THE HALF-LIVES OF
Sr^{90} AND Cs^{137}

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SCOPE AND CONTENTS: The half-lives of Sr^{90} and Cs^{137} were measured by the method of observing the disintegration rate of a known number of atoms. A 4π proportional counter was designed and constructed to measure the absolute disintegration rates of the samples used. The efficiency of the counter was tested with calibrated radioactive solutions from the National Bureau of Standards. The submicrogram quantities of the carrier-free radioisotopes used for counting were determined with the mass spectrometer along with isotope dilution techniques.

Independent evidence is given to support the half-lives found in this investigation.

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GENERAL INTRODUCTION

The nuclei of many atoms which are inherently unstable disintegrate spontaneously to yield more stable nuclear species. This process known as radioactive decay involves the release of energy by the nuclei of isotopes seeking a less energetic configuration. The disintegration rate of a given radioactive nucleus is of fundamental importance to those who investigate the properties of the atom both from experimental and theoretical points of view. It has been found that the rate of decay of a radioisotope can not be materially influenced by changes in physical conditions and it has been deduced that this rate is directly dependent on the number of atoms present. Mathematically the exponential decay of radioisotopes can be described by the following equation.

$$-dn/dt = n\lambda \qquad [1]$$

where $-dn/dt$ is the disintegration rate

n is the number of atoms present

λ is the decay constant and equals 0.6932/half-life.

Consequently the rate of radioactive disintegration for a given isotope is usually described in terms of its half-life, or the time required for one half of the atoms in a given sample to undergo decay.

Although a great many methods of measuring the rate of radioactive decay have been used, basically they are all variations of two main approaches. In one method, the change in the number of disintegrations occurring in a given sample is measured over a period of time, preferably several half-lives. The other method, applicable in the case of isotopes which decay more slowly, involves the measurement of the fraction of the total number of atoms known to be present which disintegrates in a given instant. In the calculation of half-lives both approaches make use of some form of the above equation.

It can be seen from equation [1] that the longer the half-life, the larger the number of atoms necessary for a given disintegration rate. This means that, while ordinary techniques can be used to weigh out suitable amounts of long-lived isotopes for counting rate determinations, less direct methods must be used in the case of those with short half-lives. For these radioisotopes the disintegration rate is very large for a number of atoms which are weighable by standard techniques. Thus for short-lived isotopes to which decay with time studies are applicable, it is not necessary to be able to determine the number of atoms involved. The precision of the half-lives obtained in this manner is dependent on the observed change in relation to the presicion of the measurements. From the point of view of such measurements, the longer the half-life, the poorer is the precision obtained.

There is a class of nuclides with intermediate half-lives which are difficult to determine by either of the two methods described above. Measurements of these in-between half-lives have been made both with specific disintegration rate and decay with time approaches although in almost all cases modifications to the basic methods have been necessary. For example, especially sensitive apparatus is essential if decay with time measurements are to be applied to a radioisotope with a fifty year half-life because the change even in five years is slight. Specific disintegration rate studies would also require a special way of determining the number of atoms since conventional techniques are not applicable to such an isotope.

In the fission process, two of the many radioactive products formed, i.e., Sr^{90} and Cs^{137}, belong in the class of nuclides with intermediate half-lives. Most radioactive fission fragments decay quite rapidly so that after two or three years almost all of the activity results from these two isotopes. Since they are formed in high yield in fission, large quantities of them are accumulating from the operation of nuclear reactors.

This thesis reports the measurement of the half-lives of Sr^{90} and Cs^{137}, both of which are difficult determinations using conventional techniques. Previous values based on decay with time studies have been relatively inaccurate because the decay was not followed for a sufficient length of time.

Application of the instantaneous disintegration rate method had previously been hampered by lack of an accurate method for measuring the submicrogram quantities of the isotopes which had to be used. In the present work a brass 4π proportional counter was designed and constructed to determine absolute disintegration rates and the number of atoms was measured with a mass spectrometer using isotope dilution techniques.

HISTORICAL INTRODUCTION

The term radioactivity applies to the spontaneous stabilization of the nuclei of the atoms of certain isotopes called radioisotopes. This stabilization process is known to occur in many forms such as the emission of alpha, beta or gamma radiations by the excited nuclei. There are, therefore, alpha, beta and gamma half-lives which, because of the different transformations involved, must be measured with different techniques. For example, beta half-lives range from 6 x 10^{14} years for In^{115} (1) to 0.022 seconds for B^{12} (2) (3). Short-lived nuclear isomers on the other hand often have gamma half-lives of the order of 10^{-9} seconds while alpha half-lives vary over the widest range of all. The method used for determining rates of disintegration will therefore depend on the type of emission involved.

It should be pointed out that the rate of beta decay depends on four variables (4):

(i) the energy involved in the transition

(ii) the charge remaining on the nucleus after disintegration has occurred.

(iii) the amount of angular momentum and spin carried away by the electron and the neutrino.

(iv) whether the parity of the nucleus is reversed in undergoing decay.

The influence of these factors manifests itself in the wide range of beta half-lives observed. As a result, methods which are adequate for measuring the beta half-lives of say B^{12} and P^{32} are not applicable in the cases of Rb^{87} or Cs^{137} because the former decay so much more rapidly than the latter.

This situation of widely different rates of decay is common to all three types of radioactive emission and has necessitated a wide diversity in the methods used to measure half-lives.

(A) Methods of Determining Half-Lives Based on the Decay with Time Approach

(a) Isotopic ratios measured with a mass spectrometer

If a radioactive isotope of an element is present along with a stable isotope of the same element, the change in the ratio of these two isotopes with time can be used to establish the half-life of the radioisotope. Thode and Graham (5) made the first such measurements with a mass spectrometer and found the half-life of Kr^{85} to be ten years. Later Wanless and Thode (6) published the value 10.27±0.18 years after following the changes in the Kr^{85}/Kr^{86} ratios over a period of seven years. Macnamara, Collins and Thode (7) established the half-life of Xe^{133} as 5.271±0.002 days by comparing the change with time of the ratios of this isotope and the stable Xe^{131}, Xe^{132}, Xe^{134}, Xe^{136} isotopes. By following the increase in the Cs^{133}/Cs^{137} ratio for over five years, Wiles and co-workers (8) set a value of 33 ± 2 years for the half-life of Cs^{137}. Karraker et al. (9) used the same approach to measure the half-lives of Eu^{152} (13 ± 2 years), Eu^{154} (16 ± 4 years) and Sm^{151} ($73 {}^{+25}_{-14}$ years).

Use of a mass spectrometer to measure isotopic ratios has not been restricted to decay with time studies. Strassman and Walling (10) evaluated the half-life of Rb^{87} in a lepidolite sample in the following way. They determined the total amounts of strontium and rubidium by chemical means and measured the ratios Sr^{87}/Sr_{total} and Rb^{87}/Rb_{total} with a mass spectrometer. By assuming a geological age for the mineral they obtained the value 6.3×10^{10} years. An alpha half-life of 8800 ± 600 years was determined for Am^{243} (11) from the mass spectrometric ratio of Am^{243}/Am^{241} and the ratio of alpha activities measured with a differential pulse analyzer.

(b) Measurement of amount of product formed

Early measurements of the half-life of tritium were made by counting the betas emitted from radioscale quantities of the isotope (12) while more recent measurements have involved following the decay of a larger sample with an ionization chamber (13). At the same time, Novick (14) first determined the half-life by measuring the rate of formation of He^{3}, the product of the beta decay of T. In the most recent determination Jenks and his co-workers (15) achieved greater accuracy with a modification of the latter method. A gas density balance was used to measure the quantity of tritium, and disintegration rates were determined by measuring the rate at which He^3 was formed.

Inghram (16) used a similar approach to determine the half-life of Pu^{240}, i.e.,he used the growth of the uranium daughters from a plutonium sample of known Pu^{239}/Pu^{240} isotopic composition. The mass spectrometric ratio of the U^{235} and the U^{236} formed from the alpha decay of the plutonium isotopes gives the relative half-life of Pu^{240} from the relationship

$$\frac{T_{\frac{1}{2}}\ (Pu^{240})}{T_{\frac{1}{2}}\ (Pu^{239})} = \frac{\text{moles of } U^{235}}{\text{moles of } U^{236}} \times \frac{\text{moles of } Pu^{240}}{\text{moles of } Pu^{239}}$$

A great deal of work has been done on the determination of the specific alpha activity of radium since for many years it was used as a reference material for numerous measurements in the field of radioactivity. The work has been hampered by the presence of the short-lived descendants of radium and by the fact that the half-life is too long for accurate decay with time studies. Consequently many different approaches have been tried, one of the earliest being the measurement of the rate of He evolution in Ra decay (17). Sir James Dewar determined the amount of He formed in nine months from a sample of $RaCl_2$ in a sealed tube, obtaining a value of 0.463 cu. mm. per gram of radium per day which means a half-life of 1628 years.

<u>(c) Delayed coincidence method</u>

When one nucleus undergoes beta decay the product nucleus may be in an excited or nuclear isomeric state whereas disintegration of another may lead directly to the stable ground state. Such a situation is to be expected when the two lowest states of a nuclear system have similar energies but differ considerably in angular momentum. The greater this difference the longer will be the half-life of the nuclear isomer [this can range from 10^{-13} seconds to a few months (18)] which can de-excite by the emission of either a gamma-ray or an internal conversion electron (19). The gamma half-lives of short-lived nuclear isomers are usually measured by the delayed coincidence method which utilizes an electronic apparatus capable of recording very short time intervals. The sample is placed between two counters, for example a gamma scintillation counter and a beta proportional counter. During the simultaneous stimulation of both counters, the delay time of one channel is varied with respect to the other and the per cent coincidences is plotted as a function of delay time. The identification of isomers is accomplished by searching for any asymmetry in the usually symmetrical resolution curve obtained (20) since this indicates an isomeric transition. The half-life of the nuclear isomer can then be calculated from the slope of the line obtained by plotting the logarithm of the coincidences per minute versus the delay in microseconds.

The delayed coincidence method has been used (21) (22) (23) for measurements of such isomers as Ta^{181}, Ge^{72} and with the development of scintillation counters of fast decay time, half-lives of the order of 10^{-11} seconds can be measured (24). The equipment has been adapted, however, to measure half-lives of short-lived nuclides other than nuclear isomers. Using an anthracene scintillation spectrometer with pulse height discrimination and delayed circuit timing equipment, Kline and Zaffarano (25) measured the beta half-lives of Ca^{39}, Li^{8}, He^{6} and O^{15}.

(d) Change in counting rate with time

Observation of the exponential decay of a radioisotope over a period of time has been applied to a great many half-life investigations, particularly to those in the range of fractions of a second to one or two years. These measurements often require the least complicated apparatus since there is no need to know the amount of nuclide present and often not even the geometry of the counter or the efficiency of the counting equipment. For example, Niday (26) observed over a period of about 115 days the decreasing disintegration rate of a sample of rubidium chemically separated and purified from fission products. By absorbing the beta particles from naturally radioactive Rb^{87} with aluminum he was able to measure the half-life of Rb^{86} as 18.64 ± 0.04 days. The half-lives of Na^{24}, Cl^{38}, Br^{82} and Tc^{96}, all in the 0.5 to 110-hour range, were measured (27) with a 4π ionization chamber connected to a recording potentiometer which continuously recorded the instantaneous value of the activity as a function of time.

Other varieties of apparatus have also been used for decay with time studies. The half-lives of Co^{60m} and Al^{28} (28) were found to be 10.47 ± 0.02 min. and 2.27 ± 0.02 min. respectively by means of measurements with a recording quartz fiber electroscope. The electroscope reading was automatically recorded at various intervals along with the time elapsed since the start of the experiment. This was particularly advantageous for disintegration rate studies of short-lived isotopes.

(B) Methods Involving Absolute Disintegration Rate Determinations

If the half-life being investigated is of such a length that insufficient disintegration of the radioisotope occurs to give accurate decay with time studies, it is necessary to find the absolute rate of decay of a known amount of radioisotope. The following section illustrates some of the methods which have been used.

(a) The number of atoms determined by direct weighing

The half-life of a long-lived beta emitter, Rb^{87}, has been measured by two sets of workers using two quite straightforward approaches. Curran and co-workers (29) weighed out tenth-gram samples with a semi-micro balance and measured their specific disintegration rates with a large proportional tube counter. MacGregor and Wiedenbeck (30) vaporized RbCl on to aluminized zapon films, weighed the sample and measured the specific activity with a specially designed beta counter.

An equally direct technique has been applied in the case of Tc^{99} (31). The half-life of Tc^{99}, 2.12×10^5 years, was measured by counting aliquots of solutions made by dissolving milligram quantities of NH_4TcO_4 and technetium metal. The half-life of a third long-lived beta emitter, Cl^{36}, was determined in a similar manner (32). Aliquots of HCl^{36} solution were evaporated to dryness and weighed. The specific disintegration rate of these samples was then measured with a standard mica-window G.M. counter and the half-life was found to be $(0.44 \pm 0.05) \times 10^6$ years.

Application of such a method to relatively short-lived Am^{241} ($470 \pm \overset{5}{10}$ years) was possible however, only by using the most sensitive of techniques (33). B. G. Harvey at Chalk River used americium purified by chemical means and shown spectrographically to contain less than 1 per cent impurity. He was able to weigh five microgram samples of pure Am^{241} as AmO_2 on a quartz torsion fiber ultramicrobalance and then measure the alpha activity of these samples with a methane-filled proportional counter.

(b) The number of atoms determined by isotope dilution

That class of nuclides which have what have been called intermediate half-lives require techniques other than direct weighing and counting which were successful with long-lived isotopes. A weighable amount of radioisotopes with half-lives less than about 200 years would have far too high a specific disintegration rate to be counted or handled conveniently. Therefore some way other than direct weighing must be found for measuring the number of atoms in a more moderately active sample.

Three separate determinations of the half-life of 73.6 year U^{232} have been reported, each one illustrating a different approach. The first attempt (34) was an estimate based on the growth of U^{232} alpha activity from its thirty year beta emitting parent Pa^{232}. The half-life was also estimated from the growth of U^{232} from its alpha emitting parent Pu^{236} (35) but by far the best value has been obtained by the use of an entirely new technique. Sellers et al. (36) determined the specific activity of their samples by dilution with a large excess of U^{238} so that they might use conventional weighing and counting methods. A mass spectrometer was used to measure the number of U^{232} atoms relative to the number of U^{238} atoms which had been obtained directly from the weight.

(C) Other Methods

Considerable ingenuity has been shown in many half-life determinations reported in the literature by experimenters confronted with problems unique to a particular radioisotope. They found it necessary to make use of particular facts and unusual conditions, or to design apparatus of a very specific nature because conventional instruments were inadequate. It is of interest to see some indication of the diversity of approach.

It was mentioned in section (A) that the 0.022 sec. half-life of B^{12} is one of the shortest for beta decay. Becker and Gaerttner (2) accomplished this determination by neutron activation of a boric acid target supported on an annular ring rotated at 1800 r.p.m. The half-life was measured by observing, with Lauritsen electroscopes, the activity as a function of the angular displacement from the point of bombardment. This was, of course, just a special type of decay-with-time study.

Eichelberger et al. (37) utilized the fact that because of its simple decay scheme Po^{210} makes an excellent heat source for the testing of calorimeters. These workers followed the decay of six polonium samples with four different steady-state resistance-bridge calorimeters and found the half-life to be 138.4005 ± 0.0051 days.

The two independent studies of the radioactive decay of the neutron are interesting particularly because of the differences in method and apparatus and the close similarity between results. In both cases a neutron beam from the nuclear reactor was subjected to a high voltage such that protons resulting from neutron degradation would fall on the first plate of an electron multiplier. Robson at Chalk River (38), using the values for the number of protons striking the electron multiplier, the efficiency of the collecting and focusing system and the density of neutrons in the beam, set the half-life in the range nine to twenty-five minutes. He later extended this method by including the measurement of beta particles coincident with protons and obtained a value of 12.8 ± 2.5 minutes (39). Snell and co-workers (40) used a similar approach and with corresponding measurements estimated a half-life of ten to thirty minutes.

One of the most intriguing half-life studies is that made on gaseous products of uranium fission by Overstreet and Jacobson (41). The method, called the charged wire technique, was carried out as follows. The radioactive fission product gas was made to flow down a tube, through the centre of which stretched a negatively charged copper wire. The neutral gas atoms disintegrated as they passed through the tube and the resulting positively charged daughter atoms were deposited on the wire within the immediate vicinity of their formation. Since the gas was flowing at a controlled rate, the distribution of the radioactive decay products along the wire served as a measure of the half-lives of their respective parents. Studies were carried out on short-lived xenon and kryton isotopes (41) (42) as well as on Ba^{139} (43).

(D) Prediction From Nuclear Theory

The prediction of alpha half-lives is based on the relationship between the radioactive decay constant or half-life of a radioelement and the range of the mono-energetic alpha particles emitted by it. Geiger and Nuttall (44) found experimentally that a straight line is obtained for a plot of $\log\lambda$ against the logarithm of the range ($\log R$) for a number of alpha emitters. The relationship

$$\log\lambda = b + c \log R$$

called the Geiger-Nuttall rule allows the calculation of alpha half-lives from the range of the respective alpha particles since values of the constants (b and c) have already been established in the case of alpha emitters of known half-life and range.

The prediction of beta half-lives does not benefit from any such relationship and therefore has not been nearly as successful. Lifetime estimates are reported in terms of "ft" values or comparative lifetimes where "t" is the observed half-life and "f" is the theoretical factor which corrects "t" for the effects of the nuclear charge and the energy of the transition. Agreement between "ft" estimates and experiment to within a factor of 100 in either direction is considered to be satisfactory (45).

The phenomenon of nuclear isomerism, already mentioned in part (A) (c) as the subject of much experimental work, has also been extensively studied from the theoretical point of view. It occurs when, in spite of having identical constitution, two nuclei decay with different half-lives and it is most prevalent among artificially produced nuclei (46). Metastable isomers, besides occurring in certain beta decay processes, have been produced by absorption of gamma rays and by other nuclear reactions.

Comparisons of experimental lifetime values and estimated theoretical lifetimes have been tabulated (18)(47) and although there are often large discrepancies between them, there is sufficient correspondence to strongly support the theory. For the first excited state of B^{10}, the theoretical value of 5×10^{-10} seconds is in excellent agreement with the experimentally obtained value of $(7 \pm 2) \times 10^{-10}$ seconds (48). In the case of O^{17} there is a discrepancy which the authors have explained in terms of the phenomenon of core excitation.

(E) Previous Half-Life Determinations of Sr^{90} and Cs^{137}

Strontium-90 along with its daughter yttrium-90 was first reported by R. W. Nottorf (49) in 1943 and at that time his decay with time studies indicated a half-life of 23 ± 3 years. Glendenin and Coryell on the basis of a smooth fission yield curve set the value at twenty-five years (50). The currently accepted value of 19.9 ± 0.3 years is the result of decay studies over a period of seven years using a Lauritsen electroscope (51).

The half-life of Cs^{137} was first estimated by Glendenin and Metcalf (52) from fission yield data to be thirty-three years and then by Engelkemeier to be thirty-seven years (53). The first accurate measurements were reported by Wiles, Smith, Horsley and Thode (8) and were decay with time studies as described in section (B)(a). The changes in Cs^{133}/Cs^{137}

ratios, measured for periods up to 5.4 years, were used to calculate a value of 33 ± 2 years.

EXPERIMENTAL

In this work the half-lives of Sr^{90} and Cs^{137} were measured with a specific disintegration rate determination method, one which involved the measurement of the fraction of the total number of atoms known to be present which disintegrated in a given instant. Absolute disintegration rates were measured with a 4π counter designed to operate in the proportional region, i.e.,the potential between electrodes is such that the size of the pulses is proportional to the number of ion pairs produced in the counter gas by the beta particles. The determination of the number of atoms present was accomplished with a mass spectrometer using isotope dilution techniques. Isotope dilution is the measurement of the quantitative effect on isotopic ratios caused by the addition of a known quantity of one of the isotopes (or of the same isotopes in different relative amounts). The absolute amounts can then be calculated from the relative mass spectrometric ratios, both before and after the dilution, and the amount of each isotope added.

(A) Absolute Disintegration Rate Determinations

(a) 4π counter

The use of a conventional end-window G.M. counter for the measurement of absolute disintegration rates involves so many corrections that the results obtained in this manner are of doubtful accuracy. The counting rate actually observed might well be less than 10 per cent of the absolute value because of the corrections necessary for geometry, backscatter, window and air absorption and self-absorption in the sample (54). For this work, therefore, a cylindrical 4π proportional counter, built to operate at one atmosphere of methane, was designed and constructed. Its general proportions were substantially the same as commercial models and others described in the literature (55). Details of the component parts can be seen in Figure 1. The upper and lower halves of the counter form a cylinder when assembled for operation. The anodes A which are loops of tungsten wire (0.001 in.) are soldered into the ends of copper wire B(0.040 in.) which passes through lucite insulators C. For efficient "sweeping-out" of the counting volume, C.P. methane was allowed to flow steadily into both halves through threaded nozzles D and out through similar nozzles E. Rubber "O" rings, F and G, assure a gas-tight fit between the assembled parts while dowel pins H assure reproducible alignment between the two halves. Nichrome disk sample holders J, which fit in the small indentations K in the bottom half, have eight small openings L around the central opening to allow circulation of methane. The anode connections are such that the upper and lower halves can be operated independently as well as simultaneously. A demountable clamp was used to

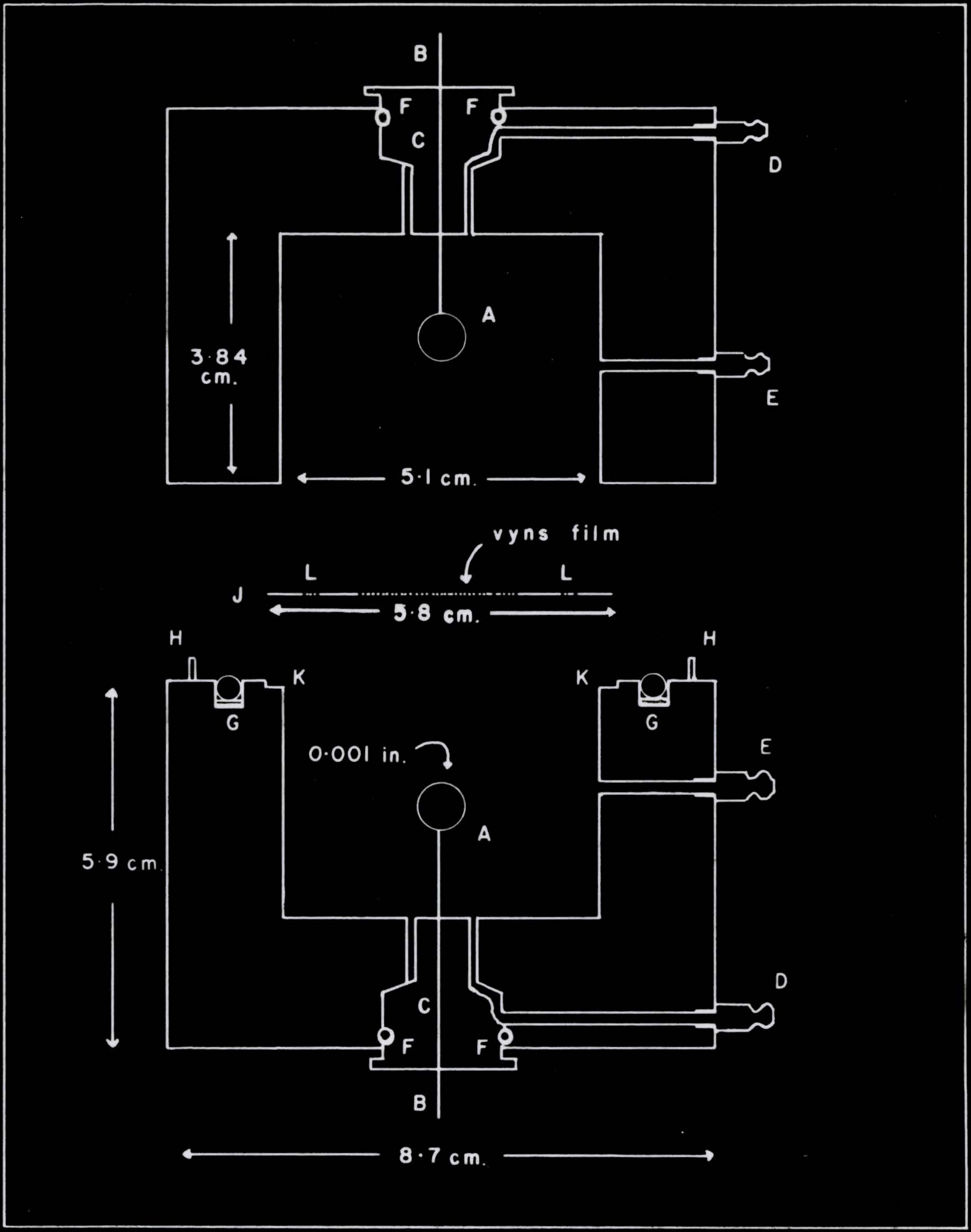


FIG. 1 - 4π PROPORTIONAL COUNTER

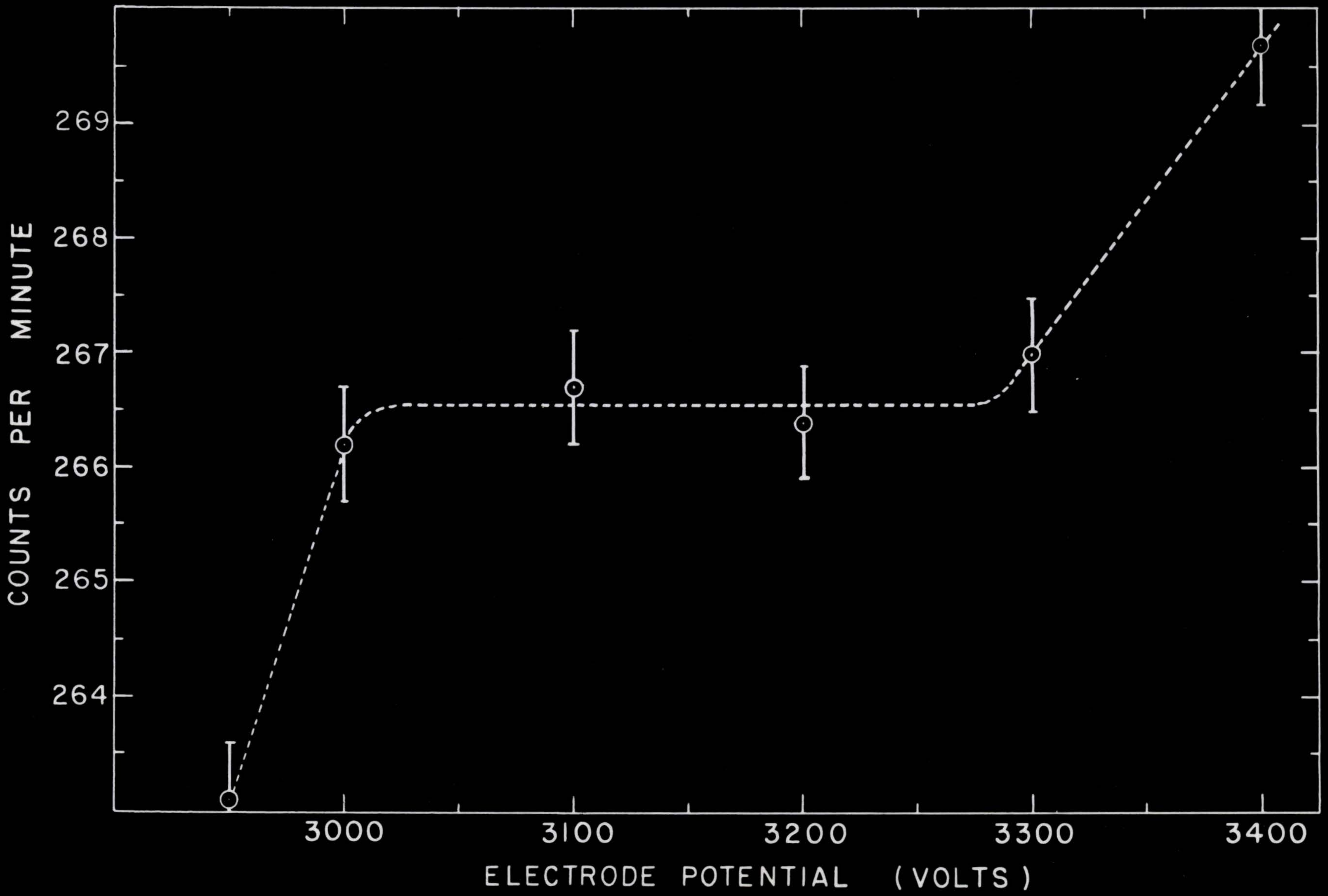
hold the halves together in such a way that they could always be placed in the same position with respect to each other. In order to reduce spurious counts to a minimum all electrical connections were shielded and all counting was carried out with the counter inside a lead castle.

(b) Counting system

An R.C.L. Mark 15 Model 1 combined pulse amplifier and high voltage supply was used to provide the electrode potential and to amplify the pulses initiated within the counter by the beta radiation. The voltage supply was continuously variable between 2300 and 5000 volts and its rated stability was 0.02 per cent change in output at 4500 volts for a 10 per cent change in the line voltage. The amplifier gain was 1860, the rise time was less than 0.5 microseconds and the decay time was less than 3.5 microseconds. The minimum input sensitivity was 2.8 millivolts and the negative output pulse was variable from zero to ten volts. The pulses after amplification were counted and recorded using an Electronic Associates scaler.

Using four different scalers and several $Sr^{90}- Y^{90}$ sources, the voltage plateau characteristics of the counting system were studied at various settings of the pulse height potentiometer of the linear amplifier. It was found that the plateau showed no measurable slope between 3000 and 3250 volts over a considerable pulse height setting range. On the basis of the plateau shown in Figure 2, the counter was operated at

FIG. 2 - 4π COUNTER VOLTAGE PLATEAU



3200 volts. The background, due mostly to cosmic rays, was of the order of one pulse per second.

The dead time of the counting system was measured experimentally by means of a split source, one that could be used with both halves together or each one separately. If C_1 and C_2 are the counting rates for the two halves separately and C_{12} is the rate for the two halves measured together then the dead time of system, t_r, can be calculated (56) from the formula

$$\frac{C_1}{1 - C_1 t_r} + \frac{C_2}{1 - C_2 t_r} = \frac{C_{12}}{1 - C_{12} t_r} \qquad [2]$$

Using a split source, C_1, C_2, and C_{12} were found to be 122.8, 123.3 and 238.7 respectively and from this a dead-time value of 2.5×10^{-4} seconds was obtained. This is in perfect agreement with the value fixed by the RC input circuit of the linear amplifier.

(c) Sample mounting

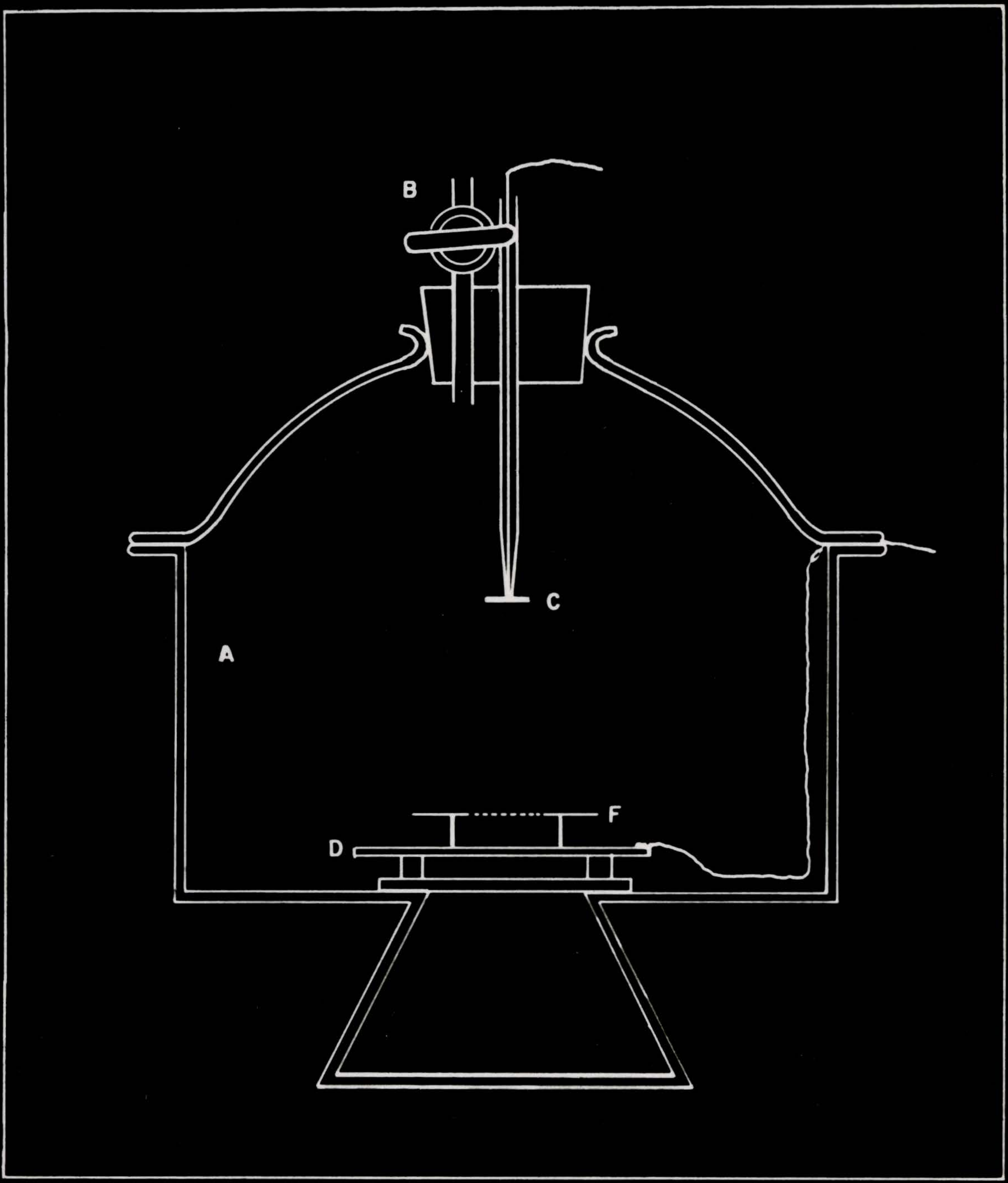
(1) Films

Disks (diameter 5.8 cm.) used for sample mounting were made from 0.005 in. sheet nichrome with central openings 2.5 cm. in diameter. It was necessary to make very thin plastic films which could be fastened across these openings and on which samples could be deposited. Many techniques for preparing thin films of such materials as polyethylene, zapon, formvar and vinyl plastics were investigated. It was found, however, that V.Y.N.S. (polyvinylchloride-acetate copolymer) plastic was most satisfactory. By "stretching" a solution of V.Y.N.S. in cyclohexanone on the surface of water, according to a technique developed by Pate and Yaffe (57), it was possible to make films of uniform thickness 140 to 210 mμ (i.e.,20 to 30 micrograms per sq. cm. superficial density). The film adhered securely to the disks when they were placed on top of the sheet of plastic about ten minutes after it had been stretched out on the surface of the water.

(2) Sputtering

If a radioactive source is placed on the surface of a non-conducting film, small charges build up as the source decays. According to Hawkings et al. (55) this would reduce the counting rate for a given source and result in the loss of voltage plateau. Consequently the films were made conducting by a process called sputtering using the apparatus shown in Figure 3. Inside a vacuum desiccator A evacuated by an oil pump through stopcock B the V.Y.N.S. films were sputtered with gold at a pressure of about 1 x 10^{-4} mm. and an electrode potential of about 3000 volts A.C. The upper electrode C consisted of a piece of 10K gold (approx. 0.63 x 0.75 x 0.05 in.) held in an aluminum frame while the lower one D was an aluminum disk. The nichrome disks F holding the thin films were supported above electrode D on a glass ring. Films were sputtered with gold on one side only to a thickness of 5 to 10 μgm. per sq. cm. at which point they were conducting.

FIG. 3 - SPUTTERING APPARATUS



Thicknesses of both plastic and gold films were estimated by means of interference patterns as described by Pate and Yaffe (57).

(d) Counting corrections and calibration of counter

In common with measurements of disintegration rates by all types of counting equipment, those by 4π counters are subject to corrections for background as well as coincidence loss which is a function of the dead time. The only other corrections which apply to 4π counting rate determinations are those for absorption losses in the source mount and for self-absorption in the source.

(1) Absorption in the source mount

Seliger and Cavallo (58) at the National Bureau of Standards have shown that reduction of the counting rate due to absorption in the source mount, if no allowance for backscatter is made, is given by

$$N_o = N_{tb}/(1 - \tau/2) \qquad [3]$$

where N_o = the absolute counting rate

$\tau = (N_t - N_b)/(N_{tb} - N_b)$, i.e., the fractional absorption in the film.

t and b refer to the upper and lower halves of the counter.

Table I shows two typical sets of values obtained using $Sr^{90} - Y^{90}$ sources for which equation [3] gives a correction of approximately 0.2 per cent.

TABLE I

Source-mount Absorption Correction

	N_t Count from Upper Half	N_b Count from Lower Half	N_{tb} Count from Both Halves	Correction Using Seliger's Formula (58)
I	141.6	141.3	283.4	0.3
II	128.6	128.2	250.0	0.4

Mann and Seliger (59) have shown that neglect of backscatter in the above relation introduces an uncertainty never greater than 1.5 per cent. However, Pate and Yaffe describe an absorption curve method which indicates an even smaller correction in the case of the beta particles dealt with in this work (i.e., with maximum energies no lower than 0.5 Mev). They maintain that both the N.B.S. method given above and the sandwich method described by Hawkings and co-workers (55) for determining the source-mount absorption correction give erroneous results. Consequently no adjustment of the measured disintegration rates was made because it would have had no significance when compared to the limits imposed by sample preparation.

(2) Self-absorption

Self-absorption in the source material itself will result in a reduction of the counting rate but this can be kept to a negligible minimum by using submicrogram quantities of carrier-free nuclides. Since the quantity of strontium or cesium on any one film was of the order of 10^{-12} gm., which even with a microgram of inherent impurities would result in negligible self-absorption, it was assumed that there was no reduction in the counting rate as a result of finite source thickness.

(3) Conversion of counting rates to disintegration rates

Sr^{90} decays by the emission of a single beta particle to Y^{90} with which it is in equilibrium so that every time a Sr^{90} nucleus expels an electron an Y^{90} nucleus also expels an electron. This means that only half a measured counting rate is due to the disintegration of $Sr^{90}$and hence all such measurements must be corrected for this effect.

In the case of cesium, however, the more complicated decay scheme necessitates additional corrections. The gamma radiation from the isomeric state of Ba^{137} (2.6 min. half-life) is not in coincidence with the beta emission. Hence the observed counting rates were corrected for the contribution of conversion electrons and for secondary electrons produced by the gamma radiation. The correction for the conversion electrons was made assuming 92 per cent decay to metastable Ba^{137} (60) and an internal conversion coefficient of 0.118 (61). In order to correct for secondary electrons the gamma radiation efficiency of the proportional counter was estimated using aluminum sandwich techniques (62) and found to be less than 0.27 per cent. This compares reasonably well with the 0.15 per cent value obtained from the data of Bradt et al. (63). A value of 0.2 per cent has been used to correct the observed counting rate and, since the correction is itself small, an uncertainty in the value has little influence on the probable error of the measured disintegration rate.

The fission product Cs^{137} used in the half-life determination was contaminated by Cs^{134} formed by an (n,γ) reaction with Cs^{133}. It was therefore necessary to subtract, from each sample counted, the contribution of Cs^{134} to the observed counting rate. Two sodium iodide scintillation spectrometers were used to determine this correction. With these spectrometers the coincidence rate due to the 0.605- and 0.796-Mev cascade gamma rays of Cs^{134} was compared under conditions of known geometry with the single-channel counting rate due to the 0.661-Mev gamma ray of Cs^{137}. The ratio of these rates was 1/1204 and the relationship between this and the relative contributions to the measured disintegration rate is given by the equation

$$\frac{\text{count rate of Cs}^{134}}{\text{count rate of Cs}^{137}} = \frac{\text{corrected coincidence rate}}{\text{corrected single-channel rate}} \times \begin{matrix}\text{relative}\\ \text{efficiencies}\end{matrix} \times \begin{matrix}\text{geometry}\\ \text{factor}\end{matrix} \qquad [4]$$

Correction of the coincidence and single channel rates for the various modes of decay was made on the basis of the latest decay schemes (61)(64). The relative efficiencies of the NaI (Tl) crystals was calculated as 1.06 on the basis of gamma ray energies from data in the Handbook of Chemistry and Physics, (33rd ed.). The geometry factor was needed to adjust the coincidence rate with respect to the single-channel rate since the former depended on the relative positions of both scintillation spectrometers and that of the source. The absolute beta disintegration rate of the source as measured by a G.M. end-window counter divided by the single channel counting rate i.e., $7.00 \times 10^5/3.46 \times 10^4$ is the geometry factor that was used. Substitution of the above values in equation [4] resulted in a 3 per cent correction to the observed beta activity with an estimated precision of ± 0.5 per cent. This agrees with the value of 3 per cent based on mass spectrometric values of the Cs^{133}/Cs^{134} ratio in the same sample measured during the latter part of 1952.

(4) Efficiency of counting system

Because the efficiency of the counter is directly dependent on its geometry the sample mounting disks were designed so that, allowing for the size of the central hole, the metal of the disk would subtend a solid angle of less than 1 per cent of 4π steradians. The effective geometry of the counter for beta particles would, however, be 4π steradians since most of the betas which strike the source mount would be expected to trigger the counting circuit.

Hawkings et al. (55) have shown how sample position affects the efficiency of the counter. Their data for a methane-filled counter indicates that if the sample were placed within a circle of 1 cm. radius at the centre of the film, efficiency would be better than 99 per cent. Since in this work the actual deposits of carrier-free activity never covered an area more than 0.5 cm. in diameter, no correction for this effect was made.

The absolute efficiency of the counter was tested by means of P^{32} and Au^{198} standard solutions obtained from the National Bureau of Standards. The results of a comparison between the observed disintegration rates for these radioactive solutions and the rates obtained previously at the N.B.S. are shown in Table II. The deviations from the average reported for this work represent uncertainties in the sampling technique since each count corresponds to a separate dilution from the standard solutions. There is also, however, further source of error resulting from the uncertainty of the half-lives since the standards were fairly old and the half-lives are short, 14.60 days for P^{32} (65) and 2.69 days for Au^{198} (66). Counts were taken through two half-lives of P^{32} and through eight half-lives of Au^{198}. On the basis of these measurements the counter was assumed to be more than 99 per cent efficient for beta particles in this energy range.

Calibration of Counter with Solution Standards

TABLE II(a)

P^{32}

Sample	Observed Disintegrations per second	Time Elapsed in Hours Since Standardization	Corrected Disintegrations per second per ml. x 10^{-6}
1	76.2	247.0	159
2	73.2	264.3	159
3	71.7	290.5	163
4	14.4	795.0	160
5	12.9	852.5	160
6	9.8	867.5	157
		Average	160 ± 1%
		N.B.S.Value	160 ± 2%

TABLE II(b)

Au^{198}

Sample	Observed Disintegrations per second	Time Elapsed in Hours Since Standardization	Corrected Disintegrations per second per ml. x 10^{-5}
1	17.6	209.0	3.80
2	14.5	228.5	3.85
3	128.2	468.0	3.90
4	97.0	492.3	3.82
5	66.3	529.5	3.90
6	64.9	530.2	3.85
		Average	3.85 ± 2%
		N.B.S.Value	3.84 ± 3%

(B) Mass Spectrometry and Isotope Dilution

The concentration determinations were carried out by isotope dilution techniques using a 90 degree sector mass spectrometer with magnetic scanning and a hot filament ion source. The filaments, made from 0.030 inch tungsten ribbon, were platinum-plated and then heated for approximately one hour under vacuum at about 2000 degrees C. Sample solutions were evaporated in a teflon dish under a heat lamp to a volume of approximately 30 μl, then transferred and evaporated on to the filament.

The isotope dilution method for finding isotope concentrations was carried out as follows. In the case of Sr^{90}, the ratio of Sr^{88} to Sr^{90} in the solution was measured; a known amount of Sr^{88} (about 0.125 μgm.) was added to 1 gm. of solution and the ratio was measured again. These mass spectrometric ratios and the weight of added Sr^{88} were then utilized to obtain the amount of Sr^{90} per unit weight of solution. The same procedure was used to obtain the amount of Cs^{137} except that in this case stable Cs^{137} was used as the isotope diluting agent.

(C) Solution Handling

The Sr^{90} for both the counting and mass spectrometry came from a solution containing about 0.1 μgm. per gm.,of fission product Sr^{90}. Since the sample was over three years old, the 55-day Sr^{89} originally present would have decayed to the extent that its contribution to the activity was less than 0.1 per cent of the total. The Cs^{137} used for the half-life determination had been isolated carrier-free from fission products by ion exchange techniques. The age of the fission products was such (over three years) that the contribution to the counting rate by 2.3-year Cs^{134} was small but measurable. For both half-life determinations one gram of solution was weighed out directly for the mass spectrometry but more dilute solutions were necessary in order that a measurable volume could be used for counting. The radioactive purity of the solutions used was found to be greater than 99 per cent by determining the aluminum absorption curve for representative aliquots. An "$Fe(OH)_3$" scavenge carried out on the Cs^{137} solution indicated the presence of nothing but alkali metal radioactivity.

Aliquots of 100μl. or less were measured with a Gilmont ultramicroburette which, on recalibration using a semimicro balance, was proved to have a reproducibility of 1μl. Adsorption losses of the carrier-free radioisotopes from the dilute solutions used could have led to serious errors. To minimize these losses, all glassware was coated with "desicote" and all solutions were kept acidic. In order to test for fractionation in the strontium work, yttrium carrier was added to some solutions used for counting, strontium carrier was added to others. This did not change the observed counting rates so it was concluded that adsorption losses were negligible. Further tests were carried out by using Sr^{88} solutions of widely differing concentrations for isotope dilution and again no change could be detected. Similar experiments using Cs^{133} indicated that there were no appreciable adsorption losses in the cesium determination.

RESULTS AND DISCUSSION

(A) Calculation of Half-Lives

(a) Disintegration rate determinations

Table III shows the activity of the solution containing Sr^{90} and Y^{90} in equilibrium proportions. The values for samples 1, 2 and 3 are equilibrium counting rates obtained 300 hours after sample preparation. A change in count with time for these samples showed that Y^{90}, adsorbed during sample preparation, was regaining its equilibrium concentration. Sample 4 was taken from a solution to which yttrium carrier had been added and sample 5 contained both strontium and yttrium carrier; these showed no change in counting rate with time.

TABLE III

Activity of Solution Containing Sr^{90} and Y^{90} in Equilibrium

Sample	Disintegrations per second on film*	Disintegrations per second per ml. of Sr^{90}-Y^{90} Equilibrium Sol'n x 10^{-6}
1	57.20	1.327
2	57.36	1.331
3	56.81	1.320
4	51.34	1.334
5	51.30	1.332
	Average	1.329 ± 0.006

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*Corrected for coincidence loss and background but not for aliquot size.

Table IV shows the corresponding results for the cesium disintegration rate determinations. Cs^{133} carrier was added to the last two samples as a test for adsorption losses. Any adsorption which did take place appears to have been negligible.

TABLE IV

Activity of Cs^{137} Solution

Sample	Disintegrations per second on film	Disintegrations per second per ml. of Cs^{137} Solution x 10^{-5} *
1	31.27	1.298
2	31.35	1.305
3	31.38	1.307
4	31.25	1.297
5	31.33	1.303
6	31.30	1.300
7	31.40	1.308
	Average	1.303 ± 0.004
	Corrected for internal conversion electrons and for secondary electrons	1.174 ± 0.004
	Corrected for Cs^{134} contribution	1.139 ± 0.004

*Corrected for coincidence loss and background

(b) Isotope dilution

The agreement between the concentration values obtained for the Sr^{90} solution in this investigation (numbers 1, 2, and 3) and those obtained by other experimenters in the same laboratory (numbers 4 and 5) is shown in Table V.

Values 1 and 2 were obtained using a solution containing 1 μgm. per ml. of Sr^{88} for isotope dilution whereas 3, 4 and 5 were obtained using Sr^{88} solution of concentration 0.1 μgm. per ml.

Concentrat

Sample	Micrograms of Sr^{90} per ml. of Solution
1	0.127
2	0.124
3	0.123
4	0.125
5	0.123
Average	0.124 ± 0.002

In Table VI, the concentration of the Cs^{137} solution is reported. Two Cs^{133} solutions with different concentrations were used for isotope dilution, one for the first three samples and the other for samples 4 and 5. This had no apparent effect on the concentration value obtained.

TABLE VI

Concentration of Cs^{137} Solution

Sample	Micrograms of Cs^{137} per ml. of Solution
1	0.0310
2	0.0315
3	0.0320
4	0.0311
5	0.0314
Average	0.0314 ± 0.0004

The limits of error shown in Tables III to VI are the standard deviations of the independently determined values in each table and are considered to represent limits imposed by the measurement of small volumes of solution.

Substitution of the absolute disintegration rate (corrected for Y^{90} contribution) from Table III and the number of atoms present from Table V into equation [1] gives a half-life of 27.7 ± 0.4 years for Sr^{90}. Similarly the average values from Tables IV and VI give a half-life for Cs^{137} of 26.6 ± 0.4 years. The precision given in both cases is the standard deviation calculated on the basis of the precision reported for the average concentrations and disintegration rates.

(B) Comparison with Currently Accepted Half-Lives

(a) Strontium 90

Powers and Voigt (51) followed the decay of a Sr^{90} sample for a period of seven years with a Lauritsen electroscope. They obtained the currently accepted value of 19.9 ± 0.3 years for the half-life, a value considerably lower than the 27.7 ± 0.4 year value resulting from this work.

Fission yield studies of U^{235} provide evidence for choosing between the 19.9 year value and the 27.7 year value. It follows from equation [1] that the fission yield of Sr^{90} may be evaluated from a determination of the Sr^{90} disintegration rate corresponding to a given amount of U^{235} fissions. Glendenin's value of 25 years (50) for the half-life of Sr^{90} assumed the yield of this isotope to be on a smooth fission yield curve. The 19.9 year half-life would lead to a fission yield some 20 per cent lower whereas the 27.7 year half-life would correspond to a value about 11 per cent above the smooth yield curve. Investigations in this laboratory (67) show that the fission yield of Sr^{90} is high and that the corresponding fission fragment of mass 144 has a yield about 7 per cent above the smooth fission yield curve. These data tend to support the value of 27.7±0.4 years for the half-life of Sr^{90}.

In a private communication, Glendenin and Steinberg (68) reported a value of 28.2 years for the half-life of Sr^{90} by using the same method employed in this investigation. They have not, however, indicated any precision for their result. In addition Dr. Voigt has expressed the opinion (69) that there may have been loss of sample during the course of the change-in-counting-rate-with-time studies leading to the 19.9 year value (51). This would have resulted in too rapid a change in the counting rate and consequently too short a half-life.

(b) Cesium 137

Recently, workers at Harwell reported $30.0 \pm {}^{0.3}_{0.4}$ years as the half-life of Cs^{137} (70). Their experiments were based on the same approach that was used in this work, i.e., an absolute disintegration rate determination and isotope dilution along with the mass spectrometer. However they used, as sample mounts, films with superficial densities as high as 250 μgm. per square cm. and yet the correction they applied for absorption in the source mount was only 1 per cent. Any of the three methods mentioned in part (A)(d)(1) of the experimental section (N.B.S., sandwich or absorption curve) would predict a larger correction. Nevertheless this discrepancy is not of sufficient magnitude to account for a difference of 3.4 years in half-life results.

The currently accepted value of 33 ± 2 years (8) for the half-life of Cs^{137} was calculated from changes in the mass spectrometric ratios of Cs^{133}/Cs^{137} for fission products differing up to 5.4 years in age. More recent studies of similar ratios have been made in this laboratory with fission products differing up to 8.3 years in age and these results, shown in Table VII, give a half-life of 26.5 ± 2 years (71). Any variation in the relative yields of the samples compared in this manner would materially affect the calculated half-life, particularly since the change in the Cs^{133}/Cs^{137} ratio, even in 8 years, is small. Both the 26.5- and 33-year values were obtained by comparison of different fission product samples and hence depend on the assumption that changes in the relative fission yields due to different irradiation conditions are small. In addition, it should be noted that all the half-lives in Table VII depend on the Cs^{137}/Cs^{133} ratio of one sample at two different ages. If this had been contaminated, all the calculated half-lives would be lower than the real value. Nevertheless, it is felt that the 26.5-year value based on completely independent measurements does support the 26.6-year half-life found in this investigation.

TABLE VII

Half-Life Values from Changes in the Cs^{137}/Cs^{133} Ratio With Time

Sample	Time Since Irradiation in Years	Age Difference in Years with respect to Sample 7	Age Difference in Years with respect to Sample 7*	Cs^{137}/Cs^{133} determined with a mass spectrometer	Half-life in years from Sample 7	Half-life in years from Sample 7*
1	1.282	6.628	8.354	0.9074	26.2	27.3
2	1.470	6.440	8.166	0.9058	25.7	26.9
3	1.767	6.143	7.869	0.925	25.0	26.5
4	1.915	5.995	7.721	0.8929	26.0	27.4
5	2.282	5.628	7.354	0.8889	25.3	26.6
6	2.360	5.550	7.276	0.8757	27.5	28.5
7	7.910			Average	26 ± 1.5	
7*	9.636				Average	27.5 ± 1.6
				Overall Average	26.5 ± 2	

7 and 7* are the same sample at two different ages.

(C) Indications of Future Research

Sr^{90} and Cs^{137} are most important because they are relatively long-lived and plentiful in fission products. Measurement of these two half-lives was therefore of considerable interest but the method reported in this thesis is by no means limited to two isotopes. The same techniques could be applied to fission product Sm^{151} as well as to many other radioisotopes with similar half-lives. For example, any half-life between that of 2.3-year Cs^{134} and 400 year-La^{137} could be measured this way. Depending on improvements in counters and counting techniques, the range could be extended much farther. Alpha emitters would be easier to work with than beta emitters since the former involve mono-energetic radiation and absolute disintegration rate determinations would be correspondingly simplified.

The scope of the isotope dilution method could include the determination of half-lives from the rate of product formation. For example, in the determination of the half-life of tritium (section (A) (b), Historical Introduction) isotope dilution could be used to measure very small amounts of He formed. Clearly, the method is applicable to a wide range of alpha, beta or gamma half-lives of both solid and gaseous radioisotopes.

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