CHEMO- AND REGIOSELECTIVE IMINO DIELS-ALDER REACTIONS: SYNTHESIS OF FUNCTIONALIZED NOVEL QUINOLIN-3-ONE AND QUINOLINE DERIVATIVES

Chander Mohan, Gaurav Bhargava, and Mohinder P. Mahajan

Department of Applied Chemistry, Guru Nanak Dev University, Amritsar – 143005, Punjab, India. Fax +91(183)225881920; E-mail: mahajanmohinderp@yahoo.co.in

Abstract – Regio- and catalyst selective imino Diels-Alder reactions of vinyl/isopropenyl pyrimidinones with N-arylimines in the presence of metal salts viz. magnesium(II) bromide, zinc(II) chloride, indium(III) chloride, yttrium triflate and scandium triflate, are described. An unprecedented oxidation of methylene to carbonyl has been shown to accompany the imino Diels–Alder reactions of isopropenyl pyrimidinones in the presence of yttrium and scandium triflates. Chemoselective effect of different Lewis acids used as catalysts in these reactions has been rationalized.

INTRODUCTION

The imino-Diels-Alder (IDA) reaction involving the coupling of imines, derived from aromatic amines, or their surrogates, with electron rich alkenes has emerged as a powerful tool for the synthesis of tetrahydroquinolines and allied substrates. Some of these substrates are known for their biologically latency viz. psychotropic, antiallergic, anti-inflammatory and estrogenic activities. There are numerous reports on the IDA reactions of N-arylimines with sterically unhindered activated alkenes, cyclopentadiene or symmetrical activated butadienes catalyzed by a variety of metal salts and protic acids. Such reactions suffered from lack of chemoselectivity and resulted in formation of a mixture of adducts. However, the reports concerning the use of acyclic un-activated and sterically hindered alkenes as dienophiles in such IDA reaction are rare.

In recent communications, we have compared the dienic properties of N-arylimines in their Lewis acid mediated cycloaddition reactions with 5-isopropenyl pyrimidinones and 3-dienyl-2-azetidinones (Figure
1). These reactions were shown to result in the formation of 5-quinoline substituted pyrimidinone/azetidinone derivatives with participation of N-aryl imines as 4π component of 2-azadiene. In continuation of these studies, we report herein a detailed account of chemo- as well as regioselective metal salts or Lewis acid catalysed IDA reactions of 5-isopropenyl/vinyl pyrimidinones with N-arylimines. Interestingly, these reactions resulted in a catalyst selective formation of novel pyrimidinone tethered quinoline, dihydroquinoline and quinolin-3-one derivatives depending upon the nature of the metal salts or Lewis acid employed. Such functionalized pyrimidinones have been reported to show antitumour, antiviral, antitubercular, antifungal, molluscidal and larvacidal activities.

Figure 1 Dienic/dienophilic behaviour of conjugated hindered alkenes and N-aryl-imines towards IDA reaction

RESULTS AND DISCUSSION

Different Lewis acids viz. magnesium(II) bromide, zinc(II) chloride, indium(III) chloride, yttrium triflate and scandium triflate were selected for examination of their effects on the chemo- and regioselectivities of the IDA reactions of 1 with N-arylimines 2.

The reactions of 1a-b with N-arylimines 2a-c in the presence of yttrium triflate as Lewis acid catalyst resulted in the formation of 5-quinoline substituted pyrimidinone derivatives 4a-f in good yields (Table 1, Entries 1-3). The reactions in the presence of scandium triflate, magnesium(II) bromide, zinc (II) chloride and indium(III) chloride (Table 1, Entries 4-15), were found to be slower and required longer reaction periods for their completion with the formation of 4a-f in moderate to good.

The products were characterized on the basis of analytical and spectral data, as described in the experimental section while the salient features are mentioned here. The compound 4a, for example, showed a [M+1]^+ peak at m/z 516 in its mass spectrum. Its IR spectrum showed a strong absorption at 1670 cm\(^{-1}\) due to the carbonyl group of the pyrimidinone ring. Its \(^1\)H NMR (300 MHz) spectrum showed singlets at \(\delta\) 7.97 and \(\delta\) 8.55 assigned to H\(_6\) of the pyrimidinone ring and H\(_8\) of the quinoline ring, respectively. The formation of pyrimidinones 4 in these reactions presumably involves regioselective IDA reactions of vinyl pyrimidinones 1 with N-arylimines 2 participating as 2-azadienes leading to the initial formation and subsequent aromatization of cycloadducts 3.
Reactions of 5-vinyl pyrimidinones 1a-b with N-aryl imines 2a-d

<table>
<thead>
<tr>
<th>Entry</th>
<th>Pyrimidinone</th>
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<th>Time (h)</th>
<th>Product</th>
<th>Yield (%)</th>
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<td>2b</td>
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a The reported yields pertain to crude products prior to recrystallization

In order to extend the scope of such studies to the IDA reactions of sterically more hindered alkenes, we have examined the reactions of 5-isopropenyl pyrimidinones 5 with N-arylimines 2 in the presence of various (lanthanide and non-lanthanide) Lewis acid catalysts. Interestingly, these reactions in the presence of 10 mol % of Y(OTf)₃ and Sc(OTf)₃ in acetonitrile resulted in regio- and chemoselective formation of...
6-oxo-1,6-dihydropyrimidin-5-yl-4H-quinolin-3-one derivatives 6 (Scheme 2; Table 2; Entries 1-8). The yields of the oxidized cycloadducts were found to be higher in presence of Y(OTf)_3 (Entries 1-4) as compared to Sc(OTf)_3 (Table 2, entry 5-7).

![Scheme 2](image)

The structure 6 was assigned to the products with the help of analytical data and spectral evidences. The pyrimidinone 6a, for example, exhibited a molecular ion peak at m/z 546 in its mass spectrum. Its IR spectrum showed the pyrimidinone and pyridone carbonyl absorptions at 1666 cm\(^{-1}\) and 1687 cm\(^{-1}\) respectively. Its \(^1\)H NMR spectrum showed the absence of methylene protons, the presence of a three proton singlet at \(\delta\) 1.73 due to the methyl protons of the quinoline-3-one ring and a characteristic singlet at \(\delta\) 8.34 for the olefinic proton of the pyrimidinone ring. Its \(^13\)C NMR spectrum showed pyrimidinone and quinolinone carbonyls at \(\delta\) 160.5 and 193.7 ppm, respectively. The assigned structure 6a was unambiguously established with the help of X-ray crystallographic studies.

The precise mechanism for the oxidation of methylene to carbonyl observed in these reactions could not be ascertained, however, it may involve a mechanism similar to the one observed in Yb(OTf)_3. Alternatively, it may take place via a charge transfer complex followed by the formation of radical cation as observed in Ce(OTf)_3. However, the reactions of pyrimidinones 5a-b with 2a-c in the presence of non-triflate Lewis acid catalysts, \textit{viz} MgBr\(_2\), InCl\(_3\), ZnCl\(_2\) and resulted in the formation of previously unreported 3,4-dihydroquinolin-4-ylpyrimidin-4-one derivatives 7a-f in good yields (Table 2; Entries 9-20). The use of magnesium (II) bromide as catalyst resulted in better yields of adducts while moderate yield of adducts 7 were obtained when indium (III) chloride and zinc (II) chloride were used as catalysts.
Thus, the oxidation of methylene to carbonyl, observed in reactions of 5a-c with 2a-d in the presence of Y(OTf)₃/Sc(OTf)₃ as catalysts, was not observed when these reactions were performed in the presence of non-triflate LA catalysts. The structure 7 was also assigned on the basis of analytical data and spectral evidences. The ¹H NMR spectrum of 7a exhibited characteristic doublets at δ 2.35 (J=16.5Hz) and δ 4.35 (J=16.5Hz) assigned to the methylene protons of the quinoline ring. The methylene carbon of the quinoline ring appeared at δ 38.4 in its ¹³C NMR. The oxidation of methylene to carbonyl in the presence of Y(OTf)₃/Sc(OTf)₃ catalysts is also supported by the independent transformation of 7 to 6 in the

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**Scheme 3**

**Table 2. Reactions of 5-vinyl pyrimidinones 5a-b with 2a-c**

<table>
<thead>
<tr>
<th>Entry</th>
<th>Pyrimidinone</th>
<th>Imines</th>
<th>Metal salts</th>
<th>Time</th>
<th>Product</th>
<th>Yields</th>
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<tr>
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<td>Y(OTf)₃</td>
<td>12/14/15</td>
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<td>6b/6f/6j</td>
<td>88/85/79</td>
</tr>
<tr>
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* The reported yields pertain to crude products prior to re-crystallization
presence of 10 mol% of Y(OTf)$_3$/Sc(OTf)$_3$ in acetonitrile at room temperature.

CONCLUSION

In conclusion, this manuscript describes the regio- and catalyst selective Imino Diels–Alder reactions of sterically hindered and geminally di-substituted alkenes \textit{viz.} 5-vinyl and isopropenyl-pyrimidinones with \textit{N}-arylimines. The reactions resulted in the synthesis of quinoline-tethered pyrimidinone derivatives in case of vinyl-pyrimidinones 1a-b using triflate as well as non-triflate Lewis acid catalysts, since aromatization of initial IDA adducts is a favored process. However, in case of isopropenyl-pyrimidinones 5a-c, where aromatization of IDA adducts is not possible due to the presence of the methyl group, the reactions accompanied an unprecedented oxidation of methylene to carbonyl in the presence of Y(OTf)$_3$/Sc(OTf)$_3$ leading to the formation of quinolone substituted pyrimidinone derivatives.

EXPERIMENTAL

GENERAL REMARKS

Melting points were determined by open capillary using a Veego Precision Digital Melting Point apparatus (MP-D) and are uncorrected. IR spectra were recorded on a Shimadzu D-8001 spectrophotometer. $^1$H NMR spectra were recorded in deuteriochloroform with a Joel (300 MHz) spectrometers using TMS as internal standard. Chemical shift values are expressed as ppm downfield from TMS and $J$ values are in Hz. Splitting patterns are indicated as s: singlet, d: doublet, t: triplet, m: multiplet, q: quartet, br: broad peak and brs: broad singlet. $^{13}$C NMR spectra were also recorded on a Joel 300 (75.0 MHz) spectrometers in deuteriochloroform using TMS as internal standard. Mass spectra were recorded on a Shimadzu GCMS-QP-2000 mass spectrometer. Elemental analyses were performed on a Heraus CHN-O-Rapid Elemental Analyzer. Column chromatography was performed on a silica gel (60–120 mesh).

GENERAL PROCEDURE

5-Vinyl/isopropenyl pyrimidinones were prepared by the reported methods.$^{11}$ The typical procedure for IDA reactions involved the addition of 10 mol % of Lewis acid to a well stirred solution of \textit{N}-arylimine (1 mmol ) in dry acetonitrile (10 mL) at rt. The solution was allowed to stir for 5-min. followed by the addition of vinyl/isopropenyl pyrimidinones (1 mmol). The progress of the reaction was monitored by tlc using pyrimidinone as the limiting reactant. On completion, the reaction mixture was quenched with a water/MeOH mixture, extracted with CH$_2$Cl$_2$ and concentrated under reduced pressure. Column
chromatography of the crude reaction mixture using a mixture of EtOAc: hexane (1:4) as an eluent resulted in the products which were recrystallized using a mixture of hexane: DCM (5:1).

5-[2-(4-Chlorophenyl)-6-methoxy-quinolin-4-yl]-2,3-diphenyl-3H-pyrimidin-4-one 4a.
Yellow solid; mp 165-166 °C; IR (KBr) ν max 1670 cm⁻¹; ¹H NMR (300 MHz, CDCl₃) : δ 3.88 (s, 3H, OCH₃), 7.06-7.50 (m, 14H, ArH), 7.97 (s, 1H, H₈), 8.12 (m, 3H, H¹¹, H¹², H¹⁴), 8.55 (s, 1H, olefinic proton), ¹³C NMR (75 MHz, CDCl₃) : 55.7 (-OCH₃), 101.8, 114.2, 122.0, 124.4, 126.3, 127.2, 127.9, 128.4, 128.8, 129.0, 129.2, 129.7, 131.4, 134.5, 134.8, 137.5, 138.6, 144.4, 150.2, 153.5, 155.4, 155.6, 158.2, 161.2.; MS m/z [M+1]⁺ : 516; Anal. Calcd for C₃₃H₂₂ClN₃O₂: C, 74.49; H, 4.30; N, 8.14. Found: C, 74.54; H, 4.82; N, 8.02.

5-(6-Methoxy-2-phenylquinolin-4-yl)-2,3-diphenyl-3H-pyrimidin-4-one 4b.
Yellow solid; mp 171-172 °C; IR (KBr) ν max 1667 cm⁻¹; ¹H NMR (300 MHz, CDCl₃) : δ 3.81 (s, 3H, OCH₃), 7.04-7.55 (m, 15H, ArH), 7.98 (s, 1H, H₈), 8.10 (m, 3H, H¹¹, H¹², H¹⁴), 8.52 (s, 1H, olefinic proton), ¹³C NMR (75 MHz, CDCl₃) : 56.2 (-OCH₃), 101.1, 113.9, 121.8, 123.8, 126.5, 127.1, 127.5, 128.3, 128.7, 129.2, 129.4, 129.8, 131.2, 134.4, 134.7, 137.6, 138.3, 144.5, 149.9, 153.6, 155.6, 155.7, 158.3, 161.2.; MS m/z [M+1]⁺ : 482; Anal. Calcd for C₃₂H₂₃N₃O₂: C, 79.81; H, 4.81; N, 8.73. Found: C, 79.74; H, 4.72; N, 8.65.

5-[6-Methoxy-2-(4-methoxyphenyl)-quinolin-4-yl]-2,3-diphenyl-3H-pyrimidin-4-one 4c.
Yellow solid; mp 159-160 °C; IR (KBr) ν max 1672 cm⁻¹; ¹H NMR (300 MHz, CDCl₃) : δ 3.79 (s, 3H, OCH₃), 3.88 (s, 3H, OCH₃), 7.10-7.46 (m, 14H, ArH), 7.98 (s, 1H, H₈), 8.10 (m, 3H, H¹¹, H¹², H¹⁴), 8.52 (s, 1H, olefinic proton), ¹³C NMR (75 MHz, CDCl₃) : 56.0 (-OCH₃), 56.2 (-OCH₃), 100.9, 114.0, 121.6, 123.5, 126.7, 126.8, 127.6, 128.4, 128.6, 129.3, 129.5, 129.9, 130.9, 133.8, 134.2, 137.2, 138.0, 144.1, 149.7, 153.5, 155.4, 155.8, 158.2, 161.3.; MS m/z [M+1]⁺ : 512; Anal. Calcd for C₃₃H₂₅N₃O₃: C, 77.48; H, 4.93; N, 8.21. Found: C, 77.35; H, 4.82; N, 8.05.

5-(6-Methoxy-2-phenylquinolin-4-yl)-2-phenyl-3-p-tolyl-3H-pyrimidin-4-one 4d.
Yellow solid; mp 162-163 °C; IR (KBr) ν max 1665 cm⁻¹; ¹H NMR (300 MHz, CDCl₃) : δ 2.28 (s, 3H, CH₃), 3.74 (s, 3H, OCH₃), 7.12-7.48 (m, 14H, ArH), 8.05 (s, 1H, H₈), 8.09 (m, 3H, H¹¹, H¹², H¹⁴), 8.35 (s, 1H, olefinic proton), ¹³C NMR (75 MHz, CDCl₃) : 16.7(CH₃), 56.2 (-OCH₃), 101.2, 114.1, 121.9, 123.8, 125.8, 126.3, 127.3, 128.0, 128.7, 129.2, 129.6, 130.3, 131.3, 134.1, 134.6, 137.5, 138.8, 144.2, 150.5, 153.5, 155.6, 155.7, 158.5, 161.0.; MS m/z [M+1]⁺ : 496; Anal. Calcd for C₃₃H₂₅N₃O₂: C, 79.98; H, 5.08; N, 8.48. Found: C, 79.73; H, 5.13; N, 8.52.
5-[2-(4-Chlorophenyl)-6-methoxy-quinolin-4-yl]-2-phenyl-3-p-tolyl-3H-pyrimidin-4-one 4e.
Yellow solid; mp 176-177 °C; IR (KBr) νmax 1668 cm⁻¹; ¹H NMR (300 MHz, CDCl₃) : δ 2.31 (s, 3H, CH₃), 3.86 (s, 3H, OCH₃), 7.02- 7.52(m, 13H, ArH), 7.94(s, 1H, H₈), 8.13 (m, 3H, H¹¹, H¹², H¹⁴), 8.55(s, 1H, olefinic proton), ¹³C NMR (75 MHz, CDCl₃) : δ 19.6 (-CH₃), 55.9 (-OCH₃), 101.6, 114.4, 122.2, 124.5, 126.6, 127.4, 127.9, 128.5, 128.7, 129.0, 129.4, 129.8, 131.5, 134.6, 134.9, 137.5, 138.6, 143.9, 153.5, 155.5, 155.7, 158.2, 161.0.; MS m/z [M+1]⁺: 530; Anal. Calcd for C₃₃H₂₄ClN₃O₂: C, 74.78 ; H, 4.56; N, 7.93. Found: C, 74.80; H, 4.62; N, 7.85.

5-[6-Methoxy-2-(4-methoxyphenyl)-quinolin-4-yl]-2-phenyl-3-p-tolyl-3H-pyrimidin-4-one 4f.
Yellow solid; mp 178-179 °C; IR (KBr) νmax 1666 cm⁻¹; ¹H NMR (300 MHz, CDCl₃) : δ 2.32 (s, 3H, CH₃), 3.88 (s, 6H, OCH₃), 7.09- 7.59(m, 13H, ArH), 7.98(s, 1H, H₈), 8.15 (m, 3H, H¹¹, H¹², H¹⁴), 8.50(s, 1H, olefinic proton), ¹³C NMR (75 MHz, CDCl₃) : δ 19.9 (-CH₃), 55.8 (-OCH₃), 56.0 (-OCH₃), 100.6, 113.9, 122.5, 123.9, 126.4, 127.7, 128.1, 128.7, 128.9, 129.2, 129.4, 129.9, 131.4, 134.3, 134.8, 137.3, 138.3, 143.7, 153.6, 155.3, 155.9, 158.4, 160.8; MS m/z [M+1]⁺: 526; Anal. Calcd for C₃₄H₂₇N₃O₃: C, 77.48 ; H, 5.18; N, 7.99. Found: C, 77.80; H, 5.22; N, 7.86.

2-(4-Chlorophenyl)-6-methoxy-4-methyl-4-(6-oxo-1,2-diphenyl-1,6-dihydropyrimidin-5-yl)-4H-quinolin-3-one 6a.
Yellow solid; mp 146-147 °C; IR (KBr) νmax 1666, 1687 cm⁻¹; ¹H NMR (600 MHz, CDCl₃) : δ 1.73(s, 3H, CH₃), 3.84 (s, 3H, OCH₃), 6.65-7.99 (m, 17H, ArH), 8.34(s,1H olefinic proton), ¹³C NMR (150 MHz, CDCl₃) : 24.8(-CH₃), 51.1(C-7, quinolin-3-one ring), 55.5(-OCH₃), 111.4, 112.7, 123.6, 127.9, 128.0, 128.4, 128.6, 128.8, 129.0, 129.2, 129.8, 129.9, 132.7, 134.3, 134.9, 135.3, 136.6, 138.8, 149.4, 154.1, 159.3, 160.3, 160.5 (C=O, pyrimidinone ring), 193.7 (C=O, quinolin-3-one ring): MS m/z [M+1]⁺: 546; Anal. Calcd for C₃₃H₂₄ClN₃O₃: C, 72.59; H, 4.43; N, 7.70. Found: C, 72.71; H, 4.54; N, 7.79.

6-Methoxy-4-methyl-4-(6-oxo-1,2-diphenyl-1,6-dihydropyrimidin-5-yl)-2-phenyl-4H-quinolin-3-one 6b.
Yellow solid; mp 149-150 °C; IR (KBr) νmax 1665, 1689 cm⁻¹; ¹H NMR (300 MHz, CDCl₃) : δ 1.73(s, 3H, CH₃), 3.86 (s, 3H, OCH₃), 6.69-7.98 (m, 18H, ArH), 8.35(s, 1H olefinic proton), ¹³C NMR (75 MHz, CDCl₃) : 24.5(-CH₃), 51.2(C-7, quinolin-3-one ring), 55.6(-OCH₃), 111.2, 112.6, 123.5, 127.9, 128.1, 128.3, 128.5, 128.6, 128.9, 129.0, 129.3, 129.7, 129.8, 132.6, 134.4, 134.8, 135.4, 136.6, 138.8, 149.5, 154.2, 159.1, 160.2, 160.5 (C=O, pyrimidinone ring), 193.5 (C=O, quinolin-3-one ring): MS m/z [M+1]⁺: 512; Anal. Calcd for C₃₃H₂₅N₃O₃: C, 77.48; H, 4.93; N, 8.21. Found: C, 77.66; H, 4.84; N, 8.34.
6-Methoxy-2-(4-methoxyphenyl)-4-methyl-4-(6-oxo-1,2-diphenyl-1,6-dihydropyrimidin-5-yl)-4H-quinolin-3-one 6c.

Yellow solid; mp 155-156 °C; IR (KBr) ν max 1665, 1688 cm⁻¹; ¹H NMR (300 MHz, CDCl₃) : δ 1.72(s, 3H, CH₃), 3.86(s, 3H, OCH₃), 3.88(s, 3H, OCH₃), 6.66-7.94 (m, 17H, ArH), 8.34(s, 1H olefinic proton), ¹³C NMR (75 MHz, CDCl₃) : 24.4(-CH₃), 51.0(C-7, quinolin-3-one ring), 55.5(-OCH₃), 55.6(-OCH₃), 111.3, 112.6, 123.6, 127.8, 128.1, 128.2, 128.4, 128.6, 128.8, 129.1, 129.4, 129.7, 129.9, 132.5, 134.4, 134.7, 135.6, 138.9, 149.4, 154.0, 159.2, 160.3, 160.4 (C=O, pyrimidinone ring), 193.6 (C=O, quinolin-3-one ring): MS m/z [M+1]⁺: 542; Anal. Calcd for C₃₄H₂₇N₃O₄: C, 75.40; H, 5.02; N, 7.76. Found: C, 75.54; H, 5.55; N, 7.45.

6-Methoxy-4-methyl-4-(6-oxo-1,2-diphenyl-1,6-dihydropyrimidin-5-yl)-2-p-tolyl-4H-quinolin-3-one 6d.

Yellow solid; mp 132-133 °C; IR (KBr) ν max 1666, 1689 cm⁻¹; ¹H NMR (300 MHz, CDCl₃) : δ 1.73(s, 3H, CH₃), 2.32 (s, 3H, CH₃), 3.86(s, 3H, OCH₃), 6.65-7.95 (m, 17H, ArH), 8.31(olefinic proton), ¹³C NMR (75 MHz, CDCl₃) : 20.4(-CH₃), 24.4(-CH₃), 51.2(C-7, quinolin-3-one ring), 55.5(-OCH₃), 111.1, 112.5, 123.4, 127.7, 128.0, 128.1, 128.4, 128.5, 128.7, 129.2, 129.5, 129.6, 129.8, 132.5, 134.5, 134.6, 135.7, 136.6, 138.8, 149.6, 154.1, 159.1, 160.2, 160.4 (C=O, pyrimidinone ring), 193.5 (C=O, quinolin-3-one ring): MS m/z [M+1]⁺: 526; Anal. Calcd for C₃₄H₂₇N₃O₃: C, 77.70; H, 5.18; N, 7.99. Found: C, 77.94; H, 5.35; N, 7.88.

2-(4-Chlorophenyl)-6-methoxy-4-methyl-4-(6-oxo-2-phenyl-1-p-tolyl-1,6-dihydropyrimidin-5-yl)-4H-quinolin-3-one 6e.

Yellow solid; mp 143-144 °C; IR (KBr) ν max 1665, 1687 cm⁻¹; ¹H NMR (300 MHz, CDCl₃) : δ 1.72(s, 3H, CH₃), 2.33 (s, 3H, CH₃), 3.89(s, 3H, OCH₃), 6.68-7.99 (m, 17H, ArH), 8.31(olefinic proton), ¹³C NMR (75 MHz, CDCl₃) : 19.3(-CH₃), 24.4(-CH₃), 51.0(C-7, quinolin-3-one ring), 55.7(-OCH₃), 111.1, 112.3, 123.5, 127.7, 128.0, 128.2, 128.3, 128.5, 128.6, 129.5, 129.6, 129.8, 129.9, 132.2, 134.3, 134.5, 135.7, 136.8, 138.6, 149.4, 154.3, 159.1, 160.2, 160.5 (C=O, pyrimidinone ring), 193.6 (C=O, quinolin-3-one ring): MS m/z [M+1]⁺: 560; Anal. Calcd for C₃₄H₂₆ClN₃O₃: C, 77.70; H, 5.18; N, 7.99. Found: C, 77.94; H, 5.35; N, 7.88.

2-(4-Chlorophenyl)-6-methoxy-4-methyl-4-(6-oxo-2-phenyl-1-p-tolyl-1,6-dihydropyrimidin-5-yl)-4H-quinolin-3-one 6f.

Yellow solid; mp 140-141 °C; IR (KBr) ν max 1665, 1687 cm⁻¹; ¹H NMR (300 MHz, CDCl₃) : δ 1.72(s, 3H, CH₃), 2.31 (s, 3H, CH₃), 3.86(s, 3H, OCH₃), 6.66-7.97 (m, 17H, ArH), 8.33(s, 1H olefinic proton), ¹³C NMR (75 MHz, CDCl₃) : 19.5(-CH₃), 24.3(-CH₃), 51.1(C-7, quinolin-3-one ring), 55.6(-OCH₃), 111.2,
112.5, 123.3, 127.6, 128.1, 128.3, 128.4, 128.6, 128.7, 129.4, 129.5, 129.7, 129.8, 132.6, 134.4, 134.5, 135.8, 136.7, 138.7, 149.5, 154.2, 159.0, 160.1, 160.5 (C=O, pyrimidinone ring), 193.7 (C=O, quinolin-3-one ring): MS m/z [M+1]^+: 526; Anal. Calcd for C_{34}H_{27}N_{3}O_{3}: C, 77.70; H, 5.18; N, 7.99. Found: C, 77.88; H, 5.12; N, 7.92.

6-Methoxy-2-(4-methoxy-phenyl)-4-methyl-4-(6-oxo-2-phenyl-1-p-tolyl-1,6-dihydropyrimidin-5-yl)-4H-quinolin-3-one 6g.

Yellow solid; mp 138-139 °C; IR (KBr) ν\text{max} 1666, 1689 cm^{-1}; ^1H NMR (300 MHz, CDCl_3): δ 1.73(s, 3H, CH_3), 2.31 (s, 3H, CH_3), 3.86(s, 3H, OCH_3), 3.87(s, 3H, OCH_3), 6.67-7.95 (m, 17H, ArH), 8.32(olefinic proton), \textsuperscript{13}C NMR (75 MHz, CDCl_3): 19.5(-CH_3), 24.5(-CH_3), 51.2(C-7, quinolin-3-one ring), 55.5(-OCH_3), 55.6(-OCH_3), 111.3, 112.5, 123.5, 127.8, 128.2, 128.3, 128.4, 128.5, 128.7, 129.4, 129.7, 129.8, 129.9, 132.1, 134.4, 134.6, 135.7, 136.7, 138.5, 149.5, 154.4, 159.2, 160.1, 160.4 (C=O, pyrimidinone ring), 193.5 (C=O, quinolin-3-one ring): MS m/z [M+1]^+: 556; Anal. Calcd for C_{35}H_{29}N_{3}O_{4}: C, 75.66; H, 5.26; N, 7.56. Found: C, 75.77; H, 5.40; N, 7.42.

6-Methoxy-4-methyl-4-(6-oxo-2-phenyl-1-p-tolyl-1,6-dihydropyrimidin-5-yl)-2-p-tolyl-4H-quinolin-3-one 6h.

Yellow solid; m.p. 134-135 °C; IR (KBr) ν\text{max} 1665, 1688 cm^{-1}; \textsuperscript{1}H NMR (300 MHz, CDCl_3): δ 1.73(s, 3H, CH_3), 2.31 (s, 3H, CH_3), 3.86(s, 3H, OCH_3), 3.87(s, 3H, OCH_3), 6.66-7.98 (m, 17H, ArH), 8.34(s, 1H olefinic proton), \textsuperscript{13}C NMR (75 MHz, CDCl_3): 19.5(-CH_3), 24.5(-CH_3), 51.1(C-7, quinolin-3-one ring), 55.7(-OCH_3), 111.1, 112.2, 123.4, 127.6, 128.3, 128.4, 128.6, 128.7, 128.9, 129.5, 129.7, 129.8, 129.9, 132.3, 134.5, 134.8, 135.7, 136.6, 138.8, 149.5, 154.5, 159.4, 160.1, 160.5 (C=O, pyrimidinone ring), 193.7 (C=O, quinolin-3-one ring): MS m/z [M+1]^+: 540; Anal. Calcd for C_{35}H_{29}N_{3}O_{4}: C, 77.90; H, 5.42; N, 7.79. Found: C, 77.84; H, 5.40; N, 7.83.

2-(4-Chlorophenyl)-6-methoxy-4-[1-(4-methoxy-phenyl)-6-oxo-2-phenyl-1,6-dihydropyrimidin-5-yl]-4methyl-4H-quinolin-3-one 6i.

Yellow solid; mp 136-137 °C; IR (KBr) ν\text{max} 1667, 1688 cm^{-1}; \textsuperscript{1}H NMR (300 MHz, CDCl_3): δ 1.71(s, 3H, CH_3), 3.86(s, 3H, OCH_3), 3.88(s, 3H, OCH_3), 6.64-7.95 (m, 17H, ArH), 8.32(s, 1H olefinic proton), \textsuperscript{13}C NMR (75 MHz, CDCl_3): 24.8(-CH_3), 51.2(C-7, quinolin-3-one ring), 55.6(-OCH_3), 111.2, 112.4, 123.6, 127.6, 128.1, 128.2, 128.5, 128.7, 128.8, 129.5, 129.6, 129.7, 129.9, 132.4, 134.6, 134.5, 135.6, 136.6, 138.8, 149.5, 154.5, 159.2, 160.1, 160.5 (C=O, pyrimidinone ring), 193.5 (C=O, quinolin-3-one ring): MS m/z [M+1]^+: 576; Anal. Calcd for C_{34}H_{26}ClN_{3}O_{4}: C, 70.89; H, 4.55; N, 7.29. Found: C, 70.81; H, 4.93; N, 7.38.
6-Methoxy-4-[1-(4-methoxyphenyl)-6-oxo-2-phenyl-1,6-dihydropyrimidin-5-yl]-4-methyl-2-phenyl-4H-quinolin-3-one 6j.
Yellow solid; mp 152-153 °C; IR (KBr) $\nu_{\text{max}}$ 1665, 1687 cm$^{-1}$; $^1$H NMR (300 MHz, CDCl$_3$) : $\delta$ 1.73 (s, 3H, CH$_3$), 3.86 (s, 3H, OCH$_3$), 3.87 (s, 3H, OCH$_3$), 6.67-7.99 (m, 17H, ArH), 8.31 (s, 1H olefinic proton), $^{13}$C NMR (75 MHz, CDCl$_3$) : 24.6 (-CH$_3$), 51.3 (C-7, quinolin-3-one ring), 55.5 (-OCH$_3$), 111.3, 112.3, 123.5, 127.7, 128.2, 128.4, 128.7, 128.8, 129.6, 129.7, 129.8, 129.9, 132.4, 134.5, 134.6, 135.8, 136.7, 138.9, 149.3, 154.4, 159.3, 160.0, 160.3 (C=O, quinolin-3-one ring); MS m/z [M+1]$^+$: 542; Anal. Calcd for C$_{34}$H$_{27}$N$_3$O$_4$: C, 75.40; H, 5.02; N, 7.76. Found: C, 75.54; H, 4.99; N, 7.81.

6-Methoxy-2-(4-methoxyphenyl)-4-[1-(4-methoxy-phenyl)-6-oxo-2-phenyl-1,6-dihydropyrimidin-5-yl]-4-methyl-4H-quinolin-3-one 6k.
Yellow solid; mp 128-129 °C; IR (KBr) $\nu_{\text{max}}$ 1665, 1689 cm$^{-1}$; $^1$H NMR (300 MHz, CDCl$_3$) : $\delta$ 1.73 (s, 3H, CH$_3$), 3.87 (s, 6H, OCH$_3$), 3.88 (s, 3H, OCH$_3$), 6.67-7.98 (m, 17H, ArH), 8.34 (s, 1H, olefinic proton), $^{13}$C NMR (75 MHz, CDCl$_3$) : 24.7 (-CH$_3$), 51.2 (C-7, quinolin-3-one ring), 55.6 (-OCH$_3$), 111.4, 112.5, 123.4, 127.6, 128.3, 128.4, 128.5, 128.6, 128.7, 129.7, 129.8, 129.9, 130.1, 132.2, 134.4, 134.5, 135.6, 136.5, 138.7, 149.4, 154.4, 159.1, 160.1, 160.4 (C=O, pyrimidinone ring), 193.6 (C=O, quinolin-3-one ring); MS m/z [M+1]$^+$: 572; Anal. Calcd for C$_{35}$H$_{29}$N$_3$O$_5$: C, 73.54; H, 5.11; N, 7.35. Found: C, 73.78; H, 5.25; N, 7.32.

6-Methoxy-4-[1-(4-methoxyphenyl)-6-oxo-2-phenyl-1,6-dihydropyrimidin-5-yl]-4-methyl-2-p-tolyl-4H-quinolin-3-one 6l.
Yellow solid; mp 130-131 °C; IR (KBr) $\nu_{\text{max}}$ 1666, 1689 cm$^{-1}$; $^1$H NMR (300 MHz, CDCl$_3$) : $\delta$ 1.71 (s, 3H, CH$_3$), 1.87 (s, 3H, CH$_3$), 2.35 (d, 1H, $J$=16.5 Hz), 3.88 (s, 3H, OCH$_3$), 4.35 (d, 1H, $J$= 16.5 Hz), 7.08-7.51 (m, 17H, ArH), 8.35 (s, 1H, olefinic proton), $^{13}$C NMR (75 MHz, CDCl$_3$) : 24.8 (-CH$_3$), 51.1 (C-7, quinolin-3-one ring), 55.5 (-OCH$_3$), 55.6 (-OCH$_3$), 111.3, 112.4, 123.5, 127.7, 128.3, 128.5, 128.6, 128.7, 128.8, 129.7, 129.8, 129.9, 130.2, 132.3, 134.5, 134.6, 135.6, 136.6, 138.8, 149.5, 154.5, 159.2, 160.3, 160.5 (C=O, pyrimidinone ring), 193.5 (C=O, quinolin-3-one ring); MS m/z [M+1]$^+$: 556; Anal. Calcd for C$_{35}$H$_{29}$N$_3$O$_5$: C, 75.66; H, 5.26; N, 7.56. Found: C, 75.75; H, 5.32; N, 7.63.

5-[2-(4-Chlorophenyl)-6-methoxy-4-methyl-3,4-dihydroquinolin-4-yl]-2,3-diphenyl-3H-pyrimidin-4-one 7a.
Yellow solid; mp 179-180 °C; IR (KBr) $\nu_{\text{max}}$ 1668 cm$^{-1}$; $^1$HN MR (300 MHz, CDCl$_3$) : $\delta$ 1.87 (s, 3H, CH$_3$), 2.35 (d, 1H, $J$=16.5 Hz), 3.88 (s, 3H, OCH$_3$), 4.35 (d, 1H, $J$= 16.5 Hz), 7.08-7.51 (m, 17H, ArH), 8.38 (s, 1H, olefinic proton), 8.35 (s, 1H, olefinic proton), 8.38 (s, 1H, olefinic proton), 8.35 (s, 1H, olefinic proton), 8.38 (s, 1H, olefinic proton), 8.35 (s, 1H, olefinic proton), 111.3, 112.4, 123.5, 127.7, 128.3, 128.5, 128.6, 128.7, 128.8, 129.7, 129.9, 130.2, 132.3, 134.5, 134.6, 135.6, 136.6, 138.8, 149.5, 154.5, 159.2, 160.3, 160.5 (C=O, pyrimidinone ring), 193.5 (C=O, quinolin-3-one ring); MS m/z [M+1]$^+$: 556; Anal. Calcd for C$_{35}$H$_{29}$N$_3$O$_5$: C, 75.66; H, 5.26; N, 7.56. Found: C, 75.75; H, 5.32; N, 7.63.
7.95(s, 1H, olefinic proton), $^{13}$CNMR (75MHz, CDCl$_3$) : 21.1(-CH$_3$), 23.6(C-7), 38.3(-CH$_2$-dihydroquinoline ring), 56.7(-OCH$_3$), 111.4, 112.0, 113.4, 121.2, 124.9, 127.3, 127.8, 128.1, 128.5, 128.6, 128.9, 129.5, 129.7, 130.6, 132.2, 132.9, 134.4, 136.1, 150.4, 159.0, 161.2; MS m/z [M+1]$^+$: 532; Anal. Caled for C$_{33}$H$_{26}$ClN$_3$O$_2$: C, 74.50; H, 4.93; N, 7.90. Found: C, 74.84; H, 4.82; N, 7.99.

5-(6-Methoxy-4-methyl-2-phenyl-3,4-dihydro-quinolin-4-yl)-2,3-diphenyl-3$^H$-pyrimidin-4-one 7b.
Yellow solid; mp 185-186 °C; IR (KBr) $\nu_{max}$ 1667 cm$^{-1}$; $^1$H NMR (300 MHz, CDCl$_3$) : $\delta$ 1.87 (s, 3H, CH$_3$), 2.37 (d, 1H, $J$=16.5 Hz), 3.89 (s, 3H, OCH$_3$), 4.37(d, 1H, $J$ = 16.5 Hz), 7.02-7.55(m, 18H, ArH), 7.99(s, 1H, olefinic proton), $^{13}$C NMR (75 MHz, CDCl$_3$) : 21.4(-CH$_3$), 23.6(C-7), 38.5(-CH$_2$-dihydroquinoline ring), 56.6(-OCH$_3$), 111.6, 112.1, 113.3, 121.4, 124.8, 127.5, 127.8, 128.2, 128.5, 128.6, 128.8, 129.6, 129.9, 130.5, 132.3, 132.8, 134.8, 136.3, 150.1, 159.2, 161.1; MS m/z [M+1]$^+$: 498; Anal. Caled for C$_{33}$H$_{27}$N$_3$O$_2$: C, 79.66; H, 5.47; N, 8.44. Found: C, 79.82; H, 5.55; N, 8.49.

5-[6-Methoxy-2-(4-methoxyphenyl)-4-methyl-3,4-dihydroquinolin-4-yl]-2,3-diphenyl-3$^H$-pyrimidin-4-one 7c.
Yellow solid; mp 188-189 °C; IR (KBr) $\nu_{max}$ 1668 cm$^{-1}$; $^1$H NMR (300 MHz, CDCl$_3$) : $\delta$ 1.88 (s, 3H, CH$_3$), 2.34 (d, 1H, $J$=16.4 Hz), 3.84 (s, 6H, OCH$_3$), 4.36(d, 1H, $J$ = 16.4 Hz), 6.99-7.48(m, 16H, ArH), 8.01(s, 1H, olefinic proton), $^{13}$C NMR (75 MHz, CDCl$_3$) : 21.4(-CH$_3$), 23.6(C-7), 38.5(-CH$_2$-dihydroquinoline ring), 56.5(-OCH$_3$), 56.6(-OCH$_3$), 111.3, 112.4, 113.7, 121.3, 124.7, 127.6, 127.7, 128.3, 128.4, 128.5, 128.8, 129.2, 129.7, 130.4, 132.4, 132.7, 134.5, 136.4, 150.0, 159.4, 161.3; MS m/z [M+1]$^+$: 528; Anal. Caled for C$_{34}$H$_{29}$N$_3$O$_3$: C, 77.40; H, 5.54; N, 7.80. Found: C, 77.50; H, 5.45; N, 7.85.

5-[2-(4-Chlorophenyl)-6-methoxy-4-methyl-3,4-dihydroquinolin-4-yl]-2-phenyl-3-p-tolyl-3$^H$-pyrimidin-4-one 7d.
Yellow solid; mp 199-200 °C; IR (KBr) $\nu_{max}$ 1668 cm$^{-1}$; $^1$H NMR (300 MHz, CDCl$_3$) : $\delta$ 1.89(s, 3H, CH$_3$), 2.33(s, 3H, CH$_3$), 2.36 (d, 1H, $J$=16.5 Hz), 3.89 (s, 3H, OCH$_3$), 4.34(d, 1H, $J$ = 16.5 Hz), 6.99-7.48(m, 16H, ArH), 8.01(s, 1H, olefinic proton), $^{13}$C NMR (75 MHz, CDCl$_3$) : 19.8(-CH$_3$), 21.4(-CH$_3$), 23.5(C-7), 38.5(-CH$_2$-dihydroquinoline ring), 56.8(-OCH$_3$), 111.6, 112.5, 113.5, 121.4, 124.6, 127.5, 127.7, 128.3, 128.4, 128.6, 128.7, 129.1, 129.3, 130.2, 132.3, 132.8, 134.7, 136.7, 151.2, 159.2, 161.1; MS m/z [M+1]$^+$: 546; Anal. Caled for C$_{34}$H$_{28}$ClN$_3$O$_2$: C, 74.78; H, 5.17; N, 6.49. Found: C, 74.70; H, 5.29; N, 6.44.

5-(6-Methoxy-4-methyl-2-phenyl-3,4-dihydro-quinolin-4-yl)-2-phenyl-3-p-tolyl-3$^H$-pyrimidin-4-one 7e.
Yellow solid; mp 190-191 °C; IR (KBr) $\nu_{max}$ 1665 cm$^{-1}$; $^1$H NMR (300 MHz, CDCl$_3$) : $\delta$ 1.87(s, 3H, CH$_3$),
5-[6-Methoxy-2-(4-methoxyphenyl)-4-methyl-3,4-dihydroquinolin-4-yl]-2-phenyl-3-p-tolyl-3H-pyrimidin-4-one 7f.

Yellow solid; mp 195-196 °C; IR (KBr) νmax 1667 cm⁻¹; ¹H NMR (300 MHz, CDCl₃) : δ 1.88(s, 3H, CH₃), 2.31(s, 3H, CH₃), 2.34 (d, 1H, J =16.4 Hz), 3.86 (s, 6H, OCH₃), 4.36(d, 1H, J = 16.4 Hz), 7.02-7.52(m, 16H, ArH), 8.01(s, 1H, olefinic proton), ¹³C NMR (75 MHz, CDCl₃) : 19.9(-CH₃), 21.5(-CH₃), 23.7(C-7), 38.8(-CH₂-dihydroquinoline ring), 56.6(-OCH₃), 56.7(-OCH₃), 111.2, 112.5, 113.6, 121.7, 124.8, 127.5, 127.7, 128.2, 128.3, 128.5, 128.8, 129.2, 129.3, 130.3, 132.4, 132.7, 134.6, 136.8, 151.1, 159.5, 161.0; MS m/z [M+1]^+ : 542; Anal. Calcd for C₃5H₃₁N₃O₃ : C, 77.77; H, 5.72; N, 7.47. Found: C, 77.77; H, 5.72; N, 7.47.

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