The Fano factor and the mean energy per ion pair in counting gases, at low x-ray energies.

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ABSTRACT

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The mean energy per ion-pair (W_i) and the Fano factor (F) are provided with high accuracy (2 and 3-4%, respectively), in C_2H_6 , C_3H_8 , $i-C_4H_{10}$, CH_4 , DME, Ar/ C_2H_6 (20:80), Ar/ $i-C_4H_{10}$ (20:80) Ar/DME (20:80) and Ar/Xe/ $i-C_4H_{10}$ (66.6/16.7 /16.7), in the x-ray energy range of 0.11 to 1.5 keV. These parameters were extracted from precise measurements of the number and temporal distribution of x-ray induced electrons, accompanied by extended simulations of the detection process. A decrease in these parameters with increasing x-ray energy was observed, accompanied by sharp increases at x-ray energies just above some atomic shells. The effect is discussed in relation to Auger electron emission. A Penning process in Ar/ C_2H_6 (20:80) and Ar/ $i-C_4H_{10}$ (20:80) is observed on the basis of comparative measurements of W_i and F in these mixtures and in the pure hydrocarbons. Ways are proposed for further improving the accuracy provided by the electron counting technique to better than 1%.

1 Introduction

The knowledge of the ionization produced by radiation in matter, is of basic importance in dosimetry and in radiation physics. It provides the link between the deposited energy and the resulting ionization electron yield, and permits the evaluation of the radiation induced damage. It also enables better optimization and conception of radiation detectors.

The stochastic ionization process is most commonly represented by two macroscopic parameters, the mean energy necessary for the creation of an electron-ion pair (W_i) in a gas (or an electron-hole pair in a solid), and the Fano factor (F) [1, 2], describing the ionization yield and its variance. They depend on the radiation type and energy, as well as

on the atomic structure of the absorption medium.

Most of the studies of F and W_i were done using charged particles, mainly electrons and α -particles, because of their close relation to dosimetry and the relative simplicity in the involved processes. It has been established [3, 4] that both W_i and F are nearly constant for energetic charged particles (for example for a few keV electrons or for a few MeV α -particles). This was explained by the independence of the electron degradation spectrum, describing the energy spectrum of both primary and secondary electrons in matter under stationary conditions, on the initial particle energy [5, 6]. For low energy particles, both F and W_i depend on the initial projectile energy as was calculated [6]- [10] and measured [11] for electrons in various gases. Kowari et al. [12] studied

the influence of the Ar L and M shells on F and W_i values. They have noticed that the inclusion of the Ar L shell ionization acts on both parameters, more significantly on F . The electron degradation spectrum is modified, though the overall influence on F and W_i is negligible due to the low Ar L shell ionization probability ($\approx 1\%$), up to a few keV electrons. In contrary to charged-particles, the photoabsorption cross-section dramatically varies when the photon energy exceeds that of an atomic shell and could lead also to significant changes in F and W_i . Samson and Haddad [13] measured W_i of xenon using both electrons and photons in the energy range between the xenon ionization threshold (12 eV) up to 90 eV. They found that while for electrons W_i smoothly decreases with the increase of electron energy, it has a discontinuity for photons at the xenon d-shell ionization energy. Using a gas scintillation proportional counter, which provides good energy resolution in noble gases only, Kowalski et al. [14] measured discontinuities in F above the xenon shell energies. Same effects were confirmed by Dias et al. [15, 16] in Monte-Carlo studies and additional experiments [17]. The discontinuities in F and W_i values following the Xe M and L shells, calculated by Dias et al., were in the order of 25-50% and 2-4% for F [15] and W_i [16], respectively. The considerably larger effect on F is in agreement with Kowari et al. [12] calculations for argon, using 2 keV electrons. The authors explain the increase in F and W_i values following the onset of Auger processes, by the change in the secondary ionization electron energy spectrum.

For hydrocarbons, the dependence of W_i and F values on the x-ray energy and on the shell structure of the gas atoms have rarely been studied with sufficient accuracy. Combecher [11] reported on a smooth decrease of W_i values in a large number of hydrocarbons with increasing electron energy from 5 to 500 eV. Suzuki and Saito [18, 19], using a gas proportional counter, observed sharp variations of W_i around the C K edge in ethylene and propane, using x-rays in a very limited energy range of 270 to 320 eV. Srdoc et al. [20, 21] and Bronic et al. [22, 23] extracted F and W_i values for several hydrocarbons at x-ray energies of 0.277, 1.48 and 5.9 keV. Their method, based on a deconvolution of the single electron distributions from x-ray spectra measured with a proportional counter, provided moderate accuracies in the order of 8% for F and 2% for W_i [24]. This and the limited number of x-ray lines, prevented a detailed evaluation of a possible dependence of F and W_i on the x-ray energy and

on the gas atomic shell structure.

We present here new accurate data on F and W_i for several hydrocarbons and some hydrocarbon mixtures with Ar and Xe. The parameters were determined with good accuracy using the technique of direct counting of x-ray induced ionization electrons deposited in low-pressure gas [25, 26, 27]. It should be noted that this technique is applicable for most counting gases provided reasonable gas gains can be reached. We confirm here the energy dependence of F and W_i , discuss the atomic shell effects on these parameters and confirm the Penning processes in Ar/C₂H₆ and Ar/i-C₄H₁₀. A comparison with other available x-ray data as well as with that of heavy charged particles is provided.

2 The experimental method

The method of extracting F and W_i combines measurements of the distributions of the number of ionization electrons deposited in a gas in proportion to the x-ray photon energy, and a Monte-Carlo simulation of the detection process [27]. The x-ray detector is a low-pressure (20 torr) electron counter (EC) described in detail in refs. [25, 26]. The EC technique has been also applied to measurements of specific primary ionization [28], soft x-ray spectroscopy [29] and microdosimetry [30]. Clusters of x-ray induced ionization electrons are expanded during a drift process in a 20 cm long gas volume at low-pressure, by an enhanced electron diffusion. Electrons are individually multiplied and the avalanche-induced pulses are counted. Photons of a given energy provide distributions of the number of the deposited electrons. The arrival time of each electron in the pulse-trail from each single photon event is recorded. The correlation between the number of counted electrons and the electron trail length provides an additional means of improving the x-ray line analysis as discussed in detail in ref. [27]. A detailed Monte-Carlo simulation taking into account all processes contributing to the x-ray interaction and electron transport and counting, allows to accurately predict the detector response and provides the electron distributions, in number and time, similar to the experimental data. F and W_i are extracted from the degree of agreement (χ^2 test) between the experimental results and the simulation calculations in which F and W_i are varied as free parameters. The errors are evaluated by varying the simulation input parameters, measured in the course of this work, within their ex-

perimental inaccuracies. A detailed description of the technique is given in our previous publication [27].

X-ray photons were generated using particle-induced x-ray emission (PIXE) technique at the Weizmann Inst. Van de-Graaf accelerator. A collimated beam of 1 MEV α -particles impinging on various low- z targets, induced characteristic x-rays at yields of a few hundred Hz, well above cosmic background (≈ 7 Hz). The low rate was kept to prevent photon event pile-up and was compatible with the readout system.

Ideally, one would prefer using targets providing monoenergetic x-rays, since contributions from other neighboring lines, or background radiation, distort the electron distributions, affecting the extracted values of F and W_i . The absence of pure low- z solid elements, such as nitrogen and oxygen, obliged us to use mixed targets such Al_2O_3 , MgO , teflon or LiF which provided carbon (277 eV), fluorine (676 eV) magnesium (1253 eV), aluminum (1486 eV) and oxygen (525 eV) x-ray lines. Pure elemental targets used were beryllium (108.5 eV), boron (183.5 eV) and carbon (277 eV). Because of the relatively poor vacuum in the target chamber (10^{-6} torr), carbon build-up on various targets occurred, adding carbon contaminant to the Be, LiF , MgO and alumina spectra. Home-made x-ray filters of 200 nm thick SiO or Al evaporated on $75\mu\text{g}/\text{cm}^2$ polypropylene foil, were used to attenuate the carbon and oxygen lines, when required. Typical SiO filter transmission efficiencies are 10^{-8} , 0.7, 27.3 and 44% for C K, O K, Mg K and Al K x-rays, respectively. A dedicated analysis technique, uniquely possible when using the EC method, enabled to isolate the fluorine and beryllium lines from the carbon line in the electron distributions [27, 29]. A demonstration of improving x-ray line resolution is given in fig.4 of ref. [27]. Attempts were made to measure the nitrogen (392 eV) line using a boron-nitride (BN) target. However, the relatively low yield of characteristic nitrogen photons as well as the carbon build-up on the target surface, prevented the sufficient resolution of this line. With the PIXE technique, we could not measure low-energy x-ray lines originating from higher shell fluorescence of high- z elements. This is due to the domination of the low-shell fluorescence yield over the higher shell one [31], affecting the stability of operation of the EC detector due to the relatively high charge induced by energetic (2 keV and above) photons.

In all experiments, the detector was operated in a flow mode using gases with purity of: argon, xenon, C_2H_6 :99.995%, $(\text{CH}_3)_2\text{O}$, C_3H_8 and $i\text{-C}_4\text{H}_{10}$: 99.9%

and CH_4 :99.999%.

3 Results

F and W_i values as function of x-ray energy were extracted in hydrocarbons: C_2H_6 , C_3H_8 , $i\text{-C}_4\text{H}_{10}$, CH_4 , $(\text{CH}_3)_2\text{O}$ (DME), in mixtures with 20% of argon: $\text{Ar}/\text{C}_2\text{H}_6$, $\text{Ar}/i\text{-C}_4\text{H}_{10}$, Ar/DME , and in a mixture containing $\text{Ar}/\text{Xe}/i\text{-C}_4\text{H}_{10}$ (66.6 /16.7 /16.7). The addition of argon permits the study of possible Penning processes, discussed elsewhere [32, 38] and the effect of Ar $L_{II,III}$ shells on F and W_i values. Forseeing the application of the EC technique to x-ray spectroscopy, we have measured F and W_i values in xenon mixture. The addition of xenon reduces the x-ray absorption length, and thus improving both the x-ray detection efficiency and the energy response of the EC detector.

The results of the present measurements are summarized in table I and in figs.1-3; the table also contains the data of C_2H_6 and $\text{Ar}/\text{C}_2\text{H}_6$ previously published in ref. [27]. A general decrease of F and W_i with increasing x-ray energy is observed, in accordance with theory. This decrease is accompanied by differential increases of both parameters for x-ray energies just above gas atomic shell levels, such as Ar $L_{II,III}$, O K and Xe M $_V$. In some of the Ar mixtures, the measured F and W_i values are lower than the pure hydrocarbon values. More details are given below.

Figure 1 presents the data for all pure gases measured, C_3H_8 , CH_4 , $i\text{-C}_4\text{H}_{10}$ and DME. Srdoc [21] results and ICRU [3] mean values for charged particles are provided when available. Except for DME, for all pure gases a decrease of both parameters with increasing x-ray energy is observed. For DME, in addition to above trend, a larger F value at 676 eV (lying above the O K shell energy) was measured compared to the 277 eV value. Such effect is less significant for W_i . Except for the W_i data in $i\text{-C}_4\text{H}_{10}$, our F and W_i values are in good agreement with both Srdoc [21] and ICRU [3].

Fig.2 presents the data measured with argon mixtures. The data of the pure gases are provided for comparison, without the experimental error bars. In the $\text{Ar}/i\text{-C}_4\text{H}_{10}$ mixture the F and W_i values measured at 277 eV, lying above the Ar $L_{II,III}$ shell energies (≈ 250 eV), are larger than that at 183.5 eV. This phenomenon was not observed for the F values in pure $i\text{-C}_4\text{H}_{10}$. In addition, the W_i and F values in $\text{Ar}/i\text{-C}_4\text{H}_{10}$ are smaller than those in pure $i\text{-C}_4\text{H}_{10}$.

In the Ar/DME mixture a few jumps are observed: W_i and F values at 277 eV, lying above the Ar $L_{II,III}$ shells, are larger than the values at 183.5 eV. A similar effect following the O K shell photoionization is observed as well, indicated by an F value at 676 eV larger than at 525 eV. The measured W_i values in Ar/DME are below that in DME, while in contrary to other mixtures, the F values are not lower.

In Ar/Xe/i-C₄H₁₀, a considerable increase in F is observed at 676 eV, lying just above the Xe M_V shell energy (676.8 eV); there is a less significant increase in the value of W_i at this energy.

4 Discussion

We have identified several effects related to the energy dependence of F and W_i , in particular effects due to the atomic shell structure of the absorbing gas.

An increase of F and W_i values following the onset of inner shell photoionization is observed in gases containing argon, xenon and oxygen. This phenomenon, intensively studied in xenon [13]–[17] is related to the change in the primary and secondary electron spectrum following the onset of Auger process. For the argon mixtures used in this work, according to the photoionization data provided by ref. [35], approximately 80% of the photoionization by 277 eV x-rays occurs in the Ar $L_{II,III}$ shells (≈ 250 eV). According to Bergmark et al. [36, 37], the Ar $L_{II,III}MM$ Auger spectrum has multiple sharp peaks between 170 and 230 eV; the intense Ar $L_{II,III}$ Auger lines have energies between 190–210 eV. Therefore when an Auger electron is emitted following an L-shell photoionization, an energy of about 40–60 eV is invested in its release, compared to an energy of 26 eV (an average W_i value for most gases) required to release an ionization electron by an energetic electron impact. In the case of photoionization by a 277 eV photon, the photoelectron, released with an energy of ≈ 29 eV has a low probability of further ionizing the gas due to the relatively large value of W_i [11]. Therefore, the W_i values at x-ray energies above the Ar L shells increase, reflecting the less efficient ionization process. The F value may increase due to the additional energy dissipation pathways, added by the new electron “sources”, namely the Auger electron and the photoelectron. The influence of the Auger process is most significant for x-ray energies close to the newly opened atomic shell energy, and diminishes when the photon energy is much larger [16]. This phenomenon

is most clearly evident in our Ar/Xe/i-C₄H₁₀ mixture in which the F K x-rays have an energy only 0.4 eV above the Xe M_V shell energy.

We did not observe an effect of the C K shell (≈ 290 eV) photoionization in the pure hydrocarbons investigated. This may be either due to the lack of experimental data with x-ray lines at close enough energies (300–400 eV), or to the less significant influence of the C K Auger process on F and W_i . In general, the Fano factor was found to be more sensitive to the initiation of an Auger process than the mean energy per ion-pair, as was calculated both for argon [12] and for xenon [15, 16].

At the largest x-ray energy investigated, for which the shell effect is negligible (see below), the W_i values of Ar/i-C₄H₁₀ and Ar/C₂H₆ are lower than the values of the corresponding hydrocarbons and of argon (26.4 eV [3]). This could indicate the existence of a Penning process, in which the de-excitation of excited argon atoms results in further ionization of the hydrocarbons. This effect leads to an efficient ionization process, resulting in smaller W_i and F values, as emphasized for example in the case of Ar/C₂H₂ (99.5/0.5) [32]. Our W_i values measured in C₂H₆ and Ar/C₂H₆(20:80) are in agreement with Hurst [38] as discussed in ref. [27].

In some of the measurements we found that the W_i and F values measured with Be K photons (108.5 eV) were lower than those measured with B K photons (183.5 eV). This phenomenon, unsupported by any other measurement or calculation known to us, could indicate a new effect or may result from a deviation of the electron distribution from the assumed gaussian shape [21] in the Monte-Carlo simulation.

In fig.3 we present the correlation between F and W_i/I_p for all gases investigated, following the approach of Bronic [39]. I_p values used [40] are summarized in table II. W_i/I_p represents the degree of inefficiency of transfer of the deposited energy into ionization, namely $W_i/I_p=1$ means a fully efficient transfer while $W_i/I_p=2$ means that only 50% of the energy goes to ionization and the rest is lost in excitations. Bronic et al. found a linear relation: $F=(0.188 \cdot W_i/I_p)-0.15$, shown in fig.3. We have found that our data are grouped into two groups (indicated by solid and hollow symbols): the gas mixture points lay above Bronic’s line, while the pure gas points are situated below. This fructication of the data could be explained by over estimated I_p values used for the mixtures, calculated using the gas constituents volume

concentration only. A more suitable procedure for estimating I_p values of mixtures should probably take into account other effects, e.g. Penning processes. This would reduce the I_p values and shift the mixture points towards the line of Bronic et al. Except for DME and Ar/DME and within the experimental errors, our data are in a general agreement with Bronic's relation and have similar spread as the data in ref. [39]. The dependence of F and W_i on shell effects is most significantly manifested for DME, Ar/DME and Ar/Xe/i-C₄H₁₀, in which F values increase

sharply but that of W_i show small change, as discussed above. The larger shift of the DME points may be due to an overestimated W_i value, that could have resulted from possible electron losses. Such losses, due to electronegative impurities in DME, have been reported in the past [41] but were not evaluated for DME in the present work.

5 Conclusion

The electron counting technique was proved to be very adequate for deriving the Fano factor and the mean energy per ion-pair creation in counting gases, other than noble ones, in the low-energy x-ray range. In this work we presented results of the energy dependence of F and W_i in several hydrocarbons and their mixtures with Ar and Xe, in the x-ray energy range of 0.1 to 1.5 keV. The influence of various atomic shells, such as Ar L_{II,III}, O K and Xe M_V, on F and W_i values was shown. The general trend, namely the decrease of F and W_i with increasing x-ray energy, is in agreement with that predicted by theory [3, 4], as well as with other experimental data derived from less accurate measurements.

The general decrease of F and W_i with energy is expected from the nature of the involved processes. At low x-ray energies, involving the production of low energy photo- and Auger electrons, the weight of each ionization channel and interaction step is significant. The opening of a new shell for ionization, therefore, gives rise to a significant increase in both parameters, as explained in details in section 4 for the case of Ar L. At larger x-ray energies, the shell effects are completely masked by the large number of energy transfer steps involved. We may conclude that soft x-rays are the only possible probe for studying the dependence of F and W_i on atomic shell details.

At its current status, the accuracy of the method

of 2-4%, though superior to that of other techniques, is not sufficient to allow detailed quantitative analysis of the observed effects. The variations of F and W_i are in the order of 1-4%, and therefore a sub-percent accuracy is required. In addition, a finer scanning in energy is needed, especially in the 300 to 500 eV range, in order to provide the details of the variation of these parameters. A synchrotron x-ray source would be the ideal tool for this task. Alternatively, a tunable electron beam, such as in electron microscope could be used, tuning the beam energy according to atomic shell energies of the target, thus producing selected x-ray lines. An attempt in this direction was already made, and indeed, electron counting experiments at a SEM yielded very good results, also in terms of low background [29].

The accuracy of the obtained F and W_i values depends mainly on their derivation process. Currently, one of the main limiting factors is the detector inefficiency caused by some overlap of single electron pulses. This significantly reduces the number of counted electrons for photons above 500 eV [25], causing non linearities in the detector response and necessitates corrections which affect the accuracy. We are currently studying the performance of two new very fast, high gain electron multipliers, the microstrip [42, 43] and the microdot [44, 45] gas chambers. The single electron avalanche pulse-width is reduced in these multipliers, from the current 40 ns to 2-5 ns FWHM. This should dramatically improve the electron counting efficiency, reducing the dependence of the results on the Monte-Carlo simulation, and thus improving the method accuracy. We believe that an improved low-pressure electron counting technique may provide an excellent tool for further studies of processes related to low amounts of radiation-induced energy deposited in gases, applicable in dosimetry and many other fields of research.

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Table I: F and W_i values derived at few x-ray energies in several gases and mixtures (error in brackets)

		x-ray energy [eV]						
gases		108.5	183.5	277	525	676	1253	1486
C ₂ H ₆	W _i	27.4(6)	28.0(4)	27.6(3)		27.2(5)		24.9(6)
	F	0.270(7)	0.280(7)	0.278(7)		0.260(10)		0.250(10)
Ar/C ₂ H ₆ (20:80)	W _i	26.8(6)	27.1(6)	28.0(3)		26.5(5)	25.1(6)	24.4(6)
	F	0.276(6)	0.255(6)	0.270(5)		0.260(6)	0.250(10)	0.240(10)
C ₄ H ₁₀	W _i	28.0(4)	27.4(3)	27.9(3)		27.0(3)	25.6(3)	
	F	0.300(10)	0.285(10)	0.280(6)		0.265(10)	0.255(10)	
Ar/C ₄ H ₁₀ (20:80)	W _i	25.5(6)	26.3(4)	26.9(3)		26.4(5)	25.0(6)	
	F	0.275(15)	0.253(6)	0.265(7)		0.250(10)	0.250(10)	
DME (CH ₃) ₂ O	W _i	28.6(5)	28.5(4)	28.6(3)		28.4(4)	27.7(4)	
	F	0.340(10)	0.320(10)	0.315(10)		0.330(10)	0.285(10)	
Ar/DME (20:80)	W _i	26.8(5)	26.9(3)	27.7(3)	26.8(5)	26.4(3)	26.8(5)	
	F	0.340(12)	0.300(10)	0.310(5)	0.300(12)	0.330(10)	0.315(12)	
C ₃ H ₈	W _i	27.7(4)	27.6(3)	27.2(3)		26.9(3)	26.2(3)	
	F	0.305(10)	0.290(10)	0.275(6)		0.270(10)	0.260(10)	
CH ₄	W _i	28.5(7)	29.7(9)	29.3(8)	29.2(8)	29.0(8)		
	F		0.285(10)	0.275(10)	0.270(10)	0.265(6)		
C ₄ H ₁₀ /Ar/Xe (66.6/16.7/16.7)	W _i		26.8(3)	26.4(3)		26.9(3)	26.0(8)	
	F		0.270(7)	0.260(5)		0.310(10)	0.270(10)	

Table II: Values of I_p ^a for gases investigated. For the mixtures the I_p values were calculated according to the volume concentration of the gas constituents, ignoring Penning effects.

gas	I_p [eV]	gas	I_p [eV]
CH_4	12.5		
C_2H_6	11.52±0.01	Ar/ C_2H_6 (20:80)	12.368
C_3H_8	10.95±0.05		
i- C_4H_{10}	11.57	Ar/i- C_4H_{10} (20:80)	12.41
(CH_3) ₂ O-DME	10.02±0.025	Ar/DME(20:80)	11.16
Ar	15.75	Ar/Xe/i- C_4H_{10}	12.14
Xe	12.13		

^a reference [40].

Figure 1: Extracted W_i and F values as a function of x-ray energy for various hydrocarbons. Solid lines are the ICRU [3] values, dashed lines mark the energy of the O K shell (≈ 525 eV).

Figure 2: Extracted W_i and F values as a function of x-ray energy for argon and argon/xenon mixtures with hydrocarbons. Dotted lines indicate the energies of the Ar $L_{II,III}$ (≈ 250 eV), O K (525 eV) and Xe M_V (676.4 eV) shells.

Figure 3: The relationship between the Fano factor and the ratio W_i / I_p in several gases. Typical errors are given for Ar/C₂H₆ only (see table 1). The linear relation suggested by Bronic [39] is indicated. I_p values are given in table 2. For gas mixtures the I_p values were calculated according to the volume concentrations.

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