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# **ORIGINAL ARTICLE**

# A highly sensitive and selective resonance Rayleigh ()scattering method for $Hg^{2+}$ based on the nanocatalytic amplification



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# **KEYWORDS**

Hg(II); Nanocatalysis; Resonance Rayleigh scattering **Abstract** In 0.2 mol/L HCl–0.22 mol/L HNO<sub>3</sub> medium, trace  $Hg^{2+}$  catalyzed NaH<sub>2</sub>PO<sub>2</sub> reduction of HAuCl<sub>4</sub> to form gold nanoparticles (AuNPs), which exhibited a strong resonance Rayleigh scattering (RRS) effect at 370 nm. With increasing of  $[Hg^{2+}]$ , the RRS effect enhanced due to more AuNP generated from the catalytic reaction. Under the chosen conditions, the enhanced RRS intensity at 370 nm is linear to  $Hg^{2+}$  concentration in the range of  $5.0-450 \times 10^{-9}$  mol/L, with a detection limit of 0.1 nmol/L. This RRS method was applied for the determination of Hg in water samples, with high sensitivity and good selectivity, and its results were agreement with that of atomic fluorescence spectrometry.

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### 1. Introduction

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As the economy development, the harm caused by toxic heavy metal pollution on the environment and human health has aroused widespread concern. Mercury has been widely recognized as one of the most hazardous pollutants and highly dangerous elements, it can cause great, long-term toxic effects on human and environment even at very low concentrations (Clarkson, 2002). Inorganic mercury can be combined with the protein sulfhydryl that would inhibit the activity of the enzyme and disruption of cell metabolism. The organic mercury such as methylmercury with strongest toxicity which can encroach the body's central nervous system and cause speech and memory disorders (Hassan et al., 2012). Thus,

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simple, rapid, sensitive, and selective detection of mercury is of great significance for biochemistry, environmental science, and medicine. At present, several methods have been reported to assay Hg<sup>2+</sup>, such as atomic spectrometry (Angeli et al., 2011; Resano et al., 2009), surface-enhanced Raman scattering spectroscopy (Ren et al., 2012; Wang and Chen, 2009), colorimetry (Fan et al., 2010; Jiang et al., 2012a,b; Lee et al., 2007), and resonance Rayleigh scattering spectroscopy (RRS) (Jiang et al., 2008; Kim et al., 2012; Rodrigues et al., 2010; Wu et al., 2012; Yin et al., 2012). Atomic spectrometry, especially atomic absorption spectrometry (AAS), is commonly used to determine trace Hg. However, the instrument is expensive and analytical time is long. The colorimetry is simple and economic, but with poor sensitivity. Surface-enhanced Raman scattering spectroscopy is difficult to quantitative analysis of Hg, with disadvantage of expensive instrument and bad reproducibility (Li et al., 2012). The RRS method has been applied in inorganic and organic analysis (Chen et al., 2012; Jiang et al., 2012a,b; Li et al., 2011; Ling et al., 2009; Liu et al., 2009; Song et al., 2012; Zou et al., 2012). Recently, a RRS method has been established to determine mercury ion, based on nanogold-labeled aptamer reaction (Jiang et al., 2008). However, the labeling step is complicated and long, and precious nucleic acid reagents were used. Up to date, there is no nanocatalytic RRS method for trace Hg. In this article, a simple, low-cost, sensitive and selective RRS spectral method has been established to determine mercury ion, which is based on the trace Hg<sup>2+</sup> catalyzing NaH<sub>2</sub>PO<sub>2</sub> reduction of HAuCl<sub>4</sub> to form AuNPs that exhibited a strong RRS peak at 370 nm.

#### 2. Materials and methods

#### 2.1. Apparatus and reagents

A model of F-7000 fluorescence spectrophotometer (Hitachi Company, Japan) was used to record the RRS intensity, with the excited and emission slit of 5.0 nm, emission filter = 1% T attenuator, and photoelectron multiple tube (PMT) voltage of 400 V, and a model of JLM-6380LV scan electron microscope (Electronic Co., Ltd, Japan) were used.

A 10 mmol/L  $Hg^{2+}$  stock standard solution was prepared as follows: a 0.2715 g HgCl<sub>2</sub> (National Pharmaceutical Group Chemical Reagents Company, China) was dissolved in 100 mL water. A 10 µmol/L  $Hg^{2+}$  working solution was obtained freshly by diluting with water. A 1.1 mol/L HNO<sub>3</sub> solution, 1 mol/L HCl solution, 4 mmol/L HAuCl<sub>4</sub>, 5 mmol/ L KMnO<sub>4</sub>, 10 mg/mL polyethylene glycol (PEG) 10000, 0.14 mol/L NH<sub>2</sub>OH•HCl, 1% trisodium citrate and 3.75 mol/L NaH<sub>2</sub>PO<sub>2</sub> solution were prepared. All regents were of analytical grade and the water was doubly distilled. The room temperature was 25 °C.

Preparation of AuNPs: In a 250 mL round-bottom flask equipped with 50 mL water and put on magnetic stirrer, adding 2.0 mL of 1% trisodium citrate after the water boiling. After the water boiling again, adding 500 µL of 1% HAuCl<sub>4</sub> quickly and maintaining the water boiling. The solution turned red within 5 min and the final color changed to brilliant red. Boiling continued for 10 min, the heating source was removed, and the colloid was not stirred until cold. Last the solution was transferred to a 50 mL volumetric flask and diluted to 50.0 mL with water. The AuNP concentration is 58 µg/mL Au, and its size is 10 nm.

#### 2.2. Procedure

A 400 µL 1.1 mol/L HNO<sub>3</sub> solution, 40 µL 10 µmol/L HgCl<sub>2</sub> solution, 100 µL 5 mmol/L KMnO<sub>4</sub> solution, 40 µL 4.0 mmol/L HAuCl<sub>4</sub> solution, 400 µL 1.0 mol/L HCl solution, 80 µL 10 mg/mL PEG10000 solution and 100 µL 0.14 mmol/L NH<sub>2</sub>OH•HCl solution were added to a 5 mL graduated tube successively, and mixed well. Then, a 100 µL 3.75 mol/L NaH<sub>2</sub>PO<sub>2</sub> solution was added, and diluted to 2.0 mL to start the reaction. The mixture was placed in room temperature (25 °C) for 15 min. The RRS spectra were recorded by means of synchronous scanning excited wavelength  $\lambda_{ex}$  and emission wavelength  $\lambda_{em}$  ( $\lambda_{ex} - \lambda_{em} = \Delta \lambda = 0$ ) on fluorescence spectrophotometer. The RRS intensity at 370 nm ( $I_{370 \text{ nm}}$ ) and the blank solution without Hg<sup>2+</sup> ( $I_0$ ) were recorded. The value of  $\Delta I = (I_{370 \text{ nm}} - I_0)$  was calculated.

#### 3. Results and discussion

#### 3.1. Principle

In 0.2 mol/L HCl-0.22 mol/L HNO<sub>3</sub> media, the NaH<sub>2</sub>PO<sub>2</sub> reduce Hg<sup>2+</sup> to form Hg nanoparticles (HgNPs) and the HgNPs catalyzed NaH<sub>2</sub>PO<sub>2</sub> reduce HAuCl<sub>4</sub> to generate small AuNPs. These formed small AuNPs also have catalysis on the reaction of NaH<sub>2</sub>PO<sub>2</sub>-HAuCl<sub>4</sub> that called as autocatalysis. The reaction of Hg<sup>2+</sup>-NaH<sub>2</sub>PO<sub>2</sub> was considered by the RRS technique and the result showed that the Hg<sup>2+</sup> was reduced by NaH<sub>2</sub>PO<sub>2</sub> to form Hg nanoparticles. The linear relationship between Hg(II) concentration and  $\Delta I$  value showed that the reaction order for Hg(II) catalyst is 1. According to our results and the reference (Chen, 1980; Jiang et al., 2008), the main reaction mechanism is as follows,

$HgCl_2 + NaH_2PO_2 \rightarrow Hg$	(fast)	(1	)	
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$hHg \rightarrow HgNPs$	(fast)	(2)	)
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 $Au^{3+} + NaH_2PO_2 \rightarrow Au^+ \quad (fast) \tag{3}$ 

$$Au^{+} + NaH_{2}PO_{2} \xrightarrow{HgNPs \text{ catalysis}} Au \quad (slow)$$
(4)

$$nAu \rightarrow AuNP$$
 (fast) (5)

$$Au^{+} + NaH_2PO_2 \xrightarrow{AuNYS catalysis} Au \quad (slow) \tag{6}$$

 $nAuNPs \rightarrow Big AuNPs$  (fast) (7)

The reduction of  $Hg^{2+}$  to Hg and  $Au^{3+}$  to  $Au^+$  is fast step in the catalytic reaction because the reducer concentration of NaH<sub>2</sub>PO<sub>2</sub> is high (0.188 mol/L) and the value of potential different is big. There are strong inter-atomic forces between metal Hg atoms or metal Au atoms or small AuNPs, that is, metal Hg atoms, metal Au atoms and small AuNPs were rapidly aggregated to HgNPs, AuNPs and big AuNPs respectively. Eqs. (4) and (6) are slow, that is, rate control steps. When the concentration of Hg<sup>2+</sup> increased, the formed nanocatalyst both HgNPs and AuNPs increased, the two rate control steps quicken, and the catalytic reaction rate enhanced that will result to form more AuNPs, the color of mixing solution quickly changed from blue to red. Thus, the AuNPs RRS intensity at 370 nm increased linearly with concentration of  $Hg^{2+}$  increasing. According to the principles of catalytic kinetic analysis and RRS (Chen, 1980; Jiang et al., 2008), we

established a new catalytic RRS method to determine trace  $Hg^{2+}$  (Fig. 1).

## 3.2. Scanning electron microscopy

According to the procedure, taken 1.5 mL of reaction solution into a 2 mL centrifuge tube and centrifuged 20 min  $(150 \times 100 \text{ r/min})$ , discarded the supernatant and added water to 1.5 mL then dispersed 30 min with ultrasonic. After centrifuged again, added 1 mL water and dispersed 30 min. Taken 2 µL sample solution by pipette dripping on silicon slice and dried naturally. The scanning electron microscope (SEM) indicated that there are AuNPs in the system with average size of 60 nm (Fig. 2). For the blank, there are very few of AuNPs which cannot be observed by SEM.

## 3.3. RRS spectra

Fig. 3 is the RRS spectra of  $Hg^{2+}$ –HAuCl<sub>4</sub>–NaH<sub>2</sub>PO<sub>2</sub> system, there are three strong Rayleigh scattering peaks at 280 nm, 370 nm and 550 nm respectively. The peak at 280 nm caused by the lamp maximal emission at 280 nm, and other two peaks are owing to the RRS effect of AuNPs. The peak at 370 nm is the strongest and the RRS intensity increased linearly with concentrations of Hg<sup>2+</sup>, other two peaks do not have good linear response. Thus, a wavelength of 370 nm was selected to detect trace Hg<sup>2+</sup>. Because the Hg(II) catalyst concentration is very low (5.0–450 × 10<sup>-9</sup> mol/L), the formed Hg nanoparticles do not affect the RRS spectra of the analytical systems.

The RRS spectra of  $Hg^{2+}$ -Na $H_2PO_2$  system were also investigated. When  $Hg^{2+}$  concentration increased, the RRS signal enhanced as in Fig. 4. This result showed that there are HgNPs in the system because the Na $H_2PO_2$  reduce  $Hg^{2+}$ to  $Hg^0$  that were aggregate to the particles. This is very significance because metal Hg sublime easily at room temperature and its scanning electron microscope (SEM) cannot be recorded.

# 3.4. Catalysis of AuNPs

In the absence of  $Hg^{2+}$ , the AuNP reaction between HAuCl<sub>4</sub> and NaH<sub>2</sub>PO<sub>2</sub> is slow. In the presence of 10 nm AuNPs, the



Figure 1 Principle of nanocatalytic RRS method for  $Hg^{2+}$ .



Figure 2 Scanning electron microscope of the nanocatalytic system. 0.22 mol/L HNO<sub>3</sub> + 450 nmol/L HgCl<sub>2</sub> + 0.5 mmol/L KMnO<sub>4</sub> + 80 µmol/L HAuCl<sub>4</sub> + 0.2 mol/L HCl + 0.4 mg/mL PEG10000 + 7 µmol/L NH<sub>2</sub>OH•HCl + 0.188 mol/L NaH<sub>2</sub>PO<sub>2</sub>.



Figure 3 RRS spectra of  $Hg^{2+}$ -HAuCl<sub>4</sub>-NaH<sub>2</sub>PO<sub>2</sub> system. a: 0.22 mol/L HNO<sub>3</sub> + 0.5 mmol/L KMnO<sub>4</sub> + 80 µmol/L HAuCl<sub>4</sub> + 0.2 mol/L HCl + 0.4 mg/mL PEG10000 + 7 µmol/L NH<sub>2</sub>OH•HCl + 0.188 mol/L NaH<sub>2</sub>PO<sub>2</sub>, incubation time for 15 min; b: a + 20 nmol/L HgCl<sub>2</sub>; c: a + 100 nmol/L HgCl<sub>2</sub>; d: a + 200 nmol/L HgCl<sub>2</sub>.



Figure 4 RRS spectra of the  $Hg^{2+}-NaH_2PO_2$  system. a: 0.22 mol/L HNO<sub>3</sub> + 0.5 mmol/L KMnO<sub>4</sub> + 0.2 mol/L HCl + 0.4 mg/mL PEG10000 + 7 µmol/L NH<sub>2</sub>OH•HCl + 0.188 mol/L NaH<sub>2</sub>PO<sub>2</sub>; b: a + 1000 nmol/L Hg<sup>2+</sup>; c: a + 2000 nmol/L Hg<sup>2+</sup>; d: a + 3000 nmol/L Hg<sup>2+</sup>.



Figure 5 Effect of HNO<sub>3</sub> concentration on the  $\Delta I$ .



Figure 6 Effect of HCl concentration on the  $\Delta I$ .



Figure 7 Effect of KMnO<sub>4</sub> concentration on the  $\Delta I$ .



Figure 8 Effect of NH<sub>2</sub>OH•HCl concentration on the  $\Delta I$ .





 $\begin{array}{lll} \mbox{Figure 10} & Effect \ of \ NaH_2PO_2 \ concentration \ on \ the \ \Delta I. \ 0.22 \ mol/L \\ L & HNO_3 + 200 \ nmol/L & HgCl_2 + 0.5 \ mmol/L & KMnO_4 + 80 \\ \mu mol/L & HAuCl_4 + 0.2 \ mol/L & HCl + 0.4 \ mg/mL & PEG10000 + 7 \\ \mu mol/L & NH_2OH \bullet HCl. \end{array}$ 



particle reaction enhanced greatly, and the  $\Delta I$  increased linearly with the AuNP concentration ( $C_{AuNP}$ ) in the range of 100– 3000 ng/mL. The regression equation is  $\Delta I = 1.76$  $C_{AuNP} + 17.6$ , with a coefficient of 0.9789. That is, with increasing of  $C_{AuNP}$ , the RRS effect enhanced linearly at 370 nm due to more big AuNPs generated from the particle



Figure 12 Influence of metal ions on the determination.  $0.22 \text{ mol/L HNO}_3 + 200 \text{ nmol/L HgCl}_2 + 0.5 \text{ mmol/L KMnO}_4 + 80 \mu \text{mol/L HAuCl}_4 + 0.2 \text{ mol/L. Ratio} = [(I_{370 \text{ nm}})_{\text{Hg}} - (I_{370 \text{ nm}})_{\text{Hg}} + \text{MI}]/(I_{370 \text{ nm}})_{\text{Hg}}$ , the MI represents metal ion.

reaction of HAuCl<sub>4</sub>–NaH<sub>2</sub>PO<sub>2</sub>. This result showed that the AuNPs exhibited catalysis of HAuCl<sub>4</sub>–NaH<sub>2</sub>PO<sub>2</sub> reaction. Thus, the formed small AuNPs have catalytic effect on the catalytic reaction of  $Hg^{2+}$ –HAuCl<sub>4</sub>–NaH<sub>2</sub>PO<sub>2</sub>.

# 3.5. Optimization conditions of the nanocatalytic system

HNO<sub>3</sub> and HCl are good media of the AuNP reaction. We have studied the effect of HNO<sub>3</sub> concentrations on the detection of 0.2 µmol/L Hg<sup>2+</sup>. The result indicated that the catalytic rate became fast when the concentration of HNO<sub>3</sub> increased, the more AuNPs formed and caused the  $\Delta I$  value increasing. When the concentration of HNO<sub>3</sub> was 0.22 mol L<sup>-1</sup>, the value of  $\Delta I$  reached its maximum. When the concentration of HNO<sub>3</sub> exceeded 0.22 mol/L, the value of  $\Delta I$  tended to decline (Fig. 5), so we chose 0.22 mol/L as the final concentration of HNO<sub>3</sub>. We also studied the effect of HCl concentration on  $\Delta I$  value reached the maximum, and then the  $\Delta I$  value tended decreased when the HCl concentration increased further (Fig. 6). So we selected 0.2 mol/L of HCl concentration in this experiment.

In the mixed acidic medium, KMnO<sub>4</sub> is a strong oxidization reagent that can oxidize organic mercury and low valence mercury to Hg<sup>2+</sup> in sample. The effect of KMnO<sub>4</sub> concentration on the  $\Delta I$  was studied. Results showed that the  $\Delta I$  value significantly increased as the KMnO<sub>4</sub> concentration increased. When the concentration of KMnO<sub>4</sub> was 0.25 mmol/L, the  $\Delta I$ value reached its maximum. When the concentration of KMnO<sub>4</sub> exceeded 0.25 mmol/L, the  $\Delta I$  tended to decline (Fig. 7), so, a 0.25 mmol/L KMnO<sub>4</sub> was chosen for use. Because NH<sub>2</sub>OH•HCl was the reductant of KMnO<sub>4</sub>, so we also studied the effects of NH<sub>2</sub>OH•HCl concentration on the  $\Delta I$ . When the NH<sub>2</sub>OH•HCl concentration was 7 µmol/L, the  $\Delta I$  value reached the maximum (Fig. 8), so a 7 µmol/L NH<sub>2</sub>OH•HCl was selected for use. The color of the sample solution appeared pale purple after addition of NH<sub>2</sub>OH•HCl, which indicated that there is no NH<sub>2</sub>OH•HCl existence before the next experimental process.

HAuCl<sub>4</sub> is an important component because it reduced catalytically by NaH<sub>2</sub>PO<sub>2</sub> to form AuNPs. We have studied the effect of HAuCl<sub>4</sub> concentrations in the presence of 0.2 µmol/ L Hg<sup>2+</sup> (Fig. 9). The results indicated that the catalytic rate became fast when the concentration of HAuCl<sub>4</sub> increased. When the HAuCl<sub>4</sub> concentration was 0.08 mmol/L, the  $\Delta I$  value reached its maximum. When the concentration of HAuCl<sub>4</sub> was exceeded 0.08 mmol/L, the value of  $\Delta I$  tended to decline, so we chose 0.08 mmol/L HAuCl<sub>4</sub>. We also studied the effect of NaH<sub>2</sub>PO<sub>2</sub> concentration on the  $\Delta I$  (Fig. 10). When the concentration of NaH<sub>2</sub>PO<sub>2</sub> was 0.188 mol/L, the  $\Delta I$  value reached the maximum. When NaH<sub>2</sub>PO<sub>2</sub> concentration was exceed 0.188 mol/L, the value of  $\Delta I$  tended to decline. Thus, we chose 0.188 mol/L NaH<sub>2</sub>PO<sub>2</sub>.

PEG10000 is a good stabilizer for the AuNPs. Results showed that When PEG10000 added concentration was 0.4 mg/mL, the value of  $\Delta I$  reached its maximum. So, a 0.4 mg/mL PEG10000 was selected. Because the blank

Water sample	Added value (nM)	Found value (nM, $n = 5$ )	RSD <sup>a</sup> (%)	Recovery (%)	AFS (nM)
Tap-water 1 – 200	No detected	-	-	No detected	
	200	216	5.0	108	
Tap-water 2	_	No detected	-	-	No detected
-	200	203	6.0	101.5	
Tap-water 3 – 300	_	No detected	-	-	No detected
	300	326	3.0	108.7	
Waste water 1	_	225	-	-	212
	300	318	4.0	106	
Waste water 2 – 250	_	308	-	-	296
	250	267	5.0	106.8	

reaction rate appeared slower compared to catalytic reaction under the room temperature (25 °C), the catalytic reaction can quickly reached a stable value too, and the room temperature was chosen for use. The result indicated that the  $\Delta I$ value increased when the reaction time became long in the range of 7–15 min, and it achieved the maximum in 15 min (Fig. 11). Then the value of  $\Delta I$  hold constant. Thus, a reaction time of 15 min at 25 °C was chosen for use. The time zero point was defined as the moment when NaH<sub>2</sub>PO<sub>2</sub> was added.

#### 3.6. Effect of foreign substances

According to the procedure, the influence of coexistent metal ions on the detection of 0.2  $\mu$ mol/L Hg<sup>2+</sup> was tested, with a relative error of  $\pm 10\%$ . Fig. 12 showed that the common metal ions such as 100 times of Pb(II), Co(II), Ca(II), Mg(II), Cr(III), 70 times of Ba(II) and 50 times of Fe(III), Zn(II) did not enhance the Hg<sup>2+</sup> catalytic reaction. This indicated the RRS method has good selectivity.

# 3.7. Working curve

Under the optimal conditions,  $\text{Hg}^{2+}$  concentration  $(C_{\text{Hg}}^{2+})$  and their corresponding  $\Delta I$  showed a good linear relationship. The linear range was 5–450 nmol/L, with a regression equation of  $\Delta I = 2.941C + 38.4$ , coefficient of 0.9801, and detection limit of 0.1 nmol/L. Compared to some reported methods for mercury (Liang et al., 2011; Chen, 1980; Wang et al., 2012; Zhou et al., 2012), this RRS method was more sensitive, and simple.

### 3.8. Sample analysis

We applied this catalytic RRS method to measure the concentration of  $Hg^{2+}$  in water samples. The water samples were pretreated by aqua regia. Then, the content of  $Hg^{2+}$  was determined according to the procedure. In the sample solutions, a known  $Hg^{2+}$  was added respectively, to measure the recovery. Table 1 showed that the recovery was in the range of 101.5–108.7%, relative standard deviation was in the range of 3–6%, and this catalytic RRS result was agreement with that of atomic fluorescence spectrometry (AFS). This indicated that this RRS assay is precision and accuracy.

# 4. Conclusion

Based on the trace  $Hg^{2+}$  catalysis of the AuNP reaction between NaH<sub>2</sub>PO<sub>2</sub> and HAuCl<sub>4</sub>, and the resonance Rayleigh scattering (RRS) effect of AuNPs at 370 nm, a simple, sensitive and selective nanocatalytic RRS method was developed for the determination of  $5.0 \times 10^{-9}$ -450  $\times 10^{-9}$  mol/L Hg<sup>2+</sup>. This new nanocatalytic RRS method was applied to detect Hg<sup>2+</sup> in water sample, with satisfactory results.

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