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Elevated temperature on leaching of arsenic and boron from excavated rocks

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ABSTRACT: This work evaluated the leaching behaviors of geogenic arsenic and boron of two excavated rocks under different temperatures. Excavated rocks with geogenic contamination are expected to be used in embankments with proper countermeasures. Their leaching behaviors might change because of seasonal and daily changes in ground temperature. However, the effect of temperature on leaching behaviors has not been well examined. Column tests at temperatures of 20 and 40°C were performed using rock samples. Elevated temperatures led to the release of greater amounts of arsenic and boron. The largest arsenic concentrations obtained from the column tests were 0.53 mg/L at 40°C, while at 20°C was 0.15 mg/L for one rock sample. The largest concentrations of arsenic and boron obtained at 40°C were at most 3.5 times larger than at 20°C. Boron concentrations decreased as electric conductivity decreased. Electric conductivity can be one index to predict the leaching behavior of boron.

1 INTRODUCTION

Large volumes of excavated soils and rocks were generated from construction projects. As a result, a certain percentage of the soils and rocks contain contaminants derived from geological processes. Geogenic contaminants, such as arsenic (As) or boron (B), are widely distributed in Japan (e.g., Tabelin et al., 2018). Such soils and rocks are expected to be used in embankments as a part of ongoing efforts to achieve sustainable soil management. However, their proper utilization is sometimes a concern, especially because contaminants can be released from the soils and rocks. Given the importance of developing economical and effective utilization methods for geogenically contaminated soils and rocks, evaluating the leaching behaviors is essential.

When excavated materials are utilized in embankments or other shallow geostructures, the effect of ground temperature on the leaching behavior of toxic elements becomes a technical concern. The daily and seasonal temperature changes in the shallow ground can be much more significant than those in the deep ground. Temperature near the ground surface can be elevated because of solar radiation. The ground temperature was often more than 30°C (Menberg et al., 2013). However, laboratory leaching tests are usually conducted at room temperature (approximately 20°C). Since leaching behaviors of geogenic contamination can be affected due to thermo-mechanical-chemical interactions, the effects of elevated temperature on leaching behaviors need to be clarified from a geoenvironmental perspective.

This work evaluated the leaching behaviors of As and B from two excavated rocks with geogenic contamination using column tests under different temperatures. Column tests were less applied to evaluate the leaching behaviors of geogenic contaminants under different temperatures, while several researchers investigated using batch leaching tests (Saito et al., 2016; Takai et al., 2020). Although batch tests have advantages in simple experimental protocol with a short testing time, the applicability of this test may be limited. Since the solute kinetics under the flow conditions can be evaluated, column tests can evaluate closer to the in-field condition than batch tests (Kato et al., 2021).

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2 MATERIALS AND METHODOLOGY

2.1 *Materials*

Two types of rocks used in this work were excavated from tunnel construction sites. The rocks were crushed to particles smaller than 2 mm for the experiments. The rock samples were stored in sealed plastic bags to prevent their oxidation by air. Table 1 highlights the fundamental physical properties of the rock samples. In addition, the total amount of As and chemical compositions of the rock samples were measured. The rock samples were air-dried at room temperature and ground into a powder with a diameter smaller than 75 μm. The samples were evaluated by X-ray fluorescence (XRF) analysis using a Shimadzu EDX 720 energy-dispersive X-ray spectrometer.

Table 1. Physical and chemical properties of rock specimens.

Parameter	Rock 1	Rock 2
Particle density, ρ_s	2.686 g/cm^3	2.749 g/cm^3
Water content, w	1.4%	4.7%
Particle size distribution		
Sand fraction [0.075–2 mm]	83%	62%
Fines fraction [<0.075 mm]	17%	38%
Average particle size	0.85 mm	0.25 mm
As content	51 mg/kg	87 mg/kg

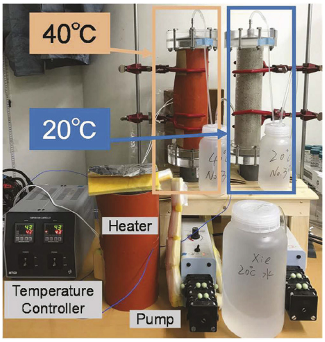

Figure 1. Setup of the column test.

2.2 *Column tests with temperature control system*

Column tests were conducted using acrylic columns (ϕ 5 cm × h 30 cm) as shown in Figure 1. A rock sample was placed in the column. Each specimen was compacted in the acrylic column in five layers with equal heights. A 125-g rammer was dropped freely from a height of 20 cm during compaction. This method was based on the corresponding ISO 21268-3 (2019). The specimen was placed between filter papers to prevent channel clogging due to fine soil particles. The dry density of the specimen for Rocks 1 and 2 were approximately 1.3 and 1.4 g/cm^3, respectively. Then distilled water was percolated in an up-flow direction using a peristaltic pump at a flow rate of approximately 15 mL/h until the specimen reached saturation. Finally, percolation was interrupted for 15 hours to achieve a saturated condition. After 15 hours, distilled water was percolated in an up-flow direction via a peristaltic pump at 15 mL/h. Leachate was collected from the outlet of the column. Column tests were conducted until the liquid-to-solid ratio (a volume of permeated solution to a dry mass of soil ratio) was larger than 10.

Temperatures of 20 and 40°C were applied to account for possible temperature changes (Menberg et al., 2013). Tests under 20°C were conducted in a constant temperature room, while 40°C were conducted by wrapping the column with rubber heaters. The temperature of the rubber heater was controlled using a controller (MTCD, Misumi Corporation). Distilled water was pre-heated to 40°C using a constant temperature bath. Tubes connected to the column were covered by glass wool (DK-089, Yamawaki Sangyo) to prevent heat dissipation. A non-contact thermometer measured the temperature of the column surface to confirm whether 40°C was achieved.

For leachate, centrifugation at 3000 rpm for 10 min and filtration with a 0.45 μm membrane filter was conducted. The filtrate electric conductivity (EC) and pH were measured using a pH/EC meter (Horiba F-54). Arsenic concentration was measured using an atomic absorption spectrophotometer (Shimadzu GFA-7000). Boron concentration was measured using an inductively coupled plasma optical emission spectrometer (Agilent Technologies ICP-OES 710).

3 RESULTS AND DISCUSSION

3.1 *Arsenic concentration and pH*

Figure 2 shows the profiles of As concentrations with pore volumes of flow (PVF) obtained from column tests. Since PVF is calculated as the volume of a permeated solution divided by a void volume, 1 PVF means one pore water is exchanged. As shown in Figure 2(a), the largest As concentration at 40°C was 0.53 mg/L, while at 20°C was 0.15 mg/L. The largest concentration at 40°C was approximately 3.5 times greater than at 20°C. As shown in Figure 2(b), As concentrations also once increased and decreased at both 40 and 20°C. The largest concentration at 40°C was 3.94 mg/L, while at 20°C was 3.19 mg/L. The largest concentration at 40°C was approximately 1.2 times greater than that at 20°C. Results show that leaching As concentration might increase as the temperature raised.

Herein, As concentrations at 1 PVF is discussed. Arsenic concentrations once increased and decreased at both 40 and 20°C. If the leaching concentration drastically decreased before 1 PVF, most contaminants flowed out as one pore water was replaced, and the leaching was considered to diminish immediately. However, even though PVF > 10, As was leached at more than 0.1 mg/L, which was 10 times larger than the regulatory limit in Japan for both rock samples. These results imply that As leching might continue relatively longer. For both Rock 1 and 2, the largest leaching concentrations were obtained after 1 PVF. Shapes of the concentration profile might not be different, but the value of the concentration might be greater at elevated temperatures.

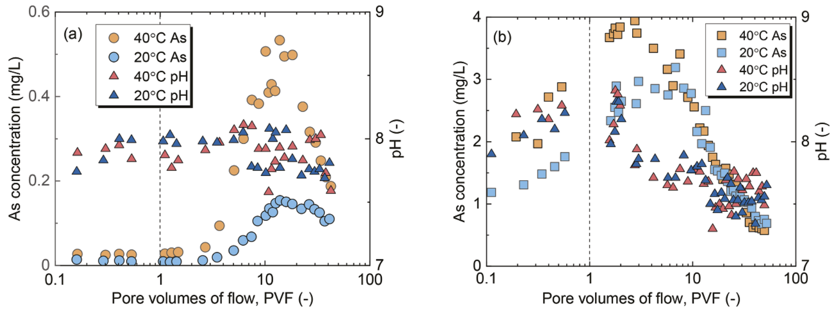

Figure 2. Arsenic concentrations and pH obtained from (a) Rock 1 and (b) Rock 2.

Figure 2 shows the profiles of pH with PVF. Alkaline pH between 7-9 was obtained from both rock samples. As shown in Figure 2(a), similar pH values were obtained regardless of temperatures, while As concentrations differed. The elevated temperature might affect not pH but As leaching behavior. According to XRF analysis, both rock samples contained some amounts of sulfide minerals. More acidic pH values should be obtained if the dissolution of sulfide minerals is promoted under elevated temperatures. However, such trends cannot be observed. Since geogenic As is usually contained in the sulfide minerals such as pyrite, dissolution of sulfide minerals is considered one of the major mechanisms of As leaching (Tabelin et al., 2018). The leaching behavior of As from these rock samples might not be related to the dissolution of sulfide minerals. In future work, the mineral composition of the rock samples and the state of the surface charge should be analyzed to investigate the leaching mechanisms of As affected by elevated temperature.

3.2 *Boron concentration and EC*

Figure 3 shows the profiles of B concentrations with PVF. Boron concentrations generally decreased as PVF increased at both 40 and 20°C. For Rock 2, the largest B concentration at 40°C was 0.14 mg/L, while at 20°C was 0.079 mg/L. The largest concentration at 40°C was approximately 1.8 times greater than at 20°C. Results show that leaching B concentration might increase as the temperature raised. In contrast to As, the largest B concentrations were obtained before 1 PVF for both rock samples. Boron leaching might diminish more immediately than As. For B contamination, even if column tests are conducted in a short period (e.g., < 1 PVF), enough to evaluate the leaching behavior because the largest concentration can be measured.

Figure 4 shows the profiles of EC vs. B concentrations. Electric conductivity decreased drastically before 1 PVF. Generally, B concentrations decreased as EC decreased. These results mean most ions in the pore water were drained by 1 PVF. Therefore, most B might be leached with other ions as one pore water was replaced. Since measuring EC is much easier than B concentration, the EC value can be helpful indices to predict B leaching behavior approximately.

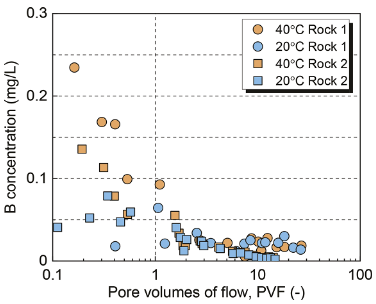

Figure 3. Boron concentrations.

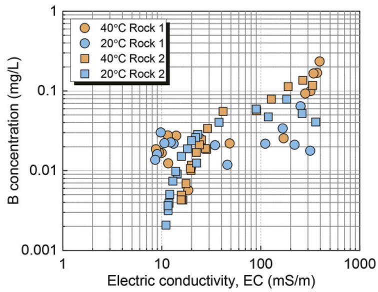

Figure 4. EC vs boron concentration.

3.3 *Geotechnical considerations for leaching behaviors under elevated temperature*

As shown in Figures 2 and 3, the largest concentrations of As and B at 40°C were at most 3.5 times larger than at 20°C. Elevated temperatures led to the release of greater amounts of As and B, but the increase was not drastic. Leaching concentration obtained from column tests is used as the boundary condition when solute transport analysis is conducted to evaluate the contamination. Since the in-situ ground temperature used by excavated materials is not always at 40°C but sometimes below 20°C, column tests conducted at 40°C may be safe side. If the solute transport analysis is performed using the As or B concentration supposed to be several times larger than obtained from the tests at room temperature, a safe prediction may conduct considering the effects of elevated temperature.

4 CONCLUSIONS

The leaching behaviors of As and B from two excavated rocks with geogenic contamination were investigated under elevated temperatures. Herein, column tests were performed at 20 and 40°C. The results support the following conclusions.

Elevated temperatures led to the release of greater amounts of As and B. The largest As concentrations obtained from the column tests were 0.53 mg/L at 40°C, while at 20°C was 0.15 mg/L for one rock sample. The largest concentrations of As and B obtained from the column tests at 40°C were at most 3.5 times larger than at 20°C. Boron concentrations decreased as EC decreased. Since measuring EC is much easier than B concentration, the EC value can be helpful indices to predict the leaching behavior of B.

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