Chemical analysis of REE in calcite and retardation effect of calcite in granite around Tono uranium ore deposit

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[Introduction]

The geological disposal in the multibarrier system that consists of the artificial barrier (vitrified waste, overpacking, and buffer material) and the natural barriers (rock) is thought to be the best method for the high-level nuclear waste disposal to the geological formation in more than 300m depth in our country . It is generally said that it will take longer than several 105 years to 109 years by the time that the radionuclides become to have the doses of radioactivity level of nature (uranium ore deposit). Natural analogue studies are methods to evaluate the long-term change of natural barriers. One of the natural analogue studies is the analysis of the behavior of the REE (lanthanide) whose chemical features are similar to radionuclides (Am, Cm) that do not exist in natural environment. As a result, evaluation of long-term safety of the natural barrier becomes possible. In this research, REE and of uranium in the carbonate mineral in granite in Tono area, Japan analyzed and the retardation effect were of carbonate for radionuclides were investigated.

[Samples and Analytical Method]

The 50 granite samples containing one fresh granite sample were collected from drilled core (06MI03 hole) from Gifu Prefecture, Tono uranium mining area. These samples were analyzed for identification of minerals by XRD, consist of major elements by XRF, and trace elements by ICP-MS. Moreover, the major elements and trace elements including REE of calcite extracted by 1M acetic acid resolution method (Okai, 1998) from these samples by AAS and ICP-AES, by ICP-MS, respectively. And 10 samples with a different content of the calcite were chosen for the extraction of REE by the pH5 acetic acid buffer solution method (Okai, 1998) whose dissolution intensity is smaller than 1M acetic acid method. These samples were analyzed in a same method.

[Conclusion and Discussion]

The average concentrations of REE and uranium in bulk samples which do not contain are lower than those of calcite. And the REE pattern normalized by fresh granite exhibits light REE enrichment compared with middle and heavy REE. That means calcite intends to take up more light REE than middle and heavy REE. This tendency thought to be due to ionic radius. Ionic radius of light REE is near about 100pm which is similar to that of Ca^{2+}. Because radionuclides (Am, Cm) have chemical similarity with light REE in terms of charge and ionic radius, the retardation effect of calcite on Am and Cm migration can be expected.

Next, REE and uranium concentrations obtained by the two extraction methods show that 1M acetic acid method was higher in abundance of calcite samples, and reverse in the middle of the samples. Moreover, the pattern shows that light REE enrichment and, the middle and heavy REE depletion, for the samples containing high amount of calcite. This indicates acetic acid complexes are stable because of a lower pH in 1M acetic acid than pH5 acetic acid buffer solution. And the reason why the pattern shows light REE enrichment reflected by the REE pattern of the calcite. In the latter case, because there are few calcites and a large amount of feldspar was dissolved, and the pH increased. Thus, a steady acetic acid buffer solution in pH5 extracted more feldspars than 1M acetic acid method. Moreover, because the stability of acetic acid complex with REE is increasing from La toward Lu, the pattern became middle and heavy REE enrichment.