



Enhanced anti-interference on electrochemical detection of arsenite with nanoporous gold in mild condition



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ABSTRACT

Determination of arsenite [As(III)] without interference in mild condition is crucial for portably assessing arsenic contamination using electrochemical method. We have developed high-surface area nanoporous gold (np-Au) with a three dimensional, interconnected ligaments and nanoporous structure for the electrochemical detection of As(III) in 0.1 M HAC-NaAc solution (pH 5.0) without using strong acidic electrolyte. Square wave anodic stripping voltammetry (SWASV) using the np-Au modified glassy carbon electrode (GCE) confirms the successful detection of As(III) with almost no interference from some commonly coexisting ions. Furthermore, the sensitivity of the np-Au modified GCE exhibited approximately 10-fold enhancement as compared to Au nanoparticles (Au NPs) modified GCE. Finally, the proposed method is successfully applicable for analysis of As(III) in real water samples with satisfactory recoveries. The np-Au modified GCE shows enhanced anti-interference and excellent sensing performance may be attribute to its special surface structure and the fast transports of analytes and electron in the interface of electrode.

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1. Introduction

The presence of arsenic (As) in food and water is a serious worldwide threat to public health, exposure to which can cause a lot of health problems, including skin lesions, keratosis (skin hardening), respiratory, mutagenic, and carcinogenic effects [1,2]. The contamination of groundwater by arsenic has been reported in 20 countries where arsenic levels in drinking water are above the World Health Organization (WHO) guideline value of 10 ppb [3–5]. In the natural environment, Arsenic is predominantly present as trivalent arsenite [As(III)] and pentavalent arsenate [As(V)], and As(III) is reported to be 40–70 fold more toxic than As(V) [6–8]. So it is of utmost urgent to develop sensitive, fast, selective, and reliable analytical methods for As(III) detection.

Up to now, a variety of methods including graphite furnace absorption spectrometry (GFAS), electro-spray MS (ES-MS) coupled to chromatographic separation (HPLC,GC), neutron-activation analysis (NAA), inductive coupled plasma mass spectrometry (ICP-MS), X-ray fluorescence (XRF), atomic emission spectrometry

(AES) generally with inductively coupled plasma (ICP-AES), and so on [2,6,9–14], have been used for the determination of As(III). Nevertheless, these techniques require expensive instruments, time-intensive, laboratory setup, and well trained technicians for their handling, which can not be routinely used for in-situ analysis. Compared to these techniques mentioned above, the low-cost electrochemical methods, particularly stripping voltammetry analysis may provide an attractive alternative for their excellent sensitivity, simple operation, and ease of portability [15,16].

Recently, many kinds of electrode materials including platinum [8,17], mercury [18,19], boron-doped diamond [20,21], modified GCE [22], and gold electrode [23–25], have been developed for determination of As(III). Because of their high hydrogen over-voltage, highly sensitive, and reversibility, the nanosized Au and Au-based materials have received more attention in the electrochemical detection of As(III). Srivastava et al. [26] determined As(III) at a graphene paste electrode modified with the thiocrown 1,4,7-trithiacyclononane (TTCN) and gold nanoparticles (AuNPs) with potentiometric stripping analysis (PSA), and obtained an LOD of 8 pM, more importantly, the modified electrode displays a 15-fold enhancement in the PSA signal (dt/dE) compared to a conventional graphene paste electrode. Using SWV, Ohsaka et al. [15] found a limit of detection of 0.28 ppb with a sensitivity of $27.27 \pm 0.01 \mu\text{A cm}^{-2} \mu\text{M}^{-1}$ for As(III) in 0.1 M phosphate buffer (PB) solution (pH 1) on a Au(111)-like poly-Au electrode. Dar

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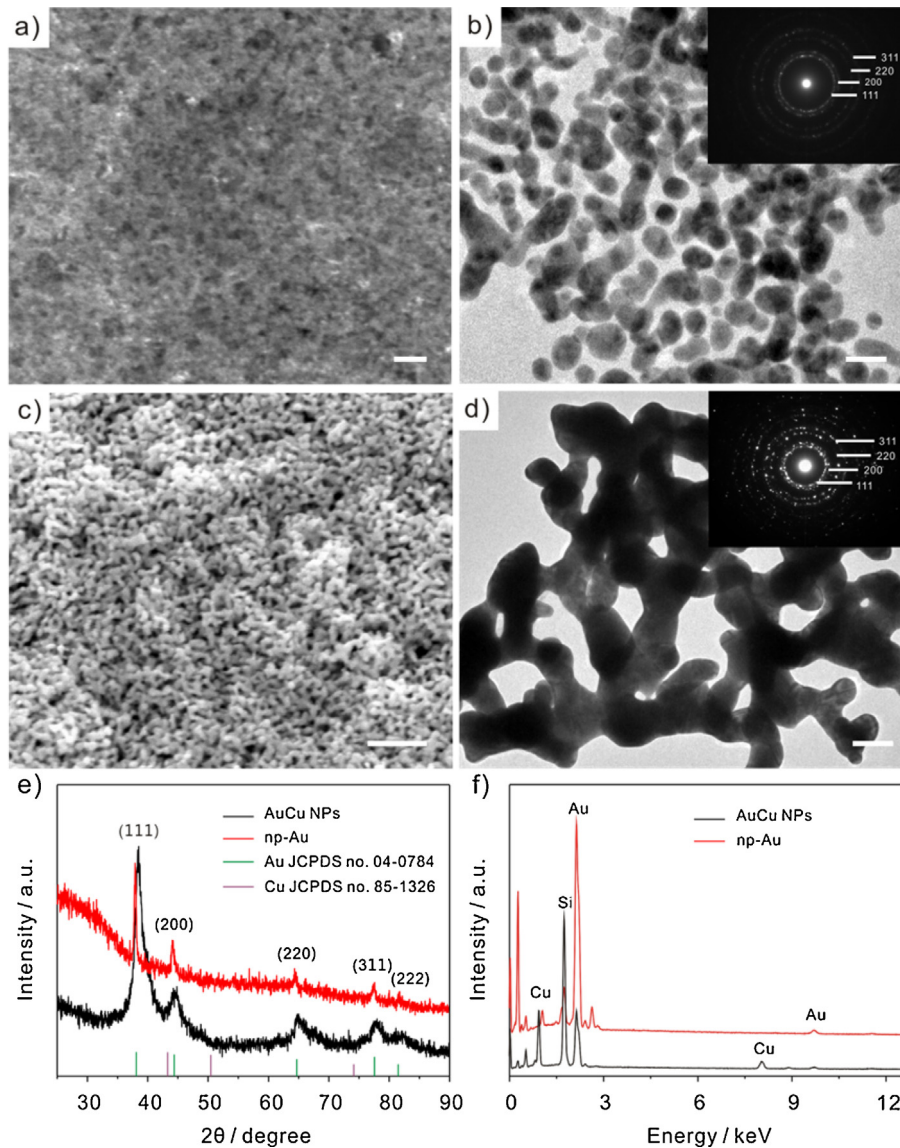


Fig. 1. Structures study of AuCu NPs and np-Au. a) and c) SEM images of the AuCu NPs and np-Au prepared through the standard procedure. b) and d) TEM images of the AuCu NPs and np-Au prepared through the standard procedure. e) XRD pattern of the AuCu NPs and np-Au. f) EDS spectrum of the AuCu NPs and np-Au. The insets in panel b and d are corresponding SAED pattern. Scale bar, a) and c) 500 nm, b) and d) 50 nm, respectively.

et al. [27] measured As(III) on silver nanoparticle (AgNPs)-graphene oxide (GO) composite modified glassy carbon electrode (GCE) in 0.1 M H_2SO_4 solution by SWV (square wave voltammetry), and obtained a sensitivity of $180.5 \mu\text{A} \mu\text{M}^{-1}$ with a LOD of 0.24 nM. Raj et al. [24] used a gold nanoelectrode ensembles (GNEEs) to determine As(III) by SWV in 1 M HCl media and obtained an LOD of 0.02 ppb with a sensitivity of $235.5 \mu\text{A} \mu\text{M}^{-1}$. Compton's group [5] investigated As(III) detection by SWV in 1 M HCl media on a gold nanoparticle modified GCE giving an LOD of 4.4 ppb with a sensitivity of $95 \mu\text{A} \mu\text{M}^{-1}$. Chen et al. [25] determined As(III) on a gold-nanoparticle embedded nafion composite modified on glassy carbon electrode using SWV in a medium containing 0.1 M EDTA and 0.1 M PB buffer (pH 5.0) and achieved an LOD of 0.047 ppb with a sensitivity of $23.98 \mu\text{A} \mu\text{M}^{-1}$. Huang et al. [28] measured As(III) in 0.1 M PB aqueous solution with 0.01 M EDTA by SWV and obtained an LOD of 0.0025 ppb with a sensitivity of $16.15 \mu\text{A} \mu\text{M}^{-1}$ on Au NPs modified GCE. Xiao et al. [16] detected As(III) in 0.1 M HCl media on a Au NPs modified carbon nanotubes and an LOD of 0.1 ppb but more importantly a sensitivity of $1985 \mu\text{A} \mu\text{M}^{-1}$ was obtained with SWV. Hossain et al. [29] prepared gold nanoparticle-modified

GCE by electrodeposition for As(III) determination in 3 M HCl by LSV (linear sweep voltammetry) with an LOD of 1.8 ppb and a sensitivity of $320 \mu\text{A} \text{cm}^{-2} \mu\text{M}^{-1}$. Other approaches include that of Huang and coauthors [30], who used Au micro wire electrodes to determine As(III) in $\text{N}_2\text{H}_4 \cdot 2\text{HCl}$ electrolyte (pH 0.5) by SWV obtained an LOD of 5.01 ppb and a sensitivity of $35.4 \text{nA} \mu\text{M}^{-1}$. Zen et al. [31] reported a poly(L-lactide) stabilized gold nanoparticles (designated as PLA-Au^{NP}) modify a disposable SPE (screen-printed carbon electrode) for the detection of As(III) by DPASV (differential pulse anodic stripping voltammeter) in 1 M HCl media and obtained an LOD of 0.09 ppb with a sensitivity of about $6.43 \mu\text{A} \mu\text{M}^{-1}$. Giacomino et al. [8] detected As(III) in 0.25 M HCl media on a lateral gold electrode with an LOD of 0.06 ppb. Through careful investigation, we found that most of the reported on As(III) detection using Au or Au-based electrodes under strongly acidic media (such as HCl, H_2SO_4 , HNO_3), which could suffer the problems from generating toxic arsine gas and the interference from H_2 evolution. Furthermore, the major problems associated with the available Au or Au-based electrodes are the interference from some commonly coexisting ions such as Cu(II), Hg(II) present in the real water sample, and the interference

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