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

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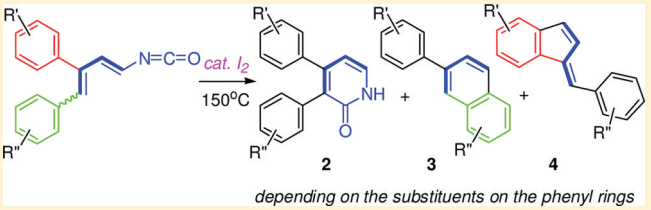
Supporting Information

**ABSTRACT:** 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.

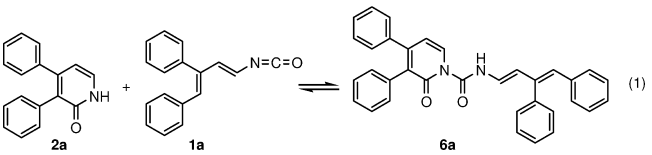



## INTRODUCTION

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.[1−8] Eloy and Washburne extensively applied the thermal cyclization of penta-2,4-dienoyl isocyanates to synthesize 1*H*-pyridin-2-ones.[1,4] 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.[9] 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.[10] 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(1*H*)-ones **2** but also phenylnaphthalenes **3** or benzylidene-1*H*-indenes **4** were obtained by the iodine-catalyzed thermal cyclization of isocyanates, generated *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(1*H*)-ones **2**, phenylnaphthalene **3**, and benzylidene-1*H*-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-(2*E*,4*Z*)-dienoyl azide [(2*E*,4*Z*)-**5a**] in diphenyl ether at 240 °C for 3 h gave 3,4-diphenylpyridin-2(1*H*)-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.[11] A less polar product, **6a**, and 2-phenylnaphthalene **3a** were obtained in the initial trial while the isocyanate of azide (2*E*,4*Z*)-**5a** disappeared within 1 h.[9] The product **6a** was deduced as *N*-(3,4-diphenylbuta-1,3-dienylcarbamoyl)-3,4-diphenyl-1*H*-pyridin-2-one, an adduct of 3,4-diphenylpyridinone **2a** and 3,4-diphenylbuta-1,3-dienyl isocyanate **1a** (eq 1). As




(1)

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.

Apparently, the unexpected product **3a** was produced via the intramolecular cyclization of the triene,[12] 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.[13,14] Unfortunately, only Curtius rearrangement products (1*E*,3*Z*)-**1a** and (1*E*,3*E*)-**1a** were obtained quantitatively from (2*E*,4*Z*)-**5a** after refluxing in *o*-

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**Scheme 1. Iodine-Catalyzed Cyclization of 3,4-Diphenylpenta-1,3-dienoyl Isocyanates 1**

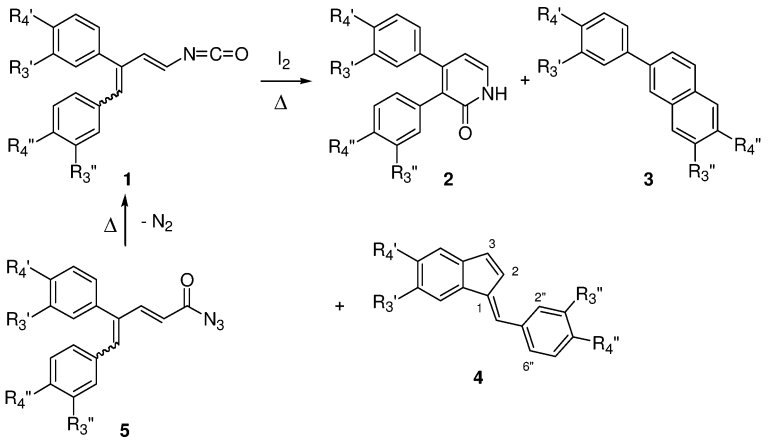


**Table 1. Formation of 2a and 3a from Dienoyl Azide (2*E*,4*Z*)-5a**

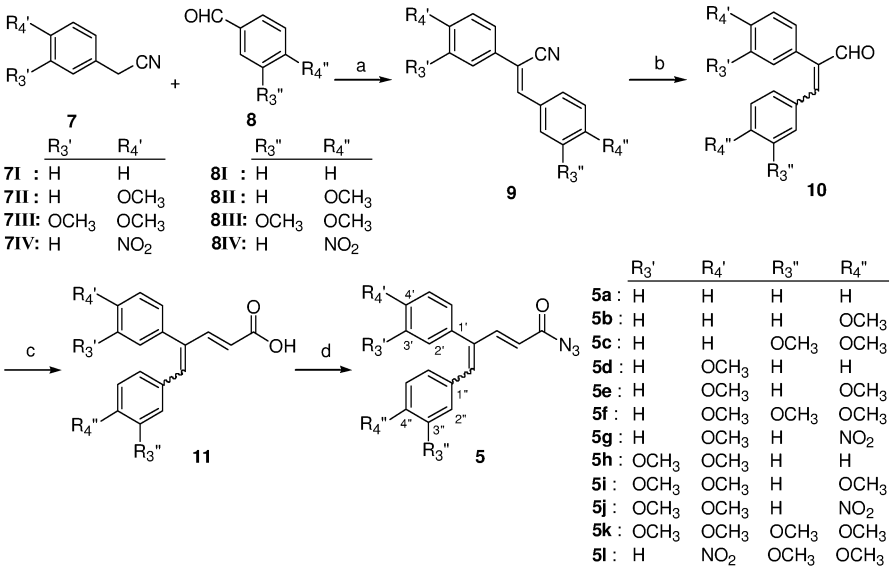
| | | | | product (%) | |
|---|---|---|---|---|---|
| entry | solvent | temp (°C) | time (h) | **2a** | **3a** |
| 1[a] | diphenyl ether | 240 | 3 | 11 | 9 |
| 2[a] | *o*-dichlorobenzene | reflux | 5 | – | – |
| 3 | *o*-dichlorobenzene | reflux | 5 | 66 | 25 |
| 4 | *o*-dichlorobenzene | 150 | 7 | 32 | 60 |
| 5 | *o*-dichlorobenzene | 130 | 15 | 45 | 40 |
| 6 | *o*-dichlorobenzene | 110 | 96 | 40 | 20 |
| 7[b] | *o*-dichlorobenzene | 150 | 7 | 31 | 60 |

[a]No iodine added. [b]Dienoyl azide (2*E*,4*E*)-**5a** was used.

dichlorobenzene (bp 180 °C) for 5 h (entry 2). We successfully used 2 mol % 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(1*H*)-one **2a** (66%) and butadienylbenzene into naphthalene **3a** (25%, entry 3). Because of the steric effects, the C-1 and C-2 isomerization of (1*E*,3*Z*)-**1a** to (1*Z*,3*Z*)-**1a** is slightly more difficult than the C-3 and C-4 isomerization of (1*E*,3*Z*)-**1a** to (1*E*,3*E*)-**1a**. Therefore, the cyclization, proceeding under milder temperature conditions (150 °C), resulted in naphthalene **3a** 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). We proposed the yield in naphthalene **3a** would decrease, probably because the loss of cyanic acid would be slightly more difficult below 150 °C. Intriguingly, heating a solution of the geometric isomer (2*E*,4*E*)-**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**.

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

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



| | $R_3'$ | $R_4'$ | $R_3''$ | $R_4''$ |
|---|---|---|---|---|
| **5a** : | H | H | H | H |
| **5b** : | H | H | H | $OCH_3$ |
| **5c** : | H | H | $OCH_3$ | $OCH_3$ |
| **5d** : | H | $OCH_3$ | H | H |
| **5e** : | H | $OCH_3$ | H | $OCH_3$ |
| **5f** : | H | $OCH_3$ | $OCH_3$ | $OCH_3$ |
| **5g** : | H | $OCH_3$ | H | $NO_2$ |
| **5h** : | $OCH_3$ | $OCH_3$ | H | H |
| **5i** : | $OCH_3$ | $OCH_3$ | H | $OCH_3$ |
| **5j** : | $OCH_3$ | $OCH_3$ | H | $NO_2$ |
| **5k** : | $OCH_3$ | $OCH_3$ | $OCH_3$ | $OCH_3$ |
| **5l** : | H | $NO_2$ | $OCH_3$ | $OCH_3$ |

Reagents and conditions: (a) Na, EtOH, 85 °C, 1h; (b) DIBAL-H, $CH_2Cl_2$, rt, 3h; (c) (i) $Ph_3P{=}CHCO_2Et$, toluene, reflux, 4 h; (ii) KOH, EtOH-$H_2O$, reflux, 3 h. (d) (i) $(COCl)_2$, toluene, 80 °C, 5 h; (ii) $NaN_3$, acetone, rt, 2 h


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**Table 2. Iodine-Catalyzed Thermal Cyclization of 1 To Give Products 2, 3, and 4**

| | | | substituent | | | | | | |
|---|---|---|---|---|---|---|---|---|---|
| entry | reactant[a] | time (h) | $R_3'$ | $R_4'$ | $R_3''$ | $R_4''$ | product (%) | | |
| 1 | **5a** (96/4) | 7 | H | H | H | H | **2a** (32) | **3a** (60) | |
| 2 | **5b** (70/30) | 4 | H | H | H | $OCH_3$ | **2b** (32) | **3b** (50) | |
| 3 | **5c** (82/18) | 6 | H | H | $OCH_3$ | $OCH_3$ | **2c** (8) | **3c** (83) | |
| 4 | **5d** (60/40) | 4 | H | $OCH_3$ | H | H | **2d** (63) | **3d** (22) | |
| 5 | **5e** (80/20) | 4 | H | $OCH_3$ | H | $OCH_3$ | **2e** (58) | **3e** (25) | |
| 6 | **5f** (88/12) | 4 | H | $OCH_3$ | $OCH_3$ | $OCH_3$ | **2f** (11) | **3f** (81) | |
| 7 | **5g** (91/9) | 7 | H | $OCH_3$ | H | $NO_2$ | **2g** (88) | | |
| 8 | **5h** (96/4) | 4 | $OCH_3$ | $OCH_3$ | H | H | **2h** (55) | | **4h** (32) |
| 9 | **5i** (94/6) | 4 | $OCH_3$ | $OCH_3$ | H | $OCH_3$ | **2i** (45) | | **4i** (25) |
| 10 | **5j** (75/25) | 4 | $OCH_3$ | $OCH_3$ | H | $NO_2$ | **2j** (30) | | **4j** (67) |
| 11 | **5k** (89/11) | 1 | $OCH_3$ | $OCH_3$ | $OCH_3$ | $OCH_3$ | **2k** (12) | **3k** (61) | |
| 12 | **5l** (100/0) | 5 | H | $NO_2$ | $OCH_3$ | $OCH_3$ | **2l** (13) | **3l** (77) | |

[a]A mixture of dienoyl azides (**2*E*,4*E***)-**5** and (**2*E*,4*Z***)-**5** was used. The (**2*E*,4*Z***)-**5**/(**2*E*,4*E***)-**5** ratio determined by $^1$H NMR spectrum is shown in parentheses.

**5** were 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**.[15,16] 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.[17] Because the identical cyclized results from (2*E*,4*E*)- and (2*E*,4*Z*)-**5a** were observed (Table 1, entries 4 and 7), the mixtures of the geometric isomers (*E*)- and (*Z*)-**10**, (2*E*,4*E*)- and (2*E*,4*Z*)-**11**, and (2*E*,4*E*)- and (2*E*,4*Z*)-**5** were used directly in the acid, azide formation, and iodine-catalyzed thermal cyclization, respectively.

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′-$OCH_3$ 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═C═O, in conjugation with the butadiene moiety, was activated by the resonance effect of the 4′-$OCH_3$ 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″-$NO_2$ 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″-$OCH_3$ *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 probably due to the steric effect of the 3″-$OCH_3$ 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″-$OCH_3$, 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′-$NO_2$ on the middle phenyl ring and a 3″-$OCH_3$ on the terminal phenyl ring gave naphthalene **3l** in a comparatively high yield (77%). This result suggests that, no matter what kinds of groups are on the middle phenyl ring in the dienoyl azide **5**, a −$OCH_3$ 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-1*H*-indene (**4h**, 32%), instead of 2-(3′,4′-dimethoxyphenyl)naphthalene, and 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 ($\delta$ 6.73) showed NOE correlations with the phenyl H-2″ and -6″ ($\delta$ 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 (see Supporting Information). We supposed that the 3′-$OCH_3$ 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″-$NO_2$) on the terminal phenyl ring as well as a 3′-$OCH_3$ 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″-$OCH_3$ 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′-$OCH_3$ on the middle phenyl ring accompanied by a 3″-$OCH_3$ 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


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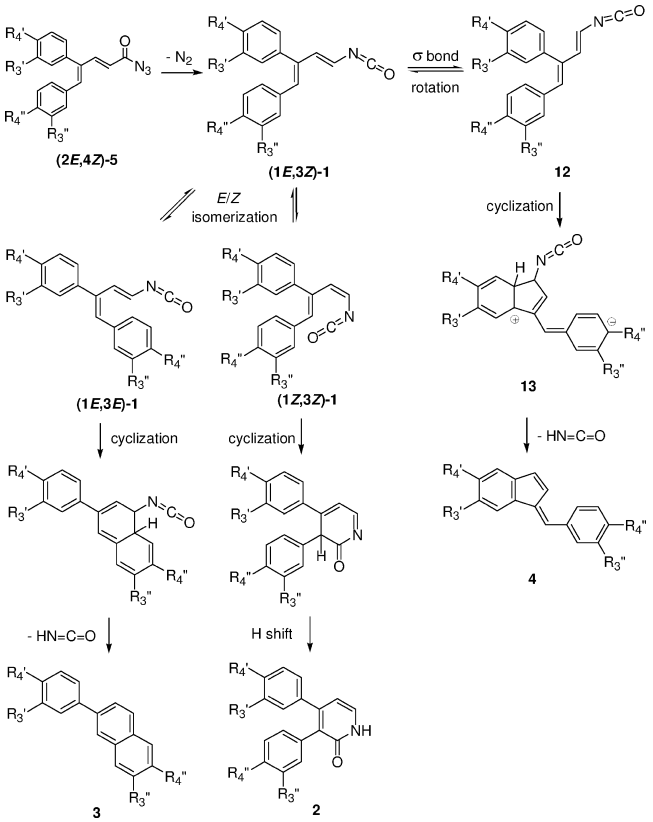
another. The cyclized product distribution depends on the properties and the location of substituents.

## 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.

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




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 been proposed.[18] Subsequently, 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 stereospecific 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 $R_3'$ = $OCH_3$. 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-1*H*-indene (**4j**) can be obtained in higher yields.

## 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 ($OCH_3$) attached to the *meta* position of the terminal phenyl ring, regardless of the other substituents, favors the formation of naphthalenes **3**. A $OCH_3$ on the *para* position of the middle phenyl ring enhances the reaction to yield pyridinones **2**. However, a $OCH_3$ 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 $NO_2$ on the *para* position of the terminal phenyl ring.

## EXPERIMENTAL SECTION

**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 (2*E*,4*E*)- and (2*E*,4*Z*)-**11**. To identify the isomers, the pure acides (2*E*,4*E*)-**11a** and (2*E*,4*Z*)-**11a**−**11l** were obtained from the correspounding aldehydes (*Z*)-**10a** and (*E*)-**10a**−**10l** for spectral analysis. The full spectral data of (2*E*,4*E*)-**11a** and (2*E*,4*Z*)-**11a**−**11l** are described as follows.

**4,5-Diphenylpenta-(2*E*,4*E*)-dienoic Acid [(2*E*,4*E*)-11a].** Yield 70% from aldehyde (*Z*)-**10a**; white granule, mp 185−186 °C (hexane−EtOAc). $^1$H NMR (500 MHz, $CDCl_3$): $\delta$ 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 (125 MHz, $CDCl_3$): $\delta$ 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 $C_{17}H_{14}O_2$: C, 81.58; H, 5.64. Found: C, 81.50; H, 5.57.

**4,5-Diphenylpenta-(2*E*,4*Z*)-dienoic Acid [(2*E*,4*Z*)-11a].** Yield 72% from aldehyde (*E*)-**10a**; white granule, mp 169−170 °C (hexane−EtOAc). $^1$H NMR (400 MHz, $CDCl_3$): $\delta$ 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 (100 MHz, $CDCl_3$): $\delta$ 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 $C_{17}H_{14}O_2$: C, 81.58; H, 5.64. Found: C, 81.52; H, 5.66.

**5-(4″-Methoxyphenyl)-4-phenylpenta-(2*E*,4*Z*)-dienoic Acid [(2*E*,4*Z*)-11b].** Yield 100% from aldehyde (*E*)-**10b**; bright yellow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granule, mp 216–217 °C (hexane–$CHCl_3$). $^1$H NMR (500 MHz, $CDCl_3$): δ 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 (125 MHz, $CDCl_3$): δ 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 $C_{18}H_{16}O_3$: C, 77.12; H, 5.75. Found: C, 77.41; H, 5.84.

**5-(3″,4″-Dimethoxyphenyl)-4-phenylpenta-(2*E*,4*Z*)-dienoic Acid [(2*E*,4*Z*)-11c].** Yield 90% from aldehyde (*E*)-**10c**; bright yellow needle, mp 219–221 °C (EtOAc). $^1$H NMR (400 MHz, $CDCl_3$): δ 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 (100 MHz, $CDCl_3$): δ 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 $C_{19}H_{18}O_4$: C, 73.53; H, 5.85. Found: C, 73.28; H, 6.02.

**4-(4′-Methoxyphenyl)-5-phenylpenta-(2*E*,4*Z*)-dienoic Acid [(2*E*,4*Z*)-11d].** Yield 85% from aldehyde (*E*)-**10d**; white granule, mp 177–178 °C (hexane–$CHCl_3$). $^1$H NMR (500 MHz, $CDCl_3$): δ 3.84 (3H, s), 5.54 (1H, d, *J* = 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 (125 MHz, $CDCl_3$): 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% from aldehyde (*E*)-**10e**; pale yellow granule, mp 189–190 °C (hexane–EtOAc). $^1$H NMR (500 MHz, $CDCl_3$): 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 (125 MHz, $CDCl_3$): 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% from aldehyde (*E*)-**10f**; pale yellow granule, mp 106–107 °C (hexane–$CHCl_3$). $^1$H NMR (400 MHz, $CDCl_3$): 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 (100 MHz, $CDCl_3$): 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, 70.57; 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% from aldehyde (*E*)-**10g**; yellow granule, mp 231–234 °C (hexane–$CHCl_3$). $^1$H NMR (500 MHz, $CDCl_3$): 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 (125 MHz, $CDCl_3$): δ 55.3, 115.0, 121.5, 123.4, 127.4, 130.2, 130.5, 136.3, 142.2, 142.9, 146.8, 150.7, 159.8, 170.6. IR (KBr): 1687 $cm^{-1}$. EIMS *m*/*z* (rel int): 325 (100, $M^+$). HREIMS *m*/*z* calcd for $C_{18}H_{15}NO_5$: 325.0950; found: 325.0956 $[M]^+$.

**4-(3′,4′-Dimethoxyphenyl)-5-phenylpenta-(2*E*,4*Z*)-dienoic Acid [(2*E*,4*Z*)-11h].** Yield 93% from aldehyde (*E*)-**10h**; pale yellow granule, mp 158–159 °C (hexane–EtOAc). $^1$H NMR (400 MHz, $CDCl_3$): 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 (100 MHz, $CDCl_3$): 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-(2*E*,4*Z*)-dienoic Acid [(2*E*,4*Z*)-11i].** Yield 85% from aldehyde (*E*)-**10i**; pale yellow granule, mp 189–190 °C (hexane–EtOAc). $^1$H NMR (500 MHz, $CDCl_3$): 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 (125 MHz, $CDCl_3$): 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-(2*E*,4*Z*)-dienoic Acid [(2*E*,4*Z*)-11j].** Yield 82% from aldehyde (*E*)-**10j**; yellow granule, mp 224–225 °C (hexane–EtOAc). $^1$H NMR (400 MHz, DMSO-$d_6$): δ 3.66 (3H, s), 3.79 (3H, 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 (100 MHz, 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-(2*E*,4*Z*)-dienoic Acid [(2*E*,4*Z*)-11k].** Yield 92% from aldehyde (*E*)-**10k**; pale yellow granule, mp 237–238 °C (hexane–EtOAc). $^1$H NMR (400 MHz, DMSO-$d_6$): δ 3.35 (3H, s), 3.69 (6H, s), 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). $^{13}$C NMR (100 MHz, DMSO-$d_6$): 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. Calcd for $C_{21}H_{22}O_6$: C, 68.10; H, 5.99. Found: C, 68.48; H, 6.12.

**5-(3″,4″-Dimethoxyphenyl)-4-(4′-nitrophenyl)penta-(2*E*,4*Z*)-dienoic Acid [(2*E*,4*Z*)-11l].** Yield 80% from aldehyde (*E*)-**10l**; pale yellow granule, mp 236–237 °C (hexane–$CH_2Cl_2$). $^1$H NMR (400 MHz, $CDCl_3$): 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). $^{13}$C NMR (100 MHz, DMSO-$d_6$): 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. Calcd for $C_{19}H_{17}NO_6$: 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 (2*E*,4*E*)- and (2*E*,4*Z*)-**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 $NaN_3$ (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–$CH_2Cl_2$ to yield a geometric mixture of azides (2*E*,4*E*)- and (2*E*,4*Z*)-**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, the pure azides (2*E*,4*E*)-**5a** and (2*E*,4*Z*)-**5a–5l** were obtained from the correspounding acids (2*E*,4*E*)-**11a** and (2*E*,4*Z*)-**11a–11l** for spectral analysis. The full spectral data of (2*E*,4*E*)-**5a** and (2*E*,4*Z*)-**5a–5l** are described as follows.

**4,5-Diphenylpenta-(2*E*,4*E*)-dienoyl Azide [(2*E*,4*E*)-5a].** Yield 100% from acid (2*E*,4*E*)-**11a**; yellow syrup. $^1$H NMR (500 MHz, $CDCl_3$): δ 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 (125 MHz, $CDCl_3$): δ 124.1, 128.1, 128.5 (2 × 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 $[M - N_2]^+$.


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**4,5-Diphenylpenta-(2*E*,4*Z*)-dienoyl Azide [(2*E*,4*Z*)-5a].** Yield 100% from acid (2*E*,4*Z*)-**11a**; yellow solid. $^1$H NMR (500 MHz, $CDCl_3$): $\delta$ 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 (125 MHz, $CDCl_3$): $\delta$ 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 $C_{17}H_{13}N_3O$: 275.1059; found: 275.1057 [M]$^+$.

**5-(4″-Methoxyphenyl)-4-phenylpenta-(2*E*,4*Z*)-dienoyl Azide [(2*E*,4*Z*)-5b].** Yield 100% from acid (2*E*,4*Z*)-**11b**; yellow solid. $^1$H NMR (500 MHz, $CDCl_3$): $\delta$ 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 (125 MHz, $CDCl_3$): $\delta$ 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 $C_{18}H_{15}N_3O_2$: 305.1164; found: 305.1157 [M]$^+$.

**5-(3″,4″-Dimethoxyphenyl)-4-phenylpenta-(2*E*,4*Z*)-dienoyl Azide [(2*E*,4*Z*)-5c].** Yield 96% from acid (2*E*,4*Z*)-**11c**; yellow solid. $^1$H NMR (400 MHz, $CDCl_3$): $\delta$ 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 (100 MHz, $CDCl_3$): $\delta$ 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 − $N_2$]$^+$.

**4-(4′-Methoxyphenyl)-5-phenylpenta-(2*E*,4*Z*)-dienoyl Azide [(2*E*,4*Z*)-5d].** Yield 100% from acid (29E,4*Z*)-**11d**; yellow solid. $^1$H NMR (500 MHz, $CDCl_3$): $\delta$ 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). $^{13}$C NMR (125 MHz, $CDCl_3$): $\delta$ 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 $C_{18}H_{15}N_3O_2$: 305.1164; found: 305.1160 [M]$^+$.

**4,5-Bis(4′-methoxyphenyl)penta-(2*E*,4*Z*)-dienoyl Azide [(2*E*,4*Z*)-5e].** Yield 91% from acid (2*E*,4*Z*)-**11e**; yellow solid. $^1$H NMR (500 MHz, $CDCl_3$): $\delta$ 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). $^{13}$C NMR (125 MHz, $CDCl_3$): $\delta$ 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 $C_{19}H_{17}N_3O_3$: 335.1270; found: 335.1265 [M]$^+$.

**5-(3″,4″-Dimethoxyphenyl)-4-(4′-methoxyphenyl)penta-(2*E*,4*Z*)-dienoyl Azide [(2*E*,4*Z*)-5f].** Yield 83% from acid (2*E*,4*Z*)-**11f**; yellow solid. $^1$H NMR (500 MHz, $CDCl_3$): $\delta$ 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). $^{13}$C NMR (125 MHz, $CDCl_3$): $\delta$ 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, 1682 cm$^{-1}$. EIMS $m/z$ (rel int): 337 (100, [M − $N_2$]$^+$). HREIMS $m/z$ calcd for $C_{20}H_{19}N_3O_4$: 337.1314; found: 337.1321 [M − $N_2$]$^+$.

**4-(4′-Methoxyphenyl)-5-(4″-nitrophenyl)penta-(2*E*,4*Z*)-dienoyl Azide [(2*E*,4*Z*)-5g].** Yield 80% from acid (2*E*,4*Z*)-**11g**; yellow solid. $^1$H NMR (500 MHz, $CDCl_3$): $\delta$ 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). $^{13}$C NMR (125 MHz, $CDCl_3$): $\delta$ 55.3, 115.0, 123.4, 123.5, 127.1, 130.1, 130.5, 137.3, 142.0, 142.8, 146.8, 150.4, 159.8, 172.0. IR (KBr): 2137, 1683 cm$^{-1}$. EIMS $m/z$ (rel int) 322 (100, [M − $N_2$]$^+$). HREIMS $m/z$ calcd for $C_{18}H_{14}N_2O_4$: 322.0954; found: 322.0959 [M − $N_2$]$^+$.

**4-(3′,4′-Dimethoxyphenyl)-5-phenylpenta-(2*E*,4*Z*)-dienoyl Azide [(2*E*,4*Z*)-5h].** Yield 93% from acid (2*E*,4*Z*)-**11h**; yellow solid. $^1$H NMR (400 MHz, $CDCl_3$): $\delta$ 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 (100 MHz, $CDCl_3$): $\delta$ 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-(2*E*,4*Z*)-dienoyl Azide [(2*E*,4*Z*)-5i].** Yield 95% from acid (2*E*,4*Z*)-**11i**; yellow solid. $^1$H NMR (400 MHz, $CDCl_3$): $\delta$ 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 (100 MHz, $CDCl_3$): $\delta$ 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 $C_{20}H_{19}NO_4$: 337.1314; found: 337.1323 [M − $N_2$]$^+$.

**4-(3′,4′-Dimethoxyphenyl)-5-(4″-nitrophenyl)penta-(2*E*,4*Z*)-dienoyl Azide [(2*E*,4*Z*)-5j].** Yield 95% from acid (2*E*,4*Z*)-**11j**; yellow solid. $^1$H NMR (400 MHz, $CDCl_3$): $\delta$ 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 (100 MHz, $CDCl_3$): $\delta$ 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 $C_{19}H_{16}N_2O_5$: 352.1059; found: 352.1051 [M − $N_2$]$^+$.

**4,5-Bis(3′,4′-dimethoxyphenyl)penta-(2*E*,4*Z*)-dienoyl Azide [(2*E*,4*Z*)-5k].** Yield 92% from acid (2*E*,4*Z*)-**11k**; yellow solid. $^1$H NMR (400 MHz, $CDCl_3$): $\delta$ 3.47 (3H, s), 3.81 (3H, s), 3.83 (3H, s), 3.91 (3H, s), 5.49 (1H, d, $J$ = 15.1 Hz), 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 (100 MHz, $CDCl_3$): $\delta$ 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 $C_{21}H_{21}N_3O_5$: 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 (400 MHz, $CDCl_3$): $\delta$ 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 (100 MHz, $CDCl_3$): $\delta$ 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$^{\pm}$). HREIMS $m/z$ calcd for $C_{19}H_{16}N_4O_5$: 380.1121; found: 380.1128 [M]$^{\pm}$.

**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 iodine (5.1 mg, 0.02 mmol) in *o*-dichlorobenzene (5 mL) was heated at 150 °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−$CH_2Cl_2$ to give 3,4-diphenylpyridin-2(1*H*)-one **2** and 2-phenylnaphthalene **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.[19] mp 284−284.5 °C). $^1$H NMR (500 MHz, 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 (125 MHz, 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 $m/z$ (rel int) 247 (58, M$^+$). Anal. Calcd for $C_{17}H_{13}NO$: C, 82.57; H, 5.30; N, 5.66. Found: C, 82.88; H, 5.65; N, 5.60.

**2-Phenylnaphthalene (3a).** Yield 60%; pale yellow granule, mp 100−101 °C (hexane−$CHCl_3$) (lit.[11] mp 100−101 °C). $^1$H NMR (400 MHz, $CDCl_3$): $\delta$ 7.37 (1H, t, $J$ = 7.3 Hz), 7.48 (4H, m), 7.73 (3H, m), 7.88 (3H, m), 8.04 (1H, s). $^{13}$C NMR (100 MHz, $CDCl_3$): $\delta$ 125.6, 125.8, 125.9, 126.2, 127.3, 127.4, 127.6, 128.2, 128.4, 128.8,


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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 $C_{16}H_{12}$: C, 94.08; H, 5.92. Found: C, 94.09; H, 6.25.

**3-(4″-Methoxyphenyl)-4-phenylpyridin-2(1*H*)-one (2b).** Yield 32%; white granules, mp 249−251 °C (hexane−$CHCl_3$). $^1H$ NMR (600 MHz, $CDCl_3$): 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). $^{13}C$ NMR (150 MHz, $CDCl_3$): 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 $C_{18}H_{15}NO_2$: 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−$CHCl_3$) (lit.[20] mp 150−151 °C). $^1H$ NMR (500 MHz, $CDCl_3$): 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). $^{13}C$ NMR (125 MHz, $CDCl_3$): 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 $C_{17}H_{14}O$: C, 87.15; H, 6.02. Found: C, 86.84; H, 6.26.

**3-(3′,4′-Dimethoxyphenyl)-4-phenylpyridin-2(1*H*)-one (2c).** Yield 8%; white granules, mp 228−230 °C (hexane−$CHCl_3$). $^1H$ NMR (500 MHz, $CDCl_3$): 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). $^{13}C$ NMR (125 MHz, $CDCl_3$): δ 55.6, 55.7, 109.6, 110.7, 114.6, 123.7, 127.5, 127.7, 128.1, 128.8, 129.4, 132.6, 139.5, 148.1, 148.2, 151.8, 164.6. IR (KBr): 1641 $cm^{-1}$. EIMS *m/z* (rel int) 307 (100, $M^+$). HREIMS *m/z* calcd for $C_{19}H_{17}NO_3$: 307.1208; found: 307.1197 $[M]^+$.

**2,3-Dimethoxy-6-phenylnaphthalene (3c).** Yield 83%; white needles, mp 136−137 °C (hexane−EtOAc) (lit.[21] mp 134−135 °C). $^1H$ NMR (400 MHz, $CDCl_3$): δ 3.98 (6H, s), 7.11 (1H, s), 7.14 (1H, s), 7.34 (1H, t, *J* = 7.5 Hz), 7.45 (2H, t, *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 (100 MHz, $CDCl_3$): δ 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, $M^+$). Anal. Calcd for $C_{18}H_{16}O_2$: C, 81.79; H, 6.10. Found: C, 81.59; H, 6.15.

**4-(4′-Methoxyphenyl)-3-phenylpyridin-2(1*H*)-one (2d).** Yield 63%; white granules, mp 245−246 °C (hexane−$CHCl_3$). $^1H$ NMR (400 MHz, DMSO-$d_6$): δ 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 (100 MHz, DMSO-$d_6$): δ 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.[22] mp 139−140 °C). $^1H$ NMR (500 MHz, $CDCl_3$): δ 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 (125 MHz, $CDCl_3$): δ 55.4, 114.3, 125.0, 125.4, 125.6, 126.2, 127.6, 128.0, 128.3, 128.4, 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−$CHCl_3$). $^1H$ NMR (600 MHz, DMSO-$d_6$): δ 3.69 (3H, s), 3.70 (3H, s), 6.19 (1H, 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 (150 MHz, DMSO-$d_6$): δ 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 $C_{19}H_{17}NO_3$: 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−$CHCl_3$) (lit.[23] mp 190 °C). $^1H$ NMR (400 MHz, $CDCl_3$): 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). $^{13}C$ NMR (100 MHz, $CDCl_3$): 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 $C_{18}H_{16}O_2$: C, 81.79; H, 6.10. Found: C, 81.42; H, 5.94.

**3-(3″,4″-Dimethoxyphenyl)-4-(4′-methoxyphenyl)pyridin-2(1*H*)-one (2f).** Yield 11%; white granules, mp 284−285 °C (hexane−$CHCl_3$). $^1H$ NMR (400 MHz, DMSO-$d_6$): 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); $^{13}C$ NMR (100 MHz, DMSO-$d_6$): δ 55.1, 55.5 (2 × C), 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 $C_{20}H_{19}NO_4$: 337.1314; found: 337.1304 $[M]^+$.

**2,3-Dimethoxy-6-(4′-methoxyphenyl)naphthalene (3f).** Yield 81%; white needles, mp 164−165 °C (hexane−$CHCl_3$) (lit.[24] mp 158−159 °C). $^1H$ NMR (400 MHz, $CDCl_3$): 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). $^{13}C$ NMR (100 MHz, $CDCl_3$): δ 55.3, 55.8 (2 × C), 106.0, 106.4, 114.2, 123.6 (2 × C), 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 $C_{19}H_{18}O_3$: C, 77.53; H, 6.16. Found: C, 77.52; H, 6.42.

**4-(4′-Methoxyphenyl)-3-(4″-nitrophenyl)pyridin-2(1*H*)-one (2g).** Yield 88%; pale yellow granules, mp 272−274 °C (hexane−$CHCl_3$). $^1H$ NMR (500 MHz, $CDCl_3$): 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). $^{13}C$ NMR (125 MHz, $CDCl_3$): 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 $C_{18}H_{14}N_2O_4$: 322.0954; found: 322.0958 $[M]^+$.

**4-(3′,4′-Dimethoxyphenyl)-3-phenylpyridin-2(1*H*)-one (2h).** Yield 55%; white granules, mp 250−252 °C (hexane−$CHCl_3$). $^1H$ NMR (400 MHz, DMSO-$d_6$): δ 3.36 (3H, s), 3.69 (3H, s), 6.28 (1H, d, *J* = 6.8 Hz), 6.47 (1H, d, *J* = 1.9 Hz), 6.74 (1H, dd, *J* = 8.3, 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). $^{13}C$ NMR (100 MHz, DMSO-$d_6$): 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 $C_{19}H_{17}NO_3$: 307.1208; found: 307.1202 $[M]^+$.

**1(*E*)-Benzylidene-5,6-dimethoxy-1*H*-indene (4h).** Yield 32%; yellow granules, mp 104−105 °C (hexane−$CHCl_3$). $^1H$ NMR (400 MHz, $CDCl_3$): 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). $^{13}C$ NMR (100 MHz, $CDCl_3$): 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 $C_{18}H_{16}O_2$: C, 81.79; H, 6.10. Found: C, 81.49; H, 6.17.

**4-(3′,4′-Dimethoxyphenyl)-3-(4″-methoxyphenyl)pyridin-2(1*H*)-one (2i).** Yield 45%; white granules, mp 286−288 °C (hexane−$CHCl_3$). $^1H$ NMR (500 MHz, DMSO-$d_6$): 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). $^{13}C$ NMR (125 MHz, DMSO-$d_6$) 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 $C_{20}H_{19}NO_4$: 337.1314; found: 337.1307 $[M]^+$.

**1(*E*)-(4″-Methoxybenzylidene)-5,6-dimethoxy-1*H*-indene (4i).** Yield 25%; yellow granules, mp 134−135 °C (hexane−$CHCl_3$). $^1H$ NMR (500 MHz, $CDCl_3$): 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). $^{13}$C NMR (125 MHz, $CDCl_3$): $\delta$ 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 $C_{19}H_{18}O_3$: C, 77.53; H, 6.16. Found: C, 77.68; H, 5.97.

**4-(3′,4′-Dimethoxyphenyl)-3-(4″-nitrophenyl)pyridin-2(1*H*)-one (2j).** Yield 30%; pale yellow granules, mp 256−257 °C (hexane−$CHCl_3$). $^1$H NMR (400 MHz, $CDCl_3$): $\delta$ 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); $^{13}$C NMR (100 MHz, $CDCl_3$) $\delta$ 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 $C_{19}H_{16}N_2O_5$: 352.1059; found: 352.1061 [M]$^+$.

**1(*E*)-(4″-Nitrobenzylidene)-5,6-dimethoxy-1*H*-indene (4j).** Yield 67%; black granules, mp 130−131 °C (hexane−$CHCl_3$). $^1$H NMR (400 MHz, $CDCl_3$): $\delta$ 3.91 (3H, s), 3.96 (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); $^{13}$C NMR (100 MHz, $CDCl_3$) $\delta$ 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 $C_{18}H_{15}NO_4$: 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(1*H*)-one (2k).** Yield 12%; white granules, mp >300 °C (hexane−$CHCl_3$). $^1$H NMR (400 MHz, $CDCl_3$): $\delta$ 3.57 (3H, s), 3.69 (3H, s), 3.85 (3H, s), 3.86 (3H, s), 6.40 (1H, d, $J$ = 6.7 Hz), 6.54 (1H, d, $J$ = 1.4 Hz), 6.71 (1H, d, $J$ = 1.1 Hz), 6.79 (4H, m), 7.39 (1H, d, $J$ = 6.7 Hz), 13.19 (1H, br s); $^{13}$C NMR (100 MHz, $CDCl_3$): $\delta$ 55.6, 55.7, 55.8 (2 × C), 109.5, 110.5, 110.8, 112.4, 114.2, 121.4, 123.5, 128.0, 128.8, 131.7, 132.8, 148.0, 148.1, 148.4, 148.6, 151.4, 164.8. IR (KBr): 1632 cm$^{-1}$. EIMS $m/z$ (rel int) 367 (100, M$^+$). HREIMS $m/z$ calcd for $C_{21}H_{21}NO_5$: 367.1420; found: 367.1426 [M]$^+$.

**2,3-Dimethoxy-6-(3′,4′-dimethoxyphenyl)naphthalene (3k).** Yield 61%; yellow granules, mp 165−166 °C (hexane−$CHCl_3$) (lit.[25] mp 168−170 °C). $^1$H NMR (400 MHz, $CDCl_3$): $\delta$ 3.94 (3H, s), 3.99 (3H, s), 4.02 (3H, s), 4.03 (3H, s), 6.98 (1H, d, $J$ = 8.2 Hz), 7.14 (1H, s), 7.18 (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). $^{13}$C NMR (100 MHz, $CDCl_3$): $\delta$ 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 $C_{20}H_{20}O_4$: C, 74.06; H, 6.21. Found: C, 74.39; H, 6.62.

**3-(3″,4″-Dimethoxyphenyl)-4-(4′-nitrophenyl)pyridin-2(1*H*)-one (2l).** Yield 13%; yellow needles, mp 243−245 °C (hexane-EtOAc). $^1$H NMR (500 MHz, $CDCl_3$) $\delta$ 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), 7.29 (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). $^{13}$C NMR (125 MHz, $CDCl_3$) $\delta$ 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 $C_{19}H_{16}N_2O_5$: 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). $^1$H NMR (400 MHz, $CDCl_3$) $\delta$ 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). $^{13}$C NMR (100 MHz, $CDCl_3$): $\delta$ 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 $C_{18}H_{15}NO_4$: 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-1*H*-pyridin-2-one (6a).** Yellow granules, mp 233−235 °C (hexane−EtOAc). $^1$H NMR (500 MHz, $CDCl_3$): $\delta$ 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). $^{13}$C NMR (125 MHz, $CDCl_3$): $\delta$ 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, 136.8, 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 $C_{34}H_{26}N_2O_2$: C, 82.57; H, 5.30; N, 5.66. Found: C, 82.35; H, 5.62; N, 5.60.

## ■ ASSOCIATED CONTENT

### Ⓢ Supporting Information

$^1$H and $^{13}$C NMR spectra of all compounds **2**−**6** and **9**−**11**, preparation of starting materials **9** and **10**, and X-ray crystallographic data as well as ORTEP structure, including CIF file for **4j**. This material is available free of charge via the Internet at http://pubs.acs.org.

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