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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 $11.2565 \times 10^{-6} \text{ cm}^2/\text{sec}$. 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% HNO₃ for at least 24 hr and rinsed several times with distilled water. The As stock solutions for batch studies were made by dissolving Na₂HAsO₄·7H₂O 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 cm², 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:

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

where, C_L and C_R (mmol/L) are the concentrations of arsenate in the source and measurement cells, respectively, while A (3.8 cm²) 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 (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

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