Innovative Alpha Radioactivity Monitor for Clearance Level Inspection Based on Ionized Air Transport Technology

CFD-Simulated and Experimental Ion Transport Efficiencies for Uranium-Attached Pipes*

Yosuke HIRATA**, Katsuhiko NAKAHARA**, Akira SANO**, Mitsuyoshi SATO***, Yoshio AOYAMA****, Yasuaki MIYAMOTO*****
Hiromi YAMAGUCHI******, Kenichi NANBU††, Hiroyuki TAKAHASHI††† and Akinori ODA†††

**Power and Industrial Systems R&D Center, Toshiba Corporation
8 Shinsugita-cho Isogo-ku Yokohama 235-8523 JAPAN
E-mail: yosuke.hirata@toshiba.co.jp

***Isogo Nuclear Engineering Center, Toshiba Corporation
8 Shinsugita-cho Isogo-ku Yokohama 235-8523 JAPAN

**** Japan Atomic Energy Agency
4-33 Muramatsu Tokai-mura Ibaraki 319-1194 JAPAN

†Tohoku University
2-1-1 Katahira Aoba-ku Sendai 980-8577 JAPAN

††The University of Tokyo
7-3-1 Hongo Bunkyo-ku Tokyo 113-8656 JAPAN

†††Nagoya Institute of Technology
Gokiso-cho Showa-ku Nagoya 466-8555 JAPAN

Abstract
An innovative alpha radioactivity monitor for clearance level inspection has been developed. This apparatus measures an ion current resulting from air ionization by alpha particles. Ions generated in a measurement chamber of about 1 m³ in volume are transported by airflow to a sensor and measured. This paper presents computational estimation of the ion transport efficiencies for two pipes with different lengths, the inner surfaces of which were covered with a thin layer of uranium. These ion transport efficiencies were compared with those experimentally obtained for the purpose of validating our model. Good agreement was observed between transport efficiencies from simulations and those experimentally estimated. Dependence of the transport efficiencies on the region of uranium coverage was also examined, based on which such characteristics of ion currents as anticipated errors arising from unknown contaminated positions are also discussed to clarify the effective operation conditions of this monitor.

Key words: Clearance Level, Radioactive Waste, Alpha Radioactivity, Uranium, Ionized Air Transportation Technology, CFD, Simulation

1. Introduction
Wastes containing uranium have been generated and accumulated in nuclear facilities and the disposal of them has been of concern. Accordingly, various studies on the
clearance and disposal of those wastes are in progress (1). Accurate determination of the radiation level is required at the time of clearance-level inspection; however, direct measurement by hand using survey meters is extremely time-consuming because of the accumulation of huge amounts of wastes, especially in the case of contamination in narrow spots.

We have been developing an innovative method for quickly measuring alpha radioactivity, in which ions generated from air by alpha radiation are detected instead of alpha rays directly (2)(3)(4)(5)(6)(7)(8)(9). The concept of the measurement is schematized in Fig. 1 (10). Alpha rays emitted from a waste ionize the air in the measurement chamber. These generated ions are transported with airflow to the ion sensor, in which an electric field is applied between the electrodes to capture the ions. The alpha radioactivity, considered to be proportional to the rate of ion generation in the measurement chamber, is thus measured indirectly in the form of ion currents.

The relation between the sensor current, \( I_s \), and the alpha radioactivity, \( \omega \), is given by

\[
I_s = \omega E_\alpha e \zeta_{\text{surf}} \zeta \eta_{\text{self}} \eta_{\text{trans}} \eta_{\text{sensor}} / w.
\]

Conversion from the ion current to the alpha radioactivity of the waste, thus, requires factors presented in Eq. (1): \( E_\alpha \), the average energy of alpha particles; \( e \), the elementary charge; \( w \), the W value of the air; \( \zeta_{\text{surf}} = 0.5 \), the efficiency of the radioactive source arising from the surface effect that 50% of the alpha rays can come out into the air; \( \zeta \), the volume fraction of ionization due to the space limitation in a pipe, etc.; \( \eta_{\text{self}} \), the effect of self-screening; \( \eta_{\text{trans}} \), the efficiency of ion transport between the measurement chamber and the ion sensor; and \( \eta_{\text{sensor}} \), the sensor efficiency. Among these factors, \( E_\alpha \), \( e \) and \( w \) are available in literature (5)(9). The sensor efficiency, \( \eta_{\text{sensor}} \), is mainly a function of airflow speed into the sensor and can be determined theoretically. The ion transport efficiency, on the other hand, varies depending on the airflow speed in the measurement chamber and the shape of the wastes, etc. Accurate estimation of the alpha radioactivity, therefore, requires information about the ion transport efficiency. And this efficiency should be high enough to attain enough ion currents to measure.

When this type of monitor is applied to practical use, the size or the shape of the chamber, as well as the operation condition, should be optimized depending on the group of wastes to inspect. This requires a proper method to design the system. Our development, therefore, focused on obtaining physical models to describe the ion transport from the measurement chamber to the sensor. Accurate physical models facilitate:

(1) Optimal design of the monitor for maximizing the ion transport efficiency; and
(2) Accurate estimation of radioactivity from measured ion currents.

Both of the above are closely interrelated and require precise knowledge about the relation...
between the radioactivity and the resultant ion currents. And the information about this relation has been pursued both from experimental and theoretical aspects (4)(8).

For this purpose, a physical model for 3-D computational fluid dynamics (CFD) and ion transport simulations has been made to include the effects of ion diffusion, neutralization and recombination. A series of CFD simulations have been performed using this model to optimize the geometry, especially of the chamber and the connecting sections between the chamber and the airflow ducts, in order to maximize the ion transport efficiency. As a result of our intensive and systematic study theoretically and experimentally, specifications of the monitoring system and its performance level have been found so far.

This paper presents, as the final stage of the development, computational estimation of the ion transport efficiencies for two pipes with different lengths, the inner surfaces of which were covered with a thin layer of uranium. The quantity of radioactive species attached to these samples was also experimentally determined to estimate the actual alpha radioactivity (11). Comparisons and discussions were made for the purpose of validating our model.

This paper is organized in the following manner: Chapter 2 deals with the computational methodology, in which our model and simulation conditions are presented. Chapter 3 offers simulation results and discussions about the ion transport efficiency as well as ion transport efficiencies to the sensor. Variations in ion transport efficiencies resulting from contaminated spots at different positions were also discussed. Summary is given in Chapter 4.

2. Computational methodology

2.1 Basic Equations

In the simulations, fluid dynamics simulations were made first; then, the ion transport on the simulated flow was calculated as a scalar function, including the effects of ion diffusion, neutralization and recombination. The $k-\varepsilon$ model was used for turbulence in the fluid calculations.

The ion transport equation can be written as

$$\frac{\partial \rho C}{\partial t} + \frac{\partial \rho u_j C}{\partial x_j} = \frac{\partial}{\partial x_j} \left[ \rho D \frac{\partial C}{\partial x_j} - \rho u_j \frac{\partial C}{\partial x_j} \right] + S - \alpha (\rho C)^2,$$

(2)

$$\rho u_j \frac{\partial C}{\partial x_j} = -\frac{\mu_t}{\sigma_t} \frac{\partial C}{\partial x_j},$$

(3)

where $C$ is the ion concentration, $\rho$ the air density, $D$ the molecular diffusion coefficient, $\mu_t$ the turbulent viscosity given in the airflow calculation, and $\sigma_t$ is the turbulent Schmidt number. $S$ is the ion source term representing air ionization by alpha particles. Self-extinction of ions, $\rho C/\tau$, was neglected since the decay time, $\tau \sim 20$ sec, is much longer than a typical transport time of about 3 sec for our operation conditions. The term, $\alpha (\rho C)^2$, represents the recombination of ions, in which $\alpha$ is the recombination coefficient. When the ions are transported to the metal surface, they are neutralized and lost. This ion neutralization on the metal wall was simulated by setting the ion concentration to zero on the walls of the chamber and ducts, as well as on the metallic surface of the waste.

2.2 Simulation Geometry

Figure 2 shows the appearance of the monitor investigated. The airflow in the measurement chamber flows from the bottom to the top. The air conveying ions is
directed upward to the sensor that is connected above directly to the outlet of the chamber. Two types of pipes used in back-end facilities of the nuclear fuel cycle, with an OD of 61 mm, an ID of 53 mm (these JIS (Japanese Industrial Standard) values were assumed) and respective lengths of 60 cm and 30 cm, were independently placed on the stage inside the chamber, perpendicular to the chamber axis (Figs. 3 and 4), and measured. In order to flush out the ions generated inside the pipes, a flexible tube was employed as a blower. The blowing speed was 1.3 m/s. For the pipe with a length of 60 cm (Fig. 3), a simple metal box was attached to one end of the pipe in order to capture small uranium-attached fragments coming off the pipe surface and falling down, so as not to contaminate the chamber.

In Fig. 5 the generated simulation meshes and the area of ion generation, as well as the conditions for the simulation, are presented. In this geometry, the chamber and ducts were divided by a plane which contains the chamber axis and mirror symmetry was assumed. In this figure, the airflow direction is from the right to the left. The speed of main flow at the sensor was set to 6.8 m/s which was the experimentally measured value. Ions are assumed generated uniformly inside the pipe since the inner surface of each pipe was assumed uniformly covered with uranium.
Fig. 3  Measurement of uranium-attached sample, a pipe with a length of 60 cm.

Fig. 4  Measurement of uranium-attached sample, a pipe with a length of 30 cm.

Fig. 5  3-D simulation model.
2.3 Simulation Conditions

The simulation conditions are summarized in Table 1. The $k$-$\varepsilon$ model was used to treat the turbulence. The coefficients of molecular diffusion and recombination of ions were $1.5 \times 10^{-5} \, [m^2/s]$ and $1.5 \times 10^{-12} \, [m^3/s]$, respectively; these values were independently obtained from other experiments (9). The simulations were conducted with a commercial CFD code, STAR-CD®.

### Table 1  Conditions for CFD simulations.

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<table>
<thead>
<tr>
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<tbody>
<tr>
<td>Air speed at chamber outlet [m/s]</td>
<td>6.8</td>
</tr>
<tr>
<td>Blower air speed [m/s]</td>
<td>1.3</td>
</tr>
<tr>
<td>Turbulence model</td>
<td>$k$-$\varepsilon$</td>
</tr>
<tr>
<td>Ion molecular diffusion coefficient [m$^2$/s]</td>
<td>$1.5 \times 10^{-5}$</td>
</tr>
<tr>
<td>Ion recombination coefficient [m$^3$/s]</td>
<td>$1.5 \times 10^{-12}$</td>
</tr>
</tbody>
</table>

In the case that the alpha source is located in such narrow spots as the inner surface of a small pipe, etc., ionization space should be considered. The average path length of alpha particles in air is about 5 cm; however, some alpha particles hit the inner wall before their full flight of 5 cm. In the case of pipes with an ID of 53 mm, calculation with the MCNPX code (12) shows that 80.9% of the total energy of emitting alpha particles is consumed to ionize atoms in the air. The rest of their energy is directly deposited in the pipe inner wall and is never used for ionization. Table 2 lists the factors appearing in Eq. (1) and the fraction of ionization space described just before. Three isotopes of uranium were considered.

### Table 2  Factors in Eq. (1) and ionization space.

<p>| | |</p>
<table>
<thead>
<tr>
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<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>Elementary charge, $e$ [C]</td>
<td>$1.6 \times 10^{-19}$</td>
</tr>
<tr>
<td>W value of air by a particle, $w$ [eV]</td>
<td>35.1</td>
</tr>
<tr>
<td>Sensor efficiency, $\eta_{\text{sensor}}$</td>
<td>1.0*</td>
</tr>
<tr>
<td>Alpha source efficiency, $\zeta_{\text{surf}}$</td>
<td>0.5</td>
</tr>
<tr>
<td>Fraction of ionization space, $\zeta$</td>
<td>0.809</td>
</tr>
<tr>
<td>Effect of self-screening, $\eta_{\text{self}}$</td>
<td>1</td>
</tr>
<tr>
<td>Average energy of a particle, $E_\alpha$ [MeV]</td>
<td>U$^{234}$ 4.773, U$^{235}$ 4.373, U$^{238}$ 4.194</td>
</tr>
</tbody>
</table>

*At the flow speed of experimental condition.

The ion source term in Eq. (2) was calculated from the uranium quantity estimated from ICP-MS (inductively coupled plasma mass spectroscopy) measurements (11). Here the surface of the sample was dissolved in acid and vaporized and the number densities of ionized $^{235}$U and $^{238}$U in the vapor were measured with mass spectroscopy. The sample contained a tiny amount of $^{234}$U that could not be measured with ICP-MS. We assumed that the number density of $^{234}$U follow the natural isotope abundance ratio of uranium (0.0055%). The radioactivity was then determined as the total amount of $^{234}$U, $^{235}$U and $^{238}$U.

The effect of self-screening, $\eta_{\text{self}}$, was assumed unity as shown in Table 2. As the reduction in the pipe weight before and after the exposure to the acid for uranium solution was about 0.1%, the thickness of the uranium-containing coating can be estimated to be about 2 $\mu$m at most (the same mass density as stainless steel was assumed). Since the
stopping length of a 5-MeV alpha particle in the same material is about 20 μm, the stopping effect was considered small.

The estimated radioactivity for the two pipes is tabulated in Table 3. Since the uranium was attached uniformly to the inner surface of the pipe, in addition, the ions were assumed generated uniformly inside the pipe. Ion generation densities were estimated assuming that half of the alpha particles from uranium actually depart from the surface ($\zeta_{\text{surf}} = 0.5$), that the fraction of ionization space, $\zeta$, is 80.9% and that ions are generated uniformly inside the pipe.

Table 3  Ion source term in the simulations.

<table>
<thead>
<tr>
<th>Sample #</th>
<th>#1</th>
<th>#2</th>
</tr>
</thead>
<tbody>
<tr>
<td>Pipe length [cm]</td>
<td>60</td>
<td>30</td>
</tr>
<tr>
<td>Alpha radioactivity [kBq]</td>
<td>39.6</td>
<td>19.2</td>
</tr>
<tr>
<td>Ion generation rate [ions/m3s]</td>
<td>$1.6 \times 10^{12}$</td>
<td>$1.6 \times 10^{12}$</td>
</tr>
</tbody>
</table>

3. Results and Discussions

3.1 Comparison of simulated and experimental ion transport efficiencies

Figures 6 and 7 show the distributions of ion concentration in the chamber for the two pipes, respectively. In these figures, the maximum values for contour plots were set lower to emphasize the region with low ion concentration. The ions are blown off out of the pipe and transferred toward the sensor by the main airflow in the chamber.

Fig. 6  Calculated ion distribution for uranium-attached sample (a pipe with a length of 60 cm).
The airflow blowing the ions out of the pipe extends toward the wall of the chamber and of the funnel-like section connecting the chamber and the outlet duct. Large part of ion loss due to neutralization arises here. Table 4 lists the simulation results including individual factors of ion loss and the final ion transport efficiencies. This table also presents ion transport efficiencies estimated experimentally. Both the simulated and experimental efficiencies were in good agreement, verifying our numerical models developed.

It is observed in Table 4 that both the simulated and experimental ion transport efficiencies differ depending on the geometry of the samples. So in order to estimate the radioactivity of the sample from the ion current accurately, this ion transport efficiency that depends on the shape of the sample should be known before measurements. A practical solution to this problem is to create a table of this efficiency for groups with similar shapes and sizes. This is very effective when the wastes are standard components.

The ion loss by recombination shows a different tendency in the simulation of the two pipes. The ion concentration in the pipe is generally higher than in the chamber. So the ion loss due to recombination is higher in Pipe #1 than in Pipe #2 since the ions stay longer in Pipe #1 than in Pipe #2. Accordingly the fraction of ions flowing out of the pipe is greater in the case of Pipe #2 than Pipe #1, resulting in the observation of higher ion loss due to neutralization on the chamber wall.
3.2 Characteristics of ion transport

In order to clarify the characteristics of ion transport and resultant ion currents on various practical parameters, various simulations were conducted. Figure 8 shows the dependence of ion transport efficiency as a function of alpha radioactivity for the pipes with respective lengths of 60 cm (#1) and 30 cm (#2). For the values of radioactivity lower than 100 Bq, the ion transport efficiency is almost constant, while it decreases as the radioactivity increases beyond 100 Bq. This may bring about complexity in determining the radioactivity from the sensor current when the waste is highly radioactive; however, in the case that radioactivity is as low as the clearance level, this tendency means that the ion current is proportional to the radioactivity and is supportive for clearance level inspection.

![Fig. 8 Ion transport efficiency dependence on alpha radioactivity.](image)

Figures 9 and 10 show simulated ion loss factors with respect to the sample radioactivity. For both sample pipes, ion recombination is dominant for radioactivity higher than 10 kBq. In contrast, ion recombination is negligibly small when the sample radioactivity is lower than 100 Bq. In this case the ion concentration is so small that the recombination term in Eq. (2) that is proportional to the square of the concentration becomes negligibly small compared with other terms. Other loss factors are constant for low radioactivity; then, as a result a linear dependence between the ion current and the radioactivity, i.e. a constant ion transport efficiency, is observed in Fig. 8 in the region below 100 Bq.

![Fig. 9 Ion loss factor dependence on alpha radioactivity (60-cm pipe).](image)
This value of 100 Bq is specific to our simulation conditions. It is reported, however, that increasing the blow-out speed increases the ion transport efficiency\(^5\) by suppressing ion recombination. This implies that to attain a certain speed in blowing will give this kind of threshold somewhere near this value and below this radioactivity a linearity between the radioactivity and the resultant ion current will be obtained.

Next, the dependence of ion transport efficiency on the contamination position is examined. This monitor essentially measures the total radioactivity; therefore, the effect of contamination position on the measured value should be important. The examination was conducted using the model presented in Fig. 11. The inside of the 60-cm-long pipe was divided into three parts, in each of which ions were generated uniformly. Two test values of total radioactivity were assumed: 15.4 kBq and 3.3 Bq.

Figure 12 shows these results. In Fig. 12, bars from left to right represent the upstream to downstream positions of ion generation, respectively, in the 60-cm-long pipe. As the position of ion generation moves downstream, the ion transport efficiency increases. This is because the ion loss due to recombination and neutralization on the pipe inner wall decreases. The mean values of the ion transport efficiency for the 60-cm and 30-cm pipes are 12.4 and 29.6%. Their variations in terms of standard deviation are 6.5% and 7.7%, respectively. Therefore, variations in the ion transport efficiency observed in Fig. 12 are about 6-8%. The dependence on the contaminated position is relatively weak. This value is considered acceptable for the specs of this monitor.
4. Summary

An innovative alpha radioactivity monitor for clearance level inspection has been developed. Computational estimation of transport efficiencies for two uranium-attached pipes was conducted and compared with experiments. Both of the transport efficiencies show good agreement, verifying our simulation models.

In order to clarify the characteristics of ion transport and resultant ion currents on various practical parameters, additional simulations were conducted. The simulated ion transport efficiencies were constant for samples with low radioactivity, below 100 Bq, while they increase for those with high radioactivity. This tendency means that the ion current is proportional to the radioactivity and is supportive for use of this monitor in clearance level inspection in which the alpha radioactivity is very low.

The variation in ion transport efficiency due to the location of contaminated positions was also examined. The ion transport efficiency is mildly dependent on the position of ion generation. The variations estimated are within 6-8%, allowing acceptable radioactivity estimation.

To overcome the geometry dependence of the ion transport efficiency, a table of pre-measured/pre-calculated correction coefficients is very effective. Our future work will, then, include the classification of wastes by their shape and size and the creation of a table for the classified groups.

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References


