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Fuel and structral materials for the VHTR have been irradiated in JAERI. The in-pile gas loop, OGL-1, in JMTR is an important irradiation facility for VHTR fuel development. A series of irradiation tests has been performed in the loop to verify a fission product(FP) retention capability of VHTR fuels. Nine fuel elements have been irradiated since the loop was put into operation in 1977.

Development of gaseous FP monitoring system and measurement of FP plate-out behavior have been also carried out, making use of FPs released from fuel element during irradiation. Described in this paper are the topical results of those fuel irradiation tests by the OGL-1.

Keywords; VHTR, In-pile Loop, Gas Loop, OGL-1, Irradiation Test,
Irradiation Facility, Fission Product, Fuel

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インパイルガスループ（OGL－1）による燃料照射試験

日本原子力研究所大洗研究所材料試験炉部

中田　宏勝・猿田　徹・寺田　博海+
露崎　典平・福田　幸朔++

（１９８６年4月２日受理）

日本原子力研究所では多目的高温ガス炉の燃料や材料についてかねてより照射試験を行って来ている。材料試験炉（JMTR）に設置されているインパイルガスループ（OGL－１）は高温ガス炉用燃料の開発に欠かせない照射装置であって，同ループにより燃料の性能確認のための一連の照射試験が行われている。

ループは昭和５２年に完成し，以来９体の燃料要素が照射されている。同ループでは，照射中の燃料から放出される核分裂生成物を利用して一次系ＦＰ濃度監視計装の開発とＦＰプレートアウト測定も行われている。

本稿ではこれら照射試験の最近の成果について報告する。

+　原子炉工学部

++　燃料工学部

本稿は，昭和６０年１２月１１日に東京で開催された「第１４回多目的高温ガス炉研究成果報告会」における「インパイルガスループ」に関する口頭発表を英文記録したものである。

Contents

目　　次

1. Introduction

Fuel and structural materials for the VHTR have been irradiated in JAERI. Described in this paper are fuel irradiation tests by the in-pile gas loop, OGL-1 installed at JMTR.1),2)

2. Description of the loop

As shown in Fig. 1, the loop consists of the in-pile tube in the JMTR core to accommodata a fuel element and a set of out-of-pile equipments. The loop is compared to a mini VHTR, if the in-pile tube and the out-of-pile equipments are regarded as a reactor vessel and a cooling system, respectively. Helium gas delivered by the circulator enters into the in-pile tube at a temperature of 650 °C, after beeing heated by the re-generative heat exchanger and then the electric heater. The gas is further heated by the high temperature return gas in the in-pile tube, and finally heated up to 1000 °C by the fuel element. Then the gas cooled to 700 °C by a low temperature entering gas in the in-pile tube returns to the circulator at a temperature of approximately 150 °C after beeing cooled by the re-generative heat exchanger and the cooler. A part of the gas delivered by the circulator flows into the purification system and returns to the main flow line. The system consists of a charcoal trap, a molecular sieve, a cold charcoal trap and a titanium trap.

Fig. 2 shows a photograph of the JMTR core. The in-pile tube of the loop is installed in the northern part of the core, as seen at the top of the photograph. The fuel element inserted in the in-pile tube is heated by neutrons emitted by the fuel region, which consists of twenty-two fuel elements and five control rods. For reference, small-diameter tubes in the photograph are protection tubes for electrical wires and mini tube of instrumented capsules, in which irradiation samples are encased. Large diameter tubes are in-pile tubes of the hydraulic rabbit facilities, in which irradiation samples are transferred to and from the core hydraulically during reactor operation. Maximum fast neutron flux density in the loop is 1.3×10^{17} n/m^2·s, and max. thermal neutron flux density is 5.9×10^{17} n/m^2·s as shown in Table 1. Dimensions of a fuel element inserted into the loop are 80 mm in diameter and 750 mm in height. As much as three fuel rods are loaded into a fuel element, depending on rod diameter. The helium gas coolant is pressurized at 3 MPa and is able to remove fuel element heat generation of 135 KW. Maximum gas temperature is

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1000 °C at fuel element outlet. This is one of main features of the loop.

The construction program of the loop started in 1970, almost simultaneously with the start of the JAERI's VHTR program. The construction of the loop was completed at the end of FY1976, after many research and development works, as shown in Table 2. At the first fuel irradiation test conducted in March 1977, gas temperature of 1000 °C at fuel element outlet was accomplished for the first time in the world.3) Since then, fuel irradiation tests have been successfully conducted mainly for the verification of the experimental VHTR's fuel.

3. Irradiation test by the loop

Fig. 3 shows structure of fuel elements of the loop and the experimental VHTR. A fuel element of the VHTR is a hexagonal graphite column loaded with fifteen or thirty-six fuel rods, in which fuel compacts containing TRISO coated particles are inserted into a graphite sleeve, while a fuel element of the loop is a circular column load with one or three fuel rods of same size as those of VHTR's.

When coated particle fuel is used in a VHTR, it is very important that a capability of fission product(FP) retention is maintained during the whole irradiation period. If the capability is lost by failure of coating layers, not only gaseous FP concentration in coolant increases, but also radiation does of maintenance personnel increases excessively due to plate-out on inner surface of pipes and equipments of cooling system. In order to verify a capability of FP retention, a series of irradiation tests has been performed in the loop for the experimental VHTR's fuel, as shown in Table 3.4)

Monitoring system for gaseous FPs also has been developed in the loop.5) FP plate-out measurement has been carried out to study plate-out characteristics of FPs, released from the fuel, at inner surface of pipes and equipments.6) Verification of the experimental VHTR's fuel is performed not only by measuring FP release rate during irradiation, but also by post-irradiation examination. Irradiation data are mainly given in this paper. Development of a monitoring system for gaseous FPs and measurement of FP plate-out are aimed to obtain information necessary to a construction and operation of the experimental VHTR, making use of FPs released from fuel element during irradiation.

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4. Test of the experimental VHTR's fuel

One fuel element has been irradiated every year since the loop was put into operation, as shown in Table 4. The nineth element is under irradiation. Irradiations up to the eighth element were aimed to test the experimental VHTR's fuel in order to prove integrity of the fuels under VHTR operation conditions. Maximum fuel temperature was 1480 °C at the sixth element, and maximum fuel burn up was 25 Gwd/t at the fifth element. Irradiation of the nineth element is aimed to test performance of a mass-produced fuel.

Table 5 shows typical concentrations of gaseous FPs in the helium gas during irradiation of each fuel element. As to Kr-85 m, concentration was 4.8 MBq/m^3 during irradiation of the third element, had varied to 0.13, 8.1, 0.2 and then 0.09 MBq/m^3 as irradiations progressed, and finally decreased to 0.03 MBq/m^3 during the eighth element irradiation. Concentrations of Kr-87, Xe-135 and Xe-138 had changed as same as that of Kr-85 m. Therefore, it is concluded that the eighth element considerably improved its FP retention capability, compared with the earlier elements.

Fig. 4 shows a change of FP release rate during irradiation of the eighth element. Vertical axis of the figure represents release ratio, R/B, where R is a release rate and B is a birth rate. The release ratios of Xe-135 and Kr-87 had changed around 10^{-6} during irradiation. Since the release ratios were about one-twentieth of those required of the experimental VHTR's fuel, it is concluded that the eighth element had kept a excellent FP retention capability.

FP distribution in a graphite sleeve of fuel rod measured in post-irradiation examination is also interested in FP release characteristics of the fuel. Fig. 5 shows FP distributions measured on the graphite sleeve of fifth element. Vertical axis of the figure represent longitudinal position on the fuel rod along helium flow, and horizontal axis represents FP radioactivity per unit weight of the graphite sleeve. Results of measurement show that Cs-137 activity was highest and was followed by those of Cs-134 and Sb-125. Ag-110 m activity was relatively higher than those of other FPs. This is due to high burn-up of the fifth element. All distribution curves show that radioactivities at the upper end of fuel rod were higher than those at the lower end, although neutron flux density at the upper end is slightly smaller than that at the lower end. It makes clear this contradiction that FPs released at the center region, where sleeve temperature is high, was plated out on the

upper surface of sleeve, where temperature is low.

5. Development of gaseous FP monitoring system

Fig. 6 shows a helium gas flow diagram of the monitoring system designed for the experimental VHTR. The system consists of a moving wire β-detector; i.e, precipitator and a Ge-detector. The precipitator measures total radioactivity of gasous FPs such as Kr and Xe. The Ge-detector measures ratios of radioactivities of these nuclides to total activity. The performance test has been carried out by flowing helium gas containing gaseous FP from outlet of the in-pile tube.

A gamma spectrum of FPs in the helium gas measured by the Ge-detector is shown in Fig. 7. Principal nuclides identified were gaseous FPs such as Kr and Xe, but also metallic FP of Cs-138 was identified. Ratio of each nuclide radioactivity to a total activity could be obtained with this kind of gamma spectrum. Fig. 8 shows how gross count rates of these two detectors changed in response to operating condition of the loop. Fuel temperature rose to 1000 °C in response to JMTR power-up at first and secondly rose to 1350 °C in response to gas temperature increase of the loop. Count rates of two detectors increased together fuel temperature ascent. The calibration shows that the precipitator has a sensitivity of 35 cps per one Ci of Kr-88 equivalent to the experimental VHTR activity inventory, and that the Ge-detector has a sensitivity of 95 cps per Ci. These detectors have enough sensitivities, since allowable max. amount of FPs in coolant of the experimental VHTR is 100 Ci equivalent to Kr-88. Large sensitivity of the precipitator and nuclide analysis capability of the Ge-detector lead to conclusion that the FP monitoring system tested would basically satisfy a specification for the experimental VHTR. The system would be further modified.

6. FP plate-out measurement

FP plate-out in the loop has been measured along gas flow at ten points between the in-pile tube outlet and the gas circulator, as shown by the arrows in Fig. 9. Length of the pipe measured is approximately 100 m from a fuel element, and temperature is in the range from 550 °C to 50 °C. Plate-out measurements have been mostly carried out during reactor down-time, but at two of ten points, also as shown by black arrows in Fig. 9, during reactor operation in order to investigate plate-out

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characteristics of short-lived FPs.

Fig. 10 shows a system set-up for plate-out measurement. Ge-detector, located at out-side of a pipe or a equipment, detects gamma rays emitted by FPs, which is plated out at in-side of pipe or equipment. Lead-made collimator is used to detect gamma-rays only from FP deposited at a specific portion. Out-put signal is analized by a multi-channel pulse height analyzer, and then plate-out radioactivity for each nuclide is calculated by a data processing unit. Plate-out coefficients obtained with standard gamma-ray sources should be entered to the data processing unit prior to the measurement. A manner shown in Fig. 10 is called the non-destructive plate-out measuring method.

Fig. 11 shows gamma spectrum measured during reactor operation and down-time. Measuring point is at middle between the in-pile tube outlet and the re-generative heat exchanger, i.e。 approximately 25 m from fuel element, and at a temperature of approximately 550 °C. Dominant nuclides detected during reactor operation were not only FPs such as I-133, I-132, and X3-138 and its daughter Cs-138, but also corrosion products such as Mn-54 and Mn-56. During reactor down-time, long-lived FPs such as I-131, Cs-137 and La-140 were dominant, and corrosion products such as Mn-56 and Co-60 were also detected.

Fig. 12 shows plate-out distribution of I-131, which was measured during reactor down-time along the primary circuit of the loop. Horizontal axis of the figure represents distance of each measuring point from fuel element, vertical axis(left) plate-out radioactivity per unit area of pipe or equipment inner surface, and vertical axis(right) gas temperature. Shown in the figure are the results measured during irradiation of the fifth element, which had shown the largest FP release rate, and the result measured during irradiation of the seventh element, which had shown the relatively lower release rate. In each case, I-131 was much more plated out around the re-generative heat exchanger, where gas temperature is in a range from 200 °C to 350 °C, than around the inpile tube outlet, where gas temperature is at approximately 550 °C. Since half-life of I-131 is approximately 8 days, these results suggest plate-out characteristics of I-131.

Fig. 13 also shows plate-out distribution of Cs-137 measured during irradiation of the fifth and the seventh element. In contrast to the case of I-131, Cs-137 was much more plated out in a region between the in-pile tube outlet, where gas temperature is 550 °C, and the re-generative heat exchanger inlet, where gas temperature is 350 °C, than other region. Plate-out distribution of Cs-137 in this region during the fifth element irradiation was rather uniform, but during the seventh element irradiation, plate-out activity decreased at the high temperature portion of 550 °C and increased at the middle temperature portion of 350 °C. Considering that half-life of Cs-137 is 30 years, this is in consequence of that Cs-137, which was plated out during irradiation of the fifth element which released much more FPs, was broken away at the high temperature portion, and shifted to and plated out at the middle temperature portion during irradiation of the seventh element, which released less FPs.

Plate-out characteristics obtained by plate-out measurements have been used to produce a FP plate-out calculation code. Considering these characteristics, could be given effective counter-measures, such as making radiation shielding or locating FP filter at a potion, where FPs tend to plate out. Fortunately, no plate out problem has been risen in the loop, since little FPs are released from fuel elements. Maximum radiation does rate around pipe and equipment of the loop is 10 mR/h even during reactor operation.

7. Conclusions

Through successful nine-year operation of the loop and fuel irradiation test, experimental VHTR's fuels have been verified up to 25 GWD/t at 1350 °C. Gaseous FP monitoring system made up of a precipitator and a Ge-detector has been developed and tested for the experimental VHTR. And valuable knowledge has been obtained on plate-out characteristics of FPs such as I-131 and Cs-137 by plate-out measurements.

Finally for the future, in-pile tube of the loop will be replaced by a new one in 1987 because of limited life time. Fuel irradiation test will be continued after the replacement relative to new type fuel or advanced fuel which is aimed at high performance.7)

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Table 1 Main characteristics of the OGL-1

Coolant	Helium gas
Operating pressure	Max. 3.0MPa
Operating temperature	Max. 1000℃
Mass flow	Max. 0.1kg/s
Fast neutron flux(>1MeV)	1.3×10^{17}n/m^2·s
Thermal neutron flux	5.9×10^{17}n/m^2·s
Limited nuclear heat in a fuel element	135kW
Max. dimensions of a fuel element	80mm(diameter) X 750mm(length)
Number of fuel rods	3

Table 2 Progress of the construction and operation of OGL-1

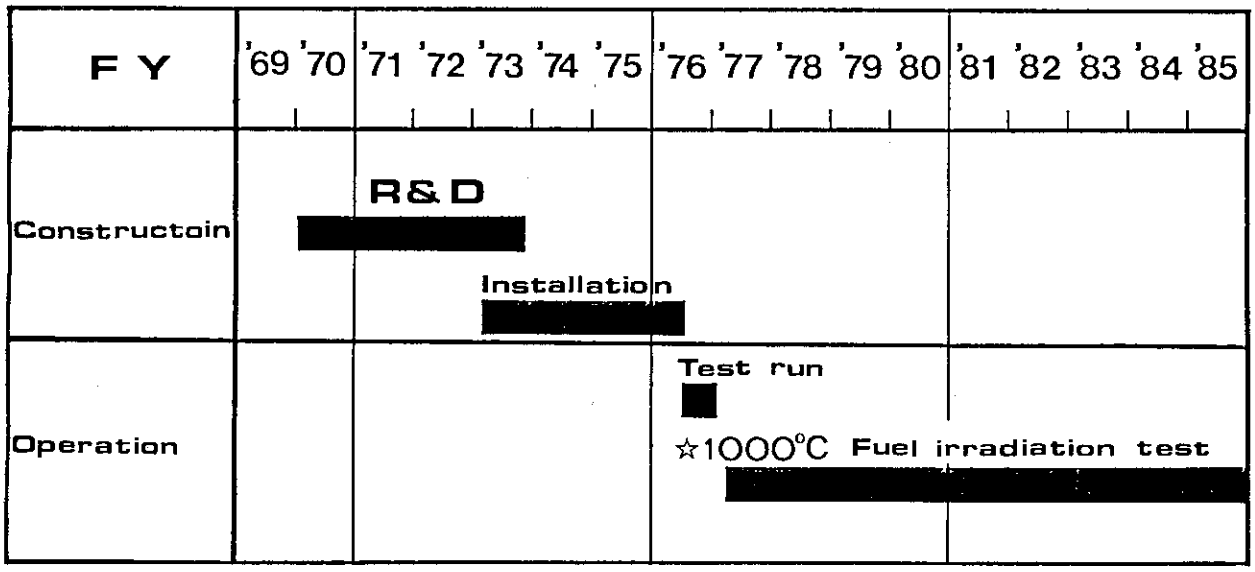

Table 3 Fuel irradiation test in the OGL-1

1. Verification of experimental VHTR's fuel
    - FP release measurement during irradiation, post-irradiation test
2. Development of gaseous FP monitoring system
3. FP plate-out measurement
    - During reactor operation, during reactor down-time

Table 4 Progress of the verification of the experimental VHTR's fuel by the OGL-1

Fuel element number	4th	5th	6th	7th	8th	9th
Year of fuel preparation	1978	1979	1980	1981	1982	1983
Purpose of irradiation	Medium burn up	High burn up	High temp. irradiation	Comparison of graphite materials	FP transport in the fuel	Performance test of Mass. products
Max. fuel temperature(°C)	1360	1350	1480	1390	1380	1340
Burn up (GWd/t)	14	25	3.5	10.4	8.2	24.5☆
Duration of irradiation(d)	78	142	22	59	54	150☆
Present status	Completed	Completed	Completed	under PIE	under PIE	under irradiation

☆ : planned

Table 5 FP concentrations in the OGL-1 herium gas during fuel irradiation tests

(MBq／m^3)

Fuel number / Nuclide	3rd	4th	5th	6th	7th	8th
^{85m}Kr	4.8	0.13	8.1	0.20	0.09	0.03
^{87}Kr	10.4	0.56	24.4	1.4	0.48	0.23
^{135}Xe	17.4	0.25	14.1	0.96	0.31	0.29
^{138}Xe	13.5	0.70	18.0	2.1	0.70	0.37

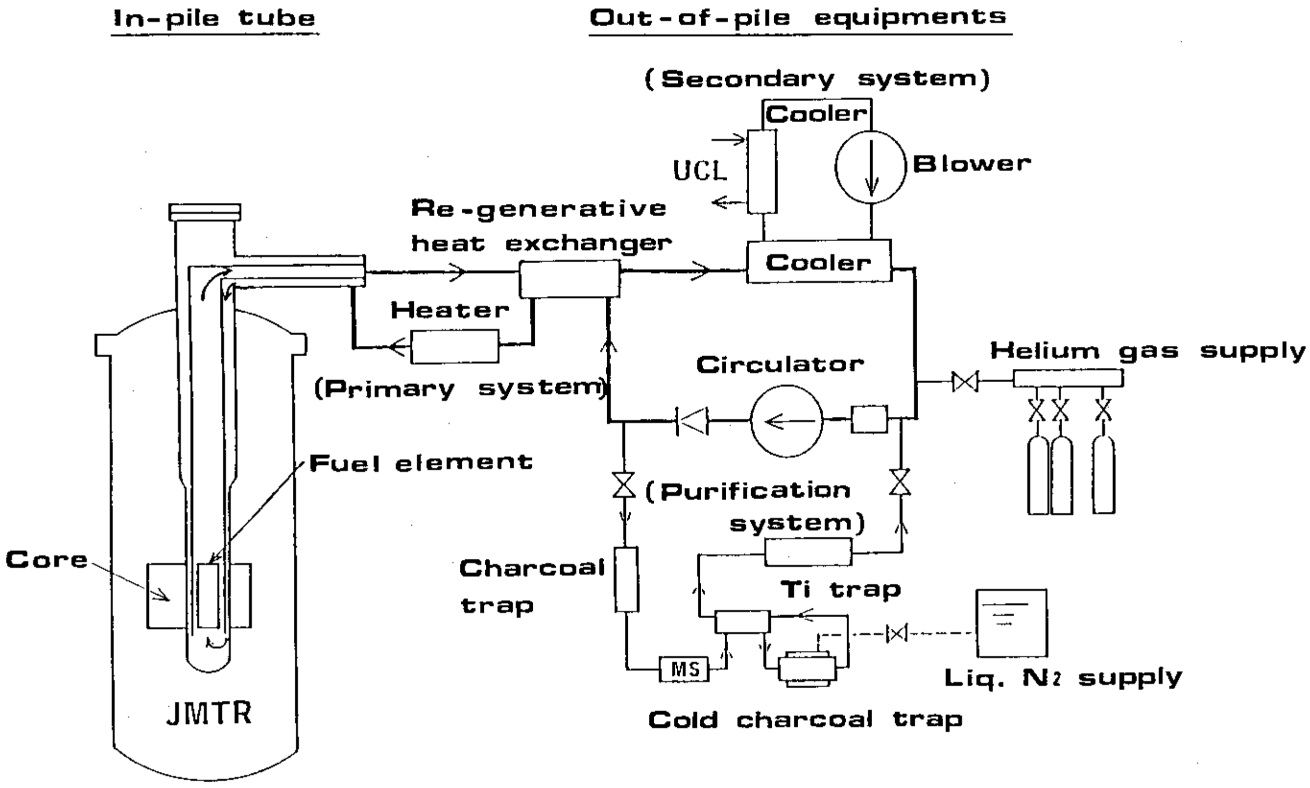

Fig. 1 Simplified flow diagram of the OGL-1

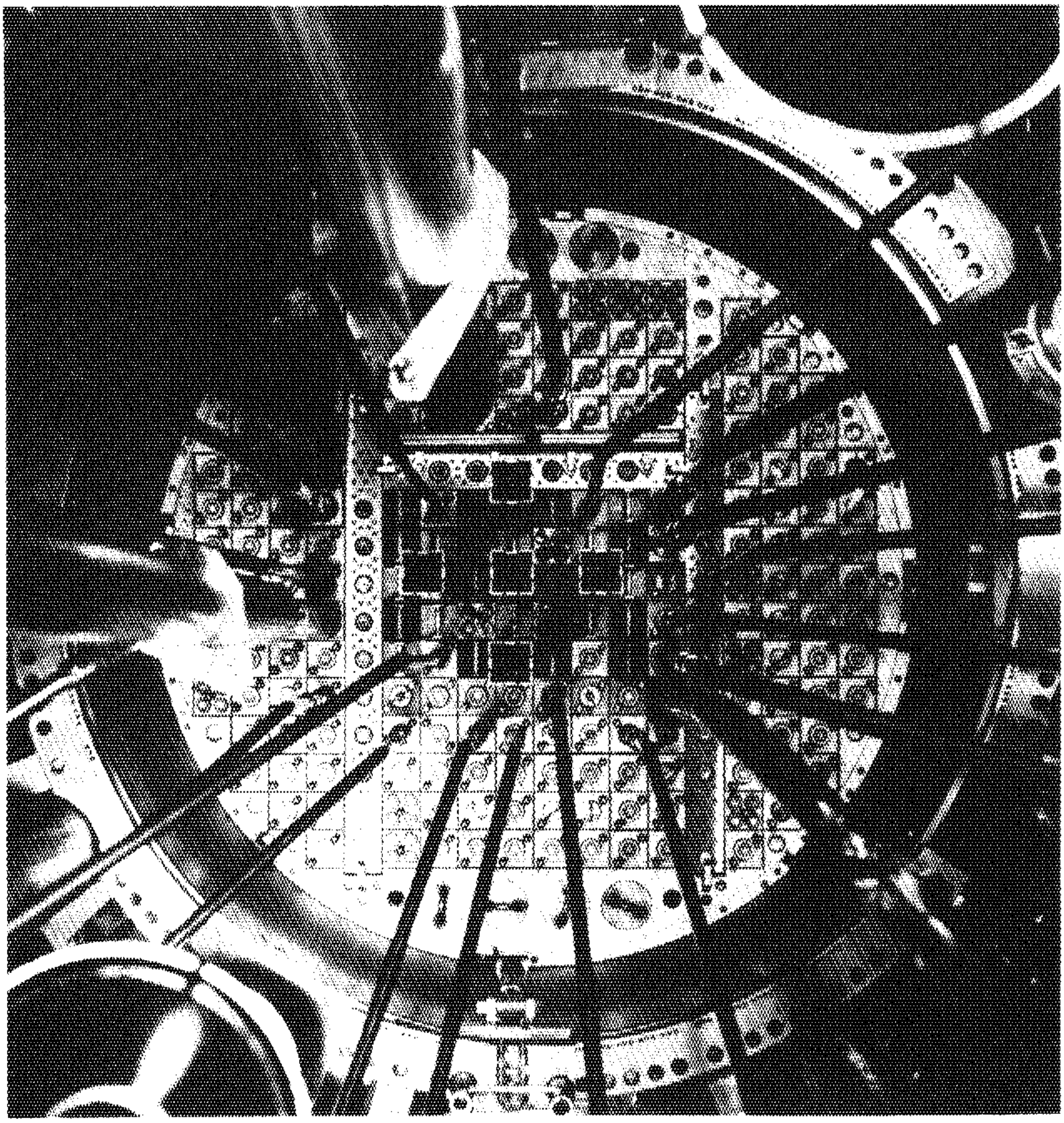

Fig. 2 JMTR Core and the OGL-1 In-pile tube

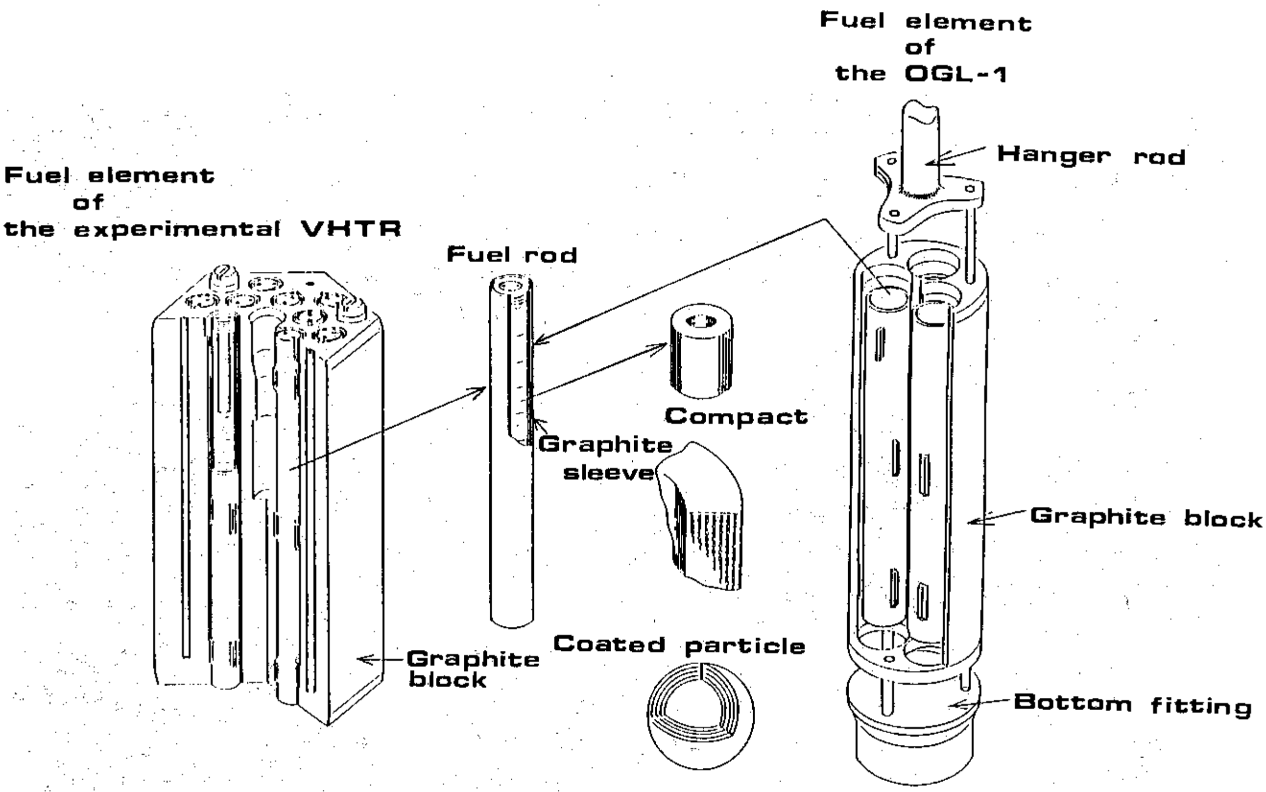

Fig. 3 Fuel elements of the OGL-1 and the experimental VHTR

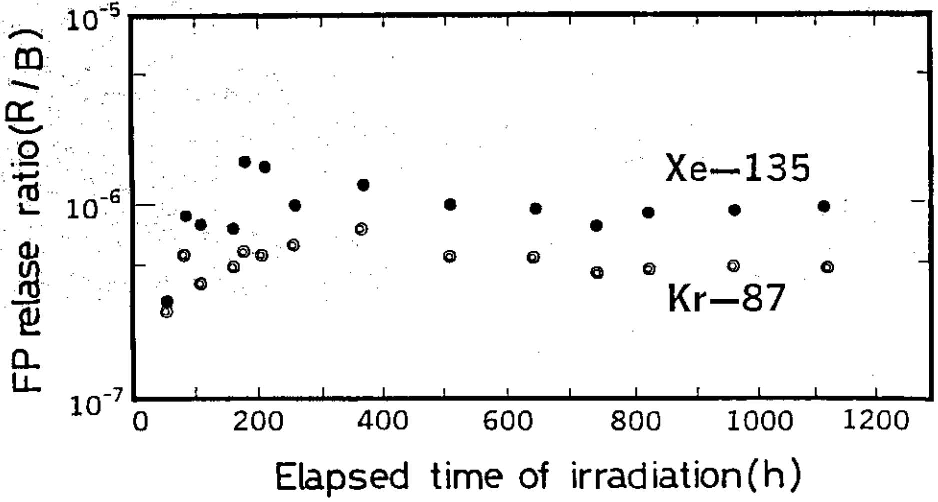

Fig. 4 Change of FP release rate during 8th fuel irradiation

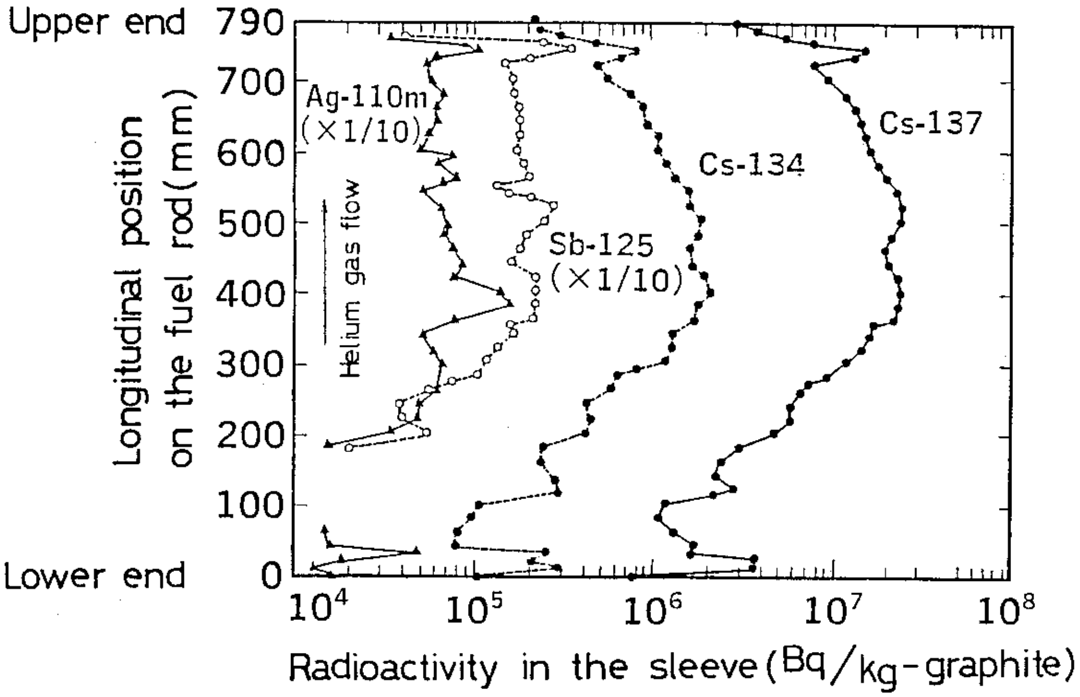

Fig. 5 FP distributions in the 5th fuel sleeve

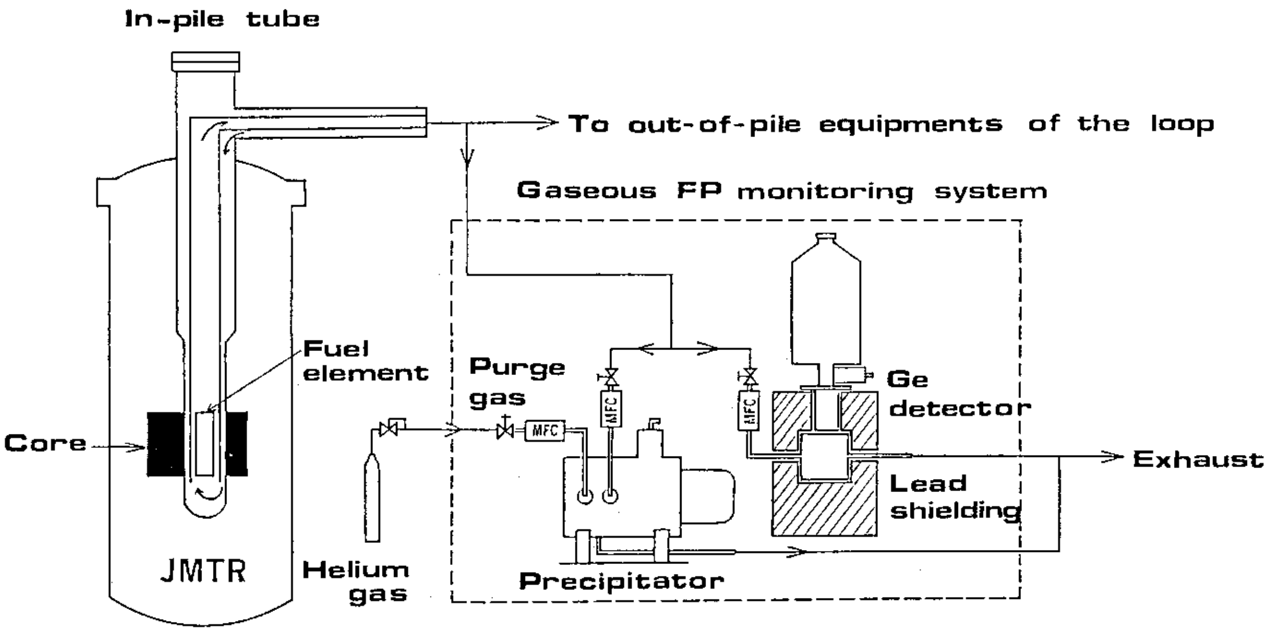

Fig. 6 Herium gas flow diagram of the gaseous FP monitoring system tested by the OGL-1

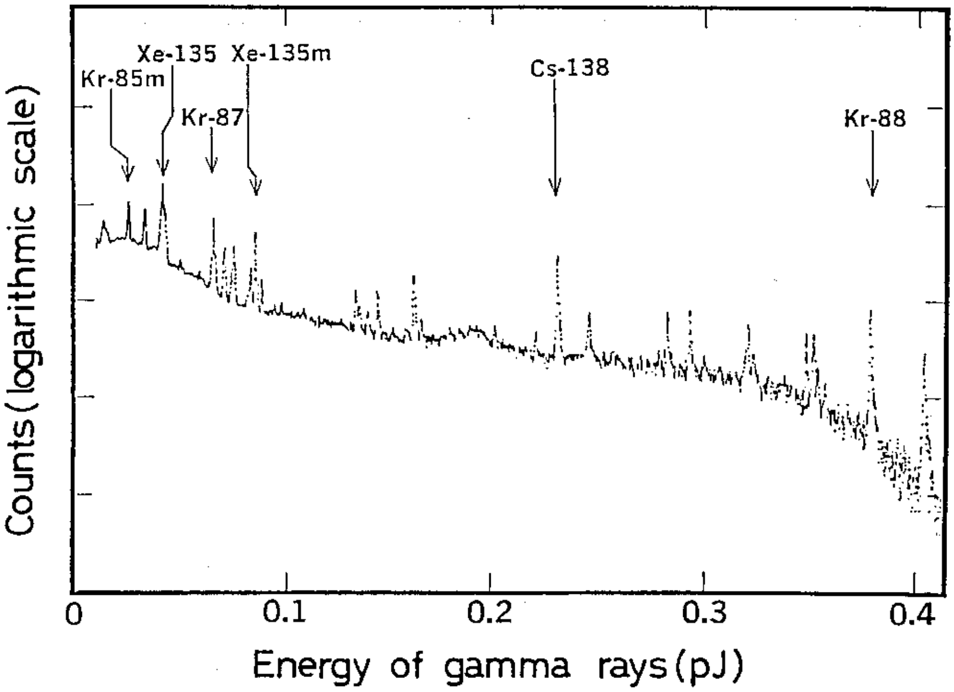

Fig. 7 FP gamma spectrum in the helium gas at the loop by the Ge detector of the monitoring system

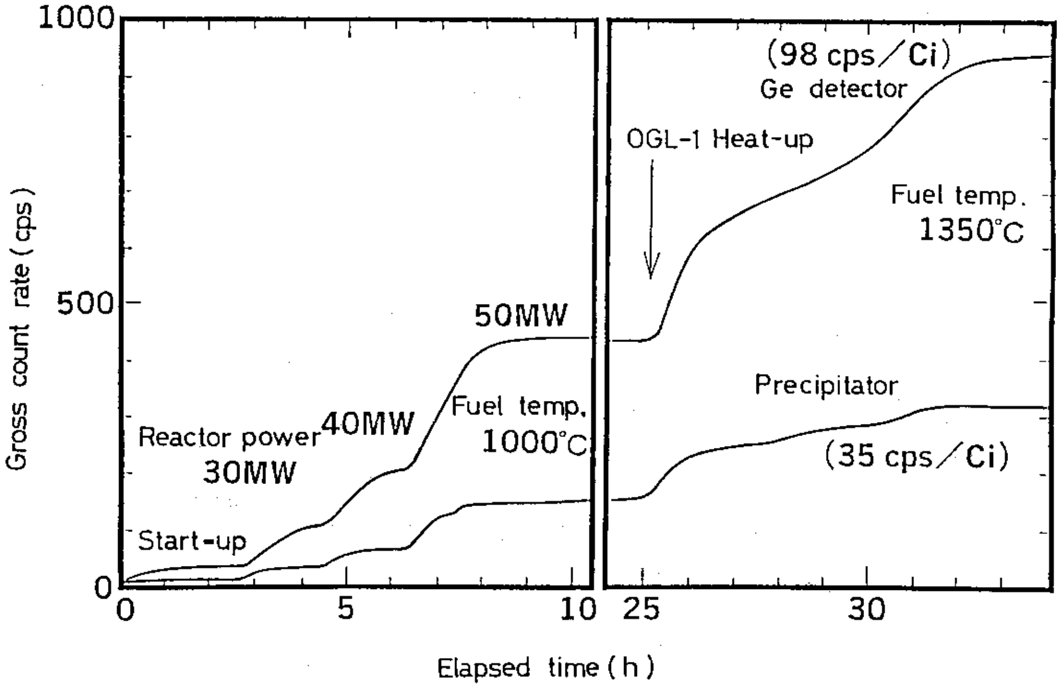

Fig. 8 A change of gross count rates of the monitoring system during the reactor start-up and the loop heat-up

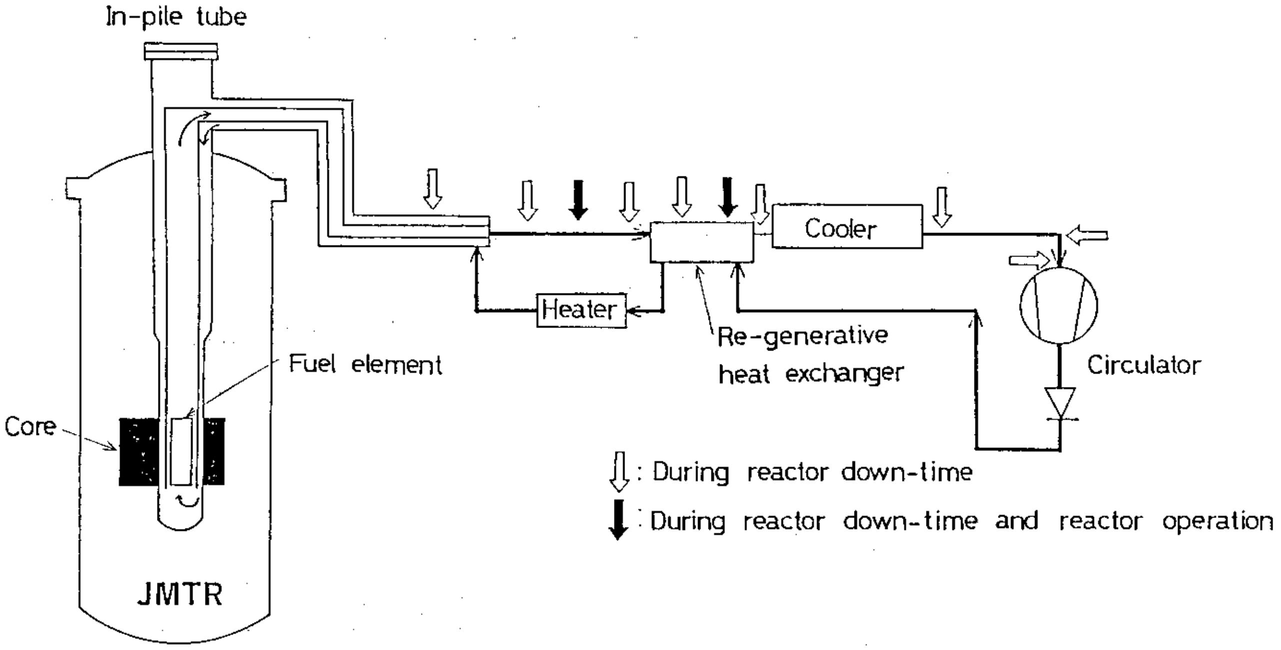

Fig. 9 FP plate-out measuring points at the OGL-1

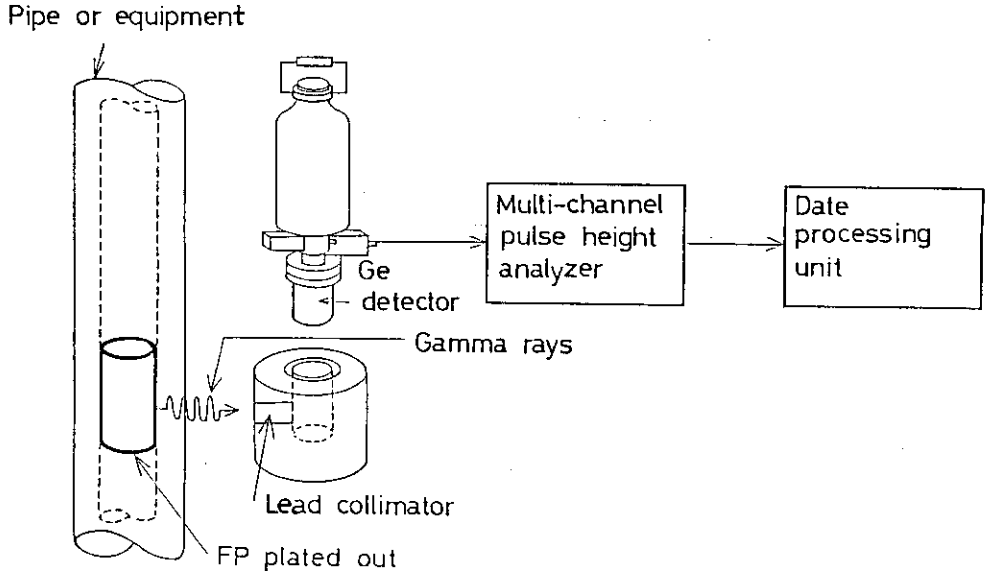

Fig. 10 FP plate-out measuring system at the OGL-1

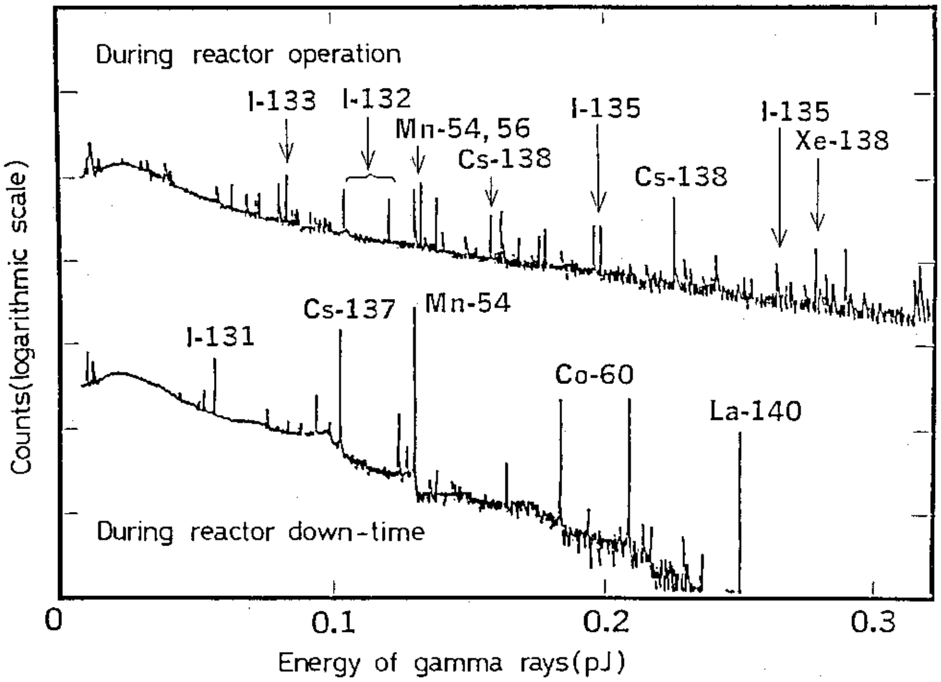

Fig. 11 Gamma spectrum of FP plated out

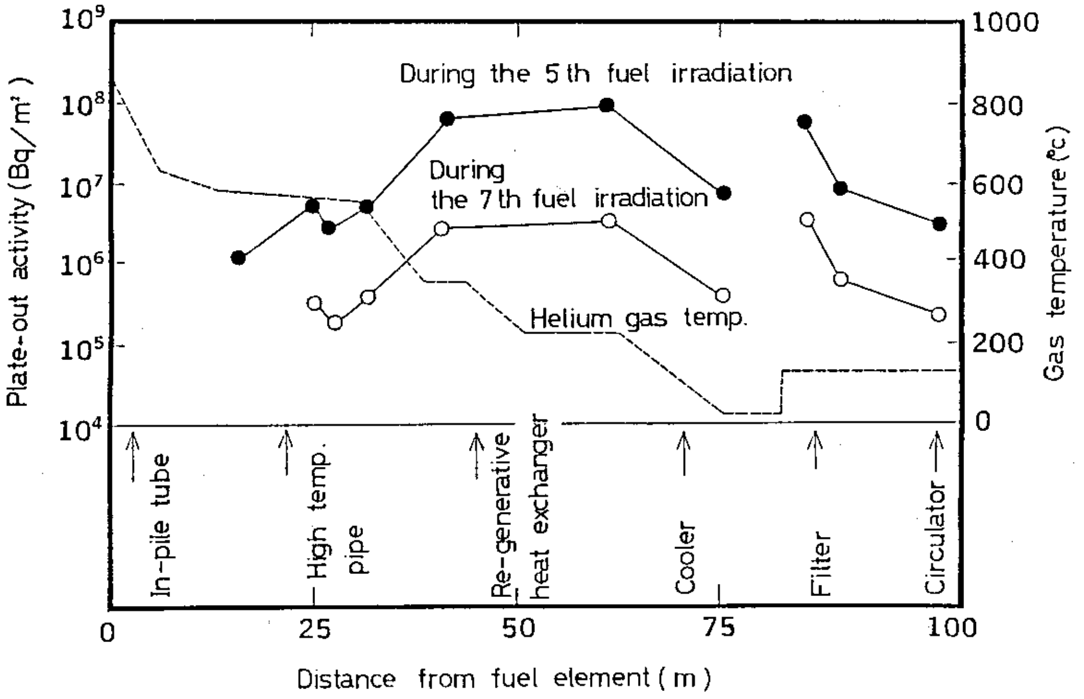

Fig. 12 Distribution of I-131 plated out during reactor down time

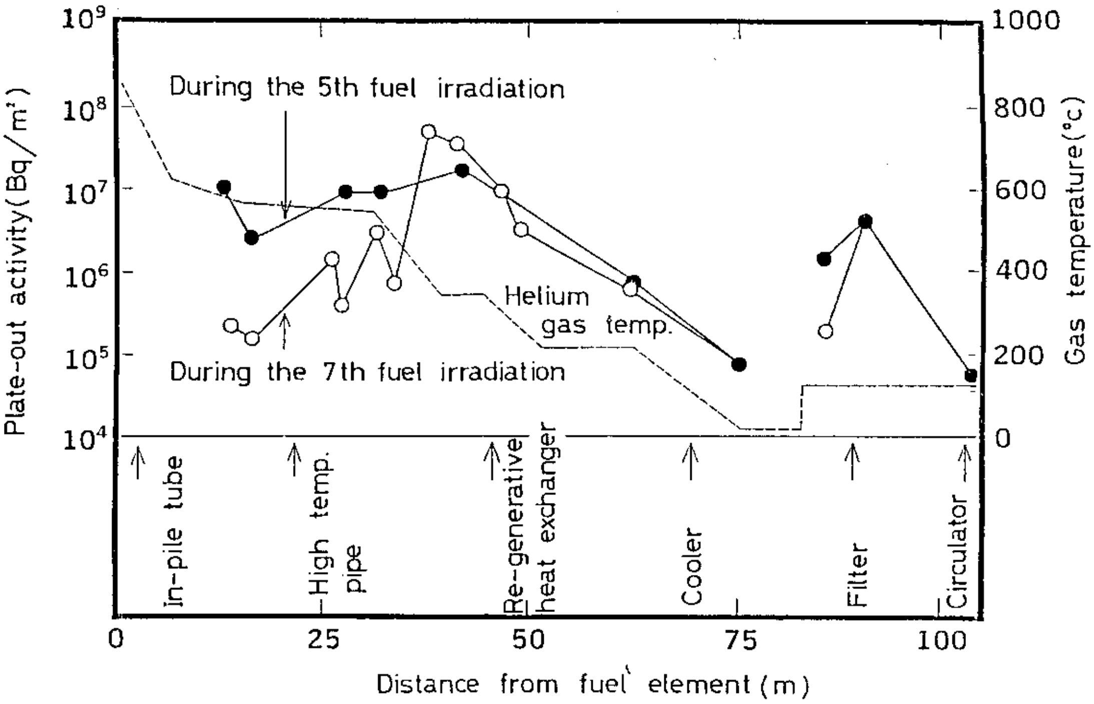

Fig. 13 Distribution of I-137 plated out during reactor down-time