Evidence From Fracture Filling Minerals for the Interpretation of Uranium Isotopes in Investigating Ancient Redox in Granitic Rock in KURT Site

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1. Introduction

The study of past groundwater condition is of particular interest in the context of potential waste repository site characterization. It is because of the need to predict the future behavior of the groundwater system over timescales of relevance to safety assessment. It is necessary to develop an understanding of how the groundwater system has changed over time in response to changing driving forces for groundwater flow and hydrogeochemical evolution. The generation of fracture filling minerals contains geochemical record of the compositions of the groundwaters from which they precipitated. The combination of isotopic characteristics of fracture filling minerals provides very important information of paleo-hydrology of a geological site. In this study, to advance the understanding of uranium isotopes in fracture filling minerals and its response to groundwater flow and chemical evolution, we measured U isotopes such as ^{238}U, ^{235}U, ^{234}U in fracture filling minerals in KURT site.

2. Methods

The study site is located at KURT (KAERI underground Research Tunnel) site in Korea Atomic Energy Research Institute, Yuseong area, the northern part of the Daejeon city, Korea. Boreholes MB1, KP3 and KP4 were drilled inside KURT, MB2 and DB2 outside KURT, respectively.

Fracture filling minerals were obtained from the drill cores from MB1, MB2, KP3, KP4 and DB2 boreholes that they were drilled into the highly fractured domain. Fracture coatings from drill cores were collected from drilled cores at various depths up to ~1000 m. Samples from the surface of fractures were crushed into powder using a pestle and mortar. Samples were prepared by MW-assisted digestion in HNO_3 + HCl + HF mixture. $^{238}U/^{235}U$ isotope ratio measurements were carried out by MC-ICP-MS (NEPTUNE, ThermoScientific). $^{235}U/^{234}U$ isotope ratio measurements were carried out by ICP-SFMS (ELEMENT XR, ThermoScientific) at ALS Scandinavia AB, Sweden. The determination of U, Fe, Mn concentrations was performed at the Korea Basic Science Institute using an ICP-AES (Perkin Elmer) and an ICP-MS(VG, PQ3).

3. Results

3.1 Redox front

The concentrations of Mn and Fe in fracture filling minerals indicated they were enriched at similar depths. Mn and Fe are believed to have formed in oxidized forms. The presence of significant concentrations of U suggests that the groundwater from which it precipitated was reducing. The enrichment patters of Mn, Fe and U indicated the location of redox front in KURT site, which was the range of 100 – 200 m depths (Fig. 1).

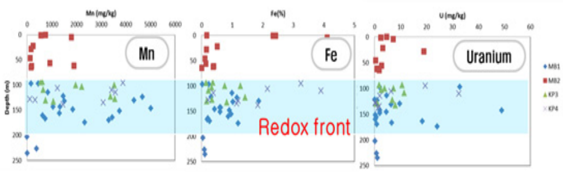

Fig. 1. Enrichment patterns of Mn, Fe and U in the KURT site indicated the presence of redox front.

3.2 $^{234}U/^{238}U$

At the shallow depths in the site, $^{234}U/^{238}U$ AR showed various values as shown in Fig. 2. At the deeper depths (below ~ 600 m depth), the values of $^{234}U/^{238}U$ AR became constant, which might indicated the secular equilibrium of U bearing minerals in fracture filling minerals. Thus, it indicated that the mobility of U was very slow to reach the secular equilibrium in the groundwater system, while the scatter patterns of $^{234}U/^{238}U$ AR resulted from weathering and precipitation. During weathering process, ^{234}U is preferred to be removed from mineral matrix and it is enriched in groundwater interacting with the solid phase [1]. The U bearing minerals was formed from groundwater that was enriched in ^{234}U resulted in higher values of $^{234}U/^{238}U$ AR, which is believed to happen continuously [1].

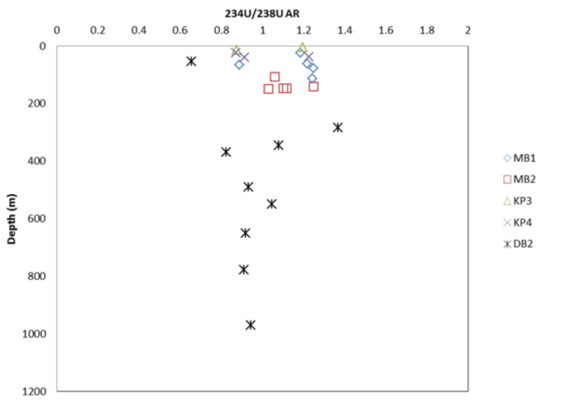

Fig. 2. Patterns of $^{234}U/^{238}U$ AR with depths in KURT site.

3.3 *$^{238}U/^{235}U$*

At the shallow depths uranium reduction process prefer ^{235}U over ^{238}U. Thus ^{238}U was enriched in U bearing fracture filling minerals by uranium reduction [1]. But the enrichment of ^{238}U is limited by microbiological uranium reduction. Thus, the large number of $^{238}U/^{235}U$ ratio in U bearing minerals indicated that U reduction was not the only process to occur but more processes involved in the formation of U bearing minerals. At shallower depths in KURT site, the values of $^{238}U/^{235}U$ ratio were scattered (Fig. 3), which shows the different sources of uranium. Thus it indicates groundwater was mixed at shallower depths. At deeper depths, the values of $^{238}U/^{235}U$ ratio became constant. It indicated that the groundwater system at the depths was not mixed and U reduction was the major process influenced $^{238}U/^{235}U$ of U bearing minerals.

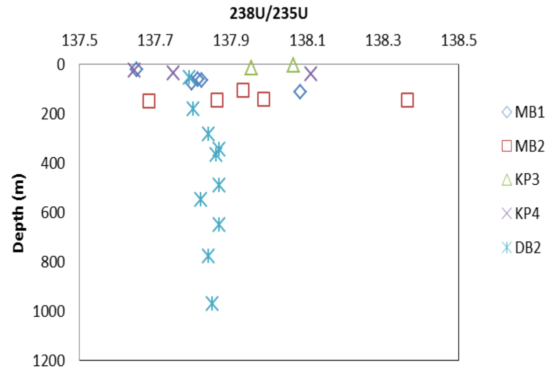

Fig. 3. Patterns of $^{238}U/^{235}U$ with depths in KURT site.

This study evaluated the location of redox front based on the patterns of concentrations of Mn, Fe and U in fracture filling minerals. Also, we evaluated paleo-hydrology condition which showed paleo-groundwater mixing, weathering and U precipitation at shower depths while very slow U mobility enough to reach the secular equilibrium at deeper depths (below ~ 600 m depths) in KURT site (Fig. 4).

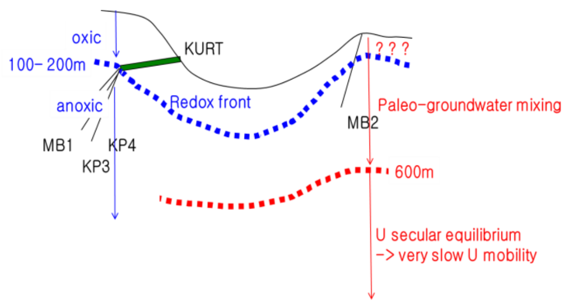

Fig. 4. A conceptual model for redox front and secular equilibrium in U, which indicated very slow U mobility to reach the secular equilibrium in KURT site.

REFERENCES

[1] D.M. Bonotto, "A comparative study of aquifer systems occurring at the Parana sedimentary basin, Brazil: U-isotopes contribution", Environ. Earth Sci., 68, 1405-1418 (2013).

[2] M. J. Murphy, C. H. Stirling, A. Kaltenbach, S.P. Turner, and B.F. Schaefer, "Fractionation of $^{238}U/^{235}U$ by reduction during low temperature uranium mineralization processes", Earth and Planetary Science Letters, 388, 306-317 (2014).