
SYNTHESIS OF NOVEL PYRAZOLE, PYRIDAZINE AND PYRIMIDINE DERIVATIVES CONTAINING SULFONAMIDO MOIETY

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Abstract

Reaction of the hydrazone **2** with hydrazine hydrate, phenylhydrazine, benzoylhydrazine and/or semicarbazide in dioxane afforded the 3,5-diaminopyrazole derivatives **3-6**, respectively. Refluxing **3** with acetic anhydride and/or benzoylchloride, furnished the N-acyl derivatives **7** and **8**. On the other hand, fusion of **3** with different aromatic aldehydes produced the Schiff bases **9a-c**, respectively. Treatment of **2** with urea, thiourea and/or guanidine hydrochloride furnished the pyrimidine and 1,3-thiazine derivatives **10-12**, respectively. Reaction of **2** with ethyl cyanoacetate, ethyl acetoacetate, malononitrile and/or piperidine yielded the pyridazine derivatives **14** and **15**, pyridopyridazine **17** and the enamionitrile **18**. Treatment of **18** with CS₂ and/or phenylisothiocyanate produced 1,3-thiazine **20**, dithioxopyrimidine **21** and the thiopyrimidine derivatives **22**, respectively.

Key words:

Sulfonamide, hydrazones, aminopyrazoles, pyrimidines and pyridazines.

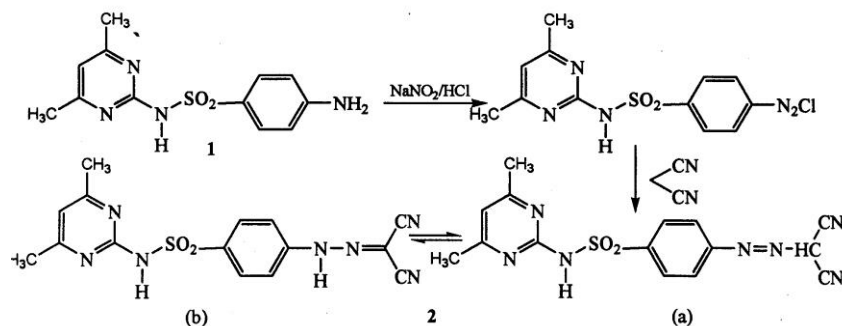
Introduction

It is well known that sulfonamides have a variety of biological activities such as antibacterial¹, carbonic anhydrase inhibitory², and antitumor activities³. Also, pyrazoles, pyrimidines and pyridazines possess biological and pharmacological activities⁴⁻¹¹. Because of these interesting activities and in continuation of our work¹²⁻¹⁶ on the synthesis of novel heterocyclic systems exhibiting biological activity, we undertook the synthesis of a new series of compounds incorporating the above mentioned biologically active moieties in one molecule.

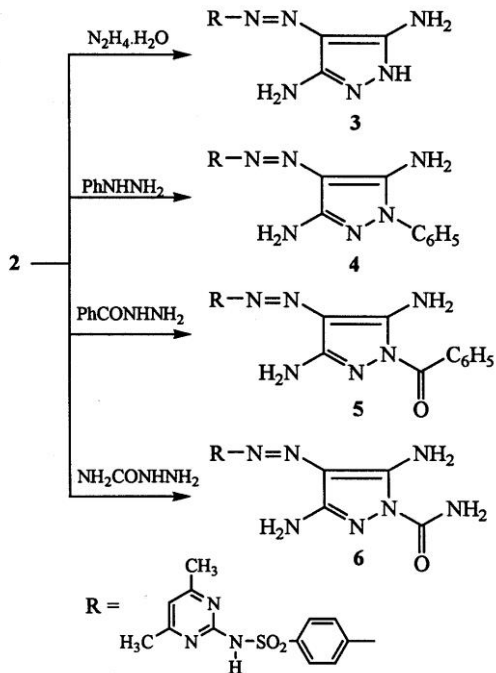
Results and Discussion

The hydrazone **2** was synthesized by diazotization of sulfamethazine, [N¹-(4,6-dimethyl-2-pyrimidinyl)sulfanilamide], **1** followed by coupling with malononitrile in the presence of sodium acetate at room temperature (Scheme 1). The spectral data revealed that this compound exists in the hydrazone form (b), where, their spectrum displayed a strong band at 2227 cm⁻¹ corresponding to C=N groups. ¹H-NMR spectrum showed two signals at 6.73 and 8.74 (exchangeable) corresponding to two

NH groups and the MS indicated the molecular ion peak at 355, which is in accordance with the molecular formula.



It was reported that β -enaminonitriles reacted with hydrazines producing pyrazole derivatives¹⁷. Thus, refluxing **2** with hydrazine hydrate in ethanol, phenylhydrazine, benzylhydrazine and semicarbazide in dioxane afforded the 3,5-diaminopyrazole derivatives **3-6** respectively (Scheme 2).

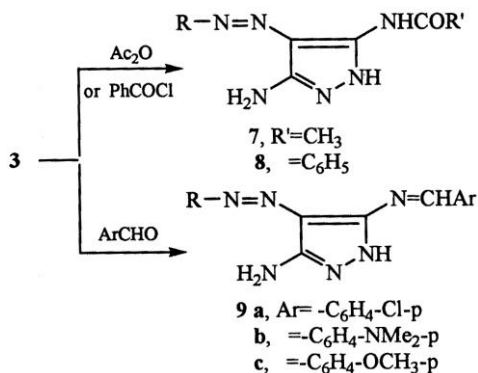


Scheme 2

The structure of compounds **3-6** were elucidated from their elemental analysis and spectral data. (cf experimental part). The $^1\text{H-NMR}$ spectra of **3** showed two broad peaks (exchangeable) at 6.05 and 6.53 corresponding to two NH_2 groups. The presence of a multiplet peak at δ 8.30-8.80 (10 protons) indicated the presence of phenyl group in compound **4**.

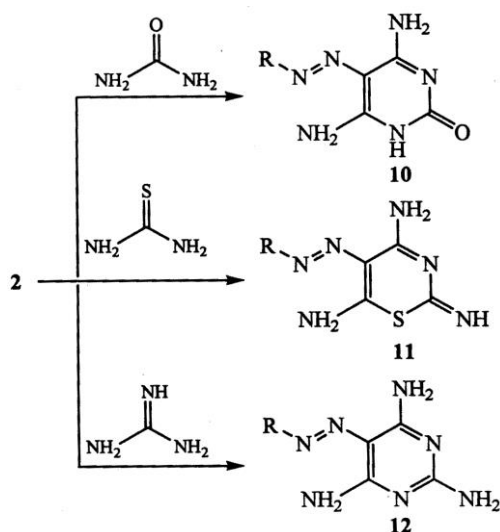
Reaction of **3** with acetic anhydride and/or benzoylchloride under reflux, furnished the N-acyl derivatives **7** and **8**. The structures of **7** and **8** were established on the basis of their spectral data, where their spectra showed bands at 1679 and 1672 cm^{-1} corresponding to the carbonyl groups, furthermore, $^1\text{H-NMR}$ spectrum of **7** showed a band at 2.43 corresponding to COCH_3 , while that of **8** exhibited a multiplet at 7.48-7.61 for the phenyl group (Scheme 3).

On the other hand, fusion of **3** with different aromatic aldehydes, in the presence of fused sodium acetate, produced the Schiff bases **9a-c** respectively. (Scheme 3).



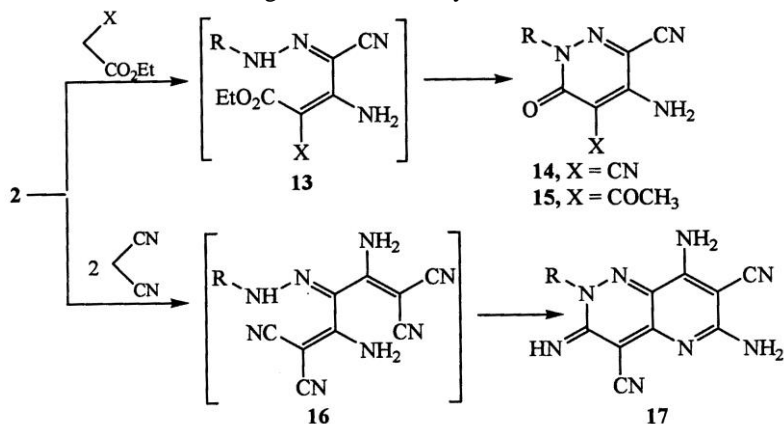
Scheme 3

Treatment of **2** with urea, thiourea, and guanidine hydrochloride, in ethanol-ethoxide, produces the 4,6-diaminopyrimidin-2-one **10**, 2-imino-4,6-diaminothiazine **11** and triaminopyrimidine derivatives **12**¹⁸, respectively (Scheme 4). The formation of these products is assumed to proceed through the cycloaddition of NH_2 or SH groups to the cyano functions of **2**. The spectral data of **10**, **11** and **12** confirmed the proposed structures, where, the spectra showed bands at 3444, 3382, 3229 and 1660 cm^{-1} , corresponding to NH_2 , NH and CO groups and the MS of **10** exhibited molecular ion peak at 415 which is in agreement with the molecular formula.



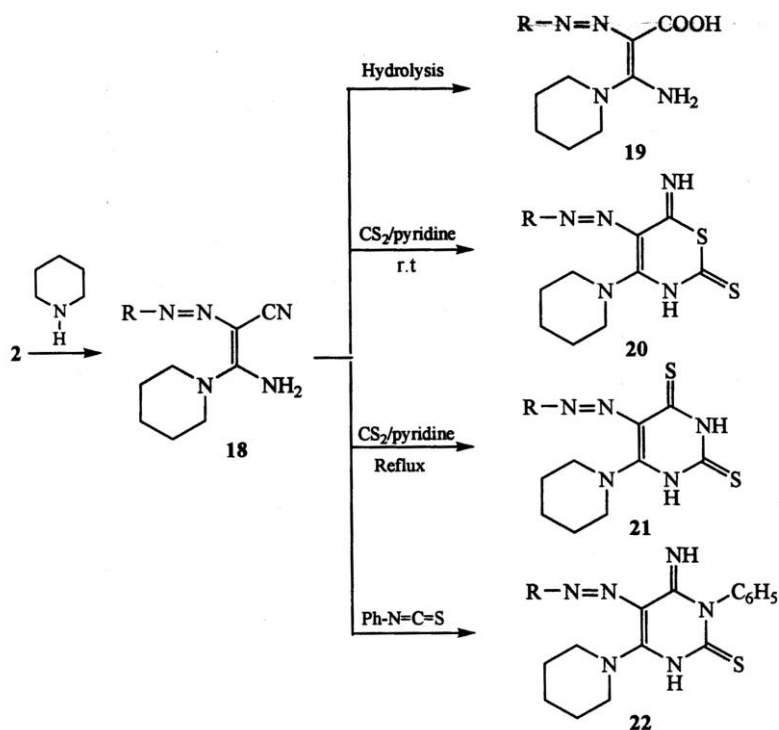
Scheme 4

The reactivity of **2** towards some active methylene reagents was studied. Thus, compound **2** reacted with ethyl cyanoacetate and/ or ethyl acetoacetate in refluxing ethanol containing triethylamine to yield the pyridazine derivatives **14** and **15** via the intermediate **13**. The structures of **14** and **15** were approved by their ir and ^1NMR spectra, where, the ^1NMR of **15** indicated the presence of a peak at δ 2.3 corresponding to COCH_3 protons. On the other hand, the reaction of **2** with two moles of malononitrile in refluxing ethanol/ Et_3N solution afforded the pyridopyridazine derivative **17**^{19,20} through the intermediate **16** (Scheme 4). The structure was elucidated through elemental analysis and MS.



Scheme 5

Refluxing **2** with piperidine in ethanol produced the acyclic enaminnitrile **18**. The $^1\text{H-NMR}$ spectrum displayed the characteristic peaks of the piperidine ring at 1.62 (m, 6H) and 3.49 (m, 4H). Furthermore, compound **18** underwent acidic hydrolysis to produce the corresponding enamino-carboxylic acid derivative **19**. Also, the behaviour of **18** towards carbon disulfide at different conditions was studied. Thus, reaction of **18** with carbon disulfide in pyridine at room temperature yielded 1,3-thiazine **20**, while, upon reflux dithioxopyrimidine **21** were formed. Finally, the pyrimidine derivative **22** was obtained when **18** was fused with phenylisothiocyanate, in an oil bath, at 140°C (Scheme 6). The structures of **19-22** were elucidated from their elemental analysis, MS and $^1\text{H-NMR}$ spectra.



Scheme 6

Experimental:

Melting points are uncorrected. The infrared absorption spectra were determined with a Pye Unicam SP 2000 infrared spectrophotometer with KBr Wafer Technique. The mass spectra were run on Shimadzu GC-NS-QP 1000 EX instrument operating at 70 eV. The $^1\text{H-NMR}$ spectra were determined on a Varian Gemini 200 ^1NMR

spectrophotometer using DMSO- d_6 as solvent, (chemical shift in δ , ppm) and TMS as internal standard. Elemental analyses were carried out by the Microanalytical Research Center, Faculty of Science, Cairo University, Egypt [satisfactory analytical data (± 0.3) were obtained for all compounds].

3,5-Diamino-4-[4-N-(4,6-dimethylpyrimidin-2-yl)sulfamoyl]phenylazomalononitrile (2).

Sulfonamide 1 (10 mmol, 2.78 g) were dissolved in a mixture of concentrated HCl (5 mL) and water (5 mL) and cooled to 5-10°C in an ice bath. A cold aqueous solution of sodium nitrite (10 mmol, 5 mL) was then added with stirring. The diazonium salt so obtained was filtered into a cold mixture of sodium acetate (4 g) and malononitrile (10 mmol, 0.66 g) in ethanol (25 mL) with stirring for one h. The resulting solid was washed with water (150 mL) and recrystallized from ethanol to afford **2** (Table 1). IR: 3243, 3192 (NH), 3001 (CHar), 2889 (CH_{al}), 2227 (C=N), 1623 (C=N), 1600 (C=C), 1543 (N=N), 1327, 1150 cm^{-1} (SO₂). ¹H-NMR (DMSO- d_6): 2.25 (s, 6H, 2 CH₃), 6.73 (s, 1H, NH exchangeable), 7.57-8.05 (m, 5H, ar-H + 1H pyrimidine ring), 8.74 (s, 1H, NH, exchangeable). MS (%): molecular ion peak at 355 (3.5) and M+ 1 at 356 (3.7).

3,5-Diamino-4-[4-N-(4,6-dimethylpyrimidin-2-yl)sulfamoyl]phenylazopyrazole (3).

To a solution of **2** (4 mmol, 1.42 g) in ethanol (50 mL), hydrazine hydrate (5 mmol, 0.25 mL) was added and the reaction mixture was refluxed for 2 h. The solid product which formed on heating was collected and recrystallized from the proper solvent to give **3** (Table 1). IR: 3474, 3370, 3203 (NH₂, NH), 1621 (C=N), 1561 cm^{-1} (N=N). ¹H-NMR (DMSO- d_6): 2.27 (s, 6H, 2 CH₃), 6.05 and 6.53 (br., 4H, 2 NH₂, exchangeable), 6.87 (s, 1H, NH exchangeable), 7.76-8.06 (m, 5H, ar-H + 1H pyrimidine ring). MS (%): 389 (3.1) M+2, 388 (2.5) M+1, 387 (1.8) M⁺.

3,5-Diamino-1-phenyl-4-[4-N-(4,6-dimethylpyrimidin-2-yl)sulfamoyl]phenylazopyrazole (4).

A mixture of **2** (4 mmol, 1.42 g) in dioxane (30 mL) and phenylhydrazine (4 mmol, 0.43 mL) was refluxed for 3 h., concentrated and left to cool at room temperature. The solid deposited was filtered off, dried and crystallized from proper solvent to produce **4** (Table 2). IR: 3395, 3276, 3205, 3163 (NH₂, NH), 1622 (C=N), 1558 cm^{-1} (N=N). ¹H-NMR (DMSO- d_6): 3.06 (s, 6H, 2 CH₃), 4.11 and 4.36 (s., 4H, 2 NH₂, exchangeable), 7.56 (s, 1H, NH exchangeable), 8.30-8.80 (m, 10H, ar H + 1H pyrimidine ring).

3,5-Diamino-1-benzoyl-4-[4-N-(4,6-dimethyl)pyrimidin-2-yl)sulfamoyl]phenylazopyrazole (5).

To 40 mL of dioxane containing the hydrazone **2** (4 mmol, 1.42 g), benzoylhydrazine (4 mmol, 0.54 g) was added and the mixture was refluxed for 6 h., concentrated and left to cool at room temperature. The solid product so formed was filtered off, dried and crystallized from proper solvent to yield **5** (Table 1). IR: 3443, 3324, 3210, (NH₂, NH), 1687 (C=O), 1604 (C=N), 1541 cm⁻¹ (N=N). ¹H-NMR (DMSO-d₆): 2.28 (s, 6H, 2 CH₃), 6.78 and 7.07 (s, 4H, 2 NH₂, exchangeable), 7.58-8.19 (m, 10H, ar-H + 1H pyrimidine ring), 10.52 (br, 1H, NH exchangeable).

3,5-Diamino-1-benzoyl-4-[4-N-(4,6-dimethylpyrimidin-2-yl)sulfamoyl]phenylazopyrazole (6).

A solution of hydrazone **2** (4 mmol, 1.42 g), semicarbazide.HCl (4 mmol, 0.45 g and triethylamine (0.5 mL) in dioxane (40 mL) was heated under reflux for 12 h. The solid product which precipitated on heating was collected and crystallized from proper solvent to give **6** (Table 2). IR: 3404, 3302, 3207, (NH₂, NH), 1680 (C=O), 1608 cm⁻¹ (C=N).

3-amino-5-acetylamino-4-[4-N-(4,6-dimethylpyrimidin-2-yl)sulfamoyl]phenylazopyrazole (7) and 3-amino-5-benzoylamino-4-[4-N-(4,6-dimethylpyrimidin-2-yl)sulfamoyl]phenylazopyrazole (8).

A mixture of pyrazole **3** (5 mmol, 1.93 g) and acetic anhydride and/or benzoyl chloride (15mL) was refluxed for 1 h. The solid product which precipitated after cooling was filtered off, washed with water (100 mL), dried and recrystallized from proper solvent to produce **7** and **8**

(Table 1). Compound **7**, IR: 3379, 3269, 3208, 3177 (NH₂, NH), 1679 (C=O), 1620 (C=N), 1603 (C=C), 1563 cm⁻¹ (N=N). ¹H-NMR (DMSO-d₆): 2.26 (s, 6H, 2 CH₃), 2.43 (s, 3H, COCH₃), 3.57 (s, 2H, NH₂, exchangeable), 6.34, 6.77 (s, 2H, 2 NH exchangeable), 7.74-8.04 (m, 5H, ar-H + 1H pyrimidine ring), 8.35 (br, 1H, NH exchangeable).

Compound **8**, IR: 3437, 3289, 3212, 3172 (NH₂, NH), 1672 (C=O), 1627 (C=N), 1595 (C=C), 1562 cm⁻¹ (N=N). ¹H-NMR (DMSO-d₆): 2.25 (s, 6H, 2 CH₃), 3.57 (s, 2H, NH₂, exchangeable), 6.37, 6.77 (s, 2H, 2 NH exchangeable), 7.48-8.14 (m, 10H, ar-H + 1H pyrimidine ring), 8.58 (br., 1H, NH exchangeable).

3-Amino-5-[4-substitutedbenzal) amino-4-[4-N-(4,6-dimethylpyrimidin-2-yl)sulfamoyl Jhenylazopyrazole (9a-c)

Fused sodium acetate (2 g) was added to a mixture of **3** (5 mmol, 1.93 g) and different aromatic aldehydes (4 mmol) namely, 4-chlorobenzaldehyde, 4-N,N-dimethylaminobenzaldehyde and/or 4-methoxybenzaldehyde. The reaction mixture was grinded and heated in a sand bath at 200-210°C for 3h. The solid product so obtained was washed with water, triturated with dioxane, filtered off, dried and crystallized from the proper solvent to furnish 9a-c, respectively (Table 1). IR (9a): 3423,3374,3214 (NH₂, NH), 1622 (C=N), 1590 (C=C), 1540 cm⁻¹ (N=N). ¹H-NMR (9a) (DMSO-d₆): 2.11 (s, 6H, 2 CH₃), 6.28 (s, 2H, NH₂ exchangeable), 7.33 (br, 1H, NH exchangeable), 7.618.04 (m, 10H, ar-H + 1H pyrimidine + N=CH), 9.26 (s., 1H, NH exchangeable).

4,6-Diamino-5-[4-N-(4,6-dimethylpyrimidin-2-yl) sulfamoyl]phenylazopyrimidin-2(1H)-one(10),4,6-Diamino-2-imino-5-[4-N-(4,6-dimethylpyrimidin-2-yl)sulfamoyl]phenylazo-1,3-thiazine (11) and 2,4,6-Triamino-5-[4-N-(4,6-dimethylpyrimidin-2-yl)sulfamoyl]phenylazopyrimidine (12).

To a mixture of **2** (4 mmol, 1.42 g) and the proper amino compound (6 mmol) (urea 0.36 g, thiourea 0.46 g, or guanidine 0.58 g) in absolute ethanol (40 mL), sodium ethoxide (0.23 g of Na in 10 mL ethanol) was added. The reaction mixture was refluxed for **6-8 h**, concentrated and cooled. The separated solid was filtered off, washed with water and crystallized from the proper solvent to afford compounds **10-12** (Table 1).

Compound (10), IR: 3437, 3372, 3224, 3154 (NH₂, NH), 1670 (CO), 1628 (C=N), 1598 (C=C), 1548 (N=N). ¹H-NMR (DMSO-d₆): 2.27 (s, 6H, 2 CH₃), 6.77 (s, 2H, NH₂ exchangeable), 7.40 (br, 2H, NH₂ exchangeable), 7.56-8.19 (m, 4H, ar-H + 1H pyrimidine), 8.40, 8.84 (br, 2H, 2NH exchangeable) .

Compound (11), IR: 3444, 3382, 3228 (NH₂, NH), 1631 (C=N), 1603 (C=C), 1543 (N=N). ¹HNMR (DMSO-d₆): 2.27 (s, 6H, 2 CH₃), 6.78 (s, 2H, NH₂ exchangeable), 7.72 (br, 2H, NH₂ exchangeable), 7.92-8.15 (m, 4H, ar-H + 1H pyrimidine), 8.70, 11.95 (br, 2H, 2NH exchangeable). Compound (12), IR: 3433, 3354, 3189 (NH₂, NH), 1621 (C=N), 1599 (C=C), 1553 cm⁻¹ (N=N). ¹HNMR (DMSO-d₆): 2.29 (s, 6H, 2 CH₃), 6.61, 6.79 and 7.47 (br, 6H, 3NH₂ exchangeable), 7.63-8.24 (m, 4H, ar-H + 1H pyrimidine), 8.83 (br., H, NH exchangeable).

4-Amino-3,5-dicyano-1-[4-N-(4,6-dimethylpyrimidin-2-yl)sulfamoyl]phenylazopyridazin-6-one (14), 5-Acetyl-4-amino-3-cyano-1-[4-N-(4,6-dimethylpyrimidin-2-yl)sulfamoyl]phenylazopyridazin-6-one(15) and 6,8-diamino-4,7-dicyano-3-imino-2-[4-N-(4,6-dimethylpyrimidin-2-yl)-sulfamoyl]phenylazopyrido[3,2-c]pyridazine (17).

A solution of **2** (4 mmol, 1.42 g) and the proper active methylene compound (ethyl cyanoacetate 4 mmol, 0.4 mL, ethyl acetoacetate 4 mmol, 0.5 mL and/or malononitrile 8 mmol, 0.53 g) in ethanol (50 mL) containing a catalytic amount of Et₃N (5 drops) was heated under reflux for **7-10** h. The solid product so obtained on cooling was collected by filtration and crystallized from the appropriate solvent to give compounds **14**, **15** and **17**, respectively (Table 1).

Compound (14), IR: 3433, 3371, 3232, 3196 (NH₂, NH), 2224 (C=N), 1671 (CO), 1621 (C=N), 1602 (C=C), 1549 cm⁻¹ (N=N). ¹H-NMR (DMSO-d₆): 2.26 (s, 6H, 2 CH₃), 6.76 (s, 2H, NH₂ exchangeable), 7.58-8.19 (m, 4H, ar-H + ¹H pyrimidine), 8.67 (br, H, NH exchangeable).

Compound (15), IR: 3448, 3381, 3275, 3106 (NH₂, NH), 2230 (C=N), 1677, 1661 (2CO), 1628 (C=N), 1595 (C=C), 1547 cm⁻¹ (N=N). ¹H-NMR (DMSO-d₆): 2.27 (s, 6H, 2 CH₃), 2.64 (s, 3H, COCH₃), 6.68 (s, 2H, NH₂ exchangeable), 7.51-8.20 (m, 4H, ar-H + ¹H pyrimidine), 9.31 (br, H, NH exchangeable).

Compound (17), IR: 3458,3377,3231,3195 (NH₂, NH), 2225 (C=N), 1627 (C=N), 1595 (C=C), 1546 cm⁻¹ (N=N). ¹H-NMR (DMSO-d₆): 2.27 (s, 6H, 2 CH₃), 6.68, 6.76 (s, 4H, 2NH₂ exchangeable), 7.59-8.22 (m, 4H, ar-H + ¹H pyrimidine), 10.34, 11.53 (br, 2H, 2NH exchangeable).

3-Amino-2-[4-N-(4,6-dimethylpyrimidin-2-yl)sulfamoyl]phenylazo-3-(piperidin-1-yl) acrylonitrile (18).

Equimolar amounts of **2** (10 mmol, 3.55 g) and piperidine (10 mmol, 1 mL), in absolute ethanol (70 mL), were refluxed for 1 h. The solid product so formed while hot was filtered off and crystallized to give **18** (Table 1). IR: 3400, 3317, 3227, 3202 (NH₂, NH), 2188 (C=N), 1629 (C=N), 1600 (C=C), 1568 cm⁻¹ (N=N). ¹H-NMR (DMSO-d₆): 1.65 (s, 6H, 3 CH₂), 2.27 (s, 6H, 2 CH₃), 3.27 (s, 4H, -CH₂-N-CH₂-), 3.60 (br, 2H, NH₂ exchangeable), 6.74 (s, ¹H, NH exchangeable), 7.537.92 (m, 4H, ar-H + ¹H pyrimidine). MS (%): 440 [M⁺] (0.9), 441 [M⁺ + 1] (2.9), 442 [M⁺ + 2] (2.7).

3-Amino-2-[4-N-(4,6-dimethylpyrimidin-2-yl)sulfamoyl]phenylazo-3-(piperidin-1-yl)acrylic acid (19).

A mixture of **18** (5 mmol, 2.20 g) and hydrochloric acid (2 M, 50 ml) was refluxed for 6 h., concentrated and left to cool. The solid deposited was collected by filtration, dried and crystallized to afford **19** (Table 1). IR: broad 3459-2723 (OH acid, NH₂), 1650 (CO acid), 1620 (C=N), 1594 (C=C), 1545 cm⁻¹ (N=N). ¹H-NMR (DMSO-d₆): 1.69 (s, 6H, 3 CH₂), 2.23 (s, 6H, 2 CH₃), 3.24 (s, 4H, -CH₂-N-CH₂-), 3.73 (br, 2H, NH₂ exchangeable), 7.40-7.88 (m, 4H, ar-H + 1H pyrimidine), 9.63 (s, 1H, NH exchangeable), 13.42 (s, 1H, COOH).

5-[4-N-(4,6-dimethylpyrimidin-2-yl)sulfamoyl]phenylazo-6-imino-4-(piperidin-1-yl)-3H-1,3thiazine-2(6H)-thione (20).

A mixture of **18** (5 mmol, 2.2 g), carbon disulfide (4 mL) and dry pyridine (10 mL) was stirred at room temperature for 24h. The reaction mixture was poured onto ice/water and neutralized by dilute HCl. The solid product was filtered off, washed with water (3x50), dried and crystallized to yield **20** (Table 1). IR: 3320,3229,3205 (NH), 1629 (C=N), 1599 (C=C), 1569 cm⁻¹ (N=N). ¹H-NMR (DMSO-d₆): 1.67 (s, 6H, 3 CH₂), 2.27 (s, 6H, 2 CH₃), 3.60 (s, 4H, -CH₂-N-CH₂-), 6.74 (s, 1H, NH exchangeable), 7.55-7.94 (m, 4H, ar-H + 1H pyrimidine), 8.84, 9.63 (br, 2H, 2NH exchangeable). MS (%): 290 [M-226] (100).

5-[4-N-(4,6-dimethylpyrimidin-2-yl)sulfamoyl]phenylazo-6-(piperidin-1-yl)pyrimidine-2,4(1H,3H)-dithione (21).

A mixture of **18** (5 mmol, 2.2 g), carbon disulfide (4 mL) and dry pyridine (15 mL) was refluxed for 3h. The reaction mixture was poured onto ice/water and neutralized by dilute HCl. The solid product was filtered off, washed with water (3x50), dried and crystallized to produce **21** (Table 1). IR: 3319, 3227, 3203 (NH), 1626 (C=N), 1598 (C=C), 1567 cm⁻¹ (N=N). MS (%): 290 [M-226] (44).

5-[4-N-(4,6-dimethylpyrimidin-2-yl)sulfamoyl]phenylazo-6-(piperidin-1-yl)pyrimidine-2(1H)-thione (22).

Phenyl isothiocyanate (5 mmol, 0.6 mL) was added to a stirred mixture of **18** (5 mmol, 2.20 g) and powdered sodium hydroxide (0.8 g) in dioxane (40 mL). After stirring for 4h., the reaction mixture was refluxed for another 4 h., then poured onto dilute acetic acid (15%, 40 mL). The precipitated solid was filtered and crystallized to afford **22** (Table 1). IR: 3322, 3223, 3209 (NH), 1626 (C=N), 1593 (C=C), 1559

cm-1 (N=N). ¹H-NMR (DMSO-d₆): 1.64 (s, 6H, 3 CH₂), 2.25 (s, 6H, 2 CH₃), 3.62 (s, 4H, -CH₂-N-CH₂-), 6.78 (s, 1H, NH exchangeable), 7.55-8.17 (m, 9H, ar-H + 1H pyrimidine), 8.89, 9.54 (br, 2H, 2NH exchangeable).

Table 1: Physical and analytical data of compounds 2-22

Comp. no.	m.p. (°C) Cryst. solvent	Colour Yield %	Mol. Form. (M.Wt)	Analyses Calcd./Found			
				C	H	N	S
2	210-1 E*	Yellow 79	C ₁₅ H ₁₃ N ₇ O ₂ S 355	50.70	3.66	27.60	9.01
				51.00	3.86	27.89	9.25
3	260-2 M/T**	Orange 78	C ₁₅ H ₁₇ N ₉ O ₂ S 387	46.51	4.39	32.56	8.27
				46.80	4.60	32.76	8.52
4	280-2 DMF/E	Yellow 92	C ₂₁ H ₂₁ N ₉ O ₂ S 463	54.42	4.54	27.21	6.91
				54.61	4.75	27.56	7.18
5	230-2 E	Orange 77	C ₂₂ H ₂₁ N ₉ O ₃ S 491	53.77	4.28	25.66	6.51
				54.00	4.44	25.88	6.73
6	243-5 D***	Yellow 75	C ₁₆ H ₁₈ N ₁₀ O ₃ S 430	44.65	4.19	32.56	7.44
				44.85	4.40	32.75	7.66
7	231-3 DMF/E	Yellow 90	C ₁₇ H ₁₉ N ₉ O ₃ S 429	47.55	4.43	29.37	7.46
				47.37	4.56	29.20	7.25
8	225-7 D	Red 80	C ₂₂ H ₂₁ N ₉ O ₃ S 491	53.77	4.28	25.66	6.52
				53.56	4.05	25.90	6.31
9a	> 300 D/H ₂ O	Brown 55	C ₂₂ H ₂₀ ClN ₉ O ₂ S 509.5	51.82	3.93	24.73	6.28
				52.02	4.24	24.55	6.49
9b	> 300 D/E****	Dark Brown 60	C ₂₄ H ₂₆ N ₁₀ O ₂ S 518	55.60	5.02	27.03	6.18
				55.89	5.30	26.84	5.91
9c	> 300 D/M	Brown 76	C ₂₃ H ₂₃ N ₉ O ₃ S 505	54.65	4.55	24.95	6.34
				54.46	4.31	24.71	6.08
10	277-9 D	Yellow 91	C ₁₆ H ₁₇ N ₉ O ₃ S 415	46.27	4.10	30.36	7.71
				46.00	4.00	30.49	7.46
11	282-4 D	Orange 62	C ₁₆ H ₁₇ N ₉ O ₂ S ₂ 431	44.55	3.94	29.23	7.42
				44.77	4.05	29.00	7.62
12	265-7 D	Brown 73	C ₁₆ H ₁₈ N ₁₀ O ₂ S 414	46.38	4.35	33.82	7.73
				46.66	4.60	34.03	8.00
14	268-70 E	Yellow 55	C ₁₈ H ₁₄ N ₈ O ₃ S 422	51.18	3.32	26.54	7.58
				51.42	3.11	26.30	7.80
15	241-2 E	Yellow 41	C ₁₉ H ₁₇ N ₇ O ₄ S 439	51.94	3.87	22.32	7.29
				51.77	4.09	22.60	7.60
17	> 300 D	Deep Red 71	C ₂₁ H ₁₇ N ₁₁ O ₂ S 487	51.75	3.49	31.62	6.57
				51.53	3.26	31.35	6.30
18	244-6 D	Orange 81	C ₂₀ H ₂₄ N ₈ O ₂ S 440	54.55	4.54	25.45	7.27
				54.77	4.77	25.74	7.00
19	271-4 D	Yellow 59	C ₂₀ H ₂₅ N ₇ O ₄ S 459	52.29	5.45	21.35	6.97
				52.00	5.69	21.60	7.21
20	> 300 D	Orange 77	C ₂₁ H ₂₄ N ₈ O ₂ S ₃ 516	48.84	4.65	21.71	18.60
				48.66	4.44	21.46	18.33
21	> 300 D	Orange 60	C ₂₁ H ₂₄ N ₈ O ₂ S ₃ 516	48.84	4.65	21.71	18.60
				48.59	4.37	21.50	18.39
22	> 300 D	Orange 56	C ₂₇ H ₂₉ N ₉ O ₂ S ₂ 575	56.35	5.04	21.91	11.13
				56.09	4.87	21.67	10.89

- * E = Ethanol
 ** M/T = Methanol-Toluene mixture
 ***D = Dioxane
 ****D/E = Dioxane- Ethanol mixture

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