Bis-enaminones as precursors for synthesis of novel 3,4-bis(heteroaryl)pyrazoles and 3,6-bis-(heteroaryl)-pyrazolo[3,4-*d*]pyridazines

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Coupling of bis-enaminones with benzenediazonium chloride and 3-diazo-1,2,4-triazole proved to be convenient routes for the synthesis of novel 3,4-bis(pyrazol-3-yl)pyrazoles and 3,6-bis(heteroaryl)pyrazolo[3,4-*d*]pyridazines which have not been reported previously. The structures of the products were elucidated on the basis of their spectral properties, elemental analyses and, wherever possible, by alternate synthesis.

Keywords: hydrazonoyl halides, heterocycles, enaminones, pyrazoles

Enaminones of type **I** have been reported to couple with arenediazonium salts to give the respective hydrazones **II** (Scheme 1).[1–4] In conjunction with our recent work on bis-enaminones of type **III**,[5–10] we decided to study their azo coupling with diazotised aromatic and heterocyclic amines and to explore the reactions of the resulting azo-coupled products **IV** and **V** with hydrazine hydrate. Our aim was to shed some light on the site selectivity in the latter reactions and explore their utility to synthesise new ter-heterocycles of type **VI** and **VII** (Fig. 1).

Results and discussion

The starting bis-enaminones **2a–c** were prepared by condensation of 3,4-diacetylpyrazole derivatives **1a–c** each with *N*,*N*-dimethylformamide dimethyl acetal (DMF-DMA) as previously described from our laboratory (Scheme 1).[10] Coupling of **2a–c** each with a benzenediazonium derivative in ethanol in the presence of sodium acetate gave the corresponding coupling products **3a–c** (Scheme 1). Although the latter compounds can exist in the *E*- and/or *Z*-forms, their ^{1}H NMR spectra revealed that they exist only in the form *Z*-**3** (Fig. 2). This is because such spectra revealed, in each case, two singlet signals in the regions δ 10.35–10.37 and 12.79–12.88 due to the resonances of the –CHO and hydrazone NH protons, respectively (see Experimental). These chemical shift values are similar to those of the *syn*-isomers (*Z*-isomers) of 3-aryl-3-oxo-2-arylhydrazononopropanals (ArCOC(CHO)=NNHAr') which were reported to exhibit their –CHO signals at δ 9.50–9.63 and hydrazone NH signals at δ 11.85–12.78.[11] The ^{1}H NMR spectra of the *anti*-isomers (*E*-isomers) of the latter compounds revealed the signals for their –CHO and NH proton resonances in the regions δ 9.96–10.17 and 13.9–14.35, respectively.[11]

Condensation of the product **3a** with hydrazine hydrate was next examined to shed some light on its site reactivity. This reaction yielded a single product that was identified, on the basis of its spectral and elemental analyses, as **4** rather than **5** (Scheme 2). The assigned structure **4** was confirmed by comparison with an authentic sample of **5**, prepared by an unambiguous synthesis as depicted in Scheme 2. Thus, reaction

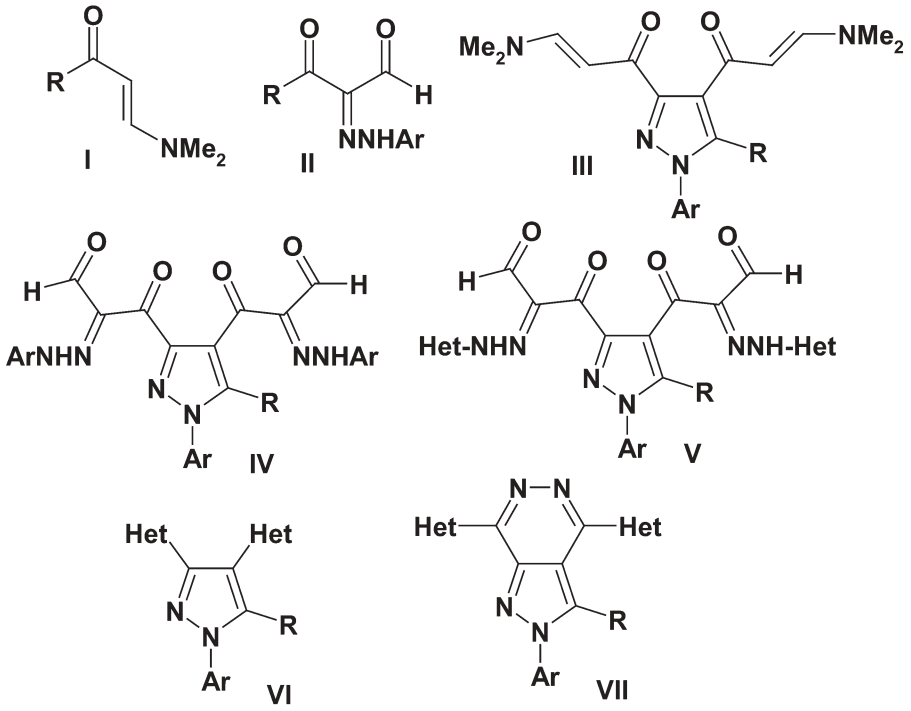

Fig. 1

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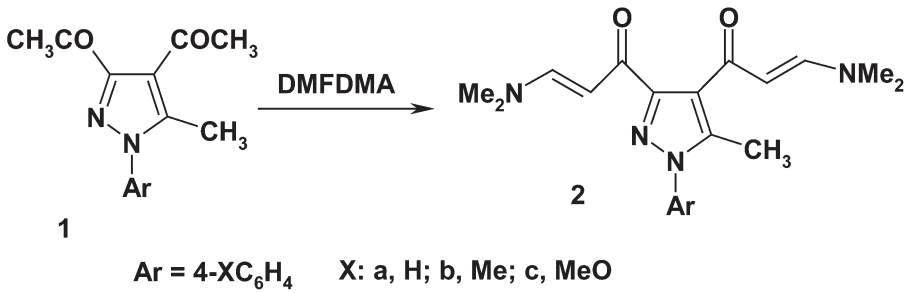


Scheme 1

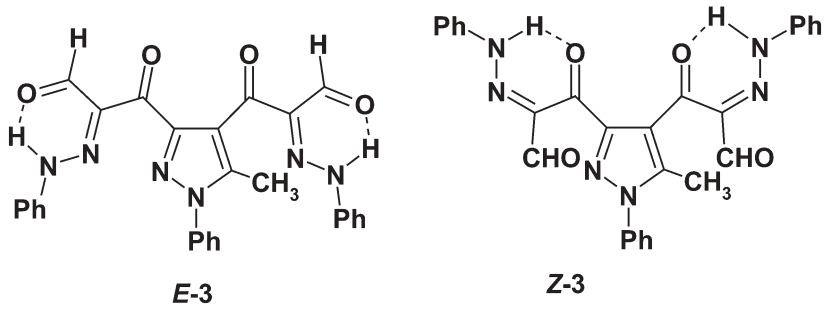
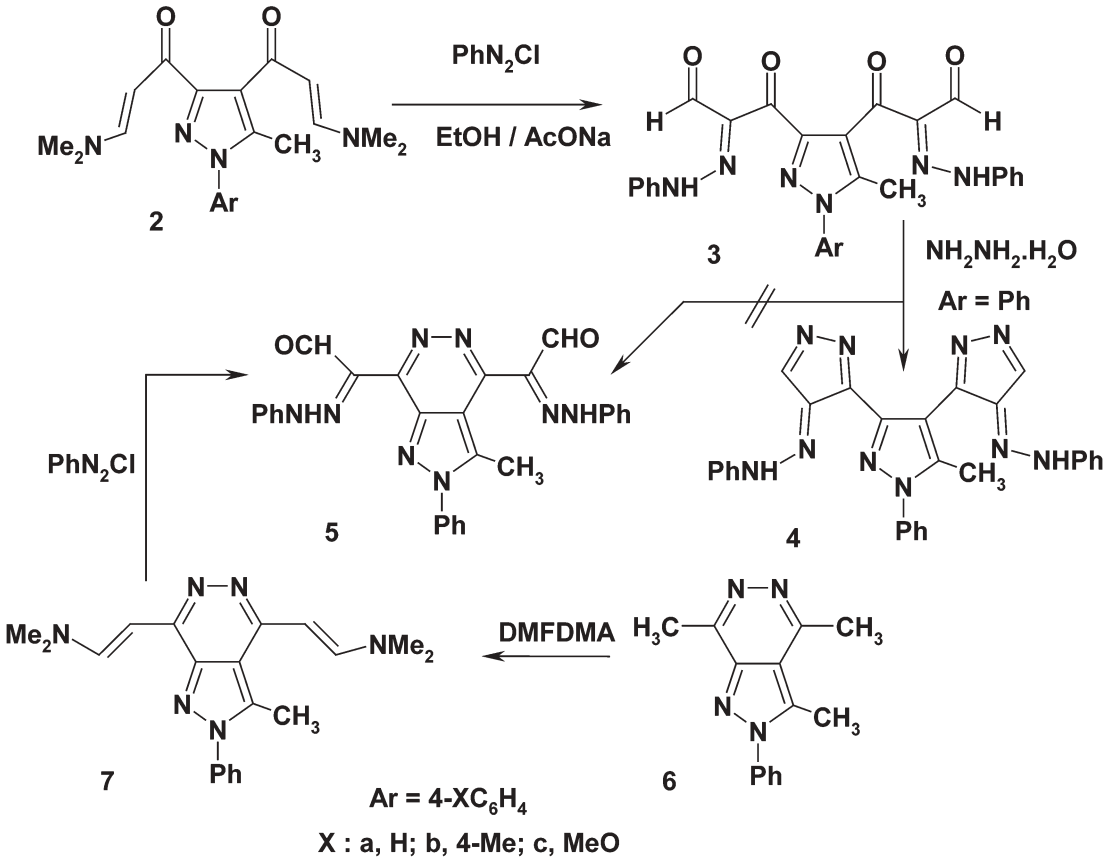


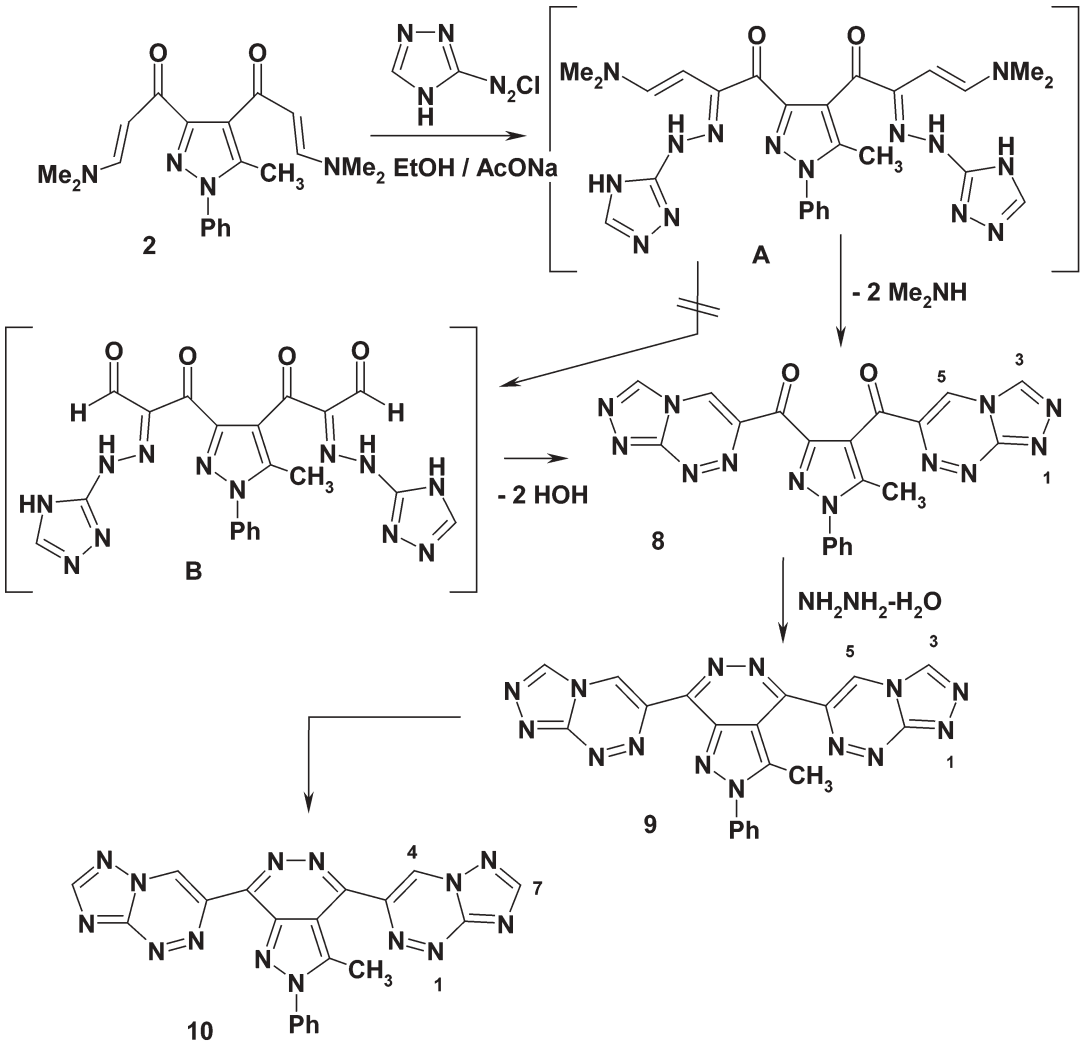
Fig. 2



Scheme 2

of 3,6-dimethylpyrazolo[3,4-*d*]pyridazine **6** with DMF-DMA yielded the bis-enamine **7**. Coupling of the latter with benzenediazonium chloride afforded **5** which proved completely different from **4** (Scheme 2). The structures of the products **4–7** were consistent with their spectral (IR, ^{1}H NMR and Ms) and microanalytical data (see Experimental).

Next, we examined coupling of **2a** with diazotised 3-amino-1,2,4-triazole in ethanol in the presence of sodium acetate. The reaction afforded a product that proved to be the respective 1-phenyl-5-methyl-3,4-bis[1,2,4-triazolo[3,4-*c*][1,2,4]triazin-6-yl)carbonyl]pyrazole **8a** (Scheme 3). Structure assignment of the latter was based on its spectral and elemental analysis

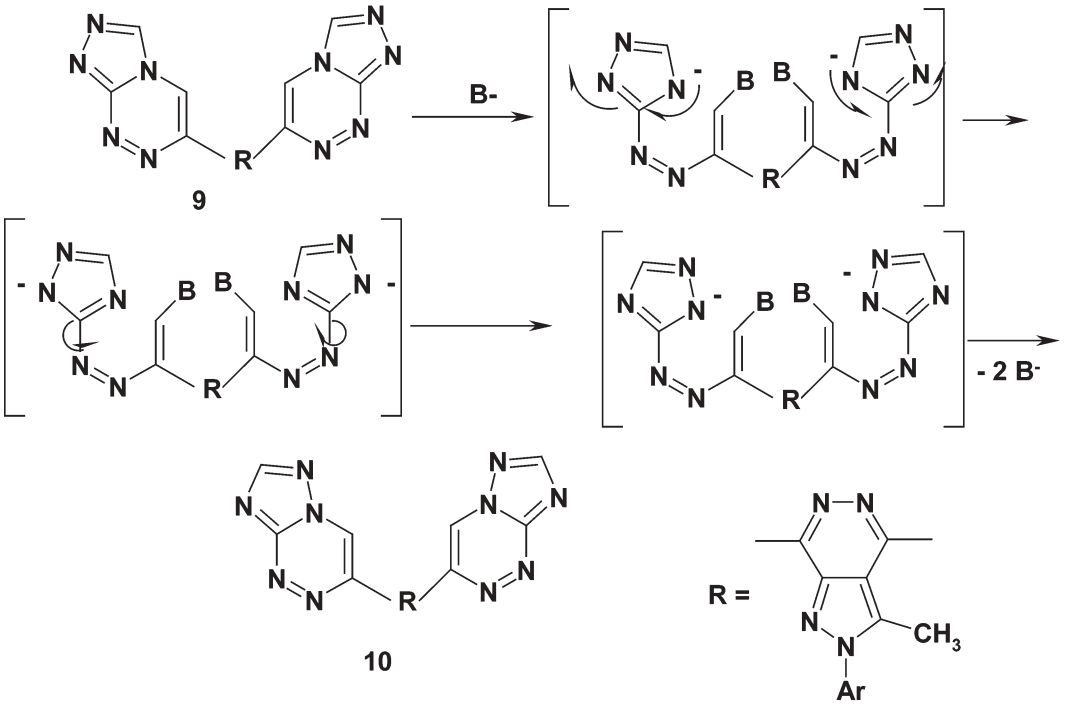


Scheme 3

data. For example, its IR spectrum showed no –CHO bands but instead exhibited a ketonic carbonyl band in the region ν 1676–1681 cm^{-1}. Also, its ^1H NMR spectrum did not show a signal characteristic of a –CHO group, they it exhibited two characteristic singlet signals at δ 7.87and 8.58 assignable to H-3 and H-5 of the [1,2,4]triazolo[3,4-*c*][1,2,4]triazine ring residue, respectively.[12]

To account for the formation of **8**, it is suggested (Scheme 3), that the initially formed azo coupled product **A** can either undergo *in situ* cyclisation *via* elimination of dimethylamine to give **8** or hydrolysis to give the bis-aldehyde derivative **B** which in turn undergoes dehydrative cyclisation to afford **8**. However, all attempts to isolate the intermediate **B** failed. This suggests that former route involving direct *in situ* cyclisation of the intermediate **A** is the predominant route.

Next, condensation of the diketone **8** with hydrazine hydrate was examined to see if it would yield 3,6-bis([1,2,4]triazolo [3,4-*c*][1,2,4]triazin-3-yl)-1-phenyl-7-methyl-pyrazolo[3,4-*d*]-pyridazine **9** or its isomer, namely 3,6-bis([1,2,4]triazolo [5,1-*c*][1,2,4]triazin-3-yl)-1-phenyl-7-methylpyrazolo[3,4-*d*]-pyridazine **10** (Scheme 3). Literature reports indicate that the [1,2,4]triazolo[3,4-*c*][1,2,4]triazine derivatives undergo readily Dimroth-type rearrangement to give the respective [1,2,4]triazolo[5,1-*c*][1,2,4]triazine isomers upon heating in the presence of a base.[13,14] However, when the diketone **8** was refluxed with hydrazine hydrate for 2 h the product was identified as the pyrazolopyridazine **9**. The structure of the latter was established on the basis of its spectral (IR, ^1H NMR, MS) and elemental analyses (see Experimental). When **9** was heated in ethanol in the presence of sodium hydroxide, it isomerised to the thermodynamically more stable [1,2,4]triazolo[5,1-*c*] [1,2,4]triazine derivative **10** *via* a Dimroth-type rearrangement through tandem ring opening and ring closure reactions (Scheme 4). This rearrangement is compatible with those reported in earlier reports on rearrangements of [1,2,4] triazolo[3,4-*c*][1,2,4]triazines and pyrimidines.[12,15] Conclusive evidence for the rearrangement of **9** into **10**, was provided by comparison of their ^1H NMR spectra. For example, the ^1H NMR spectrum of **9** revealed the H-3 proton signal at δ 8.88, whereas that of **10** showed the H-7 proton signal at δ 8.47. This feature is consistent with literature reports which indicate that the H-3 proton of [1,2,4]triazolo[3,4-*c*]pyrimidine is more deshielded than that of H-7 of [1,2,4]triazolo[5,1-*c*]pyrimidine.[14] The driving force for this rearrangement seems to be due to the fact that the [1,2,4]triazolo[5,1-*c*][1,2,4]triazine ring system is thermodynamically more stable than its isomer namely [1,2,4]triazolo[3,4-*c*][1,2,4]triazine as is the case for the [1,2,4]triazolopyrimidine isomers.[16] This suggestion was confirmed by calculating the heats of formation (ΔH_f) of both ring systems at the AM1 level using the Hyperchem program package (version 4.0).[17] The results of these calculations showed that the enthalpy of formation of [1,2,4]triazolo[3,4-*c*] [1,2,4]triazines (106.89 kcal mol^{-1}) is less than that of [1,2,4] triazolo[5,1-*c*][1,2,4]triazine (151.56 kcal mol^{-1}).



Scheme 4

Experimental

All melting points were determined on Gallenkamp apparatus. Solvents were generally distilled and dried by standard literature procedures prior to use. The IR spectra were measured on a Pye-Unicam SP300 instrument in potassium bromide discs. The ^{1}H NMR spectra were recorded on a Varian Mercury VXR-300 MHz spectrometer and the chemical shifts δ are downfield from tetramethylsilane (TMS) as an internal standard. The mass spectra were recorded on a GCMS-Q1000-EX Shimadzu and GCMS 5988-A HP spectrometers, the ionizing voltage was 70 eV. Elemental analyses were carried out by the Microanalytical Center of Cairo University, Giza, Egypt. The bis-enaminones **2** were prepared as previously described from our laboratory.[10]

Coupling of bis-enaminones 2a–c with diazotised aniline: A solution of the appropriate bis-enaminone **2** (1.76 g, 5 mmol) in ethanol (20 mL) and sodium acetate trihydrate (0.5 g) was cooled in an ice bath at 0–5°C while being stirred. Portions of the appropriate diazonium salt were added to the cold solution The salt was prepared by diazotising the appropriate aniline derivative (10 mmol) in hydrochloric acid (6M, 3 mL) with sodium nitrite (0.70 g, 10 mmol) in water (5 mL). After all the diazonium salt solution was added, the mixture was stirred for a further 30 min. while cooling in an ice-bath. The reaction mixture was then left in a refrigerator for three days. The solid that precipitated was filtered off, washed with water, dried and finally crystallised from ethanol to give the respective products **3**.

3,4-Bis(2-phenylhydrazo-1,2,3-trioxo-1-propyl)-1-phenyl-5-methylpyrazole (**3a**): Red solid, yield 1.0 g, (78 %), m.p. 122–124 °C, IR (KBr) ν_{max}/cm^{-1} 3425(NH), 1689, 1645 (C=O). ^{1}H NMR (DMSO-d_6) δ = 3.36(s, 3H, CH_3), 6.7–7.9 (m, 15H, ArH), 10.35 (s, 2H, 2HCO), 12.83 (s, 2H, 2NH). MS *m/z* (%) 506(M$^+$, 0.11), 169 (8), 153(15), 121 (4), 111 (11), 104 (11), 97 (17), 121 (14), 94 (88), 82 (14), 76 (16), 71 (18), 65 (50), 56 (25), 46 (100). Anal. Calcd for $C_{28}H_{22}N_6O_4$ (506.53) C, 66.40; H, 4.38; N, 16.59. Found: C, 66.70; H, 4.49; N, 16.72%.

3,4-Bis(2-phenylhydrazo-1,2,3-trioxo-1-propyl)-1-(4-methylphenyl)-5-methylpyrazole (**3b**): Brown solid, yield 1.0 g, (78 %), m.p, 166–168 °C, IR (KBr) ν_{max}/cm^{-1} 3427(NH), 1645, 1687(C=O). ^{1}H NMR (DMSO-d_6) δ = 2.48 (s, 3H, CH_3), 3.31(s, 3H, CH_3), 6.94–7.30(m, 14H, ArH), 10.35 (s, 2H, 2CHO), 12.88 (s, 2H, 2NH). MS *m/z* (%) 520(M$^+$, 0.1), 315 (15), 256 (16), 241 (100), 224 (27), 198 (28), 168 (19), 155 (20), 142 (20), 132 (50), 118 (14), 104 (33), 90 (93), 76 (36), 64 (59), 50 (22), 46 (10). Anal. Calcd for $C_{29}H_{24}N_6O_4$ (520.55) C, 66.91; H, 4.65; N, 16.14. Found: C, 67.15; H, 4.80; N, 16.00%.

3,4-Bis(2-phenylhydrazo-1,2,3-trioxo-1-propyl)-1-(4-methoxyphenyl)-5-methylpyrazole (**3c**): Pale brown solid, yield 1.0 g (78 %), m.p. 156–158 °C, IR (KBr) ν_{max}/cm^{-1} 3416(NH), 1683, 1731 (C=O).), 1025(C–O–C). ^{1}H NMR (DMSO-d_6) δ = 3.31(s, 3H, CH_3), 3.83 (s, 3H, OCH_3), 7.09–7.54(m, 14H, ArH), 10.37 (s, 2H, 2CHO), 12.79 (s, 2H, 2NH). MS *m/z* (%) 536(M$^+$, 0.1), 272 (38), 257 (100), 240 (30), 230 (10), 214 (26), 198 (14), 186 (18), 172 (18), 167 (17), 156 (15), 148 (43), 142 (15), 133 (10), 122 (15), 116 (17), 103 (25), 91 (64), 76 (53), 63 (35), 51 (46). Anal. Calcd for $C_{29}H_{24}N_6O_5$ (536.55) C, 64.92; H, 4.51; N, 15.66. Found: C, 65.13; H, 4.70; N, 15.82%.

3,4-Bis(4-phenylhydrazonopyrazol-3-yl)-1-phenyl-5-methyl-pyrazole (**4**): A mixture of compound **3a** (0.25 g, 0.5 mmol) and hydrazine hydrate (10 mL) in absolute ethanol was refluxed for 10 h and the reaction mixture was then cooled. The solid that precipitated was filtered off and crystallised from ethanol to give compound **4a** as an orange solid. Yield 0.99 g, (78 %), m.p. > 300 °C, IR (KBr) ν_{max}/cm^{-1} 3418 (NH), 1596 (C=N). ^{1}H NMR (DMSO-d_6) δ = 3.31 (s, 3H, CH_3), 7.60 (s, 15H, ArH), 7.83 (s, 2H, pyrazole-H), 9.92 (s, 2H, 2NH); MS *m/z* (%) 498 (M$^+$, 0.35), 301 (12), 274 (10), 238 (10), 140 (10), 118 (31), 104 (27), 92 (100), 82 (21), 76 (80), 65 (58), 50 (32). Anal. Calcd for $C_{28}H_{22}N_{10}$ (498.55) C, 67.46; H, 4.45; N, 28.09. Found: C, 67.75; H, 4.63; N, 28.27%.

1,4,5-Trimethyl-6-phenyl-6H-pyrazolo[3,4-d]-pyridazine (**6**): A mixture of the pyrazole derivative **1a** (0.25 g, 1 mmol) and hydrazine hydrate (10 mL) in absolute ethanol was refluxed for 10 h and the reaction mixture was cooled. The solid that precipitated was filtered off and crystallised from ethanol to give compound **6a** as white solid, 0.15 g, 63% yield, m.p. 254–256°C (EtOH) (lit.[16] m.p. 239–240 °C). IR (KBr) ν_{max}/cm^{-1} 1634 (C=N). ^{1}H NMR ($CDCl_3$): δ = 2.75 (s, 3H, CH_3), 2.89 (s, 6H, $2CH_3$), 7.49–7.63 (m, 5H, ArH). MS *m/z* (%): 238 (M$^+$, 100), 117 (13), 141 (12), 77 (45). Anal. Calcd for $C_{14}H_{14}N_4$ (238.29): C, 70.57; H, 5.92; N, 23.51. Found: C, 70.79; H, 6.02; N, 23.55%.

4,7-Bis(2-dimethylaminovinyl)-3-methyl-2-phenyl-2H-pyrazolo [3,4-d]pyridazine (**7**): A mixture of compound **6** (1.5 g, 6.3 mmol) and *N,N*-dimethylformamide dimethyl acetal (DMF-DMA) (4.0 g) was refluxed for 20 h then left to cool. Methanol was added to the cold mixture. The resulting solid was collected by filtration, washed with methanol, dried and finally crystallised from ethanol to afford the bis-enamine **7** as pale orange solid, (1.7 g, 78 % yield), m.p. 242–244 °C. ^{1}H NMR ($CDCl_3$) δ = 2.77 (s, 3H, CH_3), 2.87 [s, 6H, $N(CH_3)_2$], 2.93 [s, 6H, $N(CH_3)_2$], 5.86 (d, *J* = 13.0 Hz, 2H, =CH), 7.50–7.59 (m, 5H, ArH), 7.61 (d, *J* = 13.0 Hz, 2H, =CH); MS *m/z* (%): 348 (M$^+$, .74), 238

(32), 149 (22), 141 (11.26), 118 (28.31), 104 (32.7), 98 (20), 77 (45), 46 (100). Anal. Calcd for $C_{20}H_{24}N_6$ (348.45): C, 68.94; H, 6.94; N, 24.12. Found: C, 69.05; H, 7.10; N, 24.25%.

4,7-Bis[(2-oxo-1-phenylhydrazono)-3-methyl-2-phenyl-ethyl]-2H-pyrazolo[3,4-d]pyridazine (**5**): A solution of bis-enamine **7** (0.35 g, 1 mmol) in ethanol (20 mL) and sodium acetate trihydrate (0.5 g) was cooled in an ice bath at 0–5°C while being stirred. To the resulting cold solution was added portions of a cold solution of benzenediazonium chloride prepared by diazotising aniline (0.2 g, 2 mmol) in hydrochloric acid (6 M, 3 mL) with sodium nitrite (0.14 g, 2 mmol) in water (10 mL). After all the diazonium salt was added, the mixture was stirred for a further 30 min. while cooling in an ice-bath. The reaction mixture was then left in a refrigerator for three days. The solid that precipitated was filtered off, washed with water, dried and finally crystallised from the appropriate solvent to give the product **5a** as pale orange solid. Yield (1.0 g, 78 %), mp. > 300 °C, IR (KBr) ν_{max}/cm^{-1} 3428 (NH), 1603 (C=O). ^{1}H NMR (DMSO-d_6) δ = 3.32 (s, 3H, CH_3), 7.71 (s, 15H, ArH), 10.23 (s, 2H, 2CHO), 12.64 (s, 2H, 2NH); MS *m/z* (%) 502 (M$^+$, 2), 149 (58), 143 (11), 134 (11), 131 (14), 127 (11), 123 (15), 118 (30), 109 (27), 104 (40), 97 (34), 93 (50), 82 (45), 77 (83), 69 (60), 56 (100), 50 (44). Anal. Calcd for $C_{28}H_{22}N_8O_2$(502.54) C, 66.92; H, 4.41; N, 22.30. Found: C, 67.12; H, 4.63; N, 22.46%.

3,4-Bis{([1,2,4]triazolo[3,4-c][1,2,4]triazin-6-yl)carbonyl)}-1-aryl-5-methylpyrazoles (**8a–c**): A solution of bis-enaminones **2** (1.76 g, 5 mmole) in ethanol (20 mL) and sodium acetate trihydrate (0.5 g) was cooled in an ice bath at 0–5°C while being stirred. To the resulting cold solution was added portions of a cold solution of 3-diazo-1,2,4-triazole, prepared by diazotising 3-amino-1,2,4-triazole (10 mmole) in hydrochloric acid (6M, 3 mL) with sodium nitrite (0.70 g, 10 mmole) in water (5 mL). After all the diazonium salt solution was added, the mixture was stirred for further 30 minutes while cooling in an ice-bath. The reaction mixture was then left in a refrigerator for three days. The solid that precipitated was filtered off, washed with water, dried and finally crystallised from ethanol to give the respective products.

3,4-Bis{([1,2,4]triazolo[3,4-c][1,2,4]triazin-6-yl)carbonyl)}-1-phenyl-5-methylpyrazole (**8a**): Deep red solid, yield 1.71 g, (76 %), m.p. 122–124 °C, IR (KBr) ν_{max}/cm^{-1} 1665 (C=O), 1605 (C=N). ^{1}H NMR (DMSO-d_6) δ = 3.38 (s, 3H, CH_3), 7.45-7.62 (m, 5H, ArH), 7.87 (s, 2H, H-3), 8.58 (s, 2H, H-5). MS *m/z* (%) 452 (M$^+$, 0.42), 287 (18), 149 (33), 144 (11), 125 (35), 118 (17), 113 (44), 109 (24), 102 (37), 96 (94), 84 (100), 76 (52), 69 (61), 56 (93). Anal. Calcd for $C_{20}H_{12}N_{12}O_2$ (452.40) C, 53.10; H, 2.67; N, 37.15. Found: C, 53.20; H, 2.82; N, 37.30%.

3,4-Bis{([1,2,4]triazolo[3,4-c][1,2,4]triazin-6-yl)carbonyl)}-1-(4-methylphenyl)-5-methylpyrazole (**8b**): Orange solid, yield 1.86 g, (80 %), m.p. 156–158 °C, IR (KBr) ν_{max}/cm^{-1} 1681 (C=O). ^{1}H NMR (DMSO-d_6) δ = 2.43 (s, 3H, $ArCH_3$), 3.39 (s, 3H, CH_3), 7.37–7.44 (m, 4H, ArH), 7.96 (s, 2H, H-3), 8.57 (s, 2H, H-5). MS *m/z* (%) 456 (M$^+$, 0.12), 332 (20), 317 (10), 256 (30), 241 (87), 213 (14), 198 (34), 170 (26), 155 (19), 149 (10), 131 (33), 104 (61), 96 (32), 91 (64), 83 (100), 64 (57), 56 (36). Anal. Calcd for $C_{21}H_{14}N_{12}O_2$ (466.42) C, 54.08; H, 3.02; N, 36.04. Found: C, 54.15; H, 3.10; N, 36.25%.

3,4-Bis{([1,2,4]triazolo[3,4-c][1,2,4]triazin-6-yl)carbonyl)}-1-(4-methoxyphenyl)-5-methylpyrazole (**8c**): Yellow solid, yield 2.04 g, (85 %), m.p. 136–138 °C, IR (KBr) ν_{max}/cm^{-1} 1676 (C=O), 1608(C=N), 1251(C–O–C). ^{1}H NMR (DMSO-d_6) δ = 3.50 (s, 3H, CH_3), 3.83 (s, 3H, OCH_3), 7.11 (d, *J* = 8.0 Hz, 2H, ArH), 7.49 (d, *J* = 8.0 Hz, 2H, ArH), 7.53 (d, *J* = 8.0 Hz, 2H, ArH), 7.95 (s, 2H, H-3), 8.56 (d, *J* = 9.0 Hz, 2H, H-5). MS *m/z* (%) 482 (M$^+$, 0.48), 272 (41), 257 (100), 214 (16), 187 (11), 172 (13), 148 (55), 122 (32), 118 (10), 113 (32), 105 (31), 97 (70), 91 (48), 84 (60), 76 (89), 68 (55), 56 (90). Anal. Calcd for $C_{21}H_{14}N_{12}O_3$ (482.42) C, 52.28; H, 2.93; N, 34.84. Found: C, 52.45; H, 3.05; N, 35.02%.

Synthesis of 4,7-bis([1,2,4]triazolo[3,4-c][1,2,4]triazin-6-yl)-3-methyl-1-phenyl-pyrazolo[3,4-d]pyridazine (**9**): A mixture of compound **8a** (0.67 g, 1.5 mmol) and hydrazine hydrate (10 mL) in absolute ethanol was refluxed for 10 h and the reaction mixture was cooled. The solid that precipitated was filtered off and crystallised from (ethanol-dioxane) to give compound **9** as pale green solid. Yield (0.53 g, 80 %), m.p. 278–280 °C, IR (KBr) ν_{max}/cm^{-1} 1629 (C=N). ^{1}H NMR (DMSO-d_6) δ = 3.32 (s, 3H, CH_3), 7.65 (s, 5H, ArH), 7.92 (s, 2H, H-5), 8.88 (s, 2H, H-3). MS *m/z* (%) 448 (M$^+$, 0.24), 263 (77), 249 (12), 238 (23), 223 (14), 207 (15), 167 (15), 149 (12), 142 (16), 126 (16), 122 (11), 117 (37), 109 (13), 104 (52), 96 (62), 82 (100), 76 (90), 67 (80), 54 (64), 50 (51). Anal. Calcd for $C_{20}H_{12}N_{14}$ (448.41) C, 53.57; H, 2.70; N, 43.73. Found: C, 53.72; H, 2.83; N, 43.90%.

Synthesis of 4,7-bis([1,2,4]triazolo[5,1-c][1,2,4]triazin-3-yl)-3-methyl-1-phenylpyrazolo[3,4-d]pyridazine (**10**): A mixture of compound **9** (0.5 g, 1 mmol) and potassium hydroxide in absolute ethanol was refluxed for 3–5 h and the reaction mixture was cooled and poured into water. The solid that precipitated was filtered off and crystallised from (ethanol-dioxane) to give the title compound as brown solid. Yield (0.33 g, 75 %), m.p. > 300 °C, IR (KBr) ν_{max}/cm^{-1} 1629 (C=N). ^{1}H NMR (DMSO-d_6) δ = 3.25 (s, 3H, CH_3), 7.54 (s, 5H, ArH), 7.92 (s, 2H, H-4), 8.47 (s, 2H, H-7). MS *m/z* (%) 449 (M$^+$+1, 0.60), 368 (13), 263 (58), 237 (34), 223 (12), 207 (18), 193 (10), 165 (11), 151 (15), 141 (17), 135 (13), 123 (16), 117 (28), 110 (28), 105 (18), 97 (60), 83 (63), 76 (50), 68 (57), 54 (100). Anal. Calcd for $C_{20}H_{12}N_{14}$ (448.41) C, 53.57; H, 2.70; N, 43.73. Found: C, 53.72; H, 2.83; N, 43.90%.

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References

1 K.M. Al-Zaydi, E.A.A. Hafez and M.H. Elnagdi, *J. Chem. Res*., 2000, *(S)* 154, (M) 510.
2 K.M. Al-Zaydi and R.M. Borik, *Molecules*, 2007, **12**, 2061.
3 M.A. Al-Shiekh, H.Y. Medrassi, M.H. Elnagdi and E.A. Hafez, *Arkivoc*, 2008, **xvii**, 36.
4 A. Alnajjar; M.M. Abdelkhalik, A. Al-Enezi and M.H. Elnagdi, *Molecules*, 2009, **14**, 68.
5 A.S. Shawali, T.A. Farghaly, and A.R. Al-Dahshoury, *Arkivoc*, 2009, ***xiv***, 88.
6 A.S. Shawali, *J. Chem. Res*., 2010, 630.
7 A.S. Shawali, S.M. Sherif, M.A.A. Darwish and M.M. El-merzabani, *Arch. Pharm. Res.*, 2010, **33**, 55.
8 A.S. Shawali, S.M. Sherif, M.A.A. Darwish and M.M. El-merzabani, *J. Heterocycl. Chem.*, 2009, **46**, 548.
9 A.S. Shawali, T.A. Farghaly and A.R. Al-Dahshoury, *Arkivoc*, 2010, **ix**, 19.
10 A.S. Shawali and A.J.M. Habboub, *J. Heterocycl. Chem*., 2010, in press.
11 N.A. Al-Awadi, M.H. Elnagdi, Y.A. Ibrahim, K. Kaul and A. Kumar, *Tetrahedron*, 2001, **57**, 1609.
12 A.S. Shawali, H.M. Hassaneen and N. Kh. Shurrab, *J. Heterocycl. Chem*., 2008, **45**, 1825.
13 J. Daunis and M. Follett, *Bull. Soc. Chim. Fr.*, 1975, 857.
14 H. Lepez and G. Maury, *J. Org. Chem*., 1977, **42**, 1018.
15 E.S.H. Al-Ashry, E.N. Rashed and H. Assafir, *Adv. Heterocycl. Chem*., 2000, **75**, 79.
16 C.J. Shishoo, M.B. Devani, G.V. Ullas, S. Ananthan and V.S. Bhadti, *J. Heterocycl. Chem.*, 1981, **18**, 43.
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