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# DEVELOPMENT OF EQUIPMENT FOR DISPOSAL OF CONTAMINATED SODIUM(II)

May, 1977

POWER REACTOR AND NUCLEAR FUEL DEVELOPMENT CORPORATION



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Development of Equipment  
for Disposal of Contaminated Sodium (II)\*

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Abstract

The transport behavior of trace-amount of radio nuclides such as  $^{131}\text{I}$  and  $^{137}\text{Cs}$  ( $^{134}\text{Cs}$ ) in sodium have been studied on both conditions of normal sodium-steam reaction and sodium burning brought by excess steam supplying.

Trapping of these radio nuclides transported into gas phase in gas bubbler or scrubber, which is a component of waste gas treatment system, was also discussed.

Main experimental results were as follows:

- 1) Under the normal sodium-steam reaction, partition ratio of  $^{134}\text{Cs}$  transported into gas phase was  $4 \times 10^{-3}$ , while those of  $^{131}\text{I}$  and  $^{24}\text{Na}$  were one hundred times smaller than that of  $^{134}\text{Cs}$ .

Chemical forms of  $^{134}\text{Cs}$  and  $^{131}\text{I}$  transported into gas phase were estimated to be cesium hydroxide and inorganic iodine such as  $\text{NaI}$ , respectively.

- 2) When the sodium burning occurred by excess steam supplying, partition ratios of  $^{24}\text{Na}$ ,  $^{131}\text{I}$  increased 250 ~ 500 times larger than those under the normal sodium-steam reaction, while for  $^{134}\text{Cs}$ , it increased about 25 times larger than that under normal reaction.

Small amount of volatile organic form of iodine was estimated to be produced and transported into gas phase during this treatment.

- 3) Decontamination factors of  $^{131}\text{I}$  and  $^{134}\text{Cs}$  transported into gas phase both for gas bubbler and scrubber were about  $10^3$ .

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\*\* Mitsubishi Atomic Power Industries, Inc., Engineering and Research Division

## Contents

1.	Introduction .....	1
2.	Experimentals .....	2
2-1	Test apparatus .....	2
2-2	Sodium Sample .....	2
2-3	Test Condition .....	6
3.	Results .....	8
3-1	Radio Nuclides Transported into Gas Phase .....	8
3-2	Removal Test of Radioactive $^{131}\text{I}$ and $^{134}\text{Cs}$ in Waste Gas .....	12
4.	Discussion .....	23
4-1	Transport of F.P. in Sodium into Gas Phase during Sodium-Steam Reaction .....	23
4-2	Decontamination Factor of Gas Bubbler and Scrubber .	25
4-3	Decontamination Factor of Charcoal Filter and HEPA Filter .....	25
4-4	Decontamination of Waste Gas in Waste Gas Treatment System .....	28
5.	Conclusion .....	30
6.	Reference .....	32

## Figures & Tables

Fig. 1	Flow Sheet of Test Apparatus
Fig. 2	Experimental Apparatus of Evaporation to Dryness for Na <sup>131</sup> I Solution
Fig. 3	MAYPACK Sampler and Bulking Materials
Fig. 4	Schematic Diagram of Sampling System for Measurement of <sup>131</sup> I and <sup>134</sup> Cs Transported into Gas Phase
Fig. 5	Reaction Rate and Sodium Temperature in Static Nitrogen-Moist Atmosphere
Table 1	Properties of <sup>131</sup> I and <sup>134</sup> Cs
Table 2	Sampling Condition for <sup>131</sup> I Transported into Gas Phase
Table 3	Sampling Condition for <sup>134</sup> Cs Transported into Gas Phase
Table 4	Partition Ratios for <sup>131</sup> I, <sup>134</sup> Cs and <sup>24</sup> Na Transported into Gas Phase
Table 5	Radioactivities Transported into Gas Phase during Nitrogen-Moist Treatment of Sodium
Table 6	Activities Change of <sup>131</sup> I and <sup>134</sup> Cs in the Gas Phase by Sedimentation
Table 7	Distribution of Trapped <sup>131</sup> I in MAYPACK's Bulking Materials
Table 8	Results on the Qualitative Analysis Iodine Compounds Transported into Gas Phase
Table 9	Radioactivity Distribution on Waste Gas Treatment System Components
Table 10	Presumption of Decontamination Factor of Gas Bubbler and Scrubber for <sup>131</sup> I and <sup>134</sup> Cs
Table 11	Preliminary Consideration of Decontamination Factor on Waste Gas Treatment System Component

## 1. Introduction

In the previous experiment for the development of equipment for disposal of contaminated sodium (Part 1), a series of sodium-steam reaction tests in static nitrogen atmosphere had been carried out as the function of sodium temperature and steam concentration. From the result, the reactionability between sodium and steam in static nitrogen atmosphere had been clarified, and  $^{24}\text{Na}$  transported into nitrogen atmosphere during sodium-steam reaction had also been evaluated.

The purpose of this experimental study is to clarify of the transport behavior of trace amount of  $^{131}\text{I}$  and  $^{134}\text{Cs}$  ( $^{137}\text{Cs}$ ) in sodium on both conditions of normal sodium-steam reaction and sodium burning brought by excess steam supplying into nitrogen atmosphere.

And, removal of these radio nuclide transported into gas phase by gas bubbler or scrubber was also discussed.

## 2. Experimentals

### 2-1 Test apparatus

Test apparatus is composed of boiler, gas mixer, sodium treatment vessel, gas bubbler, scrubber and charcoal particle filter.

Sodium specimen was set in the center of sodium treatment vessel and atmosphere of this vessel was high purity nitrogen.

Steam was produced in boiler and sodium-steam reaction was started by introducing of moist nitrogen through the gas mixer.

Reactivity between sodium and steam was measured by the analysis of hydrogen gas content in the vessel and direct visual observation.

The flow sheet of the test apparatus was shown in Fig. 1.

### 2-2 Sodium Sample

The reactor grade sodium was used for this experiment and  $^{131}\text{I}$  was put into sodium as  $\text{Na}^{131}\text{I}$  which was diluted by  $\text{NaI}$  solution (pH9) containing 100mg- $\text{NaI}/\text{l}$ . This  $\text{NaI}-\text{Na}^{131}\text{I}$  solution was sampled out into a glass ampoule and evaporated to dryness at  $90^\circ\text{C}$  in apparatus as shown in Fig. 2.

$^{131}\text{I}$  remaining more than 99.99 % on the inner wall of glass ampoule was added into 10g of molten sodium by crushing the glass ampoule and thermal diffusion at  $200^\circ\text{C}$  for 1 hour.

The radioactivity of  $^{131}\text{I}$  in this experiment was 0.1mCi/10g-Na.

On the other hand,  $^{134}\text{Cs}$  was produced by irradiation of quartz ampouled cesium metal (purity 99.99 %) at the neutron flux of  $5 \times 10^4$  N/sec.cm<sup>2</sup> for 3 hrs.

$^{134}\text{Cs}$  was added into sodium by the same manner as that of  $^{131}\text{I}$ .

The radioactivities of  $^{134}\text{Cs}$  in this experiment was 0.15mCi/10g-Na.



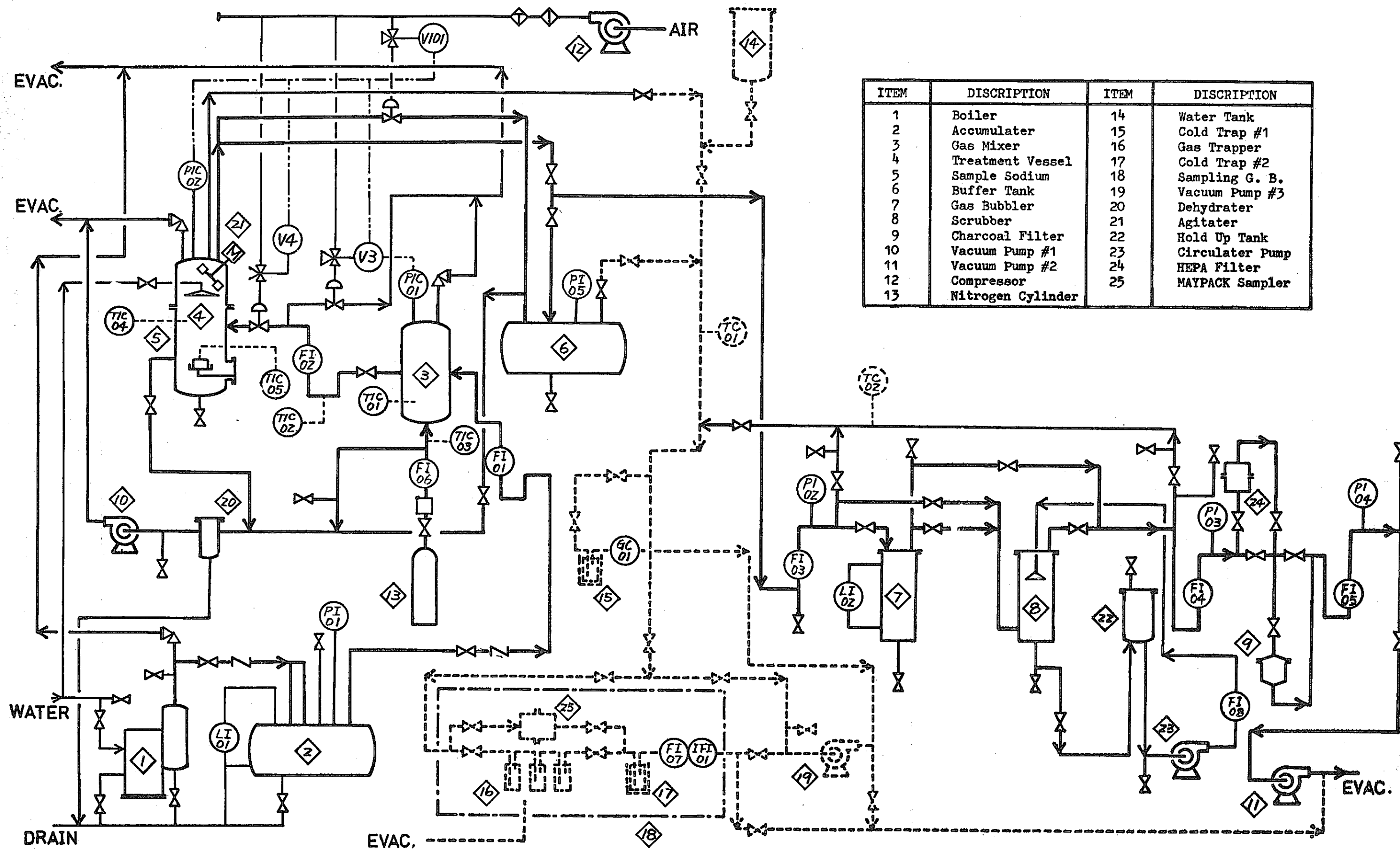


Fig. 1 Flow Sheet of Test Apparatus



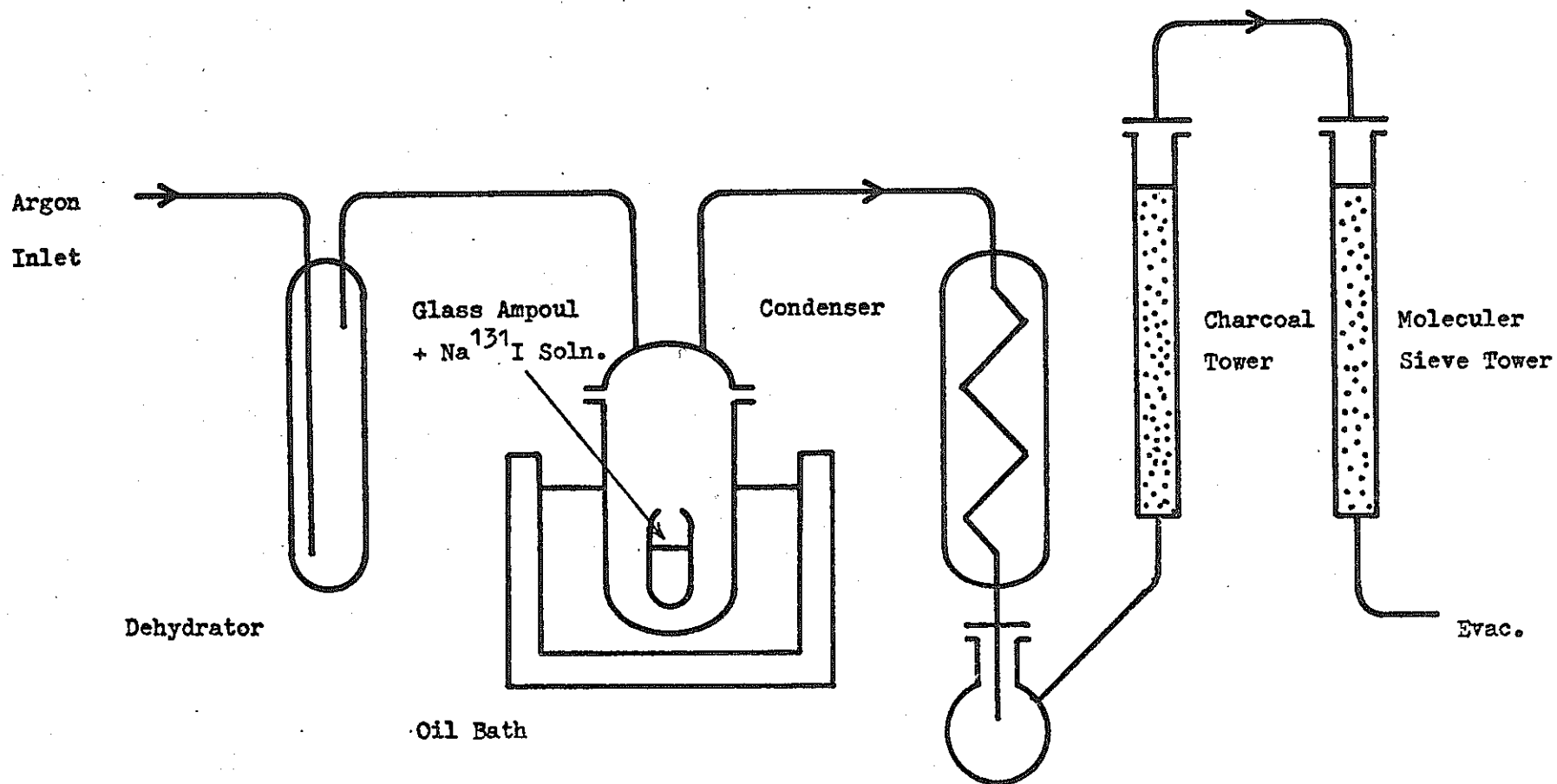


Fig. 2 Experimental Apparatus of Evaporation to Dryness for Na<sup>131</sup>I Solution

Nuclide	Half Life	Decay Pattern	Energy of $\beta$ -ray and Release Ratio
$^{131}\text{I}$	8.05 d	$\beta^-$	0.080 - 2.6 % 0.284 - 5.4 % 0.364 - 82 % 0.637 - 6.8 % 0.723 - 1.6 %
$^{134}\text{Cs}$	2.046 y	$\beta^-$	0.57 - 23 % 0.605 - 98 % 0.796 - 99 % 1.038 - 1.0 % 1.168 - 1.9 % 1.365 - 3.4 %

Table 1 Properties of  $^{131}\text{I}$  and  $^{134}\text{Cs}$

The properties of  $^{131}\text{I}$  and  $^{134}\text{Cs}$  were shown in Table 1.

## 2-3 Test Condition

### 2-3-1 Measurement of $^{131}\text{I}$ and $^{134}\text{Cs}$ transported into Gas Phase

In this experiment, tests were made under both conditions of the normal sodium-steam reaction and sodium burning in order to discuss the transport behavior of  $^{131}\text{I}$  and  $^{134}\text{Cs}$  into gas phase during reaction.

The test conditions were as follows.

#### a) Normal Reaction Test

Steam concentration:	50 %
Steam temperature:	100 °C
Initial sodium temperature:	80 °C
Initial pressure and atmosphere in the vessel:	0 kg/cm <sup>2</sup> -G, N <sub>2</sub> Gas
Wall temperature of the vessel:	as same as steam condition
Sodium:	10g
Radioactivities:	$^{131}\text{I}$ 0.1mCi/10g-Na $^{134}\text{Cs}$ 0.15mCi/10g-Na

#### b) Sodium Burning Test

Steam concentration:	50 %
Steam temperature:	100 °C
Initial sodium temperature:	80 °C
Initial pressure and atmosphere in the vessel:	0 kg/cm <sup>2</sup> G, N <sub>2</sub> Gas
Wall temperature of the vessel:	as same as steam condition
Sodium:	10g
Radioactivities:	$^{131}\text{I}$ 0.1mCi/10g-Na $^{134}\text{Cs}$ 0.15mCi/10g-Na

## 2-3-2 Removal of Radioactive $^{131}\text{I}$ and $^{134}\text{Cs}$ in Waste Gas

This test was performed for waste gas generated under the condition of sodium burning.

The test conditions were as follows.

Waste gas flow rate:	20 l/min.
Initial pressure of waste gas:	1.0 ~ 1.5 kg/cm <sup>2</sup> -G
Nitrogen carrier gas flow rate:	20 l/min.
Nitrogen carrier gas feeding volume:	1000 l
Solution filled with gas bubbler and scrubber:	1/10N NaOH solution for $^{131}\text{I}$ . 1/10N H <sub>2</sub> SO <sub>4</sub> solution for $^{134}\text{Cs}$ .
Circulation rate of solution in scrubber:	10 l/min.
Charcoal in the charcoal filter:	0.5 l (6 ~ 10 mesh)
Filter material of HEPA filter:	Air filter paper (TOYO Filter-paper Corp., Cat. No. HE-50)



### 3. Results

#### 3-1 Radio Nuclides Transported into Gas Phase

MAYPACK sampler filled with silver particle cartridge (silver particle size: 2 ~ 3 mm $\phi$ , particle layer thickness: 8 mm), air filter paper (TOYO Filter Paper Corp., Cat. No. HE-50), activated charcoal filter paper (TOYO Filter Paper Corp., Cat. No. CP-20) and activated charcoal particle cartridge (TOYO Filter Paper Corp., Cat. No. CHO-50), charcoal layer thickness: 20 mm) was used for the sampling of  $^{131}\text{I}$  in waste gas. On the other hand, sampling trap (three stages) filled with 5 ml of 1/10 N  $\text{H}_2\text{SO}_4$  solution was used for  $^{134}\text{Cs}$ .

The sample gas was introduced into and passed through these samplers. The flow diagram of sampling systems were shown in Fig. 3 and Fig. 4. After completing the sampling of gas, inner wall surface of sampling pipings were washed with pure water of constant quantity and then the radioactivity in wash-water was measured and evaluated as the radioactivity transported into gas phase.

The counting time of radioactivity for each sampling elements was set at 10 ~ 40 min for Na(I) scintillation counter, and 1,000 sec. at the maximum for Ge(Li)  $\gamma$ -ray spectrometer.

Radioactivity was determined by comparing whether these measured values were larger or smaller than the tripled values of the standard deviation ( $3\sigma$ ) of B.G.

The typical experimental results of reaction rate and the sodium temperature change during normal sodium-steam reaction are shown in Fig. 5.

The maximum temperature of sodium attained during reaction was about 240  $^{\circ}\text{C}$ , and the total reaction time was 1.5 ~ 2 hours.

All of which approximately matched those of the radioactivity

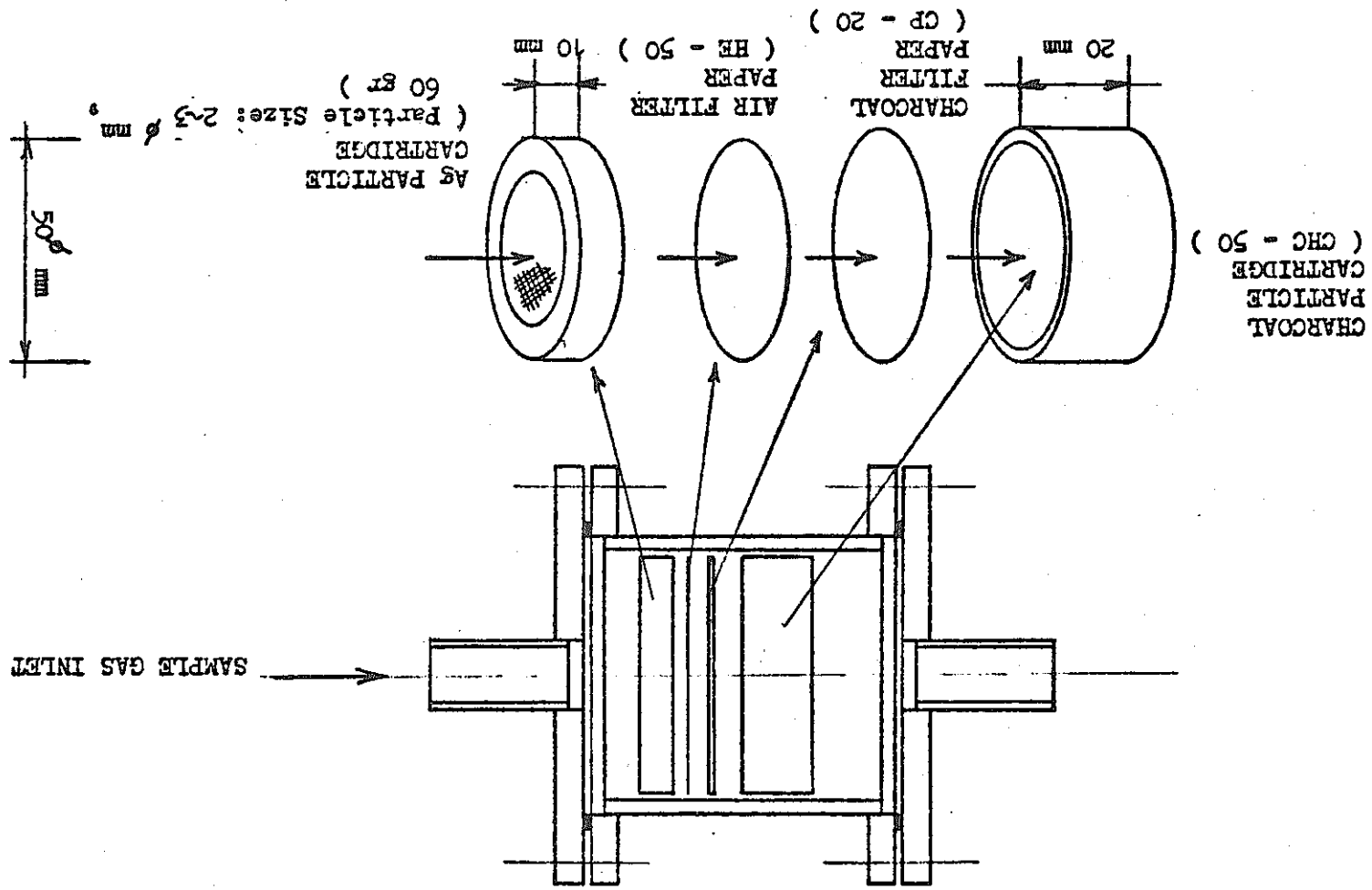


Fig. 3 MAYPACK Sampler and Bulking Materials

transport measuring test for  $^{24}\text{Na}$  performed previously.

When the steam concentration in sodium treatment vessel reached about 60 ~ 70 %, sodium ignited in only a few minutes after introduction of steam, and it continued to burn for about 1 ~ 3 minutes at the estimated temperature of 600 ~ 700 °C accompanying violent whitish smoke belching out. The fire was completely extinct after 1 ~ 3 minutes, while the smoke remained in the atmosphere for about 1.5 hours before it completely settled down.

The possibility of sodium ignition was extremely large if the introduced steam concentration was more than 50 %.

It was, however, difficult to determine whether the cause of such sodium ignition was occurred by the direct reaction between the condensed water of steam and sodium, or due to only the increase of steam concentration in the atmosphere.

Analytical results of  $^{131}\text{I}$  and  $^{134}\text{Cs}$  transported into gas phase under both of normal sodium-steam reaction and sodium burning, and also the results of  $^{24}\text{Na}$  performed previously are shown in Table 4.

It is apparently from these results that both  $^{131}\text{I}$  and  $^{134}\text{Cs}$  transport into gas phase even under normal reaction, and transport ratio of  $^{131}\text{I}$  was in the order of  $\times 10^{-5}$  and of  $^{134}\text{Cs}$  was  $4.0 \times 10^3$ .  $^{134}\text{Cs}$  transports about 1/250 of the initial amounts in sodium and it is larger than the case of  $^{24}\text{Na}$ , but the transport ratio of  $^{131}\text{I}$  is comparatively low.

When sodium ignited, transport ratios of  $^{24}\text{Na}$ ,  $^{131}\text{I}$  and  $^{134}\text{Cs}$  into gas phase increase and such increase of  $^{131}\text{I}$  was about 200 times larger than the case of normal reaction, while  $^{134}\text{Cs}$  was about 25 times larger.

Especially,  $^{134}\text{Cs}$  showed a larger transportation than  $^{24}\text{Na}$ , and it was about 10% of the initial amount in sodium.

As described in the earlier page, even in an inert gas atmosphere (oxygen content: below 1 %), sodium burns at the estimated temperature

in the range of 600 ~ 700 °C violently belching white smoke. Because of this, it is assumable that such increase of transport ratio is occurred by release of aerosols which are generated by sodium combustion, and also the evaporation of waste solution due to the temperature rise in reaction zone.

Even in this case, the transport ratio of  $^{134}\text{Cs}$  was the largest.

The results are shown in Table 5.

An instance of variations of concentration of  $^{131}\text{I}$  and  $^{134}\text{Cs}$  in the atmosphere of sodium treatment vessel caused by sedimentation of these nuclides is shown in Table 6.

The sedimentation rate of  $^{134}\text{Cs}$  is extremely large and about 90 % of transported  $^{134}\text{Cs}$  has sedimented after one hour. This decrease of  $^{134}\text{Cs}$  was consistent with visual observation that the white smoke (aerosol) which had been generated by sodium burning settled down and disappeared about 1.5 hours. It was, therefore, considered that  $^{134}\text{Cs}$  estimated to be either transported with Na aerosol or behaved like Na aerosol under sodium burning.

However, the sedimentation rate of  $^{131}\text{I}$  was so slow comparing with that of  $^{134}\text{Cs}$  and after one hour  $^{131}\text{I}$  decreased no more than 1/100 of initial transported amount. Even after Na aerosol had settled down and disappeared,  $^{131}\text{I}$  still remained in gas phase.

An instance of distributions of  $^{131}\text{I}$  on various filter elements in MAYPACK sampler is shown in Table 7.

The combination of filter elements of this MAYPACK sampler is made to separate  $^{131}\text{I}$  by its chemical form (molecular, inorganic and organic compound). But, the direct sampling of atmospheric gas caused the condensation of steam and incursion of condensed water in MAYPACK sampler, because the atmosphere of sodium treatment vessel had so high steam concentration. It is, therefore, questionable to determine the



absorption distribution of  $^{131}\text{I}$  in MAYPACK sampler as shown in Table directly to the existing ratio of iodine compound contained in waste gas. But, the existence of organic iodine compound has been estimated qualitatively.

Whereas, in order to confirm the chemical form transported iodines qualitatively, three sampling traps, as shown in Fig. 4, with 5 m of 1/10 N NaOH solution, xylene and carbontetrachloride were prepared respectively and the constant quantity of waste gas was passed through these traps.

An instance of analytical results of above mentioned solutions is shown in Table 8.

In this test, the existence of  $^{131}\text{I}$  compound which is captured by xylene and carbontetrachloride after passing through NaOH solution has been certain. Xylene and carbontetrachloride are known as the typical trapper of organic iodine compound.

So, it was considered that  $^{131}\text{I}$  compound trapped by these solution estimated to be had possibility as organic iodine compound.

### 3-2 Removal Test of Radioactive $^{131}\text{I}$ and $^{134}\text{Cs}$ in Waste Gas

The removal test of  $^{131}\text{I}$  and  $^{134}\text{Cs}$  in waste gas was only for the waste gas produced by sodium burning since the transport ratio of these nuclides into gas phase under normal reaction was so low. The capture distributions of  $^{131}\text{I}$  and  $^{134}\text{Cs}$  in the components of the waste gas treatment system is shown in Table 9.

Adsorption of  $^{131}\text{I}$  and  $^{134}\text{Cs}$  onto the inner wall surface of the piping during the transfer of waste gas to gas bubbler and scrubber was so large that such adsorption reached about 23 % for  $^{131}\text{I}$  and as much as 55 % in the case of  $^{134}\text{Cs}$  of introduced amount. Such a large adsorption

onto the wall surface of pipings was considered to have been caused by condensation of steam and also by dilution of transported  $^{131}\text{I}$  and  $^{134}\text{Cs}$ .

On the other hand,  $^{131}\text{I}$  and  $^{134}\text{Cs}$  were not detected in solution of gas bubbler and scrubber. The reason for this was assumable that the introduced radioactivity was too small for the volume of both components. The values computed from detection limit values of Na(I) scintillation counter for these nuclides were listed as maximum estimated capture values.

But,  $^{131}\text{I}$  was detected in backup charcoal filter.

$^{134}\text{Cs}$  was not also detected in HEPA Filter. It is considered that  $^{134}\text{Cs}$  was removed by gas bubbler and not able to reach HEPA filter.

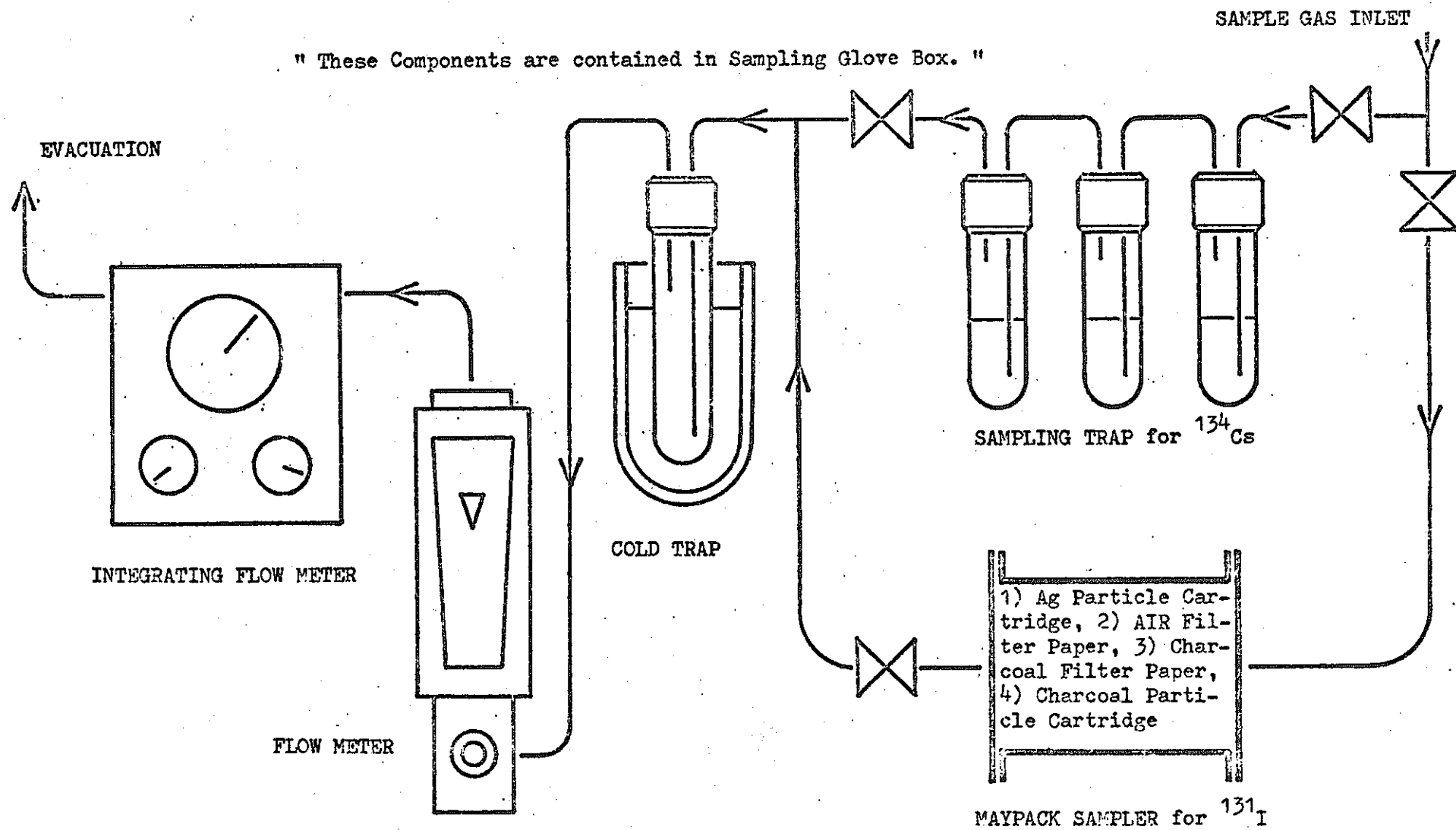


Fig. 4 Schematic Diagram of Sampling System for Measurement of  $^{131}\text{I}$  and  $^{134}\text{Cs}$  Transported into Gas Phase

Item	Condition
Flow Rate of Waste Gas	2.5 l/min
Total Sampling Gas Volume	20 l
Sampling Temperature	R.T.
MAYPACK's Bulking Material	Ag Particle Cartridge AIR Filter Paper Charcoal Filter Paper Charcoal Particle Cartridge

Table 2 Sampling Condition for  $^{131}\text{I}$  Transported into Gas Phase

Item	Condition
Flow Rate of Waste Gas	0.8 l/min
Total Sampling Gas Volume	4 l
Sampling Temperature	R.T.
Trapping Solution	1/10N $\text{H}_2\text{SO}_4$ 5 ml/Each Trap 3 pcs.

Table 3 Sampling Condition for  $^{134}\text{Cs}$  Transported into Gas Phase



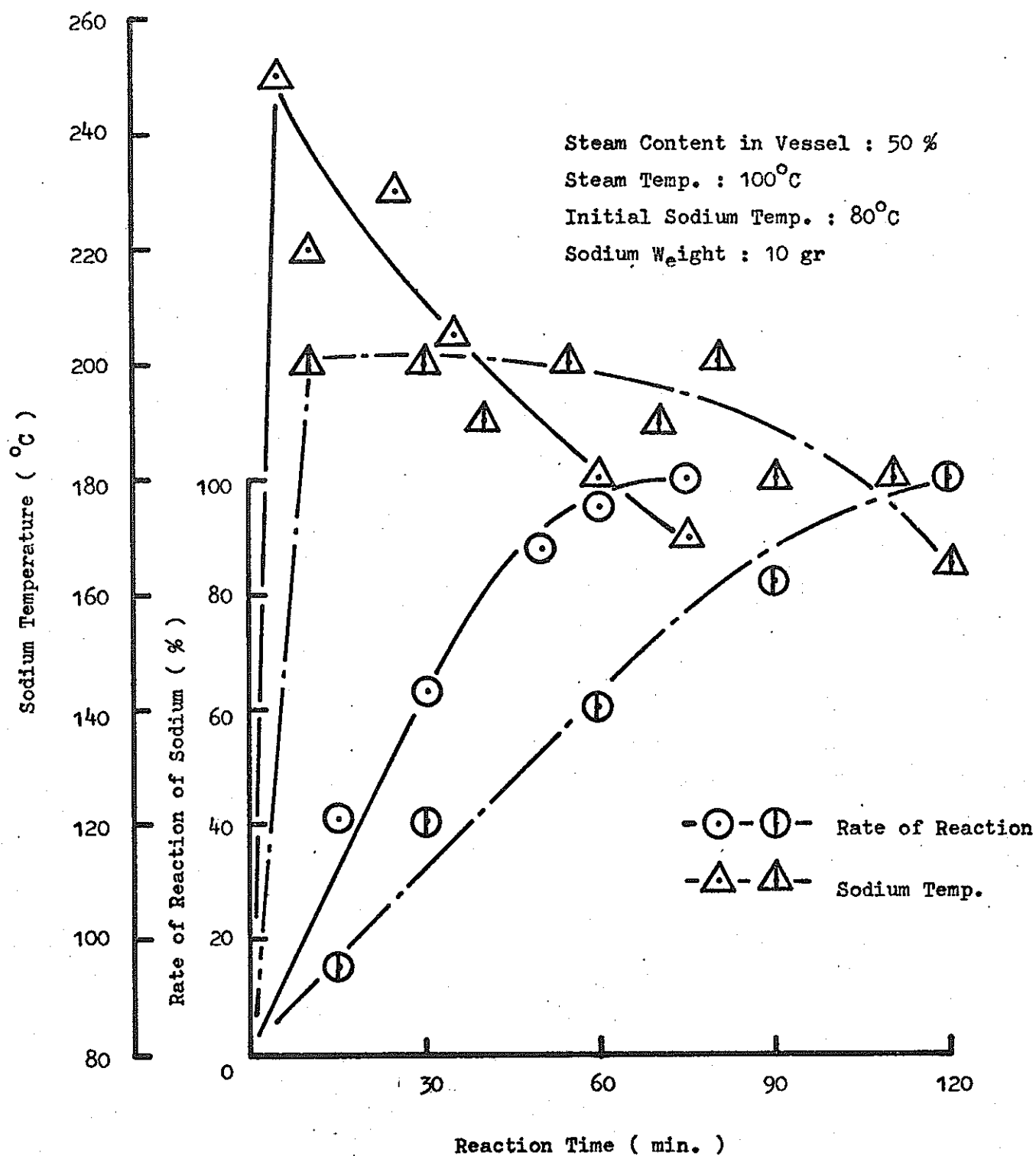


Fig. 5 Reaction Rate and Sodium Temperature in Static  
 Nitrogen-Moist Atmosphere

Radio Nuclide	Radioactivity ( mCi/10gr Na )	Reaction Behavior	Partition Ratio of Radio Nuclide Transported into Gas Phase
$^{131}\text{I}$	0.1	Normal Burning	$4.49 \times 10^{-5}$ $1.00 \times 10^{-2}$
$^{134}\text{Cs}$	0.15	Normal Burning	$4.00 \times 10^{-3}$ $9.91 \times 10^{-2}$
$^{24}\text{Na}$	0.08 0.09	Normal Burning	$2.2 \times 10^{-5}$ $1.2 \times 10^{-2}$

Table 4 Partition Ratios for  $^{131}\text{I}$ ,  $^{134}\text{Cs}$  and  $^{24}\text{Na}$  Transported into Gas Phase

Radio Nuclide	Initial Content in Na ( $\mu\text{Ci}/10 \text{ g-Na}$ )	Reaction Behavior	Transported into Gas Phase ( $\mu\text{Ci}$ )	Partition Ratio for Waste Gas	Remained in Waste Soln. ( $\mu\text{Ci}$ )
$^{131}\text{I}$	100	Normal	$4.5 \times 10^{-3}$	$4.49 \times 10^{-5}$	85.6
		Burning	1.0	$1.00 \times 10^{-2}$	97.4
$^{134}\text{Cs}$	150	Normal	$4.1 \times 10^{-3}$	$4.00 \times 10^{-3}$	124.7
		Burning	15.08	$9.91 \times 10^{-2}$	129.2
$^{24}\text{Na}$	80.6	Normal	$1.8 \times 10^{-3}$	$2.20 \times 10^{-5}$	79.6
	92.9	Burning	1.12	$1.20 \times 10^{-2}$	88.5

Table 5 Radioactivities Transported into Gas Phase during Nitrogen-Moist Treatment of Sodium

Radio Nuclide	Transported Activity by Sodium Burning ( $\mu\text{Ci}$ )	Remained Activity after 1 hr ( $\mu\text{Ci}$ )
$^{131}\text{I}$	1.0	0.986
$^{134}\text{Cs}$	15.8	1.58

Table 6 Activities Change of  $^{131}\text{I}$  and  $^{134}\text{Cs}$  in the  
Gas Phase by Sedimentation

Reaction Behavior	Ag Particle Cartridge	AIR Filter Paper	Charcoal Filter Paper	Charcoal Particle Cartridge
Normal	N. D.	N. D.	37.8 %	62.2 %
Burning	4.3 %	0.2 %	31.4 %	64.1 %

Table 7 Distribution of Trapped  $^{131}\text{I}$  in MAYPACK's Bulking Materials

Trapping Soln. ( 5 ml )	Trapped Iodine Content ( % )	Remarks
1/10N NaOH	71.5	Iodine or Inorganic Iodine Compounds
Xylene Carbon Tetrachloride	<div> <div>15.2</div> <div>13.3</div> <div>}</div> <div>29.5</div> </div>	Organic Iodine Compounds

Table 8 Results on the Qualitative Analysis Iodine Compounds  
Transported into Gas Phase

Radio Nuclide	Integrating Supplied Activity ( $\mu\text{Ci}$ )	Absorbed on Innersurface of Pipings ( $\mu\text{Ci}$ )	Trapped in G. Bubbler ( $\mu\text{Ci}$ )	Trapped in Scrubber ( $\mu\text{Ci}$ )	In C.P. Filter ( $\mu\text{Ci}$ )	In HEPA Filter ( $\mu\text{Ci}$ )
$^{131}\text{I}$	$8.2 \times 10^{-1}$	$1.9 \times 10^{-1}$	N.D. ( $< 7.44 \times 10^{-2}$ ) <sup>*</sup>	-	$1.53 \times 10^{-3}$	-
	$1.5 \times 10^{-1}$	$3.0 \times 10^{-2}$	-	N.D. ( $< 7.44 \times 10^{-2}$ ) <sup>*</sup>	$2.72 \times 10^{-4}$	-
$^{134}\text{Cs}$	$1.2 \times 10^{-1}$	$6.6 \times 10^{-2}$	N.D. ( $< 5.14 \times 10^{-2}$ ) <sup>*</sup>	-	-	N.D. ( $< 1.7 \times 10^{-5}$ ) <sup>*</sup>

\* : Converted Value from Detection Limit of Na(I) Detector.

N.D. : None Detectable

G. Bubbler : Gas Bubbler

C.P. Filter : Charcoal Particle Filter

Table 9 Radioactivity Distribution on Waste Gas Treatment System Components

#### 4. Discussion

##### 4-1 Transport of F.P. in Sodium into Gas Phase during Sodium-Steam Reaction

From the results of "Measurement of Transport of Radio Nuclides into Gas Phase",  $^{131}\text{I}$  and  $^{134}\text{Cs}$  transported into gas phase even under the normal sodium-steam reaction.

It is considered that  $^{134}\text{Cs}$  transported into gas phase by evaporation of NaOH solution as in the case of  $^{24}\text{Na}$  in the case of  $^{24}\text{Na}$  in the previous experiment, and was existing in the form of like  $\text{Na}(\text{Cs})\text{OH}$ . The separation ratio for  $^{131}\text{I}$  was in the order of  $10^{-5}$  which was about the same level of  $^{24}\text{Na}$  and was comparatively low level. In the case of  $^{134}\text{Cs}$ , its value was 100 times higher than  $^{24}\text{Na}$  and  $^{131}\text{I}$ .

In the event of any abnormal phenomena such as sodium ignition or burning during sodium treatment by steam, the transport of F.P. and  $^{24}\text{Na}$  indicated a drastic increase.

In the case of  $^{24}\text{Na}$  and  $^{131}\text{I}$ , transport ratio increased to 500 and 250 times as much as those of the normal reaction, respectively.

While for  $^{134}\text{Cs}$ , its increase was about 25 times as much as that of the normal reaction.

However,  $^{134}\text{Cs}$  transported into gas phase decreased to 1/10 after one hour. According to the visual observation of sodium-steam reaction, it was assumed that  $^{134}\text{Cs}$  transported into gas phase either with Na aerosol or aerosol itself, because Na aerosol generated by sodium burning settled down and disappeared after about 1.5 hour. Therefore, it can be presumed that  $^{134}\text{Cs}$  may be transported into gas phase in the form of  $\text{Na}(\text{Cs})$  aerosol under sodium burning.

On the other hand,  $^{131}\text{I}$  indicated small time-lapse changes of its concentration in gas phase after sodium burning, and it floated and



remained in gas phase even after Na aerosol had settled down. While sampling of  $^{131}\text{I}$  transported into gas phase was down by MAYPACK sampler, almost of  $^{131}\text{I}$  was captured by charcoal and any trace of  $^{131}\text{I}$  was not detected from the silver particle cartridge.

When a gas containing various iodine compounds is introduced into MAYPACK sampler filled with predescribed sampling elements, it is generally said that the molecular iodine is captured by silver particle cartridge,  $\text{NaI}$  is captured by activated charcoal filter paper, while the organic iodine compound is captured by activated charcoal particle cartridge. Consequently,  $^{131}\text{I}$  which remains in gas phase after the sedimentation of Na aerosol will transport in the form of iodine compound as  $\text{NaI}$ .

According to the studies of Kitani, et al.<sup>1)</sup>, iodine transports into gas phase accompanied with sodium oxide except organic iodine compound. Thus, it is noticed that it agrees with the behavior of  $^{131}\text{I}$  in this experiment.

Further, in order to analyze qualitatively the chemical form of  $^{131}\text{I}$  contained in waste gas generated during sodium burning, waste gas was passed through  $\text{NaOH}$  solution, xylene and carbon tetrachloride.  $^{131}\text{I}$  as captured by xylene and carbon tetrachloride after passing through  $\text{NaOH}$  solution was observed as shown in Table 8. The above mentioned report by Kitani, et al. confirmed that iodine was released into gas phase also in a form of organic iodine compound during sodium pool fire which was similar to the phenomena of the results of this experiment.

#### 4-2 Decontamination Factor of Gas Bubbler and Scrubber

The decontamination factors of scrubber and gas bubbler are shown in Table 10. From these results D.F. =  $4 \times 10^2$  for  $^{131}\text{I}$ , and D.F. =  $1.3 \times 10^3$  for  $^{134}\text{Cs}$  were obtained. The decontamination factor of sampling trap tube with 1/10 N  $\text{H}_2\text{SO}_4$ , as shown in Fig. was  $3.92 \times 10^3$  under normal reaction and  $1.7 \times 10^4$  under sodium burning for  $^{134}\text{Cs}$  respectively, these indicate well consistency with those of gas bubbler.

On the other hand, it is presumed that  $^{131}\text{I}$  may exist in gas phase mainly in the form of  $\text{Na}^{131}\text{I}$  under both normal and abnormal reactions and also  $^{134}\text{Cs}$  was in the form of  $\text{Na}(\text{Cs})\text{OH}$  during normal reaction. Therefore, the decontamination factor of the gas bubbler and scrubber for  $\text{Na}^{131}\text{I}$  is not so much different from for  $^{134}\text{Cs}$ , because both  $\text{NaI}$  and  $\text{Na}(\text{Cs})\text{OH}$  dissolve easily in water. In respect of  $^{131}\text{I}$ , however, organic iodine compound is not removed by gas bubbler and scrubber, and so some kind of effect might have decreased decontamination factor for  $^{131}\text{I}$  of these components.

Based on these discussion, it is considered that the gas bubbler and the scrubber, if those function well, are able to give the decontamination factor of  $10^3$  order for  $^{134}\text{Cs}$  and  $^{131}\text{I}$  excepting organic iodine compound.

#### 4-3 Decontamination Factor of Charcoal Filter and HEPA Filter

As these filters were used only for minimizing the release of RI to air, the decontamination factor was not measured for these filters in this time.

For the decontamination factors of these filters, various experiments have been done relating to air conditioning for light water reactor,

Radio Nuclide	Gas Bubbler	Scrubber
$^{131}\text{I}$	$4.1 \times 10^2$	$4.4 \times 10^2$
$^{134}\text{Cs}$	$3.1 \times 10^3$	-

Table 10 Presumption of Decontamination Factor of Gas  
Bubbler and Scrubber for  $^{131}\text{I}$  and  $^{134}\text{Cs}$

and therefore, sufficient analogical inferences are possible from these accumulated data.

It is said that the decontamination factor of charcoal particle filter for organic iodine compound is more than  $1 \times 10^3$  when the relative humidity is about 40 %. In this case, the effect of relative humidity to decontamination factor is great, and when the relative humidity rises upto 97 %, decontamination factor decreases to  $D.F. = 3.3 \times 10^1$ .

Even under such condition of high relative humidity, they retain high decontamination capacity in the order of  $D.F. = 1 \times 10^2$  for inorganic iodine compound.

As no heating was applied to the piping from the outlet of the gas bubbler and scrubber to the charcoal filter and the filter assembly itself, the relative humidity of the waste gas introduced into the charcoal filter was assumed to be close to 100 %. And so, decontamination factor was estimated to be at  $10^1$  for organic iodine compound, and about  $10^2$  for inorganic iodine compound.

However, such decontamination factor of charcoal filter as  $D.F. = 10^1$  for organic iodine compound is considered too low, and some counter-measures are necessary to be taken.

For this purpose, it is necessary to maintain the relative humidity of waste gas as low as possible. This will be realized by heating the inlet piping of the charcoal filter and the filter assembly itself. And decontamination factor for  $^{131}\text{I}$  is presumed to be raised more than  $10^3$  by this way.

It is generally said that decontamination factor of HEPA filter is  $D.F. = 3.3 \times 10^3$  when the filtrating pore size of filter paper is  $3\mu$ , and even in such case, the humidity of waste gas is desirable to be low in order to keep the integrity of the filter paper. In this experiment,

since HEPA filter's inlet piping was heated upto  $150^{\circ}\text{C}$ , the relative humidity of waste gas was presumed to have been reduced, and decontamination factor of HEPA filter for  $^{134}\text{Cs}$  was thought to be close to  $10^3$  level.

When the relative humidity of waste gas is low as above described, D.F. =  $10^3$  for charcoal filter and D.F. =  $10^3$  for HEPA filter are expected to be attainable.

#### 4-4 Decontamination of Waste Gas in Waste Gas Treatment System

From the results of above mentioned evaluation of decontamination factors for  $^{131}\text{I}$  and  $^{134}\text{Cs}$  in waste gas, decontamination factors under optimum operate conditions of various components of the test apparatus which was used in this experiment is as shown in Table 11. From these data, it is assumable that even in the event of sodium burning during sodium treatment by steam, the concentration of  $^{131}\text{I}$  and  $^{134}\text{Cs}$  in waste gas which is released from the waste gas treatment system can be maintained sufficiently low so that F.P. removal in waste gas can be effectively carried out.

Radio Nuclide	Reaction Behavior	Sedimentation ( after 1 h )	Absorption on Pipings	Gas Bubbler	Scrubber	Charcoal Filter		HEPA Filter
						R.H.*=97%	R.H.*=40%	
$^{131}\text{I}$	Normal Burning	N.G.** 1.01	N.G.** 4.3	$10^3$	$10^3$	$10^1$ (ORG.***) $10^3$ (Inorg.***)	$10^5$ (Both)	-
$^{134}\text{Cs}$	Normal Burning	N.G.** $10^1$	N.G.** 1.8	$10^3$	-	-	-	$3.3 \times 10^3$

\* R.H. : Relative Humidity

\*\* N.G. : Negligible

\*\*\* Org. : Organic Form

Iorg. : Inorganic Form

Table 11 Preliminary Consideration of Decontamination Factor on Waste Gas Treatment System Component

## 5. Conclusion

The results of the experiments and consideration may be summarized as follows:

- 1)  $^{131}\text{I}$  and  $^{134}\text{Cs}$  added into sodium transport into gas phase of somewhat higher than vapor pressure of their respective elements during a normal sodium-steam reaction.

In this case,  $^{131}\text{I}$  and  $^{134}\text{Cs}$  are considered to transport into gas phase in the form of  $\text{Na}^{131}\text{I}$  and  $\text{Na} (^{134}\text{Cs})\text{Oh}$ .

- 2) In the event of ignition and burning of sodium with the introduction of excess steam, the transport ratio of  $^{131}\text{I}$  and  $^{134}\text{Cs}$  into gas phase increased, and reached upto about 200 and 25 times as high as those of the normal reaction.

But in the case of  $^{134}\text{Cs}$ , it is presumable that it has transported into gas phase in the form of  $\text{Na} (^{134}\text{Cs})$  aerosol, since its concentration in the gas phase decreased by sedimentation, and its behavior was about the same as that of Na aerosol generated by the sodium burning.

Contrary to this, it is presumed that  $^{131}\text{I}$  has transported into gas phase as  $\text{Na}(\text{Na}^{131}\text{I})$  aerosol,  $\text{Na}^{131}\text{I}$  and organic iodine compound, and that its main substance is  $\text{Na}^{131}\text{I}$ , because  $^{131}\text{I}$  indicated hardly any sedimentation in gas phase and generation of organic iodine compound and its transport are assumable.

- 3) Generation of organic iodine compound by sodium burning is thought almost unquestionable. But as no organic iodine quantifying equipment was provided to this experimental apparatus, quantitative evaluation was not possible in this experiments.
- 4) Decontamination factor of gas bubbler and scrubber used in this experiment for  $^{131}\text{I}$  and  $^{134}\text{Cs}$  is considered at the level of  $10^3$ .

Since charcoal filter and HEPA filter were used as only backup purpose in this experiment, no measurement of decontamination factor were made.

Based on such inferences obtained from the Light Water Reactor related facilities, decontamination factor of charcoal filter for  $^{131}\text{I}$  and of HEPA filter for  $^{134}\text{Cs}$  was expected to be  $10^5$  maximum and  $3.3 \times 10^3$ , respectively.



## 6. Reference

- 1) KITANI, S. et al.: Formation of Volatile Radioiodine Compounds in Sodium Pool Burning, J. Nucl. Sci. Technol., Vol. 12, No. 11. pp 717-721, 1975.