Electroluminescence studies on the influence of percentage quantities of CO_2 added to pure xenon

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ABSTRACT: In the present work, relative measurements have been performed for the EL yields of xenon and xenon-mixtures in a uniform electric field driftless GPSC. The operational parameters of the detector, including amplitude and energy resolution, were measured as a function of the reduced electric field in the scintillation region. For instance, for an E/p value of 1.5 V cm^{-1} $torr^{-1}$, the energy resolution has values about 8.5%, 9.6% and 18% for pure xenon, Xe+0.5%CH_4, and Xe+1%CH_4, respectively. Xe-CO_2 mixtures achieve energy resolutions of about 8.6% and 9.0% for CO_2 concentration of 0.11%, and 0.33%, respectively, at an E/p value of 1.5 V cm^{-1} $torr^{-1}$. For E/p values above 1.8 V cm^{-1} $torr^{-1}$, energy resolutions of around 8.4%, 8.4% and 8.7% can be obtained in Xe-0.5%CH_4, Xe-0.11%CO_2 and Xe-0.33%CO_2 mixtures, respectively. For Xe+1%CH_4 much higher electric field values would be needed in order to achieve such energy resolutions, while with Xe+2%CH_4 it is impossible to achieve good energy resolutions. Comparing with pure xenon, Xe+0.11%CO2 and Xe+0.5%CH4 do not present significant degradation of the electroluminescence yield and associated statistical fluctuations, being the use of such mixtures in HPXe TPCs quite viable. On the other hand, other important effects such as primary electron attachment during the path in the weak electric field of the drift region, and the quenching of the primary scintillation and electroluminescence for high distances, are effects that need to be studied.

KEYWORDS: Electroluminescence; molecular additives; xenon.

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Contents

1. Introduction

Experimental studies of Xe-CH_4 mixtures have show that it is possible to add small concentrations of molecular additives to xenon without significant loss in electroluminescence yield. CO_2 would be an interesting option due to its low cost and easy handling. In addition, simulation results of diffusion in Xe-CO_2 mixtures have shown that concentrations of the order of 0.1-0.2% of CO_2 would be sufficient to reduce transversal and longitudinal diffusion to highly acceptable values. The effect of the addition of CO_2 to pure xenon is not referred in the literature and, on the other hand, it has always been assumed that the presence of CO_2 impurities is a strong killer of electroluminescence. Nevertheless, the same assumption had been made for CH_4, which we proved in former studies to be an acceptable additive, in Xe-CH_4 mixtures for CH_4 concentrations lower than 0.5%. [1]
This way, we have studied the impact of the addition of 0.1 and 0.2% of CO_2 to pure xenon on the electroluminescence yield as well as on the energy resolution achievable with these mixtures.

2. Experimental Setup

The detector is a driftless Gas Proportional Scintillation Counter (GPSC) [2,3], with a 10 cm in diameter and 2.5-cm deep scintillation region, filled with xenon or xenon-CO_2 mixtures at pressures close to 1 bar, continuously purified through SAES St-707 getters, at a temperature of 100^{o}C to avoid absorption of the CO_2. The upper part of the detector body is made of Macor, which insulates the 8-mm in diameter Kapton radiation window and its stainless steel holder. Kapton, stainless steel and Macor are epoxied to each other. The Kapton window is aluminised on the inner side to ensure electrical conductivity. The lower part of the detector is made of stainless steel and connected to the gas circulation system. The bottom of the detector is a Macor disc epoxied to a 51-mm in diameter PMT and to the detector wall. A chromium grid with line width of ~100-μm and 1000-μm spacing is vacuum-deposited onto the PMT quartz window and connected to the photocathode pin through a continuous chromium

film deposited on the side surface. The upper and lower parts of the detector are vacuum-tight by compression of an indium gasket.

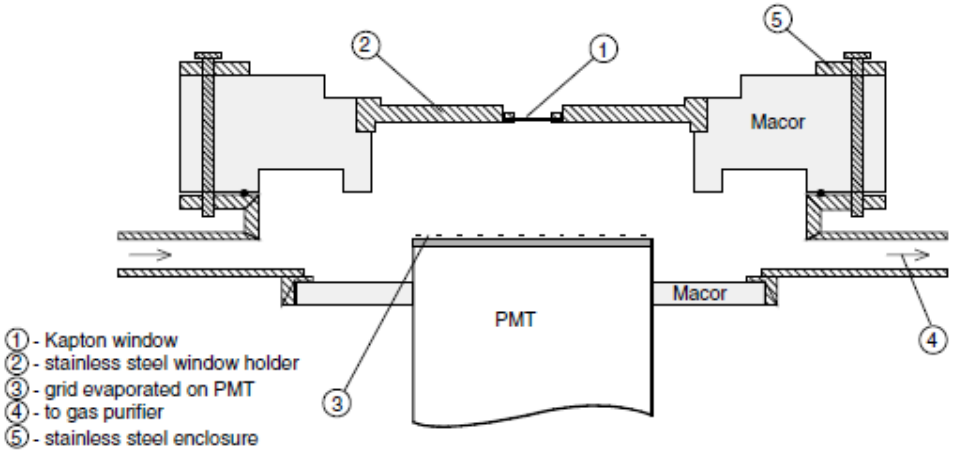

Figure 1: Scheme of the driftless GPSC used in this work [3].

The Kapton window and holder are kept at negative high voltage, while the chromium grid and PMT photocathode are kept at 0V. The window holder and the upper Macor piece were designed to ensure a uniform electric field in the scintillation region.

The charge signal from the PMT was pre-amplified with a Canberra 2004 preamplifier, with sensitivity of 9 mV/MeV and, subsequently, formatted with a HP 5582 A linear amplifier, using integration /differentiation constants of 5 μs. The formatted pulses were collected with a 1024-channel multichannel analyser (MCA). The collected signal corresponds to the total amount of EL produced in the whole scintillation region.

The different gas mixtures were achieved by adding known volumes filled with pure CO2 at known pressures to the xenon main detector chamber (figure 2).

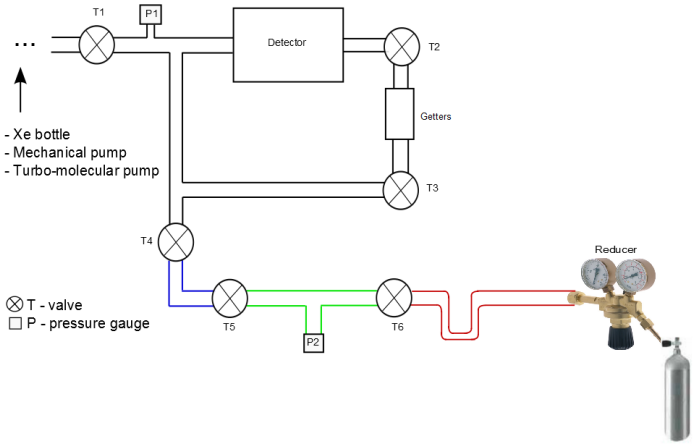

Figure 2: Scheme of both gas systems connected to the detector.

The detector was irradiated with a 5.9-keV X-ray beam from a ^{55}Fe radioactive source. A thin chromium film, placed between the radioactive source and the detector radiation window, absorbs most of the 6.4-keV x-rays of the Mn K_β line of the ^{55}Fe source.

3. Experimental Results

In a standard GPSC configuration [2], x-rays interact in a region between the detector radiation window and the scintillation region, where a weak electric field, below the gas electroluminescence threshold, is applied to guide the primary electron cloud, resulting from the x-ray interactions, towards the scintillation region. In the scintillation region, the electric field is high enough for the primary electrons to excite the noble gas atoms by electron impact, giving rise to a scintillation burst as a result of the noble gas atoms deexcitation processes.

In a driftless GPSC configuration [3], the x-ray interaction takes place directly in the scintillation region. This configuration was chosen to minimize any effect of electron attachment when the molecular additive is added to xenon. The effect of attachment during the primary electron cloud drift in the low electric field applied to the absorption/drift region of a standard GPSC could hinder the effect of the additive on the electroluminescence yield of the mixture, which is the parameter to be measured in this work. The intense electric field of the scintillation region minimizes the electron attachment, being the difference between pure xenon and xenon mixtures scintillation due to the difference in scintillation yields. On the other hand, in a driftless configuration, the amount of scintillation produced in the GPSC depends on the distance travelled by the primary electron cloud in the scintillation region and, therefore, on the x-ray interaction depth.

Figure 3 shows a typical pulse-height distribution obtained with pure xenon for 5.9-keV x-rays. The pulse-height distribution presents the typical Gaussian shape convoluted with an exponential tail towards the low-energy region, due to the x-ray penetration in the scintillation region. For 5.9-keV x-rays this tail is small, given its small absorption length in xenon, 2.7 mm at 1 bar, when compared to the thickness of the scintillation region, 25 mm. Therefore, for centroid as well as for FWHM measurements, only the right part of the distribution was fit to a Gaussian function, from which the centroid and the full-width-at-half-maximum were taken.

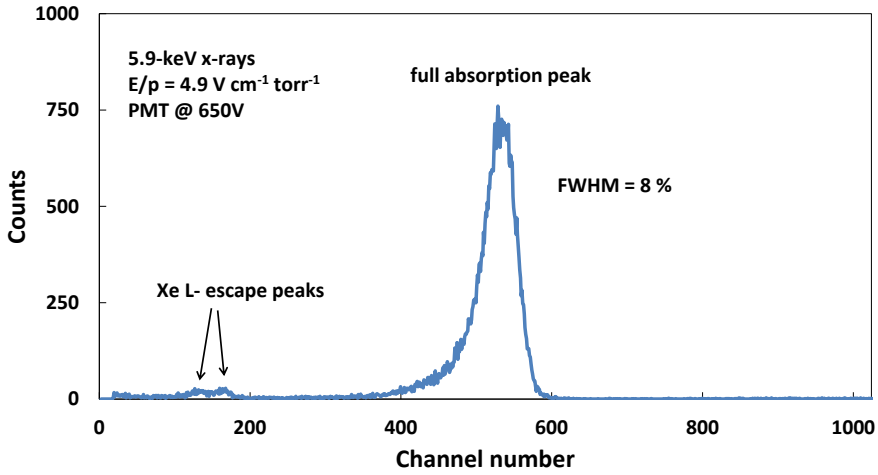

Figure 3: Pulse-height distribution for 5.9-keV x-rays absorbed in the xenon driftless GPSC. The PMT was biased at 650 V, the reduced electric field was 4.9 V cm^{-1} $torr^{-1}$ and formatting constants were 5 μs.

Figure 4 presents a typical behavior of electroluminescence relative amplitude and energy resolution as a function of E/p for a xenon filled GPSC. The amplitude exhibits a linear dependence on the reduced electric field in the scintillation region, with an intercept around 1.0 V cm^{-1} $torr^{-1}$, in agreement with the values reported in the literature [2]. For low E/p values, the

energy resolution presents a fast decrease with increasing E/p in the scintillation region due to the strong increase in the amount of electroluminescence produced, stabilizing for high reduced electric fields, as the electroluminescence reaches levels high enough for the statistical fluctuations inherent to the scintillation processes to become negligible.

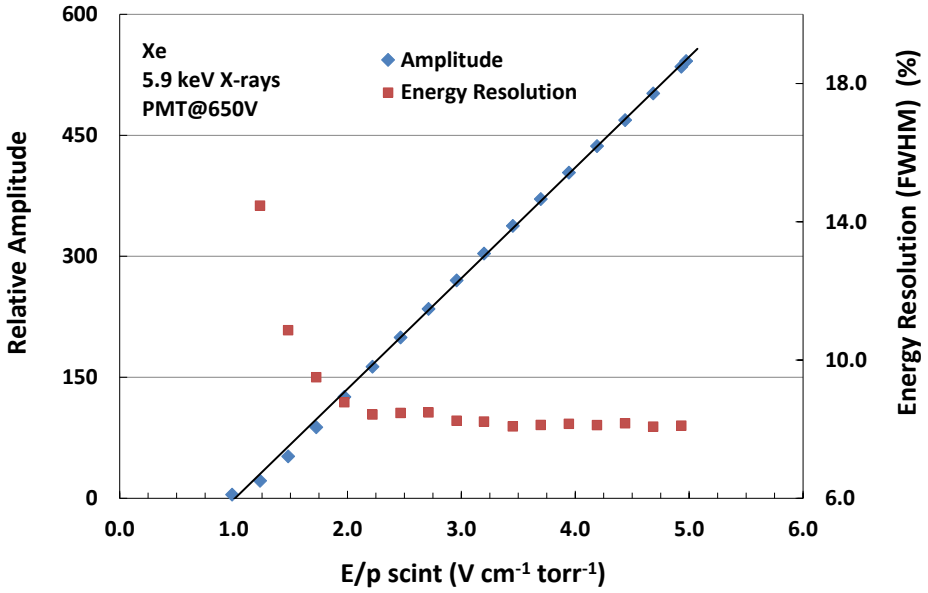

Figure 4: Relative amplitude and energy resolution (FWHM) as a function of reduced electric field, E/p, for pure xenon, for 5 μs shaping constants. A PMT bias voltage of 650 V was used and the gas pressure was 800 torr.

Mixtures of xenon and CO_2

In this section we present the results of xenon mixtures with 0.11% and 0.33% of CO_2. A comparison is made with the results for pure xenon.

Figure 5 shows the variation of the linear dependence of the amplitude on the reduced electric field with time, when 0.11% of CO_2 was added to the xenon, when the getters operation temperature was 80ºC. The excitation threshold is close to the one of pure xenon (1.0 V/cm/torr).At 80ºC the getters are not absorbing CO_2 and the mixture was stable for a long period of time.

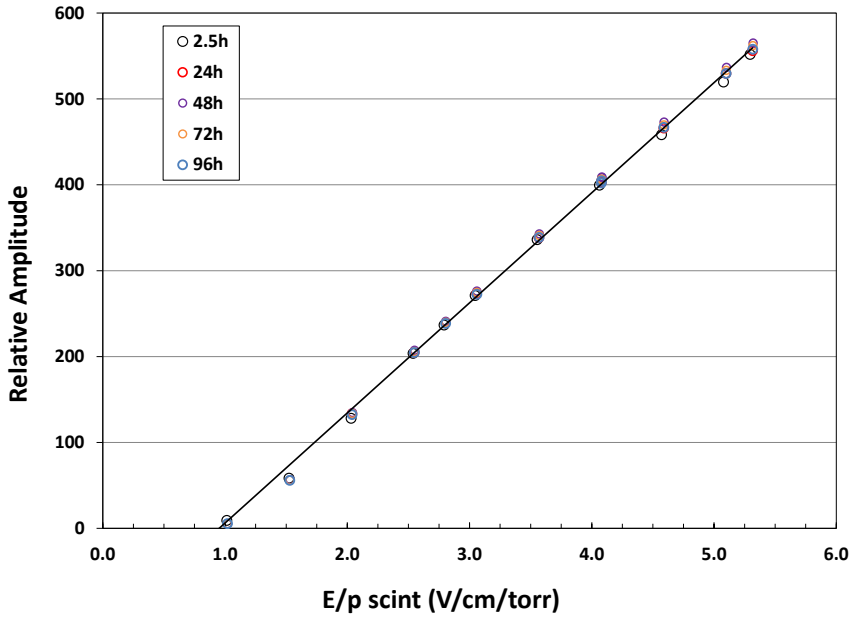

Figure 5: Relative amplitude as a function of reduced electric field, E/p, for the mixture of 99.89% Xe+0.11% CO_2 at 80ºC with a filling pressure of 750 torr, for a constant PMT bias voltage of 650 V and shaping constants of 5 μs.

Figure 6 shows amplitude and energy resolution as a function of reduced electric field for pure xenon and for the mixtures of 99.89% Xe+0.11% CO_2 and 99.67% Xe+0.33% CO_2. The getters operation temperature was 80ºC, the PMT bias voltage 650 V and the shaping constants 5 μs.

For the mixture 99.89% Xe+0.11% CO_2, the excitation threshold is close to the one of pure xenon, for the mixture of 99.67% Xe+0.33% CO_2 the threshold is around 1.2 V/cm/torr, higher than that for pure xenon, as expected. This means that a small percentage of carbon dioxide added to xenon in the driftless GPSC absorbs the energy that primary electrons gain from the electric field through inelastic collisions by exciting rotational and vibrational modes. Therefore, a high electric field is needed to produce a certain amount of scintillation.
The energy resolution still decreases with increasing reduced electric field and, at 5 V/cm/torr, is around 8.1%, 8.4% and 8.7% FWHM, for pure xenon, 99.89% Xe+0.11% CO_2 and 99.67% Xe+0.33% CO_2, respectively.

3.1 Electroluminescence Yield

Figure 7 presents the reduced EL yield, Y/N, i.e. the EL yield divided by the density of the gas, as a function of reduced electric field, E/N, in the scintillation region of the driftless GPSC. Experimental results from other authors and results from Monte Carlo simulation [4] are included for comparison. The absolute values presented in this work were obtained by normalizing the relative values measured for pure xenon to the absolute values of Monteiro et al. [5].

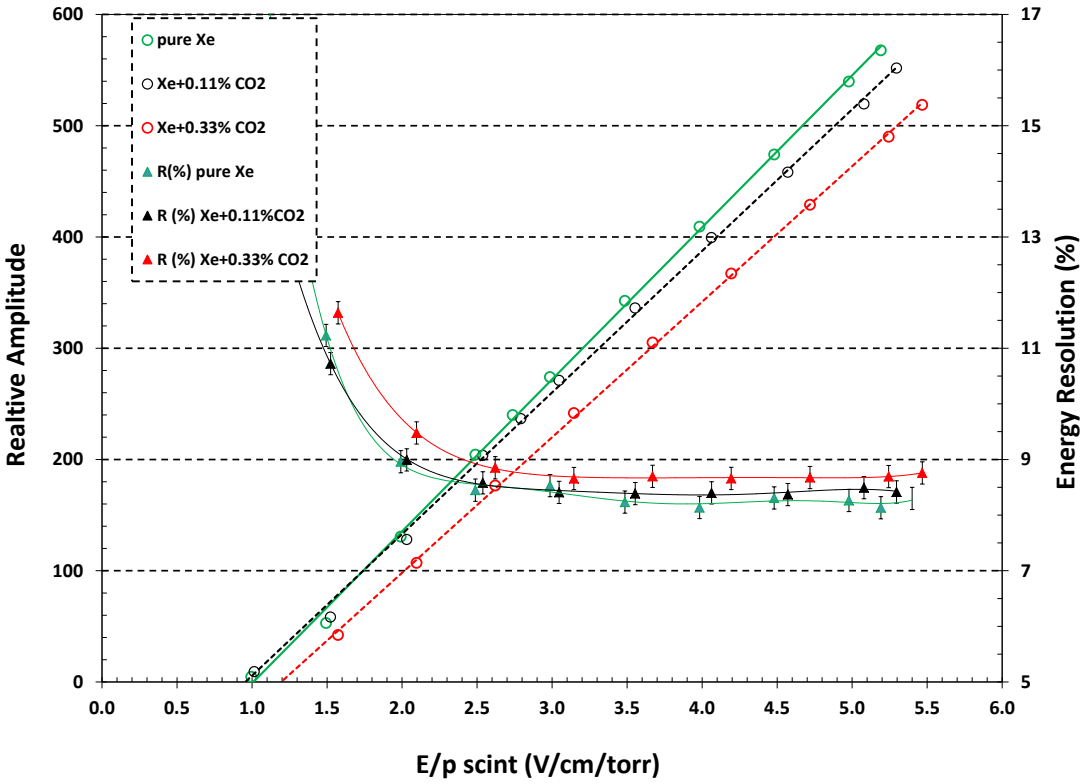

Figure 6: Relative amplitude as a function of reduced electric field, E/p, for pure xenon and for the mixtures of 99.89% Xe+0.11% CO_2 and 99.67% Xe+0.33% CO_2, for getters operating at 80°C, a constant PMT bias voltage of 650 V and shaping constants of 5 μs.

As expected, the addition of molecular gases to pure xenon reduces the electroluminescence yield of the gas mixture, decreasing with increasing molecular additive concentration. The primary electrons drift across the scintillation region undergoing a very large number of elastic collisions with the gas atoms/molecules. As the mass difference between the electron and the atoms is very high, the amount of energy lost in one elastic collision is negligible being, thus, the energy transfer from the electric field to the electrons very efficient. When the electrons acquire enough energy to excite the xenon atoms an elastic collision may occur leaving the atom in an excited state. In a collision between the electron and a polyatomic molecule, the electron energy may be lost to rotational and vibrational states, without the emission of electroluminescence, resulting in a reduced yield. As the number of inelastic collisions between two elastic collisions is higher than 10^4 [4], this effect becomes noticeable for molecular concentrations as low as few tenths of percent for CH_4 and CO_2. The addition of CO_2 has less impact on the electroluminescence yield and on the electroluminescence threshold than CH_4, which is a more complex molecule with more vibrational modes causing electrons to have higher energy losses along their path in the scintillation region.

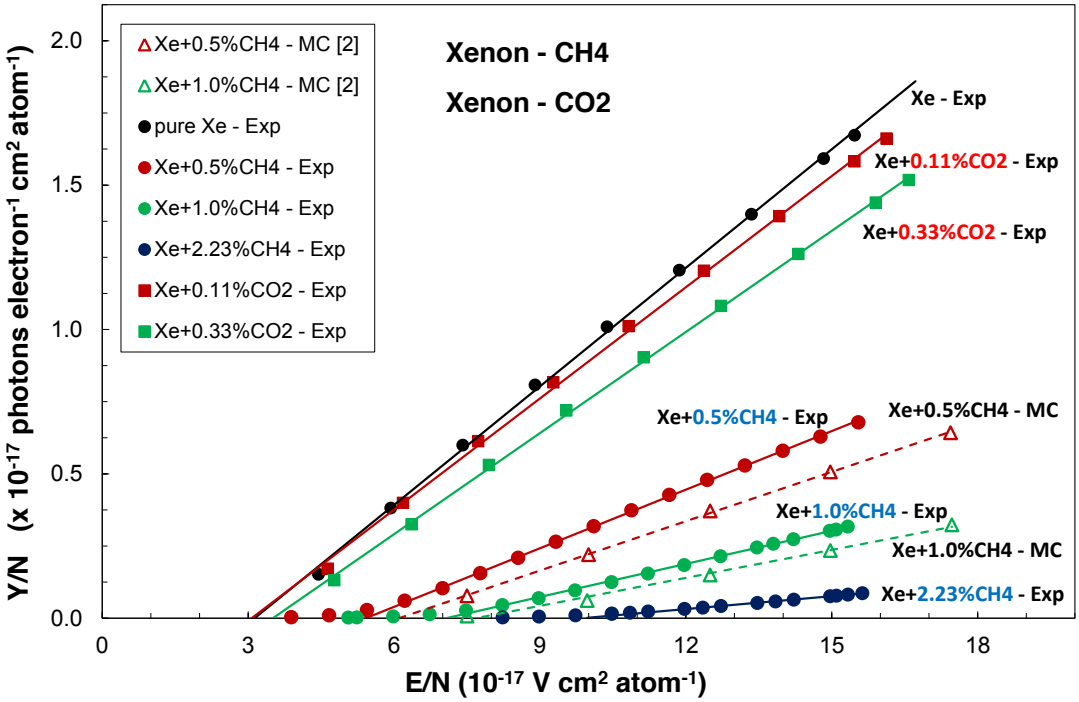

Figure 7: Xenon reduced electroluminescence yield as a function of E/N for our studies, as well as for Monte Carlo data in the literature [4].

Compared to pure xenon, the EL yield decreases to about 95% and 80% for 0.11%, and 0.33% of CO_2, respectively, for E/p between 1.5 and 2.25 V cm^{-1} $torr^{-1}$, a typical reduced electric field used in the NEXT-DEMO TPC, while for CH_4 this decrease is to about 30%, 10% and less than 3% for 0.5%, 1.0% and 2.2% CH_4, respectively. The amplification parameters and respective gas electroluminescence thresholds for the above mixtures are presented in Table I.

	Amplification parameter (photons/kV)	**Electroluminescence threshold (kV cm^{-1} bar^{-1})**
pure Xe	**137**	**1.0**
Xe+0.11%CO_2	**128**	**1.0**
Xe+0.33%CO_2	**117**	**1.2**
Xe+0.5%CH_4	**68**	**1.8**
Xe+1.0%CH_4	**38**	**2.3**
Xe+2.2%CH_4	**15**	**3.3**

3.2 Energy Resolution

The statistical fluctuations associated to the electroluminescence production and readout are an important parameter to be considered, since this is a major requirement for background reduction in double beta decay detectors. Figure 8 depicts the energy resolution (FWHM) obtained in the driftless GPSC for the different gas mixtures.

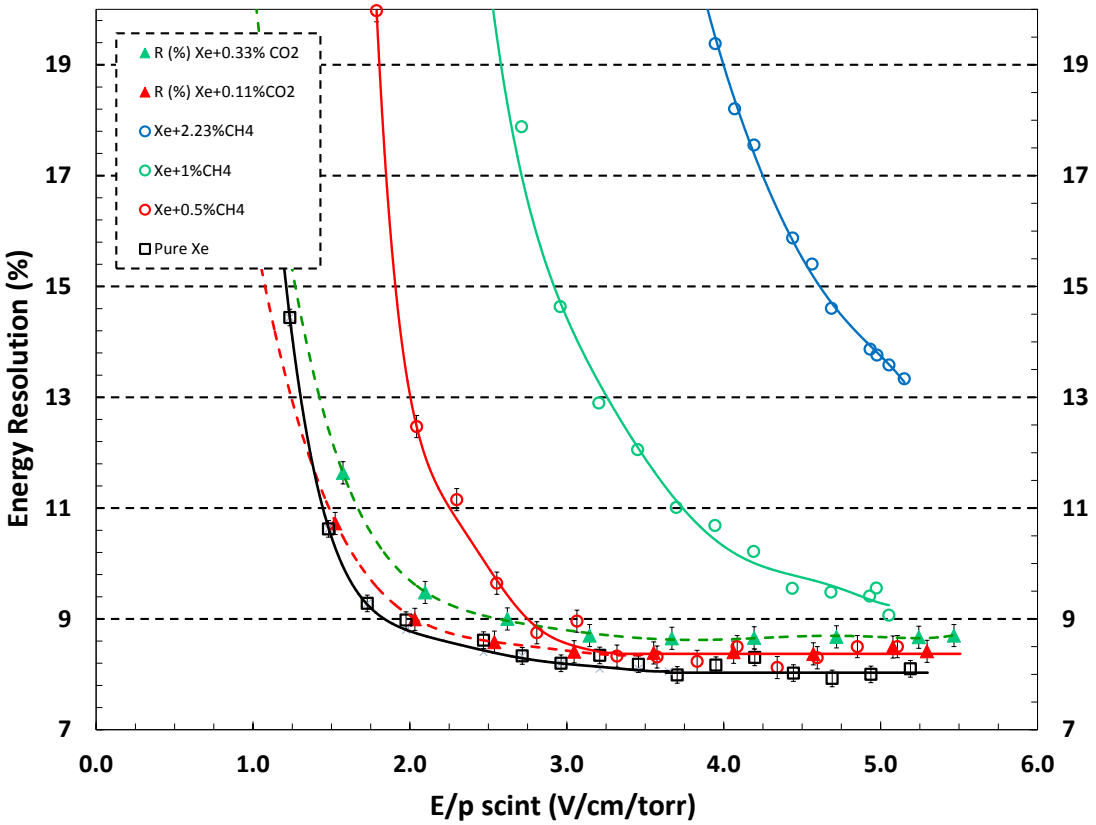

Figure 8: Energy resolution (FWHM) as a function of reduced electric field in the scintillation region, for pure xenon and for the Xe-CH_4 and Xe-CO_2 mixtures studied in this work. The lines serve only to guide the eye.

As seen in figure 8, the higher the fraction of molecular additive, the higher the energy resolution. For instance, for an E/p value of 1.5 V cm^{-1} $torr^{-1}$, the energy resolution has values of about 8.5%, 9.6% and 18% for pure xenon, Xe+0.5%CH_4, and Xe+1%CH_4, respectively. Xe-CO_2 mixtures achieve energy resolutions of about 8.6% and 9.0% for CO_2 concentration of 0.11%, and 0.33%, respectively, at an E/p value of 1.5 V cm^{-1} $torr^{-1}$. However, operating at higher reduced electric fields in the scintillation region improves the energy resolution to values close to those obtained for pure xenon. For E/p values above 1.8 V cm^{-1} $torr^{-1}$, energy resolutions around 8.4%, 8.4% and 8.7% can be obtained with Xe-0.5%CH_4, Xe-0.11%CO_2 and Xe-0.33%CO_2 mixtures, respectively. For Xe+1%CH_4 much higher electric field values would be needed in order to achieve such energy resolutions, while with Xe+2%CH_4 it is impossible to achieve good energy resolutions, as denoted in Fig.8.

The energy resolution of a GPSC is determined by the statistical fluctuations associated to the primary ionization processes, in the production of VUV scintillation light, and in the photosensor, and can be given by [2],

$$R(\%\,\mathrm{FWHM}) = 2.355\sqrt{\frac{F}{N}+\frac{1}{N}\left(\frac{J}{N_S}\right)+\left(\left(\frac{\sigma_{Ne}}{N_e}\right)^2+\frac{1}{N_e}\left(\frac{\sigma_q}{G_q}\right)^2\right)} \quad (1)$$

where N is the average number of primary electrons produced per incident x-ray, F is the relative variance of N, the Fano factor, N_S is the average number of scintillation photons produced per primary electron, J is the relative variance of N_S, $(\sigma_{Ne}/N_e)^2$ is the relative variance in the number of photoelectrons produced in the PMT per x-ray interaction, N_e, $(\sigma_q/G_q)^2$ are the fluctuations in the electron multiplication gain of the photosensor. Other contributions, e.g. of geometric nature, as those due to variations in the solid angle subtended by the PMT photocathode relative to the region where the scintillation occurred and electronic noise, are not taken into account in Eq.(1).

The second term under the radical of Eq.(1) can be neglected since $J<<F$ and $N_s>100$. The number of primary electrons produced in an x-ray interaction is given by the energy of the x-ray divided by the mean energy to produce an electron-ion pair, $N=E_x/w$. As the photoelectron production is a Poisson process, while the gain of each photoelectron in the PMT avalanche approaches an exponential distribution, the energy resolution of the GPSC can be approximated to [1]

$$R(\%\,\mathrm{FWHM}) = 2.355\sqrt{\frac{Fw}{E_x}+\frac{2}{N_e}} \quad (2)$$

As N_e is proportional to the pulse amplitude, a plot of R^2 as a function of the inverse of the relative amplitude, $N_e = kA^{-1}$, varying the reduced electric field in the scintillation region, will present a linear trend, where the detector limit resolution can be obtained by extrapolating that line to infinite light yield.

$$R^2 = 5{,}546\left(\frac{Fw}{E_x}+\frac{k}{A}\right) \quad (3)$$

Figure 9 depicts the R^2 dependence on A^{-1} for the present GPSC and for the different gas mixtures. A linear trend is observed for a certain range of amplitudes. The deviation from this trend is due to different processes occurring in the detector as the reduced electric field in the scintillation region changes. For example, the detector energy resolution degrades for high reduced electric fields due to the additional electronic fluctuations introduced by instabilities in the electric insulation and/or to the additional fluctuations introduced by electron multiplication, for electric field values above the gas ionization threshold. On the other hand, for low values of the reduced electric field, the energy resolution degradation is faster than predicted by Eq.(3) due to the poorer signal-to-noise ratio and higher diffusion of primary electrons, which introduce additional fluctuations in the measured pulse amplitudes.

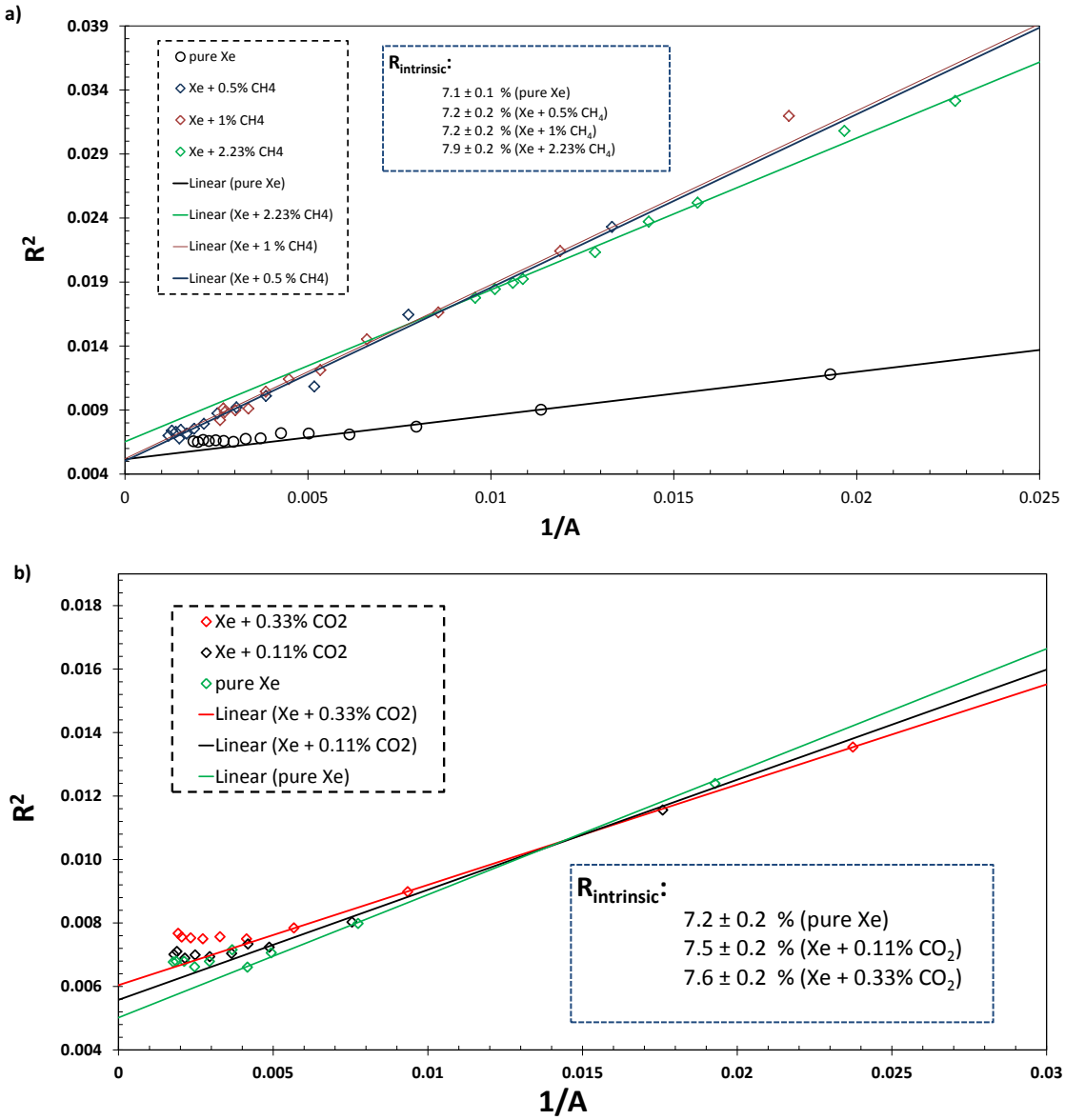

Figure 9: Dependence of R^2 on A^{-1} for the present GPSC, for the different Xe+CH_4 mixtures (a) and different Xe+CO_2 mixtures. As expected, a linear trend is observed for a certain range of amplitudes.

Table II presents the experimental values for the intrinsic energy resolution obtained for the different mixtures. The values of Table II, obtained with this method, present an upper limit, since the experimental values obtained for the GPSC energy resolution include additional contributions inherent to the present experimental setup, experimental conditions and gas purity.

	Intrinsic Energy Resolution FWHM (%)
pure Xe	7.1±0.2
Xe+0.11%CO_2	7.3±0.2
Xe+0.33%CO_2	7.7±0.2
Xe+0.5%CH_4	7.1±0.1
Xe+1.0%CH_4	7.3±0.1
Xe+2.2%CH_4	8.1±0.1

4. Conclusions

For Xe-CO_2 mixtures, the intrinsic energy resolutions are lower for low reduced electric fields, e.g. around 1.5 V cm^{-1} $torr^{-1}$, the typical field used in Next-DEMO, than those achieved with Xe+0.5%CH_4, due to the higher electroluminescence threshold and lower electroluminescence yield of the latter. For higher electric fields the intrinsic energy resolutions achieved with Xe-CO_2 are slightly higher than that of Xe+0.5%CH_4.

Comparing with pure xenon, Xe+0.11%CO_2 and Xe+0.5%CH_4 do not present significant degradation of the electroluminescence yield and associated statistical fluctuations, being the use of such mixtures in HPXe TPCs quite viable, without jeopardizing the performance for DBD detection. On the other hand, other important effects such as primary electron attachment during the path in the weak electric field of the drift region, and the quenching of the primary scintillation and electroluminescence for high distances, are effects that need to be studied in large-volume detectors, e.g. in NEXT-DEMO, now that the impact on the electroluminescence yield is known.

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