Effects of pore size and dissolved organic matters on diffusion of arsenate in aqueous solution

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ABSTRACT

Presented here is the influence of membrane pore size and dissolved organic matters on the diffusion coefficient (D) of aqueous arsenate, investigated by the diffusion cell method for the first time. The pH-dependent diffusion coefficient of arsenate was determined and compared with values from previous studies; the coefficient was found to decrease with increasing pH, showing the validity of our novel diffusion cell method. The D value increased dramatically as a function of membrane pore size at small pore sizes, and then increased slowly at pore sizes larger than 2.0 μm. Using the ExpAssoc model, the maximum D value was determined to be 11.2565×10^{-6} cm^2/sec. The presence of dissolved organic matters led to a dramatic increase of the D of arsenate, which could be attributed to electrostatic effects and ionic effects of salts. These results improve the understanding of the diffusion behavior of arsenate, especially the important role of various environmental parameters in the study and prediction of the migration of arsenate in aquatic water systems.

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Introduction

Arsenic (As), a ubiquitous and highly toxic metalloid element, has an extremely important influence on human health (Mohan and Pittman, 2007; Smedley and Kinniburgh, 2002). In particular, it has been estimated that 19.6 million people in China have been threatened by problems caused by arsenic-contaminated groundwater, according to a recent research report (Rodriguez-Lado et al., 2013). Most environmental arsenic problems result from the migration and transformation of arsenic through the combination of natural processes and anthropogenic activities (Mohan and Pittman, 2007; Smedley and Kinniburgh, 2002). Understanding and predicting the mobile behavior of arsenic under various environmental conditions are important considerations for

the development of remediation techniques for arsenic-contaminated soil and water (Tanaka et al., 2013; Wang et al., 2013).

Arsenate (As(V)) is the predominant arsenic species in near-surface environments (Mohan and Pittman, 2007; Smedley and Kinniburgh, 2002). Under oxidizing conditions, arsenate does the greatest harm to human health among all arsenic species, especially in surface drinking water. An effective way to reduce the risk of arsenate is to use Fe- or Al-based materials, through coagulation-coprecipitation and adsorption processes, to remove As(V) from natural aquatic systems (Carabante et al., 2012; Mertens et al., 2012). Extensive research has been focused on investigation of the adsorption of arsenate on Fe- or Al-based materials by combining batch adsorption experiments, powder X-ray diffraction, transmission-Fourier

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transform infrared, attenuated total reflectance-Fourier transform infrared and extended X-ray adsorption fine structure analyses (Carabante et al., 2012; Dixit and Hering, 2003; Jia et al., 2007; Mertens et al., 2012; Neupane et al., 2014; O'Reilly et al., 2001; Swedlund et al., 2014). The previous results have shown that the adsorption of arsenate can be divided into two stages, that is, a rapid adsorption step followed by a slow adsorption step, which can last several months or even longer (Swedlund et al., 2014). The long-term adsorption processes have been attributed to surface precipitation and inter-particle diffusion, and formation of inner-sphere surface complexes by ligand exchange reactions (Mertens et al., 2012). In other words, the diffusion velocity into the inner pore space of particles determines the long-term arsenate adsorption and has an impact on controlling the mobility and fate of arsenate. Thus, understanding the diffusion of arsenate in channels and pores is of great significance to the improvement of arsenate removal and kinetics. One important parameter for understanding ion migration in various aquatic systems is the diffusion coefficient (D), which has been measured for various ions (Deen et al., 1981; Furukawa and Takahashi, 2008; Furukawa et al., 2007; Tanaka et al., 2013; Velizarov, 2013; Wang et al., 2001). However, to the best of our knowledge, studies on the diffusion behavior of arsenate are still limited. Up to now, only one research study was performed on the influence of pH on the diffusion coefficients of different arsenic compounds (Tanaka et al., 2013).

On the other hand, dissolved natural organic matters are often present in soil and sediment, and can significantly affect the mobility and fixation of arsenate in the natural environment (Ikari et al., 2015; Shi et al., 2009). In particular, arsenate and dissolved organic matters can react to form a small fraction of organic arsenic compounds (Langner et al., 2011; Tanaka et al., 2013). It has been reported that complexes can be formed between metal ions and dissolved organic matters, such as EDTA and humic substances, and the diffusion coefficients of free metal ions are greatly decreased in the presence of dissolved organic matters (Furukawa and Takahashi, 2008; Furukawa et al., 2007). However, understanding of the effect of dissolved natural organic matters on the diffusion coefficients of arsenate in aquifer systems is still lacking.

Thus, the objectives of this work are: (1) to determine the diffusion coefficient of arsenate at different pH values using our diffusion cell method, compared with the D values determined in previous works; (2) to investigate the influence of pore size and dissolved organic matters on the diffusion coefficient of arsenate. The information on the diffusion of arsenate obtained by this experimental approach will have important significance in guiding the synthesis of highly efficient and low-cost adsorbents, and especially for understanding and predicting the migration and fate of arsenate under various environmental conditions.

1. Material and methods

1.1. Materials and diffusion cell method

All chemicals used were of analytical or reagent grade and purchased from commercial sources, and used to prepared solutions in deionized water without further purification. All volumetric flasks and vessels before tests were cleaned by soaking in 10% HNO_3 for at least 24 hr and rinsed several times with distilled water. The As stock solutions for batch studies were made by dissolving $Na_2HAsO_4 \cdot 7H_2O$ in deionized (DI) water and used to prepare working solutions by diluting the stock arsenate solution with DI water. The conductivity of the DI water used in our experiments was about 5.8 μS/cm, determined with a conductivity meter (model MC 230, Radiometer Analytical S.A. France).

The diffusion cell constructed for this work is made of Plexiglas, and composed of two cylindrical chambers with a membrane separating the left and right cells. The volume of each chamber is about 8 mL. The pore diameters of membranes used in this research (Hybrid Membrane Filters, Millipore) were 0.10, 0.22, 0.45, 0.65, 0.80, 1.20, 2.00 and 5.00 μm, with the exposed area of 3.8 cm^2, thickness of 13 μm, and porosity of 0.80. The membranes generally have well-distributed pores with very uniform size, which are suitable qualities for the experiment (Ding and Elmore, 2016). At the beginning of the experiment, the left chamber in the cell (source cell) was filled with a solution containing arsenate and other anions, while the right chamber (measurement cell) was filled with the same volume of DI water. The ions diffused through the membrane because of the concentration gradient between the source and measurement cells. The diffusion coefficients of the ions were determined by measuring their concentrations over time. During the diffusion experiments, the temperature of the cells was controlled by immersion in a water bath to maintain a constant temperature of 29.0 ± 0.2°C. The temperature was selected according to some environmental reports showing that the maximum temperature of surface water can reach 30°C. The solution in each chamber was vigorously stirred by a magnetic stirrer at a constant speed.

1.2. The mass transfer resistance model

The model used to determine the diffusion coefficient in this study was described in previous studies (Deen et al., 1981; Furukawa et al., 2007; Tanaka et al., 2013; Wang et al., 2001). Briefly, the researchers studied the mass transfer by a similar diffusion cell method and established a model considering the total mass transfer resistance. The equation representing the flux across the membrane was derived as follows:

$$\ln \frac{(C_L - C_R)_0}{(C_L - C_R)_t} = \frac{2Ak}{V} t \qquad (1)$$

where, C_L (mmol/L) and C_R (mmol/L) are the concentrations of arsenate in the source and measurement cells, respectively, while A (3.8 cm^2) is the membrane area in the cell, k is the mass transfer coefficient, and V (mL) is the volume of the solution in the source and measurement cells eq(8 mL in our study). By plotting $\ln(C_L - C_R)_0 / (C_L - C_R)_t$ versus time, the slope $2Ak/V$ can be determined by linear regression analysis. The k value must be calibrated to obtain the unknown diffusion coefficients, which can be determined knowing the membrane area A and solution volume V, and evaluates the

mass transfer resistance. Then, the total mass transfer resistance R_t is expressed as:

$$R_t = \frac{1}{k} = \frac{2A}{V \times slope} \tag{2}$$

In the diffusion model, the total resistance to mass transfer is described as:

$$R_t = \frac{1}{k} = R_m + 2R_b + 2R_e \tag{3}$$

where R_m is the resistance from the membrane, R_b is that of the boundary layer, and R_e is that of the end effect at the entrance and exit of the pores (Deen et al., 1981; Wang et al., 2001). In our experiments, the pore radii of the membranes used in the diffusion cells were much greater than the radius of the solute molecules. Herein, the Renkin equation shows that the diffusion coefficient in the pores equals the diffusion coefficient in the bulk solution at the infinite pore size limit, which suggests that the relationship of k to the diffusion coefficient (D) could be simplified to (Deen et al., 1981).

$$R_t = \frac{1}{k} = \frac{k_1 + k_3}{D} + \frac{k_2}{D^{2/3}} \tag{4}$$

As pointed out in previous research, the three constants (k_1, k_2, and k_3) characterizing the mass transfer in the diffusion cell should depend on the membrane properties, the cell geometry, and the stirring speed, but not on the nature of the solutes (Deen et al., 1981; Wang et al., 2001). Since the k value can be directly measured from the diffusion experiments, D can be calculated from Eq. (4) once the constants ($k_1 + k_3$) and k_2 are known. Diffusion experiments for solutes with known diffusion coefficients were carried out to determine the constants ($k_1 + k_3$) and k_2, and these constants were employed to determine the unknown diffusion coefficients of the other solutes under the same experimental conditions.

1.3. Determination of the constants ($k_1 + k_3$) and k_2

In the calibration studies, KCl, NaCl, and CaCl$_2$, whose diffusion coefficients have been reported in the literature (Weast, 1975), were employed to determine the constants ($k_1 + k_3$) and k_2. The diffusion coefficient values of these substances can be adjusted for changes in temperature by the equation:

$$D_{T2} = \frac{T_2 \mu_1}{T_1 \mu_2} D_{T1} \tag{5}$$

where μ is the viscosity of the solution (deionized water in our experiments), which can be calculated as a function of temperature (Stokes, 1950):

$$\log \frac{\mu_T}{\mu_{20}} = \frac{1.3272(20-T)-0.001053(T-20)^2}{T+105} \tag{6}$$

For the salt diffusion experiments, the salt concentration of the source cell was about 0.2 mol/L (8 mL) and the measurement cell was filled with the same volume of deionized water. At certain time intervals, 200 μL of solution was taken from the measurement cell to examine the concentrations in the right chamber. At the same time,

0.2 mL deionized water was replenished to keep the volume constant. The concentrations of NaCl, KCl and CaCl$_2$ in solution were measured with a conductivity meter (model MC 230, Radiometer Analytical S.A. France) on the basis of the linear relationship between the concentration and conductivity under the specific concentrations (Appendix A Fig. S1), and proper dilution was carried out for higher concentrations of salt if necessary. The concentrations in the source cell as a function of time were determined by the difference between the initial concentration and that in the measurement cell, as given in Eq. (7):

$$C_{Lt} = \frac{C_{L0} \times 8 - (C_N \times 8 - \sum_{i=1}^{N-1} C_i \times 0.2)}{8} \tag{7}$$

where C_{Lt} (mmol/L) and C_N (C_{Rt}) (mmol/L) are the concentrations of the solute in the lift and right chambers at a given time, respectively.

1.4. Determination of the diffusion coefficients

For the arsenate diffusion test, the source cell was filled with a solution containing 4.394 mmol/L arsenate prepared by dissolving Na$_2$HAsO$_4$·7H$_2$O in DI water. Aliquots of 0.2 mL of solution were collected at a series of time intervals from the measurement cell, and 0.2 mL deionized water was replenished after each sampling. The solutions of source and measurement cells were adjusted to the same pH using NaOH and HCl before the experiments. The concentrations of arsenate were measured using a hydride-generation atomic fluorescence spectrophotometer (AFS-2202E, Haiguang Corp., Beijing) with a detection limit of 10 μg/L, and duplicate analyses agreed within 5%.

The same experimental procedure and conditions described above were employed in the study of the effect of dissolved organic matters on diffusion of arsenate, unless otherwise specified. Five types of organic acids were purchased from commercial sources. The source cell was filled with mixtures of dissolved organic matters and arsenate, respectively, and then equilibrated at least 72 hr. Each arsenate concentration of the source cell was 0.2197 mmol/L, and the mole ratio of arsenate to dissolved organic matters was 1:20. It should be noted that the mixtures were made by the addition of the organic acid salt solids to arsenate solutions without further action. After equilibration, the pH values of the mixtures were measured. The pH of the DI water in the measurement cell was the same as that of the mixture in the source cell. All the experimental diffusion coefficients given below are the average values of triplicate runs and were used to analyze the results of the study.

2. Results and discussion

2.1. Determination of the constants ($k_1 + k_3$) and k_2

Because the constants ($k_1 + k_3$) and k_2 depended on the membrane properties, the cell geometry, and the stirring speed, but not on the nature of the solutes, three kinds of salts

with known diffusion coefficients were used to determine these constants. As shown in Fig. 1, based on the plots of $\ln(C_L - C_R)_0 / (C_L - C_R)_t$ versus time using membranes with different pore diameters, the parameters were obtained by linear simulation, as depicted in Appendix A Table S1. The results show that the data fitted well with the linear model, with the regression coefficients (R^2) higher than 0.99 in all cases. Then, R_t values could be calculated from the slopes of the lines by linear regression. Combining two solutes, several groups of the constants ($k_1 + k_3$) and k_2 were obtained through Eq. (4). Table 1 presents the calculated constants ($k_1 + k_3$) and k_2. The final values for ($k_1 + k_3$) and k_2 were obtained from the average of the sets of data, and used to interpret the diffusion experiments.

2.2. Diffusion coefficients of arsenate at various pH values

It is extremely important to investigate the influence of pH on the diffusion coefficients of arsenate, because pH plays an important role in controlling the mobility and fate of arsenate in soils and sediments (Carabante et al., 2012; Jia et al., 2007). Previously, Tanaka et al. performed *ab initio* molecular orbital calculations and Monte Carlo (MC) simulations to analyze the hydration numbers and hydration structures of arsenate species, to understand the dominant factors controlling the diffusion coefficients of

arsenic compounds under various pH conditions (Tanaka et al., 2013). Herein, we determined the diffusion coefficient of arsenate as a function of pH, which was compared with Tanaka's report to assess the accuracy of the diffusion cell method used in our experiments. Using the values for the constants ($k_1 + k_3$) and k_2 obtained from the calibration tests and R_t values from the slopes of the lines (Fig. 1a, b and Table 1), the values of the diffusion coefficient (D) of arsenate were calculated using Eq. (4) and presented in Fig. 2. As shown in Fig. 2, the D values

Table 1 – Model constants ($k_1 + k_3$) and k_2 obtained using the membranes with different pore sizes, with the known diffusion coefficient (D) values of KCl, NaCl and CaCl$_2$.

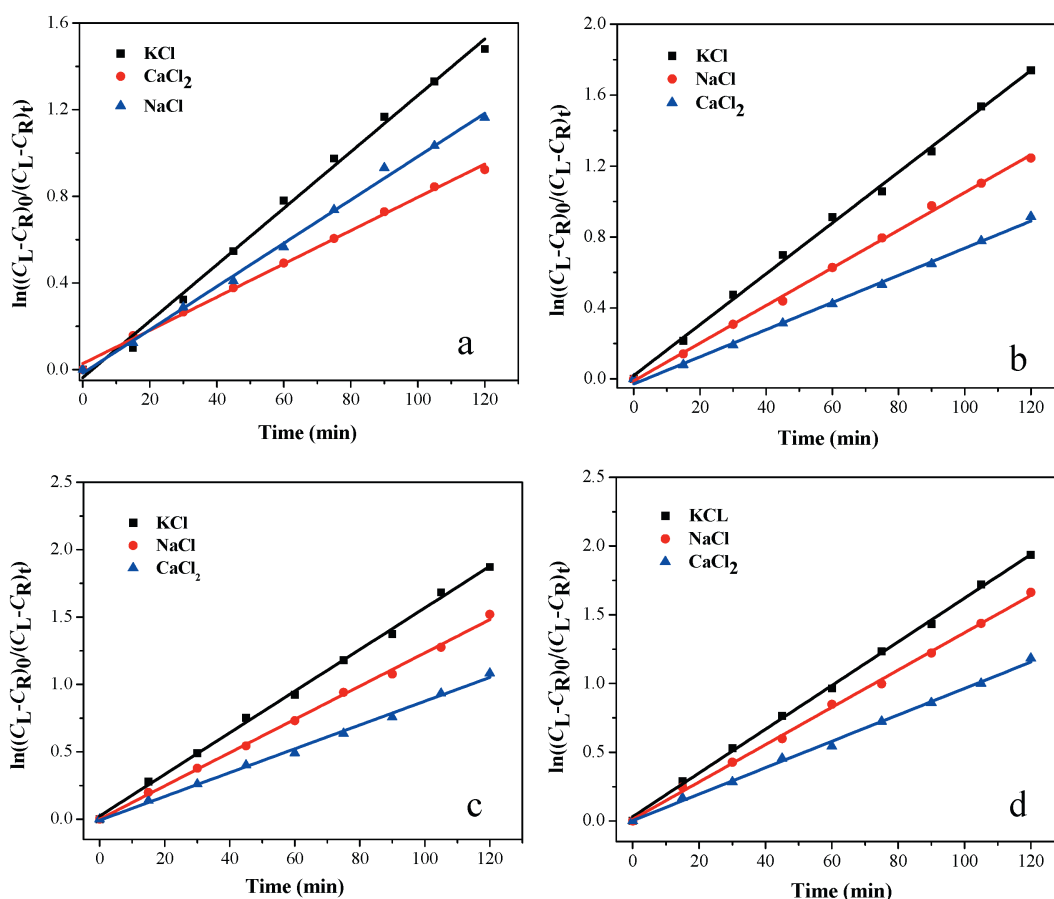| Pore size (μm)	Model constant	NaCl and KCl	KCl and CaCl$_2$	NaCl and CaCl$_2$	Average
0.22	$k_1 + k_3$ (cm)	0.0104	0.0099	0.0101	0.0102
	k_2 (cm$^{1/3}$·S$^{1/3}$)	−0.0802	−0.0746	−0.0779	−0.0776
0.80	$k_1 + k_3$ (cm)	0.0162	0.0146	0.0128	0.0145
	k_2 (cm$^{1/3}$·S$^{1/3}$)	−0.3089	−0.2453	−0.1860	−0.2467
1.20	$k_1 + k_3$ (cm)	0.0052	0.0101	0.0146	0.0997
	k_2 (cm$^{1/3}$·S$^{1/3}$)	0.0497	−0.1133	−0.2880	−0.1172
5.00	$k_1 + k_3$ (cm)	−0.0002	0.0073	0.0141	0.0071
	k_2 (cm$^{1/3}$·S$^{1/3}$)	0.2230	−0.0262	−0.2934	−0.0322

Fig. 1 – The dependence of concentration gradients in KCl, NaCl and CaCl$_2$ diffusion experiments on the elapsed time using the membranes with different pore sizes: (a) 0.22 μm; (b) 0.80 μm; (c) 1.20 μm; (d) 5.00 μm.
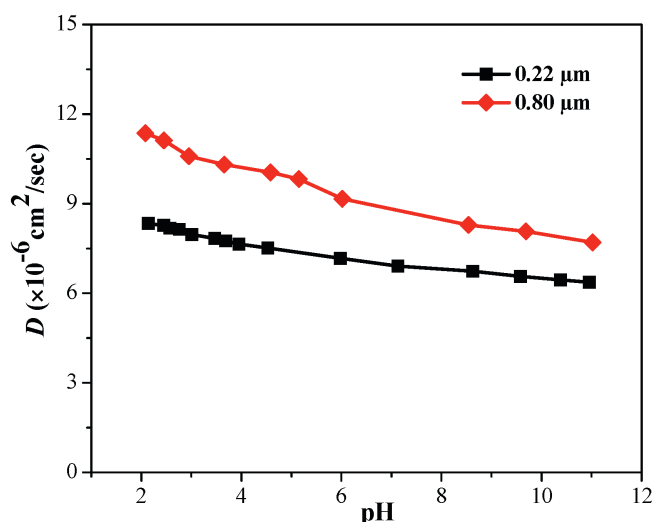
Fig. 2 – Diffusion coefficients (D) of arsenate at various pH. The pore diameters of membranes used for the diffusion experiments were 0.22 and 0.80 μm, respectively.

depended on pH and decreased with increasing pH. Using a membrane with the pore size of 0.80 μm, the D values were estimated to be 11.36×10^{-6} cm^2/sec at pH 2.08, 10.048×10^{-6} cm^2/sec at pH 4.58, 8.2836×10^{-6} cm^2/sec at pH 8.54, and 7.7034×10^{-6} cm^2/sec at pH 11.02, respectively. The D values were somewhat larger than the D values previously reported (Tanaka et al., 2013). This could be attributed to the fact that our experiments were carried out at a higher temperature. However, the D values for arsenate were estimated to be 8.3329×10^{-6} cm^2/sec at pH 2.14, 7.5162×10^{-6} cm^2/sec at pH 4.53, 6.7358×10^{-6} cm^2/sec at pH 8.63, and 6.3649×10^{-6} cm^2/sec at pH 10.96, when the pore diameter of the membrane used was 0.22 μm. These results were similar to those in the previous reports (Tanaka et al., 2013), indicating the reliability of the diffusion cell method used to determine the D values. Moreover, Fig. 2 also shows that for membranes with larger pore sizes, larger diffusion coefficients were obtained at the corresponding pH values.

2.3. Diffusion coefficients of arsenate using membranes with different pore diameters

Numerous studies have shown that the adsorption of arsenic onto iron oxides includes the transport of the adsorbate to the surface by convection and molecular diffusion, interaction with the surface, intraparticle diffusion, and surface-complex formation (Swedlund et al., 2014). According to these reports, the adsorption processes of arsenate and arsenite are known to be strong and fast, forming predominantly inner-sphere surface complexes by ligand exchange reactions (Gao et al., 2013; Jia et al., 2007). However, the subsequent processes related to adsorption are slower, which include surface precipitation or diffusion into the pore space of particles to form inner-sphere surface complexes (Mertens et al., 2012). The pore size and distribution are very significant factors in the determination of the uptake and kinetics of arsenic by

iron or aluminum-based minerals in long-term geochemical reaction processes. Thus it is necessary to study the diffusion coefficient of arsenate as a function of pore size. The variation of the diffusion coefficient of arsenate with pore diameter is shown in Fig. 3. These results indicate that the diffusion coefficient of arsenate increased with increasing pore diameter when the pore size was less than 2.0 μm. When the size was larger than 2.0 μm, the diffusion coefficient of arsenate exhibited less dependence on pore diameter. The reason for this phenomenon could be attributed to electrostatic interactions and the resistance of the boundary layer (R_b) (Furukawa and Takahashi, 2008).

The ExpAssoc model was employed to describe the relationship between the diffusion coefficient of arsenate and pore size and evaluate the maximum diffusion coefficient value, which is expressed as follows:

$$y = y_0 + A_1\left(1 - e^{-\frac{x}{t_1}}\right) + A_2\left(1 - e^{-\frac{x}{t_2}}\right) \tag{8}$$

where x is the pore size of the membrane, y_0, A_1, A_2, t_1, and t_2 are invariant constants, and the sum of y_0, A_1 and A_2 is the maximum D as the exponential functions tend to zero at large x. The result shows that the D value data was fitted well by the ExpAssoc model, with a correlation coefficient (R^2) of 0.83. The constants obtained from Eq. (8) were 5.8689, 2.6938 and 2.6938 for y_0, A_1 and A_2, respectively, indicating that the maximum diffusion coefficient value was 11.2565×10^{-6} cm^2/sec.

2.4. Effect of dissolved organic matters on diffusion coefficients of arsenate

Sodium citrate, potassium biphthalate, 5-sulfosalicylic acid, ethylenediaminetetraacetic acid disodium salt (EDTAH$_2$Na$_2$), and potassium antimony tartrate were employed to study the influence of dissolved organic matters on the diffusion coefficient of arsenate. The results were compared with the D values of arsenate *versus* pH in the absence of dissolved organic
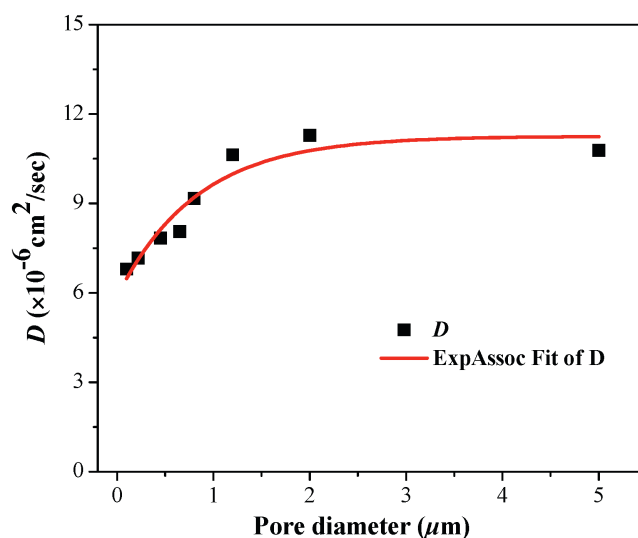

Fig. 3 – Diffusion coefficients (D) of arsenate using membranes with different pore sizes. The experiments were carried out at about pH 6.02.
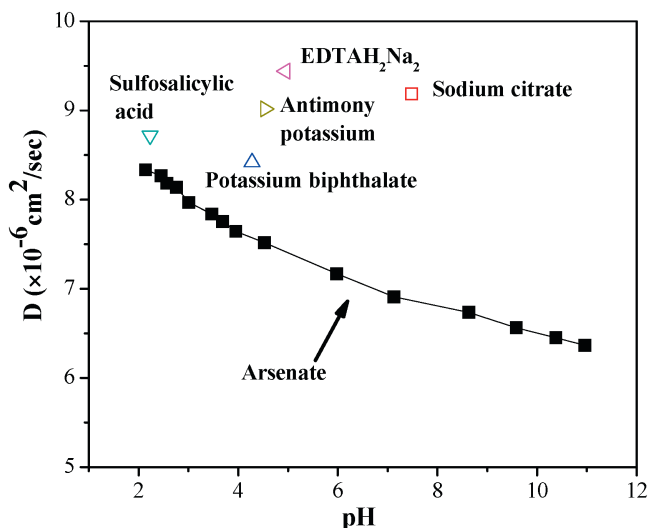
Fig. 4 – The diffusion coefficient of arsenate at different pH values in the absence of the dissolved organic matters and the effect of the dissolved organic matters on the diffusion coefficients of arsenate. The pore diameters of membranes used for all the diffusion experiments were 0.22 μm and the mole ratio of arsenate to dissolved organic matters was about 1:20.

matters using the membrane with the pore size of 0.22 μm, which showed that the diffusion coefficient of arsenate increased in the presence of dissolved organic matters (Fig. 4 and Table 2). The ability of dissolved organic matters to increase the D values ranked in the order: sodium citrate > disodium salt > potassium antimony tartrate > potassium biphthalate > 5-sulfosalicylic acid. The main factors affecting the diffusion coefficients could be ascribed to electrostatic effects and ionic effects of salts (Furukawa and Takahashi, 2008). Since diffusion coefficients in water are larger for smaller molecules, the solutes in solution have different diffusion coefficients with the order: Na^+ (H^+) > arsenate > organic anions. The concentrations of organic anions were the same in each case, while the concentrations of cations had wide discrepancies, especially for sodium citrate, with the largest cation concentration (Na^+). These wide differences among the cation concentrations of salts led to different abilities to increase the D values of arsenate in the presence of dissolved organic matters.

On the other hand, small amounts of complexes between arsenate and dissolved organic matters can co-exist in the presence of dissolved organic matters (Furukawa and Takahashi, 2008; Langner et al., 2011; Tanaka et al., 2013), which should be considered and corrected for to determine the effect of the dissolved organic matters on the diffusion

coefficient of arsenate. However, it is difficult to calculate the various dissolved species present in the solution of arsenate and dissolved organic matters, because the processes and mechanisms of the formation of these complexes are unclear; this is currently one of the difficult and hot spots in the field of arsenic chemistry.

3. Conclusions

Arsenic is ubiquitous in many natural systems, and arsenate is the predominant arsenic species under oxidizing environmental conditions. Understanding the factors which impact the diffusion of arsenate has significance in prediction of the migration and bioavailability of arsenate. The results presented here demonstrate the influence of the pore size of membranes and dissolved organic matters on the diffusion coefficient (D) of arsenate using the diffusion cell method. The results show that the D values dramatically increased with increasing membrane pore size at small pore sizes, and then increased slowly with further increases in pore size. In addition, the presence of dissolved organic matters could promote the D of arsenate, attributed to electrostatic and ionic effects among different ions in solution, indicating that the mobility of previously immobilized arsenic could be increased in the presence of dissolved organic matters. Although further studies are required to understand the mechanism of D as affected by more environmental parameters, such as different arsenic compounds, humic substances, etc., the current work undoubtedly improves the understanding of the diffusion of arsenate, which is essential in modeling the migration behavior of arsenate in various environmental systems. Moreover, the results shown in this study may contribute to removing arsenic in flow-through water-treatment processes, especially long-term adsorption processes related to diffusion into the pore space of particles.

Appendix A. Supplementary data

Supplementary data to this article can be found online at http://dx.doi.org/10.1016/j.jes.2016.05.004.

	Sodium citrate	Potassium biphthalate	5-Sulfosalicylic acid	EDTAH₂Na₂	Potassium antimony tartrate
Table 2 – D values of arsenate in the presence of dissolved organic matters in different pH conditions.					
pH	7.48	4.28	2.33	4.95	4.54
D	9.1856	8.4185	8.7175	9.4400	9.0156

D ($\times 10^{-6}$ cm²/sec): diffusion coefficients; pH were the solution pH of the mixtures after equilibration.

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