Radioactive Cs Behavioral Analysis in the Incinerator at Garbage Incineration – 15031

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ABSTRACT
About 4.7 kg cesium 137(Cs137) and about 0.3 kg cesium 134(Cs134) were released to the atmosphere around FUKUSHIMA Prefecture by the accident of the Tokyo Electric Power Company Fukushima Daiichi nuclear power plant accompanying the Great Earthquake in East Japan, which occurred on March 11, 2011. In the general incineration system which a local government owns, the garbage to which radioactive cesium (Cs) adhered (less than 8000Bq/kg) is incinerate usual time. Before incinerating the garbage to which radioactive Cs has adhered, we did the preliminary test.
As a result, if a very special case is removed, it is checked by measurement that radioactive Cs is not released from an exhaust stack of general incineration system. Our research is the elucidation of the mechanism by which radioactive Cs contained in garbage is not again released to the atmosphere with combustion gas. Concretely, we focus on the agglomeration of Cs molecular and deposition of radioactive Cs particles to the ash particle that occurs at the time of refuse incineration. And we formulated the simulation model for analyzing this phenomenon. It is presumed from an analysis result that radioactive Cs is not re-releasing by the following reasons. First, radioactive cesium hydroxide (Cs (OH)) adhering to garbage changes to radioactive cesium oxide (CsO2) by an incineration process. Furthermore, it changes to radioactive cesium chloride (CsCl) via Cs.
CsCl and Cs (OH) molecules deposit to the surrounding incineration ashes (main ashes and fly ash), in form of agglomerations. About fly ash, agglomeration and deposition are occurrence from a combustion chamber to bag filter via flue and temperature reducing tower.
All radioactive Cs deposits to fly ash and main ash particles in this process. Therefore, radioactive Cs is not re-released to the atmosphere with combustion gas. Also about the main ashes, the same phenomenon occurs.

INTRODUCTION
About 4.7 kg Cs137 and about 0.3 kg Cs134 were released to the atmosphere around FUKUSHIMA prefecture by the accident of the Tokyo Electric Power Company Fukushima Daiichi nuclear power plant accompanying the Great Earthquake in East Japan, which occurred on March 11, 2011.The thesis's concerned estimation that the chemical formation of radioactive Cs emitted to the atmosphere is Cs (OH). Moreover, Cs (OH) was contained in sulfate aerosol, and was transportation around Fukushima Prefecture. Transported Cs (OH) fell on the ground, and adhered to various things. In the general incineration system which a local government owns, the garbage to which radioactive Cs adhered (less than 8000Bq/kg) is incinerate usual time. In this situation, , a possibility that radioactive Cs would be again released to the atmosphere with the combustion gas discharged from an exhaust stack was pointed out by some residents. Before incinerating the garbage to which radioactive Cs has adhered, we did the preliminary test.
As a result, if a very special case is removed, it is checked by measurement that radioactive Cs is not released from an exhaust stack of general incineration system. Our research is the elucidation of the

mechanism by which radioactive Cs contained in garbage is not again released to the atmosphere with combustion gas. Concretely, we focus on the agglomeration of radioactive Cs molecular and deposition of radioactive Cs particles to the ash particle that occurs at the time of refuse incineration. And we formulated the simulation model for analyzing this phenomenon. It is presumed from an analysis result that radioactive Cs is not re-releasing by the following reasons. First, Cs (OH) adhering to garbage changes to radioactive CsO2 by an incineration process. Furthermore, it changes to CsCl or via radioactive Cs. The CsCl and Cs (OH) molecules deposit to the surrounding incineration ashes (main ash and fly ash), building agglomerations. Agglomeration and deposition are occurrence from a combustion chamber to bag filter via flue and temperature reducing tower.
In this process all radioactive Cs deposits to fly ash particles and main ash particles. We are going to conduct analysis that made incineration temperature and radioactive Cs concentration in garbage the parameter using this simulation. In addition, about the validity of a simulation, it is checking using operation data collected in the general incineration system in Fukushima. In this presentation, simulation models and operation data such as distribution of particle diameter and density of radioactive Cs collected in the general incineration system in Fukushima Prefecture are introduced.

INVESTIGATION OF THE ASHES

We conducted investigation of an operating condition and incineration ashes for the purpose of the construction of an analysis model. Our investigation was conducted with three facilities in Fukushima Prefecture where incineration scales differ. The investigated items are the following three points.

SUBJECT OF INVESTIGATION

Incinerator A: Stoker type incinerator Scale 110ton/day
Incinerator B: Stoker type incinerator Scale 35ton/day
Incinerator C: Stoker type incinerator Scale 40ton/day

INVESTIGATED ITEMS

Particle size distribution (main ashes and fly ash)
Specific surface area (main ashes and fly ash every particle size)
Total radioactivity (main ashes and fly ash every particle size)

Particle size distribution

The vibrational sieving machine (SW20) shown in the photograph 1 was used for measurement of particle size distribution. The minimum particle diameter in which size classification is possible is 10μm.

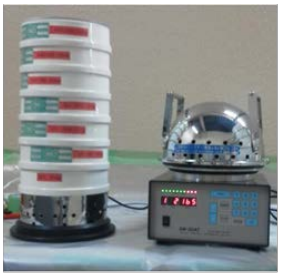


Photo.1 SW20

As a typical example, the photograph 2 shows the size classification result of incinerator A.
In addition, the sampling quantity of incineration ashes is 50 cc. This result showed the following as a feature of particle size distribution.

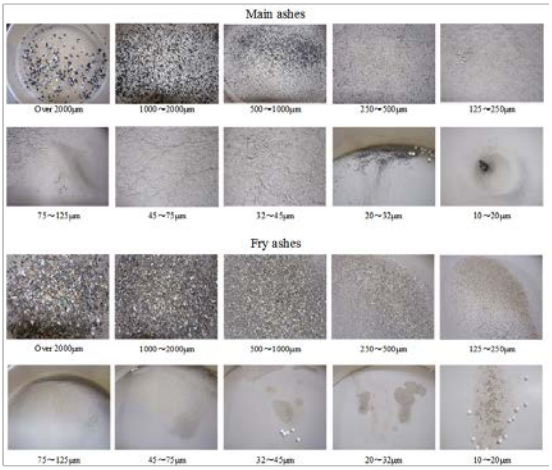


Photo.2 size classification result of incinerator A

Fly ashes, as Fig. 1 shows it, have logarithm normal distribution and average size of approximately 100μm

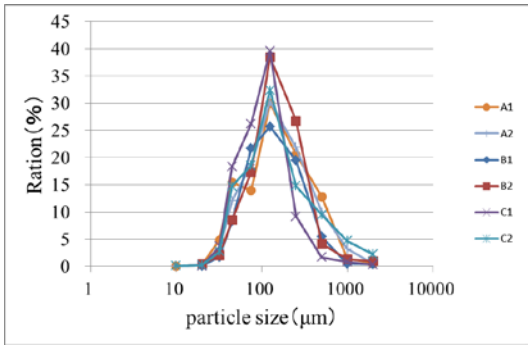


Fig.1 Particle size distribution of fly ash

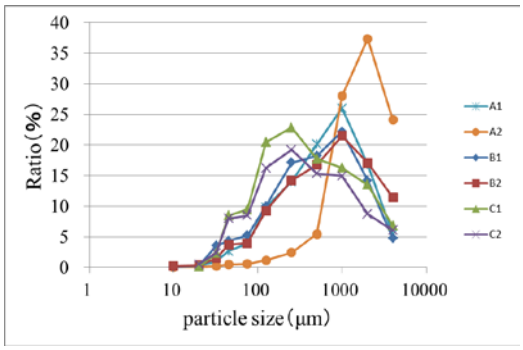


Fig.2 Particle size distribution of main ash

Main ashes (Fig.2) have a difference in particle size distribution related to the incineration system and sampling time. It is presumed that the factor is the difference in garbage composition.

Specific surface area

The specific surface area Gas adsorption (NOVA 4000e) shown in the photograph 3 was used for measurement of specific surface area. We evaluated the surface area per unit volume by the specific surface area measurement result. This result showed the following as a feature of specific surface area.

Photo.3 NOVA 4000e

Fly ashes (Fig. 3) show no correlation between particle diameter and surface area. It is presumed from this data that fly ashes are not simple globule.
Main ashes particle diameter and surface area are correlated. However, as figure 4 shows that a tendency changes with incineration conditions. It is presumed that this factor is the difference in garbage composition.

Total radioactivity

The radioactivity measurement equipment (FD-08Cs) shown in the photograph 4 was used for measurement of total radioactivity. We evaluated the radioactivity per unit volume using radioactivity measurement data. This result showed the following as a feature of radioactivity on fry ashes and main ashes.

Photo.4 FD-08Cs

Fly ashes Fig.5 shows that radioactivity and particle diameter are correlated. The radioactivity of the small fly ashes of particle diameter becomes high. And, this tendency is related also to the absolute value of radioactivity.
Main ashes Fig.6 shows that radioactivity and particle diameter are correlated. The radioactivity of the small fly ashes of particle diameter becomes high. And, this tendency is related also to the absolute value of radioactivity, too.

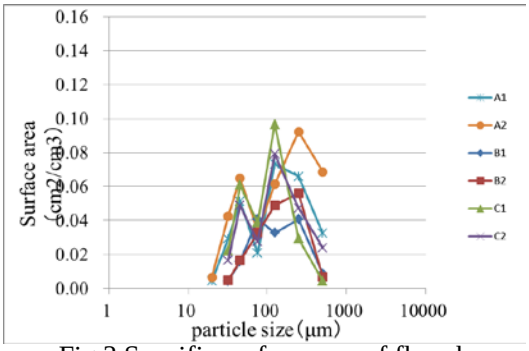


Fig.3 Specific surface area of fly ash

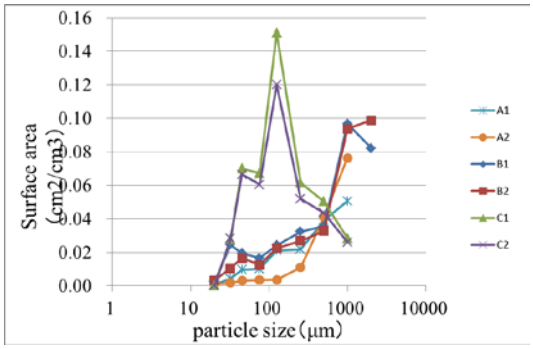


Fig.4 Specific surface area of main ash

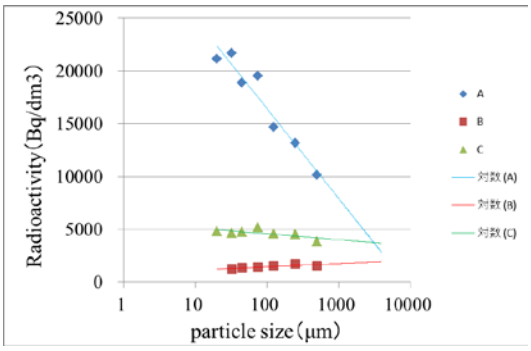


Fig.5 Total radioactivity of fly ash

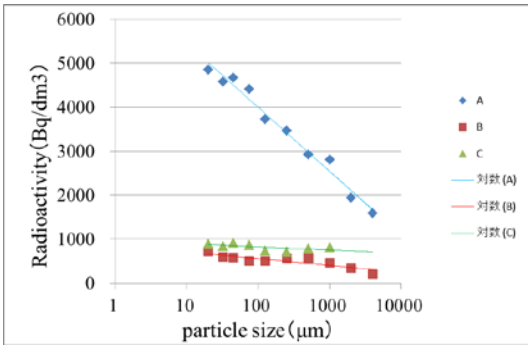


Fig.6 Total radioactivity of main ash

Consideration of the result

We performed consideration from the viewpoint of the physical model construction about the interaction of radioactive Cs and the incineration ashes based on particle size distribution, specific surface area and a total radioactivity measurement result used real ashes. A typical consideration result is shown below.

Consideration1

The surface observation by a scanning electron microscope shows that an incineration ashes particle is porous. Fig.7 and Fig.8 shows that the total radioactivity of one ash particle can be approximated by 2.7 to 3.0 power of particle diameter. It is presumed that radioactive Cs is contained from this result to the inside of an incineration ashes particle. Furthermore, it turns out that radioactive Cs deposition is dependent on ashes surface area.

Consideration2

The surface area of the ash particle to which any facility of incinerator A, incinerator B, and incinerator C exists per 1 cm^3 of combustion gas is presumed to be 0.15 to 0.45 cm^2.

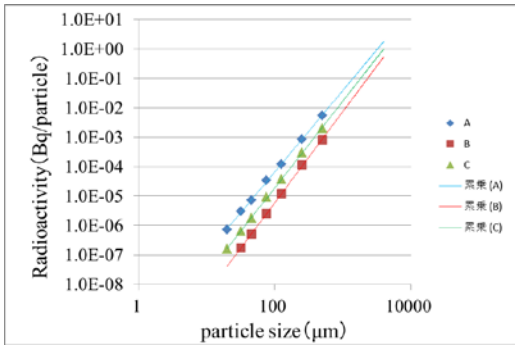


Fig.7 Radioactivity of one particle (fry ash)

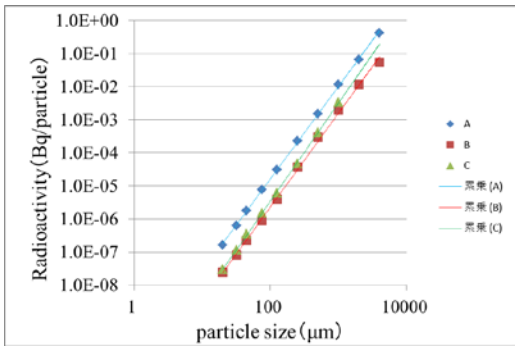


Fig.8 Radioactivity of one particle (main ash)

ANALYSIS MODEL

The analysis model assumed in our simulation code is as follows.

The chemical composition of radioactive Cs emitted by the Fukushima Daiichi nuclear power plant accident into the atmosphere is presumed to be Cs (OH). And Cs (OH) was diffused in the living sphere by sulfate aerosol. Cs(OH) adhering to plants and garbage is about not less than 700 °C, and is decomposed into Cs element via Cs_2O. Cs is combined with the chlorine contained in garbage. As a result, CsCl was generated. CsCl is condensed less than 1300 °C. And it moves in the inside incinerator, depositing to surrounding incineration ashes.

By the based on this assumption, the thermal field, flow field, chemical-species filed in an incineration

system and the amount of generation for every particle diameter of the main ashes and fly ash are calculated by combustion analyses. Also transport calculation within the incineration system of incineration ashes is performed. Naturally, this calculation is performed for every particle. And in this simulation code, it is calculated how incineration ashes are transported inside incineration system. The DEM (Distinct Element Method) is used for these analyses.

And then, Cs (OH) adhering in garbage is about not less than 700 °C, and is decomposed into Cs element via Cs_2O. Cs is combined with the chlorine contained in garbage. As a result, CsCl was generated. CsCl is condensed less than 1300 °C. CsCl is less than 1300 °C and produces condensation according to the material balance formula (formula 1) depending on density of number of CsCl molecule. Furthermore, if CsCl single molecule gas and a CsCl aggregate with an incineration ashes particle, they will adherence on the surface of incineration ashes particle according to the formula 2.

$$\frac{dn_k}{dt} = \frac{1}{2}\sum_{\substack{i=1 \\ i+j=k}}^{i=k-1} J_{ij} - \sum_{i=1}^{\infty} J_{ik} \qquad J_{ij} = \beta n_i n_j$$

n_i: Density of number of i degree agglomerates (m^{-3})
n_j: Density of number of j degree agglomerates (m^{-3})
n_k: Density of number of k degree agglomerates (m^{-3})
β : rate of condensation function ($m^3 s^{-1}$)

$$\frac{dC_m}{dt} = -C_2 \sum_{i=1}^{i=N} (\eta\, n_i m_i (\frac{A}{V})) \qquad \eta = 1 - \exp(-0.3 \times i)$$

A: Total surface area of the ash in unit space (m^2)
C_m: Cs weight in ashes particle (kg_Cs/kg_ashes)
C_2: adjustment factor (-)
I: i degree Cs agglomerates
m_i: Mass of i degree agglomerates (kg)
N_i: Density of number of i degree agglomerates (m^{-3})
V: Volume of unit space (m^3)

In addition, in our simulation code, it is not taking into consideration about chemical composition of radioactive Cs.
According to literature etc., Cs adhering to soil is heated and generates the nonvolatile cesium alumina silicate ($CsAlSiO_4$). And it is presumed that almost all $CsAlSiO_4$ is contained in the main ashes.
On the other hand, the present target of our research is investigation of the mechanism to which radioactive Cs is not re-diffusing from the exhaust column of an incineration system.
Therefore, in our analysis model, it was considered as the model that all the radioactive Cs that is in the severest state evaporates. Therefore, our analysis model was taken as the model that all the radioactive Cs evaporates.
Moreover, the analysis of amount of adhesion of radioactive Cs and incineration ashes to inside of the incinerator, such as firebrick and a flue cannot do the present model.

ANALYSIS RESULT

Flow velocity distribution, temperature distribution, radioactive single molecule gas distribution and radioactive Cs concentration distribution on incineration ashes were shown in Fig.9 as a typical example of condensation and adhesion analysis of radioactive Cs and burn analysis. In addition, an analysis facility is Incinerator A in FUKUSHIMA.
The range of analysis was charging port for incineration chamber to bag filter via flue and gas cooling tower. Moreover, the analysis conditions are as follows.

Air inflow amount: 13370Nm^3/hr.
Air temperature: 125 °C
Recirculation exhaust gas flow rate: 3000Nm^3/hr.
Gas temperature: 155 °C
Reference diameter: 0.08m
Diameter of a particle after perfect combustion:
1-6000μm(Continues distribution)
Grain density: 378kg/m^3
Charge speed: 100ton/day
Radioactive Cs density:
1800 Bq/Cs137/kg_garbage

The simulation code which we built can estimate moisture distribution, carbon dioxide distribution, methane distribution, the grain size distribution of an incineration ashes, and incineration ashes particle temperature in addition to the above.

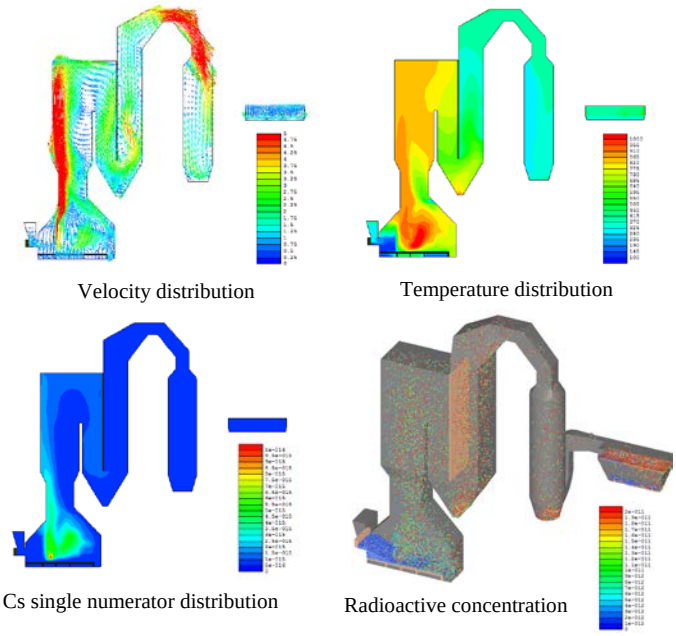


Fig.9 typical example of condensation and adhesion analysis

VALIDITY OF ANALYSIS RESULT

Validity of the analysis result was performed by comparing with the amount of generation main ashes and fly ash and particle size distribution, specific surface area, radioactivity measurement data, temperature distribution data in a furnace using incinerator A.
A verification result is shown in Table 1 and Table 2. Operation data and analysis result of the generating rate of the main ashes and fly ash correspond very well. On the other hand, there is a difference of approximately 2 times in quantity of incineration ashes. We think that these causes are the differences between garbage composition and specific burn-up.
In radioactive Cs behavior analysis, radioactive Cs concentration in the incineration gas at before bag filter was 1 Bq/m^3.
This result is in agreement with the field survey radioactive Cs which is all most all cases and is contained in an exhaust gas is below a minimum-limit-of-detection value.

Table1 Combustion analysis result and operation data comparison

	Main ash	Fry ash	Total ash	Ratio of ash
Operation data	7.60E-01	2.40E-01	1	0.08
Analysis result	7.40E-01	2.60E-01	1	0.04

Table2 Adhesion ratio of radioactiv Cs

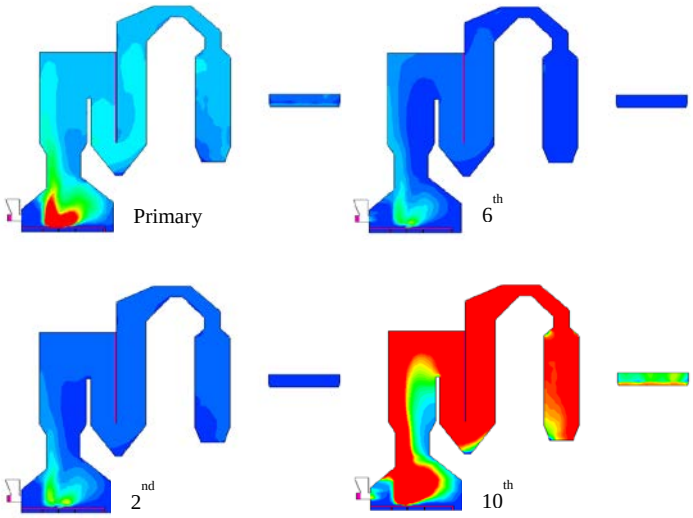
	Cs abundance		
	Combustion chamber	Befor bug filter	Adhesion ratio
	(Bq_Cs137/m3_gas)	(Bq_Cs137/m4_gas)	(%)
Analysis result	404	1	99.7

SIMULATION

We made simulations with 10 times and 100 times. increased radioactive Cs concentration in garbage We performed the simulations focusing to the concentration distribution of CsCl single molecule gas. This is because it assumes that single molecule gas has the low adhesion efficiency to incineration ashes particle. We think, the radioactive Cs concentration that can be incinerated can be presumed by our simulation. The radioactive Cs concentration in the garbage used for the simulation is as follows.

Case1: Typical survey data 1800(Bq_Cs137/kg_garbage)
Case2: 18000(Bq_Cs137/kg_garbage)
Case3: 180000(Bq_Cs137/kg_garbage)

A typical simulation result is shown in Fig.10 and Fig.11.
Fig.10 shows the condensation degrees (CsCl number of molecules) 1, 2, 6 and 10th radioactive Cs concentration distribution of case3. Moreover, the distribution inside incinerator of CsCl single molecule gas was shown in Fig.11. The following scientific knowledge is presumed from this result.



Knowledge1: In the case where radioactive Cs concentration contained in garbage is high, the condensation rate in the space from a combustion chamber to a bag filter becomes relatively high.

Knowledge2: In the case where radioactive Cs concentration in garbage is high, the CsCl single molecule gas concentration of before bag filter becomes relatively high.

Knowledge3: In the case where radioactive Cs concentration in garbage is high, radioactive Cs adhesion efficiency to incineration ashes becomes relatively high.

Fig.10 concentration distribution case3

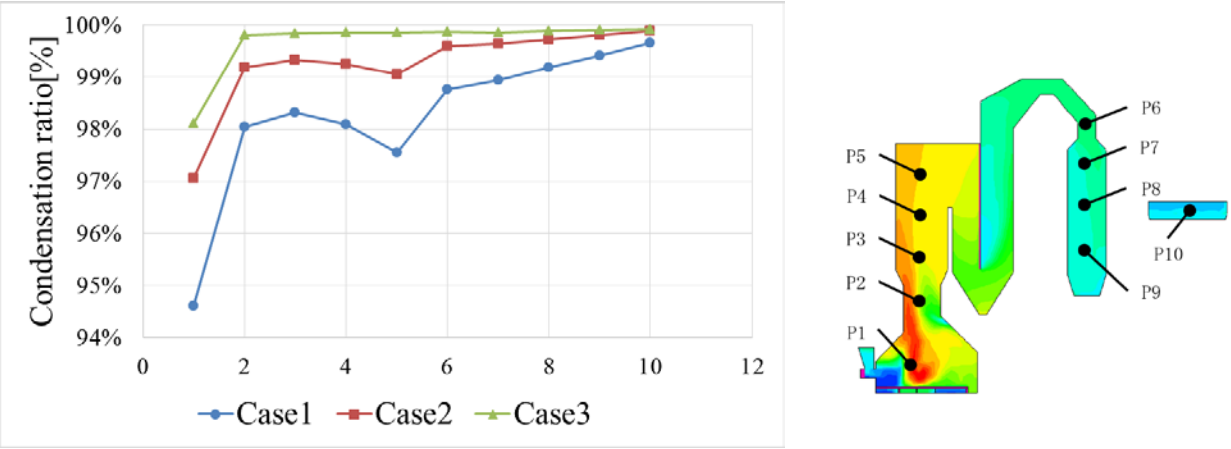


Fig.11 distribution inside incinerator of CsCl single molecule

CONCLUSIONS

We constructed the analysis code which combined combustion analysis and radioactive Cs condensation and adhesion behavior analysis.
Moreover, validity was verified using the incineration ashes and performance data that were extracted from the garbage incineration system in Fukushima.

The reliability of combustion analysis is very high. On the other hand, about Cs concentration distribution of inside incinerator, an improvement is required. We will examine the following points from now on.

Constructions of the analysis model in consideration of nonradioactive Cs for make the actual incineration conditions.

- Analysis method of nonvolatile chemical composition like as $CsAlSiO_4$.
- Review of sticking probability depends on speed (temperature) and mass in case CsCl collides with an ash particle.
- Model construction of the CsCl single molecule gas collection efficiency of a bag filter

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