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SYNTHESIS OF NEW SUBSTITUTED PYRIDOPYRAZOLOTRIAZINES

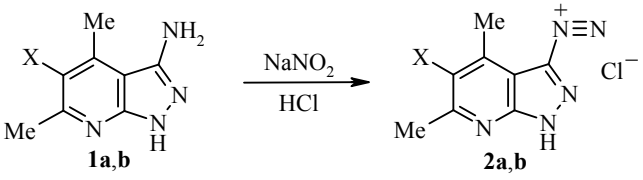
M. A. Metwally[1]*, E. Abdel-Galil[1], A. Metwally[1], and F. A. Amer[1]

Pyrazolo[3,4-b]pyridinediazonium chlorides react with a variety of active methylene-containing reagents (e.g., cyanoacetic acid arylidenehydrazide derivatives) to afford the corresponding 3-hydrazonopyrazolo[3,4-b]pyridine derivatives. The diazonium chlorides react with N'-acyl-2-cyanoacetohydrazide derivatives to give the corresponding 3-hydrazonopyrazolopyridine derivatives. The latter affords the corresponding 3-hydrazonopyrazolo[3,4-b]pyridine derivatives on reflux in acetic acid. Diazo coupling of 2,4-dimethylpyrazolo[3,4-b]pyridinediazonium chloride with ketoester, e.g., ethyl benzoylacetate, is followed by cyclization to afford pyrido[2',3':3,4]pyrazolo[5,1-c]triazine derivative. Diazo coupling of the same dimethylpyrazolo[3,4-b]pyridinediazonium chloride with unsymmetrical β-diketone, e.g., benzoylacetone, was also studied to afford the corresponding hydrazono derivative, which undergoes in situ cyclization to furnish the pyrido[2',3':3,4]pyrazolo[5,1-c]triazine derivative.

Keywords: 3-hydrazonopyrazolo[3,4-*b*]pyridines, pyrazolo[3,4-*b*]pyridinyldiazonium chlorides, pyrido[2',3':3,4]pyrazolo[5,1-*c*]triazines.

Due to the reported biological activities of triazines [1-5], as well as pyrazolopyridines [6], we were also interested in the synthesis of molecules with the pyrido[2',3':3,4]pyrazolo[5,1-*c*]triazine heterocyclic system. The latter has been used as disperse dyes (for polyester, polyamide, acrylic fibers) [7, 8], as antibacterial agents [9], and as fungicides [10].

The highly versatile 3-aminopyrazolo[3,4-*b*]pyridine derivatives **1a**,**b** have been reacted with sodium nitrite and hydrochloric acid to afford the corresponding pyrazolo[3,4-*b*]pyridinediazonium chlorides **2a**,**b**, respectively. Their versatility derives from the easy accessibility of compounds **1a**,**b**, which can be obtained from acetylacetone, hydrazine, and cyanoacetamide [11, 12]. The synthetic potentiality of compounds **2a**,**b** was investigated through their reactions with a variety of active methylene-containing reagents.



1a,**b** **2a**,**b**

1, 2 a X = H, **b** X = Br

*To whom correspondence should be addressed, e-mail: mamegs@mans.edu.eg.

[1]Chemistry Department, Faculty of Science, Mansoura University, P. O. Box 23, Mansoura, Egypt.

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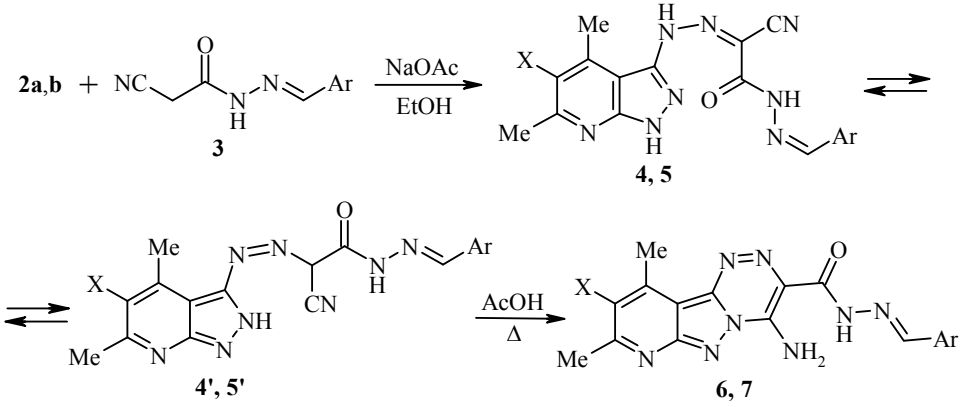
The pyrazolopyridinediazonium chlorides **2a**,**b** have been reacted with 2-cyano-*N*'-(4-methoxybenzylidene)acetohydrazide (**3**) to afford the corresponding 3-hydrazonopyrazolo[3,4-*b*]pyridine derivatives **4** and **5**, respectively.

The structure of compounds **4** and **5** is confirmed by IR and ^{1}H NMR spectroscopy, as well as by elemental analysis. The ^{1}H NMR spectra of compounds **4** and **5** include, in addition to the signals of both pyrazolopyridine and cyanoacetic arylidenehydrazide fragments, two singlet signals at 10.80 and 12.25 ppm for the three NH protons.

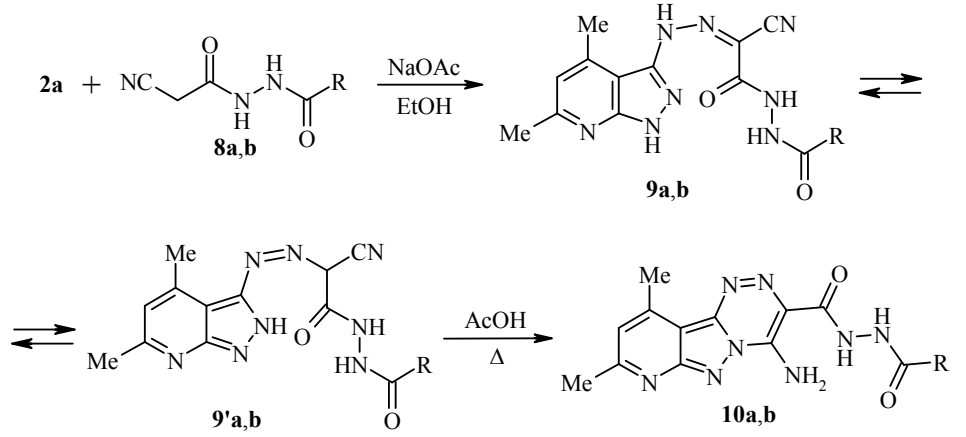


3, **4**, **4'**, **5**, **5'**, **6**, **7** Ar = 4-MeOC$_6$H$_4$; **4**, **4'**, **6** X = H; **5**, **5'**, **7** X = Br

Further confirmation of the hydrazono structures **4** and **5** was given *via* their cyclization in boiling acetic acid to give the corresponding pyrido[2',3':3,4]pyrazolo[5,1-*c*]triazine derivatives **6** and **7**, respectively. The reaction proceeds by nucleophilic attack of ring nitrogen to the cyano group in the tautomeric structures **4'** and **5'**.

The chemical structures of compounds **6** and **7** were confirmed by analytical and spectroscopic data. The IR spectra of these derivatives clearly indicated the lack of the cyano group absorption band and revealed the characteristic NH_2 and NH group absorption bands in the region of 3380-3200 cm^{-1}, in addition to the carbonyl absorption band at 1652-1661 cm^{-1}. The ^{1}H NMR and mass spectra of compounds **6** and **7** also confirm the proposed structure.

Furthermore, the diazonium chloride **2a** was reacted with *N*'-acyl-2-cyanoacetohydrazide derivatives **8a**,**b** to give the corresponding 3-hydrazonopyrazolopyridine derivatives **9a**,**b**. The structures of compounds **9a**,**b** were assigned on the basis of elemental analysis, IR, and ^{1}H NMR spectral data.

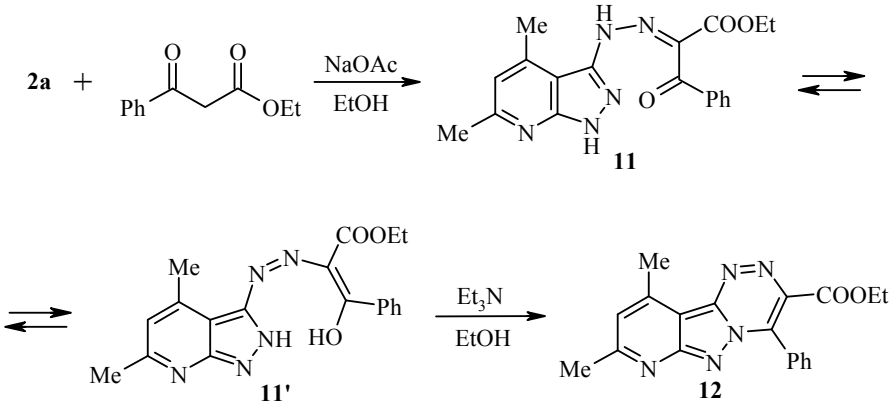


8, **9**, **9'**, **10 a** R = Me, **b** R = Ph

The IR spectrum of compound **9a** (as an example) showed characteristic absorption bands at 3403, 3282, and 3201 cm^{-1} due to NH groups, 2192 cm^{-1} for the cyano group, and 1698 and 1657 cm^{-1} for the carbonyl groups. The ^1H NMR spectrum of the same compound revealed three singlet signals at 2.10, 2.50, and 2.60 ppm for three methyl groups, a singlet signal at 7.00 ppm for the H-5 proton in addition to three singlet signals at 11.60, 12.85, and 14.25 ppm for the NH protons

When the hydrazono derivatives **9a**,**b** were refluxed in acetic acid, they underwent cyclization to afford the corresponding aminopyrido[2',3':3,4]pyrazolo[5,1-*c*]triazine derivatives **10a**,**b** respectively. The structures of the products were established based on the data of IR, ^1H NMR, MS, and elemental analyses. As in the case of cyclization of cyano derivatives **4**, **5** to compounds **6**, **7**, the IR spectrum of compounds **10a**,**b** showed the absence of the nitrile absorption band and the presence of absorption bands of the NH_2 and NH groups near 3380 and 3180 cm^{-1}.

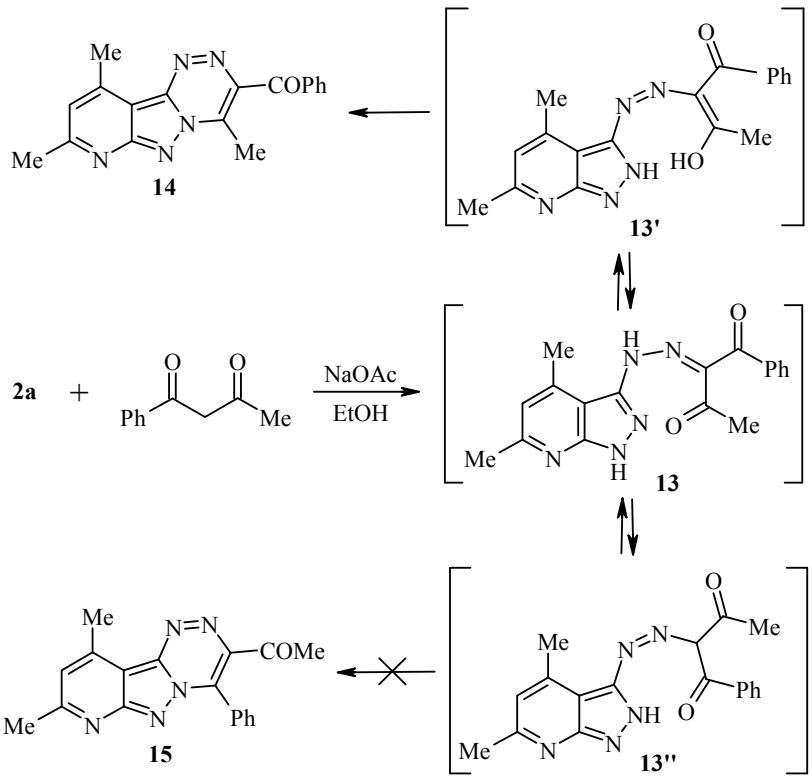
Diazo coupling of the dimethylpyrazolo[3,4-*b*]pyridinediazonium chloride **2a** with a ketoester ethyl benzoylacetate proceeded in ethanol solution of sodium acetate to furnish the corresponding hydrazono derivative **11** which underwent cyclization in ethanol containing a catalytic amount of triethylamine to afford the corresponding pyrido[2',3':3,4]pyrazolo[5,1-*c*]triazine derivative **12**. The structures of the products **11** and **12** were confirmed based on the spectral and elemental analysis data.



The IR spectrum of compound **11** showed three bands at 3147, 1685, and 1667 cm^{-1} assigned to the stretching frequency of the NH group and carbonyl groups of ester and phenyl ketone, respectively. The IR spectrum of the corresponding cyclized derivative **12** indicated the absence of the carbonyl absorption frequency (COPh) and displayed only the absorption band at 1737 cm^{-1} for the ester carbonyl group. The elemental analysis and mass spectrum of compound **12** confirm molecular composition and molecular mass of the proposed structure, indicating the loss of H_2O molecule with respect to the molecular composition of compound **11**.

The diazo coupling of dimethylpyrazolo[3,4-*b*]pyridinediazonium chloride **2a** with an unsymmetrical β-diketone benzoylacetone was also studied. The diazo-coupling reaction proceeds upon stirring at 0-5°C in ethanol solution of sodium acetate to afford the corresponding hydrazono derivative **13**, which undergoes *in situ* cyclization under the reaction conditions to furnish the pyrido[2',3':3,4]pyrazolo[5,1-*c*]triazine derivative **14** carrying the benzoyl group as substituent, instead of the other possible cyclization into the tricyclic system **15** carrying the acetyl group as substituent. Thus, it was suggested that the cyclization proceeds through water elimination by the tautomeric form **13'** rather than the other tautomeric from **13''**. The structure of the isolated product **14** is based on the spectral and elemental analysis data.

The IR spectrum of product **14** displayed the carbonyl absorption frequency at 1664 cm^{-1} assigned to benzoyl carbonyl (COPh). The mass spectrum showed the molecular ion peak at *m*/*z* 316 ($C_{18}H_{15}N_5O$); a fragment ion at *m*/*z* 105, corresponding to the benzoyl group, was also observed, providing additional confirmation of the structure **14**.



In conclusion, the synthesis of a new substituted pyridopyrazolotriazine from the reaction of pyrazolo-[3,4-*b*]pyridinyldiazonium chlorides with a variety of active methylene-containing compounds (*e g.*, cyanoacetic acid arylidene hydrazide derivatives, *N*'-acyl-2-cyanoacetohydrazide derivatives, β-ketoester and β-diketone) was described.

EXPERIMENTAL

IR spectra in KBr pellets were recorded with on a Mattson 5000 FTIR spectrometer. ^{1}H NMR spectra were measured on a Bruker WP 300 spectrometer (300 MHz), using TMS as internal standard. Mass spectra were recorded on a Finnigan MAT 212 instrument, using the electron impact ionization method. Elemental analyses were carried out in the Microanalytical Unit, Faculty of Science, University of Mansoura. Melting points (uncorrected) were determined on a Weiss-Gallenkamp electric melting point apparatus.

Starting compounds **1a**,**b** were prepared by method [11, 12]. Cyanoacetic acid hydrazides **3** and **8a**,**b** were obtained by the method described in [13-15].

Pyrazolopyridinediazonium Chlorides 2a,b (General Method). A solution of $NaNO_2$ (0.69 g, 0.01 mol) was added dropwise with stirring during 15 min to an ice-cooled sample of compound **1a** or **1b** (0.01 mol) at 0-5°C, dissolved in concentrated HCl (3 ml) and water (2 ml). After the addition, the stirring was continued for a further 15 min, and then the solution was used in further reactions.

3-Hydrazonopyrazolo[3,4-*b*]pyridine Derivatives 4 and 5 (General Method). NaOAc (about 4 g, 0.05 mol) was added to a cold solution of 2-cyano-*N*'-(4-methoxybenzylidene)acetohydrazide (**3**) (0.01 mol) in EtOH (50 ml). The solution was stirred and then the solution of diazonium salt **2a** or **2b** (0.01 mol) was added dropwise with stirring. Stirring was continued for 2 h. The products so formed were collected by filtration and washed with water, followed by cold EtOH. The isolated compounds were recrystallized from EtOH to give the corresponding 3-hydrazonopyrazolo[3,4-*b*]pyridine derivatives **4** or **5**.

2-Cyano-2-[(4,6-dimethyl-1*H*-pyrazolo[3,4-*b*]pyridin-3-yl)hydrazono]-*N*'-(4-methoxybenzylidene)-acetohydrazide (4). Yield 2.89 g (74%); mp 257-258°C. IR spectrum, ν, cm^{-1}: 3314, 3163 (2NH), 2180 (CN), 1650 (CO). ^{1}H NMR spectrum (DMSO-d_6), δ, ppm (*J*, Hz): 2.40 (3H, s, CH_3); 2.55 (3H, s, CH_3); 3.90 (3H, s, OCH_3); 6.90 (2H, d, *J* = 8.2, H Ar); 7.20 (1H, s, H-5); 7.40 (2H, d, *J* = 8.2, H Ar); 8.20 (1H, s, N=CH); 10.80 (1H, s, NH); 12.25 (2H, s, 2NH). Found, %: C 58.37; H 4.71; N 28.66. $C_{19}H_{18}N_8O_2$. Calculated, %: C 58.45; H 4.65; N 28.70.

2-[(5-Bromo-4,6-dimethyl-1*H*-pyrazolo[3,4-*b*]pyridin-3-yl)hydrazono]-2-cyano-*N*'-(4-methoxybenzylidene)acetohydrazide (5). Yield 3.19 g (68%); mp 258-259°C. IR spectrum, ν, cm^{-1}: 3325, 3214 (2NH), 2184 (CN), 1668 (CO). ^{1}H NMR spectrum (DMSO-d_6), δ, ppm (*J*, Hz): 2.40 (3H, s, CH_3); 2.50 (3H, s, CH_3); 3.85 (3H, s, OCH_3); 7.00 (2H, d, *J* = 8.0, H Ar); 7.40 (2H, d, *J* = 8.0, H Ar); 8.20 (1H, s, N=CH); 11.05 (1H, s, NH); 12.75 (2H, s, 2NH). Found, %: C 48.68; H 3.61; N 23.96. $C_{19}H_{17}BrN_8O_2$. Calculated, %: C 48.63; H 3.65; N 23.88.

8-Aminopyrido[2',3':3,4]pyrazolo[5,1-*c*][1,2,4]triazine-7-benzylideneacetohydrazide Derivatives 6 and 7 (General Method). Compound **4** or **5** (0.005 mol) in acetic acid (20 ml) was heated under reflux for 30 min, and then allowed to cool. The solid product **6** or **7** was collected by filtration, washed with cold EtOH, and recrystallized from EtOH–DMF mixture (1:1).

8-Amino-2,4-dimethylpyrido[2',3':3,4]pyrazolo[5,1-*c*][1,2,4]triazine-7-(4-methoxybenzylidene)-acetohydrazide (6). Yield 1.78 g (91%); mp > 300°C. IR spectrum, ν, cm^{-1}: 3359, 3252, 3205 (NH_2 and NH), 1660 (CO). ^{1}H NMR spectrum ($CDCl_3$–CF_3COOD), δ, ppm (*J*, Hz): 2.90 (3H, s, CH_3); 3.10 (3H, s, CH_3); 3.85 (3H, s, OCH_3); 7.00 (2H, d, *J* = 8.4, H Ar); 7.10 (1H, s, H-3); 7.40 (2H, d, *J* = 8.4, H Ar); 8.25 (1H, s, N=CH). Found, %: C 58.54; H 4.70; N 28.78. $C_{19}H_{18}N_8O_2$. Calculated, %: C 58.45; H 4.65; N 28.70.

8-Amino-3-bromo-2,4-dimethylpyrido[2',3':3,4]pyrazolo[5,1-*c*][1,2,4]triazine-7-(4-methoxybenzylidene)acetohydrazide (7). Yield 2.06 g (88%); mp > 300°C. IR spectrum, ν, cm^{-1}: 3386, 3277, 3204 (NH_2 and NH), 1661 (CO). ^{1}H NMR spectrum ($CDCl_3$–CF_3COOD), δ, ppm (*J*, Hz): 2.90 (3H, s, CH_3); 3.05 (3H, s, CH_3); 3.85 (3H, s, OCH_3); 7.00 (2H, d, *J* = 8.2, H Ar); 7.40 (2H, d, *J* = 8.2, H Ar); 8.30 (1H, s, N=CH). Found, %: C 48.72; H 3.68; N 23.93. $C_{19}H_{17}BrN_8O_2$. Calculated, %: C 48.63; H 3.65; N 23.88.

3-Hydrazonopyrazolo[3,4-*b*]pyridine Derivatives 9a,b (General Method). A freshly prepared solution of pyrazolopyridinediazonium chloride **2a** (2.1 g, 0.01 mol) was added with continuous stirring to a cold solution (0-5°C) of *N*'-acyl-2-cyanoacetohydrazide derivatives **8a** or **8b** (0.01 mol) and NaOAc (4.0 g, 0.05 mol) in EtOH (50 ml). The reaction mixture was allowed to stand in the cold for 2 h and then diluted with water. The products so formed were collected by filtration and washed with water, followed by cold EtOH. The isolated compounds were recrystallized from EtOH to give compounds **9a**,**b**.

***N*'-Acetyl-2-cyano-2-[(4,6-dimethyl-1*H*-pyrazolo[3,4-*b*]pyridin-3-yl)hydrazono]acetohydrazide (9a)**. Yield 2.17 g (69%); mp 285-286°C. IR spectrum, ν, cm^{-1}: 3403, 3282, 3201 (3NH), 2192 (CN), 1698, 1657 (2CO). ^{1}H NMR spectrum (DMSO-d_6), δ, ppm: 2.10 (3H, s, $COCH_3$); 2.50 (3H, s, CH_3); 2.60 (3H, s, CH_3); 7.00 (1H, s, H-5); 11.60 (2H, s, 2NH); 12.85 (1H, s, NH); 14.25 (1H, s, NH). Found, %: C 49.50; H 4.38; N 35.56. $C_{13}H_{14}N_8O_2$. Calculated, %: C 49.68; H 4.49; N 35.56.

***N*'-Benzoyl-2-cyano-2-[(4,6-dimethyl-1*H*-pyrazolo[3,4-*b*]pyridin-3-yl)hydrazono]acetohydrazide (9b)**. Yield 2.67 g (71%); mp 280-281°C. IR spectrum, ν, cm^{-1}: 3416, 3284, 3199 (3NH), 2124 (CN), 1667, 1658 (2CO). ^{1}H NMR spectrum (DMSO-d_6), δ, ppm: 2.45 (3H, s, CH_3); 2.60 (3H, s, CH_3); 7.00 (1H, s, H-5); 7.30–7.50 (5H, m, H Ph); 10.35 (2H, s, 2NH); 12.20 (1H, s, NH); 13.75 (1H, s, NH). Found, %: C 57.59; H 4.15; N 29.68. $C_{18}H_{16}N_8O_2$. Calculated, %: C 57.44; H 4.28; N 29.77.

8-Aminopyrido[2',3':3,4]pyrazolo[5,1-*c*]triazine-7-carboxylic Acid *N*'-acylhydrazide Derivatives 10a,b. Compound **9a**,**b** (0.005 mol) in acetic acid (20 ml) was heated under reflux for 30 min and then allowed to cool. The solid product **10a**,**b** was collected by filtration, washed with cold EtOH, and recrystallized from EtOH–DMF mixture (2:1).

8-Amino-2,4-dimethylpyrido[2',3':3,4]pyrazolo[5,1-*c*]triazine-7-carboxylic acid *N*'-Acetylhydrazide (10a). Yield 1.23 g (78%); mp > 300°C. IR spectrum, ν, cm^{-1}: 3378, 3180 (NH_2 and NH), 1641 (br., CO).

^{1}H NMR spectrum ($CDCl_3$–CF_3COOD), δ, ppm: 2.10 (3H, s, $COCH_3$); 2.80 (3H, s, CH_3); 2.95 (3H, s, CH_3); 7.10 (1H, s, H-3). Found, %: C 49.61; H 4.54; N 35.74. $C_{13}H_{14}N_8O_2$. Calculated, %: C 49.68; H 4.49; N 35.65.

8-Amino-2,4-dimethylpyrido[2',3':3,4]pyrazolo[5,1-*c*]triazine-7-carboxylic Acid *N*'-Benzoylhydrazide (10b). Yield 1.58 g (84%); mp > 300°C. IR spectrum, ν, cm^{-1}: 3386, 3189 (NH_2 and NH), 1638 (br., CO). ^{1}H NMR spectrum ($CDCl_3$–CF_3COOD), δ, ppm: 2.80 (3H, s, CH_3); 2.95 (3H, s, CH_3); 7.10 (1H, s, H-3); 7.20-7.50 (5H, m, H Ph). Found, %: C 57.40; H 4.35; N 29.81. $C_{18}H_{16}N_8O_2$. Calculated, %: C 57.44; H 4.28; N 29.77

Ethyl 2-[(4,6-Dimethyl-1*H*-pyrazolo[3,4-*b*]pyridin-3-yl)hydrazono]benzoyl Acetate (11). A freshly prepared solution of pyrazolopyridinediazonium chloride **2a** (2.1 g, 0.01 mol) was added with continuous stirring to a cold solution (0-5°C) of ethyl benzoylacetate (1.92 g, 0.01 mol) and NaOAc (4.0 g, 0.05 mol) in EtOH (50 ml). The reaction mixture was allowed to stand in the cold for 2 h, and then diluted with water. The product so formed was collected by filtration, washed with cold EtOH, and finally was recrystallized from EtOH to give compound **11** as yellow crystals. Yield 2.78 g (76%); mp 165-166°C. IR spectrum, ν, cm^{-1}: 3147 (NH), 1685 (COOEt), 1667 (COPh). ^{1}H NMR spectrum ($CDCl_3$), δ, ppm (*J*, Hz): 1.10 (3H, t, *J* = 7.2, $COOCH_2C\underline{H}_3$); 2.60 (3H, s, CH_3); 2.80 (3H, s, CH_3); 4.25 (2H, q, *J* = 7.2, $COOC\underline{H}_2CH_3$); 7.10 (1H, s, H-5); 7.40–7.70 (5H, m, H Ph); 11.65 (1H, s, NH); 13.30 (1H, s, NH). Found, %: C 62.52; H 5.32; N 19.06. $C_{19}H_{19}N_5O_3$. Calculated, %: C 62.46; H 5.24; N 19.17.

Ethyl 2,4-Dimethyl-8-phenylpyrido[2',3':3,4]pyrazolo[5,1-*c*][1,2,4]triazine-7-carboxylate (12). A solution of compound **11** (1.83 g, 0.005 mol) in EtOH (30 ml) containing catalytic amounts of Et_3N was heated under reflux for 1 h. The product so formed on heating was collected by filtration, washed with cold EtOH, and recrystallized from AcOH to afford **12** as yellowish green crystals. Yield 1.49 g (86%);mp 182-184°C. IR spectrum, ν, cm^{-1}: 1737 (CO, ester). ^{1}H NMR spectrum ($CDCl_3$), δ, ppm (*J*, Hz): 1.10 (3H, t, *J* = 7.2, $COOCH_2C\underline{H}_3$); 2.66 (3H, s, CH_3); 3.00 (3H, s, CH_3); 4.25 (2H, q, *J* = 7.2, $COOC\underline{H}_2CH_3$); 7.12 (1H, s, H-3); 7.47-7.73 (5H, m, H Ph). Mass spectrum, *m*/*z* (I_{rel}, %): 347 $[M]^+$ (90). Found, %: C 65.72; H 4.87; N 20.12. $C_{19}H_{17}N_5O_2$. Calculated, %: C 65.70; H 4.93; N 20.16.

7-Benzoyl-2,4,8-trimethylpyrido[2',3':3,4]pyrazolo[5,1-*c*][1,2,4]triazine (14). A freshly prepared solution of pyrazolopyridinediazonium chloride **2a** (2.1 g, 0.01 mol) was added with continuous stirring to a cold solution (0-5°C) of benzoylacetone (1.62 g, 0.01 mol) and NaOAc (4.0 g, 0.05 mol) in EtOH (50 ml). The reaction mixture was allowed to stand in the cold for 1 h and then diluted with water. The product so formed was collected by filtration, washed with cold ethanol, and finally was recrystallized from ethanol to give compound **14** as green crystals. Yield 2.35 g (74%); mp 170-172°C. IR spectrum, ν, cm^{-1}: 1664 (COPh). ^{1}H NMR spectrum (DMSO-d_6), δ, ppm: 2.35 (3H, s, CH_3); 2.63 (3H, s, CH_3); 2.75 (3H, s, CH_3); 7.10 (1H, s, H-5); 7.12-7.89 (5H, m, H Ph). Mass spectrum, *m*/*z* (I_{rel}, %): 317 $[M]^+$ (18), 316 (31). Found, %: C 68.18; H 4.74; N 22.03. $C_{18}H_{15}N_5O$. Calculated, %: C 68.13; H 4.76; N 22.07.

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