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Measuring the Absolute
Disintegration Rate of a
Radioactive Gas with a
Moveable Endplate Discharge
Counter (MEP) and
Theoretical Calculation of
Wall Effect

by Arthur H. Jaffey, James Gray,
William C. Bentley, and Jerome L. Lerner

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WITH A MOVEABLE ENDPLATE DISCHARGE COUNTER (MEP)
AND THEORETICAL CALCULATION OF WALL EFFECT

by

Arthur H. Jaffey, James Gray,
William C. Bentley, and Jerome L. Lerner (deceased)

Chemistry Division

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ABSTRACT

A precision built movable endplate Geiger-Müller counter was used to measure the absolute disintegration rate of a beta-emitting radioactive gas. A Geiger-Müller counter used for measuring gaseous radioactivity has <100% counting efficiency owing to two factors: (1) "end effect", due to decreased and distorted fields at the ends where wire-insulator joints are placed, and (2) "wall effect", due to non-ionization by beta-particles emitted near to and heading into the wall. The end effect was evaluated by making one end of the counter movable and measuring counting rates at a number of endplate positions. Much of the wall effect was calculated theoretically, based on known data for primary ionization of electrons as a function of energy and gas composition. Corrections were then made for the "shakeoff" effect in beta decay and for backscattering of electrons from the counter wall. Measurements and calculations were made for a sample of ^{85}Kr (beta energy, 0.67 MeV). The wall effect calculation is readily extendable to other beta energies.

1.0 INTRODUCTION AND HISTORY

The disintegration rate of a gaseous radioactive substance is conveniently measured with a gas counter, the radioactivity being mixed with an appropriate counting gas. This method has a number of advantages, provided that the chemical nature of the radioactive substance is compatible with the

proper operation of the gas counter. In the majority of cases of interest, the radioactivity involves the emission of electrons in each decay process, and the work described here is focused on this type of radioactive species. The counter often employed in such measurement is a discharge counter in the form of a cylinder with an axial wire, which is operated either as a proportional counter or a Geiger-Müller (GM) counter. The use of an internal counter avoids the problems of solid angle evaluation and correction for absorption in the counter wall or sample container which arise when the radioactive sample is measured by placing it external to a counter. If every decay yields an electron, and if every electron is counted, the disintegration rate may be derived directly from the counting rate. Most of the emitted electrons are indeed counted in such an internal counter, so it is a good choice for disintegration rate determination. We report here on a measurement of the disintegration rate of a krypton gas sample containing the radioactive species ^{85}Kr .

In making such a measurement, it has long been known that it is essential to correct for certain losses that occur in these gas counters which cause the rate at which discharges occur to be less than the disintegration rate. These losses are generally described as arising from the end-effect and the wall-effect. The end-effect causes the more important loss, and results from the fact that the electric field distribution which creates the countable discharges is not the same throughout the counter. The radial distribution of field is longitudinally uniform through a large central region of the counter, but changes radically near the ends due to the fact that the counter is there terminated by insulators which support the wire. The discharges created by ionizing particles near the ends tend to be smaller than those created near the tube center, and are not counted.

The losses that occur at the tube ends depend considerably on the design of the insulating region. In the period when gas counters were the predominant form of radioactivity detector in use, considerable research was carried out aimed at improving the field distribution at the counter ends. The gas counter is still quite convenient for measuring gaseous radioactive samples, and it is only when the actual disintegration rate is to be evaluated with some accuracy (< 5% error) that end effect losses are carefully considered.

The wall effect arises because of the stochastic process involved when an electron loses energy in passing through an absorber, in this case the counter gas. If the absorber is thin enough, there is some probability that a beta-particle emitted near the wall will not create any ionization at all before it hits the wall. As described in the Appendix, the probability of zero ions formed through ionization by a beta particle of energy E (provided it is not backscattered) is:

$$P(0) = e^{-N_a \sigma_p L} \quad [1]$$

where

N_a = number of gas molecules per cm^3

σ_p = cross section for primary ionization (emission of an electron from a gas molecule due to collision with a beta particle.)

L = distance (cm) from the point of beta decay to the tube wall, measured along the beta particle trajectory.

In order to evaluate the effect accurately, it is necessary to include all the variations of: (i) beta particle energy E , i.e., beta-particle intensity as a function of E and σ_p as a function of E ; (ii) position of emission; (iii) direction of emission; and (iv) distance to wall. The wall effect is generally of the order of 1% or less in magnitude, and a major part of it can be calculated from the known physical quantities in the variables (i-iv). An analysis of the calculation procedure is presented in Appendix A as well as the results of a particular set of calculations for the radioactive species and the gas filling discussed in this paper.

The magnitude of the end effect is larger and is not readily calculable. Correction for the effect, therefore, has generally been carried out empirically. The basic concept for the experimental evaluation has been to use two or more cylindrical counters with widely different lengths L_1 , L_2 , L_3 ..., but with identical (and uniform) internal diameters, identical wires, and identical end structures. For example, we consider a case with two counters (1,2), which are alike in all respects except length.

The first counter (of length L_1) may be treated as a large central region with counting rate per unit length k_C with two smaller end regions (of lengths ΔA and ΔB) with unit counting rates k_A and k_B (Fig. 1). On the assumption

Fig. 1. Modeling the counter as a large central section with uniform counting rate per cm, with two small end sections having smaller counting rates per cm

that the counts from the central and end sections may be treated independently, the counting rate from this counter is:

$$C_1 = k_A \Delta A + k_C (L_1 - \Delta A - \Delta B) + k_B \Delta B \quad [2A]$$

If the same concentration of radioactive gas is measured in the second counter (length L_2) and if operating conditions (gas pressure and composition, operating voltage and electronics arrangement) are the same, then

$$C_2 = k_A \Delta A + k_C (L_2 - \Delta A - \Delta B) + k_B \Delta B \quad [2B]$$

Since the counts from the ends are the same in both counters, the difference in counting rates is:

$$C = C_2 - C_1 = k_C (L_2 - L_1) \quad [2C]$$

In effect, one has measured the gas in a shorter tube with no end effects.

This method has been used by a number of investigators who wished to measure the disintegration rate of a radioactive gas. If the concentration of the radioactive isotope in the total gas is also measured, the technique then allows the determination of the specific activity (hence, the half-life) of a long-lived radioactive species such as ^{14}C . (Typical examples of the two counter approach are given in Refs. 1-6).

In the 1950's there was considerable work done by French T. ("Pete") Hagemann at the Argonne National Laboratory (ANL) to develop this technique. After extensive experiments in which a number of attempts were made to make identical end structures for the long and short counters, he succeeded in making some tubes which had end structures which were reasonably closely alike. Using two sizes of stainless steel counters (3" i.d. x 12" long and 3" i.d. x 24" long), he measured the absolute disintegration rates of several kinds of radioactive gas, including a sample of ^{85}Kr gas (here denoted as KrI).

The latter sample was of particular interest to us since some samples of ^{85}Kr have been kept at ANL as long-lived reference gases for calibrating the counting efficiencies of gas counters. The original two-counter calibration

was done in 1954 on KrI, which was later compared to a more active ^{85}Kr gas sample after KrI had considerably decayed. Comparisons were made by counting both KrI and its replacement gas in the same GM counters, with no attempt at absolute disintegration measurement. Hence, only a ratio of disintegration rates was determined. The original calibration was then periodically preserved in new ^{85}Kr gas samples as they replaced the older ones whenever these had decayed too much. The current reference gas (KrII), whose absolute disintegration rate could then be related to that of KrI, was measured in the present experiment.

After completing this work with the two-counter method, Hagemann was not satisfied that the counter ends were satisfactorily replicated to the accuracy he desired, so he attempted an alternative approach involving a single counter. With the assistance of one of the authors (J.G.) and the machinist Thomas Fallon, he tried a number of designs of moveable piston counters. In such a counter, the wire was fastened to a feedthrough insulator in a fixed position at one end and was strung through the center of a moveable insulating piston at the other end. A necessary feature of the design was that the counter should produce measurable pulses only on one side of this piston. The effective length of the counter could then be varied, and since the end structures (insulator feedthrough and piston) remained the same at each length, measurements corresponded to those in counters of different lengths with identical end effects.

A satisfactory design was developed by 1957; it was called the MEP (Moveable End-Plate) counter. Shortly after a few preliminary measurements were made, Hagemann became ill, and further work with MEP ceased. It was set aside in storage and it was not until recently that it was decided to carry out some definitive measurements with MEP to demonstrate that it could, indeed, be used for accurate measurement of disintegration rates. Measurement of the current reference ^{85}Kr gas (KrII) would allow comparison of MEP results with the previous two-counter measurement of KrI.

Other work using moveable end counters has been described in Refs. 8-10.

2.0 DESCRIPTION OF MEP

2.1 Structure. Diagrammatic drawings of the essential elements of MEP are shown in Fig. 2. The body of the counter is a heavy wall (~5 mm

Fig. 2. Diagram of MEP (Movable Endplate Counter)

thickness) tube of OFHC copper ~48 cm long and with an outside diameter of ~6 cm. Flanges for use of gold "O-rings" are attached to the ends. The mating flange at one end holds the stationary end of the counter and the emergent counter wire; at the other end, the mating flange holds the moveable piston and the precision scale. The interior of the tube was bored and reamed to complete circularity and straightness, and honed to a shiny surface. Measurement down the bore showed the tube to be accurately circular and the diameter to be very uniform, most of the values varying between 1.9944" to 1.9945" (opening up to 1.9946" near the ends). The diameter is taken to be 1.9945" (= 5.066 cm).

The endplates (Fig. 2) at both ends are identical. Each endplate is essentially a quartz disk held at its outer edge in a brass cylinder which provides support and serves as the moveable element at one end. A small gold bushing is set at the center of the disk with a highly polished hole at its center which is just large enough to allow the wire to slip through. As the wire passes through the hole, it is slightly offset to insure rubbing contact with the bushing. This serves to terminate the active wire length at the position of the disk; some such provision is necessary because the counting gas is actually present on both sides of each disk.

Back of the moveable quartz disk is a spring loaded rewind mechanism which coils the wire up as the piston is moved and keeps the wire in the counting chamber under tension. A quartz cup (not shown in the figure) which is platinized in its interior surrounds the take-up reel (Fig. 2). The metal structure of the take-up reel is connected to the platinized surface with a phosphor bronze spring. Hence, the coiled section of wire is surrounded by conducting surfaces which are at the same potential as the wire, thus preventing the occurrence of counter discharges in regions outside of the designated counting volume.

The piston is attached to a square rod which has a precision scale on one face and a fine-tooth rack on the opposite face. The vacuum-tight superstructure contains a fixed vernier scale, a quartz viewing window, a rotary bellows vacuum seal with a pinion gear to drive the rack, and a long thin tube to accommodate the rod with its scale. The accuracy of the etched lines on the scale was shop-measured with a traveling microscope and found to be at least as good as 0.01 mm, which is better than the reading achievable with the

installed vernier. A magnifying glass and focused light external to the viewing window allows ready examination of scale and vernier.

Assembly of MEP had to be carried out very carefully. After any necessary repairs or changes were made, fresh gold gaskets were inserted if necessary, and all the components (including the wire) and the interior were washed with fresh, very high purity methyl alcohol. The most difficult part of the assembly involved mounting the wire. This involved threading it through the piston and connecting it to the take-up reel, and then passing the other end through the stationary insulator and soldering it. Final assembly was followed by pumpout and leak checking.

Relative positions of the moveable endplate could be accurately measured by observing the position of the rule; displacement of the endplate could be measured as accurately as the rule could be read. For some purposes, it was desirable to know the actual length L of the tube portion between the two endplates. A measurement of L was carefully made with the unit disassembled and appropriately supported, and the rule position was then set. However, the actual value of L at a particular rule position setting depended upon the positions of the two end flanges, and these positions were slightly dependent upon how much the two gold gaskets were crushed. Since the counter was disassembled and reassembled many times, the absolute value of L was known with less accuracy than were the relative displacements of the moveable endplate.

2.2 Mode of Operation. MEP was found to be usable both as a proportional counter and as a GM counter, although the wire diameter was smaller for the former. For the present measurement, it was decided to use the GM mode, and the counter was fitted with a 0.003" (0.075 mm) soft temper stainless steel wire; the alloy was one which allowed the coil to be tightly coiled up many times without becoming distorted. Using the GM mode offered the advantage that when an ionization event occurs within the cylindrical electric field, the output pulse size is not dependent (at least in the central region) on the amount of initial ionization, hence is not a function of electron energy.

A problem arising with the GM mode is that with some gas fills it was evident that spurious pulses occasionally formed which occurred shortly after true ionization events. When the spurious pulses occurred, they were detected

through the use of an oscilloscope triggered by the GM tube output; a time-correlated secondary peak was observable a few milliseconds after the primary peak. This observation was further substantiated by measuring the distribution of time intervals between all output pulses through the use of the instrument TIDA (Time Interval Distribution Analyzer). A graphic presentation of the time interval distribution showed a peak at the few millisecond interval.

The source of such secondary pulses in GM counters had been established quite a while ago. When the transit of a beta particle ionizes an atom in the krypton gas, the released electron accelerates towards the positively charged central wire, creating a discharge, consisting of many electrons and krypton positive ions (Kr^+). The latter move to the wall, while the electrons move to the wire. There is a high probability that when a krypton ion collides with the wall, it will create a secondary electron on transferring its charge to the wall, and this electron accelerates towards the wire, creating a new discharge. The two discharges are time-correlated, the time difference being dominated by the slower transit time of Kr^+ across the tube from the vicinity of the wire where most of the positive ions are formed. To prevent the formation of this secondary discharge, it is usual to add an organic "quenching" agent, such as ethyl ether. A neutral molecule of ether exchanges charge on collision with Kr^+ , and the positively charged molecule then travels to the cathode. On transferring its charge to the wall, the ether molecule dissociates rather than extracting an electron, thus terminating the process started by the original ionization. If the charge exchange process occasionally does not occur, then some secondary discharges may be evident.

To check this interpretation, an approximate calculation was made of the Kr^+ transit time. The result indicated that for the electric field, counter gases, gas pressure and dimensions involved, this time would be expected to be in the neighborhood of 2 milliseconds.

To eliminate the nuisance of testing for the presence of secondary pulses, it was decided to modify the measuring circuit so as to eliminate the possibility of measuring secondary pulses. Since no time interval between primary and secondary discharges was ever found to be as large as 4 milliseconds, it was decided that no pulse was to be accepted for counting unless it arrived at the counting circuit more than 4 milliseconds after the

previous discharge. This requirement also served the purpose of preventing the counting of a spurious discharge which was generated secondarily from a secondary discharge, since this could occur more than 4 milliseconds after the last count. Thus, the effective deadtime was not a fixed one, but was extended by 4 milliseconds whenever a discharge occurred less than 4 milliseconds after the last discharge. Since the deadtime could then be variable, depending upon the rate of spurious discharges, it was necessary to use a standard livetimer. This measured the total time during which the counter was available to produce a countable discharge after the last count.

2.3 Circuitry. MEP was operated with the wall at ground potential and with positive high voltage applied to the center wire through a two megohm resistor. Because of limited space in the shielding tomb, the only circuitry used there was a line driver (capacitatively coupled to the center wire), which provided a low impedance signal for the external circuitry. Similarly, the outputs of the anticoincidence scintillator bank photomultipliers were delivered by line drivers to the external circuitry.

The scintillator signal was amplified and passed through a pulse height discriminator to eliminate pulses which were too small to have come from muons. The surviving pulses triggered a monostable multivibrator (MMV_1), which delivered a 100 μs square wave output for use in implementing the anticoincidence operation discussed below. MMV_1 pulses were also accumulated in the ACB (anticoincidence bank) scaler.

The MEP driver was connected to two independent circuit trains to allow use of the extending deadtime operation as referred to above as well as a conventional fixed deadtime. Thus, for fixed deadtime, the driver output passed through a discriminator, and surviving pulses triggered the multivibrator MMV_2 , with output pulse width 4 ms. This MMV was not retriggerable until the end of the 4 ms period. Immediately after MMV_2 fired, another multivibrator (MMV_3) also fired, with a 50 μs pulse-width output. The relative pulse timing was arranged so that the scintillator MMV_1 output and the MEP MMV_3 output overlapped adequately if the initiating events were time-coincident. The MMV_3 output sent enabling signals to two different multivibrators, MMV_4 and MMV_5 . MMV_4 was the BC (before coincidence) unit and always fired when activated by MMV_2 . It served to drive the BC scaler, totaling all acceptable discharges from MEP. MMV_5 , the AC (after coincidence)

unit was also connected to the MMV_1 output, and fired only if a scintillation output pulse was not time-coincident with a MEP MMV_3 output pulse. MMV_5 drove the AC scaler, totaling all acceptable MEP discharges which were not time-coincident with muon scintillations. Then $(BC - AC)$, the difference between the two scaler outputs was equal to the number of coincidence events between MEP counts and muon events.

For the extending deadtime system, the circuit train was identical, except that the discriminator fired a 4 ms MMV which could be retriggered before the termination of the 4 ms period, and then remained triggered for only another 4 ms, unless retriggered again. The other components, equivalent to MMV_3 , MMV_4 , and MMV_5 , and the BC and AC scalers, were identical to those in the other system. The AC MMV unit for the extending deadtime train was also used to provide pulses whose time interval distribution was measured with TIDA.

Each of the two deadtime systems had two timers. One was an elapsed timer, which was basically a scaler which accumulated a train of 100 pulses/min derived from the 60 cycle power line.* The other was a livetimer, which also accumulated signals from a train of 100 pulses/min derived from the 60 cycle line. However, this pulse train was interrupted by an anticoincident system during the time periods that MMV_1 or MMV_2 (or its extending deadtime equivalent) were active. Thus, these time periods were not sampled, because no activation of MMV_5 (AC output) could occur while these circuits were operating. Since the livetimer sampled only "open" time periods, calculation of the counting rate did not require knowledge of the magnitude of the actual deadtime.

TIDA, the Time Interval Distribution Analyzer, was a circuit designed to measure the distribution of time interval lengths between adjacent output pulses from a radiation detector. Aside from deadtime effects, a normally operating detector should deliver a train of pulses following a Poisson process. The distribution of interval lengths from such a process is exponential.

*Measurements against time standards had shown this time source to be quite accurate, at least over periods which are a good part of a day, as in the present experiment.

TIDA was a multichannel time analyzer with time channel boundaries adjusted on a logarithmic scale. With such an adjustment a Poisson process should yield (on the average) a uniform distribution over the various channels. Such uniformity could be perturbed by: (1) the usual fluctuations expected in any random process, (2) the fact that since TIDA is based on a digital countdown system, the logarithmic relation could only be approximated by the sequence of digital numbers chosen for the several channels, and (3) the deviation of the setting chosen for the countdown rate from that suitable for the actual mean counting process rate.

In the particular realization of the TIDA design used, an integer sequence was selected for the various channels, corresponding (with the necessary digital approximations) to 1% probability from the exponential distribution in each of the first 8 channels, and 4% probability in each of the last 23 channels, the final channel having a terminal boundary corresponding to "infinite" time. In operation, the occurrence of a count from the counting process train activated a sequence of events: The first channel integer was entered into a countdown scaler, which was counted down using an oscillator-driven pulse train of rate F . When the scaler reached zero, the second channel integer was entered, etc. When the next count from the counting process train occurred, this advancing process stopped, and a count was accumulated in the appropriate channel of a standard multichannel analyzer (MCA). Simultaneously, the first integer was again entered into the countdown channel, and the sequence advanced until the next counting process count terminated the countdown. The output consisted of the accumulated events in the MCA, which represented the distribution of the time-interval lengths in the counting process.

The frequency F of the oscillator was adjusted so that its rate corresponded to the mean rate M of the counting process, which had been evaluated with a short preliminary measurement. The desired ratio of F to M (i.e., scale factor) was fixed by the particular set of integers stored for use in the countdown process. If the desired ratio of F to M differed from the actual one, this would correspond to the various time channels actually used not being the a priori defined probability (1% and 4% bins). However, this deviation was correctable in subsequent computer analysis, in a program which made the necessary adjustments for deviations of the frequency used from

the desired value and for the integer approximations to the defined probability boundaries. Chi-square analysis and graphical presentation served to give summary results.

2.4 Two Counting Systems. Although the counting rates used were low enough so that the deadtime corrections were always modest in size, it was desirable to check the method used by independent procedures. The basic hardware elements have been described in the Section 2.3 Circuitry.

System 1.. Input with fixed deadtime (initial MMV not retriggerable), a BC scaler with total output N_{B1} , an AC scaler with total output N_{A1} , an elapsed timer with total output T_{1E} and a livetimer with total output T_{1L} . The measured counting rate could be calculated in two ways, first by using the livetime value T_{1L} , or by using the elapsed time T_{1E} and correcting for the effect of a fixed deadtime with the standard formula for a Type I counter. A GM tube with an intrinsic deadtime* smaller than the "clamping" external electronic deadtime is known to be well-fitted with the Type I relation if the counting rate is not high.

For the first method, the measured counting rate was

$$C_1 = \frac{N_{A1}}{T_{1L}} \quad [3A]$$

For the second method, the counting rate corrected for deadtime was:

$$C_1' = \frac{C_{AC}}{(1 - \tau_1 C_{BC} - \tau_2 C_{Mu})} \quad [3B]$$

where

$$C_{AC} = \frac{N_{A1}}{T_{1E}} \quad \text{and} \quad C_{BC} = \frac{N_{B1}}{T_{1E}} \quad [3C]$$

*Intrinsic deadtime as opposed to an externally-applied deadtime created by electronic circuitry, e.g., a monostable multivibrator. Intrinsic deadtime is a period following the onset of a discharge during which a new ionization event cannot create a new discharge because the traveling positive ions distort the counter's electric field.

C_{AC} was the observed net counting rate to be corrected for dead-time losses, but C_{BC} was the rate at which MEP causes deadtimes to occur. C_{Mu} was the muon counting rate (in the scintillator bank) of those muons which independently caused deadtime periods because they were not coincident with MEP counts. τ_1 was the 4 ms deadtime created by MEP's MMV₂ and τ_2 was the 100 μ s deadtime created by the scintillation bank's MMV₁. C_{Mu} was calculated from N_{ACB1} (the muon total count in the anticoincidence scintillator bank) after subtracting the number of events in which muon counts and MEP counts were coincident. Then

$$C_{Mu} = \frac{N_{ACB1} - (N_{BC} - N_{AC})}{T_{1E}} \quad [3D]$$

Since the livetimer was inactivated whenever either the anti-coincidence bank or MEP delivered an output, then

$$T_{1L} = T_{1E} - \tau_1 N_{B1} - \tau_2 [N_{ACB1} - (N_{B1} - N_{A1})] \quad [3E]$$

Since C_1' may be written as

$$\begin{aligned} C_1' &= \frac{N_{A1}/T_{1E}}{1 - \tau_1 N_{B1}/T_{1E} - \tau_2 [N_{ACB1} - (N_{B1} - N_{A1})]/T_{1E}} \\ &= \frac{N_{A1}}{T_{1E} - \tau_1 N_{B1} - \tau_2 [N_{ACB1} - (N_{B1} - N_{A1})]}, \end{aligned} \quad [3F]$$

it is evident (from Eq. [3E]) that Eq. [3F] is identical to Eq. [3A]. Hence, calculating the counting rate with either method is algebraically the same, provided that the livetimer has performed correctly, and that MEP, at the counting rates used, behaved like a Type I counter. If secondary spurious events occurred, then they were counted, and neither C_1 nor C_1' applied, since N_{A1} was inflated by the "false" counts.

System 2. Input with extending deadtime. This system had the fixed deadtime τ_1 of System 1 if none of the dead periods were extended during the initial 4 ms, so that either calculation method could be used for this system, yielding rate C_2 and C_2' from

relations equivalent to Eq. [3A] and Eq. [3B]. However, if an appreciable number of extended deadtime periods occur, then only the equivalent of Eq. [3A] was appropriate, and was correct even when spurious secondary ionization events occurred, since these were suppressed.

Measuring with the two procedures provided a check for the possible occurrence of counts from secondary pulses. Although the values actually used in the experiment were those derived from System 2 and calculated as C_2 (extending deadtime with livetimer), the results C_1 , C_1' , C_2 , and C_2' were all very close in value. Another check was made by comparing the value of T_{2L} calculated from Eq. [3E] with the electronically measured value. If the deadtimes were appreciably extended, then the values calculated from Eq. [3E] should not agree with the measured livetime, nor should the two livetimer results T_{1L} and T_{2L} agree. When discrepancies were observed in either the livetime or counting rate results, the measurements were investigated as described in Section 4.3 Some Operating Problems.

3.0 PRECISION FILLING OF MEP

It had been established in preliminary measurements that ethyl ether was a good quenching agent for MEP. The amount of ^{85}Kr activity (KrII) to use was selected as a compromise between the opposing criteria of: (a) adequate counting rate for achieving the desired total number of counts at the extreme piston position of closest approach, and (b) avoiding too much deadtime at the other extreme (wide open) piston position. In an attempt to evaluate the wall effect experimentally, measurements were made at various krypton pressures. The same amount of KrII was used in each fill, and the krypton pressure was varied by changing the amount of added "dead" krypton gas which had a small, though non-zero, disintegration rate.

3.1 Vacuum Line for Filling. To minimize potential gas contamination by adsorbed gases, the filling was carried out in a vacuum line constructed entirely of stainless steel tubing and valves except for the calibrated volumes, which were pyrex bulbs. A gas pressure, p , could be measured with a precision quartz-bourdon gauge accurate to the relative error 0.06% at $p = 100$ Torr, smaller relative errors at $p > 100$ Torr, and an absolute error of 0.05 Torr for $p < 100$ Torr (Ruska Digital Pressure Gauge, DDR-6000).

3.2 Calibrated Bulbs. Attached to the manifold with shut-off metal valves were a number of glass bulbs of various volumes which were used to prepare defined gas mixtures for MEP. The volume of each bulb was calibrated by first evacuating it and then filling it with pure xenon gas (<0.05% impurity) to a measured pressure (approximately 1 atm) and finally freezing the gas quantitatively into a preweighed and evacuated small, strong stainless steel vessel cooled to liquid nitrogen temperature. After warming, the steel vessel was weighed again with a semimicro balance, the gain in weight corresponding to the xenon previously contained in the bulb. A water bath surrounding the bulb was used to control the xenon gas temperature, which was measured to better than 0.2°C (0.07% accuracy). The weight gain, xenon temperature, and pressure sufficed to give the bulb volume, using the value of xenon density as 5.2527 g/l at 740 Torr, 25°C. This method of measuring the standard volumes is described in Ref. 7.

Each bulb was calibrated several times; the average values are shown in Table 1 (error indicated is standard error of the mean). The results in the table indicate that our goal of <0.1% error was satisfied. Each of the flasks #1 and #2 was designed to be a mixing chamber to homogenize the mixtures prepared. Each had a long freeze-down finger at the bottom which could be cooled with liquid or solid nitrogen to allow condensation of gases from the other flasks on the manifold.

3.3 Filling Gases. Quench Gas. The ethyl ether was taken from a fresh unopened container of "absolute ether". The purification process was designed to remove traces of water and oxygen. The water was removed by mixing the ether with Linde molecular sieve #4 which had been activated by heating under vacuum. After three days of drying, the ether-sieve mix was cooled with liquid nitrogen and pumped. It was then warmed up and the ether transferred to the storage bulb by distillation and cryogenic cooling. Final traces of oxygen were removed by about eight stages of freezing the ether with liquid nitrogen and pumping it. The process was complete when the vacuum gauge showed no pressure over the frozen ether.

3.4 Filling Gases. Krypton Gas. KrII, the krypton gas supply containing ^{85}Kr , and the "dead" krypton supply (KrIII) were analyzed mass spectrometrically and found to have the compositions given in Table 2. The nominally "dead" KrIII was used for diluting the specific activity of KrII.

Table 1

MEASURED BULB VOLUMES (ml)

Flask	Volume	Flask	Volume	Flask	Volume
#1	1057.44 $\pm$ 0.10	A	151.45 $\pm$ 0.11	C	219.99 $\pm$ 0.08
#2	1075.88 $\pm$ 0.03	B	215.96 $\pm$ 0.01	D	229.47 $\pm$ 0.03

Table 2

 CHEMICAL COMPOSITION OF ^{85}Kr GAS SAMPLE (KrII)
 AND "DEAD" KRYPTON (KrIII)

Species	Percent	
	KrII	KrIII
Kr	> 99.89	> 99.8
H ₂	< .05	< .07
He	--	< .01
H ₂ O	.02	.02
N ₂	< .04	< .03
CO		
O ₂	< .003	< .003
Ar		.06
CO ₂	< .002	< .006
Other	N.D.*	N.D.*

*N.D. = Not detected.

It actually had a slight amount of activity which had to be accounted for in calculating the results. This was measured in ordinary cylindrical GM counters whose counting efficiencies were calibrated with KrII. KrIII specific activity was measured as 0.0131 dpm/ml (at adopted conditions, 760 Torr, 25°C), as of June 1, 1983.

3.5 Filling Procedure. The amount of KrII to be used in the counting gas mixture was fixed by the counting rate desired. This was chosen to be about 1000 cpm when the piston was pulled all the way out, a compromise between a rate which would yield too much deadtime and a rate which would yield an adequate number of counts in a reasonable time.

The gas mixture was prepared by introducing pre-calculated pressures of the three components ethyl ether, KrII and KrIII into separate calibrated bulbs, and then transferring two of the components into the third bulb (either Flask #1 or Flask #2), where all three components were mixed. Since the mixture could not be transferred quantitatively from the mixing bulb into MEP, it was necessary to prepare more mixture than needed for MEP, allowing for the gas necessarily to be left in the manifold and the mixing bulb. Thus the ratio*

$$R_{vol} = \frac{\text{Vol. (mixing bulb)} + \text{Vol. (manifold)} + \text{vol. (MEP)}}{\text{Vol. (MEP)}}$$

was measured by entering some gas into MEP at a measured pressure P_1 , closing the valve to MEP, pumping out the manifold and mixing bulb, opening the MEP valve, and measuring the final pressure P_2 . Since $R_{vol} = P_1/P_2$, the pressure of gas in the mixing bulb needed to fill MEP to a desired pressure P_d was readily determined. When Flask #2 was used as the mixing bulb, $R_{vol} = 2.365$, so that the mixing bulb was filled to an initial pressure $(2.365)P_d$.

Two of the smaller flasks and a "one liter" flask were attached to the manifold and thermostated in the water bath. Then the desired amount of ether was transferred to one of the small flasks and the pressure precisely measured; this was chosen so that the ether pressure constituted a specified percentage (often 10%) of the total fill pressure. After clearing the

*The MEP volume included the active counting volume as well as the "dead" volumes on the outside of both insulator discs, including the extension volume containing the precision rule.

manifold, a defined pressure of KrII was entered into the other small flask and measured in the same way, KrIII was placed into the one liter flask, say Flask #2. After the manifold was pumped out, the pressure gauge was shut off, all three flasks opened to the manifold and liquid nitrogen placed around the freezedown finger of Flask #2. Solidifying the nitrogen by pumping on it condenses the ether and all krypton into the cold finger. After pressure measurement with the gauge indicated the residual pressure to be <0.01 mm, Flask #2 was isolated from the manifold.

Mixing the gases was carried out by setting up convection currents through heating the freezedown finger and cooling the top of Flask #2. After thermal equilibrium was reestablished, the gas was expanded into the evacuated and air-thermostated MEP. The gas pressure was measured and the MEP body temperature was measured at three points with platinum thermometers; readings agreed to $<0.2^{\circ}\text{C}$.

4. OPERATION OF MEP

4.1 Placement. MEP was mounted within a tomb shield with 8 inches of steel wall and inside an anticoincidence counter made of slabs of 2 inch thick solid plastic scintillator surrounding the counter space. The tomb, when closed, acted as an effective air thermostat; the temperature at various points of MEP, as measured with several Pt thermometers, was uniform to $<0.2^{\circ}\text{C}$.

The scintillator anticoincidence shield served to decrease background from very penetrating particles (muons). A muon passing through MEP would also pass through the plastic shield, resulting in an ionization discharge within MEP and a light pulse in the scintillator. Photomultipliers placed around the edges of the scintillator slabs picked up the light pulse, and a triggered anticoincidence circuit suppressed the counting of the MEP discharge which was time-coincident with this pulse. The total counting rate (KrII + muon) determined the total deadtime of the counting system, the muon rate being about 20% of that of KrII.

4.2 Setting Operating Parameters. Rather than measuring plateau characteristics, a tedious procedure, a suitable high voltage was determined by measuring the pulse height distribution from MEP. As the high voltage was increased, attainment of the Geiger region was indicated by the fact that the

pulse heights became essentially uniform. The operating point was then selected to be 25 to 50 volts higher. The actual voltage used depended upon the ethyl ether and krypton pressures, going up when either was increased. Typical values are shown in Table 3. At a constant high voltage value, measurements were made at five positions of the piston, at precision scale readings 43, 36, 29, 22, and 15 cm.

The pulse height discriminator setting was set at one-fourth the full pulse height P_F . The choice of $\frac{1}{4} P_f$ was made after observing the pulse analysis distribution of the MEP output pulses. This distribution was approximately a bell-shaped curve centering on P_F , with some tailing toward the lower pulse-height region. The tail essentially disappeared at a pulse height level well above $\frac{1}{2} P_f$. As a precaution, the discriminator was set at $\frac{1}{4} P_f$; extrapolation of the tail indicated that MEP pulses as small as $\frac{1}{4} P_f$ were of vanishingly small concentration. As expected from the nature of GM tube discharge propagation, P_F varied linearly with the "length" of the GM tube, so that P_F decreased with shorter lengths. Since the form of the distribution remained the same, the absolute value of the discriminator setting was readjusted to $\frac{1}{4} P_f$ for each piston setting.

4.3 Some Operating Problems. At times, MEP behaved erratically. This was observed either as a spread in measured values (e.g., successive "accurate" counts might differ by 3 to 4 σ (i.e., where σ = Poisson count error), or the pulse height distribution of GM pulses would show an unusually wide spread. In addition, there might be a significant difference between the corrected elapsed time and the livetime. When these observations were made, the tube was dismantled and examined carefully. In each case, something unusual was observed. For example, the wire and wall were found to have some black material deposited on them. This may have been due to decomposition products of the many GM discharges or possibly to foreign material transferred from the manifold. The system was then cleaned and reassembled with a new wire and a filter installed at the MEP inlet. Another time, it was suspected that the MEP shut-off valve carried with the MEP gas inlet may have leaked slightly. Mass spectrometric (M.S.) analysis indicated the presence of oxygen. After the single shut-off valve was replaced by two valves in series, M.S. tests at the end of each run showed no signs of leakage.

Table 3

MEP COUNTING DATA FOR FILL NO. 1

High Voltage	1550 V				
Piston Position (cm)*	43	36	29.00	22.00	15.07
Livetime (min)	1300	1300	1299	1300	4101.1
Total Count	1242604	1032255	826008	619211	1303054
CPM	955.85	794.04	635.88	476.32	317.73
	± 0.86	± 0.78	± 0.70	± 0.61	± 0.28
CPM (Decay Corr)	957.20	796.15	637.45	477.41	318.24
	± 0.86	± 0.78	± 0.70	± 0.61	± 0.28

Least Squares Slope: 22.870 ± 0.0243

Chi-Square of Fit: 2.86; Normalized Chi-Square**: -0.06

High Voltage	1600 V				
Piston Position (cm)*	43.00	36.00	29.00	22.00	15.07
Livetime (min)	950	900	900	900	3700
Total Count	917287	723849	580856	435256	1206879
CPM	965.57	804.28	645.40	483.52	326.18
	± 1.01	± 0.95	± 0.85	± 0.73	± 0.30
CPM (Decay Corr)	965.74	805.27	646.08	484.64	326.41
	± 1.01	± 0.95	± 0.85	± 0.73	± 0.30

Least Squares Slope: 22.895 ± 0.0189

Chi-Square of Fit: 1.24; Normalized Chi-Square**: -0.72

High Voltage	1625 V				
Piston Position (cm)*	43.00	36.00	29.00	22.00	15.07
Livetime (min)	1300	1300	1300	4100	3900
Total Count	1258689	1046401	838346	1992056	1277432
CPM	968.22	804.92	644.88	485.87	327.38
	± 0.86	± 0.79	± 0.70	± 0.34	± 0.29
CPM (Decay Corr)	971.48	807.77	647.28	487.97	328.53
	± 0.86	± 0.79	± 0.70	± 0.34	± 0.29

Least Squares Slope: 22.963 ± 0.0457

Chi-Square of Fit: 9.75; Normalized Chi-Square**: 2.75

*Position of fiducial marker relative to precision scale.

**Gaussian approximation of Chi-Square = $[\text{Chi-Square} - \text{DF}] / \sqrt{2 \text{ DF}}$, where DF = degrees of freedom. DF = 3 for these data.

At another disassembly, small bends were found on the center wire. It was hypothesized that this might have been due to crossover of the wire on the take-up reel. After a small spiral groove was machined on the reel surface as a guide for the wire, this problem did not recur.

5.0 MEASUREMENTS

5.1 Method. Since plateau plots (count rate vs. high voltage) had slopes of the order of ~1% per 100 V, it was not appropriate to use a single voltage operating point. Instead, measurement was made of counting rate vs. piston position at several high voltage values for which the output pulse distribution was single peaked. At any particular voltage setting, a plot of these data yielded a straight line, as expected, and the slope value was calculated by weighted least squares. The slope represented an estimate of the disintegration rate of a gas volume at the center of MEP consisting of a cylindrical slab 1 cm high and with the MEP radius. The same procedure was used for several high voltage values, and the slopes were averaged. Fig. 3 illustrates the experimental results for one particular gas mixture. Although the counting rate at a particular piston position increased a little with the high voltage value, the slope was the same at the several voltages.

The standard reference conditions used were: pressure, 760 Torr; temperature, 25°C; date, June 1, 1983. Since measurements were made over a period of a year, the ^{85}Kr decay was corrected to the reference date, using the value $T_{1/2} = 10.75$ yr (see Appendix B).

After a particular mixture was placed in MEP and the operating voltage selected, counting was carried on at this voltage at five piston positions, mostly separated by 7 cm, i.e., at rule readings 43, 36, 29, 22, and 15.46 or 15.55 cm. The last interval was constrained by the limits of available movement. For some measurements, calculation was carried out by slightly extrapolating the "15 cm" measurement to 15.00. In other measurements, the actual position was used in the calculation. MEP was counted at each piston position for one day; 300 minutes during the working day and 1000 minutes overnight. Because its counting rate was least, the "15 cm" position was left for Friday, so that it gave a 300 minute count and an approximately 3800 minute count over the weekend. The short and long counts were checked for consistency and then averaged. Time interval distribution measurements were

Fig. 3. Measurements with MEP

Lines fitted to data of counting rate vs. relative piston position for Gas Fill No. 1. (Data in Table 3.)

SLOPES: $S_A = 22.87$ Ct. Rate per Cm

$S_B = 22.90$

$S_C = 22.99$

GAS FILL No. 1: Partial Pressure (standard conditions)

KrII = 20.18 mm, Ether = 7.51%

Total Pressure = 201.8 mm

HIGH VOLTAGE: A 1550 V

B 1600 V

C 1625 V

simultaneously made with TIDA (Time Interval Distribution Analyzer). The pulse-height distribution of the output pulses was examined to check that the distribution remained suitably narrow. The short-long count check, the TIDA analysis and the output distribution check helped to alert the analyst when MEP was misbehaving in the fashion described earlier.

As an example of the results, Table 3 shows the data for Fill No. 1 with a nominal date of June 1, 1983, which was the filling date; counting results for the various fills have all been decay-corrected to this date. Decay corrections were made from the actual counting dates. The ($\pm$) values are Poisson standard deviations.

5.2 Calculation Method. Weighted Least Squares. At each high voltage, CPM is plotted against piston position. The counting rate at each point was weighted with its counting error and a weighted least squares algorithm was used to calculate the slope. The error estimates quoted for each fitted slope were derived from the data scatter about the fitted line (the so-called "external" error). The result for each fill was taken as the weighted average of the slopes for the separate high voltages used; the weights were derived from the external slope error.

5.3 Alternative Calculation Method. Minimum Chi-Square. It is interesting to compute the end effect implied by the approximate model used in Fig. 1 and Eq. [2A]. If the end effect did not exist, then the counting rate per centimeter would be calculated as

$$\text{specific counting rate} = \frac{\text{total counting rate}}{\text{actual counter length}} \quad [4A]$$

With the end effect to be allowed for, this may be modified as

$$\text{specific counting rate} = \frac{\text{total counting rate}}{\text{effective counter length}} \quad [4B]$$

where

$$\text{effective counter length} = \text{actual counter length} - \Delta \quad [4C]$$

In this formulation, it is assumed that the counting rate per unit length is constant within the effective counter length and zero in the region Δ .

The evaluation of Δ is not possible with only one value of actual counter length, but the joint results of counting with several lengths can yield an estimate of Δ , provided we assume that Δ is the same for all counter lengths -- the basic assumption of our measurement. It is shown in Appendix C how this may be most efficiently carried out with the nonlinear estimation process of minimum chi-square.

Details of the calculation results are omitted, since the resulting values of counting rate per unit length (from Eqn. [4B]) are identical to those derived from the weighted least squares estimates given in Table 6. An interesting sidelight derived from the calculations indicates that Δ decreases with increasing high voltage. If the specific counting rate is to be independent of high voltage, such a variation is necessary, since in a GM tube voltage plateau, the total counting rate does increase slowly with the high voltage value (see Eqn. [4B]). One may interpret this increased rate to be due to the more effective penetration of the electric field into the end region (at higher voltage), which increases the likelihood of forming countable pulses. In the approximate model, this shortens the length of the "dead" region Δ .

5.4 Background and "Dead" Krypton. Contributing to the counting rate of KrII in MEP were the counts arising from the background and from the added diluent "dead" krypton (KrIII). The counting rate of each varied with the piston position.

As mentioned above, ordinary GM tubes with counting efficiency calibrated with KrII were used to measure the specific activity of KrIII as 0.0131 ($\pm 8\%$) dpm/ml (at 760 Torr, 25°C) as of June 1, 1983. Since the slope was essentially the disintegration rate of an MEP cylindrical section 1 cm high and of MEP radius, the contribution to a measured slope arising from the KrIII portion of a fill mixture was calculable from the specific activity, pressure, and temperature of the added KrIII.

To evaluate the effect of background on the slope measurement, some fills were made with KrIII and ether and with argon and ether. For the former, the background was taken as the residual after subtracting the expected low specific activity effect. Since argon contains no activity, its measured slope could be taken as the background effect. The two methods gave essentially the same result, which checks the apriori assumption that the

background slope was not affected by the nature of the gas filling. Table 4 gives data for some measurements of the background.

5.5 The Gas Fills. The various gas mixtures (fills) are listed in Table 5. Since the MEP radius is 5.066/2 cm, the counter volume for 1 cm length is 20.1567 cm³. The krypton volume (760 Torr, 25°C) contained in this length is

$$V(\text{Kr}) = \frac{20.1567}{760} p(\text{Kr}) = 0.02652 p(\text{Kr})$$

where $p(\text{Kr})$ is the krypton pressure (Torr) referred to 25°C.

5.6 Measurement Results. The data and final values for each fill are given in Table 6. The unweighted average specific activity (as of June 1, 1983) was measured to be

$$\text{Uncorrected S.A.} = 42.15_2 \pm 0.09_3 \text{ dpm/ml}$$

where the error indicated is the standard error of the mean. Then

$$\left(\frac{\text{error}}{\text{average}} \right) \times 100 = 0.22\%$$

This value has not been corrected for the small wall effect, whose magnitude is calculated in Appendix A.

As shown in this appendix, the wall effect magnitude is proportional to $1/p$, where p is the gas pressure. It was originally intended that the wall effect would be determined empirically through measurements made at several values of p . A plot of "apparent specific activity" vs $1/p$ would then yield an intercept (at $p = \infty$) which would yield the wall effect. However, the behavior of MEP as a GM tube deteriorated with increasing p , so this approach was abandoned, apparently requiring considerable further study to develop stable operating conditions.

This kind of empirical approach had also been tried by several other investigators (Ref. 4,5,6,8,9) either by using the varied- p approach or by varying counter diameters at constant length. The radioactivities measured had low energy betas: ^3H , $E_{\text{max}} = 18.6 \text{ kev}$; ^{35}S , $E_{\text{max}} = 167 \text{ kev}$; ^{14}C , $E_{\text{max}} = 156 \text{ kev}$. Results from several experiments showed that for these isotopes, the wall effect was not observable, within their experimental errors. From the

Table 4

MEASUREMENTS OF SLOPE BACKGROUND

<u>Gas Fill</u>	<u>High Voltage</u>	<u>Pressure of Rare Gas (mm)</u>	<u>Measured Slope</u>	<u>Calculated Slope Due to Radioactive Gas*</u>	<u>Net Slope (Background)</u>
KrIII	1700 V	201.44	0.153	0.070	0.083
KrIII	1750 V	201.44	0.146	0.070	0.076
KrIII	1875 V	250.37	0.172	0.087	0.085
A	1700 V	258	0.097	--	0.097
Average:					0.085
Standard Deviation:					0.009

*Decay corrected to date of measurement.

Table 5

PROPERTIES OF VARIOUS GAS FILLS

Fill No.	Date	Total Gas Press. (25°C)	Percent KrII (a)	Press. of KrII (mm) (25°C)	Percent KrIII (a)	Press. of KrIII (mm) (25°C)	V ₂ ml/cm (b,c)	V ₃ ml/cm (b,d)	Spec. Act. (e) KrIII dpm/ml	dpm of Gas Fill due to 1 cm of KrIII	
										As of 6/1/83	At Count Time
1	6/1/83	201.84	10.00%	20.18	82.49%	166.50	0.5352	4.416	0.0131	0.058	0.058
2	9/12/83	201.84	10.07	20.32	82.43	166.38	0.5389	4.413	0.0129	0.058	0.057
3	10/10/83	205.48	10.00	20.55	82.51	169.54	0.5450	4.497	0.0128	0.059	0.058
4	10/26/83	277.40	7.223	20.18	85.21	236.38	0.5352	6.269	0.0128	0.082	0.080
5	3/1/84	204.51	10.00	20.45	80.004	163.62	0.5424	4.340	0.0125	0.057	0.054
6	3/19/84	335.41	6.169	20.69	83.829	281.17	0.5487	7.457	0.0124	0.098	0.093
7	4/6/84	325.93	6.267	20.43	83.734	272.91	0.5418	7.238	0.0124	0.095	0.090
8	4/26/84	324.99	6.264	20.36	83.738	272.14	0.5400	7.218	0.0124	0.095	0.089
9	6/20/84	199.90	10.00	19.99	80.00	159.92	0.5302	4.241	0.0122	0.056	0.052
10	7/6/84	224.70	8.923	20.05	81.068	182.16	0.5318	4.831	0.0122	0.063	0.059

(a) Percent by pressure.

(b) Volume (at standard conditions) contained in slab of MEP: Cylindrical section 1 cm high and with MEP radius.

(c) V₂ = Vol (KrII).

(d) V₃ = Vol (KrIII).

(e) At standard conditions, 760 mm, 25°C.

Table 6

SPECIFIC ACTIVITY RESULTS FOR KrII (Uncorrected for Wall Effect)

Fill	P(KrII) (a)	V ₂ (b)	Total Pressure (a)	Percent Ether	n(c)	Average Slope	Error (g)	Bkgnd. + "Dead" Kr (d)	Corr. Av. Slope (e)	Specific Activity (f)
1	20.18	0.5352	201.84	7.51%	3	22.89 ₄	0.01 ₃	0.143	22.75 ₁	42.50 ₉
2	20.32	0.5389	201.84	7.50	2	22.64 ₉	0.02 ₃	0.142	22.50 ₇	41.76 ₅
3	20.55	0.5450	205.48	7.50	2	22.88 ₈	0.01 ₁	0.143	22.74 ₅	41.73 ₄
4	20.18	0.5352	277.40	7.51	3	22.80 ₃	0.02 ₆	0.165	22.63 ₈	42.29 ₈
5	20.45	0.5424	204.51	10.00	2	22.80 ₇	0.01 ₈	0.139	22.66 ₈	41.79 ₂
6	20.69	0.5487	335.41	10.00	3	23.29 ₂	0.01 ₈	0.178	23.11 ₄	42.12 ₅
7	20.43	0.5418	325.93	10.00	1	23.04 ₀	0.05 ₂	0.175	22.86 ₅	42.20 ₂
8	20.36	0.5400	324.99	10.00	2	23.05 ₂	0.03 ₀	0.174	22.87 ₈	42.36 ₆
9	19.99	0.5302	199.90	10.00	2	22.59 ₆	0.04 ₀	0.137	22.46 ₂	42.35 ₉
10	20.05	0.5318	224.70	10.01	3	22.67 ₈	0.04 ₂	0.144	22.53 ₄	42.37 ₃

(a) Torr at 25°C.

(b) Volume of KrII (reduced to standard conditions, 760 Torr, 25°C) in 1 cm length of MEP.

(c) Number of slope measurements for the fill. Each slope was measured at a different high voltage value.

(d) Background slope = 0.085.

(e) (Average Slope) - (Bkgnd. + "dead" Kr) = Corrected Average Slope.

(f) S.A. = (Corr. Av. Slope)/V₂

(g) Standard Error of the Mean.

discussion in Appendix A, such a low result would be expected, since, as is evident from Equation [1], the probability for no ionization in a path length L is very small if σ_p is large, which is the case for low energy electrons.

The wall-effect calculations in Appendix A gave values which indicated that efficiency factors varied from 0.9967 to 0.9942 depending upon the fill pressure. The specific activity results of Table 6 are corrected for these wall effect values in Table 7. The corrected specific activity (as of June 1, 1983) is then

$$\text{Corrected S.A.} = 42.34_7 \pm 0.09_8$$

This is an unweighted average and the indicated error is the standard error of the mean.

5.7 Comparison of Uncorrected (for Wall Effect) Measurement with the KrI Two-Counter Calibration. As discussed earlier, the two-counter value used for the KrII specific activity was ultimately based upon the KrI two-counter 1954 calibration, through several intermediate ratio comparisons. At each comparison, the specific activity of the current standard was decay-corrected with the then accepted best value for the ^{85}Kr half-life. The value used was 10.70 yr between January 1, 1954 and January 1, 1969 and then 10.76 yr to the fiducial date June 1, 1983. The decay corrections have been recalculated for these periods using the currently preferred value of 10.75 yr (Appendix B). This correction resulted in only a modest change in the presumed value of KrII at June 1, 1983, namely, approximately 0.73%. The corrected result was 42.04, slightly larger than the hitherto presumed value of 41.88 dpm/ml. This quantity was also uncorrected for the wall effect. The original measurement data for the 2-counter calibration was no longer available, so that no error could be assigned to the data scatter contribution to the error in the 42.04 value. However, from Appendix B, it is evident that the assigned error in the ^{85}Kr half-life value results in another contribution to the error in the 42.04 value since a ^{85}Kr decay correction was made from January 1, 1954 to June 1, 1983; the one sigma error contribution is 0.35%.

The MEP and two-counter results are considerably closer than might have been expected. Thus

$$\frac{\text{Uncorrected MEP Value}}{\text{2-Counter Value}} = \frac{42.15_2}{42.04} = 1.002_7$$

Table 7

SPECIFIC ACTIVITY RESULTS FOR KrII (Corrected for Wall Effect)

Fill	Total Pressure ^(a)	Percent Ether	Uncorrected Specific Activity	P_0 ^(b)	Corrected ^(c) Specific Activity
1	201.84	7.51%	42.50 ₉	0.0058	42.75 ₇
2	201.84	7.50	41.76 ₅	0.0058	42.00 ₉
3	205.48	7.50	41.73 ₄	0.0057	41.97 ₃
4	277.40	7.51	42.29 ₈	0.0042	42.47 ₇
5	204.51	10.00	41.79 ₂	0.0054	41.93 ₀
6	335.41	10.00	42.12 ₅	0.0033	42.26 ₄
7	325.93	10.00	42.20 ₂	0.0034	42.34 ₆
8	324.99	10.00	42.36 ₆	0.0034	42.51 ₀
9	199.90	10.00	42.35 ₉	0.0056	42.59 ₈
10	224.70	10.01	42.37 ₃	0.0050	42.58 ₆

(a) Torr at 25°C

(b) Probability of no ionization from ^{85}Kr beta-particles (Table A, Appendix A).(c) $[\text{Uncorrected Specific Activity}]/[1-P_0]$.

For the wall-effect corrected result,

$$\frac{\text{Corrected MEP Value}}{\text{2-Counter Value}} = \frac{42.345}{42.04} = 1.0073$$

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APPENDIX A

CALCULATION OF "WALL-EFFECT" IN GM TUBES1.0 INTRODUCTION

The magnitude of the wall-effect in a GM counter is derived from the fraction of beta-decays within the counter (neglecting end-effects) which do not result in at least one slow electron (per decay) moving through the gas. A slow electron can have its trajectory changed by the electric field present so that it accelerates towards the wire, which process results in a countable discharge. An energetic beta particle (say, one exceeding several hundred ev), if allowed to travel indefinitely into the gas, will always create free electrons by ionization. These free secondaries lose energy by further ionization or by other inelastic collisions, and eventually slow down enough to be taken over by the electric field. The wall effect, then, occurs when a released beta particle heads towards the wall; if it is released close enough to the wall, it may enter and stay in the wall before it can ionize any gas molecule. Except at the ends of the GM tube, formation of one slow free electron suffices to allow the creation of a countable discharge,* so a pre-condition for a wall effect event is that the beta-particle does not cause any ionization.

However, a contribution to the wall effect does not always occur when a beta decay occurs close to the wall with a beta-particle headed towards the wall. Several processes must all not occur if a slow free electron is not to be formed as the result of a beta decay:

- A. A shakeoff electron must not be released. When ^{85}Kr ($Z=36$) decays to ^{85}Rb ($Z=37$), the change in nuclear charge causes a change in the binding energies of all the orbital electrons. Release of this "rearrangement" energy may result in ionization of one or more of the orbital electrons. When released, these are called shakeoff electrons. The valence shell is the most probable origin of shakeoff electrons and such an electron

*The calculation here neglects the counter end-effects, i.e., the interaction of end-effect and wall-effect is taken to be of higher order and to be neglected.

generally has a very low energy, of the order of a few ev, hence will almost certainly result in a counter discharge. Even shakeoff electrons from inner shells have energies (<20 kev) much lower than the average beta-particle energy, hence have very high cross sections for ionization, and will almost certainly cause a discharge. The probability of creating shakeoff electrons is the same for every decay event and does not depend upon the beta-particle energy, since the transition energy is the same for each decay.

- B. The decay electron must not create any ionization. Since the maximum energy of a secondary ionization electron is about one-half the beta-particle energy, and the energy distribution of the secondaries rises at least as steeply as $1/E_{\text{sec}}^2$ at the low energy end,¹⁹ a secondary electron, if formed, is most probably of low energy and almost certainly creates a discharge. As discussed below, it will be evident that only quite energetic beta-particles have a low probability of creating an ionization in the path lengths of significance.
- C. The decay electron must penetrate into the wall and must not backscatter. If it does backscatter, its trajectory will have a long path-length in the gas and it will almost certainly create ionization.

A quantitative examination of these processes offers an approach to estimation of the non-counting probability P_0 . Thus,

$$P_0 = P_A P_I \quad [\text{A-1A}]$$

where P_A is the probability of a decay without shakeoff and P_I is the probability that a beta-particle will not ionize on the way to the wall and will not backscatter (processes (B) and (C)).

The calculation of $P(B)$, the probability of process (B), can be carried out in a straightforward manner, since the functions involved have been measured (or calculated) and are available in published material. In analyzing this process, one takes into account the random distribution of the source point in the gas volume, the random distribution of θ the angle of emission, the random distribution of E , the energy of the beta-particle

(varying randomly according to the beta-spectrum distribution), and the random distribution of ionization events along the beta-particle trajectory. On the other hand, $P(C)$, the probability of process (C), although calculable in principle, is not readily calculated in detail, since the necessary data is not accessible.

Even if $P(C)$ were readily calculable, an exact calculation could not be made through independent evaluations of $P(B)$ and $P(C)$. The differential probability for a single emission not ionizing is

$$P(I_{\text{diff}}) dv = P(B_{\text{diff}}) \cdot P(C_{\text{diff}}) dv$$

where $dv = dV dE d\theta$ (volume, energy, emission angle). The quantity $P(B) = \int P(B_{\text{diff}}) dv$, multiplied by $P(C) = \int P(C_{\text{diff}}) dv$ is not equal to the value for the joint process, i.e.,

$$P_I = \int P(B_{\text{diff}}) \cdot P(C_{\text{diff}}) dv \neq P(B) \cdot P(C)$$

The terms in the joint integral are not separable into the separate integrals, since each differential element contains the common variables (θ , E), and hence are interdependent. Nevertheless, because the probability of no electron released, P_0 , is known to be small, we shall use an approximate relation

$$P_I \approx P(B) P(C) \quad [A-1B]$$

$P(B)$ is here calculated in detail, starting with the differential relations and integrating over the pertinent variables. An integral value of $P(C)$ is taken from empirical measurement results.

Since $P(A)$ does not depend upon what happens to the beta-particle, it is a separable quantity, as in Eq. [A-1]. Then

$$P_0 \approx P_A P(B) P(C) \quad [A-1C]$$

The integrated shakeoff probability has been calculated theoretically^{20,21} with results whose accuracy improves as the ratio of beta-decay

energy to the K binding energy increases. For the decay of ^{85}Kr , the theoretical value 0.22 is quite good. Hence, $P_A = 1 - 0.22 = 0.78$.

2.0 CALCULATION OF P(B)

2.1 Geometry of the Calculation. Because end-effects are to be neglected, any circular segment will have the same properties, so only beta-particles emitted within a circular segment (radius R) perpendicular to the tube axis and one cm long are to be considered (Fig. A-1). The effect of the cylindrical curvature is neglected. Fig. A-2A represents a beta-particle emission from point Q, where the projection of the initial path is long the tube axis. Fig. A-2B represents an emission whose projection of path is perpendicular to the axis direction. Paths with intermediate directions lie in between. If $z \ll R$, the approximation of treating the case in Fig. A-2B as though it were like Fig. A-2A is not important. In calculating the probability of ionization, the beta-particle path is considered straight and in the direction of the original emission. This is not actually the case for a lengthy path, since a beta-particle, as it interacts with gas molecules, may meander considerably. However, since only the first ionizing collision is of concern, and since the initial beta particle energy E is too large to allow much deflection in a weak non-ionizing collision, this approximation is a good one. The geometry of the calculation is shown in Fig. A-3. θ is the angle of L relative to the plane perpendicular to the distance vector z. From the assumption of a flat wall, the same geometry will apply for any path having the angle θ over the azimuthal angular (ϕ) range 0 to 2π .

Then

$$L = \frac{z}{\sin \theta}, \quad 0 < \phi < 2\pi \quad [\text{A-2}]$$

2.2 Outline of the Calculation. The computation involves the following steps:

- (1) Calculate the mean partial emission rate of beta particles (of a given energy E) from a volume element at a distance z from the wall into a differential solid angle fixed by the emission angle θ . Emission from the volume element is taken as isotropic.

Fig. A-1. Integration Volume. Cylindrical box made of circular segment perpendicular to the counter tube axis (radius = tube radius) and straight portion one cm along tube wall.

Fig. A-2A. Beta particle emitted from point Q along path L, which is parallel to the wire

Fig. A-2B. Beta particle emitted from point Q along path L in direction perpendicular to that in Fig. A-2A

Fig. A-3. Geometry of Calculation

- (2) Calculate the mean partial emission rate (at energy E) of those beta particles so emitted that do not cause an ionization event.
- (3) Integrate over all emission angles with an upward component (at constant E and z).
- (4) Integrate along the tube axis direction a one cm distance and around a circle of radius (R-z).
- (5) Integrate over all z (at constant E).
- (6) Integrate over all E (using beta-spectrum intensity as weighting function), yielding mean rate of non-ionizing emissions.
- (7) P(B) is the ratio of (6) to the total mean rate of all emissions in the volume of Fig. A-1.

2.3 Statistical Considerations. For a path length L cm through a pure gas, the mean number of primary ionizations from one beta-particle of energy E is

$$C = N_a L \sigma_p \quad [A-3]$$

where: N_a = number of molecules of this gas per cm^3
 $\sigma_p = \sigma_p(E)$ = cross section for primary ionization in the pure gas.

Primary ionization refers to the emission of an electron from a gas molecule resulting from collision with beta-particle. Secondary electrons resulting from collision of the primary liberated electron (or its descendants) with other molecules are not counted. Since σ_p is a function of E, of interest is the case where L is small enough so that the mean number of primary ionizations is very small, so that E (hence, σ_p) remains approximately constant over the distance L. Then the number of primary ionizations formed in the length L is Poisson-distributed, with mean C. Hence,

$$\text{Prob (k ion pairs in L)} = \frac{e^{-C} C^k}{k!} \quad [A-4]$$

and

$$P(0) = e^{-C} = e^{-N_a L \sigma_p} \quad [A-5A]$$

which was earlier written in Eq. [1].*

If several gases are mixed, the results are combined as follows:

With: N_a' = no. of molecules of Species I per cm^3
 N_a'' = no. of molecules of Species II per cm^3
 $\sigma_p' = \sigma_p'(E)$ = ionization cross section of Species I
 $\sigma_p'' = \sigma_p''(E)$ = ionization cross section of Species II

then the probabilities of no primary ionization in molecules of either gas species are

$$P_1(0) = e^{-N_a' \sigma_p' L} \quad \text{and} \quad P_2(0) = e^{-N_a'' \sigma_p'' L} \quad [\text{A-5B}]$$

Since these probabilities are independent, the probability of no ionization in molecules of either species is:

$$P(0) = P_1(0) P_2(0) = e^{-[(N_a' \sigma_p') + N_a'' \sigma_p''] L} = e^{-\Gamma L} \quad [\text{A-5C}]$$

$$\text{with } \Gamma = N_a' \sigma_p' + N_a'' \sigma_p'' \quad [\text{A-5D}]$$

In the following calculations, certain considerations concerning a class of radioactive emissions are implicit:

- (1) The disintegration rate is Poisson distributed with mean rate S
- (2) If a subclass α is considered with probability of occurrence P_α , then the number of disintegrations of the α -type is Poisson distributed with mean rate Sp_α and the probability of k events (in unit time) is Poisson with mean Sp_α .
- (3) If a class β is considered with probability P_β , then k_β is also Poisson with mean Sp_β .
- (4) If classes α and β are independent, then if $k_{\alpha\beta} = k_\alpha + k_\beta$ = number of both α and β events, $k_{\alpha\beta}$ is Poisson with mean $Sp_\alpha + Sp_\beta = S(p_\alpha + p_\beta)$.

* $P(0)$ can have a significant value only if C is small (e.g., if $C = 7$, $e^{-7} = 0.0009$). Hence, since the initial beta particle energy is measured in many kilovolts, and since energetic electrons released in primary ionization are rare, the approximation $\sigma_p \approx \text{constant}$ is a good one.

2.4 Calculation

Let W = mean emission rate (per unit time) of beta-particles from a unit volume.

$I(E)$ = Distribution function for β -particles as a function of energy E ; normalized to unity.

$W_E = W \cdot I(E) dE$ = mean emission rate from betas in interval E to $E+dE$.

$W_{EV} = W_E dV = W \cdot I(E) dE dV$ = mean emission rate in the dE and dV intervals, with dV = differential volume at emission point Q .

P_θ = probability of emission from point Q into $d\theta$ interval at angle θ , over all azimuth, into the upper hemisphere (Fig. A-4).

$$P_\theta = \frac{2\pi \cos\theta d\theta}{4\pi} = \frac{1}{2} \cos\theta d\theta, \quad 0 \leq \theta \leq \frac{\pi}{2} \quad [A-6]$$

The mean number of particles emitted from the $dEdV$ interval into the angle θ to $\theta + d\theta$ is Poisson-distributed with mean

$$M_1 = W_{EV} P_\theta = W \cdot I(E) P_\theta dV dE \quad [A-7]$$

Let $dV = dz(dydx)$

where z = vertical distance from wall

x = distance in the direction of the axis (wire)

y = distance along the perpendicular to x -axis along a circle of radius $R - z$.

From M_1 , the mean number of emissions from the intervals dz and dE into the $d\theta$ interval at θ is:

$$M_2(E, y, x, z, \theta) = W \cdot I(E) \left(\frac{1}{2} \cos\theta d\theta\right) (dydx) dz dE \quad [A-8]$$

The probability of an emission of energy E with no ion pairs into an angle θ from a pt Q a distance z from the wall is (from Eq. [A-5C] and Fig. A-3):

$$P(0) = e^{-\Gamma \frac{z}{\sin \theta}} \quad [A-9]$$

Fig. A-4. Emission into $d\theta$ interval

Hence, the mean number of emissions with no primary ionizations is at given (θ, z, E, y, x)

$$M_3(E, y, x, z, \theta) = W \cdot I(E) \left(\frac{1}{2} \cos \theta \right) d\theta e^{-\Gamma \frac{z}{\sin \theta}} (dy dx) dz dE \quad [A-10]$$

Since the event of emitting into one $d\theta$ interval is independent of emitting into another, summing over all θ gives the total mean number at constant (z, E, x, y) :

$$M_4(E, x, y, z) = \frac{1}{2} W \cdot I(E) (dy dx) dz dE A(z, E) \quad [A-10a]$$

with (since $\Gamma = \Gamma(E)$)

$$A(z, E) = \int_0^{\frac{1}{2}\pi} e^{-(\Gamma z / \sin \theta)} \cos \theta d\theta \quad [A-10b]$$

It may be noted that if Γz is very small,

$$A(z, E) \cong 1 \quad (\text{for } \Gamma z \cong 0) \quad [A-11a]$$

If Γz is very large,

$$A(z, E) \cong 0 \quad (\text{for } \Gamma z \gg 1) \quad [A-11b]$$

$A(z, E)$ is very small except when z is quite close to the wall.

Several of the variables may be analytically integrated. Thus, x is integrated from 0 to 1 (i.e., for a 1 cm cylindrical section) and y is integrated around the circle $(R - z)$ in diameter, so

$$\int_0^1 \int_{\text{circle}} dx dy = 2\pi(R - z) \approx 2\pi R \quad [A-11c]$$

since* $R \gg z$.

Then, at constant (z, E) , the mean number of emissions with no primary ionizations is:

$$M_5(z, E) = \frac{1}{2} W \cdot I(E) (2\pi R) dz \cdot A(z, E) dE \quad [A-12]$$

*The entire integrand is very small except for small z (from Eq. [A-11b]).

Integrating over z (which cannot exceed R , the mean number of emissions with no primary ionizations (at constant E) is:

$$M_6(E) = \pi R W \cdot I(E) \int_0^R A(z, E) dz$$

To a good approximation (since $A(z, E)$ is very small except for small z),

$$\int_0^R A(z, E) dz \approx \int_0^\infty A(z, E) dz$$

Then

$$\begin{aligned} M_6(E) &= \pi R W \cdot I(E) \int_0^\infty dz \int_0^{\frac{1}{2\pi}} e^{-(\Gamma z / \sin \theta)} \cos \theta d\theta \\ &= \pi R W \cdot I(E) \int_0^{\frac{1}{2\pi}} \cos \theta \left[\int_0^\infty e^{-\Gamma z / \sin \theta} dz \right] d\theta \end{aligned}$$

Let: $w = \frac{\Gamma z}{\sin \theta}$, $dw = \frac{\Gamma}{\sin \theta} dz$, $dz = \frac{\sin \theta}{\Gamma} dw$; then, with $w = 0$ at $z = 0$ and $w = \infty$ at $z = \infty$,

$$M_6(E) = \pi R W \cdot I(E) \frac{1}{\Gamma} \left(\int_0^{\frac{1}{2\pi}} \cos \theta \sin \theta d\theta \right) \left(\int_0^\infty e^{-w} dw \right) dE$$

The first integral = $\frac{1}{2}$ and the second one = 1.

Then

$$M_6(E) = \pi R W \cdot I(E) \frac{1}{2\Gamma} dE \quad [A-13]$$

Finally, integrating over E , the mean number of emissions with no primary ionizations is:

$$M_7 = \frac{1}{2} \pi R W \int_0^{E_{\max}} \frac{I(E)}{\Gamma(E)} dE \quad [A-14]$$

Since the total number of emissions (T.E.) of all types from the volume in Fig. A-1 is W multiplied by the volume, T.E. = $W (\pi R^2)$, and the probability of emissions with no primary ionization is

$$P(0) = \frac{M_7}{T.E.} = \frac{1}{2R} \int_0^{E_{\max}} \frac{I(E)}{\Gamma(E)} dE \quad [A-15]$$

N_a , the number of molecules per cm^3 , can also be written as (since the reference temperature used in this report is 25°C):

$$N_a = N_L \frac{p}{760} \frac{273.15}{298.15} = 3.2393 \times 10^{16} p = C_F p \quad [\text{A-16}]$$

where N_L = Loschmidt's number = 2.6872×10^{19} per cm^3 = number of molecules in 1 cm^3 of an ideal gas at 0°C . 760 Torr and p is measured in Torr. Then, for a mixture of two gases,

$$\Gamma = N'_a \sigma'_p + N''_a \sigma''_p = C_F p [f' \sigma'_p + f'' \sigma''_p] \quad [\text{A-17A}]$$

with

$$f' = \frac{N'_a}{N'_a + N''_a} = \frac{p'}{p} \quad \text{and} \quad f'' = \frac{N''_a}{N'_a + N''_a} = \frac{p''}{p} \quad [\text{A-17B}]$$

where p' and p'' are the partial pressures of the respective gases, and $p = p' + p''$ is the total pressure. Then Eq. [A-15] may be stated as

$$P(0) = \frac{1}{(2C_F R) p} \int_0^{E_{\max}} \frac{I(E)}{H(E)} dE \quad [\text{A-18A}]$$

with

$$H(E) = f' \sigma'_p(E) + f'' \sigma''_p(E) \quad [\text{A-18B}]$$

For a given gas sample, all the quantities in the question are constants, so

$$P(0) \propto \frac{1}{p} \quad [\text{A-18C}]$$

2.5 Computer Computation

Computer computation of Eq. [A-18A] is carried out through the approximation

$$\int_0^{E_{\max}} \frac{I(E)}{H(E)} dE \approx \sum F_i \frac{I(E_i)}{H(E_i)} \Delta E_i \quad [\text{A-19}]$$

F_i is a factor for the i -th energy interval; it is discussed below. ΔE_i -values are chosen so that the integrand is almost constant within each ΔE -

region; ΔE_i is small for small E and increases for larger E -values. The function $I(E)$, i.e., the normalized beta-spectrum of ^{85}Kr , is shown in Fig. A-5.

Values of σ_p were calculated from the relation given in Ref. 12:

$$\sigma_p = (1.874 \times 10^{-20} \text{ cm}^2) C \left[\frac{M^2}{C} x_1 + x_2 \right] \quad [\text{A-20A}]$$

$$\text{with } x_1 = \frac{1}{\beta^2} \ln \left[\frac{\beta^2}{1-\beta^2} \right] - 1, \quad x_2 = \frac{1}{\beta^2} \quad [\text{A-20B}]$$

where $\beta = v/c$, v being the velocity of the beta-particle, and c = velocity of light; β is related to the particle kinetic energy E (ev) with the relativistic relation

$$\beta^2 = 1 - \frac{1}{\left(\frac{E}{mc^2} + 1 \right)^2} \quad [\text{A-20C}]$$

where mc^2 is the energy equivalent of the electron mass, $mc^2 = 5.11 \times 10^5$ ev.

In Ref. 12, σ_p was measured for a number of gases for single electron energies varying from 100 to 2700 kev. The results were fitted to Eq. [A-20A] yielding values of M^2 and C . A linearizing plot showed the results to be valid down to 10 kev, and quite good down to a few Kev (Ref. 13). Since σ_p is very large for lower energies, the accuracy of the σ_p -values is not important in the lower energy range, where the integrand in Eq. [A-18A] is very small. Eq. [A-20A] is not valid in the lowest energy range (i.e., for small β), and since there is practically no contribution to the sum in Eq. [A-19] below 5 kev, the terms in the sum are set equal to zero for $E_i \leq 5$ kev. Thus,

$$F_i = 0 \text{ for } E_i \leq 5 \text{ kev, and } F_i = 1 \text{ for } E_i > 5 \text{ kev}$$

For krypton, the parameter values are: $M^2 = 6.09 \pm 2.6\%$, $C = 52.38 \pm 0.4\%$

For n-pentane, these are: $M^2 = 18.41 \pm 1.8\%$, $C = 184.8 \pm 0.5\%$

The σ_p -values for diethyl ether were calculated from other experimental results reported in Ref. 14. Unlike the experiment in Ref. 12, this work yielded only relative values of the ionization cross-sections for a variety of

Fig. A-5. Normalized Beta Spectrum of ⁸⁵Kr, $I(E)$ vs. E (from Ref. 11). The numerical scale of $I(E)$ was chosen so that $\int_0^{E_{\max}} I(E) dE = 1$.

gases (using the beta-particles of ^{85}Kr). The relative values were found to be closely the same over the energy range 75 ev to 500 kev. Then

$$\frac{\sigma_p(\text{diethyl ether})}{\sigma_p(\text{n-pentane})} = 0.930$$

For each E_1 -value, Eq. [A-20C] gives the β_1 -value for use in Eq. [A-20B]; hence, the $\sigma_p(E_1)$ values for pentane and krypton are derived from Eq. [A-20A]. The ratio from Ref. 14 then provides $\sigma_p(E_1)$ for diethyl ether. From these $\sigma_p(E_1)$ -values and the pressures of the gases, $H(E_1)$ is evaluated (Eq. [A-18B]) and the sum calculated as in Eq. [A-19].

Fig. A-6 presents a log-log plot of σ_p vs E for krypton and for ethyl ether. The effective σ_p for each mixture is calculated at each E -value with Eq. [A-18B].

Fig. A-7 gives a corresponding log-log plot of relative values of the integrand $I(E)/H(E)$, from Eq. [A-18A] or Eq. [A-19], as a function of E . This graph illustrates the influence of the various beta-particle energies on the wall effect calculation. Below the energy at the peak (at ~400 kev), the integrand increases rapidly with increasing E , because $I(E)$ is almost constant or slowly changing in this energy range, and $\sigma_p(E)$ is a rapidly decreasing function of E . Above the peak, $\sigma_p(E)$ decreases slowly with increasing E , while $I(E)$ becomes a rapidly decreasing function of E . It is evident that most of the contribution to the integral arises from beta particles in the approximate energy range 150 to 550 kev.

$P(B)$ was evaluated for all ten fills with Eq. [A-18A], using the approximation in Eq. [A-19] for the integral. Results are shown in Section 3.0 (see Table A). $P(B)$ is the value of P_0 calculated with neglect of the back-scattering and shakeoff effects.

3.0 DETERMINATION OF $P(C)$

Ideally, information on the back scattering effect would be most useful if its magnitude were available as a function of θ and E . In the absence of such information, we use empirical values measured for other purposes which give backscattering values integrated over θ and E .

Fig. A-6. Primary Ionization Cross Section (cm^2) as a Function of E for Krypton and Ethyl Ether

Fig. A-7. Relative Value of the Integrand Quantity $I(E)/H(E)$ as a Function of E

These measurements were made in experiments designed to determine the counting efficiency of a GM tube or proportional counter for measuring the emission rates from thin solid beta-emitting radioactive samples mounted on metal plates. In one configuration (α), a sample plate with deposited radioactivity was placed immediately outside and parallel to the very thin window of an "end-window" GM tube. The radioactivity was very close to the active discharge region, the average solid angle (relative to 4π) subtended at the sample being $\sim 25\%$. It was found that the counting efficiency exceeded the value calculable from the solid angle because some of the beta-particles emitted into the plate were backscattered through the window and were counted. Calibration of the backscattering magnitude was made by comparing the measured counting rate of activity mounted on a metal plate with that of the same amount of activity mounted on a very thin plastic film, too thin to cause backscattering. Another variation to this approach involved placing metal plates immediately below the activity deposited on a thin plastic film.

In another configuration (β), the sample plates were placed inside a flow proportional counter, in which the samples rested on or near one wall, and the relative solid angle subtended was $\sim 50\%$. The saturation counting rates were measured for backings of various atomic numbers Z and for several maximum beta energies and compared to the known disintegration rates.

The general results of these measurements were:

- (1) For a given atomic number (Z) substrate, the fraction of beta-particles emitted into the substrate that are backscattered (Fr_b) increases (with increasing substrate thickness) to a saturation value that depends mildly on the beta-particle maximum energy and more sharply upon Z . [For our application, the MEP wall thickness (of copper) is larger than the saturation value.]
- (2) Saturation Fr_b increases with the atomic number of the substrate.
- (3) The value of Fr_b at the saturation substrate thickness (at a given Z) is about the same at various values of the maximum beta-energy.
- (4) The back-scattered beta-particles are degraded in energy.
- (5) For a source outside of the counter, the angular distribution of the backscattered beta-particles is anisotropic, tending to favor the direction perpendicular to the substrate. This implies that the

percentage increase in counting rate ascribable to backscattering tends to be higher in Configuration (α) than in Configuration (β).

It seems most appropriate to use the data taken with Configuration (β), although this is not of great importance considering the variation in results between various experiments. From the various experimental values (Ref. 15-18), we take a value of ~40% for the saturation backscattering value for copper with a proposed

$$1 - P(C) = 0.40 \pm 25\%$$

$$\text{or } P(C) = 0.60 \pm 17\%$$

[A-22]

4.0 COMPUTATION RESULTS

The P_A -value from Section 1.0 and the calculation results from Sections 2.0 and 3.0 gave values of $P(B)$ and P_0 listed in Table A-1.

5.0 DISCUSSION

It is evident from Figs. A-6 and A-7 that a significant wall-effect (greater than 0.25 to 1.0%) will not occur if the beta spectrum is predominantly low energy. Thus, for a counter of about 2 inch diameter operating at about 200 Torr, and with a gas having primary ionization cross-sections comparable to those in Fig. A-6, a low energy beta-emitter would not show a sizeable wall effect. Such emitters would include ^3H ($E_{\text{max}} = 18.6 \text{ kev}$), ^{14}C ($E_{\text{max}} = 156 \text{ kev}$), and ^{35}S ($E_{\text{max}} = 167 \text{ kev}$). Only if the gas were pure hydrogen or helium (which have very low primary ionization cross sections) and the beta energy exceeded 100 kev would the wall effect be significant.

A further effect is worth noting. Implicit in the calculation above is the assumption that when a primary ionization occurs, it will always yield an electron which will be accelerated towards the wire and result in a countable discharge. This process requires that the liberated electron either slow down sufficiently so that the electric field can influence its trajectory, or that it liberate another electron in an ionization collision which can be slowed down enough to allow the formation of a discharge process. However, if the liberated electron is of high energy, and is headed towards the wall, it has a moderate probability of not slowing down or causing another ionization, i.e., it will not cause a discharge or liberate another electron before collision

with the wall. If this effect is significant, then the above calculation underestimates the wall-effect.

Except for the lowest energy liberated electrons, there is indeed a high correlation between the directions of the beta-particle and liberated electron trajectories. However, losing a discharge from an ionization electron can occur with significant probability only if the liberated electron energy E_{Le} is itself high enough. This situation can occur only with very low probability, because the liberated electron energy distribution only goes out to a maximum electron energy of one-half the beta-particle energy and the distribution has an intensity variation proportional to $1/E_{Le}^2$ with a cutoff at very low energy.¹⁹ Thus, most of the electrons liberated in primary ionization, even from the highest energy beta-particles, have low energy, and, as is evident from Fig. A-6, these will almost certainly ionize the gas.

Table A-1
COMPUTED WALL-EFFECT RESULTS

Fill No.	Total Gas Pressure (Torr)	Percent Ethyl Ether	P(B)	P_0 (a)
1	201.84	7.51%	0.00965	0.0045
2	201.84	7.50	0.00965	0.0045
3	205.48	7.50	0.00948	0.0044
4	277.40	7.51	0.00702	0.0033
5	204.51	10.00	0.00908	0.0042
6	335.41	10.00	0.00553	0.0026
7	325.93	10.00	0.00569	0.0027
8	324.99	10.00	0.00571	0.0027
9	199.90	10.00	0.00928	0.0044
10	224.70	10.01	0.00826	0.0039

(a) P_0 given in Eq. [A-1C], with $P_A = 0.78$, $P(C) = 0.6$, from Eq. [A-22].

APPENDIX B
ON THE ^{85}Kr HALF-LIFE

At present, the most reliable half-life value for ^{85}Kr is taken as

$$T_{\frac{1}{2}}(^{85}\text{Kr}) = 10.75 \pm 0.02 \text{ (one sigma)}.$$

This is a weighted average of the value 10.76 yr from Ref. 22 and 10.72 yr from Ref. 23 as recalculated in Ref. 24.

In Ref. 22, six samples were followed for 0.8 to 0.95 of a half-life, and a separate half-life value was calculated for each sample, using least squares. The half-life values from the six samples were then averaged; the one sigma error quoted (0.008 yr) was an estimate of the standard error of the mean for the six numbers ($s(x)/\sqrt{6}$). Each sample was contained in a very thin-walled sealed glass vial and β -particles were counted in an internal bell-shaped proportional counter. The β -particles emitted from metal uranium samples were used as constant reference standards; these allowed correction for the slight effect of changes in gas multiplication over a period of years. Both types of samples were counted in an alternate fashion using an automatic sample changer.

The result in Ref. 23 involved the measurement of bremsstrahlung from a single sealed ^{85}Kr sample measured in a total ionization chamber with a reentrant thimble; beta-particles did not enter the chamber. The bremsstrahlung consisted of low energy photons generated by the interaction of the ^{85}Kr betas with the sample holder and the walls of the chamber thimble, which completely absorbed the betas. Gamma-rays from a sealed ^{226}Ra source were used to provide an almost constant reference activity. Measurements were followed for 1.5 half-lives, but breakage required that the glass sample holder be replaced with a plastic one halfway through the measurement. The air-containing ionization chamber was open to the atmosphere, so the gas content was corrected for temperature and pressure variation. Least squares was used to fit the decay curve with a half-life value. The error quoted (standard deviation = 0.008 yr) was that derived from the least-squares formalism applied to the edited data from the single sample.

The results of both experiments have been combined here to give the value 10.75 yr, greater weight being given to Ref. 22, because of the redundancy of samples and the greater reliability of counting β -particles in an internal counter as compared to measuring ionization from the relatively low energy bremsstrahlung photons with an external chamber. The errors calculated from each experiment (0.008 yr) were much smaller than the difference in half-life values (~ 0.04 yr); the estimated error has therefore been arbitrarily enlarged to 0.02 yr. Other measured half-life values of ^{85}Kr have not been included in the averaging, because the results had much larger errors.

APPENDIX CON APPLYING MINIMUM CHI-SQUARE ESTIMATION TO SPECIFIC ACTIVITY DATA
DERIVED FROM POISSON-DISTRIBUTED MEASUREMENTS1.0 Gaussian, Poisson, and Chi-Square Distributions

The material in Section 1.0 is available in many advanced statistics texts (e.g. Ref. 25).

1.1 Gaussian

Measurements are represented by the random variable x , with the probability distribution, with $-\infty < x < \infty$,

$$P(x) dx = \frac{1}{\sigma \sqrt{2\pi}} \exp \left[-\frac{(x-M)^2}{2\sigma^2} \right] dx \quad [C-1A]$$

$$\text{with Mean } (x) = M, \text{ Var}(x) = \sigma^2(x) \quad [C-1B]$$

where $\text{var}(x)$ = variance of x = (standard deviation)².

1.2 Standardized Gaussian

If the data are standardized relative to the mean as origin and using σ as a scale factor, then

$$z = \frac{x - M}{\sigma(x)}, \text{ with } \sigma(x) = [\text{Var}(x)]^{\frac{1}{2}} \quad [C-2A]$$

$$P(z)dz = \frac{1}{\sqrt{2\pi}} \exp \left[-\frac{1}{2} z^2 \right] dz \quad [C-2B]$$

$$\text{with Mean } (z) = 0, \text{ Var}(z) = 1 \quad [C-2C]$$

1.3 Chi-Square

Let $x_1, x_2, \dots, x_n$ represent n independent Gaussian distributed measurements, where the (mean, variance) of the j th one is (M_j, σ_j^2) . The M_j 's and σ_j^2 's may all differ or some (or all) may be the same. When standardized, they all have the same distribution (Eqn. [C-2B]). Then the sum

$$\chi^2 = \sum_{i=1}^n z_i^2 \quad [C-3A]$$

is a random variable which has the distribution (with $0 < \chi^2 < \infty$)

$$P(\chi^2) d\chi^2 = \frac{1}{2^{D/2} \Gamma(D/2)} (\chi^2)^{D/2-1} \exp \left[-\frac{1}{2}\chi^2 \right] d\chi^2 \quad [C-3B]$$

$$\text{Mean}(\chi^2) = D, \text{ Var}(\chi^2) = 2D \quad [C-3C]$$

where D = degrees of freedom and is n in the sum in Eq. [C-3A] and Γ is the gamma function.

When the differences in the standardized variables z_i are based on differences from $\hat{M}$, an estimate of M , instead of M itself, i.e.,

$$z_i = \frac{x_i - \hat{M}}{\sigma} \quad [C-4]$$

then the sum in Eq. [C-3A] still has the chi-square distribution in Eq. [C-3B], but with $D = n-1$. The estimate $\hat{M}$, however, must be an efficient estimator. An example is the case where x_1, x_2, x_3 , and $\dots x_n$ are all independent measurements from the same Gaussian with (M, σ^2) . Then $\hat{M} = \bar{x} = \sum x_i / n$, and

$$\chi^2 = \sum_{i=1}^n \frac{(x_i - \bar{x})^2}{\sigma^2} \quad [C-5]$$

has the distribution in Eqn. [C-3B] with $D = n-1$.

1.4 Functions of Gaussian Variables

In general, if x_i is a random variable with distribution Eqn. [C-1A], then the random variable $w_i = f(x_i)$ does not have a Gaussian distribution. For example, when z_i in Eqn. [C-2A] is Gaussian, z_i^2 is not Gaussian, but has a chi-square distribution in Eqn. [C-3B] with $D = 1$. However, if the ratio σ_i/M_i is quite small, then over the region in which $P(x)$ is not very small*, the distribution of w is approximately Gaussian, with

*Roughly in the region $M - 3\sigma < x < M + 3\sigma$.

$$\text{Mean}(w) \approx f(M) \text{ and } \text{Var}(w) \approx \left[\left(\frac{df}{dx} \right)_M \right]^2 \text{Var}(x) \quad [\text{C-6}]$$

where the derivative is evaluated at $x = M$.

1.5 Poisson

In radioactive decay processes, if k = the number of disintegrations in a fixed time interval T , then k has a Poisson distribution

$$P(k) = \frac{e^{-M} M^k}{k!}, \quad \text{Mean}(k) = M, \quad [\text{C-7}]$$

where k is an integer and $\text{Var}(k) = M$. If M is large enough, say >100 , the Poisson distribution may be approximated by the Gaussian in Eqn. [C-1A], with $\text{Mean}(k) = \text{Var}(k) = M$ and standard deviation $= \sqrt{M}$ and k is treated approximately as a continuous random variable. If $U = g(k)$, then, if $\sqrt{M}/M = 1/\sqrt{M}$ is small, then U may be treated as approximately Gaussian with

$$\text{Mean}(U) \approx g(M) \text{ and} \quad [\text{C-8A}]$$

$$\text{Var}(U) \approx \left(\frac{dg}{dk} \right)^2 \text{Var}(k) = \left(\frac{dg}{dk} \right)^2 M \quad [\text{C-8B}]$$

1.6 Chi-Square with Poisson-based Data

If the M -values are large enough, then standardized variables based on Poisson-distributed quantities may be combined into the chi-square form. There is one modification, however, resulting from the fact that for Poisson data, $\text{Var}(x) = \text{Mean}(x) = M$. Thus, if the x_i 's in Eqn. [C-5] are Poisson variables, then χ^2 may be written in the form

$$\chi^2 = \sum_{i=1}^n \frac{(x_i - \bar{x})^2}{\bar{x}} \quad [\text{C-9}]$$

since (for the Poisson), $\sigma^2 = M$, hence $\bar{x}$ is shown as an estimate of $\text{Var}(x)$ as well as of M . χ^2 in Eqn. [C-9] can be shown to have the approximate chi-square distribution with $(n-1)$ degrees of freedom.

2.0 MINIMUM CHI-SQUARE FOR ESTIMATION

Consider n random variables x_i where each x_i is a random variable which is a function of a Poisson random variable k_i . Then, approximately, the x_i 's are Gaussian and the sum of the squared standardized quantities is chi-square distributed. Suppose each x_i is a function of the same unknown quantity, V , and we wish to estimate the V -value from some combination of the n values of x_i . There are various efficient schemes for such estimation, among which are maximum likelihood and minimum chi-square. Under the latter, the best estimate is that value of V which minimizes the sum representing chi-square.

3.0 APPLICATION OF MINIMUM CHI-SQUARE TO EVALUATION OF SPECIFIC ACTIVITY FROM MEP MEASUREMENTS

To formulate the appropriate form of chi-square = Q , the following definitions apply to one gas fill at a given high voltage value

- S = disintegration rate of that portion of the gas fill contained in the squat cylindrical box one cm along the tube axis and with MEP radius (see Fig. A-1).
- n = number of different measurements made with different tube lengths.
- L_i = length of tube as indicated by position of precision scale. This was calibrated at the initial disassembly of the counter.
- S_i = estimate of S , calculated from measurement at i -th position.
- Δ_1 = plus (or minus) deviation of each L_i from the actual piston-to-piston length due to nonreproducibility arising from gasket thickness variation in reassembly. Δ_1 is unknown for the various reassemblies, but is small because the gold gasket itself was <1 mm diameter. Although its value is not known, Δ_1 is the same for all sequential fills in which reassembly did not occur.
- Δ_2 = end effect decrement in length for a particular fill. It is a function of high voltage, but not of tube length.
- Δ = $\Delta_2 \pm \Delta_1$, depending on sign of Δ_1
- EL_i = $L_i - \Delta$ = effective length of tube at the i th piston position.
- N_i = total counts measured at i th position.
- T_i = livetime of i th counting period.
- c_i = N_i/T_i = measured counting rate at i th position
- t_i = elapsed time of i th count since reference date.

$D_i = e^{\lambda t_i}$ = decay correction to reference date

$C_i = D_i c_i$ = decay corrected counting rate.

$$S_i = \frac{C_i}{EL_i} = \frac{D_i c_i}{EL_i} = \frac{D_i}{EL_i} \left(\frac{N_i}{T_i} \right) = \alpha_i N_i, \quad [C-10]$$

with $\alpha_i = \frac{D_i}{T_i EL_i}$. Then,

$$\frac{\sigma^2(S_i)}{[\text{mean}(S_i)]^2} = \frac{\sigma^2(S_i)}{S_i^2} = \frac{\sigma^2(N_i)}{[\text{mean}(N_i)]^2} = \frac{\text{mean}(N_i)}{[\text{mean}(N_i)]^2} = \frac{1}{N_i} \quad [C-11A]$$

or

$$\sigma^2(S_i) = \frac{S_i^2}{N_i} \quad [C-11B]$$

Then the statistical weight for S_i is

$$w_i = \frac{1}{\sigma^2(S_i)} = \frac{N_i}{S_i^2} \quad [C-11C]$$

The weighted average as an estimate of S is

$$\bar{S} = \frac{1}{W} \sum_{i=1}^n w_i S_i, \quad \text{where } W = \sum_{i=1}^n w_i \quad [C-12]$$

Then Q , which is chi-square distributed with $DF = n-1$, is

$$Q = \sum_{i=1}^n \frac{[S_i - \text{estimate}(\text{mean}(S_i))]^2}{\text{estimate}[\sigma^2(S_i)]}$$

Hence,

$$Q = \sum_{i=1}^n \frac{(S_i - \bar{S})^2}{S_i^2 / N_i} = \sum_{i=1}^n w_i (S_i - \bar{S})^2 \quad [C-13]$$

It is evident that, since the value of each S_i depends upon the Δ -value, then Q is a function of Δ . The best estimate $\hat{\Delta}$ is the Δ -value which minimizes Q . This may be found by choosing a starting value for Δ , computing Q and then using a converging step procedure to iterate to a minimum value. Given the Δ -estimate, as the Δ -value used in the final Q calculation, the corresponding

$\bar{S}$ -value is the estimate of S . As noted in the body of the report, this value is the same as that derived by using weighted least squares to calculate the slope of the line representing the counting rate as a function of $\bar{U}_1$.

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