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SOME RADIOISOTOPE ASSAY METHODS

S. A. Reynolds

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ISOTOPES DEVELOPMENT CENTER

SOME RADIOISOTOPE ASSAY METHODS

S. A. Reynolds

Isotopes Division and
Analytical Chemistry Division

MARCH 1973

OAK RIDGE NATIONAL LABORATORY
Oak Ridge, Tennessee 37830
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SOME RADIOISOTOPE ASSAY METHODS

S. A. Reynolds

SUMMARY

A review is presented of over a quarter-century of work in calibrations for ORNL radioisotope products. Internal standardizations are surveyed, and details are given of measurements of materials produced by standardizing laboratories, particularly the National Bureau of Standards (NBS). The need for reliable nuclear data is stressed. Specific information, historical as well as current, is included on eight representative products. General suggestions are made for the benefit of others who are required to measure radionuclides. Items included are standards, personnel, equipment, and quality control.

INTRODUCTION

For nearly three decades radionuclides have been produced at Oak Ridge National Laboratory (ORNL). This report surveys their calibration and measurement. Reviews were published about one[1] and two[2] decades after initiation of the program, and little of that material is repeated. The work described here has been part of a cooperative program[3,4] of the Isotopes and Analytical Chemistry Divisions; much of the experimentation has been performed by members of the latter division. The facilities and expertise have been applied in support of activities of a number of other divisions,[5] such as Health Physics,[6] Reactor Chemistry, and Operations; but the present discussion is limited to Isotopes Division work.

Discussion is further limited to laboratory-level calibrations expressed in disintegration rate or microcurie units. Large-source methods such as calorimetry and dosimetry (expressed in rads) have been covered elsewhere.[7-10] Purity of materials needs to be considered only as it affects their measurement.

DEVELOPMENT OF THE PROGRAM

A formal quality-control program on radioisotopes has never been set up, but there have been conscientious effort and excellent cooperation among routine and specialized analytical personnel and production workers. Although occasional errors — perhaps in dilution or calculation — have been made, few legitimate challenges of local calibrations have arisen. A great majority of users' complaints about assays have been shown to be due to lack of basic nuclear or radiochemical information or their poor

standardization. The local program has required measurement of over 175 nuclides, comparable to numbers needed at other large laboratories, for example, about 170 at the British Radiochemical Centre (RCC)[11] and 125 at Pacific Northwest Laboratories.[12]

Because of the world-wide use of radioisotopes, the program has always been outwardly directed, involving cooperation with the National Bureau of Standards (NBS), International Atomic Energy Agency (IAEA), Public Health Service (PHS), American Society for Testing and Materials (ASTM), National Research Council (NRC) (particularly its Subcommittee on the Use of Radioactivity Standards[13]), and other organizations. Such cooperation included supplying of materials, assisting in preparation of documents (such as a metrology handbook[14] and ASTM methods[15]), participating in conferences,[16-19] and taking part in intercomparisons.[1,16,18,20,21] The author conducted a survey[20] for an NRC panel on standards; this survey served as the principal factual basis for the panel's report.[22]

An informal program of training in measurement techniques was carried on, largely in the years 1950-1965. Participants included personnel of the National Reactor Testing Station, the Savannah River Laboratory, PHS, etc., and scientists from Africa, Argentina, Greece, India, Thailand, and elsewhere. The author taught an informal "Analytical Radiochemistry Survey Course" 16 times during that period.

ORNL has never claimed to issue standards[5] or to calibrate routine shipments, thus negating its designation[21] as one of the "standardizing laboratories." However a number of requests for standards was received, and some users have employed shipments for calibration. On a few occasions, "analyzed samples" were furnished, e.g., to a New Zealand laboratory in 1954, to AEC Commissioner Libby in 1956,[23] and to PHS in 1960. Methods for 60 radionuclides were tabulated in a memo in 1956 and were quoted (with NBS methods for 19 nuclides) in a reference book.[24] Later information was given in a journal article,[25] the *ORNL Master Analytical Manual*,[26] and the *Isotope User's Guide*.[27]

STANDARDIZATION

Because of some discrepancies in their usage, the terms primary and absolute are avoided. Instead, direct is applied to techniques such as 4π, coincidence counting, or defined geometry (for alpha), which yield disintegration rates directly. Derived calibrations are based, e.g., on energy-efficiency curves for ionization chambers or spectrometers. A standard, or standard reference material (SRM), is furnished by a specialized laboratory such as NBS. A substitute standard is a nuclide of properties, e.g., beta energy, similar to the desired nuclide but having characteristics, such as a longer half-life, that make its use more practical. The subject of standardization has been covered thoroughly in the literature,[2,14,16-18,28-31] and no attempt will be made to review it here. Each nuclide must be considered individually, hence the detailed sections later in this report.

Direct Techniques

Calibrations at ORNL were essentially "state-of-the-art", involving such now-familiar methods as 4π beta counting, coincidence counting (including 4π beta-gamma[32,33]), and defined geometry. In addition, several direct methods of less wide application were devised, including x-gamma[34] and alpha-gamma[32] coincidence and "absolute" liquid scintillation.[35]

Outside Standards

Of necessity, calibrations at ORNL antedated work by NBS or any international organization (except on ^{226}Ra). Further, analytical requirements had short "lead times," as new radioisotope products were developed very rapidly, or especially when unexpected impurities required immediate assay. No outside laboratory could be expected to keep pace. The first NBS SRM's in 1948-1950 were used for intercomparisons, and problems were encountered.[2,16] Soon, however, the program became quite active,[21] and the standards which were issued were very reliable. A policy of not competing with commercial suppliers resulted in transfer of responsibility for certain nuclides to Nuclear-Chicago Corporation in 1957 and generally has kept the NBS program small. Some solution and point-source standards are issued,[36] and routine calibrations are offered for certain nuclides.[37] In addition, a nonroutine calibration service is said to be available, but shortage of personnel and funds and necessity of advance programming of work have resulted in the inability to fulfill most requests.[22]

The value of intercomparisons with reliable laboratories was recognized early, especially by the author, for the obvious purpose of confirming local calibrations, as well as for evaluating the SRM's offered and anticipating questions of isotope users who might calibrate by means of those SRM's. *Traceability* to NBS was especially desirable, though not dictated by regulations or by wide employment of NBS SRM's by users,[22] domestic or foreign. The practice by Nuclear-Chicago of having master solutions measured by NBS made it possible to continue to relate some local and NBS calibrations indirectly[38] until 1968. The smallness of the NBS program resulted in invasion of the domestic market in the 1960's, especially by RCC and IAEA, and quite a few of those products were measured locally. Typical early results have been summarized,[1,2] and recent ones are given in Table 1. It should be noted that agreement with NBS is comparable to that with other standardizing laboratories. (Nickel-63 is discussed in a later section.)

Radioisotope materials were gradually withdrawn from production at ORNL, and the personnel who had performed direct calibrations were transferred to other programs. It, therefore, became necessary to depend more on outside standards. In addition, the NRC standards panel strongly recommended[22] establishment of traceability to NBS, thus strengthening the trend.

Table 1. ORNL Measurements of Standards, 1965 Through 1972

Nuclide	Half-Life[a]	Origin of Standard[b]	Year of ORNL Measurement	Difference (%)
Hydrogen-3	12.3 y	NBS	1971	0.4
Carbon-14	5730 y	NBS	1967	1.0
		NBS	1971	0.0
Sodium-22	2.60 y	IAEA	1965	0.5
		IAEA	1967	1.3
		IAEA	1968	1.0
Phosphorus-32	14.3 d	NBS	1972	0.1
		RCC	1972	3.8
Sulfur-35	88 d	IAEA	1967	3.0
		IAEA	1968	0.6
		RCC	1970	0.8
		NBS	1971	2.8
		CEA	1971	2.5
		RCC	1971	2.8
Chlorine-36	0.30 My	NBS	1966	2.7
		NBS	1969	4.8
		NBS	1970	0.8
Argon-37	35 d	NBS	1972	3.5
Calcium-45	164 d	N-C	1967	5.9
		RCC	1969	6.4
		RCC	1972	0.0
Manganese-54	313 d	IAEA	1965	0.2
		IAEA	1967	1.1
		IAEA	1967	1.4
		IAEA	1968	1.3
Iron-55	2.7 y	IAEA	1969	3.6[c]
		NBS	1969	2.6[c]
Iron-59	44.6 d	NBS	1970	0.4
Cobalt-57	271 d	N-C	1966	1.2
		IAEA	1967	1.3
		IAEA	1968	0.1
		RR	1968	1.4
		RR	1969	1.7
		NBS	1972	1.1
Cobalt-60	5.27 y	IAEA	1965	0.4
		RCC	1966	0.3
		RCC	1966	0.2
		IAEA	1967	0.3
		IAEA	1968	1.8
		IAEA	1969	0.7
		IAEA	1970	0.9
		NBS	1971	1.3
		NBS	1972	2.8
Nickel-63	100 y	RCC	1967	4.6
		NBS	1969	17.3
Gallium-67	78 h	RCC	1972	0.8
Selenium-75	120 d	RR	1966	2.6
		RR	1969	3.6
Krypton-85	10.7 y	NBS	1970	1.7
Rubidium-86	18.7 d	RCC	1966	0.2
Yttrium-88	107 d	IAEA	1967	0.4
		IAEA	1968	2.2
Strontium-90	28.5 y	IAEA	1966	1.0
		IAEA	1968	1.0
Molybdenum-99	66.2 h	RCC	1967	2.7
		RR	1968	0.6

Table 1. continued

Nuclide	Half-Life[a]	Origin of Standard[b]	Year of ORNL Measurement	Difference (%)
Technetium-99m	6.02 h	RR	1968	0.6
Ruthenium-103	39.5 d	RCC	1966	2.3
Ruthenium-106	1.01 y	RCC	1966	0.3
		RCC	1968	0.4
Cadmium-109	462 d	NBS	1971	2.0
		RCC	1972	0.0
Silver-110m	250 d	RCC	1970	0.6
Indium-114m	49.5 d	RCC	1967	0.2
Iodine-125	60 d	RR	1969	1.3
		NBS	1972	1.1
Iodine-131	8.05 d	N-C	1967	1.8
		NBS	1972	0.7
Xenon-133	5.29 d	NBS	1972	~7[d,e]
Cesium-131	9.7 d	RCC	1967	10.6[d]
Cesium-134	2.06 y	RCC	1969	3.7
Cesium-137	30 y	IAEA	1965	2.5
		IAEA	1967	1.0
		IAEA	1967	0.2
		IAEA	1968	2.1
		IAEA	1969	0.3
		IAEA	1970	1.3
		NBS	1972	2.2
Barium-133	10.5 y	RCC	1966	0.0
Cerium-144	284 d	NBS	1966	3.2
Neodymium-147	11.0 d	RCC	1968	1.7
Promethium-147	2.62 y	NBS	1966	1.4
		RR	1966	1.6
		RR	1966	0.0
		RCC	1966	3.7
Thulium-170	128 d	RCC	1966	1.8
Mercury-197	64 h	RR	1966	0.0
		RR	1967	2.9
Mercury-203	46.7 d	RCC	1966	10.3
		IAEA	1967	3.0
		IAEA	1968	5.2
Americium-241	433 y	IAEA	1967	5.7[f]
		IAEA	1968	1.9[f]
		Average difference:	Overall[g]	1.9
			NBS	2.4[h]

[a]Determined or adopted by ORNL.
[b]CEA = French Commissariat a l'Energie Atomique
IAEA = International Atomic Energy Agency
N-C = Nuclear-Chicago (stocks checked by NBS)
NBS = U.S. National Bureau of Standards
RCC = British Radiochemical Centre
RR = Intercomparison; difference is between ORNL and "best" or average value.
[c]Differences in opposite senses.
[d]Suspect, rejected from average.
[e]Indirect comparison; weak sample.
[f]By gamma; alpha preferable.
[g]41 nuclides, 92 samples.
[h]22 samples; 1.7% without nickel-63.

Need for Nuclear Data

Proper use of SRM's — as well as radioisotope products themselves — requires knowledge of half-lives and decay schemes.[2] Important compilations in this field have been done by groups at Berkeley[39] and Oak Ridge.[40,41] The National Science Foundation has supported an active program, based on recommendations[42] by NRC. Compilations quickly become obsolete,[42] and most do not contain needed information such as gamma percentages (photons per 100 disintegrations). Therefore, radioisotope-analysis personnel have been forced to make selections,[2,27] for example, a table[43] and the author's chapter[44] in a handbook. In addition, many half-lives[45] and some decay-scheme parameters[2] have been determined experimentally. A study was recently made of the percentage of 88-keV gamma radiation in ^{109}Cd, a useful standard also emitting 22-keV x rays. The value was 3.8 ± 0.1%, in agreement with an estimate by D. J. Horen of the ORNL Nuclear Data Group and an unpublished result by French investigators.

APPLIED MEASUREMENTS

Establishment of the "efficiency factor" of an instrument for measurement of a particular nuclide is a simple matter when a SRM of that nuclide at an appropriate level of activity is available. Most nuclides are short-lived so that a normalizing source of similar radiation properties but of extremely long or well known half-life is measured at the same time as the SRM, and subsequent measurements of the nuclide are adjusted by means of the normalizing source. Radium-226, ^{137}Cs, ^{133}Ba, and natural uranium have been used as gamma reference sources. Cobalt-60, ^{14}C, uranium, etc., have been used for beta normalization, and ^{233}U and ^{239}Pu for alpha. SRM's and local direct calibrations have not been available for many of the radioisotopes produced at ORNL. Substitute standards and calibration curves have therefore been widely used. Most difficulty has been experienced with low-energy beta or x-ray emitters. In some cases, standardizable gamma nuclides emitting similar beta or x radiation were used to calibrate instruments, e.g., ^{95}Nb for ^{35}S and ^{60}Co for ^{45}Ca, foreshadowing the "efficiency tracing" technique.[31]

Specific Instruments

A table has been given[2] showing types of instruments and times of adoption for radioisotope work, and descriptions and principles of operation have been published.[14,26,31,46] Alpha efficiency of a 2π proportional counter for a flat thin source is essentially independent of energy and is about 51% for nickel or stainless steel backing and 52% for platinum backing. Liquid scintillation counting for weak beta emitters requires standards of the particular nuclides, and difficulties with sorption and other effects sometimes arise, requiring, for example, addition of small quantities of carriers. For other beta emitters, 2π or end-window counters are used; the latter require extrapolative correction,[47,48] using a standard of comparable energy, for the window and intervening air. X rays are counted by means of an end-window proportional[34,49] or

semiconductor[49] detector. Whenever feasible, a nuclide is measured by its gamma radiation. Standard efficiency-versus-energy curves are available for NaI(Tl) detectors,[48-50] and a curve can be prepared for a given semiconductor detector. It should be noted that for spectrometers the accuracy of the computed efficiency for a particular nuclide is dependent on the reliability of decay-scheme information to a greater degree than for most other instruments.

Special attention is given here to the 4π gamma ionization chamber (GIC), because of its precision (about 0.2%), useful range (over 10^4), relative insensitivity to sample volume, and wide use, particularly in medical ("dose calibrators") and general-assay work. The ORNL model[51-53] is filled with argon under pressure and is similar to one at NBS[28] and some of the "NPL chambers."[54] The local GIC's are calibrated with ^{60}Co solution samples from a stock whose disintegration rate has been determined by coincidence counting. Efficiencies for other nuclides[53] relative to ^{60}Co are determined by measurement of known sources of those nuclides or are calculated from information on their decay schemes coupled with an experimental efficiency plot like Fig. 1. It is apparent that the presence of a high-energy impurity would have an effect proportionately larger than its relative concentration. Response varies with temperature, and, in addition, a long-term drift may take place so that it is necessary to adjust readings by use of normalizing sources of ^{226}Ra. Principles[52] of calibration and use have remained the same since early in the radioisotope program.[51]

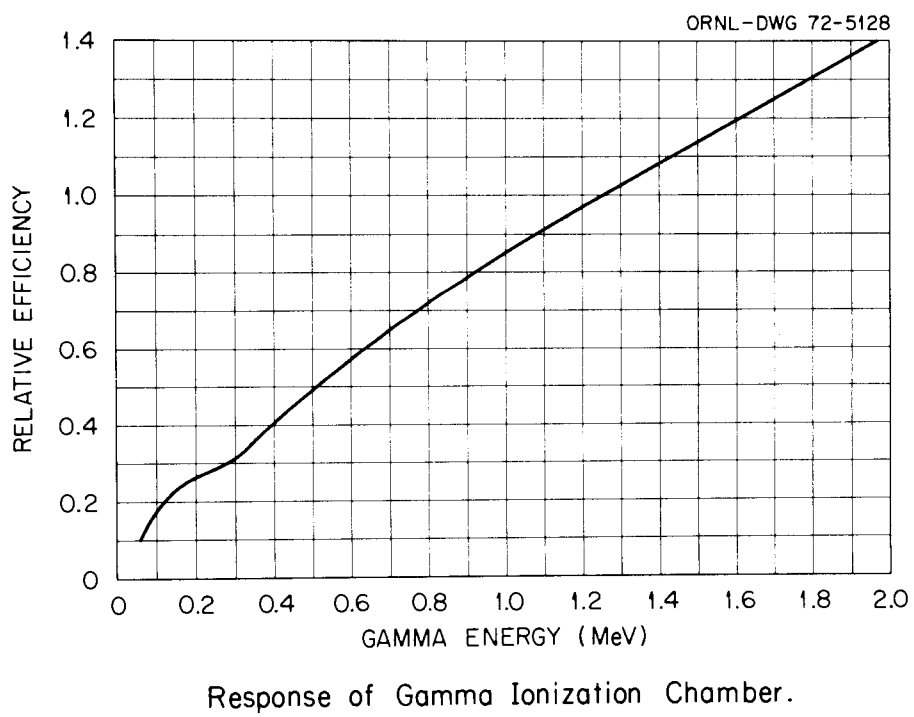

Fig. 1. Response of Gamma Ionization Chamber.

Quality Control

Quality of measurements is indicated by precision and accuracy (bias).[15] Precision is usually characterized by random variation, i.e., a "plus-or-minus" value, and can be evaluated by familiar techniques;[2,14,15,55,56] but accuracy implies an absolute, i.e., a genuine "known," which obviously does not exist. It has been pointed out[55,57] that there is no method of rigorously evaluating accuracy, since an unknown source of error may be present.

In case one has obtained a SRM or calibration from a reputable supplier, say NBS, the stated precision can be combined in quadrature with the user's own precision, and the supplier's estimate of bias then added.[55] Uncertainties in half-life[58] or decay scheme can normally be incorporated in the user's precision, provided they have been statistically evaluated.[2,41] Unwitting use of an incorrect value, presence of an unknown impurity, or improper chemical handling[11,59] could of course cause an unsuspected bias. If a standard is not available, a statistical model is difficult to apply. Response curves for instruments could be fitted and errors estimated, but such a procedure would be tedious and uncertain.

It was early recognized at ORNL that errors of activity measurements and ordinary analytical techniques would be small compared to inaccuracies in disintegration-rate determinations, and compared to the warranted[60] radioisotope-concentration uncertainties of ±3 to ±30% — typically ±10%. Therefore, systematic studies were not made, but enough information was collected to show that routine precision[25] was about 2 to 5%, depending on the characteristics of particular nuclides; selected examples are given in Table 2.

It was decided that taking aliquots by weight was not justified, i.e., the quick and simple volumetric techniques were adequate. Separate portions of samples are taken by two analysts, and limits were prescribed[26] for their agreement. Whenever feasible, measurements are made by instruments of different types, and all calculations are verified by supervisors. The data of Table 1 generally demonstrate good agreement with measurements by standardizing laboratories, and some details on individual nuclides are given in a later section.

Personnel

Responsibility for radioisotope analysis at ORNL is shared by a research group and a service group. Some development of methods, decay-scheme research, and special problems are done by the former; the routine work, actual calibrations of equipment, measurements of most standards, and formalization of methods (as in the *ORNL Master Analytical Manual*) are done by the latter. In addition, specialists are available in other divisions for consultation and special services. Analytical work is supervised by experienced people who have degrees in chemistry but generally little formal training in radiochemistry. However, all have participated in the author's survey course mentioned in an earlier

section. Most of the technicians (nonprofessional) have many years of experience in relevant work. All personnel now do other work, such as activation analysis, and all equipment is used for multiple purposes.

Table 2. Precision Values for Selected Routine Assays[a]

Nuclide	Instrument[b]	Precision (%) (95% C.L.)
Hydrogen-3	L	5
Carbon-14	L	5
Sodium-24	I,S	2
Phosphorus-32	P	2
Chlorine-36	L	3
Potassium-42	I,S	3
Calcium-47	GS	2
Chromium-51	I,S	5
Iron-55	X	10
Iron-59	I,S	2
Nickel-63	L	10
Arsenic-77	P	5
Krypton-85	GS	3
Ruthenium-103	GS	5
Ruthenium-106	P	3
Cadmium-109	GS	5
Cerium-144	P	4
Promethium-147	L	3

[a]W. S. Lyon, S. A. Reynolds, and E. I. Wyatt, *Nucleonics* 24(8), 116 (1966).

[b]Assay instruments:
GS — Gamma scintillation spectrometer
I — Gamma ionization chamber
L — Liquid scintillation counter
P — End-window proportional counter
S — Gamma scintillation counter
X — X-ray proportional counter

ASSAY OF SELECTED RADIONUCLIDES

Carbon-14

Carbon-14, the original radioisotope product, was understandably of particular concern. The first standards, in 1946, were obtained from L. D. Norris[61] of the ORNL Chemistry Division and were in the form of $BaCO_3$, as the products were and are. Assay was done by packing portions of sample and standard in wells in metal plates and counting by means of an end-window counter, interposing an absorber as necessary to limit counting rates.

Soon after an NBS SRM became available in 1950, the assay method was changed to one involving CO_2 evolution and measurement in a beta ionization chamber.[62,63] The NBS SRM was compared with the Norris material, and they were found to agree within 1%. In 1959, the present method[64] of liquid scintillation counting was adopted. NBS SRM's were purchased as needed, particularly as new types — benzoic acid in toluene, then hexadecane — were issued. Two recent measurements are listed in Table 1. Between purchases, calibration has been maintained by secondary standards made from products. Dilutions of sample and standard are made separately because use of the "standard addition" method (adding a portion of hexadecane standard to the sample mixture, then recounting) gave anomalous results, as reported by Moghissi[65] and (privately) by others.

A measure of the validity of the local activity measurements is given by occasional calculations[2] of the half-life from specific activity, consistently near the accepted "Cambridge average" of 5730 ± 40 years. A recent value[45,66] was 5736 ± 56 years.

Chlorine-36

Chlorine-36 was originally assayed by evaporating a neutralized sample, followed by routine beta counting[24,47,48] by means of an end-window counter. No external standards were used until the nuclide became available from NBS in 1962, but an early determination[67] by the author of the beta half-life yielded 0.26 ± 0.04 megayear against the presently accepted[41] 0.307 megayear, indicating probable accuracy of assay within 13% or better. Later, liquid scintillation counting was adopted; the data in Table 1 were obtained by this method.[68] Adjustment was made after the 1969 measurement, which must have been biased by accidental partial evaporation of a secondary standard.

Iron-55

Using calibrations based on coincidence-counted sources of ^{51}Cr, ^{54}Mn, and ^{65}Zn, the early measurements of ^{55}Fe were made by means of a G-M tube with krypton-ethanol filling.[69] Possible uncertainty in the measurement led to organization of an intercomparison[34] by ORNL in 1952, which showed adequate agreement. Subsequently, a proportional counter spectrometer[34,63] was used. An ^{55}Fe source was maintained for normalizing

spectrometer performance. Agreement to 1.6% was observed with an NBS SRM made available in 1961, and local measurement lay between NBS and IAEA in 1969 (Table 1).

Iron-59

Straightforward coincidence counting was used to calibrate GIC's and scintillation counters for routine measurements[70] of ^{59}Fe. Agreement with NBS (and Nuclear-Chicago Corporation) was demonstrated[38] in 1959-1960 and in 1970 (Table 1).

Nickel-63

Nickel-63 has a maximum beta energy of 0.066 MeV and is difficult to measure. For two decades, standardization[69] was based on evaluations[71] of efficiency and self-absorption in a 2π proportional counter. In 1960, liquid scintillation counting was adopted for routine measurement,[72] though the basis of calibration remained the same. Table 1 shows initial assay of a SRM in adequate agreement, but when ^{63}Ni became available from NBS,[73] the local measurement was, disappointingly, 17% higher than NBS. The calibration was, of course, adjusted to the NBS value. The early[71] and present[73] values of the half-life, 85 and 100 years, respectively, are in accord with the activity measurements.

Krypton-85

Little attention has been given radioassay of ^{85}Kr products, because they are normally dispensed on a volumetric basis after mass analysis. However, when NBS issued gamma standards of the nuclide in 1970, one was obtained and assayed by gamma spectrometry, with a result 1.7% higher then NBS. The ampul was retained as a reference source for spectrometer or GIC measurements.

Iodine-131

Because of its wide use in the biomedical field, ^{131}I was the first radioisotope product for which NBS began a standards program. Some difficulties were encountered in that the first NBS assays were made against RaDE (^{210}Pb-Bi), found later[21] to have a "forward scattering" correction of 7%. ORNL assays by coincidence counting were correspondingly higher.[1,21] Subsequently, NBS assays were improved by using 4π counting, and disagreement became insignificant[1,2] by 1951. Gamma ionization chambers and scintillation counters were calibrated for routine measurements.[74]

Promethium-147

Promethium-147 calibrations were made by 4π counting, and routine measurements were made on end-window detectors until 1954, when 2π proportional counters were adopted. Because of user difficulties, an intercomparison[75]

was arranged by ORNL in 1956. The ORNL measurement was 4.5% above the mean of six careful determinations, which had a standard deviation of 5%. Liquid scintillation counting[76] was begun in 1959, with reference solutions calibrated by 4π counting.

No external standards were obtained, but approximate confirmation of radioassays was afforded by use of weighed samples of known chemical analysis. In 1966, a discrepancy was observed, and an NBS SRM was obtained. Local measurements were found to be low, presumably because of evaporation of a reference solution. The data of Table 1 refer to measurements after making the calibration traceable to NBS. The "intercomparisons" were against samples prepared and weighed at Pacific Northwest Laboratories.

SUGGESTIONS FOR RADIONUCLIDE MEASUREMENTS

This section contains recommendations for those who make radionuclide measurements and may be producers or users of standards and calibrations, or both. Obviously the suggestions are not directed to specialists who are prepared to do a large variety of direct calibrations. Such capability is not practical for personnel of most laboratories, who typically deal with fewer than ten nuclides.[22] There is no single "best" approach to measurement capability and quality control. Circumstances such as level of funding, number and qualifications of personnel, and interactions with other parts of the organization may be determining factors. There have been no regulations governing radioassays, despite applications to radiopharmaceuticals used in humans, but imposed quality controls may come soon. There have been proposals for certification of laboratories and/or personnel (e.g., by NBS) and issuance of formal procedures for analysis.

Nuclear data is a perennial problem, often accounting for discrepancies in assays. The *Nuclear Data* journals contain the best compilations[40,41] and should be consulted first, but they necessarily lag behind current literature by as much as several years. If needed information is not found, the nuclear-medicine supplements[77] should be examined. Data in the *Table of Isotopes*[39] may be useful, but one must remember that none is newer than early 1966. Of course the recent literature contains much information, but it must be used cautiously because it is unevaluated. Correlations with earlier findings, and sometimes with theory, are desirable, and there must be compelling evidence for adopting a value which is very different from others. A convenient source for locating such papers, listed under the individual nuclide heading, is *Nuclear Science Abstracts*.

Requirements for Standards

The best form of SRM is a solution of the nuclide for which calibration is desired. This type is better than a mounted source, "point," disk, etc., because the user can prepare it in a form identical to the material to be measured, and possible systematic errors in the user's technique may be compensated. Volumetric techniques are sufficiently accurate for

most calibrations. If published efficiency curves must be used, a few points should be checked by means of single SRM's or mixtures.[78] For beta efficiencies, a better plot is obtained against average rather than maximum energy.[79] If a substitute or "mock" standard is required (such as ^{57}Co for ^{99m}Tc), it should be compared with a source of the pure nuclide under valid conditions (e.g., use of a spectrometer with small volumes of materials) and then the pure nuclide can be used to calibrate a working instrument.

For technical and statutory reasons, it is desirable to obtain SRM's and calibrations from NBS. A list of these is given in Table 3. In addition, NBS is permitted to calibrate other nuclides at cost, but that service is limited. If a particular service is unavailable or would be too much delayed, another supplier may be used. One should require certification equivalent to that recommended by NRC[58] and ICRU.[56] In general, the suggestions in the *Users' Guides for Radioactivity Standards*[59] should be followed; some chemical and physical considerations are given in later sections of this report.

Personnel

It is unrealistic to be dogmatic about types and qualifications of personnel in a radioassay group. An organization must decide what expertise and services the group must provide for itself, and what can be obtained in other groups, or even outside organizations (e.g., through service contracts with instrument companies), as well as funding and availability of the desired people.

Minimum qualifications might be as specified in an IAEA handbook[80] for a radiation-protection group, consisting of a director (supervisor), a physicist, technicians as needed, and a secretary. The handbook indicates that the supervisor should have a science or engineering degree, five years of directly relevant experience, and be familiar with statistical quality control. The physicist should have similar academic training and two years experience, while the technicians should have experience in laboratory work and it is "desirable" that it be in the subject field. ANSI[81] listed similar requirements for power plant personnel. Realistically, the supervisor, at least, should have training somewhat like that given radiopharmacists[82] (obviously with deletion of medical items), covering basic nuclear physics, radiochemistry, instruments and standards, health physics, etc. This should be supplemented by specialized courses or seminars, such as those taught by NBS and ORAU. Obviously one must keep up to date by reading the literature.

It has been suggested informally on at least two occasions that radioassay personnel be certified or licensed as are reactor[81] and fuel-plant[83] operators. Whether or not this is undertaken by a concerned agency (such as FDA for radiopharmaceuticals), laboratory management should make sure such work is being done competently and conscientiously, because of the grave responsibilities entailed.

Table 3. Standards and Routine Calibration Services of NBS[a,b]

Nuclide	Service[c]	Nuclide	Service[c]
Hydrogen-3	S	Niobium-94	P
Carbon-14	S	Niobium-95	C
Sodium-22	S,P,C	Cadmium-109	P
Sodium-24	C	Tin-113—Indium	C
Aluminum-26	S	Iodine-125	C
Phosphorus-32	C	Iodine-131	C
Sulfur-35	C	Cesium-137—Barium	P,C
Chlorine-36	S,C	Cerium-139	C
Potassium-42	C	Cerium-141	C
Calcium-45	C	Cerium-144—Praseodymium	S
Scandium-46	C	Promethium-147	S,C
Chromium-51	C	Tantalum-182	C
Manganese-54	C	Gold-198	C
Iron-55	S	Mercury-197	C
Iron-59	C	Mercury-203	C
Cobalt-57	S,C	Thallium-204	C
Cobalt-60	P,C	Polonium-210	P,C
Nickel-63	S	Radium-226	S,C
Zinc-65	C	Thorium-228	P
Krypton-85	Gas,P	Uranium-235	X
Strontium-85	C	Plutonium-238	P
Strontium-89	C	Plutonium-239	X
Strontium-90—Yttrium	C	Americium-241	P
Yttrium-88	P,C		

[a] *Standard Reference Materials*, NBS Spec. Publ. 260, Govt. Printing Office, Washington, D.C. (semiannually).

[b] *Calibration and Test Services of the National Bureau of Standards*, NBS Spec. Publ. 250, Govt. Printing Office, Washington, D.C. (1970) (revised pages and new editions as needed).

[c] C = calibration routinely available
P = point or plate source
S = solution standard
X = solid

Equipment

Much has been published[15,26,47] on apparatus for radioassay work. Ordinary laboratory equipment usually suffices. Radiation-protection provisions such as shielding and glove boxes may be needed, but less so than in facilities for processing or shipping major quantities of radioisotopes. The variety of measurement devices depends on the number and type of nuclides to be assayed. For hard-beta and gamma nuclides, an end-window counter, a gamma ionization chamber, and a NaI(Tl) or Ge(Li) spectrometer would suffice. (Some duplication for "standby" capability may be needed.) For soft-beta nuclides, a liquid scintillation counter is indicated. The same device might be used for alpha, or a 2π proportional counter might be chosen. A Si(Li) or proportional-counter spectrometer would be useful for low-energy x rays, although liquid scintillation has been used.[84]

Redundance is emphasized, especially in measuring the total activity dispensed in a secondary standard or a shipment. This can be done, in many cases, by use of a 4π ionization chamber or an external chamber, reading in dose units, and estimating content by use of the specific gamma-ray constant.[14,85]

Some Physical Sources of Error

Physical considerations in calibration have been generally discussed.[11,59] Assumptions of the wrong decay scheme or half-life clearly can cause error. "Drift" can be compensated by reading an appropriate normalizing source. If readings are made on a meter, typical "1%-of-full-scale" accuracy is not sufficient, forcing use of an expanded scale or "nulling."[52] Differences in activities of standard and unknown can lead to errors if the instrument is nonlinear and appropriate corrections are not made. Automatic live-timers sometimes malfunction and may not properly correct the data in any case.[86] The appreciable dead times of "fast" counters, typically 8 μsec[87] (i.e., about 1.3% correction at 100,000 counts/min), may be erroneously ignored. Various artifacts, such as scattering, spatial location of positron annihilation, and coincidence and random[2] summing, have been studied.[88] Summing may have both positive and negative effects by creating a spurious peak at high energy at the expense of one or more primary peaks. Conditions of use such as "geometry" (spatial relationship of source and detector) and absorption may not be made identical for standard and unknown.

The effect of a radionuclidic impurity may range from nil to very large, depending on the radiations of product and impurity and on the measurement method. Clearly, the effect may vary with time because of the different half-lives involved.

Chemical Considerations

Usual good laboratory practices must be followed, and special attention is required for items such as "contamination" and sorption, which are due in part to the extremely small masses of radioelements.[89] Manuals[14,59]

contain suggestions on chemical problems, and recent publications[90-92] give useful data. Volatility is encountered among noble gases, halogens, and some other elements such as mercury. Deposition from solution is a problem unless precautions are taken. Obviously troublesome are niobium, zirconium, rare earths, and like elements; but "soluble" sodium, rubidium, and cesium can deposit if exchange sites are afforded, e.g., glass exposed to a neutral solution. Sorption can often be minimized by adding carrier, complexing agent, or acid (for cations). "Wash-out" micropipets should be used to lessen deposition, as well as because of their accuracy.

Liquid scintillation counting is subject to interferences[93] such as quenching, chemiluminescence, and sorption, which are poorly understood and may not be anticipated in a particular case. It may be necessary to confirm measurements by a different method, to try various "cocktails," or to add carrier.

If radiochemical analysis is required (e.g., chemical separation of a minor impurity), approaches and procedures have been recommended.[26,94,95]

Quality Control

Standard quality-control methods[96] are applicable to radioisotope work; these include control charts, knowns, blind duplicates, and other devices. Intrinsic measurement precision, typified by counting statistics, must be taken into consideration[97,98] and is an essential factor in evaluating instrument performance or "constancy."[80] Other factors are usually dominant in overall accuracy;[56,57] examples are such additional sources of random error as pipetting, indeterminate ones involved with efficiency plots, and systematic errors in decay schemes or calibrations of standards. It is thus impossible to make rigorous statements about accuracy, but an estimate is made by combining precision terms in quadrature and adding the estimated systematic error.[56]

It is a mistake to assign all measurements a "blanket" warranty, e.g., "5%," since actual uncertainties vary widely.[80] Objective intercomparisons have revealed an almost universal tendency to underestimate errors, often by forgetting systematic sources.

Instruments must be calibrated when installed, when major repairs are made, or yearly in any case,[80] and performance must be tested with a reference source daily, or at every period of use. Redundance is desirable in measurement (by different types of instruments if feasible) and in calculation. A final check, e.g., with an ionization chamber or remote spectrometer, serves to detect mismeasurement or mistaken identity.

Participation in intercomparisons is urged, whether two- or multi-laboratory. Some organizations have set up very meaningful "round robins," including the NRC standards subcommittee,[13] the College of American Pathologists[99] (who arranged three in 1972), and the Society of Nuclear Medicine.[100] Traceability to NBS is an essential feature of these efforts and is now achieved through individual arrangements, but it is expected to

be handled through the Measurement Assurance Program[99] in the future. A group to study standard methodology for measurements was formed by the Atomic Industrial Forum and was recently transferred to ANSI N44.

One could continue with discussion of formalities of quality control, but the essential feature is integrity. No program can be successful without conscientious, intelligent work by concerned individuals.

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REFERENCES

1. S. A. Reynolds, *Rec. Chem. Prog.* 16, 99 (1955).

2. S. A. Reynolds, *Isotop. Radiat. Technol.* 4, 46 (1966).

3. "Cooperative Isotopes Program," in annual progress reports of the Analytical Chemistry Division, e.g., ORNL-4838, pp. 31-3 (1973).

4. Isotopes development program reports, e.g., AEC Report TID-4067, pp. 46-7 (1972).

5. A. F. Rupp, in *Nuclear Standards for Chemistry and Technology*, NBS Spec. Publ. 310 (1968).

6. S. A. Reynolds, *Health Phys.* 8, 391 (1962).

7. J. C. Posey, *Isotop. Radiat. Technol.* 1, 92 (1963).

8. S. R. Gunn, *Nucl. Instr. Methods* 29, 1 (1964) and 85, 285 (1970).

9. *Specification of High Activity Gamma-Ray Sources*, ICRU Report 18, Int. Com. Radiat. Units and Measurements, Washington, D.C. (1970).

10. Division of Applied Technology, *Isotopes Development Programs, Research and Development 1971*, TID-4067 (February 1972), pp. 134-5.

11. B. J. Wilson, ed., *The Radiochemical Manual*, 2nd ed., Radiochemical Centre, Amersham, England (1966).

12. M. R. Weiler and W. Y. Matsumoto, "Radionuclide Standards at the Pacific Northwest Laboratory," Gatlinburg Analytical Conference, Sept. 30, 1969.

13. B. Kahn, in *Nuclear Standards for Chemistry and Technology*, NBS Spec. Publ. 310 (1968).

14. *A Manual of Radioactivity Procedures*, Report 28, National Council on Radiation Protection and Measurements (NCRP), Washington, D.C. (1961); NBS Handbook 80 (revision in progress).

15. *Annual Book of ASTM Standards*, Part 30.

16. *Measurements and Standards of Radioactivity*, Nuclear Science Ser. Report 24, National Academy of Science—National Research Council Publ. 573, Washington, D.C. (1958).

17. *Metrology of Radionuclides*, International Atomic Energy Agency, Vienna (1960).

18. *Standardization of Radionuclides*, International Atomic Energy Agency, Vienna (1967).

19. W. S. Lyon, *Anal. Chem.* 36(2), 31A (1964).

20. S. A. Reynolds, *A Survey of Needs for Radioactivity Standards*, ORNL-TM-3239, Oak Ridge National Laboratory (1970).

21. G. G. Manov, *Status Report on Standardization of Radionuclides in the United States*, Preliminary Report 13, National Research Council; AEC Report NP-4664 (1953); also *Ann. Rev. Nucl. Sci.* 4, 51 (1954).

22. *National Uses and Needs for Standard Radioactive Materials*, National Academy of Science, Washington, D.C. (1970).

23. W. F. Libby, *Anal. Chem.* 29, 1566 (1957).

24. E. Segre, ed., *Experimental Nuclear Physics*, V. III, pp. 10-13, Wiley, New York (1959).

25. W. S. Lyon, S. A. Reynolds, and E. I. Wyatt, *Nucleonics* 24(8), 116 (1966).

26. *ORNL Master Analytical Manual* (especially Secs. 2 and 9), AEC Report TID-7015 and supplements, Oak Ridge National Laboratory (1957-67).

27. F. E. McKinney, S. A. Reynolds, and P. S. Baker, *Isotope User's Guide*, ORNL-IIC-19, Oak Ridge National Laboratory (1969).

28. W. B. Mann and H. H. Seliger, *Preparation, Maintenance, and Application of Standards of Radioactivity*, Circ. 594, NBS, Washington, D.C.; also *Int. J. Appl. Radiat. Isotopes* 1, 3, 215 (1956).

29. W. B. Mann and S. B. Garfinkel, *Radioactivity and Its Measurement*, Van Nostrand, Princeton (1966).

30. W. B. Mann and S. B. Garfinkel, eds., *Radioactivity Calibration Standards*, Spec. Publ. 331, NBS, Washington, D.C. (1970).

31. *Radioisotope Production and Quality Control*, STI/DOC/10/128, International Atomic Energy Agency, Vienna (1971).

32. W. S. Lyon and S. A. Reynolds, *Nucleonics* 14(12), 44 (1956).

33. P. J. Campion, in *Measurements and Standards of Radioactivity* Nuclear Science Ser. Report 24, National Academy Science—National Research Council Publ. 573, Washington, D.C. (1958); also *Int. J. Appl. Radiat. Isotopes* 4, 232 (1959).

34. W. S. Lyon and S. A. Reynolds, *Int. J. Appl. Radiat. Isotopes* 2, 80 (1957).

35. G. Goldstein, *Nucleonics* 23(3), 67 (1965).

36. *Standard Reference Materials*, NBS Spec. Publ. 260, Government Printing Office, Washington, D.C. (semiannually).

37. *Calibration and Test Services of the National Bureau of Standards*, NBS Spec. Publ. 250, Government Printing Office, Washington, D.C. (1970) (revised pages and new editions as needed).

38. W. S. Lyon, H. A. Parker, and S. A. Reynolds, in *Analytical Chemistry Division Annual Progress Report, December 31, 1961*, ORNL-3243, Oak Ridge National Laboratory, pp. 60-2.

39. C. M. Lederer, J. M. Hollander, and I. Perlman, *Tably of Isotopes*, 6th ed., Wiley, New York (1967).

40. Journal *Nuclear Data*, Parts A and B, published by Academic Press.

41. M. J. Martin and P. H. Blichert-Toft, *Nucl. Data* A8, 1 (1970).

42. *Nuclear Data Compilations*, National Academy of Science, Washington D.C. (1971).

43. G. Goldstein and S. A. Reynolds, *Nucl. Data* A1, 435 (1966).

44. S. A. Reynolds, in *Handbook of Analytical Chemistry*, L. Meites, ed., pp. 7-14 to 7-39, McGraw-Hill, New York (1963).

45. Latest publication: J. F. Emery, S. A. Reynolds, E. I. Wyatt, and G. I. Gleason, *Nucl. Sci. Eng.* 48, 319 (1972).

46. W. J. Price, *Nulear Radiation Detection*, 2nd ed., McGraw-Hill, New York (1964).

47. R. T. Overman and H. M. Clark, *Radioisotope Techniques*, McGraw-Hill, New York (1960).

48. *Annual Book of ASTM Standards*, Part 30, Method E 181; *ORNL Master Analytical Manual*, AEC Report TID-7015, Oak Ridge National Laboratory, (1957-67), Method 9 0733001.

49. F. Adams and R. Dams, *Applied Gamma-Ray Spectrometry*, 2nd ed., Pergamon Press, New York (1970).

50. R. L. Heath, *Scintillation Spectrometry Gamma-Ray Spectrum Catalogue*, IDO-16880, Phillips Petroleum Company (1964).

51. J. W. Jones and R. T. Overman, *The Use and Calibration of a 100% Geometry Ion Chamber*, AECD-2367, Oak Ridge National Laboratory (1948).

52. *ORNL Master Analytical Manual*, AEC Report TID-7015, Oak Ridge National Laboratory, (1957-67), Method 9 00360.

53. *ORNL Master Analytical Manual*, AEC Report TID-7015, Oak Ridge National Laboratory, (1957-67), Method 9 0733002.

54. J. W. G. Dale, W. E. Perry, and R. F. Pulfer, *Int. J. Appl. Radiat. Isotopes* 10, 65 (1961).

55. J. Mandel, *The Statistical Analysis of Experimental Data*, Interscience, New York (1964).

56. *Certification of Standardized Radioactive Sources*, ICRU Report 12, Int. Com. Radiat. Units and Measurements, Washington, D.C. (1968).

57. L. A. Currie, *Nucl. Instr. Methods* 100, 387 (1972).

58. S. B. Garfinkel, A. P. Baerg, and P. E. Zigman, *Certificates of Radioactivity Standards*, National Academy of Science, Washington, D.C. (1966).

59. B. Kahn, G. R. Choppin, and J. G. V. Taylor, *Users' Guides for Radioactivity Standards*, National Academy of Science, Washington, D.C. (1967) (revision in progress).

60. *Research Materials — Separated Isotopes, Radioisotopes, Special Preparations*, Oak Ridge National Laboratory (1973).

61. L. D. Norris and M. G. Inghram, *Phys. Rev.* 73, 350 (1948).

62. *ORNL Master Analytical Manual*, AEC Report TID-7015, Oak Ridge National Laboratory (1957-67), Method 9 0733172.

63. C. J. Borkowski, *Anal. Chem.* 21, 348 (1949).

64. *ORNL Master Analytical Manual*, AEC Report TID-7015, Oak Ridge National Laboratory (1957-67), Method 9 0733173.

65. A. A. Moghissi and M. W. Carter, *Anal. Chem.* 40, 812 (1968).

66. S. A. Reynolds, in *Analytical Chemistry Division Annual Progress Report, Sept. 30, 1968*, ORNL-4343, Oak Ridge National Laboratory, p. 77.

67. H. W. Wright, E. I. Wyatt, S. A. Reynolds, W. S. Lyon, and T. H. Handley, *Nucl. Sci. Eng.* 2, 427 (1957).

68. *ORNL Master Analytical Manual*, AEC Report TID-7015, Oak Ridge National Laboratory (1957-67), Method 9 0733201.

69. W. S. Lyon, S. A. Reynolds, and T. H. Handley, in *Analytical Chemistry Division Quarterly Progress Report, June 30, 1950*, ORNL-788, Oak Ridge National Laboratory.

70. *ORNL Master Analytical Manual*, AEC Report TID-7015, Oak Ridge National Laboratory (1957-67), Method 9 0733411.

71. A. R. Brosi, C. J. Borkowski, E. E. Conn, and J. C. Griess, Jr., *Phys. Rev.* 81, 391 (1951).

72. *ORNL Master Analytical Manual*, AEC Report TID-7015, Oak Ridge National Laboratory (1957-67), Method 9 0733541.

73. I. L. Barnes, S. B. Garfinkel, and W. B. Mann, *Int. J. Appl. Radiat. Isotopes* 22, 777 (1971).

74. *ORNL Master Analytical Manual*, AEC Report TID-7015, Oak Ridge National Laboratory (1957-67), Methods 9 0733391, 9 0733392, and 9 0733393.

75. S. A. Reynolds, in *Measurements and Standards of Radioactivity*, Nuclear Science Ser. Report 24, National Academy of Science—National Research Council Publ. 573, Washington, D.C. (1958), pp. 106-108.

76. *ORNL Master Analytical Manual*, AEC Report TID-7015, Oak Ridge National Laboratory (1957-67), Method 9 0733661.

77. L. T. Dillman, *J. Nucl. Med.* sup. 2 (1969) and 4 (1970) (revision in progress).

78. *SRM's* 4252 and 4253, bulletin from NBS (December 1972).

79. A. Ramaswami, S. P. Dange, S. Prakash, and M. V. Ramaniah, *Determination of Absolute Disintegration Rates of Beta-Gamma Emitters*, BARC-552, Bhabha Atomic Research Centre, Bombay (1971).

80. *Handbook on Calibration of Radiation Protection Monitoring Instruments*, STI/DOC/10/133, International Atomic Energy Agency, Vienna (1971).

81. *Selection and Training of Nuclear Power Plant Personnel*, ANSI N18.1-1971, American National Standards Institute, New York.

82. M. Tubis, in *Radiopharmaceuticals from Generator-Produced Radionuclides*, STI/PUB/294, pp. 3-9, International Atomic Energy Agency, Vienna (1971).

83. R. E. Brooksbank, D. J. Skovholt, and W. H. Lewis, *Licensing of Reprocessing Plant Operators*, ORNL-TM-2573, Oak Ridge National Laboratory (1969).

84. D. L. Horrocks, *Int. J. Appl. Radiat. Isotopes* 22, 258 (1971).

85. D. Nachtigall, *Table of Specific Gamma Ray Constants*, K. Thiemig, Munich (1969).

86. W. Filippone and F. J. Munro, *Nucl. Sci. Eng.* 47, 150 (1972).

87. C. B. Nelson, J. M. Hardin, and G. I. Coats, *Nucl. Instr. Methods* 97, 309 (1971).

88. W. S. Lyon, J. S. Eldridge, and P. Crowther, in *Radiochemical Methods of Analysis*, V. II, pp. 33-46, International Atomic Energy Agency, Vienna (1965).

89. I. M. Korenman, *Analytical Chemistry of Low Concentrations*, Israel Program for Scientific Translations, Jerusalem (1968).

90, K. Kepak, *Chem. Rev.* 71, 357 (1971).

91. E. P. Johnson and G. C. Lowenthal, *Int. J. Appl. Radiat. Isotopes* 23, 196 (1972).

92. A. G. Tuck and A. McNair, *Int. J. Appl. Radiat. Isotopes* 23, 395 (1972).

93. P. F. Davidson and L. P. Andersson, *Anal. Biochem.* 47, 253 (1972).

94. J. H. Harley, *HASL Procedures Manual*, HASL-300, Health and Safety Laboratory, New York (1972).

95. National Research Council Subcommittee on Radiochemistry, monographs on the radiochemistry of the elements, NAS-NS-3001 et seq., and radiochemical techniques, NAS-NS-3101 et seq. (1960--).

96. *ASTM Manual on Quality Control of Materials*, STP 15-C, ASTM, Philadelphia (1951).

97. H. E. Kolde, *Quality Control of Radioactivity-Counting Systems*, PHS Publ. 999-RH-15, Government Printing Office, Washignton, D.C. (1965).

98. E. G. Andresen, *Kerntechnik* 14, 226, 360 (1972).

99. L. M. Kushner, (NBS) *Tech. News Bull.* 56, 219 (1972).

100. S. Genna, *et al.*, *J. Nucl. Med.* 13, 285 (1972).

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