



A green approach to electrosynthesis of chromeno[3',4':5,6]pyrano [2,3-d] pyrimidines by electrochemistry

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ABSTRACT

Electrochemistry is a broad, useful, and selective technique method in many research fields. Among them, the investigation of performance of electrochemical methods in determination, synthesis and selective reduction/oxidation of different elements and molecules have attracted growing attention due their intrinsic advantages such as selectivity, low cost, and high yield of synthesis. Moreover, electrocatalytic synthesis of organic molecules is known as a green and environmentally benign method. In the present form, electrocatalytic multicomponent transformation of barbituric acid, aromatic aldehydes, and 4-hydroxycumarin was carried out. The electrocatalytic transformation was done in alcohols in the presence of tetrabutylammounium flouride as an electrolyte in an undivided cell containing an iron electrode as the cathode and a Pt electrode as the anode at a constant current leads to substituted chromeno[3',4':5,6]pyrano[2,3-d] pyrimidines in good to high yields (54-92%) at room temperature. The yield of reaction was obtained by gravimetric analysis and calculated upon theoretical conversion. The application of the effective electrocatalytic cascade method to the formation of chromeno-pyrano-pyrimidines is also beneficial from the viewpoint of diversity-oriented large-scale processes and represents an example of facile environmentally benign synthetic concept for electrocatalytic multicomponent reactions. The products were characterized with proper analytical methods such as elemental analysis (CHN), FT-IR, ¹H-NMR, and ¹³C-NMR spectrometry. Finally, the obtained results showed that the desired products were synthesized.

1. Introduction

Chromene derivatives have attracted great interest due to biological and pharmacological activities such as anti-coagulant, anti-cancer, spasmolytic, diuretic, anti-anaphylatic, etc. Furthermore, the chromene derivatives are widely found in natural alkaloids, flavonoids, tocopherols, and anthocyanins [1-6].

Pyrano[2,3-d]pyrimidines have also received considerable attention over the past years due to their

wide range of the diverse pharmacological action such as antitumor, cardiotoxic, hepatoprotective, antihypertensive, anticoagulant and antibronchitic activity [7-12]. Moreover, pyranopyrimidine derivatives occur widely in the structures of various natural products [13].

Thus, chromeno pyrano[2,3-d]pyrimidine system appears to be of the interest because it incorporates a chromene and a pyrano[2,3-d]pyrimidine heterocyclic ring, which are both promising with respect to biological responses.

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To the best of our knowledge, there are only a few reports on the three-component coupling of 4-hydroxycoumarin, aldehydes, and cyclic 1,3-dicarbonyl compounds [14,15].

Due to the extensive research on the electrochemistry of organic compounds, electrosynthesis has become a useful method in modern organic chemistry [16,19]. Electrochemical organosynthetic methods have received significant attention because of their benefit to the environment. In these procedures, electricity acts as a 'green' oxidative and reductive agent.

2. Experimental Procedure

2.1. Material and Methods

All reagents were purchased from Merck and Fluka and used without further purification. The melting points were obtained in open capillary tubes and were measured on an Electrothermal IA 9100 apparatus. IR spectra were recorded on KBr pellets with a Shimadzu FT-IR 8600 spectrophotometer. ^1H and ^{13}C NMR spectra were determined with a Bruker DRX-400 Avance instrument at 400 and 100 MHz. Elemental analysis were carried out on a Thermo Finnigan Flash EA 1112 series instrument.

2.2. General procedure for the synthesis of 4a-g

A mixture of barbituric acid **1** (2 mmol), aromatic aldehyde **2a-g** (2 mmol), 4-hydroxycoumarin **3** (2 mmol), and TBAF (0.04 g, 0.2 mmol) in n-PrOH/ H_2O (15/5 mL) were electrolyzed in an undivided cell equipped with a magnetic stirrer, a platinum anode and an iron cathode at room temperature under a constant current density of 4 mA/cm² ($I = 20$ mA, electrodes square 5 cm²). Progress of the reaction was monitored by thin layer chromatography. After electrolysis was finished, the precipitation of the products were obtained at pH = 7. In addition, filter cakes were washed twice with hot ethanol to give pure target products **4a-g**.

2.3. Experimental characterisation data for compounds 4a-g

7-phenyl-chromeno[3',4':5,6]pyrano[2,3-d]pyrimidine-6,8,10(7H,9H,11H)-trione (4a):

Yellow solid; mp (dec.) > 300°C. IR (KBr): $\nu_{\text{max}} = 3400, 3209, 3029, 2943, 1684, 1612, 1568, 1390, 1190$ cm⁻¹; $^1\text{H-NMR}$ (400 MHz, DMSO- d_6): $\delta = 6.11$ (s, 1H), 7.04-7.08 (m, 2H), 7.13-7.16 (m, 3H), 7.25-7.29 (m, 2H), 7.51-7.55 (m, 1H), 7.78-7.81 (m, 1H), 9.93 (s, 1H, NH), 10.12 (s, 1H, NH); $^{13}\text{C-NMR}$ (100 MHz, DMSO- d_6): $\delta = 166.2, 165.9, 165.4, 165.0, 152.6, 151.5, 143.8, 131.6, 128.0, 127.1, 125.1, 124.3, 123.6, 119.4, 116.0, 105.9, 89.8, 34.0$; Anal. Calcd for $\text{C}_{20}\text{H}_{12}\text{N}_2\text{O}_5$: C, 66.67; H, 3.36; N, 7.77. Found: C, 66.54; H, 3.24; N, 7.98.

7-(3-nitrophenyl)-chromeno[3',4':5,6]pyrano[2,3-d]pyrimidine-6,8,10(7H,9H,11H)-trione (4b):

Yellow solid; mp (dec.) > 300°C. IR (KBr): $\nu_{\text{max}} = 3400, 3211, 3072, 2980, 1684, 1612, 1570, 1525, 1351, 1282, 1183$ cm⁻¹; $^1\text{H-NMR}$ (400 MHz, DMSO- d_6): $\delta = 6.22$ (s, 1H), 7.25 (t, $J = 7.6$ Hz, 1H), 7.31 (d, $J = 8.4$ Hz, 1H), 7.46-7.58 (m, 3H), 7.80 (d, $J = 7.6$ Hz, 1H), 7.86 (s, 1H), 7.97 (d, $J = 7.6$ Hz, 1H), 10.03 (s, 1H, NH), 10.17 (s, 1H, NH); $^{13}\text{C-NMR}$ (100 MHz, DMSO- d_6): $\delta = 166.2, 164.9, 164.7, 152.7, 151.4, 148.1, 146.9, 134.2, 131.8, 129.7, 124.6, 124.4, 123.7, 123.6, 121.5, 120.6, 116.1, 104.9, 89.3, 34.2$; Anal. Calcd for $\text{C}_{20}\text{H}_{11}\text{N}_3\text{O}_7$: C, 59.27; H, 2.74; N, 10.37. Found: C, 59.22; H, 2.74; N, 10.39.

7-(4-nitrophenyl)-chromeno[3',4':5,6]pyrano[2,3-d]pyrimidine-6,8,10(7H,9H,11H)-trione (4c):

Orange solid; mp (dec.) > 300°C. IR (KBr): $\nu_{\text{max}} = 3415, 3213, 3055, 2962, 1686, 1610, 1569, 1512, 1379, 1346, 1183$ cm⁻¹; $^1\text{H-NMR}$ (400 MHz, DMSO- d_6): $\delta = 6.21$ (s, 1H), 7.25-7.32 (m, 4H), 7.54 (t, $J = 8.4$ Hz, 1H), 7.79-7.80 (m, 1H), 8.06

(d, $J = 8.8$ Hz, 2H), 9.97 (s, 1H, NH), 10.09 (s, 1H, NH); $^{13}\text{C-NMR}$ (100 MHz, DMSO- d_6): $\delta = 168.9, 166.0, 153.4, 152.8, 151.4, 145.5, 143.6, 131.8, 128.3, 124.6, 123.7, 123.5, 123.4, 119.3, 116.0, 105.0, 89.8, 34.7$; Anal. Calcd for $\text{C}_{20}\text{H}_{11}\text{N}_3\text{O}_7$: C, 59.27; H, 2.74; N, 10.37. Found: C, 59.12; H, 2.78; N, 10.50.

7-(2-chlorophenyl)-chromeno[3',4':5,6]pyrano[2,3-*d*]pyrimidine-6,8,10(7*H*,9*H*,11*H*)-trione (4d):

White solid; mp (dec.) > 300°C. IR (KBr): $\nu_{\text{max}} = 3433, 3197, 3057, 2964, 1686, 1612, 1568, 1402, 1188$ cm^{-1} ; $^1\text{H-NMR}$ (400 MHz, DMSO- d_6): $\delta = 5.99$ (s, 1H), 7.08-7.17 (m, 2H), 7.21-7.27 (m, 3H), 7.33 (d, $J = 7.2$ Hz, 1H), 7.50 (t, $J = 7.2$ Hz, 1H), 7.76 (d, $J = 7.2$ Hz, 1H), 9.86 (s, 1H, NH), 10.00 (s, 1H, NH); $^{13}\text{C-NMR}$ (100 MHz, DMSO- d_6): $\delta = 168.4, 165.6, 165.4, 164.0, 152.5, 151.5, 141.8, 133.2, 131.4, 130.7, 129.7, 127.2, 126.2, 124.2, 123.6, 119.5, 115.9, 106.0, 88.3, 34.1$; Anal. Calcd for $\text{C}_{20}\text{H}_{11}\text{ClN}_2\text{O}_5$: C, 60.85; H, 2.81; N, 7.10. Found: C, 60.74; H, 2.93; N, 7.13.

7-(4-chlorophenyl)-chromeno[3',4':5,6]pyrano[2,3-*d*]pyrimidine-6,8,10(7*H*,9*H*,11*H*)-trione (4e):

White solid; mp (dec.) > 350°C. IR (KBr): $\nu_{\text{max}} = 3429, 3217, 3068, 2920, 1686, 1612, 1572, 1373, 1188$ cm^{-1} ; $^1\text{H-NMR}$ (400 MHz, DMSO- d_6): $\delta = 6.08$ (s, 1H), 7.05 (d, $J = 8.0$ Hz, 1H), 7.07 (t, $J = 8.0$ Hz, 1H), 7.22 (d, $J = 8.4$ Hz, 2H), 7.27 (d, $J = 8.4$ Hz, 2H), 7.51 (d, $J = 7.2$ Hz, 1H), 7.78 (t, $J = 7.2$ Hz, 1H), 9.90 (s, 1H, NH), 10.07 (s, 1H, NH); $^{13}\text{C-NMR}$ (100 MHz, DMSO- d_6): $\delta = 166.0, 164.8, 152.7, 151.4, 143.1, 131.6, 129.6, 129.0, 127.9, 124.4, 123.6, 119.4, 116.0, 105.5, 89.6, 33.7$; Anal. Calcd for $\text{C}_{20}\text{H}_{11}\text{ClN}_2\text{O}_5$: C, 60.85; H, 2.81; N, 7.10. Found: C, 60.74; H, 2.93; N, 7.13.

7-(3-bromophenyl)-chromeno[3',4':5,6]pyrano[2,3-*d*]pyrimidine-6,8,10(7*H*,9*H*,11*H*)-trione (4f):

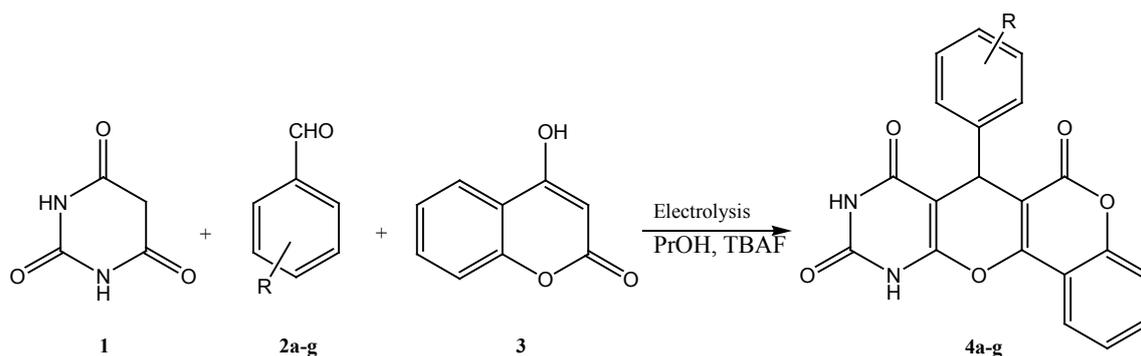
White solid; mp (dec.) > 350°C. IR (KBr): $\nu_{\text{max}} = 3333, 3167, 3060, 2933, 1684, 1612, 1576, 1468, 1398, 1188$ cm^{-1} ; $^1\text{H-NMR}$ (400 MHz, DMSO- d_6): $\delta = 6.11$ (s, 1H), 7.06-7.14 (m, 3H), 7.24-7.29 (m, 3H), 7.52-7.55 (m, 1H), 7.79 (s, 1H), 9.97-10.08 (brs, 2H, NH); $^{13}\text{C-NMR}$ (100 MHz, DMSO- d_6): $\delta = 166.1, 164.7, 152.7, 151.4, 147.4, 131.7, 130.3, 129.7, 128.0, 127.1, 126.4, 126.0, 124.4, 123.7, 121.7, 119.4, 116.0, 105.2, 89.4, 34.0$; Anal. Calcd for $\text{C}_{20}\text{H}_{11}\text{BrN}_2\text{O}_5$: C, 54.69; H, 2.52; N, 6.38. Found: C, 54.35; H, 2.64; N, 6.63.

7-(4-bromophenyl)-chromeno[3',4':5,6]pyrano[2,3-*d*]pyrimidine-6,8,10(7*H*,9*H*,11*H*)-trione (4g):

White solid; mp (dec.) > 300°C. IR (KBr): $\nu_{\text{max}} = 3400, 3210, 3084, 2930, 1684, 1612, 1568, 1377, 1184$ cm^{-1} ; $^1\text{H-NMR}$ (400 MHz, DMSO- d_6): $\delta = 6.06$ (s, 1H), 6.99 (d, $J = 8.0$ Hz, 2H), 7.23-7.29 (m, 2H), 7.32 (d, $J = 8.0$ Hz, 2H), 7.51 (d, $J = 7.6$ Hz, 1H), 7.78 (t, $J = 7.6$ Hz, 1H), 9.88 (s, 1H, NH), 10.06 (s, 1H, NH); $^{13}\text{C-NMR}$ (100 MHz, DMSO- d_6): $\delta = 168.0, 166.0, 152.5, 151.4, 148.4, 143.7, 131.6, 130.8, 129.5, 124.4, 123.6, 120.3, 119.4, 118.0, 116.0, 105.4, 89.8, 33.7$; Anal. Calcd for $\text{C}_{20}\text{H}_{11}\text{BrN}_2\text{O}_5$: C, 54.69; H, 2.52; N, 6.38. Found: C, 54.62; H, 2.45; N, 6.50.

3. Results and discussion

Simple alcohols (methanol, ethanol, propanol) or alkanes (heptane, hexane) are environmentally preferable solvents, whereas the use of dioxane, acetonitrile, acids, formaldehyde, and tetrahydrofuran is not recommendable from an environmental perspective [20, 21]. In continuation of our work [15, 22-24] on the development of efficient and convenient procedures using electrogenerated base, we were prompted to design



Scheme 1. Synthesis of chromeno pyrano[2,3-d] pyrimidine.

Table 1. Electrocatalytic transformation of barbituric acid (**1**), 3-nitrobenzaldehyde (**2b**), and 4-hydroxycoumarin (**3**) into **4b**^a.

Entry	Alcohol	I (mA)	Current density (mAcm ⁻²)	Time (min)	Electricity passed (F mol ⁻¹)	Yield (%)
1	MeOH	20	4	20	0.25	88
2	EtOH	20	4	20	0.25	90
3	PrOH	20	4	20	0.25	94
4	PrOH	10	2	40	0.25	80
5	PrOH	30	6	13.5	0.25	90
6	PrOH	50	10	8	0.25	87

^a 3-nitrobenzaldehyde (2 mmol), barbituric acid (2 mmol), 4-hydroxycoumarin (2 mmol), TBAF (0.2 mmol), alcohol/water (15/5 ml), iron cathode (5 cm²), platinum anode (5 cm²), r.t.

a green and environmentally benign methodology for the synthesis of chromeno pyrano[2,3-d] pyrimidine compounds based on electrochemically induced multicomponent reaction of barbituric acid, aromatic aldehydes and 4-hydroxycoumarin in an undivided cell containing an iron electrode as cathode and a Pt electrode as anode in the presence of tetrabutylammonium fluoride as electrolyte in alcohols at room temperature.

First, to evaluate the synthetic potential of the procedure proposed and to optimize the electrolysis conditions, the electrocatalytic multicomponent transformation of barbituric acid **1**, 3-nitrobenzaldehyde **2b**, and 4-hydroxycoumarin **3**

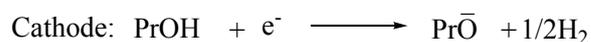
into corresponding chromeno-pyrano-pyrimidines **4b** in alcohol in an undivided cell containing an iron electrode as cathode and a Pt electrode as anode at constant current in the presence of tetrabutylammonium fluoride as an electrolyte was studied. Also, the effect of current and solvent was also examined (Table 1). As for alcohol used as solvent, PrOH is preferable to MeOH and EtOH for this electrocatalytic transformation at room temperature.

As indicated in Table 1, excellent conversions of starting compounds were obtained after 0.25 F/mol of electricity. The current density 4 mA/cm² ($I = 20$ mA, electrodes surface 5 cm²) in n-PrOH at r.t.

Table 2. Electrocatalytic transformation of barbituric acid(**1**), aromatic aldehydes (**2a-g**) and 4-hydroxycoumarin (**3**) into chromeno-pyrano-pyrimidines **4a-g**.

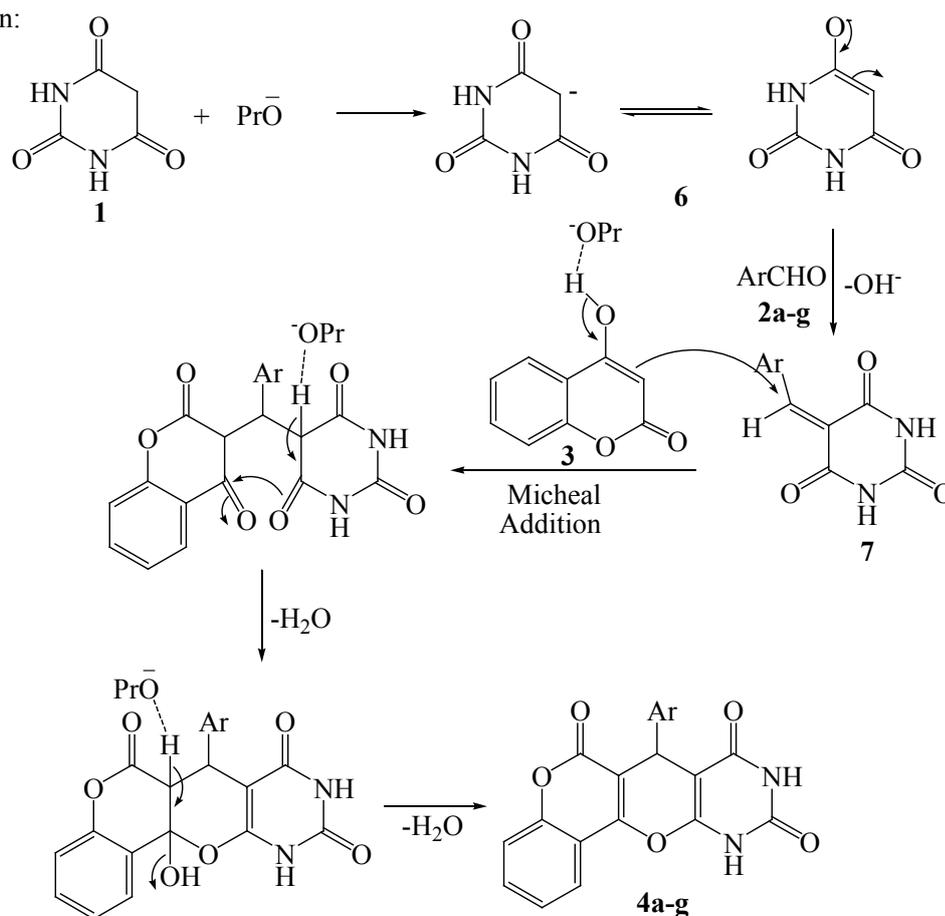
Product	Ar	Current density (mAcm ⁻²)	Time (min)	Electricity passed (F mol ⁻¹)	Yield (%)	Mp (°C)	Mp (°C) (lit. [14])
4a	C ₆ H ₅	4	45	0.56	54	> 300 dec.	> 300 dec.
4b	3-NO ₂ C ₆ H ₄	4	20	0.25	92	> 300 dec.	> 300 dec.
4c	4-NO ₂ C ₆ H ₄	4	10	0.12	82	> 300 dec.	> 300 dec.
4d	2-Cl C ₆ H ₄	4	25	0.16	88	> 300 dec.	> 300 dec.
4e	4-Cl C ₆ H ₄	4	30	0.37	90	> 350 dec.	> 350 dec.
4f	3-BrC ₆ H ₄	4	30	0.37	62	> 350 dec.	> 350 dec.
4g	4-BrC ₆ H ₄	4	30	0.37	72	> 300 dec.	> 300 dec.

^aReaction conditions: barbituric acid (2 mmol), aromatic aldehyde (2 mmol), 4-hydroxycoumarin (2 mmol), TBAF(0.2 mmol), n-PrOH/H₂O (15/5 mL), iron cathode (5 cm²), platinum anode (5 cm²), current density 4 mA/cm², room temperature.



5

in solution:

**Scheme 2.** A proposed mechanism for the electrocatalytic transformation of barbituric acid, aromatic aldehydes and 4-hydroxycoumarin into chromeno-pyrano-pyrimidines **4a-g**

was found to be optimum for the electrochemically induced chain process and allowed for the highest yield (92%) of chromeno-pyrano-pyrimidines **4b**. The increase in the current density up to 10 mA/cm² ($I = 50$ mA) results in a slight decrease of the reaction yield, which may be connected with the activation of undesired direct electrochemical processes possible under these conditions and leading to oligomerization of starting material.

Under the optimal conditions (current density 4 mA/cm², 0.25 F/mol passed, n-PrOH as a solvent), the electrolysis of barbituric acid **1**, aromatic aldehydes **2a-g**, and 4-hydroxycumarin **3** in an undivided cell gives rise to the corresponding chromeno-pyrano-pyrimidines **4a-g** with 54-92 % yield at r.t. (Scheme 1, Table 2).

Taking into consideration the above results, the following mechanism for the electrocatalytic chain transformation of barbituric acid **1**, aromatic aldehydes **2a-g**, and 4-hydroxycumarin **3** into corresponding chromeno-pyrano-pyrimidines **4a-g** is proposed.

As the initiation step of the catalytic cycle, the deprotonation of an alcohol at the cathode leads to the formation of alkoxide anion. The subsequent reaction in solution between alkoxide anion **5** and barbituric acid **1** gives rise to enolate anion **6**. Then Knoevenagel condensation of enolate anion with aromatic aldehyde takes place in the solution with elimination of hydroxide anion and formation of arylidenebarbiturate **7**. Finally, the subsequent hydroxide-promoted Michael addition of 4-hydroxycumarin **3** to electron deficient Knoevenagel adduct **7** followed by intramolecular cyclization leads to corresponding chromeno[3',4':5,6]pyrano[2,3-*d*]pyrimidines **4a-g** (Scheme 2).

4. Conclusion

The electrocatalytic transformation of barbituric acid, aromatic aldehydes, and 4-hydroxycumarin

in an undivided cell gives rise to the corresponding chromeno-pyrano-pyrimidines in comparison with conventional methods has advantages such as (i) in situ generation of base and avoidance of polluting or hazardous chemicals or the addition of base or probase, (ii) a fast one pot reaction in good to excellent yields at room temperature (iii). The procedure utilizes inexpensive reagents, green solvents, simple equipment, and an undivided cell (v). Moreover, it is easily carried out and is fully beneficial from the viewpoint of ecological organic synthesis and large-scale processes. Furthermore, this method can potentially be used for determination of such molecules.

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6. References

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