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A highly efficient CuI nanoparticles-catalyzed synthesis of tetrahydrochromenediones and dihydropyrano[*c*]chromenediones under grinding

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Abstract: A concise and CuI nanoparticle-catalyzed synthesis of tetrahydrochromenediones and dihydropyrano[*c*]chromenediones under solvent-free grinding conditions via the three-component condensation reaction of Meldrum's acid, an aromatic aldehyde, and an active methylene compound, including dimedone or 4-hydroxycoumarin, was developed. This new method gives desirable advantages such as simple reaction setup, very mild reaction conditions, production of the desired products in high yields without by-products, recyclability of the catalyst, and environmentally benign procedure.

Keywords: CuI nanoparticles; dihydropyrano[*c*]chromenediones; environmentally benign procedure; solvent-free grinding; tetrahydrochromenediones.

1 Introduction

The utility of nanostructured metal salts as catalysts in organic synthesis lies in their better properties compared to the bulk size [1, 2]. Nanosized copper iodide is one reusable Lewis acid catalyst, which has revealed several catalytic activities for the synthesis of organic compounds and others [3–12].

Chromene is a prominent structural motif found in various natural and nonnatural products, which have been reported to show a wide spectrum of useful biological properties such as antioxidant [13, 14], anticancer [15–18], antimicrobial [19–22], hypotensive [23], and local anesthetic [24]. In addition, they can be used as cognitive enhancers [25, 26], for the treatment of neurodegenerative diseases, including Alzheimer's disease [27] and schizophrenia disorder [28].

We have recently developed a series of heterocyclic syntheses using nanostructured catalysts [29–31]. Herein, we report a highly efficient and practical method for the synthesis of 4-aryl-7,7-dimethyl-4,6,7,8-tetrahydro-2*H*-chromene-2,5(3*H*)-diones (**4a–g**) and 4-aryl-3,4-dihydro-2*H*,5*H*-pyrano[3,2-*c*]chromene-2,5-diones (**6a–g**) via a condensation reaction of Meldrum's acid, aromatic aldehydes, and active methylene compounds, including dimedone or 4-hydroxycoumarin, catalyzed by CuI nanoparticles (CuI NPs) under solvent-free grinding (Scheme 1). In view of the above-mentioned useful properties of chromenes, various synthetic methods have been developed for the preparation of these compounds [32–36]. To the best of our knowledge, a synthesis of tetrahydrochromenediones and dihydropyrano[*c*]chromenediones catalyzed by CuI NPs has not been reported in the literature as yet.

2 Results and discussion

To optimize the reaction condition, benzaldehyde (**2a**) was chosen as the representative starting material in a typical reaction with Meldrum's acid (**1**) and dimedone (**3**) in the presence of CuI NPs under grinding to synthesize the corresponding chromenedione **4a** (Scheme 2 and Table 1).

Different amounts of catalyst were examined first. After 15 min with 5, 10 and 15 mol% of CuI NPs, yields of 38%, 92%, and 90%, respectively, were obtained. It is important to note that in the absence of CuI NPs, only a trace amount of product was found after 30 min (Table 1, entries 1–4).

When the grinding time was increased from 10 to 15 min, the yield of product **4a** was increased. However, further increase of the grinding time to 20 min failed to improve the yield of product **4a** (Table 1, entries 3 and 5–6).

After the completion of the model reaction, the catalyst was recovered from the reaction mixture by a procedure

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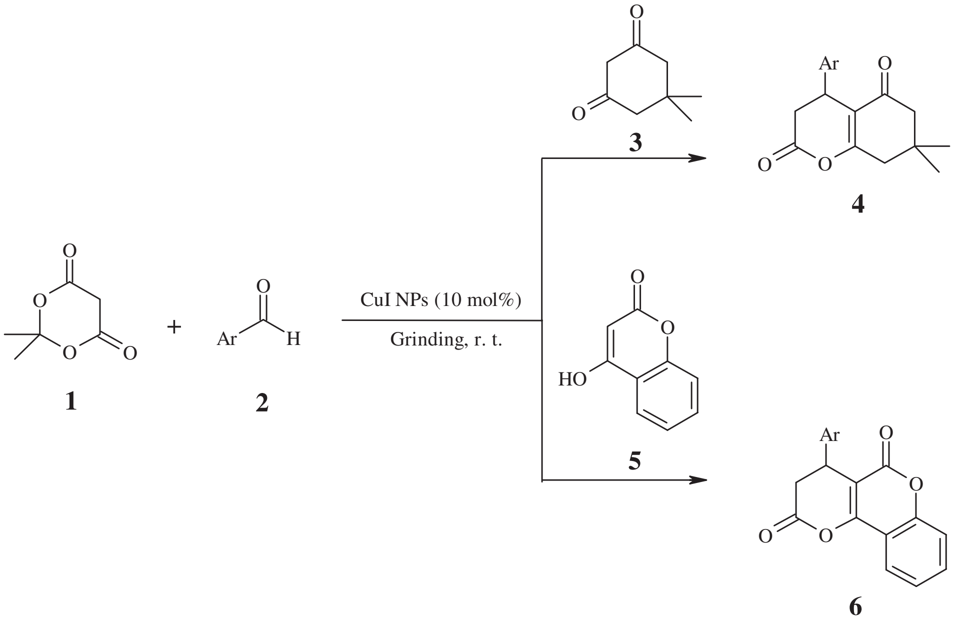
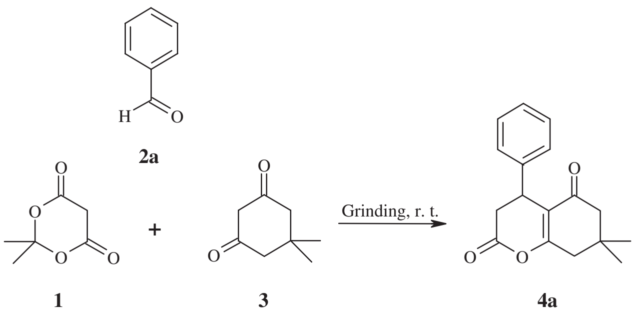


Scheme 1: Synthetic routes to chromenediones **4a–g** and pyrano[3,2-*c*]chromene-2,5-diones **6a–g**.



Scheme 2: Optimization of reaction conditions in the synthesis of chromenedione **4a**.

Table 1: Effect of different amounts of CuI NPs and grinding time on the synthesis of chromenedione **4a**.

Entry	CuI NPs (mol%)	Grinding time (min)	Yield (%)[a]
1	0	30	Trace
2	5	15	38
3	10	15	91
4	15	15	90
5	10	10	79
6	10	20	91

[a] Isolated yield.

described in the Experimental section and reused up to four times without significant loss of catalytic potential (product yields: 91%, 90%, 88%, and 85%, respectively).

The CuI NPs that were used as catalyst in this work were prepared by a literature procedure developed by Tavakoli et al. [37]. The X-ray powder diffraction (XRD) pattern and the scanning electron microscope (SEM) image of the synthesized CuI NPs have been presented in a previously published article [38]. The XRD pattern showed sharp peaks for CuI NPs matching with the standard cubic structure of crystalline CuI (cubic space group $F\bar{4}3m$, $a = 6.0545$ Å: JCPDS 82-2111). The SEM result also showed that the CuI NPs were triangle-shaped with a size range of 30–40 nm (see Abdolmohammadi and Karimpour [38] for an SEM image of the thus prepared CuI NPs).

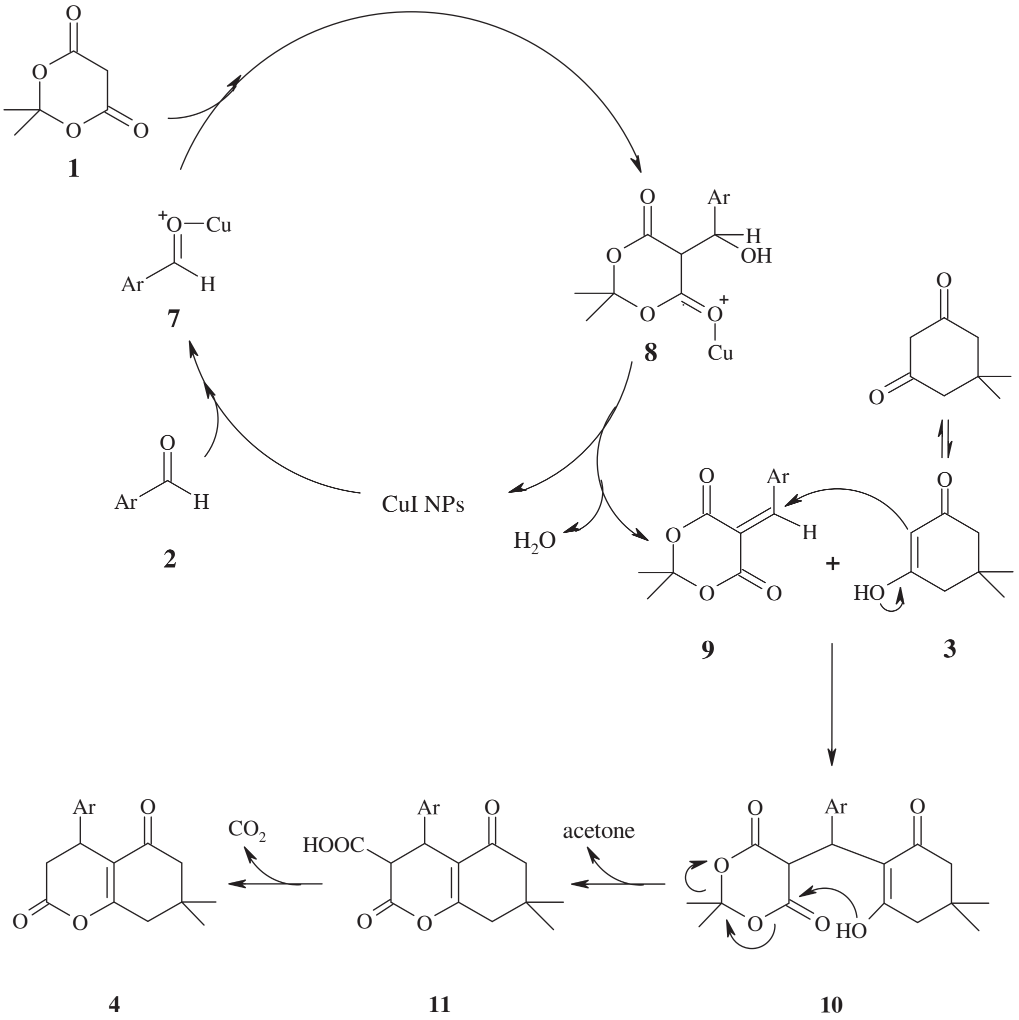
Different types of aromatic aldehydes carrying either electron donating or withdrawing substituents and dimedone or 4-hydroxycoumarin as active methylene compounds with Meldrum's acid as a model substrate were then explored under the optimized reaction conditions (Table 2), and high yields of desired products were obtained.

According to the proposed mechanism (Scheme 3), this catalytic protocol can be explained by a Knoevenagel-Michael sequence reaction. CuI NP is an effective catalyst for the formation of alkene **9**, readily prepared *in situ* from the Knoevenagel condensation of Meldrum's acid **1** and aromatic aldehyde **2**, which proceed via carbocation **7** and then intermediate **8**. The enol form of dimedone **3** then adds to alkene **9** in a Michael-type addition to generate intermediate **10**. The cyclization of **10** followed by the elimination of acetone gives product **4** after the decarboxylation of intermediate **11**. The formation of pyranochromenediones **6** can be explained analogously.

Table 2: Synthesis of chromene-2,5(3*H*)-diones **4a–g** and pyrano[3,2-*c*]chromene-2,5-diones **6a–g** catalyzed by CuI NPs.

Product	Ar	Active methylene compound	Yield (%)[a,b]	M.p. (°C)	
				Observed	Literature
4a	C_6H_5	**3**	91	101–103	99–101 [34]
4b	$2\text{-Cl-}C_6H_4$	**3**	94	131–133	134–135 [34]
4c	$4\text{-Cl-}C_6H_4$	**3**	95	156–158	160–161 [34]
4d	$4\text{-}(CH_3)_2N\text{-}C_6H_4$	**3**	89	141–143	138–140 [34]
4e	$3\text{-HO-}C_6H_4$	**3**	94	182–184	–
4f	$4\text{-}CH_3O\text{-}C_6H_4$	**3**	93	127–129	126–127 [34]
4g	$3\text{-}O_2N\text{-}C_6H_4$	**3**	96	163–165	–
6a	C_6H_5	**5**	97	168–170	169–171 [35, 36]
6b	$2\text{-Cl-}C_6H_4$	**5**	98	210–212	212–214 [35, 36]
6c	$4\text{-Cl-}C_6H_4$	**5**	93	196–197	197–199 [35, 36]
6d	$4\text{-}(CH_3)_2N\text{-}C_6H_4$	**5**	96	196–198	–
6e	$3\text{-HO-}C_6H_4$	**5**	94	184–186	–
6f	$4\text{-}CH_3O\text{-}C_6H_4$	**5**	93	146–147	143–145 [35, 36]
6g	$3\text{-}O_2N\text{-}C_6H_4$	**5**	97	181–183	–

[a] Yields refer to those of pure isolated products characterized by IR, ^1H NMR and ^{13}C NMR spectroscopy, and elemental analyses.
[b] In all cases, the reaction time was 15 min under solvent-free grinding.



Scheme 3: The possible reaction mechanism.

The structures of the compounds were confirmed by their satisfactory elemental analyses, infrared (IR), and ^{1}H and ^{13}C NMR spectroscopy. Selected spectroscopic data are given in the Experimental section. The synthesized catalyst was fully characterized by XRD and SEM techniques.

3 Conclusion

We have reported a simple route for the synthesis of tetrahydrochromenediones and dihydropyrano[*c*]chromenediones under solvent-free conditions using a grinding technique promoted by CuI NPs. Compared with known methods [34–36], this new protocol is endowed with high yields of products, short reaction time, reusability of catalyst, mild reaction conditions, and very simple operation.

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 an Electrothermal 9100 apparatus and are uncorrected (Islamic Azad University, East Tehran Branch, Tehran, Iran). Fourier transform infrared spectra were obtained using a Bruker Equinox 55 Golden Gate Micro-ATR spectrometer (Chemistry and Chemical Engineering Research Center of Iran, Tehran, Iran). ^{1}H NMR and ^{13}C NMR spectra were recorded on a Bruker DRX-500 AVANCE at 500 and 125 MHz, respectively, using tetramethylsilane (TMS) as internal standard and [D_6] dimethylsulfoxide (DMSO) as a solvent (Sharif University of Technology, Tehran, Iran). Elemental analyses were conducted using a Foss-Heraeus CHN-O-Rapid analyzer (Polymer and Petrochemical Institute, Tehran, Iran). The microscopic morphology of the catalyst was visualized by an SEM LEO 1455VP (Chemistry and Chemical Engineering Research Center of Iran, Tehran, Iran). Powder X-ray diffraction data were determined on a Philips X'Pert diffractometer using CuK_α radiation ($\lambda = 1.54$ Å) (Chemistry and Chemical Engineering Research Center of Iran, Tehran, Iran).

4.2 General procedure for the preparation of CuI NPs

$Cu(NO_3)_2 \cdot 3H_2O$ was used as the Cu source for the preparation of the catalyst by a precipitation approach. Initially, the D-(+)-glucose solution was added dropwise into the solution of copper nitrate for the reduction of the Cu^{+2} to Cu^+ under vigorous stirring, and then the LiI solution was added into the previously mentioned solution. The mixture was stirred for approximately 30 min at room temperature. After the completion of the reaction, a gray precipitate was obtained. The solid product was filtered and washed with deionized water and absolute EtOH, respectively, and finally dried in vacuo for several hours to give the pure catalyst [37].

4.3 General procedure for the synthesis of compounds 4a–g and 6a–g

Meldrum's acid (**1**, 1 mmol), aromatic aldehydes **2** (1 mmol), active methylene compounds **3** or **5** (1 mmol), and CuI NPs (1.9 mg, 10 mol%) were placed in a mortar. The mixture was ground with a mortar and pestle at room temperature for 15 min. After the completion of the reaction indicated by TLC, the resulting mixture was diluted with dimethylformadide (DMF) (5 mL) and then centrifuged for 5 min at 2000 rpm to recover the catalyst. The organic solution was then poured into ice-cold water (10 mL) to give a solid precipitate, which was filtered off and recrystallized from EtOH (95%) to afford the pure product.

4.4 Selected spectroscopic and physical data

4.4.1 4-Phenyl-7,7-dimethyl-4,6,7,8-tetrahydro-2*H*-chromene-2,5(3*H*)-dione (4a)

White solid, yield 0.246 g (91%), m. p. 101–103°C (lit: 99–101°C [34]). – IR (KBr, cm^{-1}): $\nu_{max} = 3034, 1768, 1660, 1511, 1120$. – ^{1}H NMR: $\delta = 1.09$ (s, 3H, CH_3), 1.13 (s, 3H, CH_3), 2.29 (d, 1H, H-8, $^2J_{HH} = 16.0$ Hz) 2.42 (d, 1H, H-8, $^2J_{HH} = 16.0$ Hz), 2.52 (s, 2H, H-6), 2.96 (d, 1H, H-3, $^2J_{HH} = 12.0$ Hz,), 3.02 (d, 1H, H-3, $^2J_{HH} = 12.0$ Hz), 4.30 (m, 1H, H-4), 7.22 (m, 5H, H-Ar) ppm. – ^{13}C NMR: $\delta = 26.1$ (CH_3), 29.3 (CH_3), 32.0 (CH_2), 32.8 (CH_2), 35.7 (CH_2), 40.8 (CH), 51.0 (C), 106.6 (C), 122.5 (CH), 125.8 (2CH), 129.8 (2CH), 143.1 (C), 161.6 (C=O), 163.7 (C), 196.3 (C=O) ppm. – Analysis for $C_{17}H_{18}O_3$ (270.33): calcd. C 75.53, H 6.71; found C 75.81, H 6.55%.

4.4.2 4-(2-Chlorophenyl)-7,7-dimethyl-4,6,7,8-tetrahydro-2*H*-chromene-2,5(3*H*)-dione (4b)

Lemon solid, yield 0.286 g (94%), m. p. 131–133°C (lit: 134–135 [34]). – IR (KBr, cm^{-1}): $\nu_{max} = 3046, 1755, 1697, 1534,$

1083. – ^{1}H NMR: $\delta = 1.14$ (s, 3H, CH_3), 1.16 (s, 3H, CH_3), 2.41 (s, 2H, H-8), 2. 58 (d, 1H, H-6, $^2J_{HH} = 17.4$ Hz), 2.62 (d, 1H, H-6, $^2J_{HH} = 17.4$ Hz), 2.95 (m, 2H, H-3), 4.73 (s, 1H, H-4), 7.24 (m, 4H, H-Ar) ppm. – ^{13}C NMR: $\delta = 26.8$ (CH_3), 28.7 (CH_3), 32.5 (CH_2), 33.1 (CH_2), 36.0 (CH_2), 41.2 (CH), 50.4 (C), 107.3 (C), 126.6 (CH), 128.3 (CH), 129.0 (CH), 130.1 (CH), 133.6 (C), 137.1 (C), 160.4 (C=O), 164.1 (C), 198.0 (C=O) ppm. – Analysis for $C_{17}H_{17}ClO_3$ (304.77): calcd. C 67.0, H 5.62; found C 66.83, H 5.48%.

4.4.3 4-(3-Hydroxyphenyl)-7,7-dimethyl-4,6,7,8-tetrahydro-2*H*-chromene-2,5(3*H*)-dione (4e)

Yellow solid, yield 0.269 g (94%), m. p. 182–184°C. – IR (KBr, cm^{-1}): $\nu_{max} = 3395$, 3052, 1764, 1709, 1558, 1096. – ^{1}H NMR: $\delta = 0.94$ (s, 3H, CH_3), 1.04 (s, 3H, CH_3), 2.19 (s, 2H, H-8), 2.41 (s, 2H, H-6), 2.63 (m, 2H, H-3), 4.64 (m, 1H, H-4), 7.19 (m, 2H, H-Ar), 7.32 (m, 2H, H-Ar), 9.05 (s, 1H, OH) ppm. – ^{13}C NMR: $\delta = 27.0$ (CH_3), 29.2 (CH_3), 31.9 (CH_2), 32.2 (CH_2), 37.1 (CH_2), 41.7 (CH), 50.7 (C), 105.7 (C), 114.2 (CH), 115.9 (CH), 117.0 (CH), 121.7 (CH), 140.2 (C), 146.3 (C), 160.5 (C=O), 164.5 (C), 198.2 (C=O) ppm. – Analysis for $C_{17}H_{18}O_4$ (286.33): calcd. C 71.31, H 6.34; found C 71.10, H 6.17%.

4.4.4 4-(3-Nitrophenyl)-7,7-dimethyl-4,6,7,8-tetrahydro-2*H*-chromene-2,5(3*H*)-dione (4g)

Pale yellow solid, yield 0.303 g (96%), m. p. 163–165°C. – IR (KBr, cm^{-1}): $\nu_{max} = 3085$, 1691, 1666, 1623, 1131. – ^{1}H NMR: $\delta = 1.02$ (s, 3H, CH_3), 1.16 (s, 3H, CH3), 2.07 (d, 1H, H-8, $^2J_{HH} = 15.8$ Hz), 2.12 (d, 1H, H-8, $^2J_{HH} = 15.8$ Hz), 2.38 (s, 2H, H-6), 2.62 (m, 2H, H-3), 4.54 (m, 1H, H-4), 7.33 (m, 1H, H-Ar), 8.92 (m, 3H, H-Ar) ppm. – ^{13}C NMR: $\delta = 27.2$ (CH_3), 28.9 (CH_3), 32.2 (CH_2), 33.0 (CH_2), 36.4 (CH_2), 42.4 (CH), 51.2 (C), 106.2 (C), 121.0 (CH), 125.2 (CH), 130.7 (CH), 129.6 (CH), 141.7 (C), 145.1 (C), 162.3 (C=O), 164.2 (C), 196.7 (C=O) ppm. – Analysis for $C_{17}H_{17}NO_5$ (315.32): calcd. C 64.75, H 5.43, N 4.44; found C 64.53, H 5.55, N 4.27%.

4.4.5 4-Phenyl-3,4-dihydro-2*H*,5*H*-pyrano[3,2-*c*]chromene-2,5-dione (6a)

White powder, yield 0.290 g (97%), m. p. 168–170°C (lit: 169–171 [35, 36]). – IR (KBr, cm^{-1}): $\nu_{max} = 1784$, 1702, 1659, 1087. – ^{1}H NMR: $\delta = 3.13$ (d, 1H, H-3, $^2J_{HH} = 16.1$ Hz), 3.20 (d, 1H, H-3, $^2J_{HH} = 16.1$ Hz), 4.52 (s, 1H, H-4), 7.28 (m, 3H, H-Ar), 7.29 (d, 1H, H-Ar, $^3J_{HH} = 7.0$ Hz), 7.34 (d, 1H, H-Ar, $^3J_{HH} = 7.0$ Hz), 7.39 (m, 2H, H-Ar), 7.61 (t, 1H, H-Ar, $^3J_{HH} = 7.2$ Hz), 7.91 (d, 1H, H-Ar, $^3J_{HH} = 7.4$ Hz) ppm. – ^{13}C NMR: $\delta = 36.8$ (CH_2), 37.1 (CH), 105.6 (C), 113.2 (C), 116.9 (CH), 122.8 (CH), 126.3 (CH), 128.1 (2CH), 128.8 (CH), 130.3 (2CH), 133.2 (CH), 141.2 (C), 154.1 (C), 156.7 (C), 160.1 (C=O), 163.9 (C=O) ppm. – Analysis for $C_{18}H_{12}O_4$ (299.29): calcd. C 72.24, H 4.04; found C 72.19, H 4.12%.

4.4.6 4-(Dimethylaminophenyl)-3,4-dihydro-2*H*,5*H*-pyrano[3,2-*c*]chromene-2,5-dione (6d)

White powder, yield 0.322 g (96%), m. p. 196–198°C. – IR (KBr, cm^{-1}): $\nu_{max} = 1769$, 1694, 1667, 1082. – ^{1}H NMR: $\delta = 3.04$ (s, 6H, 2NCH3), 3.09 (d, 1H, H-3, $^2J_{HH} = 15.8$ Hz), 3.17 (d, 1H, H-3, $^2J_{HH} = 15.8$ Hz), 4.48 (s, 1H, H-4), 6.87 (d, 2H, H-Ar, $^3J_{HH} = 8.0$ Hz), 7.26 (d, 2H, H-Ar, $^3J_{HH} = 8.3$ Hz), 7.35 (m, 2H, H-Ar), 7.48 (t, 1H, H-Ar, $^3J_{HH} = 8.2$ Hz), 7.80 (d, 1H, H-Ar, $^3J_{HH} = 8.6$ Hz) ppm. – ^{13}C NMR: $\delta = 35.7$ (CH2), 36.8 (CH), 45.4 (NCH_3), 104.7 (C), 112.9 (C), 114.5 (2CH), 115.9 (CH), 123.2 (CH), 124.7 (CH), 127.4 (2CH), 129.3 (C), 132.3 (CH), 149.8 (C), 152.5 (C), 159.2 (C), 161.2 (C=O), 164.4 (C=O) ppm. – Analysis for $C_{20}H_{17}NO_4$ (335.36): calcd. C 71.63, H 5.11, N 4.18; found C 71.59, H 5.28, N 4.27%.

4.4.7 4-(3-Hydroxyphenyl)-3,4-dihydro-2*H*,5*H*-pyrano[3,2-*c*]chromene-2,5-dione (6e)

White powder, yield 0.290 g (94%), m. p. 184–186°C. – IR (KBr, cm^{-1}): $\nu_{max} = 3324$, 1753, 1714, 1642, 1113. – ^{1}H NMR: $\delta = 3.12$ (d, 1H, H-3, $^2J_{HH} = 16.0$ Hz), 3.23 (d, 1H, H-3, $^2J_{HH} = 16.0$ Hz), 4.45 (s, 1H, H-4), 7. 19 (m, 2H, H-Ar), 7. 32 (m, 2H, H-Ar), 7.36 (m, 2H, H-Ar), 7.64 (t, 1H, H-Ar, $^3J_{HH} = 7.8$ Hz), 7.78 (d, 1H, H-Ar, $^3J_{HH} = 7.8$ Hz), 9.45 (s, 1H, OH) ppm. – ^{13}C NMR: $\delta = 35.3$ (CH2), 36.2 (CH), 106.1 (C), 112.5 (CH), 113.3 (C), 114.6 (CH), 115.4 (CH), 120.1 (CH), 122.3 (CH), 123.5 (CH), 125.6 (CH), 133.2 (CH), 139.1 (C), 154.1 (C), 155.6 (C), 158.7 (C), 160.8 (C=O), 163.9 (C=O) ppm. – Analysis for $C_{18}H_{12}O_5$ (308.29): calcd. C 70.13, H 3.92; found C 69.97, H 4.01%.

4.4.8 4-(4-Methoxyphenyl)-3,4-dihydro-2*H*,5*H*-pyrano[3,2-*c*]chromene-2,5-dione (6f)

White powder, yield 0.30 g (93%), m. p. 146–147°C (lit: 143–145 [35, 36]). – IR (KBr, cm^{-1}): $\nu_{max} = 1774$, 1705, 1668, 1102. – ^{1}H NMR: $\delta = 3.10$ (d, 1H, H-3, $^2J_{HH} = 16.0$ Hz), 3.19 (d, 1H, H-3, $^2J_{HH} = 16.0$ Hz), 3.71 (s, 3H, OCH_3), 4.50 (s, 1H, H-4), 6.79 (d, 2H, H-Ar, $^3J_{HH} = 8.4$ Hz), 7.17 (d, 2H, H-Ar, $^3J_{HH} = 8.4$ Hz), 7.37 (m, 2H, H-Ar), 7.69 (t, 1H, H-Ar, $^3J_{HH} = 8.0$ Hz), 7.88 (d, 1H,H-Ar, $^3J_{HH} = 8.1$ Hz) ppm. – ^{13}C NMR: $\delta = 34.8$ (CH_2), 36.7 (CH), 56.1 (OCH_3), 105.9 (C), 114.1 (C), 116.6 (CH), 117.5 (2CH), 125.8 (CH), 127.8 (CH), 130.7 (2CH), 132.8 (CH), 152.3 (C), 156.2 (C), 158.5 (C), 159.1 (C), 160.9 (C=O), 164.1 (C=O) ppm. – Analysis for $C_{19}H_{14}O_5$ (322.32): calcd. C 70.80, H 4.38; found C 70.65, H 4.16%.

4.4.9 4-(3-Nitrophenyl)-3,4-dihydro-2*H*,5*H*-pyrano[3,2-*c*] chromene-2,5-dione (6g)

White powder, yield 0.327 g (97%), m. p. 181–183°C. – IR (KBr, cm^{-1}): $\nu_{max} = 1780$, 1713, 1654, 1567, 1382, 1110. – ^1H NMR: $\delta = 3.12$ (d, 1H, H-3, $^2J_{HH} = 16.0$ Hz), 3.20 (d, 1H, H-3, $^2J_{HH} = 16.0$ Hz), 4.47 (s, 1H, H-4), 7.33 (m, 2H, H-Ar), 7.54 (t, 1H, H-Ar, $^3J_{HH} = 7.4$ Hz), 7.62 (m, 2H, H-Ar), 7.75 (m, 2H, H-Ar), 7.87 (d, 1H, H-Ar, $^3J_{HH} = 7.6$ Hz) ppm. – ^{13}C NMR: $\delta = 36.5$ (CH2), 37.7 (CH), 104.8 (C), 113.7 (C), 116.2 (CH), 120.3 (CH), 121.5 (CH), 123.8 (CH), 125.6 (CH), 130.5 (CH), 131.6 (CH), 132.4 (CH), 142.3 (C), 145.4 (C), 154.1 (C), 158.7 (C), 160.8 (C=O), 163.9 (C=O) ppm. – Analysis for $C_{18}H_{11}NO_6$ (337.29): calcd. C 64.01, H 3.29, N 4.15; found C 63.89, H 3.14, N 4.23%.

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