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A NOVEL METHOD FOR *N*3-FUNCIONALIZED AMINOIMIDAZOLINES AND AMINOPYRIMIDINES BY AZA-MICHAEL ADDITIONS OF AMINOIMIDAZOLINES AND AMINOPYRIMIDINES TO *α, β*-UNSATURATED CARBONYL COMPOUNDS

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Abstract –An efficient and convenient procedure is described for the preparation of *N3*-functionalized aminoimidazolines and aminopyrimidines through the aza-Michael additions of aminoimidazolines and aminopyrimidines with *α, β*-unsaturated carbonyl compounds catalyzed by KF/Al_2O_3. This method offers a novel and efficient protocol for the *N3*-functionalized aminoimidazolines and aminopyrimidines with good regioselectivity and high purity under mild conditions.

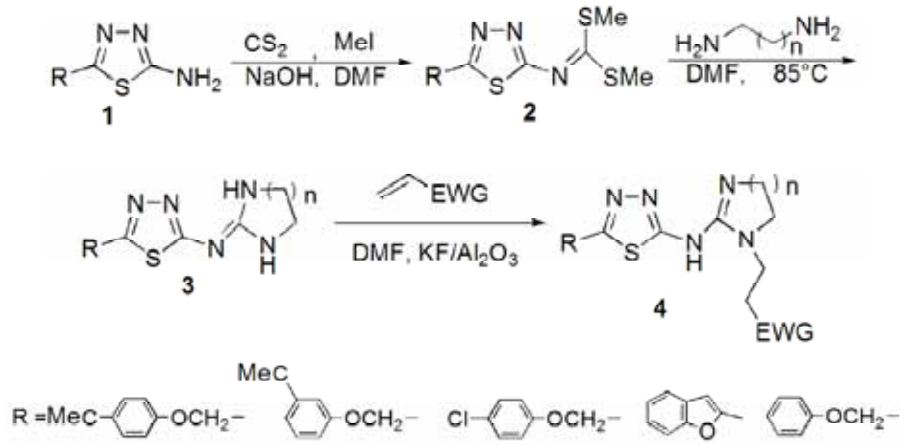
INTRODUCTION

Many compounds containing an amidine moiety (the –NH–C(R)=N– group) such as 2-amino-2-imidazoline, 2-amino-2-oxazoline and 2-amino-2-thiazoline, are known to possess interesting biological properties.[1] These compounds can be used as antivirial, antibacterial, antifungal, antihypertensive drugs and also can be used as pesticides.[2] In particularly, 2-arylamino-2-imidazolines have an interesting chemistry[3–5] and they are effective pharmacophores in medicinal chemistry. 2,6-Dichlorophenylamino-2-imidazoline (clonidine) have a pronounced hypotensive action, which is coupled with a sedative action. A hypotensive action has also been described for benzoyl derivatives of 2-arylamino-2-imidazoline, especially for the compound 1-benzoyl-2-(2′,6′-dichlorophenylamino)-2-imidazoline, in the depressant action on the central nervous system and thus the sedative action being substantially less pronounced with this compound.[6]

To our surprise, there have few reports for the synthesis of *N3*-functionalized aminoimidazolines and aminopyrimidines. Very recently, Servi *et al*. have reported *N3*-substituted aminoimidazolines by alkylation of aminoimidazolines with benzyl chloride in the presence of NaOH as base.[7] Although many important processes in organic chemistry and biochemistry involve C–N bond formation as the fundamental reaction step, some of these processes can suffer from drawbacks such as long reaction times, low yields, high pressures, high temperatures and harsh conditions, which limit its use in the synthesis of complex molecules.[8,9] A simple, efficient and atom economy approach for the synthesis of *N3*-functionalized aminoimidazoline and aminopyrimidine derivaives is our interesting.

We have previously reported the synthesis of *N3*-substituted dihydropyrimidinones, using aza-Michael addition reaction catalyzed by KF/Al_2O_3.[10] In this letter, we are interested in studying the potential and efficiency of KF/Al_2O_3 as catalyst in the addition of substituted aminoimidazolines and aminopyrimidines to *α, β*-unsaturated carbonyl compounds for the syntehsis of *N3*-functionalized aminoimidazolines and aminopyrimidines.

Scheme 1



RESULTS AND DISCUSSION

As described in Scheme 1, dimethyl N-aryldithiocarbonimidates **2** were synthesized by reaction of 2-amine-1,3,4-thiadiazole with carbon disulfide and methyl iodide. Substituted aminoimidazolines (**3a-3d)** and aminopyrimidines (**3e-3f)** were obtained by heating **2** with 1,2-diaminoethane and 1,3-diaminopropane respectively under 85 °C for 5 h.

Preliminary experiments to examine the solvent dependence of aza-Michael reaction were performed using substrate **3a** as a model system to synthesize the compound **4a** (Table 1). Firstly, various temperatures and reaction times were tested using *N*,*N*-dimethylformamide (DMF) as a solvent (Table 1, entries 1–4). All comparative reactions were conducted under optimized conditions and the compound **4a** was obtained in the presence of KF/Al_2O_3. The best yield of **4a** (82%) was obtained by carrying out the reaction in DMF at 65 °C for 6 h. Secondly, other solvents were tested including DMSO, MeCN, CH_2Cl_2 and THF (Table 1, entries 5–8). In general, the use of DMSO, DMF and MeCN resulted in faster reaction with higher yield, in contrast to reactions in less polar THF. Thirdly, we tested the reaction in the presence of various inorganic bases. As shown in Table 1 (entries 9–13), the base catalyst has a significant effect on the yield of the reaction and no desired or trace aza-Michael adduct was obtained in the presence of KOH, NaOH, $NaHCO_3$. When K_2CO_3 was used as catalyst, aza-Michael product was obtained with 68% need a longer reaction time. However, a 82% yield was obtained in the presence of KF/Al_2O_3 (10 mol %) as catalyst.

Table 1 Aza-Michael addition of 3a to methyl acrylate under various conditions.

Entry	Temp. (°C)	Base	Solvent	Time (h)	Yield[a,b](%)
1	20	KF/Al_2O_3	DMF	6	<10
2	65	KF/Al_2O_3	DMF	6	82
3	80	KF/Al_2O_3	DMF	6	76
4	65	KF/Al_2O_3	DMF	12	79
5	65	KF/Al_2O_3	DMSO	6	75
6	65	KF/Al_2O_3	MeCN	6	57
7	65	KF/Al_2O_3	CH_2Cl_2	6	51
8	65	KF/Al_2O_3	THF	6	56
9	65	K_2CO_3	DMF	12	68
10	65	Na_2CO_3	DMF	6	59
11	65	$NaHCO_3$	DMF	6	<10
12	65	NaOH	DMF	6	0
13	65	KOH	DMF	6	0

[a] The reaction was conducted with **3a** (1.0 mmol) and methyl acrylate (1.0 mmol). All the products were investigated thoroughly by ^1H NMR, ^{13}C NMR, IR and element analysis.

[b] Isolated yields.

With optimal conditions in hand, we next examined the generality of these conditions to other substrates using several aminoimidazolines and aminopyrimidines **3a-f** and *α, β*-unsaturated carbonyl compounds. The results are summarized in Table 3 (entries 1–18). Generally, the Michael addition between **3a-f** and *α,*

β-unsaturated carbonyl compounds proceeded efficiently, furnishing excellent isolated yields of desired products. The reaction of other aminoimidazolines and aminopyrimidines containing the 2-benzofuran moiety with *α, β*-unsaturated carbonyl compounds also obtained the desired products in good yields (Table 3, entries 19–24).

Finally we investigated the reusability and the recycling of the KF/Al_2O_3 and found that the catalyst could be easily recovered after completion of the reaction and reused in subsequent reactions. For the reaction of **3a** with methyl acrylate, over 65% yields for **4a** were obtained during the three recycling experiments (82%, 77% and 65%).

Table 2 The synthesis of compounds 2 and 3.

Entry	n	R	Products	Mp (°C)	Yield (%)
1		$C_6H_5OCH_2$	**2a**	80-82	83
2		4-$MeC_6H_4OCH_2$	**2b**	81-83	78
3		3-$MeC_6H_4OCH_2$	**2c**	88-89	82
4		4-$ClC_6H_4OCH_2$	**2d**	109-111	65
5		2-benzofuran	**2e**	164-166	79
6	**1**	$C_6H_5OCH_2$	**3a**	223-224	80
7	**1**	4-$MeC_6H_4OCH_2$	**3b**	221-223	71
8	**1**	3-$MeC_6H_4OCH_2$	**3c**	180-182	81
9	**1**	4-$ClC_6H_4OCH_2$	**3d**	219-221	59
10	**2**	$C_6H_5OCH_2$	**3e**	183-184	74
11	**2**	4-$MeC_6H_4OCH_2$	**3f**	186-188	76
12	**1**	2-benzofuran	**3g**	292-293	66
13	**2**	2-benzofuran	**3h**	294-296	67

The structures of compounds **3** and **4** were shown in Scheme 1. In the ^{1}H-NMR (400 MHz) spectra of **3**, the signal belonging to two CH_2 integrating for four hydrogens was observed between *δ* 3-4 ppm, only one peak belonging to two NH was observed between *δ* 7.5-8.5 ppm. In the ^{13}C-NMR (400 MHz) spectra only one peak was observed at 36.85 belonging to two CH_2. These compounds are effectively symmetrical because of imine–enamine dynamic tautomerization, which makes the CH_2 protons equivalent.

In the ^{1}H NMR spectra of compounds **4**, the signal belonging to endocyclic imida-zoline NH (between *δ* 7.5-8.5 ppm) disappeared and the signal belonging to exocyclic NH was move to *δ* 8.5-9.5 ppm. The two triples belonging to two CH_2 of compounds **4** were observed. In the ^{13}C-NMR spectra, two peaks were

found around 40 ppm. The ^{1}H NMR and ^{13}C-NMR of all products showed that the aza-Michael addition occurred exclusively at endocyclic position of substituted aminoimidazolines and that exocyclic substitution does not take place over the course of the reaction. The complete selectivity, we believe, is due to the difference in the electron density at endocyclic and exocyclic position. The higher basicity of the former resulted in exclusive alkylation at this position, in accordance with the literature precedence.[9] From the above results, we can conclude that the reaction took place via nucleophilic attack of *N* on imidazoline ring (endocyclic nitrogen atom) to *β* carbon atom of *α, β*-unsaturated compounds by useing KF/Al_2O_3 as catalyst.

Table 3 Aza-Michael addition of substituted aminoimidazolines and aminopyrimidines to *α, β*-unsaturated carbonyl compounds catalyzed by KF/Al_2O_3.

Entry[a]	n	R	EWG	Products	Yield (%)[b]
1	1	C_6H_5	CO_2Me	**4a**	82
2	1	4-MeC_6H_4	CO_2Me	**4b**	82
3	1	3-MeC_6H_4	CO_2Me	**4c**	78
4	1	4-ClC_6H_4	CO_2Me	**4d**	76
5	1	C_6H_5	CO_2Et	**4e**	79
6	1	4-MeC_6H_4	CO_2Et	**4f**	84
7	1	3-MeC_6H_4	CO_2Et	**4g**	81
8	1	4-ClC_6H_4	CO_2Et	**4h**	83
9	1	C_6H_5	CN	**4i**	75
10	1	4-MeC_6H_4	CN	**4j**	69
11	1	3-MeC_6H_4	CN	**4k**	67
12	1	4-ClC_6H_4	CN	**4l**	74
13	2	C_6H_5	CO_2Me	**4m**	77
14	2	4-MeC_6H_4	CO_2Me	**4n**	73
15	2	C_6H_5	CO_2Et	**4o**	78
16	2	4-MeC_6H_4	CO_2Et	**4p**	74
17	2	C_6H_5	CN	**4q**	60
18	2	4-MeC_6H_4	CN	**4r**	65
19	1	2-benzofuran	CO_2Me	**4s**	77
20	2	2-benzofuran	CO_2Me	**4t**	83
21	1	2-benzofuran	CO_2Et	**4u**	76
22	2	2-benzofuran	CO_2Et	**4v**	80
23	1	2-benzofuran	CN	**4w**	74
24	2	2-benzofuran	CN	**4x**	78

[a] The reaction was conducted with **3** (1.0 mmol), *α, β*-unsaturated carbonyl compounds (1.0 mmol) and DMF (5 mL) in the presence of KF/Al_2O_3 (10 mol%, 0.1 g) at 65 ^{o}C for 6 h.

[b] Isolated yields.

In conclusion, we have documented the first aza-Michael addition of aminoimidazolines and aminopyrimidines with *α, β*-unsaturated carbonyl compounds affording *N3*-functionalized aminoimidazolines and aminopyrimidines in the presence of KF/Al_2O_3 as catalyst. The KF/Al_2O_3 catalyst was readily separated from the reaction mixture by simple filtration and could be reused without appreciable losses of its high activity and regioselectivity.

EXPERIMENTAL

All reagents were obtained commercially and used without further purification. Melting points were determined on an XT-4 electrothermal micromelting point apparatus and uncorrected. IR spectra were recorded using KBr pellets on Nicolet AVATAR 36 FT-IR spectrophotometer. NMR spectra were recorded at 400 (^{1}H) and 100 (^{13}C) MHz, respectively, on a Varian Mercury plus-400 instrument using $CDCl_3$ or DMSO-d_6 as solvent and TMS as internal standard. Elemental analyses were performed on a Carlo-Erba 1106 Elemental Analysis instrument.

Compounds 2-amino-5-aryloxymethylene-1,3,4-thiadiazole[11] and 2-amino-5-(2-benzofuryl)- 1,3,4-thiadiazole[12] were prepared as described in the literature procedures.

General method for the synthesis of compounds **2a-e**. To a solution of **1** (20 mmol) in DMF (10 mL), aqueous NaOH (20 mol/L, 0.7 mL, 14 mmol) and CS_2 (0.44 mL, 7 mmol) were added with stirring at rt. After 10 min, aqueous NaOH (20 mol/L, 0.7 mL, 14 mmol) and CS_2 (0.44 mL, 7 mmol) were added again. This operation was finally repeated 10 min later and the reaction was placed in an ice bath. After 30 min, the MeI (1.37 mL, 22 mmol) was added drop wise and the stirring was continued for 4 h. The mixture was then poured onto ice-cold water and the precipitate thus obtained was filtered, washed with water, dried and recrystallized from EtOH and DMF and the products were obtained.

2a: Yellow solid. Mp 80-82 °C. IR (KBr): 3061, 2985, 2874, 1594-1394, 753. ^{1}H NMR (400 MHz, DMSO-d_6) δ (ppm): 2.62 (s, 6H, SCH_3), 5.49 (s, 2H, CH_2), 6.99-7.07 (m, 3H, ArH), 7.31-7.35 (m, 2H, ArH). Anal. Calcd for $C_{12}H_{13}N_3OS_3$: C: 46.28, H: 4.21, N: 13.49. Found: C: 46.47, H: 4.10, N: 13.33.

2b: Yellow solid. Mp 81-83 °C. IR (KBr): 3055, 2982, 2864, 1605-1394. ^{1}H NMR (400 MHz, DMSO-d_6) δ (ppm): 2.34 (s, 3H, CH_3), 2.58 (s, 6H, SCH_3), 5.43 (s, 2H, CH_2), 7.02 (d, J = 8.0 Hz, 2H, ArH), 7.18 (d, J

= 8.0 Hz, 2H, ArH). Anal. Calcd for $C_{13}H_{15}N_3OS_3$: C, 47.97; H, 4.65; N, 12.91. Found: C, 47.81; H, 4.67; N, 12.95.

2c: Yellow solid. Mp 88-89 °C. IR (KBr): 3051, 2980, 2920, 2864, 1582-1394. ^{1}H NMR (400 MHz, DMSO-d_6) δ (ppm): 2.21 (s, 3H, CH_3), 2.60 (s, 6H, SCH_3), 5.36 (s, 2H, CH_2), 6.81-6.98 (m, 3H, ArH), 7.08-7.13 (m, 1H, ArH). Anal. Calcd for $C_{13}H_{15}N_3OS_3$: C, 47.97; H, 4.65; N, 12.91. Found, C, 47.78; H, 4.66; N, 12.97.

2d: Yellow solid. Mp 109-111 °C. IR (KBr): 3072, 2997, 2885, 1623. ^{1}H NMR (400 MHz, DMSO-d_6) δ (ppm): 2.63 (s, 6H, SCH_3), 5.52 (s, 2H, CH_2), 7.23 (d, J = 8.4 Hz, 2H, ArH), 7.43 (d, J = 8.4 Hz, 2H, ArH). Anal. Calcd for $C_{12}H_{12}ClN_3OS_3$: C: 41.67, H: 3.50, N: 12.15. Found: C: 41.86, H: 3.51, N: 12.09.

2e: Yellow solid. Mp 164-166 °C. IR (KBr): 3003, 2920, 1587-1433. ^{1}H NMR (400 MHz, DMSO-d_6) δ (ppm): 2.58 (s, 6H, SCH_3), 7.31-7.41 (m, 3H, ArH), 7.66-7.72 (m, 2H, ArH). Anal. Calcd for $C_{13}H_{11}N_3OS_3$: C, 48.57; H, 3.45; N, 13.07. Found: C, 48.36; H, 3.44; N, 13.12.

General method for the preparation of compounds **3a-h**. A solution of **2** (0.04 mol) in DMF (10 mL) was added to a solution of 1,2-diaminoethane or 1,3-diaminopropane (0.05 mol) in DMF(10 mL) with stirring at rt. The reaction mixture was maintained at 85 °C for 5 h. Then the mixture was cooled and added to ice-cold water. The resulting solid was washed with water, dried and recrystallized from appropriate solvent.

3a: White crystals. Mp 223-224 °C. IR (KBr): 3256, 3147, 3091, 3040, 2953-2827, 1632. ^{1}H NMR (400 MHz, DMSO-d_6) δ (ppm): 3.58 (s, 4H, CH_2), 5.30 (s, 2H, CH_2), 6.96-7.04 (m, 3H, ArH), 7.29-7.33 (m, 2H, ArH), 8.02 (s, 2H, NH). ^{13}C NMR (100 MHz, DMSO-d_6): δ (ppm): 36.85, 64.69, 115.07, 121.55, 129.71, 156.37, 157.58, 160.51, 176.58. Anal. Calcd for $C_{12}H_{13}N_5OS$: C, 52.35; H, 4.76; N, 25.44. Found: C, 52.13; H, 4.75; N, 25.53.

3b: Yellow crystals. Mp 221-223 °C. IR (KBr): 3284, 3160, 3071, 3045, 2973-2857, 1629. ^{1}H NMR (400 MHz, DMSO-d_6) δ (ppm): 2.27 (s, 3H, CH_3), 3.55 (s, 4H, CH_2), 5.29 (s, 2H, CH_2), 6.95 (d, J = 8.4 Hz, 2H, ArH), 7.11 (d, J = 8.4 Hz, 2H, ArH), 7.94 (s, 2H, NH). ^{13}C NMR (100 MHz, DMSO-d_6): δ (ppm): 21.15, 38.88, 64.76, 115.04, 129.68, 130.12, 156.34, 157.86, 160.25, 176.45. Anal. Calcd for $C_{13}H_{15}N_5OS$: C, 53.96; H, 5.23; N, 24.20. Found: C, 53.76; H, 5.24; N, 24.31.

3c: Yellow crystals. Mp 180-182 °C. IR (KBr): 3264, 3156, 3087, 2952-2898, 1622. ^{1}H NMR (400 MHz,

DMSO-d_6) δ (ppm): 2.19 (s, 3H, CH_3), 3.55 (s, 4H, CH_2), 5.32 (s, 2H, CH_2), 6.83-6.87 (m, 3H, ArH), 6.98-7.03 (m, 1H, ArH), 7.88 (s, 2H, NH). ^{13}C NMR (100 MHz, DMSO-d_6): δ (ppm): 20.59, 36.58, 64.69, 115.48, 118.85, 126.55, 129.71, 145.36, 157.58, 160.51, 162.32, 176.58. Anal. Calcd for $C_{13}H_{15}N_5OS$: C, 53.96; H, 5.23; N, 24.20. Found: C, 53.73; H, 5.24; N, 24.30.

3d: White crystals. Mp 219-221 °C. IR (KBr): 3289, 3157, 3041, 2983-2885, 1627. ^{1}H NMR (400 MHz, DMSO-d_6) δ (ppm): 3.57 (s, 4H, CH_2), 5.36 (s, 2H, CH_2), 7.24-7.38 (m, 4H, ArH), 7.83 (s, 2H, NH). ^{13}C NMR (100 MHz, DMSO-d_6): δ (ppm): 37.88, 66.25, 115.68, 127.93, 130.87, 155.97, 158.76, 160.65, 178.27. Anal. Calcd for $C_{12}H_{12}ClN_5OS$: C: 46.53, H: 3.90, N: 22.61. Found: C: 46.71, H: 3.88, N: 22.50.

3e: White crystals. Mp 183-184 °C. IR (KBr): 3216, 3125, 3091, 3053, 2961-2871, 1623. ^{1}H NMR (400 MHz, DMSO-d_6) δ (ppm): 1.78-1.82 (m, 2H, CH_2), 3.30 (t, J = 6.4 Hz, 4H, CH_2), 5.26 (s, 2H, CH_2), 6.96-7.04 (m, 3H, ArH), 7.29-7.32 (m, 2H, ArH), 8.16 (s, 2H, NH). ^{13}C NMR (100 MHz, DMSO-d_6): δ (ppm): 20.05, 38.49, 64.73, 115.08, 121.51, 129.70, 153.62, 154.22, 157.60, 176.71. Anal. Calcd for $C_{13}H_{15}N_5OS$: C, 53.96; H, 5.23; N, 24.20. Found: C, 53.77; H, 5.25; N, 24.29.

3f: Yellow crystals. Mp 186-188 °C. IR (KBr): 3213, 3124, 3057, 2966-2867, 1631. ^{1}H NMR (400 MHz, DMSO-d_6) δ (ppm): 1.78-1.81 (m, 2H, CH_2), 2.27 (s, 3H, CH_3), 3.29 (t, J = 5.6 Hz, 4H, CH_2), 5.22 (s, 2H, CH_2), 6.91 (d, J = 8.0 Hz, 2H, ArH), 7.09 (d, J = 8.0 Hz, 2H, ArH), 8.13 (s, 2H, NH). ^{13}C NMR (100 MHz, DMSO-d_6): δ (ppm): 20.06, 20.18, 38.49, 64.88, 114.99, 130.00, 130.28, 153.60, 154.43, 155.48, 176.66. Anal. Calcd for $C_{14}H_{17}N_5OS$: C, 55.42; H, 5.65; N, 23.08. Found: C, 55.19; H, 5.67; N, 23.17.

3g: Yellow crystals. Mp 292-293 °C. IR (KBr): 3230, 3137, 3061, 2953, 2827, 1632. ^{1}H NMR (400 MHz, DMSO-d_6) δ (ppm): 3.41 (s, 4H, CH_2), 7.31-7.44 (m, 3H, ArH), 7.64-7.73 (m, 2H, ArH), 7.62 (s, 2H, NH). ^{13}C NMR (100 MHz, DMSO-d_6): δ (ppm): 41.28, 104.92, 111.63, 122.04, 123.80, 125.72, 128.69, 148.43, 149.30, 152.79, 155.10, 175.89. Anal. Calcd for $C_{13}H_{11}N_5OS$: C, 54.72; H, 3.89; N, 24.55. Found: C, 54.50; H, 3.90; N, 24.64.

3h: Yellow crystals. Mp 294-296 °C. IR (KBr): 3269, 3156, 3078, 2952, 2898, 1622. ^{1}H NMR (400 MHz, DMSO-d_6) δ (ppm): 1.81-1.86 (m, 2H, CH_2), 3.34 (t, J = 5.6 Hz, 4H, CH_2), 7.30-7.42 (m, 3H, ArH), 7.65-7.73 (m, 2H, ArH), 8.24 (s, 2H, NH). ^{13}C NMR (100 MHz, DMSO-d_6): δ (ppm): 20.15, 39.32, 104.84, 111.55, 121.89, 123.64, 125.68, 128.85, 148.54, 149.31, 152.49, 154.92, 176.06. Anal. Calcd for $C_{14}H_{13}N_5OS$: C, 56.17; H, 4.38; N, 23.40. Found: C, 55.94; H, 4.37; N, 23.50.

General procedure for the preparation of compounds **4a-x**. A suspension of compound **3** (1.0 mmol) and KF/Al_2O_3 (10 mol%, 0.1 g) in DMF(10 mL) was added with *α, β*-unsaturated compounds (1.0 mmol) and then the mixture was heated at 65 °C for 6 h. After the reaction completion, the resulting mixture was cooled and the catalyst was removed by simple filtration. Then the solvent was poured into ice-cold water and the precipitate thus obtained was filtered, to give the crude product. The crude product was recrystallized from EtOH to give the corresponding analytically pure products.

4a: White crystals. Mp 99-100 °C. IR (KBr): 3284, 3036, 2926, 2895, 1733. ^{1}H NMR (400 MHz, $CDCl_3$) δ (ppm): 2.65 (t, J = 6.8 Hz, 2H, CH_2), 3.57-3.67 (m, 6H, CH_2), 3.69 (s, 3H, CH_3), 5.30 (s, 2H, CH_2), 6.95-7.00 (m, 2H, ArH), 7.21-7.30 (m, 3H, ArH), 8.22 (s, 1H, NH). ^{13}C NMR (100 MHz, $CDCl_3$): δ (ppm): 32.73, 40.48, 40.70, 46.65, 51.82, 65.33, 114.99, 121.59, 129.53, 157.74, 158.48, 158.57, 172.41, 177.50. Anal. Calcd for $C_{16}H_{19}N_5O_3S$: C, 53.17; H, 5.30; N, 19.38. Found: C, 53.31; H, 5.28; N, 19.30.

4b: Straw yellow crystals. Mp 95-96 °C. IR (KBr): 3330, 3022, 2989-2895, 1736. ^{1}H NMR (400 MHz, $CDCl_3$) δ (ppm): 2.30 (s, 3H,CH_3), 2.61 (t, J = 6.4 Hz, 2H, CH_2), 3.38-3.42 (m, 2H, CH_2), 3.63-3.67 (m, 4H, CH_2), 3.81 (s, 3H, CH_3), 5.31 (s, 2H, CH_2), 6.86 (d, J = 7.6 Hz, 2H, ArH), 7.08 (d, J = 7.6 Hz, 2H, ArH), 8.21 (s, 1H, NH). ^{13}C NMR (100 MHz, $CDCl_3$): δ (ppm): 22.52, 32.89, 38.74, 45.37, 46.61, 51.84, 63.67, 114.86, 129.62, 130.86, 156.30, 157.72, 170.97, 178.03. Anal. Calcd for $C_{17}H_{21}N_5O_3S$: C, 54.38; H, 5.64; N, 18.65. Found: C, 54.16; H, 5.66; N, 18.72.

4c: White crystals. Mp 82-84 °C. IR (KBr): 3310, 3022, 2979-2895, 1740. ^{1}H NMR (400 MHz, $CDCl_3$) δ (ppm): 2.23 (s, 3H, CH_3), 2.60 (t, J = 6.8 Hz, 2H, CH_2), 3.58-3.67 (m, 6H, CH_2), 3.71 (s, 3H, CH_3), 5.33 (s, 2H, CH_2), 6.78-6.85 (m, 3H, ArH), 6.95-7.01 (m, 1H, ArH), 8.30 (s, 1H, NH). ^{13}C NMR (100 MHz, $CDCl_3$): δ (ppm): 21.42, 32.17, 40.28, 40.57, 46.54, 50.88, 65.53, 115.82, 118.14, 126.32, 128.97, 146.53, 157.22, 159.86, 162.32, 173.06, 177.45. Anal. Calcd for $C_{17}H_{21}N_5O_3S$: C, 54.38; H, 5.64; N, 18.65. Found: C, 54.13; H, 5.65; N, 18.72.

4d: White crystals. Mp 112-114 °C. IR (KBr): 3324, 3052, 2963-2892, 1736. ^{1}H NMR (400 MHz, $CDCl_3$) δ (ppm): 2.66 (t, J = 6.8 Hz, 2H, CH_2), 3.41-3.43 (m, 4H, CH_2), 3.68-3.71 (m, 3H, CH_3), 3.73-3.76 (m, 2H, CH_2), 5.32 (s, 2H, CH_2), 7.26-7.38 (m, 4H, ArH), 8.34 (s, 1H, NH). ^{13}C NMR (100 MHz, $CDCl_3$): δ (ppm): 32.35, 40.18, 40.52, 44.56, 51.83, 65.53, 114.57, 127.37, 130.74, 155.79, 158.61, 160.04, 172.46, 177.45. Anal. Calcd for $C_{16}H_{18}N_5O_3ClS$: C, 48.54; H, 4.58; N, 17.69. Found: C, 48.34; H, 4.59; N, 17.77.

4e: White crystals. Mp 54-56 °C. IR (KBr): 3287, 3061, 2981, 2899, 1730. ^{1}H NMR (400 MHz, $CDCl_3$) δ (ppm): 1.26 (t, J = 6.8 Hz, 3H, CH_3), 2.60 (t, J = 6.8 Hz, 2H, CH_2), 3.50-3.65 (m, 6H, CH_2), 4.14 (q, J = 6.8 Hz, 2H, CH_2), 5.29 (s, 2H, CH_2), 6.96-7.01 (m, 3H, ArH), 7.26-7.30 (m, 2H, ArH), 8.20 (s, 1H, NH). ^{13}C NMR (100 MHz, $CDCl_3$): δ (ppm): 14.14, 32.95, 40.41, 40.65, 46.56, 60.67, 65.29, 114.96, 121.54, 129.49, 157.71, 158.37, 158.54, 171.90, 177.96. Anal. Calcd for $C_{17}H_{21}N_5O_3S$: C, 54.38; H, 5.64; N, 18.65. Found: C, 54.59; H, 5.49; N, 18.52.

4f: White crystals. Mp 76-78 °C. IR (KBr): 3326, 3042, 2983-2875, 1732. ^{1}H NMR (400 MHz, $CDCl_3$) δ (ppm): 1.27 (t, J = 6.8 Hz, 3H, CH_3), 2.25 (s, 3H, CH_3), 2.63 (t, J = 6.8 Hz, 2H, CH_2), 3.62-3.69 (m, 6H, CH_2), 4.14 (q, J = 6.8 Hz, 2H, CH_2), 5.27 (s, 2H, CH_2), 6.86 (d, J = 8.0 Hz, 2H, ArH), 7.11 (d, J = 8.0 Hz, 2H, ArH), 8.38 (s, 1H, NH). ^{13}C NMR (100 MHz, $CDCl_3$): δ (ppm): 14.31, 21.23, 33.88, 40.24, 40.56, 45.56, 60.85, 64.76, 114.60, 121.76, 129.94, 157.07, 158.38, 158.56, 171.93, 176.81. Anal. Calcd for $C_{18}H_{23}N_5O_3S$: C, 55.51; H, 5.95; N, 17.98. Found: C, 55.30; H, 5.97; N, 17.91.

4g: Straw yellow crystals. Mp 56-58 °C. IR (KBr): 3254, 3078, 2958-2906, 1722. ^{1}H NMR (400 MHz, $CDCl_3$) δ (ppm): 1.25 (t, J = 7.2 Hz, 3H, CH_3), 2.31 (s, 3H, CH_3), 2.59 (t, J = 6.8 Hz, 2H, CH_2), 3.39-3.42 (m, 4H, CH_2), 3.76 (t, J = 6.8 Hz, 2H, CH_2), 4.14 (q, J = 7.2 Hz, 2H, CH_2), 5.29 (s, 2H, CH_2), 6.86-6.95 (m, 3H, ArH), 7.01-7.04 (m, 1H, ArH), 8.32 (s, 1H, NH). ^{13}C NMR (100 MHz, $CDCl_3$): δ (ppm): 15.23, 22.14, 32.36, 40.21, 40.75, 46.45, 59.54, 65.35, 115.28, 118.40, 126.21, 129.78, 145.63, 157.32, 158.96, 163.22, 172.60, 178.55. Anal. Calcd for $C_{18}H_{23}N_5O_3S$: C, 55.51; H, 5.95; N, 17.98. Found: C, 55.26; H, 5.94; N, 18.05.

4h: Straw yellow crystals. Mp 66-68 °C. IR (KBr): 3259, 3049, 2937-2858, 1730. ^{1}H NMR (400 MHz, $CDCl_3$) δ (ppm): 1.27 (t, J = 6.8 Hz, 3H, CH_3), 2.63 (t, J = 6.4 Hz, 2H, CH_2), 3.40-3.42 (m, 4H, CH_2), 3.74 (t, J = 6.4 Hz, 2H, CH_2), 4.13 (q, J = 6.8 Hz, 2H, CH_2), 5.27 (s, 2H, CH_2), 7.21-7.36 (m, 4H, ArH), 8.27 (s, 1H, NH). ^{13}C NMR (100 MHz, $CDCl_3$): δ (ppm): 13.14, 32.19, 41.24, 42.06, 46.58, 60.77, 65.78, 114.89, 127.44, 130.76, 154.79, 156.87, 160.06, 171.03, 177.73. Anal. Calcd for $C_{17}H_{20}N_5O_3ClS$: C, 49.81; H, 4.92; N, 17.09. Found: C, 49.59; H, 4.94; N, 17.15.

4i: White crystals. Mp 110-111 °C. IR (KBr): 3285, 3068, 2920, 2893, 2245. ^{1}H NMR (400 MHz, $CDCl_3$) δ (ppm): 2.70 (s, 2H, CH_2), 3.63 (s, 2H, CH_2), 3.71 (s, 4H, CH_2), 5.30 (s, 2H, CH_2), 6.99-7.01 (m, 3H, ArH), 7.27-7.31 (m, 2H, ArH), 8.29 (s, 1H, NH). ^{13}C NMR (100 MHz, $CDCl_3$): δ (ppm): 17.00, 40.76, 41.02, 46.92, 65.25, 114.94, 118.36, 121.64, 129.54, 157.67, 158.12, 158.95, 176.99. Anal. Calcd for

$C_{15}H_{16}N_6OS$: C, 54.86; H, 4.91; N, 25.59. Found: C, 55.02; H, 4.90; N, 25.48.

4j: White crystals. Mp 138-140 °C. IR (KBr): 3285, 3063, 2936-2857, 2251. ^{1}H NMR (400 MHz, $CDCl_3$) δ (ppm): 2.37 (s, 3H, CH3), 2.47 (t, J = 5.6 Hz, 2H, CH_2), 3.64-3.70 (m, 6H, CH_2), 5.23 (s, 2H, CH_2), 7.08 (d, J = 8.4 Hz, 2H, ArH), 7.27 (d, J = 8.4 Hz, 2H, ArH), 8.32 (s, 1H, NH). ^{13}C NMR (100 MHz, $CDCl_3$): δ (ppm): 17.06, 23.14, 40.67, 41.02, 46.29, 65.25, 114.85, 118.93, 129.85, 138.78, 152.54, 155.16, 157.65, 176.98. Anal. Calcd for $C_{16}H_{18}N_6OS$: C, 56.12; H, 5.30; N, 24.54. Found: C, 55.98; H, 5.31; N, 24.65.

4k: White crystals. Mp 114-116 °C. IR (KBr): 3224, 3028, 2953-2874, 2241. ^{1}H NMR (400 MHz, $CDCl_3$) δ (ppm): 2.31 (s, 3H, CH_3), 2.76 (t, J = 6.0 Hz, 2H, CH_2), 3.54-3.59 (m, 2H, CH_2), 3.65 (t, J = 6.0 Hz, 2H, CH_2), 3.84 (t, J = 6.0 Hz, 2H, CH_2), 5.30 (s, 2H, CH_2), 6.85-6.90 (m, 3H, ArH), 6.98-7.02 (m, 1H, ArH), 8.35(s, 1H, NH) ^{13}C NMR (100 MHz, $CDCl_3$): δ (ppm): 17.51, 20.76, 40.57, 41.22, 46.28, 65.43, 115.51, 118.12, 118.93, 126.74, 130.16, 143.66, 158.75, 160.23, 162.74, 177.86. Anal. Calcd for $C_{16}H_{18}N_6OS$: C, 56.12; H, 5.30; N, 24.54. Found: C, 55.88; H, 5.28; N, 24.65.

4l: White crystals. Mp 120-121 °C. IR (KBr): 3336, 3041, 2960-2907, 2247. ^{1}H NMR (400 MHz, $CDCl_3$) δ (ppm): 2.33 (s, 3H, CH_3), 2.75 (t, J = 6.0 Hz, 2H, CH_2), 3.42-3.46 (m, 2H, CH_2), 3.55 (t, J = 6.0 Hz, 2H, CH_2), 3.74 (t, J = 6.0 Hz, 2H, CH_2), 5.28 (s, 2H, CH_2), 7.19 (d, J = 8.0 Hz, 2H, ArH), 7.36 (d, J = 8.0 Hz, 2H, ArH), 8.26 (s, 1H, NH). ^{13}C NMR (100 MHz, $CDCl_3$): δ (ppm): 17.61, 40.32, 41.01, 46.46, 66.12, 114.98, 119.93, 127.46, 130.68, 155.89, 159.07, 160.96, 177.92. Anal. Calcd for $C_{15}H_{15}N_6OClS$: C, 49.65; H, 4.17; N, 23.16. Found: C, 49.44; H, 4.19; N, 23.27.

4m: Straw yellow crystals. Mp 68-70 °C. IR (KBr): 3247, 2943, 2872, 1722. ^{1}H NMR (400 MHz, $CDCl_3$) δ (ppm): 1.96-1.99 (m, 2H, CH_2), 2.66-2.71 (m, 2H, CH_2), 3.38-3.43 (m, 4H, CH_2), 3.65-3.68 (m, 3H, CH_3), 3.74-3.76, (m, 2H, CH_2), 5.27 (s, 2H, CH_2), 6.95-7.02 (m, 3H, ArH), 7.27-7.31 (m, 2H, ArH), 9.42 (s, 1H, NH). ^{13}C NMR (100 MHz, $CDCl_3$): δ (ppm): 20.63, 33.46, 39.12, 46.09, 46.95, 60.26, 64.85, 115.02, 121.49, 129.53, 152.75, 156.67, 157.72, 172.36, 177.43. Anal. Calcd for $C_{17}H_{21}N_5O_3S$: C, 54.38; H, 5.64; N, 18.65. Found: C, 54.16; H, 5.63; N, 18.72.

4n: Straw yellow crystals. Mp 83-85 °C. IR (KBr): 3251, 2951, 2857, 1732. ^{1}H NMR (400 MHz, $CDCl_3$) δ (ppm): 1.93-1.98 (m, 2H, CH_2), 2.24 (s, 3H, CH_3), 2.65-2.68 (m, 2H, CH_2), 3.39-3.42 (m, 4H, CH_2), 3.66-3.68 (m, 3H, CH_3), 3.72-3.75 (m, 2H, CH_2), 5.23 (s, 2H, CH_2), 6.88 (d, J = 7.6 Hz, 2H, ArH), 7.11 (d, J = 7.6 Hz, 2H, ArH), 9.40 (s, 1H, NH). ^{13}C NMR (100 MHz, $CDCl_3$): δ (ppm): 20.46, 21.33, 32.98,

39.20, 45.73, 46.52, 51.68, 65.54, 114.86, 130.03, 130.87, 152.74, 155.66, 157.03, 173.80, 177.29. Anal. Calcd for $C_{18}H_{23}N_5O_3S$: C, 55.51; H, 5.95; N, 17.98. Found: C, 55.39; H, 5.96; N, 18.04.

4o: Orange crystals. Mp 60-62 °C. IR (KBr): 3250, 2947, 2871, 1726. ^{1}H NMR (400 MHz, $CDCl_3$) δ (ppm): 1.25 (t, J = 7.2 Hz, 3H, CH_3), 1.93-1.96 (m, 2H, CH_2), 2.65 (t, J = 6.8 Hz, 2H, CH_2), 3.39-3.42 (m, 4H, CH_2), 3.74 (t, J = 6.8 Hz, 2H, CH_2), 4.12 (q, J = 7.2 Hz, 2H, CH_2), 5.27 (s, 2H, CH_2), 6.95-7.01 (m, 3H, ArH), 7.27-7.30 (m, 2H, ArH), 9.40 (s, 1H, NH). ^{13}C NMR (100 MHz, $CDCl_3$): δ (ppm): 14.17, 21.32, 33.26, 39.23, 45.71, 46.49, 60.56, 65.36, 114.98, 121.47, 129.49, 152.77, 156.76, 157.78, 172.37, 177.37. Anal. Calcd for $C_{18}H_{23}N_5O_3S$: C, 55.51; H, 5.95; N, 17.98. Found: C, 55.43; H, 5.93; N, 17.91.

4p: Straw yellow crystals. Mp 79-82 °C. IR (KBr): 3255, 2956-2872, 1727. ^{1}H NMR (400 MHz, $CDCl_3$) δ (ppm): 1.34 (t, J = 6.4 Hz, 3H, CH_3), 1.90-1.94 (m, 2H, CH_2), 2.42 (s, 3H, CH_3), 2.65 (t, J = 6.4 Hz, 2H, CH_2), 3.55-3.72 (m, 6H, CH_2), 4.11 (q, J = 6.4 Hz, 2H, CH_2), 5.30 (s, 2H, CH_2), 6.88 (d, J = 8.0 Hz, 2H, ArH), 7.10 (d, J = 8.0 Hz, 2H, ArH), 9.38 (s, 1H, NH). ^{13}C NMR (100 MHz, $CDCl_3$): δ (ppm): 14.16, 21.30, 23.54, 33.27, 39. 32, 45.12, 45.69, 61.56, 65.40, 114.69, 130.76, 130.85, 152.73, 156. 67, 157.61, 172.82, 178.01. Anal. Calcd for $C_{19}H_{25}N_5O_3S$: C, 56.56; H, 6.25; N, 17.36. Found: C, 56.31; H, 6.28; N, 17.29.

4q: Orange crystals. Mp 102-105 °C. IR (KBr): 3260, 2947-2873, 2243. ^{1}H NMR (400 MHz, $CDCl_3$) δ (ppm): 2.04-2.06 (m, 2H, CH_2), 2.76 (t, J = 6.4 Hz, 2H, CH_2), 3.40-3.43 (m, 2H, CH_2), 3.57 (t, J = 6.4 Hz, 2H, CH_2), 3.69 (t, J = 6.4 Hz, 2H, CH_2), 5.21 (s, 2H, CH_2), 6.97-7.02 (m, 3H, ArH), 7.28-7.32 (m, 2H, ArH), 9.43 (s, 1H, NH). ^{13}C NMR (100 MHz, $CDCl_3$): δ (ppm): 20.45, 21.34, 39.03, 46.52, 47.73, 64.72, 115.01, 117.88, 121.50, 129.72, 152.92, 154.31, 157.53, 176.81. Anal. Calcd for $C_{16}H_{18}N_6OS$: C, 56.12; H, 5.30; N, 24.54. Found: C, 55.89; H, 5.32; N, 24.65.

4r: White crystals. Mp 108-111 °C. IR (KBr): 3221, 3028, 2935, 2870, 2245. ^{1}H NMR (400 MHz, $CDCl_3$) δ (ppm): 2.01-2.07 (m, 2H, CH_2), 2.29 (s, 3H, CH_3), 2.76 (t, J = 6.0 Hz, 2H, CH_2), 3.43-3.46 (m, 2H, CH_2), 3.54 (t, J = 6.0 Hz, 2H, CH_2), 3.73 (t, J = 6.0 Hz, 2H, CH_2), 5.27 (s, 2H, CH_2), 6.89 (d, J = 8.0 Hz, 2H, ArH), 7.08 (d, J = 8.0 Hz, 2H, ArH), 9.50 (s, 1H, NH). ^{13}C NMR (100 MHz, $CDCl_3$): δ (ppm): 16.96, 20.48, 21.31, 39.24, 46.25, 47.37, 65.49, 114.84, 118.78, 129.96, 130.87, 152.46, 155.61, 157.61, 176.94. Anal. Calcd for $C_{17}H_{20}N_6OS$: C, 57.28; H, 5.66; N, 23.58. Found: C, 57.08; H, 5.68; N, 23.66.

4s: Yellow crystals. Mp 154-156 °C. IR (KBr): 3261, 3054, 2946, 2873, 1735. ^{1}H NMR (400 MHz, $CDCl_3$) δ (ppm): 2.68 (t, J = 6.4 Hz, 2H, CH_2), 3.49-3.54 (m, 4H, CH_2), 3.68 (s, 3H, CH_3), 3.75 (t, J = 6.4 Hz, 2H, CH_2), 7.27-7.38 (m, 3H, ArH), 7.56-7.64 (m, 2H, ArH), 8.23 (s, 1H, NH). ^{13}C NMR (100 MHz,

$CDCl_3$): δ (ppm): 32.37, 40.41, 40.72, 46.45, 52.02, 105.24, 111.56, 120.98, 123.24, 125.82, 127.98, 147.14, 149.21, 152.84, 155.02, 171.97, 176.53. Anal. Calcd for $C_{17}H_{17}N_5O_3S$: C, 54.97; H, 4.61; N, 18.86. Found: C, 54.75; H, 4.62; N, 18.94.

4t: Yellow crystals. Mp 136-138 °C. IR (KBr): 3237, 3121, 2945, 2859, 1722. ^{1}H NMR (400 MHz, $CDCl_3$) δ (ppm): 1.96-2.02 (m, 2H, CH_2), 2.72 (t, J = 6.4 Hz, 2H, CH_2), 3.44 (t, J = 6.0 Hz, 4H, CH_2), 3.70 (s, 3H, CH_3), 3.80 (t, J = 6.4 Hz, 2H, CH_2), 7.24-7.35 (m, 3H, ArH), 7.53-7.62 (m, 2H, ArH), 9.51 (s, 1H, NH). ^{13}C NMR (100 MHz, $CDCl_3$): δ (ppm): 21.27, 33.05, 39.25, 45.79, 46.52, 51.72, 104.82, 111.52, 121.50, 123.40, 125.29, 128.26, 148.35, 149.23, 152.90, 154.96, 172.75, 176.13. Anal. Calcd for $C_{18}H_{19}N_5O_3S$: C, 56.09; H, 4.97; N, 18.17. Found: C, 55.88; H, 4.99; N, 18.25.

4u: Yellow crystals. Mp 121-123 °C. IR (KBr): 3301, 3042, 2976, 2888, 1730. ^{1}H NMR (400 MHz, $CDCl_3$) δ (ppm): 1.26 (t, J = 7.2 Hz, 3H, CH_3), 2.63 (t, J = 6.4 Hz, 2H, CH_2), 3.38-3.43 (m, 4H, CH_2), 3.73 (t, J = 6.4 Hz, 2H, CH_2), 4.13 (q, J = 7.2 Hz, 2H, CH_2), 7.28-7.38 (m, 3H, ArH), 7.54-7.61 (m, 2H, ArH), 8.19 (s, 1H, NH). ^{13}C NMR (100 MHz, $CDCl_3$): δ (ppm): 14.31, 32.90, 40.42, 40.57, 46.53, 60.62, 105.18, 111.42, 121.38, 123.23, 126.07, 128.66, 148.70, 149.82, 153.02, 155.63, 172.23, 176.61. Anal. Calcd for $C_{18}H_{19}N_5O_3S$: C, 56.09; H, 4.97; N, 18.17. Found: C, 55.85; H, 4.99; N, 18.25.

4v: Yellow crystals. Mp 103-105 °C. IR (KBr): 3255, 3122, 3063, 2936, 2851, 1727. ^{1}H NMR (400 MHz, $CDCl_3$) δ (ppm): 1.27 (t, J = 7.2 Hz, 3H, CH_3), 1.96-2.02 (m, 2H, CH_2), 2.70 (t, J = 6.8 Hz, 2H, CH_2), 3.42-3.46 (m, 4H, CH_2), 3.80 (t, J = 6.8 Hz, 2H, CH_2), 4.15 (q, J = 7.2 Hz, 2H, CH_2), 7.24-7.35 (m, 3H, ArH), 7.53-7.62 (m, 2H, ArH), 9.51 (s, 1H, NH). ^{13}C NMR (100 MHz, $CDCl_3$): δ (ppm): 14.19, 21.28, 33.33, 39.26, 45.75, 46.49, 60.60, 104.82, 111.52, 121.50, 123.40, 125.28, 128.27, 148.38, 149.21, 152.93, 154.97, 172.33, 176.16. Anal. Calcd for $C_{19}H_{21}N_5O_3S$: C, 57.13; H, 5.30; N, 17.53. Found: C, 56.88; H, 5.31; N, 17.61.

4w: Brown crystals. Mp 193-195 °C. IR (KBr): 3285, 3065, 2921, 2889, 2243. ^{1}H NMR (400 MHz, $CDCl_3$) δ (ppm): 2.70 (t, J = 6.4 Hz, 2H, CH_2), 3.64 (t, J = 6.4 Hz, 2H, CH_2), 3.69 (s, 4H, CH_2), 7.24-7.34 (m, 3H, ArH), 7.52-7.60 (m, 2H, ArH), 8.20 (s, 1H, NH). ^{13}C NMR (100 MHz, $CDCl_3$): δ (ppm): 17.30, 40.75, 41.02, 46.96, 104.84, 111.61, 118.83, 121.74, 123.44, 126.20, 128.62, 147.77, 149.08, 153.17, 154.27, 176.36. Anal. Calcd for $C_{16}H_{14}N_6OS$: C, 56.79; H, 4.17; N, 24.84. Found: C, 56.55; H, 4.16; N, 24.94.

4x: Yellow crystals. Mp 173-175 °C. IR (KBr): 3226, 3121, 3064, 2948, 2872, 2246. ^{1}H NMR (400 MHz, $CDCl_3$) δ (ppm): 1.84-1.91 (m, 2H, CH_2), 2.74 (t, J = 6.0 Hz, 2H, CH_2), 3.42-3.46 (m, 2H, CH_2), 3.54 (t, J = 5.6 Hz, 2H, CH_2), 3.73, (t, J = 6.0 Hz, 2H, CH_2), 7.23-7.35 (m, 3H, ArH), 7.52-7.62 (m, 2H, ArH), 9.46 (s, 1H, NH). ^{13}C NMR (100 MHz, $CDCl_3$): δ (ppm): 17.01, 20.51, 39.38, 46.52, 47.71, 104.48 111.55, 118.68, 121.92, 123.54, 125.86, 128.58, 148.45, 149.13, 152.91, 154.62, 176.01. Anal. Calcd for $C_{17}H_{16}N_6OS$: C, 57.94; H, 4.58; N, 23.85. Found: C, 57.71; H, 4.60; N, 23.95.

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