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## Nano-scale metrology for extra long-term prediction of global environmental change

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Global environmental change and energy problems are unavoidable issue for every human being. Although the utilization of nuclear power and fossil fuel are standing oppositely for emission of green-house gas, they are sharing common problem, wastes. Underground as the final destination of the wastes causes dissolution and precipitation of minerals toward the equilibrium. In the engineering fields for radioactive waste disposal and CCS (carbon capture and storage), long-term safety is predicted by computation based on limited kinetic data of interaction among groundwater, country-rocks, cement-buildings, and barrier-clay and on hydraulic behaviors.

In order to predict the underground condition for extra long-term period (~100 kyr), we deed to obtain precise kinetic data in the order of 1E-5 nm/s. During this period, rock and minerals should cause a few cm displacements, so that the groundwater flow can be changed. Latest metrological instruments (AFM and interferometer) are able to detect such an ultra-slow phenomenon. Interferometer is sometimes useful to reproduce the realistic reaction with in-situ measurement. I will here report the latest examples of (1) mineral dissolution measurements, (2) erosional dispersion of molecular layer by flow and (3) growth measurement of calcite, by using micro-interferometer with phase-shit optics.

Plagioclase, one of the most common rock-forming minerals, is poorly understood in its fundamental mechanisms of dissolution. In the most groundwater system, slightly alkaline pH promotes the dissolution. Dissolution process of Ca-bearing phases is important, because calcite and Ca-zeolite can be precipitated as the secondary phase. Dissolution experiments of anorthite (Satoh et al., 2007) and hornblende (Sato et al., in prep.) at alkaline conditions provided information about interface process accompanying with step retreat velocity, step density, and pH. Another experiment on dissolution measurement on zoned plagioclase suggested that solid-solution is controlling dissolution rates. Secondary phase precipitation was recognized in the process of high-pH dissolution of hornblende. Aggregates of colloidal precipitates deposited near etch pit and inhibited subsequent dissolution. Similar observation was made on olivine surface immersed in acidic solution. Amorphous Si-rich phase was recognized as secondary phase, which affects the dissolution process.

In natural system, clay can be eroded and dispersed into groundwater. Molecular layer can be lifted-up by swelling and DLVO repulsive force, and colloidized by flow. Interferometric measurement estimated possible colloid concentration ( $mol/m^3$ ) of smectite, based on the observed colloidization rate ( $mol/m^2/s$ ) and flow (m/s). Under a few hundred um/s of groundwater flow, ~10 ppb of colloid can be expected.

Growth rate of calcite is the most important information for CCS engineering. In the georeactor process by  $CO_2$ -injection into hot-dry rock, carbon is fixed as calcite (Ueda et al., 2009). The growth rate of calcite is key information to evaluate the life-time of storage and the capacity. Laboratory in-situ measurement of calcite growth using high-T and high-P cell, and on-site experiment using Crystal Growth Sonde (Satoh, 2011) which enables deep-diving and in-situ growth were conducted to examine the system. The results showed that calcite growth is inhibited in the actual georeactor system in spite of sufficient supersaturation.

Thus, the mineral dissolution and growth in natural system tend to show slower rates. As a possible reason, widely existing and important, but forgotten impurity, colloid needs to be considered.

References

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