# Synthesis of 2,4-disubstituted furans and 4,6-diaryl-substituted 2,3-benzo-1,3a,6a-triazapentalenes

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**Dedicated to our long-time friend Binne Zwanenburg**

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**Abstract**

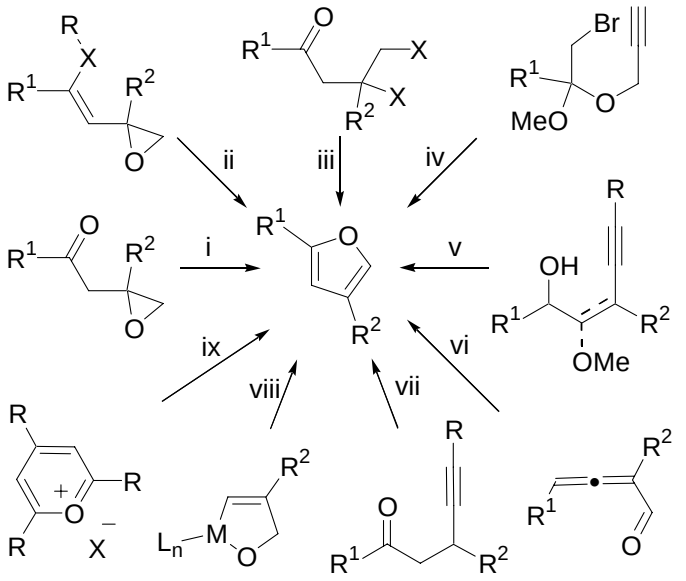
Reactions of acylacetylenes **1a–h** with benzotriazole **2** give intermediates **3a–h**. The treatment of **3a–h** with trimethylsulfonium iodide in the presence of base give intermediate oxiranes **4a–h** and 2,3-benzo-1,3a,6a-triazapentalenes **7d–g** depending on substituent. Acid-catalyzed rearrangement of crude **4a–h** give 2,4-disubstituted furans **5a–h**.

**Keywords:** 2,4-Disubstituted furans, heteropentalenes, 2,3-benzo-1,3a,6a-triazapentalenes, oxiranes

## Introduction

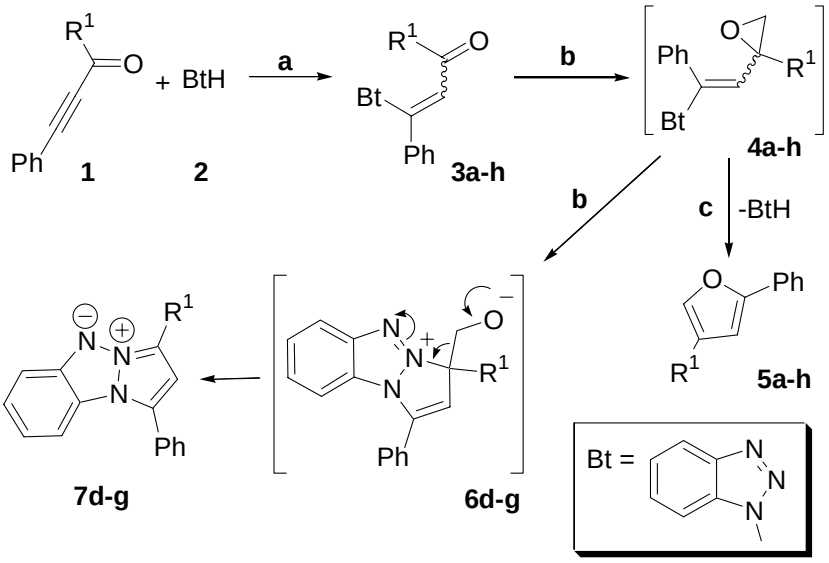
The furan ring is an important structural unit in many biologically active and naturally occurring compounds.[1] Furan syntheses and their applications have been reviewed in detail.[1d,2] The available strategies for the preparation of 2,4-disubstituted furans (Scheme 1) utilize ring construction via: (i) acid-catalyzed rearrangement of oxiranyl ketones,[1c,3] and (ii) their enol (or thioenol) ethers;[1a,1e–g,4] (iii) thermal annulation of 3,4-dihaloketones;[5] (iv) radical cyclization – reductive demethoxylation of bromoketals;[6] palladium-catalyzed cyclization of (v) 2-methoxy- or 2-unsaturated pent-4-yn-1-ol,[7] and (vi) allenyl aldehydes;[8] (vii) base-catalyzed cyclization of γ-alkynyl ketones,[9] (viii) carbon insertion into cyclic enol – metal complexes;[10] and (ix) an oxidative ring transformation of pyrylium salts.[11]



**Scheme 1**

To develop a procedure for regioselective preparation of unsymmetrical 2,4-disubstituted furans, we have investigated the reactivity of 2-oxiranyl-vinyl benzotriazoles **4** (Scheme 2) toward rearrangement followed by elimination of benzotriazole to afford furans **5**. We now disclose the preparation of 2,4-disubstituted furans **5a–h** in two steps from acylacetylenes **1a–h** in methodology that utilizes the leaving capability of the benzotriazole group[12] and thus compliments the Garst-Spencer furan synthesis.[1a] In the course of our investigation we also have found and now report the preparation of heteropentalenes **7d–g** from benzoylacetylenes **1d–g** via a mechanism which may involve intermediates **6d–g**.



**Scheme 2.** For designation of $R^1$ see Table 1. a) *t*-BuOK (0.05 equivalent), toluene, 110 °C, 3–4 h; b) $Me_3S^+I^-$, *n*-$Bu_4NI$, $CH_2Cl_2$ – 50% NaOH, 20–40 °C; c) *p*-TsOH (0.03 equivalent) THF, 50–60 °C.

**Table 1.** Preparation of compounds **3a–h**, furans **5a–h**, and 2,3-benzo-1,3a,6a-triazapentalenes **7d–g**

| Entries | $R^1$ | Yields, % | | |
|---|---|---|---|---|
| | | **3**[a] | **5**[b] | **7**[b] |
| **a** | Me | 67 | 38 | – |
| **b** | Et | 81 | 33 | – |
| **c** | *t*-Bu | 84 | 28 | – |
| **d** | *p*-Tol | 83 | 27 | 71 |
| **e** | 4-Cl-$C_6H_4$ | 72 | 50 | 8 |
| **f** | 2-Thienyl | 84 | 12 | 42 |
| **g** | 4-MeO-$C_6H_4$ | 64 | 18 | 42 |
| **h** | 4-$NO_2$-$C_6H_4$ | 89 | 83 | – |

[a] Mixture of cis/trans isomers. The combined yields are given. [b)] Yields are based on intermediates **3**.

## Result and Discussion

Acylacetylenes **1a–h** and benzotriazole **2** (Scheme 2) react in the presence of catalytic amounts of *t*-BuOK in toluene under reflux to give *β*-benzotriazolyl unsaturated ketones **3a–h** as mixtures of *cis*/*trans*-isomers in good yields. Structures **3a–h** were supported by their $^1$H and $^{13}$C NMR spectra. Compounds **3a–h** were converted to oxiranes **4a–h** by treatment with trimethylsulfonium iodide (1.3–2.0 equivalents) in methylene chloride – 50% aqueous NaOH two-phase system in the presence of tetrabutylammonium iodide at 20–40 °C for 12–48 h.[13] Crude oxiranes **4a–h** rearranged in the presence of *p*-toluenesulfonic acid in THF solution at 50–60 °C to afford furans **5a–h**. To support the reaction pathway proposed, intermediate **4a** was isolated, but because of its instability only $^1$H and $^{13}$C NMR spectra of **4a** were acquired and used to deduce its structure. The $^1$H NMR spectrum of **4a** shows a new set of signals: two doublets at 2.49 ppm and 2.60 ppm were assigned to the oxirane ring protons and the singlet at 1.28 ppm was assigned to the methyl group in **4a**. The $^{13}$C spectrum of **4a** shows no carbonyl group signal, while two new signals at 54.8 ppm and 55.0 ppm were assigned to oxirane ring carbons.

The structures of furans **5a–h** were supported by their $^1$H and $^{13}$C NMR spectra. Their $^1$H NMR spectra no longer show signals in the range 7.0–8.2 ppm corresponding to the benzotriazolyl group in **3a–h** and **4a**, nor signals for oxirane ring protons as for intermediate **4a**. In the $^{13}$C NMR spectra of **5a–h**, the signals around 181–204 ppm corresponding to the carbonyl group in **3a–h**, as well as signals around 111 ppm, 120 ppm, 133 ppm, and 146 ppm assigned to benzotriazolyl groups in **3a–h** are no longer present. New signals in the $^{13}$C NMR spectra of **5a–**

**h** in the ranges 103.3–107.7 ppm, 137.1–139.0 ppm, and 153.8–155.8 ppm are distinctive for 2,4-disubstituted furans[10b,14] and were assigned to the carbon atoms of the furan ring formed.

The treatment of **3d–g** with trimethylsulfonium iodide in the presence of base gave, in addition to compounds **5d–g**, isolable byproducts. The $^{1}$H NMR spectra of these byproducts no longer appear to be typical for N-substituted benzotriazoles with $^{1}$H signals around 8.0–8.2 ppm as in **3a–h**. However, the $^{13}$C NMR spectra of the byproducts show signals near 111 ppm, 120 ppm and 147 ppm, which together with elemental analyses suggested the presence of a benzotriazole moiety. The $^{13}$C NMR spectra also show two new signals in the range 105.0–105.5 ppm and 113.1–113.7 ppm. A single crystal X-ray structure determination for the product obtained from **3e** unambiguously demonstrated it to be the 2,3-benzo-1,3a,6a-triazapentalene **7e** (Figure 1). The bonding geometry within the molecule is similar to that in the only other reported X-ray structure containing this heterocyclic ring system.[15] Heteropentalenes of this type cannot be represented by a single valence bond description. Based on the fact that the 4-chlorophenyl ring is approximately coplanar with the benzopentalene system [angle between meanplanes = 5.9(1)º] whereas the phenyl ring is twisted out of this plane [by 45.2(1)º] it is probable that the atom labelled C3 carries significant charge. In accord with this proposal is the observation that the C3-C10 bond is significantly shorter than the C1-C16 bond.

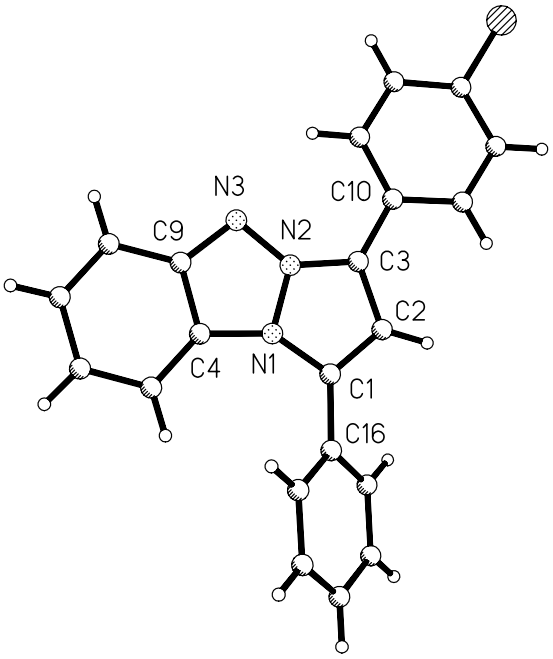


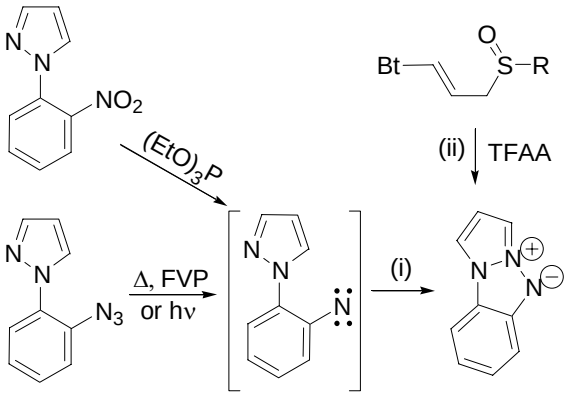

**Figure 1.** Perspective view and selected atom labelling of the X-ray crystal structure of **7e**. Selected bond lengths (Å): N1-N2 1.380(2), N1-C1 1.384(2), N1-C4 1.397(2), N2-N3 1.356(2), N2-C3 1.368, N3-C9 1.373(2), C1-C2 1.380(2), C2-C3 1.410(2), C1-C16 1.471(2), C3-C10 1.456(2).

The $^{1}H$ and $^{13}C$ NMR spectra of these byproducts also support the heteropentalene structures **7d–g**. The reaction mechanism for the formation of **7d–g** probably involves nucleophilic attack on the epoxide in **4d–g** from the 2-position benzotriazole nitrogen to give intermediate **6d–g**. Intermediates **6d–g** then eliminate formaldehyde with the formation of 4,6-diarylsubstituted 2,3-benzo-1,3a,6a-triazapentalenes **7d–g** in 8–71 % yields. Interestingly, for compound **3e**, the corresponding heteropentalene **5e** was obtained only in 8 % yield while the formation of **7h** was not detected.

To examine the possible differences in the reactivity of individual isomers of intermediates **3**, we separated mixtures of *cis*/*trans*-isomers **3a,c,e–h**. However, subsequent reactions of the individual *cis* and *trans* isomers of **3** with trimethylsulfonium iodide and *p*-toluenesulfonic acid gave the same final product composition in all cases.

## Conclusions

New routes for the preparation of 2,4-disubstituted furans and 4,6-diarylsubstituted 2,3-benzo-1,3a,6a-triazapentalenes have been established. This methodology provides easy access to 2-aryl-4-alkylsubstituted furans as well as to 2,4-diarylsubstituted furans with electron withdrawing 4-aryl group in reasonable yields and has practical importance for the synthesis of unsymmetric furans. The procedure also avoids the use of sulfur containing starting materials in the original Garst-Spencer furan synthesis.[1a] Unfortunately, the present procedure could not be extended to 4-alkyl substituted furans: Alkyl-substituted *β*-Benzotriazolyl-α,*β*-unsaturated allcylketones gave complex reaction mixtures on treatment with trimethyl sulfonium iodide in a basic medium. Simultaneously, the present procedure allows the synthesis of 4,6-diarylsubstituted 2,3-benzo-1,3a,6a-triazapentalenes with electron donating 6-aryl group in good overall yields. Available methods for the preparation of pyrazolo[1,2-a]benzotriazoles (Scheme 3) include (i) intramolecular annulation of 1-(2-nitrenophenyl)pyrazoles generated by deoxygenation of the corresponding 1-(2-nitrophenyl)pyrazoles,[16] thermolysis or photolysis of 1-(2-azidophenyl)pyrazoles;[16b,17] and (ii) recently reported, the multistep preparation via Pummerer-type annulation of γ-(benzotriazol-1-yl)allyl sulfoxides to 6-sulfanyl-2,3-benzo-1,3a,6a-triazapentalenes.[15]




**Scheme 3**

## Experimental Section

**General Procedures.** Melting points were determined on a hot-stage apparatus and are uncorrected. $^1$H and $^{13}$C NMR spectra were recorded in $CDCl_3$ with TMS as the internal standard for $^1$H (300 MHz) or a solvent as the internal standard for $^{13}$C (75 MHz). THF was dried over sodium / benzophenone and used freshly distilled. Column chromatography was conducted on silica gel 200–425 meshes.

**Materials.** The acylacetylenes **1b–h** were prepared according to the following published procedures:[18] 1-phenyl-1-pentyn-3-one **(1b)**;[18a] 4,4-dimethyl-1-phenyl-1-pentyn-3-one **(1c)**;[18b] 1-(4-methylphenyl)-3-phenyl-2-propyn-1-one **(1d)**;[18b] 1-(4-chlorophenyl)-3-phenyl-2-propyn-1-one **(1e)**;[18b] 3-phenyl-1-(2-thienyl)-2-propyn-1-one **(1f)**;[18b] 1-(4-methoxyphenyl)-3-phenyl-2-propyn-1-one **(1g)**;[18b] 1-(4-nitrophenyl)-3-phenyl-2-propyn-1-one **(1h)**.[18b]

**1-Phenyl-1-pentyn-3-one (1b).** Yellow oil[19] (45%); $^1$H NMR δ 7.58–7.55 (m, 2H), 7.45–7.35 (m, 3H), 2.70 (q, *J* = 7.4 Hz, 2H), 1.21 (t, *J* = 7.4 Hz, 3H); $^{13}$C NMR δ 8.0, 38.7, 87.5, 90.5, 119.9, 128.5, 130.6, 132.9, 188.5.

**4,4-Dimethyl-1-phenyl-1-pentyn-3-one (1c).** Yellow oil[20] (98%); $^1$H NMR δ 7.60–7.57 (m, 2H), 7.48–7.35 (m, 3H), 1.28 (s, 9H); $^{13}$C NMR δ 26.1, 44.8, 86.0, 92.2, 120.2, 128.6, 130.5, 132.9, 194.2.

**1-(4-Methylphenyl)-3-phenyl-2-propyn-1-one (1d).** White microcrystals from ethyl acetate / hexanes (76%), mp 69–70 °C (lit.[18b] 86–88 °C); $^1$H NMR δ 8.11 (d, *J* = 8.2 Hz, 2H), 7.69–7.66 (m, 2H), 7.47–7.38 (m, 3H), 7.30 (d, *J* = 8.2 Hz, 2H), 2.44 (s, 3H); $^{13}$C NMR δ 21.8, 86.9, 92.5, 120.2, 128.6, 129.3, 129.6, 130.6, 133.0, 134.5, 145.2, 177.6. Anal. Calcd for $C_{16}H_{12}O$: C, 87.25; H, 5.49; Found: C, 87.14; H, 5.37.

**1-(4-Chlorophenyl)-3-phenyl-2-propyn-1-one (1e).** White microcrystals from ethyl acetate / hexanes (39%), mp 101–102 °C; $^1$H NMR δ 8.15 (d, *J* = 8.5 Hz, 2H), 7.69–7.66 (m, 2H), 7.52–7.40 (m, 5H); $^{13}$C NMR δ 86.6, 93.6, 119.9, 128.8, 129.0, 130.9, 131.0, 133.1, 135.3, 140.7, 176.7. Anal. Calcd for $C_{15}H_9ClO$: C, 74.85; H, 3.77; Found: C, 74.83; H, 3.55.

**3-Phenyl-1-(2-thienyl)-2-propyn-1-one (1f).** Brown microcrystals from methanol (90%), mp 56–57 °C; $^1$H NMR δ 8.01 (dd, *J* = 3.8, 1.0 Hz, 1H), 7.73 (dd, *J* = 4.8, 1.0 Hz, 1H), 7.67–7.64 (m, 2H), 7.51–7.39 (m, 3H), 7.20–7.18 (m, 1H); $^{13}$C NMR δ 86.4, 91.7, 119.9, 128.3, 128.6, 130.8, 133.0, 135.0, 135.2, 144.9, 169.7. Anal. Calcd for $C_{13}H_8OS$: C, 73.56; H, 3.80; Found: C, 73.16; H, 3.67.

**1-(4-Methoxyphenyl)-3-phenyl-2-propyn-1-one (1g).** Brown microcrystals from ethyl acetate / hexane (35%), mp 93–94 °C; $^1$H NMR δ 8.21–8.17 (m, 2H), 7.69–7.65 (m, 2H), 7.47–7.39 (m, 3H), 7.00–6.97 (m, 2H), 3.89 (s, 3H); $^{13}$C NMR δ 55.5, 86.9, 92.2, 113.8, 120.3, 128.6, 130.2, 130.5, 131.9, 132.9, 164.4, 176.6. Anal. Calcd for $C_{16}H_{12}O_2$: C, 81.34; H, 5.12; Found: C, 81.13; H, 5.18.

**1-(4-Nitrophenyl)-3-phenyl-2-propyn-1-one (1h).** Brown microcrystals from ethyl acetate / hexanes (77%), mp 160–161 °C (lit.[18b] 162–163 °C); $^1$H NMR δ 8.42–8.34 (m, 4H), 7.73–7.70 (m, 2H), 7.57–7.44 (m, 3H); $^{13}$C NMR δ 86.5, 95.3, 119.3, 123.8, 128.8, 130.4, 131.4, 133.2, 141.0, 150.8, 175.8.

**General procedure for the preparation of 3a–h**

A catalytic amount of potassium *tert*-butoxide (5–10 mg) was added to a solution of acylacetylene **1a–h** (10 mmol) and benzotriazole (**2**) (1.55 g, 13 mmol) in toluene and the reaction mixture was refluxed for 3–5 h under a nitrogen atmosphere. Then the reaction mixture was cooled down to room temperature and washed with saturated aqueous solution of sodium carbonate to remove excess benzotriazole. The organic layer was dried over magnesium sulfate, and the solvent was evaporated in vacuum. The crude product (mixture of *cis*/*trans*-isomers) was purified by column chromatography to give **3a–h** (the crude product may be used on the next step without additional purification).

**4-(1*H*-Benzotriazol-1-yl)-4-phenyl-3-buten-2-one (3a). First isomer.** Yellow needles from diethyl ether (45%), mp 131–132 °C; $^1$H NMR δ 8.09 (d, *J* = 8.4 Hz, 1H), 7.63 –7.57 (m, 1H), 7.54–7.48 (m, 2H), 7.42 (d, *J* = 7.3 Hz, 2H), 7.35 (t, *J* = 7.7 Hz, 1H), 7.29–7.24 (m, 1H), 7.03 (s, 1H), 6.43 (d, *J* = 8.4 Hz, 1H), 2.17 (s, 3H); $^{13}$C NMR δ 31.3, 111.9, 120.7, 121.1, 124.9, 128.6, 129.3, 130.1, 131.5, 132.3, 132.8, 145.8, 147.0, 197.9. Anal. Calcd for $C_{16}H_{13}N_3O$: C, 72.99; H, 4.98; N, 15.96. Found: C, 73.17; H, 5.00; N, 16.15.

**Second isomer.** Colorless needles from diethyl ether (22%), mp 118–119 °C; $^1$H NMR δ 8.17–8.12 (m, 1H), 7.52–7.37 (m, 5H), 7.25–7.23 (m, 2H), 7.06–7.01 (m, 1H), 6.79 (s, 1H), 1.91 (s, 3H); $^{13}$C NMR δ 30.0, 110.8, 120.5, 124.7, 124.8, 127.6, 128.7, 129.4, 131.6, 133.6, 134.0, 142.4, 146.2, 196.7. Anal. Calcd for $C_{16}H_{13}N_3O$: C, 72.99; H, 4.98; N, 15.96. Found: C, 73.18; H, 4.97; N, 16.17.

**1-(1*H*-Benzotriazol-1-yl)-1-phenyl-1-penten-3-one (3b).** Yellow plates from ethyl acetate / hexanes (mixture of isomers) (81%), mp 63–64 ºC; $^1$H NMR δ 8.15–8.06 (m, 2H), 7.51–7.45 (m, 2H), 7.43–7.31 (m, 8H), 7.27–7.22 (m, 4H), 7.06–7.00 (m, 2H), 6.82 (s, 1H, major), 6.38 (d, *J* = 8.4 Hz, 1H, minor), 2.54 (q, *J* = 14.5, 7.3 Hz, 2H, minor), 2.34 (q, *J* = 14.5, 7.3 Hz, 2H, major), 1.07 (t, *J* = 7.3 Hz, 3H, minor), 0.99 (t, *J* = 7.3 Hz, 3H, major); $^{13}$C NMR δ (for the mixture of isomers): 7.7, 8.0, 36.4, 37.3, 110.6, 111.7, 119.7, 120.2, 120.3, 123.4, 124.3, 124.5, 127.3, 128.2, 128.3, 128.8, 129.1, 129.7, 130.9, 131.2, 132.0, 132.6, 133.3, 133.9, 141.5, 144.9, 145.8, 146.7, 199.2, 200.2. Anal. Calcd for $C_{17}H_{15}N_3O$: C, 73.63; H, 5.45; N, 15.15. Found: C, 73.72; H, 5.48; N, 15.07.

**1-(1*H*-Benzotriazol-1-yl)-4,4-dimethyl-1-phenyl-1-penten-3-one (3c). First isomer.** Yellow microcrystals from ethyl acetate / hexanes (37%), mp 74–75 °C; $^1$H NMR δ 8.08 (d, *J* = 8.2 Hz, 1H), 7.57–7.52 (m, 1H), 7.49–7.30 (m, 6H), 7.26–7.20 (m, 1H), 6.31 (d, *J* = 8.4 Hz, 1H), 1.26 (s, 9H); $^{13}$C NMR δ 26.4, 44.5, 111.8, 115.9, 120.4, 124.5, 128.2, 128.7, 129.6, 130.6, 132.3, 132.6, 145.9, 146.8, 204.0. Anal. Calcd for $C_{19}H_{19}N_3O$: C, 74.73; H, 6.27; N, 13.76. Found: C, 74.61; H, 6.34; N, 13.75.

**Second isomer.** Yellow microcrystals from ethyl acetate / hexanes (37%), mp 147–148 °C; $^1$H NMR δ 8.12–8.09 (m, 1H), 7.51–7.33 (m, 5H), 7.27 (d, *J* = 7.4 Hz, 2H), 7.11 (s, 1H), 6.99–6.96 (m, 1H), 1.26 (s, 9H); $^{13}$C NMR δ 26.3, 44.3, 110.8, 120.2, 120.4, 124.0, 127.3, 127.9, 129.1, 131.1, 133.5, 134.4, 142.3, 145.9, 203.8. Anal. Calcd for $C_{19}H_{19}N_3O$: C, 74.73; H, 6.27; N, 13.76. Found: C, 74.74; H, 6.37; N, 13.75.

**3-(1*H*-Benzotriazol-1-yl)-1-(4-methylphenyl)-3-phenyl-2-propen-1-one (3d).** Yellow microcrystals from ethyl acetate / hexanes (mixture of isomers) (83%), mp 105–108 °C; $^1$H NMR δ 8.11 (d, *J* = 8.2 Hz, 1H, minor), 8.00 (d, *J* = 7.1 Hz, 1H, major), 7.91 (d, *J* = 8.2 Hz, 2H, minor), 7.70 (d, *J* = 8.2 Hz, 2H, major), 7.60–7.32 (m, 13H), 7.27–7.22 (m, 4H), 7.09–7.01 (m, 3H), 6.50 (d, *J* = 8.4 Hz, 1H, minor), 2.39 (s, 3H, minor), 2.31 (s, 3H, major); $^{13}$C NMR δ (for the mixture of isomers): 21.5, 21.6, 110.8, 111.7, 118.7, 120.0, 120.3, 121.3, 124.1, 124.5, 127.5, 128.0, 128.2, 128.4, 128.7, 128.9, 129.0, 129.1, 129.2, 129.6, 130.6, 131.2, 132.2, 132.6, 133.4, 134.1, 134.5, 135.1, 142.8, 144.0, 144.2, 145.2, 145.8, 146.7, 189.7, 189.9. Anal. Calcd for $C_{22}H_{17}N_3O$: C, 77.86; H, 5.05; N, 12.38. Found: C, 77.58; H, 5.13; N, 12.43.

**3-(1*H*-Benzotriazol-1-yl)-1-(4-chlorophenyl)-3-phenyl-2-propen-1-one (3e). First isomer.** Yellow microcrystals from ethyl acetate / hexanes (39%), mp 156–157 °C; $^1$H NMR δ 8.12 (d, *J* = 8.24 Hz, 1H), 7.95–7.93 (m, 2H), 7.60 (s, 1H), 7.53–7.48 (m, 1H), 7.44–7.34 (m, 7H), 7.30–7.25 (m, 1H), 6.43 (d, 8.4 Hz, 1H); $^{13}$C NMR δ 111.8, 117.5, 120.5, 124.7, 128.4, 128.9, 128.9, 129.7, 130.1, 131.0, 132.0, 132.6, 136.0, 139.7, 146.3, 146.8, 189.1. Anal. Calcd for $C_{21}H_{14}ClN_3O$: C, 70.10; H, 3.92; N, 11.68. Found: C, 70.16; H, 3.78; N, 11.68.

**Second isomer.** White microcrystals from ethyl acetate / hexane (33%), mp 140–141 °C; $^1$H NMR δ 8.02–7.99 (m, 1H), 7.72–7.69 (m, 2H), 7.59–7.52 (m, 1H), 7.48–7.43 (m, 2H), 7.40–7.34 (m, 4H), 7.26–7.21 (m, 2H), 7.16 (s, 1H), 6.99–6.96 (m, 1H); $^{13}$C NMR δ 110.8, 120.3, 120.7, 124.4, 127.6, 128.2, 128.6, 129.3, 129.6, 131.5, 133.3, 133.7, 135.4, 139.4, 143.6, 146.0, 189.3. Anal. Calcd for $C_{21}H_{14}ClN_3O$: C, 70.10; H, 3.92; N, 11.68. Found: C, 70.18; H, 3.80; N, 11.70.

**3-(1*H*-Benzotriazol-1-yl)-3-phenyl-1-(2-thienyl)-2-propen-1-one (3f). First isomer.** Yellow microcrystals from ethyl acetate / hexanes (50%), mp 137–138 °C; $^1$H NMR δ 8.11 (d, *J* = 8.4 Hz, 1H), 7.85–7.83 (m, 1H), 7.67–7.65 (m, 2H), 7.57–7.44 (m, 5H), 7.38–7.33 (m, 1H), 7.28–7.23 (m, 1H), 7.14–7.11 (m, 1H), 6.39 (d, *J* = 8.4 Hz, 1H); $^{13}$C NMR δ 111.9, 116.5, 120.4, 124.6, 127.9, 128.3, 128.4, 128.7, 129.8, 130.9, 132.0, 132.7, 134.5, 145.7, 146.4, 146.8, 181.6. Anal. Calcd for $C_{19}H_{13}N_3OS$: C, 68.86; H, 3.95; N, 12.68. Found: C, 68.90; H, 3.77; N, 12.56.

**Second isomer.** Yellow prisms from ethyl acetate / hexanes (34%), mp 161–162 °C; $^1$H NMR δ 8.09–8.06 (m, 1H), 7.69 (d, *J* = 3.7 Hz, 1H), 7.59 (d, *J* = 4.9 Hz, 1H), 7.55–7.50 (m, 1H), 7.46–7.34 (m, 6H), 7.29 (s, 1H), 7.07–7.01 (m, 2H); $^{13}$C NMR δ 110.8, 119.7, 120.2, 124.2, 127.6, 128.0, 128.2, 129.2, 131.5, 132.3, 133.5, 134.3, 134.5, 143.5, 144.8, 145.8, 180.9. Anal. Calcd for $C_{19}H_{13}N_3OS$: C, 68.86; H, 3.95; N, 12.68. Found: C, 69.02; H, 3.86; N, 12.59.

**3-(1*H*-Benzotriazol-1-yl)-1-(4-methoxyphenyl)-3-phenyl-2-propen-1-one (3g). First isomer.** Yellow microcrystals from ethyl acetate / hexanes (38%), mp 133–134 °C; $^1$H NMR δ 8.11 (d, *J* = 8.2 Hz, 1H), 8.00 (d, *J* = 8.8 Hz, 2H), 7.57 (s, 1H), 7.48–7.25 (m, 7H), 6.91 (d, *J* = 8.5 Hz, 2H), 6.52 (d, *J* = 8.4 Hz, 1H); $^{13}$C NMR δ 55.5, 111.8, 113.8, 119.0, 120.4, 124.5, 128.2, 128.7,

129.6, 130.6, 130.6, 131.2, 132.3, 132.6, 144.8, 146.7, 163.8, 188.9. Anal. Calcd for $C_{22}H_{17}N_3O_2$: C, 74.35; H, 4.82; N, 11.82. Found: C, 73.96; H, 4.80; N, 11.75.

**Second isomer.** Orange oil (26%); $^1$H NMR δ 8.02–8.00 (m, 1H), 7.80 (d, *J* = 8.8 Hz, 2H), 7.53–7.30 (m, 7H), 7.25 (s, 1H), 7.05–7.02 (m, 1H), 6.77 (d, *J* = 8.8 Hz, 2H), 3.78 (s, 3H); $^{13}$C NMR δ 55.4, 110.8, 113.6, 120.1, 121.6, 124.1, 127.4, 128.0, 129.1, 130.1, 130.7, 131.1, 133.5, 134.3, 142.5, 145.8, 163.6, 188.6. Anal. Calcd for $C_{22}H_{17}N_3O_2$: C, 74.35; H, 4.82; N, 11.82. Found: C, 74.27; H, 4.85; N, 11.74.

**3-(1*H*-Benzotriazol-1-yl)-1-(4-nitrophenyl)-3-phenyl-2-propen-1-one (3h). First isomer.** Yellow microcrystals from ethyl acetate / hexanes (48%), mp 153–154 °C; $^1$H NMR δ 8.26–8.23 (m, 2H), 8.14–8.10 (m, 3H), 7.65 (s, 1H), 7.55–7.50 (m, 1H), 7.46–7.35 (m, 5H), 7.30–7.25 (m, 1H), 6.36 (d, *J* = 8.4 Hz, 1H); $^{13}$C NMR δ 111.8, 116.4, 120.6, 123.7, 124.9, 128.7, 129.0, 129.6, 129.8, 131.4, 131.6, 132.6, 142.5, 147.0, 147.8, 150.1, 188.8. Anal. Calcd for $C_{21}H_{14}N_4O_3$: C, 68.10; H, 3.81; N, 15.13. Found: C, 68.12; H, 3.56; N, 15.25.

**Second isomer.** Yellow microcrystals from ethyl acetate / hexanes (41%), mp 164–165 °C; $^1$H NMR δ 8.03–8.00 (m, 2H), 7.95–7.93 (m, 1H), 7.84–7.81 (m, 2H), 7.61–7.55 (m, 1H), 7.51–7.31 (m, 6H), 7.11 (s, 1H), 6.95–6.93 (m, 1H); $^{13}$C NMR δ 110.8, 120.2, 120.3, 123.3, 124.6, 127.8, 128.5, 128.8, 129.4, 131.9, 133.1, 141.7, 144.6, 146.1, 149.7, 189.4. Anal. Calcd for $C_{21}H_{14}N_4O_3$: C, 68.10; H, 3.81; N, 15.13. Found: C, 68.21; H, 3.62; N, 15.14.

## General procedure for the preparation of 5a–h and 7d–g

A vigorously stirred mixture of **3a–h** (4 mmol) with trimethylsulfonium iodide (1.06 g, 5.2 mmol) in the presence of tetrabutylammonium iodide (50 mg, 0.13 mmol) in dichloromethane (15 mL) and 50% aqueous NaOH (15 mL) was refluxed under nitrogen atmosphere for 6–36 h (the reaction progress was monitored by TLC; the addition of extra trimethylsulfonium iodide (1.0–2.0 mmol) may be required). When **3a–h** was consumed, the product was extracted with dichloromethane. The extract was dried over magnesium sulfate and the solvent was evaporated to give the crude intermediate **4a–c,h** or a mixture of **4d–g** and **7d–g**. The residue was dissolved in anhydrous THF, a catalytic amount of *p*-toluenesulfonic acid (10–20 mg) was added and the reaction mixture was refluxed for 0.5–1 h under nitrogen atmosphere. Then the reaction mixture was concentrated in vacuum and the residue was subjected to column chromatography on silica gel to give **5a–h** and **7d–g**.

**4-Methyl-2-phenylfuran (5a).** Colorless needles from hexanes (38%), mp 37–39 °C (lit.[4a] 38–40 °C); $^1$H NMR δ 7.64–7.61 (m, 2H), 7.38–7.32 (m, 2H), 7.25–7.19 (m, 2H), 6.51 (s, 1H), 2.05 (d, *J* = 0.7 Hz, 3H); $^{13}$C NMR δ 9.82, 107.7, 121.9, 123.6, 127.1, 128.6, 131.0, 138.8, 153.8.

**4-Ethyl-2-phenylfuran (5b).** Oil (33%); $^1$H NMR δ 7.63 (d, *J* = 7.4 Hz, 2H), 7.37–7.32 (m, 2H), 7.24–7.19 (m, 2H), 6.55 (s, 1H), 2.46 (q, *J* = 7.6 Hz, 2H), 1.21 (t, *J* = 7.6 Hz, 3H); $^{13}$C NMR δ 14.3, 18.3, 106.3, 123.6, 127.1, 128.6, 129.1, 131.1, 137.9, 153.8. Anal. Calcd for $C_{12}H_{12}O$: C, 83.69; H, 7.02; Found: C, 83.46; H, 7.05.

**4-*tert*-Butyl-2-phenylfuran (5c).** White microcrystals from ethyl acetate / hexane (28%), mp 168–169 °C; $^1$H NMR δ 7.66–7.63 (m, 2H), 7.38–7.33 (m, 2H), 7.26–7.20 (m, 2H), 6.61 (s, 1H),

1.26 (s, 9H); $^{13}$C NMR δ 29.9, 30.8, 104.6, 123.6, 127.1, 128.6, 131.1, 136.3, 138.3, 153.8. Anal. Calcd for $C_{14}H_{16}O$: C, 83.96; H, 8.05; Found: C, 83.63; H, 7.93.

**4-(4-Methylphenyl)-2-phenylfuran (5d).** White microcrystals from ethyl acetate / hexanes (27%), mp 111–112 °C; $^1$H NMR δ 7.72–7.68 (m, 3H), 7.42–7.35 (m, 4H), 7.28–7.23 (m, 1H), 7.18 (d, *J* = 8.1 Hz, 2H), 6.92 (s, 1H), 2.35 (s, 3H); $^{13}$C NMR δ 21.1, 104.0, 123.8, 125.7, 127.5, 128.3, 128.7, 129.5, 130.7, 136.8, 137.6, 154.7. Anal. Calcd for $C_{17}H_{14}O$: C, 87.15; H, 6.02; Found: C, 86.65; H, 5.94.

**4-(4-Chlorophenyl)-2-phenylfuran (5e).** White microcrystals from ethyl acetate / hexanes (50%), mp 127–128 °C; $^1$H NMR δ 7.73–7.69 (m, 3H), 7.47–7.25 (m, 7H), 6.91 (d, *J* = 0.8 Hz, 1H); $^{13}$C NMR δ 103.7, 123.9, 127.0, 127.3, 127.7, 128.7, 128.9, 130.4, 130.9, 132.7, 137.9, 155.1. Anal. Calcd for $C_{16}H_{11}ClO$: C, 75.45; H, 4.35; Found: C, 75.70; H, 4.41.

**2-Phenyl-4-(2-thienyl)furan (5f).** White microcrystals from ethyl acetate / hexanes (12%), mp 71–72 °C; $^1$H NMR δ 7.71–7.68 (m, 3H), 7.42–7.37 (m, 2H), 7.30–7.25 (m, 1H), 7.23–7.19 (m, 1H), 7.15–7.13 (m, 1H), 7.03 (dd, *J* = 4.9, 3.6 Hz, 1H), 6.85 (s, 1H); $^{13}$C NMR δ 104.4, 122.4, 123.4, 123.7, 123.9, 127.6, 127.7, 128.7, 130.4, 134.8, 137.5, 154.8. Anal. Calcd for $C_{14}H_{10}OS$: C, 74.31; H, 4.45; Found: C, 74.70; H, 4.30.

**4-(4-Methoxyphenyl)-2-phenylfuran (5g).** White microcrystals from ethyl acetate / hexanes (18%), mp 122–123 °C; $^1$H NMR δ 7.72–7.67 (m, 3H), 7.47–7.37 (m, 4H), 7.30–7.24 (m, 1H), 6.95–6.91 (m, 3H), 3.83 (s, 3H); $^{13}$C NMR δ 55.3, 104.0, 114.2, 123.8, 125.0, 126.9, 127.5, 128.0, 128.7, 130.7, 137.1, 154.7, 158.8. Anal. Calcd for $C_{17}H_{14}O_2$: C, 81.58; H, 5.64; Found: C, 81.55; H, 5.37.

**4-(4-Nitrophenyl)-2-phenylfuran (5h).** Red microcrystals from ethyl acetate / hexanes (83%), mp 148–149 °C; $^1$H NMR δ 8.25 (d, *J* = 8.8 Hz, 2H), 7.88 (s, 1H), 7.74–7.65 (m, 4H), 7.46–7.41 (m, 2H), 7.35–7.30 (m, 1H), 6.98 (s, 1H); $^{13}$C NMR δ 103.3, 124.0, 124.3, 126.0, 126.6, 128.1, 128.8, 130.0, 139.0, 139.5, 146.6, 155.8. HRMS Calcd for $C_{16}H_{11}NO_3$ [M]: 265.0739; Found: 265.0740.

**6-(4-Methylphenyl)-4-phenyl-2,3-benzo-1,3a,6a-triazapentalene (7d).** Yellow microcrystals from ethyl acetate / hexanes (71%), mp 142–143 °C; $^1$H NMR δ 8.21 (d, *J* = 8.2 Hz, 2H), 7.74 (d, *J* = 7.3 Hz, 2H), 7.66–7.53 (m, 4H), 7.48–7.43 (m, 1H), 7.39–7.32 (m, 3H), 7.07 (s, 1H), 6.90–6.85 (m, 1H); $^{13}$C NMR δ 21.4, 105.4, 110.8, 113.3, 116.1, 118.5, 119.6, 122.5, 124.7, 125.6, 126.1, 127.5, 128.5, 128.9, 129.1, 129.5, 136.9, 147.7. Anal. Calcd for $C_{22}H_{17}N_3$: C, 81.71; H, 5.30; N, 12.99. Found: C, 81.30; H, 5.32; N, 12.65.

**6-(4-Chlorophenyl)-4-phenyl-2,3-benzo-1,3a,6a-triazapentalene (7e).** Yellow microcrystals from ethyl acetate / hexane (8%), mp 172–173 °C; $^1$H NMR δ 8.27 (d, *J* = 8.6 Hz, 2H), 7.75–7.72 (m, 2H), 7.66–7.55 (m, 4H), 7.50–7.46 (m, 3H), 7.38 (t, *J* = 7.8 Hz, 1H), 7.07 (s, 1H), 6.94–6.89 (m, 1H); $^{13}$C NMR δ 105.5, 110.9, 113.6, 116.8, 117.1, 119.6, 122.6, 125.8, 126.3, 126.9, 127.5, 128.7, 129.0, 129.2, 132.3, 147.5. Anal. Calcd for $C_{21}H_{14}ClN_3$: C, 73.36; H, 4.10; N, 12.22. Found: C, 72.97; H, 3.96; N, 12.00.

**Crystal data for 7e.** $C_{21}H_{14}N_3Cl$, FW 343.80, triclinic, space group P-1, *a* = 6.9146(6), *b* = 8.9169(8), *c* = 14.3617(13) Å, *α* = 88.783(1), *β* = 81.345(1), *γ* = 71.669(1)º, V = 830.7(1) Å$^3$,

F(000) = 356, Z = 2, T = –95 ºC, μ (MoKα) = 0.238 $mm^{-1}$, $D_{calcd}$ = 1.375 $g.cm^{-3}$, crystal size 0.75 x 0.23 x 0.18 mm, $2\theta_{max}$ 53º (CCD area detector, MoKα radiation, 98.4% completeness), GOF = 1.05, $wR(F^2)$ = 0.0971 (all 3354 data), R = 0.0365 (2990 data with I > 2σI).

**4-Phenyl-6-(2-thienyl)-2,3-benzo-1,3a,6a-triazapentalene (7f).** Brown microcrystals from methanol / dichloromethane (42%), mp 140–141 °C; $^1$H NMR δ 7.89 (dd, *J* = 3.7, 0.9 Hz, 1H), 7.72–7.69 (m, 2H), 7.63 (t, *J* = 7.5 Hz, 2H), 7.53 (t, *J* = 7.5 Hz, 2H), 7.46–7.41 (m, 1H), 7.38–7.31 (m, 2H), 7.17 (dd, *J* = 4.9, 3.7 Hz, 1H), 6.95 (s, 1H), 6.88 (t, *J* = 7.9 Hz, 1H); $^{13}$C NMR δ 105.0, 110.8, 113.7, 114.1, 116.6, 120.0, 122.5, 123.4, 123.6, 126.1, 127.4, 127.8, 128.6, 128.6, 129.1, 130.1, 147.7. Anal. Calcd for $C_{19}H_{13}N_3S$: C, 72.36; H, 4.15; N, 13.32. Found: C, 72.32; H, 3.99; N, 13.19.

**6-(4-Methoxyphenyl)-4-phenyl-2,3-benzo-1,3a,6a-triazapentalene (7g).** Yellow prisms from methanol / dichloromethane (42%), mp 182–183 °C; $^1$H NMR δ 8.24–8.21 (m, 2H), 7.72–7.69 (m, 2H), 7.62–7.50 (m, 4H), 7.45–7.41 (m, 1H), 7.36–7.31 (m, 1H), 7.05–7.02 (m, 2H), 6.97 (s, 1H), 6.86–6.81 (m, 1H); $^{13}$C NMR δ 55.3, 105.0, 110.7, 113.1, 114.2, 115.9, 118.3, 119.6, 121.2, 122.4, 126.0, 126.2, 127.4, 128.4, 128.9, 129.0, 147.7, 158.7. Anal. Calcd for $C_{22}H_{17}N_3O$: C, 77.86; H, 5.05; N, 12.38. Found: C, 77.78; H, 4.97; N, 12.31.

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**Supplementary Information Available.** Crystallographic data for compound **7e**. This material is available free of charge online. See page 219.

## References

1. (a) Garst, M. E.; Spencer, T. A. *J. Am. Chem. Soc.* **1973**, *95*, 250. (b) Desimoni, G.; Tacconi, G.; Barco, A.; Pollini, G. P. *Natural Products Synthesis Through Pericyclic Reactions*; American Chemical Society: Washington, 1983. (c) Maurer, B.; Hauser, A. *Tetrahedron Lett.* **1984**, *25*, 1061. (d) Donnelly, D. M. X.; Meegan, M. J. In *Comprehensive Heterocyclic Chemistry*; Katritzky, A. R.; Rees, C. W., Eds; Pergamon: Oxford, 1984; Vol. 4, p 705. (e) Zoretic, P. A.; Shen, Z.; Wang, M.; Ribeiro, A. A. *Tetrahedron Lett.* **1995**, *36*, 2925. (f) Zoretic, P. A.; Wang, M.; Zhang, Y; Shen, Z.; Ribeiro, A. A. *J. Org. Chem.* **1996**, *61*, 1806. (g) Francesconi, I.; Wilson, W. D.; Tanious, F. A.; Hall, J. E.; Bender, B. C.; Tidwell, R. R.; McCurdy, D.; Boykin, D. W. *J. Med. Chem.* **1999**, *42*, 2260.
2. (a) Bosshard, P.; Eugster, C. H. *Adv. Heterocycl. Chem.* **1966**, *7*, 377. (b) Barton, D.; Ollis, W. D. Eds.; *Comprehensive Organic Chemistry*; Pergamon: Oxford, 1979. (c) Dean, F. M. *Adv. Heterocycl. Chem.* **1982**, *30*, 167. (d) Dean, F. M. *Adv. Heterocycl. Chem.* **1982**, *31*,

237. (e) D'Auria, M.; Racioppi, R. *ARKIVOC* **2000**, (*ii)*, 133. (f) Lewkowski, J. *ARKIVOC* **2001**, (*i)*, 17.
3. (a) Cormier, R. A.; Francis, M. D. *Synth. Commun.* **1981**, *11*, 365. (b) Molina, P.; Lorenzo, A.; Fresneda, P. M. *Synthesis* **1983**, 49. (c) Kosugi, M.; Takano, I.; Hoshino, I.; Migita, T. *J. Chem. Soc., Chem. Commun.* **1983**, 989. (d) Cormier, R. A.; Grosshans, C. A.; Skibbe, S. L. *Synth. Commun.* **1988**, *18*, 677. (e) Padmanabhan, S.; Ogawa, T.; Suzuki, H. *Bull. Chem. Soc. Jpn.* **1989**, *62*, 2114. (f) Araki, S.; Butsugan, Y. *Bull. Chem. Soc. Jpn.* **1991**, *64*, 727. (g) Barba, F.; de la Fuente, J. L. *Tetrahedron Lett.* **1992**, *33*, 3911. (h) Kel'in, A. V.; Kulinkovich, O. G. *Zh. Org. Khim.* **1994**, *30*, 197. (i) Zoretic, P. A.; Zhang, Y.; Ribeiro, A. A. *Tetrahedron Lett.* **1995**, *36*, 2929.
4. (a) Harris, C. M.; Cleary, J. J.; Harris, T. M. *J. Org. Chem.* **1974**, *39*, 72. (b) Nishio, T.; Sugawara, M.; Omote, Y. *J. Heterocycl. Chem.* **1979**, *16*, 815. (c) Kurosu, M.; Marcin, L. R.; Grinsteiner, T. J.; Kishi, Y. *J. Am. Chem. Soc.* **1998**, *120*, 6627. (d) Kurosu, M.; Marcin, L. R.; Kishi, Y. *Tetrahedron Lett.* **1998**, *39*, 8929.
5. (a) Ibragimov, I. I.; Mamedov, E. I.; Ismailov, A. T.; Aliev, A. G.; Mekhtieva, Sh. Z.; Dzhafarov, V. G.; Belyaeva, V. I. *Zh. Org. Khim.* **1990**, *26*, 1648. (b) Mamedov, E. I.; Ismailov, A. G.; Kozhushkov, S. I.; Zefirov, N. S. *Chem. Heterocycl. Compd. (Engl. Transl.)* **1992**, 260.
6. (a) Srikrishna, A.; Pullaiah, K. C. *Tetrahedron Lett.* **1987**, *28*, 5203. (b) Srikrishna, A.; Viswajanani, R.; Yelamaggad, C. V. *Tetrahedron* **1997**, *53*, 10479.
7. (a) Wakabayashi, Y.; Fukuda, Y.; Shiragami, H.; Utimoto, K.; Nozaki, H. *Tetrahedron* **1985**, *41*, 3655. (b) Gabriele, B.; Salerno, G.; Lauria, E. *J. Org. Chem.* **1999**, *64*, 7687.
8. (a) Marshall, J. A.; Robinson, E. D. *J. Org. Chem.* **1990**, *55*, 3450. (b) Marshall, J. A.; Wang, X.-J. *J. Org. Chem.* **1991**, *56*, 960. (c) Hashmi, A. S. K.; Schwarz, L.; Choi, J.-H.; Frost, T. M. *Angew. Chem., Int. Ed.* **2000**, *39*, 2285.
9. MaGee, D. I.; Leach, J. D.; Setiadji, S. *Tetrahedron* **1999**, *55*, 2847.
10. (a) Ratier, M.; Drouillard, S.; Trouve, B.; Duboudin, J. G. *Synth. Commun.* **1986**, *16*, 1509. (b) Kataoka, Y.; Tezuka, M.; Takai, K.; Utimoto, K. *Tetrahedron* **1992**, *48*, 3495. (c) Forgione, P.; Wilson, P. D.; Fallis, A. G. *Tetrahedron Lett.* **2000**, *41*, 17.
11. Balaban, T. S.; Hiegemann, M. *Tetrahedron* **1992**, *48*, 9827.
12. (a) Katritzky, A. R.; Lan, X.; Yang, J. Z.; Denisko, O. V. *Chem. Rev.* **1998**, *98*, 409. (b) Katritzky, A. R.; Cui, X.; Long, Q.; Mehta, S.; Steel, P. J. *ARKIVOC* **2000**, (*iv)*, 471. (c) Katritzky, A. R.; Ji, Y.; Tymoshenko, D. O.; Fang, Y.; Hylton, K.-G. *ARKIVOC* **2001**, (*iv)*, 20. (d) Katritzky, A. R.; Silina, A.; Tymoshenko, D. O.; Qiu, G.; Nair, S. K.; Steel, P. J. *ARKIVOC* **2001**, (*vii)*, 138.
13. (a) Bermand, C.; Comel, A.; Kirsch, G. *ARKIVOC* **2000**, (*ii)*, 128. (b) Katritzky, A. R.; Kirichenko, K.; Ji, Y.; Steel, P. J.; Karelson, M. *ARKIVOC* **2003**, (*vi)*, 49.
14. (a) Padwa, A.; Kassir, J. M.; Xu, S. L. *J. Org. Chem.* **1997**, *62*, 1642. (b) Francesconi, I.; Patel, A.; Boykin, D. W. *Synthesis* **1999**, 61.
15. Kim, T.; Kim, K.; Park, Y. J. *Eur. J. Org. Chem.* **2002**, 493.

16. (a) Lynch, B. M.; Hung, Y.-Y. *J. Heterocycl. Chem.* **1965**, *2*, 218. (b) McRobbie, I. M.; Meth-Cohn, O.; Suschitzky, H. *Tetrahedron Lett.* **1976**, *17*, 925. (c) Albini, A.; Bettinetti, G. F.; Minoli, G. *J. Org. Chem.* **1983**, *48*, 1080.
17. (a) Albini, A.; Bettinetti, G.; Minoli, G. *J. Am. Chem. Soc.* **1991**, *113*, 6928. (b) Clark, B. A. J.; McNab, H.; Sommerville, C. C. *Chem. Commun.* **1996**, 1211.
18. (a) Verkruijsse, H. D.; Heus-Kloos, Y. A.; Brandsma, L. *J. Organomet. Chem.* **1988**, *338*, 289. (b) Chowdhury, C.; Kundu, N. G. *Tetrahedron* **1999**, *55*, 7011.
19. Katritzky, A. R.; Lang, H. *J. Org. Chem.* **1995**, *60*, 7612.
20. Barbee, T. R.; Albizati, K. F. *J. Org. Chem.* **1991**, *56*, 6764.