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Kinetic study of silica dissolution in aqueous solutions of aromatic organic electrolytes



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HIGHLIGHTS

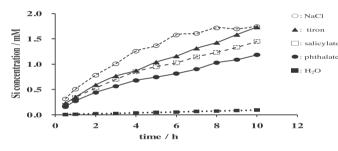
G R A P H I C A L A B S T R A C T

- Rate constant in organic electrolyte solutions 18–30 times larger than in water.
- Compared with water, organic electrolytes increase the dissolution rate of silica.
- Compared with NaCl, organic electrolytes decrease the dissolution rate of silica.
- Pure organic anions decrease the dissolution rate of silica by adsorbed on surface.

ARTICLE INFO

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Keywords: Dissolution rate of silica Kinetic analysis Rate constant Organic electrolyte Adsorption The silicic acid concentration in the organic electrolyte solution was larger than in the water, but less than in the NaCl solution. The facts suggest that organic electrolyte accelerates the dissolution rate of silica compared with water, but retard the dissolution rate of silica compared with NaCl. In fact, organic anions containing in the organic electrolytes are retarding the dissolution rate of silica. The concentration of silicic acid with time varies in the different kinds of solutions. The concentration of sodium ion in each solution is same $(0.2 \text{ mol } \text{L}^{-1})$.



ABSTRACT

To evaluate the effect of organic compounds on the dissolution rate of amorphous silica, the dissolution of amorphous silica in pure water and in organic electrolyte solutions were investigated at room temperature at pH 6. Based on the kinetic analyses, the dissolution rate constants of amorphous silica in the organic electrolyte solutions were 18–30 times larger than in pure water but less than those in the NaCl solution, which contained the same Na⁺ concentration as the organic electrolyte solution. The order of dissolution rate constants was $N_{NaCl} > N_{tiron} > N_{phthalate} > N_{salicylate} > N_{water}$. In these experiments, the dried samples were analyzed by FT-IR, and the results indicated that all of the organic electrolyte solution was adsorbed on the surface of the silica. Compared with pure water, the cations in the organic electrolyte solution rate of silica decreased in the aromatic organic electrolyte solution due to the adsorption of the organic electrolytes on the surface of the silica as well as to a decrease in the exposed surface. In fact, organic anions containing in the organic electrolytes are retarding the dissolution rate of silica. (Due to the surface of the silica as well as to a decrease in the exposed surface. In fact, organic anions containing in the organic electrolytes are retarding the dissolution rate of silica.

1. Introduction

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http://dx.doi.org/10.1016/j.colsurfa.2014.07.048 0927-7757/© 2014 Elsevier B.V. All rights reserved. The reaction of silica with water is of interest for the chemical, geological, biological and technical sciences. Extensive studies of the dissolution rate and solubility of silica have been reported in the past two decades from the viewpoint of mineral weathering

[1–5]. Silica dissolves in water as silicic acid through the breakage of siloxane bonds by the attack of water molecules. The dissolution of silica in water is primarily determined by temperature, pH and the ionic strength of the electrolytes [4]. Inorganic salts can enhance the dissolution rate of silica by a specific sorption of cations, which disrupts the surface structure and facilitates the direct access of water to Si–O–Si bonds [3,6]. Some anions, such as sulfate, can also accelerate the dissolution rate of amorphous silica through specific adsorption [7]. The surfaces of quartz grains from soil which is rich in organic materials have been significantly altered by chemical etching, indicating that organic materials promote the dissolution of guartz [8]. Natural water contains not only various types of inorganic salts but also numerous organic compounds produced during the biodegradation of marine and freshwater organic matter. Dissolved organic compounds in natural water consist primarily of complex organic acids, including aliphatic, aromatic, keto-, and other partially oxidized carbon species [9]. The interaction of these organic compounds with silica and other silicate minerals at near neutral pH could help to understand the weathering process of rock in the environment. However, the influence of organic compounds on the dissolution or mobility of silica in aqueous systems is still poorly documented. The influence of oxalate on the dissolution rate of quartz can be negligible [10]. Our research group have found that the solubility of amorphous silica increases two-fold by tiron (disodium 4,5-dihydroxy-1,3-benzenedisulfonate) through the formation of a complex at near neutral pH [11]. Nevertheless, the influence of tiron on the dissolution rate of amorphous silica has not yet been reported. Therefore, to understand the behavior of silica in environmental aqueous systems, it is necessary to investigate the effect of various types of organic matter on the dissolution of silica.

The aim of this work was to investigate the effect of a group of structurally similar aromatic organic electrolytes (Supplementary 1) on the dissolution rate of amorphous silica. This investigation may provide new and valuable information for understanding the details of the dissolution mechanism of silica in the presence of organic electrolytes and for developing greater knowledge of the weathering processes of silicate minerals.

2. Experimental

2.1. Reagents and sample solutions

Silica gel powder for chromatography (Mallinckrodt, 100 mesh) was used as a source of amorphous silica. Tiron, sodium salicylate (sodium 2-hydroxybenzonate), disodium phthalate (disodium benzene-1,2-dicarboxylate) and NaCl (a special grade) were purchased from Wako Pure Chemical Industries, Ltd. All sample solutions were prepared with ultra pure water.

2.2. Dissolution of amorphous silica in each organic electrolyte solution

To prevent volatilize and photo-oxidation of the organic matters, all experiments were performed in closed containers in a dark room. The 0.1 M (M: mol dm⁻³) solutions of tiron, disodium phthalate, sodium salicylate and 0.2 M NaCl (volume of each solution was 500 cm³) were prepared by dissolving them in ultra pure water. To regulate the concentration of Na⁺ in sodium salicylate solution, NaCl was added to the sodium salicylate solution to maintain a constant Na⁺ concentration of 0.2 M. Silica gel powder (5.0 g) was added to each solution and stirred by a shaker in a dark room at 25 °C. The pH of each solution was monitored using a pH meter. After an adequate interval, an aliquot of the suspended solution was taken out and filtered with a 0.45 μ m membrane filter. The silicic acid

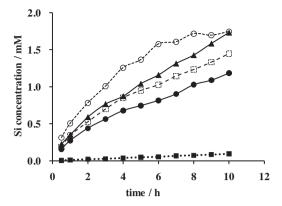


Fig. 1. Variation of silicic acid concentration with time; \bigcirc : in 0.2 M NaCl; \blacktriangle : 0.1 M tiron; \Box : in 0.1 M disodium phthalate; \bullet : in 0.1 M sodium salicylate; \blacksquare : in pure water; pH: 6.2.

concentration in the filtrate was determined by inductively coupled plasma atomic emission spectrometry (ICP-AES, Varian 730-ES). Here, the dissolution of amorphous silica in pure water was used as a control.

2.3. Measurement of infrared spectrum of silica gel

To identify the interaction between the silica gel particles and each organic compound, IR measurements were performed for silica gel using a Fourier Transform IR spectrophotometer (BRUKER TENSOR 27). Fifty milligrams of silica gel powder was added to each organic compound solution and stirred magnetically for 10 h at 25 °C. Then, the silica gel was filtered through a 0.45 μ m membrane filter, rinsed three times with ultra pure water and air dried at room temperature. FT-IR spectra in the wave number range from 400 to 4000 cm⁻¹ were obtained using the KBr pellet technique. For comparison, the IR of silica gel alone was also measured using the same method.

3. Results and discussion

3.1. Dissolution rate of amorphous silica in each organic electrolyte solution

To evaluate the effect of the individual organic electrolyte on the dissolution rate of amorphous silica, dissolution experiments of amorphous silica were conducted in each organic electrolyte solution with a constant Na⁺ concentration and in pure water. As the dissolution rate of amorphous silica depends on pH [12], the experiments were carried out at pH 6. To compare the dissolution rate of silica in the organic electrolyte solutions and the NaCl solution, the dissolution experiment of silica in NaCl solution was also conducted under the same conditions. Fig. 1 shows the variation of silicic acid concentration with reaction time. The concentration of silicic acid increased with time in all cases. The concentration of silicic acid in pure water was considerably lower than the concentration in organic electrolyte solutions, suggesting that the organic electrolyte can accelerate the dissolution of silica. These results are consistent with the literature [13]. Berger et al. reported that cations can increase the dissolution rate of silica [14]. Therefore, compared with pure water, the presence of Na⁺ in the organic electrolyte solutions may be a main factor in enhancing the dissolution rate of silica.

According to the theory that Na⁺ can increase the dissolution of silica and Cl⁻ cannot affect the dissolution of silica, we chose the NaCl solution as reference solution to determine the effect of pure organic compositions on the dissolution of silica. The Download English Version:

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