Green synthesis of tri/tetrasubstituted 1*H*-imidazoles and 2,3-dihydroquinazolin-4(1*H*)-ones using nano aluminium nitride as solid source of ammonia

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Abstract. A simple, green and cost-effective protocol was achieved for the synthesis of tri/tetrasubstituted-1*H*-imidazoles and 2,3-dihydroquinazolin-4(1*H*)-ones using nano aluminium nitride. The reaction was carried out under catalyst-free conditions and the products were isolated in good to excellent yield.

Keywords. Imidazoles; 2,3-dihydroquinazolin-4(1*H*)-ones; nano aluminium nitride; multicomponent; one-pot.

1. Introduction

Nitrogen-heterocyclic compounds (NHCs) are produced for a variety of applications, including pharmaceuticals, cosmetics, pesticides, disinfectants, agrochemicals, dyestuffs and antifreeze.¹ In particular, derivatives of imidazoles and hydroquinazolins are used as reagents in industry.² The five-membered imidazole ring is one of the important substructures found in a large number of natural products and pharmacologically active heterocyclic compounds.³ The members of this class of compounds are known to possess NO synthase inhibition and antifungal, antimycotic, antibiotic, antiulcerative, antibacterial, antitumor, and CB1 receptor antagonistic activities.⁴ Among the imidazoles, 2,4,5-triphenylimidazoles can be used as light-sensitive materials in photography, as inhibitors of IL-1 or p38 MAP kinase, fungicides and herbicides and plant growth regulators.^{5,6}

There are numerous methods in the literature for the synthesis of highly substituted imidazoles: condensation of 1,2-diones, aldehydes, primary amines and ammonia in the presence of several catalysts such as FeCl₃·6H₂O,⁷ ionic liquids,⁸ K₅CoW₁₂O₄₀·3H₂O,⁹ HClO₄·SiO₂,¹⁰ NaH₂PO₄,¹¹ heteropolyacids,¹² InCl₃·3H₂O,¹³ Bronsted acidic ionic liquid,¹⁴ MCM-41 or p-TsOH.¹⁵ In addition, they can also be accessed by hetero-Cope rearrangement, condensation of a 1,2-diketone with an aryl nitrile and primary amine under microwave irradiation, and N-alkylation of trisubstituted imidazoles.¹⁶

Many of the synthetic methods for imidazoles suffer from one or more disadvantages such as low yields, harsh reaction conditions, prolonged time period and application of hazardous and often expensive acid catalysts and expensive catalysts long reaction times.¹⁷ Moreover, the synthesis of these compounds is usually carried out in polar solvents such as ethanol, methanol, acetic acid, dimethylformamide (DMF), and dimethylsulfoxide (DMSO), leading to complex isolation and recovery procedures.¹⁴

2,3-Dihydroquinazolin-4(1*H*)-ones, an important class of heterocyclic compounds, influence numerous cellular processes.¹⁸ These exhibit a wide range of biological activities, such as antitumor, antibiotic, antidefibrillatory, antipyretic, analgesic, antihypertonic, diuretic, antihistamine, antidepressant, and vasodilating behavior.¹⁹ Furthermore, quinazolinone skeleton is frequently found in various natural products. Some examples include the anticancer compound trimetrexate, the sedative methaqualone, the alpha adrenergic receptor antagonist such as doxazosin and the antihypertensive agent ketanserin.²⁰

Environmental problems are expected to become more serious in the 21st century. Therefore, the development of simple, efficient, clean, high-yielding, and environmentally friendly approaches without the use of a catalyst or an organic solvent for the synthesis of these compounds is an important task for organic chemists.

In recent years, polymer composite materials with excellent thermal conductivity have attracted increasing attention. Among them, nano-sized aluminium nitride (nano-AlN) is a nontoxic, low cost substance, readily available as high purity powder.²¹

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2. Experimental

Chemicals were purchased from Sigma-Aldrich, Fisher, and Merck. The tri/tetrasubstituted imidazole and 2,3-dihydroquinazolin-4(1H)-one derivatives were characterized by ^1H and ^{13}C NMR spectra (^1H NMR, 400 MHz; ^{13}C NMR, 100 MHz) and IR. Scanning electron microscopy (SEM) studies were conducted on a JSM 740 scanning electron microscope.

2.1 Procedure for preparation of 2-(4-chlorophenyl)-4,5-diphenyl-1H-imidazole **1a**

To the mixture of 0.102 g (5 mmol) of aluminum nitride, 0.140 g (1 mmol) of 4-chlorobenzaldehyde in a few drops of water (0.3 mL), 0.210 g (1 mmole) benzil was added at room temperature. The reaction vessel was sealed and allowed to warm to 130°C for 8 h with constant stirring (the progress of the reaction was monitored by TLC). After completion of the reaction, the reaction mixture was cooled down to room temperature and the crude product extracted by acetone. Acetone was removed by simple evaporation. Finally, crude product 2-(4-chlorophenyl)-4,5-diphenyl-1H-imidazole **1a** was obtained by recrystallization from hot EtOH; yield, 0.273 g (83%).

2.2 Procedure for preparation of 1,2-bis(4-chlorophenyl)-4,5-diphenyl-1H-imidazole **2a**

To the mixture of 0.102 g (5 mmol) of aluminum nitride, 0.140 g (1 mmol) of 4-chlorobenzaldehyde in a few drops of water (0.3 mL), 0.210 g (1 mmol) benzil and 0.127 g (1 mmol) 4-chloroaniline were added at room temperature. The reaction vessel was sealed and allowed to warm to 130°C for over 8 h with constant stirring (the progress of the reaction was monitored by TLC). After completion of the reaction, the reaction mixture cooled down to room temperature and the crude product extracted by acetone. Acetone was removed by simple evaporation. Finally crude product 1,2-bis(4-chlorophenyl)-4,5-diphenyl-1H-imidazole **2b** were obtained by recrystallization in the hot EtOH, yield 0.316 g (72%) and M.p. 185–186°C.

2.3 Procedure for preparation of 2-(4-chlorophenyl)-2,3-dihydroquinazolin-4(1H)-one **3a**

A mixture of isatoic anhydride (0.234 g, 1 mmol), aluminum nitride (1.5 mmol), 4-chlorobenzaldehyde (1 mmol) was heated at 80°C for 6 h. The reaction was monitored by TLC analysis using petroleum and

n-hexan-acetate (4:1, v/v). After completion of the reaction, the product was dissolved in hot ethanol, filtered and from the filtrate ethanol was removed by evaporation. Finally, crude product 2-(4-chlorophenyl)-2,3-dihydroquinazolin-4(1H)-one **3a** were obtained by recrystallization in the EtOH and water, yield (66%).

2.4 Characterization data of selected compounds

2-(4-chlorophenyl)-4,5-diphenyl-1H-imidazole **1a**: M.p.: 254–256°C. FT-IR-ATR (ν_{max} , cm^{-1}): 3413, 3063, 2838, 1635, 1481, 1323, 1152, 830, 695. ^1H NMR (400 MHz CDCl_3): δ 7.87 (d J = 8.4 Hz 2H) 7.54 (d J = 6.8 Hz 4H) 7.45–7.30 (m 8H) ppm. ^{13}C NMR (100 MHz CDCl_3): δ 144.8 134.9 129.1 128.7 127.9 127.7 126.7 ppm.

2-(4-bromophenyl)-4,5-diphenyl-1H-imidazole **1b**: M.p.: 240–244°C. FT-IR-ATR (ν_{max} , cm^{-1}): 3323, 3061, 2835, 1645, 1481, 1322, 827, 694. ^1H NMR (400 MHz, DMSO): δ 7.32–7.42 (m, 6H), 7.53–7.55 (m, 4H), 7.70 (d, 2H, J = 8.4 Hz), 8.05 (d, 2H, J = 8.4 Hz), 12.85 (br, 1H) ppm.

2-(3-Nitrophenyl)-4,5-diphenyl-1H-imidazole **1c**: M.p.: 312–314°C. FT-IR-ATR (ν_{max} , cm^{-1}): 3383, 3320, 3069, 1695, 1524, 1348, 1070, 697.

2-(naphthalen-2-yl)-4,5-diphenyl-1H-imidazole **1d**: M.p.: 267–268°C. ^1H NMR (400 MHz, DMSO): δ 7.26 (t, J = 14, 1H), 7.35 (m, 2H), 7.41 (t, J = 14 Hz, 1H), 7.46–7.49 (m, 2H), 7.55–7.62 (m, 7H), 7.95–8.04 (m, 3H), 8.27 (d, J = 8.4 Hz, 1H), 8.63 (s, 1H) ppm. ^{13}C NMR (100 MHz, DMSO): δ 123.9, 124.2, 126.8, 127.1, 127.2, 127.6, 128.2, 128.3, 128.6, 128.7, 128.8, 128.9, 129.2, 130, 131.4, 133.2, 133.5, 135.6, 137.9, 138.5, 145.9 ppm.

4,5-diphenyl-2-(p-tolyl)-1H-imidazole **1e**: M.p.: 227–230°C. FT-IR-ATR (ν_{max} , cm^{-1}): 3418, 3034, 2987, 2793, 1600, 1586, 1321, 764. ^1H NMR (400 MHz, CDCl_3): δ 2.43 (s, 3H), 7.24–7.40 (m, 8H), 7.58 (br, 4H), 7.83 (d, 2H, J = 8 Hz), 9.30 (br, 1H) ppm.

2,4,5-Triphenyl-1H-imidazole **1f**: M.p.: 259–261°C. FT-IR-ATR (ν_{max} , cm^{-1}): 3404, 3038, 2962, 2852, 1601, 1408, 1071, 696.

2-(3,4-dimethoxyphenyl)-4,5-diphenyl-1H-imidazole **1g**: M.p.: 183–185°C. FT-IR-ATR (ν_{max} , cm^{-1}): 3055, 2937, 1602, 1484, 1263, 1024, 698.

2-(anthracene-9-yl)-4,5-diphenyl-1H-imidazole **1h**: M.p.: 219–220°C. FT-IR-ATR ($\nu_{\max}$, cm^{-1}): 3381, 3048, 3069, 1598, 1443, 1342, 1079, 695. ^1H NMR (400 MHz CDCl_3): δ 7.34–7.50 (m 10H), 7.96 (d, J = 7.2 Hz, 4H), 7.93 (d, J = 8.8 Hz, 2H), 8.02 (d, J = 8.4 Hz, 2H), 8.50 (s, 1H), 9.87 (b, 1H) ppm. ^{13}C NMR (100 MHz, CDCl_3): δ 143.6, 131.5, 131.1, 129, 128.6, 128.5, 127.9, 127.5, 126.6, 125.8, 125.4, 125 ppm.

5-bromo-2-(4,5-diphenyl-1H-imidazol-2-yl)phenol **1i**: M.p.: 221–224 °C. FTIR (KBr, cm^{-1}): 3417, 3392, 3210, 2680, 1644, 1487, 712.

2-(4-methoxyphenyl)-4,5-diphenyl-1H-imidazole **1j**: M.p.: 221–224°C. FTIR (KBr, cm^{-1}): 3034, 2987, 1600, 1321, 1261, 764. ^1H NMR (400 MHz, DMSO): δ 3.82 (s, 3H), 7.06 (d, J = 8.8 Hz, 2H), 7.37 (m, 6H), 7.52 (d, J = 6.4 Hz, 4H), 8.03 (d, J = 8.8 Hz, 2H), 12.5 (br, 1H) ppm. ^{13}C NMR (100 MHz, DMSO): δ 55.6, 114.6, 123.2, 127.3, 127.7, 128.2, 128.9, 146.2, 159.9 ppm.

1-(4-chlorophenyl)-2-(4-chlorophenyl)-4,5-diphenyl-1H-imidazole **2a**: Isolated Yield = 72%. M.p.: 185–186°C. ^1H -NMR (CDCl_3 , 400MHz): δ 6.99 (d, 2H, J = 8.4), 7.14–7.16 (m, 2H), 7.23–7.33 (m, 10H), 7.39 (d, 2H, J = 8.8), 7.61 (d, 2H, J = 8.4) ppm.

1-benzyl-2-(4-chlorophenyl)-4,5-diphenyl-1H-imidazole **2b**: M.p.: 161–164°C. FTIR (KBr, cm^{-1}): 3059, 3030, 2938, 1662, 1446, 1085, 693.

1-benzyl-2-(4-bromophenyl)-4,5-diphenyl-1H-imidazole **2c**: M.p.: 162–165°C. FTIR (KBr, cm^{-1}): 3055, 1597, 1496, 1071, 694.

3-(1-benzyl-4,5-diphenyl-1H-imidazol-2-yl)phenol **2d**: M.p.: 229–231°C. FTIR (KBr, cm^{-1}): 3059, 3031, 1598, 1457, 1234, 695.

1-benzyl-2-(3,4-dimethoxyphenyl)-4,5-diphenyl-1H-imidazole **2e**: M.p.: 161–163°C. FTIR (KBr, cm^{-1}): 3322, 3052, 3001, 1602, 1529, 1484, 1263, 698.

1-benzyl-2-(2-nitrophenyl)-4,5-diphenyl-1H-imidazole **2f**: M.p.: 144–148°C. FTIR (KBr, cm^{-1}): 3059, 3031, 2925, 1602, 1497, 1300, 1076, 692.

2-(4-bromophenyl)-1,4,5-triphenyl-1H-imidazole **2g**: M.p.: 179–183°C. FTIR (KBr, cm^{-1}): 3055, 1597, 1477, 1446, 1071, 831, 694.

1,4,5-triphenyl-2-(p-tolyl)-1H-imidazole **2h**: M.p.: 175–178°C. FTIR (KBr, cm^{-1}): 3042, 2919, 1595, 1414, 1024, 695.

2-(4-chlorophenyl)-1,4,5-triphenyl-1H-imidazole **2i**: M.p.: 172–174°C. FTIR (KBr, cm^{-1}): 3060, 1597, 1496, 1446, 1082, 837, 698.

4-(1,4,5-Triphenyl-1H-imidazol-2-yl)-phenol **2j**: M.p.: 279–280°C. FTIR (KBr, cm^{-1}): 3448, 3057, 2974, 2613, 1604, 1492, 1273, 696.

2-(4-chlorophenyl)-2,3-dihydroquinazolin-4(1H)-one **3a**: M.p.: 204–207°C. ^1H NMR (250 MHz, $\text{DMSO}-d_6$): δ 8.29 (s, 1H), 7.61–7.41 (m, 5H), 7.26–7.2 (t, 1H), 7.12 (s, 1H), 6.75–6.63 (m, 2H), 5.75 (s, 1H) ppm. ^{13}C NMR (62 MHz, $\text{DMSO}-d_6$): δ 163.9, 148.1, 141.1,

Scheme 1. Synthesis of 2,4,5-trisubstituted imidazoles.

Scheme 2. Synthesis of 1,2,4,5-tetrasubstituted imidazoles.

133.8, 133.4, 129.2, 128.7, 127.8, 117.7, 115.4, 114.9, 66.2 ppm.

2-(4-methylphenyl)-2,3-dihydroquinazolin-4(1H)-one 3d: M.p.: 220–225°C. ¹H NMR (250 MHz, DMSO-*d*₆): δ 8.21 (s, 1H), 7.62–7.59 (d, 1H), 7.38–7.35 (d, 2H), 7.26–7.16 (m, 3H), 7.03 (s, 1H), 6.75–6.63 (m, 2H), 5.71 (s, 1H), 2.49–2.42 (s, 3H) ppm. ¹³C NMR (62 MHz, DMSO-*d*₆): δ 164.1, 148.4, 139.1, 138.2, 133.7, 129.3, 127.8, 127.2, 117.5, 115.4, 114.9, 66.8, 21.2 ppm.

3. Results and Discussion

In continuing our investigations on the synthetic organic reactions,^{22–25} in efforts to develop an efficient procedure for the synthesis of substituted imidazole and quinazolin-4(1H)-one derivatives, we report the synthesis of tri/tetrasubstituted imidazole and quinazolin-4(1H)-ones, *via* one-pot and multi-component condensation using aluminium nitride under solvent-free or water as solvent conditions.

It was found that benzil could be condensed with aromatic aldehydes and nano aluminum nitride to smoothly afford 2,4,5-trisubstituted-1H-imidazole derivatives, **1a–1j** with excellent yields at 80°C under solvent-free condition in the presence a few drops of water (0.3 mL) (scheme 1). The same reaction with an additional amine yielded 1,2,4,5-tetrasubstituted-1H-imidazole derivatives **2a–2j** (scheme 2).

Table 1. Optimization of reaction time^a.

Entry	AlN (mmol)	Time (h)	Yield (%)
1	4	3	10
2	4	5	50
3	5	6	68
4	4	8	69
5	5	8	83

^aMolar ratio of the reagents: 4-chlorobenzaldehyde 1 mmol, benzyl 1 mmol.

^bIsolated yield.

Initially, the effect of reaction time on the yield was investigated (table 1, entry 1–5). The reaction took place quickly at first and gradually reached the balance after 8 h. The yield changed little after 8 h. Also, the effect of reaction time on the condensation, a one-pot reaction of isatoic anhydride, aldehyde and aluminum nitride for the synthesis of substituted quinazolin-4(1H)-ones,^{32,33} **3a–3e** in water was investigated (scheme 3). When reaction time was increased gradually from 3 to 6 h, the rate of the reaction increased from 2 to 63%. Increase of time to 7 h has less effect on the yield (66%) of the reaction.

We carried out the study in search for optimal amount of aluminum nitride for the proposed transformation using as a model the reaction involving 1 mmol of 4-chlorobenzaldehyde, 1 mmol of benzil for 2,4,5-trisubstitution and also 1 mmol of 4-chloroaniline for synthesis of 1,2,4,5-tetrasubstituted imidazole derivatives in the presence of different amounts of nano aluminum nitride in few drops of water (0.3 mL) at 130°C was conducted, and the significant data obtained as a function of the amount of aluminum nitride are shown in (figure 1, 1a and 2b). The best results were obtained with 5 mmol of aluminum nitride in the presence of a few drops of water (0.3 mL) and under solvent-free conditions after 8 h.

Figure 1. Optimization of the amount of aluminum nitride for the synthesis of substituted imidazole.

Scheme 3. Synthesis of substituted quinazolin-4(1H)-ones.

Figure 2. Optimization of the amount of aluminum nitride for the synthesis of substituted quinazolin-4(1H)-ones.

In order to study we optimized the synthesis of substituted quinazolin-4(1H)-ones with various amount (mmol) of nano AlN. The condensation, a one-pot reaction of isatoic anhydride (1 mmol), 3-nitrobenzaldehyde (1 mmol) was chosen as a model reaction (figure 1).

Aluminum nitride plays a major role in the product yield. It was observed that increasing the loading of the nano AlN from 1 to 1.5 mmol gave an improved yield of 63% of the product (figures 2, 3e). Further increase of aluminum nitride loading leads to lower reaction yields.

Under optimized conditions for aluminum nitride, the scope of synthesis of 2,4,5-trisubstituted imidazole derivatives with other aldehydes was found to be quite good. As shown in (table 2), the method has the ability to tolerate a variety of functional groups such as chloro, nitro, methoxy and methyl.

Then, under these conditions, 1,2,4,5-tetrasubstituted imidazole derivatives were formed cleanly, The results (table 3) clearly indicate the feasibility of four-component reaction. The products were synthesized in good yield and a variety of different substituted aldehydes and amines were subjected to this reaction.

The optimized reaction conditions were extended to the condensation of other aldehydes with isatoic anhydride and aluminum nitride at 80°C. Aromatic aldehydes bearing both electron-deficient and electron-rich substituents afforded the desired 2,3-disubstituted dihydroquinazolin-4(1H)-one in good yields (table 4).

Major advantage of this protocol is the simplicity of operation and isolation of the products, which have been achieved by simple washing and crystallization of the crude products.

The scanning electron microscope (SEM) images of nano aluminium nitride showed that the resulting particles were irregular sphere structure with the particle size is about 40 nm (figure 3a). In the presence of water, the nano AlN will be decomposed to form

Figure 3. SEM images: (a) nano AlN, (b) nano AlN in reaction and (c) Al(OH)_3 after reaction.

Table 2. Synthesis of 2,4,5-trisubstituted imidazole derivatives^a.

Entry	R	Product	Yield ^b (%)	Mp (°C) found (reported)
1	4-ClC ₆ H ₄	1a	83	254-256 (259-269) ²⁶
2	4-BrC ₆ H ₄	1b	70	240-244(252-254) ²⁶
3	3-NO ₂ C ₆ H ₄	1c	79	312-314(315-317) ²⁷
4	C ₁₀ H ₇	1d	80	267-268(273-276) ²⁸
5	4-CH ₃ C ₆ H ₄	1e	74	227-230(226-228) ²⁶
6	C ₆ H ₅	1f	72	259-261(267-269) ²⁹
7	3,4-(OCH ₃) ₂ C ₆ H ₃	1g	80	183-185(215) ⁶
8	C ₁₄ H ₉	1h	98	219-220
9	5-Br-2-OHC ₆ H ₃	1i	86	311-314(186-187) ²⁹
10	4-OCH ₃ C ₆ H ₄	1j	84	221-224(228-231) ²⁶

^aMolar ratio of the reagents: aluminum nitride/aldehyde/benzil (5 mmol/1 mmol /1 mmol).

^bIsolated yield.

Table 3. Synthesis of 1,2,4,5-tetrasubstituted imidazole derivatives^a.

Entry	R ¹	R ²	Product	Yield (%) ^b	Mp (°C) found (reported)
1	4-ClC ₆ H ₄	4-ClC ₆ H ₄	2a	72	185-186 (187-189) ¹⁴
2	4-ClC ₆ H ₄	C ₆ H ₅ CH ₂	2b	77	161-164(161-163) ²⁹
3	4-BrC ₆ H ₄	C ₆ H ₅ CH ₂	2c	83	162-165(172-175) ¹⁶
4	3-OHC ₆ H ₄	C ₆ H ₅ CH ₂	2d	93	229-231(232-235) ³
5	3,4-(OCH ₃) ₂ C ₆ H ₃	C ₆ H ₅ CH ₂	2e	93	161-163
6	2- NO ₂ C ₆ H ₄	C ₆ H ₅ CH ₂	2f	81	144-148(152-155) ³⁰
7	4-BrC ₆ H ₄	C ₆ H ₅	2g	70	179-183(191-193) ⁶
8	4-CH ₃ C ₆ H ₄	C ₆ H ₅	2h	71	175-178(187-189) ²⁵
9	2-ClC ₆ H ₄	C ₆ H ₅	2i	76	172-174(160-162) ¹⁰
10	4-OHC ₆ H ₄	C ₆ H ₅	2j	81	279-280(280-281) ¹⁰

^aMolar ratio of the reagents: aluminum nitride/aldehyde/amine/benzil (5 mmol/1 mmol/1 mmol/1 mmol).

^bIsolated yield.

Table 4. Synthesis of 2,3- dihydroquinazolin-4(1H)-one derivatives^a.

Entry	R	Product	Yield ^b (%)	M.p. (°C) found (reported)
1	4-ClC ₆ H ₄	3a	66	204-207(205-2206) ³¹
2	3-NO ₂ C ₆ H ₄	3b	73	213-216 (216-217) ³¹
3	4-OCH ₃ C ₆ H ₄	3c	66	189-193 (192-193) ³¹
4	4-CH ₃ C ₆ H ₄	3d	62	220-225(223-224) ³¹
5	C ₆ H ₅	3e	63	218-222(218-219) ³¹

^aMolar ratio of the reagents: aluminum nitride/aldehyde/isatoic anhydride (1.5 mmol/1 mmol/1 mmol).

^bIsolated yield.

aluminum hydroxide and ammonia. These particles are connected with each other without obvious boundaries. SEM image showed that diameters of the particles in the reaction mixture are approximately 1 μ m (figure 3b). After the reaction, the particle size increased and the average size of amorphous aluminum hydroxide is in the range of 500 nm (figure 3c).

According to the results, the reaction can be mechanistically considered as aluminum nitride generating a solution of ammonia by reaction with water (scheme 4). Based on this mechanism, the Al(OH)₃ as a Lewis acid participates in the reaction which activates the aldehydes, benzil and isatoic anhydride followed by nucleophilic addition of amines.

Scheme 4. Possible mechanism.

4. Conclusion

In conclusion, we have developed a new strategy that provides an efficient way for the synthesis of tri/tetra-substituted-1H imidazoles and 2,3-dihydroquinazolin-4(1H)-ones using nano AlN as a source of ammonia. Our method involves mild reaction conditions, very simple and accessible starting materials. The key advantages of this process are high yields, elimination of organic solvents and catalysts, easy work-up and purification of products by non-chromatographic methods.

Supplementary Information

Additional information pertaining to characterization of the tri/tetrasubstituted imidazole and 2,3 dihydroquinazolin-4(1H)-one derivatives, namely, IR spectra, ^1H and ^{13}C NMR spectra are available at www.ias.ac.in/chemsci.

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References

- Xing X, Zhu X, Li H, Jiang Y and Ni J 2012 *Chemosphere* **86** 368
- Mohammadizadeh M R, Hasaninejad A and Bahramzadeh M 2009 *Synth. Commun.* **39** 3232
- Niknam K, Deris A, Naeimi F and Majleci F 2011 *Tetrahedron Lett.* **52** 4642
- Li W and Lam Y 2005 *J. Comb. Chem.* **7** 644
- Karimi A R, Alimohammadi Z and Amini M M 2010 *Mol. Divers.* **14** 635
- Das B, Kashanna J, Kumar R A and Jangili P 2013 *Monatsh Chem.* **144** 223
- Heravi M M, Derikvand F and Haghighi M 2008 *Monatsh Chem.* **139** 31
- Shaterian H R and Ranjbar M J 2011 *Mol. Liq.* **160** 40
- Nagarapu L, Apuri S and Kantevari S 2007 *J. Mol. Catal. A: Chem.* **266** 104
- Kantevari S, Vuppalapati S V N, Biradar D O and Nagarapu L 2007 *J. Mol. Catal. A: Chem.* **266** 109
- Karimi-Jaberi Z and Barekat M 2010 *Chin. Chem. Lett.* **21** 1183
- Heravi M M, Derikvand F and Bamoharram F F 2007 *J. Mol. Catal. A: Chem.* **263** 112
- Sharma S D, Hazarika P and Konwar D 2008 *Tetrahedron Lett.* **49** 2216
- Davoodnia A, Heravi M M, Safavi-Rad Z and Tavakoli-Hoseini N 2010 *Synth. Commun.* **40** 2588
- Hekmat Shoar R, Rahimzadeh G, Derikvand F and Farzaneh M 2010 *Synth. Commun.* **40** 1270
- Emrani A, Davoodnia A and Tavakoli-Hoseini N 2011 *Bull. Korean Chem. Soc.* **32** 2385
- Teimouria A and Najafi A 2011 *J. Mol. Catal. A: Chem.* **346** 39
- Murthy P, Rambabu D, Krishna G R, Reddy C M, Prasad K R S, Basaveswara Rao M V and Pal M 2012 *Tetrahedron Lett.* **53** 6923
- Subba Reddy B V, Venkateswarlu A, Madan C and Vinu A 2011 *Tetrahedron Lett.* **52** 1891
- Vilas B L, Shinde P V and Shingare M S 2013 *Tetrahedron Lett.* **54** 5778

21. Yu J H, Duan J K, Peng W Y, Wang L C, Peng P and Jiang P K 2011 *Polymer Lett.* **5** 132
22. Ghorbani-Choghamarani A, Zolfigol M A, Salehi P, Ghaemi E, Madrakian E, Nasr-Isfahanid H and Shahamirian M 2008 *Acta Chim. Slov.* **55** 644
23. Ghorbani-Choghamarani A and Taghipour T 2011 *Lett. Org. Chem.* **8** 470
24. Ghorbani-Choghamarani A, Nikoorazm M, Goudarziafshar H, Shokr A and Almasi H 2011 *J. Chem. Sci.* **123** 453
25. Ghorbani-Choghamarani A, Zolfigol M A, Hajjami M, Goudarziafshar H, Nikoorazm M, Yousefi S and Tahmasbi B 2011 *J. Brazil. Chem. Soc.* **22** 525
26. Shaabani A and Rahmati A 2006 *J. Mol. Cat. A: Chem.* **249** 246
27. Acke D, Orru R and Stevens C 2006 *Qsar. Comb. Sci.* **25** 474
28. Safari J, Dehghan Khalili S and Banitaba S H 2011 *Synthetic Commun.* **41** 2359
29. Samai S, Nandi G, Singh P and Singh M S 2009 *Tetrahedron* **65** 10155
30. Sadeghi B, Mirjalili B B F and Hashemi M 2008 *Tetrahedron Lett.* **49** 2575
31. Rostami A and Tavakoli A 2011 *Chin. Chem. Lett.* **22** 1320
32. Wang M, Zhang T T, Liang Y and Gao J J 2011 *Chin. Chem. Lett.* **22** 1423
33. Kanhe N S, Tekale S U, Kulkarni N V, Nawale A B, Das A K, Bhorkaskar S V, Ingle R D and Pawar R P 2012 *J. Iran. Chem. Soc.* **10** 243