Effects of Weathering on Impregnated Charcoal Removal Efficiency for Radioactive Methyl Iodide

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1. Introduction

Standby Gas Treatment System (SGTS) is one of the engineered safety features installed in BWR and ABWR plants to limit the discharge of radioactivity into the environment during the reactor accident.

KI_3 impregnated charcoal (iodized charcoal) filter is used for removing of radioactive iodine species such as molecular iodine and methyl iodide.

It is well known that the removal capability of impregnated charcoals for radioactive methyl iodide deteriorates by various pollutants and humidity in flowing air through impregnated charcoals beds. [1][2]

This paper describes the experimental results of the weathering effects on radioactive methyl iodide removal capability of iodized charcoals under the condition of Loss of Coolant Accident (LOCA), and the weathering evaluation of the actual plants under the normal condition.

2. Measuring and Calculation Method of Removal Efficiency

In BWR of Japan, the removal coefficient "μ" is used to evaluate the performance of charcoal filter.

The overview of the test method of the removal efficiency is shown in Figure.1. The test equipments consist of a methyl iodide generator, a humidifier and a adsorbent bed installed in a thermostated oven as shown in Figure.2. Figure.3 shows the construction of the bed and the stainless steel filter elements assembled into the bed. The filter elements have internal dimensions of 60mm diameter by 10mm long and both sides are covered with a stainless steel screen of 50mm in diameter.

The tests are conducted in the order of pre-flow step, methyl iodide injection step and after-flow step.

Pre-flow step: Air with controlled relative humidity is run through the bed until the adsorbents reaches equilibrium. The mass flow meter measures controlled flow rate of dry air. Relative humidity can be controlled in the range of 0 ~ 95 percent and temperature of the adsorbent bed in the thermostated oven can be controlled in the range of 20 ~ 90℃.

Methyl iodide injection step: After the pre-flow step, Nitrogen gas which contained methyl iodide tagged with I^{131} is being injected into humid air flow for one hour.

After-flow step: Following the methyl iodide injection step, humid air is passed through the bed at the same condition as in the pre-flow step for one hour.

After these steps, the piping and the bed were swept with dry air, and then the bed is disassembled into the filter elements to measure radioactivity collected in each filter element with a Ge (Li) detector.

Test Conditions are determined according to the LOCA condition of BWR as follow.

Temperature:	66℃
Relative Humidity:	70%
Face Velocity:	20cm/sec
Packing Density:	approximately Apparent Density
CH_3I-131+CH_3I-127 Concentration:	0.10mg/in^3
Bed Depth:	1cm × 10

It is well known that radioactive intensity of the filter elements is to be inversely exponential to bed depth. The typical profile of radioactive methyl iodide-131 along with bed depth was expressed in the following equation.

$$Qi \propto e^{-\mu Li} \qquad (1)$$

where

Qi: radioactive intensity of the ith filter element

Li: distance of the ith filter element from inlet of the bed

i: number of filter element (I=1〜10)

μ: removal coefficient

The removal efficiency is presented by the equation.

$$\eta = 1- e^{-\mu L} \qquad (2)$$

where

η: removal efficiency

L: bed length

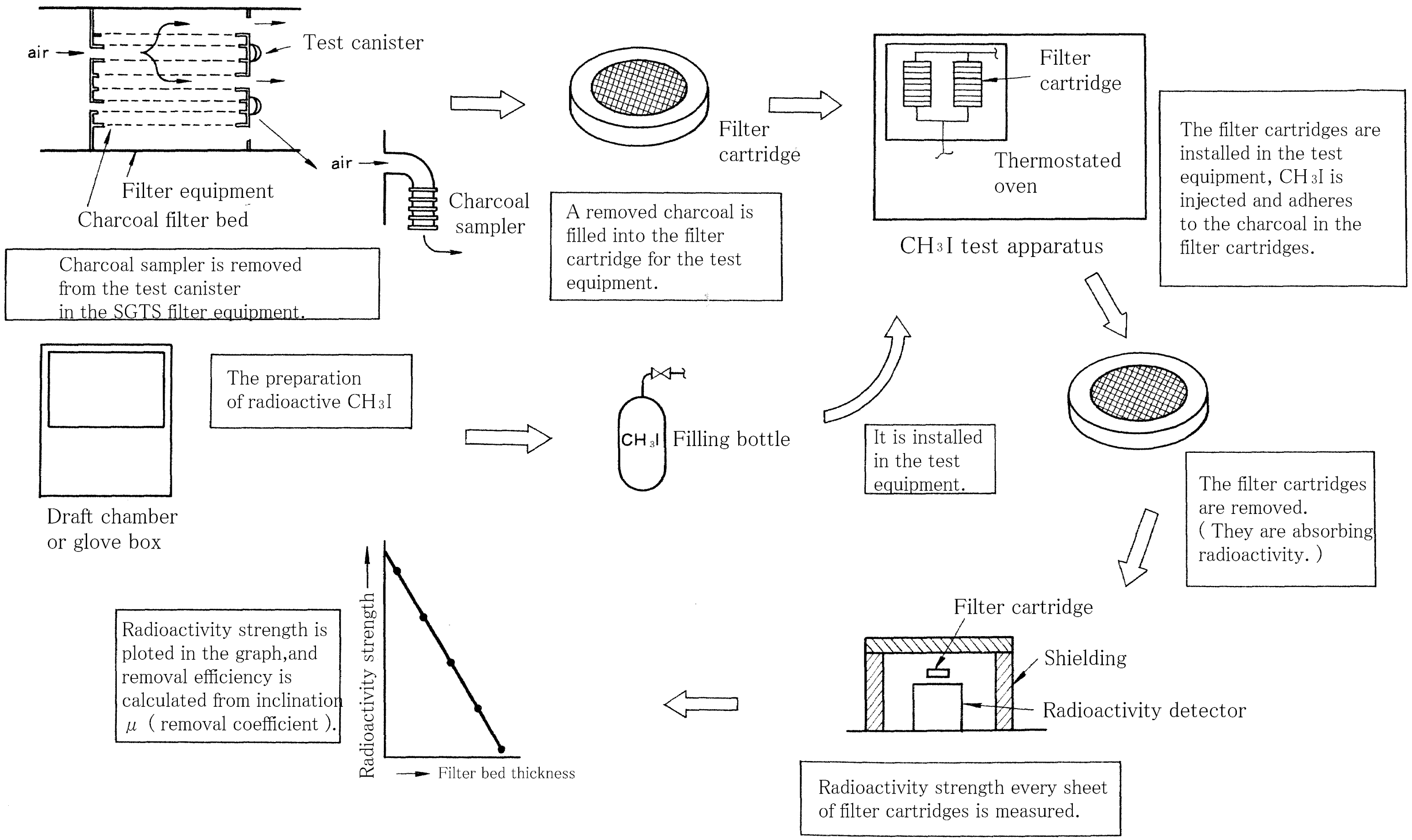


Fig.1 Measuring Method of Removal Efficiency

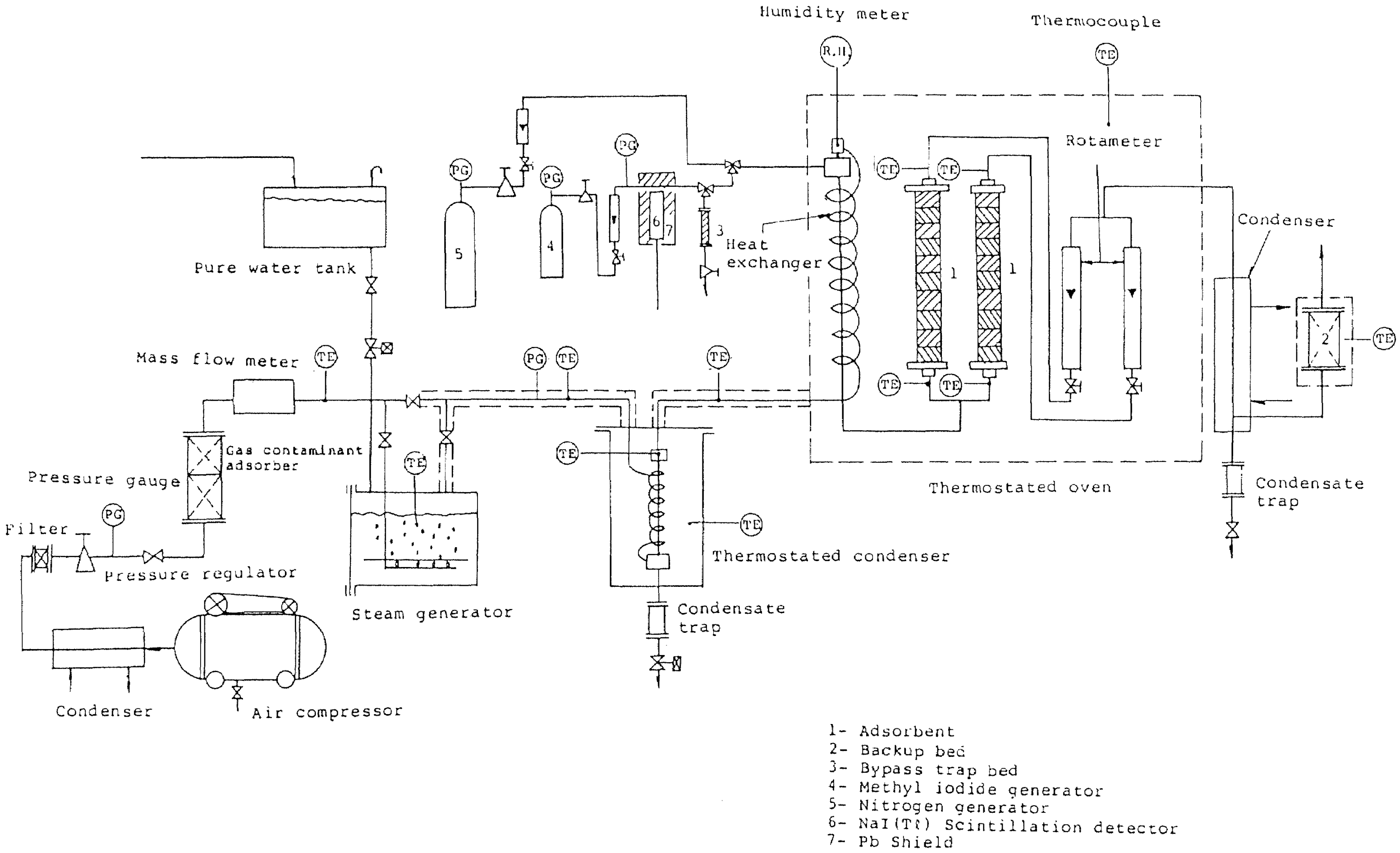
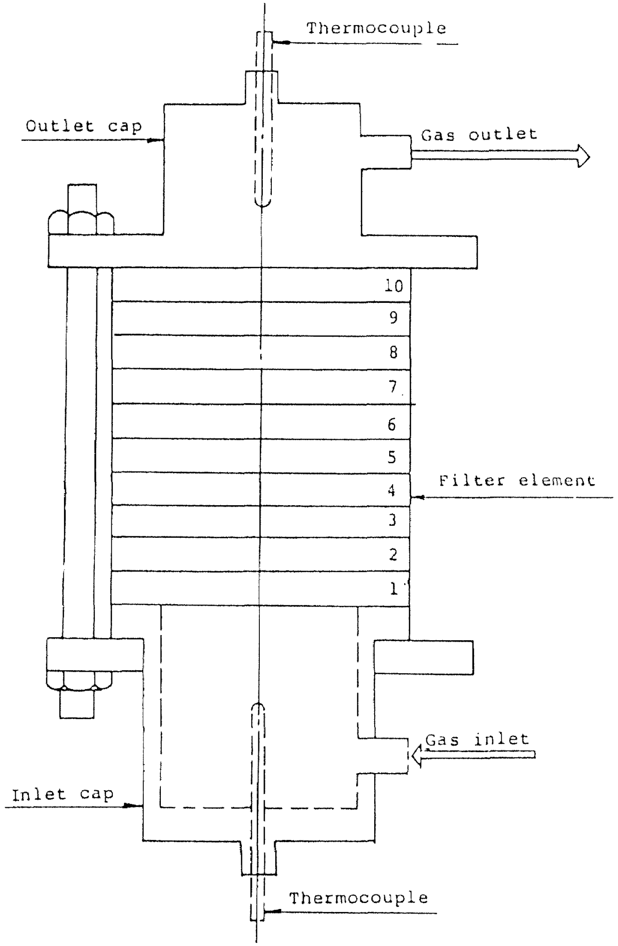
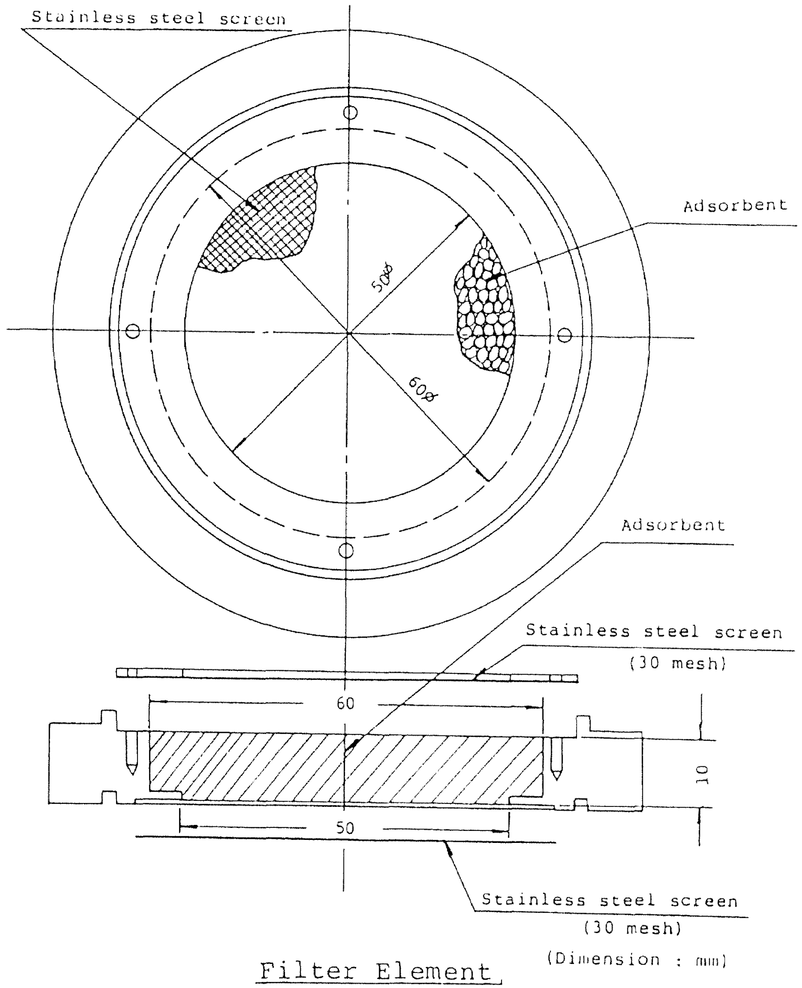


Fig.2 Schematic Diagram of CH3I Test Apparatus



Adsorbent Bed



Filter Element

Fig.3 Filter Element and Filter Bed Assembly

3. Weathering Experiment under the accident condition

3.1 Test Method

The test samples are BC-727 and SS 208 C 5K1$_3$ which are KI$_3$ impregnated charcoals. BC-727 and SS 208C 5K1$_3$ are both used for SGTS in BWR and ABWR of Japan. Impregnated charcoal to be tested was loaded in ten pieces of filter cartridge, dimensions of 5 cm inside diameter by 1cm long, and then which were assembled in a test bed.

Weathering experiments were performed with the weathering apparatus as shown in Figure. 4 under the simulant air condition during LOCA in BWR, which was given in Table.1. And then removal efficiency against radioactive methyl iodide of weathered samples was measured with the apparatus described in Section 2.

Table. 1 Conditions of Weathering Experiment

Weathering air	: indoor air
Contaminants of air	: NO_X< 0.01ppm, SO_X< 0.01ppm
Temperature	:66±3 ℃
Relative humidity	:70±15 %
Air velocity (superficial)	:20±2 cm/sec
Weathering term	:0～140 days

Table. 2 Values of Constant in the Experimental Equation (3)

Absorbent	Constant	
	μ (cm^{-1})	α (day^{-1})
BC-727 M-3034	1.39	4.02×10^{-3}
BC-727 M-3138	1.45	4.02×10^{-3}
SS 208C 5KI$_3$ 471/1	1.90	3.42×10^{-3}

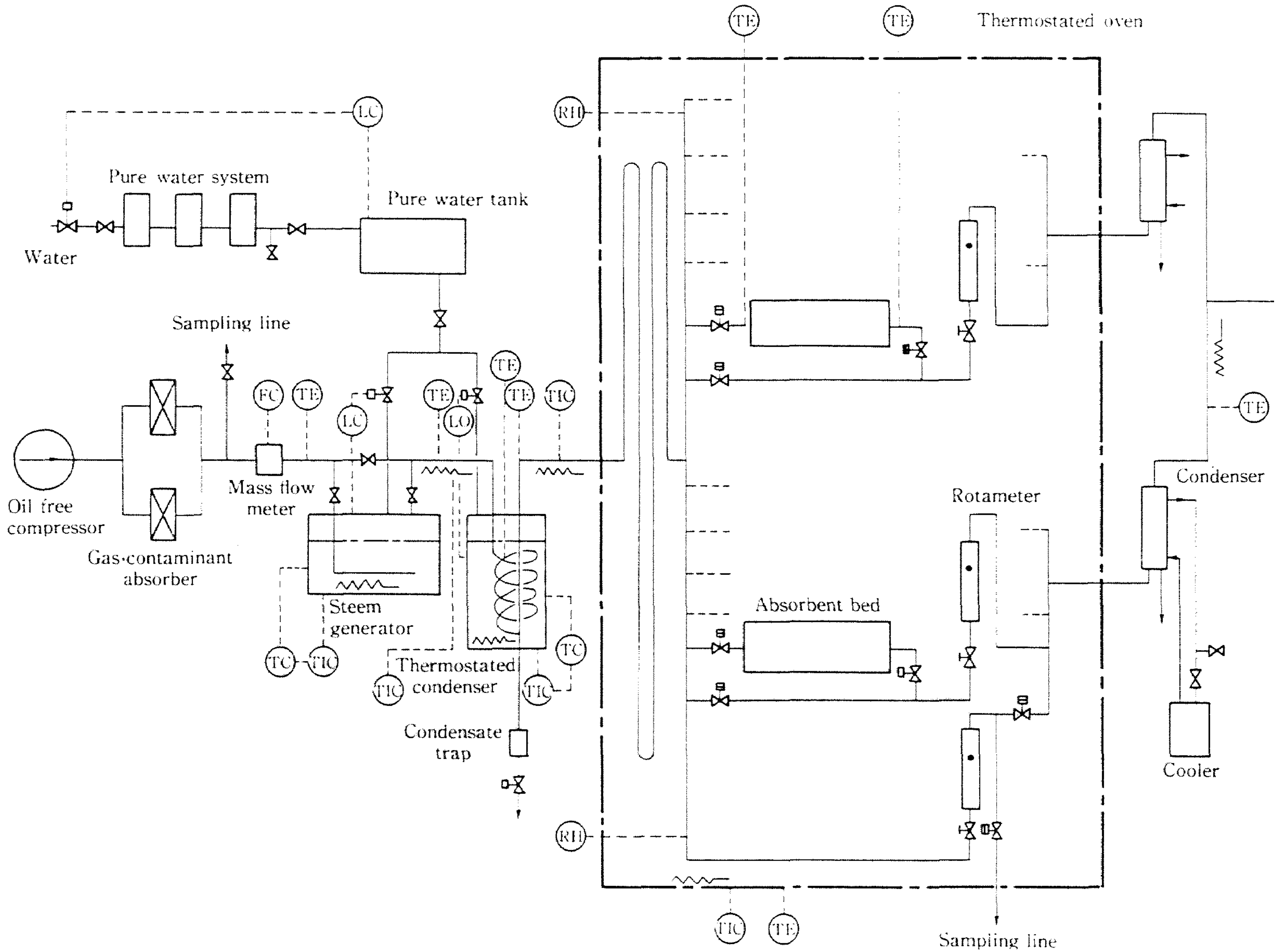


Fig.4 Schematic Diagram of Weathering Apparatus

3.2. Results and Discussion

The typical profile which shows the weathering effects on the removal efficiency of each one-centimeter layer is given in Figure. 5. From the experiment it was suggested that qualitatively

1) weathering deterioration increases with weathering period, and

2) there are two type mechanisms for deterioration in a filter bed, such as

Weathering Effect I: increasing deterioration uniformly over the bed length without relationship of depth from the filter bed entrance, and

Weathering Effect II : increasing deterioration from front to back of filter bed.

Figure. 6 illustrates change of removal coefficient and removal efficiency in five centimeters bed depth for radioactive methyl iodide. As shown in Fig. 6, the removal coefficient is similar to be inversely exponential to weathering term. This tendency was expressed with the following experimental equation and the values of constant in the equation for each weathered charcoals summarized in Table. 2.

$$\mu = \mu_0 \exp(-\alpha \cdot t) \qquad (3)$$

Where

μ : removal coefficient of weathered charcoals (cm^{-1})

μ_0:removal coefficient of new charcoals (cm^{-1})

α :constant (namely weathering coefficient) (day^{-1})

t :weathering period (day)

Table. 2 appears that BC-727 and SS 208 C 5 KI_3 have similar weathering slopes, and so it is seen that both charcoals would have the same weathering characteristics. Figure. 6 shows that the bed five centimeters thick has removal efficiency of 99% even under 80-day air flowing condition and the efficiency is greater than the designed value of the SGTS, 97%.

In order to examine the factor of weathering deterioration, the pollutants adhered to weathered samples were analyzed. They were extracted with carbon tetrachloride and methyl alcohol and were analyzed with the gas chromatography and the infra-red spectroscopy.

The analytical results show that a large quantity of organic pollutants has been adsorbed by entrance layer in comparison with the other layers. From this result, it was found that the important factor of Weathering Effect II was the atmospheric pollutants, which had been adsorbed by charcoals. However, The factor of Weathering Effect I couldn’t be clarified in this study.

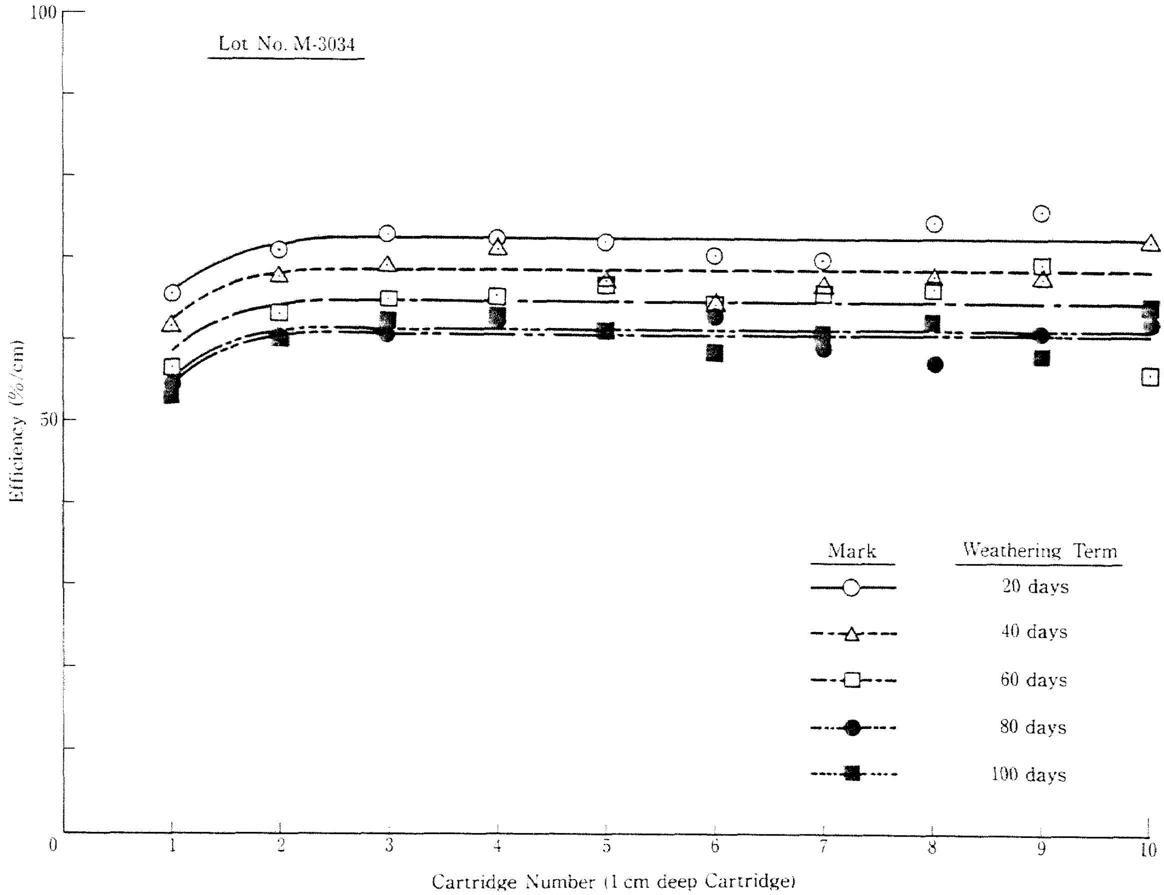


Fig.5 Experimental Depth Profile of Removal Efficiency of $CH_3{}^{131}I$ with BC-727 in Ten One-Centimeter Layers

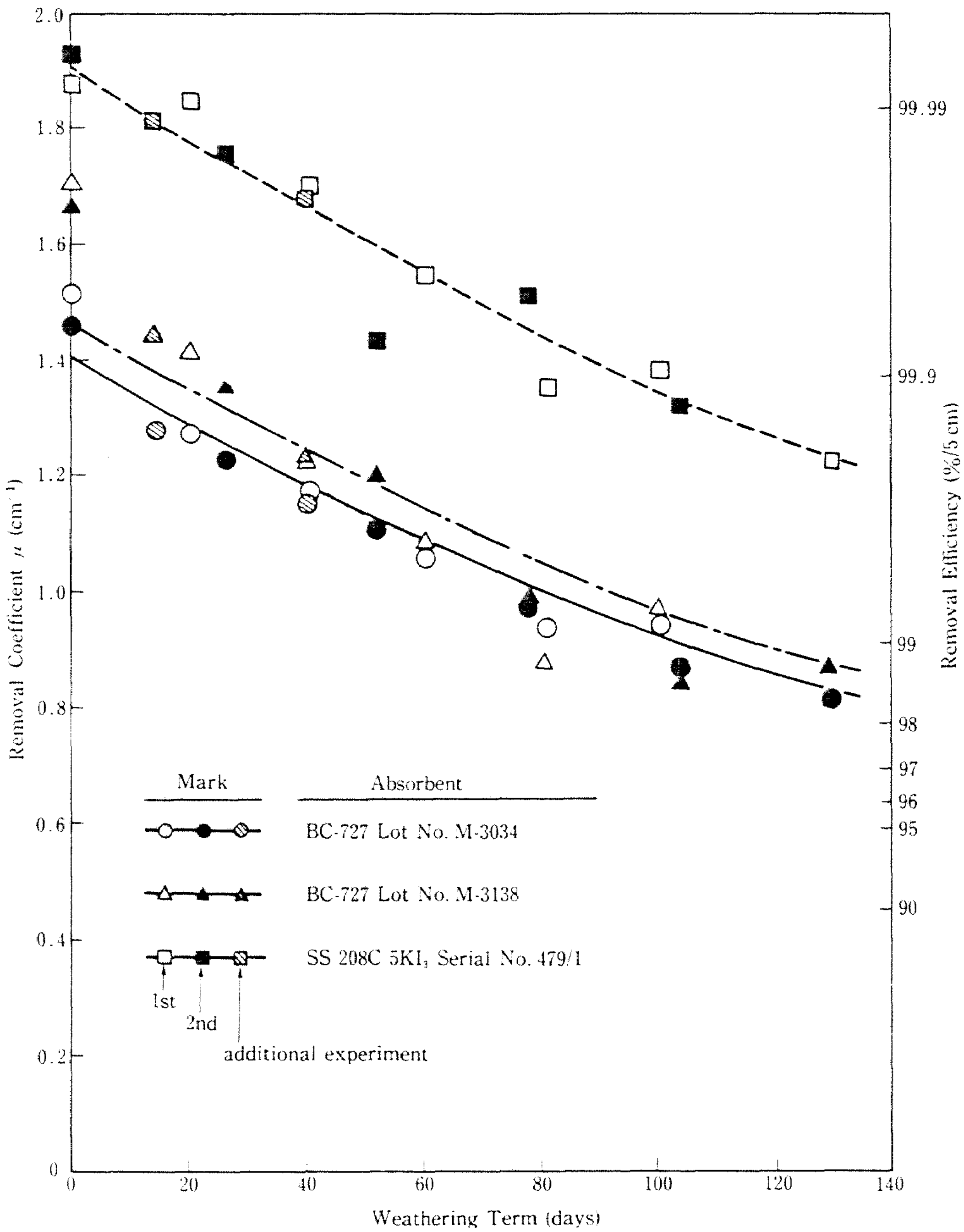


Fig.6 Effect of Weathering on $CH_3{}^{131}I$ Removal Capability of Iodized Charcoals
(Experiment under Accident Condition)

4. Weathering evaluation of the actual plants under the normal condition

The charcoal filter of SGTS is kept in a filter unit heated with space heaters under the standby condition, while SGTS is operated at least once per month for surveillance test, and once per year for the periodic inspection of SGTS itself and the leak rate test of the secondary containment. Weathering effect of the normal condition has to be considered for both standby and operating conditions. Normal condition of actual plants is shown in Table. 3.

The degradation trend can be obtained from plotting the removal coefficient of every year's laboratory test result of SGTS filters at periodic inspections. The laboratory test method is the same as written in Section 2. The example of the degradation trend of removal coefficient is shown in Figure. 7. From accumulating data, the renewal timing of the charcoal filter is estimated. This tendency is expressed in the same equation of Section 3.2. But weathering coefficient (α) becomes bigger than coefficient written in Table.2, when the weathering period (t) is substituted with the actual operating time of SGTS. Therefore it is considered that the weathering effect of the periodic operation is different from the continuous operation.

The examples of indoor atmosphere analyzed in some plants are shown in Table4. Because the low volatility organic compounds are considered major pollutants for the charcoal filter in the indoor atmosphere of the nuclear power plant, limiting of painting work before the scheduled operation of SGTS is recommend to user.

Table. 3　Normal Condition of Actual Plants

Operating condition	
Temperature	:30～50 ℃
Relative humidity	: ～50 %
Air velocity (superficial)	:< 20 cm/sec

Standby condition	
Temperature	:70～90 ℃
Relative humidity	: ～30 %
Air velocity (superficial)	: －

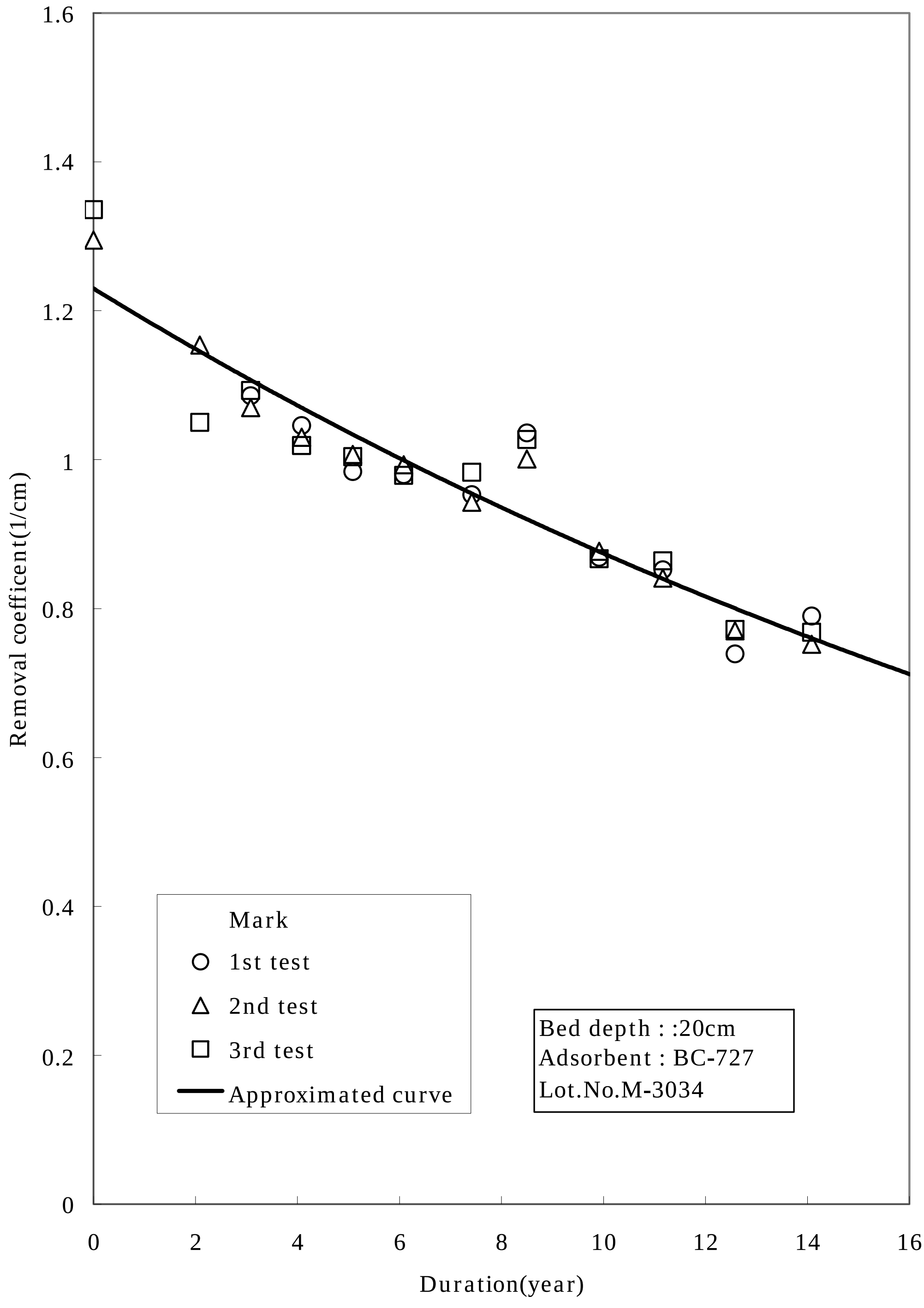


Fig. 7 Effect of Weathering on $CH_3{}^{131}I$ Removal Capability of Iodized Charcoals
(Sample of Actual Plant under Normal Condition)

Table. 4 Pollutants detected from the atmosphere in plants

<Sample of A plant's data>

Impurities	boiling point (℃)	Concentration (ppb)	Assumed cause
acetaldehyde	20	4.6	fungicide, solvent
acetone	56	6.3	wax, lacquer
methanol	65	23	material of formalin, solvent
methyl ethyl ketone	80	43	
toluene	111	860	paint
1-butanol	117	36	paint
phenol	182	63	

< Sample of B plant's data >

Impurities	boiling point (℃)	Concentration (ppb)	Assumed cause
acetonitrile	-46	1.5	material of synthetic fiber
chlorodifluorometane (R-22)	-41	3.3	refrigerant
iso-butane	-12	1.1	air spray
n-butane	-0.5	3	air spray
acetaldehyde	20	2.1	fungicide, solvent
acetone	56	4.6	wax, lacquer
methanol	65	15	material of formalin, solvent
tetrahydrofuran	66	2.5	
ethanol	78	2.7	solvent, varnish
methyl ethyl ketone	80	1	
n-heptane	98	3.8	bond, thinner, solvent
toluene	111	3.4	paint
1-butanol	117	30	paint

5. Conclusion

To evaluate the performance of the SGTS impregnated charcoal filter, the influence on weathering was evaluated for both accident condition and normal condition.

As for the weathering experiment of accident condition, it found the following.

Air stimulant under the condition of temperature and relative humidity equivalent to atmosphere during LOCA was continuously passed through KI_3 impregnated charcoals for a long term, and the removal efficiency of charcoals for radioactive methyl iodide was measured. The summarized results are as follows.

(1) The KI_3 impregnated charcoal beds keep removal efficiency of 99%/ 5cm even under 80-day air flowing condition. The efficiency is greater than the designed value of the SGTS, 97%.

(2) A fall of removal efficiency is classified into next two groups.

- Uniform deterioration which progresses slowly extend over the all bed length, and
- Large deterioration which is brought by organic pollutants in air progresses from the upper stream to the lower stream in the bed.

As for the weathering effect of the normal condition, the periodical inspection data of operating plants were evaluated. SGTS is operated for surveillance test and periodical inspections under the normal condition. The tendency is expressed in the same equation of the weathering experiment. But the weathering coefficient of the periodic operation is different from that of the continuous operation. Because the low volatility organic compounds are considered major pollutants for the charcoal filter in the indoor atmosphere of the nuclear power plant, limiting such as painting work before the scheduled operation of SGTS is recommended.

References

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