



Visual and light scattering spectrometric method for the detection of melamine using uracil 5'-triphosphate sodium modified gold nanoparticles

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ARTICLE INFO

Article history:

Received 11 April 2016

Received in revised form 22 August 2016

Accepted 25 August 2016

Available online 27 August 2016

Keywords:

Melamine

Gold nanoparticles

UTP

Hydrogen-bonding

ABSTRACT

A highly selective method was presented for colorimetric determination of melamine using uracil 5'-triphosphate sodium modified gold nanoparticles (UTP-Au NPs) in this paper. Specific hydrogen-bonding interaction between uracil base (U) and melamine resulted in the aggregation of AuNPs, displaying variations of localized surface plasmon resonance (LSPR) features such as color change from red to blue and enhanced localized surface plasmon resonance light scattering (LSPR-LS) signals. Accordingly, the concentration of melamine could be quantified based on naked eye or a spectrometric method. This method was simple, inexpensive, environmental friendly and highly selective, which has been successfully used for the detection of melamine in pretreated liquid milk products with high recoveries.

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1. Introduction

Melamine (1, 3, 5-triazine-2, 4, 6-triamine, $C_3H_6N_6$), containing 66% nitrogen by mass, is a synthetic compound, which has been commonly used in the production of polymer resins and plastics for tableware, laminates, coatings, commercial filters or can lining [1,2]. However, due to its low cost and high nitrogen level, melamine was fraudulently added to milk, infant formula and other related food to increase apparent protein content, as protein concentrations are typically measured by analysis of nitrogen [3,4]. Although the toxicity of melamine is low, the excessive intake will lead to the formation of insoluble cyanurate crystals in kidneys and finally cause renal failure in animals and humans [5]. The death of thousands of dogs and cats in the USA in 2007, and the occurrence of kidney stones in thousands of infants across China in 2008 were two examples resulted directly from the consumption of milk, infant formula or related products adulterated with melamine [2, 4,6]. Therefore, determination of melamine is of biological, clinical and food industry importance, and a reliable method is needed to examine melamine residues in food and particularly in dairy products for children.

Currently, several methods have been used for the detection of melamine, including gas chromatography-mass spectrometry (GC-MS) [4], liquid chromatography-mass spectrometry (LC-MS) [7,8], high-

performance liquid chromatography (HPLC) [1], capillary electrophoresis (CE) [9,10], electro-chemiluminescence [11] and electrochemical method coupled with enzyme colorimetric assay [12]. Most of above methods require expensive apparatus. Furthermore, other methods, such as surface enhanced Raman spectroscopy (SERS) [13–16], fluorescence spectroscopy [17,18], surface plasmon resonance [19], nuclear magnetic resonance (NMR) spectroscopy, dynamic light scattering (DLS) [20] and enzyme-linked immunosorbent assay (ELISA) [21], are also emerging. However, some of these methods are time consuming and labor-intensive because of the complex pretreatments containing extraction, purification, derivatization and so on. Hence, a simple, rapid, low-cost and highly selective analytical method to detect melamine is of particular importance.

Due to the ease of preparation, biocompatibility and stability, gold nanoparticles (Au NPs) have attracted considerable attention [14,22, 23]. Especially, Au NPs owns interesting physical and chemical properties, and has potential applications that are quite different from those of bulk solid [24]. In the visible wavelength range, their localized surface plasmon resonance absorption (LSPR-A) and localized surface plasmon resonance light scattering (LSPR-LS) features are strong with extremely high extinction coefficients ($\sim 3 \times 10^{-11} \text{ cm}^{-1}$) [2]. The wavelength corresponding to the extinction maximum of the SPR is highly dependent on the sizes, shapes, and dielectric properties of the metal nanoparticles [25]. Consequently, the color of Au NPs can change from wine red (dispersion state) to blue (aggregation state) and the light scattering intensity can be enhanced [26]. Based on this unique characteristic, the

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results can be directly observed by naked eye, therefore offering a simple and promising method used for on-site screening melamine contamination in milk products or at home diagnosis. With this excellent property of Au NPs, our group has reported one novel method with polythymine-stabilized AuNPs for the visual detection of melamine on the basis of the formation of triple H-bonds between thymine and melamine through stepwise reactions [26]. Similar with thymine base, the uracil base (U) can also form the triple H-bonds with melamine due to the diimide moiety [27]. Thereby, on the basis of the above work, we further established a new and simple visual and light scattering spectrometric method for melamine assay based on uracil 5'-triphosphate sodium modified gold nanoparticles (UTP-AuNPs). After the addition of melamine, the UTP-AuNPs solution showed a color change from red to blue. Other nucleotides modified Au NPs couldn't show the similar color change, demonstrating the specific response of UTP-AuNPs to melamine. Comparing with our previously reported method [26], this proposed method is much simpler because the detection of melamine could be completed by one step. Up to now, although Au NPs have been modified with p-nitroaniline [28], cyanuric acid (CA) [27], 4-mercaptopyridine [29], 2, 4, 6-trinitrobenzene sulfonic acid and uracil-5-carboxylic acid [30] for the detection of melamine, these modification methods could introduce the environmental toxicity or biological hazards. Herein, as one vital member of the nucleotides, UTP is environmental friendly for the detection of melamine in real samples, displaying the merit of this method. Besides, it exhibited excellent selectivity against other interferences. To testify its practicality, the present method has been further applied for the detection of melamine in real samples successfully.

2. Experimental

2.1. Apparatus

Localized surface plasmon resonance light scattering (LSPR-LS) signals and extinction spectra were measured with F-4500 spectrofluorometer and UV-3010 spectrophotometer (Hitachi Ltd., Tokyo, Japan), respectively. Scanning electron microscopic (SEM) measurements were performed on an S-4800 electron microscope (Hitachi Ltd., Tokyo, Japan). A Zetasizer Nano-ZS90 instrument (Malvern Inc.) was used to detect the size of the aggregation species in solution based on the dynamic light scattering (DLS) principle. AQL-901 vortex mixer (Haimen, China) was employed to blend the solutions in 1.5-mL tubes. pH of the test solution was measured with Oakton pH 510 meter (Singapore).

2.2. Materials

Melamine was commercially purchased from J&K Chemical Ltd. (Beijing, China). Stock solution was prepared by dissolving melamine

with 5.0 mL methanol firstly and then diluted to the final concentration of 3.0×10^{-4} M with water. Guanosine 5'-triphosphate sodium salt (GTP), adenosine 5'-triphosphate sodium salt (ATP), cytosine 5'-triphosphate sodium salt (CTP) and uracil 5'-triphosphate sodium salt (UTP) were purchased from Sigma-Aldrich (St. Louis, MO, USA). Stock solutions of GTP, ATP, CTP and UTP were prepared by dissolving the sodium salts in water and diluted to the final concentration of 3.0×10^{-4} M. All the nucleic acids solutions were stored in refrigerator at 4 °C and used within one month. HEPES buffer (50 mM, pH 7.2) was used to control the acidity. All reagents were analytic grade and used without further purification. Milli-Q purified water (18.2 Ω M) was used for all sample preparations.

2.3. Preparation of citrate-stabilized Au nanoparticles (CT-AuNPs)

The CT-AuNPs were prepared according to previous references by reducing $\text{HAuCl}_4 \cdot 4\text{H}_2\text{O}$ with citrate [26,31]. Briefly, 48 mL of Milli-Q purified water was mixed with 2 mL of 1% (w/w) HAuCl_4 solution to make the final concentration of $\text{HAuCl}_4 \cdot 4\text{H}_2\text{O}$ be 1 mM. The mixture was then heated under magnetic stirring until it began to boil, and 1 mL of 5% trisodium citrate was added to the solution. Under continuous stirring and boiling, the color of the mixture gradually changed to deep red within 3 min. After boiling for another 5 min, the solution was cooled to room temperature (25 °C) under vigorous magnetic stirring, and was filtered by 0.22 μm filter membrane. After that, this solution was transferred for the UV-vis absorption and SEM measurements, and then stored in 4 °C refrigerator. The size of prepared CT-AuNPs was about 13 nm.

2.4. Surface modification of CT-AuNPs with nucleotides

For the surface modification of CT-AuNPs with mononucleotides, an aliquot of 30 μL (2.0 mM) UTP, CTP, ATP or GTP was added to 970 μL AuNPs colloidal solutions according to previous references [32]. After incubating for 3 h at 4 °C, the resulting mixture was centrifuged at the speed of 12,000 rpm for 30 min, and then the supernatant fluid was removed. The obtained AuNPs precipitate was dissolved with 400 μL Milli-Q H_2O . Finally, these nucleotides-modified AuNPs solutions were stored in 4 °C refrigerator for further use.

2.5. Sample preparation

The liquid milk bought from a local super market was pretreated according to the general procedure. Firstly, trichloroacetic acid and methanol solution were added to the liquid milk to eliminate proteins and extracted analyte, then the mixture was centrifuged at 10,000 rpm for 4 min after 15 min sonication and 10 min shaking, and the supernatant was filtrated. Lastly, the filtrate was concentrated and filtered through a 0.45 μm microporous membrane.

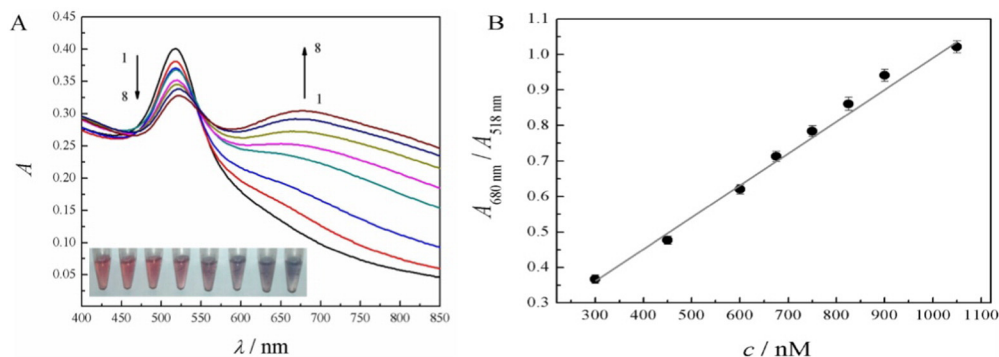


Fig. 1. (A) The absorption spectra of UTP-AuNPs upon the addition of increasing melamine (inset was the color change of gold nanoparticles); (B) the linear calibrated curve between the intensity ratios of 680 nm to 518 nm and the concentrations of melamine. Concentrations: UTP-AuNPs, 1.5 nM; melamine (Curves 1–8, nM): 0, 300, 450, 600, 675, 750, 825, 900; pH 7.2. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

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