Synthesis and tautomeric structure of novel 3-arylazo-imidazo[1,2-*b*]pyrazolo[4,3-*d*]pyridazines

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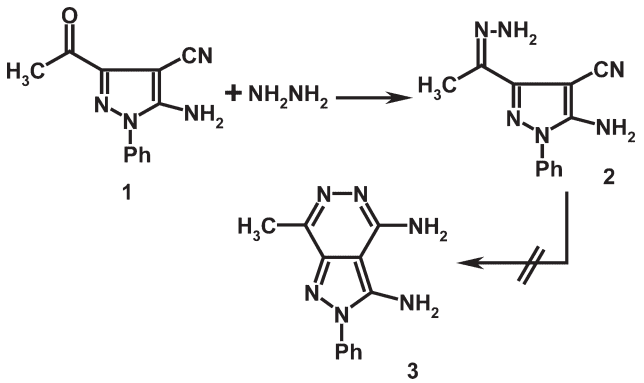
Treatment of 3-acetyl-5-amino-4-cyano-1-phenylpyrazole with hydrazine hydrate afforded the hydrazone derivative. Reaction of the latter hydrazone with hydrazonoyl chlorides was found to be site selective as it afforded the corresponding formazan and not the amidrazone derivatives. The ^{1}H NMR spectra of the formazan derivatives indicated that they exist as the bis-hydrazone tautomers. Acid-catalysed cyclisation of these dihydroformazans afforded the respective 9-amino-3-arylazo-6-methyl-8-phenyl-2-substituted-8*H*-imidazo[1,2-*b*]pyrazolo[4,3-*d*]pyridazines whose electronic absorption spectra revealed that they exist predominantly in the arylazo tautomeric form. The structures of all compounds prepared were confirmed by spectral and elemental analyses. Also the mechanism of the reactions studied are discussed.

Keywords: hydrazonoyl halides, amidrazones, hydrazones, formazans

In recent years, arylazo derivatives of various heterocyclic ring systems have received much attention from organic chemists and dye manufacturers.[1] In conjunction with our continuing interest in azo-hydrazone tautomerism of azo colouring materials[2] and the chemistry of hydrazonoyl halides,[3–13], we studied the utility of the latter for the synthesis of arylazo derivatives of a new triheterocyclic system namely imidazo[1,2-*b*]pyrazolo[4,3-*d*]pyridazine which has not been reported previously. In addition, our objective with such a study was to shed some light on the site-selectivity in the reactions to be studied and to investigate the tautomerism of the target colorants.

Results and discussion

The required precursors namely 3-acetyl-5-amino-4-cyano-1-phenyl-pyrazole **1** and hydrazonoyl halides **4a–h** were prepared as previously described from our laboratory.[14–16] Treatment of **1** with hydrazine hydrate in refluxing ethanol yielded a product that was identified as the hydrazone derivative **2** (Scheme 1). The possible cyclised product **3** was not formed under the reaction conditions employed (Scheme 1). This finding is consistent with other literature reports on the hydrazinolysis of 1,5-disubstituted-3-acetyl-4-cyanopyrazole derivatives.[17,18] The structure of the product **2** was assigned on the basis of its spectral data (IR, MS and ^{1}H NMR) and elemental analyses (see Experimental). For example, its IR spectrum exhibited characteristic bands at ν 3415, 3334, 3237, 3191 (2 NH_2) and 2211 (CN) cm^{-1}. The ^{1}H NMR spectrum of **2** revealed, in addition to aromatic proton signals, characteristic signals at δ 1.97 (s, 3H, CH_3), 6.46 (s, 2H, NH_2), 6.62 (s, 2H, NH_2).



Scheme 1 (a) *m*-CPMA/p-TsOH; (b) *p*-TsOH.

When compound **2** was treated with each of the hydrazonoyl halides **4** in 1,4-dioxane in the presence of triethylamine, it afforded, in each case, a single product that was identified as the respective formazan derivative **5** (Scheme 2). In all cases, the amidrazone derivatives **6** were not formed (Scheme 2). This finding indicates that the reaction is site specific. The formation of **5** rather than **6** can be attributed to the lower nucleophilicity of the 5-amino group that results from being conjugated with the electron withdrawing cyano group. This renders the amino-nitrogen of the hydrazone moiety more nucleophilic and in turn its attack on the carbon atom of the hydrazonoyl halide is more favourable to give **5** as the final product. The isolated products **5** can have either the bis-hydrazone structure **5** or its tautomeric structure **5A** (Scheme 2). However, their ^{1}H NMR spectra are consistent with the former tautomeric form **5**. For example, their ^{1}H NMR spectra revealed in each case, in addition to CH_3, NH_2 and aromatic protons signals, two common singlets in the regions δ 8.98–9.42 and 10.76–11.08 assignable to the two NH groups of structure **5**. On the basis of this finding, the tautomeric structure **5A** was discarded.

When each of the formazan derivatives **5** was heated in glacial acetic acid, they underwent dehydrative cyclisation and afforded the respective 9-amino-3-arylazo-6-methyl-8-phenyl-2-substituted-8*H*-imidazo[1,2-*b*]pyrazolo[4,3-*d*]pyridazines **8** (Scheme 3). The structures of the isolated products **8** were confirmed by their spectra (IR, MS and ^{1}H NMR) and by elemental analyses (see Experimental). For example, their IR spectra, while they revealed the absence of both the nitrile and carbonyl absorption bands present in the spectra of their precursors **5**, they showed instead two common bands in the regions 3410–3354 and 3390–3241 cm^{-1} due to the symmetric and asymmetric stretching vibrations of the –NH_2 group.

As the latter products **8** can have either the amino-azo structure **8**, or less likely, the imino-hydrazone structure **9** (Scheme 4), their electronic absorption spectra were also studied to shed some light on their actual tautomeric structure. Their visible spectra in dioxane revealed, in each case, two absorption bands in regions 430–488 and 325–375 nm (Table 1). Such an absorption pattern is similar to that reported for arylazo compounds.[18] Furthermore, the spectra of **8f**, taken as an example of the series prepared, in solvents of different polarities, revealed little, if any, change indicating that such compounds exist only in the depicted azo tautomeric form **8** (Scheme 3).[19]

Conclusions

A series of new formazan derivatives **5** were synthesised and cyclised into aryazoheterocycles **8**. The reactions leading to **5**

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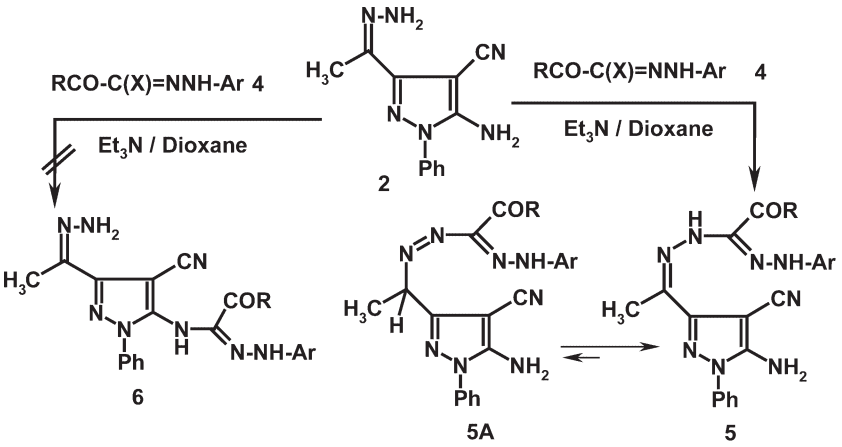
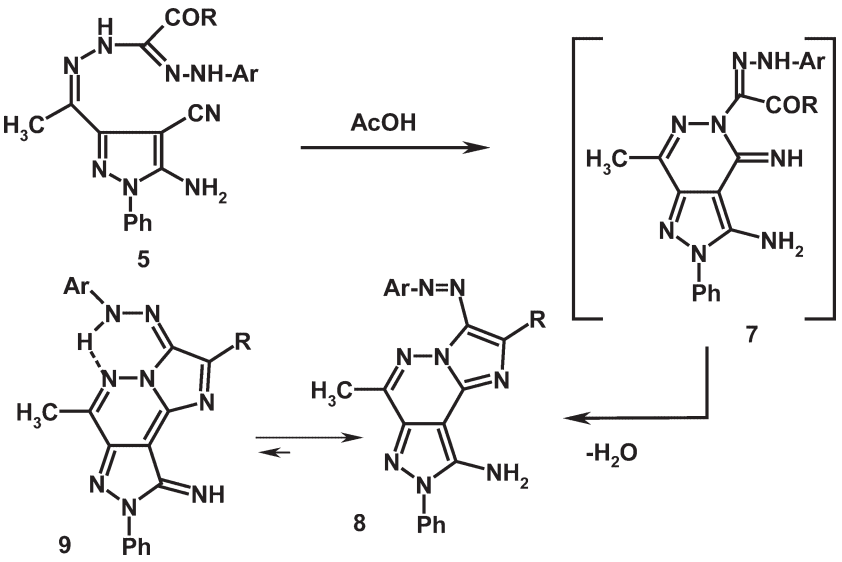


X = Cl or Br Ar = C_6H_4Y

R / Y: a, R = CH_3 , Y, 3-CH_3; b, R = CH_3 , Y= H; c, R = CH_3, Y= 4-Cl;
d, R = CH_3 , Y = 3-NO_2; e, R = CH_3 , Y = 4-CH_3; f, R = Ph ,Y = H;
g, R = 2-naphthyl , Y = H; h, R = 2-naphthyl , Y = 4-Cl

Scheme 2



Ar = C_6H_4Y

a, R =CH_3 , Y= 3-CH_3; c, R = CH_3 , Y, 4-Cl; d, R = CH_3 , Y= 3-NO_2;
e, R = CH_3 , Y = 4-CH_3; g, R = Ph , Y = H; h, R = 2-naphthyl , Y = H

Scheme 3

proved to be site-selective. Compounds **5** have a bis-hydrazone structure whereas compounds **8** exist predominantly as the azo tautomeric form.

Table 1 UV spectra of compounds **8** in 1,4-dioxane

Compd. No.	λ_{max} (log ε)
8a	430 (4.86), 351 (4.45)
8c	453 (3.88), 350 (3.54)
8d	487 (4.70), 375 (4.24)
8e	434 (4.71), 340 (3.59)
8f[a]	479 (4.62), 370 (3.78)
8g	488 (4.36), 325 (3.08)

[a] Solvent: λ_{max} (log ε): acetic acid 478 (4.39), 369 (3.59); chloroform 479 (4.51), 371 (3.78); DMF 480 (4.19), 372 (3.46); ethanol 479 (4.29), 368 (4.00).

Experimental

All melting points were determined on an Electrothermal apparatus and are uncorrected. Solvents were generally distilled and dried prior to their 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 spectrometer (300 MHz for ^{1}H NMR) and the chemical shifts were related to that of the solvent DMSO-d_6. The mass spectra were recorded on a GCMS-Q1000-EX Shimadzu and GCMS 5988-A HP spectrometers, the ionising voltage was 70 eV. Elemental analyses were carried out at the Microanalytical Centre of Cairo University, Giza, Egypt. 3-Acetyl-5-amino-4-cyano-1-phenylpyrazole **1** and the hydrazonoyl halides **2** were prepared as previously described in Literature.[14–16]

5-Amino-3-acetyl-4-cyano-1-phenylpyrazole hydrazone (**2**): A mixture of 3-acetyl-5-amino-4-cyano-1-phenylpyrazole (1) (2.5 g, 0.01 mol) and an excess of hydrazine hydrate (16 g, 0.5 mol) was refluxed for 25 h. The reaction mixture was cooled and the solid that precipitated was filtered off and crystallised from ethanol to give compound 2 as

yellow crystals, yield (1.97 g, 82%), m.p. 255 °C; IR (KBr) ν = 3415, 3334, 3237, 3191 ($2NH_2$), 2211 (CN) cm^{-1}; ^1H NMR (DMSO-d_6) δ 1.97 (s, 3H, CH_3), 6.46 (s, 2H, NH_2), 6.62 (s, 2H, NH_2), 7.39–7.53 (m, 5H, ArH) ppm; MS, *m/z* (%) 240 (M^+, 100), 210 (25), 184 (4), 155 (14), 134 (4), 103 (13), 77(46). Anal. Calcd for $C_{12}H_{12}N_6$ (240.26): C,n59.99; H, 5.03; N, 34.98. Found: C, 60.32; H, 5.23, N, 35.06%.

Reaction of compound ***2*** *with hydrazonoyl halides* **4**

To a mixture of **2** (0.6 g, 2.5 mmol) and the appropriate hydrazonoyl halide **4** (2.5 mmol) in 1,4-dioxane (20 mL) was added triethylamine (0.35 mL) and the mixture was refluxed for 15 h, then cooled. The solution was poured onto ice and concentrated hydrochloric acid. The solid that produced was filtered off and crystallised from the appropriate solvent to give the corresponding compound **5**. The products **5a–h** prepared together with their physical constants are listed below.

3-Acetyl-5-(m-tolyl)-1-[5-amino-4-cyano-1-phenylpyrazol-3-yl)-ethylidene]-1,2-dihydroformazan (**5a**): Yellow solid, yield (0.77 g, 74%), m.p. 250 °C; (ethanol); IR (KBr) ν 3411, 3330 (NH_2), 3232, 3156 (2NH), 2207 (CN), 1668 (CO) cm^{-1}; ^1H NMR (DMSO-d_6) δ 2.27 (s, 3H, CH_3), 2.28 (s, 3H, CH_3), 2.49 (s, 3H, $COCH_3$), 6.83 (s, 2H, NH_2), 7.15–7.57 (m, 9H, ArH), 8.99 (s, 1H, NH), 10.77 (s, 1H, NH) ppm; MS, *m/z* (%) 414 (M^+, 87), 399 (68), 367 (11), 295 (15), 240 (13), 226 (76), 209 (28), 118 (48), 105 (78), 91 (100). Anal. Calcd for $C_{22}H_{22}N_8O$ (414.46): C,63.75; H, 5.35; N, 27.04. Found: C, 63.54; H, 5.39; N, 27.08%.

3-Acetyl-5-phenyl-1-[5-amino-4-cyano-1-phenylpyrazol-3-yl)-ethylidene]-1,2-dihydroformazan (**5b**): Yellow crystals, yield (0.75 g, 75%), m.p. 230 °C; (ethanol); IR (KBr) ν 3403, 3323 (NH_2), 3230 (NH), 3047 (NH), 2212 (CN), 1667 (CO) cm^{-1}; ^1H NMR (DMSO-d_6) δ 2.27 (s, 3H, CH_3), 2.42 (s, 3H, $COCH_3$), 6.84 (s, 2H, NH_2), 7.23–7.57 (m, 10H, ArH), 9.01 (s, 1H, NH), 10.83 (s, 1H, NH) ppm; MS, *m/z* (%) 400 ($M^{+\cdot}$ 95), 384 (53), 175 (17), 141 (15), 119 (28), 104 (39), 77 (100). Anal. Calcd for $C_{21}H_{20}N_8O$ (400.44): C, 62.99; H, 5.03; N, 27.98. Found: C, 63.04; H, 5.25; N, 27.88%.

3-Acetyl-5-(p-chlorophenyl)-1-[5-amino-4-cyano-1-phenyl-pyrazol-3-yl)ethylidene]-1,2-dihydroformazan (**5c**): Pale brown crystals, yield (0.87 g, 80%), m.p. 220 °C; (ethanol); IR (KBr) ν 3403 (NH_2), 3327, 3232 (2NH), 2211 (CN), 1668 (CO) cm^{-1}; ^1H NMR (DMSO-d_6) δ 2.27 (s, 3H, CH_3), 2.49 (s, 3H, $COCH_3$), 6.86 (s, 2H, NH_2), 7.26 (d, *J* = 9 *Hz*, 2H, ArH), 7.36 (d, *J* = 9 *Hz*, 2H, ArH), 7.47–7.56 (m, 5H, ArH), 9.03 (s, 1H, NH), 10.83 (s, 1H, NH) ppm; MS, *m/z* (%) 436 (M^++2, 29), 434 (M^+, 64), 419 (53), 240 (16), 226 (100), 210 (62), 184 (33), 141 (40), 111 (42), 77 (77). Anal. Calcd for $C_{21}H_{19}ClN_8O$ (434.88): C, 58.00; H, 4.40; N, 25.77. Found: C, 58.21; H, 4.30; N, 25.89%.

3-Acetyl-5-(m-nitrophenyl)-1-[5-amino-4-cyano-1-phenyl-pyrazol-3-yl)ethylidene]-1,2-dihydroformazan (**5d**): Green crystals, yield (0.87 g, 78%), m.p 236 °C; (ethanol); IR (KBr) ν 3420, 3331 (NH_2), 3210, 3150 (2NH), 2211 (CN), 1674 (CO) cm^{-1}; ^1H NMR (DMSO-d_6) δ 2.29 (s, 3H, CH_3), 2.49 (s, 3H, $COCH_3$), 6.89 (s, 2H, NH_2), 7.46–8.16 (m, 9H, ArH), 9.08 (s, 1H, NH), 10.98 (s, 1H, NH) ppm; MS, *m/z* (%) 445 (M^+, 73), 427 (47), 295 (14), 225 (100), 209 (65), 184 (66), 156 (33), 118 (60), 91 (64), 76 (91). *Anal*. Calcd for $C_{21}H_{19}N_9O_3$ (445.43): C, 56.62; H, 4.30; N, 28.30. Found: C, 56.45; H, 4.22; N, 28.24%.

3-Acetyl-5-(p-tolyl)-1-[5-amino-4-cyano-1-phenylpyrazol-3-yl)-ethylidene]-1,2-dihydroformazan (**5e**): Golden yellow crystals, yield (0.79 g, 76%), m.p. 226-228 °C; (ethanol); IR (KBr) ν 3412, 3381 (NH_2), 3329 (NH), 3231 (NH), 2212 (CN), 1620 (CO) cm^{-1}; ^1H NMR (DMSO-d_6) δ 2.24 (s, 3H, CH_3), 2.26 (s, 3H, CH_3), 2.46 (s, 3H, $COCH_3$), 6.84 (s, 2H, NH_2), 7.05 (d, *J* = 9 Hz, 2H, ArH), 7.26 (d, *J* = 9 *Hz*, 2H, ArH), 7.47–7.56 (m, 5H, ArH), 8.98 (s, 1H, NH), 10.76 (s, 1H, NH) ppm; MS, *m/z* (%) 414 (M^+, 80), 399 (55), 240 (14), 226 (74), 190 (20), 119 (56), 90 (100), 77 (59). Anal. Calcd for $C_{22}H_{22}N_8O$ (414.46): C,63.75; H, 5.35; N, 27.04. Found: C, 63.84, H, 5.21; N, 27.00%.

3-Benzoyl-5-phenyl-1-[5-amino-4-cyano-1-phenylpyrazol-3-yl)-ethylidene]-1,2-dihydroformazan (**5f**): Yellow crystals, yield (0.92 g, 80%), m.p. 175 °C; (ethanol); IR (KBr) ν 3436, 3340 (NH_2), 3302, 3215 (2NH), 2213 (CN), 1659 (CO) cm^{-1}; ^1H NMR (DMSO-d_6) δ 2.34 (s, 3H, CH_3), 6.82 (s, 2H, NH_2), 7.17–8.42 (m, 15H, ArH), 9.29 (s, 1H, NH), 11.04 (s, 1H, NH) ppm; MS, *m/z* (%) 462 (M^+, 1), 301 (15),131 (10), 105 (29), 77 (100). Anal. Calcd for $C_{26}H_{22}N_8O$ (462.51): C, 67.52; H, 4.79; N, 24.23. Found: C, 67.29; H, 4.67; N, 24.18%.

3-(2-Naphthoyl)-5-phenyl-1-[5-amino-4-cyano-1-phenylpyrazol-3-yl)ethylidene]-1,2-dihydroformazan (**5g**)**:** Green crystals, yield (0.97 g, 76%), m.p. 180 °C; (ethanol/dioxan); IR (KBr) ν 3429, 3312 (NH_2), 3250, 3049 (2NH), 2210 (CN), 1635 (CO) cm^{-1}; ^1H NMR (DMSO-d_6) δ 2.36 (s, 3H, CH_3), 6.82 (s, 2H, NH_2), 7.16–8.13 (m, 16H, ArH), 8.69 (s, 1H, naphthoyl-H), 9.37 (s, 1H, NH), 11.08 (s, 1H, NH) ppm; MS, *m/z* (%) 512 (M^+, 2), 494 (39), 405 (59), 153 (39), 149 (10), 143 (4), 126 (28), 82 (100), 76 (96). *Anal*. Calcd for $C_{30}H_{24}N_8O$ (512.56): C, 70.30; H, 4.72; N, 21.86. Found: C, 70.11; H, 4.66; N,21.57%.

3-(2-Naphthoyl)-5-(p-chlorophenyl)-1-[5-amino-4-cyano-1-phenyl-pyrazol-3-yl)ethylidene]-1,2-dihydroformazan (**5h**): Brown crystals, yield (0.96 g, 70%), m.p. 188 °C; (ethanol); IR (KBr) ν 3426, 3321 (NH_2), 3219, 3055 (2NH), 2213 (CN), 1626 (CO) cm^{-1}; ^1H NMR (DMSO-d_6) δ 2.37 (s, 3H, CH_3), 6.75 (s, 2H, NH_2), 6.87 (d, *J* = 9 *Hz*, 2H, ArH), 7.08 (d, *J* = 9 Hz, 2H, ArH), 7.45–7.79 (m, 11H, ArH), 8.04 (s, 1H, naphthoyl-H), 9.42 (s, 1H, NH), 11.00 (s, 1H, NH) ppm; MS, *m/z* (%) 547 (M^+, 1), 527 (32), 447 (50), 433 (79), 238 (12, 209 (18), 126 (18), 91 (19), 77 (100). Anal. Calcd for $C_{30}H_{23}N_8OCl$ (547.01): C, 65.87; H, 4.24; N, 20.48. Found: C, 65.90; H, 4.33; N, 20.40%.

*Synthesis of 2,6,8-trisubstituted-9-amino-3-arylazo-imidazo[1,2-b] pyrazolo-[4,3-d]pyridazines (***8***)*

A solution of the appropriate formazan **5** (1 mmol) in glacial acetic acid (10 mL) and two drops of HCl was refluxed for 50–60 h (monitored by TLC) then cooled. The cold reaction mixture was poured onto ice cold solution of sodium acetate. The precipitate formed was filtered off and crystallised from the appropriate solvent to give the respective imidazo[1,2-*b*]pyrazolo[4,3-*d*]pyridazine **8**. The compounds **8** together with physical constants are listed below.

*2,6-Dimethyl-3-[m-tolylazo]-8-phenyl-9-amino-imidazo[1,2-*b*] pyrazolo[4,3-*d*]pyridazine* (**8a**): Dark red solid, yield (0.19 g, 47%), m.p. 294–298 °C; (ethanol/1,4-dioxane); IR (KBr) ν 3354, 3241 (NH_2) cm^{-1}; ^1H NMR ($CDCl_3$) δ 2.32 (s, 3H, CH_3), 2.35 (s, 3H, CH_3), 2.58 (s, 3H, CH_3), 6.65 (s, 2H, NH_2), 7.22–7.58 (m, 9H, ArH) ppm; MS, *m/z* (%) 396 (M^+, 23), 367 (29), 226 (35), 190 (46), 142 (19), 132 (20), 119 (55), 91 (100), 77 (97). Anal. Calcd for $C_{22}H_{20}N_8$ (396.45): C,66.65; H, 5.08; N, 28.26. Found: C, 66.43, H, 5.19; N, 28.02%.

*2,6-Dimethyl-3-[p-chlorophenylazo]-8-phenyl-9-amino-imidazo [1,2-*b*]pyrazolo[4,3-*d*]pyridazine* (**8c**): Pale brown solid, yield (0.16 g, 38%), m.p. > 300 °C; (1,4-dioxane); IR (KBr) ν 3384, 3310 (NH_2) cm^{-1}; ^1H NMR (DMSO-d_6) δ 2.27 (s, 3H, CH_3), 2.49 (s, 3H, CH_3), 6.80 (s, 2H, NH_2), 7.27 (d, *J* = 9 *Hz*, 2H, ArH), 7.30 (d, *J* = 9 *Hz*, 2H, ArH), 7.36–7.56 (m, 5H, ArH) ppm; MS, *m/z* (%) 418 (M^++2, 19), 416 (M^+, 27), 337 (25), 335 (42), 320 (46), 226 (33), 209 (22), 152 (25), 127 (44), 111 (61), 91 (28), 77 (100). Anal. Calcd for $C_{21}H_{17}ClN_8$ (416.87): C, 60.50; H, 4.11; N, 26.88. Found: C, 60.45; H, 4.23; N, 26.78%.

*2,6-Dimethyl-3-[m-nitrophenylazo]-8-phenyl-9-amino-imidazo [1,2-*b*]pyrazolo[4,3-*d*]pyridazine* (**8d**): Brown solid, yield (0.15 g, 36%), m.p 280–282 °C; (ethanol); IR (KBr) ν 3400, 3390 (NH_2) cm^{-1}; ^1H NMR (DMSO-d_6) δ 2.29 (s, 3H, CH_3), 2.54 (s, 3H, CH_3), 6.92 (s, 2H, NH_2), 7.36–8.15 (m, 9H, ArH) ppm; MS, *m/z* (%) 427 (M^+, 34), 346 (54), 331 (66), 226 (23), 224 (31), 163 (29), 119 (37), 92 (48), 85 (39), 77 (100). Anal. Calcd for $C_{21}H_{17}N_9O_2$ (427.42): C, 59.01; H, 4.01; N, 29.49. Found: C, 59.27; H, 4.05; N, 29.34%.

*2,6-Dimethyl-3-[p-tolylazo]-8-phenyl-9-amino-imidazo[1,2-*b*]-pyrazolo[4,3-*d*]pyridazine* (**8e**): Dark red solid, yield (0.17 g, 42%), m.p. > 300 °C; (ethanol / 1,4-dioxane); IR (KBr) ν 3410, 3336 (NH_2) cm^{-1}; ^1H NMR ($CDCl_3$) δ 2.27 (s, 3H, CH_3), 2.29 (s, 3H, CH_3), 2.57 (s, 3H, CH_3), 6.65 (s, 2H, NH_2), 7.08 (d, *J* = 8 *Hz*, 2H, ArH), 7.27 (d, *J* = 8 *Hz*, 2H, ArH), 7.35–7.85 (m, 5H, ArH) ppm; MS, *m/z* (%) 396 (M^+, 10), 226 (30), 121 (22), 119 (34), 106 (22), 91 (100), 77 (70). Anal. Calcd for $C_{22}H_{20}N_8$ (396.45): C,66.65; H, 5.08; N, 28.26. Found: C, 66.51, H, 5.30; N, 28.14%.

*2,8-Diphenyl-6-methyl-3-phenylazo-9-amino-imidazo[1,2-*b*] pyrazolo[4,3-*d*]pyridazine* (**8f**): Dark red solid, yield (0.14 g, 31%), m.p. > 300 °C; (DMF); IR (KBr) ν 3380, 3356 (NH_2) cm^{-1}; ^1H NMR (DMSO-d_6) δ 2.25 (s, 3H, CH_3), 6.65 (s, 2H, NH_2), 7.34–8.02 (m, 15H, ArH) ppm; MS, *m/z* (%) 444 (M^+, 9), 105 (54), 91 (84), 77 (100). Anal. Calcd for $C_{26}H_{20}N_8$ (444.49): C, 70.26; H, 4.54; N, 25.21. Found: C, 70.39; H, 4.44; N, 25.08%.

*2-(2-Naphthyl)-6-methyl-3-[phenylazo]-8-phenyl-9-amino-imidazo [1,2-*b*]pyrazolo[4,3-*d*]pyridazine* (**8g**): Brown solid, yield (0.19 g, 39%), m.p. 276–278 °C; (ethanol/1,4-dioxane); IR (KBr) ν 3405, 3335 (NH_2) cm^{-1}; ^1H NMR (DMSO-d_6) δ 2.29 (s, 3H, CH_3), 6.72 (s, 2H, NH_2), 7.29–8.04 (m, 16H, ArH), 8.58 (s, 1H, naphthyl-H) ppm; MS, *m/z* (%) 494 (M^+, 62), 127 (39), 105 (29), 91 (64), 77 (100). Anal. Calcd for $C_{30}H_{22}N_8$ (494.55): C, 72.86; H, 4.48; N, 22.66. Found: C, 72.95; H, 4.68; N,22.42%.

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