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Silver (I)-coordinated bis(trimethoxysilylpropyl)amine Polycondensate for Adsorptive Removal of Iodide from Aqueous Solutions

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Abstract. Silver-coordinated bis(trimethoxysilylpropyl)amine polycondensate (TSPA-$AgNO_3$) was prepared and used to adsorb iodide ions in aqueous solutions. Batch adsorption studies were performed to investigate the effects of pH, temperature and coexisting NaCl on adsorption behavior. The results show that TSPA-$AgNO_3$ is easy to prepare and remarkably efficient in adsorbing iodide in water, especially in acidic solutions. Furthermore, increased temperature accelerated the adsorption, while coexisting NaCl inhibited the adsorption. TSPA-$AgNO_3$ also proved to be chemically stable in simulated environmental situations, which reveals a promising potential for applying this method to the disposal of radioactive iodide in environment water.
Keywords: Bis(trimethoxysilylpropyl)amine; Iodide; Adsorption; Coordination

1 Introduction

Radioactive iodine is released into the environment during the course of normal nuclear power plant operation and by nuclear accidents such as that in 2011 at Fukushima, Japan. Beyond that, radioactive iodide is increasingly used in biological experiments, medical treatments and disease diagnostics, creating radioactive iodide waste that is collected and stored [1,2]. Radioiodide has well-known risks. It can be readily accumulated in the body, especially in the thyroid, where it causes a significant health hazard. Different substances have been discussed as potential adsorbents to aid in removing radioactive iodide from experimental liquid waste and water contaminated by nuclear waste. For example, one study placed pure metallic silver membranes in a screwable holder for the removal of free radioiodide from I-radiopharmaceuticals [1], but the difficult preparation process may constrain this method's application in radioiodide separation from radioactive waste. Anion exchange cellulose paper membranes, which can remove radioactive iodide and effectively

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store it until it can be discarded, usually comprise dimethylaminoethyl groups as well as trimethylhydroxypropylammonium [2,3]. Iodide ions selectively permeate throughout the anion exchange paper membrane, forming ion pairs with cations in the solution. However, the anion exchange paper membrane method is not applicable to the problem of immobilizing radioactive iodide released into the environment, although it holds promising potential for reducing the volume of experimental waste.

Organophilic bentonite, zeolite, surfactant-modified zeolite and chemically pretreated clinoptilolite have been investigated for removing radioiodide from simulated environmental water [4-7]. A group of minerals including 11 Å tobermorite, attapulgite, allophone, clinoptilolite, ferrihydrite, goethite, pyrite and marcasite were assessed with respect to iodide retention in high-level radioactive waste (HLRW) repositories. Of these, organic surfactant modified bentonite (by hexadecylpridinium chloride, quaternary alkylmmonium salts, distearyldimethyl ammonium chloride, and others) was found to be the most promising candidate material because of high iodide adsorption capacity as well as selectivity of organoclays [8]. However, the required thermal stability of organoclays demands further discussion. Meanwhile, silver-impregnated porous solid adsorbents have proven to be quite effective in removing and sequestering radioactive iodine in the form of silver-iodine compounds [9-12]. Previous works have investigated porous solid materials such as styrene-divinylbenzene copolymer, silica gel, zeolite and active carbon. However, silver-impregnated porous solid adsorbents continue to present shortcomings such as a complex preparation process and chemical instability and uniformity, probably due to the lack of an especially strong affinity between silver ions and the porous material.

Bis(trimethoxysilylpropyl)amine (TSPA) is a silane coupling agent with applications as a controlled release carrier for water-soluble drugs; a surface treatment agent for enhancing corrosion resistance in steel; an adhesion promoter; and an optical, dielectric and protective coating [13, 14]. TSPA gels quickly in water and forms xerogel with good mechanical strength after drying. Taking advantage of these properties as well as the –NH– groups in TSPA, we prepared silver-coordinated bis(trimethoxysilylpropyl)amine polycondensate (hereafter referred to as TSPA-$AgNO_3$) for adsorbing iodide from aqueous solutions. We investigated the adsorption behavior of TSPA-$AgNO_3$ towards iodide, and we studied the effects of different parameters including solution pH, temperature and coexisting NaCl on the adsorption process in order to identify the optimal conditions for the adsorptive removal of iodide from aqueous solutions.

2 Experimental Details

2.1 Materials.

All reagents except TSPA were of analytical grade and used as received from Shanghai Boer Chemical Reagent Co. Ltd., China, and TSPA was purchased from Nanjing Capatue Chemical Co., Ltd., China, without further purification. This study used stable iodide (I-127) as a surrogate for radioactive iodide (I-129).

2.2 Preparation of TSPA-AgNO3.

We added 1.7 g (0.01 mol) of $AgNO_3$ into 7.5 mL (0.02 mol) of TSPA, and soon afterwards the liquid became dark brown. Then the mixture was sealed up and shaken for 48 hours to ensure that the $AgNO_3$ was adequately dissolved into the TSPA at ambient temperature. Afterwards, the solution was mixed into 30 mL of distilled water under

magnetic stirring until gelation. After 24 hours of aging, the gel composite was dried at 40 ˚C in an oven. It was ground up and repeatedly washed with distilled water until the washing liquid tested negative for $AgNO_3$ due to precipitation of AgCl. It was then dried again. Finally, the TSPA-$AgNO_3$ was pulverized using a mortar and pestle to isolate 20-60 mesh sized particles, which were stored in a brown sample bottle until the adsorption experiments.

2.3 Analytical methods.

Ultraviolet visible spectrophotometry (TU-1810, Beijing Purkinje General Instrument Co., Ltd.) determined the iodide ion concentration in the aqueous solution at the absorption wavelength of iodide aqueous solutions (226 nm; 220. 0 and 231.5 nm) [15,16]. The pH of point of zero charge (pH_{PZC}) for TSPA-$AgNO_3$ was detected using the pH equilibrium method as described in the reference work [17]. Solution pH was measured with a Sartorius PB-10 pH meter. Fourier Transform Infrared Spectrometry (FTIR) spectra of the TSPA gel and TSPA-$AgNO_3$ were obtained to analyze the coordination between silver ions and –NH– groups in TSPA. In addition, X-ray diffraction (XRD) and scanning electron microscopy (SEM) analyses were conducted to characterize the TSPA-$AgNO_3$ before and after adsorption.

2.4 Batch adsorption experiments.

Batch adsorption experiments were conducted as follows. Approximately 0.1 g of TSPA-$AgNO_3$ was placed into 100 mL iodide solutions at different initial pH levels (1, 3, 5, 7, 9, 11), temperatures (25, 35, 45, 55 ^{o}C) and coexisting NaCl concentrations (0, 1, 2, 3, 4 5 mol/L). The solutions were in a water bath with a constant shaking speed of 170 rpm. Solution pH was adjusted using HCl and NaOH solutions. The adsorption quantity of iodide onto TSPA-$AgNO_3$ is calculated using Eq. 1:

$$q_t = \frac{V(C_0 - C_t)}{m} \quad . \tag{1}$$

In Eq.1, q_t is the adsorption quantity of iodide onto the adsorbent (mmol/g) at time t, V is the volume of the solution (L), and m is the mass of the adsorbent (g). C_0 and C_t represent iodide concentrations in the solution (mmol/L) at initial and time t, respectively. When equilibrium iodide concentration C_e is substituted for C_t, Formula 1 yields equilibrium adsorption quantity, q_e (mmol/g).

3 Results and Discussion

3.1 Preparation of TSPA-AgNO3 composite adsorbent.

One secondary amine group lies in the middle with a trimethoxysilylpropyl group on either end of TSPA. In light of previous research on complexation between Ag^+ and amino-containing compounds [18-21], we inferred that $AgNO_3$ could dissolve into TSPA. Furthermore, the FTIR analyses of TSPA gel and TSPA-$AgNO_3$ gel displayed in Fig. 1 also evidence the formation of the Ag^+- amine complex. Various changes occurred with the transformation of TSPA gel to TSPA-$AgNO_3$ gel, including the following: (1) Similar to the results of previous work [22], N-H stretching vibration and intermolecular hydrogen bonding stretching vibration at 3500-3100 cm^{-1} split, N-H stretching vibration receded to 3240 cm^{-1}, and N-H bending vibration at 1545 cm^{-1} disappeared; (2) C-H stretching vibration at 2858

cm^{-1} strengthened; and (3) A characteristic absorption peak of NO_3^- at 1385 cm^{-1} appeared. These results show the weakening of N-H bonds and the slight strengthening of C–H bonds in TSPA-$AgNO_3$ gel compared to TSPA gel, confirming the formation of complexing action between Ag^+ and –NH– groups.

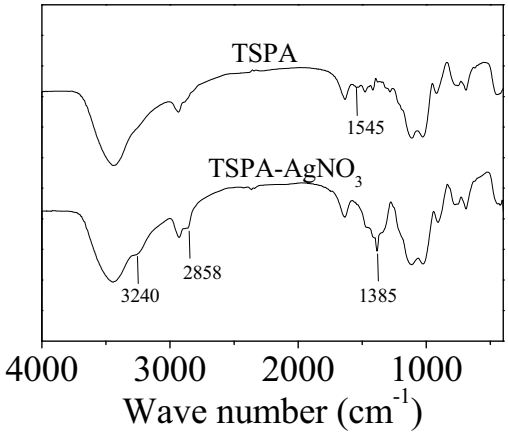


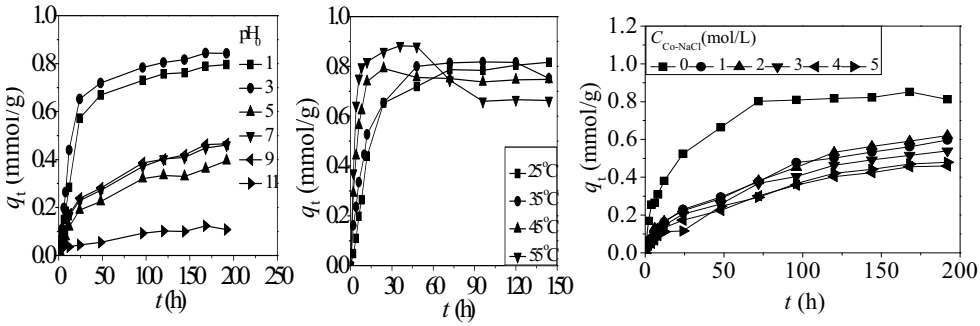
Fig. 1 FTIR spectra of TSPA and TSPA-$AgNO_3$

3.2 Effect of solution initial pH.

Fig. 2A displays the effect the solution pH exerts on the adsorption of iodide ions onto TSPA-$AgNO_3$. The solution pH remarkably affects both the adsorption rate and the adsorption quantity. The adsorption rate and quantity decline sharply as pH increases from 1 to 11. When pH is between 1 and 3, the equilibrium adsorption quantity reaches about 0.8 mmol/g, but this figure is 0.4 mmol/g when pH is between 5 and 9, and it is only 0.1 mmol/g when pH is 11. Furthermore, according to the slope of the adsorption curve, the average adsorption rate in the first 24 hours for strong acidic solutions was higher than that in strong basic solutions. The pH_{PZC} of TSPA-$AgNO_3$ is 9.5, and TSPA-$AgNO_3$ consumes H^+ in solution and changes the solution pH from 5-9 to 8-9.5. Therefore, when the solution pH is 11, the surface of TSPA-$AgNO_3$ is negatively charged and repels I^-; in contrast, a positively charged surface and plentiful H^+ in solution is likely to promote the adsorption reaction between iodide and TSPA-$AgNO_3$ when pH is between 1 and 3. Therefore, the pH range of 1 to 3 should be considered optimal for the adsorption of iodide onto TSPA-$AgNO_3$.

3.3 Effect of temperature.

Our study also observed the effect of temperature on adsorption in acidic solutions (pH_0 = 3). As Fig. 2B shows, the average adsorption rate in the first 24 hours increases as the temperature rises from 25 ^oC to 55 ^oC. However, the adsorption quantity over 144 hours, which is considered to approximate the thermodynamic equilibrium adsorption quantity, declines from 0.82 mmol/g at 25 ^oC to 0.66 mmol/g at 55 ^oC. It appears that raising the temperature promotes the diffusion of ions and molecules in aqueous solutions, and consequently accelerates adsorption reactions. The difference between approximate equilibrium adsorption quantities at different temperatures probably results from the fact that adsorption products in acidic solutions, in this case silver iodide, tend to be more unstable at higher temperatures. Given the efficiency of the batch adsorption operation, we suggest that temperature and adsorption time be regulated at 25-35 ^oC and 72 hours, respectively.



A B C

Fig. 2 Effect of initial solution pH (A), temperature (B) and coexisting NaCl (C) on the adsorption of iodide onto TSPA-AgNO3

3.4 Effect of coexisting NaCl.

In most actual systems, high Cl^- content is a main interfering factor for adsorbing and separating iodide because of their similar properties. Therefore, the adsorption experiments with different concentrations of coexisting NaCl (0, 1, 2, 3, 4, 5 mol/L) were conducted at 25 ^oC in acidic solutions (pH= 3). The results displayed in Fig. 2C indicate that coexisting NaCl inhibits the adsorption of iodide onto TSPA-$AgNO_3$, results that are in agreement with the previous related work [7]. The quantity adsorbed over 72 hours decreases sharply from 0.8 mmol/g to 0.3 mmol/g when the concentration of coexisting NaCl increases from 0 mol/L to 5 mol/L. It is possible that a higher adsorption quantity could be achieved if the time were extended; For example, 0.4 mmol/g might be adsorbed in a 5 mol/L coexisting NaCl solution after 144 hours.

3.5 Adsorption dynamic and thermodynamic models.

The experimental kinetic and equilibrium data at 25 ^oC shown in Figures 5, 6 and 7 were analyzed using the kinetic and thermodynamic models (Formulas 2-5) proposed previously [23-25].

Pseudo-first-order model: $$q_t = q_e - \frac{q_e}{e^{k_L t}} \quad (2)$$

Pseudo-second-order model: $$q_t = \frac{k_H q_e^{\,2} t}{1 + k_H q_e t} \quad (3)$$

Langmuir isotherm: $$\frac{C_e}{q_e} = \frac{1}{K_L Q_m} + \frac{C_e}{Q_m} \quad (4)$$

Freundlich isotherm: $$\ln q_e = \ln K_F + \frac{1}{n} \ln C_e \quad (5)$$

In these equations, q_t and q_e are the adsorption quantities at time t and at equilibrium, respectively; k_L and k_H are the adsorption rate constants for the pseudo-first-order and pseudo-second-order models, respectively; Q_m is the adsorption capacity; K_L is the Langmuir coefficient; $1/n$ is the heterogeneity factor of the adsorbent; and K_F is the Freundlich equation constant.

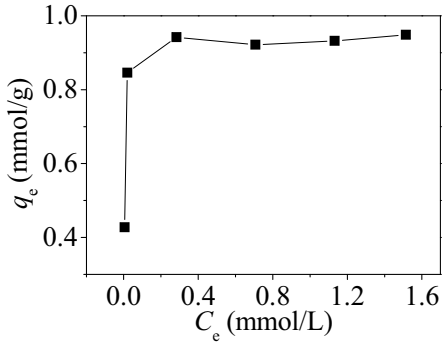
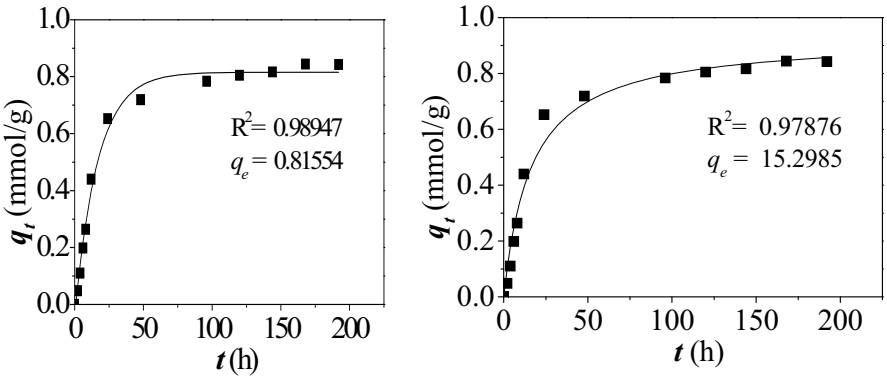
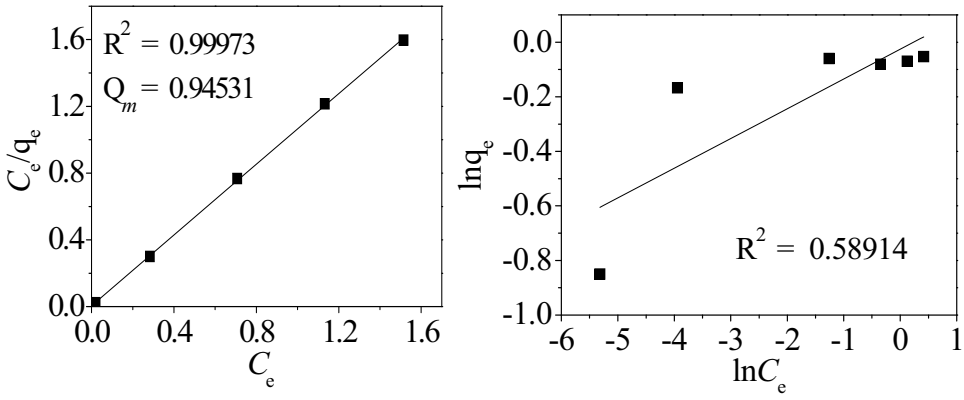


Fig. 3 Adsorption isotherm at 25 °C in acidic solution



(a) (b)

Fig. 4 Kinetic models for the adsorption of iodide onto TSPA-AgNO3 at 25 °C, (a) pseudo-first-order model; (b) pseudo-second-order model



(a) (b)

Fig. 5 Isotherm models for the adsorption of iodide onto TSPA-AgNO3 at 25 °C, (a) Langmuir model; (b) Freundlich model

Fig. 4 and Fig.5 show the fitting results for the kinetic and thermodynamic models. Based on the correlation coefficients and the degree of closeness between the calculated equilibrium adsorption quantities (0.8 mmol/g for pseudo-first-order and 15 mmol/g for

pseudo-second-order) and the experimental equilibrium adsorption quantity (0.8 mmol/g), the pseudo-first-order dynamic model and Langmuir isotherm model appear to describe and predict accurately the adsorption behaviors of iodide onto TSPA-$AgNO_3$ in strong acidic solutions (pH=1-3). The pseudo-first-order dynamic model lumps together all steps of the adsorption process, including external diffusion, internal diffusion, and adsorption [26]. Therefore, the overall adsorption process of iodide onto TSPA-$AgNO_3$ appears similar to a first-order reaction. The Langmuir model assumes that the solute is adsorbed onto a finite number of binding sites with the same energy levels found on a homogenous surface, and that the adsorption occurs on a monolayer surface with no interaction between adsorbed molecules [27].

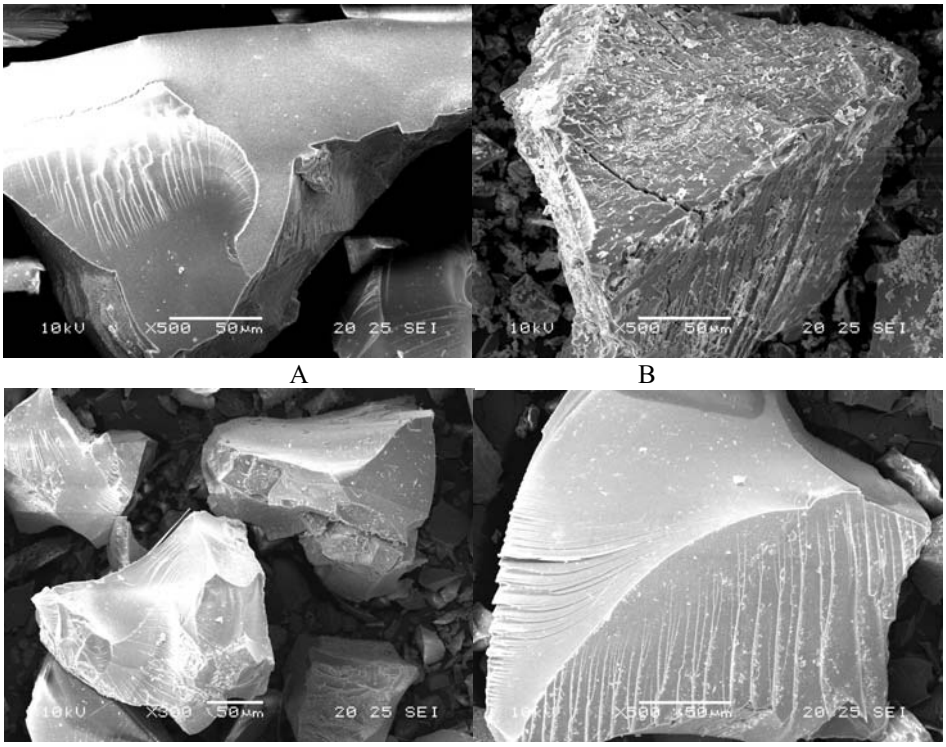


Fig. 6 SEM images of TSPA-AgNO3 before (A) and after adsorption (pH=3 were B; pH = 7 were C; pH= 11 were D)

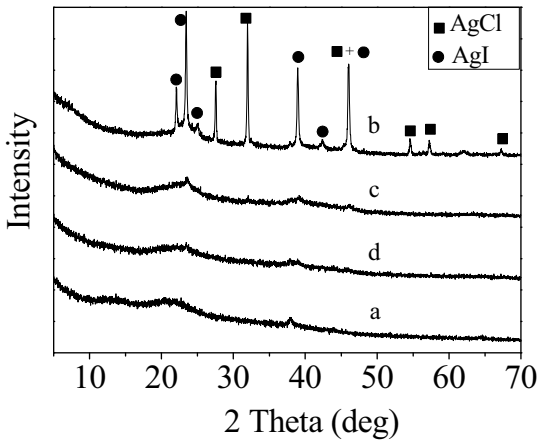


Fig. 7 XRD of TSPA-AgNO3 before (a) and after adsorption (pH = 3 was b; pH = 7 was c; pH = 11 was d)

4 Adsorption mechanism.

SEM and XRD analysis were performed for TSPA-$AgNO_3$ before and after adsorption to explore further the adsorption behavior of iodide ions. Figures 8 and 9 display SEM and XRD analysis results, which can be summarized as follows: (1) TSPA-$AgNO_3$ is uniform, compact and amorphous before adsorption, and no crystal $AgNO_3$ exists in TSPA-$AgNO_3$ adsorbent, further illustrating the formation of Ag^+- amine coordination. (2) Many silver iodide and silver chloride agglomerates occur over the matrix after adsorption in strong acidic solutions adjusted using HCl. Furthermore, nitrate ions infiltrate into the solution when iodide ions are adsorbed onto TSPA-$AgNO_3$.

Based on the effects of solution pH and coexisting NaCl, as well as the analysis results listed above, we conclude that the adsorption of iodide ions onto TSPA-$AgNO_3$ involves two processes: (1) NO_3^-, the counterbalance ion of the Ag^+- amine coordination complex, is exchanged with anions such as I^-, Cl^-, or OH^- in solution. (2) In strong acidic solutions (pH=1-3), the interaction of the coexisting H^+ and the secondary amine group in the matrix results in the weakening of the Ag^+- amine coordination, consequently promoting the reaction between Ag^+ and the precipitating agent like I^- or Cl^- that diffuses into the gel matrix to form AgI or AgCl. Therefore, the immobilization of iodide using TSPA-$AgNO_3$ in acidic aqueous solutions is closely related to the precipitation of silver iodide on the surface of TSPA-$AgNO_3$ (see Eq. 6). In neutral or slightly alkaline solutions, NO_3^- is exchanged with I^- as the counterbalance ion of the Ag^+- amine coordination complex (Eq. 7).

$$[\text{TSPA-Ag}]^+NO_3^- + I^- \overset{H^+}{=} \text{TSPA-AgI} + NO_3^- \tag{6}$$

$$[\text{TSPA-Ag}]^+NO_3^- + I^- = [\text{TSPA-Ag}]^+I^- + NO_3^- \tag{7}$$

5 Summary

Silver-coordinated bis(trimethoxysilylpropyl)amine polycondensate (TSPA-$AgNO_3$) was prepared and used to adsorb iodide ions in aqueous solutions. The main conclusions of these experiments are as follows:

(1) Silver nitrate can dissolve into TSPA, forming a uniform dark brown TSPA-$AgNO_3$ complex.

(2) TSPA-$AgNO_3$ is easy to prepare and remarkably efficient in adsorbing iodide ions in water, especially in acidic solutions. The adsorption capacity of iodide ions onto TSPA-$AgNO_3$ is nearly 0.9 mmol/g when the solution pH is between 1 and 3, and it declines to 0.1 mmol/g as the solution pH increases to 11. Furthermore, increased temperature can accelerate the adsorption rate. However, coexisting NaCl can inhibit adsorption, and the quantity adsorbed over 72 hours decreases from 0.8 mmol/g to 0.3 mmol/g when the concentration of coexisting NaCl increases from 0 mol/L to 5 mol/L. The pseudo-first-order dynamic model and Langmuir isotherm model well describe and predict the adsorption behaviors of iodide onto TSPA-$AgNO_3$ in acidic solutions (pH=1-3).

Acknowledgements

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