4. Studies on Fuel Failure Detection in Rikkyo Research Reactor

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ABSTRACT

Studies on fuel failure detection have been made since 1986 in Rikkyo Research Reactor. One of the methods is the monitoring of the trace concentration of fission products appearing in the air on the surface of the water tank of the reactor. The interested radionuclides here are ^{89}Rb and ^{138}Cs, which are the daughter nuclides of the FP rare gas nuclides, ^{89}Kr and ^{138}Xe, respectively and have the half lives of 15.2 min and 32.2 min respectively. They are detected on a filter paper attached on a conventional dust sampler, by sucking the air of the surface of the water for 15 ~30 min during reactor operation (100 kW). In this presentation are reported the results of an attempt to increase the sensitivity of detecting these nuclides by introducing nitrogen gas bubbles into the water. The bubbling of the gas increased the sensitivity as much as several times compared with the case without bubbling. These measurements are giving us the "backgound" concentration, the order of which is almost unchanged for these several years,--in 10 $^{-6}$ Bq/cm^3. The origin of these nuclides is considered to be not from the fuel but from the uranium contained as an impurity in the reactor material in the core.

INTRODUCTION

Studies on fuel failure detection have been made since 1986 in Rikkyo Research Reactor (TRIGA, 100kW) in four approaches: 1) monitoring of fission products (FP) in the air covering the reactor cooling water, (2) monitoring of FP in the water, (3) the development of a "sniffer" device for detecting the location of failure fuel element when some anomaly is found in routine monitoring, and (4) the development of "sipping" inspection method of the fuel element.[1,2]

In the first method, the fission-originating two radionulides, ^{89}Rb, and ^{138}Cs, contained as an aerosol in the air covering the water in the reactor tank, are caught on a filter paper through which the air is sucked for sampling. These are the daughters of the rare gas fission products, ^{89}Kr ($T_{1/2}$=3.18 min) and ^{138}Xe ($T_{1/2}$ =14.1 min) respectively, which are produced somewhere in the reactor core and dissolved into the water. This method has been regarded to be the most sensitive for the routine monitoring of the fuel elements in several TRIGA facilities.[3,4]

We have recently attempted to improve the detection efficiency in this method, by introducing nitrogen gas into the reactor water to purge the fission rare gases dissolved in the water. By employing this "bubbling method", the sensitivity of

detection increased as much as several times, compared with the cases without bubbling of the gas. The results obtained up to present is reported in this paper.

EXPERIMENTAL

Fig. 1 shows the schematic setup of the system. During reactor irradiation (100kW), nitrogen gas was introduced into the reactor water through a porous exit in various flow rate (from ～0.2 to ～10 liter/min). The depth of the nitrogen gas exit in the reactor tank was varied also (from～430 to ～100 cm, from the top of the water surface). During introducing nitrogen gas, the air which covers the primary cooling water was sucked by a conventional dust sampler equipped with a filter paper by the flow rate of ～500 liter/min. After sampling the air for 15 or 30 min, the radioactivity of the filter paper was measured with a Ge-Li detector with MCA, by counting the photo-peaks of 1032 and 1434 keV due to ^{89}Rb and ^{138}Cs, respectively. The data were normalized to the time of the end of sampling, using the half lives of these nuclides, i.e. 15.2 min and 32.2 min respectively. The experiment without bubbling was also made, and the results were compared.

RESULTS

Preliminary experiments

Table 1 shows the data in the experiments done in fiscal 1990. From these data the followings were observed:
(1) The concentration of ^{138}Cs is almost always higher than that of ^{89}Rb.
(2) The bubbling of nitrogen gas clearly increased the detecting sensitivity of these nuclides, as shown in the ratio of the activities when bubbled to those without bubbling. The ratio, which is shown in the parentheses in the table, increased about three times compared with the case of no bubbling on the average.

Dependence on the position of the exit and flow rate of introducing gas

The dependence on the position of the exit of the introducing gas and on the flow rate was explored in the experiments done on June 14 and July 26, 1991. The data are shown in Figs. (a) and (b). The experiments were done at three different depths, 427cm, 214cm, and 105cm from the top of water level. The flow rate was varied from～0.5 to ～5 l/min. What were observed are:
(1) By the bubbling, the detection sensitivity increased as much as 4 or 5 times compared with the case of no bubbling.
(2) As the depth of exit of the bubbling gas increases, the ratio increased.
(3) The effect of radial direction at the same depth of the exit in the water seems to be unimportant. Also, the effect of on-or-off of the cooling pump did not change the efficiency of collecting the radioactivity.

We are attempting to continue the study on these effects more extensively.

Annual change of the FP concentration

In Fig. 3 is plotted the average concentration of these two radionuclides

Table 1. Radioactivity in the air and the effect of bubbling

Date determined	^{89}Rb (cpm)*	^{138}Cs (cpm)*	Condition of bubbling
			Depth(m) Flow rate(l/min)
April 16, 1990	11.0	12.5	(no Bubbling)
Dec. 5, 1990	3.0	5.9	(no Bubbling)
	20.8 (6.3)	17.0 (2.8)	4.5 ~0.2
Dec. 13, 1990	8.0	10.1	(no Bubbling)
	22.0 (2.7)	40.7 (4.1)	1.8 ~5
Jan. 23, 1991	11.5	6.8	(no Bubbling)
	23.7 (2.1)	25.8 (3.8)	1.8 ~10
Feb. 6, 1991	5.9	6.5	(no Bubbling)
	13.9 (2.4)	19.6 (3.0)	1.8 ~2
Average	7.9	8.4	(no Bubbling)
	20.1	25.5	

*The number shows the activities (normalized to the value just after the end of sampling) of the radionuclides collected on a filter paper, through which the air was sampled for 30 minutes by a dust sampler in the flow rate of 500 ml/min. The underlined value is for the case of nitrogen bubbling into the water. The value in the parentheses is the ratio compared with the case of no bubbling.

detected for the case of no bubbling as a function of calendar year, since 1986. It can be seen that the concentration detected in the air is almost constant in the order, 10^{-7} 10^{-6} Bq/cc.

OTHER INFORMATION AND CONSIDERATION

As to the origin of the FP found in the reactor coolant

It is important to investigate where is the origin of these fission products. What seems to be plausible at present is that these fission products come from the uranium which is contained as an impurity in the constructing materials in the core such as aluminum, not from the fuel. There are several reasons:

(1) Some FP, such as ^{131}I, are found in the cooling water of our reactor[1,2] and in Musashi Reactor,[5] which is the same type as ours. It is to be noted that in the latter reactor, where all the aluminum-clad fuel elements were replaced by stainless steel-clad ones, the FP can still be detected.[6]

(2) In JMTR Tokai, the FP (^{131}I) found in the primary cooling system was identified as from the uranium contained in the amount of as much as 42 ppm as an impurity in the beryllium reflector.[7]

(3) It has been reported that the "reactor grade" aluminum manufactured about thirty five years ago contains 1.7 ppm uranium as an impurity.[8]

Cases of fuel failure in TRIGA

Table 2 shows the cases of fuel failure which were reported in the literature.

This shows that most cases of the fuel failure occurred in the "pulsed" reactors. (These data have already been reported on the occasion of ref. 2.)

CONCLUDING REMARKS

To be sure its saftey, most research reactor facilities have the monitoring system for observing the state of the fuel elements, called CAM (continuous air monitor). The present study will be a useful information for constructing such a system, which has an improved efficiency in detecting FP. We hope to develop such a system, in which the routine monitoring for fuel element will be performed semi-automatically with high sensitivity.

From another point of view, as an active source term of the short lived fission products easily available during reactor operation, the present study may afford an interesting example to be performed in relation to the reactor safety, such as on the behavior of short lived FP in the water.

Acknowledgements

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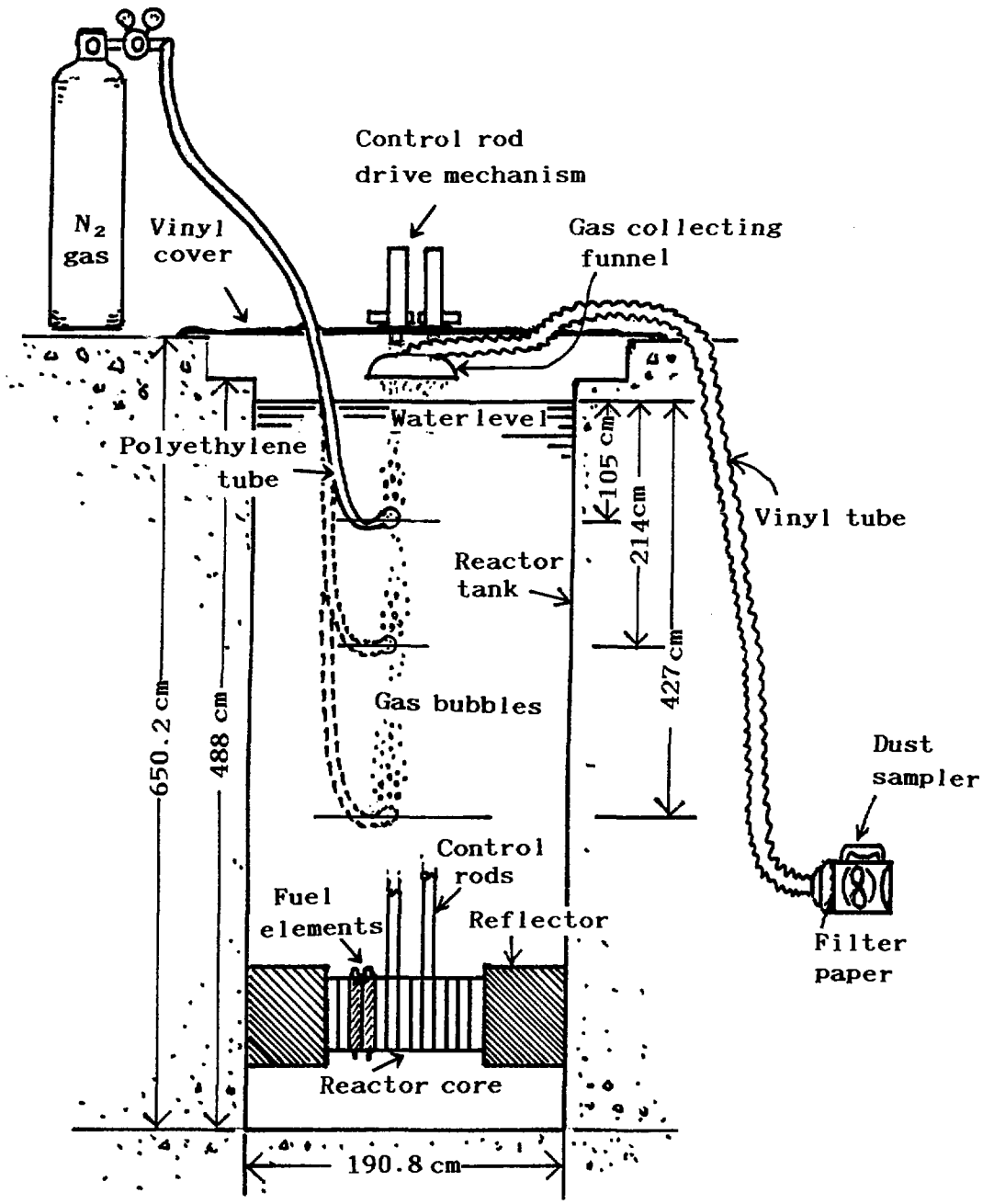

Fig.1 Schematic setup of dust sampling bubbled with N_2 gas. (Bubbling method)

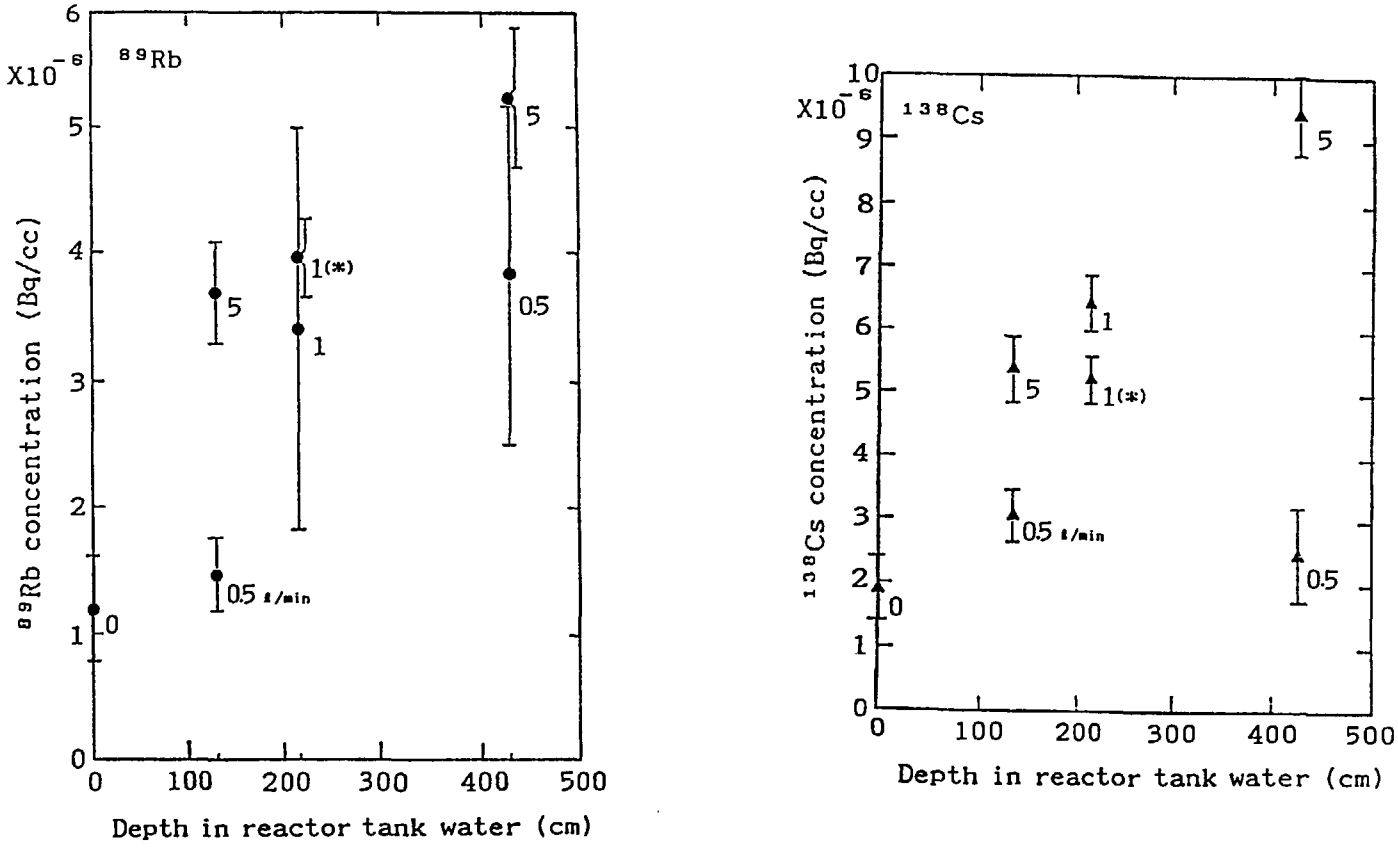

Fig. 2. Radioactive concentration in the air on the water surface of the reactor tank with and without N_2 gas bubbling. (Number shows a flow rate of the gas.) (a) ^{89}Rb, (b) ^{138}Cs.

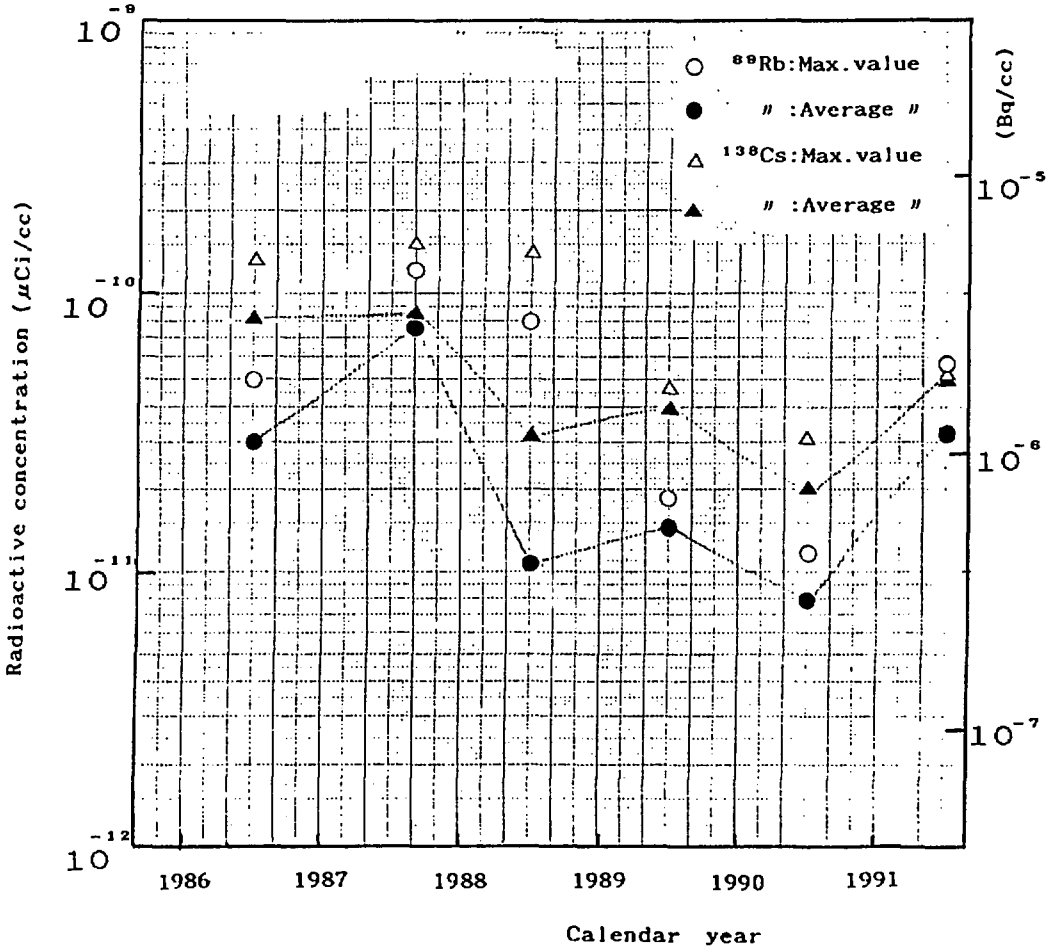

Fig. 3. Radioactive concentration of ^{89}Rb and ^{138}Cs in the air on the water surface of the reactor tank as a function of calendar year.

Table 2. Cases of fuel failure in TRIGA

	Location (Country) Type or Power (Date of Criticality)		Pulse (Peak Power)	Cladding	Date in Use	clad. (failed)	Discovery & Status	Date occurred	Ref.
1	Mainz(FRG) 250kW(1965-)		pulse	Al		Al	2 elongations 2 leak gas buble 2 swelling	1970	T- 721 (1970)
2	Pavia(Italy) 250kW(1965-)		pulse (250MW)	Al		Al	8 elongations/ 5 years (2 removed from core)	1970*	T- 721
3	Ljubliana(Yugoslavia)		steady	Al		Al	5 elongations (more than 2 cm)	1973*	TOC- 6 (1974)
4	München(FRG) 1 MW(1972-)		pulse	SS		SS	1 swelling (3cm) +Release of FP	1977	TOC-11 (1978)
5	Texas A & M (USA)		pulse	SS		SS	4 deformations	1982*	TOC-15 (1982)
6	Helsinki(Finland) 250kW(1967-)		pulse	Al		SS	1 crack+FP detected	1981	TOC-15
7	GA San Diego (USA)	250kW Mark I (1958-)	steady	23 SS 48 Al	1958-	SS	7 bent >1.6 mm	1982*	TOC-15
						Al	1 leaker 2 grew >1.8 cm 3 bent >1.6 mm		
		1.5MW Mark F (1960-)	pulse	100 Al 70-100 SS 65-100 FLIP	1960-1962 1962-1973 1973-	SS	54 bent >1.6 mm 1)		
						Al	35 bent 1) (including 1 leaker)		
		2 MW Mark III (1960-1973)	"	100 SS 100 FLIP	1960-1971 1971-1973	SS	1 leaker		
8	Vienna(Austria)		pulse	Al			1 swelling + gas bubling + FP detection 5 elongations or bents	Dec.1981	TOC-15
9	GA-ORR Oak-Ridge(USA) (Fuel test 1979-)		steady	SS (Incoloy 800)		SS (FLIP)	1 pin hole (/ 45 elements /4 year	1983	TOC-17 (1984)
10	Berkeley 1 MW(1966-1986)		pulse (2800MW)	SS		SS	1 FP detected (removed from core)	Oct.1986	TOC-18 (1986)

* reported date
1) Nearly all resulting from high power pulse development tests (up to $ 5.00)
2) Nearly all resulting from pulse development tests