Restraint of Radinuclides Migration by Cementitious Materials containing Fly Ash

Background

In the concept of sub-surface repository system for the low-level radioactive waste disposal in Japan, cementitious materials are considered for use as an engineered barrier to restrain the migration of radionuclides (Fig. 1). Especially, organic carbon-14 and iodine-129 have potentially large contributions to the dose evaluation, but they are not sorbed so strongly on cementitious materials. Low-heat Portland cement containing 30 wt% fly ash (FAC) is a candidate cement material for the construction of sub-surface repository because of its high density structure. Therefore, the diffusion of radionuclides in FAC materials is a very important parameter for the performance assessment when considering the release of those radionuclides from wastes and its migration in a cementitious repository environment.

Objectives

The purpose of this study is to estimate effective diffusion coefficients for organic carbon and iodine by diffusion experiment, and clarify the barrier effects of FAC materials.

Principal Results

1. Change of diffusion rates in FAC

The diffusion of acetate, which is one of typical chemical form of organic carbon, and iodide in FAC hardened cement paste was examined by the through-diffusion experiment (Fig. 2). As a comparison, ordinary Portland cement (OPC) was also used for the diffusion experiments. The water-to-cement weight ratio was 0.35. For OPC hardened cement paste, diffusion rates of trace ions were constant (Fig. 3(a)). On the other hand, diffusion rates for FAC gradually became smaller (Fig. 3(b)). This is probably due to the addition of pozzolanic materials such as fly ash, and it is possible that the structure of FAC gradually becomes more complicated due to the pozzolanic reaction.

2. Effective diffusion coefficients

Effective diffusion coefficients, D_e , of trace ions for FAC were estimated to be 10^{-13} m² s⁻¹ order at the beginning of diffusion experiments. Then, the diffusion of trace ions slowed down over the experimental period of 4-18 months, and D_e were estimated to be 10^{-14} m² s⁻¹ order. On the other hand, the D_e for OPC were estimated to be 10^{-12} m² s⁻¹ order.

These suggest that FAC can have further effective barrier performance for the diffusion of trace ions as time passes.

Future Developments

As future works, it is necessary to clarify the mechanism of the restraint of radinuclides migration and consider the relation between the long-term pozzolanic reaction and the diffusion behavior of trace ions in FAC materials.

Main Researcher: Taiji Chida, Ph. D.,

Senior Research Scientist, Radiation Safety Research Center, Nuclear Technology Research Laboratory

Reference

T. Chida and D. Sugiyama, 2008, "Diffusion of Organic Carbon and Iodine in Low-heat Portland Cement containing Fly Ash", CRIEPI Report L07018 (in Japanese)

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Fig.1 Concept of sub-surface disposal.







Fig.3 The diffusion of iodide through the hardened cement pastes.