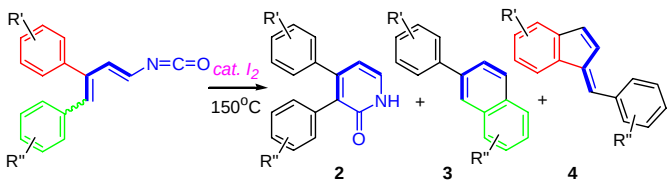
Substituent Effects on the Iodine-Catalyzed Thermal Cyclization of 3,4-Diphenylbuta-1,3-dienyl Isocyanates: Mechanistic Studies

Ta-Hsien Chuang, Wei-Yu Chang, Chien-Fu Li, Yu-Chia Wen and Chia-Chen Tsai*

School of Pharmacy, China Medical University, No. 91, Hsueh-Shih Road, Taichung, 40402, TAIWAN, Republic of China

thchuang@mail.cmu.edu.tw

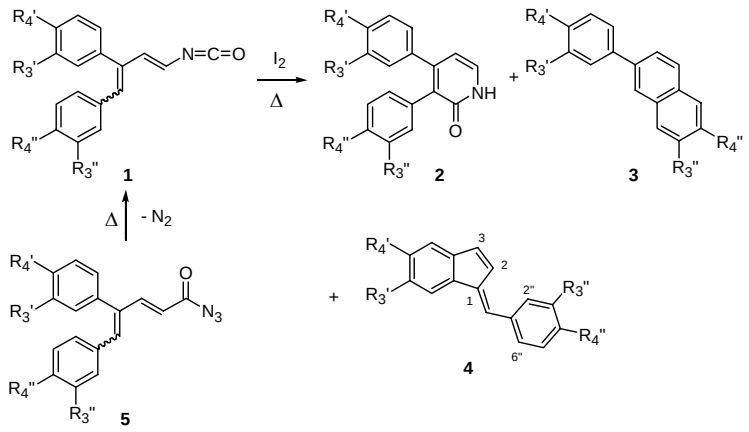


The thermal cyclization of 3,4-diphenylbuta-1,3-dienyl isocyanates **1**, generated *in situ* from the corresponding azides, was investigated using iodine as a catalyst. Diphenylpyridinones **2**, phenylnaphthalenes **3** and indenes **4** were produced via intramolecular ring closure. The nature of 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.[1-7] Eloy and Washburne extensively applied the thermal cyclization of penta-2,4-dienoyl isocyanates to synthesize 1*H*-pyridin-2-ones.[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.[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.[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(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.

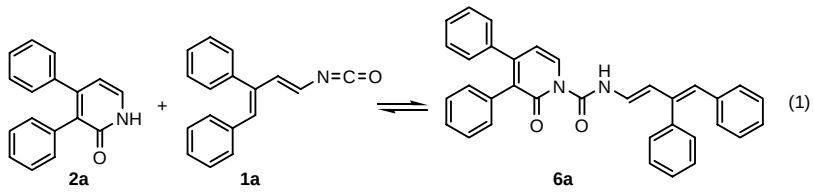
Scheme 1. Iodine-Catalyzed Cyclization of 3,4-Diphenylpenta-1,3-dienoyl Isocyanates 1



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.[10] 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.[8] 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 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.



2a **1a** **6a** (1)

Apparently, the unexpected product **3a** was produced via the intramolecular cyclization of the triene[11], 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 (1*E*,3*Z*)-**1a** and (1*E*,3*E*)-**1a**, were obtained quantitatively from (2*E*,4*Z*)-**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(1*H*)-one **2a** (66%) and butadienylbenzene into naphthalene **3a** (25%, entry 3). Due to 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 **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 (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**.

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

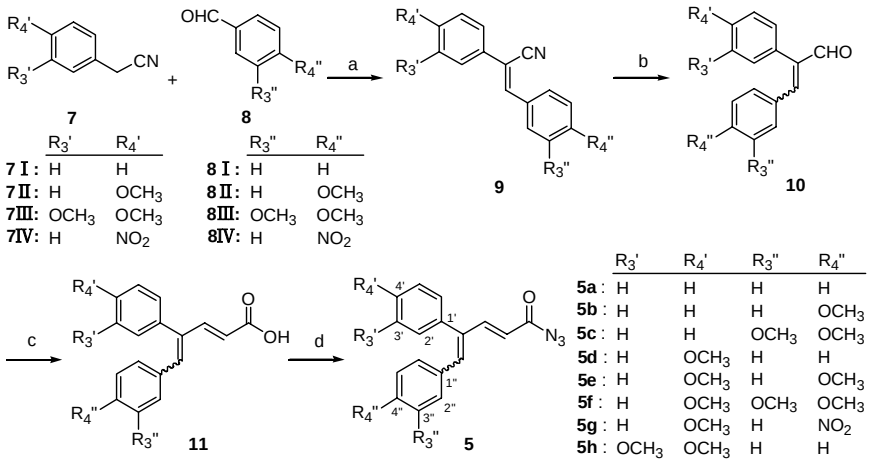
entry	solvent	temp (°C)	time (h)	product (%)	
				2a	**3a**
1[a]	diphenyl ether	240	3	11	9
2[a]	*o*-dichlorobenzene	reflux	5	-	-
3	*o*-dichlorobenzene	reflux	5	66	25
4	*o*-dichlorobenzene	150	7	32	60
5	*o*-dichlorobenzene	130	15	45	40
6	*o*-dichlorobenzene	110	96	40	20
7[b]	*o*-dichlorobenzene	150	7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

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

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**.[14,15] 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.[16] 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** as well as (2*E*,4*E*)- and (2*E*,4*Z*)-**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



	R_3'	R_4'
7Ⅰ:	H	H
7Ⅱ:	H	OCH_3
7Ⅲ:	OCH_3	OCH_3
7Ⅳ:	H	NO_2

	R_3''	R_4''
8Ⅰ:	H	H
8Ⅱ:	H	OCH_3
8Ⅲ:	OCH_3	OCH_3
8Ⅳ:	H	NO_2

	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 ^oC, 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 ^oC, 5 h; (ii) NaN_3, 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'-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 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 kind 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, 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'-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 with 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 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

entry	reactant*	time (h)	substituent				product (%)		
			R_3'	R_4'	R_3"	R_4"			
1	**5a**	7	H	H	H	H	**2a** (32)	**3a** (60)	
2	**5b**	4	H	H	H	OCH_3	**2b** (32)	**3b** (50)	
3	**5c**	6	H	H	OCH_3	OCH_3	**2c** (8)	**3c** (83)	
4	**5d**	4	H	OCH_3	H	H	**2d** (63)	**3d** (22)	
5	**5e**	4	H	OCH_3	H	OCH_3	**2e** (58)	**3e** (25)	
6	**5f**	4	H	OCH_3	OCH_3	OCH_3	**2f** (11)	**3f** (81)	
7	**5g**	7	H	OCH_3	H	NO_2	**2g** (88)		
8	**5h**	4	OCH_3	OCH_3	H	H	**2h** (55)		**4h** (32)
9	**5i**	4	OCH_3	OCH_3	H	OCH_3	**2i** (45)		**4i** (25)
10	**5j**	4	OCH_3	OCH_3	H	NO_2	**2j** (30)		**4j** (67)
11	**5k**	1	OCH_3	OCH_3	OCH_3	OCH_3	**2k** (12)	**3k** (61)	
12	**5l**	5	H	NO_2	OCH_3	OCH_3	**2l** (13)	**3l** (77)	

* A mixture of dienoyl azides **(2*E*,4*E*)-5** and **(2*E*,4*Z*)-5** was used.

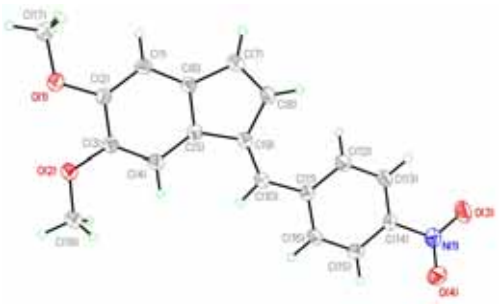


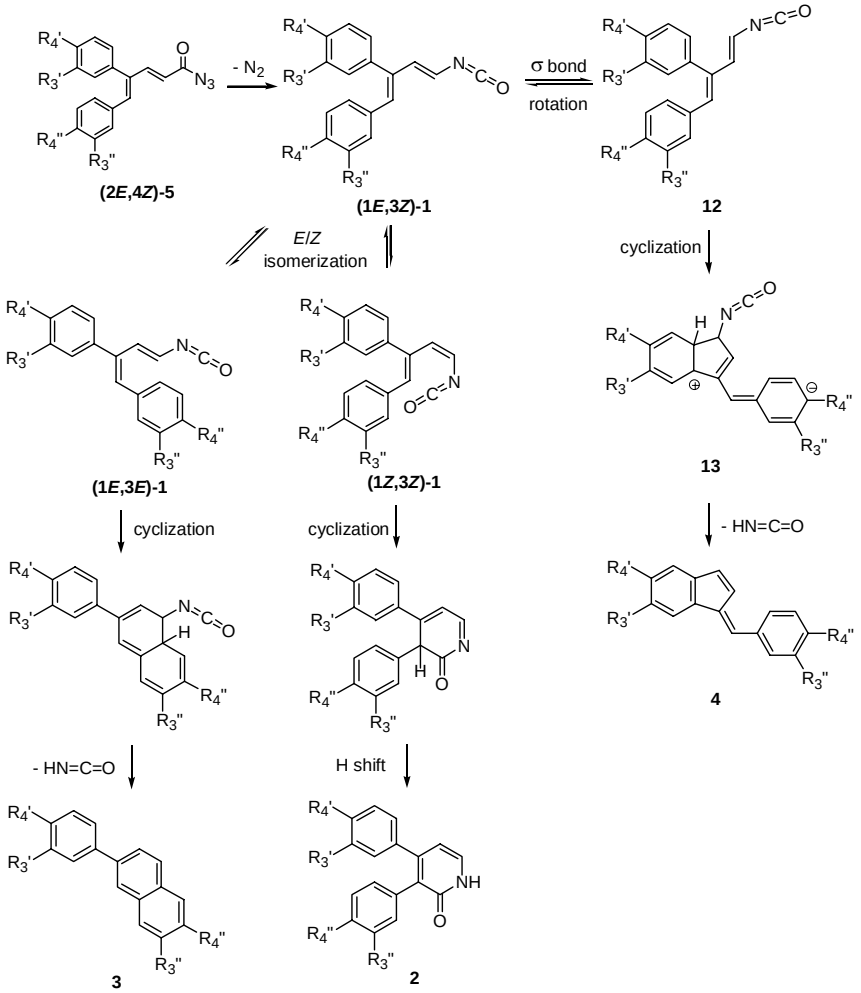
Figure 1. ORTEP (Oak Ridge Thermal-Ellipsoid Plot Program) plot of molecule of **4j** drawn with the thermal ellipsoids at the 20% probability level, small circles represent hydrogen atoms

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.[17] 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 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 $R_3' = OCH_3$. 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.

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

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 N_2 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.,[18] 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.,[19] 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.,[20] mp 88°C). ^{1}H NMR (400MHz, $CDCl_3$) δ 3.94 (3H, s), 3.97 (3H, s), 6.92 (1H, d, J = 8.3 Hz), 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.,[19] 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.,[21] mp 113°C). ^{1}H NMR (400MHz, $CDCl_3$) δ 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 = 8.8Hz) ; ^{13}C NMR (100MHz, $CDCl_3$) δ 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^{-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, 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-$CHCl_3$) (lit.,[22] mp 105°C). ^{1}H NMR (400MHz, $CDCl_3$) δ 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); ^{13}C NMR (100MHz, $CDCl_3$) δ 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^{-1}; EIMS *m/z* (rel int) 295 (100, M^+). Anal. Calcd for $C_{18}H_{17}NO_3$: 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). ^{1}H NMR (400MHz, $CDCl_3$) δ 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); ^{13}C NMR (100MHz, $CDCl_3$) δ 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^{-1}; EIMS *m/z* (rel int) 280 (100, M^+). Anal. Calcd for $C_{16}H_{12}N_2O_3$: 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-$CHCl_3$) (lit.,[23] mp 85−86°C). ^{1}H NMR (400MHz, $CDCl_3$) δ 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); ^{13}C NMR (100MHz, $CDCl_3$) δ 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^+). Anal. Calcd for $C_{17}H_{15}NO_2$: 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.,[24] mp 128–130°C). ^{1}H NMR (400MHz, $CDCl_3$) 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); ^{13}C NMR (100MHz, $CDCl_3$) 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. Calcd for $C_{18}H_{17}NO_3$: 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-$CHCl_3$). ^{1}H NMR (400MHz, $CDCl_3$) 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); ^{13}C NMR (100MHz, $CDCl_3$) δ 56.0, 56.1, 108.8, 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. Calcd for $C_{17}H_{14}N_2O_4$: 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.,[14] mp 154−155°C). ^{1}H NMR (400MHz, $CDCl_3$) 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); ^{13}C NMR (100MHz, $CDCl_3$) 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^+). Anal. Calcd for $C_{19}H_{19}NO_4$: C, 70.14; H, 5.85; N, 4.31. Found: C, 70.24; H, 5.76; N, 4.55.

(*Z*)-3-(3'',4''-Dimethoxyphenyl)-2-(4'-nitrophenyl)acrylonitrile (9l). Yield 91%; yellow granule, mp 152–153°C (hexane-EtOAc). ^{1}H NMR (400MHz, $CDCl_3$) 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, $CDCl_3$) δ 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 *m/z* (rel int) 310 (100, M^+). Anal. Calcd for $C_{17}H_{14}N_2O_4$: 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_2Cl_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°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_2O layer was extracted with CH_2Cl_2 (2 × 100 mL), and the combined extracts were washed with H_2O (3 × 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°C (hexane-EtOAc) (lit.,[25] mp 119.5−120.5°C). ^{1}H NMR (400MHz, $CDCl_3$) δ 7.38 (10H, m), 7.84 (1H, s), 10.10 (1H, s); ^{13}C NMR (100MHz, $CDCl_3$) δ 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 *m/z* (rel int) 208 (34, M^+). Anal. Calcd 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°C (hexane-EtOAc) (lit.,[26] mp 93°C). ^{1}H NMR (400MHz, $CDCl_3$) δ 7.18 (6H, m), 7.23 (1H, m), 7.35 (4H, m), 9.72 (1H, s); ^{13}C NMR (100MHz, $CDCl_3$) δ 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"-Methoxyphenyl)-2-phenylacrylaldehyde [(*Z*)-10b]. Yield 32%; white granule, mp 108–109°C (hexane-EtOAc). ^1H NMR (400MHz, $CDCl_3$) δ 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, $CDCl_3$) δ 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"-Methoxyphenyl)-2-phenylacrylaldehyde [(*E*)-10b]. Yield 41%; white granule, mp 123–124°C (hexane-EtOAc). ^1H NMR (400MHz, $CDCl_3$) δ 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, $CDCl_3$) δ 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",4"-Dimethoxyphenyl)-2-phenylacrylaldehyde [(*Z*)-10c]. Yield 22%; pale yellow granule, mp 109–110°C (hexane-$CHCl_3$). ^1H NMR (500MHz, $CDCl_3$) δ 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, $CDCl_3$) δ 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",4"-Dimethoxyphenyl)-2-phenylacrylaldehyde [(*E*)-10c]. Yield 40%; pale yellow granule, mp 123–124°C (hexane-$CHCl_3$). ^1H NMR (400MHz, $CDCl_3$) δ 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, $CDCl_3$) δ 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^+). Anal. Calcd 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.,[27] mp 114–115°C). ^1H NMR (500MHz, $CDCl_3$) δ 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, $CDCl_3$) δ 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. Calcd 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). ^1H NMR (400MHz, $CDCl_3$) δ 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, $CDCl_3$) δ 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. Calcd 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). ^1H NMR (500MHz, $CDCl_3$) δ 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, $CDCl_3$) δ 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. Calcd 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). ^1H NMR (500MHz, $CDCl_3$) δ 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, $CDCl_3$) δ 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. Calcd 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, $CDCl_3$) δ 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, $CDCl_3$) δ 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, $CDCl_3$) δ 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 = 8.7Hz), 7.29 (1H, s), 9.71 (1H, s); ^{13}C NMR (100MHz, $CDCl_3$) δ 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, 6.25.

(*Z*)-2-(4'-Methoxyphenyl)-3-(4"-nitrophenyl)acrylaldehyde [(*Z*)-10g]. Yield 7%; yellow needle, mp 149–150°C (hexane-EtOAc). ^{1}H NMR (500MHz, $CDCl_3$) δ 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 = 8.3Hz), 10.07 (1H, s); ^{13}C NMR (125MHz, $CDCl_3$) δ 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, $CDCl_3$) δ 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, $CDCl_3$) δ 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, 128.8, 129.4, 130.2, 134.0, 140.6, 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 (6H, 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 $C_{18}H_{18}O_4$: 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). ^{1}H NMR (400MHz, $CDCl_3$) 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); ^{13}C NMR (100MHz, $CDCl_3$) 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 $C_{17}H_{15}NO_5$: 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). ^{1}H NMR (400MHz, $CDCl_3$) 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); ^{13}C NMR (100MHz, $CDCl_3$) 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 $C_{17}H_{15}NO_5$: 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). ^{1}H NMR (500MHz, $CDCl_3$) 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); ^{13}C NMR (125MHz, $CDCl_3$) 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, 150.6, 192.5; IR (KBr) 2837, 1682 cm^{-1}; EIMS *m/z* (rel int) 328 (100, M^+); HREIMS *m/z* calcd for $C_{19}H_{20}O_5$: 328.1311, found: 328.1300 [M^+].

(*E*)-2,3-Bis(3',4'-dimethoxyphenyl)acrylaldehyde [(*E*)-10k]. Yield 87%; yellow syrup. ^{1}H NMR (400MHz, $CDCl_3$) 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)*-10l]. 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-(2*E*,4*E*)-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, $CDCl_3$) δ 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 [(*2E,4Z)*-11a]. Yield 72%; white granule, mp 169–170°C (hexane-EtOAc). ^{1}H NMR (400MHz, $CDCl_3$) δ 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, $CDCl_3$) δ 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 [(*2E,4Z*)-11b]. Yield 100%; bright yellow granule, mp 216–217°C (hexane-$CHCl_3$). ^{1}H NMR (500MHz, $CDCl_3$) 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 (125MHz, $CDCl_3$) 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%; bright yellow needle, mp 219–221°C (EtOAc). ^{1}H NMR (400MHz, $CDCl_3$) 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 (100MHz, $CDCl_3$) 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 [(*2E,4Z*)-11d]. Yield 85%; white granule, mp 177–178°C (hexane-$CHCl_3$). ^{1}H NMR (500MHz, $CDCl_3$) 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 (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 [(*2E,4Z*)-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 [(*2E,4Z*)-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, 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%; 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,

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%; pale yellow granule, mp 158–159°C (hexane-EtOAc). ^1H 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-(2*E*,4*Z*)-dienoic acid [(*2E,4Z*)-11i]. Yield 85%; pale yellow granule, mp 189–190°C (hexane-EtOAc). ^1H 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-(2*E*,4*Z*)-dienoic acid [(2*E*,4*Z*)-11j]. Yield 82%; yellow granule, mp 224–225°C (hexane-EtOAc). ^1H NMR (400MHz, 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 (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-(2*E*,4*Z*)-dienoic acid [(2*E*,4*Z*)-11k]. Yield 92%; pale yellow granule, mp 237–238°C (hexane-EtOAc). ^1H NMR (400MHz, 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 (100MHz, 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%; pale yellow granule, mp 236–237°C (hexane-CH_2Cl_2). ^{1}H NMR (400MHz, $CDCl_3$) 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 (100MHz, 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 (*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 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 (*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*E*,4*E*)-dienoyl azide [(*2E,4E)*-5a]. Yield 100% from acid **(***2E,4E*)-**11a**; yellow sygrup. ^{1}H NMR (500MHz, $CDCl_3$) 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 (125MHz, $CDCl_3$) 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, $[M\text{-}N_2]^+$); HREIMS *m/z* calcd for $C_{17}H_{13}NO$: 247.0997, found: 247.0991 $[M\text{-}N_2]^+$.

4,5-Diphenylpenta-(2*E*,4*Z*)-dienoyl azide [(*2E,4Z)*-5a]. Yield 100% from acid (*2E,4Z*)-**11a**; yellow solid. ^{1}H NMR (500MHz, $CDCl_3$) δ 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, $CDCl_3$) δ 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: 275.1059 $[M]^+$.

5-(4"-Methoxyphenyl)-4-phenylpenta-(2*E*,4*Z*)-dienoyl azide [(*2E,4Z*)-5b]. Yield 100% from acid (*2E,4Z*)-**11b**; yellow solid. ^{1}H NMR (500MHz, $CDCl_3$) δ 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, $CDCl_3$) δ 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 (*2E,4Z*)-**11c**; yellow solid. ^{1}H NMR (400MHz, $CDCl_3$) δ 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, $CDCl_3$) δ 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, $[M\text{-}N_2]^+$); HREIMS *m/z* calcd for $C_{19}H_{17}NO_3$: 307.1208, found: 307.1219 $[M\text{-}N_2]^+$.

4-(4'-Methoxyphenyl)-5-phenylpenta-(2*E*,4*Z*)-dienoyl azide [(*2E,4Z*)-5d]. Yield 100% from acid (*2E,4Z*)-**11d**; yellow solid. ^{1}H NMR (500MHz, $CDCl_3$) δ 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 (125MHz, $CDCl_3$) δ 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 [(*2E,4Z*)-5e]. Yield 91% from acid (*2E,4Z*)-**11e**; yellow solid. ^{1}H NMR (500MHz, $CDCl_3$) δ 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 (125MHz, $CDCl_3$) δ 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 [(*2E,4Z*)-5f]. Yield 83% from acid (*2E,4Z*)-**11f**; yellow solid. ^{1}H NMR (500MHz, $CDCl_3$) δ 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 (125MHz, $CDCl_3$) δ 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 (*2E,4Z*)-**11g**; yellow solid. ^{1}H NMR (500MHz, $CDCl_3$) δ 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 (125MHz, $CDCl_3$) δ 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 (*2E,4Z*)-**11h**; yellow solid. ^1H NMR (400MHz, $CDCl_3$) δ 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, $CDCl_3$) δ 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 [(*2E,4Z*)-5i]. Yield 95% from acid (*2E,4Z*)-**11i**; yellow solid. ^1H NMR (400MHz, $CDCl_3$) δ 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, $CDCl_3$) δ 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 $C_{20}H_{19}N_3O_4$: 337.1314, found: 337.13223 $[M-N_2]^+$.

4-(3',4'-Dimethoxyphenyl)-5-(4"-nitrophenyl)penta-(2*E*,4*Z*)-dienoyl azide [(2*E*,4*Z*)-5j]. Yield 95% from acid (*2E,4Z*)-**11j**; yellow solid. ^1H NMR (400MHz, $CDCl_3$) δ 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, $CDCl_3$) δ 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 (*2E,4Z*)-**11k**; yellow solid. ^1H NMR (400MHz, $CDCl_3$) δ 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, $CDCl_3$) δ 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 (*2E,4Z*)-**11l**; yellow solid. ^{1}H NMR (400MHz, $CDCl_3$) δ 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, $CDCl_3$) δ 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 $C_{19}H_{16}N_4O_5$: 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°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.,[29] mp 284–284.5°C). ^{1}H NMR (500MHz, DMSO-d_6) δ 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) δ 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.,[10] mp 100−101°C). ^{1}H NMR (400MHz, $CDCl_3$) 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); ^{13}C NMR (100MHz, $CDCl_3$) 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 $C_{16}H_{12}$: 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-$CHCl_3$). ^{1}H NMR (600MHz, $CDCl_3$) 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 (150MHz, $CDCl_3$) 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.,[30] mp 150−151°C). ^{1}H NMR (500MHz, $CDCl_3$) 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 (125MHz, $CDCl_3$) 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-$CHCl_3$). ^{1}H NMR (500MHz, $CDCl_3$) 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 (125MHz, $CDCl_3$) 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.,[31] mp 134−135°C). ^1H NMR (400MHz, $CDCl_3$) 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 (100MHz, $CDCl_3$) 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 (400MHz, 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 (100MHz, 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.,[32] mp 139−140°C). ^1H NMR (500MHz, $CDCl_3$) 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 (125MHz, $CDCl_3$) 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 (600MHz, 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 (150MHz, 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.,[33] mp 190°C). ^{1}H NMR (400MHz, $CDCl_3$) 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 (100MHz, $CDCl_3$) 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-$CHCl_3$). ^{1}H NMR (400MHz, 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 (100MHz, DMSO-d_6) 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 $C_{20}H_{19}NO_4$: 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.,[34] mp 158–159°C). ^{1}H NMR (400MHz, $CDCl_3$) 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 (100MHz, $CDCl_3$) 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-$CHCl_3$). ^{1}H NMR (500MHz, $CDCl_3$) 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 (125MHz, $CDCl_3$) 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-$CHCl_3$). ^{1}H NMR (400MHz, 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 (100MHz, 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-$CHCl_3$). ^{1}H NMR (400MHz, $CDCl_3$) 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 (100MHz, $CDCl_3$) 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-$CHCl_3$). ^{1}H NMR (500MHz, 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 (125MHz, 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 (500MHz, $CDCl_3$) 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 (125MHz, $CDCl_3$) 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 $C_{19}H_{18}O_3$: 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-$CHCl_3$). ^1H NMR (400MHz, $CDCl_3$) 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); ^{13}C NMR (100MHz, $CDCl_3$) 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 $C_{19}H_{16}N_2O_5$: 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-$CHCl_3$). ^1H NMR (400MHz, $CDCl_3$) 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); ^{13}C NMR (100MHz, $CDCl_3$) 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 $C_{18}H_{15}NO_4$: 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-$CHCl_3$). ^1H NMR (400MHz, $CDCl_3$) δ 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 (100MHz, $CDCl_3$) δ 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.,[35] mp 168−170°C). ^{1}H NMR (400MHz, $CDCl_3$) δ 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 (100MHz, $CDCl_3$) 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 $C_{20}H_{20}O_4$: 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 (500MHz, $CDCl_3$) 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), 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 (125MHz, $CDCl_3$) 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 $C_{19}H_{16}N_2O_5$: 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 (400MHz, $CDCl_3$) 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); ^{13}C NMR (100MHz, $CDCl_3$) 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 $C_{18}H_{15}NO_4$: 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 (500MHz, $CDCl_3$) 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); ^{13}C NMR (125MHz, $CDCl_3$) 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, 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.

X-ray crystallographic data of 4j. Colorless, single crystals of **4j** suitable for X-ray diffraction study were grown by recrystallization from hexane-CH_2Cl_2. 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-1*H*-indene (**4j**): $C_{18}H_{15}NO_4$, Mr = 309.31; Crystal size 0.60 × 0.26 × 0.17 mm^3; 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 $Å^3$; Z = 8; Dc = 1.381 Mg/m^3. The structures were refined by full-matrix least-squares on F^2 using SHELEXL-97 (Sheldrick, 2008). Final discrepancy indices of R_1 = 0.0431, wR_2 = 0.0699 and GOOF = 0.957 were obtained for observed data with $I > 2\sigma(I)$. 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 (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

(1) Eloy, F.; Deryckere, A. *J. Heterocycl. Chem.* **1970**, *7*, 1191-1193.

(2) Mikol, G. J.; Boyer, J. H. *J. Org. Chem.* **1972**, *37*, 724-726.

(3) MacMillan, J. H.; Washburne, S. S. *J. Org. Chem.* **1973**, *38*, 2982-2984.

(4) Ozaki, S. *Chem. Rev.* **1972**, *72*, 457-496.

(5) Luo, L.; Bartberger, M. D.; Dolbier, Jr., W. R. *J. Am. Chem. Soc.* **1997**, *119*, 12366-12367.

(6) Kobayashi, K.; Kitamura, T.; Yoneda, K.; Morikawa, O.; Konishi, H. *Chem. Lett.* **2000**, *29*, 798-799.

(7) Choshi, T.; Hibino, S. *Heterocycles* **2011**, *83*, 1205-1239.

(8) Chuang, T. H.; Wu, P. L. *J. Chin. Chem. Soc.* **2006**, *53*, 413-420.

(9) Jiang, B.; Li, C.; Tu, S. J. Shi, F. *J. Comb. Chem.* **2010**, *12*, 482-487.

(10) Qin, C.; Lu, W. *J. Org. Chem.* **2008**, *73*, 7424-7427.

(11) Aubert, C.; Betschmann, P.; Eichberg, M. J.; Gandon, V.; Heckrodt, T. J.; Lehmann, J.; Malacria, M.; Masjost, B.; Paredes, E.; Vollhardt, K. P. C.; Whitener, G. D. *Chem. Eur. J.* **2007**, *13*, 7443-7464.

(12) Cheng, C. C. *Med. Hypotheses* **1986**, *20*, 157-172.

(13) Heim, R.; Lucas, S.; Grombein, C. M.; Ries, C.; Schewe, K. E.; Negri, M.; Müller-Vieira, U.; Birk. B.; Hartmann, R. W. *J. Med. Chem.* **2008**, *51*, 5064-5074.

(14) Buckley III, T. F.; Rapoport, H. *J. Org. Chem.* **1983**, *48*, 4222-4232.

(15) Sugiyama, Y.; Suzuki, Y.; Mitamura, S.; Nishiyama, T. *Bull. Chem. Soc. Jpn.* **1993**, *66*, 687-691.

(16) Chuang, T. H.; Lee, S. J.; Yang, C. W.; Wu, P. L. *Org. Biomol. Chem.* **2006**, *4*, 860-867.

(17) Ideses, R.; Shani, A. *J. Am. Oil Chem. Soc.* **1989**, *66*, 948-952.

(18) McMurry, J. E.; Silvestri, M. G.; Fleming, M. P.; Hoz, T.; Grayston, M. W. *J. Org. Chem.* **1978**, *43*, 3249-3255.

(19) Kulp, S. S.; Caldwell, C. B. *J. Org. Chem.* **1980**, *45*, 171-173.

(20) Kiewit, T. D.; Stephen, H. *J. Chem. Soc.* **1931**, 639-640.

(21) Loupy, A.; Pellet, M.; Petit, A.; Vo-Thanh, G. *Org. Biomol. Chem.* **2005**, 3, 1534-1540.

(22) Niederl, J. B.; Ziering, A. *J. Am. Chem. Soc.* **1942**, *64*, 885-886.

(23) Yue, Y.; Fang, H.; Wang, M.; Wang, Z.; Yu, M.; *J. Chem. Res.* **2009**, *6*, 377-380.

(24) Yamashita, S.; Tokuda, M.; Orito, K.; Kurono, N.; Senboku, H. *Eur. J. Org. Chem.* **2009**, *8*, 1173-1180.

(25) Nalesnik, T. E.; Fish, J. G.; Horgan, W.; Orchin, M. *J. Org. Chem.* **1981**, *46*, 1987-1990.

(26) Molander, G. A.; Fumagalli, T. *J. Org. Chem.* **2006**, *71*, 5743-5747.

(27) Cacchi, S.; Fabrizi, G.; Marinelli, F.; Moro, L.; Pace, P. *Tetrahedron* **1996**, *52*, 10225-10240.

(28) Grieco, P. A.; Parker, D. T. *J. Org. Chem.* **1988**, *53*, 3325-3330.

(29) Masamune, S.; Fukumoto, K. *Tetrahedron Lett.* **1965**, *6*, 4647-4654.

(30) Shishido, K.; Yamashita, A.; Hiroya, K.; Fukumoto, K. *J. Chem. Soc. Perkin Trans 1* **1990**, 469-475.

(31) Cooke, R. G.; Johnson, B. G.; Segal, W. *Aust. J. Chem.* **1958**, *11*, 230-235.

(32) Cho, C. H.; Park, H.; Park, M. A.; Ryoo, T. Y.; Lee, Y. S.; Park, K. *Eur. J. Org. Chem.* **2005**, *15*, 3177-3181.

(33) Mewshaw, R. E.; Edsall, R. J.; Yang, C.; Manas, E. S.; Xu, Z. B.; Henderson, R. A.; Keith, J. C.; Harris, H. A. *J. Med. Chem.* **2005**, *48*, 3953-3979.

(34) Gopinath, K. W.; Govindachari, T. R.; Nagarajan, K.; Viswanathan, N. *J. Chem. Soc.* **1957**, 4760-4764.

(35) Dominguez, E.; Iriondo, C.; Laborra, C.; Lopez, P.; Martinez, J.; Zamora, F. *J. Heterocycl. Chem.* **1989**, *26*, 549-551.