Analysis of Radioactive Fall-out in Air at High Altitude by the Method of γ ray Spectroscopy

γ線スペクトルによる高層の放射能塵の核種分析

By Tatsuo URAI*

Analysis of radioactive fall-out at high altitude was made by the method of γ ray spectroscopy. The sample was collected in Chubu district of Japan on Nov. 10, 1961 by a F-86-F aircraft of Japan Air Self Defense Force. The method of collection was described in the previous paper (1).

From the γ ray spectrum it seems that this sample is composed of fall-out which was probably caused by the nuclear test of 50 Mt. nuclear weapon of U.S.S.R.

I. INTRODUCTION

On Nov. 10, 1961, a very intense radioactive sample (about 44,000 μC**) was collected at the altitude of 12 km in Chubu district (at Gifu Pref.). The γ ray spectrum of this sample was measured on Dec. 1, 1961 by the 512 channels γ ray spectrometer. This spectrum was analyzed by comparing with the spectra of standard γ ray sources of 22Na, 45Mn, 137Cs and 133Ba by the same spectrometer.

II. EXPERIMENTAL

1. Sample

The sample was collected by the F-86F aircraft of Japan Air Self Defense Force and treated with the method described in the previous paper (1) and put in a test-tube as shown in Fig. 1. The γ ray spectrum of this sample was measured by the 512 channels γ ray spectrometer of Nuclear Data Co., with a 13/4 φ×2 φ NaI scintillator.

2. Standard γ ray sources

As standard γ ray sources, 22Na (0.0289 μC±10%), 54Mn (0.0191 μC±10%), 137Cs (0.06044 μC±10%) and 133Ba (0.0952 μC±10%) were used. They were prepared by New England Co., USA and activities were calculated as of Dec. 1, 1961. Fig. 2 shows the size of each standard source.

3. Apparatus

The arrangement of samples and the spectrometer is shown in Fig. 3. Each sample was placed 9 mm from the scintillator surface on its central axis.


** It corresponds to 4,700 μμC/m³.
III. EXPERIMENTAL RESULTS

The spectrum of the sample of fall-out is shown in Fig. 4. Those of standard γ ray sources are shown in Figs. 5~8.

Fig. 4 γ ray spectrum of the collected sample

In Fig. 4 there are peaks at 1.6, 0.77, 0.51, 0.34 and 0.22 MeV. Each peak corresponds to the following γ rays:

- 1.6 MeV: $^{140}$La (1.596 MeV)
- 0.77 MeV: $^{95}$Nb (0.768 MeV), $^{95}$Zr (0.724, 0.758 MeV), $^{140}$La (0.815 MeV)
- 0.51 MeV: $^{103}$Ru (0.498 MeV), $^{147}$Nd (0.532 MeV), $^{140}$Ba (0.537 MeV), Anihilation of positron
- 0.34 MeV: $^{131}$I (0.364 MeV)
- 0.22 MeV: $^{239}$Np (0.31 MeV)

As shown in Figs. 5~8 full-energy-peaks of standard spectra are as follows.

<table>
<thead>
<tr>
<th>γ ray energy (MeV)</th>
<th>Nuclide</th>
<th>full-energy-peak (counts/30 min)</th>
<th>Half-width (keV)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1.27</td>
<td>$^{22}$Na</td>
<td>3,593</td>
<td>80</td>
</tr>
<tr>
<td>0.842</td>
<td>$^{46}$Na</td>
<td>4,004</td>
<td>70</td>
</tr>
<tr>
<td>0.662</td>
<td>$^{137}$Cs</td>
<td>15,080</td>
<td>60</td>
</tr>
<tr>
<td>0.357</td>
<td>$^{133}$Ba</td>
<td>41,463</td>
<td>47</td>
</tr>
<tr>
<td>0.300</td>
<td>$^{40}$K</td>
<td>10,785</td>
<td>—</td>
</tr>
</tbody>
</table>

Fig. 5 γ ray spectrum of $^{22}$Na

Note: Half-width of the peak of 1.27 MeV is 80 keV.

Fig. 6 γ ray spectrum of $^{46}$Mn

Note: Half-width of the peak of 0.84 MeV is 70 keV.
IV. Calculation

First, the ratio $\alpha$ of the area of the full-energy-peak to the number of emitted $\gamma$-photons of that peak energy must be calculated. This value is indicated by the formula.

$$\alpha = \frac{N_p}{N_d \times \epsilon}, \quad (1)$$

where $N_p$: area of full-energy-peak

$N_d$: number of disintegrations of nuclide

$\epsilon$: abundance of $\gamma$ ray of the peak energy.

In case of standard $\gamma$ sources these values are shown in Table 1. These values of $\alpha$ are plotted in Fig. 9. They fall on a straight line\(^5\). From this figure it is shown that $\alpha$ for 1.596 MeV is 0.0055. For this energy $N_p$ is 5,676 counts/80 min (Fig. 4) and $\epsilon$ is 100\%\(^4\). It is calculated by Eq. (1) as follows; $N_d = 5,676$ counts/80 min, $\alpha = 0.0055$, $\epsilon = 100\%$

\[Nd = \frac{N_p}{\epsilon \alpha} = \frac{5,676}{0.0055} = 1,032,000 \text{ counts/80 min} = 215 \text{ disintegrations/sec} = 5,811 \mu \text{mc (2)}\]

Since total activity of the collected sample is 44,000 $\mu \text{mc}$, the partial activity of $^{140}\text{La}$ is $5,811/44,000 = 13\%$.

$^{140}\text{La}$ emits also 0.815 MeV $\gamma$ rays. In this case, $\epsilon$ is estimated to be 20\% by Prikhodtseva, et al.\(^4\). $N_d$ is 1,032,000 counts/80 min from Eq. (2) and $\alpha$ is 0.013 from Fig. 9, so $N_p$ is calculated to be 2,683 counts/80 min by Eq. (1).

As shown in Fig. 4 the peak of 0.77 MeV contains not only the $\gamma$ rays of 0.77 MeV,
but also the $\gamma$ rays of 0.815 MeV from $^{140}$La. Therefore, the net peak of 0.77 MeV must be estimated by subtracting the contribution of 0.715 MeV $\gamma$ rays from the total peak area.

As shown in Fig. 4, the total area of the full-energy-peak of 0.77 MeV is 43,775 counts/80 min, and the contribution of 0.815 MeV $\gamma$ rays is 2,683 counts/80 min as shown above. The net peak is calculated as follows:

$$43,775 - 2,683 \text{ counts/80 min} = 41,092 \text{ counts/80 min} = 8.56 \text{ counts/sec.}$$

This value is caused by photons other than those photons which are emitted from $^{140}$La. They are probably emitted from $^{95}$Nb (0.768 MeV) and $^{95}$Zr (0.724, 0.757 MeV). This fact will be discussed later again.

For $^{95}$Nb (0.768 MeV) $\varepsilon$ is 100% and for $^{95}$Zr (0.757 + 0.724 MeV) $\varepsilon$ is 98-100%. Fig. 9 shows that $\alpha$ is 0.014 for 0.77 MeV. The sum of the partial activity of $^{95}$Nb and that of $^{95}$Zr is calculated as follows:

$$N_p = 8.56 \text{ counts/sec as shown above}$$

$$\varepsilon = 100\%, \quad \alpha = 0.014$$

$$N_p = \frac{N_p}{\varepsilon \alpha} = 611 \text{ d/sec}$$

$$= 16,500 \mu\text{mc.}$$

The total activity of the collected sample is 44,000 $\mu\text{mc.}$ So the partial activity of $^{95}$Zr-$^{95}$Nb is about 37.5%.

V. DISCUSSIONS

Hunter and Ballou investigated relative activities of fission product nuclides of $^{235}$U. These are shown in Fig. 10, and from this figure Table 2 has been made.

<table>
<thead>
<tr>
<th>Nuclide</th>
<th>Contribution to total activity (%)</th>
<th>$\gamma$ ray energy (MeV)</th>
<th>half life (day)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{144}$Ce, $^{144}$Pr</td>
<td>2.0</td>
<td>2.7</td>
<td></td>
</tr>
<tr>
<td>$^{133}$Xe</td>
<td>2.5</td>
<td>—</td>
<td></td>
</tr>
<tr>
<td>$^{131}$I</td>
<td>3.5</td>
<td>2.2</td>
<td></td>
</tr>
<tr>
<td>$^{147}$Nd</td>
<td>4.0</td>
<td>3.0</td>
<td></td>
</tr>
<tr>
<td>$^{95}$Nd</td>
<td>4.0</td>
<td>6.4</td>
<td>0.768 (100%)</td>
</tr>
<tr>
<td>$^{103}$Rh</td>
<td>5.4</td>
<td>6.2</td>
<td></td>
</tr>
<tr>
<td>$^{108}$Ru</td>
<td>5.5</td>
<td>6.4</td>
<td></td>
</tr>
<tr>
<td>$^{89}$Sr</td>
<td>6.7</td>
<td>8.0</td>
<td></td>
</tr>
<tr>
<td>$^{91}$Y</td>
<td>7.4</td>
<td>9.0</td>
<td></td>
</tr>
<tr>
<td>$^{95}$Zr</td>
<td>8.0</td>
<td>9.8</td>
<td>0.757 (98%)</td>
</tr>
</tbody>
</table>

The partial activity of $^{140}$La of the collected sample is 13% as described previously. It seems that this sample is mainly composed of fission products 30-40 days after the fission. Therefore this sample is thought to have come from the nuclear test of the 50 Mt. weapon of U.S.S.R. at the end of Oct. 1951.

By Table 2 the sum of the partial activity of $^{95}$Nb and that of $^{95}$Zr should be 14% at 30 days after fission and 16.2% at 40 days after fission. In our case, however, the collected sample shows, as described in Chap. IV, that the sum of the partial activity of $^{95}$Nb and that of $^{95}$Zr is 37.5%. It is probable that the collected sample is composed mainly of fission products 30-40 days after fission as described previously, and includes some other fission products. Fig. 10 shows that the contributions of $^{95}$Nb and $^{95}$Zr to the total activities continue to increase for 100-150 days after fission. Therefore the above explanation may be thought acceptable.

Fig. 4 shows that in case of 0.77 MeV peak, the ratio of the half-width to the square root of peak-energy is 2.95. On the other hand the ratio's for standard sources are 2.40(Figs. 5-8). This means that the peak at 0.77
MeV is composed of two or more kinds of γ photons. They are thought to be 0.815 MeV (140La), 0.768 MeV (95Nb), 0.757 MeV (95Zr) as described previously.

The author wishes to express his sincere thanks to Col. Rose of 5th Air Force USA, in Japan, Dr. T. Suzuki and Prof. Y. Nishiwaki for their encouragement and advice, and to Itochu Shoji Co., for the use of the γ ray spectrometer and to Mr. Zartman for reviewing the article.

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—REFERENCES—


研究論文

331. Experimental Study on Neutron Energy Spectra during Moderation in the Thermal Reactor

減速中の中性子エネルギースペクトルに関する実験研究

By Hideo KUROI*

Effects of neutron leakage and absorption on neutron energy spectra during moderation were studied experimentally by using resonance foils of In, Au, Co and Mn and also by using the information obtained with BF3 and 238U fission counters.

The influence of neutron leakage and absorption on the neutron energy spectrum were measured separately by activation measurements with these foils at the center of critical systems using UO2SO4 and D2O as fuel and moderator respectively, and it was indicated that the neutron spectrum φ(u) had a break point in the vicinity of 100 eV due to the combination of these two effects, especially in the systems of high fuel concentration.

The space dependence of intermediate neutron energy spectra in the reflected systems was also investigated. A considerably wide 1/E neutron spectrum was observed at a small distance outside the core-reflector interface and a peak of neutron leakage was also observed at a small distance inside the same interface. In the reflected system, the characteristics of the intermediate neutron energy spectrum as a function of "space" could be understood fairly well.

I. INTRODUCTION

Usually, the 1/E neutron energy spectrum is used as the neutron energy spectrum during moderation in a reactor system. Strictly