Analytica Chimica Acta 845 (2014) 92–97

Contents lists available at ScienceDirect

Analytica Chimica Acta

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

Hydrogen-bonding recognition-induced aggregation of gold nanoparticles for the determination of the migration of melamine monomers using dynamic light scattering

Long Wu [1], Kun Chen [1], Zhicheng Lu, Tingting Li, Kang Shao, Feng Shao, Heyou Han *

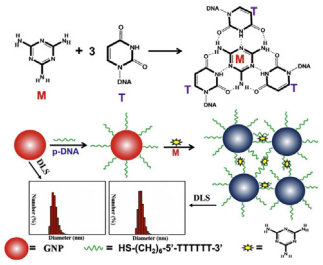
State Key Laboratory of Agricultural Microbiology, College of Science, Huazhong Agricultural University, Wuhan 430070, PR China

HIGHLIGHTS

- The p-DNA modified GNPs were synthesized to recognize melamine.
- The strategy was developed in the measurement of MMMQ by DLS technique.
- The influence of four solvents, temperature, and time on MMMQ was investigated.
- The method provides good linearity ($R \geq 0.9970$) and high sensitivity ($2\ \mu g\ L^{-1}$).

GRAPHICAL ABSTRACT

Melamine (M) can hybridize with thymine (T) through the hydrogen-bonding recognition. The combination of melamine with the p-DNA (containing 6 T) modified GNPs induces the aggregation of GNPs. The change in the size of GNPs can be distinguished by DLS technology with high sensitivity.



ARTICLE INFO

Article history:
Received 3 March 2014
Received in revised form 17 July 2014
Accepted 21 July 2014
Available online 24 July 2014

Keywords:
Migration of melamine monomers
Oligonucleotides
Gold nanoparticles
Dynamic light scattering
Melamine tableware

ABSTRACT

The migration of melamine monomers from food contact materials has aroused particular attention since the 2008 melamine-tainted milk scandal in China. However, the determination of melamine monomer's migratory quantity (MMMQ) has remained an open question because of the complex sample pretreatment and the low sensitivity. Based on the hydrogen bonding interaction between DNA thymine and melamine, this paper described a simple and rapid method focusing on the measurement of MMMQ from melamine tableware by gold nanoparticles (GNPs) and dynamic light scattering (DLS). With the presence of probe DNA (p-DNA), the GNPs were stable in NaCl solution (0.06 M), whereas they became aggregated when the p-DNA hybridized with melamine. The change in the hydrodynamic diameter of GNPs could be detected by DLS technology. Under the optimal conditions, the average diameter increased linearly with the concentration of melamine over the range from 5.0 to 320.0 $\mu g\ L^{-1}$, and showed a detection limit of 2.0 $\mu g\ L^{-1}$ (3σ/slope). The MMMQ was investigated within a range from 6.00×10^{-4} to 2.58×10^{-1} mg dm^{-2} ($n \geq 3$) in four different food simulants at different temperatures and time points. The results suggest that the DLS method has great potential in the analysis of the migration of melamine monomers.

1. Introduction

Melamine (1,3,5-triazine-2,4,6-triamine, $C_3H_6N_6$) is an industrial chemical frequently applied in the production of melamine-formaldehyde resins and the manufacture of durable plastics [1]. Owing to their advantages of being light, cheap, durable, and easy to clean, the melamine resins were widely used at home and

* Corresponding author. Tel.: +86 027 87282043; fax: +86 027 87282043.
E-mail address: hyhan@mail.hzau.edu.cn (H. Han).
[1] These authors contributed equally to this work.

http://dx.doi.org/10.1016/j.aca.2014.07.036
0003-2670/

restaurants in the form of cups and plates [2]. Such plastic products, mainly made from melamine and formaldehyde by polycondensation, are known as melamine tableware. In many cases, residual melamine monomers can remain in the plastic articles after polymerization [3]. Besides, the melamine could also be released from the finished articles with the breakdown of polymers and migrate to the foodstuffs, which would be harmful to people's health with constant intake [4,5]. Recently, due to a series of food poisoning incidents such as 2008 "Sanlu milk powder incident" in China, food safety concerns including the MMMQ have became the focus of attention worldwide [6,7]. The migration of melamine monomers from food contact materials was strictly regulated in many countries. To our knowledge, the MMMQ allowed in European Union is less than 5 mg dm^{-2} (EC 2002) and not above 0.2 mg dm^{-2} in China [8,9].

The study for the migration of melamine monomers is still in the infancy, and only a few articles have been reported on the determination of MMMQ, making the availability of such information difficult for more readers [10,11]. To detect melamine in various matrices, many methods have been developed, such as gas chromatography/mass spectrometry (GC/MS) [12], high performance liquid chromatography (HPLC) [13], liquid chromatography with tandem mass spectrometry (LC–MS/MS) [14], and the indirect competitive enzyme-linked immunosorbent assay (IC-ELISA) method [15]. However, these methods are either labor-intensive or time-consuming (more than 15 min per sample), or lacking sensitivity, and almost none of them were applied to measure the MMMQ because of their complex sample pretreatment procedures. Thus, it is imperative to develop a simple and sensitive approach to detect the MMMQ from melamine tableware. DLS, also known as photon correlation spectroscopy, can be an alternative method. DLS is a technique used widely in the particle size and size distribution studies [16]. This technique coupled with GNPs was employed extensively as an ultra-sensitive assay for chemical and biological detection [16–18].

Melamine is known as a noted molecular recognition module in many contexts [19–21]. It was demonstrated in some papers that the reaction of thymine (T) and melamine (M) could form stable complex by NH—O and NH—N hydrogen bonds [22,23]. The interaction of the triple hydrogen bonding was considered to be specific and directional [24]. Based on the recognition model, we constructed a new route to detect the migration of melamine monomers (see Fig. 1). As shown in Fig. 1(B), the single-strand DNA (p-DNA) (HS-$(CH_2)_6$-5′-T6-3′) was firstly modified onto the GNPs surface, and then the p-DNA modified GNPs began to aggregate when hybridized with melamine. The aggregation of GNPs grew larger with the increase of the concentration of melamine. The relationship between the size change of GNPs and the concentration of melamine was investigated by DLS. Thus, the migration of melamine monomers can be detected using DNA functionalized GNPs and DLS technology. This work focused on the development of a simple method of testing the melamine tableware and measuring their MMMQ in different food simulants.

2. Experimental

2.1. Reagents and chemicals

Melamine (≥99% purity), formaldehyde solution (analytical reagent), acetic acid (≥99.8%), ethanol (≥99.7%), isopropanol

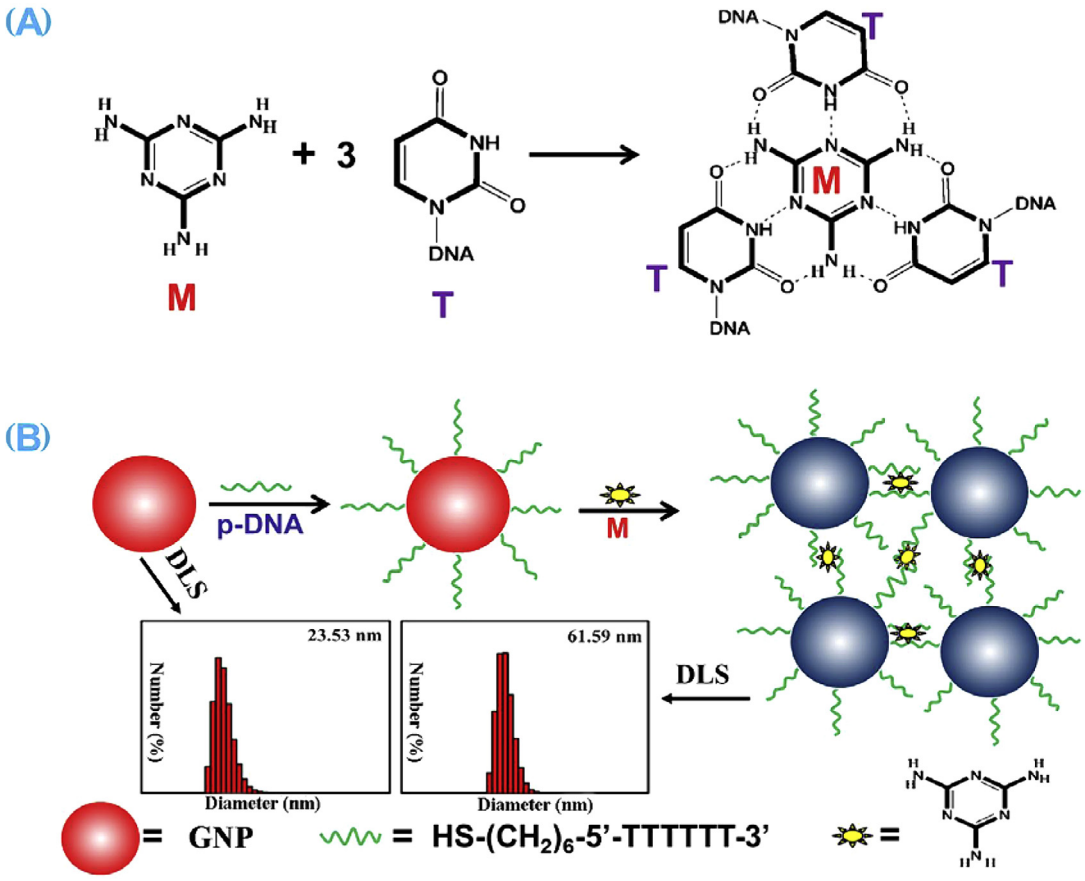


Fig. 1. Illustration of the detection of melamine using GNPs probes and DLS, (A) hydrogen-bonding recognition between melamine (M) and thymine (T), and (B) the aggregation of the p-DNA modified GNPs recognized with melamine.

(≥99.7%) and olive oil were purchased from Sinopharm Chemical Regent Co., Ltd. (Shanghai, China). The melamine formaldehyde resin food containers (bottles) and polystyrene plastic bottles were purchased from a local school supermarket. The oligonucleotide (HS-$(CH_2)_6$-5′-T6-3′) was obtained from Augct Biotechnology Company (Beijing, China). The oligonucleotide stock solution was prepared using phosphate buffer solution (pH 6.5). $HAuCl_4 \cdot 4H_2O$ was obtained from Shanghai Chemical Reagent Co., Ltd. (Shanghai, China). Sodium citrate and sodium chloride (NaCl) were purchased from Tianjin Chemical Reagent Company (Tianjin, China). Other reagents and chemicals were of analytical grade. Ultrapure water (Mill-Q, Millipore, 18.2 MΩ cm) was used throughout the experiments.

2.2. Instruments

The UV–vis absorption spectra were acquired on a Thermo Nicolet Corporation Model Evolution 300 spectrophotometer coupled with a 1.0 cm quartz cell. The transmission electron microscopy (TEM) and high-resolution transmission electron microscopy (HRTEM) images were obtained using a FEI Tecnai G20 transmission electron microscope. The hydrodynamic size was measured on Malvern Zetasizer Nanoseries with 633 nm laser excitation at 25 °C.

2.3. Preparation of GNPs

All glassware used in this experiment was thoroughly cleaned in a bath of freshly prepared aqua regia solution (HCl:HNO_3, 3:1), and then rinsed with ultrapure water and oven-dried. The synthesis of GNPs was performed by reducing $HAuCl_4$ with citrate as described in our previous report [25]. Briefly, 40 mL of 0.6 mM $HAuCl_4$ solution was heated to boiling, followed by addition of 10 mL of 1% (w/w) sodium citrate solution under vigorous stirring, resulting in a color change from light yellow to wine red, and then the solution was boiled for another 5 min to complete the reduction. After cooling to room temperature, the final solution was stored at 4 °C. The concentration of GNPs was calculated to be 0.8 nM as previously reported by Haiss' group [26].

2.4. Preparation of GNPs–DNA conjugates

Before the preparation of GNPs–DNA conjugates, we optimized the concentration of p-DNA and NaCl solution, and the pH value of sodium phosphate buffered saline (PBS) solution.

GNPs–DNA conjugates were prepared according to our previously reported method [25]. Briefly, GNPs (0.5 mL, 0.8 nM) were mixed with 2.5 μL p-DNA (17.6 mM) for 12 h under continuous stirring. The mixture solution was purified by centrifugation at 7000 rpm for 10 min and the pellets were re-suspended in PBS solution (0.01 M, pH 6.5) containing 0.06 M NaCl and stored at 4 °C for use.

2.5. Preparation of standard solutions

To calculate the MMMQ in different food storage conditions, linear equations were formulated from calibration standards of melamine in different food simulants, which stand for the corresponding soaking-out solution [27]. The preparations of stock solution and different standard solutions are described in Supporting Information.

2.6. Exposure of sample to food simulants

This procedure was carried out by exposure of sample to different food simulants using the conventional direct immersion method [27]. The most commonly used solvents are considered to be water, alcohol, vinegar, and cooking oil. Thus, the food simulants used in this experiment were water, acetic acid (4%, v/v), ethanol (10%, v/v), and olive oil. Among them, water was defined as the aqueous solution (such as milk or coffee), acetic acid as the acid solution with causticity (for example, lemon juice or apple juice), ethanol as the volatile organic solution (such as wine), and olive oil as the oil solution.

In this report, the resin containers were respectively immersed in water, ethanol (10%, v/v), acetic acid (4%, v/v), and olive oil to obtain the MMMQ with different time periods, namely 0.5, 1, 2, 4, and 12 h, and different temperatures, namely 5, 20, 40, 70, 100, and 130 °C. Besides, the polystyrene plastic bottles immersed in acetic acid (4%, v/v) were adopted as control (Fig. S1). The detailed immersion procedures were described in Supporting Information.

2.7. Procedures for DLS detection of gold NPs aggregation

The DLS detection of gold NPs aggregation was performed according to the following procedures [18]. First, the as-prepared GNPs–DNA conjugates were mixed with three times volume of the solution to be assayed, and then the mixture was allowed to stand at room temperature for 15 min. Subsequently, about 1 mL of the balanced mixture was pipetted into to a dust-free DTS0012-Disposable sizing cuvette. Finally, the DLS measurements were carried out at 25 °C, with the equilibration time set for 2 min to maintain temperature balance, and a fixed 10 runs for each sample.

The DLS analyses of all the samples were performed in the aforementioned four simulants (containing 0.2 nM GNPs–DNA and 12 mM NaCl) with different standard operation procedures (SOPs), but the same SOP was adopted for the same solvent. The parameters of different SOPs were elaborated in Supporting Information. All measurements were carried out on at least three parallel sample sets, and each reported particle size was the average of three measurements.

3. Results and discussion

3.1. Optimization of GNPs solution

As known to us all, the concentrations of p-DNA and NaCl solutions and the pH of PBS solution have great influence on the stability of GNPs [28]. Thus, we investigated their influences on GNPs and obtained the optimal conditions.

Fig. 2 shows the optimization results of the concentration of NaCl (C_{NaCl}) solution, the pH of PBS, the concentration of p-DNA, and the aggregation time of GNPs–DNA conjugates combining with melamine. Based on the results, the pH value of PBS and the concentration of NaCl solution were chosen to be 6.5 (Fig. 2(A)) and 60 mM (Fig. 2(B)), respectively. Considering the adequate binding site of p-DNA with melamine and the stability of GNPs, the concentration of p-DNA ($C_{p\text{-}DNA}$) was chosen to be 87 μM (pH 6.5, C_{NaCl} = 60 mM) (Fig. 2(C)). Additionally, the aggregation time of GNPs–DNA conjugates combining with different concentrations of melamine (pH 6.5, C_{NaCl} = 60 mM, $C_{p\text{-}DNA}$ = 87 μM) was chosen to be 15 min (Fig. 2(D)), because the average hydrodynamic diameter of GNPs–DNA conjugates increased steadily as time was prolonged, and then became balanced after 15 min.

3.2. Characterization of GNPs

The stability of the GNPs–DNA conjugates usually affected the detection results of melamine. Compared with the electrostatic interaction, the Au—S chemical bonds provided the stronger conjugation between DNA and the GNPs [29]. The preparation process in this study was simple and efficient without any coupling

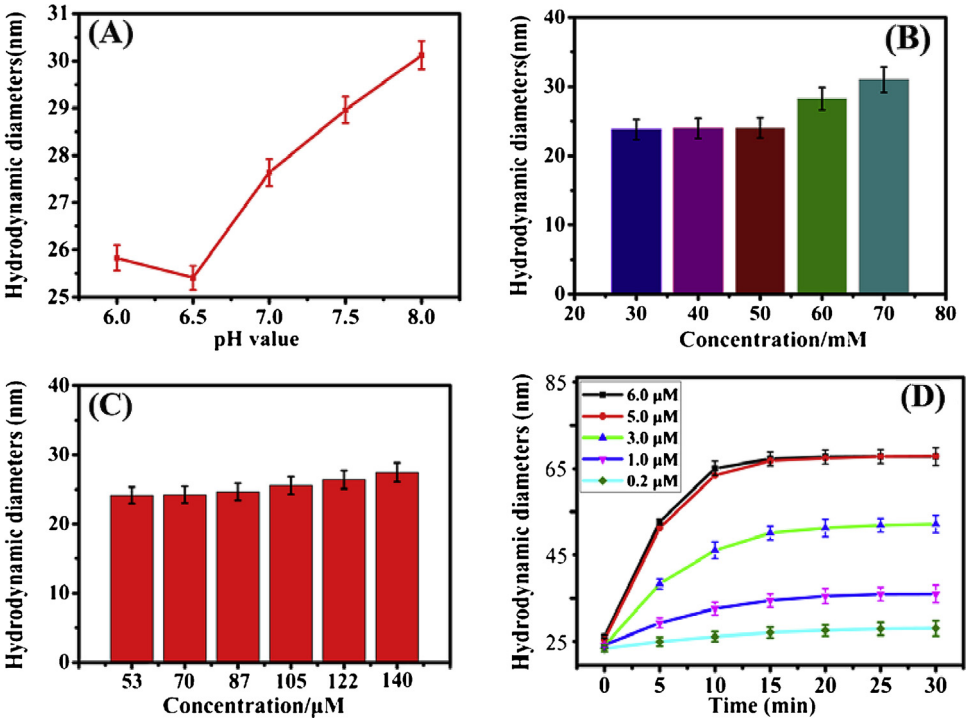


Fig. 2. Optimization results of GNPs. (A) The pH value of PBS, (B) the concentration of sodium chloride solution (pH 6.5), (C) the concentration of DNA (pH 6.5, C_{NaCl} = 60 mM), and (D) the time of GNPs–DNA conjugates (Cp-DNA = 87 μM) combining with different concentrations of melamine (from bottom to up: 0.2, 1.0, 3.0, 5.0, 6.0 μM; pH 6.5, C_{NaCl} = 60 mM). All the error bars were calculated based on the standard deviation of three measurements.

agent. The obtained GNPs were characterized by UV–vis absorption spectra, TEM, and DLS.

To demonstrate the aforementioned mechanism of specific hybridization between GNPs–DNA conjugates and melamine, different samples were prepared as control, and the color change of the solution is displayed in Fig. 3(A). Furthermore, the results of UV–vis absorption spectrum (Fig. 3(B)) were consistent with those in Fig. 3(A). In Fig. 3(B), a typical UV–vis absorption spectrum of GNPs was observed with a sharp peak located at 521 nm (curve a, curve b). After combining with the p-DNA, the intensity of the peak at 521 nm remained nearly intact (curve c). As expected, after the addition of formaldehyde (3.0 μM), the location of the peak still remained unchanged (curve d). However, after hybridization with melamine (3.0 μM), the peak slightly shifted from 521 nm to 525 nm, and its intensity decreased at 521 nm and increased at 700 nm (curve e). The results demonstrated that the GNPs aggregated after the hybridization of the GNPs–DNA conjugates and melamine.

TEM images of GNPs are shown in Fig. 4, and the TEM sample preparation is presented in Supporting Information. The synthesized gold NPs were uniform and well-dispersed with an average diameter of approximately 23 nm (Fig. 4(A)). Initially, the GNPs were well dispersed in PBS with p-DNA (Fig. 4(B)) or melamine (Fig. 4(C)). However, the GNPs–DNA conjugates, when recognized by melamine, were bound to it (Fig. 4(E)), forming a sharp contrast to the GNPs–DNA conjugates alienated from formaldehyde (Fig. 4(D)). The results were in agreement with the analysis of UV–vis absorption spectra.

DLS was used to monitor the size change of GNPs–DNA conjugates. The average hydrodynamic diameter of the as-prepared GNPs was 23.53 nm. When the p-DNA and melamine were separately mixed with GNPs, the average size turned out to be 23.81 nm and 23.95 nm, respectively, indicating no significant difference from the blank. When GNPs–DNA conjugates (0.8 nM) were mixed with formaldehyde (3.0 μM), the average size showed insignificant

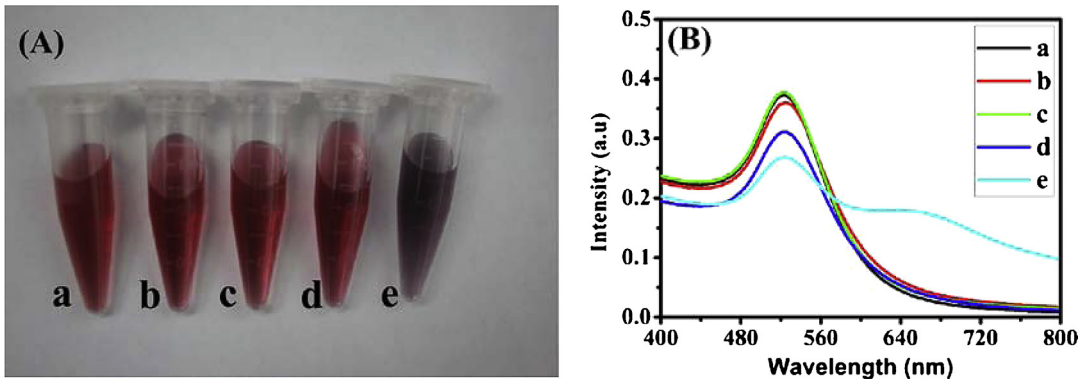


Fig. 3. (A) Visual color change of optimized GNPs: (a) GNPs without any addition; (b) GNPs in the presence of melamine (3.0 μM); (c) GNPs–DNA conjugates ($C_{p\text{-}DNA}$ = 87 μM); (d) mixture of GNPs–DNA conjugates ($C_{p\text{-}DNA}$ = 87 μM) and formaldehyde (3.0 μM); (e) mixture of GNPs–DNA conjugates ($C_{p\text{-}DNA}$ = 87 μM) and melamine (3.0 μM). (B) UV–vis absorption spectra of the GNPs corresponding to the triple dilutions in (A).

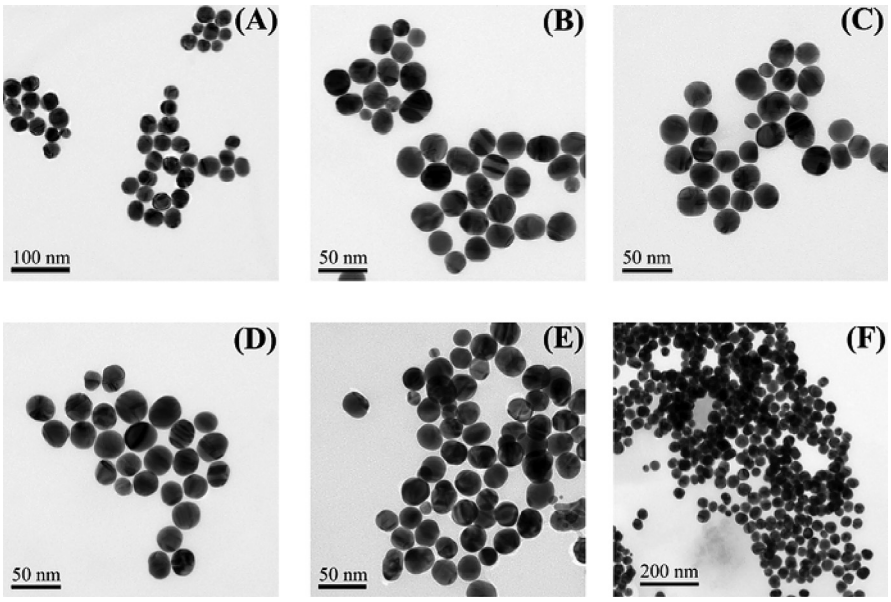


Fig. 4. HRTEM images of (A) GNPs, (B) GNPs with melamine (3.0 μM), (C) GNPs–DNA conjugates ($C_{p\text{-}DNA}$ = 87 μM), (D) GNPs–DNA conjugates ($C_{p\text{-}DNA}$ = 87 μM) with formaldehyde (3.0 μM), (E) GNPs–DNA conjugates ($C_{p\text{-}DNA}$ = 87 μM) with melamine (3.0 μM), and (F) a zoomed out version of (E).

change (24.66 nm). However, the size increased significantly from 23.81 nm to 61.59 nm after the hybridization of GNPs–DNA conjugates (0.8 nM) with melamine (3.0 μM), suggesting that the aggregation of GNPs–DNA conjugates was caused by the combination with melamine rather than formaldehyde.

3.3. Linearity of standard solutions

DLS measurements for the prepared standard solutions were carried out as described in Section 2.7. The GNPs–DNA conjugates were rather stable with nearly constant sizes (Fig. S1). With the specific recognition of GNPs–DNA and melamine, the average size of GNPs gradually increased and became balanced after 15 min (Fig. 2(D)). The balanced size was proportional to the concentration of melamine and put as linear equations in each solvent.

The quantities related to the use of melamine standard linear equations are listed in Table 1. All the equations present a good correlation coefficient of above 0.9970 within the concentration range of 5.0 μg L^{-1}–320.0 μg L^{-1}. Moreover, the method showed high precision (RSD ≤ 3%) and accuracy, with a relative error below 5%. From the slope depicted in Table 1, it can be concluded that the average diameter of GNPs in water, ethanol (10%, v/v) and isopropanol (10%, v/v) had a slow change with the increase of melamine concentration. However, in acetic acid solvent (4%, v/v), the appearance of a larger slope indicated that GNPs possessed a greater change rate with the increase of melamine concentration. Interestingly, it was found to have a good linearity with the correlation coefficient of 0.9971.

The above results were probably related to the solvent properties such as pH and molecular polarity. The pH value was found to have great influence on the stability of GNPs–DNA conjugates (discussed in Section 3.1), and molecular polarity possibly affected their dispersity. Thus, the greater change rate and wider rangeability of size in acetic acid solvent (4%, v/v) were probably due to the stronger pH (2.84) and molecular polarity of acetic acid.

3.4. Method comparison

Table 2 lists the comparison of the proposed method and other reported techniques for the detection of melamine. Clearly, the sensitivity (2.0 μg L^{-1}) of the proposed method was 50 times higher than that of the traditional HPLC method [13], and also significantly higher than that of HPLC/MS/MS [14], capillary electrophoresis diode with array detection [31], and colorimetric sensing method [32]. In addition, the proposed method was accurate and precise with the least limit of quantity (LOQ) (5.0 μg L^{-1}).

3.5. MMMQ in different solvents

Fig. S2 displays the results of DLS signals upon melamine in different solvents under the conditions of different temperatures and different time periods. With the increase of immersion temperature and time, a significant change can be observed in the average hydrodynamic diameter of GNPs, indicating that the melamine monomer migration rate (MMMR) accelerated with the increases of temperature. However, the average hydrodynamic diameter of GNPs nearly remained unchanged (Fig. S1) when the polystyrene plastic bottles were immersed in acetic acid (4%, v/v) at different temperatures.

Table 1
The equations to the standard melamine solution in different solvents.

Solvent	pH	Equation (m = 6)	R	$\pm C_b/\pm C_a$ ($1-\alpha$ = 0.95)
Water	6.55	(1): $d = 0.05796c + 23.87$	0.9987	±0.002914/±0.4394
4% (v/v) acetic acid	2.84	(2): $d = 0.3356c + 24.25$	0.9971	±0.01782/±2.6871
10% (v/v) ethanol	6.73	(3): $d = 0.04255c + 23.89$	0.9982	±0.002449/±0.3693
10% (v/v) isopropanol	6.53	(4): $d = 0.03967c + 23.92$	0.9970	±0.002954/±0.4455

c: melamine concentration (μg L^{-1}); d: Z-average diameter (nm); m: total number of points; $\pm C_b$: confidence limits about the slope; $\pm C_a$: confidence limits about the intercept; $1-\alpha$: confidence level.

Table 2
Comparison of the proposed method with the reported techniques.

Method	LOD ($\mu g\,mL^{-1}$)	LOQ ($\mu g\,mL^{-1}$)	Reference
HPLC	0.1	0.2	[13]
LC/MS/MS	–	0.05	[14]
RPHLC	–	0.1	[30]
Capillary electrophoresis	0.047	0.05	[31]
Colorimetric sensing	0.006	0.05	[32]
DLS	0.002	0.005	This work

LOD: limit of detection; LOQ: limit of quantity.

The results of MMMQ in different solvents are shown in Supporting Information (Tables S1–S4), and Fig. S3 displays the corresponding calibration plots of immersion time vs. logarithm of MMMQ (lg MMMQ). From the results shown in Fig. S3, it can be concluded that the MMMQ largely depends on the immersion temperature and time, and the MMMQ value is higher and harder to balance in a solvent with high polarity. Based on the results ($<0.2\,mg\,dm^{-2}$ at 70 °C for 2 h), all the samples measured have reached the National Criterion of the People's Republic of China, and in polystyrene plastic bottles, melamine is not detectable under the same conditions (see Fig. S1).

4. Conclusions

A simple and reliable method using DLS technique has been developed for the detection of MMMQ from melamine resin food containers without any separation and purification of the samples. The proposed method shows good linearity ($R \geq 0.9970$) between the size change of GNPs and the concentration of melamine, and more than 93 samples were quickly detected (less than 3 min for each sample) with satisfactory results. Besides, the influences of four typical solvents, temperature, and time on the MMMQ have been investigated. All the results suggest that the method is simple in operation and remarkable in sensitivity, and has potential in food testing and supervision. Moreover, this work has demonstrated the potential application of DLS assay as a simple, rapid, and sensitive method for detecting melamine extracted from the melamine tableware. However, the sustained MMMQ from the melamine resin containers, for example 3-day or longer immersion duration, and different brands or shapes of melamine plastic containers, remains to be explored.

Acknowledgment

We gratefully acknowledge the financial support from National Natural Science Foundation of China (21375043, 21175051).

Appendix A. Supplementary data

Supplementary data associated with this article can be found, in the online version, at http://dx.doi.org/10.1016/j.aca.2014.07.036.

References

[1] N.N. Yazgan, İ.H. Boyacı, A. Topcu, U. Tamer, Detection of melamine in milk by surface-enhanced Raman spectroscopy coupled with magnetic and Raman-labeled nanoparticles, Anal. Bioanal. Chem. 403 (2012) 2009–2017.
[2] L.C. Mecker, K.M. Tyner, J.F. Kauffman, S. Arzhantsev, D.J. Mans, C.M. Gryniewicz-Ruzicka, Selective melamine detection in multiple sample matrices with a portable Raman instrument using surface enhanced Raman spectroscopy-active gold nanoparticles, Anal. Chim. Acta 733 (2012) 48–55.
[3] M. Ibáñez, J.V. Sancho, F. Hernández, Determination of melamine in milk-based products and other food and beverage products by ion-pair liquid chromatography–tandem mass spectrometry, Anal. Chim. Acta 649 (2009) 91–97.
[4] C.C. Liu, C.F. Wu, B.H. Chen, S.P. Huang, G. William, H.H. Lee, Y.H. Chou, W.J. Wu, C.H. Huang, J. Shiea, C.H. Lee, K.Y. Wu, M.T. Wu, Low exposure to melamine increases the risk of urolithiasis in adults, Kidney Int. 80 (2011) 746–752.
[5] B.M. Kuehn, Melamine scandals highlight hazards of increasingly globalized food chain, JAMA 301 (2009) 473–475.
[6] N. Poovarodom, P. Tangmongkollert, T. Jinkarn, V. Chonhenchob, Survey of counterfeit melamine tableware available on the market in Thailand, and its migration, Food Addit. Contam. 28 (2011) 251–258.
[7] K.H. Lund, J.H. Petersen, Migration of formaldehyde and melamine monomers from kitchen and tableware made of melamine plastic, Food Addit. Contam. 23 (2006) 948–955.
[8] National Criterion of the People's Republic of China [S], Hygienic standard for melamine-formaldehyde products used as food containers and packaging materials, GB/T 9690–2009.
[9] E.L. Bradley, V. Boughtflower, T.L. Smith, D.R. Speck, L. Castle, Survey of the migration of melamine and formaldehyde from melamine food contacts articles available on the UK market, Food Addit. Contam. 22 (2005) 597–606.
[10] X.D. Pan, P. Wu, D.J. Yang, L.Y. Wang, X.H. Shen, C.Y. Zhu, Simultaneous determination of melamine and cyanuric acid in dairy products by mixed-mode solid phase extraction and GC–MS, Food Control 30 (2013) 545–548.
[11] Y.T. Wu, C.M. Huang, C.C. Lin, W.A. Ho, L.C. Lin, T.F. Chiu, D.C. Tarng, C.H. Lin, T.H. Tsai, Oral bioavailability, urinary excretion and organ distribution of melamine in Sprague-Dawley rats by high-performance liquid chromatography with tandem mass spectrometry, J. Agric. Food Chem. 58 (2010) 108–111.
[12] X. Xu, Y. Ren, Y. Zhu, Z. Cai, J. Han, B. Huang, Y. Zhu, Direct determination of melamine in dairy products by gas chromatography/mass spectrometry with coupled column separation, Anal. Chim. Acta 650 (2009) 39–43.
[13] G. Venkatasami, J.R. Sowa Jr., A rapid, acetonitrile-free HPLC method for determination of melamine in infant formula, Anal. Chim. Acta 665 (2010) 227–230.
[14] M.S. Filigenzi, B. Puschner, L.S. Aston, R.H. Poppenga, Diagnostic determination of melamine and related compounds in kidney tissue by liquid chromatography/tandem mass spectrometry, J. Agric. Food Chem. 56 (2008) 7593–7599.
[15] Z. Wang, X. Ma, L. Zhang, W. Yang, L. Gong, P. He, Z. Li, Screening and determination of melamine residues in tissue and body fluid samples, Anal. Chim. Acta 662 (2010) 69–75.
[16] Q. Dai, X. Liu, J. Coutts, L. Austin, Q. Huo, A one-step highly sensitive method for DNA detection using dynamic light scattering, J. Am. Chem. Soc. 130 (2008) 8138–8139.
[17] X. Miao, L. Ling, X. Shua, Ultrasensitive detection of lead (II) with DNAzyme and gold nanoparticles probes by using a dynamic light scattering technique, Chem. Commun. 47 (2011) 4192–4194.
[18] D. Gao, Z. Sheng, H. Han, An ultrasensitive method for the detection of gene fragment from transgenics using label-free gold nanoparticle probe and dynamic light scattering, Anal. Chim. Acta 696 (2011) 1–5.
[19] Y. Yuan, S. Wei, G. Liu, S. Xie, Y. Chai, R. Yuan, Ultrasensitive electrochemiluminescent aptasensor for ochratoxin A detection with the loop-mediated isothermal amplification, Anal. Chim. Acta 811 (2014) 70–75.
[20] X. Li, J. Li, H. Kuang, L. Feng, S. Yi, X. Xia, H. Huang, Y. Chen, C. Tang, Y. Zeng, An ultrasensitive method for the determination of melamine using cadmium telluride quantum dots as fluorescence probes, Anal. Chim. Acta 802 (2013) 82–88.
[21] J. Li, Z. Chen, Y. Li, A strategy for constructing sensitive and renewable molecularly imprinted electrochemical sensors for melamine detection, Anal. Chim. Acta 706 (2011) 255–260.
[22] D.C. Sherrington, K.A. Taskinen, Self-assembly in synthetic macromolecular systems via multiple hydrogen bonding interactions, Chem. Soc. Rev. 30 (2001) 83–93.
[23] K. Ai, Y. Liu, L. Lu, Hydrogen-bonding recognition-induced color change of gold nanoparticles for visual detection of melamine in raw milk and infant formula, J. Am. Chem. Soc. 131 (2009) 9496–9497.
[24] Y. Zeng, Y. Pratumyot, X. Piao, D. Bong, Discrete assembly of synthetic peptide-DNA triplex structures from polyvalent melamine–thymine bifacial recognition, J. Am. Chem. Soc. 134 (2012) 832–835.
[25] K. Chen, H. Han, Z. Luo, Y. Wang, X. Wang, A practicable detection system for genetically modified rice by SERS-barcoded nanosensors, Biosens. Bioelectron. 34 (2012) 118–124.
[26] W. Haiss, N.T.K. Thanh, J. Aveyard, D.G. Fernig, Determination of size and concentration of gold nanoparticles from UV–vis spectra, Anal. Chem. 79 (2007) 4215–4221.
[27] Food contact materials-Polymer-Determination of 2,4,6-triamino-1,3,5-triazine in food simulants-High performance liquid chromatography, GB/T 23,296-15-2009.
[28] L.M. Demers, C.A. Mirkin, R.C. Mucic, R.A. Reynolds, R.L. Letsinger, R. Elghanian, G. Viswanadham, A fluorescence-based method for determining the surface coverage and hybridization efficiency of thiol-capped oligonucleotides bound to gold thin films and nanoparticles, Anal. Chem. 72 (2000) 5535–5541.
[29] L. Vigderman, P. Manna, E.R. Zubarev, Quantitative replacement of cetyl trimethylammonium bromide by cationic thiol ligands on the surface of gold nanorods and their extremely large uptake by cancer cells, Angew. Chem. Int. Ed. 50 (2011) 1–7.
[30] H. Sun, L. Wang, L. Ai, S. Liang, H. Wu, A sensitive and validated method for determination of melamine residue in liquid milk by reversed phase high-performance liquid chromatography with solid-phase extraction, Food Control 21 (2010) 686–691.
[31] Z. Chen, X. Yan, Simultaneous determination of melamine and 5-hydroxymethyl -furfural in milk by capillary electrophoresis with diode array detection, J. Agric. Food Chem. 57 (2009) 8742–8747.
[32] H. Guan, J. Yu, D. Chi, Label-free colorimetric sensing of melamine based on chitosan-stabilized gold nanoparticles probes, Food Control 32 (2013) 35–41.