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Effect of Initial Arsenic Concentration on Sediment Adsorption of Arsenic

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ABSTRACT

Aiming at exploring the effect of initial arsenic concentration on sediment adsorption, at first, an orthogonal experiment of four factors and four levels was carried out to determine the optimal experimental conditions. Based on orthogonal experiments, the single factor experiments of different sampling times under the optimal experimental conditions were conducted to explore the influence of the initial arsenic concentration on adsorption of sediment of different concentrations. To confirm the arsenic's adsorption equilibrium time, the experiments with different sediment concentrations were conducted under the optimal experimental conditions, when sampling times respectively, were 5, 10, 15, 30, 45, 60, 90 and 120 mins. The results showed that (1) the optimal conditions for the sediment adsorption of arsenic: temperature is 40°C, vibration velocity is 250 r·min^{-1}, pH is 9, and sediment diameter ranges from 0.15 to 0.3 mm; (2) when the sampling time and the sediment concentration are constant, with the increase of initial arsenic concentration, the amount of the unit sediment adsorption increases gradually; when the concentration of the initial arsenic and sediment is constant, adsorption capacity of per unit mass of the sediment increases with time; (3) the time of arsenic adsorption equilibrium from sediment is 60 mins.

INTRODUCTION

At present, arsenic (As) contamination of water bodies has posed a serious problem internationally (Baig et al. 2013, Das et al. 2013, Freikowski et al. 2013). Arsenic contamination in drinking water sources threatens the health of some 200 million people worldwide (Baig et al. 2013). In natural water, As exists in inorganic forms with the oxidation states of -3, 0, +3 and +5 (Smedley et al. 2002). Predominantly, the species arsenic [As (III)] and arsenate [As (V)] exist in ground and surface water respectively. It is reported that As (III) is more toxic to biological systems than As (V) (Ferguson & Gavis 1972). As (III) is a well known water contaminant that causes toxicological and carcinogenic effects (Shipley et al. 2010). It is very essential, therefore, to remove As from water (Nitzsche et al. 2015).

Precipitation of authigenic minerals with As as a constituent ion is unlikely, as most As minerals are too soluble to precipitate under oxic condition (Wagemann 1978, Webster 1990, Bowell 1994). The sediment is the important carrier of the transportation and transformation to the main restricted factors in the eutrophic water bodies, such as heavy metals. Adsorption is a common physico-chemical process used to remove heavy metals (Sounthararajah et al. 2015). It plays a very important role in the transport and fate of pollutants, especially in sediment (Aksentijeviæ et al. 2012).

Unlike authigenic mineral precipitation, sorption or desorption may be an important process in controlling the dissolved As concentrations in sediment.

The previous researches about the adsorption of sediment have mostly been focused on Cr and Cu pollution (Jin et al. 2009, Renkou et al. 2009, Shipley et al. 2010, Terbouche et al. 2011, XIA et al. 2011, Zhang et al. 2011, Seco-Reigosa et al. 2013), while few studies were concentrated on As pollution. The main factors that control the adsorption of As in water and solid phases remain unclear. The aims of the present work were to investigate the effect of initial As concentration on sediment adsorption and make sure the As solution's adsorption equilibrium time.

MATERIALS AND METHODS

Materials: The experimental materials are the sediments taken from shallow water near the bank of the Yellow River at Huayuankou during the process of natural air drying. The dried sands were crushed by the pulverizer in 2 ~ 3 mins and got through 325 mesh sample sieve to obtain sediment with the size of 0.0375~0.088 mm. The sands were baked for 6 hours at 105°C and cooled to the room temperature in a dryer. The sediment composed of 1% organic matter and 10% CaCO$_3$. The pH of the sediment was 7.9~8.3.

Methods: Atomic fluorescence spectrophotometer (model

8220) was used to determine the concentration of As (III). As (III) (GSB 04-1714-2004) was purchased from the market. Quantification of As (III) was based upon the calibration curves of standard solutions of As (III) ion. The equation of calibration curve was $y=44.268x-33.533$, and the detection limit of As (III) was 0.01 mg·L^{-1}. The correlation coefficient was approximately 0.9998. The relative standard deviation of three replicates was always below 1%. At the same time, four water samples were selected, which were set as four parallel samples respectively, in order to run the addition and recovery experiment. The results indicated that the recovery rate was in the range of 97.489% to 105.303%. The accuracy of the method was good and satisfied the accuracy requirement of the analysis method. An Orion-Ross combination glass electrode was used to measure the pH of the solution. The glass electrode was calibrated at 25°C using 4, 7, and 10 pH buffers.

The As (III) removal from the water was calculated as:

As (III) removal rate (%) = $100\% (C_0 - C_t)/C_0$

and the adsorptive uptake of As (III) by sediment (g·L^{-1}) (mg·g^{-1}), was calculated as

$qt = (C_0 - C_t)V/W$

Where, C_0 is the initial As (III) concentration (mg·L^{-1}), C_t is the As (III) concentration (mg·L^{-1}) at any time, V is the volume of the solution (L) and W is the mass of the adsorbent (g).

RESULTS AND DISCUSSION

The determination of the optimum experimental condition: The preinstall initial concentration of As was 0.1 mg·L^{-1}, the concentration of sediment was 10 kg·m^{-3}, and the orthogonal experiment of four factors (temperature, velocity, pH and sediment particle size) and four levels was designed according to the orthogonal array as shown in Table 1.

The greater the removal rate is, the better the adsorption effect is. As can be seen from the Table 1, temperature factor column: K2>K4>K1>K3; vibration velocity factor column: K4>K3>K2>K1; pH column: K4>K1>K3>K2; sediment diameter: K4>K3>K2>K1. Therefore, the optimal conditions for the experiment: temperature of 40°C, vibration velocity of 250 r·min^{-1}, pH of 9, and sediment diameter of 0.15 ~ 0.3 mm.

The effects of initial concentration of As under the optimal experimental condition on the sediment adsorption of As: To explore the influence of initial As concentration at different sampling times (15, 30 and 60 mins) on different concentrations of sediment adsorption of As, under the optimal condition determined through the orthogonal experiments of sediment adsorption of As, the initial As concentration value series (0.1, 0.15, 0.2, 0.25, 0.3, 0.35 and 0.4

mg·L^{-1}) and sediment concentration series (5, 10, 15, 20 and 25 kg·m^{-3}) were selected to carry on the single factor experiment at the sampling times of 15, 30 and 60 mins.

Concrete experiment plan: The sediment of particle size of 0.15~0.3 mm was placed as an adsorbent in 5 numbered volumetric flasks of 500 mL, in which the dry masses of the sediment were respectively 2.5, 5, 7.5, 10, and 12.5 g, which were added with water to the scale line, formulating as the sediment solutions with different concentrations of 5, 10, 15, 20 and 25 kg·m^{-3}. 0.5 mL of the As standard solution of 100 mg·L^{-1} was added in the volumetric flasks respectively. The value of pH was adjusted to 9. Another five volumetric flasks were taken with identical concentration sediment solutions and pH, but without As standard solution as the blank samples. The ten volumetric flasks were sealed with paraffin and placed in the whole temperature vibrator at vibration velocity of 250 r/min and 40°C for 15, 30 and 60 mins. Samples (10 mL) were taken from the vessel and filtered through a 0.45 μm Nalgene SCFA filter. The above method successively changed the concentration of the As solution to 0.15, 0.2, 0.25, 0.3, 0.35 and 0.4 (mg·L^{-1}), and sampling times were 15, 30 and 60 minutes.

Hydride atomic fluorescence spectrophotometry was used to determine the concentration of As in the water samples and calculate adsorption of per unit mass of the sediment to obtain change curves of adsorption capacity of per unit mass of the sediment with the initial concentration of As at the sampling time of 15 mins (Fig. 1a), 30 mins (Fig. 1b) and 60 mins (Fig. 1c) in the sediment solution of different concentrations.

As can be seen from the Fig. 1a, under the same sediment concentration, adsorption capacity of per unit mass of the sediment changed with the initial concentration of As. When the sediment concentration was 5 kg·m^{-3} and the initial concentration of As changed from 0.1 mg·L^{-1} to 0.4 mg·L^{-1}, the adsorption capacity of per unit mass of the sediment increased gradually from 0.010 mg·g^{-1} to 0.042 mg·g^{-1}, increased by 3.2 times; when the concentrations of sediment were 10, 15, 20 and 25 kg·m^{-3}, the adsorption capacity of per unit mass of the sediment increased by 3.3, 3.7, 3.2 and 3.1 times respectively.

As can be seen from the Fig. 1b, under the same sediment concentration, adsorption capacity of per unit mass of the sediment changed with the initial concentration of As. When the sediment concentration was 5 kg·m^{-3} and the initial concentration of As changed from 0.1 mg·L^{-1} to 0.4 mg·L^{-1}, adsorption capacity of per unit mass of the sediment increased gradually from 0.010 mg·g^{-1} to 0.043 mg·g^{-1}, increased by 3.3 times; when the concentrations of sediment were 10, 15, 20 and 25 kg·m^{-3}, the adsorption capacity of per unit mass

Table 1: The analysis of the orthogonal experiment of four factors and four levels of temperature, vibration velocity, pH and sediment particle size.

Number	Temperature (°C)	pH	Vibration velocity (r·min^{-1})	Sediment diameter (mm)	As removal rate (%)
1	20	6	100	<0.088	40.813
2	20	8	0	0.088~0.15	51.648
3	20	7	200	>0.3	69.968
4	20	9	250	0.15~0.3	71.748
5	40	6	0	>0.3	73.571
6	40	8	100	0.15~0.3	73.157
7	40	7	250	<0.088	79.829
8	40	9	200	0.088~0.15	86.858
9	10	6	200	0.15~0.3	54.415
10	10	8	250	>0.3	58.192
11	10	7	100	0.088~0.15	38.369
12	10	9	0	<0.088	53.207
13	30	6	250	0.088~0.15	84.538
14	30	8	200	<0.088	61.761
15	30	7	0	0.15~0.3	68.116
16	30	9	100	>0.3	64.424
K1	234.177	253.34	216.763	235.61	
K2	313.415	244.76	246.542	261.413	
K3	204.183	256.28	273.002	266.155	
K4	278.839	276.24	294.307	267.436	
k1	58.54425	63.334	54.19075	58.9025	
k2	78.35375	61.19	61.6355	65.35325	
k3	51.04575	64.071	68.2505	66.53875	
k4	69.70975	69.059	73.57675	66.859	
Range R	27.308	7.8698	19.386	7.9565	

of the sediment increased by 3.3, 3.6, 3.3 and 3.2 times respectively.

As can be seen from the Fig. 1c, under the same sediment concentration, adsorption capacity of per unit mass of the sediment changed with the initial concentration of As. When the sediment concentration was 5 kg·m^{-3} and the initial concentration of As changed from 0.1 mg·L^{-1} to 0.4 mg·L^{-1}, the adsorption capacity of per unit mass of the sediment increased gradually from 0.013 mg·g^{-1} to 0.052 mg·g^{-1}, increased by 3.0 times; when the concentrations of sediment were 10, 15, 20 and 25 kg·m^{-3}, the adsorption capacity of per unit mass of the sediment increased by 3.3, 3.1, 3.3 and 3.2 times respectively.

As can be seen from the Fig. 2, when the concentration of the initial As and sediment was constant, adsorption capacity of per unit mass of the sediment increased with time; when the initial concentration of As changed from 0.1 to 0.35 mg·L^{-1}, the difference among adsorption capacity of per unit mass of the sediment of the three sampling times increased gradually; when the concentration of As was 0.4 mg·L^{-1}, it decreased.

This is mainly because that the adsorption of sediment on heavy metal As is a physical and chemical reaction oc-

curring in the solid-liquid two-phase interface. No matter it is the physical or chemical adsorption, it can be understood that there are adsorption sites on the surface of the solid phase. When the concentration of sediment is constant, the number of adsorption sites must remain. When the initial As concentrations in the water samples increases, the amount of adsorbate around the adsorption sites increases, and the probability of As aggregation in sediment surface also increases, and at the same time it also ensures the adsorption sites have enough concentration of As for adsorption. That is, with the increase of the initial concentration of As, the unit sediment adsorption is also changing.

The adsorption of sediment on heavy metal is a gradual process. There is an adsorption equilibrium time in the variety of adsorption process. Within the adsorption saturation time, with the increase of the sampling time, there is more As accumulation on the surface of sediment, and the physical and chemical adsorption becomes more and the amount of sediment adsorption also increases. This is consistent with the general study of adsorption. At a low initial concentration of As, the effect of sampling time on the unit sediment adsorption is not obvious, this is because the adsorption process of the low concentration of As has been largely completed in a relatively short period of time. There-
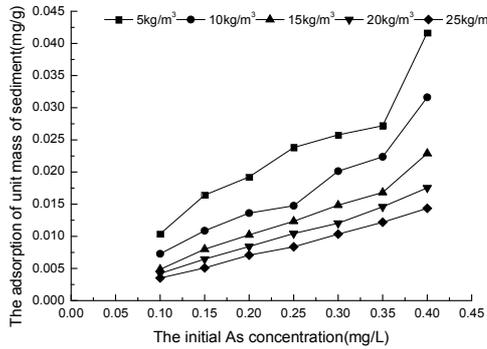
(1a) the sampling times is 15 mins
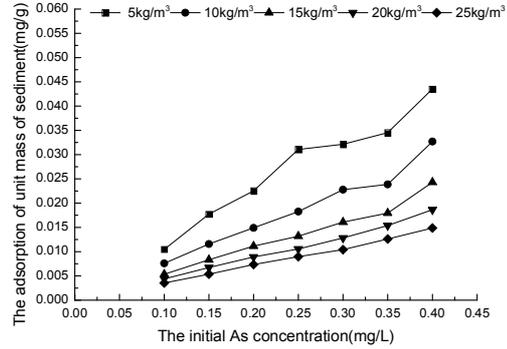
(1b) the sampling times is 30 mins
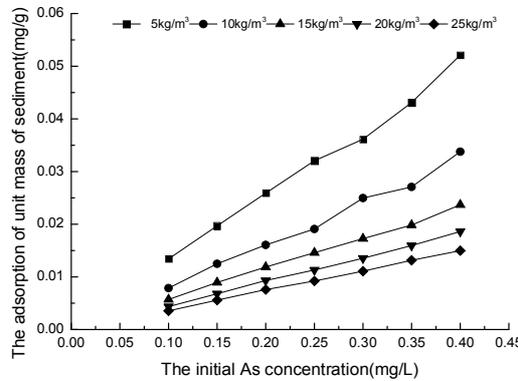
(1c) the sampling times is 60 mins

Fig. 1: The influence of initial As concentration on the unit sediment adsorption of different sediment concentrations when the sampling times are 15, 30 and 60 mins.
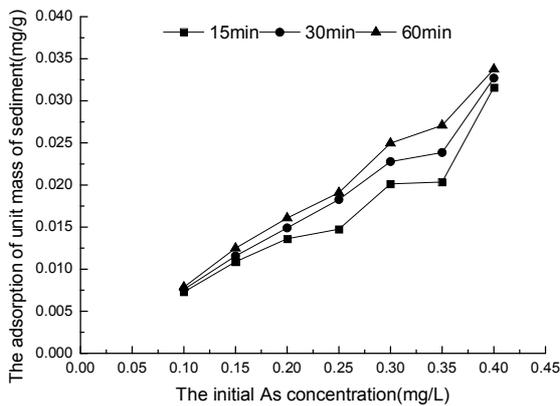

Fig. 2: The variation of adsorption capacity of per unit mass of the sediment of 10 kg·m^{-3} with initial As concentration under different sampling times.

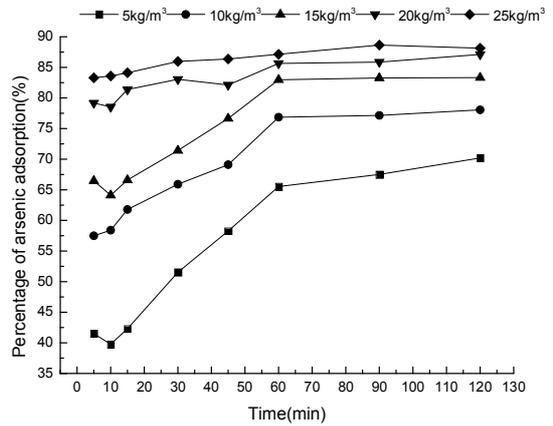

Fig. 3: The As removal rate changes with time under different sediment concentrations when the concentration of As is 0.1 mg/L.

fore as the concentration of As increased, the difference among the amount of sediment adsorption of the unit sediment adsorption in the three sampling times becomes larger. When the initial concentration of As as 0.4 mg·L^{-1}, the As

concentration is too high, which results in the low probability of the contact of a part of As and sediment. So the effect of the sampling time on the unit sediment adsorption becomes lesser.

Adsorption equilibrium time: To make sure of the adsorption equilibrium time of As solution, the experiments were conducted under the condition of 25°C, pH of 9.00±0.04, vibration velocity of 250 r·min^{-1}, and size of sediment of 0.15~0.3 mm. Sorbent doses of 5 kg·m^{-3}, 10 kg·m^{-3}, 15 kg·m^{-3}, 20 kg·m^{-3} and 25 kg·m^{-3} of sediment concentrations were prepared respectively. Typically, samples were collected at 5, 10, 15, 30, 45, 60, 90 and 120 mins. The As removal rate curve of different sediment concentrations was obtained and shown in Fig. 3.

The Fig. 3 indicated that the percentage of As adsorption did not appreciably change with the change of time from 5 to 10 mins. This instability of As adsorption potentially could be related to the fact that the reaction which was in an adaptation period was just beginning. When the time rose from 10 to 60 mins, the percentage of As adsorption of different sediment concentrations increased with time. Whereas in 60 to 120 mins, the As adsorption rate was approximately stable. This result showed that As adsorption equilibrium time from sediment was 60 mins.

CONCLUSIONS

1. The results of the orthogonal experiment of four factors (temperature, vibration velocity, pH and sediment particle size) and four levels demonstrate that when the initial concentration of As is 0.1mg·L^{-1} and the concentration of sediment is 10 kg·m^{-3}, the optimal condition for the sediment adsorption of As is that temperature is 40°C, vibration velocity is 250 r·min^{-1}, pH is 9, and sediment diameter is 0.15 ~ 0.3 mm.

2. The influence of initial As concentration at different sampling times on adsorption of heavy metal As can be drawn: when the sampling time and the sediment concentration are constant, with the increase of initial As concentration, adsorption capacity of per unit mass of the sediment increases gradually; when the concentration of the initial As and sediment are constant, adsorption capacity of per unit mass of the sediment increases with time; when the initial concentration of As changes from 0.1 to 0.35 mg·L^{-1}, the difference among adsorption capacity of per unit mass of the sediment of the three sampling times increases gradually; when the concentration of As is 0.4 mg·L^{-1}, it decreases.

3. When the time changes from 10 to 60 mins, the percentage of As adsorption of different sediment concentrations increases with time. Whereas in 60 to 120 mins, the As adsorption rate is approximately stable. This result shows that As adsorption equilibrium time from sediment is 60 mins.

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