

# A Novel Route to Imidoylbenzotriazoles and Their Application for the Synthesis of Enaminones

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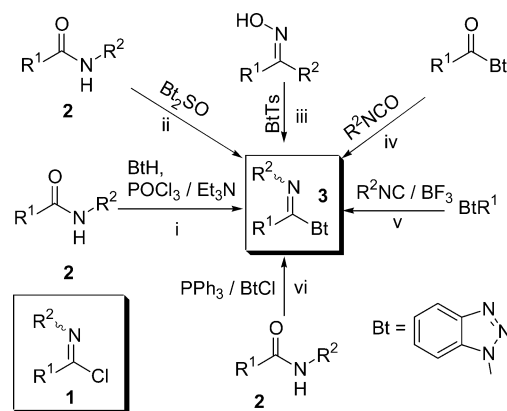
**Abstract:** Reactions of secondary amides **2a–i** with 1-chloro-1*H*-benzotriazole and triphenylphosphine give imidoylbenzotriazoles **3a–i**. The treatment of **3a,b,e,g** with silyl enol ethers **5a,b** in the presence of potassium *tert*-butoxide provides a new general approach to enaminoketones **6a–h**.

Imidoyl chlorides **1** (Scheme 1) are important intermediates and precursors for the synthesis of numerous functionalities, including amidines,<sup>2</sup> imidates,<sup>2a,3</sup> amidoximes,<sup>4</sup> amidrazones,<sup>4c,d</sup> imidonitriles,<sup>5</sup> thioamides,<sup>6</sup> diacylamines,<sup>3b</sup> imines,<sup>7</sup> and heterocyclic compounds.<sup>4c,d,5a</sup>

Imidoylbenzotriazoles **3** have become important as stable alternatives to the corresponding imidoyl chlorides **1**.<sup>8</sup> Major synthetic strategies utilized for the preparation of imidoylbenzotriazoles **3** include the following (Scheme 1): (i) the reaction of secondary amides **2** with benzotriazole and POCl<sub>3</sub> in the presence of triethylamine;<sup>8a</sup> (ii) the reaction of secondary amides **2** and in situ prepared 1,1'-sulfinyldibenzotriazole;<sup>8b</sup> (iii) the one-pot reaction of oximes with 1-(*p*-toluenesulfonyl)benzotriazole;<sup>9</sup> (iv) the condensation of isocyanates with 1-acyl-benzotriazoles;<sup>10</sup> (v) the reaction of isonitriles with *N*-(aminoalkyl)benzotriazoles in the presence of boron trifluoride etherate.<sup>8d</sup>

The enaminone class of compounds<sup>11</sup> represents versatile and useful building blocks for the synthesis of

SCHEME 1



heterocyclic compounds, such as 1,5-benzodiazepine,<sup>12</sup> 1,4-dihydropyridine,<sup>13</sup> furoisoquinoline,<sup>14</sup> indole,<sup>15</sup> isoxazole,<sup>16</sup> pyrrolo-1,2,4-triazine,<sup>17</sup> pyrimidine,<sup>18</sup> pyridinone,<sup>19</sup> and quinoline,<sup>19b,20</sup> derivatives.

The available methods for the preparation of enaminones can be classified according to the bond formed (Scheme 2). Five approaches form bond **a**: (i) reactions of 1,3-diketones with amines in the presence of catalysts;<sup>15,21</sup> (ii) addition of amines to acylacetylenes;<sup>15,20a,22</sup> (iii) palladium-assisted amination of  $\alpha$ -keto olefines<sup>15,23</sup> or amination with *O*-methylhydroxylamines in the presence of base;<sup>24</sup> (iv) additions of amines to 3-oxo-2,3-dihydrothiophene 1,1-dioxides with extrusion of sulfur dioxide;<sup>25</sup> and (v) substitution of a  $\beta$ -functional group, such as alkylthio,<sup>26</sup> imidazolyl,<sup>27</sup> and methoxy.<sup>28</sup> Oxidative cleavage of pyridinium perchlorates with hydrogen

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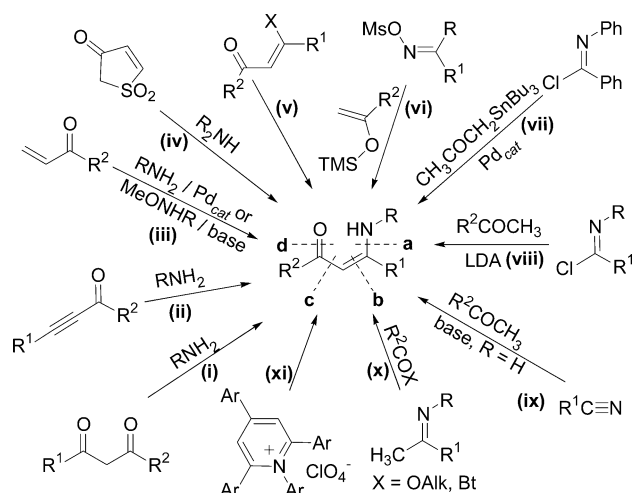
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SCHEME 2



peroxide (route xi)<sup>29</sup> provides enaminones via bond **d**. However, the formation of bonds **a** and **d** shows limited convergence as the “enaminone carbon skeleton” is part of a starting material and this limits the selective construction of unsymmetrical enaminones. Therefore the preparation of enaminones via formation of **b** or **c** carbon–carbon bond is frequently preferable. For the preparation via bond **b**, the following approaches are available: (vi) coupling of silyl enol ethers with oxime sulfonates;<sup>30</sup> (vii) reaction of imidoyl chlorides with acetonyltributyltin in the presence of palladium catalyst,<sup>31</sup> (viii) similar reaction with enolates of ketones;<sup>32</sup> and (ix) reactions of nitriles with ketones in the presence of base.<sup>33</sup> Existing methods (vi–ix) for construction of bond **b** show some limitations: (a) the oxime route (vi) involves a C → N migration and is limited to symmetric oximes;<sup>30</sup> (b) for imidoyl chlorides (routes vii–viii), a low yield and formation of byproduct in a reaction with acetonyltributyltin (single example)<sup>31</sup> was reported and the reaction with enolates was described only for fluorinated imidoyl chlorides;<sup>32</sup> (c) reactions of nitriles with ketones (route ix) were described only for nitriles of aromatic<sup>33a</sup> and fluorinated<sup>33b</sup> acids. The only approach presently available for the synthesis of diverse enaminones in good yields is route (x): the reaction of ketimine anions with esters<sup>16,32,34</sup> or acylbenzotriazoles,<sup>35</sup> in which bond **c** is formed.

We now disclose the one-step preparation of imidoylbenzotriazoles **3a–i** by reactions of secondary amides **2a–i** with 1-chloro-1*H*-benzotriazole in the presence of triphenylphosphine (Scheme 1, route vi, Table 1). This

new approach makes a variety of imidoylbenzotriazoles readily available by a simple procedure in good yields. We also report the synthesis of enaminones **6a–h** via substitution of the benzotriazolyl group in imidoylbenzotriazoles **3a,b,e,g** with silyl enol ethers **5a,b** (Scheme 3, Table 2): this represents a convergent synthesis of enaminones in which bond **b** is formed.

**Preparation of Imidoylbenzotriazoles (3).** Secondary amides **2a–i** were treated with a mixture of 1-chloro-1*H*-benzotriazole and triphenylphosphine in THF under reflux to give the corresponding imidoylbenzotriazoles **3a–i** in good yields (Scheme 1, Table 1). Structures **3a–i** are supported by their <sup>1</sup>H NMR and <sup>13</sup>C NMR spectra. The <sup>1</sup>H NMR spectra of **3a–i** showed the appearance of a new set of signals at 7.3–8.6 ppm, characteristic of the benzotriazolyl group.<sup>8,9,10</sup> The <sup>13</sup>C NMR spectra of **3a–i** also showed the disappearance of the amide signals and the appearance of the new signals around 115, 120, 124, 127, 131, and 146 ppm, corresponding to the *N*-substituted benzotriazoles, and at 153–155 ppm, corresponding to the imine carbon.<sup>8,9,10</sup>

**Preparation of Enaminoketones (6) from Imidoylbenzotriazoles (3).** Imidoylbenzotriazoles **3a,b** were reacted with enolates generated from corresponding ketones **4a,b** by treatment with base. Previously investigated reactions of imidoylbenzotriazoles with 2-methyl-2-oxazoline and 2-methyl-2-thiazoline in the presence of LDA or *n*-BuLi gave products of substitution in high yields and demonstrated the stability of imidoylbenzotriazoles in the presence of strong bases at low temperatures.<sup>8e</sup> Reactions of compounds **3a,b** with ketones **4a,b** in the presence of LDA (2.0–2.5 equiv) at temperatures ranging from –78 to 0 °C resulted in condensation of the ketones and recovery of the imidoylbenzotriazoles **3a,b**. Treatment of **3b** with ketones **4a,b** (2 equiv) in the presence of potassium *tert*-butoxide (2.5–3.0 equiv) in THF or diethyl ether at temperatures from 0 to 40 °C (reflux in diethyl ether) gave enaminoketones **6c,d** in low yields (20–30%). The major byproduct of these reactions was the corresponding amide **2b**. The formation of **2b** is ascribed to the hydrolysis of **3b** in the presence of a strong base. Water is formed from the aldol condensation of ketones, resulting in the hydrolysis of **3b**. Generally, higher rates of these reactions were observed at higher temperatures with similar selectivity.

On the other hand, treatment of **3a,b,e,g** with silyl enol ethers **5a,b** (2–2.5 equiv) in the presence of potassium *tert*-butoxide (2.5–3.0 equiv) in THF at temperatures from 20 to 40 °C or in diethyl ether at 20–35 °C gave enaminoketones **6a–h** in good yields (Scheme 3, Table 2). The major byproducts observed in these reactions were the corresponding amides **2a,b,e,g**.

Structures **6a–h** were supported by their <sup>1</sup>H and <sup>13</sup>C NMR spectra. Their <sup>1</sup>H NMR spectra no longer showed distinctive signals in the range 7.0–8.2 ppm corresponding to the benzotriazolyl group in **3a,b,e,g**.<sup>8,9,10</sup> The singlets at 12.9–13.10 ppm and at 13.74–14.18 ppm in the <sup>1</sup>H NMR spectra of **6a,c,e,g** and **6b,d,f,h**, respectively, were assigned to the NH proton. The singlets at 5.81–6.08 ppm in the spectra of **6a,c,e,g** were assigned to olefinic proton. For **6b,d,f,h**, the new set of signals that

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TABLE 1. Preparative Yields of Imidoylbenzotriazoles 3a–i: Comparison with Previous Methods

|    | R <sup>1</sup>                                                                    | R <sup>2</sup>                          | BtCl/PPh <sub>3</sub> | POCl <sub>3</sub> | BtSOBt          | Oximes/BtTs      | ArNCO           |
|----|-----------------------------------------------------------------------------------|-----------------------------------------|-----------------------|-------------------|-----------------|------------------|-----------------|
|    |                                                                                   |                                         | vi <sup>a</sup>       | i <sup>a</sup>    | ii <sup>a</sup> | iii <sup>a</sup> | iv <sup>a</sup> |
| 3a | Ph                                                                                | Ph                                      | 90                    |                   | 41              | 70               | 71              |
| 3b | 4-Me-C <sub>6</sub> H <sub>4</sub>                                                | 4-MeO-C <sub>6</sub> H <sub>4</sub>     | 75                    |                   | 40              |                  |                 |
| 3c | Ph                                                                                | 4-Cl-C <sub>6</sub> H <sub>4</sub>      | 80                    |                   |                 |                  |                 |
| 3d | Ph                                                                                | 2-Me-5-Cl-C <sub>6</sub> H <sub>3</sub> | 80                    |                   |                 |                  |                 |
| 3e | Bn                                                                                | 4-Me-C <sub>6</sub> H <sub>4</sub>      | 83                    |                   |                 |                  |                 |
| 3f | Me                                                                                | Et                                      | 40                    |                   |                 | 45               |                 |
| 3g | Me                                                                                | Ph                                      | 90                    | 96                | 49              | 60               |                 |
| 3h | Me                                                                                | 4-MeO-C <sub>6</sub> H <sub>4</sub>     | 81                    |                   |                 |                  |                 |
|    | Me                                                                                | 4-EtO-C <sub>6</sub> H <sub>4</sub>     |                       |                   | 25              |                  |                 |
| 3i |  | Ph                                      | 82                    |                   |                 |                  |                 |

<sup>a</sup> Corresponding methods for the preparation in Scheme 1.

SCHEME 3

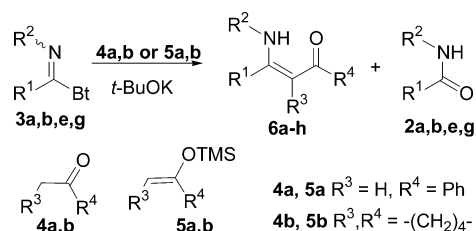


TABLE 2. Preparation of Enaminoketones 6a–h

|       | R <sup>1</sup>                     | R <sup>2</sup>                      | R <sup>3</sup>                     | R <sup>4</sup> | yield of <b>6</b> , % |
|-------|------------------------------------|-------------------------------------|------------------------------------|----------------|-----------------------|
| 3a 6a | Ph                                 | Ph                                  | H                                  | Ph             | 80                    |
| 3a 6b | Ph                                 | Ph                                  | -(CH <sub>2</sub> ) <sub>4</sub> - |                | 54                    |
| 3b 6c | 4-Me-C <sub>6</sub> H <sub>4</sub> | 4-MeO-C <sub>6</sub> H <sub>4</sub> | H                                  | Ph             | 58                    |
| 3b 6d | 4-Me-C <sub>6</sub> H <sub>4</sub> | 4-MeO-C <sub>6</sub> H <sub>4</sub> | -(CH <sub>2</sub> ) <sub>4</sub> - |                | 65                    |
| 3e 6e | Bn                                 | 4-Me-C <sub>6</sub> H <sub>4</sub>  | H                                  | Ph             | 55                    |
| 3e 6f | Bn                                 | 4-Me-C <sub>6</sub> H <sub>4</sub>  | -(CH <sub>2</sub> ) <sub>4</sub> - |                | 36                    |
| 3g 6g | Me                                 | Ph                                  | H                                  | Ph             | 40                    |
| 3g 6h | Me                                 | Ph                                  | -(CH <sub>2</sub> ) <sub>4</sub> - |                | 19                    |

appeared in their <sup>1</sup>H NMR spectra in the ranges 1.53–1.82 ppm, 2.11–2.28 ppm, and 2.38–2.51 ppm were assigned to aliphatic protons of the cyclohexanone ring. The <sup>13</sup>C NMR spectra of **6a,c,e,g** showed new signals at 188.6–189.6 ppm as well as at 94.2–97.0 ppm and 161.4–164.6 ppm, corresponding to carbonyl carbon and the vinyl fragment of **6a,c,e,g**, respectively. Similarly, the <sup>13</sup>C NMR spectra of **6b,d,f,h** showed new signals at 196.0–199.0 ppm and at 159.5–161.2 ppm, corresponding to carbonyl carbon and the vinyl carbon next to amine, respectively. Unlike **3a,c,e,g**, the <sup>13</sup>C NMR spectra of **6a–h** no longer showed either the characteristic benzotriazole signals around 115, 120, and 146 ppm or the imine carbon signal at 153–155 ppm.

An efficient and simple route to imidoylbenzotriazoles has been developed using secondary amides and 1-chloro-1*H*-benzotriazole. The procedure uses no aggressive reagents and occurs under mild reaction conditions. This method provides easy access to imidoylbenzotriazoles **3a–i** in good yields and complements previous preparations of imidoylbenzotriazoles. The reaction of imidoylbenzotriazoles with silyl enolates opens a new way for the synthesis of β-enaminoketones; it is especially useful as a synthetic method for the preparation of

unsymmetrical β-enaminoketones, unavailable by other methods.

## Experimental Section

**General Procedure for the Preparation of Imidoylbenzotriazoles 3a–i.** To a stirred solution of triphenylphosphine (524 mg, 2 mmol) in THF (20 mL) was added 1-chloro-1*H*-benzotriazole (307 mg, 2 mmol) at room temperature, and the reaction mixture was stirred for 1 h. The corresponding secondary amide **2a–i** (1 mmol) was added to the reaction mixture which was then heated under reflux for 20 h. Progress of the reaction was monitored by TLC; the addition of extra triphenylphosphine and 1-chloro-1*H*-benzotriazole may be required. After the starting amide **2a–i** had been consumed, the solvent was evaporated under reduced pressure, and the residue was purified by flash column chromatography on silica gel (for **3f**, on alumina) using hexanes/ethyl acetate gradients to give **3a–i**.

**N-[Benzotriazol-1-yl(phenyl)methylidene]aniline (3a):** yellow microcrystals from ethyl acetate/hexanes (90%); mp 130–131 °C (lit.<sup>8b</sup> 130–132 °C); <sup>1</sup>H NMR δ 8.48 (d, *J* = 8.1 Hz, 1H), 8.14 (d, *J* = 8.1 Hz, 1H), 7.65–7.59 (m, 1H), 7.53–7.48 (m, 1H), 7.42–7.35 (m, 5H), 7.25–7.20 (m, 2H), 7.06–7.01 (m, 1H), 6.85 (d, *J* = 7.8 Hz, 2H); <sup>13</sup>C NMR δ 115.3, 120.0, 121.5, 124.2, 125.6, 128.2, 128.8, 129.3, 130.1, 130.2, 130.4, 132.1, 146.4, 147.0, 153.8.

**N-[Benzotriazol-1-yl(4-methylphenyl)methylidene]-4-methoxyaniline (3b):** yellow plates from methylene chloride (75%); mp 71–73 °C (lit.<sup>8e</sup> mp 56–58 °C); <sup>1</sup>H NMR δ 8.43 (d, *J* = 8.2 Hz, 1H), 8.14 (d, *J* = 8.2 Hz, 1H), 7.64–7.46 (m, 2H), 7.29–7.17 (m, 5H), 6.83–6.76 (m, 4H), 3.77 (s, 3H), 2.38 (s, 3H); <sup>13</sup>C NMR δ 21.6, 55.4, 114.1, 115.3, 119.9, 122.4, 123.0, 125.3, 127.6, 129.0, 129.0, 130.1, 132.1, 140.1, 140.6, 146.3, 153.2, 156.5. Anal. Calcd for C<sub>21</sub>H<sub>18</sub>N<sub>4</sub>O: C, 73.67; H, 5.30; N, 16.36. Found: C, 73.55; H, 5.29; N, 16.67.

**N-[1-(Benzotriazol-1-yl)-2-phenylethylidene]-4-methylaniline (3c):** white microcrystals from methylene chloride (83%); mp 123–125 °C; <sup>1</sup>H NMR δ 8.52 (d, *J* = 8.2 Hz, 1H), 8.08 (d, *J* = 8.2 Hz, 1H), 7.60–7.55 (m, 1H), 7.48–7.42 (m, 1H), 7.22–7.13 (m, 7H), 6.87 (d, *J* = 8.2 Hz, 2H), 4.61 (s, 2H), 2.38 (s, 3H); <sup>13</sup>C NMR δ 20.9, 34.7, 115.6, 119.7, 120.1, 125.4, 126.8, 128.6, 128.7, 129.2, 129.8, 131.5, 134.0, 135.3, 144.4, 146.6, 154.6. Anal. Calcd for C<sub>21</sub>H<sub>18</sub>N<sub>4</sub>: C, 77.28; H, 5.56; N, 17.17. Found: C, 77.39; H, 5.55; N, 17.44.

**N-[1-(Benzotriazol-1-yl)ethylidene]aniline (3g):** white microcrystals from ethyl acetate/hexanes (90%); mp 106–107 °C (lit.<sup>9</sup> mp 107–108 °C); <sup>1</sup>H NMR δ 8.54 (d, *J* = 8.4 Hz, 1H), 8.13 (d, *J* = 8.4 Hz, 1H), 7.62–7.57 (m, 1H), 7.50–7.40 (m, 3H), 7.22–7.17 (m, 1H), 6.95 (d, *J* = 7.4 Hz, 2H), 2.75 (s, 3H); <sup>13</sup>C NMR δ 16.1, 115.6, 119.6, 120.1, 124.2, 125.2, 129.1, 129.1, 131.1, 146.5, 147.2, 153.8.

**General Procedure for the Preparation of Enaminoketones 6a–h.** To a stirred solution of the corresponding imidoylbenzotriazole **3a,b,e,g** (1 mmol) in THF or diethyl ether (20 mL), the corresponding silyl enol ether **5a,b** (2–2.5 mmol) and then potassium *tert*-butoxide (0.28–0.34 g, 2.5–3.0 mmol) were added at room temperature. The reaction mixture was stirred for 12–40 h either at room temperature (if THF was used as solvent) or while refluxing (if diethyl ether was used as solvent). Progress of the reaction was monitored by TLC. After the reaction was complete, water (40 mL) was added to the reaction mixture, which was then extracted with diethyl ether (5 × 25 mL). The combined extracts were dried over anhydrous magnesium sulfate. The solvent was removed under reduced pressure and the remaining residue was purified by gradient column chromatography on silica gel (hexanes–ethyl acetate/hexanes 1:15) to give **6a–h**.

**3-Anilino-1,3-diphenyl-2-propen-1-one (6a):** yellow-orange microcrystals from methylene chloride (80%); mp 95–97 °C (lit.<sup>26</sup> mp 101–102 °C); <sup>1</sup>H NMR δ 12.90 (s, 1H), 7.96 (d, *J* = 6.9 Hz, 2H), 7.50–7.26 (m, 8H), 7.14–7.08 (m, 2H), 7.00–6.94 (m, 1H), 6.78 (d, *J* = 8.0 Hz, 2H), 6.08 (s, 1H); <sup>13</sup>C NMR δ 97.0, 123.1, 124.1, 127.2, 128.3, 128.5, 128.7, 129.6, 131.3, 135.8, 139.4, 139.8, 161.4, 189.6. Anal. Calcd for C<sub>21</sub>H<sub>17</sub>NO: C, 84.25; H, 5.72; N, 4.68. Found: C, 84.50; H, 5.86; N, 4.30.

**2-[Anilino(phenyl)methylidene]cyclohexanone (6b):** yellow microcrystals from methylene chloride (54%); mp 125–127 °C (lit.<sup>36</sup> mp 135–136 °C); <sup>1</sup>H NMR δ 13.74 (s, 1H), 7.35–7.30 (m, 3H), 7.20–7.16 (m, 2H), 7.06–6.97 (m, 2H), 6.91–6.85 (m, 1H), 6.61 (d, *J* = 8.0 Hz, 2H), 2.51–2.44 (m, 2H), 2.13 (t, *J* = 6.3 Hz, 2H), 1.82–1.71 (m, 2H), 1.63–1.53 (m, 2H); <sup>13</sup>C NMR δ 22.6, 23.9, 27.4, 38.5, 103.7, 122.9, 123.5, 128.4, 128.5, 128.6, 134.2, 139.7, 142.2, 159.5, 199.0. Anal. Calcd for C<sub>19</sub>H<sub>19</sub>NO: C, 82.28; H, 6.90; N, 5.05. Found: C, 81.91; H, 7.24; N, 4.81.

**3-(4-Methoxyanilino)-3-(4-methylphenyl)-1-phenyl-2-propen-1-one (6c):** yellow-orange microcrystals from methylene chloride (58%); mp 126–128 °C; <sup>1</sup>H NMR δ 12.90 (s, 1H), 7.99–7.91 (m, 2H), 7.50–7.37 (m, 3H), 7.26 (d, *J* = 8.0 Hz, 2H), 7.11 (d, *J* = 7.7 Hz, 2H), 6.76 (d, *J* = 8.8 Hz, 2H), 6.68 (d, *J* = 9.1 Hz, 2H), 6.03 (s, 1H), 3.71 (s, 3H), 2.34 (s, 3H); <sup>13</sup>C NMR δ 21.3, 55.3, 95.9, 113.9, 124.8, 127.1, 128.3, 128.4, 129.1, 131.0, 132.6, 132.8, 139.7, 140.1, 156.4, 162.2, 189.1. Anal. Calcd for C<sub>23</sub>H<sub>21</sub>NO<sub>2</sub>: C, 80.44; H, 6.16; N, 4.08. Found: C, 80.53; H, 6.42; N, 3.98.

**2-[(4-Methoxyanilino)(4-methylphenyl)methylidene]cyclohexanone (6d):** yellow-orange microcrystals from methylene chloride (28%); mp 119–121 °C; <sup>1</sup>H NMR δ 13.78 (s, 1H), 7.12 (d, *J* = 7.8 Hz, 2H), 7.03 (d, *J* = 7.8 Hz, 2H), 6.62–6.55 (m, 4H) 3.68 (s, 3H), 2.47 (t, *J* = 6.7 Hz, 2H), 2.33 (s, 3H), 2.11 (t, *J* = 6.2 Hz, 2H), 1.79–1.70 (m, 2H), 1.60–1.54 (m, 2H); <sup>13</sup>C NMR δ 21.3, 22.7, 24.0, 27.4, 38.4, 55.2, 102.9, 113.7, 124.8, 128.5, 129.1, 131.3, 132.9, 138.3, 156.0, 160.8, 198.1. Anal. Calcd for C<sub>21</sub>H<sub>23</sub>NO<sub>2</sub>: C, 78.47; H, 7.21; N, 4.36. Found: C, 78.00; H, 7.52; N, 3.98.

**1,4-Diphenyl-3-(4-toluidino)-2-buten-1-one (6e):** yellow microcrystals from diethyl ether (55%); mp 87–89 °C; <sup>1</sup>H NMR δ 13.05 (s, 1H), 7.85–7.82 (m, 2H), 7.44–7.35 (m, 3H), 7.28–7.20 (m, 3H), 7.14–7.08 (m, 4H), 6.98 (d, *J* = 8.2 Hz, 2H), 5.81 (s, 1H), 3.72 (s, 2H), 2.32 (s, 3H); <sup>13</sup>C NMR δ 20.9, 38.5, 94.3, 125.5, 126.7, 127.0, 128.2, 128.5, 128.8, 129.6, 130.8, 135.5, 136.0, 136.7, 140.0, 164.6, 188.8. Anal. Calcd for C<sub>23</sub>H<sub>21</sub>NO: C, 84.37; H, 6.46; N, 4.28. Found: C, 84.49; H, 6.64; N, 4.28.

**2-[2-Phenyl-1-(4-toluidino)ethylidene]cyclohexanone (6f):** orange oil (36%); <sup>1</sup>H NMR δ 14.17 (s, 1H), 7.31–7.19 (m, 3H), 7.12 (d, *J* = 6.9 Hz, 2H), 7.01 (d, *J* = 8.2 Hz, 2H), 6.88 (d, *J* = 8.2 Hz, 2H), 3.75 (s, 2H), 2.44 (t, *J* = 6.6 Hz, 2H), 2.28–2.21 (m, 5H), 1.75–1.67 (m, 2H), 1.65–1.57 (m, 2H); <sup>13</sup>C NMR δ 20.8, 22.7, 23.8, 25.7, 34.2, 38.1, 102.9, 125.0, 126.4, 127.7, 128.7, 129.6, 135.5, 136.0, 136.6, 161.5, 196.8. Anal. Calcd for C<sub>21</sub>H<sub>23</sub>NO: C, 82.58; H, 7.59; N, 4.59. Found: C, 82.21; H, 7.95; N, 4.25.

**3-Anilino-1-phenyl-2-buten-1-one (6g):** orange microcrystals from diethyl ether (42%); mp 104–106 °C (lit.<sup>26</sup> mp 105–106 °C); <sup>1</sup>H NMR δ 13.10 (br s, 1H), 7.93–7.90 (m, 2H), 7.46–7.39 (m, 3H), 7.35 (d, *J* = 8.0 Hz, 2H), 7.24–7.16 (m, 3H), 5.89 (s, 1H), 2.14 (s, 3H); <sup>13</sup>C NMR δ 20.4, 94.2, 124.8, 125.7, 127.0, 128.2, 129.1, 130.9, 138.6, 140.0, 162.2, 188.7.

**2-[1-Anilinoethylidene]cyclohexanone (6h):** yellow oil<sup>21b</sup> (19%); <sup>1</sup>H NMR δ 14.06 (s, 1H), 7.37–7.31 (m, 2H), 7.19 (t, *J* = 7.4 Hz, 1H), 7.08 (d, *J* = 7.7 Hz, 2H), 2.42–2.37 (m, 4H), 1.99 (s, 3H), 1.77–1.72 (m, 4H); <sup>13</sup>C NMR δ 16.2, 22.8, 23.9, 26.5, 38.2, 102.8, 125.2, 125.4, 129.0, 139.1, 160.5, 196.0.

**Supporting Information Available:** Characterization data for imidoylbenzotriazoles **3c,d,f,h,i**. This material is available free of charge via the Internet at <http://pubs.acs.org>.

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