GAS PROPORTIONAL SCINTILLATION COUNTER WITH XENON AND XENON MIXTURES

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Summary

The light output of the recently developed gas proportional scintillation counter - a gas scintillation counter with light multiplication produced by a cylindrical geometry electric field - is investigated for heavy gaseous media under alpha particle excitation with a 56 UVP phototube. The gases used are Xe, and Xe-Ar and Xe-N_2 mixtures for a wide range of concentrations, at a total pressure of about 965 Torr.

The light output of the Xe-Ar mixtures shifts towards the ultraviolet ($\lambda \lesssim$ 3250 Å) region as the electric field intensity increases. Xe-Ar mixtures with Xe concentrations ranging from about 1 to 10% give - with a p-quaterphenyl wavelength shifter - a light output more than two orders of magnitude larger than that of a CsI(Tl) scintillator.

In Xe-N_2 mixtures - with wavelength shifter - nitrogen has a quenching effect.

For the mixtures of noble gases the rise time of the secondary component of the light pulse is slow, typically 15 μs. Methane and nitrogen while having a quenching effect, reduce this rise time.

Introduction

Gas scintillation counters, specially those using noble gases, are well known for their good time resolution and versatility of the detection media but, in general, the energy resolution is poor[1-3].

Their behaviour when an uniform electric field is applied, has also been investigated[4-6] and it was observed that, following the fast primary scintillation it appears another and much slower component which can be used for energy resolution measurements. The uniform electric field geometry, although convenient to study the mechanism of the process, gives rise to different pulse heights depending on the orientation and location of the charged particle track. Therefore the energy resolution is relatively poor.

The use of a cylindrical geometry electric field has lead to the development of the gas proportional scintillation counter - PS counter[7-12]. Such a counter by confining the region of light multiplication to the immediate vicinity of the central anode, can give pulses with amplitude independent of the position of the initial track. Their amplitude rises with the electric field. These pulses - secondary scintillation pulses - have a rise time dependent on the electron migration time which is slow. When the electric field is strong enough there might appear tertiary and higher order components[10]. These are the consequence of avalanches resulting from photoelectrons ejected from the walls of the counter by ultraviolet light produced in the scintillation processes. We call light multiplication pulse the total scintillation pulse minus the contribution of the primary scintillation, i.e. the pulse arising from those components due to the electric field. Its rise time being slow cannot be used for fast coincidence work but, as the primary scintillation is not affected, there is still the possibility of obtaining the fast time resolution characteristic of gas scintillators with no electric field.

To the increase of the light output, which can be quite large, corresponds an improvement in the energy resolution, which is limited by the appearance of the higher order components[8-10]. The experimental energy resolution, for the argon-nitrogen mixture that gives the largest light output seems to be limited by the amount of light reaching the phototube, provided that those higher order components are avoided. The energy resolutions already obtained are comparable with those of a proportional ionization counter[8,9].

The performance of PS counters is surpassed by semiconductor detectors, but for special applications, those for which gas scintillation counters and proportional ionization counters are still used, the PS counter might be advantageous even at the present stage of development.

As little work has bee done with PS counters using xenon - an useful medium for X and gamma ray detection - we carried out this investigation aimed at obtaining data on the performance of pure xenon counters and of xenon counters with variable amounts of argon under alpha particle excitation. Xenon mixtures are particularly interesting due to their large light output[12]. Small amouts of nitrogen and methane were added in order to reduce the pulse rise time.

Experimental techniques

The gases used (commercially available) are: Xenon of purity 99.8% (impurities: Kr 2000 v.p.m., N_2 20 v.p.m., H_2O 10 v.p.m., O_2 10 v.p.m., $CO+CO_2$ 10 v.p.m., CH_4 5 v.p.m.), Argon of purity 99.99% (humidity < 10 mg/m^3), Nitrogen 99.9% ($O_2 \leq 5$ p.p.m., $H_2O \leq 5$ p.p.m. - "dew point" < -72 °C, argon≤1000 p.p.m.), Methane 99.98% (N_2 0.02%).

The noble gases were continuously purified with calcium turnings at 370 °C, in the system described before[10]. When working with nitrogen or carbon dioxide mixtures the temperature of the purifier was reduced to 250 °C after a previous purification at 370 °C of the noble gases. It was verified that when working with methane at these temperatures there was a slow consumption of this gas. So mixtures with methane were made with continuous circulation and the purifier temperature reduced to 125°C. For all mixtures the total working pressure was always 965 Torr.

The geometry of the PS counter is the one of a proportional counter, apart from the system of light collection and detection. The light is observed with a 56 UVP photomultiplier tube through a quartz window 1 cm thick. Detailed diagrams of the counters I and II used in this work are found in refs. 10 and 11 respectively, counter I being longer (17cm) than counter II (7cm). Counter I provides a much better electric field geometry than counter II, but has the disadvantage of a smaller light collection efficiency. The diameter of the anode nickel wire used is 0.005" for counter I and 0.010" for counter II. Both counters have side aperture 2mm diameter through which alpha particles are injected. Counter I aperture has a 0.001" thick Mylar window while counter II has no window. The sources used were ThC+ThC' and Am-241. The inside surface of the quartz window is covered with p-quaterphenyl (≤ 100 μg/cm^2) and the walls of counter I (stainless steel) with a layer of the same wavelength shifter (≥ 100 μg/cm^2). The walls of counter II (brass) have no deposit.

The photomultiplier anode resistance was 4.7 MΩ ; this allows a detailed observation of the pulse structure. The pulses were studied with a 585 Tektronix oscilloscope. Energy resolution measurements were made feeding the photomultiplier pulses through a White cathode follower into a linear amplifier (integration time 5 μs, differentiation time off) and analysed with a single channel analyser (HP 5583A).

Experimental results

Pure Xenon PS counter

Pulse shapes for pure xenon under Am-241 alpha particle excitation in counter II with no p-quaterphenyl wavelength shifter, are shown in fig. 1. Fig. 1 a, for no electric field, shows only the primary scintillation fast pulse. Fig. 1 b for 300 Volts in the central anode shows the secondary scintillation pulse which follows that fast pulse after a time lag of 12 μs, its rise time being about 10 μs. The time lag between the primary and secondary scintillation is well defined on account of the short range of alpha particles in xenon. Otherwise only for alpha particles parallel to the central anode can the time lag be well defined[10]. When the anode voltage rises there is a decrease of the time lag and of the rise time of the secondary component and an increase of its amplitude. Fig. 2 shows the variation with anode voltage of the light multiplication pulse amplitude. The plotted amplitude is the total amplitude minus the contribution of the primary scintillation, rather than the total scintillation pulse[9-12]. This correction is made in order to enhance the influence of the anode voltage on the light multiplication process.

By interposing filters with cut-off wavelengths of 3250 and 3800 Å [10] the pulse amplitudes V_1 and V_2 corresponding to the light that passed through the respective filter were compared to V_o, the pulse amplitude with no filters. V_o corresponds to wavelengths above the sensitivity limit (1800 Å) of the phototube. The ratios V_1/V_o and V_2/V_o suitably corrected for the transmittance of the filters and the contribution of the primary scintillation, are plotted, as r_c(%), in fig. 2, as a function of the anode voltage. The r_c for $\lambda > 3250$ Å and for $\lambda > 3800$ Å are approximately the same. This means that, for any anode voltage, the contribution of the light within the 3250-3800 Å range is small. For less than about 0.5 kV, r_c decrea-

ses sharply but it remains approximately constant for more than 1 kV. This implies a pronounced shift towards shorter wavelengths, so that from 1 kV upwards only about 10% of the pulse height is due to light with $\lambda > 3250$ Å. This shifting is attributed to the excitation of higher energy levels by more energetic electrons.

In view of this results it is to be expected that, as far as the pulse height is concerned, a wavelength shifter is advisable, even when a quartz window phototube is used. A layer of p-quaterphenyl ($\leq 100\ \mu g/cm^2$) was vacuum deposited on the inside surface of the quartz window of the counter. Fig. 2 shows also the ratio, q, of the light multiplication pulse height with and without p-quaterphenyl as a function of the anode voltage. In the region where there is a fast decrease of r_c, q increases rapidly, remaining practically constant for larger anode voltages. Here the effect of the wavelength shifter is to increase the pulse height by a factor 8.

Xenon-Argon PS counter

In order to study the behaviour of xenon-argon PS counters, several xenon-argon mixtures were prepared, continuously purified and circulated through counter II.

The light multiplication pulse amplitude(corrected for the energy loss of the Am-241 alpha particles in the gas channel corresponding to the thickness of the counter wall) is plotted in fig. 3, as a function of the central anode voltage, for various xenon--argon mixtures. Two sets of curves are shown: one for data with the inside surface of the quartz window covered with a p-quaterphenyl layer ($\leq 100\ \mu g/cm^2$) and another for data without such a layer. The effect of the wavelength shifter is to increase the pulse amplitude by about an order of magnitude. Besides the data shown in fig. 3, measurements were also made for the following xenon percentages: 99.2, 69.7, 23.1, 9.9, 2.5, 2.3, 0.25 and 0.05, without p-quaterphenyl, and 9.7 and 2.5, with p-quaterphenyl. These curves are not plotted for the sake of clearness. For each curve the maximum voltage used was limited either by a sudden and strong distortion of the pulse shape or by sparking.

The general behaviour of the curves is the same whether there is a wavelength shifter or not: a fast increase of the secondary pulse amplitude for voltages up to about 0.4 kV followed by an increase at a slower rate. For xenon concentrations of a few percent the rate of increase rises again for voltages over about 1.5 kV. Two mixtures with a small concentration of one of the components, Ar-0.05% Xe and Ar-99.2% Xe, give the same light output than the pure gas, i.e. argon and xenon respectively. But while mixtures very rich in argon like Ar-43.8% Xe, give about the same light output as does pure xenon, mixtures like Ar-0.51% Xe, that have a small content of xenon, give a light output much larger than that from pure argon (fig.3).

The variation of the light output with the concentration is voltage dependent as can be seen in fig. 4 where a family of curves for 1.0, 1.4 and 1.8 kV is plotted. The light output is practically constant for xenon concentrations larger than about 40%. It rises sharply from 0.1 to a few percent. For a xenon concentration of around 3% the light output, at relatively high voltages, reaches a maximum that is a sharper for 1.8 kV than for 1.4 kV, specially when the wavelength shifter is used(fig. 4). As it has already been pointed out[12] it is for argon-xenon mixtures in this region of concentrations that a gas proportional scintillation counter gives one of the largest light yields.

Fig. 5 shows the ratio, r_c (%), for $\lambda > 3250$ Å for three anode voltages as a function of the xenon concentration. As it should be expected, at any concentration there is a decrease of r_c when the voltage increases, the curve for 1.8 kV lying below the one for 1.0 kV and this one below the curve for 0.5 kV; this corresponds to a shifting towards shorter wavelengths. The decrease of r_c for xenon concentrations from about 0.1 to a few percent implies a relative increase of the light output from 1800 to 3250 Å. Further increase in the xenon content does not change appreciaby the proportion of light within this range. The comparison of figs. 4 and 5 shows that the ranges of concentrations for which these features occur are the same as those for which the pulse amplitude first rises sharply and thereafter remains constant for not very high anode voltages.

Xenon-Nitrogen PS counter

Counter I was used as before with its window and walls covered with p-quaterphenyl in order to study the behaviour of xenon--nitrogen mixtures. The light multiplication pulse amplitude versus anode voltage is plotted in fig. 6. For large nitrogen concentrations (dashed curves for 16.5 and 33.8% N_2) the plotted pulse amplitude includes the primary scintillation because this was too small and of the order of the noise. Whatever the anode voltage nitrogen has a quenching effect as it had for the argon-nitrogen counter des-

cribed before[11]. Nevertheless the nitrogen quenching effect is less accentuaded for xenon than it was for argon. Fig. 7 shows the effect of nitrogen on the amplitude and the rise time of the light multiplication pulse for 2.2 and 1.4 kV. Nitrogen concentrations of about 1% have a practical interest since they reduce the rise time from about 18 to 7 μs, while reducing the amplitude only ~ 30%. Larger concentrations do not change appreciably the rise time while decreasing drastically the pulse amplitude. The observed decrease in rise time is to be expected on the grounds that, as it is well known, the drift velocity of electrons in noble gases is reduced when impurities like nitrogen and methane are added. This suggests the study of the effect of these impurities in a mixture giving a larger light output like the Ar-5% Xe one.

Fig. 8 shows the rise time and the light multiplication pulse amplitude versus nitrogen or methane content in this mixture. The quenching effect of nitrogen is stronger for this mixture than it was for pure xenon and the rise times are about the same. Methane while reducing the rise time more than nitrogen has a very strong quenching effect. For example, for 2% of methane the rise time is only 1.5 μs, but the pulse amplitude has decreased already by three orders of magnitude.

A similar behaviour was observed when methane was added to pure xenon.

Practical light output and energy resolutions

The arbitrary units scale used in this work for pulse amplitudes is the same (within 10%) as for the previous works[11,12]. In this scale the primary scintillation pulse amplitude for pure xenon was found to be 2.8 units in counter I and 6.4 units in counter II (counters with wavelength shifter and the anode at zero Volts). The different outputs of the two counters result from different light collection efficiencies.

Pure xenon in counter I at 1.4 kV gives an amplitude of 400 units, a gain of about 140 over the primary scintillation; the same gain is observed in counter II. This is about 80 times the pulse amplitude from a CsI(Tl) scintillator[12]. At 1.8 kV this last figure rises to 120 but it was difficult to see whether tertiary components were present. From our observations we estimate that the practical light output - defined as the output to which corresponds the largest pulse obtained with no tertiary components[12] - is, for pure xenon, about two orders of magnitude larger than the CsI(Tl) light output.

Xenon-argon mixtures can give larger pulse amplitudes, over a thousand units, specially on the region 3 to 5% of xenon content. The practical light output of an Ar-5% Xe mixture (counter I) is reached for 1.46 kV to which corresponds an amplitude of 1800 units i.e. over 300 times the one for CsI(Tl).

To practical light outputs as large as these should correspond a substantial improvement in the energy resolution. The best resolution obtained (counter I) was 3.3% for an Ar-5% Xe mixture at 1.36 kV and E_{α} = 6.7 MeV; for pure xenon and Xe-1% N_2 mixture the energy resolutions were only 5 or 6%. These figures are not corrected for the straggling in the Mylar window.

Due to the low light collection efficiency of the counter, the amount of light that reaches the phototube is only a fraction of the total light. This fact and the possible instabilities in the multiplication processes may explain the relatively poor resolutions obtained.

Conclusions

Gas proportional scintillation counters using xenon and xenon-argon mixtures are found to give, with a wavelength shifter, light outputs that may be more than two orders of magnitude larger than the ones from standard inorganic scintillators.

The maximum light output for xenon-argon mixtures is obtained for a xenon concentration of 3-5%. It is interesting to point out that this is the range of concentrations for which Golubnichii and Yakolev[13] while investigating xenon-argon mixtures for spark chambers found a marked minimum for the so-called breakdown potential. The variation of the breakdown potential with xenon concentration seems to be related with the variation we found for the light multiplication pulse amplitude.

The spectral distribution of the secondary scintillation light is found to depend on the anode voltage. For xenon this is in apparent disagreement with the results of Teyssier et al.[14] which did not observe such a dependence for uniform electric fields. However, in PS counters the electric field at the anode surface is typically 20 kV/cm which is much stronger than the fields used by those authors. Any future attempt to interpret the mechanism of a PS counter has to take into account the scintillation spectra for such high fields. At present there is no data available on this subject.

The slow pulse rise time of PS coun-

ters with pure noble gases and their mixtures can be reduced by the addition of small amounts of nitrogen and methane but this is achieved at the expense of a large reduction in the pulse amplitude. Nevertheless pulse rise times of 7 to 5 μs are attainable with no drastic reduction in the pulse amplitude if 1 or 2% of nitrogen is added to pure xenon. These mixtures might have practical interest for X and gamma ray detection.

The energy resolutions obtained are comparable with those from standard scintillators. However, we expect this characteristic to be improved when a more efficient light collection system is devised and when the problem of the suspected instabilities of the light multiplication process is dealt with. Work in progress at this Laboratory is being carried out along these lines.

Thanks are due to Prof. J.R. de Almeida Santos for his interest in this work. Financial support given by Comissão de Estudos de Energia Nuclear do Instituto de Alta Cultura, Portugal, as well as funds supplied by the Calouste Gulbenkian Foundation, Lisbon, and by the NATO Scientific Committee are gratefully acknowledged.

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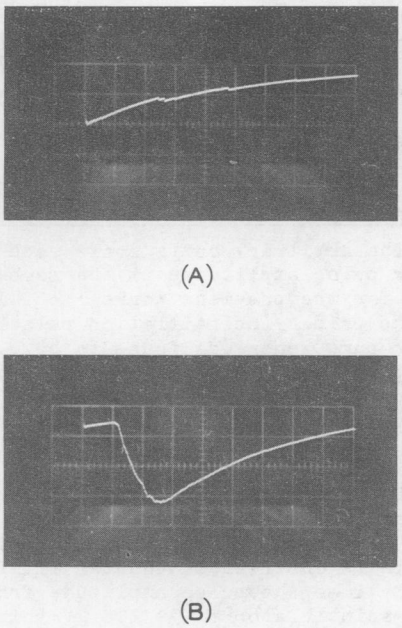

Fig. 1. Pulse shape for Am-241 alpha particles in xenon at 965 Torr (counter II without wavelength shifter): (a) no electric field (hor.: 10 μs/div.). (b) 300 V anode voltage (hor.: 10 μs/div.).

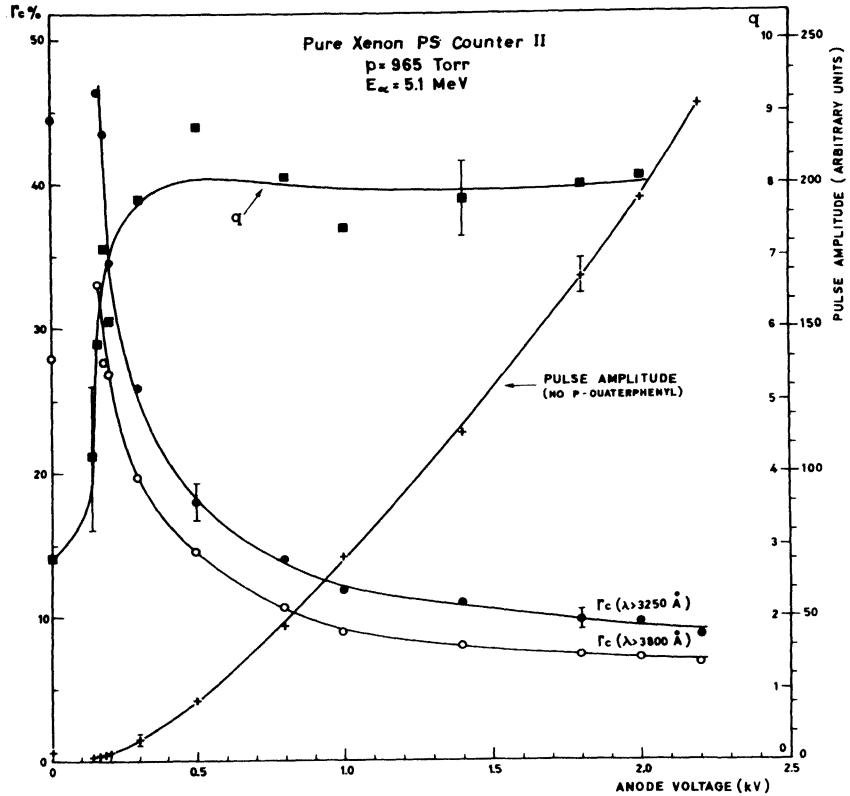

Fig. 2. Characteristics of the pure xenon proportional scintillation counter as a function of the anode voltage (counter II). The errors shown are estimated.
crosses: light multiplication pulse amplitude (no p-quaterphenyl).
full circles: ratio, r_c, (see text) of the amplitudes with and without filter for $\lambda >$ 3250 Å.
open circles: ratio, r_c, for $\lambda >$ 3800 Å.
full squares: wavelength shifter efficiency parameter, q, (see text).

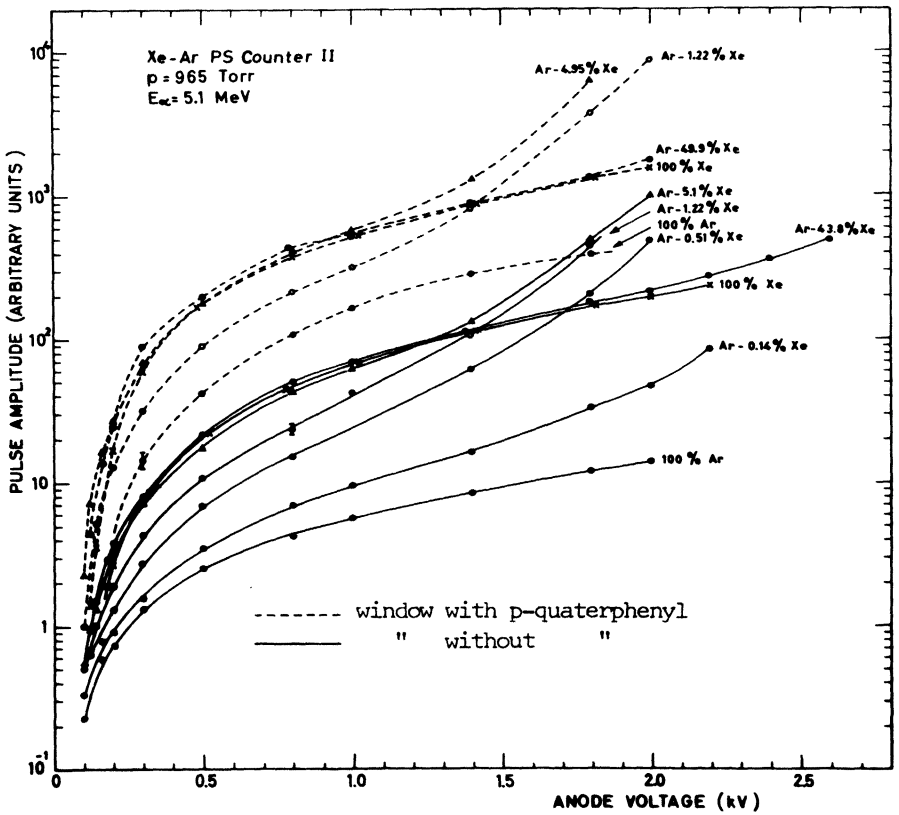

Fig. 3. Light multiplication pulse amplitude versus anode voltage for various xenon-argon mixtures (counter II with and without p-quaterphenyl). The errors shown are estimated.

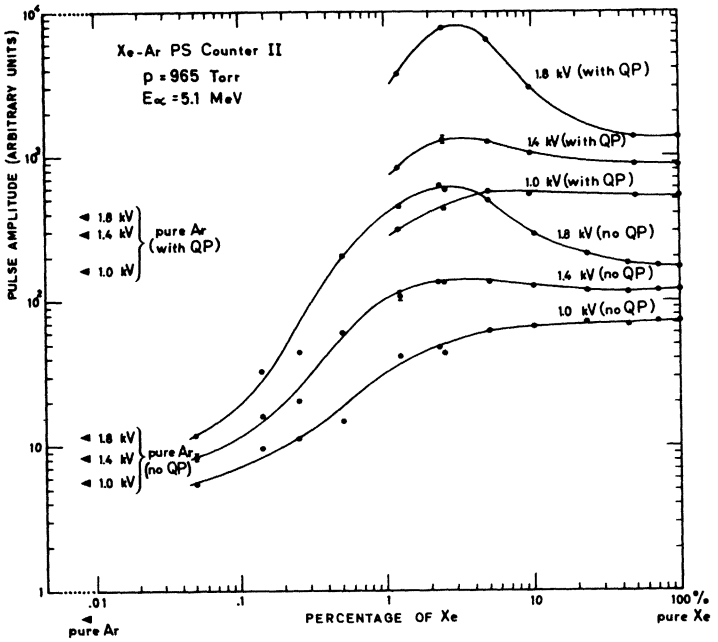

Fig. 4. Light multiplication pulse amplitude versus xenon concentration, for various anode voltages (counter II with and without p-quaterphenyl). The errors shown are estimated.

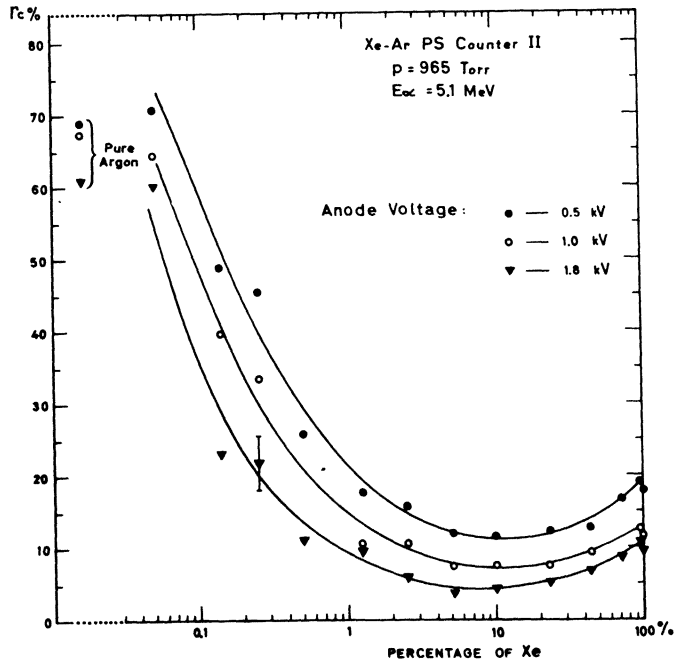

Fig. 5. Ratios, r_c, (see text) of the amplitudes with and without filter ($\lambda > 3250$ Å) versus xenon concentration in argon, for 0.5, 1.0 and 1.8 kV (counter II with no p-quaterphenyl). The errors shown are estimated.

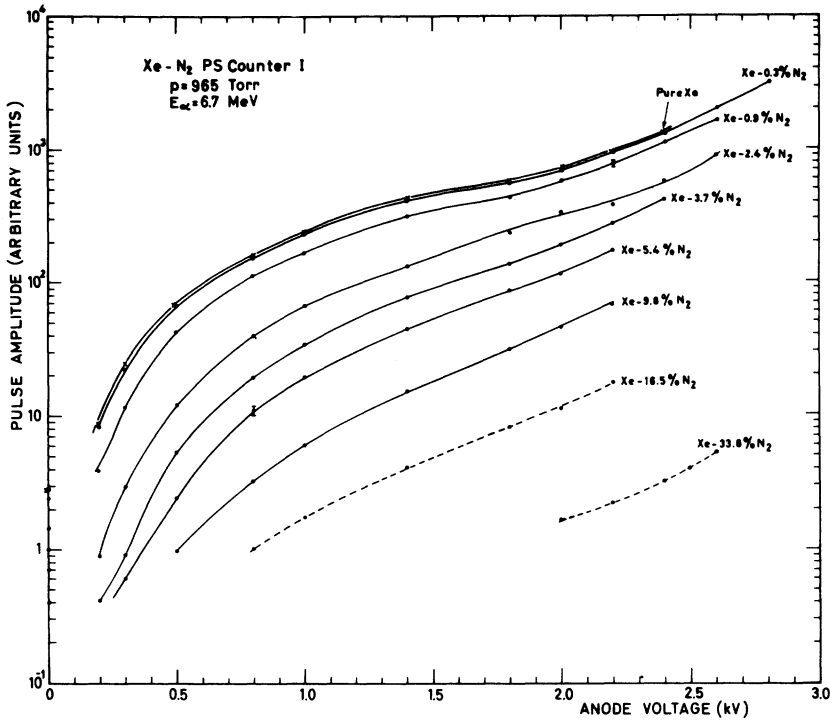

Fig. 6. Light multiplication pulse amplitude versus anode voltage for various xenon-nitrogen mixtures (p-quaterphenyl in counter I window and walls). Dashed curves are not corrected for the primary scintillation. The errors shown are estimated.

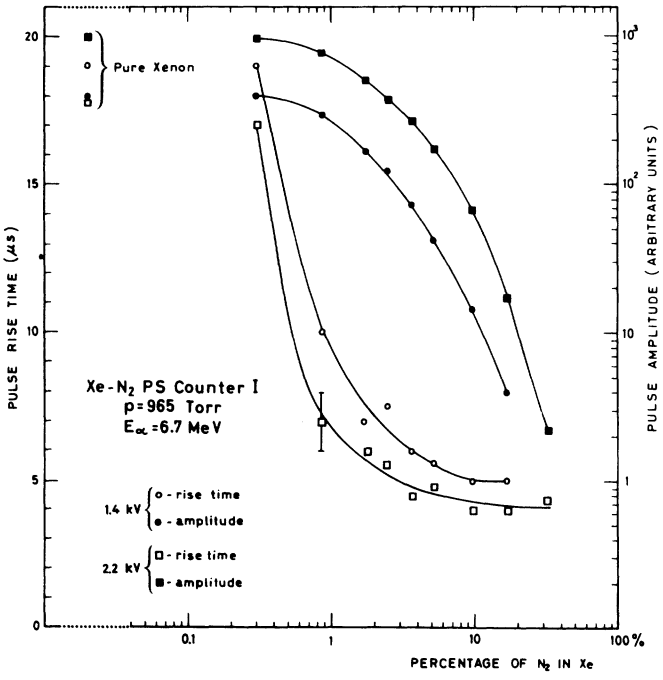

Fig. 7. Rise time and amplitude of the light multiplication pulse, versus nitrogen content in xenon for 1.4 and 2.2 kV anode voltage (counter I with p-quaterphenyl). The errors shown are estimated.

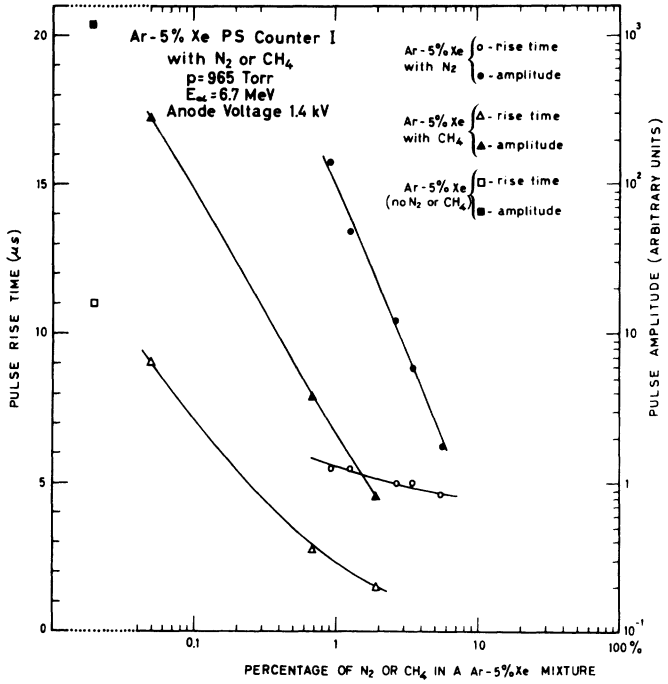

Fig. 8. Rise time and amplitude of the light multiplication pulse, verus nitrogen or methane content in an Ar-5% Xe mixture for 1.4 kV anode voltage (counter I with p-quaterphenyl).