



A novel strategy to synthesize Au nanoplates and their application for enzymeless H₂O₂ detection

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ABSTRACT

The present communication reports a novel and simple strategy to synthesize Au nanoplates (AuPs) by heating an aqueous solution of H₂AuCl₄ in the presence of poly [(2-ethyl dimethylammonioethyl methacrylate ethyl sulfate)-co-(1-vinylpyrrolidone)] (PQ11). Direct placing of the resultant dispersion on a glassy carbon electrode (GCE) surface gives a very stable AuPs-containing film exhibiting remarkable catalytic performance toward both the oxidation and reduction of H₂O₂. This enzymeless H₂O₂ sensor shows a fast amperometric response time of less than 3 s and the corresponding linear range and detection limit are estimated to be from 0.1 mM to 50 mM ($r = 0.999$) and 4 μ M at a signal-to-noise ratio of 3, respectively.

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1. Introduction

It has been established that H₂O₂ as a metabolic intermediate is involved in many biological reactions and plays an important role in the fields of chemistry, biology, clinical control and environmental protection [1–3]. This has raised extensive demands for establishing protocols for H₂O₂ detection. Till now, various techniques including spectrometry, titrimetry, chemiluminescence, and electrochemistry have been employed for determination of H₂O₂ [4–8]. Among them, electrochemical methods have attracted considerable attention owing to their high sensitivity, good selectivity, low-cost, and ease of operation [9]. Most previous studies on this subject involved the use of enzymes which can accelerate the electron transfer between the electrodes and H₂O₂ [10,11]. Their application, however, is limited because enzymes are expensive and easily denatured [12]. With the rapid development in nanotechnology, nanomaterials such as noble metal nanoparticles, carbon nanotubes, and reduced graphene oxide were designed as catalysts to catalyze the oxidation or reduction of H₂O₂ [13–18], leading to enzymeless H₂O₂ sensors.

On the other hand, Au nanomaterials have been widely used in biomedical fields due to their excellent biocompatibility, nontoxicity, catalytic activity and offer a hospitable environment for biomolecules [19]. Hence, there is increasing research attention to biosensors based on Au nanomaterials [20,21]. Au nanoparticles provide a stable immobilization of biomolecules retaining their bioactivity and facilitating electron transfer between the target molecules and electrode substrates and hence have advantages for the construction of electrochemical biosensors with enhanced analytical performance [22,23]. Although Au nanoparticles based electrochemical biosensing has been largely studied [24–26], little attention has been paid to electrochemical sensing application of two-dimensional Au nanoplates (AuPs) [27]. More recently, we have prepared AuPs in aqueous solution at room temperature using tannic acid (TA) as a reducing agent and found that these AuPs exhibit notable catalytic performance toward the oxidation and reduction of H₂O₂ [28]. In order to obtain stable AuPs films, however, chitosan was used as an immobilization support matrix which may block electron transfer at the modified electrode [29].

In this study, we report on a novel and simple strategy to synthesize AuPs by heating an aqueous solution of H₂AuCl₄ in the presence of poly [(2-ethyl dimethylammonioethyl methacrylate ethyl sulfate)-co-(1-vinylpyrrolidone)] (PQ11). A very stable AuPs-containing film exhibiting remarkable catalytic performance

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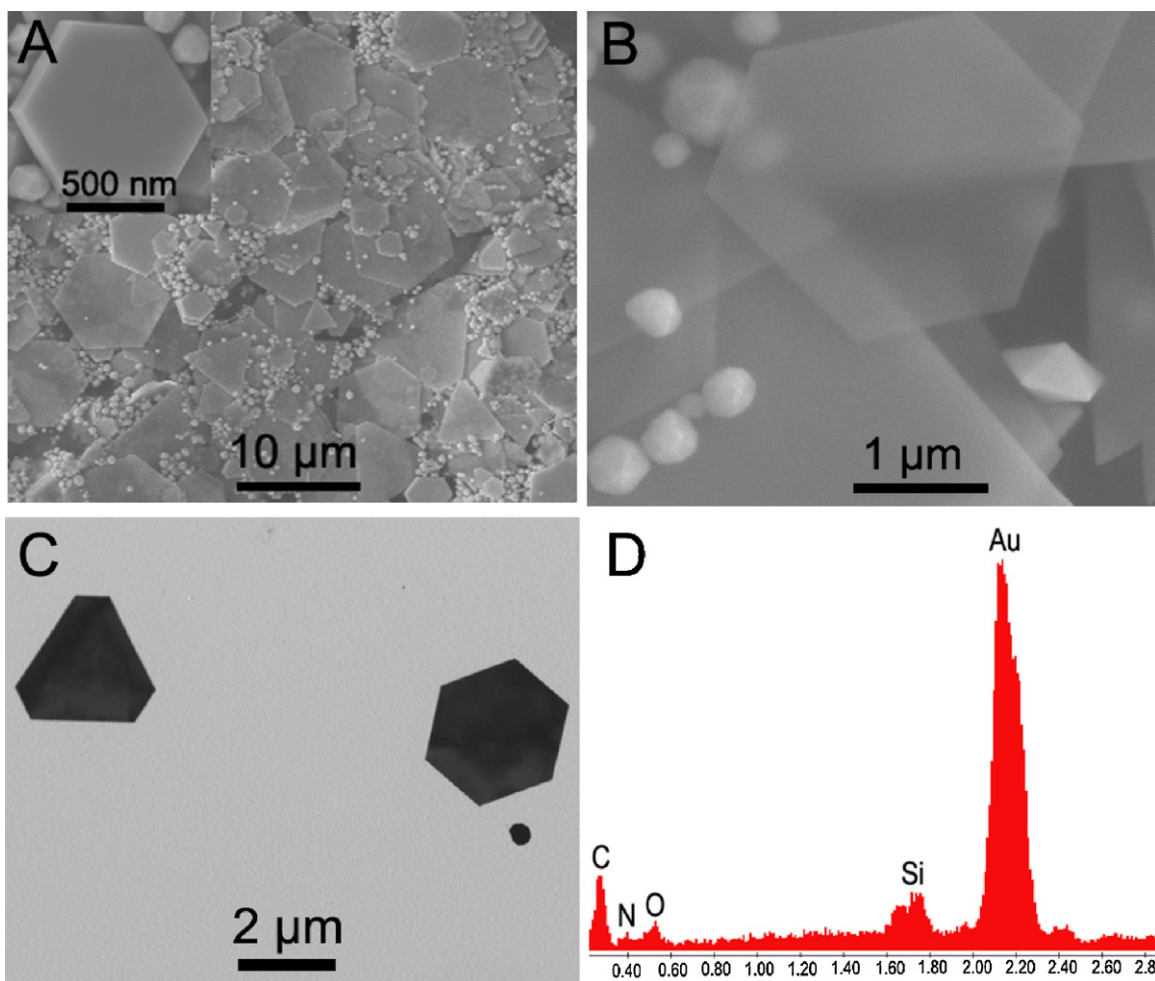


Fig. 1. (A and B) SEM images, (C) TEM image, and (D) EDS of the precipitates. The inset in A shows that the edge length of hexagonal Au nanoplate is about 500 nm.

toward both the oxidation and reduction of H_2O_2 was formed on bare substrates without the use of an immobilization support matrix. As-constructed enzymeless H_2O_2 sensor on a glassy carbon electrode (GCE) shows a fast amperometric response time of less than 3 s and the corresponding linear range and detection limit are estimated to be from 0.1 mM to 50 mM ($r=0.999$) and 4 μM at a signal-to-noise ratio of 3, respectively.

2. Experimental

2.1. Reagents and materials

PQ11 (20 wt% in H_2O), H_2O_2 (30 wt%), Na_2HPO_4 , and NaH_2PO_4 were purchased from Aladin Ltd. (Shanghai, China). HAuCl_4 were purchased from Aldrich Chemical Comp. All chemicals were used as received without further purification. The water used throughout all experiments was purified through a Millipore system. Phosphate buffer saline (PBS) was prepared by mixing stock solutions of NaH_2PO_4 and Na_2HPO_4 and a fresh solution of H_2O_2 was prepared daily.

2.2. Synthesis of AuPs

In a typical experiment, 2 mL of 0.8 M PQ11 aqueous solution was mixed with 6 mL of H_2O , and then 3 mL of 24.3 mM HAuCl_4 aqueous solution was added into the above diluted PQ11 aqueous

solution at room temperature. The resulting solution was subjected to heat treatment at 94 °C for 40 min, leading to a large amount of sand-like golden precipitates. The resulting precipitates were centrifuged and washed twice with distilled water, and then redispersed in water for characterization and further use.

2.3. Characterization

Transmission electron microscopy (TEM) measurement was made on a HITACHI H-8100 EM (Hitachi, Tokyo, Japan) with an accelerating voltage of 200 kV. Scanning electron microscopy (SEM) measurements were made on a XL30 ESEM FEG scanning electron microscope at an accelerating applied potential of 20 kV. The sample for SEM characterization was prepared by placing a drop of the dispersion on a bare indium tin oxide coated glass substrate (ITO) and air-dried at room temperature. Electrochemical measurements are performed with a CHI 660D electrochemical analyzer (CH Instruments, Inc., Shanghai). A conventional three-electrode cell was used, including a GCE (geometric area = 0.07 cm^2) as the working electrode, a Ag/AgCl (saturated KCl) electrode as the reference electrode, and platinum foil as the counter electrode. The sensor was fabricated by placing 3- μL dispersion on a bare GCE surface and air-dried for 10 min at room temperature, followed by rinsing the film formed on the electrode surface with water. All the experiments were carried out at ambient temperature.

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