A STUDY OF ARGON–ISOBUTANE MIXTURES IN A PROPORTIONAL COUNTER: GAS AMPLIFICATION, W VALUE, AND ENERGY RESOLUTION

I. Krajcar Bronić*
Rudjer Bošković Institute
PO Box 1016, 10001 Zagreb, Croatia

Abstract — The mean gas amplification factor, the single electron spectra, the mean energy required to form an ion pair (W), and the energy resolution were measured in argon–isobutane mixtures at various pressures over a wide range of the reduced electric field strengths in a cylindrical proportional counter. In mixtures with low isobutane partial pressure, gas gain is an over-exponential function of the applied high voltage. The relative variance of single electron spectra, the apparent W value and the energy resolution are functions of both gas gain and isobutane partial pressure in the mixture. A better energy resolution is obtained with mixtures containing less isobutane at low gas gains whereas at the higher gas gains usually applied in X ray spectroscopy the resolution is degraded.

INTRODUCTION

Rare gas with hydrocarbon mixtures are often used as counting gases for proportional counters because of their attractive properties when compared with the pure gases. In Penning mixtures de-excitation of metastable states of rare gas atoms may result in the ionisation of admixed polyatomic gas molecules if their ionisation energy is lower than the excitation energy of the rare gas metastable state. Due to this enhanced ionisation in Penning mixtures as compared to that in pure gases and in regular mixtures the Townsend ionisation coefficient is higher, gas gain is increased, starting and operating potentials are lower, W value (mean energy required to form an ion pair) and the Fano factor F (a measure of statistical fluctuations in the number of primary ion pairs) are lowered. It has also been generally believed that the relative variance (f) of the gas amplification factor, i.e. of the so-called single electron (SE) spectrum, is lowered. The energy resolution R of the counter is a function of W, F and f, $R^2 \propto W(F+f)$, so it is expected that by using a Penning mixture one can significantly improve the energy resolution of the proportional counter, which is an important fact in spectroscopy of low energy X rays[1–3].

The argon–isobutane Penning mixture, in which excited argon atoms can transfer their energy to isobutane molecules and thus ionise them, was chosen for a comprehensive study of gas mixture characteristics in a proportional counter[4]. The mean gas amplification factor M (gas gain), the statistical fluctuations f of the gas amplification, the W value, and the energy resolution R were studied. The main aim was to determine the dependence of all mentioned quantities on the isobutane concentration fraction and on the total pressure of the mix-

ture, to find the dependence of f and R on the gas gain, and to define mixture parameters (composition, pressure) which would result in an optimal performance of proportional counters. A summary of the most important experimental results is presented here, while the detailed discussions on each measured quantity will be presented in forthcoming papers.

EXPERIMENT

The experimental method follows that of Srdoč[5,6], Srdoč and Clark[7], and Srdoč et al[8]. A specially designed proportional counter for low energy photon detection was used. Pulse height distributions produced by multiplication of initial charges in a gas-filled proportional counter were measured. The X rays from a ^{55}Fe source (5.89 keV) entered the active counter volume through a 5 μm beryllium window. Single electrons incapable of producing immediate ionisation were released from a thin Al film deposited on a quartz rod and irradiated by a UV lamp. A source of single electrons enables calibration of the ^{55}Fe spectra in terms of number of electrons per ionising event. The method was used for W value measurements in pure gases and regular gas mixtures[8], and the results obtained agreed well with the W values measured by other methods. The electronic part of the experimental set-up consisted of a high voltage supply, a preamplifier, a shaping amplifier, and a multichannel analyser with the on-line VAX computer.

Argon of purity >99.9996% and isobutane of purity >99.95% were used. For total pressures of 10, 20, 30, 60 and 90 kPa several mixtures with varying concentration fractions of isobutane were prepared. The lowest partial pressure of isobutane was 1 kPa.

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RESULTS

Gas amplification

For each gas mixture a mean gas amplification factor was measured as a function of the applied high voltage. Several gas gain curves are presented in Figure 1. In pure isobutane the gas amplification is, at all pressures, an exponential function of the applied voltage in the whole range of gas gains.

In argon–isobutane mixtures the high voltage required for the same gas amplification decreases with decreasing isobutane concentration fraction in the mixture. At the same time, the slope of the gas gain curve becomes steeper. In gas mixtures with isobutane partial pressure, pp > 15 kPa, gas gain is an exponential function of the applied voltage, up to the highest gain obtained, $M = 2 \times 10^4$. As the concentration fraction of isobutane decreases, the range of linearity of the gas gain becomes narrower, and the upper limit of the gas gain still in the linear region becomes lower. Above that limit, the gas gain increases faster than exponentially with high voltage. This over-exponential effect is more pronounced at lower total pressures indicating that the linearity region is not defined by the concentration fraction of isobutane in the mixture, but rather by its total amount, i.e. by its partial pressure[9]. The upper limit of the exponential functional dependence of the gas gain decreases from $M \approx 5000$ at pp = 12 kPa, to only $M \approx 700$ at pp = 1 kPa. The over-exponential increase

of the gas gain is attributed to an 'avalanche chain' formation[10]. The amount of isobutane is not sufficient to quench all the photons emitted by excited argon atoms. These photons initiate additional avalanches that join the main avalanche, thus forming the avalanche chains and increasing the measured gas amplification.

Single electron spectra

A single electron (SE) spectrum represents the statistical distribution of avalanche sizes triggered by single electrons with insufficient energy to produce immediate ionisation in a gas. The relative variance f of this distribution may contribute towards a better understanding of the working mechanisms of proportional counters[10]. Each SE spectrum was fitted by a Polya distribution[5] of the form $P(x) = a\,x^b\,e^{-cx}$, where x is a channel number, and a, b, and c are constants characteristic of a gas. The relative variance f was calculated as

$$f = \frac{1}{1+b} \tag{1}$$

In pure isobutane the SE spectrum was a peaked-shaped distribution with f < 1 over the whole range of gas gains and f was increasing slightly with gas gain. The increase of f with increasing gas gain was also observed in all argon–isobutane mixtures. At low gains, $M \leq 1000$, all SE spectra have a peaked shape (f < 1). The peaked shape remains at all gas gains (up to 2×10^4) for mixtures with at least 6 kPa of isobutane.
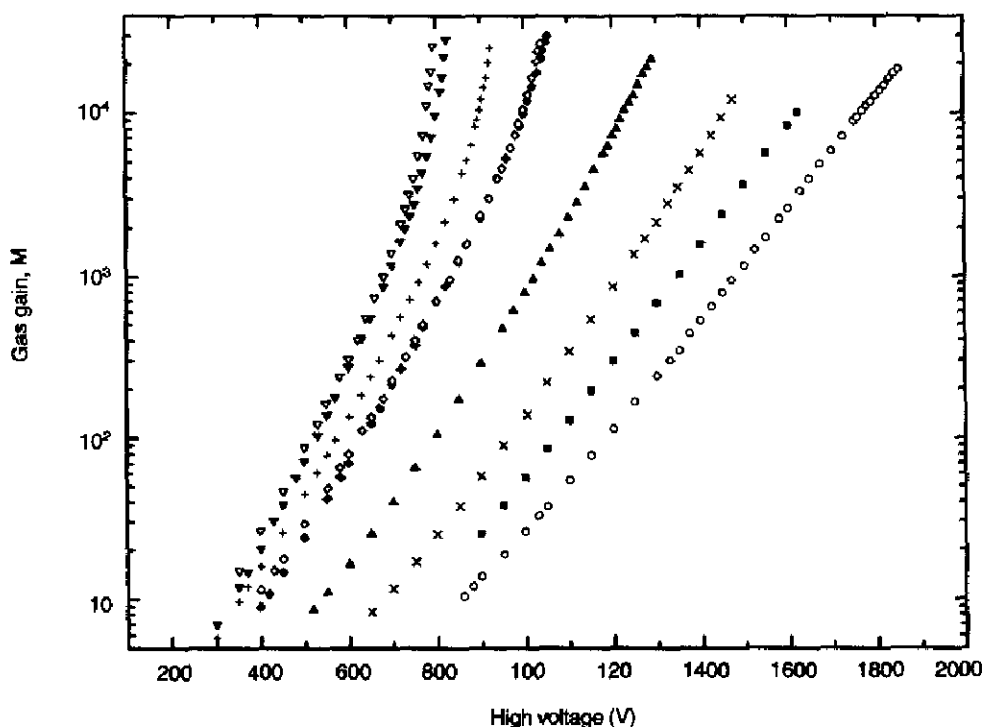

Figure 1. Gas gain curves for several argon–isobutane mixtures: (○) 30% isobutane, total pressure 90 kPa; (■) pure isobutane, p = 30 kPa; (×) 20% isobutane, 60 kPa; (▲) 6.6%, 90 kPa; (◆) 10%, 30 kPa; (◇) 30%, 10 kPa; (+) 20%, 10 kPa; (▼) 3.3%, 30 kPa; (∇) 10%, 10 kPa.

The SE spectra become exponential (f = 1) or hyperbolic (f > 1) in shape at higher gas gains in mixtures with pp < 6 kPa. The increase of f with gas gain is faster in mixtures with lower isobutane partial pressure. This effect is again caused by avalanche chain formation. For a given gas gain f is a function of isobutane partial pressure (Figure 2) regardless of the total pressure.
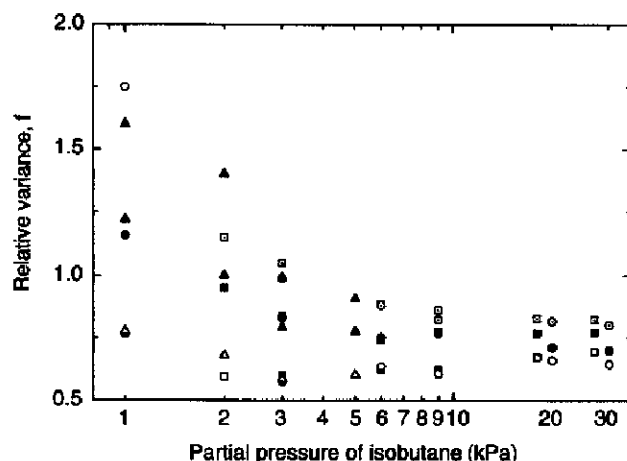

Figure 2. Relative variance f of the single electron spectra at three different gas gains as a function of isobutane partial pressure. Gas gain M = 1000: (□) p = 90 kPa, (○) 30 kPa, (△) 10 kPa. M = 3500: (■) 90 kPa, (●) 30 kPa, (▲) 10 kPa. M = 10⁴: (▨) 90 kPa, (⊙) 30 kPa, (▵) 10 kPa.

Apparent W value

The mean energy required to form an ion pair (W) as determined by the proportional counter method[6] determined as:

$$W = T\frac{x_{SE}}{x_{Fe}}\frac{G_{Fe}}{G_{SE}} \qquad (2)$$

where T is the photon energy (5.89 keV), x_{SE} is the mean value of the SE spectrum calculated as $x_{SE} = (1+b)/c$ (b and c are coefficients of the Polya distribution), x_{Fe} is the mean value of the measured Fe pulse height spectrum averaged over its frequency distributions, and G_{SE} and G_{Fe} are the corresponding electronic gains. Equation 2 is valid if the proportional counter is operated under strictly proportional working conditions, and if both SE and Fe spectra are taken at the same gas gain. Gas gain ranged from 10^3 to 2×10^4.

The measured quantity given by Equation 2 is found to be dependent on the gas gain (Figure 3). Because of its dependence on the measuring conditions this quantity cannot represent a real W value of the gas. Therefore, we call the quantity defined in Equation 2 an 'apparent W value'. The initial decrease of the apparent W value at low gas gains is more pronounced in mixtures with lower isobutane partial pressure, and is attributed to the secondary effects (avalanche chain formation). A relatively constant value is measured for the gas gain range 2000–8000. For the same concentration fraction of isobutane, this value is lower in mixtures at lower total pressure. At higher gas gains the apparent W value
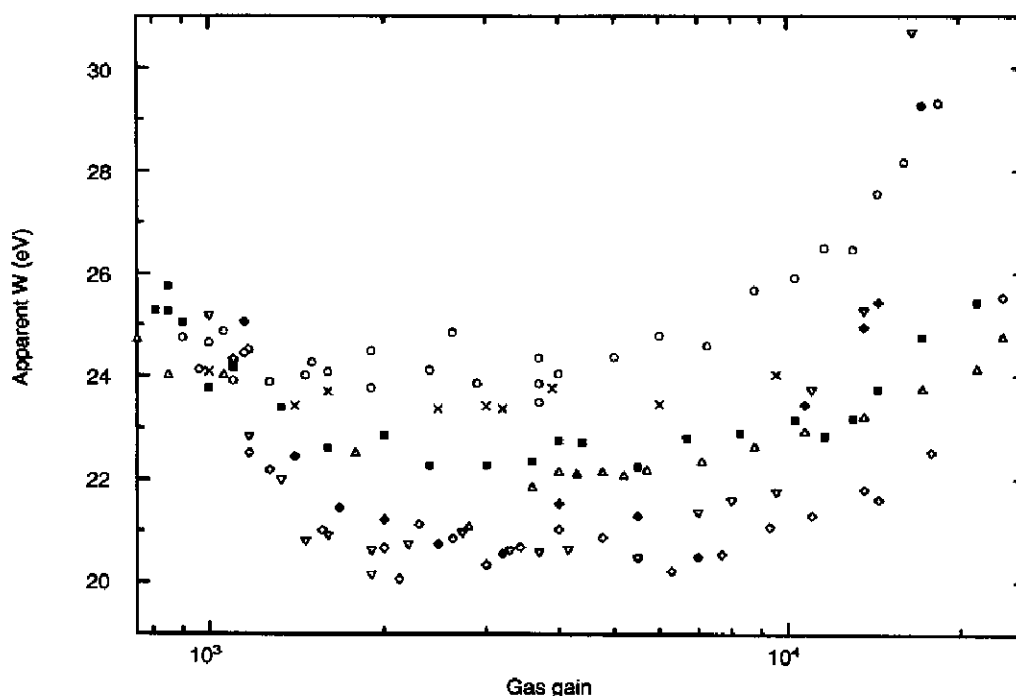

Figure 3. The apparent W value measured by a proportional counter in isobutane and in several argon–isobutane mixtures as a function of the gas gain: (×) pure isobutane, p = 30 kPa; (○) 30% isobutane, p = 90 kPa; (△) 30%, 30 kPa; (◇) 30%, 10 kPa; (■) 10%, 90 kPa; (◆) 10%, 10 kPa; (▽) 3.3%, 30 kPa.

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increases in all mixtures, as well as in pure isobutane due to the space charge effect. We can conclude that the proportional counter method for W value measurement is not applicable to the Penning gas mixtures because of the secondary effects in the counter caused by insufficient quenching.

Energy resolution

Energy resolution of the counter at 5.9 keV was measured as the ratio of the full width at half maximum (FWHM) and the position of the Fe peak. Energy resolution is a function of gas gain (Figure 4). The best values are obtained at very low gas gains, from $M \approx 30$ to $M \approx 200$, while at gas gains usually applied in low
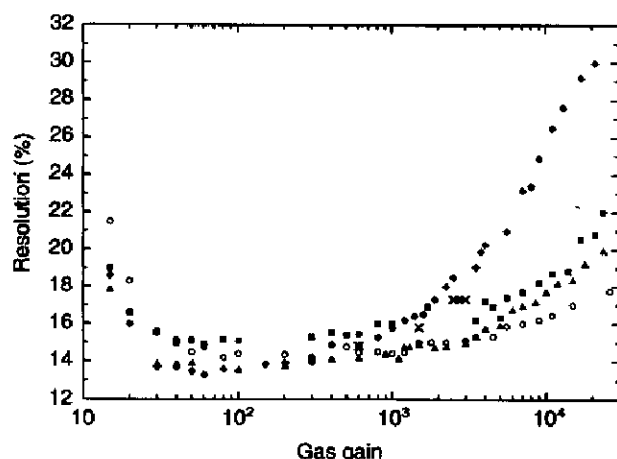

Figure 4. Energy resolution at 5.9 keV as a function of gas gain in a proportional counter filled with isobutane and argon–isobutane mixtures at total pressure of 30 kPa: (×) pure isobutane, (■) 30% isobutane, (○) 20%, (△) 10%, (♦) 3.3%.

energy X ray spectroscopy (several thousands) it is degraded. Mixtures with a lower content of isobutane give better energy resolution in the low gas gain region, but the gas gain range with the optimal resolution is narrow and the degradation of energy resolution is faster, which is caused by faster increase of the f values in such mixtures.

The measured energy resolution is a combination of the intrinsic resolution proportional to W(F+f), and the instrumental resolution[8]. Any attempt at derivation of the Fano factor F from the measured energy resolution is meaningless unless the exact instrumental contribution to R is known.

CONCLUSION

Although very promising, the Penning mixtures should not be used in a proportional counter at high gas gains and low gas pressures due to secondary effects arising from insufficient quenching. Avalanche chain formation in mixtures with low isobutane content influences primarily the statistics of gas amplification and thus it becomes the dominating factor in the energy resolution degradation at high gas gains. The present results contribute to the knowledge about new Penning gas mixtures and can help to improve performances of gas-filled detectors by choosing the gas mixture advantageous for a specific application.

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