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**TiO$_2$–SiO$_2$ nanocomposite as a highly efficient catalyst for the solvent-free cyclocondensation reaction of isatins, cyclohexanones, and urea**

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**Abstract:** A TiO$_2$–SiO$_2$ nanocomposite catalyzes the one-pot cyclocondensation reaction of 1H-indole-2,3-diones (isatins), 1,3-cyclohexanediones, and urea at $T=70^\circ$C under solvent-free conditions with high efficiency. The present method affords the corresponding spiro products in high yields and short reaction time. The unique features of the TiO$_2$–SiO$_2$ nanocomposite are high catalytic activity, ease of preparation, non-toxicity, low loading, and recyclability.

**Keywords:** catalyst recyclability; green procedure; isatins; solvent-free; TiO$_2$–SiO$_2$ nanocomposite.

**1 Introduction**

In the last decade, spiroheterocycles containing 1H-indole-2,3-diones (isatins) as versatile building blocks have gained attention because of their interesting pharmacological properties [1–3]. These compounds have been found to possess a variety of biological activities such as antimicrobial [4], antiviral [5], antitumor [6], mutagenicity [7], antiproliferative [8], and as sex pheromones [9].

Nanostructured metal oxides are generally used in heterogeneous catalysis of different organic transformations, owing to their salient features such as optimum porous size and an increase in the surface area [10–13]. TiO$_2$ nanoparticles (TiO$_2$ NPs) have been easily prepared in different compositions or crystalline structures and used widely as efficient heterogeneous catalysts to enhance the rate of various organic reactions [14–21]. Moreover, the catalytic efficiency of TiO$_2$ NPs is significantly improved by loading them on silica shells as support. The TiO$_2$ NPs loaded on silica shells (TiO$_2$–SiO$_2$ nanocomposite) have unique electronic properties and large specific surface areas [22].

There are many studies in the literature for the synthesis of spiroheterocycles, due to their wide range of useful biological activities. During our ongoing studies toward the development of TiO$_2$–SiO$_2$ nanocomposites as an effective heterogeneous catalyst for the synthesis of heterocyclic compounds [23–26], we prepared a set of eight diverse spiro[3, 4]1,3-dihydro-2H-indol-2-one-4′,6′,7′,8′-tetrahydro-2′,5′(1′H,3′H)-quinazoline-diones (4a–h) in high yields via a Biginelli-type reaction of isatins 1, 1,3-cyclohexanediones 2, and urea 3, in the presence of a catalytic amount of TiO$_2$–SiO$_2$ nanocomposite, at $T=70^\circ$C under solvent-free conditions (Scheme 1).

**2 Results and discussion**

The TiO$_2$–SiO$_2$ nanocomposite catalyst was prepared according to a reported non-thermal sol–gel procedure [27]. By powder X-ray diffraction (XRD), the crystalline structure and purity of the synthesized nanocomposite were clearly confirmed. The XRD pattern of the TiO$_2$–SiO$_2$ nanocomposite shows crystalline TiO$_2$ NPs (anatase modification) in the amorphous silica matrix. The morphology of the synthesized nanocomposites was also revealed by transmission electron microscopy (TEM). The TEM image of the nanocomposite shows the grain size of the synthesized nanocomposites to be in the range of 5–9 nm. The chemical composition of the nanocomposite was determined by X-ray fluorescence (XRF), and the results show that the composite consists of 55% TiO$_2$ and 45% SiO$_2$. Representative XRD and TEM images of the TiO$_2$–SiO$_2$ nanocomposite produced that way have already been shown in a previously published article [24].

Various parameters such as the effect of catalyst, temperature, and reaction media were investigated under different reaction conditions using the reaction of 5-bromo-1H-indole-2,3-dione (1b), 1,3-cyclohexanedione (2b), and urea 3 as a model reaction (Scheme 2 and Table 1).
When this reaction was performed at \( T=70^\circ C \) in the absence of a catalyst and under solvent-free conditions, only trace amounts of product 4b were obtained (thin-layer chromatography [TLC]; Table 1, entry 1). Subsequently, the model reaction was performed with the addition of different amounts of TiO\(_2\)-SiO\(_2\) nanocomposite as a catalyst under the same conditions. It was found that the best yield of product 4b is obtained using 15 mol.% of catalyst (Table 1, entries 2–4).

The study on the reaction media revealed that under solvent-free conditions, the best yield of product 4b is achieved (Table 1, entries 3 and 5–8). Finally, we tried to optimize the reaction temperature. It was shown that there is no significant improvement of the yield of product 4b by increasing the temperature up to 70°C (Table 1, entries 3 and 9–10).

To investigate the scope and generality of this new procedure, some 5-substituted isatins were examined in the reaction with 1,3-cyclohexanenediones and urea to obtain the expected products in high yields under optimized conditions (Table 2). The compounds 4a–h were cleanly isolated and characterized by IR, \(^1\)H NMR, and \(^13\)C NMR and also by elemental analyses.

![Scheme 1](image1.png)

**Scheme 1:** Synthesis of spiro[3, 4′]1,3-dihydro-2H-indol-2-one-4′,6′,7′,8′-tetrahydro-2′,5′(1′H,3′H)-quinazoline-diones (4a–h) using a TiO\(_2\)-SiO\(_2\) nanocomposite as a catalyst. For the substituents R and R′ and the corresponding compound numbering see Table 2.

![Scheme 2](image2.png)

**Scheme 2:** Synthesis of spiro[3, 4′]5-bromo-1,3-dihydro-2H-indol-2-one-4′,6′,7′,8′-tetrahydro-2′,5′(1′H,3′H)-quinazoline-dione (4b) as a model reaction for the screening of the reaction conditions and the catalyst.

**Table 1:** Synthesis of the spiroheterocycle 4b (R=Br) in the reaction of 5-bromo-1H-indole-2,3-dione (1a; R=Br), 1,3-cyclohexanenedione (2b), and urea (3) in the presence of a TiO\(_2\)-SiO\(_2\) nanocomposite (1:1) under different conditions (Scheme 2).

<table>
<thead>
<tr>
<th>Entry</th>
<th>Amount of TiO(_2)-SiO(_2) nanocomposite as a catalyst (%)</th>
<th>Solvent</th>
<th>Temp. (°C)</th>
<th>Time (h)</th>
<th>Yield (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>No catalyst</td>
<td>Neat</td>
<td>70</td>
<td>7</td>
<td>Trace</td>
</tr>
<tr>
<td>2</td>
<td>10</td>
<td>Neat</td>
<td>70</td>
<td>4</td>
<td>79</td>
</tr>
<tr>
<td>3</td>
<td>15</td>
<td>Neat</td>
<td>70</td>
<td>4</td>
<td>90</td>
</tr>
<tr>
<td>4</td>
<td>20</td>
<td>Neat</td>
<td>70</td>
<td>4</td>
<td>91</td>
</tr>
<tr>
<td>5</td>
<td>15</td>
<td>H(_2)O</td>
<td>Reflux</td>
<td>3</td>
<td>81</td>
</tr>
<tr>
<td>6</td>
<td>15</td>
<td>EtOH</td>
<td>Reflux</td>
<td>4</td>
<td>79</td>
</tr>
<tr>
<td>7</td>
<td>15</td>
<td>CH(_2)Cl(_2)</td>
<td>Reflux</td>
<td>4</td>
<td>78</td>
</tr>
<tr>
<td>8</td>
<td>15</td>
<td>DMF</td>
<td>Reflux</td>
<td>3</td>
<td>85</td>
</tr>
<tr>
<td>9</td>
<td>15</td>
<td>Neat</td>
<td>60</td>
<td>6</td>
<td>73</td>
</tr>
<tr>
<td>10</td>
<td>15</td>
<td>Neat</td>
<td>90</td>
<td>4</td>
<td>90</td>
</tr>
</tbody>
</table>

*According to the molar ratio of TiO\(_2\) to SiO\(_2\) in the TiO\(_2\)-SiO\(_2\) nanocomposite (1:1), the catalyst loadings with respect to TiO\(_2\) were calculated to be 14, 21, and 28 mg for 10, 15, and 20 mol.%, respectively. †Isolated yields.

<table>
<thead>
<tr>
<th>Product</th>
<th>R</th>
<th>R′</th>
<th>Yield (%)</th>
<th>Time (h)</th>
<th>m.p. (°C)</th>
</tr>
</thead>
<tbody>
<tr>
<td>4a</td>
<td>H</td>
<td>H</td>
<td>93</td>
<td>4</td>
<td>&gt;300 345–347 [28]</td>
</tr>
<tr>
<td>4b</td>
<td>Br</td>
<td>H</td>
<td>90</td>
<td>4</td>
<td>&gt;300  –</td>
</tr>
<tr>
<td>4c</td>
<td>OCH(_3)</td>
<td>H</td>
<td>92</td>
<td>4</td>
<td>&gt;300  –</td>
</tr>
<tr>
<td>4d</td>
<td>NO(_2)</td>
<td>H</td>
<td>95</td>
<td>4</td>
<td>&gt;300  –</td>
</tr>
<tr>
<td>4e</td>
<td>H</td>
<td>CH(_3)</td>
<td>94</td>
<td>3</td>
<td>&gt;300 350–352 [28]</td>
</tr>
<tr>
<td>4f</td>
<td>Br</td>
<td>CH(_3)</td>
<td>92</td>
<td>3</td>
<td>&gt;300  –</td>
</tr>
<tr>
<td>4g</td>
<td>OCH(_3)</td>
<td>CH(_3)</td>
<td>94</td>
<td>3</td>
<td>&gt;300  –</td>
</tr>
<tr>
<td>4h</td>
<td>NO(_2)</td>
<td>CH(_3)</td>
<td>97</td>
<td>3</td>
<td>&gt;300  –</td>
</tr>
</tbody>
</table>

*Yields of pure isolated products.
A plausible mechanism for the formation of product 4 is given in Scheme 2. It is conceivable that the TiO$_2$–SiO$_2$ nanocomposite catalyzes the formation of alkene 7 by a Knoevenagel-type condensation between isatin 1 and 1,3-cyclohexanedione 2 via the intermediates 5 and 6. This occurs by the coordination of the acidic sites of TiO$_2$ NPs with the oxygen of carbonyl group in isatin and increase the reactivity of isatin carbonyl group. Subsequently, urea 3 is added to alkene 7 to generate the Michael adduct 8. Cyclization of 8 and further dehydration of the cyclized compound 9 furnish product 4.

Furthermore, we studied the catalyst recyclability in the synthesis of product 4b. After completion of the reaction, the catalyst was successfully recovered in a simple manner as described in the general procedure and reused subsequently four times without significant loss of activity (product yields: 90%, 89%, 87%, and 86%, respectively).

3 Conclusion

In summary, we developed an efficient new procedure for the synthesis of spiro[3, 4’][1,3-dihydro-2H-indol-2-one-4′,6′,7′,8′-tetrahydro-2′,5′(1′H,3′H)-quinazoline-diones

Scheme 2: The proposed mechanism for the formation of 4.
in high yields via a Biginelli-type reaction of isatins 1, 1,3-cyclohexanediones 2, and urea (3), in the presence of a catalytic amount of TiO₂–SiO₂ nanocomposite at \( T = 70^\circ C \) under solvent-free conditions. In comparison with previous methods [28, 29], this procedure has several advantages such as facile synthesis, high product yields, short reaction times, easy separation, and reusability of the catalyst.

4 Experimental section

4.1 Materials and methods

All chemicals used in this work were purchased from Merck and Fluka in high purity (Kimiaexir Chemical Company, Tehran, Iran). Melting points were determined with Electrothermal 9100 apparatus (East Tehran Branch, Islamic Azad University, Tehran, Iran). FT-IR spectra were obtained using a Bruker Equinox 55 Golden Gate Micro-ATR spectrometer (Chemistry and Chemical Engineering Research Center of Iran, Tehran, Iran). ¹H NMR and ¹³C NMR spectra were recorded on a Bruker Gate Micro-ATR spectrometer (Chemistry and Chemistry Research Institute AEOI, Tehran, Iran). Elemental analyses were carried out using a Foss-Heraeus CHN-O-Rapid analyzer (Polymer and Petrochemical Institute, Tehran, Iran). The TEM images of the catalyst were obtained with a Philips EM208 transmission electron microscope under acceleration (Nuclear Science and Technology Research Institute AEOI). Powder XRD data were determined by XRF spectroscopy using Oxford ED 2000 apparatus (Nuclear Science and Technology Research Institute AEOI). The composition of the catalyst was determined by XRF spectroscopy using Oxford ED 2000 apparatus (Nuclear Science and Technology Research Institute AEOI).

4.2 General procedure for the synthesis of compounds 4a–h

Mixtures of isatins 1 (1 mmol), 1,3-cyclohexanediones 2 (1 mmol), and urea (3, 1.2 mmol), and a TiO₂–SiO₂ nanocomposite (21 mg, 15 mol.%), were heated under magnetic stirring at \( T = 70^\circ C \) for the given reaction times (Table 2) under solvent-free conditions. After completion of the reactions, as indicated by TLC (ethyl acetate as eluent), the reaction mixtures were dissolved in hot ethanol (5 mL) and then centrifuged for 5 min at 2000–3000 rpm to remove the catalyst. The catalyst was then washed with ethanol and dried in air at ambient temperature for more than 2 h for subsequent reuse. The resulting solutions were allowed to cool to room temperature. The white pure title products were obtained by dilution with H₂O (1 mL) and subsequent crystallization.

4.3 Spectroscopic and physical data

4.3.1 Spiro[3, 4’7]-1,3-dihydro-2H-indol-2-one-4’,6’,7’, 8’-tetrahydro-2’-quinazoline-dione (4a)

White powder; yield: 0.263 g (93%), m.p. >300°C (lit. 345–347°C [28]). – IR (KBr): \( \nu_{\text{max}} = 3379, 3348, 3201, 1709, 1645, 1629, 1525 \text{ cm}^{-1}. – ¹H NMR (500 MHz, DMSO-d₆): \( \delta = 2.20 (2 \text{ H, m, CH}_2), 3.03 (2 \text{ H, m, CH}_2), 3.49 (2 \text{ H, m, CH}_2), 6.95–7.53 (4 \text{ H, m, HAr}), 7.68 (1 \text{ H, s, HAr}), 8.21 (1 \text{ H, s, HAr}), 9.03 (1 \text{ H, s, NH}) \text{ ppm}. – ¹³C NMR (125 MHz, DMSO-d₆): \( \delta = 21.9, 27.1, 36.8, 82.5, 106.1, 114.3, 116.7, 120.4, 121.5, 123.2, 139.0, 157.7, 159.1, 165.3, 190.6 \text{ ppm}. – Analysis calcd for C₂₃H₁₇N₃O₃ (408.39): C 68.00, H 4.70, N 15.27; found C 67.93, H 4.74, N 15.30%

4.3.2 Spiro[3, 4’7]-1,3-dihydro-2H-indol-2-one-4’,6’,7’, 8’-tetrahydro-2’-quinazoline-dione (4b)

White powder; yield: 0.326 g (90%), m.p. >300°C. – IR (KBr): \( \nu_{\text{max}} = 3356, 3353, 3197, 1710, 1637, 1615, 1554 \text{ cm}^{-1}. – ¹H NMR (500 MHz, DMSO-d₆): \( \delta = 2.28 (2 \text{ H, m, CH}_2), 2.86 (2 \text{ H, m, CH}_2), 3.51 (2 \text{ H, m, CH}_2), 7.08–7.50 (2 \text{ H, m, HAr}), 7.71 (1 \text{ H, s, HAr}), 8.08 (1 \text{ H, s, HAr}), 8.16 (1 \text{ H, s, NH}), 9.12 (1 \text{ H, s, NH}) \text{ ppm}. – ¹³C NMR (125 MHz, DMSO-d₆): \( \delta = 22.3, 26.8, 36.1, 80.7, 105.6, 114.0, 116.8, 120.5, 122.4, 130.1, 141.2, 155.9, 158.5, 164.8, 191.1 \text{ ppm}. – Analysis calcd for C₂₃H₁₇BrN₃O₃ (362.18): C 49.74, H 3.34, N 11.60; found C 49.91, H 3.45, N 11.39%

4.3.3 Spiro[3, 4’7]-1,3-dihydro-5-methoxy-2H-indol-2-one- 4’,6’,7’, 8’-tetrahydro-2’-quinazoline-dione (4c)

White powder; yield: 0.288 g (92%), m.p. >300°C. – IR (KBr): \( \nu_{\text{max}} = 3356, 3334, 3183, 2967, 1714, 1640, 1622, 1546 \text{ cm}^{-1}. – ¹H NMR (500 MHz, DMSO-d₆): \( \delta = 2.28 (2 \text{ H, m, CH}_2), 2.98 (2 \text{ H, m, CH}_2), 3.42 (2 \text{ H, m, CH}_2), 3.63 (3 \text{ H, s, CH}_3), 7.08–7.50 (4 \text{ H, m, HAr}), 7.68 (1 \text{ H, s, HAr}), 8.21 (1 \text{ H, s, HAr}), 9.03 (1 \text{ H, s, NH}) \text{ ppm}. – ¹³C NMR (125 MHz, DMSO-d₆): \( \delta = 21.9, 27.1, 36.8, 82.5, 106.1, 114.3, 116.7, 120.4, 121.5, 123.2, 139.0, 157.7, 159.1, 165.3, 190.6 \text{ ppm}. – Analysis calcd for C₂₅H₁₉O₃BrN₃ (449.39): C 66.54, H 4.31, N 13.77; found C 66.64, H 4.28, N 13.69%
4.3.4 Spiro[3,4′]1,3-dihydro-5-nitro-2H-indol-2-one-4′,6′,7′,8′-tetrahydro-2′,5′(1′H,3′H)-quinazoline-dione (4d)

White powder; yield: 0.312 g (95%), m.p. >300°C. – IR (KBr): $\nu_{\text{max}}$ = 3361, 3352, 3204, 1719, 1639, 1569, 1555 cm⁻¹. – 1H NMR (500 MHz, DMSO-d₆): $\delta$ = 2.16 (2 H, m, CH₂), 3.01 (2 H, m, CH₂), 3.39 (2 H, m, CH₂), 7.38 (1 H, m, HAr), 7.59 (1 H, s, CH₃), 7.99–8.36 (2 H, m, HAr), 8.41 (1 H, s, NH), 9.26 (1 H, s, NH) ppm. – 13C NMR (125 MHz, DMSO-d₆): $\delta$ = 23.0, 27.5, 36.5, 82.2, 105.1, 117.2, 119.9, 123.7, 124.7, 139.8, 146.6, 156.4, 160.2, 164.2, 191.3 ppm. – Analysis calcd for C₁₇H₁₇N₃O₃ (311.34): C 65.58, H 5.59, N 13.55%.

4.3.5 Spiro[3,4′]1,3-dihydro-5-nitro-2H-indol-2-one-7′,7′-dimethyl-4′,6′,7′,8′-tetrahydro-2′,5′(1′H,3′H)-quinazoline-dione (4e)

White powder; yield: 0.293 g (94%), m.p. >300°C (lit. 350–352°C [28]). – IR (KBr): $\nu_{\text{max}}$ = 3376, 3285, 3167, 2905, 1720, 1678, 1615, 1533 cm⁻¹. – 1H NMR (500 MHz, DMSO-d₆): $\delta$ = 0.97 (3 H, s, CH₃), 1.01 (3 H, s, CH₃), 2.07 (2 H, m, CH₂), 2.38 (2 H, m, CH₂), 7.03–7.22 (4 H, m, HAr), 7.68 (1 H, s, NH), 9.40 (1 H, s, NH), 10.13 (1 H, s, NH) ppm. – 13C NMR (125 MHz, DMSO-d₆): $\delta$ = 27.8, 30.1, 37.6, 49.0, 79.0, 104.5, 115.4, 116.1, 120.6, 121.4, 122.3, 149.8, 158.7, 160.3, 168.5, 191.3 ppm. – Analysis calcd for C₁₇H₁₇N₃O₃ (311.34): C 65.58, H 5.50, N 13.50; found C 65.47, H 5.59, N 13.52%.

4.3.6 Spiro[3,4′]5-bromo-1,3-dihydro-5-nitro-2H-indol-2-one-7′,7′-dimethyl-4′,6′,7′,8′-tetrahydro-2′,5′(1′H,3′H)-quinazoline-dione (4f)

White powder; yield: 0.359 g (92%), m.p. >300°C. – IR (KBr): $\nu_{\text{max}}$ = 3357, 3307, 3143, 2881, 1716, 1686, 1630, 1545 cm⁻¹. – 1H NMR (500 MHz, DMSO-d₆): $\delta$ = 0.94 (3 H, s, CH₃), 1.02 (3 H, s, CH₃), 2.03 (1 H, d, $J$ = 15.9 Hz, CH₂), 2.14 (1 H, d, $J$ = 15.8 Hz, CH₂), 2.45 (2 H, m, CH₂), 7.41 (1 H, d, $J$ = 7.8 Hz, HAr), 7.58 (1 H, d, $J$ = 7.7 Hz, HAr), 7.65 (1 H, s, NH), 8.01 (1 H, m, HAr), 9.38 (1 H, s, NH), 10.11 (1 H, s, NH) ppm. – 13C NMR (125 MHz, DMSO-d₆): $\delta$ = 28.1, 30.5, 38.2, 48.6, 78.2, 109.7, 116.5, 120.1, 123.3, 124.4, 131.9, 149.8, 159.2, 161.2, 168.8, 193.2 ppm. – Analysis calcd for C$_{17}$H$_{17}$BrN$_3$O$_3$ (390.24): C 52.32, H 4.13, N 10.77; found C 52.20, H 4.32, N 10.91%.

4.3.7 Spiro[3,4′]1,3-dihydro-5-methoxy-2H-indol-2-one-7′,7′-dimethyl-4′,6′,7′,8′-tetrahydro-2′,5′(1′H,3′H)-quinazoline-dione (4g)

White powder; yield: 0.321 g (94%), m.p. >300°C. – IR (KBr): $\nu_{\text{max}}$ = 3380, 3290, 3152, 2898, 1723, 1665, 1627, 1529 cm⁻¹. – 1H NMR (500 MHz, DMSO-d₆): $\delta$ = 0.93 (3 H, s, CH₃), 1.05 (3 H, s, CH₃), 2.11 (2 H, m, CH₂), 2.39 (2 H, m, CH₂), 2.75 (3 H, s, OCH₃), 5.79–7.22 (2 H, m, HAr), 7.66 (1 H, s, NH), 9.37 (1 H, s, NH), 10.11 (1 H, s, NH) ppm. – 13C NMR (125 MHz, DMSO-d₆): $\delta$ = 28.5, 30.4, 37.5, 50.1, 56.7, 78.5, 111.7, 114.6, 116.8, 117.4, 123.9, 148.3, 152.0, 158.5, 160.1, 170.1, 192.4 ppm. – Analysis calcd for C$_{17}$H$_{17}$N$_3$O$_4$ (341.36): C 63.33, H 5.61, N 12.31; found C 63.14, H 5.48, N 12.13%.

4.3.8 Spiro[3,4′]1,3-dihydro-5-nitro-2H-indol-2-one-7′,7′-dimethyl-4′,6′,7′,8′-tetrahydro-2′,5′(1′H,3′H)-quinazoline-dione (4h)

White powder; yield: 0.346 g (97%), m.p. >300°C. – IR (KBr): $\nu_{\text{max}}$ = 3366, 3283, 3149, 2911, 1718, 1680, 1631, 1550 cm⁻¹. – 1H NMR (500 MHz, DMSO-d₆): $\delta$ = 0.99 (3 H, s, CH₃), 1.02 (3 H, s, CH₃), 2.02 (1 H, d, $J$ = 16.1 Hz, CH₂), 2.10 (1 H, d, $J$ = 16.1 Hz, CH₂), 2.37 (1 H, d, $J$ = 16.8 Hz, CH₂), 2.44 (1 H, d, $J$ = 16.6 Hz, CH₂), 1.756 (1 H, d, $J$ = 7.6 Hz, HAr), 7.65 (1 H, s, NH), 7.88 (1 H, d, $J$ = 7.4 Hz, HAr), 8.12 (1 H, s, HAr), 9.41 (1 H, s, NH), 10.14 (1 H, s, NH) ppm. – 13C NMR (125 MHz, DMSO-d₆): $\delta$ = 29.3, 32.1, 38.4, 50.9, 80.1, 104.3, 115.6, 120.1, 124.7, 125.6, 149.1, 150.1, 159.0, 160.3, 169.8, 193.2 ppm. – Analysis calcd for C$_{17}$H$_{17}$N$_3$O$_4$ (356.34): C 57.30, H 4.53, N 15.72; found C 57.19, H 4.63, N 15.88%.

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References