Complex Behavior of Radionuclides in a Disposal Environment

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1. Introduction

A study on the complex behavior of radionuclides in a deep geological disposal environment for radioactive wastes is necessary to predict their migration and retardation. A disposal system for radioactive waste was mainly consists of a disposal canister, buffer/backfill material and natural barrier. It is generally known that the geochemical behavior between radionuclides and the components of disposal system is a main retardation process of radionuclide migration in a deep geological environment. The geochemical reaction of radionuclides is highly affected by various geochemical conditions such as pH, redox potential, ionic strength, microbe and ionizing radiation.

This work is focused on the reaction between radionuclides and corrosion products, the alteration of bentonite and its effect on the sorption/diffusion of radionuclides, the microbial effect on the migration of radionuclides and the combined reaction of radiation and microbe.

2. Materials and Methods

2.1 Materials

Copper disks (99.90% Cu), Kyungju bentonite, KURT granite and goethite were used as a component of a geological disposal of radioactive wastes. All the solutions were prepared with deoxygenated ultrapure water with a resistivity of 18.3 MΩ/cm (Milli-Q system, Millipore) under anaerobic conditions in a glove box filled with Ar.

2.2 Methods

Gas generation was measured by gas chromatography (7890A, Agilent) using a packed column and thermal conductivity detector. The composition and morphology of corrosion products on copper specimen was analyzed using XRD and SEM. XRD patterns were taken using Cu-Kα radiation and a Rigaku D/MAX 2200 Ultima X-ray diffractometer. The surface morphology was observed using FE-SEM (JSM-7000F, JEOL) and the chemical composition of corrosion products formed on the copper specimen were analyzed using Energy Dispersive X-ray analysis (EDS). Copper in aqueous solution dissolved from the copper sample was measured using ICP-MS (Ultramass 700, Varian).

3. Results

The distance of the interlayers of bentonite can be reduced when Cu^{2+} exchanged with ions (Na^{+}, Ca^{2+}) located in the interlayer of the raw bentonite owing to the differences of the hydration energy among Cu^{2+}, Na^{+}, and Ca^{2+}. Consequently, the contraction of the basal plane spacing resulted in the reduction of swelling pressure of bentonite. However, copper exchanged bentonite sorbed uranium and cesium more than the raw bentonite. The exchange of copper with ions of bentonite interlayer could reduce the mobility of radionuclides.

A γ-irradiation increased the H_2 gas generation and the dissolution of copper ions from the metallic copper. The generation of H_2 gas was mainly affected by the total irradiation dose and chloride concentration. After γ-irradiation, cuprite (Cu_2O) was observed as a major corrosion product and very few tenorite (CuO) was also observed. A microbial activity induced the generation of H_2(g) and H_2S(g) and formed the corrosion products of copper and irons as chalcocite(Cu_2S) and mackinawite(FeS) respectively.

The microbes inhabiting in Kyungju bentonite were isolated and identified and the changes in the microbial community existed in fracture filling minerals were observed. Different kinds of sulfate reducing bacterium were identified from Kyungju bentonite and the *Veillonella* genus was observed from the matured fracture fillings. In the presence of bacteria and H_2, the sorption of uranium by Kyungju bentonite and fracture fillings increase as a function of the growth time due to the changes in the microbial community in natural minerals.

Gamma-irradiation did not affect the viability of bacteria and the combined reaction between gamma-irradiation and the microbial activity resulted in the increase of uranium sorption due to the changes in redox environments and forming a strong binding between uranium and newly formed secondary mineral from goethite.

The formation of CuI induced by the microbial activity immobilized the iodide in a copper exchanged bentonite. Microbial activity also reduced the mobility of uranium in the compacted bentonite by the formation of mackinawite with a high sorption affinity to uranium when the sulfate was added.

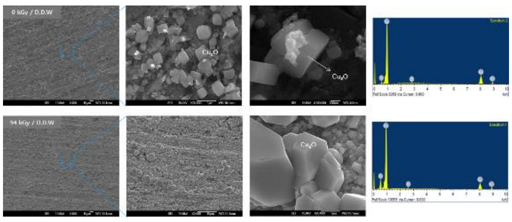

Fig. 1. Effect of γ-irradiation on the formation of copper corrosion products.

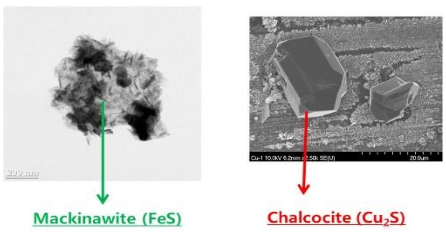

Fig. 2. Effect of microbial activity on the formation of iron/copper corrosion products.

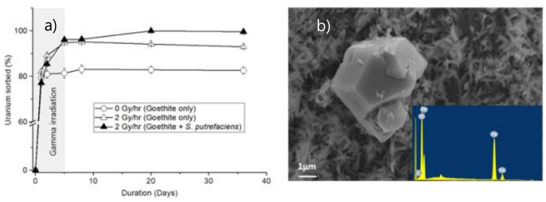

Fig. 3. Effect of microbe and γ--irradiation on U sorption and formation of magnetite.

4. Conclusion

The results of a study on the complex behavior of radionuclides and the relative technologies can be directly utilized for the disposal of the spent fuel generated in Korea. Models and data obtained from this work can be also applied to the development of the safety case. Moreover, the reliability of the safety for the disposal of radionuclides can be enhanced by presenting the scientific evidences on the complex behavior of radionuclides in a deep geological environment to the public.

REFERENCES

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