Performance of Argon-Xenon Mixtures in a Gas Proportional Scintillation Counter for the 0.1–10 keV X-Ray Region

S. J. C. do Carmo, F. I. G. M. Borges, and C. A. N. Conde, *Life Member, IEEE*

***Abstract*—The performance of a gas proportional scintillation counter filled with either pure Xe or Ar-Xe mixtures is described for the 0.1–10 keV X-ray energy range. It is shown that the spectra tail distortion exhibited by Xe filled gaseous detectors for soft X-rays below 2 keV is reduced if Ar-Xe mixtures are used. The peak-to-valley ratio increases by a factor of about 2 or 3 as the Xe concentration is reduced from 100% to 5%. Moreover, the energy resolutions for such mixtures are similar to the ones for pure Xe.**

***Index Terms*—Argon detectors, energy resolution, gas detectors, proportional gas scintillation detectors, x-ray detectors, x-ray spectroscopy, xenon detectors.**

I. INTRODUCTION

GAS Proportional Scintillation Counters (GPSC) are room temperature gaseous radiation detectors with many advantages, like their good energy resolution and large detectable energy range (typically from 0.1 to 100 keV), which allow their use in many fields such as X-ray spectrometry or high energy physics [1]–[3]. Soft X-ray detection is an area where GPSCs are particularly competitive since they can outperform even solid-state detectors in the low energy range [4]. The techniques developed for large area detectors using relatively small photosensors for the scintillation light [5], [6] present an advantage that can be used for all types of radiation detected.

GPSCs are usually filled with pure Xe at slightly above atmospheric pressure and the consequent short absorption length of soft X-rays (few hundred μm for energies below 1 keV) is at the origin of the loss of primary electrons to the detector entrance window by backscattering [7]. Spectra distortion thus appears as a low energy tail which consequently worsens the energy resolution and the peak-to-valley ratio.

Previous research works have been carried out in order to attenuate this undesirable spectra distortion, either by increasing the electric field in the absorption region [8] or by eliminating the absorption region with the so called driftless GPSC [9]. The latter authors have shown very good results in attenuating this distortion, but the technique requires elaborate electronic pulse processing. Alternatively, performance improvements were also observed by filling the detector with a lighter gaseous mixture in order to increase the X-ray absorption length [10]. Indeed, the addition of a lighter gas to Xe leads not only to longer X-ray absorption lengths and so to reduced tails [7] but can also lead to Penning mixture effects which originate improved Fano factors and w-values. The use of Ne-Xe mixtures as an alternative to pure Xe has already been studied with improved results in reducing the low energy tail distortion but they do not exhibit the expected energy resolution improvement [10]. This was attributed to the lower secondary scintillation yields and larger w-values of Ne-Xe mixtures in comparison with pure Xe ones.

Ar-Xe mixtures have been considered as valid alternatives to pure Xe for GPSCs [11] since, not only they provide longer absorption lengths than pure Xe, but they also simultaneously present similar or even improved energy resolutions, w-values, Fano factors and secondary scintillation yields in comparison to pure Xe [12]–[16]. However, the increase in the absorption length, apart for the 250–630 eV energy range, is not expected to be as large as for Ne-Xe mixtures [7].

Due to the continued interest in soft X-ray detectors for general purpose applications and to our particular interest in studying low energy and low atomic number PIXE, we have carried out the present work, which consists in the study of the performance of a GPSC filled with pure Xe and Ar-Xe mixtures for X-rays in the 0.1–10 keV energy range. Particular attention was paid to the 0.1–2 keV energy range, where the improvements are expected to be more significant.

In Section II, we describe the experimental set-up used to perform this study and the results obtained for the energy resolutions and for the peak-to-valley ratios (PVR).

II. EXPERIMENTAL RESULTS

The set-up of the GPSC used for the present work is represented in Fig. 1. This uses a 10 dynode EMI 9266QB photomultiplier tube (PMT) and has already been described in detail in [13]. The detector was filled at a pressure slightly above atmospheric one, 780 Torr, with either pure Xe or various Ar-Xe mixtures. The filling gases were continuously purified by convection with ST707 getters from SAES.

The detector window was manufactured from a 0.9 μm thick Mylar film glued onto a 4 mm thick stainless steel frame. This frame had 6 holes 1.5 mm in diameter circularly distributed 1.5 mm apart around a similar seventh hole centered in the middle of the frame. These low area holes enable the Mylar film to stand the atmospheric pressure when vacuum operations are

---

Manuscript received November 13, 2008; revised January 13, 2009. Current version published April 08, 2009. This work was carried out in the Atomic and Nuclear Instrumentation Group of the Instrumentation Center (Unit 217/94) of Departamento de Física da Universidade de Coimbra, Portugal. This work was supported by FEDER and Programme POCI 2010 through Project POCI/FP/81918/2007 of Fundação para a Ciência e a Tecnologia (FCT). S.J.C. do Carmo acknowledges doctoral Grant SFRH/BD/28534/2006 from Fundação para a Ciência e a Tecnologia (FCT).

The authors are with Departamento de Física da Universidade de Coimbra, P-3004-516 Coimbra, Portugal (e-mail: sergiocarmo@gian.fis.uc.pt).

Digital Object Identifier 10.1109/TNS.2009.2013952

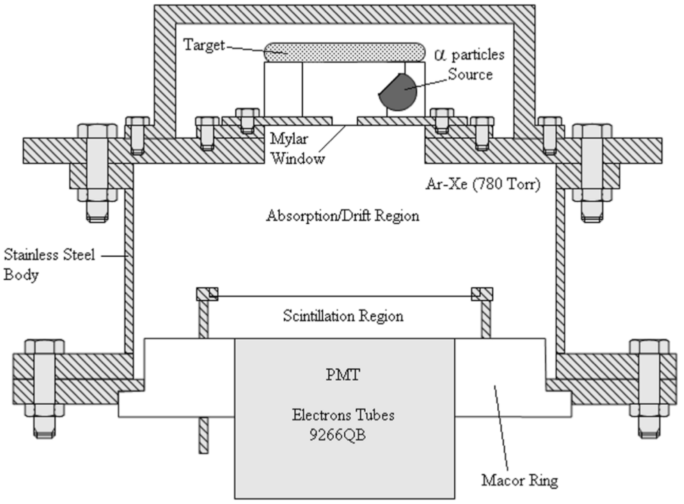

Fig. 1. Schematic of the GPSC used in the present work.

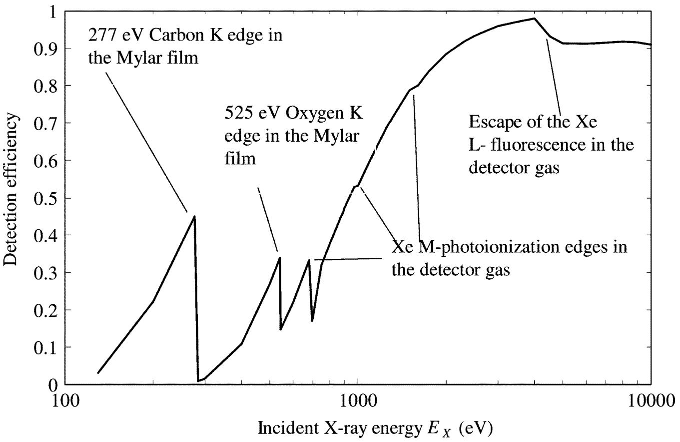

Fig. 2. Calculated detection efficiency of the present GPSC filled with pure Xe as a function of the energy E_X of the incident X-ray.

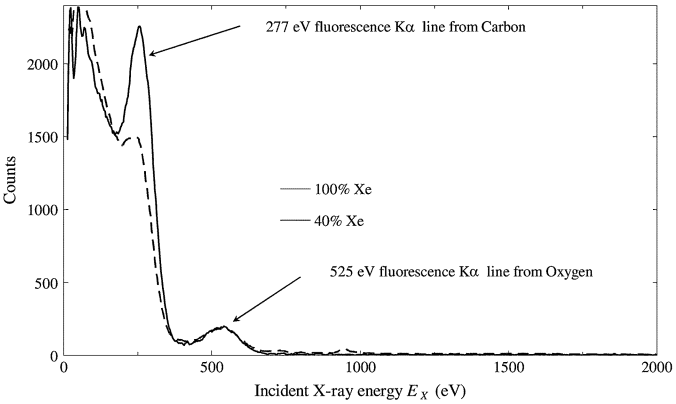

Fig. 3. Experimental pulse height-distributions in pure Xe and 60%Ar-40%Xe for the 277 and 525 eV Kα fluorescence lines from C and O respectively.

needed. A 40 nm thick aluminum layer was vacuum evaporated onto the Mylar film to electrically ground the window.

The detection efficiency of the detector (Fig. 2) was calculated in a way described in [4] by taking into account the transmission of both Mylar and Al layers [17] and the loss of primary electrons by backscattering to the detector window [7], which depends on the filling gas used.

For each Ar-Xe mixture studied the reduced electric fields in both absorption and scintillation regions were chosen in order to reach the optimal energy resolution [12], keeping the PMT voltage constant. X-ray spectra were obtained for several X-ray energies E_X and for several Ar-Xe mixtures with 100%, 40%, 20%, 10% and 5% Xe concentrations, as shown in Table I and Figs. 3–4. These different X-ray energies E_X correspond to the unfiltered Kα and Kβ fluorescence lines of the various elements, obtained by exciting pure or compound element targets, from C to Ge, with α-particles from a 30 kBq ^{244}Cm radioactive source placed under vacuum. As we can observe in Fig. 4, the spectra present high counting rates in the very low energy region. This behaviour is due to the presence of electronic noise and of the C and O X-ray fluorescence K-lines resulting from the interaction of the incident X-ray beam with the C and O atoms present in the Mylar window.

The experimental pulse height distributions were fitted to Gaussian distributions in order to determine the position of their centroid c and their Full Width at Half Maximum (FWHM) H. The performance of the GPSC is evaluated in terms of the energy resolution $R = H/c$, and the Peak-to-Valley ratio (PVR) [10], which is defined as the ratio of the counts at the centroid c and the average number of counts for 5 points located at a distance of 1.5 FWHM from the peak [18]. The PVR gives a quantitative measurement of the distortion of the spectrum in the low energy region.

Table II shows that the PVR generally improves by a factor of about 2 or 3 as the Ar concentration increases. This behaviour shows that the loss of primary electrons to the detector window is reduced with the addition of Ar, as it can be seen in the spectra shown in Fig. 4. However, these improvements in the reduction of the spectra distortion do not result in the expected improvement of the energy resolution, R, as shown in Table I. Indeed, R remains approximately constant for pure Xe and the Ar-Xe mixtures. This might be explained by the slightly lower scintillation yield of Ar-Xe when compared to pure Xe observed in [12].

Moreover, this improvement in the PVR is not achieved for the C and O Kα lines (277 eV and 525 eV respectively) (Fig. 3) since, as we have already mentioned, the absorption length is shorter in Ar than in Xe in the 250–630 eV energy range. This also explains why, for this range, the energy resolution R worsens with increasing Ar concentrations (Table I). In fact, the C peak becomes almost undistinguishable from the low energy tail, becoming impossible to determine its energy resolution for low Xe concentrations (Table I).

Fig. 5 confirms that the energy resolution R is approximately proportional to the square root of the reciprocal X-ray energy $1/E_X$ [1], in exception for the data corresponding to very low energy X-rays which are above the linear behaviour. This is due to the significant worsening of R below approximately 1 keV because of the low energy tail distortion. Small deviations apart this linear behaviour can also be attributed to the expected discontinuities in the Fano factors F and the w-values for Ar-Xe mixtures with the X-ray energy E_X. Indeed, such variations in F and w have been studied before for pure Xe [19] and Ne-Xe

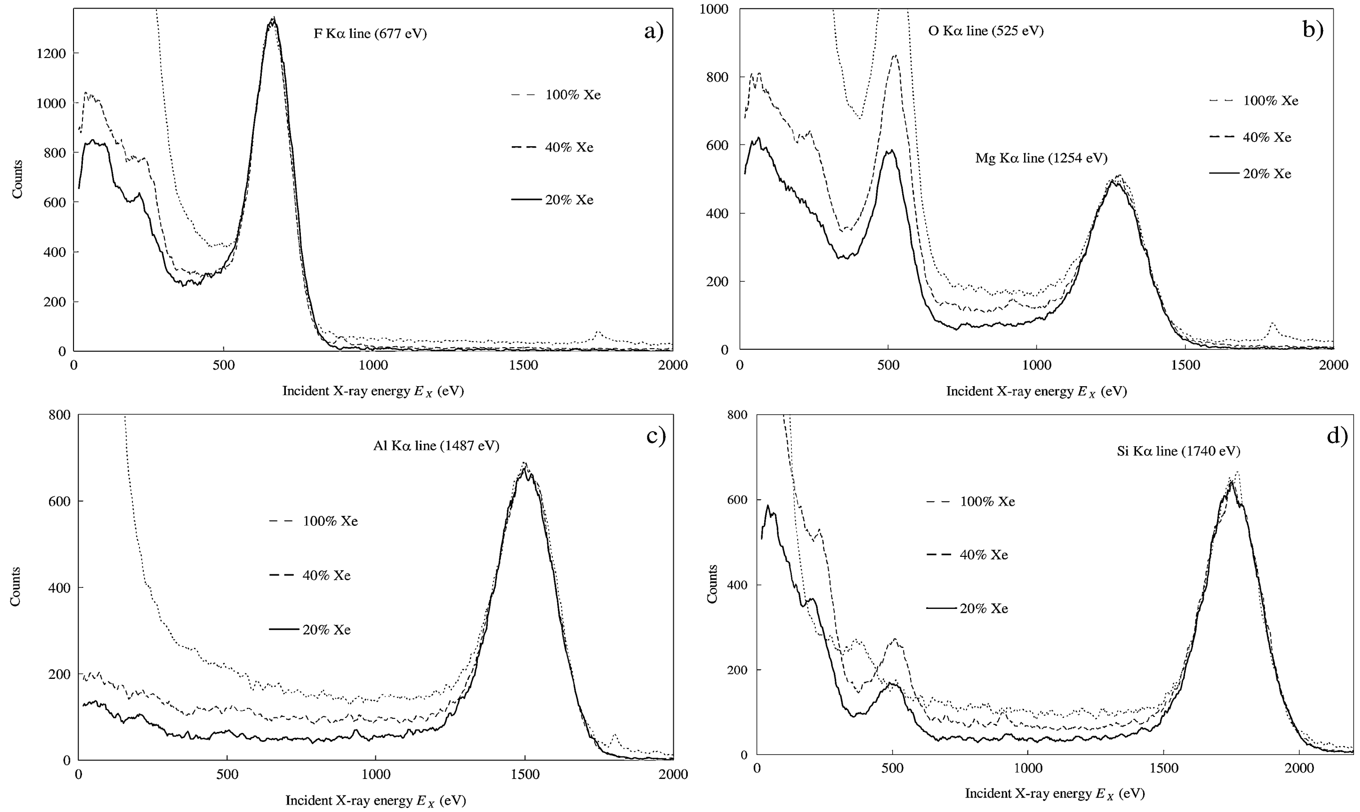

Fig. 4. Experimental pulse height-distributions in pure Xe, 60%Ar-40%Xe and 80%Ar-20%Xe for (a) 677 eV, (b) 525 eV and 1254 eV, (c) 1487 eV and (d) 1740 eV X-rays (F, O, Mg, Al and Si Kα fluorescence lines respectively).

TABLE I
ENERGY RESOLUTIONS, R, OBTAINED FOR THE GPSC FILLED WITH SEVERAL AR-XE MIXTURES FOR THE Kα X-RAYS FROM LOW ATOMIC NUMBER ELEMENTS

Element	Kα line energy E_x (eV)	R (%) ± 0.1				
		5% Xe	10% Xe	20% Xe	40% Xe	100% Xe
C	277	-	-	57.5	42.6	37.2
O	525	34.5	33.4	30.4	24.7	24.1
F	677	24.4	24.0	24.3	22.3	21.9
Mg	1254	17.5	17.5	18.3	18.1	16.9
Al	1487	15.3	15.9	15.7	15.3	15.7
Si	1740	14.7	14.7	14.5	14.4	13.7
S	2307	13.2	12.9	12.9	12.5	11.7
Cl	2622	12.9	12.8	12.3	11.8	11.5
Ca	3690	10.2	10.0	9.7	9.7	9.4
Ti	4508	9.5	9.5	9.4	9.1	8.8
V	4949	9.4	9.4	9.3	9.2	8.8
Cr	5411	9.3	9.0	9.0	9.0	8.7
Mn	5895	9.2	8.6	8.6	8.6	8.4
Co	6925	8.7	7.9	8.1	8.5	7.6
Ni	7422	8.5	8.1	7.9	7.8	7.6
Cu	8041	8.4	8.1	7.8	7.5	7.4
Zn	8631	8.1	7.9	7.7	7.5	7.3
Ge	9876	7.9	7.8	7.5	7.5	7.1

mixtures [20], but no data is available in the literature for Ar-Xe mixtures.

We have also observed, as illustated in Fig. 6, that the response of the detector is linear within the experimental error, apart for the discontinuities at the Xe L-, Xe M- and Ar K-edges [21]. We have also noticed that the discontinuities at the Xe L- and M-edges decrease as the Xe concentration decreases while the Ar K-edge becomes more pronounced as the Ar concentration increases. The values of these discontinuities in the linearity were calculated in a way described in [19]. Discontinuities of 63 and 37 eV were obtained in pure Xe for the Xe L- and M-edges respectively for instance, values which are in good agreement with the published data [19], [22], [23].

TABLE II
PEAK-TO-VALLEY RATIOS FOR DIFFERENT SOFT X-RAY ENERGIES AND FOR THE GPSC FILLED WITH XE AND AR-XE MIXTURES

% Xe	Peak-to-Valley ratios			
	F	Mg	Al	Si
100	2.9	2.9	4.8	6.2
40	4.4	3.4	6.7	8.8
20	4.9	6.9	9.7	14.2
10	3.2	8.2	10.5	14.7
5	2.7	6.9	11.0	15.7

III. CONCLUSION

We have shown that gas proportional scintillation counters filled with Ar-Xe mixtures present energy resolutions for soft X-rays that, apart from the 250–630 eV energy range, are almost as good as the ones for pure Xe, confirming the preliminary results presented in [11].

Moreover, the peak-to-valley ratios, that measure the distortion present in soft X-ray spectra, generally improve as the Xe concentrations decrease from 100% down to 5%. Although these improvements in the peak-to-valley ratios are not as

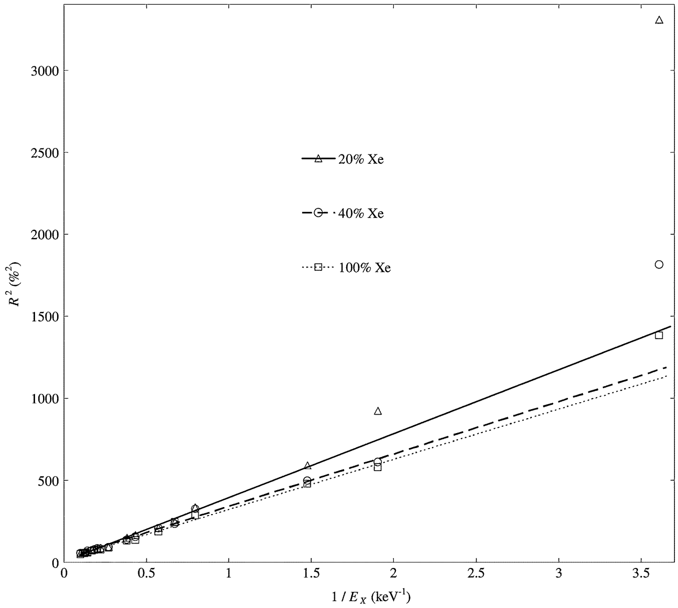

Fig. 5. Plot of R^2 as a function of the reciprocal X-ray energy, $1/E_X$.

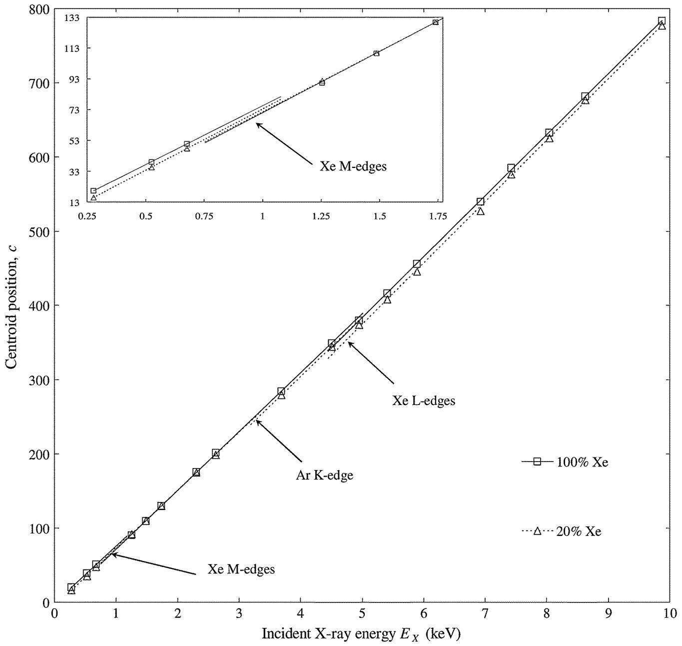

Fig. 6. Centroid position c as a function of the incident X-ray energy E_X for pure Xe and Ar-20%Xe.

important as the ones observed in [10] for Ne-Xe mixtures, Ar-Xe represent a better alternative to pure Xe due to the better energy resolutions reachable.

REFERENCES

[1] C. A. N. Conde, "Gas Proportional Scintillation Counters for X-Ray Spectrometry," in *X-Ray Spectrometry: Recent Technological Advances*, K. Tsuji, J. Injuk, and R. Van Grieken, Eds. New York: Wiley, 2004, pp. 195–215.

[2] E. Aprile, A. E. Bolotnikov, A. I. Bolozdynya, and T. Doke*, Noble Gas Detectors*. New York: Wiley-VCH, 2006.

[3] A. J. P. L. Policarpo, "The gas proportional scintillation counter," *Space Sci. Instrum.*, vol. 3, pp. 77–107, 1977.

[4] F. I. G. M. Borges, J. M. F. dos Santos, T. H. V. T. Dias, F. P. Santos, and C. A. N. Conde, "A gas proportional scintillation counter for x-ray spectrometry in the 0.1–3 keV range," *X-Ray Spectrom.*, vol. 33–2, pp. 124–127, 2004.

[5] C. A. N. Conde, J. M. F. dos Santos, and A. C. S. S. M. Bento, "Gas Proportional Scintillation Counters for Ionizing Radiation With Medium and Large Size Radiation Windows and/or Detection Volumes" U.S. Patent 5 517 030, May 14, 1996 [Online]. Available: http://www.uspto.gov/patft/index.html, or in http://www.google.com/patents?id=h7knAAAAEBAJ&dq=5517030

[6] C. A. N. Conde, J. M. F. dos Santos, and A. C. S. S. M. Bento, "Gas Proportional Scintillation Counter for Ionizing Radiation With Medium and Large Size Radiation Windows and/or Detection Volumes," European Patent EP 0616722 BI, Dec. 30, 1998 [Online]. Available: http://ep.espacenet.com

[7] F. I. G. M. Borges, F. P. Santos, T. H. V. T. Dias, P. J. B. M. Rachinhas, C. A. N. Conde, and A. D. Stauffer, "Xenon-neon gas proportional scintillation counters for x-rays below 2 keV: A Monte Carlo simulation study," *IEEE Trans. Nucl. Sci.*, vol. 49, no. 3, pp. 917–922, Jun. 2002.

[8] F. P. Santos, J. M. F. dos Santos, T. H. V. T. Dias, and C. A. N. Conde, "Pulse-Height spectrum distortion in Xenon gaseous detectors for soft X-rays," *IEEE Trans. Nucl. Sci.*, vol. 42, no. 4, pp. 611–614, Aug. 1995.

[9] D. G. Simons and P. A. J. de Korte, "Soft X-ray energy resolution and background rejection in a driftless gas scintillation proportional counter," *Nucl. Instrum. Methods Phys. Res. A*, vol. A277, pp. 642–656, 1989.

[10] F. I. G. M. Borges, F. P. Santos, J. M. F. dos Santos, T. H. V. T. Dias, P. J. B. M. Rachinhas, and C. A. N. Conde, "Xenon-neon gas proportional-scintillation counters for X-rays below 2 keV: Experimental results," *IEEE Trans. Nucl. Sci.*, vol. 50, no. 4, pp. 842–846, Aug. 2003.

[11] H. Inoue, K. Koyama, M. Matsuoka, T. Ohashi, Y. Tanaka, and H. Tsunemi, "Properties of gas scintillation proportional counters for soft X-rays," *Nucl. Instrum. Methods*, vol. 157, pp. 295–300, 1978.

[12] S. J. C. do Carmo, A. M. F. Trindade, and F. I. G. M. Borges, "Ar-Xe mixtures and their use in curved grid gas proportional scintillation counters for X-rays," *J. Instrum.*, Mar. 2009.

[13] S. J. C. do Carmo, F. I. G. M. Borges, F. L. R. Vinagre, and C. A. N. Conde, "Experimental study of the *w*-values and Fano factors of gaseous xenon and Ar-Xe mixtures for x-rays," *IEEE Trans. Nucl. Sci.*, vol. 55, no. 5, pp. 2637–2642, Oct. 2008.

[14] S. Kubota, "Non-metastable Penning effect in the alpha-particle ionization of inert gas mixtures," *J. Phys. Soc. Jpn.*, vol. 29, no. 4, pp. 1017–1029, 1970.

[15] T. Takahashi, S. Himi, M. Suzuki, J. Ruan, and S. Kubota, "Emission spectra from Ar-Xe, Ar-Kr, Ar-N2, Ar-CH4, Ar-CO2 and Xe-N2 gas scintillation proportional counters," *Nucl. Instrum. Methods*, vol. 205, pp. 591–596, 1983.

[16] H. Brunet, A. Birot, H. Dijols, J. Galy, P. Millet, and Y. Salamero, "Spectroscopic and kinetic analysis of the VUV emissions of argon and argon-xenon mixtures: II. Energy transfer in Ar-Xe mixtures," *J. Appl. Phys B*, vol. 15, pp. 2945–2967, 1982.

[17] [Online]. Available: http://henke.lbl.gov/optical_constants

[18] R. M. Keyser, Characterization of Room Temperature Detectors Using the Proposed IEEE Standard, Oak Ridge, TN [Online]. Available: http://www.ortec-online.com/papers/roomtemp_det.pdf

[19] T. H. V. T. Dias, J. M. F. dos Santos, J. P. B. M. Rachinhas, F. P. Santos, C. A. N. Conde, and A. D. Stauffer, "Full-energy absorption of X-ray energies near the Xe L- and K-photoionization thresholds in xenon gas detectors: simulation and experimental results," *J. Appl. Phys.*, vol. 82, no. 6, pp. 2742–2753, 1997.

[20] F. Santos, T. H. V. T. Dias, P. J. B. M. Rachinhas, C. A. N. Conde, and A. D. Stauffer, "Monte Carlo simulation study of the Fano factor, w-value, and energy resolution for the absorption of soft x rays in xenon-neon gas mixtures," *J. Appl. Phys.*, vol. 89, no. 12, pp. 8202–8213, 2001.

[21] [Online]. Available: http://www.csrri.iit.edu/periodic-table.html

[22] J. M. F. dos Santos, C. A. N. Conde, and A. C. S. S. M. Bento, "The energy linearity of gaseous xenon radiation detectors for X-rays with energies between 2 and 60 keV: Experimental results," *Nucl. Instrum. Methods Phys. Res. A*, vol. A324, pp. 611–613, 1993.

[23] F. I. G. M. Borges, T. H. V. T. Dias, F. P. Santos, P. J. B. M. Rachinhas, J. M. F. dos Santos, and C. A. N. Conde, "An investigation of the M-discontinuities in the response of xenon gas detectors to X-rays in the 0.1–2 keV range," *Nucl. Instrum. Methods Phys. Res. A*, vol. A505, pp. 242–246, 2003.