MONITORING TECHNIQUE FOR RADIOIODINE IN SODIUM AEROSOL

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Abstract

Radioiodine monitoring in a liquid-metal fast breeder reactor is different from that in the other types of reactor because radioiodine is always associated with the radioactive sodium aerosol. In this connection, experiments were made on behavior of the iodine and collection characteristics of an air sampler (JAERI type).

In the experiment, the fume of sodium containing sodium iodide was produced by vaporization of the liquid sodium, labelled with ^{24}Na and ^{131}I. It was then introduced through a fume transport tube into an ageing chamber with control of the temperature and relative humidity of air.

Characteristics of the airborne iodine in the sodium aerosol in the chamber were examined with May packs. About 97 % of iodine in the aerosol at room temperature is in particulate form (as sodium iodide), and percentage of gaseous iodine increases with rise of the fume temperature to about 30 % at 400°C.

More than 99 % of the iodine in the aerosol can be collected by a particulate and a charcoal filter paper in the sampler. In order to detect the radioiodine in sodium aerosol, however, the iodine should be collected separate from the sodium aerosol. For monitoring radioiodine in aerosol, a sampling technique with an arc discharge was contrived. It consists in the decomposition of aerosol, followed by the iodine sampling, the portion of sodium and of iodine thus separated are collected in the particulate and the charcoal filter paper, respectively.

1. Introduction

Monitoring of airborne radioiodine is important for radiation protection in a liquid metal fast breeder reactor as well as in the other types of reactor.[1] Air contamination due to radioiodine in a sodium cooled reactor, which is produced mainly by sodium pool fire or sodium spray fire[2], is associated with the radioactive sodium aerosol because radioiodine released from the failed fuel is trapped effectively as sodium iodide in liquid sodium.[3,4]

There are several experiments made for the pool fire, which is caused by major sodium leakage. In this case, the ratio of I/Na in vapor and liquid is reported to be $0.3\sim1.0$[5~8] and the iodine behaves as sodium iodide which can be effectively collected by a dust sampling filter paper such as cellulose asbestos filter paper. However, no experiments are made on the behavior of airborne radioiodine released by spray fire, in which sodium is introduced explosively into an oxidizing atmosphere by defect in primary system piping. It is considered that the characteristics of radioiodine in this case are different from those of a pool fire, because in spray fire, sodium is released into air in the form of fine particles of a high temperature and oxygen and moisture in

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the atmosphere is sufficient to oxidize the sodium. Air contamination in liquid cooled reactor may often be caused by this type of fire.

To establish the techniques of sampling the airborne radioiodine it is important to find the physical and chemical forms of airborne radioiodine when a small amount of sodium containing radioiodine is released into the air or inert atmosphere.

In the present study, assuming that radioiodine is trapped in the liquid sodium, the forms of airborne iodine were investigated by May pack, so named after its originator Fred May,[9] for the aerosol produced when Na-NaI was sprayed into the inert gas or air at various fume temperatures. And also collection performance of the JAERI-type air sampler for iodines was tested.

In this paper, the experimental results obtained are described and also discussed a new technique for sampling radioiodine separate from sodium aerosol.

2. Experimental

2.1 Generation of Na-NaI aerosol

A mixture of sodium (40g) and sodium iodide (40 μg of I), labelled with ^{24}Na (2 mCi) and ^{131}I (2 mCi), respectively, was prepared in a stainless steel can. The mixture in the can was melted in an electric induction furnace as shown in Fig. 1 which shows schematic diagram of the generator of sodium aerosol contaminated with iodine. In order to homogenize sodium iodide in the liquid sodium, the furnace was frequently vibrated at 350°C.

The temperature of liquid sodium was raised to 400$^{\circ}\sim$550°C, with argon as the sweeping gas at 3 liter/min. Then, the temperature of sodium fume from the can was controlled to 25$^{\circ}\sim$ 400°C by passing through the fume transport-tube with tape heater. The fume was sprayed into the air in a reaction cell with control of the relative humidity at 25°C, and the produced aerosol was introduced into the chamber of volume about one cubic meter, at the same humidity, aged for 100 sec in it.

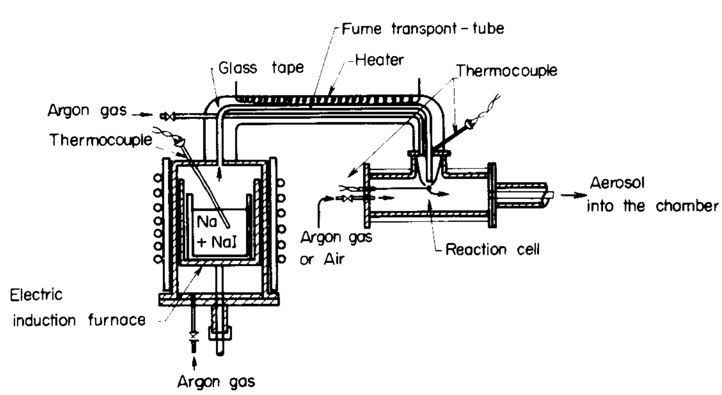

Fig.1 Aerosol generator

2.2 Identification of iodine form by May pack

The aerosol was sampled by two May packs placed in parallel at the chamber exit. The respective arrangements of components are shown in Fig. 2, the components are listed below.

1) One Toyo HE-40 cellulose asbestos filter paper (particulate filter paper)
2) Four silver plated copper gauzes (80 mesh)
3) One Toyo CP-7 charcoal-loaded filter paper
4) Two 10 mm thick activated charcoal cartridges

Sampling was made for 30 min at the flow of 15 liter/min (face velocity 30 cm/sec).

The forms of iodine in sodium aerosol were estimated from the distribution of sodium and iodine collected in the components of May pack. The distribution was determined by measuring ^{131}I and ^{24}Na as the tracers. Radio-activities were measured by a γ-ray spectrometer with a 1-3/4"ϕx2" NaI(Tl) well type detector connected to a 200 channel analizer.

Identification of the iodine forms was made on the basis of collection performance of the May pack.[10,11] Iodine in particulate filter paper is particulate; in silver gauzes, elemental iodine; in charcoal filter paper,

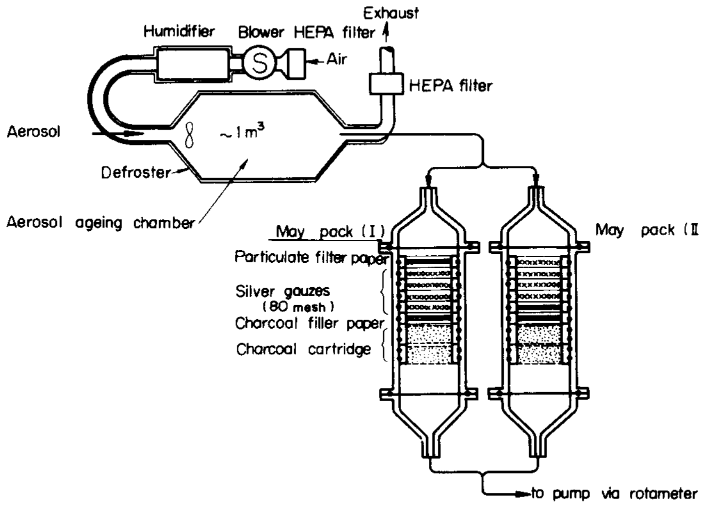

Fig.2 Apparatus for identifying iodine forms in sodium aerosol.

iodine compounds;and organic iodide such as methyl iodide is in the charcoal cartridge. In this method, it is necessary to determine the amount of gaseous iodine collected by sodium aerosol in the particulate filter paper. This was made by comparing the fractional distribution of iodine in the May pack when the gauzes were placed before the particulate filter paper and when placed after it.

3. Properties of iodine in sodium aerosol

3.1 Iodine form in an inert gas

Sample was taken at the fume generator exit as shown in Fig.1, in the absence of air. Fig.3 shows the fractional distributions of sodium and iodine in components of May packs for the sodium fume in an inert atmosphere.

As seen in the figure, the iodine penetrating through the particulate filter paper is less than 0.1%, and the ratio of iodine to sodium fractions in the filter paper is the same for both the May packs and the iodine and sodium fractions in the four silver gauzes are the same in May pack (I). Therefore, the iodine form in sodium fume under an inert atmosphere is considered to be particulate such as Na-NaI particles. Fractional distributions of iodine and sodium at various temperatures of sodium fume were the same as shown in Fig.3. Most of the iodine (99.9%) is presumed to be particulate, independent of the temperature of sodium fume.

3.2 Forms of iodine in the air

Fig.4 shows the fractional distributions of sodium and iodine in the components of May packs for the aerosol produced from the fume of 300°C. The fraction of iodine trapped in the particulate filter paper is the same in both the May packs, and also the fractional distributions of iodine in the four silver gauzes and in the charcoal filter paper were similarly the same. This shows that gaseous iodine, including elemental iodine,is not trapped in the filter paper loaded with a large quantity of sodium aerosol. Consequently, the forms of iodine and the respective percentages in the aerosol were estimated by the May pack (I).

Fig.5 shows the abundances of gaseous iodine in the sodium aerosol at the sodium fume temperatures of 25° to 400°C. The gaseous iodine increases with fume temperature except near 100°C, 37% at 400°C and 1.5~3% at 25°C.

The iodine trapped in the silver gauzes, charcoal filter paper and charcoal cartridges are approximately 30, 70 and 2% of the gaseous iodine, respectively, at all temperatures, except 100°C. The percentage of gaseous iodine at the fume temperature of 100°C was about 10 times as large as that on the "general" curves of abundance and temperature. However, the elemental iodine trapped in the silver gauzes was about only 3 times as large. It is thus seen that the abundance of less reactive iodine increases remarkably near 100°C.

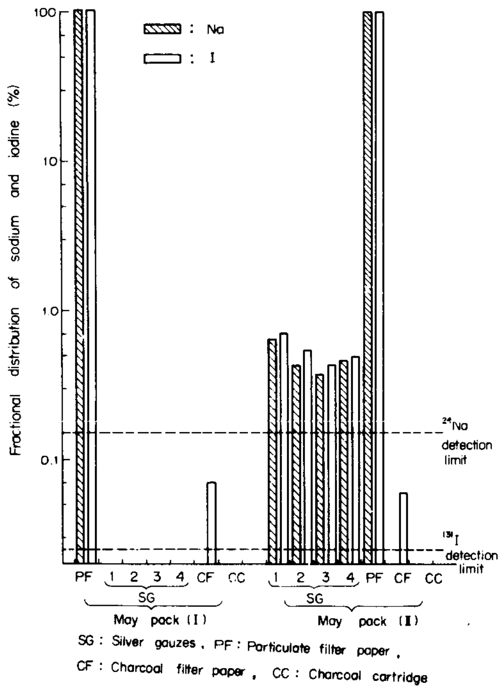

Fig.3 Fractional distribution of sodium and iodine (in inert atmosphere at 450°C)

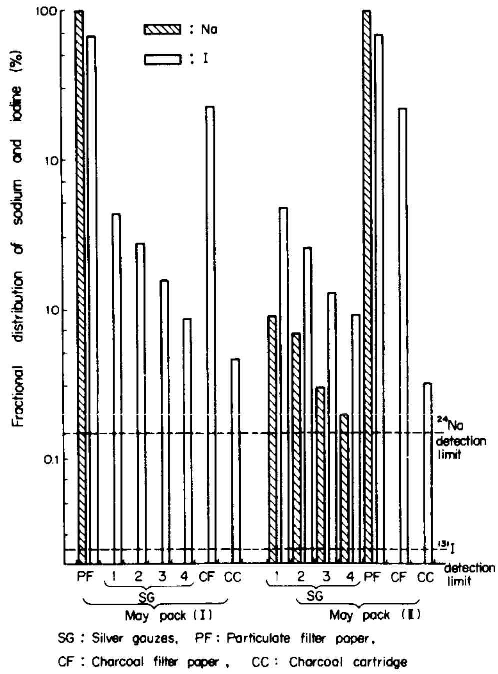

Fig.4 Fractional distribution of sodium and iodine (in air atmosphere at 300°C)

The percentages of particulate and gaseous iodines under various conditions are shown in Table 1. It is seen from the table that abundance of gaseous iodine decreases with increase of the humidity of the air.

4. Collection performance

The air sampler consists of a particulate filter paper and a charcoal filter paper in this order. The collection efficiencies of these two components were estimated for radioiodine in the sodium aerosol described in Sec.3. The results are shown in Table 2 together with the overall efficiency of the sampler.

In the sodium fume containing sodium iodide, sprayed into the inert atmosphere of 25°C, more than 99% of the total iodine is collected in the particulate filter paper together with the sodium, because radioiodine exists as Na-NaI particles.

When the sodium fume of 25°C is sprayed into the air, on the other hand, 3% and 1% of the iodine are trapped alone (i.e., with no sodium) in the charcoal filter paper at the air humidity of 40% and 80%, respectively. At the fume temperature of 300°C, the fraction of iodine collected in the charcoal filter paper increases to 29% and 5% at the humidity of 40% and 80%, respectively. The overall collection efficiency for iodine is more than 99% for both the inert and air atmospheres, but the amount of radioiodine is difficult to measure because of the large amount of radioactive sodium. The amount of gaseous iodine in the sodium aerosol can be estimated by measuring the activity in the charcoal filter paper, separate from that in the particulate filter paper. However, the total iodine in the aerosol is difficult to measure from the activity in the charcoal filter paper, because the fraction of gaseous iodine varies largely with temperature of the sodium fume and the air humidity.

In order to improve the detection sensitivity for the radioiodine in the radioactive sodium aerosol, the iodine should be collected separate from the sodium aerosol. This can be made by decomposition of the sodium iodide using an arc discharge[12] tube upstream the sampler. By the arc discharge technique,

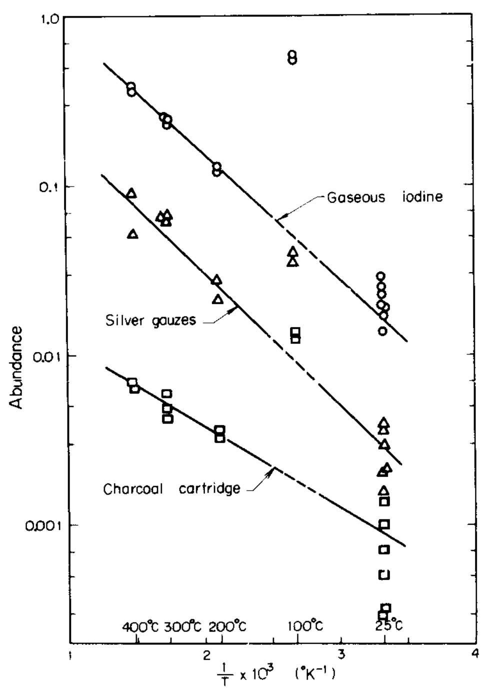

Fig.5 Effect of temperature of released sodium on abundance of gaseous iodines

80~90% of the iodine in the aerosol was decomposed into gaseous iodine, which can be collected efficiently in the charcoal filter paper. The collection efficiencies of the sampler using this technique are shown on the right side of Table 2. As seen in Table 2, 80~90% of the iodine is selectively collected in the charcoal filter paper from the sodium aerosol, regardless of the sodium fume temperature and the air humidity. Thus, the concentration of total radioiodine in the radioactive sodium aerosol can be measured sensitively and accurately from the iodine activity collected in the charcoal filter paper.

Sodium iodide in the inert atmosphere, however, was not decomposed by arc discharge technique, as seen in Table 2. To use this technique, therefore, arc discharge must be made in air.

Table 1 Percentage of particulate and gaseous iodines in the sodium aerosol

Atmosphere	Temperature of sodium fume (°C)	Relative humidity (%)	Run number	Percentage of iodine (%)	
				particulate	gaseous
Argon	25	dry	8	99.9	0.1
	300	dry	4	99.9	0.1
Air	25	40~95	21	98	2
	100	40	2	42	58
	200	40	2	88	12
	300	40~60	10	74	26
		80	4	95	5
		95	2	98	2
	400	40	2	63	37

Table 2 Collection efficiency of the sampler for the radioiodine in sodium aerosol

Temperature of sodium fume (°C)	Atmosphere	Relative humidity (%)	Collection efficiency (%)					
			Without arc discharge			With arc discharge		
			Overall *	particulate filter paper	charcoal filter paper	Overall *	particulate filter paper	charcoal filter paper
25~400	argon	dry	99	99	<1	99	99	<1
25	air	40	99	96	3	99	19	80
		80	99	98	1	99	11	88
300	air	40	99	70	29	99	22	77
		80	99	94	5	—	—	—

* sum of collection efficiencies for two filter papers

5. Conclusion

The forms of airborne iodine, when the sodium fume containing 1 ppm of sodium iodide is sprayed in the inert and air atmospheres, were examined experimentally. The results obtained are summarized as follows.

1) When the Na-NaI fume is sprayed into the inert atmosphere(Ar), no gaseous iodine is produced;the airborne iodine is considered as sodium iodide.

2) When sprayed into the air, the percentage of the gaseous iodine increases with increase of the fume temperature, about 30% at 400°C and 3% at 25°C at the air humidity of 40% and the percentage also depends on the humidity. The fraction of elemental iodine and inorganic iodide in the gaseous iodine are about 30% and 70%, respectively. At the fume temperature of near 100°C, however, the percentage of gaseous iodine increases to 60%, the forms are presumed to be different from those at other temperatures.

3) In monitoring radioactive iodine in the sodium aerosol such as in sodium coolant release from LMFBR, the sodium iodide with sodium aerosol is collected in the particulate filter paper and the gaseous iodine in the charcoal filter

paper. The percentage of iodine collected in the charcoal filter paper depends largely on the release conditions and atmospheric conditions. More than 99% of the iodine can be collected in the sampler consisting of the particulate and charcoal filter papers.

4) An approximately 90% of the iodine can be changed to the gaseous state by arc discharge technique, independent of the atmospheric conditions. The gaseous iodine is selectively collected in the charcoal filter paper from the sodium aerosol. Therefore, it is possible to measure airborne radioiodine accurately in the highly radioactive sodium aerosol, using the air sampler with the arc discharge tube.

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