Substituent Effects on the Iodine-Catalyzed Thermal Cyclization of 3,4-Diphenylbuta-1,3-dienyl Isocyanates: Mechanistic Studies

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The thermal cyclization of 3,4-diphenylbuta-1,3-dienyl isocyanates 1, generated in situ from the corresponding azides, was investigated using iodine as a catalyst. Diphenylpyridinones 2, phenylnaphthalenes 3 and indenes 4 were produced via intramolecular ring closure. The nature of the substituents on the phenyl rings was found to be crucial to the distribution of cyclized products 2-4. The mechanism of the reaction is also discussed.
Introduction

The modes of intramolecular cyclization of conjugated isocyanates have attracted considerable attention because they could provide the key intermediates for the syntheses of many biological pyridines, isoquinolines, and other heterocyclic derivatives.\textsuperscript{1-7} Eloy and Washburne extensively applied the thermal cyclization of penta-2,4-dienoyl isocyanates to synthesize $1H$-pyridin-2-ones.\textsuperscript{1,3} Recently, our laboratory utilized the intramolecular cyclization of styryl isocyanates to produce isoquinolinones and discovered that the reaction temperature could be lowered to the refluxing temperature of $o$-dichlorobenzene in the presence of mercury(II) acetate.\textsuperscript{8} In addition, molecular iodine has proven to be a useful Lewis acid catalyst for the activation of carbonyl compounds, including in the $[2+2]$ cyclization of isocyanates with alkenes.\textsuperscript{9} Therefore, we envisioned that iodine would facilitate the cyclization of conjugated isocyanates. This led us to begin a systematic investigation of the feasibility of the intramolecular cyclization of 3,4-diphenylbuta-1,3-dienyl isocyanates 1. To our surprise, not only 3,4-diphenylpyridin-2($1H$)-ones 2 but also phenylnaphthalenes 3 or benzylidene-$1H$-indenones 4 were obtained by the iodine-catalyzed thermal cyclization of isocyanates, generated \textit{in situ} from the corresponding azides 5 (Scheme 1). We found that the cyclized product distribution depends on the substituents of the two phenyl rings, and a mechanistic rationale for the formation of 3,4-diphenylpyridin-2($1H$)-ones 2, phenylnaphthalene 3 and benzylidene-$1H$-indene 4 is also presented.
Results and Discussion

The initial model compound was unsubstituted 3,4-diphenylbuta-1,3-dienyl isocyanate (1a). The effects of various reaction conditions on the thermal cyclization of 1a are summarized in Table 1. First, heating a solution of the geometrically pure 4,5-diphenylpenta-(2E,4Z)-dienoyl azide [(2E,4Z)-5a] in diphenyl ether at 240 °C for 3 h gave 3,4-diphenylpyridin-2(1H)-one (2a) in 11% yield via the cyclization of the butadienyl isocyanate moiety and also furnished another intriguing compound in 9% yield, which was identified as 2-phenylnaphthalene (3a, entry 1). The latter compound 3a was confirmed by comparing its spectral data with literature values.10 A less polar product, 6a, and 2-phenylnaphthalene 3a were obtained in the initial trial while the isocyanate of azide (2E,4Z)-5a disappeared within 1 h.8 The product 6a was deduced as N-(3,4-diphenylbuta-1,3-dienylcarbamoyl)-3,4-diphenyl-1H-pyridin-2-one, an adduct of 3,4-diphenylpyridinone 2a and 3,4-diphenylbuta-1,3-dienyl isocyanate 1a (eq 1). As long as the transformation of isocyanate was completed, the spot of a more
polar pyridinone 2a on the TLC plate developed gradually, while 6a simultaneously faded away slowly with prolonged heating. We are confident that 1a produces both 2a and 3a. The reaction could be monitored by the disappearance of the spots of isocyanate 1 and dimer 6 on the TLC plate.

\[ \text{N} \quad \text{C} \quad \text{O} \quad \text{+NH} \quad \text{O} \quad 2a \]

\[ \text{N} \quad \text{O} \quad \text{H} \quad \text{N} \quad 6a \]

\[(1)\]

Apparently, the unexpected product 3a was produced via the intramolecular cyclization of the triene\(^1\), composed of the two double bonds of the 1,3-butadiene and one double bond of terminal phenyl ring (4-phenyl group), followed by the loss of cyanic acid (HNCO). It is well known that the tricyclic chemical structural pattern, consisting of a phenyl ring attached to the 2-position of a naphthalene moiety, displays various pharmacological activities.\(^12,13\) Unfortunately, only Curtius rearrangement products \((1E,3Z)-1a\) and \((1E,3E)-1a\), were obtained quantitatively from \((2E,4Z)-5a\) after refluxing in o-dichlorobenzene (bp 180 °C) for 5 h (entry 2). We successfully used a flake of iodine to catalyze the cyclization of isocyanates 1a in refluxing o-dichlorobenzene. The result revealed that iodine is an efficient catalyst to promote the conversion of butadienyl isocyanate into pyridin-2(1H)-one 2a (66%) and butadienylbenzene into naphthalene 3a (25%, entry 3). Due to the steric effects, the C-1 and C-2 isomerization of \((1E,3Z)-1a\) to \((1Z,3Z)-1a\) is slightly more difficult than the C-3 and C-4 isomerization of \((1E,3Z)-1a\) to \((1E,3E)-1a\). Therefore, the cyclization, proceeding under milder temperature conditions (150 °C), resulted in naphthalene 4a as the major product (60%, entry 4). When the reactions were performed at temperatures below 150 °C, the reaction time was longer, and the combined yield of 2a and 3a was slightly less (entries 5 and 6). Intriguingly, heating a solution of the geometric isomer \((2E,4E)-\)
5a under the same conditions as entry 4 afforded the same cyclized products 2a and 3a in essentially the same yields (entry 7). This phenomenon led us to make the preliminary conclusion that the identical cyclized result was obtained regardless of the E or Z geometry of the terminal double bond in the dienoyl azide 5.

Table 1. Formation of 2a and 3a from Dienoyl Azide (2E,4Z)-5a

<table>
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<tr>
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<th>solvent</th>
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<th>product (%)</th>
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<td>5</td>
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<td>o-dichlorobenzene</td>
<td>150</td>
<td>7</td>
<td>31</td>
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</table>

*a* No iodine added; *b* dienoyl azide (2E,4E)-5a was used

To investigate the effect of substituents on the phenyl rings on the thermal cyclization of 3,4-diphenylbuta-1,3-dienyl isocyanates 1, a series of 4,5-diphenylpenta-2,4-dienoyl azides 5 was synthesized. The strategy for the preparation of dienoyl azides 5 is shown in Scheme 2. First, a Knoevenagel condensation between phenylacetonitriles 7 and benzaldehydes 8 generated thermodynamically stable (Z)-2,3-diphenylacrylonitriles 9. Subsequently, reduction of 9 with DIBAL-H yielded a geometric mixture of (E)- and (Z)-2,3-diphenylacrylaldehydes 10. The two isomers, (Z)- and (E)-10, were carefully separated by column chromatography. Compound 10 was predominantly
in the E-form, as identified by the NOE correlation between aldehydic proton and olefinic proton. The Wittig reaction between 2,3-diphenylacrylaldehydes 10 and (carboethoxymethylene)triphenylphosphorane, followed by hydrolysis, produced the conjugated acids 11. Finally, 4,5-diphenylpenta-2,4-dienoyl azides 5 were readily prepared quantitatively by the reaction of the corresponding acids 11 with oxalyl chloride followed by the addition of sodium azide. Because the identical cyclized results from (2E,4E)- and (2E,4Z)-5a were observed (Table 1, entries 4 and 7), the mixtures of the geometric isomers (E)- and (Z)-10, (2E,4E)- and (2E,4Z)-11 as well as (2E,4E)- and (2E,4Z)-5 were used directly in the acid, azide formation, and iodine-catalyzed thermal cyclization, respectively.

Scheme 2. Synthesis of 4,5-Diphenylpenta-2,4-dienoyl Azides 5

Reagents and conditions: (a) Na, EtOH, 85 °C, 1 h; (b) DIBAL-H, CH2Cl2, rt, 3 h; (c) (i) Ph3P=CHCO2Et, toluene, refluix, 4 h; (ii) KOH, EtOH-H2O, refluix, 3 h. (d) (i) (COCl)2, toluene, 80 °C, 5 h; (ii) NaN3, acetone, rt, 2 h

The results of the iodine-catalyzed thermal cyclization of 4,5-diphenylpenta-2,4-dienoyl azides 5 at 150 °C in o-dichlorobenzene are shown in Table 2. The reactions were monitored using TLC. First, in the cases of the dienoyl azides 5d and 5e, possessing the 4'-OCH3 on the middle phenyl ring (3-phenyl...
group), the yields of pyridinones 2d and 2e increased, while the yields of naphthalenes 3d and 3e decreased (see entries 1 and 4 and entries 2 and 5). The electrophilic N=O=O in conjugation with the butadiene moiety was activated by the resonance effect of the 4'-OCH₃ and facilitated the cyclization of buta-1,3-dienyl isocyanates to produce pyridinones. Furthermore, when the dienoyl azide 5g bearing a deactivated terminal phenyl ring with a 4''-NO₂ was utilized, the cyclization of butadienylbenzene was retarded, and only product pyridinone 2g was obtained in the good yield (88%, entry 7). Second, the dienoyl azides 5c and 5f, bearing the electron-donating group 3''-OCH₃ para to the C-6'' on the terminal phenyl ring, produced the regiospecific 6-aryl-2,3-dimethoxynaphthalenes 3c and 3f in high yields (83% and 81%, respectively). The regiospecific selection is due to the steric effect of the 3''-OCH₃ which hinders the attack on the C-2''. The results reveal that the cyclization of butadienylbenzene can be promoted by the resonance effect of 3''-OCH₃, which increased the electron density on the C-6'' position (compare entries 2 and 3 and entries 5 and 6). Even in the case of entry 12, dienoyl azide 5l with a 4'-NO₂ on the middle phenyl ring and a 3''-OCH₃ on the terminal phenyl ring gave naphthalene 3l in a comparatively high yield (77%). This result suggests that no matter what kind of groups are on the middle phenyl ring in the dienoyl azide 5, a -OCH₃ on the C-3'' position of terminal phenyl ring increases the formation of naphthalene 3. To our surprise, when 4-(3',4'-dimethoxyphenyl)-5-phenylpentadienoyl azide (5h) was subjected to the same conditions, it was found that 1-benzylidene-5,6-dimethoxy-1H-indene (4h, 32%), instead of 2-(3',4'-dimethoxyphenyl)naphthalene, combined with pyridinone 2h (55%) were obtained (entry 8). A similar result was also obtained when the dienoyl azides 5i and 5j were used (entries 9 and 10). The stereochemistry of the indenes 4 was determined by 2D-NMR analysis. For example, in the NOESY spectrum of 4j, the indenyl H-2 (δ 6.73) showed NOE correlations with the phenyl H-2'' and -6'' (δ 7.65), suggesting that the geometry of 4j was in an E configuration. This configuration was also confirmed by a single-crystal X-ray diffraction study (Figure 1). We supposed that the 3'-OCH₃ could activate the C-6' position of the middle phenyl ring by the
resonance effect and that it not only promoted the intramolecular cyclization of the middle phenyl ring to the C=C double bond to produce indenes 4 but also inhibited the production of naphthalene derivatives 3. Moreover, in the case of entry 10, the dienoyl azide 5j, bearing a strong electron-withdrawing group (4”-NO2) on the terminal phenyl ring as well as a 3’-OCH3 on the middle phenyl ring, led to further encouragement of the cyclization based on the yields of indene 4j (67%). Finally, as mentioned above, 3”-OCH3 could increase the yield of naphthalenes 3; therefore, naphthalene 3k (61%) and pyridinone 2k (12%) were produced by the thermal reaction of 5k with a 3’-OCH3 on the middle phenyl ring accompanied with a 3”-OCH3 on the terminal phenyl ring (entry 11), as in the case of 5c, 5f and 5l. Based on the above analysis, the intramolecular cyclization of a butadienyl isocyanate moiety to form pyridinone 2, a butadienylbenzene moiety to form naphthalene 3, and a 3-phenylethene moiety to form indene 4 competed with one another. The cyclized product distribution depends on the properties and the location of substituents.

Table 2. Iodine-catalyzed Thermal Cyclization of 1 to Give Products 2, 3 and 4

<table>
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<th>entry</th>
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*A mixture of dienoyl azides (2E,4E)-5 and (2E,4Z)-5 was used.*
Mechanistic study

Based on the above results, a complete mechanistic route for the formation of pyridinone 2, naphthalene 3, and indene 4 through an iodine-catalyzed thermal cyclization of 3,4-diphenylbuta-1,3-dienyl isocyanates 1, generated in situ from corresponding azides 5, is proposed as depicted in Scheme 3. First, the reactants 5 underwent a Curtius rearrangement to form the corresponding isocyanates 1. The $(1^E,3^Z)$-1 should isomerize to the $(1^E,3^E)$-1 and $(1^Z,3^Z)$-1 required for the intramolecular ring closure by molecular iodine. The iodine-catalyzed $E/Z$-isomerization of conjugated dienes involves a thermal radical mechanism that has previously already been proposed. The cyclization of the butadienylbenzene moiety in $(1^E,3^E)$-1 followed by the loss of cyanic acid produced naphthalene 3. The intramolecular cyclization can be promoted when $R_3''$ is an electron-donating methoxy group, and the product 3 is then obtained in higher yields. Alternatively, the cyclic reaction of the butadienyl isocyanate moiety in $(1^Z,3^Z)$-1 followed by a hydrogen shift produced pyridinone 2. Because the isocyanate involved in the cyclic reaction was apparently electron-deficient, the cyclization can be facilitated when the substituent $R_4'$ is a methoxy group, and its competing reaction to produce naphthalene will be inhibited when the substituent $R_4''$ is a nitro group. Therefore, the product $2g$ was obtained in the best yield. In addition, the conformer 12 of isocyanate $(1^E,3^Z)$-1 was formed via rotation.
about the C-2 and C-3 single bond. The intramolecular addition of $s$-cis geometric isomer 12 gave the cycloadduct 13 and, after the loss of cyanic acid, produced the sterospecific indenes 4. The carbocation of the intermediate 13 can be stabilized by the resonance effect of the electron-donating substituent on the middle phenyl ring, such as $R_3^* = \text{OCH}_3$. However, the carbanion of the intermediate 13 can also be stabilized by the resonance effect of the electron-withdrawing substituent, NO$_2$ in the $para$ position of the terminal phenyl ring. Hence, $1(E)$-(4''-nitrobenzylidene)-5,6-dimethoxy-$1H$-indene (4j) can be obtained in higher yields.

Scheme 3. Mechanism of the Iodine-Catalyzed Cyclization of ($1E,3Z$)-Dienyl Isocyanate
Conclusion

This paper describes, to our knowledge, the first observation of the intramolecular cyclization of 3,4-diphenylbuta-1,3-dienyl isocyanates 1, generated from 4,5-diphenylpenta-2,4-dienoyl azides 5, to give naphthalenes 3, indenes 4, and pyridinones 2 using iodine as a catalyst under thermal conditions. The effect of the substituents on the two phenyl rings in the reaction of butadienyl isocyanates is shown. An electron-donating group (OCH₃) attached to the meta position of the terminal phenyl ring, regardless of the other substituents, favors the formation of naphthalenes 3. A OCH₃ on the para position of the middle phenyl ring enhances the reaction to yield pyridinones 2. However, a OCH₃ on the meta position of the middle phenyl ring will inhibit the production of naphthalenes 3 and facilitate the production of indenes 4, especially with an extra electron-withdrawing group NO₂ on the para position of the terminal phenyl ring.

Experimental Section

General
All reagents were purchased and used without further purification. Nuclear magnetic resonance spectra were recorded on 400 MHz, 500 MHz and 600 MHz FT-NMR spectrometers; all chemical shifts were reported in ppm from tetramethylsilane as an internal standard. Column chromatography was carried out using 230–400 mesh silica gel and 70–230 mesh neutral aluminum oxide.

General Procedure for the Preparation of (Z)-2,3-diphenylacrylonitrile 9. To a stirred solution of the mixture of phenylacetonitrile 7 (20 mmol) and benzaldehyde 8 (20 mmol) in absolute EtOH (100 mL) at room temperature was added in one portion of freshly prepared 1 M NaOEt (absolute EtOH, 22 mL). The mixture was heated to 85 °C under N₂ for 1 h. After cooling, the precipitate was filtered and washed with small portions of cold EtOH, affording the (Z)-2,3-diphenylacrylonitrile 9.
(Z)-2,3-Diphenylacrylonitrile (9a). Yield 71%; white granule, mp 88–88.5°C (hexane-EtOAc) (lit., mp 88°C). $^1$H NMR (400MHz, CDCl$_3$) δ 7.42 (6H, m), 7.52 (1H, s), 7.67 (2H, dt, $J = 7.1$, 1.5 Hz), 7.88 (2H, m); $^{13}$C NMR (100MHz, CDCl$_3$) δ 111.7, 117.9, 125.9, 128.9, 129.0, 129.1, 129.2, 130.5, 133.7, 134.4, 142.2; IR (KBr) 2218 cm$^{-1}$; EIMS $m/z$ (rel int) 205 (100, M$^+$). Anal. Calcd for C$_{15}$H$_{11}$N: C, 87.77; H, 5.40; N, 6.82. Found: C, 87.61; H, 5.35; N, 6.90.

(Z)-3-(4"-Methoxyphenyl)-2-phenylacrylonitrile (9b). Yield 76%; white granule, mp 93–93.5°C (hexane-CHCl$_3$) (lit., mp 93.5–94.5°C). $^1$H NMR (400MHz, CDCl$_3$) δ 3.84 (3H, s), 6.96 (2H, d, $J = 8.8$ Hz), 7.34 (1H, t, $J = 7.6$ Hz), 7.41 (3H, m), 7.63 (2H, d, $J = 7.6$ Hz), 7.87 (2H, d, $J = 8.8$ Hz); $^{13}$C NMR (100MHz, CDCl$_3$) δ 55.4, 108.6, 114.3, 118.5, 125.7, 126.4, 128.7, 128.9, 131.1, 134.8, 141.8, 161.4; IR (KBr) 2208 cm$^{-1}$; EIMS $m/z$ (rel int) 235 (100, M$^+$). Anal. Calcd for C$_{16}$H$_{13}$NO: C, 81.86; H, 5.57; N, 5.95. Found: C, 81.46; H, 5.71; N, 5.66.

(Z)-3-(3",4"-Dimethoxyphenyl)-2-phenylacrylonitrile (9c). Yield 83%; pale yellow granule, mp 87–88°C (hexane-EtOAc) (lit., mp 88°C). $^1$H NMR (400MHz, CDCl$_3$) δ 7.36 (2H, m), 7.42 (3H, m), 7.65 (2H, d, $J = 8.6$ Hz), 7.71 (1H, d, $J = 2.1$ Hz); $^{13}$C NMR (100MHz, CDCl$_3$) δ 55.9, 56.0, 108.7, 110.8, 111.0, 118.6, 124.4, 125.8, 126.7, 128.7, 129.0, 134.8, 142.1, 149.1, 151.2; IR (KBr) 2203 cm$^{-1}$; EIMS $m/z$ (rel int) 265 (100, M$^+$). Anal. Calcd for C$_{17}$H$_{15}$NO$_2$: C, 76.96; H, 5.70; N, 5.28. Found: C, 77.28; H, 5.56; N, 5.31.

(Z)-2-(4"-Methoxyphenyl)-3-phenylacrylonitrile (9d). Yield 81%; pale yellow granule, mp 91–91.5°C (hexane-CHCl$_3$) (lit., mp 92.5–93.5°C). $^1$H NMR (400MHz, CDCl$_3$) δ 3.83 (3H, s), 6.94 (2H, d, $J = 8.4$ Hz), 7.43 (4H, m), 7.59 (2H, d, $J = 8.4$ Hz), 7.84 (2H, d, $J = 7.5$ Hz); $^{13}$C NMR (100MHz, CDCl$_3$) δ 55.4, 111.2, 114.4, 118.1, 126.9, 127.2, 128.8, 129.0, 130.1, 133.9, 140.0, 160.4; IR (KBr) 2215 cm$^{-1}$; EIMS $m/z$ (rel int) 235 (100, M$^+$). Anal. Calcd for C$_{16}$H$_{13}$NO: C, 81.68; H, 5.57; N, 5.95. Found: C, 81.60; H, 5.36; N, 5.64.
(Z)-2,3-Bis(4′-methoxyphenyl)acrylonitrile (9e). Yield 81%; pale yellow granule, mp 110–111°C (hexane-EtOAc) (lit.,\textsuperscript{21} mp 113°C). \textsuperscript{1}H NMR (400MHz, CDCl\textsubscript{3}) \(\delta\) 3.82 (3H, s), 3.84 (3H, s), 6.92 (2H, d, \(J = 8.8\) Hz), 6.94 (2H, d, \(J = 8.8\) Hz), 7.32 (1H, s), 7.55 (2H, d, \(J = 8.8\) Hz), 7.82 (2H, d, \(J = 8.8\) Hz); \textsuperscript{13}C NMR (100MHz, CDCl\textsubscript{3}) \(\delta\) 55.3 (2×C), 108.2, 114.2, 114.3, 118.6, 126.7, 127.0, 127.2, 130.8, 139.8, 160.0, 161.0; IR (KBr) 2212 cm\textsuperscript{-1}; EIMS \(m/z\) (rel int) 265 (100, M\textsuperscript{+}). Anal. Calcd for C\textsubscript{17}H\textsubscript{15}NO\textsubscript{2}: C, 76.96; H, 5.70; N: 5.28. Found: C, 76.92; H, 5.52; N, 5.08.

(Z)-3-(3″,4″-Dimethoxyphenyl)-2-(4′-methoxyphenyl)acrylonitrile (9f). Yield 81%; pale yellow granule, mp 109–110°C (hexane-CHCl\textsubscript{3}) (lit.,\textsuperscript{22} mp 105°C). \textsuperscript{1}H NMR (400MHz, CDCl\textsubscript{3}) \(\delta\) 3.83 (3H, s), 3.92 (3H, s), 3.95 (3H, s), 6.89 (1H, d, \(J = 6.7\) Hz), 6.93 (2H, d, \(J = 7.1\) Hz), 7.31 (2H, m), 7.56 (2H, d, \(J = 7.1\) Hz), 7.66 (1H, s); \textsuperscript{13}C NMR (100MHz, CDCl\textsubscript{3}) \(\delta\) 55.3, 55.8, 55.9, 108.2, 110.6, 110.8, 114.3, 118.7, 123.9, 126.8, 126.9, 127.2, 140.0, 148.9, 150.7, 160.0; IR (KBr) 2209 cm\textsuperscript{-1}; EIMS \(m/z\) (rel int) 295 (100, M\textsuperscript{+}). Anal. Calcd for C\textsubscript{18}H\textsubscript{17}NO\textsubscript{3}: C, 73.20; H, 5.80; N, 4.74. Found: C, 73.40; H, 5.70; N, 4.50.

(Z)-2-(4′-Methoxyphenyl)-3-(4″-nitrophenyl)acrylonitrile (9g). Yield 60%; yellow granule, mp 105–106°C (hexane-EtOAc). \textsuperscript{1}H NMR (400MHz, CDCl\textsubscript{3}) \(\delta\) 3.87 (3H, s), 6.98 (2H, d, \(J = 8.9\) Hz), 7.47 (1H, s), 7.64 (2H, d, \(J = 8.9\) Hz), 7.98 (2H, d, \(J = 8.9\) Hz), 8.27 (2H, d, \(J = 8.9\) Hz); \textsuperscript{13}C NMR (100MHz, CDCl\textsubscript{3}) \(\delta\) 55.4, 114.6, 115.4, 117.1, 124.0, 125.8, 127.6, 129.6, 136.3, 139.9, 147.8, 161.2; IR (KBr) 2223 cm\textsuperscript{-1}; EIMS \(m/z\) (rel int) 280 (100, M\textsuperscript{+}). Anal. Calcd for C\textsubscript{16}H\textsubscript{12}N\textsubscript{2}O\textsubscript{3}: C, 68.56; H, 4.32; N, 9.99. Found: C, 68.59; H, 4.36; N, 9.66.

(Z)-2-(3″,4″-Dimethoxyphenyl)-3-phenylacrylonitrile (9h). Yield 77%; pale yellow granule, mp 85–85.5°C (hexane-CHCl\textsubscript{3}) (lit.,\textsuperscript{23} mp 85–86°C). \textsuperscript{1}H NMR (400MHz, CDCl\textsubscript{3}) \(\delta\) 3.92 (3H, s), 3.95 (3H, s), 6.92 (1H, d, \(J = 8.4\) Hz), 7.15 (1H, d, \(J = 2.2\) Hz), 7.26 (1H, dd, \(J = 8.4, 2.2\) Hz), 7.45 (4H, m), 7.87 (2H, d, \(J = 8.4\) Hz); \textsuperscript{13}C NMR (100MHz, CDCl\textsubscript{3}) \(\delta\) 55.8, 55.9, 108.6, 111.1, 111.2, 118.0, 118.9, 127.1, 128.7,
128.9, 130.0, 133.7, 140.2, 149.1, 149.9; IR (KBr) 2214 cm⁻¹; EIMS m/z (rel int) 265 (100, M⁺).

Caled for C₁₇H₁₅NO₂: C, 76.96; H, 5.70; N: 5.28. Found: C, 76.55; H, 5.70; N, 5.24.

(Z)-2-(3',4'-Dimethoxyphenyl)-3-(4''-methoxyphenyl)acrylonitrile (9i). Yield 82%; pale yellow granule, mp 129–130°C (hexane-EtOAc) (lit., mp 128–130°C).

**1H NMR (400MHz, CDCl₃) δ 3.84 (3H, s), 3.90 (3H, s), 3.94 (3H, s), 6.88 (1H, d, J = 8.4 Hz), 6.95 (2H, d, J = 8.6 Hz), 7.12 (1H, s), 7.21 (1H, d, J = 8.4 Hz), 7.34 (1H, s), 7.84 (2H, d, J = 8.6 Hz);**

**13C NMR (100MHz, CDCl₃) δ 55.3, 55.9 (2×C), 108.4, 108.6, 111.2, 114.2, 118.5, 118.6, 126.5, 127.6, 130.8, 140.0, 149.2, 149.6, 161.0; IR (KBr) 2210 cm⁻¹; EIMS m/z (rel int) 295 (100, M⁺).

Anal. Caled for C₁₈H₁₇NO₃: C, 73.20; H, 5.80; N, 4.74. Found: C, 73.40; H, 6.08; N, 4.36.

(Z)-2-(3',4'-Dimethoxyphenyl)-3-(4''-nitrophenyl)acrylonitrile (9j). Yield 84%; yellow granule, mp 145–146°C (hexane-CHCl₃).

**1H NMR (400MHz, CDCl₃) δ 3.94 (3H, s), 3.97 (3H, s), 6.94 (1H, d, J = 8.5 Hz), 7.17 (1H, d, J = 2.1 Hz), 7.31 (1H, dd, J = 8.5, 2.1 Hz), 7.49 (1H, s), 7.99 (2H, d, J = 8.7 Hz), 8.28 (2H, d, J = 8.7 Hz);**

**13C NMR (100MHz, CDCl₃) δ 56.0, 56.1, 108.8, 110.8, 111.3, 1115.5, 117.2, 119.7, 124.0, 126.1, 129.6, 136.6, 139.8, 147.9, 149.4, 150.9; IR (KBr) 2218 cm⁻¹; EIMS m/z (rel int) 310 (100, M⁺).

Anal. Caled for C₁₉H₁₄N₂O₄: C, 65.80; H, 4.55; N, 9.03. Found: C, 65.41; H, 4.52; N, 9.08.

(Z)-2,3-Bis(3',4'-dimethoxyphenyl)acrylonitrile (9k). Yield 95%; bright yellow granule, mp 155–156°C (hexane-EtOAc) (lit., mp 154–155°C).

**1H NMR (400MHz, CDCl₃) δ 3.92 (3H, s), 3.94 (3H, s), 3.96 (3H, s), 3.97 (3H, s), 6.91 (1H, d, J = 8.4 Hz), 6.92 (1H, d, J = 8.4 Hz), 7.13 (1H, d, J = 2.0 Hz), 7.23 (1H, dd, J = 8.4, 2.0 Hz), 7.34 (1H, s), 7.35 (1H, dd, J = 8.4, 2.0 Hz), 7.66 (1H, d, J = 2.0 Hz);**

**13C NMR (100MHz, CDCl₃) δ 55.9, 56.0 (3×C), 108.6, 108.8, 110.8, 111.0, 111.4, 118.7, 118.8, 123.9, 126.9, 127.7, 140.3, 149.0, 149.3, 149.8, 150.9; IR (KBr) 2209 cm⁻¹; EIMS m/z (rel int) 325 (100, M⁺).


(Z)-3-(3″,4″-Dimethoxyphenyl)-2-(4′-nitrophenyl)acrylonitrile (9l). Yield 91%; yellow granule, mp 152–153°C (hexane-EtOAc).

**1H NMR (400MHz, CDCl₃) δ 3.98 (3H, s), 3.99 (3H, s), 6.97 (1H, d, J
= 8.4 Hz), 7.43 (1H, dd, J = 8.4, 2.1 Hz), 7.61 (1H, s), 7.79 (1H, d, J = 2.1 Hz), 7.83 (2H, d, J = 8.9 Hz), 8.31 (2H, d, J = 8.9 Hz); \( ^{13} \)C NMR (100MHz, CDCl\(_3\)) \( \delta \) 55.8, 55.9, 105.7, 110.6, 110.8, 117.7, 124.1, 125.5, 125.7, 126.1, 140.8, 145.1, 147.2, 148.9, 152.0; IR (KBr) 2207 cm\(^{-1}\); EIMS \( \text{m/z} \) (rel int) 310 (100, M\(^+\)). Anal. Caled for C\(_{17}\)H\(_{14}\)N\(_2\)O\(_4\): C, 65.80; H, 4.55; N, 9.03. Found: C, 66.09; H, 4.46; N, 9.33.

**General Procedure for the Preparation of 2,3-diphenylacrylaldehyde 10.** To a solution (Z)-2,3-diphenylacrylonitrile 9 (15 mmol) in CH\(_2\)Cl\(_2\) (120 mL) was added 20% diisobutylaluminum hydride in hexane (21 mmol). After the addition of the hydride was complete, the solution was stirred for an additional 30 min at \(-78 \, ^{\circ}\)C and then allowed to come to room temperature over 3 h. A 10% HCl solution (80 mL) was added, and the mixture was stirred for 30 min. The H\(_2\)O layer was extracted with CH\(_2\)Cl\(_2\) (2 \( \times \) 100 mL), and the combined extracts were washed with H\(_2\)O (3 \( \times \) 50 mL), dried with anhydrous MgSO\(_4\), and filtered. The filtrate was concentrated, and the residue was purified by column chromatography over silica gel and eluted with hexane-EtOAc to give a geometric mixture of aldehydes (E)- and (Z)-10. To identify the isomers, we took a small amount of the aldehyde mixture 10 and rechromatographed to give pure aldehydes (E)- and (Z)-10 for spectral analysis. The full spectral data of (E)- and (Z)-10 were described as follows.

**(Z)-2,3-Diphenylacrylaldehyde [(Z)-10a].** Yield 20%; white granule, mp 120–121\(^{\circ}\)C (hexane-EtOAc) (lit.,\(^{25}\) mp 119.5–120.5\(^{\circ}\)C). \(^1\)H NMR (400MHz, CDCl\(_3\)) \( \delta \) 7.38 (10H, m), 7.84 (1H, s), 10.10 (1H, s); \(^{13} \)C NMR (100MHz, CDCl\(_3\)) \( \delta \) 128.3, 128.5, 128.7, 129.5, 130.2, 133.9, 136.2, 141.1, 147.1, 192.1; IR (KBr) 2852, 1683 cm\(^{-1}\); EIMS \( \text{m/z} \) (rel int) 208 (34, M\(^+\)). Anal. Caled for C\(_{15}\)H\(_{12}\)O: C, 86.51; H, 5.81; Found: C, 86.60; H, 5.70.

**(E)-2,3-Diphenylacrylaldehyde [(E)-10a].** Yield 50%; white granule, mp 93.5–94\(^{\circ}\)C (hexane-EtOAc) (lit.,\(^{26}\) mp 93\(^{\circ}\)C). \(^1\)H NMR (400MHz, CDCl\(_3\)) \( \delta \) 7.18 (6H, m), 7.23 (1H, m), 7.35 (4H, m), 9.72 (1H, s); \(^{13} \)C NMR (100MHz, CDCl\(_3\)) \( \delta \) 128.1, 128.3, 128.6, 129.2, 130.0, 130.5, 133.2, 133.8, 141.6, 149.9,
193.6; IR (KBr) 2849, 1665 cm\(^{-1}\); EIMS \(m/z\) (rel int) 208 (100, M\(^+\)). Anal. Calcd for C\(_{15}H_{12}O\): C, 86.51; H, 5.81. Found: C, 86.39; H, 5.67.

\((Z)-3-(4''\text{-Methoxyphenyl})-2\text{-phenylacrylaldehyde} \ ([Z]-10b)\). Yield 32\%; white granule, mp 108–109°C (hexane-EtOAc). \(^1\)H NMR (400MHz, CDCl\(_3\)) \(\delta\) 3.84 (3H, s), 6.94 (2H, d, \(J = 8.6\) Hz), 7.37 (7H, m), 7.77 (1H, s), 10.09 (1H, s); \(^{13}\)C NMR (100MHz, CDCl\(_3\)) \(\delta\) 55.3, 114.0, 126.6, 128.0, 128.2, 128.7, 132.1, 136.5, 139.6, 146.9, 160.9, 192.0; IR (KBr) 2847, 1667 cm\(^{-1}\); EIMS \(m/z\) (rel int) 238 (33, M\(^+\)). Anal. Calcd for C\(_{16}H_{14}O_2\): C, 80.65; H, 5.92. Found: C, 80.92; H, 6.02.

\((E)-3-(4''\text{-Methoxyphenyl})-2\text{-phenylacrylaldehyde} \ ([E]-10b)\). Yield 41\%; white granule, mp 123–124°C (hexane-EtOAc). \(^1\)H NMR (400MHz, CDCl\(_3\)) \(\delta\) 3.74 (3H, s), 6.72 (2H, d, \(J = 8.8\) Hz), 7.14 (2H, d, \(J = 8.8\) Hz), 7.19 (2H, d, \(J = 7.9\) Hz), 7.30 (1H, s), 7.38 (3H, m), 9.69 (1H, s); \(^{13}\)C NMR (100MHz, CDCl\(_3\)) \(\delta\) 55.2, 113.9, 126.6, 128.0, 128.8, 129.3, 132.6, 133.8, 139.7, 150.0, 161.2, 193.7; IR (KBr) 2839, 1668 cm\(^{-1}\); EIMS \(m/z\) (rel int) 238 (100, M\(^+\)). Anal. Calcd for C\(_{16}H_{14}O_2\): C, 80.65; H, 5.92. Found: C, 80.65; H, 5.97.

\((Z)-3-(3''\text{,4''-Dimethoxyphenyl})-2\text{-phenylacrylaldehyde} \ ([Z]-10c)\). Yield 22\%; pale yellow granule, mp 109–110°C (hexane-CHCl\(_3\)). \(^1\)H NMR (500MHz, CDCl\(_3\)) \(\delta\) 3.92 (3H, s), 3.95 (3H, s), 6.93 (2H, m), 7.02 (1H, dd, \(J = 8.3, 1.3\) Hz), 7.39 (5H, m), 7.79 (1H, s), 10.13 (1H, s); \(^{13}\)C NMR (125MHz, CDCl\(_3\)) \(\delta\) 56.0 (2xC), 110.9, 113.1, 124.3, 127.0, 128.2, 128.3, 128.8, 136.5, 140.1, 147.1, 148.9, 150.7, 192.2; IR (KBr) 2837, 1668 cm\(^{-1}\); EIMS \(m/z\) (rel int) 268 (100, M\(^+\)). Anal. Calcd for C\(_{17}H_{16}O_3\): C, 76.10; H, 6.01. Found: C, 76.09; H, 6.26.

\((E)-3-(3''\text{,4''-Dimethoxyphenyl})-2\text{-phenylacrylaldehyde} \ ([E]-10c)\). Yield 40\%; pale yellow granule, mp 123–124°C (hexane-CHCl\(_3\)). \(^1\)H NMR (400MHz, CDCl\(_3\)) \(\delta\) 3.39 (3H, s), 3.84 (3H, s), 6.58 (1H, d, \(J = 1.8\) Hz), 6.77 (1H, d, \(J = 8.4\) Hz), 6.96 (1H, dd, \(J = 8.4, 1.8\) Hz), 7.23 (2H, d, \(J = 7.3\) Hz), 7.32 (1H, s), 7.36 (1H, t, \(J = 7.3\) Hz), 7.44 (2H, t, \(J = 7.3\) Hz), 9.71 (1H, s); \(^{13}\)C NMR (100MHz, CDCl\(_3\)) \(\delta\) 54.9, 55.6, 110.4, 112.0, 125.9, 126.6, 127.9, 128.8, 129.3, 133.9, 139.7, 148.1, 150.2, 150.7, 193.6; IR (KBr) 2839,
1651 cm$^{-1}$; EIMS m/z (rel int) 268 (100, M$^+$$\cdot$). Anal. Caled for C$_{17}$H$_{16}$O$_3$: C, 76.10; H, 6.01. Found: C, 76.43; H, 5.73.

(Z)-2-(4'-Methoxyphenyl)-3-phenylacrylaldehyde [(Z)-10d]. Yield 48%; pale yellow granule, mp 114–115°C (hexane-EtOAc) (lit.,$^{22}$ mp 114–115°C). $^1$H NMR (500MHz, CDCl$_3$) $\delta$ 3.83 (3H, s), 6.93 (2H, d, $J$ = 8.8 Hz), 7.40 (7H, m), 7.81 (1H, s), 10.08 (1H, s); $^{13}$C NMR (125MHz, CDCl$_3$) $\delta$ 55.3, 113.8, 128.5, 128.6, 129.4, 130.0, 130.3, 134.2, 140.6, 145.8, 159.8, 192.5; IR (KBr) 2837, 1672 cm$^{-1}$; EIMS m/z (rel int) 238 (100, M$^+$). Anal. Caled for C$_{16}$H$_{14}$O$_2$: C, 80.65; H, 5.92. Found: C, 80.66; H, 5.67.

(E)-2-(4'-Methoxyphenyl)-3-phenylacrylaldehyde [(E)-10d]. Yield 50%; white granule, mp 112–113°C (hexane-EtOAc). $^1$H NMR (400MHz, CDCl$_3$) $\delta$ 3.82 (3H, s), 6.93 (2H, d, $J$ = 8.7 Hz), 7.13 (2H, d, $J$ = 8.7 Hz), 7.25 (5H, m), 7.33 (1H, s), 9.74 (1H, s); $^{13}$C NMR (100MHz, CDCl$_3$) $\delta$ 55.1, 114.3, 125.2, 128.4, 130.0, 130.5, 130.6, 134.2, 141.3, 149.8, 159.5, 194.2; IR (KBr) 2833, 1670 cm$^{-1}$; EIMS m/z (rel int) 238 (100, M$^+$). Anal. Caled for C$_{16}$H$_{14}$O$_2$: C, 80.65; H, 5.92. Found: C, 80.57; H, 5.66.

(Z)-2,3-Bis(4'-methoxyphenyl)acrylaldehyde [(Z)-10e]. Yield 20%; pale yellow granule, mp 139–140°C (hexane-EtOAc). $^1$H NMR (500MHz, CDCl$_3$) $\delta$ 3.83 (3H, s), 3.86 (3H, s), 6.93 (2H, d, $J$ = 8.8 Hz), 6.95 (2H, d, $J$ = 8.8 Hz), 7.36 (4H, m), 7.74 (1H, s), 10.08 (1H, s); $^{13}$C NMR (125MHz, CDCl$_3$) $\delta$ 55.3, 55.4, 113.8, 114.0, 126.9, 128.9, 129.9, 132.1, 139.2, 145.8, 159.6, 160.8, 192.4; IR (KBr) 2843, 1672 cm$^{-1}$; EIMS m/z (rel int) 268 (100, M$^+$). Anal. Caled for C$_{17}$H$_{16}$O$_3$: C, 76.1; H, 6.01. Found: C, 75.90; H, 5.74.

(E)-2,3-Bis(4'-methoxyphenyl)acrylaldehyde [(E)-10e]. Yield 71%; pale yellow granule, mp 102–103°C (hexane-EtOAc). $^1$H NMR (500MHz, CDCl$_3$) $\delta$ 3.79 (3H, s), 3.84 (3H, s), 6.76 (2H, d, $J$ = 8.8 Hz), 6.96 (2H, d, $J$ = 8.6 Hz), 7.13 (2H, d, $J$ = 8.6 Hz), 7.21 (2H, d, $J$ = 8.8 Hz), 7.28 (1H, s), 9.70 (1H, s); $^{13}$C NMR (125MHz, CDCl$_3$) $\delta$ 55.2, 55.3, 114.0, 114.4, 125.7, 126.9, 130.6, 132.6, 139.4, 149.9, 159.4, 161.1, 194.2; IR (KBr) 2839, 1676 cm$^{-1}$; EIMS m/z (rel int) 268 (44, M$^+$). Anal. Caled for C$_{17}$H$_{16}$O$_3$: C, 76.10; H, 6.01. Found: C, 76.24; H, 6.22.
(Z)-3-(3",4"-Dimethoxyphenyl)-2-(4'-methoxyphenyl)acrylaldehyde [(Z)-10f]. Yield 14%; pale yellow granule, mp 105–106°C (hexane-EtOAc). $^1$H NMR (400MHz, CDCl$_3$) $\delta$ 3.81 (3H, s), 3.89 (3H, s), 3.92 (3H, s), 6.91 (4H, m), 6.97 (1H, dd, $J = 8.3, 1.7$ Hz), 7.36, (2H, d, $J = 8.8$ Hz), 7.73 (1H, s), 10.09 (1H, s); $^{13}$C NMR (100MHz, CDCl$_3$) $\delta$ 55.1, 55.8 (2xC), 110.7, 112.9, 113.6, 124.0, 127.0, 128.7, 129.8, 139.4, 145.6, 148.7, 150.3, 159.5, 192.2; IR (KBr) 2845, 1688 cm$^{-1}$; EIMS $m/z$ (rel int) 298 (100, M$^+$. Anal. Calcd for C$_{18}$H$_{18}$O$_4$: C, 72.47; H, 6.08. Found: C, 72.55; H, 6.15.

(E)-3-(3",4"-Dimethoxyphenyl)-2-(4'-methoxyphenyl)acrylaldehyde [(E)-10f]. Yield 60%; pale yellow granule, mp 101–102°C (hexane-EtOAc). $^1$H NMR (400MHz, CDCl$_3$) $\delta$ 3.50 (3H, s), 3.83 (3H, s), 3.87 (3H, s), 6.73 (1H, d, $J = 2.0$ Hz), 6.79 (1H, d, $J = 8.4$ Hz), 6.98 (3H, m), 7.16 (2H, d, $J = 8.7$Hz), 7.29 (1H, s), 9.71 (1H, s); $^{13}$C NMR (100MHz, CDCl$_3$) $\delta$ 55.2, 55.3, 55.8, 110.6, 112.4, 114.4, 125.7, 125.9, 127.1, 130.8, 139.6, 148.4, 150.5, 150.9, 159.5, 194.0; IR (KBr) 2827, 1669 cm$^{-1}$; EIMS $m/z$ (rel int) 298 (100, M$^+$. Anal. Calcd for C$_{18}$H$_{18}$O$_4$: C, 72.47; H, 6.08. Found: C, 72.45; H, 6.15.

(Z)-2-(4'-Methoxyphenyl)-3-(4"-nitrophenyl)acrylaldehyde [(Z)-10g]. Yield 7%; yellow needle, mp 149–150°C (hexane-EtOAc). $^1$H NMR (500MHz, CDCl$_3$) $\delta$ 3.85 (3H, s), 6.96 (2H, d, $J = 8.9$ Hz), 7.40 (2H, d, $J = 8.9$ Hz), 7.56 (2H, d, $J = 8.3$ Hz), 7.78 (1H, s), 8.29 (2H, d, $J = 8.3$Hz), 10.07 (1H, s); $^{13}$C NMR (125MHz, CDCl$_3$) $\delta$ 55.4, 114.1, 123.7, 127.6, 130.0, 130.8, 140.6, 141.6, 142.8, 147.9, 160.4, 191.5; IR (KBr) 2868, 1684 cm$^{-1}$; EIMS $m/z$ (rel int) 283 (100, M$^+$. Anal. Calcd for C$_{16}$H$_{13}$NO$_4$: C, 67.84; H, 4.63; N, 4.94. Found: C, 67.91; H, 4.33; N, 4.79.

(E)-2-(4'-Methoxyphenyl)-3-(4"-nitrophenyl)acrylaldehyde [(E)-10g]. Yield 38%; yellow granule, mp 164–165°C (hexane-EtOAc). $^1$H NMR (500MHz, CDCl$_3$) $\delta$ 3.85 (3H, s), 6.94 (2H, d, $J = 7.9$ Hz), 7.10 (2H, d, $J = 7.9$ Hz), 7.39 (3H, m), 8.09 (2H, d, $J = 8.1$ Hz), 9.82(1H, s); $^{13}$C NMR (125MHz, CDCl$_3$) $\delta$ 55.2, 114.5, 123.5, 123.9, 130.6, 130.9, 140.7, 143.9, 145.5, 147.7, 160.1, 193.5; IR (KBr) 2839, 1674 cm$^{-1}$; EIMS $m/z$ (rel int) 283 (100, M$^+$. Anal. Calcd for C$_{16}$H$_{13}$NO$_4$: C, 67.84; H, 4.63; N, 4.94. Found: C, 67.84; H, 4.76; N, 4.90.
(Z)-2-(3',4'-Dimethoxyphenyl)-3-phenylacrylaldehyde [(Z)-10h]. Yield 11%; pale yellow granule, mp 89–89.5°C (hexane-EtOAc). $^1$H NMR (500MHz, CDCl$_3$) δ 3.91 (6H, s), 6.91 (1H, d, $J = 8.3$ Hz), 7.00 (1H, s), 7.02 (1H, d, $J = 8.3$ Hz), 7.42 (5H, m), 7.84 (1H, s), 10.08 (1H, s); $^{13}$C NMR (125MHz, CDCl$_3$) δ 55.9 (2×C), 111.0, 111.9, 121.3, 128.4, 128.9, 130.2, 134.0, 146.0, 148.6, 149.3, 192.4; IR (KBr) 2839, 1674 cm$^{-1}$; EIMS $m/z$ (rel int) 268 (100, M$^+$). Anal. Calcd for C$_{17}$H$_{16}$O$_3$: C, 76.10; H, 6.01. Found: C, 76.20; H, 5.82.

(E)-2-(3',4'-Dimethoxyphenyl)-3-phenylacrylaldehyde [(E)-10h]. Yield 43%; pale yellow granule, mp 100–101°C (hexane-EtOAc). $^1$H NMR (400MHz, CDCl$_3$) δ 3.77 (3H, s), 3.91 (3H, s), 6.69 (1H, d, $J = 1.9$ Hz), 6.78 (1H, dd, $J = 8.2$, 1.9 Hz), 6.92 (1H, d, $J = 8.2$ Hz), 7.28 (5H, m), 7.35 (1H, s), 9.76 (1H, s); $^{13}$C NMR (100MHz, CDCl$_3$) δ 55.8 (2×C), 111.5, 112.3, 121.9, 125.5, 128.5, 130.2, 130.7, 134.1, 141.4, 149.0, 149.2, 150.0, 194.1; IR (KBr) 2835, 1670 cm$^{-1}$; EIMS $m/z$ (rel int) 268 (100, M$^+$). Anal. Calcd for C$_{17}$H$_{16}$O$_3$: C, 76.10; H, 6.01. Found: C, 76.00; H, 5.85.

(Z)-2-(3',4'-Dimethoxyphenyl)-3-(4''-methoxyphenyl)acrylaldehyde [(Z)-10i]. Yield 18%; yellow granule, mp 132–133°C (hexane-EtOAc). $^1$H NMR (400MHz, CDCl$_3$) δ 3.87 (3H, s), 3.91 (3H, s), 6.90 (1H, d, $J = 8.2$ Hz), 6.97 (3H, m), 7.00 (1H, dd, $J = 8.2$, 2.0 Hz), 7.36 (2H, d, $J = 8.7$ Hz), 7.77 (1H, s), 10.08 (1H, s); $^{13}$C NMR (100MHz, CDCl$_3$) δ 55.3, 55.8 (2×C), 111.0, 112.0, 113.9, 121.3, 126.7, 129.2, 132.0, 139.2, 146.0, 148.6, 149.1, 160.8, 192.3; IR (KBr) 2839, 1667 cm$^{-1}$; EIMS $m/z$ (rel int) 298 (100, M$^+$). Anal. Calcd for C$_{18}$H$_{18}$O$_4$: C, 72.47; H, 6.08. Found: C, 72.35; H, 5.99.

(E)-2-(3',4'-Dimethoxyphenyl)-3-(4''-methoxyphenyl)acrylaldehyde [(E)-10i]. Yield 75%; yellow needle, mp 114–115°C (hexane-EtOAc) (lit.,$^{28}$ mp 110–112°C). $^1$H NMR (400MHz, CDCl$_3$) δ 3.78 (3H, s), 3.79 (3H, s), 3.92 (3H, s), 6.70 (1H, d, $J = 1.8$ Hz), 6.77 (3H, m), 6.93 (1H, d, $J = 8.2$ Hz), 7.21 (2H, d, $J = 8.0$ Hz), 7.29 (1H, s), 9.70 (1H, s); $^{13}$C NMR (100MHz, CDCl$_3$) δ 55.3, 55.8, 55.9, 111.7, 112.3, 114.0, 121.8, 126.1, 126.8, 132.7, 139.5, 148.9, 149.3, 150.0, 161.2, 194.0; IR (KBr) 2840, 1666 cm$^{-1}$;
EIMS m/z (rel int) 298 (100, M⁺). Anal. Calcd for C₁₈H₁₈O₄: C, 72.47; H, 6.08. Found: C, 72.14; H, 6.10.

(Z)-2-(3',4'-Dimethoxyphenyl)-3-(4''-nitrophenyl)acrylaldehyde [(Z)-10j]. Yield 6%; yellow granule, mp 156–157°C (hexane-EtOAc). ¹H NMR (400MHz, CDCl₃) δ 3.92 (3H, s), 3.93 (3H, s), 6.93 (1H, d, J = 8.4 Hz), 7.00 (1H, d, J = 2.1 Hz), 7.05 (1H, dd, J = 8.4, 2.1 Hz), 7.57 (2H, d, J = 8.6 Hz), 7.79 (1H, s), 8.30 (2H, d, J = 8.6 Hz), 10.07 (1H, s); ¹³C NMR (100MHz, CDCl₃) δ 55.9, 56.0, 111.1, 111.9, 121.6, 123.7, 127.9, 130.8, 140.5, 141.8, 142.9, 148.0, 148.9, 150.1, 191.5; IR (KBr) 2844, 1685 cm⁻¹; EIMS m/z (rel int) 313 (100, M⁺). Anal. Calcd for C₁₇H₁₅NO₅: C, 65.17; H, 4.83; N, 4.47. Found: C, 65.44; H, 4.99; N, 4.62.

(E)-2-(3',4'-Dimethoxyphenyl)-3-(4''-nitrophenyl)acrylaldehyde [(E)-10j]. Yield 35%; yellow granule, mp 166–167°C (hexane-EtOAc). ¹H NMR (400MHz, CDCl₃) δ 3.78 (3H, s), 3.91 (3H, s), 6.69 (1H, d, J = 1.8 Hz), 6.74 (1H, dd, J = 8.2, 1.8 Hz), 6.91 (1H, d, J = 8.2 Hz), 7.40 (1H, s), 7.41 (2H, d, J = 8.9 Hz), 8.08 (2H, d, J = 8.9 Hz), 9.82 (1H, s); ¹³C NMR (100MHz, CDCl₃) δ 55.7, 55.8, 111.6, 112.1, 121.9, 123.4, 124.2, 130.9, 140.5, 143.9, 145.5, 147.7, 149.3, 149.5, 193.3; IR (KBr) 2839, 1674 cm⁻¹; EIMS m/z (rel int) 313 (100, M⁺). Anal. Calcd for C₁₇H₁₅NO₅: C, 65.17; H, 4.83; N, 4.47. Found: C, 64.77; H, 5.09; N, 4.16.

(Z)-2,3-Bis(3',4'-dimethoxyphenyl)acrylaldehyde [(Z)-10k]. Yield 2%; yellow granule, mp 55.0–55.5°C (hexane-EtOAc). ¹H NMR (500MHz, CDCl₃) δ 3.91 (6H, s), 3.92 (3H, s), 3.95 (3H, s), 6.92 (3H, m), 7.00 (3H, m), 7.77 (1H, s), 10.10 (1H, s); ¹³C NMR (125MHz, CDCl₃) δ 55.9 (2×C), 56.0 (2×C), 110.9, 111.1, 112.0, 113.0, 121.4, 124.2, 127.1, 129.3, 139.7, 146.1, 148.7, 148.9, 149.3, 193.3; IR (KBr) 2837, 1682 cm⁻¹; EIMS m/z (rel int) 328 (100, M⁺); HREIMS m/z calcd for C₁₉H₂₀O₅: 328.1311, found: 328.1300 [M⁺].

(E)-2,3-Bis(3',4'-dimethoxyphenyl)acrylaldehyde [(E)-10k]. Yield 87%; yellow syrup. ¹H NMR (400MHz, CDCl₃) δ 3.51 (3H, s), 3.81 (3H, s), 3.87 (3H, s), 3.90 (3H, s), 6.77 (4H, m), 6.96 (2H, m),
7.29 (1H, s), 9.71 (1H, s); $^{13}$C NMR (100MHz, CDCl$_3$) δ 55.2, 55.7, 55.8, 55.9, 110.6, 111.7, 112.5 (2×C), 121.9, 125.7, 126.2, 126.9, 139.6, 148.4, 148.9, 149.4, 150.0 150.9, 193.8; IR (KBr) 2837, 1676 cm$^{-1}$; EIMS m/z (rel int) 328 (100, M$^+$); HREIMS m/z calcd for C$_{19}$H$_{20}$O$_5$: 328.1311, found: 328.1308 [M$^+$].

(E)-2-(4'-Nitrophenyl)-3-(3'',4''-dimethoxyphenyl)acrylaldehyde [(E)-10]. Yield 43%; yellow syrup. $^1$H NMR (500MHz, CDCl$_3$) δ 3.52 (3H, s), 3.88 (3H, s), 6.31 (1H, d, $J = 2.1$ Hz), 6.79 (1H, d, $J = 8.5$ Hz), 6.89 (1H, dd, $J = 8.5, 2.1$ Hz), 7.59 (3H, m), 8.30 (2H, d, $J = 8.8$ Hz), 9.72 (1H, s); $^{13}$C NMR (125MHz, CDCl$_3$) δ 55.4, 56.0, 111.0, 112.8, 123.9, 125.7, 125.9, 131.0, 137.7, 141.2, 147.6, 148.8, 151.6, 151.7, 192.3; IR (KBr) 2837, 1680 cm$^{-1}$; EIMS m/z (rel int) 313 (39, M$^+$); HREIMS m/z calcd for C$_{17}$H$_{15}$NO$_5$: 313.0950, found: 313.0954 [M$^+$].

**General Procedure for the Preparation of 4,5-diphenylpentadienoic acid 11.** A mixture of aldehyde 10 (10 mmol) and (carboethoxymethylene)triphenylphosphorane (12 mmol) in toluene (50 mL) was refluxed under N$_2$ for 4 h. After cooling, the resulting solution was directly purified by column chromatography over silica gel, eluting with hexane-EtOAc to give ethyl 4,5-diphenylpentadienoate. Subsequently, a solution of 1 N KOH (20 mL) was added to a solution of the above ester (8 mmol) in EtOH (40 mL), and the reaction mixture was heated to reflux for 3 h. After cooling, the solution was evaporated, and the residue was dissolved in water (50 mL), acidified with 10% HCl and extracted with EtOAc (5 × 50 mL). The combined extracts were dried with anhydrous MgSO$_4$, filtered, and evaporated under reduced pressure to give a geometric mixture of acids (2E,4E)- and (2E,4Z)-11. To identify the isomers, we took a small amount of the mixture of azides and rechromatographed to give pure (2E,4E)- and (2E,4Z)-11a and (2E,4Z)-11b - 11l for spectral analysis. The full spectral data of (2E,4E)- and (2E,4Z)-11a and (2E,4Z)-11b - 11l are described as follows.

4,5-Diphenylpenta-(2E,4E)-dienoic acid [(2E,4E)-11a]. Yield 70%; white granule, mp 185–186°C (hexane-EtOAc). $^1$H NMR (500MHz, CDCl$_3$) δ 5.91 (1H, d, $J = 15.7$ Hz), 6.95 (1H, s), 7.39 (10H, m),
8.07 (1H, d, \( J = 15.7 \) Hz), 11.10 (1H, br s); \(^{13}\)C NMR (125MHz, CDCl₃) \( \delta \) 122.4, 128.0, 128.3, 128.4, 128.5, 129.0, 129.8, 136.2, 138.6, 138.8, 140.3, 144.5, 172.8; IR (KBr) 1684 cm\(^{-1}\); EIMS \( m/z \) (rel int) 250 (43, \( M^+ \)). Anal. Calcd for C₁₇H₁₄O₂: C, 81.58; H, 5.64. Found: C, 81.50; H, 5.57.

**4,5-Diphenylpenta-(2E,4Z)-dienoic acid [(2E,4Z)-11a]**. Yield 72%; white granule, mp 169–170°C (hexane-EtOAc). \(^1\)H NMR (400MHz, CDCl₃) \( \delta \) 5.50 (1H, d, \( J = 15.4 \) Hz), 6.96 (3H, m), 7.13 (5H, m), 7.40 (3H, m), 7.76 (1H, d, \( J = 15.4 \) Hz), 11.10 (1H, br s); \(^{13}\)C NMR (100MHz, CDCl₃) \( \delta \) 119.3, 128.0, 128.2, 128.5, 129.1, 129.2, 130.1, 135.5, 136.6, 139.4, 139.7, 151.9, 172.8; IR (KBr) 1678 cm\(^{-1}\); EIMS \( m/z \) (rel int) 250 (38, \( M^+ \)). Anal. Calcd for C₁₇H₁₄O₂: C, 81.58; H, 5.64. Found: C, 81.52; H, 5.66.

**5-(4"-Methoxyphenyl)-4-phenylpenta-(2E,4Z)-dienoic acid [(2E,4Z)-11b]**. Yield 100%; bright yellow granule, mp 216–217°C (hexane-CHCl₃). \(^1\)H NMR (500MHz, CDCl₃) \( \delta \) 3.73 (3H, s), 5.42 (1H, d, \( J = 15.3 \) Hz), 6.66 (2H, d, \( J = 8.9 \) Hz), 6.89 (2H, d, \( J = 8.9 \) Hz), 6.90 (1H, s), 7.16 (2H, d, \( J = 7.4 \) Hz), 7.39 (1H, t, \( J = 7.4 \) Hz), 7.43 (2H, t, \( J = 7.4 \) Hz), 7.75 (1H, d, \( J = 15.3 \) Hz), 11.46 (1H, br s); \(^{13}\)C NMR (125MHz, CDCl₃) \( \delta \) 55.2, 113.7, 118.0, 127.9, 128.3, 129.2, 129.3, 131.8, 137.0, 137.4, 139.6, 152.3, 159.8, 172.6; IR (KBr) 1676 cm\(^{-1}\); EIMS \( m/z \) (rel int) 280 (99, \( M^+ \)). Anal. Calcd for C₁₈H₁₆O₃: C, 77.12; H, 5.75. Found: C, 77.41; H, 5.84.

**5-(3"4"-Dimethoxyphenyl)-4-phenylpenta-(2E,4Z)-dienoic acid [(2E,4Z)-11c]**. Yield 90%; bright yellow needle, mp 219–221°C (EtOAc). \(^1\)H NMR (400MHz, CDCl₃) \( \delta \) 3.36 (3H, s), 3.83 (3H, s), 5.44 (1H, d, \( J = 15.3 \) Hz), 6.33 (1H, s), 6.74 (2H, m), 6.90 (1H, s), 7.21 (2H, d, \( J = 7.3 \) Hz), 7.38 (1H, t, \( J = 7.3 \) Hz), 7.46 (2H, t, \( J = 7.3 \) Hz), 7.76 (1H, d, \( J = 15.3 \) Hz); \(^{13}\)C NMR (100MHz, CDCl₃) \( \delta \) 55.0, 55.7, 110.4, 111.6, 118.0, 124.9, 127.9, 128.4, 129.3, 129.4, 137.1, 137.4, 139.6, 148.1, 149.4, 152.1, 172.6; IR (KBr) 1679 cm\(^{-1}\); EIMS \( m/z \) (rel int) 310 (M\(^+\), 42). Anal. Calcd for C₁₉H₁₈O₄: C, 73.53; H, 5.85. Found: C, 73.28; H, 6.02.

**4-(4'-Methoxyphenyl)-5-phenylpenta-(2E,4Z)-dienoic acid [(2E,4Z)-11d]**. Yield 85%; white granule, mp 177–178°C (hexane-CHCl₃). \(^1\)H NMR (500MHz, CDCl₃) \( \delta \) 3.84 (3H, s), 5.54 (1H, d, \( J = 7.3 \) Hz), 6.33 (1H, s), 6.74 (2H, m), 6.90 (1H, s), 7.21 (2H, d, \( J = 7.3 \) Hz), 7.38 (1H, t, \( J = 7.3 \) Hz), 7.46 (2H, t, \( J = 7.3 \) Hz), 7.76 (1H, d, \( J = 15.3 \) Hz); \(^{13}\)C NMR (100MHz, CDCl₃) \( \delta \) 55.0, 55.7, 110.4, 111.6, 118.0, 124.9, 127.9, 128.4, 129.3, 129.4, 137.1, 137.4, 139.6, 148.1, 149.4, 152.1, 172.6; IR (KBr) 1679 cm\(^{-1}\); EIMS \( m/z \) (rel int) 310 (M\(^+\), 42). Anal. Calcd for C₁₉H₁₈O₄: C, 73.53; H, 5.85. Found: C, 73.28; H, 6.02.
15.3 Hz), 6.92 (1H, s), 6.95 (2H, d, J = 8.7 Hz), 6.99 (2H, d, J = 7.9 Hz), 7.06 (2H, d, J = 8.7 Hz), 7.14 (3H, m), 7.75 (1H, d, J = 15.3 Hz), 11.90 (1H, br s); $^{13}$C NMR (125MHz, CDCl$_3$) δ 55.2, 114.7, 119.2, 128.2, 128.3, 128.6, 130.1, 130.3, 135.7, 139.1, 139.7, 152.2, 159.2, 172.9; IR (KBr) 1682 cm$^{-1}$; EIMS m/z (rel int) 280 (100, M$^+$. Anal. Calcd for C$_{18}$H$_{16}$O$_3$: C, 77.12; H, 5.75. Found: C, 76.92; H, 5.66.

4,5-Bis(4'-methoxyphenyl)penta-(2$^E$,4$^Z$)-dienoic acid [(2$^E$,4$^Z$)-11e]. Yield 91%; pale yellow granule, mp 189–190°C (hexane-EtOAc). $^1$H NMR (500MHz, CDCl$_3$) δ 3.75 (3H, s), 3.86 (3H, s), 5.46 (1H, d, J = 15.3 Hz), 6.68 (2H, d, J = 9.0 Hz), 6.88 (1H, s), 6.94 (2H, d, J = 9.0 Hz), 6.97 (2H, d, J = 8.6 Hz), 7.07 (2H, d, J = 8.6 Hz), 7.73 (1H, d, J = 15.3 Hz), 11.43 (1H, br s); $^{13}$C NMR (125MHz, CDCl$_3$) δ 55.2, 55.3, 113.7, 114.8, 117.8, 128.5, 129.0, 130.4, 131.7, 137.1, 139.6, 152.6, 159.2, 159.8, 172.5; IR (KBr) 1676 cm$^{-1}$; EIMS m/z (rel int) 310 (100, M$^+$. Anal. Calcd for C$_{19}$H$_{18}$O$_4$: C, 73.53; H, 5.85. Found: C, 73.72; H, 5.64.

5-(3"',4"'-Dimethoxyphenyl)-4-(4'-methoxyphenyl)penta-(2$^E$,4$^Z$)-dienoic acid [(2$^E$,4$^Z$)-11f]. Yield 83%; pale yellow granule, mp 106–107°C (hexane-CHCl$_3$). $^1$H NMR (400MHz, CDCl$_3$) δ 3.45 (3H, s), 3.83 (3H, s), 3.84 (3H, s), 5.49 (1H, d, J = 15.3 Hz), 6.42 (1H, d, J = 1.5 Hz), 6.70 (1H, d, J = 8.4 Hz), 6.74 (1H, dd, J = 8.4, 1.5 Hz), 6.87 (1H, s), 6.99 (2H, d, J = 8.6 Hz), 7.11 (2H, d, J = 8.6 Hz), 7.74 (1H, d, J = 15.3 Hz), 11.83 (1H, br s); $^{13}$C NMR (100MHz, CDCl$_3$) δ 55.1, 55.3, 55.7, 110.6, 111.9, 114.8, 118.0, 124.6, 128.7, 129.1, 130.6, 137.2, 139.7, 148.2, 149.4, 152.4, 159.3, 172.8; IR (KBr) 1681 cm$^{-1}$; EIMS m/z (rel int) 340 (84, M$^+$. Anal. Calcd for C$_{20}$H$_{20}$O$_5$: C, 73.57; H, 5.92. Found: C, 70.80; H, 5.90.

5-(3"',4"'-Dimethoxyphenyl)-4-(4'-methoxyphenyl)penta-(2$^E$,4$^Z$)-dienoic acid [(2$^E$,4$^Z$)-11f]. Yield 83%; pale yellow granule, mp 106–107°C (hexane-CHCl$_3$). $^1$H NMR (400MHz, CDCl$_3$) δ 3.45 (3H, s), 3.83 (3H, s), 3.84 (3H, s), 5.49 (1H, d, J = 15.3 Hz), 6.42 (1H, d, J = 1.5 Hz), 6.70 (1H, d, J = 8.4 Hz), 6.74 (1H, dd, J = 8.4, 1.5 Hz), 6.87 (1H, s), 6.99 (2H, d, J = 8.6 Hz), 7.11 (2H, d, J = 8.6 Hz), 7.74 (1H, d, J = 15.3 Hz), 11.83 (1H, br s); $^{13}$C NMR (100MHz, CDCl$_3$) δ 55.1, 55.3, 55.7, 110.6, 111.9, 114.8, 118.0, 124.6, 128.7, 129.1, 130.6, 137.2, 139.7, 148.2, 149.4, 152.4, 159.3, 172.8; IR (KBr) 1681 cm$^{-1}$; EIMS m/z (rel int) 340 (84, M$^+$. Anal. Calcd for C$_{20}$H$_{20}$O$_5$: C, 73.57; H, 5.92. Found: C, 70.80; H, 5.90.

4-(4'-Methoxyphenyl)-5-(4"'-nitrophenyl)penta-(2$^E$,4$^Z$)-dienoic acid [(2$^E$,4$^Z$)-11g]. Yield 82%; yellow granule, mp 231–234°C (hexane-CHCl$_3$). $^1$H NMR (500MHz, CDCl$_3$) δ 3.87 (3H, s), 5.67 (1H, d, J = 15.5 Hz), 6.97 (3H, m), 7.05 (2H, d, J = 8.8 Hz), 7.12 (2H, d, J = 8.8 Hz), 7.73 (1H, d, J = 15.5 Hz), 8.00 (2H, d, J = 8.8 Hz); $^{13}$C NMR (125MHz, CDCl$_3$) δ 55.3, 115.0, 121.5, 123.4, 127.4, 130.2, 130.5,
4-(3’,4’-Dimethoxyphenyl)-5-phenylpenta-(2E,4Z)-dienoic acid [(2E,4Z)-11h]. Yield 93%; pale yellow granule, mp 158–159°C (hexane-EtOAc). $^1$H NMR (400MHz, CDCl$_3$) δ 3.79 (3H, s), 3.94 (3H, s), 5.57 (1H, d, $J$ = 15.3 Hz), 6.65 (1H, d, $J$ = 1.6 Hz), 6.72 (1H, dd, $J$ = 8.1, 1.6 Hz), 6.94 (2H, m), 7.01 (2H, m), 7.16 (3H, m), 7.75 (1H, d, $J$ = 15.3 Hz); $^{13}$C NMR (100MHz, CDCl$_3$) δ 55.9, 66.0, 111.9, 112.2, 119.2, 121.5, 128.2, 128.5, 129.0, 130.1, 135.6, 139.1, 139.7, 148.7, 149.7, 152.0, 172.4; IR (KBr) 1682 cm$^{-1}$; EIMS m/z (rel int) 310 (100, M$^+$). Anal. Calcd for C$_{19}$H$_{18}$O$_4$: C, 73.53; H, 5.85. Found: C, 73.21; H, 6.21.

4-(3’,4’-Dimethoxyphenyl)-5-(4”-methoxyphenyl)penta-(2E,4Z)-dienoic acid [(2E,4Z)-11i]. Yield 85%; pale yellow granule, mp 189–190°C (hexane-EtOAc). $^1$H NMR (500MHz, CDCl$_3$) δ 3.76 (3H, s), 3.81 (3H, s), 3.95 (3H, s), 5.50 (1H, d, $J$ = 15.2 Hz), 6.70 (4H, m), 6.88 (1H, s), 6.95 (3H, m), 7.74 (1H, d, $J$ = 15.2 Hz); $^{13}$C NMR (125MHz, CDCl$_3$) δ 55.2, 55.9, 56.0, 112.0, 112.1, 113.8, 117.8, 121.5, 128.3, 129.3, 131.8, 137.1, 139.5, 148.6, 149.7, 152.5, 159.8, 172.5; IR (KBr) 1682 cm$^{-1}$; EIMS m/z (rel int) 340 (100, M$^+$). Anal. Calcd for C$_{20}$H$_{20}$O$_5$: C, 70.57; H, 5.92. Found: C, 70.44; H, 5.96.

4-(3’,4’-Dimethoxyphenyl)-5-(4”-nitrophenyl)penta-(2E,4Z)-dienoic acid [(2E,4Z)-11j]. Yield 82%; yellow granule, mp 224–225°C (hexane-EtOAc). $^1$H NMR (400MHz, DMSO-$d_6$) δ 3.35 (3H, s), 3.69 (6H, s), 5.46 (1H, d, $J$ = 15.4 Hz), 6.64 (1H, d, $J$ = 7.9 Hz), 6.70 (1H, s), 7.03 (1H, d, $J$ = 7.9 Hz), 7.21 (3H, m), 7.61 (1H, d, $J$ = 15.4 Hz), 8.02 (2H, d, $J$ = 7.9 Hz), 12.39 (1H, br s); $^{13}$C NMR (100MHz, DMSO-$d_6$) δ 55.6, 55.8, 112.4, 112.7, 121.2, 123.1, 123.5, 128.1, 130.6, 135.5, 142.8, 142.9, 146.4, 148.4, 149.0, 149.6, 167.5; IR (KBr) 1681 cm$^{-1}$; EIMS m/z (rel int) 355 (100, M$^+$). Anal. Calcd for C$_{19}$H$_{17}$NO$_6$: C, 64.22; H, 4.82; N, 3.94. Found: C, 64.54; H, 5.04; N, 3.70.

4,5-Bis(3’,4’-dimethoxyphenyl)penta-(2E,4Z)-dienoic acid [(2E,4Z)-11k]. Yield 92%; pale yellow granule, mp 237–238°C (hexane-EtOAc). $^1$H NMR (400MHz, DMSO-$d_6$) δ 3.35 (3H, s), 3.69 (6H, s), 5.46 (1H, d, $J$ = 15.4 Hz), 6.64 (1H, d, $J$ = 7.9 Hz), 6.70 (1H, s), 7.03 (1H, d, $J$ = 7.9 Hz), 7.21 (3H, m), 7.61 (1H, d, $J$ = 15.4 Hz), 8.02 (2H, d, $J$ = 7.9 Hz), 12.39 (1H, br s); $^{13}$C NMR (100MHz, DMSO-$d_6$) δ 55.6, 55.8, 112.4, 112.7, 121.2, 123.1, 123.5, 128.1, 130.6, 135.5, 142.8, 142.9, 146.4, 148.4, 149.0, 149.6, 167.5; IR (KBr) 1681 cm$^{-1}$; EIMS m/z (rel int) 355 (100, M$^+$). Anal. Calcd for C$_{19}$H$_{17}$NO$_6$: C, 64.22; H, 4.82; N, 3.94. Found: C, 64.54; H, 5.04; N, 3.70.
3.80 (3H, s), 5.28 (1H, d, J = 15.3 Hz), 6.45 (1H, d, J = 1.7 Hz), 6.67 (1H, dd, J = 8.1, 1.7 Hz), 6.72 (2H, m), 6.82 (1H, d, J = 8.1 Hz), 7.01 (1H, s), 7.07 (1H, d, J = 8.1 Hz), 7.56 (1H, d, J = 15.3 Hz); 13C NMR (100MHz, DMSO-d6) δ 55.0, 55.8, 56.0, 56.1, 111.8, 112.7, 113.0, 113.2, 119.7, 121.6, 124.4, 128.8, 129.8, 137.3, 138.6, 148.3, 148.8, 149.6, 149.9, 150.1, 168.0; IR (KBr) 1665 cm⁻¹; EIMS m/z (rel int) 370 (100, M⁺). Anal. Caled for C21H22O6: C, 68.10; H, 5.99. Found: C, 68.48; H, 6.12.

5-(3",4"-Dimethoxyphenyl)-4-(4'-nitrophenyl)penta-(2E,4Z)-dienoic acid [(2E, 4Z)-11l]. Yield 80%; pale yellow granule, mp 236–237°C (hexane-CH2Cl2). 1H NMR (400MHz, CDCl3) δ 3.48 (3H, s), 3.83 (3H, s), 5.34 (1H, d, J = 15.5 Hz), 6.35 (1H, d, J = 1.8 Hz), 6.62 (1H, dd, J = 8.5, 1.8 Hz), 6.70 (1H, d, J = 8.5 Hz), 7.00 (1H, s), 7.42 (2H, d, J = 8.7 Hz), 7.75 (1H, d, J = 15.5 Hz), 8.33 (2H, d, J = 8.7 Hz); 13C NMR (100MHz, DMSO-d6) δ 54.9, 55.7, 111.7, 112.6, 120.1, 124.2, 124.9, 127.8, 131.2, 135.3, 139.4, 145.1, 147.4 (2×C), 148.4, 149.7, 167.9; IR (KBr) 1678 cm⁻¹; EIMS m/z (rel int) 355 (42, M⁺). Anal. Caled for C19H17NO6: C, 64.22; H, 4.82; N, 3.94. Found: C, 64.15; H, 4.92; N, 3.93.

General Procedure for the Preparation of 4,5-diphenylpentadienoyl azide 5. A mixture of the acids (2E,4E)- and (2E,4Z)-11 (5 mmol) and oxalyl chloride (10 mmol) in toluene (50 mL) was heated for 5 h at 80 °C. After cooling, the resulting mixture was concentrated under reduced pressure to afford 4,5-diphenylpenta-2,4-dienoyl chloride. Subsequently, the pentadienoyl chloride was added immediately into a suspension of NaN₃ (15 mmol) in dry acetone (30 mL) in an ice bath. The reaction mixture was stirred gently for 2 h at room temperature and filtered. The solvent was evaporated in vacuo, and the residue was purified by column chromatography over silica gel and eluted with hexane-CH₂Cl₂ to yield a geometric mixture of azides (2E,4E)- and (2E,4Z)-5. Because the isomers produced the same products in the next step, it was not necessary to separate them, and they could be used directly. To identify the isomers, we took a small amount of the mixture of azides and rechromatographed to give pure (2E,4E)- and (2E,4Z)-5a and (2E,4Z)-5b - 5l for spectral analysis. The full spectral data of (2E,4E)- and (2E,4Z)-5a and (2E,4Z)-5a - 5l are described as follows.
**4,5-Diphenylpenta-(2\text{E},4\text{E})-dienoyl azide \[ (2\text{E},4\text{E})-5\text{a}]**. Yield 100% from acid \((2\text{E},4\text{E})-11\text{a}\); yellow syrup. $^1$H NMR (500MHz, CDCl$_3$) $\delta$ 5.90 (1H, d, $J = 15.6$ Hz), 6.99 (1H, s), 7.36 (10H, m), 8.04 (1H, d, $J = 15.6$ Hz); $^{13}$C NMR (125MHz, CDCl$_3$) $\delta$ 124.1, 128.1, 128.5 (2\times C), 128.6, 129.0, 129.9, 136.1, 138.9, 139.6, 140.1, 144.2, 172.4; IR (KBr) 2143, 1686 cm$^{-1}$; EIMS $m/z$ (rel int) 247 (5, [M-N$_2$]$^+$); HREIMS $m/z$ calcd for C$_{17}$H$_{13}$NO: 247.0997, found: 247.0991 [M-N$_2$]$^+$. 

**4,5-Diphenylpenta-(2\text{E},4\text{Z})-dienoyl azide \[ (2\text{E},4\text{Z})-5\text{a}]**. Yield 100% from acid \((2\text{E},4\text{Z})-11\text{a}\); yellow solid. $^1$H NMR (500MHz, CDCl$_3$) $\delta$ 5.48 (1H, d, $J = 15.3$ Hz), 6.96 (2H, d, $J = 7.3$ Hz), 6.99 (1H, s), 7.16 (5H, m), 7.42 (3H, m), 7.74 (1H, d, $J = 15.3$ Hz); $^{13}$C NMR (125MHz, CDCl$_3$) $\delta$ 121.1, 128.1, 128.3, 128.7, 129.1, 129.3, 130.3, 135.4, 136.4, 139.4, 140.9, 151.7, 172.2; IR (KBr) 2152, 1679 cm$^{-1}$; EIMS $m/z$ (rel int) 275 (8, M$^+$); HREIMS $m/z$ calcd for C$_{17}$H$_{13}$N$_3$O: 275.1059, found: 275.1059 [M$^+$].

**5-(4"-Methoxyphenyl)-4-phenylpenta-(2\text{E},4\text{Z})-dienoyl azide \[ (2\text{E},4\text{Z})-5\text{b}]**. Yield 100% from acid \((2\text{E},4\text{Z})-11\text{b}\); yellow solid. $^1$H NMR (500MHz, CDCl$_3$) $\delta$ 3.71 (3H, s), 5.41 (1H, d, $J = 15.2$ Hz), 6.65 (2H, d, $J = 8.9$ Hz), 6.89 (2H, d, $J = 8.9$ Hz), 6.92 (1H, s), 7.12 (2H, d, $J = 8.0$ Hz), 7.39 (3H, m), 7.72 (1H, d, $J = 15.2$ Hz); $^{13}$C NMR (125MHz, CDCl$_3$) $\delta$ 55.1, 113.7, 119.6, 127.9, 128.0, 129.1, 129.3, 131.9, 136.7, 137.2, 140.8, 152.0, 160.0, 172.0; IR (KBr) 2140, 1680 cm$^{-1}$; EIMS $m/z$ (rel int) 305 (4, M$^+$); HREIMS $m/z$ calcd for C$_{18}$H$_{15}$N$_3$O$_2$: 305.1164, found: 305.1157 [M$^+$].

**5-(3"4"-Dimethoxyphenyl)-4-phenylpenta-(2\text{E},4\text{Z})-dienoyl azide \[ (2\text{E},4\text{Z})-5\text{c}]**. Yield 96% from acid \((2\text{E},4\text{Z})-11\text{c}\); yellow solid. $^1$H NMR (400MHz, CDCl$_3$) $\delta$ 3.35 (3H, s), 3.81 (3H, s), 5.42 (1H, d, $J = 15.2$ Hz), 6.31 (1H, s), 6.70 (1H, d, $J = 8.4$ Hz), 6.76 (1H, d, $J = 8.4$ Hz), 6.92 (1H, s), 7.18 (2H, d, $J = 7.4$ Hz), 7.38 (1H, t, $J = 8.4$ Hz), 7.45 (2H, t, $J = 7.4$ Hz), 7.73 (1H, d, $J = 15.2$ Hz); $^{13}$C NMR (100MHz, CDCl$_3$) $\delta$ 54.8, 55.6, 110.3, 111.4, 119.6, 125.1, 127.8, 128.1, 129.2, 129.3, 136.7, 137.1, 140.8, 148.0, 149.6, 151.7, 171.9; IR (KBr) 2133, 1682 cm$^{-1}$; EIMS $m/z$ (rel int) 307 (86, [M-N$_2$]$^+$); HREIMS $m/z$ calcd for C$_{19}$H$_{17}$NO$_3$: 307.1208, found: 307.1219 [M-N$_2$]$^+$. 

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4-(4'-Methoxyphenyl)-5-phenylpenta-(2E,4Z)-dienoyl azide [(2E,4Z)-5d]. Yield 100% from acid (2E,4Z)-11d; yellow solid. \(^1\)H NMR (500MHz, CDCl\(_3\)) \(\delta\) 3.85 (3H, s), 5.52 (1H, d, \(J = 15.3\) Hz), 6.95 (2H, d, \(J = 8.6\) Hz), 6.96 (1H, s), 6.99 (2H, d, \(J = 6.7\) Hz), 7.05 (2H, d, \(J = 8.6\) Hz), 7.15 (3H, m), 7.73 (1H, d, \(J = 15.3\) Hz); \(^1\)C NMR (125MHz, CDCl\(_3\)) \(\delta\) 55.3, 114.7, 120.9, 128.2, 128.3, 128.6, 130.2, 130.3, 135.5, 139.1, 140.9, 152.1, 159.4, 172.2; IR (KBr) 2142, 1681 cm\(^{-1}\); EIMS \(m/z\) (rel int) 305 (5, \([M]^+\); HREIMS \(m/z\) calcd for C\(_{18}\)H\(_{15}\)N\(_3\)O\(_2\): 305.1164, found: 305.1160 \([M]^+\).

4,5-Bis(4'-methoxyphenyl)penta-(2E,4Z)-dienoyl azide [(2E,4Z)-5e]. Yield 91% from acid (2E,4Z)-11e; yellow solid. \(^1\)H NMR (500MHz, CDCl\(_3\)) \(\delta\) 3.74 (3H, s), 3.86 (3H, s), 5.45 (1H, d, \(J = 15.2\) Hz), 6.68 (2H, d, \(J = 8.8\) Hz), 6.94 (5H, m), 7.05 (2H, d, \(J = 8.8\) Hz), 7.71 (1H, d, \(J = 15.2\) Hz); \(^1\)C NMR (125MHz, CDCl\(_3\)) \(\delta\) 55.2, 55.3, 113.8, 114.8, 119.6, 128.3, 128.7, 130.3, 131.9, 137.0, 140.9, 152.5, 159.3, 160.0, 172.2; IR (KBr) 2137, 1675 cm\(^{-1}\); EIMS \(m/z\) (rel int) 335 (3, \([M]^+\); HREIMS \(m/z\) calcd for C\(_{19}\)H\(_{17}\)N\(_3\)O\(_3\): 335.1270, found: 335.1265 \([M]^+\).

5-(3",4"-Dimethoxyphenyl)-4-(4'-methoxyphenyl)penta-(2E,4Z)-dienoyl azide [(2E,4Z)-5f]. Yield 83% from acid (2E,4Z)-11f; yellow solid. \(^1\)H NMR (500MHz, CDCl\(_3\)) \(\delta\) 3.44 (3H, s), 3.83 (3H, s), 3.84 (3H, s), 5.47 (1H, d, \(J = 15.1\) Hz), 6.41 (1H, s), 6.71 (1H, d, \(J = 8.4\) Hz), 6.75 (1H, d, \(J = 8.4\) Hz), 6.91 (1H, s), 6.99 (2H, d, \(J = 8.7\) Hz), 7.09 (2H, d, \(J = 8.7\) Hz), 7.72 (1H, d, \(J = 15.1\) Hz); \(^1\)C NMR (125MHz, CDCl\(_3\)) \(\delta\) 55.1, 55.3, 55.7, 110.5, 111.9, 114.8, 119.7, 124.9, 128.5, 128.8, 130.5, 137.1, 140.9, 148.2, 149.7, 152.2, 159.3, 172.1; IR (KBr) 2129, 1675 cm\(^{-1}\); EIMS \(m/z\) (rel int) 337 (100, \([M-N_2]^+\); HREIMS \(m/z\) calcd for C\(_{20}\)H\(_{19}\)N\(_3\)O\(_4\): 337.1314, found: 337.1321 [M-N\(_2\)]\(^+\).

4-(4'-Methoxyphenyl)-5-(4"-nitrophenyl)penta-(2E,4Z)-dienoyl azide [(2E,4Z)-5g]. Yield 80% from acid (2E,4Z)-11g; yellow solid. \(^1\)H NMR (500MHz, CDCl\(_3\)) \(\delta\) 3.87 (3H, s), 5.64 (1H, d, \(J = 15.2\) Hz), 6.97 (3H, m), 7.03 (2H, m), 7.12 (2H, d, \(J = 8.8\) Hz), 7.72 (1H, d, \(J = 15.2\) Hz), 8.00 (2H, d, \(J = 8.8\) Hz); \(^1\)C NMR (125MHz, CDCl\(_3\)) \(\delta\) 55.3, 115.0, 123.4, 123.5, 127.1, 130.1, 130.5, 137.3, 142.0, 142.8,
4-(3',4'-Dimethoxyphenyl)-5-phenylpenta-(2E,4Z)-dienoyl azide [(2E,4Z)-5h]. Yield 93% from acid (2E,4Z)-11h; yellow solid. $^1$H NMR (400MHz, CDCl$_3$) $\delta$ 3.79 (3H, s), 3.05 (3H, s), 5.55 (1H, d, $J$ = 15.2 Hz), 6.62 (1H, d, $J$ = 1.9 Hz), 6.69 (1H, dd, $J$ = 8.1, 1.9 Hz), 6.93 (1H, d, $J$ = 8.1 Hz), 6.96 (1H, s), 7.01 (2H, dd, $J$ = 7.6, 1.8 Hz), 7.16 (3H, m), 7.73 (1H, d, $J$ = 15.2 Hz); $^{13}$C NMR (100MHz, CDCl$_3$) $\delta$ 55.9, 56.0, 111.9, 112.0, 121.0, 121.4, 128.3, 128.7, 128.8, 130.2, 135.4, 139.1, 140.8, 148.8, 149.7, 151.9, 172.2; IR (KBr) 2141, 1686 cm$^{-1}$; EIMS $m/z$ (rel int) 307 (100, [M-N$_2$]$^+$); HREIMS $m/z$ calcd for C$_{19}$H$_{17}$NO$_3$: 307.1208, found: 307.1202 [M-N$_2$]$^+$.

4-(3',4'-Dimethoxyphenyl)-5-(4''-methoxyphenyl)penta-(2E,4Z)-dienoyl azide [(2E,4Z)-5i]. Yield 95% from acid (2E,4Z)-11i; yellow solid. $^1$H NMR (400MHz, CDCl$_3$) $\delta$ 3.76 (3H, s), 3.81 (3H, s), 3.95 (3H, s), 5.48 (1H, d, $J$ = 15.1 Hz), 6.63 (1H, s), 6.69 (3H, m), 6.94 (4H, m), 7.71 (1H, d, $J$ = 15.1 Hz); $^{13}$C NMR (100MHz, CDCl$_3$) $\delta$ 55.1, 55.8, 55.9, 111.9, 112.0, 113.8, 119.6, 121.3, 128.1, 129.0, 131.9, 136.9, 140.7, 148.6, 149.7, 152.3, 160.0, 172.1; IR (KBr) 2137, 1674 cm$^{-1}$; EIMS $m/z$ (rel int) 337 (100, [M-N$_2$]$^+$); HREIMS $m/z$ calcd for C$_{20}$H$_{19}$N$_3$O$_4$: 337.1314, found: 337.13223 [M-N$_2$]$^+$.

4-(3',4'-Dimethoxyphenyl)-5-(4''-nitrophenyl)penta-(2E,4Z)-dienoyl azide [(2E,4Z)-5j]. Yield 95% from acid (2E,4Z)-11j; yellow solid. $^1$H NMR (400MHz, CDCl$_3$) $\delta$ 3.80 (3H, s), 3.95 (3H, s), 5.67 (1H, d, $J$ = 15.2 Hz), 6.61 (1H, d, $J$ = 1.9 Hz), 6.68 (1H, dd, $J$ = 8.2, 1.9 Hz), 6.95 (1H, d, $J$ = 8.2 Hz), 7.00 (1H, s), 7.15 (2H, d, $J$ = 8.9 Hz), 7.72 (1H, d, $J$ = 15.2 Hz), 8.00 (2H, d, $J$ = 8.9 Hz); $^{13}$C NMR (100MHz, CDCl$_3$) $\delta$ 55.8, 55.9, 111.6, 112.0, 121.2, 123.3, 123.4, 127.4, 130.5, 137.1, 141.8, 142.8, 146.8, 149.2, 149.8, 150.2, 171.8; IR (KBr) 2141, 1684 cm$^{-1}$; EIMS $m/z$ (rel int) 352 (100, [M-N$_2$]$^+$); HREIMS $m/z$ calcd for C$_{19}$H$_{16}$N$_2$O$_5$: 352.1059, found: 352.1051 [M-N$_2$]$^+$.

4,5-Bis(3',4'-dimethoxyphenyl)penta-(2E,4Z)-dienoyl azide [(2E,4Z)-5k]. Yield 92% from acid (2E,4Z)-11k; yellow solid. $^1$H NMR (400MHz, CDCl$_3$) $\delta$ 3.47 (3H, s), 3.81 (3H, s), 3.83 (3H, s), 3.91
(3H, s), 5.49 (1H, d, J = 15.1Hz), 6.46 (1H, s), 6.67 (1H, s), 6.72(3H, m), 6.91 (1H, s), 6.96 (1H, d, J = 8.1 Hz), 7.72 (1H, d, J = 15.1 Hz); $^{13}$C NMR (100MHz, CDCl$_3$) $\delta$ 54.9, 55.5, 55.8 (2×C), 110.4, 111.8, 111.9, 112.0, 119.6, 121.4, 124.8, 128.2, 128.9, 136.9, 140.7, 148.1, 148.5, 149.6, 149.7, 151.9, 171.9; IR (KBr) 2136, 1687 cm$^{-1}$; EIMS $m/z$ (rel int) 395 (1, M$^+$); HREIMS $m/z$ calcd for C$_{21}$H$_{21}$N$_3$O$_5$: 395.1481, found: 395.1488 [M$^+$].

5-(3",4"-Dimethoxyphenyl)-4-(4'-nitrophenyl)penta-(2$^E$,4$^Z$)-dienoyl azide [(2$^E$, 4$^Z$)-5l]. Yield 96% from acid (2$^E$,4$^Z$)-11l; yellow solid. $^1$H NMR (400MHz, CDCl$_3$) $\delta$ 3.48 (3H, s), 3.83 (3H, s), 5.32 (1H, d, J = 15.4 Hz), 6.35 (1H, s), 6.63 (1H, d, J = 8.4 Hz), 6.70 (1H, d, J = 8.4 Hz), 7.03 (1H, s), 7.41 (2H, d, J = 8.4 Hz), 7.73 (1H, d, J = 15.4 Hz), 8.33 (2H, d, J = 8.4 Hz); $^{13}$C NMR (100MHz, CDCl$_3$) $\delta$ 55.2, 55.8, 110.9, 112.3, 119.9, 124.5, 124.6, 127.3, 130.8, 134.8, 141.8, 144.2, 147.6, 148.5, 150.2, 150.3, 171.7; IR (KBr) 2133, 1682 cm$^{-1}$; EIMS $m/z$ (rel int) 380 (1, [M-N$_2$]$^+$); HREIMS $m/z$ calcd for C$_{19}$H$_{16}$N$_4$O$_5$: 380.1121, found: 380.1128 [M-N$_2$]$^+$. 

**General Procedure for the Iodine-Catalyzed Cyclization of an 4,5-Diphenylpenta-2,4-dienoyl Azide, 5.** A mixture of azide 5 (1 mmol) and a flake of iodine in o-dichlorobenzene (5 mL) was heated at 150 $^\circ$C under N$_2$ for 1-7 h. Reaction completion was monitored by the disappearance of the spots of isocyanate 1 and dimer 6 on the TLC plate. After cooling, the resulting solution was directly purified by column chromatography over silica gel and eluted with hexane-CH$_2$Cl$_2$ to give 3,4-diphenylpyridin-2(1$H$)-one 2 and 2-phenynaphthalene 3 or 1($E$)-benzylidene-1$H$-indene 4. The full spectral data of these compounds are described as follows.

**3,4-Diphenylpyridin-2(1$H$)-one (2a).** Yield 32%; white granules, mp 284–285°C (hexane-CHCl$_3$) (lit.,$^{29}$ mp 284–284.5°C). $^1$H NMR (500MHz, DMSO-$d_6$) $\delta$ 6.22 (1H, d, J = 6.7 Hz), 7.04 (4H, m), 7.15 (6H, m), 7.41 (1H, d, J = 6.7 Hz), 11.78 (1H, br s); $^{13}$C NMR (125MHz, DMSO-$d_6$) $\delta$ 107.9, 126.7, 127.6, 127.8, 128.2, 128.9, 129.3, 131.2, 134.0, 136.1, 139.5, 150.5, 162.1; IR (KBr) 1643 cm$^{-1}$; EIMS
2-Phenylnaphthalene (3a). Yield 60%; pale yellow granule, mp 100–101°C (hexane-CHCl₃) (lit.,¹⁰ mp 100–101°C). ¹H NMR (400MHz, CDCl₃) δ 7.37 (1H, t, J = 7.3 Hz), 7.48 (4H, m), 7.73 (3H, m), 7.88 (3H, m), 8.04 (1H, s); ¹³C NMR (100MHz, CDCl₃) δ 125.6, 125.8, 125.9, 126.2, 127.3, 127.4, 127.6, 128.2, 128.4, 128.8, 132.6, 133.7, 138.5, 141.1; IR (KBr) 2925, 1597 cm⁻¹; EIMS m/z (rel int) 204 (100, M⁺). Anal. Calcd for C₁₆H₁₂: C, 94.08; H, 5.92. Found: C, 94.09; H, 6.25.

3-(4"-Methoxyphenyl)-4-phenylpyridin-2(1H)-one (2b). Yield 32%; white granules, mp 249–251°C (hexane-CHCl₃). ¹H NMR (600MHz, CDCl₃) δ 3.77 (3H, s), 6.35 (1H, d, J = 6.6 Hz), 6.77 (2H, d, J = 6.6 Hz), 7.10 (4H, m), 7.21 (3H, d, J = 6.6 Hz), 7.36 (1H, d, J = 6.6 Hz); ¹³C NMR (150MHz, CDCl₃) δ 55.1, 109.7, 113.4, 127.4, 127.6, 128.0, 128.9, 129.2, 132.2, 132.7, 139.4, 151.7, 158.5, 164.8; IR (KBr) 1637 cm⁻¹; EIMS m/z (rel int) 277 (81, M⁺). Anal. Calcd for C₁₈H₁₅NO₂: C, 77.96; H, 5.45; N, 5.05. Found: C, 78.25; H, 5.10; N, 5.16.

2-Methoxy-6-phenylnaphthalene (3b). Yield 50%; pale yellow granules, mp 151–152°C (hexane-CHCl₃) (lit.,³⁰ mp 150–151°C). ¹H NMR (500MHz, CDCl₃) δ 3.92 (3H, s), 7.16 (2H, m), 7.34 (1H, t, J = 7.7 Hz), 7.46 (2H, t, J = 7.7 Hz), 7.70 (3H, m), 7.77 (1H, d, J = 8.2 Hz), 7.79 (1H, d, J = 8.2 Hz), 7.96 (1H, s); ¹³C NMR (125MHz, CDCl₃) δ 55.3, 105.6, 119.1, 125.6, 126.0, 127.0, 127.2, 127.3, 128.8, 129.2, 129.7, 133.8, 136.4, 141.2, 157.8; IR (KBr) 2960, 1603 cm⁻¹; EIMS m/z (rel int) 234 (100, M⁺). Anal. Calcd for C₁₇H₁₄O: C, 87.15; H, 6.02. Found: C, 86.84; H, 6.26.

3-(3',4'-Dimethoxyphenyl)-4-phenylpyridin-2(1H)-one (2c). Yield 8%; white granules, mp 228–230°C (hexane-CHCl₃). ¹H NMR (500MHz, CDCl₃) δ 3.60 (3H, s), 3.85 (3H, s), 6.36 (1H, d, J = 6.7 Hz), 6.60 (1H, s), 6.78 (1H, d, J = 8.3 Hz), 6.88 (1H, d, J = 8.3 Hz), 7.10 (2H, m), 7.21 (3H, m), 7.38 (1H, d, J = 6.7 Hz), 12.90 (1H, br s); ¹³C NMR (125MHz, CDCl₃) δ 55.6, 55.7, 109.6, 110.7, 114.6,
2,3-Dimethoxy-6-phenylnaphthalene (3c). Yield 83%; white needles, mp 136–137°C (hexane-EtOAc) (lit., mp 134–135°C). \(^1\)H NMR (400MHz, CDCl\(_3\)) \(\delta\) 3.98 (6H, s), 7.11 (1H, s), 7.14 (1H, s), 7.34 (1H, \(J = 7.5\) Hz), 7.45 (2H, \(J = 7.5\) Hz), 7.58 (1H, dd, \(J = 8.4, 1.7\) Hz), 7.70 (3H, m), 7.88 (1H, s); \(^13\)C NMR (100MHz, CDCl\(_3\)) \(\delta\) 55.8 (2×C), 105.9, 106.4, 123.7, 124.3, 126.7, 126.9, 127.1, 128.3, 128.7, 129.3, 136.8, 141.2, 149.4, 149.7; IR (KBr) 2963, 1601 cm\(^{-1}\); EIMS \(m/z\) (rel int) 264 (100, M\(^+\)).


4-(4’-Methoxyphenyl)-3-phenylpyridin-2(1\(H\))-one (2d). Yield 63%; white granules, mp 245–246°C (hexane-CHCl\(_3\)). \(^1\)H NMR (400MHz, DMSO-\(d_6\)) \(\delta\) 3.69 (3H, s), 6.21 (1H, \(d, J = 6.8\) Hz), 6.74 (2H, \(d, J = 8.6\) Hz), 6.99 (2H, \(d, J = 8.6\) Hz), 7.05 (2H, \(d, J = 7.6\) Hz), 7.16 (3H, m), 7.36 (1H, \(d, J = 6.8\) Hz), 11.57 (1H, br s); \(^13\)C NMR (100MHz, DMSO-\(d_6\)) \(\delta\) 55.0, 107.7, 113.5, 126.3, 127.3, 128.6, 130.0, 130.9, 131.4, 133.5, 136.2, 149.8, 158.7, 161.9; IR (KBr) 1639 cm\(^{-1}\); EIMS \(m/z\) (rel int) 277 (74, M\(^+\)). Anal. Calcd for C\(_{18}\)H\(_{15}\)NO\(_2\): C, 77.96; H, 5.45; N, 5.05. Found: C, 77.91; H, 5.68; N, 5.05.

2-(4’-Methoxyphenyl)naphthalene (3d). Yield 22%; pale yellow granules, mp 138–139°C (hexane-EtOAc) (lit., mp 139–140°C). \(^1\)H NMR (500MHz, CDCl\(_3\)) \(\delta\) 3.87 (3H, s), 7.02 (2H, \(d, J = 8.7\) Hz), 7.47 (2H, m), 7.66 (2H, \(d, J = 8.7\) Hz), 7.71 (1H, \(d, J = 8.7\) Hz), 7.86 (3H, m), 7.98 (1H, s); \(^13\)C NMR (125MHz, CDCl\(_3\)) \(\delta\) 55.4, 114.3, 125.0, 125.4, 125.6, 126.2, 127.6, 128.0, 128.3, 132.3, 133.6, 133.8, 138.2, 159.3; IR (KBr) 2947, 1597 cm\(^{-1}\); EIMS \(m/z\) (rel int) 234 (100, M\(^+\)); HREIMS \(m/z\) calcd for C\(_{17}\)H\(_{14}\)O: 234.1045, found: 234.1039 [M\(^+\)].

3,4-Bis(4’-methoxyphenyl)pyridin-2(1\(H\))-one (2e). Yield 58%; white granules, mp 254–256°C (hexane-CHCl\(_3\)). \(^1\)H NMR (600MHz, DMSO-\(d_6\)) \(\delta\) 3.69 (3H, s), 3.70 (3H, s), 7.02 (2H, \(d, J = 6.6\) Hz), 6.75 (2H, \(d, J = 8.4\) Hz), 6.78 (2H, \(d, J = 8.4\) Hz), 6.97 (2H, \(d, J = 9.0\) Hz), 7.01 (2H, \(d, J = 9.0\) Hz), 7.35 (1H, \(d, J = 6.6\) Hz), 11.66 (1H, br s); \(^13\)C NMR (150MHz, DMSO-\(d_6\)) \(\delta\) 54.9, 55.0, 107.8, 112.9,
113.5, 128.1 (2×C), 130.1, 131.6, 132.1, 133.1, 149.4, 157.7, 158.5, 162.1; IR (KBr) 1636 cm⁻¹; EIMS m/z (rel int) 307 (95, M⁺). Anal. Calcd for C₁₉H₁₇NO₃: C, 74.25; H, 5.58; N, 4.56. Found: C, 73.97; H, 5.87; N, 4.65.

2-Methoxy-6-(4'-methoxyphenyl)naphthalene (3e). Yield 25%; white granules, mp 201–202°C (hexane-CHCl₃) (lit., ³³ mp 190°C). ¹H NMR (400MHz, CDCl₃) δ 3.86 (3H, s), 3.93 (3H, s), 7.00 (2H, d, J = 8.8 Hz), 7.16 (2H, m), 7.63 (2H, d, J = 8.8 Hz), 7.67 (1H, dd, J = 8.5, 1.9 Hz), 7.77 (1H, d, J = 8.5 Hz), 7.78 (1H, d, J = 8.5 Hz), 7.91 (1H, d, J = 1.9 Hz); ¹³C NMR (100MHz, CDCl₃) δ 55.3, 55.4, 105.6, 114.3, 119.1, 124.9, 125.9, 127.2, 128.2, 129.3, 129.5, 133.4, 133.8, 136.0, 157.6, 159.0; IR (KBr) 2932, 1605 cm⁻¹; EIMS m/z (rel int) 264 (100, M⁺). Anal. Calcd for C₁₈H₁₆O₂: C, 81.79; H, 6.10. Found: C, 81.42; H, 5.94.

3-(3″,4″-Dimethoxyphenyl)-4-(4'-methoxyphenyl)pyridin-2(1H)-one (2f). Yield 11%; white granules, mp 284–285°C (hexane-CHCl₃). ¹H NMR (400MHz, DMSO-d₆) δ 3.51 (3H, s), 3.70 (6H, s), 6.18 (1H, d, J = 6.6 Hz), 6.62 (2H, m), 6.77 (3H, m), 7.02 (2H, d, J = 8.4 Hz), 7.33 (1H, d, J = 6.6 Hz), 11.50 (1H, br s); ¹³C NMR (100MHz, DMSO-d₆) δ 55.1, 55.5 (2xC), 107.7, 111.3, 113.5, 115.8, 123.6, 128.4, 128.5, 129.9, 131.8, 133.1, 147.6, 147.8, 149.6, 158.6, 161.9; IR (KBr) 1636 cm⁻¹; EIMS m/z (rel int) 337 (100, M⁺); HREIMS m/z calcd for C₂₀H₁₉NO₄: 337.1314, found: 337.1304 [M⁺].

2,3-Dimethoxy-6-(4′-methoxyphenyl)naphthalene (3f). Yield 81%; white needles, mp 164–165°C (hexane-CHCl₃) (lit., ³⁴ mp 158–159°C). ¹H NMR (400MHz, CDCl₃) δ 3.83 (3H, s), 3.98 (3H, s), 3.99 (3H, s), 6.99 (2H, d, J = 8.7 Hz), 7.10 (1H, s), 7.14 (1H, s), 7.55 (1H, dd, J = 8.4, 1.5 Hz), 7.61 (2H, d, J = 8.7 Hz), 7.70 (1H, d, J = 8.4 Hz), 7.83 (1H, s); ¹³C NMR (100MHz, CDCl₃) δ 55.3, 55.8 (2xC), 106.0, 106.4, 114.2, 123.6 (2xC), 126.7, 128.0, 128.1, 129.5, 133.8, 136.5, 149.3, 149.7, 159.0; IR (KBr) 2963, 1605 cm⁻¹; EIMS m/z (rel int) 294 (100, M⁺). Anal. Calcd for C₁₉H₁₈O₃: C, 77.53; H, 6.16. Found: C, 77.52; H, 6.42.
4-(4'-Methoxyphenyl)-3-(4''-nitrophenyl)pyridin-2(1H)-one (2g). Yield 88%; pale yellow granules, mp 272–274°C (hexane-CHCl₃). ¹H NMR (500MHz, CDCl₃) δ 3.77 (3H, s), 6.41 (1H, d, J = 6.8 Hz), 6.75 (2H, d, J = 8.6 Hz), 6.99 (2H, d, J = 8.6 Hz), 7.40 (3H, m), 8.11 (2H, d, J = 8.6 Hz), 12.93 (1H, s); ¹³C NMR (125MHz, CDCl₃) δ 55.2, 109.9, 113.9, 123.1, 126.9, 130.2, 130.3, 132.2, 133.9, 143.0, 146.6, 152.8, 159.8, 163.8; IR (KBr) 1639 cm⁻¹; EIMS m/z (rel int) 322 (87, M⁺); HREIMS m/z calcd for C₁₈H₁₄N₂O₄: 322.0954, found: 322.0958 [M⁺].

4-(3',4'-Dimethoxyphenyl)-3-phenylpyridin-2(1H)-one (2h). Yield 55%; white granules, mp 250–252°C (hexane-CHCl₃). ¹H NMR (400MHz, DMSO-d₆) δ 3.36 (3H, s), 3.69 (3H, s), 6.28 (1H, d, J = 8.3 Hz), 6.74 (1H, d, J = 1.9 Hz), 6.47 (1H, d, J = 1.9 Hz), 6.82 (1H, d, J = 8.3 Hz), 7.05 (2H, d, J = 8.3 Hz), 7.18 (3H, m), 7.39 (1H, d, J = 6.8 Hz), 11.7 (1H, br s); ¹³C NMR (100MHz, DMSO-d₆) δ 55.1, 55.5, 107.7, 111.4, 113.3, 121.4, 126.5, 127.6, 128.7, 131.1, 131.4, 133.7, 136.7, 147.9, 148.5, 150.0, 162.1; IR (KBr) 1636 cm⁻¹; EIMS m/z (rel int) 307 (94, M⁺); HREIMS m/z calcd for C₁₉H₁₇NO₃: 307.1208, found: 307.1202 [M⁺].

1(⁵)-Benzylidene-5,6-dimethoxy-1H-indene (4h). Yield 32%; yellow granules, mp 104–105°C (hexane-CHCl₃). ¹H NMR (400MHz, CDCl₃) δ 3.91 (3H, s), 3.96 (3H, s), 6.89 (3H, m), 7.27 (1H, s), 7.32 (1H, t, J = 7.3 Hz), 7.37 (1H, s), 7.41 (2H, t, J = 7.3 Hz), 7.58 (2H, d, J = 7.3 Hz); ¹³C NMR (100MHz, CDCl₃) δ 56.1, 56.4, 104.0, 104.9, 124.8, 127.8, 128.1, 128.6, 130.0, 130.1, 134.1, 135.4, 137.0, 140.4, 147.5, 149.3; IR (KBr) 2970, 1597 cm⁻¹; EIMS m/z (rel int) 264 (100, M⁺). Anal. Calcd for C₁₉H₁₆O₂: C, 81.79; H, 6.10. Found: C, 81.49; H, 6.17.

4-(3',4'-Dimethoxyphenyl)-3-(4''-methoxyphenyl)pyridin-2(1H)-one (2i). Yield 45%; white granules, mp 286–288°C (hexane-CHCl₃). ¹H NMR (500MHz, DMSO-d₆) δ 3.42 (3H, s), 3.69 (6H, s), 6.25 (1H, d, J = 6.8 Hz), 6.51 (1H, d, J = 1.8 Hz), 6.72 (1H, dd, J = 8.3, 1.8 Hz), 6.77 (2H, d, J = 8.7 Hz), 6.83 (1H, d, J = 8.3 Hz), 6.97 (2H, d, J = 8.7 Hz), 7.35 (1H, d, J = 6.8 Hz), 11.64 (1H, s); ¹³C NMR (125MHz, DMSO-d₆) δ 55.2 (2×C), 55.5, 107.8, 111.4, 113.1, 113.2, 121.4, 128.4, 128.6, 131.7, 132.2,
133.3, 147.9, 148.4, 149.7, 158.0, 162.3; IR (KBr) 1636 cm⁻¹; EIMS m/z (rel int) 337 (100, M⁺); HREIMS m/z calcd for C₂₀H₁₉NO₄: 337.1314, found: 337.1307 [M⁺].

1(E)-(4''-Methoxybenzylidene)-5,6-dimethoxy-1H-indene (4i). Yield 25%; yellow granules, mp 134–135°C (hexane-CHCl₃). ¹H NMR (500MHz, CDCl₃) δ 3.86 (3H, s), 3.93 (3H, s), 3.97 (3H, s), 6.91 (3H, m), 6.96 (2H, d, J = 8.7 Hz), 7.26 (1H, s), 7.33 (1H, s), 7.57 (2H, d, J = 8.7 Hz); ¹³C NMR (125MHz, CDCl₃) δ 55.3, 56.1, 56.4, 103.7, 104.7, 114.2, 124.6, 127.7, 129.7, 130.3, 131.6, 133.4, 135.1, 138.5, 147.4, 149.0, 159.8; IR (KBr) 2928, 1601 cm⁻¹; EIMS m/z (rel int) 294 (100, M⁺). Anal. Calcd for C₁₉H₁₈O₃: C, 77.53; H, 6.16. Found: C, 77.68; H, 5.97.

4-(3',4'-Dimethoxyphenyl)-3-(4''-nitrophenyl)pyridin-2(1H)-one (2j). Yield 30%; pale yellow granules, mp 256–257°C (hexane-CHCl₃). ¹H NMR (400MHz, CDCl₃) δ 3.59 (3H, s), 3.85 (3H, s), 6.45 (1H, d, J = 6.7 Hz), 6.50 (1H, d, J = 1.9 Hz), 6.69 (1H, dd, J = 8.3, 1.9 Hz), 6.74 (1H, d, J = 8.3 Hz), 7.41 (3H, m), 8.12 (2H, d, J = 8.9 Hz), 13.11 (1H, br s); ¹³C NMR (100MHz, CDCl₃) δ 55.7, 55.8, 109.8, 110.9, 112.3, 121.8, 123.1, 126.8, 130.4, 132.1, 134.1, 143.1, 146.6, 148.6, 149.4, 152.9, 163.9; IR (KBr) 1641 cm⁻¹; EIMS m/z (rel int) 352 (100, M⁺); HREIMS m/z calcd for C₁₉H₁₆N₂O₅: 352.1059, found: 352.1061 [M⁺].

1(E)-(4''-Nitrobenzylidene)-5,6-dimethoxy-1H-indene (4j). Yield 67%; black granules, mp 130–131°C (hexane-CHCl₃). ¹H NMR (400MHz, CDCl₃) δ 3.91 (3H, s), 3.96 (3H, s), 3.94 (3H, s), 6.73 (1H, d, J = 5.6 Hz), 6.86 (1H, s), 6.93 (1H, d, J = 5.6 Hz), 7.23 (1H, s), 7.29 (1H, s), 7.65 (2H, d, J = 8.6 Hz), 8.22 (2H, d, J = 8.6 Hz); ¹³C NMR (100MHz, CDCl₃) δ 56.0, 56.3, 104.4, 105.1, 123.7, 123.8, 124.4, 129.3, 130.4, 135.6, 136.4, 143.4, 143.5, 146.8, 147.7, 149.9; IR (KBr) 2988, 1589, 1513 cm⁻¹; EIMS m/z (rel int) 309 (100, M⁺). Anal. Calcd for C₁₉H₁₅NO₄: C, 69.89; H, 4.89; N, 4.53. Found: C, 69.55; H, 5.02; N, 4.26.

3,4-Bis(3',4'-dimethoxyphenyl)pyridin-2(1H)-one (2k). Yield 12%; white granules, mp 300°C (hexane-CHCl₃). ¹H NMR (400MHz, CDCl₃) δ 3.57 (3H, s), 3.69 (3H, s), 3.85 (3H, s), 3.86 (3H, s),
2,3-Dimethoxy-6-(3',4'-dimethoxyphenyl)naphthalene (3k). Yield 61%; yellow granules, mp 165–166°C (hexane-CHCl₃) (lit., mp 168–170°C). ¹H NMR (400MHz, CDCl₃) δ 3.94 (3H, s), 3.99 (3H, s), 4.02 (3H, s), 6.98 (1H, d, J = 8.2 Hz), 7.14 (1H, s), 7.23 (1H, m), 7.26 (1H, s), 7.57 (1H, dd, J = 8.2, 1.8 Hz), 7.74 (1H, d, J = 8.2 Hz), 7.85 (1H, s); ¹³C NMR (100MHz, CDCl₃) δ 55.9 (2×C), 56.0 (2×C), 106.1, 106.5, 110.7, 111.6, 119.5, 123.7, 123.8, 126.8, 128.2, 129.5, 134.5, 136.9, 148.5, 149.3, 149.5, 149.9; IR (KBr) 2928, 1601 cm⁻¹; EIMS m/z (rel int) 324 (100, M⁺). Anal. Calcd for C₂₀H₂₀O₄: C, 74.06; H, 6.21. Found: C, 74.39; H, 6.62.

3-(3",4"-Dimethoxyphenyl)-4-(4'-nitrophenyl)pyridin-2(1H)-one (2l). Yield 13%; yellow needles, mp 243–245°C (hexane-EtOAc). ¹H NMR (500MHz, CDCl₃) δ 3.70 (3H, s), 3.85 (3H, s), 6.34 (1H, d, J = 6.6 Hz), 6.69 (1H, s), 6.72 (1H, d, J = 8.2 Hz), 6.76 (1H, d, J = 8.2 Hz), 6.79 (2H, d, J = 8.3 Hz), 7.44 (1H, d, J = 6.6 Hz), 8.09 (2H, d, J = 8.3 Hz), 13.36 (1H, br s); ¹³C NMR (125MHz, CDCl₃) δ 55.7, 55.8, 108.7, 110.9, 114.1, 123.4, 123.7, 126.5, 129.8, 130.2, 133.4, 146.1, 147.1, 148.5, 148.6, 149.4, 164.5; IR (KBr) 1636 cm⁻¹; EIMS m/z (rel int) 352 (100, M⁺). Anal. Calcd for C₁₉H₁₆N₂O₅: C, 64.77; H, 4.58; N, 7.95. Found: C, 64.51; H, 4.84; N, 7.63.

2,3-Dimethoxy-6-(4'-nitrophenyl)naphthalene (3l). Yield 77%; yellow granules, mp 169–170°C (hexane-EtOAc). ¹H NMR (400MHz, CDCl₃) δ 4.01 (6H, s), 7.13 (1H, s), 7.18 (1H, s), 7.56 (1H, dd, J = 8.5, 1.9 Hz), 7.77 (3H, m), 7.91 (1H, s), 8.26 (2H, d, J = 8.5 Hz); ¹³C NMR (100MHz, CDCl₃) δ 55.8 (2×C), 106.0, 106.7, 123.1, 124.0, 125.1, 127.2, 127.5, 129.2, 129.3, 134.2, 146.7, 147.7, 150.1, 150.3;
IR (KBr) 2966, 1591 cm⁻¹; EIMS m/z (rel int) 309 (100, M⁺). Anal. Calcd for C₁₈H₁₅NO₄: C, 69.89; H, 4.89; N, 4.53. Found: C, 69.90; H, 5.15; N, 4.73.

**N-(3,4-Diphenylbuta-1,3-dienylcarbamoyl)-3,4-diphenyl-1H-pyridin-2-one (6a).** Yellow granules, mp 233–235°C (hexane-EtOAc). ¹H NMR (500MHz, CDCl₃) δ 6.42 (1H, d, J = 14.2 Hz), 6.46 (1H, s), 6.53 (1H, d, J = 7.7 Hz), 6.56 (1H, dd, J = 14.2, 10.0 Hz), 6.84 (2H, m), 7.06 (3H, m), 7.10 (2H, m), 7.13 (2H, m), 7.22 (6H, m), 7.27 (2H, m), 7.38 (3H, m), 8.41 (1H, d, J = 7.8 Hz), 12.68 (1H, d, J = 10.0 Hz); ¹³C NMR (125MHz, CDCl₃) δ 110.6, 123.1, 124.8, 126.6, 127.7 (2×C), 127.9, 128.1, 128.2, 128.5, 128.8, 129.1, 129.2, 129.4, 129.7, 130.0, 130.8, 131.3, 134.6, 135.6, 137.7, 137.9, 139.7, 149.7, 151.9, 164.8; IR (KBr) 1721, 1634 cm⁻¹; ESIMS m/z (rel int) 495 (100, [M+1]+). Anal. Calcd for C₃₄H₂₆N₂O₂: C, 82.57; H, 5.30; N, 5.66. Found: C, 82.35; H, 5.62; N, 5.60.

**X-ray crystallographic data of 4j.** Colorless, single crystals of 4j suitable for X-ray diffraction study were grown by recrystallization from hexane-CH₂Cl₂. Data were obtained on an Oxford Gemini S diffractometer with graphite-monochromated Mo Kα radiation, operating at 50 kV and 40 mA at 297 K over a 2θ range of 5.68 to 58.48°. Data were processed on a Pentium III PC using the Bruker AXS SHELXTL, NT software package. 1(E)-(4"-nitrobenzylidene)-5,6-dimethoxy-1H-indene (4j):

C₁₈H₁₅NO₄, Mr = 309.31; Crystal size 0.60 × 0.26 × 0.17 mm³; monoclinic, space group P2/c; Unit cell dimensions: a = 15.717, b = 12.299, c = 15.454 Å, α = 90, β = 95.11, γ = 90°; Volume: 2975.4 Å³; Z = 8; Dc = 1.381 Mg/m³. The structures were refined by full-matrix least-squares on F² using SHEEXL-97 (Sheldrick, 2008). Final discrepancy indices of R₁ = 0.0431, wR₂ = 0.0699 and GOOF = 0.957 were obtained for observed data with I > 2σ(I). Crystallographic data for the structure 4j (CCDC-837686) reported in this paper have been deposited with the Cambridge Crystallographic Data Centre. Copies of the data can be obtained free of charge via the Internet at [http://www.ccdc.cam.ac.uk/conts/retrieving.html](http://www.ccdc.cam.ac.uk/conts/retrieving.html) (or from the CCDC, 12 Union Road, Cambridge CB2 1EZ, U.K.; fax: +44 1223 336033; e-mail: deposit@ccdc.cam.ac.uk)
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**Supporting Information Available:** The copies of the $^1$H and $^{13}$C NMR spectra of all compounds 2-6 and 9-11, and X-ray crystallographic data, including CIF file for 4j, are available free of charge via the Internet at http://pub.acs.org.

**References and Footnotes**


