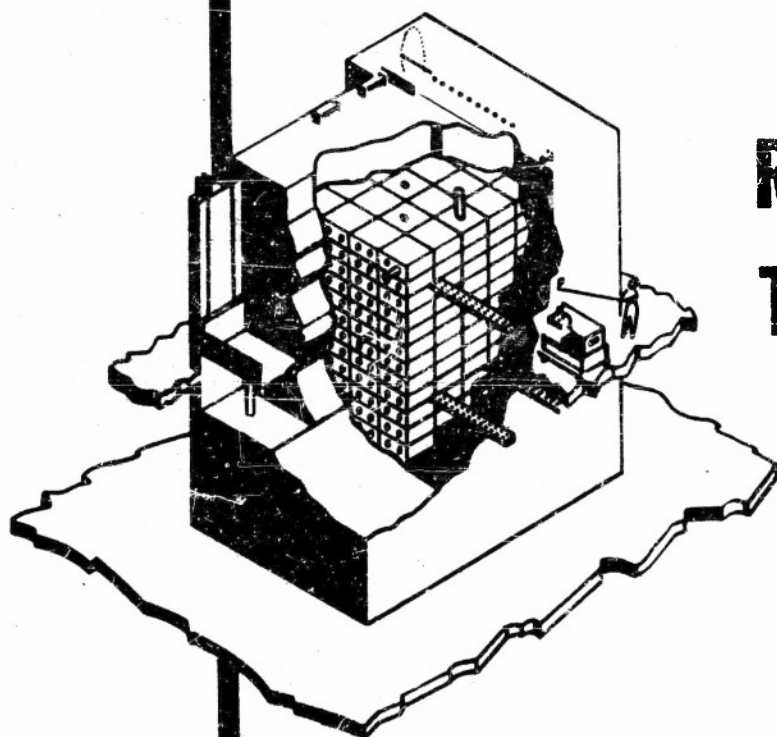


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Status Report on Standardization of Radionuclides in The United States

by
George G. Manov



Division of Physical Sciences

NATIONAL RESEARCH COUNCIL

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STATUS REPORT ON STANDARDIZATION OF RADIONUCLIDES IN THE UNITED STATES

By

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Atomic Energy Commission

Washington, D. C.

PREPARATION SPONSORED BY THE
SUBCOMMITTEE ON BETA AND GAMMA RAY MEASUREMENTS AND STANDARDS

OF THE

COMMITTEE ON NUCLEAR SCIENCE
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I. INTRODUCTION

In the past decade, increasing attention has been paid to the problem of establishing primary standards of radioactivity. The task has been by no means simple, for not only have the required chemical separations been complex, but the degree of radiochemical and/or radioisotopic purity needed¹ has been extremely high. Where chemical methods have been involved in the assay, interference has been found to occur from analogous compounds of the same element (e.g., $\text{Na}_2\text{HP}^{32}\text{O}_4$ and $\text{Na}_2\text{HP}^{32}\text{O}_3$) as well as from different radio-species of the same element (e.g., $\text{Na}_2\text{HP}^{32}\text{O}_4$ and $\text{Na}_2\text{HP}^{33}\text{O}_4$).

Improved sensitivity of instrumentation, particularly in coincidence counting, has led to the discovery of greater complexities in decay schemes previously thought to have been rather simple. Only a few decay schemes, on the other hand, have been simplified. Experimental re-investigation of problems thought to have been satisfactorily solved have disclosed new and unsuspected sources of error. The discovery of a 12-day metastable Xe-131 in Iodine-131 (1) and the discovery of Phosphorus-33 (2) (25-day half-life) in Phosphorus-32 are but two examples.

Over a thousand radioactive species are known to exist, but only a comparatively few satisfy the requirements for primary standards. The required characteristics are: known decay scheme, satisfactory half-life and disintegration energy, radiochemical and radioisotopic purity, and ease of preparation and availability.

Primary standards of radioactivity are, in some ways, analogous to fixed points in the thermometer scale in that each can be used for comparison with "unknowns" not too far removed from the standard, for example, in terms of the type and energy of the radiation.

Whereas it had originally been considered

that a relatively few standards covering the range of beta and gamma energies of interest would suffice, the thought has been recently expressed that standards may be required for many additional radionuclides. For example, discrepancies in measurements using end-window counters may arise because of the difference in the distribution of energies in the beta-ray spectra of two isotopes that have approximately the same maximum beta energy.² Similarly, although a given beta-gamma standard may be suitable for determining quantitatively the activity of a second radionuclide by comparison of their respective beta activities, it may be totally inapplicable, for example, for use in measurements involving comparative gamma-ray counting.

The value of beta and gamma ray standards to research and education needs no elaboration. It is surprising, however, how quickly new information regarding energies and decay schemes of certain radionuclides has been applied to other fields.

The quantity of tetraethyl lead in ethyl gasoline can be determined by a measurement of the percent absorption of the beta radiation from Sr-90 by the lead in solution. In the field of medicine, 28-day Chromium-51 is injected into cancerous human tissue. The dosage delivered to the tissue of the patient is primarily that originating from the soft K-capture electrons, and is measured external to the patient by counting the gamma radiation escaping the tissue. The disintegration of Cr-51 heretofore (3) has been considered to be primarily (97%) by K-capture directly to the ground state and approximately 3% through K-capture followed by a 0.32 mev gamma radiation to the ground state. A small uncertainty in the branching

¹ Attainment of radiochemical purity involves the complete separation of the radionuclide in question from other radioactive species. Radioisotopic purity involves the absence of different species of the same element, e.g., Fe-55 in Fe-59.

² Additional questions have been raised recently as to the theoretical shape of the spectra of soft beta radiation (n_β as a function of E) for isotopes of less than 0.5 mev maximum beta energy. Many laboratories use end-window tubes for standardization purposes, and the correction to zero thickness of total absorber, for example, will depend on the spectral distribution of the beta rays from the two isotopes under comparison.

ratio (e.g., 96% vs 97%) would make approximately 30% difference in the calculated dose delivered to the patient if reliance is placed only on the measurements made of the gamma radiation escaping the patient's body. Recently, it has been found (4) that the gamma branching is approximately 8% rather than 3%.

A general survey of the problems involved in the preparation and certification of primary standards of radioactivity indicates major progress in the determination of the absolute disintegration rate of beta and of beta-gamma emitters. The situation is much improved over the period 1947-49, and the availability of standards within the United States such as C-14, Co-60, I-131, P-32, Ra-226 and Pb-210 (Radium D-E) is one measure of the advances made.

The progress in the acceptance of definitions for radioactivity units and standards has been reported by R. D. Evans (5a,b). The use of the term "rutherford" to denote 10^6 dis/sec was abandoned in 1950.

Radium was formerly unique in that it was

the only radionuclide for which the establishment of radiochemical purity was equivalent to primary standardization. The curie was officially redefined, however, as reported by Evans (5b) and Paneth (6) at the July, 1950, meeting of the Joint Commission on Standards, Units, and Constants of Radioactivity as "the quantity of any radioactive nuclide in which the number of disintegrations per second is 3.700×10^{10} ." This new definition divorces the curie unit from a special association with radon and radium.

Assembly by the National Bureau of Standards (7) of published information on the decay scheme, half-life and energy distribution of many radionuclides has served as a spur for additional research in these fields. Prompt publication of such summaries is now a routine matter (8), and trials are under way to provide some of the information in the form of punched cards.³

³ Private communication from Dr. A. F. Thompson, Chief, Technical Information Service, AEC, March 20, 1963. A similar set of cards will be available shortly from the Radioactive Products, Inc., Detroit, Michigan.

II. INFORMATION NEEDED TO PREPARE AND CERTIFY PRIMARY STANDARDS

All of the common sources of radionuclides supply materials for the preparation of standards: Ra-226 and Pb-210 from natural ores, Na-22 from cyclotron targets, I-131 from fission products and Co-60 from neutron irradiation. In every case, chemical separations, some simple, some complex (9) are necessary to obtain a radiochemically pure product.

When radiochemical and radioisotopic purity are assured and a sufficient supply of the

material is at hand, the properties of the radionuclide are then studied in greater detail. Information generally needed involves a determination of the half-life, the shape of the beta spectrum, the energy of the beta and of the gamma radiation and a detailed decay scheme. Other important or useful information may include a measurement of the number of radioactive atoms per gram of material (determined, for example, by mass-spectrographic analysis), self-absorption coefficients, etc.

III. ADVANCE IN TECHNIQUES OF MEASUREMENTS

A. DETERMINATION OF THE HALF-LIFE OF RADIONUCLIDES

The determination of the half-life of a nuclide by any method at constant, but unknown, efficiency is essentially a straightforward procedure. If R_1 and R_2 are the observed counting rates obtained with a given detector during a time interval, t , the half-life, $T_{1/2}$ can be calculated:

$$T_{1/2} = \frac{0.693t}{2.3 \log (R_1/R_2)} \quad \text{Eq 1)}$$

The method is best adapted to those nuclides for which the initial and final measurements can be obtained in a reasonable length of time.

Half-lives have been measured to four significant figures by use of a differential ionization chamber and a vibrating-reed amplifier with feedback (10, 11). With this device, it has been possible to obtain significant measurements over a period of 12 half-lives with some short-lived radionuclides.

If a nuclide is very long-lived (such as C-14), the half-life can be calculated from the expression

$$T_{1/2} = 0.693 N / (-dN/dt), \quad \text{Eq 2)}$$

where N is the total number of radioactive atoms present (determined by mass-spectrographic analysis and the weight of the sample present) and dN/dt is the measured rate of disintegration.

Calorimetric methods have been used to determine directly the half-life of radionuclides such as P-32 (12).

B. DETERMINATION OF THE RATE OF DISINTEGRATION

Measurements⁴ of the rate of disintegration can be made by (a) coincidence counting (either beta-gamma or gamma-gamma), (b) G-M or other type of counters under conditions of known geometry and (c) calorimetry. Coincidence counting (16, 17, 18, 19, 20) involves

the use of two counters; one for detecting the beta (or gamma) radiation, and the other for detecting the (second) gamma radiation. Certain conditions must be fulfilled and various small corrections to the observed counting rates must be applied. If N_1 and N_2 are the counting rates of the beta (or gamma) and of the gamma counters, respectively, and N_{12} is the coincidence rate, the absolute rate of disintegration $-dN/dt$ of the sample is given by

$$-dN/dt = N_1 N_2 / N_{12} \quad \text{Eq 3)}$$

This method can be applied to the determination of the activity of long-lived as well as moderately short-lived nuclides.

The technique of "known geometry" with end-window counting for example, involves measuring the rate of decay of a sample under conditions in which the counter "sees" a known solid angle at a known distance from the source. In principle the method is simple but corrections must be applied (15) for resolution or dead time losses; for backscattering from the support; for sidescattering from the walls of the shield enclosing the sample, the support and the counter; for absorption by the window of the counter; etc. The method of end-window counting is applicable preferably to beta emitters with moderate or long half-lives. A variation of the method of known geometry is that in which the sample to be measured is completely inside the counter or may even be part of the filling gas. This type of counter has been successfully used in measurements involving C-14 and H-3.

The rate of charge-loss has been measured in isolated systems containing a radionuclide by using a Lindemann electrometer and a compensating condenser (21). The method has been applied to P-32 and Na-22.

Internal counters with 4-pi geometry suggested in 1947 by Borkowski (22) have been refined and used extensively (23, 24a, 24b, 25) for absolute beta measurements. Proportional 4-pi counters have been widely used in Canada (26) for standardization.

⁴ Reference is made to "Measurements of Radioactivity" (13), to "The Geiger-Mueller Counter" (14), to the Conference on Absolute Beta Counting (15) for a more detailed explanation of the methods used.

Panel discussions (15) on the techniques of absolute beta-counting have brought out a number of small, hitherto unsuspected sources of error regarding the effects of backscattering from supports, self-absorption, etc. New measurements have been made of the backscattering of positrons and electrons from various kinds of supporting materials (27, 28).

The method of Marinelli (29) can be used for standardization if the decay scheme has been established. The rate of disintegration of a radionuclide can also be determined (30) if the average energy per disintegration, $\bar{E}$, is known.

Calorimetric methods for the determination of activity involve measurement of the rate of heat evolved, dQ/dt , by absorption of the energy emitted during the decay of the radionuclide. The method involves the usual thermal and other corrections applicable to calorimetry and is used preferably for beta emitters or for other isotopes where the radiation to be measured can be completely absorbed in the calorimeter. From the expression

$$dQ/dt = \bar{E}_\beta dN/dt \text{ ----- (Eq 4)}$$

either $-dN/dt$ or $\bar{E}_\beta$, the average energy per beta disintegration, can be computed if one of them is known.

The availability of intense, high specific-activity sources and the development of suitable calorimeters (12, 31) operating at liquid air temperatures has enabled estimation of the half-lives of isotopes such as C-14.

C. DECAY SCHEMES

A clearer understanding of the techniques involved and of the errors inherent in coincidence counting (16, 17) the method of "delayed coincidence" counting (32), the development (33, 34) of amplifiers with very short resolving times (10^{-9} sec.) and the virtual elimination of spurious coincidences have permitted closer examination of many complex decay schemes.

A beta-beta (conversion electron) coincidence spectrometer has been devised (35) in which a Geiger-Mueller counter is placed at the "exit" end of the spectrometer and a scintillation counter is placed near the source. Coincidences are measured between the two counters corresponding to a selected electron "line" of known energy and all other beta radiation. The arrangement is said to be satisfactory for estab-

lishing decay schemes by beta-beta (conversion electron) coincidences.

Internal conversion coefficients have been measured by gamma-X ray coincidence counting (36, 37) as well as by the double-coil, thin lens magnetic beta-ray spectrometer (38). A revised table of K-shell internal-conversion coefficients is available (39).

D. SPECTROMETRY

Crystal spectrometers (40) have been developed for the determination of gamma-ray energies to four or more significant figures. Scintillation spectrometers and counters for the measurement of the energy distribution and the half-life (1 to 30 seconds) of beta-ray emitters have been described (41). A new type of scintillation spectrometer has been devised (42) in which incident photons are detected in a primary detecting NaI(Tl) crystal; the radiations not completely absorbed in the primary crystal are absorbed in a surrounding cluster of NaI(Tl) crystals which act as an anticoincidence circuit. The arrangement is said to yield the gamma-ray spectrum directly without any interpretation of results which might become laborious, particularly if the incident gamma radiation has a complex spectrum. The apparatus has been used to study Cs-137, Na-22 and other radionuclides.

Pair spectrometers have been constructed (43, 44) in which rotation of the image in a short magnetic lens is used to separate negatrons and positrons by putting the pair converter a little to one side of the optical axis of the lens. It is then possible to obtain coincidence "lines." The apparatus has been used to study the 2.62 mev gamma-ray of ThC" (Tl-208) and the 2.76 mev gamma-ray of Na-24. A recent study has been made of the shapes of beta spectra (45) and tables for the analysis of beta spectra are available (46).

E. MASS ABSORPTION COEFFICIENTS

New measurements have been reported for the gamma-ray absorption coefficients (47, 48, 49, 50) over a range of energies from 0.1 to 6 mev (Co-60, I-131 and others) in absorbers of atomic number from 13 to 82. The calculated values are in good agreement with those measured experimentally. This information is useful in correcting for the attenuation of the gamma radiation passing through the walls of the GM counter or ion chamber.

IV. STATUS OF INDIVIDUAL RADIONUCLIDES ⁵

For purposes of presentation, the status of individual radionuclides is divided into three categories: (a) nuclides for which standards in the United States are available or are in preparation, (b) nuclides for which no standards are available, but considerable information on whose nuclear properties exists, and (c) nuclides for which standards would be desirable but considerably more information of various kinds is needed.

In each category, the information listed includes (when available): the method of production, method of obtaining radio-chemical purity, the half-life, the decay scheme (or schemes), intercomparison of the activity between different laboratories and the availability of standard samples. The nuclides are listed alphabetically within each category.

A. AVAILABLE STANDARDS

Summaries are presented for the following radionuclides: Carbon-14, Cobalt-60, Iodine-131, Phosphorus-32, Radium-226 and Lead-Bismuth-210 (Radium D-E).

1. Carbon-14

Most of the C-14 is now produced by neutron irradiation of beryllium nitride, Be_3N_2 . The C-14 is separated as active CO_2 and precipitated as BaCO_3 . Specific activities of 250 to 1500 mc/gm of BaCO_3 are routinely available (51).

Full agreement has not yet been reached for the value of the half-life of C-14. Because of the long half-life of this radio-nuclide, a direct measurement is not possible, but it can be calculated from the relation expressed in Eq. 2. Values ranging from a high of 7200 to a low of 4700 years have been obtained. Two general methods have been used: (a) end-window counting of solid barium carbonate and (b) counting of gaseous C-14 either as carbon dioxide or as methane in which the radio-active carbon forms a portion of the filling gas of the

counter. Generally speaking, gas-phase counting has proven to be more reliable than counting of solid barium carbonate.

In 1949, Hawkings, Hunter, Mann and Stevens (52) reported a half-life of 6360 ± 200 years. The procedure involved a mass spectrographic analysis of the active CO_2 and the use of a compensated pair of CO_2 - CS_2 filled (53, 54) internal gas counters of varying diameters. The active carbon dioxide for the determination of the isotopic ratio was evolved by heating active BaCO_3 in a quartz tube to 1000 - 1200°C . For the disintegration-rate experiments, the active carbon dioxide was generated by heating with an excess of lead chloride, the dried residue obtained from a solution containing active sodium carbonate and a known amount of inactive Na_2CO_3 . Checks were made to determine the completeness of the evolution of carbon dioxide under these conditions.

Jones (55), using alcohol-argon as a filling gas in an internal G-M counter, obtained 5589 ± 75 years for the half-life of C-14. Libby and co-workers (56, 57) also used alcohol-argon as the filling gas and employed counters of various ratios of length to diameter. The value for the half-life first reported, 5720 ± 20 years (56) was lowered to 5580 ± 45 years (57) after further corrections were made for the end- and the wall-effects of the counters. It was assumed that the counting efficiency was 100%.

In each of the above cases, the investigator worked with his own sample of active carbonate and measured (or had measured for him) the isotopic ratio from which N in Eq. 2 could be calculated, and each performed his own measurements of dN/dt . The percentage of active CO_2 in the barium carbonate ranged from 1.28 to 39.4% C-14.

In 1949 the National Bureau of Standards (NBS) undertook a cooperative venture to determine the half-life of C-14 and to prepare standard samples of C-14 for distribution. A sample of specially purified BaCO_3 treated to remove silica and other inert impurities was

⁵ For the purpose of this report, a cut-off date of April 1, 1953, has been used.

prepared by the Chemistry Division of the Oak Ridge National Laboratory (ORNL), thoroughly mixed to insure homogeneity, and sealed in 3 separate glass ampoules. One of these ampoules was retained by ORNL; the other two were sent to NBS. One of the NBS samples was put away unopened for reference. Details of the NBS measurements have been reported by Manov and Curtiss (58).

The CO_2 from a portion of this active barium carbonate was generated by heating an intimate mixture of the BaCO_3 with PbCl_2 at first to 500°C and then to 1100°C . The evolved CO_2 was mixed thoroughly by repeated condensation with liquid air and evaporation into a small, closed flask and allowed to stand for several hours. Samples of this CO_2 were sent to several cooperating laboratories for mass spectrographic analysis.

An additional portion of the same active barium carbonate was mixed with a known weight of inert barium carbonate, the CO_2 evolved with perchloric acid and absorbed in an excess of sodium-hydroxide—sodium-carbonate solution. The solution was diluted to a total volume of 39.00 ± 0.05 liters. The final solution contained all of the active carbon obtained from 0.1066 ± 0.0001 gm of active barium carbonate in 0.100 M inert Na_2CO_3 with free sodium hydroxide to the extent of approximately 0.001 M. Approximately 1200 flame-sealed ampoules containing 25.00 ± 0.03 ml of this solution were prepared,⁶ and samples were distributed to the participating laboratories for counting.

It is to be noted that the problem of determining the half-life of Carbon-14 had been divided into two independent parts: (a) a collaborative determination of the isotopic ratio from which N in Eq. 2 could be calculated, (b) a collaborative determination of $-dN/dt$. The results were reported in February, 1950 (59a).

Measurements obtained for the isotopic ratio were as follows:

⁶ Experiments by A. Schwebel using an ion chamber and a vibrating-read electrometer demonstrated that 0.1 M sodium carbonate carrier was sufficient to prevent the loss of any significant quantity of active carbonate by adsorption on the walls of the ampoule.

Experiments also showed that the ampoules could be flame-sealed without introducing CO_2 from the torch. For this purpose $\text{Ba}(\text{OH})_2$ was substituted for the $\text{NaOH-Na}_2\text{CO}_3$ solutions and the sealed ampoule examined for the presence of a Tyndall beam. None was found.

Argonne	3.93%
McMaster Univ.	3.74%
NBS	3.88%
Sloane Kettering	3.99%

Average 3.88%

The arithmetical average, 3.88%, happened to agree with the value obtained by the NBS that had an estimated precision of $\pm 0.01\%$ and an estimated accuracy of $\pm 0.04\%$. Because the accuracy of the other measurements could not be estimated satisfactorily, the NBS value and its accuracy were accepted (59a). The abundance of C^{14} in the sample distributed was taken as $3.88 \pm 0.04\%$ C-14.

The following results were obtained for the activity of the sodium carbonate solution:

Chalk River	1330 dis/sec/ml
MIT	1410
NBS	1280

Average 1340 dis/sec/ml

In each case, gas-phase counting was performed with a compensated pair of $\text{CO}_2\text{-CS}_2$ filled counters. It is known (52, 53, 54) that the resolving time of the $\text{CO}_2\text{-CS}_2$ counters is of the order of 1000 microseconds, and at counting rates sufficiently high (50 - 100 counts/second) to obtain results of statistical significance in a reasonable length of time, the resolution-time losses are of the order of several percent.

From the average value of 3.88% for the isotopic ratio and the counting measurements reported by each institution, the following half-lives can be calculated:

Chalk River	5170 Years
MIT	4870
NBS	5370

Average 5140

The counting rates obtained by Chalk River and by MIT were, at the time, provisional values. As an interim value pending further work, it was therefore suggested (59a) that the average isotopic ratio of the active barium carbonate used in the cooperative investigation be considered 3.88%, the activity of the ampoules be tentatively assigned 1280 dis/sec/ml, and the half-life of C-14 be estimated at 5400 years, with an uncertainty of perhaps ± 200 years.

Subsequent to this suggestion, two additional values for the half-life using other samples

TABLE I*

Relative Activity of Nuclides Assayed by Various Workers

Isotope	Harwell	M.R.C. Hammer- smith	Bureau of Standards (U.S.A.)	Oak Ridge (U.S.A.)	M.I.T. (U.S.A.)	Chalk River Canada	N.P.L. Teddington
Co-60	1.00 (a)	1.02 (d)	1.025	---	1.0 (g)	---	1.0 (m)
Na-24	1.00 (a)	1.00 (d)	---	---	---	---	---
P-32	1.00 (b)	0.95 (e)	0.91	0.99 (g)	1.85 (h)	0.91 to 1.12 (k)	---
I-131	1.00 (c)	0.96 to 1.02 (f)	0.98	1.05 (g)	1.04 (h)	0.80 (l)	---

- (a) Measured by beta-gamma coincidences.
 (b) Measured by defined solid angle.
 (c) Defined solid angle. Conversion electrons neglected.
 (d) Gamma-ray ionization (also approximately checked by beta-gamma coincidences).
 (e) Beta-ray ionization.
 (f) Beta-ray ionization and gamma-ray ionization (using breakdown scheme as Figure 3B); Metzger & Deutsch, Phys. Rev. **74**, 1640 (1948).
 (g) Based on first sample supplied to M.R.C. (Iodine figures include a correction that 1.75 times ordered quantity was sent.)
 (h) Calibrated sample sent to M.R.C., September 1947. (Iodine figures corrected for change in M.I.T. measurement 11/2/49.)
 (k) Based on two samples received by Dr. Popjak (M.R.C. Hampstead).
 (l) Mean of four shipments received by Dr. Popjak (M.R.C. Hampstead).
 (m) Exchange with M.R.C., Hammersmith.

*Taken from "Absolute Measurements of the Activity of Beta Emitters,"
 J. L. Putman, J. Brit. Radiology, **23**, 46 (1950).

TABLE II

Results of Intercomparison of Co-60 by Different Laboratories*

	$4\pi\beta$ counter Conducting film	$4\pi\beta$ counter Mirror image	$\beta-\gamma$ coincidence	$\gamma-\gamma$ coincidence
Cobalt-60: United States NBS	0.785 mc./ml.	0.782 mc./ml.	0.787 mc./ml.	(0.790 mc./ml. 0.800
ORNL			0.774	
Canada AECL			0.790	

*Taken from "Refinements in Standardization by 4π Beta Counting"
 W. B. Mann and H. H. Seliger, J. Research, **NBS 50** #4 April (1953)
 RP 2049

TABLE III

Results of Intercomparisons of I^{131} Solution Standards Prepared and Distributed by the National Bureau of Standards*

Values are given in disintegrations per second per milliliter $\times 10^{-5}$ as of 8AM on the zero date

Zero Date	UNITED STATES			CANADA		ENGLAND		Averages
	NBS 4πβ Counter	ORNL β-γ Coincidence	AECL Chalk River 4 π Counter	Hammersmith Absolute β Ion Chamber	Hammersmith 4πGM Counter			
Dec. 13, 1949	250			248		249		
June 8, 1950	299		304	300		301		
Jan. 15, 1951	193		200	196		197		
June 7, 1951	243		254	246	261	250		
Jan. 14, 1952	180		182			181		
Mar. 15, 1952	2580				2690	2635		
June 3, 1952	271	267	272	261	264	267		

*H. H. Seliger, Private Communication.

TABLE IV

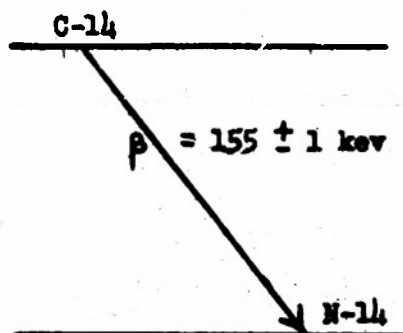
Results of intercomparisons of P^{32} Solution Standard Prepared and Distributed by the National Bureau of Standards*

Values are given in disintegrations per second per milliliter $\times 10^{-3}$ as of 0800 on the zero date

UNITED STATES				CANADA		ENGLAND		Averages
Zero Date	NBS 4π β Counter	ORNL Defined Solid Single	Columbia U Parallel Plate Chamber	AEC Chalk River 4π β Counter	Hammersmith Absolute β Ion Chamber			
Feb. 27, 1950	327	317		322		322		
Sept. 14, 1950	240	235	241	243	229	238		
Mar. 20, 1951	633	631				632		
Sept. 1, 1951	383			386		385		
Mar. 31, 1952	246		248		243	246		
Sept. 1, 1952	489	488		497		491		
Mar. 1, 1953	249			249		249		
Mar, 10, 1953	161					(161)		

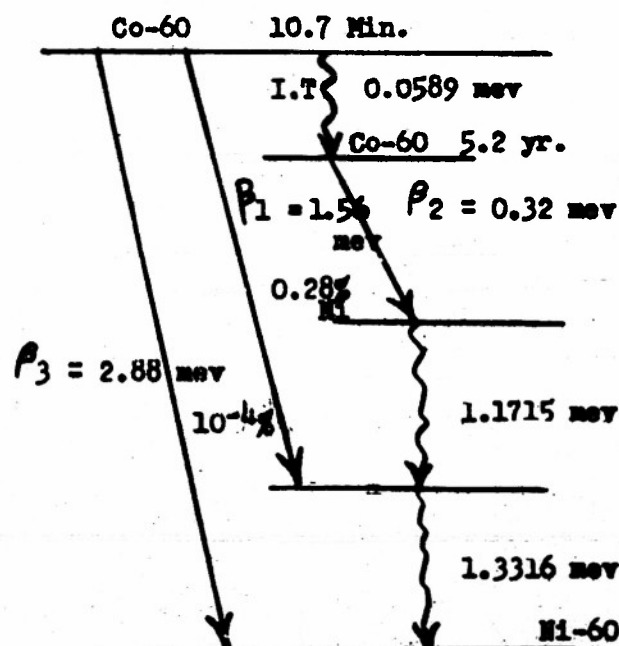
*H. H. Seliger, Private Communication.

Figure 1



Max. beta energy: S. D. Warshaw, Phys. Rev. 80, 111 (1950).

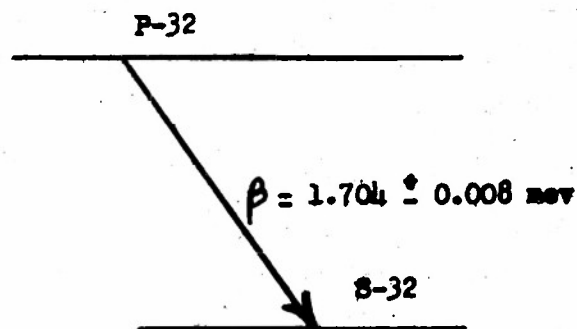
Figure 2



Decay scheme: Deutsch and Goldhaber, Phys. Rev. 83, 1059 (1951).

Gamma energies: DuMond et al, Phys. Rev. 76, 1838 (1949).

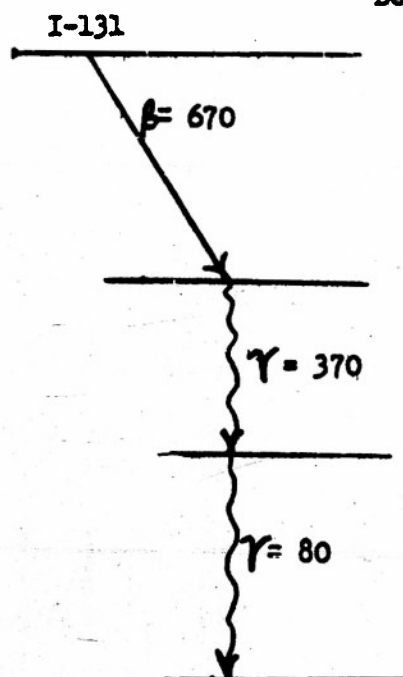
Figure 4



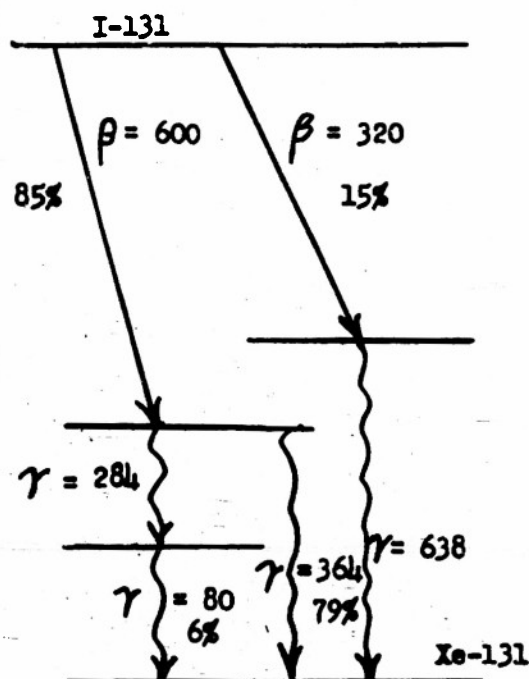
Jensen and Nichols, AECU 1264, April, 1951.

See Figure 3 for decay scheme of I-131.

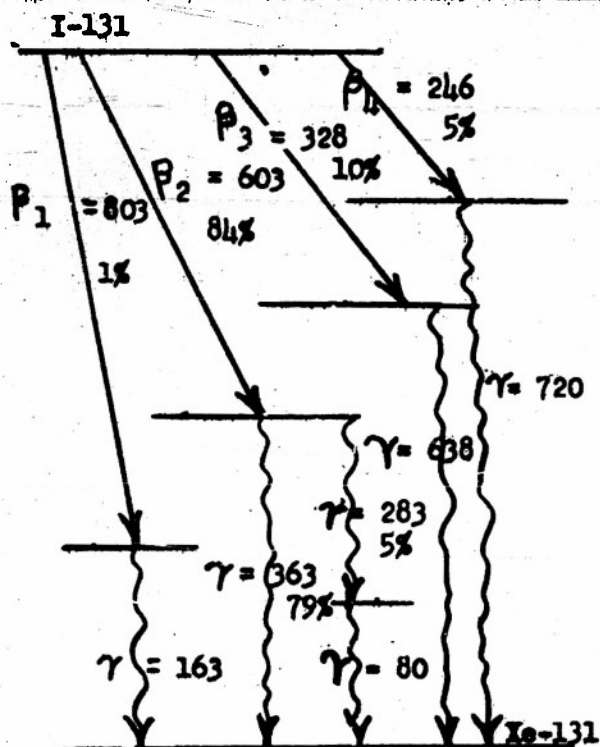
Figure 5
Decay Schemes for I-131
(Energies in kev)



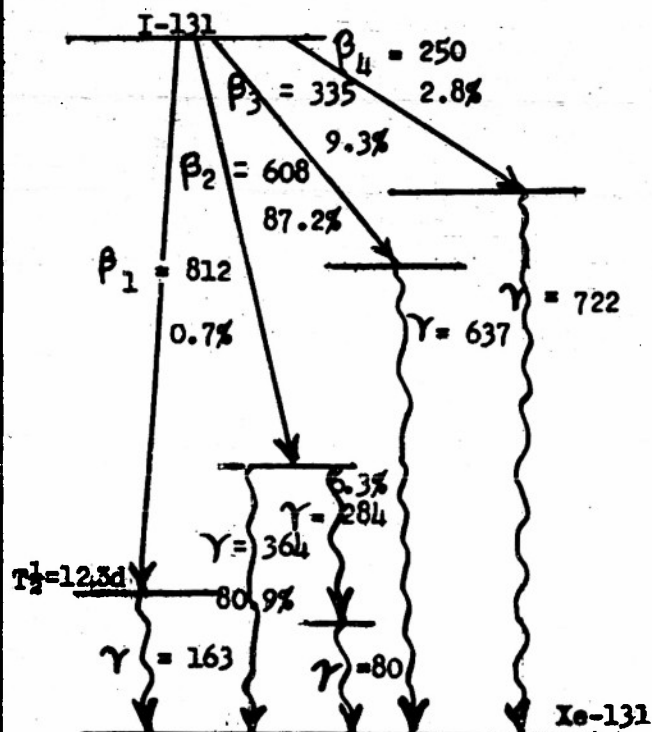
Downing, et al; Phys. Rev. 61, 686 (1942).



Metzger and Deutsch, Phys. Rev. 74, 1640 (1948)

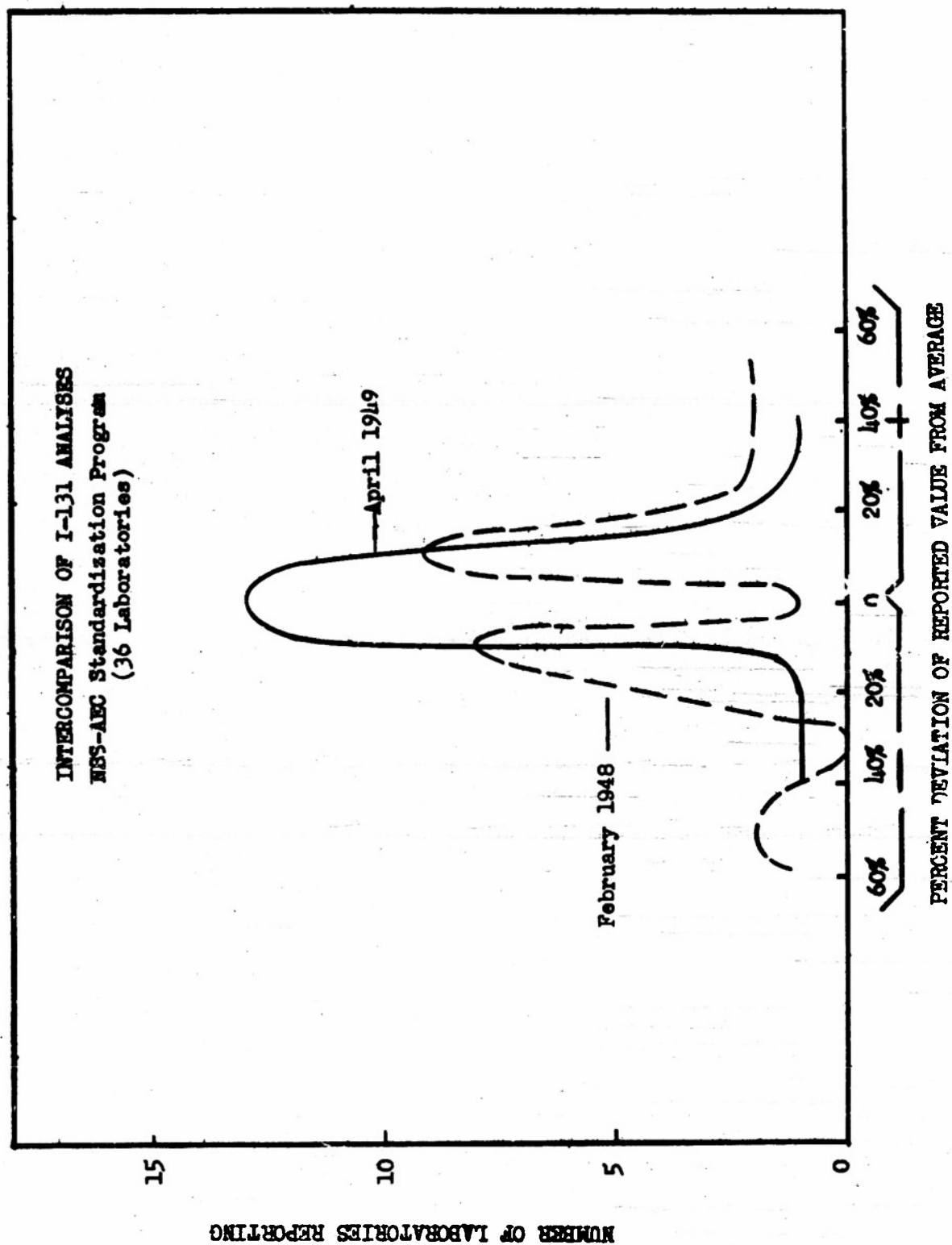


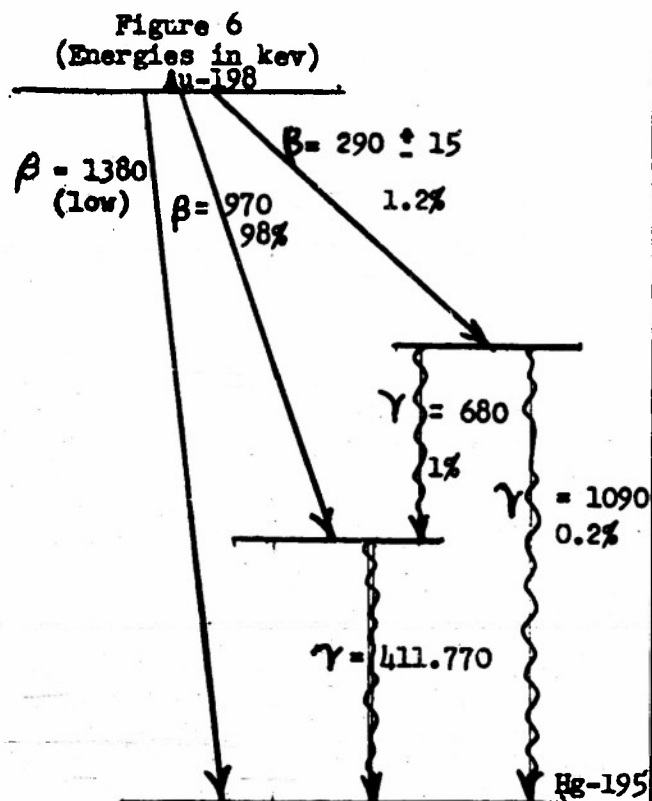
Cavanagh, Phil. Mag. (7) 43, 221 (1952).



Bell and Graham, Phys. Rev. 86, 212 (1952).
See also Haskins and Karbatov, Phys. Rev. 88, 884 (1952).

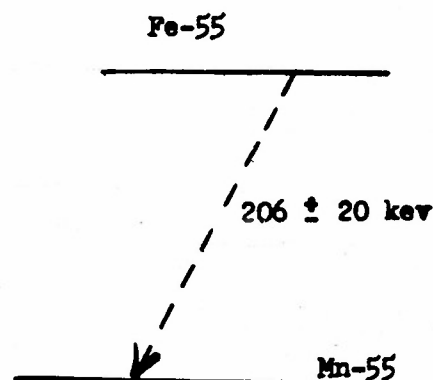
Figure 5





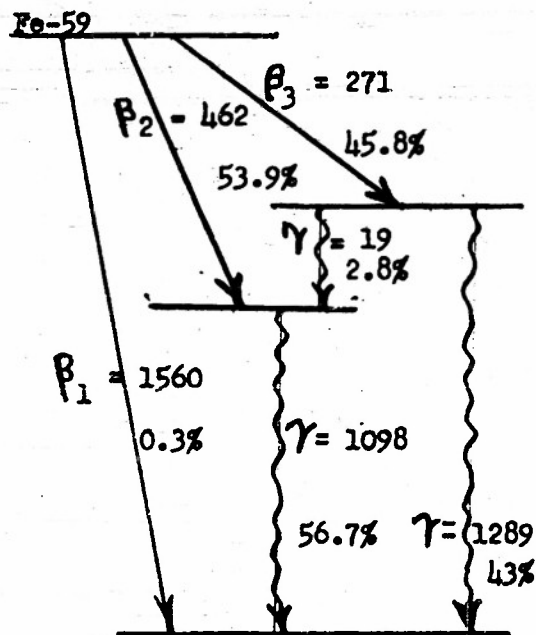
Decay scheme: Brosi, Ketelle, Zeldes and Fairstein, Phys. Rev. 84, 586 (1951).
Gamma energy: DuMond, et al, Phys. Rev. 88, 775 (1952).

Figure 7



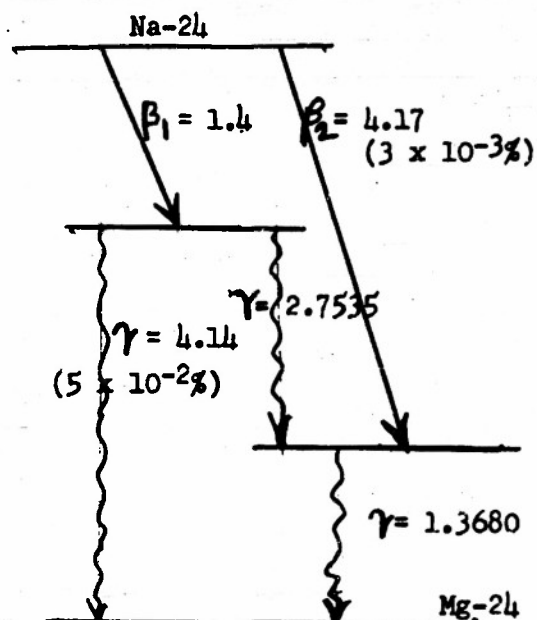
P. R. Bell, J. M. Jauch and J. Cassidy, Science 115, 12 (1952).

Figure 8
(Energies in kev)



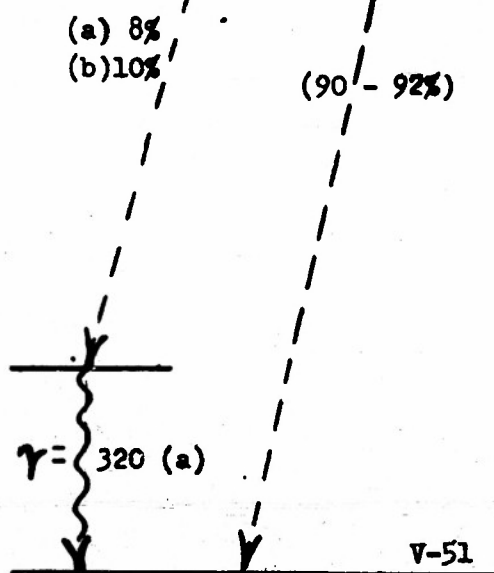
Metzger, Phys. Rev. 88, 1360 (1952).

Figure 9
(Energies in mev)



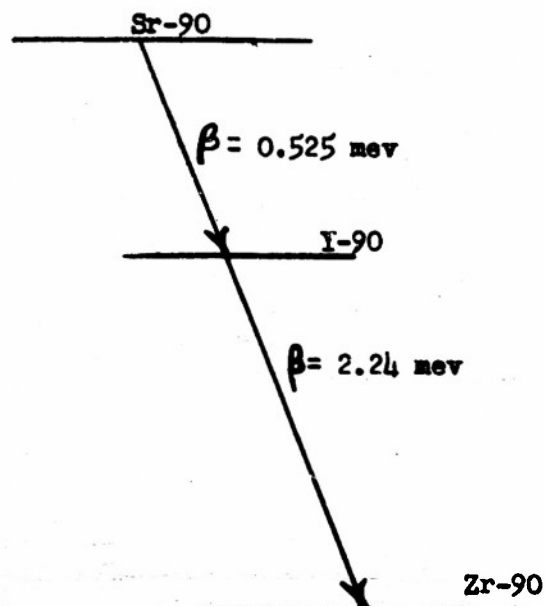
Decay scheme: Turner and Cavanagh; Phil. Mag. (7) 42, 636 (1951).
Gamma energy: Hedgran and Lind, Arkiv

Figure 10
(Energies in kev)
Cr-51 28 day



(a) W. S. Lyon; Phys. Rev. 87, 1126 (1952).
(b) A. W. Sunyar; unpublished data (1952).

Figure 11



Sr-90 Ruth A. Cohn; Ph.D. Thesis, Ohio State University (1950).
Y-90 L. R. Shepherd, Ph.D. Thesis, University of Cambridge (1950).

Figure 12

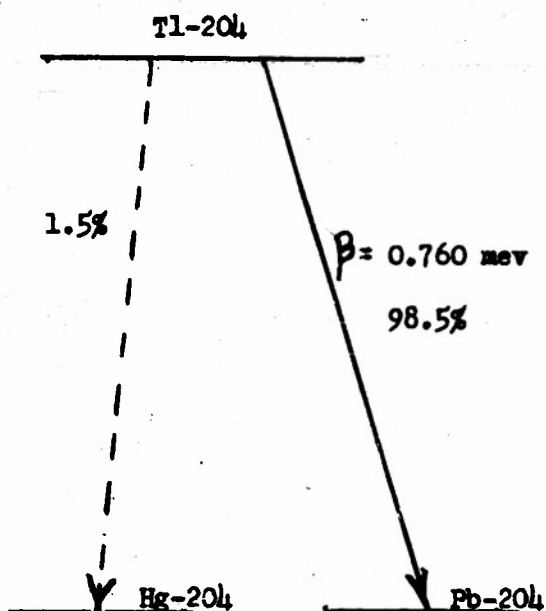
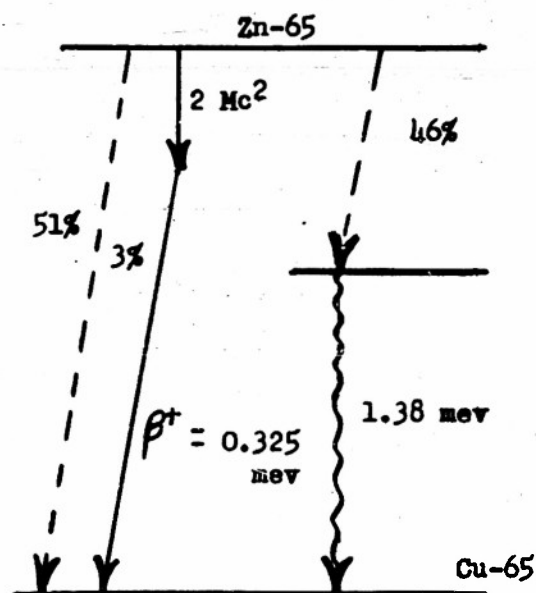


Figure 13



Mann, Phys. Rev. 76, 1719 (1949).

of BaCO_3 have been reported by Bernstein and Ballentine (60a) in which it appears that gas-phase counting in the proportional region and in the G-M region yield 5513 and 6360 years, respectively. This discrepancy appears to have been partially resolved in a later paper (60b), in which the hypothesis was advanced that "over-quenching" of the G-M counter may have occurred which led to a significant loss (13.9%) in the counting rate. If this observation and the explanation are correct, it would seem appropriate to reduce the value for the half-life obtained (60a) in the G-M region to 6360/1.139 or 5580 years. Gas-phase counting therefore leads to an estimate of 5550 years for the two sets of measurements.

Calorimetric measurements of the heat evolved in the disintegration of C-14 have been reported (31) but incomplete knowledge of the average energy of the beta radiation (45-49 kev) permits only an approximate calculation of the half-life, 5500-6000 years.

Evidently the last work on the half-life of Carbon-14 has not yet been said.

The maximum energy for the beta spectrum of C-14 is 155 ± 1 kev (61), and the decay scheme involves only a simple beta emission (see Figure 1).

2. Cobalt-60

Co-60 is generally prepared by neutron absorption in Co-59; sample preparation consists of solution in hydrochloric or other acids (with or without added cobalt carrier) and dilution to the appropriate activity.

A 10.7 minute isomeric branching has been reported to the extent of 0.28 ± 0.06 percent, decaying by a 1.56 mev beta emission to the first excited state (1.33 mev) of Ni-60. The ratio of the neutron-capture cross-sections is $\sigma_{5.2 \text{ yr.}} / \sigma_{10.7 \text{ min.}} = 1.4 \pm 0.6$. Agreement has not been fully reached for the half-life of Co-60. Recent determinations yield 5.26 ± 0.17 , 5.27 ± 0.07 , 5.25 ± 0.21 and 4.95 ± 0.04 years (63, 11, 64, 65). The last value was obtained by use of a quartz fibre electroscope over a period of 8 months.

The decay scheme involves a simple beta decay with $E_{\text{max}} = 0.306 \pm 0.005$ mev (66), followed by two gamma rays in cascade whose energies (40) are 1.1715 ± 0.0010 and 1.3316 ± 0.0010 mev, respectively (see Figure 2).

Intercomparisons of the strength of a stock

solution of Cobalt-60 by coincidence counting were made in 1947 between NBS, MIT, ORNL and NRC (Canada). These measurements resulted in the preparation by the National Bureau of Standards of two series of 5 ml ampoules with radiocobalt activities, respectively, of 1.59×10^6 dis/sec (series I) and 0.159×10^6 dis/sec (Series II) as of July 24, 1947. The estimated error was $\pm 2\%$. In 1949, several ampoules were sent to Dr. L. H. Gray of the Medical Research Council, Hammersmith, England, and to others, who measured the activity by means of gamma-ray ionization and checked it approximately by beta-gamma coincidence counting. The agreement (23) between Harwell and NBS was within 2.5%, both using beta-gamma coincidence counting (See Table I).

Recently, Barnothy and Forro (17) re-assayed the (1947) NBS ampoule (Series II) using coincidence counting and found an activity of $(1.033 \pm 0.026) \times 10^5$ dis/sec at the center of their measuring period November 7, 1950. Assuming the value 5.26 ± 0.17 years for the half-life as reported by Brownell (63), (Barnothy and Forro used Brownell's preliminary value of 5.2 years); the calculated activity for the NBS source on November 7, 1950, would be 1.031×10^5 . The agreement between the two sets of measurements is satisfactory.

Subsequent work at the Bureau of Standards has involved the preparation of another set of ampoules which are more dilute with respect to total solids (i.e., approximately 1 ug/ml, rather than 1 mg/ml), followed by collaborative measurements of the activity of these ampoules by coincidence, 4-pi and other counting methods. Preliminary results, reported by W. B. Mann and H. H. Seliger (28) are given in Table II. The average of the NBS measurements by the four methods is 0.789 mc/ml.

3. Iodine-131

I-131 is presently separated from fission products by oxidation to elemental iodine and volatilization from an aqueous solution. The radionuclide is distributed in the form of a weekly basic iodide solution containing a trace of NaHSO_3 (51).

The half-life has been reported by Sreb (67) to be 8.1409 ± 0.0062 days; 8.04 ± 0.04 days by Sinclair and Halloway (64); 8.06 ± 0.02 days by Lockett and Thomas (65); $8.075 \pm$

0.022 days by Seliger, Cavallo and Culpepper (68); and 8.05 ± 0.01 days by Hawkings and co-workers (69).

Decay schemes for I-131 have recently been reported by Bell, Cassidy and Kelley (70) and by Cork and co-workers (71). Both schemes recognize the existence of the 12-day metastable state of Xe-131 reported by Zeldes, Brosi, and Ketelle (1), but associate it with different beta energies. Additional complexities in the decay scheme have since been discovered. Verster (72), Ketelle (73), Cavanagh (74), and Bell (75) and Kurbatov (76) have confirmed the existence of two additional beta energies, 250 kev (2%), and 807 kev (1%). Rose (77) has measured internal conversion electrons but apparently did not observe the 250 kev beta energy. Bell (78) has recently found a previously unknown gamma energy of about 0.5 mev that does not fit into the presently accepted decay scheme. A summary of the decay schemes proposed for I-131 is given in Figure 3.

Caswell (30) has found the average energy of the beta radiation to be 0.189 ± 0.008 mev.

A number of methods are in use for the standardization of I-131: known geometry, coincidence counting, 4-pi counting and ionization methods prevail (15). The status of the intercomparison of iodine standards is much more promising than it was five years ago. Distribution was made by the Bureau of Standards in January, 1948, of identical samples of I-131 solution to approximately 75 laboratories throughout the United States. Statistical analysis of the values reported indicated marked divergencies—differences as great as 200% were not uncommon. (The values ranged from a low of 43×10^6 dis/sec/ml of solution to a high of 231×10^6 , with a most probable value of 146×10^6 .)

Because of the importance of this radioisotope in medicine, a joint NBS-AEC survey was made which involved individual visitation and discussion with the personnel of fifty-five of the participating laboratories. In April, 1949, another sample was distributed to thirty-six of the fifty-five laboratories visited. The results of the two sets of analyses (see Figure 5) show that it is now possible for a large number of laboratories to agree fairly well with each other.

As a result of this work, the National Bureau of Standards since 1950 has been furnishing on

a regular schedule 3-ml ampoules of I-131 (and of P-32) with a known activity in terms of disintegrations per second per milliliter of solution. The activity is of the order of 100,000 dis/sec/ml, with higher activities occasionally available by special arrangement. Provision is made for pH control, the samples are low in total solids (approximately 10 ug/ml), and instructions for use accompany each sample. 4-pi proportional counting is used for standardization (24a). The presence of the metastable, 12 day Xe-131 introduces an error which can be neglected in end-window beta counting or in ionization chamber measurements where an overall accuracy of $\pm 5\%$ is sufficient.

Approximately 200 ampoules are distributed at six-month intervals to users within the United States, Australia, Canada and Great Britain. The value of this service to the average isotope user is considerable. Frequent interchecks between NBS, ORNL, and other standardizing laboratories serve to maintain the requisite accuracy in standardization.

Samples of I-131 (and other isotopes) were exchanged in 1949 with L. H. Gray of the Medical Research Council of Hammersmith, England. The results have been published by Putman (23). The details are presented in Table I. A more recent intercomparison by Seliger (24b) is presented in Table III and indicates better agreement.

Samples of I-131 are being exchanged in an attempt to establish an international standard. The present agreement appears to be of the order of $\pm 2\%$.

4. Phosphorus-32

Neutron irradiation of sulfur, followed by chemical oxidation and separation, is used to obtain P-32 (51).

If sulfur-containing compounds are irradiated to form phosphorus, the chemistry of separation is rather involved because of the different radiochemical forms in which the phosphorus may be found. For example, if calcium sulfate is irradiated, metaphosphates (PO_3^-), ortho-phosphates (PO_4^{---}), pyro-phosphates ($\text{P}_2\text{O}_7^{---}$) and phosphites (HPO_3^{--}) are formed. Failure to recognize the chemistry involved may lead to anomalous results for the half-life (64) and to apparently abnormal tracer behavior (79).

New measurements have been reported for

the half-life of P-32: 14.30 ± 0.05 days by Bayley (12) and 14.50 ± 0.04 days by Lockett and Thomas (65). The maximum beta energy has been found to be 1.704 ± 0.008 mev. (80).

The existence of P-33 in reactor-produced P-32 has been reported by Jensen (2, 80) and confirmed by Sheline (81) and others. The half-life is 24.8 ± 0.5 days, and the maximum beta energy is 0.26 ± 0.02 mev (80).

Collaborative determinations of the absolute strength of P-32 samples have been carried out for a number of years between several standardizing laboratories, and the results have recently been summarized by Seliger (24b) as shown in Table IV.

Ampoules containing known amounts of Phosphorus-32 have been available since 1950 from the National Bureau of Standards. These ampoules contain approximately 100,000 dis/sec/ml of P-32 with 0.001 M H_3PO_4 added as carrier. The effect of P-33 on the measurements is considered to be negligible if the time interval involves less than two half-lives. 4-pi proportional counting is used (24b). The availability and the method of distribution⁷ for P-32 and I-131 are identical.

5. Radium-226

Radium standards have been available for many years, but minor uncertainties still persist in determining the activity of one gram of radium. Careful measurements of the specific activity of radium were made by Kohman (83) in 1947, yielding $(3.608 \pm 0.028) \times 10^{10}$ alpha disintegrations per second per gram of radium, and a half-life of 1620 ± 13 years. One curie (of radium or of any other nuclide) was redefined in 1950 to be equivalent to 3.700×10^{10} dis/sec (5b, 6).

6. Pb-Bi-210 (Ra D-E)

Pb-210 (RaD) is obtained by chemical separation from the equilibrium radium mixture, and an organic reagent, dithizone, is useful (84) for this purpose. Electroplating under carefully controlled conditions (85) is also employed to effect the separation.

The spectrum of Pb-210 is of theoretical interest, and the results of several investiga-

tions (86a, b, c) have been reported. Cohen and Jaffe (87) have found that over 90% of the beta radiation has an energy of less than 5 kev.

Bi-210 (RaE) is obtained as the daughter product of Pb-210. Values of 5.02 ± 0.02 (88) and 4.989 ± 0.013 (65) days have been reported for the half-life of Radium-E.

Details of the preparation and calibration of RaE sources have been discussed by Novey (89). The original method for preparing these standards was the subject of discussion between the National Bureau of Standards and the National Research Council. The method decided upon at the time, early in 1946, was a compromise between (a) accurate standards desirable for the future, and (b) the immediate need for a fairly large number of reproducible standards with a somewhat lesser degree of accuracy.

The NBS standards are composed of a circular deposit of active and inert PbO_2 weighing approximately 1 milligram; the material is deposited electrolytically in the center of a palladium-faced silver disk approximately 1 inch in diameter and $\frac{1}{16}$ -inch in thickness. Calibration was initially made by assaying accurately a radium ore for activity, extracting the total lead therefrom, and then electrolytically depositing a portion of the lead as lead peroxide. Several hundred such standards have been prepared and distributed.

In 1950, Burt (90, 91) and others found that there was a forward scattering of the beta particles from RaE because of the small thickness of the PbO_2 deposit. The scattering amounted to approximately 7% under the particular conditions of geometry, backscatter, sidescatter, etc., used by him. Seliger (27) has made additional measurements of backscattering from various sources.

These refinements in measurement point up a fact that has long been recognized; i.e., that measurements made by comparison of the beta activities between a standard of a given radionuclide and the "unknown" should be made under identical conditions of geometry, thickness of deposit, backscattering and that the energies of the standard and of the "unknown" and the shapes of their beta spectra should be as nearly alike as possible.

An independent calibration of the strength of the RaE sources can be obtained by measuring the growth of the alpha activity from RaF. Such measurements have been made (89) and

⁷ Ten microcurie quantities of radionuclides with a half-life of less than 30 days and 1 microcurie quantities with a half-life over 30 days may now be interchanged between laboratories without prior approval by the U. S. Atomic Energy Commission (82). This action has served to stimulate intercomparisons not only of materials proposed as standards, but of various intermediate preparations. It applies, however, only to transfers within the United States.

indicate that if proper corrections are applied, the agreement between beta and alpha counting is within 2%. National Bureau of Standards Pb-Bi-210 standards are available in activities approximating 100, 200, and 500 and 1000 dis/sec, and have been widely used in this country and abroad.

The international standardization of Pb-Bi-210 has not yet been attempted.

B. DESIRABLE STANDARDS; SOME NUCLEAR INFORMATION AVAILABLE

Summaries are presented for the following radionuclides: Au-198, Fe-55, Fe-59, Na-24 and H-3.

1. Gold-198

Au-198 is prepared by neutron irradiation of metallic gold. Radio-gold is used medically in the treatment of some forms of cancer. It is interesting to note that 65 curies of Au-198 have been used for this purpose.

The half-life of Au-198 has been found to be: 2.69 ± 0.01 (10), 2.73 ± 0.01 (64), 2.697 ± 0.003 (65), and 2.74 ± 0.04 (66) days.

The decay scheme has been studied recently by a number of workers (66, 93, 94, 95, 96). The major portion (98%) of the gold decays by a simple beta emission, 0.97 ± 0.01 mev (66), followed by a gamma ray of 0.411770 ± 0.000036 mev (92), and the minor portion (1.2%) with a beta energy of 0.290 mev and a gamma energy of 1.09 mev. The work of Brosi (94) confirms that of Cavanagh (93) (see Figure 6). It has been determined (34) that the half-life of excited (411 kev) Hg-198 resulting from the beta-decay of Au-198 is less than 3×10^{-11} seconds.

Hill and Mihelich (96) observed two weak gamma-rays of 159 and 209 kev which have been attributed to Au-199 formed by the reaction: $\text{Au-197} + n_0 = \text{Au-198}$; $\text{Au-198} + n_0 = \text{Au-199}$. The half-life for Au-199 was observed to be 3.3 days, the maximum beta-energy is 0.32 mev (7) and the calculated cross section for Au-198 for thermal neutrons is estimated to be about 3.5×10^4 barns. The relative proportions of Au-198/Au-199 were estimated⁸ to be 95/1 for a sample irradiated for 8 days in a flux of 10^{12} n/cm²/sec (96).

Standards for Au-198 are needed. Presently,

⁸ A recalculation using the data supplied by Hill and Mihelich (96) indicates that the ratio of Au-198 to Au-199 for an infinitely long irradiation time is approximately 1000 to 1. (Private communication from H. H. Seliger of NBS).

measurements of the strength of these sources are generally made by comparison with a radium needle using an ionization chamber counter or electroscope. ORNL assays Au-198 routinely by means of a high pressure ionization chamber calibrated periodically by 4-pi coincidence counting of the same sample. Because of its comparatively short half-life, it is doubtful that standard samples of Au-198 can be distributed in the usual sense.

Two courses of action may be open: (a) standardization of a Au-198 source by coincidence counting and simultaneous comparison with radium in an ionization chamber under rigidly specified conditions; the gold might then be certified in terms of radium-equivalents, (b) search for an isotope of reasonably long half-life that has either the same beta or gamma energy as Au-198. Secondary standards might then be distributed for comparison with either the beta or the gamma radiation, as appropriate.

2. Iron-55

Fe-55 is of interest because the decay is thought to proceed almost entirely by K-capture. Scintillation counting (97) has shown that the continuous radiation has an upper limit of 0.205 mev. A later value (37) of 0.206 ± 0.020 mev has been reported. The decay scheme is shown in Figure 7. A new value for the half-life, 3.0 ± 0.15 yr. has been reported (98) in agreement with an earlier one of 2.94 years (63).

Preliminary results on intercomparison of samples of Fe-55 reported by Reynolds (99) indicate that an agreement of the order of $\pm 15\%$ is possible. Details of the techniques used are to be published shortly.

3. Iron-59

Fe-59 is generally produced by cyclotron bombardment. Two values for the half-life have been reported: 45.1 ± 0.2 (98) and 47.1 ± 0.5 (11) days.

Recent decay schemes by Rose and co-workers (100) and by Metzger (101) for Fe-59 are shown in Figure 8.

4. Sodium-24

Na-24 is produced by neutron irradiation of stable sodium. The half-life of Na-24 has been reported by Sreb (102) to be 15.060 ± 0.039 hr., and by Lockett and Thomas (65) at 14.97 ± 0.02 hr. The earlier value of Wilson and Bishop (103) was re-calculated by Sreb and yielded

14.96 ± 0.10 hr. The three values agree within the limits of error.

Considerable interest has been manifested in the decay scheme of Na-24. Turner and Cavanagh (104) have found evidence for a 4.17 mev beta group and estimate the branching ratio to be $3 \times 10^{-3}\%$. Siegbahn and Johansson (43) have used a new type of pair spectrometer in which rotation of the image in a short magnetic lens is used to separate negatrons from positrons by putting the pair-converter a little aside from the optical axis. The simultaneous emission of three particles (a decay beta-ray and an electron-positron pair) from Na-24 has been examined (105) and the value of the pair-conversion coefficient for the 2.78 mev gamma-ray has been determined to be $(7.6 \pm 1.9) \times 10^{-4}$. A pair conversion coefficient of $(8.25 \pm 1.05) \times 10^{-4}$ was obtained for positrons stopped in beryllium.

Hedgran and Lind (106) have measured the energy of the gamma rays from Na-24 and report 1.3680 ± 0.0010 and 2.7535 ± 0.0010 mev.

Absolute measurements of the activity of Na-24 have been performed by using cavity-type ionization chambers (25) and have been compared with the results of coincidence counting (23). The results are shown in Table I. It will be difficult to set up standards for such a short-lived material.

The decay scheme for Na-24 is shown in Figure 9.

5. Tritium

Two recent values for the half-life of tritium yield 12.46 ± 0.1 and 12.41 ± 0.20 years (107, 108). Recent measurements of the maximum beta energy by Langer and Moffat (109) yield 17.95 ± 0.10 kev with an average energy (107) of 5.69 ± 0.04 kev. A bibliography of the unclassified literature on tritium has been compiled (110).

The low energy of the beta radiation from tritium makes quantitative measurements difficult or impractical by ordinary methods such as end-window counting and recourse must be had to gas-phase measurements (108, 111, 112).

For samples which are essentially pure tritium, the method of gas-density (107) can be used and it is claimed that an accuracy of ± 0.05 percent can be attained within the range of 50-100% T_2 . A check on the composition of tritium-deuterium-hydrogen mixtures is ob-

tained by mass-spectrographic analysis. The tritium can be stored on spongy zirconium or palladium, and released by gentle heating.

For lower concentrations of tritium, internal gas-counting can be used. The counters are of brass with flat end-plates similar to those used by Libby (56) with alcohol-argon in the ratio of 1 to 9 at a total pressure of 10 cm as the filling gas. The tritium is quantitatively diluted with ordinary hydrogen to give a reasonable counting rate in the G-M tube. A counting efficiency of 97% is assumed, and the length of the plateau is approximately 200 V with a 1000 V threshold and a slope of approximately 3% per 100 V. Backgrounds of approximately 200 c/min on a counter of $1\frac{1}{2}'' \times 12''$ are obtained. Unfortunately, some memory effects are associated with this type of counter, and the background has been found to increase approximately 200 c/min for each filling. The counters can be cleaned thoroughly by rinsing with diluted aqueous ammonia and washing with distilled water.

Robinson (111) has used a mixture of tritium and methane as a filling gas. The advantage is that the resolving time is very short, that counting can proceed at a high rate of speed without undue coincidence losses, and it is claimed that the memory effects are smaller.

It should also be possible to use a vibrating-reed electrometer and an ion chamber for routine measurements. Such a chamber could be calibrated at high concentrations of tritium by means of the gas-density balance and at low concentration by G-M gas-phase counting.

It was recently proposed (59b) that standard samples of tritium be prepared in the form of tritiated water with an approximate activity of 10^6 to 10^7 dis/sec/ml.

C. DESIRABLE STANDARDS; LITTLE NUCLEAR INFORMATION AVAILABLE

Summaries are presented for the following radionuclides: Cr-51, Sr-90, Tl-204, and Zn-65.

1. Chromium-51

Cr-51 is produced by neutron irradiation of Cr-50. A value of 27.75 ± 0.3 days for the half-life has been reported recently by Lyon (4), and the decay scheme has been examined (4, 113). (See Figure 10). Reynolds (59b) has reported measurements of the disintegration rate of the same sample of Cr-51 (a) by X-gamma coincidence counting and (b) by X-ray

counting with a proportional counter standardized by means of a source of Mn-54 of known disintegration rate. Results by the two methods agreed within 3%.

2. Strontium-90

Sr-90, obtained by separation from fission products, is a pure beta-ray emitter which decays to Y-90 and then to stable Zr-90. Contemplated usage of Sr-90 in luminous buttons may amount to millions of units, totalling perhaps hundreds of curies. Standards of Sr-90 will be needed for research, production, and testing.

A value of 19.97 ± 0.3 years (114) has been reported by Powers and Voigt for the half-life. Measurements were made over a period of seven years. The value is significantly different from the currently accepted figure of 25 years.

The maximum beta-energies of Sr-90 and Y-90 according to Cohn (115) are 0.525 and 2.24 mev, respectively; for Y-90, Moreau and Corba (116) found 2.270 ± 0.02 mev. Similar measurements have been made by Shepherd (117) on Y-90. The average beta energy of Y-90 is 0.895 ± 0.035 mev (30). The decay scheme for Sr-Y-90 is shown in Figure 11.

3. Thallium-204

Tl-204 is prepared by neutron irradiation of Tl-203 and is made available as an aqueous solution of the nitrate. Various difficulties have been encountered in the preparation of samples with reproducible counting rates. Tl-204 has chemical oxidation states of +1 and +3 in aqueous solution, and it has been reported by Peacock (59b) that electroplating is not satisfactory as a method of separation from other electrolytes. Thallous nitrate solutions in dilute nitric acid have been found by Reynolds (59b) to be more stable than neutral or alkaline solutions of thallic ion or halide solutions of thallous ion.

Values for the maximum beta energy of thallium-204 have been reported to be $0.765 \pm$

0.010 mev by Lidofsky, Macklin and Wu (118) and 0.760 ± 0.01 mev by der Mateosian and Smith (119). The Kurie plot is linear down to 0.150 mev (118) and the decay scheme is shown in Figure 12.

A value of 3.9 yr. for the half-life of Tl-204 has been reported by Harbottle (120), compared to two other values (7) of 2.7 and 3.5 yr.

Reynolds (59b) has reported the results of a limited intercomparison (6 laboratories) of a solution of thallium-204 whose activity was approximately 1×10^{-3} uc/ml. The following methods were used (the results obtained are given in parenthesis): 4-pi counting (1.00×10^{-3} uc/ml); 2-pi counting corrected for backscatter and self-absorption (0.98×10^{-3}) and comparison with RaD-E and G-M end-window counting, (0.91×10^{-3}). The results appear promising, with a deviation of $\pm 5\%$. Further work is underway.

4. Zinc-65

Zn-65 is prepared by irradiation of metallic zinc and is made available as aqueous zinc chloride (51).

No new measurements of the half-life, approximately 250 days (7), have been reported.

A small percentage of the disintegrations are by positron emission, the main decay process being by K-capture. Furberg (121) has determined the branching ratio⁹ $K_2/(K_1+K_2)$ to be 0.44 ± 0.03 . Major (122) has found 0.45 for the branching ratio. Griffiths (123) by coincidence measurements found that 1 positron was emitted for every 65 ± 5 gamma rays.

Mann and co-workers (124) measured the beta spectrum by means of a thin lens type magnetic spectrometer and found 0.325 ± 0.002 mev for the positron and 1.38 ± 0.03 mev for the gamma radiations. The decay scheme is shown in Figure 13.

⁹ Where K_1 = number of capture processes leading to the ground state, and K_2 = number leading to the excited state.

V. SUMMARY

1. Working standards are available for C-14, Co-60, I-131, P-32 and Ra D-E. Progress made in this respect is such as to warrant formal exchange of samples for intercomparison and standardization leading to international standards for these isotopes. Suitable standards for radium and radon already exist.

2. Additional experimental work needs to be done on nuclides such as Fe-55, Fe-59, Au-198, Na-24, and H-3.

3. Radionuclides for which standards may be desirable in the foreseeable future are Cr-51, Sr-90, Tl-204 and Zn-65.

VI. REFERENCES

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