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Copper Effects on the Retention of Mobile Iodine in a Compacted Bentonite

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1. Introduction

The spent fuel derived from the nuclear reactor facilities may be finally disposed in a deep underground below 500 m [1]. It mainly consists of uranium and also has iodine and technetium in minority, which are typical anionic radionuclides. In particular, radioiodine has higher mobility from its spent fuel source. It has been well known that it could freely pass through a compacted bentonite that is one of underground engineering barriers that are used to retard some nuclides' migration from the spent fuel. We installed a small laboratory apparatus that was similar to the underground repository by using an anaerobic glove box and evaluated an iodine mobility in the compacted bentonite with or without copper. The copper-bentonites were prepared in two types, a copper ion-exchanged form and a copper nanoparticle-mixed one. In our study, we additionally tried to find an effect of sulfate to retard mobile iodine from the compacted bentonite for a long-term period.

2. Methods and Results

2.1 Methods

We made a small repository module (Fig. 1) by using a granite column with 5 cm height and 5.2 cm diameter that was cut around the center, where the inside part was also excavated in a dimension of 2 cm height and 3 cm diameter as a purpose for putting a compacted bentonite into it later on. The granite used was a rock boring core obtained from the KURT (Kaeri Underground Research Tunnel) site. The bentonite used as a compacted buffer material was from Kyongju, Gyeonsang province. It was prepared by exchanging the interlayer cations with copper(II) ions as much as 1 CEC (cation exchange capacity) or by mixing copper nanoparticles (60~80 nm) with it in 1:9 ratio. The prepared bentonite samples were compacted by a density of 1.6 g/cm^3 in column (2 cm height and 3 cm diameter), where iodine powder, NaI (45 mg), was put into the center as a replica of radioiodine in the spent fuel waste. The prepared granite + compacted bentonite modules were immersed into plastic containers with an anaerobic solution containing $NaHCO_3$ (1 mM), Na_2SO_4 (2 mM), and Na-lactate (10 mM). The whole apparatus was placed in the glove box filled with N_2 gas. Some aqueous solution was periodically sampled from the plastic containers and measured for pH, Eh, and dissolved ions such as iodine during several months. After the experiment, the solid bentonite sample was cut and examined for the release of iodine using microprobe analyzer.

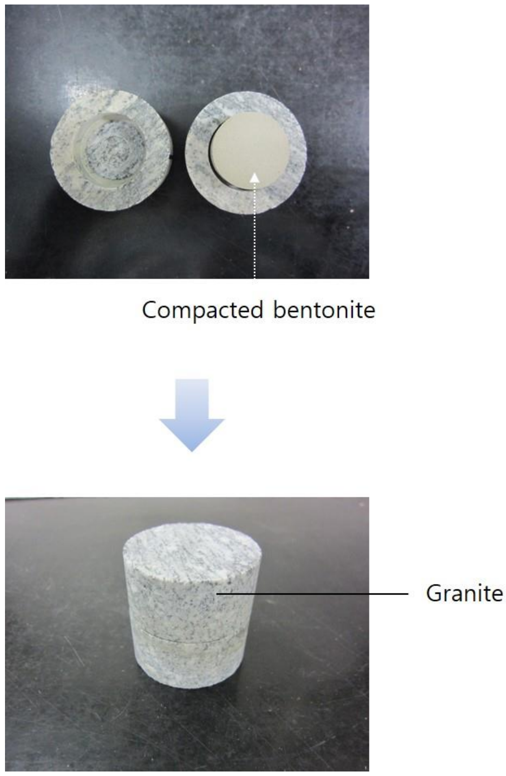

Fig. 1. A small module of KURT granite + compacted bentonite with iodine (center) was used in our experiment, which conceptually represents a specific radwaste geological repository.

2.2 Iodine Release from the Compacted Bentonite

The solution pH and Eh were initially 6.5 and 190 mV, respectively, exhibiting a little acidic and anaerobic condition. The initial pH value was little changed, but a significant change for Eh was observed for the case of presence of aqueous sulfate, displaying its decline from 190 to 74 mV during 5 months.

The iodine powder in the compacted raw bentonite was gradually dissolved and released to the surrounding solution as it became saturated with water. Its release rate was initially fast and then became slow with time in a buffer solution of $NaHCO_3$ alone. However, in a specific condition both with Cu-exchanged bentonite and aqueous sulfate, the releasing rate of iodine was relatively slower and nearly zero at the late stage (Fig.

2). It means that the leaching of iodine from the compacted bentonite was strongly affected by both the copper component and the aqueous sulfate. The sulfate can be reduced to sulfide by SRB (sulfate-reducing bacteria) living in the bentonite, probably affecting the iodine diffusion and migration in the system. In general, bentonite could have some indigenous bacteria including SRB [2], which have been recognized as regulating agents to be able to change the geochemical properties in the bentonite.

For a Cu nanoparticle-mixed bentonite, the iodine release from the compacted bentonite was rarely detected. Most soluble iodine ions in the compacted bentonite were transformed to a solid phase, which was identified by X-ray diffractometer and electron microscopy. The iodine in the solid phase was mostly coexisted with copper component in the bentonite. It means that in a specific condition the highly mobile iodine can be effectively immobilized by using copper additives.

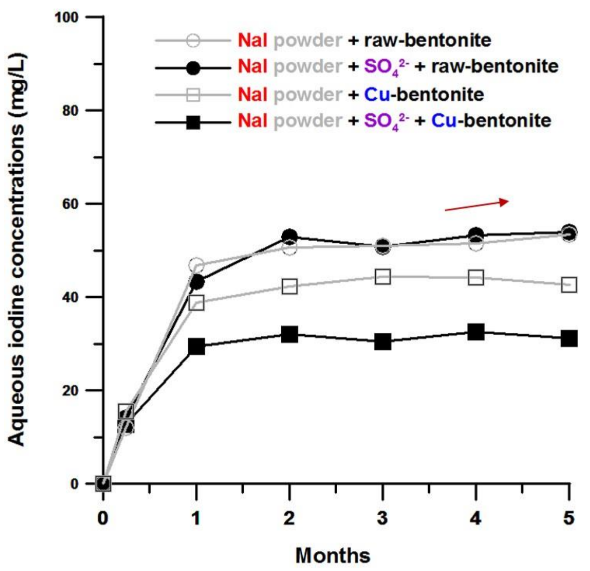

Fig. 2. Releases of iodine from the compacted bentonites with or without copper in them and aqueous sulfate in solution.

3. Conclusions

We found an effective way to limit the iodine release from the compacted bentonite. This condition can be achievable by exchanging the bentonite interlayer cations with copper ions or by mixing copper nanoparticles with bentonite. In those cases, soluble iodine can be easily immobilized as a solid phase by combining with copper component via the role of indigenous SRB in bentonite.

REFERENCES

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