Food Chemistry 174 (2015) 473–479

Contents lists available at ScienceDirect

Food Chemistry

journal homepage: www.elsevier.com/locate/foodchem

Analytical Methods

Colorimetric detection of melamine based on methanobactin-mediated synthesis of gold nanoparticles

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ARTICLE INFO

Article history:
Received 17 April 2014
Received in revised form 21 October 2014
Accepted 16 November 2014
Available online 23 November 2014

Keywords:
Methanobactin
Gold nanoparticles
Melamine
Colorimetric detection
Liquid milk

ABSTRACT

A simple and rapid field-portable colorimetric method for the detection of melamine in liquid milk was reported. Methanobactin (Mb) could reduce Au (III) to Au (0) and mediate the synthesis of gold nanoparticles (Au-NPs). Upon the addition of melamine, melamine interacted with oxazolone ring of Mb, which interrupted the formation of Au-NPs. Melamine could also stimulate the aggregation of formed Au-NPs. In this paper, these characteristics have been used to detect melamine in liquid milk by naked eyes observation with a detection limit of 5.56×10^{-6} M (0.7 mg/kg). Further, the plasmon absorbance of the formed Au-NPs allowed the quantitative detection of melamine by UV–vis spectrometer. A linear correlation was existed between the absorbance and the melamine concentration ranging from 3.90×10^{-7} M to 3.97×10^{-6} M with a correlation coefficient of 0.9685. The detection limit (3σ) obtained by UV–vis spectrum was as low as 2.38×10^{-7} M (i.e., 0.03 mg/kg).

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1. Introduction

Melamine (1,3,5-triazine-2,4,6-triamine, $C_3H_6N_6$) is a triazine heterocyclic organic compound, which is widely used in the production of melamine resins, flame retardants, fertilizer and other products. Because of the high nitrogen content (66% nitrogen by mass), melamine was illegally and unethically adulterated in protein-rich ingredients to increase the apparent protein content measured by Kjeldahl protein analysis (Mauer, Chernyshova, Hiatt, Deering, & Davis, 2009). Melamine can be hydrolyzed to cyanuric acid which in turn associates with melamine to form insoluble complexes, resulting in the formation of insoluble crystals in the kidneys and subsequent tissue injury (Reimschuessel & Puschner, 2010). It is not allowed to use as a food additive in human food or animal feeds. A safety limit of melamine (2.5 mg/kg) in milk and milk-based products was set by Food and Drug Administration (FDA) of USA and European Union (Zhu, Gamez, Chen, Chingin, & Zenobi, 2009). Therefore, there is an increasing need for rapid and reliable methods to detect melamine in milk and other food products. Currently there are a number of analytical techniques for detecting melamine in milk and milk-based products. Amongst them, the most common methods are high performance liquid chromatography (HPLC) (Ehling, Tefera, & HO, 2007), mass spectrum (MS) (Yang et al., 2009), gas chromatography/mass spectrum (GC/MS) (Yokley, Mayer, Rezaaiyan, Manuli, & Cheung, 2000), high performance liquid chromatography/mass spectrum (HPLC/MS) (Kim et al., 2008), capillary zone electrophoresis/mass spectrum (CE/MS) (Vo et al., 2008), surface enhanced Raman spectroscopy (SERS) (Lin et al., 2008) and fluorescence polarisation immunoassay (Wang et al., 2011) etc. Although these methods have high sensitivity, many of them require expensive apparatus (such as MS) and time-consuming sample pretreatment (such as derivatisation or extraction) and are not readily adaptable to on-site detection.

UV-spectrophotometry is also a convenient and widely used method for quantitative analysis. However, the spectrophotometric method requires the analyte presented in the given sample to have different absorption spectrum from other unknown coexistent substances with low spectral overlapping. If the method is used to determine melamine in milk directly, the problem seems more intractable. Numerous nutrition components in milk could interfere the quantitative determination of melamine (Liu, Deng Jian, Liang, Chen, & Wang, 2011).

Recently, gold nanoparticles (Au-NPs) have been widely used as colorimetric probes for chemical sensing and biosensing because of their solution colour, strongly size-dependent and distance-dependent optical property, and extremely high extinction coefficients (10^8–10^{10} M^{-1} cm^{-1}, which is about 3–5 orders of

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http://dx.doi.org/10.1016/j.foodchem.2014.11.098
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magnitude higher than common organic molecules) (Jin, Wu, Li, Mirkin, & Schatz, 2003; Kuang et al., 2011). There are a few reports on the colorimetric detection of melamine based on the fact that Au-NPs are induced to aggregate by interparticle crosslinking (Ai, Liu, & Lu, 2009; Chi, Liu, Guan, Zhang, & Han, 2010; Kuang et al., 2011). However, the complex modification of nanoparticles limits their potential application. More simple colorimetric methods for the detection of melamine by unmodified nanoparticles were proposed by Li, Li, Cheng, and Mao (2010). But the tedious sample preparation by solid phase extraction is disadvantageous for on-site detection. Colorimetric detection of melamine during the formation of gold nanoparticles has also been reported. However, most of them are based on the interaction between melamine and reducer, which leading to weakening of the Au (III) reducing ability (Cao et al., 2010).

Methanotrophs are a group of gram-negative eubacteria that utilise methane as the sole carbon and energy source (Hanson & Hanson, 1996). It is known that the amount of bioavailable copper regulates the methane monooxygenase (MMO), which is used by methanotrophs to oxidise methane (Hakemian & Rosenzweig, 2007). Methanobactin (Mb) is a copper-binding small peptide that appears to function as an agent for copper sequestration and uptake in methanotrophs (Balasubramanian & Rosenzweig, 2008). The crystal structure of copper-loaded Mb (Cu–Mb) from *Methylosinus trichosporium* OB3b revealed a 1217 Da molecule with a chemical composition of $C_{45}N_{12}O_{14}H_{62}S_5Cu$ (Balasubramanian & Rosenzweig, 2008). Mb can coordinate a single Cu (II) ion by its nitrogens from two oxazolone rings and sulfurs from two enethiol groups and then reduce Cu (II) to Cu (I) (Choi et al., 2006). Mb can also bind and reduce Au (III) to Au (0), which result in the formation of Au-NPs (Xin et al., 2013).

In this paper, we have developed a colorimetric method for the detection of melamine in liquid milk based on the phenomena that melamine can interfere the formation of Au-NPs by Mb. The detection of melamine can be achieved during the formation of Au-NPs, i.e., the synthesis of Au-NPs and the analysis of melamine can occur in one-step just by using Mb as mediator for the generation Au-NPs. This one-step synthesis offers fast, sensitive and convenient colorimetric detection of melamine with the naked eyes or UV–vis spectroscopy within 50 min. The proposed scheme reduces the volume of waste generated during Au-NPs synthesis and purification, and can be an alternative means for onsite detection of melamine without costly instruments.

2. Materials and methods

2.1. Chemicals and materials

Melamine was obtained from Sigma -Aldrich (St. Louis, MO, USA). Chloroauric acid ($HAuCl_4{\cdot}4H_2O$) was obtained from Sinopharm Chemical Reagent Co. Ltd. (Shanghai, China). Other chemicals were of analytical reagent grade and used without further purification. All stock solution were prepared daily with distilled water. The market milk products were bought from local supermarket. The general composition was showed below:

Lipids: 3.3 g/100 g; milk solids-not-fat (MSNF): 8.1 g/100 g; proteins: 2.9 g/100 g (market milk from Inner Mongolia Meng Niu Dairy Co., Ltd. China).

Lipids: 3.4 g/100 g; milk solids-not-fat (MSNF): 8.1 g/100 g; proteins: 2.9 g/100 g (Heilongjiang Province Wondersun Dairy Co., Ltd. China).

Lipids: 3.5 g/100 g; milk solids-not-fat (MSNF): 8.1 g/100 g; proteins: 3.0 g/100 g (Heilongjiang Province Wondersun Dairy Co., Ltd. China) (Inner Mongolia Yili Industrial Group Co. Ltd. China).

2.2. Organism and culture conditions

Methylosinus trichosporium 3011 was obtained from the Institute of Microbiology and Virology (Kiev. Ukraine) and was cultivated with a mineral salt medium according to Xin et al. (2010). Methanol was added to 0.2% (v/v) and supplied on-line to keep the same concentration. Cells were grown at 28–30 °C and an agitation rate of 250–300 rpm. Ambient air was bubbled through the fermentor continuously at 0.5–0.8 L/min. The cultures were grown to stationary phase for Mb production.

2.3. Isolation and quantification of Mb

Mb from the spent medium of *Methylosinus trichosporium* 3011 was isolated as previously described for *Methylococcus capsulatus* Bath by Choi et al. (2005). The cells were removed by centrifugation at 10,000g for 30 min. The supernate was loaded onto a 2.5 × 20 cm Diaion HP-20 column (Mitsubishi Chemical Holdings, Japan). The bound Mb was washed with two column volumes of H_2O and eluted with 40% methanol: 60% H_2O. The eluant was lyophilized for concentration and storage. The freeze-dried samples following chromatography on Diaion HP-20 columns were the source of Mb used in this study. The amount of Mb in the sample was quantified according to Xin et al. (2013).

2.4. Gold coordination

Gold coordination was performed using 0.1 mM aqueous solutions of Mb. Before gold coordination, 50 μL of 10 mM melamine or the same volume of dH_2O were added to 5 mL of Mb solutions and shaken for 1 min. Stock solution of $AuHCl_4$ (10 mM) was added gradually in 0.1 mM aqueous solutions of Mb, mixed and incubated for 10 min before spectral determinations. UV–visible absorption spectroscopy was performed by using a UV-2550 spectrophotometer (Shimadzu, Japan). Fluorescence excitation spectra were recorded on Hitachi F-7000 fluorescence spectrometer. The valence states of elements were analysed by using X-ray photoelectron spectroscopy (ESCALAB210 , VG Scientific). The samples for XPS were prepared by adding 0.2 mL aqueous solution of the Au (III) & Mb mixture onto glass plate and allowing water to completely evaporate. transmission electron microscope (TEM) technique was employed to visualise the size and shape of gold nanoparticles. The samples for TEM were prepared by adding 10 μL of a 1:5 (stock : H_2O) dilute solution of the sample onto carbon-coated copper grids and allowing water to completely evaporate. TEM images were recorded in a Hitachi H-7650 transmission electron microscope.

2.5. Synthesis of AuNPs and colorimetric detection of melamine

Au-NPs were synthesised according to previous method (Xin et al., 2013). Mb was added into the solution of $HAuCl_4$ (0.5 mM) and the mixture was further reacted at room temperature until the wine-red solution of Au-NPs was obtained. The detection of melamine was conducted under the optimal conditions for the synthesis of Au-NPs. First, In 5 mL centrifuge tube, 0.2 mL of Mb (1 mM) and 0.1 mL of different concentrations of melamine solutions were mixed in a 5 mL centrifugal tube, NaOH (0.1 M) was added to adjust pH in the range of 5.0–5.4 and kept for 1 min. Then, 0.1 mL of $HAuCl_4$ (10 mM) and 0.4 mL HQ (1 mM) were added to the tube. The mixture solution was diluted with dH_2O to 2–5 mL and was further incubated at room temperature for 50 min. The absorbance (at 539 nm) of the mixture solution in the presence and in the absence of melamine were recorded, respectively. The spiked-recovery detection of melamine in raw milk was manipulated in the same step.

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