

準安定なカルシウム炭酸塩鉱物への陰イオン吸着とその後の挙動 Anion adsorption and post-adsorption behavior of metastable calcium carbonate polymorph

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In Japanese transuranic (TRU) waste disposal facilities, I-129 is the most important radionuclide that must be considered in long-term safety assessments of the repository. However, the degradation of cement materials used in the repositories can produce high pH pore fluids that can affect to anion transport behavior. Therefore, it is necessary to understand the behavior of anions such as I⁻ in hyperalkaline conditions. Examples of I⁻ behavior in natural hyperalkaline environments, such as in Oman, show that I⁻ is taken up by aragonite, opening up the possibility of calcium carbonates as inhibitors of I⁻ migration. This concept is currently being applied in the development of the Advanced Liquid Processing System (ALPS), which employs carbonate coprecipitation to treat contaminated waters resulting from the Fukushima Daiichi nuclear power plant accident. However, the stability of the carbonate phases precipitated in this system as well as the anion uptake capacities of these phases are poorly understood. In a previous study, (Kasahara, 2012), it was found that monohydrocalcite (MHC), a precursor of aragonite, affects the iodine capacity of aragonite, making it a possibly important material that can control the behavior of anions. The objective of this study therefore, is to investigate the sorption capacity of MHC for anions and its stability. MHC ($Mg^{2+}/Ca^{2+}=6$; $Ca^{2+}/CO_3^{2-}=1$) was synthesized and used for sorption experiments involving F⁻, Br⁻, I⁻, IO₃⁻, SO₄²⁻, CrO₄²⁻, HAsO₄²⁻, and phase transformation experiments. Results show that K_d values of HAsO₄²⁻ and F⁻ on MHC are high, while IO₃⁻, SO₄²⁻ are relatively low. On the other hand, Br⁻, I⁻, NO₃⁻, CrO₄²⁻ were not taken up. It is because MHC has high chemical reactivity and high specific surface (4 times large of aragonite, 15 times large of calcite), in addition MHC is most low density of calcium carbonate, so MHC can take up relatively large amount of anions than other calcium carbonate. And other thing, MHC involves Mg²⁺ abundantly. This study indicates that Mg²⁺ form fluoride adsorption site. Results of the transformation experiments show that MHC with no adsorbed anions easily transforms into a stable phase, whereas MHC loaded with increasing amounts of anions transform after longer durations. It is because the driving force for the transformation decreases with the anions content in the solution. In conclusion, MHC can take up fluoride and oxyanions that ionic radii is similar to carbonate but larger than that. In addition, MHC is stabilized as a function of uptake amount of anions.