Synthesis and Characterization of 1,3,4-Oxadiazole-Triazolopyridinone Hybrid Derivatives as New Blue-Greenish Photoluminescent Materials

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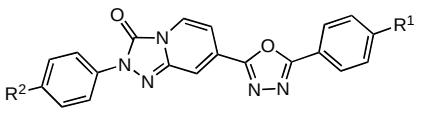

5a. R^1 = H, R^2 = H
5b. R^1 = H, R^2 = Me
5c. R^1 = H, R^2 = OMe
5d. R^1 = Me, R^2 = H
5e. R^1 = Me, R^2 = Me
5f. R^1 = Me, R^2 = OMe
5g. R^1 = Cl, R^2 = H
5h. R^1 = Cl, R^2 = Me
5i. R^1 = Cl, R^2 = OMe

Recently, New functionalized oxadiazole-triazolopyridinone hybrid compounds were investigated as photoluminescent materials. In this work, we introduce triazolopyridinone to synthesize a series of oxadiazole-triazolopyridinone hybrid derivatives as potential photoluminescent materials and explore the effect of modification of the triazolopyridinone moiety. The λ_{max} values of the photoluminescence (PL) spectra of 1,3,4-oxadiazole–triazolopyridinone hybrids are promoted to longer wavelengths (470–486 nm) than the traditional 1,2,3-triazole derivatives (410–425 nm) in solutions. PL spectra **5a**, **5d**, and **5g** of the vacuum evaporated films on quartz substrates, with a maximum at 487 nm, shows a red-shift (~15–20 nm), with respect to the solution spectrum. The solution fluorescence quantum yields (Φ_f) were measured, all of which fell into the range 0.65–0.76, and were determined relative to that of 2-phenyl-5-(4-biphenyl)-1,3,4-oxadiazole in benzene (Φ_f = 0.80). 1,3,4-Oxadiazole–triazolopyridinone hybrid derivatives show clearly non-reversible reduction processes in cyclic voltammogram measurements. Following spectroscopic studies and observation of the electrochemical behaviors, 1,3,4-oxadiazole–triazolopyridinone derivatives were determined to be potential efficient blue-greenish photoluminescent materials.

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INTRODUCTION

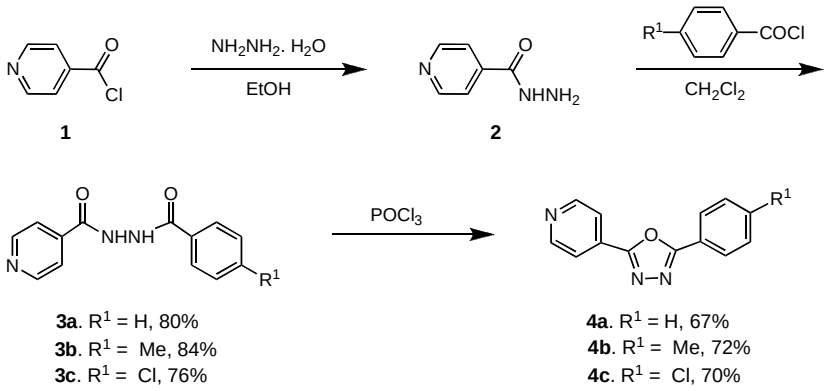
Organic light-emitting devices (OLEDs) have been an important subject in recent years due to their applications in displays and electronic industries [1-3]. Furthermore, blue light-emitting materials have been essential because of their wild applicability [4-7]. Adachi, *et al*. reported 2-(biphenyl-4-yl)-5-(4-*tert*-butylphenyl)-1,3,4-oxadiazole (PBD) as an excellent electron transport material (ETM) in an organic multilayer electroluminescent (EL) diode [8]. After this report, 1,3,4-oxadiazole derivatives have been widely exploited as electron-transporting, hole-blocking (ETHB) materials in electroluminescent (EL) devices due to their electron-deficient nature, high thermal stability and high photoluminescence quantum yield (PLQL) [9-10]. 1,3,4-Oxadiazole-based heterocyclic compounds were also enthusiastically investigated. For example, 1,3,4-oxadiazole-pyridine hybrids [11], 1,3,4-oxadiazole-pyrimidine hybrids [11], 1,3,4-oxadiazole-carbazole [12] and 1,3,4-oxadiazole-spirobifluorene [13] were well studied. The heterocyclic moieties on the molecular structure can provide improved hole/injection, transport properties and confer rigidity.

In this work, novel 1,3,4-oxadiazole-triazolopyridinone hybrid derivatives were synthesized in order to explore the effect of modification of the triazolopyridinone moiety [14]. We synthesized a series of 8-(1,3,4-oxadiazol-2-yl)-[1,2,4]triazodone derivatives (**5a**–**5i**) in which *N*1-phenyl group and the substituted phenyl in 1,3,4-oxadiazole ring were modified in several ways. Following the spectroscopic studies and the measurements of cyclic voltammogram, 1,3,4-oxadiazole-triazolopyridinone derivatives were highly efficient blue-greenish electroluminescent.

RESULTS AND DISCUSSION

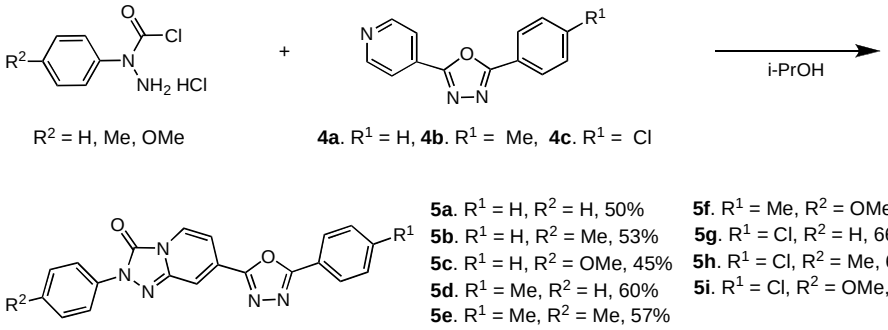
Synthesis of 1,3,4-oxadiazol-2-yl-[1,2,4]triazolo[4,3-a]pyridin-3(2*H*)-one derivatives (5a–5i). The synthetic route of 4-(5-phenyl-1,3,4-oxadiazol-2-yl)pyridine derivatives (**4a**–**4c**) is shown in Scheme 1. Isonicotinyl

chloride **1** reacted with hydrazine monohydrhydrate to generate isonicotinyl hydrazine **2** [15]. Then various benzoyl chloride (para-R^1 = H, Me, and Cl) were treated with isonicotinyl hydrazine **2** to give the corresponding 1-isonicotinyl-2-nicotinyl hydrazines **3a**–**3c** [16]. 1-Isonicotinyl-2-nicotinyl hydrazines **3a**–**3c** were performed by the dehydration–cyclization using fresh $POCl_3$ to produce 1,3,4-oxadiazol-2-ylpyridine derivatives **4a**–**4c**.



Scheme 1

α-Chloroformylarylhydrazines hydrochloride were synthesized through our previous published procedures [17]. 1,3,4-Oxadiazol-2-ylpyridine derivatives **4a**–**4c** reacted with α-chloroformylarylhydrazine in *i*-PrOH at 80 °C for 2 h to generate 1,3,4-oxadiazole-triazolopyridinone derivatives **5a**–**5i** in 45–66 % yields (see Scheme 2).



Scheme 2

Photophysical properties. The UV-vis spectra of 1,3,4-oxadiazol-2-ylpyridine **4a**–**4c** and 1,3,4-oxadiazole-triazolopyridinone derivatives **5a**–**5i** was measured in CH_2Cl_2 solutions. The λ_{max} values of **4a**–**4c** are between 310 and 314 nm. 1,3,4-oxadiazole-triazolopyridinone derivatives **5a**–**5i** with the moiety of triazolopyridinone have gradual red shifts to 300–318 nm due to extension of conjugation (see Table 1). The photoluminescence (PL) spectra of 1,3,4-oxadiazol-2-ylpyridine derivatives **4a**–**4c** and 1,3,4-oxadiazole-triazolopyridinone derivatives **5a**–**5i** are shown in Table 1. The emission wavelengths for **4a**–**4c** are between 410 nm and 425 nm in $CHCl_3$ solution. The λ_{max} values of photoluminescence are around 470–486 nm of 1,3,4-oxadiazole-triazolopyridinone derivatives **5a**–**5i** with the moiety of triazolopyridinone exhibit blue-greenish fluorescence in $CHCl_3$ solution. The long range of substitution effects on R^1 and R^2 positions are not clearly function for absorption and emission spectra (see Table 1 and Figure 1). The solution fluorescence quantum yields (Φ_f) of **5a**–**5i**, all of which fall in the range 0.66–0.76, were determined relative to that of 2-phenyl-5-(4-biphenyl)-1,3,4-oxzdiazole in benzene (Φ_f = 0.80, see Table 1) [18]. The PL spectra **5a**, **5d**, and **5g** of the vacuum evaporated films on quartz substrates, with a maximum at 487 nm, shows a red-shift (~15–20 nm), with respect to the solution spectrum as shown in Figure 2.

Cyclic Voltammetry Measurements: The electrochemical behavior of 1,3,4-oxadiazole-triazolopyridinone derivatives **5a**–**5i** were investigated by cyclic voltammetry. The measurements were carried out with a platinum electrode in CH_2Cl_2 containing tetrabutylammonium hexafluorophosphate ($TBAPF_6$). The potential was measured against Ag/AgCl as reference electrode and each measurement was calibrated with an internal

Table 1. UV-Vis Absorption Maxima and Photoluminescence Peak Wavelength of 1,3,4-oxadiazole-triazolopyridinone derivatives **5a**–**5i**

Compound			λ_{max} /nm of UV-Vis	λ_{max} /nm of PL	Φ_f [a]
	R^1	R^2	CH_2Cl_2	$CHCl_3$	
4a	H	-	314	425	-
4b	Me	-	310	425	-
4c	Cl	-	312	410	-
5a	H	H	314	479	0.76
5b	H	Me	308	476	0.71
5c	H	OEt	302	470	0.72
5d	Me	H	316	480	0.71
5e	Me	Me	318	486	0.69
5f	Me	OEt	304	486	0.73
5g	Cl	H	304	484	0.66
5h	Cl	Me	301	482	0.65
5i	Cl	OEt	300	486	0.71

[a] Φ_f: Fluorescence quantum efficiency, relative to 2-phenyl-5-(4-biphenyl)-1,3,4-oxadiazole in benzene (Φ_f = 0.8).

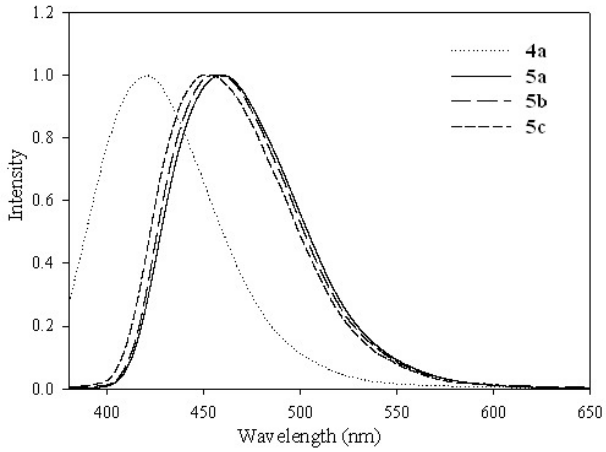


Figure 1. Normalized Photoluminescence Spectra of **4a**, **5a**, **5b**, and **5c** in $CHCl_3$ solution.

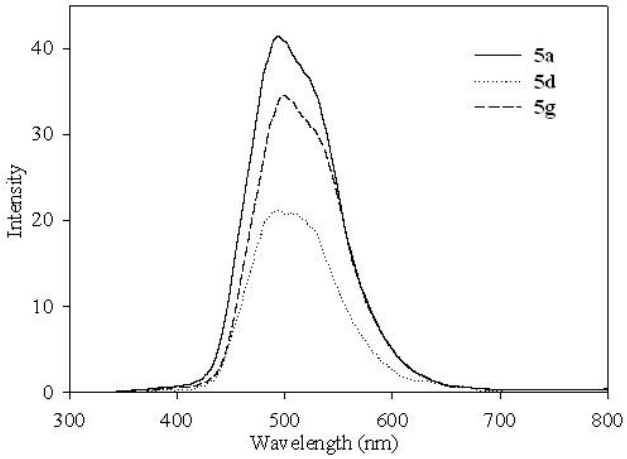


Figure 2. Normalized Photoluminescence Spectra of **5a**, **5d**, and **5g** (Vacuum evaporated Film).

standard, ferrocene/ferrocenium (Fc) redox system [19-20]. The data are summarized in Table 2 and the HOMO energy (*Ip*) for 1,3,4-oxadiazole-triazolopyridinone derivatives **5a**–**5i** were calculated based on the value of –4.8 eV for Fc with respect to zero vacuum level [19]. Upon the anodic sweep, **5a**–**5i** showed irreversible reduction processes. As an example, the cyclic voltammogram of **5e** is shown in Figure 3. In the case of **5e**, the reversibility of oxidation was estimated and the HOMO value is –5.35 eV with respect to Ag/AgCl. The bandgap energies (3.56–3.69 eV) of 1,3,4-oxadiazole-triazolopyridinone derivatives **5a**–**5i** were estimated from the onset wavelength (λ_{onset}) of the UV-vis absorption. The substitution effects on R^1 and R^2 positions do not promote the electronic properties characterization of 1,3,4-oxadiazole-triazolopyridinone derivatives **5a**–**5i**. From the high electron affinities, **5a**–**5i** possess the potential of electron-transporting and highly efficient blue-greenish photoluminescent materials.

Table 2. Electrochemical properties of 1,3,4-oxadiazole-triazolopyridinone derivatives **5a**–**5i**

Compound	*E*onset[a] (v)	*E'*onset[b] (v)	*Ip*[c,d] = E_{HOMO} (eV)	*Eg*[d,f] = Bandgap energy (eV)	*Ea*[d,f] = E_{LUMO} (eV)
5a	1.00	0.82	–5.62	3.66	–1.91
5b	1.00	0.82	–5.62	3.69	–1.93
5c	1.00	0.81	–5.51	3.65	–1.86
5d	0.75	0.57	–5.37	3.58	–1.79
5e	1.22	1.04	–5.35	3.57	–1.78
5f	0.80	0.62	–5.42	3.56	–1.86
5g	0.67	0.49	–5.39	3.57	–1.82
5h	0.63	0.45	–5.35	3.58	–1.77
5i	0.60	0.42	–5.32	3.57	–1.75

[a] Measured vs. ferrocene/ferrocenium. [b] *E'*onset = *E*onset – 0.19 eV (Measured vs. Ag/AgCl). [c] *Ip* = - (*E'*onset + 4.8), [d] *Eg*: the bandgap energy estimated from the onset wavelength of UV-vis absorption. [e] *Ea* = *Ip* + *Eg*. [f] *1 eV = 96.5 kJ/mol*

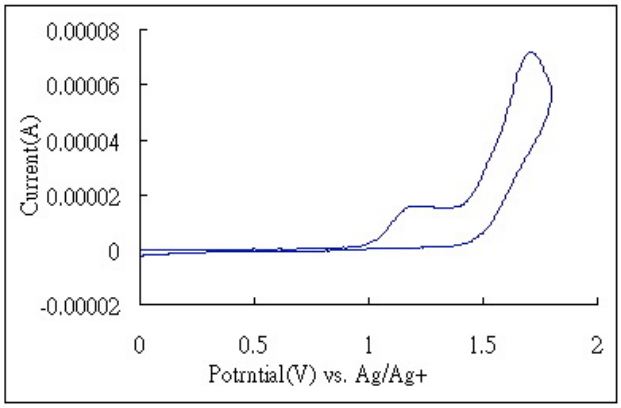


Figure 3. Cyclic voltammogram of **5e** in CH_2Cl_2 containing 0.1 M $TBAPF_6$ at a scan rate of 0.01 eV/s.

We successfully prepared a series of 1,3,4-oxadiazole-triazolopyridinone hybrid compounds as the blue electroluminescent materials by using 1,3,4-oxadiazole-pyridine derivatives with α-chloroformylarylhydrazine hydro-

chloride. Triazolopyridinone moiety plays an excellent assistant role in controlling fundamental photolytic process

EXPERIMENTAL

General. Nicotinohydrazide [14] and α-chloroformylaryl-hydrazine hydrochlorides [21] were synthesized according to literature procedures. All chemicals were reagent grade and used as purchased. All reactions were carried out under nitrogen atmosphere and monitored by TLC analysis. Flash column chromatography was carried out on silica gel (230-400 mesh). Commercially available reagents were used without further purification unless otherwise noted. Ethyl acetate, dimethyl sulfoxide, diisopropyl ether, hexanes, glacid acetic acid, and methanol were purchased from Mallinckrodt Chemical Co. Tetrahydrofuran (reagent grade) was purchased from Aldrich. The following compounds were purchased from Acoros Chemical Co: Benzoyl chloride, *p*-toluoly chloride, 4-chloro-benzoyl chloride, sodium hydrogen carbonate, sodium hydroxide and tributyl amine.

Analytical thin-layer chromatography (TLC) was performed on precoated plates (silica gel 60 F-254), purchased from Merck Inc. Mixtures of ethyl acetate and hexanes were used as eluants. Purification by gravity column chromatography was carried out by use of Merck Reagents Silica Gel 60 (particle size 0.063–0.200 mm, 70–230 mesh ASTM). Infrared (IR) spectra were measured on a Bomem Michelson Series FT-IR spectrometer. The wavenumbers reported are referenced to the polystyrene 1601 cm^{-1} absorption. Absorption intensities are recorded by the following abbreviations: s, strong; m, medium; w, weak. UV-visible spectra were measured with an HP 8452A diode-array spectrphotometer. Photolumimesence (PL) spectra were obtained on a Perkin-Alemer fluorescence spectrophotometer (LS 55). Proton NMR spectra were obtained on a Varian Unity-400 (400 MHz) or a Bruker-300 (400 MHz) spectrometer by use of chloroform-*d* as solvent. Elemental analyses were carried out on a Heraeus CHN–O RAPID element analyzer.

Cyclic Voltammetry Measurements: Cyclic voltammetry (CV) and differential pulse voltammetry (DPV) measurements were performed on a PGSTAT 20 electrochemical analyzer. The oxidation and reduction measurements were carried out, respectively, in anhydrous CH_2Cl_2 and anhydrous THF containing 0.1 *M* tetrabutylammonium hexafluorophosphate ($TBAPF_6$) as the supporting electrolyte at a scan rate of 50 mV s^{-1}. The potentials were measured against an Ag/Ag^+ (0.01 *M* AgCl) reference electrode using ferrocene as the internal standard. The onset potentials were determined from the intersection of two tangents drawn at the rising current and background current of the cyclic voltammogram [19].

Standard Procedure for Acylation [16]. A solution of nicotinohydrazide and pyridine was mixed and stirred in CH_2Cl_2 solution at room temperature. Benzoyl chloride was added dropwise into the reaction mixture and stirred at room temperature for 3 h. After the reaction was completed, the reaction mixture was filtered and washed with CH_2Cl_2. The filtrate solid was dried in vacuum oven for overnight and crystallized from CH_2Cl_2 to give the desired product (**3a**–**3c**).

Isonicotinic acid hydrazide (3a). A solution of isonicotinohydrazide (**2**, 2.81 g, 20.5 mmole, 1.0 equiv.) and pyridine (142 mg, 22.6 mmol, 1.1 equiv.) was mixed and stirred in CH_2Cl_2 (50.0 mL) solution at room temperature. Benzoyl chloride (175 mg, 22.8 mmol, 1.1 equiv.) was added dropwise into the reaction mixture and stirred at room temperature for 3 h. After the reaction was completed, the reaction mixture was filtered and washed with CH_2Cl_2 (150 mL). The filtrate solid was dried in vacuum oven for overnight and crystallized from CH_2Cl_2 to give the pure **3a** as white powder in 80% yield (3.95 g, 16.3 mmol): ^{1}H NMR (DMSO-d6, 200 MHz) δ 7.46–7.65 (m, 3H, ArH), 7.72–7.95 (m, 4H, ArH), 8.82 (d, *J* = 4.6 Hz, 2H), 10.50 (s, 1H, NH), 10.64 (s, 1H, NH).

4-Methylbenzoic acid(pyridine-4-carbonyl)hydrazide (3b). The standard procedure was followed and the desired product **3b** was obtained as white powder in 84% yield: ^{1}H NMR (DMSO-d6, 200 MHz) δ 2.37 (s, 3H, CH_3), 7.32 (d, *J* = 8.1 Hz, 2H, ArH), 7.78–7.85 (m, 4H, ArH), 8.79 (d, *J* = 5.9 Hz, 2H, ArH), 10.55 (s, 1H, NH), 10.83 (s , 1H, NH)

4-Chlorobenzoic acid(pyridine-4-carbonyl)hydrazide (3c). The standard procedure was followed and the desired product **3c** was obtained as white powder in 76% yield: ^{1}H NMR (DMSO-d6, 200 MHz) δ 7.59 (d, *J* = 8.5 Hz, 2H, ArH), 7.76–7.96 (m, 4H, ArH), 8.85 (d, *J* = 4.9 Hz, 2H, ArH), 10.76 (s, 1H, NH), 10.97 (s, 1H, NH).

Standard Procedure for Dehydroxyl-cyclolization [16]. A solution of *N*'-benzoylisonicotinohydrazide compounds **3a**–**3c** in $POCl_3$ (15 mL) was stirred at 100 °C for 2–4 h. After the reaction was complete, the reaction mixture was added to cold water (50 mL) and neutralized with NaOH aqueous solution (50 mL) to form precipitate. The product was washed with cold water, collected by filtration and dried in a vacuum oven overnight to give the desired product (**4a**–**4c**).

4-(5-Phenyl-1,3,4-oxadiazol-2-yl)pyridine (4a). A solution of *N*'-benzoylisonicotinohydrazide (**3a**, 3.26 g, 13.5 mmole, 1.0 equiv.) in $POCl_3$ (15 mL) was stirred at 100 °C for 2–4 h. After the reaction was completed, the reaction mixture was added to cold water (50 mL) and neutralized with NaOH aqueous solution (50 mL) to precipitate. The product was washed with cold water, collected by filtration and dried in vacuum oven overnight to obtain a pure **4a** (2.02 g, 9.03 mmol) as white solid in 67% isolated yield: ^{1}H NMR (DMSO-d6, 200 MHz) δ 7.63–7.68 (m, 3H, ArH), 8.04–8.08 (m, 2H, ArH), 8.13–8.18 (m, 2H, ArH), 8.85 (d, *J* = 4.5 Hz, 2H, ArH); FABMS *m/z* (relative intensity) 224 (M+1, 31), 223 (M, 54), 77 (100). *Anal*. Calcd for $C_{13}H_9N_3O$: C, 69.95; H, 4.06; N, 18.82. Found: C, 69.99; H, 4.03; N, 18.82.

4-(5-(4-Methylpheny)l-1,3,4-oxadiazol-2-yl)pyridine (4b). The standard procedure was followed and the desired product **4b** was obtained as white powder in 72% yield: ^{1}H NMR (DMSO-d6, 200 MHz) δ 2.40 (s, 3H, CH_3), 7.44 (d, *J* = 8.1 Hz, 2H, ArH), 8.02–8.06 (m, 4H, ArH), 8.85 (d, *J* = 5.9 Hz, 2H, ArH); FABMS *m/z* (relative intensity) 238 (M+1, 24), 237 (M, 44). *Anal*. Calcd for $C_{14}H_{11}N_3O$: C, 70.87; H, 4.67; N, 17.71. Found: C, 70.89; H, 4.66; N, 17.74.

4-(5-(4-Chloropheny)l-1,3,4-oxadiazol-2-yl)pyridine (4c). The standard procedure was followed and the desired product **4b** was obtained as white powder in 70% yield: ^{1}H NMR (DMSO-d6, 200 MHz) δ 7.70 (d, *J* = 7.9 Hz, 2H, ArH), 8.05–8.17 (m, 4H, ArH), 8.87 (d, *J* = 5.8 Hz, 2H , ArH) ; FABMS *m/z* (relative intensity) 258 (M+1, 31), 257 (M, 93).*Anal*. Calcd for $C_{13}H_8ClN_3O$: C, 60.60; H, 3.13; N, 16.31. Found: C, 60.59; H, 3.16; N, 16.36.

Standard Procedure for Arcycloaddition [16]. A solution of 3-phenyl-1,3,4-oxadiazolpyridine (**4a**–**4c**) was stirred in *i*-PrOH (15 mL) and triethylamine (1.0 mL) solution. The reaction mixture was heated up to 80 °C. α-Chloroformylaryl-hydrazine hydrochloride (0.36 g, 1.82 mmole, 1.1 equiv) was

added into the reaction mixture. After the reaction was completed, hot-filtration was performed and the material washed with cold ethanol (10 mL) to isolate the solid crude product. The crude product was dried and crystallized from CH_2Cl_2 to give pure **5a**–**5i** as light yellow solids in 45–66% yields.

2-Phenyl-7-(5-phenyl-1,3,4-oxadiazol-2-yl)-[1,2,4]triazolo-[4,3-*a*]pyridin-3(2*H*)-one (5a). A solution of 4-(5-phenyl-1,3,4-oxadiazol-2-yl)pyridine (**4a**, 0.36 g, 1.61 mmole, 1.0 equiv) was stirred in *i*-PrOH (15 mL) and triethylamine (1.0 mL) solution. The reaction mixture was heated up to 80 °C. α-Chloroformylaryl-hydrazine hydrochloride (0.35 g, 1.76 mmole, 1.1 equiv) was added into the reaction mixture. After the reaction was completed, hot-filtration was performed and the material was washed with cold ethanol (10 mL) to isolate the solid crude product. The crude product was dried and crystallized from $CHCl_3$ to give pure **5a** as a light yellow solid in 50% yield (313 mg, 885 mmol): mp 257–259 °C ($CHCl_3$); ^{1}H NMR (DMSO-d6, 200 MHz) δ 7.20–7.33 (m, 2H, ArH), 7.37–7.47 (m, 5H, ArH), 8.05–8.27 (m, 6H, ArH); IR (KBr) 1710 (m, C=O) cm^{-1}; FABMS *m/z* (relative intensity) 356 (M+1, 20), 355 (M, 39), 77 (100). *Anal.* Calcd for $C_{20}H_{13}N_5O_2$: C, 67.60; H, 3.69; N, 19.71. Found: C, 67.63; H, 3.70; N, 19.68.

2-(4-Methylphenyl)-7-(5-phenyl-1,3,4-oxadiazol-2-yl)-[1,2,4]-triazolo[4,3-*a*]pyridin-3(2*H*)-one (5b). The standard procedure was followed and the desired product **5b** was obtained as a light yellow in 53% yield: mp 250–252 °C ($CHCl_3$); ^{1}H NMR (DMSO-d6, 200 MHz) δ 2.22 (s, 3H, CH_3), 7.06–7.15 (m, 4H, ArH), 7.43–7.62 (m, 6H, ArH), 8.03–8.09 (m, 2H, ArH); IR (KBr) 1711 (m, C=O) cm^{-1}; FABMS *m/z* (relative intensity) 370 (M+1, 31), 369 (M, 83), 91 (40). *Anal.* Calcd for $C_{21}H_{15}N_5O_2$: C, 68.28; H, 4.09; N, 18.96. Found: C, 68.31; H, 4.11; N, 18.98.

2-(4-Methoxyphenyl)-7-(5-phenyl-1,3,4-oxadiazol-2-yl)-[1,2,4]triazolo[4,3-a]pyridin-3(2*H*)-one (5c). The standard procedure was followed and the desired product **5c** was obtained as a light yellow solid in 45% yield: mp 253–255 °C ($CHCl_3$); ^{1}H NMR (DMSO-d6, 200 MHz) δ 3.80 (s, 3H, CH_3) , 7.11 (d, *J* = 8.8 Hz, 2H, ArH), 7.21 (d, *J* = 7.4 Hz, 1H, ArH), 7.64–7.68 (m, 3H, ArH), 7.95 (d, *J* = 8.8 Hz, 2H, ArH), 8.11–8.23 (m, 4H, ArH); IR (KBr) 1709 (m, C=O) cm^{-1}; FABMS *m/z* (relative intensity) 386 (M+1, 31), 385 (M, 55). *Anal.* Calcd for $C_{21}H_{15}N_5O_3$: C, 65.45; H, 3.92; N, 18.17. Found: C, 65.41; H, 3.95; N, 18.22.

2-Phenyl-7-(5-(4-methylphenyl)-1,3,4-oxadiazol-2-yl)-[1,2,4]-triazolo[4,3-a]pyridin-3(2*H*)-one (5d). The standard procedure was followed and the desired product **5d** was obtained as a light yellow solid in 60% yield: mp 255–257 °C ($CHCl_3$); ^{1}H NMR (DMSO-d6, 200 MHz) δ 2.31 (s, 3H, CH_3), 7.31–7.53 (m, 7H, ArH), 7.92–8.16 (m, 5H, ArH); IR (KBr) 1715 (m, C=O) cm^{-1}; FABMS *m/z* (relative intensity) 370 (M+1, 28), 292 (M, 79). *Anal.* Calcd for $C_{21}H_{15}N_5O_2$: C, 68.28; H, 4.09; N, 18.96. Found: C, 68.31; H, 4.11; N, 18.97.

2-(4-Methylphenyl)-7-(5-(4-methylphenyl)-1,3,4-oxadiazol-2-yl)-[1,2,4]triazolo[4,3-a]pyridin-3(2*H*)-one (5e). The standard procedure was followed and the desired product **5e** was obtained as a light yellow solid in 57% yield: mp 241–243 °C ($CHCl_3$); ^{1}H NMR (DMSO-d6, 200 MHz) δ 2.27 (s, 3H, CH_3), 2.41 (s, 3H, CH_3), 7.08–7.20 (m, 5H, ArH), 7.43 (d, *J* = 8.2 Hz, 2H, ArH), 7.56 (d, *J* = 8.5 Hz, 2H, ArH), 8.01 (d, *J* = 8.2 Hz, 2H, ArH); IR (KBr) 1715 (m, C=O) cm^{-1}; FABMS *m/z* (relative intensity) 384 (M+1, 19), 292 (M, 43). *Anal.* Calcd for $C_{22}H_{17}N_5O_2$: C, 68.92; H, 4.47; N, 18.27. Found: C, 68.91; H, 4.43; N, 18.25.

2-(4-Ethoxyphenyl)-7-(5-(4-methylphenyl)-1,3,4-oxadiazol-2-yl)-[1,2,4]triazolo[4,3-a]pyridin-3(2*H*)-one (5f). The standard procedure was followed and the desired product **5f** was obtained as a light yellow solid in 50% yield: mp 236–238 °C ($CHCl_3$); ^{1}H NMR (DMSO-d6, 200 MHz) δ 2.07 (s, 3H, CH_3), 3.80 (s, 3H, CH_3), 7.09 (d, *J* = 8.8 Hz, 2H, ArH), 7.22 (d, *J* = 7.3 Hz, 1H, ArH), 7.63–7.67 (m, 3H, ArH), 7.95 (d, *J* = 8.8 Hz, 2H, ArH), 8.11–8.24 (m, 3H, ArH); IR (KBr) 1718 (m, C=O) cm^{-1}; FABMS *m/z* (relative intensity) 400 (M+1, 22), 399 (M, 43). *Anal.* Calcd for $C_{22}H_{17}N_5O_3$: C, 66.16; H, 4.29; N, 17.53. Found: C, 66.12; H, 4.31; N, 17.51.

2-Phenyl-7-(5-(4-chlorophenyl)-1,3,4-oxadiazol-2-yl)-[1,2,4]-triazolo[4,3-a]pyridin-3(2*H*)-one (5g). The standard procedure was followed and the desired product **5g** was obtained as a light yellow solid in 66% yield: mp 263–265 °C ($CHCl_3$); ^{1}H NMR (DMSO-d6, 200 MHz) δ 7.51–7.78 (m, 7H, ArH), 8.05–8.25 (m, 5H, ArH); IR (KBr) 1725 (m, C=O) cm^{-1}; FABMS *m/z* (relative intensity) 390 (M+1, 34), 389 (M, 69). *Anal.* Calcd for $C_{20}H_{12}ClN_5O_2$: C, 61.63; H, 3.10; N, 17.97. Found: C, 61.60; H, 3.12; N, 17.98.

2-(4-Methylphenyl)-7-(5-(4-chlorophenyl)-1,3,4-oxadiazol-2-yl)-[1,2,4]triazolo[4,3-a]pyridin-3(2*H*)-one (5h). The standard procedure was followed and the desired product **5h** was obtained as a light yellow solid in 65% yield: mp 238–239 °C ($CHCl_3$); ^{1}H NMR (DMSO-d6, 200 MHz) δ 2.27 (s, 3H, CH_3), 7.08–7.20 (m, 3H, ArH), 7.56 (d, *J* = 8.6 Hz, 2H, ArH), 7.68–7.72 (m, 4H, ArH), 8.13 (d, *J* = 8.6 Hz, 2H, ArH); IR (KBr) 1726 (m, C=O) cm^{-1}; FABMS *m/z* (relative intensity) 404 (M+1, 32), 403 (M, 72). *Anal.* Calcd for $C_{21}H_{14}ClN_5O_2$: C, 62.46; H, 3.49; N, 17.34. Found: C, 62.50; H, 3.47; N, 17.30.

2-(4-Ethoxyphenyl)-7-(5-(4-chlorophenyl)-1,3,4-oxadiazol-2-yl)-[1,2,4]triazolo[4,3-a]pyridin-3(2*H*)-one (5i). The standard procedure was followed and the desired product **5i** was obtained as a light yellow solid in 63% yield: mp 223–225 °C ($CHCl_3$); ^{1}H NMR (DMSO-d6, 200 MHz) δ 3.85 (s, 3H, CH_3), 7.01 (d, *J* = 8.9 Hz, 2H, ArH), 7.28 (d, *J* = 7.2 Hz, 2H, ArH), 7.56–7.59 (m, 3H, ArH), 7.97–8.16 (m, 5H, ArH); IR (KBr) 1726 (m, C=O) cm^{-1}; FABMS *m/z* (relative intensity) 420 (M+1, 42), 419 (M, 89), 179 (36). *Anal.* Calcd for $C_{21}H_{14}N_5O_3$: C, 60.08; H, 3.36; N, 16.68. Found: C, 60.11; H, 3.33; N, 16.69.

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