Dissolution rate of chalcedony in alkaline condition

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The dissolution of minerals composed in the bentonite is important to evaluate a long-term behavior of the engineered barrier in a radioactive waste disposal. Specifically, the dissolution of montmorillonite is a key issue because it contributes to the low-permeability of the bentonite. Also, dissolution of SiO_2 minerals are noted as a resource of Si ion in terms of the saturation of montmorillonite. While the dissolurion kinetics of quartz and amorphous silica was well-known, it is difficult to understand that of chalcedony due to microcrystalline form of SiO_2. Therefore, this study investigated the dissolution rate of the chalcedony in the alkaline condition.

The flow-through experiment was carried out using the chalcedony and quartz. Initial pH of NaOH-NaCl solution was from 8.9 to 13.5. The ionic strength of the solution was adjusted to 0.3 mol/L. The temperature was 15, 25 and 50 degree C. The dissolution rate was calculated using the Si concentration in steady state. After the experiments, the sample was observed by SEM.

The relation between the dissolution rate, *Rate*, and the hydrogen ion activity, a_{H+}, was obtained as a following function; $Rate=k\times a_{H+}^{-n}$. The reaction constant, k, and the reaction order, n, are listed below. The higher temperature represented larger dissolution rate of the chalcedony. The dissolution rate of the chalcedony was almost same as that of quartz at same temperature. After the experiments, SEM observation showed that the rough surface of primary chalcedony partly changed to smooth surface like as quartz. It is supposed that the rough surface of primary chalcedony was rapidly dissolved because of a low crystallinity. The dissolution rate obtained is supposedly applicable to the highly crystalline SiO_2 of chalcedony.

Table 1 Reaction constant and reaction order

Sample	Solution	Temp (℃)	k	n
Chalcedony	NaOH-NaCl	15	$10^{-13.72}$	-0.30
	NaOH-NaCl	25	$10^{-13.02}$	-0.30
	NaOH-NaCl	50	$10^{-12.68}$	-0.34
Quartz	NaOH-NaCl	50	$10^{-11.67}$	-0.23