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# One-pot Vilsmeier reagent-mediated multicomponent reaction: A direct synthesis of oxazolones and Erlenmeyer azlactones from carboxylic acids

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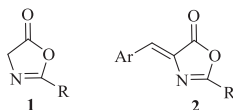
## ABSTRACT

N-Acylation of glycine with carboxylic acids was carried out by using the Vilsmeier reagent. The resulting *N*-acyl- $\alpha$ -amino acids were subsequently cyclodehydrated into oxazol-5-ones via the Vilsmeier reagent. Finally, treatment of oxazol-5-ones with aldehydes in the presence of the Vilsmeier reagent delivered Erlenmeyer azlactones. By combining these steps, using the Vilsmeier reagent allowed direct one-pot conversion of carboxylic acids into oxazol-5-ones and direct one-pot conversion of carboxylic acids into Erlenmeyer azlactones in the presence of aldehydes. These Vilsmeier reagent-mediated multicomponent reactions proceeded smoothly in reasonable chemical yields at room temperature. The chemical structure of the title compounds was confirmed by spectral data.

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## 1. Introduction

Oxazol-5-ones (oxazolones) **1** and 4-arylmethylene-oxazolones (Erlenmeyer azlactones) **2** are versatile organic compounds with applications in different fields of research [1].



Oxazolone derivatives possess important biological activities [1,2]. Some prepared oxazolones showed inhibition of tyrosinase [3] and Human NK1 receptor antagonist [4]. Also, they are used as antioxidant and anticorrosive

additives for lubricant oils [5]. In addition, oxazolones are used as versatile building blocks in synthetic organic chemistry for the synthesis of several heterocycles and biologically active molecules [6].

According to their importance, many efforts made for the synthesis of oxazolones and azlactones have been reported [7]. The general method for the preparation of oxazolones involves the classical cyclization of an *N*-acyl- $\alpha$ -amino acid with acetic anhydride under reflux conditions, where *N*-acyl- $\alpha$ -amino acids are prepared separately from acyl chlorides and glycine [8].

In a pioneering work, Erlenmeyer described the synthesis of azlactone by condensation of benzaldehyde with *N*-acetyl glycine in the presence of acetic anhydride and sodium acetate [9]. To avoid the acidic and harsh conditions using acetic anhydride [10], other dehydrating agents such as carbodiimides [11], ethyl chloroformate [12], and aryl-sulfonyl chloride [13] have been reported.

Some drawbacks in these methods are multistep reactions from carboxylic acid to oxazolones, the need for low or high temperatures, lower yield of the products, harsh

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conditions, unnecessary waste, and chromatographic separations of products.

Chloromethylenedimethylammonium chloride (Vilsmeier reagent) **3** is easily prepared by the reaction of DMF and oxalyl chloride or thionyl chloride in dry dichloromethane [14]. This reagent has been identified as a formylating agent [15], a coupling reagent in the synthesis of esters [16], amides [17], acid chlorides [18],  $\beta$ -sultams [19],  $\beta$ -lactams [20], diacylhydrazines [21], 1,3,4-oxadiazoles [22], and 1,3,4-thiadiazoles [23].

To the best of our knowledge, the Vilsmeier reagent has not been used for the synthesis of oxazolones. Hence, in this article, we discussed the application of the Vilsmeier reagent for the conversion of carboxylic acid to oxazolones and Erlenmeyer azlactones.

## 2. Results and discussion

### 2.1. Synthesis

Hippuric acid **4a** used as a starting material was simply prepared from benzoic acid and glycine in the presence of the Vilsmeier reagent. The prepared hippuric acid **4a** was stirred with the Vilsmeier reagent in the presence of triethylamine in DMF to yield 2-phenyloxazol-5-one **5a** in 66% yield. The progress of the reaction was monitored by thin-layer chromatography (TLC) until all hippuric acid **4a** was consumed in the reaction. The reaction mixture was then quenched with water and ethyl acetate. The organic layer was washed with saturated sodium bicarbonate and dried over  $\text{Na}_2\text{SO}_4$ . After removal of the solvent and recrystallization from ethanol, 2-phenyloxazol-5-one **5a** was obtained as pale yellow crystals.

To optimize the reaction conditions for the synthesis of 2-phenyloxazol-5-one **5a** from hippuric acid **4a**, various solvents and different molar amounts of the Vilsmeier reagent as well as temperature were investigated (Table 1). It was found that the yield of **5a** in chloroform was higher

than in other solvents. In addition, the best temperature for this reaction was room temperature giving highest yield of the desired product. As it is shown in Table 1, an equimolar amount of the Vilsmeier reagent was sufficient for completion of the reaction.

Treatment of *N*-acyl- $\alpha$ -amino acids **4a–l** with the Vilsmeier reagent and triethylamine in chloroform at room temperature gave oxazol-5-ones **5a–l** with good to excellent yield after crystallization from ethanol (Scheme 1, Table 2). The structures of all compounds **5a–l** were confirmed by elemental analysis and spectroscopic methods (IR,  $^1\text{H}$  NMR, and  $^{13}\text{C}$  NMR).

Analysis of the IR spectrum of oxazol-5-ones **5a–l** showed two characteristic signals at about  $1650$  and  $1800\text{ cm}^{-1}$ , which were ascribed to the presence of  $\text{C}=\text{N}$  and  $\text{C}=\text{O}$  bonds, respectively. Of course, the NH, CO amide, OH, and CO carboxylic acid peaks of *N*-acyl- $\alpha$ -amino acids **4a–l** had vanished. Furthermore, the  $^1\text{H}$  NMR and  $^{13}\text{C}$  NMR spectra showed characteristic  $\text{CH}_2$ ,  $\text{C}=\text{N}$ , and CO peaks.

Transformation of oxazol-5-ones and aldehydes to Erlenmeyer azlactones in chloroform has been previously reported [8d]. Then, oxazol-5-one **5a** was converted to Erlenmeyer azlactone **6a** in the presence of benzaldehyde and of the Vilsmeier reagent in 84% yield (Scheme 2).

These results prompted us to develop an easy method for the one-pot synthesis of oxazol-5-ones and azlactones from carboxylic acids. Reaction of benzoic acid (1.0 mmol) with glycine (1.0 mmol) in the presence of the Vilsmeier reagent **3**, triethylamine, and benzaldehyde (1.0 mmol) in dry chloroform at room temperature for 3 h allowed direct conversion into the crude azlactone **6a**. The mixture was washed with saturated  $\text{NaHCO}_3$  solution. Crystallization from 70% ethanol gave pure azlactone **5a** as a yellow solid. In the absence of benzaldehyde, oxazol-5-one **5a** was obtained. Then, using similar procedures, various pure oxazol-5-ones **5a–l** and azlactones **6a–m** were prepared directly from carboxylic acids (Scheme 3, Tables 2 and 3).

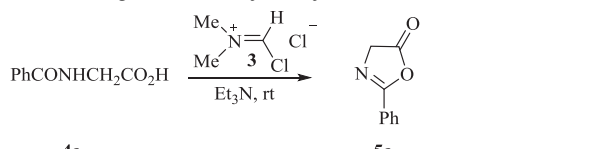
The synthesized compounds were characterized by IR,  $^1\text{H}$  NMR,  $^{13}\text{C}$  NMR, and elemental analysis.

The method was clean and convenient giving moderate to excellent isolated yields. In addition, the byproducts (DMF and salts) could be easily removed from the reaction mixture by aqueous work up.

## 3. Conclusion

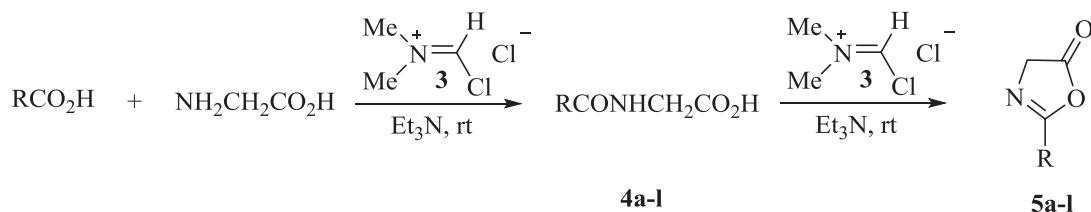
To sum up, we have demonstrated the effectiveness and efficiency of the Vilsmeier reagent for the synthesis of oxazol-5-ones (oxazolones) and 4-arylmethylene-oxazolones (Erlenmeyer azlactones) proceeding via the cyclization of *N*-acyl- $\alpha$ -amino acids. Ready availability, high reactivity, and low cost make the Vilsmeier reagent a versatile and useful reagent for these reactions. In addition, we have developed facile and efficient methods for the synthesis of aliphatic, (hetero)-aromatic oxazol-5-ones, and 4-arylmethyleneoxazolones directly from carboxylic acids in a one-pot procedure. This methodology offers the advantage of mild reaction condition, good to excellent yield of products, and simplicity of work. Pure products were obtained by simple aqueous work up and crystallization.

**Table 1**  
Vilsmeier reagent-mediated cyclodehydration of **4a**.



Entry	Solvent	Temperature	Vilsmeier reagent (mmol)	Yield (%)
1	DMF	rt	1.0	66
2	$\text{CH}_2\text{Cl}_2$	rt	1.0	76
3	Benzene	rt	1.0	34
4	$\text{CH}_3\text{CN}$	rt	1.0	58
5	$\text{CHCl}_3$	rt	1.0	81
6	DMSO	rt	1.0	61
7	$\text{CHCl}_3$	rt	0.8	55
8	$\text{CHCl}_3$	rt	1.3	76
8	$\text{CHCl}_3$	$0\text{ }^\circ\text{C}$	1.0	— <sup>a</sup>
9	$\text{CHCl}_3$	$61\text{ }^\circ\text{C}$	1.0	70

<sup>a</sup> Trace amount of the product was observed on TLC monitoring but it was not isolated.

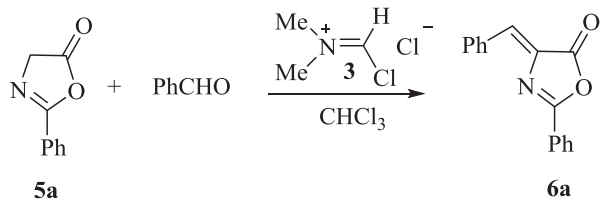


**Scheme 1.** Synthesis of oxazol-5-ones **5a–l** from *N*-acyl- $\alpha$ -amino acids **4a–l**.

**Table 2**

Synthesis of *N*-acyl-glycines **4a–l** and oxazol-5-ones **5a–l** using the Vilsmeier reagent.

Entry	R	RCONHCH <sub>2</sub> CO <sub>2</sub> H	Isolated yield (%)	Oxazol-5-one	Isolated yield (%)	Isolated yield (%) (one-pot synthesis)
1	Ph	<b>4a</b>	85	<b>5a</b>	81	86
2	4-ClC <sub>6</sub> H <sub>4</sub>	<b>4b</b>	83	<b>5b</b>	88	89
3	4-MeOC <sub>6</sub> H <sub>4</sub>	<b>4c</b>	75	<b>5c</b>	78	84
4	4-NO <sub>2</sub> C <sub>6</sub> H <sub>4</sub>	<b>4d</b>	90	<b>5d</b>	86	91
5	4-MeC <sub>6</sub> H <sub>4</sub>	<b>4e</b>	83	<b>5e</b>	82	80
6	C <sub>6</sub> H <sub>5</sub> CH <sub>2</sub>	<b>4f</b>	79	<b>5f</b>	72	78
7	4-(Me <sub>2</sub> N)C <sub>6</sub> H <sub>4</sub>	<b>4g</b>	74	<b>5g</b>	77	73
8	4-Pyridyl	<b>4h</b>	72	<b>5h</b>	70	79
9	2-MeOC <sub>6</sub> H <sub>4</sub>	<b>4i</b>	81	<b>5i</b>	86	88
10	3-ClC <sub>6</sub> H <sub>4</sub>	<b>4j</b>	84	<b>5j</b>	85	90
11	2-Naphthyl	<b>4k</b>	89	<b>5k</b>	87	92
12	CH <sub>3</sub> -CH=CH	<b>4l</b>	74	<b>5l</b>	79	76



**Scheme 2.** Transformation of oxazol-5-ones to Erlenmeyer azlactones.

## 4. Experimental section

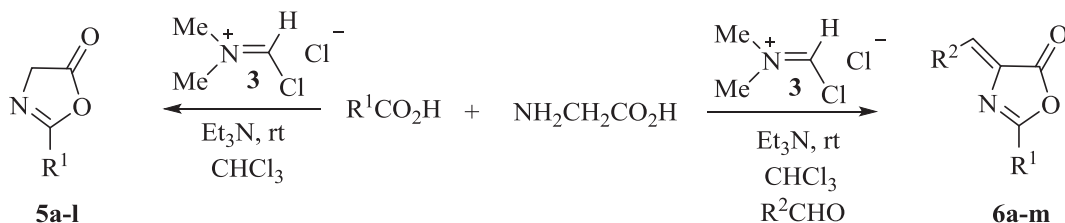
### 4.1. Chemical reagents

All needed chemicals were purchased from Merck, Fluka, and Acros chemical companies. IR spectra were run on a Shimadzu FT-IR 8300 spectrophotometer. <sup>1</sup>H NMR and <sup>13</sup>C NMR spectra were recorded in DMSO-*d*<sub>6</sub> and CDCl<sub>3</sub> using a Bruker Avance DPX instrument (<sup>1</sup>H NMR 250 MHz, <sup>13</sup>C NMR 62.9 MHz). Chemical shifts were reported in ppm (δ)

downfield from tetramethylsilane. All of the coupling constants (*J*) are in Hertz. The mass spectra were recorded on a Shimadzu GC-MS QP 1000 EX instrument. Elemental analyses were run on a Thermo Finnigan Flash EA-1112 series. Melting points were determined in open capillaries with Buchi 510 melting point apparatus. TLC was carried out on silica gel 254 analytical sheets obtained from Fluka.

### 4.2. General procedure for the conversion of *N*-acyl- $\alpha$ -amino acids **4a–l** to oxazol-5-ones **5a–l**

A solution of *N*-acyl- $\alpha$ -amino acids **4a–l** (1.0 mmol), Vilsmeier's reagent **3** (1.0 mmol), and Et<sub>3</sub>N (2.0 mmol) in dry chloroform (15 mL) was stirred at room temperature for 1.5 h. Water (15 mL) was added and the organic layer was separated. The organic layer was washed with brine (20 mL), dried (Na<sub>2</sub>SO<sub>4</sub>), filtered, and the solvent was removed under reduced pressure to give the crude products. The crude residues were purified by crystallization from 95% ethanol. Spectral data for **5a–f** and **5i–j** have been previously reported [24].



**Scheme 3.** One-pot synthesis of oxazol-5-ones and azlactones from carboxylic acids.

**Table 3**

One-pot synthesis of azlactones **6a–m** from carboxylic acids using the Vilsmeier reagent.

Entry	R <sup>1</sup>	R <sup>2</sup>	Product	Isolated yield (%)
1	Ph	Ph	<b>6a</b>	92
2	Ph	4-ClC <sub>6</sub> H <sub>4</sub>	<b>6b</b>	94
3	Ph	4-MeOC <sub>6</sub> H <sub>4</sub>	<b>6c</b>	87
4	Ph	CH <sub>3</sub>	<b>6d</b>	69
5	4-MeOC <sub>6</sub> H <sub>4</sub>	2-Furyl	<b>6e</b>	82
6	4-ClC <sub>6</sub> H <sub>4</sub>	4-MeOC <sub>6</sub> H <sub>4</sub>	<b>6f</b>	88
7	4-NO <sub>2</sub> C <sub>6</sub> H <sub>4</sub>	4-NO <sub>2</sub> C <sub>6</sub> H <sub>4</sub>	<b>6g</b>	85
8	C <sub>6</sub> H <sub>5</sub> CH <sub>2</sub>	4-MeC <sub>6</sub> H <sub>4</sub>	<b>6h</b>	90
9	4-(Me <sub>2</sub> N)C <sub>6</sub> H <sub>4</sub>	Ph	<b>6i</b>	87
10	4-(Me <sub>2</sub> N)C <sub>6</sub> H <sub>4</sub>	4-ClC <sub>6</sub> H <sub>4</sub>	<b>6j</b>	94
11	4-Pyridyl	4-MeOC <sub>6</sub> H <sub>4</sub>	<b>6k</b>	66
12	2-Naphthyl	4-MeC <sub>6</sub> H <sub>4</sub>	<b>6l</b>	85
13	CH <sub>3</sub> –CH=CH	Ph	<b>6m</b>	78

#### 4.2.1. 2-(4-(Dimethylamino)phenyl)oxazol-5(4H)-one (**5g**)

Yellow solid, mp 129–131 °C. IR (KBr) cm<sup>-1</sup>: 1802 (C=O), 1631 (C=N). <sup>1</sup>H NMR (CDCl<sub>3</sub>) δ 3.17 (2 Me, s, 6H), 4.35 (CH<sub>2</sub>, s, 2H), 6.84 (ArH, d, 2H, *J* = 7.6), 7.47 (ArH, d, 2H, *J* = 7.6). <sup>13</sup>C NMR (CDCl<sub>3</sub>) δ 44.8 (Me), 57.6 (CH<sub>2</sub>), 109.4, 125.2, 129.9, 154.6 (aromatic carbons), 164.5 (C=N), 176.2 (C=O). Anal. Calcd for C<sub>11</sub>H<sub>12</sub>N<sub>2</sub>O<sub>2</sub>: C, 64.69; H, 5.92; N, 13.72. Found: C, 64.81; H, 6.08; N, 13.66.

#### 4.2.2. 2-(Pyridin-4-yl)oxazol-5(4H)-one (**5h**)

Pale-yellow solid, mp 113–115 °C. IR (KBr) cm<sup>-1</sup>: 1811 (C=O), 1634 (C=N). <sup>1</sup>H NMR (CDCl<sub>3</sub>) δ 4.18 (CH<sub>2</sub>, s, 2H), 7.33 (ArH, d, 2H, *J* = 7.9), 8.46 (ArH, d, 2H, *J* = 7.9). <sup>13</sup>C NMR (CDCl<sub>3</sub>) δ 55.9 (CH<sub>2</sub>), 119.7, 134.1, 151.5 (aromatic carbons), 167.1 (C=N), 178.4 (C=O). Anal. Calcd for C<sub>8</sub>H<sub>6</sub>N<sub>2</sub>O<sub>2</sub>: C, 59.26; H, 3.73; N, 17.28. Found: C, 59.35; H, 3.84; N, 17.31.

#### 4.2.3. 2-(Naphthalen-2-yl)oxazol-5(4H)-one (**5k**)

Orange solid, mp 154–156 °C. IR (KBr) cm<sup>-1</sup>: 1805 (C=O), 1639 (C=N). <sup>1</sup>H NMR (CDCl<sub>3</sub>) δ 4.53 (CH<sub>2</sub>, s, 2H), 7.26–7.29 (ArH, m, 2H), 7.60–7.65 (ArH, m, 3H), 7.72–7.75 (ArH, m, 2H). <sup>13</sup>C NMR (CDCl<sub>3</sub>) δ 53.7 (CH<sub>2</sub>), 108.4, 114.7, 119.4, 121.2, 125.9, 127.5, 128.3, 131.7, 138.6, 144.9 (aromatic carbons), 166.2 (C=N), 174.5 (C=O). Anal. Calcd for C<sub>13</sub>H<sub>9</sub>N<sub>2</sub>O<sub>2</sub>: C, 73.92; H, 4.30; N, 6.63. Found: C, 74.02; H, 4.44; N, 6.55.

#### 4.2.4. 2-(Prop-1-en-1-yl)oxazol-5(4H)-one (**5l**)

Colorless viscous oil. IR (KBr) cm<sup>-1</sup>: 1808 (C=O), 1626 (C=N). <sup>1</sup>H NMR (CDCl<sub>3</sub>) δ 1.39 (Me, m, 3H), 5.26 (vinylic H, d, 1H, *J* = 15.7), 5.94 (vinylic H, m, 1H). <sup>13</sup>C NMR (CDCl<sub>3</sub>) δ 20.6 (Me), 56.1 (CH<sub>2</sub>), 116.8, 138.5 (C=C), 159.4 (C=N), 177.1 (C=O). Anal. Calcd for C<sub>6</sub>H<sub>7</sub>N<sub>2</sub>O<sub>2</sub>: C, 57.59; H, 5.64; N, 11.19. Found: C, 57.51; H, 5.71; N, 11.16.

### 4.3. General procedure for one-pot synthesis of oxazol-5-ones **5a–l** from carboxylic acids

Glycine (1.0 mmol) was added to a solution of carboxylic acid (1.0 mmol), Vilsmeier's reagent (2.0 mmol), and Et<sub>3</sub>N (4.0 mmol) in dry chloroform (15 mL) at room temperature. The mixture was stirred at room temperature 2 h. Saturated NaHCO<sub>3</sub> (20 mL) was added and the organic layer was separated. It was washed with brine (20 mL), dried

(Na<sub>2</sub>SO<sub>4</sub>), filtered, and the solvent was removed under reduced pressure to give the crude products. The crude residues were purified by crystallization from 95% ethanol.

### 4.4. General procedure for one-pot synthesis of azlactones **6a–m** from carboxylic acids

The corresponding aldehyde (1.0 mmol) was added to a solution of the corresponding carboxylic acid (1.0 mmol), Vilsmeier's reagent (3.0 mmol), and Et<sub>3</sub>N (5.0 mmol) in dry chloroform (20 mL) at room temperature. After 2 h, saturated NaHCO<sub>3</sub> (20 mL) was added and the organic layer was separated. It was washed with brine (20 mL), dried (Na<sub>2</sub>SO<sub>4</sub>), filtered, and the solvent was removed under reduced pressure to give the crude products. The crude residues were purified by crystallization from 70% ethanol. Spectral data for **6a–g**, **6i**, and **6m** have been previously reported [8d,25].

#### 4.4.1. 2-Benzyl-4-(4-methoxybenzylidene)oxazol-5(4H)-one (**6h**)

Orange powder, mp 173–175 °C. IR (KBr) cm<sup>-1</sup>: 1631 (C=N), 1793 (C=O). <sup>1</sup>H NMR (CDCl<sub>3</sub>) δ 3.11 (CH<sub>2</sub>, s, 2H), 3.57 (OMe, s, 3H), 7.06 (ArH, d, 2H, *J* = 7.9), 7.28–7.32 (ArH, m, 5H), 7.73 (ArH, s, 1H), 7.87 (ArH, d, 2H, *J* = 7.9). <sup>13</sup>C NMR (CDCl<sub>3</sub>) δ 33.1 (CH<sub>2</sub>), 55.3 (OMe), 112.6, 114.9, 119.0, 122.4, 123.0, 124.7, 130.1, 138.3, 143.4, 154.8 (C=C, aromatic carbons), 162.8 (C=N), 166.4 (C=O). Anal. Calcd for C<sub>18</sub>H<sub>15</sub>NO<sub>3</sub>: C, 73.71; H, 5.15; N, 4.78. Found: C, 73.81; H, 5.29; N, 4.83.

#### 4.4.2. 4-(4-Chlorobenzylidene)-2-(4-(dimethylamino)phenyl)-oxazol-5(4H)-one (**6j**)

Bright yellow solid, mp 186–188 °C. IR (KBr) cm<sup>-1</sup>: 1637 (C=N), 1801 (C=O). <sup>1</sup>H NMR (CDCl<sub>3</sub>) δ 3.06 (Me, s, 3H), 6.91 (ArH, d, 2H, *J* = 8.1), 7.33–7.39 (ArH, m, 4H), 7.77–7.81 (ArH, m, 3H). <sup>13</sup>C NMR (CDCl<sub>3</sub>) δ 42.5 (Me), 111.2, 115.7, 119.3, 124.4, 126.1, 127.5, 129.8, 133.6, 145.3, 151.7 (C=C, aromatic carbons), 161.3 (C=N), 165.8 (C=O). Anal. Calcd for C<sub>18</sub>H<sub>15</sub>ClN<sub>2</sub>O<sub>2</sub>: C, 66.16; H, 4.63; N, 8.57. Found: C, 66.11; H, 4.71; N, 8.54.

#### 4.4.3. 4-(4-Methoxybenzylidene)-2-(pyridin-4-yl)oxazol-5(4H)-one (**6k**)

Orange solid, mp 174–176 °C. IR (KBr) cm<sup>-1</sup>: 1630 (C=N), 1791 (C=O). <sup>1</sup>H NMR (CDCl<sub>3</sub>) δ 3.69 (OMe, s, 3H), 7.17 (ArH, d, 2H, *J* = 7.7), 7.49 (ArH, d, 2H, *J* = 7.7), 7.75–7.81 (ArH, m, 3H), 8.30 (ArH, d, 2H, *J* = 7.6). <sup>13</sup>C NMR (CDCl<sub>3</sub>) δ 56.5 (OMe), 110.6, 114.1, 117.8, 122.6, 127.3, 132.4, 133.9, 147.5, 155.3 (C=C, aromatic carbons), 162.6 (C=N), 167.3 (C=O). Anal. Calcd for C<sub>16</sub>H<sub>12</sub>N<sub>2</sub>O<sub>3</sub>: C, 68.56; H, 4.32; N, 9.99. Found: C, 68.64; H, 4.43; N, 10.05.

#### 4.4.4. 4-Benzylidene-2-(naphthalen-2-yl)oxazol-5(4H)-one (**6l**)

Orange solid, mp 195–197 °C. IR (KBr) cm<sup>-1</sup>: 1636 (C=N), 1797 (C=O). <sup>1</sup>H NMR (CDCl<sub>3</sub>) δ 7.29–7.37 (ArH, m, 5H), 7.44–7.47 (ArH, m, 2H), 7.71–7.76 (ArH, m, 4H), 7.98–8.01 (ArH, m, 2H). <sup>13</sup>C NMR (CDCl<sub>3</sub>) δ 107.4, 111.8, 120.6, 123.1, 125.8, 126.1, 126.5, 127.9, 128.5, 132.6, 132.9, 134.7, 138.4, 142.0, 146.2, 147.9 (C=C, aromatic carbons), 164.3 (C=N),

166.1 (C=O). Anal. Calcd for C<sub>20</sub>H<sub>13</sub>NO<sub>2</sub>: C, 80.25; H, 4.38; N, 4.68. Found: C, 80.36; H, 4.53; N, 4.60.

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