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RESEARCH ARTICLE

SYNTHESIS OF SOME NEW AZOLOTRIAZINE, 4-ARYLAZOPYRAZOLE AND PYRIDINE DERIVATIVES CONTAINING 1,2,3-TRIAZOLE MOIETY

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Abstract

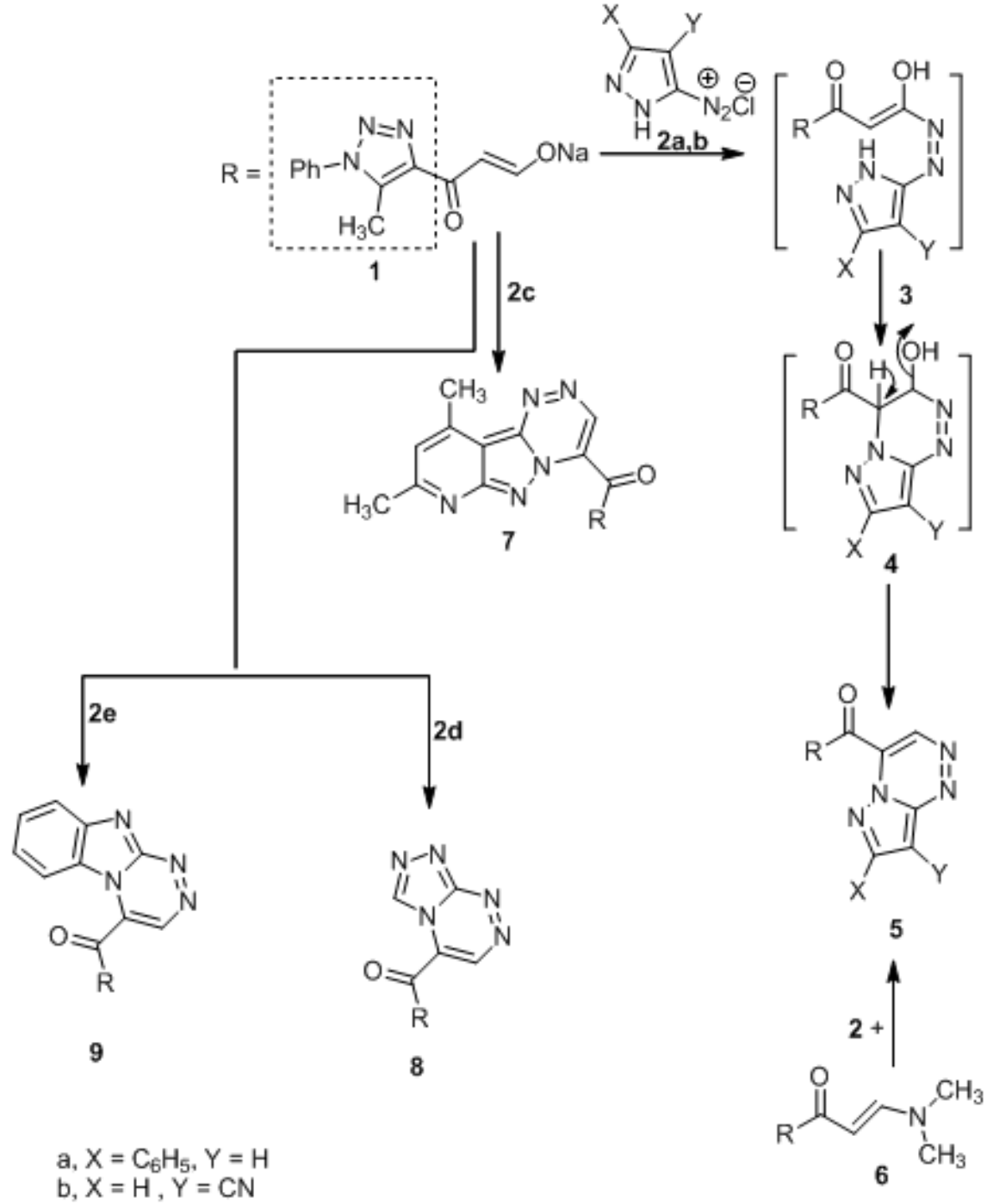
Pyrazolo[5,l-*c*]triazines, [l,2,4]triazolo[4,3-*c*]triazine, benzo[4,5]-imidazo[2,1-*c*][1,2,4]triazine, pyridine, urea and carbamate derivatives containing 1,2,3,-triazole moiety were synthesized in a good yields *via* sodium salt of 3-hydroxy-1-(5-methyl-1-phenyl-1*H*-1,2,3-triazol-4-yl)prop-2-en-1-one. The newly synthesized derivatives were elucidated by elemental analysis, spectral data and alternative synthetic routes whenever possibile.

1. Introduction

In recent years in various publications, certain compounds having a 1,2,3-triazole nucleus have been reported as antibacterials [1], antifungals [2], antivirals [3], anti-inflammatories, and analgesics [4]. Recently, some new 1,3,4-triazole derivatives have been synthesized as possible anticonvulsants [5] and plant growth regulators [6], 1,2,3-triazole derivatives have been synthesized to inhibit tumor proliferation, invasion, metastasis [7], and have shown anti-HIV activity [8-13]. For this reason and continuous of our work [14-21], we report here the convenient synthesis of azolo[5,1-*c*]triazines, 4-arylazopyrazoles, pyrazolo[3,4-*b*]pyridine and pyridines containing 1,2,3-triazole moiety.

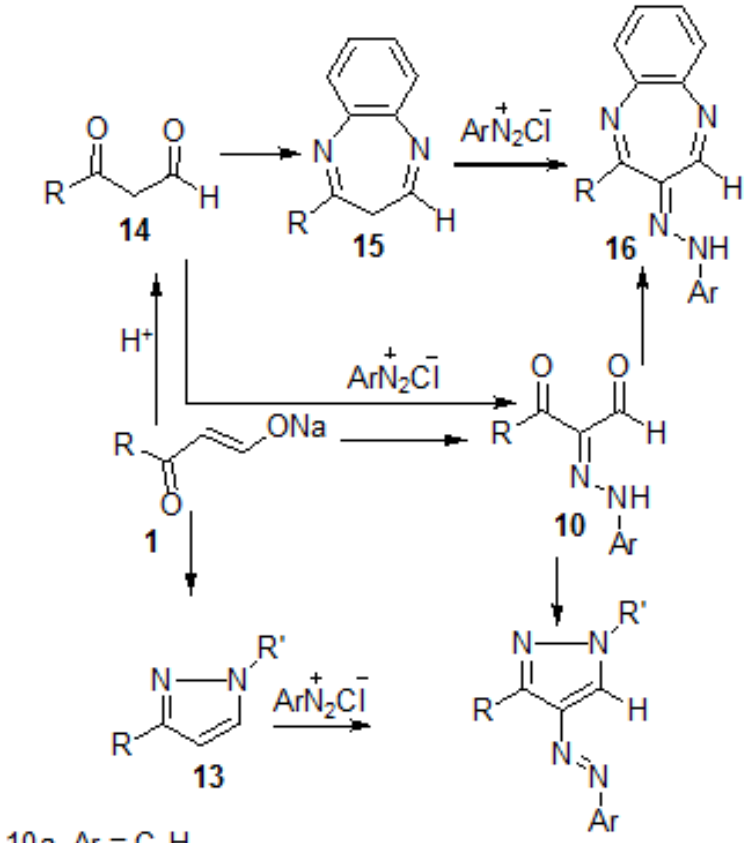
2. Results and Discussion

Treatment of the diazotized 3-amino-5-phenylpyrazole (**2a**) with sodium salt of 3-hydroxy-1-(5-methyl-1-phenyl-1*H*-1,2,3-triazol-4-yl)prop-2-en-1-one (**1**), in ethanolic sodium acetate solution gave (5-methyl-1-phenyl-1*H*-1,2,3-triazol-4-yl)(7-phenylpyrazolo[5,1-*c*][1,2,4]triazin-4-yl)methanone (**5a**) in good yield, (Scheme 1). Analogously, treatment of the appropriate diazonium salt of aminopyrazoles **2b-c** with **1** in ethanolic sodium acetate afforded pyrazolo[5,1-*c*]triazines (**5b**) and pentaazafluorene (**7**), respectively (Schemes 1)**.** Structure **5a** was elucidated by elemental analysis, spectral data and alternative synthetic route. The formation of **5a** accorded via coupling diazonium chloride **2a** with **1** to form the intermediate **3** which converted to another intermediate **4.** The latter afforded the final product **5** through elimination of one molecule of water. Meanwhile, treatment of 3-(dimethylamino)-1-(5-methyl-1-phenyl-1*H*-1,2,3-triazol-4-yl)prop-2-en-1-one (**6**) with **2a** in ethanolic sodium acetate as a buffer solution gave product identical in all aspects (mp., mixed mp. and spectra) with **5a** (Scheme 1).
Analogously, treatment of **1** or **6** with the appropriate 4-cyanopyrazol-3-yldiazonium chloride (**2b**), 4,6-dimethyl-2*H*-pyrazolo[3,4-*b*]pyridine-yldiazonium chloride (**2c**), 1,2,4-triazol-3-yldiazonium nitrate (**2d**) or benzimidazol-2-diazonium sulphate (**2e**) in cold solution of ethanolic sodium acetate gav4-(5-methyl-1-phenyl-1H-1,2,3-triazole-4-carbonyl)pyrazolo[5,1-c][1,2,4]triazine-8-carbonitrile (**5b**), (8,10-dimethylpyrido[2',3':3,4]pyrazolo[5,1-c][1,2,4]triazin-4-yl)(5-methyl-1-phenyl-1H-1,2,3-triazol-4-yl)methanone (**7**), ([1,2,4]triazolo[3,4-c][1,2,4]triazin-5-yl)(5-methyl-1-phenyl-1*H*-1,2,3-triazol-4-yl)methanone (**8**) and benzo[4,5]imidazo[2,1-*c*][1,2,4]triazin-4-yl-(5-methyl-1-phenyl-1*H*-[1,2,3]triazol-4-yl)-methanone (**9**), respectively in good yield (Scheme 1).



Scheme 1

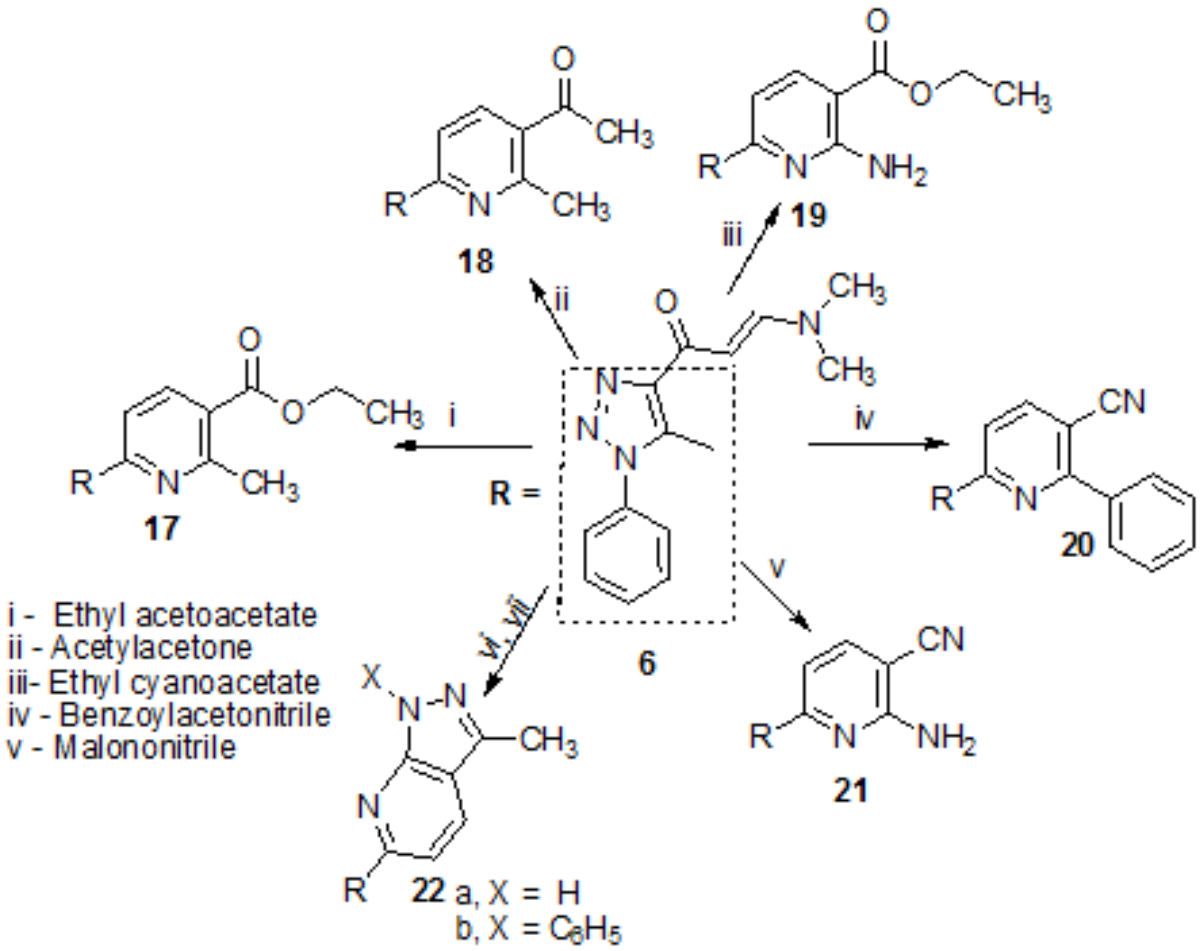
Next, treatment of **1** or **6** with arenediazonium chloride in ethanol containing sodium acetate as a buffer solution yielded 2-(2-arylhydrazono)-3-(5-methyl-1-phenyl-1*H*-1,2,3-triazol-4-yl)-3-oxopropanal **10** (Scheme 2). Structure **10a** was confirmed by elemental analysis, spectral data, alternative synthetic route and chemical transformations. Also, 3-(5-methyl-1-phenyl-1*H*-1,2,3-triazol-4-yl)-3-oxopropanal (**14**) was coupled with benzenediazonium chloride gave product identical in all respects (mp., mixed mp., and spectra) with **10a**. ^{1}H NMR spectrum of **10a** showed signal at δ = 2.06 (s, 3H, CH_3), 7.26-8.20 (m, 10H, ArH's), 9.75 (s, 1H, -CHO) and 14.39 (s, br., 1H, NH). Compound **10a** was reacted with hydrazine hydrate in boiling ethanol under reflux gave 1-(3-(5-methyl-1-phenyl-1*H*-1,2,3-triazol-4-yl)-1*H*-pyrazol-4-yl)-2-phenyldiazene (**11a**) (Scheme 2).



Scheme 2

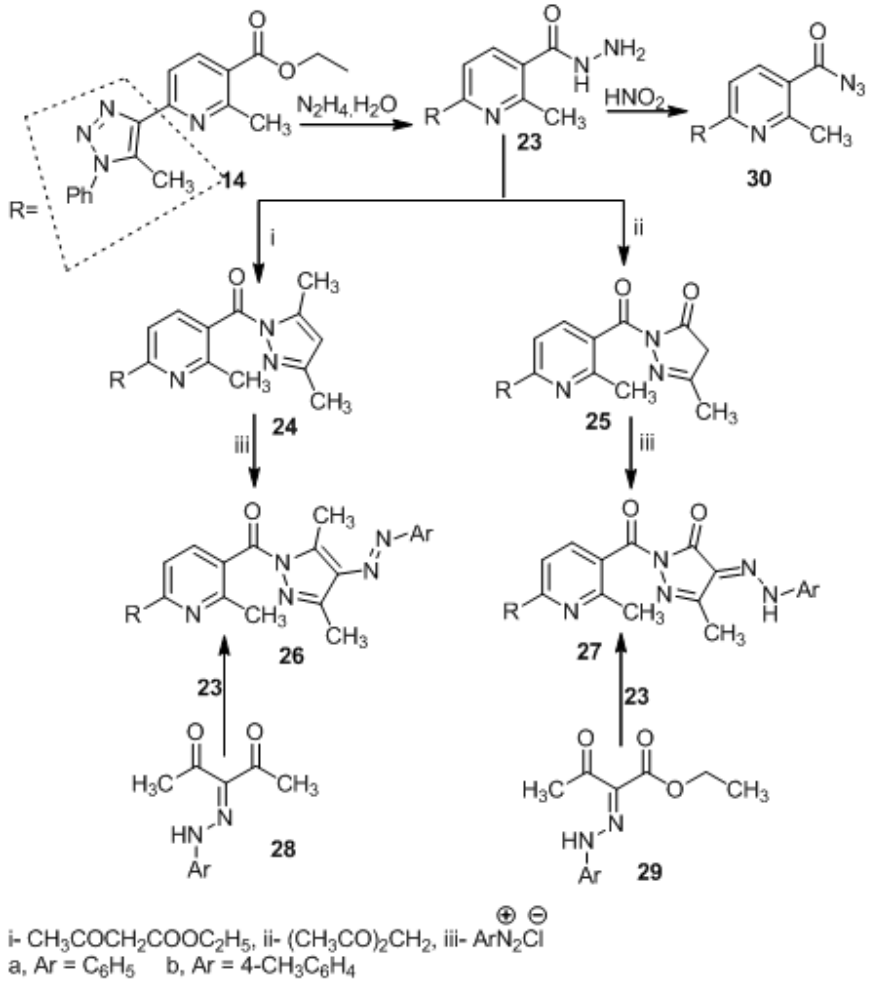
The structure **11a** was confirmed by elemental analysis, spectral data and alternative synthetic route. Thus, compound **1** reacted with hydrazine hydrate gave 5-methyl-1-phenyl-4-(1*H*-pyrazol-3-yl)-1*H*-1,2,3-triazole (**13a**). The latter was reacted with benzenediazonium chloride in ethanolic sodium acetate solution to afford product identical in all aspects (mp., mixed mp. and spectra) with **11a**. Similar, *p*-toluenediazonium chloride reacted with **1** in ethanolic sodium acetate gave **10b**, which converted to pyrazole **12a** by heating with hydrazine hydrate. Compound **10a** reacted with *o*-phenylenediamine afforded 2-(5-methyl-1-phenyl-1*H*-1,2,3-triazol-4-yl)-3-(2-phenylhydrazono)-3*H*-benzo[*b*][1,4]diazepine (**16a**). Structure **16** was confirmed by elemental analysis, spectral data and alternative synthetic route. Thus, 2-(5-methyl-1-phenyl-1*H*-1,2,3-triazol-4-yl)-3*H*-benzo[*b*][1,4]diazepine (**15**), which prepared by reaction of **14** with *o*-phenylenediamine, was reacted with benzenediazonium chloride in ethanolic sodium acetate solution gave product identical in all aspect (mp., mixed mp., and spectra) with **16a**.

Also, treatment of 3-(dimethylamino)-1-(5-methyl-1-phenyl-1*H*-1,2,3-triazol-4-yl)prop-2-en-1-one (**6**) with each of ethyl acetoaetate, acetylacetone, ethyl cyanoacetate, benzoylacetonitrile, malononitrile or 3-methyl-1-substituted pyrazol-5-one in boiling acetic acid containing ammonium acetate under reflux gave ethyl 2-methyl-6-(5-methyl-1-phenyl-1*H*-1,2,3-triazol-4-yl)pyridine-3-carboxylate (**17**),



Scheme 3

1-(2-methyl-6-(5-methyl-1-phenyl-1*H*-1,2,3-triazol-4-yl)pyridin-3-yl)ethanone (**18**), ethyl 2-amino-6-(5-methyl-1-phenyl-1*H*-1,2,3-triazol-4-yl)pyridine-3-carboxylate (**19**), 6-(5-Methyl-1-phenyl-1H-1,2,3-triazol-4-yl)-2-phenylpyridine-3-carbonitrile (**20**), 2-amino-6-(5-methyl-1-phenyl-1H-1,2,3-triazol-4-yl)pyridine-3-carbonitrile (**21**), and 3-methyl-6-(5-methyl-1-phenyl-1*H*-1,2,3-triazol-4-yl)-1-substituted 1*H*-pyrazolo[3,4-*b*]pyridine (**22**), respectively (Scheme 3). Thus, the IR spectra of **19** showed the absence of bands between "2100-2300" cm^{-1} due to the absence of CN group but showed the presence of bands at 3448 and 3274 cm^{-1} due to the presence of an amino group. Also, ^{1}H-NMR spectrum of **19** revealed signals at δ = 1.32 (t, 3H) and 4.31 (q, 2H), indicate the presence of ethoxy group (*cf*. Experimental Part). Structure **17** was confirmed by elemental analysis, spectral data and chemical transformation. Thus, it reacted with hydrazine hydrate to give 2-methyl-6-(5-methyl-1-phenyl-1*H*-1,2,3-triazol-4-yl)pyridine-3-carbohydrazide (**23**) in a good yield.

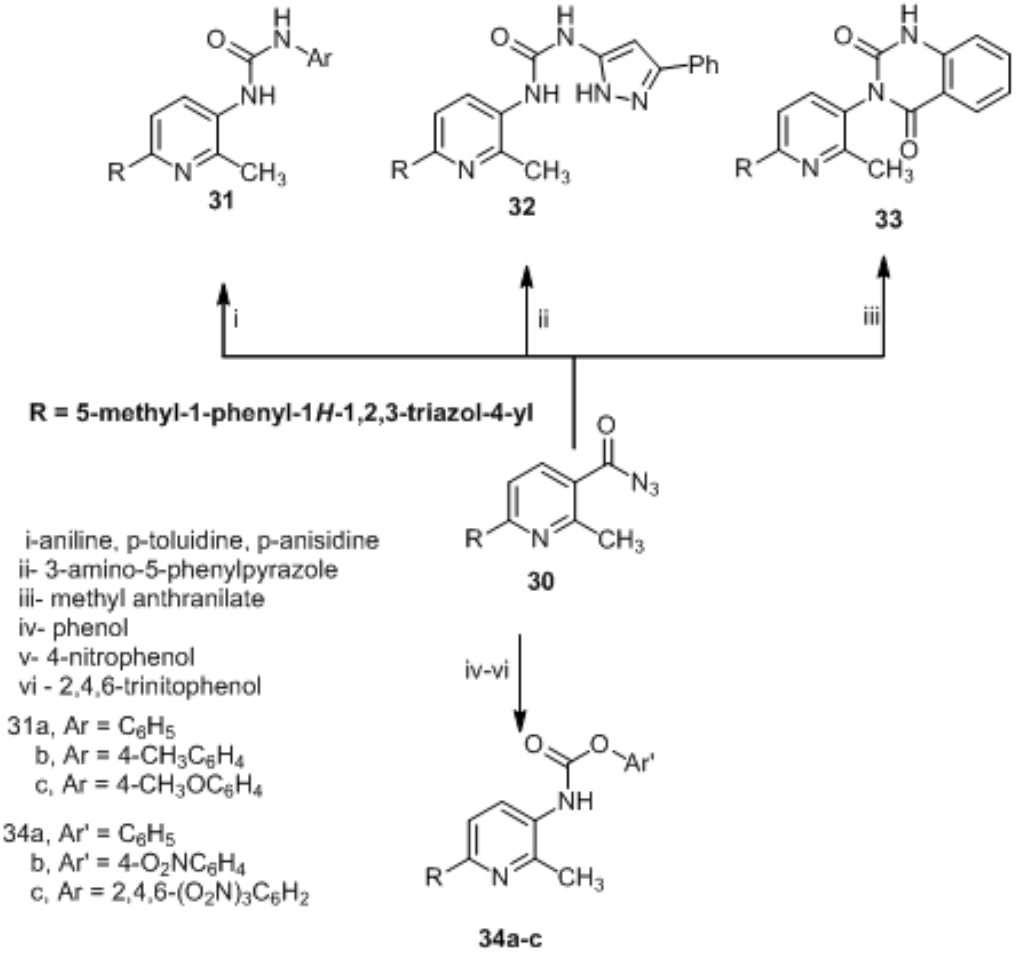


Scheme 4

Compound **23** reacted with each of acetylacetone, ethyl acetacetate or with sodium nitrite in the presence of hydrochloric acid to afford (3,5-dimethyl-1*H*-pyrazol-1-yl)(2-methyl-6-(5-methyl-1-phenyl-1*H*-1,2,3-triazol-4-yl)pyridin-3-yl)methanone (**24**), 5-methyl-2-[2-methyl-6-(5-methyl-1-phenyl-1*H*-[1,2,3]triazol-4-yl)-pyridine-3-carbonyl]-2,4-dihydro-pyrazol-3-one (**25**) and azido(2-methyl-6-(5-methyl-1-phenyl-1*H*-1,2,3-triazol-4-yl)pyridin-3-yl)methanone (**30**), respectively (Scheme 4).

Meanwhile, each compounds **24** and **25** were reacted with benzenediazonium chloride in a cold solution of ethanol containing sodium acetate as a buffer solution to give(3,5-dimethyl-4-(phenyldiazenyl)-1H-pyrazol-1-yl)(2-methyl-6-(5-methyl-1-phenyl-1H-1,2,3-triazol-4-yl)pyridin-3-yl)methanone (**26a**) and 5-methyl-2-[2-methyl-6-(5-methyl-1-phenyl-1*H*-[1,2,3]triazol-4-yl)-pyridine-3-carbonyl]-4-(phenyl-hydrazono)-2,4-dihydro-pyrazol-3-one (**27a**) (structure 4). The structure of compounds **26** and **27** were confirmed by alternative synthesis by treatment of the hydrazide **23** with each of 3-(2-phenylhydrazono)pentane-2,4-dione (**28**) and ethyl 3-oxo-2-(phenylhydrazono)butanoate (**29**) in boiling acetic acid to afford a products which were found identical in all aspects (mp., mixed mp, and spectra) with **26a** and **27a.** Analogously, *p*-tolyldiazonium chloride was reacted with each **24** and **25** gave (3,5-dimethyl-4-(p-tolyldiazenyl)-1*H*-pyrazol-1-yl)(2-methyl-6-(5-methyl-1-phenyl-1*H*-1,2,3-triazol-4-yl)pyridin-3-yl)methanone (**26b**) and3-methyl-1-(2-methyl-6-(5-methyl-1-phenyl-1*H*-1,2,3-triazol-4-yl)nicotinoyl)-4-(2-(p-tolyl)hydrazono)-1*H*-pyrazol-5(4H)-one (**27b**), respectively.

Azido(2-methyl-6-(5-methyl-1-phenyl-1*H*-1,2,3-triazol-4-yl)pyridin-3-yl)methanone (**30**) can be converted into urea derivatives, **31a-c**, **32** and 3-(2-methyl-6-(5-methyl-1-phenyl-1*H*-1,2,3-triazol-4-yl)pyridin-3-yl)quinazoline-2,4(1*H*,3*H*)-dione (**33**) by its boiling with the appropriate aromatic amines, 3-amino-5-phenylpyrazole or methyl anthranilate (anthranilic acid) in dry dioxane, respectively. Also, aryl 2-methyl-6-(5-methyl-1-phenyl-1*H*-1,2,3-triazol-4-yl)pyridin-3-ylcarbamate **34a-c** can obtained by boiling the azido **30** with substituted phenols in dry benzene (Scheme 5).



Scheme 5

3. Experimental

All melting points were determined on an electrothermal apparatus and are uncorrected. IR spectra were recorded (KBr discs) on a Shimadzu FT-IR 8201 PC spectrophotometer. NMR spectra were recorded in $CDCl_3$ and $(CD_3)_2SO$ solutions on a Burker AM 400Mz and a Varian Gemini 300 MHz spectrometer and chemical shifts are expressed in δ ppm units using TMS as an internal reference. Mass spectra were recorded on a GC-MS QP1000 EX Shimadzu, QP-2010 Plus Shimadzu. Elemental analyses were carried out at the Microanalytical Center of Cairo University.

3.1. Sodium salt of 3-hydroxy-1-(5-methyl-1-phenyl-1H-1,2,3-triazol-4-yl)prop-2-en-1-one (1).

In three-necked flask (250 mL) take of sodium methoxide (0.54 g, 10 mmoles) and ether (20 mL) and pour over it through separating funnel the 1-(5-methyl-1-phenyl-1*H*-1,2,3-triazol-4-yl)ethanone (2.1g, 10 mmoles) with ethyl formate (0.74 g, 10 mmoles) with efficient stirring. The solid product was collected and used directly in the reactions.

3.2. (5-Methyl-1-phenyl-1*H*-1,2,3-triazol-4-yl)(7-phenylpyrazolo[5,1-*c*][1,2,4]triazin-4-yl)methanone (5a), 4-(5-Methyl-1-phenyl-1*H*-[1,2,3]triazole-4-carbonyl)-pyrazolo[5,1-*c*][1,2,4]triazine-8-carbonitrile (5b), 2,4 - Dimethyl-1,5,6,8a,9-pentaaza-fluoren-8-yl)-(5-methyl-1-phenyl-1H-[1,2,3]triazol-4-yl)-methanone (7), ([1,2,4]triazolo[3,4-c][1,2,4]-triazin-5-yl)(5-methyl-1-phenyl-1*H*-1,2,3-triazol-4-yl)methanone (8) and benzo[4,5]imidazo[2,1-c][1,2,4]triazin-4-yl-(5-methyl-1-phenyl-1H-[1,2,3]triazol-4-yl)-methanone (9).

Method A. A solution of the appropriate diazonium salt of heterocyclic amines 3-amino-5-phenylpyrazole (**2a**), 3-amino-4-cyanopyrazole (**2b**), 4,6-dimethyl-2*H*-pyrazolo[3,4-*b*]pyridin-3-amine (**2c**), 3-amino-1,2,4-triazole (**2d**), 2-amino-benzimidazole (**2e**) (5 mmole) was added to a mixture of sodium salt of 3-hydroxy-1-(5-methyl-1-phenyl-1*H*-1,2,3-triazol-4-yl)prop-2-en-1-one (**1**) (1.25 g, 5 mmole), sodium acetate (0.65 g, 5 mmole) in ethanol (30 mL) at 0–5^{o}C while stirring. The solid so formed after 3 h was collected, washed with water, and recrystallized to give **5a**, **5b**, **7**, **8** and **9**.

Method B. A solution of the appropriate diazonium salt of heterocyclic amines (3-amino-5-phenylpyrazole (**2a**), 3-amino-4-cyanopyrazole (**2b**), 4,6-dimethyl-2*H*-pyrazolo[3,4-*b*]pyridin-3-amine (**2c**), 3-amino-1,2,4-triazole (**2d**), 2-amino-benzimidazole (**2e**) (5 mmole) was added to a mixture of 3-(dimethylamino)-1-(5-methyl-1-phenyl-1*H*-1,2,3-triazol-4-yl)prop-2-en-1-one (**6**) (1.28 g, 5 mmole), sodium acetate (0.65 g, 5 mmole) in ethanol (30 mL) at 0–5_C while stirring. The resulting solid which formed after 3 h was collected, washed with water, and recrystallized from proper solvent to give products identical in all aspects mp., mixed mp., and spectra with the corresponding obtained in method A.

3.2.2. (5-Methyl-1-phenyl-1*H*-1,2,3-triazol-4-yl)(7-phenylpyrazolo[5,1-*c*][1,2,4]-triazin-4-yl)methanone (5a). Orange crystals from benzene, yield (88%), mp: 248–50^{o}C; IR (KBr): 3059 (CH, aromatic), 2974, 2939 (CH, aliphatic), 1681 (CO), 1647 (C=N), and 1512 (C=C); ^{1}H NMR ($CDCl_3$): δ 2.64 (s, 3H, CH_3), 6.33 (s, 1H, pyrazole H-4), 7.48-7.98 (m, 10H, ArH's) and 9.61 (s, 1H, ArH); MS: m/z = 381 (M, 9.8%), 380 (5.6%), 352 (10%), 325 (30%), 223 (20%), 130 (26.7%), 118 (43.3%), 103 (21.1%), 77 (100%), 66 (21.1%), 51 (50%); Anal. Calcd. For $C_{21}H_{15}N_7O$ (381.39) C, 66.13; H, 3.96; N, 25.71 Found: C, 66.24; H, 4.10; N, 25.79 %.

3.2.3. 4-(5-Methyl-1-phenyl-1H-[1,2,3]triazole-4-carbonyl)-pyrazolo[5,1-c][1,2,4]-triazine-8-carbonitrile (5b). Brown crystals from benzene, yield (92%), mp: >300^{o}C; IR (KBr): 3028 (CH, aromatic), 2912, 2827 (CH, aliphatic), 2233 (CN), 1670 (CO) and 1596 (C=C); ^{1}H NMR ($CDCl_3$): δ 2.61 (s, 3H, CH_3), 7.33-8.08 (m, 5H, ArH's), 8.24 (s, 1H, pyrazole H-3) and 8.42 (s, 1H, ArH); MS: m/z = 330 (M, 1%), 273 (2%), 219 (3%), 165 (4%), 155 (5%), 145 (14%), 130 (18.3%), 123 (17%), 117 (11%), 103 (26.7%), 93 (93%), 77 (100%), 64 (91%), 80 (90.2%); Anal. Calcd. For $C_{16}H_{10}N_8O$ (330.3) C, 58.18; H, 3.05; N, 33.92 Found: C, 58.25; H, 3.17; N, 34.10%.

3.2.4. (2,4-Dimethyl-1,5,6,8a,9-pentaaza-fluoren-8-yl)-(5-methyl-1-phenyl-1H-[1,2,3]triazol-4-yl)-methanone (7). Yellow crystals from EtOH, yield (82%), mp: >300^{o}C; IR (KBr): 3066 (CH, aromatic), 2924 (CH, aliphatic), 1678 (CO) and 1604 (C=C); ^{1}H NMR ($CDCl_3$): δ 2.61 (s, 3H, CH_3), 2.69 (s, 3H, CH_3), 2.93 (s, 3H, CH_3), 6.78 (s, 1H, pyridine H-5), 7.13-7.44 (m, 5H, ArH's) and 8.34 (s, 1H, triazine H-6); MS: m/z = 384 (M, 6%), 274 (95%), 357 (83%), 345 (76%), 317 (83%), 300 (83%), 247 (90%), 225 (83%), 200 (89%), 197 (93%), 186 (83%), 185 (18%), 174 (83%), 165 (88%), 149 (87%), 142 (87%), 130 (33%), 121 (76%), 90 (100%), 77 (21%); Anal. Calcd. For $C_{20}H_{16}N_8O$ (384.39) C, 62.49; H, 4.20; N, 29.15 Found: C, 63.17; H, 4.31; N, 29.24%.

3.2.5. ([1,2,4]Triazolo[3,4-c][1,2,4]triazin-5-yl)(5-methyl-1-phenyl-1H-1,2,3-triazol-4-yl)methanone (8). Brown crystals from benzene, yield (89%), mp: 210-12^{o}C; IR (KBr): 3058 (CH, aromatic), 2920, 2850 (CH, aliphatic), 1689 (CO), 1609 (C=N) and 1551 (C=C); ^{1}H NMR ($CDCl_3$): δ 2.43 (s, 3H, CH_3), 7.33-7.62 (m, 4H, ArH's), 7.65-7.68 (m, 1H, ArH)), 7.85 (s, 1H, ArH) and 7.93 (s, 1H, ArH); MS: m/z = 306 (M, 2%), 250 (15.7%), 130 (31%), 118 (15%), 103 (8%), 96 (6%), 77 (100%), 76 (25%), 65 (10%), 53 (18%); Anal. Calcd. For $C_{14}H_{10}N_8O$ (306.28) C, 54.90; H, 3.29; N, 36.59 Found: C, 54.90; H, 3.29; N, 36.59%.

3.2.6. Benzo[4,5]imidazo[2,1-c][1,2,4]triazin-4-yl-(5-methyl-1-phenyl-1H-[1,2,3-]triazol-4-yl)-methanone (9). Beige crystals from benzene, yield (74%), mp: >300^{o}C; IR (KBr): 1712 (CO) and 1512 (C=C); ^{1}H NMR ($CDCl_3$): δ 2.48 (s, 3H, CH_3), 7.22-7.53 (m, 9H, ArH's), and 7.96 (s, 1H, ArH); MS: m/z = 356 (M-1, 4%), 214 (4%), 108 (6%), 92 (11%), 91 (100%), 77 (7%); Anal. Calcd. For $C_{19}H_{13}N_7O$ (355.35) C, 64.22; H, 3.69; N, 27.59 Found: C, 64.39; H, 3.76; N, 27.62%.

3.2.7. 2-(2-Arylhydrazono)-3-(5-methyl-1-phenyl-1H-1,2,3-triazol-4-yl)-3-oxo-propanal 10a,b

A solution of the appropriate arenediazonium chloride (5 mmole) was added to a mixture of sodium salt of 3-hydroxy-1-(5-methyl-1-phenyl-1*H*-1,2,3-triazol-4-yl)prop-2-en-1-one (**1**) (1.25 g, 5 mmole),, sodium acetate (0.65 g, 5 mmole) in ethanol (30 mL) at 0–5^{o}C while stirring. The resulting solid which formed after 3 h was collected, washed with water and recrystallized to give **10a** and **10b**, respectively.

***3.2.8. 2-(2-Phenylhydrazono)-3-(5-methyl-1-phenyl-1H-1,2,3-triazol-4-yl)-3-oxo-propanal (10a)*.** Brown crystals from EtOH, yield (76%), mp: 172-74^{o}C; IR (KBr): 3352 (NH), 3066, 2920 (CH), 2866, 2754 (CHO), 1678 (CO), 1643 (C=N) and 1600 (C=C); ^{1}H NMR ($CDCl_3$): δ 2.60 (s, 3H, CH_3), 7.47-7.78 (m, 10H, ArH's), 8.81 (s, br., 1H, NH) and 13.59 (s, 1H, CHO); MS: m/z = 333 (M, 1.2%), 283 (10%), 158 (7%), 131 (16%), 130 (40%), 118 (31%), 103 (41%), 91 (47%), 89 (14%), 78 (15%), 76 (94%), 63 (37%), 50 (100%); Anal. Calcd. For $C_{18}H_{15}N_5O_2$ (333.34) C, 64.86; H, 4.54; N, 21.01 Found: C, 64.81; H, 4.62; N, 21.14%.

***3.2.9. 2-(2-p-Tolylhydrazono)-3-(5-methyl-1-phenyl-1H-1,2,3-triazol-4-yl)-3-oxo-propanal (10b*).** Orange crystals from EtOH, yield (74%), mp: 178-80^{o}C; IR (KBr): 3352 (NH), 3066, 2920 (CH), 2866, 2754 (CHO), 1678 (CO), 1643 (C=N) and 1601 (C=C); ^{1}H NMR ($CDCl_3$): δ 2.26 (s, 3H, CH_3), 2.60 (s, 3H, CH_3), 7.38-7.69 (m, 9H, ArH's), 8.75 (s, br., 1H, NH) and 9.75 (s, 1H, CHO); MS: m/z = 348 (M+1, 1.03%), 304 (5%), 300 (8%), 283 (30%), 270 (7%), 238 (12.7%), 208 (21%), 194 (5%), 160 (5%), 149 (6%), 130 (20%), 106 (30%), 90 (99%), 76 (86%); Anal. Calcd. For $C_{19}H_{17}N_5O_2$ (347.37) C, 65.69; H, 4.93; N, 20.16 Found: C, 65.72; H, 5.13; N, 20.30%.

3.3. 1-(3-(5-Methyl-1-phenyl-1*H*-1,2,3-triazol-4-yl)-4*H*-pyrazol-4-ylidene)-2-arylhydrazine 11a-c and 12a-c

Method A: A mixture of the appropriate **10a** and **10b** (5 mmole) and the appropriate the appropriate of hydrazine hydrate, phenylhydrazine or p-nitrophenylhydrazine (5 mmole) (5 mmole) in ethanol (15 mL) was refluxed for 2 h. The resulting solid was collected and recrystallized afforded **11a-c** and **12a-c**, respectively.

Method B**::** A solution of the appropriate arenediazonium chloride (5 mmole) was added to a mixture of the appropriate **13a-c** (5 mmole), sodium acetate (0.65 g, 5 mmole) in ethanol (30 mL) at 0–5^{o}C while stirring. The resulting solid which formed after 3 h was collected, washed with water and recrystallized gave identical in all aspects (mp: mixed mp., and spectra) with the corresponding obtained **11a–c** and **12 a-c**.

3.3.1. 1-(3-(5-Methyl-1-phenyl-1*H*-1,2,3-triazol-4-yl)-1*H*-pyrazol-4-yl)-2-phenyldiazene (11a).

Brown crystals from EtOH, yield (83%), mp: 208-10^{o}C; IR (KBr): 3201 (NH), 3062, 2923, 2854 (CH), 1643 (C=N) and 1596 (C=C); ^{1}H NMR ($CDCl_3$): δ 2.62 (s, 3H, CH_3), 7.18-7.87 (m, 10H, ArH's), 8.75 (s, 1H, pyrazole H-5) and 10.84 (s, br., 1H, NH); MS: m/z = 329 (M, 0.6%), 294 (13%), 292 (12%), 267 (14%), 266 (98%), 264 (100%), 249 (15%), 247 (12%), 225 (29%), 213 (17%), 211 (18%), 184 (11%), 182 (13%), 140 (18%), 114 (10%), 102 (16%), 77 (6%), 75 (12%), 43 (49%); Anal. Calcd. For $C_{18}H_{15}N_7$ (329.36) C, 65.64; H, 4.59; N, 29.77 Found: C, 65.57; H, 4.47; N, 29.69%.

3.3.2. 1-(3-(5-Methyl-1-phenyl-1H-1,2,3-triazol-4-yl)-1-phenyl-1H-pyrazol-4-yl)-2-phenyldiazene (11b).

Orange crystals from AcOH, yield (81%), mp: 200-202^{o}C; IR (KBr): 3055, 2923, 2854 (CH), and 1604 (C=C); ^{1}H NMR ($CDCl_3$): δ 2.62 (s, 3H, CH_3), 7.18-7.99 (m, 16H, ArH's and pyrazole H-5); MS: m/z = 405 (M, 6%), 373 (38%), 328 (11%), 253 (14%), 215 (11%), 185 (77%), 156 (9%), 147 (7%), 225 (29%), 144 (11%), 131 (14%), 118 (77%), 113 (10%), 104 (10%), 97 (14%), 91 (11%), 85 (35%), 77 (100%), 71 (51%), 69 (17%), 65 (10%); Anal. Calcd. For $C_{24}H_{19}N_7$ (405.45) C, 71.09; H, 4.72; N, 24.18 Found: C, 71.15; H, 4.79; N, 24.22%.

3.3.3. 1-(3-(5-Methyl-1-phenyl-1H-1,2,3-triazol-4-yl)-1-(4-nitrophenyl)-1H-pyrazol-4-yl)-2-phenyldiazene (11c).

Orange crystals from dioxane, yield (89%), mp: 222-24^{o}C; IR (KBr): 3062, 2920, 2850 (CH), 1593 (C=C) and 1535, 1392 (NO_2); ^{1}H NMR ($CDCl_3$): δ 2.62 (s, 3H, CH_3), 7.18-7.99 (m, 13H, ArH's and pyrazole H-5), 8.37-8.39 (d, 2H, *J* = 8Hz, ArH's); MS: m/z = 450 (M, 6%), 449 (19%), 421 (28%), 118 (55%), 80 (17%), 77 (100%), 64 (17%); Anal. Calcd. For $C_{24}H_{18}N_8O_2$ (450.45) C, 63.99; H, 4.03; N, 24.88 Found: C, 63.89; H, 4.12; N, 24.81%.

3.3.4. 1-(3-(5-Methyl-1-phenyl-1H-1,2,3-triazol-4-yl)-1H-pyrazol-4-yl)-2-p-tolyldiazene (12a).

Brown crystals from EtOH, yield (75%), mp: 240-42^{o}C; IR (KBr): 3213 (NH), 3066, 2923, 2854 (CH), and 1600 (C=C)); ^{1}H NMR ($CDCl_3$): δ 2.38 (s, 3H, 4-$CH_3C_6H_4$), 2.60 (s, 3H, CH_3), 7.10-7.82 (m, 9H, ArH's), 8.68 (s, 1H, pyrazole H-5), and 10.84 (s, br., 1H, NH); MS: m/z = 343 (M, 1%), 222 (23%), 105 (87%), 100 (100%), 87 (33%), 77 (35%); Anal. Calcd. For $C_{19}H_{17}N_7$ (343.39) C, 66.46; H, 4.99; N, 28.55 Found: C, 66.49; H, 4.91; N, 28.37%.

3.3.5. 1-(3-(5-Methyl-1-phenyl-1H-1,2,3-triazol-4-yl)-1-phenyl-1H-pyrazol-4-yl)-2-p-tolyldiazene (12b).

Yellow crystals from dioxane, yield (88%), mp: 160-62^{o}C; IR (KBr): 3028, 2916, 2854 (CH), and 1620 (C=N), 1562 (C=C); ^{1}H NMR ($CDCl_3$): δ 2.36 (s, 3H, 4-$CH_3C_6H_4$), 2.62 (s, 3H, CH_3), 7.10-7.92 (m, 15H, ArH's and pyrazole H-

5); MS: m/z = 419 (M, 31%), 390 (58%), 345 (16%), 296 (21%), 252 (16%), 207 (18%), 195 (16%), 183 (17%), 157 (17%), 118 (35%), 94 (16%), 77 (100%), 68 (17%), 65 (19%); Anal. Calcd. For $C_{25}H_{21}N_7$ (419.19) C, 71.58; H, 5.05; N, 23.37 Found: C, 71.50; H, 5.10; N, 23.45%.

3.3.6. 1-(3-(5-Methyl-1-phenyl-1H-1,2,3-triazol-4-yl)-1-(4-nitrophenyl)-1H-pyrazol-4-yl)-2-p-tolyldiazene (12c).

Brick red crystals from dioxane, yield (89%), mp: 238-40^{o}C; IR (KBr): 3031, 2916, 2858 (CH), 1597 (C=C) and 1539, 1392 (NO_2); ^{1}H NMR ($CDCl_3$): δ 2.37 (s, 3H, 4-$CH_3C_6H_4$), 2.62 (s, 3H, CH_3), 7.18-7.99 (m, 12H, ArH's and pyrazole H-5), 8.37-8.39 (d, 2H, *J* = 8Hz, ArH's); MS: m/z = 465 (M+1, 1.2%), 464 (M, 4%), 463 (12%), 435 (14%), 317 (6%), 186 (16%), 131 (9%), 118 (80%), 107 (22%), 104 (11%), 61 (48%), 77 (100%), 64 (31%); Anal. Calcd. For $C_{25}H_{20}N_8O_2$ (464.17) C, 64.65; H, 4.34; N, 24.12 Found: C, 64.69; H, 4.43; N, 24.24%.

3.4. 5-Methyl-1-phenyl-4-(1-aryl-1H-pyrazol-3-yl)-1H-1,2,3-triazoles 13a-c

A mixture of sodium salt of 3-hydroxy-1-(5-methyl-1-phenyl-1*H*-1,2,3-triazol-4-yl)prop-2-en-1-one (**1**) (1.25 g, 5 mmole) and the appropriate of hydrazine hydrate, phenylhydrazine or *p*-nitrophenylhydrazine (5 mmole) in ethanol (15 mL) was refluxed for 2 h. The resulting solid was collected and recrystallized from acetic acid to give **13a-c.**

3.4.1. 5-Methyl-1-phenyl-4-(1H-pyrazol-3-yl)-1H-1,2,3-triazole (13a).

White crystals from AcOH, yield (73%), mp: 270-72^{o}C; IR (KBr): 3220 (NH), 3058, 2916, 2858 (CH), 1608 (C=C); ^{1}H NMR ($CDCl_3$): δ 2.62 (s, 3H, CH_3), 6.34 (d, 1H, *J* = 4 Hz, pyrazole H-4), 7.18-7.44 (m, 5H, ArH's), 7.54-7.55 (d, 1H, *J* = 4Hz, pyrazole H-5), 7.65-769 (m, 1H, ArH)); MS: m/z = 224 (M-1, 13%), 103 (20%), 98 (33%), 97 (40%), 91 (19%), 85 (40%), 84 (27%), 72 (36%), 63 (84%), 60 (53%), 55 (71%), 52 (124%); Anal. Calcd. For $C_{12}H_{11}N_5$ (225.25) C, 63.99; H, 4.92; N, 31.09 Found: C, 64.10; H, 5.12; N, 31.00%.

3.4.2. 5-Methyl-1-phenyl-4-(1-phenyl-1H-pyrazol-3-yl)-1H-1,2,3-triazole (13b).

Orange crystals from AcOH, yield (82%), mp: 240-42^{o}C; IR (KBr): 3058, 2920, (CH), 1608 (C=C); ^{1}H NMR ($CDCl_3$): δ 2.62 (s, 3H, CH_3), 7.18-7.22 (m, 12H, ArH's and pyrazole proton); Anal. Calcd. For $C_{18}H_{15}N_5$ (225.25) C, 63.99; H, 4.92; N, 31.09 Found: C, 64.10; H, 5.12; N, 31.00%.

3.4.3. 5-Methyl-4-(1-(4-nitrophenyl)-1H-pyrazol-3-yl)-1-phenyl-1H-1,2,3-triazole (13c).

Red crystals from AcOH, yield (89%), mp: 188-90^{o}C; IR (KBr): 3058, 2920 (CH), 1608 (C=C) and 1539, 1392 (NO_2); ^{1}H NMR ($CDCl_3$): δ 2.62 (s, 3H, CH_3), 7.10-7. 62 (m, 11H, ArH's and pyrazole proton); Anal. Calcd. For $C_{18}H_{14}N_6O_2$ (346.12) C, 62.42; H, 4.07; N, 24.27 Found: C, 62.49; H, 4.17; N, 24.35%.

3.5. 3-(5-Methyl-1-phenyl-1H-1,2,3-triazol-4-yl)-3-oxopropanal (14).

A solution of sodium salt of 3-hydroxy-1-(5-methyl-1-phenyl-1*H*-1,2,3-triazol-4-yl)prop-2-en-1-one (**1**) (1 g) in water (20mL) was acidified with hydrochloric acid (6 M). The resulting solid was collected and recrystallized from ethanol gave **14** as yellow crystals. yield (84%), mp: 104-106^{o}C; IR (KBr): 3062, 2924 (CH), 1680 (CO), 1629 (C=N), 1595 (C=C); ^{1}H NMR ($CDCl_3$): δ 2.64 (s, 3H, CH_3), 3.82 (s, 2H, CH_2), 6.60-6.61 (d, 1H, *J* = 4Hz, CH), 7.22-7.72 (m, 5H, ArH's)); MS: m/z = 301 (M, 0.15%), 276 (0.85%), 201 (10%), 158 (14%), 131 (50%), 130 (56%), 118 (100%), 108 (17%), 103 (14%), 91 (34%), 77 (69%), 63 (23%); Anal. Calcd. For $C_{12}H_{11}N_3O_2$ (229.23) C, 62.87; H, 4.84; N, 18.33 Found: C, 62.92; H, 4.78; N, 18.28%.

3.6. 2-(5-Methyl-1-phenyl-1H-1,2,3-triazol-4-yl)-3H-benzo[b][1,4]diazepine (15)

A mixture of **14** (1.14 g, 5 mmole) and o-phenylendiamine (0.55 g, 5 mmol) in ethanol (20mL) was heated under reflux for 2h. The resulting solid was collected and recrystallized from ethanol gave **15** as yellow crystals, yield (69%), mp: 180-82^{o}C; IR (KBr): 3062, 2974, 2858 (CH), 1647, 1620 (C=N), 1593 (C=C); ^{1}H NMR ($CDCl_3$): δ 2.62 (s, 3H, CH_3), 2.68-2.82 (m, 2H, CH_2), 7.22-7.72 (m, 10H, ArH's)); MS: m/z = 229 (M, 6%), 222 (24%), 218 (25%), 213 (24%), 208 (27%), 201 (67%), 195 (23%), 185 (26%), 170 (24%), 161 (26%), 154 (37%), 140 (27%), 125 (25%), 115 (18%), 108 (22%), 99 (26%), 88 (10%); Anal. Calcd. For $C_{18}H_{15}N_5$ (301.35) C, 71.74; H, 5.02; N, 23.24 Found: C, 71.78; H, 5.11; N, 23.32%.

3.7. 2-(5-Methyl-1-phenyl-1H-1,2,3-triazol-4-yl)-3H-benzo[b][1,4]diazepin-3-ylidene)-2-substituted hydrazine 16a and 16b.

A solution of the appropriate arenediazonium chloride (5 mmole) was added to a mixture of **14** (5 mmole), sodium acetate (0.65 g, 5 mmole) in ethanol (30 mL) at 0–5°C while stirring. The resulting solid which formed after 3 h was collected, washed with water and recrystallized from acetic acid to give **16a** and **16b**, respectively.

3.7.1. 2-(5-methyl-1-phenyl-1H-1,2,3-triazol-4-yl)-3-(2-phenylhydrazono)-3H-benzo[b][1,4]diazepine (16a).
Red crystals from AcOH, yield (78%), mp: 192-94°C; IR (KBr): 3309 (NH), 3031 (CH), 1620 (C=N), 1555(C=C); ^{1}H NMR ($CDCl_3$): δ 2.64 (s, 3H, CH_3), 6.85 – 7.66 (m, 15 H, ArH's), 11.23 (s, br., 1H, NH); MS: m/z = 405 (M, 5%), 395 (26%), 379 (17.4%), 372 (11%), 365 (52%), 85342 (35%), 339 (35%), 322 (44%), 318 (16%),306 (36%), 289 (11%), 268 (36%), 241 (39%), 227 (15%), 214 (47%), 176 (35%), 148 (37%), 113 (36%); Anal. Calcd. For $C_{24}H_{19}N_7$ (405.45) C, 71.09; H, 4.72; N, 24.18 Found: C, 71.12; H, 4.85; N, 23.98%.

3.7.2. 2-(5-Methyl-1-phenyl-1H-1,2,3-triazol-4-yl)-3H-benzo[b][1,4]diazepin-3-ylidene)-2-p-tolylhydrazine (16b).
Orange crystals from AcOH, yield (89%), mp: 198-200°C; IR (KBr): 3379 (NH), 3028 (CH), 2977, 2920 (CH), 1612 (C=N), 1562(C=C); ^{1}H NMR ($CDCl_3$): δ 2.32 (s, 3H, 4-$CH_3C_6H_4$), 2.64 (s, 3H, CH_3), 7.24 – 7.66 (m, 14 H, ArH's), 11.23 (s, br., 1H, NH); MS: m/z = 420 (M+1, 5%), 408 (21%), 406 (86%), 384 (11%), 374 (11%), 368 (12%), 358 (38%), 338 (6%), 317 (32%), 289 (22%), 271 (14%), 219 (13%), 206 (18%), 201 (35%), 169 (10%), 144 (26%), 131 (21%), 118 (78%), 105 (18%), 90 (14%), 77 (100%), 64 (17%); Anal. Calcd. For $C_{25}H_{21}N_7$ (419.48) C, 71.58; H, 5.05; N, 23.37 Found: C, 71.58; H, 5.05; N, 23.37%.

3.8. Pyridine derivatives 17-21 and pyrazolo[3,4-b]pyridine 22a and 22b.
A mixture of the appropriate of ethyl acetoacetate, acetylacetone, ethyl cyanoacetate, benzoylacetonitrile, malononitrile, 3-methyl-1*H*-pyrazol-5(4*H*)-one or 3-methyl-1-phenyl-1*H*-pyrazol-5(4*H*)-one (5 mmole), 3-(dimethylamino)-1-(5-methyl-1-phenyl-1*H*-1,2,3-triazol-4-yl)prop-2-en-1-one (**6**) (1.32 g, 5 mmole) and ammonium acetate (0.37 g, 5 mmole) in acetic acid (30 mL) was reflux for 4 h. The resulting solid which formed was collected and recrystallized to give **17-22b**.

3.8.1. Ethyl 2-methyl-6-(5-methyl-1-phenyl-1H-1,2,3-triazol-4-yl)pyridine-3-carboxylate (17).
White crystals from EtOH, yield (83%), mp: 108-110°C; IR (KBr): 3070 (CH), 2974, 2927 (CH), 1716 (CO), 1589 (C=C); ^{1}H NMR ($CDCl_3$): δ 1.35 (t, 3H, $CH_2\mathbf{CH_3}$), 2.63 (s, 3H, CH_3), 2.71 (s, 3H, CH_3), 4.22 (q, 2H, $\mathbf{CH_2}CH_3$), 7.11 – 8.32 (m, 7 H, ArH's); MS: m/z = 322 (M, 7%), 321 (34%), 281 (11%), 177 (19%), 149 (100%), 121 (12%), 104 (12%), 103 (23%), 76 (8%), 65 (9%); Anal. Calcd. For $C1_8H_{18}N_4O_2$ (322.36) C, 67.07; H, 5.63; N, 17.38 Found: C, 67.15; H, 5.52; N, 17.29%.

3.8.2. 1-(2-Methyl-6-(5-methyl-1-phenyl-1H-1,2,3-triazol-4-yl)pyridin-3-yl)ethanone (18).
White crystals from EtOH, yield (91%), mp: 178-180°C; IR (KBr): 3058 (CH), 2958, 2920 (CH), 1678 (CO), 1589 (C=C); ^{1}H NMR ($CDCl_3$): δ 2.48 (s, 3H, CH_3), 2.49 (s, 3H, CH_3), 2.63 (s, 3H, CH_3), 7.11-8.23 (m, 7 H, ArH's); MS: m/z = 292 (M, 7%), 291 (16%), 264 (95%), 264 (95%), 221 (33%), 206 (25%), 180 (17%), 160 (56%), 130 (55%), 118 (18%), 110 (12%), 91 (20%), 77 (100%), 65 (19%), 51 (75%); Anal. Calcd. For $C_{17}H_{16}N_4O$ (292.34) C, 69.85; H, 5.52; N, 19.17 Found: C, 69.75; H, 5.48; N, 19.26%.

3.8.3. Ethyl 2-amino-6-(5-methyl-1-phenyl-1H-1,2,3-triazol-4-yl)pyridine-3-carboxylate (19).
Yellow crystals from AcOH, yield (73%), mp: 284-86°C; IR (KBr): 3467, 3325 (NH_2), 3066 (CH), 2923, 2854 (CH), 1708 (CO), 1624 (C=N), 1550 (C=C); ^{1}H NMR ($CDCl_3$): δ 1.27 (t, 3H, $CH_2\mathbf{CH_3}$), 2.60 (s, 3H, CH_3), (q, 2H, $\mathbf{CH_2}CH_3$), 7.11-8.23 (m, 9 H, ArH's); MS: m/z =323 (M, 3%), 295 (42%), 248 (38%), 221 (22%), 193 (35%), 166 (13%), 153 (18%), 152 (20%), 130 (28%), 118 (14%), 91 (14%), 77 (100%), 64 (20%), 51 (80%); Anal. Calcd. For $C_{17}H_{17}N_5O_2$ (323.35) C, 63.15; H, 5.30; N, 21.66 Found: C, 63.21; H, 5.19; N, 21.54%.

3.8.4. 6-(5-Methyl-1-phenyl-1H-1,2,3-triazol-4-yl)-2-phenylpyridine-3-carbonitrile (20).
Yellow crystals from EtOH, yield (73%), mp: 268-70°C; IR (KBr): 3066 (CH), 2923, 2827 (CH), 2233 (CN), 1624 (C=N), 1600 (C=C); ^{1}H NMR ($CDCl_3$): δ 2.62 (s, 3H, CH_3), 7.11-8.23 (m, 12 H, ArH's); MS: m/z = 373 (M, 0.6%), 294 (13%), 292 (12%), 267 (14%), 266 (98%), 264 (100%), 249 (15%), 247 (12%), 225 (29%), 213 (17%), 211 (18%), 154 (11%), 140 (18%), 102 (15%), 75 (12%), 43 (49%); Anal. Calcd. For $C_{21}H_{15}N_5$ (337.38) C, 74.76; H, 4.48; N, 20.76 Found: C, 74.67; H, 4.38; N, 20.62%.

3.8.5. 2-Amino-6-(5-methyl-1-phenyl-1H-1,2,3-triazol-4-yl)pyridine-3-carbonitrile (21).

Yellow crystals from EtOH, yield (73%), mp: 268-70°C; IR (KBr): 3413, 3340 (NH_2), 3028 (CH), 2954, 2823 (CH), 2237 (CN), 1643 (C=N), 1566 (C=C); ^{1}H NMR ($CDCl_3$): δ 2.62 (s, 3H, CH_3), 6.22 (s, 2H, NH_2), 7.11-8.23 (m, 7 H, ArH's); MS: m/z = 277 (M+1, 4.8%), 276 (M, 23%), 220 (17%), 219 (100%), 90 (6%); Anal. Calcd. For $C_{15}H_{12}N_6$ (276.3) C, 65.21; H, 4.38; N, 30.42 Found: C, 65.18; H, 4.44; N, 30.56%.

3.8.6. 3-Methyl-6-(5-methyl-1-phenyl-1H-1,2,3-triazol-4-yl)-1H-pyrazolo[3,4-b]pyridine (22a).

Yellow crystals from EtOH, yield (86%), mp: >300°C; IR (KBr): 3406 (NH), 3047 (CH), 2920, 2850 (CH), 1624 (C=N), 1546 (C=C); ^{1}H NMR ($CDCl_3$): δ 2.63 (s, 6H, $2CH_3$), 7.11-7.65 (m, 6 H, ArH's), 8.59-8.61 (d, 1H, *J* = 12Hz, ArH), 10.24 (s, br., 1H, NH); MS: m/z = 277 (M+1, 4.8%), 276 (M, 23%), 220 (17%), 219 (100%), 90 (6%); Anal. Calcd. For $C_{16}H_{14}N_6$ (290.32) C, 66.19; H, 4.86; N, 28.95 Found: C, 66.27; H, 4.82; N, 29.11%.

3.8.7. 3-Methyl-6-(5-methyl-1-phenyl-1H-1,2,3-triazol-4-yl)-1-phenyl-1H-pyrazolo[3,4-b]pyridine (22b).

Orange crystals from EtOH, yield (83%), mp: 192-94°C; IR (KBr): 3047 (CH), 2920, 2850 (CH), 1620 (C=N), 1589 (C=C); ^{1}H NMR ($CDCl_3$): δ 2.63 (s, 3H, CH_3), 2.73 (s, 3H, $2CH_3$), 7.11-8.05 (m, 12 H, ArH's); MS: m/z = 366 (M, 3%), 319 (4%), 279 (4%), 138 (12%), 125 (9%), 111 (15%), 109 (20%), 95 (18%), 81 (20%); Anal. Calcd. For $C_{22}H_{18}N_6$ (366.42) C, 72.11; H, 4.95; N, 22.94 Found: C, 72.11; H, 4.95; N, 22.94 %.

3.9. 2-Methyl-6-(5-methyl-1-phenyl-1H-1,2,3-triazol-4-yl)pyridine-3-carbohydrazide (23).

Equimolar amounts of ethyl 2-methyl-6-(5-methyl-1-phenyl-1*H*-1,2,3-triazol-4-yl)pyridine-3-carboxylate (**17**) and hydrazine hydrate (5 mmol for each) in ethanol (10 mL) were refluxed for 5 h. The resulting solid, was cooled and recrystallized to give **23** as white crystals from ethanol. Yield (89%), mp: 184-86°C; IR (KBr): 3296, 3190 (NH, NH2), 3062 (CH), 2920, 2850 (CH), 1685 (CO), 16270 (C=N), 1593 (C=C); ^{1}H NMR ($CDCl_3$): δ 2.63 (s, 3H, CH_3), 2.73 (s, 3H, CH_3), 6.24 (s, br., 3H, NH, NH_2), 7.11-8.38 (m, 7 H, ArH's); MS: m/z = 308 (M, 0.5%), 150 (20%), 123 (6%), 43 (12%), 28 (100%); Anal. Calcd. For $C_{16}H_{16}N_6O$ (308.34) C, 62.32; H, 5.23; N, 27.26 Found: C, 62.32; H, 5.23; N, 27.26 %.

3.10. (3,5-Dimethyl-1H-pyrazol-1-yl)(2-methyl-6-(5-methyl-1-phenyl-1H-1,2,3-triazol-4-yl)pyridin-3-yl)methanone (24) and 5-Methyl-2-[2-methyl-6-(5-methyl-1-phenyl-1H-[1,2,3]triazol-4-yl)-pyridine-3-carbonyl]-2,4-dihydro-pyrazol-3-one (25).

Equimolar amounts of 2-methyl-6-(5-methyl-1-phenyl-1*H*-1,2,3-triazol-4-yl)pyridine-3-carbohydrazide (**23**) and acetylacetone or ethyl acetoacetate (4 mmol for each) in ethanol (10 mL) with two drops of acetic acid were refluxed for 4 h. The resulting solid, so formed, was cooled and recrystallized from diluted acetic acid to give the corresponding **24** and **25**, respectively

3.10.1. (3,5-Dimethyl-1H-pyrazol-1-yl)(2-methyl-6-(5-methyl-1-phenyl-1H-1,2,3-triazol-4-yl)pyridin-3-yl)methanone (24).

White crystals from ethanol, yield (75%), mp: 200-202°C; IR (KBr): 3058 (CH), 2954, 2824, 2854 (CH), 1701 (CO), 1589 (C=C); ^{1}H NMR ($CDCl_3$): δ 2.28 (s, 3H, CH_3), 2.35 (s, 3H, CH_3), 2.62 (s, 3H, CH_3), 2.72 (s, 3H, CH_3), 6.87 (s, 1H, pyrazole H-4), 7.11-7.81 (m, 5 H, ArH's), 8.81-8.83 (d, 1H, *J* = 8 Hz, ArH), 9.11-9.13 (d, 1H, *J* = 8 Hz, ArH); MS: m/z = 373 (M+1, 25%), 372 (M, 100%), 355 (81%), 337 (10%), 310 (79%), 294 (8%), 282 (16%), 254 (80%), 226 (49%), 200 (6%), 155 (27%), 141 (10%), 113 (43%), 100 (6%); Anal. Calcd. For $C_{21}H_{20}N_6O$ (372.42) C, 67.73; H, 5.41; N, 22.57 Found: C, 67.67; H, 5.36; N, 22.64%.

3.10.2. 5-Methyl-2-[2-methyl-6-(5-methyl-1-phenyl-1H-[1,2,3]triazol-4-yl)-pyridine-3-carbonyl]-2,4-dihydro-pyrazol-3-one (25).

White crystals from ethanol, yield (72%), mp: 290-92°C; IR (KBr): 3058 (CH), 2920 2850 (CH), 1710 (CO), 1604 (C=C); ^{1}H NMR ($CDCl_3$): δ 2.21 (s, 3H, CH_3), 2.62 (s, 3H, CH_3), 2.72 (s, 3H, CH_3), 3.41 (q, 1H, CH_2 (pyrazole)), 3.62 (q, 1H, CH_2 (pyrazole)), 7.11-8.45 (m, 7 H, ArH's); MS: m/z = 374 (M, 0.01%), 153 (86%), 137 (100%), 106 (31%), 90 (18%), 89 (43%), 87 (30%), 77 (29%), 69 (15%); Anal. Calcd. For $C_{20}H_{18}N_6O_2$ (374.4) C, 64.16; H, 4.85; N, 22.45 Found: C, 64.08; H, 4.78; N, 22.53%.

3.11. (4-Arylazo 3,5-dimethyl-pyrazol-1-yl)-[2-methyl-6-(5-methyl-1-phenyl-1H-[1,2,3] triazol-4-yl)-pyridin-3-yl]-methanone (26) and 4-(Arylhydrazono)-5-methyl-2-[2-methyl-6-(5-methyl-1-phenyl-1H-[1,2,3]triazol-4-yl)-pyridine-3-carbonyl]-2,4-dihydro-pyrazol-3-one (27).

Method A. arenediazonium chlorides (5 mmole), which is prepared via reaction of the appropriate aniline or *p*-toluidene (5 mmole), hydrochloric acid (3 mL, 6 M), and sodium nitrite (0.37 g, 5 mmole) at 0–5_C, was added to a mixture of the appropriate **24** or **25** (5 mmole) and sodium acetate (0.66 gm, 5 mmole) in ethanol (30 mL) at 0–5^{o}C, while stirring. The reaction mixture was stirred for 3 h. The resulting solid, was collected, washed with water and recrystallized to give **26a,b** and **27a,b**, respectively.
Method B. A mixture of **23** and the appropriate 3-(2-arylhydrazono)pentane-2,4-dione or ethyl 2-arylazo-3-oxo-4-butanoate (5 mmol for each) in ethanol (20 mL) and catalytic amount of acetic acid (2 drops) was refluxed for 2h. The resulting solid, so formed, was collected and recrystallized from acetic acid to give product identical in all aspects with corresponding products obtained in Method A.

3.11.1. (4-Phenylazo-3,5-dimethyl-pyrazol-1-yl)-[2-methyl-6-(5-methyl-1-phenyl-1H-[1,2,3] triazol-4-yl)-pyridin-3-yl]-methanone (26a).

Brown crystals from ethanol, yield (70%), mp: 240-42^{o}C; IR (KBr): 3058 (CH), 2850 (CH), 1700 (CO), 1647 (C=N), 1547 (C=C); ^{1}H NMR ($CDCl_3$): δ 2.28 (s, 3H, CH_3), 2.35 (s, 3H, CH_3), 2.62 (s, 3H, CH_3), 2.72 (s, 3H, CH_3), 7.11-7.81 (m, 10 H, ArH's), 8.81-8.83 (d, 1H, *J* = 8 Hz, ArH), 9.11-9.13 (d, 1H, *J* = 8 Hz, ArH); MS: m/z = 476 (M, 11%), 475 (5%), 432 (7%), 418 (6%), 376 (11%), 362 (14%), 348 (17%), 334 (27%), 320 (35%), 305 (18%), 278 (36%), 237 (37%), 220 (17%), 207 (85%), 195 (10%), 191 (15%), 181 (32%), 165 (100%), 119 (14%), 69 (7%), 43 (36%); Anal. Calcd. For $C_{27}H_{24}N_8O$ (476.53) C, 68.05; H, 5.08; N, 23.51 Found: C, 68.12; H, 5.10; N, 23.68%.

3.11.2. (4-p-Tolylazo-3,5-dimethyl-pyrazol-1-yl)-[2-methyl-6-(5-methyl-1-phenyl-1H-[1,2,3] triazol-4-yl)-pyridin-3-yl]-methanone (26b).

Brown crystals from acetic acid, yield (79%), mp: 222-24^{o}C; IR (KBr): 3058 (CH), 2927 (CH), 1701 (CO), 1647 (C=N), 1589 (C=C); ^{1}H NMR ($CDCl_3$): δ 2.11 (s, 3H, CH_3), 2.28 (s, 3H, CH_3), 2.35 (s, 3H, CH_3), 2.62 (s, 3H, CH_3), 2.72 (s, 3H, CH_3), 7.11-7.81 (m, 9 H, ArH's), 8.81-8.83 (d, 1H, *J* = 8 Hz, ArH), 9.11-9.13 (d, 1H, *J* = 8 Hz, ArH); MS: m/z = 490 (M, 8%), 489 (8%), 419 (11%), 353 (22%), 333 (11%), 308 (9%), 290 (11%), 269 (14%), 250 (13%), 223 (14%), 169 (16%), 156 (25%), 138 (21%), 129 (19%), 121 (24%), 115 (20%), 97 (35%), 96 (48%), 85 (26%), 82 (26%), 71 (72%, 69 (99%)); Anal. Calcd. For $C_{28}H_{26}N_8O$ (490.56) C, 68.55; H, 5.34; N, 22.84 Found: C, 68.62; H, 5.42; N, 22.94%.

3.11.3. 4-(Phenylhydrazono)-5-methyl-2-[2-methyl-6-(5-methyl-1-phenyl-1H-[1,2,3]triazol-4-yl)-pyridine-3-carbonyl]-2,4-dihydro-pyrazol-3-one (27a).

Brown crystals from ethanol, yield (74%), mp: 268-70^{o}C; IR (KBr): 3386 (NH), 3074 (CH), 2920, 2850 (CH), 1716, 1658 (CO), 1593 (C=C); ^{1}H NMR ($CDCl_3$): δ 2.01 (s, 3H, CH_3), 2.62 (s, 3H, CH_3), 2.72 (s, 3H, CH_3), 7.11-7.65 (m, 12 H, ArH's), 13.78 (s, br., 1H, NH); MS: m/z = 478 (M, 87%), 349 (27%), 321 (24%), 304 (19%), 293 (16%), 277 (26%), 276 (25%), 275 (20%), 249 (41%), 232 (24%), 219 (18%), 205 (36%), 192 (21%), 179 (13%), 166 (29%), 152 (57%), 140 (14%), 125 (14%), 77 (6%), 29 (100%); Anal. Calcd. For $C_{26}H_{22}N_8O_2$ (478.51) C, 65.26; H, 4.63; N, 23.42 Found: C, 65.19; H, 4.57; N, 23.26%.

3.11.4. 4-(p-Tolylhydrazono)-5-methyl-2-[2-methyl-6-(5-methyl-1-phenyl-1H-[1,2,3]triazol-4-yl)-pyridine-3-carbonyl]-2,4-dihydro-pyrazol-3-one (27b).

Beige crystals from acetic acid, yield (52%), mp: 252-54^{o}C; IR (KBr): 3352 (NH), 3074 (CH), 2927, 2854 (CH), 1701, 1658 (CO), 1589 (C=C); ^{1}H NMR ($CDCl_3$): δ 2.01 (s, 3H, CH_3), 2.31 (s, 3H, CH_3), 2.62 (s, 3H, CH_3), 2.72 (s, 3H, CH_3), 7.11-7.65 (m, 11 H, ArH's), 14.11 (s, br., 1H, NH); MS: m/z = 493 (M+1, 9%), 396 (15%), 381 (11%), 357 (9%), 349 (10%), 327 (9%), 320 (14%), 305 (11%), 286 (11%), 195 (9%), 135 (13%), 123 (13%), 113 (18%), 104 (10%), 98 (15%), 60 (26%), 86 (8%); Anal. Calcd. For $C_{27}H_{24}N_8O_2$ (492.53) C, 65.84; H, 4.91; N, 22.75 Found: C, 65.92; H, 4.87; N, 22.64%.

3.12. Azido(2-methyl-6-(5-methyl-1-phenyl-1H-1,2,3-triazol-4-yl)pyridin-3-yl)methanone (30).

To a stirred solution of 2-methyl-6-(5-methyl-1-phenyl-1*H*-1,2,3-triazol-4-yl)pyridine-3-carbohydrazide (**23**) (5 mmole) in acetic acid (15 mL) at 0–5^{o}C, sodium nitrite was added portion-wise tell effervescence ended. The reaction mixture stirred for 1 h. The resulting solid, was collected, filtered, washed with water, and recrystallized to give the corresponding **30**, as buff crystals, from acetic acid, yield (86%), mp: 150-52^{o}C; IR (KBr): 3066 (CH), 2974 (CH), 2137 (azide group), 1685 (CO), 1500 (C=C); ^{1}H NMR ($CDCl_3$): δ 2.63 (s, 3H, CH_3), 2.68 (s, 3H, CH_3), 7.62-7.66 (m, 5 H, ArH's), 7.92-7.94 (m, 2H, ArH's), 9.18 (s, 1H. ArH); MS: m/z = 319 (M, 45%), 285 (16%), 270 (45%), 240 (10%), 225 (14%), 193 (8%), 179 (22%), 166 (14%), 108 (100%), 80 (7%), 77 (7%); Anal. Calcd. For $C_{16}H_{13}N_7O$ (319.12) C, 60.18; H, 4.10; N, 30.70 Found: C, 60.24; H, 4.17; N, 30.62 %.

3.13. 1-(2-Methyl-6-(5-methyl-1-phenyl-1H-1,2,3-triazol-4-yl)pyridin-3-yl)-3-substituted urea 31a-c and 32.
A mixture of **30** (1.59 g, 5 mmol) and appropriate aniline, *p*-toluidine, *p*-anisidine or 3-amino-5-phenylpyrazole (5 mmol) in dry dioxane (20 mL) was refluxed for 4 h. The resulting solid, so formed, was collected and recrystallized to give **31a-c** and **32**, respectively.

3.13.1. 1-(2-Methyl-6-(5-methyl-1-phenyl-1H-1,2,3-triazol-4-yl)pyridin-3-yl)-3-phenylurea 31a.
White crystals, from DMF, yield (77%), mp: 240-42^{o}C; IR (KBr): 3375 (NH), 3058 (CH), 2920 (CH), 1705 (CO), 1596 (C=C); ^{1}H NMR ($CDCl_3$): δ 2.21 (s, 3H, CH_3), 2.64 (s, 3H, CH_3), 7.11-7.67 (m, 11 H, ArH's), 8.29-8.31 (m, 1H, ArH's), 8.88 (s, br., 2H. 2NH); MS: m/z = 384 (M, 0.1%), 276 (13%), 233 (5%), 131 (11%), 91 (100%), 79 (11%); Anal. Calcd. For $C_{22}H_{20}N_6O$ (384.17) C, 68.73; H, 5.24; N, 21.86 Found: C, 68.81; H, 5.324; N, 21.79%.

3.13.2. 1-(2-Methyl-6-(5-methyl-1-phenyl-1H-1,2,3-triazol-4-yl)pyridin-3-yl)-3-p-tolylurea 31b.

Grey crystals, from DMF, yield (91%), mp: 248-50^{o}C; IR (KBr): 3278 (NH), 3062 (CH), 2923 (CH), 1700 (CO), 1639 (C=N), 1581 (C=C); ^{1}H NMR ($CDCl_3$): δ 2.15 (s, 3H, CH_3), 2.26 (s, 3H, CH_3), 2.64 (s, 3H, CH_3), 7.11-7.67 (m, 10 H, ArH's), 8.29-8.31 (m, 1H, ArH's), 8.88 (s, br., 2H. 2NH); MS: m/z = 389 (M, 0.1%), 280 (15%), 268 (13%), 267 (63%), 148 (28%), 105 (100%), 77 (27%); Anal. Calcd. For $C_{23}H_{22}N_6O$ (398.46) C, 69.33; H, 5.57; N, 21.09 Found: C, 69.29; H, 5.48; N, 21.12%.

3.13.3 1-(4-Methoxyphenyl)-3-(2-methyl-6-(5-methyl-1-phenyl-1H-1,2,3-triazol-4-yl)pyridin-3-yl)urea 31c.

Brown crystals, from DMF, yield (73%), mp: 278-80^{o}C; IR (KBr): 3298 (NH), 3062 (CH), 2920 (CH), 1695 (CO), 1643 (C=N), 1604 (C=C); ^{1}H NMR ($CDCl_3$): δ 2.46 (s, 3H, CH_3), 2.64 (s, 3H, CH_3), 3.68 (s, 3H, OCH_3), 7.11-7.67 (m, 10 H, ArH's), 8.29-8.31 (m, 1H, ArH's), 8.33 (s, br., 1H. 1NH), 8.85 (s, br., 1H. 1NH); MS: m/z = 414 (M, 8%), 351 (9%), 295 (30%), 254 (11%), 253 (55%), 237 (11%), 235 (11%), 209 (8%), 207 (14%), 185 (14%), 184 (100%), 168 (11%), 127 (6%); Anal. Calcd. For $C_{23}H_{22}N_6O_2$ (414.18) C, 66.65; H, 5.35; N, 20.28 Found: C, 66.61; H, 5.39; N, 20.34%.

3.13.4. 1-(2-Methyl-6-(5-methyl-1-phenyl-1H-1,2,3-triazol-4-yl)pyridin-3-yl)-3-(3-phenyl-1H-pyrazol-5-yl)urea (32).
Beige crystals, from EtOH, yield (85%), mp: 210-12^{o}C; IR (KBr): 3275 (NH), 3062 (CH), 2923, 2854 (CH), 1690 (CO), 1577 (C=C); ^{1}H NMR ($CDCl_3$): δ 2.47 (s, 3H, CH_3), 2.64 (s, 3H, CH_3), 6.56 (s, 1H, pyrazole H-4), 7.42-7.99 (m, 12 H, ArH's), 8.47 (s, br., 1H. 1NH), 9.57 (s, br., 1H. 1NH), 12.54 (s, br., 1H. 1NH); MS: m/z = 450 (M, 3.4%), 447 (5%), 432 (7%), 404 (9%), 390 (9%), 376 (11%), 362 (14%), 334 (27%), 320 (35%), 319 (11%), 306 (20%), 392 (17^), 278 (36%), 237 (37%), 220 (17%), 207 (85%), 191 (15%), 181 (32%), 165 (100%), 119 (14%), 69 (7%)57 (11%), 43 (36%); Anal. Calcd. For $C_{25}H_{22}N_8O$ (450.5) C, 66.65; H, 4.92; N, 24.87 Found: C, C, 66.69; H, 5.12; N, 24.72%.

3.14. 3-(2-Methyl-6-(5-methyl-1-phenyl-1H-1,2,3-triazol-4-yl)pyridin-3-yl)quinazoline-2,4(1H,3H)-dione (33).
A mixture of **30** (1.59 g, 5 mmol) and appropriate anthranilic acid or methyl anthranilate (5 mmol) in dry dioxane (20 mL) was refluxed for 4 h. The resulting solid, so formed, was collected and recrystallized to give **33** as white crystals, from DMF, yield (77%), mp: 240-42^{o}C; IR (KBr): 3278 (NH), 3062 (CH), 2920, 2850 (CH), 1662 (CO), 1593 (C=C); ^{1}H NMR ($CDCl_3$): δ 2.47 (s, 3H, CH_3), 2.64 (s, 3H, CH_3), 7.21-7.99 (m, 11 H, ArH's), 10.54 (s, br., 1H. 1NH); MS: m/z = 410 (M, 77%), 396 (29%), 395 (100%), 205 (14%), 197 (10%), 108 (20%); Anal. Calcd. For $C_{23}H_{18}N_6O_2$ (410.43) C, 67.31; H, 4.42; N, 20.48 Found: C, 67.18; H, 4.32; N, 20.55%.

3.15. ***Aryl 2-methyl-6-(5-methyl-1-phenyl-1H-1,2,3-triazol-4-yl)pyridin-3-ylcarbamate 34a-c.***
A mixture of **30** (1.59 g, 5 mmol) and appropriate phenol, p-nitrophenol or picric acid (5 mmol) in dry benzene (20 mL) was refluxed for 4 h. The resulting solid, so formed, was collected and recrystallized to give **34a-c**, respectively.

3.16.1. Phenyl 2-Methyl-6-(5-methyl-1-phenyl-1H-1,2,3-triazol-4-yl)pyridin-3-ylcarbamate (34a) as beige crystals, from dioxane, yield (77%), mp: 224-26^{o}C; IR (KBr): 2897 (CH), 1669 (CO), 1624 C=N); ^{1}H NMR ($CDCl_3$): δ 2.11 (s, 3H, CH_3), 2.64 (s, 3H, CH_3), 6.70-6.77 (m, 11H, ArH's), 8.28-8.30 (d, 1H, J = 8Hz, ArH), 9.44 (s, br., 1H, NH);

MS: m/z = 385 (M, 11%), 384 (M-1, 23%), 383 (M-2, 100%), 171 (14%), 170 (100%), 168 (20%), 154 (15%), 149 (18%), 127 (10%), 122 (5%); Anal. Calcd. For $C_{22}H_{19}N_5O_2$ (385.42) C, 68.56; H, 4.97; N, 18.17; O Found: C, 68.622; H, 5.14; N, 18.25; O%.

3.16.2. 4-Nitrophenyl 2-methyl-6-(5-methyl-1-phenyl-1H-1,2,3-triazol-4-yl)pyridin-3-ylcarbamate (34b). as yellow crystals, from AcOH, yield (81%), mp: >300^oC; IR (KBr):3433 (NH), 3062 (CH), 1700 (CO), 1627 (C=N), 1512, 1373 (NO_2); ^1H NMR ($CDCl_3$): δ 2.21 (s, 3H, CH_3), 2.65 (s, 3H, CH_3), 6.58 (s, br., 1H, NH), 7.17-8.22 (m, 11H, ArH's); MS: m/z = 430 (M, 0.06%), 139 (69%), 109 (28%), 93 (40%), 81 (33%), 69 (14%), 65 (100%), 63 (25%); Anal. Calcd. For $C_{22}H_{18}N_6O_4$ (430.42) C, 61.39; H, 4.22; N, 19.53 Found: C, 61.39; H, 4.22; N, 19.53%.

***3.16.3. 2,4,6-Trinitrophenyl 2-methyl-6-(5-methyl-1-phenyl-1H-1,2,3-triazol-4-yl)-pyridin-3-ylcarbamate (34c*)** as beige crystals, from dioxane, yield (70%), mp: 282-84^oC; IR (KBr): 3274 (NH), 3060, 2916, 2850 (CH), 1700 (CO), 1635 (C=N), 1585 (C=C); ^1H NMR ($CDCl_3$): δ 2.21 (s, 3H, CH_3), 2.65 (s, 3H, CH_3), 6.88 (s, br., 1H, NH), 7.17-7.66 (m, 7H, ArH's), 9.11 (s, 2H, ArH's); MS: m/z = 521 (M+1, 37%), 505 (36%), 504 (100%), 492 (13%), 491 (17%), 478 (10%), 373 (9%), 270 (48%), 269 (42%), 252 (17%), 223 (11%), 176 (20%), 121 (41%), 106 (12%), 83 (11%), 71 (11%), 57 (28%), 43 (46%) 41 (26%); Anal. Calcd. For $C_{22}H_{16}N_8O_8$ (520.11) C, 50.77; H, 3.10; N, 21.53 Found: C, 50.82; H, 3.17; N, 21.48%

4. Antimicrobial Activity

Antimicrobial activity of the tested samples was determined using a modified Kirby-Bauer disc diffusion method.[22] Briefly, 100 µl of the test bacteria/fungi were grown in 10 ml of fresh media until they reached a count of approximately 108 cells/ml for bacteria or 105 cells/ml for fungi [23] 100 µl of microbial suspension was spread onto agar plates corresponding to the broth in which they were maintained.

Isolated colonies of each organism that might be playing a pathogenic role should be selected from primary agar plates and tested for susceptibility by disc diffusion method [24]**.**

Of the many media available, NCCLS recommends Mueller-Hinton agar due to: it results in good batch-to-batch reproducibility

Disc diffusion method for filamentous fungi tested by using approved standard method (M38-A) developed by the [25] for evaluating the susceptibilities of filamentous fungi to antifungal agents.

Disc diffusion method for yeasts developed by using approved standard method (M44-P) [26].

Plates inoculated with filamentous fungi as ***Aspergillus fumigatus*** at 25°C for 48 hours; Gram (+) bacteria as ***Staphylococcus aureus, Bacillus subtilis;*** Gram (-) bacteria as ***Escherichia coli, Salmonella typhimurium*** they were incubated at 35-37°C for 24-48 hours and a diploid fungus as ***Candida albicans*** incubated at 30°C for 24-48 hours and, then the diameters of the inhibition zones were measured in millimeters [22]**.**

Standard discs of **Tetracycline** (Antibacterial agent), **Clotrimazole** (Antifungal agent) served as positive controls for antimicrobial activity but filter discs impregnated with 10 µl of solvent (distilled water, chloroform, DMSO) were used as a negative control.

The agar used is Meuller-Hinton agar that is rigorously tested for composition and pH. Further the depth of the agar in the plate is a factor to be considered in the disc diffusion method. This method is well documented and standard zones of inhibition have been determined for susceptible and resistant values.

Blank paper disks (Schleicher & Schuell, Spain) with a diameter of 8.0 mm were impregnated l0µ, of tested concentration of the stock solutions.

When a filter paper disc impregnated with a tested chemical is placed on agar the chemical will diffuse from the disc into the agar. This diffusion will place the chemical in the agar only around the disc. The solubility of the chemical and its molecular size will determine the size of the area of chemical infiltration around the disc. If an organism is placed on the agar it will not grow in the area around the disc if it is susceptible to the chemical. This area of no growth around the disc is known as a **"Zone of inhibition"** or" **Clear zone".**

For the disc diffusion, the zone diameters were measured with slipping calipers of the National Committee for Clinical Laboratory Standards [27].

Agar-based methods such as Etest and disk diffusion can be good alternatives because they are simpler and faster than broth-based methods [28, 29].

Table 1: Response of various microorganisms to some synthesized compounds in *vitro* culture.

Inhibition Zone diameter (mm/mg sample)
Antimicrobial Activity%(Inhibition Zone%)

SAMPLE	*A.fumigatus*	*C.albicans*	*S.aureus*	*B.subtilis*	*E.coli*	*S.typhimurium*
DMSO (positive control)	0.0	0.0	0.0	0.0	0.0	0.0
Tetracycline (Antibacterial agent)	– –	– –	30	29	31	30
Clotrimazole (Antifungal agent)	24	22	– –	– –	– –	– –
	5 21%	7 32%	17 57%	21 72%	28 90%	26 87%
	3 13%	4 18%	0.0 0.0%	0.0 0.0%	0.0 0.0%	0.0 0.0%
	14 58%	18 82%	29 97%	27 93%	25 81%	27 90%
	12 50%	17 77%	27 90%	22 76%	29 94%	25 83%
	13 54%	17 77%	9 30%	10 34%	5 16%	3 10%
	21 88%	20 91%	18 60%	21 72%	19 61%	25 83%
	6 25%	0.0 0.0%	5 17%	4 14%	2 6%	3 10%

-Strong Antimicrobial activity means: Antimicrobial activity % ≥60 %
- Medium Antimicrobial activity means: 30% ≥Antimicrobial activity % > 60%
- Weak Antimicrobial activity means: 0.0% >Antimicrobial activity %> 30%

5. Conclusion

The studies described above clearly demonstrate that the new pyrazolo[5,l-*c*]triazines, [l,2,4]triazolo[4,3-*c*]triazine, benzo[4,5]-imidazo[2,1-*c*][1,2,4]triazine, pyridines, urea and carbamate derivatives containing 1,2,3,-triazole moiety can be synthesized in a good yields *via* sodium salt of 3-hydroxy-1-(5-methyl-1-phenyl-1*H*-1,2,3-triazol-4-yl)prop-2-en-1-one

6. References

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