Effect of Methylal Quenching Gas for Self-Quenching Streamer (SQS) Tube with Ar-isoC₄H₁₀-Methylal Mixture

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Received December 14, 1992
Revised April 12, 1993

Properties of a self-quenching streamer (SQS) tube with an Ar-isoC₄H₁₀(75:25) mixture containing 2.5% methylal vapor have been investigated for uniform γ-ray irradiation. The effect of electro-negative methylal as the second quenching gas has been studied for avalanche sizes, pulse shapes of current signals, dead zones, and counting-rate characteristics of the SQS tube. The dead zone and counting-rate characteristics are improved very much by the use of the mixture containing methylal.

KEYWORDS: gas counter, self-quenching streamer mode, self-quenching streamer tube, GM tube, gamma radiation, radiation monitors, methylal, counting rates

I. INTRODUCTION

In our previous papers(1), it has been shown that self-quenching streamer (SQS) tubes, i.e. gas counters operated in the SQS mode, have good performance as a radiation monitoring device in high counting-rate condition. This paper describes further studies on performance of an SQS tube with a mixture containing second quenching gas vapor. Basic characteristics of the SQS mode have been investigated by Alekseev et al.(2), Atac et al.(3) and others(4). The following has been revealed in the SQS mode in comparison with the other operation modes of gas counters: high gas gains associated with a leap in the gas multiplication curve, fast rise time of signals, localization of streamers on the anode, and small dead zones. Because of these characteristics attributable to SQS formation, the SQS-mode operation of gas counters may have an advantage in their use in high counting-rate condition.

Until now, we have studied the characteristics of SQS mode in a gas counter filled with Ar-mixtures at 101.3 kPa using α-, β- and X-ray sources(5). Furthermore, it has been recently found that most of rare-gas (He, Ne, Ar, Kr or Xe) mixtures with a quenching gas of CH₄, C₂H₆, C₃H₈, isoC₄H₁₀ or CO₂ can be used for the SQS-mode operation except for Ne-CH₄, Ne-CO₂, He-CH₄ and He-CO₂ mixtures(6). Dead zone characteristics in the SQS-mode operation with Ar-isoC₄H₁₀ and Ne-isoC₄H₁₀ mixtures have been reported and compared with those in the GM-mode operation(1).

The effect of the second quenching gas vapor of methylalcohol, ethanol(7), and photoionizing vapors(8) of triethylamine and benzene has been studied for stabilizing the characteristics of the SQS mode and for lowering the transition voltage from the proportional mode to the SQS mode. The effect on
counting-rate characteristics, however, has not been studied yet. The counting-rate characteristics can be improved by decreasing the dead zone with an electro-negative gas vapor instead of the quenching gases listed above. Since methylal is known to be slightly electro-negative through studies on the operating gas mixture for multiwire proportional counters, it is interesting to study further its effect on the counting-rate characteristics and the dead zone of the SQS tube.

This paper will describe the effect of methylal as the second quenching gas vapor on the gas multiplication properties, the pulse shape of current signals, the dead zone, and the counting-rate characteristics for improvement of the performance of the SQS tube. The performance will be compared with that of a GM tube as a reference.

II. APPARATUS

The SQS and GM tubes used were a slightly modified model of a commercially available GM tube (Aloka 2008 GM). The SQS and GM tubes with a gas inlet were made of a coval cathode of 20 mm in inner diameter, and a tungsten anode wire of 50 μm in diameter stretched along the axis to provide an effective counter length of 81 ± 1 mm. The filling gas mixture for the SQS tube was chosen to be an Ar-isoC₆H₁₄ (75:25) mixture of 101.3 kPa on the basis of our previous work, which showed the SQS mode is stable with the mixture (6). Then, a small amount of electro-negative methylal vapor (CH₃(OCH₃)₂) was added to the mixture as the second quenching gas vapor. The GM tube was filled with an Ar-ethylformate (HCOOC₂H₅) (96:4) mixture at 20 kPa, which was the same gas filling as that for Aloka 2008 GM tubes. The tubes were irradiated with ⁶⁷Co γ-rays.

The measurement was performed with almost the same apparatus as in the previous work (1). The schematic block diagram for the measurement is shown in Fig. 1. The pulse height of signals from a charge sensitive preamplifier followed by a main shaping amplifier was converted to the avalanche size.
corresponding to the number of electrons or ions produced in an avalanche. Pulse shapes of the signal current flowing through a 200 Ω resister in a fast current-to-voltage converter were observed with an oscilloscope and recorded on photographs. In order to determine the dead zone of the SQS and GM tubes, the counting loss as a function of time intervals between successive pulses was measured with a pulse-interval-time-to-amplitude converter (PI-TAC)\(^{(18)}\).

III. RESULTS AND DISCUSSION

1. Gas Multiplication Properties

Avalanche-size curves for the SQS and GM modes are compared in Fig. 2(a). As seen in the figure, the SQS mode obtained with the Ar-isoC\(_4\)H\(_{10}\)(75:25) mixture of 101.3 kPa is characterized by a large leap in the avalanche-size curve: Amplitudes of output pulses continuously increase by more than one order of magnitude above 2.7 kV. A typical GM-mode operation obtained with the GM tube (equivalent to Aioka 2008 GM tube) represents larger gas gain at much lower applied voltages than the tube operated in the SQS mode.

The effect of methylal as the second quenching gas vapor was investigated for the gas multiplication in the SQS tube. After the mixing ratio of Ar and isoC\(_4\)H\(_{10}\) was fixed at 75:25, methylal was added to it by a proportion of 2.5, 5.0 and 7.5% in volume. Since methylal is slightly electro-negative, avalanche size decrease with increasing the methylal proportion as shown in Fig. 2(b). The leap corresponding to the SQS transition is diminished at the 7.5% methylal proportion. By addition of 2.5% methylal, avalanche sizes in the SQS mode are decreased by a factor of about 2 for the mixture without methylal.

The avalanche size in the limited proportional region is almost independent of the methylal proportion, and is about \(2.5 \times 10^6\) electrons. This means that approximately the same avalanche size is necessary for the SQS transition. The methylal proportion was fixed at 2.5% for the following studies in the present work: The mixture consisted of 97.5% of Ar-isoC\(_4\)H\(_{10}\)(75:25) mixture and 2.5% of methylal in volume.

2. Pulse Shapes of Current Signals

In Fig. 3 pulse shapes of SQS signal current flowing through a 200 Ω resister are compared for the Ar-isoC\(_4\)H\(_{10}\) mixtures without methylal and with 2.5% methylal. The signals in the SQS mode show a sharp exponential-decay-like shape of a width of about 100 ns fwhm (about 140 ns in decay time) with the Ar-isoC\(_4\)H\(_{10}\) mixture, as seen in Fig. 3(a) and (b). Figure 3(c) shows that the width of the signal shape decreases by a factor of about 2 with the mixture containing 2.5% methylal.

Since the current signal represents the time variation of induced charges\(^{(11)}\), the process of signal formation is related to the signal shape. The shape of SQS signals is given by the convolution of induced current from a string of electrons and ions (i.e. a streamer) produced perpendicularly to the anode. For
The conventional definition of dead time is insufficient to describe the properties of the SQS tube. As discussed in our previous paper \(^{(10)}\), the dead zone is written for uniform \(\gamma\)-ray irradiation over the whole tube as follows:

\[
\text{Dead length: } \delta(t) = L(1 - N(t)/N_0(t)) \geq L, \quad (1)
\]

\[
\text{Dead zone: } A = \int \delta(t) dt, \quad (2)
\]

where \(N_0(t)\) is the number of expected events at a time interval \(t\) between successive pulses in the case of no dead zone, \(N(t)\) the number of observed events at the time interval, and \(L\) the length of the tube. These formulae are also applied for GM tubes; since GM tubes are insensitive in the dead length of \(L\) during a dead time \(T\), the dead zone of GM tubes is to be \(L \cdot T\).

For determination of the dead zone, counting losses were measured as a function of the time interval between successive pulses induced by \(\gamma\)-rays from a \(^{60}\)Co source with a pulse-interval-time-to-amplitude converter (PI-TAC). Figure 4 shows an example of the measured interval distribution (i.e. the number of counts as a function of the time interval) for the SQS tube with the Ar-isoC\(_4\)H\(_{10}\) mixture. In order to calculate the dead length distribution with Eq. (1), \(N_0(t)\) is obtained by a
least square fit in a region of \( t \) from 1,000 to 2,000 \( \mu s \), where the dead zone is not effective, as indicated by a solid line in Fig. 4. Comparing \( N(t) \) and \( N_0(t) \) in the figure, it is found that the counting loss in the SQS tube is small even at very short time-intervals. Dead length distributions are indicated as a function of the time interval for the SQS and GM tubes in Fig. 5. The dead length distribution in Fig. 5(a) was obtained from the data shown in Fig. 4. As seen in Fig. 5(c), the GM tube is completely insensitive during time intervals less than about 400\( \sim \)500 \( \mu s \). The dead length distributions derived from the measured interval distributions were used for evaluation of the dead zone. The obtained dead zones are 750 \( \mu s \cdot cm \) for the SQS tube with the Ar-isoC\(_{16}H_{10}\) mixture, 140 \( \mu s \cdot cm \) for the SQS tube with the Ar-isoC\(_{16}H_{10}\) mixture containing 2.5\% methylal, and 5,100 \( \mu s \cdot cm \) for the GM tube. The result suggests that the dead zone of the SQS tube is minimized by mixing a small amount of methylal to the mixture.

The dead zones obtained for the Ar-isoC\(_{16}H_{10}\) mixture are rather larger than those obtained for the same gas mixture in another counter, whose cathode is 14 mm in diameter and effective length is 108 mm, by means of a collimated \( \beta \)-ray source\(^{(1)} \). In that work, dead zone values were 290 \( \mu s \cdot cm \) for the SQS mode and 2,450 \( \mu s \cdot cm \) for the GM mode. The difference in the dead zone is caused by the drift time of ions formed in the streamer toward the cathode and the electric field strength depending on the counter geometry, and not by the difference between \( \beta \)- and \( \gamma \)-rays\(^{(10)} \). In Table 1, the measured dead zones are summarized for different mixtures and counters with the previous results\(^{(1)} \).

4. Counting-rate Characteristics

Since the dead zone of the SQS tube with the mixture containing 2.5\% methylal is much smaller than that of the GM tube, the SQS tube can be employed under high exposure rates. Figure 6(a) shows the output counting rate as a function of the exposure rate of \( \gamma \)-rays. In the measurement the discriminator level was set at 1/10 of the SQS- or GM-signal amplitude obtained at a low exposure rate less than 1 mR/h \((0.258 \mu C/kg/h)\). The counting efficiency of the SQS tube was equal to that of the GM tube at the exposure rate. As shown in Fig. 6(a), the output counting rates from the SQS and GM tubes are proportional to the exposure rate up to about
Table 1  Dead zones for different mixtures and counters

<table>
<thead>
<tr>
<th>Mixture</th>
<th>Pressure (kPa)</th>
<th>Mode</th>
<th>Dead zone $d$ ($\mu$m·cm)</th>
<th>Mixture</th>
<th>Pressure (kPa)</th>
<th>Mode</th>
<th>Dead zone $d$ ($\mu$m·cm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ar-isoC$<em>4$H$</em>{10}$(75:25)</td>
<td>101.3</td>
<td>SQS</td>
<td>290</td>
<td>Ar-isoC$<em>4$H$</em>{10}$(75:25) + 2.5% methylal</td>
<td>101.3</td>
<td>SQS</td>
<td>230</td>
</tr>
<tr>
<td></td>
<td>20</td>
<td>GM</td>
<td>2,450</td>
<td>Ne-isoC$<em>4$H$</em>{10}$(75:25)</td>
<td>101.3</td>
<td>SQS</td>
<td>~1,500</td>
</tr>
<tr>
<td>Aloka 2008 GM Ar-ethylformate (96:4)</td>
<td>20</td>
<td>GM</td>
<td>5,100</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

$d$: Counter diameter,  $L$: Anode length

30 mR/h. Above this exposure rate the proportionality is lost for the GM tube, while is kept up to about 200 mR/h for the SQS tube. Counting loss of 50% is caused at 70 mR/h for the GM tube and at 250 mR/h for the SQS tube with the Ar-isoC$_4$H$_{10}$(75:25) mixture as shown in Fig. 6(b).

For the SQS tube with the Ar-isoC$_4$H$_{10}$(75:25) mixture containing 2.5% methylal, the exposure rate of the 50% counting loss is about 700 mR/h. The counting-rate characteristics of the SQS tube with this mixture, therefore, is improved by a factor of about 3 for the mixture without methylal as compared in Fig. 6(b). Consequently, the SQS tube with the mixture containing methylal can be applied in a region of exposure rates from the background level to about 700 mR/h.

The counting loss may be caused from a space-charge effect due to the SQS formation. Since the interval distribution of $\gamma$-rays from a source is generally represented by an exponential form, whose decay constant is proportional to the mean event-rate$^{(12)}$, events occurred within short intervals increase with the mean event-rate: The counting loss due to the dead zone (i.e. a local space-charge effect) is increased as the counting rate increases. From the measured dead zone of the SQS tube with the Ar-isoC$_4$H$_{10}$ mixture containing 2.5% methylal, the counting loss for uniform $\gamma$-ray irradiation was calculated as a function of exposure rate using a Monte Carlo method. As shown by a solid line in Fig. 6(b), the calculated result explains well the measured one at exposure rates less than 100 mR/h. Since $\gamma$-ray events at higher counting rates distribute in shorter time intervals where the dead zone is caused, the counting loss becomes nearly constant for exposure rates higher than 100 mR/h for uniform $\gamma$-ray irradiation of the tube.

At exposure rates higher than 100 mR/h, the calculation could not reproduce the measured counting losses. It may be necessary to
take into account the decrease in the pulse height of signals by another space-charge effect, because signals with pulse heights less than the discriminator level are not counted. Positive ions slowly drifting toward the cathode are accumulated in the whole counter volume during the irradiation at very high exposure rates, and hence produce space charges in the counter. The space charges cause the decrease in the pulse height of signals; this is called an averaged space-charge effect. Even for the SQS tube having very small dead zones, the decrease in the pulse height due to the averaged space-charge effect may be significant at very high exposure rates, although detailed discussion is not made in this paper.

IV. CONCLUSION

The properties of an SQS tube with the Ar-isoC,H / (75:25) mixture containing 2.5% methylal were investigated for uniform γ-ray irradiation. By the use of the mixture with methylal, avalanche sizes are shrunk, the streamer length is shortened, and the dead zone is decreased very much. Slightly electronegative methylal vapor as the second quenching gas is effective to improve the dead zone and counting-rate characteristics of the SQS tube. The SQS tube can be applied in a region of exposure rates from the background level to about 700 mR/h within 50% counting loss. The counting loss as a function of the exposure rate was explained by the local space-charge effect due to the dead zone up to about 100 mR/h. It is necessary to take into account the averaged space-charge effect to explain the counting loss at very high exposure rates.

ACKNOWLEDGMENT

We are grateful to Messrs. T. Oshima, S. Matsubara and M. Yoshizumi, Aloka Co. Ltd. for fabrication of a modified model of Aloka 2008 GM tube, and to Dr. T. Sakae for his helpful discussion. This work was supported in part by a grant-in-aid for scientific research from the Ministry of Education, Science and Culture, Japan.

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