An Efficient Synthesis of 1-Thia-5-azaspiro[5.5]undec-2-ene and its Recyclization to 1,5-Diazaspiro[5.5]undec-2-ene and/or Spiro-Thieno[2,3-*d*]pyrimidin-4(1*H*)-one Derivatives

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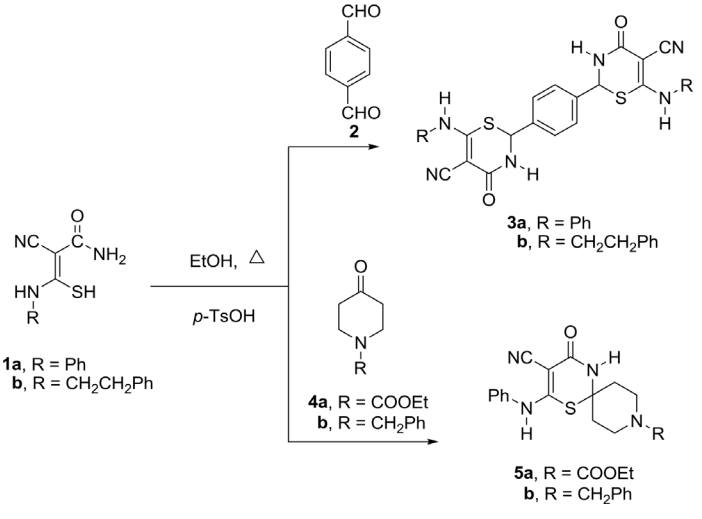
An efficient and direct synthesis of 1-thia-5-azaspiro[5.5]undec-2-ene is described, and the base-catalyzed recyclization of this compound was studied. The products have been characterized by elemental analyses, and IR, MS, ^{1}H NMR, and ^{13}C NMR spectroscopy.

Key words: 4-Oxo-1-phenylcyclohexanecarbonitrile, 1-Thia-5-azaspiro[5.5]undec-2-ene, Spiro-1,3-thiazine, Spiropyrimidine, Spiro-thieno[2,3-*d*]pyrimidine

Introduction

Spiro-heterocyclic compounds are well known to posses various pharmacological activities, and hence their synthesis has always been a challenge and of attraction to organic chemists. These compounds may display pronounced antimicrobial [1], analgesic [2], antiinflammatory [2], antimycobacterial [3], antifungal [4], antitumor [5, 6], and antiviral [5, 6] activities. Furthermore, 1,3-thiazines are an important type of heterocycles showing also a wide variety of related properties [7 – 11].

Recently, we reported an efficient and direct procedure for the synthesis of 2,2′-(1,4-phenylene)bis-3,4-dihydro-2*H*-1,3-thiazin-4-ones **3** [12] and 1-thia-5,9-diaza-spiro[5.5]undec-2-enes **5** [13] *via* the cyclocondensation of 3-arylamino-2-cyano-3-mercapto-acrylamide (**1**) with terephthalaldehyde (**2**) and/or 4-oxo-piperidine derivatives **4** in the presence of catalytic amounts of *p*-toluenesulfonic (*p*-TsOH) acid in boiling ethanol (Scheme 1). Compounds **3** and **4** could have interesting effects on biological targets.

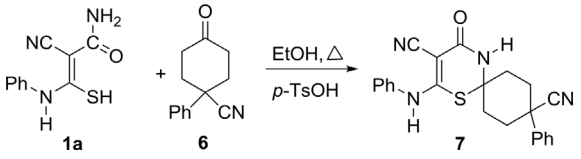


Scheme 1. Synthesis of bis-1,3-thiazines **3** and 1-thia-5,9-diaza-spiro[5.5]undec-2-enes **5**.

Prompted by the aforesaid biological and pharmaceutical activities, and as a part of our continuing interest in the synthesis of new polyfunctionally substituted heterocyclic [14 – 25] and spiro-heterocyclic compounds [26 – 29] of expected biological activity, we report here the versatile and hitherto unreported synthesis of a spiro-fused 1,3-thiazine-4-one **7** and its base-catalyzed recyclization to spiro-fused pyrimidin-4-one and spiro-thieno[2,3-*d*]pyrimidin-4-one derivatives with the purpose of investigating in the future their possible biological activity.

Results and Discussion

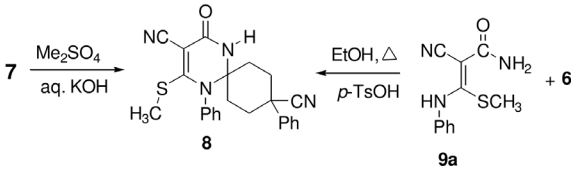
Scheme 2 outlines the synthesis of 1-thia-5-azaspiro[5.5]undec-2-ene **7** from the cyclocondensation of 2-cyano-3-mercapto-3-phenylamino-acrylamide (**1a**) with 4-oxo-1-phenyl-cyclohexanecarbonitrile (**6**) in the presence of catalytic amounts of *p*-toluenesulfonic acid in boiling ethanol.



Scheme 2. Synthesis of the spiro-1,3-thiazine **7**.

The structure of compound **7** was determined on the basis of elemental analysis, spectral data and the chemical transformations outlined below. Thus, compound **7** exhibits an IR spectrum with strong absorption bands at 3174 (NH), 2208 (CN) and 1636 cm^{-1} (CO). Its ^1H NMR spectrum shows characteristic singlets at δ = 10.24 and 8.43 ppm for the exocyclic and endocyclic NH proton, respectively, in addition to an eight-proton multiplet in the region of δ = 2.01 – 2.43 ppm of the cyclohexyl protons, and a multiplet at δ = 7.23 – 7.57 ppm for the aromatic protons. The structure **7** was further confirmed by a ^{13}C NMR spectrum which revealed resonances at δ = 63.38 and 163.86 ppm consistent with the quaternary sp^3 carbon (spiro carbon) and the carbonyl group of the thiazine ring, respectively. Furthermore, the structure assigned for compound **7** was fully supported by its mass spectrum, which showed a molecular formula $C_{23}H_{20}N_4OS$ (*m/z* = 400 (6.5 %), $[M]^+$).

Next, we moved on to study the alkylation of **7** using dimethyl sulfate, ethyl iodide, benzyl chloride, ethyl bromoacetate, and bromoacetonitrile as alkylating agents under basic conditions (Schemes 3 – 5).

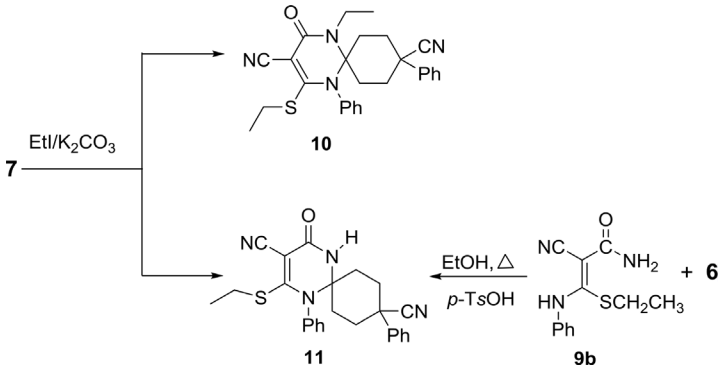


Scheme 3. Formation of the *S*-methylated spiro-fused pyrimidine **8**.

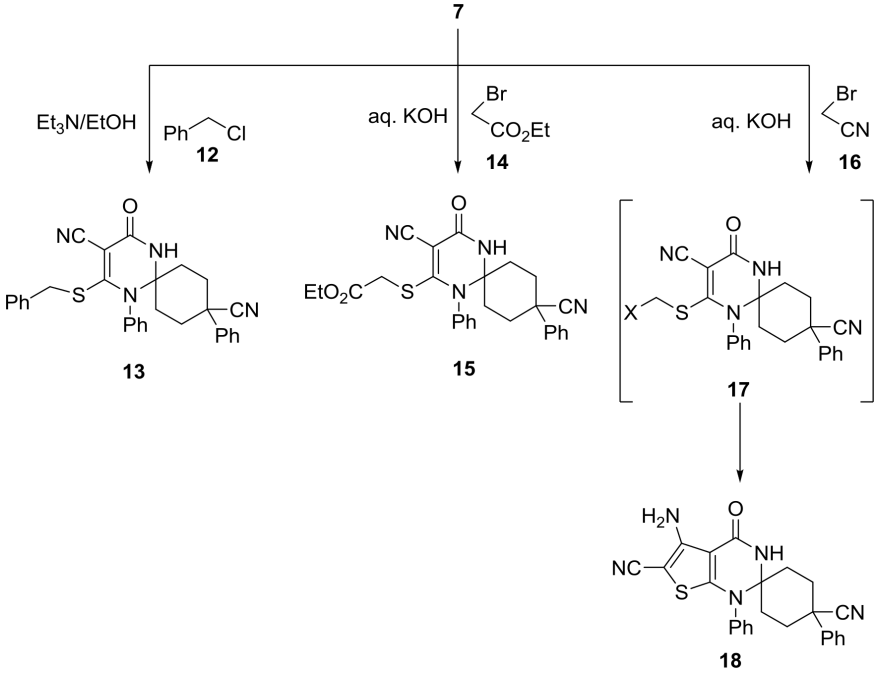
The reaction of **7** with dimethyl sulfate involved the sulfur atom thus affording the *S*-methylated derivative **8** (Scheme 3). The structural assignment of compound **8** was confirmed by its spectroscopic data. A distinction between the thiazine and pyrimidine structure types is clearly manifested in the ^1H and ^{13}C NMR spectra. The ^1H NMR spectrum of **8** showed the absence of an exocyclic NH proton, and in the ^{13}C NMR spectrum the resonance of the aminal carbon atom (δ = 73.87 ppm) is shifted downfield from that of the thioaminal carbon atom in compound **7**. The mass spectrum of **8** showed the molecular ion peak at *m/z* = 414, corresponding to the molecular formula $C_{24}H_{22}N_4OS$. Compounds **8** was prepared independently by the reaction of 3-(phenylamino)acrylamide **9a** with compound **6** in boiling ethanol and in the presence of catalytic amounts of *p*-toluenesulfonic acid. Both products had the same melting point, IR, and ^1H NMR data (Scheme 3).

The reaction of the spirothiazine **7** with ethyl iodide in DMF at r. t. and in the presence of anhydrous potassium carbonate did not afford the expected *S*-ethylated derivative **11**, but rather the diethylated species **10** (Scheme 4). The elemental analysis and the spectral data of compound **10** are in good agreement with the proposed structure. The IR and ^1H NMR spectra of **10** revealed the absence of NH groups and of signals attributable to the endo- and exocyclic NH protons of **7**, respectively. Also, the mass spectrum of **10** showed the molecular ion peak at *m/z* = 456, corresponding to the molecular formula $C_{27}H_{28}N_4OS$.

The *S*-ethylated derivative **11** was prepared by the reaction of 2-cyano-3-(ethylthio)-3-(phenylamino)acrylamide (**9b**) with compound **6** in boiling ethanol and in the presence of catalytic amounts of *p*-toluenesulfonic acid (Scheme 4). The structure of compound **11** was proven by elemental analysis and spectral data. The formula, $C_{25}H_{24}N_4OS$, of **11** was confirmed by the mass spectrum, which exhibited the molecular ion at *m/z* = 428. Furthermore, the IR and ^1H NMR spectra of **11** revealed the presence of an NH group and a characteristic singlet at δ = 8.51 ppm attributable to



Scheme 4. Synthesis of *S*-ethylated and diethylated spiro pyrimidines **10**, **11**.



Scheme 5. Synthesis of the thio- and thienopyrimidines **13**, **15** and **18**.

the endocyclic NH proton which was exchangable with D_2O.

Also, benzylation of **7** with benzyl chloride (**12**) in ethanol and in the presence of triethylamine gave the the *S*-benzylated compound **13** in high yield (Scheme 5). The structure of **13** was deduced on the basis of analytical and spectral data. The ^{1}H NMR spectrum revealed signals at δ = 4.19 and 8.41 ppm attributable to the SCH_2 and NH protons, respectively. Moreover, the mass spectrum of **13** exhibited a molecular ion peak at *m/z* = 490, and other significant peaks were as expected.

Similarly, the alkylation of **7** with ethyl bromoacetate (**14**) in aqueous KOH gave the spiro-fused *S*-substituted thiopyrimidine **15**. Its structure was deduced by IR, ^{1}H NMR, ^{13}C NMR, and mass spectra as well as by elemental analysis.

On the other hand, the reaction of **7** with bromoacetonitrile (**16**) under the same conditions gave the thienopyrimidine **18** (Scheme 5). The elemental analysis and the spectral data are in good agreement with the proposed structure. The ^{1}H NMR spectrum of **18** revealed the absence of signals attributable to the exocyclic NH protons of **7** and showed the presence of signals attributable to NH_2 protons. The structure of **18** is rationalized in terms of the initial formation of the intermediate **17**, which on subsequent intramolecular cyclization affords the final product (Scheme 5).

Experimental Section

General procedures

Melting points were measured with a Gallenkamp apparatus and are uncorrected. The reactions and the purity of the products were monitored by thin layer chromatography (TLC) on aluminum plates coated with silica gel with fluorescence indicator (Merck, 60 F_{254}) using $CHCl_3/CH_3OH$ (10 : 1) as eluent. The infrared spectra were recorded on a Jasco FT/IR-450 Plus infrared spectrophotometer. The NMR spectra were obtained on a JHA-LAA 400 WB-FT spectrometer (300 MHz for ^{1}H NMR, 75 MHz for ^{13}C NMR), with deuterated chloroform ($CDCl_3$) or dimethylsulfoxide ([D_6]DMSO) as solvents. Chemical shifts are quoted in δ and are referenced to TMS or the solvent signal. The mass spectra were recorded on a Trace GC 2000/Finngan Mat SSQ 7000 and a Shimadzu GCMS-QP-1000EX mass spectrometer at 70 eV. Elemental analyses were measured with a Vario EL III CHNOS elemental analyzer, in the Microanalytical Center of Cairo University. Compounds **1a** [28], **1b** [12], **9a** [29], and **9b** [12] were synthesized using the published procedures.

*3,9-Dicyano-4-oxo-9-phenyl-2-(phenylamino)-1-thia-5-azaspiro[5.5]undec-2-ene (**7**)*

A mixture of compound **1a** (2.19 g, 0.01 mol), 4-oxo-1-phenylcyclohexanecarbonitrile (**6**) (1.99 g, 0.01 mol) and *p*-toluenesulfonic acid (0.038 g, 0.002 mol) in ethanol (20 mL) was refluxed. A pale-yellow precipitate was formed after 3 h, and the reaction was continued for 5 h. The precipitate was filtered off, washed with ethanol, dried and recrystallized from DMF/EtOH. Pale-yellow powder, yield: 82 %, m. p. 237 – 238 °C. – IR (film): ν = 3174, 3032, 2943, 2208, 1636, 1551 cm^{-1}. – ^{1}H NMR (300 MHz, [D_6]DMSO): δ = 2.01 – 2.43 (m, 8H, 4CH_2), 7.23 – 7.57 (m, 10H, ArH), 8.43 (s, 1H, NH, D_2O-exchangeable), 10.24 (s, 1H, NH, D_2O-exchangeable). – ^{13}C NMR (75 MHz, [D_6]DMSO): δ = 31.27 (CH_2), 32.55 (CH_2), 34.42 (CH_2), 36.27 (CH_2), 42.51 (C-CN), 63.38 (C-6), 76.82 (C-3), 116.09 (CN), 121.85 (CN), 125.39, 126.73, 126.79, 128.17, 128.97, 129.11, 137.96, 139.55, 154.31 (C-Ar), 163.86 (C=O), 164.48 (C-2). – MS (EI, 70 eV): *m/z* (%) = 400 (6) $[M]^+$. – Anal. for $C_{23}H_{20}N_4OS$ (400.50): calcd. C 68.98, H 5.03, N 13.99, S 8.01; found C 68.86, H 5.19, N 13.87, S 7.91.

*3,9-Dicyano-2-methylthio-4-oxo-1,9-diphenyl-1,5-diazaspiro[5.5]undec-2-ene (**8**)*

To a stirred 0.75 N aqueous KOH solution (20 mL), compound **7** (0.40 g, 10 mmol) and dimethyl sulfate (0.25 g, 20 mmol) were added successively. The resulting precipitate was filtered off, washed with water, dried and recrystallized from ethanol. Colorless powder, yield: 77 %, m. p. 242 – 244 °C. – IR (film): ν = 3181, 3059, 2209, 1652, 1524 cm^{-1}. – ^{1}H NMR (300 MHz, $CDCl_3$): δ = 2.03 – 2.12 (m, 4H, 2CH_2), 2.37 – 2.48 (m, 4H, 2CH_2), 2.52 (s, 3H, SCH_3), 7.24 – 7.66 (m, 10H, ArH), 9.28 (s, 1H, NH). – ^{13}C NMR (75 MHz, $CDCl_3$): δ = 17.73 (SCH_3), 32.11 (CH_2), 32.19 (CH_2), 43.53 (CH_2), 44.36 (CH_2), 44.61 (C-CN), 73.87 (C-6), 76.85 (C-3), 115.8 (CN), 121.34 (CN), 125.52, 125.82, 125.84, 125.95, 128.06, 128.83, 129.43, 129.50, 138.52, 139.08 (C-Ar), 173.78 (C=O), 173.79 (C-2). – MS (EI, 70 eV): *m/z* (%) = 414 (3.61) $[M]^+$. – Anal. for $C_{24}H_{22}N_4OS$ (414.52): calcd. C 69.54, H 5.35, N 13.52, S 7.74; found C 69.48, H 5.41, N 13.45, S 7.67.

*Alternative synthesis of **8***

A mixture of compound **6** (1.99 g, 0.01 mol), **9a** (2.33 g, 0.01 mol), and *p*-toluenesulfonic acid (0.076 g, 0.01 mol) in ethanol (20 mL) was refluxed. A yellow precipitate was formed after 30 min, and stirring was continued for 2 h. The precipitate was filtered off, washed with ethanol, dried and recrystallized from the appropriate solvents.

*3,9-Dicyano-5-ethyl-2-ethylthio-1,9-diphenyl-4-oxo-1,5-diazaspiro [5.5]undec-2-ene (**10**)*

Ethyl iodide (0.31 g, 20 mmol) was added to a mixture of **7** (0.40 g, 10 mmol) and anhydrous potassium carbonate (0.28 g, 20 mmol) in DMF (5 mL). The reaction mixture was stirred for 18 – 20 h at r. t. and then poured into cold water. After stirring for 15 min, the precipitated product was collected by filtration, washed with water, dried and recrystallized from ethanol. Yellow crystals, yield: 76 %, m. p. 190 – 192 °C. – IR (film): ν = 2940, 2200, 1638 cm^{-1}. – ^{1}H NMR (300 MHz, [D_6]DMSO): δ = 1.14 (t, 3H, CH_3, *J* = 1.8 Hz), 1.30 (t, 3H, CH_3, *J* = 1.8 Hz), 1.79 – 2.39 (m, 8H, 4CH_2), 2.87 (q, 2H, SCH_2, *J* = 7.2 Hz), 4.22 (q, 2H, NCH_2, *J* = 7.2 Hz), 7.34 – 7.51 (m, 10H, ArH). – MS (EI, 70 eV): *m/z* (%) = 456 (8) $[M]^+$. – Anal. for $C_{27}H_{28}N_4OS$ (456.60): calcd. C 71.02, H 6.18, N 12.27, S 7.02; found C 70.92, H 6.24, N 12.21, S 6.94.

*3,9-Dicyano-2-ethylthio-4-oxo-1,9-diphenyl-1,5-diazaspiro-[5.5]undec-2-ene (**11**)*

A mixture of compound **6** (1.99 g, 0.01 mol), **9b** (2.47 g, 0.01 mol), and *p*-toluenesulfonic acid (0.076 g, 0.01 mol) in ethanol (20 mL) was refluxed. A colorless precipitate was formed after 30 min, and stirring was continued for 2 h. The precipitate was filtered off, washed with ethanol, dried and recrystallized from DMF/EtOH. Colorless crystals yield: 74 %, m. p. 248 – 250 °C. – IR (film): ν = 3161, 3042, 2965, 2207, 1644, 1504 cm^{-1}. – ^{1}H NMR (300 MHz, [D_6]DMSO): δ = 1.11 (t, 3H, CH_3, *J* = 7.2 Hz), 1.86 – 1.92 (m, 2H, CH_2), 2.02 – 2.07 (m, 2H, CH_2), 2.21 – 2.25

(m, 4H, $2CH_2$), 2.91 (q, 2H, SCH_2, J = 7.2 Hz), 7.36 – 7.55 (m, 10H, ArH), 8.51 (s, 1H, NH, D_2O-exchangeable). – MS (EI, 70 eV): *m/z* (%) = 428 (36.46) $[M]^+$. – Anal. for $C_{25}H_{24}N_4OS$ (428.55): calcd. C 70.07, H 5.64, N 13.07, S 7.48; found C 69.96, H 5.72, N 12.95, S 7.41.

*2-Benzylthio-3,9-dicyano-1,9-diphenyl-4-oxo-1,5-diazaspiro[5.5]undec-2-ene (**13**)*

A mixture of compound **7** (4.00 g, 0.01 mol), benzyl chloride (1.68 g, 0.011 mol), and triethylamine (2.1 mL, 0.015 mol) in ethanol (20 mL) was refluxed, and a colorless precipitate was formed after half an hour. The reaction was continued for 2 h, and the resulting precipitate was filtered off, dried, and recrystallized from DMF / EtOH. Colorless crystals yield: 79 %, m. p. 261 – 262 °C. – IR (film): ν = 3162, 3053, 2209, 1651, 1530 cm^{-1}. – ^1H NMR (300 MHz, $[D_6]$DMSO): δ = 1.72 – 1.81 (m, 2H, CH_2), 1.97 – 2.24 (m, 6H, $3CH_2$), 4.19 (s, 2H, SCH_2), 7.08 – 7.52 (m, 15H, ArH), 8.41 (s, 1H, NH, D_2O-exchangeable). – MS (EI, 70 eV): *m/z* (%) = 490 (5.6) $[M]^+$. – Anal. for $C_{30}H_{26}N_4OS$ (490.62): calcd. C 73.44, H 5.34, N 11.42, S 6.54; found C 73.36, H 5.42, N 11.32, S 6.48.

*Ethyl 2-(3,9-dicyano-4-oxo-1,9-diphenyl-1,5-diazaspiro-[5.5]undec-2-en-2-ylthio)acetate (**15**)*

To a stirred 0.75 N aqueous KOH solution (20 mL), compound **7** (0.40 g, 10 mmol) and ethyl bromoacetate (**14**) (0.33 g, 20 mmol) were added successively. The resulting precipitate was filtered off, washed with water, dried and recrystallized from ethanol. Colorless powder, yield: 77 %, m. p. 251 – 252 °C. – IR (film): ν = 3202, 3059, 2212, 1671, 1633, 1585 cm^{-1}. – ^1H NMR (300 MHz, $[D_6]$DMSO): δ = 1.21 (t, 3H, CH_3, J = 7.2 Hz), 1.88 – 2.23 (m, 8H, $4CH_2$), 3.36 (s, 2H, SCH_2), 4.12 (q, 2H, OCH_2, J = 7.2 Hz), 7.37 – 7.51 (m, 10H, ArH), 8.55 (s, 1H, NH). – ^{13}C NMR (75 MHz, $[D_6]$DMSO): δ = 13.89 (CH_3), 31.36 (CH_2), 31.82 (CH_2), 34.84 ($2CH_2$), 42.67 (SCH_2), 61.50 (C-CN), 73.73 (C-6), 86.87 (C-3), 116.30 (CN), 121.60 (CN), 125.52, 128.12, 128.90, 129.42, 129.57, 138.35, 139.45 (C-Ar), 160.66 (CO), 162.27 (CO), 167.42 (C-2) . – MS (EI, 70 eV): *m/z* (%) = 486 (20.33) $[M]^+$. – Anal. for $C_{27}H_{26}N_4O_3S$ (486.59): calcd. C 66.65, H 5.39, N 11.51, S 6.59; found C 66.47, H 5.51, N 11.42, S 6.53.

*5′-Amino-4,6′-dicyano-1′,4-diphenyl-4′-oxo-3′,4′-dihydro-1′H-spiro[cyclohexane-1,2′-thieno[2,3-d]pyrimidine] (**18**)*

To a stirred 0.75 N aqueous KOH solution (20 mL), compound **7** (0.40 g, 10 mmol) and bromoacetonitrile (**16**) (0.24 g, 20 mmol) were added successively. The resulting precipitate was filtered off, washed with water, dried and recrystallized from ethanol. Grey powder, yield: 80 %, m. p. 330 – 332 °C. – IR (film): ν = 3429 – 3310, 3202, 2185, 1642, 1514 cm^{-1}. – ^1H NMR (300 MHz, $[D_6]$DMSO): δ = 1.85 – 2.36 (m, 8H, $4CH_2$), 6.82 (s, 2H, NH_2), 7.34 – 7.59 (m, 10H, ArH), 8.50 (s, 1H, NH). – MS (EI, 70 eV): *m/z* (%) = 439 (46) $[M]^+$. – Anal. for $C_{25}H_{21}N_5OS$ (439.53): calcd. C 68.32, H 4.82, N 15.93, S 7.30; found C 68.24, H 4.93, N 15.84, S 7.18.

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