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STUDY OF SOURCE TERM EVALUATION FROM FUEL SOLUTION UNDER SIMULATED NUCLEAR CRITICALITY ACCIDENT IN TRACY

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INTRODUCTION

A criticality accident at the dissolver is assumed one of the design basis accidents (DBA) for the fuel reprocessing plant. In the accident, various fission products and radiolysis gases will be produced in the fuel solution and volatile radioactive nuclides (iodine, xenon and krypton) and radiolysis gases (hydrogen (H_2) and nitrogen oxide (NOx)) will be released into vent-gas spontaneously. Moreover, other non-volatile nuclides will be released from the fuel solution as radioactive aerosol (mist) with bursting bubbles at surface of the solution. Bubbles are produced from the radioactive and radiolysis gases in the solution. The vent-gas including the radioactive materials will be transported in a ventilation system having air-cleaning filters and, finally, released to the atmosphere. Therefore, quantitative estimation of release and transport behavior of the radioactive materials from the fuel solution as source term is very important for safety evaluation of the plant. NUCEF-TRACY is a transient criticality experimental facility for studying the transient criticality characteristics of low-enriched uranium fuel solution and the characteristics are investigated under the various conditions of the excess reactivity and reactivity addition rate. To verify the confinement capability for the released radioactive materials of the ventilation system in the plant and the safety margin for the values adopted in the safety evaluation of the plant, data concerning the release and transport of these materials will be acquired during simulation experiments in TRACY.

Design and establishment of various experimental devices have been carried out since fiscal 1994 and three experiments for evaluating release behavior of the radioactive materials and radiolysis gases from the fuel solution were performed in fiscal 1996 and 1997. In the present paper, experimental methods and results about the release behavior of the H_2, radioactive aerosol and iodine species from the fuel solution are reported.

EXPERIMENTAL

Experimental devices and methods

In TRACY, there is a vent-gas line connected to the core tank and vent gas circulates through the line. The radioactive materials and radiolysis gases, released from the fuel solution, effuse into the line during the transient criticality. To observe release and transport behavior of those compounds, the line is equipped with a cascade impactor for sampling mist and iodine (Mst), Maypack samplers for iodine (M), Andersen-type cascade impactors for aerosol particles (C) and gas concentration measurement devices for H_2, NOx and radioactive noble gases (G) as shown in Figure 1.

Mst can collect directly the mist and aerosol released from the fuel solution to the gas phase in the TRACY core tank. Therefore, it can be regarded that the concentration of radioactive

materials estimated with Mst represents one in the gas phase in the core tank. Mst consists of a four stage impactor for collecting the mists with liquid collector, one membrane filter and iodine collection element (ICE), such as five silver plated fibrous filters, two silver nitrate impregnated alumina beds. Inorganic iodine species such as I_2 and organic iodine species such as CH_3I are collected separately in the respective components of ICE. Mst is equipped with back-up samplers including ten silicagel beds for collecting moisture in the sampling gas. The radioactive aerosol and iodine species, which effuse into the vent-gas line from the core tank, are collected with C and M samplers. The each C sampler consists of a seven stage impactor for collecting the aerosols, one membrane filter and ICE, and the each M sampler consists of ICE. Sampling lines at upstream of the all samplers were heated up to 80 ℃. After sampling, the respective samplers was taken apart to each component and the radioactive nuclides in the components were determined by gamma-ray spectrum analysis.

As the H_2 concentration measurement device, a catalytic combustion type thermal conductivity detector (Yanako-ogi, TC-201S) is adopted. CLM-500 (Shimazu-seisakujyo) measures the NOx gas concentration in the vent-gas by using chemiluminescence method for oxidative reaction from NO to NO_2 by O_3.

Moreover, several thermometers, pressure gauges and flow meters are also attached to the vent-gas line for observing thermofluid conditions of the vent-gas.

Experimental conditions

Three experiments with different reactivity conditions have been performed using the devices as shown in Table 1. Figure 2 and Figure 3 show the time-course of fission number per second of the respective experiments. R75 was performed under the ramp withdrawal mode of a transient rod (300 cm/min). Since the insertion time of the rod was put off than other two experiments, the time-course of fission number per second oscillated and several fission peaks occurred for about 120 sec. On the other hand, in two other experiments, which were performed under the pulse withdrawal mode of the rod (about 36,000 cm/min in average), one sharp fission peak with a few milliseconds wide was observed.

To collect radioactive iodine which is emerge from the solution with a time lag after the transient criticality, the fuel solution had been kept in the core tank for 5 h after the transient criticality. The volumes of the fuel solution and the gas phase in the core tank were 0.2 m^3 and 0.18 m^3, respectively. The volume flow rate of the vent-gas was about 0.28 m^3/h.

RESULTS AND DISCUSSIONS

Release behavior of H_2 gas from the fuel solution

Figure 4 shows time-courses of the H_2 concentration in the vent-gas obtained from R69 and R75 experiments. The H_2 concentrations in the vent-gas attained to the peak just after the transient criticality and decreased exponentially with time. The maximum concentrations increased with increasing the total fission number. The maximum concentrations and total volumes of H_2 for 5 hours after the transient criticality were 9,000 ppm (0.9 %) and 0.23 m^3 for R69, and 14,000 ppm (1.4%) and 0.33 m^3 for R75, respectively. The maximum concentrations were lower than the lower limit concentration of combustion of H_2, about 4 %.

Release behavior of radioactive aerosol and iodine species

By gamma-ray spectrum analysis of the radioactivity adhered to each stage of Mst, ^{140}Ba-^{140}La, daughter nuclides of ^{140}Xe, and ^{131}I were only detected. Other non-volatile radioactive nuclides produced by fission in the fuel solution were not detected. This fact indicated that

detectable amount of mist was not released from the solution.

The concentration of ^{140}Ba in the gas phase of the core tank attained to the maximum just after the transient criticality and decreased exponentially with time as shown in Figure 5. Although the total fission number on R75 was larger than that on R69, the first peak height of fission number per second on R75 was lower than that on R69 because of the difference in reactivity addition mode between them. This fact explains that the concentration of ^{140}Ba on R75 early in the time-course after the transient criticality was lower than that on R69.

The concentrations of ^{131}I in the gas phase of the core tank began to increase with a time lag of several minutes from the transient criticality and attained approximately constant values, as shown in Figure 6. The values were different according to the total fission number of each experiment. The values at 85 min after the transient criticality were 7.3×10^{2} Bq/m^3 for R35, 4.5×10^{4} Bq/m^3 for R69 and 3.0×10^{5} Bq/m^3 for R75, respectively.

Evaluation of release ratio of ^{131}I from the fuel solution

In this study, the time-course of the number of iodine atoms in the fuel solution and that in the gas phase in the core tank were calculated by using the simultaneous equations as follows.

$$\frac{dI_{sol}}{dt} = Fs \times \eta_I + \lambda_p \times P_{sol} - (\lambda_I + \xi_I) \times I_{sol} \quad (1)$$

$$\frac{dI'_{sol}}{dt} = Fs \times \eta_I + \lambda_p \times P_{sol} \quad (2)$$

$$\frac{dI_{gas}}{dt} = \xi_I \times I_{sol} - (\lambda_I + \psi) \times I_{gas} \quad (3)$$

$$\frac{dI'_{gas}}{dt} = \xi_I \times I_{sol} \quad (4)$$

where, Fs (fissions/s) is the fission number per second, η (-) is the fission yield, λ (1/s) is the radioactive decay constant, ξ (1/s) is the release constant from the fuel solution, ψ (1/s) is the dilution constant in the gas phase of the core tank by the vent gas. The subscripts "I" and "P" mean the values of ^{131}I atom and parent nuclides of ^{131}I, and, "sol" and "gas" mean the values in the fuel solution and in the gas phase of the core tank. I'_{sol} and I'_{gas} are the time-integrated numbers of ^{131}I atoms in the fuel solution and those in the gas phase. They were calculated by omitting decrease of the number of ^{131}I atoms by radioactive decay and release from the fuel solution or dilution and discharge by the vent gas. Moreover, the release ratio, R (%), of the ^{131}I from the fuel solution to the gas phase was defined as the following equation;

$$R = \frac{I'_{gas}}{I'_{sol}} \times 100 \quad (5)$$

If it is assumed that the time trend of I_{gas} in Figure 6 can be also represented by Eq(6) and Eq(7), ξ_I is calculated from Eq(8) by combining Eq(3) and Eq(7).

$$I_{gas} = \alpha \times exp(\beta \times t) \quad (6)$$

$$\frac{dI_{gas}}{dt} = \alpha \times \beta \times exp(\beta \times t) \quad (7)$$

$$\xi_I = \frac{\alpha \times exp(\beta \times t) \times (\beta + \lambda_I + \psi)}{I_{sol}} \quad (8)$$

At first, α (-) and β (-) were estimated from relations between time and I_{gas} as shown in Figure 6, next, ξ_I was calculated by using Eq(8). Moreover, R was estimated by solving from Eq(1) to Eq(5) simultaneously by Runge-Kutta-Gill method. As for the nuclear data of ^{131}I, η and λ, respective values in JNDC nuclear data library[1] were used. ψ was determined from an experiment using the mock-up vessel that has the same dimension and ventilation flow as the TRACY core tank. ψ could be estimated as 2.53×10^{-2} (1/s) from the slope of linear relationship between time and the aerosol number concentration in the vessel shown in Figure 7. Fs for R35 and R69 was estimated by assuming that it was equal to total fission number at just transient criticality point (t=0) and zero in the time range after the point. For R75, Fs was estimated directly by approximation of the oscillated time-course of fission number per second, as shown in Figure 8.

The estimated results of R of ^{131}I are shown in Figure 9. As seen in Figure 8, R is large when the total number of fission is large. The values of R are 6.5×10^{-4} % for R35 and 4.7×10^{-2} % for R75 at 4.5 h after the transient criticality. These values were much lower than the value, 25 %, which was adopted in the DBA scenario.

CONCLUSION

To evaluate release behavior of radioactive materials and radiolysis gases from the fuel solution under the simulated criticality accident, design and establishment of various experimental devices have been carried out and three simulation experiments were performed.

As the results of the experiments, release patterns of H_2, ^{140}Ba and ^{131}I could be grasped. Concentrations of H_2 in the vent-gas and ^{140}Ba in the gas phase in the core tank attained to the peak just after the transient criticality and decreased exponentially with time. On the other hand, concentrations of ^{131}I in the gas phase in the core tank began to increase with a time lag of several minutes from the transient criticality and attained approximately constant values. Maximum concentrations of H_2 and ^{131}I increased with increasing total fission number. Moreover, a time-course of release ratio of ^{131}I from the fuel solution could be estimated by assuming simultaneous equations for representing time-course of number of ^{131}I atoms.

In future, release behavior of the radioactive noble gases will be investigated as function of various reactivity conditions because they have large effect on the public dose. And, comprehensive and detailed calculation models for simulating release behavior of the materials will be estimated.

ACKNOWLEDGMENTS

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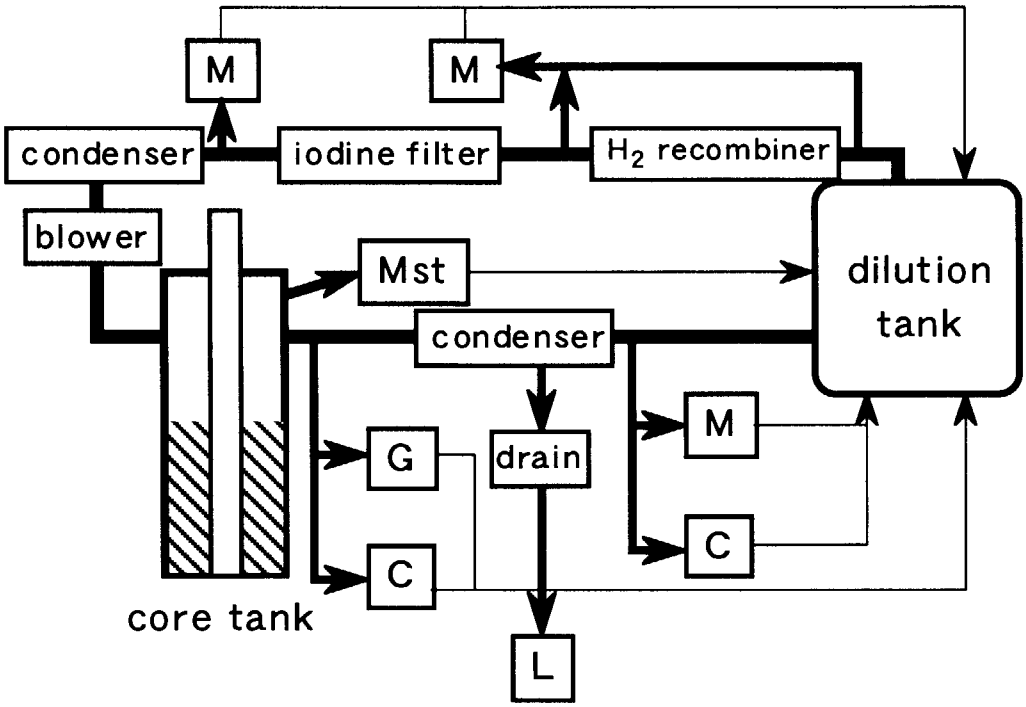
REFERENCES

(1) TASAKA.K., *et al.* : *JAERI* 1320 (1990).

Table 1 Parameters of respective experiments

Run No.	Reactivity($)	Reactivity addition mode	Total fission number
R35	1.8	pulse withdrawal	2.9×10^{17}
R69	2.9	pulse withdrawal	5.7×10^{17}
R75	2.9	ramp withdrawal (300cm/min)	6.9×10^{17}



Mst : Cascade impactor for mist and iodine
M : Maypack sampler for iodine
C : Andersen-type cascade impactor for aerosol particles
G : Gas monitors (for H_2, NOx and radioactive noble gases)
L : Drain sampler

Figure 1 A scheme of the vent-gas line of TRACY and sampling and measuring devices.

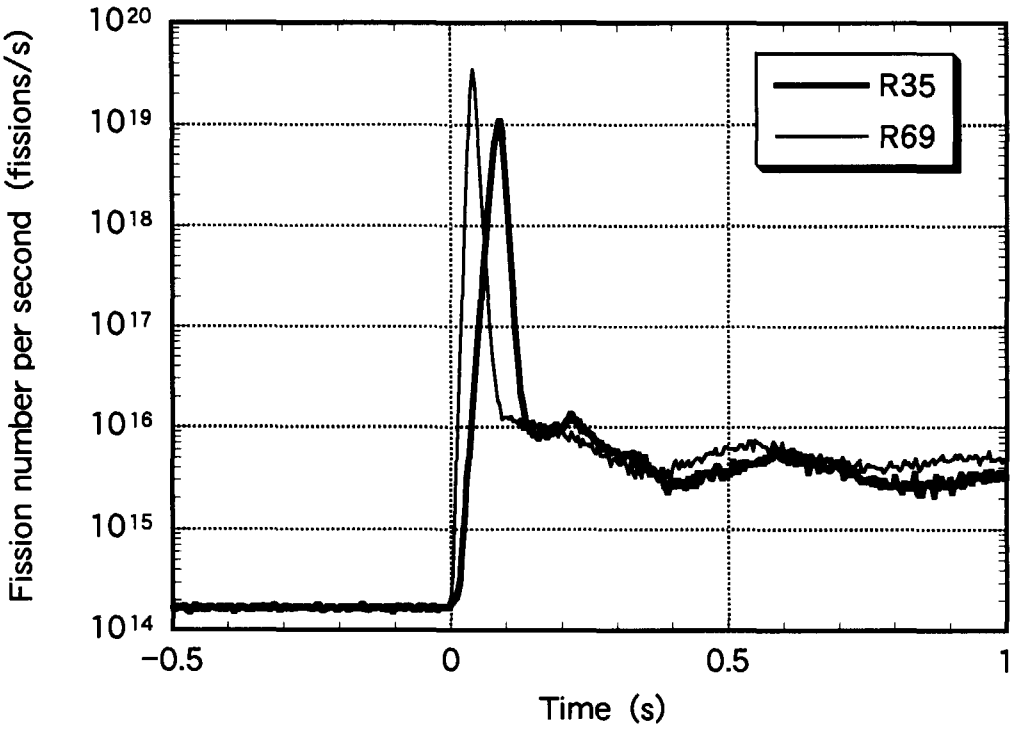


Figure 2 Time-course of fission number per second on R35 and R69

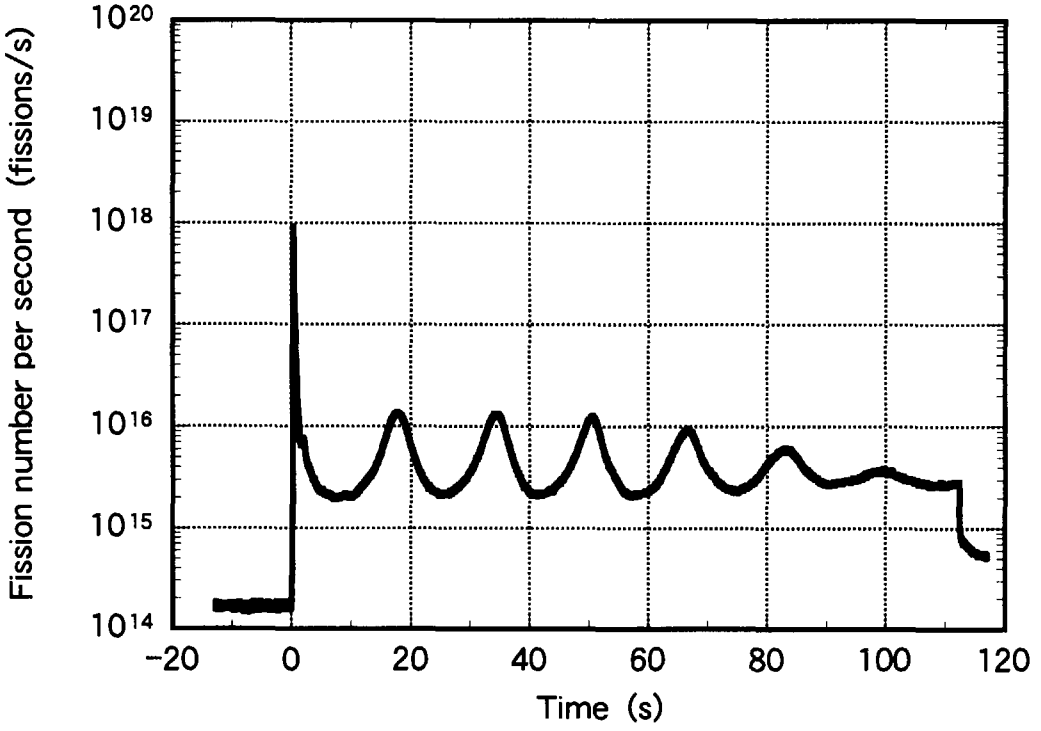


Figure 3 Time-course of fission number per second on R75

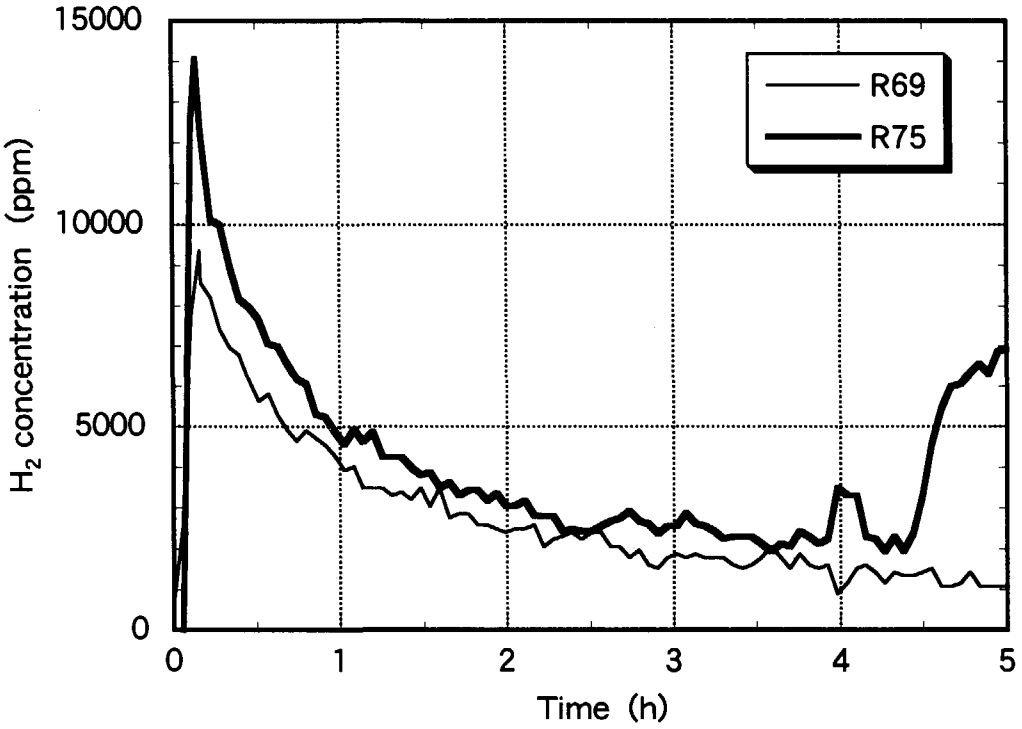


Figure 4 Time-course of H_2 concentration in the vent-gas on R69 and R75

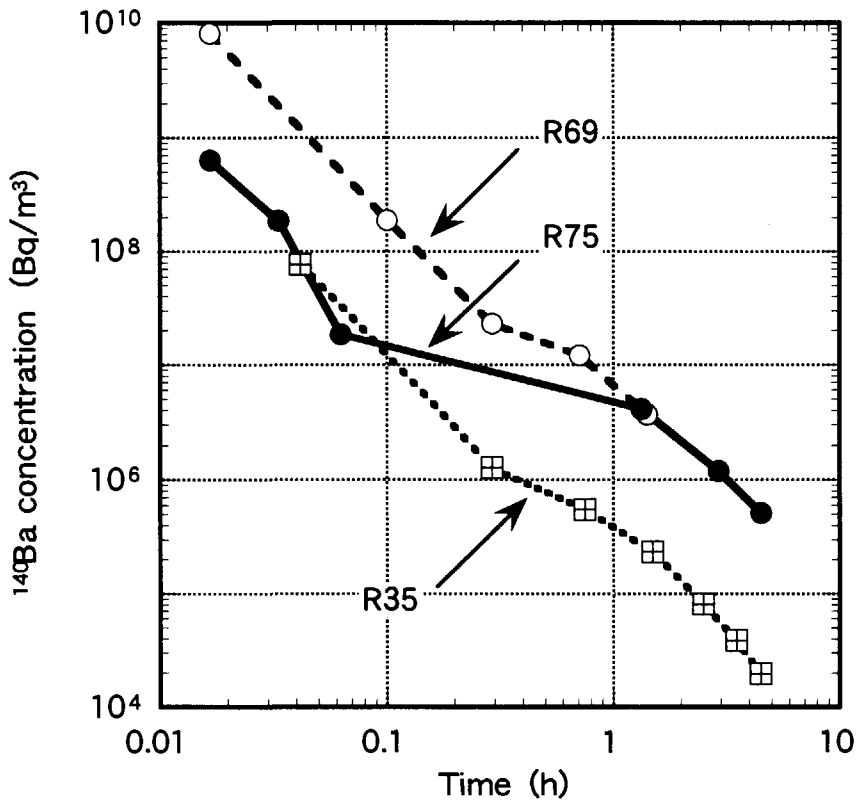


Figure 5 Time-course of ^{140}Ba concentration in the gas phase in the core tank on R35, R69 and R75

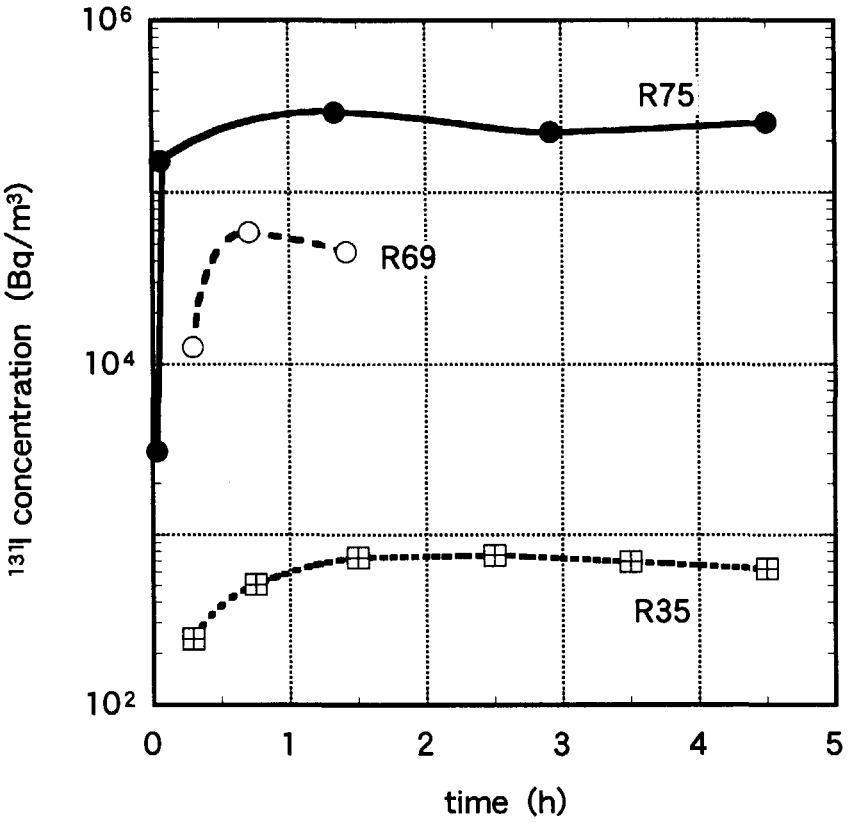


Figure 6 Time-course of ^{131}I concentration in the gas phase of the core tank on R35, R69 and R75

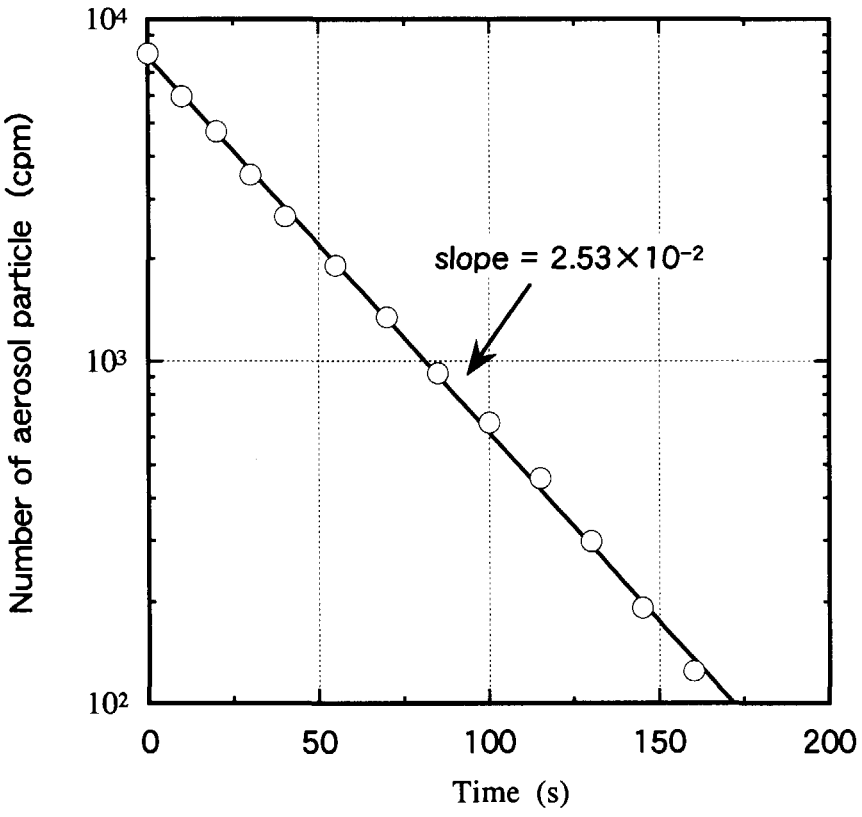


Figure 7 Time-course of the number of aerosol particle in the mock-up vessel

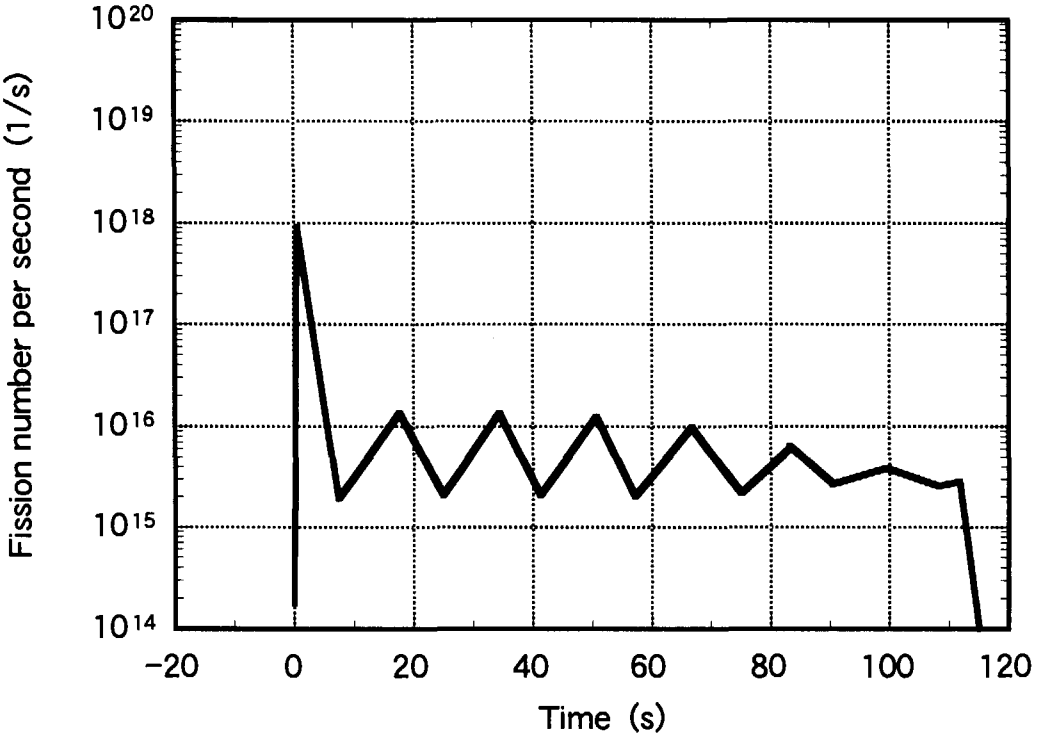


Figure 8 Approximation of the oscillated time-course of fission number per secound on R75

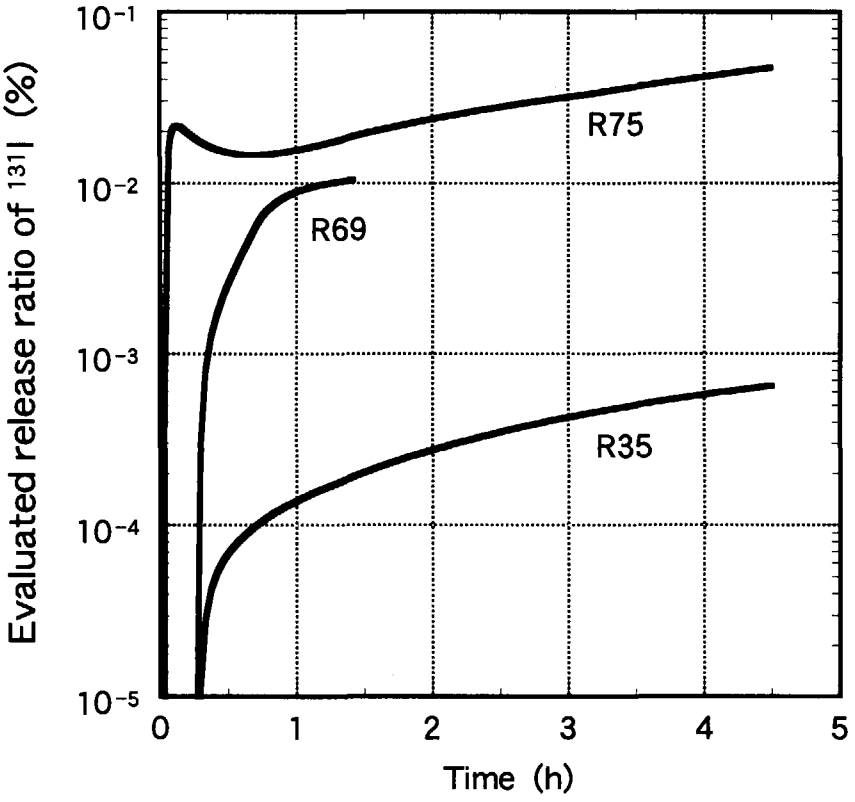


Figure 9 Time-course of evaluated release ratio of ^{131}I from the fuel solution to the gas phase in the core tank