

Geochemistry of carbonate minerals in sedimentary rocks around Tono uranium deposit, central Japan

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A retardation effect (an effect to retard migration of a radioactive element in the underground) by mineral adsorption has been studied a lot by a conventional natural analogue study, but a few studies of retardation effect by mineral incorporation (formation of solid solution) has been studied. It is expected that mineral incorporation could be a long-term retardation effect. Therefore, in this study, carbonate mineral incorporation is considered, because carbonate mineral is one of the most ubiquitous authigenic mineral in the sedimentary rocks. In addition, the rare-earth elements are used as chemically analogue elements to radioactive elements such as Am and Cm don't occur in natural environment.

C1 chondrite-normalized REE pattern of the carbonate shows the W-type effects. M-type tetrad effects can occur in the REE pattern of weathered granitic rocks, while the W-type tetrad effects, the counterpart of fraction occur in the groundwater by water-rock interactions in the Tono area (Takahashi et al., 2002). The REE pattern of sedimentary rocks shows W-type tetrad effect by REE provided from groundwater (Takahashi et al. 2002). Therefore, it is thought that the rare-earth element is incorporated into carbonate from the groundwater, resulting to W-type tetrad effect of carbonate as well as the groundwater.

In particular the light rare-earth element concentration in carbonate is high, compared to that of bulk rock. Because light rare-earth element is chemically analogue to radioactive element such as Am and Cm, it is likely that a carbonate mineral is easily to incorporate these especially. Therefore, it is expected that a carbonate mineral incorporation can retard long-term migration of radioactive elements such as Am and Cm.

Groundwater-normalized partition coefficient was calculated to discuss REE incorporation into carbonate. From the relationship between the ionic radius and the partition coefficient of elements in the carbonate minerals in Tono sedimentary rocks, it is understood that the partition coefficients of free cations whose ionic radius are closer to that of Ca^{2+} and charges are higher are higher. For example, partition coefficient of trivalent REEs is higher than that of monovalent alkali elements or divalent alkaline earth elements.

Moreover because it is thought that chemical species of the REE in the solution influences partitioning of elements into the carbonate minerals, REE speciation was calculated assuming that all REEs in the solution are carbonate complexes. As a result, it is found that $\text{REE}(\text{CO}_3)_2^-$ is dominant among REE species in the groundwater.

It is thought that the ionic radius and the charge influence significantly incorporation of radioactive elements (Am, Cm) into carbonate minerals under actual underground geological environment.