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SELF-QUENCHING STREAMERS IN MIXTURES WITH PHOTOIONIZATION VAPOURS

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Abstract

The basic properties of self-quenching streamers in a proportional counter with a thick anode wire are investigated. In particular, the influence of mixing benzene and triethylamine to argon-isobutane mixtures is studied regarding thresholds for streamer production, pulse shapes and dead time. There is a lowering of the high voltage threshold for streamer production associated with low concentrations of triethylamine and benzene. Higher concentrations of benzene increase that threshold. Triethylamine leads to narrower pulses. Stable operation is observed for a much broader range of high voltages than in the absence of photoionization vapours.

1. Introduction

On the last few years some remarkable properties associated with the self-quenching streamer (SQS) mode (or limited streamer mode) have been studied¹⁻⁹, following the first observation of unconventional gas multiplication phenomena under certain conditions¹⁰⁻¹². The SQS mode in which narrow streamers, a few hundred μm wide are formed perpendicularly to the anode wires for steady high voltages, is characterized by large wire signals ($\geq 10 \text{ mV}/50 \Omega$), at least one order of magnitude larger than those of the proportional mode, relatively narrow amplitude distribution and short duration of a few tens of nanoseconds. The mode is stable, i.e., there is a relatively wide range of high voltage with a counting rate plateau. This, together with the localization capability and the large amplitude of the fast signals, which greatly simplifies the electronics, are attractive features of this regime.

For some HV values proportional mode pulses and SQS are observed simultaneously; around this transition voltage no intermediate pulse heights are observed between those corresponding either to the proportional or the streamer state modes.

The transition from the proportional to the SQS mode seems to occur at some definite value of the charge in the primary avalanche^{1,4} and the most probable mechanism associated with the SQS relies on the decisive role of the photoionization processes in the formation of secondary avalanches and on the influence of the space charge of the primary avalanche. According to Atac¹, during the process of the primary avalanche development, when space charge saturation becomes important, for example for fillings with argon the radiative recombination of Ar^+ and cooled e^- originates the emission of energetic photons capable of ionizing molecules present in the medium. These photoelectrons can originate secondary avalanches on the high field region on the tip of previous avalanches and form the streamer, provided that the electric field of the primary avalanche is of the order of magnitude of the external electric field. There seems to be some evidence¹ that the electrons produced by recombination photons, which mediate the SQS, result from ionizing the organic additive and not argon atoms. The mean free path of the photons is a very important parameter, and it must be

similar to the dimensions of the avalanche, such that the photoionization can take place in the immediate vicinity of the primary avalanche. Of course, to what extent this condition is satisfied depends on the composition of the gas mixture and pressure.

Argon with organic additives as quenchers have been largely studied, as these gases are commonly used in proportional chambers. Some detailed studies on argon-ethane-ethyl alcohol mixtures^{1,2}, argon-methane-methylal⁴⁻⁷ and argon-isobutane⁵ have been performed. The general features of the SQS are the same for all these mixtures. Even in conditions of large charge multiplication, mixtures of argon with small concentrations of benzene or triethylamine or acetone seem to feature photon spectra in which there is some contribution of the argon continua¹³. To our knowledge no spectra of the photons associated to the SQS mode are available and the noble gas continua may still be present. And, as this streamer process is decisively mediated by photoionization, in the present work the argon-isobutane gas mixtures are retaken and the effect of additives with photoionization efficiencies and photo-absorption cross-sections reasonably known around the region of the argon continua emissions, namely benzene and triethylamine (TEA), is investigated.

2. Experimental System

The detector used in this work is a proportional counter with a stainless steel cathode 17 cm long and 4 cm diameter, and a nichrome wire anode 60 μm thick, stretched along its axis. The entrance window for the bombarding radiation, X rays from a ^{55}Fe source, is a 10 mm diameter 0.001" thick beryllium foil. The chamber is operated at atmospheric pressure in a continuous flow regime. The composition of the gas filling is achieved by mixing pure argon (purified with a BOC purifier) with isobutane and argon saturated with the photoionizing vapour, either benzene or TEA. The concentrations quoted in this paper were not corrected for differences of pressure and viscosity and correspond to relative volumes as obtained from the readings in the flowmeters and vapour pressures of the liquids.

The measurements of the charge characteristics of the chamber were made using a conventional charge amplifier electronic system and multichannel analyzer.

The study of the anode current pulses, through a 50 Ω resistance, was made using an oscilloscope to observe the output of a fast timing filter amplifier fed with those signals.

The counting characteristics of the chamber were observed with the electronic set up used for the current amplitudes plus a discriminator to reject the proportional pulses or noise.

3. Experimental Results

Argon-isobutane in 2:1 proportion (ratio of direct flowmeter readings) is used throughout this work; this proportion is kept even when TEA or benzene are used. The concentrations of TEA were 1.1% and 4.5% and

of benzene 1.5%, 3.4% and 7%. The effect of the isobutane was studied only through the influence of the high voltage on the current and the charge amplitudes for argon-isobutane mixtures in proportions 100/40 (28.5%), 100/50 (33.3%) and 100/60 (37.5%).

3.1 Amplitude spectra

Figs. 1,2,3 and 4 show the average collected charge and current pulse amplitudes from the chamber as a function of the high voltage starting from the proportional region, for argon-isobutane with several concentrations of TEA or benzene. The discontinuous transition from saturated proportional to the SQS mode is observed in all cases. In a few cases, for higher voltages, a second amplitude leap was observed, though it is much smaller than the first one. This is represented in the figures showing the current behaviour; in the figures displaying charges, just an average value of smaller and higher amplitude streamers, anyway not too different, is shown. Again, to make the figures simpler, in those referring to current no data are represented without the photoionization vapour, as the general trend is identical to the data concerning charge.

A few features seem to be apparent. Space charge saturation effects, on the region of $\sim 4pC$, are associated with the transition to the SQS mode, either for argon and isobutane in 2/1 ratio or for this mixture with benzene and TEA.

From Figs. 1 and 2 where the effect of the addition of TEA and benzene on the 2/1 argon-isobutane mixture is displayed, it is clear that on the proportional mode TEA 4.5% does not seem to influence the mean charge pulse amplitudes, while 1.1% increases their amplitude relative to argon-isobutane. Benzene 1.5% behaves very much like TEA 1.1%, but as the % of benzene increases the mean charge pulse amplitude decreases relatively to argon-isobutane alone.

On the SQS mode the same general pattern as in the proportional region is followed by all the mixtures with benzene and by the 1.1% TEA mixture, but the 4.5% TEA mixture in the SQS mode decreases significantly the mean charge pulse amplitude relative to argon-isobutane (the opposite happened in the proportional region), and has an effect very similar to that of 7% benzene.

As far as the threshold voltages for streamer production are concerned, there is a lowering with the addition of either benzene or triethylamine in very small amounts (1.1% TEA and 1.5% benzene). However, for the higher concentrations either of TEA or benzene that threshold increases relatively to the 2/1 mixture of argon-isobutane. Data from Figs. 3 and 4 concerning the current pulse, yield similar conclusions.

To have some possibility of comparing the effect of benzene and TEA as additives, relative to the pure isobutane quencher, the study of the argon-isobutane mixtures 28.5%, 33.3% and 37.5% on the current mode was made (see Fig. 5), pulse amplitudes being studied as a function of the high voltage. Increasing the organic additive displaces the curves in both the proportional and the SQS modes: higher threshold voltages for streamer production and decreased pulse amplitudes are observed for higher isobutane percentage.

3.2 Pulse shapes

The current pulses for all fillings used are narrow, and their f.w.h.m. decrease slightly with the addition of the photoionizing vapours, being narrower for TEA than for benzene: $\sim 50ns$ for argon-isobutane, $\sim 45ns$ for argon-isobutane-benzene, and $\sim 35-40ns$ for Argon-isobutane-TEA.

3.3 Stability of operation

Although the addition of TEA and benzene, in the percentages studied, does not change very much the mean pulse amplitude, the pulse time duration or the threshold for SQS formation, it does alter very drastically the stability of the SQS mode.

Figs. 6 and 7 show the plateau of the counting characteristics for the various argon-isobutane-TEA and argon-isobutane-benzene mixtures used, expanding with the addition of the photoionizing vapour. The plateau increases from about 100 V for the 2/1 argon-isobutane mixture to more than 1400 V for benzene and more than 1000 V for TEA 4.5%. In fact, with this higher concentration of TEA and with benzene 7% the end of the plateau was not reached at the highest voltages used.

The initial rise of the curves in Figs. 6 and 7 corresponds to the transition between proportional and limited streamer pulse generation and at the beginning of the plateau such a transition is completed. The counting rate increase at higher voltages is due to the generation of afterpulses. These arise from the emission of secondary electrons from the cathode walls due to ultraviolet photon production in the primary discharge region in the case of the argon-isobutane mixture. The delay time between the primary pulses and the successive afterpulses was seen to be constant $\sim 500ns$, in agreement with previous observations of the same phenomena, as it corresponds to the electron drift time across the counter radius ⁵.

With 1.1% of TEA the afterpulses are not time localized and seem to correspond to photoionization processes in the gas filling. With higher concentrations of TEA the effect is qualitatively the same, although afterpulses are only observed for much higher high voltages. With benzene practically no afterpulses are observed, and again seem to be associated with photoionization of the gas.

3.4 Dead time

In this work the time structure of the dead zone ⁴ was qualitatively investigated and some preliminary data on its possible dependence on the high voltage and on the nature of the gas mixture were obtained. Our experimental procedure was similar to that used in ref. 4. Essentially the probability distribution of the time intervals between successive streamers is measured by sending the current signals through a short delay ($\sim 4\mu s$) to the start, and directly to the stop, of a time to amplitude converter unit; its output is seen by a multichannel analyser.

As can be easily shown these distributions are described, in the region where essentially there are no losses due to the dead time, by a decreasing exponential with a decay constant that is the counting rate. So the curves plotted in a semilog scale should fit a straight line for long enough times and the deviations from a straight line for smaller times will reflect the time behaviour of the dead zone. This behaviour was experimentally found in all our measurements the calculated decay constant being, within the experimental errors, in good agreement with the measured counting rate.

Fig. 8 displays some data referring to the argon-isobutane - 7% benzene mixture. For 4100 V, just on the beginning of the counting rate plateau, Fig. 8 a) shows clearly the effect of the dead time for smaller times, which is not visible in Fig. 8 b) for the higher voltage of 4300 V. This corresponds to a clear decrease of the dead time; its effect is nevertheless shown in Fig. 8 c), still for the same voltage, where data for a smaller time range are displayed. For higher voltages, see Figs. 8 d) and e), no strong variation of the dead time is observed, its main effect lasting for about 50 μs .

Fig. 9 displays the effect of the concentration of benzene in the dead time, for a constant voltage of 4300 V, that for all mixtures corresponds to the counting rate plateau. The higher concentrations of 3.4 and 7% show smaller dead time effects than the lower concentration of 1.5%.

4. Concluding Remarks

The charge values at the transition from proportional to SQS mode were estimated from the experimental results and are represented in table 1: Q_1 and Q_2 are the mean charges in the limited proportional and SQS mode, respectively, at the transition. Q_1 can be interpreted as a critical charge at which the transition from the Townsend's avalanche to streamer occurs; HV_{tr} is the transition high voltage, at which one half of the pulses belongs to the SQS mode and the second half to the proportional mode; this value was estimated from the single rates counting curves.

Table 1

Mixture Composition *	HV_{tr} (V)	Q_1 (pC)	Q_2 (pC)	Q_1 ($e^- \times 10^7$)	Q_2 ($e^- \times 10^7$)
Ar-1	3700	4.6	19.7	2.9	12.3
Ar-1-1.5% B	3650	5.9	23.6	3.7	14.8
Ar-1-3.4% B	3750	4.1	19.7	2.5	12.3
Ar-1-7% B	3850	4.1	22.4	2.5	14.0
Ar-1-1% TEA	3600	3.8	22.4	2.4	14.0
Ar-1-4.5% TEA	3650	3.8	15.8	2.4	9.9

* I = isobutane ; B = benzene

Within experimental errors Q_1 does not depend on the composition of the mixture and is $\sim 3 \times 10^7$ ion pairs. Also Q_2 does not seem to be much affected by the presence of the photoionizing vapour, except perhaps with 4.5% TEA, where the observed decrease may be significant. The results agree, in general, with those obtained in ref. 4 for Ar-methane-methylal mixtures, $\sim 2 \times 10^7$ ion pairs for Q_1 . A lower value, $\sim 8 \times 10^6$ ion pairs is indicated in ref. 1 for argon-ethane-ethyl alcohol. All these values approach the Raether's condition of 10^8 ion pairs.

The effect of the photoionizing vapour on the length of the counting rate plateau is clearly much more important than that of higher concentrations of isobutane in argon-isobutane mixtures⁵, and probably also more important than the effect of methylal in argon-methane fillings⁴.

Concerning the dead time characteristics, the data presented is being quantitatively analyzed. Nevertheless, qualitatively, the sharp decrease in the dead time when the high voltage increases near the transition region, and the fact that it remains approximately constant for higher voltages within the plateau, as observed in this work (Fig. 8), is in general agreement with the data of ref. 5 for pure argon-isobutane mixtures. Also, Fig. 9, there is a decrease of the dead time with increasing benzene concentrations, again in agreement with the effect of methylal in argon-methane mixtures⁴.

The results obtained are far from conclusive concerning the mechanism for streamer production. Clearly the effect both of benzene and TEA on the streamer high voltage threshold is very small and on the other hand they decisively affect afterpulse formation. One difficulty associated with the interpretation of these results is the lack of photoabsorption coefficients and quantum efficiencies for isobutane.

With argon SQS are observed with isobutane (IP=10.57 eV) and ethane (IP=11.5 eV). To photons able to photoionize these components correspond, for ethane,

a photoabsorption cross section of ~ 50 Mb. For the concentration of this additive normally used the mean free path for photons would be of the order of a few tens of μm . Probably isobutane will feature lower mean free paths, even on the region of the second argon continuum, and one can tentatively associate this or smaller dimensions with the mechanism for streamer development. Benzene and TEA on the region of the argon continuum where these additives feature larger quantum efficiencies, (benzene $\sim 60\%$, TEA $\geq 10\%$), and for the concentrations that we have used, feature mean free paths for photoabsorption at least five times larger. This being so if the argon continuum is present the small effect of these additives for the streamer formation could be explained by that much larger mean free path. Of course these considerations do not explain the very efficient suppression of afterpulses by both additives, specially by benzene.

Acknowledgments

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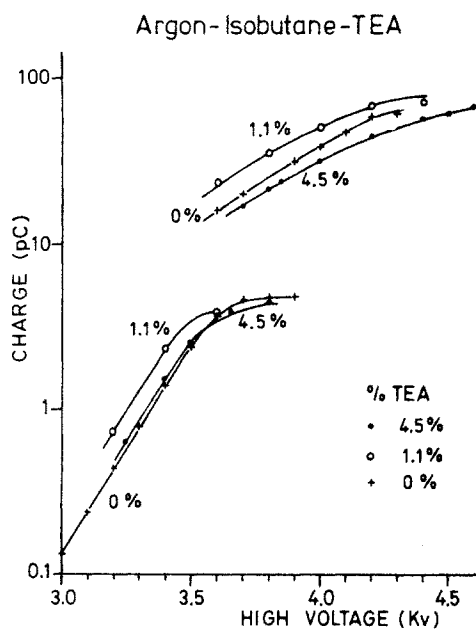


Fig. 1. A comparison of charge characteristics of the chamber for various argon-isobutane-TEA mixtures.

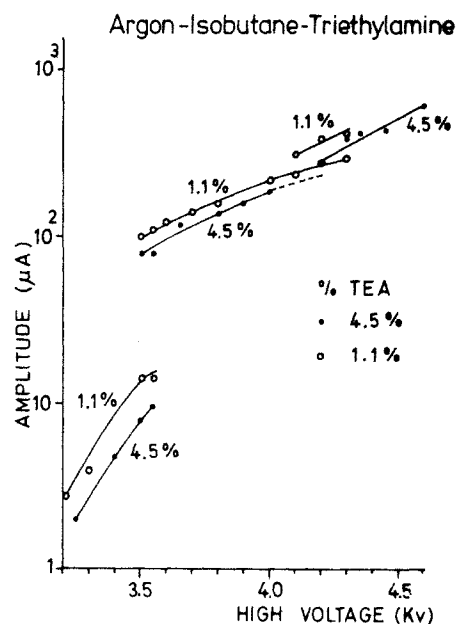


Fig. 3. The average amplitudes of the current pulses from the chamber for two different argon-isobutane-TEA mixtures.

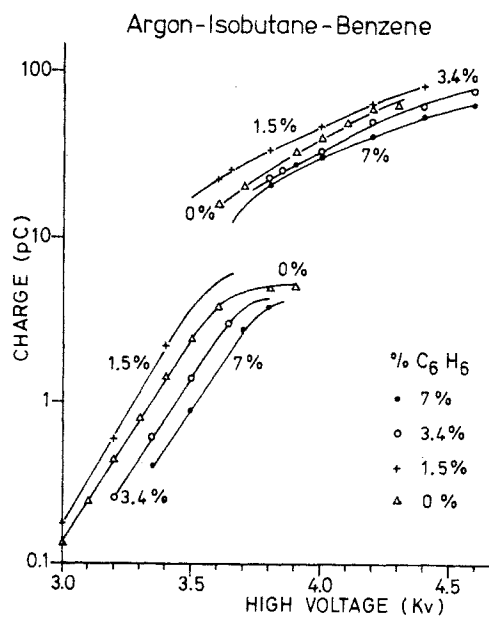


Fig. 2. A comparison of charge characteristics of the chamber for various argon-isobutane-benzene mixtures.

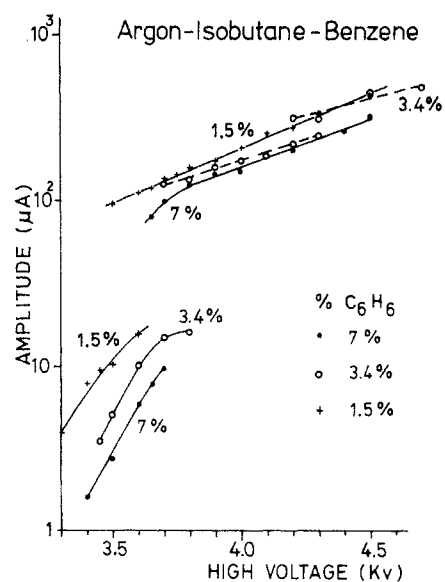


Fig. 4. The average amplitudes of the current pulses from the chamber for the different argon-isobutane-benzene mixtures used.

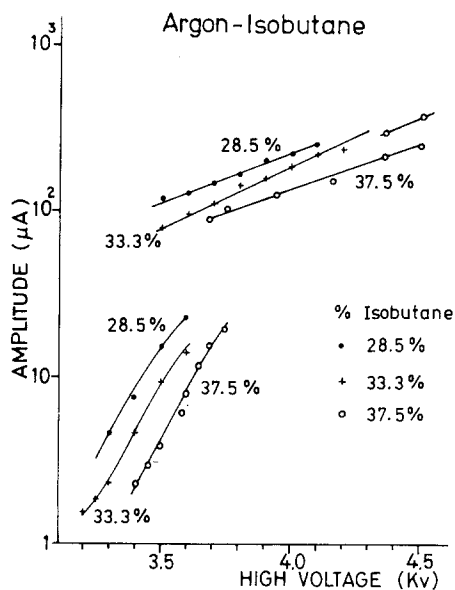


Fig. 5. The average amplitudes of the current pulses from the chamber for various concentrations of argon and isobutane.

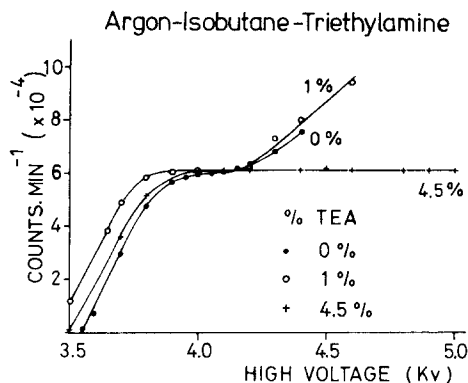


Fig. 6. Counting rate (⁵⁵Fe source) versus high voltage for various argon-isobutane-TEA mixtures.

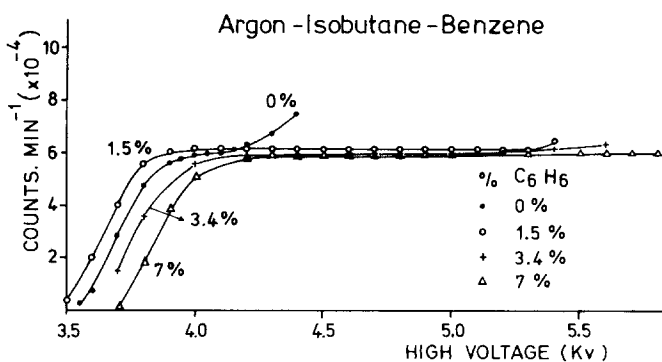


Fig. 7. Counting rate (⁵⁵Fe source) versus high voltage for various argon-isobutane-benzene mixtures.

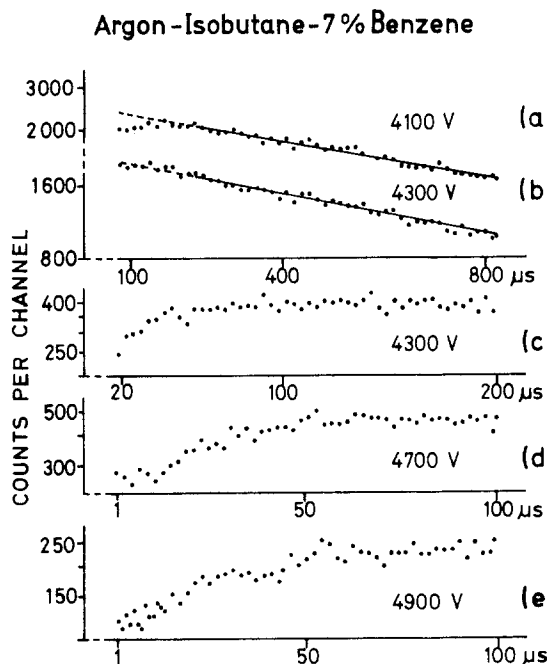


Fig. 8. Distributions of the time intervals between successive streamers. The time ranges of the time to amplitude converter unit are indicated. For 800 μs time range linear fits are displayed.

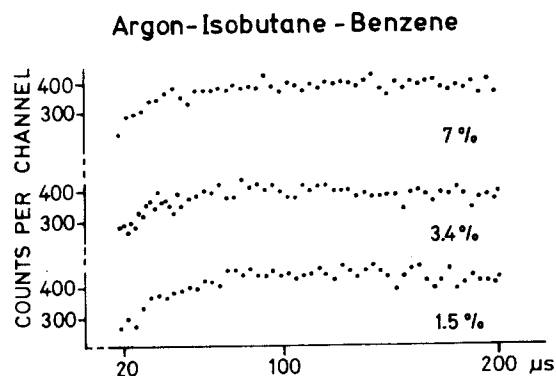


Fig. 9. Influence of the benzene concentration on the distribution of the time intervals between successive streamers for 4300 volts.