

Dissolution Characteristics of Uranium From the KURT Granite in Various Geochemical Conditions

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1. Introduction

The studies on long-term behavior of radionuclide have been progressing in the field of radioactive waste disposal. After a long period of radioactive waste disposal, radionuclides released from a radioactive waste repository will be contacted with groundwater and radionuclides will be dissolved by groundwater. Therefore, the study on dissolution characteristics of radionuclide is important part in understanding the long-term behavior of radionuclide in disposal environment.

The objective of this study is to understand dissolution characteristics of uranium from KURT (KAERI Underground Research Tunnel) granite by groundwater in relevant geochemical conditions.

2. Samples and dissolution test

2.1 Samples

Samples for dissolution experiments were obtained from DB-1 core of KURT granite (depth of sampling: 108 ~ 109 m), Granite core samples from KURT were grinded into particles with a size less than 74 μm (200 mesh) and then homogeneously mixed. After washing the samples 5 times, samples and deionized water were mixed in 1 (20 g) : 20 (400 mL) ratio. After the mixed samples were precipitated for 10 minutes by gravitation, suspending turbid water was removed. The washed samples were dried in oven at 60 °C for 48 h and then completely dried at 90 °C for 6 h.

2.2 Leaching test

Seven reaction solutions were prepared considering the relevant geochemical conditions of KURT groundwater in order to confirm the dissolution characteristics by groundwater (Table 1). Samples and seven reaction solutions were reacted in 1 : 10 (5 g : 50 mL) ratio and stirred in an agitator at 100 rpm

continuously. Dissolution tests were carried out during 8 different time steps (0 day, 2 day, 5 day, 1 week, 2 weeks, 1 month, 3 months, 5 months). After the end of reaction periods, pH of reaction solutions were measured and supernatant samples (2.5 mL) were taken and then analyzed by ICP-MS after filtration with 0.22 μm syringe filter (Fig. 1).

Table 1. Scheme and conditions of the dissolution experiments

Code	Solution	What to see
UD-BI (pH 1)	0.1M HNO ₃	Effects of blank solution on U dissolution
UD-Bg (pH 8.8)	0.6mM NaHCO ₃ 0.4mM CaCO ₃ 0.3mM Na ₂ SiO ₃ ·9H ₂ O	Effects of Background solution on U dissolution
UD-CO ₃ (pH 8.8)	0.5 mM NaHCO ₃ 0.5mM CaCO ₃	Effects of Carbonate on U dissolution
UD-Na (pH 8.8)	1mM NaClO ₄	Effects of Na ⁺ on U dissolution
UD-Ca (pH 8.8)	1mM Ca(ClO ₄) ₂	Effects of Ca ²⁺ on U dissolution
UD-CaSi i (pH 8.8)	0.5mM CaCO ₃ 0.5mM NaSiO ₃	Effects of (Ca+SiO ₂) on U dissolution
UD-Si (pH 8.8)	1mM NaSiO ₃	Effects of SiO ₂ on U dissolution

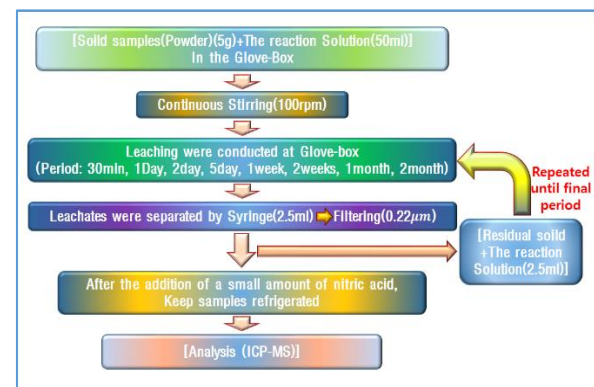


Fig. 1. Leaching test procedure.

3. Results and Discussion

As expected, uranium showed a higher amount of dissolution in the reaction solution (UD-BI). Since uranium is well dissolved in mixed solution with HNO₃[1], the uranium dissolved by HNO₃ shows several tens to hundreds times greater concentration than other solutions in the UD-BI. The dissolved amount of uranium is increased up to 5 day and then decreased after 1 week with a fluent curve. This result may be due to the fact that dissolution of uranium tends to decrease since the dissolution of uranium will be limited by the solubility of uranium in a given condition. The amount of uranium dissolved in other solutions (UD-Bg, UD-CO₃, UD-Na, UD-Ca, UD-CaSi) increases with increasing time although the amount of uranium dissolved is very small (less than 7 ug/L) (Table 2). Because uranium⁺ forms stable complexes with carbonates (CO₃²⁻) [1], the samples UD-Bg and UD-CO₃ show higher amount of dissolved uranium than other reaction solution (UD-Na, UD-Ca, UD-CaSi). But the dissolution rate is decreased as the reaction (contact) time increases because the formation of uranium carbonate complex decreases by decreasing carbonate concentration as the reaction proceeds.

In addition, the dissolved amount of uranium shows a tendency to increase consistently with increasing reaction time in the solutions of UD-Na, UD-Ca, and UD-CaSi although the amount of uranium dissolved is very small. This result implicates that the presence of Na⁺, Ca²⁺, and SiO₂ would not greatly influence on the dissolution of uranium from the KURT granite samples.

Table 2. The concentrations of dissolved uranium from the KURT granite samples with increasing contact time (ug/L)

U	UD-BI	UD-Bg	UD-CO ₃	UD-Na	UD-Ca	UD-CaSi	UD-Si
0 D	20.53	0.78	0.68	0.35	0.09	0.44	0.73
1 D	188.46	2.80	2.68	0.82	0.38	0.71	0.81
2 D	251.73	3.73	4.20	1.03	0.50	0.85	0.72
5 D	298.11	4.30	4.60	1.37	0.81	0.81	0.60
1 W	298.71	4.58	5.13	1.84	1.03	0.86	0.54
2 W	287.79	4.52	5.01	2.42	1.52	0.99	0.50
1 M	277.66	5.31	5.96	3.63	2.36	1.66	0.57
2 M	262.84	5.74	6.29	5.26	3.62	3.11	0.75
3 M	247.29	-	-	-	-	-	-
5 M	247.95	9.25	10.06	10.15	9.95	7.60	5.25

4. Conclusion

Our result shows that dissolution of uranium from the KURT granite is estimated to be affected by at the presence of CO₃²⁻. According to collected KURT groundwater properties and composition at DB-1 core of KURT (91.7 ~ 199.7 m), pH and HCO₃⁻ of groundwater were identified as 8.3 ~ 8.5 and 64.1 ~ 85.4 mg/L, respectively, and the main chemical species of uranium was a calcium-uranyl-carbonate complex, Ca₂UO₂(CO₃)₃(aq) [2]. Since CaCO₃ is also present at KURT granite, KURT groundwater is estimated to have a sufficient condition for uranium to be dissolved by CO₃²⁻. Therefore, under the geochemical conditions of KURT, uranium can be dissolved from the KURT granite by interactions with groundwater. Furthermore, the dissolved uranium by interaction with groundwater is able to migrate through fractures in the KURT granite. Besides, our result can explain why the concentration of uranium in groundwater shows an anomalously higher value in a certain part of DB-1 borehole.

REFERENCES

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- [2] M. H. Baik et al., "Determination of uranium concentration and speciation in natural granitic groundwater using TRLFS", J. Radioanal. Nucl. Chem., Vol.305, p.589 (2015).