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Synthesis and regiospecific methylation of new benzimidazolyl β-ketosulfones and β-hydroxysulfones

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Condensation of 2-mercaptobenzimidazoles **1a-e** with α-bromoacetylbenzene **2** resulting in benzimidazolyl β-ketosulfides **3a-e** followed by oxidation with hydrogen peroxide giving β-ketosulfones **7a-e** and subsequent reduction with sodium borohydride yielding β-hydroxysulfones **9a-e** is described. Reaction of **3a**, **8a, 7a** and **9a** with DMS in the presence of potassium carbonate as base and tetrabutylammonium bromide (TBAB) as phase transfer catalyst, results in the regiospecific N-methylation of the NH of benzimidazole moiety giving **11a**, **10a**, **13a** and **12a** respectively.

Keywords: Regiospecific alkylation, phase-transfer catalyst, 2-mercaptobenzimidazole

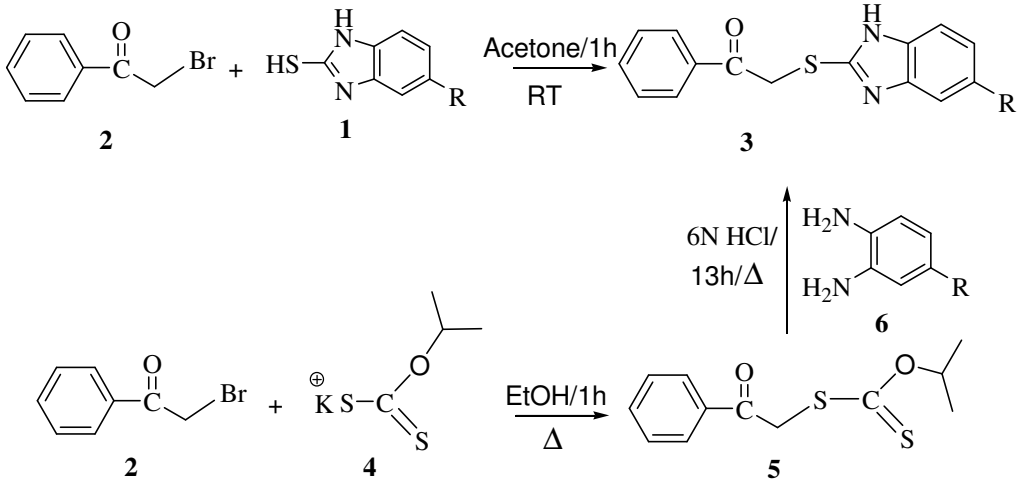
The β-ketosulfone moiety is readily available from a variety of precursor functionalities[1] and displays a broad range of synthetic versatility[2-6]. These are also important group of intermediates in Michael and Knoevenagel reactions[7], and are valuable precursors in the synthesis of acetylenes, allenes, chalcones[8] and vinylsulfones[9]. The β-ketosulfone moiety may be synthesized from a variety of precursors, for example from cyanosulfones *via* the Thorpe reaction[10], by oxidation of sulfides or sulfoxides[11], or from esters through Claisen condensations[12]. Reductions of β-ketosulfone have attracted considerable attention from synthetic chemist in the past decade[13]. The chiral β-hydroxy-sulfones formed, are of great utility in organic synthesis; they have been used in the preparation of many compounds, but find practical use in the synthesis of chiral lactones[14] and 2,5-disubstitutedtetrahydrofurans[15]. Benzimidazoles are very important class of compounds due to their wide spectrum of biological activity, behaving as anti-hypertensive, anti-viral, anti-fungal, anti-tumor and anti-helminthic agents in veterinary medicine[16]. We report herein the synthesis of novel benzimidazole containing β-hydroxysulfones from its corresponding β-ketosulfones by performing the reductions with $NaBH_4$ in a protic solvent, *i.e.*, methanol[17], and also report on regiospecific alkylation of the products.

Results and Discussion

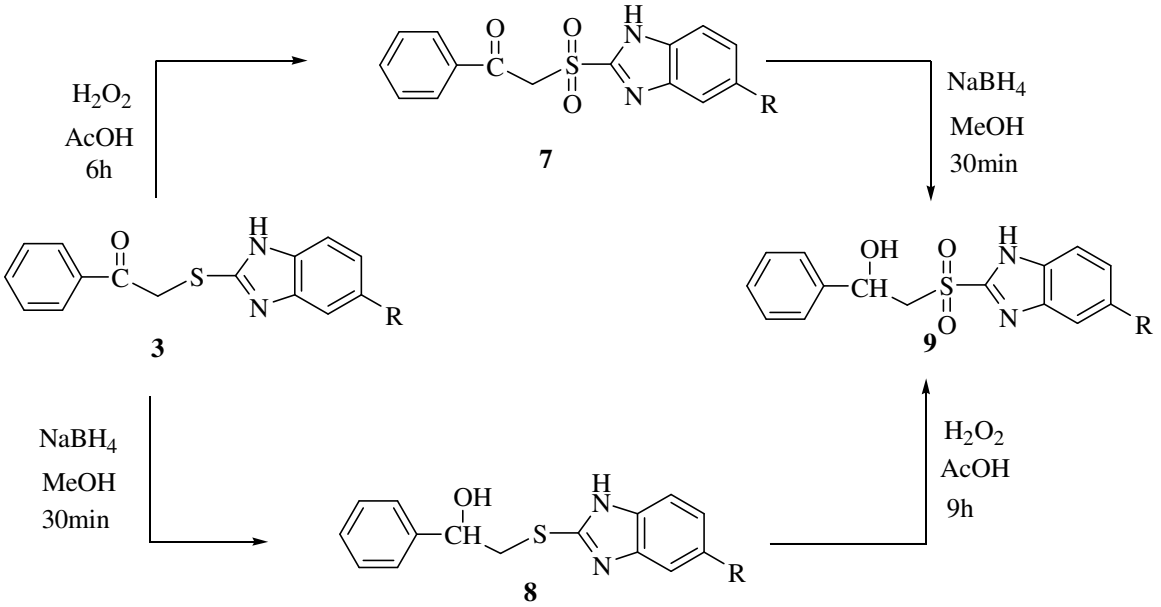
Condensation of 5-substituted-2-mercaptobenzimidazole **1** (ref. 18) with α-bromoacetylbenzene **2** (ref. 19) gave a product, which has been characterized as 2-benzoylmethylthiobenzimidazole **3**. Alternatively, the compound **3**, could also be prepared by the initial reaction of **2** with isopropyl xanthate **4** to obtain **5**, followed by condensation of the latter with 4-substituted-o-phenylenediamines **6** (*i.e.*, R= H, CH_3, OCH_3, NO_2, Cl) under Phillip's conditions[20] (**Scheme I**).

Treatment of **3** with excess hydrogen peroxide in acetic acid gave the corresponding β-ketosulfone **7**. The latter on treatment with $NaBH_4$ in methanol at RT gave the corresponding β-hydroxysulfone **9**. It is evident that $NaBH_4$ does not touch the sulfone functionality but reduces the keto group to the secondary hydroxyl group, an observation found to be in conformity with the general behaviour[21]. Alternatively, **9** could also be prepared by the initial reduction of **3** with $NaBH_4$ in methanol yielding β-hydroxysulfide **8** followed by oxidation with hydrogen peroxide in acetic acid. These reactions are briefly summarized in **Scheme II**.

The compound 2-(1*H*-benzimidazol-2-sulfonyl)-1-phenyl-ethanone **7a** (*i.e.*, **7**, R=H) on treatment with dimethyl sulfate in the presence of potassium carbonate as base and tetrabutylammonium bromide (TBAB) as phase transfer catalyst gave a product



Scheme I



R = H, CH_3, OCH_3, NO_2, Cl

Scheme II

which has been characterized as the *N*-methylated derivative *i.e.*, 2-(*N*-methyl-benzimidazole-2-sulfonyl)-1-phenyl-ethanone **10a** (*i.e.*, **10**, R = H) rather than the C-methylated derivative involving methylation of the α carbon of β-ketosulfone, on the basis of spectral data. Thus, the reactions seems to proceed regiospecifically such that methylation takes place only on benzimidazole –NH– rather than on the α carbon of β-ketosulfone. To put all doubts at rest, **10a** could also be prepared alternatively by treatment of 1-(*N*-methyl-benzimidazol-2-sulfayl)-1-phenyl-ethanone **11a** (*i.e.*, **11**, R = H) with H_2O_2 in acetic acid. The latter *i.e.*, **11a** in turn was synthesized by the reaction of **3a** (*i.e.*, **3**, R = H) with DMS under phase transfer catalyst conditions.

Reaction of 2-(1*H*-benzimidazol-2-ylsulfanyl)-1-phenyl-ethanol **8a** (*i.e.*, **8**, R = H) with dimethyl sulfate using potassium carbonate as base and

tertabutylammonium bromide as phase-transfer catalyst gave a TLC pure product, which could be either of the regiospecifically methylated derivatives, *i.e. N*-methyl or *O*-methyl derivative. The product was, however, found to be the region-specifically *N*-methylated derivative *i.e.*, 2-(*N*-methyl-benzimidazol-2-sulfanyl)-1-phenylethanol **12a** (*i.e.*, **12**, R = H) on the basis of spectral data. Furthermore, **12a** could also be prepared alternatively from **11a**, when treated with sodium borohydride in methanol at RT.

The compound **12a** on oxidation with hydrogen peroxide in acetic acid gave the corresponding β-hydroxysulfone **13a**. Alternatively, the compound **13,** could also be independently prepared by the reduction of **10a** with $NaBH_4$ and reaction of **9a** (*i.e.*, **9**, R = H) with dimethyl sulfate. All the above reactions are nicely shown in the summary **Scheme III**.

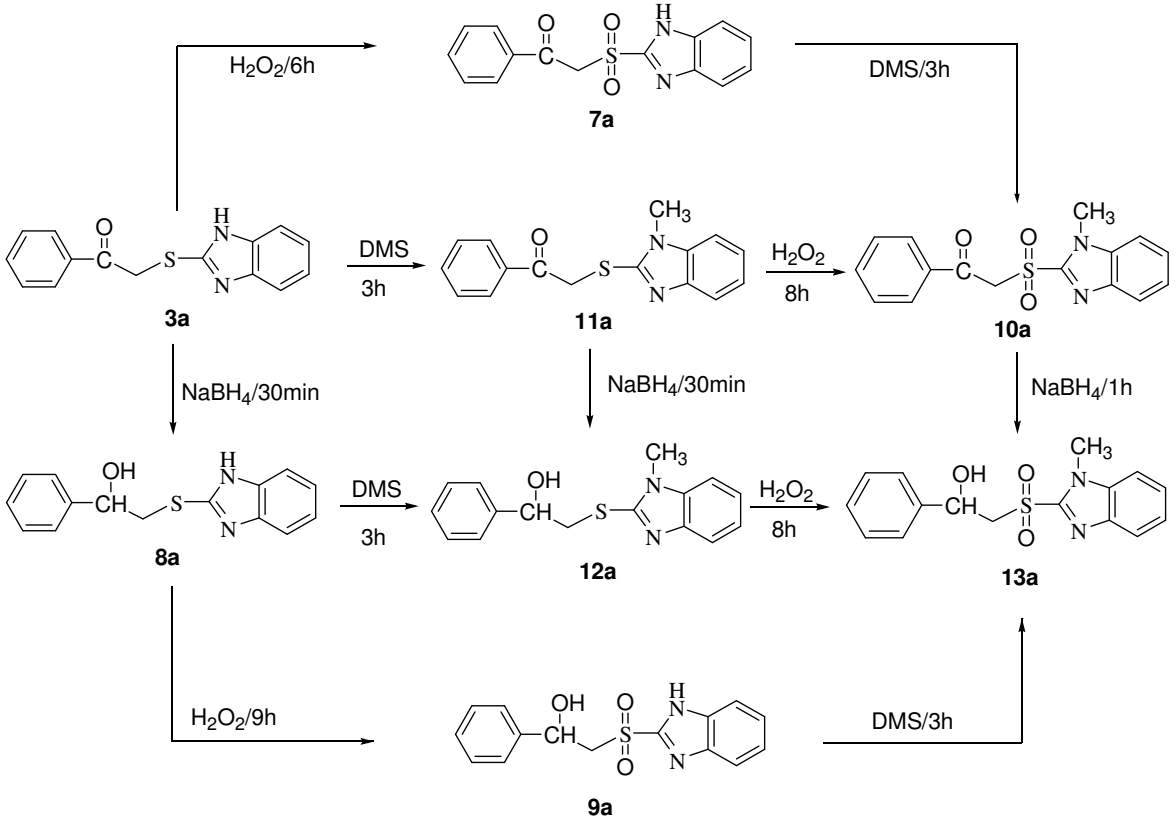
In conclusion we have synthesized novel β-hydroxysulfones from β-ketosulfones by using sodium borohydride, a mild reducing agent in a protic solvent like methanol. The research paper also emphasizes the regiospecific alkylation studies of β-hydroxy-sulfides and β-hydroxysulfones using a phase transfer catalyst.

Experimental Section

All reagents were obtained commercially and were of the LR or AR quality and used as such without further purification. Solvents were freshly distilled and used. Melting points were determined in open capillaries in sulfuric acid bath and are uncorrected. TLC analyses were done on glass plates coated with silica gel GF-254 and spotting was done using Iodine/UV lamp. IR spectra were recorded on a Perkin-Elmer model 446 instrument in KBr phase. ^{1}H NMR were recorded in $CDCl_3$/DMSO using 400 MHz Varian Gemini spectrometer and Mass spectra were recorded on a LCMS spectrometer, model HP-5989A.

Synthesis of 3 from 1 and 2: A mixture of **1** (0.01 mol), **2** (1.99 g, 0.01 mol) and acetone (50 mL) was stirred at RT for 1 hr. At the end of this period, the reaction-mixture was poured into ice water. The separated solid was filtered, washed with water, dried and recrystallized from ethyl acetate to obtain pure **3.**

3a (*i.e.*, **3**, R = H); Yield 2.19 g (82%), m.p. 165-68°C; IR (KBr): 3057-2804 (-NH-), 1680 (-CO-) cm^{-1}; ^{1}H NMR (DMSO-d_6/TMS): δ 5.26 (s, 2H, -CH_2-S-), 7.22-8.09 (m, 8H, Ar-H); ^{13}C NMR (DMSO-d_6/TMS): δ 40.17 (-CH_2-S-), 122.1-150.2 (Aromatic carbons), 194.10 (-CO-); MS (CI): *m/z* 269 $[M+H]^+$.



Scheme III

3b (*i.e.*, **3**, R = CH_3). Yield 2.34 g (83%), m.p. 176-78°C (EtOAc); IR (KBr): 3200-2935 cm^{-1}; ^1H NMR spectrum (DMSO-d_6/TMS): δ 2.30 (s, 3H, Ar-CH_3), 4.82 (s, 2H, -CH_2-S-), 7.20-7.90 (m, 8H, Ar-H), 12.60 (s, 1H, D_2O exch., –NH); MS (CI): *m/z* 283 $[M+H]^+$. Anal. Calcd. for $C_{16}H_{14}N_2OS$: C, 68.06; H, 5.00; N, 9.92. Found: C, 65.08; H, 4.24; N, 9.31%.

3c (*i.e.*, **3**, R = OCH_3). Yield 2.62 g (80%), m.p. 150-55°C (EtOAc); IR (KBr): 3430-2933 cm^{-1} (–NH-); PMR spectrum (DMSO-d_6/TMS): δ 3.00 (s, 3H, Ar-OCH_3), 4.80 (s, 2H, -CH_2-S-), 7.00-8.00 (m, 8H, Ar-H), 12.50 (s, 1H, D_2O exch., –NH); MS (CI): *m/z* 329 $[M+H]^+$.

3d (*i.e.*, **3**, R = NO_2). Yield 2.59 g (83%), m.p. 128-32°C (EtOAc); IR (KBr): 3207-2859 cm^{-1} (–NH-); PMR spectrum (DMSO-d_6/TMS): δ 4.90 (s, 2H, -CH_2-S-), 7.30-8.40 (m, 8H, Ar-H), 12.92 (s, 1H, D_2O exch., –NH); MS (CI): *m/z* 314 $[M+H]^+$.

3e (*i.e.*, **3**, R = Cl). Yield 2.27 g (75%), m.p. 198-04°C (EtOAc); IR (KBr): 3437 cm^{-1} (–NH-); PMR spectrum (DMSO-d_6/TMS): δ 4.87 (s, 2H, -CH_2-S-), 7.25-8.30 (m, 8H, Ar-H), 12.87 (s, 1H, D_2O exch., –NH); MS (CI): *m/z* 303 $[M+H]^+$.

Synthesis of 5 from 2 and 4: A mixture of **2** (0.01 mol), potassium isopropylxanthate **4** (1.60 g, 0.01 mol) and ethanol (30 mL) was refluxed for 1 hr. Then the reaction-mixture was cooled to RT. The mixture was filtered to remove KBr salt and washed with ethanol (5 mL). The filtrate was distilled off and the residue diluted with water. The separated solid was filtered, washed with water and dried. It was recrystallized from ethanol to obtain pure **5**.

5a; Yield 2.0 g (83%), m.p. 145-47°C; IR (KBr): 1610 (-CO-), 1250 (-(C=S)-), 1048 (-S-(C=S)-) cm^{-1}; ^1H NMR (DMSO-d_6/TMS): δ 1.19 (d, 6H, -CH-$(CH_3)_2$), 4.72 (s, 2H, -CO-CH_2), 5.55 (q, 1H, -OCH-), 7.53-8.02 (m, 5H, Ar-H); MS (CI): *m/z* 243 $[M+H]^+$.

Alternate synthesis of 3 from 5 and 6: A mixture of **6** (0.01 mol) and **5** (0.01 mol) was refluxed in 6 N HCl (100 mL) for 12-14 hr. At the end of this period, the reaction mass was diluted with water and *p*H was adjusted to ≥7.0 with aq. NH_3. The separated solid was filtered, washed with water and dried to obtain the crude product.

3a (*i.e.*, **3**, R = H). Yield 2.30 g (85%), m.p. 165-67°C (EtOH).

3b (*i.e.*, **3**, R = CH_3). Yield 2.00 g (70%), m.p. 173-77°C (MeOH).

3c (*i.e.*, **3**, R = OCH_3). Yield 2.3 g (67%), m.p. 151-55°C (MeOH).

3d (*i.e.*, **3**, R = NO_2). Yield 2.28 g (72%), m.p. 127-32°C (MeOH).

3e (*i.e.*, **3**, R = Cl). Yield 2.33 g (77%), m.p. 199-02°C (MeOH).

General procedure for oxidation with H_2O_2: To a suspension of compound (0.01 mol) in acetic acid (25 mL) was added H_2O_2 (30%) (2.0 mL) at 5-10°C. After the completion of addition, mixture was heated on a steam-bath (at 100°C) until the reaction is complete. At the end of the reaction, the acetic acid was evaporated, and the reaction-mixture was diluted with ice-water. The separated solid was filtered, washed with water, dried and the crude product was purified by column chromatography over basic alumina using hexane:ethyl acetate mixture (100:0 to 90:10) as eluent to afford pure oxidized compound.

7a (*i.e.*, **7**, R = H); Yield 2.19 g (73%), m.p. 174-78°C; IR (KBr): 3428-2926 (-NH-), 1694 (-CO-), 1331 cm^{-1} (-SO_2-); ^1H NMR (DMSO-d_6/TMS): δ 5.36 (s, 2H, -CH_2-S), 6.91-7.86 (m, 9H, Ar-H), 13.80 (s, 1H, D_2O exch., –NH); MS (CI): *m/z* 301 $[M+H]^+$.

7b (*i.e.*, **7**, R = CH_3); Yield 2.07 g (66%), m.p. 126-29°C; IR (KBr): 3200-2935 (-NH-), 1686 (-CO-), 1332 cm^{-1} (-SO_2-); ^1H NMR (DMSO-d_6/TMS): δ 2.30 (s, 3H, Ar-CH_3), 5.22 (s, 2H, -CH_2-S), 7.20-7.90 (m, 8H, Ar-H), 12.60 (s, 1H, D_2O exch., –NH); MS (CI): *m/z* 315 $[M+H]^+$.

7c (*i.e.*, **7**, R = OCH_3); Yield 2.37 g (72%), m.p. 222-25°C; IR (KBr): 3430-2933 (-NH-), 1680 (-CO-), 1340 cm^{-1} (-SO_2-); ^1H NMR (DMSO-d_6/TMS): δ 2.40 (s, 3H, Ar-OCH_3), 5.10 (s, 2H, -CH_2-S), 7.01-8.00 (m, 8H, Ar-H), 12.50 (s, 1H, D_2O exch., –NH); MS (CI): *m/z* 331 $[M+H]^+$.

7d (*i.e.*, **7**, R = NO_2); Yield 2.38 g (69%), m.p. 128-33°C; IR (KBr): 3207-2859 (-NH-), 1685 (-CO-), 1338 cm^{-1} (-SO_2-); ^1H NMR (DMSO-d_6/TMS): δ 5.05 (s, 2H, -CH_2-S), 7.30-8.40 (m, 8H, Ar-H), 12.93 (s, 1H, D_2O exch., –NH); MS (CI): *m/z* 346 $[M+H]^+$.

7e (*i.e.*, **7**, R = Cl); Yield 2.27 g (68%), m.p. 187-92°C; IR (KBr): 3437-2850 (-NH-), 1680 (-CO-), 1331, 1134 cm^{-1} (-SO_2-); ^1H NMR (DMSO-d_6/TMS): δ 5.28 (s, 2H, -CH_2-S), 7.25-8.30 (m, 8H, Ar-H), 13.00 (s, 1H, D_2O exch., –NH); MS (CI): *m/z* 335 $[M+H]^+$.

9a (*i.e.*, **9**, R = H); Yield 1.99 g (66%), m.p. 228-30°C; IR (KBr): 3150-2700 (-NH- and -OH), 1321, 1144 cm^{-1} (-SO_2-); ^1H NMR (DMSO-d_6/TMS): δ 4.10 (m, 2H, -CH_2), 5.01 (m, 1H, -CH-), 6.31 (d, 1H, D_2O exch., –OH), 7.10-7.77 (m, 9H, Ar-H); MS (CI): *m/z* 303 $[M+H]^+$.

9b (*i.e.*, **9**, R = CH_3); Yield 2.26 g (72%), m.p. 221-23°C; IR (KBr): 3200-2935 (-NH- and -OH), 1312, 1150 cm^{-1} (-SO_2-); ^{1}H NMR (DMSO-d_6/TMS): δ 2.20 (s, 3H, Ar-CH_3), 4.82 (d, 2H, -CH_2-S), 6.22 (d, 1H, D_2O exch., –OH), 7.20-7.90 (m, 8H, Ar-H), 12.60 (s, 1H, D_2O exch., –NH); MS (CI): *m/z* 315 $[M+H]^+$.

9c (*i.e.*, **9**, R = OCH_3); Yield 2.49 g (75%), m.p. 158-61°C; IR (KBr): 3430-2933 cm^{-1} (-NH- and -OH), 1322, 1139 cm^{-1} (-SO_2-); ^{1}H NMR (DMSO-d_6/TMS): δ 2.30 (s, 3H, Ar-OCH_3), 4.80 (s, 2H, -CH_2-S), 4.98 (m, 1H, -CH-), 6.28 (d, 1H, D_2O exch., –OH), 7.00-8.00 (m, 8H, Ar-H), 12.50 (s, 1H, D_2O exch., –NH); MS (CI): *m/z* 333 $[M+H]^+$.

9d (*i.e.*, **9**, R = NO_2); Yield 2.42 g (70%), m.p. 175-77°C; IR (KBr): 3207-2859 cm^{-1} (-NH- and -OH), 1312, 1140 (-SO_2-) cm^{-1}; ^{1}H NMR (DMSO-d_6/TMS): δ 4.90 (d, 2H, -CH_2-S), 4.95 (m, 1H, -CH-), 6.28 (d, 1H, D_2O exch., –OH), 7.30-8.40 (m, 8H, Ar-H), 12.92 (s, 1H, D_2O exch., –NH); MS (CI): *m/z* 348 $[M+H]^+$.

9e (*i.e.*, **9**, R = Cl); Yield 2.08 g (62%), m.p. 184-87°C; IR (KBr): 3437-2730 (-NH- and -OH), 1321, 1143 cm^{-1} (-SO_2-); ^{1}H NMR (DMSO-d_6/TMS): δ 4.87 (d, 2H, -CH_2-S), 5.13 (m, 1H, -CH-), 6.29 (d, 1H, D_2O exch., –OH), 6.95-7.69 (m, 7H, Ar-H), 12.87 (s, 1H, D_2O exch., –NH);; MS (CI): *m/z* 337 $[M+H]^+$.

10a (*i.e.*, **10**, R = H); Yield 2.66 g (85%), m.p. 146-48°C; IR (KBr): 1680 (-CO-), 1329 cm^{-1} (-SO_2-); ^{1}H NMR (DMSO-d_6/TMS): δ 3.76 (s, 3H, -N-CH_3), 5.31 (s, 2H, -CH_2-S-), 7.10-7.90 (m, 9H, Ar-H); MS (CI): *m/z* 315 $[M+H]^+$.

13a (*i.e.*, **13**, R = H); Yield 2.78 g (88%), m.p. 178-80°C; IR (KBr): 3437-2924 (-OH), 1324, 1155 cm^{-1} (-SO_2-); ^{1}H NMR (DMSO-d_6/TMS): δ 3.83 (s, 3H, -N-CH_3), 4.15 (m, 2H, -CH_2-S-), 5.09 (m, 1H, -CH-), 6.30 (d, 1H, D_2O exch., –OH), 7.00-7.84 (m, 9H, Ar-H); MS (CI): *m/z* 317 $[M+H]^+$.

General procedure for Reduction with $NaBH_4$: To a suspension/solution of compound (0.01 mol) in methanol (30 mL) was added $NaBH_4$ (0.44 g, 0.012 mol) in 5-6 lots at 0-5°C. After maintaining the temperature for 5-10 min, the temperature was raised to RT and stirred for 30 min. At the end of this period, the reaction-mixture was poured into ice-water. The separated solid was filtered, washed with water, dried and recrystallized from ethanol to obtain the pure reduced compound.

8a (*i.e.*, **8**, R = H); Yield 2.18 g (81%), m.p. 177-80°C; IR (KBr): 3059-2606 cm^{-1} (-NH- and -OH); ^{1}H NMR (DMSO-d_6/TMS): δ 3.55 (d of q, 2H, -CH_2-S-, *J* = 5.2 Hz), 4.90 (m, 1H, -CH-), 6.00 (d, 1H, D_2O exch., –OH, *J* = 4.7 Hz), 7.10-7.64 (m, 9H, Ar-H), 12.50 (s, 1H, D_2O exch., –NH-); MS (CI): *m/z* 271 $[M+H]^+$.

8b (*i.e.*, **8**, R = CH_3); Yield 1.98 g (70%), m.p. 225-27°C; IR (KBr): 3200-2935 (-NH- and -OH) cm^{-1}; ^{1}H NMR (DMSO-d_6/TMS): δ 2.10 (s, 3H, Ar-CH_3), 4.79 (m, 2H, -CH_2-S-), 5.09 (m, 1H, -CH-), 6.34 (d, 1H, D_2O exch., –OH), 7.20-7.90 (m, 8H, Ar-H), 12.60 (s, 1H, D_2O exch., –NH-); MS (CI): *m/z* 285 $[M+H]^+$.

8c (*i.e.*, **8**, R = OCH_3); Yield 2.1 g (70%), m.p. 150-54°C; IR (KBr): 3430-2933 cm^{-1} (-NH- and -OH); ^{1}H NMR (DMSO-d_6/TMS): δ 2.40 (s, 3H, Ar-OCH_3), 4.80 (m, 2H, -CH_2-S-), 5.19 (m, 1H, -CH-), 6.38 (d, 1H, D_2O exch., –OH), 7.00-8.00 (m, 8H, Ar-H), 12.50 (s, 1H, D_2O exch., –NH-); MS (CI): *m/z* 301 $[M+H]^+$.

8d (*i.e.*, **8**, R = NO_2); Yield 2.23 g (71%), m.p. 128-30°C; IR (KBr): 3207-2859 cm^{-1} (-NH- and -OH); ^{1}H NMR (DMSO-d_6/TMS): δ 4.90 (m, 2H, -CH_2-S-), 5.18 (m, 1H, -CH-), 6.33 (d, 1H, D_2O exch., –OH), 7.30-8.40 (m, 8H, Ar-H); MS (CI): *m/z* 316 $[M+H]^+$.

8e (*i.e.*, **8**, R = Cl); Yield 2.18 g (72%), m.p. 160-64°C; IR (KBr): 3437-2720 cm^{-1} (-NH- and -OH); ^{1}H NMR (DMSO-d_6/TMS): δ 4.87 (m, 2H, -CH_2-S-), 5.15 (m, 1H, -CH-), 6.24 (d, 1H, D_2O exch., –OH), 7.25-8.30 (m, 8H, Ar-H), 12.87 (s, 1H, D_2O exch., –NH-); MS (CI): *m/z* 305 $[M+H]^+$.

9a (*i.e.*, **9**, R = H); Yield 2.08 g (69%), m.p. 228-30°C.

9b (*i.e.*, **9**, R = CH_3); Yield 2.35 g (75%), m.p. 221-23°C.

9c (*i.e.*, **9**, R = OCH_3); Yield 2.49 g (75%), m.p. 158-61°C.

9d (*i.e.*, **9**, R = NO_2); Yield 2.29 g (66%), m.p. 175-77°C.

9e (*i.e.*, **9**, R = Cl); Yield 2.41 g (72%), m.p. 184-87°C.

12a (*i.e.*, **12**, R = H); Yield 2.18 g (77%), m.p. 156-58°C; IR (KBr): 3196 (-OH-) cm^{-1}; ^{1}H NMR (DMSO-d_6/TMS): δ 3.54 (s, 3H, -N-CH_3), 3.65 (d of q, 2H, -C(H_AH_B)-S-), 5.20 (q, 1H, -CH-), 6.80 (d, 1H, D_2O exch., –OH), 7.20-7.66 (m, 9H, Ar-H); MS (CI): *m/z* 285 $[M+H]^+$.

13a (*i.e.*, **13**, R = H); Yield 2.74 g (87%), m.p. 179-82°C.

General procedure for alkylation with DMS: A mixture of compound (0.01 mol), K_2CO_3 (1.63 g, 0.012 mol), TBAB (0.2 g) and acetonitrile (30 mL)

was stirred at RT for 30 min. Later, dimethyl sulfate (0.011 mol) was added and the entire mixture stirred at RT for 3-4 hr. At the end of this period, the reaction-mixture was poured in to ice-water. The separated solid was filtered, washed with water, dried and the crude product was purified by column chromatography over basic alumina using hexane:ethyl acetate mixture (100:0 to 95:5) as eluent to afford pure methylated product.

10a (*i.e.*, **10**, R = H); Yield 2.67 g (85%), m.p. 147-50°C.

11a (*i.e.*, **11**, R = H); Yield 2.39 g (85%), m.p. 174-77°C; IR (KBr): 1687 cm^{-1} (-CO-); ^{1}H NMR (DMSO-d_6/TMS): δ 3.78 (s, 3H, -N-CH_3), 5.02 (s, 2H, -S-CH_2), 7.20-8.10 (m, 9H, Ar-H); MS (CI): *m/z* 283 $[M+H]^+$.

12a (*i.e.*, **12**, R = H); Yield 2.18 g (77%), m.p. 156-58°C.

13a (*i.e.*, **13**, R = H); Yield 2.78 g (88%), m.p. 180-83°C.

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