

## Dissolution processes and mechanisms of chlorite inferred from FIB-TEM analyses

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Dissolution processes and mechanisms of chlorite (clinochlore) were examined by TEM, focusing especially on the structural modification of crystal edges in experimentally dissolved samples. Focused-ion beam (FIB) TEM sample preparation was applied to crystals dissolved in a flow-through reaction system at pH 3.0 and 25 degrees for 31 days.

Solution analyses showed that release rates of cations reached constant after 20 days. Silicon dissolution was at a rate of  $-11.5$  ( $\log R$ ,  $\text{mol}/\text{m}^2\text{s}$ ) in almost agreement with previous experimental studies, but accompanied by a higher magnesium rate ( $-11.1$   $\text{mol}/\text{m}^2\text{s}$ ), implying that the dissolution was incongruent.

TEM observation of dissolved samples revealed a development of amorphous zone of 20 - 30 nm in thickness at an outermost rim of the crystal. Comparison between dissolved and undissolved specimen evidently demonstrated that this is not an artifact but a product during the dissolution processes. EDX analyses indicated depletions of Mg and Al relative to Si in this amorphous zone. Morphological characteristics indicate that the Si-rich amorphous zone was produced by selective reaching of metal ions except for Si, rather than the re-precipitation of Si gel from the experimental solution. The microtexture and chemistry have indicated the amorphous zone is a surface leached layer. At the boundary between the leached layer and intact chlorite, several alteration features indicating preferential destruction of hydroxyl layers in chlorite were observed by high-resolution TEM: ex-foliation of TOT layers, and amorphization or collapse of hydroxyl layers. Chlorite dissolution may proceed via the retreat of the leached layer through which Mg and Al are successively released to the solution.