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Ex-situ decoration of graphene oxide with palladium nanoparticles for the highly sensitive and selective electrochemical determination of chloramphenicol in food and biological samples

Thangavelu Kokulnathan[a], Tata Sanjay Kanna Sharma[a], Shen-Ming Chen[a,*], Tse-Wei Chen[a], Bose Dinesh[b]

[a] Electroanalysis and Bioelectrochemistry Lab, Department of Chemical Engineering and Biotechnology, National Taipei University of Technology, No.1, Section 3, Chung-Hsiao East Road, Taipei 106, Taiwan, ROC

[b] Nano and Bioelectrochemistry Research Laboratory, Department of Chemistry, School of Advanced Sciences, Vellore Institute of Technology, Vellore 632014, Tamil Nadu, India

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ABSTRACT

The global usage of antibiotics drug chloramphenicol (CPL) in pharmaceuticals and food-producing animals may cause severe threats to both human health and animals. Therefore, the development of highly selective detection of chloramphenicol has turned the hottest issue to protect human health and another living organism. In this work, we have developed well dispersive palladium nanoparticles decorated with graphene oxide (Pd NPs/GO) nanocomposite, by facile ice bath method. Moreover, the successful formation of Pd NPs/GO nanocomposite was deep-rooted by various analytical and spectroscopic methods. The electrochemical comportment of the electrochemical sensor was investigated by cyclic voltammetry and amperometric method. The Pd NPs/GO nanocomposite shows an excellent electrocatalytic behavior in terms of higher cathodic peak current and lower peak potential than that of other modified and unmodified glassy carbon electrode (GCE). Furthermore, the Pd NPs/GO modified GCE displayed an excellent linear response range (0.007 to 102.68 µM), good sensitivity (3.0479 µA µM^{-1} cm^{2}) and lower detection limit (0.001 µM), correspondingly. The Pd NPs/GO/GCE spectacles its excellent selectivity for the CPL sensing in the presence of other potentially interfering compounds. Additionally, the nanocomposite is efficaciously applied to the biological and food samples for the determination of CPL and obtained good recoveries values.

1. Introduction

The universal consumption of antibiotics from last decades to continue contributing to the treatment of bacterial diseases to the human and animals. All the pharmaceutical antibiotics are not safer for treating bacterial infection and it also has some pose a threat to health [1]. Especially, chloramphenicol (CPL) is well-known antibiotics drug commonly used in human and veterinary medicine since the 1949s [2]. The CPL is naturally isolated by David Gotlieb in 1947 from the micro-organism *Streptomyces Venezuela* and it belongs to the *Actinomycetes*, which are abundantly found soil-bacteria in worldwide [3]. Due to cheapness and high efficiency, it is widely applied to the animal food production, domestic poultry, therapeutic & prophylactic agent of bacteria, superficial eye infection, aqua agriculture farming [4,5]. However, the accumulation of CPL in food-producing animals has lethal threat the health of human beings. Besides, CPL can cause human health hazards such as blood dyscrasias (myelosuppression, thrombocytopenia, granulocytopenia, hypoplastic anemia, aplastic anemia), neurotoxic reactions (mental confusion, delirium, headache, depression), gastrointestinal disorders (diarrhea, enterocolitis, nausea, vomiting) and hypersensitivity (anaphylaxis, fever, vesicular rashes, urticaria, angioedema), respectively [6,7]. Additionally, CPL use is strictly prohibited to women pregnant and also breastfeeding. Because, CPL is potentially harmful to fetal with adverse effects such as gray syndrome, irregular respiration, pallid cyanosis, vasomotor collapse and abdominal distension with or without emesis. It also easily passes through breast milk to the infant [4,8].

Remarkably, CPL is highly risk consumption with other drugs, which can affect or reduce blood cell counts (clozapine). For above reasons, many countries including United States, China, Australia, Japan and the European Union (EU) are banned this CPL drug [9]. The European Union (1994) has also established a minimum re-

* Corresponding author.
E-mail addresses: smchen78@ms15.hinet.net, smchen@ntut.edu.tw (S.-M. Chen).

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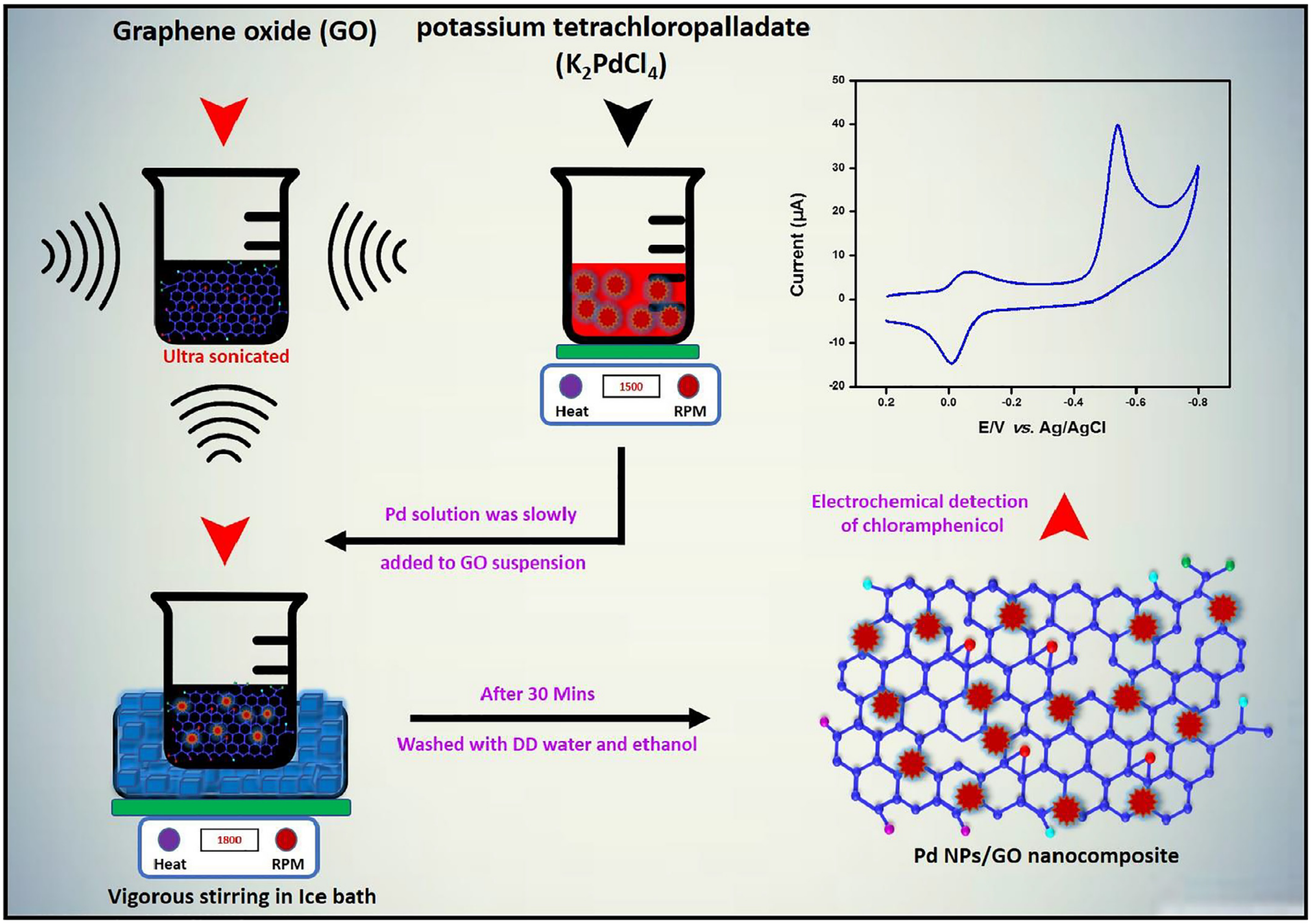

Scheme 1. The overall synthesis of Pd NPs/GO nanocomposite and its electrochemical application towards detection of CPL.

quired performance limits (MRPL) for all matrices for CPL at a level of 3 μg/kg [10]. Still, some emerging countries are illegitimately rummage-sale CPL for animal food production and treatment for infectious diseases. Bestowing to EU decisions, aquaculture and fishery food products are imported from China, Indonesia, Vietnam, Myanmar, and Thailand are regularly examined to ensure the absence of CPL filtrates [11]. Due to the importance of the CPL drug and their extensive global dangerous, the detection and measurement of the food merchandises in a human body is an important analytical challenge. Henceforward, analytical chemists have taken this as a challenge to develop a rapid methodology for the selective and sensitive determination of CPL. Consequently, many approaches have been proposed for CPL detection, such as colorimetric, immune sensors, electroanalysis method, high-pressure liquid chromatography, and fluorescence detection and microbiological [4–8]. These techniques possess some limitations of expertise monitoring, high cost, cumbersome, time-consuming, tedious extraction process, complicated routine analysis, low sensitivity, and selectivity are affecting the response. The electrochemical sensor as an appropriate and efficient technique has been a reliable analytical method and attracted due to its simplicity, low cost, rapid response, high selectivity, excellent sensitivity and spatial controlment [12,13]. Howbeit, the direct electrochemical reduction of CPL on the conventional electrode suffers with the poor electron transfer and sluggish response as well as over reduction potential. Therefore, the nanostructured and carbonaceous material is the most fruitful nominees for enhanced electrochemical performance.

GO is made up of a single layer of graphite and directly generated via chemical treatment graphite through oxidation. It can effortlessly disperse in water and other appropriate organic solvents. In the intervening time, the abundant oxygen functional groups located on GO's basal plane and edges (epoxide, hydroxyl, phenol, lactone, quinone, carbonyl/carboxyl groups), individually [14,15]. Many electrochemists pay more consideration to it because of its admirable conductivity, less weight, strong mechanical properties, high surface area and superior chemical stability [16]. However, the electrochemical application of GO has some hitches due to its insulation properties which are interrelated to the manifestation of oxygen functionalities. To overcome this issue, GO was composite with metal particles, metal oxide, conductive polymers, and biopolymers [14,17]. As mentioned above, small sized metal particles have been acknowledged as a good mediator on the fabrication of electrochemical sensors due to their biocompatibility, fast electron transfer rate, and excellent conductivity. Therefore, decoration of small-sized metal nanoparticles on the surface of GO sheets is proposed to not only further resolve the insulation properties problem of GO but also significantly increase the electrochemical activity towards CPL detection.

Incontrovertibly, noble metals (Ag, Au, Ru, Pd, Pt) is a vital electrocatalysis with nanosize have momentous interest because of their sophisticated properties (large surface area, high activity, reactivity) and the numeral of edges and corner atoms [18–21]. Among the innumerable noble metals, Pd NPs have received particular attention owing to their excellent physio-chemical properties. The Pd NPs have some compensations such as fast electron