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Kinetic study on the adsorption of arsenic in the Yellow River sediment with different gradation

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ABSTRACT

The aim of this article is to understand the adsorption mechanism and adsorption kinetics law of Yellow River sediment on arsenic with different gradations. Adsorbent particles with different sizes and states were tested in order to acquire better insight into the adsorption process. Different kinetic models, such as the first order kinetic model, the two-level kinetic model and the particle diffusion model, were used to fit the experimental results. The correlation coefficient of the silver sand were all above 0.9200 in the first kinetic fitting curves and that of the coarse sand were all above 0.9900 in the two-level kinetic fitting curves. Both of the first order kinetic and the two-level kinetics model fitting results indicated that silver sand and the coarse sand had good correlation. The mass transfer kinetic model fitting results indicated that the adsorption reaction was mainly controlled by intra particle diffusion. By fitting the experimental results of silver sand in static and turbulent conditions, concluded that the average values of correlation coefficient were 0.9600 and 0.9368 in static and turbulent conditions respectively, which manifested that under static conditions had better correlation, at the same time, because the value of c was not equal to 0, the adsorption reaction was mainly controlled by intra particle diffusion, liquid film diffusion and surface adsorption.

Keywords: Yellow River sediment; Gradation; Adsorption; Kinetics

1. Introduction

Nowadays, the discharge of heavy metal ions in natural water by industrial activities is increasing, which is one of the most serious environmental problems even at very low concentrations [1]. Arsenic is a kind of metalloid, but the toxicity of arsenic ions in aqueous solution is very tragic, which is equal to the heavy metal. So far, there are many methods of removing arsenic ions and other heavy metal ions from water, such as reverse osmosis, electrodialysis, electrochemical technology and so on [2–7]. However, these methods have many shortcomings, such as high material consumption, chemical residues. In order to solve this problem, at the same time to easily take the material and to realize innovation, this paper mainly studies the adsorption of arsenic by the Yellow River sediment. As the muddiest river in the world, the Yellow River is a significant water source in the northwest and north of China [8]. In recent years, the pollution of the Yellow River appeared with a rapidly expanding economy of Yellow River basin, and arsenic pollution [9,10] has became a big problem on water safety of Yellow River, therefore arsenic pollution control and management of water resources were already a research focus [11,12]. The research of adsorption mechanism and adsorption kinetics law of Yellow River sediment on arsenic will provide a theoretical basis for the pollution control and the management of water resources of Yellow River.

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There are so many papers already published on the removal of pollutant using different adsorbents, such as activated lignin–chitosan extruded blends, agricultural waste-based activated carbon, novel guar gum/Al_2O_3 nanocomposite, ZrO_2/Fe_3O_4/chitosan nanomaterials and so on [13–18]. However, the study on the adsorption of arsenic by sediment is rare. In this study, the experimental materials can be easily obtained and then we can use the existing materials effectively to study the adsorption of arsenic. The Yellow River sediment particles have large specific surface area. There are many kinds of active substances on the particle surface, which can combine with heavy metal (including metalloid) pollutants [19]. As an effective carrier for the migration, transformation and diffusion of heavy metals in water, sediment affects heavy metal pollutants valence state, transformation and water environment capacity. The contaminants adsorption of sediments can significantly affect the bioavailability, toxicity and mobility of contaminants [20]. The sediment adsorption changes with hydrodynamic conditions. Therefore, the study of the water and sediment system of heavy metals will be conducive to understanding the characteristics and rules of migration and transformation of pollutants in water and sediment.

Equilibrium experimentation and modeling, as well as kinetic studies are widely used to study the adsorption process [21]. Adsorption kinetics model study is an important way to understand the mechanism of the adsorption process and the potential rate limiting step, which can be used to express the process of chemical reaction, diffusion and mass energy transfer [22]. The adsorption kinetics, such as the adsorption dynamic equilibrium and adsorption rate measurements, can theoretically illustrate the relationship between adsorption capacity and adsorption time [23]. The first order kinetics model, the two-level dynamic model and the mass transfer kinetics model are widely used in dynamic models study of adsorption process [24]. In this paper, the arsenic adsorption of the Yellow River sediment was studied with dynamic fitting of the arsenic adsorption in different gradation of the sediment.

2. Materials and methods

2.1. Materials

The sand used in this study was collected from the river shoal near Huayuankou section of the Yellow River. The sampling uniformity of the sediment was taken into account during the sampling process. After naturally drying in the absence of light, the impurities were removed and then the sediment was divided into coarse sand, medium sand and silver sand according to size distribution. The medium sand consisted of coarse sand and silver sand with the quality ratio of 1:1. The grain size of coarse sand and silver sand was observed and analyzed by photoelectric particle size analyze, the median particle diameters of silver sand, medium sand and coarse sand were 0.01, 0.017 and 0.036 mm, respectively. The detail is listed in Table 1.

2.2. Methods

According to the sediment gradation requirements (coarse sand, medium sand and silver sand) and concentration (1, 5 and 10 kg/m^3 requirements), dry sand was put into 500 mL conical flask, then activated with high purity water 24 h. Water samples of different concentrations prepared with arsenic reserves were added to conical flask, so that the total volume can be 1 L. The adsorption experiments were carried out in the static and turbulent conditions. When saturated adsorption was reached, the sample was filtered with 0.45 μm ultrafiltration membrane to remove the adsorption interference of colloid and organic macromolecules, and then 5.00 mL of filtrate was used for arsenic concentration measurement with standard method.

In order to better understand the surface characteristics of sand, scanning the washed sand by using scanning electron microscope (SEM). To further reveal the mechanism of the adsorption, different kinetic models were used to fit the experimental results, which were shown below.

3. Results and discussion

3.1. Characterization of the sand

3.1.1. Characterization of microstructure by SEM

The surface characterization of sediment particles is important to the arsenic adsorption in aqueous phase. As shown in Fig. 1, the morphologies of the cleaned sand and the sand after adsorption equilibrium were characterized by SEM. The surface of the cleaned sand particle was rough and irregular, there were some fine flakes or blocks attach to the depression or flat place of the particle. And the surface texture of cleaned sand was clear and complex, and there were a lot of pores on the particles. This feature was due to the composition of the sediment and the forces acting during migration. After adsorption, the surface of the sand became smooth and fine, with fractured pores and small spherical particles. Because there were varieties of organic and inorganic pollutants, the different pollutants and the components of sediment itself affected each other in the process of adsorption, and there were constant migration and transformation under the water flow, which made the surface morphologies of sediment particles became more complex and changeable.

Table 1
Grain size analysis of sediment used in the experiments

Sample	The percentage of particles smaller than a certain size ϕ (%)									Median particle size (μm)	Average particle size (μm)
	Particle size (μm)										
	2	8	16	25	50	75	100	250	1,000		
Silver sand	12.62	43.26	64.27	76.84	91.93	97.11	99.03	100.00	100.00	10.02	17.75
Medium sand	8.85	31.13	47.81	61.21	84.46	93.83	97.21	99.26	100.00	17.32	28.25
Coarse sand	2.55	7.06	15.05	30.20	70.19	89.00	95.90	99.11	100.00	35.84	43.76

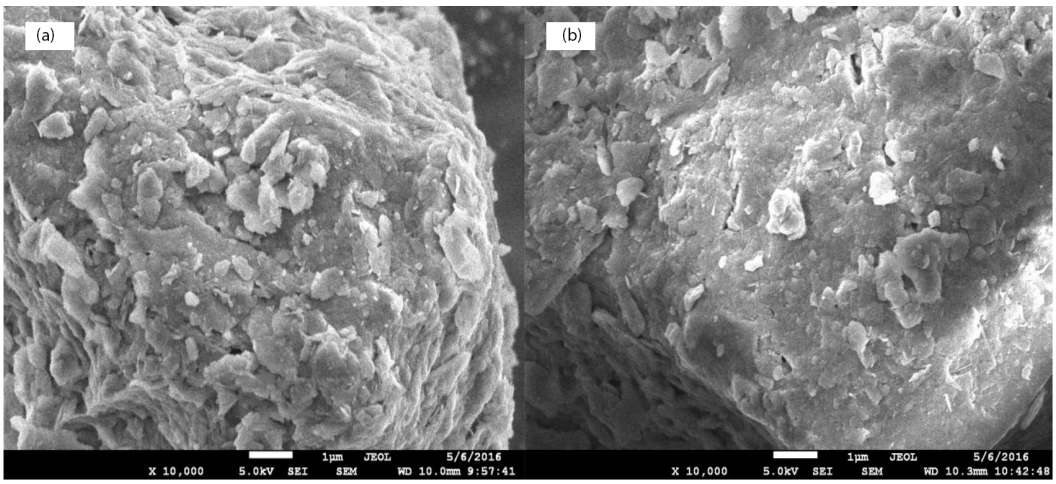
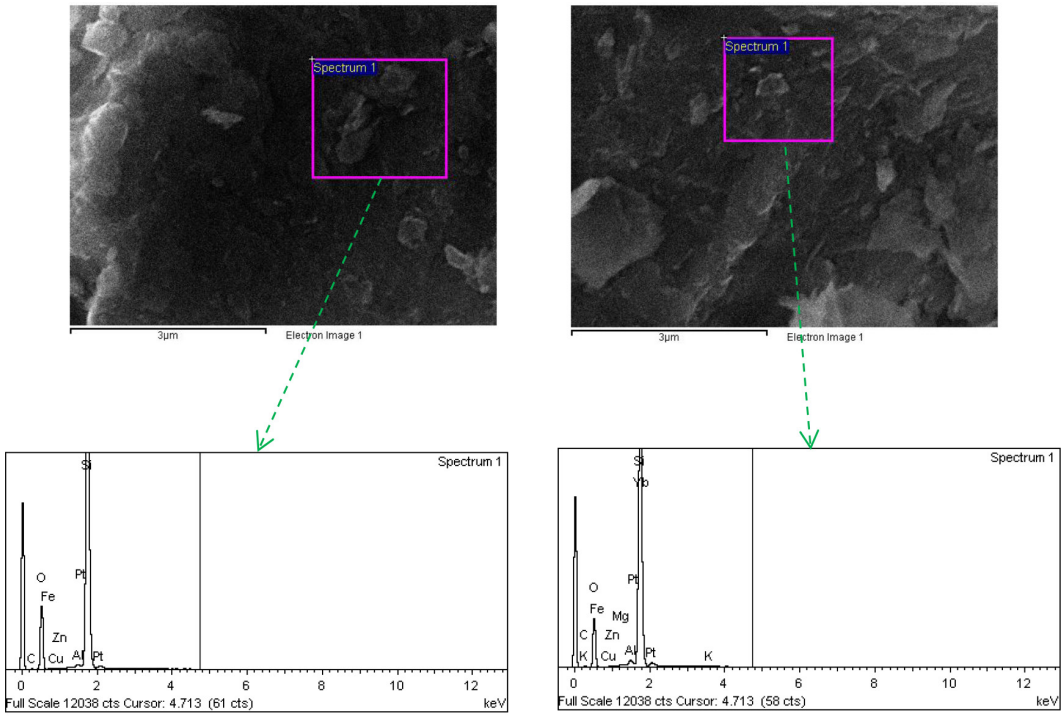


Fig. 1. SEM images of (a) cleaned sand and (b) sand after adsorption equilibrium.



Part 1 Part 2

Fig. 2. SEM-EDS images of cleaned sand.

3.1.2. Characterization of microstructure by SEM-EDS

SEM-EDS can be used to detect and analyze the local range of the particle surface and to detect the percentage of mass fraction (W) and the number of atoms (A) in the region. As seen from the Figs. 2 and 3, the main atoms in the particle detection area were C, O, Al and Si, in which the content of O and Si was higher. It was consistent with the conclusion that the sediment particles are mainly composed of SiO_2. It was obvious that the content of arsenic on the sediment surface increased after adsorption, which indicated that the adsorption capability of the sediment for arsenic was good.

The classical method to investigate the adsorption mechanism and kinetics law of adsorption process is kinetic model fitting with the adsorption results under different conditions. In this study, the experimental results of different sediment gradation were fitted with the first order kinetic model, two-level kinetic model and intra particle diffusion model to obtain the first order kinetic equation, two-level kinetic equation and intra particle equation.

3.2. First order kinetic model

The first order kinetic model is the kinetic equation of the relationship between the reaction rate and the concentration of the reactant in the system; the expression of which is as follows:

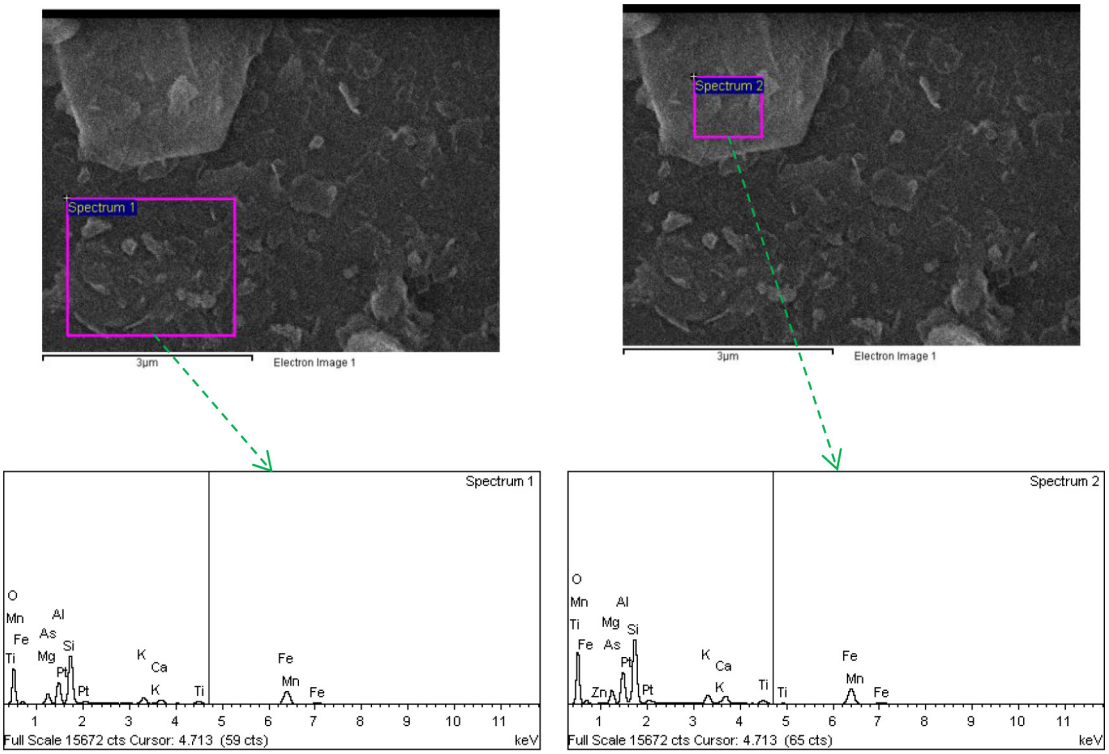


Fig. 3. SEM-EDS images of the sand after adsorption equilibrium.

$$\frac{dq_t}{dt} = k_1\left(q_e - q_t\right) \tag{1}$$

The boundary condition is when $t = 0$; $q_t = 0$; when $t = t$; $q = q_e$, the integration is taken over boundary conditions on both sides of Eq. (1), it becomes:

$$\ln(q_e - q_t) = \ln q_e - k_1 t \tag{2}$$

where q_e is the adsorption capacity of unit adsorbent at equilibrium condition (μg/g); q_t is the adsorption capacity of unit adsorbent at t moment (μg/g); k_1 is the kinetic adsorption rate constant for first order kinetics (g/μg·min^{-1}).

When we plot $\ln(q_e - q_t)$ vs. time and perform linear fitting, the slope of the fitting straight line provides the rate constant of the first order kinetic adsorption, and the intercept provides the equilibrium adsorption capacity. The fitting curves are shown in Fig. 4, and the corresponding first order kinetic parameters are shown in Table 2.

As seen in Table 2, the correlation coefficients of coarse sand were 0.7066, 0.7786 and 0.8374; the correlation coefficients of medium sand were 0.9198, 0.8481 and 0.9902; and the correlation coefficients of silver sand were 0.9624, 0.9769 and 0.9217 at the concentration of 1, 5 and 10 kg/m^3, respectively, which showed that the correlation coefficient obtained for the silver sand was better than the medium sand and coarse sand. The correlation coefficient generally increases with the increase of initial aqueous phase arsenic concentration.

The first order kinetic model cannot express the arsenic adsorption process well, because the difference between the equilibrium adsorption capacity q_e and the experimental results q_e* was relatively large, that is the value of q_e obtained in the adsorption experiment deviated from the theoretical model fitting value.

3.3. Two-level kinetic model

The two-level kinetic model is used to describe the divalent metal ions adsorption process, assuming that the adsorption is proportional to the amount of adsorbent on adsorption site; the expression of which is as follows:

$$\frac{dq_t}{dt} = k_2\left(q_e - q_t\right)^2 \tag{3}$$

The boundary condition is $t = 0$; $q_t = 0$; $t = t$; $q = q_e$, integration is taken over boundary conditions on both sides of Eq. (3), it can be obtained as:

$$\frac{t}{q_t} = \frac{1}{k_2 q_e^2} + \frac{t}{q_e} \tag{4}$$

where q_e is the adsorption capacity of unit adsorbent at equilibrium condition (μg/g); q_t is the adsorption capacity of unit adsorbent at t moment (μg/g); k_2 is the kinetic adsorption rate constant for two-level kinetics (g/μg·min^{-1}).

A plot of the two-level kinetic model can be obtained by taking the time as the abscissa and t/q_t as the ordinate, the equilibrium adsorption capacity can be calculated according to the intercept. The adsorption rate constant can be obtained

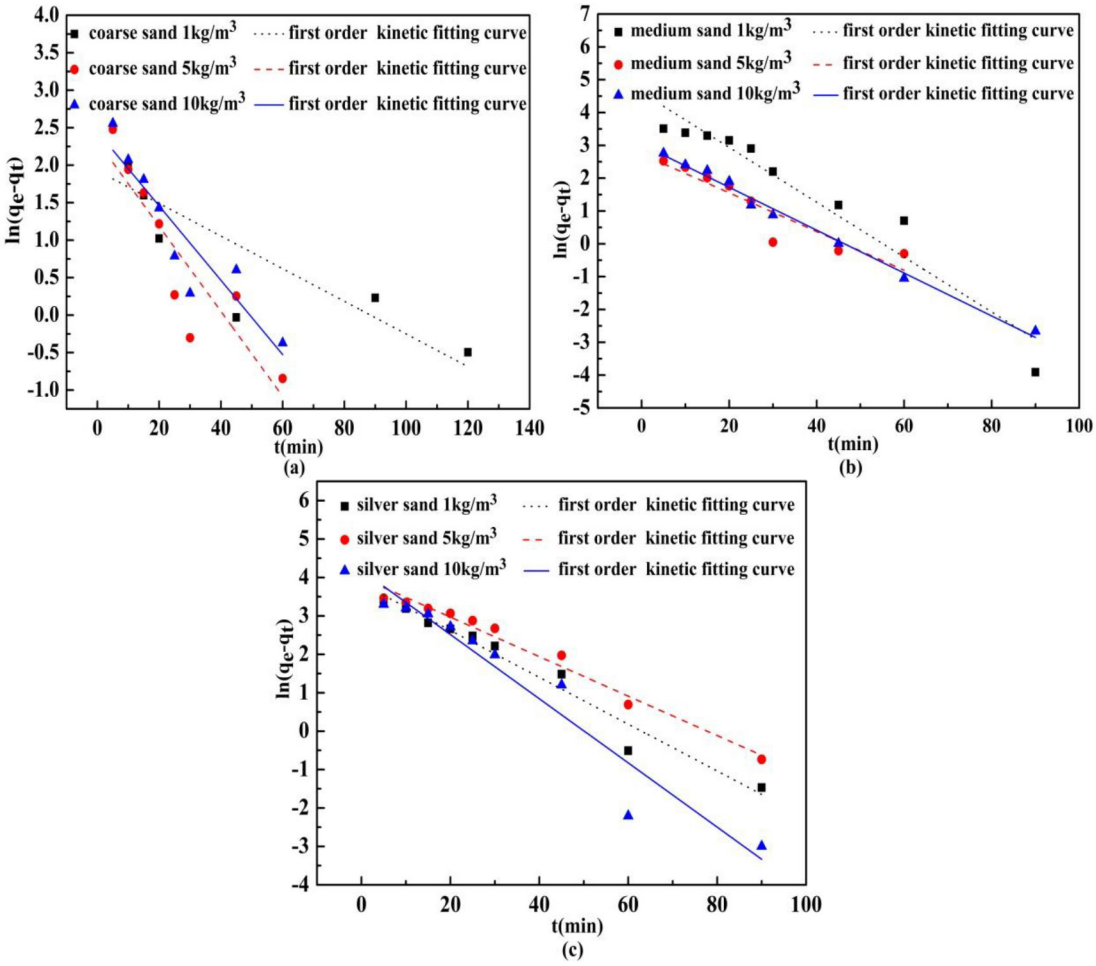


Fig. 4. Fitting curves of the first order kinetics with different sediment gradation: (a) coarse sand, (b) medium sand, and (c) silver sand.

Table 2
Parameters of the first order kinetic model with different sediment gradation

Sediment gradation	Sediment concentration (kg/m^3)	First order kinetic model			
		k_1	q_e	q_e*	R^2
Coarse sand	1	0.0218	6.8569	19.6300	0.7066
	5	0.0567	10.1736	18.2200	0.7786
	10	0.0496	11.5742	17.3200	0.8374
Medium sand	1	0.0836	100.3195	44.1400	0.9198
	5	0.0415	15.6143	23.9900	0.8481
	10	0.0653	20.6768	20.9100	0.9902
Silver sand	1	0.0610	46.5078	52.6400	0.9624
	5	0.0514	54.2900	44.9700	0.9769
	10	0.0835	65.7559	38.1700	0.9218

from the intercept and the equilibrium adsorption capacity. Compared with the first order kinetic model, the two-level kinetic model reveals the entire absorption process which is consistent with the rate control step.

The experimental results were fitted with the two-level kinetic model. The fitting curves are shown in Fig. 5, and the corresponding first order kinetic parameters are shown in Table 3.

It can be seen that the difference between the equilibrium adsorption capacity q_e and the experimental results q_e* was small, so by fitting the experimental results with the two-level kinetic equation can well describe the arsenic adsorption process. It also can be seen from Table 4 that the two-level kinetic model correlation coefficient of coarse sand, medium sand and silver sand could reach more than 0.98, among them, the coarse sand has better correlation and the

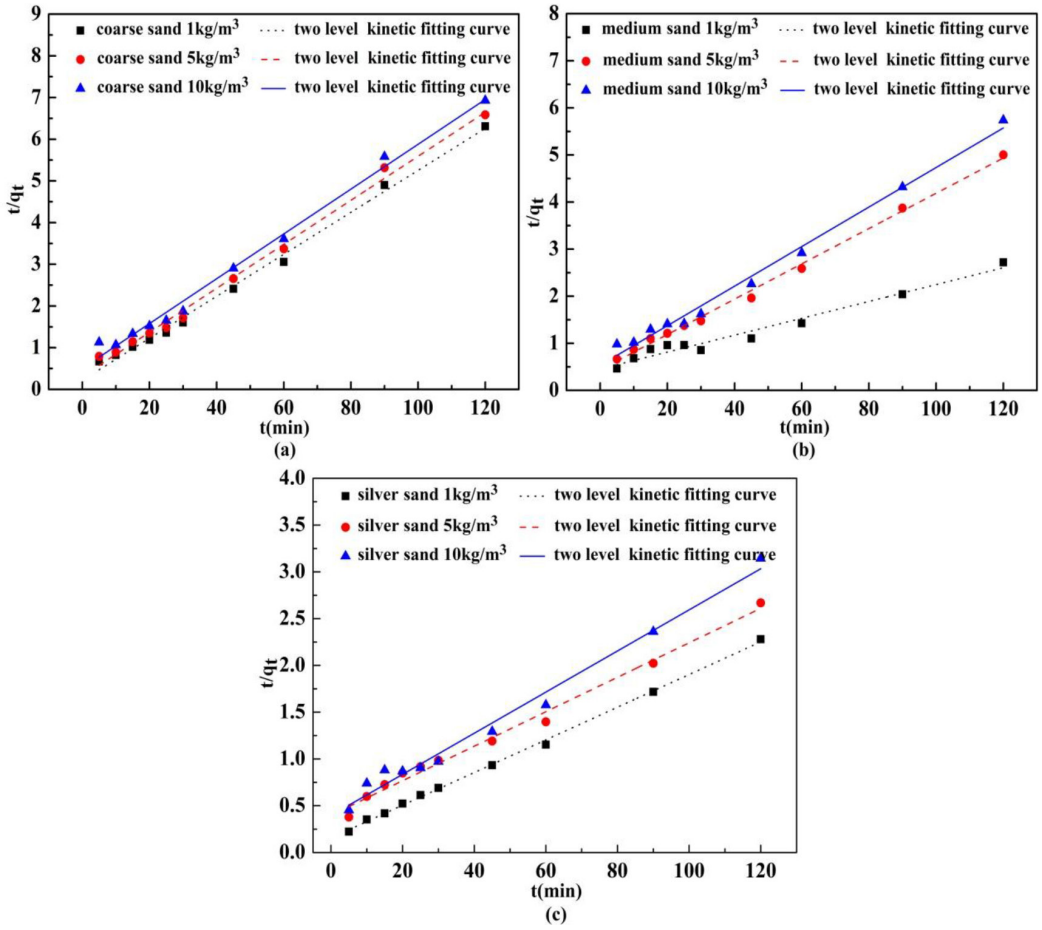


Fig. 5. Two-level kinetics fitting curve of the different sediment gradation: (a) coarse sand, (b) medium sand, and (c) silver sand.

Table 3
Parameters of the two-level kinetic model under different gradation

Sediment gradation	Sediment concentration (kg/m³)	Two-level kinetic model			
		K_2	q_e	q_e^*	R^2
Coarse sand	1	0.0115	19.8650	19.6300	0.9949
	5	0.0089	18.9466	18.2200	0.9941
	10	0.0057	18.6047	17.3200	0.9909
Medium sand	1	0.0007	55.8347	44.1400	0.9662
	5	0.0032	26.6667	23.9900	0.9958
	10	0.0033	23.8095	20.9100	0.9897
Silver sand	1	0.0019	57.2738	52.6400	0.9985
	5	0.0009	54.2888	44.9700	0.9886
	10	0.0012	45.4959	38.1700	0.9834

correlation coefficient can reach above 0.99, which indicated that the two-level kinetic fitting has a good linear correlation.

3.4. Mass transfer kinetics

The particle diffusion model can be expressed as [25]:

$$q_t = k_i t^{1/2} + C \quad (5)$$

where q_t is the adsorption capacity of unit adsorbent at t moment (μg/g); k_i is the diffusion rate constant (μg g^{-1}·$min^{1/2}$); c is the characterization of the boundary layer effect and the degree of membrane diffusion. The parameter c has positive correlations with the external diffusion effect.

A plot of the intra particle diffusion model can be obtained by taking $t^{1/2}$ as the abscissa and q_t as the ordinate, the slope is rate constant k_i and intercept is parameter c. The parameter c is considered to characterize the boundary layer

effect and membrane diffusion degree; its value can reflect the impact of liquid film diffusion (external surface diffusion) in the adsorption rate controlling step. In general, the value of parameter c increases with the thickness of the diffusion boundary layer, the liquid film diffusion rate decreases with the increase of mass transfer resistance, which can enhance the effect of liquid film diffusion.

3.4.1. Mass transfer kinetics model

The simulation results of the mass transfer kinetics model can be used to determine the controlling factors of specific adsorption process. Intra particle diffusion model is one of the mass transfer kinetic models, which is used most widely, therefore, in this study, the intra particle diffusion model is used to study the adsorption process in order to further reveal the adsorption mechanism. The experimental data and the fitting curve of intra particle diffusion model are shown in Fig. 6, and the fitting parameters are shown in Table 4.

It can be seen from Table 4 that the average value of correlation coefficient R^2 with different sediment gradation were 0.7280, 0.7745 and 0.8501 respectively, which means the correlation coefficient increases with decrease of particle size. The fitting results show that silver sand is better

Table 4
Parameters of the intra particle diffusion model under different gradation

Sediment gradation	Sediment concentration (kg/m^3)	k_i	c	R^2	Average value of R^2
Coarse sand	1	1.4582	7.0865	0.7345	0.7280
	5	1.1109	7.6661	0.6914	
	10	1.2489	5.3729	0.7491	
Medium sand	1	4.3428	3.9053	0.8351	0.7745
	5	1.8254	7.3080	0.7755	
	10	1.6953	5.8861	0.7130	
Silver sand	1	3.430	21.1829	0.8258	0.8501
	5	4.0575	6.2199	0.9138	
	10	3.4324	6.9459	0.8108	

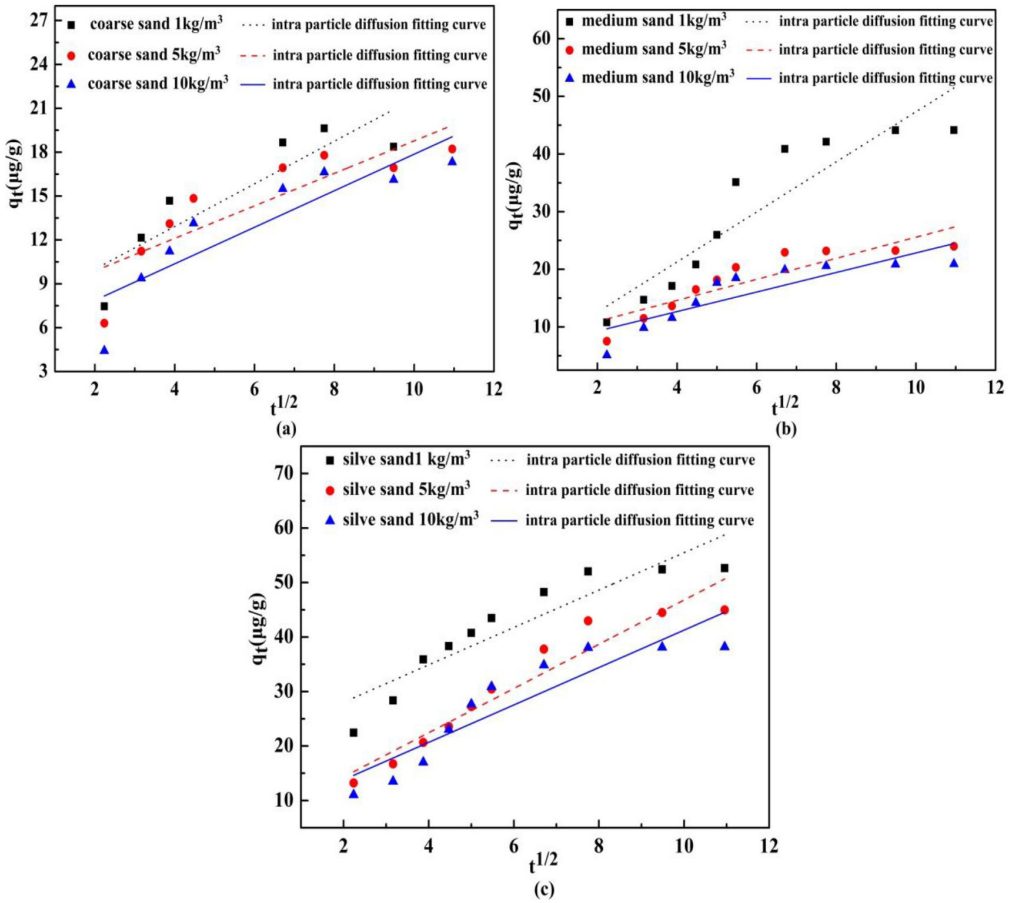


Fig. 6. Intra particle diffusion fitting curve of the different sediment gradation: (a) coarse sand, (b) medium sand, and (c) silver sand.

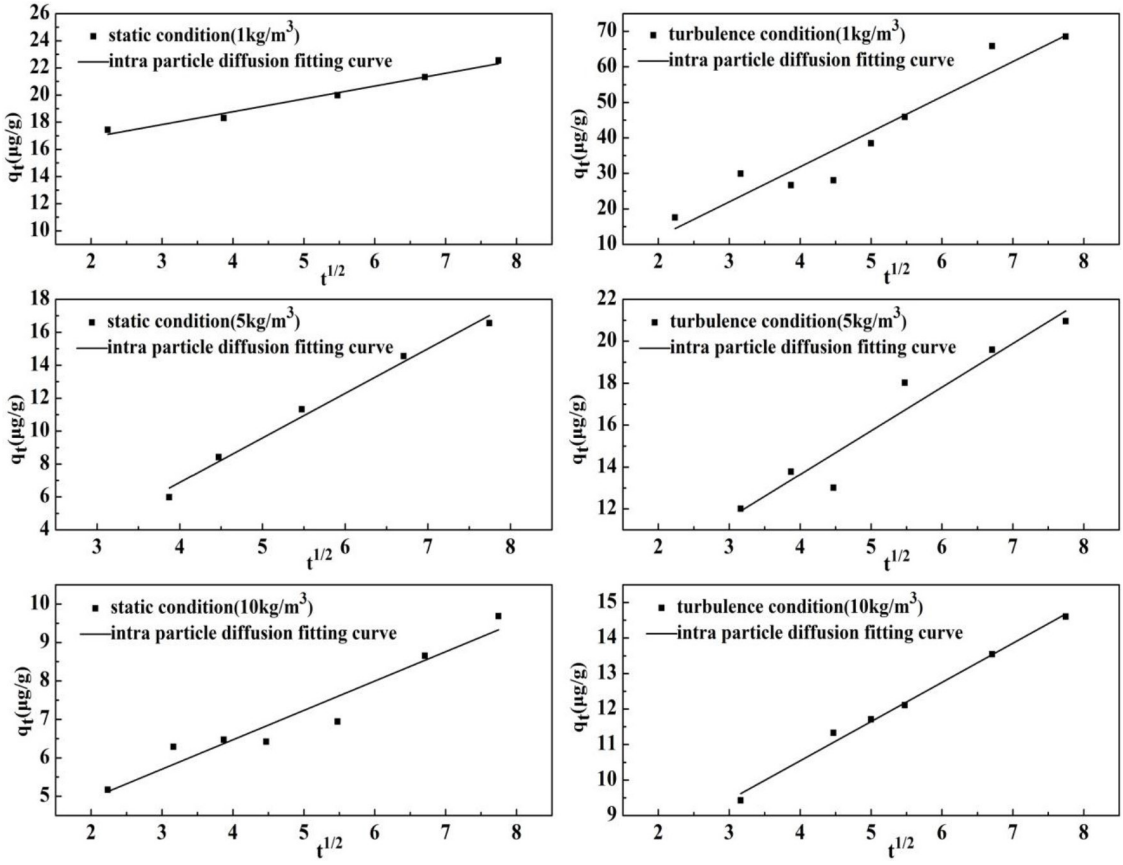


Fig. 7. Mass transfer kinetic model fitting in static and turbulent conditions.

than medium sand and coarse sand. Because the value of parameter c is not equal to 0, which means the adsorption reaction is mainly controlled by the intra particle diffusion, but the particle diffusion is not the only rate control step in the adsorption process, the adsorption rate is also affected by liquid film diffusion and surface adsorption.

3.4.2. Mass transfer kinetic model fitting curve in static and turbulent conditions

As shown above, the experimental results of silver sand fitted well with the mass transfer kinetic model. For a deep study of the mass transfer rules, the results of adsorption experiment of silver sand, which were carried out at both static and turbulent conditions, were fitted with the mass transfer kinetic model. The fitting results are shown in Fig. 7 and the fitting parameters are shown in Table 5.

As shown in Fig. 7 and Table 5, the average values of correlation coefficient R^2 were 0.9598 and 0.9368 in static and turbulent conditions respectively, the fitting correlation coefficient R^2 of the silver sand at a concentration of 10 kg/m^3 in turbulent conditions was 0.991, which indicated that the fitting results are very good and the fitting effect performs better in static condition than that in turbulent condition. This is often observed in experimental mass transfer work. Moreover, because parameter c is the characterization of the boundary layer effect and the degree of membrane diffusion, at the same time, the value of parameter c is not equal to 0, so the adsorption reaction is mainly controlled by the diffusion of the particles and the adsorption rate is also affected by liquid film diffusion and surface adsorption.

Table 5
Parameters of intra particle diffusion model in static and turbulent conditions

Experimental condition	Sediment concentration (kg/m^3)	k_i	c	R^2
Static	1	0.9452	14.9929	0.9760
	5	2.7067	–3.9470	0.9836
	10	0.7632	3.4174	0.9198
Turbulent	1	9.8748	–7.6413	0.9042
	5	2.0828	5.3115	0.9154
	10	1.1036	6.1275	0.9910

4. Conclusions

In this paper, the effects of different gradation of the Yellow River sediment on arsenic adsorption were analyzed. The first order kinetic model, the two-level kinetic model and the intra particle diffusion model were used to fit the experimental results, through the fitting curves and the parameters of different kinds of kinetics model and mass transfer model, the results are shown as follows:

1. The first order kinetic model was used to fit the experimental results, which showed that the correlation coefficient performance as the silver sand was better compared with coarse and medium sand. From the difference between the equilibrium adsorption capacity q_e and the experimental results q_e*, it can be seen that the error was relatively large.
2. The correlation coefficient of coarse sand was larger than medium sand and silver sand in the two-level kinetic model fitting parameters, which revealed that the coarse sand has better adsorption for arsenic. The difference between the equilibrium adsorption capacity q_e and the experimental results q_e* was small, which showed the error of two-level kinetic fitting was small.
3. The average value of correlation coefficient of different sediment gradation were 0.7280, 0.7745 and 0.8501, respectively, by fitting with the intra particle kinetic model, the fitting results manifested as silver sand was better than medium sand and coarse sand. Because the value of parameter c was not equal to 0, so the adsorption reaction was mainly controlled by the intra particle diffusion.
4. The intra particle kinetic model in static and turbulent conditions was used to fit the experimental results, which showed that the average values of correlation coefficient were 0.9598 and 0.9368 in static and turbulent conditions, indicating that the fitting in static conditions had good correlation. Because the value of parameter c was not equal to 0, it further revealed that the adsorption reaction was affected by the intra particle diffusion, liquid film diffusion and surface adsorption.

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