Alteration of bentonite by hyperalkaline fluids

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Introduction

Cementitious materials and bentonite will be used as backfill and buffer materials respectively for a geological disposal of radioactive wastes. Cement hydrates will gradually dissolve and leach out from cementitious materials over long-term period on contact with the groundwater. Bentonite may alter and its nuclide transport properties, e.g. permeability etc.. may also change by alkaline fluids. This study aims at qualitatively investigating the alteration by batch experiment with hyperalkaline solutions.

Experimental

$Ca(OH)_2$ solution, mixed solution of $Ca(OH)_2$, NaOH, KOH and low-alkaline cement leachate with pH in the range 10.5 to 13 were provided as hyperalkaline fluids. 1g of bentonite sample was soaked in 1L of the solution in resinous container for each case. The containers were kept under the different temperature conditions (50, 80, 100 °C). During batch experiment, pH control had been done to keep it nearly the expected pH by adding alkaline solution with a few exceptions. Solid-liquid separation was carried out at the planned periods (30 to 720days). The mineral composition of solid phase was analyzed by XRD, TG-DTA and surface morphology was examined by SEM. Methylene blue adsorption was also measured to estimate the quantity of montmorillonite. The element concentrations of the liquid phase were measured by atomic absorption spectrometry.

Results

(1) $Ca(OH)_2$ solution [pH12.5, 11.5, 10.5]

In case of pH12.5 and 100 °C, montmorillonite dissolved in 14 days and calcium silicate minerals such as CSH and CASH formed in 7days. In case of pH12.5 and 80 °C, dissolution of montmorillonite and formation of secondary minerals were also observed but it took more time to show these changes. However, the formation of secondary minerals were not observed under lower pH (<10.5) conditions.

(2) Mixed solution of $Ca(OH)_2$, NaOH and KOH [pH13]

Dissolution of montmorillonite and formation of CSH, CASH were observed under any temperature condition. The higher temperature became, the earlier these phenomena occurred.

(3) Low-alkaline cement leachate [pH11]

Although the exchangeable cation of montmorillonite transformed into Ca dominant type from Na dominant type under any temperature condition, dissolution of montmorillonite and formation of calcium silicate mineral were not observed.

Discussion

The batch experiment for 720days showed that the higher pH(>11.5) and temperature became, the more dissolution of montmorillonite and formation of secondary minerals were accelerated. It is likely that low-alkaline cement is favorable material for the stability of repository environment.

Percolative segregation of iron melts during core formation

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Introduction

Physical processes on formation of metallic cores in the Earth have controlled the chemical composition of the core and mantle. Previous high-pressure experimental studies up to 25 GPa have indicated that metal segregation by permeable flow is prevented due to high dihedral angle between liquid iron and silicate minerals. Here we report the connectivity of liquid iron in $(Mg,Fe)SiO_3$-perovskite aggregates at higher pressures corresponding to the lower mantle depths.

Experimental methods

High pressure and temperature experiments were performed using both multi-anvil apparatus and laser-heated diamond-anvil cell (LHDAC). A mixture of iron (99.9 % purity and less than 2µm in size) and gel powder, with a composition of $Mg_{0.9}Fe_{0.1}SiO_3$, was used as starting materials (8:92 in weight). Chemical analysis and texture observations of recovered samples were performed using analytical transmission electron microscopy (ATEM).

Results and Discussions

The experiments demonstrate that the measured dihedral angles of liquid iron are reduced to 56° with increasing pressure to 47 GPa. This value is smaller than the critical angle of 60°, allowing the formation of interconnected iron melt networks within the perovskite-bearing rock, even at small melt fractions. This change in wetting behavior could be due to the reduction of perovskite-iron interfacial energies by increasing temperature of iron melt and by dissolving oxygen and silicon in molten iron from silicate perovskite as pressure increases. The separation of molten iron during core formation may have proceeded by percolation in a deep perovskite-bearing rocks. Oxygen and silicon incorporated during the core formation is an important to understand the light elements in the Earth’s core. And this core separation mechanism can shed light on the explanation for the observed “Si depletion” of the upper mantle.