Available online at www.sciencedirect.com
ScienceDirect
Tetrahedron 64 (2008) 3017–3024
Tetrahedron
www.elsevier.com/locate/tet

Lewis acid promoted imino Diels–Alder reactions of 5-dienyl pyrimidinones with *N*-aryl/naphthyl imines: synthesis of novel quinoline/benzoquinoline derivatives

Gaurav Bhargava, Chander Mohan, Mohinder P. Mahajan*

Department of Applied Chemistry, Guru Nanak Dev University, Amritsar 143005, Punjab, India

Received 4 September 2007; received in revised form 2 January 2008; accepted 10 January 2008
Available online 16 January 2008

Abstract

The chemo- as well as regioselective imino Diels–Alder reactions of 5-dienyl pyrimidinones with *N*-aryl as well as *N*-naphthyl imines in the presence of a different Lewis acid catalysts resulting in novel quinoline and benzoquinoline derivatives are reported.

1. Introduction

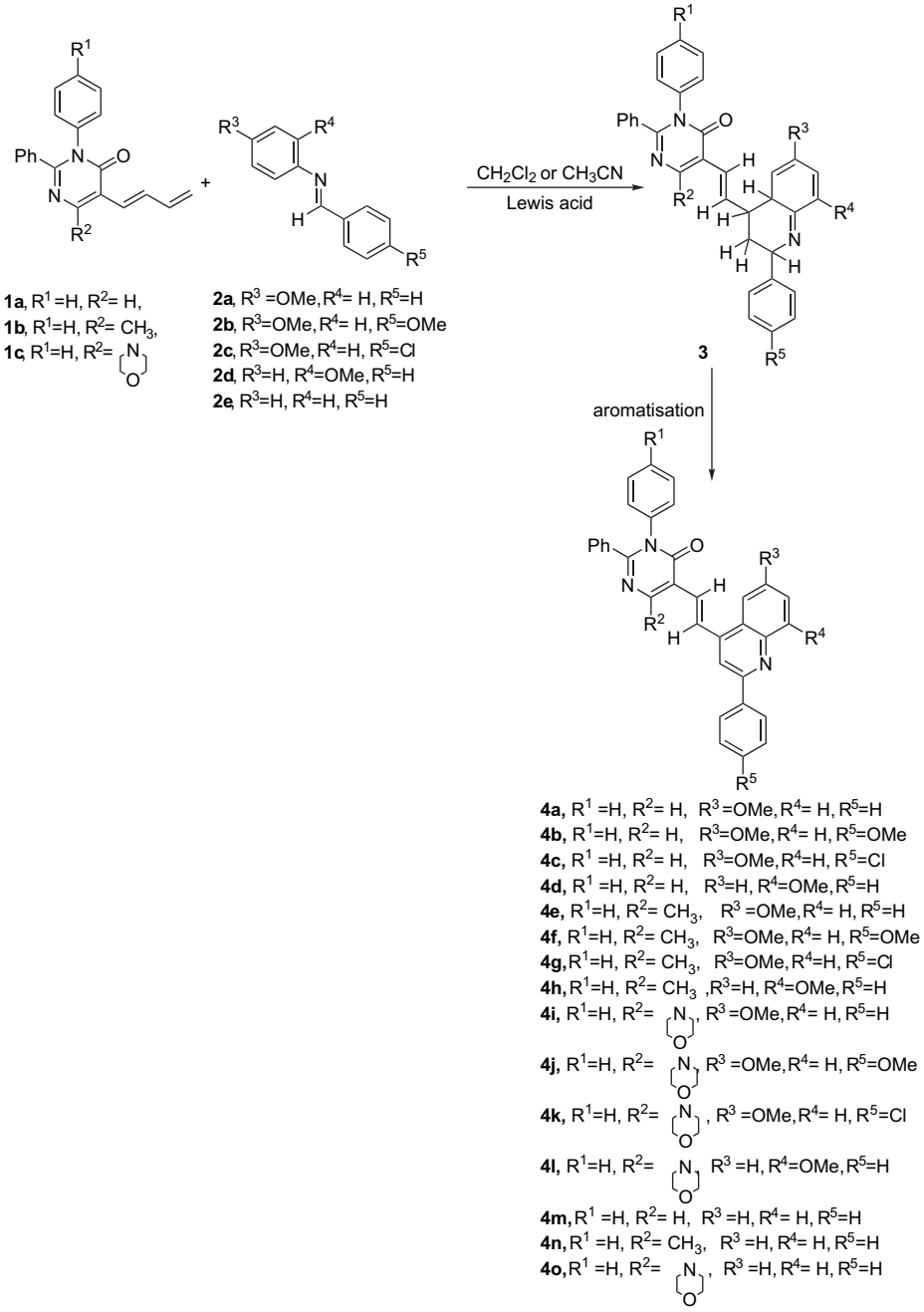
The imino Diels–Alder reaction (IDA) provides an efficient route for the construction of nitrogen containing functionalized as well as fused ring heterocycles.[1,2] In recent years, this reaction is attracting an increasing attention and special efforts are directed towards activating imine systems for cycloadditions by increasing their electron-deficient character.[3] Also, the transition metal catalysed IDA reactions have been explored as convenient routes for the formation of a wide variety of heterocycles.[4] Earlier reports in this direction described the participation of unhindered activated alkenes, cyclopentadiene or symmetrical activated butadienes in IDA cycloaddition reactions with simple *N*-aryl imines leading to the synthesis of tetrahydro/dihydroquinoline and di/tetrahydropyridine derivatives, respectively.[5] However, such reactions suffered from lack of chemoselectivity.[6] There are numerous reports on the biological importance of functionalized pyrimidinone derivatives for their antitumour,[7] antiviral,[8] antitubercular,[9] antifungal,[10] molluscicidal[11]and larvacidal[11] activity and activity against positive strand (vesicular stomatitis virus) RNA virus.[12] Further, the quinoline and [3*H*]benzo(*g*)quinoline ring and its derivatives are known for their diverse biological activities and its analogues behaved as agonist at this D_2-site in schizophrenia[13] and several have been evaluated for their antimicrobial activity.[14]

In a recent communication, we have reported the Lewis acid catalysed IDA reactions of 1-substituted unactivated butadienes with *N*-aryl imines.[15,16] The reactions were shown to result in the chemo- as well as regioselective formation of 5-vinyl quinoline substituted pyrimidinone derivatives in good yields. Herein, we report a detailed account of chemo- as well as regioselective transition metal salt catalysed IDA cycloaddition reactions of 5-dienyl pyrimidinone derivatives with *N*-aryl and previously unexplored *N*-naphthyl imines in the presence of the different Lewis acids.

Lewis acids such as magnesium(II) bromide, zinc(II) chloride, indium(III) chloride, yttrium triflate and scandium triflate were selected and examined for their effects on the chemo- and regioselectivities of the reactions. The treatment of **1a–c** in the presence of different Lewis acids resulted in the chemo- as well as regioselective formation of fluorescent 5-vinyl quinoline ring substituted pyrimidinone derivatives **4a–o** in good yields. The formation of such pyrimidinone derivatives in these reactions is presumably the result of usual oxidative aromatisation of the imino DA adducts **3**, as intermediate initially formed via participation of *N*-aryl imines and dienyl pyrimidinones as 4π and 2π components, respectively (Scheme 1). Interestingly, the use of all these Lewis acids

* Corresponding author. Tel.: +91 0183 2258802 09x3320; fax: +91 0183 2258819 20.
E-mail address: mahajanmohinderp@yahoo.co.in (M.P. Mahajan).

0040-4020/$ - see front matter © 2008 Elsevier Ltd. All rights reserved.
doi:10.1016/j.tet.2008.01.041



Scheme 1.

promoted remarkably high chemo- as well as regioselectivity and high resolution ^{1}H NMR spectra (300 MHz) of even crude reaction mixtures of the adducts did not show the presence of detectable amount of any other isomer (Scheme 1).

The reactions of **1a**–**c** with **2a**–**d** in the presence of yttrium triflate (Table 1; entries 4, 9, 14, 19) and indium(III) chloride (Table 1; entries 3, 8, 13, 18) in dry acetonitrile resulted in the formation of corresponding 5-quinoline substituted pyrimidinones derivatives **4a**–**l** in comparatively lower yields. The comparative increase in the yields of the corresponding 5-vinyl quinoline substituted pyrimidinone derivatives **4a**–**l** was noticed when scandium triflate was used as catalyst (Table 1; entries 5, 10, 15, 20). The reactions of **1a**–**c** with **2a**–**d** in the presence of zinc(II) chloride resulted in the poor to moderate yields of the products **4a**–**l** (Table 1; entries 2, 7, 12, 17).

Best results in terms of yields and selectivity were obtained with the use of magnesium(II) bromide as catalyst (Table 1; entries 1, 6, 11, 16). The reactions of **1a**–**c** with anisidine derived imines **2a**–**d**, in the presence of magnesium(II) bromide resulted in the exclusive formation of 5-vinyl quinoline substituted pyrimidinone derivatives **4a**–**n** in good yields. The overall comparative increase in yields was also noticed when 6-morpholine substituted 5-dienyl pyrimidinone **1c**, was used as a dienophile (Table 1; entries 1–20).

Table 1
Lewis acid catalysed reactions of **1a–c** with different imines **2a–d**[17]

Entry	Pyrimidinone	Imine	LA	Overall yield (%)
1	**1a/b/c**	**2a**	$MgBr_2$	83/81/88
2	**1a/b/c**	**2a**	$ZnCl_2$	43/44/49
3	**1a/b/c**	**2a**	$InCl_3$	69/65/72
4	**1a/b/c**	**2a**	$Y(OTf)_3$	72/73/79
5	**1a/b/c**	**2a**	$Sc(Tf)_3$	75/74/81
6	**1a/b/c**	**2b**	$MgBr_2$	82/88/90
7	**1a/b/c**	**2b**	$ZnCl_2$	43/52/60
8	**1a/b/c**	**2b**	$InCl_3$	63/62/62
9	**1a/b/c**	**2b**	$Y(OTf)_3$	68/72/78
10	**1a/b/c**	**2b**	$Sc(Tf)_3$	68/70/74
11	**1a/b/c**	**2c**	$MgBr_2$	62/86/90
12	**1a/b/c**	**2c**	$ZnCl_2$	45/42/51
13	**1a/b/c**	**2c**	$InCl_3$	65/61/70
14	**1a/b/c**	**2c**	$Y(OTf)_3$	60/67/74
15	**1a/b/c**	**2c**	$Sc(Tf)_3$	66/61/76
16	**1a/b/c**	**2d**	$MgBr_2$	75/77/86
17	**1a/b/c**	**2d**	$ZnCl_2$	42/43/48
18	**1a/b/c**	**2d**	$InCl_3$	71/70/78
19	**1a/b/c**	**2d**	$Y(OTf)_3$	65/69/78
20	**1a/b/c**	**2d**	$Sc(Tf)_3$	67/71/84

The reactions of **1a–c** with *N*-benzylideneaniline **2e** resulted in poor yields of the corresponding imino Diels–Alder cycloadducts **4m–o** with the use of the traditional Lewis acid (Scheme 1, Table 2; entries 1–5). However, these reactions when performed in the presence of magnesium(II) bromide containing catalytic amount of aluminium(III) chloride resulted in the isolation of 5-vinyl quinoline substituted pyrimidinone derivatives **4m–o** in good yields (Scheme 1, Table 2; entry 6). This is reported earlier probably due to the formation of an active $MgBr^+$ cation, which is sufficiently acidic to activate the imines (Eq. 1).

$$AlCl_3 + MgBr_2 \rightarrow [MgBr]^+[AlCl_3Br]^- \quad (1)$$

Its worth mentioning that the IDA reactions described above did not proceed in the presence of stoichiometric amount of aluminium(III) chloride possibly due its strong acidity, leading to the hydrolysis of the imines.

The 5-vinyl quinoline substituted pyrimidinone derivatives **4a–n** were characterised with the help of analytical data and spectral evidences, the details of which are described in Section 2.

In continuation of these studies and in an attempt to generalise and to firmly establish the dienophilic character of the terminal double bond of the 5-dienyl pyrimidinones in imino Diels–Alder cycloaddition reactions, we have also examined their reactions with *N*-naphthyl imine **5** in the presence of different Lewis acids. All these reactions as envisaged resulted in novel 5-vinyl benzoquinoline ring substituted pyrimidinone derivatives **7a–c** in fair to moderate yields. The pyrimidinone derivatives are once again presumably to be formed by the usual oxidative aromatisation of the imino DA adducts **6**, formed initially via participation of *N*-naphthyl imines and dienyl pyrimidinones as 4π and 2π components, respectively (Scheme 2).

Table 2
Lewis acid catalysed reactions of **1a–c** with benzylideneaniline **2e**[17]

Entry	Pyrimidinone	Imine	Lewis acid	Yield (%)
1	**1a/b/c**	**2e**	$MgBr_2$	8/5/10
2	**1a/b/c**	**2e**	$ZnCl_2$	9/10/12
3	**1a/b/c**	**2e**	$InCl_3$	11/15/14
4	**1a/b/c**	**2e**	$Y(OTf)_3$	16/19/19
5	**1a/b/c**	**2e**	$Sc(OTf)_3$	14/16/20
6	**1a/b/c**	**2e**	$MgBr_2$/cat. $AlCl_3$	74/75/75

Here again, the use of traditional transition metal salts resulted in the moderate yields of the corresponding imino Diels–Alder cycloadducts (Table 3; entries 1–5). The reactions failed in the presence of zinc(II) chloride and aluminium(III) chloride as catalysts (Table 3; entry 2) and resulted in poor yields of corresponding products **7** in the presence of magnesium(II) chloride and indium(III) chloride (Table 3; entries 1, 3) while moderate yields of the **7a–c** were obtained by the use of scandium and yttrium triflate (Table 3; entries 4, 5). However, fairly good yields of the products **7a–f** were obtained in the presence of magnesium(II) bromide containing catalytic amount of aluminium(III) chloride in dry dichloromethane as catalyst (Table 3; entry 6).

The 5-vinyl benzoquinoline substituted pyrimidinone derivatives **7a–c** were characterised with the help of analytical data and spectral evidences, the details of which are described in Section 2.

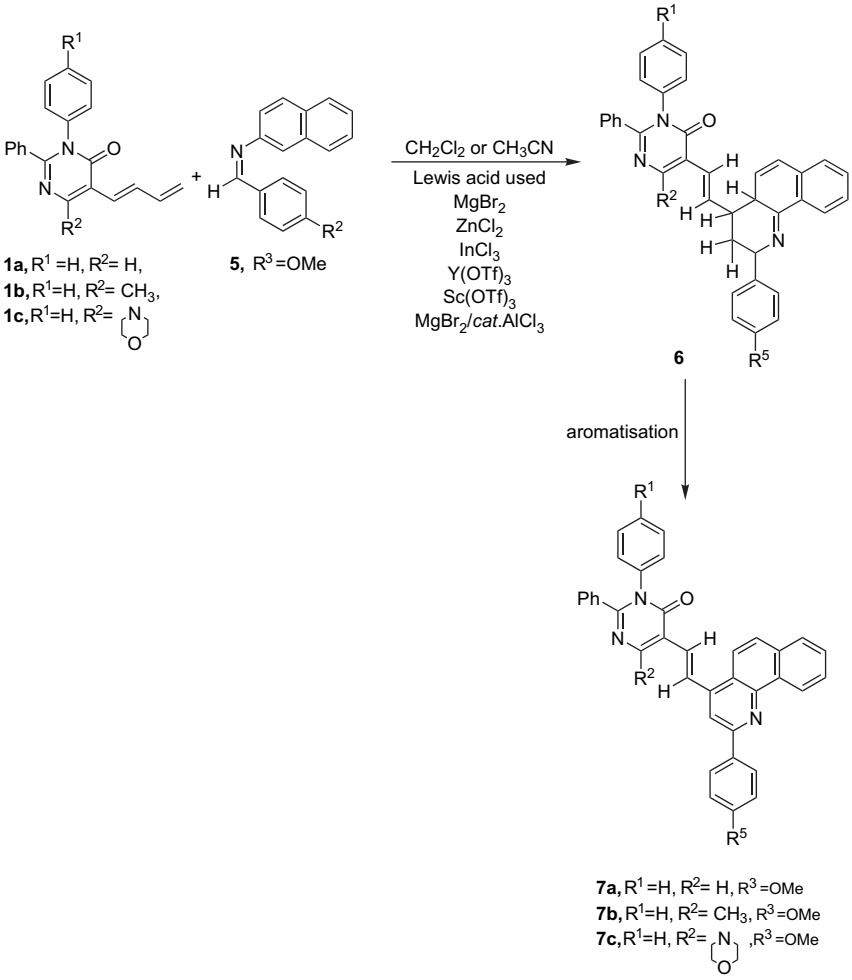
In conclusion, a Lewis acid/mixed Lewis acid mediated imino DA reaction of unactivated *N*-aryl imines with unactivated acyclic 1,3-butadienes resulting in good yields of a variety of novel quinoline, benzoquinoline/pyrimidinone derivatives is reported. A possible reason for the unusual 2π-behaviour of 1-substituted butadienes in such aza Diels–Alder cycloaddition reactions may be due to the steric interaction expected between the pyrimidinone ring and the aryl group present at the nitrogen of the *N*-aryl imines in normal DA adducts when diene group participates as a 4π component. Alternatively, it may be due to activation of the terminal double bond by the slight polarisation of the dienyl group involving the non-bonding electrons present on N^3 of the pyrimidinone ring. It is also likely that the ring closure leading to the formation of 5-quinoline substituted pyrimidinone derivatives may be the result of a sequence of electrophilic and substitution reactions (Scheme 3). In this scheme it is assumed that the path-a is favoured and path-b is ruled out owing to the strong steric interaction between the nitrogen substituents of the *N*-aryl/naphthyl imines with pyrimidinone ring (Scheme 3).

In order to distinguish between these mechanistic possibilities, further work is in progress in our laboratory.

2. Experimental

2.1. General

Melting points were determined by open capillary using Veego Precision Digital Melting Point apparatus (M.P-D) and are uncorrected. IR spectra were recorded on a Shimadzu



Scheme 2.

D-8001 spectrophotometer. ^{1}H NMR spectra were recorded in deuterochloroform with Jeol JE 300 (300 MHz) spectrometers using TMS as internal standard. Chemical shift values are expressed as parts per million downfield from TMS and *J* values are in hertz. Splitting patterns are indicated as s: singlet, d: doublet, t: triplet, m: multiplet, q: quartet and br: broad peak. ^{13}C NMR spectra were also recorded on Jeol JE 300 (75.4 MHz) spectrometers in deuterochloroform using TMS as internal standard. Mass spectra were recorded on Shimadzu GC–MS-QP-2000 mass spectrometer. Elemental analyses were performed on Heraus CHN-O-Rapid Elemental Analyzer. Column chromatography was performed on a silica gel (60–120) mesh.

Table 3
Lewis acid catalysed reactions of **1a–c** with *N*-naphthyl imine **5**[17]

Entry	Pyrimidinone	Imine	Lewis acid	Yield (%)
1	**1a/b/c**	**5**	$MgBr_2$	12/18/16
2	**1a/b/c**	**5**	$ZnCl_2$	—
3	**1a/b/c**	**5**	$InCl_3$	15/28/30
4	**1a/b/c**	**5**	$Y(OTf)_3$	37/28/32
5	**1a/b/c**	**5**	$Sc(OTf)_3$	35/31/35
6	**1a/b/c**	**5**	$MgBr_2$/cat. $AlCl_3$	68/70/64

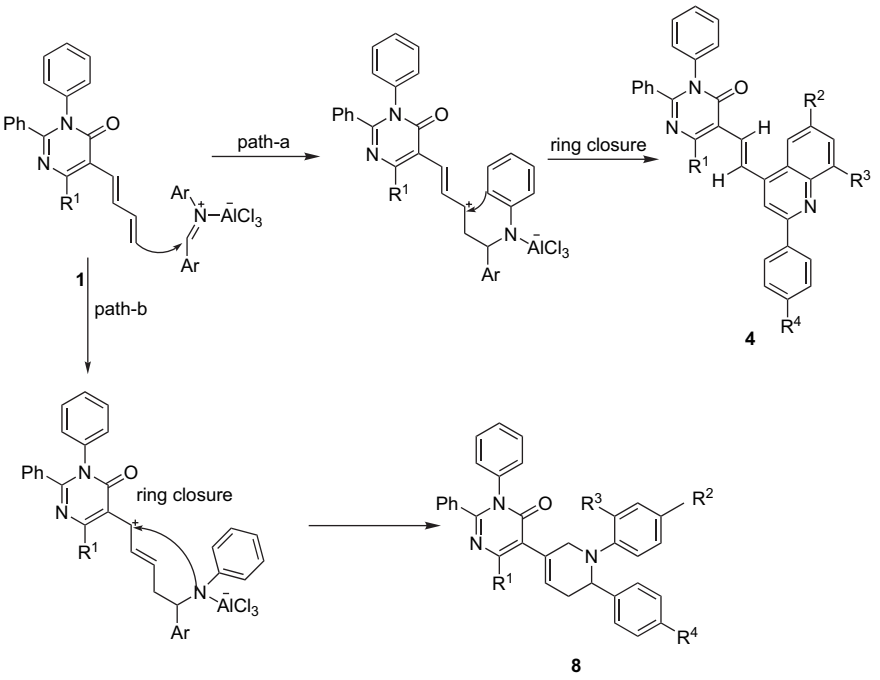
2.2. General procedure

2.2.1. General procedure using simple metal catalysts

The procedure for imino Diels–Alder reactions involved the addition of Lewis acid (15 mmol %) to a well-stirred solution of imines (2 mmol) in dry dichloromethane (20 mL) or acetonitrile at the reaction temperature. The solution was allowed to stir for 20 min followed by the addition of diene (1 mmol). The progress of the reaction was monitored by TLC taking diene as the limiting reactant. The reaction mixture was quenched with the addition of solution of water and methanol. The resulted solution was extracted with dichloromethane and dried over sodium sulfate and concentrated under reduced pressure. The crude products obtained were recrystallised from ethyl acetate–hexane mixture (1:5).

2.2.2. General procedure using mixed metal catalyst ($MgBr_2$/cat. $AlCl_3$)

The procedure involved the addition of magnesium(II) bromide (15 mol %) to a well-stirred solution of imines (2 mmol) in dry dichloromethane (20 ml) at the reaction temperature. The solution was allowed to stir for 5 min followed by the addition of catalytic amount of aluminium(III) chloride



Scheme 3.

(2 mol %). The reaction was allowed to stir further for 20 min followed by addition of diene (1 mmol). The progress of the reaction was monitored by TLC taking diene as the limiting reactant. The reactions mixture was quenched with the addition of solution of water and methanol. The resulted solution was extracted with dichloromethane and dried over sodium sulfate and concentrated under reduced pressure. The crude products obtained were recrystallised from ethyl acetate–hexane mixture (1:5).

2.2.2.1. 5-[2-(6-Methoxy-2-phenyl-quinolin-4-yl)-vinyl]-2,3-diphenyl-3H-pyrimidin-4-one ***4a***. Yellow solid, mp 145–146 °C; δ_H ($CDCl_3$, 600 MHz): 3.92 (s, 3H, $-OCH_3$), 7.11 (d, J=15.8 Hz, 1H, H_8), 7.14–7.29 (m, 15H, aromatic), 7.35 (dd, J=2.1, 7.9 Hz, 1H, H_{14}), 7.49 (d, J=2.1 Hz, 1H, H_{16}), 8.03 (s, 1H, pyrimidinone), 8.16 (d, J=7.9 Hz, 1H, H_{13}), 8.32 (s, 1H, H_{10}), 8.7 (d, J=15.8 Hz, 1H, H_7); δ_C ($CDCl_3$, 75 MHz): 55.7, 101.9, 114.2, 120.0, 123.5, 124.5, 126.5, 127.5, 127.9, 128.7, 128.5, 128.8, 129.0, 129.3, 129.7, 129.9, 131.4, 134.5, 135.8, 137.3, 138.7, 144.6, 144.9, 153.2, 155.69, 157.9, 161.7, 162.8; *m/z* 507 (M^+); ν_{max} (KBr)/cm^{-1} 1688, 1608, 1544, 1485, 1246, 1174. Anal. Calcd for $C_{34}H_{25}N_3O_2$: C, 80.45; H, 4.96; N, 8.28%. Found: C, 80.59; H, 5.12; N, 8.17%.

2.2.2.2. 5-{2-[6-Methoxy-2-(4-methoxy-phenyl)-quinolin-4-yl]-vinyl}-2,3-diphenyl-3H-pyrimidin-4-one ***4b***. Yellow solid, mp 153–154 °C; δ_H ($CDCl_3$, 300 MHz): 3.90 (s, 3H, $-OCH_3$), 3.94 (s, 3H, $-OCH_3$), 7.15 (d, J=16.0 Hz, 1H, H_8), 7.22–7.38 (m, 14H, aromatic), 7.41 (dd, J=2.3, 8.2 Hz, 1H, H_{14}), 7.53 (d, J=2.3 Hz, 1H, H_{16}), 8.1 (s, 1H, pyrimidinone), 8.19 (d, J=8.2 Hz, 1H, H_{13}), 8.29 (s, 1H, H_{10}), 8.63 (d, J=16.0 Hz, 1H, H_7); δ_C ($CDCl_3$, 75 MHz): 55.2, 55.4, 99.2, 110.2, 121.0, 122.3, 123.8, 126.2, 126.5, 127.5, 128.3, 128.7, 129.1, 129.30, 129.5, 130.1, 130.8, 130.9, 133.8, 136.2, 138.0, 138.2, 145.1, 145.7, 153.5, 155.8, 158.2, 160.3, 161.4; *m/z* 537 (M^+); ν_{max} (KBr)/cm^{-1} 1682, 1601, 1559, 1502, 1229, 1179. Anal. Calcd for $C_{35}H_{27}N_3O_3$: C, 78.19; H, 5.06; N, 7.82%. Found: C, 78.31; H, 5.20; N, 7.93%.

2.2.2.3. 5-{2-[2-(4-Chloro-phenyl)-6-methoxy-quinolin-4-yl]-vinyl}-2,3-diphenyl-3H-pyrimidin-4-one ***4c***. Yellow solid, mp 166–167 °C; δ_H ($CDCl_3$, 300 MHz): 3.83 (s, 3H, $-OCH_3$), 7.09 (d, J=15.8 Hz, 1H, H_8), 7.15–7.30 (m, 14H, aromatic), 7.47 (dd, J=2.2, 8.7 Hz, 1H, H_{14}), 7.56 (d, J=2.2 Hz, 1H, H_{16}), 8.08 (s, 1H, pyrimidinone), 8.23 (d, J=8.7 Hz, 1H, H_{13}), 8.32 (s, 1H, H_{10}), 8.53 (d, J=15.8 Hz, 1H, H_7); δ_C ($CDCl_3$, 75 MHz): 55.8, 107.2, 114.2, 121.8, 122.6, 123.9, 126.8, 127.1, 127.8, 128.0, 128.2, 128.9, 129.5, 130.2, 130.7, 131.5, 132.0, 135.2, 137.7, 137.8, 138.1, 145.2, 145.5, 154.1, 155.2, 158.1, 160.1, 160.7; *m/z* 541 (M^+); ν_{max} (KBr)/cm^{-1} 1670, 1598, 1523, 1483, 1246, 1231. Anal. Calcd for $C_{34}H_{24}ClN_3O_2$: C, 75.34; H, 4.46; N, 7.75%. Found: C, 75.46; H, 4.59; N, 7.60%.

2.2.2.4. 5-[2-(8-Methoxy-2-phenyl-quinolin-4-yl)-vinyl]-2,3-diphenyl-3H-pyrimidin-4-one ***4d***. Yellow solid, mp 180–181 °C; δ_H ($CDCl_3$, 300 MHz): 3.95 (s, 3H, $-OCH_3$), 7.06 (d, J=7.74 Hz, 1H, H_{14}), 7.12 (d, J=15.8 Hz, 1H, H_8), 7.18–7.44 (m, 15H, aromatic), 7.81 (d, J=7.74 Hz, 1H, H_{16}), 7.53 (t, J=7.68 Hz, 1H, H_{15}), 8.0 (s, 1H, pyrimidinone), 8.15 (s, 1H, H_{10}), 8.81 (d, J=15.8 Hz, 1H, H_7); δ_C ($CDCl_3$, 75 MHz): 55.2, 101.2, 115.0, 116.4, 121.0, 124.5, 127.0, 127.4, 128.4, 128.3, 128.7, 129.2, 129.6, 130.5, 131.4, 132.5, 133.1, 136.8, 137.1, 137.3, 138.4, 145.8, 145.4, 153.9, 155.7, 158.5, 160.4, 161.5; *m/z* 507 (M^+); ν_{max} (KBr)/cm^{-1} 1664, 1534, 1509, 1323. Anal. Calcd for $C_{34}H_{25}N_3O_2$: C, 80.45; H, 4.96; N, 8.28%. Found: C, 80.53; H, 5.07; N, 8.17%.

2.2.2.5. 5-[2-(6-Methoxy-2-phenyl-quinolin-4-yl)-vinyl]-6-methyl-2,3-diphenyl-3H-pyrimidin-4-one ***4e***. Yellow solid, mp 195–196 °C; δ_H ($CDCl_3$, 200 MHz): 2.7 (s, 3H, −Py CH_3), 3.94 (s, 3H, $-OCH_3$), 7.09 (d, J=15.7 Hz, 1H, H_8), 7.17–7.36 (m, 15H, aromatic), 7.86–7.91 (m, 3H, $H_{13,14,16}$), 8.11 (s, 1H, H_{10}), 8.79 (d, J=15.7 Hz, 1H, H_7); δ_C ($CDCl_3$, 75 MHz): 22.7, 55.8, 99.8, 114.3, 121.5, 122.8, 125.2, 127.3, 127.9, 128.0, 128.6, 128.9, 129.3, 129.4, 129.3, 130.2, 130.8, 131.5, 134.5, 137.3, 140.0, 144.1, 144.3, 144.7, 154.8, 156.3, 157.2, 160.8, 161.1; *m/z* 521 (M^+); ν_{max} (KBr)/cm^{-1}1679, 1522, 1233, 1156. Anal. Calcd for $C_{35}H_{27}N_3O_2$: C, 80.59; H, 5.22; N, 8.06%. Found: C, 80.72; H, 5.36; N, 8.19%.

2.2.2.6. 5-{2-[6-Methoxy-2-(4-methoxy-phenyl)-quinolin-4-yl]-vinyl}-6-methyl-2,3-diphenyl-3H-pyrimidin-4-one ***4f***. Yellow solid, mp 194–195 °C; δ_H ($CDCl_3$, 200 MHz): 2.7 (s, 3H, −Py CH_3), 3.82 (s, 3H, $-OCH_3$), 3.90 (s, 3H, $-OCH_3$), 7.12 (d, J=15.0 Hz, 1H, H_8), 7.23–7.55 (m, 14H, aromatic), 7.87–7.94 (m, 3H, $H_{13,14,16}$), 8.07 (s, 1H, H_{10}), 8.93 (d, J=15.0 Hz, 1H, H_7); δ_C ($CDCl_3$, 75 MHz): 22.5, 54.7, 55.8, 100.2, 116.3, 121.3, 122.4, 125.6, 127.8, 127.3, 128.7, 128.9, 129.2, 129.5, 129.8, 130.2, 130.8, 131.1, 132.8, 134.3, 136.2, 136.8, 137.4, 146.1, 146.9, 155.0, 155.9, 159.6, 160.0, 160.4; *m/z* 551 (M^+); ν_{max} (KBr)/cm^{-1} 1681, 1548, 1432, 1473, 1400, 1546. Anal. Calcd for $C_{36}H_{29}N_3O_3$: C, 78.38; H, 5.30; N, 7.62%. Found: C, 78.49; H, 5.42; N, 7.51%.

2.2.2.7. 5-{2-[2-(4-Chloro-phenyl)-6-methoxy-quinolin-4-yl]-vinyl}-6-methyl-2,3-diphenyl-3H-pyrimidin-4-one ***4g***. Yellow solid, mp 173–174 °C; δ_H ($CDCl_3$, 600 MHz): 2.7 (s, 3H, −Py CH_3), 3.94 (s, 3H, $-OCH_3$), 7.20 (d, J=15.71 Hz, 1H, H_8), 7.29–7.44 (m, 10H, aromatic), 7.80 (m, 2H, $H_{14,16}$), 7.50 (d, J=9.2 Hz, 2H, H_{20}), 7.9 (s, 1H, H_{10}), 8.05 (d, J=9.4 Hz, 1H, H_{13}), 8.12 (d, J=9.2 Hz, 2H, H_{19}), 8.72 (d, J=15.7 Hz, 1H, H_7); δ_C ($CDCl_3$, 75 MHz): 22.7, 55.7, 102.0, 115.2, 118.2, 122.5, 126.7, 126.9, 128.1, 128.4, 128.6, 128.7, 128.8, 128.9, 129.1, 129.2, 129.9, 130.5, 131.7, 134.1, 135.3, 138.2, 146.5, 144.9, 153.2, 156.4, 157.8, 161.0, 161.2; *m/z* 555 (M^+); ν_{max} (KBr)/cm^{-1} 1675, 1619, 1531, 1235, 1211, 1129. Anal. Calcd for $C_{35}H_{26}ClN_3O_2$: C, 75.60; H, 4.71; N, 7.56%. Found: C, 75.77; H, 4.84; N, 7.68%.

2.2.2.8. 5-[2-(8-Methoxy-2-phenyl-quinolin-4-yl)-vinyl]-6-methyl-2,3-diphenyl-3H-pyrimidin-4-one ***4h***. Yellow solid, mp 179–180 °C; δ_H ($CDCl_3$, 300 MHz): 2.7 (s, 3H, Py−CH_3), 3.92 (s, 3H, $-OCH_3$), 7.09 (d, J=7.2 Hz, 1H, H_{14}), 7.21 (d, J=15.3 Hz, 1H, H_8), 7.22–7.43 (m, 15H, H, aromatic), 7.53 (unresolved d, J=7.2 Hz, 1H, H_{15}), 7.81 (d, J=7.2 Hz, 1H, H_{16}), 7.95 (s, 1H, H_{10}), 8.92 (d, J=15.3 Hz, 1H, H_7); δ_C ($CDCl_3$, 75 MHz): 22.7, 55.2, 107.2, 115.4, 115.9, 118.2, 126.2, 126.6, 127.7, 128.09, 128.7, 128.8, 129.1, 129.13, 129.2, 129.8, 130.2, 134.5, 137.3, 140.1, 140.7, 144.3, 144.8, 145.0, 155.7, 156.0, 153.6, 161.0, 161.1; *m/z* 521 (M^+); ν_{max} (KBr)/cm^{-1} 1684, 1643, 1548, 1506. Anal. Calcd for $C_{35}H_{27}N_3O_2$: C, 80.59; H, 5.22; N, 8.06%. Found: C, 80.67; H, 5.39; N, 7.95%.

2.2.2.9. 5-[2-(6-Methoxy-2-phenyl-quinolin-4-yl)-vinyl]-6-morpholin-4-yl-2,3-diphenyl-3H-pyrimidin-4-one ***4i***. Red solid, mp 146–147 °C; δ_H ($CDCl_3$, 600 MHz): 3.75 (m, 4H, H, morpholine), 3.84 (m, 4H, H, morpholine), 3.91 (s, 3H, $-OCH_3$), 7.18 (d, J=16.3 Hz, 1H, H_8), 7.19–7.32 (m, 15H, aromatic), 7.35 (dd, J=2.7, 9.0 Hz, 1H, H_{14}), 7.41 (d, J=2.7 Hz, 1H, H_{16}), 7.58 (d, J=9.0 Hz, 1H, H_{13}), 7.87 (s, 1H, H_{10}), 8.49 (d, J=16.3 Hz, 1H, H_7); δ_C (150 MHz, $CDCl_3$): 49.8 (−C morpholine), 55.7 ($-OCH_3$), 67.1 (−C morpholine), 99.2, 101.9, 114.1, 122.1, 124.6, 126.4, 127.3, 127.9, 128.4, 128.5, 128.9, 129.0, 129.0, 129.2, 129.9, 131.4, 134.6, 134.8, 137.5, 138.6, 144.5, 144.7, 153.5, 155.6, 157.7, 161.7, 162.7; *m/z* 592 (M^+); ν_{max} (KBr)/cm^{-1} 1665, 1702, 1492, 1385. Anal. Calcd for $C_{38}H_{32}N_4O_3$: C, 77.01; H, 5.44; N, 9.45%. Found: C, 77.19; H, 5.56; N, 9.56%.

2.2.2.10. 5-{2-[6-Methoxy-2-(4-methoxy-phenyl)-quinolin-4-yl]-vinyl}-6-morpholin-4-yl-2,3-diphenyl-3H-pyrimidin-4-one ***4j***. Fluorescent yellow solid, mp 203–204 °C; δ_H ($CDCl_3$, 600 MHz): 3.70 (m, 4H, morpholine), 3.89 (m, 4H, morpholine), 3.83 (s, 3H, $-OCH_3$), 3.90 (s, 3H, OCH_3), 7.16 (d, J=15.9 Hz, 1H, H_8), 7.19–7.25 (m, 14H, aromatic), 7.27 (dd, J=2.5, 9.2 Hz, 1H, H_{14}), 7.53 (d, J=2.5 Hz, 1H, H_{16}), 7.59 (d, 1H, J=9.2 Hz, H_{13}), 8.15 (s, 1H, H_{10}), 8.43 (d, 1H, J=15.9 Hz, H_7); ^{13}C NMR (75 MHz, $CDCl_3$): δ 49.4 (−C morpholine), 55.0 ($-OCH_3$), 55.3 ($-OCH_3$), 67.2 (−C morpholine), 99.3, 102.3, 122.1, 124.6, 126.4, 127.3, 128.0, 128.1, 128.3, 128.6, 128.9, 129.0, 129.0, 129.2, 129.9, 131.4, 134.6, 134.8, 137.5, 138.6, 144.5, 144.7, 153.5, 155.6, 157.2, 160.8, 162.1; *m/z* 622 (M^+); ν_{max} (KBr)/cm^{-1} 1663, 1583, 1510, 1430, 1133. Anal. Calcd for $C_{39}H_{34}N_4O_4$: C, 75.22; H, 5.50; N, 9.00%. Found: C, 75.36; H, 5.63; N, 8.94%.

2.2.2.11. 5-{2-[2-(4-Chloro-phenyl)-6-methoxy-quinolin-4-yl]-vinyl}-6-morpholin-4-yl-2,3-diphenyl-3H-pyrimidin-4-one ***4k***. Dark yellow solid, mp 158–159 °C; δ_H ($CDCl_3$, 600 MHz): 3.71–3.73 (m, 4H, morpholine), 3.81–3.83 (m, 4H, morpholine), 3.93 (s, 3H, $-OCH_3$), 7.11 (d, 1H, J=16.4 Hz, H_8), 7.21–7.28 (m, 14H, aromatic), 7.30 (dd, J=2.5, 9.1 Hz, 1H, H_{14}), 7.49 (d, J=2.5 Hz, 1H, H_{16}), 7.50 (d, J=9.1 Hz, 1H, H_{13}), 7.92 (s, 1H, H_{10}), 8.55 (d, J=16.4 Hz, 1H, H_7); ^{13}C NMR (75 MHz, $CDCl_3$): δ 49.8 (C morpholine), 55.7 ($-OCH_3$), 67.1 (C morpholine), 99.3, 102.3, 114.9, 122.1, 124.6, 126.4, 127.3, 128.0, 128.1, 128.3, 128.6, 129.0, 129.0, 129.2, 129.9, 131.4, 134.6, 134.8, 137.5, 138.6, 144.5, 144.7, 153.5, 155.6, 157.2, 160.8, 162.5; *m/z* 626 (M^+); ν_{max} (KBr)/cm^{-1} 1650, 1620, 1581, 1544, 1488, 1417, 1232, 1120. Anal. Calcd for $C_{38}H_{31}ClN_4O_3$: C, 72.78; H, 4.98; Cl, 5.65; N, 8.93%. Found: C, 72.89; H, 5.12; N, 8.81%.

2.2.2.12. 5-[2-(8-Methoxy-2-phenyl-quinolin-4-yl)-vinyl]-6-morpholin-4-yl-2,3-diphenyl-3H-pyrimidin-4-one ***4l***. Yellow solid, mp 177–178 °C; δ_H ($CDCl_3$, 200 MHz): 3.73–3.74 (m, 4H, morpholine), 3.84–3.85 (m, 4H, morpholine), 3.94 (s, 3H, $-OCH_3$), 7.13 (d, J=7.8 Hz, 1H, H_{14}), 7.28 (d, J=16.0 Hz, 1H, H_8), 7.30–7.48 (m, 15H, aromatic), 7.53 (m, 1H, H_{15}), 7.81 (d, J=7.8 Hz, 1H, H_{16}), 7.97 (s, 1H, H_{10}),

8.78 (d, J=16.0 Hz, 1H, H_7); δ_C ($CDCl_3$, 75 MHz): 49.8, 55.4, 67.6, 100.1, 102.3, 115.7, 122.4, 124.8, 125.3, 126.4, 126.7, 127.5, 128.1, 128.3, 128.5, 128.9, 129.0, 129.2, 129.8, 131.2, 135.4, 137.0, 138.5, 142.0, 143.9, 153.4, 154.3, 156.2, 160.8, 162.2; *m/z* 592 (M^+); ν_{max} (KBr)/cm^{-1} 1659, 1603, 1527, 1400, 1193. Anal. Calcd for $C_{38}H_{32}N_4O_3$: C, 77.01; H, 5.44; N, 9.45%. Found: C, 77.21; H, 5.58; N, 9.55%.

2.2.2.13. 2,3-Diphenyl-5-[2-(2-phenyl-quinolin-4-yl)-vinyl]-3H-pyrimidin-4-one ***4m***. Yellow solid, mp 162–163 °C; δ_H ($CDCl_3$, 200 MHz): 7.38–7.53 (m, 16H, aromatic and $1H_8$), 8.02 (s, 1H, H_{10}), 8.12 (s 1H, Py–H), 8.24–8.35 (m, 4H, $H_{13,14,15,16}$), 8.74 (d, J=15.4 Hz, 1H, H_7); δ_C ($CDCl_3$, 75 MHz): 101.7, 114.9, 120.8, 122.4, 125.3, 126.9, 127.2, 127.8, 128.1, 128.5, 128.72, 128.75, 128.9, 129.0, 129.6, 129.8, 130.2, 131.8, 134.9, 137.7, 140.0, 143.5, 144.8, 157.4, 157.8, 160.2, 161.3; *m/z* 477 (M^+); ν_{max} (KBr)/cm^{-1} 1674, 1536, 1502, 1431. Anal. Calcd for $C_{33}H_{23}N_3O$: C, 83.00; H, 4.85; N, 8.80%. Found: C, 83.16; H, 4.99; N, 8.65%.

2.2.2.14. 6-Methyl-2,3-diphenyl-5-[2-(2-phenyl-quinolin-4-yl)-vinyl]-3H-pyrimidin-4-one ***4n***. Yellow solid, mp 165–166 °C; δ_H ($CDCl_3$, 200 MHz): 2.70 (s, 3H, Py CH_3), 7.15–7.74 (m, 16H, 15H, aromatic and $1H_7$), 8.10 (s, 1H, H_{10}), 8.10–8.27 (m, 4H, $H_{13,14,15,16}$), 8.99 (d, J=15.8 Hz, 1H, H_8); δ_C ($CDCl_3$, 75 MHz): 22.72, 98.4, 107.3, 115.8, 120.4, 126.3, 126.9, 127.4, 127.5, 128.3, 128.9, 129.0, 129.1, 129.3, 129.9, 130.1, 133.4, 135.6, 137.0, 137.8, 140.3, 140.8, 145.2, 155.6, 157.8, 158.3, 161.2, 161.4; *m/z* 491 (M^+); ν_{max} (KBr)/cm^{-1} 1653, 1566, 1509, 1421, 1361. Anal. Calcd for $C_{34}H_{25}N_3O$: C, 83.07; H, 5.13; N, 8.55%. Found: C, 83.19; H, 5.30; N, 8.42%.

2.2.2.15. 6-Morpholin-4-yl-2,3-diphenyl-5-[2-(2-phenyl-quinolin-4-yl)-vinyl]-3H-pyrimidin-4-one ***4o***. Yellow solid, mp 171–172 °C; δ_H ($CDCl_3$, 200 MHz): 3.72–3.74 (m, 4H, morpholine), 3.88–3.90 (m, 4H, morpholine), 7.14 (d, J=15.9 Hz, 1H, H_8), 7.21–7.82 (m, 15H, H, aromatic), 7.97 (s, 1H, H_{10}), 8.19–8.30 (m, 4H, $H_{13,14,15,16}$), 8.94 (d, J=15.9 Hz, 1H, H_7); δ_C ($CDCl_3$, 75 MHz): 49.5, 67.4, 100.2, 102.4, 115.6, 120.8, 123.9, 125.8, 126.4, 127.4, 127.6, 128.0, 128.3, 128.4, 128.9, 129.1, 129.8, 130.0, 131.3, 134.7, 138.7, 138.9, 144.2, 145.7, 154.2, 155.8, 156.9, 161.2, 162.3; *m/z* 562 (M^+); ν_{max} (KBr)/cm^{-1} 1677, 1603, 1524, 1381, 1209. Anal. Calcd for $C_{37}H_{30}N_4O_2$: C, 78.98; H, 5.37; N, 9.96%. Found: C, 79.11; H, 5.52; N, 9.81%.

2.2.2.16. 5-{2-[2-(4-Methoxy-phenyl)-benzo[h]quinolin-4-yl]-vinyl}-2,3-diphenyl-3H-pyrimidin-4-one ***7a***. Yellow solid, mp 212–213 °C; δ_H ($CDCl_3$, 200 MHz): 3.95 (s, 3H, $-OCH_3$), 7.10 (d, J=8.4 Hz, 2H, aromatic), 7.18–7.40 (m, 11H, aromatic and H_8), 7.45–7.49 (m, 6H, benzoquinoline), 7.93 (s, 1H, H_{10}), 8.12 (s, 1H, Py–H), 8.15 (d, J=8.4 Hz, 2H, aromatic), 8.92 (d, J=15.6 Hz, 1H, H_7); δ_C ($CDCl_3$, 75 MHz): 55.6, 99.2, 113.9, 122.3, 125.4, 125.5, 126.0, 127.3, 128.0, 128.6, 128.65, 128.9, 129.3, 129.5, 129.9, 131.4, 131.9, 134.5, 134.6, 137.8, 138.1, 144.4, 145.1, 151.7, 152.3, 154.2, 154.8, 157.3, 161.2, 163.8; *m/z* 557 (M^+); ν_{max} (KBr)/cm^{-1} 1670, 1565, 1578, 1432, 1256. Anal. Calcd for $C_{38}H_{27}N_3O_2$: C, 81.85; H, 4.88; N, 7.54%. Found: C, 81.98; H, 5.01; N, 7.36%.

2.2.2.17. 5-{2-[2-(4-Methoxy-phenyl)-benzo[h]quinolin-4-yl]-vinyl}-6-methyl-2,3-diphenyl-3H-pyrimidin-4-one ***7b***. Yellow solid, mp 224–225 °C; δ_H ($CDCl_3$, 300 MHz): 2.75 (s, 3H, Py–CH_3), 3.93 (s, 3H, $-OCH_3$), 7.10 (d, J=8.7 Hz, 2H, aromatic), 7.17–7.52 (m, 10H, aromatic), 7.67–7.72 (m, 4H, $H_{13,14,15,16}$), 7.87 (d, J=7.8 Hz, 1H, H_{18}), 8.12 (s, 1H, H_{10}), 8.14 (d, J=7.8 Hz, 1H, H_{17}), 8.16 (d, J=8.7 Hz, 2H, aromatic), 8.38 (d, J=15.8 Hz, 1H, H_8), 8.97 (d, J=15.8 Hz, 1H, H_7); δ_C ($CDCl_3$, 75 MHz): 22.7, 55.4, 100.4, 114.1, 122.8, 125.1, 126.0, 126.8, 127.9, 128.0, 128.3, 128.6, 128.5, 129.2, 129.4, 129.4, 129.7, 130.0, 130.3, 134.3, 135.9, 136.3, 139.1, 144.0, 146.4, 151.2, 151.5, 155.9, 156.1, 157.1, 162.2, 164.1; *m/z* 571 (M^+); ν_{max} (KBr)/cm^{-1} 1662, 1569, 1503, 1240. Anal. Calcd for $C_{39}H_{29}N_3O_2$: C, 81.94; H, 5.11; N, 7.35%. Found: C, 82.11; H, 5.29; N, 7.21%.

2.2.2.18. 5-{2-[2-(4-Methoxy-phenyl)-benzo[h]quinolin-4-yl]-vinyl}-6-morpholin-4-yl-2,3-diphenyl-3H-pyrimidin-4-one ***7c***. Yellow solid, mp 205–206 °C; δ_H ($CDCl_3$, 200 MHz): 3.73–3.75 (m, 4H, morpholine), 3.89–3.91 (m, 4H, morpholine), 3.98 (s, 3H, $-OCH_3$), 7.12 (d, J=8.1 Hz, 2H, aromatic), 7.26–7.30 (m, 11H, H, aromatic and H_8), 7.49–7.52 (m, 4H, benzoquinoline), 7.61–7.64 (m, 2H, benzoquinoline), 8.10 (s, 1H, H_{10}), 8.15 (d, J=8.1 Hz, 2H, aromatic), 8.74 (d, J=16.3 Hz, 1H, H_7); δ_C ($CDCl_3$, 75 MHz): 55.9, 49.7, 67.3, 101.3, 116.4, 122.6, 123.2, 125.3, 125.8, 126.1, 127.4, 128.0, 128.6, 129.1, 129.2, 129.1, 130.2, 130.5, 130.7, 134.2, 134.5, 135.8, 137.4, 140.2, 142.5, 147.8, 149.9, 151.5, 154.1, 155.3, 157.5, 161.3, 165.7; *m/z* 642 (M^+); ν_{max} (KBr)/cm^{-1} 1669, 1538, 1521, 1204. Anal. Calcd for $C_{42}H_{34}N_4O_3$: C, 78.48; H, 5.33; N, 8.72%. Found: C, 78.60; H, 5.47; N, 8.56%.

Acknowledgements

We are thankful to the Prof. Takao Saito, Department of Applied Chemistry, Faculty of Science, Tokyo University of Science, Kagurazaka, Shinjuku-ku, Tokyo 162-8601, Japan for providing 600 MHz NMR spectra.

References and notes

1. For leading references, see: (a) Daly, J. W.; Spande, T. F. *Alkaloids: Chemical and Biological Perspectives*; Pelletier, S. W., Ed.; Wiley: New York, NY, 1986; Vol. 4, pp 1–254; (b) Fodor, G. B.; Colasanti, B. *Alkaloids: Chemical and Biological Perspectives*; Pelletier, S. W., Ed.; Wiley: New York, NY, 1985; Vol. 3, pp 1–91.
2. (a) Boger, D.; Weinreb, S. M. *Hetero Diels–Alder Methodology in Organic Synthesis*; Academic: San Diego, CA, 1987; Chapter 2, pp 34–70; Chapter 9, pp 239–299 and references therein; (b) Boger, D. L.; Patel, M. *Progress in Heterocyclic Chemistry*; Suschitzky, H., Scriven, E. F. V., Eds.; Pergamon: New York, NY, 1989; pp 1–30; (c) Kametani, T.; Hibino, S. *Adv. Heterocycl. Chem.* **1987**, *42*, 246–333.
3. For recent reviews see: (a) Buonora, P.; Olsen, J.-C.; Oh, T. *Tetrahedron* **2001**, *57*, 6099; (b) Jørgensen, K. A. *Angew. Chem., Int. Ed.* **2000**, *39*, 3558 and references cited therein; (c) Orr, R. K.; Calter, M. A. *Tetrahedron* **2003**, *59*, 3545 and references cited therein.

4. For recent examples see: (a) Takasu, K.; Shindoh, N.; Tokuyama, H.; Ihara, M. *Tetrahedron* **2006**, *62*, 11900; (b) Manian, R. D. R. S.; Jayashankaran, J.; Ramesh, R.; Raghunathan, R. *Tetrahedron Lett.* **2006**, *47*, 7571; (c) Yadav, J. S.; Reddy, B. V. S.; Kondaji, G.; Sowjanya, S.; Nagaiah, K. *J. Mol. Catal. A: Chem.* **2006**, *258*, 361; (d) Prenzel, A. H. G. P.; Deppermann, N.; Maison, W. *Org. Lett.* **2006**, *8*, 1681; (e) Zhou, Y.; Jia, X.; Li, R.; Liu, Z.; Liu, Z.; Wu, L. *Tetrahedron Lett.* **2005**, *46*, 8937; (f) Rodrý'guez, M.; Ochoa, M. E.; Santillan, R.; Farfa'n, N.; Barba, V. *J. Organomet. Chem.* **2005**, *690*, 2975; (g) Garcia, A.-B.; Valdes, C.; Cabal, M.-P. *Tetrahedron Lett.* **2004**, *45*, 4357; (h) Danieli, B.; Lesma, G.; Passarella, D.; Piacenti, P.; Sacchetti, A.; Silvani, A.; Virdis, A. *Tetrahedron Lett.* **2002**, *43*, 7155.
5. (a) Tietze, L. F.; Kettschau, G. *Top. Curr. Chem.* **1997**, *189*, 1–120; (b) Kobayashi, S. *Eur. J. Org. Chem.* **1999**, 15–27; (c) Panunzio, M.; Zarantonello, P. *Org. Process Res. Dev.* **1988**, *2*, 49–59; (d) Enders, D.; Meyer, O. *Liebigs Ann. Chem.* **1996**, 1023–1035; (e) Waldmann, H. *Organic Synthesis Highlights II*; Waldman, H., Ed.; VCH: New York, NY, 1995; pp 37–47; (f) Waldmann, H. *Synthesis* **1994**, 535–551; (g) Waldmann, H.; Braun, M. *Gazz. Chim. Ital.* **1991**, *121*, 277–284; (h) Waldmann, H. *Synlett* **1995**, 133–141; (i) Barluenga, J.; Joglar, J.; Gonzalez, F. J.; Fustero, S. *Synlett* **1990**, 129–138; (j) Parker, D. T. *Organic Synthesis in Water*; Grieco, P. A., Ed.; Blackie Academic and Professional: London, 1998; Vol 2, pp 47–81; (k) Barluenga, J.; Tomas, M. *Adv. Heterocycl. Chem.* **1993**, *57*, 1–80.
6. (a) Kobayashi, S.; Ishitani, H.; Nagayama, S. *Synthesis* **1995**, 1195–1202; (b) Kobayashi, S.; Nagayama, S. *J. Am. Chem. Soc.* **1996**, *118*, 8977–8978.
7. (a) Szczech, G. M.; Furman, P.; Painter, G. R.; Barry, D. W.; Borroto-Esoda, K.; Grizzle, T. B.; Blum, M. R.; Sommadossi, J.-P.; Endoh, R.; Niwa, T.; Yamamoto, M.; Moxham, C. *Antimicrob. Agents Chemother.* **2000**, *44*, 123; (b) Dieterle, F.; Müller-Hagedon, S.; Liebich, H. M.; Gauglitz, G. *Artif. Intell. Med.* **2003**, *28*, 265.
8. Rowley, D. C.; Hansen, M. S. T.; Rhodes, D.; Striffer, C. A.; Ni, H.; McCammon, J. A.; Bushman, F. D.; Fenical, W. *Bioorg. Med. Chem.* **2002**, *10*, 3619.
9. (a) Molina, P.; Aller, E.; Lorenzo, A.; Lopez-Cremades, P.; Rioja, I.; Ubeda, A.; Terencio, M. C.; Alcaraz, M. J. *J. Med. Chem.* **2001**, *44*, 1011; (b) Kumar, A.; Sinha, S.; Chauhan, P. M. S. *Bioorg. Med. Chem. Lett.* **2002**, *12*, 667.
10. (a) Bher, J.-B.; Gourlain, T.; Helimi, A.; Guillerm, G. *Bioorg. Med. Chem. Lett.* **2003**, *13*, 1713; (b) De Koning, H. P.; Al-Salabi, M. I.; Cohen, A. M.; Coombs, G. H.; Wastling, J. M. *Int. J. Parasitol.* **2003**, *33*, 821.
11. Hosni, H. M.; Basyouni, W. M.; El-Nahas, H. A. *J. Chem. Res., Synop.* **1999**, 646.
12. Botta, M.; Occhionero, F.; Nicoletti, R.; Mastromarino, P.; Conti, C.; Margini, M.; Saladino, J. *Bioorg. Med. Chem.* **1999**, *9*, 1925.
13. Seeman, P.; Guan, H.-C.; Nobrega, J.; Jiwa, D.; Markstein, R.; Balk, J.-H.; Picetti, R.; Borrelli, E.; Tol, H. H. M. V. *Synapse* **1997**, *25*, 137.
14. Elslager, E. F.; Short, W. F.; Tendick, H. F. *J. Heterocycl. Chem.* **1968**, *5*, 599.
15. Bhargava, G.; Kumar, V.; Mahajan, M. P. *Tetrahedron Lett.* **2007**, *48*, 2365.
16. (a) Goel, R. K.; Kumar, V.; Mahajan, M. P. *Bioorg. Med. Chem. Lett.* **2005**, *15*, 2145; (b) Bedi, P. M. S.; Kumar, V.; Mahajan, M. P. *Bioorg. Med. Chem. Lett.* **2004**, *14*, 5211; (c) Goel, R. K.; Thind, J. S.; Bal, C. S.; Mahajan, M. P.; Kulkarni, S. K. *Die Pharmazie* **2005**, 369–374; (d) Bhargava, G.; Mahajan, M. P.; Saito, T.; Otani, T.; Kurashima, M.; Sakai, K. *Eur. J. Org. Chem.* **2005**, 2397; (e) Mohan, C.; Bhargava, G.; Pannu, A. P. S.; Mahajan, M. P. *Tetrahedron Lett.* **2007**, *48*, 1711.
17. All the reactions were conducted at room temperature, for indium(III) chloride, scandium triflate and yttrium triflate, dry acetonitrile was used as solvent and for magnesium(II) chloride, zinc(II) chloride and $MgBr_2$/cat. $AlCl_3$, dry dichloromethane was used as a solvent.