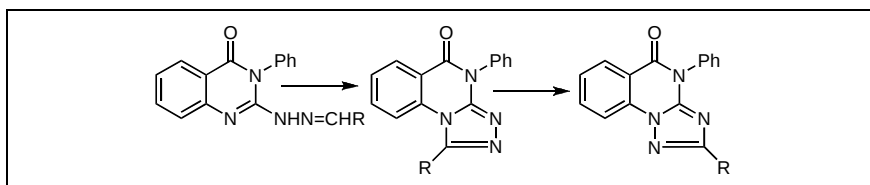


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A novel series of 2,4-disubstituted-1,2,4-triazolo[1,5-*a*]quinazolin-5(4*H*)-ones were prepared by Dimroth rearrangement of their respective isomers namely 1,4-disubstituted-[1,2,4]triazolo[4,3-*a*]quinazolin-5(4*H*)-ones. The latter were prepared *via* new synthetic strategy based on 1,5-electrocyclization of the respective *N*-(4-oxo-3-phenylquinazolin-2-yl)nitrilimines.

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INTRODUCTION

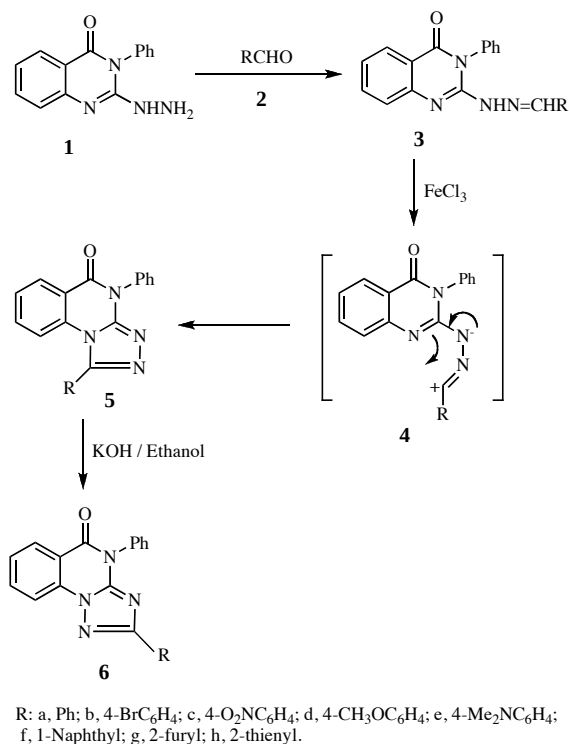
Recently, it was reported that some 1,4-disubstituted-[1,2,4]triazolo[4,3-*a*]quinazolin-5(4*H*)-ones exhibit promising antihistaminic activity against histamine-induced bronchospasm on conscious guinea pigs *in vivo* model [1]. Some other derivatives showed negligible sedation compared to chlorpheniramine maleate and could therefore serve as lead molecules for further modification to obtain a clinically useful class of non-sedative antihistamines [1] [2]. In addition, 1-(*p*-chlorophenyl-amino)-4-methyl-1,2,4-triazolo[4,3-*a*]quinazolin-5-one was reported to exhibit antitoxoplasmosis activity [3]. These findings aroused our interest in exploring the biological activity of the title compounds which are the isomers of 1,4-disubstituted-[1,2,4]triazolo[4,3-*a*]quinazolin-5(4*H*)-ones. However, prior to this exploration, it was thought necessary to develop a new synthetic strategy for synthesis of 1,4-disubstituted-[1,2,4]triazolo[4,3-*a*]quinazolin-5(4*H*)-ones and their isomeric 2,4-disubstituted-1,2,4-triazolo[1,5-*a*]quinazolin-5(4*H*)-ones. This is because the synthetic studies of 1,2,4-triazolo[1,5-*a*]quinazoline ring systems have been rather limited and the reaction of 2-hydrazinobenzoic acid with *N*-cyanoimides seems to offer so far the best route to such ring system and its derivatives [4].

In continuation of our ongoing studies dealing with the chemistry of nitrilimines and their precursors [5-9], we wish to report herein the results of our study of 1,5-electrocyclization of *N*-(4-oxo-3-phenylquinazolin-2-yl)-nitrilimines to give 1,2,4-triazolo[4,3-*a*]quinazolin-5(4*H*)-ones and Dimroth rearrangement of the latter to give the respective 1,4-disubstituted-1,2,4-triazolo[1,5-*a*]quinazolin-5(4*H*)-ones.

RESULTS AND DISCUSSION

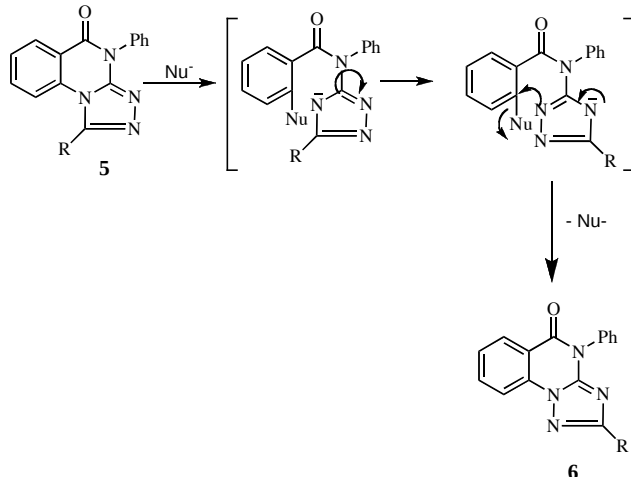
The required aldehyde *N*-(3-phenyl-4-oxoquinazolin-2-yl)hydrazones **3** were prepared by condensation of 2-hydrazino-3-phenylquinazolin-4(3*H*)-one (**1**) with the appropriate aldehydes **2** (Scheme 1). Three of the latter hydrazones namely **3a**, **3c** and **3d** are known [10] whereas the other hydrazones have not been reported hitherto. The structures of the new derivatives were confirmed by their elemental analyses and spectral (MS, IR and ¹H NMR) data (see Experimental). For example, their ¹H NMR in DMSO-*d*₆ revealed, in each case, two characteristic signals in the regions δ 8.0 - 8.2 and 10.3 - 10.7 corresponding to the -N=CH- and hydrazone -NH-N=C protons, respectively. Treatment of each of the hydrazones **3** with equivalent amount of iron(III) chloride in ethanol for 30 min gave a single product as evidenced by TLC analysis which was isolated as a crystalline solid. Elemental analyses and mass spectra revealed that each of the isolated products has two less hydrogen atoms than the respective hydrazone. This feature was confirmed by ¹H NMR spectroscopy which indicated the absence of the signals attributed to the -N=CH- and hydrazone -NH-N=C functions. In the IR spectra the compounds showed a peak for carbonyl (C=O) around 1680 cm⁻¹. On the basis of these findings, the isolated products were assigned the structure of 1,4-disubstituted-1,2,4-triazolo[4,3-*a*]quinazolin-5(4*H*)-one **5** (Scheme 1). The conversion of **3** into **5** is reminiscent of other related oxidative cyclization of aldehyde *N*-heteroarylhydrazones with iron(III) chloride, which was reported to proceed *via* generation of the respective nitrilimines **4** which undergo *in situ* 1,5-electrocyclization to give the respective fused heterocycles [11,12] (Scheme 1).

Scheme 1



Treatment of **5** with potassium hydroxide in refluxing ethanol yielded, in each case, one product that was identified as the respective 2,4-disubstituted-1,2,4-triazolo[1,5-*a*]quinazolin-5(4H)-one **6** (Scheme 1). This isomerization is similar to the Dimroth rearrangement of 1,2,4-triazolo[4,3-*a*]pyrimidine into 1,2,4-triazolo[1,5-*a*]pyrimidine [15]. To account for the rearrangement of **5** into **6**, it is suggested that it occurs through ring opening and ring closure as depicted in Scheme 2.

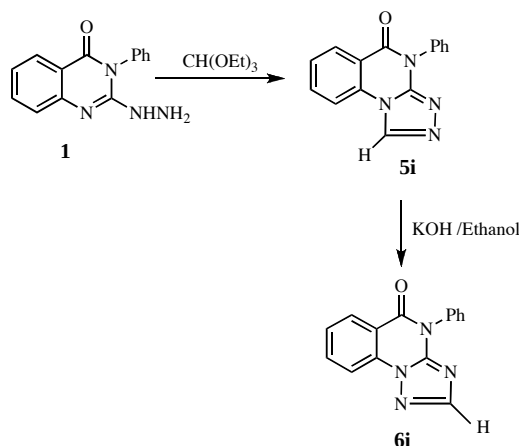
Scheme 2



The structure of the products **6** was verified by elemental analyses and spectral (MS, IR, ¹H NMR) data (see Experimental) and alternate synthesis of the two isomers **5i** and **6i**. Thus, reaction of **1** with triethyl orthoformate gave **5i** [10]. Heating the latter in ethanol in the presence of potassium hydroxide gave **6i** (Scheme 3).

¹H NMR spectra of **5i** and **6i** revealed their triazole proton signals at δ 9.5 and 8.3, respectively. This finding indicates that the triazole (C3-H) of **5i** is more deshielded than that of the triazole (C2-H) of **6i**. Such feature is consistent with literature reports which indicate that the triazole proton of triazolo[4,3-*c*]pyrimidine (C3-H) is more deshielded than that of [1,5-*c*] isomers (C2-H) [16] [17] and provides thus a conclusive evidence of the rearrangement of **5** into **6** under the reaction conditions employed.

(Scheme 3)



EXPERIMENTAL

All melting points were determined on an electrothermal Gallenkamp apparatus and are uncorrected. The IR spectra were measured on a Pye-Unicam SP300 instrument in potassium bromide discs. The ¹H-NMR spectra were recorded on a Varian Mercury VXR-300 spectrometer (300 MHz) in DMSO-*d*₆ or CDCl₃. The mass spectra were recorded on a GCMS-Q1000-EX Shimadzu and GCMS 5988-A HP spectrometers, the ionizing voltage was 70 eV. Elemental analyses were carried out by the Microanalytical Center of Cairo University, Giza, Egypt. The starting 2-hydrazino-3-phenylquinazolin-4(3H)-one (**1**) was prepared by hydrazinolysis of 3-phenyl-2-thioxo-quinazoline-4(1H,3H)-one according to literature method [14].

Aldehyde N-(3-phenyl-4-oxoquinazolin-2-yl)hydrazones (3). General Procedure. To a mixture of 2-hydrazino-3-phenylquinazolin-4(3H)-one **3** (1.3 g, 5 mmole) and the appropriate aldehyde **2** (5 mmole) in ethanol (50 mL), few drops of acetic acid were added and the reaction mixture was refluxed for 3 h then cooled. The precipitate, formed upon cooling, was collected by filtration, washed with water then ethanol and finally crystallized from the appropriate solvent to give the corresponding hydrazone derivative **3**. The various hydrazone

derivatives **3a-h** prepared are listed in the following together with their physical constants and spectral data.

Benzaldehyde *N*-(3-phenyl-4-oxoquinazolin-2-yl)-hydrazine (3a). Colorless crystals (yield 86%), m.p. 226–228°C (ethanol), Lit. m.p. 232°C [10], 205–206°C [18], IR: (KBr) ν 3426, 3344, 1697, 1612 cm^{-1} . ^1H NMR (DMSO): δ 7.10 – 8.0 (m, 14 H, ArH), 8.10 (s, 1H, $-\text{N}=\text{CH}$), 10.5 (s, 1H, NH). MS: m/z (%): 341 ($\text{M}^+ + 1$, 13), 340 (M^+ , 49), 339 (60), 263 (93), 235 (55), 192 (10), 118 (27), 91 (37), 89 (100), 77 (93), 62 (36). Anal Calcd. for $\text{C}_{21}\text{H}_{16}\text{N}_4\text{O}$ (340.39): C, 74.10; H, 4.74; N, 16.46. Found: C, 73.94; H, 4.77; N, 16.53 %.

4-Bromobenzaldehyde *N*-(3-phenyl-4-oxoquinazolin-2-yl)-hydrazine (3b). Pale yellow crystals (yield 85%), m.p. 285°C (ethanol-dioxane). IR: (KBr) ν 3215, 1667, 1618 cm^{-1} . ^1H NMR (DMSO): δ 7.31 – 7.80 (m, 9 H, ArH), 7.35 (d, 2H, ArH), 7.90 (d, 2H, ArH), 8.00 (s, 1H, $-\text{N}=\text{CH}$), 10.60 (s, 1H, NH). MS: m/z (%): 421 ($\text{M}^+ + 2$, 11), 420 ($\text{M}^+ + 1$, 42), 419 (M^+ , 18), 263 (100), 236 (58), 193 (15), 119 (27), 90 (27), 77 (44). Anal Calcd. for $\text{C}_{21}\text{H}_{15}\text{BrN}_4\text{O}$ (419.28): C, 60.16; H, 3.61; N, 13.36. Found: C, 60.15; H, 3.65; N, 13.38 %.

4-Nitrobenzaldehyde *N*-(3-phenyl-4-oxoquinazolin-2-yl)-hydrazine (3c). Orange crystals (yield 86%), m.p. 269°C (ethanol-dioxane), Lit. m.p. 260–262°C [18], IR: (KBr) ν 3350, 1692, 1618 cm^{-1} . ^1H NMR (DMSO): δ 7.20 – 7.85 (m, 9 H, ArH), 7.90 (d, 2H, ArH), 8.15 (d, 2H, ArH), 8.25 (s, 1H, $-\text{N}=\text{CH}$), 10.73 (s, 1H, NH). MS: m/z (%): 386 ($\text{M}^+ + 1$, 14), 385 (M^+ , 48), 262 (100), 235 (51), 192 (12), 119 (44), 91 (37), 77 (74). Anal Calcd. for $\text{C}_{21}\text{H}_{15}\text{N}_5\text{O}_3$ (385.39): C, 65.45; H, 3.92; N, 18.17. Found: C, 65.35; H, 4.21; N, 18.03 %.

4-Methoxybenzaldehyde *N*-(3-phenyl-4-oxoquinazolin-2-yl)-hydrazine (3d). Colorless crystals (yield 90%), m.p. 236°C (ethanol), Lit. m.p. 238°C [10], 274–276°C [18], IR: (KBr) ν 3235, 1696, 1618 cm^{-1} . ^1H NMR (DMSO): δ 3.88 (s, 3H, OCH_3), 7.10 (d, 2H, ArH), 7.50 – 7.90 (m, 9 H, ArH), 8.00 (s, 1H, $-\text{N}=\text{CH}$), 8.26 (d, 2H, ArH), 10.49 (s, 1H, NH). MS: m/z (%): 371 ($\text{M}^+ + 1$, 34), 370 (M^+ , 100), 263 (57), 236 (66), 120 (29), 91 (27), 77 (48). Anal Calcd. for $\text{C}_{22}\text{H}_{18}\text{N}_4\text{O}_2$ (370.40): C, 71.34; H, 4.90; N, 15.13. Found: C, 71.64; H, 5.18; N, 15.30 %.

4-Dimethylaminobenzaldehyde *N*-(3-phenyl-4-oxoquinazolin-2-yl)-hydrazine (3e). Orange crystals (yield 81%), m.p. 260°C (ethanol-dioxane). IR: (KBr) ν 3328, 1689, 1606 cm^{-1} . ^1H NMR (DMSO): δ 3.00 (s, 6H, $\text{N}(\text{CH}_3)_2$), 6.68 – 7.74 (m, 13 H, ArH), 7.86 (s, 1H, $-\text{N}=\text{CH}$), 10.40 (s, 1H, NH). MS: m/z (%): 384 ($\text{M}^+ + 1$, 73), 383 (M^+ , 100), 382 (21), 263 (25), 236 (58), 223 (14), 146 (63), 132 (49), 91 (21), 77 (34). Anal Calcd. for $\text{C}_{23}\text{H}_{21}\text{N}_5\text{O}$ (383.46): C, 72.04; H, 5.52; N, 18.26. Found: C, 72.37; H, 5.23; N, 18.48 %.

1-Naphthaldehyde *N*-(3-phenyl-4-oxoquinazolin-2-yl)-hydrazine (3f). pale green crystals (yield 82%), m.p. 230°C (ethanol-dioxane). IR: (KBr) ν 3342, 1684, 1613 cm^{-1} . ^1H NMR (DMSO): δ 7.13 – 8.67 (m, 16 H, ArH), 8.77 (s, 1H, $-\text{N}=\text{CH}$), 10.67 (s, 1H, NH). MS: m/z (%): 391 ($\text{M}^+ + 1$, 28), 390 (M^+ , 72), 263 (100), 236 (75), 140 (46), 127 (15), 119 (12), 91 (12), 77 (32). Anal Calcd. for $\text{C}_{25}\text{H}_{18}\text{N}_4\text{O}$ (390.45): C, 76.91; H, 4.65; N, 14.35. Found: C, 77.01; H, 4.36; N, 14.32 %.

2-Furfuraldehyde *N*-(3-phenyl-4-oxoquinazolin-2-yl)-hydrazine (3g). colorless crystals (yield 66%), m.p. 214°C (ethanol). IR: (KBr) ν 3210, 1681, 1629 cm^{-1} . ^1H NMR (DMSO): δ 6.6 (dd, 1H, $J = 4$ Hz, Furyl-C4-H), 6.8 (d, 1H, $J = 4$ Hz, Furyl-C3-H), 7.7 (d, 1H, $J = 4$ Hz, Furyl-C5-H), 7.55 (–8.50 (m, 9H, Ar-H), 7.90 (s, 1H, $-\text{N}=\text{CH}$), 10.40 (s, 1H, NH). MS: m/z (%): 331 ($\text{M}^+ + 1$, 12), 330 (M^+ , 59), 329 (72),

235 (53), 118 (26), 92 (30), 77 (64), 51 (100). Anal Calcd. for $\text{C}_{19}\text{H}_{14}\text{N}_4\text{O}_2$ (330.35): C, 69.08; H, 4.27; N, 16.96. Found: C, 69.15; H, 4.21; N, 16.91 %.

2-Thiophenealdehyde *N*-(3-phenyl-4-oxoquinazolin-2-yl)-hydrazine (3h). pale brown crystals (yield 82%), m.p. 252°C (ethanol-dioxane). IR: (KBr) ν 3349, 1698, 1614 cm^{-1} . ^1H NMR (DMSO): δ 6.9 (d, 1H, $J = 4$ Hz, thienyl-C3-H), 7.3 (dd, 1H, $J = 4$ Hz, thienyl-C4-H), 7.6 (d, 1H, $J = 4$ Hz, thienyl-C5-H), 7.6 – 8.5 (m, 9 H, ArH), 8.21 (s, 1H, $-\text{N}=\text{CH}$), 10.29 (s, 1H, NH). MS: m/z (%): 347 ($\text{M}^+ + 1$, 19), 346 (M^+ , 82), 235 (53), 193 (14), 166 (12), 144 (16), 119 (40), 109 (20), 95 (94), 77 (100). Anal Calcd. for $\text{C}_{19}\text{H}_{14}\text{N}_4\text{OS}$ (346.41): C, 65.88; H, 4.07; N, 16.17. Found: C, 65.77; H, 4.27; N, 15.96 %.

1,4-Disubstituted-1,2,4-triazolo[4,3-*a*]quinazolin-5(4*H*)-ones (5). **General procedure.** To a solution of the appropriate aldehyde hydrazone **3** (2.5 mmole) in ethanol (150 mL), was added a solution of ferric chloride (2 *M*, 2 mL) and the mixture was refluxed for 30 min then left overnight at room temperature. The solution was evaporated under reduced pressure and water was added to it. The solid that precipitated was collected by filtration, washed with water, dried and finally crystallized from ethanol to give the respective 1,4-substituted-1,2,4-triazolo[4,3-*a*]quinazolin-5(4*H*)-one **5**. The compounds prepared together with their physical constants and spectral data are listed below.

1,4-Diphenyl-1,2,4-triazolo[4,3-*a*]quinazolin-5(4*H*)-one (5a). Colorless crystals (yield 60%), m.p. 267°C (ethanol), (Lit. m.p. 238°C [10]), IR: (KBr) ν 1691, 1607 cm^{-1} . ^1H NMR (DMSO): δ 7.11 (d, 2H, ArH), 7.52 – 7.73 (m, 10H, ArH), 8.3 (d, 2H, ArH). MS: m/z (%): 339 ($\text{M}^+ + 1$, 16), 338 (M^+ , 67), 234 (66), 205 (10), 178 (55), 103 (44), 101 (15), 77 (72), 76 (100). Anal Calcd. for $\text{C}_{21}\text{H}_{14}\text{N}_4\text{O}$ (338.37): C, 74.54; H, 4.17; N, 16.56. Found: C, 74.64; H, 4.32; N, 16.57 %.

1-(4-Bromophenyl)-4-phenyl-1,2,4-triazolo[4,3-*a*]quinazolin-5(4*H*)-one (5b). Pale yellow crystals (yield 64%), m.p. 285°C (ethanol). IR: (KBr) ν 1680, 1611 cm^{-1} . ^1H NMR (CDCl_3): δ 7.27 – 8.94 (m, ArH). MS: m/z (%): 419 ($\text{M}^+ + 2$, 6), 418 ($\text{M}^+ + 1$, 27), 417 (M^+ , 33), 258 (12), 234 (56), 177 (19), 103 (19), 77 (58), 76 (100). Anal Calcd. for $\text{C}_{21}\text{H}_{13}\text{BrN}_4\text{O}$ (417.27): C, 60.45; H, 3.14; N, 13.43. Found: C, 60.58; H, 3.03; N, 13.48 %.

1-(4-Nitrophenyl)-4-phenyl-1,2,4-triazolo[4,3-*a*]quinazolin-5(4*H*)-one (5c). Orange crystals (yield 57%), m.p. 310°C (ethanol). IR: (KBr) ν 1683, 1608 cm^{-1} . ^1H NMR (DMSO): δ 7.15 – 8.04 (m, ArH). MS: m/z (%): 384 ($\text{M}^+ + 1$, 30), 383 (M^+ , 30), 234 (49), 224 (19), 177 (21), 103 (20), 77 (54), 76 (100). Anal Calcd. for $\text{C}_{21}\text{H}_{13}\text{N}_5\text{O}_3$ (383.37): C, 65.79; H, 3.42; N, 18.27. Found: C, 65.86; H, 3.34; N, 18.54 %.

1-(4-Methoxyphenyl)-4-phenyl-1,2,4-triazolo[4,3-*a*]quinazolin-5(4*H*)-one (5d). Colorless crystals (yield 68%), m.p. 290°C (ethanol), (Lit. m.p. 282°C [10]). IR (KBr) ν 1681, 1612 cm^{-1} . ^1H NMR (CDCl_3): δ 3.94 (s, 3H, CH_3O), 7.09 – 8.66 (m, 13H, ArH). MS: m/z (%): 369 ($\text{M}^+ + 1$, 9), 368 (M^+ , 34), 367 (43), 234 (44), 165 (10), 132 (36), 102 (43), 90 (41), 77 (58), 75 (100). Anal Calcd. for $\text{C}_{22}\text{H}_{16}\text{N}_4\text{O}_2$ (368.40): C, 71.73; H, 4.38; N, 15.21. Found: C, 71.90; H, 4.43; N, 15.31 %.

1-(4-Dimethylaminophenyl)-4-phenyl-1,2,4-triazolo[4,3-*a*]quinazolin-5(4*H*)-one (5e). Orange crystals (yield 66%), m.p. 280°C (ethanol). IR: (KBr) ν 1685, 1614 cm^{-1} . ^1H NMR (CDCl_3): δ 3.09 (s, 6H, $(\text{CH}_3)_2\text{N}$), 6.83 – 8.42 (m, 13H, ArH). MS: m/z (%): 382 ($\text{M}^+ + 1$, 17), 381 (M^+ , 64), 380 (100), 234 (18), 145 (50), 144 (50), 103 (20), 77 (28), 76 (50), 75 (65). Anal Calcd. for $\text{C}_{23}\text{H}_{19}\text{N}_5\text{O}$ (381.44): C, 72.42; H, 5.02; N, 18.36. Found: C, 72.48; H, 5.13; N, 18.51 %.

1-(1-Naphthyl)-4-phenyl-1,2,4-triazolo[4,3-a]quinazolin-5(4H)-one (5f). Pale green crystals (yield 65%), m.p. 255°C (ethanol). IR: (KBr) ν 1689, 1610 cm^{-1} . ^1H NMR (CDCl_3): δ 6.65 – 8.03 (m, ArH). MS: m/z (%): 389 ($\text{M}^+ + 1$, 12), 388 (M^+ , 52), 387 (100), 235 (10), 227 (14), 152 (30), 125 (15), 77 (43), 76 (57). *Anal Calcd.* for $\text{C}_{25}\text{H}_{16}\text{N}_4\text{O}$ (388.43): C, 77.31; H, 4.15; N, 14.42. Found: C, 76.79; H, 4.27; N, 14.14 %.

1-(2-Furyl)-4-phenyl-1,2,4-triazolo[4,3-a]quinazolin-5(4H)-one (5g). Colorless crystals (yield 86%), m.p. 250°C (ethanol). IR: (KBr) ν 1684, 1601 cm^{-1} . ^1H NMR (CDCl_3): δ 6.7 (dd, 1H, J = 4 Hz, Furyl-C4-H), 7.4 (d, 1H, J = 4 Hz, Furyl-C3-H), 7.9 (d, 1H, J = 4 Hz, Furyl-C5-H), 7.7 – 8.2 (m, 9H, ArH). MS: m/z (%): 345 ($\text{M}^+ + 1$, 0.3), 344 (M^+ , 10), 327 (100), 234 (39), 168 (47), 104 (11), 92 (31), 77 (51), 75 (90). *Anal Calcd.* for $\text{C}_{19}\text{H}_{12}\text{N}_4\text{O}_2$ (328.33): C, 69.51; H, 3.68; N, 17.06. Found: C, 69.48; H, 3.34; N, 17.18 %.

1-(2-Thienyl)-4-phenyl-1,2,4-triazolo[4,3-a]quinazolin-5(4H)-one (5h). Pale brown crystals (yield 60%), m.p. 242°C (ethanol). IR: (KBr) ν 1684, 1601 cm^{-1} . ^1H NMR (CDCl_3): δ 7.4 (dd, 1H, J = 4 Hz, thienyl-C4-H), 7.8 (d, 1H, J = 4 Hz, thienyl-C3-H), 8.10 (d, 1H, J = 4 Hz, thienyl-C5-H), 7.3 – 8.2 (m, 9H, ArH). MS: m/z (%): 345 ($\text{M}^+ + 1$, 21), 344 (M^+ , 77), 343 (100), 234 (72), 184 (62), 168 (47), 109 (33), 77 (57), 75 (90). *Anal Calcd.* for $\text{C}_{19}\text{H}_{12}\text{N}_4\text{OS}$ (344.40): C, 66.26; H, 3.51; N, 16.27. Found: C, 66.20; H, 3.33; N, 16.05 %.

4-Phenyl-1,2,4-triazolo[4,3-a]quinazolin-5(4H)-one (5i). A mixture of 2-hydrazino-3-phenylquinazolin-4(3H)-one **1** (1.3 g, 5 mmole) and triethyl orthoformate (20 mL) was refluxed for 9 h then cooled. The precipitate that formed upon cooling was collected by filtration, washed with water then ethanol and finally crystallized from ethanol to give **5i** brown solid, 0.85 g, 65% yield, m.p. 336 °C (Lit. m.p. 332-334°C [10]. IR: (KBr) ν 1685, 1601 cm^{-1} . ^1H NMR (CDCl_3): δ 7.5-8.3 (m, 9H, ArH), 9.5 (s, 1H, triazole). MS: m/z (%): 263 ($\text{M}^+ + 1$, 74), 262 (M^+ , 100), 235 (78), 103 (44), 90 (15), 77 (50). *Anal Calcd.* for $\text{C}_{15}\text{H}_{10}\text{N}_4\text{O}$ (262.27): C, 68.69; H, 3.84; N, 21.36. Found: C, 68.38; H, 3.99; N, 21.53 %.

2,4-Disubstituted-1,2,4-triazolo[1,5-a]quinazolin-5(4H)-ones (6). **General procedure.** To a solution of **5** (1 mmole) in absolute ethanol (15 mL) was added potassium hydroxide (0.25 g) and the mixture was refluxed for 4 h and cooled. The solid, that precipitated upon neutralization of the mixture with hydrochloric acid (6 M) was collected by filtration, washed with water, dried and finally crystallized from appropriate solvent. The various products that were isolated are listed below together with their physical constants and spectral data.

2,4-Diphenyl-1,2,4-triazolo[1,5-a]quinazolin-5(4H)-one (6a). Greenish crystals (yield 82%), m.p. 218°C. IR: (KBr) ν 1695, 1600 cm^{-1} . ^1H NMR (DMSO): δ 6.84 – 8.02 (m, ArH). MS: m/z (%): 339 ($\text{M}^+ + 1$, 56), 338 (M^+ , 100), 235 (45), 179 (34), 104 (12), 103 (20), 90 (11), 77 (36). *Anal Calcd.* for $\text{C}_{21}\text{H}_{14}\text{N}_4\text{O}$ (338.37): C, 74.54; H, 4.17; N, 16.56. Found: C, 74.62; H, 4.10; N, 16.46 %.

2-(4-Bromophenyl)-4-phenyl-1,2,4-triazolo[1,5-a]quinazolin-5(4H)-one (6b). Colorless crystals (yield 82%), m.p. 240°C (ethanol-DMF). IR: (KBr) ν 1682, 163 cm^{-1} . ^1H NMR (DMSO): δ 6.83 – 8.18 (m, ArH). MS: m/z (%): 419 ($\text{M}^+ + 2$, 19), 418 ($\text{M}^+ + 1$, 100), 417 (M^+ , 55), 416 (89), 336 (15), 259 (23), 235 (88), 181 (11), 178 (23), 104 (10), 103 (17), 102 (29), 90 (18), 77 (41), 76 (51). *Anal Calcd.* for $\text{C}_{21}\text{H}_{13}\text{BrN}_4\text{O}$ (417.27): C, 60.54; H, 3.14; N, 13.43. Found: C, 60.40; H, 3.03; N, 13.14 %.

2-(4-Nitrophenyl)-4-phenyl-1,2,4-triazolo[1,5-a]quinazolin-5(4H)-one (6c). Brown crystals (yield 82%), m.p. 243°C

(ethanol-DMF). IR (KBr) ν 1702, 1601 cm^{-1} . ^1H NMR (DMSO): δ 6.86 – 8.30 (m, ArH). MS: m/z (%): 384 ($\text{M}^+ + 1$, 9), 383 (M^+ , 12), 369 (9), 353 (9), 288 (12), 265 (15), 234 (9), 215 (14), 175 (9), 145 (8), 118 (20), 105 (30), 104 (25), 93 (19), 79 (16), 76 (29), 55 (100). *Anal Calcd.* for $\text{C}_{21}\text{H}_{13}\text{N}_5\text{O}_3$ (383.37): C, 65.79; H, 3.42; N, 18.27. Found: C, 65.75; H, 3.21; N, 18.60 %.

2-(4-Methoxyphenyl)-4-phenyl-1,2,4-triazolo[1,5-a]quinazolin-5(4H)-one (6d). Colorless crystals (yield 85%), m.p. 240°C (ethanol-DMF). IR (KBr) ν 1680, 1604 cm^{-1} . ^1H NMR (DMSO): δ 3.71 (s, 3H, CH_3O), 6.83 – 8.06 (m, 13H, ArH). MS: m/z (%): 369 ($\text{M}^+ + 1$, 24), 368 (M^+ , 100), 235 (38), 234 (12), 209 (23), 133 (26), 103 (22), 90 (24), 76 (42). *Anal Calcd.* for $\text{C}_{22}\text{H}_{16}\text{N}_4\text{O}_2$ (368.40): C, 71.73; H, 4.38; N, 15.21. Found: C, 71.64; H, 4.40; N, 15.00 %.

2-(4-Dimethylaminophenyl)-4-phenyl-1,2,4-triazolo[1,5-a]quinazolin-5(4H)-one (6e). White crystals (yield 86%), m.p. 212°C (ethanol-DMF). IR: (KBr) ν 1607, 1587 cm^{-1} . ^1H NMR (DMSO): δ 2.86 (s, 6H, $(\text{CH}_3)_2\text{N}$), 6.54 – 8.05 (m, 13H, ArH). MS: m/z (%): 382 ($\text{M}^+ + 1$, 83), 381 (M^+ , 100), 235 (25), 145 (27), 77 (20), 76 (17). *Anal Calcd.* for $\text{C}_{23}\text{H}_{19}\text{N}_5\text{O}$ (381.44): C, 72.42; H, 5.02; N, 18.36. Found: C, 72.68; H, 5.07; N, 18.20 %.

2-(1-Naphthyl)-4-phenyl-1,2,4-triazolo[1,5-a]quinazolin-5(4H)-one (6f). White crystals (yield 81%), m.p. 198°C (ethanol-water). IR: (KBr) ν 1667, 1600 cm^{-1} . ^1H NMR (DMSO): δ 6.24 – 8.22 (m, ArH). MS: m/z (%): 389 ($\text{M}^+ + 1$, 82), 388 (M^+ , 82), 387 (100), 235 (14), 228 (14), 153 (38), 103 (15), 77 (26), 76 (33). *Anal Calcd.* for $\text{C}_{25}\text{H}_{16}\text{N}_4\text{O}$ (388.43): C, 77.31; H, 4.15; N, 14.42. Found: C, 77.63; H, 4.10; N, 14.22 %.

2-(2-Furyl)-4-phenyl-1,2,4-triazolo[1,5-a]quinazolin-5(4H)-one (6g). White crystals (yield 78%), m.p. 244°C (ethanol). IR: (KBr) ν 1690, 1603 cm^{-1} . ^1H NMR (DMSO): δ 6.5 (dd, 1H, J = 4 Hz, Furyl-C4-H), 7.4 (d, 1H, J = 4 Hz, Furyl-C3-H), 7.7 (d, 1H, J = 4 Hz, Furyl-C5-H), 7.5 – 8.2 (m, 9H, ArH). MS: m/z (%): 330 ($\text{M}^+ + 2$, 15), 329 ($\text{M}^+ + 1$, 39), 328 (M^+ , 100), 325 (36), 197 (17), 169 (21), 90 (10), 77 (20), 76 (32). *Anal Calcd.* for $\text{C}_{19}\text{H}_{12}\text{N}_4\text{O}_2$ (328.33): C, 69.51; H, 3.68; N, 17.06. Found: C, 69.60; H, 3.51; N, 17.16 %.

2-(2-Thienyl)-4-phenyl-1,2,4-triazolo[1,5-a]quinazolin-5(4H)-one (6h). Pale brown crystals (yield 84%), m.p. 255°C (ethanol-DMF). IR: (KBr) ν 1678, 1601 cm^{-1} . ^1H NMR (DMSO): δ 7.3 (dd, 1H, J = 4 Hz, thienyl-C4-H), 7.9 (d, 1H, J = 4 Hz, thienyl-C5-H), 8.0 (d, 1H, J = 4 Hz, thienyl-C3-H), 7.6 – 8.2 (m, 9H, ArH). MS: m/z (%): 346 ($\text{M}^+ + 2$, 9), 345 ($\text{M}^+ + 1$, 25), 344 (M^+ , 100), 317 (25), 235 (38), 185 (34), 109 (26), 103 (13), 90 (14), 77 (30). *Anal Calcd.* for $\text{C}_{19}\text{H}_{12}\text{N}_4\text{OS}$ (344.40): C, 66.26; H, 3.51; N, 16.27. Found: C, 66.55; H, 3.85; N, 16.51 %.

4-Phenyl-1,2,4-triazolo[1,5-a]quinazolin-5(4H)-one (6i). Colorless crystals (yield 82%), m.p. 352°C (ethanol-DMF). IR: (KBr) ν 1690, 1602 cm^{-1} . ^1H NMR (CDCl_3): δ 6.8-8.2 (m, 9H ArH), 8.3 (s, 1H, triazole). MS: m/z (%): 263 ($\text{M}^+ + 1$, 14), 262 (M^+ , 73), 235 (100), 206 (11), 103 (47), 90 (18), 77 (20). *Anal Calcd.* for $\text{C}_{15}\text{H}_{10}\text{N}_4\text{O}$ (262.27): C, 68.69; H, 3.84; N, 21.36. Found: C, 68.42; H, 3.63; N, 21.42 %.

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