2–4 Accident Simulation: HLLW Evaporation to Dryness

−Evaluating the Restraint Effect on the Release of Volatile Ruthenium−

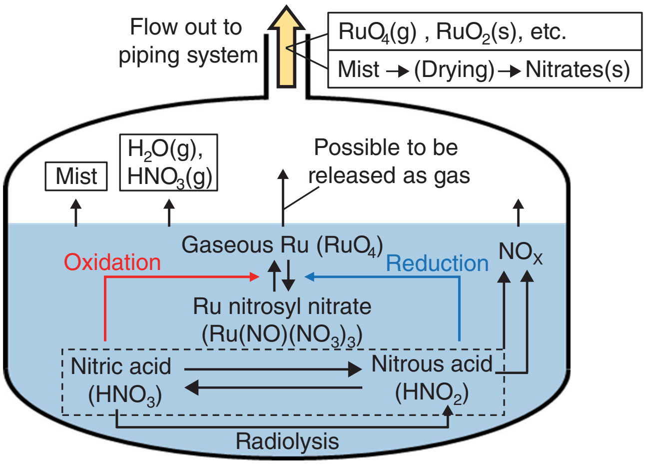

Fig.2-12 HLLW tank during an evaporation and dryness accident
The flow of gases and mists from high-level liquid waste (HLLW) is summarized here. During this type of accident, water vapor (H_2O(g)), nitric acid vapor (HNO_3(g)), NO_x, HLLW mist, and volatile compounds comprising mainly RuO_4 could be released.

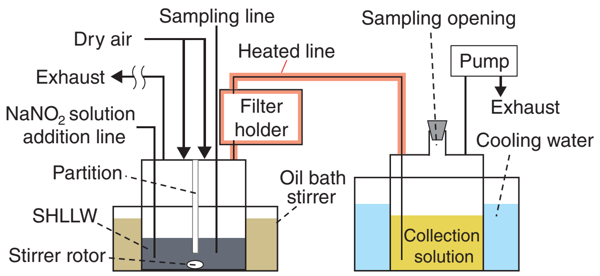

Fig.2-13 Experiment device for boiling SHLLW and collecting RuO_4
The simulated HLLW (SHLLW) was heated to keep a constant release rate of vapor and maintained at a temperature of approximately 103 °C. Generated gases were carried to the collection solution by the input dry air ventilation. The filter on the heated line catches mist, thus allowing only gaseous compounds to be passed to the collection solution. By adding sodium nitrite ($NaNO_2$) solution in the SHLLW, NO_x are generated and they cause additional mist. The partition minimized the effect of releasing mist caused by adding $NaNO_2$ solution.

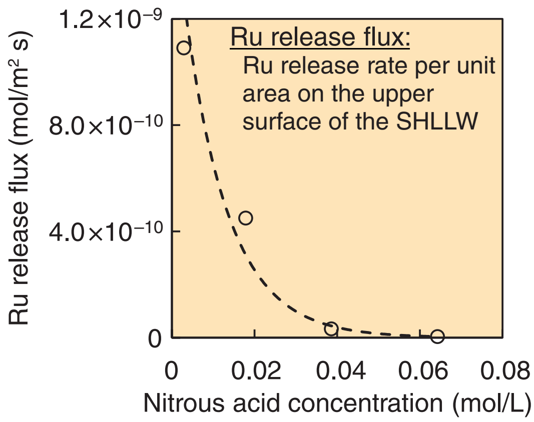

Fig.2-14 Relationship between nitrous acid concentration and Ru release flux
The circles show experimental data and the dotted line shows exponential fitting line. The Ru release flux decreased exponentially as the concentration of nitrous acid in the SHLLW increased.

High-level liquid waste (HLLW) containing radioactive elements is temporarily stored in reprocessing plants, where uranium and plutonium are extracted from the spent nuclear fuels and the HLLW must be cooled continuously to remove decay heat. If the cooling functions are lost and countermeasures do not work sufficiently, the temperature of the HLLW may increase above the boiling point, the water will evaporate and the HLLW will dry up. This evaporation to dryness due to the loss of cooling functions has been defined as a severe accident (i.e., beyond the design basis) of the reprocessing plant; sufficient countermeasures to suppress the release of radioactive materials must be developed.

Since ruthenium (Ru) forms volatile compounds, such as ruthenium tetroxide (RuO_4), more Ru would be released from the HLLW to out of the reprocessing plant than other elements in such an accident because RuO_4 cannot be trapped in filters. Therefore, properly assessing the safety of a reprocessing plant requires an understanding of the release behavior of Ru.

RuO_4 is generated by the oxidation of ruthenium nitrosyl nitrate by nitric acid in the HLLW. On the other hand, RuO_4 can be reduced by nitrous acid, which is generated by the radiolysis of nitric acid in the HLLW, thereby reducing the release of RuO_4 (Fig.2-12).

To clarify the relationship between the nitrous acid concentration and the amount of RuO_4 released from boiling HLLW, simulated HLLW (SHLLW) prepared with non-radioactive compounds was heated under fixed conditions; the experimental design is summarized in Fig.2-13. For the nitrous acid concentration in the SHLLW keeping constant, sodium nitrite solution was continuously added. The amount of RuO_4 released from the boiling SHLLW was trapped in the collection solution and measured.

As shown in Fig.2-14, increasing the nitrous acid concentration caused less RuO_4 to be released. More specifically, increasing the nitrous acid concentration led to an exponential decrease in the Ru release flux. This result contributes to the quantitative analysis of released radioactive isotopes during reprocessing plant accidents.

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Reference

Yoshida, R. et al., Restraint Effect of Coexisting Nitrite Ion in Simulated High Level Liquid Waste on Releasing Volatile Ruthenium under Boiling Condition, Journal of Nuclear Science and Technology, vol.58, issue 2, 2021, p.145–150.