Smectitic Alterations in Drill Cores obtained from the campus of Tokai University, Hiratsuka, Kanagawa Prefecture

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In the geological disposal of radioactive waste, cementitious materials and bentonite, which contains smectite as a major mineral component, are to be used as part of the engineered barrier system. Natural analogues of the reactions between cementitious materials and bentonite have been studied to understand reactions on the timescale of tens of thousands of years and longer. Drill cores were obtained from the campus of Tokai University in Hiratsuka, Kanagawa Prefecture, for a natural analogue study of smectitic alterations in the calcium-rich water released by cementitious materials.

A sandy tuff with smectite was observed in drill cores from depths of 239.7 to 250 m, the deepest of the drill cores. Foraminifera microfossils were found in the sandy tuff, suggesting that the tuff was deposited under the sea. Fission-track and uranium-lead dating of the zircon in the sandy tuff were performed to obtain ages of ca. 10 Ma, and thus, it was estimated that the sandy tuff was deposited at or later than ca. 10 Ma. The altitude of the drilling location was assumed to have come above sea level at ca. 0.13 Ma (Oka, 1980), and so the sandy tuff was thought to have been under the sea for a maximum period of ca. tens of millions of years.

Two types of smectite were observed coexisting with zeolite in the sandy tuff, suggesting that two different types of smectite were formed in at least two different periods.

a) Smectite formed as a result of zeolite alteration: Electron probe micro-analysis (EPMA) indicated that the smectite belonged to dioctahedral montmorillonite. The oxygen-isotope geothermometer, obtained from the isotopic equilibrium of stable oxygen isotopes in the smectite and the coexisting zeolite, indicated temperatures ca. 40 degrees C.

b) Smectite with calcite: Both minerals are included in the zeolite crystal, and EPMA indicated that the smectite was trioctahedral saponite. The oxygen-isotope geothermometer indicated temperatures ca. 165 degrees C.

The chemical compositions of smectite and plagioclase in the sandy tuff were analyzed using EPMA. The results show that variations of Ca/(Ca+Na) at different depths were similar in the smectite and the plagioclase, and suggested that a possible source of the calcium in the smectite was from the dissolution of the plagioclase.

A numerical analysis of long-term alteration was performed to examine whether the mineral composition of the sandy tuff could be explained by calculations based on thermodynamic data. In this analysis, a hypothetical mineral composition calculated based on the chemical composition of non-altered rock near the drilling location was used as the starting material, with seawater as the reacting fluid. The deposit of only Na-type montmorillonite was permitted, and the number of sites for ion exchange was decided by the amount of Na-type montmorillonite. Other types of montmorillonite including Ca, K and Mg, were formed in the exchange of ions between the reacting fluid and each type, based on the selectivity coefficient for ion-exchange reactions.

From the numerical analysis, it was found that the mineral composition became stable after ca. 10 000 years. The mineral composition resulting from the analysis included Ca-type montmorillonite and saponite as the dominant smectites, followed by Na-type montmorillonite and saponite. The above sequence of smectites was coincident with the sequence observed in the sandy tuff. The concentrations of each element in the reaction fluid in these results were coincident with the measured concentrations of the water in a hot spring near the drilling location, showing differences within two orders of magnitude.

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Reference


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