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Colorimetric detection of glucose based on gold nanoparticles coupled with silver nanoparticles

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ABSTRACT

We have coupled gold nanoparticles (AuNPs) with silver nanoparticles (AgNPs) to assemble a plasmonic sensing platform for colorimetric detection of glucose. In this system, small AuNPs (~4 nm) can act as glucose oxidase (GOD) mimic enzyme to catalytically oxidize glucose in the presence of oxygen, producing hydrogen peroxide, which dissolves AgNPs to lead the color changes. Glucose can be detected not only by naked eyes (from yellow to red) but also by spectrophotometer in the concentration range of 5–70 μM, with detection limit of 3 μM. More importantly, we found that L-cysteine added in the system can markedly improve the selectivity for the detection of glucose. The proposed method was used to application for the detection of glucose in human serum with satisfactory results. This system is simple and low cost without using any enzymes and organic chromogenic agents.

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1. Introduction

Glucose is a very important molecule in metabolic homeostasis, energy source and human bodily functioning [1,2]. Especially, the blood glucose is one of the important indicators of the health of human, especially for patients with diabetes. Therefore, glucose detections have attracted much attention in the research and application because diabetes is a major health problem in the world [3–5]. In many technologies, colorimetric method has attracted great attention due to its simplicity, low-cost instruments, portability, and practicality. The typical colorimetric detection of serum glucose relies on the use of glucose oxidase (GOD) [4,6]. GOD catalyzes the oxidation of glucose by oxygen to produce gluconic acid and hydrogen peroxide (H_2O_2). The product of H_2O_2 is measured by oxidation of chromogenic agents in the presence of peroxidase. However, natural enzymes have some serious shortcomings [7,8], such as high expense, insufficient stability, and easy inhibition of the catalytic activity.

In recent years, a wide variety of nanomaterials have been developed in the application for the colorimetric detection of glucose because of their particular characterization [4,9,10]. For examples, some of nanomaterials, such as Fe_3O_4 [8], $ZnFe_2O_4$ magnetic nanoparticles [11,12], nitrogen-doped grapheme quantum dots [13], C_{60}-carboxyfullerenes [14], graphite-like carbon nitrides [15], and CuS nanoparticles [16], exhibit intrinsic peroxidase like activity similar to that found in HRP and apply to colorimetric detection of glucose with help of GOD. Moreover, gold nanoparticles (AuNPs) and silver nanoparticles (AgNPs) were also used as chromogenic agent to sensing H_2O_2 [17,18].

Among the nanomaterials, AuNPs and AgNPs have been greatly applied in optical sensing for detection of glucose [19–22]. The main methods for colorimetric detection of glucose includes: (1) Formation of metal nanoparticles reduced directly by glucose in an alkaline medium and high temperature (70-80 °C) [23,24]; (2) Aggregation/anti-aggregation of metal nanoparticles by concanavalin A (Con A, a well-known glucose binding protein) [25,26]; (3) Color change of AuNPs based on the decreasing pH value by the production of gluconic acid in the glucose and GOD reaction [27]. (4) Measurement of the H_2O_2 product in the GOD-catalyzed system by the aggregation of gold nanoparticles (Au NPs) [28,29], etching of gold or silver nanoparticles [30–34], and enlargement of Au NPs [6,35].

Recently, it was reported that small Au NPs (~3.5 nm) were possessed intrinsic GOD-like activity, which also catalyzes aerobic oxidation of glucose to generate gluconic acid and H_2O_2 [36–38]. On the basis of this mimic GOD, Fan et al. developed colorimetric method for detection of glucose using AuNPs coupled with horseradish peroxidase (HRP) and organic chromogenic agent [39]. Liu et al. also investigated this glucose oxidation reaction based on nanoceria but extremely toxic substance of KCN was used to dissolve the AuNPs in the process [40]. In this work, we report a simple colorimetric method for detection of glucose based on small Au NPs coupled with silver nanoparticles (Ag NPs). We also demonstrated that the proposed method can be used to detection of serum glucose in clinic samples. The sensing mechanism is exhibit in Scheme 1. In the presence of oxygen, Au NPs catalyze the oxidation of glucose to form gluconic acid and H_2O_2. Then the Ag NPs

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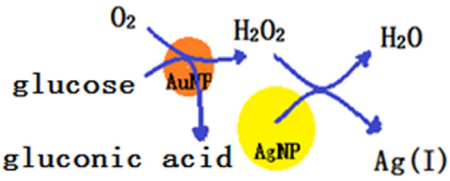

Scheme 1. Schematic illustration of the reactions for colorimetric detection of glucose based on Au NPs coupled with Ag NPs.

added in the solution are etched by the product of H_2O_2, which induces a change of the localized surfaceplasmon resonance (LSPR) band strength [31,33]. The original color of the AuNPs-AgNPs solution was yellow, which mainly come from the color of AgNPs. The yellow of AgNPs was gradually fading with etching of the AgNPs. When AgNPs were completely dissolved by the H_2O_2 which was produced by the oxidation of glucose, the red color of AuNPs was observed. Therefore, this method provides a colorimetric detection of glucose using the couple of Au NPs and Ag NPs without any enzymes and organic chromogenic agents.

2. Experimental Section

2.1. Materials

Silver nitrates ($AgNO_3$) was purchased from shanghai reagent factory (China). Chloroauric acid ($HAuCl_4 \cdot 4H_2O$), sodium citrate ($Na_3C_6H_5O_7 \cdot 2H_2O$), citric acid ($C_6H_8O_7$), disodium hydrogen phosphate(Na_2HPO_4), sodium borohydride ($NaBH_4$), hydrogen peroxide (H_2O_2), glucose, fructose, L-cysteine, ascorbic acid (AA), uric acid(DA), paracetamol (PA), bovine serum albumin (BSA), glycine, were acquired from Sinopharm Chemical Reagent Co., Ltd. (China). All chemicals were of analytical grade. All aqueous solutions were prepared using ultrapure water (18.2 MΩ cm) obtained by a Milli-Q system.

2.2. Apparatus

An Agilent Cary-60 spectrophotometer was used to record the UV–vis absorption spectra. TEM images were taken using a Tecnai G20 (FEI, U.S.A.) transmission electron microscope operated at 200 kV. Ultrasonic cleaner from Kunshan ultrasonic instrument company was used for dispersing solutions.

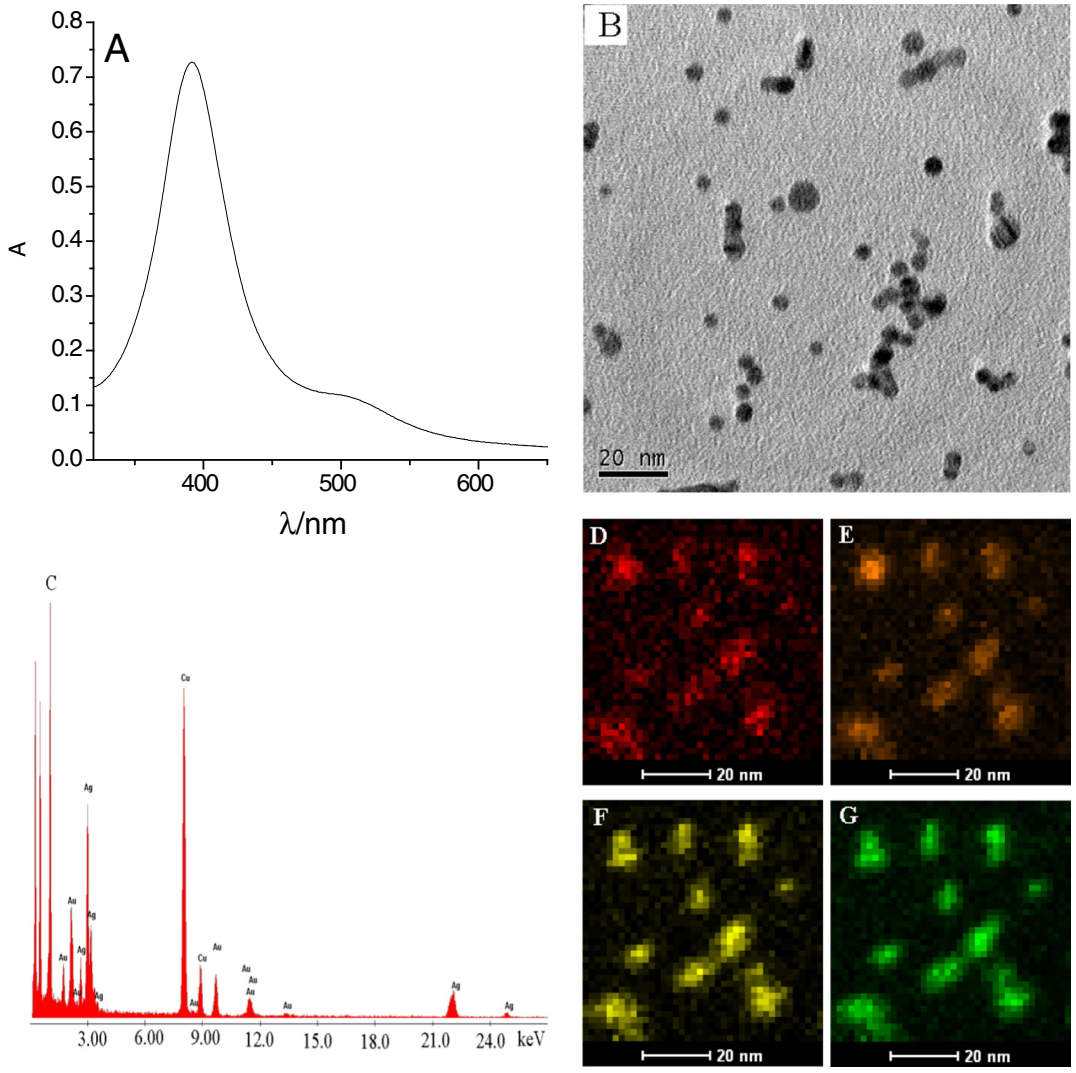

Fig. 1. The UV–vis absorption spectrum of the AuNPs-AgNPs solution (A), TEM image of the AuNPs-AgNPs (B), the EDX pattern of AuNPs-AgNPs (C), the EDX elemental mapping images of Ag-K (D), Ag-L (E), Au-L (F), and Au-M (G).

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