1–1 Evaluation of Uranium Leaching from Fuel Debris in Water

—Leaching Rate Evaluation Considering Effects of Concrete Components, and Comparison with Spent Fuel—

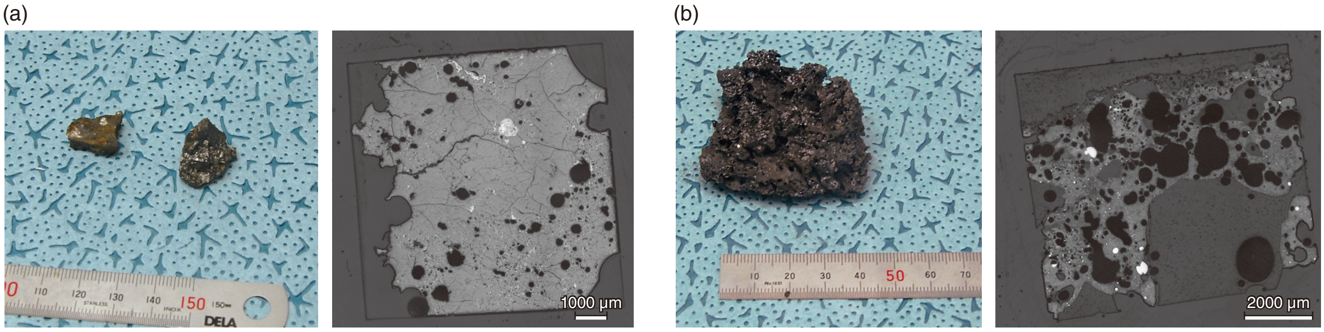

Fig.1-2 Observed appearance (left) and cross section (right) of simulated fuel debris samples from the leaching test
Two types of simulated fuel debris were used. The resulting leaching rates are 2.53 mg/m^2/d and 2.86 mg/m^2/d with/without concrete components. This is comparable to rates of spent fuels.
(a) Without concrete: A sample containing U, Zr, Fe, rare earths, etc. was heated at 2760 °C for approximately 45 minutes in reducing atmosphere (Ar–H_2). (b) With concrete: A sample containing U, Zr, Fe, rare earth, etc. and additive Si, Ca, Mg as concrete components was heated at 2000 °C for approximately 10 minutes under N_2 atmosphere. In each, the outer appearance is on the left and a cross-sectional optical microscope image is on the right.

Fuel debris at the TEPCO's Fukushima Daiichi NPS (1F) is assumed to be stored in water during transportation and interim storage for cooling after removal from reactors. Therefore, it is important to understand the leaching behavior of fuel debris components into water, especially nuclear materials such as uranium (U), for a safety evaluation of fuel debris transportation and interim storage in water. However, few data on U leaching in water are available; none of these available data come from fuel debris rates. Thus, a leaching test using simulated fuel debris was performed to acquire U leaching rates from fuel debris.

Some samples of the simulated fuel debris are shown in Fig.1-2. Two types of fuel debris have been assumed to be present in 1F: melt of fuels and structural materials, which is generated in reactor pressure vessels, and molten core–concrete interaction (MCCI) products generated in the lower parts of pressure containment vessels. Thus, samples with and without concrete components were prepared.

The simulated fuel debris were immersed in deionized water at approximately 25 °C and irradiated with gamma rays from a ^{60}Co source (approximately 80 Gy/h) in air for 100 days. Here, gamma irradiation was used to generate hydrogen peroxide by radiolysis of water, which is expected to affect leaching rates. Furthermore, to limit the production of nitric acid from nitrogen (N_2) in air due to the gamma rays, the volume ratio of the gas phase to the water phase was set to approximately 0.1.

The resulting colloid in the sampled solution was dissolved by adding nitric acid; the U concentration in the solution was then analyzed using inductively coupled plasma mass spectrometry (ICP-MS). After removing the solution, the container and sample holder were immersed in 0.5 N nitric acid for 24 h for precipitates dissolution; the U concentration in the immersing solution was analyzed. The total amount of U in the solution and precipitate was obtained from these analyses, allowing the leached amount of U to be calculated. The leaching rate was then calculated by dividing the leached amount by the surface area of the pre-test sample and the leaching period. The test was conducted at ATALANTE facility of the French Alternative Energies and Atomic Energy Commission (CEA) with the cooperation of CEA.

The resulting leaching rates of U in the samples with and without concrete components were 2.53 and 2.86 mg/m^2/d respectively, and were found to be close. Previous studies have reported U leaching rates of spent fuels in water are 2 mg/m^2/d or 83 mg/m^2/d according to two typical reports. The U leaching rates of simulated fuel debris obtained in this study therefore do not differ from those of spent fuels.

The leaching rates of fuel debris in 1F thus concluded similar to those of spent fuels regardless of the presence of concrete components. When transportation and storage are applied for 1F fuel debris, the equipment and facilities of spent fuels will be helpful. In addition, the U leaching rates obtained in this study can be utilized as basic data to evaluate behavior for various situations in which fuel debris may come into contact with water, such as transportation, storage, retrieval, and disposal.

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Reference
Nakayoshi, A. et al., Leaching Behavior of Prototypical Corium Samples: A Step to Understand the Interactions between the Fuel Debris and Water at the Fukushima Daiichi Reactors, Nuclear Engineering and Design, vol.360, 2020, p.110522-1–110522-18.