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RELATIVE DETERMINATION OF W-VALUES FOR ALPHA PARTICLES IN TISSUE EQUIVALENT AND OTHER GASES

GARY L. KRIEGER

June 1976

SAFETY AND ENVIRONMENTAL PROTECTION DIVISION

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RELATIVE DETERMINATION OF W-VALUES
FOR ALPHA PARTICLES IN TISSUE
EQUIVALENT AND OTHER GASES

Gary L. Krieger

Abstract

W (the average energy to form an ion pair) for 5.4 MeV ^{241}Am alpha particles in a Rossi-type tissue equivalent (T.E.) gas, argon and methane was determined to an accuracy better than 0.2% using a new automated data handling system. A vibrating reed electrometer and current digitizer were used to measure the current produced by completely stopping the alpha particles in a large cylindrical ionization chamber. A multichannel analyzer, operating in a slow multiscaler mode, was used to store pulses from the current digitizer. The dwell time, on the order of 60 minutes per channel, was selected with an external timer gate. Current measurements were made at reduced pressures (~ 200 Torr) to reduce ion-recombination. The average current, over many repeated measurements, was compared to the current produced in nitrogen and its previously published W-value of 36.39 ± 0.04 eV/ion pair. The resulting W-values were (in eV/ion pair): 26.29 ± 0.03 for argon, 29.08 ± 0.03 for methane and 30.72 ± 0.04 for T.E. gas, which had an analyzed composition of 64.6% methane, 32.4% CO_2 , and 2.7% nitrogen. Although the methane and argon values agree within 0.1% with previously published values, the value for T.E. is 1.2% lower than the single previously reported value.

I. Introduction:

The objective of this research was to determine within a 0.2% accuracy the W-value (average energy required to form an ion pair) for 5.4 MeV, ^{241}Am alpha particles in a Rossi-type¹ tissue equivalent (T.E.) gas, methane and argon. A comparison method was used to calculate W to nitrogen and its associated W-value as the reference.

W is widely used in radiation dosimetry to calculate the energy deposited in gas filled detectors. Experiments in microdosimetry using ionization chambers employ T.E. gases so that the energy deposited in tissue can be more easily inferred. Therefore, any error associated with the W-value will result in a proportional error in the energy loss calculation.

Measurements of total ionization in gases, to which W is related, have been made since the earliest studies of radioactivity. One of the first to make these measurements was Rutherford² at McGill University, Montreal in 1899. Working with Thomson,³ he developed the theory that, "...rays in passing through the gas produce positive and negatively charged particles..." The term ion, which was derived from an analogy with electrolytic conduction, was applied to these charged particles.

Following this development, Rutherford went on to measure the total ionization produced in various gases from "uranium radiation" relative to that produced in air. The term ion pair was adopted at a later date to describe the process of completely removing an electron from the atom and forming two separate entities.

Considerable work was done to make accurate measurements of W during the 1950's and 1960's. Jesse,⁴ at St. Procopious College, Illinois, measured W-values for a number of gases using absolute and relative methods. In these studies, current formed by individual alpha particles,

from a ^{210}Po source, in a cylindrical ionization chamber operated at low pressures, was measured using a vibrating reed electrometer which was connected to a strip chart recorder. By measuring the magnitude of many pulses and comparing them to a calibrated pulse, he was capable of determining W with a 0.2% error.

Another major group, Bay, et al.⁵ at the National Bureau of Standards, made absolute measurements of W for various gases with a different technique than that used by Jesse. Ionization current from a ^{210}Po alpha source was measured using a vibrating reed electrometer. Their apparatus also simultaneously counted the number of alpha particles producing that current. By knowing the overall capacitance on the chamber, they were able to determine W with the same accuracy as Jesse. The results published by the two investigators were in excellent agreement.

Measurements were also made by Bortner and Hurst⁶ at Oak Ridge National Laboratory. The charge, produced by ^{239}Pu alpha particles in a parallel plate ionization chamber, was measured by determining the voltage change on an accurately calibrated capacitor in series with a potentiometer and an electrometer as a null detector. They used highly purified gases in their experiments and examined the effects of impurities on various gases. A discussion on the effects of impurities on gases will be described in the Theory section.

Kemmochi⁷ reported W for alpha particles in Rossi-type T.E. gas as 31.1 ± 0.1 eV/ion pair. His procedure was a modification of Jesse's method. Instead of a strip chart recorder, he used a peak holder and a multichannel analyzer to store pulses. He compared the mean value of pulse height spectra from alpha particles in each gas, using a collimated ^{241}Am source, with nitrogen as the reference gas.

A recent survey on W-values for alpha particles in a variety of gases was made by Myer⁸. Although his tabulation is extensive, no W-value for alpha particles in Rossi T.E. gas is presented. Table 1 lists previously published W-values for alpha particles in the gases examined in this research. The weighted mean is a composite value calculated by Whyte⁹ from the results of the listed experimenters.

In the research described here, different procedures were employed than those used by the above-mentioned scientists to measure W and the associated error for alpha particles in a T.E. gas. A comparison of the determinations made in this research with the accepted published W-values for argon and methane establishes a level of confidence used to estimate the accuracy of the W-value determined for T.E. gas.

Table 1. W-VALUES (eV/ion pair) FOR ALPHA PARTICLES IN VARIOUS GASES

<u>Researcher</u>		<u>N₂</u>	<u>CH₄</u>	<u>Argon</u>	<u>Rossi T.E.</u>
Valentine & Curran	(10)	36.0±0.7	29.0±0.6	25.9±0.5	-
Jesse & Sadauskis	(11)	36.6	29.2	26.4	-
Sharpe	(12)	36.4±0.4	29.1±0.4	26.3±0.3	-
Haeberli, et al.	(13)	36.3±0.15	-	26.25±0.12	-
Biber, et al.	(14)	36.5±0.15	29.0±0.15	-	-
Bortner & Hurst	(6)	36.3±0.4	29.4±0.3	26.4±0.3	-
Jesse	(4)	36.39±0.07	-	-	-
Bay, et al.	(5)	36.38±0.07	-	-	-
Genin	(15)	36.6±0.5	-	26.5±0.3	-
Kemmochi	(7)	-	-	-	31.1±0.1
Weighted Mean	(9)	36.39±0.04	29.1±0.1	26.3±0.1	31.1±0.1

II. Theory:

A. Ionization and Excitation

When an ionizing particle passes through a gas, part of its energy is lost in ionizing the gas. The rest is consumed in non-ionizing processes (e.g., excitation). Because energy is lost in non-ionizing processes, the energy required to form an ion pair will be greater than the ionization potential for a particular gas. The energy expenditure to form an ion pair varies from one atom to the next and for most cases the number of ion pairs is large, so that an average value is useful. The parameter W is defined¹⁶ as the average energy spent to form an ion pair.

$$W = \frac{E}{N_i} \quad (1)$$

Where N_i is the number of ion pairs formed when the energy, E , of the particle is completely absorbed.

Platzman¹⁷ developed the energy-balance equation, which described the processes involved in energy loss, for the case of helium:

$$E = N_i \bar{E}_i + N_{ex} \bar{E}_{ex} + N_i \bar{\epsilon} \quad (2)$$

where

E = total charged particle energy

N_i = number of ion pairs

$\bar{E}_i$ = average energy expended to form an ionized atom

N_{ex} = number of excited atoms produced

$\bar{E}_{ex}$ = average energy expended to cause excited atoms

$\bar{\epsilon}$ = average kinetic energy of sub-excitation electron

Equations 1 and 2 when combined define W as:

$$W = \frac{E}{N_i} = \bar{E}_i + \left(\frac{N_{ex}}{N_i} \right) \bar{E}_{ex} + \bar{\epsilon} \quad (3)$$

Platzman suggested that the magnitude of W be evaluated relative to the ionization potential I. He found, for the noble gases, W/I is close to 1.7, whereas for molecular gases including hydrogen W/I lies between 2.1 and 2.6. The ratios are higher for molecular gases because energy is transferred to molecular potential energy and the higher probability of multiple electron transitions.

B. Gas Impurities

There are two known processes by which impurities cause a decrease in the measured W-value.

(1) The Jesse effect (ionization of impurities by excited atoms). Jesse and Sadauskis¹¹ found that small amounts of argon (0.1 mole %) in helium and neon decreased the measured W-value by 30%. This effect is caused by an energy transfer from the excited atoms to argon, and because the ionization potential is lower for argon than for neon or helium, more ionization is produced.

(2) Ionization of impurities by sub-excitation electrons. Electrons of impurities with lower binding energy than the lowest possible excitation potential of the parent gas, can be ionized.

C. Gas Mixtures

Many types of ionization chambers currently in use employ a mixture of two or more gases to achieve the desired operating characteristics. Bortner and Hurst⁶ investigated the effect on W for a mixture of gases. They found the W-value for a mixture of two gases is given by:

$$\frac{I}{W} = \left(\frac{I}{W_1} - \frac{I}{W_2} \right) Z + \frac{1}{W_2} \quad (4)$$

where $Z = p_1/(p_1 + ap_2)$, p_1 , p_2 = partial pressures of the two gases in the mixture and a is an empirically determined constant for each mixture.

In 1963, the formula for the composite W-value was modified slightly by Strickler¹⁸ for three gases as follows:

$$W_{ij} = (W_i - W_j)Z''_{ij} + W_j \quad (5)$$

where

$$Z''_{ij} = p_i / (p_i + f_{ij}p_j)$$

p_i, p_j = partial pressures of the gases i and j respectively;

f_{ij} is also an empirically determined constant.

Strickler assumed that,

$$f_{ij} \approx \frac{f_{kj}}{f_{ki}} \quad (6)$$

where i , j and k refer to any three gases.

Kemmochi⁷ expanded equation (5) to a mixture of more than two gases.

$$W_{1,2,\dots,n} = \frac{\sum_{i=1}^n p_i f_i W_i}{\sum_{i=1}^n p_i f_i} \quad (7)$$

By comparing the calculated and measured W-values for a variety of gas mixtures he demonstrated the validity of equation (7). If the presence of a secondary gas does not contribute ionization to the primary gas, then equation (7) can be employed to find the effective W of the mixture.⁶

D. Recombination

When a charged particle passes through a gas, positive and negative ions are formed. After a period of time, in the absence of an electrical field, the ions will recombine. Recombination is the process by which ions of opposite charge neutralize themselves. Thomson³ at the Cavendish Laboratory was the first to study the loss of ions by recombination. Since that time, various aspects of recombination have been investigated under a variety of conditions. A thorough reference to the various types of recombination under different conditions is given by Loeb.¹⁹

Under the operating conditions of this experiment two types of recombination, columnar and volume, were dominant.

(1) Columnar Recombination.

Bragg and Kleeman²⁰ in their experiments on the range and ionization of alpha particles, noticed that it was more difficult than predicted to achieve saturation current in their ionization chamber. Their explanation, although erroneous, was based on the assumption of an initial recombination effect, whereby ions originating from the same molecules reunite. Moulin²¹ demonstrated that what Bragg and Kleeman observed was not initial recombination, but the result of ions belonging to the same 'column' (alpha particle track) recombining. He also suggested that the ions were not distributed uniformly. Around the same time, Jaffe²² developed the theory of columnar ionization and recombination. He suggested that ionization of a gas by an alpha particle occurs in a restricted region (column) along the alpha particle's path. He assumed that, in the initial ionization along the path, the ion distribution is very dense and declines radially in a Gaussian distribution from the

axis of the column. Columnar recombination is defined as the recombination that occurs between the ions distributed within the same column. Under normal circumstances the true saturation current cannot be measured even for very large fields. Before the ions can be drawn out of the column, some will diffuse and be lost by recombination.

(2) Volume Recombination.

Basically, volume recombination occurs between the ions of different tracks. A theory was developed by Thomson that would describe the total saturation curve. He developed differential equations describing the motion of ions under the influence of an electric field. Thomson³ reasoned that the number of ions, n , that are lost by recombination per unit time is proportional to the square of the number present in a given volume.

$$\frac{dn}{dt} = -\alpha n^2 \quad (8)$$

where α is defined as the recombination coefficient. Many theoreticians and experimenters have improved upon Thomson's initial differential equations. A complete history of the development of these equations can be found in the report by Tate and East.²³

In order to determine the amount of recombination, the ratio of measured current, I , to saturation current (expected for an infinite field) I_∞ is employed. The fractional recombination is therefore:

$$\Delta = 1 - \frac{I}{I_\infty} \quad (9)$$

To find current produced for an infinite field, a method similar to Zanstra's,²⁴ first published by Hess,²⁵ is employed. The procedure is described in the section on experimental methods.

III. Experimental Methods:

A. Apparatus

The ionization chamber used in this experiment was originally designed by Baum²⁶ for LET_r studies of heavy ion beams at reduced pressures. The physical construction of the chamber was described in detail by Varma and Baum.²⁷ For the purpose of this research, the chamber was modified slightly to measure W for alpha particles in a variety of gases. It consisted of a cylindrical aluminum ionization chamber, 1 meter long with a diameter of 30 centimeters (see Fig. 1). Parallel to the central axis of the chamber was a segmented cylindrical brass collecting electrode. The electrode contained 6 segments, each 15 cm long, supported on a ground electrode. Selection of the desired segments was made by magnetically operated reed switches (Hamlin, Lake Mills, Wisconsin). This permitted selection of any one or a combination of up to 6 segments to collect ions along the particle track. Batteries (two Everready 300V) were used to supply the high voltage to the chamber. Batteries were chosen because of their high stability and their low noise characteristics.

A description of the experimental design is found in Fig. 2. Ionization current resulting from collection of ions within the chamber was measured with the aid of a vibrating reed electrometer (Victoreen Model 475A) over the range of 10^{-15} to 10^{-7} amperes. It had a low input capacitance (25 picofarads) and a high input impedance (10^{10} ohm). The amplified current was converted to pulses using a current digitizer (Ortec Model 439). This instrument produced square wave pulses of +5 volts, 500 nanoseconds wide corresponding to each 10^{-6} coulombs of charge collected. The pulses were stored in a multichannel analyzer

Figure 1. Schematic of ionization chamber.

Figure 2. Experimental apparatus.

(TMC Model 404) operated in a multiscaler mode. Pulses were stored in one channel for a selected length of time, then a channel advance pulse from the dual timer/gate (Canberra Model 1776) signals the analyzer to advance one channel. The timing interval, usually 60 minutes, was controlled by a crystal time base clock which was accurate to 0.0005% over the operating temperature range of 0 to 50°C. Finally, the number of pulses stored in the multichannel analyzer was printed out with a paper type printer (TMC Model 500).

The chamber was evacuated overnight to 5 millitorr and was filled with the gas under investigation and pumped down to 30 millitorr five times before each measurement. This process was followed to reduce any possible contamination from the insulators and walls of the chamber.

To have maximum alpha particle range and a minimum of ion recombination, gas pressure was adjusted to cause the range to be slightly less than 15 cm. This was to insure that no energy was lost to the walls of the chamber. This range limitation of the alphas was insured by making certain that no current was produced in the second 15 cm electrode segment above the background current.

The gases examined were (1) ultra pure nitrogen (99.996%), (2) methane (99.0%), (3) argon (99.998%), (4) tissue equivalent gas* (64.4% methane, 32.4% CO₂ and 3.2% N₂). The actual partial pressures for T.E. gas found in this study are given on page 36. Gas samples were analyzed for impurities by the Brookhaven National Laboratory Analytical Services Section using mass spectroscopy.

Nitrogen was selected as the reference gas for various reasons.

* Specified by Matheson Gas Products, E. Rutherford, N.J.

Nitrogen has a low recombination coefficient which makes saturation easily attainable, and the W-value for nitrogen is widely accepted as 36.39 ± 0.04 eV/ion pair.⁹ The nitrogen used was of ultra high purity grade. According to the analysis at Brookhaven National Laboratory (BNL) impurities accounted for 50 ppm (parts per million). The effect of this amount of impurity was negligible since impurities in the published data were 100 ppm.

An americium-241 ($T_{1/2} = 458$ years) alpha source with a measured energy peak of 5.40 MeV was employed. The ^{241}Am was electrodeposited on a stainless steel disc measuring 1.5 cm diameter. The source strength, determined with a large area Au-barrier type Si detector, was 1.93×10^5 disintegrations per minute per 2π .²⁸ Absolute disintegration rate was not important since measurements were made by the comparison method. The source was positioned on the inside center of the chamber end plate. No collimator was used because it would have greatly reduced the intensity of the source which would have required longer observation times to produce the same accuracy.

B. Calibration of Equipment

Before each measurement, a calibration check of the electrometer, current digitizer and multichannel analyzer was made using a standard current source (Victoreen Model 475-429, Serial 176). The expected current produced by the standard was 1.09×10^{-11} amperes $\pm 1\%$. The purpose of the calibration check was to insure that the system was stable and that no drift in the electrometer or in the associated electronics was taking place.

C. Saturation Characteristics

For each gas a saturation curve (current vs. voltage) was plotted, for the collection of both positive and negative ions. To correct for ion recombination within each gas, it was necessary to extrapolate to the current expected for an infinite field. For current I and voltage V , one plots I/V vs. I and extrapolates as $V \rightarrow \infty$, $I/V \rightarrow 0$.²⁵ The extrapolated value would be the current collected for an infinite field strength. This method to correct for ion recombination is similar to that of Zanstra,²⁴ who suggests plotting $1/V$ vs. $1/I$ as $V \rightarrow \infty$. The results of these extrapolations are identical except that by using Hess' method I_∞ is extrapolated directly. A least squares fit²⁹ was applied to the linear portion of the extrapolation curve (5 highest voltage points) and a linear regression was used to calculate the intercept and its uncertainty.³⁰ This method of finding the intercept was used to reduce the possible errors resulting from a graphical extrapolation. The linear regression formula was also used to calculate the standard deviation of the intercept which is an indication of the magnitude of uncertainty in the intercept. Current produced at each voltage was divided by the extrapolated current (I_∞) to produce relative saturation curves for each gas for both polarities.

IV. Results and Discussion:

A. Determination of Optimum Conditions

In order to achieve optimum conditions for measuring W , it was necessary to stop essentially all of the alpha particles within the first 15 cm electrode segment of the chamber and at the same time reduce ion-recombination as much as possible. The optimum range of

the alphas was determined by adjusting the pressures in the chamber.

Fig. 3 shows experimental saturation curves at various pressures in air. Decreasing the pressure from 760 to 200 Torr caused saturation to be attained at a lower voltage and a greater current was collected, which indicated recombination was being minimized. Reducing the pressure further showed a decrease in current because energy was being lost to the walls of the chamber. At pressures <100 Torr, and voltages >400 volts, electrical arcing occurred between the chamber wall and the center electrode. For each gas under investigation, a similar routine was performed to insure optimum conditions.

Saturation characteristics of this chamber for each gas were determined for both polarities at pressures from 100 to 760 Torr. Higher current was collected using negative polarities because of gas amplification by electrons accelerated by the applied electric field. For the same field strength, positive ions caused less amplification because of their greater mass and resulting lower velocity. It was decided from this preliminary experiment that current formed by positive ions was to be used to determine W.

B. Recombination Corrections

At the desired pressure and voltage for each gas, a fractional recombination coefficient was measured and used to correct for current lost by recombination. Employing Hess' method,²⁵ a series of curves, as shown in Figs. 4 and 5, was used to correct for recombination. Fig. 4 shows the overall shape of the extrapolation curve (I vs. I/V) for positive and negative ions in nitrogen at 200 Torr. It can be seen that higher currents resulted from the collection of negative ions. The

Figure 3. Saturation curves for both polarities in air at various pressures.

Figure 4. Total extrapolation curves with outer chamber wall positive and negative.

curves become linear as $I/V \rightarrow 0$ and it was in this region the curves were extrapolated to find the current that would be expected for an infinite electric field.

An expanded view of the linear portion of the extrapolation curve is shown in Fig. 5. Solid and open points represent negative and positive polarities respectively. Error bars signify one standard deviation from the mean of three separate measurements. The solid lines represent least square fits and are extrapolated to $I/V \rightarrow 0$. Because of the similarity of the extrapolation curves for each gas investigated, only the nitrogen data have been shown for illustrative purposes.

Results of the extrapolated currents and their standard deviation are summarized in Table 2. In each case, the current for negative ion collection was higher than that for positive ions. The values in Table 2 were used to find the fractional recombination (Eq. 9) and also to construct relative saturation curves.

Table 2.

CURRENT AND ITS STANDARD DEVIATION
EXPECTED FOR AN INFINITE FIELD

<u>Gas</u>	<u>Expected Current ($\times 10^{-11}$ amps)</u>	
	<u>$I_{\infty}(+)$</u>	<u>$I_{\infty}(-)$</u>
N ₂	6.306 $\pm$ 0.005	6.341 $\pm$ 0.009
T.E.	7.492 $\pm$ 0.008	7.526 $\pm$ 0.006
CH ₄	7.923 $\pm$ 0.007	7.955 $\pm$ 0.004
Ar	8.776 $\pm$ 0.006	8.833 $\pm$ 0.011

Relative saturation curves (I/I_{∞} vs. voltage) for each gas are shown in Fig. 6 for the pressures used. The fraction of ions collected at various voltages for each gas are summarized in Table 3.

Figure 5. Expanded extrapolation curve in the linear region as I/V approaches zero for nitrogen.

Figure 6. Relative saturation curves (I/I_{∞} vs. Voltage) for each gas investigated.

Table 3.

RELATIVE SATURATION (I/I_∞) FOR EACH GAS
AT VARIOUS VOLTAGES AT THE PRESSURES USED

Gas & Polarity	Applied Voltage (Volts)					
	150	200	250	300	350	400
N ₂ (+)	0.993±0.001*	0.993±0.001	0.996±0.002	0.996±0.001	0.997±0.001	0.997±0.001
" (-)	0.996±0.002	0.997±0.002	0.997±0.002	0.997±0.002	0.997±0.002	0.999±0.002
T.E. (+)	0.991±0.002	0.995±0.003	0.995±0.001	0.996±0.002	0.997±0.001	0.998±0.001
" (-)	0.988±0.001	0.990±0.002	0.992±0.003	0.994±0.001	0.994±0.001	0.995±0.001
CH ₄ (+)	0.992±0.002	0.993±0.001	0.995±0.002	0.995±0.001	0.996±0.001	0.997±0.001
" (-)	0.990±0.001	0.993±0.001	0.994±0.001	0.995±0.003	0.966±0.001	0.997±0.001
Ar (+)	0.996±0.002	0.997±0.001	0.997±0.001	0.997±0.001	0.997±0.001	0.998±0.001
" (-)	0.994±0.002	0.997±0.001	0.996±0.001	0.997±0.001	0.999±0.001	0.998±0.001

*Uncertainties are one standard deviation limits

The fraction of ions lost to recombination was higher (~.3%) for positive than negative ions in nitrogen and argon. With T.E. gas it was just the opposite, and for methane the values were approximately the same. At saturation voltages (200 to 400 volts), recombination accounted for only about 0.4% of the total ions collected. The values listed in Table 3 were used to correct all current measurements for recombination losses.

C. W-Value Results

W was determined using the mean value of many repeated current measurements as described in the Experimental Methods. Current was collected over periods ranging from 15 to 100 hours. Data collected during each hour represented one measurement.

Certain problems were encountered during these measurements. The lower curve of Fig. 7 shows how current measurements were affected by changes in background radiation produced by operation of the High Flux Beam Reactor (HFBR) and Alternating Gradient Synchrotron (AGS) facilities at BNL. Increases and decreases correspond to start-up and shut-down procedures of those facilities. An increase of 6 μ R/hr above natural background produced a 0.4% increase in current measurements. The upper curve of Fig. 7 shows data from a typical measurement made when both the HFBR and AGS were operating continuously. All successive measurements were made under conditions similar to those found in the upper curve.

To insure reproducibility, measurements were repeated three times for each gas, each measurement being separated by an interval of two weeks. The results for each gas are listed in Table 4. Average cur-

Figure 7. Typical measurements when background was constant and when background was changing.

rents corrected for background and percent saturation are reported as mean values and associated standard error.³¹ The average current, of the three measurements for each gas, was compared to the average current produced in nitrogen and its previously published W-value to determine W.

Ratios of the current produced in each gas to the current produced in nitrogen are listed in column 6. If a more precise W-value for nitrogen can be found, these ratios can be employed to calculate W for each gas to a better precision since about 50% of the listed uncertainty is due to uncertainty in the previously published W-value for nitrogen.

The W-values calculated for each measurement plus the calculated mean of the three measurements are listed in column 7. Each determined W-value has an uncertainty of about 0.1 to 0.2%. Deviations within a single day were no greater than 0.05%. There was very good agreement between measurements from day to day, except for the single case of argon. The maximum deviations were 0.34% for argon, 0.06% for methane, 0.16% for T.E. gas and 0.09% for nitrogen. One value of argon falls just outside the 95% (2σ) confidence level of the mean value. Fig. 8 shows a typical frequency distribution of current collected for 100 hours. A Chi-square test was used to determine the probability, for repeated observations, that deviations greater than 1σ from the normal distribution would occur. The probability that a measurement would show a greater deviation was calculated as 27%. Therefore, in 3 out of 10 similar observations a greater fluctuation (1σ) could be observed. It is believed that the exceptional argon result mentioned above was within the limits of this statistical uncertainty.

Table 4.

EXPERIMENTAL RESULTS

Gas	P(Torr)	Applied Potential	Number of Observations	Avg. Ion Current* (10^{-11} amps)	Ratio of Current to N_2 Current	W-Values
N_2	200	350	60	6.3376 ± 0.0008 ****	-	-
"	200	300	48	6.3269 ± 0.0011	-	-
"	200	300	64	6.3314 ± 0.0009	-	-
			mean	6.3320 ± 0.0025	-	$(36.39 \pm 0.04)^9$
Rossi T.E.	200	∞ **	18 ****	7.4920 ± 0.002	0.8452 ± 0.0004 ****	30.76 ± 0.04 ****
"	200	350	24	7.4981 ± 0.002	0.8445 ± 0.0004	30.73 ± 0.04
"	200	300	17	7.5124 ± 0.003	0.8429 ± 0.0005	30.67 ± 0.04
			mean	7.5008 ± 0.0061	0.8442 ± 0.0008	30.72 ± 0.04
CH_4	300	∞ **	18 ****	7.923 ± 0.004	0.7992 ± 0.0005	29.08 ± 0.04
"	300	300	23	7.9280 ± 0.002	0.7987 ± 0.0004	29.07 ± 0.03
"	300	300	16	7.9191 ± 0.003	0.7996 ± 0.0004	29.10 ± 0.03
			mean	7.9234 ± 0.003	0.7992 ± 0.0004	29.08 ± 0.03
Ar	250	∞ **	18 ****	8.7760 ± 0.001	0.7215 ± 0.0003	26.26 ± 0.03
"	250	300	20	8.7760 ± 0.002	0.7213 ± 0.0003	26.25 ± 0.03
"	250	300	15	8.7335 ± 0.002	0.7250 ± 0.0003	26.38 ± 0.03
			mean	8.7627 ± 0.014	0.7226 ± 0.0010	26.29 ± 0.05

* Corrected for background and % saturation.

** Extrapolated to ∞ voltage

*** Number of points in extrapolation

**** Errors are standard errors

Figure 8. Typical frequency distribution of current measurements showing mean value and associated standard deviations.

D. Gas Analysis

Actual analysis of the gases investigated in this research reported by the Analytical Services Section were:

Rossi type T.E.:	CH_4	64.6%
	CO_2	32.4%
	N_2	2.7%
	C_2H_6	0.28%
	O_2	0.02%
	plus some trace organics	

N_2 :	N_2	99.995%
	O_2	0.005%

CH_4 :	CH_4	98.981%
	N_2	0.64%
	CO_2	0.20%
	O_2	0.05%
	C_2H_6	0.07%
	C_3H_8	0.02%
	Ar	0.009%
	other organics 0.02 - 0.03%	

Ar:	Ar	99.998%
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Since the impurities in the gases did not contribute to the Jesse effect, Strickler's modified equation (Eq. 7) along with the above values were used to calculate W for each mixture of gases. The calculated values are listed in column 3 of Table 5.

Table 5.

COMPARISON OF W-VALUES (eV/ion pair)

<u>Gas</u>	<u>Experimental Result</u>	<u>Gas Mixture Formula</u>	<u>Published Values</u>
N ₂	-	36.35	36.39±0.04 ⁹
T.E.	30.72±0.04	30.83	31.1±0.1 ⁷
CH ₄	29.08±0.03	29.14	29.1±0.1 ⁹
Ar	26.29±0.05	26.30	26.3±0.1 ⁹

E. Comparison of W-Values

W for methane and argon are in excellent agreement (better than 0.1%) with calculated and previously published W-values. This was an indication of the reliability of this technique for precise determinations of W. There was slightly less agreement (0.3%) between experimental and calculated W-values for T.E. gas. The experimentally determined W-value for T.E. gas is 1.2% lower than the previously reported W-value. The values do not agree within their limits of uncertainty. The values determined here are in closer agreement with the values predicted by Strickler's equation.

F. Error Analysis

The total error in the determination of W was estimated to be about 0.2%. The various sources of error are summarized in Table 6.

Table 6.

SUMMARY OF ERRORS

<u>Source</u>	<u>Estimated Error (%)</u>
Electrometer reproducibility	0.05
Voltage determination	0.01
Extrapolation (includes above)	0.1
Reference value	0.109
Average reproducibility error	0.1
Total uncertainty	0.18

Not included in Table 6 was the uncertainty ($\pm 0.1\%$) in the output of the current calibration source. Because determinations were made by comparisons of currents, any such systematic error in the absolute current calibration was cancelled out. The variations in the measured current from the standard source is included in the above reproducibility errors.

The largest sources of error were the extrapolated current (0.1%), reproducibility (0.1%), and the uncertainty in the reference value for nitrogen (0.1%). Uncertainty in the extrapolated current was calculated using analysis of variance³⁰ for the 5 points on the curve. Variations from the least squares fit, for each point, were reflected in the uncertainty of the extrapolated value. The average deviation in the reproducibility for determining W was 0.1%. The largest variation from the mean value was the single measurement for argon which was 0.34%.

V. Conclusion:

The W-value for T.E. gas determined in this study was 1.2% lower than the value published by Kemmochi, and shows better agreement with

the predicted value obtained using Strickler's equation. This lower value will reflect a proportional increase in the calculated dose delivered to a tissue equivalent ionization chamber.

The automated techniques employed in this study reduce the time required to analyze the data without sacrificing precision. The excellent agreement (0.1%) between the previously published values for methane and argon, and those found in this study demonstrates the reliability of this method for routine precise determination of W.

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