



A new label-free electrochemical immunosensor based on dendritic core-shell AuPd@Au nanocrystals for highly sensitive detection of prostate specific antigen

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ABSTRACT

Herein, bimetallic dendritic core-shell AuPd@Au nanocrystals (AuPd@Au NCs) were prepared by a simple one-pot aqueous method using xanthine as a green growth-directing agent. By virtue of the enhanced peak currents in the H₂O₂ reduction catalyzed by AuPd@Au NCs, a label-free immunosensor was constructed for the detection of prostate specific antigen (PSA). The immunosensor exhibited significantly improved analytical performance for the assay of PSA with wide linear range of 0.1–50 ng mL⁻¹ and low detection limit of 0.078 ng mL⁻¹ (S/N = 3), coupled with the improved stability, reproducibility and selectivity. It provides a promising platform for clinical research and diagnosis.

1. Introduction

In the past few years, electrochemical immunosensors have attracted much interest by virtue of their advantages such as high specificity and sensitivity, as well as facile operation (Yu et al., 2013). Thus, it has great potential applications in clinic diagnostics for prediction and monitoring of diseases (Han et al., 2017). Particularly, label-free electrochemical immunosensor is promising because of their advantages including the simplified working procedure and the direct detection of antibody (Ab)-antigen binding (Li et al., 2011), accompanied by eliminating the time-consuming and complicated labeling processes compared with the sandwich-typed counterparts (Yang et al., 2014).

As it is well known, the electrode materials are essential for the design and construction of the label-free immunosensor (Zhu et al., 2015), which are closely associated with the immobilization of antibodies and the signal amplification (Moon et al., 2014). Nowadays, numerous nanomaterials were employed as platform for signal amplification such as metal nanoparticles (NPs) (Wang et al., 2014), graphene (Yan et al., 2012), conducting polymers (Sundaram et al., 2008), carbon nanotubes (Prieto-Simón et al., 2015) and their derivatives (Jang et al., 2015). Among them, metal NPs especially bimetallic NPs have attracted great interest due to their good catalytic activity, large surface area, and high electrical conductivity (Guo et al., 2010).

Early stage detection and evaluate the reappear of the disease after

treatment play the significant roles in clinic analysis of cancer biomarkers (Chen et al., 2013). Prostate cancer is the third most common disease in human (Jie et al., 2008). It is known that prostate specific antigen (PSA) is the premier biomarker in the diagnosis and the target of the prostate cancer at the early stage (Kingsmore, 2006), whose common concentration is 4.0 ng mL⁻¹, and the above of 10 ng mL⁻¹ is considered as abnormal (Jin et al., 2015). Thus, the detection of PSA plays the key role in the prediction and diagnosis of prostate cancer.

Xanthine is one of purine bases (Fig. S1, Supporting Information, SI), which is usually found in most human body tissues, fluids and other organisms (Plekan et al., 2012). In this study, a facile one-pot wet-chemical approach was developed to prepare dendritic core-shell AuPd@Au nanocrystals (NCs) with the assistance of xanthine. A new PSA electrochemical immunosensor was constructed based on the enhanced catalytic currents by reducing H₂O₂ at AuPd@Au NCs modified glassy carbon electrode (GCE).

2. Experimental

2.1. Preparation of dendritic AuPd@Au nanocrystals (NCs)

For typical construction of AuPd@Au NCs, xanthine (10 mM) was firstly dissolved into 10 mL of water containing 150 µL of NaOH (1 M). Then, 2 mM HAuCl₄ and 2 mM H₂PdCl₄ solutions were successively

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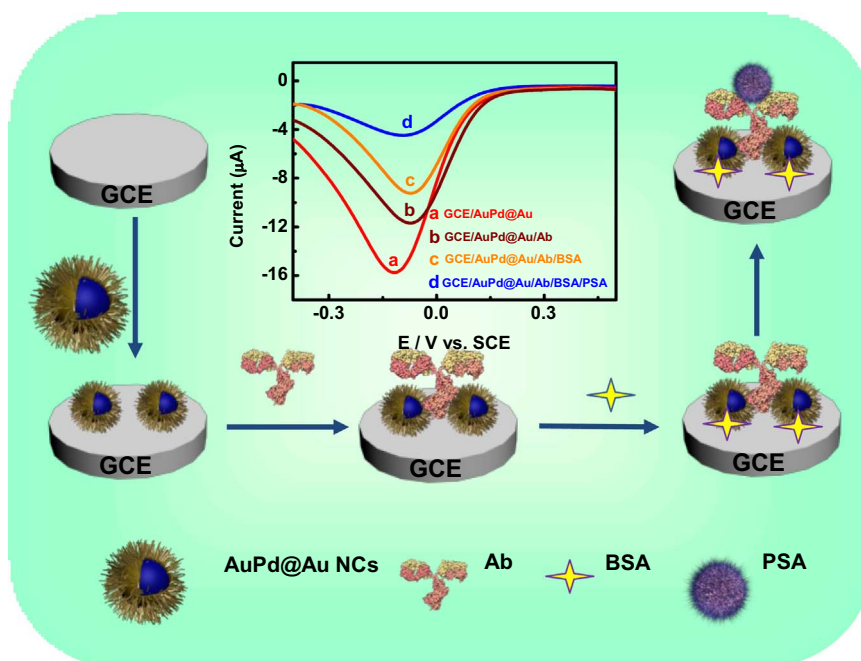


Fig. 1. Schematic illustration of the construction of an immunosensor based on AuPd@Au NCs. Inset shows the DPV curves of AuPd@Au NCs (curve a), AuPd@Au NCs/Ab (curve b), AuPd@Au NCs/Ab/BSA (curve c) and AuPd@Au NCs/Ab/BSA/PSA (curve d) modified electrodes in N_2 -saturated PBS containing 5 mM H_2O_2 .

put into the mixed solution under stirring at 60 °C, followed by the addition of 10 mM freshly-prepared ascorbic acid (AA). After reacting for 40 min, the hydrazine solution (16.7 M) was dropwise injected into the reaction system and kept reacting for 0.5 h under stirring. The prepared product was ultimately collected by centrifugation, completely washed with ethanol, and dried in a vacuum at 60 °C for further characterization.

2.2. Fabrication of the immunosensor

Fig. 1 describes the scheme for the construction of the AuPd@Au NCs based immunosensor. Firstly, 12 μL of the AuPd@Au NCs suspension (2 mg mL^{-1}) was dropped onto the clean glass carbon electrode (GCE, 3 mm in diameter) and dried in air, accompanied with the subsequent deposition of 6 μL of the Ab solution ($10 \mu\text{g mL}^{-1}$)

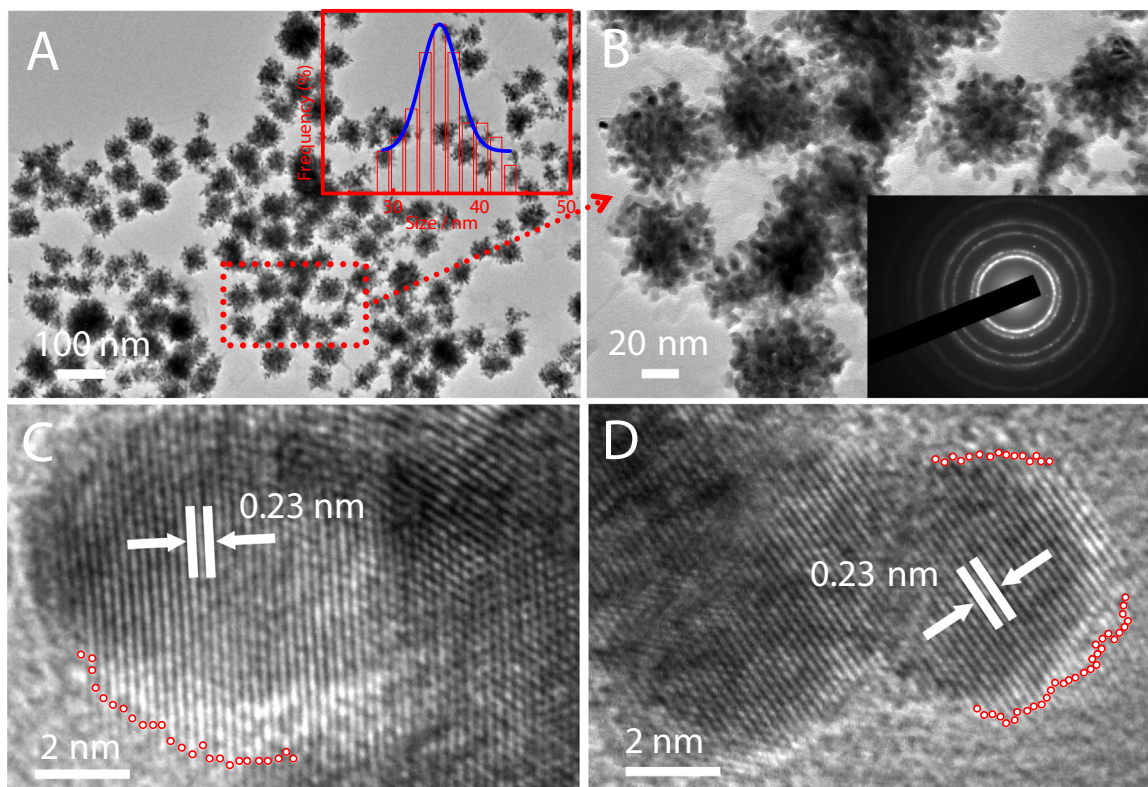


Fig. 2. (A) Low- and (B) medium-magnification TEM images of AuPd@Au NCs prepared with 10 mM xanthine. (C–D) The corresponding high-resolution TEM images. Insets in Fig. 2A and B show the associated particle-size histogram and SAED pattern, respectively.

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