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Improvement in efficiency of iron leaching from green tuff using sonication and oxalic acid

超音波補助によるシュウ酸を用いた緑色凝灰岩からの鉄抽出効率の改善

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1. Introduction

Quarry waste generated from quarrying and cutting of stone is not ideal for use as an building materials as it consists of irregularly crushed rocks and fine particles [1-3]. One effective use of quarry waste is to extract raw iron by leaching from the stone. Lee et al. extracted iron from ore using oxalic acid and a mechanical stirrer [4]. According to their report, the extraction of iron from ore is highly temperature dependent. In this study, we focused on the physics of ultrasound irradiation to enhance the amount of iron leached from green tuff quarry [5, 6]. We discuss here, the difference between leaching efficiency obtained using ultrasound irradiation and that using mechanical stirring.

2. Experimental

An ultrasonic generator (TA-4021: Kaijo, Japan) produced ultrasonic waves at a frequency of 200 kHz, with an output power of 200 W. An oscillator was put in a tank filled with water, and a flask containing an experimental solution was placed immediately above the oscillator. The temperature of the irradiated solution was controlled by water circulating around the flask. Green tuff (Akita, Japan) was chosen as the sample for iron leaching. The chemical composition of green tuff quarried in Akita, Japan was shown in **Table I**. The diameter range of the sample was 1 to 2.83 mm. Iron-leaching solution containing 10 g green tuff and 0.4 mol/l oxalic acid was prepared. Leaching was conducted by ultrasound irradiation or mechanical stirring for 1 and 3 h, respectively, at a solution temperature of 90 °C. Leaching efficiencies were evaluated from the concentration of iron ions found in the filtered solutions following the ultrasound irradiation or mechanical stirring, as measured by inductively -coupled plasma emission spectrometry (SPS5510). A schematic diagram of the experimental apparatus is shown **Fig. 1**.

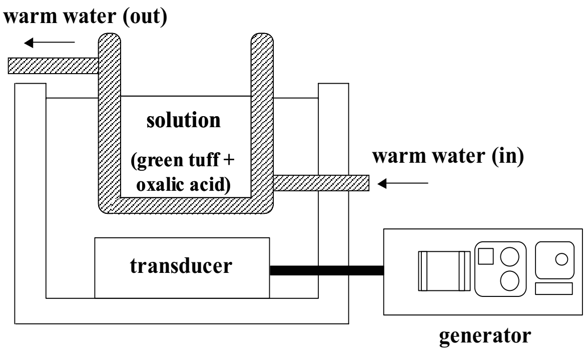

Fig. 1 Schematic diagram of the experimental apparatus

Table I Chemical composition of Green tuff (Akita, Japan)

SiO_2	Al_2O_3	Na_2O	Fe_2O_3	
72.1	12.7	4.10	3.23	
K_2O	CaO	MgO	TiO_2	Residual
2.30	1.30	1.00	0.49	2.78

Unit : wt%

Table II Amount of iron leached from green tuff by mechanical stirring process

Temperature (°C)	Process	Fe (ppm)
20	Stirring + Oxalic acid	56.53
90	Stirring	0.45
90	Stirring + Oxalic acid	257.87

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3. Results and Discussion

Table II shows the amounts of iron leached from green tuff by mechanical stirring under various conditions (treatment temperatures are 20 °C or 90 °C, oxalic acid was added in two of three instances).We were able to confirm that the amount of iron leached from green tuff using oxalic acid increased at high temperature. These findings are similar to the results of Lee et al. [4]. Moreover, we confirmed that the leaching of iron from green tuff was difficult in the absence of oxalic acid. **Fig. 2** shows the amount of iron leached from green tuff when treated by ultrasound irradiation and by stirring, at 90 °C. Comparing the amount of iron leached using the two techniques, ultrasound treatment leached higher amounts than stirring for the same treatment time. Oxalic acid reacted with iron ions and deposited iron oxalate on the surface of the stone. The leaching efficiency of the iron reduces when the surface of the stone is covered with the deposited iron oxalate. It is thought that jet flow caused by the collapse of cavities during ultrasound irradiation prevents and strips the deposits of iron oxalate. A large number of jet flows generate very rapid flow at local areas in the solution, a phenomenon that enhances the amount of iron leached. The second reason for the increase is thought to be that ultrasound irradiation breaks down the green tuff into fine particles. As a result, the total surface area of green tuff increases and it comes into contact more easily with hot water, which functions as a solvent of iron ions from the green tuff. Changes in particle size distribution were measured using 5 μm grain (average size) after ultrasound irradiation and mechanical stirring for 3 h; the results are shown in **Fig. 3**. The particle diameter generated by ultrasound irradiation was smaller than that generated by mechanical stirring. It is thought that the increased amount of iron leached by ultrasound irradiation is due to the formation of finer particles.

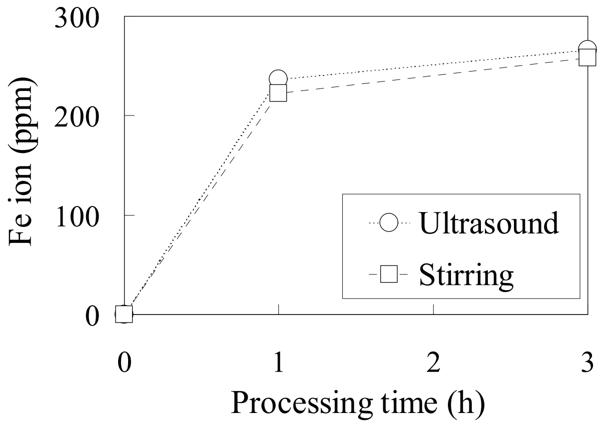

Fig. 2 Variations in the amount of iron leached from green tuff into the oxalic acid solution treated by ultrasound irradiation or mechanical stirring for 3 h at 90 °C

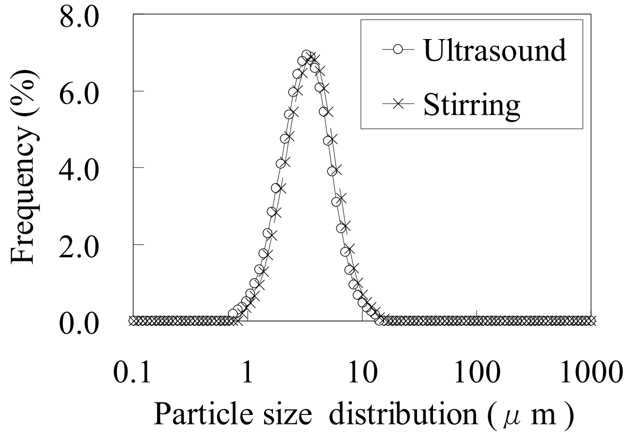

Fig. 3 Particle-size distribution of green tuff generated by ultrasound irradiation and by mechanic stirring

4. Conclusions

This study of the leaching of iron from green tuff was conducted using ultrasound irradiation and oxalic acid. Ultrasound irradiation contributed to an increase in the amount of iron leached from tuff. In future studies, we plan to use low-frequency ultrasound, which we expect shall result in finer particles, ultimately resulting in even greater amounts of iron leached.

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