Colorimetric Method for the Detection of Mercury Ions Based on Gold Nanoparticles and Mercaptophenyl Boronic Acid

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In the present study, we found that phenylboronic acid derivatives including benzene-1,4-diboronic acid (BDBA) and mercaptophenyl boronic acid (MPBA) can induce the aggregation of citrate-capped gold nanoparticles (Au NPs). However, the speed of Au NP aggregation induced by MPBA was much faster than that of BDBA. The reaction between MPBA and Hg2+ ions resulted in the formation of MPBA-Hg2+-MPBA, which was similar to BDBA having two free boronic acid groups. Based on the above phenomenon, a sensitive and selective colorimetric method for the detection of mercury ions (Hg2+) in aqueous solution was developed. The linear range for the detection of Hg2+ ions was from 0.08 to 1.25 μmol dm−3 with a detection limit of 37 nmol dm−3. The strategy offered excellent selectivity toward Hg2+ against other metal ions. Meanwhile, this simple and cost-effective sensor was applied to determine the Hg2+ in the lake water samples with satisfactory recoveries (91.3 – 100.7%).

Keywords Colorimetric detection, Hg2+, gold nanoparticles, phenylboronic acid derivatives

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Introduction

Mercury ion (Hg2+) is one of the highly toxic heavy metal pollutants that exerts an adverse effect on the environment and also on human health.1,2 Mercury exists in a variety of different forms such as inorganic mercury, mercury vapors, and organic mercury derivatives.3 Inorganic mercury can cause serious damage to the kidneys, brain, endocrine system and central nervous system of both humans and animals.4 Also, water-soluble Hg2+ ions can be transformed to methyl mercury, which is the most common organic source of mercury through the microbial biomethylation process.5 Hence, routine detection of Hg2+ ions in the aqueous ecosystem is significant for environmental monitoring of rivers and larger bodies of water. Concerns over toxic exposure to mercury have motivated the exploration of new methods for the detection of water-soluble Hg2+ ions. Although traditional methods such as atomic absorption/emission spectrometry and inductively coupled plasma-mass spectrometry (ICP-MS) provide high sensitivity and selectivity for the detection of Hg2+ ions, these methods suffer from the need for expensive, sophisticated instrumentation and time-consuming sample preparation processes.6,7 Alternatively, many electrochemical and optical sensing systems capable of providing real-time Hg2+ determination with reduced analysis time and cost have been developed, based on organic fluorophores,8 oligonucleotides,9–13 proteins,14,15 polymers,16 metal nanoparticles,17–20 semiconductor quantum dots,21–23 graphene oxide,24 single-walled carbon nanotubes25 and so on. Among these methods, colorimetric methods are extremely attractive because the results can be easily read out with the naked eye in some cases at the point of use. Several colorimetric methods have been developed for the simple and rapid detection of Hg2+ ions.6–25 For example, Mirkin et al.26 a colorimetric method to detect Hg2+ ions using DNA-functionalized Au NPs, in which the concentration of Hg2+ ions can be determined by the change of the solution color at a given temperature or the melting temperature of the DNA-Au NP aggregates. Chang et al.27 reported another colorimetric assay system for Hg2+ ions based on poly T, ss-DNA and 13-nm Au NPs. The above methods were sensitive and selective for the detection of Hg2+ ions. However, they still needed strictly controlled experimental conditions such as accurate temperature control and salt concentration, and use of specific DNA sequences, which limited the on-site application of the proposed methods.

Recently, Au NPs have been widely employed in developing biosensing systems for the detection of proteins,28 sequence-specific DNA29 and small molecules,30 because of their unique properties such as facile synthesis, large specific surface area, high chemical stability and biocompatibility. The major advantages of using Au NPs for the detection of Hg2+ ion include the elimination of the labeling process and the high sensitivity afforded arising from the extremely high extinction coefficients of their surface plasmon resonance (SPR) absorption effects. In the present study, we found that mercaptophenyl boronic acid (MPBA) is capable of inducing the aggregation of citrate-capped Au NPs. We speculated that the driving force for the MPBA-induced nanoparticle aggregation might arise from the interaction between the citrate and boronic acids of MPBA and formation of Au-S bond between Au surface and -SH group of MPBA.31,32 Interestingly, we found that under the same experimental condition, benzene-1,4-diboronic acid (BDBA)
exhibited a much lower ability to induce the aggregation of citrate-capped Au NPs. Based on the above phenomenon, a simple colorimetric method for the detection of Hg²⁺ ions was developed (Scheme 1). In the absence of Hg²⁺ ions, MPBA triggered the aggregation of citrate-capped Au NPs resulting in a red-to-blue color change of the colloidal solution. In contrast, in the presence of Hg²⁺ ions in solution, Hg²⁺ ions competitively react with -SH group of MPBA forming MPBA-Hg²⁺-MPBA, whose structure is similar to BDBA having two free boric acid groups. The resulting product was incapable of changing the color of the colloidal solution and the extinction spectrum. The proposed method is capable of quantitatively detecting aqueous Hg²⁺ ions with both high sensitivity and selectivity.

Experimental

Reagents and chemicals

All chemicals were of analytical reagent grade and were used as received. Ultrapure water (18.2 MΩ cm⁻¹) was used throughout the current work. MPBA and BDBA were obtained from Sigma-Aldrich (Beijing, China). Tetrachloroaurate(III) tetrahydrate (HAuCl₄·4H₂O) and trisodium citrate dihydrate (C₆H₅O₇Na₃·2H₂O) were obtained from Sinopharm Chemical Reagent Co., Ltd. (Beijing, China). Hg(NO₃)₂ was purchased from Aladdin Biochemical Technologies Inc (Shanghai, China). Other chemicals such as BaCl₂·2H₂O, CaCl₂·2H₂O, CdCl₂·H₂O, CoCl₂·6H₂O, CuCl₂·3H₂O, CrCl₃·6H₂O, FeCl₃·6H₂O, FeCl₂·4H₂O, MgCl₂·6H₂O, ZnCl₂, NaCl, KCl, NiCl₂ and MnCl₂·4H₂O were purchased from Tianjin Chemical Co., Ltd (Tianjin, China). Stock solutions of MPBA and BDBA were prepared daily at a concentration of 20 mM in ethanol and the working solution was made by gradually diluting the stock solution with K₂HPO₄–KH₂PO₄ buffer solution (pH 8.0).

Apparatus

UV-Vis measurements were performed with a multiscan spectrometer (Tecan Infinite M200, Switzerland).

Preparation of Au NPs

Au NPs were prepared by citrate-mediated reduction of HAuCl₄. Briefly, 1 mL of 1% (w/v) hydrogen tetrachloroaurate solution was added to 99 mL of water. The solution was brought to boil and stirred vigorously. Then, 4 mL of 1% (w/v) sodium citrate was added to the boiling solution quickly. The solution was continuously stirred until the solution color turned to wine red. After another 5 min of stirring, the heat source was removed and the solution was cooled down to room temperature. The average size of gold nanoparticles identified by a Zetasizer nanoparticle analyzer (Malvern Instruments, UK) was about 16 ± 1 nm. In order to remove free sodium citrate in the solution, the Au NPs were isolated by centrifugation (12000 rpm for 20 min) and resuspended in ultrapure water. The centrifugation-resuspension-recentrifugation cycle was repeated 2 times. The red oily precipitate was resuspended in ultrapure water, and was one-tenth of its original volume. The concentration of the Au NPs was calculated to be 7.36 nmol dm⁻³ according to the Lambert-Beer law.

Assay procedure for colorimetric detection of Hg²⁺

For the colorimetric detection of Hg²⁺, 50 μL of a reaction solution containing MPBA and a desired concentration of Hg²⁺ was transferred to the well of a 96-well plate and incubated at room temperature for 20 min. Then, 50 μL of Au NPs was added to the reaction solution. And the resulting reaction mixture was incubated at room temperature for 20 min. Finally, the UV-Vis absorption at 520 and 690 nm was recorded by a multiscan spectrometer.

Results and Discussion

Aggregation of citrate-capped Au NPs induced by MPBA and BDBA

Au NPs that were prepared by citrate-mediated reduction of HAuCl₄ are stable in aqueous solution, because of the electrostatic repulsion of the negative capping agent (e.g. citrate ion) against the van der Waals attraction between Au NPs. The prepared Au NP solution had a wine red color and displayed SPR peak located at 520 nm. Interestingly, we found that MPBA was capable of triggering the aggregation of citrate-capped Au NPs. The addition of MPBA into the Au NPs caused the color of the colloidal solution to change from red to blue. The presence of MPBA also caused a decrease in the strength of the SPR peak at 520 nm and a new broad SPR peak located at 690 nm, which were characteristics of gold nanoparticle aggregation (Fig. 1B). The extinction coefficients of the solutions at 520 and 690 nm are related to the quantities of dispersed and aggregated Au NPs, respectively. Thus, the $A_{520}/A_{690}$ value was used in the following study to express the
molar ratio of dispersed and aggregated Au NPs. Then, the effect of MPBA concentration on the aggregation of Au NPs was investigated (Fig. S1A, Supporting Information) by using a fixed concentration of Au NPs (1 mM). As the concentration of MPBA was raised from 0 to 5 μM, the absorbance (A$_{520}$/A$_{690}$) value of the MPBA-Au NPs reaction system progressively decreased, indicating a higher concentration of MPBA induced higher aggregation level of Au NPs. And then the A$_{520}$/A$_{690}$ value leveled off when the concentration of MPBA was above 1.25 μM. The results demonstrated that 1.25 μM MPBA can induce the aggregation of 1 nM Au NPs efficiently. Tseng et al. reported the boronic acids of BDBA could react with the α-hydroxycarboxylate of citrate, triggering the aggregation of citrate-capped Au NPs. Based on the report, we speculated that the driving force for the MPBA-induced nanoparticle aggregation could arise from the binding of citrate to boronic acids of MPBA and formation of Au-S bond between Au surface and -SH group of MPBA.

In the next experiment, we investigated the influence of the concentration of BDBA on the aggregation of Au NPs. As shown in Fig. S1B (Supporting Information), much higher concentration of BDBA was needed to trigger the aggregation of Au NPs. In other words, compared with MPBA, BDBA, which has two boronic groups, showed lower ability to induce the aggregation of citrate-capped Au NPs. We speculated that the stronger aggregation effect of MPBA on citrate-capped Au NPs may be attributed to the -SH group of MPBA, which has a higher binding ability to the surface of Au NPs.

**Mechanism of the label-free colorimetric detection of Hg$^{2+}$ ions**

Based on the above observation, a label-free colorimetric method for the detection of Hg$^{2+}$ ions was developed in the present study. In the absence of Hg$^{2+}$ ions, MPBA was capable of inducing the aggregation of Au NPs. In contrast, in the presence of Hg$^{2+}$ ions, Hg$^{2+}$ ions were capable of reacting with -SH group of MPBA and resulting in the formation of MPBA-Hg$^{2+}$-MPBA whose structure was similar to BDBA having two boronic acid groups and the incapability of triggering the aggregation of Au NPs under the same experimental conditions. We verified the feasibility of the designed label-free colorimetric platform for the detection of Hg$^{2+}$ ions. Figures 1A and 1B shows the photographs and UV-Vis spectra of Au NPs under different conditions. In the presence of 2.5 μM BDBA, the color change of the 2 mM Au NP solution from red to blue implied the aggregation of Au NP, while in the presence of the same concentration of BDBA, the color of the Au NP solution remained red. When 1.25 μM MPBA was incubated with 2.5 μM MPBA, the resulting product (MPBA-Hg$^{2+}$-MPBA) was incapable of changing the color of the colloidal solution and the extinction spectrum. Then, we calculated the corresponding A$_{520}$/A$_{690}$ value under different conditions (Fig. 1C). A low A$_{520}$/A$_{690}$ value was obtained when MPBA was incubated with Au NPs for 20 min, indicating the aggregation of Au NPs in the presence of 2.5 μM MPBA. Under the same experimental conditions, the A$_{520}$/A$_{690}$ value of Au NP solution in the presence of 2.5 μM MPBA was almost the same as the control colloidal solution in which the buffer solution instead of the phenylboronic acid derivatives was added. The A$_{520}$/A$_{690}$ value (refers to the A$_{520}$/A$_{690}$ value of Au NPs in the blank group minus A$_{520}$/A$_{690}$ value of Au NPs in the presence of phenylboronic acid derivatives) was employed to reflect the dispersion status difference between Au NPs in the blank group and Au NPs in the presence of phenylboronic acid derivatives. Figure S2 (Supporting Information) shows the time dependence of A$_{520}$/A$_{690}$ value of Au NPs under different conditions. When 2.5 μM MPBA was incubated with 2 mM Au NPs, the A$_{520}$/A$_{690}$ value increased with the increase of incubation time within 10 min indicating MPBA can trigger the aggregation of Au NPs quickly and then levelled off when the Au NPs response time ranged from 10 to 20 min. Then, the A$_{520}$/A$_{690}$ value decreased gradually because of the increase of blank signal, which is attributed to the instability of Au NPs in buffer solution when the incubation time is longer than 20 min. In addition, the TEM images in Figure S3 (Supporting Information) also revealed that Au NPs formed aggregates in the presence of MPBA. In contrast, the Au NPs are monodispersed in the absence of MPBA and in the presence of MPBA-Hg$^{2+}$-MPBA and BDBA. The above results demonstrated that Hg$^{2+}$ ions could react with the -SH group of MPBA forming MPBA-Hg$^{2+}$-MPBA, which could not stimulate the aggregation of Au NPs under the same experimental conditions. Based on the above observations, a simple colorimetric sensing system was established for the detection of Hg$^{2+}$ ions in aqueous solution.

**Optimization of experimental conditions for colorimetric detection of Hg$^{2+}$ ions**

The assay conditions such as the concentration of MPBA and Au NPs, the pH of the reaction solution, and the response time
of Au NPs were investigated systematically for the sensitive and selective detection of Hg$^{2+}$. Firstly, the concentration of MPBA incubated with Hg$^{2+}$ ions plays an important role for the sensitive detection of Hg$^{2+}$ ions. The effect of MPBA concentration on $\Delta A_{520}/A_{690}$ (refers to the $A_{520}/A_{690}$ value of sample minus the $A_{520}/A_{690}$ value of blank) was studied in the range of 0 – 5 $\mu$mol dm$^{-3}$ by using a fixed concentration of Au NPs (1 nmol dm$^{-3}$). As shown in Fig. 2a, the $\Delta A_{520}/A_{690}$ increased with increasing MPBA concentration in the range of 0 – 1.25 $\mu$mol dm$^{-3}$, and the signal decreased dramatically when the concentration of MPBA was above 1.25 $\mu$mol dm$^{-3}$. The results demonstrated that although a high concentration of MPBA was beneficial to the aggregation of Au NPs, an excess amount of MPBA may decrease the detection sensitivity for Hg$^{2+}$ ions. Hence, 1.25 $\mu$mol dm$^{-3}$ MPBA, which was corresponding to 1 nmol dm$^{-3}$ Au NPs, was selected to incubate with Hg$^{2+}$ ions for the following studies.

The Au NP response time is significant for the sensitive detection of Hg$^{2+}$ ions. In the absence of Hg$^{2+}$ ions, MPBA could induce the aggregation of Au NPs rapidly. However, in the presence of MPBA-Hg-MPBA, which is the reaction product of MPBA and Hg$^{2+}$ ions, the aggregation speed of Au NPs would slow down. As shown in Fig. 2b, the maximum $\Delta A_{520}/A_{690}$ value was obtained at 20 min. Hence, 20 min was selected as the response time of Au NPs for the following experiments.

The effect of solution pH on the colorimetric detection of Hg$^{2+}$ ions was studied in the range of pH 6 to 10. KH$_2$PO$_4$–K$_2$HPO$_4$ buffer and K$_2$HPO$_4$–NaOH buffer were employed to control the pH of the reaction solution. We found that the blank signal increased with the decrease of solution pH, indicating acidic condition was inappropriate for MPBA to trigger the aggregation of Au NPs. The reason may be because the acidic condition may be not conductive to the dissociation of –SH group of MPBA and to the esterification reaction of boronic acid of MPBA and citrate on the surface of Au NPs. As shown in Fig. 2c, the $\Delta A_{520}/A_{690}$ increased rapidly when the pH of the reaction solution was increased from 6 to 8, and then decreased gradually when the pH value was above 8. Hence, the optimal pH of the reaction solution was set at 8.

Finally, the influence of the concentration of Au NPs on the $\Delta A_{520}/A_{690}$ was examined in the range of 0.25 – 4 nmol dm$^{-3}$ by using a fixed MPBA to Au NPs ratio (1.25 $\mu$M MPBA corresponding to 1 nmol dm$^{-3}$ Au NPs). It was found that the $\Delta A_{520}/A_{690}$ increased with the increasing concentration of Au NPs from 0.25 to 2 nmol dm$^{-3}$, and then remained in the range of 2 – 4 nmol dm$^{-3}$ (Fig. 2d). Hence, 2 nmol dm$^{-3}$ Au NPs was selected for the following experiments.

Sensitivity, repeatability, and selectivity for the detection of Hg$^{2+}$ ions

Once the optimum experimental conditions were determined, the quantitative ability of the colorimetric sensor for the detection of Hg$^{2+}$ ions was evaluated by adding a different concentration of Hg$^{2+}$ ions under stringently controlled. In a typical experiment, the reaction solution containing 2.5 $\mu$mol dm$^{-3}$ MPBA and different concentrations of Hg$^{2+}$ ions was incubated at room temperature for 20 min, and the resulting products were incubated with citrate-capped Au NPs for 20 min.
As shown in Fig. 3a, with the increase of Hg$^{2+}$ concentration, an increase in the ratio of $A_{520}/A_{690}$ can be observed. It indicated that the aggregation of Au NPs decreased with the increase of Hg$^{2+}$ concentration, and a progressive color change from blue to red was observed (inset of Fig. 3a). A good linear relationship between the ratio of $A_{520}/A_{690}$ and the concentration of Hg$^{2+}$ ions was obtained over the range of 0.08 – 1.25 μmol dm$^{-3}$ with a correlation coefficient of 0.9969. When the concentration of Hg$^{2+}$ ions was above 1.25 μM, the signal increased slowly with the increase of Hg$^{2+}$ concentration, indicating the binding ability of MPBA may reach saturation. The limit of detection (LOD) was defined as the concentration of Hg$^{2+}$ ions, which gave a colorimetric signal 3$\times$ the standard deviation of a blank (which was detected eight times parallelly). The LOD of the proposed sensing system was calculated to be 37 nmol dm$^{-3}$, which is comparable with previously reported optical Hg$^{2+}$ assays (Table 1). Under the optimum conditions, the color of the citrate-capped Au NPs changed from red to blue in the presence of 1.25 μmol dm$^{-3}$ of 14 different interferential metal ions, while the colloidal color remained red in the presence of an equal concentration of Hg$^{2+}$ (the inset of Fig. 4). In all cases, the Hg$^{2+}$ ions showed 10 – 130 fold higher colorimetric signal ($\Delta A_{690}/A_{520}$) than that of any of the other metal ions (Fig. 4). The high selectivity of the sensing system may be attributed to the selective reaction between –SH group of MPBA and Hg$^{2+}$ ions. These results demonstrated that our strategy shows outstanding selectivity toward Hg$^{2+}$ sensing.

### Practical application

Lake water samples obtained from Jingye Lake on the campus of Tianjin University were employed to evaluate the potential of the proposed colorimetric sensing system for the analysis of Hg$^{2+}$ ions in environmental samples. The lake water samples were first filtered through a 0.22-μm membrane to remove soil and other particles. A standard addition method was employed to determine the concentration of Hg$^{2+}$ ions spiked in the lake water samples, and then tested using the proposed colorimetric sensing system. The results are summarized in Table 2, and the recoveries ranged from 91.3 to 100.7%. The results clearly confirmed that this colorimetric nanosensor was potentially applicable for the determination of Hg$^{2+}$ in real water samples.

### Table 1 Comparison between the colorimetric method and other optical techniques for Hg$^{2+}$ detection

| Technique | Linear range/μmol dm$^{-3}$ | LOD/nmol dm$^{-3}$ | Reference |
|-----------|-----------------------------|---------------------|-----------|
| Colorimetry | 0.08 – 1.25 | 37 | This work |
| UV-Vis | 0.5 – 50 | 85 | 4 |
| Colorimetry | 9 – 50 | 1350 | 19 |
| Colorimetry | 2 – 24 | 89 | 26 |
| Colorimetry | 0.2 – 60 | 30 | 34 |
| Fluorescent | 0 – 70 | 234 | 36 |
| Fluorescent | 0 – 25 | 230 | 37 |
| Colorimetric | 0 – 1 | 10 | 38 |

Fe$^{2+}$, Cu$^{2+}$, Ba$^{2+}$, Zn$^{2+}$, K$^+$, Cr$^{3+}$ and Na$^+$ were prepared. The colorimetric signals of the interferential ions were determined under the optimum experimental conditions of the case of Hg$^{2+}$ ions. The results indicated that the proposed assay exhibited very high selectivity for Hg$^{2+}$ ions (Fig. 4). Under the optimum conditions, the color of the citrate-capped Au NPs changed from red to blue in the presence of 1.25 μmol dm$^{-3}$ of 14 different interferential metal ions, while the colloidal color remained red in the presence of an equal concentration of Hg$^{2+}$ (the inset of Fig. 4). In all cases, the Hg$^{2+}$ ions showed 10 – 130 fold higher colorimetric signal ($\Delta A_{690/A_{520}}$) than that of any of the other metal ions (Fig. 4). The high selectivity of the sensing system may be attributed to the selective reaction between –SH group of MPBA and Hg$^{2+}$ ions. These results demonstrated that our strategy shows outstanding selectivity toward Hg$^{2+}$ sensing.
Table 2  Analytical results for Hg\textsuperscript{2+} spiked in lake water

| Sample | Spiked/\textmu mol dm\textsuperscript{-3} | Found/\textmu mol dm\textsuperscript{-3} | Recovery |
|--------|---------------------------------------|---------------------------------------|----------|
| 1      | 0.6250                                | 0.5708 ± 0.0179                       | 0.9133 ± 0.0286 |
| 2      | 0.3125                                | 0.3145 ± 0.0040                       | 1.0065 ± 0.0129 |
| 3      | 0.1563                                | 0.1438 ± 0.0186                       | 0.9202 ± 0.1156 |

Conclusions

In conclusion, we described a low-cost, rapid, highly selective, and sensitive colorimetric assay for detecting Hg\textsuperscript{2+} using unmodified Au NPs. Experiment results showed that MPBA was capable of triggering the aggregation of citrate-capped Au NPs. However, the Hg\textsuperscript{2+} ions-treated MPBA cannot stimulate the aggregation of Au NPs. Therefore, we can detect the amount of Hg\textsuperscript{2+} by the aggregation degree of Au NPs. The assay is particularly attractive because it does not rely on organic solvents, enzymatic reactions, light-sensitive dye molecules, lengthy protocols and sophisticated instrumentation. Most importantly, the method exhibited promising results for the detection of Hg\textsuperscript{2+} ions in lake water. It would open new opportunities for development of miniaturized detection technologies for Hg\textsuperscript{2+} in environmental applications.

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Supporting Information

The effect of MPBA and BDBA concentration on the aggregation of Au NPs (Fig. S1). The time dependence of \Delta A_{520/460} of Au NPs under different conditions (Fig. S2), and TEM images of the Au NP under different experimental conditions (Fig. S3).

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