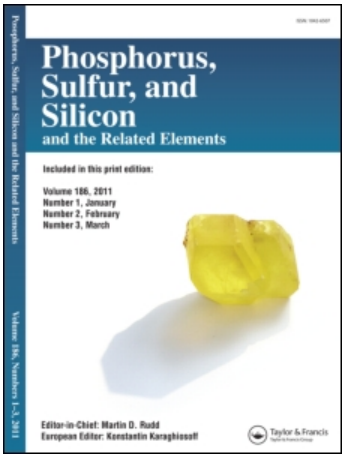


Phosphorus, Sulfur, and Silicon and the Related Elements
Publication details, including instructions for authors and subscription information:
http://www.informaworld.com/smpp/title~content=t713618290

A NEW GENERAL METHOD FOR SUBSTITUTED 4-ALKYLTHIO-*N*-ARYLSULPHONYL-AMINO-2-PYRIDONES: REACTION OF KETENE-*SS*-ACETALS WITH ARYLSULPHONYLHYDRAZIDES

Galal H. Elgemeie[a]; Hosny A. Ali[b]; Ahmed H. Elghandour[b]; Hassan M. Abdel-aziz[b]
[a] Department of Chemistry, Faculty of Science, Helwan University, Helwan, Cairo, Egypt [b] Department of Chemistry, Faculty of Science, Cairo University, Bani Suef, Egypt

To cite this Article Elgemeie, Galal H. , Ali, Hosny A. , Elghandour, Ahmed H. and Abdel-aziz, Hassan M.(2001) 'A NEW GENERAL METHOD FOR SUBSTITUTED 4-ALKYLTHIO-*N*-ARYLSULPHONYL-AMINO-2-PYRIDONES: REACTION OF KETENE-*SS*-ACETALS WITH ARYLSULPHONYLHYDRAZIDES', Phosphorus, Sulfur, and Silicon and the Related Elements, 170: 1, 171 — 179
To link to this Article: DOI: 10.1080/10426500108040593
URL: http://dx.doi.org/10.1080/10426500108040593

Phosphorus, Sulfur and Silicon, 2001, Vol. 170, pp. 171-179
Reprints available directly from the publisher
Photocopying permitted by license only

A NEW GENERAL METHOD FOR SUBSTITUTED 4-ALKYLTHIO-*N*-ARYLSULPHONYL-AMINO-2-PYRIDONES: REACTION OF KETENE-*SS*-ACETALS WITH ARYLSULPHONYLHYDRAZIDES

GALAL H. ELGEMEIE[a*], HOSNY A. ALI[b], AHMED H. ELGHANDOUR[b] and HASSAN M. ABDEL-AZIZ[b]

[a]Department of Chemistry, Faculty of Science, Helwan University, Helwan, Cairo, Egypt and [b]Department of Chemistry, Faculty of Science, Cairo University, (Bani Suef Branch), Bani Suef, Egypt

(Received June 31, 2000; In final form November 12, 2000)

A novel and efficient method for the synthesis of substituted 4-alkylthio-*N*-arylsulphonylamino-2-pyridones via the reaction of ketene-*SS*-acetals with *N*-cyanoacetoarylsulfonylhydrazides has been investigated. 1-Arylsulfonylamino-pyrazolo[3,4-*c*]pyridine-2(1*H*)-ones have also been prepared from the reaction of 4-alkylthio-*N*-arylsulfonylamino-2-pyridones with hydrazines.

During the course of our studies directed toward exploring the synthetic potential of ketene dithioacetals for synthesizing new classes of novel antimetabolites, [1-3] we have recently reported different successful approaches for synthesis of mercaptopurine and thioguanine analogues by the reaction of ketene dithioacetals with amino- and oxo-azoles.[4-6] The present work describes a new one-pot synthesis of 4-alkylthio-*N*-arylsulphonylamino-2-pyridones by the reaction of ketene dithioacetals with *N*-cyanoacetoarylsulfonylhydrazides. As far as we know, this is the first N-sulfonamide example to be reported for 2-pyridones. Thus, it has been found cyanoacetohydrazide **1** reacts with arylsulfonyl chloride **2** in pyridine to afford the corresponding *N*-cyanoacetoarylsulfonylhydrazides **3** in

* Corresponding Author

good yields. The structures of **3** were established and confirmed on the basis of their elemental analyses and spectral data. Compounds **3** reacted with both [bis(methylthio)methylene]malononitrile (**4a**) and ethyl 2-cyano-3,3-bis(methylthio)acrylate (**4b**) in dioxane containing a catalytic amount of potassium hydroxide to yield products for which the 4-alkylthio-*N*-arylsulphonylamino-2-pyridones **6** were assigned. The structures of **6** were established on the basis of their elemental analyses and spectral data (MS, IR, ^{1}H NMR). Thus, the mass spectrum of **6a** was compatible with the molecular formula $C_{14}H_{11}N_5O_3S_2$ (M^+ 361).

^{1}H NMR spectroscopy was used to confirm this structure for the product. Thus, ^{1}H NMR analysis revealed a singlet at $\delta = 2.51$ assigned to SCH_3 group, a multiplet at $\delta = 7.56$–7.81 assigned for aromatic protons, and a broad band at $\delta = 8.79$ assignable for an NH_2 group. The formation of **6** from **3** and **4** is assumed to proceed via addition of the active methylene group of **3** to the double bond of **4** to give intermediate Michael adducts. The latter lost elements of CH_3SH to yield the intermediate **5**, which cyclized to yield the novel 4-methylthio-*N*-arylsulphonylamino-2-pyridone derivatives **6**.

In order to explore the possibility that this reaction my occur with other classes of ketene dithiacetals, we investigated the reaction of cyanoacetoarylsulfonylhydrazides **3** with *N*-substituted bis(methylthiomethylene)cyanoacetamides **10**, the latters were prepared from the reaction of substituted acetanilide derivatives **9** with carbon disulfide in the presence of sodium ethoxide followed by the alkylation with methyl iodide. Thus, we treated **3** with one equivalent of dithioacetals **10** in dioxane containing a catalytic amount of potassium hydroxide for 24 h, and obtained the corresponding substituted 4-alkylthio-*N*-arylsulfonylamino-2-pyridone derivatives **11** in moderate yields. The structures of **11** were established on the basis of their elemental analyses and MS, IR and ^{1}H NMR data. Compounds **6** and **11** bearing latent functional substituents were found useful for further chemical transformations. Thus, it was found that compounds **6** and **11** reacted with hydrazine hydrate in refluxing ethanol containing a catalytic amount of piperdine to give the corresponding pyrazolo[3,4-*c*]pyridone derivatives **7** and **12**, respectively. Structures **7** and **12** were established and confirmed on the basis of their elemental analyses and spectral data. Thus, the mass spectrum of **7a** was compatible with the molecular formula $C_{13}H_{11}N_7O_3S$ (M^+ 345), and the ^{1}H NMR contained two broad singlets at $\delta = 8.23$ and 11.8 assignable for two NH groups.

Compounds **6**, when fused with aniline, led to the formation of the corresponding 4-anilino-*N*-arylsulphonylamino-2-pyridone derivatives **8**. The structures of **8** were established by mass spectroscopy, IR, and ^{1}H NMR data. The ^{1}H NMR spectrum for **8a** revealed a broad signal at $\delta = 8.6$ assignable to NH group.

In summary, we have achieved a regiospecific synthesis of interesting 4-alkythio-*N*-sulphonylaminated-2-pyridones by the reaction of certain ketene dithioacetals with *N*-cyanoacetylarylsulphonylhydrazides. The compounds obtained seem promising as high potential intermediates for synthesizing a variety of heterocycles.

EXPERIMENTAL

All melting points are uncorrected. The IR spectra were obtained (KBr disk) on a Perkin Elmer/1650 FT-IR instrument. The ^{1}H NMR spectra were measured on a Varian 400 MHz spectrometer for solutions in $(CD_3)_2SO$ using $Si(CH_3)_4$as an internal standard. Mass spectra were recorded on a Varian MAT 112 spectrometer. Analytical data were obtained from the Microanalytical Data Center at Cairo University

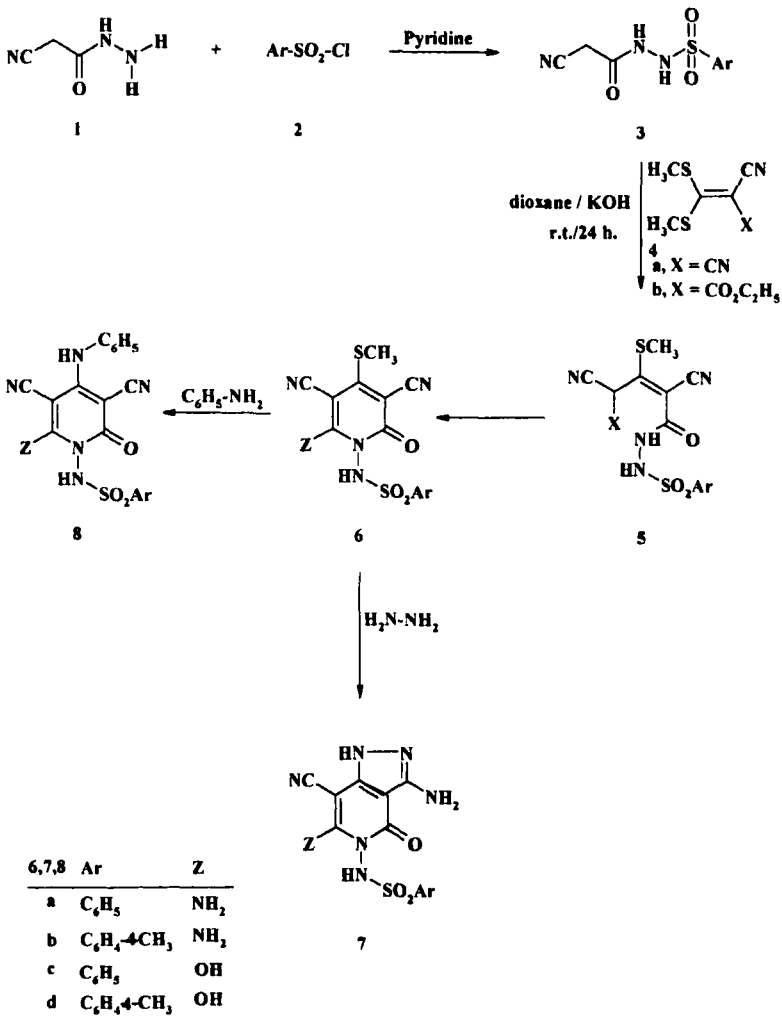
Arylsuphonylcyanoacetohydrazide 3a, b

A mixtur of cyanoacetohydrazide **1** (0.01 mol) and arenesulfonyl chloride (0.01 mol) in ethanol (30 ml) was stirred at room temprature for 24 h. The resulting solid product was collected by filtration and recrystallized from ethanol.

3a: (80 %), m.p. 170 °C; IR (KBr) 3407, 3284 (NH, NH), 2215 (CN), and 1672 cm^{-1} (CO); ^{1}H NMR δ 3.63 (s, 2H, CH_2), 7.56–7.86 (m, 5H, C_6H_5), 10.11 (s, br, 1H, NH) and 10.40 (s, br, 1H, NH); MS: M^+ 239. Calcd for $C_9H_9N_3O_3S$: C, 45.19; H, 3.77; N, 17.57%. Found: C, 45.0; H, 3.5; N, 17.3%. **3b**: (85 %), m.p. 180 °C; Calcd for $C_{10}H_{11}N_3O_3S$: C, 47.43; H, 4.35; N, 16.60%. Found: C, 47.0; H, 4.1; N, 16.3%.

N-Arylsulphonylamino-4-methylthio-2-pyridones 6. General procedure

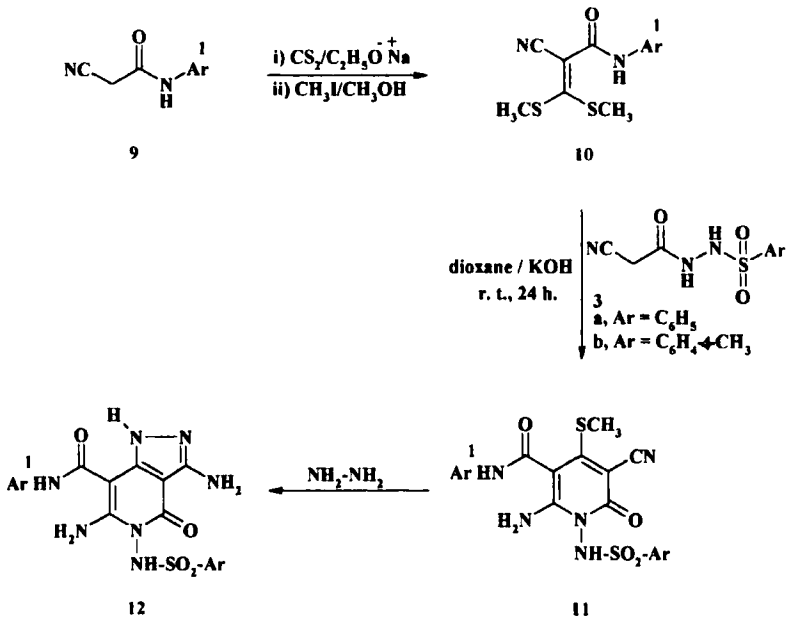
A mixture of [bis(methylthio)methylene]malononitrile (**4a**) or ethyl 2-cyano-3,3-(methylthio)acrylate (**4b**) (0.01 mol), *N*-cyanoacetoarylsulfo-



6,7,8	Ar	Z
a	C_6H_5	NH_2
b	C_6H_4-4-CH_3	NH_2
c	C_6H_5	OH
d	C_6H_4-4-CH_3	OH

CHART 1

nylhydrazides (**3a,b**) (0.01 mol), potassium hydroxide (0.12 mol) and dioxane (50 ml) was stirred at room temperature for 24 h. The resultant product was acidified with hydrochloric acid. The precipitate formed was collected by filtration, dried, and then crystallized from the appropriate solvent. **6a**: Orange, from MeOH, m.p. 250 °C, yield 75%; ν_{max}/cm^{-1}



9,10	Ar^1	11,12	Ar	Ar^1	11,12	Ar	Ar^1
a	C_6H_5	a	C_6H_5	C_6H_5	e	C_6H_5	C_6H_4-4-CH_3
b	C_6H_4-4-Cl	b	C_6H_4-4-CH_3	C_6H_5	f	C_6H_5	C_6H_4-4-Cl
c	C_6H_4-4-CH_3	c	C_6H_4-4-CH_3	C_6H_4-4-CH_3	g	C_6H_5	C_6H_4-4-OCH_3
d	C_6H_4-4-OCH_3	d	C_6H_4-4-CH_3	C_6H_4-4-Cl	h	C_6H_4-4-CH_3	C_6H_4-4-OCH_3

CHART 2

(KBr) 3570, 3382 (NH, NH_2), 2216 (CN), 1700 (CO). ^1H NMR [$(CD_3)_2SO$] δ 2.51 (s, 3H, SCH_3), 7.56–7.81 (m, 5H, C_6H_5), 8.79 (s, br, 2H, NH_2), 9.65 (s, br, 1H, NH); $C_{14}H_{11}N_5O_3S_2$ m/z 361; Calcd: C, 46.54; H, 3.05; N, 19.40%. Found: C, 46.3; H, 2.9; N, 19.2%. **6b**: Buff, from EtOH, m.p. 265 °C, yield 80%; ν_{max}/cm^{-1} (KBr) 3298, 3204 (NH, NH_2); 2212 (CN); 1695 (CO). ^1H NMR [$(CD_3)_2SO$] δ 2.41 (s, 3H, CH_3), 2.70 (s, 3H, SCH_3), 7.37–7.69 (m, 4H, C_6H_4), 8.78 (s, br, 2H, NH_2), 10.03 (s, br, 1H, NH). $C_{15}H_{13}N_5O_3S_2$ m/z 375. Calcd: C, 48.00; H, 3.47; N, 18.66%. Found: C, 47.7; H, 3.2; N, 18.5%. **6c**: Buff, from EtOH, m.p. 270 °C, yield

73%; ν_{max} /cm^{-1} (KBr) 3203–3000 (OH, NH), 2208 (CN), 1689 (CO), ^{1}H NMR [$(CD_3)_2SO$] δ 2.58 (s, 3H, SCH_3), 7.37–7.69 (m, 4H, C_6H_4), 9.22 (s, br, 1H, OH), 10.03 (s, br, 1H, NH). $C_{14}H_{10}N_4O_4S_2$ Calcd: C, 46.41; H, 2.76; N, 15.47%. Found: C, 46.2; H, 2.6; N, 15.7%. **6d**: Buff, from EtOH, m.p. 230 °C, yield 70%; ν_{max} / cm^{-1} (KBr) 3520 (NH), 2211 (CN), 1697 (CO), $C_{15}H_{12}N_4O_4S_2$ Calcd: C, 47.87; H, 3.19; N, 14.89%. Found: C, 47.6; H, 3.0; N, 14.6%.

N-Arylsulphonylamino-4-methylthio-2-pyridones 11a-h. General procedure

A mixture of N-substituted bis(methylthiomethylene)(cyano)acetamide derivatives **10a-d** (0.01 mol) and *N*-cyanoacetoarylsulfonylhydrazides **3a,b** (0.01 mol) was refluxed in DMF (50 ml) containing potassium carbonate (0.05 mol) for 5 h. The reaction mixture was then cooled, poured over an ice-water mixture, and neutralized with dil. HCl, the performed product was collected by filtration and recrystallized from the appropriate solvent. **11a**: Brown, from DMF, m.p. >300 °C, yield 70%; ν_{max} /cm^{-1} (KBr) 3570, 3382 (NH, NH_2), 2216 (CN), 1689 (CO). ^{1}H NMR [$(CD_3)_2SO$] δ 2.29 (s, 3H, SCH_3), 7.15–7.50 (m, 10H, $2C_6H_5$), 7.97 (s, br, 2H, NH_2), 9.62 (s, br, 1H, NH), 10.46 (s, br, 1H, NH). $C_{20}H_{17}N_5O_4S_2$ m/z 455, Calcd: C, 52.75; H, 3.74; N, 15.38%. Found: C, 52.3; H, 3.7; N, 15.4%. **11b**: Brown, from DMF-EtOH, m.p. >300 °C, yield 75%; ν_{max}/cm^{-1} (KBr) 3520, 3154 (NH, NH_2), 2211 (CN), 1672 (CO), ^{1}H NMR [$(CD_3)_2SO$] δ 2.41 (s, 3H, CH_3), 2.76 (s, 3H, SCH_3), 7.23–7.70 (m, 9H, C_6H_5 and C_6H_4), 8.69 (s, br, 2H, NH_2), 11.91 (s, br, 1H, NH). $C_{21}H_{19}N_5O_4S_2$ Calcd: C, 53.73; H, 4.05; N, 14.93%. Found: C, 53.3; H, 3.8; N, 15.1%. **11c**: Brown, from DMF-EtOH, m.p. >300 °C, yield 75%; ν_{max} /cm^{-1} (KBr) 3269, 3102 (NH, NH_2), 2259 (CN), 1669 (CO). ^{1}H NMR [$(CD_3)_2SO$] δ 2.38 (s, 3H, CH_3), 2.49 (s, 3H, CH_3), 2.66 (s, 3H, SCH_3), 7.23–7.70 (m, 8H, $2C_6H_4$), 8.36 (s, br, 2H, NH_2), 10.88 (s, br, 1H, NH). $C_{22}H_{21}N_5O_4S_2$ Calcd: C, 54.66; H, 4.35; N, 14.49%. Found: C, 54.4; H, 4.1; N, 14.2%. **11d**: Brown, from DMF-EtOH, m.p. >300 °C, yield 75%; ν_{max}/cm^{-1} (KBr) 3448, 3122 (NH, NH_2), 2207 (CN), 1662 (CO). $C_{21}H_{18}ClN_5O_4S_2$ Calcd: C, 50.05; H, 3.57; N, 13.90%. Found: C, 50.0; H, 3.3; N, 13.6%. **11e**: Brown, from DMF-EtOH, m.p. >300 °C, yield 75%; ν_{max} /cm^{-1}(KBr) 3306, 3068 (NH, NH_2), 2177 (CN), 1680 (CO). $C_{21}H_{19}N_5O_4S_2$ Calcd: C, 53.73; H, 4.05; N, 14.93%. Found: C,

53.4; H, 3.8; N, 14.6%. **11f**: Brown, from DMF-EtOH, m.p. >300 °C, yield 75%; ν_{max} /cm^{-1} (KBr) 3335, 3158 (NH, NH_2), 2174 (CN), 1687 (CO). $C_{20}H_{16}ClN_5O_4S_2$ Calcd: C, 49.03; H, 3.27; N, 14.30%. Found: C, 49.3; H, 3.1; N, 14.2%. **11g**: Brown, from DMF-EtOH, m.p. >300 °C, yield 75%; ν_{max}/cm^{-1} (KBr) 9239, 3291 (NH, NH_2), 2202 (CN), 1621 (CO). $C_{21}H_{19}N_5O_5S_2$ Calcd: C, 51.96 H, 3.92; N, 14.43%. Found: C, 51.7; H, 3.7; N, 14.2%. **11h**: Brown, from DMF-EtOH, m.p. >300 °C, yield 75% ν_{max}/cm^{-1} (KBr) 3336, 3157 (NH, NH_2), 2179 (CN), 1690 (CO). ^{1}H NMR [$(CD_3)_2SO$] δ 2.44 (s, 3H, CH_3), 2.70 (s, 3H, SCH_3), 2.88 (s, 3H, OCH_3), 7.11–7.68 (m, 8H, $2C_6H_4$), 8.12 (s, br, 2H, NH_2), 10.88 (s, br, 1H, NH). $C_{22}H_{21}N_5O_5S_2$ Calcd: C, 52.91; H, 4.21; N, 14.03%. Found: C, 52.6; H, 4.0; N, 13.8%.

1-Arylsulfonylamino-3-cyano-pyrazolo[3,4-c]pyridine-2-(1H)-ones 7a-d. General procedure

A mixture of equivalent amounts of **6a-d** (0.01 mol) and hydrazine hydrate (0.01 mol) were heated in ethanol (30 ml) for 4 h. The solid product formed was collected by filtration and recrystallized from the appropriate solvent. **7a**: Buff, from EtOH-DMF, m.p. >300 °C, yield 75%; ν_{max} /$^{-1}$(KBr) 3450, 3350 (NH, NH_2), 2205 (CN), 1698 (CO). ^{1}H NMR [$(CD_3)_2SO$] δ 5.8 (s, 2H, NH_2), 7.38–7.65 (m, 5H, C_6H_5), 8.23 (s, 2H, NH_2), 9.11 (s, br, 1H, NH), 11.8 (s, br, 1H, NH). $C_{13}H_{11}N_7O_3S$ Calcd: C, 45.22; H, 3.19; N, 28.41%. Found: C, 44.8; H, 3.5; N, 28.1%. **7b**: Buff, from EtOH-DMF, m.p. >300 °C, yield 75%; ν_{max} /cm^{-1} (KBr) 3573, 3416 (NH, NH2), 2217 (CN), 1700 (CO), ^{1}H NMR [$(CD_3)_2SO$] δ 2.20 (s, 3H, CH_3), 5.61 (s, 2H, NH_2), 7.51–7.65 (m, 4H, C_6H_4), 8.23 (s, br, 2H, NH_2), 11.85 (s, br, 1H, NH), 12.25 (s, br, 1H, NH) $C_{14}H_{13}N_7O_3S$ m/z 359, Calcd: C, 46.80; H, 3.62; N, 27.30%. Found: C, 46.4; H, 3.7; N, 27.0%. **7c**: Buff, from EtOH-DMF, m.p. >300 °C, yield 80%; ν_{max} /cm^{-1}(KBr) 3749, 3182 (NH, NH_2), 2206 (CN), 1695 (CO). $C_{13}H_{10}N_6O_4S$, Calcd: C, 45.08; H, 2.89; N, 24.28%. Found: C, 44.8; H, 3.0; N, 24.0%. **7d**: Buff, from EtOH, m.p. >300 °C, yield 77%; ν_{max}/cm^{-1}(KBr) 3451, 3352 (NH, NH_2), 2205 (CN), 1794 (CO). ^{1}H NMR [$(CD_3)_2SO$] δ 5.3 (s, 2H, NH_2), 7.22–7.98 (m, 5H, C_6H_5), 8.55 (s, 1H, OH), 10.37 (s, br, 1H, NH), 11.35 (s, br, 1H, NH) $C_{14}H_{12}N_6O_4S$, Calcd: C, 46.66; H, 3.33; N, 23.33%. Found: C, 46.5; H, 3.3; N, 23.0%.

1-Arylsulfonylamino-pyrazolo[3,4-c]pyridine-2-(1H)-ones 12-a-h. General procedure

A mixture of equivalent amounts of **11a-h** (0.01 mol) and hydrazine hydrate (0.01 mol) were heated in DMF (30 ml) containing a catalytic amount of piperdine for 4 h. The solid product formed was collected by filteration and recrystallized from the appropriate solvent. **12a**: Brown, from DMF, m.p. >300 °C, yield 70%; ν_{max} /cm^{-1}(KBr) 3425, 3285 (NH, NH_2), 1671 (CO). $C_{19}H_{17}N_7O_4S$, Calcd: C, 51.94; H, 3.87; N, 22.32%. Found: C, 51.6; H, 3.8; N, 21.9%. **12b**: Brown, from DMF-EtOH, m.p. >300 °C, yield 75%; ν_{max}/cm^{-1} (KBr) 3430, 3298 (NH, NH_2), 1652 (CO). $C_{20}H_{19}N_7O_4S$ (453), Calcd: C, 53.00; H, 4.19; N, 21.63%. Found: C, 52.6; H, 4.1; N, 21.4%. **12c**: Brown, from DMF-EtOH, m.p. >300 °C, yield 75%; ν_{max}/cm^{-1} (KBr) 3418, 3284 (NH, NH_2), 1655 (CO). $C_{21}H_{21}N_7O_4S$, Calcd: C, 53.96; H, 4.50; N, 20.99%. Found: C, 53.6; H, 4.5; N, 20.7%. **12d**: Brown, from DMF-EtOH, m.p. >300 °C, yield 75%; ν_{max}/cm^{-1} (KBr) 3418, 3270 (NH, NH_2), 1660 (CO). $C_{20}H_{18}ClN_7O_4S$ Calcd: C, 49.23; H, 3.69; N, 20.10%. Found: C, 49.6; H, 4.0; N, 20.3%. **12e**: Brown, from DMF-EtOH, m.p. >300 °C, yield 75%; ν_{max} /cm^{-1} (KBr) 3470, 3229 (NH, NH_2), 1681 (CO). $C_{20}H_{19}N_7O_4S$, Calcd: C, 52.98; H, 4.19; N, 21.63%. Found: C, 52.8; H, 3.6; N, 22.1 %. **12f**: Brown, from DMF, m.p. >300 °C, yield 75%; ν_{max} /cm^{-1} (KBr) 3337, 3212 (NH, NH_2), 1689 (CO). $C_{19}H_{16}ClN_7O_4S$ Calcd: C, 48.21; H, 3.43; N, 20.72%. Found: C, 47.9; H, 3.3; N, 20.5%. **12g**: Brown, from DMF, m.p. >300 °C, yield 75%; ν_{max} /cm^{-1} (KBr) 3418, 3166 (NH, NH_2), 1623 (CO). $C_{20}H_{19}N_7O_5S$ Calcd: C, 51.17; H, 4.08; N, 20.90%. Found: C, 51.0; H, 4.0; N, 20.7%. **12h**: Brown, from DMF, m.p. >300 °C, yield 75%; ν_{max} /cm^{-1} (KBr) 3336, 3198 (NH, NH_2), 1699 (CO). $C_{21}H_{21}N_7O_5S$ Calcd: C, 52.17; H, 4.35; N, 20.29%. Found: C, 53.5; H, 4.6; N, 20.7%.

N-Arylsulphonylamino-4-anilino-2-pyridones 8. General procedure

A mixture of **6a-d** (0.01 mol) and pure aniline (0.01 mol) was heated for one hour in an oil bath (150 C). The reaction mixture was diluted with ethanol. The resulting solid product was filtered off and crystallized from the appropriate solvent. **8a**: Brown, from EtOH-DMF, m.p. >300 °C, yield 72%; ν_{max}/cm^{-1} (KBr) 3186, 2924 (NH, NH_2), 2207 (CN), 1694 (CO). ^{1}H

NMR [$(CD_3)_2SO$): 7.00–7.65 (m, 10H, 2 C_6H_5), 8.48 (s, br, 2H, NH_2), 8.60 (s, br, 1H, NH), 9.56 (s, br, 1H, NH). $C_{19}H_{14}N_6O_3S$ Calcd: C, 56.16; H, 3.45; N, 20.69%. Found: C, 56.7; H, 3.1; N, 20.5%. **8b**: Buff, from EtOH-DMF, m.p. >300 °C, yield 78%; ν_{max} /cm^{-1} (KBr) 3453, 3287 (NH, NH_2), 2209 (CN), 1671 (CO). $C_{20}H_{16}N_6O_3S$ m/z 420, Calcd: C, 57.14; H, 3.81; N, 20.00%. Found: C, 56.9; H, 3.5; N, 19.7%. **8c**: Yellow, from EtOH-DMF, m.p. >300 °C, yield 68%; ν_{max} /cm^{-1}(KBr) 3315 (NH), 2200 (CN), 1655 (CO). $C_{19}H_{13}N_5O_4S$ Calcd: C, 56.02; H, 3.19; N, 17.22%. Found: C, 56.5; H, 3.5; N, 16.8%. **8d**: Buff, from EtOH-DMF, m.p. >300 °C, yield 65%; ν_{max} /cm^{-1}(KBr) 3311 (NH), 2216 (CN), 1651 (CO). $C_{20}H_{15}N_5O_4S$ Calcd: C, 57.01; H, 3.56; N, 16.63%. Found: C, 56.8; H, 3.4; N, 16.4%.

References

1. G. E. H. Elgemeie, A. H. Elghandour, A. M. Elzanate and S. A. Ahmed, *J. Chem. Soc. Perkin Trans. 1*, 3285 (1997).
2. G. E. H. Elgemeie, A. H. Elghandour, A. M. Elzanate, and S. A. Ahmed, *J. Chem. Research (S)*, 162 (1998).
3. G. E. H. Elgemeie, A. H. Elghandour, A. M. Elzanate, and W.A. Masoud, *J. Chem. Research (S)*, 164 (1998).
4. G. E. H. Elgemeie, H. A. Ali, and A. M. Elzanaty, *J. Chem. Research (S)*, 340 (1996).
5. G. E. H. Elgemeie, A. H. Elghandour, A. M. Elzanaty and A. A. Hussein, *J. Chem. Research (S)*, 260 (1997).
6. G. E. H. Elgemeie, S. R. El-Ezbawy, H. A. Ali, and A.K. Mansour, *Bull. Chem. Soc. Jpn.*, *67*, 738 (1994).