CO10-1 Application of KURAMA-II to Radiation Monitoring of Soil Separation Facilities in Fukushima Prefecture

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INTRODUCTION: KURAMA (Kyoto University RAdiation MApping system)-II is a radiation measurement system characterized by its compactness, autonomous operation, and acquisition of pulse-height spectrum data (Fig. 1) [1]. KURAMA-II measures ambient dose equivalent rate (hereafter referred to as air dose rate) and GPS position and automatically transmits them to a dedicated cloud server. We used a backpack-style KURAMA-II (Fig. 2) for the radiation monitoring of soil separation facilities in an interim storage facility to assess whether the radioactive material was scattered under demolition.

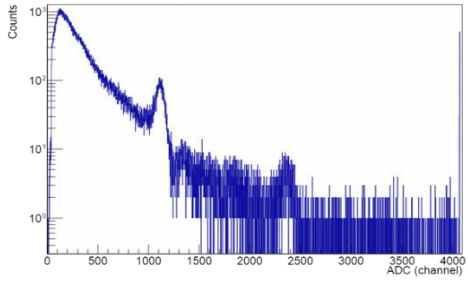


Fig. 1. A typical example of pulse-height spectrum obtained by KURAMA-II measurement.

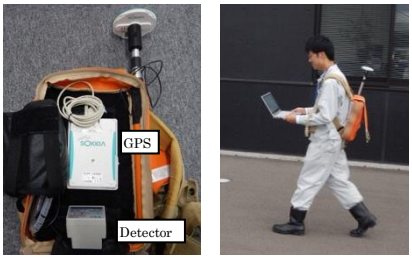


Fig. 2. KURAMA-II in a backpack.

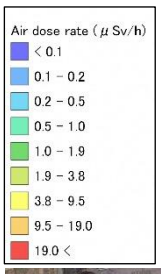
EXPERIMENTS: The air dose rates of two soil separation facilities (facility A and facility B) were measured on foot with a KURAMA-II in a backpack. The date of the measurement and the demolition period are shown in Table 1.

Table 1. The date of the measurement and the demolition period.

	Facility A	Facility B
Demolition	Apr. 2022 - Nov. 2022	Jun. 2022 - Oct. 2022
1st measurement	13 Jul. 2022	13 Jul. 2022
2nd measurement	16 Dec. 2022	16 Dec. 2022

A CsI (Tl) scintillation detector (C12137-4034, Hamamatsu Photonics) was used for measurement. The air dose rate and GPS position were measured every second. For 1st measurement, the air dose rate was measured by walking along the facility's boundary under demolition. After the demolition, 2nd measurement was carried out in the whole area of the facility. The measurement data were averaged in a 15-meter mesh and visualized to the colored air dose rate maps using GIS software (QGIS 3.20.3). Several points of the boundaries of facilities were measured by a NaI (Tl) scintillation survey meter.

RESULTS: The air dose rate maps are shown in Fig.3-6, and the ranges of the air dose rate are summarized in Table 2. No apparent differences were found between 1st and 2nd measurements at both facilities. KURAMA-II results were comparable to those of the survey meter. As shown in Fig.5 and 6, no apparent contamination was found in the study area. In conclusion, no clear effect of the demolition of the soil separation facilities on the air dose rate was found in the present study.



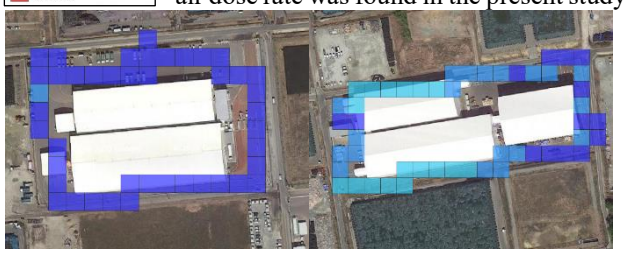


Fig. 3. 1st measurement at facility A.

Fig. 4. 1st measurement at facility B.

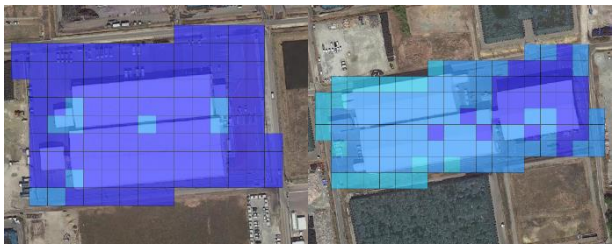


Fig. 5. 2nd measurement at facility A.

Fig. 6. 2nd measurement at facility B.

Table 2. The air dose rates measured by KURAMA-II and by survey meter (μSv/h). The range of KURAMA-II (1st) was calculated from the mesh measured in both measurements.

	Method	Facility A	Facility B
1st	KURAMA-II	0.06-0.11	0.07-0.40
	Survey meter	0.08-0.11	0.11-0.28
2nd	KURAMA-II	0.07-0.11	0.09-0.35
	Survey meter	0.08-0.12	0.10-0.33

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CO10-2 Effective Measures on Safety, Security, Hygiene and Disaster Prevention in Laboratories

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INTRODUCTION:

Important aspects of the study can be found in the following keywords, such as safety, security, hygiene and disaster prevention. Nuclear research reactor is one of representative facilities together with these keywords under their operation. It is effective to investigate the latest status on practical measures on these keywords in various facilities including nuclear research reactors, to compare each other among facilities, and to discuss more optimized ones for our positive safety management. Through this process, it is also essential to investigate the latest international and/or national regulations and the movement of revision of them. In addition, development of human resource and public literacy on nuclear science and technology is also within the scope of the research. The total discussion contents and their fruits are directly useful for all relating laboratories.

RESEARCH APPROACH:

General research approach is as follows.

- Measures of safety management during operation or standstill status of the real facilities would be investigated. This information would be used for our research discussion on the positive and more optimized safety management.
- It would not be a single year research, but maybe two to three years research for one theme.
- Information source of facilities would not be only KUR, KUCA or the other facilities in Kyoto University, but also the Kindai university research nuclear reactor or the facility of National Institute of Fusion Science, etc. This research is an active joint-research with these relating facilities and positive researchers on safety management.
- One of the distinctive features of this research is to involve office staffs as cooperators as well as researchers and technical staffs. In The University of Tokyo, most of the members in Division for Environment, Health and Safety are office staffs who knows real situation of safety management in laboratories very well.

Concrete discussion target in FY of 2022 was determined as the followings; “developing a set of educational videos for safety managers and researchers in universities using accelerators” and “analysis of effects on radiation education in secondary schools.

DEVELOPING EDUCATIONAL VIDEOS IN UNIV. FOR USERS OF ACCEARATORS:

A refresher training videos "Accelerator Safety Application for Radiation Workers in The University of Tokyo” were produced. With the cooperation of the managers and experts of the off-campus facilities (e.g. J-PARC and KEK), the contents were carefully examined and included features and safety points for each facility that should be communicated to radiation workers in advance, not only for radiation workers of the facilities at The University of Tokyo, but also for ones sent to the off-campus facilities.

The educational items were subdivided into the following six issues, which were presented as short videos of 5 to 8 minutes each, with a maximum overall duration of 45 minutes or less. These are (1) types and characteristics of accelerators, (2) safe handling of accelerators, (3) examples of accidents and troubles related to accelerators, (4) on-campus accelerator facility - MALT section, (5) domestic accelerator facilities - J -PARC, and (6) domestic accelerator facilities -KEK. Each facility section of (4) to (6) includes facility features, main research achievements, safety considerations and a message from the administrator.

ANALYSIS OF RADIATION EDUCATION EFFECTS IN SECONDARY SCHOOL

A questionnaire survey of secondary schools and their teachers in eight countries in the Asia-Pacific region, including Japan, was conducted from around the end of 2020 to the end of 2021. The survey asked about the teachers' values on radiation, an overview of the actual implementation of the lessons (e.g. the content of each item), and the students' impressions, knowledge and interest in radiation.

We analyzed the results of questionnaires on the effectiveness of radiation education in secondary schools. The degree to which students perceive radiation as ”Interesting” became stronger after the STEAM radiation lecture. Radiation education with the WOW factor is effective. In the analysis of changes in students’ impressions based on teacher characteristics, teachers who perceive radiation as ”Simple” or ”Easy to be understood” are more likely to teach radiation basics attractively. On the other hand, in the overall analysis, the degree to which students perceive radiation as ”Interesting” or ”Easy to understand” decreased when taught by the teacher who perceive it as “Good”. It is possible the students were influenced negatively by the teachers' strong impressions and messages.

CO10-3 Study of Penetration/Leaching Behavior from Concrete Contaminated with Cs -Analysis of changes in mortar structure accompanied by dry-wet cycles-

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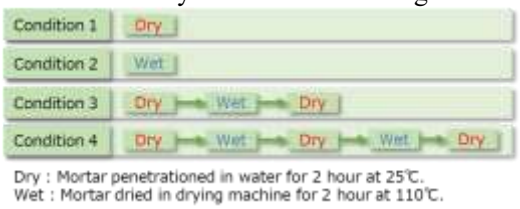
INTRODUCTION: Cesium (Cs) penetration/leaching experiments on concrete of reactor containment structural materials are contributed to understanding of contamination mechanism by radioactive materials, which is one of six important issues identified by the Nuclear Damage Compensation and Decommissioning Facilitation Corporation (NDF). The contaminated concretes are exposed to the environment changes, which means that Cs concentration behavior may changes.

EXPERIMENTS: Mortar specimens (without aggregates) were made with ordinary Portland cement (W/C=0.37, S/C=2.1) in a cubic of 15mm×15mm×15mm. They were embedded into acrylic resin, so that only one surface remains for penetration/leaching of Cs. The Cs penetration solution (15 ml) was CsOH solution adjusted to 10^{-2}M. Table 1 shows four dry and wet conditions given to mortars. For example, a penetration experiment using mortar given condition 1 was named P1 and a leaching experiment was named L1, and P1-P4 and L1-L4 were prepared. The mortars of P1-P4 were soaked in CsOH solution for 15 days for penetration. Mortars of L1-L4 were soaked in CsOH solution for 15 days for penetration and then in water for 15 days for leaching. After the experiments, the mortars were scraped with sandpaper (particle size: #60) to powder. The powders were analyzed by INAA to determine amounts of Cs remaining in them.

RESULTS&DISCUSSION: Fig.1 shows results of the penetration experiments. The number of times the mortar was dried is written after "d " in the figure, and the number of times it was wetted after "w". The penetration experiment results for P1-P4 showed the amount of Cs was greater at depth than at the surface. This is possibly due to the fact only the powder collected at a depth of 0.5 mm from the mortar surface had a higher percentage of acrylic resin, resulting in smaller amount of Cs. Fig.2 shows the results of the leaching experiments. In the leaching experiment CsOH solution in contact with the mortar surface would change from CsOH solution to water containing no Cs, resulting in a larger concentration gradient. If the migration of Cs was considered to proportional to the concentration gradient, it is assumed that Cs concentration decreases near the surface and is relatively higher inside, like a "mountain" distribution. Then, the Cs concentration distribution "flatten" by concentration gradient. The leaching experiments showed such a profile, but the position of the "mountain" differed depending on the number of dry/wet cycle. If the mortar given a dry/wet cycle is regarded to migrate Cs greater [1], leaching is more advanced in profiles L3 and L4 than in profiles L1 and L2. It suggests a change in the position of "mountain". As the cause of this penetration and leaching behavior, it is possible that the different number of dry/wet cycles may complicate the diffusion phenomenon by physically and chemically changing the mortar structure. Examples include capillary action and hydration reactions. It is thought Cs migration to be influenced not only by diffusion through water filled paths, but also by the dry/wet cycle and moisture conditions [2].

In the future, our team conduct the following to study the detailed concrete structure and Cs penetration/leaching behavior. (a) Simulation and calculation of Cs permeation/leaching behavior (FEM analysis).
(b) Estimation of Cs and major elements (Fe, Ca, Si, Al) of concrete by setting irradiation time and decay time in detail with INAA. (c) Investigate microstructure by pouring fluorescent paint into concrete.

Table.1. Four dry and wet conditions given to the mortar.



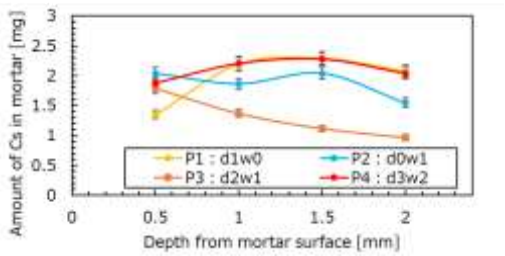


Fig.1. Amount of Cs present at each mortar depth in the penetration experiment.

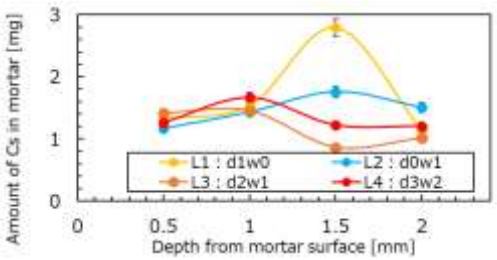


Fig.2. Amount of Cs present at each mortar depth in the leaching experiment.

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CO10-4 Modeling of slope-directed migration of Cs in forest soils

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INTRODUCTION: A previous study on quantitative evaluation of air dose rates in forest was conducted by Miyata et al.[1] for the evaluation of the effectiveness of decontamination in a forest using the point-decay nuclear integration method. In this calculation, the distribution of Cs concentration in soil in the forest was assumed to be the same in all locations, and the distribution in the direction of the slope was not considered. As a result, the calculated air dose rates underestimated the measured ones, especially at the monitoring points located at the bottom of the slope, and the error of air dose rate between the calculated and measured values was large. In this study, we developed a model to estimate the slope directional distribution of radioactive Cs in forest soils, and validated it by comparing it with the measured air dose rate.

MODEL FOR SLOPE MOVEMENT OF Cs: Advection of Cs-sorbed soil particles by subsoil flow was considered as a mechanism that causes the distribution of radioactive Cs concentrations in forest soils in the slope direction. In this study, the slope is divided into n compartments, and Cs in soil particles is considered to move from the top to the bottom of the slope due to the movement of soil particles from the solid phase to the liquid phase caused by rainfall and the movement of water from the top to the bottom of the slope. We then construct a simultaneous ordinary differential equation that shows the amount of change in radioactivity per unit time in each compartment due to advection. By solving the equations, the Cs concentration in the soil at the end of rainfall is calculated. Then, assuming that all soil particles in the water return to the solid phase after the end of rainfall, we calculate the distribution of Cs concentration in the soil at the desired date and time by repeating the simulation of multiple rainfall events. The amount of Cs transferred per unit time into the water in the i-th compartment due to the transfer of soil particles from the solid phase to the liquid phase, P_i, is calculated by the following equation:

$$P_i = \alpha (1-\phi) DL\cos\theta \cdot \rho S_i^{o} e^{-\alpha t} \quad (1)$$

where, S_i^{o}:Radioactivity concentration in unit soil mass in the i-th compartment at the start of rainfall; t:Time since the start of rainfall; D:Cross-sectional area of the x-sectional slope; L:Slope length of each compartment, ϕ:Porosity of soil, ρ :Density of soil solid phase; θ : Maximum slope angle; α :Coefficient of radioactive runoff. α is a coefficient that expresses how much of the amount of radioactivity in a unit volume is flown up into the flowing water in a unit time, and is calculated by the following formula.

$$\alpha = (E_s S_m)/((1-\phi)\rho S_x) \quad (2)$$

where, α :Coefficient of radioactive runoff (1/s); E_s: Amount of soil flow per unit volume and per unit time ($g/m^3/s$); S_m:Radioactivity concentration in unit mass of soil which flowed away (Bq/g); S_x:Radioactivity concentration in unit mass of soil before the flow (Bq/g); $(1-\phi)\rho$:Apparent density of soil (g/m^3).

The unit time transfer of Cs from the i-th compartment to the i+1-th compartment due to the movement of water from the top to the bottom of the slope, $A_{i,i+1}$, is calculated by the following formula:

$$A_{i,i+1} = \phi \gamma C_i q_i D \quad (3)$$

where, C_i:Cs concentration in water in the i-th compartment; q_i: Flow rate per unit area in the i-th compartment; D:Cross-sectional area of slope in x-section; Φ:Porosity of soil; γ :Coefficient of velocity of particle movement relative to water.

The flow rate q_i in the i-th compartment due to rainfall shown in equation (3) is assumed to be calculated by the following equation:

$$q_i = (p' x_i)/d \quad (4)$$

where, q_i:Flow rate per unit area in the i-th compartment; x_i:x-coordinate of the center of the i-th compartment; d:Slope depth at which soil particle movement occurs due to rainfall; p':(Daily precipitation during rainfall) - (average daily evaporation).

As shown in equation (4), the flow rate q_i is assumed to increase in proportion to the distance x from the top of the slope, but we assumed that the flow rate q_i reach maximum value at about 40m from the top of the slope. The slope depth d, at which soil particles move due to rainfall, is set to 10 cm in this study.

RESULTS AND DISCUSSION: The soil runoff E_s in equation (2) was measured in an actual forest. Other parameters and rainfall values were obtained from the model of Miyata et al. The calculated values were compared with the distribution of Cs concentration in soil obtained by fitting the measured values of air dose rate, assuming a linear change of Cs concentration in soil in the slope direction. As a result, the distribution of Cs concentration in soil was significantly different from the distribution of Cs concentration obtained by fitting to air dose rates. The value of α that produces a distribution similar to the Cs concentration distribution in soil obtained by fitting to the measured air dose rates was 1200 times larger than the value of α determined from the measured soil flux. One of the reason for the much larger value of α was thought that Cs was more abundant near the ground surface immediately after the nuclear power plant accident, and the amount of Cs migration due to slope movement of soil particles caused by rainfall may have been much larger than the average soil runoff velocity obtained in this study.

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CO10-5 Geological Standard Samples for Elemental Analysis of Accelerator Concrete

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INTRODUCTION: Concrete, which constitutes the building of accelerator facilities, accounts for the almost mass in the facility. Therefore, whether the concrete is activated or not will affect the amount of radioactive waste generated, which will become apparent as a cost when the facility is decommissioned. Especially, long-lived nuclides of ^{152}Eu and ^{60}Co would become problematic as waste. We have investigated the elemental concentrations of natural Eu and Co in concrete from various accelerator facilities in Japan, the origin of ^{152}Eu and ^{60}Co, by neutron activation analysis [1]. The elemental concentrations have been determined as relative values using two types of igneous rock standards, JA-1 and JG-3. Whereas concrete is a mixture of cement and some aggregates, and it might contain various materials other than igneous rock. To further discuss the elemental composition of accelerator concrete, we have irradiated various geological standard samples such as sedimentary rock, sand, and fly ash and compared the irradiation results with those of concrete.

EXPERIMENTS: The irradiated samples are as follows. Seven igneous rocks: JA-1, JB-1b, JG-1, JGb-1, JGb-2, JR-1, JR-2. Two sedimentary rocks: JSl-1, JSl-2. Three river and lake sediments: JLk-1, JSd-2, JSd-3. Three soil and other samples: JSO-1, JCFA-1, JMn-1. These are summarized in Table 1. All samples were weighed 100 mg of each after drying, then sealed with double-layered polyethylene films. Neutron irradiations to the samples were performed in February 2023 in the pneumatic transport tubes (Pn-2 and Pn-3) at KUR. After 10 seconds of irradiation with Pn-3 at a thermal power of 1 MW, γ-ray spectrometry was performed with a Ge detector within 5 minutes. The same samples were then irradiated with Pn-2 at a thermal power of 5 MW for 50 minutes. After 10 days of irradiation, all samples were measured with a Ge detector within 20 minutes.

RESULTS: The γ-ray spectrometry results for Pn-3 irradiated samples (1MW-10s) showed that the peaks of ^{24}Na, ^{28}Al, ^{42}K, ^{49}Ca, ^{51}Ti, ^{52}V, and ^{56}Mn were identified. Although the results of Pn-2 irradiated samples (5MW-50min) are still under analysis, we were able to identify ^{82}Br (half-life: 35.3h), ^{140}La (half-life: 40.3h), ^{153}Sm (half-life: 46.3h), and other nuclides with half-lives of less than 2 days. These rare earth and halogen elements could not be determined in our previous experiments due to their difficulty in γ-ray spectrometry. It is suggested that the determination of these elements is possible even in concrete samples.

Table. 1. Representative geological standard samples for reference of elemental concentration analysis of accelerator concrete materials [2-5].

Name	Kinds of rocks	Samping place
	Igneous rock	
JA-1	Andesite	Hakone volcano
JB-1b	Basalt	Kitamatsuura basalt
JG-1	Granodiorite	Sori granodiorite
JGb-1	Gabbro	Utsushigatake
JGb-2	Gabbro	Tsukuba-san leucogabbro
JR-1	Rhyolite	Wada Toge obsidian
JR-2	Rhyolite	Wada Toge obsidian
	Sedimentary rock	
JSl-1	Slate	Toyoma clay slate
JSl-2	Slate	Toyoma clay slate
	Sediments	
JLk-1	Lake sediment	Lake Biwa
JSd-2	Stream sediment	Eastern region, Ibaraki
JSd-3	Stream sediment	Central region, Ibaraki
	Soil	
JSO-1	Soil	Machida, Tokyo
	Coal fly ash	
JCFA-1	Coal fly ash	Isogo, Yokohama
	Ore	
JMn-1	Manganese Nodule	Southern Central Pacific Basin

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CO10-6 Particle Size Measurement for Aerosol Particles Generated from Molten Gold Using a High-Frequency Induction Furnace System

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INTRODUCTION:

Recently high intensity accelerators have been developed for medical application and isotope production. Target melting accidents, such as the J-PARC accident in 2013, could occur by mishandling of a high-intensity beam. Various radionuclides were released from a molten radioactive gold target in the J-PARC accident. The characteristics of the released radionuclides were very important to estimate both external and internal doses. The radioactive aerosol particles are formed by incorporation of radioactive nuclides into the aerosol particles formed through condensation process of evaporated target metal material. Particle size of the radioactive aerosol particles gives us useful information on the formation mechanism. To simulate the target accidents it is necessary to develop a high temperature furnace dedicated to aerosol sampling.

A high-frequency induction furnace system has been developed [1] for collection and size analysis of radioactive aerosol particles formed from molten radioactive metal samples. In this work, particle size was studied for gold aerosol particles formed from molten gold using the furnace system/

EXPERIMENTS:

Furnace system: Metal samples were heated using the high-frequency induction furnace system which was specially designed for aerosol collection. The furnace system consists of the induction quartz tube furnace and a low-pressure impactor for particle size analysis (Fig. 1). Highly pure carbon (graphite) crucibles were used for induction heating in the furnace. A disadvantage of induction heating in aerosol experiments is that high concentration fine particles are often produced from heated material. In this experiment, emission of the fine particles was successfully suppressed by long preheating of the crucibles under vacuum and in pure argon.

Heating of samples and collection of aerosols: Granular gold samples were heated in the preconditioned carbon crucible in the furnace up to 1,750 ºC in a flow of highly pure argon. The generated aerosols were introduced to the impactor by the argon flow. The impactor used was a so-called "jack-up" or "drawer" type low pressure impactor. It consists of 13 collection stages and a back-up filter. Teflon-binder glass fiber filter was used as the collection substrate.

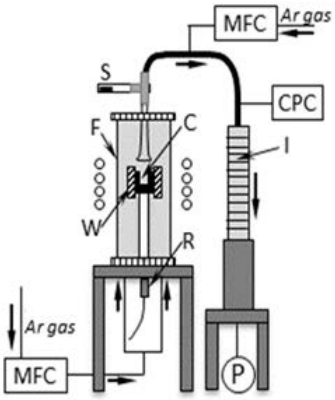

Fig.1 High Frequency Induction Furnace System

Particle size analysis: Weight of gold aerosol particles collected on each collection stage of the impactor was determined by activation analysis. Small piece of the collection substrate containing five spots of collected particles were cut from the substrate filter of each collection stage. The pieces were subjected to neutron irradiation in the Pn-1 pneumatic tube of the Kyoto University Research Reactor (KUR). The activity of ^{198}Au was measured with Ge semiconductor detectors.

RESULTS AND DISCUSSION:

Neutron activation analysis was adopted for determination of gold because the dispersion ratio from molten gold was estimated to be very small at 1,750 ºC. Weight of gold of each collection stage was determined with high accuracy. Mass medium aerodynamic diameter (MMAD) was found to be approximately 1 μm. The MMAD was almost the same as that of aluminum [2] although their vapor pressure is quite different.

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