Multielement Position Sensitive Proportional Counter for Measurement of Tritium Labeled Gas Movement

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Received March 9, 1987
Revised November 14, 1987

A multielement position sensitive proportional counter was developed for the measurement of $^3$H labeled gas movement in a pipe by internal counting. The counter consists of an outer cylindrical pipe with an inner diameter of 40 mm, seven resistive stainless steel anode wires with a diameter of 22 μm, and 24 cathode wires with a diameter of 120 μm. These wires divide the radial cross section of the pipe into seven regions. Since each region independently works as a position sensitive proportional counter and the seven resistive anode wires are connected in series through lump resistors, the region and the axial position where a β-decay event occurred can be known by a simple operation upon the charge signals obtained from two preamplifiers. The axial position resolution for $^3$H β-rays was 9 mm FWHM. Demonstrative experiments were performed for the flow and diffusion of $^3$H labeled ethane in the counter pipe.

KEYWORDS: position sensitive counters, multielement proportional counters, charge division, internal counting, gas multiplication factor, tritium labeled gases, gas movement, gas flow, gas diffusion, tritium

I. INTRODUCTION

Various types of position sensitive proportional counters (PSPC) have been developed including two dimensional detectors\(^{(1)}\)(\(^{(2)}\)) or three dimensional ones such as parallel plate multiwire proportional counters\(^{(3)}\)(\(^{(4)}\)). These PSPC's are used mainly for incident radiations from the outside of the counter or the nuclear reaction products in the counter caused by incident radiations and not for internal radioactive gas counting. On the other hand, proportional counters which have been used for internal radioactive gas counting\(^{(5)}\)(\(^{(6)}\)) have not had position sensitive capability.

Recently, new measuring methods of the absolute radioactivity of $^3$H labeled gas\(^{(7)}\)(\(^{(8)}\)) and $^{14}$C labeled gas\(^{(9)}\) have been developed. In these new methods the information on the movement of the labeled gas in the counter is very important to determine the appropriate time to start the activity measurement after the injection of the labeled gas into the counter. This information is also useful for the measurement of radioactive gaseous samples using ionization chambers\(^{(10)}\). The authors have already measured $^3$H labeled gas movement using a PSPC with a single anode wire\(^{(11)}\). The counter could not detect the radial distribution of the labeled gas in a pipe.

A PSPC with a multielement counter construction was fabricated, which was able to give some information on the radial distribution in addition to the axial distribution of $^3$H labeled gas in a cylindrical pipe. In this paper the characteristics of the counter are described.

II. MULTIELEMENT POSITION SENSITIVE PROPORTIONAL COUNTER

1. Measuring Principle

Figure 1(a) shows the radial cross section of the counter. The cross section of a brass pipe with an inner diameter of 40 mm was
divided by cathode wires with a diameter of 120 μm into seven hexagonal regions. A stainless steel wire as a resistive anode with a diameter of 22 μm and a resistance of 23 Ω/cm was set at the center of each region to make each work as an individual element PSPC. The regions and the anodes are numbered from A1 to A7 as shown in Fig. 1(a). A similar geometrical structure of a multielement proportional counter was presented by Povinec et al.\textsuperscript{[13][14]}, but it was not a position sensitive counter. They used it for the purpose of decreasing background counts in \textsuperscript{3}H or \textsuperscript{14}C counting by taking coincidence and anti-coincidence counts among different anode signals.

The average and maximum energies of \textsuperscript{3}H β-rays are 5.7 and 18 keV, respectively, and the corresponding penetration ranges in the present counting gas (Ar 50%, CH\textsubscript{4} 50%) with atmospheric pressure are about 1 and 10 mm, respectively. These penetration ranges are effectively short when compared with the dimension of each region. Hence, most β-rays ionize the counting gas in one region. Consequently the region in which the β-decay event occurred can be identified by adopting the multiwire structure mentioned above. On the other hand, the axial distribution of \textsuperscript{3}H labeled gas can be measured by wiring resistive anodes as shown in Fig. 1(b). The lump resistors between anodes were needed to separate the position signals of each region. The resistance (300 Ω) was chosen to be 1/5 that of an anode wire. The effect of lump resistors on the pulse-propagation time constant (about 0.4 ms) is therefore only about 1/5. Since the applied high voltage to A1 was higher than that of the other six anodes, the capacitor C\textsubscript{12} was needed for the separation of the high voltages. The equivalent impedance of this capacitor for the signal pulse was 150 Ω, so that the resistance R\textsubscript{12} was chosen to be 150 Ω. The lump resistors R\textsubscript{L} and R\textsubscript{R} at both ends were introduced to bring a uniform detection efficiency over the anode wire chain\textsuperscript{[13]}.  

2. Counter Fabrication

The electrode consisting of a Teflon plate and Kovar sleeves was set at each end of a brass pipe. The length of each anode wire was 550 mm. On the upper side of the counter, there was a window with a width of 5 mm and a length of 500 mm along the axial direction covered with an aluminum-coated Mylar film as shown in Fig. 1(a). Through the window, collimated X-rays could be injected for the position calibration. At the bottom of the counter, three gas ports were set; at the center, 200 mm to the left and 250 mm to the right from the center. They were used for the insertion of a position calibration \textsuperscript{3}H source and the injection of \textsuperscript{3}H labeled gas.

III. EXPERIMENTAL RESULTS AND DISCUSSIONS

1. Performance of Multielement PSPC

The \textsuperscript{55}Mn KX-rays collimated to a diameter of 1 mm were injected at the center of the coun-
through the Mylar film window shown in Fig. 1(a). The counting gas was a mixture of Ar (50%) and CH₄ (50%). Figure 2 shows the pulse height spectra of the X-rays when an equal high voltage (1,400 V) was applied to all anodes. Figure 2(a) shows the spectrum obtained from the central counter A₁ and (b) from one of the peripheral counters A₂. The pulse height obtained from A₂ is about 30% higher than that from A₁. The difference in pulse height between the element counters causes the difference in detection efficiency. This is because, β-rays bring continuous pulse height spectrum and the pulse height discriminator used for the purpose of cutting noise signals brings different effective discrimination levels for different pulse height spectra of ³H β-rays.

Fig. 2 Pulse height distributions of ⁵⁵Mn KX-rays obtained from: (a) central counter A₁; (b) One of peripheral counters A₂ under same applied high voltage (1,400 V)

It is desirable for the pulse height distributions to be the same for all element counters in order to obtain a uniform detection efficiency. The difference between gas multiplication factors or the pulse heights of the central counter A₁ and the peripheral counters is explained by the following approximate calculation using a simple electric field model. The method to obtain the same gas multiplication factor will be described next.

In the electrode system shown in Fig. 3, when the distance Rᵢ between the centers of an anode A (radius a) and a cathode B (radius b) is very large compared with the radii a and b, the equipotential curves near the thin wire electrodes are almost completely circular, irrespective of the shape of the other electrodes (15)(16). This means that the electric charge distribution on each electrode surface is uniform. Hence, the charge can be considered as an infinitely thin wire charge in the calculation of electric field strength and the principle of the superposition of the electric field can be applied. In the hexagonally symmetrical electrode system, when rectangular co-ordinates are chosen as shown in Fig. 3 the y component of the electric field E(r) at r on the x co-ordinate is zero due to the cancellation by the symmetry of each y component. The electric field at r is, hence, expressed as follows:

\[
E(r) = \frac{\sigma_A}{2\pi \varepsilon r} + \sum \frac{\sigma_{B_i}}{2\pi \varepsilon R_i^2 + r^2 - 2rR_i \cos \theta_i} \cos \theta_i
\]

where \(\sigma_A\) and \(\sigma_{B_i}\) are the charges per unit length of electrodes A and B, respectively, \(\varepsilon\) is the dielectric constant of the counting gas.

Fig. 3 Electrode system with thirteen wires in Cartesian co-ordinate

As an approximation of the electric field strength of the A₁ counter shown in Fig. 1, we calculated the electric field of the infinite system which consists of an infinite number
of elemental counters with hexagonal shape. In the infinite system the number of cathode wires is five times of that of the anode wires and there is no leakage of the lines of electric force, so that \( \sigma_A + 2\sigma_{B1} + 3\sigma_{B2} = 0 \) is held, where \( \sigma_{B1} \) and \( \sigma_{B2} \) are the charges on the cathodes at \( R_1 \) and \( R_2 \), respectively, shown in Fig. 3. Since \( R_1 \) is nearly equal to \( R_2 \), the average cathode charge \( \sigma_B \) \( \equiv (2\sigma_{B1} + 3\sigma_{B2})/5 \) can be introduced and \( \sigma_B \) is, therefore, equal to \( -\sigma_A/5 \). The calculated result as a function of the distance \( r \) by the use of Eq. (1) is shown in Fig. 4(a) by a solid line for the applied voltage of 1,400 V. The electric field of the system which has a cylindrical metal wall with radius of \( (R_1 + R_2)/2 \) is also shown by a broken line as a comparison.

\[
V_A = \frac{1}{2\pi \varepsilon} \left\{ \sigma_A \ln \frac{R_2-b}{a} - \frac{1}{2} \sum \sigma_{Bi} \ln \frac{R_i^2 + (R_2-b)^2 - 2(R_2-b)R_i \cos \theta_i}{R_i^2 + a^2 - 2aR_i \cos \theta_i} \right\} \quad (2)
\]

The summation was done using only the adjoining six hexagonal counters in consideration of the negligible effect of far distant counters on \( V_A \). The \( \sigma_A \) can, thus, be calculated by substituting the actual applied voltage \( V_A \). When the applied voltage is equal in both systems, \( \sigma_A \) in the system with the thin wire cathode is smaller than \( \sigma_A \) in the system with the metal wall cathode. The charge difference can be explained through the second term of Eq. (2), so that the electric field near the anode surface in the former system is weaker than the latter by this amount. In the counter with a cylindrical metal wall cathode with a radius of \( R \), the next relation holds:

\[
\frac{V_A}{\ln \frac{R}{a}} = \frac{\sigma_A}{2\pi \varepsilon},
\]

and we can calculate the gas multiplication factor \( M \) by using Diethorn's equation:

\[
M = \exp \left\{ \frac{V_A}{\ln \frac{R}{a}} \frac{\ln 2}{\Delta V} \left( \ln \frac{\sigma_A}{2\pi \varepsilon pa \ln \frac{R}{a}} - \ln K \right) \right\},
\]

where \( \Delta V \) and \( K \) are the specific constants of the counting gas and \( \rho \) is the pressure of the gas. Through Eq. (3), Eq. (4) can be rewritten in terms of the anode charge \( \sigma_A \), or the electric field strength around the anode wire,

\[
M = \exp \left\{ \frac{\sigma_A}{2\pi \varepsilon} \frac{\ln 2}{\Delta V} \left( \ln \frac{\sigma_A}{2\pi \varepsilon \rho a \ln \frac{R}{a}} - \ln K \right) \right\},
\]

In the peripheral counter which has a metal wall cathode in part, the anode charge \( \sigma_p \) was estimated by the following equation:

\[
\sigma_p = \frac{\sigma_m + 5\sigma_w}{6},
\]

where \( \sigma_m \) and \( \sigma_w \) are the charges induced on the anodes in the counter with a metal wall over the whole cathode (Eq. (3)) and in that with a thin wire wall over the whole cathode.
In Eq. (6) the ratio, about 1/6, of the metal wall portion to the total cathode wall including the wire wall was taken into account. The gas multiplication factor was calculated by substituting $\sigma_m$ and $\sigma_p$ for $\sigma_A$ in Eq. (5) in each counter, respectively. The results were listed in Table 1. The values in the table were normalized to the value of the counter with the cylindrical metal wall cathode. The absolute gas multiplication factors are given in parentheses.

Though a mixture gas of Ar (50%) CH$_4$ (50%) was used as a counting gas in this experimental work, the calculation was done for the PR-gas because there was no data on $\Delta V$ and $K$ for the present gas. Each calculated value agreed well with the experimental value.

### Table 1

<table>
<thead>
<tr>
<th>Cathode Structure</th>
<th>$\sigma_m$</th>
<th>$\sigma_p$</th>
<th>$\sigma_A$</th>
<th>$\sigma_0$</th>
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<tbody>
<tr>
<td>Physical property</td>
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<td></td>
<td></td>
<td>0.95</td>
</tr>
<tr>
<td>Anode charge</td>
<td>$\sigma_m \times 1$</td>
<td>$\sigma_p \times 0.94$</td>
<td>$\sigma_A \times 5.94$</td>
<td>$0.95$</td>
</tr>
<tr>
<td>Relative gas multiplication factor</td>
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<td>0.40</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Relative pulse height, experimental</td>
<td>0.31</td>
<td>0.38</td>
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<td></td>
</tr>
</tbody>
</table>

As mentioned before, the pulse height is desirable to be the same in all regions to give the same detection efficiency for $\beta$-rays. A higher voltage was, therefore, applied to the anode A1 of the central region than to the others A2~A7, and an equal amount of charge was induced on all of the anodes. For the purpose of obtaining the same pulse height, the ratio of the applied voltage of A1 to the voltage of other anodes was experimentally determined and was 1.020, whereas the theoretical ratio determined by Eq. (5) was 1.022. Such high voltage application resulted in the same pulse height in all of the counters. The capacitor C$_{12}$ shown in Fig. 1 was introduced to receive this voltage difference and its impedance for the signal was desirable to be as small as possible.

**Figure 5** shows the pulse height spectra of $^3$H $\beta$-rays obtained from the bottom counter A5 with the insertion of a $^3$H labeled nucleic acid source with 430 Bq into the central calibration hole. The spectrum (a) with the applied voltage of 1.7 kV is nearly the same as the energy spectrum of $^3$H $\beta$-rays. In this situation the gas multiplication factor about $3 \times 10^3$ is so small that the signal to noise ratio, hence the position resolution, is poor. It is also difficult to expect a high detection efficiency because of the presence of many pulses whose amplitudes are less than the discrimination level of the pulse processing system. In the spectrum (b) with the applied voltage of 2.3 kV, the gas multiplication factor being about $5 \times 10^4$, saturated for the $\beta$-rays with higher energy but did not for those with lower energy$^{(12)(18)}$. The pulse height distribution was different from that of (a). The detection efficiency became more than 98% in (b) by setting the pulse height discrimination at the level shown by the arrow in the figure. Furthermore, since the amount of charge per event was larger than that in (a), the signal-to-noise ratio was greatly improved.

**Fig. 5** Pulse height distributions of $^3$H $\beta$-rays with applied voltage of: (a) 1,700 V; (b) 2,350 V

(2) Position Resolution and Position Linearity

The best position resolutions were 6 mm
FWHM for $^{55}\text{Mn}$ KX-rays collimated to a width of 0.4 and 9 mm FWHM for $^3\text{H}$ $\beta$-rays emitted from a circular source with a diameter of about 1 mm. The resolution of 9 mm for $^3\text{H}$ $\beta$-rays was enough for the purpose of the measurement of labeled gas movement in the counter with an effective length of about 550 mm.

A collimated Mn KX-ray beam was injected into the all of the regions through the window and the relation between the channel number of position signals and the position of X-ray incidence were obtained. The lineality of the position signal against the X-ray incident position was within $\pm 0.2\%$ over $7 \times 550$ mm.

2. Demonstrative Experiment with $^3\text{H}$ Labeled Gas

Figure 7 shows the flow patterns of $^3\text{H}$ labeled ethane ($\text{C}_2\text{H}_6$ 70%, $\text{CF}_4$ 30%) in the counting gas (Ar 50%, $\text{CH}_4$ 50%) in regions A2, A3, A5, A6 measured for 5 s at $5, 10, 40$ s, $3 \text{ min}$ 10 s, $7 \text{ min}$ 10 s after the injection of about $1 \text{ cm}^3$ (370 Bq) of ethane through the right side gas port shown in Fig. 1(a). It was clearly observed that the ethane was flowing with axial diffusion and cross sectional diffusion from A5 to other regions. Figure 8 shows the diffusion of $^3\text{H}$ labeled ethane of about 0.75 cm$^3$ injected through the central port. In this case the flow of the counting gas had been stopped. At 5 s after the injection the largest count was observed at A5 and then $^3\text{H}$ labeled gas diffused cross-sectionally and axially. The concentration of the labeled gas finally became uniform over all of the regions.

The axial diffusion of $^3\text{H}$ labeled ethane injected at the axial center of a pipe with a length of $2X_0$ can be expressed as follows by modifying the diffusion equation

$$C(x) = \frac{f(x+\xi)}{2 \pi D t} \sum_{n=-\infty}^{\infty} \exp \left\{ -\frac{-(4n+1)^2 X_0^2 + x^2 + \xi^2}{4Dt} \right\} d\xi, \quad \text{(7)}$$

The concentration of the labeled gas finally became uniform over all of the regions.
Fig. 7 Movement of $^3$H labeled ethane in each region at various elapsing times after injection of labeled gas

Fig. 8 Diffusion of $^3$H labeled ethane
$B/(X^2 + A)$ to the experimental initial axial distribution which is the sum of the seven areas. Constants $A$ and $B$ were determined to be $6.383$ (cm$^2$) and $1.915$ (counts·cm/5s) respectively. Because of the difficulty of finding out the numerical value of $D$ of ethane in Ar-CH$_4$ mixture gas, $D$ was chosen to be $0.400$ (cm$^2$/s) at 760 mmHg and 293 K by referring the present experimental results and reference values$^{(20)}$ of H$_2$ in CH$_4$, 0.156 of Ar in Ar, and 0.240 of CH$_4$ in CH$_4$. The calculated results shown by solid lines in the “Total” in Fig. 8 agree fairly well with the experimental ones shown by dots.

(2) Evaluation of Influence of Range of Tritium $\beta$-ray

The $^3$H $\beta$-ray has a maximum range of about 1 cm in this counting gas, so that it sometimes ionizes the gas in more than one region. In such case the position signal obtained does not correspond to the true position where the $\beta$-decay occurred. Therefore the fraction of such events must be evaluated. Such events can be identified when each resistor $R_{ij}$ between anodes is set at the value of $15 \Omega$, which is ten times that of an anode.

Figure 9 shows the position signal obtained when the $^3$H labeled gas uniformly diffused in the counter under the above setting. The signal originated by a $\beta$-ray which ionized the counting gas in only one region, for example A2, is registered at the corresponding peak A2 in the figure, even if a $\beta$-ray is detected at any different position along the anode A2.

On the other hand, when a $\beta$-ray ionized the counting gas in two regions and the number of ion pairs in one region is more than 1/10 that of the other regions, the signal is registered between the peaks, since each resistance of $R_{ij}$ between anodes is ten times that of an anode. By using Fig. 9 the fraction of such an event was determined to be 1/7 of the total events. This fraction, of course, depends on the diameter of a counter, the kind and the pressure of the counting gas, and the energy of $\beta$-rays.

IV. CONCLUSION

A multielement position sensitive proportional counter was constructed to detect the axial and radial movements of $^3$H labeled gas in a cylindrical pipe by dividing the cross section of the pipe into seven regions with many cathode wires and seven resistive anode wires. The difference in pulse heights among regions was theoretically explained under a simple electric field model. To make the difference disappear, different high voltages were applied. The axial position resolution for $^3$H $\beta$-rays was 9 mm. Detection efficiency was more than 98%, which was achieved by making the saturation of the gas multiplication factor for higher energy $^3$H $\beta$-rays. The fraction of $^3$H $\beta$-rays which ionized the counting gas in more than one region was evaluated to be 1/7 of the total $\beta$-rays. The diffusion and flow of $^3$H labeled ethane in the counter were measured and the results agreed fairly well with simplified calculated results.

ACKNOWLEDGMENT

The authors would like to thank Dr. K. Hoizumi, Mr. N. Takeuchi and the staffs of the radioactivity standardization group of Japan Atomic Energy Reserch Institute for their valuable discussions.

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