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SHORT COMMUNICATION

POTASSIUM 2-OXOIMIDAZOLIDINE-1,3-DIIDE: AN EFFECTIVE AND NEW CATALYST FOR THE GRINDING SYNTHESIS OF (1*H*-INDOL-3-YL)METHYL-2*H*-INDAN-1,3-DIONES

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ABSTRACT. Potassium 2-oxoimidazolidine-1,3-diide (POImD) is easily used as an efficient and recyclable catalyst for the synthesis of 2*H*-indene-1,3-dione derivatives by a reaction between indoles, 2*H*-indene-1,3-dione and aldehydes at room temperature. All reactions are performed in humid POImD in high to excellent yield during short reaction time. Further, the catalyst can be reused and recovered for several times without loss of activity.

KEY WORDS: 1,3-Indandione, Indole, POImD, One pot reaction

INTRODUCTION

In recent years, an increasing interest has been focused on the synthesis of 2-substituted 2*H*-indene-1,3-dione compounds owing to their significant biological activity [1, 2]. It is well known that indandione and related compounds exhibit a wide range of biological activities such as antitumor, anticancer, sedative, hypertensive and neuromuscular blocking activities [3-6]. The synthesis of various 2-substituted-1,3-indandiones has initially been stimulated by their known anticoagulant and pharmacological properties [7, 8]. In the other hand, the interest in 2*H*-indene-1,3-diones has moved from pure synthetic chemistry to the field of material science [9]. As a very strong electron acceptor, 2*H*-indene-1,3-dione is a part in new dipolar molecules exhibiting interesting optical properties and used as dopants in novel functional materials [10, 11]. A famous example is *N,N*-dimethylamino-benzylidene-1,3-indandione (DMABI) and its derivatives, whose photo physical and non-linear optical properties were extensively studied [12-14].

However, most of these reported methods for the synthesis of 2*H*-indene-1,3-diones suffer from environmental pollution, exotic reaction conditions, tedious preparation procedure, long reaction time, expensive reagents or catalysts, unsatisfactory yields and complicated operations. In order to make this reaction simple and green, herein, we used POImD to synthesis of 2-substituted-1,3-indandiones by the three-component reaction of indole, 2*H*-indene-1,3-dione and various arylaldehydes at room temperature in aqueous media (Scheme 1).

Water has a unique media in chemistry and is one of the best solvents, owing to its features such as being eco-friendly, clean, green, nontoxic, non-flammable, safe, low-cost and readily available in organic transformations. Also, the use of aqua media not only diminishes the risk entailed in the use of organic solvents but also improves the rate of many chemical reactions [15-18].

RESULTS AND DISCUSSION

As part of our ongoing interest for the development of efficient and environmentally friendly procedures for the synthesis of heterocyclic and pharmaceutical compounds [19-25], herein we

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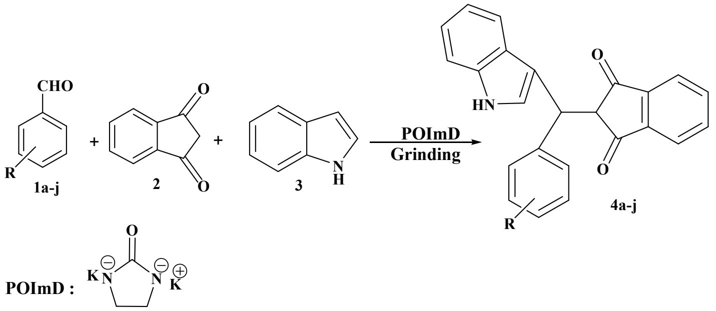
*Corresponding author. E-mail: nikpassand@iaurasht.ac.ir wish to report our experimental results on the synthesis of new generation of 2*H*-indene-1,3-dione using various aromatic aldehydes, indole, 1,3-indandione in the presence of novel catalyst (POImD) (Scheme 1) and the results were listed in Table 1. With the best catalyst in hand, we moved to study the effects of catalyst amount on the model reaction and the results were listed in Table 2. 0.02 mmol of POImD is sufficient to push the reaction forward completion and 0.01 mmol of catalyst was not enough. Higher amount of catalyst did not lead to significant change in the reaction yields.



Scheme 1. Synthesis of 2-((indol-3-ylmethyl)-1*H*-indene-1,3-diones using of POImD.

Table 1. Synthesis of 2-((indol-3-ylmethyl)-1*H*-indene-1,3-diones **4a-j** using POImD.

Entry	Product	R	Time (min)	Yield (%)^a
1	4a	4-Cl	30 (180)^b (720)^d	90 (70)^b (60)^d
2	4b	4-NO_2	25 (180)^b (180)^c	93 (68)^b (60)^c
3	4c	3-NO_2	30 (360)^b (360)^c	89 (69)^b (75)^c
4	4d	4-Br	30 (600)^d	90 (65)^d
5	4e	3-OH	45 (270)^b (240)^c	85 (80)^b (75)^c
6	4f	4-SCH_3	60	87
7	4g	pyridin-3-yl	30	86
8	4h	2-Cl-5-NO_2	25	89
9	4i	4-OCH_3	60 (300)^b (240)^c	83 (75)^b (75)^c
10	4j	4-$N(CH_3)_2$	60 (270)^b (480)^d	72 (80)^b (70)^c

[a]Isolated yield, [b]in the presence of [BMIM]Br, [c]in the presence of K10, [d]in a catalyst free reaction.

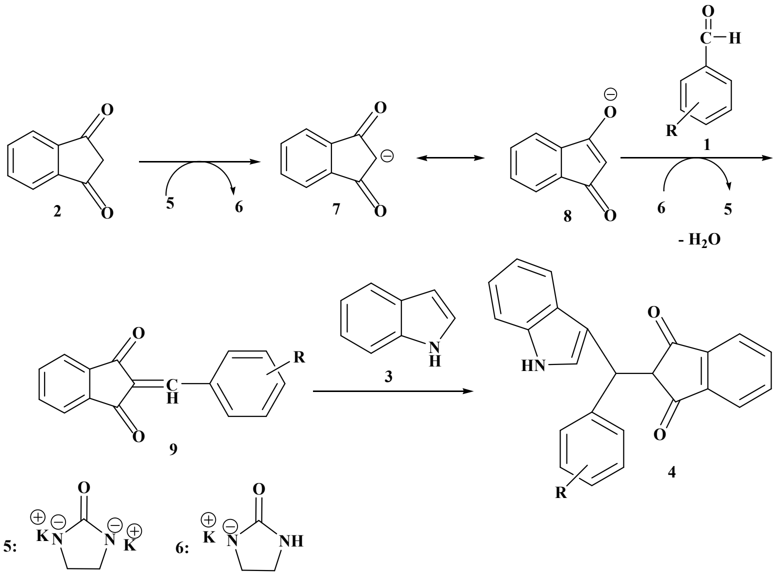
After reaction, the catalyst is easily separated from the reaction medium by washing with distilled water (catalyst is soluble in water). The washed catalyst is distilled under vacuum to recover solvent for reuse in subsequent reactions. Our experiments also indicated that after six runs, the catalytic activities of the reagents were almost the same as those of fresh catalysts.

Table 2. The effect amount of POImD when synthesizing **4a**.

Entry	Catalyst amount (mmol)	Time (min)	Yield (%)^a
1	0.01	120	37
2	0.02	30	90
3	0.03	30	89
4	0.04	30	90

[a]Isolated yield.

Several aromatic aldehydes could be converted to the corresponding products in good to high yield using POImD. Benzaldehyde and other aromatic aldehyde containing electron withdrawing groups (such as nitro, halide) or electron releasing groups (such as hydroxyl, alkoxyl group) were employed and reacted well to give the corresponding 2*H*-indene-1,3-diones in the yields ranging from 72 to 93% (Table 1). In a plausible mechanism, potassium 2-oxoimidazolidine-1,3-diide (POImD) (**5**) converted 1,3-indandione (**2**) to active form **7** or **8** by hydrogen abstraction. Then, the nucleophilic attack of C-2 anion **7** or **8** via resonance to arylaldehyde (**1**) leads to compound **9**. Finally, after nucleophilic addition of indole **3**, compound **9** was conformed to product **4**. It is mentionable that under this procedure catalyst POImD reversibly converts from **5** to **6** (Scheme 2).



Scheme 2. Proposed mechanism for synthesis of 2-((indol-3-ylmethyl)-1*H*-indene-1,3-diones using of POImD.

CONCLUSIONS

In conclusion, we develop an efficient and convenient procedure for the synthesis of 1,3-indandiones through three component synthesis of aldehydes, indole and 2*H*-indene-1,3-dione catalyzed by POImD as a reusable and catalyst. The remarkable advantages offered by this method are: catalyst is inexpensive, non-toxic, easy handling and reusable, simple work-up procedure, short reaction time, high yields of product with better purity and green aspect by avoiding toxic catalyst and hazardous solvent. Apart from the mild conditions of the process and its excellent results, the simplicity of product isolation and the possibility to recycle the catalyst offer a significant advantage. To the best of our knowledge this is the first report on synthesis of POImD.

EXPERIMENTAL

Chemicals were purchased from Merck and Fluka and used as instructed. Melting points were measured on an Electro-thermal 9100 apparatus and are uncorrected. NMR spectra were obtained on a Bruker DRX 400 Avance spectrometer in DMSO-d_6 and $CDCl_3$ as solvent and with TMS as internal standard. FT-IR spectra were recorded on a Shimadzu FT-IR-8400S spectrometer.

General procedure for the synthesis of 2-oxoimidazolidine-1,3-diide; POImD. A mixture of imidazolidin-2-one (20 mmol), KOH (20 mmol) and H_2O (10 mL) was stirred overnight. Following the completion of the reaction, as indicated by TLC, potassium 2-oxoimidazolidine-1,3-diide (POImD) was separated from the reaction mixture by filtration. POImD was purified by recrystallization from EtOH to afford pure products.

General procedure for preparation of 2H-indene-1,3-diones. A mixture containing aldehyde (1 mmol), indole (1 mmol), 1,3-indandione (1 mmol), 0.01 mmol of POImD and 5 drops of H_2O were ground at room temparature for the required reaction times. The progress of the reaction was monitored by TLC (EtOAc: petroleum ether 1:3). After completion of the reaction, the product was extracted with $CHCl_3/H_2O$ and preferred the organic phase. After evaporation and recrystallization, the pure product was obtained. The aqueous phase was evaporated also to produce recycled bis POImD.

*2-((4-Chlorophenyl)(1H-indol-3-yl)methyl)-2H-indene-1,3-dione (**4a**)*. Yellow-orange solid; mp 282-283 °C; IR (KBr) (ν_{max}, cm^{-1}): 3380, 1700, 1652, 1552, 1253 cm^{-1}; ^1H NMR ($CDCl_3$, 400 MHz) δ: 4.31 (d, $J = 2.4$ Hz, 1H), 5.14 (d, $J = 2.4$ Hz, 1H), 6.85 (td, $J = 8.0$ Hz, $J = 0.8$ Hz, 1H), 7.02 (td, $J = 8.0$ Hz, $J = 0.8$ Hz, 1H, ArH), 7.13 (d, $J = 8.0$ Hz, 1H, ArH), 7.20–7.25 (m, 4H, ArH), 7.30–7.35 (m, 3H, ArH), 7.85–7.92 (m, 3H, ArH), 10.96 (s, 1H, NH); ^{13}C NMR ($CDCl_3$, 100 MHz) δ: 37.2, 58.5, 111.9, 114.4, 119.0, 121.6, 123.2, 123.2, 124.8, 126.7, 126.7, 128.4, 130.8, 136.6, 136.6, 140.9, 142.6, 200.3 (C=O) ppm. Anal. calcd. for $C_{24}H_{16}ClNO_2$: C, 74.71; H, 4.18; N, 3.63. Found: C, 74.65; H, 4.21; N, 3.58.

*2-((1H-indol-3-yl)(4-nitrophenyl)methyl)-2H-indene-1,3-dione (**4b**)*. Off white solid, mp 217-219 °C; IR (KBr) (ν_{max}, cm^{-1}): 3379, 1701, 1591, 1510, 1348, 1257 cm^{-1}; ^1H NMR ($CDCl_3$, 400 MHz) δ: 4.45 (d, $J = 4.0$ Hz, 1H), 5.30 (d, $J = 4.0$ Hz, 1H), 6.88 (td, $J = 7.8$ Hz, $J = 0.8$ Hz, 1H, ArH), 7.02 (td, $J = 8.0$ Hz, $J = 1.2$ Hz, 1H, ArH), 7.17 (d, $J = 8.0$ Hz, 1H, ArH), 7.24 (d, $J = 2.4$ Hz, 1H), 7.30 (d, $J = 8.0$ Hz, 1H, ArH), 7.36 (s, 1H, ArH), 7.55 (d, $J = 7.8$ Hz, 2H, ArH), 7.85–7.95 (m, 3H, ArH), 8.07 (d, $J = 7.8$ Hz, 2H, ArH), 11.01 (s, 1H, NH) ppm. Anal. calcd. for $C_{24}H_{16}N_2O_4$: C, 72.72; H, 4.07; N, 7.07. Found: C, 72.67; H, 4.12; N, 7.11.

*2-((1H-indol-3-yl)(3-nitrophenyl)methyl)-2H-indene-1,3-dione (**4c**)*. Off white solid, mp 305-307 °C; IR (KBr) (ν_{max}, cm^{-1}): 3320, 1700, 1655, 1525, 1345, 1243 cm^{-1}; ^1H NMR ($CDCl_3$, 400 MHz) δ: 4.44 (d, $J = 3.0$ Hz, 1H), 5.34 (d, $J = 3.0$ Hz, 1H), 6.86 (t, $J = 8.0$ Hz, 1H, ArH), 7.00 (t, $J = 8.0$ Hz, 1H, ArH), 7.22–7.24 (m, 2H, ArH), 7.30 (d, $J = 8.4$ Hz, 1H, ArH), 7.35–7.36 (m, 1H, ArH), 7.52 (d, $J = 8.0$ Hz, 1H, ArH), 7.77 (d, $J = 8.0$ Hz, 1H, ArH), 7.86–7.94 (m, 3H, ArH), 8.0 (dd, $J = 8.4$ Hz, $J = 1.6$ Hz, 1H, ArH), 8.17 (d, $J = 2.0$ Hz, 1H, ArH), 10.99 (s, 1H, NH); ^{13}C NMR ($CDCl_3$, 100 MHz) δ: 35.6, 58.3, 112.0, 113.5, 119.0, 119.2, 121.8, 122.0, 123.3, 123.3, 123.6, 124.8, 130.0, 135.8, 136.4, 136.7, 142.4, 144.6, 147.9, 200.2 (C=O) ppm. Anal. calcd. for $C_{24}H_{16}N_2O_4$: C, 72.72; H, 4.07; N, 7.07. Found: C, 72.77; H, 4.00; N, 7.12.

*2-((4-Bromophenyl)(1H-indol-3-yl)methyl)-2H-indene-1,3-dione (**4d**)*. Off white solid, mp 269-271 °C; IR (KBr) (ν_{max}, cm^{-1}): 3392, 1702, 1650, 1597 cm^{-1}; ^1H NMR ($CDCl_3$, 400 MHz) δ: 4.31 (d, $J = 3.2$ Hz, 1H), 5.12 (d, $J = 3.2$ Hz, 1H), 6.83–6.87 (m, 1H), 6.99–7.03 (m, 1H, ArH), 7.12 (d, $J = 8.0$ Hz, 1H, ArH), 7.16 (d, $J = 8.4$ Hz, 2H, ArH), 7.30–7.32 (m, 2H, ArH), 7.34 (d, $J = 8.4$ Hz, 2H, ArH), 7.84–7.93 (m, 4H, ArH), 10.95 (s, 1H, NH); ^{13}C NMR ($CDCl_3$, 100 MHz) δ: 58.5, 78.8, 111.9, 114.3, 118.9, 119.0, 119.9, 121.6, 123.2, 124.7, 131.2, 131.2, 136.4, 136.5, 136.6, 141.4, 142.6, 200.3 (C=O) ppm. Anal. calcd. for $C_{24}H_{16}BrNO_2$: C, 66.99; H, 3.75; N, 3.26. Found: C, 67.05; H, 3.69; N, 3.24.

*2-((3-Hydroxyphenyl)(1H-indol-3-yl)methyl)-2H-indene-1,3-dione (**4e**)*. Light yellow solid, mp 311-313 °C; IR (KBr) (ν_{max}, cm^{-1}): 3720, 3454, 1700, 1630, 1550, 1354, 1270 cm^{-1}; ^1H NMR

($CDCl_3$, 400 MHz) δ: 4.20 (s, 1H), 5.04 (s, 1H), 6.41 (dd, *J* = 8.0 Hz, *J* = 2.4Hz, 1H), 6.57 (t, *J* = 6.0 Hz, 1H), 6.61 (d, *J* = 8.0 Hz, 1H), 6.83–6.92 (m, 2H), 7.00–7.04 (m, 1H, ArH), 7.13 (d, *J* = 7.6 Hz, 1H, ArH), 7.31 (d, *J* = 8.0 Hz, 1H, ArH), 7.41 (d, *J* = 2.0 Hz, 1H, ArH), 7.83–7.92 (m, 4H, ArH), 10.92 (s, 1H, OH), 10.93 (s, 1H, NH); ^{13}C NMR ($CDCl_3$, 100 MHz) δ: 30.1, 58.7, 111.8, 113.9, 115.1, 116.1, 118.9, 119.0, 119.6, 121.5, 123.2, 124.8, 127.0, 129.2, 136.4, 142.4, 142.8, 143.1, 157.2, 200.5 (C=O) ppm. Anal. calcd. for $C_{24}H_{17}NO_3$: C, 78.46; H, 4.66; N, 3.81; Found: C, 78.40; H, 4.72; N, 3.86.

*2-((1H-indol-3-yl)(4-(methylthio)phenyl)methyl)-2H-indene-1,3-dione (**4f**)*. Off white solid, mp 275-277 °C; IR (KBr) (ν_{max}, cm^{-1}): 3367, 1701, 1593, 1488, 1344, 1261 (aromatic C-S stretch) cm^{-1}; ^1H NMR ($CDCl_3$, 400 MHz) δ: 2.34 (s, 3H), 4.27 (d, *J* = 2.6 Hz, 1H), 5.10 (d, *J* = 2.6 Hz, 1H), 6.85 (t, *J* = 7.2 Hz, 1H), 7.0 (d, *J* = 8.0 Hz, 2H, ArH), 7.13 (d, *J* = 8.0 Hz, 2H, ArH), 7.30 (d, *J* = 8.0 Hz, 1H, ArH), 7.37 (s, 1H, ArH), 7.86 (d, *J* = 5.6 Hz, 2H, ArH), 7.91 (d, *J* = 4.4 Hz, 2H, ArH), 10.94 (s, 1H, NH); ^{13}C NMR ($CDCl_3$, 100 MHz) δ: 14.8, 37.2, 58.7, 111.9, 114.9, 119.0, 121.5, 123.2, 124.7, 125.8, 125.8, 126.8, 129.6, 136.4, 136.5, 138.4, 142.4, 200.4 (C=O) ppm. Anal. calcd. for $C_{25}H_{19}NO_2S$: C, 75.54; H, 4.82; N, 3.52. Found: C, 75.60; H, 4.78; N, 3.59.

*2-((1H-indol-3-yl)(pyridin-3-yl)methyl)-2H-indene-1,3-dione (**4g**)*. light yellow solid; IR (KBr) (ν_{max}, cm^{-1}): 3134, 1703, 1585, 1425 cm^{-1}; ^1H NMR ($CDCl_3$, 400 MHz) δ: 4.38 (d, *J* = 2.8 Hz, 1H), 5.21 (d, *J* = 2.8 Hz, 1H), 6.85 (t, *J* = 7.2 Hz, 1H), 7.00 (t, *J* = 7.2 Hz, 1H), 7.19–7.23 (m, 2H), 7.27 (d, *J* = 1.6 Hz, 1H), 7.30 (d, *J* = 7.2 Hz, 2H), 7.64 (d, *J* = 7.6 Hz, 1H), 7.87–7.94 (m, 3H), 8.31 (d, *J* = 7.6 Hz, 1H), 8.50 (s, 1H), 10.99 (s, 1H, NH); Anal. calcd. for $C_{23}H_{16}N_2O_2$: C, 78.39; H, 4.58; N, 7.95; Found: C, 78.43; H, 4.51; N, 8.00.

*2-((2-Chloro-5-nitrophenyl)(1H-indol-3-yl)methyl)-2H-indene-1,3-dione (**4h**)*. light yellow solid; IR (KBr) (ν_{max}, cm^{-1}): 3367, 1697, 1575, 1340, 1259 cm^{-1}; ^1H NMR ($CDCl_3$, 400 MHz) δ: 4.33 (d, *J* = 3.6 Hz, 1H), 5.64 (d, *J* = 3.6 Hz, 1H), 6.87 (d, *J* = 2.4 Hz, 1H), 7.00 (t, *J* = 5.2 Hz, 1H, ArH), 7.23 (d, *J* = 8.0 Hz, 1H, ArH), 7.28 (d, *J* = 8.0 Hz, 1H, ArH), 7.37 (s, 1H, ArH), 7.77 (d, *J* = 8.8 Hz, 1H, ArH), 7.91–7.97 (m, 4H, ArH), 8.13 (dd, *J* = 8.8 Hz, *J* = 2.8 Hz, 1H, ArH), 8.61 (d, *J* = 2.8 Hz, 1H, ArH), 10.96 (s, 1H, NH) ppm. Anal. Calcd. For $C_{24}H_{15}ClN_2O_4$: C, 66.91; H, 3.51; N, 6.50; Found: C, 67.00; H, 3.56; N, 6.55.

*2-((1H-indol-3-yl)(4-methoxyphenyl)methyl)-2H-indene-1,3-dione (**4i**)*. Off white solid, mp 270-272 °C; IR (KBr) (ν_{max}, cm^{-1}): 3335, 1712, 1576, 1492, 1340, 1210 (aromatic C-O stretch) cm^{-1}; ^1H NMR ($CDCl_3$, 400 MHz) δ: 3.49 (s, 3H), 4.31 (d, *J* = 2.7 Hz, 1H), 5.18 (d, *J* = 2.8 Hz, 1H), 6.92 (t, *J* = 7.4 Hz, 1H, ArH), 7.11 (d, *J* = 8.2 Hz, 2H, ArH), 7.27 (d, *J* = 8.2 Hz, 2H, ArH), 7.41 (d, *J* = 8.2 Hz, 1H, ArH), 7.50 (s, 1H, ArH), 7.81 (d, *J* = 7.0 Hz, 2H, ArH), 7.98 (d, *J* = 6.2 Hz, 2H, ArH), 11.09 (s, 1H, NH); ^{13}C NMR ($CDCl_3$, 100 MHz) δ: 15.5, 38.2, 62.1, 110.0, 113.2, 118.1, 122.0, 122.9, 125.7, 125.8, 127.1, 128.8, 129.5, 136.1, 136.3, 138.4, 148.1, 199.1 (C=O) ppm. Anal. calcd. for $C_{25}H_{19}NO_3$: C, 78.72; H, 5.02; N, 3.67. Found: C, 78.68; H, 4.98; N, 3.69.

*2-((4-(Dimethylamino)phenyl)(1H-indol-3-yl)methyl)-2H-indene-1,3-dione (**4j**)*. Off white solid, mp 280-282 °C; IR (KBr) (ν_{max}, cm^{-1}): 3325, 1689, 1573, 1489, 1340 cm^{-1}; ^1H NMR ($CDCl_3$, 400 MHz) δ: 2.42 (s, 6H), 4.20 (d, *J* = 2.4 Hz, 1H), 5.21 (d, *J* = 2.6 Hz, 1H), 6.80 (t, *J* = 7.4 Hz, 1H, ArH), 7.20 (d, *J* = 8.2 Hz, 2H, ArH), 7.32 (d, *J* = 8.2 Hz, 2H, ArH), 7.39 (d, *J* = 8.2 Hz, 1H, ArH), 7.41 (s, 1H, ArH), 7.78 (m, 2H, ArH), 7.91 (d, *J* = 6.2 Hz, 2H, ArH), 10.31 (s, 1H, NH); ^{13}C NMR ($CDCl_3$, 100 MHz) δ: 15.2, 37.1, 43.7, 112.9, 113.7, 119.0, 122.1, 123.1, 124.2, 125.0, 125.8, 127.1, 129.0, 136.0, 137.2, 138.8, 146.1, 199.2 (C=O) ppm. Anal. calcd. for $C_{26}H_{22}N_2O_2$: C, 79.16; H, 5.62; N, 7.10. Found: C, 79.23; H, 5.67; N, 7.02.

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