

Efficient, One-Pot Synthesis of Pyrido[3,2-*d*]pyrimidine Derivatives

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Abstract: A mild and efficient method for the synthesis of pyrido[3,2-*d*]pyrimidine derivatives via three-component domino reaction of amines, aldehydes, and terminal unactivated alkynes is described using $\text{BF}_3 \cdot \text{OEt}_2$ as Lewis acid catalyst in one pot. The features of this procedure are mild reaction conditions, good to high yields, and shorter reaction time with operational simplicity.

Key words: multicomponent reaction, $\text{BF}_3 \cdot \text{OEt}_2$, pyrido[3,2-*d*]pyrimidine, aromatic aldehyde, phenylacetylene

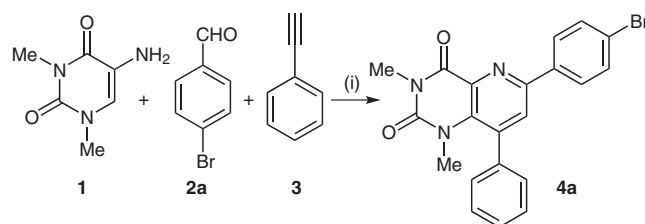
As a consequence of their useful biological properties and their role as pharmacophores of considerable historical importance, simple nitrogen containing heterocycles have received considerable attention in literature. Among the nitrogen heterocycles, pyrimidines, being an integral part of DNA and RNA, play important roles as analgesic,¹ antihypertensive,² antipyretic,³ antiviral,⁴ and anti-inflammatory drugs.⁵ Also they are used in agriculture as pesticides⁶ and plant growth regulators.⁷ Pyridopyrimidines, biologically significant annulated pyrimidines connected with purine pteridines system,^{8–10} are of great interest due to their biological and medicinal applications such as: antifolate,¹¹ antibacterial,¹² tyrosine kinase activity,¹³ antimicrobial,¹⁴ calcium channel antagonists,¹⁵ anti-inflammatory and analgesic,¹⁶ antileishmanial,¹⁷ tuberculostatic,¹⁸ anticonvulsant,¹⁹ diuretic and potassium-sparing,²⁰ antiaggressive,²¹ and antitumor.²² Therefore, activity towards developing convenient and practical synthetic protocol for these compounds still remains an area of contemporary research interest.

The efficiency of a chemical synthesis depends on the parameters such as (i) selectivity and overall yield, (ii) raw materials, (iii) reaction time, (iv) human resources, (v) energy requirements, (vi) toxicity and hazard of the chemicals, and (vii) the protocols involved. Thus, design of highly efficient chemical reaction sequences that provide maximum structural complexity and diversity with a minimum number of synthetic steps to synthesize compounds with interesting properties²³ is important for drug discovery and natural product like compounds. Recently, multicomponent reactions (MCRs) have attracted considerable attention in combinatorial and medicinal chemistry and have been designed to produce elaborate biologically active compounds.²⁴ Multicomponent reactions²⁵ are advantageous compared to linear stepwise synthesis because of

possible structural variations, simplicity of a one-pot procedure, atom economy, and convergent character.^{24d,26}

In view of the importance of pyridopyrimidine derivatives, several methods were developed for the synthesis of pyridopyrimidine derivatives.^{27–30} However, many of these synthetic protocols reported so far suffer from disadvantages, such as harsh reaction conditions, multistep reaction, expensive reagents and longer reaction time, requirement of large amount of catalyst, and lower yields of the reaction. Therefore, a new protocol with reagent economy, one-pot reaction, cheaper catalyst, and improved yields is desirable. In continuation of our work on the synthesis of pyridopyrimidine derivatives³¹ and bioactive heterocycles,³² we became interested to develop an efficient methodology for the synthesis of pyrido[3,2-*d*]pyrimidine derivatives. Herein, we present our recent investigation.

Treatment of 5-amino-1,3-dimethylpyrimidine-2,4(1*H*,3*H*)-dione, an aromatic aldehyde, and phenylacetylene in the presence of 10 mol% of $\text{BF}_3 \cdot \text{OEt}_2$ in toluene at reflux condition afforded the expected pyridopyrimidine derivatives via the aza-Diels–Alder reaction in high yields. In our initial study, the reaction of 5-amino-1,3-dimethylpyrimidine-2,4(1*H*,3*H*)-dione (**1**), 4-bromobenzaldehyde (**2a**), and phenylacetylene (**3**) were used as a model reaction to optimize the reaction conditions (Scheme 1) and the results are presented in Table 1.



Scheme 1 Reagents and conditions: (i) $\text{BF}_3 \cdot \text{OEt}_2$, toluene, reflux, 4 h.

It was found that no product was formed without any catalyst when the reactions were carried out in toluene at room temperature and refluxing condition (Table 1, entries 1, 2). Similar reaction was carried out in the presence of 5, 10, 20 mol% of $\text{BF}_3 \cdot \text{OEt}_2$ (entries 15, 4, 14, respectively) and it was found that use of 10 mol% $\text{BF}_3 \cdot \text{OEt}_2$ at reflux in toluene for four hours (entry 4) gave the highest yield of the product. Lower or higher loading of the catalyst had no significant effect on the reaction yield. Several Lewis and Brønsted acids were used under the same reaction conditions (entries 5–9) for optimization of the reac-

Table 1 Optimization of the Reaction Conditions

Entry	Temp (°C)	Acid catalyst (mol%) ^a	Solvent	Time (h)	Yield ^b (%)
1	r.t.	—	toluene	24	0
2	reflux	—	toluene	24	0
3	r.t.	BF ₃ ·OEt ₂ (10)	toluene	24	0
4 ^c	reflux	BF ₃ ·OEt ₂ (10)	toluene	4	92
5	reflux	Yb(OTf) ₃ (10)	toluene	4	66
6	reflux	CuBr (10)	toluene	6	36
7	reflux	CuI (10)	toluene	6	42
8	reflux	CF ₃ CO ₂ H (10)	toluene	4	59
9	reflux	PTSA (10)	toluene	4	58
10	reflux	BF ₃ ·OEt ₂ (10)	MeCN	4	82
11	reflux	BF ₃ ·OEt ₂ (10)	THF	4	63
12	reflux	BF ₃ ·OEt ₂ (10)	DMF	4	78
13	reflux	BF ₃ ·OEt ₂ (10)	EtOH	4	63
14	reflux	BF ₃ ·OEt ₂ (20)	toluene	4	89
15	reflux	BF ₃ ·OEt ₂ (5)	toluene	4	87

^a Amount (mol%) of acid catalyst required relative to the phenylacetylene in all the entries.

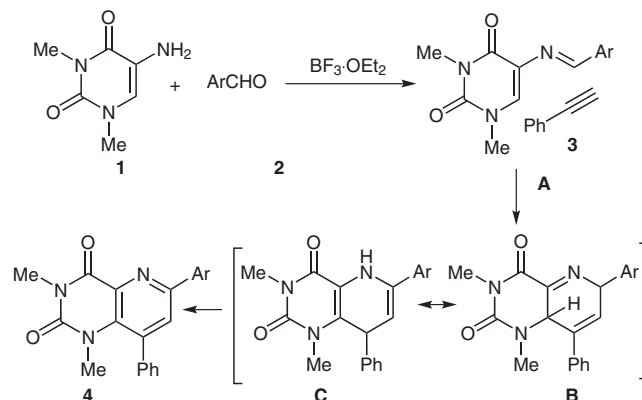
^b Isolated yields.

^c Optimized reaction conditions.

tion conditions. BF₃·OEt₂ furnished better yield compared to other Lewis acids. Brønsted acids gave much lower yield compared to BF₃·OEt₂. In addition, acetonitrile, tetrahydrofuran, *N,N*-dimethylformamide, ethanol (entries 10–13) were also tested as solvents. In these cases product **4a** was formed in lower yield. Variation of the catalyst and solvent showed that running the reaction in refluxing toluene using 10 mol% BF₃·OEt₂ provides the best result.

A small excess (1.1 equiv) of the amine and aromatic aldehyde with respect to phenylacetylene was required to obtain the maximum yield. Further increase in molar amounts resulted in side reactions. Having established the optimized reaction conditions, various aldehydes **2a–l** were subjected to undergo the reaction with 5-amino-1,3-dimethylpyrimidine-2,4(1*H*,3*H*)-dione (**1**) to give various pyrido[3,2-*d*]pyrimidine derivatives **4** in high yields within few hours. It was observed that both electron-donating and electron-withdrawing substituents on the aromatic aldehydes and heterocyclic aldehydes gave the desired pyridopyrimidine derivatives in high yields. *Ortho*, *meta* and *para* substituents as well as disubstitution on the aromatic ring of the aldehydes gave almost similar results. Alkynes like hex-1-yne and hept-1-yne and aliphatic aldehydes like formaldehyde and acetaldehyde did not give the desired pyridopyrimidine derivatives.

Mechanistic rationalization for the formation of pyrido[3,2-*d*]pyrimidine derivatives is outlined in Scheme 2. Initially, a BF₃-catalyzed imine **A** is formed, which possesses the aza-heterodiene moiety. This undergoes intermolecular aza-Diels–Alder reaction with the alkyne, which is activated by BF₃·OEt₂ to give the products **4** via **B** ↔ **C**.

**Scheme 2** The proposed mechanism for the synthesis of pyridopyrimidine derivatives

Chiral Lewis acid or chiral Brønsted acid catalyzed large number of asymmetric aza-hetero Diels–Alder reactions are well known where active preformed dienes are used as a reaction component.^{33–38} The direct asymmetric aza-hetero Diels–Alder reaction of alkynes, which avoids the use of preformed dienes, has been less extensively studied.³⁹

Table 2 Pyridopyrimidine Derivatives Prepared

Entry	Aldehyde 2 Ar	Product 4 Ar	Yield (%)
1	2a 4-BrC ₆ H ₄	4a 4-BrC ₆ H ₄	92
2	2b Ph	4b Ph	89
3	2c 2-BrC ₆ H ₄	4c 2-BrC ₆ H ₄	88
4	2d 2-thienyl	4d 2-thienyl	83
5	2e 2-ClC ₆ H ₄	4e 2-ClC ₆ H ₄	86
6	2f 4-ClC ₆ H ₄	4f 4-ClC ₆ H ₄	90
7	2g 4-MeC ₆ H ₄	4g 4-MeC ₆ H ₄	86
8	2h 4-MeOC ₆ H ₄	4h 4-MeOC ₆ H ₄	81
9	2i 3-ClC ₆ H ₄	4i 3-ClC ₆ H ₄	87
10	2j 2-MeO-5-ClC ₆ H ₄	4j 2-MeO-5-ClC ₆ H ₄	84
11	2k 2,4-(MeO) ₂ C ₆ H ₄	4k 2,4-(MeO) ₂ C ₆ H ₄	80
12	2l 2-MeO-4-MeC ₆ H ₄	4l 2-MeO-4-MeC ₆ H ₄	81

Very recently, some example can be found in the literature for the synthesis of substituted quinolines,⁴⁰ isomeric ellipticine,⁴¹ and pyranoquinoline⁴² derivatives in one pot. However, there is no report of the synthesis of pyrido[3,2-*d*]pyrimidine derivatives avoiding preformed dienes by multicomponent reaction. To the best of our knowledge, this is the first example of multicomponent aza-Diels–Alder reaction of unactivated alkynes in the absence of any copper catalyst under conventional heating avoiding microwave irradiation.

In summary, we have demonstrated a simple, atom-economical, and efficient approach for the synthesis of potentially biologically active pyrido[3,2-*d*]pyrimidine derivatives by one-pot, three-component aza-Diels–Alder reaction using readily available starting materials. The advantages of this method are short reaction times, high yields, mild reaction conditions, easy purification, and use of cheaper catalyst.

Melting points were determined in open capillaries and are uncorrected. IR spectra (cm^{-1}) were recorded on a Perkin–Elmer L 120-000A spectrometer on KBr disks. ^1H NMR and ^{13}C NMR spectra were recorded on a Bruker DPX-400 or Bruker DPX-300 spectrometer in CDCl_3 with TMS as internal standard (chemical shift in δ). CHN analyses were obtained on a 2400 series II CHN PerkinElmer analyzer. MS were recorded on a Q-TOF microTM instrument at the Indian Institute of Chemical Biology, Kolkata and HRMS were recorded on a QTOF MicroYA 263 instrument at the Indian Association for the Cultivation of Science, Kolkata. Silica gel [(60–120, 230–400 mesh), Rankem, India] was used for chromatographic separation. Silica gel G [CDH, (India)] was used for TLC.

Pyridopyrimidines 4a–l; General Procedure

A mixture of heterocyclic amine **1** (167.2 mg, 1.078 mmol, 1.1 equiv) and aromatic aldehyde **2a–l** (1.078 mmol, 1.1 equiv) was stirred in toluene at r.t. for 10 min. After the addition of 10 mol% $\text{BF}_3\cdot\text{OEt}_2$ (mol% calculated relative to phenylacetylene), phenylacetylene (**3**; 100 mg, 0.980 mmol, 1 equiv) was added and the reaction mixture was refluxed for 4 h. After completion of the reaction as monitored by TLC, the reaction mixture was cooled, diluted with sat. aq. NaHCO_3 (50 mL), and extracted with EtOAc (3×25 mL). The combined organic extracts were washed with brine (50 mL) and dried (Na_2SO_4). The solvent was distilled off and the resulting crude product was purified by column chromatography over silica gel (60–120 mesh) using hexane–EtOAc (4:1) as eluent to give compounds **4a–l**.

4a

Yield: 92%; colorless solid; mp 218–220 °C.

IR (KBr): 1477, 1663, 1711, 3057 cm^{-1} .

^1H NMR (400 MHz, CDCl_3): δ = 3.00 (s, 3 H), 3.55 (s, 3 H), 7.41–7.43 (m, 2 H), 7.52 (d, J = 5.2 Hz, 3 H), 7.59 (d, J = 8.4 Hz, 2 H), 7.81 (s, 1 H), 8.00 (d, J = 8.4 Hz, 2 H).

^{13}C NMR (100 MHz, CDCl_3): δ = 29.0, 38.3, 124.1, 127.6, 128.4, 128.5, 129.0, 129.1, 131.9, 133.9, 136.0, 136.2, 138.3, 139.8, 151.5, 151.8, 160.5.

HRMS: m/z calcd for $\text{C}_{21}\text{H}_{16}\text{BrN}_3\text{O}_2$ [$\text{M} + \text{H}$]⁺, [$\text{M} + \text{H} + 2$]⁺: 422.0499, 424.0481; found: 422.0469, 424.0469.

4b

Yield: 89%; colorless solid; mp 196–198 °C.

IR (KBr): 1477, 1664, 1710, 3053 cm^{-1} .

^1H NMR (400 MHz, CDCl_3): δ = 3.00 (s, 3 H), 3.55 (s, 3 H), 7.41–7.51 (m, 8 H), 7.84 (s, 1 H), 8.11 (d, J = 7.6 Hz, 2 H).

^{13}C NMR (100 MHz, CDCl_3): δ = 29.0, 38.3, 127.0, 127.9, 128.5, 128.8, 129.0, 129.0, 129.5, 133.8, 136.0, 137.2, 138.5, 139.7, 151.8, 152.7, 160.6.

MS: m/z = 344 ($\text{M} + \text{H}$)⁺.

Anal. Calcd for $\text{C}_{21}\text{H}_{17}\text{N}_3\text{O}_2$: C, 73.45; H, 4.99; N, 12.24. Found: C, 73.67; H, 4.92; N, 12.13.

4c

Yield: 88%; colorless solid; mp 168–170 °C.

IR (KBr): 1456, 1667, 1710, 3055 cm^{-1} .

^1H NMR (400 MHz, CDCl_3): δ = 3.04 (s, 3 H), 3.56 (s, 3 H), 7.27 (t, J = 7.6 Hz, 1 H), 7.40–7.44 (m, 3 H), 7.47–7.50 (m, 3 H), 7.65 (d, J = 8.0 Hz, 1 H), 7.75 (dd, J = 1.6, 7.8 Hz, 1 H), 7.83 (s, 1 H).

^{13}C NMR (100 MHz, CDCl_3): δ = 29.0, 38.6, 121.8, 127.8, 128.5, 129.0, 129.1, 130.3, 132.1, 132.6, 133.1, 133.7, 136.2, 138.1, 138.4, 139.2, 139.6, 151.9, 153.6, 160.5.

Anal. Calcd for $\text{C}_{21}\text{H}_{16}\text{BrN}_3\text{O}_2$: C, 59.73; H, 3.82; N, 9.95. Found: C, 59.57; H, 3.93; N, 9.91.

4d

Yield: 83%; grey-colored solid; mp 238–240 °C.

IR (KBr): 1477, 1660, 1706, 3058 cm^{-1} .

^1H NMR (400 MHz, CDCl_3): δ = 3.01 (s, 3 H), 3.54 (s, 3 H), 7.11 (t, J = 5.2 Hz, 1 H), 7.40–7.44 (m, 3 H), 7.52 (d, J = 5.6 Hz, 3 H), 7.66 (d, J = 3.6 Hz, 1 H), 7.72 (s, 1 H).

^{13}C NMR (100 MHz, CDCl_3): δ = 29.0, 38.4, 125.7, 126.7, 128.1, 128.4, 128.5, 129.0, 129.1, 133.6, 135.8, 138.3, 139.7, 142.7, 148.5, 151.8, 160.2.

Anal. Calcd for $\text{C}_{19}\text{H}_{15}\text{N}_3\text{O}_2\text{S}$: C, 65.31; H, 4.33; N, 12.03. Found: C, 65.55; H, 4.28; N, 12.18.

4e

Yield: 86%; colorless solid; mp 146–148 °C.

IR (KBr): 1463, 1668, 1715, 3060 cm^{-1} .

^1H NMR (400 MHz, CDCl_3): δ = 3.03 (s, 3 H), 3.56 (s, 3 H), 7.34–7.40 (m, 2 H), 7.42–7.45 (m, 3 H), 7.47–7.50 (m, 3 H), 7.81 (dd, J = 2.0, 7.2 Hz, 1 H), 7.86 (s, 1 H).

^{13}C NMR (100 MHz, CDCl_3): δ = 29.0, 38.5, 127.3, 128.5, 129.0, 129.1, 129.9, 130.2, 132.1, 132.5, 133.8, 136.1, 137.2, 138.1, 138.5, 151.9, 152.2, 160.5.

Anal. Calcd for $\text{C}_{21}\text{H}_{16}\text{ClN}_3\text{O}_2$: C, 66.76; H, 4.27; N, 11.12. Found: C, 66.55; H, 4.32; N, 11.29.

4f

Yield: 90%; colorless solid; mp 138–140 °C.

IR (KBr): 1461, 1663, 1712, 3046 cm^{-1} .

^1H NMR (400 MHz, CDCl_3): δ = 3.02 (s, 3 H), 3.58 (s, 3 H), 7.43 (t, J = 8.8 Hz, 4 H), 7.53 (d, J = 5.6 Hz, 3 H), 7.83 (s, 1 H), 8.09 (d, J = 8.8 Hz, 2 H).

^{13}C NMR (75 MHz, CDCl_3): δ = 29.0, 38.3, 124.1, 127.6, 128.4, 128.5, 129.0, 129.1, 131.9, 133.9, 136.0, 136.2, 138.3, 139.8, 151.5, 151.8, 160.5.

^{13}C DEPT (75 MHz, CDCl_3): δ = 29.0, 38.3, 127.6, 128.2, 128.4, 129.0, 129.0, 129.1.

Anal. Calcd for $\text{C}_{21}\text{H}_{16}\text{ClN}_3\text{O}_2$: C, 66.76; H, 4.27; N, 11.12. Found: C, 66.62; H, 4.38; N, 11.19.

4g

Yield: 86%; colorless solid; mp 178–180 °C.

IR (KBr): 1477, 1660, 1711, 3043 cm⁻¹.¹H NMR (400 MHz, CDCl₃): δ = 2.40 (s, 3 H), 2.99 (s, 3 H), 3.55 (s, 3 H), 7.27 (d, *J* = 7.6 Hz, 2 H), 7.40–7.42 (m, 2 H), 7.51 (d, *J* = 6.0 Hz, 3 H), 7.81 (s, 1 H), 8.02 (d, *J* = 8.0 Hz, 2 H).¹³C NMR (100 MHz, CDCl₃): δ = 21.3, 29.0, 38.3, 126.8, 127.6, 128.5, 129.0, 129.5, 133.7, 134.4, 135.8, 138.6, 139.6, 139.7, 151.9, 152.8, 160.6.Anal. Calcd for C₂₂H₁₉N₃O₂: C, 73.93; H, 5.36; N, 11.76. Found: C, 74.16; H, 5.30; N, 11.82.**4h**

Yield: 81%; colorless solid; mp 140–142 °C.

IR (KBr): 1477, 1661, 1705, 3007 cm⁻¹.¹H NMR (400 MHz, CDCl₃): δ = 3.00 (s, 3 H), 3.56 (s, 3 H), 3.86 (s, 3 H), 7.00 (d, *J* = 8.8 Hz, 2 H), 7.40–7.43 (m, 2 H), 7.51 (d, *J* = 6.8 Hz, 3 H), 7.80 (s, 1 H), 8.10 (d, *J* = 9.2 Hz, 2 H).¹³C NMR (100 MHz, CDCl₃): δ = 27.9, 37.3, 54.3, 113.1, 126.2, 127.3, 127.4, 127.9, 128.7, 132.6, 134.5, 137.5, 138.6, 150.8, 151.5, 159.7, 159.8.MS: *m/z* = 374 (M + H)⁺.Anal. Calcd for C₂₂H₁₉N₃O₃: C, 70.76; H, 5.13; N, 11.25. Found: C, 70.58; H, 5.25; N, 11.14.**4i**

Yield: 87%; colorless solid; mp 206–208 °C.

IR(KBr): 1457, 1666, 1712, 3057 cm⁻¹.¹H NMR (400 MHz, CDCl₃): δ = 3.01 (s, 3 H), 3.51 (s, 3 H), 7.41–7.43 (m, 2 H), 7.52 (d, *J* = 5.2 Hz, 3 H), 7.59 (d, *J* = 8.4 Hz, 2 H), 7.81 (s, 1 H), 8.00 (d, *J* = 8.4 Hz, 2 H).¹³C NMR (75 MHz, CDCl₃): δ = 29.0, 38.3, 125.1, 127.0, 127.9, 128.4, 129.0, 129.1, 129.4, 130.0, 133.8, 134.9, 136.3, 138.2, 138.9, 139.8, 151.2, 151.7, 160.4.¹³C DEPT (75 MHz, CDCl₃): δ = 29.0, 38.3, 125.1, 127.0, 127.9, 128.4, 129.0, 129.1, 129.4, 130.0.Anal. Calcd for C₂₁H₁₆ClN₃O₂: C, 66.76; H, 4.27; N, 11.12. Found: C, 66.99; H, 4.31; N, 11.04.**4j**

Yield: 84%; colorless solid; mp 228–230 °C.

IR (KBr): 1471, 1668, 1712, 3058 cm⁻¹.¹H NMR (400 MHz, CDCl₃): δ = 3.01 (s, 3 H), 3.56 (s, 3 H), 3.84 (s, 3 H), 6.92 (d, *J* = 8.4 Hz, 1 H), 7.33 (dd, *J* = 2.4, 8.8 Hz, 1 H), 7.42 (d, *J* = 7.2 Hz, 2 H), 7.51 (d, *J* = 6.4 Hz, 3 H), 7.98 (d, *J* = 2.4 Hz, 1 H), 8.01 (s, 1 H).¹³C NMR (100 MHz, CDCl₃): δ = 29.0, 38.4, 56.0, 112.6, 126.5, 128.5, 128.9, 130.1, 131.2, 132.6, 133.7, 135.9, 138.5, 150.4, 152.0, 155.6, 160.5.Anal. Calcd for C₂₂H₁₈ClN₃O₃: C, 64.79; H, 4.45; N, 10.30. Found: C, 64.62; H, 4.58; N, 10.39.**4k**

Yield: 80%; grey-colored solid; mp 138–140 °C.

IR (KBr): 1463, 1668, 1712, 3069 cm⁻¹.¹H NMR (400 MHz, CDCl₃): δ = 2.99 (s, 3 H), 3.56 (s, 3 H), 3.84 (s, 3 H), 3.86 (s, 3 H), 6.53 (d, *J* = 1.5 Hz, 1 H), 6.65 (dd, *J* = 1.8, 4.8 Hz, 1 H), 7.40–7.42 (m, 2 H), 7.46–7.56 (m, 3 H), 8.05 (d, *J* = 3.0 Hz, 1 H), 8.07 (s, 1 H).¹³C NMR (75 MHz, CDCl₃): δ = 28.9, 38.3, 55.4, 55.6, 98.7, 105.3, 120.0, 128.5, 128.7, 128.8, 132.2, 132.6, 133.5, 135.1, 138.2, 138.8, 151.7, 152.0, 158.2, 160.8, 161.8.¹³C DEPT (75 MHz, CDCl₃): δ = 28.9, 38.3, 55.4, 55.6, 98.7, 105.3, 128.5, 128.7, 128.8, 132.2, 132.6.Anal. Calcd for C₂₃H₂₁N₃O₄: C, 68.47; H, 5.25; N, 10.42. Found: C, 68.59; H, 5.28; N, 10.29.**4l**

Yield: 81%; grey-colored solid; mp 138–140 °C.

IR (KBr): 1468, 1654, 1702, 3039 cm⁻¹.¹H NMR (400 MHz, CDCl₃): δ = 2.35 (s, 3 H), 3.00 (s, 3 H), 3.56 (s, 3 H), 3.82 (s, 3 H), 6.88 (d, *J* = 8.0 Hz, 1 H), 7.18 (d, *J* = 7.2 Hz, 1 H), 7.42–7.48 (m, 5 H), 7.82 (s, 1 H), 8.04 (s, 1 H).¹³C NMR (75 MHz, CDCl₃): δ = 20.5, 29.0, 38.4, 55.8, 111.3, 126.7, 128.5, 128.8, 128.9, 130.6, 131.0, 131.9, 132.8, 133.6, 135.6, 138.2, 138.7, 152.0, 155.0, 160.8.Anal. Calcd for C₂₃H₂₁N₃O₃: C, 71.30; H, 5.46; N, 10.85. Found: C, 71.55; H, 5.39; N, 10.68.**Acknowledgment**

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