

## Crystal growth in clay solidification: zeolitization of smectite

\*Hisao Satoh<sup>1</sup>

1.Naka Energy Research Laboratory, Mitsubishi Materials Corporation

At the Earth's surface, rock-forming minerals are weathered and altered to be fine particles of clay. Though the clay particles stably exist in the burial condition, unsolidified mud composed of clays is re-equilibrated with groundwater and diagenetically stabilized as solidified sedimentary rock. One of the most friendly clay material in our life is smectite. Since smectite under subsurface condition suffers from alkaline groundwater and can be altered into zeolite, the natural bentonite rocks composed of smectite always contain zeolite.

Investigating growth kinetics of zeolite at various solution, temperature and pressure conditions is important to estimate the time-scale of stabilization of clay materials. We conducted alteration experiment of Na-montmorillonite in 1.0 M NaOH at 120 °C and measured dissolution and growth rates by in-situ phase-shift interferometry [1]. The simplified reaction can be scripted as:  $\text{Al}_2\text{Si}_4\text{O}_{10}(\text{OH})_2 + 2\text{NaOH} = 2\text{NaAlSi}_2\text{O}_6(\text{H}_2\text{O})$ . At this condition, dissolution of montmorillonite proceeds at  $6.9\text{E-}6$  m/s calculated from rate equation [2] and molar volume. Growth rate of analcime was measured to be  $1.1\text{E-}10$  m/s. Since the reactive surface area of montmorillonite is reducible from 1/10 to 1/1000 by layer-stacking, this observed growth rate of analcime is mostly consistent. However, total volume change of before and after reaction gives  $193.96 - 173.91 = +20.05 \text{ cm}^3/\text{mont mol}$ , which suggests that this reaction is suppressed in the limited space. Further, reaction system in the presence of large excess NaOH with a syringe compaction cell produced zeolite as hydro-sodalite scripted as:  $3\text{Al}_2\text{Si}_4\text{O}_{10}(\text{OH})_2 + 20\text{NaOH} = \text{Na}_8\text{Al}_6\text{Si}_6\text{O}_{24}(\text{OH})_2 + 6\text{Na}_2\text{SiO}_3 + 12\text{H}_2\text{O}$ . Its volume change is calculated to be  $+54.67 \text{ cm}^3/\text{mont mol}$ , which suggests that this reaction can be more suppressed than low-alkaline system. Thus, alkali plays important role for stabilization by zeolitization but this alteration cannot be achieved entirely except at pores.

[1] Satoh et al. (2007) American Mineralogist, 92, 503-509.

[2] Sato et al. (2005) Proc. of the Int. Workshop on Bentonite-Cement Interaction in Repository Environments, A3-38-41.

Keywords: smectite, zeolitization, growth rate, volume change