A new technique for studying the Fano factor and the mean energy per ion pair in counting gases

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ABSTRACT

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A new method is presented for deriving the Fano factor, F , and the mean energy per ion pair, W_i , in counting gases. It is based on the technique of individual counting of single ionization electrons induced in low-pressure gas samples by soft x-ray photons. A correlation of the experimental data with a detailed simulation of the electron deposition and counting process permits the extraction of the Fano factor and the mean energy per ion pair values. We present data of F and W_i for C_2H_6 and Ar/C_2H_6 over the energy range of 100-1500 eV. The energy dependence of these parameters reflects the atomic level structure of the gases. We discuss in detail the accuracy of this technique and its advantages and limitations. Ways are proposed for improving of the technique and for broadening the energy range.

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1 Introduction

A fundamental topic in radiation physics and dosimetry deals with the number of ion pairs generated in matter and its fluctuation, following the absorption of an energy quantum. The dissipation of energy released by charged particles involves primary ionization followed by additional ionization and excitation phenomena. In the case of x-rays, additional processes exist related to photo-absorption. These processes include the Auger/Coster-Kronig transitions, fluorescence, electron shake-up and electron shake-off. Both the charged-particle energy loss and the x-ray energy absorption can be described by two energy-dependent parameters: the mean energy per ion pair (W_i) and the Fano factor (F) [1, 2].

Most of the theoretical studies of these two parameters were done using charged particles, because of their close relation to dosimetry and the relative simplicity of the involved processes. The common theoretical approaches are the Monte-Carlo method (see for example ref. [3]), and an analytical approach using the Spencer-Fano equation [4, 5, 6]. The solution of this equation provides the electron degradation spectrum, which is used to calculate F and W_i . Under a stationary condition (constant impinging radiation flux), the electron degradation spectrum represents the energy spectrum of all electrons (primaries, secondaries etc..) released in the matter, and thus provides an important tool for understanding the processes involved. For exact mathematical definitions see ref. [4].

However, both methods require the knowledge of all the involved interaction cross-sections, thus limiting their usefulness to a few simple gases such as Ar, He, H₂, and tissue-equivalent (TE) mixtures. All theoretical studies have shown that both W_i and F values increase when the electron energy decreases below a few hundreds eV, and reach asymptotic values for electron energy above a few keV. This behavior of W_i and F was explained by the weak dependence of the electron degradation spectrum on the source electron energy above 2 keV, as was calculated by Douthat [7] for He. The energy dependence of W_i was verified in many experiments using different types of charged particles, as summarized in the ICRU report [8]. However, the energy dependence of the Fano factor was not measured for charged particles due to the lack of accurate experimental tools. The effect of different shell ionizations on W_i and F was studied by Kowari et al. [9] in argon. It was shown that even though the electron degradation spectrum was modified by the existence of L-shell ionizations, the effect on W_i and F was negligible due to the low L-shell ionization probability.

Unlike charged-particles energy loss, the photoabsorption cross-section dramatically changes when the photon energy exceeds an atomic shell energy. This may lead to detectable changes in F and W_i . Samson and Haddad [10] have measured W_i of xenon using both electrons and photons in the energy range between the xenon ionization threshold upto 90 eV. They found that for electrons W_i smoothly decreases with the increase of electron energy, while for photons, W_i has a discontinuity at the xenon d-shell ionization energy. Using a gas scintillation proportional counter which is considered a reliable tool

for the study of the Fano factor, Kowalski et al. [11] have measured discontinuities in F above the xenon shell energies. This was later confirmed by a Monte-Carlo study done by Dias et al. [12, 13] and by additional experiments as well [14].

Contrary to xenon, hydrocarbons widely used in gaseous detectors have rarely been studied. This is due on one hand to the ignorance of the interaction cross-sections, and on the other hand to the lack of appropriate experimental tools. Most of the W_i and F measurements in hydrocarbons were conducted using high energy particles and provided only asymptotic values. Suzuki and Saito [15, 16] have measured relative mean energy per ion pair values for ethylene and propane in the x-ray energy range of 270-320 eV. They extracted the relative W_i values from pulse-height spectra and have shown that they oscillate above the C-K shell energy. A similar study was done by Srdoc [17] in propane and TE mixtures, showing a decrease of W_i with an increase of x-ray energy. Later on, Srdoc et al. [18] de-convoluted the ionization electron distributions from proportional counter pulse-height spectra, providing both F and W_i . This work was done for several hydrocarbons at three x-ray energies: 277, 1486 and 5890 eV; therefore it did not provide the full dependence of F and W_i on the x-ray energy.

We developed a unique experimental tool which permits the determination with great accuracy, in a nearly direct way, of the values of W_i and F in the ultrasoft-to-soft x-ray energy range. The method is applicable for all counting gases, including their mixtures as well as mixtures comprising noble gases. The method allows one to study the shell effects on W_i and F of hydrocarbons and low Z gases such as Ar and N_2 . In this article, we describe the method and present first results for C_2H_6 and Ar/ C_2H_6 (20:80).

2 The experimental method

Our method of extracting W_i and F combines an experimental measurement of the distribution of the number of ionization electrons deposited by an x-ray photon of a given energy in a gas, and a Monte-Carlo simulation of the detection process. The measurements are done with an Electron Counting (EC) detector, described in detail in refs. [19, 20]. Due to the non-linear electron counting efficiency [19] of the EC detector, the values of W_i and F cannot be determined directly. These parameters can be determined by simulating this inefficiency and analyzing the agreement between the experimental and simulated distributions of the number of counted electrons. This is done mathematically using a χ^2 test, varying the W_i and F parameters.

In the following section we will describe the experimental setup and the basic algorithm of the Monte-Carlo code.

2.1 The electron counting detector

A schematic view of the Electron Counting (EC) detector is shown in fig.1. It is composed of a 200 mm long radiation conversion and electron drift volume coupled to a double-stage parallel plate electron multiplication element. The active area of the detector is 50 cm².

The ionization electrons created by an absorbed x-ray photon, the number of which is proportional to the photon energy, slowly drift towards the amplification element. A low drift field is maintained along the conversion volume by two parallel meshes and equally spaced metal rings, connected to a resistor chain. Diffusion in the gas, enhanced by the low pressure operation and by the long drift path, causes the initial electron cloud to expand during the drift process. Therefore, the electrons arrive at the amplification element at different time intervals and are individually multiplied and counted. The single electron multiplication factor is higher than 10^6 , which guarantees high detection efficiency. Electron losses to the drift volume walls are maintained at a negligible level by controlling the transverse diffusion and by a proper collimation of the x-ray beam. Single electron avalanche-induced pulses are amplified using a fast (9 ns rise time), high gain current amplifier and are measured by a LeCroy 5100M digitizing oscilloscope. Each of the single photon-induced electron trails (a sequence of single electron pulses induced by one x-ray photon) is transferred to a PC at a rate of up to 40 Hz. The primary electron pulses are automatically recognized and counted using an off-line correlation method. The method is based on a software correlation between a single electron pulse-shape function and the digitized induced electron trail. The resulting correlation function contains a trail of bipolar spikes, corresponding to highly correlated electron pulses. This form allows an efficient automatic single electron recognition and counting. A detailed description of this method is given in ref. [21, 22]. An example of a digitized fluorine-K induced electron pulse trail and its correlated function is shown in fig.2.

2.2 The x-ray source

The detector vessel, kept under low pressure, is attached to the target chamber through a thin ($\approx 75\mu\text{g}/\text{cm}^2$) polypropylene foil. In a previous setup [20], we used a commercial α -induced x-ray source, providing us with a collimated x-ray photon beam of 15-100 photons/s, depending on the target material. This rate was comparable to the cosmic background rate, and since the detection was not triggered by the x-ray source, it was not possible to accurately extract F and W_i .

We are currently employing Particle Induced X-ray Emission (PIXE) technique, at our Van De-Graaf accelerator. The EC detector is attached to the target chamber in a backward direction (120°), in order to reduce the probability of direct illumination by back-scattered particles. Characteristic soft x-rays are induced by a 0.5-50 nA beam of 1 MeV α -particles. We used several targets providing the following lines: Be (110 eV), B (185

eV), Carbon (277 eV), LiF (54.7, 676 eV), Mg (1254 eV) and Al_2O_3 (525, 1486 eV). The detected x-ray photon rate was a few hundreds/s for all targets, which is sufficiently above the cosmic background rate.

Due to carbon build-up on the targets, thin (200 nm) Al and SiO filters were used to cut the C-K line. Yet, at this stage, the carbon build-up still prevents the measurements of the N-K and O-K lines.

2.3 The Monte-Carlo code

A Monte-Carlo program simulates all the processes occurring in the EC detector, starting with the absorption process of an x-ray in the drift volume up to the single electron counting and recognition. The program involves the following: x-ray absorption cross sections, primary ionization statistics, electron drift and diffusion, electron losses in the drift volume, amplification statistics, single electron pulse shape and correlation analysis parameters.

The simulation procedure is as follows:

The absorption depth of each photon is generated from an exponential distribution, with a proper energy-dependent attenuation length, taken from ref. [23]. A number of primary electrons is randomly generated from a Gaussian distribution, with an average number $\bar{n} = E_\gamma/W_i$ and a width $\sigma = (F \cdot \bar{n})^{0.5}$. E_γ is the x-ray photon energy. W_i and F are free parameters in the simulation procedure. The arrival time of each electron at the amplification element, after drifting along the conversion region, is randomly generated using the drift velocity and the longitudinal diffusion, carefully measured (see below) for each gas mixture studied. An electron pulse-trail (see definition in sec.2.1) is generated: The amplitude of each electron pulse is generated out of an experimental single electron pulse-height spectrum measured using the EC technique. The shape of individual electron pulses was derived from our experimental data; a random noise with a proper level was also added to the pulse-trail. The pulse-trail data are stored in a digital form, identical to that of the experimental data. Finally, the pulses in each simulated pulse-trail are recognized and counted by the correlation technique with the same parameters applied to the experimental data.

Typical examples of experimental and simulated 677 eV x-ray induced electron pulse-trails are shown in fig.2.

3 Experimental results

3.1 Measurements procedure

For each gas under study we recorded the x-ray-induced electron distribution at different x-ray energies, using different targets. In addition, for each gas we measured the single electron pulse-height spectrum and the electron drift velocity and diffusion. All these provided the parameters incorporated in the simulation program.

Typical distributions of the number of counted electrons measured at 20 Torr of Ar/C₂H₆ (20:80) for the two extreme x-ray energies: Be-K 110 eV and Al-K 1486 eV, are shown in fig.3. The Al-K spectrum was measured using a 200 nm Al filter foil which absorbed both the oxygen and carbon lines. The arrows indicate the expected average number of counted electrons for Be-K and Al-K lines, assuming 100% counting efficiency and W_i of 26 eV. They emphasize the known non-linear counting efficiency of the EC technique, dominated by the overlap of electron pulses at large electron densities [22, 19]. One can also notice the carbon line, around 9 electrons, due to carbon build-up on the Be target. An additional effect, also seen in fig.3, is the tailing of the Al line towards the low number of counted electrons. This is due to photons absorbed deep within the drift volume, resulting in a shorter drift path and therefore lower diffusion, causing a reduction of the counting efficiency. The combined effect of a large photon attenuation length (37 cm for the Al-K at 20 Torr of Ar/C₂H₆) and a large number of induced electrons (≈ 60) is well emphasized in a contour plot (fig.4) of the number of counted electrons versus the electron trail length: a tail of short electron trail length extending to the axis origin is seen for Al-K, but such an effect does not exist in the case of the C-K, also shown in fig.4.

Single electron pulse-height spectra needed as input for the simulation were measured using the EC technique by recording the pulse-height of each of the individually recognized electrons. The measurement was done using low energy x-rays, and applying an extremely low drift field to avoid pulse overlap. In these measurements, the pulse-height distributions do not directly contain the electronic noise and pedestal, but have a low cut-off due to the threshold in the correlation algorithm. The lower part of the pulse-height spectrum is therefore extrapolated by fitting it to a Polya function. An example of a single-electron pulse-height spectrum, measured in Ar/C₂H₆ (20:80) is given in fig.5 together with its Polya fit. The single electron pulse-height measurement method can be employed as an efficient tool to determine gas amplification parameters [24].

The electron drift velocity and longitudinal diffusion were measured using a pulsed H₂ lamp. UV-induced pulses originating from electrons photo-induced on the drift volume mesh electrodes were recorded. The electron drift velocity and diffusion were extracted from the pulse arrival time and its distribution. A detailed account of the technique is given elsewhere [25].

3.2 Data analysis, results and discussion

The variance of the distribution of the counted number of electrons contains both the intrinsic Fano fluctuations and fluctuations resulting from the EC technique. The present detector configuration with a 200 mm long drift volume is optimized for ultrasoft x-ray photons having a small attenuation length. Therefore, the fluctuations resulting from the EC technique are dominant for high energy photons as is clearly reflected in the tailed distribution of the Al-K x-ray photons in fig.3. In order to reduce the influence of these type of fluctuations, and to improve the sensitivity of the method for determining the Fano factor, we applied an event selection procedure: only electron trails originating from events subjected to large diffusion were selected for the χ^2 analysis. An example of such an event selection, performed on the experimental and on the simulated electron distributions, is shown in fig.6. It is important to note that there is a very good agreement between the experimental and simulated distributions, both for selected and non-selected events, and that this event selection is not for covering up a deficiency of the simulation, but to reduce the relative influence of experimental fluctuations and thus improve the sensitivity to intrinsic Fano fluctuations. Contour plots of χ^2 values comparing experimental and simulated distributions of the number of counted electrons are shown in fig.7 for Al-K at 20 Torr of Ar/C₂H₆ for the two cases. A global minimum is indeed observed here for the selected events, indicating the most probable F and W_i values. In fig.7a, where all events were considered in the χ^2 analysis, only the W_i , but not the F value could be determined, due to the very weak dependence of the χ^2 values on F. An event selection procedure was actually needed only for tailed distributions, namely for energetic photons. In order to verify that the selection of events does not introduce a bias on the extracted F and W_i , we varied the selection conditions over a large range; We have satisfied ourselves that the fluctuations of the derived F and W_i values due to event selection conditions were below 1% and 0.5%, respectively.

Our first results of F and W_i as a function of the photon energy, measured for C₂H₆ and Ar/C₂H₆ (20:80), are given in fig. 8. In both gases a decrease of F and W_i with increasing energy is well observed. Plotting the extracted F values as a function of that of W_i (fig.9), a clear correlation between them is observed. Such a correlation was also pointed out by Bronic [26]. An increase of W_i and F for the C-K line compared to the B-K line can be seen only in the Ar/C₂H₆ gas mixture. This increase is a result of the change in the electron degradation spectrum induced by ionization of the L₂ and L₃ edges of argon lying at 250.6 and 248.4 eV, respectively. The existence of a similar effect expected in C₂H₆ above the C-K 282.4 eV edge could not be observed due to lack of experimental data in this energy range. The observed increase of the F and W_i values at 185 eV (B-K energy) compared to that of 110 eV (Be-K), is so far unexplained.

A comparison between the results of Srdoc et al. [18] and ours for the C-K and Al-K lines in C₂H₆ is given in table I. The discrepancies between the results do not exceed 10%, and it should be noted that no discussion of the source and magnitude of the errors is

provided in their reference. In our experience, the error in measurements of single-electron pulse-height spectrum is rather large, especially in the low energy region. Srdoc's analysis relies on this spectrum, which may be the source of the discrepancies between their results and ours.

To our knowledge, only Hurst et al. [27] have measured W_i values in Ar/C₂H₆ gas mixtures, using 5.3 MeV α -particles. Their measured W_i values are 26.6 eV and 25.4 eV for C₂H₆ and Ar/C₂H₆, respectively. Our W_i values, at the highest x-ray energy (Al-K), are 24.9 and 24.4 eV for C₂H₆ and Ar/C₂H₆ (20:80), respectively. In both Hurst's and our measurements, the W_i value of Ar/C₂H₆ is lower than that of C₂H₆, due to a Penning effect described in that reference. However, the W_i values of Hurst et al. are higher than ours. This may be due to the fact that with α -particles W_i reaches its asymptotic value at energies near 10 MeV [8]. With photons, W_i reaches its asymptotic value for photon energies much larger than the nearest atomic shell energy which can be ionized, as in the case of Al-K photon and C₂H₆ and Ar/C₂H₆.

3.3 Error evaluation

Both experimental, statistical and analysis errors contribute to the overall error of F and W_i .

Experimental errors

The experimental errors are the errors involved in the electron counting technique and also the errors involved in the measurement of parameters employed in the simulation, such as electron drift velocity and diffusion, pulse-height spectra etc. The errors induced on W_i and F by the uncertainties of the above parameters increase with the x-ray energy, due to the decrease in electron counting efficiency. Therefore, these induced errors were evaluated for each energy by repeating the fitting procedure (χ^2 calculation) between the simulated and experimental distributions of counted electrons, and varying the parameter values within their own experimental uncertainties. We will present here the minimal and maximal errors for the two extreme x-ray energies investigated (110 and 1486 eV).

The simulation input parameters can be divided into three categories according to the importance of their influence on W_i and F determination:

- Pulse shape and pulse recognition algorithm (correlation) parameters. Their values are taken directly from the experimental data, without any errors.
- Photon attenuation lengths, for which the data are taken from ref. [23]. According to our calculations using the errors provided in ref. [23], the variations in the simulation results due to the error in the photon attenuation length are negligible ($\leq 0.1\%$).
- Single electron pulse-height spectra, electron losses due to attachment and electron transport parameters represented in the simulation by the electron drift velocity

and diffusion. Their influence on the results are discussed in detail below.

The mean values of the pulse-height spectra were measured with an accuracy of 4%. The induced error on F and W_i varies between 0.6% to 1.5% for Be and Al lines, respectively.

Electron drift velocities were measured with an accuracy of 1%, inducing an error on W_i and F of 0% to 0.6%. The electron diffusion coefficients were measured with an accuracy of 5%, inducing an error of 0% to 2% on W_i and F .

Electron losses in the drift volume were found to be below 1%. They were estimated by comparing the ratios of counting efficiencies of C-K distributions of counted number of electrons to their corresponding simulated distributions at 3 different drift velocities ranging from 0.5-1.5 cm/ μ s. The maximal induced errors on W_i and F resulting from possible electron losses are - 0.5%.

Statistical errors

The number of experimental and simulated events measured/simulated for each x-ray energy was between $6 \cdot 10^4$ to $1.4 \cdot 10^5$ for the two extreme energies, respectively. The statistical error of the mean and the standard deviation of the distributions of counted number of electrons is thus below 0.3%. The induced errors in F and W_i are $\approx 0.4\%$.

Analysis errors

These refer to fluctuations of F and W_i resulting from event selection criteria imposed on the data. We estimate the errors to be 1.5% and 0.5% for F and W_i , respectively.

To summarize, the uncertainties on the W_i values vary between 1.1% and 2.7%, while those on the F values vary between 1.7% to 3.1%, for photons in the respective energy range of 110-1486 eV.

4 Conclusions

We have described a new method for measuring the Fano factor and the mean energy per ion pair in counting gases, in the ultrasoft-to-soft x-ray energy range. It is applicable to hydrocarbons and other gas mixtures, for which no other accurate experimental or theoretical tools exist.

Measurements of F and W_i were done for several energies ranging from the Be-K 110 eV line to the Al-K 1486 eV line, in C_2H_6 and Ar/ C_2H_6 (20:80). A decrease in F and W_i with increasing x-ray energies was observed in both gases. An increase of F and W_i values above the argon L shell energy was observed in Ar/ C_2H_6 (20:80). Such a dependence of F and W_i on the gas shell levels was also observed by others in xenon [12, 13], while oscillations of the W_i values were described by Suzuki et al. [15, 16] for ethylene and propane. This effect is due to the change in the electron degradation spectrum following

the opening of a new channel of photoabsorption by an inner shell level. In addition, higher energy is required to release an Auger electron than the average energy required to release an outer shell electron. For example, the argon L_2 , L_3 energies are 250.7 and 248.6 eV while the main $L_{2,3}-M_x, M_y$ Auger energies lie between 190-210 eV [28], x, y are argon's M shell indices. Consequently 40-60 eV are used to release an Auger electron, compared to ≈ 27 eV which are required to further release electrons by a photoelectron impact. This leads directly to an increase in W_i , and modification of F.

Presently, we cannot analyze N-K and O-K data due to a dominant yield of C-K x-rays. In future measurements, we will reduce the rate of carbon build-up by using a cold finger (liquid nitrogen temperature) which will be placed at the vicinity of the targets. This hopefully will allow us to have two more data points at 392 eV and 525 eV.

At its present stage, using an electron multiplier with parallel grid amplification element, our method provides W_i and F determination with an accuracy of 3%. The error is mainly due to the large influence of the uncertainties of the simulation input parameters, especially at high x-ray energies, where the electron counting efficiency is low.

We are investigating a new electron multiplier, based on a microstrip amplification element [29], to improve our electron counting technique. First measurements, using a parallel plate device followed by a gas microstrip amplification element, provided single electron pulses having a FWHM of 10 ns, compared to 40-50 ns in our current setup, with an improvement of the signal-to-noise ratio by a factor of 10. With this new multiplication element, the electron counting efficiency approaches 100% at energies up to 277 eV, allowing a direct measurement of F and W_i .

In the future, we are planning to study F and W_i values in a large number of gases.

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Table I: A comparison of the Fano factor (F) and the mean energy per ion pair (W_i) values measured in C_2H_6 by Srdoc et al^a. and in this work.

	C-K 277 eV		Al-K 1486 eV	
	Our data	Results of Srdoc et al.	Our data	Results of Srdoc et al.
F	0.275 ± 0.005	0.296	0.250 ± 0.008	0.276
W_i	27.6 ± 0.3	25.87	24.9 ± 0.6	24.82

^aReference 18.

Figures caption.

1. A schematic layout of the Electron Counting detector.
2. Typical 676 eV, fluorine x-ray events: (a) a digitized experimental electron trail, (b) the corresponding correlated trail. (c) a simulated electron trail. Electron pulses recognized by the correlation technique (b) are circled in (a) and (c). The solid horizontal line in (b) is the threshold chosen for the data analysis. Ar/C₂H₆(20:80) at 20 Torr.
3. Distributions of the counted number of electrons, induced by Be-K and Al-K photons in Ar/C₂H₆(20:80) at 20 Torr. A C-K induced peak, around 8-9 electrons, is observed on the Be spectrum. The expected average numbers of counted electrons, assuming 100% efficiency and a W_i value of 26 eV, are indicated in the figure. Solid lines connecting the experimental points are added to guide the eye.
4. Contour plots of the counted number of electrons as a function of the pulse-trail length for Al-K (a) and C-K (b) x-rays. The window shown in (a) corresponds to selected events, employed for the determination of F and W_i .
5. A single electron pulse-height spectrum measured in Ar/C₂H₆(20:80) at 20 Torr. The solid line is a Polya fit to the spectrum.
6. Distributions of experimental (squares) and simulation (lines) counted number of electrons. (a) includes all events, while (b) includes only events within the 2D window shown in fig.4. Note that the simulation remarkably reproduces the experimental data in both cases.
7. Contour plot of χ^2 values, representing the degree of agreement between experimental and simulated distributions of counted electrons of Al-K measured in Ar/C₂H₆(20:80) at 20 Torr. (a) without event selection, (b) with events selected within the window shown in fig.4. The location of the minimal χ^2 value in (b) provides the values of F and W_i .
8. Extracted W_i and F values as function of x-ray energy for Ar/C₂H₆ (20:80) and C₂H₆.
9. The relation between the Fano factor and the mean energy per ion pair W_i in C₂H₆ and Ar/C₂H₆ gas mixture.

Fig.1

Fig.2

Fig.3

Fig.4

Fig.5

Fig.6

Fig.7

Ar/C₂H₆ (20:80)

C₂H₆

Fig.8

Fig.9