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Solid state PhI(OAc)$_2$-Al$_2$O$_3$ Mediated Efficient Synthesis and Characterization of series 2-[1, 8] naphthyridin-3-yl)-5-(substituted -(thiophen-2-yl)--1,3,4-oxadiazoles

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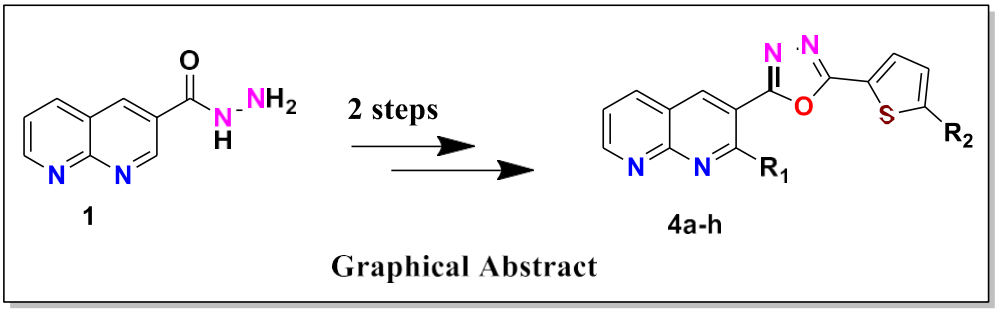
Abstract

Reaction in between 1,8-naphthyridine-3-carbohydrazide **1** and thiophene-2-carbaldehyde **2** with *p*-TSA was ground by pestle and mortar to give hydrazones **3.** Furthermore, hydrazones **3** on oxidative cyclization with PhI(OAc)$_2$-Al$_2$O$_3$ in the solid state at RT under grinding conditions afforded the respective 2-(2-substituted [1,8]-naphthyridin-3-yl)-5-(thiophen-2-yl)-1,3,4-oxadiazoles **4a-h** in good yields (**Scheme I**).

Keywords: 1,8-Naphthyridines, *p*-TSA, solid PhI(OAc)$_2$-Al$_2$O$_3$



Graphical Abstract

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Introduction

Pyridine-like analog to naphthalene, the first naphthyridine derivative was synthesized and named by Arnold Reassert[1-3]. The name "naphthyridine" was exclusively designated to the fused-ring system resulting from the fusion of twopyridine rings through two adjacent carbon atoms, with each ring containing only one nitrogen atom. Also known with other names, such as diazanaphthalenes or pyridopyridines, "naphthyridine" remains the most commonly used name for this classof compounds. Various review articles summarize the synthesis, structure, physicochemical properties, and pharmacological role of naphthyridines [4–10]. Six different isomeric forms of naphthyridines are described based on the position of the nitrogens in the bicyclicsystem (Shown in Fig I & II).

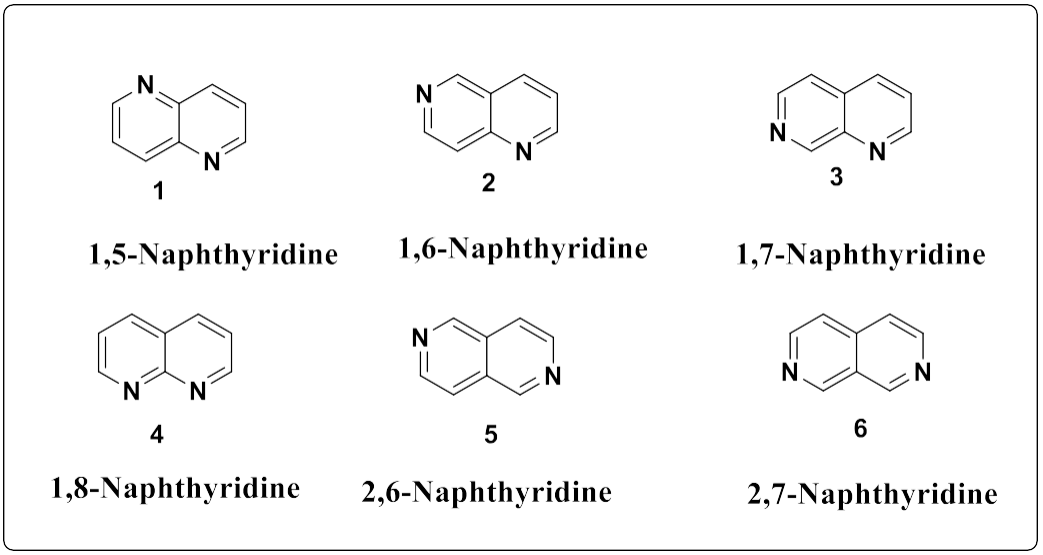


Fig. I Structures of isomers of Naphthyridines

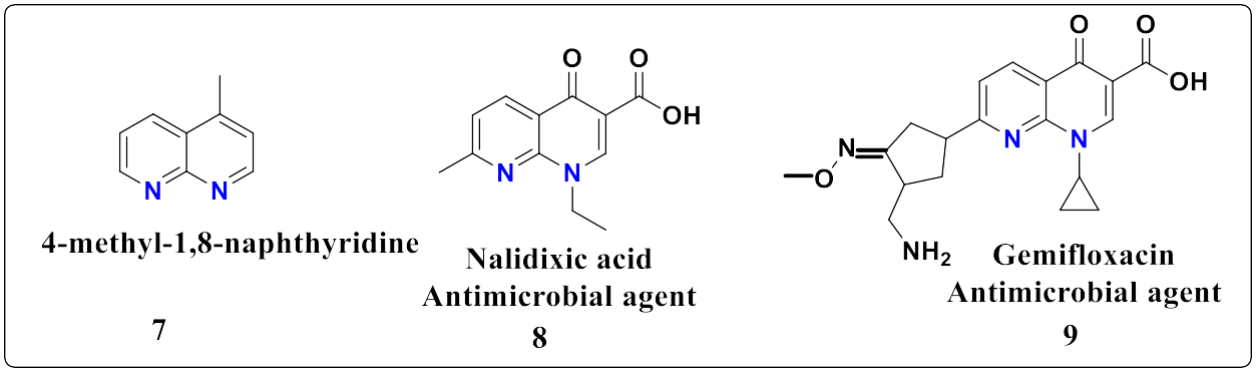


Fig. II Biologically active 1,8-naphthyridines

Herein, we have synthesized the Solid state PhI(OAc)$_2$-Al$_2$O$_3$ mediated synthesis and characterization of new series2-[1,8] naphthyridin-3-yl)-5-(substituted -(thiophen-2-yl)--1,3,4-oxadiazoles in accordingly systematic protocols reference cited. [11–24]

Experimental section

Melting points were determined using a Cintex melting point apparatus and are uncorrected. TLC was performed by using Merck silica gel 60F254 precoated plates (0.25 mm) and column chromatography was performed by using Silica gel (particle size 100-200 mesh). Proton nuclear Magnetic Resonance (400 MHz) and Carbon Nuclear Magnetic Resonance (100 MHz)

spectrums were logged on Bruker AC-300spectrophotometer in $CHCl_3$with *TMS* asreference. Mass spectrum was documented on JEOL SX-102 spectrophotometer. All the chemicals and reagents used in present investigation were purchased from Sigma- Aldrich Chemical Company.

I General procedure for the Synthesis of 2-Methyl-*N'*-(thiophen-2-ylmethylene)-1,8-naphthyridine-3-carbohydrazides 3

A mixture of 1,8-naphthyridine-3-carbohydrazide **1** (0.01 mole), thiophene-2-carbaldehyde **2** (0.01 mole) and *p*-TSA (0.015 mole) was ground by pestle and mortar at RT. On completion of the reaction (monitored by TLC), the reaction-mixture was treated with ice-cold water. The solid thus obtained was filtered, washed with water and purified by recrystallization from ethanol to give **3.**

II General procedure for the synthesis 2-(2-methyl-1,8- naphthyridin-3-yl)-5-(thiophen-2-yl)-1,3,4-oxadiazoles 4

A mixture of a finely powdered appropriate hydrazone **3** (0.01 mole) and PhI(OAc)$_2$-Al$_2$O$_3$ (0.01 mole) was ground in a mortar by pestle at RT for specified time. After completion of the reaction (indicated by TLC), the reaction mixture was treated with cold water. The resultant product was filtered, washed with water and purified byrecrystallization from ethanol to furnish **4**.

Preparation of alumina-supported iodobenzene diacetate [PhI(OAc)$_2$- Al$_2$O$_3$][25]

Iodobenzene diacetate (0.01 mole) per gram of neutral alumina is ground using a pestle and mortar; the recovered alumina after removal of the products is reused without any loss of activity.

Results and discussions

I Synthesis of *N'*-(thiophen-2-ylmethylene)-1,8-naphthyridine-3- carbohydrazides 3

Condensation of 2-substituted-1,8-naphthyridine-3-carboxylic acid hydrazides**1** with thiophene-2-carbaldehyde **2** in the presence of catalytic amount of PTSA insolvent-free grinding conditions at RT furnished the corresponding *N'*-(thiophen-2-ylmethylene)-1,8-naphthyridine-3- carbohydrazide **3** in excellent yields.

In a typical case, an equimolar mixture of 2-methyl-1,8-naphthyridine-3-carboxylic acid hydrazide **1a** (R^1=CH_3)**,**2-furaldehyde **2** (R^2=H) and PTSA wasground in mortar by pestle at RT for 2.0min. Aftercompletion of the reaction(monitored by TLC) the reaction mixturewas treated with ice-cold water. After usualwork-up 2-methyl-*N'*- (thiophen-2-ylmethylene)-1,8-naphthyridine-3- carbohydrazide **3a** (R^1=CH_3; R^2=H) was obtained in 94% yield.

The above condensation reaction was found to be general one and proceeded smoothly with thiophene-2-carbaldehydes giving the respective*N*'3-[1-(substituted 2-furyl) methylidene]- 2-sustituted [1,8] naphthyridine-3 carbohydrazides **3b-h** [(R^1=CH_3; R^2= NO_2),(R^1=CF_3; R^2= H), (R^1=CF_3; R^2= NO_2), (R^1=C_6H_5; R^2= H), (R^1=C_6H_5; R^2= NO_2), (R^1=4-$NO_2C_6H_4$; R^2= H), (R^1=4-$NO_2C_6H_4$;R^2= NO_2)].

II Synthesis of 2-(2-substituted [1, 8]-naphthyridin-3-yl)-5-(thiophen-2-yl)-1,3,4-oxadiazoles 4

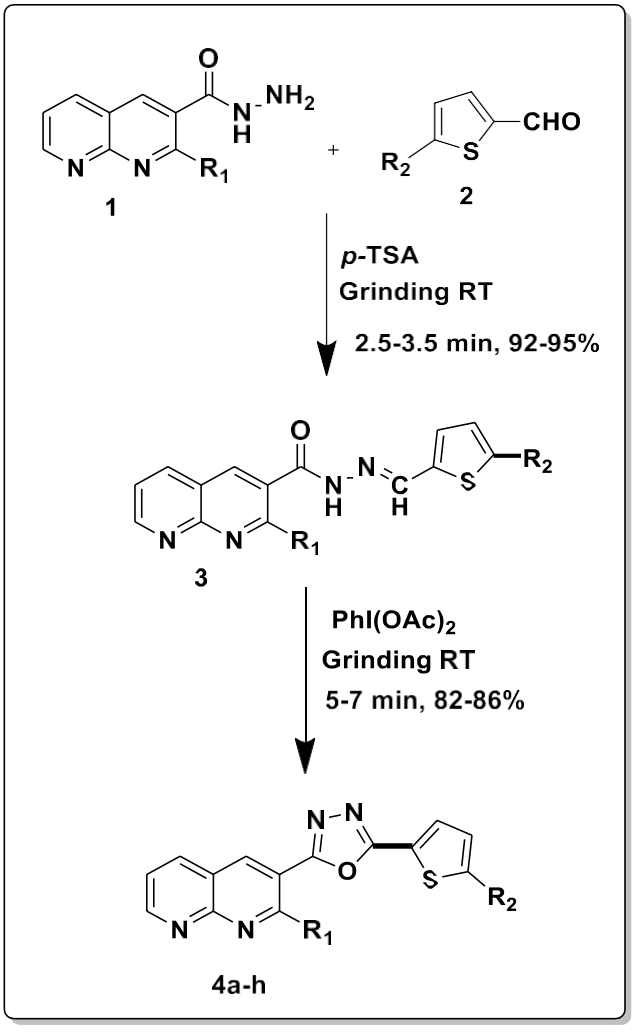
The hydrazones **3** on oxidative cyclization with PhI(OAc)$_2$-Al$_2$O$_3$ in the solid state at RT under grinding conditions afforded the respective 2-(2-substituted [1,8]- naphthyridin-3-yl)-5-(thiophen-2-yl)-1,3,4- oxadiazoles **4** in good yields (**Scheme I**). Reactions are not consuming and the yieldsof the products are good. The reaction conditions and work-up procedures are

mild, simple, convenient and efficient. The products were obtained with high purity by this procedure. The process isenvironmentally benign. The experimental procedure is very simple and avoids sophistication.

In a typical case, a mixture of 2-methyl-*N*'-(thiophen-2-ylmethylene)-1,8- naphthyridine-3-carbohydrazide **3a** (R^1=CH_3; R^2=H) and PhI(OAc)$_2$-Al_2O_3 was ground in a mortar by pestle at RT for 5-7 min.
On completion of the reaction (indicated by TLC), the reaction mixture istreated with cold water followed by simple processing afforded 2methyl-(2- substituted [1,8]-naphthyridin-3-yl)-5-(thiophen-2-yl)-1,3,4-oxadiazole **4a** (R^1=CH_3; R^2=H) in 86% yield.

The generality of the facile oxidative transformation was established by treating other hydrazones 3b-h with PhI(OAc)$_2$- Al_2O_3 under solid state grinding conditionsto get the corresponding 2-(2-substituted [1,8]-naphthyridin-3-yl)-5-(thiophen-2-yl)-1,3,4-oxadiazoles **4b-h** in 82-85% yields.

The structures of compounds **4a-h** wereestablished on the basis of their elemental analyses and spectral (IR, ^{1}H NMR ^{13}CNMR and MS) data. The simple operation, high purity of the products, good yields, mild reaction conditions and non-toxicity of the reagent are notable advantages of this protocol.



Scheme I

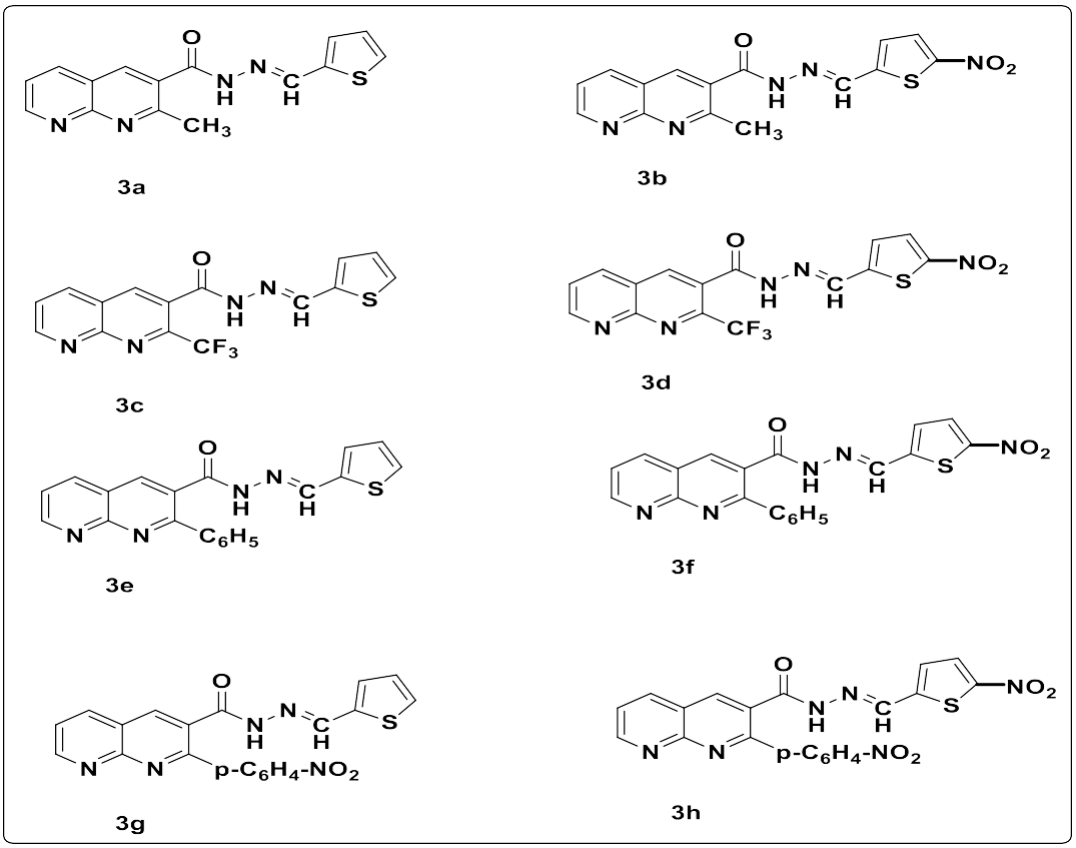


Fig. III Structures of compounds 3a-h

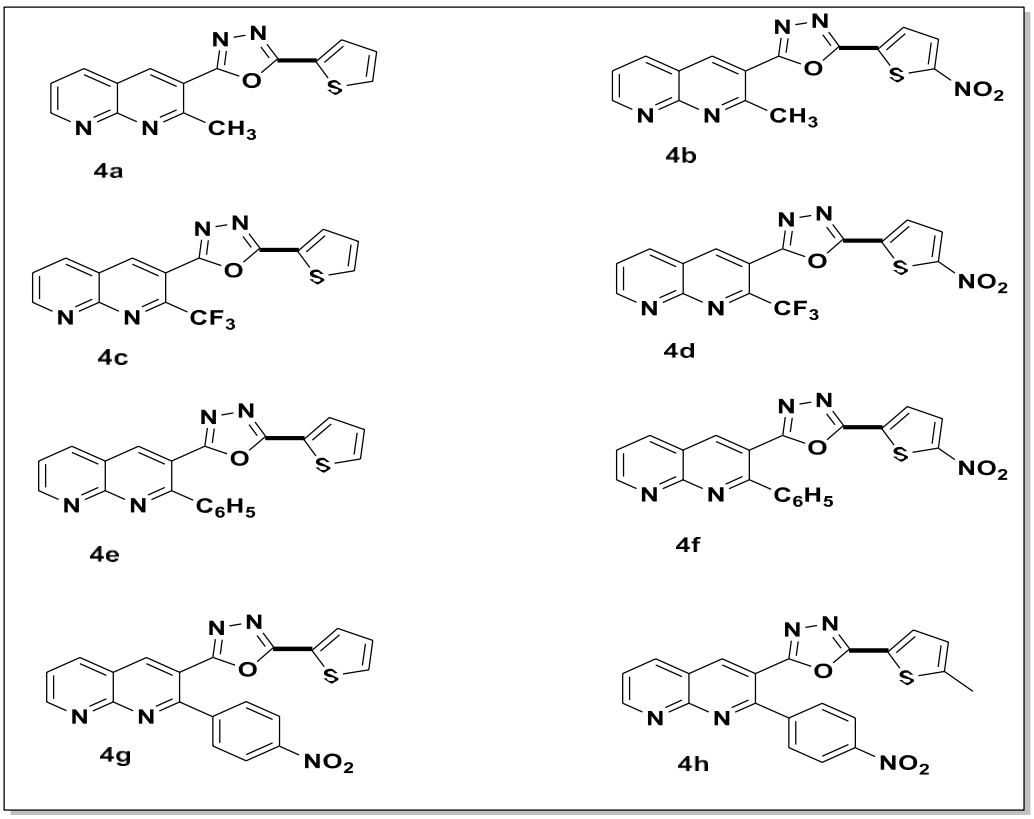


Fig. IV Structures of compounds 4a-h

Table I – IR and mass spectral data of 2-methyl-*N'*-(thiophen-2-ylmethylene)-1,8-naphthyridine-3-carbohydrazides (3a-f)

Entry	R^1	R^2	ν_{max} in cm^{-1}			MS (ESI) $[M+H]^+$ *m/z*
			NH	C=O	C=N	
3a	CH_3	H	3469	1660	1616	297
3b	CH_3	NO_2	3442	1675	1619	342
3c	CF_3	H	3471	1661	1618	351
3d	CF_3	NO_2	3448	1675	1617	396
3e	C_6H_5	H	3458	1655	1615	357
3f	C_6H_5	NO_2	3454	1699	1618	402
3g	4-$NO_2C_6H_4$	H	3452	1644	1620	402
3h	4-$NO_2C_6H_4$	NO_2	3427	1664	1617	416

Table II – ^{1}H NMR spectral data of 2-methyl-*N'*-(thiophen-2-ylmethylene)-1,8-naphthyridine-3-carbohydrazides (3a-f)

Entry	R^1	R^2	^{1}H NMR (300 MHz, $CDCl_3$) (δ, ppm)
3a	CH_3	H	2.78(s, 3H, CH_3), 8.16(m, 1H, C_6-H), 8.51 (m, 2H, C_4-H, C_5-H), 9.14 (m, 1H, C_7-H), 8.62(s, 1H, N=CH), 7.42-8.05 (m, 3H, C_3-H, C_4-H, C_5-H of furan), 12.55(s, 1H, NH).
3b	CH_3	NO_2	2.76(s, 3H, CH_3), 8.15(m, 1H, C_6-H), 8.43 (m, 2H, C_4-H, C_5-H), 9.06 (m, 1H, C_7-H), 8.57(s, 1H, N=CH), 7.58(d, 1H, C_3-H of furan), 7.90(d, 1H, C_4-H of furan), 12.12(s, 1H, NH).
3c	CF_3	H	7.78(m, 1H, C_6-H), 8.20 (m, 2H, C_4-H, C_5-H), 9.10 (m, 1H, C_7-H), 8.87(s, 1H, N=CH), 7.23-7.42 (m, 3H, C_3-H, C_4-H, C_5-H of furan), 12.43(s, 1H, NH).
3d	CF_3	NO_2	7.94 (m, 1H, C_6-H), 8.25 (m, 2H, C_4-H, C_5-H), 9.18 (m, 1H, C_7-H), 8.66(s, 1H, N=CH), 7.62(d, 1H, C_3-H of furan), 8.10(d, 1H, C_4-H of furan), 12.46 (s, 1H, NH).

3e	C_6H_5	H	7.83(m, 1H, C_6-H), 8.22 (m, 2H, C_4-H, C_5-H), 9.20 (m, 1H, C_7-H), 8.78(s, 1H, N=CH), 6.40-7.22 (m, 3H, C_3-H, C_4-H, C_5-H of furan), 7.48-7.80 (m, 5H, Ar-H), 12.25(s, 1H, NH).
3f	C_6H_5	NO_2	7.77 (m, 1H, C_6-H), 8.60 (m, 2H, C_4-H, C_5-H), 9.22 (m, 1H, C_7-H), 8.73(s, 1H, N=CH), 7.24(d, 1H, C_3-H of furan), 7.40(d, 1H, C_4-H of furan), 7.47-7.74(m, 5H, Ar-H), 12.42(s, 1H, NH).
3g	4-$NO_2C_6H_4$	H	7.86(m, 1H, C_6-H), 8.58 (m, 2H, C_4-H, C_5-H), 9.20 (m, 1H, C_7-H), 8.80(s, 1H, N=CH), 6.52-7.30 (m, 3H, C_3-H, C_4-H, C_5-H of furan), 7.72-7.84(m, 4H, Ar-H), 12.35(s, 1H, NH).
3h	4-$NO_2C_6H_4$	NO_2	7.92 (m, 1H, C_6-H), 8.30 (m, 2H, C_4-H, C_5-H), 9.15 (m, 1H, C_7-H), 8.82(s, 1H, N=CH), 7.20(d, 1H, C_3-H of furan), 7.62(d, 1H, C_4-H of furan), 7.68-7.87(m, 4H, Ar-H), 12.40(s, 1H, NH).

Table III – IR and mass spectral data of 2-(2-substituted [1,8]-naphthyridin-3-yl)-5-(thiophen-2-yl)-1,3,4-oxadiazoles 4a-h

Entry	R^1	R^2	ν_{max} in cm^{-1}	MS (ESI)
			C=N	$[M+H]^+$ *m/z*
4a	CH_3	H	1600	295
4b	CH_3	NO_2	1602	340
4c	CF_3	H	1603	349
4d	CF_3	NO_2	1601	394
4e	C_6H_5	H	1604	357
4f	C_6H_5	NO_2	1605	402
4g	4-$NO_2C_6H_4$	H	1603	402
4h	4-$NO_2C_6H_4$	NO_2	1601	416

Table IV – ^{1}H NMR spectral data of **2-(2-substituted [1,8]-naphthyridin-3-yl)-5-(thiophen-2-yl)-1,3,4-oxadiazoles 4a-h**

Entry	R^1	R^2	^{1}H NMR (300 MHz, $CDCl_3$) (δ, ppm)
4a	CH_3	H	3.10(s, 3H, CH_3), 8.02(m, 1H, C_6-H), 8.63 (m, 1H, C_5-H), 8.35 (s, 1H, C_4-H), 9.18 (m, 1H, C_7-H), 7.23-7.80 (m, 3H, C_3-H, C_4-H, C_5-H of furan).
4b	CH_3	NO_2	3.12(s, 3H, CH_3), 8.10(m, 1H, C_6-H), 8.70(m, 1H, C_5-H), 8.42 (s, 1H, C_4-H), 9.22 (m, 1H, C_7-H), 7.43 (d, 1H, C_3-H of furan), 8.02(d, 1H, C_4-H of furan).
4c	CF_3	H	8.12(m, 1H, C_6-H), 8.68 (m, 1H, C_5-H), 8.46 (s, 1H, C_4-H), 9.20(m, 1H, C_7-H), 7.27-7.82 (m, 3H, C_3-H, C_4-H, C_5-H of furan).
4d	CF_3	NO_2	7.97 (m, 1H, C_6-H), 8.82 (m, 1H, C_5-H), 8.95 (s, 1H, C_4-H), 9.20 (m, 1H, C_7-H), 7.40(d, 1H, C_3-H of furan), 7.97(d, 1H, C_4-H of furan).
4e	C_6H_5	H	8.06(m, 1H, C_6-H), 8.45 (m, 1H, C_5-H), 8.65 (s, 1H, C_4-H), 9.25 (m, 1H, C_7-H), 7.30-7.87 (m, 3H, C_3-H, C_4-H, C_5-H of furan), 7.52-7.95(m, 5H, Ar-H).
4f	C_6H_5	NO_2	8.10 (m, 1H, C_6-H), 8.65 (m, 1H, C_5-H), 8.72(s, 1H, C_4-H), 9.28 (m, 1H, C_7-H), 7.35(d, 1H, C_3-H of furan), 7.58 (d, 1H, C_4-H of furan), 7.50-7.84(m, 5H, Ar-H).
4g	4-$NO_2C_6H_4$	H	7.88(m, 1H, C_6-H), 8.72 (m, 1H, C_5-H), 8.98 (s, 1H, C_4-H), 9.23 (m, 1H, C_7-H), 7.40-7.60 (m, 3H, C_3-H, C_4-H, C_5-H of furan), 7.65-7.80 (m, 4H, Ar-H).
4h	4-$NO_2C_6H_4$	NO_2	8.30 (m, 1H, C_6-H), 8.78 (m, 1H, C_5-H), 9.08 (s, 1H, C_4-H), 9.24 (m, 1H, C_7-H), 7.17(d, 1H, C_3-H of furan), 7.65(d, 1H, C_4-H of furan), 7.70-7.90(m, 4H, Ar-H).

Table V — Physical and analytical data of ***N'*-(thiophen-2-ylmethylene)-1,8-naphthyridine-3-carbohydrazides 3a-h**

Entry	R^1	R^2	Reaction time (min)	m.p. ^oC	Yield (%)	Mol. formula	Found (%) (Calcd)		
							C	H	N
3a	CH_3	H	2.0	210	94	$C_{15}H_{12}N_4OS$	64.41 (64.28	4.34 4.32	20.03 19.99)
3b	CH_3	NO_2	1.5	253	93	$C_{15}H_{11}N_5O_3S$	55.51 (55.39	3.42 3.41	21.58 21.53)
3c	CF_3	H	2.5	230	95	$C_{15}H_9F_3N_4OS$	54.04 (53.90	2.73 2.71	16.80 16.76)
3d	CF_3	NO_2	2.0	238	92	$C_{15}H_8F_3N_5O_3S$	47.63 (47.51	2.14 2.13	18.52 18.47)
3e	C_6H_5	H	2.0	248	94	$C_{20}H_{14}N_4OS$	70.31 (70.17	4.14 4.12	16.41 16.37)
3f	C_6H_5	NO_2	1.5	174	92	$C_{20}H_{13}N_5O_3S$	62.15 (62.02	3.39 3.38	18.13 18.08)
3g	4-$NO_2C_6H_4$	H	2.5	283	93	$C_{20}H_{13}N_5O_3S$	62.16 (62.02	3.40 3.38	18.12 18.08)
3h	4-$NO_2C_6H_4$	NO_2	2.0	276	92	$C_{20}H_{12}N_6O_5S$	55.69 (55.56	2.81 2.80	19.50 19.44)

Table VI — Physical and analytical data of 2-(2-substituted [1,8]-naphthyridin-3-yl)-5-(thiophen-2-yl)-1,3,4-oxadiazoles 4a-h

Entry	R^1	R^2	Reaction time (min)	m.p. ^oC	Yield (%)	Mol. formula	Found (%) (Calcd)		
							C	H	N
4a	CH_3	H	6.5	300	86	$C_{15}H_{10}N_4OS$	64.82 (64.74	3.64 3.62	20.18 20.13)
4b	CH_3	NO_2	6.0	214	84	$C_{15}H_9N_5O_3S$	55.86 (55.73	2.83 2.81	21.70 21.66)
4c	CF_3	H	6.0	302	85	$C_{15}H_7F_3N_4OS$	54.36 (54.23	2.14 2.12	16.91 16.86)
4d	CF_3	NO_2	5.5	192	84	$C_{15}H_6F_3N_5O_3S$	47.90 (47.76	1.61 1.60	18.60 18.56)
4e	C_6H_5	H	6.5	263	85	$C_{20}H_{12}N_4OS$	70.71 (70.58	3.57 3.55	16.53 16.48)
4f	C_6H_5	NO_2	6.0	233	84	$C_{20}H_{11}N_5O_3S$	62.49 (62.34	2.90 2.88	18.22 18.17)
4g	4-$NO_2C_6H_4$	H	7.0	218	84	$C_{20}H_{11}N_5O_3S$	62.47 (62.34	2.89 2.88	18.23 18.17)
4h	4-$NO_2C_6H_4$	NO_2	6.5	257	82	$C_{21}H_{13}N_5O_3S$	55.94 (55.82	2.36 2.34	19.57 19.53)

Table VII— ^{13}C NMR spectral data of 2-(2-substituted [1,8]-naphthyridin-3-yl)-5-(thiophen-2-yl)-1,3,4-oxadiazoles 4a-h

Entry	^{13}C NMR chemical shift data (δ)
4a	160.28, 159.57, 156.60, 152.70, 150.17, 137.50, 131.98, 131.30, 130.53, 130.04, 127.40, 126.92, 119.88, 114.65, 23.54.
4b	160.53, 159.57, 156.60, 153.41, 152.70, 150.17, 137.23, 133.49, 131.98, 126.92, 121.22, 119.88, 114.65, 23.54.
4c	160.28, 156.79, 151.98, 141.92, 141.71, 141.50, 141.28, 137.28, 131.30, 130.57, 130.04, 129.18, 127.40, 126.46, 124.36, 122.27, 121.68, 120.17, 115.81.
4d	160.53, 156.79 , 153.41, 151.93, 141.71, 137.28, 133.49, 130.40 (s), 129.18, 126.46, 124.36, 122.27, 121.68, 121.22, 120.17, 115.81
4e	160.28, 156.75, 153.07, 152.41, 137.72, 137.28, 136.16, 131.30, 131.07, 130.53, 130.04, 128.75, 128.58, 127.40, 121.64, 115.81.
4f	160.53, 156.75, 153.41, 153.07, 152.41, 151.98, 137.72, 136.16, 133.49, 131.07, 128.75, 128.58, 121.68, 121.22, 115.81
4g	160.28, 156.75, 153.07, 152.41, 151.98, 149.98, 141.21, 137.28, 130.53, 127.40, 124.09, 121.64, 115.81.
4h	160.53, 156.75, 153.07, 151.98, 149.98, 143.32, 141.21, 137.72, 137.28, 136.47, 131.65, 130.85, 125.14, 124.09, 121.64, 115.81, 16.83.

Conclusion

Herein, we have reported an efficient synthesis of (2-substituted [1,8]-naphthyridin-3-yl)-5-(thiophen-2-yl)-1,3,4-oxadiazoles under Solid state mediated PhI(OAc)$_2$-Al$_2$O$_3$.

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