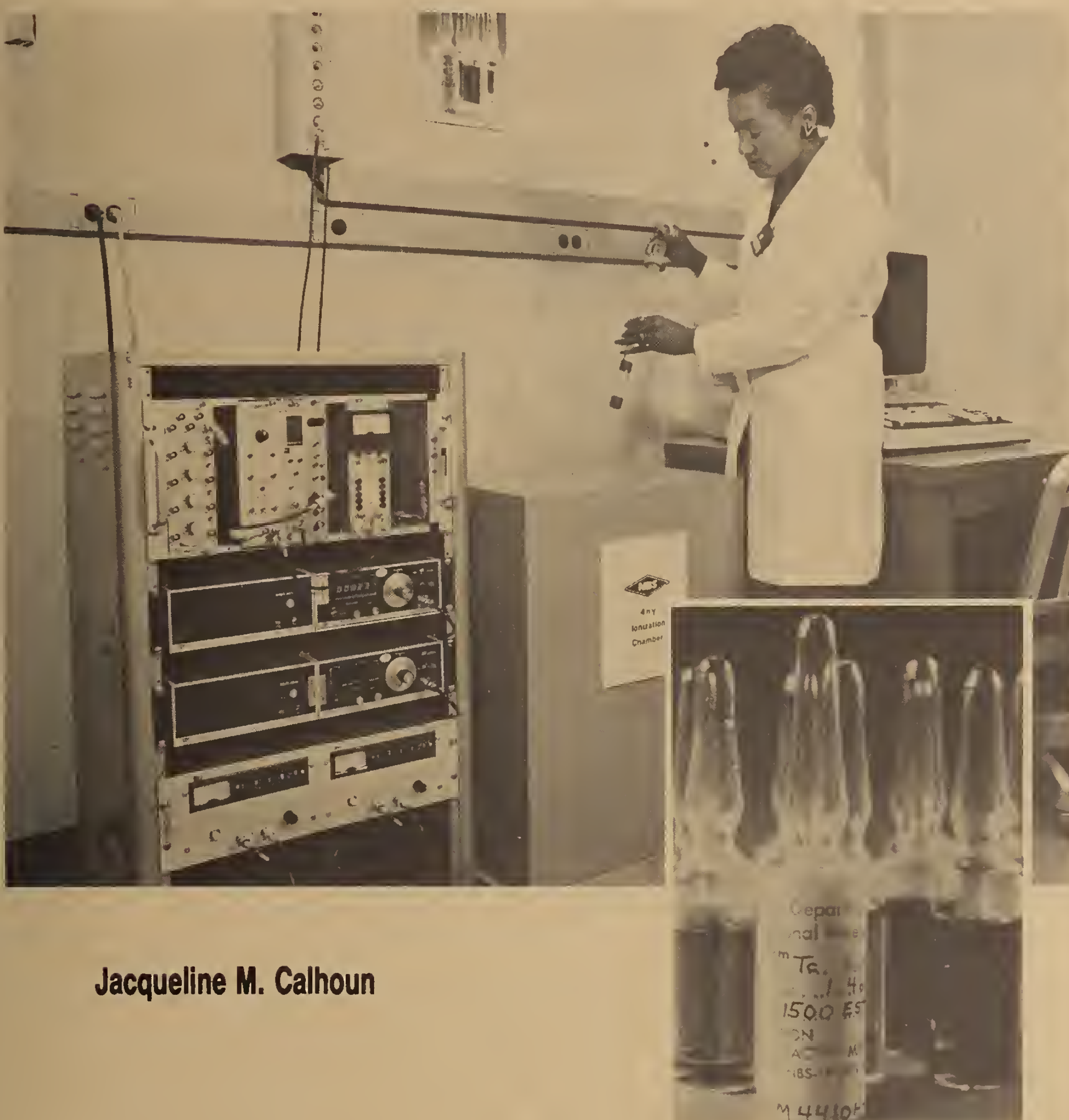
NBS Measurement Services:

Radioactivity Calibrations with the "4π" Gamma Ionization Chamber and Other Radioactivity Calibration Capabilities

NBS
Special
Publication
250-10



Jacqueline M. Calhoun

U.S. Department of Commerce
National Bureau of Standards

Center for Radiation Research

The Center for Radiation Research is a major component of the National Measurement Laboratory in the National Bureau of Standards. The Center provides the Nation with standards and measurement services for ionizing radiation and for ultraviolet, visible, and infrared radiation; coordinates and furnishes essential support to the National Measurement Support System for ionizing radiation; conducts research in radiation related fields to develop improved radiation measurement methodology; and generates, compiles, and critically evaluates data to meet major national needs. The Center consists of five Divisions and one Group.

Atomic and Plasma Radiation Division

Carries out basic theoretical and experimental research into the spectroscopic and radiative properties of atoms and highly ionized species; develops well-defined atomic radiation sources as radiometric or wavelength standards; develops new measurement techniques and methods for spectral analysis and plasma properties; and collects, compiles, and critically evaluates spectroscopic data. The Division consists of the following Groups:

- Atomic Spectroscopy
- Atomic Radiation Data
- Plasma Radiation

Radiation Physics Division

Provides the central national basis for the measurement of far ultraviolet, soft x-ray, and electron radiation; develops and disseminates radiation standards, measurement services, and data for for these radiations; conducts theoretical and experimental research with electron, laser, ultraviolet, and soft x-ray radiation for measurement applications; determines fundamental mechanisms of electron and photon interactions with matter; and develops advanced electron- and photon-based measurement techiques. The Division consists of the following Groups:

- Far UV Physics
- Electron Physics
- Photon Physics

Radiometric Physics Division

Provides national measurement standards and support services for ultraviolet, visible, and infrared radiation; provides standards dissemination and measurement quality assurance services; conducts research in optical radiation, pyrometry, photometry, and quantum radiometry; and develops spectroradiometric and spectrophotometric standards and calibration procedures. The Division consists of the following Groups:

- Spectral Radiometry
- Spectrophotometry
- Radiometric Measurement Services

Radiation Source and Instrumentation Division

Develops, operates, and improves major NBS radiation facilities including the electron Linac and race track microtron; develops, designs, and builds electronic and mechanical instrumentation for radiation programs and facilities; provides national leadership in the standardization of nuclear instrumentation; and develops new competence in radiation sources and instrumentation. The Division consists of the following Groups:

- Accelerator Research
- Linac Operations
- Electronic Instrumentation
- Mechanical Instrumentation

Ionizing Radiation Division

Provides primary national standards, measurement services, and basic data for applications of ionizing radiation; develops new methods of chemical and physical dosimetry; conducts theoretical and experimental research on the fundamental physical and chemical interactions of ionizing radiation with matter; provides essential standards and measurement support services to the National Measurement Support System for Ionizing Radiation; and develops and operates radiation sources needed to provide primary radiation standards, fields, and well-characterized beams of radiation for research on radiation interactions and for development of measurement methods. The Division consists of the following Office and Groups:

- Office of Radiation Measurement
- Radiation Theory
- Radiation Chemistry and Chemical Dosimetry
- Neutron Measurements and Research
- Neutron Dosimetry
- Radioactivity
- X-Ray Physics
- Dosimetry

Nuclear Physics Group

Engages in forefront research in nuclear and elementary particle physics; performs highly accurate measurements and theoretical analyses which probe the structure of nuclear matter; and improves the quantitative understanding of physical processes that underlie measurement science.

NBS MEASUREMENT SERVICES: RADIOACTIVITY CALIBRATIONS WITH THE "4π" GAMMA IONIZATION CHAMBER

AND OTHER RADIOACTIVITY CALIBRATION CAPABILITIES

Jacqueline M. Calhoun

Center for Radiation Research
National Measurement Laboratory
National Bureau of Standards
Gaithersburg, MD 20899

U.S. DEPARTMENT OF COMMERCE, Clarence J. Brown, Acting Secretary
NATIONAL BUREAU OF STANDARDS, Ernest Ambler, Director

Issued October 1987

Library of Congress Catalog Card Number: 87–619870

National Bureau of Standards Special Publication 250-10
Natl. Bur. Stand. (U.S.), Spec. Publ. 250-10, 41 pages (Oct. 1987)
CODEN: XNBSAV

Commercial products—materials and instruments—are identified in this document for the sole purpose of adequately describing experimental or test procedures. In no event does such identification imply recommendation or endorsement by the National Bureau of Standards of a particular product; nor does it imply that a named material or instrument is necessarily the best available for the purpose it serves.

U.S. GOVERNMENT PRINTING OFFICE
WASHINGTON: 1987

For sale by the Superintendent of Documents, U.S. Government Printing Office, Washington, DC 20402–9325

PREFACE

The calibration and related measurement services of the National Bureau of Standards are intended to assist the makers and users of precision measuring instruments in achieving the highest possible levels of accuracy, quality, and productivity. NBS offers over 300 different calibration, special test, and measurement assurance services. These services allow customers to directly link their measurement systems to measurement systems and standards maintained by NBS. These services are offered to the public and private organizations alike. They are described in NBS Special Publication (SP) 250, NBS Calibration Services Users Guide.

The Users Guide is being supplemented by a number of special publications (designated as the "SP 250 Series") that provide a detailed description of the important features of specific NBS calibration services. These documents provide a description of the: (1) specifications for the service; (2) design philosophy and theory; (3) NBS measurement system; (4) NBS operational procedures; (5) assessment of measurement uncertainty including random and systematic errors and an error budget; and (6) internal quality control procedures used by NBS. These documents will present more detail than can be given in an NBS calibration report, or than is generally allowed in articles in scientific journals. In the past NBS has published such information in a variety of ways. This series will help make this type of information more readily available to the user.

This document (SP 250-10), NBS Measurement Services: Radioactivity Calibrations with the "4π" Gamma Ionization Chamber and Other Radioactivity Calibration Capabilities, by J. M. Calhoun, is the tenth to be published in this new series of special publications. It describes the use of the NBS "4π" gamma ionization chamber to measure the activity of gamma-ray-emitting radionuclides against national standards (see test numbers 43010C and 43020D in the SP 250 Users Guide). It also reviews NBS capabilities for making direct radioactivity calibrations. Inquiries concerning the technical content of this document or the specifications for these services should be directed to the author or one of the technical contacts cited in SP 250.

The Center for Radiation Research (CRR) is in the process of publishing 21 documents in this SP 250 series, covering all of the calibration services offered by CRR. A complete listing of these documents can be found inside the back cover.

NBS would welcome suggestions on how publications such as these might be made more useful. Suggestions are also welcome concerning the need for new calibration services, special tests, and measurement assurance programs.

George A. Uriano
Director
Measurement Services

Chris E. Kuyatt
Director
Center for Radiation Research

ABSTRACT

This paper describes the use of the NBS "4π"γ ionization chamber — an instrument which provides an indirect method of comparing the activity (decays per second) of gamma-ray-emitting radionuclides with national standards for a routine calibration service by the National Bureau of Standards Radioactivity Group. A description of the chamber's construction and characteristics, the operational procedure, and the associated equipment is included.

A description of NBS capabilities for direct radioactivity calibrations is also presented. Many of these capabilities are used to establish calibration factors for the "4π"γ ionization chamber.

Key Words: activity; calibration; gamma ray; ionization chamber; radionuclides; radium reference source; standard

TABLE OF CONTENTS

LIST OF TABLES

LIST OF FIGURES

1. Introduction

1.1. General Information

The NBS "4π"γ ionization chamber (IC) is a well-type pressurized instrument which is used for measuring the activity (decays per second) of gamma- ray-emitting radionuclides. Scheduled calibration services for submitted samples of gamma-ray-emitting radionuclides are offered as a complement to the Standard Reference Material (SRM) program as described in NBS Special Publication 260 [1]. These calibration services are advertised in the NBS Special Publication 250 (SP250) [2] as the "old" 8.2C (for radionuclides with half lives greater than 15 days)and 8.2D (for radionuclides with half lives less than 15 days) which are now the "new" 43010C and 43020C, respectively. The range of activity for the gamma-ray-emitting radionuclides is 0.4 - 60 MBq, and the nominal uncertainty range is 0.7 - 3.4 percent (corresponding to 3 standard deviations). Special tests for beta-particle-emitting solution sources are also available. These special tests are advertised in the SP250 as the old 8.2Q, R, and presently 43060S and 43070S. The specifics of these special tests are not discussed in this paper, but general information is given about the direct calibration of the beta-particle emitting radionuclides in section 2, Design Philosophy and Theory.

The "4π"γ chamber uses calibration factors for each radionuclide previously established by direct methods of activity measurements (1,2). A description of the instrumentation used to make direct measurements is given in the section on Design Philosophy and Theory. Many IC calibrations have been checked with those of other national standardizing laboratories through the Bureau International des Poids et Mesures (BIPM).

The gamma-ray-emitting radionuclides which may be submitted to NBS for "4π"γ ionization chamber calibrations are:

Na-22	Fe-59	Mo-99	I-123	Ba-140	Au-195
Na-24	Co-60	Tc-99m	I-131	Ce-141	Hg-197
Sc-46	Ga-67	Cd-109	Ba-133	Eu-152	Au-198
Cr-51	Se-75	Ag-110m	Cs-134	Eu-154	Tl-201
Mn-54	Sr-85	In-111	Cs-137	Eu-155	Hg-203
Co-57	Y-88	Sn-113	Ce-139	Yb-169	Pb-203

1.2 Description of Service

1.2.1 Purpose and Results of Service. The calibration service for the gamma-ray-emitting radionuclides is available for the technical user who must make measurements consistent with national standards or who requires higher-accuracy calibrations than are available with commercial standards. When the calibration is completed, the technical user is provided with a report stating:

a) the principal radionuclide
b) reference time and date
c) method of calibration
d) certified value of activity
e) decay-scheme assumptions
f) assessment of radionuclidic purity
g) overall uncertainty

When the calibration service is completed, the technical users are notified, and the calibrated sample returned, if desired by customer. Short-lived radionuclide samples are usually measured by the customer only before transmittal to NBS, for the activity would be too low for measurement after the NBS calibration.

1.3 Specifications for Samples Submitted for Calibration in the NBS "4π"γ Ionization Chamber

1.3.1 Submitted Solution Specifications. Samples submitted for calibration must be chemically and physically stable. Tables 1 and 2 list the chemical forms which are suggested for these solution sources. Tables 1 and 2 also list the activity ranges and their associated nominal uncertainties.

The solution sample should consist of a specific radionuclide in 5.0 ± 0.2 milliliters of liquid flame-sealed in a NBS-type borosilicate-glass ampoule. The ampoule will be provided by the Radioactivity Group.

SPECIFICATIONS FOR CALIBRATION OF GAMMA-RAY EMITTING SOLUTIONS WHICH ARE SUBMITTED FOR CALIBRATION

Radionuclides having half lives greater than 15 days

TABLE 1

Radionuclide	Nominal "3σ" Uncer. of Ionization-Chamber Calibration[a]	Activity Range[b] (MBq)	Suggested Chemical Form[c] Carrier	Suggested Chemical Form[c] Solution
^{22}Na	1.6%	0.4–40	$NaCl$	1 M HCl
^{46}Sc	0.8%	0.4–40	$ScCl_3$	1 M HCl
^{51}Cr	1.0%	2–60	$CrCl_3$	0.5 M HCl
^{54}Mn	1.2%	2–60	$MnCl_2$	1 M HCl
^{57}Co	0.8%	2–60	$CoCl_2$	1 M HCl
^{59}Fe	1.4%	0.4–40	$FeCl_3$	1 M HCl
^{60}Co	0.8%	0.4–40	$CoCl_2$	1 M HCl
^{75}Se	2.4%	2–60	H_2SeO_3	1 M HNO_3
^{85}Sr	2.0%	2–60	$SrCl_2$	1 M HCl
^{88}Y	0.7%	0.4–40	YCl_3	1 M HCl
$^{109}Cd - ^{109m}Ag$[d]	1.7%	2–60	$CdCl_2$	1.3 M HCl*
$^{110m}Ag - ^{110}Ag$	0.9%	0.4–40	$AgNO_3$	1 M HNO_3
$^{113}Sn - ^{113m}In$	3.0%	2–60	$SnCl_2$ or $SnCl_4$	4 M HCl
^{133}Ba	1.5%	2–60	$BaCl_2$	1 M HCl
^{134}Cs	1.0%	2–60	$CsCl$	1 M HCl
$^{137}Cs - ^{137m}Ba$	1.5%	2–60	$CsCl$	1 M HCl
^{139}Ce	1.0%	2–60	$CeCl_3$	1 M HCl
^{141}Ce	2.0%	2–60	$CeCl_3$	1 M HCl
^{152}Eu	1.6%	0.4–40	$EuCl_3$	1 M HCl
^{154}Eu	0.8%	0.4–40	$EuCl_3$	4 M HCl
^{155}Eu	1.5%	2–60	$EuCl_3$	4 M HCl
^{169}Yb	2.5%	2–60	$YbCl_3$	0.1 M HCl*
^{195}Au	2.3%	2–60	$KAu(CN)_4$	$10 gL^{-1}$ KCN* $10 gL^{-1}$ KCl*
^{203}Hg	1.4%	2–60	$Hg(NO_3)_2$	0.1 M HNO_3

(a) The total estimated uncertainty will depend upon the activity level and chemical form.

(b) The source activity should be in the indicated range when it arrives at NBS. The calibration scheduling must be coordinated with the NBS technical contact.

(c) This information is based in large part on the NBS Standard Reference Materials for these radionuclides. For those radionuclides marked with an asterisk, the carrier should be discussed with the NBS technical contact.

(d) The calibration for $^{109}Cd - ^{109m}Ag$ is in terms of gamma-ray-emission rate rather than activity.

SPECIFICATIONS FOR CALIBRATION OF GAMMA-RAY SOLUTION SOURCES

Radionuclides having half lives less than 15 days

TABLE 2

Radionuclide	Nominal "3σ" Uncer. of Ionization-Chamber Calibration(a)	Activity Range(b) (MBq)	Suggested Chemical Form(c) Carrier	Solution
^{24}Na	0.8%	0.4–40	NaCl	1 M HCl
^{67}Ga	1.4%	0.4–40	$GaCl_3$	2 M HCl
$^{99}Mo - ^{99m}Tc$	1.6%	2–60	Molybdate	4 M HNO_3
^{99m}Tc	1.5%	2–60	No carrier added/ pertechnetate	Saline
^{111}In	1.3%	2–60	$InCl_2$	3 M HCl
^{123}I	1.5%	2–60	KI, Na_2SO_3	0.01 M LiOH*
^{131}I	1.3%	2–60	KI, Na_2SO_3	0.01 M LiOH*
$^{140}Ba - ^{140}La$	3.4%	0.4–40	$Ba(NO_3)_2$, La $(NO_3)_3$	1 M HCl
^{197}Hg	2.4%	2–60	$Hg(NO_3)_2$	0.1 M HNO_3
^{198}Au	1.3%	2–60	$KAu(CN)_4$	10 gL^{-1} KCN* 10 gL^{-1} KCl
^{201}Tl	1.9%	2–60	$Tl(NO_3)_3$	0.9 M HNO_3
^{203}Pb	1.7%	2–60	$PbCl_2$	0.5 M HCl

(a) The total estimated uncertainty will depend upon the activity level and chemical form.
(b) The source activity should be in the indicated range when it arrives at NBS. The calibration scheduling must be coordinated with the NBS technical contact.
(c) This information is based in large part on the NBS Standard Reference Materials for these radionuclides. For those radionuclides marked with an asterisk, the carrier should be discussed with the NBS technical contact.

1.3.2 Packaging. Packaging for all sources must be in compliance with the Department of Transportation (DOT) and Nuclear Regulatory Commission (NRC) regulations. Copies of these regulations may be obtained from Operations Division, Office of Hazardous Materials, Department of Transportation, Washington, DC 20590. Postal regulations prohibit mailing radioactive materials which require a caution label under DOT regulations.

1.3.3 NBS Shipping Address. All samples submitted for calibration are checked by the NBS Health Physics Group for radiation level and source integrity before transferring to the NBS Radioactivity Group for calibration. The following shipping address must be used:

National Bureau of Standards
Atten: Name of NBS contact
Health Physics (Radioactivity Group)
Quince Orchard and Clopper Road
Gaithersburg, Maryland 20899

2. Design Philosophy and Theory

2.1 Physical Principles and Restrictions

The function and status of the "4π"γ ionization-chamber calibration service can be better appreciated if the difficulties in maintaining national standards for the activity of radionuclides is considered. Each of the 80-100 radionuclides for which activity calibrations are sometimes required in the United States presents a separate challenge for a "direct" calibration, independent of existing standards. Such calibrations, discussed in a section to follow, are usually time-consuming and not applicable to routine submitted samples. Because radionuclides by definition decay, a comparative measurement against an existing, laboriously directly- calibrated, sample may not be possible after even a few days.

What is needed is a stable, easy-to-use instrument to serve as a repository once the direct calibration has been performed for a particular radionuclide. With due precautions, an ionization chamber serves this purpose if gamma rays of sufficient energy and probability are emitted as part of the decay process. Gamma-ray interactions in a gas, pressurized to maximize their probability, produce ion-electron pairs in proportion to the gamma-ray energy. A voltage between two electrodes causes these charges to be collected; the resulting current is also proportional to the number of decays per second, called the "activity" and assigned the name bequerel (Bq). One bequerel of a given radionuclide always produces the same current if all conditions are the same. Hence a calibration, or "K" value can be assigned for that radionuclide, on a particular ionization chamber.

As a precaution against changes in the chamber (loss of gas; movement of electrodes) or the sensitive electronics required to measure the small currents produced by the usual-activity samples, the current for the sample is measured relative to that from a long-lived-radionuclide sample which was also measured when the original "K" value was determined. To account for possible changes in leakage currents or differences in external radiation, "background" measurements with no sample are also interleaved with sample and reference source runs during a measurement series.

Samples which are too low in activity produce a current insufficiently different from background; samples which are too strong may produce so many ion pairs that recombination can occur before the electric field sweeps them away. Any change in container, solution nature, or density can cause changes in gamma-ray absorption. Sample location must always be the same; in order to achieve this, and to maximize the current, the chamber is made re-entrant so that the sample can be placed at the center (see fig. 1).

The chamber cannot distinguish between currents produced by different radionuclides. Hence all samples must be examined with calibrated gamma-ray-spectrometry systems, and the effect of any impurity radionuclides removed. The amount (activity) of the impurity present is deduced from the spectrometry measurements, but a calibration factor may not have been established for some uncommon radionuclides. In this case, a response (current) can be synthesized from the observed gamma-ray-emission rates at different energies and an ionization chamber "response" curve constructed from the measured response of radionuclides emitting one or two gamma rays of known probability per decay (see fig. 2a and 2b).

Obviously, the ionization chamber is not suitable for radionuclides which emit only alpha particles or electrons, or for which NBS direct calibrations of sufficient accuracy have not yet been performed. However, the chamber offers a useful means of propagating national standards for, at present, some 36 radionuclides, including many for which SRM's are not regularly available.

2.2 Direct Calibrations

The following excerpt is from an article [1] that summarizes most of the direct calibrations which have been performed at NBS, including those used in establishing the National Bureau of Standards IC calibration values. All methods and radionuclides are given to indicate other possible NBS services beyond those listed in SP250. The uncertainties given in the article, obviously different for each radionuclide, usually contribute the major uncertainty in a calibration certificate. A change in the method of specifying the uncertainties retains their "3σ" significance, as given in this article on pages 40-44. Detailed descriptions of the direct activity measurement techniques are given in Ref. [2].

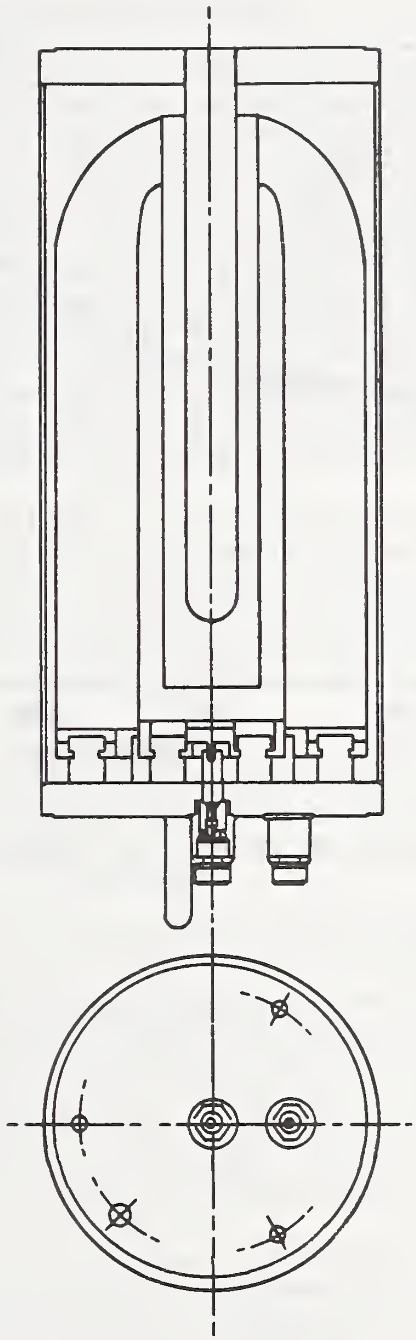


Fig. 1. Sectional view of a one-inch diameter reentrant-well IG = 11 pressure ionization chamber. (From Centronic, 20th Century Electronics Ltd., King Henry's Drive, New Addington, Croydon CR9 0BG, England.)

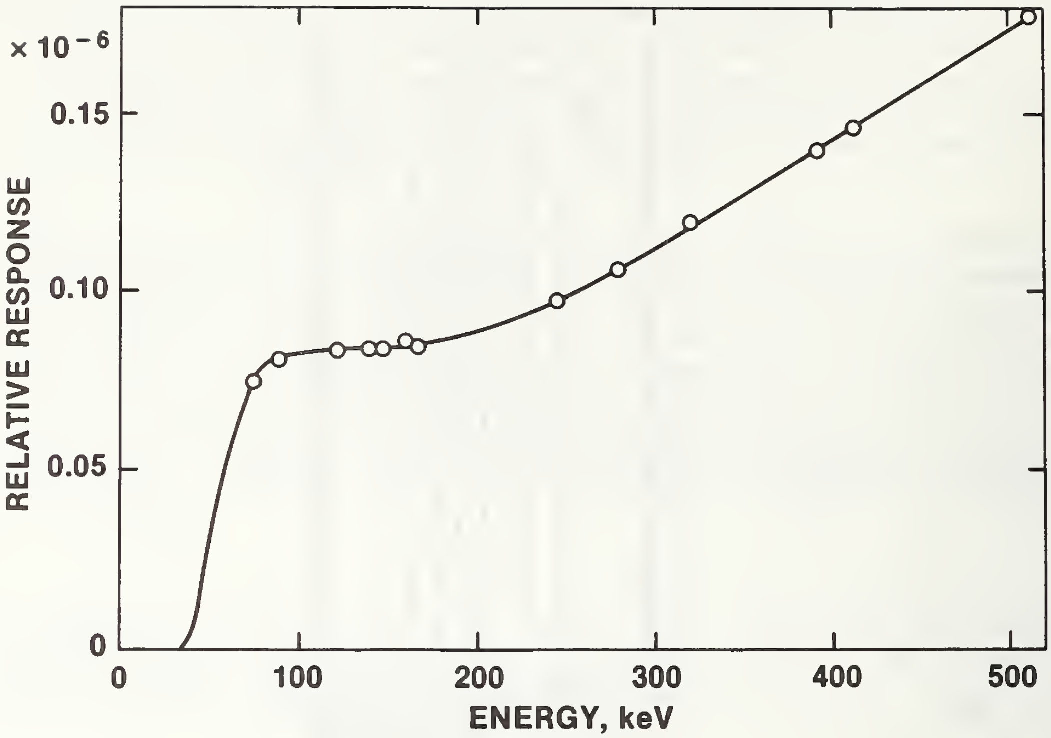


Fig. 2a. Relative Response of Chamber vs. Gamma-Ray Energy 0-500 keV.

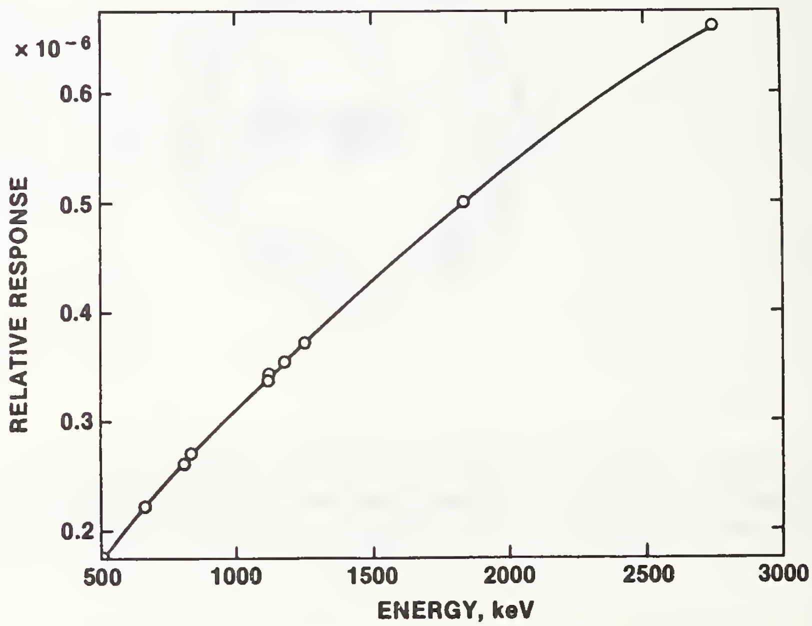


Fig. 2b. Relative Response of Chamber vs. Gamma-Ray Energy 500-3000 keV.

2.3 Comments on Direct Activity Measurements

All direct measurements depend to some degree on existing nuclear-decay-scheme data. For example, calorimetric measurements require a knowledge of the average energy per decay. The associated uncertainty for this method is trivial for some radionuclides emitting only alpha particles of a single energy which has been measured to five significant figures, but can be quite significant for beta decays for which the spectral distribution and maximum energy have not been measured well. Many of the methods developed for radionuclides with cascade radiations do not depend sensitively on details of the decay. In this article it is accepted that a requirement for nuclear data will not rule out a method being called direct if the sensitivity to those data, and the certainty of the values, will permit an acceptable overall uncertainty. Usually at least one nuclear datum, the half life, is required if the measured value is to have significance at any earlier or later time.

It should also be noted that all of the direct methods assume that no impurity radionuclide is present. This can be tested by repeated measurements over an appreciable fraction of a half life, but in practice a careful search for impurities by other means is a prudent first step in any calibration. Another very important topic not discussed here is the quantitative preparation of suitable and stable sources and solutions.

Techniques discussed here are assigned, somewhat arbitrarily, to one of two general classes: (1) those where the detector efficiency can be calculated from geometrical and physical information, without the use of information obtained from cascade radiations; and (2) those where the cascade radiations form the basis of the method.

2.4 Nuclear Radiations and Direct Activity Measurements

In order to relate the counting rate or current from a detector to the activity of a sample of a radionuclide being measured without use of a reference standard, several points usually have to be addressed: (1) What is the radiation to be detected, what is its energy, what is its probability per decay, and what is the effect of other radiations?; (2) What is the effect of the source matrix and support?; and (3) What fraction of the emitted radiations interact in the detector and what is the response?

Some of the methods discussed later can bypass these questions, but to see the difficulty in devising methods to answer the questions with sufficiently small uncertainty, it is useful to consider the properties of different radiations. Alpha particles are absorbed readily in thick sources and are accompanied by nuclear recoils that can cause confusion, but they have large and unique energies, backscatter from supports only at near-grazing angles, and can be restricted by a defining aperture with little contribution from edge effects. Beta particles are emitted with a spectrum of energies from zero to some characteristic maximum energy, with energy-dependent absorption or scattering in sources or supports. Positrons share these characteristics and produce annihilation photons. Positron decays also have electron-capture decays competing. Electron capture, which happens even when there is insufficient energy for positron emission, can occur in different atomic shells, and the subsequent atomic deexcitation produces both characteristic x rays and Auger electrons.

Subsequent to the primary decay, excited levels in the daughter nuclide then decay by gamma-ray emission or by internal conversion. The latter process also gives rise to x rays and Auger electrons. Long-lived excited states can contribute extra counts for the fraction of decays occurring beyond the resolving times of counting circuits. Radionuclides decaying only by isomeric transitions, without alpha or beta radiations, must also be calibrated.

The many types of radiations, and the large range of energies and half lives encountered, suggest that there is no universal method for direct calibrations, but rather that each radionuclide must be considered as a separate challenge. Also, most standardizing laboratories consider it prudent to check a calibration with an alternate method wherever possible. The reduction in the uncertainty of nuclear-decay data in recent years, together with improvements in technique, make measurements with efficiency-calibrated germanium-spectrometry systems a common indirect way of making a final check for many radionuclides.

2.5 Direct Methods Using Calculated Efficiencies

Methods using calculated efficiencies can be divided into those for which the efficiency is very high ("4π" methods) and those for which the efficiency is smaller, usually because the solid angle is appreciably smaller, but where the latter can be specified with sufficient accuracy ("defined-geometry" methods). Both methods carry a requirement that at least the sum of the probabilities per decay for each of the radiations to be detected must be known with an acceptable uncertainty. Both usually require some extrapolation or correction for events too low in energy to be separated from system noise.

2.5.1 High-Efficiency Counters. Two instruments commonly used for "4π" measurements are gas-proportional counters and liquid-scintillation counters. In the former, attenuation in the source and backing materials is the usual limitation for solid sources. In the latter, difficulties may arise from extrapolation complexity, resolution limitations, "quenching" (reduction of light output with the addition of the radioactive material), and the sensitivity to radiations other than the ones desired.

For radionuclides emitting beta particles of sufficiently high energy, 4π proportional counters can yield results with a satisfactory uncertainty. Suitable precautions in preparing thin conducting support films and thin sources must be observed.

For alpha particles, liquid-scintillation counting avoids the problem of energy loss in a support, but any energetic electrons may interfere with a measurement because of the much higher light output for a given energy absorption relative to alpha particles. If such interferences are taken into account, simple liquid-scintillation counting can be quite effective.

2.5.2 Calorimetry. If the average energy per decay for a radionuclide is known and almost all of this energy can be transformed into heat in a calorimeter, measurements of low uncertainty are possible, even for radionuclides emitting low-energy beta particles.

2.5.3 Internal Gas Counters. Radionuclides in the gaseous form can be introduced directly into the counting gas of proportional counters. With precautions for quantitative mixing and handling, and compensation for boundary effects, low uncertainties can be obtained even for rather low-energy beta spectra.

2.5.4 Defined-Geometry Counters. Counters with a 2π geometry are widely used in routine counting, but backscattering from a thick backing often makes them unsuitable for a direct measurement of beta particles, except under special circumstances. However, in the case of alpha particles, quantitative investigations of source and backing scattering have provided sufficient information so that an activity can be deduced from a 2π alpha-counting rate with moderate accuracy. The predominant emission of backscattered alpha particles almost parallel to the backing suggests, however, that more restricted geometries will give results of lower uncertainty.

Defined-geometry counters with restricting apertures are not usually considered suitable for electron counting because of the ill-defined scattering from edges. They are suitable for emission-rate measurements of low-energy photons, but the large uncertainties in probabilities per decay usually prohibit conversion into useful activity values.

2.6 Direct Methods Using Cascade Radiations

The methods discussed so far are most often used for radionuclides without appreciable branching to excited nuclear levels. However, more complex decays can give enough information to yield efficiency values for the detectors as part of the activity measurement. The essence of most methods is that not only are two radiations detected, each with some unknown efficiency, but the simultaneous detection of both (which depends on the product of efficiencies) is also recorded or inferred. With three measured quantities expressed as functions of the efficiencies of the two detectors and the activity of the sample, enough information exists for the three unknowns to be calculated. As with the previously discussed counting methods, the effects of backgrounds, dead-times, and pulse pileup must be considered, but with cascade radiations there may also be timing and resolving-time considerations.

2.6.1 Coincidence Counting. For a simple alpha-gamma or beta-gamma cascade, with each of two detectors sensitive to one radiation and not the other, corrections and analysis are straightforward. Many systems using proportional or liquid-scintillation counters to detect the particles, and NaI(Tl) or germanium detectors for photons, have been described.

Usually analysis is simplified if the detector for the particles has a high efficiency, but sometimes the circumstances require other choices. The reduction in gamma-ray rate due to branching from a level, or to internal conversion, is incorporated into an apparent detection efficiency, but explicit corrections must be made for other branchings from the level originating the cascade. Extra counts in the particle detector due to the detection of gamma rays or internal-conversion electrons must be taken into account. The effect of dead times for moderate counting rates was treated approximately for many years with small error, but recent formulations allow accurate correction even at high rates.

2.6.2 Anticoincidence Counting. If the efficiency of one detector is very high, then it is reasonable to count those pulses from the other detector which do not have a coincident pulse. As applied, this method has the technical advantage that the timing is not critical and the effect of long-lived states, which is difficult to measure by the coincidence method, can be measured directly.

2.6.3 Anticoincidence and Coincidence Counting with Efficiency Extrapolation. One must normally correct for other radiations that can produce counts in the beta detector when a beta particle is not detected. As the efficiency for detection of the beta particle (or positron or Auger electrons and x rays) approaches unity, however, these corrections go to zero. Thus one can introduce solids into the source or add absorbing films to vary the efficiency (as measured by the ratio of the coincidence rate to the gamma-ray rate) to obtain an experimental function of beta rate versus efficiency. This function can then be extrapolated to 100-percent efficiency, and corrections for the extraneous counts no longer have to be made explicitly.

If the pulse height from the particle detector is monotonically related to the energy of the particle detected, as it can be for liquid scintillators and high-pressure gas proportional counters, pulse-height discrimination can be used to vary the efficiency.

For a simple decay, without alternate beta branches or other complications, the extrapolation function of beta-particle count rate versus efficiency should be a straight line. In general the extrapolation function is taken to be a simple polynomial, with the extrapolated value considered more reliable if a straight line gives a satisfactory fit. For complex decays, explicit corrections may yet have to be made for differences in branch decay properties in the extrapolation region. Radionuclides which have no coincident radiations can be "efficiency traced" with quantitative amounts of another radionuclide which has a similar particle spectrum, but with coincident gamma radiations.

Efficiency extrapolation has become the method of choice for radionuclides to which it can be applied. In an international intercomparison of ^{134}Cs activity measurements organized by the Bureau International des Poids et Mesures (BIPM) in 1978, the total range of values from 22 laboratories was only 0.7 percent [5].

2.6.4 Sum-Peak Counting. This method, applicable to simple two- or three-photon cascades, also does not use coincidence or anticoincidence circuitry. The radiations are detected in the same spectrometry system, with the probability of the simultaneous detection of one of each energy equal to the product of the efficiencies for detecting each radiation singly. The result is a spectral peak at the sum of their energies. The areas of the single-photon and sum peaks are used to deduce directly the activity.

2.6.5 High-Efficiency NaI(Tl) Counting. Although individual probabilities per decay and detection efficiencies for gamma rays and x rays in cascade decays may not be known with sufficient accuracy, the probability of not detecting any radiation from a decay in large well-type NaI(Tl) detectors may be small enough that a calibration of suitable uncertainty is possible.

2.7 Radioactivity Group Instrumentation at the U.S. National Bureau of Standards

Some 40 counting systems are in use by the NBS Radioactivity Group for the direct calibration of radionuclides, checking for impurities, retaining calibrations, counting special samples, measuring half lives and the probabilities per decay of radiations, and intercomparing samples of production SRMs. Some of these systems have been described in the past or are similar to instruments described by others. In many cases the detector or source-handling portions were constructed at NBS, but in general commercial modular electronics are used, with provision for direct or indirect transfer of data to a minicomputer for analysis.

Table 3 lists and describes some of the equipment currently used for establishing basic calibrations without using cascade radiations. Table 4 is a similar list for instruments using cascade radiations.

2.8 Present Basic Radionuclide Calibrations at NBS

Finally, Table 5 lists most of the radionuclides for which the NBS Radioactivity Group has developed calibrations in the past three decades, together with the methods used, the estimated uncertainties as given on recent certificates, and the conformity with results from other national standardizing laboratories in many cases. NBS uncertainties in Table 5 are given as the sum of half of the 99-percent confidence interval of the mean, plus the linear sum of estimates of the limits of suspected systematic uncertainties [4].

Some radionuclides have been compared bilaterally with other nations, but for gamma-ray-emitting radionuclides the ionization chambers maintained at the Bureau International des Poids et Mesures have proven an efficient mechanism. NBS has 27 radionuclides recorded on these chambers at the present time. The uncertainties shown for the international intercomparisons in Table 5 are the simple estimated standard deviations of the means.

3. Principles of Operation

3.1 Construction of the NBS "4π"γ Ionization Chamber

The detecting element is a "4π" (re-entrant) cylindrical ionization chamber in which the current produced by gamma rays from the sample is compared with current from long-lived reference sources (see fig. 1). The chamber used for most of the calibration services was constructed at the Chalk River Nuclear Laboratories of Atomic Energy of Canada, Limited, but a commercial version from 20th Century Electronics, Ltd. in England is used in a later automatic-sample-changing system on which basic NBS calibrations are also being stored. The automatic chambers may later be used in the calibration service in the same way as the present manual chamber.

All chambers are filled with 20 atmospheres of dry argon. A 4-inch-thick lead housing surrounds the chamber presently used for service calibrations to reduce environmental background effects and prevent a change of efficiency from movable scattering material in the vicinity. The voltage for this chamber is 600 volts. A rigid tight-fitting holder made of low-Z material is used for the samples and reference sources to insure reproducibility of position.

TABLE 3 INSTRUMENTS FOR NBS BASIC CALIBRATIONS NOT USING CASCADE RADIATIONS

Counting System	Label	Description and References
$0.1\pi\alpha$ scintillation counter	$0.1\pi\alpha$	A carefully measured external baffle about 11 cm in diameter establishes the solid angle subtended by a CaF(Eu) scintillator in an evacuated tube. As with the following alpha-particle-counting systems, extrapolation to zero energy is used to account for the small fraction of the alpha spectrum masked by noise or the detection of recoil nuclei or other radiations.
0.8π Robinson α scintillation counter	$0.8\pi\alpha$	The solid angle subtended by a CsI(Tl) scintillator greater than 15cm in diameter is limited by a precise hole in a diaphragm and by a central stepped cone. The latter minimizes corrections for source size (up to 1.6-cm diameter) and displacement. Intervening air is replaced by hydrogen to reduce energy losses.
$2\pi\alpha$ proportional counter	$2\pi\alpha$	A 12-cm-diameter hemispherical counter with a wire-loop electrode is used for measuring the alpha-particle-emission rate of thin sources up to 9 cm in diameter. Measured and calculated effects of source thickness permit calculation of activity from alpha-particle-emission rate.
Liquid scintillation counters	LSa	A commercial sample-changing system with two refrigerated phototubes viewing 10-ml vials. Outputs of the two tubes are summed, with a coincidence requirement. Pulses are sent through an external amplifier and then analyzed.
	LSb	A commercial sample-changing system with refrigerated phototubes, logarithmic amplifier, and microprocessor control and analysis.
	LSc	A refrigerated NBS-built system with the sample in a quartz spectrophotometer cell optically coupled to a phototube on each end.
	LSd	The "LS Cascade" system but without gamma-ray-detection requirements.

Table 3 cont.

4π proportional counter	$4\pi\beta$	A metallized film carrying a thin source forms the boundry between two hemispherical proportional counters, each with a loop electrode. It is used primarily for high-energy pure-beta-ray-emitting radionuclides.
Pressurized 4π proportional counter	PPCX	Similar to the above 4π proportional counter, but modified for pressures of a few atmospheres. It is used for measuring x-ray-emission rates of pure electron capture radionuclides of low atomic number.
Radon pulse-ionization chambers	Radon PIC	Radon gas is quantitatively flushed from radium solutions or introduced from a collection flask. Water and oxygen are removed, and the gas transferred to one of four 3.6-l cylindrical pulse-ionization counters.
Microcalorimeter	Cal	A twin-cup radiation balance is used to compare compact similar sources. Radionuclides with radiations that can be totally absorbed are compared with a Peltier current.
Internal-gas counters	Gas	Radioactive gases are quantitatively mixed with counting gas and introduced into a common chamber containing 3 copper-walled proportional counters of different length and a comparable set of 3 counters with walls of stainless steel. Spectral extrapolation of the difference in counting rates for counters of different length is used.
Defined-geometry system for low-energy photons	Photon Def. Geom.	A 0.015-cm-thick Pt diaphragm with a hole 0.9505 ± 0.0020 cm in diameter quantitatively establishes the area subtended by a 1.6-mm-thick NaI(Tl) crystal with a 0.13-mm-thick Be window. Carefully measured source positions are arranged in a vacuum housing containing the apparatus. The emission rate of photons between 5 and 40 keV can be measured with an uncertainty of less than one percent.
Pin-well NaI(Tl)	Pin Well	Two-inch-diameter NaI(Tl) detectors with thin-walled wells or central passages less than 1 cm in diameter are used for measuring photon-emission rates in the energy region of 50 to 150 keV.

TABLE 4 INSTRUMENTS FOR BASIC NBS CALIBRATIONS USING CASCADE RADIATIONS

Counting System	Label	Description and References
Thin-crystal scintillation system	Thin-Xtal Cascade	A NaI(Tl) crystal 1.6mm thick is used with a thin plastic scintillator for measurement of gamma rays in coincidence with low-energy beta rays, or in summation with another thin NaI(Tl) crystal for sum-peak counting of ^{125}I.
Large sodium iodide system	8" NaI	Two 8" by 4" NaI(Tl) crystals with shallow wells are brought together to form a system for sum-peak or high-efficiency cascade counting.
Liquid scintillation system	LS Cascade	A hemisphere containing a liquid-scintillation cocktail is optically coupled to the phototube on which it sits. A 4" NaI(Tl) crystal with a 2" well is inverted over the hemisphere. Coincidence or anticoincidences are used to measure the efficiency for selected beta or electron-capture branches as a function of energy.
Early atmospheric proportional counter - gamma-ray coincidence system	PC-γ	A spherical counter has opposing wire loops in top and bottom, with the source at the center plane on a gold-coated collodion film at ground potential. The 2-in x 2-in NaI(Tl) crystal is external to the counter.
Atmospheric proportional counter system with source changer	PC Wheel	Two "pill-box" proportional counters operating at atmospheric pressure are separated by a thin metallized film carried in a slide. Shielded 3" x 3" NaI(Tl) gamma-ray detectors are above and below the proportional counters. Coincidences are recorded. An automatic sample changer inserts in turn one of 30 samples. Cover films are added for efficiency extrapolation.
Pressure-proportional counter system	PPC Cascade	Two "pill-box" proportional counters operating at pressures of about 1.5 MPa are separated by a slide carrying a metalized thin film on which the source is deposited. Shielded 3" by 3" gamma-ray detectors are above and below the proportional counters. The system is operated in the coincidence and anticoincidence modes, with discriminator efficiency variation for extrapolation.

TABLE 5 RADIONUCLIDE MEASUREMENT CAPABILITIES IN THE RADIOACTIVITY GROUP AT THE U.S. NATIONAL BUREAU OF STANDARDS

Radio-nuclide	System Label of Table 2 or 3	Typical+ Uncertainty (%)	International Check: S_m of others (%)*	International Check: NBS deviation from mean (%)	Remarks, References
^{22}Na	PC Cascade	1.6	0.1	−0.36	
^{24}Na	LS Cascade	0.8			
^{26}Al	8" NaI Sum	1.1			
^{46}Sc	LS Cascade	0.8	0.1	+0.05	
^{51}Cr	PPC Cascade	0.9	0.1	+0.14	
^{54}Mn	PPC Cascade	1.2	0.1	−0.06	
^{55}Fe	Photon Def. Geom.	2.7	1.0	+0.13	X-ray-emission rate
^{56}Mn	PC-γ	----			
^{57}Co	PPC Cascade	0.8	0.1	+0.74	
^{59}Fe	PC Wheel	1.4	0.1	+1.03	
^{60}Co	PPC Cascade	0.8	0.04	+0.18	
^{63}Ni	Cal	1.0	~1	−0.7	
^{67}Ga	PPC Cascade	1.5	0.16	−0.05	Delayed state
^{75}Se	PC-γ	2.6			Delayed state
^{85}Kr	Gas	0.9			
^{85}Sr	Photon Def. Geom	1.5	0.16	+0.08	
^{88}Y	PPC Cascade	0.5	0.14	−0.25	

Table 5 cont.

Radio-nuclide	System Label of Table 1 or 2	Typical+ Uncertainty (%)	International Check: S_m of others (%)*	International Check: NBS deviation from mean (%)	Remarks, References
^{94}Nb	8" NaI Sum	1.5			
^{99}Mo	PPC Cascade, PC wheel	1.6	1	+0.49	
^{99m}Tc	PP-1C Cascade, PC wheel	1.0			
^{108m}Ag	8" NaI	1.5			Electron capture decay only
^{109}Cd	Pin Well, Photon Def. Geom	2.0			X-, γ-ray-emission rates
^{110m}Ag	PC wheel	0.7			
^{111}In	PPC Cascade	1.3	0.13	+0.50	
^{113}Sn	NaI, Ge	2.4			Gamma-ray-emission rate
^{121m}Sn	Ge	2.8			Gamma-ray-emission rate
^{123}I	PPC Cascade	1.4			
^{125}I	Thin-Xtal Cascade	1.7			
^{125}Sb	PPC, LS Cascade	2.0	2	+1.9	Only gamma-ray-emission rates
^{127}Xe	Gas	1.5			
^{131}I	PPC Cascade	0.7	0.20	+0.03	
^{131m}Xe	Gas	3.3			

Table 5 cont.

Radio-nuclide	System Label of Table 1 or 2	Typical[+] Uncertainty (%)	International Check: S_m of others (%)*	International Check: NBS deviation from mean (%)	Remarks, References
^{133}Ba	PPC Cascade	1.4	0.05	+0.95	
^{133}Xe	Gas	1.3			
^{134}Cs	LS Cascade, PPC Cascade	0.7	0.11	+0.29	
^{137}Cs	PPC Cascade	1.4	0.24	+0.20	Efficiency traced with ^{134}Cs
^{139}Ce	PPC Cascade	0.7	0.14	+0.29	
^{140}Ba	PPC Cascade	2.8			With ^{140}La in equilibrium
^{140}La	PPC Cascade	1.5			
^{141}Ce	PC−γ	2.0			
^{147}Pm	4πβ	2.5			
^{152}Eu	8" NaI	1.5	0.3	−0.43	
^{154}Eu	PPC Cascade, LS Cascade	0.6	1.0	−0.81	
^{155}Eu	LS Cascade	1.4			
^{169}Yb	PPC Cascade	1.8	1.3	+0.83	Delayed state
^{195}Au	PPC Cascade	1.7			
^{197}Hg	PC−γ	2.0			
^{198}Au	PC−γ	1.3	0.2	+0.31	

Table 5 cont.

Radio-nuclide	System Label of Table 1 or 2	Typical+ Uncertainty (%)	International Check: S_m of others (%)*	International Check: NBS deviation from mean (%)	Remarks, References
^{201}Tl	PPC Cascade	1.8			
^{203}Hg	PC-γ	1.0	0.08	+1.04	
^{203}Pb	PPC Cascade	1.7			
^{204}Tl	$4\pi\beta$	1.0	0.6	+0.5	
^{207}Bi	8" NaI Sum	1.5			
^{222}Rn	Radon PIC				
^{226}Ra	Cal	1.2			

+ Usually half the 99-percent confidence interval of the mean plus the linear sum of estimates of suspected systematic uncertainties.
* Estimated standard deviation of the mean, in percent.

3.2 Characteristics and Functions

Several important characteristics of the NBS "4π"γ ionization chamber include: sensitivity, long-term stability, simplicity of operation, and simple source preparation [2]. The instrument is not only used to maintain results of direct standardization, but also provides checks of consistency between results of direct measurements made at different times. It is also useful for testing dilutions and sampling methods. It also serves to check or establish half-life data for most gamma-ray-emitting radionuclides for which basic calibrations have been performed. All samples measured are carefully checked for impurities with germanium gamma-ray spectrometers and appropriate corrections are applied.

3.3 Current Measurements

The electrometer is used as a coulombmeter, where the voltage developed across a capacitor is measured [5]. As shown in figure 3 this output voltage is fed into the comparator, which triggers a timer scalar to start counting the elapsed time between a low and high voltage reading on the capacitor. The triggering of the timer scalar is typically one volt to start the scalar and 10 volts to stop it. The critical point is that the comparator circuit is designed to always measure a fixed difference in voltage, i.e., nine volts, whether from 1 to 10 or from 2 to 11 volts, in order to minimize temperature fluctuations in the comparator circuits. After measuring this elapsed time, the comparator triggers a print control circuit which transmits the timer scalar and calendar scalar data to the computer for storage and further processing.

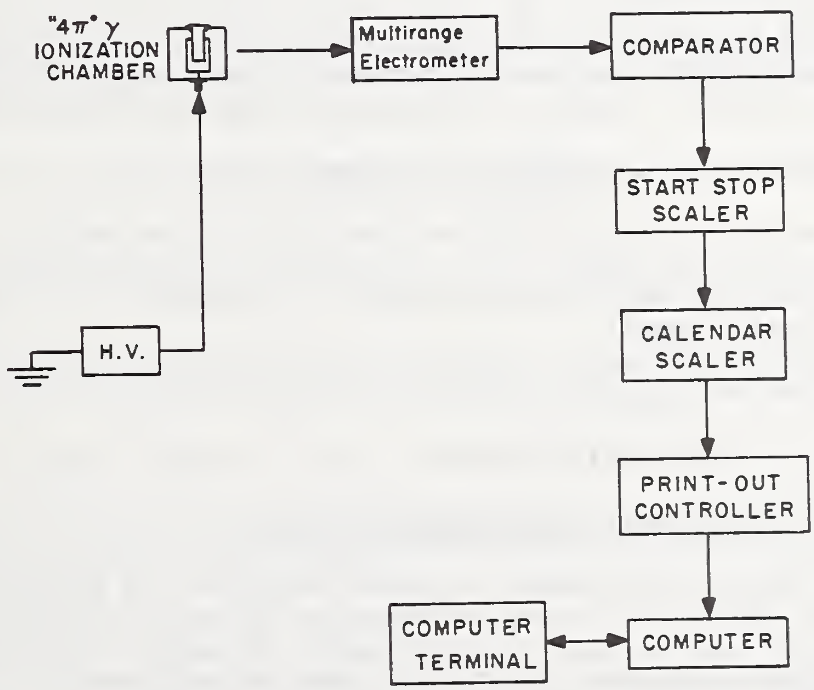


Fig. 3 Schematic Diagram of Electronic System for the "4π"γ Ionization Chamber.

The electrometer has a sensitivity stated to be 10μV. The input amplifier provides high input resistance (2×10^{14} ohms), low offset current (less than 5×10^{-15} A), and low noise. The double-shielded input section permits floating operations up to 1000 V above chassis. The construction of the electrometer keeps unshielded capacitance from input HI to chassis ground below 0.1 pF.

3.4 Source Container and Contents

The solution to be assayed in the NBS "4π"γ ionization chamber should have as nearly as possible the same composition as the standards used to calibrate the chamber [4]. The gamma-ray-emitting radionuclide solution should be 5.0 ± 0.2 mL in a standard 5-mL ampoule. The specifications of these ampoules are as follows:

body diameter	16.5 ± 0.5 mm
wall thickness	0.60 ± 0.4 mm
barium content	less than 2.5 percent
lead oxide content	less than 0.02 percent
other heavy elements	trace quantities

Where variation in sample volume cannot be avoided, corrections should be made from data obtained by investigation of changes in the pertinent parameters [4]. Corrections can be empirically determined for different solution heights, the function being only slightly dependent on the energy of the radiation.

4. Operational Procedures

The operational procedures for calibrations using the "4π"γ ionization chamber has been divided into several sections. Each section is designed to provide instructions for a specific operation. These sections are:

4.1 Description of the actual measurement process

4.2 Receiving instructions for radioactive solution samples

4.3 Step-by-step operating instructions for the NBS "4π"γ Pressure Ionization Chamber

4.4 Data Analysis

4.5 Format of Calibration Report

4.1 Description of the Actual Measurement Process

The stability of this chamber, although excellent, is made moot by measuring its response to a reference source of ^{226}Ra in equilibrium with its daughters whenever calibrations or measurements are made. The activity of the sample is taken to be proportional to the ratio of the chamber response (current) for the sample to that for the reference source. The relative calibration factor, K_R, is given by R_R/R, the ratio of the measured responses of the ^{226}Ra reference source and a known one bequerel of a stated radionuclide.

The activity, A, of the radionuclide in a sample, S, to be measured is given by $K_R R_S$, where R_S is the response (current) for the sample relative to that for the radium source. Background currents measured just before and after counting the sample and radium must be subtracted from the response for each. K_R diminishes from the value at the time of the original calibration with the 1600-year half life of the ^{226}Ra, and a correction for this must be made.

4.2 Receiving Instructions for Radioactive Solution Samples

All radioactive sources arriving at NBS are delivered to the Health Physics Group where they are examined to determine the radiation level and possible contamination of containers and packaging.

After Health Physics releases the source to the analyst, the analyst in turn should follow a quality control procedure to verify that the correct radionuclide has been shipped in the activity range which has been previously discussed with the customer. Quality control procedures that should be implemented by the analyst are as follows:

a. The analyst should re-examine the source for pin holes of any type and check the ampoule seal.

b. The analyst should inspect the label attached to the primary container. This helps to insure that it is the required radionuclide and the activity amount is correct.

c. Technical data sheets and package inserts accompanying the radionuclide package should be read carefully for special instructions on stability, quality inspection, and chemistry of the product. This information should conform to the calibration guidelines specifying the physical and chemical properties of the submitted sample solution, or see Tables 1 and 2.

Providing that no defects are observed, the sample may be transported for measurement in the NBS "4π"γ ionization chamber.

4.3 Step-by-Step Operating Instructions for the NBS "4π"γ Pressure Ionization Chamber

4.3.1 Starting of the Ionization Chamber System. The system is started from the control terminal using the following procedure:

Log on as user- "ondata" (lower case)

Password is _______(blank), carriage return. "datax" will be displayed on screen in lower case letters

Type "iconline", carriage return

Answer Y or y for default para., N or n to change and supply answers (generally use default values)

Carriage return after each answer

Questions are as follows:

New Bkg-0; Skip 1

BKGS=-1

Time: Day#_______

Input Start of Calender Scalar HR,MIN,SEC

Reset clock on clock scalar

Computer Time: HR:MIN:SEC

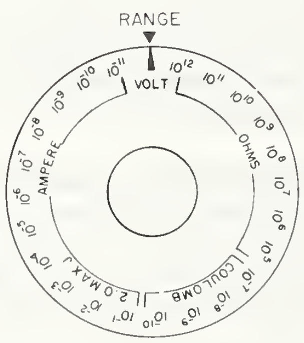
Statement of agreement between computer and calender clock

Answer Y or y if OK, N or n to reenter cal. start time

Enter Sample #: 0-BKG, 999-DONE, -999 change CAPR, PTS.

4.3.2 Background Measurements. Background measurements are made initially and should be examined before continuing. If the background is inconsistent, or abnormally different, constant repetitive measurements may be necessary.

4.3.3 Setting the Electrometer. Set the coulomb dial on the electrometer to 10^{-10} for background or 10^{-9} for a sample.

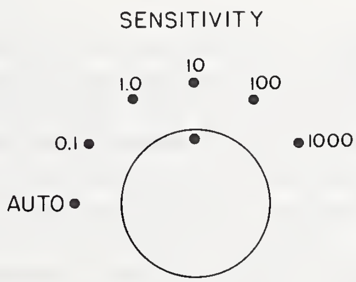


Set the sensitivity dial to the proper setting. The range of sensitivity is 0.1 to 1000. The sensitivity setting for background is 0.1. Refer to Table 6 for determining the 1-1000 range settings.

Table 6. Sensitivity Range Settings

Capacitor Ratio	Radium Reference Source*
1.0	Background
10.0	3, 10, 20
100.0	20, 50, 100, 200
1000.0	200, 500, 1000, 5000

*The number of the radium reference source used in approximately equivalent to the number of micrograms of radium per source.



Note: The capacitor ratio and the ^{226}Ra reference source to be used must be determined based on the activity provided by the supplier when the sources are submitted, with appropriate corrections for decay.

4.3.4 Measuring Reference Source. The ^{226}Ra reference source is then placed into a rigid, tight-fitting Lucite holder and lowered into the chamber.

After the radium reference source is placed in the chamber, change the timer controller switch from the manual to the automatic mode. This step is repeated each time a radium reference source or solution sample is measured.

After the designated number of measurements have been made on the ^{226}Ra reference source, or a sample, type on the terminal "-1", then press line feed key. This should be done at the end of every set of measurements. This will display the average count time for the sample after measurement. The sample number will be requested for the next sample which is to be counted.

Remove the holder from the chamber and replace the ^{226}RA reference source with the sample which is to be assayed.

4.3.5 Measuring the Sample. When placing the solution sample in the chamber, observe the ampoule to make certain that no liquid is in the neck of the ampoule. The sample is then counted the same number of times in each set of measurements as the radium reference source. At least two sets of measurements should be made on the sample, with an equivalent number of measurements made on the radium reference source.

After all measurements have been completed on the submitted sample and the radium reference source, it is recommended that background measurements be repeated.

4.4 Data Analysis

4.4.1 Data Collection. The data collection is under control of a FORTRAN program running on a multi-user super-microcomputer. The start of the calendar scalar and other default parameters are entered as prompted, by the operator. The operator enters the sample number, number of data points to be collected, and capacitor ratio. The data is collected and stored to a disk for off-line analysis. The programs are available from the Radioactivity Group, upon request.

4.4.2 Processing the Data. The data processing is done using a FORTRAN program. The program averages the data for each sample and corrects them for background and radioactive decay (using the established half life). The sample activity is calculated using the entered K-value for that radium reference source producing a current approximating that of the sample. The results, with intermediate averages if desired, are printed out.

4.4.3 Correction for Impurities. Figure 2a and 2b show the relative response curves for the NBS "4π"γ pressure ionization chamber for monoenergtic gamma rays. To construct the response for an impurity radionuclide for which there is no measured calibration factor, one uses the relative response for energy E_i and a specific ^{226}Ra reference source to calculate:

$$A = \frac{R}{R_R} \times \sum_i \frac{1}{(P_\gamma)_{E_i} (X\ )_{E_i}}$$

where A is the impurity activity, R_R is the reading for the reference source, R is the reading for the impurity radionuclide emitting gamma rays of energies E_i with a probability of emission per decay of $(P\gamma)_{E_i}$, and $(X\gamma)_{E_i}$ is the response for a gamma ray of energy E_i.

The sharp drop-off of response at approximately 100 keV is due to the absorption of low-energy gamma rays in the 1/8" brass liner in the reentrant tube in which the source is placed (See fig. 1). The advantage of this liner is the much lower response to low-energy x-rays, resulting in a more linear response curve for most nuclides having considerable x-ray emissions. However, a disadvantage is the low response to nuclides such as ^{125}I.

4.5 Format of Calibration Report

The following two pages show an example of a Report of Calibration. As stated in section 1.2, at the end of a calibration exercise, the technical user will be given a Report of Calibration stating primarily [3]:

a) the principal radionuclide

b) reference time and date

c) method of calibration

d) certified values for activity

e) decay-scheme assumptions

f) assessment of radionuclidic purity

g) overall uncertainty determinations for calibration

5. Assessment of Uncertainty

5.1 Precautions and Uncertainties in Service Calibrations

Measurements performed with the "4π"γ ionization chamber (IC) must be done under completely fixed conditions. This requires a stable background, no saturation losses due to incomplete charge collection because of recombination at high rates, and corrections for all impurities. Ampoules must be of the standard 5-mL type, the volume in ampoules must be 5.0 ± 0.2 mL, and the ^{226}Ra reference source must be corrected for decay.

Insufficient volume in the NBS standard 5-mL ampoule can be adjusted by adding an inactive carrier solution to adjust the height of the solution. Observed variations with height are used to estimate the uncertainty due to inexact fillings. Uncertainty in activity measurements may be increased if the contribution of the background is variable, large, or both. The uncertainty quoted is derived from measurement of the variations. The operator of the chamber should make frequent measurements of the background and look for the source of any unusual variations.

Radionuclidic impurities may or may not have a significant effect, depending on the radiations of the principal and impurity radionuclide and the amount of impurity present. If their half-lives are relatively short, measurements at different times may be used to check consistency. Germanium detector systems are used to identify and quantify the significant radionuclidic impurities that may be present. The uncertainty in these measurements is propagated through the calculated effect on the IC results.

The uncertainty in the ratio of radium reference sources used when the initial sample and test sample differ greatly in activity were determined by measuring different radium sources against the same and various radionuclides.

NBS-443
(Rev. 2-85)

U.S. DEPARTMENT OF COMMERCE
NATIONAL BUREAU OF STANDARDS
GAITHERSBURG, MARYLAND 20899

REPORT OF CALIBRATION

for

COMPANY NAME
ADDRESS

Radionuclides	Indium-111
Source identification	X5001
Source description	Liquid in NBS borosilicate glass ampoule (1)*
Solution composition	Stated to be carrier-free in 0.1 M HCl
Activity	4.157×10^{6} Bq
Reference time	1200 EST February 19, 1985
Overall uncertainty	0.73 percent (2)
Photon-emitting impurities (Activity ratios at reference time)	$^{114m}In/^{111}In$: $(2.0 \pm 0.6) \times 10^{-4}$ (3)
Half life	2.8048 ± 0.0005 days (4)
Measuring instrument	NBS pressurized "4π"γ ionization chamber calibrated by $4\pi(A,x)-\gamma$ coincidence efficiency-extrapolation

Dale D. Hoppes, Group Leader
Radioactivity Group
Center for Radiation Research

Randall S. Caswell, Chief
Nuclear Radiation Division
Center for Radiation Research

Gaithersburg, MD 20899
February 26, 1985
NBS Test No.

*Notes on next page

NOTES

(1) Approximately five milliliters of solution. Ampoule specifications:

body diameter	16.5 ± 0.5 mm
wall thickness	0.60 ± 0.04 mm
barium content	less than 2.5 percent
lead oxide content	less than 0.02 percent
other heavy elements	trace quantities

(2) The overall uncertainty was formed by taking three times the quadratic combination of the standard deviations of the mean, or approximations, thereto, for the following:

a) 59 ionization-chamber measurements on this sample	0.02 percent
b) standard deviation of the mean of six coincidence measurements	0.06 percent
c) dead time	0.01 percent
d) resolving time	0.02 percent
e) background	0.17 percent
f) gravimetric measurements	0.10 percent
g) standard deviation of the mean of the original ionization chamber measurement	0.05 percent
h) photon-emitting impurities	0.03 percent
i) efficiency extrapolation	0.10 percent
j) half-life	0.05 percent

(3) Limits of detection as a percentage of the gamma-ray-emission rate of the 245-keV gamma rays emitted in the decay of indium-111 are:

0.1 percent between 30 and 240 keV
0.01 percent between 250 and 1900 keV,

provided that the impurity photons are separated in energy by five keV or more from photons emitted in the decay of indium-111.

(4) NBS-measured half-life value. NCRP Report No. 58, 2nd Edition, February 1985, lists a half-life value of 2.805 ± 0.001 days.

For further technical information please contact J.M. Calhoun at 301-921-2383.

The origin of the uncertainty estimates (as standard deviations of means, or suitable approximations) in a direct comparison are shown in the following excerpt from the reporting form for an international intercomparison of activity measurements of ^{133}Ba. Measurement was by anticoindences between electron capture radiations (Auger electrons and x rays) and gamma rays measured by NaI(Tl) detectors. The efficiency of the proportional counter was varied and an extrapolation of its rate to an efficiency of 100% was made to give the activity.

^{133}Ba Uncertainty Components

Due to:	Uncertainty (%)	Obtained by:
counting statistics	0.07	standard deviation of the mean
weighing	0.1	calculated effect of limits
dead time	0.02	measured accuracy of live-time determination
background	0.1	calculation using estimated background uncertainty
timing	0.004	measured oscillator accuracy
fitting procedure	0.13	error on intercepts
absorption	0.05	limit of measured effects
radionuclidic impurities	0.001	limit of ^{134}Cs fraction
half-life	0.01	calculated effect of half-life uncertainty
Combined Uncertainty	0.21	(Square root of summed squares)

5.2 Significance of Uncertainty Statements

The uncertainties given in most certificates prepared in past years were divided into "random" and "systematic" parts. The former, derived from the well-defined estimated standard deviation of the mean of a stated number of measurements and the assumption of a normal distribution, was given as one-half the 99-percent confidence interval of the mean. The "systematic" part was given as the linear addition of "limits" of other possible errors in the measurement system. The "systematic" part was clearly dominant in most radioactivity calibrations. If an overall uncertainty was required, the two parts were added linearly; if the source uncertainty needed to be propagated as a component of the uncertainty in a subsequent measurement, it was sometimes divided by three and treated as a standard deviation of the mean.

Discussions among representatives of several national standardizing laboratories, following the distribution of a questionnaire about uncertainty statements by the Bureau International des Poids et Mesures, led to a suggestion that an approach akin to that used for those components susceptible to ordinary statistical analysis be applied to those that are not. This suggestion followed at least the spirit of the article by J. W. Müller Ref. [8]. With the idea that even the subsidiary quantities have distributions (perhaps measured poorly or not at all), an approximation of a standard deviation should be devised for them. A possible virtue is that deducing, say, the two-thirds coverage of an ill-defined distribution may be less subjective than setting a "99% limit." Moreover, if <u>all</u> uncertainties are taken to be equivalent to the corresponding standard deviations (or standard deviations of the mean, if appropriate), the propagation of uncertainties is straightforward and an approximation to the "total standard deviation of the mean," called the combined uncertainty, is easily formed. A more conservative overall uncertainty formed by taking three times this quantity is roughly equivalent to the former total uncertainty in many cases, and this is done in present certificates. No matter which method is used, the uncertainty for a radioactivity calibration cannot be considered an accurate quantity, but rather a best estimate.

5.3 Examples of Uncertainty Components

The uncertainties which have been determined for the NBS "4π"γ ionization chamber originate predominantly from the direct calibration method for the particular radionuclide. Uncertainty is also introduced in the operation of the ionization chamber and in the comparison with radium reference sources to preserve the initial activity calibration of a specific radionuclide. As examples, consider two samples submitted to NBS for measurements on the IC. The overall uncertainty was formed by taking three times the quadratic combination of the standard deviations of the mean, or approximations thereto, for the following:

<u>^{111}In</u>

		percent
a)	59 ionization-chamber measurements on this sample	0.02
b)	standard deviation of the mean of six coincidence measurements	0.06
c)	dead time	0.01
d)	resolving time	0.02
e)	background	0.17
f)	gravimetric measurements	0.10
g)	standard deviation of the mean of the original ionization chamber measurements	0.05
h)	photon-emitting impurities	0.03
i)	efficiency extrapolation	0.10
j)	half-life	0.05
	overall uncertainty = ($3 \times \sqrt{\Sigma\, u^2}$)	0.73

^{123}I

		percent
a)	60 ionization-chamber measurements on this sample	0.01
b)	solution composition and density	0.05
c)	five coincidence measurements	0.11
d)	efficiency extrapolation	0.10
e)	gravimetric measurements	0.03
g)	resolving time	0.05
h)	background	0.01
i)	half life	0.20
j)	original ionization-chamber measurements	0.01
k)	photon-emitting impurities in original calibration	0.04
l)	radium 5000 to radium 1000 reference sources ratio	0.25

overall uncertainty = 1.10
$(3 \times \sqrt{\Sigma u^2})$

6. References

1. NBS Special Publication 260, National Bureau of Standards, Standard Reference Materials Catalog 1986-1987 ed. [Available from the Superintendent of Documents, U.S. Government Printing Office, Washington, D.C. 20402].

2. NBS Special Publication 250, National Bureau of Standards Calibration Services Users Guide 1986-1988 ed. [Available from the Superintendent of Documents, U.S. Government Printing Office, Washington, D.C. 20402].

3. Hoppes, D. D., Basic Radionuclide Measurements at the United States National Bureau of Standards, Environmental Int. 10, 99-107 (1984).

4. NCRP Report 58, A Handbook of Radioactivity Measurements Procedures, National Council on Radiation Protection and Measurement, Bethesda, Maryland, (1985).

5. Rytz, A., Activity Measurements of a Solution of ^{134}Cs: Report of an International Conparison, Nucl. Instrum. Methods 192, 427-431 (1982).

6. Keithly Digital Electrometer Instruction Manual, Model 6616, Keithly Instruments Cleveland, Ohio, April 1976. (Mention of a commercial product does not imply recommendation or endorsement by the National Bureau of Standards, nor does it imply that the product identified is necessarily the best available for the purpose.)

7. Radioactivity Calibrations Catalog, 1986 (unpublished).

8. Müller, J. W., J. Nucl. Instrum. Methods 163, 241-253 (1979).

NBS-114A (REV. 2-80)

U.S. DEPT. OF COMM.
BIBLIOGRAPHIC DATA SHEET *(See instructions)*

1. PUBLICATION OR REPORT NO. NBS/SP-250/10

2. Performing Organ. Report No.

3. Publication Date October 1987

4. TITLE AND SUBTITLE

NBS Measurement Services: Radioactivity Calibrations with the "4π" Gamma Ionization Chamber and Other Radioactivity Calibration Capabilities

5. AUTHOR(S)

Jacqueline M. Calhoun

6. PERFORMING ORGANIZATION *(If joint or other than NBS, see instructions)*

NATIONAL BUREAU OF STANDARDS
DEPARTMENT OF COMMERCE
~~WASHINGTON, D.C. 20234~~
Gaithersburg, MD 20899

7. Contract/Grant No.

8. Type of Report & Period Covered

Final

9. SPONSORING ORGANIZATION NAME AND COMPLETE ADDRESS *(Street, City, State, ZIP)*

Same as item #6.

10. SUPPLEMENTARY NOTES

Library of Congress Catalog Card Number: 87-619870

☐ Document describes a computer program; SF-185, FIPS Software Summary, is attached.

11. ABSTRACT *(A 200-word or less factual summary of most significant information. If document includes a significant bibliography or literature survey, mention it here)*

This paper describes the use of the NBS "4π"γ ionization chamber — an instrument which provides an indirect method of comparing the activity (decays per second) of gamma-ray-emitting radionuclides with national standards for a routine calibration service by the National Bureau of Standards Radioactivity Group. A description of the chamber's construction and characteristics, the operational procedure, and the associated equipment is included.

A description of NBS capabilities for direct radioactivity calibrations is also presented. Many of these capabilities are used to establish calibration factors for the "4π"γ ionization chamber.

12. KEY WORDS *(Six to twelve entries; alphabetical order; capitalize only proper names; and separate key words by semicolons)*

activity; calibration; gamma ray; ionization chamber; radionuclides; radium reference source; standard

13. AVAILABILITY

☒ Unlimited
☐ For Official Distribution. Do Not Release to NTIS
☒ Order From Superintendent of Documents, U.S. Government Printing Office, Washington, D.C. 20402.
☐ Order From National Technical Information Service (NTIS), Springfield, VA. 22161

14. NO. OF PRINTED PAGES

41

15. Price

PUBLICATIONS IN THE SP 250 SERIES FROM THE CENTER FOR RADIATION RESEARCH

SP 250-1 NBS Measurement Services:
Spectral Radiance Calibrations
J.H. Walker, R.D. Saunders, and A.T. Hattenburg

SP 250-2 NBS Measurement Services:
Far Ultraviolet Detector Standards
L.R. Canfield and N. Swanson

SP 250-3 NBS Measurement Services:
Radiometric Standards in the Vacuum Ultraviolet
J.Z. Klose, J.M. Bridges, and W.R. Ott

SP 250-4 NBS Measurement Services:
Irradiation of Ferrous-Ferric Dosimeters
(Fricke- Dosimetry)
C.G. Soares, E.L. Bright and M. Ehrlich

SP 250-5 NBS Measurement Services:
Alpha-Particle Calibrations
J.M.R. Hutchinson

SP 250-6 NBS Measurement Services:
Regular Spectral Transmittance
K.L. Eckerle, J.J. Hsia, K.D. Mielenz,
and V.R. Weidner

SP 250-7 NBS Measurement Services:
Radiance Temperature Calibrations
W.R. Waters, J.H. Walker, and A.T. Hattenburg

SP 250-8 NBS Measurement Services:
Spectral Reflectance
V.R. Weidner and J.J. Hsia

SP 250-9 NBS Measurement Services:
Calibration of Beta-Particle-Emitting
Ophthalmic Applicators
J.S. Pruitt

SP 250-10 NBS Measurement Services:
Radioactivity Calibrations with the "4π"
Gamma Ionization Chamber and Other
Radioactivity Calibration Capabilities
J.M. Calhoun

SP 250-11 NBS Measurement Services:
Dosimetry for High-Dose Applications
J.C. Humphreys, D. Hocken, and W.L. McLaughlin

SP 250-12 NBS Measurement Services:
Neutron Personnel Dosimetry
R.B. Schwartz

SP 250-13 NBS Measurement Services:
Activation Foil Irradiation
with Californium Fission Sources
G.P. Lamaze and J.A. Grundl

SP 250-14 NBS Measurement Services:
Activation Foil Irradiation by Reactor
Cavity Fission Sources
G.P. Lamaze and J.A. Grundl

SP 250-15 NBS Measurement Services:
Photometric Calibrations
R.L. Booker and D.A. McSparron

SP 250-16 NBS Measurement Services:
Calibration of X-Ray and Gamma-Ray
Measuring Instruments
P.J. Lamperti, T.P. Loftus, and R. Loevinger

SP 250-17 NBS Measurement Services:
The NBS Photodetector Spectral Response
Calibration Transfer Program
E.F. Zalewski

SP 250-18 NBS Measurement Services:
Neutron Source Strength Calibrations
E.D. McGarry and E.W. Boswell

SP 250-19 NBS Measurement Services:
Calibration of Gamma-Ray-Emitting
Brachytherapy Sources
J.T. Weaver, T.P. Loftus, and R. Loevinger

SP 250-20 NBS Measurement Services:
Spectral Irradiance Calibrations
J.H. Walker, R.D. Saunders, J.K. Jackson,
and D.A. McSparron

SP 250-21 NBS Measurement Services:
Calibration of Beta-Particle
Radiation Instrumentation and Sources
J.S. Pruitt, C.G. Soares, and M. Ehrlich

U.S. Department of Commerce
National Bureau of Standards
Gaithersburg, MD 20899