Differences of Detection Efficiency among Several Nasal Swab Samples Simulated for Nuclear Emergency Accident

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Abstract. At nuclear emergency accident such as inhalation intake of alpha nuclide, an indispensable nasal swab method has not been used for the precise internal dose estimation. One of the reasons is uncertainty in its radiation measurement, so that precise measurement with alpha spectrometry was examined for filter samples simulating nasal swab. It was confirmed that the alpha spectrometry made possible the distinction between solution and particulate in addition to the nuclide identification. The alpha activity in swab sample was precisely evaluated only when the detection efficiency was determined considering the self-absorption with filter fibers. Another big problem of wiping efficiency in nasal swabbing is still remain, but this study certainly raised the usefulness of the nasal swab method for rapid response in emergency.

KEYWORDS: nasal swab, detection efficiency, plutonium, alpha spectrometry

1. Introduction

A nasal swab sample is taken from internally contaminated patient in nuclear emergency accident, but currently its major use is limited to confirmation of possible inhalation of alpha radionuclides. NCRP Report No.65 mentions that nasal swabs are useful because of their early availability but they should always be followed by more definitive tests, such as in vivo measurements with lung monitor or whole body counter and bioassay measurements with urine or feces [1]. In vivo measurement is not effective for measurement of pure alpha emitter such as plutonium-239, and the bioassay methods take much time over several days to get results. However, emergency medical treatment for decorporation, such as chelation therapy, should be carried out as soon as possible. From this point of view, nasal swab method is expected as rapid measurement to give objective evidence of inhalation intake. In recent year, some papers reevaluated nasal swab data for rapid internal dose assessment [2,3]. The special emphasis is placed on precise measurements for the nasal swab samples.

The nasal swab sample used to be measured with gross counter such as ZnS(Ag) scintillation counter cause of confirmation of possible inhalation. In this paper, alpha spectrometry, which is an established technique known to produce accurate results, was used to compare with gross counter. Alpha particles lose significant energy while passing through the filter, and some alpha loses all their energy. Some paper shows the results with concerning to under-reporting of alpha activity on filter [4] and the self-absorption in fiberglass filter [5]. For precise measurement of nasal swab sample, it should be examined to confirm energy loss in filter paper. And also all procedures from pre-treatment to alpha measurement were re-examined with plutonium nitrate and oxide.

2. Material and Methods

2.1 Preparation of simulated samples for nasal swab

The detection of alpha activity is strongly dependent on the sample codition and the geometry of sample to detector. A nasal swab is collected on a moist, clean, cotton-tipped applicator or on filter paper on a swabstick [1]. If a cotton applicator is used, the cotton fibers are teased off the applicator and spread out evenly for alpha counting. Therefore, it is difficult to fix the geometry constantly. On the other hand,

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the shape of filter paper is relatively stable even if it were wet. In this experiment, filter paper was used for our nasal swab sample on account of geometrical advantage.

The quantitative filter papers of ashless grades were selected. Whatman 40 (Whatman International Ltd., England), which is prepared for nasal swab in emergency medicine in NIRS, was used for simulated samples. The filter papers were cut in a circle of 25mm diameter before preparing simulated samples. To reproduce the state of moist, the distilled water of 30µl was dropped on the filter. The contaminant was dropped immediately after that.

The contaminants were prepared on the assumption that an inhalation accident might occur in a plutonium facility. Plutonium nitrate (Pu(NO₃)₄) solution and plutonium oxide (PuO₂) suspension were arranged for mist and dust inhalation accidents, respectively. The dropped volume into filter paper was settled from 1 to 10µl assuming the difference of nasal mucus condition. Seven samples were prepared for each volume. To measure the dropping concentration, stainless disks were similarly made. Our plutonium is mainly composed of ²³⁹Pu, and slightly contains ²⁴⁰Pu and ²⁴¹Pu. Therefore, total alpha activity includes ingrown ²⁴¹Am. The activity for simulated samples was evaluated by alpha spectrometry or scintillation alpha counter as total amount of alpha activity for ²³⁹Pu, ²⁴⁰Pu and ²⁴¹Am. The Pu(NO₃)₄ solution was diluted to 2N nitrate solution from 8N solution. From the measurement of Pu(NO₃)₄ solution dropped to stainless disks with the alpha spectrometer, the concentration of the solution was determined to be 8503 ± 306 Bq/ml. The PuO₂ suspension was made by which PuO₂ particles captured in air filters were resuspended in distilled water using an ultrasonic bath. The activity median aerodynamic diameter (AMAD) of PuO₂ had been 0.466 µm (geometric diameter; 0.1 µm, geometric standard deviation (GSD); 2.13). The concentration of PuO₂ suspension was determined to be 312 ± 13.2 Bq/ml measured with alpha spectrometer. The simulated swab samples were dried, and then were wrapped with Diafoil membrane (Mitsubishi Polyester Film Corp., Tokyo, Japan) of 1.5µm in thickness to protect surface contamination of alpha detector from recoiled alpha particles.

2.2 Alpha measurement procedure

For alpha spectrometry, alpha spectrometer with 12 alpha PIPS detectors (Alpha Analyst Model S570 and Model A1200-37AM, Canberra Industries, Inc.) was used. The effective area of the detector is 1200 mm², and the average counting efficiency of alpha particles is 0.37.

Gross alpha counting was made for comparison to alpha spectrometry. For gross alpha count, ZnS(Ag) scintillation counter (LUDLUM Model 43-10 Alpha Sample Counter, LUDLUM Measurements, Inc.) was used. Our LUDLUM counter was modified to protect of detector from surface contamination. The scintillator was replaced to the disposable scintillating sheet. The effective area of the detector is 2000 mm², and the average counting efficiency is 0.44.

3. Results and Discussion

3.1 Effect of mucus volume

The nasal mucus volume may be influence the spread of activity through the filter paper, and thus the detection efficiency might be decrease. For check this problem, the dropped volume of plutonium into filter paper was settled from 1 to 10µl. Figure 1 shows the detection efficiency on several volume of plutonium. Detection efficiency is given by following:

\[
\text{Detection Efficiency} = \frac{\text{measured count of filter paper}}{\text{decay number of deposited activity}} \quad (1)
\]

The result showed that nasal mucus volume had no influence of the detection efficiency. It was suggested that nasal mucus volume had no problem in the measurement if swab sample dried surely.
Figure 1: The detection efficiency on several volume of plutonium

![Detection Efficiency Graph](image)

### 3.2 Difference of alpha spectrum between Pu(NO$_3$)$_4$ and PuO$_2$

Figure 2 shows the typical spectra for Pu(NO$_3$)$_4$ solution and PuO$_2$ suspension. The spectrum for PuO$_2$ sample showed clear peaks for plutonium and americium, and a wide tail to the end of low energy. It was considered that PuO$_2$ particles adhered to the surface of the fiber, indicating that the energy of peak corresponded to the proper energy of plutonium and americium. Some PuO$_2$ got into the paper medium received the self-absorption by the fibers, and their alpha had formed the spectrum with a wide tail of lower energy side. In this result, the sharp peak was formed from about 40% in alpha count. This result showed that energy spectrum analysis was useful to identify the nuclide. It was also suggested to identify the nuclide of particular material easily even if it was contamination accompanied with two or more nuclides. On the other hand, the spectrum for Pu(NO$_3$)$_4$ sample showed different shape with comparison to that for PuO$_2$. The shape inclined gently toward low energy region. The peak is not clear in these samples, but it is important to estimate the nuclide based on the highest energy of a right edge. Considering the differences in spectrum form between PuO$_2$ suspension and Pu(NO$_3$)$_4$ solution, it was suggested that the characters of contaminants could be estimated from alpha spectrometry.

Figure 2: Typical spectra of simulated swab samples for Pu(NO$_3$)$_4$ solution and PuO$_2$ suspension

![Spectra Graph](image)

The detection efficiency of simulated swab sample was lower as shown in Figure 1. The detection efficiency would be the same as counting efficiency if deposited activity could be detected perfectly. It indicated that the alpha activity would be underestimate if it has been estimated using counting efficiency. The detection efficiency for Pu(NO$_3$)$_4$ solution was lower than those for PuO$_2$ suspension. It was affected by shape of spectrum. The shape of spectrum showed that the almost Pu(NO$_3$)$_4$ solution was infiltrated in the fiber while some PuO$_2$ particle adhered to the surface of the fiber. The lower detection efficiency for Pu(NO$_3$)$_4$ was result of large energy loss by filter fiber It suggested that the alpha activity of filter paper should be corrected by appropriate detection efficiency.

### 3.3 Comparison among different pressure conditions

The measurement of alpha spectrometry is done under vacuum condition usually. On the other hand, the gross counter is used at atmospheric pressure. The alpha particle loses energy by passing air layer. The difference of detection efficiency between under vacuum condition and atmospheric pressure was examined when the geometry of sample to detector was fixed. Table 1 shows the difference of detection efficiency for Pu(NO$_3$)$_4$ solution and PuO$_2$ suspension. The detection efficiency was
calculated when energy range of interest was changed. For PuO₂ suspension, about 40% count contributed to the peak formation among total count. The counts of each energy channel were small in the low energy area, and dependence of detection efficiency on energy range of interest was limited. On the other hand, the counts of each energy channel were constantly high in the low energy area for Pu(NO₃)₄ solution. Therefore, the detection efficiency decreased rapidly with decreasing energy range of interest. The distance from sample to detector was 5mm in this experiment. The air layer of 5mm affected the detection efficiency following energy loss by air layer thickness. The detection efficiency of Pu(NO₃)₄ solution at atmospheric pressure was affected more than that of PuO₂ suspension cause of same reason about spectrum shape. If the energy range cut at some level for avoiding high background, it is necessary to check the detection efficiency carefully based on these results.

Table 1: Differences of detection efficiency for Alpha Analysis depending on the energy range of interest

<table>
<thead>
<tr>
<th></th>
<th>1-6 MeV</th>
<th>2-6 MeV</th>
<th>3-6 MeV</th>
<th>4-6 MeV</th>
</tr>
</thead>
<tbody>
<tr>
<td>Under vacuum</td>
<td>Pu(NO₃)₄</td>
<td>0.078 ± 0.002</td>
<td>0.066 ± 0.002</td>
<td>0.049 ± 0.001</td>
</tr>
<tr>
<td></td>
<td>PuO₂</td>
<td>0.110 ± 0.014</td>
<td>0.100 ± 0.013</td>
<td>0.086 ± 0.012</td>
</tr>
<tr>
<td>Atmospheric pressure</td>
<td>Pu(NO₃)₄</td>
<td>0.062 ± 0.002</td>
<td>0.050 ± 0.002</td>
<td>0.033 ± 0.002</td>
</tr>
<tr>
<td></td>
<td>PuO₂</td>
<td>0.095 ± 0.013</td>
<td>0.083 ± 0.012</td>
<td>0.066 ± 0.011</td>
</tr>
</tbody>
</table>

Table 2 shows the detection efficiencies among different instruments at atmospheric pressure. The decreasing of detection efficiency for Pu(NO₃)₄ solution was clear comparing with PuO₂ suspension. The reason was mentioned above. For the LUDELUM counter, the detection efficiencies was higher than alpha spectrometer at atmospheric pressure condition, and its value was close to the highest detection efficiency for Alpha Analyst at under vacuum. The thickness of air layer mainly affected the detection efficiency at atmospheric pressure condition. The air thickness for LUDELUM counter was almost zero, and it was absolutely small comparing with the 5mm for Alpha Analyst. Modified LUDELUM, in which the scintillating sheet contacted to the sample, might cause improvement of the high detection efficiency. This result indicated that ZnS(Ag) scintillation counter would be expected to give the similar detection efficiency as under vacuum condition if the distance from sample to detector could be reduced to a minimum.

Table 2: Detection efficiencies among different instruments at atmospheric pressure

<table>
<thead>
<tr>
<th></th>
<th>Alpha spectrometer</th>
<th>ZnS(Ag) scintillation counter</th>
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<tbody>
<tr>
<td></td>
<td>Alpha Analyst</td>
<td>Modified LUDELUM</td>
</tr>
<tr>
<td>Pu(NO₃)₄</td>
<td>0.062 ± 0.002</td>
<td>0.079 ± 0.002</td>
</tr>
<tr>
<td>PuO₂</td>
<td>0.095 ± 0.013</td>
<td>0.110 ± 0.015</td>
</tr>
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</table>

4. Conclusion

An alpha spectrometry has big advantage to measure the individual activity of radionuclide comparing with gross alpha counting. In the alpha spectrometer measurement for simulated nasal swab samples, it was confirmed that not only the nuclide identification but also the properties identification of the radioactive substance was possible. The alpha activity in swab sample was precisely evaluated with sufficient detection efficiency. In the case of dust inhalation, the possibility that the nuclide could be identified even by two or more nuclides was suggested. The nasal swab sample could give more useful information at first response of emergency when the sample was measured with the alpha spectrometer.

REFERENCES


