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# Measurements of electron attachment by oxygen molecule in proportional counter

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#### ABSTRACT

We present pulse height measurements for 5-keV Auger electrons from a radioactive <sup>55</sup>Fe source mounted at the inner cathode surface of cylindrical proportional counter, which is operated with  $CH_4$  admixed dry air or  $N_2$ . A clear shift of the pulse height has been observed by varying the amount of the admixtures; the number of electrons, created in the primary ionization by Auger electrons, is decreased by the electron attachment of the admixtures during their drift from the place near the source to the anode wire. The large gas amplification (typically  $10^4$ ) in the secondary ionization of proportional counter makes it possible to investigate a small change in the number of primary electrons. The electron attenuation cross-section of  $O_2$  has been evaluated by analyzing the shifts of the pulse height caused by the electron attachment to dry air and  $N_2$ .

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# 1. Introduction

We have recently developed a proportional counter for the measurement of tritium gas in air with minute concentration below  $10^{-3}$  Bq per 1-cm<sup>3</sup> air. In this work, we have faced a problem on electron attachments of air in the counter gas consisting of CH<sub>4</sub>+10%Air, which shows apparent peak-shift on energy spectrum depending on the air concentration. The precise measurements of the distribution such as the peak position and the area are very important to get information on nuclide identification and its evaluating concentration. The peak-shift phenomenon is well-known as the negative-ion formation by the molecules such as O<sub>2</sub> and H<sub>2</sub>O contaminated in the counter gas, which causes some deterioration of time resolution and detection efficiency. Recently these influences in a long drift counter and in gaseous plasma have been investigated to optimize the performance of the detection system [1].

We have observed a similar phenomenon of electron attachments varying the concentration of dry air. For our original aim of developing proportional counter, we need to search the effect of electron attachments in measuring the concentration of tritium in the air atmosphere. Since the electron attachment is mainly caused by O<sub>2</sub> in the air, we have systematically measured pulse heights of 5-keV Auger electrons from thin <sup>55</sup>Fe source by varying the amount of dry air and N<sub>2</sub> in the counter gas, of which the main

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component was CH<sub>4</sub>; we avoided a risk of direct measurement using O<sub>2</sub> gas, because an admixed gas with O<sub>2</sub> and CH<sub>4</sub> indicates an explosion hazard in the discharge mode of operating proportional counter. In the present counter, the primary electrons, which are created near the cathode surface by ionization of counter gas by the Auger electrons, drift to the anode wire through the counter gas with small amount of O<sub>2</sub>. Then, they are multiplied by a factor of  $10^3-10^4$  by secondary gas amplification known as the Townsend avalanche [2]. Therefore, the output from the proportional counter is very sensitive to the electron attachment, which can be observed as the peak shift on the spectrum. Using this basic mechanism of proportional counter, we have extracted attenuation cross-section of O<sub>2</sub> by the subtraction of the result of N<sub>2</sub> from that of dry air.

In this paper, we present our experimental method to observe the peak shift caused by the electron attachment of admixture gases with positive electron affinity. The analysis to deduce electron attenuation cross-sections of admixture gases from the peak shift is also explained.

# 2. Experimental and method

We have measured the energy spectra of 5-keV Auger electrons from the <sup>55</sup>Fe source with the proportional counter, which consists of a cylindrical cathode of 50-cm length and 5-cm inner diameter and a tungsten wire anode of 30-µm diameter. The <sup>55</sup>Fe source was set on the inner wall of the counter. Main component of the counter gas was CH<sub>4</sub>. Although <sup>55</sup>Fe also emits 5.9-keV Mn K X-ray, CH<sub>4</sub> is not effectively ionized by the X-rays comparing with





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5-keV Auger electrons. The energy spectra have been obtained as a function of partial pressure of admixtures, i.e., dry air or N<sub>2</sub>, which was ranged from 0% to about 16% within the total pressure of 0.5 atm. The counter gas was sealed in the cathode vessel. The gas pressure was measured by an instrument which has a gas blending system with fine control-regulators and a column-absorption for removing water vapor. The uncertainty of gas pressure was estimated to be within 3%. The 5-keV Auger electron makes electron–ion pairs by the ionization of counter gas. The total number of ion pairs by the ionization is estimated to be 180 pairs, because the *W*-value of CH<sub>4</sub> is 27.3 eV [3]. The primary electron cloud was created at very close area of the <sup>55</sup>Fe source, only several millimeters away. A high voltage of 2000 V was applied to the anode wire, which corresponds to the gas-amplification factor of about 8000.

The output voltage *V*, i.e., the pulse height of output from a proportional counter is given by

$$V = \frac{Q}{C},\tag{1}$$

where *Q* is the total electric charge created at the anode wire and *C* is a capacity of the proportional counter. The value of *Q* changes due to the attachment of the primary electrons by admixture gases during drifting from the <sup>55</sup>Fe source to the anode wire. An attenuation factor is denoted by  $\exp(-\sigma \cdot t)$ , where  $\sigma$  is the cross-section of attenuation in the unit of cm<sup>2</sup> and *t* is the density of admixture gas in the unit of cm<sup>-2</sup>. Thus the value of *Q* is given by

$$Q = n_0 \cdot e \cdot \exp(-\sigma \cdot t) \cdot M, \tag{2}$$

where  $n_0 \cdot e$  is the total charge of primary electrons and *M* is the gasamplification factor which depends on gas pressure and anode voltage. Both  $n_0 \cdot e$  and *M* in Eq. (2) are constant under a given condition in the present measurements, i.e., the total pressure and anode voltage were respectively maintained at constant values of 0.5 atm and 2000 V. For the gas-amplification, the secondary ionized ion-pairs created around anode wire are hard to recombine in the strong electrical field. Then the avalanche electrons quickly move to the anode wire. Therefore the gas-amplification factor *M* is considered as a constant. Thus the observed change of *Q* results from the attenuation of electrons expressed by the factor of  $exp(-\sigma \cdot t)$  in Eq. (2).

#### Air = 14.5% 13 2% 2000 Air Counts/Channel 9 21% 1500 7.89% 6 58% 5 26% % 2.63% 1.32% <sub>0%</sub> 1000 500 0 200 400 600 800 1000 Channel number

**Fig. 1.** Energy spectra of the Auger electrons measured by the proportional counter filled with mixture of  $CH_4$  and dry air; the concentration of dry air is changed from 0% to 14.5% within the total pressure of 0.5 atm.

## 3. Results and discussion

Fig. 1 shows energy spectra of the Auger electrons obtained with the present proportional counter; the counter gas is a mixture of main component CH<sub>4</sub> and dry air of which partial pressure ratio is ranging from 0% to 14.5%. Fig. 2 shows the spectra obtained with the admixture of N<sub>2</sub> ranging from 0% to 15.8%. The peak positions in both figures decrease with increasing the amount of the admixtures; with the increase of dry air in the counter, the number of primary electrons arrived at the anode wire is more decreased because of the electron attachment by dry air. Peak profiles are not symmetry. The spread tail in the left side of peak is caused by an energy loss at the surface of <sup>55</sup>Fe source. The width of the peak profile of Auger electrons obtained from the right side of the peak,  $\Delta Q$ , is given by

$$\left(\frac{\Delta Q}{Q}\right)^2 = \left(\frac{\Delta n_0}{n_0}\right)^2 + \left(\sigma \cdot \Delta t\right)^2 + \left(\frac{\Delta M}{M}\right)^2,\tag{3}$$

where  $\Delta n_0$ ,  $\Delta t$  and  $\Delta M$  indicate statistical fluctuations of  $n_0$ , t and M in Eq. (2), respectively.

The response function of peak profile was obtained by fitting spectrum data for pure CH<sub>4</sub> with polynomial equation, as shown by the blue curve (air = 0%) in Fig. 1 or the curve (N<sub>2</sub> = 0%) in Fig. 2. Other spectrum data were fitted with this response function to obtain peak positions and gross area normalized to that of the response function; fitted profiles, which are normalized to the height of corresponding observed spectrum, are shown by red curves in Figs. 1 and 2. As seen in these figures, the gross area is almost same for all the profiles, meaning that the total number of counted events is same for all spectra.

It is seen in Fig. 1 that, as the concentration of dry air is larger, the width of observed spectrum is wider comparing with that of the corresponding fitted profiles. This large fluctuation of Q is explained by the second term ( $\sigma \cdot \Delta t$ ) in Eq. (3), not first and third terms.

The attenuation factor given by  $\exp(-\sigma \cdot t)$  in Eq. (2) is experimentally obtained by a ratio of the peak position to that of the spectrum with pure CH<sub>4</sub> (air = 0% or N<sub>2</sub> = 0%). Results are shown in Fig. 3; the solid circle is the attenuation factor for dry air and the open circle is that for N<sub>2</sub>. The error was estimated to be 2–5% for each point. In Fig. 3, the attenuation factor is plotted as a function of the density of molecules, evaluated from the partial pressure of admixture gas within the total pressure of 0.5 atm for the counter gas. The density was calculated from the measured



**Fig. 2.** Energy spectra of the Auger electrons measured by the proportional counter filled with mixture of  $CH_4$  and  $N_2$ ; the concentration of  $N_2$  is changed from 0% to 15.8% within the total pressure of 0.5 atm.

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