NBS CIRCULAR 594

Preparation, Maintenance, and Application of Standards of Radioactivity

UNITED STATES DEPARTMENT OF COMMERCE

NATIONAL BUREAU OF STANDARDS

The National Bureau of Standards

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NATIONAL BUREAU OF STANDARDS • A. V. Astin, *Director*

Preparation, Maintenance, and Application of Standards of Radioactivity

W. B. Mann and H. H. Seliger

National Bureau of Standards Circular 594
Issued June 11, 1958

For sale by the Superintendent of Documents, U. S. Government Printing Office
Washington 25, D. C. - Price 35 cents

Preface

The present Circular, which is a revision and unification of two review articles that appeared in the International Journal of Applied Radiation and Isotopes, attempts to survey the current position in the field of radioactivity standardization.

The most important aspects of this revision have been the inclusion of recent work to bring the text up to date, the use of a modified Harvard system of references and the incorporation of figures that illustrate specifically the methods used at the National Bureau of Standards in the field of radioactivity.

The National Bureau of Standards wishes to thank the Editors of the International Journal of Applied Radiation and Isotopes and the Pergamon Press Limited for their kind permission to use material in the two review articles, referred to above, in this Circular.

A. V. Astin, *Director.*

Contents

Preparation, Maintenance, and Application of Standards of Radioactivity

W. B. Mann and H. H. Seliger

The methods available for the preparation and maintenance of primary and secondary standards of radioactivity are reviewed, and the applications of such standards to problems in physics are discussed.

1. Introduction

The oldest standards of radioactivity are those of radium. Just 62 years ago, in February 1896, the radioactivity of uranium was discovered by Henri Becquerel, and before the turn of the century Marie Curie, investigating the high activity arising from Bohemian pitchblende, had isolated polonium and radium.

With the great outburst of research activity that these startling new discoveries generated, it soon became essential to establish international radium standards in order that the results obtained in different countries might be related one to another. Accordingly, in 1910, the Congress of Radiology and Electricity, held in Brussels, appointed a committee to arrange for the preparation of an international primary standard of radium and of international secondary standards.

In August 1911, Mme. Curie prepared, in Paris, a standard of radium sealed in a thin-walled glass tube and comprised of 21.99 mg of the pure anhydrous radium chloride that she had used to determine the atomic weight of radium to be 226.0. At the same time, Professor Otto Hönigschmid, at the Academy of Sciences in Vienna, prepared three standards of radium containing 10.11, 31.17, and 40.43 mg of radium chloride from material that he had used to obtain a value of 225.97 for the atomic weight of radium. In March 1912 the committee that had been appointed by the Congress of Radiology and Electricity in 1910, met in Paris to compare the radium standard of Mme. Curie with those of Professor Hönigschmid.

As a result of these comparisons, the Paris standard was accepted as the international primary standard of radium, while that consisting of 31.17 mg of radium chloride was designated as the international secondary standard. These two standards, which are now generally referred to as the 1911 Paris and Vienna standards, were placed respectively in the custody of the Bureau International des Poids et Mesures at Sèvres and of the Institut für Radiumforschung in Vienna. All subsequently prepared national radium standards were, for the next two decades, compared by measuring the relative gamma-ray activities with the 1911 Paris and Vienna standards.

By 1934, however, some concern was beginning to be felt for the safety of the old international primary and secondary standards, on account of the possibility of their breakage due to the devitrification under radiation of the glass tubes in which they were sealed and the accumulation of helium and chlorine within them. Accordingly, the International Radium Standards Commission invited Hönigschmid to prepare a new set of standards, using the radium salt with which he was making, at that time, in Munich, a precise determination of the atomic weight of radium. This salt, the chloride of radium, had been purified by Hönigschmid to such a point that spectroscopic analysis by Gerlach showed a maximum of not more than 0.002 to 0.003 atom percent, of barium. The purity of the radium chloride is, so to speak, defined by the value then obtained by Hönigschmid for the atomic weight of radium, namely 226.05, which is the value that is still current.

On the afternoon of 2 June 1934, Hönigschmid prepared twenty new standards of radium by sealing into thin-walled glass tubes quantities varying from 12.53 mg to 137.34 mg of this highly purified anhydrous chloride of radium from which the radium D had been separated on 25 May 1934 (Meyer, 1945; Hönigschmid, 1945).* One of these, containing as of that date, 22.23 mg of radium chloride, was selected as the new international primary standard of radium for custody in Paris, and its value was carefully compared with that of the 1911 Paris standard by gamma-ray measurements over a period of 4 years (Chamié, 1940). The remaining Hönigschmid standards were designated for use as international secondary or national standards of radium.

With the advent of controlled nuclear fission and the consequent large-scale production and use of artificially prepared radioactive materials, the need for radioactivity standards of elements other than radium quickly arose. In the first place, approximate standards were often prepared in the establishments in different countries that were producing such radioactive materials in order to provide them with a "yardstick" for the supply of radioactive materials in bulk. However, with the increasingly important applications of radioactive materials to research and therapy, accurate standards of radioactivity became as essential as, some 40 years earlier, they had become with radium. Once more the standards were required not only for comparison of results between laboratories in one country, but also for international comparisons.

Accordingly, in 1951, representatives of the

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*See references on page 42.

three countries that were at that time chiefly interested in the creation of international standards of radioactivity, Canada, the United Kingdom, and the United States of America, met at the Atomic Energy Research Establishment at Harwell, England, to discuss the preparation and maintenance of radioactivity standards of the artificially prepared radioactive isotopes of elements. As a result, it was agreed to exchange for intercomparison purposes suitable samples of the following radioactive nuclides, namely, carbon-14, sodium-24, phosphorus-32, cobalt-60, bromine-82, strontium-90-yttrium-90, iodine-131, thallium-204, and gold-198. Responsibility for the establishment of such standards in the three countries was vested in the National Bureau of Standards at Washington, D. C., the National Physical Laboratory at Teddington, and the National Research Council at Chalk River. The National Research Council's responsibilities were subsequently assumed, however, by Atomic Energy of Canada Limited with regard to the artificially prepared radioactive nuclides, while the Canadian National Radium Standard continued in the custody of the National Research Council at Ottawa.

Within the United Kingdom and the United States three advisory committees or subcommittees have now been set up to provide a forum for discussion amongst those interested in the field of radioactivity standards. These are the British National Physical Laboratory's Advisory Committee on Radioactive Standards; the U. S. National Research Council's Nuclear Science Committee's Subcommittee on Beta- and Gamma-ray Measurements and Standards; and the U. S. National Committee on Radiation Protection and Measurement's Subcommittee on Standards and Measurements of Radioactivity for Radiological Use. These latter include Canadian representation, and the National Research Council's subcommittee meetings in Washington, D. C., in 1954, at Chalk River, Ontario, in 1955, at Richmond and Berkeley, California, in 1956, and at Easton, Maryland, in 1957, were also attended by British representatives. The name of the National Research Council's subcommittee was changed, subsequent to its 1956 California meeting, to the Subcommittee on Measurements and Standards of Radioactivity in order that it might not be precluded in its deliberations from considering other than beta and gamma emitters.

It had, however, always been the aim that the cooperation that had been created between the United States, Canadian, and British laboratories should be extended as quickly as possible to all interested countries. Accordingly, in 1953, at its meeting in Copenhagen, the International Commission on Radiological Units created a Subcommittee on Standards of Radioactivity, to be composed in the first instance of representatives of the Federal Republic of Germany, France, The Netherlands, Sweden, the United Kingdom, and the United States. This subcommittee met in Frankfurt-am-Main in August 1955 to discuss the preparation, preservation, and propagation of international standards of radioactivity, and the hope was expressed at that meeting that these activities, that were so intimately related to the progress, prosperity, and health of all mankind, might soon be extended to all those countries, that were interested, throughout the world.

In April 1956 the International Commission on Radiological Units met again in Geneva and received the report of its subcommittee on radioactivity standards which met concurrently with the main commission. At this time the name of the subcommittee was changed to Committee I on Standards and Measurement of Radioactivity for Radiological Use and its membership rotated and extended to include a representative of Norway, in place of the representative of Sweden, and a representative of Uruguay. The report of the subcommittee at Geneva has now been published in the National Bureau of Standards Handbook 62, "Report of the International Commission on Radiological Units and Measurements (ICRU) 1956."

For convenience, the consideration in this Circular of methods of measurement will be broken down into sections dealing with primary and secondary methods of standardization. The dividing line between a primary method and a secondary method is, however, often extremely thin and difficult to place. It might be argued, for example, that the calorimetric method for determining disintegration rates is "secondary" because it requires a knowledge of the average energy per disintegration; it will be found, however, that in the case of those radioactive elements for which the the details of the disintegration scheme are known, calorimetry has been placed in the "primary" group, whereas in the case of radium in equilibrium with its products, where the disintegration data are complicated, if not nebulous, it has been considered to be a "secondary" method. On reflection, it might be said that in general a primary method is one by which the initial standardization is made or one in which a fundamental quantity such as disintegrations per second, or rate of energy emission per gram is measured. A secondary method, on the other hand, is one in which a calibration is related to an earlier primary calibration or in which a fundamental quantity is measured in terms of a similar property that has already been measured for another radioactive nuclide, such as radium in equilibrium with its products. But lest the adherents to any particular method should feel that less than justice might have been done to it, let it be immediately placed on record that no kind of stigma whatsoever, beyond that of mere convenience, attaches to either classification.

No attempt has been made within the confines of this Circular and its bibliography to deal in detail with the wide field of radioactivity instrumentation. Many excellent and well-known textbooks are available that deal with the subject of counter- and ionization-chamber techniques. Only in cases where an instrument or method of measurement may have had a somewhat specific application to radioactivity standardization, have references been given in this review.

2. Methods of Measurement

2.1. Secondary Standardization of the Naturally Occurring Radioactive Elements

The international primary standards of radium are standards by mass (figs. 1, 2, and 3). Other radium preparations are normally compared with these by comparison of the gamma-ray effects after the preparation has been sealed about thirty days to enable the radium to come into equilibrium with its early daughter products; the principal gamma-ray contribution comes from the excited levels of radium C′ in the disintegration of radium C. Alternatively, radium preparations and standards may be intercompared calorimetrically or by the method of radon analysis.

The radioactivity of a given mass of radium is measured in terms of the curie. This unit was initially defined as the quantity of radon in equilibrium with 1 gram of radium. This quantity of radon was also found to be emitting alpha particles at a rate of about 3.7×10^{10} disintegrations per second, although in later years, with the improvements in measuring techniques, the measured value of this emission rate has departed significantly from 3.7×10^{10} disintegrations per second, with a trend toward lower values.[1] A recent and very careful determination of the specific activity of radium, which corresponds to the curie as first defined for radon in equilibrium with radium, has given a value of $(3.608 \pm 0.028)\times10^{10}$ alpha particles per second per gram (Kohman, Ames, and Sedlet, 1949). Consequently, in 1950, at the VIth International Congress of Radiology in London, the definition of the curie was changed to apply to the measurement of the disintegration rate of radioactive substances other than radium and radon, being defined as the quantity of any radioactive nuclide in which the number of disintegrations per second is 3.700×10^{10}.

One of the earliest and simplest methods for measuring the relative gamma-ray effects of radium preparations was that using the gold-leaf electroscope. This instrument has indeed been retained even till the present time in national standardizing laboratories (Curtiss, 1928; Perry, 1936) and gives good reproducibility with a standard deviation of the order of 0.5 percent, or even less with care, and is ideal for long-term trouble-free routine service (fig. 4). Ionization chambers, either singly or as a balanced pair (Rutherford and Chadwick, 1912) have also been used for the precise comparison of the gamma-ray effects from radium in equilibrium with its daughter products. The use of a balanced pair of ionization chambers has recently been carried to a high degree of precision, using the decay of a sealed source of radon in the one chamber to achieve an exact balance (Kipfer, 1953; Curie, 1954). The present position is that the relative gamma-ray effects of different preparations can be measured with a precision that is better than the uncertainties arising from the unknown self-absorption in the sources themselves, which is difficult to calculate.

The calorimetric method, in which the energy emission due to the alpha, beta, and gamma radiations is measured, is less dependent on differences in the gamma-ray absorption of the sources themselves. Some 93 percent of the energy emission of a radium sample in equilibrium with its products is corpuscular and, provided that most of the particles are absorbed, the measured relative values of different sources will be quite insensitive to any small changes of the order of 1 or 2 percent in the absorption of the gamma radiation.

The earliest experiments to measure the rate of energy emission of radium and its products by their heating effects were carried out within a few years of the discovery of radium, and published in 1903 (Curie and Laborde). Subsequently, attempts were made to measure the heating effects of radium and those of its products that were alpha emitters (Rutherford and Barnes, 1903; Schweidler and Hess, 1908; Meyer and Hess, 1912; Hess, 1912; Rutherford and Robinson, 1912; Rutherford and Robinson, 1913; Watson and Henderson, 1928; Zlotowski, 1935). Recently, the calorimetric method has been developed to compare radium preparations with a standard deviation of about 0.1 percent (Mann, 1954a and 1954b) (figs. 5 and 6).

It should perhaps be emphasized that where the electroscope can measure only relative quantities by the gamma-ray effect, the calorimeter, in the comparison of one radium preparation with another or with a standard, does measure absolutely a property of radium and its products, namely, the rate of energy emission per milligram of radium element.

The radon method of comparing radium preparations requires that the radon in equilibrium with the radium shall be removed and measured separately, nowadays usually by counting the radon alpha particles or those arising from its decay products (Rutherford and Boltwood, 1905 and 1906; Strutt, 1905; Paneth and Koeck, 1931; Evans, 1935; Urry, 1936; Urry and Piggot, 1941; Curtiss and Davis, 1943; Bryant and Michaelis, 1952; Cowper and Simpson, 1953). It is thus unsuited to the comparison of permanently sealed

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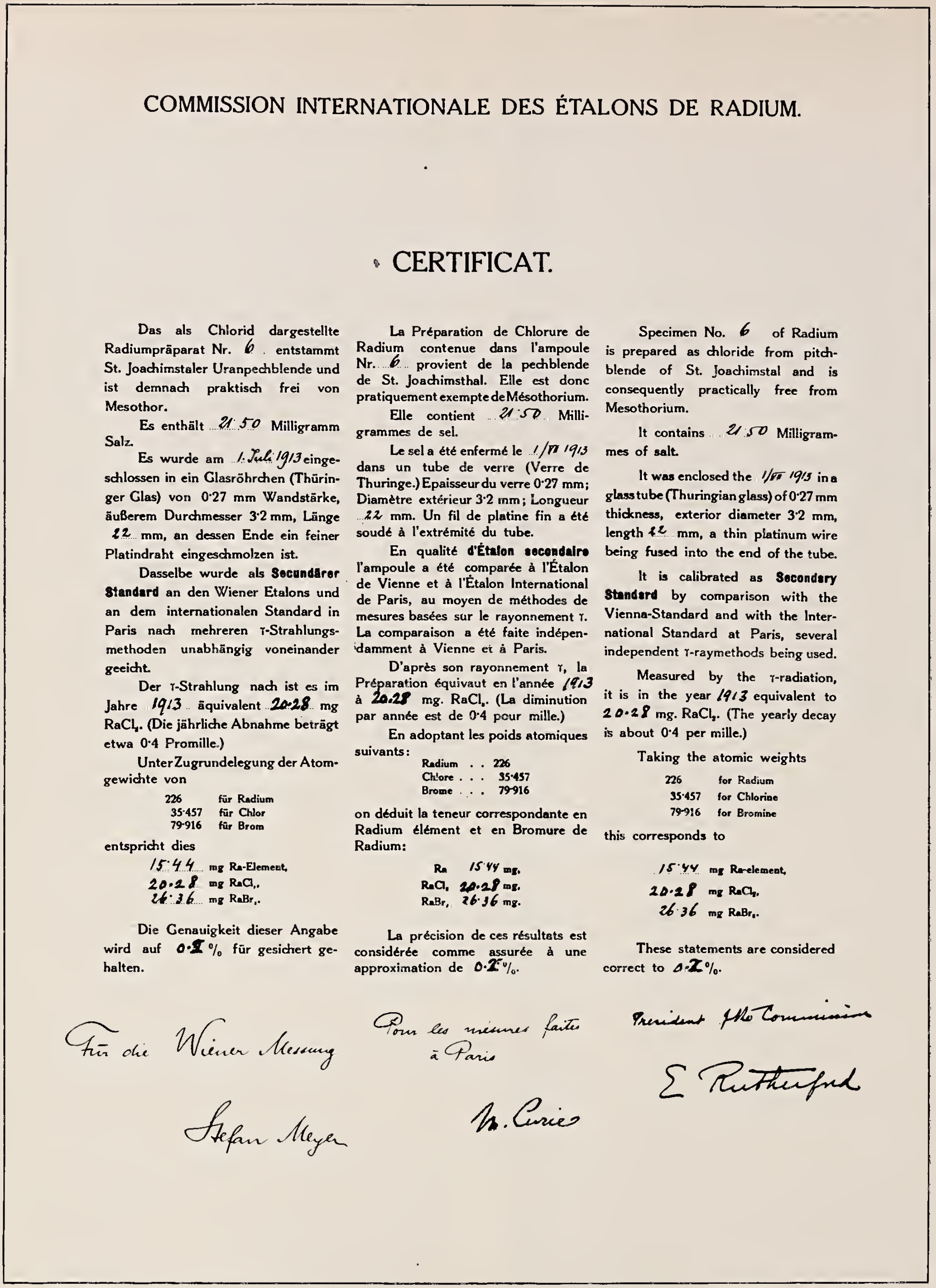
[1] A complete bibliography of these is given by Kohman, Ames, and Sedlet (1949).



COMMISSION INTERNATIONALE DES ÉTALONS DE RADIUM.

CERTIFICAT.

Das als Chlorid dargestellte Radiumpräparat Nr. 6 entstammt St. Joachimstaler Uranpechblende und ist demnach praktisch frei von Mesothor.

Es enthält 21·50 Milligramm Salz.

Es wurde am 1. Juli 1913 eingeschlossen in ein Glasröhrchen (Thüringer Glas) von 0·27 mm Wandstärke, äußerem Durchmesser 3·2 mm, Länge 22 mm, an dessen Ende ein feiner Platindraht eingeschmolzen ist.

Dasselbe wurde als **Secundärer Standard** an den Wiener Etalons und an dem internationalen Standard in Paris nach mehreren γ-Strahlungsmethoden unabhängig voneinander geeicht.

Der γ-Strahlung nach ist es im Jahre 1913 äquivalent 20·28 mg RaCl$_2$. (Die jährliche Abnahme beträgt etwa 0·4 Promille.)

Unter Zugrundelegung der Atomgewichte von

226	für Radium
35·457	für Chlor
79·916	für Brom

entspricht dies

15·44 mg Ra-Element,
20·28 mg RaCl$_2$,
26·36 mg RaBr$_2$.

Die Genauigkeit dieser Angabe wird auf 0·2 % für gesichert gehalten.

Für die Wiener Messung

Stefan Meyer

La Préparation de Chlorure de Radium contenue dans l'ampoule Nr. 6 provient de la pechblende de St. Joachimsthal. Elle est donc pratiquement exempte de Mésothorium.

Elle contient 21·50 Milligrammes de sel.

Le sel a été enfermé le 1/VII 1913 dans un tube de verre (Verre de Thuringe.) Epaisseur du verre 0·27 mm; Diamètre extérieur 3·2 mm; Longueur 22 mm. Un fil de platine fin a été soudé à l'extrémité du tube.

En qualité **d'Étalon secondaire** l'ampoule a été comparée à l'Étalon de Vienne et à l'Étalon International de Paris, au moyen de méthodes de mesures basées sur le rayonnement γ. La comparaison a été faite indépendamment à Vienne et à Paris.

D'après son rayonnement γ, la Préparation équivaut en l'année 1913 à 20·28 mg. RaCl$_2$. (La diminution par année est de 0·4 pour mille.)

En adoptant les poids atomiques suivants:

Radium	226
Chlore	35·457
Brome	79·916

on déduit la teneur correspondante en Radium élément et en Bromure de Radium:

Ra 15·44 mg.
RaCl$_2$ 20·28 mg.
RaBr$_2$ 26·36 mg.

La précision de ces résultats est considérée comme assurée à une approximation de 0·2 %.

Pour les mesures faites à Paris

M. Curie

Specimen No. 6 of Radium is prepared as chloride from pitchblende of St. Joachimstal and is consequently practically free from Mesothorium.

It contains 21·50 Milligrammes of salt.

It was enclosed the 1/VII 1913 in a glass tube (Thuringian glass) of 0·27 mm thickness, exterior diameter 3·2 mm, length 22 mm, a thin platinum wire being fused into the end of the tube.

It is calibrated as **Secondary Standard** by comparison with the Vienna-Standard and with the International Standard at Paris, several independent γ-raymethods being used.

Measured by the γ-radiation, it is in the year 1913 equivalent to 20·28 mg. RaCl$_2$. (The yearly decay is about 0·4 per mille.)

Taking the atomic weights

226	for Radium
35·457	for Chlorine
79·916	for Bromine

this corresponds to

15·44 mg Ra-element,
20·28 mg RaCl$_2$,
26·36 mg RaBr$_2$.

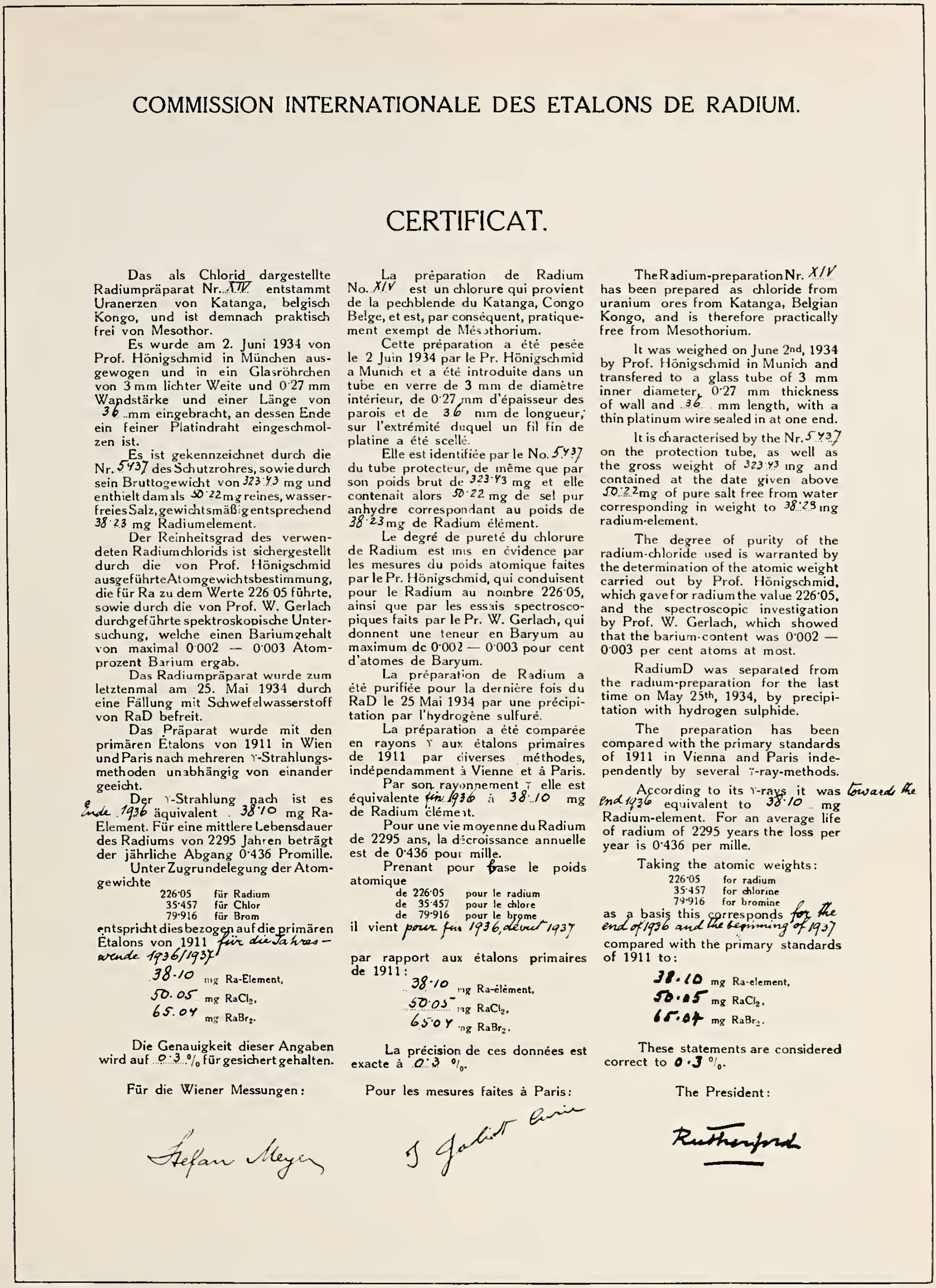
These statements are considered correct to 0·2 %.

President of the Commission

E Rutherford

FIGURE 1. *Certificate of the International Radium Standards Commission for the first United States Primary Radium Standard, Vienna No. 6 and International Commission No. IV, which was brought to America in 1913 by Mme. Marie Curie.*

It is interesting to note the somewhat large difference in the mass of the radium chloride as determined by weighing and gamma-ray measurement.



COMMISSION INTERNATIONALE DES ETALONS DE RADIUM.

CERTIFICAT.

Das als Chlorid dargestellte Radiumpräparat Nr. XIV entstammt Uranerzen von Katanga, belgisch Kongo, und ist demnach praktisch frei von Mesothor.

Es wurde am 2. Juni 1934 von Prof. Hönigschmid in München ausgewogen und in ein Glasröhrchen von 3 mm lichter Weite und 0·27 mm Wandstärke und einer Länge von 36 mm eingebracht, an dessen Ende ein feiner Platindraht eingeschmolzen ist.

Es ist gekennzeichnet durch die Nr. 5437 des Schutzrohres, sowie durch sein Bruttogewicht von 323·43 mg und enthielt damals 50·22 mg reines, wasserfreies Salz, gewichtsmäßig entsprechend 38·23 mg Radiumelement.

Der Reinheitsgrad des verwendeten Radiumchlorids ist sichergestellt durch die von Prof. Hönigschmid ausgeführte Atomgewichtsbestimmung, die für Ra zu dem Werte 226·05 führte, sowie durch die von Prof. W. Gerlach durchgeführte spektroskopische Untersuchung, welche einen Bariumgehalt von maximal 0·002 — 0·003 Atomprozent Barium ergab.

Das Radiumpräparat wurde zum letztenmal am 25. Mai 1934 durch eine Fällung mit Schwefelwasserstoff von RaD befreit.

Das Präparat wurde mit den primären Étalons von 1911 in Wien und Paris nach mehreren γ-Strahlungsmethoden unabhängig von einander geeicht.

Der γ-Strahlung nach ist es Ende 1936 äquivalent 38·10 mg Ra-Element. Für eine mittlere Lebensdauer des Radiums von 2295 Jahren beträgt der jährliche Abgang 0·436 Promille.

Unter Zugrundelegung der Atomgewichte

226·05 für Radium
35·457 für Chlor
79·916 für Brom

entspricht dies bezogen auf die primären Étalons von 1911 für die Jahreswende 1936/1937

38·10 mg Ra-Element,
50.05 mg $RaCl_2$,
65.04 mg $RaBr_2$.

Die Genauigkeit dieser Angaben wird auf 0·3 % für gesichert gehalten.

Für die Wiener Messungen:

Stefan Meyer

La préparation de Radium No. XIV est un chlorure qui provient de la pechblende du Katanga, Congo Belge, et est, par conséquent, pratiquement exempt de Mésothorium.

Cette préparation a été pesée le 2 Juin 1934 par le Pr. Hönigschmid a Munich et a été introduite dans un tube en verre de 3 mm de diamètre intérieur, de 0·27 mm d'épaisseur des parois et de 36 mm de longueur, sur l'extrémité duquel un fil fin de platine a été scellé.

Elle est identifiée par le No. 5437 du tube protecteur, de même que par son poids brut de 323·43 mg et elle contenait alors 50·22 mg de sel pur anhydre correspondant au poids de 38·23 mg de Radium élément.

Le degré de pureté du chlorure de Radium est mis en évidence par les mesures du poids atomique faites par le Pr. Hönigschmid, qui conduisent pour le Radium au nombre 226·05, ainsi que par les essais spectroscopiques faits par le Pr. W. Gerlach, qui donnent une teneur en Baryum au maximum de 0·002 — 0·003 pour cent d'atomes de Baryum.

La préparation de Radium a été purifiée pour la dernière fois du RaD le 25 Mai 1934 par une précipitation par l'hydrogène sulfuré.

La préparation a été comparée en rayons γ aux étalons primaires de 1911 par diverses méthodes, indépendamment à Vienne et à Paris.

Par son rayonnement γ elle est équivalente fin 1936 à 38·10 mg de Radium élément.

Pour une vie moyenne du Radium de 2295 ans, la décroissance annuelle est de 0·436 pour mille.

Prenant pour base le poids atomique

de 226·05 pour le radium
de 35·457 pour le chlore
de 79·916 pour le brome

il vient pour fin 1936, début 1937

par rapport aux étalons primaires de 1911:

38·10 mg Ra-élément,
50·05 mg $RaCl_2$,
65·04 mg $RaBr_2$.

La précision de ces données est exacte à 0·3 %.

Pour les mesures faites à Paris:

I Joliot Curie

The Radium-preparation Nr. XIV has been prepared as chloride from uranium ores from Katanga, Belgian Kongo, and is therefore practically free from Mesothorium.

It was weighed on June 2nd, 1934 by Prof. Hönigschmid in Munich and transfered to a glass tube of 3 mm inner diameter, 0·27 mm thickness of wall and 36 mm length, with a thin platinum wire sealed in at one end.

It is characterised by the Nr. 5437 on the protection tube, as well as the gross weight of 323·43 mg and contained at the date given above 50·22 mg of pure salt free from water corresponding in weight to 38·23 mg radium-element.

The degree of purity of the radium-chloride used is warranted by the determination of the atomic weight carried out by Prof. Hönigschmid, which gave for radium the value 226·05, and the spectroscopic investigation by Prof. W. Gerlach, which showed that the barium-content was 0·002 — 0·003 per cent atoms at most.

RadiumD was separated from the radium-preparation for the last time on May 25th, 1934, by precipitation with hydrogen sulphide.

The preparation has been compared with the primary standards of 1911 in Vienna and Paris independently by several γ-ray-methods.

According to its γ-rays it was toward the end 1936 equivalent to 38·10 mg Radium-element. For an average life of radium of 2295 years the loss per year is 0·436 per mille.

Taking the atomic weights:

226·05 for radium
35·457 for chlorine
79·916 for bromine

as a basis this corresponds for the end of 1936 and the beginning of 1937 compared with the primary standards of 1911 to:

38·10 mg Ra-element,
50·05 mg $RaCl_2$,
65·04 mg $RaBr_2$.

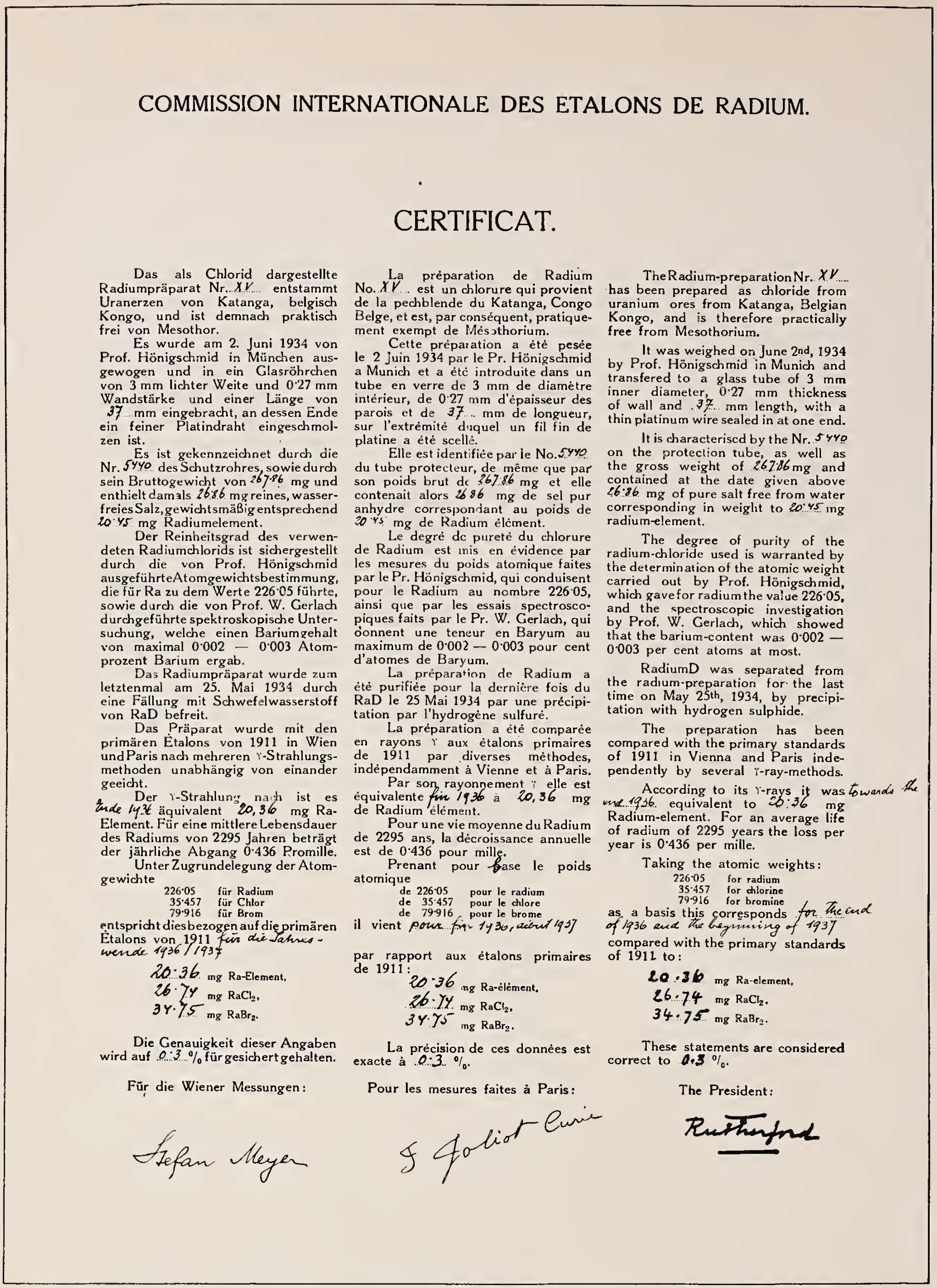
These statements are considered correct to 0·3 %.

The President:

Rutherford

FIGURE 2. *Certificate of the International Radium Standards Commission for United States Primary Radium Standard No. XIV.*

This standard which is Hönigschmid standard No. 5437, is actually about 36.7 mm in length. According to Stefan Meyer (1945), the Paris and Vienna gamma-ray measurements of the amount of radium chloride in this standard were respectively 50.03 and 50.08 mg.



COMMISSION INTERNATIONALE DES ETALONS DE RADIUM.

CERTIFICAT.

Das als Chlorid dargestellte Radiumpräparat Nr. XV entstammt Uranerzen von Katanga, belgisch Kongo, und ist demnach praktisch frei von Mesothor.

Es wurde am 2. Juni 1934 von Prof. Hönigschmid in München ausgewogen und in ein Glasröhrchen von 3 mm lichter Weite und 0·27 mm Wandstärke und einer Länge von 37 mm eingebracht, an dessen Ende ein feiner Platindraht eingeschmolzen ist.

Es ist gekennzeichnet durch die Nr. 5440 des Schutzrohres, sowie durch sein Bruttogewicht von 267·86 mg und enthielt damals 26·86 mg reines, wasserfreies Salz, gewichtsmäßig entsprechend 20·45 mg Radiumelement.

Der Reinheitsgrad des verwendeten Radiumchlorids ist sichergestellt durch die von Prof. Hönigschmid ausgeführte Atomgewichtsbestimmung, die für Ra zu dem Werte 226·05 führte, sowie durch die von Prof. W. Gerlach durchgeführte spektroskopische Untersuchung, welche einen Bariumgehalt von maximal 0·002 — 0·003 Atomprozent Barium ergab.

Das Radiumpräparat wurde zum letztenmal am 25. Mai 1934 durch eine Fällung mit Schwefelwasserstoff von RaD befreit.

Das Präparat wurde mit den primären Etalons von 1911 in Wien und Paris nach mehreren γ-Strahlungsmethoden unabhängig von einander geeicht.

Der γ-Strahlung nach ist es Ende 1936 äquivalent 20,36 mg Ra-Element. Für eine mittlere Lebensdauer des Radiums von 2295 Jahren beträgt der jährliche Abgang 0·436 Promille.

Unter Zugrundelegung der Atomgewichte

226·05	für Radium
35·457	für Chlor
79·916	für Brom

entspricht dies bezogen auf die primären Etalons von 1911 für die Jahreswende 1936/1937

20·36 mg Ra-Element,
26·74 mg $RaCl_2$,
34·75 mg $RaBr_2$.

Die Genauigkeit dieser Angaben wird auf 0·3 % für gesichert gehalten.

Für die Wiener Messungen:

Stefan Meyer

La préparation de Radium No. XV est un chlorure qui provient de la pechblende du Katanga, Congo Belge, et est, par conséquent, pratiquement exempt de Mésothorium.

Cette préparation a été pesée le 2 Juin 1934 par le Pr. Hönigschmid a Munich et a été introduite dans un tube en verre de 3 mm de diamètre intérieur, de 0·27 mm d'épaisseur des parois et de 37 mm de longueur, sur l'extrémité duquel un fil fin de platine a été scellé.

Elle est identifiée par le No. 5440 du tube protecteur, de même que par son poids brut de 267·86 mg et elle contenait alors 26·86 mg de sel pur anhydre correspondant au poids de 20·45 mg de Radium élément.

Le degré de pureté du chlorure de Radium est mis en évidence par les mesures du poids atomique faites par le Pr. Hönigschmid, qui conduisent pour le Radium au nombre 226·05, ainsi que par les essais spectroscopiques faits par le Pr. W. Gerlach, qui donnent une teneur en Baryum au maximum de 0·002 — 0·003 pour cent d'atomes de Baryum.

La préparation de Radium a été purifiée pour la dernière fois du RaD le 25 Mai 1934 par une précipitation par l'hydrogène sulfuré.

La préparation a été comparée en rayons γ aux étalons primaires de 1911 par diverses méthodes, indépendamment à Vienne et à Paris.

Par son rayonnement γ elle est équivalente fin 1936 à 20,36 mg de Radium élément.

Pour une vie moyenne du Radium de 2295 ans, la décroissance annuelle est de 0·436 pour mille.

Prenant pour base le poids atomique

de 226·05	pour le radium
de 35·457	pour le chlore
de 79·916	pour le brome

il vient pour fin 1936, début 1937

par rapport aux étalons primaires de 1911:

20·36 mg Ra-élément,
26·74 mg $RaCl_2$,
34·75 mg $RaBr_2$.

La précision de ces données est exacte à 0·3 %.

Pour les mesures faites à Paris:

F Joliot Curie

The Radium-preparation Nr. XV has been prepared as chloride from uranium ores from Katanga, Belgian Kongo, and is therefore practically free from Mesothorium.

It was weighed on June 2nd, 1934 by Prof. Hönigschmid in Munich and transfered to a glass tube of 3 mm inner diameter, 0·27 mm thickness of wall and 37 mm length, with a thin platinum wire sealed in at one end.

It is characterised by the Nr. 5440 on the protection tube, as well as the gross weight of 267·86 mg and contained at the date given above 26·86 mg of pure salt free from water corresponding in weight to 20·45 mg radium-element.

The degree of purity of the radium-chloride used is warranted by the determination of the atomic weight carried out by Prof. Hönigschmid, which gave for radium the value 226·05, and the spectroscopic investigation by Prof. W. Gerlach, which showed that the barium-content was 0·002 — 0·003 per cent atoms at most.

RadiumD was separated from the radium-preparation for the last time on May 25th, 1934, by precipitation with hydrogen sulphide.

The preparation has been compared with the primary standards of 1911 in Vienna and Paris independently by several γ-ray-methods.

According to its γ-rays it was towards the end 1936 equivalent to 20·36 mg Radium-element. For an average life of radium of 2295 years the loss per year is 0·436 per mille.

Taking the atomic weights:

226·05	for radium
35·457	for chlorine
79·916	for bromine

as a basis this corresponds for the end of 1936 and the beginning of 1937 compared with the primary standards of 1911 to:

20·36 mg Ra-element,
26·74 mg $RaCl_2$.
34·75 mg $RaBr_2$.

These statements are considered correct to 0·3 %.

The President:

Rutherford

FIGURE 3. *Certificate of the International Radium Standards Commission for United States Primary Radium Standard No. XV.*

This standard which is Hönigschmid standard No. 5440 is about 36.3 mm in length. The Paris and Vienna gamma-ray measurements of the amount of radium chloride in this standard, if one neglects an obvious misprint in the original paper (Meyer, 1945), were respectively 26.73 and 26.76 mg. Figures 1, 2, and 3 provide an interesting historical glimpse of the personal permanence of the Commission over a period of nearly a quarter of a century.

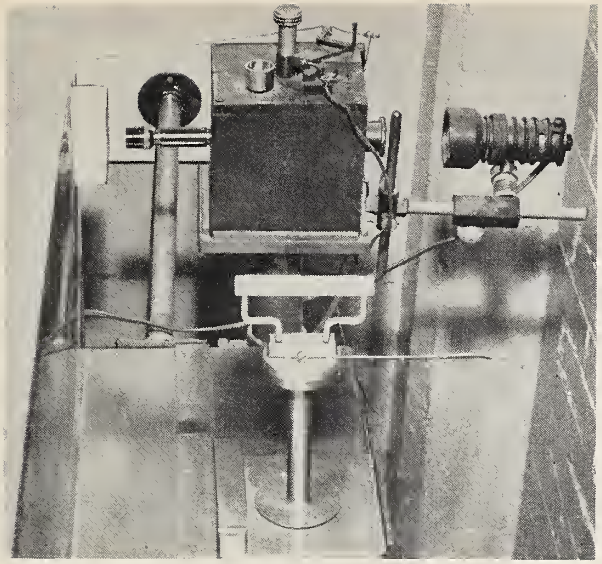


FIGURE 4. *The National Bureau of Standards standard gold-leaf electroscope (Curtiss, 1928).*

The ionization chamber consists of a 10-cm-cube free-air volume, with walls made of 1-cm thick lead sheet with ½-cm thick aluminum inner lining. A gold leaf is suspended near the center of the chamber and a quartz fiber, 10μ in diameter, at the free end of the leaf provides a fine line for optical projection, with a magnification of 100, onto a metric scale. Transit times of the quartz-fiber image between two fixed points on the scale 6-cm apart are normally measured.

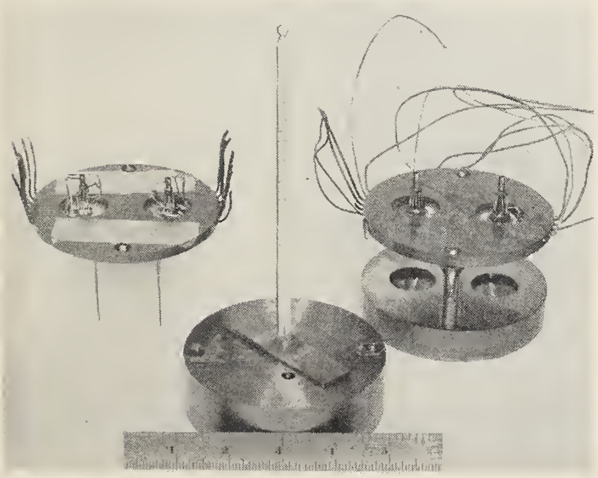


FIGURE 5. *Two early versions of the radiation balance or twin-cup Peltier-effect microcalorimeter (Mann, 1954a).*

The two copper disks on which the microcalorimeter cups are mounted both fit the same copper base and copper cover, the latter with thermometer. The microcalorimeter assembly on the left actually has four Peltier junctions attached to each cup, two above and two below the equatorially-located thermopile junctions. The assembly on the right has only one Peltier junction attached to the base of each cup. Provided that the thermal conduction in the cups is adequate, it appears that no difference in results is obtained by distributing the cooling along the cup. A precision of some ±0.5 percent was obtained with the single Peltier-junction microcalorimeter working with no thermal isolation of the microcalorimeter from its surroundings. The lighter shaded metal on the left-hand disk is of lead and was used to assure good thermal contact between the disks and the copper base and cover when assembled.

international or national radium standards, either with each other or with other radium preparations. In order to compare radium preparations with a radium primary standard, using the radon method, it is necessary that a secondary radon standard be prepared by comparing a suitable amount of radium salt with a primary radium standard, calorimetrically or by gamma-ray measurements. From this calibrated radium salt an aqueous solution is then prepared, from which equilibrium amounts of radon can be obtained by boiling or by bubbling through the solution an inert gas such as nitrogen. Initially, electroscopic, balanced ionization-chamber or ionization-current measuring methods were used to measure the relative amounts of radon collected (Rutherford and Boltwood, 1905 and 1906; Strutt, 1905; Paneth and Koeck, 1931; Evans, 1935; Urry, 1936; Urry and Piggot, 1941), but latterly these amounts have been determined by counting the alpha particles in ionization chambers (Curtiss and Davis, 1943) (figs. 7 and 8). Recently, however, scintillation counting techniques have been applied to the comparison of amounts of radon by measuring the alpha particles arising from radium-A recoils in a gas, these ionized radium-A atoms being attracted electrostatically to an extremely thin aluminum foil and counted by means of a thin scintillator of zinc sulfide, adjacent to the aluminum foil, used in conjunction with an electron-multiplier phototube (Bryant and Michaelis, 1952; Cowper and Simpson, 1953) (fig. 9). The radon method of analysis of radium gives results that are reproducible with a standard deviation of the order of 1.0 percent, and can, in addition, be used to determine the amount of radium present, for example, in water or in uranium ores, in the latter case giving a measure of their uranium content, provided that there has been no leaching and that the radium is in equilibrium with the uranium.

Other daughters of radium, and members of the uranium family, that are of importance in standardization are radium D+E, for beta-reference sources, and radium D+E+F, or radium F (polonium-210) alone, for alpha-ray standards. Radium D sources can be electrolytically or chemically deposited onto palladium-clad silver disks, and polonium-210 can either be grown from radium D or else plated from polonium-210 solution onto the same disks. In all three cases extremely thin sources of beta or alpha activity are formed. The disintegration rate of the radium D (lead-210), in equilibrium with the energetic beta emitter radium E, is determined by electrodepositing a known amount of lead that has been quantitatively extracted from a pitchblende, the radium content of which has previously been determined by the radon method. Secondary sources of radium D+E can then be calibrated by comparison with one of these primary radium D+E standards by means of an end-window Geiger-Müller counter. The alpha emission from thin alpha standards of

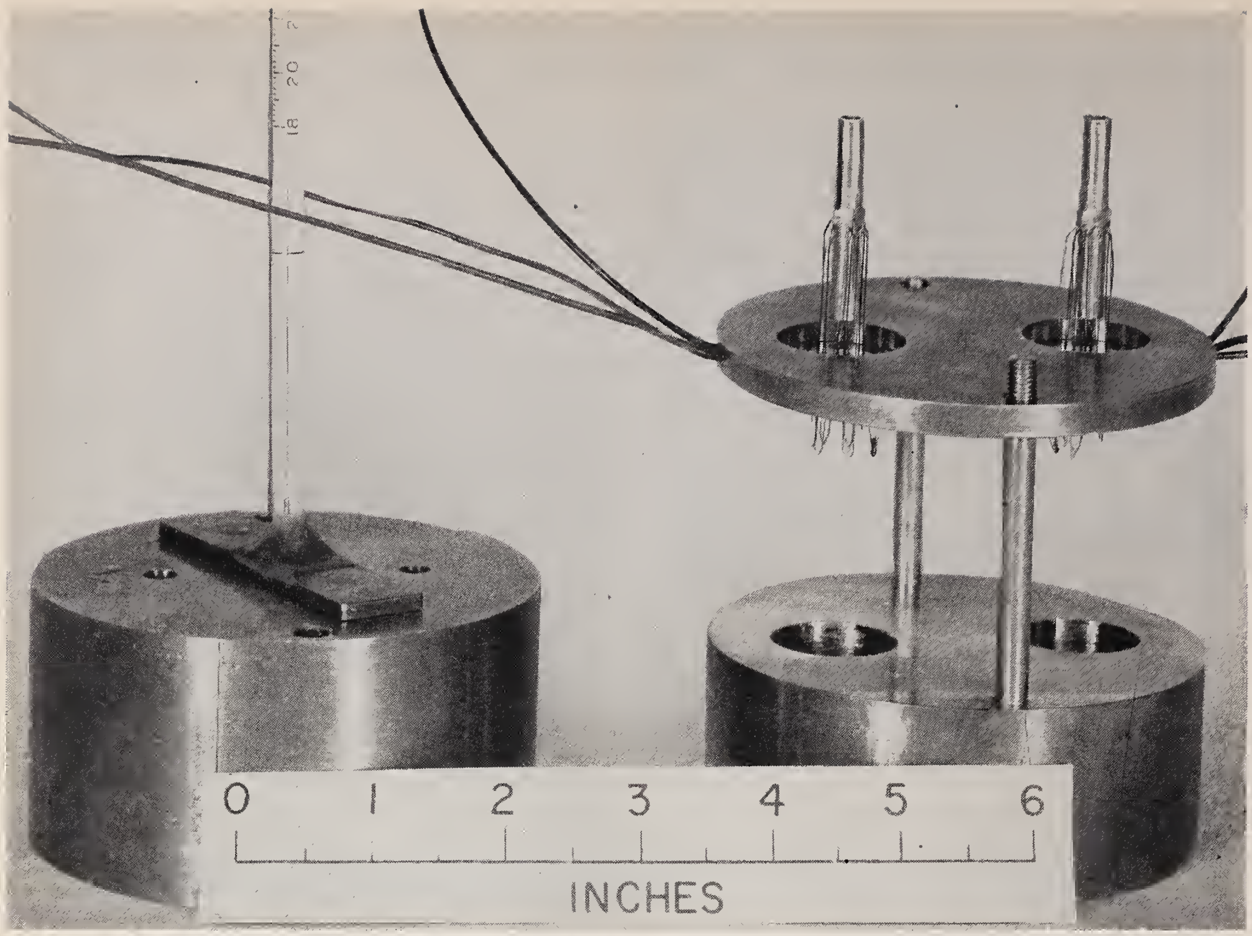


FIGURE 6. *The radiation balance or twin-cup Peltier-effect microcalorimeter that was constructed for the comparison of a number of national Hönigschmid radium standards (Mann, 1954b; Davenport, McCraven, Mann, and Smith, 1954; Connor and Youden, 1954; Loftus, Mann, Paolella, Stockmann, and Youden, 1957).*

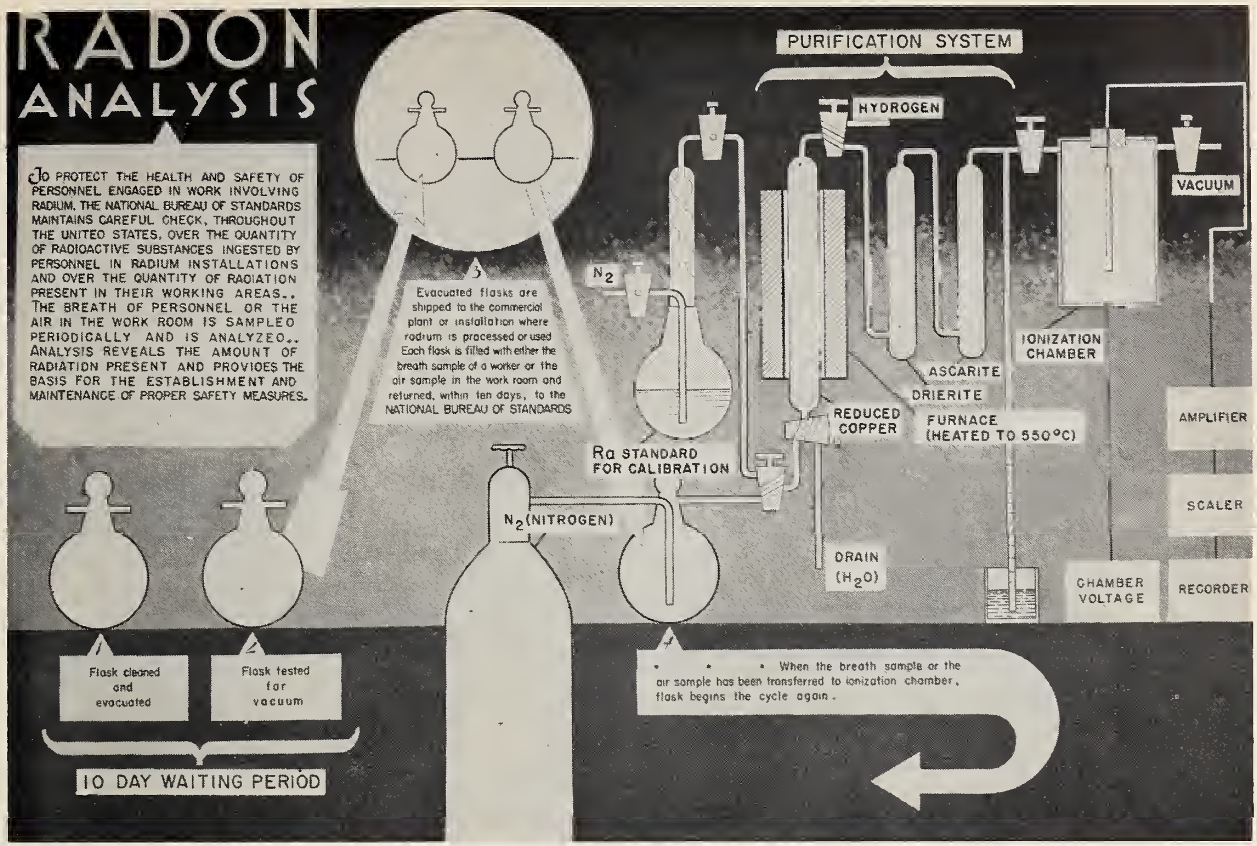
The cups of this radiation balance were of gold and it was enclosed in a temperature-attenuating enclosure to give a precision in the comparison of the national standards of about ±0.1 percent. The scale sensitivity, which limits the precision of the lowest rate of energy dissipation measurement, is about 3 mm/μw.

radium D+E+F or of radium F (polonium-210) can be determined by means of a $2\pi\alpha$-proportional-flow counter (fig. 10). Unless, however, the sources are thin, considerable straggling in the source will occur and give rise to plateaus that are not flat. Polonium-210 sources can also be standardized by determining their total gamma-ray emission, about 1 in every 10^5 alpha disintegrations giving rise to a less energetic alpha transition to an excited level of lead-206 with subsequent emission of a 0.804-Mev gamma ray. This alpha-branching ratio has recently been determined with a view to applying the value to the standardization of strong polonium-210 alpha sources (Grace, Allen, West, and Halban, 1951; Hayward, Hoppes, and Mann, 1955; Rojo, Hakeem, and Goodrich, 1955; Ascoli, Asdente, and Germagnoli, 1956).

Apart from radium and its products, there is little interest in the standardization of the naturally occurring radioactive elements with the possible exception of mesothorium or radiothorium as a generator of thorium X, an alpha emitter, for incorporation into a solution or ointment for the treatment of skin lesions. The demand for this is so inconsiderable, however, that the standardizations are normally carried out, as the need arises, by measuring the number of 2.62-Mev gamma rays from thorium C″, and calculating their build-up in relation to the thorium X. This standardization can be preserved in terms of an ampoule containing a solution of about 10 μg of radium chloride solution by means of an ionization chamber in the manner that is to be discussed later in connection with the maintenance of standards of the artificially produced nuclides.

2.2. Primary Standardization of Artificially Produced Beta-, Gamma-, and X-ray Emitters

When one turns to the problem of standardizing the artificially produced radioactive nuclides, the methods of measurement become as varied as the properties of the radiations that are emitted by such nuclides. There are low-energy and high-



TO PROTECT THE HEALTH AND SAFETY OF PERSONNEL ENGAGED IN WORK INVOLVING RADIUM, THE NATIONAL BUREAU OF STANDARDS MAINTAINS CAREFUL CHECK, THROUGHOUT THE UNITED STATES, OVER THE QUANTITY OF RADIOACTIVE SUBSTANCES INGESTED BY PERSONNEL IN RADIUM INSTALLATIONS AND OVER THE QUANTITY OF RADIATION PRESENT IN THEIR WORKING AREAS.. THE BREATH OF PERSONNEL OR THE AIR IN THE WORK ROOM IS SAMPLED PERIODICALLY AND IS ANALYZED.. ANALYSIS REVEALS THE AMOUNT OF RADIATION PRESENT AND PROVIDES THE BASIS FOR THE ESTABLISHMENT AND MAINTENANCE OF PROPER SAFETY MEASURES.

Evacuated flasks are shipped to the commercial plant or installation where radium is processed or used Each flask is filled with either the breath sample of a worker or the air sample in the work room and returned, within ten days, to the NATIONAL BUREAU OF STANDARDS

Flask cleaned and evacuated

Flask tested for vacuum

• • • When the breath sample or the air sample has been transferred to ionization chamber, flask begins the cycle again.

Figure 7. *Flow chart illustrating the method of radon analysis in use at the National Bureau of Standards (Curtiss and Davis, 1943).*

energy pure beta emitters, each calling for its own method of measurement, beta-gamma emitters and also electron capturers or X-ray emitters. Each particular case must be treated on its merits and may call for exhaustive prior investigations of the mode of disintegration of the nuclide. Thus a pure beta emitter may call for one kind of measurement, whereas a radioactive nuclide that decays by emitting an electron, followed immediately by the emission of a gamma ray from an excited state of the daughter nucleus, may be dealt with by a variety of methods, depending both on the degree of certainty with which the particular nuclear decay scheme is known and its complexity.

For beta emitters with maximum energy greater than about 300 kev, the best, and most widely used, method of primary standardization is that of $4\pi\beta$ counting, either in the proportional or Geiger regions. This method involves the use of a thin source mounted on as thin a conducting film as possible and looked at on both sides by two hemispherical, semicylindrical or "pill-box" counters each subtending a solid angle of essentially 2π at the source. The use of this method was suggested first in 1944, but the counter then described (Simpson) appears to have had somewhat appreciably less than a completely 4π geometry. The first such counter to attain to what is now regarded as essentially a 4π geometry was designed by Schutzmeister-Meyer and described in 1948 (Haxel and Houtermans). The use of such counters has since been carried to a high standard of working efficiency by a number of investigators (Putman, 1949; Cohen, 1949 and 1952; Sawyer and Wiedenbeck, 1950; Borkowski, 1950; Charpak and Suzor, 1950; Seliger and Cavallo, 1951; Suzor and Charpak, 1951, 1952a, and 1952b; Hawkings, Merritt, and Craven, 1952; Houtermans, Meyer-Schutzmeister, and Vincent, 1952; Meyer-Schutzmeister and Vincent, 1952; Gray, 1952; Mann and Seliger, 1953; Smith, 1953 and 1954; Smith and Wildblood, 1953; Pate and Yaffe, 1955a, 1955b, 1955c, 1955d, and 1956) (figs. 11, 12, and 13).

To avoid source self-absorption of electrons at the low-energy end of the beta spectrum, it is essential to reduce solids in the source to a minimum. For this reason, it is often important to precipitate the source material chemically from the drop of solution that has been deposited on the thin-film source holder, or to "freeze-dry" it in position. In the process of simple evaporation, the slow reduction in physical size of the drop of solution results in an ever-increasing concentration of solute in the residual drop, and a final deposition of solids from the vanishing drop. The structure of precipitated low-solids sources has

Figure 8. *A rack of six ionization chambers with associated linear amplifiers and scalers.*

The outputs of the scalers, giving the numbers of alpha particles counted, are recorded on traffic recorders hourly throughout the night. The transfer of the samples from the test flasks (shown on the bottom shelf) to the ionization chambers is effected during the day. Oxygen is removed, to reduce recombination effects, by passing the gas samples over heated copper in electric furnaces, which can be seen in the top left-hand corner.

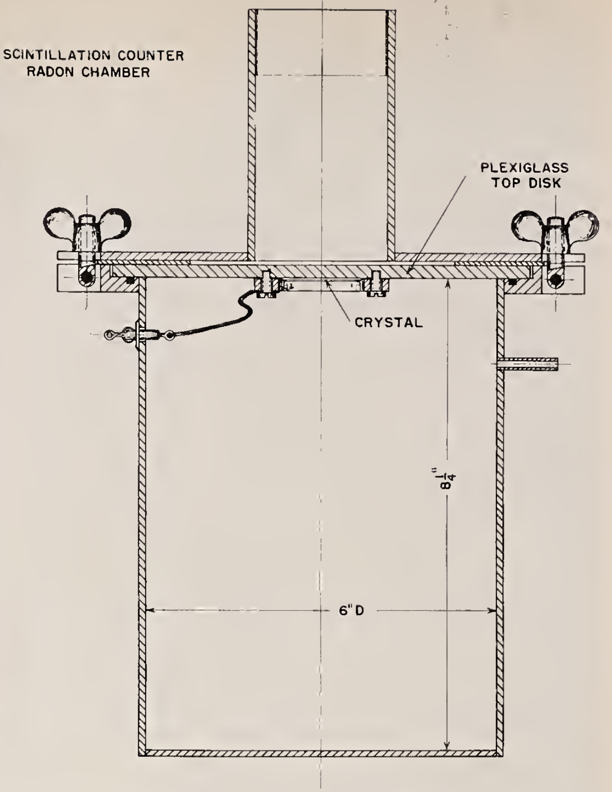


Figure 9. *Cross-sectional drawing of scintillation counter for radon analysis constructed at the National Bureau of Standards and based on the method of Bryant and Michaelis (1952).*

The multiplier phototube is located in the upper tube and looks at the scintillator consisting of a thin layer of zinc-sulfide crystals. An aluminum foil close to and parallel to the zinc-sulfide layer is maintained at a negative potential of some 2,000 v. with respect to the surroundings. On admitting a radon sample into the chamber, radium-A recoils are attracted electrostatically to the aluminum foil and the subsequent alpha particles striking the zinc-sulfide screen are counted. The equipment is calibrated by admitting a sample of radon from a radium solution standard.

been investigated by the method of electron-shadow-micrography in the case of silver-iodide (iodine-131) sources (Seliger and Schwebel, 1954), and it has been found that the deposition of such sources occurs in almost spherically shaped "islands" surrounded by large vacant areas (fig. 14). The addition of solids therefore merely serves to increase the number of "islands" without essentially changing their size and, thus, to fill in the empty spaces without increasing the source self-absorption in any manner that can be subjected to analysis by the method of extrapolation. The counting rate on addition of inactive solids can, in fact, up to a certain point remain unchanged. It has been shown that in the case of cobalt-60, which has a maximum beta energy of 310 kev, the source self-absorption can be as high as 10 percent in $4\pi\beta$-proportional counting (Seliger and Schwebel, 1954). The method has, on the other hand, been used for sulfur-35 (Pate and Yaffe, 1955c; Seliger, Mann, and Cavallo, 1958), sources of which can be prepared with practically negligible solids, even though the maximum beta energy is considerably lower than that of cobalt-60.

Measurements of the charge carried away from a source of radium have been made by several investigators (Wien, 1903, Rutherford, 1905, Makower, 1909; Moseley, 1912). Recently, however, this method has been applied to the standardization of beta emitters (Clarke, 1950; Gross and Failla, 1950; Keene, 1950). In 1950, experiments were described using both spherical (Clarke) and parallel-plate (Gross and Failla) geometry. The former achieved agreement with National Bureau of Standards standard samples of phosphorus-32 to within 5 percent, but has not since been in general use. In the parallel-plate geometry the source is a very thin deposit of the emitter in question on one plate of a parallel-plate collecting system, the second plate being sufficiently close to balance the secondary electron emission and backscattering effects. Low-energy secondary electrons are bent back by means of a weak magnetic field, and the measurement is made in a vacuum to eliminate air ionization. General agreement with the National Bureau of Standards standard samples of phosphorus-32 to within ±2 percent has been obtained by this method over a number of years (Seliger and Schwebel, 1954). In another development of the charge-measuring

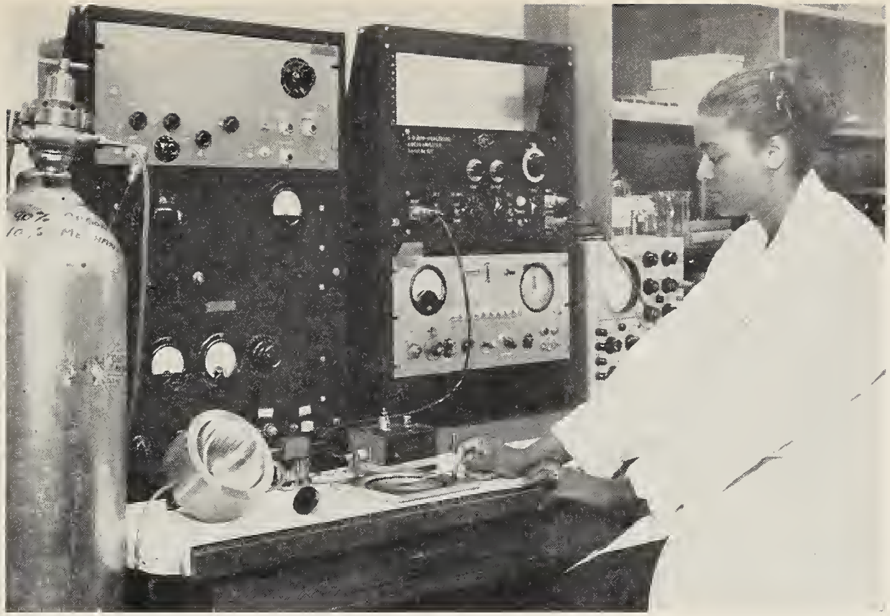


FIGURE 10. *The $2\pi\alpha$-proportional counter used for the measurement of alpha-particle standards.*

The diameter of the steel hemisphere of the counter is 11.9 cm and the counter has been found, by surveying with an approximately point alpha-particle source, to give a flat response over an area in the center, which is considerably greater than that of the alpha-particle sources to be measured. Hemispheres of smaller diameter had previously been found to give too rapid a fall-off in response with distance from the center.

method (Keene, 1950), the source is deposited on a thin aluminum foil that is completely surrounded by an open-mesh wire-grid enclosure. This experiment, like the first, is carried out in a vacuum, and the current from the foil to the grid is measured for different positive and negative voltages between them. Agreement to within about 10 percent for the calibration of a phosphorus-32 source from the Atomic Energy Research Establishment at Harwell is quoted.

For low-energy beta emitters there are two general methods of primary standardization available. There are the methods of gas counting and of calorimetry. In the former the radioactive nuclide such as hydrogen-3, carbon-14, or sulfur-35 is admitted in the form of a gas into a cylindrical-cathode gas counter operating in either the proportional or Geiger regions. With the investigators who have used this method there are two general philosophies, namely, that which advocates the use of long counters having negligible or calculable end corrections due to the fall-off in electric field or change in active volume (Miller, 1947; Brown and Miller, 1947; Anderson, Libby, Weinhouse, Reid, Kirshenbaum, and Grosse, 1947; Engelkemeir, Hamill, Inghram, and Libby, 1949; Jones, 1949; Bernstein and Ballentine, 1950; Engelkemeir and Libby, 1950), and that which promotes the simultaneous use of two or more counters identical in every respect except for the cathode and anode wire lengths (Mann and Parkinson, 1949; Hawkings, Hunter, Mann, and Stevens, 1949; Hawkings, Hunter, and Mann, 1949; Manov and Curtiss, 1951; Mann, 1958). By taking the difference in counting rates for such a compensated set of counters, the counting rates are obtained for ideal cylinders, equal in length to the differences in the lengths of the individual counters (figs. 15, 16, 17, and 18). Investigators of the former predilection have investigated the effect of varying length, and most of the workers in both disciplines have experimented with counters of varying radii, with a view to determining the magnitude of the wall correction arising from the fact that beta particles proceeding towards the wall from a finite layer of gas contiguous to the wall may not be counted. The thickness of this layer will depend on the number of ion pairs formed per unit length of beta path, and upon the minimum number of ion pairs required to give a count. As the number of ion pairs formed per unit path-length will be proportional to gas pressure, this may also be varied in an investigation of the wall effect.

It is of importance in gas counting to be assured that any chemical reactions used in the preparation of the counting gas from the radioactive sample shall have been carried to completion, so that the measured radioactivity of the gas may be quantitatively related to the radioactivity of the original sample independent of any change in isotopic abundance that might occur in an incomplete reaction. Alternatively, it may be more satisfactory to convert the counting gas itself quantitatively back to compounds that can be totally absorbed or trapped after the counting experiment has been completed. Thus, in the standardization of hydrogen-3 or carbon-14, the quantitative preparation of, say, radioactive acetylene, in which the hydrogen-3 or carbon-14

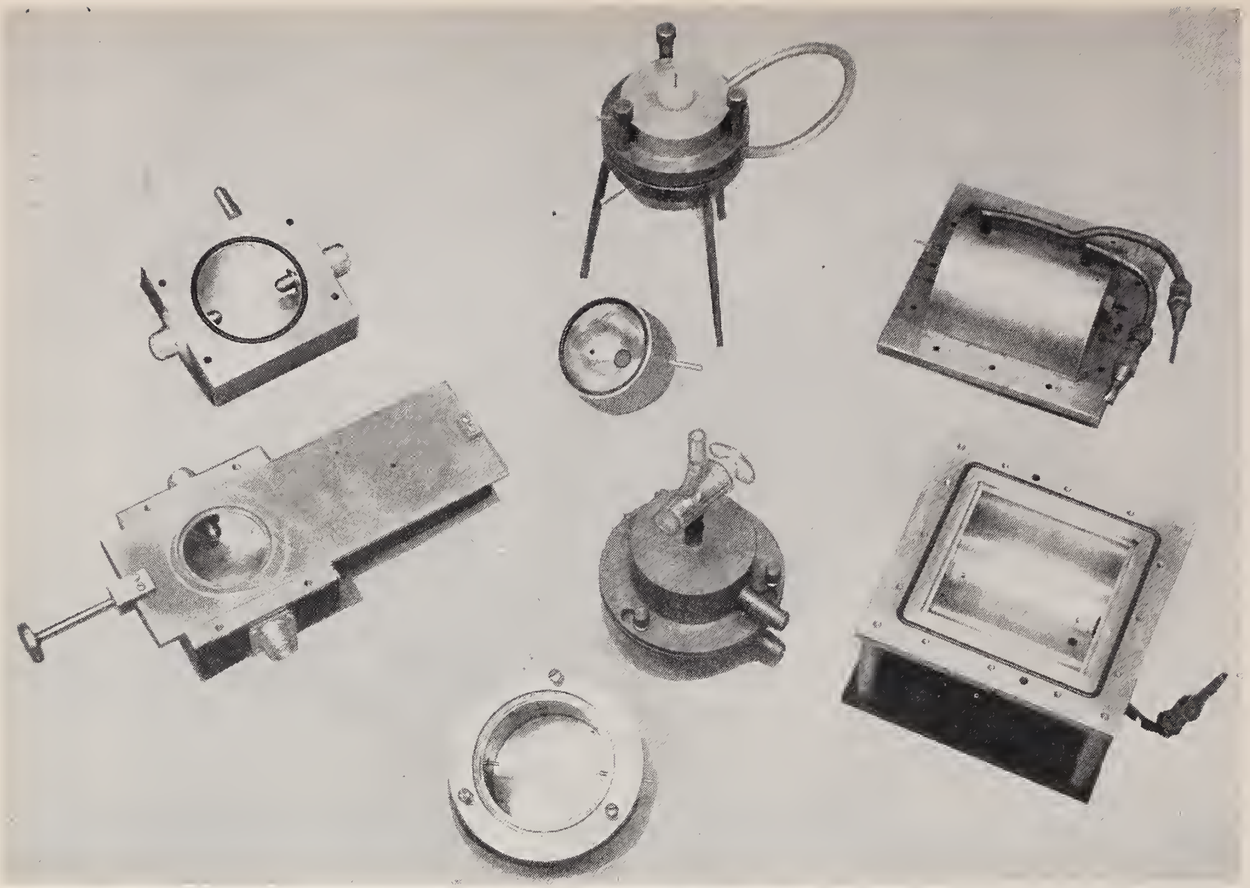


FIGURE 11. *A selection of the $4\pi\beta$-proportional counters that have been used at the National Bureau of Standards for the calibration of beta-ray standards (Seliger and Cavallo, 1951; Mann and Seliger, 1953; Seliger and Schwebel, 1954).*

The parts of the type currently in use are shown in the center of the photograph, and in figures 12 and 13.

is incorporated in the acetylene molecule, may be difficult. Any sample of radioactive acetylene that has not been quantitatively prepared can, however, be quantitatively converted by complete combustion, after counting, to water and carbon dioxide. The measured radioactivity of this water or carbon dioxide can then be compared with the original sample by relative counting techniques, such as the liquid scintillation counter or the gas ionization chamber. The only slight drawback to this method would lie in the fact that the amount of the primary standard available would be somewhat limited.

Electron-capturing nuclides that exist in suitable gaseous compounds should be susceptible of primary standardization by gas-counting methods, provided that adequate information regarding the fluorescence yield is available.

In some cases where macroscopic amounts of the disintegrating nuclei can be obtained, the rate of disintegration can be determined by collecting and measuring the amounts of the decay products. Such techniques, that have in the past been used to determine the ages of rocks containing the naturally occurring radioactive elements, have recently been most successfully applied (Jones, 1955) to obtaining the half-life of tritium by determining the initial isotopic abundance and the number of disintegrations in a given time, the latter being measured by collecting the helium-3 formed and assaying it mass-spectrometrically. This method provides an absolute measurement of the disintegration rate, but it has application in only a limited number of specific cases.

Microcalorimetric methods have been used at the Chalk River laboratories of Atomic Energy of Canada Limited to measure the disintegration rate of hydrogen-3 (Hawkings and Merritt, 1954), and at the National Bureau of Standards to determine the disintegration rate of both hydrogen-3 and sulfur-35 (Gross, Wingate, and Failla, 1956 and 1957; Bay, Mann, Seliger, and Wyckoff, 1957). As there is no published account of the hydrogen-3 standardization carried out at the National Bureau of Standards, it may be appropriate to mention that the rate of energy emission from about 4.8 curies of tritiated water contained in a small quartz tube was measured over a period of some 4 months in the radiation balance developed to compare the 1934 Hönigschmid radium standards (Mann, 1954b). The tritiated water had been sealed into the quartz tube under some 2-cm-Hg pressure of

FIGURE 12. *A closeup view of the $4\pi\beta$-proportional counter in use at the National Bureau of Standards showing details of the source mounted on plastic supported by aluminum foil.*

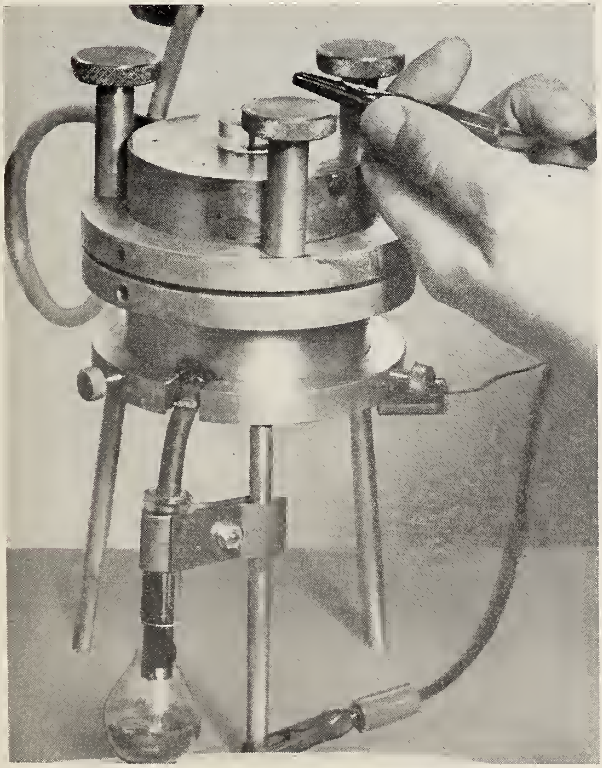


FIGURE 13. *The National Bureau of Standards $4\pi\beta$-proportional counter assembled for use.*

FIGURE 14. *Chromium-shadowed electron micrograph of a silver-iodide source deposited on a plastic film.*

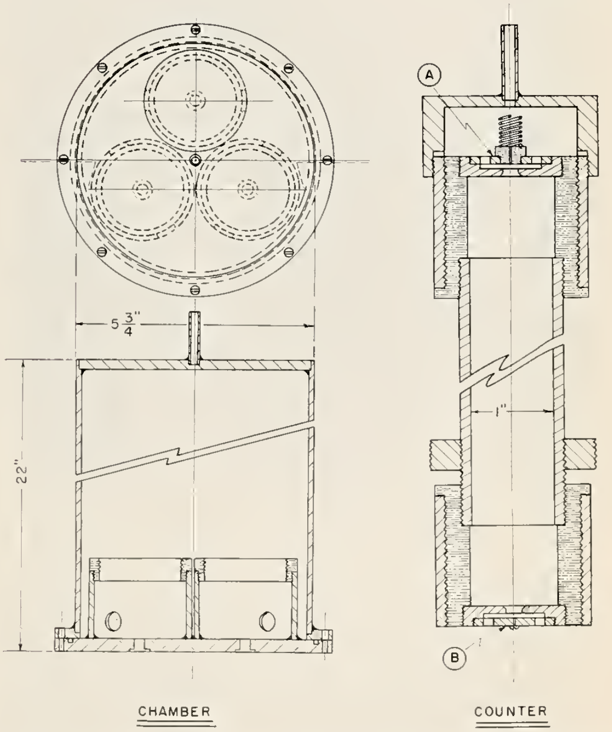


FIGURE 15. *Workshop drawing of a set of compensated gas counters that have been constructed with copper anodes with Teflon-insulated stainless-steel anode-wire supports at each end (Mann, 1958).*

The tolerances of the end construction have been kept to within a few thousandths of an inch so as to achieve accurate compensation for end effects. The internal diameter of the copper anodes is 1 in. The anode lengths are 6.0012, 8.0017, and 16.0003 in.

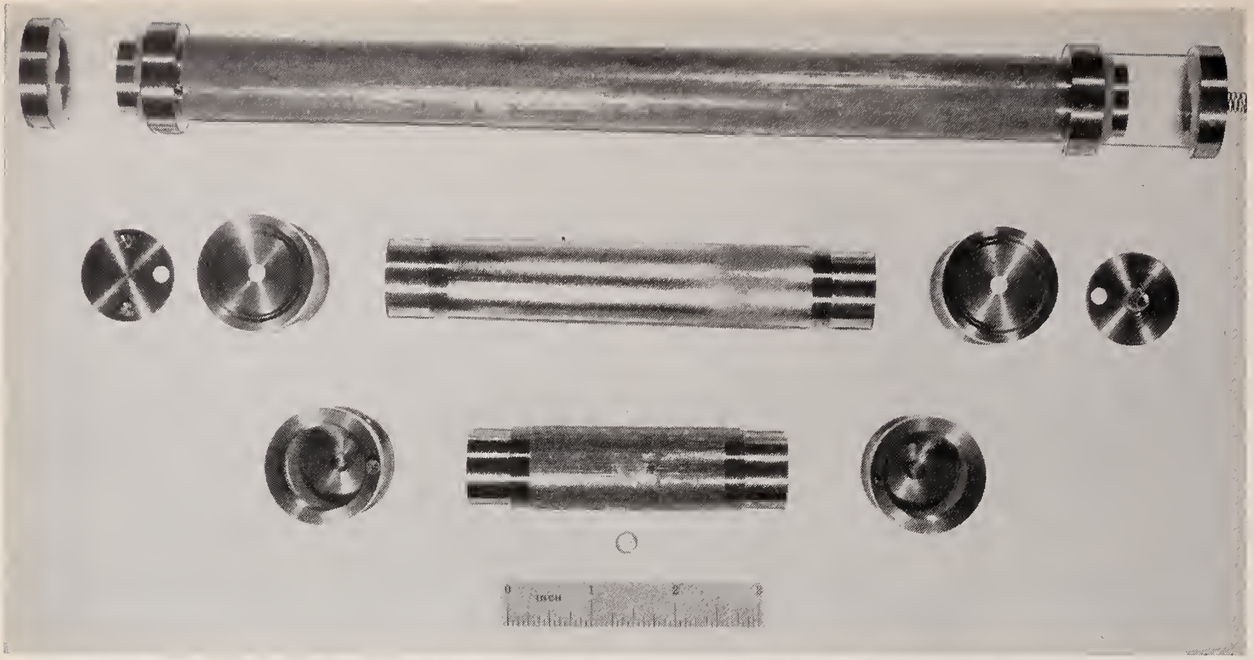


FIGURE 16. *A set of three stainless-steel compensated counters with glass and stainless-steel ends (Mann, 1958)*

The glass insulators are cemented into stainless-steel rings which are in turn screwed on to the stainless-steel cathodes by means of carefully cut micrometer threads. The ends were mounted onto the cathodes using a traveling microscope to insure that the distance from the end of each cathode to the stainless-steel disk supporting the tungsten anode wire was the same in all of the counters. The anode lengths of the stainless-steel counters are 3.7586, 5.7410, and 11.7623 in.

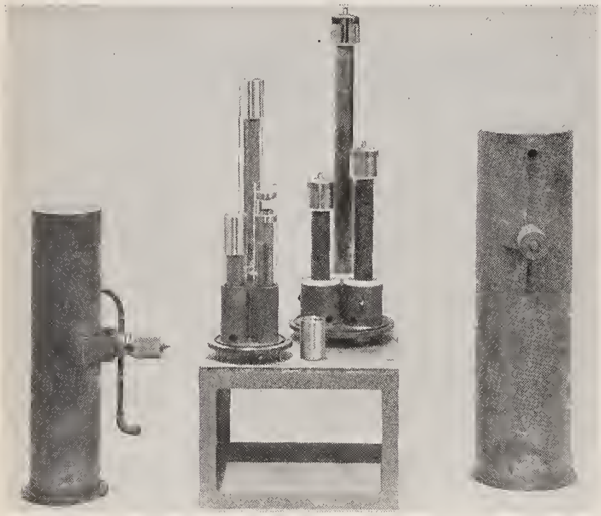


FIGURE 17. *The compensated gas counters described in figures 15 and 16 assembled ready for enclosure in their respective vacuum containers.*

The stainless-steel counters are on the left and the copper on the right.

hydrogen to inhibit the formation of H and OH radicals as a result of irradiation of the water (Ghormley and Allen, 1948). After measurement in the microcalorimeter, this tube of tritiated water was crushed under 1 kg of water in a specially constructed stainless steel container, and the master solution so obtained diluted to give standards of activity around 10^4 and 10^6 dps/ml. The exact disintegration rate was obtained from the energy emission rate per milliliter and the assumed mean energy of 5.67 kev per disintegration for hydrogen-3 [2] (Jenks, Ghormley, and Sweeton, 1949).

For beta emitters that are associated with one or more gamma rays in coincidence, the method of respectively beta-gamma or gamma-gamma coincidence counting is one of the most powerful standardizing tools available. The earliest known application of this method was by H. Geiger and A. Werner (1924), to determine the efficiencies of two observers looking at the same zinc-sulfide screen through two microscopes and counting the number of scintillations due to alpha particles. The application of the method of coincidence counting to the determination of the efficiencies of beta or gamma counters (Dunworth, 1940) has the added advantage that these efficiencies normally remain constant and do not suffer from such human frailties as fatigue and susceptibility to diversion. The method of coincidence counting has been used in conjunction with $4\pi\beta$ counters (Smith, 1953 and 1954) in the standardization of cobalt-60. In the standardization of cobalt-60 by the method of gamma-gamma coincidence counting, using sodium-iodide (thallium-activated) crystals as the detectors in each channel, the effects of backscattered gamma rays from one counter giving rise to a coincidence count in the other is readily eliminated by setting the discrimination levels to exclude all such backscattered gamma rays. By introducing varying resolving times by the use of delay lines of varying length, it is also possible to obtain a linear plot of coincidence counting rate against time (or delay-line length) and to

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[2] The mean energy for disintegration is the mean beta-ray energy less the difference in the total electron binding energies of hydrogen-3 and helium-3.

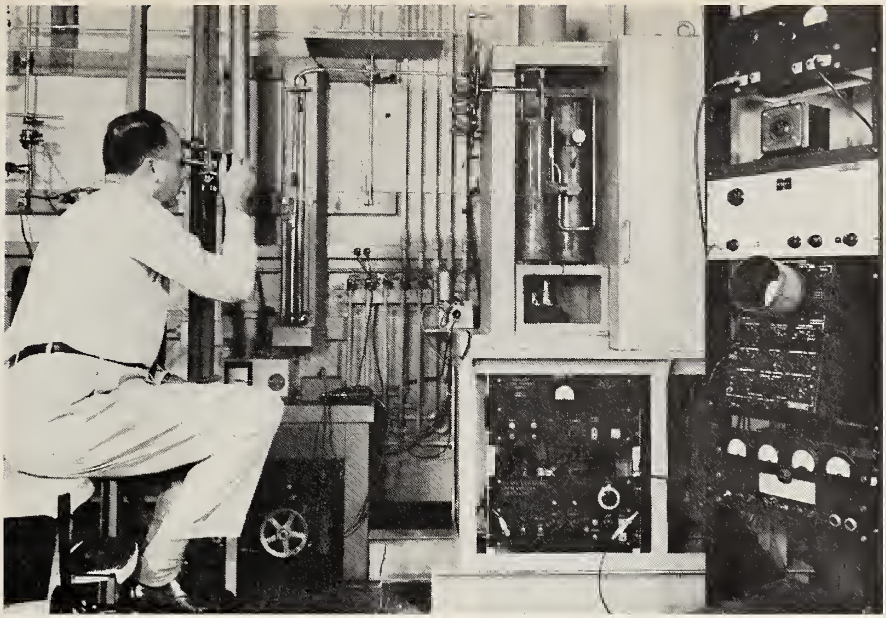

FIGURE 18. *The National Bureau of Standards compensated gas counters for the counting of low-energy beta emitters and electron-capturing nuclides assembled within a steel shield and with their auxiliary gas-handling equipment.*

extrapolate to zero resolving time (or zero length) to obtain the true coincidence rate without accidentals (Hayward, 1953). In conjunction with fast circuitry, this method is of great use in the standardization of such nuclides as cobalt-60 and sodium-24 (fig. 19).

In the application of beta-gamma coincidence scintillation counting it is important to minimize the efficiency of the beta detector to gamma rays. This can readily be done by grinding the crystal detector used in the beta channel down to a thickness of about 0.5 mm (Seliger and Schwebel, 1954) (fig. 20), although there is always a finite effect due to Compton electrons produced in the glass-faced photocathode of the electron-multiplier phototube. While it had been tacitly assumed that the source, in the coincidence technique, was a point source it was later shown that the method is equally applicable to extended sources provided that the efficiency of either detector is the same for all parts of the source (Putman, 1950). Such would be the case using a $4\pi\beta$ counter as the detector in the beta channel (Campion, 1958).

Electron-capturers that emit a nuclear gamma ray in coincidence with the K and L X-rays can also be standardized by the method of X-ray—gamma-ray coincidence counting.

The principle of $4\pi\beta$ counting has recently been extended to crystal scintillation counting by forming a sandwich of two anthracene crystals with the source deposited between the sandwich on one of the crystals (Smith, Seliger, and Steyn, 1956; Mann and Seliger, 1956) (fig. 21). Two phototubes look at the sandwich from opposite sides, and true counts in the crystal are recorded as coincidences between the two phototube channels. Efficiencies of better than 99 percent have been obtained by this method for phosphorous-32, strontium-90–yttrium-90, and thallium-204. The low threshold of about 4 or 5 kev of this method enables one to hope that it will be possible to use it to standardize electron-capturing nuclides.

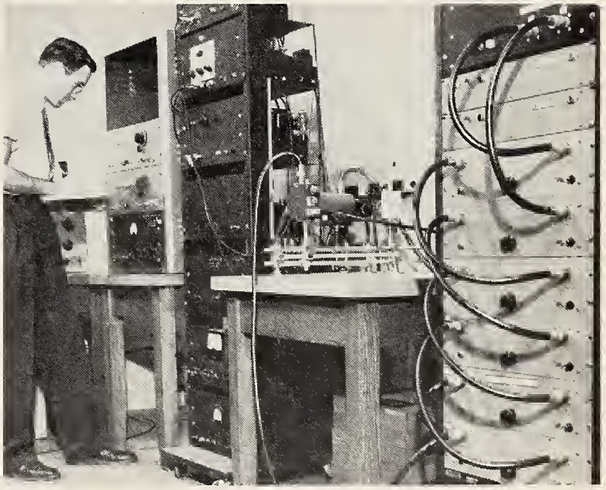

FIGURE 19. *Equipment for the determination of the disintegration rate of a source of cobalt-60 by the gamma-gamma coincidence method (Hayward, 1953).*

The angular correlation between the directions of emission of the two gamma rays is used to obtain the disintegration rate under different experimental conditions such that the presence of many spurious effects may be detected easily.

The method of $4\pi\beta$-liquid-scintillation counting has been applied to the standardization of alpha and beta emitters (Belcher 1953; Basson and Steyn, 1954; Steyn 1956, Seliger 1958a, 1958b, and 1958c). Belcher (1953), using beta emitters, supported the source in the liquid scintillator between a sandwich of two thin foils. The absorption

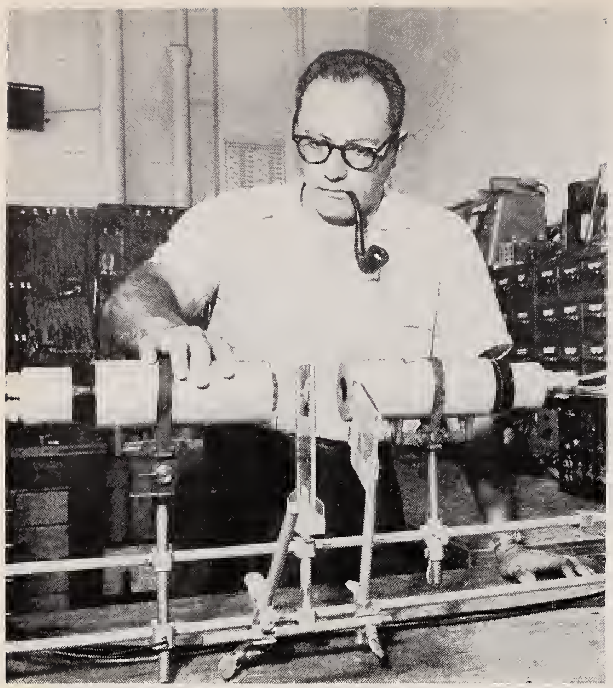

FIGURE 20. *Photograph of the low-geometry β-γ scintillation coincidence-counting arrangement at the National Bureau of Standards.*

The source is mounted on a thin plastic film between the beta detector on the right and the gamma detector on the left. The aluminum absorbers that are used to estimate the gamma-counting rate in the beta detector can be rotated to be directly in front of the beta detector which is an anthracene crystal ground down to a thickness of approximately 0.5 mm. The gamma detector is a sodium-iodide (thallium-activated) crystal. Coincidence resolving times of the order of 0.5 μsec are usually used. In actual practice, the β, γ, and β-γ channels are always counted over the same interval. In this way the disintegration rate obtained is independent of any changes in efficiency of the β or γ detector due to slight drifts in the electronic circuits.

correction for these foils and for source self-absorption can, however, be quite high. Basson and Steyn (1954) and Seliger (1958c) have standarized alpha particle solutions of polonium-210 by incorporating the solution directly into the liquid scintillator, and Steyn (1956) and Seliger (1958a and 1958b) have pushed the technique to the standardization of beta-emitting nuclides incorporated as a solution in the liquid scintillator.

2.3. Secondary Standardization of Artificially Produced Beta-, Gamma-, and X-ray Emitters

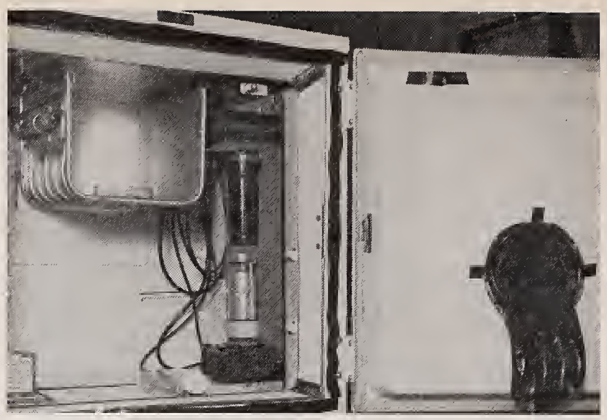

FIGURE 21. *Photograph of the 4π-crystal-sandwich-coincidence-counting arrangement (Smith, Seliger, and Steyn, 1956; Mann and Seliger, 1956).*

The source is delivered directly to the polished face of one anthracene crystal. The second crystal is placed directly on top and the sandwich surrounded by an aluminum-foil cylinder. The flat phototube faces make contact with the crystal faces during the measurement. The refrigerator also serves as a light-tight enclosure.

The simplest way of effecting a comparison between a primary and secondary standard of radioactivity is by means of a substitution method under reproducible conditions of geometry, the relative intensities of radioactivity being measured by any instrument that records either individual beta or gamma rays or else measures their ionizing effects. Such a method, whatever the nature of the recording device, can, in the hands of a competent observer, be relied upon to give results having a standard deviation of from 0.1 to 1.0 percent. It is also to be understood that within the meaning of "reproducible conditions of geometry" is to be included the method of source preparation. The method of comparing radioactive sources of unknown strength with calibrated radium D+E sources is no longer very acceptable, on account of the fact that the geometrical conditions are, on the whole, somewhat irreproducible and involve considerations of absorption and backscattering. The method has, however, been shown to give consistent results with the use of "weightless" sources mounted on thin films (Novey, 1950). Radium D+E sources are now, in general, only recommended for use as reference sources for the maintenance of the calibration of measuring equipment by the method of substitution in which the initial calibration has been made using a standard sample of the nuclide in question.

The recording device used in such a substitution method can be a gold-leaf or quartz-fibre electroscope; a beta ionization chamber or gamma ionization chamber used in conjunction with a Lindemann-Ryerson, or vibrating-reed electrometer, or any other low current-measuring device; any kind of beta-, gamma-, or X-ray counter; or a formamide solution in a $2\pi\beta$-proportional counter.

At the National Bureau of Standards the primary standardizations of the short-lived nuclides are maintained by means of a $2\pi\beta$-ionization chamber in conjunction with radium D+E reference sources, and a $4\pi\gamma$-ionization chamber in conjunction with reference standards of radium consisting of radium bromide solution in flame-sealed glass ampoules (Seliger and Schwebel, 1954). More recently both types have been replaced by radium reference sources, with the radium salt sealed in stainless steel. Primary standards of the short-lived nuclides sodium-24, phosphorus-32, potassium-42, iodine-131, and gold-198 are calibrated by $4\pi\beta$ or coincidence counting. These primary standards are then used to effect a calibration of the appropriate ionization chamber in

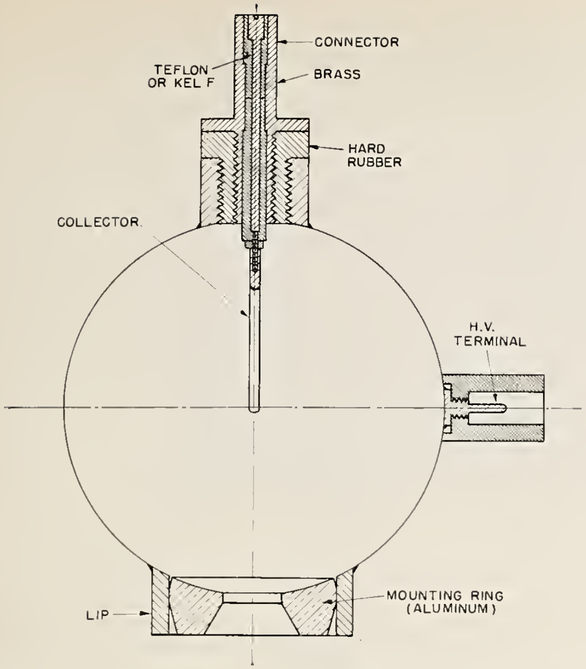

FIGURE 22. *Schematic drawing showing the construction details of the NBS 2πβ-ionization chamber (Seliger and Schwebel, 1954).*

The chamber proper is made from a flagpole ball graciously donated by Dr. S. Feitelberg of Mt. Sinai Hospital, New York, N. Y. The body is a gold-plated sphere approximately 6 in. in diameter. At the base is a cylindrical shoulder which slides smoothly over the source-mounting ring. The center of the source-mounting ring is recessed to hold reproducibly a 1-in. diameter disk on which an aliquot of the active solution is evaporated. The length of the ⅛-in. diameter brass-collecting electrode has been adjusted empirically so that a point source of beta rays can be displaced slightly from the center of the source mount without affecting the observed ionization current. This ensures that no error will be introduced into the secondary standardization due to off-center drying of solutions delivered to the disks. Current measurements are made with a Lindemann-Ryerson electrometer in a Townsend rate-of-charge null method. The chamber is mounted together with the Lindemann-Ryerson electrometer and a compensating condenser in such a manner that the entire assembly can be raised several inches above the aluminum mounting ring, into which the sources are placed for measurement. Collecting voltages are applied to the chamber so that the collector is always very close to ground potential during a reading.

conjunction with a radium reference source. Further standards of these artificially produced nuclides are then issued as secondary standards by calibration against the reference sources in these chambers (figs. 22, 23, and 24).

A cavity ionization chamber has also been used to determine, to within ±5 percent, the disintegration rate of a beta or gamma emitter (Gray, 1949), the former being incorporated in a gelatin lining to the chamber and the latter being in the form of an external source. The gamma rays from radium in equilibrium with its products were used to calibrate the chamber. The relative stopping powers of the wall and air, and the energy loss per ion pair formed in air for the gamma rays of the radium in equilibrium with its daughters and for the beta or gamma rays of the nuclide in question, can then be used to calculate its rate of disintegration (Gray, 1949; Robinson, 1954; Perry, 1954a). Any gamma emitter other than radium may be used to calibrate the ionization chamber, provided that the dose rate in air to which it gives rise is known in terms of its disintegration rate.

In this connection it is interesting to note that in addition to using counting methods to standardize such gamma emitters as sodium-24, potassium-42, iron-59, cobalt-60, and bromine-82, some laboratories in the United Kingdom have also maintained standards of such nuclides by determining the roentgens per hour at 1 cm from samples standardized by counting methods. This was partially to accommodate medical and therapeutic needs. As a result, the British standard of radioactive iodine-131 is a standard that has been defined by measurement of its gamma-ray effect, in terms of roentgens per hour at 1 cm, using a graphite-wall ionization chamber that had been calibrated with radium (Bullard, 1953), assuming a conversion factor of 8.3 roentgens per hour per milligram at 1 cm. However, in order to derive the millicurie content of the iodine-131, it is necessary to adopt an arbitrary conversion factor to correlate the response of the chamber with absolute counting measurements. This factor is 2.25 roentgens per hour in air at 1 cm from 1 mc of iodine-131, and compares favorably with the factor 2.29 deduced from recent determinations of the disintegration scheme (Cavanagh, 1952; Bell and Graham, 1952a and 1952b) if the average energy lost in producing one ion pair in air is assumed to be 32.5 ev and the conversion factor for radium is assumed as above. However, recent measurements (Jesse and Sadaukis, 1957; Gross, Wingate, and Failla, 1957; Bay, Mann, Seliger, and Wyckoff, 1957) have shown the energy per ion pair in air to be around 33.7 ev. The use of this value would reduce the factor of 2.29 to 2.21 but leave the agreement with experiment unchanged. None of these assumptions affects the value of the standard in millicuries, which is ultimately based on counting methods, once the arbitrary conversion factor has been adopted. It would therefore seem that this method, while preserving the primary standard, is capable only of producing secondary standards for future use. The dose-rate as measured in air will be unchanged by changes in other atomic and nuclear data, and is therefore the same for the clinician, but is relative to the corresponding figure for radium, so long as this is used to calibrate the graphite-walled chamber.

For low-energy beta emitters, such as hydrogen-3 or carbon-14, comparisons with the primary standards can be carried out by liquid scintillation counting (Raben and Bloembergen, 1951; Audric and Long, 1952 and 1953; Ziegler, Seliger, and Jaffe, 1956; Steyn, 1956; Seliger and Ziegler, 1957) (fig. 25). The method of formamide counting (Schwebel, Isbell, and Karabinos, 1951; Schwebel, Isbell, and Moyer, 1954) is particularly useful for the rapid comparison of the activities of aqueous solutions of salts of the radioactive nuclides. In this method up to 10 percent of an aqueous solution of radioactive material can be dissolved in the organic solvent, formamide, and this resulting solution can be introduced into a $2\pi\beta$-proportional flow counter without in any way impairing the electrical or counting properties of the counter. A great advantage of this method is that it completely eliminates problems arising from differences in source self-absorption.

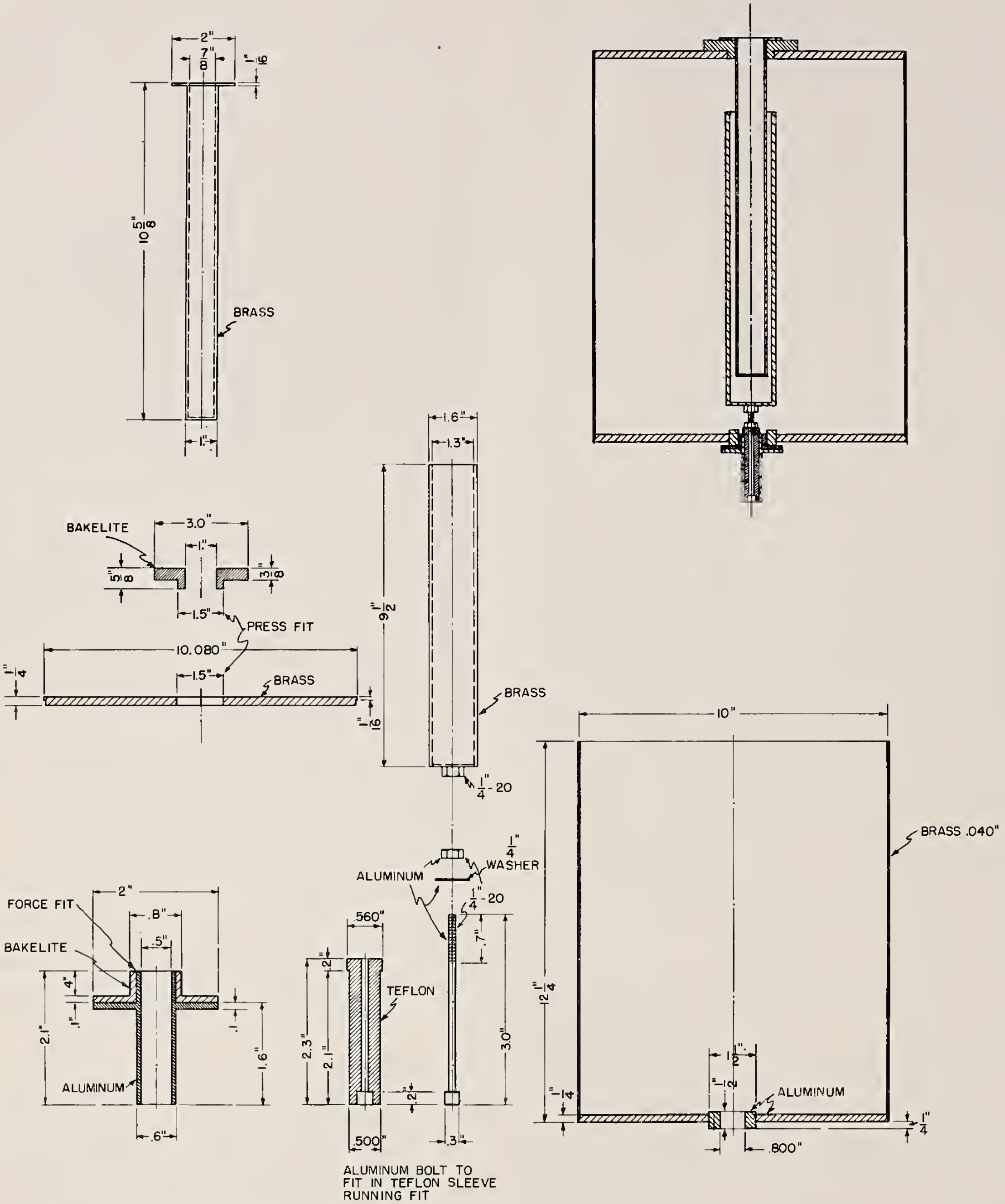

Figure 23. *Schematic drawing showing the construction details of the NBS $4\pi\gamma$-ionization chamber* (*Smith and Seliger, 1953*).

The chamber is constructed of brass and aluminum and has a volume of approximately 15 liters. The reentrant cylinder is separated from the main body of the chamber and is grounded. Sources are inserted into the cylinder and supported at the geometrical center on a Lucite spacer. If a point source of gamma radiation is moved vertically through ±4 cm about the geometrical center of the chamber, the response remains uniform to ±1 percent. Thus it is possible to standardize solutions of different volumes provided self-absorption is negligible. High voltage is applied to the chamber so that the collector cylinder is always very close to ground potential during a reading. Current measurements are made with a Lindemann-Ryerson electrometer in a Townsend rate-of-charge null method.

FIGURE 24. *Photograph of the NBS $2\pi\beta$- and $4\pi\gamma$-ionization chambers (Garfinkel, 1958).*

The $4\pi\gamma$ chamber is mounted inside a 1-in. thick lead shield to reduce the background which is less than 4×10^{-14} ampere. A source is being inserted into the $4\pi\gamma$-ionization chamber. On the left are the manual controls for applying compensating charge and for measuring the timing interval. On the right is a semiautomatic timing and charge-compensating circuit. Diode photocells, actuated by the projected shadow of the electroscope fiber, switch on and off a decade scaler and a motor-driven 10-turn precision potentiometer, respectively. The decade scaler is driven by the NBS standard 1,000-cycle frequency and the compensating voltage is read by means of the precision potentiometer.

3. Problems Arising in the Preparation and Distribution of Radioactivity Standards

In some countries where the use of radioactive materials is still relatively on a small scale, facilities exist for the standardizing of each consignment *en route* to the laboratory to which it is being consigned. In the United States and the United Kingdom this practice is no longer feasible, so that both the National Bureau of Standards and the National Physical Laboratory produce radioactivity standards for distribution to laboratories using radioactive materials for experimental purposes in order that these laboratories may standardize their own calibration equipment. The standardization of such equipment for any particular radioactive nuclide, having radiations unique to itself and therefore giving rise to a definite efficiency for measurement, is then preserved in terms of a long-lived radioactive substance such as radium. Incoming shipments of radioactive materials can then be routinely calibrated and, provided that there is no change in the characteristics of the calibrating equipment, only an occasional recheck is necessary against a radioactivity standard of the nuclide under consideration. From the point of view of permanence and consistency of operation, ionization methods can be seen to be superior to counting methods that depend on both the life of the counter, if it is of the sealed variety, and the maintenance of the auxiliary electronic equipment.

Most of the radioactivity standards of the National Bureau of Standards, as can be seen from the list of table 1, are solution standards to which sufficient carrier has been added to render negligible the adsorption of the active material to the container walls. With five exceptions, these are standards of relatively long life that are prepared in batches of several hundred ampoules at a time and kept in stock for distribution as required (figs. 26, 27, and 28). The standards of the short-lived nuclides sodium-24, phosphorus-32, potassium-42, iodine-131, and gold-198 were, up to 1957, each distributed twice a year. Some 6 weeks before each distribution, notification cards were sent to a mailing list consisting of some 350 potential users in North America. Normally between 50 and 100 orders were received, and the distribution planned accordingly, an appropriate number of glass ampoules being filled with the same solution (prepared by suitable dilution from a master solution) and flame-sealed. Several of these ampoules, taken at random, were then calibrated (fig. 29) and the distribution made either by air express (sodium-24, potassium-42, and gold-198) or railway express (phosphorus-32 and iodine-131).

In May of 1957, however, standard solutions of these short-lived nuclides were made available commercially and the National Bureau of Standards accordingly withdrew from the field. It continues, however, to maintain the primary standards of these nuclides and to compare them with those of other national standardizing laboratories.

In view of the difficulties that had to be surmounted in the distribution of the short-lived radioactivity standards, there have been a number of

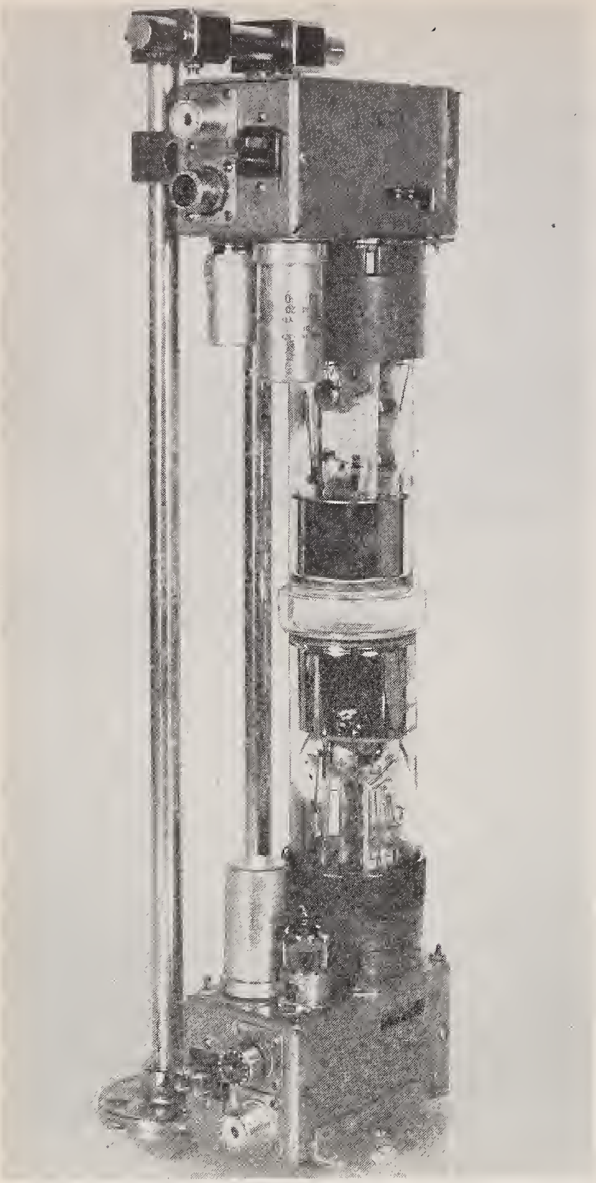

Figure 25. *Experimental arrangement for 4π-liquid-scintillation counting which is similar to that for 4π-sandwich-crystal counting shown in figure 21.*

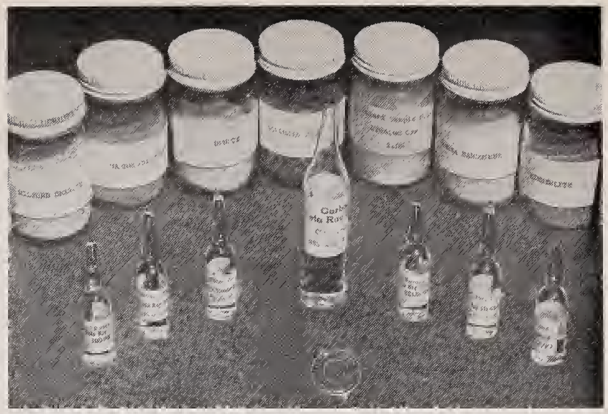

Figure 26. *A selection of radioactivity standards issued by the National Bureau of Standards.*

In the foreground is a radium D+E reference standard flanked by representative radioactivity solution standard samples. In the background are a number of radium rock ore standards and one uranium ore standard. The distribution of uranium and also of thorium ore standards has, however, been discontinued but these can currently be obtained from the Atomic Energy Commission's New Brunswick Laboratory.

attempts to provide substitute standards for these. Thus, in The Netherlands it has been shown that, using a beta counter with sodium-22, the absorption of the positrons, together with the gamma rays and annihilation radiation, in aluminum is essentially the same as that for the beta particles and gamma rays from iodine-131 up to about 90 mg/cm^2 of aluminum (Aten, 1954). The backscattering from a thick layer of aluminum was practically the same for each nuclide. Thus, iodine-131 may be assayed in terms of a standardized sample of sodium-22. In the same way, thulium-170 has been shown to have the same absorption coefficient as gold-198 for beta counting, and potassium-40 is suggested as a good substitute standard, by beta counting, for sodium-24 (Aten, 1954). Recently a remarkably close gamma-ray analogue for iodine-131, whose principal gamma rays have energies of 335 and 608 kev, has been produced by mixing the correct proportions of barium-133 (82-, 300-, and 375-kev gamma rays) and cesium-137—barium-137 (662-kev gamma rays) (Brucer, 1955). When such a simulated standard is used to calibrate a true iodine-131 sample, its effect as measured by any kind of gamma-sensitive equipment will be approximately the same as for iodine-131. It may thus have uses for calibration to within say ±3 to 5 percent when standards of short-lived iodine-131 itself do not happen to be available. A set of such "mock-iodine" standards consisting of two nominal 10-microcurie and two 50-microcurie ampoules was calibrated in the summer of 1955 at the National Bureau of Standards, the Max-Planck-Gesellschaft at Göttingen, and the National Physical Laboratory. The results, obtained by ionization chambers, Geiger-Müller and scintillation counters at these three laboratories did not differ by more than ±4 percent from the mean value.

At the National Physical Laboratory the problem of preserving short-lived radioactivity standardization outside the national standardizing laboratories is being approached by investigating the possibility of factory-producing a combined beta- and gamma-ionization chamber (Perry, Dale, and Pulfer, 1956). Four such combined chambers have already been tested at the National Physical Laboratory. It is hoped that when they are ultimately produced in quantity they will be so similar in construction that a separate calibration of each combined chamber for each nuclide will be unnecessary and that users of radioactive materials will always have available means for the calibration of individual nuclides to within ±3 percent.

TABLE 1. *Primary alpha, beta, and gamma radioactivity standards maintained by the National Bureau of Standards*

Sample No.	Radiation	Nuclide	Nominal activity[a]	Volume	Chemical form of standard	Method of primary standardization	Method of secondary standardization
4900	α	Polonium-210[b]	200 dps	(c)	Polonium plated from polonium-chloride solution.	$2\pi\alpha$-prop. counting	$2\pi\alpha$-prop. counting.
4901	α	Polonium-210[b]	500 dps	(c)	do	do	Do.
4902	α	Polonium-210[b]	1,000 dps	(c)	do	do	Do.
4903	α	U_3O_8[d]	15 dps	(e)		do	Do.
4910	$\beta(\alpha)$	RaD+E[f]	200 dps	(c)	Lead peroxide	Quantitative extraction from pitchblende.	Defined-geometry G.-M. counting.
4911	$\beta(\alpha)$	RaD+E[f]	500 dps	(c)	do	do	Do.
4912	$\beta(\alpha)$	RaD+E[f]	1,000 dps	(c)	do	do	Do.
4913	$\beta(\gamma)$	Cobalt-60	10^4 dps/ml	(g)	Chloride in HCl solution	$4\pi\beta$-prop. counting; coinc. counting.	$4\pi\gamma$-ion. chamber; formamide counting.
4914	$\gamma(\beta)$	Cobalt-60	10^5 dps	5.0 ml	do	do	Do.
4915	$\gamma(\beta)$	Cobalt-60	10^6 dps	5.0 ml	do	do	Do.
4916	β	Phosphorus-32	10^5 dps/ml	(g)	Phosphoric acid solution	$4\pi\beta$-prop. counting	$2\pi\beta$-ion. chamber; formamide counting.
4917	$\beta(\gamma)$	Iodine-131	10^5 dps/ml	(g)	Sodium iodide solution	$4\pi\beta$-prop. counting; coinc. counting.	$2\pi\beta$-$4\pi\gamma$-ion. chambers; formamide counting.
4918	$\beta(\gamma)$	Gold-198	10^5 dps/ml	(g)	Auric cyanide solution	do	Do.
4919	β	Strontium-90 Yttrium-90	10^4 dps/ml	(g)	Chloride in HCl solution	$4\pi\beta$-prop. counting	$2\pi\beta$-ion. chamber; formamide counting.
4920	β	Thallium-204	10^4 dps/ml	(g)	Thallic nitrate in HNO_3 solution.	do	Do.
4921	$\beta(\gamma)$	Sodium-22	10^4 dps/ml	(g)	Chloride in HCl solution	$4\pi\beta$-prop. counting; coinc. counting; Co^{60} comparison.	$4\pi\gamma$-ion. chamber.
4922	$\gamma(\beta)$	Sodium-22	10^6 dps	5.0 ml	do	do	Do.
4923	$\beta(\gamma)$	Sodium-24	10^5 dps/ml	(g)	do	$4\pi\beta$-prop. counting; coinc. counting.	Do.
4924	β	Carbon-14	10^3 dps/ml	25.0 ml	Sodium carbonate solution	Compensated gas counting	Formamide counting; gas ion. chamber; liq. scint. counting.
4925	β	Carbon-14[h]	10^4 dps/g	(g)	Benzoic acid in toluene	do	Do.
4926	β	Hydrogen-3	10^4 dps/ml	25.0 ml	Tritiated water solution	Calorimeter; compensated gas counting.	Liq. scint. counting; gas ion. chamber.
4927	β	Hydrogen-3	10^5 dps/ml	(g)	do	do	Do.
4928	β	Sulfur-35	10^4 dps/ml	(g)	Sodium sulfate solution	Calorimeter; $4\pi\beta$-prop. counting.	Liq. scint. counting; formamide counting.
4929	K	Iron-55	10^5 dps/ml	(g, i)	Ferric chloride in HCl solution.	Liq. scint. counting	Liq. scint. counting; X-ray counting.
4930	$K(\gamma)$	Zinc-65	10^5 dps/ml	(g)	Chloride in HCl solution	Liq. scint. counting; coinc. counting.	Do.
4931	$\gamma(\beta)$	Cesium-137 Barium-137	10^4 dps	5.0 ml	do	$4\pi\beta$-prop. counting; coinc. counting.	$4\pi\gamma$-ion. chamber.
4932	$\gamma(\beta)$	Mercury-203	10^6 dps	5.0 ml[i]	Mercurous nitrate solution	do	Do.
4933	$\gamma(\beta)$	Potassium-42	10^5 dps/ml	(g)	Chloride in HCl solution	do	Do.
4934	$\gamma(\beta)$	Tantalum-182	10^5 dps/ml	(g)	Fluoride in HCl solution	$4\pi\beta$-prop. counting	Do.
4935	β	Krypton-85	10^4 dps	(j)	Gaseous krypton	Compensated gas counting	Gas ion. chamber; gas counting.

[a] The disintegration rate as of the reference data is given on a certificate accompanying the standard.

[b] Samples consist of a practically weightless deposit of polinium-210 on a silver disk 1 in. in diameter, 1/16 in. thick, and faced with 0.002 in. of palladium.

[c] Deposited source.

[d] Samples consist of U_3O_8 deposited on a 0.1-mm-thick platinum foil and mounted on an aluminum disk, 1¼ in. in diameter and 1/32 in. thick. The alpha-ray disintegration rate as of the date of calibration and in the forward hemisphere is indicated on the certificate accompanying the standard.

[e] Evaporated source.

[f] Standards consist of Pb-210 and Bi-210 in equilibrium, deposited on a silver disk 1 in. in diameter, 1/16 in. thick, and faced with 0.002 in. of palladium.

[g] Approximately 3 ml of low-solids carrier solution containing the active nuclide in a flame-sealed ampoule.

[h] Benzoic acid in toluene for use in liquid scintillation counters.

[i] In preparation.

[j] Approximately 10 ml of krypton at STP in a break-seal ampoule.

Radium standards (for radon analysis)

Sample No.	Radium content (in grams)	Volume (in ml)
4950	10^{-9}	100
4951	10^{-11}	100
4952	Blank solution	100

Samples are sealed in glass containers. They were prepared by determining the radium content of a purified sample of radium chloride by direct comparison with the U. S. Primary Radium Standards by means of the gold-leaf electroscope and radiation balance (Mann, Stockmann, Youden, Schwebel, Mullen, and Garfinkel, 1958). This radium chloride was then transferred quantitatively to a carrier solution consisting of 0.2 percent by weight $BaCl_2 \cdot 2H_2O$ in a 5 percent by weight solution of HCl.

Radium gamma-ray standards

Sample No.	Radium content (in grams)	Volume (in ml)
4955	0.1×10^{-6}	5
4956	0.2	5
4957	0.5	5
4958	1.0	5
4959	2.0	5
4960	5.0	5
4961	10	5
4962	20	5
4963	50	5
4964	100	5

Samples are contained in flame-sealed glass ampoules. They were prepared by determining the radium content of a purified sample of radium bromide by direct comparison with the U. S. Primary Radium Standards by means of the gold-leaf electroscope. The radium bromide was then dissolved quantitatively in a 5 percent by weight solution of HNO_3.

Figure 27. *NBS Radiochemical Laboratory of the Radioactivity Section showing the experimental arrangement adopted for the dispensing of the carbon-14-labeled benzoic-acid-in-toluene standards for liquidscintillation counting.*

The master solution is shown in the triple-necked flask from which it is transferred by air pressure to the modified 25-ml volumetric flask on the right. From this latter, 3-ml aliquots are transferred to the glass ampoules by means of a spring-loaded hypodermic syringe. The ampoules are then placed in a dry-ice-alcohol bath and are flame-sealed, the toluene having been cooled to a point where its vapor pressure is less than 1 mm of mercury.

Table 1. *Radium rock standards—Continued*

Sample No.	Rock	Average radium content (gram of radium per gram of rock)
4975	Dunite	0.009±0.004×10^{-12}
4976	Carthage limestone	0.15 ±0.03
4977	Berea sandstone	0.24 ±0.02
4978	Columbia River basalt	0.33 ±0.03
4979	Chelmsford granite	2.96 ±0.08
4980	Quartzite	0.06 ±0.01
4981	Graniteville granite	3.3 ±0.2
4982	Gabbro-diorite	0.18 ±0.02
4983	Milford granite	0.23 ±0.02
4984	Triassic diabase	0.18 ±0.03
4985	Deccan trap	0.21 ±0.04
4986	Kimberlite	0.59 ±0.04

Each sample consists of 100 g of pulverized rock taken from bulk material analyzed for radium content. Petrographic data and approximate chemical analysis of a representative sample of each rock also given in a certificate accompanying each sample.

Contemporary standard for carbon-14-dating laboratories

Sample No.	Description	Quantity
4990	Oxalic acid	5 lb.

Figure 28. *Radioactivity standards are stored in an outside brick room on shelves with brick and concrete lips having a thickness of 8 inches.*

In this photograph carbon-14 and radium-226 standards can be seen reflected in the mirror at the back of such a shelf. The radium-226 ampoules are packaged in plastic containers with absorbent cotton.

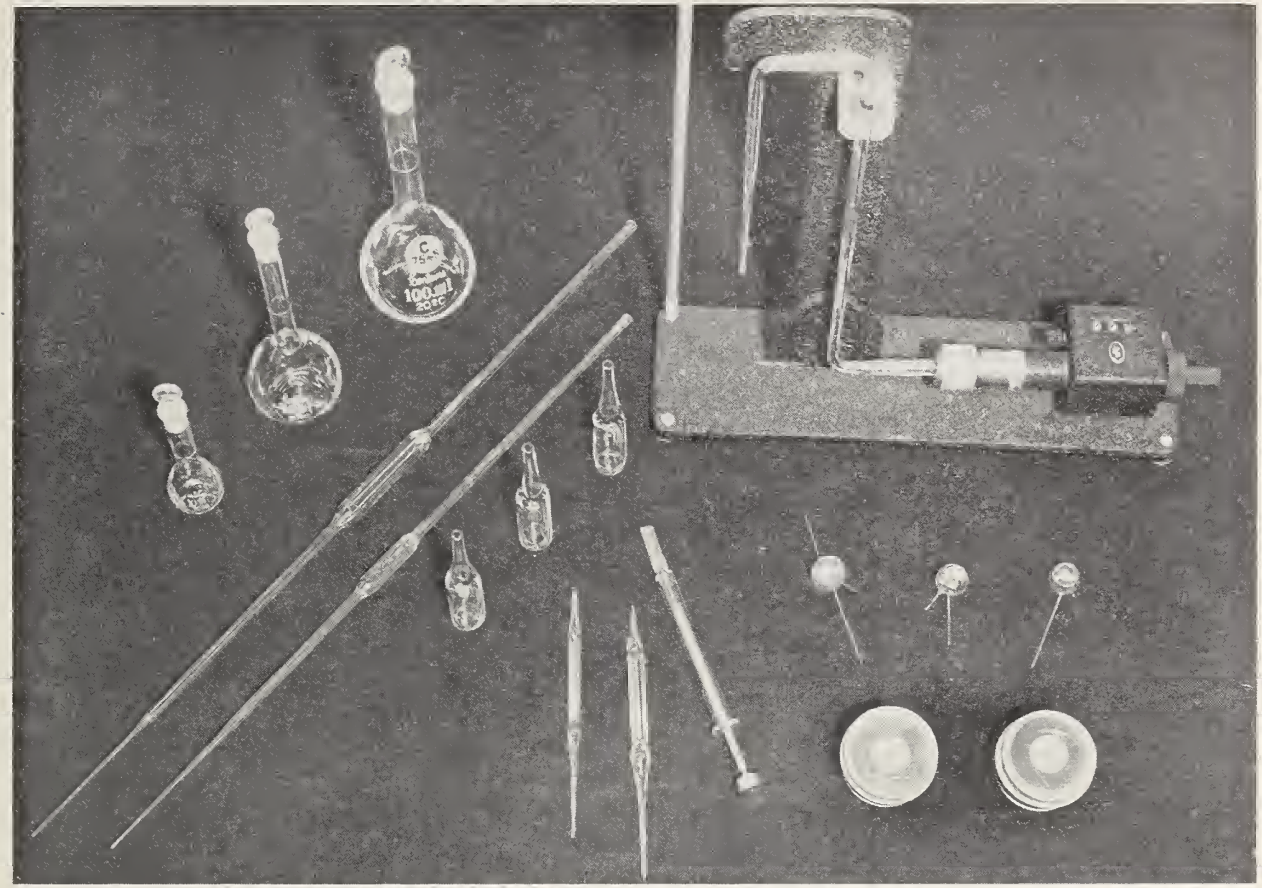

Figure 29. *Apparatus used for preparation of sources for $4\pi\beta$ counting.*

In the right rear is a mercury-displacement type ultramicroburet capable of delivering 0.1 ml in 1,000 divisions. The displacement of an accurately-machined stainless steel plunger is read directly on a micrometer dial. In the right center are extremely light pycnometers also used for delivering aliquots of solutions to $4\pi\beta$-counter source mounts in the foreground. The fine wire by which the pycnometer is suspended from the balance arm can be seen attached to the one farthest to the left. The pycnometer is filled by first heating the body with the tip immersed in the active solution. A small soldering-iron tip, brought *near* the body while it is suspended from the balance arm can control the delivery of single droplets. The long capillary neck permits the pycnometer to be used for hours with negligible change of weight due to evaporation.

4. Use of Radioactivity Standards in the Laboratory

Determinations of the number of alpha particles emitted by radium per second per gram were first made by Rutherford and Geiger (1908), using their newly developed spark counter (fig. 30), and by Sir James Dewar (1910), who measured the rate of production of helium from about 70 mg of pure radium chloride. In the same year (1908), Regener attempted to detect alpha particles quantitatively by counting the scintillations produced in zinc sulfide and using the emission constant determined by Rutherford and Geiger, and by Dewar. The determination of the efficiency of the zinc-sulfide scintillation counter for alpha particles can therefore be considered to be the first direct application of a standard of radioactivity.

Also in 1908, Soddy (1908) measured the production of helium in purified uranium, leading to a value for the half-life of the latter element. This measurement had a direct application in answering one of the oldest questions of mankind, namely, how old is the earth? Radioactive materials and particularly uranium provided a process wherein the rate of progress of the radioactive decay was independent of external physical or chemical conditions and in which the element was sufficiently long lived so as not to have decayed appreciably during the entire process.

In 1912, Mosely (1912), using radium B+C, determined that within his experimental error there was only one electron emitted per beta disintegration. This result, while not a standardization *per se,* forms an essential link in the interpretation of the subsequent results of Ellis and Wooster (1927), who determined the mean energy per radium E beta disintegration. To do this they measured in a sensitive calorimeter the total rate of energy emission of a separated radium E sample and its subsequent polonium-210 daughter. From this they determined the ratio of the rate of energy emission of radium E to polonium-210, using the known decay constants. They also assumed the energy of the polonium-210 alpha particles to be 5.22 Mev.

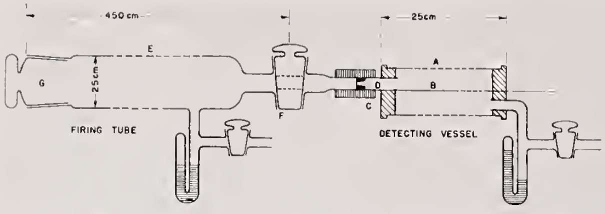

FIGURE 30. *A schematic drawing of the original Geiger-Rutherford alpha-particle counter in which for the first time, in 1912, it was shown to be practicable to detect a single particle by means of its electrical effect.*

A source of alpha particles on a thin film was placed in the large tube E which was then evacuated. When the stopcock F was opened a small fraction of the alpha particles entered the aperture into the counter which contained air at a few centimeters of mercury pressure. If the voltage was adjusted properly, the ionization by the alpha particle in the vicinity of the wire B caused an electrical breakdown of the air which was detected by means of an electrometer connected to ground through a high resistance.

From the primary applications of standards of radioactivity referred to above, namely (a) determination of absolute efficiencies of detectors, (b) determination of long half-lives, and (c) determination of mean energies, there have been derived today a number of related applications. There are measurements of absolute cross sections, activation analyses, and activity measurements for medical, industrial, and military uses and uranium prospecting, all based on (a): measurements of historical and geologic dates, based on (b); and measurements of W (the energy expended per ion pair), electron penetration, and depth-dose-rate for medical applications, based on (c). The use of the half-life of carbon-14 for historical dating has become an archaeological tool of extremely high resolving power, the latest and most important contribution being the accurate dating of the Dead Sea Scrolls.

It is important to distinguish between the applications of *radioactivity* and the applications of *standards of radioactivity*. Any report encompassing the former would be monumental, and would only mostly describe the same kind of precise relative measurements. However, even in these cases, for radiation safety, for planning experimental details, and for ordering and disposing of active material, a knowledge of the disintegration rates of the material is essential. The experimenter must account for all of the initial radioactivity in order to obtain a material balance in his experimental results. Thus the total efficiencies under differing experimental conditions of his various radiation detectors must be known. In the following a summary of the applications of standards of radioactivity will be presented. A large part will be devoted to suggested methods for the use of radioactivity standards for calibration purposes, and the various methods of measurement of radioactivity will be discussed. The remainder of the sections will be concerned with long-half-life measurements, historical and geologic dating, and measurements based on the mean energy of beta disintegration. The literature references are by no means exhaustive but are representative of the applications of standards of radioactivity.

The *total efficiency*, ϵ, of a detector for a source prepared from a radioactivity standard is defined as:

$$\epsilon = \frac{\text{counts per second observed for source}}{\text{disintegrations per second in source}}. \qquad (1)$$

The total efficiency is compounded of the *geometry* (i. e., the fractional solid angle subtended by the detector) and the *detector efficiency* (i. e., the probability of a measurable response from a radiation striking the detector).

Beta-ray solution standards distributed by the National Bureau of Standards are contained in flame-sealed glass ampoules and consist of approximately 3 ml of an aqueous carrier solution of the active nuclide. A certificate specifies the disintegration rate per second per milliliter as of the zero date. Ampoules intended for use as gamma-ray standards contain an accurately known volume of solution (5 ml), and in these cases the total disintegration rate and the exact volume of solution at a given temperature are also specified. Radium D+E beta-ray reference sources, radium F (polonium-210) alpha-ray standards, and uranium oxide (U_3O_8) alpha-ray standards consist of the active material already deposited on source mounts.

In table 2 are listed most of the important radionuclides, their decay properties, and the preferred methods to be used in the laboratory for the routine assay of samples. Nuclides now standardized or shortly to be standardized at the National Bureau of Standards are printed in *italic* type. The beta-ray energies available cover the range from the 18.9-kev spectrum of hydrogen-3 to the 3.58-Mev spectrum of potassium-42. Gamma-ray energies cover the range from the 87-kev line of cadmium-109 to the 2.758-Mev line of sodium-24, as well as the X-rays from the electron capturers such as iron-55 and zinc-65.

5. Methods of Measurement

Table 3 lists the most general methods available for the measurement of alpha, beta, and gamma emitters, together with a brief description of the counting conditions and the range of applicability of each method. In this connection the term Background Equivalent Activity (BEA) has been found to be a convenient method of expressing the intrinsic sensitivity of a detector (Seliger and Schwebel, 1954). In all measurements of radioactivity there is a background effect without the source which must be subtracted from the observed reading taken with the source present. The magnitude of this background, or more specifically the ratio of the source effect to background effect, may place practical time limitations upon the conduct of an experiment. It is for this reason that the term Background Equivalent Activity (BEA) was devised, this being the disintegration rate of the

Table 2. *Preferred methods of measurement for selected radionuclides*

Nuclide	$E_{\beta_{\max}}$ (Mev)	Principal E_γ (Mev)	$T_{1/2}$	Suggested methods for routine assay [a]
H^{3}	0.0189	--------------	12.3 y	Liquid scintillation counter; internal gas counter; internal gas ion chamber.
C^{14}	0.155	--------------	5600 y	$2\pi\beta$ windowless counter; liquid scintillation counter; internal gas counter; internal gas ion. chamber.
Na^{22}	0.542	0.511 1.28	2.565 y	$2\pi\beta$ windowless counter; well-type scintillation counter.
Na^{24}	1.39	2.758 1.380	14.97 h	Do.
P^{32}	1.71	--------------	14.3 d	$2\pi\beta$ windowless counter; well-type scintillation counter (bremsstrahlung).
S^{35}	0.167	--------------	87.1 d	$2\pi\beta$ windowless counter; liquid scintillation counter.
Cl^{36}	0.713	--------------	4×10^{5} y	Do.
A^{37}	EC	Cl$K\alpha$	35.0 d	Internal gas counter; internal gas ion. chambers.
K^{40}	1.33 EC	1.45, A$K\alpha$	1.3×10^{9} y	$2\pi\beta$ windowless counter; well-type scintillation counter.
K^{42}	2.04 3.58	1.51	12.44 h	Do.
Ca^{45}	0.254	--------------	152 d	$2\pi\beta$ windowless counter; liquid scintillation counter.
Sc^{46}	0.36 1.2 0.5%	1.12, 0.89, 0.89, 0.32	85 d	$2\pi\beta$ windowless counter; well-type scintillation counter.
Cr^{51}	EC	V$K\alpha$	27.8 d	X-ray counter; well-type scintillation counter; liquid scintillation counter.
Mn^{54}	EC	0.84 Cr$K\alpha$	291 d	Do.
Fe^{55}	EC	Mn$K\alpha$	2.94 y	Do.
Fe^{59}	0.260 0.460	1.3 1.1	45.1 d	$2\pi\beta$ windowless counter; well-type scintillation counter.
Co^{60}	0.310	1.17 1.33	5.26 y	Do.
Ni^{59}	EC	Co$K\alpha$	8×10^{4} y	X-ray counter; well-type scintillation counter.
Ni^{63}	0.067	--------------	85 y	$2\pi\beta$ windowless counter; liquid scintillation counter.
Zn^{65}	EC 0.325 1.5%	1.11, Cu$K\alpha$ 0.511	240 d	X-ray counter; well-type scintillation counter.
Ga^{67}	EC	Complex Zn$K\alpha$	77.9 h	Do.
As^{74}	β^-1.36, 0.69 β^+1.53, 0.92	0.596 0.547, 0.787	17.5 d	$2\pi\beta$ windowless counter; well-type scintillation counter.
Br^{82}	0.465	Complex	35.9 h	Do.
Kr^{85}	0.695	0.54 0.6%	10.2 y	Well-type scintillation counter; gas counter; internal gas ion. chamber.
Rb^{86}	0.716 1.822	1.081	19.5 d	$2\pi\beta$ windowless counter; well-type scintillation counter.
Rb^{87}	0.130	0.034	6×10^{10} y	Do.
Sr^{85}	EC	0.51	65 d	X-ray counter; well-type scintillation counter.
Sr^{89}	1.46	--------------	53 d	$2\pi\beta$ windowless counter; well-type scintillation counter (bremsstrahlung).
Sr^{90}	0.61	--------------	19.9 y	Do.
Y^{90}	2.180	--------------	61 h	Do.
Zr^{95}	0.371 0 84 isomer	0.721	65 d	$2\pi\beta$ windowless counter; well-type scintillation counter.
Nb^{95}	0.160	0.745	35 d	Do.
Tc^{99}	0.290	--------------	2×10^{5} y	Do.
Ru^{103}	0.217 0.698	0.498	39.8 d	Do.
Ru^{106}	0.039	0.52 0.62	1 y	$2\pi\beta$ windowless counter; liquid scintillation counter.
Ag^{110}	Complex	Complex	270 d	$2\pi\beta$ windowless counter; well-type scintillation counter.
Cd^{109}	EC	0.087, Ag$K\alpha$	470 d	X-ray counter; well-type scintillation counter.
Sn^{113}	EC	In$K\alpha$	112 d	Do.
I^{126}	0.850 1.27	0.382	13 d	$2\pi\beta$ windowless counter; well-type scintillation counter.
I^{121}	0.255 0.600	0.080 0.364 0.638	8.08 d	Do.
Cs^{134}	0.090 0.648	0.602 0.794	2.3 y	Do.
Cs^{137}	0.510 1.20	--------------	30 y	Do.
Ba^{137}	--------------	0.661	2.6 m	Do.
Ba^{133}	EC	0.085, Cs$K\alpha$	10 y	X-ray counter; well-type scintillation counter.
Ce^{141}	0.442 0.581	0.141	33.1 d	$2\pi\beta$ windowless counter; well-type scintillation counter.
Ce^{144}	0.300, 0.170	Complex	282 d	Do.
Pr^{144}	3.0	1%	17 m	Do.

TABLE 2. *Preferred methods of measurement for selected radionuclides*—Continued

Nuclide	$E_{\beta_{\max}}$ (Mev)	Principal E_γ (Mev)	$T_{1/2}$	Suggested methods for routine assay [a]
Pm^{143}	EC	0.067, Nd$K\alpha$	200 d	X-ray counter; well-type scintillation counter.
Eu^{154}	0.300 0.700 1.900	0.778 1.2 0.336	16 y	$2\pi\beta$ windowless counter; well-type scintillation counter.
Tm^{170}	0.866 0.970	--------------	129 d	Do.
Ta^{182}	0.250 0.530	Complex	115 d	Do.
Ir^{192}	0.670	Complex	74.4 d	Do.
Au^{198}	0.970	0.411	2.698 d	Do.
Hg^{203}	0.208	0.279	47.9 d	Do.
Tl^{204}	0.783	--------------	4.0 y	$2\pi\beta$ windowless counter.
Pb^{210}	0.026	0.007	22 y ?	$2\pi\beta$ windowless counter; liquid scintillation counter.
Bi^{210}	1.17	--------------	5.0 d	$2\pi\beta$ windowless counter; well-type scintillation counter (bremsstrahlung).
Po^{210}	α5.3	0.773 10^{-3}%	138.40 d	$2\pi\beta$ windowless counter; ZnS α scintillation counter.
Ra^{226}	α4.8	Complex	1620 y	$4\pi\gamma$ ionization chamber; well-type scintillation counter.

[a] The methods suggested are the most efficient. This does not preclude the use of the quartz-fiber electroscope or the standard G-M counter for routine measurements.

particular isotope required to equal the background reading.

If we let A be the observed sample-plus-background counts observed in a given time and let B equal the background counts observed with no sample present, then the standard deviation of the net counting rate $A-B$ is given by

$$\sigma=(A+B)^{1/2}, \tag{2}$$

and the relative error of the net counting rate is

$$\frac{\sigma}{(A-B)}=\frac{(A+B)^{1/2}}{A-B}. \tag{3}$$

Thus the relative error of a measurement of a BEA is always $171/\sqrt{B}$ percent where $\sqrt{B}$ is the standard deviation of the background count.

The $2\pi\beta$ windowless-gas-flow proportional counter is considered to be the most sensitive and versatile apparatus for the routine counting of all types of beta sources, followed closely by the $2\pi\beta$-windowless electroscope (fig. 31). The well-type NaI (Tl) scintillation counter has the highest sensitivity for gamma rays of any detector now available (Baskin, Demorest, and Sandhaus, 1954; Haigh, 1954). With these instruments one can carry out almost any type of measurement involving radioactive materials. Where cost and trouble-free maintenance are overriding factors, the electroscope and the end-window G-M counter are efficient substitutes for the windowless flow counter, and a cylindrical arrangement of several gamma-ray G–M counters surrounding the source can substitute for the well-type scintillation counter. For these substitute methods the BEA is, of course, increased, especially so for the gamma-ray arrangement.

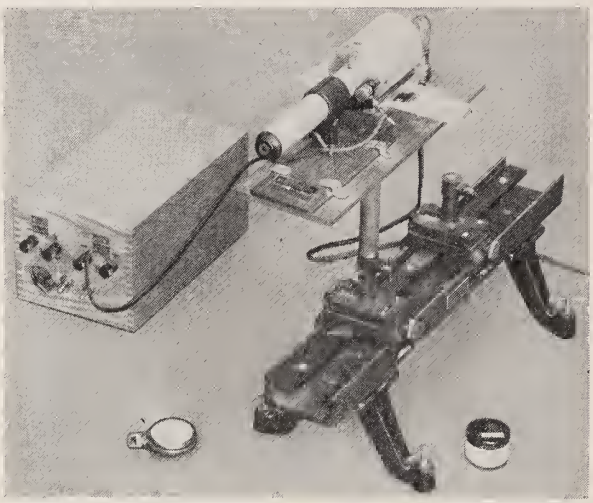

FIGURE 31. *Photograph of $2\pi\beta$ windowless electroscope used for secondary standardization and for general relative counting.*

The instrument is an adaptation of the Lauritsen electroscope as described by Hunter and Mann (1948). Inside the partly inserted sliding shelf is an NBS radium D+E reference source. If the same region of the scale is traversed for each timing interval such a high degree of precision can be obtained with a relatively simple and inexpensive instrument.

The liquid scintillation counter has recently come into limited use for the routine measurement of low-energy beta emitters. At the present time, under proper experimental conditions, it is possible to prepare a liquid scintillator which is more

Table 3. *General methods of measurement for alpha, beta, and gamma emitters*

Detector	Auxiliary equipment required [a]	Types of sources	Range of applicability and BEA [b]
1. $2\pi\beta$-air-ionization chamber (recommended for secondary standardization at a large installation).	High-voltage supply, Vibrating-reed electrometer, *or* Lindemann - Ryerson electrometer Projector Calibrated condensers Voltmeter Stopwatch	Dried on planchet; liquid in glass or metal cup forming part of periphery of chamber.	Sensitive mainly to alpha and beta rays. Higher intensity gamma-ray sources can also be measured. Small self-absorption effects for low-energy beta emitters. BEA for P^{32} dried sources—0.001 μc. BEA for P^{32} liquid sources—0.01 μc.
2. $4\pi\gamma$-air-ionization chamber (recommended for secondary standardization at a large installation).	High-voltage supply, Vibrating-reed electrometer, *or* Lindemann - Ryerson electrometer Projector Calibrated condensers Voltmeter Stopwatch	Liquid in ampoule; test-tube inserted into reentrant cylinder.	Beta rays are completely absorbed within the reentrant cylinder. Ionization is due to bremsstrahlung and to gamma rays. Big advantage is nondependence of reading on position or volume of source within the chamber. Can be used to compare high-energy beta emitters from bremsstrahlung measurements. BEA for I^{131} in solution—1.4 μc. BEA for P^{32} in solution—20 μc. High-pressure chambers are more sensitive.
3. $2\pi\beta$-windowless-proportional flow counter (recommended for all beta-ray measurements on a routine basis).	High-voltage supply Non-overloading amplifier Discriminator Scaler 90% argon, 10% methane *or* helium isobutane, *or* 100% methane.	Dried on planchet; liquid, in formamide or other low-vapor-pressure solvent, delivered to stainless-steel cup.	Most sensitive method for beta rays. Higher-intensity gamma-ray sources can also be measured. The efficiency for beta rays is practically 100% over the central region, becoming 90% at periphery. Simple to operate and to decontaminate. Alpha particles can be counted in presence of betas at lower voltages in the proportional region. BEA for P^{32} dried source—0.0001 μc. BEA for P^{32} as thick source—0.001 μc. BEA for C^{14} as liquid—0.001 μc.
4. Well-type NaI scintillation counter (recommended for all gamma-ray measurements on a routine basis).	Precision high-voltage supply Nonoverloading linear amplifier Discriminator Multiplier Phototube Scaler Single-channel pulse-height analyzer	Liquid in ampoule; test-tube inserted into well. Source could also be external in conditions of "good" geometry.	Beta rays are completely absorbed in walls of crystal holder. Light output to phototube is due to bremsstrahlung and to gamma rays. Most sensitive method for gamma rays. Tremendous increase in sensitivity over air-ionization chamber. Light output is directly proportional to energy absorbed—permits identification of isotopes by gamma photopeaks. Window pulse-height analyser permits counting one photopeak to *exclusion* of others in mixtures of nuclides. Relatively independent of source position or size inside well. BEA for I^{131} in 5-ml ampoule—0.0003 μc. BEA for Cr^{51} in 5-ml ampoule—0.003 μc. BEA for P^{32} in 5 ml ampoule—0.02 μc.
5. Liquid scintillation counter (recommended for double beta-labeling experiments. Useful but not essential for low-energy beta emitters and electron capturers).	Precision high-voltage supply Non-overloading linear amplifier Discriminator Multiplier phototube Scaler Single-channel pulse-height analyser	Liquid incorporated into solution of scintillator as water - alcohol - toluene, water-dioxane, or an organic tagged molecule such as C^{14}-benzoic acid.	Useful for measurement of low-energy beta-emitters such as H^3, Ni^{63} and C^{14} and electron-capturing nuclides such as Cr^{51}, Fe^{55}, Ni^{59}—eliminates self-absorption effects. Useful for double- and triple-labeling experiments because of linear response to energy absorbed. Can discriminate between α's and β's. BEA for H_2^3O in 5-ml cell—0.01 μc. BEA for C^{14} benzoic acid in 5-ml cell—0.004 μc.
6. Mica end-window G-M counter (lower-priced substitute for $2\pi\beta$-windowless counter).	High-voltage supply Scaler	Dried on planchet; liquid in glass or metal cup directly below window.	Almost as sensitive to betas as $2\pi\beta$-flow counter. 100% sensitive to electrons entering sensitive volume. Air and window absorption lower efficiency for low-energy beta emitters. Does not discriminate between α's and β's as in 2π flow counter. Efficiency for gamma rays same as $2\pi\beta$-flow counter (1–2%). Halogen self-quenching tube gives long life, trouble-free service. Standard shelf arrangement gives reproducible geometry. BEA for P^{32} dried source in top shelf position—0.0001 μc.

TABLE 3. *General methods of measurement for alpha, beta, and gamma emitters*—Continued

Detector	Auxiliary equipment required [a]	Types of sources	Range of applicability and BEA [b]
7. Thin-window argon, krypton, or xenon X-ray counter (specifically for use with electron-capturing nuclides).	High-voltage supply Linear amplifier Discriminator Scaler Single-channel pulse-height analyser	Dried on planchet; liquid in glass or metal cup directly below window.	Makes use of critical or near-critical absorption of gas for X-rays near critical absorption limit. Useful for pure electron-capturing nuclides. Window-absorption reduces efficiency for low-energy X-rays. Linear response makes *identification* and *differential* counting of mixtures of gamma-emitting nuclides possible. BEA for Fe^{55} dried source—0.0003 μc.
8. Quartz-fiber electroscope (lower priced substitute for $2\pi\beta$ windowless counter).	Battery for charging Battery for pilot light Stopwatch	Dried on planchet, on slide mounted external or internal to electroscope. Internal liquid sources not recommended due to difficulty in decontamination.	Very-low-capacity system slightly more sensitive to internal beta sources than $2\pi\beta$-air-ionization chamber. Low sensitivity to gamma rays. Displacement of fiber is slightly nonlinear. Excellent for relative measurements over same region of scale. Simple, trouble-free operation with care. Can be used for low-energy beta emitters with same precautions as $2\pi\beta$-air-ionization chamber and for higher intensity external gamma sources. BEA for P^{32} dried source—0.001 μc.
9. Internal-gas-G-M counter	High-voltage supply Scaler Chemical conversion system	Nuclide converted to inert or counting gas; small amount present in vapor phase so as not to interfere with operation.	Useful in some cases for routine analyses of tritium or carbon-14 where material can be converted to gaseous phase. Can also be used for counting small amounts of THO as vapor. 100% efficient for beta particles. Small end and wall effects. BEA for C^{14} as C^{14}O$_2$—0.00003 μc.
10. Internal-gas-ionization chamber.	High-voltage supply Vibrating-reed electrometer Chemical conversion system	Nuclide converted to gaseous phase.	Less sensitive than internal gas counter. However, the technique is less critical as to the type of gas present. Good G-M characteristics are not required so that larger amounts of the isotope as a gas may be counted. Sensitivity similar to $2\pi\beta$-air-ionization chamber. BEA for C^{14} as C^{14}O$_2$—0.001 μc.
11. ZnS alpha scintillation counter (used where β or γ counting rates interfere with α detection).	Precision high-voltage supply Linear amplifier Discriminator Multiplier phototube Scaler	Dried on planchet	Detects α's in presence of high intensity β's and γ's. Efficiency is practically 100% for α particles striking ZnS film.

[a] All equipment listed is available commercially in U. S. A.

[b] BEA (Background Equivalent Activity) is a measure of the intrinsic sensitivity of a detector. It is the number of disintegrations per second of the particular nuclide that will produce an effect equal to the background effect.

efficient than anthracene (Seliger and Ziegler, 1956a, 1956b, and 1957). The most efficient liquid scintillator combinations reported to date are approximately 4 g/liter 2,5-diphenyloxazole (DPO) with 0.1 g/liter 1,4-di-(2(5-phenyloxazole))-benzene (POPOP) in toluene, and approximately 8 g/liter phenylbiphenyloxadiazole (PBD) zene (POPOP) in toluene (Hayes, Orr, and Kerr, 1956). Under carefully controlled conditions of solution preparation and measurement it has recently been possible to intercompare tritiated-water standards received from Atomic Energy of Canada Limited, Chalk River, Canada, and Los Alamos Scientific Laboratory, Los Alamos, New Mexico, with the NBS tritiated-water standard with a precision of a few tenths of 1 percent (fig. 32). In this case a water: alcohol: xylene ratio of 1: 50: 250 was used in the system with 4 g/liter DPO as the scintillating solute. For certain other tracer applications involving lower activities of material and therefore requiring the measurement of larger volumes of water, the use of dioxane as a solvent will permit a much larger percentage of water to be incorporated into the measuring cell (Langham, Eversole, Hayes, and Trujillo, 1956).

5.1. Considerations in Beta-Ray Calibrations

To use beta-ray solution standards it is necessary to open the standard ampoule and to prepare a standard source for comparison, or for calibration of a counting system. Assuming that the electronic equipment is suitable and the detector adequately stable, the following are the most important considerations:

a. Source Self-Absorption

Most beta-ray solution standards distributed by the National Bureau of Standards have a minimum of total solids present, consistent with chemical stability and freedom from adsorption effects. In some instances, however, where the

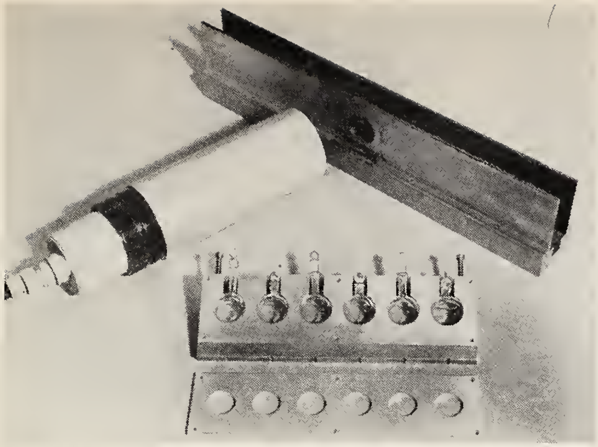

Figure 32. *Sliding sample arrangement used for comparing liquid scintillating samples.*

The slots on the bottom make for reproducibility of setting. The black felt on the sides of the slide holder prevents any but the light from the sample being viewed from reaching the photocathode of the phototube. The cells are 1-inch-diameter Pyrex absorption cells. The flame-sealed sample at the extreme right is a blank solution for determination of background. With this arrangement, a precision of 0.1 percent was obtained.

original material may be of low specific activity, as, for example, in the carbon-14 solution standards, the absorption of beta rays from a dried source may be quite appreciable on account of the amount of solids present. Because of the low energy in the case of the carbon-14 beta-ray spectrum ($E_{max}=155$ kev), the observed counting rate will be a strong function of the amount of solids present *as well as the method of source preparation.* Even in the case of practically weightless deposits of cobalt-60 ($E_{max}=310$ kev), Seliger and Schwebel (1954) have found as much as a 10-percent difference in observed counting rate in a $4\pi\beta$ counter, depending on the method of preparation of the source. This was observed independently by Smith and Wildblood (1953). It is therefore suggested in the cases of low-energy beta emitters that comparison sources be prepared from carrier solutions of nearly identical solid concentrations when low to medium solid sources are desired. Otherwise "infinitely" thick liquid or solid sources should be prepared. The $2\pi\beta$-gas-flow proportional counter has almost 100 percent efficiency for a beta particle entering the sensitive volume. A method of counting, using this type of counter for infinitely thick liquid samples has been described by Schwebel, Isbell, and Karabinos (1951) and Schwebel, Isbell, and Moyer (1954), in which the radioactive material is dissolved in formamide, an organic solvent having a very low vapor pressure compared with water. In this method a diluting solution of formamide containing 1 part by volume of carrier solution to 99 parts of formamide is prepared. The radioactive solution to be standardized is then diluted by a factor of 100 with the formamide-carrier solution to reduce the water concentration to 1 percent (fig. 33). At this low concentration water vapor does not appear to change the counting characteristics of the $2\pi\beta$ flow counter. Usually 1 ml of the 1-percent solution is pipetted into a shallow stainless-steel cell. As the thickness of the solution is greater than the beta-particle saturation thickness, the counting rate is proportional only to the solution activity and to the area of the cell. If the volume and hence the depth of solution is always the same, any effects due to gamma rays will be constant. Thus the formamide method of liquid counting can also be used for beta emitters that have accompanying gamma rays.

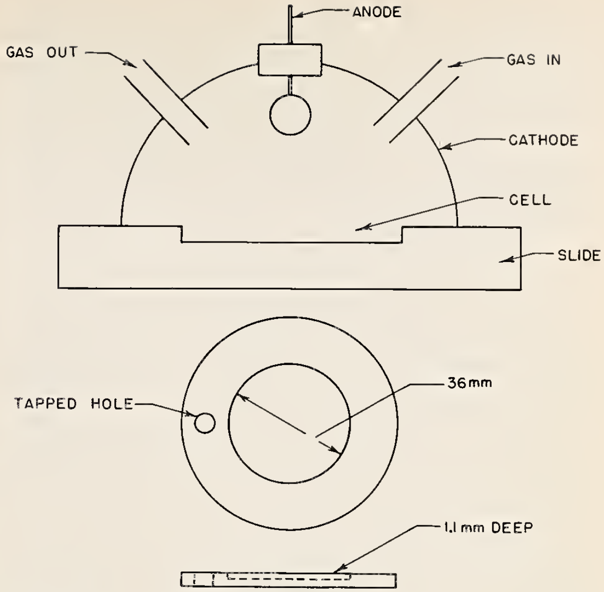

Figure 33. *Schematic drawing of the NBS formamide counter (Schwebel, Isbell, and Karabinos, 1951; Schwebel, Isbell, and Moyer, 1954).*

The cell, to which the active material in 99-percent formamide solution is added, is made of stainless steel. At 1-percent concentration in formamide, water vapor apparently does not change the counting characteristics of the $2\pi\beta$-gas-flow counter.

"Infinitely" thick solid samples can be prepared uniformly from slurries of active material containing carrier. Care must be taken in the use of slurries or partial digestions or wet ashings because of slight differences in self-absorption and scattering due to changes in deposition of colloidal particles and possible settling. It may be necessary to stir the sample continuously or to use rotating planchets to average differences in concentration over the source. Even in the cases of the infinitely thick solid and liquid sources it must be remembered that the effective atomic number of the material will to some extent determine the amount of self-scattering and therefore the self-absorption (Nervik and Stevenson, 1952; Walton, Thompson, and Croall, 1953; Bowles and Walton, 1954; Suttle and Libby, 1955). In order to obtain reproducible measurements the chemical compositions of the samples being compared must be as alike as possible. For example, if identical areas of infinitely thick sources of carbon-14 as sodium carbonate and of carbon-14 as barium carbonate are compared, the barium-carbonate carbon-14 will give the higher reading even though the sources have the same specific activity. For beta emitters of $E_{\max}$ greater than 300 kev the problem of source self-absorption is not so important, and samples can be intercompared by drying aliquots on planchets and inserting these in position in the $2\pi\beta$-flow counter. However, there is yet another important consideration.

b. Backscattering

For the routine analysis of dried samples of material various workers have used planchets or cups made of aluminum, glass, copper, stainless steel, palladium, or platinum, on which aliquots of solution were dried. The conditions of the experiment and the economic limitations of the cost of sample holders will determine the type of backings used. In any case, one must carry out all the measurements using the same type of backing, since the backscattering of beta rays is dependent on the atomic number of the backing material (fig. 34). This atomic-number dependence has been investigated for both electrons and positrons (Seliger, 1950 and 1952).

The effect of the self-scattering on observations made with end-window and 2π-geometry counters was shown very strikingly by Bowles and Walton (1954) and is reproduced in figure 35.

Under certain conditions the use of a nonconductor such as glass as a source backing can induce electric charges inside a counter. At the National Bureau of Standards sources are dried on palladium-faced silver disks. Palladium is less expensive than either gold or platinum, is close to silver in atomic number and therefore in backscattering properties and, unlike aluminum, copper, steel, and silver itself, is inert to most acids and bases.

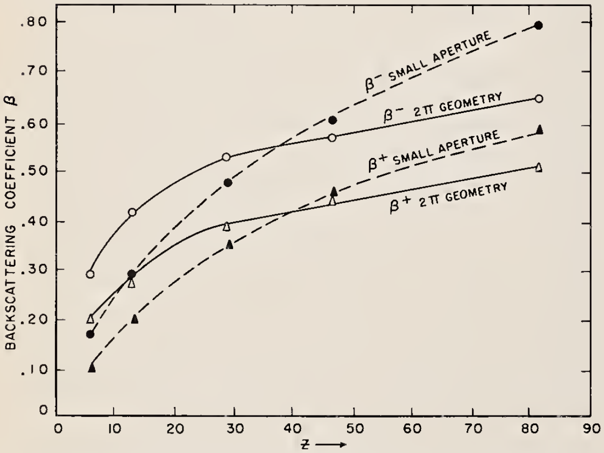

FIGURE 34. *Backscattering coefficients for positrons and electrons as functions of the atomic number of the backing material for both end-window (low) geometry and 2π geometry (Seliger 1952).*

The dependence of the backscattering coefficient upon the geometry of the measurement is due to the anisotropy in the angular distribution of the backscattered radiation. In the comparison of different samples, the source backings should be of the same atomic number.

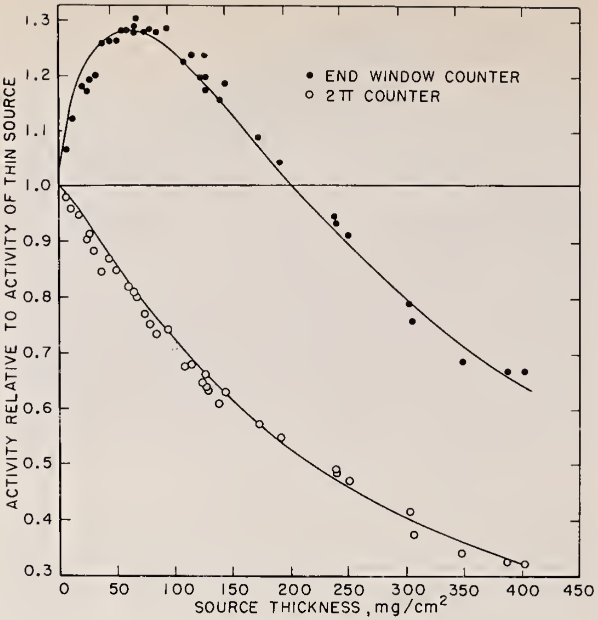

FIGURE 35. *Results obtained by Bowles and Walton (1954) showing the effect of geometry and source self-scattering on the counting rates observed from thick sources.*

The counting rate relative to that of a thin, almost weightless, source is plotted as a function of the thickness of the source, with the activity of the source remaining constant. From symmetry considerations the monotonic decreasing curve obtained with the $2\pi\beta$ counter is to be expected since an increase in solids can lead only to an increase in source self-absorption. However in the case of the end-window counter, with a low geometry, it is possible for beta rays to be anisotropically scattered into the forward direction, producing the anomalous effect shown in the figure. The same type of curve can be obtained for a thin source when an absorption curve in aluminum is plotted if the aluminum absorbers are placed on top of the source instead of directly in front of the counter window. Here again the self-focusing is initially more important than the absorption. Some examples of anisotropic angular distributions of backscattered beta rays have been given by Seliger (1952). (Reproduced, by courtesy, from AERA Report C/R 1463.)

c. Low-Solids Source Preparation

For the delivery of a small-volume source there is available a delivery-type mercury-displacement ultramicroburet that can deliver quickly and precisely volumes of solution as small as 0.01 ml with an accuracy of 0.5 percent. No particular problems are encountered in the preparation of most medium- and high-energy beta sources. The technique of air drying at room temperature has been quite satisfactory. For low-energy beta emitters and even in those cases of the higher-energy beta emitters such as $Na^{22}Cl$ and $Sr^{90}Cl_2$, which form crystalline salts, it is necessary to prepare sources carefully to avoid self-absorption effects. Methods that are employed to obtain minimum self-absorption include freeze drying (Perry, 1954b); precipitation by addition of specific salts (such as $AgNO_3$ added to NaI^{131} solutions to precipitate AgI^{131}); slow evaporation in a saturated ammonia atmosphere (Seliger and Schwebel, 1954), in which ammonia induces precipitation in the form of an ammonium complex; vacuum distillation from organic chelate compounds (Pate and Yaffe, 1956); vacuum evaporation at higher temperatures (Stockendal and

Bergkvist, 1957); and adsorption of radioactive gases onto thin evaporated films (Pohm, Waddell, Powers, and Jensen, 1954).

5.2. Consideration in Gamma-Ray Calibrations

Except under special conditions, it is not intended that a gamma-ray solution standard be opened or destroyed. The following procedure is suggested for the application of a 5-ml gamma-ray standard. A stock solution of the same isotope is diluted so that 5 ml of the final dilution contains approximately the same activity as the standard ampoule. A "laboratory standard" ampoule of the same dimensions as the standard ampoule is prepared containing an accurately known volume close to 5 ml. The laboratory standard is then compared directly with the primary standard under identical conditions of geometry, e. g., in a well-type scintillation counter, thus standardizing the entire solution. In this way the radioactivity standard can be retained as a check on further solutions, or in case of any doubts as to chemical stability or losses from the solutions used for the actual experiments. The detector efficiencies can be determined again as in eq (1) from the newly-prepared "laboratory standard" or else other types of special sources can be prepared from it.

Some electron-capturing nuclides such as manganese-54, zinc-65, and cadmium-109 have nuclear gamma rays associated with their decay. These therefore can be used for standardization in the same manner as other gamma-ray emitters.

These characteristic X-radiations following the process of electron capture can be used as a source of low-energy monochromatic gamma rays for the calibration of detectors. In table 4 are given those X-ray emitters of suitable half-life for practical measurements, together with the *K* X-ray quantum energy of the daughter atom and *K*-branching ratio. Electron capturers, such as iron-55 and nickel-59, have only low-energy daughter-atom X-rays associated with their decay. In these latter cases the method of measurement may involve the opening of the ampoule for the preparation of dried sources on planchets for X-ray counting and $2\pi\beta$ counting, or for the preparation of liquid sources for liquid scintillation counting. The same precautions should be observed here as for the low-energy beta emitters.

5.3. Simulated Standards and Reference Sources

It is sometimes advantageous in the cases of the short-lived radioactive nuclides to have available a simulated standard with which intermediate checks can be made of the detectors and of the total efficiency during the course of an extended experiment. A radium D+E reference source (with a half-life of 22 years) serves this purpose adequately for beta-emitters in the medium- and high-energy range.

As, however, recent measurements at the National Bureau of Standards have cast some doubt on the half-life of radium D it is necessary to recalibrate a radium D+E reference source at intervals depending on the accuracy required.

TABLE 4. *Electron-capturing nuclides*

Nuclide	Half-life	*K* X-ray energy (kev)	% *K*-capture
A^{37}	34.1 days	2. 82	100
Ca^{41}	1.2×10 years	2. 61	100
Cr^{51}	27.8 days	5. 47	92
Fe^{55}	2.94 years	6. 54	100
Ni^{59}	7.5×10^4 years	7. 71	100
Zn^{65}	245 days	8. 98	98. 5
Ge^{71}	11.4 days	10. 39	100
As^{73}	76 days	11. 10	100
Se^{75}	127 days	11. 87	100
Y^{88}	105 days	16. 10	99. 8
Tc^{95}	60 days	20. 00	99. 6
Cd^{107}	6.7 hours	25. 54	99. 3
Cd^{109}	470 days	25. 54	78
I^{126}	13.1 days	31. 82	58
Cs^{131}	9.6 days	34. 55	100
Ba^{131}	12 days	35. 97	100
Ba^{133}	9.5 years	35. 97	100
Ce^{134}	72 hours	38. 92	100
Ce^{139}	140 days	38. 92	100
Nd^{140}	3.3 days	41. 99	100
$Pm^{142,\ 143}$	Approx. 250 days	43. 57	100
$Pm^{143,\ 144}$	Approx. 300 days	43. 57	100
Sm^{145}	410 days	45. 2	100
Eu^{145}	5 days	46. 85	100
Eu^{147}	24 days	46. 85	99+
Eu^{148}	59 days	46. 85	100
Gd^{149}	9 days	48. 51	99+
Gd^{153}	236 days	48. 51	100
Tb^{153}	5.1 days	50. 2	100
Tb^{155}	190 days	50. 2	100
Tb^{157}	4.7 days	50. 2	100
Dy^{159}	134 days	52. 0	100
Ho^{162}	65 days	53. 8	85
Ho^{163}	5.2 days	53. 8	100
Tm^{167}	9.6 days	57. 5	100
Tm^{168}	85 days	57. 5	98
Yb^{169}	31.8 days	59. 4	100
Lu^{171}	600 days	61. 3	100
Hf^{175}	70 days	63. 3	100
Ta^{179}	600 days	65. 4	100
W^{178}	21.5 days	67. 5	100
W^{181}	140 days	67. 5	100
Re^{183}	155 days	69. 5	100
Re^{184}	50 days	69. 5	100
Os^{185}	97 days	71. 7	74
Ir^{190}	12.6 days	73. 9	100
Pt^{191}	3 days	76. 1	100
Au^{195}	180 days	78. 4	100
Au^{196}	5.6 days	78. 4	95
Hg^{197}	65 hours	80. 7	100
Tl^{202}	12.5 days	83. 1	100

Calibrated radium D+E sources from the National Bureau of Standards can also be used for standardization that is accurate to ±5 to 10 percent for nuclides whose beta-ray spectra are not too dissimilar to the radium-E beta-ray spectrum. The procedure briefly is as follows: An unknown source is prepared from a stock solution on a palladium-faced silver disk similar to the standard disk, and an absorption curve in aluminum is obtained for each source with say, an end-window beta-ray G–M counter in a fixed geometry. Extrapolation of these curves to zero total absorber will correct to a first approximation for the differences in the beta energies and for slight differences in geometry from one calibration to the next (figs. 36 and 37). Since the backscattering in the energy range 300 kev to 1.7 Mev is independent of the energy distribution, one may assume that the fractional backscattering for the unknown source and for the radium-E beta rays is the same. The disintegration rate of the unknown source is given by

$$A=\frac{\text{c/s zero total absorber (unknown)}}{\text{c/s zero total absorber (Ra D+E)}}\times(\text{Ra D+E disintegration rate}). \qquad (4)$$

The radium D+E disintegration rate is given on a certificate accompanying the standard and represents the total number of radium-D disintegrations per second contained in the source. Thirty days after preparation of the standard, the radium E will be in equilibrium with the radium D. Therefore this number is also the disintegration rate of the radium E. The use of a minimum of 6 mg/cm^2 of total absorber between the source and the counter will insure that there is no contribution from either low-energy radium-D beta rays or from the radium-F alphas which are also present.

The method can be made exact by performing the above measurements using as an "unknown", a source prepared from a *standard solution* of the nuclide. In this case a reference number for the nuclide can be assigned to the radium D+E

$$\text{Ra D+E reference number}=\text{activity of standard source}\times\frac{\text{(c/s) extrapolated (Ra D+E)}}{\text{(c/s) extrapolated (std. source)}}. \qquad (5)$$

These reference numbers may be slightly different for each nuclide. Once more it should be emphasized that the very important considerations of stability of electronic equipment, technique of source measurement, and statistical design are always present, although they are not discussed in this Circular.

In some cases there exist long-lived beta emitters whose beta spectra are nearly identical to those of the short-lived nuclides. Sources prepared from these long-lived beta emitters can be assigned reference numbers as in eq (5), with the simplification that the absorption curves in aluminum are parallel, i. e., the ratio at only one thickness of absorber is necessary. Aten (1954), using end-window G–M counters, has found that the absorption of sodium-22 positrons is the same as that of iodine-131 electrons up to 90 mg/cm^2 of aluminum absorber, and that thulium-170 electrons are absorbed in the same manner as gold-198 electrons between 20 mg/cm^2 and 100 mg/cm^2 of aluminum absorber. At the present time there is a sufficient range of energies of beta-ray standards available for use as substitute standards, so that practically any beta emitter can be standardized to ±10 percent routinely in the laboratory. Some representative transmission curves in aluminum, using end-window geometry are shown in figures 38 and 39. Mixtures of fission-product nuclides can be assayed, using thallium-204 as a substitute standard (Reynolds and Brooksbank, 1953; Whalen, Reynolds, and Brooksbank, 1954).

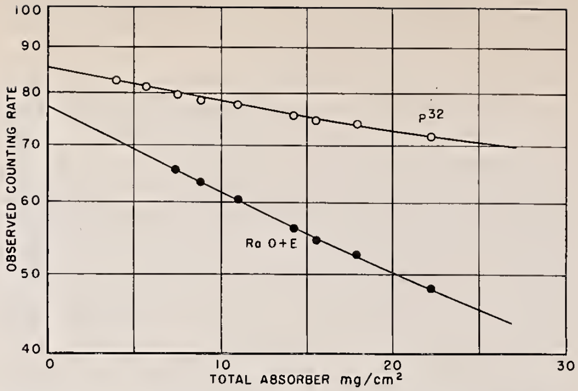

Figure 36. *Approximate method for determining the disintegration rate of a phosphorus-32 sample by comparison with an NBS radium D+E reference source.*

Sources are mounted on palladium-faced silver disks and absorption curves in aluminum are measured using an end-window Geiger counter. The window thickness can range from 1.4 to 4 mg/cm^2. The geometry is shown in figures 38 and 39. Since the beta-ray energies are different, it is necessary to extrapolate back to zero *total* absorber. There is a slight curvature below about 20 mg/cm^2.

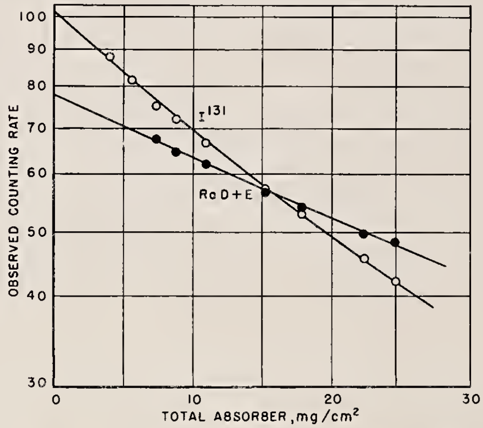

Figure 37. *Approximate method for determining the disintegration rate of an iodine-131 sample by comparison with an NBS radium D+E reference source.*

Sources are mounted on palladium-faced silver disks and absorption curves in aluminum are measured using an end-window Geiger counter. The window thickness can range from 1.4 to 4 mg/cm^2. The geometry is shown in figures 38 and 39. Since the beta-ray energies are different, it is necessary to extrapolate back to zero *total* absorber. There is a slight curvature below about 20 mg/cm^2.

There are several simulated gamma-ray standards available, the most notable of which is the "mock iodine-131" standard developed by Brucer (1955; see also Brucer, Oddie, and Eldridge, 1956), In this case an empirically determined mixture of barium-133 and cesium-137–barium-137 produces, with proper filtration, a gamma-ray spec-

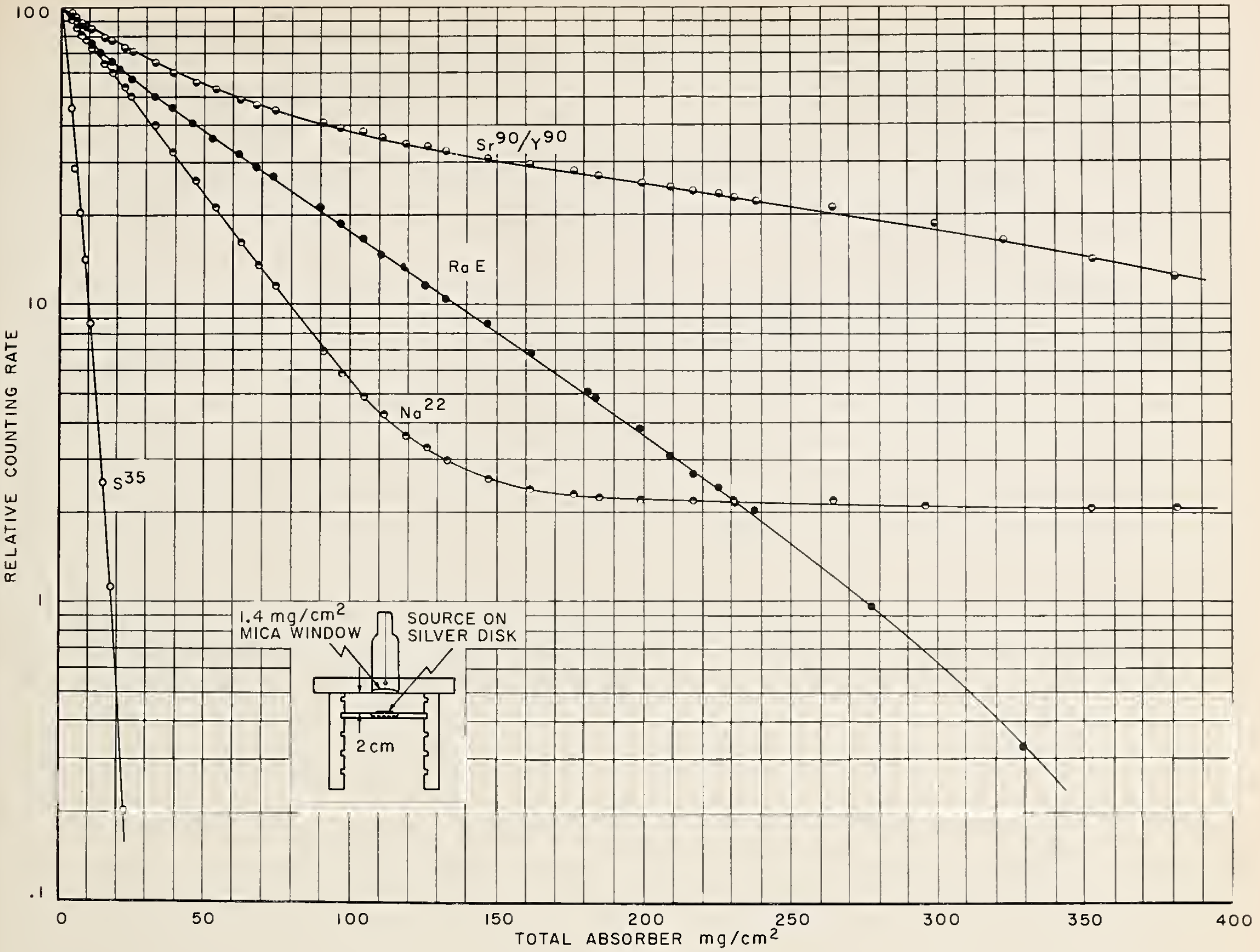

Figure 38. *Transmission of beta rays of the radionuclides sodium-22, sulfur-35, strontium-90/yttrium-90, and radium E through aluminum.*

The geometry is shown in the figure. These curves can be used as qualitative checks on the radioactive purity of a sample. The shapes of the curves are very similiar. There is a slight concave-upwards portion at the beginning, a straight-line (exponential) portion over most of the range and then a tailing off into a gamma-ray portion as in the case of sodium-22 or to negligible transmission for pure beta emitters. All sources used were mounted on palladium-faced silver disks. Absorbers were placed as close to the counter as practicable to minimize self-focusing effects similar to those illustrated in figure 35.

trum almost identical with that of iodine-131. Two sets of such "standards" have been "standardized" in a $4\pi\gamma$-ionization chamber by direct comparison with iodine-131 previously standardized in the $4\pi\beta$ counter. The long half-lives of the barium-133 and the cesium-137 will permit the simulated standards to be used for approximately 10 years before there is much change in the spectrum due to the differences in decay. Not only is a long-lived simulated standard advantageous as a check on calibrations during the course of an experiment, but the material can be incorporated into actual phantoms and into various organ shapes in order to serve as a reference in, say, iodine-uptake studies in human beings. At the present time such simulated standards of iodine-131 are being used throughout the United States in intercomparisons of iodine-131 uptake measurements, in an effort to make diagnoses more precise and to standardize the method of measurement.

Another example of the use of a substitute standard for gamma rays is that of cobalt-60 whose gamma rays are suitable for the standardization of sodium-22, iron-59, and zinc-65. Since the response of a NaI(Tl) crystal, and of an ionization chamber to some extent, is dependent upon the energy of the gamma rays being detected, it is possible, using previously standardized nuclides having widely separated gamma-ray lines associated with their decay, to determine empirically a smooth curve of response per unit activity versus energy for a given NaI(Tl) scintillation counter (Kahn and Lyon, 1953) or with a given ionization chamber. With this smooth curve it should then be possible to standardize, by interpolation or extrapolation, gamma emitters of practically any energy. This will be of special interest to nuclear spectroscopists and those interested in determining absolute gamma-ray yields from nuclear reactions.

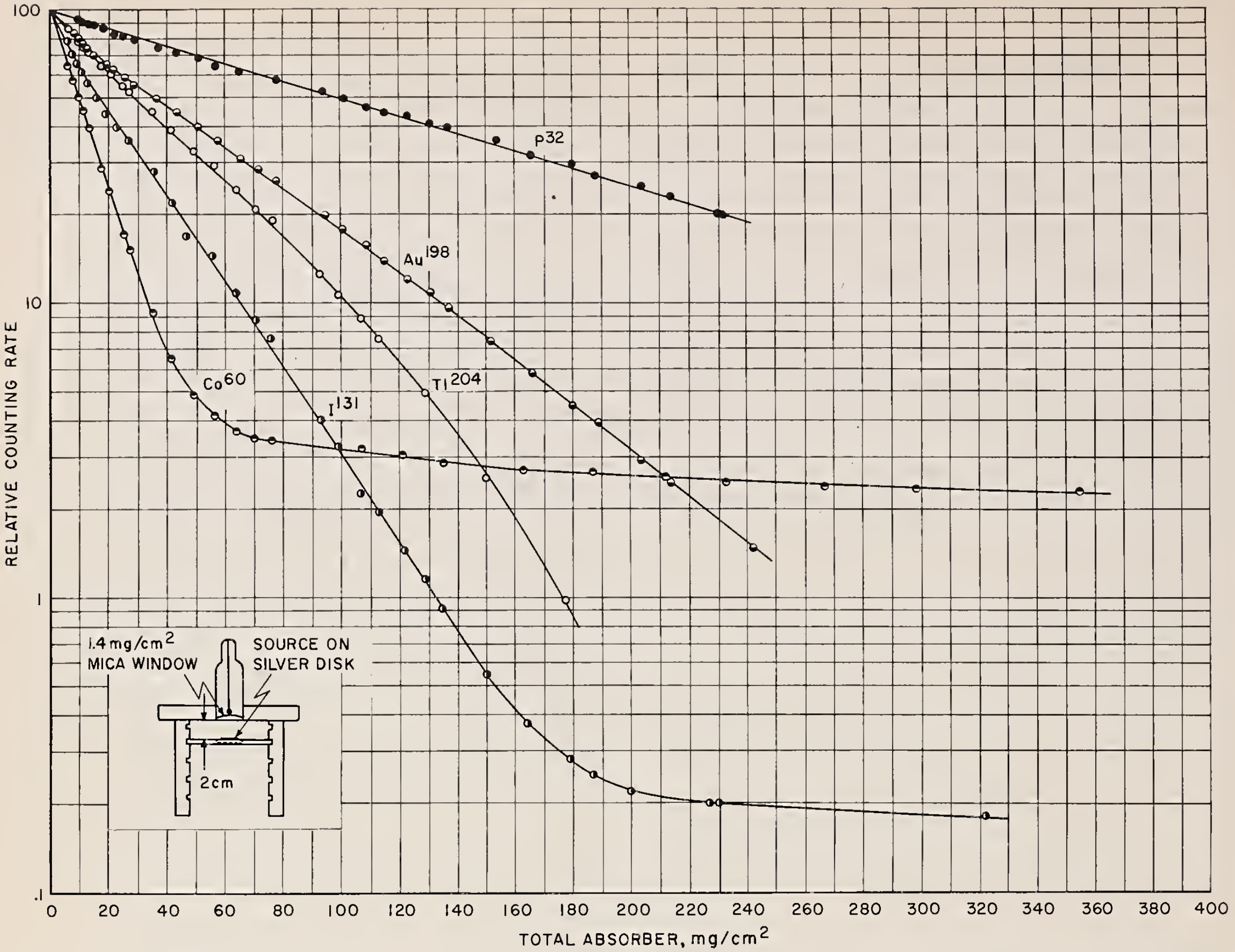

FIGURE 39. *Transmission of beta rays of the radionuclides phosphorus-32, cobalt-60, iodine-131, gold-198, and thallium-204 through aluminum.*

The geomety is shown in the figure. These curves can be used as qualitative checks on the radioactive purity of a sample. The shapes of the curves are very similar. There is a slight concave-upwards portion at the beginning, a straight-line (exponential) portion over most of the range and then a tailing off into a gamma-ray portion as in the case of cobalt-60 and iodine-131 or to negligible transmission for pure beta emitters. All sources used were mounted on palladium-faced silver disks. Absorbers were placed as close to the counter as practicable to minimize self-focusing effects similar to those illustrated in figure 35.

The currents per unit activity as functions of energy are given in figures 40 and 41 for the National Bureau of Standards $2\pi\beta$- and $4\pi\gamma$-ionization chambers, respectively. This technique, in principle, can be applied to other types of detectors.

5.4. Complex Standardization Procedures

There arise many cases in which standardizations are to be made of mixtures of radionuclides. Fission products, for which thallium-204 serves as a substitute standard (Reynolds and Brooksbank, 1953; Whalen, Reynolds, and Brooksbank, 1954) are an example of a mixture being standardized by means of a single average standard. In other cases, mainly in medical and biological research, the *simultaneous use* of two or more different tracers is of great assistance to the researcher. In this way each tracer nuclide, whose effects are to be observed separately, will have had the same host under exactly the same experimental conditions. Double-labeling experiments involving hydrogen-3 and carbon-14 make use of the difference in the beta spectra of these two nuclides. The method of measurement involves (a) incorporating the samples in a liquid-scintillator solution and (b) a two-channel pulse-height analysis of the observed pulses. The relative positions and widths of the two windows of the two-channel pulse-height analyser are adjusted, using standard solutions of the nuclides, so that most of the hydrogen-3 counts occur in the lower window and most of the carbon-14 counts occur in the upper window. With the hydrogen-3 and carbon-14 standard solutions the channels and the response can there-

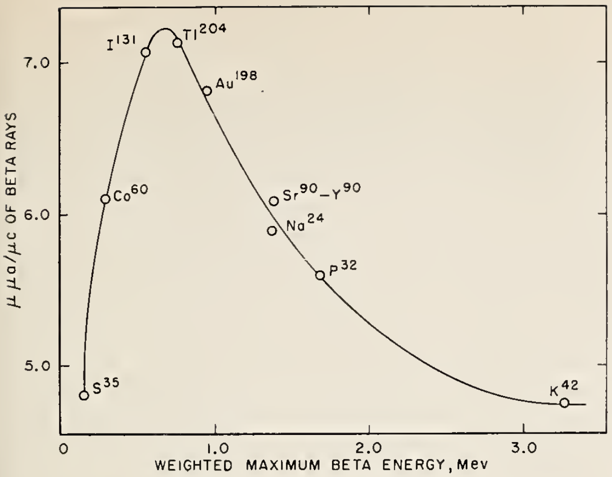

FIGURE 40. *Curve showing the energy response of the NBS $2\pi\beta$-ionization chamber.*

The current per unit activity has been plotted as a function of the maximum beta-ray energy. Where there is beta-ray branching this energy value is the weighted average of the energies of the branches. The response curve appears to be a smooth curve on both sides of a peak at a maximum energy of approximately 0.7 Mev. The existence of a peak and the anomolous decrease above $E_{max}=0.7$ Mev are due solely to the finite dimensions of the chamber. The high-energy beta rays of potassium-42 with their consequently smaller specific ionization will strike the walls and be absorbed before producing as many ion pairs as the lower-energy beta rays of gold-198. Most of the sulfur-35 beta rays are absorbed in the gas of the $2\pi\beta$-ionization chamber. However, these are quite low in energy and thus produce relatively few ion pairs.

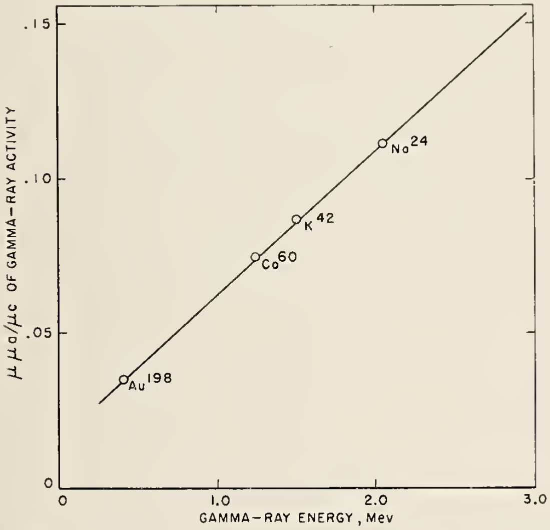

FIGURE 41. *Curve showing the energy response of the NBS $4\pi\gamma$-ionization chamber.*

The current per-unit activity has been plotted as a function of the gamma-ray energy. Where two gamma rays appear in cascade, the energy values have been averaged. Since most of the ionization current measured is the result of Compton scattering, one would expect the energy dependence shown. It should therefore be possible, knowing the decay scheme of a particular radionuclide, to interpolate and to standardize the disintegration rate of a sample of the nuclide by measurement of the current only.

fore be calibrated and the overlap reduced to a minimum. A description of the simultaneous use of sodium-24 and potassium-42 as tracers is given by Tait and Williams (1952). One method of separating the components of a mixture of beta emitters is the graphical analysis of beta-absorption curves (Harley and Hallden, 1955). Harley and Hallden (1955) give a table of the semi-logarithmic slopes of a large number of beta emitters relative to phosphorus-32 and calcium-45.

A number of nuclides emit several gamma rays with different energies. In these cases the various gamma rays can be separated, using the well-type scintillation counter with pulse-height analysis. Similarly, mixtures of gamma-emitting nuclides can be separated by counting under the photopeaks corresponding to the most suitable gamma rays. By means of an efficiency curve obtained with gamma-ray standards the components of the mixture can be analyzed quantitatively. The simultaneous use of Cr^{51}-Fe^{59} and Na^{24}-K^{42} samples is described by Hine, Burrows, Apt, Poelycave, Ross, and Sarkes (1955). A similar application of the energy dependence of the light output of the NaI(Tl) crystal was made by Upson, Connally, and Leboeuf (1955) in counting plutonium-239 gamma rays in the presence of fission-product gamma rays.

5.5. Bremsstrahlung

In addition to its versatility and high efficiency for nuclear gamma rays, the well-type scintillation counter can be used to compare beta sources by measurement of the internal and external bremsstrahlung. Loevinger and Feitelberg (1955) have recently described a method of counting phosphorus-32 routinely by means of the bremsstrahlung from solutions of this nuclide in a well-type NaI(Tl) crystal. The efficiency is given as 0.8 count per 100 disintegrations. For a beta emitter an approximate expression for the energy radiated as bremsstrahlung (Wyard, 1955) measured in million electron volts per beta ray, is

$$B=1.23\times10^{-4}(\overline{Z}+3)E_\beta^2 \tag{6}$$

when $\overline{Z}$ is the effective atomic number of the material in which the beta particles are brought to rest and E_β is the maximum energy of beta disintegration. As a rule of thumb, 85 percent of the total intensity of bremsstrahlung from a beta emitter is emitted in the range 0 to 0.3 E_β, in the ratio 45:25:15 for 0 to 0.1 E_β, 0.1 to 0.2 E_β, and 0.2 to 0.3 E_β, respectively. From eq (6) the energy lost per disintegration from sulfur-35 (E_β=166 kev) would be one-hundredth that of phosphorus-32. In addition, 45 percent of the bremsstrahlung would have energies less than 16 kev, and would therefore be highly absorbed in the housing of the scintillation crystal. The technique should therefore be used for the higher-energy beta emitters. The only disadvantage of the method of measurement is its extreme sensitivity to gamma-emitting impurities, although this can be turned into an asset in the determination of radioactive purity.

6. Determination of Absolute Efficiencies of Detectors

A number of examples of the use of beta- and gamma-ray standards for the determination of the absolute efficiencies of detectors has been reported. Smith, Seliger, and Steyn (1956) have used standardized solutions of phosphorus-32, cobalt-60, strontium-90—yttrium-90, iodine-131, and thallium-204 to determine the absolute efficiency of the 4π-crystal-scintillation-counting technique for beta rays of varying end-point energies.

Gamma-ray standards have been applied to the absolute calibration of experimental equipment used to investigate Coulomb excitation of nuclei. Heydenberg and Temmer (1954), Temmer and Heydenberg (1955), and Fagg, Wolicki, Bondelid, Dunning, and Snyder (1955) have made use of the gamma rays from standards of sodium-22, iodine-131, cerium-141, and gold-198 in order to calibrate their equipment and so determine absolute gamma-ray yields. In the Coulomb-excitation experiment a very thin layer of the target material is laid on top of a Na I(Tl) crystal and is bombarded with alpha particles or protons. The gamma rays emitted from any low-lying excited levels are then detected by means of a scintillation spectrometer. To determine the absolute efficiency, a standardized gamma-ray source of the same size as the beam cross section is delivered to the target material. The ratio of source strength to area under the photopeak then yields one point of an absolute photopeak efficiency curve. A similar application has been made by Swann and Metzger (1955) for the absolute calibration of their gamma-ray detectors. These were used in investigating the cross sections for the production of isomeric states excited in nuclei by inelastic scattering of monoenergetic neutrons. Heft and Libby (1955) have measured absolute cross sections for deuterons on beryllium from absolute measurements of the disintegration rate of the tritium produced in the reaction.

7. Determination of Long Half-Lives

In determining the half-life of a radioactive nuclide by counting measurements for several half-lives, the familiar relationship

$$N=N_0e^{-\lambda t} \tag{7}$$

is reduced to

$$\ln\ \lambda N=\ln\ \lambda N_0-\lambda t \tag{8}$$

and the disintegration constant λ is found from the slope of a linear plot, whence the half-life T is given by

$$T=\frac{\ln 2}{\lambda}. \tag{9}$$

When it is impracticable to measure for a sufficiently long period, the disintegration constant can be obtained by using the more fundamental decay law

$$dN/dt=-\lambda N. \tag{10}$$

If the half-life is so long that the disintegration rate dN/dt of a sample does not change appreciably during a measurement, λ can be deduced from a knowledge of N, the number of nuclei present at the beginning (or end) of the experiment, and of dN/dt.

For isotopically pure elements such as radium the determination of N can be made by gravimetric means. For isotopically pure elements that can be converted to the gas phase a measurement of pressure and temperature in a defined volume will yield N. In most cases, however, due to the presence of other isotopes of the same element, isotopic analysis by means of the gas-density balance or mass spectrometer is necessary.

The determination of N in the measurement of the half-life of carbon-14 is an example of these latter procedures. The carbon-14 half-life is of particular importance in archaeology for the accurate dating of organic relics. In principle the procedure for the determination of the half-life is as follows:

A sample of, say, carbon-14-tagged carbonate is converted to carbon dioxide. A small but accurately-known fraction of this gas is mixed thoroughly with an appropriate counter gas at a definite pressure, and the sample is counted with nearly 100-percent efficiency. Appropriate end and wall corrections are made as discussed previously. The sensitive volume of the counter together with the pressure and temperature give the total number of molecules of carbon dioxide within the sensitive volume of the counter but do not differentiate among carbon-12, carbon-13, and carbon-14. The isotopic abundance ratio of the carbon-dioxide gas must also be determined in the mass spectrometer or in the gas density balance. Since the quantitative count in the gas counters is due only to carbon-14 and the isotopic abundance ratio yields $C^{14}/(C^{12}+C^{13}+C^{14})$ we have dN/dt (C^{14}) and $N(C^{14})$ as required by eq (10).

At the present time the reported values of the carbon-14 half-life range from 5,360 years (Manov and Curtiss, 1951) to 6,360 years (Hawkings, Hunter, Mann, and Stevens, 1949).

In an analogous manner to the carbon-14 half-life determination, Jones (1951) has used a disintegration-rate standard of tritium to determine the half-life of tritium. In this case the isotopic abundance ratio measurement can be quite accurate because of the large relative differences in the atomic weights of the hydrogen isotopes. As has already been mentioned in this Circular, Jones later (1955) also determined dN/dt mass spectrometrically by assaying the helium-3 collected in a given time.

A determination of the half-life of potassium-40 has recently been made by Suttle and Libby (1957). In this case the gamma rays from a standardized sample of cobalt-60 were used to determine the absolute gamma efficiency of the experimental arrangement, a known amount of cobalt being homogeneously incorporated into the sample. A small correction was necessary for the differences in gamma-ray efficiencies of their counters between the 1.17- and 1.33-Mev gamma rays of cobalt-60 and the 1.46-Mev gamma rays from argon-40.

8. Historical and Geological Dating From Half-Lives

The constancy of the disintegration constant of any group of nuclei, independent of external physical or chemical environment, forms the basic assumption of all age determinations based on radioactivity decay. From investigations of pleochroic haloes in mica it has been shown that the ranges of alpha particles from a number of natural radioactive species have always remained constant. Since it has also been shown, both empirically (Geiger and Nuttall, 1911) and theoretically (Gamow, 1928; Gurney and Condon, 1928), that the decay constant is uniquely related to the energy and therefore to the range of the alpha particles ($\log \lambda = a + bE$), it follows that the decay constant has not changed. In particular, it is 1.54×10^{-10} year^{-1} for uranium-238 corresponding to a half-life of 4.49×10^{9} years.

With this constancy definitely established, the half-life becomes a unit of time measurement. For example, in the naturally occurring uranium series there are eight successive alpha transformations from uranium-238 to lead-206. Each alpha particle at the end of its range becomes a helium atom, and since 1 cm^3 of helium contains 2.687×10^{19} atoms, it can readily be shown that 1 g of uranium-238 in secular equilibrium with all daughter products produces 1.16×10^{-7} cm^3 of helium per year.

Assuming no loss of daughter products, the age of the rock is given by

$$8.6 \times \frac{\text{Volume of He}^4 \text{ in cm}^3}{\text{Mass of U}^{238} \text{ in grams}} \text{ million years.}$$

Alternatively, the age can be determined from the Pb^{206}/U^{238} ratio, the Pb^{207}/Th^{232} ratio, or the Pb^{206}/Pb^{204} ratio (Nier, 1939). In each of these cases no loss of daughter products must have occurred between the time of formation of the rock and the time of measurement. A review of this field and a fairly complete reference list can be found in articles by Burling (1952) and Kulp, Broecker, and Eckelmann (1953). A more recent summary of the field and of the use of separated isotopes for the determination of geologic ages by means of the K^{40}-A^{40} and the Rb^{87}-Sr^{87} ratios has been given by Aldrich (1956).

Near the other end of the dating scale is the carbon-14 method developed by Libby (1955). In this case the time unit is approximately 5,600 years instead of 4.49×10^{9} years. There is an important further qualification to the fundamental assumptions of constancy of decay, however. In the naturally radioactive series the material is incorporated into the rock and the quantitative amount of decay is measured. Carbon-14 on the other hand is a relatively short-lived nuclide produced by cosmic-ray bombardment in the reaction $N^{14}(n,p)C^{14}$. Only under the further assumption that the intensity of cosmic rays has been constant throughout the dating period is the carbon-14 dating method applicable. Living organic matter exchanges carbon dioxide with the atmosphere which is in equilibrium with the $C^{14}O_2$ produced in the atmosphere by cosmic rays. When the organism dies, the $C^{14}O_2$ already present is trapped, and loss of carbon-14 by decay is no longer compensated for by exchange of carbon dioxide with the atmosphere. The ratio of the amount of carbon-14 per gram present in any dead organic matter to the amount of carbon-14 per gram present today in living organic matter, together with the knowledge of the half-life of carbon-14, permits the determination of the "age" of the specimen. The sensitivity of the carbon-14 method has been extended to 40,000 years by means of large-volume liquid-scintillator counting (Arnold, 1954), where the carbon is incorporated into the liquid scintillator, and by high-pressure gas-phase counting of acetylene (Suess, 1954) and of carbon dioxide (Fergusson, 1955).

The limitations of available isotopes with appropriate half-lives and in proper chemical composition leaves a wide gap in the dating scale between 40 thousand and 40 million years ago. In this intermediate range, ages have been determined from both thorium 230 (half-life, 83,000 years) and chlorine-36 (half-life 4.4×10^{5} years) measurements, the former for deep-sea sediments and the latter for relatively young surface rocks less than 1 million years old. The thorium-230 method makes use of the thorium-uranium ratio, assuming that the rate of adsorption of uranium by the sediment has been constant. The chlorine-36 in surface rocks is built up by the $Cl^{35}(n\gamma)\ Cl^{36}$ reaction due to slow neutrons from cosmic-ray showers. Both of these methods suffer from difficulties due to leaching and erosion.

9. Determination of Mean Energies and Energy Per Ion Pair

When the beta disintegration-rate standardization is combined with a measurement of the total energy-emission rate, one obtains the average energy $\overline{E}$ defined by

$$\overline{E}=\frac{\int_0^{E_{\max}} N(E)E\,dE}{\int_0^{E_{\max}} N(E)\,dE}, \tag{11}$$

where $N(E)\,dE$ is the number of beta particles emitted in unit time having energies between E and $E+dE$. The only direct experimental method for the determination of the quantity

$$\int_0^{E_{\max}} N(E)E\,dE$$

is by calorimetry. The quantity $\int_0^{E_{\max}} N(E)\,dE$ is the disintegration rate of the source.

Generally it is easier to measure the disintegration rate of a source than to measure directly the total energy-emission rate. Thus for biological applications where one is interested in energy delivered to a particular volume, the quantity $\lambda N\overline{E}$ is of interest, $\overline{E}$ having been determined from eq (11).

In actual practice, calorimetric measurements of the rate of energy absorption in any medium are difficult to make. However, with a knowledge of the stopping-power ratio for the medium in question to air, use can be made of the ionization produced in air by the radiation, which is related to the rate of energy absorption in air by the ratio W_{air}, the mean energy in electron volts expended in the production of an ion pair in air. Since the roentgen, in X- and gamma-ray dosage, is defined in terms of ionization in air, and since most dosimeters, with the exception of the chemical dosimeters, make use of this ionizing property, it has been of considerable interest to obtain accurate values of W_{air} for electrons. Experimentally one obtains

$$W_{\mathrm{air}}=\frac{\int_0^{E_{\max}} N(E)E\,dE}{J_{\mathrm{air}}}, \tag{12}$$

where J_{air} is the total current of ion pairs produced by the source and $\int_0^{E_{\max}} N(E)EdE$ is determined with a calorimeter. W_{air} for sulfur-35 beta rays has recently been determined using eq (12). The rate of energy emission of an approximately 400-mc source was measured in a Peltier microcalorimeter (Mann, 1954b) and the total ionization current from specially prepared sources was measured in a large cylindrical ionization chamber (Bay, Mann, Seliger, and Wyckoff, 1957). The value for W_{air} 33.7 ±0.3 ev per ion pair agrees quite well with the value of 33.6 ev per ion pair, based on measurements made in an extrapolation ionization chamber by Gross, Wingate, and Failla (1957) and the most recent value of 33.9 ev per ion pair reported by Jesse and Sadaukis (1957). A review of the entire field of the measurement of W for other gases as well as for air and also for alpha particles has been given by Binks (1954).

Standards of radioactivity form an essential link between the measurement and the interpretation of physical and chemical phenomena in both the biological and the physical sciences. Their importance will undoubtedly grow as the increasing availability of radioactive materials makes their use more widespread.

10. International Comparisons of Radioactivity Standards

Within the last 5 years a great many intercomparisons of radioactivity standards have been carried out between the standardizing laboratories of Canada, the United Kingdom, and the United States (Astin, Bullard, and Lewis, 1956a and 1956b). The results that are available of these intercomparisons are summarized as follows:

10.1. Radium-226

In the summer of 1952 one of the two United States Hönigschmid radium standards (No. 5440) was taken to the National Physical Laboratory and compared with the British Hönigschmid radium standard (No. 5432) (Perry, 1955). In February 1954 the British Hönigschmid standard (No. 5432) was brought to the National Bureau of Standards and compared with the United States Hönigschmid standards (Nos. 5437 and 5440) (Davenport, McCraven, Mann, and Smith, 1954; Connor and Youden, 1954). In November and December 1955 the Hönigschmid standard of the Federal Republic of Germany (No. 5426) and the Canadian Hönigschmid standard (No. 5425) (Garrett and Geiger, 1956) were brought to the National Bureau of Standards for comparison with the United States standards (Loftus, Mann, Paolella, Stockmann, and Youden, 1957) (fig. 42); and between these last two measurements the Federal Republic of Germany standard was compared at the National Research Council's laboratories in Ottawa with the Canadian standard. The measurements of these standards carried out at the National Bureau of Standards using the electroscope and calorimeter agreed to within 0.3 percent with the results of Hönig-

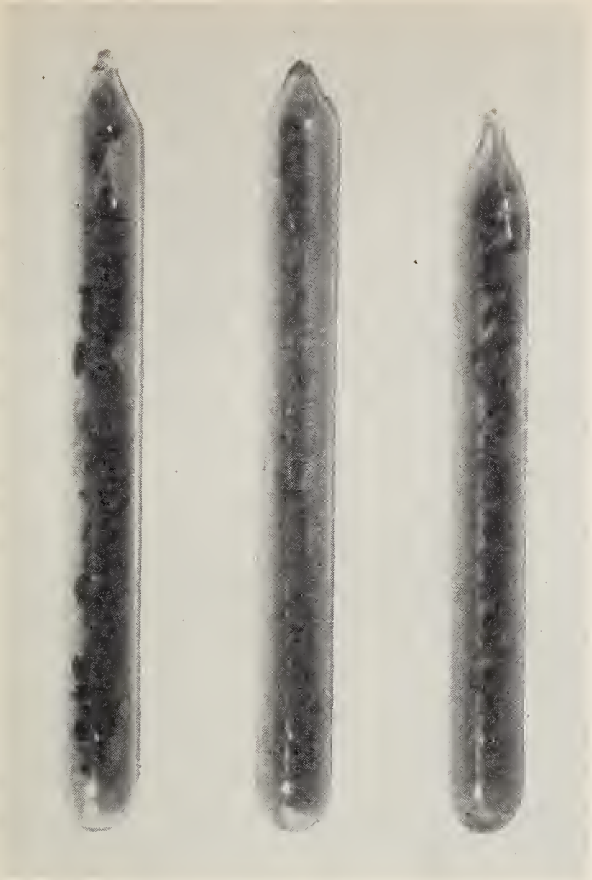

Figure 42. *Three primary national radium standards that were prepared by Hönigschmid in 1934.*

The standards are, from left to right, United States Primary Radium Standards numbers XIV (Hönigschmid No. 5437) and XV (Hönigschmid No. 5440) and the Primary Radium Standard of the Federal Republic of Germany (Hönigschmid No. 5426). The standards are shown with the grains of radium chloride distributed along the lengths of the glass vials as they would normally be arranged for measurement by the National Bureau of Standards standard electroscope. ■

schmid's weighings. By combining all three determinations at the National Bureau of Standards with Hönigschmid's own weighings, "best estimates" of the masses of radium have been made (Loftus, Mann, Paolella, Stockmann, and Youden, 1957).

In the autumn of 1952 the United States Hönigschmid radium standard (No. 5440) was taken to Ottawa for comparison, by the method of gamma-ray ionization measurement, at the National Research Council's laboratories with the old Canadian National Radium Standard (XIII), which was not one of the original Hönigschmid standards (Michel and Whyte, 1955). As the Hönigschmid standards are loosely packed and the old Canadian standard tightly packed, it was necessary to calculate absorption corrections for each of the two standards. In the description of this intercomparison by Michel and Whyte (1955) the results of all previous measurements of the Canadian standard are quoted and the mean given for all values determined by the gamma-ray ionization method. None of the individual ionization values differs from this mean by more than ±0.1 percent, but the calorimetric determination at the National Bureau of Standards is noted to have given a value for the radium content that was 0.4-percent higher. Subsequently, in 1957, the United States standard No. 5440 was compared with the new Canadian standard in Ottawa (Geiger, 1957).

A number of very precise comparisons of Hönigschmid standards, including the new Canadian standard (No. 5425), with the 1911 Paris (Marie Curie) and Vienna standards and with the new international standard (Hönigschmid No. 5430) has also recently been carried out at Brussels (Kipfer, 1956) by the balanced-ionization-chamber method. The precision of the results obtained, of a few parts in 10,000, takes one, however, at least one order of magnitude beyond the uncertainties involved in the self-absorption of the sources themselves.

10.2. Hydrogen-3

Hydrogen-3 standards, in the form of tritiated water, were distributed to a number of laboratories for intercomparison by Atomic Energy of Canada Limited in 1953. At the National Bureau of Standards the Canadian and United States standards were compared by the method of liquid scintillation counting and found to agree to within 1.3 percent. The Canadian standard had been calibrated by the method of gas counting (Hawkings and Merritt, 1954) and that of the United States calorimetrically.

10.3. Carbon-14

Solution standards of this nuclide have been distributed internationally, but the general situation leaves much to be desired. The appropriate method of primary standardization would appear to be that of gas counting, but the disagreements obtained can be matched only with those still extant for its half-life; to which latter disagreement, however, the contributions of mass spectrometry would appear to be not wholly trivial. At the National Bureau of Standards, new and carefully designed compensated gas counters of copper and of stainless steel (figs. 15, 16, 17, and 18) have been constructed with a view to recalibrating its carbon-14 standards (Mann, 1958).

10.4. Sodium-22

Standard samples of sodium-22 have been distributed on an international basis, but to date no intercomparison results are available.

10.5. Sodium-24

Since the inception of the half-yearly distribution of standard solutions of this nuclide by the National Bureau of Standards in the autumn of 1954, samples have been sent regularly to Canada and the United Kingdom for intercomparison. In spite of its short half-life of only 15 hours, comparisons have been carried out by the methods of $4\pi\beta$ counting and $\beta\gamma$-coincidence counting, with an over-all agreement of ±2 percent from the mean value for each distribution. The cooperation of the United Kingdom Supply and Treasury Delegation and Scientific Mission in getting these solution standards to the National Physical Laboratory on the day after dispatch from Washington, D. C., has been an essential factor in the success of these intercomparisons. Normally, zero dates for measurement are set at definite times on the second, third, and fourth days after dispatch from Washington. This is the shortest-lived nuclide used for international comparisons, and it is doubtful whether standards of much shorter-lived nuclides could be conveniently prepared for international or domestic distribution. For individual or special requirements, however, standardizations could probably be carried out with nuclides having half-lives of an order of magnitude less than that of sodium-24, but such standardizations would probabably call for simultaneous measurements a day or so after despatch of a sample that is some 10^7 or 10^8 times stronger than is ultimately required for measurement.

10.6. Phosphorus-32

Semiannual intercomparisons of this nuclide with Canada and the United Kingdom have been carried out since February 1950 (Seliger and Schwebel, 1954). Most measurements have been by the method of $4\pi\beta$ counting (in the proportional or Geiger regions), and over-all agreement appears to be settling down to about ±1 percent from the mean value for each distribution.

10.7. Cobalt-60

Early in 1952, solution standards of this nuclide were made available for international-comparison measurements by the National Bureau of Standards. The results obtained by Canada, the United Kingdom, and the United States by $\beta\gamma$- and $\gamma\gamma$-coincidence counting agreed to within ±1 percent from the mean value (Seliger and Schwebel, 1954). A somewhat larger spread arose, however, in the results obtained by $4\pi\beta$ counting (both proportional and Geiger), and it was subsequently shown (Seliger and Schwebel, 1954) that source self-absorption could cause a decrease in the observed counting rate by as much as 10 percent. By taking the most extreme precautions in source preparation and precipitating the cobalt from solution in the 0.05-ml drop that was placed on the formvar-polystyrene film by means of ammonia, the $4\pi\beta$-proportional counter gave results that were in agreement with those obtained by coincidence counting (Seliger and Schwebel, 1954). The National Physical Laboratory has also shown, by $4\pi\beta\gamma$-coincidence counting, that the losses due to source self-absorption in the $4\pi\beta$ counting of cobalt-60 can be of the order of 5 percent (Perry, 1953). For beta counting, therefore, the solids in solution must be kept low and a minimum of carrier used. It would appear that $4\pi\beta$ counting is not generally to be recommended for radioactive nuclides with beta spectra having maximum energies below 300 kev.

10.8. Zinc-65

A standard solution of this electron-capturing nuclide was sent by the National Bureau of Standards to the Atomic Energy Research Establishment, Harwell, in 1957 for comparison, and agreement was obtained to within 1.5 percent.

10.9. Strontium-90 in Equilibrium With Yttrium-90

Standard solutions of this nuclide were distributed by the Atomic Energy Research Establishment, Harwell, in 1953. By means of $4\pi\beta$ counting, over-all agreement within the limits of ±2 percent from the mean value was obtained (Seliger and Schwebel, 1954). It was shown, however, at the National Bureau of Standards that in sources prepared for $4\pi\beta$-proportional counting the source self-absorption for evaporated sources could be as much as 6-percent higher than that for precipitated sources of this nuclide. As the maximum energy of the yttrium-90 beta rays is 2.2 Mev, this is surprising, and emphasizes the precautions that must be taken in source preparation.

10.10. Iodine-131

Semiannual intercomparisons of this nuclide have been made with Canada, or the United Kingdom, or both, ever since December 1949, using the standards prepared for the regular distributions of the National Bureau of Standards (Seliger and Schwebel, 1954). Agreement has been obtained in the determinations of the basic counting rates to within ±1 percent from the mean value for each distribution. The National Bureau of Standards has, however, made a calculation for the source self-absorption based on shadow electron micrographs of silver-iodide source deposits (Seliger and Schwebel, 1954) (fig. 14). These calculations give a source self-absorption of about 2 percent, and accordingly all National Bureau of Standards calibrations by $4\pi\beta$-proportional counting are corrected by this amount for source self-absorption. The standardized activity is thus 2-percent higher than that determined by $4\pi\beta$-proportional counting.

10.11. Gold-198

The first intercomparison of gold-198 was carried out in May 1953 on samples distributed to Canada and the United States by the Atomic Energy Research Establishment, Harwell. Over-all agreement was not better than ±4 percent from the mean value, the calibrations being carried out by $4\pi\beta$-proportional and $4\pi\beta$-Geiger counting, $\beta\gamma$-coincidence counting, γ-reference chamber, and β-reference chamber. Subsequently, this agreement has been improved to about ±2 percent, using the solution standards made available semiannually by the National Bureau of Standards, but in December 1956 an intercomparison was carried out between Atomic Energy of Canada Limited, the National Physical Laboratory (United Kingdom), the National Physical Research Laboratory (Union of South Africa) and the National Bureau of Standards, which supplied the sources, and results were obtained none of which differed by more than ±1 percent from the mean value.

10.12. Thallium-204

Standard solutions of this nuclide, calibrated by $4\pi\beta$-proportional counting, have been distributed by the National Bureau of Standards for intercomparison. Some difficulties that may possibly be ascribed to adsorption on the glass or to exchange have, however, been experienced, and work is proceeding currently on the subject. These difficulties may arise from the same source as gives rise to the two groups of quite different half-lives for thallium-204 that appear in the literature.

10.13. Summary

It will thus be seen that, of the original nine nuclides that were proposed for intercomparison at the 1951 Harwell conference, satisfactory agreement has been achieved in the case of six. Two others, namely hydrogen-3 and zinc-65, have also been satisfactorily intercompared.

In addition to the three nuclides still outstanding, the difficult field of electron-capturing nuclide standardization remains relatively untouched from an international point of view. Even on a purely national basis, the attempts to produce radioactivity standards of such nuclides have been somewhat sporadic, and it yet remains to achieve the kind of accuracies and reproducibility of results that have been aimed at in international comparisons. The methods available for calibrating electron-capturing nuclides have recently been reviewed by Allen (1957) who has himself used, as described in his review, high-pressure 4π-Geiger-Müller and 4π-proportional counters to measure the rate of emission of X-rays from electron-capturing nuclides. He did not, however, obtain complete absorption of the X-rays. The use of a proportional counter for the assay of such X-ray-emitting nuclides has also been described by Lyon and Reynolds (1957) while Campion and Merritt (1957) have used a 4π-proportional counter operated with argon and methane up to pressures of approximately 7 atmospheres to measure the rate of X-ray emission from manganese-55 in the decay of iron-55.

Another problem that is presently engaging the attention of the National Bureau of Standards is the production of gamma-ray standards kits with a variety of standards to cover, in suitable intervals, the energy range from 0.1 to 1.0 Mev. These standards, which are required for nuclear-physics research, will probably be issued in the form of disks calibrated in terms of the gamma-ray emission in a 2π geometry, from the surface of the disk on which the source is deposited. It is anticipated, from the interest already shown in them, that these standards will also, in the future, be considered for international comparison.

11. International Organization

As has been noted already, the first meeting of the Subcommittee on Radioactivity Standards of the International Commission on Radiological Units was held in Frankfurt-am-Main on 5 and 6 August 1955. At this meeting it was recognized that, in the field of radioactivity standardization, there were three general groupings of the countries that were then, or were in the future likely to be. interested. These groupings were: (1) Those countries that were producing regularly primary or secondary standards of radioactivity and were in a position to supply other countries with such standards; (2) those countries that, while being in a position to produce their own standards of radioactivity, might yet find it more convenient to obtain standards from countries in group (1); and (3) those countries that would find it difficult to produce primary or secondary standards and would prefer to meet their needs by relying wholly on relative measurements using standards supplied by countries in group (1). It was recognized that the National Bureau of Standards (U. S. A.) and the National Physical Laboratory (U. K.) were the only laboratories at present preparing and distributing radioactivity standards in quantity, and it was accordingly agreed that the Subcommittee should prepare a combined catalogue of the radioactivity standards available from these two standardizing organizations. This combined catalogue has now been published as appendix III to the National Bureau of Standards Handbook 62 (Report of the International Commission on Radiological Units and Measurements 1956).

It is eventually hoped that other countries will participate in international comparisons of radioactivity standards, and accordingly the directors of the three standardizing laboratories concerned, in the United States, United Kingdom, and in Canada, have, in the closing paragraph of a joint statement (Astin, Bullard, and Lewis, 1956a and 1956b) extended an invitation to those who are interested in such intercomparisons to write to the most conveniently situated of the three directors.

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WASHINGTON, April 1, 1958.

U. S. GOVERNMENT PRINTING OFFICE: 1958

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