Absolute Energy to Produce an Ion Pair by Beta Particles from S^{35}†

WILLIAM P. JESSE, *St. Procopius College, Lisle, Illinois*

AND

JOHN SADAUKIS, *Argonne National Laboratory, Lemont, Illinois*

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The absolute average energy W_β necessary to form an ion pair in nitrogen, ethylene, and ethane was determined for S^{35} beta particles by the use of a sample assayed in a 4π counter. The total number of ions produced per second for each gas was determined by a measurement of total ionization in a chamber. The average energy per beta particle was calculated from the known shape of the distribution curve for S^{35}. Corrected values of W_β were found to be 35.3, 26.2, and 24.7 ev/ion pair for N_2, C_2H_4, and C_2H_6, respectively. These values yield a corresponding value for air of 34.1 ev/ion pair. All these are in excellent agreement with values previously predicted from relative beta- and alpha-ray measurements in various gases.

INTRODUCTION

In an earlier paper[1] results have been given for the relative ionization in a number of gases by the beta particles from tritium and from Ni^{63}. To these measurements now been added new measurements for the beta particles from C^{14}, which are in excellent agreement with the earlier results.

From certain regularities in these relative measurements the assumption has been made that in the noble gases and hydrogen the energy required to form an ion pair is the same for alpha and beta particles; hence W_β, the absolute energy per ion pair for beta ionization, is taken to be 26.4 ev/ion pair in argon, identical, that is, to the absolute W_α value found for polonium alpha particles in argon. It is then possible from the ionization measurements made relative to argon in various gases to determine numerical values of W_β for the various gases measured. Such measurements are given in the first four columns of Table I. As has been noted before, the W_β values exhibit no variation with beta energy larger than the experimental error throughout a range of average energies extending from an estimated energy of 3 to 5 kev for tritium to about 50 kev for C^{14}. This constancy of W_β with beta-particle energy is in agreement with the results of Valentine[2] taken over a more limited range of energies. On the basis of the assumption that W_β is invariant with beta-particle energy, the average W_β from columns 2 to 4 is computed and shown in column 5 of Table I. For the sake of completeness the absolute W_α values for Po^{210} alpha particles are listed in column 6. In column 7 is shown the ratio W_α/W_β. The constancy of this ratio within each of the three groups of gases indicated by the spacing is quite striking.

It is interesting to note that the mean W_β in air is about 34 ev/ion pair, a value considerable higher than the 32.5 ev/ion pair which has in the past been commonly quoted in the literature.

It should be emphasized at this point that the W_β values in column 5 are the result of purely relative beta particle measurements and of the assumption that in argon the W value for beta particles is the same as that for polonium alpha particles—an assumption as yet unverified. The verification of this assumption requires some form of absolute beta particle ionization measurements. The experiments described in this paper constitute a preliminary attempt at such absolute measurements.

METHOD AND APPARATUS

In this first attempt at absolute measurements, the somewhat obvious method was adopted of using a S^{35} source which had been accurately counted in a 4π proportional counter and then making an absolute measurement of the total ionization current produced by that sample in an ionization chamber filled with the desired gas. The average energy per beta particle can be calculated theoretically from a knowledge of the S^{35} beta-ray spectrum. The energy emission per second from the sample is then this average beta particle energy multiplied by the sample disintegration rate.

TABLE I. Summary of W values in ev/ion pair for beta particles of different energies.

Gas	W_β for Ni^{63}	W_β for tritium	W_β for C^{14}	Mean W_β for beta particles	Mean W_α for α particles from Po^{210}	Ratios W_α/W_β
He		42.3		42.3	42.7	1.0095
Ne		36.6		36.6	36.8	1.0054
A	(26.4)	(26.4)	(26.4)	(26.4)	26.4	(1.0000)
Kr	24.1	24.2	24.2	24.2	24.1	0.9959
Xe	21.8	22.2	22.1	22.0	21.9	0.9955
H_2		36.3		36.3	36.3	1.000
Air	34.1	33.9	34.0	34.0	35.5	1.044
N_2		34.7	35.2	35.0	36.6	1.046
O_2		30.9		30.9	32.5	1.052
CO_2	33.1	32.8	32.9	32.9	34.5	1.049
C_2H_4	26.3		26.2	26.2	28.0	1.069
C_2H_6	24.9	24.6		24.8	26.6	1.073
CH_4		27.3		27.3	29.2	1.070
C_2H_2		26.1	25.8	25.9	27.5	1.062

† This work was supported in part by the U. S. Atomic Energy Commission.

[1] W. P. Jesse and J. Sadauskis, Phys. Rev. **97**, 1668 (1955).

[2] J. M. Valentine, Proc. Roy. Soc. (London) **A211**, 75 (1952).

If this product is then divided by the number of ion pairs produced per second, as derived from the ionization chamber measurements, the value of W_β is at once obtained.

The accuracy with which W_β can be determined depends, of course, upon both the accuracy of the ionization measurements and the accuracy of the absolute counting methods. The accuracy of the calculation of the average beta-particle energy will also affect the result, but this error is probably much smaller than that encountered in either of the other two measurements.

The authors were fortunate in being able to obtain[3] S^{35} sources counted by the method of Pate and Yaffe.[4] The active material, benzidene sulfate, was distilled onto a VYNS film (mass thickness 30–40 micrograms per cm^2) on which a layer of gold (5 micrograms per cm^2) had been deposited. The samples were counted in a 4π proportional counter according to the technique described in the references cited. It seems probable that the counting technique developed by Pate and Yaffe will yield as accurate results as are obtainable anywhere by counting methods at the present time.

For the most accurate determination of W, two of the prepared samples were chosen: these were sample A, which initially gave an uncorrected disintegration rate of $1.36\pm0.01\times10^5$ dis/minute, and sample B, which, as of the same date, gave $1.96\pm0.02\times10^5$ dis/minute. A correction factor of two percent was in each case added for absorption in the gold-coated films.

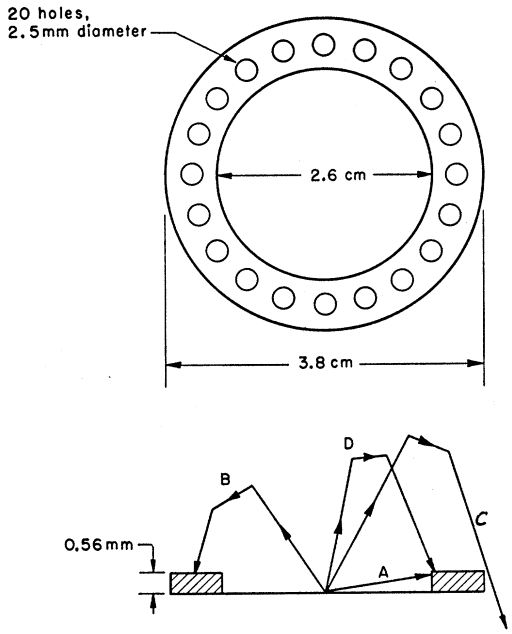

FIG. 1. Schematic diagram of sample mounting. A, B, C, and D represent possible beta-particle paths.

[3] The authors are greatly indebted to Professor Leo Yaffe for his kindness in furnishing these counted samples.

[4] B. D. Pate and L. Yaffe, Canadian Journal of Chemistry **33**, 610 (1955); **33**, 929, 1656 (1955); **34**, 265 (1956).

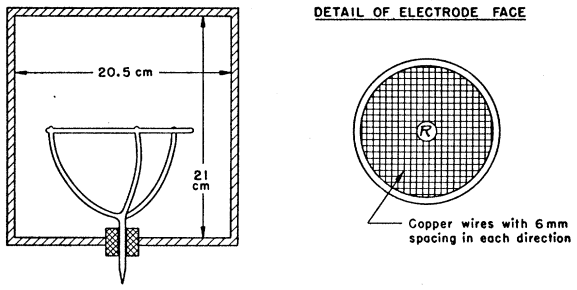

FIG. 2. Schematic diagram of ionization chamber for beta-particle current measurements.

The VYNS films were mounted on aluminum disks as is shown in Fig. 1. These disks were annular in shape, of thickness 0.56 mm, and perforated at intervals with circular holes as indicated. The diameter of the sample deposit at the film center was estimated to be of the order of one centimeter.

The technique of making the ionization measurements for the S^{35} samples was almost the same as that described in earlier experiments.[1] However, because of the higher energy of the S^{35} beta particles, a somewhat larger chamber was employed—a cylindrical one of brass of inside diameter 20.5 cm and height 21.0 cm (Fig. 2). The collecting electrode at the center of the cylinder was in the form of a ring 17 cm in diameter, supported by three wire stays of 0.3 cm diameter rising from the central insulated shaft. In the plane of the collecting ring and extending over most of the interior, was a square gridwork of copper wires 6 mils in diameter and spaced 6 mm apart in each direction. The use of this gridded construction was to minimize the effect of the backscattering of the beta particles from the gas itself. In the center of the electrode face the grid wires were cut away and a very narrow brass retaining ring R inserted. This ring was recessed to hold either the beta-ray sample or a flat brass circular disk inserted for chamber background measurements.

The general technique of ionization measurement was the same as in previous experiments,[1] where the ionization chamber was coupled to a vibrating-reed electrometer, which in turn fed into a Brown strip-chart recorder. Here again a null method was used, the drift of the electrometer being compensated by an applied counter potential from an external potentiometer. Since for absolute ionization current measurements one must know the capacitance of the system, this was measured periodically by the use of a double-ended standard condenser. Here one employs the known capacity difference between two possible electrode assemblies for the condenser. This capacity difference has been determined at the National Bureau of Standards four separate times within the past five years and twice within the progress of the present experiments. The agreement for all four measurements is within the experimental error of calibration—about 0.2%.

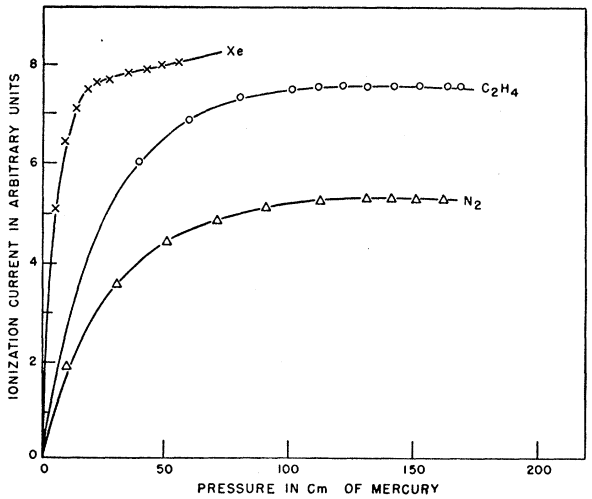

FIG. 3. Measured beta-ray ionization current in arbitrary units as a function of gas pressure.

In these experiments extensive measurements were made of the ionization produced by the two S^{35} samples in nitrogen, ethylene, and ethane. Less extensive measurements were made in argon, krypton, and xenon. For each gas it was the practice to make a preliminary run of the ionization current as a function of gas pressure, so that one could be sure that the final measurements would be made at a pressure where the total beta energy was absorbed. With increasing pressure (Fig. 3) the ionization current at voltage saturation rose at first rapidly and then more slowly until for nitrogen, ethylene, and ethane the current ceased to increase with further increase of pressure. For the denser gases, argon, krypton, and xenon, such a condition was never reached within the range of pressures convenient to use in this chamber. Even for the higher range of pressures the ionization current continued to increase slowly with pressure. Past experience showed these pressures to be well above those necessary to prevent the beta particles from striking the walls of the chamber. It is believed that this slow rise is probably due to a variation with pressure of the backscattering of the beta particles from the gas to the annular specimen mounting ring. This subject will be discussed more fully below in connection with the estimation of correction terms.

In Table II is given a summary of the data taken and the steps in the calculation of W_β for N_2, C_2H_4, and C_2H_6. In column 2 are listed the manufacturer's estimates of the purity of the gases given in column 1. These gases were obtained in steel cylinders. No further purification of the N_2 was carried out, after a preliminary experiment with the nitrogen continuously circulated over hot calcium at 250°C to remove possible traces of oxygen showed no change in ionization current from the untreated gas. The two hydrocarbons were at each use frozen in a bulb immersed in liquid nitrogen and pumped until all traces of incondensible gas disappeared. This method is effective in removing such incondensible gases, but probably will not remove the last minute traces of other hydrocarbon gases with similar physical characteristics.

Column 5 of the table gives the net ionization currents measured. The chamber background has in each case been subtracted. In general, the background of such a lead-shielded chamber is of the order of one percent of the total measured current. For a few of the runs, where the walls of the chamber became slightly contaminated, apparently with S^{35}, the background was higher. In such cases the background was checked very frequently. The total number of ion pairs collected per second is listed in column 6. This value is, of course, derived from column 5 by dividing by the value of the electronic charge in coulombs.

In column 7 are listed the disintegration rates of Samples *A* and *B*, corrected for film absorption. These

TABLE II. Data summary for absolute beta measurements.

Gas	Maker's estimate of purity	Sample	Run	Net current through chamber in amperes	Total ion-pairs per second	Sample disintegration rate, beta part./sec	Emission of beta energy, ev/per sec	Uncorrected W_β value, ev/ion pair
N_2	99.99%	*A*	1	3.912×10^{-13}	244.2×10^{4}	1.851×10^{3}	90.12×10^{6}	36.9
N_2		*A*	2	3.880×10^{-13}	242.2×10^{4}	1.836×10^{3}	89.40×10^{6}	36.9
N_2		*B*	1	5.384×10^{-13}	336.1×10^{4}	2.522×10^{3}	122.8×10^{6}	36.5
N_2		*B*	2	2.755×10^{-13}	172.0×10^{4}	1.292×10^{3}	62.92×10^{6}	36.6
Mean								36.7
C_2H_4	99.5%	*A*	1	5.576×10^{-13}	348.1×10^{4}	1.939×10^{3}	94.42×10^{6}	27.1
C_2H_4		*A*	2	5.327×10^{-13}	332.5×10^{4}	1.862×10^{3}	90.68×10^{6}	27.3
C_2H_4		*B*	1	7.279×10^{-13}	454.4×10^{4}	2.522×10^{3}	122.8×10^{6}	27.0
C_2H_4		*B*	2	3.571×10^{-13}	222.9×10^{4}	1.241×10^{3}	60.44×10^{6}	27.1
C_2H_4		*B*	3	3.540×10^{-13}	221.0×10^{4}	1.231×10^{3}	59.95×10^{6}	27.1
Mean								27.1
C_2H_6	99.75%	*B*	1	7.721×10^{-13}	482.0×10^{4}	2.502×10^{3}	121.8×10^{6}	25.3
C_2H_6		*B*	2	3.742×10^{-13}	233.6×10^{4}	1.222×10^{3}	59.49×10^{6}	25.5
C_2H_6		*B*	3	3.722×10^{-13}	232.4×10^{4}	1.213×10^{3}	59.06×10^{6}	25.4
Mean								25.4

are calculated for the dates in question on the basis of a half-life of S^{35} of 87.1 days.[5] This half-life value is in accord with values derived from the decay of ionization current with time in our own measurements.

In column 8, Table II, are found the values of the total beta energy emitted by the sample per second in units of electron volts/second. These values were obtained by multiplying the beta disintegration rate in the previous column by 48.7 kev, the calculated value for the average beta-particle energy for S^{35}.

This value of the average energy[6] was obtained by means of a graphical integration of the calculated beta-ray spectral distribution curve, with the assumption of an allowed shape and a maximum particle energy of 167 kev. In the above calculation the values of the Fermi differential function, $f(Z,n)$, were taken from the tables[7] of the Bureau of Standards. The function $f(Z,n)$ was corrected for screening by a fraction of a percent at low energy values by the use of the ENIAC calculations reported by Reitz.[8] The derived value of E_{Av}/E_{max} is 0.2916 with an estimated error of 0.2%. This derived value for E_{Av}/E_{max} is only slowly dependent upon the value chosen for E_{max}. A change of 1% in the latter produces only a 0.2% change in the former. The final derived value of E_{Av} is 48.7 kev, which has an estimated error of the order of $\frac{1}{2}$% if one assumes that the experimental value for E_{max} of 167.0 kev is good to $\frac{1}{2}$%.

In the last column of Table II are given the uncorrected experimental values for W_β. These are seen to be somewhat larger than the values predicted from the relative beta measurements shown in column 5 of Table I. The discrepancy is 4.9% for N_2, 3.4% for C_2H_4, and 2.4% for C_2H_6. Even larger discrepancies were found for argon and xenon in measurements not recorded in Table II. The values for these two gases were larger than the predicted values by eight and ten percent, respectively. Although such discrepancies are large, they are in the direction to be expected, since almost every error in ionization measurements has the effect of reducing the ionization current, that is, of increasing the measured value of W_β.

DETERMINATION OF CORRECTIONS FOR IONIZATION MEASUREMENTS

There are at least three corrections which must be applied to the experimental W_β values determined above. The first of these results from the loss of energy in those beta particles which pass through the thin film supporting the sample. The second corrects for the loss of energy by the beta particles which actually strike the sample mount directly after their initial ejection from the sample. The third deals with the loss in energy in those particles which are backscattered to the sample mount by the gas in the chamber.

1. A very crude calculation of the beta energy absorbed in the sample film gives a value of the order of less than $\frac{1}{2}$% of the total emitted energy. An experimental determination of this correction was carried out in the following way. An electrode was prepared identical with the one shown in Fig. 2, except that no center retaining ring was inserted, and the wire gridwork remained intact throughout the central portion of the electrode. The cross at the geometrical center of the electrode, where the two central wires intersected, was coated with a very thin radioactive coating by immersion in a droplet of a solution of elemental S^{35} dissolved in benzene. The total length of each wire coated was one centimeter, to accord with the dimensions of the active area on the original sample film. A blank film similar to those supporting the samples was available for these absorption measurements. This film was slightly larger than that shown in Fig. 1, being 5 cm in diameter and mounted on an aluminum ring of 7.5-cm external diameter. The center of the film was held in position directly over the active cross wires by means of very narrow circular retaining ring, itself held in position on the gridwork by three grooved feet. The height of the film was adjusted so it just failed to touch the active spot.

Successive readings were made with nitrogen in the chamber, first with the film and its mounting in position and then replaced by an exactly similar aluminum mounting ring, which bore no film. The interchange was then repeated. The experimental conditions duplicated those of the main experiment.

The ratio of the ionization current obtained with the film in the chamber to that without the film is given for two runs in column three of Table III. The two runs are in excellent agreement and indicate a total drop of ionization current of about 0.4%. To insure that this small change was not merely the effect of a slight increase in the capacitance of the chamber, occasioned by the introduction of the film on the surface of the electrode, the capacitance of the system was carefully measured with each configuration, and no difference could be detected.

It is interesting to note the difference in magnitude of this ionization film correction in comparison with the estimated two percent loss due to film absorption in the original 4π counting of these samples. This difference would seem to follow from the truism, that in the counter measurements the beta particles most susceptible to a counting loss by film absorption are the particles of minimum energy. In the ionization chamber, which essentially measures energy, the loss of such particles contributes but little to a loss in total measured energy.

2. The second correction to be determined results

[5] Hollander, Perlman, and Seaborg, Revs. Modern Phys. **25**, 469 (1953).

[6] The authors are greatly indebted to Dr. John H. Marshall of the Argonne Laboratory for his kindness in carrying out this calculation for them.

[7] *Analysis of Beta Spectra*, Bureau of Standards Applied Mathematics Series No. 13 (U. S. Government Printing Office, Washington, D. C., 1952).

[8] John R. Reitz, Phys. Rev. **77**, 10 (1950).

TABLE III. Ionization correction factors.

Gas	Run	Film absorption ratio	Correction ratio, backscatter plus mount interception	Uncorrected W_β from Table II	Corrected absolute W_β	W_β predicted from relative gas measurements	Corresponding absolute W_β for air
N_2	1	0.997_1	0.967_5				
	2	0.995_8	0.966_7				
	3		0.964_7				
Mean		0.996_2	0.966_3	36.7	35.3	35.0	34.3
C_2H_4	1		0.973_4				
	2		0.969_4				
Mean			0.971_4	27.1	26.2	26.2	34.0
C_2H_6	1		0.974_9				
	2		0.978_0				
Mean			0.976_5	25.4	24.7	24.8	33.9

from the interception by the wall of the aluminum sample ring of a very small number of beta particles emerging from the source. The path of one such beta particle is indicated at A in Fig. 1. From a knowledge of the solid angle subtended at the source by the wall of the ring, the gas pressure, and the stopping power of the gas, one can make a crude estimate of such an energy loss. Though this will vary, of course, with the pressure and the nature of the gas, for the three gases above under the experimental conditions used, the energy loss is of the order of 1% of the total energy. However, the experimental determination of such a correction is included in the determination of the third correction, which follows immediately.

3. Probably the greatest source of error in the ionization measurements described above results from the backscattering of the beta particles from the gas in the chamber to the sample mount. This effect of backscattering from the chamber has already been noted[1] and a series of measurements[9] has recently been made to investigate various phenomena connected with it.

A beta particle, B in Fig. 1, on emerging from the source may, by either single or multiple scattering in the gas, be deflected back to the ring and thus lose part of its energy. The measured loss has been found to depend upon the size and conformation of the electrode, the pressure of the gas, and upon the atomic number of the gas—the higher the atomic number the higher the energy loss from backscattering. For gases of high atomic number the energy losses can be very large. Thus with a large surface electrode in xenon the ionization energy loss may be as large as 40%.

It should be noted that with the ring electrode of the present experiment the energy losses, and hence the measured ionization current losses, are a function of the radial distribution around the central axis of the chamber of the backscattering beta particles. Such losses are also a function of the gas pressure, since the latter determines the scale on which such a radial distribution diagram is drawn. Thus the scattered particle at C in Fig. 1 may at a slightly higher pressure hit the outside edge of the ring and thus lose remaining energy without ionization. Similarly a scattered particle at D may at a slightly higher pressure cease to hit the ring and pass through the film, giving an increased ionization. Thus the final observed constancy of ionization with pressure (Fig. 3), as is the case of N_2 and C_2H_4, may not indicate a condition of no loss of energy by backscattering, but it may merely indicate a balance between the energy lost and gained for a given increment of pressure change. Similarly the slow rise of ionization with pressure observed in argon, krypton, and xenon may merely indicate an asymmetry in the distribution of backscattered beta particles under these particular conditions, such that for a given increment of pressure increase the ionization energy gained is larger than that lost. Finally it should be noted that with a gas of high atomic number at sufficiently high pressure it should be possible, at least theoretically, to shrink the backscattered beta-ray pattern to such a point that the rays strike only the central film. Under these conditions the correction for backscattering from the gas should then be virtually eliminated.

Since attainment of such high pressures was not possible in the present measurements with N_2, C_2H_4, and C_2H_6, corrections for backscattering were made by a method similar to that used for determining the film absorption. Once more the auxiliary mesh electrode with the active center wires was employed. With this bare electrode in the chamber ionization current measurements were made under the same conditions which obtained in the original experiments. In most cases the gas pressure was the highest available—162 cm of mercury. Alternate runs were also made with a "dummy" ring assembly identical to that supporting the standardized sources but carrying no film, placed on the grid wire face of the electrode and accurately centered about the active spot. The sample ring assembly included a dummy brass ring identical to the

[9] A paper dealing with such measurements is now in preparation.

one shown at R in Fig. 2, to simulate accurately the conditions of the original experiment.

Here again the ratio of the currents obtained with the two electrode configurations was very reproducible, as is shown by the results for different runs in column 4 of Table III. From the mean ratios it is seen that the correction term is 3.4, 2.9, and 2.4% for N_2, C_2H_4, and C_2H_6, respectively. This sequence of values is again in accord with our previous experience, that the back-scattering from gases rises in value with the effective atomic number. It should be pointed out, however, that the measured corrections include also our second correction, that is the correction for the energy directly intercepted by the mounting ring. Since this is relatively small and should not vary markedly for the three gases in question, it is improbable that the rising sequence of values would be altered.

By a use of the mean correction ratios in columns 3 and 4 of Table III, it is possible to correct the direct experimental W_β given in the last column of Table II and for convenience repeated in column 5 of Table III. These last values are successively multiplied by the ratios in columns 3 and 4 to give the corrected absolute values in column 6. The ratio, 0.996 for film absorption, determined only in nitrogen, has been assumed the same in all three gases.

It is interesting to compare the corrected absolute values of W_β with the values predicted from relative gas measurements on the assumption that W_β and W_α are identical for the noble gases. These latter values, taken from column 5 of Table I, are for ease of comparison retabulated in column 7 of Table III. The agreement between the values in columns 6 and 7 is excellent, the maximum difference in the case of nitrogen being less than 1%. Such agreement is much better than one has a right to expect, when one reviews the possible errors in the absolute measurements. With an estimated error of 1% in the sample counting and an error of the same order in the over-all ionization measurements, a combined error of almost 1½% would not be unexpected. The almost exact agreement must therefore be regarded as in some degree fortuitous.

Since absolute values of W_β for air were not directly determined here, and since such values are always of especial interest, they were calculated from the present results and are shown in column 8 of Table III. These values were obtained from the determined absolute values for the three gases in question by multiplying each by the ratio $W_\beta(\text{air})/W_\beta(\text{gas})$, the W_β values being taken from the mean values in column 5, Table I. It should be emphasized that this multiplying ratio is fundamentally an experimentally determined ratio of two ionization currents and is in no way dependent upon any assumptions as to the equality of W_α and W_β in the noble gases. Each of the three absolute W_β values gives rise to a value of W_β for air. The agreement between these last values is very good, with a mean value of 34.1 ev/ion pair. The agreement is also good with the predicted value for air of 34.0 ev/ion pair from Table I.

The above results are in good agreement with the recent results of Weiss and Bernstein,[10] who for x-rays of an estimated average energy of 1 Mev found W_β values of 26.4, 34.6, and 33.9 ev/ion pair for ethylene, nitrogen, and air, respectively. In addition to these published values, the present authors have become aware through discussions during the past year of a number of researches which indicate a probable value of W_β for air in the neighborhood of 34 ev/ion pair—in good agreement with the result given above. However, since most of this work has not as yet appeared in final detailed publication, any critical comparison with the above results would seem at this moment premature.

CONCLUSION

The results of this preliminary determination of absolute W_β values are in excellent agreement for all three gases used with the values predicted from relative measurements in different gases with the assumption of the equality of W_β and W_α in the noble gases. However, even though they can be determined with apparently a high degree of precision, the corrections of several percent necessary in the above absolute measurements are somewhat disturbing. It should be pointed out, however, that these preliminary experiments suggest a method of eliminating such ionization corrections by a redesign of the sample ring, which shrinks both its width and thickness to a minimum. A mounting of the sample film upon a circular loop of relatively fine wire would be an ideal arrangement and should virtually eliminate the need for all ionization current corrections except the very small one for film absorption.

It is a pleasure to express our grateful appreciation to a large number of friends and colleagues who have helped in various ways in this work. We should like to express our thanks to Professor Leo Yaffe who furnished the counted S^{35} sources which made this work possible and to Dr. John H. Marshall who carried out the calculations to determine the mean beta-ray energy in S^{35}. For many stimulating discussions of this subject we are greatly indebted to Professor Robert L. Platzman, Dr. Francis R. Shonka, Mr. John E. Rose, and Mr. L. D. Marinelli.

[10] J. Weiss and W. Bernstein, Phys. Rev. **98**, 1828 (1955); **103**, 1253 (1956).