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Removal of Arsenous Acid from Sulfuric Acidic Solutions using Ultrasound Oxidation and Iron Oxide

超音波による酸化と酸化鉄を用いた硫酸酸性溶液中の亜砒酸の除去

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1. Introduction

Arsenic is used in many different fields such as production of medicine and semiconductors. However, arsenic has a toxic activity for human body. Therefore, it is necessary to treat arsenic in the waste materials and factory effluent using appropriate methods. The values of effluent standard for arsenic in Japan are established as 0.1 ppm. The precipitation of arsenic acid (As(V)) with trivalent iron compounds such as iron hydroxide and ferric oxide has been used as a method to remove arsenic from aqueous solution onto the surface of the trivalent iron compounds. Compared to the removal processes of arsenous oxide (As(III)) using trivalent iron compounds, the removal processes of As(V) are easier to handle. Thus, As(III) is required the oxidation to As(V) using oxidants. Oxidants and bacteria have both been commonly used as oxidation methods of As(III). However, the use of oxidants and the use of bacteria have concerns for environment and maintenance respectively. Thus, we focused on the ultrasound oxidation to oxidize As(III) to As(V). The oxidation effect of radicals generated by ultrasound irradiation has already been used for As(III). In our previous study, we tried to remove arsenious acid from the solution using ultrasound oxidation and iron hydroxide. [1)] As(III) was easily oxidized with ultrasound. However, pH value of the solution became low due to the generation of nitric acid under air atmosphere. Iron hydroxide was dissolved the solution irradiated by ultrasound, and As(V) was released from the surface of iron hydroxide. In the argon and oxygen atmosphere, we could remove arsenic using iron hydroxide because pH value of the irradiated solution was stayed at neutral. Thus, we investigated trivalent iron compounds witch solve little in acidic solution (pH=2). In this study, goethite was used as a trivalent iron compound for the removal of arsenic because it did not dissolve in acidic aqueous solution and did not break down by ultrasound irradiation. We performed the arsenic removal in As(III) and As(V) acidic solution using goethite and ultrasound oxidation process.

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2. Experimental

As_2O_3 and $Na_2HAsO_4 \cdot 7H_2O$ were used to make solutions of As(III) and As(V) (10 ppm). H_2SO_4 was used for pH adjustment. Ion-exchange water was used for all experiments. A synthesized goethite (FeO(OH)) was used to remove arsenic oxide and arsenous oxide from solutions. The schematic diagram of the experimental apparatus is shown in **Fig.1**. The sonication was performed with an ultrasonic generator (Kaijo TA-4021) and a submersible transducer (Kaijo, diameter 65mm). The output and the frequency of these devices were adjusted to 200 W and 200 kHz respectively. The temperature of the irradiated solution was controlled at about 20 °C using a cooling system.

Arsenic removal experiments were conducted as follows. First, we confirmed the removal ratio of As(III) or As(V) in the solution using goethite and a stirrer without using ultrasound. Goethite(200 mg) was added to both As(III) and As(V) solution (10 ppm, 100 ml, pH 2.2) and magnetic stirrers were used at 400 rpm for 30 min to stir these solutions. The precipitates from the above process were filtered through a 0.45 μm membrane filter. The residual As concentration in the filtered solution was measured using an inductively coupled plasmaoptical emission spectroscopy (ICP-OES) instrument and the removal ratio of arsenic was calculated from the results. Second, we performed As(III) removal using goethite and ultrasound irradiation. As(III) solution (10 ppm, 100 ml, pH 2.2) was sonicated for 30 min, followed by the addition of goethite and stir with the same condition as control experiment described above (30 min, 400 rpm). Arsenic removal rate was evaluated using ICP-OES.

3. Results and Discussion

Figure 2 shows removal ration of arsenic in the As(III) and As(V) solutions treated with goethite and stirring without ultrasound irradiation. The As removal ratio in As(V) solution showed 96.9% as opposed to only 26.3% in As(III) solution. Thus, As(III) was more difficult to remove compared to As(V) using the goethite with stirrer. In the case of using goethite and ultrasound for As(III) solution, As removal ratio showed 89.4%. As removal ratio in As(III) using goethite and ultrasound irradiation was higher than that using goethite and stirring, reflecting the difference in the amount of As(V) oxidized by ultrasound irradiation or stirring.

Figure 3 shows the arsenic removal ratio in the As(III) solution using goethite addition before and after ultrasound irradiation. We changed the order of ultrasound irradiation and goethite addition to treat the acidic solution including As(III) and evaluated the two treatment order:(1) the addition of the goethite after ultrasound irradiation for 30min, (2) the addition of the goethite before ultrasound irradiation (30min). Method (1) showed the arsenic removal ratio of 89.4% wheres method (2) showed 82.4%. Therefore, addition of goethite after ultrasound irradiation is better than that before ultrasound irradiation. This different As(III) removal ratio may reflect the difference in their ultrasound power reached to solutions. We deduced that the ultrasound was disturbed by the added goethite and reduced oxidation radicals and H_2O_2. For future study, we will perform As removal in acidic solution including As(III) using goethite and ultrasound by varying sonication conditions..

4. Conclusion

As(III) was more difficult to remove compared to As(V) in the acidic solution using the goethite and stirrer. Using ultrasound irradiation and goethite to remove As(III) in the acidic solution, the removal rate was increased because As(III) was oxidized to As(V) by generated oxidants We also evaluated arsenic removal ratio in the As(III) acidic solution with different treatment orders. Ultrasound irradiation followed by the addition of goethite showed higher As removal ratio than the addition of goethite followed by ultrasound irradiation.

Acknowledgment

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Reference

1. R. Hosokawa, T. Saito, and H. Okawa: Jpn. J. Appl. Phys. 50 (2011)07HE11.

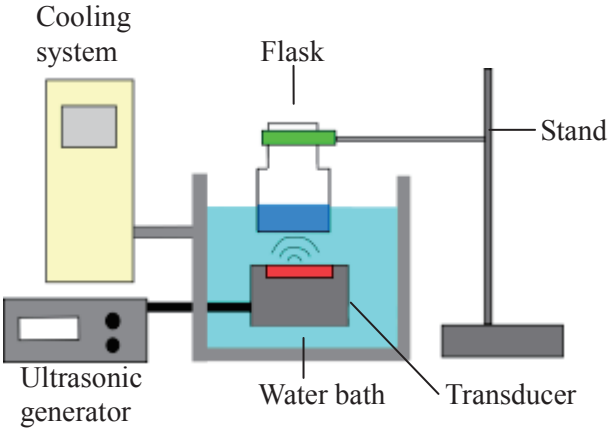

Fig.1 Schematic design of the experimental apparatus

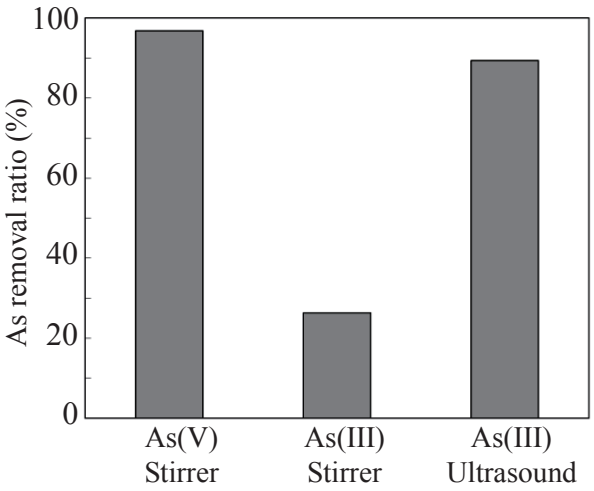

Fig.2 Removal ratio of As in As(III) and As(V) solution using goethite with stirring or ultrasound irradiation

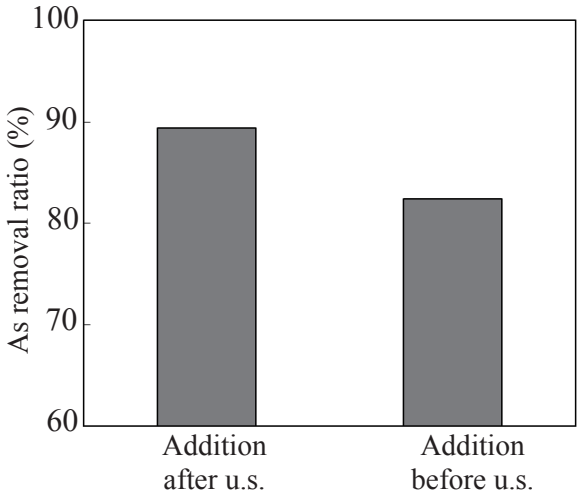

Fig.3 Removal ratio of As in As(III) solution using goethite addition after and before ultrasound irradiation