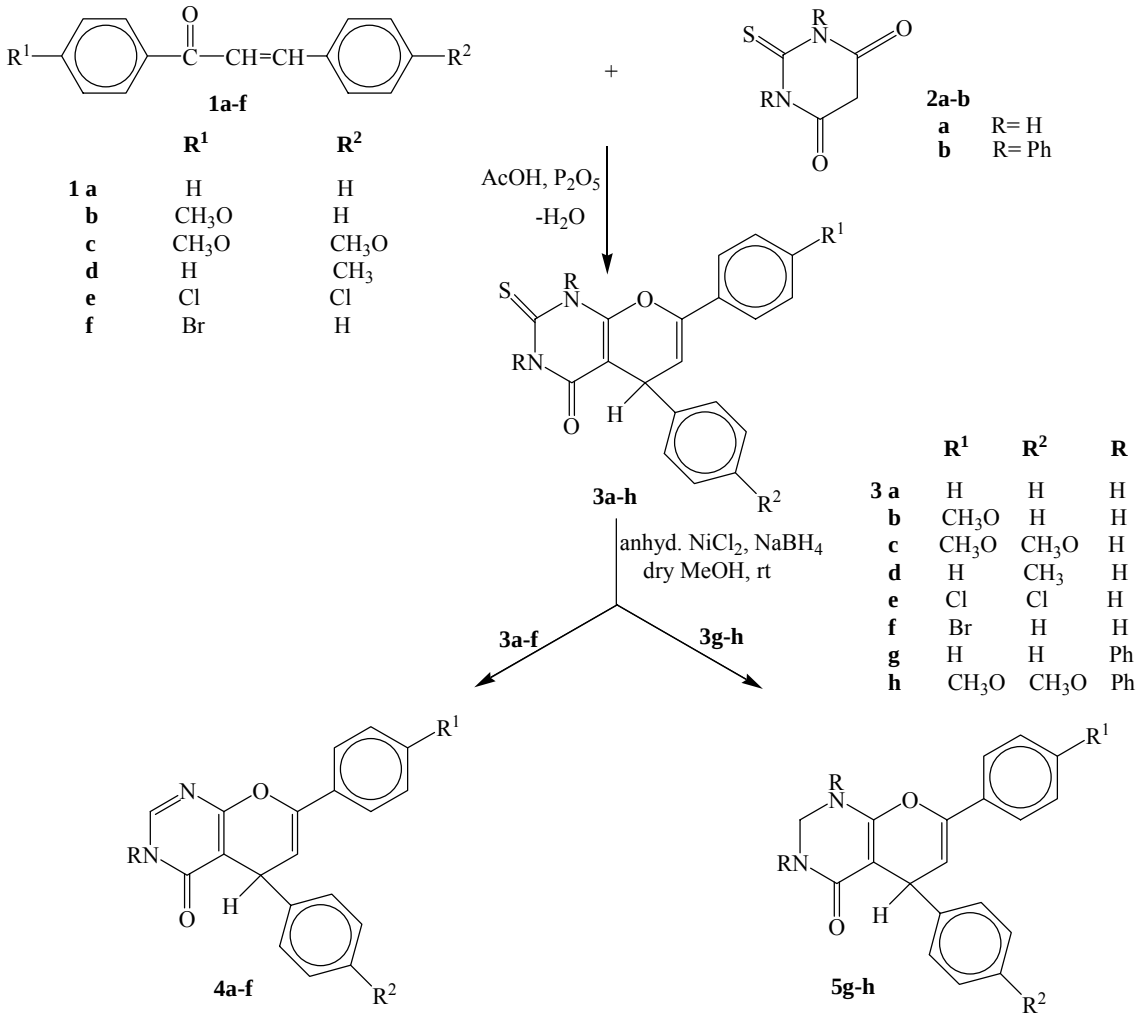
Table 1: Preparation of 4-oxo-2-thioxo-5*H*-pyrano[2,3-*d*]pyrimidines

S. No.	Chalcones (1)	2-Thiobarbituric acid (2)	2-Thioxo-5*H*-pyrano[2,3-*d*] pyrimidines (3)	Yield (%)
1.	**1a**	**2a**	**3a**	95
2.	**1b**	**2a**	**3b**	94
3.	**1c**	**2a**	**3c**	80
4.	**1d**	**2a**	**3d**	86
5.	**1e**	**2a**	**3e**	89
6.	**1f**	**2a**	**3f**	85
7.	**1a**	**2b**	**3g**	80
8.	**1c**	**2b**	**3h**	68

We have already reported[6,7] reductive desulfurization of 2-thiobarbiturates and 2-thioxo-4(3*H*)-quinazolonones with nickel boride prepared *in situ*. Therefore reaction of **3a** was carried out with nickel boride in methanol by changing the molar ratio of substrate: nickel chloride: sodium borohydride. It was observed that the reaction of **3a** completed in 90 min using a molar ratio of 1:10:10 ($S:NiCl_2:NaBH_4$) in dry methanol and gave **4a** in good yield which was characterized by IR, MS and NMR spectra. IR showed the absence of C=S stretch at 1560.56 cm^{-1} and appearance of C=N stretch at 1608.62 cm^{-1}. FAB MS spectra gives M^++1 molecular ion peak at 303. NMR showed a new signal at δ 8.0646 due to H-2 proton which confirms the formation of –CH=N-. With lower molar ratios of substrate to nickel chloride to sodium borohydride, the reaction was incomplete though it showed the formation of product **(4a)**. The reaction of **3a** with nickel boride in dry THF, acetonitrile and ethanol was incomplete even after 24 h. Reaction of **3a** in dry DMF was complete in 1 h but produced a complex mixture of products as observed by TLC. Removal of traces of DMF from the reaction mixture also posed some difficulties. Therefore dry methanol was employed in all further reactions. There was very little reaction of **3a** with $NaBH_4$ alone even after 24 h and the starting material was recovered unchanged when treated with $NiCl_2$ only. The desulfurization is thus undoubtedly proceeding due to the involvement of nickel boride formed *in situ*.

Whereas 2-thioxo-5*H*-pyrano[2,3-*d*]pyrimidines **(3b-f)** underwent reductive desulfurization with nickel boride to give 3,4-dihydro-4-oxo-5*H*-pyrano[2,3-*d*]pyrimidines **(4b-f)** as confirmed by spectral data **3g** and **3h** yielded 1,2,3,4-tetrahydro-4-oxo-5*H*-pyrano[2,3-*d*]pyrimidines**(5g** and **5h)** (Scheme I) respectively as the desulphurised product because both the nitrogens are blocked with a phenyl group. This is evident from disappearance of C=S streatching band in IR spectra and apperance of 2 protons of C-2 at ~ δ 5.17 and ~ δ 5.49 which coupled to each other (J=11.5). All the compounds are new products and have been well characterized by NMR, IR and MS spectra. These results are summarized in Table 2. Nickel boride shows high selectivity towards desulfurization and does not affect the amide carbonyl group. Nickel

boride also did not affect the chloro and bromo groups in the halogenated substrates **(3e** and **3f)**.



Scheme 1

Table 2: Preparation of 1,3,5,7-tetraaryl-3,4-dihydro-4-oxo-5*H*-pyrano[2,3-*d*]pyrimidines

S. No.	2-Thioxo-5*H*-pyrano[2,3-*d*] pyrimidines (3)	Molar ratio S:$NICL_2$:$NaBH_4$	Time (h)	Product (4)	Yield (%)
1.	**3a**	1:10:10	1.5	**4a**	89
2.	**3b**	1:10:10	1	**4b**	86
3.	**3c**	1:10:10	1	**4c**	82
4.	**3d**	1:10:10	2	**4d**	84
5.	**3e**	1:15:15[a]	1.5	**4e**	86
6.	**3f**	1:15:15[a]	1	**4f**	86
7.	**3g**	1:25:25[b]	8	**5g**	72
8.	**3h**	1:40:40[c]	6	**5h**	69

a: Reaction was incomplete in 1:10:10 so carried out in 1:15:15.
b: Reaction started with 1:10:10 and gradually increased the ratio to 1:25:25
c: Reaction started with 1:10:10 and gradually increased the ratio to 1:40:40.

We conclude that 5,7-diaryl-3,4-dihydro-4-oxo-5*H*-pyrano[2,3-*d*]pyrimidines and 1,3-diphenyl-5,7-diaryl-1,2,3,4-tetrahydro-4-oxo-5*H*-pyrano[2,3-*d*]pyrimidines can be prepared by condensation of chalcones with 2-thiobarbiturates followed by reductive desulfurization with nickel boride in good yields.

EXPERIMENTAL

The melting points were determined on a Tropical Labequip apparatus and are uncorrected. IR spectra were recorded on a Perkin Elmer FT-IR Spectrum 2000 and NMR spectra were recorded on Bruker avance spectrospin dpx-300 MHz instrument, respectively. The FAB mass spectra were recorded on JOEL SX 102/DA-6000 Spectrometer using Argon/Xenon as the FAB gas. Chalcones were prepared by condensation of acetophenones with benzaldehydes by the general procedure of Kohler and Chadwell.[8] 1,3-Diphenyl-2-thiobarbituric acid was prepared by condensation of malonic acid and N, N'-diphenylthiourea.[9] 2-Thiobarbituric acid (Merck) was used as such.

General Procedure for Preparation of 5,7-Diphenyl-1,2,3,4-tetrahydro-4-oxo-2-thioxo-5*H*-pyrano[2,3-*d*]pyrimidines

In a typical procedure, a solution of chalcone (**1a**, 0.01 mol) and 2-thiobarbituric acid (**2a**, 0.01 mol) in glacial acetic acid (30 mL) was taken in a 100 mL RB flask and P_2O_5 (10 g) was added to it. The contents were refluxed for 30 min with constant stirring. The mixture was cooled and poured onto crushed ice. The solid thus separated was filtered at pump, washed with dilute acetic acid, dried and crystallized from EtOH to give **3a** as white crystals. Yield: 95%. mp 290-291°C[5]; ^{1}H-NMR (DMSO-d_6): 4.4914-4.5071 (d, J= 4.71, 1H, H-5), 5.9453-5.9609 (d, J= 4.68, 1H, H-6), 7.1970-7.7174 (m, 10H, H-Ar); IR (KBr) ν_{max}: 1567.76 (C=S), 1674.72 (C=O), 3433.88 (NH), 2840.28 (C–H), 1137.95 cm^{-1} (C–O–C); FAB MS: 335(M^++1).

Compounds (**3b-h**) was obtained in a similar way. Spectroscopic data of all the new compounds are reported here in.

3c: Crystallized from EtOH as light brown crystals. Yield: 80%. mp 248-250°C; ^{1}H-NMR (DMSO-d_6): δ 3.698-3.857 (2 x OCH_3, 6H), 4.439-4.455 (d, J = 4.80 Hz, 1H, H-5), 5.658 (s, 1H, H-6), 6.798-6.919 (dd, 4H, 2′′,3′′,5′′ & 6′′), 7.205-7.231 (d, J=7.80, 2H, 3′ & 5′), 7.913-7.938 (d, J= 7.50, 2H, 2′ & 6′); IR (KBr) ν_{max}: 1560.56 (C=S), 1668.23 (C=O), 3448.79 (NH), 2956.15 (C–H), 1034.65 (C–O–C), 833.53 cm^{-1} (p-substituted aromatic); FAB MS: 395 (M^++1); Anal. Calcd for $C_{21}H_{18}N_2O_4S$: C 63.94, H 4.59, N 7.10, S 8.13. Found: C 63.79, H 4.67, N 7.02, S 8.24.

3d: Crystallized from EtOH as white crystals. Yield: 86%. mp 272-274°C; ^{1}H-NMR (DMSO-d_6) : δ 2.279 (s, 3H, CH_3), 4.496 (s, 1H, H-5), 5.755 (s, 1H, H-6), 7.086-7.368 (m, 11H, Ar′-H, Ar′′-H & 2N-H);

IR (KBr) ν_{max}: 3429.96 (NH), 2957.64 (C-H), 1666.63 (C=O), 1563.35 (C=S), 1054.12 (C–O–C), 859.37 cm^{-1} (p-substituted aromatic); FAB MS: 349 (M^++1); Anal. Calcd for $C_{20}H_{16}N_2O_2S$: C 68.94, H 4.62, N 8.04, S 9.20. Found: C 68.76, H 4.67, N 7.95, S 9.24.

3e: Crystallized from EtOH as light yellow crystals. Yield: 89%. mp 280-285°C; ^{1}H-NMR (DMSO-d_6) :δ 4.537-4.552 (d, J = 4.50, 1H, H-5), 5.746-5.763 (d, J = 5.10, 1H, H-6), 7.235-7.637 (m, 10H, Ar′-H, Ar′′-H & 2N-H); IR (KBr) ν_{max}: 1672.12 (C=O), 1561.40 (C=S), 1092.53 (C–C1), 826.39 cm^{-1} (p-substituted aromatic); FAB MS: 403 (M^+); Anal. Calcd for $C_{19}H_{12}N_2O_2Cl_2S$: C 56.58, H 2.99, N 6.94, S 7.95. Found: C 56.60, H 3.02, N 6.82, S 7.97.

3f: Crystallized from EtOH as brick red powder. Yield: 84.7%. mp 290°C (decomp.); ^{1}H-NMR (DMSO-d_6):δ 4.527-4.544 (d, J = 5.10, 1H, H-5), 5.821-5.838 (d, J = 5.10, 1H, H-6), 7.200-7.305 (m, 6H, Ar′′-H, 1N-H), 7.497-7.589 (m, 5H, Ar′-H, 3N-H); IR (KBr) ν_{max}: 3435.11 (NH), 2955.07 (C-H), 1674.02 (C=O), 1568.70 (C=S), 845.31 cm^{-1} (p-substituted aromatic); FAB MS: 413 (M^+), 415 (M^++2); Anal. Calcd for $C_{19}H_{13}N_2O_2BrS$: C 55.21, H 3.17, N 6.81, S 7.76. Found: C 55.33, H 3.24, N 6.74, S 7.85.

3h: Crystallized from EtOH as light brown powder. Yield: 68%. mp 295°C (decomp.); ^{1}H-NMR (DMSO-d_6) :δ 4.629 (s, 1H, H-5), 5.697 (s, 1H, H-6), 3.777 (s, 2 x OCH_3, 6H), 6.863 (s, 4H, 3′,5′,3′′ & 5′′ Ar-H), 7.145-7.478 (m, 4H, 2′, 6′, 2′′ & 6′′ Ar-H), 7.617 (m, 5H, N^1Ar-H) 7.789-7.825 (m, 5H, N^3Ar-H). . IR (KBr) ν_{max}: 1689.95 (C=O), 1511.41 (C=S), 839.86 cm^{-1} (*p*-substituted aromatic); FAB MS: 546 (M^+); Anal. Calcd for $C_{33}H_{26}N_2O_4S$: C 72.50, H 4.79, N 5.12, S 5.86. Found: C 72.42, H 4.89, N 5.17, S 5.90.

General Procedure for Preparation of 5,7-Diphenyl-3,4-dihydro-4-oxo-5*H*-pyrano[2,3-*d*]pyrimidines

In a typical procedure, **3a** (0.2 g, 0.5988 mmol) and dry MeOH (10 mL) were placed in a 100 mL round bottomed flask. The flask was mounted over a magnetic stirrer and the contents stirred. Anhyd. $NiCl_2$ (0.7724 g, 5.9880 mmol) was added to the flask followed by $NaBH_4$ (0.2275 g, 5.9880 mmol) at room temperature. Addition of $NaBH_4$ is highly exothermic. After complete disappearance of starting material as monitored by TLC (90:10 AcOEt: benzene; v/v), the reaction mixture was filtered through a celite pad (~ 1 inch). Nickel boride precipitate was washed with methanol (2 x 10 mL). The combined filtrate was diluted with water (~ 40 mL) and extracted with AcOEt (3 x 60 mL). The combined AcOEt extract was dried over anhyd. $MgSO_4$, filtered and concentrated to afford **4a** as white powder. Yield: 89%. mp 233°C; ^{1}H-NMR (DMSO-d_6) :δ 4.6306-4.6454 (d, J=4.44, 1H, H-5), 5.8757-5.8906 (d, J=4.47, 1H, H-6), 7.1818-7.6904 (m, 11H, Ar′-H, Ar′′-H & N-H), 8.0646 (s, 1H, H-2); IR (KBr) ν_{max}: 1646.99 (C=O), 1608.62 (C=N); FAB MS: 303 (M^++ 1); Anal. Calcd for $C_{19}H_{14}N_2O_2$: C 75.48, H 4.66, N 9.26. Found: C 75.43, H 4.59, N 9.37.

Compounds (**4b-f**) and (**5g-h**) were obtained in similar way.

4b: White powder. Yield: 86%. mp 236-237°C; ^{1}H-NMR (DMSO-d_6): δ 3.7774 (s, 3 x O-CH_3), 4.5856-4.6001 (d, J = 4.35, 1H, H-5), 5.7842-5.7996 (d, J = 4.62, 1H, H-6), 6.9634-6.9916 (d, J= 8.46, 2H, 3′ & 5′-H), 7.1867-7.2688 (m, 5H, Ar′′-H), 7.5973-7.6254 (d, J= 8.43, 2H, 2′ & 6′-H), 8.0940 (s, 1H, H-2); IR (Nujol) ν_{max}: 1642.00 (C=O), 1607.75 (C=N); FAB MS: 333 (M^++1); Anal. Calcd for $C_{20}H_{16}N_2O_3$: C 72.27, H 4.85, N 8.42. Found: C 72.42, H 4.79, N 8.37.

4c: Light yellow powder. Yield: 82%. mp 247-250°C; ^{1}H-NMR (DMSO-d_6) :δ 3.6994 (s, 3H, 4′-OCH_3), 3.7808 (s, 3H, 4′′-OCH_3), 4.5210-4.5358 (d, J = 4.44, 1H, H-5), 5.7573-5.7723 (d, J = 4.50, 1H, H-6), 6.8308-6.8576 (d, J= 8.05, 2H, 3′ & 5′-H), 6.9670-6.9943 (d, J= 8.19, 2H, 3′′ & 5′′-H), 7.1550-7.1818 (d, J=8.04, 2H, 2′′ & 6′′-H), 7.5949-7.6224 (d, J=8.25, 2H, 2′ & 6′-H), 8.0751 (s, 1H, H-2); IR (KBr) ν_{max}: 1642.60 (C=O), 1608.71 (C=N); FAB MS: 363 (M^++1); Anal. Calcd for $C_{21}H_{18}N_2O_4$: C 69.60, H 3.89, N 7.73. Found: C 69.49, H 3.91, N 7.57.

4d: White powder. Yield: 84%. mp 258-260°C; ^{1}H-NMR (DMSO-d_6) :δ 2.2837 (s, 3H, CH_3), 4.6216-4.6369 (d, J = 4.59, 1H, H-5), 5.7364-5.7524 (d, J = 4.80, 1H, H-6),7.0681-7.0930 (d, J= 7.47,2H, 2′′ & 6′′-H), 7.1856-7.2116 (d, J= 7.80, 2H, 5′′ &3′′-H), 7.3653-7.3892 (m, 3H, 3′,4′& 5′-H), 7.6534-7.6749 (d, J=6.45, 2H, 2′ & 6′-H), 7.9444 (s, 1H, H-2); IR (KBr) ν_{max}: 1667.96 (C=O), 1562.83 (C=N); FAB MS: 317 (M^++1); Anal. Calcd for $C_{20}H_{16}N_2O_2$: C 75.93, H 5.09, N 8.85. Found: C 75.76, H 5.02, N 8.69.

4e: White powder. Yield: 86%. mp 205°C (decompose); ^{1}H-NMR (DMSO-d_6) :δ 4.7204-4.7359 (d, J = 4.65, 1H, H-5), 6.0709-6.0870 (d, J = 4.83, 1H, H-6), 7.3612-7.4496 (q, 5H, 2′′, 3′′, 5′′, 6′′ & N^3-H), 7.5626-7.5909 (d, J= 8.49, 2H, 3′ & 5′-H), 7.7758-7.8037 (d, J= 8.37, 2H, 2′ & 6′-H), 8.2028 (s, 1H. H-2); IR (KBr) ν_{max}: 1641.91 (C=O), 1607.04 (C=N); FAB MS: 371 (M^+); Anal. Calcd for $C_{19}H_{12}N_2O_2Cl_2$: C 61.47, H 4.34, N 7.54. Found: C 61.65, H 4.29, N 7.64.

4f: Light yellow powder. Yield: 86%. mp 255°C (decompose); ^{1}H-NMR (DMSO-d_6): δ 4.6805-4.6941 (d, J= 4.08,1H. H-5), 6.0937-6.1093 (d, J= 4.68, 1H, H-6), 7.2646-7.7025 (m, 10 H, 4 Ar′ -H, 5 Ar′′ -H, 1 N^3-H), 8.1814 (s, 1H, H-2); IR (KBr) ν_{max}: 1650.33 (C=O), 1610.88 (C=N); FAB MS: 381 (M^++1); Anal. Calcd for $C_{19}H_{13}N_2O_2Br$: C 60.02, H 3.44, N 7.36. Found: C 59.84, H 3.33, N 7.49.

5g: White powder. Yield: 72%. mp 192-194 °C; ^{1}H-NMR (DMSO-d_6): δ 4.8582-4.8761 (d, J= 5.37,1H, H-5), 5.1757-5.2143 (d, J= 11.58, 1H, H-2), 5.5245-5.5630 (d, J= 11.55, 1H, H-2), 5.9033-5.9213 (d, J= 5.4, 1H, H-6), 7.1226-7.4303 (m, 20 H, 5 Ar′ -H, 5 Ar′′ -H, 10 N-Ar-H); IR (Nujol) ν_{max}: 1614.68 (C=O); FAB MS: 457 (M^++1); Anal. Calcd for $C_{31}H_{24}N_2O_2$: C 81.55, H 5.29, N 6.13. Found: C 81.44, H 5.27, N 6.20.

5h: Light brown powder. Yield: 69%. mp 186-189°C; ^{1}H-NMR (DMSO-d_6): δ 3.7670 (s, 6H, 2x OCH_3), 4.7578-4.7756 (d, J= 5.34,1H. H-5), 5.1766-5.2155 (d, J= 11.67, 1H, H-2), 5.4969-5.5355(d, J= 11.58, 1H, H-2), 5.7448-5.7629 (d, J= 5.43, 1H, H-6), 7.0892-7.4526 (m, 18 H, 4 Ar′ -H, 4 Ar′′ -H, 10 N-Ar-H); IR (KBr) ν_{max}: 1653.54 (C=O); FAB MS: 516 (M^++1); Anal. Calcd for $C_{33}H_{28}N_2O_4$: C 76.72, H 5.46, N 5.42. Found: C 76.81, H 5.39, N 5.35.

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