Talanta 82 (2010) 1791–1796

Contents lists available at ScienceDirect

Talanta

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

Solid phase extraction of trace Hg(II) on silica gel modified with 2-(2-oxoethyl)hydrazine carbothioamide and determination by ICP-AES

Xiaoli Chai[a,b], Xijun Chang[a,b,*], Zheng Hu[a,b], Qun He[a,b], Zhifeng Tu[a,b], Zhenhua Li[a,b]

[a] Department of Chemistry, Lanzhou University, Tianshui Road 222, Lanzhou, Gansu 730000, PR China
[b] Key Laboratory of Nonferrous Metals Chemistry and Resources Utilization, Gansu Province, PR China

ARTICLE INFO

Article history:
Received 27 April 2010
Received in revised form 23 July 2010
Accepted 28 July 2010
Available online 5 August 2010

Keywords:
Silica gel
2-(2-Oxoethyl)hydrazine carbothioamide
Solid phase extraction
Hg(II)
Inductively coupled plasma atomic emission spectrometry (ICP-AES)

ABSTRACT

In this work, a new 2-(2-oxoethyl)hydrazine carbothioamide modified silica gel (SG-OHC) sorbent was prepared and applied for preconcentration of trace mercury(II) prior to the measurement by inductively coupled plasma atomic emission spectrometry (ICP-AES). The optimization of some analytical parameters affecting the adsorption of the analyte such as acidity, shaking time, sample flow rate and volume, eluent condition, and interfering substances were investigated. At pH 3, the maximum static adsorption capacity of Hg(II) onto the SG-OHC was 37.5 mg g^{-1}. The quantitative recovery (>95%) of Hg(II) could be obtained using 2 mL of 0.5 mol L^{-1} HCl and 1% CS(NH$_2$)$_2$ solution as eluent. Common coexisting substances did not interfere with the separation of mercury(II) under optimal conditions. The detection limit of present method was 0.10 ng mL^{-1}, and the relative standard deviation (RSD) was lower than 4.0% ($n = 8$). The prepared sorbent was successfully applied for the preconcentration of trace Hg(II) in certified and water samples with satisfactory results.

1. Introduction

Mercury is a global pollutant and is identified as a highly toxic element because of its accumulative and persistent character in the environment and living organisms [1]. Therefore, determination of mercury becomes increasingly important, especially in water system. However, due to both the low concentration of mercury in water and the complexity of the sample, a separation and preconcentration step is required prior to the determination by inductively coupled plasma atomic emission spectrometry (ICP-AES).

Different methods, viz., cloud point extraction [2], electroanalytical techniques [3], liquid–liquid extraction [4], membrane filtration [5], ion exchange [6], solid phase extraction (SPE) [7], coprecipitation [8], resin chelation [9] and foam-flotation [10] have been reported for preconcentration of trace mercury(II). Of all these methods, SPE has gained rapid acceptance because of its merits of simplicity, rapidity, high enrichment factor, ease of automation, low consumption of organic solvents and the ability of combination with different detection techniques [11,12]. The basic principle of SPE is the transfer of the analyte from the aqueous phase to bind to active sites of the adjacent solid phase [13]. A variety of solid phase have been applied in SPE, such as active carbon [14], silica gel [15], microcrystalline naphthalene [16], Amberlite XAD resin [17], polyurethane foam [18] and alumina [19]. Among these solid phases, silica gel modified with organic compounds as metal chelating agent exhibits some definite advantages such as porosity, large specific surface area, good mechanics, the thermal and chemical stability [20,21]. Hence, silica gel is selected as the solid phase in this work.

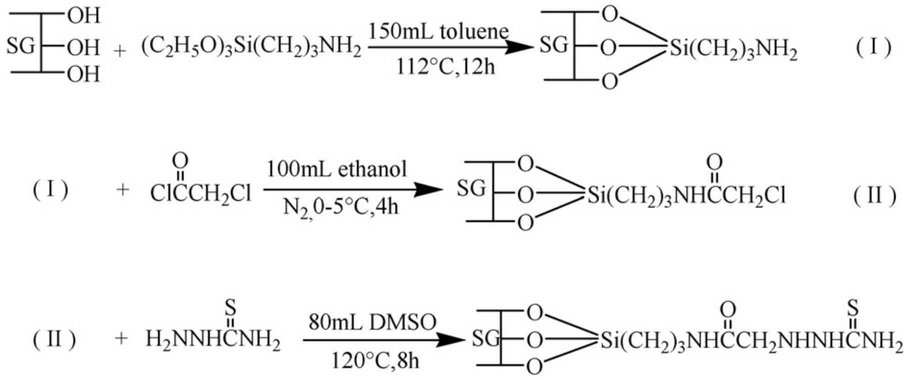
In general, mercury(II) can complex strongly with ligands containing nitrogen and sulfur atoms [22,23]. 2-(2-Oxoethyl)hydrazine carbothioamide, which contains nitrogen, sulfur and oxygen atoms, can probably form chelate with Hg(II).

In this study, a new silica gel modified with 2-(2-oxoethyl) hydrazine carbothioamide was synthesized and characterized. The prepared sorbent possessed a high adsorption selectivity for mercury(II). Parameters that affect the quantitative recoveries of the analytes were studied in detail. The proposed method was validated by analyzing the certified reference materials (GBW 08301, river sediment and GBW 08303, polluted farming soil) and applied to the analysis of natural water samples with satisfactory results. The proposed method showed simple, fast and accurate characteristics for separation and preconcentration of Hg(II) from environmental samples.

---

* Corresponding author at: Tel.: +86 931 891 2422; fax: +86 931 891 2582.
E-mail address: gscxl_love@126.com (X. Chang).

0039-9140/$ – see front matter © 2010 Elsevier B.V. All rights reserved.
doi:10.1016/j.talanta.2010.07.076



Scheme 1. Synthetic route of the SG-OHC.

2. Experimental

2.1. Instruments and apparatus

An IRIS Advantage ER/S inductively coupled plasma emission spectrometer (TJA, USA) (RF power supply: 1.15 kW; Ar carrier gas flow rate: 0.6 L min^{-1}; Ar auxiliary gas flow rate: 1.0 L min^{-1}; Ar coolant gas flow rate: 14.0 L min^{-1}; viewing height: 15 mm; wavelength of Hg:194.227 nm) was employed for the metal ion determination. The pH was adjusted by a pHs-3C digital pH meter (Shanghai Lei Ci Device Works, Shanghai, China). Infrared spectra (4000–400 cm^{-1}) in KBr were recorded on a Nicolet NEXUS 670 FT-IR spectrometer (Nicolet, Madison, WI, USA). An YL-110 peristaltic pump (General Research Institute for Non-ferrous Metals, Beijing, China) was used in the preconcentration process. A PTFE (polytetrafluoroethylene) column (45 mm × 3.0 mm i.d.) was used in this work.

2.2. Chemicals and reagents

Unless otherwise stated, all reagents used were of analytical grade and all solutions were prepared with doubly distilled deionized water. Standard labware and glassware used were repeatedly cleaned with HNO_3 and rinsed with double distilled water, according to a published procedure [24].

Silica gel (100–200 mesh) was purchased from Qingdao Hailang Chemical Factory (Qingdao, SD, China). 3-Aminopropyltrimethoxysilane was obtained from Chemical Engineering Corporation of Ocean University of China (Qingdao, China). Chloroacetyl chloride (The First Reagent Factory, Shanghai, China) and thiosemicarbazide (Beijing Chemical Industry, Beijing, China) were used in this work.

Standard stock solution of Hg(II) (1 mg mL^{-1}) were prepared by dissolving spectral pure-grade $HgCl_2$ (The First Reagent Factory, Shanghai, China) in double distilled water with the addition of hydrochloric acid (The First Reagent Factory, Shanghai) and further diluted daily prior to use. The standard reference material (GBW 08301, river sediment and GBW 08303, polluted farming soil) was provided by the National Research Center for Certified Reference Materials (Beijing, China).

2.3. Sample preparation

River water was collected from Yellow River, Lanzhou, China. The water samples were filtered through a 0.45 μm polytetrafluoroethylene (PTFE) millipore filter and acidified to a pH of about 2 with concentrated HCl prior to storage for use. Tap water samples were taken from our laboratory without pretreatment before determination, the pH value was adjusted to 3 with 0.1 mol L^{-1} HCl or 0.1 mol L^{-1} $NH_3{\cdot}H_2O$ prior to use. The water samples were analyzed within 2 weeks after collection.

The reference materials (GBW 08301 and GBW 08303) were treated according to the following method: to oxidize organic matter such as humic acid, the sample was digested by oxidizing UV-photolysis in the presence of 1% H_2O_2 using a low pressure Hg-lamp which was integrated in a closed quartz vessel [25], and then the digested samples were immediately filtered through a millipore cellulose nitrate membrane, pore size 0.45 μm, acidified to pH 3 with hydrochloric acid and stored in precleaned polyethylene bottles prior to use [26].

2.4. Preparation of new sorbent

2.4.1. Synthesis of silica gel modified with 3-aminopropyltrimethoxysilane (APSG)

In order to remove any adsorbed metal ions and increase the content of –OH, silica gel (SG) was activated by refluxing with concentrated hydrochloric acid under stirring for 8 h, then the activated silica gel was filtered and washed with doubly distilled water to neutral and dried under vacuum at 70 °C for 8 h. To prepare APSG, 10 g of activated silica gel was dispersed into 150 mL toluene, and then 10 mL 3-aminopropyltrimethoxysilane was gradually added into the solution with continuous stirring. The mixture was refluxed for over night. The final product was filtered off, washed with toluene, alcohol and diethyl ether, and then dried under vacuum at 70 °C for 8 h.

2.4.2. Synthesis of silica gel modified with 2-(2-oxoethyl)hydrazine carbothioamide (SG-OHC)

The dried APSG was transferred into the flask, then 10 mL chloroacetyl chloride and 100 mL of dried ethanol were added in sequence. The reaction mixture was stirred for 4 h at 0–5 °C under nitrogen atmosphere, filtered under vacuum, washed with ethanol and dried under vacuum at 60 °C for 6 h. Finally, 1.4 g of thiosemicarbazide was dissolved in 80 mL dimethyl sulfoxide (DMSO), and 5.0 g of the dried product was added to the solution. The mixture reacted at 120 °C for 8 h. The resulting reaction product (SG-OHC) was filtered, washed with a small amount of DMSO, ethanol and dried in an oven at 80 °C for 8 h. The synthetic route of the SG-OHC is schematically represented in Scheme 1.

2.5. General procedures

2.5.1. Batch method

Portions of 10 μg mL^{-1} (1 mL) standard or sample solutions containing Hg(II) were transferred into a 25 mL beaker, and the pH

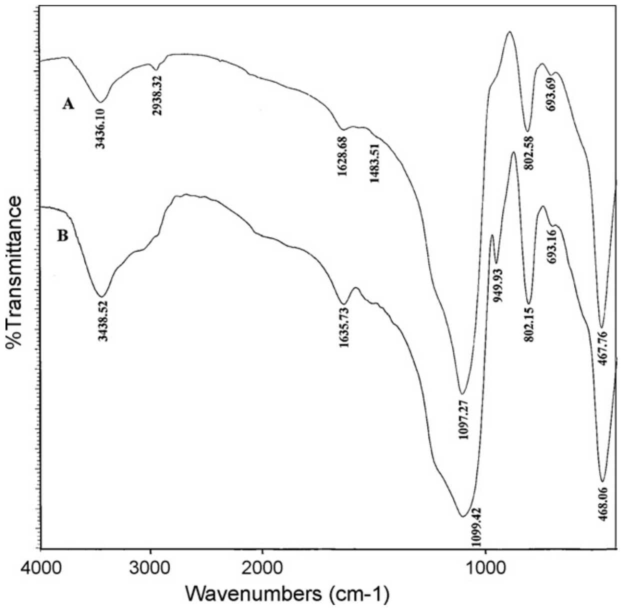


Fig. 1. IR spectra of APSG (A) and SG-OHC (B).

value was adjusted to the desired value with 0.1 mol L^{-1} HCl and 0.1 mol L^{-1} $NH_3 \cdot H_2O$. Then the volume was adjusted to 10 mL with double distilled water. 30 mg of SG-OHC was added, and the mixture was shaken vigorously for 30 min to facilitate adsorption of the metal ions onto the sorbents. After centrifugation, the concentrations of the metal ions in the solution were directly determined by ICP-AES. The adsorbent was then eluted with $CS(NH_2)_2$ and HCl under optimum conditions. Finally, the desorbed metal ions were measured by ICP-AES.

2.5.2. *Column procedure*

A total 50 mg of SG-OHC was packed in the PTFE column plugged with a small portion of glass wool at both ends. Before using, 0.5 mol L^{-1} HCl and doubly distilled deionized water were successively passed through the microcolumn in order to equilibrate, clean and neutralize it. Portions of aqueous standard or sample solutions containing Hg(II) were prepared, and the pH value was adjusted to 3 with 0.1 mol L^{-1} HCl and 0.1 mol L^{-1} $NH_3 \cdot H_2O$. Each solution was passed through the column at a flow rate of 2.0 mL min^{-1} controlled by a peristaltic pump. Afterwards, the bound metal ions were stripped off from the column with 1% $CS(NH_2)_2$ and 0.5 mol L^{-1} HCl solution. The analytes in the elution were determined by ICP-AES.

3. Results and discussion

3.1. *FT-IR spectra and elemental analysis*

The modified silica gel was confirmed by IR analysis as shown in Fig. 1. The peaks of APSG at 467.76, 802.58, 1097.27, 1628.68, 2938.32 and 3436.10 cm^{-1} were attributed to δ(Si–O–Si), ν(Si–O–Si), longitudinal SiO_2 lattice vibration, $\delta(H_2O)$, $\nu(CH_3)$ and ν(Si–OH), respectively [28]. The peak appeared at 3436.10 cm^{-1}, was assigned to ν(N–H) [27]. Compared with that of APSG, the IR of SG-OHC showed remarkable spectral changes. According to the literature [27], the peak at 1099.42 cm^{-1} was due to $\nu_{\text{C–N}}$, the peak at 1635.73 cm^{-1} was caused by $\nu_{\text{C=O}}$, the band around 3438.52 cm^{-1} could be assigned to $\nu_{\text{N–H}}$. In theory, $\nu_{\text{C=S}}$ is at 1190–1270 cm^{-1} and its intensity is weak, so the peak was not obvious in the spectrum.

Elemental analysis indicated 6.89% carbon, 1.97% nitrogen and 1.913% hydrogen in SG-OHC. It could be calculated that 1 g silica gel contained 0.01478 g 2-(2-oxoethyl)hydrazine carbothioamide. This was caused by the reason that only the surface of silica gel was modified.

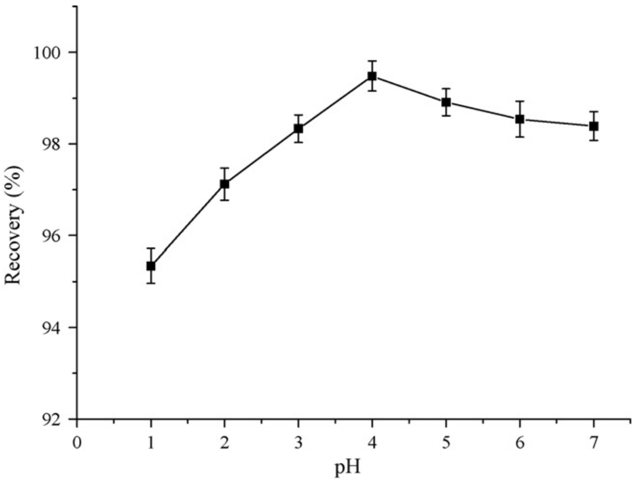


Fig. 2. Effect of pH on adsorption of 1.0 μg mL^{-1} Hg(II) on SG-OHC. Other conditions: shaking time 30 min, temperature 25 °C.

Consequently, the above experimental results suggest that silica gel is successfully modified by 2-(2-oxoethyl)hydrazine carbothioamide.

3.2. *Effect of pH*

Solution acidity affects metal adsorption: proton in acid solution can protonate binding sites of the chelating molecules, and hydroxide in basic solution may complex and precipitate many metals [28]. In order to determine this parameter, the effect of the pH on the static adsorption of Hg(II) was tested by equilibrating 30 mg of SG-OHC sorbent with 10 mL of the buffer solutions containing 1.0 μg mL^{-1} of Hg(II), Cr(III), Fe(III), Co(II), Ni(II), Cu(II), Zn(II), Cd(II), Mn(II) and Pb(II) in range of pH 1–7 using the batch procedure. It could be seen in Fig. 2 that quantitative extraction (>95%) of Hg(II) occurred from pH 1–7. The results indicated that the SG-OHC possessed excellent adsorptive selectivity towards Hg(II) at pH < 4 than any other ions. To avoid hydrolyzing at higher pH values and get higher recovery, pH 3 was selected as the enrichment acidity for further study. In addition, Zn(II) was not enriched by SG-OHC at pH 3, Cr(III), Co(II), Cd(II) and Pb(II) could be adsorbed by SG-OHC about 10–16% at pH 3, Ni(II), Mn(II), Fe(III) and Cu(II) could be adsorbed by SG-OHC about 31–67% at pH 3, but they did not interfere with enrichment and determination of Hg(II).

3.3. *Effect of shaking time*

The shaking time is considered to be another important factor in determining the availability of the newly synthesized SG-OHC for the selective extraction of metal ions. In this work, different shaking time (ranged from 2 to 40 min) was studied for the percentage extraction of Hg(II) by SG-OHC. It is found in Fig. 3 that only 10 min was needed for maximum recovery (>95%), which indicated that SG-OHC possessed rapid adsorption kinetics for Hg(II). Therefore, it is suitable for application in flow system.

3.4. *Effect of flow rate*

In the column SPE system, the flow rate of the sample solution was an important parameter not only affecting the retention of cations on the adsorbent, but also controlling the time of analysis. Consequently, the effect of flow rate on adsorption of Hg (II)

Table 1
Elution recovery (%) for Hg(II) adsorbed on SG-OHC sorbent.

Concentration of HCl (mol L^{-1})[a]	0.5	1	2	4	5
Recovery (%)					
HCl	9.4	20.6	29.7	69.7	100
HCl + 1% $CS(NH_2)_2$	100	100			
Eluent volume (mL)[b]	2	3	4	5	6
Recovery (%)	96.1	97.3	98.6	100	100

[a] 5 mL.
[b] 0.5 mol L^{-1} HCl + 1% $CS(NH_2)_2$.

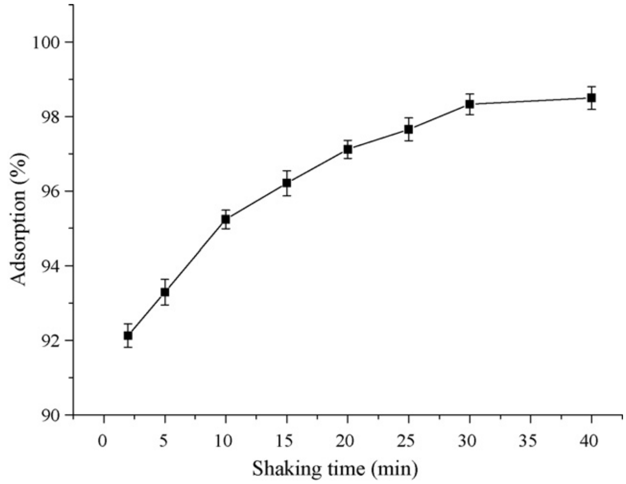


Fig. 3. Effect of shaking time on adsorption of 1.0 μg mL^{-1} Hg(II) on SG-OHC. Other conditions: pH 3, temperature 25 °C.

was examined under the optimum conditions (pH, eluent, etc.) by passing 10 mL of sample solution through the microcolumn. The flow rates were adjusted in the range of 0.5–3.5 mL min^{-1} controlled by a peristaltic pump. The results are shown in Fig. 4. It was found that the retention of the studied ion was practically not changed up to 2.0 mL min^{-1} flow rate. The recovery of the analyte decreased slightly when the flow rate was over 2.0 mL min^{-1}. Thus, a flow rate of 2.0 mL min^{-1} was selected throughout the column experiment.

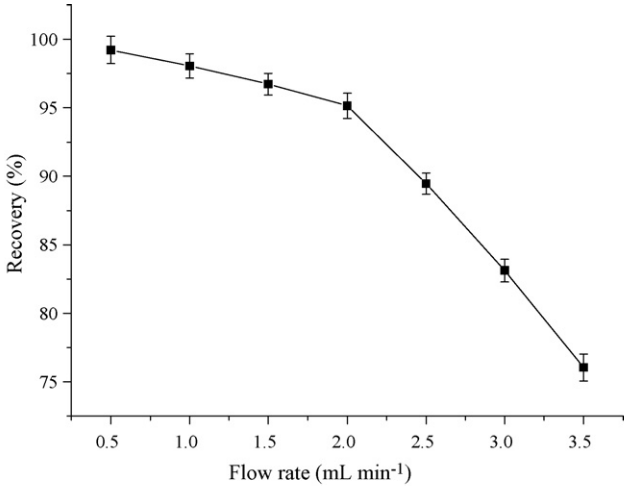


Fig. 4. Effect of solution flow rates on adsorption of 1.0 μg mL^{-1} Hg(II) on SG-OHC. Other conditions: 50 mg of SG-OHC, volume 10 mL, pH 3, temperature 25 °C.

Table 2
Effect of coexisting substances on percent recovery of 1.0 μg mL^{-1} Hg(II) adsorbed on SG-OHC.

Coexisting substances	Concentration (μg mL^{-1})	Recovery of Hg(II) (%)
NO_3^-	3000	97.12
Cl^-	3000	99.34
K^+	2000	95.82
Na^+	2000	98.74
SO_4^{2-}	1000	100
Ca^{2+}	500	96.41
Mg^{2+}	300	100
Fe^{3+}	50	100
Cr^{3+}	50	100
Cd^{2+}	50	100
Zn^{2+}	50	100
Ni^{2+}	50	100
Cu^{2+}	50	100
Pb^{2+}	50	100
Mn^{2+}	50	100
Co^{2+}	20	95.73
Humic acid	2000	95.97

Table 3
Analytical results for the determination of Hg(II) in certified reference materials.

Sample	Concentration of Hg(II) (μg g^{-1})		
	Found by present method[a]	Certified value	The data of *t*-test
GBW08301	0.22 ± 0.06	0.22 ± 0.01	0
GBW08303	2.13 ± 0.05	2.11 ± 0.04	0.89

[a] The value following "±" is the standard deviation (*n* = 5).

3.5. Maximum sample volume, enrichment factor and elution condition

In order to explore the possibility of concentrating low concentration of Hg(II) from large volumes, the maximum applicable volume must be determined. For this purpose, 50, 100, 150, 200, 250 and 300 mL of sample solutions containing 1.0 μg mL^{-1} Hg(II) were

Table 4
Analytical results for the determination of trace Hg(II) in natural water samples.

Water samples	Concentration of Hg(II) (μg L^{-1})		Recovery (%)
	Added	Found[a]	
Yellow River water	0	0.94 ± 0.12	
	5	5.90 ± 0.17	99.2
	10	11.02 ± 0.09	100.8
Tap water	0	–	
	5	4.86 ± 0.21	97.2
	10	9.98 ± 0.16	99.8

[a] The value following "±" is the standard deviation (*n* = 5).

Table 5
Comparison of figures of merit for determination of Hg(II).

Solid support	Ligand	pH	Capacity (mg g^{-1})	Ref.
MAA	TMPTM-TAN	7	6.42	[31]
Bicarbonate-treated peanut hull carbon	–	3.5–10.0	28.57	[32]
Amberlite XAD-4 resin	Brilliant green	–	4.12	[17]
Ordered mesoporous silica	Chitosan	6.5	13.5	[33]
Silica gel	Diphenylcarbazone	7	0.8	[34]
Sol–gel silicas	1,5 Diphenylcarbazide	6	5.6	[35]
Silica gel	2-(2-Oxoethyl)hydrazine carbothioamide	3	37.5	Present work

passed through the microcolumn with 50 mg sorbent at the optimum flow rate. Quantitative recovery (>95%) was obtained for the sample volume of 100 mL for Hg(II), and at greater volume percent of recovery decreased.

The elution condition was also studied by using various concentrations and volumes of thiourea and HCl solution for the desorption of retained Hg(II). The obtained results showed (Table 1) that 5 mL of 5 mol L^{-1} HCl or 2.0 mL of 1% (w/w) $CS(NH_2)_2$ and 0.5 mol L^{-1} HCl solution was sufficient for complete elution, but in order to avoid using high concentration of HCl, so 2 mL of 0.5 mol L^{-1} HCl and 1% $CS(NH_2)_2$ solution was chosen as eluent for stripping off the adsorbed Hg(II). When the flow rate for desorption of Hg(II) was varied from 0.2 to 2.0 mL min^{-1}, the results showed that target ion could be quantitatively eluted at flow rate below 0.5 mL min^{-1}. Therefore, the enrichment factor of 50 for Hg(II) was obtained (because maximum sample volume was 100 mL and eluent volume was 2 mL).

3.6. Adsorption capacities

The capacity study was adopted from the paper recommended by Maquieira et al. [29]. 10 mL of Hg(II) solutions of a series of concentrations (50–300 μg mL^{-1}) were adjusted to the appropriate pH and followed the batch method. A breakthrough curve was gained by plotting the concentration (μg mL^{-1}) vs. the microgram of Hg(II) adsorbed per gram of SG-OHC. From the breakthrough curve, the saturated adsorption amount of SG-OHC for Hg(II) was found to be 37.5 mg g^{-1} at pH 3.

3.7. Effect of coexisting substances

The effect of different substances on the adsorption of Hg(II) on SG-OHC were investigated. In these experiments, solutions of 1.0 μg mL^{-1} of Hg(II) containing the added interfering substances were prepared according to the batch procedure. The results represented in Table 2 showed that 3000-fold NO_3^-, Cl^-, 2000-fold K^+, Na^+, 1000-fold SO_4^{2-}, 500-fold Ca^{2+}, 300-fold Mg^{2+}, 50-fold Fe^{3+}, Cr^{3+}, Cd^{2+}, Zn^{2+}, Ni^{2+}, Cu^{2+}, Pb^{2+}, Mn^{2+}, 20-fold Co^{2+} and 2000-fold humic acid did not interfere with the separation and determination of the analyte. The high recoveries (>95%) of Hg(II) in the presence of interfering substances revealed that the present solid phase extractant (SG-OHC) allowed the interference-free extraction of trace Hg(II) in the environmental samples.

3.8. The detection limits and analytical precision

Under the selected conditions, eight portions of standard solutions were enriched and analyzed simultaneously following the general procedure. In accordance with the definition of IUPAC, the detection limit of the method was calculated based on three times the standard deviation of eight runs of the blank solution [30]. The detection limits (3σ) were found to be 0.10 ng mL^{-1} for Hg(II). The relative standard deviation (RSD) of the eight replicate determinations was lower than 4.0%, which indicated that the method possessed good precision for the analysis of trace Hg(II) in solution samples.

3.9. Application of the method

The proposed method has been applied to the determination of trace Hg(II) in certified reference materials (GBW 08301, river sediment and GBW 08303, polluted farming soil). According to the results listed in Table 3, the estimated content by the proposed method were in good agreement with the certified values of Hg(II). For the analysis of Yellow River water and tap water samples, the standard addition method was used. The results listed in Table 4 shows that the recoveries of Hg(II) were in range of 97.2–100.8%. The obtained results indicated the suitability of the SG-OHC for the preconcentration of Hg(II) from environmental samples prior to ICP-AES analysis.

3.10. Comparison with other methods

Table 5 shows the comparison of the suggested method with other SPE methods reported in the literature [31–35,17]. As seen from the data, the present method possessed the advantages of higher enrichment acidity and adsorption capacity than the reported other SPE methods.

4. Conclusion

In this work, a simple, rapid, accurate and reliable method was developed for determination of trace level of Hg(II) in environmental samples based on adsorption of silica gel modified with 2-(2-oxoethyl)hydrazine carbothioamide. This sorbent exhibited good characteristics for separation and preconcentration of Hg(II) in aqueous solution, such as excellent selectivity, fast adsorption equilibrium, easy elution, good adsorption capacity and interference-free extraction of trace Hg(II). Moreover, the preparation of SG-OHC was relatively simple and convenient. The proposed method was successfully applied to the analysis of trace Hg(II) in certified and environmental samples with satisfactory results. In summary, the new sorbent shows good analytical potential for analysis of trace Hg(II) from environmental samples.

References

[1] Z.J. Li, Q. Wei, R. Yuan, X. Zhou, H.Z. Liu, H.X. Shan, Q.J. Song, Talanta 71 (2007) 68.
[2] J.G. Chen, H.W. Chen, X.Z. Jin, H.T. Chen, Talanta 77 (2009) 1381.
[3] M.S. Çelebi, H. Özyörük, A. Yıldız, S. Abacı, Talanta 78 (2009) 405.
[4] E.M. Martinis, P. Bertón, R.A. Olsina, J.C. Altamirano, R.G. Wuilloud, J. Hazard. Mater. 167 (2009) 475.
[5] H. Bessbousse, T. Rhlalou, J.-F. Verchère, L. Lebrun, J. Membr. Sci. 325 (2008) 997.
[6] P. Houserová, P. Kubáň, V. Kubáň, Electrophoresis 27 (2006) 4508.
[7] L.N. Zhang, X.J. Chang, Z. Hu, L.J. Zhang, J.P. Shi, R. Gao, Microchim. Acta 168 (2010) 79.
[8] M. Vircavs, V. Rone, A. Pelne, D. Vircava, Anal. Chim. Acta 299 (1994) 291.
[9] A.A. Atia, A.M. Donia, K.Z. Elwakeel, React. Funct. Polym. 65 (2005) 267.
[10] M.S. Hosseini, H. Hashemi-Moghaddam, Talanta 67 (2005) 555.
[11] K. Pyrzynska, M. Trojanowicz, Crit. Rev. Anal. Chem. 29 (1999) 313.
[12] Q. He, X.J. Chang, Q. Wu, X.P. Huang, Z. Hu, Y.H. Zhai, Anal. Chim. Acta 605 (2007) 192.

[13] E.M. Soliman, M.B. Saleh, S.A. Ahmed, Anal. Chim. Acta 523 (2004) 133.
[14] Q. He, Z. Hu, Y. Jiang, X.J. Chang, Z.F. Tu, L.N. Zhang, J. Hazard. Mater. 175 (2010) 710.
[15] Z.F. Tu, Z. Hu, X.J. Chang, L.J. Zhang, Q. He, J.P. Shi, R. Gao, Talanta 80 (2010) 1205.
[16] C.M. Xiong, Z.C. Jiang, B. Hu, Anal. Chim. Acta 559 (2006) 113.
[17] H. Hashemi-Moghaddam, H.A. Panahi, M.N. Nezhati, Anal. Lett. 42 (2009) 1911.
[18] N. Burham, Cent. Eur. J. Chem. 6 (2008) 641.
[19] S.A. Ahmed, J. Hazard. Mater. 156 (2008) 521.
[20] M. Kooshki, E. Shams, Anal. Chim. Acta 587 (2007) 110.
[21] G.A.E. Mostafa, M.M. Hassanien, K.S. Abou-El-Sherbini, V. Gorlitz, Anal. Sci. 19 (2003) 1151.
[22] L. Hakim, A. Sabarudin, K. Oshita, M. Oshima, S. Motomizu, Talanta 76 (2008) 1256.
[23] X.P. Zhu, S.D. Alexandratos, Ind. Eng. Chem. Res. 44 (2005) 7490.
[24] D.P.H. Laxen, R.M. Harrison, Anal. Chem. 53 (1981) 345.
[25] C. Guéguen, C. Belin, B.A. Thomas, F. Monna, P.Y. Favarger, J. Dominik, Anal. Chim. Acta 386 (1999) 155.
[26] X.J. Chang, N. Jiang, H. Zheng, Q. He, Z. Hu, Y.H. Zhai, Y.M. Cui, Talanta 71 (2007) 38.
[27] S.W. Li, R.L. Fan, Practical Organic Chemistry Handbook, Shanghai Science and Technology Press, 1981, pp. 557–564.
[28] L.J. Zhang, X.J. Chang, Y.H. Zhai, Q. He, X.P. Huang, Z. Hu, N. Jiang, Anal. Chim. Acta 629 (2008) 84.
[29] A. Maquieira, H.A.M. Elmahadi, R. Puchades, Anal. Chem. 66 (1994) 3632.
[30] G.L. Long, J.D. Winefordner, Anal. Chem. 55 (1983) 712A.
[31] I. Dakova, I. Karadjova, V. Georgieva, G. Georgiev, Talanta 78 (2009) 523.
[32] C. Namasivayam, K. Periasamy, Water Res. 27 (1993) 1663.
[33] D.H. Chen, B. Hu, C.Z. Huang, Talanta 78 (2009) 491.
[34] J. Fan, Y.X. Qin, C.L. Ye, P.G. Peng, C.L. Wu, J. Hazard. Mater. 150 (2008) 343.
[35] A. Khan, F. Mahmood, M.Y. Khokhar, S. Ahmed, React. Funct. Polym. 66 (2006) 1014.