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Synthesis of [1,2,4]Triazolo[3,4-a]isoquinolines and Pyrrolo[2,1-a]isoquinolines Using α-Keto Hydrazonoyl Halides

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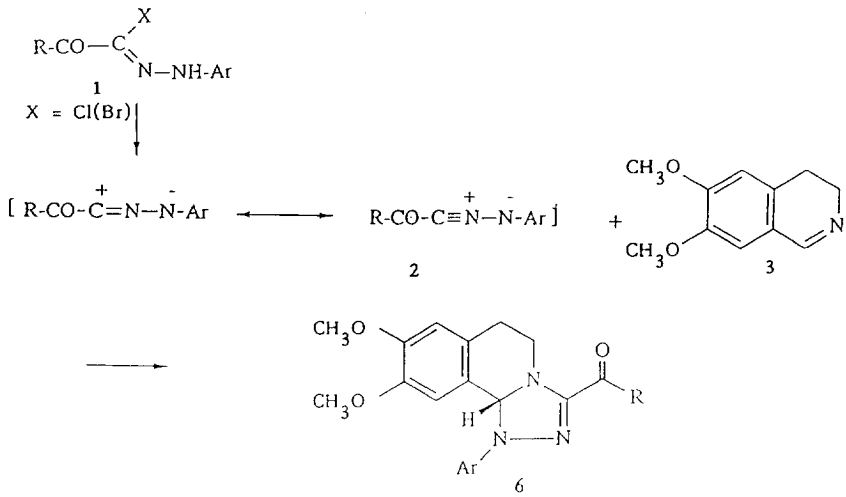
Abstract : Treatment of α-keto hydrazonoyl halides **1** with 3,4-dihydro-6,7-dimethoxyisoquinoline **3** and its 1-methyl derivative **4** in the presence of triethylamine in tetrahydrofuran at reflux afforded the corresponding cycloadducts **6** and **9**, respectively. The same halides **1** react with 1-cyanomethyl-3,4-dihydro-6,7-dimethoxyisoquinoline **5** and afforded pyrrolo[2,1-a]isoquinoline derivatives **12** in high yield.

The considerable pharmacological activities of [1,2,4]triazolo[3,4-a]isoquinolines and pyrrolo[2,1-a]isoquinolines as cardiovascular,[1] antiinflammatory[2] and antidepressant[3] have stimulated interest in the synthesis of these ring systems. The aim of the present study is on one hand to introduce an efficient one-pot synthesis for the title compounds which utilizes readily inexpensive reactants and gives good yields, and on the other hand to prepare compounds that might have expected pharmacological activity. In addition, this investigation indicates that 1-cyanomethyl-3,4-dihydro-6,7-dimethoxyisoquinoline **5** exhibits different behaviour from 3,4-dihydro-6,7-dimethoxyisoquinoline **3** and its 1-methyl derivative **4** towards α-keto hydrazonoyl halides **1**.

Reactions of 3,4-dihydro-6,7-dimethoxyisoquinoline **3** with nitrilimines **2**, generated in situ by the action of triethylamine on the corresponding α-keto hydrazonoyl halides **1**, were carried out by heating at reflux equimolar amounts of the reactants in tetrahydrofuran for 6 hr. In all cases only one cycloadduct was formed as was shown by TLC and ^{1}H NMR of the crude reaction mixture. All the isolated cycloadducts **6** gave satisfactory elemental analyses and spectroscopic data (IR, ^{1}H NMR, MS) for the proposed structures. For example, the ^{1}H NMR spectrum of each compound showed .a singlet signal at δ 6.6 ppm assignable to the proton at position 10b (Scheme 1). This chemical shift value is higher than that of CH-1 in the starting dipolarophile 3.[4]

Next , the reaction of nitrilimines **2** with dipolarophiles having active groups at C-1, namely, 1-methyl-3,4-dihydro-6,7-dimethoxyisoquinoline **4** and 1-cyanomethyl-3,4-dihydro-6,7-dimethoxyisoquinoline **5** were studied. The ability of 1-methyl-3,4-dihydro-6,7-dimethoxyisoquinoline **4** to behave as cyclic ketimine **4B** or as a secondary enamine **4A** has been discussed by many investigators.[5,6] Our aim point of interest whether addition of nitrilimines occured on enamine double bond of **4A** or the C=N double bond of ketimine structure **4B** (Scheme 2). Thus, when these reactions were carried out in a similar manner as that of **2** with **3**, they gave products whose elemental analyses were compatible with spiropyrazoline **7**, triazine derivatives **8** or the triazole derivatives **9**. The structures **7** and **8** were discarded on the basis of ^{1}H NMR evidence. For example, while



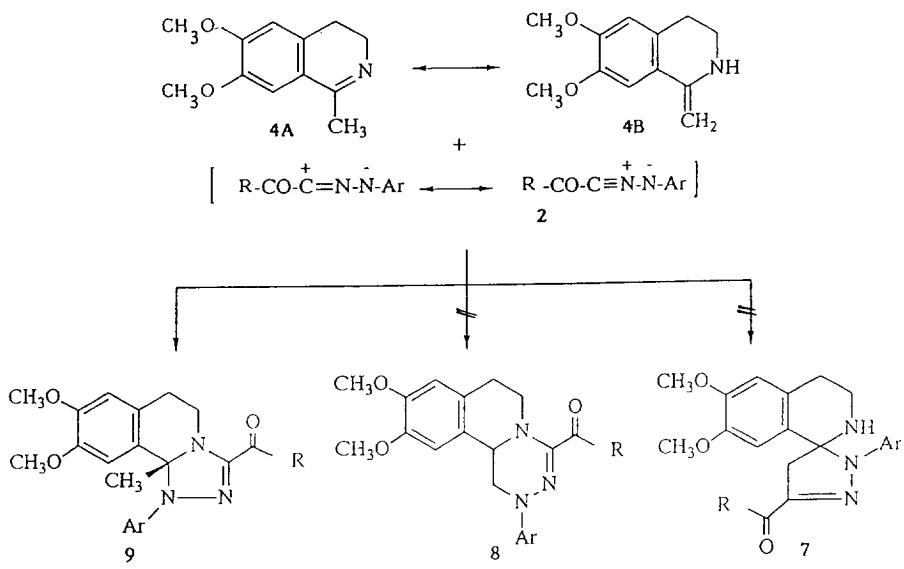
	R/Ar
a	CH_3/C_6H_5
b	CH_3/4-$CH_3C_6H_4$
c	C_6H_5/C_6H_5
d	2-naphthyl/ C_6H_5

	R/Ar
e	2-naphthyl/4-$CH_3C_6H_4$
f	2-thienyl/C_6H_5
g	2-thienyl/4-$CH_3C_6H_4$

Scheme 1

structure **8** is expected to reveal doublet and triplet signals assignable to C1-H_2 and C11b-H protons respectively, the other isomeric structure **7** will reveal two singlet signals assignable to CH_2 and NH protons. Such signals were absent in the ^{1}H NMR spectra of the product isolated from the reaction of **2** with **4**. Instead of these signals, the ^{1}H NMR spectra of the latter products showed one singlet signal of δ 1.9 ppm. The presence of such a signal is compatible with the assigned structure **9**. Indeed the proton resonance of the moiety -N=C(CH_3)- appear in the ^{1}H NMR spectra of the dipolarophile **4** at δ 2.37 ppm. This resonance was shifted to higher field in the ^{1}H NMR spectra (δ 1.9 ppm) of the cycloadducts **9** indicating the conversion of such moiety to the saturated moiety -N-C(CH_3) due to cycloaddition. On the basis of these finding the products isolated from the reaction mixture were assigned structure **9** (Scheme 2).

Treatment of α-keto hydrazonoyl halides **1** with 1-cyanomethyl - 3,4-dihydro-6,7-dimethoxyisoquinoline **5** in tetrahydrofuran in the presence of triethylamine at reflux afforded, in each case , a single product as evidenced by TLC and ^{1}H NMR of the crude reaction product (Scheme 3). The cycloadduct structure **13** for the product was ruled out on the basis of elemental analyses and spectroscopic data (IR, ^{1}H NMR, MS,UV). For example, the IR spectra of the products showed conjugated nitrile absorption band near 2210 cm^{-1} and absence of carbonyl absorption band. The mass spectra of all compounds exhibit a molecular ion peak with high intensity. Furthermore the electronic absorption spectrum of **12** in ethanol revealed, in each case, four intense maxima at λ near 480, 410, 330 and 250 nm assignable to arylazo chromophore. On the basis of these findings, the products isolated from reaction of **1** with **5** were assigned structure **12**.



	R/Ar		R/Ar
a	C_6H_5/C_6H_5	c	2-thienyl/C_6H_5
b	C_6H_5/4-$CH_3C_6H_4$	d	2-thienyl/4-$CH_3C_6H_4$
		e	C_6H_5NH/C_6H_5

Scheme 2

The reaction pathway that seems to account for the formation of **12** from **5** and **2** is outlined in Scheme 3. It is proposed that the reaction involves initial nucleophilic substitution to give **10**. The latter intermediate **10** tautomerizes to give **11** which cyclized *via* elimination of the elements of water to give **12**.

EXPERIMENTAL

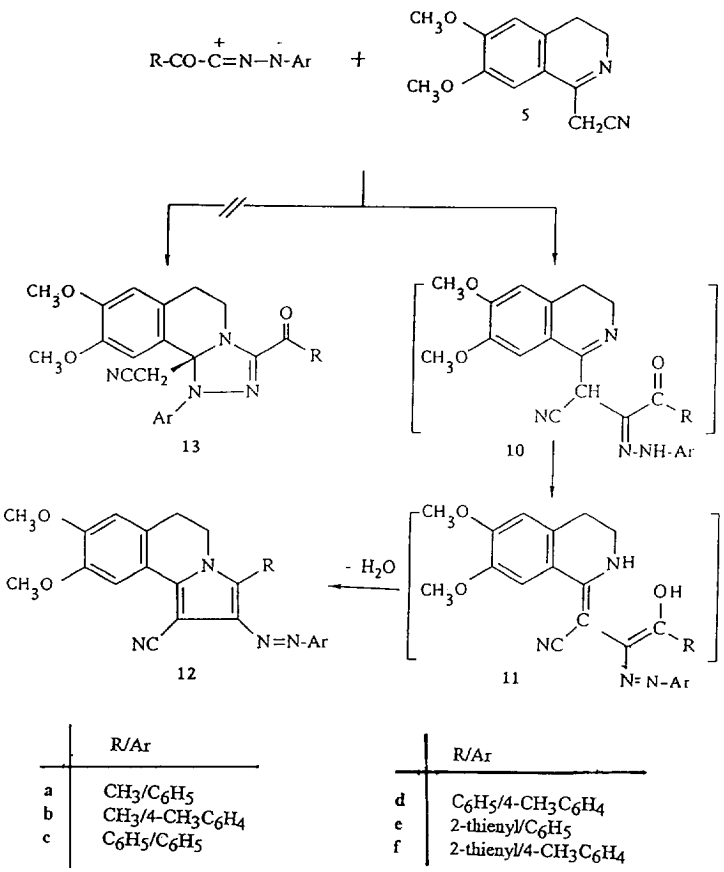
Melting points were measured on a Gallenkamp melting point apparatus. The infrared spectra were recorded on a Perkin-Elmer 1430 spectrometer. The ^{1}H NMR spectra were recorded in deuterated chloroform or DMSO-d6 on a Varian T-60 NMR spectrometer using tetramethylsilane as internal reference. Mass spectra were recorded on GCMS-QP1000 EX spectrometer. Elemental analyses were carried out at the Microanalytical Laboratory, University of Cairo, Giza, Egypt.

3,4-Dihydo-6,7-dimethoxyisoquinolines **3**[6], **4**[7], **5**[8], α-keto hydrazonoyl halides **1a**, **b**[9], **1c**, **d**[10], **1e**, **f**[11], **1g**, **h**[12] were prepared following the procedure reported in literature.

[1,2,4]triazolo[3,4-a]isoquinolines 6 and 9

General procedure.

To a solution of hydrazonoyl halides **1** (5 mmol) and the appropriate dipolarophile **3** or **4** (5 mmol) in tetrahydrofuran (40 ml) was added triethylamine (1.4 ml, 10 mmol) at room temperature. The reaction mixture was refluxed for 6 hr. The mixture was washed three times with water, the organic layer was collected, dried over anhydrous soduim sulfate and then filtered. The solvent was evaporated under reduced pressure and the residue was triturated with methanol (10 ml) where it solidified. The crude product was collected and crystallized from suitable solvents. The products **6** and **9** were obtained in 70-85 % yield and their physical constants are given below:



Scheme 3

6a: Yield (70 %); m.p. 130°C (ethanol); IR (KBr) υ 1660 (C=O) cm^{-1}; ^1H NMR ($CDCl_3$) δ 6.9-7.4 (m, 5H), 6.7 (s, 1H), 6.6 (s, 1H), 6.5 (s, 1H), 4.0 (m, 2H), 3.7 (s, 3H), 3.6 (s, 3H), 2.7 (m, 2H), 2.5 (s, 3H) ppm; MS m/e (%) 351 (100.0), 335 (9.0), 309 (7.0), 293 (8.0), 190 (8.0), 176 (8.0), 104 (4.0), 91 (15.0), 77 (16.0), 51 (8.0); (Calcd for $C_{20}H_{21}N_3O_3$: C, 68.4; H, 6.0; N, 12.0. Found: C, 68.4; H, 5.8; N, 11.8).

6b: Yield (71.0 %); m.p. 135°C (ethanol); IR (KBr)υ 1661 (C=O) cm^{-1}; ^1H NMR ($CDCl_3$) δ 7.1-7.3 (m, 4H), 6.7 (s, 1H), 6.6 (s, 1H), 6.5 (s, 1H), 4.0 (m, 2H), 3.8 (s, 3H), 3.6 (s, 3H), 2.7 (m, 2H), 2.5 (s, 3H), 2.3 (s, 3H) ppm; MS m/e (%) 365 (34.0), 364 (100.0), 348 (9.0), 323 (7.0), 252 (3.0), 295 (4.0), 192 (8.0), 176 (5.0), 132 (5.0), 105 (12.0), 91 (8.0), 77 (4.0), 51 (7.0); (Calcd for $C_{21}H_{23}N_3O_3$: C, 69.0; H, 6.3; N, 11.6. Found: C, 68.8; H, 6.0; N, 11.2).

6c: Yield (85 %); m.p. 151°C (acetic acid); IR (KBr) υ 1665 (C=O) cm^{-1}; ^1H NMR ($CDCl_3$) δ 7.0-8.2 (m, 10H), 6.8 (s, 1H), 6.7 (s, 1H), 6.6 (s, 1H), 4.0 (m, 2H), 3.9 (s, 3H), 3.6 (s, 3H), 2.9 (m, 2H) ppm; (Calcd for $C_{25}H_{23}N_3O_3$: C, 72.6; H, 5.6; N, 10.2. Found: C, 72.4; H, 5.3; N, 10.4).

6d: Yield (73 %); m.p. 171-172°C (ethanol); IR (KBr) υ 1660 (C=O) cm^{-1}; ^1H NMR ($CDCl_3$) δ 6.8-8.8 (m, 12H), 6.8 (s, 1H), 6.7 (s, 1H), 6.6 (s, 1H), 4.0 (m, 2H), 3.9 (s, 3H), 3.7 (s, 3H), 2.9 (m, 2H) ppm; (Calcd for

$C_{29}H_{25}N_3O_3$: C, 75.1; H, 5.4; N, 9.1. Found: C, 75.3; H, 5.1; N, 8.9).

6e: Yield (75 %); m.p. 150°C (ethanol); IR (KBr) υ 1660 (C=O) cm^{-1}; ^1H NMR ($CDCl_3$) δ 7.1-8.8 (m, 11H), 6.8 (s, 1H), 6.7 (s, 1H), 6.6 (s, 1H), 4.0 (m, 2H), 3.9 (s, 3H), 3.7 (s, 3H), 2.9 (m, 2H), 2.3 (s, 3H) ppm; MS m/e (%) 478 (46./0), 477 (92.0), 321 (18.0), 296 (13.0), 280 (4.0), 267 (19.0), 253 (14.0), 239 (8.0), 217 (8.0), 190 (6.0), 155 (96.0), 133 (4.0), 127 (100.0), 103 (10.0), 91 (20.0), 77 (12.09), 65 (14.0), 51 (7.0); (Calcd for $C_{30}H_{27}N_3O_3$: C, 75.4; H, 5.7; N, 8.8. Found: C, 75.6; H, 5.5; N, 8.9).

6f: Yield (70 %); m.p. 274-276°C (acetic acid); IR (KBr) υ 1650 (C=O) cm^{-1}; ^1H NMR (DMSO-d_6) δ 7.9-8.5 (m, 8H), 7.4 (s, 1H), 7.3 (s, 1H), 6.3 (s, 1H), 4.8 (m, 2H), 3.9 (s, 3H), 3.4 (s, 3H), 2.5 (m, 2H) ppm; MS m/e (%) 419 (16.0), 417 (81.0), 307 (20.0), 280 (19.)), 222 (4.0), 111 (100.0), 91 (3.0), 77 (16.0), 51 (10.0); (Calcd for $C_{23}H_{21}N_3O_3S$: C, 65.9,; H, 5.1; N, 10.1; S, 7.6. Found: C, 65.7; H, 4.9; N, 9.9; S, 7.5).

6g: Yield (75 %); m.p. 158°C (acetic acid); IR (KBr) υ 1650 (C=O) cm^{-1}; ^1H NMR ($CDCl_3$) δ 7.1-8.2 (m, 7H), 6.7 (s, 1H), 6.6 (s, 1H), 6.5 (s, 1H), 4.0 (m, 2H), 3.9 (s, 3H), 3.6 (s, 3H), 2.8 (m, 2H), 2.3 (s, 3H) ppm; MS m/e (%) 433 (35.0), 432 (100.0), 417 (8.0), 322 (5.0), 293 (2.0), 267 (5.0), 217 (5.0), 190 (4.0), 174 (1.0), 111 (74.0), 91 (8.0), 77 (6.0), 51 (4.0); (Calcd for $C_{24}H_{23}N_3O_3S$: C, 66.5; H, 5.4; N, 9.7; S, 7.4. Found: C, 66.5; H, 5.1; N, 9.8; S, 7.2).

9a: Yield (74 %); m.p. 170°C (ethanol); IR (KBr) υ 1640 (C=O) cm^{-1}; ^1H NMR ($CDCl_3$) δ 7.2-8.1 (m, 10H), 6.5 (s, 1H), 5.9 (s, 1H), 4.6 (m, 2H), 3.8 (s, 3H), 3.2 (s, 3H), 2.5 (m, 2H), 2.2 (s, 3H) ppm; MS m/e (%) 427 (1.0), 412 (100.0), 396 (12.0), 306 (3.0), 204 (5.0), 190 (5.0), 103 (65.0), 91 (5.0), 77 (62.0), 51 (13.0); (Calcd for $C_{26}H_{25}N_3O_3$: C, 73.1; H, 5.9; N, 9.9. Found: C, 73.2; H, 5.6; N, 9.7).

9b: Yield (78 %); m.p. 137°C (ethanol); IR (KBr) υ 1640 (C=O) cm-1; ^1H NMR ($CDCl_3$) δ 7.1-8.0 (m, 9H), 6.5 (s, 1H), 5.9 (s, 1H), 4.7 (m, 2H), 3.8 (s, 3H), 3.3 (s, 3H), 3.0 (m, 2H), 2.8 (s, 3H), 2.2 (s, 3H) ppm; (Calcd for $C_{27}H_{27}N_3O_3$: C, 73.4; H, 6.2; N, 9.6. Found: C, 73.5; H, 6.0; N, 9.3).

9c: Yield (72 %); m.p. 176°C (ethanol); IR (KBr) υ 1640 (C=O) cm^{-1}; ^1H NMR ($CDCl_3$) δ 7.9-8.2 (m, 8H), 6.4 (s, 1H), 5.9 (s, 1H), 4.7 (m, 2H), 3.8 (s,3H), 3.3 (s,3H), 2.5 (m, 2H), 2.2 (s, 3H) ppm; MS m/e (%) 434 (1.0), 419 (100.0), 403 (13.0), 294 (1.0), 231 (1.0), 190 (3.0), 111 (56.0), 77 (15.0), 51 (6.0); (Calcd for $C_{24}H_{23}N_3O_3S$: C, 66.5; H, 5.4; N, 9.7; S, 7.4. Found: C, 66.4; H, 5.1; N, 9.5; S, 7.2).

9d: Yield (74 %); m.p. 173-174°C (ethanol); IR (KBr) υ 1645 (C=O) cm^{-1}; 1H NMR ($CDCl_3$) δ 7.1-8.1 (m, 7H), 6.4 (s, 1H), 5.9 (s, 1H), 4.7 (m, 2H), 3.8 (s, 3H), 3.3 (s, 3H), 3.0 (m, 2H), 2.3 (s, 3H), 2.2 (s, 3H) ppm; (Calcd for $C_{25}H_{25}N_3O_3S$: C, 67.1; H, 5.6; N, 9.4; S, 7.2. Found: C, 67.4; H, 5.4; N, 9.2; S, 7.1).

9e: Yield (84 %); m.p. 140°C (ethanol); IR (KBr) υ 3288 (NH), 1680 (C=O) cm^{-1}; ^1H NMR ($CDCl_3$) δ 8.6 (s, 1H), 7.1-7.6 (m, 10H), 6.5 (s, 1H), 5.8 (s, 1H), 4.9 (m, 2H), 3.8 (s, 3H), 3.3 (s, 3H), 2.5 (m, 2H), 2.1 (s, 3H) ppm; MS m/e (%)), 442 (100.0), 336 (3.0), 306 (10.0), 279 (3.0), 232 (7.0), 207 (5.0), 191 (7.0), 175 (4.0), 145 (2.0), 132 (3.0), 118 (3.0), 91 (15.0), 77 (15.0), 51 (4.0); (Calcd for $C_{26}H_{26}N_4O_3$: C, 70.6; H, 5.9; N, 12.7. Found: C, 70.8; H, 6.0; N, 12.5).

Pyrrolo[2, 1-a]-5, 6- dihydroisoquinoline derivatives 12a-f

The compounds **12a-f** were prepared in 75- 85 % yield by the same method described for the synthesis of **6** and **9** using 3, 4- dihydro - 6, 7- dimethoxyisoquinoline 1- carbonitriles **5** in place of **3** or **4**. The compounds prepared with their physical constants are listed below.

12a: Yield (75 %); m.p. 283-285°C (DMF); IR (KBr) υ 2222 (C≡N) cm^{-1}; ^1H NMR (DMSO-d_6) δ 7.4-7.9 (m, 5H), 7.3 (s, 1H), 6.7 (s, 1H), 4.0 (m, 2H), 3.98 (s, 3H), 3.94 (s, 3H), 3.1 (m, 2H), 2.6 (s, 3H) ppm; MS m/e (%) 372 (100.0), 267 (8.0), 251 (15.0), 236 (7.0), 223 (14.0), 210 (3.0), 192 (5.0), 180 (4.0), 154 (4.0), 133 (3.0),

127 (4.0), 114 (2.0), 102 (2.0), 90 (2.0), 77 (45.0), 51 (18.0); UV (ethanol), λ_{max} (nm): 480, 397, 324, 241 (Calcd for $C_{22}H_{20}N_4O_2$: C, 71.0; H, 5.4; N, 15.0. Found: C, 71.2; H, 5.5; N, 15.2).

12b: Yield (80 %); m.p. 262-264°C (DMF); IR (KBr) υ 2203 (C≡N) cm^{-1}; ^1H NMR (DMSO-d_6) δ 7.6-7.9 (m, 4H), 7.4 (s, 1H), 7.2 (s, 1H), 4.2 (m, 2H), 3.8 (s, 3H), 3.4 (s, 3H), 3.2 (m, 2H), 2.5 (s, 3H), 2.3 (s, 3H) ppm; MS m/e (%) 386 (100.0), 267 (79.0), 252 (13.0), 236 (7.0), 223 (11.0), 193 (12.0), 181 (4.0), 154 (5.0), 133 (1.0), 127 (4.0), 115 (2.0), 103 (2.0), 91 (46.0), 77 (5.0), 65 (21.0), 51 (6.0); UV (ethanol), λ_{max} (nm): 485,398, 324, 244 (Calcd for $C_{23}H_{22}N_4O_2$: C, 71.5; H, 5.7; N, 14.5. Found: C, 71.3; H, 5.5; N, 14.2).

12c: Yield (78 %); m.p. 296-298°C (DMF); IR (KBr) υ 2208 (C≡N) cm^{-1}; ^1H NMR (DMSO-d_6) δ 7.5-7.8 (m 10H), 7.5 (s, 1H), 7.4 (s, 1H), 3.9 (s, 3H), 3.8 (m, 2H), 3.7 (s, 3H), 2.6 (m, 2H) ppm; MS m/e (%) 434 (100.0), 345 (10.0), 329 (49.0), 298 (72.0), 285 (22.0), 271 (9.0), 255 (28.0), 242 (14.0), 227 (5.0), 190 (3.0), 135 (3.0), 127 (3.0), 114 (4.0), 108 (4.0), 91 (3.0), 77 (77.0), 65 (4.0), 51 (31.0); UV (ethanol) λ_{max} (nm): 485, 410, 325, 255 (Calcd for $C_{27}H_{22}N_4O_2$: C, 74.7; H, 5.1; N, 12.9. Found: C, 74.4; H, 4.9; N, 12.7).

12d: Yield (80 %); m.p. 298-300°C (DMF); IR (KBr) υ 2205 (C≡N) cm^{-1}; ^1H NMR (DMSO-d_6) δ 7.5-7.8 (m, 9H), 7.5 (s, 1H), 7.4 (s, 1H), 3.9 (s, 3H), 3.8 (m, 2H), 3.7 (s, 3H), 2.6 (m, 2H), 2.4 (s, 3H) ppm; Ms m/e (%) 449 (88.0), 448 (100.0), 346 (13.0), 330 (41.0), 300 (16.0), 299 (56.0), 286 (17.0), 266 (18.0), 244 (10.0), 227 (4.0), 191 (2.0), 151 (2.0), 127 (4.0), 114 (3.0), 107 (5.0), 91 (50.0), 65 (23.0), 51 (8.0); UV (ethanol) λ_{max} (nm) : 480, 412, 329, 260 (Calcd for $C_{28}H_{24}N_4O_2$: C, 75.0; H, 5.4; N, 12.6. Found: C, 74.8; H, 5.2; N, 12.2).

12e: Yield (77 %); m.p. 278-280°C (DMF); IR (KBr) υ 2208 (C≡N) cm^{-1}; MS m/e (%) 440 (96.0), 439 (100.0), 351 (12.0), 335 (34.0), 319 (2.0), 304 (31.0), 291 (13.0), 261 (12.0), 234 (4.0), 220 (10.0), 213 (4.0), 190 (4.0), 153 (2.0), 132 (3.0), 127 (3.0), 114 (2.0), 102 (2.0), 93 (4.0), 77 (73.0), 51 (25.0);UV (ethanol),λ_{max} (nm): 482, 418, 330, 256 (Calcd for $C_{25}H_{20}N_4O_2S$: C, 68.2; H, 4.6; N, 12.7; S, 7.3. Found: C, 68.5; H, 4.8; N, 12.5; S, 7.5).

12f: Yield (85 %); m.p. 295-297°C (DMF); IR (KBr) υ 2210 (C≡N) cm^{-1}; ^1H NMR (DMSO-d_6) δ 7.5-8.0 (m,7H), 7.4 (s, 1H), 7.2 (s, 1H), 4.4 (m, 2H), 3.9 (s, 3H), 3.8 (s, 3H), 3.2 (m, 2H), 2.3 (s, 3H) ppm; MS m/e (%) 454 (100.0), 364 (2.0), 336 (26.0), 320 (17.0), 305 (19.0), 291 (8.0), 262 (10.0), 234 (2.0), 222 (2.0), 214 (1.0), 191 (1.0), 153 (1.0), 130 (1.0), 119 (1.0), 91 (29.0), 65 (12.0), 51 (3.);UV (ethanol), λ_{max} (nm): 487, 420, 335, 250 (Calcd for $C_{26}H_{22}N_4O_2S$: C, 68.7; H, 4.9; N, 12.3; S, 7.1. Found: C, 69.0; H, 4.6; N, 12.1; S, 7.0).

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