THE DESIGN AND OPERATION OF A DIFFERENTIAL IONIZATION
CHAMBER SYSTEM FOR HALFLIFE MEASUREMENTS

by

RICHARD JOHN HEINRICH SNEED

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TABLE OF CONTENTS

INTRODUCTION

Statement of Purpose

Many methods have been introduced to determine the life-times of isotopes ranging from a 10^{-10} seconds to billions of years. Each of these methods, was designed for a particular time range. Much work has been done lately to improve the methods used in the extreme regions as a result of the present interest in the characteristics of the short lived, artificially created isotopes and in the information which could be gained from long-lived isotopes as for example, in the field of Archeology or Geophysics. There has been a definite need for improved measurements in the intermediate ranges as illustrated by recent investigations of Eu^{154}. Several investigators taking measurements over periods of from six months to four years determined its half life as $5 - 8$ years, $20 +$ years, 5.4 years, and 16 ± 4 years, (7, 15, 9, 11).

The purpose of this thesis is to describe the design and the operation of a differential method which is particularly applicable to medium-long ranges both for absolute and comparative measurements. The first recorded application of the differential ionization method was made by Rutherford (Tobailem, 34). Segre (27) first recommended this method particularly for half life measurements in the medium-long to long region. Segre (27) himself, and later Bainbridge et al. (1) and Bouchez et al. (5) were more specifically interested in detecting the very minute changes in half life which were apparently brought about by

a change in the chemical state of isotopes. Tobailem (34) very recently
published a number of half lives determined by a method similar to that
suggested by Segre (27) and quite analagous to the method described in
this thesis, which clearly shows the advantages and the reliability of
this apparatus. Most previous differential chamber systems relied on
rather complex electronic equipment. The emphasis in the design here
described was on simplicity of circuitry and operation, with no loss of
accuracy and speed.

Summary of Some of the Methods Used in Determining
Half Lives of All Magnitudes

Very Short Half Lives. (10^{-10} to 10^{-3} seconds). The methods used
in this region have been designed to determine the time delay between
the advent of an isotope and its disintegration. Each of these events
was recorded by detecting the radio-active emission accompanying them.
For example, Ra-C was found to decay into a Ra-C' recoil atom after
emitting a beta particle. The decay of Ra-C' into Ra-D was in turn
signaled by an alpha emission. Consequently, the time between the oc-
currence of the beta particle and that of the alpha particle represents
the life time of Ra-C'. As early as 1924, Jacobsen (10) made use of
these phenomena. He measured the density of alpha particles along, and
perpendicular to the path of the recoil atoms of Ra-C' and plotted this
density versus the distance from the source. Calculating the recoil ve-
locity of Ra-C' as 8.4×10^5 cm/sec from the known masses and the momentum

of the emitted beta particle and estimating the mean path of the recoil atoms from the observations led to an approximate half life of 10^{-6} seconds. Jacobsen (Rowland, 23) improved this method later by detecting the beta and alpha particles with individual geiger tubes. The impulses were fed, into a moving-iron mirror-oscillograph system of mutually perpendicular mirrors. Lightbeams from each mirror were reflected onto a slowly moving film. He was able to calculate a half life of $2X10^{-4}$ seconds from the delay time observed between beta particle signals and alpha particle signals, which was in very good agreement with the presently accepted values of about $1.6X10^{-4}$ seconds (31).

The modern approach to measurements of very short half lives brought about the increased application of coincidence circuitry. Rowland (23) described three methods to detect the delay between signals arising at the recorder (counter or oscilloscope) (Plate I, Fig. 1). In the first one, accredited to Rotblat (Rowland, 23) advantage was taken of lengthening, in steps, the pulse due to the initial particle, e.g. beta particle in case of Ra-C' measurements. The increase of coincidences with pulse lengthening was recorded and this produced a curve representing the inverse of the normal decay curve. Corresponding to the finite length of the original pulses half life measurements of greater than ten microseconds could be undertaken. The second method, introduced by Jacobsen (Rowland, 23), gave a more direct measurement of the delay time. Here, instead of lengthening the initial pulse, the pulse was delayed. Plotting coincidences against delay time produced a curve resembling the decay curve. It was subsequently shown by Newton (18), however, that it

is safe to assume this curve to follow the true decay curve only for relatively large delay times. Newton made an analytical evaluation of the process, taking into account the response of the apparatus to simultaneous emission of particles $P(x)$ and the probability of occurrence of the delayed particles in a time interval of $f(t)dt$, producing the "delayed coincidence resolution curve":

$$F(x) = \int_{-\infty}^{\infty} f(t) \, P\,(x-t) \, dt \quad (1)$$

where x was the artificial delay time and $F(x)$, $P(x)$, $f(t)$ all were normalized to enclosed unit area (Plate I, Fig. 2). For a single decay product present, $f(t) = 0$ for $t < 0$ and $f(t) = \lambda \, e^{-\lambda t}$ for $t > 0$; letting $y = x-t$,

$$F(x) = \lambda \, e^{-\lambda x} \int_{-\infty}^{x} e^{-\lambda y} \, P(y) \, dy \quad (2)$$

and

$$\frac{dF(x)}{dx} = -\lambda \left[P(x) - F(x) \right] \quad (3)$$

or

$$\frac{d \ln}{dx} F(x) = -\lambda \left[1 - \frac{P(x)}{F(x)} \right] \quad (4)$$

(3) indicated the position of the maximum to be at the intersection of $P(x)$ and $F(x)$ and (4) showed how, as $F(x)$ became larger than $P(x)$, i.e. as the delay time became larger, the curve $F(x)$ more closely represented the true decay curve.

In order to perform calculations from points other than at the "tail" of $F(x)$, (3) was integrated from x=A to x=B:

$$\lambda = \left[F(A) - F(B) \right] \Big/ \int_{A}^{B} \left[F(x) - P(x) \right] dx$$

i.e.

$$\lambda = \left[F(A) - F(B) \right] \Big/ \text{Area between x=A, } F(x), \, P(x) \text{ and x=B.}$$

Fig. 1. Illustration of Methods for Measuring Very Short Half Lives
(Ra-C' Measurement) (23)

Legend:

C   Coincidence

NC   No Coincidence

DR   Delay Recorded

(a) Pulse Lengthening Method
        (Beta pulses lengthened 50 microseconds)

(b) Delayed Coincidence Method
        (Beta pulses delayed 50 microseconds)

(c) Oscilloscope Method
        (Beta pulses initiate 200 microsecond oscilloscope
        sweep.)

Fig. 2. Explanatory Graph to Newton's (18) Calculation of the
Delayed Coincidence Distribution.

    P(x)   Response of Apparatus to simultaneous emmission.

    F(x)   Response of Apparatus to delayed coincidences.
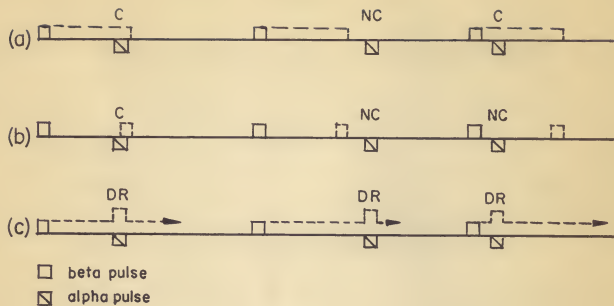
PLATE I

(a) — C — NC — C —

(b) — C — NC — NC —

(c) — DR → — DR → — DR →

□ beta pulse
◩ alpha pulse

Fig. 1

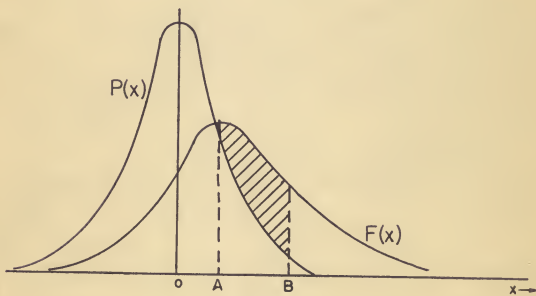
Fig. 2

Bell and Graham (3) applied this method and measured the half life of
83 Kev transition in Yb^{170} as $1.6\pm .2 \times 10^{-9}$ sec. The P(x) coincidence
curve was determined from the coincidences of the beta rays of Au^{198}
and the electrons of the excited state of Hg^{198}. The third method was
developed by Rowland (23) and was closely related to Jacobsen's mirror-
oscillograph method by taking advantage of modern highspeed equipment.
Again, Ra-C' was used for the initial trial. The beta pulses triggered
the single sweep of the oscillograph while the alpha pulses were fed into
the vertical deflection plates. The sweeptime was previously calibrated
and, thus, the position of the alpha pulses on the screen enabled a dir-
ect observation of the delay time. Plotting the number of pulses ob-
served during a certain time interval versus their position or delay
time, produced the decay curve of Ra-C'.

Short Half Lives. (10^{-3} seconds to 10 seconds). Great ingenuity
was also required in devising some of the methods for measuring half
lives in this range. The difficulties which arose were mainly due to
the fact that one was dealing with artificially produced isotopes so
that transport time had to be cut to a minimum and that the time of each
individual observation is long compared to the half life and requires
correction. The first difficulty was overcome by either measuring the
activities directly where they were produced or, where this was impossible,
the isotopes were, e.g., transported directly from the target chamber
which was kept under pressure, by means of evacuated tubes to the obser-
vation station (Sommers and Sherr, 30). In 1911 Moseley and Fajan (17)
measured the half lives of Actinium A (.002 sec.) and Thorium A (.14 sec.)

to a fair degree of accuracy. They deposited Actinium and Thorium emanation on a negatively charged disk which rotated under two stationary ionization boxes. These boxes were so designed as to allow only the alpha particles emitted to enter. Recording the change in ionization current as a function of angular displacement and taking into consideration the angular velocity of the disc permitted the calculation of the investigated half lives. Nearly 30 years later Becker and Gaerttner (2) used a similar method to measure the half life of B^{12}. They mounted a target of boric acid on a disk rotating at 1800 rpm and bombarded it with a well-focused, stationary beam of deuterons. The activity of the target was then measured by electroscopes as a function of angular displacement from the point of bombardment. The resulting value was .022 ± .002 sec. for the half life of B^{12}.

A mathematically and experimentally simple method has been introduced by Wagner (35) which helped overcome the need for corrections with respect to the time required for observing, or counting, activities of short lived isotopes. His only experimental requirement was that all observations (counts) took exactly the same length of time. Mathematically he deduced as follows:

$$- \frac{dN}{dt} = \lambda N = \lambda N_o \, e^{-\lambda t}$$

Considering A as the number of disintegrations during the first period of observation $(t_1 - 0)$

$$A = N_o \int_0^{t_1} e^{-\lambda t} \, dt = N_o (1 - e^{-\lambda t_1})$$

Also, taking B as the number of disintegrations during a second period
of observation, $(t_3-t_2) = (t_1-0)$

$$B = \lambda N_0 \int_{t_2}^{t_3} e^{-\lambda t} = N_0(e^{-\lambda t_2} - e^{-\lambda t_3}) = N_0 e^{-\lambda t_2} \left[1-e^{-\lambda(t_3-t_2)}\right]$$

or $$B = N_0 e^{-\lambda t_2} (1-e^{-\lambda t_1})$$

Therefore $\frac{A}{B} = e^{\lambda t_2}$ And the half life $t_{\frac{1}{2}} = .693 t_2 \ln \frac{A}{B}$

which led to the linear relation:

$$\ln B = \ln A - \frac{.693\ t_2}{t_{\frac{1}{2}}}$$

This method was not only recommended for measuring short half lives but
also for the measurement of very weak activities which might require a
rather long observation time for recording a statistically significant
number of disintegrations.

Medium Short Life Times. (minutes to months). This range could
conveniently and directly be observed over several half lives with a great
degree of accuracy for many years, provided uncontaminated sources were
available. For these measurements the activity or the log of the activi-
ty was plotted versus the time. The time during which the activity
dropped to half its initial value could be read directly off the graph
or the disintegration constant could be evaluated from the slope of the
straight line resulting from semi-log plot. Most commonly, the half life
in this range was determined by measuring the ionization current produced
by the sources in an ionization chamber. This was done by means of an

electroscope, electrometer or other electronic measuring devices. The difficulty here consisted normally in maintaining constant geometry over the required observation time and in providing chambers of sufficient sensitivity to deal with weak sources. The separation of compound activities either before they excited the detector or on the final graph was another problem. For accurate results it has recently appeared desirable to separate complex activities in mass spectrometers (Thode, 32), or beta ray spectrometers (Katz and Lee, 13) and then to evaluate the complex decay curves. Robson (20) applied a method very similar to that of Jacobsen (10) in determining the half life of neutrons. Robson mounted an electron multiplier tube perpendicular to the path of a neutron beam behind the aperture in a negatively charged cylinder enclosing the beam. An earlier source (Snell, et al., 29) had shown experimentally that the neutrons decayed into electrons and protons. Robson attracted the protons to the electron multiplier tube by the negative charge on the cylinder and determined the number of decays per unit time per unit volume. The measurement of the neutron density of the beam and the estimation of that volume of the beam which supplied protons to the tube caused a rather large uncertainty in the result of 12.8 minutes. Robson suspected this factor to yield about an 18 percent error in the result.

Medium Long Half Lives. (months to 50 years). As already indicated above, the methods applied thus far in this range were either very time consuming or quite unreliable. Krisberg, et al. (15) who assigned a "minimum" half life of 20 years to Eu^{154} took direct measurements with a geiger counter for four years, while others (7, 9) evaluating the same

isotope for only several months arrived at half lives of five to eight
years. The use of a mass spectrometer improved these results. Hayden
et al. (9) were able to separate Eu^{154} from Eu^{152}. He inserted activated
europium in the mass spectrometer and, thus, the mass spectrum line of
Eu^{154} consisted of a radio active deposit which was transferred to a
second film and then served as a source for half life measurements. With-
in six months he arrived at a half life of 16 ± 4 years for Eu^{154} and,
simultaneously, he assigned a half life of approximately 13 years to
Eu^{152}. Fajan and Voigt (8) measured the long period in thallium for 300
days with a geiger counter. Two sources of different intensity indicated
a half life of $4.0 \pm .5$ and $3.1 \pm .4$ years, from which these investigators
deduced an average half life of $3.5 \pm .5$ years. The first great step to-
ward precision and speed in the measurement of half lives in the medium
long region was achieved rather recently, when Tobailem (34) published
in January of 1955, (actual measurements apparently date back to 1951 (33)),
several half lives determined with a differential ionization chamber system.
With data taken for 32 days he obtained a value of $5.27 \pm .07$ years for
cobalt 60, and with data taken for 140 days the half life of actinium 227
was determined as 21.6 ± 0.4 years. In the meantime Lockett and Thomas
(16) re-measured Co^{60} in an electroscope for eight months and arrived
at $4.95 \pm .04$ years. They felt their value to be the more accurate one
because of the duration of the measuremnt but they were repudiated by
Kastner and Whyte (12) who measured Co^{60} at intervals from six to twelve
months over a period of three years in what they consider a very stable
ionization chamber which was calibrated by radium at all times and they

found a half life of 5.21 ± .04 years.

Very Long Half Lives. (50 years to 10^{12} years). Up to a limit of perhaps 10^3 years, the differential method should provide a means for direct and relatively fast measurements provided that rather strong sources were available, that the geometry of the system could be kept constant for a period of many months and that a source of practically infinite half life such as uranium is at hand to serve both as a standard to check the chamber sensitivity and as a comparative source.

The methods used thus far gave some surprisingly accurate results with respect to significant numbers assigned, yet in most cases their reliability was hampered by the inclusion of certain suppositions and by indirect measurements which could introduce a chain of experimental errors. The equilibrium method was often used. This method could be applied when minerals were investigated in which a parent activity of long life was in secular equilibrium with the activity of its daughter which was of considerably shorter life. When equilibrium was obtained the decay rates of both isotopes, that is the products of (λN) parent and (λN) daughter were equal. If the relative abundance of each isotope was known and the decay constant of the daughter could be determined, the decay constant of the parent could also be calculated. According to Rowland (23) this method was used in 1907 by Meyer and Achweidler to determine the half life of Ra-D (22 years). This was calculated from a known mass of Ra-D in equilibrium with a known mass of radon (half life 3.825 days). More commonly, the specific activity of the material to be investigated has been determined. The basis for this method was the simple equation

$dN/dt = -\lambda N$. By this method Rutherford and Geiger (26) using a count-
er similar to the Geiger-Muller counter developed later, were able to de-
termine the half life of radium. They found the total number of alpha
particles emitted by one gram of radium to be 3.4×10^{10} per second (25)
and from Avogadros number they calculated the number of atoms present in
one gram of radium. The result was a half life of 1760 years. Both the
equilibrium and specific activity methods have been considerably improved
by the use of the mass spectrograph.

Nier (19) determined the abundance of U^{238}, U^{235}, and U^{234} in a
uranium sample and then applied the equilibrium method to find the half
lives, 4.56×10^{9}, 7.13×10^{8}, and 2.7×10^{5} years respectively. As a basis
for his calculations he used the value of λ for U^{238} determined by
Kovarik (14) and found by careful counting of alpha particles in a pro-
cedure similar to that of Rutherford and Geiger (25). Chamberlain et al.
(6) with a mass spectrograph of higher resolution defined the abundance
ratios somewhat better than Nier (19) and then determined the specific
activity of the uranium sample. Deducting the activity due to U^{238} and
U^{235} he arrived at a value of $(2.35 \pm .14) \times 10^{5}$ years as the half life of
U^{234}. Another method of measuring specific activity was that of deter-
mining the heating effect (Rutherford, et al. 24). For example, it was
shown in a Bunsen-ice calorimeter that one gram of radium emitted heat
at the rate of 100 calories per hour. This energy was expected to re-
present the kinetic energy of all particles emitted if they were absorbed
within the calorimeter. This suggested the equation

$$E = \frac{1}{2} MQ \sum \left[(1 + \frac{M}{m}) \ v2 \right] + E_1$$

Where: E = radiation energy measured, M = mass of the alpha particle, Q = number of alpha particles emitted per second per gram of radium; v = velocity of each group of alpha particles present; m = mass of the recoil atom and E_1 = energy due to beta and gamma rays per gram of radium.

Rutherford et al. (24), having at his disposal the values of v due to radon, radium A, and radium C, which were found from the ranges of the alpha particles emitted by each isotope, and estimating E_1, was able to calculate Q from this equation, as 3.72×10^{10} alpha particles emitted per second per gram of radium, which is somewhat higher than the value he determined electronically (26). As late as 1947, Stout and Jones (Rowland, 23) used this method to evaluate the half life of plutonium 239 as 2.411×10^4 years.

THEORY

The development of the theory for determining medium long to long half lives with differential ionization chambers is rather simple. Effectively it is based on the null method which has been used in many other fields where minute quantities were to be measured. In this case the minute quantity consists of a small time rate of change in the activity of the investigated radio active sources. Depending on whether comparative or absolute measurements are desired the experimental arrangements and the calculations vary slightly. Bainbridge et al. (1), for instance, were interested as stated previously, in the slight change in the half life of an isotope due to the change in electron structure of an atom when in a chemically different compound. Therefore, they were interested

in strictly comparative measurements. For this kind of measurement two sources of nearly equal activity and life time were mounted in two ion chambers of as nearly equal geometry and response as possible. The applied potentials on the chambers were of opposite sign. Accordingly, the ionization currents produced in the chambers were also of opposite sign. If the currents were not exactly matched a small difference current existed. This difference current changed as the activity of one source varied with respect to that of the other. Therefore, the time rate of change of the difference current indicated the difference in decay of the two sources. Mathematically this may be expressed as follows: for two sources - a and b

$$i(t)_a = i(o)_a \, e^{-\lambda a t}; \; i(t)_b = i(o)_b \, e^{-\lambda b t}$$

if: $\lambda_a = \lambda_b + \Delta\lambda$, $i(t)_a - i(t)_b = e^{-\lambda a t} \left[i(o)_a - i(o)_b \, e^{-\Delta\lambda \, t} \right]$ (I) since $\Delta\lambda \, t \ll 1$ for two sources of nearly equal life time, an expansion results in

$$i(t)_a - i(t)_b = e^{-\lambda a t} \left[i(o)_a - i(o)_b + i(o)_b \, \Delta\lambda \, t \right]$$

or - $e^{\lambda a t} \, i(t) = i(o) + i(o)_b \, \Delta\lambda \, t$ (II)

where $i(t)$ and $i(o)$ are the difference currents measured at time $t = t$ and $t = 0$, respectively. For absolute measurements equation (II) can be even more simplified provided the observation time is kept small compared to the life time which is to be measured. As the standard source, an isotope of practically infinite half life as compared to that of the investigated source is used, so that, $\lambda_a \cdot t \ll 1$ and, therefore, $e^{-\lambda a t}$ = 1. Also, $\lambda_a \ll \lambda_b$ and, accordingly $\Delta\lambda = \lambda_b$. Thus (II) becomes the

simple linear relation $i(t) = i(o) + i(o)_b \lambda_b t$ (III). For relatively long observation times, however, it is more accurate to use –

$$\log \frac{i(t)-i(o)}{i(o)_b} = \log \frac{i(o)_a}{i(o)_a-i(o)} = \lambda t \quad (IV)$$

The form using $i(o)_a$ rather than $i(o)_b$ is to be preferred because the former term can always be rechecked and $i(o)$ can be exactly evaluated from the least square method of fitting curves. In the actual experiment ionization potentials rather than currents were measured. But these potentials were measured across the same resistors. Therefore, the currents i could be replaced by the corresponding voltages v in the equation. Bainbridge et al. (1) and Tobailem (34) arrived at somewhat different equations. Bainbridge exchanged the sources after each reading to account for any systematic drift in the chamber system. He arrived at:

$$i = (i_{12}-i_{21}) / 2 = \overline{S} \ (I_1-I_2)_e - \lambda \ t + \overline{S}I_2 e^{-\lambda} t_{\lambda} \lambda t \quad (V)$$

where: i_{12} and i_{21} were the difference currents measured with interchanged sources, $\overline{S} = (S_A + S_B)/2$ was the mean sensitivity of the system, and I_1 and I_2 were the initial ionization currents of either source. In the present design it was felt, that an interchange of sources would bring about greater irregularities than the possible drift of the system. The latter could easily be checked and compensated for by a standard source, as was done by Tobailem (34). In addition, certain design features guarded against major drifts in either chamber alone. Tobailem (34), who was interested in absolute measurements, obtained an equation of the form – $\log \left[I_o / (I_o-i) \right] = -\lambda (t-t_o)$ (VI)

where I_o was the initial current due to the standard source. The reason for the apparent dissimilarity to equation (IV) was due to Tobailem's insistence on starting his time scale with perfectly matched sources at the point $t = o$. He realized, however, that this was experimentally not quite feasible. Still avoiding the term i_o used in (IV) he simply shifted his time scale so that the actually recorded time became $t-t_o$, t_o being a constant.

DESIGN AND OPERATION

The Complete System

In Plate II the complete circuit is schematically represented. From the collecting electrodes of the two ionization chambers a heavy cable was connected directly to a Wulff bifilar electrometer. This cable was completely suspended in air to avoid any leakage to ground between the connections. The cable could be disconnected from either chamber by means of a simple connection. This proved very valuable for isolating either chamber from the system for individual measurements without removing the potential from the isolated chamber. A copper pipe system was connected to the interior of both chambers to maintain equal atmospheric conditions in either chamber (this system was also installed for possible future operation with inert gases rather than atmospheric air as was used to date). The sources were mounted inside the chamber and space was available to shield the two chambers from cross-ionization

when sources of high energy rays were used. Across the electrometer, resistances were mounted to change the current sensitivity of the electrometer as desired. The voltage divider maintained equal potentials of opposite sign on either chamber. A Simpson voltmeter was kept in the circuit at all times during operation to keep the resistance of the supply circuit constant. By means of the rheostat the output of the voltage supply was controlled and a power regulating transformer took care of the line voltage variations. The voltage supply itself was not regulated. Its output was about 6000 volts with no load applied. With a circuit resistance of 25 megohms a potential of about 4000 volts was obtained. Two high capacity filters were added to overcome a rather annoying 60-cycle pickup in the unshielded section of the chamber circuit. Shielding of the collector-electrometer cable had to be omitted because no insulation was available which would have prevented leakage currents. However, besides the 60-cycle pickup from the voltage supply, only body capacity, while adjusting the electrometer, had any noticeable effect. This was corrected by a small copper shield between the electrometer adjustment knob and the ionization current conductor. A battery supplied either positive or negative potential to the auxiliary capacitor of the electrometer. The entire system, except the voltage supply, was mounted on a wooden frame which provided easy access to all parts (Plate III).

EXPLANATION OF PLATE II
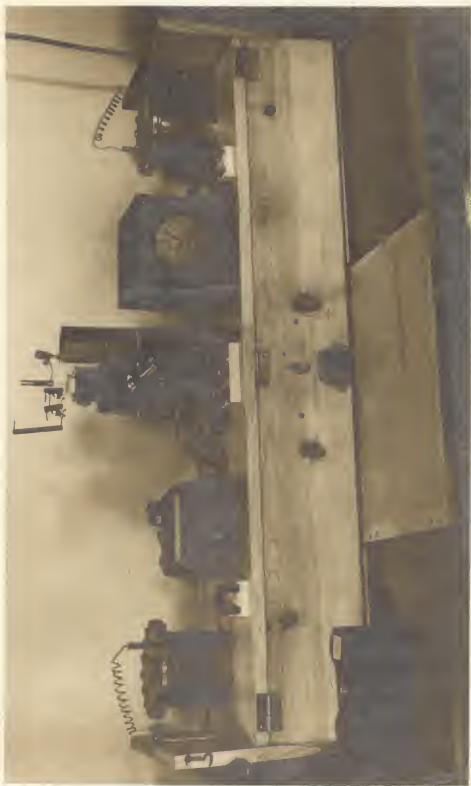
Schematic Circuit Diagram

20
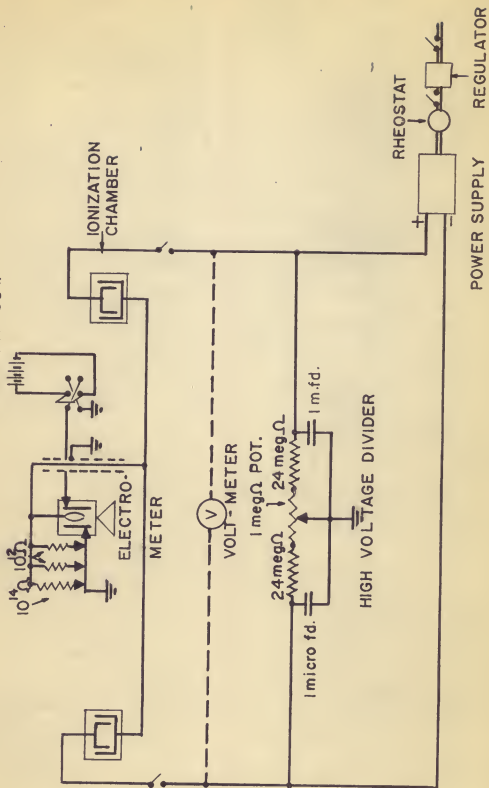
PLATE II

EXPLANATION OF PLATE III

The Differential Ionization Chamber Assembly

PLATE III

The Ionization Chambers

The chambers, (Plate IV), were designed to be sensitive to both beta
and gamma rays. Further consideration was given to constant geometry,
electrical insulation, and the possibility of applying a low pressure
(2 to 3 atmospheres) if this should be required. The high potential and
collector electrodes consisted of two concentric copper cylinders with
the collector cylinder being closed at the bottom. Gamma rays produced
secondary electrons in the walls of the high potential electrode, while
beta rays entered the chamber proper through the open end of the high po-
tential cylinder. The outer, grounded, cylindrical shell of the chamber
served both as a guard ring and as a protective cover. To predict the
approximate yield of each chamber some approximate, preliminary calcu-
lations were made which were later confirmed experimentally. Assuming
a source of 1.7 MEV beta rays and an activity of 3.7×10^7 disintegrations
per second (one millicure) the expected ionization current I was calcu-
lated from the equation −

$$I = e\bar{N}gn/3 \times 10^9 \text{ amperes}$$

e = 4.8×10^{-10}
N = 3.7×10^7 disintegrations per second
g = geometric factor = approximately 0.25 for beta particles
n = number of ion pairs formed per beta particle = 180
g   was estimated from the physical dimension of the chamber
n   was determined for an average 4 cm path from Rossi's table
    (22) of specific ionization.

This calculation resulted in an expected value of I = 2.67×10^{-10}
amps/mc. This was checked with a P^{32} source whose activity was determined
accurately by the isotope laboratory. The source had an activity of 2.17×10^6

EXPLANATION OF PLATE IV

Cross Section of Ionization Chamber - Scale 1:1

1   3/16" bolts, brass

2   Chamber top with source holder insert, brass

3   Stupakoff seal, high potential connector

4   Polyethylene gasket

5   Source holder, brass

6   Lucite spacer

7   High potential electrode, .08" copper sheet

8   Source backed by zapon film

9   Source cup of .06" brass cylinder with rubber seal
    (.6 mg/cm^2) at bottom

10 Outer shell of chamber, brass

11 Rubber film seal, (.6 mg/cm^2)

12 Collector electrode, .08" copper sheet

13 Ground bus, brass

14 Lucite support for collector electrode

15 Needle valve, brass

16 Stupakoff seal

17 Screw connector for chamber-electrometer conductor

PLATE IV
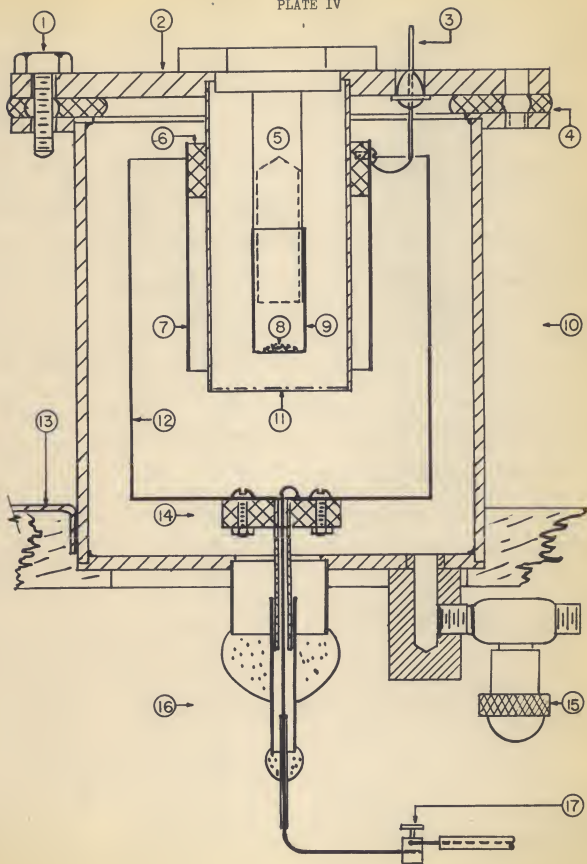
disintegrations per second. The resulting ioncurrent in the chambers
of this system was approximately 1.1×10^{-11} amps. This corresponded
to 1.88×10^{-10} amps/mc., in reasonably good agreement with calculations.
The small absorption due to the two layers of .6 mg/cm^2 rubber and the
enclosed air space in the path of the beta rays as well as self absorp-
tion were neglected, their influence being less than one percent, which
is small compared even to the error introduced in the measurement of the
ionization current alone ($\pm$ 5 percent). Also in this crude calculation
no estimate was made as to possible backscattering, recombination or sec-
ondary ionization. For gamma rays, the geometry factor had to be raised
to about .5, but an efficiency factor η of production of secondary elec-
trons had to be included. From Rossi's data (21) η was estimated to be
about 8.0×10^{-3} for 1.7 MEV photons in the brass chambers. This was a
modification of the value given by Rossi. His data referred to a beam
of photons which were incident on the chamber from the outside. Also
the average path of secondary electrons was only about 1.5 cm for this
case. The ions formed per cm remained about the same as in the first
calculation. The revised formula is -

$$I = \eta \ e \ N \ g \ n/3 \times 10^9 = 1.72 \times 10^{-12} \text{ amps/mc}$$

From these calculations recommendations could be deduced for the econom-
ical operation of the system as stated below.

The chambers were to be operated at saturation. For this purpose
ionization curves were plotted (Plate V) which indicated a saturation
plateau when 1800 to 1900 volts were applied to the chamber. The curves
also indicated a near equality of sensitivity for the two chambers. The

EXPLANATION OF PLATE V

Ionization Curve of Both Ionization Chambers.
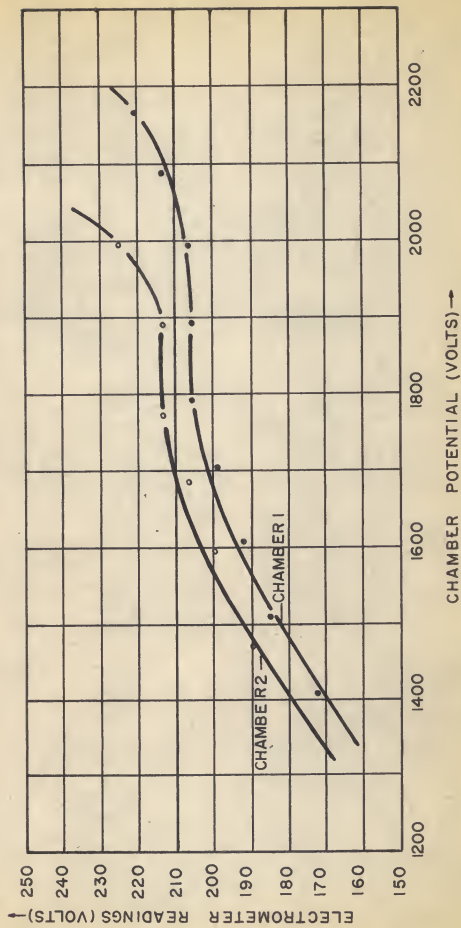
10^14 ohm resistor across electrometer

Same Ra-D source in either chamber

Same scale for both curves

At saturation, approximately 5% deviation
of response (might be due to source
position as well as chamber geometry).

PLATE V

relatively long path for ions formed by the beta rays of the Ra-D
source used for the determination of the ionization curve was considered
as the possible cause for the rather high saturation potential. The
spacing of the electrodes was large enough to avoid any danger of corona
discharge, therefore no particular precautions, such as polishing surfaces,
were taken to prevent discharges.

The Electrometer

The Wulff bifilar electrometer with auxiliary capacitor proved
satisfactory from the standpoint of stability and rigidity. Its sen-
sitivity, about .5 divisions per volt in the linear range, as well as
the extent of this linear range, 40 volt to 200 volt, was somewhat of
a handicap. Also, the leakage resistance of the electrometer's insul-
ators set an upper limit on the external resistance of the circuit. A
calibration curve (Plate VI) indicated the unlinearity of the low-voltage
range. To take readings in this low voltage range a potential was applied
to the auxiliary capacitor which encloses the fibers. If this potential
was of opposite sign to that of the fiber it increased the attractive
force on the fibers enough to bring about linear response. The auxi-
liary capacitor had also the advantage of indicating the sign of the
ionization current, which could otherwise not be determined with this
electrometer.

EXPLANATION OF PLATE VI

Calibration Curve of Wulff Bifilar Electrometer

PLATE VI

FIBRE SEPARATION (DIVISIONS) →

POTENTIAL APPLIED (VOLTS) →
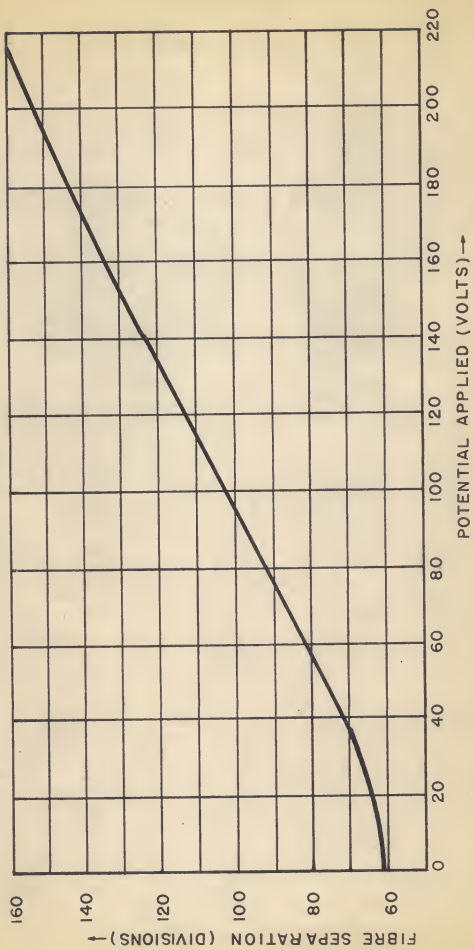
The Source Holders

A source cup consisting of a 6/100" thick brass cylinder, closed on one side with a thin film of rubber, took up the source to be investigated. For improved spatial stability and as a protection against diffusion the source was backed by a drop of liquid Zapon which quickly evaporated to form a film. The cup was then slipped over the partially hollow source holder which determined the position of the source in the chamber. The cup and the holder were marked to assure the same spacing. Also, the hexagonal top of the source holder bore markings at each point so that the angular position could always be reproduced by matching the respective points with a line engraved on the top of the chamber.

Circuit Resistances and Capacities

One of the major problems in the design of this system was that of obtaining optimum resistances to avoid leakage currents and to get the largest possible IR drop across the electrometer. To utilize the differential system fully the small changes in the difference current were to produce relatively large changes in the electrometer voltage, for fast and accurate measurements. With well matched sources the initial magnitude of the difference currents is somewhere in the neighborhood of 10^{-14} amperes and with activities of long life this current increases only slowly. Therefore the original aim was to use a resistance of about 10^{-14} ohm across the electrometer. However, when establishing the resis-

tances of the circuit components, it was found that the resistance of
the Stupakoff seals was about 10^{-12} ohms and that of the electrometer
less than 10^{-14} ohms. Application of ceresin wax upped all resistors
to 10^{-14} ohms. This produced a combined resistance of 2.5×10^{13} ohms,
all resistors being in parallel. Having component resistances of the
same magnitude as the standard resistor made for very unstable measure-
ments, because the components, despite the ceresin wax cover, were in-
fluenced by extreme humidity and temperature changes. Another disad-
vantage of using the 10^{14} ohm resistor as standard was the very long
time constant produced. The combined capacity of the system was mea-
sured as 70 micromicrofarad. Therefore with this large resistor the
RC product amounted to about 7000 seconds or nearly two hours. Despite
lowered sensitivity a resistor of 10^{12} ohm was applied across the elec-
trometer during actual measurements.

All resistance measurements were made by the RI drop method. A
known voltage was applied to a completely isolated, i.e. air suspended,
resistor arrangement which always included the electrometer. Using var-
ious combinations, the respective resistances could be calculated from
the voltage readings on the previously calibrated electrometer. The
capacities were measured by a time rate of change of potential method.

High Voltage Supply

In lieu of two individual supplies providing chamber potentials of
opposite sign one power supply was operated above ground. This had the

advantage that, due to the constant voltage division, any fluctuation
in potential was evenly shared by both chambers and always in the same
direction. The power supply was enclosed in a wooden case to prevent
accidental contact by the operator.

Operation

It was required to check the response of the system rather fre-
quently. This involved a recalibration of the electrometer every few
days and whenever accidental electrical or mechanical shocks affected
the fiber tension. Further on, during each measurement the chamber re-
sponse had to be calibrated against a standard source of long life.
For measurements of short duration this had to be done at least before
and after the measurement, for long lasting measurements, at least a
daily check was required. The chamber potential was constantly observ-
ed and corrected by means of the rheostat. Occasional check of the volt-
age division was also found to be necessary. As a protection against
large temperature changes the room was kept air conditioned 24 hours a
day. Minor temperature variations produced no observable influence.
"Dryrite" was placed in the neighborhood of all critical insulators to
control the humidity. Due to a very large time delay (about 12 hours) in
obtaining constant response of the system, the potential had to be con-
stantly applied to the chambers. This was possibly due to the time re-
quired to polarize the various chamber components, particularly the
Stupakoff seals, before the final field configuration had been established.

To measure the ionization of difference currents, the same resistor was used across the electrometer if possible. If it was required to change resistors for measuring currents of very different magnitudes, no effort was made to establish the absolute resistances. An arbitrary current was fed into the completely assembled system with one resistor in place and the resulting voltage was observed in the electrometer. Thereafter, the resistor was exchanged for any other which was to be used in consequent measurements. The new voltage readings were determined and multiplication factors established with respect to the first resistor. Thus, for example, one of the working equations might have become

$$V(t)/m_1 = V(o)/m_2 + V(o)_B \lambda t$$

if $V(o)_B$ was measured with the standard resistor and the difference potentials $V(t)$ and $V(o)$ were measured with resistors of multiplication factors m_1 and m_2 respectively.

As previously implied, closely matched sources were desired at the beginning of any measurement. However, it was found if the initial difference was too small the experimental error of each reading was greatly increased. In this case, it was required to apply both positive and negative potentials on the auxiliary capacitor of the electrometer and to evaluate the potential from the difference. This entailed at least a doubling of the normal error.

According to the sensitivity of the chambers a source activity of a few millicuries seemed to be recommendable. This insured an operating current which provided sufficiently large electrometer reading, with a

parallel resistor of the order of 10^{12} ohm or less. Consideration had to be given though, to the energy and kind of radiation emitted.

Recommended Changes to Improve Operating Conditions

To improve the speed and accuracy of the system an electrometer of greater voltage sensitivity such as the Wulff Unifilar Electrometer (1000 divisions per volt and the same capacity as Bifilar electrometer) might be installed in the system. A shielded ion current collector cable with an insulation such as "teflon" would provide better protection against induced currents without the danger of establishing a leakage path. A source of much longer half life than that of Radium D (22 years) and of somewhat greater activity than that used now (.25 mc) would be desirable. With regard to the power supply, a regulated and variable output would make the operation easier. For example, to calibrate the electrometer or to make resistance-capacitance measurements, a second supply of lower, variable output had to be employed. Some consideration might also be given to replace the Stupakoff seals by prefabricated Teflon seals. Should the system be operated with a sufficiently low standard resistor (10^{12} ohm or less) this change might prove unnecessary.

SIMPLE APPLICATION

A trial measurement was made to evaluate the reliability of the system. The two sources used were Ra-D and P^{32}. The half life of P^{32} was accepted to be 14.3 days (31). The sources were not closely matched because the available activity of the P^{32} source was very small, (.08 mc). A 10^{14} ohm resistor had to be placed across the electrometer due to the small P^{32} source. Readings were taken over a period of nearly fifty hours. Each individual reading was repeated for thirty minutes at three-minute intervals. The mean electrometer voltages observed at the mean time of observation were recorded in Table 1 and plotted on a linear scale (Plate VII).

Table 1. Trial Measurement of half life of P^{32}.

No. :	Date :	Hour :	Voltage observed
1	June 8,	9:15	176.70
2	June 8,	11:17	186.55
3	June 8,	13:28	199.44
4	June 8,	15:51	190.30
5	June 8,	18:03	204.70
6	June 9,	12:12	212.41
7	June 9,	14:31	215.80
8	June 9,	17:10	217.50
9	June 10,	9:36	225.10
10	June 10,	12:11	211.80

EXPLANATION OF PLATE VII

Decay Curve of P^{32}

(Straight line fitted visually)

PLATE VII
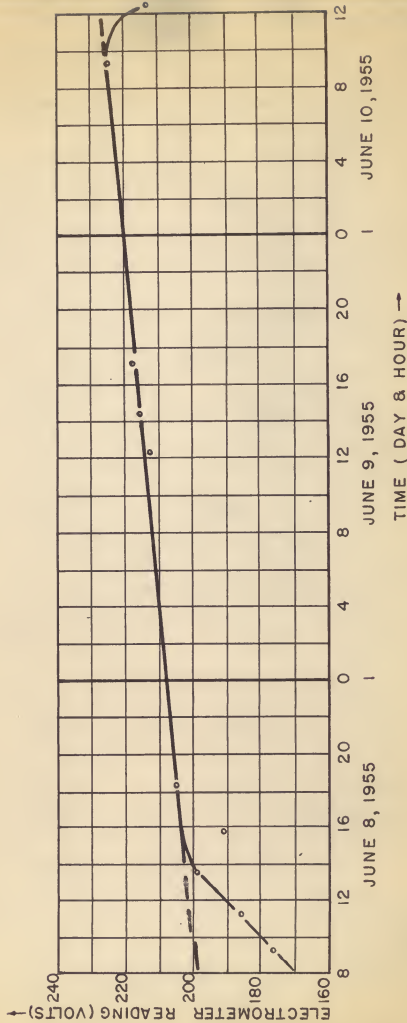
The straight line connecting these points was drawn by visual approximation, and a slope of 12.72 volt per day was found. With a 10^{12} ohm resistor in parallel with the electrometer and 1800 volt applied to each chamber $V(o)$ of Ra-D was found to be 18.20 volts and $V(o)$ of P^{32} amounted to 11.0 volts. This produced a difference potential of 7.20 volt. With a 10^{14} ohm resistor in the system the difference voltage was measured to be 176.20 volt. Thus, the change in resistors gave a multiplication (M) of 24.55.

Now
$$\frac{V(t)}{M} = \frac{V(o)}{M} + V(o)_p \; \lambda \; t$$

or
$$\lambda = \frac{\frac{V(t) - (Vo)}{t}}{M \; V(o)_p} = \frac{12.72}{24.55 \times 11.00}$$

and
$$t_{\frac{1}{2}} = 14.68 \text{ days.}$$

Making corrections for the fact that the P^{32} source had decayed since its initial measurement at the time $t = t(o) - 9$ hours, and also for the fact that the actual multiplication factor (M) had to be interpolated to the extension of the apparent decay curve (Plate VII), the final result became 14.48 days, which was fairly close to the accepted value of 14.30 days. The initial large slope of the decay curve was probably caused by a waiting period, after applying the potential to the chambers, which was too short to produce final chamber response. The drop off at the end of the curve was to be expected. Since the total voltage applied to the system is kept constant rather than the voltage applied to each chamber, the increase in electrometer voltage is added to the chamber potential of opposite sign and deducted from

that of like sign. In this case the chamber containing the Ra-D source
experienced a decline of potential which eventually dropped below the
saturation potential. It was concluded from this that a closer match-
ing of the sources was essential. Despite this shortcoming the results
implied a satisfactory operation of the whole system.

Subsequently, a second, more precise measurement of the half life
of P^{32} was made. A stronger source of undetermined activity was acquired
for this measurement. This permitted the use of a 10^{12} ohm resistor in
parallel with the electrometer. 3800 volt was maintained across the two
chambers at all times and the standard source was frequently checked to
correct for possible drifts. Each measurement consisted of 10 readings
taken within three minutes. The time of the first reading was taken as
the reference time. From the ten readings, the mean electrometer volt-
age and a root mean square deviation was calculated. To each point
a weight was assigned and by the method of least squares the decay curve
was plotted (Plate VIII).

Table 2. Data taken for accurate measurement of the half life of P^{32}.

No.	time (hrs.)	observed voltage	RMS deviation(volts)	weight
1	00.08	30.62	.06	1.00
2	18.17	25.88	.10	-
3	25.41	36.36	.19	.56
4	41.07	35.80	.13	.10
5	43.17	37.62	.16	.21
6	45.00	38.03	.09	.14
7	47.03	37.73	.09	.44
8	49.18	36.40	.09	.44
9	50.69	36.38	.08	.56
10	65.00	38.38	.08	.56
11	67.20	38.22	.10	.36
12	69.58	38.09	.12	.25
13	72.08	38.58	.10	.36
14	73.13	38.28	.06	1.00
15	74.60	38.38	.13	.21

EXPLANATION OF PLATE VIII

Decay Curve of P^{32}

(Data taken for 75 hours)

PLATE VIII

$X = TIME (HOURS) \longrightarrow$

$Y = \left[V(t)_{Ra-D} - V(t)_{P32} \right]$ (VOLTS) $\longrightarrow$

POINT 2

43
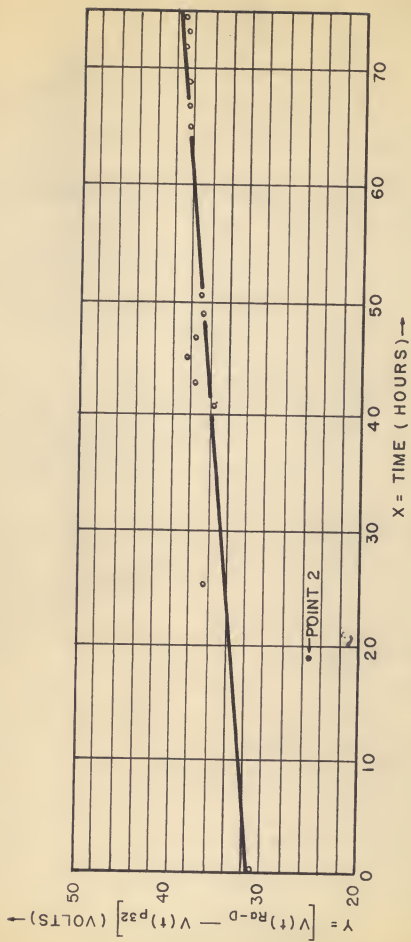
Point 2 was neglected in the calculations, because its deviation from the decay curve was more than five times the average deviation. The reason for the large deviation of this point was established as being due to a very short waiting period after inserting the source. The weights were determined from the formula $W_i = C/p_i^2$ (Birge, 4) where C = constant (arbitrary). Here C was set equal to p_1^2. The first calculation was made assuming linearity of the decay curve. The required equation was then of the form: $y = a + bx$, where $y = V(t)$; $x = t$; $b = -\lambda V(o)_p$ and $a = V(o)$. By the method of least squares, (Birge, 4) -

$$a = \frac{(\Sigma wy)(\Sigma wx^2) - (\Sigma wx)(\Sigma wxy)}{D} \; ; \; b = \frac{(\Sigma w)(\Sigma wxy) - (\Sigma wx)(\Sigma wy)}{D}$$

$$D = (\Sigma w)(\Sigma wx^2) - (\Sigma wx)^2$$

The resulting values were:

$$a = 31.76 \text{ volt} \qquad \text{and} \qquad b = .0982 \text{ volts/hr}$$

$V(o)_{Ra-D}$ remained constant at 80.80 volts and was used in the calculation rather than the experimental values of $V(o)_{P32}$

$$V(o)_P = V(o)_R - a = 80.80 - 31.76 = 49.04 \text{ volts}$$

Therefore, $t_{\frac{1}{2}} = \frac{.693 \times 49.04}{2400 \times .0982} = 14.42$ days.

The error in b was calculated from:

$$P_b = .675 (\frac{\Sigma w}{D} \cdot \frac{\Sigma wv^2}{N-2})^{\frac{1}{2}} \qquad \text{(Birge, 4)}$$

where v was the residue of each point. This resulted in

$$P_b = 6.450 \times 10^{-4} \text{ which was .66\% of b.}$$

The deviation of $(V(o)-a)$ was .08% of the extrapolated values, so that the total error of $t_{\frac{1}{2}}$ was .74%. In addition, the initial approximation $\Delta\lambda = \lambda_b$ (see section on Theory) produced a systematic error of .13% or a decrease of the calculated half life of .02 days. The corrected value became

$$t_{\frac{1}{2}} = 14.40 \pm .11 \text{ days}$$

The decay of Ra-D was also taken into consideration. Taking the half life of Ra-D as 22 years and the duration of the measurement as 3.12 days

$$V(t) = 80.8e^{-\frac{.693 \times 3.12}{22 \times 365}} = 80.8(1-2.69 \times 10^{-4})$$

i.e., a change of .03% took place, which amounts to only .0042 days and could be neglected. Another calculation was made with the same values and by the same method to check whether the assumption that the decay curve was a straight line was a sufficiently accurate. The basic formula used was: $\ln\left[V(o)_R - V(t)\right] = \ln V(o)_p - \lambda t$

the result was: $t_{\frac{1}{2}} = 14.31 \pm .14$ days,

and, again correction for the decay of Ra-D was neglected. A third value was obtained by using the logarithmic relation without giving weight to the individual points. This value was - $t_{\frac{1}{2}} = 14.50 \pm .07$ days.
To further check the results a new trial was started and measurements were taken for 11 days; i.e. nearly a full half life. The stability of the system was checked against the Ra-D source, and appropriate corrections were made. Because the Ra-D source was treated as being constant no decay correction was made. It would, in any case, still have been

too small to have any effect on the significant numbers of the result.
Table 3 shows the data taken and Plate IX illustrates the decay curve.
The calculations were made by the least square method from the equation:

$$\ln \left[V(o)_R - V(t) \right] = \ln V(o)_R - \lambda t$$

$V(o)_R = 78.39 \pm .08$ volts at time $t(o) = 13:10$ o'clock, June 30.

Table 3. Data for a measurement of the half life of P^{32} taken over a
relatively long span of time (11 days).

No.	time (days)	$V(o)_R - V(t)$ (volt)	RMS Dev.(volt)	weight
1	0.110	38.27	.15	46.28
2	0.841	37.61	.21	17.08
3	0.894	34.85	.60	1.82
4	1.049	36.87	.11	59.17
5	1.222	35.59	.15	30.19
6	1.891	33.55	.11	59.17
7	3.974	35.49	.22	14.24
8	4.843	28.11	.10	42.17
9	5.014	31.69	.13	37.18
10	6.676	30.05	.11	40.57
11	7.125	25.59	.14	17.80
12	7.888	28.19	.13	25.25
13	9.000	23.79	.11	24.75
14	10.009	26.63	.10	37.64
15	11.006	24.32	.12	21.84

The result was: $t_{\frac{1}{2}} = 14.44 \pm .16$ days.

The somewhat higher experimental error was due to having to account for
two deviations at each point, that of the difference voltage and that
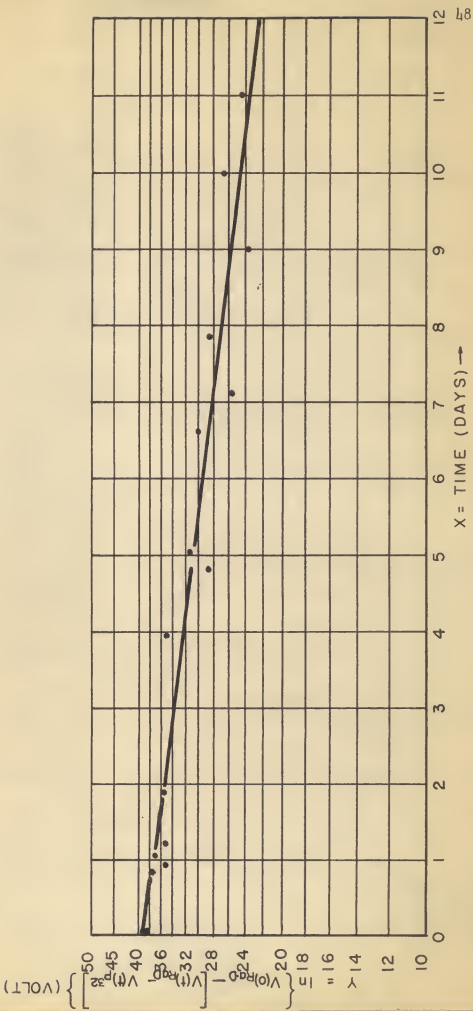of the Ra-D voltage.

EXPLANATION OF PLATE II

Decay Curve of P^{32}

(Data taken for 11 days)

PLATE IX

DISCUSSION OF RESULTS AND CONCLUSION

The results are somewhat above the most generally accepted one of 14.30 days. However, Sinclair and Holloway (28) measured the half life of P^{32} over three half lives and found for two different sources the values $14.60 \pm .05$ days and $14.59 \pm .03$ days. Thus, the three most reliable values found in this experiment: $14.40 \pm .11$ days, $14.31 \pm .14$ days, and $14.44 \pm .14$ days, with their overlapping deviations were not considered unreasonable. The average of these values was $14.38 \pm .14$ days. The conclusion drawn from these results was that the performance of the apparatus justified the expectation that medium long half lives could be measured with a fair degree of accuracy (1 to 2%). With sources somewhat stronger than used here, measurements of this accuracy should be obtainable in 1/20th of the half life to be investigated, using a standard resistor not higher than 10^{12} ohm. If the operating conditions are carefully maintained the speed might be increasable to 1/50 to 1/100 of a life time by using a 10^{14} ohm standard resistor. More promising effects on speed and reliability could be expected by giving consideration to the improvements suggested above.

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THE DESIGN AND OPERATION OF A DIFFERENTIAL IONIZATION
CHAMBER SYSTEM FOR HALF LIFE MEASUREMENTS

by

RICHARD JOHN HEINRICH SNEED

B. A., and B. S., St. Mary's University, San Antonio, Texas, 1953

AN ABSTRACT OF A THESIS

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1955

As a literature survey showed, various methods have been designed to experimentally determine half lives of magnitudes ranging from 10^{-10} seconds to 10^{12} years. There was, however, an apparent lack of methods permitting accurate and reasonably short measurements of half lives ranging from several months to about 50 years. Segre suggested that the differential ionization method, introduced by Rutherford, would make possible both comparative and absolute measurements of the decay of long lived isotopes within a fraction of their half life. The reliability and speed of this method was verified very recently by Tobailem.

For comparative measurements two radioactive sources of nearly equal half life are required, one being the standard source of known half life. The sources produce ion currents in separate chambers. The potential applied to the two chambers is of opposite sign, as are the resulting ionization currents which are fed into a common conductor. The difference of these currents is measured and the time rate of change of this difference current represents the change of the activity of one source with respect to that of the other. Mathematically this can be expressed by the following equations:

$$i_1 = i_{o1}e^{-\lambda_1 t}; \quad i_2 = i_{o2}e^{-\lambda_2 t} = i_{o2}e^{-(\lambda_1 + \Delta\lambda)t}$$

$(i_1 - i_2) = e^{-\lambda_1 t}(i_{o1} - i_{o2}e^{-\Delta\lambda t}); \Delta\lambda\, t \ll 1$ for sources of nearly equal half lives and for a relatively short observation time. Therefore,

$$(i_1 - i_2) = e^{-\lambda_1 t}(i_{o1} - i_{o2}) + e^{-\lambda_1 t} i_{o2}\,\Delta\lambda\, t,$$

For absolute measurements the standard is an isotope of practically infinite half life with respect to that of the source to be investigated

and, therefore, $e^{-\lambda_1 t} = 1$ and $\lambda_2 - \lambda_1 = \Delta\lambda = \lambda_2$, i.e., $(i_1 - i_2) = (i_{o1} - i_{o2}) + i_{o2} \lambda t$.

In this experiment the sources were mounted inside two cylindrical ionization chambers and the resulting difference currents were measured with a Wulff Bifilar electrometer. The half life of P^{32} was measured for the primary purpose of evaluating the operating characteristics of the system. However, the experiment was conducted as carefully as possible and the data was treated in such a manner as to render the results reliable. The first measurement indicated a half life of 14.48 days for P^{32}. Ra-D (half life - 22 years) was used as a standard source. The data were plotted on a linear graph and the decay curve was visually fitted. The result of a second measurement was obtained by treating the data which were taken over a three day period and with a different P^{32} source by the least square method of fitting curves. The data were fitted according to the linear relation, $(i_1 - i_2) = (i_{o1} - i_{o2}) + i_{o2} \lambda t$, and, according to the relation, $\log[(i_1 - i_2) - (i_{o1} - i_{o2})] = \log i_{o2} - \lambda t$. This resulted in the two values, $14.40 \pm .11$ and $14.31 \pm .14$ days for the half life of P^{32}. Another measurement, data being taken for 11 days, produced the result of $14.44 \pm .16$ days for the same half life. All results are in reasonable agreement with values determined by other investigators (14.30 days to 14.60 days). The performance of the equipment showed satisfactory stability. A greater speed in taking measurements could be obtained by increasing the sensitivity of the system, in particular that of the current measuring device.