

# Preparation of pyran derivatives promoted by $\text{FeCl}_3 \cdot 6\text{H}_2\text{O}$ in ionic liquid

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The condensation reaction of chalcone and 1,3-cyclohexanedione was smoothly carried out in  $[\text{bmim}][\text{BF}_4]$  with catalysis by  $\text{FeCl}_3 \cdot 6\text{H}_2\text{O}$ . A series of substituted pyrans were obtained by this reaction, in high yields under mild conditions through a simple operational procedure. In addition, the ionic liquid together with the catalyst used can be conveniently recovered and efficiently reused.

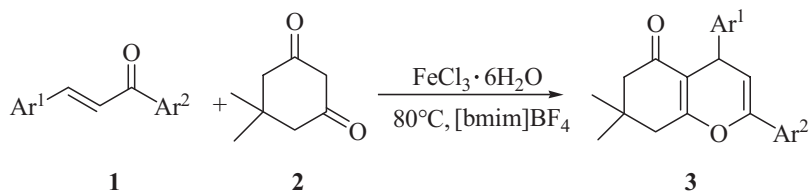
**Keywords:** pyran derivatives, ionic liquid,  $\text{FeCl}_3 \cdot 6\text{H}_2\text{O}$ , green synthesis

Pyran derivatives are of considerable interest because of their wide range of biological properties, such as spasmolytic, diuretic, anti-coagulant, anti-cancer, anti-anaphylactic activity. In addition, they can be used as cognitive enhancers for the treatment of neurodegenerative disease, including Alzheimer's disease, amyotrophic lateral sclerosis, and Parkinson's disease.<sup>1-5</sup> In particular, the 4*H*-benzopyran moiety is a constituent of the structures of a series of natural products with interesting biological and pharmacological activities.<sup>6,7</sup> Consequently the development of novel methods for the synthesis of these heterocyclic compounds has been of considerable interest in both organic and medicinal fields. The condensation of  $\alpha$ ,  $\beta$ -unsaturated ketones with 1,3-cyclohexanedione promoted by various catalysts has appeared as a novel alternative.<sup>8</sup> In this regard, anhydrous zinc chloride was recently utilised to catalyse this reaction to afford substituted benzopyran derivatives. However, this method needs to be further improved because of the low yields. It has also been reported that a combination of glacial acetic acid and phosphoric anhydride can promote the condensation of  $\alpha$ ,  $\beta$ -unsaturated ketones with 1,3-cyclohexanedione, but the use of strongly acidic compounds conflicts with the principles of green chemistry and may exclude some acid sensitive substrates.<sup>9</sup> As a result, the development of efficient, milder and greener synthetic methods for the preparation of 4*H*-benzopyran derivatives is still required.

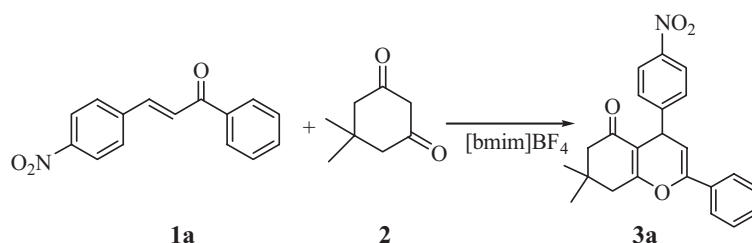
Green chemistry technologies based on ionic liquid which circumvent the use of volatile organic compounds are attractive in organic synthesis. The unique properties of ionic

liquids, such as good solvating capability, wide liquid range, negligible vapour pressure, and ease of recycling, offer an environmentally acceptable organic synthesis. Many classical organic processes have successfully been carried out in ionic liquids. Recently, we have developed novel methods for the preparation of various biologically important heterocyclic compounds, such as 1,4-DHPs, xanthenedione and quinolines in multi-component reactions by using ionic liquids as novel reaction medium and/or promoter.<sup>10-13</sup> Herein we report our study of a novel preparation of substituted pyran derivatives from reaction of chalcones (**1**) with 1,3-cyclohexanedione (**2**) in the presence of  $\text{FeCl}_3 \cdot 6\text{H}_2\text{O}$  in an ionic liquid, 1-butyl-3-methylimidazolium tetrafluoroborate ( $[\text{bmim}][\text{BF}_4]$ ) (Scheme 1).

Our study was initiated by the treatment of 3-(4-nitrophenyl)-1-phenylprop-2-en-1-one (**1a**), previously prepared from 4-nitrobenzaldehyde and acetophenone, with 5,5-dimethyl-1,3-cyclohexanedione (**2**) in  $[\text{bmim}][\text{BF}_4]$  at room temperature without added catalyst. TLC analysis showed that no reaction took place after the mixture was stirred at room temperature for several hours. The reaction was then attempted at elevated temperatures and it turned out that when it was run at 80°C, it gradually gave a new substance. However, the reaction was still incomplete after the mixture had been stirred at 80°C for 2 days. At this point, the reaction was interrupted and the new substance was separated and identified. <sup>1</sup>H NMR spectroscopy together with mass spectrometry result showed it is the desired product **3a** with a yield of 35% (Scheme 2).



Scheme 1



Scheme 2

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To improve the above reaction with regard to reaction time and yield, several mild Lewis acid type catalysts including  $\text{InCl}_3 \cdot 4\text{H}_2\text{O}$ ,  $\text{FeCl}_3 \cdot 6\text{H}_2\text{O}$ ,  $\text{CeCl}_3 \cdot 7\text{H}_2\text{O}$ ,  $\text{Fe}_2(\text{SO}_4)_3 \cdot x\text{H}_2\text{O}$  and  $\text{ZnCl}_2$  were then tried by using **1a** and **2** as model substrates. The results were listed in Table 1.

From the results listed in Table 1, it can be concluded that the above reaction was significantly improved by the addition of Lewis acid catalyst. Of the catalysts examined,  $\text{FeCl}_3 \cdot 6\text{H}_2\text{O}$  turned out to be the most effective one. For example, in the presence of 20 mol%  $\text{FeCl}_3 \cdot 6\text{H}_2\text{O}$ , reaction of **1a** and **2** gave **3a** exclusively after the mixture was stirred at 80°C for 6 h with a yield of 80%. In the presence of other catalysts, the yields were somewhat lower and the reaction needed a longer time to complete. As for the loading of  $\text{FeCl}_3 \cdot 6\text{H}_2\text{O}$ , the results listed in Table 1 showed that 20 mol % of  $\text{FeCl}_3 \cdot 6\text{H}_2\text{O}$  gave a good result and increasing this amount to 50 mol % did not make any obvious difference in terms of yield and reaction time. Therefore, 20 mol % was chosen as the optimum loading of  $\text{FeCl}_3 \cdot 6\text{H}_2\text{O}$ .

Encouraged by the results obtained with **1a** and **2**, a variety of substituted chalcones were tested to investigate the scope and generality of this novel method. The results are listed in Table 2. It showed that a wide range of chalcones reacted smoothly with **2** in  $[\text{bmim}][\text{BF}_4]$  and gave **3** in good yields. It is worth noting that the electronic properties of the aromatic ring of the chalcones has some effects on the yield of the

products and the reaction time. For example, for substrates bearing electron-withdrawing groups on the aromatic rings, shorter reaction time was needed and higher yields were obtained (for **3a**, **3b**, **3f** and **3j**). On the other hand, a longer reaction time was needed and lower yields were usually obtained with substrates bearing electron-donating groups on the aromatic rings (for **3i**).

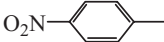
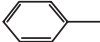
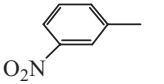
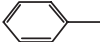
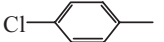
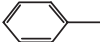
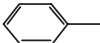
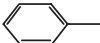
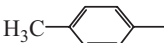
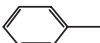
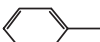
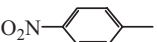
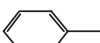
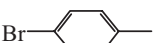
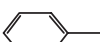
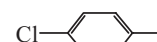
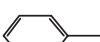
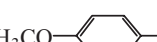

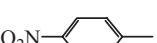
It is well documented that, compared with classical organic solvents, reactions carried out in ionic liquids possess the advantages not only of green chemistry, but are often simpler processes and show enhanced reactivity and better yields. This was demonstrated by comparing  $[\text{bmim}][\text{BF}_4]$  with several classical organic solvents including ethanol, toluene and THF. The following is the results (shown in Table 3) of investigations by using **1a** and **2** as model substrates for the preparation of **3a** (Scheme 2).

It can be seen that of the four solvents studied,  $[\text{bmim}][\text{BF}_4]$  gave the highest yields for **3a** (80%, Table 3, entry 1) at 80°C. Moreover, using  $[\text{bmim}][\text{BF}_4]$  as the reaction medium also made the separation process much easier. For example, in  $[\text{bmim}][\text{BF}_4]$ , **3a** is in solid state and could be conveniently isolated with high purity by filtration. On the other hand, when ethanol, toluene or THF were used, a complicated mixture was formed and **3a** could only be obtained by column chromatography.

**Table 1** Reaction of **1a** and **2** in  $[\text{bmim}][\text{BF}_4]$  with various catalysts

| Entry | Catalyst/mol%   | Time/h | Temperature/°C | Yield/% |
|-------|---|--------|----------------|---------|
| 1     | –   | 24     | 80             | 28      |
| 2     | $\text{InCl}_3 \cdot 4\text{H}_2\text{O}$ (20)              | 8      | 80             | 68      |
| 3     | $\text{FeCl}_3 \cdot 6\text{H}_2\text{O}$ (20)              | 6      | 80             | 80      |
| 4     | $\text{CeCl}_3 \cdot 7\text{H}_2\text{O}$ (20)              | 12     | 80             | 60      |
| 5     | $\text{Fe}_2(\text{SO}_4)_3 \cdot x\text{H}_2\text{O}$ (20) | 10     | 80             | 70      |
| 6     | $\text{ZnCl}_2$ (20)  | 15     | 80             | 58      |
| 7     | $\text{FeCl}_3 \cdot 6\text{H}_2\text{O}$ (50)              | 6      | 80             | 82      |
| 8     | $\text{FeCl}_3 \cdot 6\text{H}_2\text{O}$ (10)              | 6      | 80             | 60      |
| 9     | $\text{FeCl}_3 \cdot 6\text{H}_2\text{O}$ (5)               | 6      | 80             | 36      |

**Table 2** Preparation of **3** promoted by  $\text{FeCl}_3 \cdot 6\text{H}_2\text{O}$  (20% mol) in ionic liquid

| Products  | Ar <sup>1</sup>   | Ar <sup>2</sup>   | Time/h | Temperature/°C | Yield/% |
|-----------|---|---|--------|----------------|---------|
| <b>3a</b> |  |  | 6      | 80             | 80      |
| <b>3b</b> |  |  | 6      | 80             | 81      |
| <b>3c</b> |  |  | 8      | 80             | 75      |
| <b>3d</b> |  |  | 8      | 80             | 70      |
| <b>3e</b> |  |  | 10     | 80             | 70      |
| <b>3f</b> |  |  | 6      | 80             | 85      |
| <b>3g</b> |  |  | 8      | 80             | 76      |
| <b>3h</b> |  |  | 8      | 80             | 75      |
| <b>3i</b> |  |  | 10     | 80             | 62      |
| <b>3j</b> |  |  | 6      | 80             | 82      |

**Table 3** Preparation of **3a** in different solvents

| Entry | Product   | Solvent                  | Temperature/°C | Time/h | Yield <sup>a</sup> /% |
|-------|-----------|--------------------------|----------------|--------|-----------------------|
| 1     | <b>3a</b> | [bmim][BF <sub>4</sub> ] | 80             | 6      | 80                    |
| 2     | <b>3a</b> | Ethanol                  | Reflux         | 6      | 28                    |
| 3     | <b>3a</b> | Toluene                  | 80             | 6      | 40                    |
| 4     | <b>3a</b> | THF                      | Reflux         | 6      | 46                    |

<sup>a</sup>In the presence of FeCl<sub>3</sub>·6H<sub>2</sub>O (20 mol%).

**Table 4** Reusability of [bmim][BF<sub>4</sub>] and catalyst for the preparation of **3a**

| Entry | Round | IL recovered/% | Time/h | Temperature/°C | Yield/% |
|-------|-------|----------------|--------|----------------|---------|
| 1     | 1     | 96             | 6      | 80             | 80      |
| 2     | 2     | 95             | 6      | 80             | 82      |
| 3     | 3     | 92             | 6      | 80             | 78      |
| 4     | 4     | 90             | 6      | 80             | 75      |

<sup>a</sup>In the presence of FeCl<sub>3</sub>·6H<sub>2</sub>O (20 mol%).

The possibility of the recovery and reutilisation of the catalyst and solvent was then studied. Upon completion, the product was in fact in a solid state and could be simply collected by suction. The filtrate containing [bmim][BF<sub>4</sub>] together with the immobilised Fe (III) was then dried at 100°C. Investigations by using **1a** and **2** as model substrates again showed that successive reuse of the recovered ionic liquid and catalyst in the same reaction gave **3a** with a yield of 82% (Table 4, Entry 2). It was also observed that in the fourth round, **3a** could be obtained with good yield by using the ionic liquid and the catalyst recovered from the third round (Table 4, entry 4).

In conclusion, we have demonstrated that the condensation reaction between chalcone and 1,3-cyclohexanedione could be carried out smoothly in [bmim][BF<sub>4</sub>] with catalysis by FeCl<sub>3</sub>·6H<sub>2</sub>O. In this reaction, substituted pyrans were obtained efficiently. It should be noted that this procedure offers several advantages including the use of low-loading of a green catalyst, green solvent, improved yields and simple experimental procedures. In addition, easy workup has been realised compared with the use of conventional organic solvent in that the products can be obtained by filtration upon completion of the reaction.

## Experimental

Melting points were measured by a Kofler micromelting point apparatus and were uncorrected. <sup>1</sup>H NMR spectra were determined on a Bruker AC 400 spectrometer as CDCl<sub>3</sub> or DMSO-*d*<sub>6</sub> solutions. Chemical shifts (δ) were expressed in ppm downfield from the internal standard tetramethylsilane and coupling constants *J* were given in Hz. Mass spectra were obtained in ESI mode using a Bruker Esquire 3000 mass spectrometer. Elemental analyses were performed on an EA-1110 instrument.

### General procedure for the preparation of substituted pyran derivatives (**3**)

To [bmim][BF<sub>4</sub>] 1 ml were added chalcone 1 mmol, 1, 3-cyclohexanedione 1 mmol and FeCl<sub>3</sub>·6H<sub>2</sub>O (20 mol%). The reaction mixture was stirred at 80°C until the reaction was complete (monitored by TLC). Then, the mixture was cooled to room temperature and 50% ethanol water 1 ml was added. The solids were collected by suction and rinsed with water and ethanol, and then dried to give **3** high purity. The ionic liquid layer was dried at 100°C under reduced pressure to recover the ionic liquid together with the catalyst.

**3a**: M.p. 131–132°C; IR (KBr) (cm<sup>-1</sup>): 3090, 3051, 2960, 2874, 1680, 1652, 1590, 1516; <sup>1</sup>H NMR (CDCl<sub>3</sub>): 1.09 (s, 3H, CH<sub>3</sub>), 1.18 (s, 3H, CH<sub>3</sub>), 2.22–2.61 (m, 4H, 2 × CH<sub>2</sub>), 4.65 (d, 1H, *J* = 4.8 Hz, CH), 5.67 (d, 1H, *J* = 4.8 Hz, =CH), 7.40–7.42 (m, 3H, ArH), 7.52 (d, 2H, *J* = 8.0 Hz, ArH), 7.61–7.62 (m, 2H, ArH), 8.17 (d, 2H, *J* = 8.0 Hz, ArH); MS: *m/z* 398 [M + Na]<sup>+</sup>. Anal. Calcd for C<sub>23</sub>H<sub>21</sub>NO<sub>4</sub>: C 73.6, H 5.6, N 3.7; found: C 73.8, H 5.4, N 3.6%.

**3b**: M.p. 139–141°C; IR (KBr) (cm<sup>-1</sup>): 3075, 3066, 2960, 2868, 1681, 1660, 1631, 1529; <sup>1</sup>H NMR (CDCl<sub>3</sub>): 1.11 (s, 3H, CH<sub>3</sub>), 1.18 (s, 3H, CH<sub>3</sub>), 2.23–2.67 (m, 4H, 2 × CH<sub>2</sub>), 4.66 (d, 1H, *J* = 4.8 Hz,

CH), 5.68 (d, 1H, *J* = 4.8 Hz, =CH), 7.37–7.49 (m, 4H, ArH), 7.62 (d, 2H, *J* = 7.6 Hz, ArH), 7.72 (d, 1H, *J* = 7.6 Hz, ArH), 8.07 (d, 1H, *J* = 8.4 Hz, ArH), 8.19 (s, 1H, ArH); MS: *m/z* 398 [M + Na]<sup>+</sup>. Anal. Calcd for C<sub>23</sub>H<sub>21</sub>NO<sub>4</sub>: C 73.6, H 5.6, N 3.7; found: C 73.8, H 5.6, N 3.7%.

**3c**: M.p. 110–111°C; IR (KBr) (cm<sup>-1</sup>): 3080, 3050, 2950, 2871, 1679, 1658, 1627, 1500; <sup>1</sup>H NMR (CDCl<sub>3</sub>): 1.08 (s, 3H, CH<sub>3</sub>), 1.16 (s, 3H, CH<sub>3</sub>), 2.22–2.58 (m, 4H, 2 × CH<sub>2</sub>), 4.51 (d, 1H, *J* = 4.8 Hz, CH), 5.70 (d, 1H, *J* = 4.8 Hz, =CH), 7.26–7.31 (m, 4H, ArH), 7.35–7.42 (m, 3H, ArH), 7.61 (d, 2H, *J* = 7.6 Hz, ArH); MS: *m/z* 387 [M + Na]<sup>+</sup>. Anal. Calcd for C<sub>23</sub>H<sub>21</sub>ClO<sub>2</sub>: C 75.7, H 5.8; found: C 75.5, H 6.0%.

**3d**: M.p. 140–141°C (lit.<sup>8</sup> 140°C); IR (KBr)(cm<sup>-1</sup>): 3068, 3030, 2946, 2872, 1679, 1658, 1627, 1488; <sup>1</sup>H NMR (CDCl<sub>3</sub>): 1.06 (s, 3H, CH<sub>3</sub>), 1.17 (s, 3H, CH<sub>3</sub>), 2.23–2.59 (m, 4H, 2 × CH<sub>2</sub>), 4.49 (d, 1H, *J* = 4.8 Hz, CH), 5.70 (d, 1H, *J* = 4.8 Hz, =CH), 7.14–7.18 (m, 1H, ArH), 7.25–7.38 (m, 7H, ArH), 7.57–7.59 (m, 2H, ArH).

**3e**: M.p. 84–86°C; IR (KBr) (cm<sup>-1</sup>): 3050, 3026, 2955, 2871, 1680, 1656, 1628, 1512; <sup>1</sup>H NMR (CDCl<sub>3</sub>): 1.06 (s, 3H, CH<sub>3</sub>), 1.13 (s, 3H, CH<sub>3</sub>), 2.18–2.58 (m, 7H, CH<sub>2</sub>, 2 × CH<sub>2</sub>), 4.56 (d, 1H, *J* = 4.8 Hz, CH), 5.69 (d, 1H, *J* = 4.8 Hz, =CH), 7.08 (d, 2H, *J* = 7.6 Hz, ArH), 7.21 (d, 2H, *J* = 7.6 Hz, ArH), 7.30–7.37 (m, 3H, ArH), 7.58 (d, 2H, *J* = 8.0 Hz, ArH). MS: *m/z* 367 [M + Na]<sup>+</sup>. Anal. Calcd for C<sub>24</sub>H<sub>24</sub>O<sub>2</sub>: C 83.7, H 7.0; found: C 83.5, H 6.9%.

**3f**: M.p. 148–150°C; IR (KBr) (cm<sup>-1</sup>): 3068, 3025, 2958, 2866, 1658, 1625, 1594, 1516; <sup>1</sup>H NMR (CDCl<sub>3</sub>): 1.12 (s, 3H, CH<sub>3</sub>), 1.18 (s, 3H, CH<sub>3</sub>), 2.24–2.67 (m, 4H, 2 × CH<sub>2</sub>), 4.57 (d, 1H, *J* = 4.8 Hz, CH), 5.94 (d, 1H, *J* = 4.8 Hz, =CH), 7.22–7.33 (m, 5H, ArH), 7.77 (d, 2H, *J* = 8.4 Hz, ArH), 8.24 (d, 2H, *J* = 8.4 Hz, ArH); MS: *m/z* 398 [M + Na]<sup>+</sup>. Anal. Calcd for C<sub>23</sub>H<sub>21</sub>NO<sub>4</sub>: C 73.6, H 5.6, N 3.7; found: C 73.8, H 5.4, N 3.5%.

**3g**: M.p. 183–184°C; IR (KBr) (cm<sup>-1</sup>): 3070, 3026, 2955, 2869, 1669, 1628, 1489, 1456; <sup>1</sup>H NMR (CDCl<sub>3</sub>): 1.10 (s, 3H, CH<sub>3</sub>), 1.17 (s, 3H, CH<sub>3</sub>), 2.22–2.62 (m, 4H, 2 × CH<sub>2</sub>), 4.51 (d, 1H, *J* = 4.8 Hz, CH), 5.73 (d, 1H, *J* = 4.8 Hz, =CH), 7.19–7.22 (m, 1H, ArH), 7.29–7.34 (m, 4H, ArH), 7.46–7.53 (m, 4H, ArH); MS: *m/z* 431 [M + Na]<sup>+</sup>. Anal. Calcd for C<sub>23</sub>H<sub>21</sub>BrO<sub>2</sub>: C 67.5, H 5.2; found: C 67.2, H 5.4%.

**3h**: M.p. 172–174°C (lit.<sup>8</sup> 175–176°C); IR (KBr) (cm<sup>-1</sup>): 3079, 3046, 2958, 2870, 1688, 1655, 1626, 1490; <sup>1</sup>H NMR (CDCl<sub>3</sub>): 1.09 (s, 3H, CH<sub>3</sub>), 1.16 (s, 3H, CH<sub>3</sub>), 2.22–2.63 (m, 4H, 2 × CH<sub>2</sub>), 4.51 (d, 1H, *J* = 4.8 Hz, CH), 5.74 (d, 1H, *J* = 4.8 Hz, =CH), 7.20–7.34 (m, 5H, ArH), 7.36 (d, 2H, *J* = 8.4 Hz, ArH), 7.53 (d, 2H, *J* = 8.4 Hz, ArH).

**3i**: M.p. 133–134°C; IR (KBr) (cm<sup>-1</sup>): 3066, 3018, 2971, 2870, 1686, 1658, 1620, 1460; <sup>1</sup>H NMR (DMSO-*d*<sub>6</sub>): 1.06 (s, 3H, CH<sub>3</sub>), 1.13 (s, 3H, CH<sub>3</sub>), 2.18–2.58 (m, 4H, 2 × CH<sub>2</sub>), 3.81 (s, 3H, OCH<sub>3</sub>), 4.47 (d, 1H, *J* = 4.8 Hz, CH), 5.57 (d, 1H, *J* = 4.8 Hz, =CH), 6.89 (d, 2H, *J* = 9.6 Hz, ArH), 7.14–7.18 (m, 1H, ArH), 7.25–7.33 (m, 4H, ArH), 7.51 (d, 2H, *J* = 9.6 Hz, ArH); MS: *m/z* 383 [M + Na]<sup>+</sup>. Anal. Calcd for C<sub>24</sub>H<sub>24</sub>O<sub>3</sub>: C 80.0, H 6.7; found: C 80.2, H 6.7%.

**3j**: M.p. 202–204°C; IR (KBr) (cm<sup>-1</sup>): 3065, 3012, 2966, 2868, 1670, 1656, 1627, 1512; <sup>1</sup>H NMR (CDCl<sub>3</sub>): 1.10 (s, 3H, CH<sub>3</sub>), 1.18 (s, 3H, CH<sub>3</sub>), 2.23–2.60 (m, 4H, 2 × CH<sub>2</sub>), 4.54 (d, 1H, *J* = 4.8 Hz, CH), 5.89 (d, 1H, *J* = 4.8 Hz, =CH), 7.25–7.30 (m, 4H, ArH), 7.76 (d, 2H, *J* = 8.8 Hz, ArH), 8.25 (d, 2H, *J* = 8.8 Hz, ArH); MS: *m/z* 432 [M + Na]<sup>+</sup>. Anal. Calcd for C<sub>23</sub>H<sub>20</sub>ClNO<sub>4</sub>: C 67.4, H 4.9, N 3.4; found: C 67.4, H 4.7, N 3.6%.

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