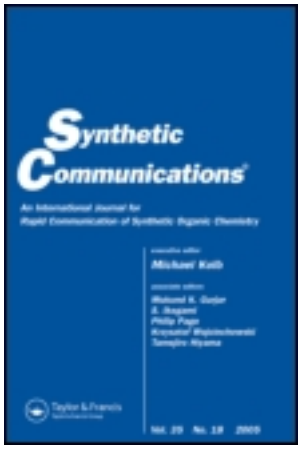

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Synthesis of Novel Thioamidoalkyl- and Thiocarbamidoalkyl Naphthols via a Three-Component Condensation Reaction Using Heterogeneous Catalyst of Ferric Hydrogensulfate

Hossein Eshghi [a], Gholam Hossein Zohuri [a] & Saman Damavandi [a]

[a] Department of Chemistry, Ferdowsi University of Mashhad, Mashhad, Iran

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SYNTHESIS OF NOVEL THIOAMIDOALKYL- AND THIOCARBAMIDOALKYL NAPHTHOLS VIA A THREE-COMPONENT CONDENSATION REACTION USING HETEROGENEOUS CATALYST OF FERRIC HYDROGENSULFATE

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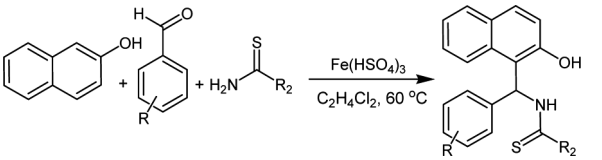
Department of Chemistry, Ferdowsi University of Mashhad, Mashhad, Iran

GRAPHICAL ABSTRACT



Abstract *Ferric hydrogensulfate [$Fe(HSO_4)_3$] was used as a suitable heterogeneous catalyst for the one-pot multicomponent reaction of β-naphthol, aromatic aldehydes, and thioamide derivatives to obtain the corresponding thioamidoalkyl naphthols. Various novel thioamidoalkyl naphthols and thiocarbamidoalkyl naphthols were synthesized in good yields from thioacetamide and thiourea. The heterogeneous nature of the catalyst made it reusable for further chemical reactions.*

Keywords Ferric hydrogensulfate; heterogeneous catalyst; multicomponent reaction; thioacetamide; thioamidoalkyl naphthol

INTRODUCTION

Multicomponent reactions (MCRs) have become an efficient and powerful tool for the construction of complex molecules because products are formed in a one-pot reaction without isolation of intermediates or modification the reaction conditions.[1–3] MCRs are particularly useful for generating diverse chemical libraries of drug-like molecules for biological screening.[4] One-pot reactions provide the possibility of synthesizing the complex directly, without isolating the intermediates, which prompts researchers to design such reactions using different catalysts. Therefore, the reaction times are reduced and both the energy and raw materials are saved.[5,6]

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Address correspondence to Hossein Eshghi, Department of Chemistry, Ferdowsi University of Mashhad, Mashhad 91775-1436, Iran. E-mail: heshghi@ferdowsi.um.ac.ir

Compounds bearing 1,3-amino oxygenated functional groups are biologically important natural products and potential drugs, including a number of nucleoside antibiotics and HIV protease inhibitors, such as ritonavir and lipinavir.[7] Aminonaphthols have been reported to exhibit antihypertensive, adrenoceptor blocking, and Ca^{+2} channel blocking, activities.[8] Furthermore, amidoalkyl naphthols can be converted to useful synthetic building blocks and 1-aminomethyl-2-naphthols, which exhibit depressor and bradycardiac activity.[9,10]

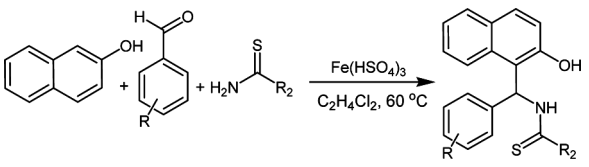
Many studies have been devoted to the preparation of amidoalkyl naphthols by multicomponent condensation of aldehydes, β-naphthol, and amide in the presence of various Lewis or Brønsted acid catalysts.[11–18] However, to the best of our knowledge, only two works with limited examples have reported on the replacement of amide with thioamide or thiourea.[19,20]

In the present study, ferric hydrogenesulfate [$Fe(HSO_4)_3$] was introduced as an efficient catalyst for synthesis of the thioamidoalkyl naphthols and thiocarbamidoalkyl naphthols by one-pot, three-component coupling of β-naphthol, aromatic aldehydes, and thioacetamide or thiourea (Scheme 1).

Synthesis of thioamidoalkyl and thiocarbamidoalkyl naphthols was performed successfully by the three-component condensation of β-naphthol, aryl aldehydes, and thiourea or thioacetamide in the presence of heterogeneous catalyst of ferric hydrogenesulfate.[21] To find the optimum conditions, the one-pot reaction of mentioned components was carried out in different conditions. First of all, one-pot, three-component reaction of β-naphthol, 4-nitrobenzaldehyde, and thiourea in the presence of ferric hydrogenesulfate was carried out as an experimental model with different solvents, while the reaction time and the amount of used catalyst were kept constant.

As it can be seen in Table 1, the greatest yield was obtained using dimethylsulfoxide (DMSO) as a solvent. Because of the good yield and easier workup procedure, 1,2-dichloroethane was used for further studies of multicomponent reactions. It should be noted that the model reaction was carried out with three different amounts of catalyst to find the optimum molar ratio of the catalyst (Table 1). However, addition of more than 10% of the catalyst only slightly increased the yield of the reaction. Thus, further studies were carried out in the presence of 10% molar ratio of ferric hydrogensulfate as a catalyst.

Our previous study showed that the ferric hydrogensulfate is a durable catalyst for six subsequent reactions.[22] However, in the present work, reusability of the catalyst was studied in the reaction of β-naphthol, 4-nitrobenzaldehyde, and thiourea in the presence of the catalyst in 1,2-dichloroethane as a solvent. After completion of each run, the catalyst was recovered simply. The hydrated catalyst was washed



Scheme 1. Ferric hydrogensulfate–catalyzed three-component synthesis of thioamidoalkyl and thiocarbamidoalkyl naphthols.

Table 1. Influence of the solvent, mol% of the catalyst, and its reusability in the three-component reaction of β-naphthol, 4-nitrobenzaldehyde, and thiourea[a]

Entry	Solvent	Catalyst (%)	Yield (%)[b]
1	CH_3CN	$Fe(HSO_4)_3$ (10)	70
2	C_2H_5OH	$Fe(HSO_4)_3$ (10)	72
3	CH_3OH	$Fe(HSO_4)_3$ (10)	75
4	CH_2Cl_2[c]	$Fe(HSO_4)_3$ (10)	76
5	$CHCl_3$[c]	$Fe(HSO_4)_3$ (10)	80
6	DMSO	$Fe(HSO_4)_3$ (10)	95
7	$C_2H_4Cl_2$	$Fe(HSO_4)_3$ (10)	92
8	$C_2H_4Cl_2$	$Fe(HSO_4)_3$ (8)	85
9	$C_2H_4Cl_2$	$Fe(HSO_4)_3$ (5)	80
10[d]	$C_2H_4Cl_2$	$Fe(HSO_4)_3$ (10)	92
11[e]	$C_2H_4Cl_2$	$Fe(HSO_4)_3$ (10)	92
12[f]	$C_2H_4Cl_2$	$Fe(HSO_4)_3$ (10)	88
13[g]	$C_2H_4Cl_2$	$Fe(HSO_4)_3$ (10)	85
14	$C_2H_4Cl_2$	$FeCl_3$ (10)	20
15	$C_2H_4Cl_2$	$NaHSO_4$ (10)	Trace
16	$C_2H_4Cl_2$	$NaHSO_4$ (30)	35
17	$C_2H_4Cl_2$	$FeCl_3$ (10) + $NaHSO_4$ (30)	62

[a]All reactions were carried out at 60 °C for 8 h.
[b]Isolated yields.
[c]Reflux condition.
[d–g]Reusability of the recovered catalyst in new runs.

several times with methanol and dried in an oven at 80 °C for 2 h before reusing. As can be seen in Table 1, the catalyst could be reused without significant loss of its catalytic activity at least four times.

To find out which part of the catalyst is responsible for promoting the reaction, we studied this reaction in the presence of catalytic amounts of $FeCl_3$ and $NaHSO_4$. Comparing the of catalytic efficiency of $Fe(HSO_4)_3$ (10%) with $NaHSO_4$ (10% and 30%) and $FeCl_3$ (10%) or combining them showed that ferric hydrogensulfate was acting even better than a bifunctional catalyst (Table 1, entries 14–17). As can be seen in Table 1, it seems that both parts of the catalyst are involved in catalyzing the reaction. It is noteworthy that by monitoring the reaction with thin-layer chromatography (TLC), we realized that the *ortho*-quinone methide (*o*-QM) intermediate in the presence of $FeCl_3$ appeared faster than in the reactions using $NaHSO_4$; however the yield was lower. It is conceivable that the first stage catalyzes better in the presence of ferric because of chelate formation, but the next stage can be catalyzed by both of them.

Various aromatic aldehydes, thioacetamide, thiourea, or urea were reacted with β-naphthol in 1,2-dichloroethane to obtain corresponding thioamidoalkyl naphthols in good yield using catalytic amount of ferric hydrogensulfate. The results are illustrated in Table 2. Although aromatic aldehydes bearing either electron-donating or electron-withdrawing substituted groups reacted successfully and afforded the products in good yields, reaction times were profoundly varied. As expected, the aromatic aldehydes with electron-withdrawing groups reacted faster than the aromatic aldehyde with electron-releasing groups.

Table 2. Results of thioamidoalkyl and thiocarbamidoalkyl naphthol synthesis

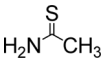
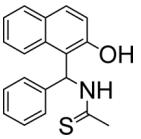
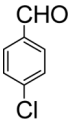
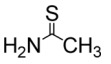
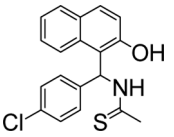
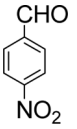
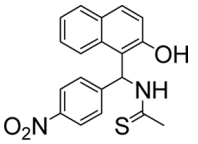
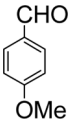
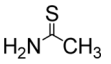
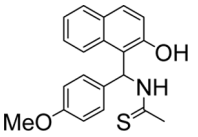
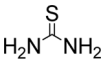
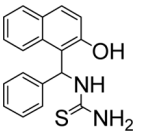
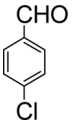
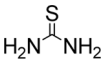
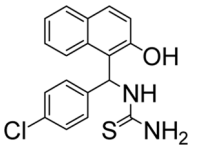
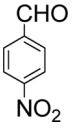
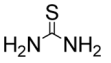
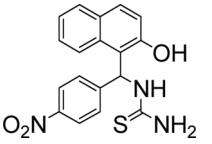
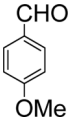
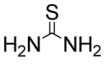
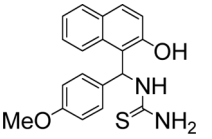
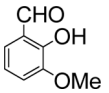
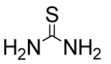
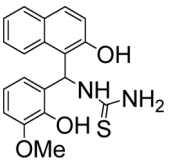
Entry	Aldehyde	Amide	Catalyst (%mol)	Product	Time (h)	Yield (%)[a]	MP(°C) [lit.]
1			10		6.5	88	240–242 [not reported][20]
2			10		6	88	246–248
3			10		5.5	89	243–245
4			10		8	79	192–194
5			12		6	88	179–181 [180–182][19]
6			12		6	90	175–177
7			12		5	91	176–178
8			12		8	83	136–138
9			12		5.30	83	165–167

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Table 2. Continued

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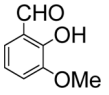
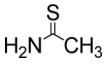
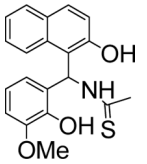
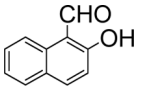
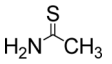
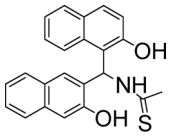
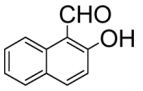
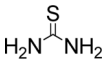
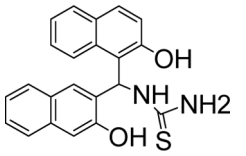
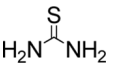
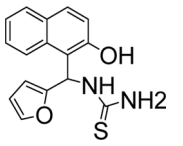
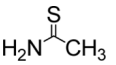
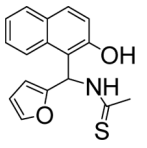
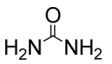
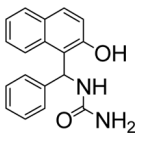
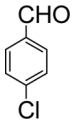
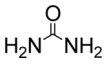
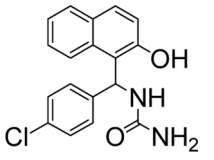
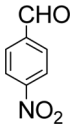
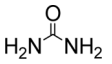
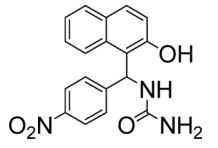
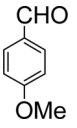
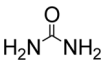
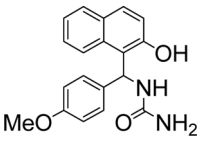
10			10		6	81	202–204
11			10		7	78	218–220
12			10		7	75	170–172
13			10		8	35	185–187
14			10		8	25	225–227
15			10		6.5	88	175–176 [174–175][15]
16			10		5	92	168–169 [168–169][15]
17			10		5	94	179–180 [181–182][14]

(*Continued*)

Table 2. Continued

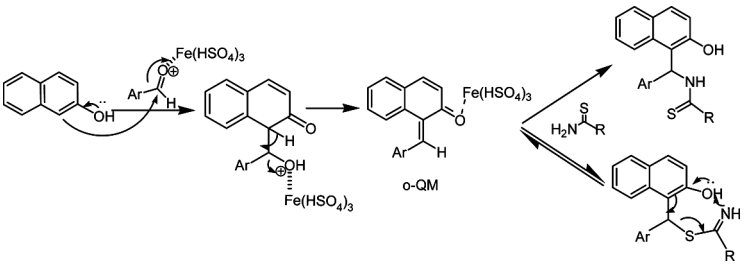
Entry	Aldehyde	Amide	Catalyst (%mol)	Product	Time (h)	Yield (%)[a]	MP(°C) [lit.]
18			10		8	84	186–187 [184–186][11]

[a]Isolated yields.

The adopted mechanism[11] in the synthesis of amidoalkyl naphthol compounds is outlined in Scheme 2 for thio derivatives: in situ generation of *ortho*-quinone methides (*o*-QMs) and addition of thioamide to form thioamidoalkyl naphthol derivatives. In fact, the nucleophilicity of nitrogen atoms of thiourea or thioacetamide is very weak because of the push–pull effect, which increases the nucleophilicity of sulfur atoms. We suggest that sulfur can attack kinetically to *o*-QMs, but reversibility of the reaction (unstable product) in this case favored attack by nitrogen to form a thermodynamically stable product. Beside, the interaction of *o*-QMs with $Fe(HSO_4)_3$ increases its hardness and provides a condition for the attack of the hard nitrogen (weak nucleophile) rather than soft sulfur (strong nucleophile) attack. Because the reactions of β-naphthol, aldehydes, and thiourea failed in the presence of some reported catalysts such as I_2[23] and oxalic acid,[24] we suggest that the second stage is an acid-catalyzed nucleophilic substitution.

The poor reaction with heterocyclic aldehydes can be explained by low catalytic activation of the furfural carbonyl group by acid because conjugation with the heterocyclic heteroatom lowers the carbonyl carbon's electrophilicity. Many studies reported obtaining only trace amounts of the corresponding products in the reaction of heterocyclic aldehydes such as pyridine-4-carboxaldehyde, indole-3-carboxaldehyde, and furfural with 2-naphthol and acetamide.[2,4,11] However, in the presence of ferric hydrogensulfate, the corresponding products in the reaction of furfural, β-naphthol, and thioacetamide or thioamide were obtained in better yields (Table 2, entries 13 and 14).

In conclusion, we have elaborated an efficient, simple, and mild one-pot procedure for the synthesis of thioamidoalkyl and thiocarbamidoalkyl naphthol



Scheme 2. Proposed mechanism for thioamidoalkyl naphthol synthesis.

derivatives. This new approach provided moderate to good yield of products with the simple experimental and workup procedure. Since the $Fe(HSO_4)_3$ catalyst is an inexpensive, easy to produce and handle, and readily removable and reusable, our method seems to be attractive process for the convenient synthesis of substituted thioamidoalkyl and thiocarbamidoalkyl naphthols through a three-component reaction.

EXPERIMENTAL

Chemicals were either prepared in our laboratories or purchased from Merck, Fluka, and Aldrich Chemical Companies. All yields refer to isolated products. The reactions were monitored by thin-layer chromatography (TLC) carried out on silica plates. The products were characterized by comparison of their physical data with those of known samples or by their spectral data. Infrared (IR) spectra were recorded on a Shimadzu IR 470 spectrophotometer. ^{1}H NMR spectra was recorded on a Bruker 100-MHz spectrometer in dimthylsulfoxide (DMSO) as the solvent and tetramethylsilane (TMS) as internal standard. Elemental analyses were obtained on a Thermo Finnigan Flash EA microanalyzer. Silica gel 60 (230–400 mesh) was purchased from Merck. Ferric hydrogensulfate was prepared according to previously reported procedure.[21,22]

General Procedure to Synthesize Thioamidoalkyl and Thiocarbamidoalkyl Naphthols

Aldehyde (1 mmol), thioacetamide or thiourea (1.2 mmol), and ferric hydrogensulfate [$Fe(HSO_4)_3$] (10 mol%) were added successively to a mixture of β-naphthol (1 mmol) in 1,2-dichloroethane (10 mL). The mixture was stirred at 60 °C, and the reaction was followed by TLC. After completion of the reaction, the catalyst was simply recovered through filtration. The hydrated catalyst was washed several times with methanol and dried in oven at 80 °C for 2 h before reuse. The solvent was evaporated, and the solid residue was washed with chloroform and water several times respectively to obtain the almost pure thioamidoalkyl and thiocarbamidoalkyl naphthols. For preparation with high purity for elemental analysis, the final product was purified by short column chromatography (eluent; hexane–ethyl acetate 4:1).

Spectral Data of the Prepared Compounds

Entry 1. ^{1}H NMR (100 MHz, DMSO-d_6): 9.95 (br, 1H), 8.50–8.45 (ss, 1H), 7.85–7.75 (m, 4H), 7.35–7.15 (m, 8H), 1.75 (s, 3H); IR (KBr, cm^{-1}): 3396, 3137, 2681, 1590, 1461, 1388 (C=S), 1072 (C=S), 744. Anal. calcd. for $C_{19}H_{17}NOS$: C, 74.23; H, 5.57; N, 4.56; S, 10.43. Found: C, 74.32; H, 5.59; N, 4.59; S, 10.45.

Entry 2. ^{1}H NMR (100 MHz, DMSO-d_6): 10.35 (br, 1H), 8.55–8.50 (ss, 1H), 7.95–7.85 (m, 3H), 7.65–7.55 (m, 3H), 7.35–7.20 (m, 7H), 6.50 (s, 1H); IR (KBr, cm^{-1}): 3426, 2064, 1519, 1377 (C=S), 1086 (C=S), 805. Anal. calcd. for $C_{19}H_{16}ClNOS$: C, 66.75; H, 4.72; N, 4.10; S, 9.38. Found: C, 66.83; H, 4.79; N, 4.17; S, 9.30.

Entry 3. ^{1}H NMR (100 MHz, DMSO-d_6): 10.08 (br, 1H), 8.30 (s, 1H), 8.05–7.90 (m, 4H), 7.70–7.60 (m, 4H), 7.40–7.25 (m, 5H), 6.75 (s, 1H); IR (KBr, cm^{-1}): 3369, 3262, 3162, 1598, 1468, 1421 (C=S), 1087 (C=S), 740. Anal. calcd. for $C_{19}H_{16}N_2O_3S$: C, 64.76; H, 4.58; N, 7.95; O, 13.62; S, 9.10. Found: C, 64.72; H, 4.69; N, 8.05; S, 9.17.

Entry 4. ^{1}H NMR (100 MHz, DMSO-d_6): 10.12 (br, 1H), 8.15 (s, 1H), 7.95–7.80 (m, 3H), 7.75–7.60 (m, 4H), 7.40–7.25 (m, 6H), 6.55 (s, 1H), 3.55 (s, 3H); IR (KBr, cm^{-1}): 3292, 3085, 2647, 1480 (C=S), 1381(C=S), 970. Anal. calcd. for $C_{20}H_{19}NO_2S$: C, 71.19; H, 5.68; N, 4.15; O, 9.48; S, 9.50. Found: C, 71.25; H, 5.74; N, 4.08; S, 9.54.

Entry 5. ^{1}H NMR (100 Hz, DMSO-d_6): δ 10.25 (br, 1H), 8.25–8.15 (m, 2H), 7.95–7.80 (m, 3H), 7.40–7.25 (m, 10H); IR (KBr, cm^{-1}): 3442, 3310, 1614, 1514, 1435 (C=S), 1262 (C=S), 1173, 811. Anal. calcd. for $C_{18}H_{16}N_2OS$: C, 70.10; H, 5.23; N, 9.08; S, 10.40. Found: C, 69.98; H, 5.16; N, 9.03; S, 10.54.

Entry 6. ^{1}H NMR (100 Hz, DMSO-d_6): δ 10.12 (br, 1H), 8.35–8.30 (ss, 1H), 8.10 (s, 1H), 7.85–7.74 (m, 4H), 7.50–7.10 (m, 8H); IR (KBr, cm^{-1}): 3471, 3285, 1605, 1515, 1436 (C=S), 1260 (C=S), 1144, 807; Anal. calcd. for $C_{18}H_{15}ClN_2OS$: C, 63.06; H, 4.41; N, 8.17, S; 9.35. Found: C, 63.18; H, 4.49; N, 8.28, S; 9.31.

Entry 7. ^{1}H NMR (100 MHz, DMSO-d_6): 10.10 (br, 1H), 8.35–8.30 (m, 3H), 7.90–7.75 (m, 2H), 7.50–7.25 (m, 9H), 7.05-6.95 (m, 1H), 2.30 (s, 3H); IR (KBr, cm^{-1}): 3390, 3149, 1602, 1557, 1386 (C=S), 1081 (C=S), 883. Anal. calcd. for $C_{18}H_{15}N_3O_3S$: C, 61.18; H, 4.28; N, 11.89; S, 9.07. Found: C, 61.22; H, 4.34; N, 11.95; S, 9.11.

Entry 8. ^{1}H NMR (100 MHz, DMSO-d_6): 11.10 (br, 1H), 8.45–8.40 (ss, 1H), 8.10–7.95 (m, 2H), 7.70–7.60 (m, 6H), 7.30–7.25 (m, 1H), 7.05-6.95 (m, 4H), 3.70 (s, 3H); IR (KBr, cm^{-1}): 3455, 3262, 2054, 1642, 1379 (C=S), 1141 (C=S), 814. Anal. calcd. for $C_{19}H_{18}N_2O_2S$: C, 67.43; H, 5.36; N, 8.28; S, 9.47. Found: C, 67.40; H, 5.41; N, 8.20; S, 9.52.

Entry 9. ^{1}H NMR (100 MHz, DMSO-d_6): 9.95 (br, 1H), 8.45–8.30 (m, 2H), 7.70–7.60 (m, 3H), 7.30–7.10 (m, 3H), 6.75-6.60 (m, 6H), 3.65 (s, 3H); IR (KBr, cm^{-1}): 3351, 3231, 1577, 1491, 1413 (C=S), 1037 (C=S), 865. Anal. calcd. for $C_{19}H_{18}N_2O_3S$: C, 64.39; H, 5.12; N, 7.90; S, 9.05. Found: C, 64.47; H, 5.19; N, 7.97; S, 8.95.

Entry 10. ^{1}H NMR (500 MHz, DMSO-d_6): 10.28 (b, 3H), 7.91 (d, 0.13H, non-bonded free-NH), 7.90 (dd, 1H, $J_1 = 7.5$ Hz, $J_2 = 1.5$ Hz), 7.76 (dt, 1H, $J_1 = 7.5$ Hz, $J_2 = 1.5$ Hz), 7.53 (dd, 1H, $J_1 = 7.5$ Hz, $J_2 = 1.5$ Hz), 7.40 (t, 1H, $J = 8$ Hz), 7.30 (m, 2H), 6.8–7.2 (m, 4H), 3.63 (s, 3H), 2.25 (s, 3H). IR (KBr, cm^{-1}): 3369, 3262, 3162, 1598, 1468, 1421 (C=S), 1087 (C=S), 740. Anal. calcd. for $C_{20}H_{19}NO_3S$: C, 67.97; H, 5.42; N, 3.96; S, 9.07. Found: C, 67.92; H, 5.39; N, 4.01; S, 9.01.

Entry 11. ^{1}H NMR (500 MHz, DMSO-d_6): ^{1}H NMR (500 MHz, DMSO-d_6): 10.88 (b, 2H, OH), 7.91 (d, 1H, $J = 7.5$ Hz), 7.78 (m, 1H), 7.61 (m, 1H), 7.53 (d, 1H, $J = 7.5$ Hz), 7.44 (t, 1H, $J = 9$ Hz), 7.37 (t, 1H, $J = 8.5$ Hz), 7.27 (m, 2H), 7.18 (m, 2H), 7.08 (m, 4H), 2.26 (s, 3H). IR (KBr, Cm^{-1}): 3359, 1629, 1525, 1474, 1420

(C=S), 1257, 1067 (C=S), 814, 742. Anal calcd. for $C_{23}H_{19}NO_2S$: C, 73.97; H, 5.13; N, 3.75; S, 8.59. Found: C, 73.89; H, 5.02; N, 3.81; S, 8.51.

Entry 12. ^{1}H NMR (100 MHz, DMSO-d_6): δ 11.30 (bs, 2H), 8.50 (bs, 1H), 6.80–8.25 (m, 12H), 6.50 (s, 1H), 3.75 (s, 3H); IR (KBr, cm^{-1}): 3347, 3180, 1586, 1444, 1409 (C=S), 1033 (C=S), 917. Anal. calcd. for $C_{22}H_{18}N_2O_2S$: C, 70.57; H, 4.85; N, 7.48; S, 8.56. Found: C, 70.62; H, 4.89; N, 7.53; S, 8.61.

Entry 13. ^{1}H NMR (100 MHz, DMSO-d_6): δ: 10.20 (br, 1H), 7.92 (bs, 1H), 7.75–7.05 (m, 10H), 6.95–6.90 (m, 2H); IR (KBr, cm^{-1}): 3362, 3099, 1580, 1473, 1288 (C=S), 1011 (C=S), 822. Anal. calcd. for $C_{16}H_{14}N_2O_2S$: C, 64.41; H, 4.73; N, 9.39; S, 10.75. Found: C, 64.52; H, 4.79; N, 9.43; S, 10.89.

Entry 14. ^{1}H NMR (100 MHz, DMSO-d_6): 10.28 (br, 1H), 8.05 (s, 1H), 7.80–7.70 (m, 3H), 7.45–7.30 (m, 3H), 7.05-6.95 (m, 4H), 2.30 (s, 3H); IR (KBr, cm^{-1}): 3420, 3095, 1580, 1431, 1265 (C=S), 1233, 1119 (C=S), 856. Anal. calcd. for $C_{17}H_{15}NO_2S$: C, 68.66; H, 5.08; N, 4.71; S, 10.78. Found: C, 68.62; H, 5.14; N, 4.65; S, 10.85.

AKNOWLEDGMENT

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