Research Article

Nucleosides 8 [18]: Ribosylation of Fused Quinazolines—Synthesis of New [1,2,4]Triazolo[5,1-*b*]- and [1,2,4]Triazino[3,2-*b*]quinazoline Nucleosides of Fluorescence Interest

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[1,2,4]Triazolo[5,1-*b*]- and [1,2,4]triazino[3,2-*b*] quinazolines have been ribosylated by coupling with 1-*O*-acetyl-2,3,5-tri-*O*-benzoyl- β -D-ribofuranose and by using the silylation method, followed by debenzoylation by methanolic sodium methoxide to afford the corresponding free N-nucleosides. Nucleosides obtained have been identified by their spectral analysis. From the UV-visible and fluorescence studies of some nucleosides synthesized, it is found that they have fluorescence properties.

1. Introduction

Most of the modified nucleosides prepared over the last years have been reported to be potential antiviral and chemotherapeutic agents. Quite a few are fluorescent, although most were not originally examined for their photophysical properties and were not employed as fluorescent probes [1–4]. Other fluorescent nucleotide analogues have been developed but, when incorporated into DNA, they are quenched, destabilizing, or of limited sensitivity to environmental change. As a consequence, there is a strong demand for new fluorescent nucleoside analogues with improved spectroscopic properties and this is the purpose of intense research [5–8]. In recent years there has been an increasing interest in the chemistry of 4(3*H*)-quinazolinones because of their biological importance. Many of them show antifungal, antibacterial, anticancer, anti-inflammatory, anticonvulsant, immunotropic, hypolipidemic, antitumor, antiulcer, analgesic, antiproliferative activities and inhibitory effects of thymidylate synthase and poly(ADP-ribose) polymerase (PARP) [9].

A variety of nucleoside derivatives have been prepared through the deletion or change in nature of the functional group present on the heterocyclic base or their sugar moieties. Such analogs permit the synthesis of oligonucleotides in which a single functional group at a preselected position has been deleted or otherwise altered.

Quinazoline nucleosides were first synthesized by Stout and Robins in 1968 [10] as pyrimidine nucleoside analogs and consequent synthetic studies were contributed by Dunkel and Pfeleiderer in the 1990s [11–13]. More recently, several quinazoline-2,4-dione nucleosides have been incorporated into oligonucleotides as written or thymidine substitutes to study the binding affinity and base-pairing selectivity [14–16].

In the view of our observations of the aforementioned biological and fluorescence [1–4, 15, 17] importance of quinazoline derivatives and as part of our continuing interest in the synthesis of new nucleosides [18–24] as expected as new fluorescent molecules, it has encouraged us to study a synthetic strategy of new derivatives of [1,2,4]triazolo[5,1-*b*]- and [1,2,4]triazino[3,2-*b*]quinazoline nucleosides of fluorescence

interest, which have not been yet reported in the literature. Our final goal will be directed to find new nucleosides as expected to be fluorescent and to study their biological activity in further investigations.

2. Experimental

2.1. General. Melting points were determined on a Galenkamp apparatus and are uncorrected. IR spectra were recorded in potassium bromide pellets using Perkin Elmer FTIR 1650 and Pye Unicam SP300 infrared spectrophotometers. ^1H -NMR and ^{13}C -NMR spectra were recorded in deuterated chloroform (CDCl_3) or deuterated dimethyl sulfoxide ($\text{DMSO}-d_6$) using a Varian Gemini 300 NMR spectrometer. Thin layer chromatography was performed on silica gel sheets F 1550 LS 254 of Schleicher & Schuell and column chromatography on Merck silica gel 60 (particle size 0.063–0.20 mm). Mass spectra were recorded on a GCMS-QP 1000 EX Shimadzu and GCMS 5988-A HP spectrometers. UV-Vis spectra were measured by model 2450 a spectrophotometer (Shimadzu) and fluorescence spectra were measured by model RF-5301pc a spectrofluorometer (Shimadzu). Elemental analyses were carried out in the Microanalytical Laboratory of Cairo University, Giza, Egypt. The starting materials 2,3-diaminoquinazolin-4-one (**1**) [25] and ethyl benzoylpyruvate (**18**) [26] were prepared as in the literature reported methods.

2.2. Synthesis of 2–5, 15, and 19

2.2.1. 3H[1,2,4]Triazolo[5,1-*b*]quinazolin-9(1H)-one (2). A mixture of 2,3-diaminoquinazolin-4-one (**1**) (5 g, 0.028 mol) and triethyl orthoformate (150 mL) was heated under reflux for 7 h then left to cool. The solid product formed was filtered and crystallized from EtOH/ H_2O (1:2 v/v) mixture to give 3H[1,2,4]triazolo[5,1-*b*]quinazolin-9(1H)-one (**2**).

Yield (75%); white crystals; m.p. 323–326°C; IR (KBr cm^{-1}): ν 3287 (NH), 1623 (C=O); ^1H NMR ($\text{DMSO}-d_6$): δ 7.36 (d, 2H, aromatic protons, $J = 8.0$ Hz), 8.00 (d, 2H, aromatic protons, $J = 8.0$ Hz), 8.34 (s, 1H, 2-CH), 11.90 (s, 1H, NH); MS (m/z) = 186 (M^+ , 100%). Anal. Calcd for $\text{C}_9\text{H}_6\text{N}_4\text{O}$ (186.17): C, 58.06; H, 3.25; N, 30.09 (%); Found: C, 58.35; H, 3.20; N, 29.70 (%).

2.2.2. 2-Methyl- and 2-Phenyl-3H[1,2,4]triazolo[5,1-*b*]quinazolin-9(1H)-one (3 and 4). To a solution of 2,3-diaminoquinazolin-4-one (**1**) (5 g, 0.028 mol) in 30 mL of dry pyridine, add acetic anhydride (3.0 g, 0.03 mol) or benzoyl chloride (4.2 g, 0.03 mol) in an ice bath under stirring for one hour. The mixture was stirred at room temperature overnight (15 h), left to cool, and then poured into cold water with constant stirring. The solid product that separated out was filtered off, thoroughly washed with water, dried and then recrystallized from acetic acid to give beige crystals of 2-methyl or 2-phenyl-3H[1,2,4]triazolo[5,1-*b*]quinazolin-9(1H)-one (**3**) or (**4**), respectively.

Compound 3. Yield (68%); white crystals; m.p. 333–336°C; IR (KBr cm^{-1}): ν 3380 (NH), 1625 (C=O); ^1H NMR ($\text{DMSO}-d_6$)

δ 2.60 (s, 3H, CH_3), 7.25 (d, 2H, aromatic protons, $J = 8.0$ Hz), 7.80 (d, 2H, aromatic protons, $J = 8.0$ Hz), 11.50 (s, 1H, NH); MS (m/z) = 200 (M^+ , 85%). Anal. Calcd for $\text{C}_{10}\text{H}_8\text{N}_4\text{O}$ (200.20): C, 59.99; H, 4.03; N, 27.99; Found: C, 60.00; H, 4.30; N, 27.80 (%).

Compound 4. Yield (60%); white crystals; m.p. 364–365°C; IR (KBr cm^{-1}): ν 3385 (NH), 1625 (C=O); ^1H NMR ($\text{DMSO}-d_6$) δ 7.20 (d, 2H, aromatic protons, $J = 8.0$ Hz), 7.70 (d, 2H, aromatic protons, $J = 8.0$ Hz), 7.90 (m, 5H, aromatic protons), 11.50 (s, 1H, NH); MS (m/z) = 262 (M^+ , 70%). 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, 68.50; H, 3.66; N, 20.95 (%).

2.2.3. 2,3-Dihydro-2-thioxo-[1,2,4]triazolo[5,1-*b*]quinazolin-9(1H)-one (5). To a stirred solution of 2,3-diaminoquinazolin-4-one (**1**) (10 g, 0.056 mol) in DMF (100 mL), CS_2 (30 mL, 0.47 mol) was added. The reaction mixture was refluxed for 16 h, and the formed precipitate was filtered, washed with methanol, and dried. The product is insoluble in most organic solvents; hence, it was purified by dissolving in 5% KOH. The alkaline solution was cooled in an ice bath and then rendered acidic by addition of conc. HCl under stirring. The precipitate was filtered, washed with distilled water, and dried to yield 2,3-dihydro-2-thioxo-[1,2,4]triazolo[5,1-*b*]quinazolin-9(1H)-one (**5**).

Yield (74%); pale yellow crystals; m.p. 350–352°C; IR (KBr cm^{-1}): ν 3390, 3250 (2NH), 1630 (C=O), 1210 (C=S); ^1H NMR ($\text{DMSO}-d_6$) δ 7.30 (d, 2H, aromatic protons, $J = 8.0$ Hz), 7.96 (d, 2H, aromatic protons, $J = 8.0$ Hz), 11.00 (s, 1H, NH), 11.90 (s, 1H, NH); MS (m/z) = 218 (M^+ , 70%). Anal. Calcd for $\text{C}_9\text{H}_6\text{N}_4\text{OS}$ (214.24): C, 49.53; H, 2.77; N, 25.67; S, 14.69; Found: C, 49.20; H, 3.00; N, 25.80; S, 14.50 (%).

2.2.4. 1,2-Dihydro-4H[1,2,4]triazino[3,2-*b*]quinazolin-3,10-dione (15). A mixture of **1** (5 g, 0.028 mol) and ethyl chloroacetate (3.4 g, 0.028 mol) in a mixture of 30 mL pyridine and 30 mL absolute ethanol was refluxed for 5 hr. After cooling the reaction mixture was poured onto ice water and the solid obtained was filtered and then recrystallized from dioxane to give **15**.

Yield (60%); pale yellow crystals; m.p. 320–322°C; IR (KBr cm^{-1}): ν 3410, 3240 (2NH), 1710, 1665 (2C=O); ^1H -NMR ($\text{DMSO}-d_6$): 4.30 (s, 2H, CH_2), 7.00 (d, 2H, aromatic protons, $J = 8.0$ Hz), 7.50 (d, 2H, aromatic protons, $J = 8.0$ Hz), 10.51 (s, 1H, NH), 11.4 (s, 1H, NH); MS (m/z) = 216 (M^+ , 55%). Anal. Calcd for $\text{C}_{10}\text{H}_8\text{N}_4\text{O}_2$ (216.20): C, 55.55; H, 3.73; N, 25.91; Found: C, 55.20; H, 3.88; N, 25.66 (%).

2.2.5. 2-Phenacyl-(3H)[1,2,4]triazino-[3,2-*b*]quinazoline-2,6(1H)dione (19). A mixture of 2,3-diamino-4(3H)quinazolinone (**1**) (8.93 g, 0.05 mol) and ethyl benzoylpyruvate (**18**) (11.0 g, 0.05 mol) was heated under reflux in 100 mL of dry pyridine and 100 mL absolute ethanol for 3 hours and cooled. The solid precipitated was collected, washed with ethanol, and crystallized from acetic acid to give pale yellow crystals of 2-phenacyl-(3H)[1,2,4]triazino-[3,2-*b*]quinazoline-2,6(1H)dione (**19**).

Yield 65%, m.p. 305°C (Lit [24] m.p. 305–307°C); IR (KBr) ν 3350 (N–H), 3050 (C–H aromatic), 1670, 1600 (2C=O); ^1H

NMR (DMSO- d_6): δ 6.85 (s, 1H, =CH), 7.50–7.80 (m, 9H, Ar-H), 14.00 (s, 2H, 2N-H); ^{13}C NMR (DMSO- d_6): δ 115.1, 122.0–134.0, 137.2, 145.1, 155.0, 160.2, 162.1, 164.0, 181.0. EIMS, m/z : 332 (M^+), Anal. Calcd for $\text{C}_{18}\text{H}_{12}\text{N}_4\text{O}_3$ (332.31); calcd. C, 65.06; H, 3.64; N, 16.86; Found C, 65.38; H, 3.58; N, 16.72 (%).

2.3. Ribosylation of 2–5, 15 and 19. Synthesis of 4-(2',3',5'-Tri-*O*-benzoyl- β -D-ribofuranosyl)-3*H*[1,2,4]triazolo[5,1-*b*]quinazolin-9(1*H*)-one (7), 2-Methyl-4-(2',3',5'-tri-*O*-benzoyl- β -D-ribofuranosyl)-3*H*[1,2,4]triazolo[5,1-*b*]quinazolin-9(1*H*)-one (8), 2-Phenyl-4-(2',3',5'-tri-*O*-benzoyl- β -D-ribofuranosyl)-3*H*[1,2,4]triazolo[5,1-*b*]quinazolin-9(1*H*)-one (9), 2,3-Dihydro-2-thioxo-4-(2',3',5'-tri-*O*-benzoyl- β -D-ribofuranosyl)-[1,2,4]triazolo[5,1-*b*]quinazolin-9(1*H*)-one (10), 4-(2',3',5'-Tri-*O*-benzoyl- β -D-ribofuranosyl)-1,2-dihydro-4*H*[1,2,4]triazino[3,2-*b*]quinazolin-3,10-dione (16), 2-Phenacyl-4-(2',3',5'-tri-*O*-benzoyl- β -D-ribofuranosyl)-(3*H*)[1,2,4]triazino-[3,2-*b*]quinazoline-2,6(1*H*)dione (20).

General Procedure. A mixture of each [1,2,4]triazolo[5,1-*b*]quinazolines (2–5), [1,2,4]triazino[3,2-*b*]quinazolines (15) and (19) (0.02 mol) and dry hexamethyldisilazane (150 mL) was heated under reflux for 10–15 h with a catalytic amount of ammonium sulfate (0.1 g). After the clear solution was cooled, it was evaporated to dryness under anhydrous condition to give the silylated derivative, which directly was dissolved in 50 mL of dry 1,2-dichloroethane. To this was added a solution of 1-*O*-acetyl-2,3,5-tri-*O*-benzoyl- β -D-ribofuranose (6) (9.6 g, 0.02 mol) in dry 1,2-dichloroethane (50 mL) was then added. The mixture was cooled in ice bath and a solution of trimethylsilyl trifluoromethanesulfonate (Triflate) (4 mL, 0.02 mol) in dry 1,2-dichloroethane (20 mL) was added dropwise. It was stirred at room temperature for 24 h, and then diluted with chloroform (500 mL), washed with a saturated solution of aqueous sodium bicarbonate (200 mL), water (3 $\times$ 150 mL), and dried over anhydrous sodium sulfate. The solvent was removed in vacuo and the residue was chromatographed on silica gel with chloroform as eluent to afford white solid which was crystallized from EtOH/1,4-dioxane (v/v 1:1) to yield colorless crystals of the corresponding nucleoside derivatives 7–10, 16, and 20, respectively.

2.3.1. Compound (7). Yield (62%), m.p. 185°C–187°C; IR (KBr) ν cm^{-1} : 1730, 1630; ^1H NMR (CDCl_3): δ 4.60–4.90 (m, 3H, 2H-5', H-4'), 6.10 (d, 1H, H-1', $J_{1',2'} = 7.5$ Hz), 6.25–6.30 (m, 1H, H-3'), 6.40–6.45 (m, 1H, H-2'), 7.36 (d, 2H, aromatic protons, $J = 8.0$ Hz), 8.00 (d, 2H, aromatic protons, $J = 8.0$ Hz), 8.10–8.15 (m, 15H, Ar-H), 8.34 (s, 1H, 2-CH); ^{13}C NMR (CDCl_3): 59.1, 71.5, 78.3, 84.2, 93.0, 121.2, 125.0–140.0, 151.2, 162.1, 168.4; Anal. Calcd. for $\text{C}_{35}\text{H}_{26}\text{N}_4\text{O}_8$ (630.60): C, 66.66; H, 4.16; N, 8.88 (%); Found: C, 66.59; H, 4.30; N, 8.70 (%).

2.3.2. Compound (8). Yield (60%), m.p. 168°C–169°C; IR (KBr) ν cm^{-1} : 1724, 1630; ^1H NMR (CDCl_3): δ 2.55 (s, 3H, CH_3), 4.65–4.92 (m, 3H, 2H-5', H-4'), 6.15 (d, 1H, H-1',

$J_{1',2'} = 7.5$ Hz), 6.25–6.30 (m, 1H, H-3'), 6.40–6.45 (m, 1H, H-2'), 7.20 (d, 2H, aromatic protons, $J = 8.0$ Hz), 7.85 (d, 2H, aromatic protons, $J = 8.0$ Hz), 8.00–8.15 (m, 15H, Ar-H); ^{13}C NMR (CDCl_3): 35.2, 59.1, 71.0, 78.3, 84.4, 93.2, 121.0, 125.1–140.0, 151.3, 160.4, 168.1; Anal. Calcd. for $\text{C}_{36}\text{H}_{28}\text{N}_4\text{O}_8$ (644.63): C, 67.07; H, 4.38; N, 8.69 (%); Found: C, 67.30; H, 4.20; N, 8.50 (%).

2.3.3. Compound (9). Yield (55%), m.p. 160°C–161°C; IR (KBr) ν cm^{-1} : 1730, 1625; ^1H NMR (CDCl_3): δ 4.60–4.90 (m, 3H, 2H-5', H-4'), 6.10 (d, 1H, H-1', $J_{1',2'} = 7.5$ Hz), 6.25–6.30 (m, 1H, H-3'), 6.40–6.45 (m, 1H, H-2'), 7.20 (d, 2H, aromatic protons, $J = 8.0$ Hz), 7.70 (d, 2H, aromatic protons, $J = 8.0$ Hz), 7.90–8.00 (m, 20H, aromatic protons); ^{13}C NMR (CDCl_3): 59.2, 71.4, 78.1, 84.2, 93.2, 12.2, 120.0–135.0, 151.2, 160.2, 168.0; Anal. Calcd. for $\text{C}_{41}\text{H}_{30}\text{N}_4\text{O}_8$ (706.7): C, 69.68; H, 4.28; N, 7.93 (%); Found: C, 69.50; H, 4.15; N, 7.65 (%).

2.3.4. Compound (10). Yield (58%), m.p. 203°C–204°C; IR (KBr) ν cm^{-1} : 1730, 1620; ^1H NMR (CDCl_3): δ 4.60–4.95 (m, 3H, 2H-5', H-4'), 6.15 (d, 1H, H-1', $J_{1',2'} = 7.5$ Hz), 6.20–6.30 (m, 1H, H-3'), 6.35–6.40 (m, 1H, H-2'), 7.30 (d, 2H, aromatic protons, $J = 8.0$ Hz), 7.96 (d, 2H, aromatic protons, $J = 8.0$ Hz), 8.00–8.15 (m, 15H, Ar-H), 11.25 (s, 1H, NH); ^{13}C NMR (CDCl_3): 59.5, 71.2, 78.1, 84.0, 93.2, 121.4, 125.0–140.2, 151.2, 159.2, 168.0, 175.2 (C=S); Anal. Calcd. for $\text{C}_{35}\text{H}_{26}\text{N}_4\text{O}_8\text{S}$ (662.67): C, 63.44; H, 3.95; N, 8.45; S, 4.84 (%); Found: C, 63.50; H, 4.00; N, 8.20; S, 4.55 (%).

2.3.5. Compound (16). Yield (55%), m.p. 190°C–192°C; IR (KBr) ν cm^{-1} : 1725, 1630, 1600; ^1H NMR (CDCl_3): δ 4.25 (s, 2H, CH_2), 4.60–4.95 (m, 3H, 2H-5', H-4'), 6.10 (d, 1H, H-1', $J_{1',2'} = 7.5$ Hz), 6.20–6.30 (m, 1H, H-3'), 6.35–6.40 (m, 1H, H-2'), 7.30 (d, 2H, aromatic protons, $J = 8.0$ Hz), 7.95 (d, 2H, aromatic protons, $J = 8.0$ Hz), 8.10–8.15 (m, 15H, Ar-H), 11.00 (s, 1H, NH); ^{13}C NMR (CDCl_3): 29.0, 50.2, 59.1, 71.1, 78.4, 84.2, 93.0, 121.2, 125.5–140.3, 151.0, 159.2, 164.1, 168.0; Anal. Calcd. for $\text{C}_{36}\text{H}_{28}\text{N}_4\text{O}_9$ (660.63): C, 65.45; H, 4.27; N, 8.48 (%); Found: C, 65.30; H, 4.10; N, 8.15 (%).

2.3.6. Compound (20). Yield (58%), m.p. 188–190°C; IR (KBr) ν cm^{-1} : 1735, 1635, 1620, 1590; ^1H NMR (CDCl_3): δ 4.10 (s, 2H, CH_2), 4.60–4.95 (m, 3H, 2H-5', H-4'), 6.15 (d, 1H, H-1', $J_{1',2'} = 7.5$ Hz), 6.20–6.30 (m, 1H, H-3'), 6.35–6.40 (m, 1H, H-2'), 6.90 (s, 1H, =CH), 7.10 (d, 2H, aromatic protons, $J = 8.0$ Hz), 7.30 (d, 2H, aromatic protons, $J = 8.0$ Hz), 7.70–7.85 (m, 20H, Ar-H), 12.5 (s, 1H, NH); ^{13}C NMR (CDCl_3): 59, 71, 78, 84, 93, 121, 125–140, 151, 159, 163, 168, 180. Anal. Calcd. for $\text{C}_{44}\text{H}_{32}\text{N}_4\text{O}_{10}$ (776.75): C, 68.04; H, 4.15; N, 7.21 (%); Found: C, 67.88; H, 4.00; N, 7.00 (%).

2.4. Deprotection of 7–10, 16, and 20. Synthesis of 4-(β -D-Ribofuranosyl)-3*H*[1,2,4]triazolo[5,1-*b*]quinazolin-9(1*H*)-one (11), 2-Methyl-4-(β -D-ribofuranosyl)-3*H*[1,2,4]triazolo[5,1-*b*]quinazolin-9(1*H*)-one (12), 2-Phenyl-3-(β -D-ribofuranosyl)-3*H*[1,2,4]triazolo[5,1-*b*]quinazolin-9(1*H*)-one (13),

2,3-Dihydro-2-thioxo-4-(β -D-ribofuranosyl)-[1,2,4]triazolo[5,1-*b*]quinazolin-9(1*H*)-one (**14**), 4-(β -D-Ribofuranosyl)-4*H*[1,2,4]triazino[3,2-*b*]quinazolin-3,10-dione (**17**), 2-Phenacyl-4-(β -D-ribofuranosyl)-(3*H*)[1,2,4]triazino-[3,2-*b*]quinazoline-2,6(1*H*)dione (**21**).

General Procedure. A mixture of each protected nucleoside **7–10**, **16**, or **20** (0.001 mol for each), absolute methanol (20 mL) and sodium methoxide (0.06 g, 0.0011 mol) was stirred at room temperature for 48 h. Evaporation of the solvent under vacuum gave a colorless solid, which was dissolved in hot water and neutralized with acetic acid. The precipitate was filtered off and afforded upon crystallization from water/dioxane (v/v 1 : 1) to give the corresponding nucleosides **11–14**, **17**, and **21**, respectively, as colorless crystals.

2.4.1. Compound (11). Yield (48%), m.p. 245°C–246°C; IR (KBr) ν cm⁻¹: 3350 (OH), 1600; ¹H NMR (DMSO-*d*₆): δ 3.40–3.55 (m, 2H, 5',5'-H), 3.85–3.95 (m, 1H, 4'-H), 4.10–4.15 (m, 1H, 3'-H), 4.30–4.40 (m, 1H, 2'-H), 4.65–4.75 (t, 1H, 5'-OH), 5.0–5.10 (d, 1H, 3'-OH), 5.25 (s, 1H, H-5), 5.35–5.45 (d, 1H, 2'-OH), 6.10 (d, 1H, $J_{1',2'} = 7.0$ Hz, 1'-H), 6.95 (d, 2H, aromatic protons, $J = 8.0$ Hz), 7.2 (d, 2H, aromatic protons, $J = 8.0$ Hz), 8.10 (s, 1H, 2-CH). Anal. Calcd. for C₁₄H₁₄N₄O₅ (318.28): C, 52.83; H, 4.43; N, 17.60 (%); Found: C, 52.60; H, 4.25; N, 17.50 (%).

2.4.2. Compound (12). Yield (40%), m.p. 230°C–232°C; IR (KBr) ν cm⁻¹: 3380 (OH), 1595; ¹H NMR (DMSO-*d*₆): δ 2.30 (s, 3H, CH₃), 3.5–3.6 (m, 2H, 5',5'-H), 3.9–4.0 (m, 1H, 4'-H), 4.15–4.2 (m, 1H, 3'-H), 4.35–4.45 (m, 1H, 2'-H), 4.7–4.8 (t, 1H, 5'-OH), 5.1–5.2 (d, 1H, 3'-OH), 5.30 (s, 1H, H-5), 5.4–5.5 (d, 1H, 2'-OH), 6.10 (d, 1H, $J_{1',2'} = 7.0$ Hz, 1'-H), 6.95 (d, 2H, aromatic protons, $J = 8.0$ Hz), 7.2 (d, 2H, aromatic protons, $J = 8.0$ Hz). Anal. Calcd. for C₁₅H₁₆N₄O₅ (332.31): C, 54.21; H, 4.85; N, 16.86 (%); Found: C, 54.10; H, 4.55; N, 17.00 (%).

2.4.3. Compound (13). Yield (40%), m.p. 222°C–223°C; IR (KBr) ν cm⁻¹: 3380 (OH), 1585; ¹H NMR (DMSO-*d*₆): δ 3.55–3.65 (m, 2H, 5',5'-H), 3.95–4.10 (m, 1H, 4'-H), 4.15–4.25 (m, 1H, 3'-H), 4.40–4.50 (m, 1H, 2'-H), 4.75–4.85 (t, 1H, 5'-OH), 5.15–5.25 (d, 1H, 3'-OH), 5.35 (s, 1H, H-5), 5.45–5.55 (d, 1H, 2'-OH), 6.10 (d, 1H, $J_{1',2'} = 7.0$ Hz, 1'-H), 6.95 (d, 2H, aromatic protons, $J = 8.0$ Hz), 7.2 (d, 2H, aromatic protons, $J = 8.0$ Hz), 7.70–7.80 (m, 5H, Ar-H). Anal. Calcd. for C₂₀H₁₈N₄O₅ (394.38): C, 60.91; H, 4.60; N, 14.21 (%); Found: C, 60.85; H, 4.50; N, 14.10 (%).

2.4.4. Compound (14). Yield (40%), m.p. 256°C–258°C; IR (KBr) ν cm⁻¹: 3380 (OH), 1600; ¹H NMR (DMSO-*d*₆): δ 2.30 (s, 3H, CH₃), 3.5–3.6 (m, 2H, 5',5'-H), 3.9–4.0 (m, 1H, 4'-H), 4.15–4.2 (m, 1H, 3'-H), 4.35–4.45 (m, 1H, 2'-H), 4.7–4.8 (t, 1H, 5'-OH), 5.1–5.2 (d, 1H, 3'-OH), 5.30 (s, 1H, H-5), 5.4–5.5 (d, 1H, 2'-OH), 6.10 (d, 1H, $J_{1',2'} = 7.0$ Hz, 1'-H), 6.95 (d, 2H, aromatic protons, $J = 8.0$ Hz), 7.2 (d, 2H, aromatic protons, $J = 8.0$ Hz), 11.5 (s, 1H, NH). Anal. Calcd. for C₁₄H₁₄N₄O₅S (350.35): C, 47.99; H, 4.03; N, 15.99; S, 9.15 (%); Found: C, 47.80; H, 3.99; N, 16.20; S, 8.95 (%).

2.4.5. Compound (17). Yield (48%), m.p. 280°C–281°C; IR (KBr) ν cm⁻¹: 3350 (OH), 1595; ¹H NMR (DMSO-*d*₆): δ 3.55–3.65 (m, 2H, 5',5'-H), 3.95–4.10 (m, 1H, 4'-H), 4.20–4.25 (m, 1H, 3'-H), 4.30 (s, 2H, CH₂), 4.35–4.45 (m, 1H, 2'-H), 4.7–4.8 (t, 1H, 5'-OH), 5.1–5.2 (d, 1H, 3'-OH), 5.30 (s, 1H, H-5), 5.4–5.5 (d, 1H, 2'-OH), 6.15 (d, 1H, $J_{1',2'} = 7.0$ Hz, 1'-H), 7.00 (d, 2H, aromatic protons, $J = 8.0$ Hz), 7.4 (d, 2H, aromatic protons, $J = 8.0$ Hz), 11.00 (s, 1H, NH). Anal. Calcd. for C₁₅H₁₆N₄O₆ (348.31): C, 51.72; H, 4.63; N, 16.09 (%); Found: C, 51.60; H, 4.45; N, 15.80 (%).

2.4.6. Compound (21). Yield (40%), m.p. 265°C–267°C; IR (KBr) ν cm⁻¹: 3350 (OH), 1600; ¹H NMR (DMSO-*d*₆): δ 3.5–3.6 (m, 2H, 5',5'-H), 3.9–4.0 (m, 1H, 4'-H), 4.15–4.2 (m, 1H, 3'-H), 4.35–4.45 (m, 1H, 2'-H), 4.7–4.8 (t, 1H, 5'-OH), 5.1–5.2 (d, 1H, 3'-OH), 5.30 (s, 1H, H-5), 5.4–5.5 (d, 1H, 2'-OH), 6.15 (d, 1H, $J_{1',2'} = 7.0$ Hz, 1'-H), 6.85 (s, 1H, =CH), 7.00 (d, 2H, aromatic protons, $J = 8.0$ Hz), 7.4 (d, 2H, aromatic protons, $J = 8.0$ Hz), 7.50–7.73 (m, 5H, Ar-H), 11.5 (s, 1H, NH). Anal. Calcd. for C₂₃H₂₀N₄O₇ (464.43): C, 59.48; H, 4.34; N, 12.06 (%); Found: C, 59.22; H, 4.15; N, 11.89 (%).

3. Results and Discussion

Refluxing of the starting material 2,3-diaminoquinazolin-4-one (**1**), which is prepared according to the literature reported method [25], in triethyl orthoformate for 7 hours afforded one product, which is identified to be 3*H*[1,2,4]triazolo[5,1-*b*]quinazolin-9(1*H*)-one (**2**) [25, 27] (Scheme 1). Reaction of **1** with acetic anhydride or benzoyl chloride in pyridine yielded the corresponding 2-methyl- or 2-phenyl-3*H*[1,2,4]triazolo[5,1-*b*]quinazolin-9(1*H*)-one (**3** or **4**), respectively (Scheme 1). Also, heating of **1** in carbon disulphide in ethanol containing equivalent amount of potassium hydroxide gave 2,3-dihydro-2-thioxo-[1,2,4]triazolo[5,1-*b*]quinazolin-9(1*H*)-one (**5**) (Scheme 1).

Although the products **2–5** are reported in the literature [25, 27], no complete characterization data are found in the hand.

So, the structures of **2–5** were established and confirmed on the basis of their elemental analyses and spectral data (see Section 2).

With the aim of expanding the synthetic potential of the 2,3-diaminoquinazolin-4-one (**1**) formed, it is interesting to study the reaction of **1** with ethyl chloroacetate. When such reaction is carried out in relaxing pyridine/ethanol (v/v 1 : 1) yielded the new 4*H*[1,2,4]triazino[3,2-*b*]quinazolin-3,10-Dione (**15**) (Scheme 3). The structure of the product **15** was based on the assumption that the reaction in basic condition allowed it to proceed through elimination of sodium chloride from the basic 3-NH₂, followed by cyclisation [28].

Furthermore, the reaction of **1** with ethyl benzoylpyruvate (**18**) [26] in a mixture of pyridine/ethanol (v/v 1 : 1) yielded as reported of our laboratory [28], 2-phenacyl-(3*H*)[1,2,4]triazino-[3,2-*b*]quinazoline-2,6(1*H*)dione (**19**) (Scheme 3).

The formation of nucleoside derivatives of the above compounds, **2–5**, **15**, and **19** has been studied, hence ribosylation of each **2–5**, **15**, and **19** with 1-*O*-acetyl-2,3,5-tri-*O*-benzoyl- β -D-ribofuranose (**6**) was carried out by the silylation method

SCHEME 1: Synthesis of 4-β-D-ribofuranosyl-[1,2,4]triazolo[5,1-b]quinazoline-9(3H)one derivatives (11-14).

according to Vorbruggen method [29]. Each of such compounds, 2-5, 15 and 19 was refluxed in hexamethyldisilazane (HMDS) with ammonium sulfate as a catalyst followed by stirring of the silylated product with ribose derivative 6 in dry acetonitrile and TMS-triflate ($\text{CF}_3\text{SO}_2\text{OSiMe}_3$) at room temperature for 24 h. This method yielded products in 53–62% of the corresponding benzoylated nucleosides 7-10 (Scheme 1), 16 and 20 (Scheme 3), respectively.

The structures of the latter products 7-10, 16, and 20 were established and confirmed on the bases of their elemental analyses and spectral data. Thus, their ^1H NMR spectra in CDCl_3 showed a doublet signal at δ 6.10–6.15 assigned to the anomeric proton of the ribose moiety with a spin-spin coupling constant equal to 7.5 Hz, which corresponds to a diaxial orientation for the 1'- and 2'-H protons; that is, the β-configuration (high $J_{1',2'}$) [18–24]. ^{13}C NMR Spectrum in CDCl_3 of 7 as an example revealed signals at δ 59.1, 71.5, 78.3, 84.2, 93.0, 121.2, 125.0–140.0, 151.2, 162.1, 168.4 (CO), while that of nucleoside 10 revealed a characteristic signal

of C=S chemical shifts at δ 175.2. A distinction between the O-, N- and S-nucleosides was possible by comparison of the ^1H NMR and ^{13}C NMR spectra with those of literature data of similar compounds [30]. $^{13}\text{C}=\text{S}$ chemical shifts of δ 172.0 were reported for cycloalkyl[4,5]-thieno[2,3-*d*]pyrimidin-4-one-2-thione, while 2-alkylthio-cycloalkyl[4,5]thieno[2,3-*d*]pyrimidin-4-one show chemical shifts of C-2 (=C-S-) around δ 159 ppm.

The structure of nucleoside 10 may exist in two forms, thiol form (10A) and thione form (10B or 10C) (Scheme 2). The results reported above of ^{13}C NMR spectra of compound 10 of C=S at δ 175 ppm indicate the formation of N-nucleosides as thione form (10B or 10C) [30–33].

Also, the structure of the protected nucleosides 16 and 20 was confirmed by their IR, ^1H & ^{13}C NMR spectra, and microanalyses. Their IR spectra revealed three characteristic bands for (C=O)-stretching bands of product 16 and four bands for product 20 (see Section 2). The ^1H NMR spectra in CDCl_3 of 16 and 20 showed as in the case of that

SCHEME 2: Tautomeric forms of **10**.TABLE 1: Optical properties of the products **2**, **5**, **7**, **11**, **15**, **16**, **17**, **19**, and **20**.

Compound no.	2	5	7	11	15	16	17	19	20
$\lambda_{\max}$ abs. (nm)	318	325	330	342	318	298	286	368	328
$\lambda_{\max}$ em. (nm)	388	376	374	379	390	331	335	382	376
Bandgap (ev)	3.9	3.81	3.76	3.63	3.9	4.17	4.34	3.37	3.78

of compounds **7–10** signals at δ 6.10–6.15 assigned to the anomeric proton of the ribose moiety with a spin-spin coupling constant equal to 7.5 Hz (β -configuration [18–24]). ^{13}C NMR Spectrum in CDCl_3 of **16** as an example revealed signals at δ 29, 50, 59, 71, 78, 84, 93, 121, 125–140, 151, 159, 164, 168 (CO).

However, debenzoylation of each of the blocked nucleosides, **7–10**, **16**, and **20** with methanolic sodium methoxide at room temperature for 48 h, yielded the corresponding free N-nucleosides, **11–14** (Scheme 1), **17** and **21** (Scheme 3), respectively. The ^1H NMR spectra of the free nucleosides **11–14**, **17**, and **21** showed the expected base moiety protons in addition to the sugar moiety protons (see Section 2).

4. Optical Properties

4.1. Introduction. Some quinazoline-based fluorescent nucleosides have been synthesized for photophysical studies and applications in probing nucleic acid structure, dynamics, and recognition [34]. These size-expanded U analogues exhibit fluorescent emission wavelengths that span 155 nm, from 335 to 490 nm. Each nucleoside has unique characteristic response to changes in its microenvironment. These distinct features lead to a variety of applications in biological assays, many of which have been explored [34].

As part of our continuing interest are optical properties [35–41]. We just perform the UV-visible and fluorescence

SCHEME 3: Synthesis of 4- β -D-ribofuranosyl-[1,2,4]triazino[3,2-*b*]quinazoline-3,10(4*H*)dione (**17**) and 4- β -D-ribofuranosyl-2-phenacyl[1,2,4]triazino[3,2-*b*]quinazoline-3,10(1*H*)dione (**21**).

FIGURE 1: UV-Vis absorption spectra of (a) samples 7, 16, 19, 20, and (b) samples 2, 5, 11, 15, 16, and 17 (all samples are kept at 0.02 M).

FIGURE 2: The fluorescence emission spectra of (a) samples: 2, 7, 15, 16, and 17, (b) samples: 5, 11, 19 and 20 (all samples are kept at 0.02 M).

measurements of a series of fused quinazoline nucleosides prepared. Further studies of optical properties of such nucleosides are being under investigation and the results will be reported in due time.

4.2. UV-Visible Study. Absorption spectra of all samples 2, 5, 7, 11, 15, 16, 17, 19, and 20 show an intense lowest energy charge-transfer absorption band in the UV-visible region. The position of this band is strongly influenced by the structure of the compounds, for example, by the type of λ system (aryl or heteroaryl groups). The shifts of the absorption maxima are proportional to the intramolecular

charge-transfer between the electron-releasing and withdrawing groups. In general, the stronger the donor and/or acceptor group, the smaller the energy difference between ground and excited states, and the longer the wavelength of absorption according to Chemla and Zyss [42].

As shown in Figure 1, the absorption spectra for all samples 2, 5, 7, 11, 15, 16, 17, 19 and 20 were measured as small volume samples in methanol in quartz cuvettes. The concentration of all samples are kept at 0.02 M in methanol. The effect of different solvents are simple, so we used one solvent for illustration the $\lambda_{\max}$ abs. and $\lambda_{\max}$

em. The absorption maximum and estimated optical band gaps (E_g^{op}) are summarized in Table 1. It is well known that there are a lot of methods to determine band gap (E_g) [43–45]; the optical band gap can be calculated also on the basis of the optical absorption spectrum [46, 47]. The low-wavelength absorption peak less than (349–405 nm) is attributed to the π - π^* transition of donor units whereas the high-wavelength peak (483–575 nm) is believed to be related to the intramolecular π - π^* transition between the ground and the excited states [48].

4.3. Fluorescence Study. As shown in Figure 2, fluorescence spectra were carried out with all samples **2**, **5**, **7**, **11**, **15**, **16**, **17**, **19**, and **20** were kept at 0.02 M concentration in methanol. In order to relate the fluorescence properties to the nature of the nature of the donating and the acceptor groups attached to these moieties [49].

From the forgoing UV data (λ_{max} , in methanol, 286–368 nm) reported for some of our products, they are almost consistent with that of similar quinazolines derivatives which exhibit absorption wavelengths (λ_{max} , in methanol or acetone) from 280 to 399 nm [50, 51]. However, our products exhibit fluorescent emission wavelengths from 331 to 390, while some quinazoline-based fluorescent nucleosides have been synthesized for photophysical studies that exhibit fluorescent emission wavelengths from 335 to 490 nm [34], depending upon their chromophores.

5. Conclusion

Silylation method was found to be a convenient method for the ribosylation of [1,2,4]triazolo[5,1-*b*]quinazoline derivatives **2–5** and [1,2,4]triazino[3,2-*b*]quinazoline derivatives, **15** and **19** with 1-*O*-acetyl-2,3,5-tri-*O*-benzoyl- β -D-ribofuranose (**6**) to get the corresponding benzoylated N-nucleosides **7–10**, **16** and **20**, respectively. Deprotection of the latter by using methanolic sodium methoxide gave new free N-nucleosides **11–14**, **17**, and **21**, respectively, in moderate yields. Nucleosides obtained have been identified by their spectral analysis. In agreement with the results obtained from the UV-visible, fluorescence studies, the samples **2**, **5**, **7**, **11**, **15**, **16**, **17**, **19**, and **20** synthesized can be applied for the manufacture of new materials with nonlinear optical (NLO) properties [52], as fluorescent markers, due to their strong fluorescence, or as light emitters in organic light emitting devices (OLEDs) [53].

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