Sensors and Actuators B 193 (2014) 623–629

Contents lists available at ScienceDirect

Sensors and Actuators B: Chemical

journal homepage: www.elsevier.com/locate/snb

Highly stable pyridinic nitrogen doped graphene modified electrode in simultaneous determination of hydroquinone and catechol

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ARTICLE INFO

Article history:
Received 6 August 2013
Received in revised form
20 November 2013
Accepted 5 December 2013
Available online 17 December 2013

Keywords:
Graphene
Pyridinic nitrogen
Hydroquinone
Catechol
Simultaneous determination
Differential pulse voltammetry

ABSTRACT

A highly stable pyridinic nitrogen doped graphene (pyridine-NG) was used as an excellent electrocatalyst for the construction of electrochemical sensor for simultaneous determination of hydroquinone (HQ) and catechol (CC) in 0.20 M pH 5.5 acetate buffer solution. At the pyridine-NG modified electrode, both HQ and CC can cause a pair of quasi-reversible redox peaks and the potential difference of oxidation peaks between HQ and CC was 103 mV. Under the optimized condition, the oxidation peak current of HQ was linear over the range from 5 to 200 μM in the presence of 100 μM CC, and the oxidation peak current of CC was linear over the range from 5 to 200 μM in the presence of 100 μM HQ. The detection limit is 0.38 μM for HQ and 1 μM for CC (S/N = 3). This proposed sensor was successfully applied to the simultaneous determination of HQ and CC in artificial sample, and the results were good stability and high reproducibility. The excellent electrocatalysis of pyridine-NG can be due to the π–π interactions between the benzene ring of CC and graphene layer, the hydrogen bonds formed between hydroxyl in HQ molecule and pyridinic nitrogen atoms within graphene layers, especially the less density distribution of π electron cloud in pyridinic-NG in acidic condition.

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1. Introduction

Graphene, a two-dimensional system of carbon chicken-wire, rises as a "superstar" material due to its unique structure and electronic properties since its discovery in 2004 [1,2]. This unique property holds great promise for potential applications in various fields, such as physics, chemistry, material science, biology [3–5]. However, graphene is hydrophobic and tends to form irreversible agglomerates due to van der Waals interactions and strong π–π stacking [6]. The prevention of aggregation is of particular importance for graphene sheets because most of their unique properties are only associated with individual sheets. Although aggregation can be reduced by the attachment of surfactants [7], polymers [8] or metal nanoparticles [9–11] onto the sheets, the presence of foreign stabilizers is undesirable for most applications. Such, the direct dispersion of hydrophobic graphene sheets in water without the assistance of dispersing agents has generally been considered to be an insurmountable challenge.

Up to now, chemical reduction of graphene oxide (GO), a soft chemical synthesis route, is regarded as the efficient approach to producing relatively clean graphene sheets in bulk quantity while keeping them individually separated [6]. In this method, reducing agents commonly used, such as hydrazine hydrate [6,12] and sodium borohydride [13], are highly toxic chemicals. Other substitutes for hydrazine, such as hexamethylenetetramine [14], ethylenediamine [15] and vitamin C [16,17], are also exploited. However, stable graphene suspension in mono-layered or few-layered sheets is still a challenge. In our previous work, the GO sheets can be reduced by dimethyl ketoxime at the same time as nitrogen was doped into the graphene plane [18]. More importantly, the suspension can readily form stable aqueous colloids and display long-term stability due to electrostatic repulsion between the terminal carboxylic acid remained in the graphene sheets. It can be placed for at least six months without obviously aggregation [18]. This makes it suitable for exploiting most proposed applications, especially for modified electrode in sensors.

Dihydroxybenzene, as a fundamental chemical compound and synthetic intermediate, has been widely used to produce dyes, photostabilizer, developer, cosmetic, pesticides and some pharmaceuticals. However, this compound is toxic to many biological species and difficult to be degraded, thus threatening the environments and endangering the health of both humans and animals. Simultaneous determination of hydroquinone (1,4-dihydroxy-benzene, HQ) and catechol (1,2-dihydroxybenzene, CC) has become one of the important studies for environmental analysis because they have similar structures and properties. Electrochemical method is

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http://dx.doi.org/10.1016/j.snb.2013.12.018

Table 1
Comparison of major characteristics of different materials electrochemical sensors for simultaneous determination dihydroxybenzene isomers.

Electrode materials	Supporting electrolyte	Method	Isomer	Linear range (μM)	Detection limit (nM)	References
Single-wall carbon nanotube	pH 7.4 PBS	2.5th-order differential voltammetry	HQ	0.4–10; 40–100	120	[19]
			CC	0.4–10; 40–100	260	
			RC	0.4–10; 40–100	300	
Graphene/BMIMPF$_6$ nanocomposite	0.1 M pH 5.0 ABS	DPV	HQ	0.5–50	10	[20]
		DPV	CC	0.5–50	20	
Thermally reduced graphene oxide	0.02 M pH 6.0 PBS^4	DPV	HQ	1–500	750	[24]
		DPV	CC	1–500	800	
Polydopamine-reduced graphene oxide nanocomposite	0.1 M pH 4.5 PBS	DPV	HQ	1–230	720	[25]
		DPV	CC	1–250	820	
Gold nanoparticle and graphene composite film co-electrodeposited with carbon ionic liquid	0.1 M pH 2.5 PBS	DPV	HQ	0.06–800	18	[26]
Graphene oxide–mesoporous MnO$_2$ nanocomposite	0.1 M pH 7.0 PBS	DPV	HQ	0.01–0.7	7	[27]
		DPV	CC	0.03–1	10	
Mixture of graphene nanosheets and carbon nanospheres	0.1 M pH 8.0 PBS^3	DPV	HQ	0.1–100	10	[28]
				100–600		
Electrodeposition of reduced graphene oxide	0.2 M pH 5.8 ABS	DPV	HQ	6–200	200	[29]
		DPV	CC	1–200	100	
Nafion-(4-ferrocenylethyne) phenylamine-carbon nanoparticles-graphene	0.2 M pH 5.4 ABS	DPV	HQ	0.3–90	100	[30]
		DPV	CC	0.6–100	700	
		DPV	RC	4–300	200	
Gold nanoparticle/sulfonated graphene composites	0.1 M pH 5.6 ABS	DPV	HQ	5–100	1000	[31]
		DPV	CC	5–100	2000	
Poly(brilliant cresyl blue)	0.1 M pH 7.0 PBS^2	DPV	HQ	1–250	60	[32]
				1–250	50	
Graphene-gold nanocomposite film	0.1 M pH 7.0 PBS^1	DPV	HQ	0.016–120	5.2	[33]
			RC	0.01–2	2.2	
Poly(3,4-ethylenedioxy-thiophene)/graphene oxide hybrid film	0.1 M pH 6.0 PBS^2	DPV	HQ	2.5–200	1600	[34]
			CC	2–400	1600	
Carbon nanoparticle – chitosan composite	0.1 M pH 7.0 PBS^2	DPV	HQ	0.8–100	200	[35]
		DPV	CC	0.8–100	200	
		DPV	RC	8–1000	3000	
Poly(thionine)	0.1 M pH 7.0 PBS^2	DPV	HQ	1–120	30	[36]
			CC	1–120	25	
Pyridinic nitrogen doped graphene	0.1 M pH 5.5 ABS	DPV	HQ	5–30; 30–200	380	This work
		DPV	CC	5–200	1000	

ABS, HAc–NaAc; PBS^1, Na_2HPO_4–KH_2PO_4–NaCl–KCl; PBS^2, Na_2HPO_4–KH_2PO_4; PBS^3, Na_2HPO_4–NaH_2PO_4; PBS^4, Na_2HPO_4–KH_2PO_4–NaCl.

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