Gas Gain in Air-mixed Argon-Propane (1%)-based Proportional Counters

D. Mazed\* and R. Ciolini

Dipartimento di Ingegneria Meccanica, Nucleare e della Produzione, Università di Pisa
Via Diotisalvi, 2, I-56126, Pisa, Italy. E-mail: dahmane.mazed@studenti.ing.unipi.it

Abstract. For the design of a special gas-flow proportional counter intended for a new on-line measurement method of airborne radon ($^{222}$Rn) concentration, the amplification properties of four gas compositions were investigated in air-mixed Argon-propane (1%)-based proportional counters. These four gas compositions are: Ar-C$_3$H$_8$ (1%); Ar-C$_3$H$_8$ (1%) + 2% air; Ar-C$_3$H$_8$ (1%) + 6% air and Ar-C$_3$H$_8$ (1%) + 10% air. The influence of the electron attachment effect due to oxygen on the saturation and gas amplification characteristics was examined in particular. The semi-microscopic gas gain formula was found to describe accurately the gas gain data obtained in each of the gas mixtures analyzed allowing the determination of the gas constants. The use of air-mixed counting gases in a specially designed gas-flow proportional counter for airborne radon concentration measurements appeared quite viable.

1. Introduction

Being a radioactive noble gas, Radon ($^{222}$Rn) presents some very particular properties: it diffuses easily out of soils or building materials and easily spreads out throughout the environment. Consequently, different concentrations of $^{222}$Rn and of its progenies can be found everywhere around us. Since it is an inert gas, $^{222}$Rn does not react chemically with other atoms or molecules. This makes its chemical trapping impossible and its physical trapping also difficult. Due to the importance of radon activity concentration for radiation protection, several detection systems and various measurement methods were developed in the past aiming at an accurate assessment of its concentration in air. However, most methods present the limitation of not being able to measure on-line the radon activity concentration directly, being often affected by somewhat low detection efficiency. Therefore, they did not provide the performance required in practice for an effective on-line measurement of low and medium radon activity concentrations (1 Bq·m$^{-3}$ to 10 kBq·m$^{-3}$). This feature is indeed of fundamental relevance for the study of the time variations of radon airborne concentrations and expositions, especially when a correlation with environmental parameters is sought.

2. The use of gas-filled detectors for radon concentration measurement in ambient air

Few laboratories worldwide focus their research on the use of gas-flow proportional counters for the continuous monitoring of radon concentration in air. In fact, the detection of $\alpha$ particles in air is quite difficult to achieve with acceptable accuracy, due to the presence of electronegative oxygen, water vapour and other electronegative gas traces. Nevertheless, some research groups [1-6] have already made significant progress in this direction. Their major results are briefly reviewed below.

Baltzer et al. [1] reported about a pulse–counting ionization chamber for measuring radon concentration in air. Their device is based on both positive and negative ion collection (due to electron attachment), thus it is severely limited by the excessively long collection times involved. Klein et al. [2] mentioned the possibility of using proportional counters for in-situ radon level characterization, but gave no details. Zikowsky [3] tried to measure radon concentrations in air by performing alpha counting of solid radioactive progenies ($^{218}$Po and $^{214}$Po mainly) deposited on the inner walls of a proportional counter. He first filled the proportional counter with an air sample, and then evacuated the counter after periods of time ranged between 30 and 216 min, immediately introducing an appropriate counting gas (P-10 gas) to carry out alpha counting of the particles emitted by the radon decay products. This method is not as sensitive as direct radon counting. Indeed, in addition to the fact that it cannot provide on-line measurements, it has at least a major inconvenient: the equilibrium factors are not known exactly, which is of fundamental importance [4].

\*Also: Commissariat à l’Energie Atomique, Centre de Recherche Nucléaire de Birine, BP.180 Ain-Ousséra, 17200, Djelfa, Algeria.
In order to make on-line low- and medium-level measurements of the radon activity, Rottger et al. [5] also used a wire ionization chamber similar to the one described by Baltzer et al. Their device was designed to permit adequately large volumes (from 5 to 13 litres) and presented a special electrode configuration (Archimedean spiral layout). However, the device was also based on ion collection. Therefore, it is still subjected to considerable limitations such as microphonics, high collection times and poor energy resolution. Nevertheless, the authors indicated that their device can be able to separate between alpha particles emitted from $^{222}\text{Rn}$ and those originating from $^{218}\text{Po}$ in the recorded pulse height spectra.

Recently, Busch et al. [6] reported on the first use of a gas-flow proportional counter for performing direct radon concentration measurements. The radon gas however emanates from a radium source cell. Indeed, the gas-flow proportional counter used does not allow measurement in air; there is henceforth no electron attachment problem due to oxygen. Instead, the counter is filled with an adequately chosen counting gas (P-10 gas: Argon-methane 90%-10%) in which a known fraction of radon gas emanation from the radium cell is also mixed.

To the best of our knowledge, no attempt was reported about the use of gas-flow proportional counters directly and specifically designed to measure the radon concentration in air by counting only the $\alpha$ particles emitted by $^{222}\text{Rn}$ itself. This feature indeed would imply that the counter sensitive volume must be partly, if not wholly, filled by a certain fraction of the air sample to be monitored. This prompted us to make a subsequent investigation on the amplification characteristics of air-mixed counting gases in proportional counters.

The amplification properties of four gas compositions were thus investigated in air-mixed Argon-propane (1%)-based single wire proportional counters for a new on-line measurement method of airborne radon ($^{222}\text{Rn}$) concentration. These four gas compositions are: Ar-C$_3$H$_8$ (1%); Ar-C$_3$H$_8$ (1%) + 2% air; Ar-C$_3$H$_8$ (1%) + 6% air and Ar-C$_3$H$_8$ (1%) + 10% air.

3. The gas amplification and electron attachment in air-mixed gases

When operated in the ionization regime, an air-filled detector delivers too small electric pulses and these are also characterized by strong pulse height fluctuations. Because of these limitations, electron collection-based air-filled ionization chambers can no longer be used for our purpose. Indeed, the electron attachment by the oxygen molecules contained in air will reduce drastically the number of the primary electrons produced within the sensitive volume and, consequently, a reliable linear response of the recorded counting and height pulses becomes very difficult to achieve in practice. However, in proportional counters a number of primary electrons not attached by electronegative molecules will reach the anode wire where they are amplified up to $10^3$ or even $10^4$ times by the high voltage applied to the anode. The attached electrons lost during the drift step will then be largely compensated through subsequent gas multiplication. This way, the detection of charged particles becomes quite possible.

When released in the sensitive volume of gas-filled detectors, primary electrons undergo well-known elastic and inelastic processes [7]. Among these processes one has to pay careful consideration to the electron attachment by electronegative molecules such as oxygen, especially in the case when an important fraction of this gas, contained in atmospheric air, is intentionally mixed with the main counting gas used in the counter. The attachment effect is well known, both fundamentally [7, 8] and also regarding its practical implications in usual gas-filled detectors [9,10] such as ionization chambers, proportional counters, drift chambers etc. In case of proportional counters, we can identify two main evolving regions: the drift space region and the gas amplification region, also called Townsend avalanche region. In the former, the electric-field-to-gas-pressure ratio, $S(r) = E(r)/P$, is characterized by rather low values, typically not higher than 35 to 45 V/cm torr. The primary electron energy is always lower than the first ionization potential of the main filling gas and there is no electron multiplication. The electron attachment however, because of the lower values of $S(r)$, has such a considerable effect that even the detection process of the counter could be totally compromised. By contrast, within the Townsend avalanche region $S(r)$ generally has sufficiently high values, such that a great fraction of the avalanche electrons can ionize the gas and create further electrons along their last transit step, just before reaching the anode wire. In this region, the influence of electron attachment effect is minor. This is due to the shorter distances travelled by avalanche electrons,
typically 5 to 10 times the wire radius, and also to the higher energies reached by the electrons constituting the avalanches. The electron attachment cross section indeed is quite negligible for electrons of energy higher than 9 eV, which are predominant in that region. This is illustrated in figure 1, which shows the electron energy dependence of the dissociative electron attachment cross section of the oxygen molecule. Consequently, one can expect that the electron attachment process introduces only a negligible effect on the gas amplification characteristics, even in the presence of an important fraction of electronegative gas molecules, as is the case here.

**FIG. 1. The electron energy dependence of the dissociative electron attachment cross section of oxygen**

Under these conditions, and according to the semi-microscopic gas gain formula taken to its 3rd order approximation, the natural logarithm of the resulting gas gain \( A \) in a single wire proportional counter, is given by [11]:

\[
\frac{\ln A}{P \cdot a \cdot S_o} = K \left[ (1+m) \ln \left( \frac{S_o}{S_0} \right) + \frac{1}{1!} \left( \frac{S_0}{S_o} \right)^{(1+m)} - \frac{1}{2!} \left( \frac{S_0}{S_o} \right)^{(2+m)} + \frac{1}{3!} \left( \frac{S_0}{S_o} \right)^{(3+m)} \right] - L \quad (1)
\]

with,

\[
S_o = \frac{V}{P \cdot a \cdot \ln(b/a)} \quad (2),
\]

where \( V \) is the applied voltage, \( P \) is the total gas pressure in the counter, \( a \) and \( b \) are respectively the anode and cathode radii. The expression in brackets, which depends only on external, physical and electrical parameters, is called the control variable and is noted \( X \). It does not in any way depend on the gas mixture nature, whereas the constants \( S_0, m, K, L \) characterize the physical nature of the gas mixture inside the counter. These are determined on the basis of a suitable fitting analysis of the experimental set of gas gain data at hand. When the left hand side term of equation (1) is plotted versus the control variable \( X \), a straight line should be obtained. Then, by least squares fitting, the constants \( K \) and \( L \) are easily determined. The physical significance of the gas constants and also the method used for the determination of the primary gas constants \( m \) and \( S_0 \), are extensively discussed in our previous contributions [11-13]. The moderation parameter \( m \) is related to the shape of the cross sections of the major inelastic processes involved, whereas \( S_0 \) is interpreted as the critical value of the electric field to pressure ratio for which the gas amplification process starts-up and its value is connected to the value taken by the moderation parameter \( m \). We can also deduce another more explicit gas constant, \( \Delta V_{max} \), given by:

\[
\Delta V_{max} = \frac{1}{(1+m)K} \quad (3)
\]

This can be interpreted as the maximum potential difference travelled by an avalanche electron between two successive ionizing collisions. By studying the gas constants values of the air-mixed argon-propane (1%) mixtures, one can assess better the net effect of the air on the amplification properties, in order to choose the most suitable composition for the above-mentioned purpose.
4. Experimental set-up and procedure

The proportional device used in the gas gain measurements is a gas-flow proportional counter with 0.001 inch (25.4 micron) tungsten wire and a stainless steel cathode tube of 32 mm diameter. The counter was operated at atmospheric pressure and at room temperature (∼296°K) for all the gas mixtures examined. The gas-flow rate was set at 1 l/min and kept constant during the whole measurements. The counter and the gas mixture manifold are shown in figure 2. A silicagel cell is inserted in the air circuit in order to remove evenly the excess water vapour present in the atmospheric air and then prevents to involve high humidity rate air samples. The ionizing radiation source was placed directly inside the counter, on the cathode surface, at the mid point of the anode wire. Two radiation sources were used: a multi-alpha source formed by a mixture of \( ^{239}\text{Pu} \), \( ^{241}\text{Am} \) and \( ^{244}\text{Cm} \) radioisotopes, for low- and medium- gas gain levels, and \( ^{55}\text{Fe} \) radioisotope emitting 5.9 keV X-rays for the higher gas gain values, in order to avoid excessive space charge accumulation around the anode wire. First, we examined the amplification characteristics in the Argon-propane (1%) counting gas mixture free from air then, successively added 2 %, 6% and 10 % of atmospheric air. For each gas composition, the amplification characteristics were measured.

The pulse signals delivered by the proportional counter were fed to a low-noise charge-sensitive preamplifier (ORTEC 142 PC). The preamplifier output is in turn fed to the built-in amplifier of the MCA (Silena, SNIP 204G). The pulse shaping time was fixed at 2 µs and the amplifier gain was varied according to the pulse height delivered by the preamplifier. Typically, two amplifier gain settings were used: 200 and 10. A gain of 200 was chosen when very low amplitude signals from the preamplifier were processed, when the counter operates in the ionization chamber regime and also when it operates at low gas gain values in the proportional regime. A gain of 10 was adopted for higher gas gain operation. The experimental gas gain measurement method we used is based on the measurement of the channel number corresponding to the centroid of the main peak in the spectrum as a function of the high voltage value. At low gas gain values, when the multialpha source was used, the gas gain \( A(V) \) at the set voltage \( V \) was simply computed by:

\[
A(V) = \frac{N^\alpha(V) G^\alpha}{N^\alpha_s G^\alpha_s} \quad (4),
\]
where, $N_S^\alpha$ is the channel number of the alpha reference peak when the counter operates at the saturated ionization chamber regime and $N_S^\alpha(V)$ is the channel number of the same peak at the set value $V$ of the high voltage, whereas, $G_v^\alpha$ and $G_V^\alpha$ are the amplifier gain respectively in the saturated ionization chamber regime and when the counter is operated at the high voltage $V$, using the multialpha radiation source. At higher voltage values, we replaced the multialpha source by the 5.9 keV X-ray $^{55}$Fe source, the gas gain $A(V)$ was computed by:

$$A(V) = \frac{N_S^X(V)}{N_S^\alpha} \frac{G_v^\alpha}{G_V^\alpha} \frac{E_X}{E_\alpha} \frac{W_X}{W_\alpha}$$

(5),

where, $N_S^X(V)$ is channel number corresponding to the 5.9 keV peak recorded for the high voltage $V$, $G_v^\alpha$ is the adopted amplifier gain when the counter was polarized at voltage $V$, whereas, $E_X$ and $E_\alpha$ are the energy expended by the ionizing radiation ($X$ and $\alpha$, respectively) in the sensitive gas. $W_X$ and $W_\alpha$ are the energies necessary for one electron-ion pair production in the sensitive gas mixtures respectively by X-ray photons and by $\alpha$ particles. However, we neglected the small differences that exit between the two values, and set the ratio $(W_X/W_\alpha)$ equal to unity.

**FIG.3. Evolution of the gas amplification characteristics in the air-mixed argon-propane (1%) based gas-mixtures**

### 5. Results and discussion

For each gas mixture examined, the $A(V)$ amplification characteristics was plotted, as shown in figure 3, where we can notice the important effect of air admixture. Using the Ar-$C_3H_8$ (1%) free from air and setting the high voltage to about 800 volts, it is possible to achieve a gas gain of 100. But, the addition of just 2 % air to the counting gas mixture, requires increasing the high voltage up to 1200 volts in order to get the same level of gas gain. For 6 % air admixture, a gas gain of 100 is achieved at a voltage of about 1400 volts, and when 10 % of air is admixed to the counting gas, the high voltage value must be set at 1600 volts, twice the value used in air-free argon-propane (1%). This is expected, since the electrons need much more energy to compensate the losses through inelastic collisions which they increasingly undergo during their avalanche process. In other words, the increased voltage value has the net effect to preserve the total number of avalanche electrons able to produce subsequent secondary electrons, at almost the same fraction level as it was in case of air-free argon-propane (1%) gas mixture. The effect of the electron attachment, particularly predominant in the drift space, can be noticed in figure 4, which enlarges the recombination region and the saturated ionization chamber.
FIG. 4. Evolution of the saturation plateau slope in air-mixed argon-propane (1%) based gas-mixtures

FIG. 5. The fitting curves of the 3rd order approximation of the semi-microscopic formula to the gain data in the examined air-mixed Ar-C₃H₈ (1%) based gas mixtures

regime of the amplification characteristics (figure 3). We can notice that the effect resulting from electron attachment is a progressive degradation of the saturation characteristics. This translates in a progressive increase of the saturation plateau slope when we compare the saturation curves achieved increasing the air fraction in the mixture from 0% to 10%. For a given high voltage value, this effect is to be expected since the greater the oxygen fraction in the gas mixture, the lower the number of primary electrons which cross the drift space without being attached by an oxygen molecule.

The drift space is the region between their electron production and their entry in the avalanche region, located in the immediate vicinity of the central anode wire. Table 1 summarises the saturation plateau slopes as a function of air fraction in the gas mixture. We can notice the expected increase of the saturation plateau slope as a result of the increase of the air fraction in the gas mixture. By increasing the high voltage value, and hence the electric field to pressure ratio, the electron energy distribution function in the drift space is shifted to higher energy side. Thus, the net effect is a reduction of the electron attachment rate, since this effect is predominant at lower electron energies. Therefore, the fraction of collected electrons is then re-established again without further gas amplification.

The gas amplification characteristics are studied using the semi-microscopic formula given by equation 1. The fitting curves from this formula for our measured gas gain data are shown in figure 5. As expected, the 3rd order approximation of the semi-microscopic gas gain formula describes well the obtained gas gain data, despite the attachment effect due to oxygen. The correlation coefficient of the

<table>
<thead>
<tr>
<th>Air fraction admixed</th>
<th>0%</th>
<th>2%</th>
<th>6%</th>
<th>10%</th>
</tr>
</thead>
<tbody>
<tr>
<td>Saturation Plateau Slope (% / 100 V)</td>
<td>8</td>
<td>10</td>
<td>14</td>
<td>19</td>
</tr>
</tbody>
</table>

TABLE II. The gas gain constants obtained from the fitting analysis of the measured gas gain data in Ar-C₃H₈ (1%) and air-mixed Ar-C₃H₈ (1%) based proportional counters.

<table>
<thead>
<tr>
<th>Air fraction admixed</th>
<th>0%</th>
<th>2%</th>
<th>6%</th>
<th>10%</th>
</tr>
</thead>
<tbody>
<tr>
<td>m</td>
<td>0.333</td>
<td>0.500</td>
<td>0.530</td>
<td>0.550</td>
</tr>
<tr>
<td>S₀ (V/cm torr)</td>
<td>43.5</td>
<td>65</td>
<td>75</td>
<td>80</td>
</tr>
<tr>
<td>Correlation coefficient</td>
<td>0.9991</td>
<td>0.9992</td>
<td>0.9958</td>
<td>0.9997</td>
</tr>
<tr>
<td>K (10⁻³ V⁻¹)</td>
<td>47.1</td>
<td>24.8</td>
<td>18.7</td>
<td>15.8</td>
</tr>
<tr>
<td>L (10⁻³ V⁻¹)</td>
<td>28.7</td>
<td>13.7</td>
<td>9.2</td>
<td>8.2</td>
</tr>
<tr>
<td>ΔVₘₐₓ (V)</td>
<td>15.9</td>
<td>26.9</td>
<td>35.0</td>
<td>40.8</td>
</tr>
</tbody>
</table>
linear fitting is greater than 0.99 in all cases and 0.9997 is even achieved in case of Ar-C\textsubscript{3}H\textsubscript{10} (1%) + 10% air. The obtained gas constants are summarised in table 2. With the increase of the air fraction, the values taken by the moderation parameter $m$ increase from 0.33 up to 0.55; the trend is almost the same for $S_0$, the critical value for gas amplification. These trends can be interpreted considering that the increasing air fraction in the gas mixture contributes to increase the mean fraction of the total energy that the electrons lose through inelastic non-ionizing collisions. As a result, the electron temperature in the avalanches decreases (the cooling effect or moderation effect). This involves an increase of the parameter $m$, introduced in the semi-microscopic formula. The total energy losses through inelastic collisions excite the vibration and optical states of the nitrogen and oxygen molecules present in the gas with an increasing relative density. On the other hand, the increase of $S_0$, shown in figure 6-a, means that the volume of the avalanche region decreases and collapses around the central wire, since the electric field distribution around the central anode wire is $1/r$, where $r$ is the radial position. Therefore this results in the reduction of the gas gain for a fixed value of the high voltage. This effect appears quiet well in figure 6-b, showing the evolution of the maximum potential difference $\Delta V_{\text{max}}$ travelled by an avalanche electron between two successive ionizing impacts.

$$\text{FIG. 6. Evolution of the gas constants } S_0 (\text{a}) \text{ and } \Delta V_{\text{max}} (\text{b}) \text{ in air-mixed Ar-propane(1\%) based gas mixtures as a function of the admixed air fraction.}$$

6. Conclusion

To achieve more reliable on-line measurements of airborne radon concentration using gas-filled detectors based on alpha spectrometry technique, it is required to perform the measurement of the radon concentration within a sensitive volume by counting directly only the alpha particles emitted by radon nuclides. This implies the need to mix with the counting gas at least a small fraction of the air sample to be assessed. Therefore, the air sample becomes part of the sensitive gas mixture. Unfortunately, the presence of oxygen, a highly electronegative element, introduces serious problems related in particular to the attachment effect causing an excessive electron charge loss within the drift space. However, the electron attachment effect can be counterbalanced by gas amplification in the proportional counters. In this paper, we present the gas amplification characteristics of three air-mixed argon-propane (1\%)-based gas mixtures. Our results show that the attachment effect does not introduce a critical perturbation within the gas amplification region, but it affects only the charge collection process within the drift space of the proportional counter, where the gas amplification process can compensate for the primary electrons lost through the attachment effect. Hence, it seems quite possible to reduce the electron losses by designing special proportional counters that minimize the drift space dimensions. The presence of a fraction of air as high as 10\% does not seem to penalise excessively the detection process if an appropriately designed gas-flow proportional counter is used.

Acknowledgements

We would like to thank particularly Dr. F. d’Errico for his useful comments and Mr. A. Del Gratta and F. Pazzaglia for their precious technical assistance in the preparation of our apparatus.
References


