Principal Research Results


Background

Research and development on specifications of disposal concepts and a reliability of safety assessments of TRU waste disposal are addressed by the Federation of Electric Power Companies and Japan Nuclear Cycle Development Institute, as given in their responsibility for management of TRU waste. Cementitious material is a potential waste packaging, backfilling and constructing material for the disposal of TRU waste (Fig.1) and is expected to provide both chemical and physical containment. For the long term performance assessment of the containment properties of cementitious materials, it is necessary to develop a series of predictive calculation models of the long-term evolution of the cementitious repository system. In past performance assessment studies in Japan, the evolution of cementitious materials was discussed only using the homogeneous porosity transport model, however, an evaluation method to discuss the alteration of cementitious materials in a fractured repository should be also developed from a realistic viewpoint.

Objectives

To develop a coupling transport and chemical equilibrium calculation code to predict the chemical alteration behaviour of the fractured cement material in the TRU waste repository environment.

Principal Results

1. Development of calculation code to predict the alteration behaviour of fractured cementitious materials

Coupled Chemical equilibria-mass transport code for Fractured-media (CCT-F), was developed to predict the alteration behaviour of fractured cementitious materials in a groundwater flow condition. In CCT-F, the thermodynamic incongruent dissolution/precipitation model of calcium-silicate hydrate (C-S-H) developed by us is coupled with a quasi-two-dimensional transport model in which the transport in the cracks is described by only advection and dispersion and transport within the cement solid matrix is constrained to be diffusive and perpendicular to the direction of the crack (Fig.2).

2. A flow-through leaching experiment using an artificial fractured cement hydrate and verification of the CCT-F code

A flow-through leaching experiment using an artificial fractured ordinary Portland cement (OPC) column sample and distilled water was carried out and the composition of the discharged solution and the distribution of components in the solid matrix were analysed. It showed that calcium leached from the surface due to dissolution of Ca(OH)₂ and C-S-H and the Ca/Si ratio of the solid decreased in the surface region (Fig.2). As the amount of discharged water increased, calcium concentration decreased and silica concentration increased with time due to the incongruent dissolution of C-S-H, with decreasing Ca/Si ratio (Fig.3). The alteration behaviour of the artificial fractured OPC was reasonably well predicted with the proposed model, therefore, we can conclude that CCT-F code is applicable to the discussion on possible alteration of cementitious materials along a crack.

Future Developments

Sophisticated model and code considering a network of plural fractures will be developed based on the above model, and will be applied to predict the chemical alteration behaviour of the fractured cementitious facility in the TRU waste repository. Long term performance of the containment properties of the facility will be also evaluated to take radionuclide retardation into account.

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References:
**Fig.1** Concept of the TRU waste disposal facility

**Fig.2** Schematic representation of transport model in fractured cement materials and experimental design of flow-through leaching experiments with calcium concentration profile at cross section of leached OPC sample

A prototype coupling transport and chemical equilibrium calculation code was developed by describing the transport in fractured cement solid as a quasi-two-dimensional transport in which the transport in the cracks is described by only advection and dispersion and the transport within the cement solid matrix is constrained to be diffusive and perpendicular to the direction of the crack.

In the flow-through experiment, leaching of calcium from the surface and the distribution of Ca/Si ratio in the solid matrix was observed.

**Fig.3** Measured and predicted aqueous compositions of discharged water

Measured Ca and Si concentrations in the flow-through leaching experiment using an artificial fractured OPC hydrate were reasonably well predicted by CCT-F.