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Note

Volume Reduction by the Incineration of the Combustible Radioactive Solid Samples from Radioisotope Usage at the Utilization Facility

——Estimation of the Distribution of Low Energy β-Emitter Using the Imaging Plate——†

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We want to establish a system of volume reduction by the incineration of the combustible radioactive solid wastes from radioisotope usage at the utilization facility. We have been performing experiments using an experimental incineration system to examine the distribution of radionuclides during incineration and to collect basic data. To reproduce the realistic conditions of incineration of low-level radioactive wastes in an experimental system, we adopted new incineration methods in this study. Low level radioactive samples (LLRS) were set up in a mesh container of stainless steel and incinerated at high temperature (over 800 ℃) generated by two sets of high calorie gas burners. Low energy β-emitters ^{35}S, ^{45}Ca, ^{33}P, *and a high energy β-emitter* ^{32}P *were used for the experiment. Their translocation percentages in exhaust air and dust were estimated using the Imaging Plate. Distribution of radionuclides during the incineration was similar to that estimated by conventional methods by our study or to that reported in incineration of liquid scintillation cocktail waste. We concluded that the use of the Imaging Plates is a simple and reliable method for estimation of the distribution of low energy β-emitters in incineration gas and ash.*

Key Words: low level radioactive sample (LLRS), low energy β-emitter, Imaging Plate, experimental incineration system

1. Introduction

1·1 Purpose

It is defined in Japanese law that any solid waste contaminated with radioisotopes should be preserved in a can to be collected by a licensed dealer of radioactive wastes[1]. It is unreasonable to treat in such a way the waste in which radioisotopes have decayed out to background radioactivity levels due to disintegration. We believe it is reasonable to reduce by combustion the volume of such wastes which contain various radioisotopes of short half life. It is important to exclude radioisotopes of long half life. Thus it is important to establish a system for the quantification of radioisotopes in solid waste. We want to establish a system of volume reduction by the incineration of the combustible radioactive solid wastes from radioisotope usage at the utilization facility. We have been performing experiments using an experimental incineration system to examine the distribution of radionuclides during incineration and to collect basic data for realization of the incineration of solid wastes in the near future[2),3)]. In this

† 使用施設における可燃性固体 RI 試料の焼却減容——低エネルギーβ線放出核種のイメージングプレートによる評価——。湯本泰弘，花房直志，永松知洋，岡田　茂：岡山大学アイソトープ総合センター，700-8558　岡山市鹿田町 2-5-1。

experiment, we investigated the use of the Imaging Plate for the distribution of radionuclides which emit low energy β-rays that cannot be detected easily by conventional methods.

1·2 Experimental incineration system

In a typical incineration, samples are put into a high temperature environment, over 800°C, and burned to ash. It is important to reconstruct realistic conditions in an experimental system. In an earlier experiment[2], a low level radioactive sample (LLRS) was put into the furnace after the furnace temperature was raised to 300°C by the incineration of materials which contained non-radioactive combustible materials. Huge volumes of furnace ash remained after the experiment and a few LLRS remained intact. The incomplete incineration was due to the fall of LLRS to the furnace ash collector. Therefore, we introduced a new system to guarantee a complete and high temperature incineration of LLRSs. One more high-calorie burner was added to the existing one to heat LLRS directly, and LLRS in a stainless mesh container was positioned between the two

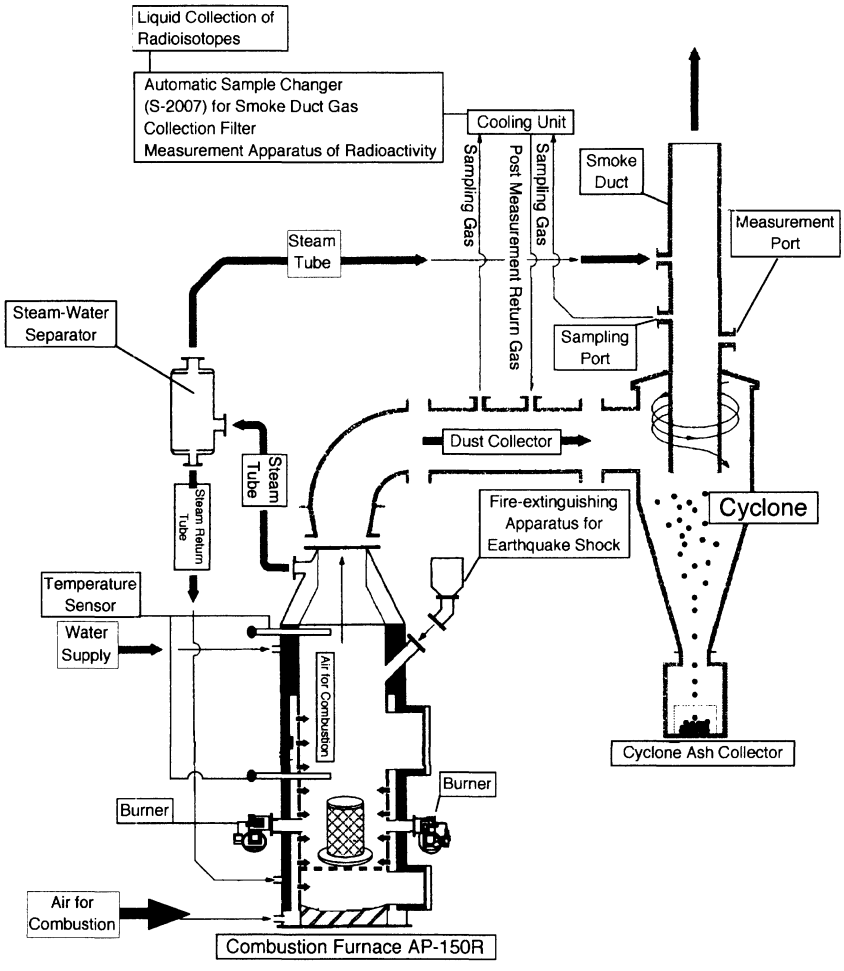

Fig. 1 Experimental incineration system.
Experimental samples were settled in a stainless steel container and incinerated with high calorie burners.

burners (Fig. 1). The problem of incomplete incineration caused by the fall of LLRS to the furnace ash collector was eliminated. Scattering of LLRSs was prevented by the stainless mesh container. Also, all furnace ash could be recovered and analyzed.

2. Materials and Methods

2·1 Materials

LLRS contains low energy β-emitter ^{35}S, ^{45}Ca, ^{33}P and high energy β-emitter ^{32}P.

2·2 Making LLRSs

Radionuclides used for LLRS were diluted with an appropriate buffer solution and the solution divided to 100 RIA test tubes (IUCHI: 22-459-01). These LLRSs were adjusted to about 5 kg in a stainless steel mesh container (35 cm in diameter, 40 cm in height, 0.5 cm mesh) with non radioactive plastic samples.

2·3 Incineration

The container was located on stainless tray (40 cm in diameter) between the two burners. The container was incinerated for 60 min, and the air was blown for 2 h without burner until burned out completely. The furnace was left for 3 h to cool down.

2·4 Collection of the samples

Exhaust air samples were collected by an automatic sample changer. Samples were trapped by a glass fiber filter (GB100R™, 60 mm in diameter ADVANTEC Co. Ltd.) for 5 or 10 min in each filter. Exhaust gas containing radioactive samples which passed through the filters was blown into a liquid collection apparatus for complete collection of radionuclides. Furnace ash was collected by a vacuum cleaner (compact cleaner, THE1105™, Chiyoda Technol Co. Ltd.). All ash on the furnace internal wall was collected by a vacuum cleaner, but ash on the inner wall of the smoke duct and cyclone was not collected. Ash in the cyclone ash collector was also collected by a vacuum cleaner.

2·5 Measurement by Imaging Plate

Radioactive filter samples were placed on an Imaging Plate (BAS-III™, Fuji Photo Film Co. Ltd.) along with standard samples for comparison, exposed for 48 to 72 h and read by a BAS imaging scanner (BAS-2000II™, Fuji Photo Film Co. Ltd.). Ash samples from furnace residue, furnace wall and cyclone were collected in a GB100R™ filter by suction with a dust sampler (Portable Air Sampler, NR2010™, Chiyoda Technol Co. Ltd.). After measuring the weight of the filter, the samples were placed on an Imaging Plate with standard samples, exposed for 72 h and then the Imaging Plates were analyzed by a BAS imaging scanner.

2·6 Correction of geometric configuration

To match the geometric configuration of standard samples to other samples, the standard samples were made by adsorbing radioactive solution to activated carbon powder, thereafter the samples were air dried. The radioactive samples were collected on a GB100R™ filter by a dust sampler.

2·7 Correction of thickness of samples

It is important to correct the effect of self adsorption of sample radiation for accurate measurement of low energy β-emitter. We made a standard curve showing the relationship between thickness of samples and counting efficiency.

2·8 Calculation of recovered amount of radioactivities

There exist linear correlations between the

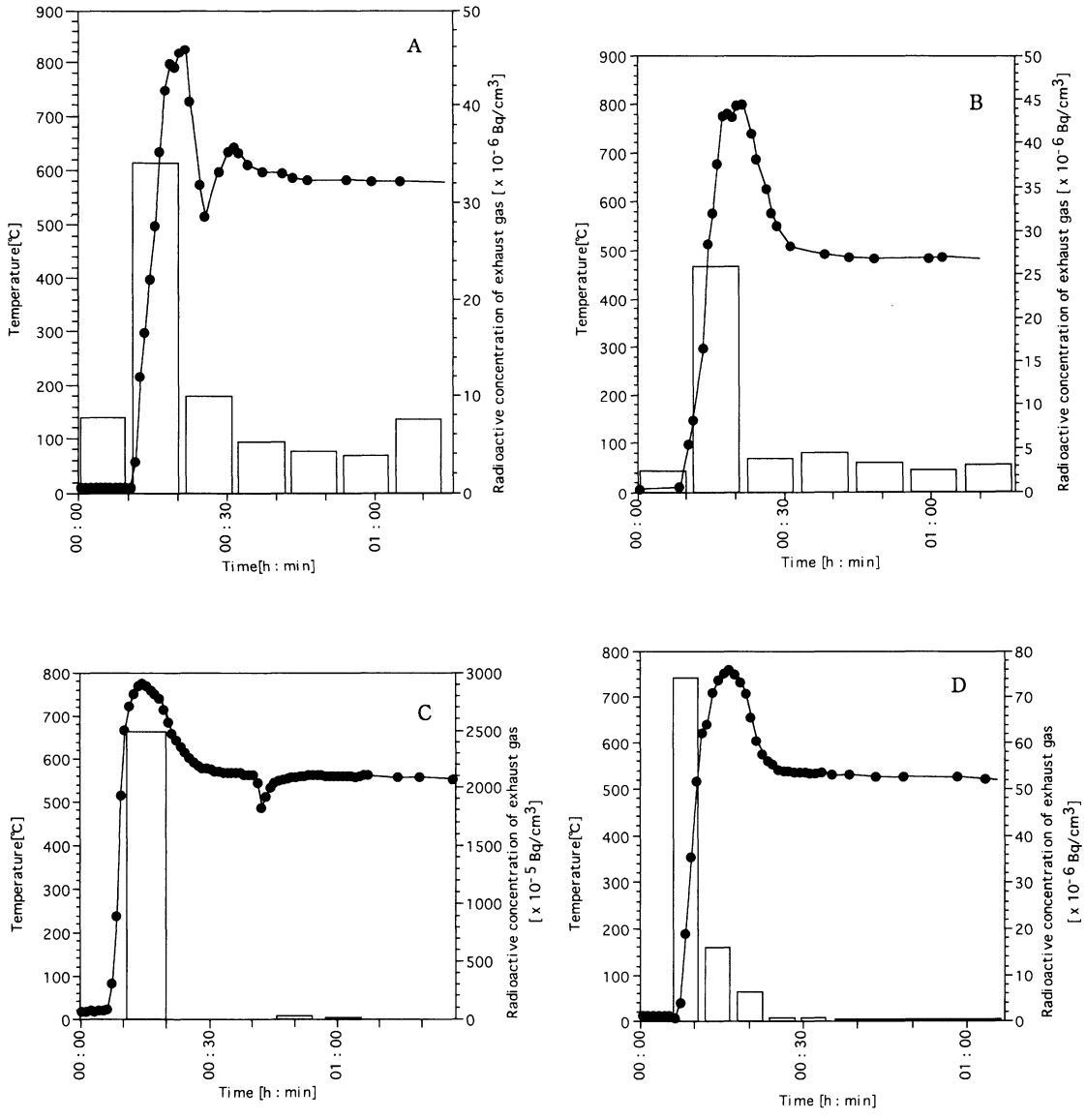

Fig. 2 Temperature of the furnace and exhaust gas concentration in experimental incineration.
A ; ^{35}S, B ; ^{45}Ca, C ; ^{33}P, D ; ^{32}P.
Temperature is indicated by a closed circle and radioactive concentration of the exhaust gas is indicated by an open box column.

photostimulated luminescence(PSL) values and the radioactivities in the sample of the same geometric configuration on the same Imaging Plate. Radioactivities of the samples were calculated from the ratios of PSL values between the samples and the standards of known radioactivities. Recovered amount of the residues and ash was calculated from the radioactivities per weight of the samples. Recovered amount of the exhaust air was calculated from the concentration of the radioactivities of the samples.

3. Results

3・1 Incineration

Experimental incineration was performed for nuclides ^{35}S, ^{45}Ca, ^{33}P and ^{32}P separately. Experimental samples were set in the furnace and air blast, automatic sample changer, liquid collection apparatus, measurement apparatus of radioactivity, data logger for measurement of the temperature and air velocity were started. After collecting background samples for 5 min, 2 burners were ignited. Six or 7 samples were collected for 5 or 10 min in each filter with an interval of 1 min. The furnace temperature reached about 800°C after 10 min of ignition, thereafter it reached a plateau of 500 to 600°C (Fig. 2). Flames of the burner did not reach the temperature sensor because it was set 10 cm over the containers. As the temperature of the flames of the burner reached 1 000°C the color of container changed to bright orange. We concluded that the temperature of the container reached a temperature over 800°C. Almost all samples were burned out in 20 min and ashing was completed after 1 h of incineration. As radioactivity was detected for 10 min just after ignition, we thought that the samples burned out in a short time and then scattered (Fig. 2) into the smoke duct gas.

3・2 Collection of samples

Radionuclides were released as radioactive gas or dust in exhaust air. In this process, some parts adhered to the furnace wall, smoke duct, or cyclone wall. Most residue samples remained on the tray but some ash scattered as dust and was recovered by the cyclone. To evaluate the exact distribution of radionuclides, we collected exhaust air, furnace residue, cyclone ash and ash of the furnace wall. Radioactive samples in the exhaust air were collected by a filter and liquid trap. Residue and ash samples were collected to the dust bag of vacuum cleaner.

3・3 Correction of measured radioactive values

All samples for measurement were of the same geometry, in which a fine grain was adsorbed on the filter surface. It is most important to fit the geometric configuration of the standard and samples for accurate evaluation by an Imaging Plate[4),5)]. A solution of radionuclides for standard samples was prepared and it was adsorbed to activated carbon and air dried. The resultant carbon powders were collected by a GB100R™ filter by a dust sampler. In the case of ^{35}S standard samples, linear correlation between the thickness of samples and PSL values existed up to 3 mg/cm^2 thickness. When the thickness exceed 10 mg/cm^2, PSL values reached plateau regardless of a change in thickness (Fig. 3).

3・4 Measurement of sample radioactivity

Thickness of filter samples which adsorbed exhaust gas did not exceed 0.4 mg/cm^2, so we considered that there was no need to correct the measured value by thickness. Filter samples and

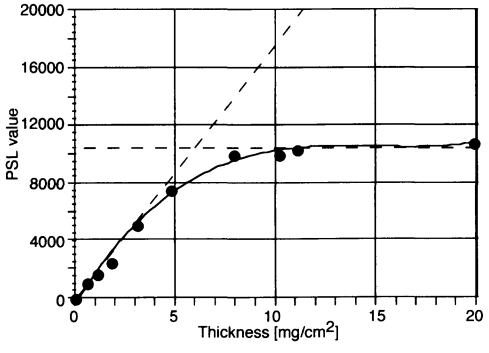

Fig. 3 Change of PSL value by the thickness of the samples.
PSL value of each thickness of the standard sample which contains ^{35}S nuclide is indicated by a closed circle.

Table 1 Distribution of low energy β-emitter during the experimental incineration, in and out of the furnace

Nuclide	Input amount (kBq)	Recovered amount						
		Residue (kBq)	Ash of wall and duct (kBq)	Cyclone ash (kBq)	Exhaust air (kBq)	Exhaust air concentration (Bq/cm^3)	Incineration time (min)	Recovery (%)
^{35}S(dCTP)	500	11.1 (8%)*	90.2 (65%)	2.70 (2%)	33.8 (25%)	9.95E-06	77	28
^{14}C(proline)	500	1.95 (0.6%)	0.95 (0.3%)	0.27 (0.1%)	304 (99%)	3.14E-05	66	61.7
$^{45}Ca(CaCl_2)$	150	72.9 (60%)	41.0 (34%)	1.00 (1%)	5.70 (5%)	6.43E-06	77	80.4
^{33}P(dCTP)	500	326 (82%)	46.5 (12%)	0.39 (0.1%)	23.3 (5.9%)	3.69E-06	88	79.3
^{32}P(dCTP)	75	36.3 (82%)	20.2 (29%)	8.95 (13%)	3.70 (5%)	3.58E-06	66	92

*Percentage of each recovered amount to tatal recovered amount is indiceted in parenthesis.

standard samples which matched the geometric configuration were set on the same Imaging Plate, exposed for a definite time and read by a BAS imaging scanner Dust and ash samples from the furnace, furnace wall and cyclone were adsorbed to a GB100R™ filter by a dust sampler and the weight of this filter was measured. Samples in which weight did not exceed 3 mg/cm^2 were used for analysis. In some samples, standards which were made by dropping a solution of a radionuclide to GB100R™ and then air dried, were used. In this condition, we corrected the measured values using the defined relation between this standard and the same geometric standard samples.

4. Discussion

The results of experimental incineration of ^{35}S, ^{45}Ca, ^{33}P, and ^{32}P are shown with the result of ^{14}C which was performed using a liquid scintillation counter in Table 1. Recovery rate of nuclides which do not vaporize (^{45}Ca, ^{33}P and ^{32}P) exceeded 80%, 60 to 80% of recovered radioactivity was detected in furnace ash and about 5% was emitted in the exhaust air.

We detected about 30% radioactivity in wall and cyclone ash. The recovery rate of nuclides which vaporize by incineration (^{14}C, ^{35}S) was lower than those of no vaporization. Almost all ^{14}C samples were emitted as gas. The recovery rate of ^{35}S was only 30%. These results were reproduced by independent experiments. We could not determine the cause of this low recovery, but we considered that ^{35}S in the gas form was re-adsorbed to the smoke duct and cyclone wall and could not be recovered. An example of adsorption of ^{35}S-compounds to the inner wall of the apparatus during dry-distillation of radioactive wastes is reported[6]. We obtained nearly the same result for the nuclides which did not vaporize (^{45}Ca, ^{33}P and ^{32}P)[2),3),7)]. We conclude that these results support the reliance of the experimental incineration system and the measurement method used in this study. Distribution of β-emitter shown in this study did not show the marked discrepancy in the results of our earlier experiments and in an incineration experiment of organic liquid wastes[7]. Therefore, we concluded that the measurement method used in this study was effective for further experiments. We also used plastic materials in this

study as incineration samples. About 5 kg of samples was reduced to less than 100 g of ash and dust. The reduction rate was less than 0.02% (1/5 000). We showed the effectiveness of incineration for weight reduction.

We conclude that the use of the Imaging Plates is a simple and reliable method for estimation of the radioactivity of low energy β-emitter in incineration gas and ash. There was no need to make samples for liquid scintillation counting which is a rather complicated process. We can process many samples at a time using Imaging Plates. We demonstrated that the counting efficiency by an Imaging Plate of low energy β-emitter is high as compared to that of the liquid scintillation counter and that the use of an Imaging Plate is ideal for measurement of ash and dust radioactive samples.

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