RESEARCH ON IN-PILE RELEASE OF FISSION PRODUCTS FROM COATED PARTICLE FUELS

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ABSTRACT

Coated particle fuels fabricated in accordance with VHTR(Very High Temperature gas-cooled Reactor) fuel design have been irradiated by both capsules and an in-pile gas loop(OGL-1), and data on the fission products release under irradiation were obtained for loose coated particles, fuel compacts and fuel rods in the temperature range between 800°C and 1600°C. For the fission gases, temperature- and time dependences of the fractional release(R/B) were measured. Relation between release and failure fraction of the coated particles was elucidated on the VHTR reference fuels. Also, measured was tritium concentration in the helium coolant of OGL-1. In-pile release behavior of the metallic fission products was studied by measuring the activities of the fission products adsorbed in the graphite sleeves of the OGL-1 fuel rods and the graphite fuel container of the sweep gas capsules in the PIE. Investigation on palladium interaction with SiC coating layer was included.

INTRODUCTION

In the VHTR development program at JAERI, we have developed the coated particle fuels for use in this reactor. The fuels fabricated in accordance with VHTR design have been tested in the in-pile and out-of-pile works. In the study relating to the fission product migration, the fuels have been irradiated by the sweep gas capsules and and an in-pile gas loop(OGL-1), aiming principally to evaluate the fission product release under normal and transient conditions of VHTR operation. In OGL-1 irradiation experiments, tritium concentration in the helium coolant was also measured during OGL-1 operation to elucidate origin and chemical form of tritium.

Interaction of palladium,Pd, with SiC layer is the most noteworthy phenomena in the HTR TRISO fuel behavior nowadays, because the interaction deteriorates the SiC coating layer, resulting remakable fission product release. At JAERI the interaction was studied on the irradiated coated particles by EPMA (Electron Probe Micro Analyser).

IRRADIATION EXPERIMENT

Specifications of the coated particle fuels concerning the irradiation experiments are listed in Table 1. Two different sizes of LEU-TRISO coated particles consolidated into two different sizes of the annular compacts were tested. Irradiation experiments by OGL-1 and the sweep gas capsules are summarized in Tables 2 and 3, respectively. Three types of the fuels were tested in the irradiation experiments; the loose coated particles and the compacts were irradiated by the sweep gas capsules, and the fuel rods, by OGL-1.

Eight OGL-1 experiments have been finished already and 9th experiment is going on. Two types of the fuel elements were employed in these experiments as illustrated in Fig. 1, one of which was composed of three fuel rods in a graphite block and the other, of single rod[1]. In a series of OGL-1 experiments the fractional releases of fission gases from the fuel element have been obtained by measuring the fission gas concentration in the coolant. In both the 8th and 9th experiments tritium concentration was measured at several gas-sampling nozzles in the gas purge system of the loop. Measurements of the metallic fission product distributions were also made on the graphite sleeves of the fuel rods and the block after disassembly of the elements in the PIE(Post Irradiation Examination).

Irradiation experiments by the sweep gas capsules have been performed 7 times(Table 3). While the OGL-1 experiments were characterized by low fast neutron fluence(less than 0.5×10^{21} n/cm^2, E$>$29 fJ), the fluence in these experiments was relatively high, attaining up to 4×10^{21} n/cm^2(E$>$29fJ). In addition to the fission gas measurements during irradiation, metallic fission product releases were measured from the fission product concentrations in the graphite holder for the fuels.

For EPMA observation on Pd interaction , the coated particles irradiated by the sweep gas capsules, OGL-1 and the other capsules were prepared in the PIE. EPMA observation was made on more than 30 particles but the Pd interaction was detected in only 8 particles. This observation is going on.

FISSION GAS RELEASE

Time dependence

Time dependence of ^{85m}Kr release, R/B(Release rate/Birth rate), from the fuel compacts and the fuel rods measured by the sweep gas capsules and OGL-1, respectively are depicted in Fig. 2. The releases from the fuel compacts increased with irradiation time, where increasing rate was rather influenced by BOL R/B, connecting deeply with initial failure of the coated particles, than by irradiation temperature. R/B from the fuel rods, on the other hand, decreased gradually with time up to about 50 days, followed by levelling off to a certain value depending on initial failure of the coated particles in the fuel rods. The fission gas releases in SSL-1[2], GAIL loop[3,4] and Peach Bottom reactor[5] exhibited initial rapid increase caused by premature failure of the coated particles. The present initial decrease, being inconsistent with these results, might be considered to be caused by the trapping of the fission gases in the graphite matrix or the property change of the graphite matrix. However, these hypotheses could not explain the results by the sweep gas capsules, where the compacts fabricated in the same batch with OGL-1 experiment were irradiated. Presence of the graphite sleeves is suspected to relate with this decrease, but details are not known.

Temperature dependence

Temperature dependence of the typical releases from the three forms of the fuels are displayed in Figs. 3(a) and (b). In Fig. 3(a) it is seen that the releases from all of the fuels increase monotonously with temperature where the release levels depend on the degree of the exposed uranium. Gradient of the release to the temperature is the greatest for the loose coated particles among the fuels. R/B controlled by diffusion is expressed as

$$R/B = 3\left(\frac{D}{\lambda a^2}\right)^{1/2} \quad \cdots\cdots\cdots \quad (1)$$

and by substituting $D=D_0 \exp(-\frac{\Delta E}{RT})$ in this equation,

$$\lambda (R/B)^2 = A \exp(-\frac{\Delta E}{RT}) \quad \cdots\cdots\cdots \quad (2)$$

is obtained, where

D= diffusion coefficient, cm^2/s
D_0= pre-exponential factor, cm^2/s
λ= decay constant, s^{-1}
a = equivalent sphere radius, cm
ΔE = activation energy for diffusion, kJ/mol
R = gas constant, 8.314 kJ/mol
T = temperature, K
A = constant , s^{-1} .

The releases in Fig. 3(a) are plotted according to Eq. 2 in Fig. 3(b), where the activation energies are included. From the energies for the fuel compacts and the fuel rods, both of which are around 100 kJ/mol, it is evident that the releases are controlled by bulk diffusion probably in the graphite grains of the compact matrix. The energies agreed roughly with the activation energy for ^{85m}Kr release(ΔQ=69.6 kJ/mol) from Fort St. Vrain fuel rods[6], where $\Delta Q = \Delta E/2$ should be considered. The energy for the release from the loose coated particles, 252 kJ/mol above 1150°C was fairly the same as that of Xe diffusion in PyC[7], implying that the release was controlled by bulk diffusion in OLTI-PyC of the particles, where fission recoil release from the contaminated uranium in the OLTI-PyC was involved. The low energy less than 60 kJ/mol was owing to the gas phase diffusion[6].

Release and coating failure

Various attempts have been made for prediction of the failure fraction of the coated particles[2,8,9] under irradiation. For this object, a relation between R/B at EOL and mean failure fraction of the coated particles was studied and the results are depicted in Fig. 4. It is seen that the release expresses a gentle slope in the region of the failure fraction less than 10^{-3}, which originated from the contaminated uranium, and a relatively steep increase at the higher failure fraction by coating failure. This relation agreed with the computed results in the VHTR design, indicating that the computation for the release was verified experimentally.

Tritium measurement

The results of measurements on tritium concentration in the coolant of OGL-1 are shown in Fig. 5. In spite of remakable difference of the fission gas releases in the 8th and the 9th experiments(Fig. 2), the tritium

concentraions in the both experiments were the same together, implying that the source of tritium was not the fuels. Preliminary calculation suggested that the major sources were ^{6}Li impurity in the graphite and ^{3}He in the helium coolant. From the tritium concentration measurements at the several points in the gas purge system, it was found that chemical form of tritium was hydrogen gas, H-T, since the tritium concentration decreased after passing through only the titanium trap as shown in Fig. 5, which could remove hydrogen.

METALLIC FISSION PRODUCT RELEASES

Profiles of the release in the graphite sleeves of the fuel rods

As for the metallic fission product behavior in the fuel rod, profiles of ^{137}Cs fractional releases into the graphite sleeves of the fuel rods are depicted in Fig. 6. Indicated in this figure is that levels of the fractional releases depended roughly on the average failure fractions(EOL) of the coated particles, where the irradiation temperatures were nearly the same together except the 6th experiment(Table 2). The high releases at the top end of the sleeves were supposedly due to adsorption from the coolant. The fractional releases of various fission products are compared in Fig. 7, where the 5th experiment is presented as a typical example. Ag-110m release was remakably large, of which level was about two orders of magnitude higher than these of Cs. Eu-155 release was exceptionally high at the center of the sleeve, suggesting strong temperature dependence of this release.

Release in the sweep gas capsules

In-pile releases of metallic fission products measured by the sweep gas capsules are shown in Fig. 8. The horizontal axis of this figure is expressed as non-dimensional diffusion parameter,D't, where D' is a reduced diffusion coefficient of Cs in UO_2[10] and t, irradiation time. For plotting Ag release, D' of Ag was assumed to be the same as that of Cs. Cs release remained in the region of order of 10^{-2}, whereas Ag release was about one order of magnitude higher than that of Cs. In Table 4, failure fractions of total coatings and SiC layer of the fuels designated as 75F5A-1 and 75F5A-2 are listed with the releases and the irradiation conditions. The failure fractions of the total coatings for both the fuels were not so different, whereas the SiC layer failure fraction of 75F5A-2 was about 3 times as large as that of 75F5A-1. Despite of such difference in the SiC layer failure fraction, the Cs and Ag releases from 75F5A-2 were about 20 times and 10 times higher than those from 75F5A-1, respectively. This might imply that retention ability for these fission products reduced rather by increasing irradiation temperature from 1200°C to 1400°C than by increasing the SiC layer failure fraction. As for the Ag release from TRISO coated particles, Nabielek et al.[11] concluded that irradiation temperature should be kept at less than 1250°C for Ag retention. Our results suggested generally their conclusion, despite that Ag release at present showed a high fraction. Present case might be affected by fractuation of irradiation temperature.

Palladium interaction with the SiC coating layer

EPMA observations on coated particles suffered severely by Pd in our experiemnts are presented in Fig. 9. These particles consolidated in the compacts were irradiated in 78F3A capsule(Table 3). Two conspicuous features are noted in the observations; firstly, Pd accumulation occurred along boundaries of SiC/ILTI-PyC, buffer PyC/ILTI-PyC and the other place where probably the density of ILTI-PyC changed. Secondly, small carbon precipitations were found in the Pd interaction zone of the SiC layer. These features might be foundation for elucidation of Pd interaction mechanism. Pd penetration rates into the SiC layer obtained at JAERI are plotted in Fig. 10, where the reported values of the particles having oxide kernels[12-16] are included. Our results agreed fairly with those oxide values. This experiment is going on.

SUMMARY

Irradiation experiments on in-pile release of the fission products from VHTR reference fuels have been performed by the sweep gas capsule and the in-pile gas loop, OGL-1.

In the fission gas release behavior, time dependence of the releases from the compacts and the fuel rods exhibited different manners. Temperature dependence, on the other hand, revealed that the releases from the comapcts and the fuel rods above 1150°C were controlled by bulk diffusion probably in the graphite grains of the compact matrix. Furthermore, relation between the release and failure fraction of the coated particles was studied. By tritium measurement in the coolant of OGL-1, it was found that tritium source was not the fuel, and its chemical form was hydrogen gas,H-T.

Concerning the metallic fission product release, the profiles of the fractional releases of the fission products were obtained in OGL-1 experiments, and the fractional releases were also measured by the sweep gas capsules. As for Pd interaction with SiC layer, the data on temperature dependence of Pd penetration into the SiC layer were obtained.

ACKNOWLEDGMENT

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Table 1 Specifications of the fuels used for irradiation.

Coated Particles		
Coated Particle Sort	A	B
VHTR Design referred	Preliminary Design	Mk-III Design
Kernel Material	LEU-Oxide	LEU-Oxide
Diameter (μm)	500	600
Coating Thickness(μm)		
Buffer	40	60
ILTI- PyC	30	30
SiC	25	25
OLTI-PyC	45	45
Particle Diameter(μm)	780	920
Fuel Compact		
Compact Sort	Small Size	Large Size (Mk-III Design)
Coated particles included	A or B	B
Outer Diameter(mm)	24	36
Inner Diameter(mm)	8	18
Length (mm)	36 or 40	36

Table 2 Irradiation experiments by OGL-1

OGL-1 Irradiation Experiment	Object of Irrdiation	Type of fuel element*	Irradiation Conditions		
			Max. Temperature (°C)	Max. Burnup (% FIMA)	Irrad. Time (EFPD)
1st	Test for loop	M	1360	0.6	39.0
2nd	Preliminary irradiation of Mk-III fuel	M	1425	1.0	59.4
3rd	Fuel rod bowing	S	1360	0.5	40.7
4th	Burnup to medium level in VHTR design	M	1400	1.5	78.0
5th	Burnup to VHTR design level	M	1370	2.6	142.1
6th	Irradiation for reactor transient condition	S	1480	0.3	21.9
7th	Comparison of compact matrix material	M	1390	1.1	60.0
8th	Fission product migration in element	S	1340	0.7	53.8
9th**	Irradiation for fuels fabricated on scale-up	S	(1320)	(2.5)	(150)

* M-type fuel element contains 3 fuel rods in a graphite block, which load 60 compacts in total.
S-type fuel element contains single fuel rod in a graphite block, which loads 20 compacts in total.

** 9th experiment is going on.

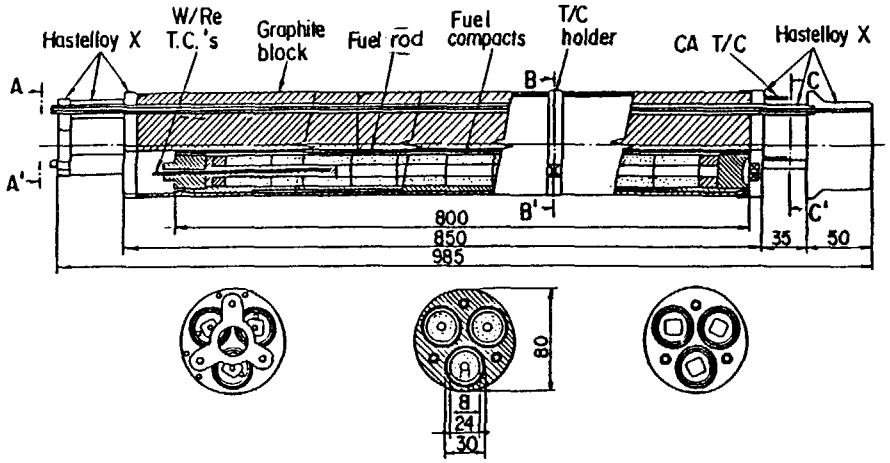
Table 3 Irradiation experiments by sweep gas capsules

Sweep Gas Capsule Irradiation	Object of Irradiation	Type of Fuel Sample	Irradiation Conditions			
			Max. Temp. (°C)	Max. Burnup (%FIMA)	Irrad. Time (EFPD)	Fast Neutron (cm^{-2}, E > 29fJ)
74F9J	Test for facility	Particles	1600	2.3	50.5	$0.7^{x10^{21}}$
75F4A	Temperature variation	Particles	1500	3.5	83.1	1.1
75F5A	Temperature variation	Compacts	1600	1.3	78.0	1.1
76F4A	High flux irrad.	Particles	1180	0.8	36.5	0.9
76F5A	High flux irrad.	Compacts	1380	3.2	101.0	4.0
78F3A	High temperature	Compacts	1550	2.7	81.9	2.9
80F4A	Fuel rod irrad.	Compacts	1300	2.9	80.0	2.4

Table 4 Relation between coating failure and release

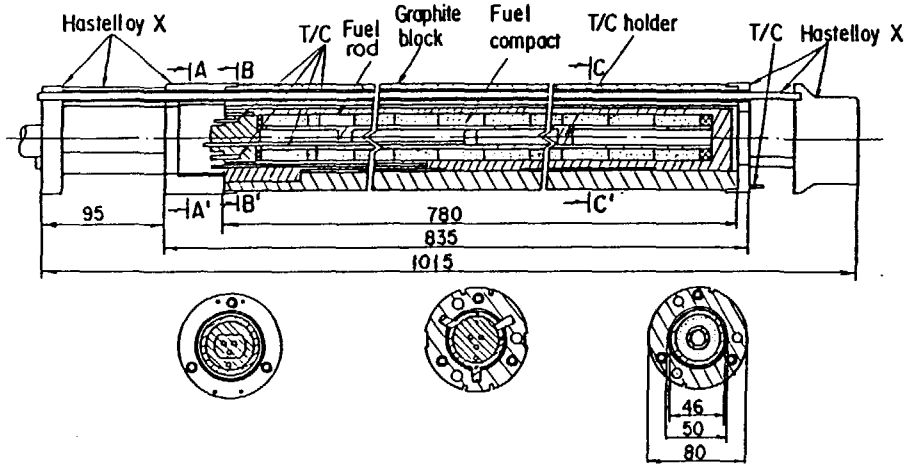
Fuel Sample	75F5A-1	75F5A-2
Burnup (%FIMA)	0.8	1.3
Irradiation Temperature(°C)	800 - 1500	900 - 1600
Averaged Temperature(°C)	1200	1400
Fast Neutron Fluence (cm^{-2}, E > 29fJ)	$0.7x10^{21}$	$1.1x10^{21}$
Total Coating Failure Fraction(EOL)	$5.9x10^{-4}$	$7.8x10^{-4}$
SiC layer Failure Fraction(EOL)	$1.6x10^{-3}$	$5.6x10^{-3}$
Fractional Release of		
^{137}Cs	$4.5x10^{-5}$	$1.3x10^{-3}$
^{134}Cs	$5.1x10^{-5}$	$1.2x10^{-3}$
^{110m}Ag	$7.0x10^{-3}$	$7.7x10^{-2}$

[A] OGL-1 fuel body holding 3 fuel rods



A-A' cross section B-B cross section C-C' cross section

[B] OGL-1 fuel body holding single fuel rod



A-A' cross section B-B' cross section C-C' cross section

Fig. 1 Structure of OGL-1 fuel element.

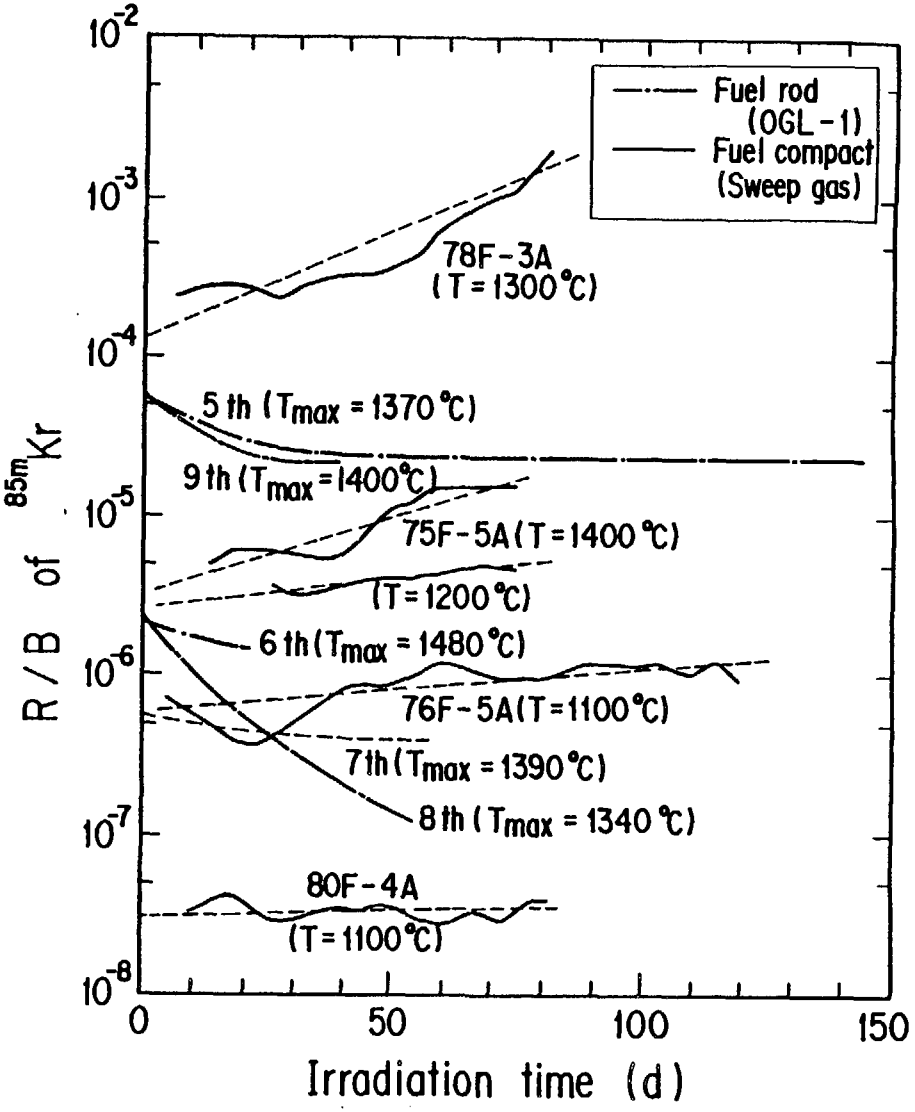


Fig. 2 Time dependence of fission gas releases from the fuel rods and the fuel compacts.

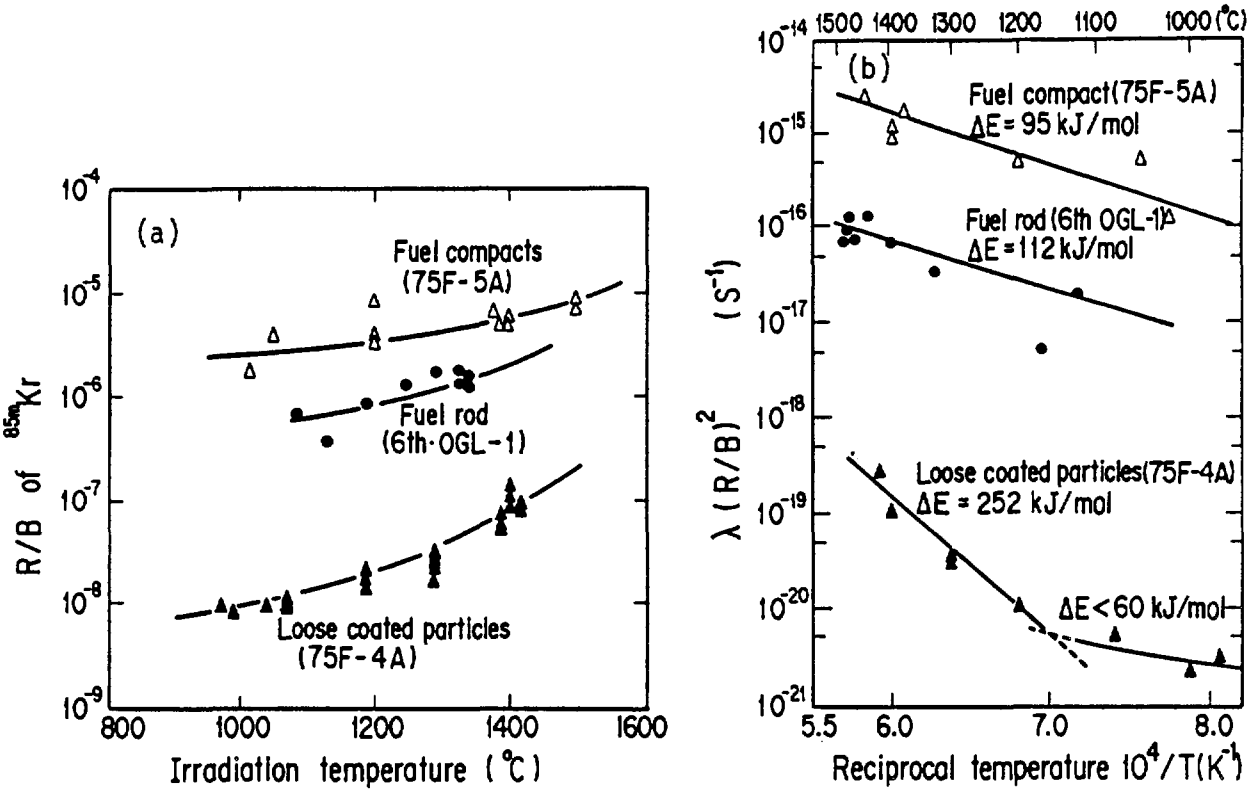


Fig. 3 Temperature dependence of fission gas releases from the loose coated particles, the fuel compacts and the fuel rods.

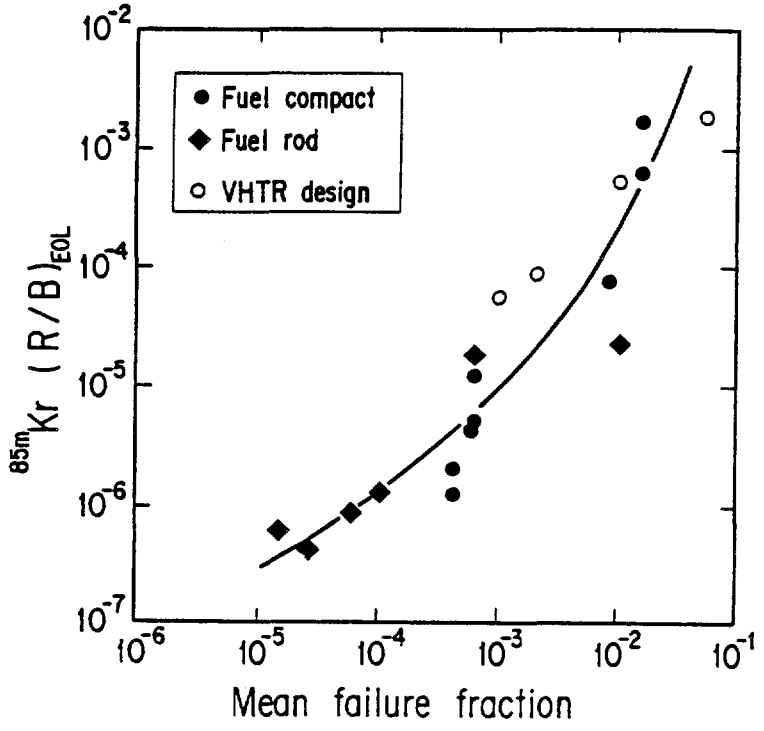


Fig. 4 Relation between fission gas release and failure fraction.

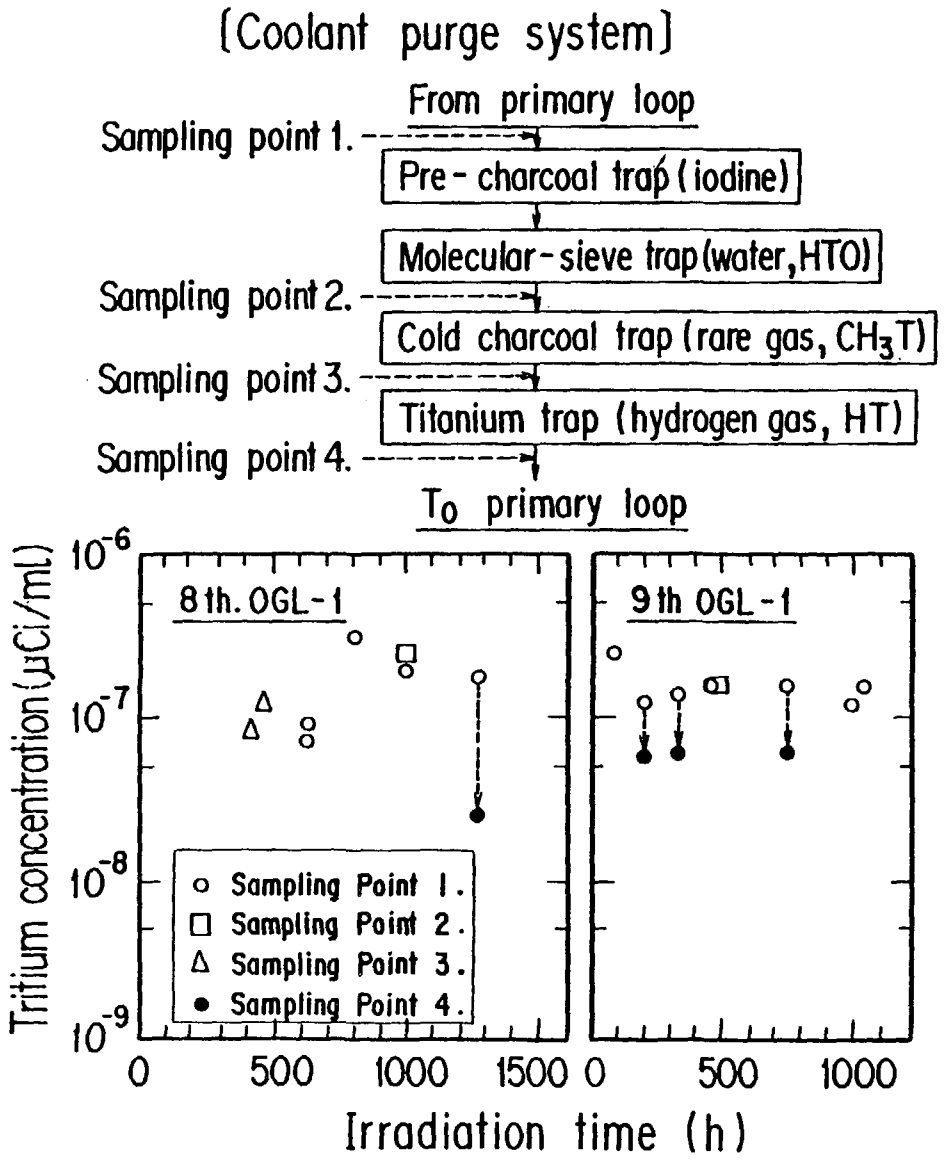


Fig. 5 Tritium concentration in the coolant of OGL-1 loop.

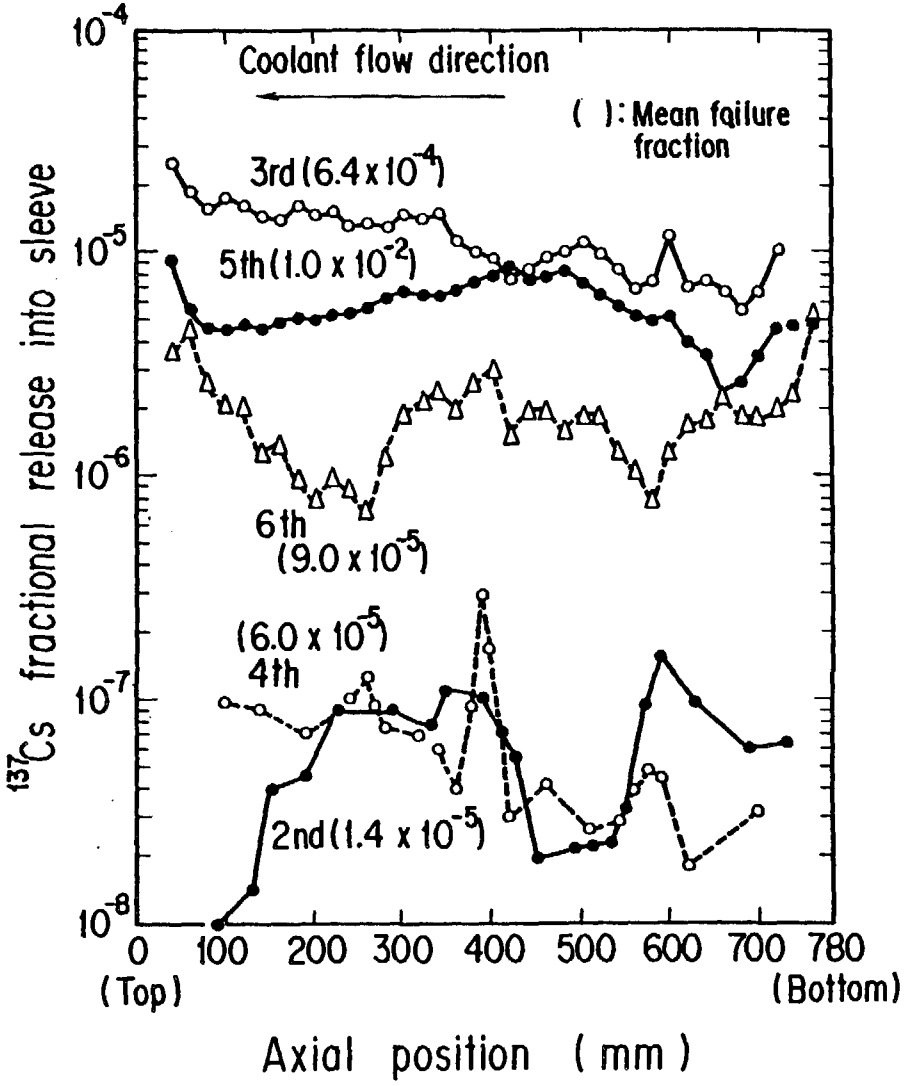


Fig. 6 ^{137}Cs fractional releases from the fuel compacts into the sleeves in several OGL-1 experiments.

Fig. 7 Fractional releases of metallic fission products into the sleeves in the 5th OGL-1 experiment.

Fig. 8 Metallic fission product releases from VHTR fuels measured by the sweep gas capsules.

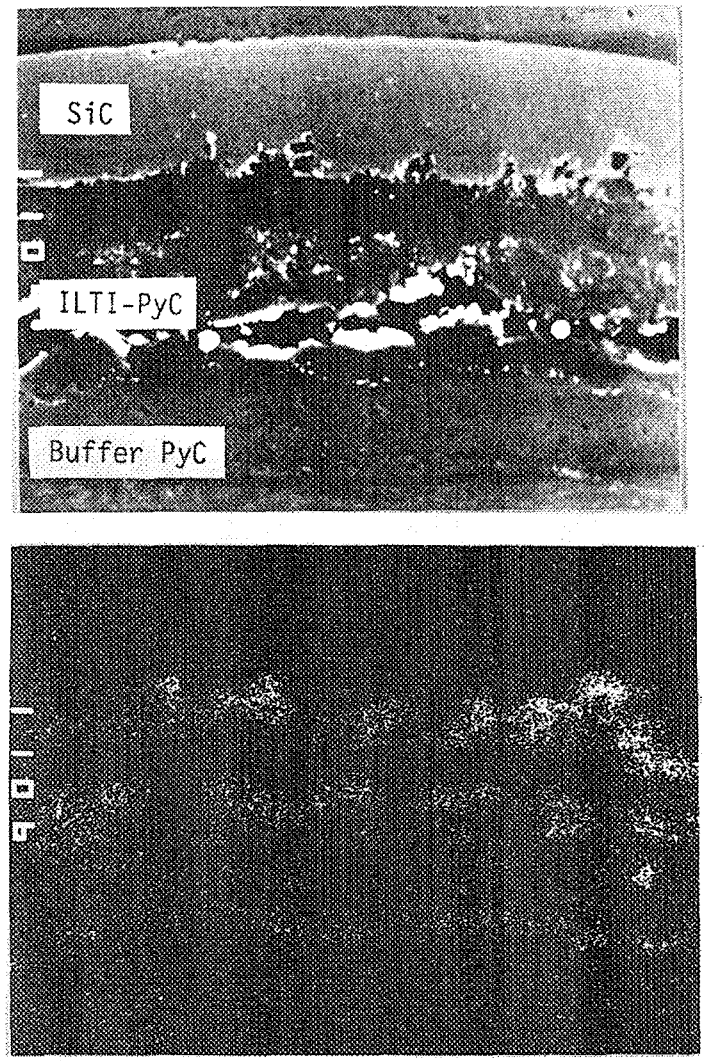


Fig. 9 Electron Prove Micro Analysis of palladium/SiC layer interaction in the irradiated coated particles; (upper) secoundary electron image, (lower) Pd-L image of EPMA.

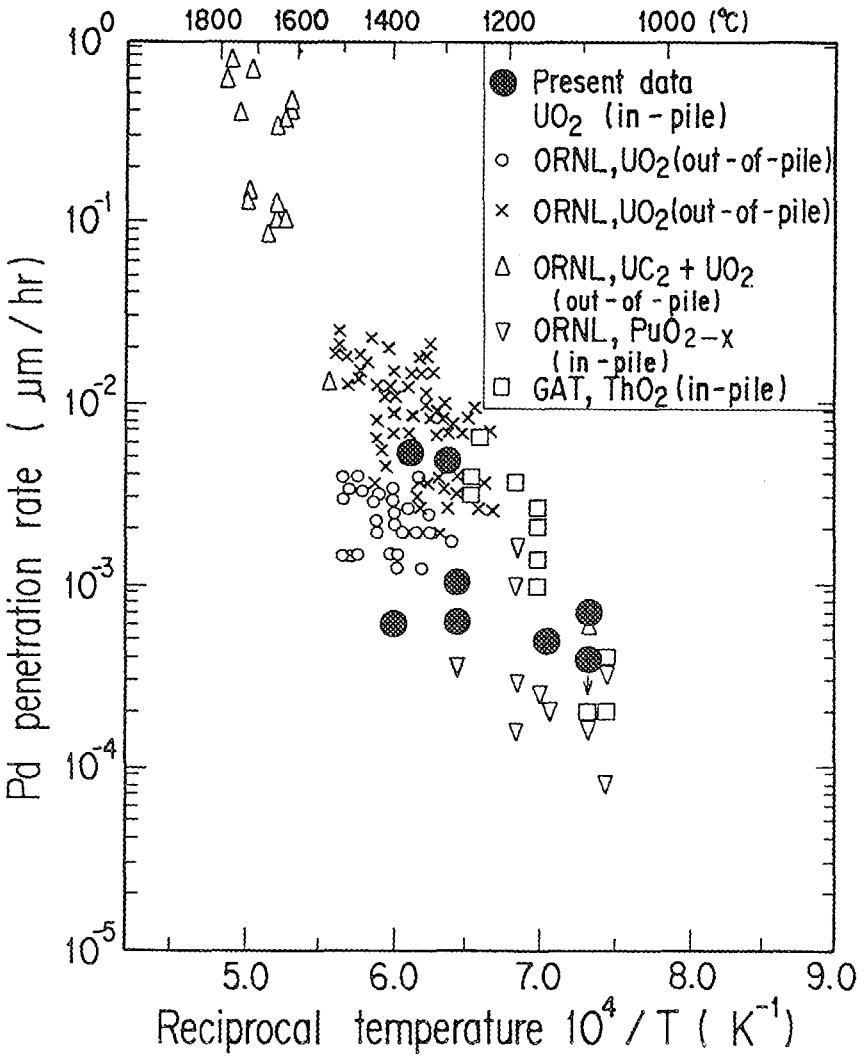


Fig. 10 Pd penetration rate into SiC layer

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SiC layer Failure Fraction(EOL)	1.6×10^{-3}	5.6×10^{-3}
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