JAERI-M
93-097

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March 1993

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Japan Atomic Energy Research Institute

編集兼発行　日本原子力研究所
印　　　刷　いばらき印刷㈱

Analysis of the Plate-out Distribution in the VAMPYR-I Experiments by PLAIN Code

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(Received March 10, 1993)

Fission products plate-out analysis code, PLAIN, was developed to calculate the plate-out distribution in the primary cooling system of High Temperature Gas-cooled Reactor (HTGR).

As one of the verification works of the PLAIN code, calculations of the plate-out densities in VAMPYR-I experiments were carried out. The VAMPYR-I was a test equipment installed in the experimental HTGR in Germany, AVR, to investigate fission products plate-out behavior. The calculated plate-out densities were smaller than measured ones in the upper region of the test-tube of VAMPYR-I and it is considered that the difference is caused by the dust effect.

Keywords: Plate-out, Fission Product, HTGR, PLAIN, VAMPYR-I, Dust

PLAINコードによるVAMPYR－I実験の沈着分布解析

日本原子力研究所大洗研究所高温工学試験研究炉開発部
沢　和弘・塩沢　周策・馬場　治

(1993年3月10日受理)

高温ガス炉の1次冷却系における核分裂生成物の沈着挙動を解析するために，PLAINコードが開発された。

PLAINコードの検証作業の一環として，VAMPYR－I実験における沈着密度計算を行った。VAMPYR－Iは，ドイツの高温ガス実験炉AVRに沈着挙動研究のために設置された実験装置である。沈着密度の計算値は，VAMPYR－Iのテストチューブの上流で測定値を下回り，その違いはダストの効果によるものと考えられることが分かった。

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1. Introduction

In high temperature gas-cooled reactors (HTGRs), some amounts of fission products are released mainly from fuel with failed coatings and are transported in primary cooling system with primary coolant during normal operation. In that case, condensable fission products plate-out on the inner surface of components in the primary cooling system and they affect the shielding design and safety evaluation(1). Therefore, the plate-out behavior of fission product has been investigated and a fission product plate-out analysis code, PLAIN(2), has been developed. The PLAIN code has been used to calculate the plate-out distribution in the primary cooling system of the High Temperature Engineering Test Reactor (HTTR) for its shielding design and safety evaluation(1),(3). Verification works of the PLAIN code were mainly carried out by comparison with experimental data of fission products plate-out distributions obtained in the Oarai Gas Loop No.1 (OGL-1), which is an in-pile loop simulating the conditions of the primary cooling system of HTGR.

Calculation works were also carried out by using experimental data obtained in the VAMPYR-I to investigate the in-situ plate-out behavior. The VAMPYR-I was installed in AVR, which was an experimental HTGR with pebble bed-type fuel element, in Germany. The calculation works of the VAMPYR-I were carried out based on the "JAERI's Participation in the AVR Investigation Programme, HTA-11".

This paper describes calculated results of plate-out distributions in the VAMPYR-I by the PLAIN code together with discussions.

2. VAMPYR-I Experiments(4)

The aims of VAMPYR-I were

(1) measurement of deposition and diffusion profiles at different materials for the determination of plate-out parameters

and

(2) determination of the concentrations of the non gaseous fission products in the hot coolant gas of the AVR reactor.

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There were three tubes for exchangeable thermocouples extending into the hot coolant flow. A hole was bored on the front side of one tube facing the coolant flow. Via this tube, a part of the coolant gas was extracted and led through a removable sampling tube which was smoothly inserted into the thermocouple tube. Then, the gas was led to following absolute filters. The gas flow rate was 15 Nm^3/h, i.e. 0.7 g/s. The flow characteristic was laminar. Figure 1 shows a cross-section of the reactor showing the installed place of the VAMPYR-I.

A schematic presentation of the facility is drawn in Fig. 2. The sampling tube, i.e. test-tube was about 2.3 m in length and its inner diameter was about 20 mm. The sampling tube and the absolute filter-1 were one unit, so called the test section, which could be removed into a connectable transport container via two ball valves. The test section was removed by pneumatic treatment using the pressure drop between the primary circuit and the transport container. Removal and exchange of the test section were carried out during a short shut down period.

The experiment was usually operated for a period of about 4 weeks and the operational conditions such as temperature of the coolant gas, gas flow through the facility, etc. were kept constant during the test period.

The test tube could be made from different steel materials. However, there was a restriction for the insertion of high temperature metals because of high neutron flux in the front part of about 1.5 m in length of the test tube. This caused high activation of the most materials and it was very difficult to determine deposited fission products in that region, especially for short lived isotopes. Thus for measurement on specific activity in the coolant or on dust, the insertion was restricted to materials having low activation cross-section, titanium for example.

After the test run, the test section and control filter-2 were exchanged and examined. This examination includes determination of total activity and activity profiles on test tube and filters. Activity measurements were done by gamma spectrometry of the total fission product spectrum.

Calculations works for the verification of PLAIN code were carried out for the experimental runs of V09 and V12, which were taken in 1973/74, respectively, since there were sufficient and accurate post experimental data for calculations. Main test conditions and total shut-down activities of experimental runs of V09 and V12 are listed in Table 1. Calculations were carried out for Cs-137 and I-131, which are important from the view point of HTGR design. As shown in Table 1, test tube materials were

Cr-Mo steel and Ti in V09 and V12 test, respectively. Cr-Mo steel is used as the material of reactor pressure vessel of HTTR. On the other hand, since Ti is not used in the HTTR, material data of Ti were added to the internal material data of PLAIN code to calculate plate-out distribution of V12 test. Temperature distributions along the test tube used in the calculations are shown in Fig. 3.

3. Result and Discussion

The analytical model of PLAIN code is shown in Appendix-1.

Calculated results of I-131 and Cs-137 are shown in Fig. 4(a) and Fig. 4(b), respectively, and the following results are obtained.

(1) Calculated plate-out distribution of I-131 in V09 test shows good agreement with measured one.

(2) Calculated plate-out activities of I-131 in V12 test show smaller values than measured ones in the upper stream region of the test-tube.

(3) Calculated plate-out distributions of Cs-137 both in V09 and V12 tests show smaller values than measured ones in the upper stream region of the test-tube.

The difference between calculated and measured plate-out distributions is large in the upper stream region of the test-tube, where temperature is high and thus fisson products easily diffuse into the base metal. On the other hand, in the down stream region of the test-tube, where temperature is low, adsorption of fission products on the surface of the test-tube is dominant. In down stream region, the calculated and measured plate-out distributions show good agreement. From these results, it seems that the amount of fission products, which diffuse into the base metal, are underestimated.

In order to examine the sensitivity of diffusion constant in the base metal, parameter survey was carried out. In the survey calculation, diffusion constants in the base metals were changed parametrically. The calculated results of Cs-137 are shown in Fig. 5. Since the half-life of I-131 is relatively short, it disintegrates before diffusing into the base metal and the diffusion constant does not drastically affect the plate-out distribution of I-131. From the parameter survey, the following results

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(1) The diffusion constant in the Cr-Mo steel must be about 100 times larger than the base value to fit calculated plate-out distribution to measured one in the upper stream region of test-tube.

(2) The diffusion constant in the Ti must be more than 100 times larger than base value to adjust calculated plate-out distribution to measaured one in the upper stream region of test-tube.

These results are not acceptable because the diffusion constants in the base metal are physically determined as the combinations of nuclides and the materials, and the differences of the diffusion constants are too large to fit calculated plate-out profiles to measured ones. Therefore, the differences between calculated and measured plate-out distributions in the upper stream region of the test-tube can not be concluded to be caused from the differences of the diffusion constants in base metal.

On the other hand, PLAIN code can calculate the plate-out distributions based on given/fixed penetration coefficients. The penetration coefficients used in the calculation were taken from the literatures(5),(6). The calculated plate-out distributions of I-131 and Cs-137 with constant penetration coefficients are shown in Fig. 6(a) and Fig. 6(b), respectively, and the following results are obtained.

(1) Calculated plate-out distributions of I-131 in V09 and V12 tests become larger than measured values in the upper stream region of the test-tube.

(2) Calculated plate-out distributions of Cs-137 in V09 and V12 tests show good agreement with measured ones.

Thus, the following considerations are extracted from the calculations.

(1) In the upper stream region of the test-tube of VAMPYR-I, there is an effect which causes hundred or more times larger plate-out densities than calculated ones by PLAIN code. This effect is more drastic for Cs-137 than for I-131. The penetration coefficients described in the literatures(5),(6) might be determined including this effect.

(2) If this effect is supposed to be caused by the dust which is contained in the helium coolant of AVR, the behavior of the measured plate-out distributions would be qualitatively explained as follows. If there are some amounts of graphite or carbon dusts in the primary coolant, they might fall down and/or adhere on the surface of the

test-tube. Since cesium has larger sorptivity to the graphite than that of iodine, the dusts contain some amounts of cesium, and the measured plate-out activities in the upper stream region would include the activity of cesium on the dusts. On the other hand, the calculated values by PLAIN code are not considered the dust effect. Though the similar effect could also occur for iodine, because of its low sorptivity to the graphite compared with cesium ($\sim\times10^{-6}$)(7), observed plate-out distributions of I-131 would not be affected by the dust effect. As shown in Appendix-2, the PLAIN code can calculate the plate-out distribution in the OGL-1, because there is no graphite abrasion mechanism in the OGL-1, and the similar situation could be expected in the HTTR.

(3) In order to confirm the cause of differences between measured and calculated plate-out distributions in the VAMPYR-I, the following works should be carried out: (a) calculation of plate-out distributions in VAMPYR-II experiment, which is expected to contain smaller amount of dust than the VAMPYR-I, and (b) calculation of plate-out distributions in the VAMPYR-I by the code, in which dust plate-out model is introduced. In this case, measured plate-out activities should be divided into plate-out activities on dust and on/in the metal wall.

4. Conclusions

The plate-out distributions of I-131 and Cs-137 in the VAMPYR-I experiments in AVR were calculated by the PLAIN code, and the following conclusions were obtained.

(1) In the upper stream region of test-tube of VAMPYR-I, there is an effect which causes hundred or more larger times plate-out densities than calculated ones by PLAIN code. This effect is considerably larger for Cs-137 than for I-131.

(2) Since there are some amounts of graphite dust on which cesium absorbs in helium coolant of AVR, the difference between measured and calculated plate-out distributions is supposed to be caused by the dust effect.

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Acknowledgment

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Table 1 Main Experimental Data of V09 and V12.

	V09	V12
Pressure (ata)	11	11
Concentration at inlet (n/cm^3)	I–131: 2.60X10^9 Cs–137: 9.63X10^{10}	I–131: 2.71X10^9 Cs–137: 1.68x10^{11}
Temperature (°C)	Fig. 3	Fig. 3
Flow rate (g/s)	0.66	0.66
Experimental duration (h)	799	822
Test tube material	Cr–Mo	Ti

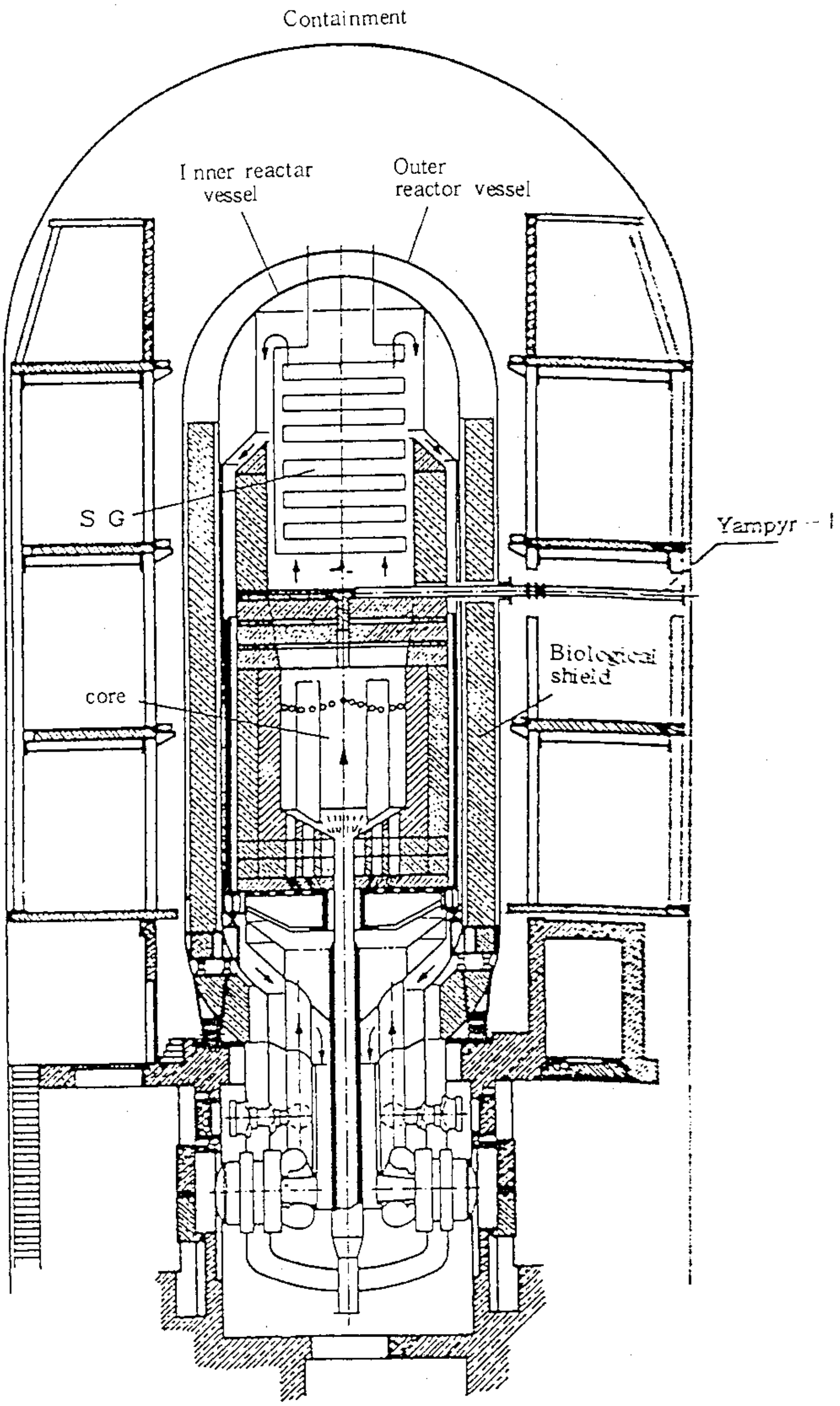


Fig. 1 Schematic View of AVR and VAMPYR-I.

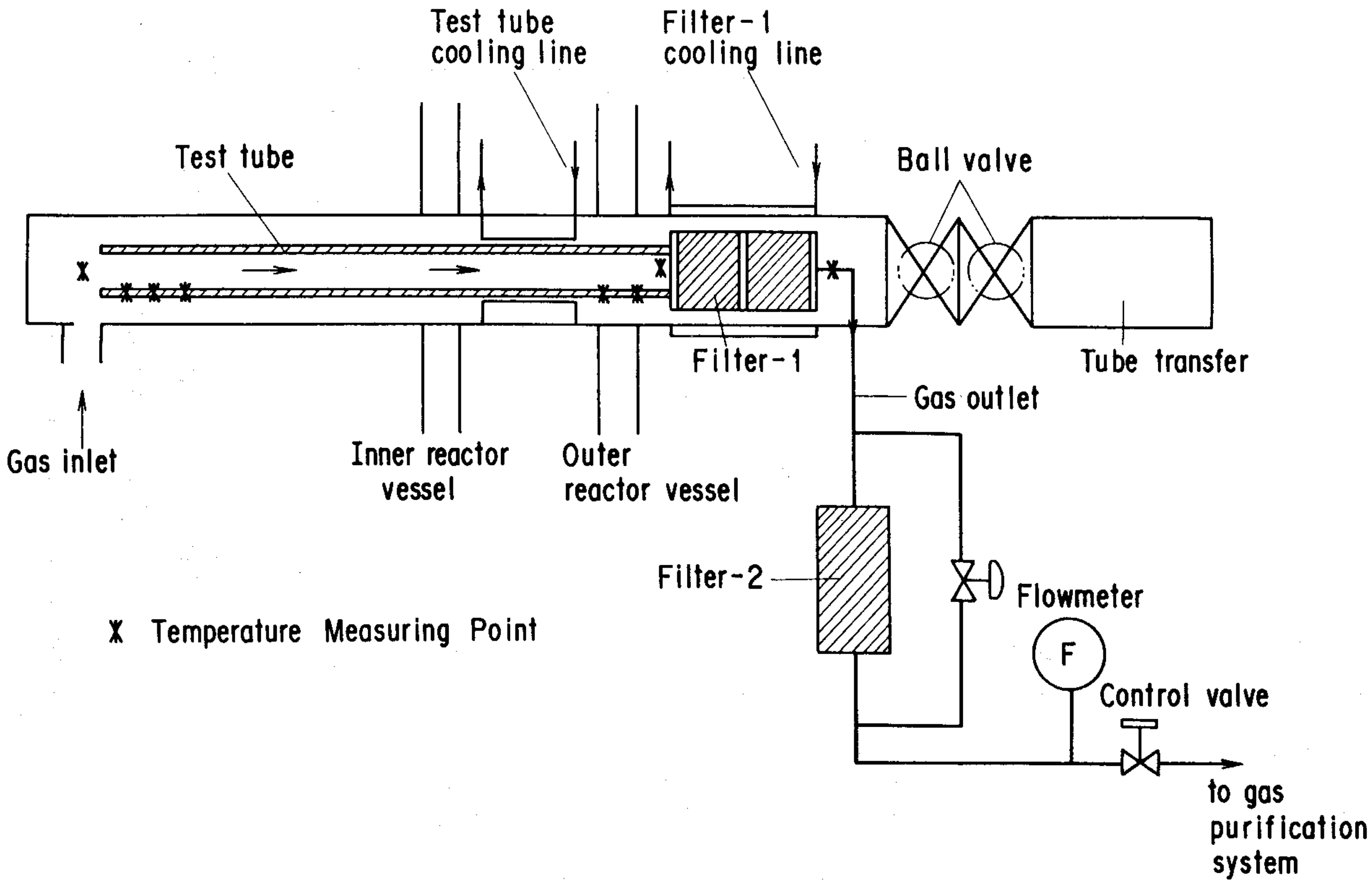


Fig. 2 Flow Diagram of VAMPYR-I.

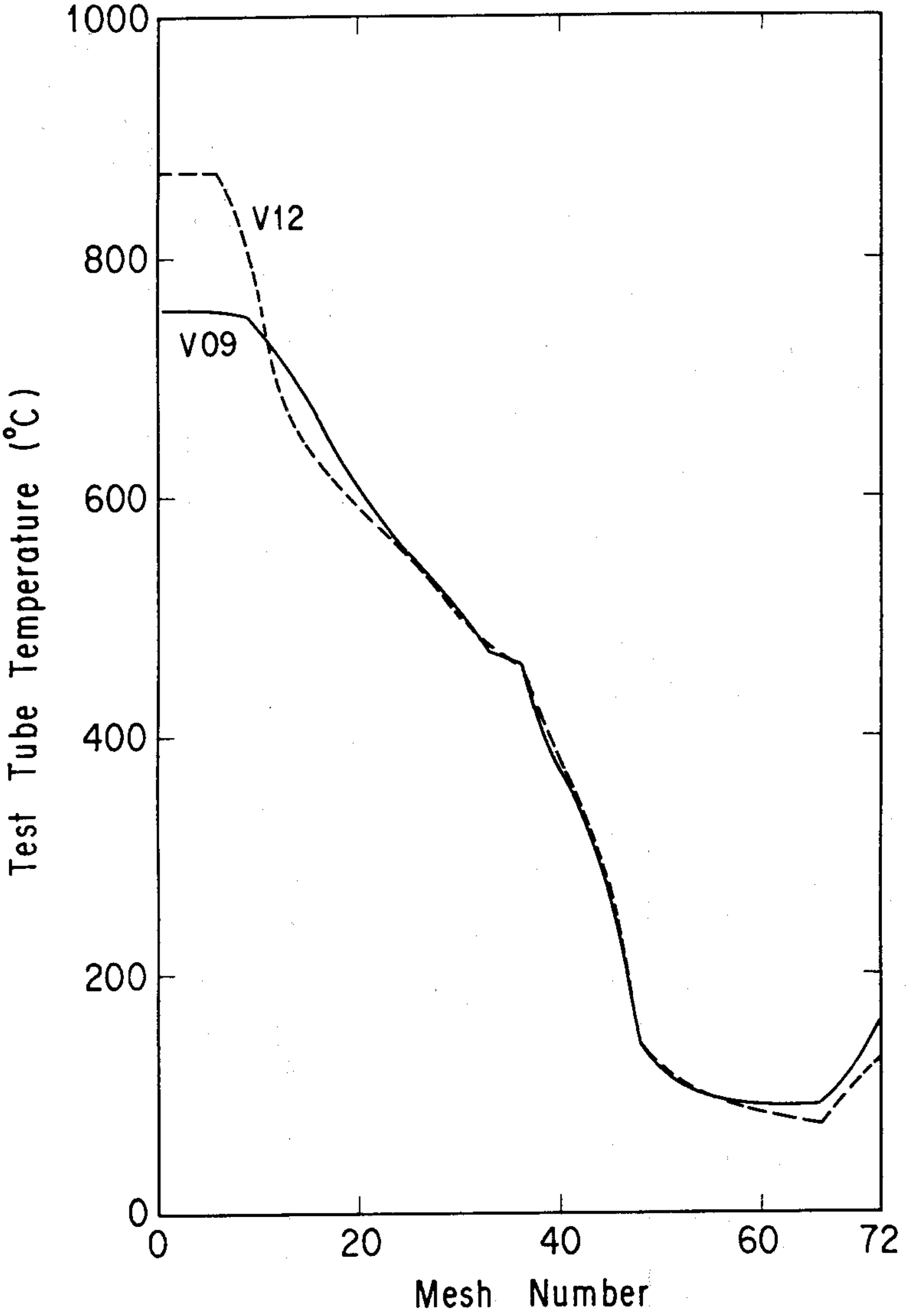
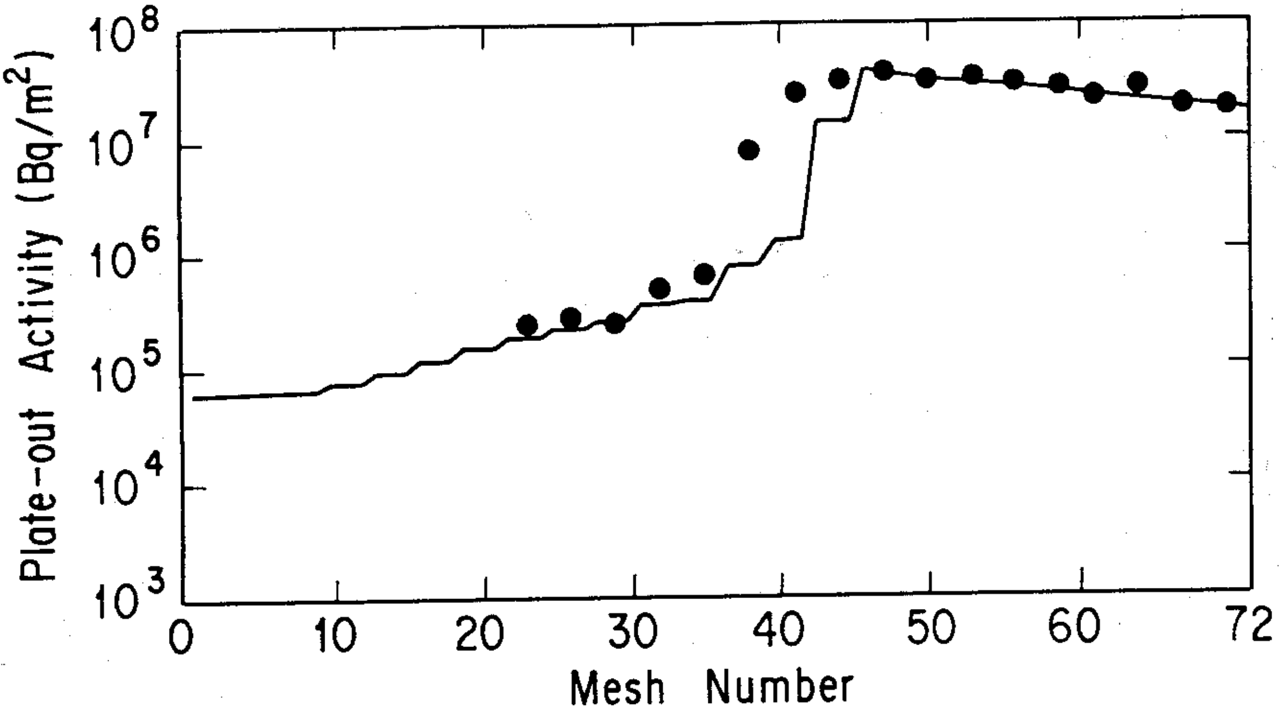
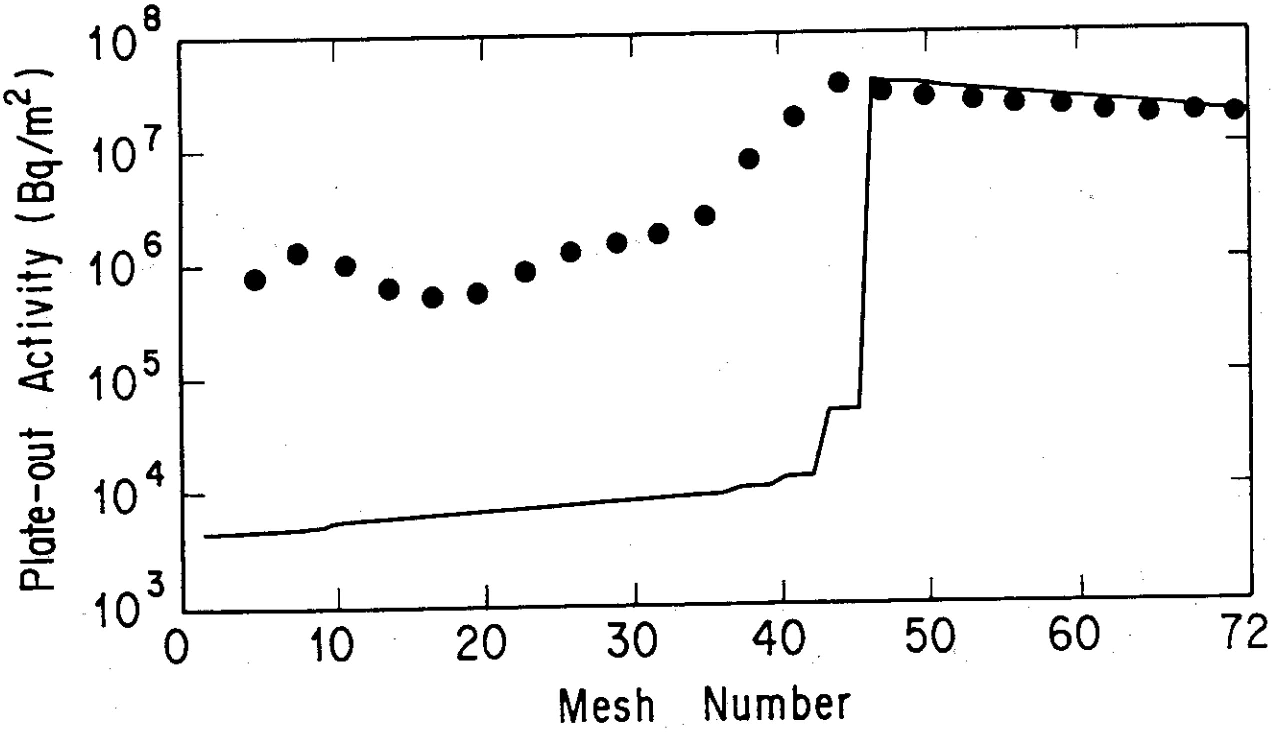


Fig. 3 Temperature Distributions of V09 and V12.

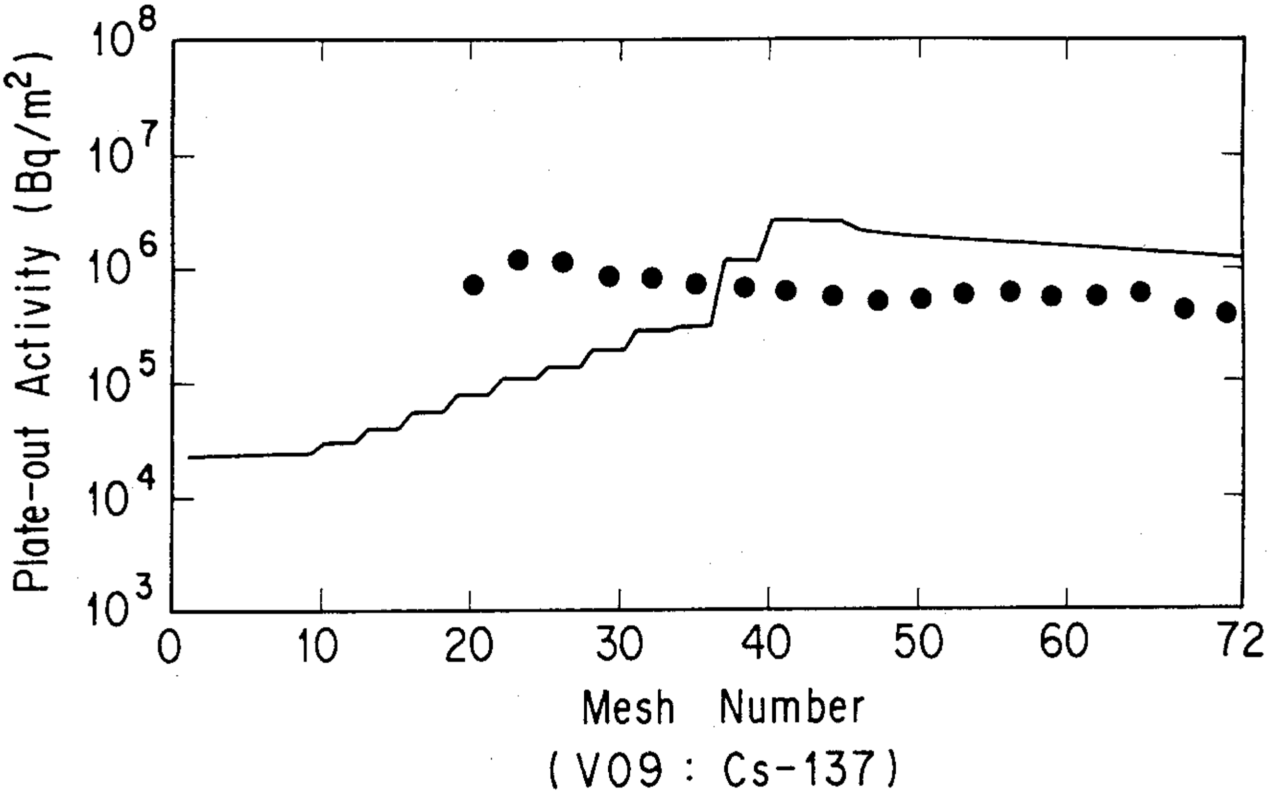


(V09 : I-131)



(V12 : I-131)

Fig. 4(a) Results of I-131 Plate-out Distributions in VAMPYR-I.



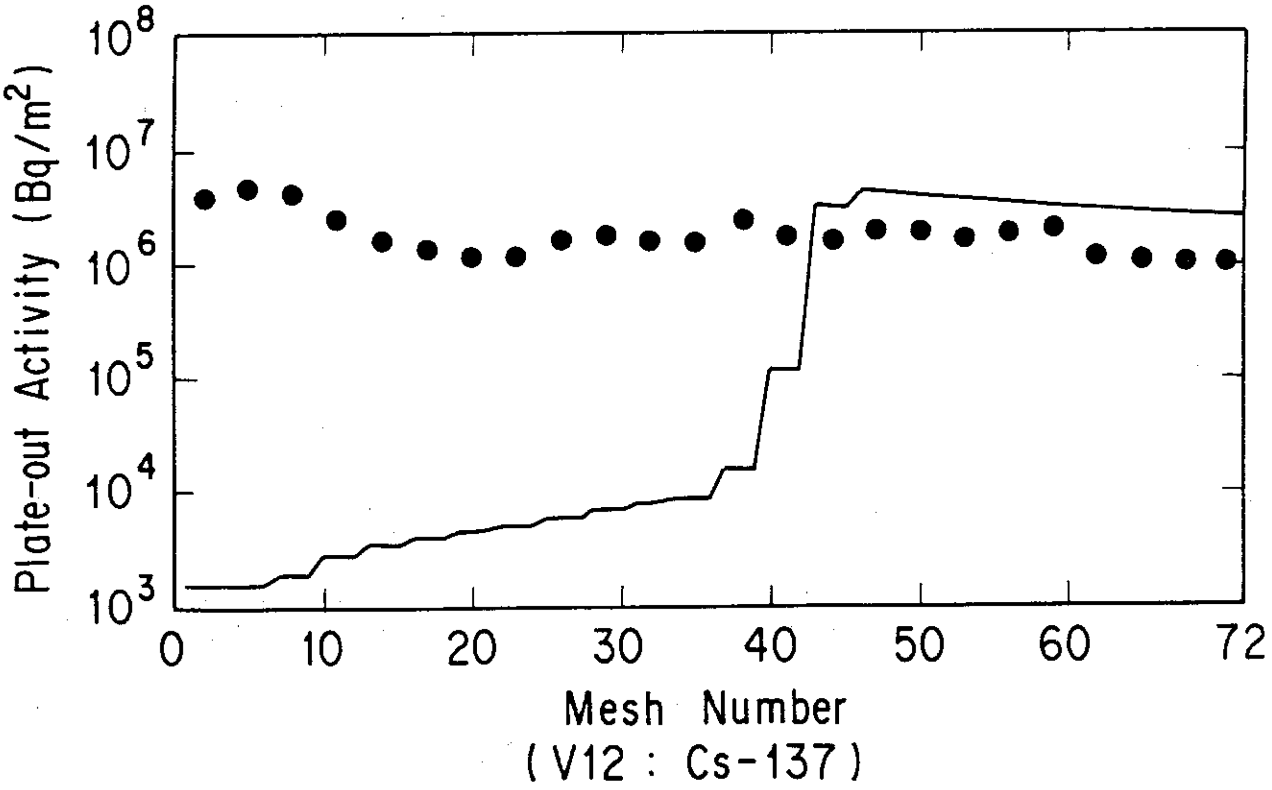


Fig. 4(b) Results of Cs-137 Plate-out Distributions in VAMPYR-I.

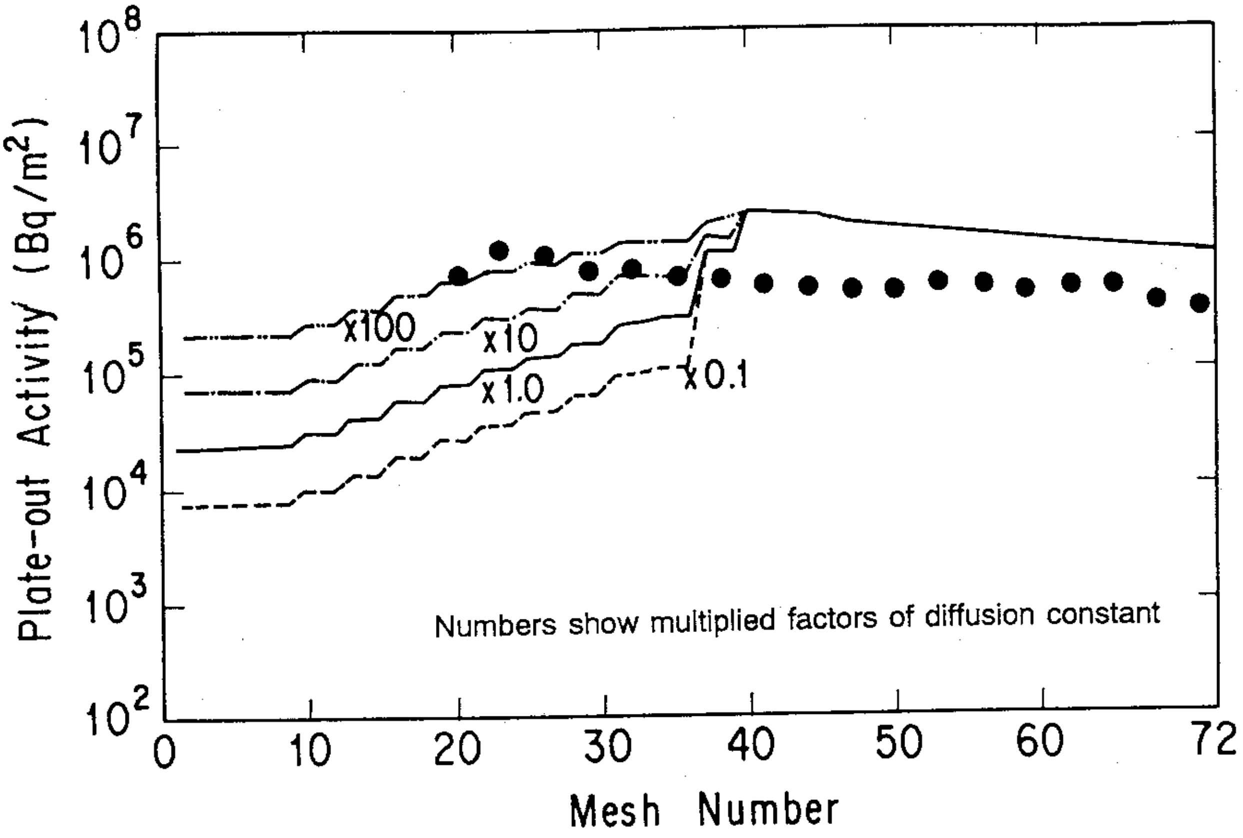


Fig. 5(a) Results of Parameter Survey of Cs-137 (V09).

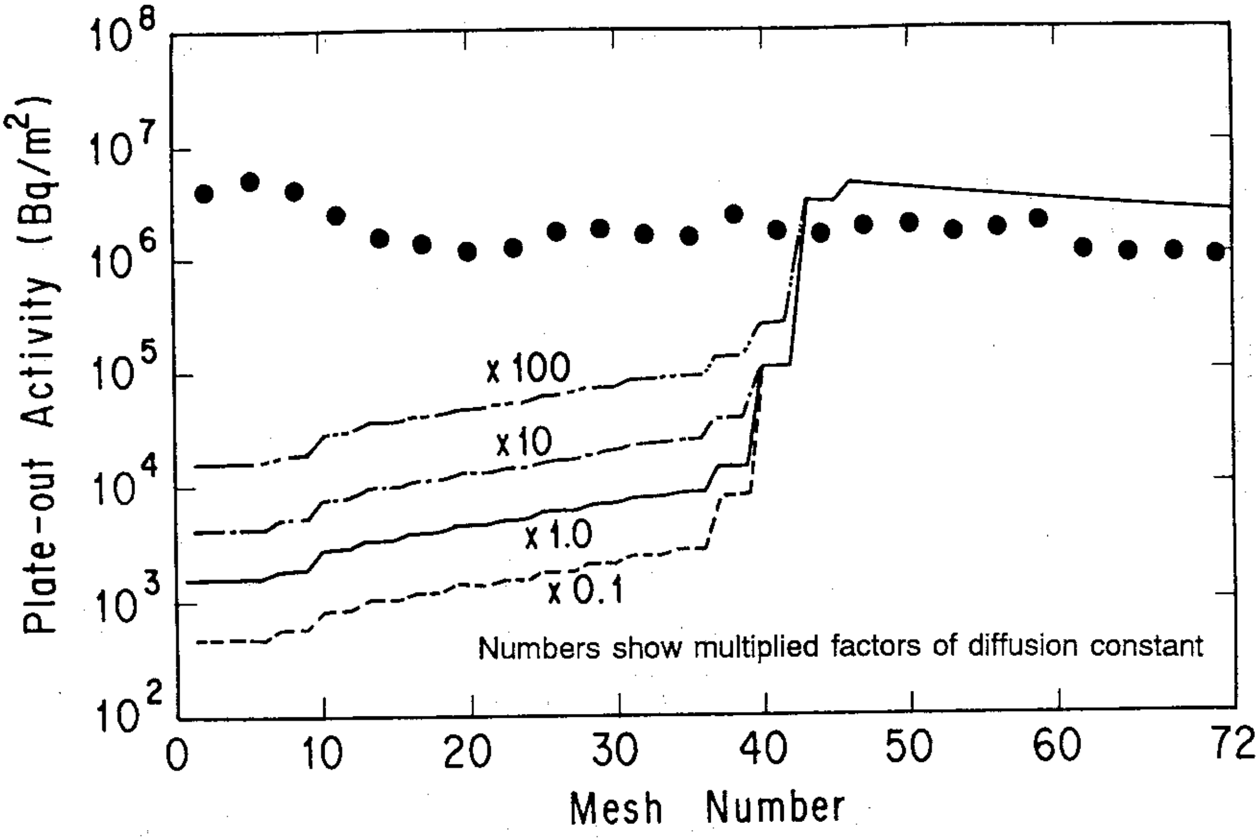
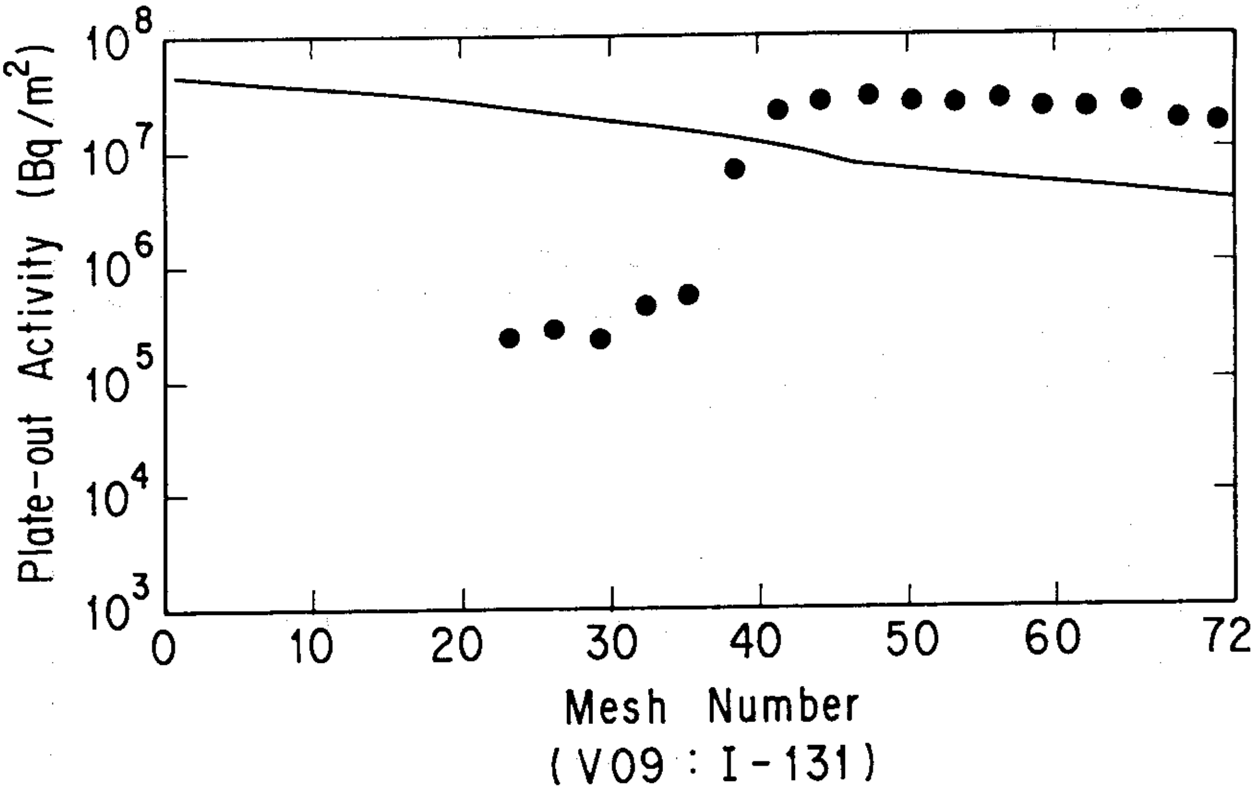


Fig. 5(b) Results of Parameter Survey of Cs-137 (V12).



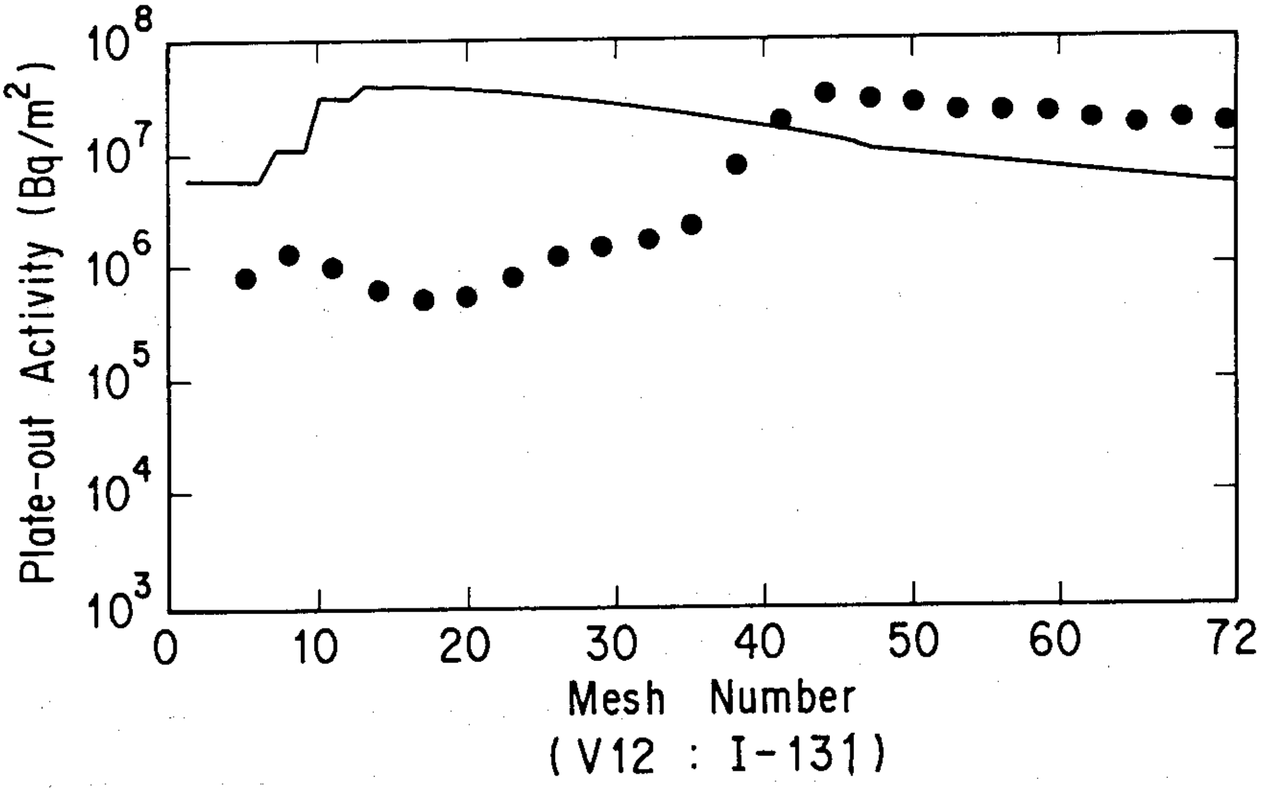


Fig. 6(a) Results of I-131 Plate-out Distributions by Iniotakis model.

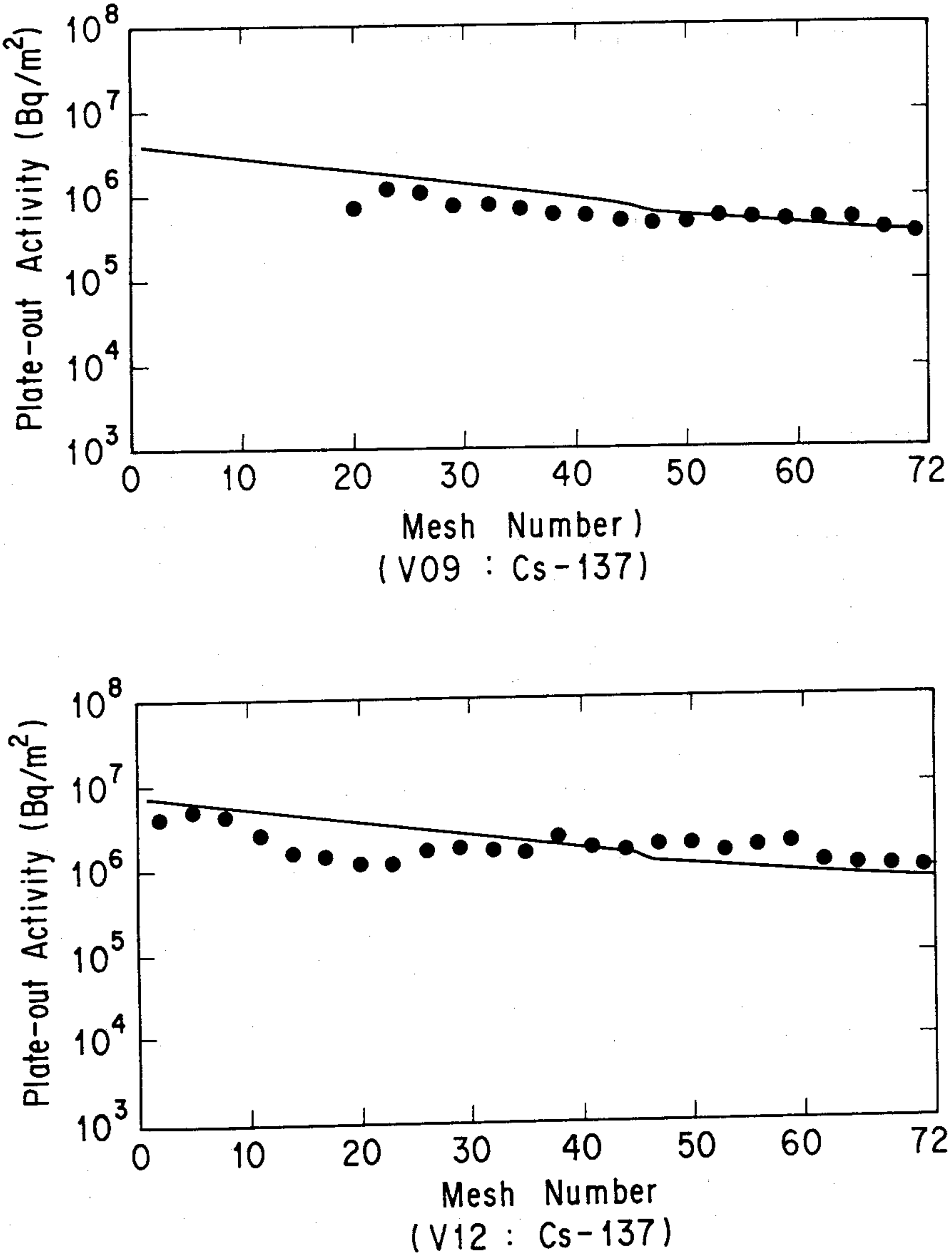


Fig. 6(b) Results of Cs-137 Plate-out Distributions by Iniotakis model.

Appendix 1 Analytical Model of PLAIN Code(A1-1),(A1-2)

A computer code, PLAIN, has been developed to calculate fission products plate-out distributions in the primary cooling system of HTGR(A1-1). PLAIN is based on Iniotakis model(A1-3) but has a modified model in the fission product penetration process into the base metal reflecting the experimental results of the OGL-1 which is installed in the Japan Materials Testing Reactor (JMTR).

1. Analytical Model

The following two mechanisms are assumed in the PLAIN code.

(1) Adsorption and desorption
Fission products which collide on the inner surfaces of components in primary cooling system adsorb on them with certain probability and adsorbed fission products desorb by the thermal excitation of the metal surface of the components.

(2) Diffusion into and from base metal
The adsorbed fission products diffuse into the base metal according to the lattice vibration of base metal and again diffuse out when the surface concentration becomes low.

The behavior of fission products is modeled into following three regions as shown in Fig. A1-1.

(1) Main flow region (Main stream)
Fission products flow together with coolant and their concentrations are assumed to be uniform, e.g. the diffusion flow is neglected relative to the macroscopic transport flow.

(2) Laminar sub-layer (Material transport region)
Fission product concentration shows a linear decrease in the radial direction, e.g. the macroscopic transport flow is neglected relative to the diffusion flow.

(3) Wall flow (Layer of the wall)
This region has a thickness of the mean free path of fission products, in which the thermal motion of the fission products is the decisive factor.

The basic equations employed in the PLAIN code are as follows.

$$\frac{\partial N_i}{\partial t} + \frac{4h_i}{d}(N_i - N_{w,i}) + \lambda_i N_i + V\frac{\partial N_i}{\partial x} - \lambda_{i-1}N_{i-1} = 0 \qquad \text{(A1-1)}$$

$$- h_i(N_i - N_{w,i}) - \theta_i M_{o,i} + \alpha^* N_{w,i} = 0 \qquad \text{(A1-2)}$$

$$\frac{\partial M_o}{\partial t} + (\theta_i + \lambda_i + \gamma_i)M_{o,i} - \lambda_{i-1}M_{o,i-1} - \alpha^* N_{w,i} - \eta_i \phi_{i,R_i} = 0 \qquad \text{(A1-3)}$$

$$\frac{\partial \phi_i}{\partial t} - D_i\left(\frac{\partial^2 \phi_i}{\partial \rho^2} + \frac{1}{\rho}\frac{\partial \phi_i}{\partial \rho}\right) + \lambda_i \phi_i - \lambda_{i-1}\phi_{i-1} = 0 \qquad \text{(A1-4)}$$

$$\frac{\partial M_{D,i}}{\partial t} + \lambda_i M_{D,i} - \lambda_{i-1}M_{D,i-1} = - D_i\left(\frac{\partial \phi_i}{\partial \rho_{R_i}} - \frac{R_o}{R_i}\frac{\partial \phi_i}{\partial \rho_{R_o}}\right) \qquad \text{(A1-5)}$$

where,

N : fission product concentration in main stream (n/m^3)

N_w : fission product concentration in layer of the wall (n/m^3)

M_o : number of fission product on the wall metal (n/m^3)

ϕ : concentration of diffused fission product in the wall metal (n/m^3)

M_D : equivalent number of diffused fission product on coolant flow surface (n/m^3)

$$M_D = \frac{\int_{R_i}^{R_o} \phi \cdot 2\pi r \cdot dr}{2\pi r_i}$$

λ : decay constant (s^{-1})

α : accommodation factor (-)

$$\alpha^* = \alpha\sqrt{\frac{RT}{2\pi m}}$$

η : sublimation rate (s^{-1})

γ : in-diffusion rate (s^{-1})

θ : desorption rate (s^{-1})

m : mass number of fission product (g/mol)

D : diffusion coefficient of fission product in wall metal (m^2/s)

h : mass transfer coefficient of fission product in coolant (m/s)

d : equivalent hydraulic diameter of flow region (m)

v : velocity of coolant flow (m/s)

x : distance in flow direction (m)

t : time (s)

R : gas constant (J/mol•K)

R_i, R_o: inner and outer diamter of flow path (m)

$i, i-1$: fission product and its precursor

T : temperature (K)

Brief descriptions of the physical constants employed in the PLAIN code are described below(A1-2).

Diffusion constants in base metal

$$D = D_o e^{-\frac{Q_D}{RT}} \tag{A1-6}$$

where,

D_o : vibration factor (m^2/s)

Q_D : activation energy (J/mol)

Diffusion constant in coolant

Arnold's(A1-4) or Hirshfelder's(A1-5) formula can be selected. The calculations were carried out by using the following Hirshfelder's formula.

$$Dg = \frac{0.00092916\ T^{2/3}}{P\cdot(\gamma_{12})^2\cdot\psi}\sqrt{\frac{1}{M_1}+\frac{1}{M_2}} \tag{A1-7}$$

where,

P : pressure

γ_{12} : mean value of coolant and fission product molecular radius (Å)

ψ : collision function of fission product

M_1, M_2: mass number of fission product and coolant, respectively (g/mol)

Mass transfer coefficient

For Re < 2300,

$$h = 0.023\,\frac{Dg}{d}\,Re^{0.8}Sc^{0.4} \tag{A1-8a}$$

For Re > 2300,

$$h = \frac{Dg}{d}\left(3.66 + \frac{0.0668(\ell/d)Re\cdot Sc}{1 + 0.04(\frac{\ell}{d}\cdot Re\cdot Sc)^{2/3}}\right) \tag{A1-8b}$$

where,

Re : Reynolds number in the main flow

Sc : Schumidt number in the main flow

ℓ : length of flow path

Desorption rate

$$\theta = \omega e^{-\frac{Q_d}{RT}} \tag{A1-9}$$

where,

Q_d : desorption energy (J/mol)

ω : Debye constant (s^{-1})

In-diffusion rate

$$\gamma = \omega e^{-\frac{Q_D}{RT}} \tag{A1-10}$$

Sublimation rate

$$\eta = \frac{D}{a} e^{-\frac{Q_d}{RT}} \tag{A1-11}$$

where,

a : lattice constant of wall metal (m)

References of Appendix 1

A1-1. O. BABA, et.al.: "Fission Products Plate-out Analysis Code in the HTGR - PLAIN-", JAERI-M 88-266, (1988) (in Japanese).

A1-2. JAERI: "Fission Product Plate-out Studies in JAERI", 2-4, Dec., (1991).

A1-3. N. INIOTAKIS, et.al.: "Initial Results of Investigation into Fission Product Deposition In-pile Experiments", Nucl. Eng. Des., 34, (1975).

A1-4. J.H. ARNOLD: Ind. Eng. Chem., 22, 1091, (1930).

A1-5. J.O. HIRSHFELDER: Chem. Reviews, 44, 205, (1949).

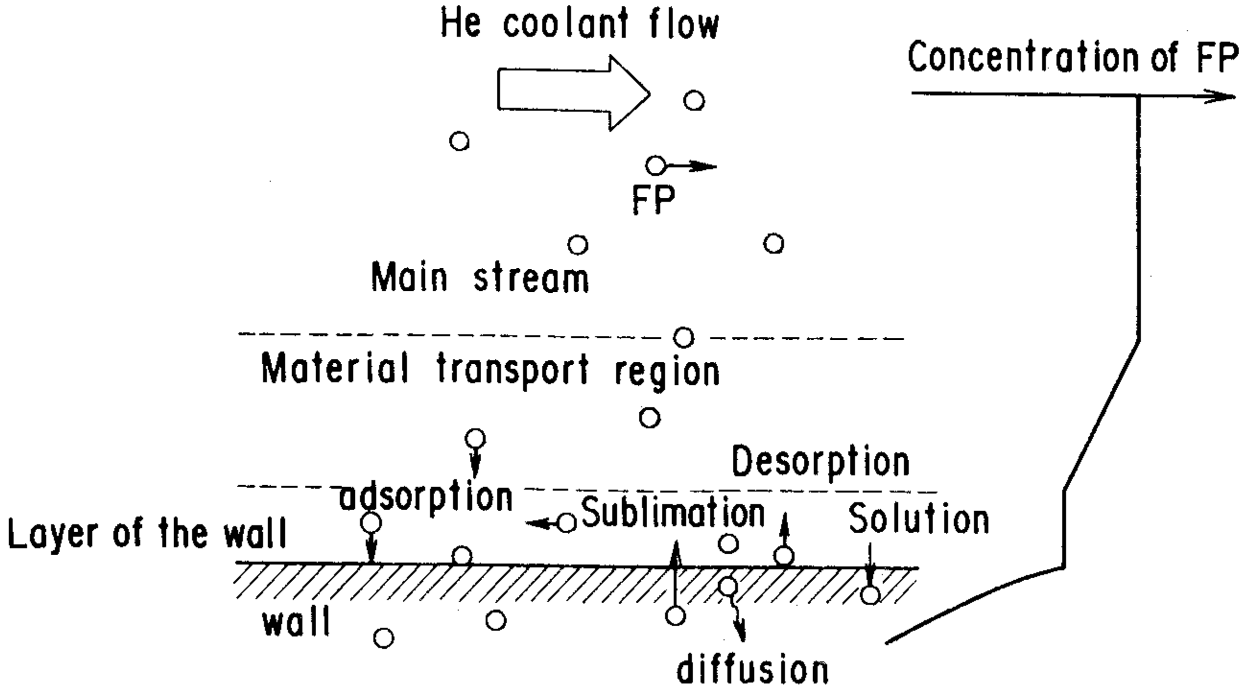


Fig. A1-1 Plate-out Model of PLAIN Code.

Appendix 2 Verification Results of PLAIN Code by OGL-1 Experiments(A2-1)

The verification works have been carried out by comparison with experimental data of fission products plate-out distributions obtained in the OGL-1 experiments, which simulate the primary cooling system condition of HTGR. The plate-out distribution in OGL-1 has been measured from the outside of the primary pipe after every operational cycle using Ge-detector, and the range of helium gas temperature is from 1000°C at fuel exit to room temperature at down stream region. Flow condition is turbulent. The flow diagram of OGL-1 is shown in Fig. A2-1.

Examples of verification results of I-131 and Cs-137 are shown in Fig. A2-2. In the figure, lines and dots show calculated and measured plate-out densities, respectively. From the results, it is concluded that the calculated profiles show good consistency with the measured values as a whole, in spite of the complicated temperature distribution and flow diagram as shown in Fig. A2-1.

Reference of Appendix 2

A2-1. K.SAWA, et.al.: "The Verification of Fission Products Plate-out Analysis Code for HTGR -PLAIN-", JAERI-M 91-084, (1991) (in Japanese).

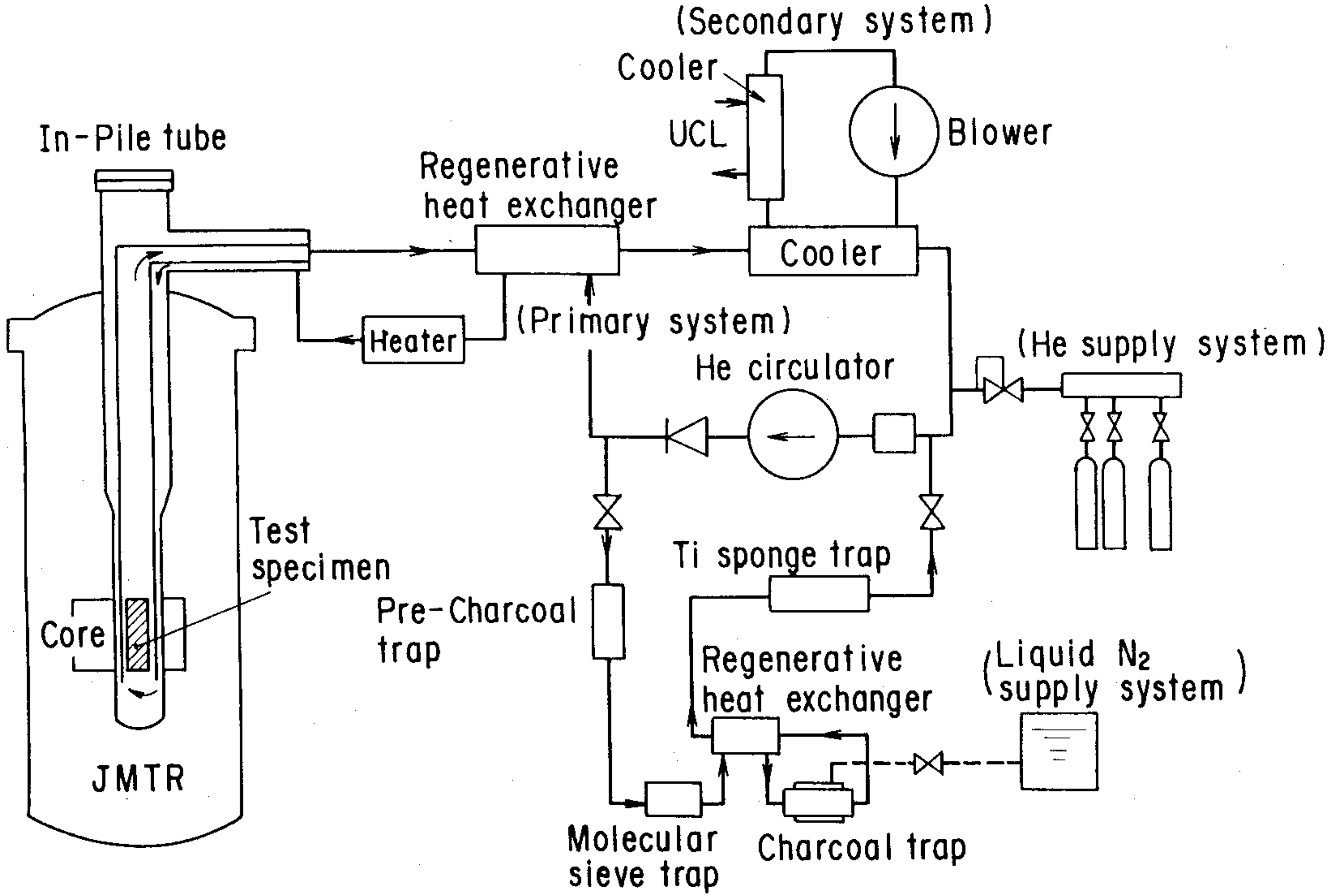


Fig. A2-1 Flow Diagram of OGL-1.

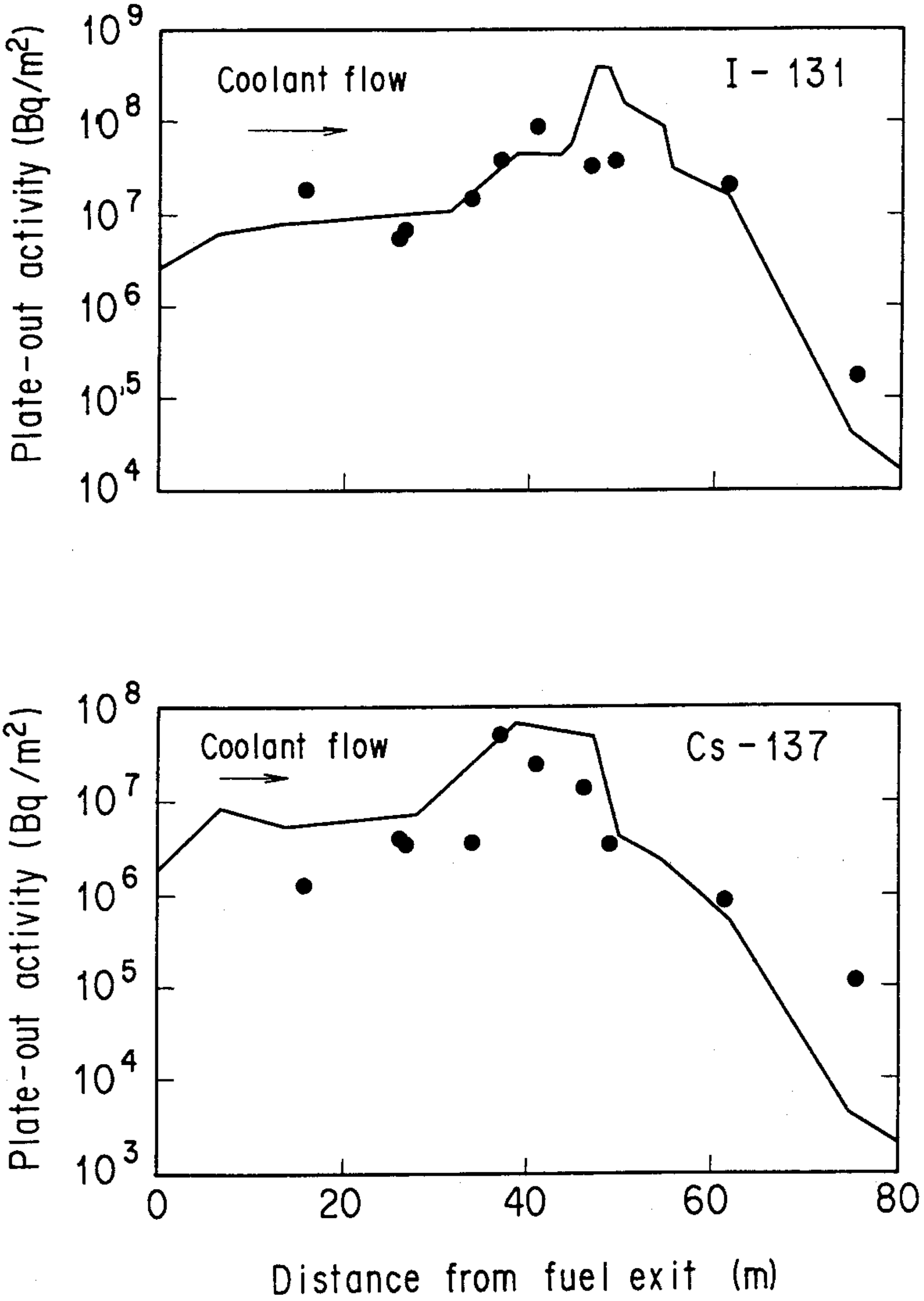


Fig. A2-2 Examples of Verification of PLAIN Code in OGL-1.