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Synthesis of various heterocycles using coumarinyl isothiocyanates

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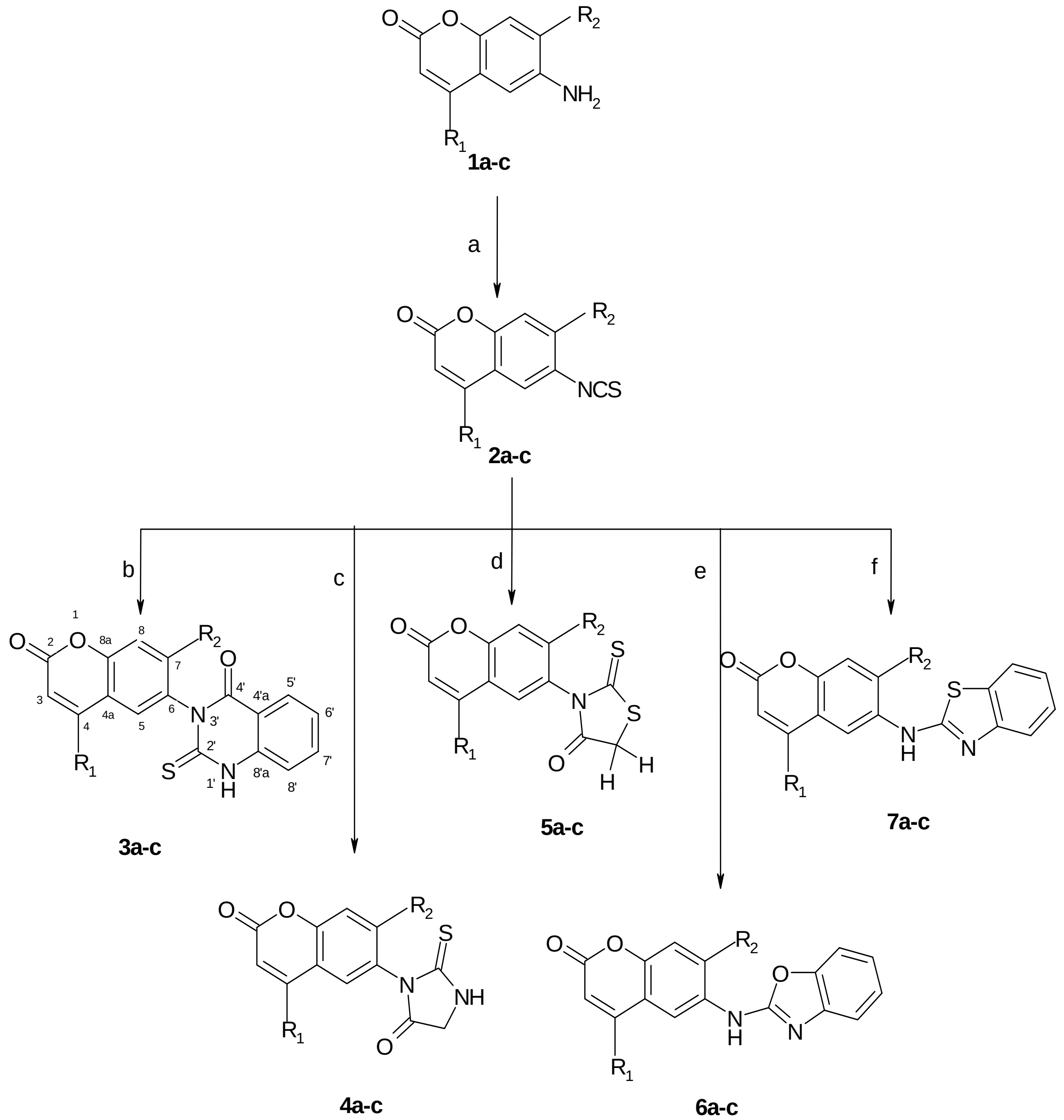
Coumarinyl isothiocyanates **2a-c** are obtained from 6-amino coumarins **1a-c** used as a starting materials for the construction of heterocycles. 6-Coumarinyl isothiocyanate on treatment with anthranilic acid, glycine, thioglycolic acid, *o*-aminophenol, *o*-aminothiophenol separately yielded 3-(2-oxo-*2H*-benzopyran-6-yl)-2-thioxo-2,3-dihydro-*1H*-quinazolin-4-one **3a-c**, 3-(2-oxo-*2H*-benzopyran-6-yl)-2-thioxo-imidazolidin-4-one **4a-c**, 3-(2-oxo-*2H*-benzopyran-6-yl)-2-thioxo-thiazolidin-4-one **5a-c**, 6-(benzooxazol-2-ylamino)-benzopyran-2-one **6a-c** and 6-(benzothiazol-2-ylamino)-benzopyran-2-one **7a-c** respectively. The structures of all the compounds are confirmed on the basis of spectral and analytical data. All the above compounds are screened for their antimicrobial activities and some of them have found to show significant antimicrobial activity.

Keywords: 6-aminocoumarin, quinazolinone, thiohydatoins, thiazolidinone, benzooxazole, benzothiazole, antimicrobial activity

Isothiocyanates remain very important starting materials for the construction of heterocycles[1-10]. The concept of counterattack reagent proposed[11], can be extended to isothiocyanates since many of their adducts with compounds carrying an active hydrogen atom undergo cyclization spontaneously or can be manipulated to yield heterocycles[12]. Coumarin chemistry has become more important since many years because of the discovery of the varied biochemical properties[13], industrial uses[14] and analytical applications[15] of these compounds. Coumarins are widely distributed in nature and known to exhibit various physiological activities[16,17]. For this purpose coumarinyl isothiocyanate is synthesized and used for construction of various heterocycles like quinazolinone, thiohydatoins, thiazolidinone, benzooxazole, benzothiazole derivatives.

Results and Discussion

Coumarinyl isothiocyanate **2a-c** were prepared by using 6-amino coumarin **1a-c** with carbon disulphide and iodine in pyridine. Condensation of the anthranillic acid with 6-coumarinyl isothiocyanate resulted in the formation of 3-(2-oxo-*2H*-benzopyran-6-yl)-2-thioxo-2,3-dihydro-*1H*-quinazolin-4-one **3a-c**. The IR spectrum of compound **3b** in KBr showed band at 3175 cm^{-1} for >NH stretching, at 1705 cm^{-1} for >C=O stretching, along with other bands at 1623, 1541, 1418, 1202, 758 cm^{-1} etc. The ^{1}H NMR spectrum of compound **3b** in $CDCl_3$ showed presence of singlet at δ 2.28 for three protons of methyl group at C_4, a singlet at 2.40 for three protons of methyl group at C7. Singlet observed at 6.32 for a proton at C_3. Doublet observed at 7.23 (J = 9Hz) was assigned a proton at $C_{8'}$. It showed multiplet between δ 7.37-7.41 for three protons of C_8, C_5 and C_7. respectively. Triplet observed at δ 7.72 (J = 9Hz) for a proton at $C_{6'}$ and doublet observed at 8.20 (J = 9Hz) for a proton at $C_{5'}$. Broad singlet observed at δ 10.70 was observed for one proton of >NH which was D_2O exchangeable. The ^{13}C NMR spectrum of compound **3b** in $CDCl_3$ showed signal at δ 17.93 and 18.58 for two methyl carbon at C_4 and C_7 respectively. Signal at δ 159.78, 160.71 and 176.09 for two >C=O of amide, coumarin and one >C=S respectively and other signal at 153.48 (C_{8a}), 151.99 (C_4), 140.66, 139.01, 136.15, 134.10, 128.91, 128.43, 125.41, 124.68, 119.41, 116.12, 115.03, 114.88. The mass spectrum of **3b** showed molecular ion peak at *m/z* 350. The peaks of fragment ions appearing at *m/z* 335, 317, 289, 162, 145, 119 and 92 gave further evidence in support of the structure assigned. Condensation of the glycine and thioglycolic acid separately with 6-coumarinyl isothiocyanate resulted in the formation of 3-(2-oxo-*2H*-benzopyran-6-yl)-2-thioxo-imidazolidin-4-one **4a-c** and 3-(2-oxo-*2H*-benzopyran-6-yl)-2-thioxothiazo



1a-7a. $R_1 = H$, $R_2 = CH_3$
1b-7b. $R_1 = CH_3$, $R_2 = CH_3$
1c-7c. $R_1 = H$, $R_2 = H$

a) CS_2, I_2/pyridine b) anthranilic acid c) glycine, TEA d) thioglycolic acid, TEA e) *o*-aminophenol, pyridine f) *o*-aminothiophenol, DMF

Scheme I

lidin-4-one **5a-c** respectively. The IR spectrum of compound **4c** in KBr showed band at 3214 cm^{-1} for >NH stretching, at 1763, 1733 cm^{-1} for >C=O stretching, along with other bands at 1572, 1533, 1442, 1281, 1183, 1102, 823, cm^{-1} *etc*. The ^1H NMR spectrum of compound **4c** in DMSO-d_6 showed presence of singlet at δ 4.30 for two protons of –CO-CH_2- group. A doublet at 6.53 (J = 9Hz) for one proton at C_3 and doublet observed at 8.09 (J = 9Hz) for one proton at C_4. It showed multiplet between δ 7.52-7.67 for three protons of C_5, C_8 and C_7 respectively. Singlet observed at δ 10.49 was observed for one proton of >NH which was D_2O exchangeable. The mass spectrum of **4c** showed molecular ion peak at *m/z* 260 itself has been observed as the base peak, along with other *m/z* peaks at 249, 227, 203, 188, 175, 160, 146,

Table I —- Characterization data of compounds **3-7a-c**

Compd	Mol. Formula	m.p. °C	Yield (%)	Found (%) (Required)			
				C	H	N	S
3a	$C_{18}H_{12}N_2O_3S$	248-50	61	64.12	3.49	8.20	9.24
				(64.27	3.60	8.33	9.53)
3b	$C_{19}H_{14}N_2O_3S$	284-86	68	65.29	4.14	8.01	9.23
				(65.13	4.03	7.99	9.15)
3c	$C_{17}H_{10}N_2O_3S$	228-31	63	63.23	3.27	8.88	10.02
				(63.35	3.13	8.69	9.95)
4a	$C_{13}H_{10}N_2O_3S$	218-20	74	56.80	3.88	10.44	11.54
				(56.92	3.67	10.21	11.69)
4b	$C_{14}H_{12}N_2O_3S$	252-54	69	58.14	4.14	9.54	10.99
				(58.32	4.20	9.72	11.12)
4c	$C_{12}H_8N_2O_3S$	164-66	72	55.66	3.21	11.02	12.16
				(55.38	3.10	10.76	12.32)
5a	$C_{13}H_9NO_3S_2$	280-83	67	53.59	3.11	4.81	22.01
				(53.55	3.10	4.83	22.11)
5b	$C_{14}H_{11}NO_3S_2$	292-95	66	55.07	3.63	4.59	21.00
				(55.12	3.68	4.56	21.03)
5c	$C_{12}H_7NO_3S_2$	233-36	72	51.97	2.54	5.05	23.12
				(51.87	2.53	5.03	23.11)
6a	$C_{17}H_{12}N_2O_3$	158-60	60	69.66	4.50	9.55	
				(69.86	4.14	9.58	-)
6b	$C_{18}H_{14}N_2O_3$	169-72	61	70.33	4.55	9.22	
				(70.58	4.61	9.15	-)
6c	$C_{16}H_{10}N_2O_3$	145-47	64	69.14	3.66	10.11	
				(69.06	3.62	10.07	-)
7a	$C_{17}H_{12}N_2O_2S$	160-62	65	66.29	3.89	9.04	10.49
				(66.22	3.92	9.08	10.40)
7b	$C_{18}H_{14}N_2O_2S$	175-78	59	67.16	4.34	8.64	9.99
				(67.06	4.38	8.69	9.95)
7c	$C_{16}H_{10}N_2O_2S$	155-57	64	65.21	3.44	9.55	10.89
				(65.29	3.42	9.52	10.89)

Table II — Antibacterial activity of compounds **3-7a-c**

Compd	Zone of inhibition in mm					
	S. aureus		*S. typhi*		*E. coli*	
	50μg	100μg	50μg	100μg	50μg	100μg
3a	16	18	17	19	17	20
3b	17	19	15	16	18	20
3c	14	15	15	17	15	17
4a	15	17	13	15	16	17
4b	13	14	12	14	13	15
4c	11	12	13	14	12	14
5a	14	15	13	15	15	17
5b	13	15	12	14	15	16
5c	12	14	13	15	15	17
6a	11	15	13	14	16	20
6b	14	16	15	17	17	21
6c	10	14	13	16	16	18
7a	15	16	18	20	18	20
7b	16	18	17	21	18	20
7c	12	14	14	16	15	18

Disc size: 6.35mm Standard: streptomycin Control: DMSO
Duration: 24 hr resistant (11mm/less) ensitive(15mm/more) intermediate(12-14 mm)

133, 120, 117, 103 and 89. Condensation of the *o*-amino phenol with 6-coumarinyl isothiocyanate resulted in the formation of 6-(benzooxazol-2-ylamino)-benzopyran-2-one **6a-c** (**Scheme I**) The IR spectrum of compound **6b** in KBr showed band at 3360 for >NH stretching, at 1691 for >C=O stretching, along with other bands at 1556, 1447, 1386, 1258, 1053, 861 cm^{-1}, *etc*. The ^1H NMR spectrum of compound **6b** in $CDCl_3$ showed presence of singlet at δ 2.19 for three protons of methyl group at C_4, a singlet at 2.30 for three protons of methyl group at C_7. Singlet observed at 6.15 for one proton at C_3. It showed multiplet between δ 6.74-7.31 for six protons of C_5, C_8 and four aromatic protons. Broad singlet observed at 3.61 was observed for one proton of >NH which was D_2O exchangeable. The ^{13}C NMR spectrum of compound **6b** in $CDCl_3$ showed signal at δ 17.81 and 18.67 for two methyl carbon at C_4 and C_7 respectively. Signal at 161.75 for >C=O of coumarin and other signal at 152.23(C_{8a}), 146.77(C_4), 141.42, 128.37, 125.08, 124.46, 124.06, 118.65, 118.32, 114.71, 114.29, 110.32, 110.20, 108.29, 108.03. The mass spectrum of **6b** showed molecular ion peak at *m/z* 306 itself has been observed as the base peak, along with other peaks at *m/z* 289, 277, 261, 249, 206, 199, 189, 171, 160, 145, 124, 115, 103, 91, and 77. Condensation of the *o*-aminothiophenol with 6-coumarinyl isothiocyanate resulted in the formation of 6-(benzothiazol-2-ylamino)-benzopyran-2-one **7a-c** (**Table I**) The IR spectrum of compound **7a** in KBr showed band at 3348 cm^{-1} for >NH stretching, at 1722 cm^{-1} for >C=O stretching, along with other bands at 1623, 1533, 1444, 1186, 1123, 1016, 824, 750 cm^{-1} etc. The ^1H NMR spectrum of the same in $CDCl_3$ showed singlet at δ 2.45 for three protons of methyl group at C_7. Doublet observed at 6.42 (J = 9Hz) and 7.70 (J = 9Hz) for a proton at C_3 and C_4 respectively. Multiplet observed at 7.14-7.60 for five protons of C_8 and four aromatic protons. Singlet observed at 7.94 for one proton of C_5. Singlet at 4.38 was observed for one proton of >NH which was D_2O exchangeable. The mass spectrum of **7a** showed molecular ion peak at *m/z* 308 itself has been observed as the base peak, along with other peaks at *m/z* 293, 275, 251, 216, 185, 157, 146, 126, 93, and 77.

Antimicrobial activity

All the synthesized compounds **3a-c**, **4a-c**, **5a-c**, **6a-c**, and **7a-c** were screened for their antibacterial activity (**Table II**) against *S. aureus*, *S. typhi* and *E. coli* (**Table I**) by the drug diffusion method[18]. The zone of inhibition was measured in mm and was compared with standard drug. DMSO was used as a blank and streptomycin was used as antibacterial standard. All the compounds were tested at 50 μgm/mL and 100 μg/mL concentration.

From the antimicrobial screening of the compounds **3-7a-c** it could be observed that quinazolinone and benzothiazole derivatives have significant anti-bacterial activities and other heterocycles have comparable antibacterial activities.

Experimental Section

General: Melting points were taken in open capillaries and are uncorrected. Purity of the compounds was checked on TLC. IR spectra (cm^{-1}) were recorded on a Perkin-Elmer FTIR; NMR (^1H and ^{13}C) on a 300 MHz JEOL NMR AL300 using TMS as standard; and mass spectra on a Shimadzu GC-MS QP-2010.

Synthesis of 3-(2-oxo-*2H*-benzopyran-6-yl)-2-thioxo-2,3-dihydro-*1H*-quinazolin-4-one 3a-c

General Procedure

Coumarinyl isothiocyanate **2a-c** (0.01 mole) and anthranilic acid (0.01 mole) in ethanol (20 mL) was refluxed on water-bath for 6 hr, it was then allowed to cool at room temperature. The solid obtained was filtered, washed with water, dried and crystallized from proper solvent to give 3-(2-oxo-*2H*-benzopyran-6-yl)-2-thioxo-2,3-dihydro-*1H*-quinazolin-4-one **3a-c**.

Synthesis of 3-(2-oxo-*2H*-benzopyran-6-yl)-2-thioxo-imidazolidin-4-one 4a-d and 3-(2-oxo-*2H*-benzopyran-6-yl)-2-thioxo-thiazolidin-4-one 5a-d

General Procedure

A mixture of glycine or thioglycolic acid (0.002 mole) and coumarinyl isothiocyanate **2a-c** (0.002 mole) and TEA (1 mL) in ethanol (15 mL) was refluxed for 6 hr, excess of ethanol was removed by distillation and neutralize by using 1:1 HCl, then reaction-mixture was poured into ice-cold water, precipitate was filtered off and recrystallized from proper solvent to give 3-(2-oxo-*2H*-benzopyran-6-yl)-2-thioxoimidazolidin-4-one **4a-d** and 3-(2-oxo-*2H*-benzopyran-6-yl)-2-thioxo-thiazolidin-4-one **5a-d** respectively.

Synthesis of 6-(benzooxazol-2-ylamino)-benzopyran-2-one 6a-c

General Procedure

A mixture of *o*-aminophenol (0.002 mole) and coumarinyl isothiocyanate **2a-c** (0.002 mole) and pyridine (1 mL) in absolute alcohol (15 mL) was refluxed for 24 hr, excess of ethanol was removed by distillation. After cooling, the reaction-mixture was poured into ice-cold water. The precipitate was filtered off and recrystallized from ethanol.

Synthesis of 6-(benzothiazol-2-ylamino)-benzopyran-2-one 7a-c. General Procedure

A mixture of *o*-aminothiophenol (0.002 mole) and the coumarinyl isothiocyanate **2a-c** (0.002 mole) in DMF (15 mL) was refluxed for 8 hr. After cooling, the reaction-mixture was poured into ice-cold water, The precipitate was filtered off and recrystallized from ethanol.

Spectral Data

3a: IR (KBr): 3160 (-NH), 3033, 2984 (-CH), 1715 (>C=O), 1633, 1585, 1414, 1192, 750 cm^{-1}; ^1H NMR ($CDCl_3$): δ 2.45 (s, 3H, C_7-CH_3), 6.49 (d, 1H, *J* = 9Hz, C_3-H), 7.24 (d, 1H, *J* = 9Hz, $C_{8'}$ -H), 7.35-7.64 (m, 3H, C_8-H, C_5-H, C_4-H and $C_{7'}$-H), 7.74 (t, 1H, *J* = 9Hz, $C_{6'}$-H), 8.21 (d, 1H, *J* = 9Hz, $C_{5'}$-H), 10.71 (br, 1H, NH-exchangeable); **3b**: IR (KBr): 3175 (-NH), 3019, 2965 (-CH), 1705 (>C=O), 1623, 1541, 1418, 1202, 758 cm^{-1}; ^1H NMR ($CDCl_3$): δ 2.28 (s, 3H, C_4-CH_3), 2.40 (s, 3H, C_7-CH_3), 6.32 (s, 1H, C_3-H), 7.23 (d, 1H, *J* = 9Hz, $C_{8'}$ -H), 7.37-7.41 (m, 3H, C_8-H, C_5-H and $C_{7'}$-H), 7.72 (t, 1H, *J* = 9Hz, $C_{6'}$-H), 8.20 (d, 1H, *J* = 9Hz, $C_{5'}$-H), 10.70 (br, 1H, NH-exchangeable); ^{13}C NMR ($CDCl_3$): δ 17.93 (C_4-CH_3), 18.58 (C_7-CH_3), 159.78 (-N-C=O), 160.71 (C_2-C=O), 176.09 (>C=S), 153.48 (C_{8a}), 151.99 (C_4), 140.66, 139.01, 136.15, 134.10, 128.91, 28.43, 125.41, 124.68, 119.41, 116.12, 115.03, 114.88; Mass (*m/z* %): M^+ 350(95), 335(59), 317(100), 305(4), 289(5), 162(7), 145(9), 119(8), 92(10); **3c**: IR (KBr): 3165 (-NH), 3023, 2979 (-CH), 1720 (>C=O), 1622, 1565, 1444, 1252, 733 cm^{-1}; ^1H NMR ($CDCl_3$): δ 6.55 (d, 1H, *J* = 9Hz, C_3-H), 7.24 (d, 1H, *J* = 9Hz, $C_{8'}$ -H), 7.36-7.69 (m, 5H, C_8-H, C_5-H, C_7-H, C_4-H and $C_{7'}$-H), 7.73 (t, 1H, *J* = 9Hz, $C_{6'}$-H), 8.20 (d, 1H, *J* = 9Hz, $C_{5'}$-H), 10.69 (br, 1H, NH-exchangeable); **4a**: IR(KBr): 3188 (-NH), 3024, 2978 (-CH), 1748,1696 (>C=O), 1625, 1543, 1493, 1247, 1198, 1125, 1050, 904 cm^{-1}; ^1H NMR (DMSO-d_6): δ 2.18 (s, 3H, C_7-CH_3), 4.35 (s, 2H, –CO-CH_2-), 6.46 (d, 1H, *J* = 9Hz, C_3-H), 7.43 (s, 1H, C_8-H), 7.56 (s, 1H, C_5-H), 8.03 (d, 1H, *J* = 9Hz, C_4-H), 10.51 (br, 1H, NH-exchangeable); **4b**: IR(KBr): 3176 (-NH), 2943 (-CH), 1733, 1685(>C=O), 1634, 1557, 1424, 1266, 1134, 1122, 1059, 745 cm^{-1}; **4c**: IR (KBr): 3214 (-NH), 2918 (-CH), 1763,1733(>C=O), 1572, 1533, 1442, 1281, 1183, 1102, 823 cm^{-1}; ^1H NMR (DMSO-d_6): δ 4.30 (s, 2H, –CO-CH_2-), 6.53 (d, 1H, *J* = 9Hz, C_3-H), 7.52-7.67 (m, 3H, C_5-H, C_8-H and C_7-H), 8.09 (d, 1H, *J* = 9Hz, C_4-H), 10.49 (br, 1H, NH-exchangeable); Mass (*m/z* %): M^+ 260(100), 249(14), 203(33), 188(17), 175(39), 160(46), 146(13), 133(17), 117(14), 103(14), 89(26), 73(23), 63(22); **5a**: IR(KBr): 2981, 2926 (–CH), 1734 (>C=O), 1625, 1260, 1239, 1223, 1192 cm^{-1}; ^1H NMR (DMSO-d_6): δ 2.12 (s, 3H, C_7-CH_3), 4.38, 4.55 (2d, each 1H, *J* = 21Hz, S-CH_2), 6.47 (d, 1H, *J* = 9Hz, C_3-H), 7.46 (s, 1H, C_8-H), 7.59 (s, 1H, C_5-H), 8.01 (d, 1H, *J* = 9Hz, C_4-H); Mass (*m/z* %): M^+ 291(100), 276(16), 258(37), 249(13), 230(9), 217(47), 185(41), 173(10), 157(32), 130(12), 102(14), 89(6), 77(16); **5b**: IR(KBr): 2978, 2944 (–CH), 1723 (>C=O), 1646, 1276, 1268, 1224, 1157, 745 cm^{-1}; ^1H NMR ($CDCl_3$): δ 2.21 (s, 3H, C_4-CH_3), 2.40 (s, 3H, C_7-CH_3), 4.26 (s, 2H, S-CH_2), 6.30 (s, 1H, C_3-H), 7.30 (s, 1H, C_8-H), 7.34 (s, 1H, C_5-H); **5c**: IR(KBr): 2987, 2936 (–CH), 1744 (>C=O), 1632, 1246, 1221, 1153 cm^{-1}; ^1H NMR ($CDCl_3$): δ 4.23 (s, 2H, S-CH_2), 6.49 (d, 1H, *J* = 9Hz, C_3-H), 7.38-7.47 (m, 3H, C_5-H, C_8-H and C_7-H), 7.69 (d, 1H, *J* = 9Hz, C_4-H); **6a**: IR(KBr): 3347 (-NH), 1709 (>C=O), 1526, 1467, 1345, 1288, 1032, 768 cm^{-1}; ^1H NMR (DMSO-d_6): δ 2.14 (s, 3H, C_7-CH_3), 6.26 (d, 1H, *J* = 9Hz, C_3-H), 6.77-7.35 (m, 6H, C_8-H, C_5-H, and four CH- aromatic protons.), 7.84 (d, 1H, *J* = 9Hz, C_4-H), 5.01 (br, 1H, NH-exchangeable); **6b**: IR(KBr): 3360 (-NH), 1691 (>C=O), 1556, 1447, 1386, 1258, 1053, 861 cm^{-1}; ^1H NMR ($CDCl_3$): δ 2.19 (s, 3H, C_4-CH_3), 2.30 (s, 3H, C_7-CH_3), 6.15 (s, 1H, C_3-H), 6.74-7.31 (m, 6H, C_8-H, C_5-H, and four aromatic protons.), 3.61 (br, 1H, NH-exchangeable); ^{13}C NMR ($CDCl_3$): 17.81 (C_4-CH_3), 18.67 (C_7-CH_3), 161.75 (C_2-C=O), 152.23 (C_{8a}), 146.77 (C_4), 141.42, 128.37, 125.08, 124.46, 124.06, 118.65, 118.32, 114.71, 114.29, 110.32, 110.20, 108.29, 108.03; Mass (*m/z* %): M^+ 306(100), 289(16), 277(37), 261(13), 249(9), 206(47), 199(41), 189(10), 171(32), 160(12), 145(14), 124(6), 115(16), 103(16), 91(16), 77(16); **6c**: IR(KBr): 3364(-NH), 1721(>C=O), 1543, 1424, 1366, 1223, 1046, 766 cm^{-1}; **7a**: IR(KBr): 3348 (-NH), 3061, 2926 (–CH), 1722 (>C=O), 1623, 1533, 1444, 1186, 1123, 1016, 824, 750 cm^{-1}; ^1H NMR ($CDCl_3$): δ 2.45 (s, 3H, C_7-CH_3), 6.42 (d, 1H, *J* = 9Hz, C_3-H), 7.14-7.60 (m, 5H, C_8-H and four aromatic protons), 7.70 (d, 1H, *J* = 9Hz, C_4-H), 7.94 (s, 1H, C_5-H), 4.38 (br, 1H, NH-exchangeable); Mass (*m/z* %): M^+ 308(100), 293(20), 275(14), 251(6), 216(6), 185(8), 157(7), 146(7), 126(11), 93(7), 77(7); **7b**: IR(KBr): 3258 (-NH), 1702 (>C=O), 1624, 1571, 1530, 1446, 1421, 1158, 1053, 754 cm^{-1}; ^1H NMR ($CDCl_3$): δ 2.44 (s, 6H, C_4, C_7-CH_3), 6.31(s, 1H, C_3-H), 7.17-7.63 (m, 5H, C_8-H and four CH- aromatic protons), 8.01 (s, 1H, C_5-H), 4.43 (br, 1H, NH-exchangeable); **7c**: IR(KBr): 3299 (-NH), 1707 (>C=O), 1618, 1585, 1542, 1444, 1259, 1182, 1149, 751 cm^{-1}; ^1H NMR ($CDCl_3$): δ 6.47 (d, 1H, *J* 9Hz, C_3-H), 7.21-7.75 (m, 7H, C_8-H, C_4-H, C_7-H and four aromatic protons), 7.95 (s, 1H, C_5-H), 4.38 (br, 1H, NH-exchangeable).

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