

Regular Article

Ex-situ decoration of ordered mesoporous carbon with palladium nanoparticles via polyoxometalates and for sensitive detection of acetaminophen in pharmaceutical products

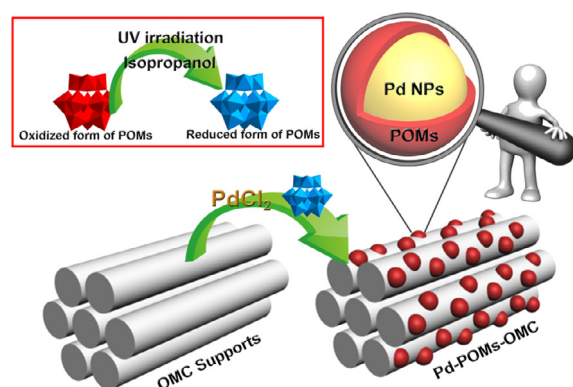


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GRAPHICAL ABSTRACT

A facile and green method is described for decoration of ordered mesoporous carbon (OMC) with palladium nanoparticles (Pd NPs) through a polyoxometalates (POMs).



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ABSTRACT

In this study, a facile and green method is described for decoration of ordered mesoporous carbon (OMC) with palladium nanoparticles (Pd NPs) through a polyoxometalates (POMs). Serving as both reducing and stabilizing agent, the Keggin-type POMs are used for synthesis of stable colloidal Pd NPs and then link them to the OMC at mild temperature. Current research focuses on the use of Pd-POMs-OMC as an effective sensing template for the detection of acetaminophen (AP) has been studied. Linear calibration curves in the range of 0.10–33.15 μM ($R^2 = 0.998$) was obtained for AP with a sensitivity of $985.4 \mu\text{AmM}^{-1}$; the limit of detection was calculated to be 0.069 μM . Importantly, the successful fabricated Pd-POMs-OMC device accurately measured the amount of AP in pharmaceutical samples.

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1. Introduction

Acetaminophen (AP) is a major ingredient in numerous cold and influenza medications [1–4]. Conversely, the overdoses of AP can cause hepatic toxicity and kidney damage. Hence, the determination of AP receives much more attention in biological samples and also in pharmaceutical formulations. A variety of techniques has been developed for the determination of acetaminophen, including titrimetric [5,6], chemiluminescent [7,8], high performance-liquid chromatography [9], and UV–Vis spectrometry [10]. Commonly, these techniques are costly, complicated, time consuming and operated by highly skilled technicians. However, electrochemical methods can provide a simple, rapid, and selective detection with low-cost of systems and eco-friendly [11–14].

Noble-metal based electro-catalysts (such as Pt, Ru, Ir, Au and Pd) have received considerable attention owing to their excellent optical and electrical properties. Among the noble-metal based electro-catalysts studied to date, Pd, which is considerably less expensive than other noble-metals, has been widely investigated [15–20].

Polyoxometalates (POMs), which are polyatomic anionic ion clusters composing of d-block transitional metal-oxides, as catalysts has been the focus of intensive research on catalysis, medicine and materials sciences [21–23]. This is because POMs exhibit a wide range of structural, redox, and photochemical characteristics and readily participate in reversible multielectron redox transformations [24]. In reduced forms, their electron and proton transfer and/or storage abilities may act as efficient donors or acceptors of several electrons without structural changes [25,26]. In general, POMs-passivated metal NPs have been prepared through ligand place-exchange reactions, reducing the metal precursors in the presence of POMs, or reducing metal precursors by using reduced POMs as reducing agents [27,28].

Recently, the synergetic effect on heterogeneous composite catalysts has attracted great attention because the combination of catalytic component and support materials can sometimes endows the composite catalysts with unexpectedly improved catalytic properties [29–31]. Ordered mesoporous carbon (OMC) exhibits uniform tailored and extremely well-ordered pore structure, high specific surface area, large pore volume, excellent conductivity, good thermal stability and chemical inertness, which make them suitable for applications in electrocatalysis and design of electrochemical sensors [32–38]. In particular, favourable chemical and electronic interaction between noble-metal NPs and OMC leads to a synergistic enhancement of electrocatalytic activity [39–42].

In this work, relatively uniform Pd NPs attached on OMC surface were prepared through a green and facile method using $\text{H}_3\text{PW}_{12}\text{O}_{40}$ (PW12, henceforth POMs for convenience) as both reductant and bridging molecules. The as-prepared novel tri-component nanohybrids of Pd-POMs-OMC extend the applications of support materials and provide new features of electrocatalytic activities. The new type of Pd-POMs-OMC nanocomposite modified electrode has been fabricated, which was employed for an excellent electrochemical sensor of AP.

2. Experimental

2.1. Chemical reagents

AP, PdCl_2 , POMs, isopropanol, and N, N'-dimethylformamide (DMF) (HPLC grade) were used as purchased from MACKLIN Reagent Co. Ltd. The 0.1 M phosphate buffer solution (PBS pH 7.0), which was made up from NaH_2PO_4 , Na_2HPO_4 , and H_3PO_4 , was employed as a supporting electrolyte. All other reagents were of analytical grade, and all solutions were prepared using double distilled water.

2.2. Instrumentation

All the electrochemical experiments were performed with an Autolab Electrochemistry Workstation (PGSTAT 302N, Metrohm, Switzerland). Electrochemical impedance spectroscopy (EIS) was conducted using the Autolab electrochemical analyzer in a 0.1 M KCl solution containing 5.0 mM $\text{K}_3\text{Fe}(\text{CN})_6/\text{K}_4\text{Fe}(\text{CN})_6$, from 0.1 Hz to 10.0 kHz. Scanning electron microscopy (SEM) image was determined with a Philips XL-30 ESEM, operating at 3.0 kV. Transmission electron microscopy (TEM) images and energy-dispersive X-ray (EDX) spectra were obtained using a JEM-2100F transmission electron microscope JEOL (Japan) operating at 200 kV. Nitrogen adsorption-desorption isotherms were measured on ASAP 2020 Micromeritics (USA) at 77 K. The Brunauer–Emmett–Teller (BET) method was utilized to calculate the specific surface area. X-ray diffraction (XRD) patterns were obtained on an X-ray D/max-2200vpc (Rigaku Corporation, Japan) instrument operated at 40 kV and 20 mA using Cu $\text{K}\alpha$ radiation ($\lambda = 0.15406$ nm). X-ray photoelectron spectroscopy (XPS) measurements were performed with a thermo ESCA LAB spectrometer (USA). A conventional three electrode cell was used; the working electrode was glassy carbon electrode (GCE) or the modified electrode; a platinum electrode was used as the counter electrode whereas an Ag/AgCl (in saturated KCl solution) electrode served as a reference electrode. All potentials in this paper were measured and reported versus Ag/AgCl.

2.3. Synthesis of tri-component nanohybrids of Pd-POMs-OMC

OMC was prepared according to the method reported by Ryoo et al. [43]. For Pd-POMs-OMC, firstly, 1.5 mL of POMs (10 mg/mL) was added in a quartz container and mixed with 30 μL of isopropanol; the mixed solution was irradiated under ultra-violet light (500 W) for 30 min and the color of the solution became blue. Secondly, PdCl_2 (12 μL , 100 mgmL^{-1}) and OMC suspension (0.2 mL, 5 mgmL^{-1}) were adding to the solution of reduced POMs with stirring 5 min. At last, the resulting Pd-POMs-OMC nanomaterial was isolated, thoroughly washed with doubly distilled water, and finally dried under vacuum overnight. The whole reaction was performed at room temperature and ambient pressure. Illustration of the preparation of Pd-POMs-OMC is presented in Scheme 1.

2.4. Preparation of the modified electrodes

Prior to the modification, GCE (model CHI104, 3 mm diameter) was polished before each experiment with 1, 0.3 and 0.05 μm alumina powder, respectively, rinsed thoroughly with double distilled water between each polishing step, and then sonicated successively in 1:1 nitric acid, absolute alcohol, double distilled water. The cleaned electrode was dried with nitrogen stream for the next modification. To prepare the modified electrodes, 5 mg of the electrode materials were dispersed into 1 mL DMF to give homogeneous suspension upon bath sonication. A 5 μL of the suspension was dropped onto GCE and the electrode was then dried at room temperature.

2.5. Pharmaceutical real samples solution preparation

The tablets of AP were finely powdered in a mortar with pestle. Calculated amounts of the tablets required for AP were transferred into a 25 mL volumetric flask and were dissolved in 0.1 M PBS (pH 7.0). The solution was filtered and a suitable aliquot of the clear filtrate was collected and stored in the refrigerator for further uses.

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