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RELEASE RATE OF NON-VOLATILE FISSION PRODUCTS DURING SODIUM-CONCRETE REACTION

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POWER REACTOR AND NUCLEAR FUEL DEVELOPMENT CORPORATION

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RELEASE RATE OF NON-VOLATILE FISSION PRODUCTS DURING SODIUM-CONCRETE REACTION

S. Miyahara, K. Haga and Y. Himeno

Safety Engineering Division
O-arai Engineering Center
Power Reactor and Nuclear Fuel Development Corporation
4002 Narita, O-arai machi, Ibaraki 311-13, Japan

ABSTRACT

A series of tests was conducted to study the mechanical release of non-volatile fission products during sodium-concrete reaction, in which hydrodynamic break-up by the hydrogen bubble bursting is predominant at the sodium pool surface.

In the tests, non-radioactive materials, namely strontium oxide, europium oxide, and ruthenium particles whose sizes ranged from some microns to several tens of microns, were used as simulated non-volatile fission products.

The following results were obtained from the present study:
(1) The sodium aerosol release rate during the sodium-concrete reaction was larger than that of natural evaporation. The difference, however, became smaller with the increase in the sodium temperature; nearly 10 times at 400 ℃ and 3 times at 700 ℃.
(2) Retention factors of these non-volatile materials in sodium pool increased in the range of 0.5 to 10^4 with the increase in the sodium temperature from 400 ℃ to 700 ℃.

INTRODUCTION

In the vessel melt-through sequences of hypothetical core disruptive accidents in a liquid metal fast breeder reactor, it is postulated that the hot sodium containing fission products drops on a steel liner in the reactor cavity. When the dropped sodium is heated up to a very high temperature by decay heat, the steel liner may experience failure leading to a sodium-concrete reaction. During the reaction, hydrogen gas bubbles emerging from the reaction layer agitate the sodium pool, and enhance the sodium aerosol generation due to the hydrodynamic break-up by the bubble bursting at the pool surface. Involved in the released aerosols, the particles of the non-volatile fission products (n-VFPs) will also be released from the pool.

So far, many experimental and code development studies[1-6] have been conducted on the thermal and chemical processes of the sodium-concrete reaction. However, no experimental data was available on the releases of sodium aerosols and n-VFPs that were essential for assessing the source term in the later phase of the accidents.

Therefore, two types of test have been conducted to determine the release rate of sodium aerosol and the retention factor (R.F. : the ratio of the mass concentration of n-VFPs in the sodium pool to that in the sodium aerosol) of n-VFPs in sodium pool; one is the sodium-concrete reaction test to demonstrate the increase in sodium aerosol release and the retention capability of n-VFPs in liquid sodium pool, and the other is the simplified test in which agitation of sodium pool by hydrogen gas during the sodium-concrete reaction was simulated by gas bubbles from a nozzle located at the bottom of the sodium pool. The later is to interpret the effects of sodium pool temperature and bubbling gas flow rate on the aerosol release rate and R.F. because the results of sodium-concrete reaction test were obtained in the complicated conditions.

I. EXPERIMENT

1. Test Rig

(1)Sodium-concrete reaction test

Figure 1 shows an arrangement of the test rig for the sodium-concrete reaction. The test rig mainly consists of three parts; a concrete block, a sodium pool, and a cover gas space.

The concrete block of graywacke aggregates was prepared in a stainless steel bucket. The block is 300 mm high and 203.3 mm in diameter. Seven thermocouples were embedded at various axial locations in the concrete to measure the temperature profiles during the test.

The instruments in the sodium pool are three thermocouples for measuring temperature in the sodium pool and a wave guide with an acoustic sensor attached on the outer surface of the test vessel for monitoring hydrogen evolution regime.

The cover gas space over the sodium pool is about 850 mm high and about 27.5 litters in volume. Thermocouples are installed to measure the temperature of the cover gas space and the lower surface of the top flange of test vessel. A pressure gauge, a gas flow meter, settling aerosol samplers, and a rupture disk are mounted directly on the vessel or in the vent line that connects the vessel with a gas scrubber. Aerosol sampling filters made of sintered stainless steel are connected to the vent line on the both sides of the flange.

The test data is recorded automatically by using a computer controlled data acquisition system.

(2)Simulation test by gas bubbling

Figure 2 shows an arrangement of the test rig for the simulation test. The main part of the test rig is a stainless steel vessel with its axial length of 1,200 mm and with its inner diameter of 202.3 mm. A gas bubbling nozzle was installed in the bottom of the vessel and nitrogen gas was blown into sodium pool from the bubbling nozzle to simulate hydrogen evolution in sodium-concrete reaction.

Other arrangement of the test rig is almost the same as that of the sodium-concrete reaction test rig described above.

2. Test Conditions and Procedures

Test conditions in the sodium-concrete reaction test and the simulation test are shown in Table I.

(1)Sodium-concrete reaction test

Non-radioactive particles of strontium oxide (SrO), europium oxide (Eu_2O_3), and ruthenium (Ru) were scattered at the same time on the concrete surface as simulated n-VFPs. Then, about 10 kg of sodium at the temperature of 600 ℃ were poured onto the concrete surface and were being heated to keep the temperature by electrical heaters surrounding the test rig. The cover gas and the top flange of the test rig were maintained at a constant temperature of 300 ℃ during the test. Released aerosols and simulated n-VFPs were collected by using both aerosol sampling filters and settling samplers.

(2)Simulation test by gas bubbling

The bubbling gas flow rate and the sodium temperature ranged from 1 $Nl/m^2/sec$ to 10 $Nl/m^2/sec$ and from 400 ℃ to 700 ℃, respectively. Regarding the simulated n-VFPs, particles of SrO and Eu_2O_3 with various diameter were used to obtain data on the effect of particle size on the R.F.s.

II. RESULTS AND DISCUSSION

Because the detailed explanation on thermal and chemical processes of the sodium-concrete reaction has been given in papers,[1-6] only the results related to the aerosol release rate and R.F. will be described below.

1. Aerosol Release Rate

Figure 3 shows (a) temperature changes of sodium pool and concrete, and (b) changes of hydrogen evolution rate and aerosol release rate during a typical sodium-concrete reaction test (Run-2). Here, the hydrogen evolution rate and the aerosol release rate are expressed by the static sodium pool surface area because the free surface area during the reaction is unknown. The hydrogen evolution rate and aerosol release rate during the first 40 minutes are relatively low because an energetic sodium-concrete reaction, in which the chemical reaction between the ingredients of the concrete and sodium hydroxide is predominant,[1] does not occur yet. As soon as the energetic reaction starts, these rates increase rapidly. The peak values of these rates, however, were not obtained because of the difficulty of the sampling technique in use. As the reaction proceeds, the hydrogen evolution rate decreases gradually and reaches a nearly constant value of 2 to 3 $Nl/m^2/sec$, which is larger than the value during the first 40 minutes. On the other hand, the aerosol release rate decreases to a lower value than the initial one.

Several simulation tests by gas bubbling were conducted to interpret the above-mentioned sodium-concrete reaction phenomena. The measured aerosol release rate are shown in Fig. 4 together with the results under natural evaporation. In these results, the aerosol release rates under the gas bubbling condition increase with the increase in the flow rate of bubbling gas. The values are larger than those of the natural evaporation, but the difference becomes smaller with the increase in the sodium temperature; nearly 10 times at 400 °C and 3 times at 700 °C. This is explained by the fact that sodium evaporation, which does not contribute to mechanical aerosol release due to hydrodynamic break-up by bubble bursting, becomes a dominant source of sodium aerosol formation at high sodium temperature.

Figure 5 compares the results of Run-2 with those of the simulation tests. The numbers above the results of the sodium-concrete reaction test show the sampling order which are consistent with the numbers in Fig. 3. From the figure, the aerosol release rates obtained in the first half of the sodium-concrete reaction test agree comparatively well with those in simulation tests. The reason why the rates obtained after the initiation of the energetic reaction became lower than those in the simulation tests could be explained by the effect of reaction products in the sodium pool which could prevent the sodium evaporation and the hydrodynamic break-up by changing the physical properties of the liquid sodium, e.g., viscosity and/or surface tension.

2. Retention Factor (R.F.)

Figure 6 shows the relation between hydrogen evolution rate and the R.F.s of SrO, Eu_2O_3, and Ru during another sodium-concrete reaction test (Run-4). Here, for obtaining R.F.s, the mass concentrations of simulated n-VFPs in the sodium pool were calculated based on the total quantity of particles added in the pool. From these results, the R.F.s of simulated n-VFPs seem to be independent of the hydrogen evolution rate and nearly constant during the period measured.

Figure 7 shows the R.F.s from simulation tests together with the results of the sodium-concrete reaction test. The results of a NARA-II experiment conducted under natural evaporation condition by Sauter et al.,[7] are also presented in the figure. From the figure, it is seen that the results of the simulation tests scattered within the range of two orders in magnitude. This is due to both the settling effect of the particles in the sodium pool and the formation of condensed layer of them near the pool surface which were observed during and after the tests. The effects of particle sizes, flow rates of bubbling gas, and chemical species upon the R.F., therefore, were not clearly observed from these results. However, sodium temperature effects on the R.F., which increases with the increase in the sodium temperature in the range of 0.5 at 400 °C to 10^4 at 700 °C, is observed from the figure. The R.F.s obtained from the sodium-concrete reaction test agree well with those obtained from the simulation tests. On the other hand, the results of NARA-II indicate the upper bound of the results of this test series. The measured R.F.s of n-VFPs in sodium-concrete reaction are smaller than those in natural evaporation.

The mass concentrations of n-VFPs in sodium aerosols are shown in Fig. 8. This figure shows that the concentration decreases with the increase in the sodium temperature. As explained in the former section, this is caused by the fact that sodium evaporation, which does not contribute to the mechanical release of sodium aerosol containing n-VFPs, is a dominant source of sodium aerosol formation at high sodium temperature. This reflects the obtained R.F.s increase with the increase in the sodium temperature as shown in Fig. 7. The nearly constant R.F. value measured in the sodium-concrete reaction test is attributed to the fact that all data were measured in the condition of almost constant temperature of sodium pool.

III. CONCLUSIONS

A sodium-concrete reaction test and a simulation test using a gas bubbling nozzle were conducted in order to study mechanical release of non-volatile fission products from sodium pool during a sodium-concrete reaction. In these tests, non-radioactive materials, namely strontium oxide (SrO), europium oxide (Eu_2O_3), and ruthenium (Ru) particles whose sizes ranged from some microns to several tens of microns, were used as simulated non-volatile fission products (n-VFPs), then the retention factor (R.F. : ratio of mass concentration of n-VFPs in sodium pool to that in sodium aerosol) of these materials were measured with the aerosol release rate. The range of bubbling gas flow rate and sodium pool temperature studied were from 1 $Nl/m^2/sec$ to 10 $Nl/m^2/sec$ and from 400 ℃ to 700℃, respectively. The following conclusions were obtained from the present study:

(1) The increase in aerosol release rate due to the hydrodynamic break-up by bubble bursting at liquid sodium surface was confirmed. Although the sodium aerosol release rate during the sodium-concrete reaction was larger than that of the natural evaporation, the difference became smaller with the increase in the sodium temperature; about 10 times at 400 ℃ and about 3 times at 700 ℃.

(2) The effects of particle sizes, bubbling gas flow rates, and chemical species upon R.F. were not clearly observed. Obtained R.F.s increased from 0.5 to 10^4 with the increase in the sodium temperature from 400 ℃ to 700 ℃.

REFERENCES

1. MARTEN G. BARKER and PAUL G. GADD, "A Chemical Study of the Sodium-Concrete Reaction," in *Proc. the LMFBR Safety Topical Mtg., III*, Lyon (1982), p 91.

2. J. DUFRESNE, et al., "Sodium Concrete Interaction," in *Proc. the LMFBR Safety Topical Mtg., III*, Lyon (1982), p 109.

3. L. D. MUHLESTEIN and A. K. POSTMA, "Application of Sodium-Concrete Reaction Data on Breeder-Reactor Safety Analysis," *Nuclear Safety*, *25*, No.2, 212 (1984).

4. DONG H. NGUYEN and L. D. MUHLESTEIN, "SOCON: A Computer Model for Analyzing the Behavior of Sodium-Concrete Reactions," in *Proc. the Int. Topical Mtg. on FAST REACTOR SAFETY*, 2, Knoxville (1985), p 747.

5. H.-W.FRITZKE, et al., "Experimental Investigation of Hydrogen Release as a Consequence of Sodium-Concrete Interaction," in *Proc. Int. Conf. on Sci. and Technol. of FAST REACTOR SAFETY*, *1*, Guernsey (1986), p 87.

6. M. BOLVIN, et al., "Research on Concrete with Acceptable Safety Characteristics under Sodium Attack," in *Proc. Int. Conf. on Sci. and Technol. of FAST REACTOR SAFETY*, *1*, Guernsey (1986), p 97.

7. H. SAUTER and W. SCHUETZ, "Fuel and Fission Product Release from a Hot Sodium Pool," KfK3800/CSNI95, 90 (1984).

Table 1 Test Conditions and Test Parameters

	Sodium – Concrete Reaction Test			Sodium Bubbling Test	
Mass of Sodium Pool (kg)	8.9			8.9	
Sodium Pool Temperature (℃)	350~700 (600 before discharge)			400 500 600 700	
Depth of Sodium Pool (cm)	32~35			32~35	
Temperature of Vessel Ceiling (℃)	300 (constant)			300 (constant)	
Bubbling Gas	Hydrogen (H_2)			Nitrogen (N_2)	
Gas Evolution Rate (N ℓ/m²/sec)	1~20* (own course)			1 5 10	
n-VFPs	SrO	Eu_2O_3	Ru	SrO	Eu_2O_3
Mass Mean Diameter of Particles (μm)	30.7	3.4	7.3	30.7 12.6 9.7	3.4 1.4
Quantity added to Sodium Pool (g)	25			25	
Mass Concentration of Simulated FPs in Sodium Pool (mg/g-Na)**	2.80			2.80	

* assumed peak value
** based on total quantity of simulated FPs added in pool

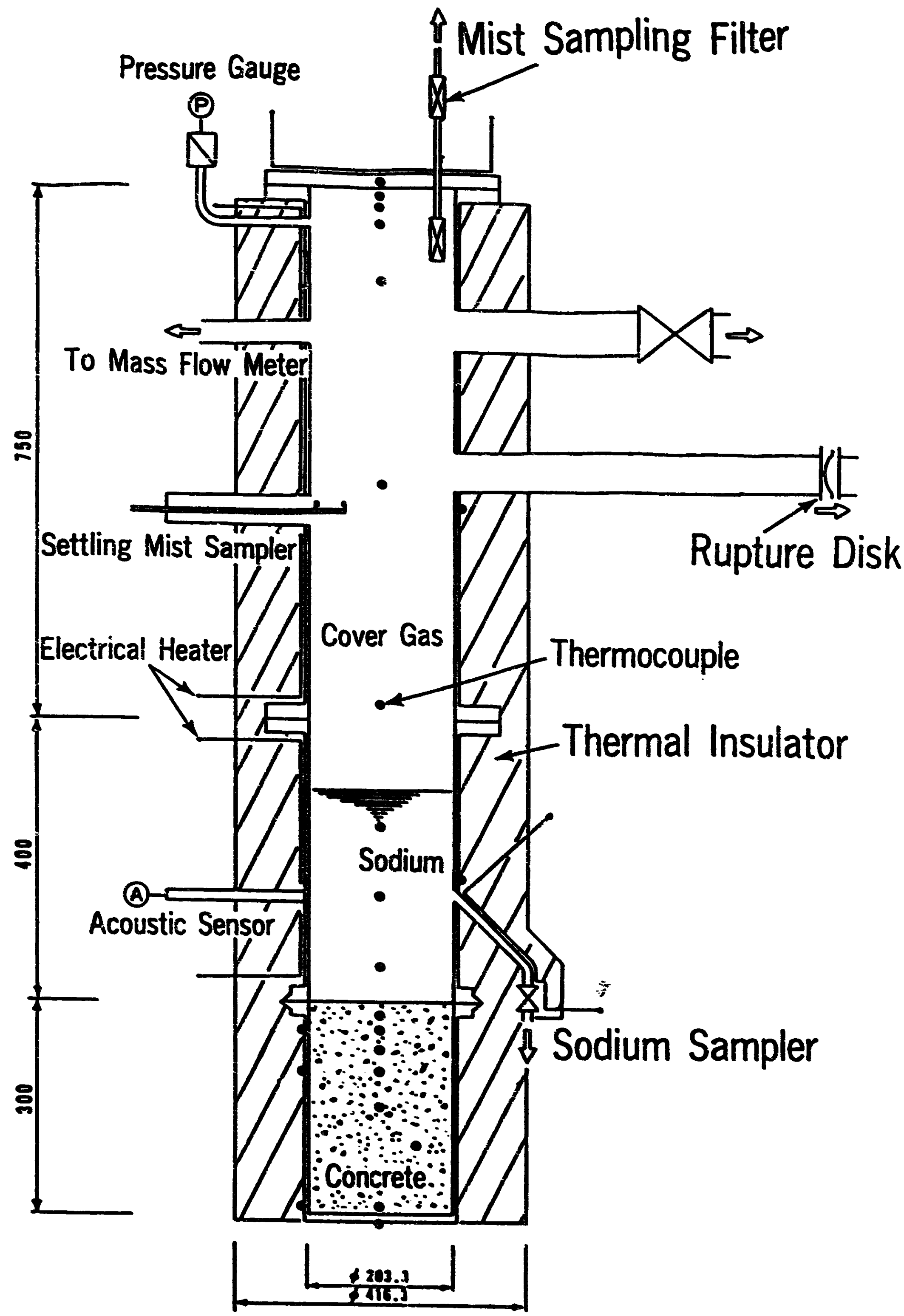


Fig.1 Arrangement of Test Rig for Sodium Concrete Reaction

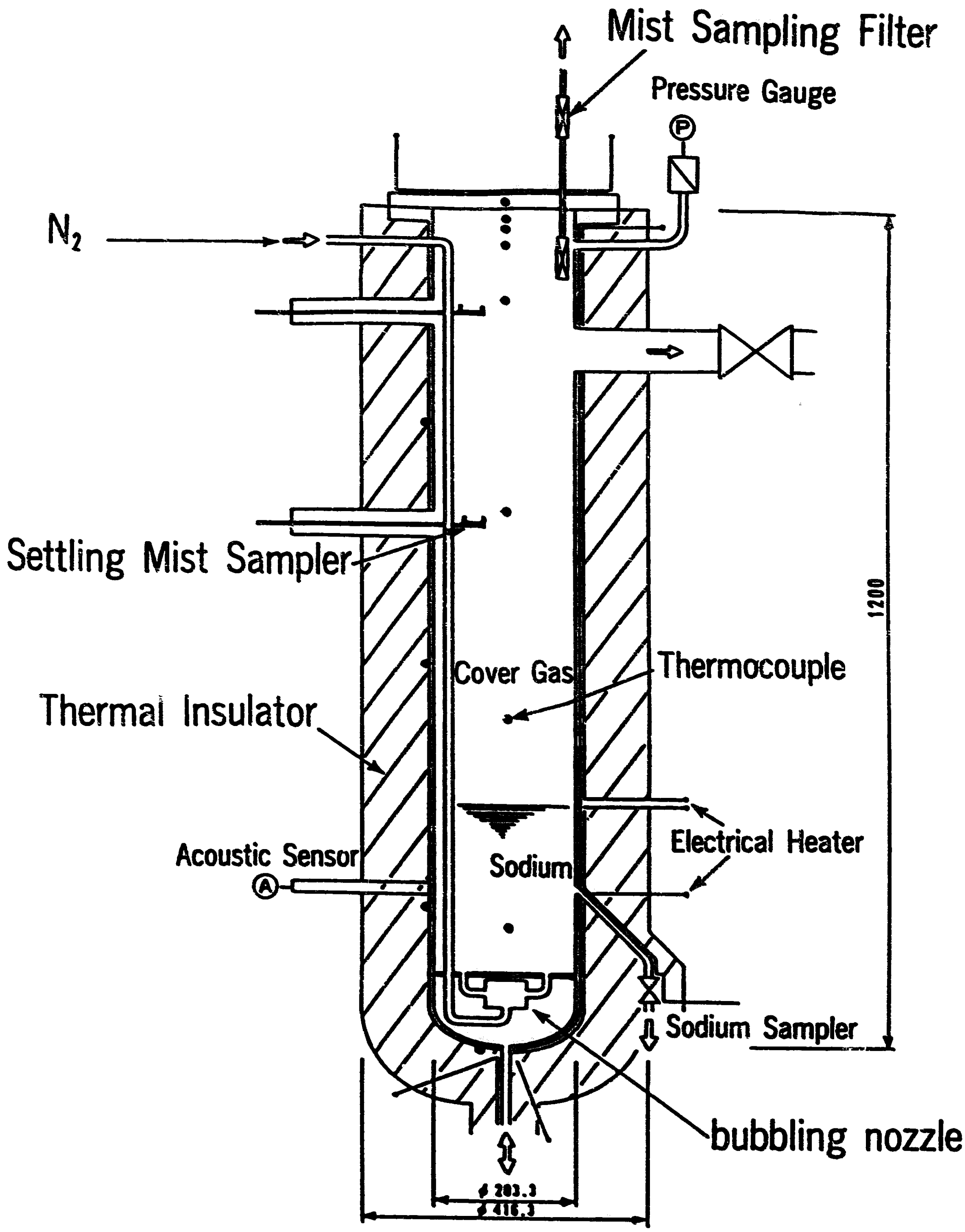
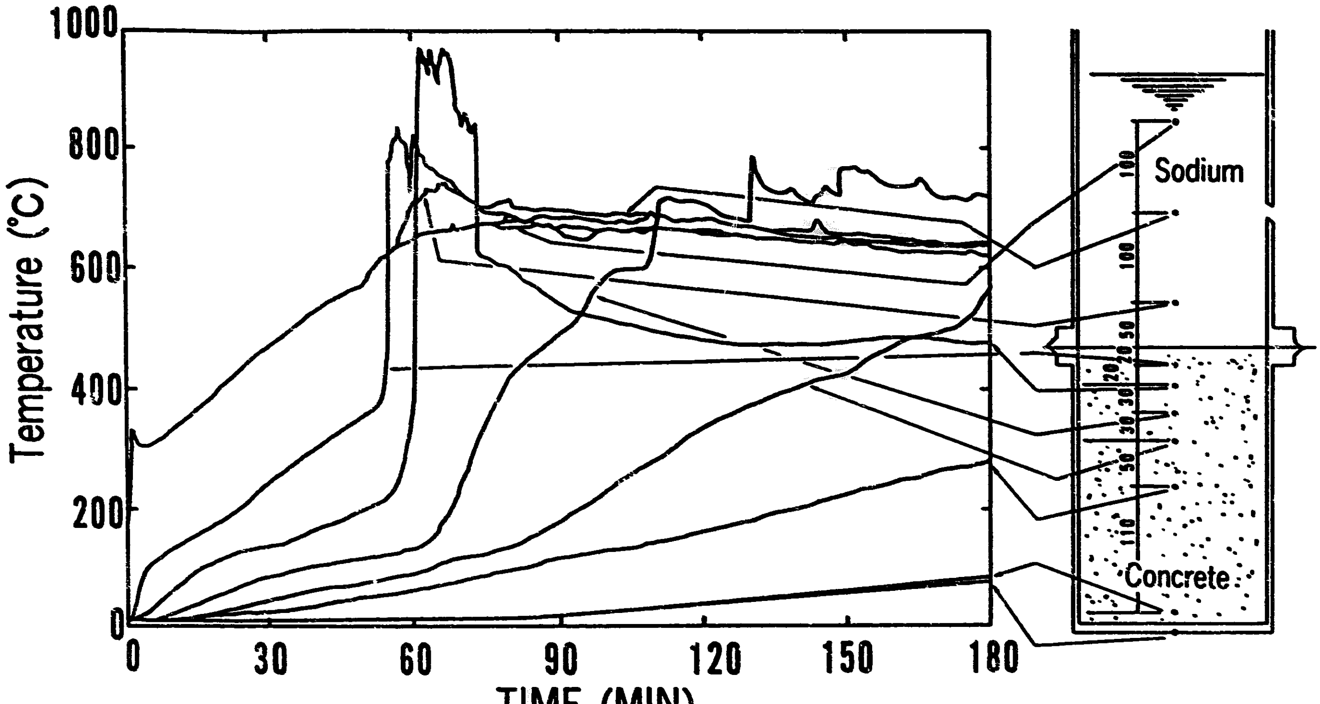
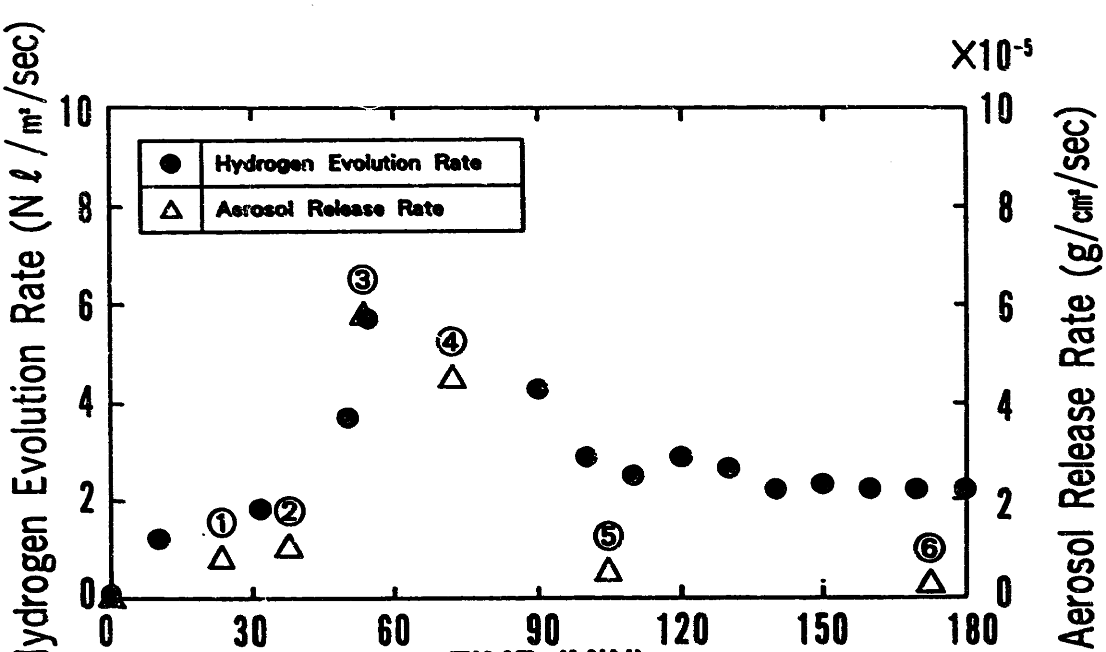


Fig.2 Arrangement of Test Rig for Sodium Bubbling



(a) Temperature Changes of Sodium Pool and Concrete



(b) Changes of Hydrogen Evolution Rate and Aerosol Release Rate

Fig.3 Test Results of Temperatures of Sodium Pool and Concrete, Hydrogen Evolution Rate, and Aerosol Release Rate during Sodium — Concrete Reaction ; Run — 2

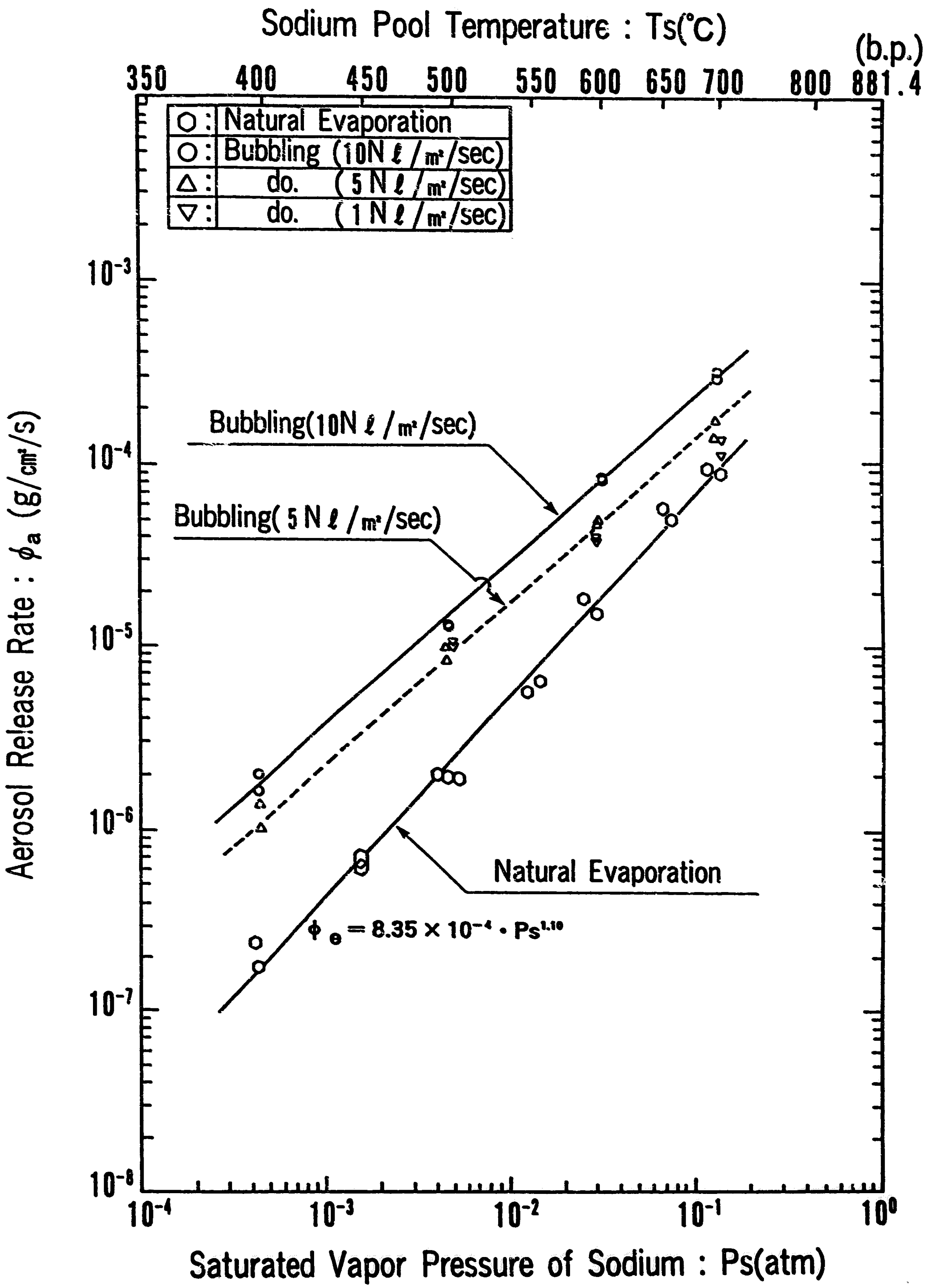


Fig.4 Aerosol Release Rate in Sodium Bubbling Test

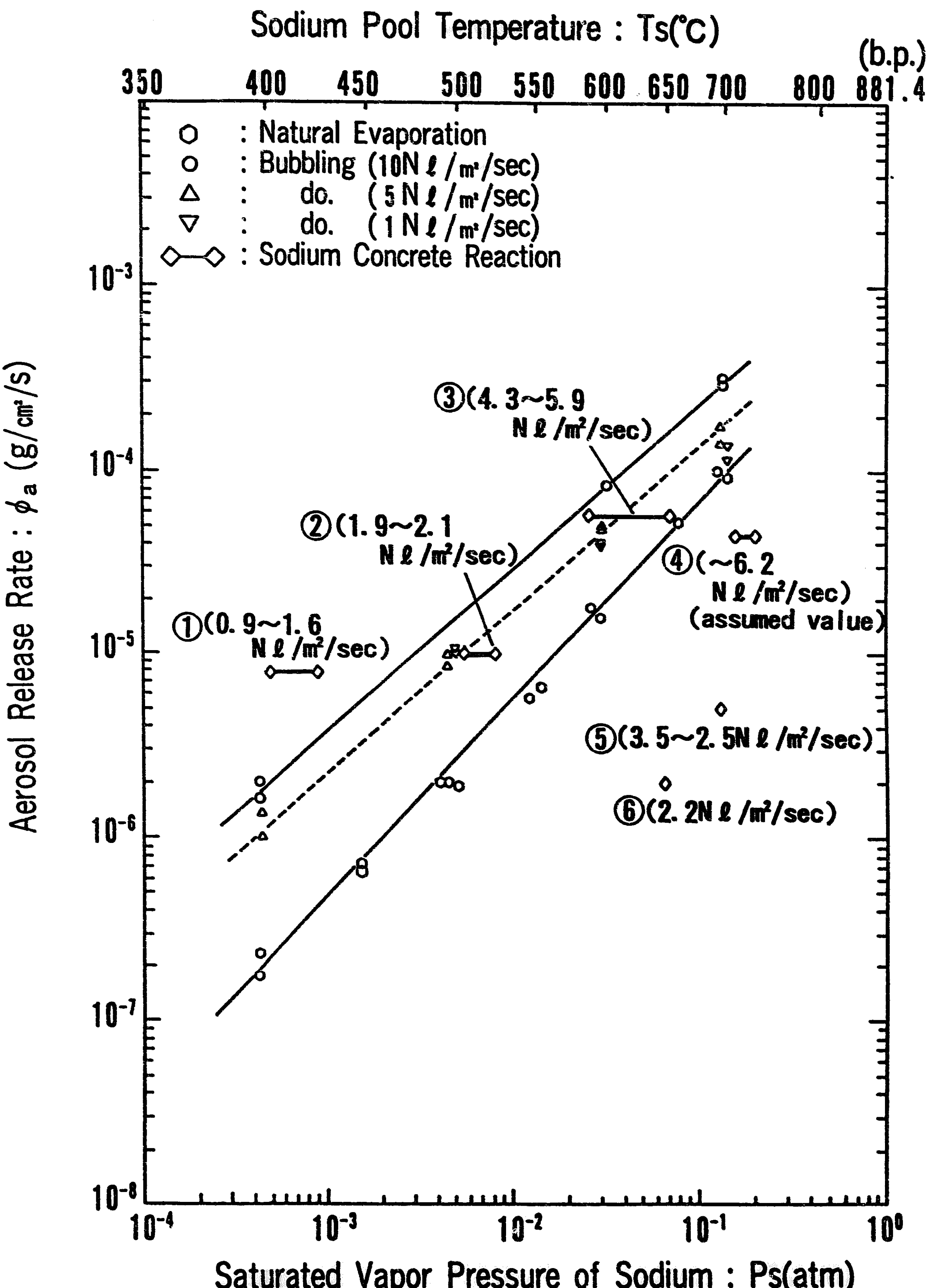


Fig.5 Aerosol Release Rate in Sodium Bubbling Test and Sodium Concrete Reaction Test ; Run − 2

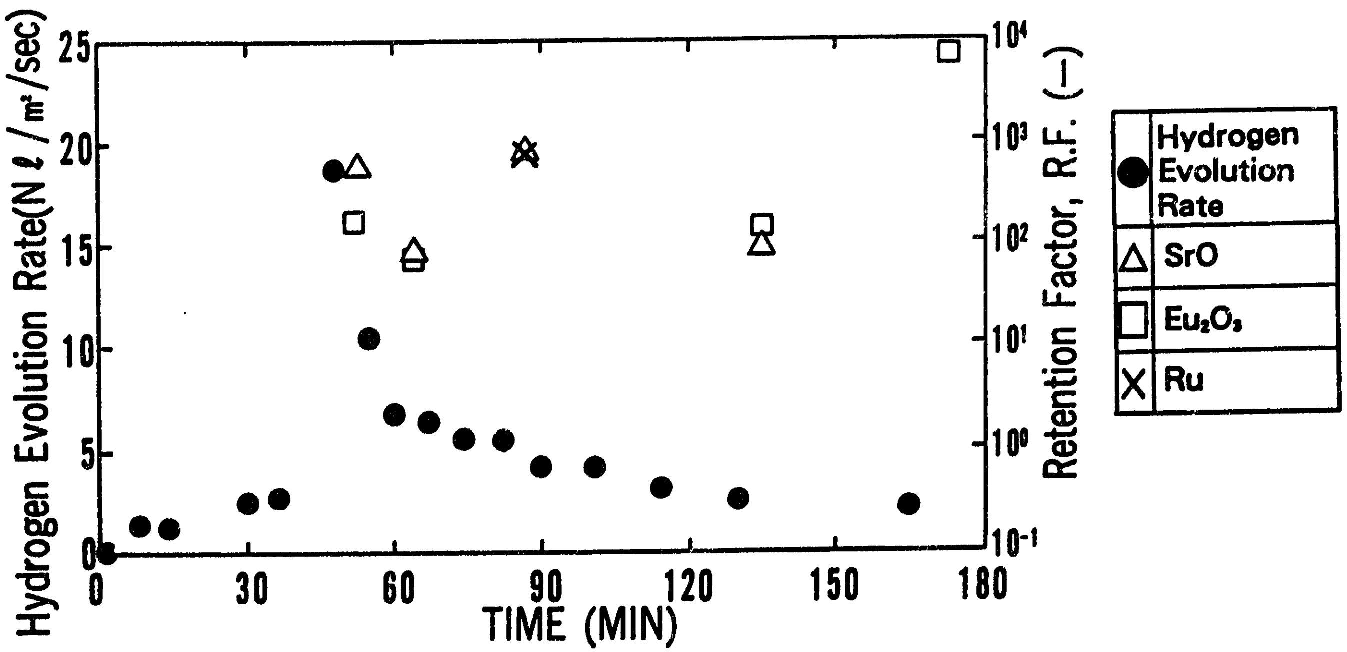


Fig. 6 Test Results of Hydrogen Evolution Rate and Retention Factor during Sodium — Concrete Reaction ; Run — 4

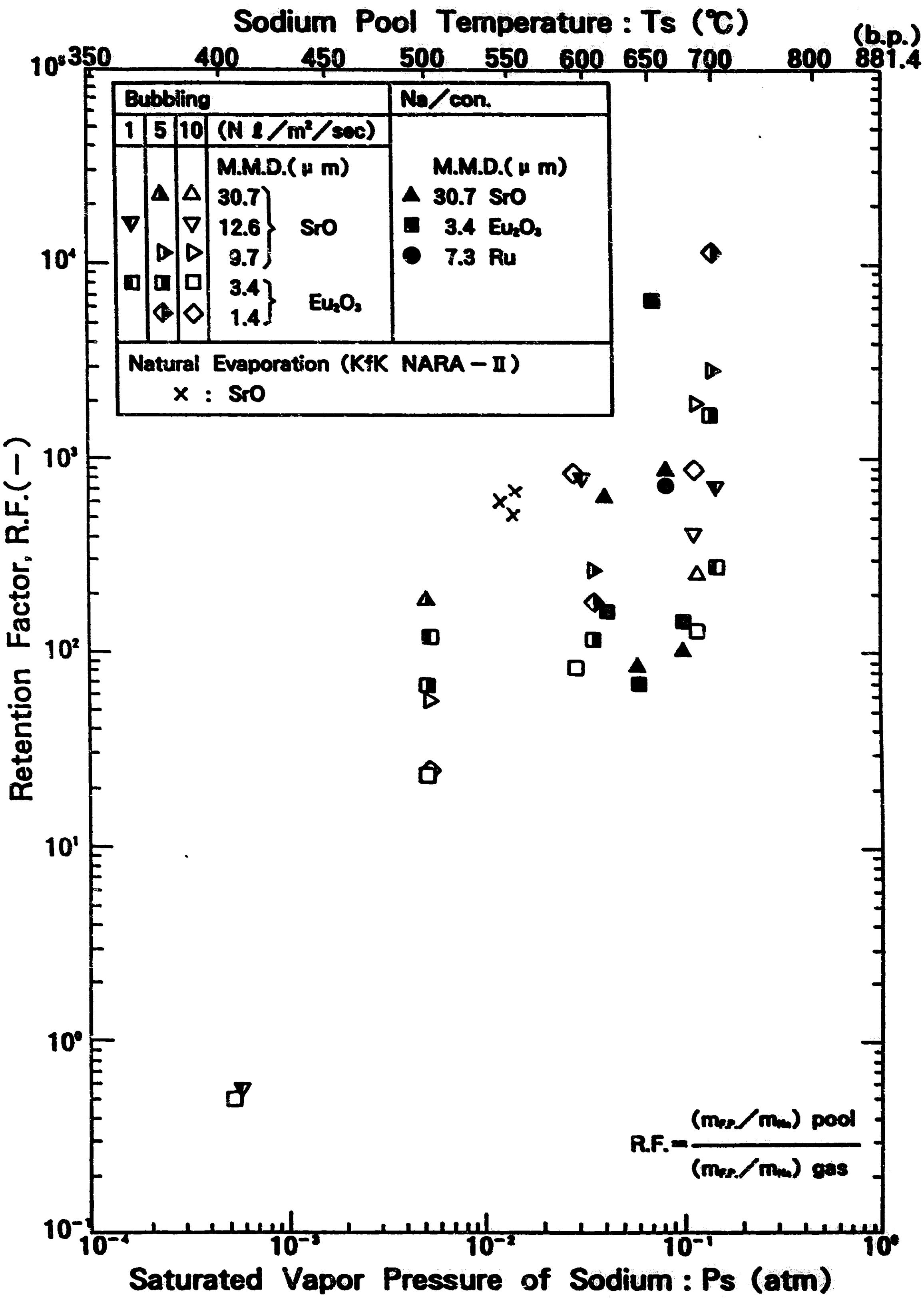


Fig.7 Obtained Retention Factors of Simulated non − volatile Fission Products

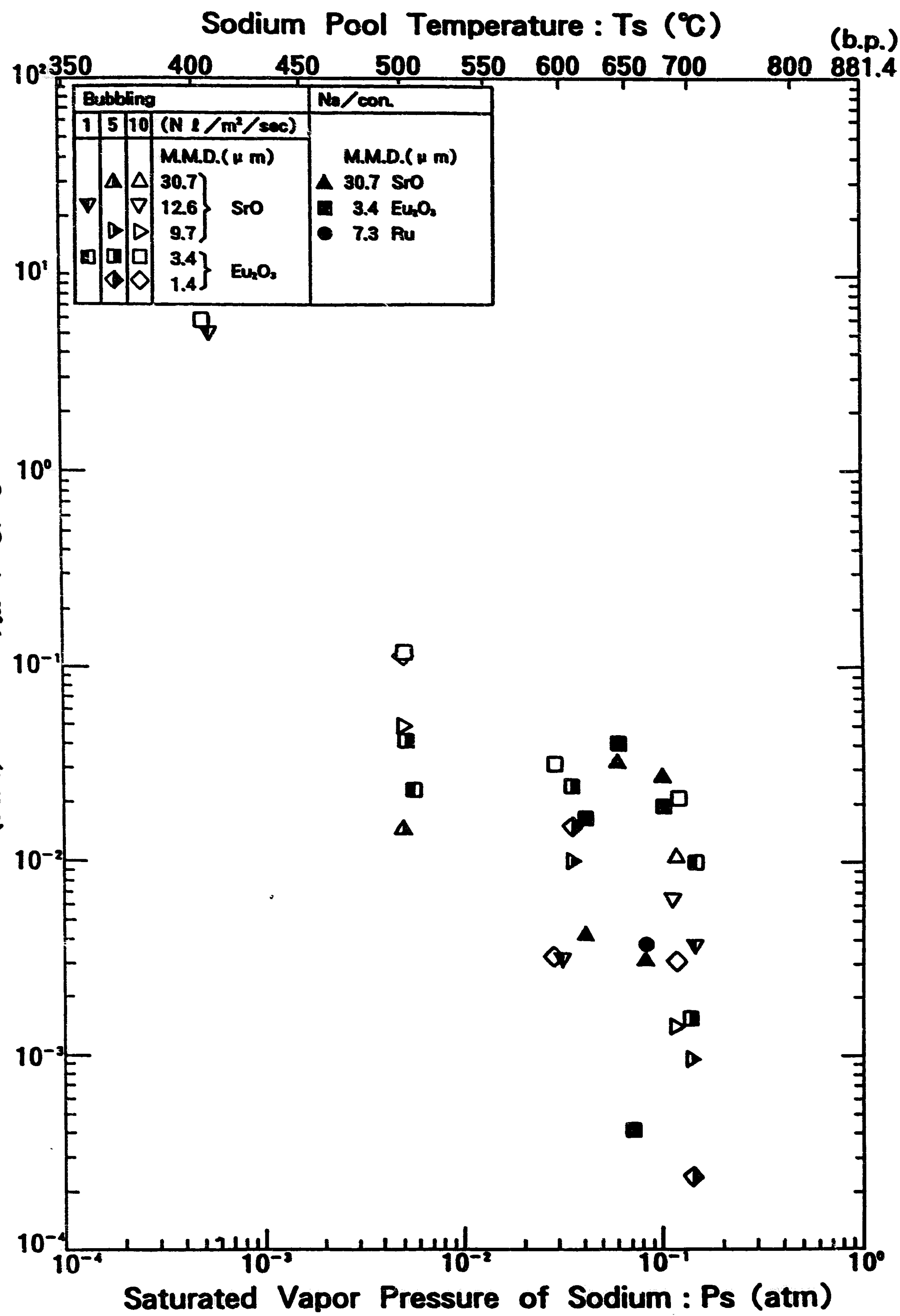


Fig.8 Concentrations of simulated non-volatile Fission Products in Sodium Aerosol