



# A microfluidic approach to water-rock interactions using thin rock sections: Pb and U sorption onto thin shale and granite sections



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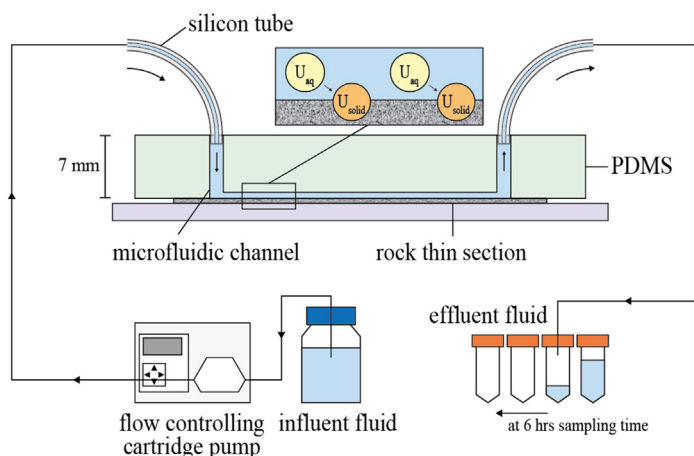
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## HIGHLIGHTS

- Microfluidic tests were used to investigate water-rock (mineral) interactions.
- Pb and U sorption onto thin shale and granite sections was evaluated.
- Pb removal by thin shale section is related primarily to Fe-containing minerals.
- A slightly larger amount of U was removed onto the thin granite section with Fe-containing minerals.

## GRAPHICAL ABSTRACT



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## ABSTRACT

The feasibility of using microfluidic tests to investigate water-rock (mineral) interactions in fractures regarding sorption onto thin rock sections (i.e., shale and granite) of lead (Pb) and uranium (U) was evaluated using a synthetic  $\text{PbCl}_2$  solution and uranium-containing natural groundwater as fluids. Effluent composition and element distribution on the thin rock sections before and after microfluidic testing were analyzed. Most Pb removal ( $9.8 \text{ mg/cm}^2$ ) occurred within 3.5 h (140 PVF), which was 74% of the total Pb removal ( $13.2 \text{ mg/cm}^2$ ) at the end of testing (14.5 h, 560 PVF). Element composition on the thin shale sections determined by  $\mu\text{-XRF}$  analysis indicated that Pb removal was related primarily to Fe-containing minerals (e.g., pyrite). Two thin granite sections (biotite rich, Bt-R and biotite poor, Bt-P) exhibited no marked difference in uranium removal capacity, but a slightly higher amount of uranium was removed onto the thin Bt-R section ( $266 \text{ }\mu\text{g/cm}^2$ ) than the thin Bt-P section ( $240 \text{ }\mu\text{g/cm}^2$ ) within 120 h (4800 PVF). However, uranium could not be detected by micro X-ray fluorescence ( $\mu\text{-XRF}$ ) analysis, likely due to the detection limit. These results suggest that microfluidic testing on thin rock sections enables quantitative evaluation of rock (mineral)-water interactions at the micro-fracture or pore scale.

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## 1. Introduction

Safe disposal of radioactive waste is a crucial global issue, because nuclear power plants produce potentially extremely haz-

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ardous wastes (e.g., spent fuel), despite requiring only a small amount of radioactive material. Among the various types of radioactive waste repository—space, seabed, ice cap, and underground disposal [1]—underground geological disposal is considered the best approach in most countries.

Crystalline rocks (e.g., granite) are the preferred host rocks for the underground geological disposal of radioactive waste [2]. Fractures developed in crystalline rocks act as flow paths for groundwater. As groundwater flows through fractures, groundwater interacts with adjacent fracture-filling materials and host rocks. Water-rock interactions affect the transport of radionuclides, such as uranium (U), cesium (Cs), and strontium (Sr), which may be released from radioactive waste repositories. When radionuclides in groundwater migrate through fractures in crystalline rocks, their mobility is affected by interactions between the groundwater and geologic media.

Interactions within the fracture system involve diverse processes, for example, ion exchange, precipitation, oxidation/reduction, and surface complexation [3]. The sorption, diffusion, and reduction processes occur at the water-rock (mineral) interface in the fracture system, resulting in retardation of contaminants in groundwater [4]. Transport of radionuclides can be retarded by sorption onto the surface of geologic media [5]. The minerals that comprise adjacent rocks can play important roles in radionuclide sorption.

Batch-type experiments have been widely used for radionuclide sorption tests, using various powdered materials [4–8]. However, a few radionuclide sorption experiments have been performed that used thin rock sections [9,10]. In previous studies, thin rock sections were placed in solutions of radionuclides under stationary conditions for a designated period, as a batch-type test. For example, Ticknor et al. [9] investigated the sorption of radionuclides ( $^{90}\text{Sr}$ ,  $^{137}\text{Cs}$ ,  $^{144}\text{Ce}$ ,  $^{241}\text{Am}$ ,  $^{75}\text{Se}$ , and  $^{95\text{m}}\text{Tc}$ ) onto thin sections from the core rocks of fracture zones. The sorption capacities of the radionuclides onto various minerals (chlorite, hornblende, biotite, laumontite, kaolinite, orthopyroxene, quartz, and altered plagioclase) were evaluated. Hornblende and biotite had high affinity for most of the radionuclides, except selenium, which exhibited marked sorption to chlorite.

The technologies of “microfluidics”, or “lab-on-a-chip (LOC)”, appeared at the beginning of the 1980s [11]. Microfluidics tests have been applied in biochemistry, biotechnology, nanotechnology, and energy engineering [12–16]. Microfluidic tests have many advantages compared to other methods: (1) experiments can be miniaturized, thus less space is needed, (2) instruments are cost-effective for production, and portable, (3) the flow condition can be easily controlled using a pump as a power source, and (4) reactions can be observed in small-scale microfluidic channels [12].

However, few studies have investigated water-rock (mineral) interactions using microfluidic tests. In nature, contaminant retardation in fracture zones involves adjacent fracture-filling materials or host rocks, and groundwater flowing through fractures. Microfluidic tests facilitate simulation of the reactions between rocks (minerals) and fluid, as the flow conditions can be controlled, and there is contact between the fluid and reactant.

The primary objective of this study was to evaluate the feasibility of a microfluidic test to investigate water-rock (mineral) interactions using thin rock sections. To mimic water-rock interactions in fracture zones, microfluidic tests were conducted using thin rock sections (i.e., shale and granite). A synthetic  $\text{PbCl}_2$  solution and U-containing natural groundwater were used as permeants. The effluent composition and element distribution on the thin rock sections obtained before and after microfluidic testing were analyzed by inductively coupled plasma-atomic emission spectrometry (ICP-AES), inductively coupled plasma-mass spectrometry (ICP-MS), and micro X-ray fluorescence ( $\mu$ -XRF). Microfluidic tests can more

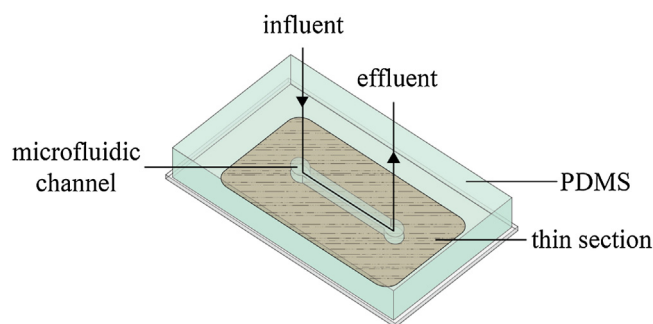


Fig. 1. A schematic diagram of PDMS with a micro-channel placed on a thin rock section.

closely mimic water-rock (mineral) interactions, and  $\mu$ -XRF can be used to analyze the mineral surface after interactions with fluid, without disturbance of the sample. Furthermore, flow experiments on a small scale can be performed, as can the continuous reaction between groundwater and rocks (minerals) within very narrow flow paths, resulting in long-term interactions.

## 2. Experimental

### 2.1. Materials

Shale and granite were used to prepare thin rock sections to evaluate the feasibility of the microfluidic sorption test. Shale, which generally has a high clay mineral content, was selected to evaluate Pb sorption. Previous studies have reported that clay minerals are effective for the uptake of Pb [17–19].

Granite was used to evaluate the U sorption capacity. The Korea Atomic Energy Research Institute (KAERI) constructed the KAERI Underground Research Tunnel (KURT) in Daejeon, South Korea in 2006 to develop site characterization technologies for the underground disposal of radioactive waste [20]. Host rocks near the KURT are composed of Mesozoic two-mica granite, and unknown-age schistose granite intruded by two-mica granite [20]. The core rock was collected from a borehole at the KURT site. Two thin rock sections (i.e., biotite poor, Bt-P; and biotite rich, Bt-R) derived from the core rock samples obtained from the fracture zone of the KURT site were used to evaluate the effect of the presence of biotite in granite on U sorption.

A synthetic  $\text{PbCl}_2$  solution and U-containing natural groundwater were used as permeants in the microfluidic tests. The  $\text{PbCl}_2$  solution ( $7.42 \times 10^{-3} \text{ M}$ ) was prepared by dissolving analytical-grade  $\text{PbCl}_2$  powder (Sigma-Aldrich Co.) in deionized (DI) water. The initial pH of the  $\text{PbCl}_2$  solution was 5.0. The groundwater sample was collected from a borehole at the KURT site in Daejeon, South Korea, and filtered through a  $0.45 \mu\text{m}$  membrane filter before chemical analysis.

### 2.2. Methods

#### 2.2.1. Microfluidic tests

A microfluidic chip with a micro-channel (width =  $3000 \mu\text{m}$ , length =  $20 \mu\text{m}$ , and height =  $2500 \mu\text{m}$ ) was manufactured from polydimethylsiloxane (PDMS). The PDMS was prepared with Sylgard 184 silicone elastomer base and a curing agent at a 10:1 weight ratio [21]. The PDMS chip was placed on the thin rock section (Fig. 1). To prevent fluid leaking from the flow channel through gaps between the PDMS chip and the thin rock section, the PDMS chip and the thin rock section were pressed upward and downward, respectively, using an acrylic board fixed with screws. The procedure of the microfluidic test was described in detail in Oh [22]. The

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