Synthesis of Some New Pyridones, Fused Pyrimidines, and Fused 1,2,4-Triazines

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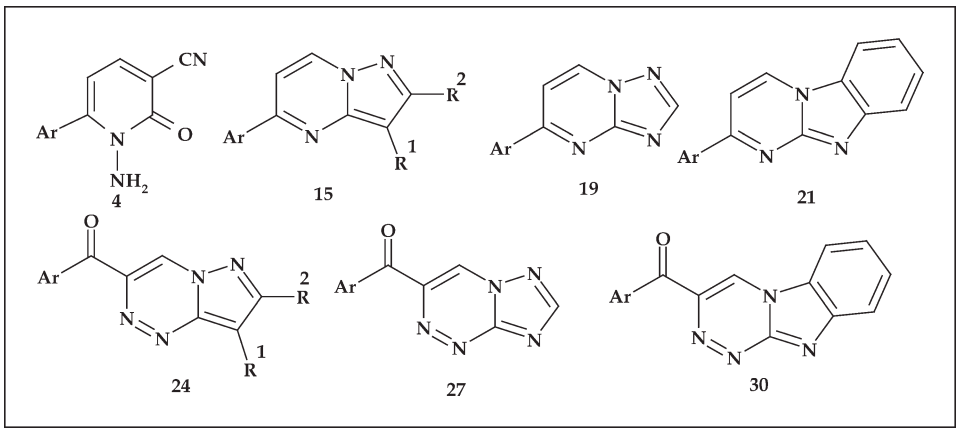

Cyclocondensation of hydrazides, 3-aminopyrazoles, 3-amino-1,2,4-triazole, 2-aminobenzoimidazole, pyrazole-3-diazonium salts, 1,2,4-triazol-3-diazonium salt, or benzoimidazole-2-diazonium salt with sodium salt of 3-hydroxy-1-(2-naphthyl)prop-2-en-1-one gave 2-pyridones, pyrazolo[1,5-*a*]pyrimidines, triazolo[1,5-*a*]pyrimidine, benzo[4,5]imidazo[1,2-*a*]pyrimidine, (pyrazolo[5,1-*c*][1,2,4]triazin-3-yl)-methanone, [1,2,4]triazolo[5,1-*c*][1,2,4]triazin-3-yl-methanone, or benzo[4,5]imidazo[2,1-*c*] [1,2,4]triazin-3-yl-methanone derivatives, respectively, which will be tested for anti-tumor and anti-cancer activates.

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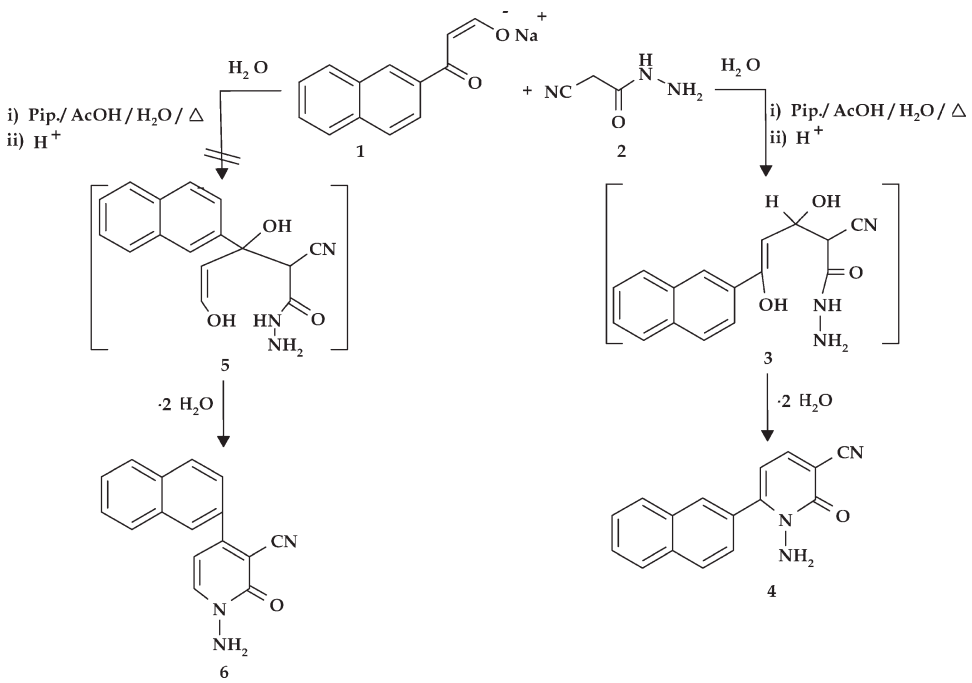
INTRODUCTION

Chemotherapeutic anticancer agents continue to be an active area of research at many companies and research centers [1–3]. So that, pyridine, pyrazolopyrimidine, and trazine derivatives have received considerable attention due to their wide range of applications in chemotherapy, such as anti-inflammatory, anti-tumor, anti-mycobacterial, anti-fungal, and anti-viral activities [4–6]. Recently, it has been reported that many pyridine and pyrazolopyrimidine derivatives showed strong cytotoxicity against several human cancer cell lines [6–9]. Pyrazolo[4,3-*d*]pyrimidine and pyrazolo[1,5-*a*]pyrimidine derivatives were reported to be inhibitors of tyrosine kinase and cyclin-dependent kinases (CDK) which are involved in mediating the transmission of mitogenic signals and numerous other cellular events [3,10–15], including cell proliferation, migration, differentiation, metabolism, and immune response. It was also found that many of these derivatives may block proliferation of various cancer cell lines [16]. In view of these reports and in continuation with the previous work, here a successful trial for synthesizing new derivatives of pyridones, pyrazolo[1,5-*a*]pyrimidines, triazolo[1,5-*a*]pyrimidine, imidazo[1,2-*a*]pyrimidine, pyrazolo[5,1-*c*]triazine, triazolo[5,1-*c*]triazine, and imidazo [2,1-*c*]triazine derivatives which will be tested for anti-tumor and anti-cancer activates are reported.

RESULTS AND DISCUSSION

As a part of our program directed for the development of efficient and simple procedures for the synthesis of antimetabolites [17–19], we have recently reported different and successful approaches for the synthesis of 2-pyridones [17,19]. The synthesized compounds act as intermediates for synthesis of deazafolic acid ring system and deazapyrimidine nucleosides, which reported to be significantly active, both *in vitro* and *in vivo* [20,21]. They also act as inhibitor of dihydrofolate reductase [22], cytotoxicity against various experimental tumors as potential as methotrexate [23,24], and one of the most effective antimetabolites currently used in treatment of various solid tumors [25,26]. This prompted our interest to the synthesis and study of the chemistry of this class of compounds [17,18]. Although *N*-amino-2-pyridones have proved to be useful synthetic intermediates, there are few procedures for their preparation and they are usually obtained in low yield by the reaction of hydrazine with 2-pyridones [27,28]. I have reported in this part, one step synthesis of 2-pyridones from the reaction

Scheme 1



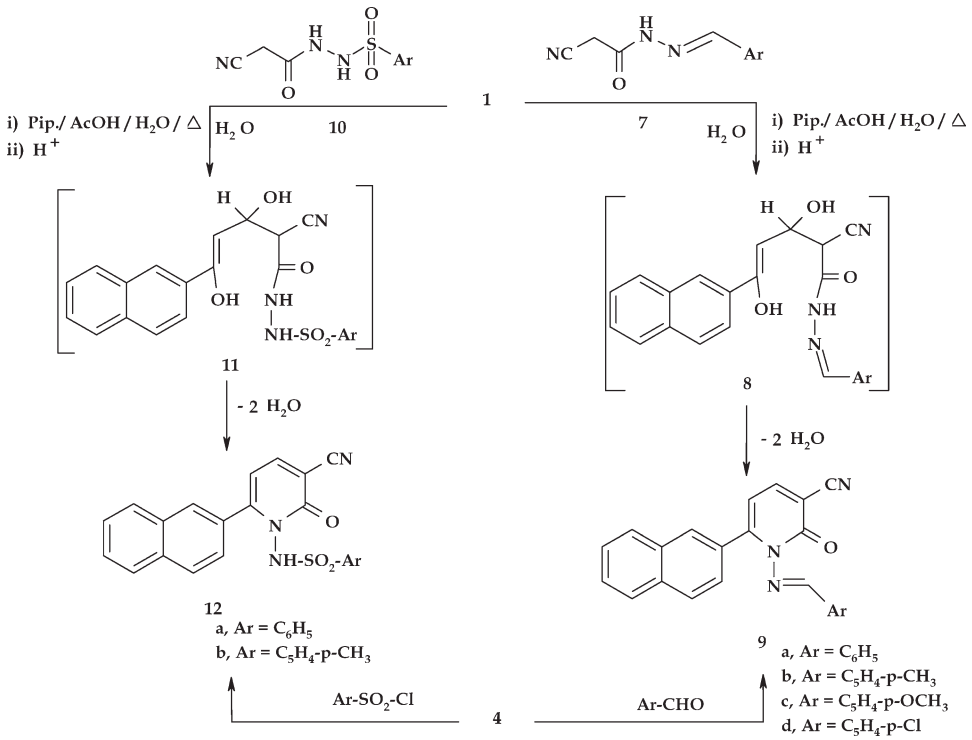
of 3-hydroxy-1-(2-naphthyl)prop-2-en-1-one **1** with different hydrazides. Thus, it has been found that salt **1** reacted with cyanoacetic acidhydrazide **2** to give a trisubstituted 2-pyridone. Two modes of cyclization are feasible, giving a 1,2,3,6- or 1,2,3,4-tetrasubstituted products, as outlined in Scheme 1. First, initial attack by a methylene carbanion takes place at the formyl group of the salt **1** and subsequent Michael cyclization followed by elimination of two moles of water to give the 1,2,3,6-trisubstituted product **4**. Second, initial nucleophilic attack by the methylene carbon takes place at the ketonic group, followed by cyclization and elimination of water giving 1,2,3,4-trisubstituted isomer **6**. In fact, only one isomer was obtained, which was suggested to be **4** due to the fact that initial attack of the active methylene carbon at the unhindered formyl group leading to **4** being much more probable than attack at the hindered and electronically disfavored ketonic group [29]. Spectral studies did not allow us to distinguish between structures **4** and **6**. ^{1}H-NMR for the product revealed the absence of CH_2 and the presence of 2-naphthyl protons from $\delta = 7.54$–8.66 ppm whereas the amino group has two protons at $\delta = 2.70$ ppm in solution. No significant amounts of the alternative regioisomers could be detected. To establish structure of the product, the crystal structure of a similar previous work has been reported [19,29,30]. The X-ray analysis of this work confirms the exclusive presence of the regioisomer **4** [29].

The reaction of sodium salts of 3-hydroxy-1-(2-naphthyl)prop-2-en-1-one **1** with arylidenecyanoacetohydrazide **7** and arylsulfonylcyanoacetohydrazide **10** derivatives represents a novel, one step, synthesis of *N*-arylideneamino- and *N*-arylsulphonylamino-2-pyridones, respectively. Thus, it has been found that, salt **1** reacted with arylidenecyanoacetohydrazide **7** to give *N*-arylideneamino-2-pyridones **9,** whereas **1** reacted with arylsufonylcyanoacetohydrazide **10** to give *N*-arylsulfonylamino-2-pyridones **12**. The structure of compound **12b** was established on the basis of its elemental analysis and spectral data (IR, ^{1}H-NMR, and MS). Thus, the mass spectra of **12b** was compatible with the molecular formula $C_{23}H_{17}N_3O_3S$ (M^+ 415) and the ^{1}H-NMR showed a signal at $\delta = 2.35$ ppm assigned for methyl group. Moreover, both **7** and **12** can be obtained from the reaction of **4** with aldehydes or arylsufonylchlorides, respectively, as shown in Scheme 2.

Analogously, treatment of sodium 3-hydroxy-1-(2-naphthyl)prop-2-en-1-one **1** with appropriate amounts of 3-aminopyrazoles **13** [31], in the presence of piperidine acetate afforded pyrazolo[1,5-*a*]pyrimidines **15**. Structure of the product **15b** was confirmed by elemental analysis and spectral data (IR, ^{1}H–NMR, and MS). The mass spectra of **15b** was compatible with the molecular formula $C_{23}H_{17}N_3$ (M^+ 335) and ^{1}H NMR for the product revealed the presence of a CH_3 protons at $\delta = 2.41$ ppm and methine H of pyrazole at $\delta = 6.22$ ppm and methine 2H of pyrimidine at $\delta = 8.52$ and 8.78 ppm in solutions as shown in Figure 1.

The reaction seemed to proceed via initial nucleophilic attack by the exocyclic amino group of

Scheme 2



aminopyrazoles **13** at the ketonic group, which formed *in situ* from salt **1** with water, followed by cyclization and elimination of two moles of water to give the products **15** Scheme 3. It has been suggested that the formation of the alternative isomeric products **17** is based on the initial attack of endocyclic amino group at the ketonic group. The latter suggestion is excluded due to the higher nucleophilicity of the exocyclic primary amino group than the endocyclic amino group which previously reported [17,29,30].

Similarly, treatment of sodium 3-hydroxy-1-(2-naphthyl)prop-2-en-1-one **1** with 3-amino-1,2,4-triazole or 2-aminobenzoimidazole in the presence of piperidine acetate afforded 5-(naphthalen-2-yl)[1,2,4]triazolo[1,5-*a*]pyrimidine **19** or 2-(naphthalen-2-yl)benzo[4,5]imidazo[1,2-*a*]pyrimidine **21**, respectively, Scheme 4. The structure of **19** was confirmed by elemental analysis and spectral data (IR, ^{1}H–NMR, and MS). The mass spectra of **19** was compatible with the molecular formula $C_{15}H_{10}N_4$ (M^+ 246) and ^{1}H NMR for the product revealed the presence methine H of triazole at $\delta = 8.27$ ppm and methine 2H of pyrimidine at $\delta = 7.54$ and 8.52 ppm.

Finally, the reaction of sodium 3-hydroxy-1-(2-naphthyl)prop-2-en-1-one **1** with pyrazole-3-diazonium salts **22**, 1,2,4-triazol-3-diazonium salt **25,** or benzoimidazole-2-diazonium salt **28** in the presence of piperidine acetate afforded naphthalen-2-yl-(pyrazolo[5,1-*c*][1,2,4] triazin-3-yl)-methanone derivatives **24,** naphthalene-2-yl-[1,2,4]triazolo[5,1-*c*][1,2,4]triazin-3-yl-methanone **27,** or benzo[4,5] imidazo[2,1-*c*][1,2,4]triazin-3-yl-naphthalen-2-yl-methanone **30,** respectively, Scheme 5. The structure of **24** was confirmed by elemental analysis and spectral data (IR, ^{1}H-NMR, and MS). The mass spectra of **24a** was compatible with the molecular formula $C_{22}H_{14}N_4O$ (M^+ 350) and IR for the product revealed the presence carbonyl group at 1,665 cm^{-1}.

EXPERIMENTAL

All melting points are uncorrected IR spectra were obtained (KBr disk) on a Perkin Elmer 11650 FT-IR instrument. The ^{1}H-NMR spectra were measured on a Varian 400 MHz spectrometer for solutions in $(CD_3)_2$ SO using $Si(CH_3)_4$ as an internal standard. Mass spectra were recorded on a Varian MAT 112 spectrometer. Analytical data were obtained from the Microanalytical Data Center at Cairo University.

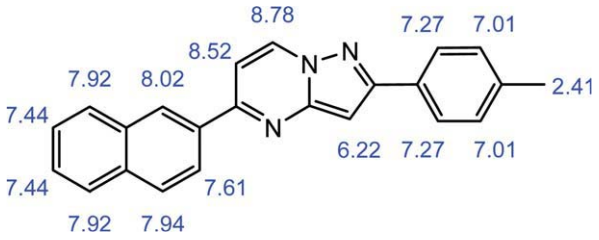
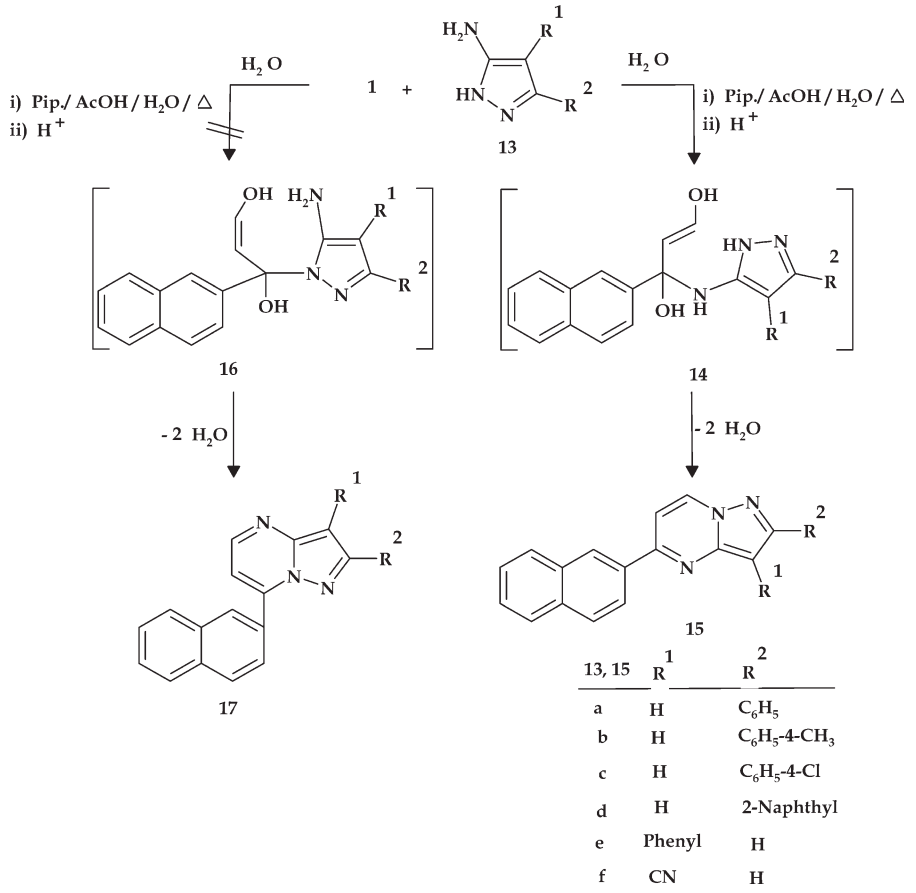


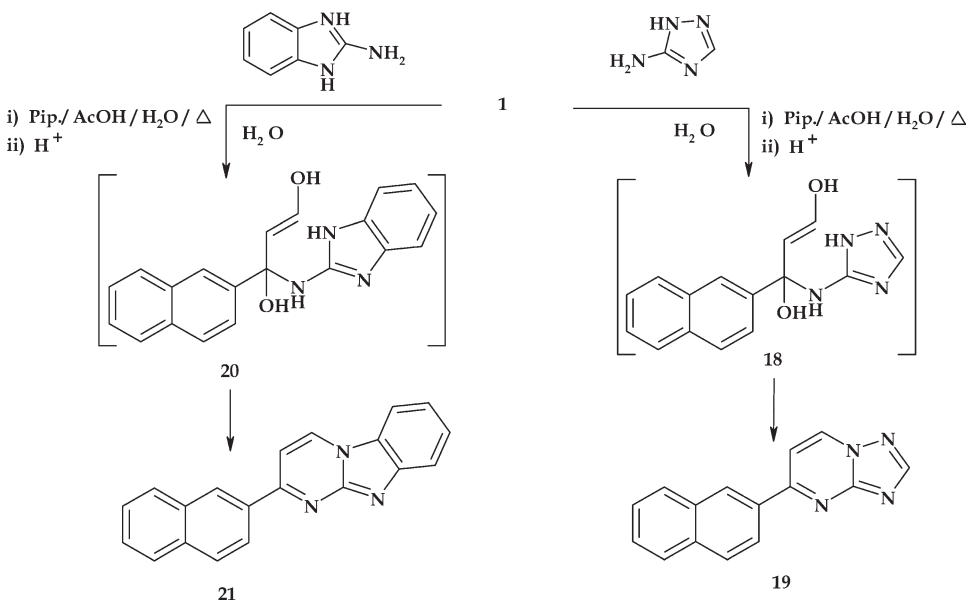
Figure 1. ^{1}H NMR δ values for **15b.**

Scheme 3

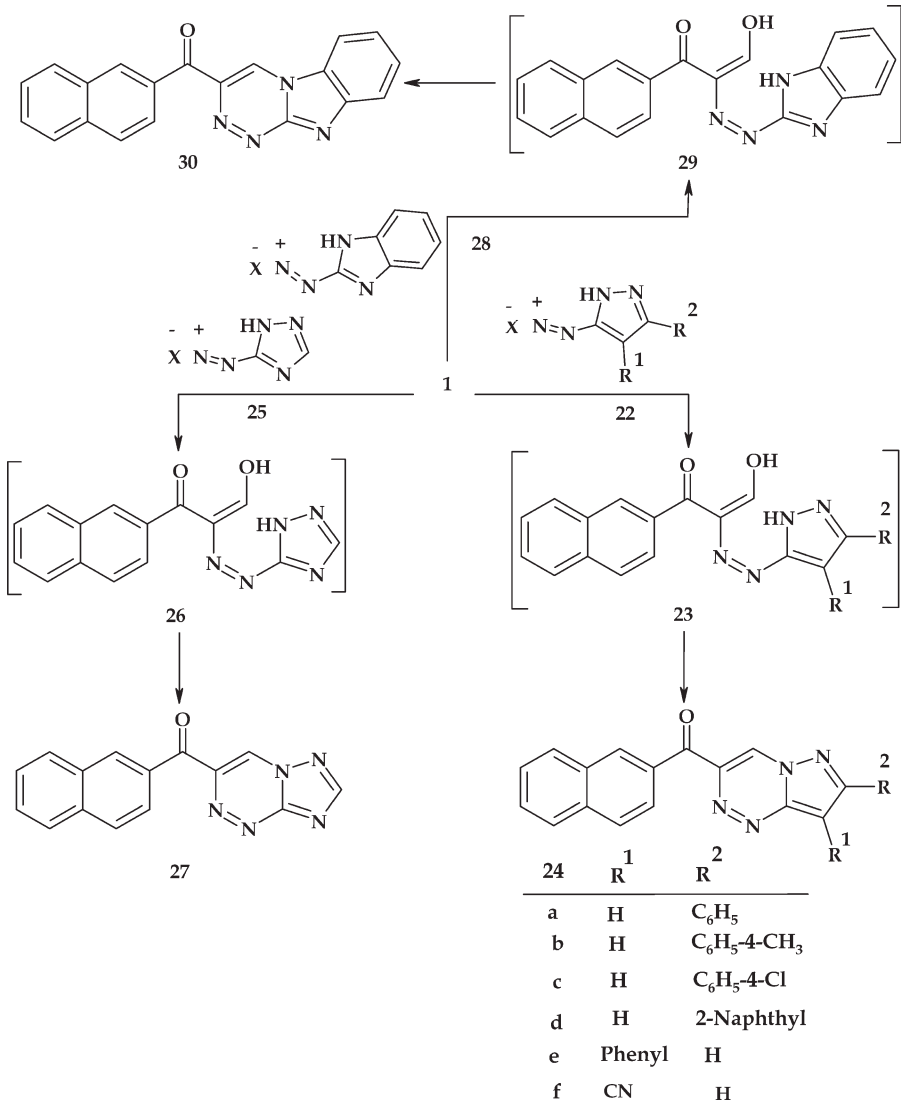


13, 15	R^1	R^2
a	H	C_6H_5
b	H	C_6H_5-4-CH_3
c	H	C_6H_5-4-Cl
d	H	2-Naphthyl
e	Phenyl	H
f	CN	H

Scheme 4



Scheme 5



2-Pyridones (4), (9a–d), (12a,b), Pyrazolo[1,5-*a*]pyrimidines (15a–f), Triazolo[1,5-*a*]pyrimidine (19), Benzo[4,5]imidazo[1,2-*a*]pyrimidine (21), (pyrazolo [5,1-*c*][1,2,4] triazin-3-yl)-methanones (24a–f), [1,2,4] Triazolo[5,1-*c*] [1,2,4]triazin-3-yl-methanone (27), or Benzo[4,5] imidazo [2,1-*c*][1,2,4]triazin-3-yl-methanone (30). ***General method (A).*** A mixture of 3-hydroxy-1-(2-naphthyl)prop-2-en-1-one **1** (0.01 mole), cyanoacitic acidhydrazide **2**, arylidenecyanoacetohydrazides **7,** arylsulfonylcyanoacetohydrazides **10**, 3-aminopyrazoles **13**, 3-amino-1,2,4-triazole, 2-aminobenzoimidazole, pyrazole-3-diazonium salts **19,** 1,2,4-triazol-3-diazonium salts **22,** or benzoimidazole-2-diazonium salts **25** (0.01 mole) in piperidine acetate (1 mL), {pipridine + acetic acid + H_2O}, and H_2O (3 mL) was refluxed for 5 min. Acetic acid (1.5 mL) was added to the hot solution and the performed solid product was filtered off and recrystallized from ethanol.

Method (B) for preparation of 2-pyridones (9a–d) or (12a,b). An equimolar amount of aldehydes or arylsulfonylchlorides was added to a cold solution of compounds **4** in pyridine. The mixture was stirred for 12h and then poured over ice-water mixture and neutralized with dil. HCl. The formed solid product was filtered off to produce 2-pyridones **9** or **12**, respectively.

2-Pyridone (4). Yellow crystals from EtOH, (yield 72%), m.p. 278–280°C, ν_{max}/cm^{-1} (KBr) 3388 and 3168 (NH_2), 3043 (CH, aromatic), 2210.9 (CN), 1643 (CO); ^{1}H-NMR (300 MHz, DMSO-d6): δ = 2.49 (s, 2H, NH_2), 7.54–8.73 (m, 9H, aromatic); m/z 261 (Calcd for $C_{16}H_{11}N_3O$ (261.29):C, 73.55; H, 4.24; N, 16.08%. Found: C, 73.37; H, 4.36; N, 15.92%).

2-Pyridones (9a–d). *9a.* Pale yellow crystals from EtOH, (yield 74%), m.p. 235°C, ν_{max}/cm^{-1} (KBr) 3048 (CH, aromatic), 2206.3 (CN), 1662 (CO); ^{1}H-NMR (300 MHz, DMSO-d6): δ = 6.13–7.71 (m, 14H, aromatic), 8.10 (s, 1H, =CH); m/z 349 (Calcd for $C_{23}H_{15}N_3O$ (349.40): C, 79.07; H, 4.33; N, 12.03%. Found: C, 79.16; H, 4.14; N, 12.12%).

9b. Pale yellow crystals from EtOH, (yield 79%), m.p. 230°C, ν_{max}/cm^{-1} (KBr) 3033 (CH, aromatic), 2198.3 (CN), 1675 (CO); ^{1}H-NMR (300 MHz, DMSO-d6): δ = 2.35 (s, 3H, CH_3), 6.27–7.70 (m, 13H, aromatic), 8.07 (s, 1H, =CH); m/z 363 (Calcd for $C_{24}H_{17}N_3O$ (363.42): C, 79.32; H, 4.72; N, 11.56%. Found: C, 79.38; H, 4.64; N, 11.62%).

9c. Pale yellow crystals from EtOH, (yield 77%), m.p. 220°C, ν_{max}/cm^{-1} (KBr) 3064 (CH, aromatic), 2208.3 (CN), 1671 (CO); ^{1}H-NMR (300 MHz, DMSO-d6): δ = 3.76 (s, 3H, CH_3), 6.29–7.73 (m, 13H, aromatic), 8.09 (s, 1H, =CH); m/z 379 (Calcd for $C_{24}H_{17}N_3O_2$ (379.42): C, 75.98; H; 4.52; N, 11.07%. Found: C, 75.92; H; 4.61; N, 11.12%).

9d. Pale yellow crystals from EtOH, (yield 71%), m.p. 200°C, ν_{max}/cm^{-1} (KBr) 3051 (CH, aromatic), 2199.1 (CN), 1661 (CO); ^{1}H-NMR (300 MHz, DMSO-d6): δ = 6.31-7.72 (m, 13H, aromatic), 8.12 (s, 1H, =CH); m/z 383 (Calcd for $C_{23}H_{14}ClN_3O$ (383.84): C, 71.97; H, 3.68; Cl, 9.24; N, 10.95%. Found: C, 72.03; H, 3.64; Cl, 9.31; N, 11.02%).

2-Pyridones (12a,b). *12a.* Pale yellow crystals from EtOH, (yield 73%), m.p. 220°C, ν_{max}/cm^{-1} (KBr) 3350 (NH), 3061 (CH, aromatic), 2208.1 (CN), 1666 (CO); ^{1}H-NMR (300 MHz, DMSO-d6): δ = 2.0 (s, 1H, NH), 6.13–7.93 (m, 14H, aromatic); m/z 401 (Calcd for $C_{22}H_{15}N_3O_3S$ (401.45): C, 65.82; H, 3.77; N, 10.47; S, 7.99%. Found: C, 65.93; H, 3.65; N, 10.54; S, 8.05%).

12b. Pale yellow crystals from EtOH, (yield 75%), m.p. 290°C, ν_{max}/cm^{-1} (KBr) 3361 (NH), 3064 (CH, aromatic), 2210.1 (CN), 1664 (CO); ^{1}H NMR (300 MHz, DMSO-d6): δ = 2.1 (s, 1H, NH), 2.35 (s, 3H, CH_3), 6.15–7.94 (m, 13H, aromatic); MS: m/z = 415 (2.56, M), 345 (36.34%), 280 (53.54%), 238 (85.33%), 234 (39.34%), 121 (80.65%), 92 (90.54%), 69 (100%); (Calcd for $C_{23}H_{17}N_3O_3S$ (415.47): C, 66.49; H, 4.12; N, 10.11; S, 7.72%. Found: C, 66.52; H, 4.07; N, 10.21; S, 7.80%).

Pyrazolo[1,5-a]pyrimidines (15a–f). *15a.* Colorless crystals from EtOH, (yield 78%), m.p. 210°C, ν_{max}/cm^{-1} (KBr) 3073 (CH, aromatic); ^{1}H NMR (300 MHz, $CDCl_3$): δ = 6.53 (s, 1H), 7.22–7.89 (m, 13H, aromatic), 8.50 (d, 1H); m/z 321 (Calcd for $C_{22}H_{15}N_3$ (321.38): C, 82.22; H, 4.70; N, 13.07%. Found: C, 82.20; H, 4.77; N, 13.13%).

15b. Colorless crystals from EtOH, (yield 75%), m.p. 194°C, ν_{max}/cm^{-1} (KBr) 3082 (CH, aromatic); ^{1}H NMR (300 MHz, $CDCl_3$): δ = 2.41 (s, 3H, CH_3), 6.21 (s, 1H), 7.01–8.05 (m, 11H, aromatic), 8.52 (d, 1H), 8.78 (d, 1H); m/z 335 (Calcd for $C_{23}H_{17}N_3$ (335.41): C, 82.36; H, 5.11; N, 12.53%. Found: C, 82.27; H, 5.04; N, 12.56%).

15c. Colorless crystals from EtOH, (yield 71%), m.p. 225°C, ν_{max}/cm^{-1} (KBr) 3051 (CH, aromatic); ^{1}H-NMR (300 MHz, $CDCl_3$): δ = 6.23 (s, 1H), 7.02–8.02 (m, 11H, aromatic), 8.57 (d, 1H), 8.75 (d, 1H); m/z 355 (Calcd for $C_{22}H_{14}ClN_3$ (355.83): C, 74.26; H, 3.97; Cl, 9.96; N, 11.81%. Found: C, 74.15; H, 4.05; Cl, 9.87; N, 11.90%).

15d. Colorless crystals from EtOH, (yield 82%), m.p. 220°C, ν_{max}/cm^{-1} (KBr) 3062 (CH, aromatic); ^{1}H NMR (300 MHz, $CDCl_3$): δ = 6.53 (s, 1H), 7.22–7.89 (m, 14H, aromatic), 8.51 (d, 1H), 8.78 (d, 1H); m/z 371 (Calcd for $C_{26}H_{17}N_3$ (371.45): C, 84.07; H, 4.61; N, 11.31%. Found: C, 84.16; H, 4.52; N, 11.36%).

15e. Colorless crystals from EtOH, (yield 73%), m.p. 190°C, ν_{max}/cm^{-1} (KBr) 3073 (CH, aromatic); ^{1}H NMR (300 MHz, $CDCl_3$): δ = 7.22–7.89 (m, 12H, aromatic), 8.12 (d, 1H), 8.51 (s, 1H), 8.62 (d, 1H); m/z 321 (Calcd for $C_{22}H_{15}N_3$ (321.38): C, 82.22; H, 4.70; N, 13.07%. Found: C, 82.26; H, 4.65; N, 13.02%).

15f. Colorless crystals from EtOH, (yield 75%), m.p. 225°C, ν_{max}/cm^{-1} (KBr) 3053(CH, aromatic), 1223 (CN); ^{1}H NMR (300 MHz, $CDCl_3$): δ = 7.22–7.89 (m, 7H, aromatic), 7.60 (d, 1H), 8.15 (s, 1H), 8.51 (d, 1H); m/z 270 (Calcd for $C_{17}H_{10}N_4$ (270.30): C, 75.54; H, 3.73; N, 20.73%. Found: C, 75.51; H, 3.82; N, 20.80%).

Triazolo[1,5-a]pyrimidine (19). Colorless crystals from EtOH, (yield 72%), m.p. 210°C, ν_{max}/cm^{-1} (KBr) 3053(CH, aromatic); ^{1}H NMR (300 MHz, $CDCl_3$): δ = 7.32–7.89 (m, 7H, aromatic), 7.52 (d, 1H), 8.27 (s, 1H), 8.50 (d, 1H); m/z = 246 (Calcd for $C_{15}H_{10}N_4$ (246.27): C, 73.16; H, 4.09; N, 22.75%. Found: C, 73.19; H, 4.13; N, 22.64%).

Benzo[4,5]imidazo[1,2-a]pyrimidine (21). Colorless crystals from EtOH, (yield 78%), m.p. 230°C, ν_{max}/cm^{-1} (KBr) 3053(CH, aromatic); ^{1}H NMR (300 MHz, $CDCl_3$): δ = 7.31–7.89 (m, 11H, aromatic), 7.52 (d, 1H), 8.50 (d, 1H); m/z 295 (Calcd for $C_{20}H_{13}N_3$ (295.35): C, 81.34; H, 4.44; N, 14.23%. Found: C, 81.39; H, 4.38; N, 14.27%).

(Pyrazolo[5,1-c][1,2,4]triazin-3-yl)-methanones (24a–f). *24a.* Colorless crystals from AcOH, (yield 75%), m.p. 210°C, ν_{max}/cm^{-1} (KBr) 3073 (CH, aromatic), 1665 (CO); ^{1}H NMR (300 MHz, DMSO-d6): δ = 6.53 (s, 1H), 7.22–8.22 (m, 12H, aromatic), 9.56 (s, 1H); m/z 350 (Calcd for $C_{22}H_{14}N_4O$ (350.38): C, 75.42; H, 4.03; N, 15.99%. Found: C, 75.45; H, 4.12; N, 16.04%).

24b. Colorless crystals from AcOH, (yield 73%), m.p. 194°C, ν_{max}/cm^{-1} (KBr) 3082 (CH, aromatic), 1665 (CO); ^{1}H NMR (300 MHz, DMSO-d6): δ = 2.35 (s, 3H, CH_3), 6.21 (s, 1H), 7.12–8.22 (m, 11H, aromatic),

9.46 (s, 1H); m/z 364 (Calcd for $C_{23}H_{16}N_4O$ (364.41): C, 75.81; H, 4.43; N, 15.37%. Found: C, 75.87; H, 4.52; N, 15.41%).

24c. Colorless crystals from AcOH, (yield 71%), m.p. 225°C, ν_{max}/cm^{-1} (KBr) 3051 (CH, aromatic), 1663 (CO); ^1H NMR (300 MHz, DMSO-d6): δ = 6.53 (s, 1H), 7.22–7.86 (m, 11H, aromatic), 9.42 (s, 1H); m/z 384 (Calcd for $C_{22}H_{13}ClN_4O$ (384.83): C, 68.67; H, 3.41; Cl, 9.21; N, 14.56%. Found: C, 68.53; H, 3.36; Cl, 9.30; N, 14.62%).

24d. Colorless crystals from AcOH, (yield 79%), m.p. 220°C, ν_{max}/cm^{-1} (KBr) 3062 (CH, aromatic), 1654(CO); ^1H NMR (300 MHz, DMSO-d6): δ = 6.53 (s, 1H), 7.32–8.22 (m, 14H, aromatic), 9.46 (s, 1H); m/z 400 (Calcd for $C_{26}H_{16}N_4O$ (400.44): C, 77.99; H, 4.03; N, 13.99%. Found: C, 78.05; H, 4.11; N, 14.04%).

24e. Colorless crystals from AcOH, (yield 77%), m.p. 226–230°C, ν_{max}/cm^{-1} (KBr) 3073 (CH, aromatic), 1646 (CO); ^1H NMR (300 MHz, DMSO-d6): δ = 7.22–7.89 (m, 12H, aromatic), 8.40 (s, 1H), 9.46 (s, 1H); MS: m/z = 350 (34.57%, M^+ 1), 350 (100%, M), 321 (9.27%), 154 (85.33%), 126 (58.26%), 100 (3.93%), 69 (4.95%), 55 (8.15%) (Calcd for $C_{22}H_{14}N_4O$ (350.38): C, 75.42; H, 4.03; N, 15.99%. Found: C, 75.37; H, 4.06; N, 15.92%).

24f. Colorless crystals from AcOH, (yield 77%), m.p. 225°C, ν_{max}/cm^{-1} (KBr) 3064(CH, aromatic), 1653 (CO), 1221 (CN); ^1H NMR (300 MHz, DMSO-d6): δ = 7.32–8.22 (m, 7H, aromatic), 7.63 (s, 1H), 9.36 (s, 1H); m/z 299 (Calcd for $C_{17}H_9N_5O$ (299.29): C, 68.22; H, 3.03; N, 23.40%. Found: C, 68.15; H, 3.23; N, 23.64%).

[1,2,4]Triazolo[5,1-c][1,2,4]triazin-3-yl-methanone (27) Color-less crystals from AcOH, (yield 74%), m.p. 260°C, ν_{max}/cm^{-1} (KBr) 3053(CH, aromatic), 1665 (CO); ^1H NMR (300 MHz, DMSO-d6): δ = 7.32–8.21 (m, 7H, aromatic), 8.27 (s, 1H), 9.46 (s, 1H); m/z = 275 (Calcd for $C_{15}H_9N_5O$ (275.27): C, 65.45; H, 3.30; N, 25.44%. Found: C, 65.36; H, 3.22; N, 25.51%).

Benzo[4,5] imidazo[2,1-c][1,2,4]triazin-3-yl-methanone (30) Colorless crystals from AcOH, (yield 76%), m.p. 230°C, ν_{max}/cm^{-1} (KBr) 3053(CH, aromatic), 1665 (CO); ^1H NMR (300 MHz, $CDCl_3$): δ = 7.32–8.22 (m, 11H, aromatic), 9.45 (s, 1H); m/z 324 (Calcd for $C_{20}H_{12}N_4O$ (324.34): C, 74.06; H, 3.73; N, 17.27%. Found: C, 74.13; H, 3.67; N, 17.32%).

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