

The Effect of Solvent on the Synthesis of Pyridazinones and Some Reactions of the New Compounds

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The reaction of 4-aryl-4-oxo-2-(5-oxo-1,3-diphenyl-2-pyrazolin-4-yl) butanoic acids with hydrazine hydrate in ethanol afforded the 4-(5-oxo-2-pyrazolin-4-yl)-3(2*H*)-pyridazinones and in 1-butanol a pyrazolo[3,4-*c*]pyridazino[4,3-*e*]pyridazine derivative was obtained while in acetic acid the 6-aryl-3(2*H*)-pyridazinone was the product. Probable mechanism has been proposed. The behavior of the 4-(5-oxo-2-pyrazolin-4-yl)-3(2*H*)-pyridazinones towards dimethyl sulfate, diethyl sulfate, ethyl bromoacetate, *p*-anisaldehyde, phosphoryl chloride and phosphorus pentasulfide has been studied.

It was stated that the reaction of an equimolar amount of 4-aryl-4-oxo-2-(5-oxo-1,3-diphenyl-2-pyrazolin-4-yl) butanoic acids with hydrazine hydrate in ethanol gave the 4-(5-oxo-2-pyrazolin-4-yl)-4,5-dihydropyridazin-3(2*H*)-one derivatives.¹⁾ The object of the present work is to replace ethanol as a solvent with acetic acid or 1-butanol to study the influence of the solvent on the type of reaction products and also to study the behavior of the resulting pyridazinones towards several reagents (Schemes 1, 2, and 3).

Thus, the reaction of 4-oxo-4-(*p*-chloro- and *p*-methylphenyl)-2-butenic acids (**1a** and **1b**) with 1,3-diphenyl-2-pyrazolin-5-one (**2**) in dry benzene gave 4-aryl-4-oxo-2-(5-oxo-1,3-diphenyl-2-pyrazolin-4-yl)butanoic acids (**3a** and **3b**). Their structures were derived from their IR[#] spectra showing the absorption bands of $\nu_{\text{C=O}}$ (acid) at 1730–1715, $\nu_{\text{C=O}}$ at 1670–1645, and $\nu_{\text{C=N}}$ at 1605–1590. The ¹H NMR (DMSO-*d*₆) spectrum of **3a** exhibited signals at δ =7.88–7.40 (14H, m, ArH) and 4.28–3.28 (4H, m, CH₂–CH+COCH).

The reaction of **3a** and **3b** with hydrazine hydrate and/or phenylhydrazine in boiling ethanol gave 6-aryl-4-(5-oxo-2-pyrazolin-4-yl)-4,5-dihydropyridazin-3(2*H*)-ones (**4a** and **4b**) and 2,6-diaryl-4-(5-oxo-2-pyrazolin-4-yl)-4,5-dihydropyridazin-3(2*H*)-ones (**4c** and **4d**), respectively. The structure of **4** was derived from their IR spectra showing the bands of $\nu_{\text{C=O}}$ at 1690–1640, $\nu_{\text{C=N}}$ at 1600–1590 and ν_{NH} at 3450–3380 for compounds **4a** and **4b**.

The reaction of **3a** with hydrazine hydrate in acetic acid gave the 6-(*p*-chlorophenyl)-3(2*H*)-pyridazinone (**5**). The reaction presumably proceeds through the normal condensation of hydrazine hydrate with the keto acid to give the intermediate **4a** with subsequent loss of the pyrazolone ring according to the mechanism given in Scheme 4.

The structure of compound **5** was supported by the following facts:

1. By similarity of melting point with the authentic compound prepared alternatively by the reaction of the corresponding 4-aryl-4oxo-2-butenic acid

(**1a**) with hydrazine hydrate in ethanol.²⁾

2. By refluxing **4a** with acetic acid, compound **5** and 1,3-diphenyl-2-pyrazolin-5-one were obtained.
3. The ¹H NMR (DMSO-*d*₆) spectrum of **5** showed signals at δ =8.00–6.84 (6H, m, ArH+2H-pyridazinone) and 13.20 (1H, s, NH).
4. The IR spectrum showed the absorption bands of $\nu_{\text{C=O}}$ at 1640, $\nu_{\text{C=N}}$ at 1590, and ν_{NH} at 3420.

On the other hand, the reaction of **3b** with two moles of hydrazine hydrate in boiling 1-butanol for 10 h gave 6,9,9a,9b-tetrahydro-8-(*p*-tolyl)-1,3-diphenyl-3*H*-pyrazolo[3,4-*c*]pyridazino[4,3-*e*]pyridazine (**6**). The reaction presumably proceeds through normal condensation of one mole of hydrazine hydrate with **3b** to give the pyridazinone derivative **4b**, which undergo condensation involving both the cyclic ketone moiety with a second mole of hydrazine hydrate to give **6**. The structure of **6** was derived from the IR spectrum showing the bands of $\nu_{\text{C=N}}$ at 1585, ν_{NH} at 3240, and being devoid of $\nu_{\text{C=O}}$.

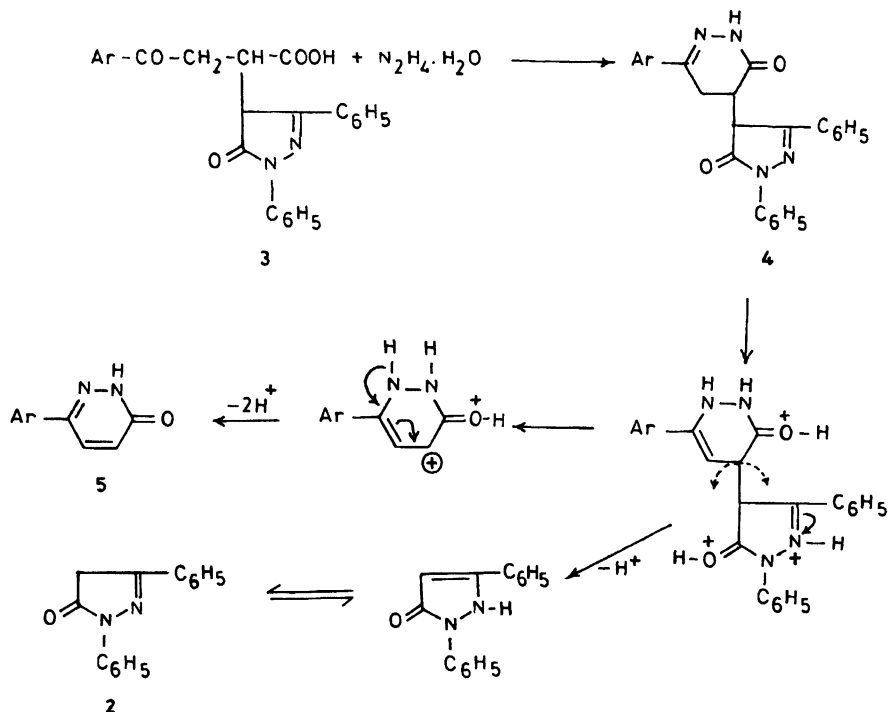
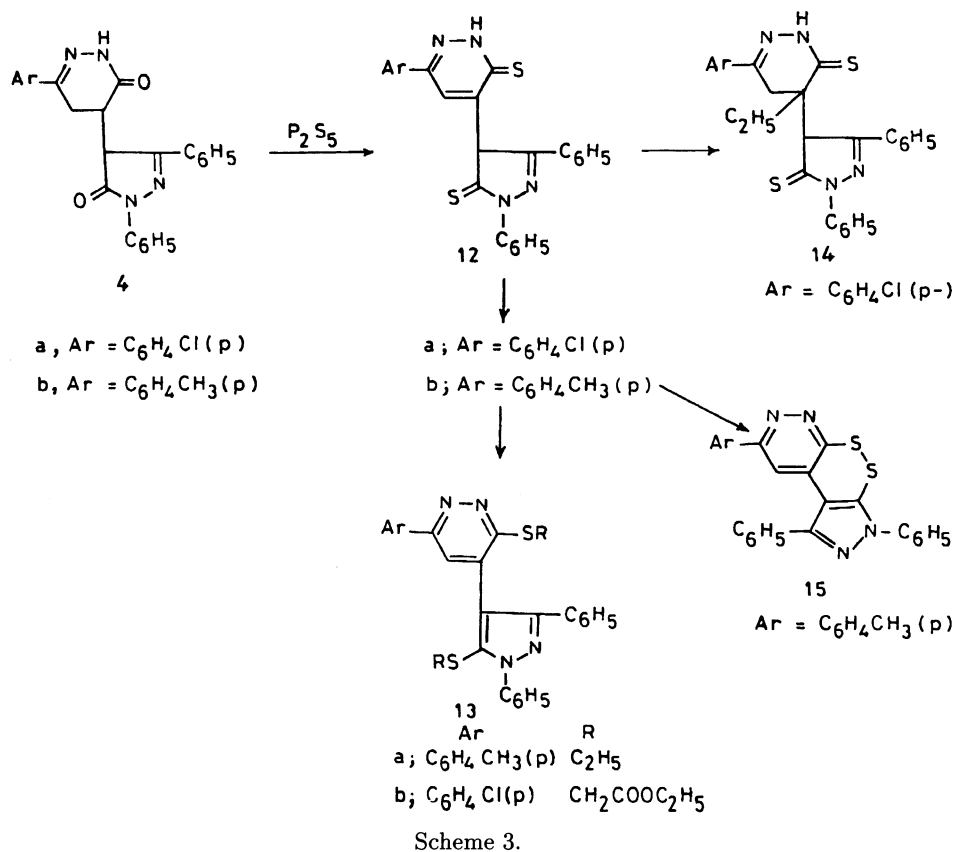
Compounds **4a** and **4b** were subjected to further studies. Thus, reactions of **4a** and **4b** with dimethyl sulfate, diethyl sulfate or ethyl bromoacetate gave the *N*-substituted products **4e–h**. The IR spectra showed the bands of $\nu_{\text{C=O}}$ at 1660, $\nu_{\text{C=N}}$ at 1660–1590. An additional band at 1740 for 4 h is attributable to $\nu_{\text{C=O}}$ (carboxylic ester).

Condensation of **4b** with *p*-anisaldehyde in the presence of ethanolic KOH took place at the 5-position to give a 4,5,6-trisubstituted pyridazin-3(2*H*)-one **7**.

Then we investigated the behavior of the 4-(5-oxo-pyrazolin-4-yl)pyridazin-3(2*H*)-ones **4** towards electrophilic reagents like POCl₃. Treatment of **4a** and **4b** with POCl₃ gave the 3-chloro-4-(5-chloro-1,3-diphenyl-2-pyrazolin-4-yl)-6-(4-substituted phenyl) pyridazines **8a** and **8b**, respectively, by substitution of the enolic hydroxyl group with chlorine together with dehydrogenation. The phenomenon of dehydrogenation is not strange since it is observed in the reactions of pyridazinones with P₂S₅³⁾ and Grignard reagents.⁴⁾ Their structures were derived from their IR spectra showing the band of $\nu_{\text{C=N}}$ at 1610–1600 and being devoid of $\nu_{\text{C=O}}$.

[#]IR ν_{max} here and elsewhere in the paper in cm^{–1}.

ethanol gave the dihydrazino (**9a** and **9b**), the dianilino (**9d** and **9e**) and the dimorpholino (**9f**) derivatives, respectively. Their structures were derived from their IR spectra showing the bands of $\nu_{\text{C=N}}$ at 1610–1595 and $\nu_{\text{N-H}}$ at 3480–3420. The ^1H NMR ($\text{DMSO-}d_6$) spectrum of **9a** showed signals at 8.00–7.00 (15H, m, Ar-



H+H-pyridazine). The 1H NMR ($CDCl_3$) spectrum of **9e** showed signals at $\delta=7.89-7.07$ (25H, m, Ar-H+H-pyridazine) and at $\delta=2.40$ (3H, s, CH_3).

Structure of **9b** was further established by its reaction with salicylaldehyde to give bis(*N'*-salicylidenehydrazino)

no) derivative **9c**, its IR spectrum showed the bands of $\nu_{C=N}$ at 1610 and ν_{NH} , ν_{OH} at 3540 (broad).

Compounds **9a** and **9b** were used for the preparation of compounds containing both triazolo and tetrazolo rings.

Thus the reaction of the dihydrazino derivatives **9a** and **9b** with nitrous acid yielded the 4-(pyrazolo[*d*]-1,2,3-triazol-3a-yl)-tetrazolo[*b*]pyridazine derivatives **10a** and **10b**, respectively. Their structures were derived from their IR spectra showing the band of $\nu_{\text{C}=\text{N}}$ at 1600 and by their preparation through an alternative route by the reaction of the dichloro compounds **8a** and **8b** with sodium azide. The similarity of these compounds was identified by IR spectra and by mixed melting point determination with the samples prepared above.

Anthranilic acid reacted with **8a** at 150°C to give compound **11**. Its IR spectrum showed the band of $\nu_{\text{C}=\text{O}}$ at 1690 and $\nu_{\text{C}=\text{N}}$ at 1630.

Compounds **4a** and **4b** with phosphorus pentasulfide in dry xylene gave the corresponding 4-(1,3-diphenyl-5-thioxo-pyrazolin-4-yl)-6-(4-substituted phenyl)-3(2*H*)-pyridazinethiones (**12a** and **12b**), respectively, a reaction in which thioketone formation together with dehydrogenation take place. The IR spectra of **12** exhibited characteristic absorption bands for $\nu_{\text{N}=\text{C}=\text{S}}$ at 1470, $\nu_{\text{C}=\text{S}}$ at 1380, $\nu_{\text{C}=\text{N}}$ at 1600–1595, and ν_{NH} at 3480–3420.

Treatment of **12a** and **12b** with diethyl sulfate or ethyl bromoacetate in dry acetone in presence of anhydrous potassium carbonate yielded the corresponding S-substituted derivatives **13a** and **13b**, respectively. Their structures were derived from their IR spectra showing the band of $\nu_{\text{C}=\text{N}}$ at 1600–1590 in addition to a strong band at 1730 characteristic of the $\nu_{\text{C}=\text{O}}$ of ester of compound **13b**. The ^1H NMR (DMSO-*d*₆) spectrum of **13a** showed signals at δ =8.18–7.28 (15H, m, Ar-H+H-pyridazine), 3.34 (4H, q, 2×CH₂ of ethyl group), 2.44 (3H, s, CH₃), and 1.48 (6H, t, 2×CH₃ of ethyl group).

A study of the reaction of compound **12** with a Grignard reagent was also undertaken. When **12a** was treated with ethylmagnesium iodide compound **14** was obtained. This reaction takes place by 1,4-addition of the Grignard reagent to the unsaturated C=N. This is in accordance with the previous results.³⁾ The ^1H NMR (CDCl₃) spectrum of **14** showed signals at δ =8.20–7.30 (14H, m, Ar-H), 5.10 (1H, s, pyrazole proton), 4.40 (2H, s, pyridazine protons), 3.40 (2H, q, CH₂ of ethyl group), 1.40 (3H, t, CH₃ of ethyl group), and 13.00 (1H, s, NH).

Interestingly, compound **12b** was easily oxidized to the cyclic disulfide **15** by an iodine solution. Its IR spectrum showed the band of $\nu_{\text{C}=\text{N}}$ at 1600. The ^1H NMR (DMSO-*d*₆) spectrum of **15** showed signals at δ =8.02–7.26 (15H, m, Ar-H+H-pyridazine) and 2.98 (3H, s, CH₃). The characteristic feature of the mass spectrum of **15** is the absence of the parent ion but showed the cation ($\text{M}^+ - \text{C}_6\text{H}_5$) at m/z =373 (22.3%).

Experimental

Melting points are uncorrected. IR spectra (KBr) were recorded with a Unicam SP 1200 spectrophotometer. ^1H NMR were measured with a Varian VN 1009 (S-60 T) instrument using TMS as internal standard and mass spectra were measured with an AET-MS 902 mass spectrometer at

70 eV, 6 kV accelerating voltage at 130°C ion source temperature using a direct insertion probe.

Reaction of **1a** and **1b** with the Pyrazolinone:

Formation of **3a and **3b**:** To a solution of **1a** or **1b** (0.01 mol) in dry benzene (20 cm³), 1,3-diphenyl-2-pyrazolin-5-one (0.01 mol) was added and the reaction mixture refluxed for 10 h. The solid that separated on cooling was crystallized from toluene (**3a**) or ethanol (**3b**).

3a: mp 183°C; yield 95%. Calcd for C₂₅H₁₉ClN₂O₄: C, 67.18; H, 4.28; N, 6.27%. Found: C, 66.8; H, 4.3; N, 6.4%. MS m/z 446 (M^+).

3b: mp 173°C; yield 93%. Calcd for C₂₆H₂₂N₂O₄: C, 73.22; H, 5.20; N, 6.57%. Found: C, 73.32; H, 5.2; N, 6.21%. MS m/z 426 (M^+).

Reaction of **1a**, **3a**, **3b**, **8a**, **8b**, and **9b** with Hydrazines, Aniline, Morpholine, and Salicylaldehyde:

Formation of **4a–d, **5,6** and **9a–f**:** To a solution of **1a**, **3a**, **3b**, **8a**, **8b**, or **9b** (0.01 mol) in ethanol, acetic acid or 1-butanol (20 cm³), hydrazine hydrate, phenylhydrazine, aniline, morpholine, or salicylaldehyde (0.01 mol or 0.02 mol) was added and the reaction mixture refluxed for 5 or 10 h. The solid that separated on cooling was crystallized from ethanol (**4a–d**, **6**), benzene (**5**, **9a**, **9b**, **9d**, **9e**), acetic acid (**9c**), or petroleum ether (bp 90–100°C) (**9f**).

4a: mp 223°C; yield 87%. Calcd for C₂₅H₁₉ClN₄O₂: C, 67.79; H, 4.32; N, 12.65%. Found: C, 67.3; H, 4.4; N, 12.6%. MS m/z 442 (M^+).

4b: mp 212°C; yield 83%. Calcd for C₂₆H₂₂N₄O₂: C, 73.91; H, 5.25; N, 13.26%. Found: C, 73.56; H, 5.28; N, 12.85%. MS m/z 422 (M^+).

4c: mp 211°C; yield 75%. Calcd for C₃₁H₂₃ClN₄O₂: C, 71.74; H, 4.46; N, 10.79%. Found: C, 71.8; H, 4.2; N, 10.6%. MS m/z 518 (M^+).

4d: mp 197°C; yield 82%. Calcd for C₃₂H₂₆N₄O₂: C, 77.08; H, 5.21; N, 11.23%. Found: C, 77.4; H, 5.6; N, 11.4%. MS m/z 498 (M^+).

5: mp 262°C; yield 32%. Calcd for C₁₀H₇ClN₂O: C, 58.12; H, 3.41; N, 13.56%. Found: C, 58.2; H, 3.3; N, 13.2%. MS m/z 206 (M^+).

6: mp 239°C; yield 43%. Calcd for C₂₆H₂₂N₆: C, 74.61; H, 5.30; N, 20.08%. Found: C, 74.5; H, 5.6; N, 19.7%. MS m/z 418 (M^+).

9a: mp 143°C; yield 70%. Calcd for C₂₅H₂₁ClN₈: C, 64.03; H, 4.51; N, 23.88%. Found: C, 64.3; H, 4.6; N, 24.0%. MS m/z 468 (M^+).

9b: mp 161°C; yield 72%. Calcd for C₂₆H₂₄N₈: C, 69.62; H, 5.39; N, 24.98%. Found: C, 69.2; H, 5.4; N, 24.6%. MS m/z 448 (M^+).

9c: mp 286°C; yield 52%. Calcd for C₄₀H₃₂N₈O₂: C, 73.15; H, 4.91; N, 17.06%. Found: C, 73.4; H, 4.7; N, 17.2%. MS m/z 656 (M^+).

9d: mp 230°C; yield 52%. Calcd for C₃₇H₂₇ClN₆: C, 75.17; H, 4.60; N, 14.21%. Found: C, 75.3; H, 4.7; N, 14.4%. MS m/z 590 (M^+).

9e: mp 195°C; yield 51%. Calcd for C₃₈H₃₀N₆: C, 79.97; H, 5.29; N, 14.72%. Found: C, 80.2; H, 5.3; N, 14.7%. MS m/z 570 (M^+).

9f: mp 160°C; yield 48%. Calcd for C₃₃H₃₁ClN₆O₂: C, 68.44; H, 5.39; N, 14.51%. Found: C, 68.5; H, 5.5; N, 14.73%. MS m/z 578 (M^+).

Reaction of **4a** with Acetic Acid: Formation of

5: A solution of **4a** (0.01 mol) in acetic acid (20 cm³) was

refluxed for 5 h, concentrated and cooled. The separated solid was filtered and fractionally crystallized from benzene to give **5** and the residue was crystallized from ethanol to give 1,3-diphenyl-2-pyrazolin-5-one which was identified by melting point determination.

Reaction of 4a, 4b, 12a and 12b with Dimethyl Sulfate, Diethyl Sulfate, and Ethyl Bromoacetate: Formation of 4e—h and 13a,b: A mixture of **4a** or **4b**, **12a**, **12b** (0.01 mol), anhydrous potassium carbonate (0.03 mol), dimethyl sulfate, diethyl sulfate, or ethyl bromoacetate (0.03 mol) and dry acetone (50 cm³) was refluxed for 20 h and excess solvent removed. The product was crystallized from petroleum ether (bp 90–100°C) (**4e**, **4f**, **4h**, **13a**) or ethanol (**4g**, **13b**).

4e: mp 205°C; yield 53%. Calcd for C₂₆H₂₁ClN₄O₂: C, 68.34; H, 4.6; N, 12.26%. Found: C, 68.06; H, 4.23; N, 12.57%. MS *m/z* 456 (M⁺).

4f: mp 215°C; yield 55%. Calcd for C₂₇H₂₄N₄O₂: C, 74.29; H, 5.54; N, 12.83%. Found: C, 73.9; H, 5.4; N, 12.5%. MS *m/z* 436 (M⁺).

4g: mp 220°C; yield 62%. Calcd for C₂₈H₂₆N₄O₂: C, 74.64; H, 5.81; N, 12.43%. Found: C, 74.7; H, 5.8; N, 12.6%. MS *m/z* 450 (M⁺).

4h: mp 83°C; yield 67%. Calcd for C₂₉H₂₅ClN₄O₄: C, 65.84; H, 4.76; N, 10.59%. Found: C, 66.1; H, 4.3; N, 10.9%. MS *m/z* 528 (M⁺).

13a: mp 90°C; yield 56%. Calcd for C₃₀H₂₈N₄S₂: C, 70.83; H, 5.54; N, 11.01%. Found: C, 70.7; H, 5.7; N, 10.8%. MS *m/z* 508 (M⁺).

13b: mp 210°C; yield 48%. Calcd for C₃₃H₂₉ClN₄O₄S₂: C, 61.42; H, 4.53; N, 8.68%. Found: C, 61.7; H, 4.22; N, 8.8%. MS *m/z* 644 (M⁺).

Condensation of *p*-Anisaldehyde with 4b: Formation of 7: A warm solution of **4b** (0.01 mol) in ethanol (20 cm³) was treated with ethanol KOH solution (25 cm³; 4%) and then *p*-anisaldehyde (0.01 mol) was added dropwise with continuous shaking. The reaction mixture was refluxed for 2 h, cooled, poured into cold water and the solid obtained was crystallized from benzene to give **7**.

7: mp 215°C; yield 46%. Calcd for C₃₄H₂₈N₄O₃: C, 75.53; H, 5.22; N, 10.36%. Found: C, 75.3; H, 5.1; N, 10.7%. MS *m/z* 540 (M⁺).

Reaction of 4a and 4b with POCl₃: Formation of 8a and 8b: A mixture of **4a** or **4b** (0.01 mol) and POCl₃ (10 cm³) was gently refluxed for 30 min, cooled, treated with crushed ice and the precipitated solid filtered and crystallized from petroleum ether (bp 90–100°C) (**8a**) or ethanol (**8b**).

8a: mp 193°C; yield 45%. Calcd for C₂₅H₁₅Cl₃N₄: C, 62.85; H, 3.16; N, 11.72%. Found: C, 63.1; H, 3.2; N, 11.8%. MS *m/z* 477 (M⁺).

8b: mp 150°C; yield 43%. Calcd for C₂₆H₁₈Cl₂N₄: C, 68.27; H, 3.96; N, 12.25%. Found: C, 68.3; H, 4.1; N, 12.5%. MS *m/z* 457 (M⁺).

Reaction of 9a and 9b with Nitrous Acid: Formation of 10a and 10b: An aqueous solution of sodium nitrite (0.03 mol in 10 cm³ H₂O) is added dropwise with stirring to a solution of **9a** (or **9b**) (0.01 mol) in 4 equiv acetic acid (10 cm³) and stirring is continued for 1 h. The solid formed is filtered and crystallized from petroleum ether (60–80°C) (**10a**) or ethanol (**10b**).

10a: mp 207°C; yield 43%. Calcd for C₂₅H₁₅ClN₁₀: C,

61.17; H, 3.08; N, 28.52%. Found: C, 60.9; H, 3.2; N, 28.8%. MS *m/z* 490 (M⁺).

10b: mp 196°C; yield 45%. Calcd for C₂₆H₁₈N₁₀: C, 66.37; H, 3.85; N, 29.77%. Found: C, 66.6; H, 3.9; N, 29.5%. MS *m/z* 470 (M⁺).

Reaction of 8a and 8b with Sodium Azide: Formation of 10a and 10b: A mixture of **8a** or **8b** (1 g), sodium azide (2 g) water (5 cm³) and *N,N*-dimethylformamide (20 cm³) was boiled for 2 h and then cooled. The solid obtained upon dilution with water was filtered and crystallized from petroleum ether (bp 60–80°C) (**10a**) or ethanol (**10b**).

Reaction of 8a with Anthranilic Acid: Formation of 11: A mixture of **8a** (0.01 mol) and anthranilic acid (0.02 mol) was heated in an oil bath at 150°C for 1 h, cooled and triturated with ethanol. The solid obtained was crystallized from ethanol to give **11**.

11: mp 275°C; yield 70%. Calcd for C₃₉H₂₃ClN₆O₂: C, 72.82; H, 3.60; N, 13.06%. Found: C, 72.5; H, 3.5; N, 13.4%. MS *m/z* 642 (M⁺).

Reaction of 4a and 4b with P₂S₅: Formation of 12a and 12b: A solution of **4a** (or **4b**) (0.01 mol), P₂S₅ (0.03 mol), and dry xylene (50 cm³) was boiled under reflux for 6 h. The reaction mixture was filtered while hot and then concentrated. The product which separated on cooling was crystallized from xylene (**12a**) or ethanol (**12b**).

12a: mp 228°C; yield 62%. Calcd for C₂₅H₁₇ClN₄S₂: C, 63.47; H, 3.62; N, 11.84%. Found: C, 63.6; H, 3.7; N, 11.5%. MS *m/z* 472 (M⁺).

12b: mp 197°C; yield 67%. Calcd for C₂₆H₂₀N₄S₂: C, 68.99; H, 4.45; N, 12.38%. Found: C, 69.2; H, 4.5; N, 12.4%. MS *m/z* 452 (M⁺).

Reaction of Grignard Reagent with 12a: Formation of 14: A solution of ethylmagnesium iodide (prepared from 0.03 mol of ethyl iodide and 0.03 atom of magnesium) was added to a solution of **12a** (0.01 mol) in dry ether. The resultant solution was refluxed for 4 h in a boiling water bath and left overnight. The reaction mixture was then hydrolyzed with a saturated solution of ammonium chloride, extracted with ether and the solvent removed to give a solid product. The solids were crystallized from petroleum ether (bp 90–100°C) to give **14**.

14: mp 104°C; yield 45%. Calcd for C₂₇H₂₃ClN₄S₂: C, 64.45; H, 4.60; N, 11.13%. Found: C, 64.8; H, 4.7; N, 11.4%. MS *m/z* 502 (M⁺).

Oxidation of 12b with Iodine: Formation of 15: A solution of iodine (0.02 mol) in 5% KI solution (100 cm³) was added dropwise with stirring to a solution of **12b** (0.01 mol) in 10% aqueous sodium hydroxide (10 cm³) until the color of iodine persists. The solid formed was filtered off and crystallized from DMF to give **15**.

15: mp 225°C; yield 60%. Calcd for C₂₆H₁₈N₄S₂: C, 69.30; H, 4.02; N, 12.43%. Found: C, 69.4; H, 4.3; N, 12.7%. MS *m/z* 373 (M⁺ – C₆H₅).

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