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Synthesis of 4-Thioquinazoline Compounds in Aqueous Media Catalyzed by Indium

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An efficient and facile reaction of 4-chloroquinazoline and thiols was achieved under indium mediation in water, providing a simple method for the synthesis of substituted 4-thioquinazoline compounds in good yield. The structures of the title compounds were characterized by elemental analyses as well as IR, ^{1}H and ^{13}C NMR spectroscopies. Preliminary biological tests showed that compound **3d** exhibited good antifungal activity against *Fusarium oxysporum* with EC_{50} = 17.08 μg/mL.

Key Words: Quinazoline, Green synthesis, Indium, Antifungal activity.

INTRODUCTION

In recent years, quinazoline compounds have been found to possess biological activities in medicine and pesticide[1-4]. As part of our ongoing research on heterocyclic compounds that may serve as leads for designing novel antifungal agents, we are particularly interested in 4-substituted thioquinazolines[5-9]. Indium-mediated organic reactions in aqueous media have become so attractive that several studies and reviews were published in the past several years[10-12]. In this paper, a series of 4-thioquinazoline compounds were prepared with a metal catalyst in water (Fig. 1). The structures of the 4-thioquinazoline compounds were characterized by elemental analyses as well as IR, ^{1}H and ^{13}C NMR spectroscopies. Preliminary biological tests showed that some of the compounds exhibited good antifungal activities.

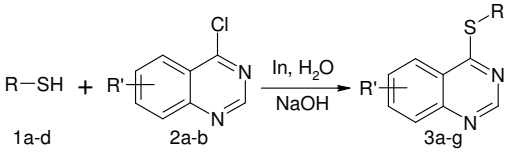


Fig. 1. Synthetic route of 4-thioquinazoline compounds

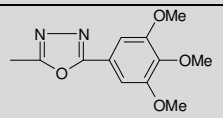
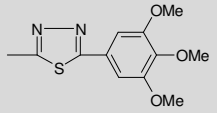
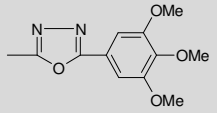
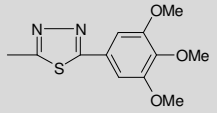
EXPERIMENTAL

Unless otherwise indicated, all common reagents and solvents were used as obtained from commercial supplies without further purification. All product melting points were determined on a XT-4 binocular microscope (Beijing Tech Instrument Co., China) and were not corrected. Infrared spectra were recorded on a Bruker VECTOR22 spectrometer in KBr disks. The ^{1}H and ^{13}C NMR spectra were recorded on a spectrometer Varian-Inova 500 MHz (500 and 125 MHz, respectively) at room temperature in DMSO-d_6 using TMS as an internal standard. Elemental analysis was performed by an Elementar Vario-III CHN analyzer. The following compounds were prepared as described in literature: 4-chloroquinazoline (**2a**): white needle crystal, yield 54.5 %, m.p. 94-95 ºC (lit.[13], m.p. 96.5-97.5 ºC); 4-chloro-6,7,8-trimethoxy quinazoline (**2b**): white solid, yield 62.5 %, m.p. 100-102 ºC (lit.[14], m.p. 101-103 ºC).

General procedure for the traditional method of preparing compounds 3a-3g (method A): Compounds **1a**-**1d** (3 mmol) and potassium carbonate (5.5 mmol) were added to absolute acetone (15 mL). The mixture was stirred at room temperature for 10 min and then compounds **2a** or **2b** (3 mmol) were slowly added. The reaction mixture was refluxed for 8 h and 80 mL of water was added to the reaction mixture. After filtering the mixture, the solid was washed with water to pH 7 and recrystallized from absolute ethanol to give the desired products **3a**-**3g**.

General procedure for the preparation of compounds 3a-3g without catalyst (method B): Compounds **1a**-**1d** (3 mmol) were added to water (15 mL) and sodium hydroxide (150 mg) was added. The mixture was stirred at room temperature for 10 min and then compounds **2a**-**2b** (3 mmol) was

TABLE-1
PHYSICAL DATA OF COMPOUNDS **3a-3g**

Compound	R	R'	Yield[a] (%)			m.p. (ºC)
			Method A[b]	Method B[c]	Method C[d]	
3a		H	13.5	9.6	65.3	184-185
3b		H	36.4	13.8	82.4	200-201
3c		H	14.0	12.1	70.6	68-69
3d	Allyl	H	45.8	15.6	76.9	39-41
3e		6,7,8-Trimethoxy	10.7	0	66.1	178-180
3f		6,7,8-Trimethoxy	9.9	0	57.2	192-193
3g		6,7,8-Trimethoxy	36.2	11.4	85.5	99-100

[a]Average yields of isolated products. [b]Traditional method: **1a-1d** (3 mmol), K_2CO_3 (5.5 mmol), 15 mL acetone, **2a-2b** (3 mmol), reflux, 8 h. [c]Without catalyst in NaOH and water: **1a-1d** (3 mmol), NaOH (150 mg) + 15 mL H_2O, rt, **2a-2b** (3 mmol), 6-12 h. [d]In-catalyst method in water: **1a-1d** (3 mmol), NaOH (150 mg) + 15 mL H_2O, rt, **2a-2b** (3 mmol), 0.25 mg indium, 6-12 h.

slowly added. The reaction mixture was stirred in a water bath at room temperature or 60 ºC and then filtered after 6-12 h. The solid was washed with sodium hydroxide (10 %, w/w), and then with water to pH 7 and purified by silica gel column chromatography (petroleum ether-ethyl acetate, 4:1, v:v) to give the desired products **3a**-**3g**.

General procedure for the preparation of compounds 3a-3g catalyzed by indium (method C): Compounds **1a**-**1d** (3 mmol) were added to water (15 mL) and sodium hydroxide (150 mg) was added. The mixture was stirred at room temperature for 1 h and then compounds **2a**-**2b** (3 mmol) and indium (0.25 mg) were slowly added. The remaining steps were the same as method B.

Table-1 shows the physical data of compounds **3a**-**3g**. Table-2 comprises the reaction conditions for preparation of compounds **3a**. Table-3 shows the IR spectra and elemental analysis of compounds **3a**-**3g**. Table-4 shows the ^{1}H NMR spectra of compounds **3a**-**3g**. Table-5 shows the ^{13}C NMR spectra of compounds **3a**-**3g**.

TABLE-2
REACTION CONDITIONS FOR
PREPARATION OF COMPOUNDS **3a**

Entry	Metal	Reaction temperature	Phase transferred catalyst	Yield[a] (%)
1	Indium	Room temp.	–	65.3
2	Zinc	Room temp.	*n*-Bu_4NBr	12.7
3	Zinc	56 ºC	*n*-Bu_4NBr	50.7
4	Tin	Room temp.	*n*-Bu_4NBr	15.2
5	Tin	56 ºC	*n*-Bu_4NBr	53.9

[a]Average yields of isolated products.

Antifungal assay: The antifungal activities of compounds **3a**-**3g** were tested against *F. graminearum*, *F. oxysporum* and *C. mandshurica* by the mycelium growth inhibition method[15]. Compounds **3a**-**3g** were dissolved in acetone before mixing with 90 mL of potato dextrose agar (PDA). The final concentration of compounds **3a**-**3g** in the medium was 50 μg/mL. All kinds of fungi were incubated in PDA at 27 ± 1 ºC for 4 days to obtain new mycelia for the antifungal assay. Then, mycelium dishes approximately 4 mm in diameter were cut from the culture medium and one of them was picked up with a sterilized inoculation needle and inoculated in the center of the PDA plate aseptically. The inoculated plates were incubated at 27 ± 1 ºC for 5 days. Acetone in sterile distilled water served as a control, whereas hymexazole served as a positive control. For each treatment, three replicates were conducted. The radial growth of fungal colonies was measured and the data were statistically analyzed. The inhibition effects of the tested compound *in vitro* on these fungi were calculated by the formula I (%) = [(C – T)/(C – 0.4)] × 100, where C and T represent the diameters of fungus growth on untreated and treated PDA and I is the inhibition rate. The EC_{50} values were estimated statistically by Probit analysis with the Probit package of SPSS 11.5 software using a personal computer[16]. The average EC_{50} (μg/mL) was taken (effective dose for 50 % inhibition) from at least three separate analyses for the inhibition of growth using the basic LD_{50} program version 1.1. The inhibition effects of compounds **3a**-**3g** are shown in Table-6. The toxicities of compounds **3a**-**3g** are shown in Table-7.

RESULTS AND DISCUSSION

Table-1 shows the reaction results using the traditional method (method **A**), without catalyst (method **B**) and with In-catalyst in water (method **C**). The In-catalyst method produced

TABLE-3
IR SPECTRA AND ELEMENTAL ANALYSIS OF COMPOUNDS **3a-3g**

Compound	IR (cm^{-1})	Elemental analysis (%): Found (calcd.)		
		C	H	N
3a	2966.5(ν_{asCH_3}), 2831.5 (ν_{sCH_3}), 1597.1-1454.3 (Ar skeleton vibration), 1238.3 ($\nu_{asAr\text{-}O\text{-}C}$), 1134.1 ($\nu_{sAr\text{-}O\text{-}C}$), 765.7 ($\delta_{Ar\text{-}H}$)	57.36 (57.57)	3.90 (4.07)	13.94 (14.13)
3b	3008.9 ($\nu_{Ar\text{-}H}$), 2958.8, 2937.6 (ν_{asCH_3}), 2835.4 (ν_{sCH_3}), 1585.5~1456.3 (Ar skeleton vibration), 1232.5 ($\nu_{asAr\text{-}O\text{-}C}$), 1132.2 ($\nu_{sAr\text{-}O\text{-}C}$), 759.9 ($\delta_{Ar\text{-}H}$)	55.64 (55.32)	4.02 (3.91)	13.50 (13.58)
3c	3010.9 ($\nu_{Ar\text{-}H}$), 2960.7 (ν_{asCH_3}), 2827.6 (ν_{sCH_3}), 1591.3-1467.8 (Ar skeleton vibration), 1469.8 (δ_{asCH_3}), 1377.2 (δ_{sCH_3}), 1240.2 ($\nu_{asAr\text{-}O\text{-}C}$), 1136.1 ($\nu_{sAr\text{-}O\text{-}C}$), 765.7 ($\delta_{Ar\text{-}H}$)	67.03 (67.14)	4.81 (4.51)	10.52 (10.44)
3d	3078.4 ($\nu_{=CH2}$), 3037.9 ($\nu_{Ar\text{-}H}$), 2978.1 ($\nu_{=CH2}$), 2922.2 (ν_{CH2}), 1635.6 ($\nu_{C=C}$), 1612.5-1483.3 (Ar skeleton vibration), 993.3 ($\delta_{\text{-}CH=}$), 920.1 ($\delta_{=CH2}$), 759.9 ($\delta_{Ar\text{-}H}$)	65.03 (65.32)	5.18 (4.98)	14.09 (13.85)
3e	2987.7 (ν_{asCH_3}), 2837.3 (ν_{sCH_3}), 1600.9~1469.7 (Ar skeleton vibration), 1240.2 ($\nu_{asAr\text{-}O\text{-}C}$), 1136.1 ($\nu_{sAr\text{-}O\text{-}C}$), 781.2 ($\delta_{Ar\text{-}H}$)	53.94 (54.31)	4.68 (4.56)	11.64 (11.52)
3f	2980.0 (ν_{asCH_3}), 2835.4 (ν_{sCH_3}), 1604.8~1467.8 (Ar skeleton vibration), 1249.9 ($\nu_{asAr\text{-}O\text{-}C}$), 1132.2 ($\nu_{sAr\text{-}O\text{-}C}$), 785.0 ($\delta_{Ar\text{-}H}$)	52.56 (52.58)	4.54 (4.41)	11.11 (11.15)
3g	3072.6 ($\nu_{Ar\text{-}H}$), 2968.5 (ν_{asCH_3}), 2835.4 (ν_{sCH_3}), 1604.8-1481.3 (Ar skeleton vibration), 1481.3 (δ_{asCH_3}), 1375.3 (δ_{sCH_3}), 1247.9 ($\nu_{asAr\text{-}O\text{-}C}$), 1124.5 ($\nu_{sAr\text{-}O\text{-}C}$), 779.2 ($\delta_{Ar\text{-}H}$)	60.43 (60.32)	5.15 (5.06)	8.01 (7.82)

TABLE-4
^{1}H NMR SPECTRA OF COMPOUNDS **3a-3g**

Compound	δ, ppm (*J*, Hz)
3a	8.97 (s, 1H, quinazoline H-2), 8.27 (d, *J* = 8.55 Hz, 1H, quinazoline H-8), 8.15 (t, *J* = 8.0 Hz, 1H, quinazoline H-7), 8.09 (d, *J* = 8.6 Hz, 1H, quinazoline H-5), 7.89 (t, *J* = 7.45 Hz, 1H, quinazoline H-6), 7.33 (s, 2H, Ph-2,6-H), 3.88 (s, 6H, Ph-3,5-site 2CH_3O), 3.77 (s, 3H, Ph-4-site CH_3O)
3b	9.14 (s, 1H, quinazoline H-2), 8.17 (d, *J* = 8.0 Hz, 1H, quinazoline H-8), 8.09 (d, *J* = 8.0 Hz, 1H, quinazoline H-5), 7.99 (t, *J* = 7.2 Hz, 1H, quinazoline H-7), 7.75 (t, *J* = 7.7 Hz, 1H, quinazoline H-6), 7.28 (s, 2H, Ph-2,6-H), 3.99 (s, 6H, Ph-3,5-site 2CH_3O), 3.94 (s, 3H, Ph-4-site CH_3O)
3c	8.86 (s, 1H, quinazoline H-2), 8.25 (d, *J* = 8.55 Hz, 1H, quinazoline H-8), 8.05 (t, *J* = 7.4 Hz, 1H, quinazoline H-7), 7.99 (d, *J* = 8.0 Hz, 1H, quinazoline H-5), 7.80 (t, *J* = 7.45 Hz, 1H, quinazoline H-6), 7.46 (t, *J* = 8.32 Hz, 1H, Ph-5-H), 7.24, 7.23 (d, 2H, Ph-2-H+ Ph-6-H), 7.13 (dd, *J* = 8.27 Hz, *J* = 2.85 Hz, 1H, Ph-4-H), 3.80 (s, 3H, Ph-3-site CH_3O)
3d	8.98 (s, 1H, quinazoline H-2), 8.04 (d, *J* = 8.0 Hz, 1H, quinazoline H-8), 7.94 (d, 1H, *J* = 8.0 Hz, quinazoline H-5), 7.79~7.82 (m, 1H, quinazoline H-7), 7.52~7.55 (m, 1H, quinazoline H-6), 5.99~6.05 (m, 1H, CH = C), 5.18~5.41 (m, 2H, C = CH_2), 4.05 (d, *J* = 7.5 Hz, 2H, CH_2)
3e	9.36 (s, 1H, quinazoline H-2), 8.35 (s, 1H, quinazoline H-5), 7.37 (s, 2H, Ph-2,6-H), 3.96-3.90 (m, 18H, 6CH_3O)
3f	9.03 (s, 1H, quinazoline H-2), 7.34 (s, 2H, Ph-2,6-H), 7.24 (s, 1H, quinazoline H-5), 4.07-3.76 (m, 18H, 6CH_3O)
3g	8.73 (s, 1H, quinazoline H-2), 7.45 (t, *J* = 8.0 Hz, 1H, Ph-5-H), 7.21, 7.20 (d, 3H, Ph-6-H+Ph-2-H+ quinazoline H-5), 7.11 (dd, *J* = 8.85 Hz, *J* = 1.7 Hz, 1H, Ph-4-H), 4.04-3.97 (t, 9H, quinazoline-6,7,8-site 3CH_3O), 3.80 (s, 3H, Ph-3-site CH_3O)

TABLE-5
^{13}C NMR SPECTRA OF COMPOUNDS **3a-3g**

Compound	δ (ppm)
3a	171.9 (Oxdiazole-5-C), 167.8 (oxdiazole-2-C), 155.9 (quinazoline C-4), 153.5 (2C, Ph-3,5-C), 152.6 (quinazoline C-2), 148.3 (quinazoline C-9), 141.0 (Ph-4-C), 135.6 (quinazoline C-7), 129.2 (quinazoline C-8), 128.6 (quinazoline C-6), 123.7 (Ph-1-C), 122.1 (quinazoline C-5), 117.8 (quinazoline C-10), 104.1 (2C, Ph-2,6-C), 60.2 (4-CH_3OPh), 56.1 (2C, 3,5-$(OCH_3)_2Ph$)
3b	171.2 (Thiadiazole-5-C), 165.1 (thiadiazole-2-C), 156.4 (quinazoline C-4), 153.8 (2C, Ph-3,5-C), 152.8 (quinazoline C-2), 148.8 (quinazoline C-9), 140.9 (Ph-4-C), 134.9 (quinazoline C-7), 129.3 (quinazoline C-8), 128.6 (quinazoline C-6), 125.2 (Ph-1-C), 123.3 (quinazoline C-5), 122.9 (quinazoline C-10), 105.2 (2C, Ph-2,6-C), 61.1 (4-CH_3OPh), 56.5 (2C, 3,5-$(OCH_3)_2Ph$)
3c	170.3 (Ph-3-C), 159.6 (Quinazoline C-4), 153.5 (quinazoline C-2), 147.7 (quinazoline C-9), 134.6 (Ph-1-C), 130.3 (quinazoline C-7), 128.4 (Ph-5-C), 128.3 (quinazoline C-8), 127.8 (quinazoline C-6), 127.4 (quinazoline C-5), 123.5 (quinazoline C-10), 122.3 (Ph-6-C), 120.9 (Ph-2-C), 115.7 (Ph-4-C), 55.3 (3-CH_3OPh)
3d	170.8 (Quinazoline C-4), 153.5 (quinazoline C-2), 147.9 (quinazoline C-9), 133.7 (quinazoline C-7), 132.8 (-CH=), 128.8 (quinazoline C-8), 127.3 (quinazoline C-6), 123.9 (quinazoline C-5), 123.8 (quinazoline C-10), 118.6 (=CH_2), 32.2 (CH_2)
3e	171.5 (Oxdiazole-5-C), 164.2 (oxdiazole-2-C), 156.1 (quinazoline C-4), 153.8 (quinazoline C-2), 153.1 (2C, Ph-3,5-C), 150.0 (quinazoline C-6), 145.7 (quinazoline C-9), 143.6 (quinazoline C-7), 139.8 (quinazoline C-8), 139.5 (Ph-4-C), 129.8 (Ph-1-C), 119.2 (quinazoline C-10), 103.9 (2C, Ph-2,6-C), 96.7 (quinazoline C-5), 62.0, 60.8, 56.4 (3C, quinazoline 6,7,8-$(CH_3O)_3$), 59.0 (4-CH_3OPh), 55.7 (2C, 3,5-$(OCH_3)_2Ph$)
3f	170.8 (Thiadiazole-5-C), 161.5 (thiadiazole-2-C), 156.6 (quinazoline C-4), 154.0 (quinazoline C-2), 153.4 (2C, Ph-3,5-C), 150.2 (quinazoline C-6), 148.0 (quinazoline C-9), 146.7 (quinazoline C-7), 140.5 (quinazoline C-8), 140.2 (Ph-4-C), 124.5 (Ph-1-C), 119.1 (quinazoline C-10), 105.0 (2C, Ph-2,6-C), 97.1 (quinazoline C-5), 62.2, 61.1, 56.5 (3C, quinazoline 6,7,8-$(CH_3O)_3$), 60.1 (4-CH_3OPh), 56.1 (2C, 3,5-$(OCH_3)_2Ph$)
3g	168.0 (Ph-3-C), 160.2 (Quinazoline C-4), 153.9 (quinazoline C-2), 151.6 (quinazoline C-6), 147.9 (quinazoline C-9), 147.4 (quinazoline C-7), 140.5 (Ph-1-C), 130.9 (quinazoline C-8), 128.5 (Ph-5-C), 128.3 (quinazoline C-10), 121.4 (Ph-6-C), 119.9 (Ph-2-C), 116.1 (Ph-4-C), 97.8 (quinazoline C-5), 62.7, 61.7, 56.8 (3C, quinazoline 6,7,8-$(CH_3O)_3$), 55.9 (3-CH_3OPh)

TABLE-6
INHIBITION EFFECT OF COMPOUND **3a-3g** AGAINST THREE KINDS OF PHYTOPATHOGENIC FUNGI *in vitro*

Compound	Con. (μg mL^{-1})	Inhibition (%)[a]		
		F. graminearum	*F. oxysporum*	*C. mandshurica*
1a	50	11.2 ± 1.83[b]	11.2 ± 0.94[b]	16.8 ± 1.50[b]
1b	50	14.6 ± 1.92[b]	22.7 ± 1.12[b]	13.2 ± 1.75[b]
1c	50	43.2 ± 1.70[b]	49.4 ± 0.86[b]	39.4 ± 1.64[b]
1d	50	69.5 ± 1.66[b]	71.9 ± 0.99[b]	70.8 ± 1.52[b]
1e	50	24.2 ± 1.58[b]	12.1 ± 1.13[b]	8.1 ± 1.55[b]
1f	50	32.3 ± 1.73[b]	44.1 ± 1.10[b]	42.5 ± 1.60[b]
1g	50	42.0 ± 1.53[b]	37.2 ± 0.92[b]	46.8 ± 1.52[b]
Hymexazole[c]	50	63.6 ± 1.63[b]	51.5 ± 0.91[b]	57.8 ± 1.46[b]

[a]Average of three replicates. [b]Compared with acetone control, Hymexazole and **3a-3g** treatment showed statistically significant inhibition ($p < 0.05$). [c]Positive control.

TABLE-7
TOXICITY OF COMPOUND **3d** ON THREE KINDS OF PHYTOPATHOGENIC FUNGI *in vitro*

Fungi	EC_{50}[a] (μg mL^{-1})	Toxic regression equation[a]	r
F. graminearum	25.88 ± 3.81[b]	y = 2.09x + 2.08	0.995
F. oxysporum	17.08 ± 3.62[b]	y = 1.86x + 2.34	0.984
C. mandshurica	28.77 ± 4.37[b]	y = 1.80x + 2.37	0.993

[a]Average of three replicates. [b]The values were estimated statistically by SPSS 11.5 software using a personal computer. 95 % confidence limits for EC_{50}.

both facile reactions and higher yields. The yields of synthesized compound **3a**-**3g** increased by 31.1-56.6 % under indium mediation in water.

To optimize the reaction parameters, we selected compound **3a** for further study under different conditions with a metal catalyst and the results are shown in Table-2. When the etherification was mediated by indium in water, the reaction went smoothly at room temperature without any promoter. When zinc or tin phase-transferred catalysts were used, heat was usually required. Reactant **1a** was accordingly tested and the results are shown in Table-2. With this method, compounds **3a**-**3g** were synthesized in high yields and easily purified by silica gel column chromatography. An organic co-solvent was also not necessary. In the absence of indium and water, the reaction was much slower and the yield was lower when the reaction mixture was heated in DMF or acetonitrile in the presence of K_2CO_3 and triethylamine. Under this condition, by-products were produced because 4-chloroquinazoline was sensitive to the basic aqueous medium and heating.

A preliminary bioassay suggested that compound **3d** had good antifungal activity against *Fusarium graminearum* with EC_{50} = 25.88 μg/mL, *Fusarium oxysporum* with EC_{50} = 17.08 μg/mL and *Cytospora mandshurica* with EC_{50} = 28.77 μg/mL.

Conclusion

4-Thioquinazoline compounds **3a**-**3g** were synthesized by a novel method under indium mediation in water. The method offers several advantages, such as convenient reaction and excellent yields, over the traditional synthesis method that involves organic solvents and low yields. The compounds were evaluated for their *in vitro* antifungal activity against *F. graminearum*, *F. oxysporum* and *C. mandshurica*. Compound **3d** inhibited *F. graminearum* with EC_{50} = 25.88 μg/mL, *F. oxysporum* with EC_{50} = 17.08 μg/mL and *C. mandshurica* with EC_{50} = 28.77 μg/mL. Unfortunately, the other tested compounds exhibited low antifungal activities against *F. graminearum*, *F. oxysporum* and *C. mandshurica*.

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