Release Behavior of Radioactive Materials at a Boiling Accident of High Active Liquid Waste in Reprocessing Plants - 14079

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ABSTRACT

The experimental study for source term data of radioactive materials has been conducted at a boiling accident of high active liquid waste (HALW) in reprocessing plants. In the study, three kinds of tests have been conducted including a cold small scale test, a cold engineering scale test and a hot small scale test.

In the cold small scale test using a non-radioactive simulated HALW, the release behavior of fission product (FP) elements such as Ru and Cs from the simulated HALW were investigated under various boiling accident conditions. As a result of the test, Ru was found to be a volatile element under the accident conditions and to be released into the gas phase in the form of both mist and gas. Also other FP elements such as Cs were found to be non-volatile and to be released into the gas phase in the form of mist.

In the cold engineering scale test, the release behavior of FP elements at boiling accident conditions was investigated mainly as a spatial function in the vessel type equipment. The major part of FPs released from the simulated HALW to the gas phase was deposited on the surface of inner liner of the vessel type equipment. The release fraction of Ru obtained in the vessel type equipment was almost the same as that of the cold small scale test.

In the hot small scale test using a radioactive simulated HALW, the release behavior of radioactive materials were obtained under typical boiling accident conditions. The release fractions of Ru and non-volatile FPs obtained in the hot small scale test were almost the same as those of the cold small scale test.

INTRODUCTION

Renewed Japanese nuclear safety standards for reprocessing plants which reflect lessons learned from the accident at the Fukushima Dai-ichi Nuclear Power Plants of Tokyo Electric Power Company (TEPCO) requires serious accident management for the first time. Accidents such as boiling of high active liquid waste (HALW), criticality, fire of organic solvents, and explosion of hydrogen gas that occur beyond Design Basis Accident (DBA) conditions are included as serious accidents in reprocessing plants. Studies on scenarios and source terms of serious accidents in reprocessing plants are needed for the development of evaluation methods of effectiveness of accident management measures. It is reported that a boiling accident of a HALW tank occurs in a reprocessing plant if it should happen that the cooling function of the tank is lost for more than 15 hours [1]. If the loss of cooling function continues further, HALW reaches dryness. Source term data for the evaluation of consequences of a boiling accident of HALW tanks were little reported [2]. Therefore, this study has been conducted to develop experimental data of radioactive material source terms at a boiling accident of HALW in a reprocessing plant under an agreement among Japan Atomic Energy Agency (JAEA), Japan Nuclear Fuel Ltd. (JNFL) and Japan Nuclear Energy Safety Organization (JNES) [3-8].

In the study, three kinds of tests are conducted including a cold small scale test, a cold engineering scale test and a hot small scale test. In the cold small scale test using a non-radioactive simulated HALW, the release and transport behavior of radioactive materials such as Ru and Cs has been investigated under the various conditions at a boiling accident of HALW. Also, differential thermal analysis (TG-DTA) has been conducted mainly to take the thermal decomposition rate data of nitrate of FP elements in solid state. In the cold engineering scale test using a non-radioactive simulated HALW, the release behavior of radioactive materials at a boiling accident has been investigated mainly as a spatial function using vessel type test equipment under various boiling accident conditions, and the transport behavior of non-radioactive materials has been investigated using a duct type test equipment. In the hot small scale test using actual reprocessing waste as a simulated HALW, the release behavior of radioactive materials at boiling accident has been investigated under typical accident conditions. The aim of the hot test is to find the difference between a radioactive simulated HALW and a non-radioactive simulated HALW on the release behavior of radioactive materials.

This paper describes the results on the release behavior of radioactive materials at a boiling accident of HALW in the three kinds of tests.

COLD SMALL SCALE TEST

Experimental

The composition of the simulated HALW used in the test was based on the composition of HALW in a reprocessing plant [9].

The cold small scale test equipment consisted of a Pyrex glass vessel, a condenser, an air filter, an absorber (1 mol/l NaOH), a cascade low pressure impactor and a pump [8]. The simulated HALW (100 ml) in the vessel was heated up to about 300 ^{o}C under the constant air ventilation rate of about 1 l/min. During the tests, the vapor flow rate was 1.3 cm/s at boiling step. FP elements released from the simulated HALW were analyzed with ICP-MS. The chemical form of Ru in the condensate was analyzed by using a visible/UV spectrophotometry. The released material was flowed to a cascade low pressure impactor over 150 ^{o}C to measure particle size distribution data at drying step.

Results and Discussion

FP elements such as Ru and Cs released from the simulated HALW were distributed mainly in the condensate and the rest of them were distributed in the air filter, absorber and connecting pipes between the vessel and condenser. The time dependency of the release fraction (RF) of Ru and Cs, the temperature of simulated HALW and the cumulative volume of condensed water in the condenser are shown in Fig. 1. The RF was defined as the fraction of amount of FP element in the condensate to initial amount of FP element in the simulated HALW in Eq. (1), as FP elements were mainly distributed in the condensate.

$$\text{Release fraction (RF)} = \frac{\text{Amount of FP element in the condensate}}{\text{Initial amount of FP element in the simulated HALW}} \qquad \text{Eq. (1)}$$

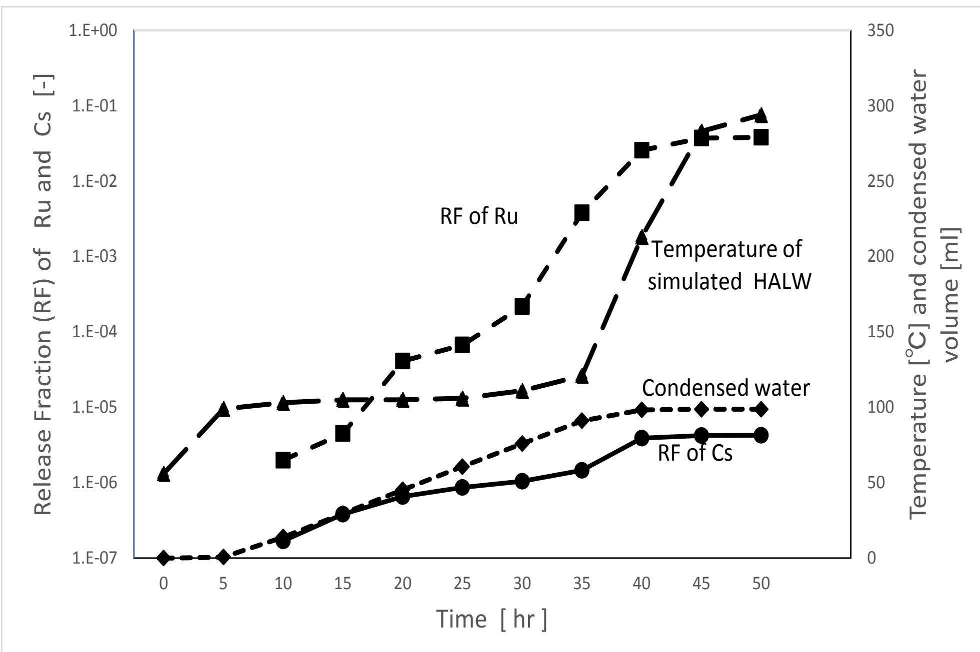

Fig. 1. Time dependency of temperature of simulated HALW, cumulative condensate volume, and the RF of ruthenium and cesium.

The temperature of simulated HALW increased to about 100 ^{o}C after about 5 minutes from the start of heating, and the simulated HALW boiled for 30 minutes. The cumulative condensate volume of water increased linearly with time until about 35 minutes from the start of heating. The simulated HALW changed to solid from liquid. The RF of Ru went up to about 2 x 10^{-2} at the end of the heating. The RF of Ru was more than that of Cs by a factor of more than 1,000. Cesium was considered to be a non-volatile element under these experimental conditions [10]. Cesium released into the gas phase was mainly in the form of mist. Ruthenium released into the gas was in the form of both mist and gaseous product which was considered to be in the chemical form of RuO_4 [10, 11]. It was considered that $RuNO(NO_3)_3$ was oxidized to gaseous RuO_4 by nitric acid in the simulated HALW and that volatilized RuO_4 was considered to form nitrosyl ruthenium by H_2O, HNO_3, NOx and O_2 in the gas phase [12].

COLD ENGINEERING SCALE TEST

Experimental

The composition of the simulated HALW used in the cold engineering scale test was the same as that of the cold small scale test. The cold engineering scale test equipment consisted of a SUS vessel with SUS inner liner, a condenser, an air filter, an absorber (1 mol/l NaOH), a cascade low pressure impactor, a mass flow controller and a pump. The simulated HALW (400 ml) was heated up to about 300 ^{o}C under the constant air ventilation rate of about 10 l/min. The temperature of the wall of the vessel was heated to prevent the deposition of water vapor released from the simulated HALW. During the tests, the vapor flow rate was about 1.1 cm/s at boiling step. FP elements released from the simulated HALW were analyzed with ICP-MS.

Results and Discussion

FP elements released from the simulated HALW were distributed in inner liner of the vessel, a condenser, an air filter, an absorber and connecting pipes. Ruthenium was distributed mainly in the condensate at the condenser. The amount profiles of FP elements deposited on the surface of inner liner of the vessel are shown against vessel height in Fig. 2. The amount of Ru was almost the same as those of other FPs such as Cs and Nd. It was considered that FP elements deposited on the inner liner corresponds to the entrainment as mist released from the simulated HALW. About 99% of Ru released from the simulated HALW was in the form of nitrosyl ruthenium in the condensate and about 75% of non-volatile FP elements such as Cs and Nd released from the simulated HALW was deposited on the inner liner of the vessel in the test.

The RFs of Ru and Cs defined in Eq. (1) were Ru: 2.2×10^{-2}, Cs: 1.5×10^{-5} at the end of the heating, respectively. The RF of volatile Ru in the cold engineering scale test was almost the same as that of the cold small scale test under the same heating conditions. On the other hand, the RFs of non-volatile FPs were a little smaller than those of cold small scale test.

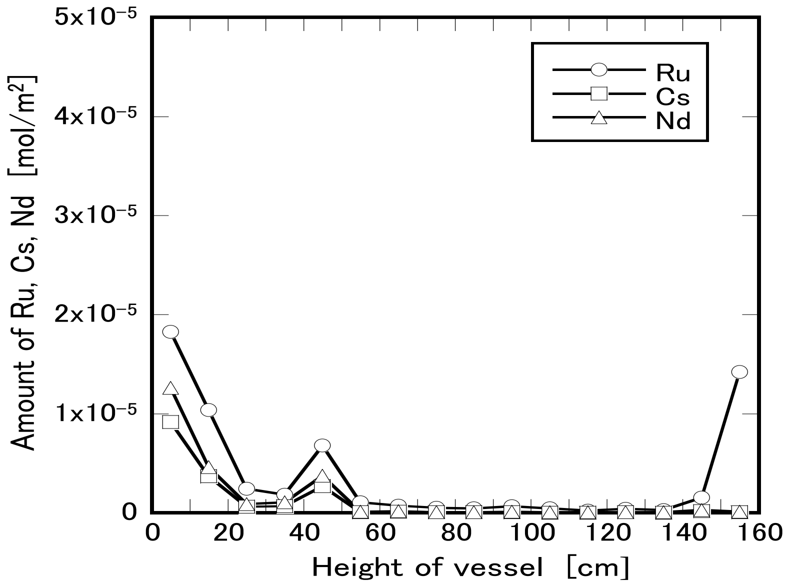

Fig. 2 Amount of Ru, Cs and Nd deposited on the inner liner of vessel against height of vessel.

HOT SMALL SCALE TEST

Experimental

A radioactive raffinate solution generated from a reprocessing experiment of spent fuel of Fugen reactor, a prototype advanced thermal reactor in JAEA was used as a simulated HALW. The hot small scale test equipment is shown in Fig. 3. The test equipment was installed in a hot cell as shown in Fig. 4. The

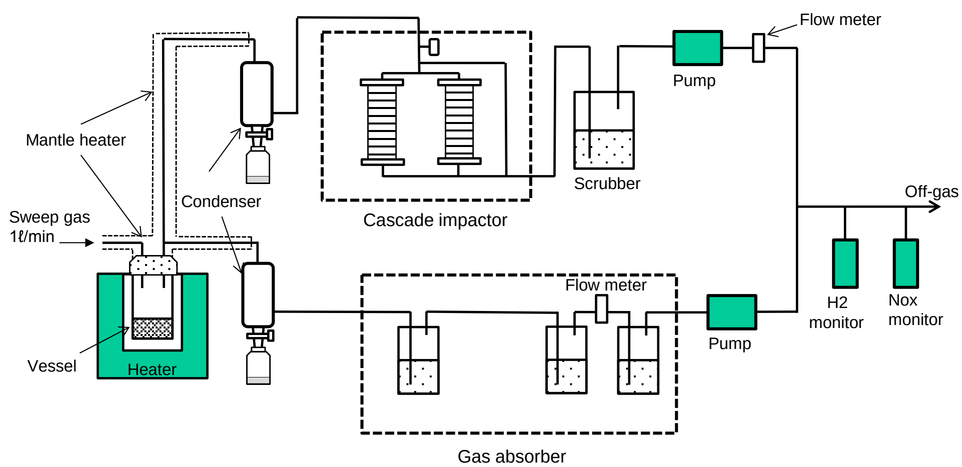

Fig.3 Hot small scale test equipment.

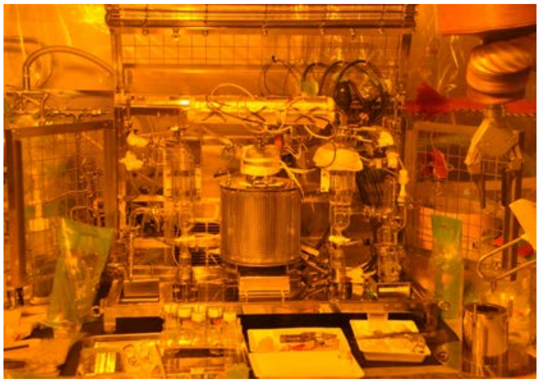

Fig. 4 Alpha-gamma hot cell where the test equipment was installed.

actual radioactive HALW (about 50 ml) was heated up to about 400 ^{o}C under the constant air ventilation rate of about 1 l/min. During the tests, the vapor flow rate was about 1.3 cm/s at boiling step. FPs and alpha nuclide released from the HALW were analyzed with ICP-MS, ICP-AES and gamma- and alpha-ray radioactivity measurements.

Results and Discussion

FP elements such as Ru and Cs released from the radioactive simulated HALW were distributed mainly in the condensate in the same as those of the cold small scale test. The time dependency of the temperature of radioactive simulated HALW and cumulative volume of condensed water is shown in Fig. 5. The time dependency of the RF of Ru defined in Eq. (1) is shown in Fig. 6. The RFs of FPs gradually

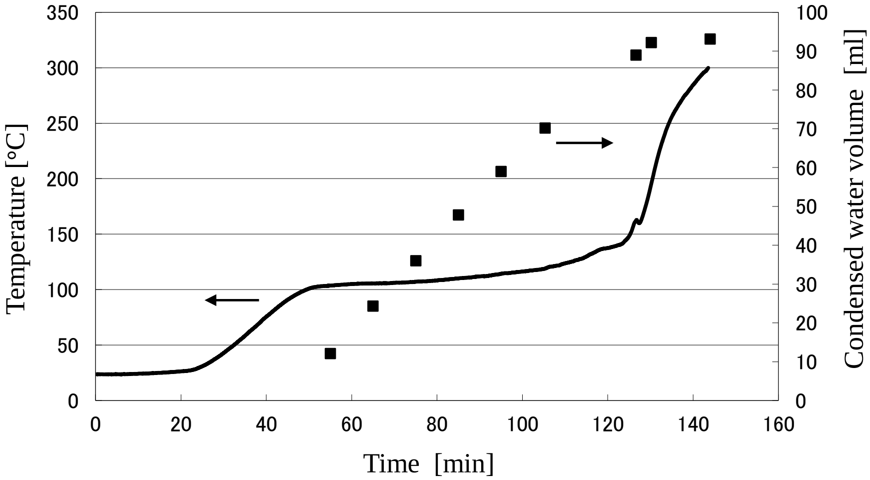

Fig. 5 Time dependency of temperature of radioactive simulated HALW and cumulative volume of condensed water.

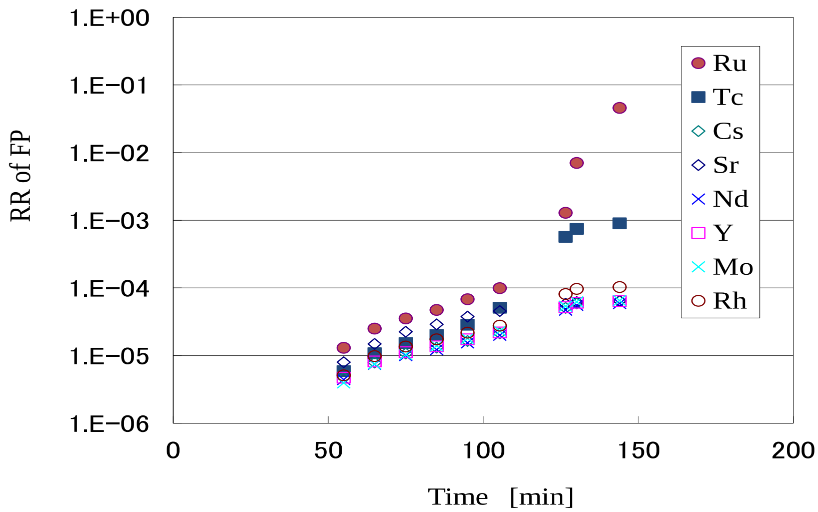

Fig. 6 Time dependency of RF of FPs.

increased in the same way until about 120 minutes (about 120 ^{o}C) but after that Ru and Tc increased more rapidly than other FPs such as Cs and Nd. The RFs of Ru and Tc went up to about 5 x 10^{-2} and about 1 x 10^{-3}, respectively at the end of the heating. The RF of FPs other than Ru and Tc were almost about 1 x 10^{-4}. The RFs of alpha nuclides such as Am-241, Pu-238 and Cm-242 were almost the same as those of Cs and Nd. Alpha nuclides and FPs other than Ru and Tc were considered to be non-volatile nuclides under these experimental conditions [10]. It was considered that those nuclides released into the gas phase were mainly in the form of mist and that Ru and Tc released were in the form of not only mist but also gaseous products.

Comparing the time dependency of RFs of FPs between hot test result (Fig. 6) and cold test result (Fig. 1), the RF of non-volatile FP increasing gradually with time in the hot test is almost the same as that of cold small scale test. However, the RF of Ru in the boiling step in the hot is depressed compared to that of the cold test. The RF of Ru in the boiling step in the hot test is the almost the same as that of non-volatile FPs in the cold small scale test. It was considered that nitrous acid of a radiolysis product of nitric acid in the highly radioactive solution prevented nitric acid from oxidizing $RuNO(NO_3)_3$ to RuO_4 [8].

The RFs of Ru and non-volatile FPs obtained under typical conditions in the hot small scale test were almost the same as those of the cold small scale test. It was found that there was little difference between a radioactive simulated HALW and a non-radioactive simulated HALW on the release behavior of FP elements.

These test results will contribute to appropriate evaluation of accident consequences of HALW boiling in reprocessing plants. Further study is required to improve experimental data of FP source terms at the accident.

CONCLUSIONS

The release behavior of radioactive materials at a boiling accident of HALW in a reprocessing plant were studied for the improving experimental data of source terms of the accident. The results are as follows:

In the cold small scale test using a non-radioactive simulated HALW, the release behavior of FP elements such as Ru and Cs was obtained under various conditions at boiling accident conditions of a simulated HALW. Ruthenium was released into the gas phase in the form of both mist and gas. Non-volatile Cs was released into the gas phase in the form of mist.

In the cold engineering scale test, the release behavior of FP elements at a boiling accident was investigated mainly as a spatial function in the vessel type equipment. The RF of Ru obtained in the vessel type equipment was almost the same as that in the cold small scale test.

In the hot small scale test using an actual reprocessing waste, the release behavior of radioactive materials at a boiling accident was obtained under typical boiling accident conditions. The RFs of Ru and non-volatile FPs obtained in the hot small scale test were almost the same as those of the cold small scale test. It was found that there was little difference between a radioactive simulated HALW and a non-radioactive simulated HALW on the release behaviors of FP elements.

These test results will contribute to improve experimental data of FP source terms for appropriate evaluation of accident consequences of HALW boiling in reprocessing plants.

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