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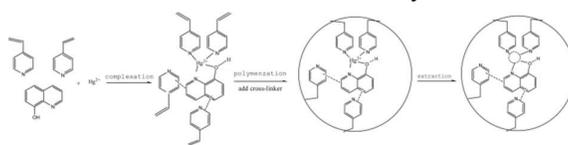
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Hg(II)-ion-imprinted polymer for the sensitive and selective determination of mercury in water samples



Synthesis and characterization of Hg(II)-ion-imprinted polymer and its application for the determination of mercury in water samples

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A novel Hg(II) ion imprinted polymer (IIP) was synthesized by bulk polymerization using Hg(II) ion as a template, 4-vinylpyridine (4-VP) as a monomer, 8-hydroxyquinoline as a specific ligand for Hg(II), and ethylene glycol dimethacrylate (EDMA) as a crosslinker. The Hg(II) IIP was systematically characterized by adsorption test, FTIR spectroscopy, pore size analysis and SEM studies. Under the optimized condition, the maximum adsorption of Hg (II) IIP were 143.8 mg/g, nearly three times higher than that of the non ion-imprinted polymer (NIP) (52.6 mg/g). The synthesized Hg (II) IIP had a good selectivity for Hg (II) ion compared with other ions. The trace amounts of Hg (II) in tap water and river water samples were measured by the sequential injection-vapor generation-atomic fluorescence spectrometry (SI-VG-AFS) in a linear range of 0.01-1 µg/L, with the limit of detection (LOD) of 0.005 µg/L.

1. Introduction

Mercury has been considered as a human health hazard because it may cause kidney toxicity, neurological damage, paralysis, chromosome breakage and birth defects.¹ Due to the high toxicity and accumulative character of mercury, the determination of trace amounts of mercury in environmental samples is of great importance. Highly sensitive methods are demanding for the determination of Hg-species at extremely low levels in natural waters after suitable separation and preconcentration steps. A great variety of analytical procedures for Hg(II) preconcentration and separation have been reported such as liquid-liquid extraction,^{2,3} coprecipitation,^{4,5} and solid phase extraction (SPE).⁶⁻¹¹ SPE is one of the most common method utilized for preconcentration of heavy metals due to its flexibility, absence of emulsion and simplicity.¹² Ion imprinted polymers (IIPs) have distinct advantages such as large number of binding sites, high surface area and porosity, which make them better adsorbents for the selective collection of mercury from hazardous waste

solutions.¹³

IIPs have been demonstrated their unique application for selective preconcentration and separation of Hg(II) ions.¹³⁻¹⁸ For example, Singh et al.¹⁹ prepared Hg(II) imprinted copolymer by the formation of a binary complex of mercury (imprint ion) with 4-(2-thiazolylazo) resorcinol (TAR) and thermally copolymerizing with methacrylic acid (monomer) and ethylene glycol dimethacrylate (EDMA, crosslinker), and the adsorption capacity of Hg (II) on IIP and NIP is 25.1 and 11.6 mg/g, respectively. Liu et al.⁸ reported Hg (II) imprinted copolymers by copolymerizing mercury chloride, diazoaminobenzene (DAAB) and 4-vinylpyridine (4-VP) using EDMA as crosslinker in the presence of 2,2-azobisisobutyronitrile (AIBN) as initiator. The Hg-(II) imprinted copolymers had a higher adsorption capacity of 41.1 mg/g, 3.5 times higher compared with non-imprinted copolymers (11.8 mg/g). However, to the best of our knowledge, there is no previous report concerning the use of 8-hydroxyquinoline, a good chelating agent for metal ions,²⁰ for the synthesis of any metal ion imprinted polymers to improve the adsorption capacity and selectivity.

In this paper, we report the synthesis of imprinted polymer using 4-vinylpyridine (4-VP) as a monomer, ethylene glycol dimethacrylate(EDMA) as a crosslinker and 8-hydroxyquinoline

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as a specific ligand for Hg(II), and the adsorption characteristics of the Hg(II) IIP and NIP have been investigated. The maximum adsorption capacity of IIP and NIP towards Hg(II) were 143.8 mg/g and 52.6 mg/g, respectively, which was consistent with the Claudio Baggiani's hypothesis that if a NIP shows binding properties toward a target molecule, MIP would show a significant imprinting effect.²¹ In addition, the IIP shows excellent selectivity for Hg(II) compared with other metal ions. The application of the developed IIP in the determination of mercury in water samples was also studied using SI-VG-AFS with high sensitivity.

2. Experimental

2.1. Instrument and Reagent

4-Vinylpyridine (4-VP), 8-hydroxyquinoline (HQ) and ethylene glycol dimethacrylate (EDMA) were obtained from Alfa (Tianjin, China). 2,2-azobisisobutyronitrile (AIBN), mercury(II) chloride and potassium borohydride were purchased from Tianjin Chemical Reagent Company, China. The 1000 mg/L stock solution of mercury was obtained from the National Research Center for Certified Reference Materials (NRCCRM, China). Working solutions were freshly prepared from the stock solutions by stepwise dilution just before use. The potassium tetrahydroborate (III) solutions for the determination of mercury were prepared daily by dissolving the reagent in 0.5% potassium hydroxide solution. All reagents were analytical grade without further purification. Ultrapure water was used throughout all the experiment. The IR spectra were measured by an Avatar 360 FTIR spectrophotometer (Nicolet, USA). The morphological structure of the polymers were characterized by a Quanta 450 scanning electronic microscopy (SEM, Bruker, Germany). The surface roughness of the IIP and NIP were investigated by Nanoscope IIIa scanning probe microscope (Bruker, Germany). The concentration of the Hg(II) after the treatment of EDTA of IIP was detected by Hydra II AA Automated Mercury Analyzer (TELEDYNE Leeman Labs, USA). The adsorption data were obtained from TriStar II 3020 (V1.03) surface area and porosity measurement system (Micromeritics Inc, USA). AFS-922 sequential injection vapor generation 100 double-channel non-dispersive atomic fluorescence spectrometry (Titan Instruments Co. Ltd., Beijing, China) was used for the measurement of trace amounts of Hg.

2.2. Preparation of Hg(II) Ion Imprinted Polymer (IIP)

HgCl₂ (1 mmol), 8-hydroxyquinoline (1 mmol), 4-vinylpyridine (4 mmol) and EDMA (16 mmol) and AIBN (0.24 mmol) were dissolved in 15 mL dimethyl sulfoxide (DMSO). Please note that the crosslinker EDMA was added in large excess in this reaction to ensure the complete polymerization. After fully dispersing and deoxygenating with sonication and nitrogen gas, the mixture was heated at 60 °C with water bath for 24 h. The resulting polymers were crushed, ground and sieved with a 0.2 mm sieve. Subsequently the synthesized polymers were treated with EDTA (0.1 mol/L) for 2 h to remove the Hg (II) ion, followed by washing with methanol and water, respectively. The final Hg(II) IIP was dried in vacuum at 60 °C. The non-imprinted polymer (NIP) was prepared using the same protocol as described above without adding Hg (II) in the polymerization mixture. In order to test whether the Hg(II) in IIP was completely removed, we measured the Hg(II) concentration after the EDTA treatment. Briefly, 30 mg IIP was weighted, and treated with 10.00 mL EDTA (0.1 mol/L) for 3 times, and then washed with 10.00 mL 10% HNO₃ (v/v). The analytical results showed that the concentration of Hg(II) was 0.012 µg/L in the washed EDTA solution, and the concentration of Hg(II) in the washed HNO₃ was not found, demonstrating the complete removal of Hg(II) in IIP after the treatment with EDTA.

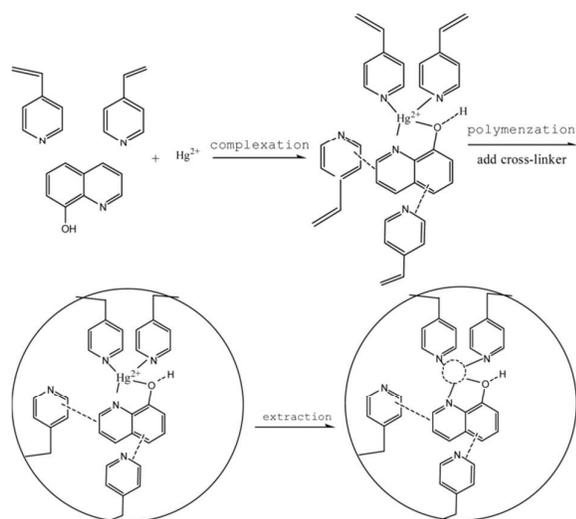
2.3. Absorption test

An amount of 10 mg Hg(II) IIP was incubated with 10.0 mL Hg (II) with a certain concentration for 25 min. Then the solution was centrifuged and filtrated to detect the Hg (II) concentrations left in solution by EDTA titration monitored by a UV-Vis spectrophotometer. The quantity of adsorption (Q) can be calculated based on the equation $Q = (C_0 - C)V/w$, in which Q is the quantity of adsorption (mg/g), C₀ is the Hg(II) ion concentration of the solution before absorption (mg/L), C is the Hg(II) concentration of the solution after absorption (mg/L), w is the weight of the adsorbent (g), and V is the volume of the solution (L).

3. Results and Discussion

3.1. Characterization of the IIP

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Scheme 1 Schematic representation of the method applied for IIP preparation

As shown in the Scheme 1, the IIP was formed by the reaction between 4-vinyl pyridine and 8-hydroxyquinoline in the presence of Hg(II) as described in the experimental section. The binary complex of Hg(II) ion with 8-hydroxyquinoline was prepared under the similar condition in the absence of 4-vinylpyridine. The IR spectra of Hg(II)-IIP (curve a), NIP (curve b) and Hg(II)-IIP without extraction (curve c) were shown in Fig.1. The IR spectra of Hg(II)-IIP (curve a) is identical to that of NIP (curve b), indicating the Hg²⁺ was removed from IIP after washed with EDTA (0.1 mol/L) and methanol. The bonds in the region of 3,200-3,600 cm⁻¹ corresponds to the OH groups in 8-hydroxyquinoline, which appeared in NIP, Hg(II)-IIP without extraction and Hg(II)-IIP after washing with EDTA (0.1 mol/L) and with methanol, demonstrating the presence of 8-hydroxyquinoline in the IIP structure. It can be seen that the unleached IIP represents a strong IR band at 1163 cm⁻¹ (curve c) other than the band of leached IIP at 1137 cm⁻¹ (curve b). This band is probably related to ν_{C-O} stretching vibration of 8-hydroxyquinoline due to the banding of Hg²⁺ and OH group of 8-hydroxyquinoline. A strong IR band at 2990 cm⁻¹ is found in the unleached IIP (curve c), and instead of the IIP (curve a) and NIP (curve b), indicating that coordination interaction between Hg²⁺ and intramolecular chelated OH group in 8-hydroxyquinoline in the unleached IIP.²² It can also be observed that the unleached IIP with a IR band at 1639 cm⁻¹ (curve c) is different from the band of

leached IIP at 1622 cm⁻¹ (curve b), which is another evidence for the coordination of N-atom of 8-hydroxyquinoline and 4-vinylpyridine with Hg²⁺.

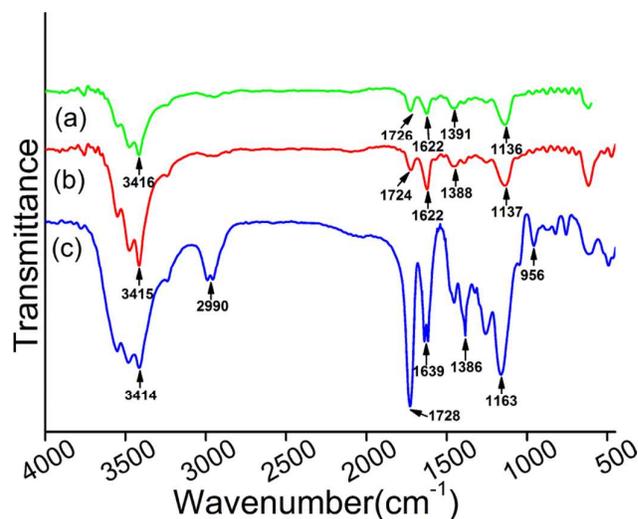


Fig.1 IR spectra of IIP (curve a), NIP (curve b), and unleached IIP (curve c)

Surface area and pore size of Hg(II) IIP were examined by TriStar II3020(V1.03).surface area and porosity measurement system (Micromeritics Inc., USA). As shown in Table 1 and Fig.2, Hg(II) IIP has a high pore specific surface area with the pore size less than 140 Å.

Table 1 The surface and pore size of Hg(II)-IIP

Item	Surface Area (m ² g ⁻¹)	Item	Pore Size (Å)
Single point surface area at (P/Po = 0.199822709)	234.3917	Adsorption average pore width (4V/A by BET)	67.3139
BET Surface Area	241.5468	BJH Adsorption average pore diameter (4V/A)	139.104
Langmuir Surface Area	332.9128	BJH Desorption average pore diameter (4V/A)	125.137
t-Plot Micropore Area	49.938		
t-Plot External Surface Area	191.6088		
BJH Adsorption cumulative (D = 17–3,000 Å)	167.427		
BJH Desorption cumulative (D = 17–3,000 Å)	182.8403		

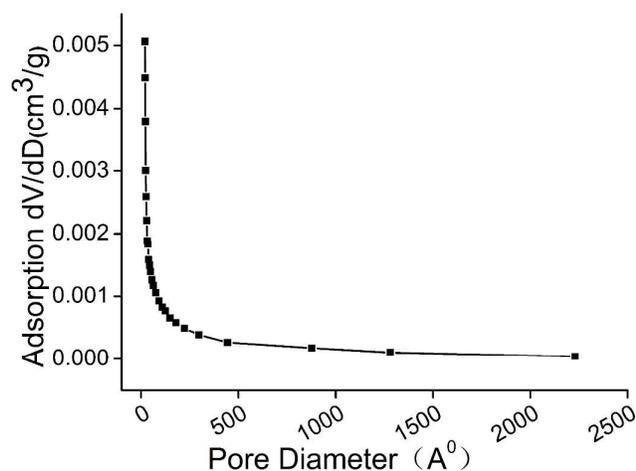


Fig. 2 Adsorption pore distribution

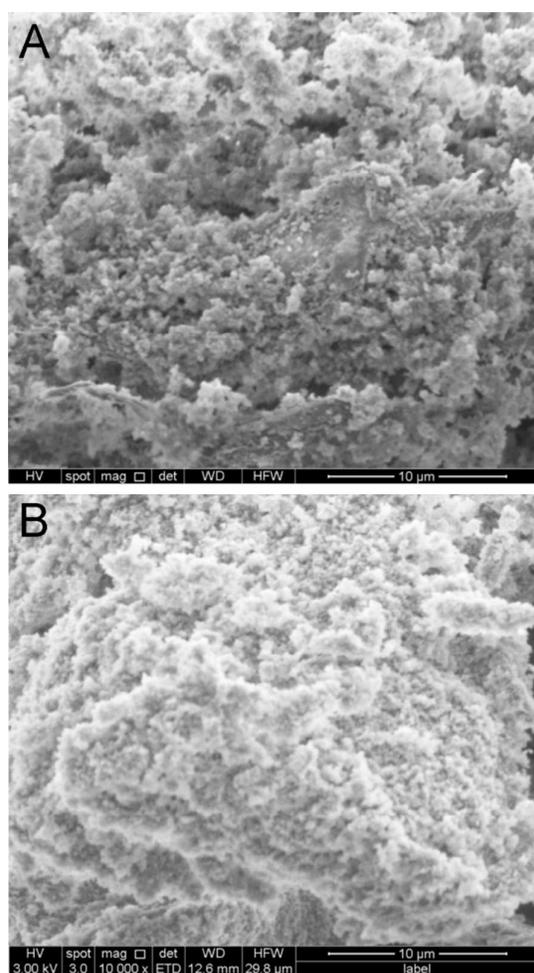


Fig. 3 SEM images of IIP (A) and NIP (B)

The morphological structure characterization of the IIP and NIP were studied by BRUKER Quanta 450 scanning electronic microscopy (SEM). As shown in Fig. 3, both the IIP (Fig. 3A) and NIP (Fig. 3B) exhibited rough surface, which are beneficial to the homogeneous binding of template ions. The roughness of the surface is of paramount importance in adsorption processes that makes the mass transfer of metal ions to the polymer surface easier, enhancing the metal ion adsorption. The morphological structure of IIP (Fig. 3A) showed more cavities than that of NIP (Fig. 3B), therefore the IIP had much higher adsorption of Hg(II) ion than that of NIP, which is consistent with the high imprinting ability of IIP toward Hg(II) ion. We also used Atomic Force Microscopy (AFM) to calculate the surface roughness of the IIP (Fig. 4A) and NIP (Fig. 4B), and the results showed that the mean roughness of IIP (~ 6.22 nm) was about 5 times higher than that of NIP (~ 1.24 nm), which was consistent with the results from the SEM images.

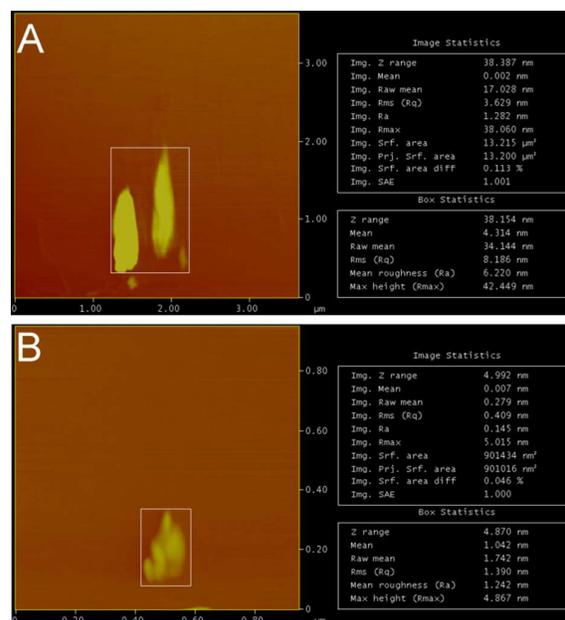


Fig. 4 Surface roughness of IIP (A) and NIP (B)

3.2 Adsorption Test

Adsorption time of Hg(II) ion in Hg(II) IIP was investigated. As shown in Fig. 5, the amount of Hg(II) ion adsorption increases with time, and reaches an adsorption equilibrium at 25 min. Therefore, the adsorption time of 25 min was selected in the following studies.

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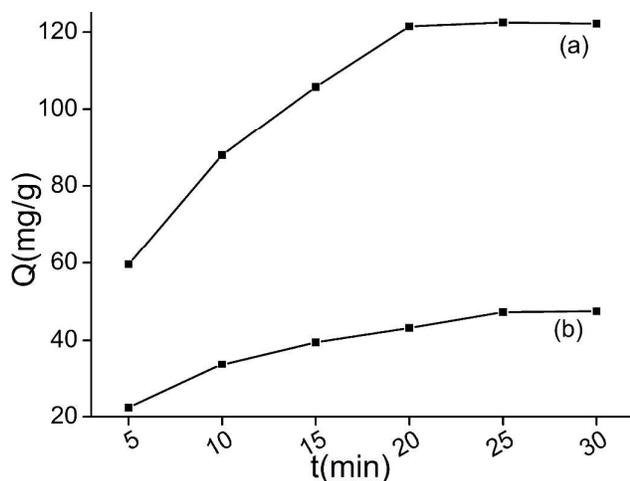


Fig. 5 Effect of time on Hg (II) adsorption Hg (II) concentration: 150 mg/L; pH 3.5. a. Hg(II) on Hg(II) IIP, b. Hg(II) on NIP.

The effect of pH on the amount of Hg (II) ion adsorption was also investigated. As shown in Fig.6, Hg(II) ion adsorption quantity reached its maximum at pH 3.5. The nitrogen atom of 8-hydroxyquinoline and pyridine group would be protonated at pH < 3, resulting in the decrease of the Hg(II) ion adsorption. In contrast, Hg(II) ion could be hydrolyzed at pH > 4, and thus the Hg(II) ion adsorption amount would be decreased. Therefore, the optimum pH is 3.5.

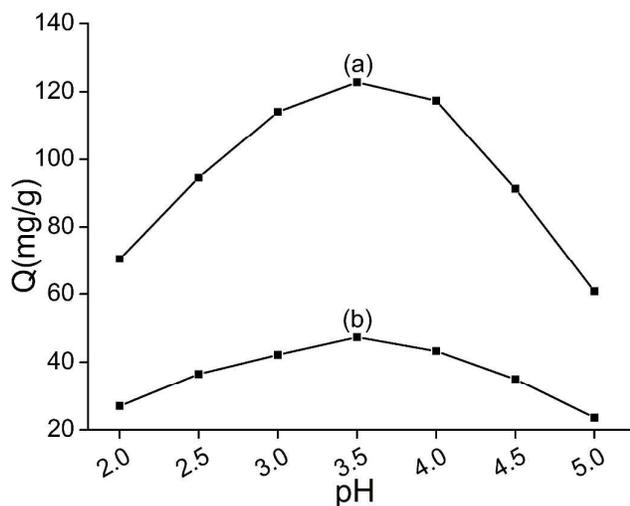


Fig. 6 Effect of pH on Hg(II) sorption with Hg(II) concentration of 150 mg/L. a. Hg(II) on Hg(II) IIP, b. Hg(II) on NIP.

Adsorption isotherm of Hg(II) on the IIP Equilibrium adsorption isotherm is typically described by Langmuir model¹⁸

$$C/Q = C/Q_{\max} + 1/(Q_{\max} \cdot K) \quad (1)$$

In the Eq.1, C is Hg(II) ion concentration at equilibrium, Q and Q_{\max} refer to metal adsorption amount at equilibrium and maximum metal adsorption amount (mg/g) respectively, and K is the Langmuir isotherm constant. The relationship between C and Q is shown in Fig. 7. As shown in Fig. 8, the curve of C/Q and C has a good linear relationship, demonstrating that the adsorption of Hg(II) IIPs for Hg(II) ion can be well fitted to the Langmuir model. The isotherm equation is as follows:

$$C/Q = 0.00638C + 0.1963 \quad (R=0.998) \quad (2)$$

The adsorption quantity at equilibrium is 156.7 mg/g calculated from Langmuir model by the Eq. 2, which is close to the maximum adsorption quantity measured by the experiment (143.8 mg/g). On the contrary, the adsorption amount of NIP for Hg(II) ion is 52.6 mg/g, further demonstrating the imprinting ability of the synthesized Hg(II) IIP.

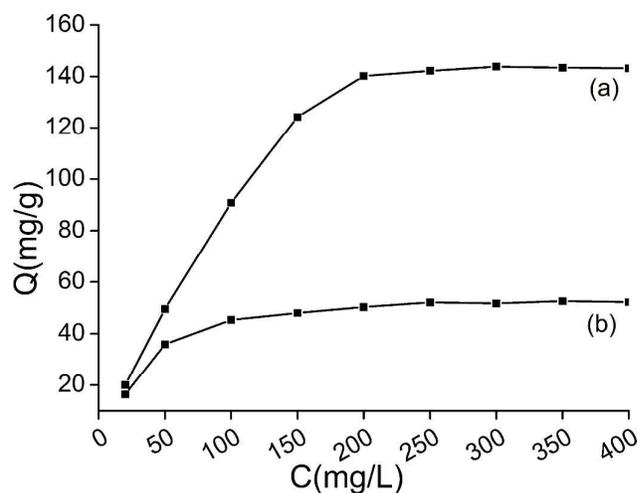


Fig. 7 Adsorption isotherm of Hg(II) IIP (a) and NIP (b) (T=25°C, pH=3.5)

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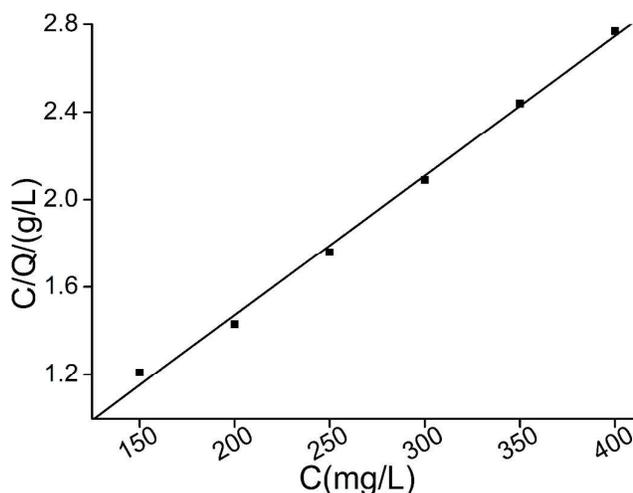


Fig. 8 Relation between C and Q

D(L/g)	0	0	0.0483	0.0489	0	0	0.06	0.0153	2.185
$S_{\text{Hg(II)/M}}$	∞	∞	45.2	44.7	∞	∞	36.4	142.8	1.00

Table 3 Selectivity coefficient and distribution ratio of NIP

Element	Cd^{2+}	Zn^{2+}	Pb^{2+}	Cu^{2+}	Mg^{2+}	Ca^{2+}	Al^{3+}	Fe^{3+}	Hg^{2+}
$D(\text{L g}^{-1})$	0.0598	0.0209	0.0503	0.0495	0.0593	0	0.057	0.0153	0.871
$S_{\text{Hg(II)/M}}$	14.6	41.7	17.3	17.6	14.7	∞	15.3	56.9	1.00

3.3. Selectivity of the IIP

The selectivity of IIP for Hg(II) over other foreign metal ions was investigated. The selectivity coefficient ($S_{\text{Hg(II)/M}}$) is defined as¹⁹:

$$S_{\text{Hg(II)/M}} = D_{\text{Hg}}/D_{\text{M}} \quad (1)$$

In the above Eq.1, D_{Hg} and D_{M} are the distribution ratios of the polymer with Hg(II) and the foreign metal ions. These distribution ratios were calculated using the following equation:

$$D = (C^i - C^f)v/(mC^f) \quad (2)$$

In the Eq. 2, C^i and C^f are the concentrations of the metal ion (mg/L) before and after the adsorption, respectively. v is the volume of the solution, and m is the mass of the polymer. As shown in Table 2, the Hg(II) ion adsorption amount of the Hg(II) IIP was higher than that of the eight foreign metal ions, indicating that the Hg(II) IIP has a good selectivity. Compared with the selectivity of Hg(II) NIP (Table 3), Hg(II)-IIP has a better selectivity than that of NIP, further demonstrating the successful synthesis of the IIP.

Table 2 Selectivity coefficient and distribution ratio of Hg(II)-IIP

Element	Cd^{2+}	Zn^{2+}	Pb^{2+}	Cu^{2+}	Mg^{2+}	Ca^{2+}	Al^{3+}	Fe^{3+}	Hg^{2+}
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3.4. Analytical applications

Real water sample of 200 mL (pH 3.5, adjusted by HNO_3) was stirred with 30 mg Hg (II) IIP for 30 min. After centrifugation (1500 rpm) the supernatant was removed and Hg (II) was eluted with 10.00 mL 10% HNO_3 (v/v) from the polymer. The 5.00 mL eluate was transferred into 10.00 mL volumetric flask. The trace amounts of Hg (II) in tap water and river water samples were measured by the sequential injection-vapor generation-atomic fluorescence spectrometry (SI-VG-AFS) under optimal instrumental parameters.¹⁹ The results showed a good linear relationship from 0.01 to 1.0 $\mu\text{g/L}$, with linear equation of $I=1813.2C-38.4$ (Hg (II), $\mu\text{g/L}$), and the linear coefficient of 0.999 and the limit of detection (LOD) of 0.005 $\mu\text{g/L}$. The results were showed in Table 4 and the recoveries were found in the range of 92-104%. These results demonstrated the suitability of the synthesized Hg(II) imprinted polymer for the quantitative recovery of Hg(II) from natural water samples, and the sensitivity of method was improved more than 20 times after the extraction. Compared with other SPE based methods (Table 5), the proposed IIP showed a improved performance as SPE sorbents for preconcentration of Hg^{2+} .

Table 4 Determination of Hg in natural water samples

Sample	Hg(II) Added	Hg(II) Found	RSD%	Average
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	($\mu\text{g/L}$)	($\mu\text{g/L}$)	(n=5)	Recovery (%)
Tap water	0	0.031	3.0	
	0.025	0.054	3.8	92.0
	0.050	0.079	2.9	96.0
River water	0	0.053	4.2	
	0.025	0.079	3.5	104.0
	0.050	0.102	4.8	98.0

Table 5 Comparison with other analytical methods of IIPs for the determination of mercury

Detection methods	Linear range ($\mu\text{g/L}$)	Detection limit ($\mu\text{g/L}$)	Recovery (% n=5)	References
IIP-SPE-VGA-AAS	10-50	2.875	99.7-101.7	19
IIP-SPE-CV-AAS	0.02-1	0.006		5
	0.05-2	0.02	95-98%	23
	0.13-25	0.05	94-105%	8
IIP-Electrochemical Sensor	0.5-100	0.1		24
	2.0-320	0.61	99.60%	25
	0.2-32	0.02	93.1-108.7	26
IIP-SPE-AFS	0.08-50	0.03	95.2-116.3	27
	0.05-15	0.015	93.0-105.2	28
	0.01-1	0.005	92.0-104.0	This work

4. Conclusions

The Hg ion-imprinted polymer was successfully synthesized by bulk polymerization using Hg(II) ion as a template, 4-vinylpyridine (4-VP) as a monomer, 8-hydroxyquinoline as a specific ligand for Hg(II) and ethylene glycol dimethacrylate as a crosslinker, and 2, 2-azobisisobutyronitrile as the initiator in dimethyl sulfoxide. The imprinting mechanization and characterization of polymers were studied by FTIR spectroscopy, SEM, adsorption test and pore size

analysis. The results showed that the imprinted polymer has very large adsorption amount, good selectivity for Hg(II), the adsorption capacity of Hg(II)-IIP is higher than that of the reported IIPs, and the selectivity studies showed that possible quantitative separation of Hg(II) from Zn(II), Cd(II), Cu(II) and Pb(II). We also demonstrated its application as an adsorbent for the selective separation and preconcentration of Hg(II) followed by the determination using AFS in the water samples

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Notes and references

- P. H. Fair, W. J. Dougherty and S. A. Braddon, *Toxicol. Appl. Pharmacol.*, 1985, **80**, 78-96.
- M. Andaç, S. Mirel, S. Şenel, R. Say, A. Ersöz and A. Denizli, *Int. J. Biol. Macromol.*, 2007, **40**, 159-166.
- D. K. Singh and S. Mishra, *Desalination*, 2010, **257**, 177-183.
- W. S. Qu, Y. H. Zhai, S. M. Meng, Y. Q. Fan and Q. Zhao, *Microchim. Acta*, 2008, **163**, 277-282.
- I. Dakova, I. Karadjova, V. Georgieva, G. Georgiev, *Talanta*, 2009, **78**, 523-529.
- L. B. Escudero, R. A. Olsina and R. G. Wuilloud, *Talanta*, 2013, **116**, 133-140.
- M. Firouzzare and Q. Q. Wang, *Talanta*, 2012, **101**, 261-266.
- Y. W. Liu, X. J. Chang, D. Yang, Y. Guo and S. G. Meng, *Anal. Chim. Acta*, 2005, **538**, 85-91.
- M. Shamsipur, J. Fasihi and K. Ashtari, *Anal. Chem.*, 2007, **79**, 7116-7123.
- M. Shamsipur, J. Fasihi, A. Khanchi, R. Hassani, K. Alizadeh and H. Shamsipur, *Anal. Chim. Acta*, 2007, **599**, 294-301.
- M. Shamsipur and A. B. Seidani, *React. Funct. Polym.*, 2011, **71**, 131-139.
- M. Shamsipur, B. S. Abbas, J. Fasihi, H. Sharghi, *Talanta*, 2010, **83**, 674-681.
- M. Soleimani and M. G. Afshar, *J. Anal. Chem.*, 2015, **70**, 5-12.
- M. Shamsipur and H. R. Rajabi, *Microchim. Acta*, 2013, **180**, 243-252.
- V. Vatanpour, S. S. Madaeni, S. Zinadini and H. R. Rajabi, *J. Membr. Sci.*, 2011, **373**, 36-42.
- G. H. Wu, Z. Q. Wang, J. Wang, C. Y. He, *Anal. Chim. Acta*, 2007, **582**, 304-310.
- S. Büyüktiryaki, R. Say, A. Denizli and A. Ersöz, *Talanta*, 2007, **71**, 699-705.
- T. Y. Guo, Y. Q. Xia, G. J. Hao, M. D. Song and B. H. Zhang, *Biomaterials*, 2004, **25**, 5905-5912.
- D. K. Singh and S. Mishra, *Desalination*, 2010, **257**, 177-183.
- C. Deraeve, C. Boldron, A. Maraval, H. Mazarguil, H. Gornitzka, L. Vendier, M. Pitie and B. Meunier, *Chem. Eur. J.*, 2008, **14**, 682-696.

Paper

- 21 C. Baggiani, C. Giovannoli, L. Anfossi, C. Passini, P. Baravalle and G. Giraudi, *J. Am. Chem. Soc.*, 2012, **134**, 1513-1518.
- 22 W. K. Zhao, *Instrumental Analysis*, Higher Education Press, 2001, pp. 159.
- 23 I. Dakova, T. Yordanova, I. Karadjova, *J. Hazard. Mater.*, 2012, **231–232**, 49–56.
- 24 T. Alizadeh, M. R. Ganjali and M. Zare, *Anal. Chim. Acta*, 2011, **689**, 52–59.
- 25 Z. H. Liu, S. Y. Huan, J. H. Jiang, G. I. Shen and R. Q. Yu, *Talanta*, 2006, **68**, 1120–1125.
- 26 X. C. Fu, X. Chen, Z. Guo, C. G. Xie, L. T. Kong, J. H. Liu and X. J. Huang, *Anal. Chim. Acta*, 2011, **685**, 21-28.
- 27 S. F. Xu, L. X. Chen, J. H. Li, Y. F. Guan and H. Z. Lu, *J. Hazard. Mater.*, 2012, **237-238**, 347-354.
- 28 Z. Zhang, J. H. Li, X. L. Song, J. P. Ma and L. X. Chen, *RSC Adv.*, 2014, **4**, 46444-46453.