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LOW-LEVEL MEASUREMENTS AND THEIR APPLICATIONS TO ENVIRONMENTAL RADIOACTIVITY

Editors

M. GARCIA-LEON

G. MADURGA



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RADON MEASUREMENTS AND CALIBRATION STANDARDS

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R. Collé and J.M.R. Hutchinson
Center for Radiation Research
National Bureau of Standards
Gaithersburg, Maryland 20899, USA

ABSTRACT

Recent and increasing awareness of the potential health hazards of radiation exposure to the general public due to indoor radon has made the topic of radon measurements and calibration standards one of considerable significance. In the U.S., as in other countries, various remedial action programs and wide-scale measurement surveys involving tens of thousands of measurements have been instituted. This paper, in the context of the radon control activities in the U.S., describes the most widely employed methods used for the measurement of radon concentration in air, radon concentration in water, and radon flux density. The primary measurement standards, transfer calibration standards, and calibration support services needed to support these measurements are also described.

1. INTRODUCTION

Before directly addressing the topic of radon measurements and calibration standards, it may be insightful and useful to briefly summarize some of the ongoing activities involving radon in the U.S. In this way, one may obtain a better understanding of why radon probably has become the most significant area of environmental radioactivity.

2. OVERVIEW OF RADON CONTROL PROGRAMS IN THE U.S.

In the past ten years, indoor radon^{*} has been identified and continues to be one of the more significant radiation protection problem areas facing many countries (cf., Hopke^{1]}). This has resulted in a variety of national remedial action programs and wide-scale measurement surveys within these countries. In the United States, this continuing concern about exposure of the public to radon (^{222}Rn) and its decay progeny has resulted in the development of a voluntary Federal radon control program, as well as an increased interest in the quality of radon measurements used to support this program^{2]}.

Unlike some of the regulatory control programs proposed or established in the European community, the U.S. has adopted a policy, supported by the Congress, that programs addressing indoor radon should be nonregulatory, providing technical support to the States. The Federal agencies role is to develop information on risk to the general population from radon exposure, provide advice on remediation and mitigation methods, and support the activities of State and local authorities.

Most of the Federal effort on indoor radon is performed by the U.S. Department of Energy (DOE) and the U.S. Environmental Protection Agency (EPA). DOE programs primarily are directed at fundamental research on health effects, radon sources and pathways into buildings, and on remedial measures to reduce indoor radon levels; whereas EPA activities mainly involve implementation programs that provide assistance to State and local governments, and applied research for these programs. The activities of other agencies within the U.S. government, e.g., the U.S. Geological Survey (USGS), Centers for Disease Control (CDC) in the Department of Health and Human Services, and the National Bureau of Standards (NBS) are less extensive, but provide vital research and support services.

*A brief review and summary of the physical and chemical properties of radon and its progeny as it governs their natural occurrence, their pathways into buildings, and their behavior in indoor environments may be found in Colle^{3]}.

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To provide guidance to the public and the States on indoor radon control, EPA in cooperation with the CDC developed a set of recommended remedial action guidelines^{4]}. The guidance consists of a graded scale of annual average radon concentrations in four broad categories in which the level determines the urgency for remedial action. In the lowest category below about 4 pCi/L (0.02 WL) it is suggested that reductions in radon concentrations may be "difficult, and sometimes impossible to achieve" although "exposures in this range do present some risk of lung cancer". In the range of 4 to 20 pCi/L (0.02 to 0.1 WL) it is recommended that remedial action be taken "within a few years, sooner if levels are at the upper end of this range." For indoor radon concentrations in two higher ranges, above about 20 pCi/L (0.1 WL) and above about 200 pCi/L (1.0 WL), more urgent action is recommended. With these guidelines and with the risk information also contained in the guidance document^{4]}, homeowners should have enough information to make informed decisions about whether and how quickly they may need to take action to reduce their exposure to indoor radon. The level of 4 pCi/L is often blindly used as an absolute remedial action fiducial benchmark even though the guidance was given in ranges to avoid this type of unreasonable and strict interpretation.

To date, many thousands of indoor radon measurements have been made in the U.S. These results are forming the basis for a comprehensive database on how widespread and serious the radon problem is in the U.S. Much of the early data was obtained for research studies by universities and national laboratories. More recently however, nearly twenty of the States, some with EPA assistance, have conducted or presently are performing screening surveys of indoor radon concentrations in homes; and many individual homeowners have privately contracted with commercial radon measurement vendors to make measurements in their homes. A national survey which will be conducted by EPA next year is also being planned. This survey is intended to characterize the frequency distribution of indoor radon levels across the U.S. — in homes as well as schools and workplaces.

Available data indicate that indoor radon levels in the U.S. vary widely and the problem is more widespread than initially believed. Nearly 25% of all homes tested have radon concentrations exceeding the lower end

of the 4 to 20 pCi/L action level range. This has been found in virtually every State. Similarly, very high levels exceeding 100 pCi/L have been found in virtually every State. The initial surveys were undertaken to try to identify high-risk areas, and to give the states an idea of the distribution within the States. It appears, however, that even States with low overall radon distributions, can have very localized "hotspots."

As a result, testing individual buildings is the only way to ensure a relatively low indoor radon concentration.

Assuring the quality of radon and radon progeny measurements is, of course, a necessary and vital element of any effective radon control program. Within the U.S., the EPA was instrumental in developing standardized measurement protocols and in establishing a nationwide measurement proficiency program. The interim protocols^{5]} for making "field" are nearly universally used throughout the country screening measurements of radon and radon progeny in buildings, thereby insuring that measurements made anywhere within the U.S. are consistent and compatible. EPA also established a voluntary Measurement Proficiency Program for commercial radon measurement vendors^{6]}. This program is very important, particularly because of the proliferation of vendors offering measurement services to the public. It is administered by EPA, but relies upon the calibration services of several national laboratories. The program provides States and the public with information concerning the proficiency of hundreds of measurement firms and analytical laboratories across the country.

The role of NBS in this overall U.S. radon control program derives from its primary function which is "the custody, maintenance, and development of the national standards of measurement, and the provision of means and methods for making measurements consistent with those standards...."^{7]}. NBS not only maintains a national radon measurement standard, but also provides various transfer calibration standards and calibration services that are directly relatable to this national standard, and conducts measurement proficiency tests and measurement intercomparisons with other laboratories. In addition, NBS continually monitors the need for new transfer standards and services, and maintains a limited program to develop and disseminate them.

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3. RADON MEASUREMENT METHODS

3.1 Introduction

It is important to recognize that the monitoring and characterization of indoor radon sources may involve the measurement of several very different quantities. These quantities include: radon concentration in air; potential alpha energy concentration (in units of working levels); individual radon progeny concentrations in air; radon exhalation rate or flux density from surfaces; radon concentration in water; thoron (^{220}Rn) and thoron progeny concentrations; and special aerosol measurements, such as condensation nuclei, particle size distributions, uncombined fractions, etc.

For each of these quantities, there usually are several measurement methods utilizing widely different protocols and instrumentation. Although there are common elements in some methods, in many cases the specific method for the measurement of a given quantity will have to be considered and treated as a unique entity. Just in terms of sampling times, the various methods include "instantaneous" (grab-sample) methods in which a sample is obtained nearly instantaneously over a few-minute sampling period, time-integral methods which provide a single, average value for an extended period of time ranging from a few days to a week or longer, and continuous monitoring methods which provide individual short-term measurement results continuously over long time intervals. As a result, the subject of radon measurements is one of considerable diversity.

This diversity can be easily illustrated by the results of a 1981 survey of radon measurement activities in State radiation control programs^{8]}. The survey identified that 13 different methods were used by 19 States to measure radon concentration in air; 5 different methods were used by 24 States to measure potential alpha energy concentrations; at least 4 different methods were used by 16 states to measure individual radon progeny concentrations in air; 3 different methods were used by 11 States to measure radon flux density; and 3 different methods were used by 24 States to measure radon concentration in water.

As suggested, methods exist for nearly every conceivable type of necessary radon measurement. They range, of course, from very sophisticated methods and instruments used in research applications (e.g., in understanding the sources, pathways, and aerosol characteristics of radon and its progeny in buildings) to relatively simple passive devices used for screening measurements in homes. Many of these methods have been summarized by Budnitz⁹, by Breslin¹⁰, by George¹¹, by Colle¹², and more recently in a NCRP Report¹³.

This paper will briefly consider only those measurement methods that are most widely used and directly applicable to indoor radon assessments made in the U.S.

As recently as only a few years ago, it was widely held that indoor radon assessments should be based on measurements of potential alpha energy concentrations (working level) or radon progeny concentrations. This was believed because the dose to the human lung resulting from exposure to radon can be predominately attributed to the alpha particles emitted in the decay of the radon progeny, and not that from the radon itself. At the present time, however, most indoor radon assessments are based exclusively on measurements of average radon concentrations. This simplification arose with the understanding that the potential radiation hazard due to radon and its progeny was not only dependent on the degree of radioactive equilibrium between radon and its progeny (i.e., the equilibrium ratio that relates the radon concentration to the working level), but also on such important factors as the relative attached and unattached fraction and the aerosol size distribution. Each of these is time dependent and is influenced by a large number of environmental variables. Except for research applications, such a detailed radon assessment would be hopelessly complex and impractical for surveying large numbers of buildings. Furthermore, there is little advantage to a working level measurement over an average radon concentration measurement since the equilibrium ratio in nearly all indoor environments is approximately 50%, and the ratio is rarely outside the range of 40% to 70%. In general, measurements of radon concentrations are also more reliable in terms of consistency, ease of calibration, and accuracy. As a result, passive screening devices for the measurement of average radon concentrations have become the workhorses of indoor radon assessments in the U.S.

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Two other important applications that will be considered here are the measurement of radon concentration in water and the radon flux density from surfaces.

3.2 Radon-in-Air

Scintillation-counting methods are widely used in a variety of configurations for the assay of radon in air. For these methods the radon sample is introduced into a suitable vessel (e.g., a chamber or a flask) containing a silver-activated zinc sulfide phosphor which emits light that can be detected with photomultiplier tubes when the alpha particles from decay of the radon and progeny interact with the phosphor. The basic physics and operating characteristics of scintillation detection devices are described in many elementary texts and other references^{14,15,16}. Several distinct techniques are employed.

In the electrostatic-collection technique, the initially positively-charged recoiling daughter atoms from the decay of radon are collected by means of an electric field onto a ZnS(Ag)-coated electrode within the chamber containing the radon sample.

In the second technique, the gas sample is contained in a vessel whose walls are coated with a ZnS(Ag) phosphor and viewed by one or two photo tubes. The scintillation flask, popularly known as the Lucas cell¹⁷, is extensively used both in the laboratory and for field measurements. Scintillation flasks, both commercial and home-made, come in a variety of sizes (nominally 50 mL to 650 mL), are constructed of either glass or plastic walls, and are equipped with one or two valves. The flasks are usually pre-evacuated for collection of a sample of whole air, and then mounted on the phototube assembly for counting, usually after a delay of a few hours to allow the radon progeny to come into radioactive equilibrium with the radon. For field use, the flasks may be returned to a laboratory for counting, or may be measured entirely in the field with the use of portable, battery-powered photomultiplier assemblies.

Continuous flow-through scintillation monitors are also popular. The instruments consist of a Lucas-type scintillation flask with two ports that allow air to be continuously drawn through the flask. It is permanently coupled to a photomultiplier tube and pulse counter, and can

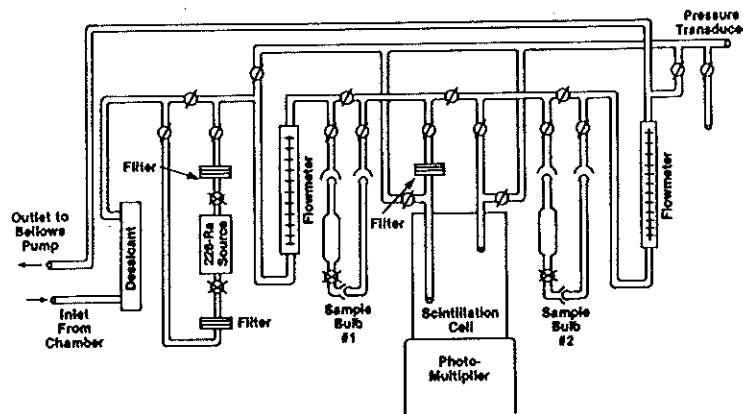


Fig. 1. Schematic diagram of an NBS gas-line sampling and monitoring manifold for radon. The monitoring system uses a flow-through scintillation cell. The manifold also contains a 226-Ra source for generating radon samples.

thus continuously monitor changes in the radon concentration in the air drawn through the flask. Figure 1 illustrates the use of this type of flow-through scintillation cell which is installed at NBS on a gas-line sampling manifold. When it is used in a continuous mode, even if the inlet air stream is filtered, radon progeny radionuclides produced within the flask "plate out" and deposit within the detecting volume. As a result, the count rate is affected by the plated-out progeny that arise from radon sampled in the previous several hours. A complicated decay correction procedure is required to obtain a true instantaneous radon concentration reading.

For screening measurements in buildings the two most widely employed methods are charcoal adsorption and nuclear track techniques. Both of these methods rely on passive monitors and yield a time-integrated radon concentration. Their principle advantage compared to other methods is the relatively low measurement cost for wide-scale deployment.

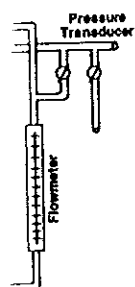
The charcoal adsorption method uses a canister containing activated charcoal. The method relies upon the passive diffusion of ambient radon into the canister and its adsorption onto the charcoal. The quantity of radon collected is then determined by directly assaying the charcoal for the radon progeny radionuclides by gamma-ray spectrometry. Several

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different sizes and types of canisters have been used. They typically range from 20 to 80 grams of charcoal. Some canisters also use diffusion barriers to eliminate the entrance of thoron and to minimize the effect of radon concentration transients, and desiccant packages to minimize humidity effects. Before the exposure period, adsorbed moisture is removed by heating the canister for several hours. The canister is then sealed for delivery to the sampling location where a lid or top of the canister is removed and radon allowed to adsorb onto the charcoal. The canisters are deployed for a period of 3 to 5 days at which time they are resealed and returned to the laboratory for analysis. The analysis is performed by placing the canister on a NaI(Tl) scintillation detector and measuring either three or four gamma rays from ^{214}Pb and ^{214}Bi (242, 294, 352, and 609 keV). Typical detection levels for the method are a few tenths of a pCi/L. The charcoal canister method requires a very careful control on calibration and correction factors. The correction for water adsorption (obtained by weighing the charcoal before and after the exposure period) alone can be as high as 50%.

The nuclear track film technique is the second time-integrated, passive, screening measurement method. For this method, a small piece (nominally 1 cm) of a suitable plastic film, such as cellulose nitrate, is exposed for long period of time in the air to be monitored. Alpha particles, incident on the film, damage the surface. By etching the film in suitable alkaline solutions (e.g., sodium hydroxide), each point of impact is enlarged. The resultant tracks and pits are then counted with a microscope or by other electro-optical means to provide a measure of the total number of alpha particles incident on the film during the exposure period. Various types of membranes and filters have been used with the films to achieve different hold-up times that can be used to exclude thoron, and to filter the particulate radon progeny radionuclides. The method can be calibrated and used to measure radon concentration or potential alpha energy concentration. Direct calibrations of each batch of films in known radon atmospheres to relate the track density to the quantity of interest must be performed for each specific application. At typical indoor radon concentrations, long exposure periods (3 months to 1 year) are needed to obtain sufficient track density; and the precision



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is poor (about 25 to 50%) for the shorter exposure periods for the field sizes typically counted. Because of its simplicity, low cost, and long integration length, however, the nuclear track film technique is a very useful screening method for obtaining average indoor radon concentrations.

Additional information and details on these methods, (including some of the underlying assumptions for the methods, their applications and limitations, and their calibration requirements) can be found in References 12 and 13.

3.3 Radon-in-Water

The most common procedure for assaying radon in water is based on liquid scintillation (LS) counting. Although other methods (primarily based on the de-emanation of radon by aeration and the quantitative collection of the released radon for subsequent assay) are available, the LS method has been extensively used in the U.S. to conduct a nationwide survey of radon concentration in private, commercial, agricultural, public and community water supplies[18]. In this survey, more than 2500 public water supplies, representing about 5% of the total number of groundwater supplies in the contiguous 48 States and nearly 45% of the water consumed by U.S. groundwater users, were sampled.

For the LS method, an unaerated water sample is taken into a syringe, and injected beneath the surface of a scintillation cocktail contained in a LS counting vial. The vial is then shaken in order to equilibrate the radon between the air space and the gel which is formed from mixing the water and cocktail, and measured with a LS counting system. The LS sample mixtures typically consist of 5 to 10 g of water with 10 g of cocktail in 15 or 20 mL vials. Suitable scintillation cocktails that have been used include mineral oil, toluene-based, and xylene-surfactant-based phase-combining mixtures^{19,20}. These cocktails are miscible with water over a wide range, and have counting efficiencies of 90% to nearly 100% for radon. The LS counting technique and operation of the LS counter are fairly routine, and will not be described here. These details may be found in standard texts^{14,15,16}. Since radon is readily released from water on aeration, one of the most important aspects in measuring radon in water is the procedure used to collect the water sample. The procedure used for the U.S. survey consisted of connecting a tube from the water supply outlet to the bottom of a wide-mouth funnel, letting the water flow upward through the funnel and over the mouth of the funnel so as to not

aerate the water stream. While the water is continuously flowing, the sampling syringe is inserted into the funnel mouth and used to remove the water sample which is then injected into the LS vial.

3.4 Radon Flux Density

Radon flux density is just the rate of exhalation of radon from a given surface area. Although most of the methods for measuring radon flux density were developed for studying radon exhalation from the earth's surface, the methods may, with minor variation, be applied to measure the flux density from any surface. These measurements are often useful in evaluating the release rate of radon from soils at building sites, and may also be used indoors on structural surfaces to determine the entrance pathways of radon into buildings.

Many methods for the measurement of radon flux density are known. These methods have been reviewed in detail by Collé, et al.^{12]}. The two major methods are the accumulation method and the adsorption method.

The accumulation method involves the direct accumulation of radon in an open-faced vessel which is inverted and placed (and possibly sealed) on the surface. In this way, the vessel and surface forms a closed container for the accumulation of exhaled radon. On soil surfaces, the vessel is normally imbedded several centimeters into the ground. On other surfaces, such as building materials, the vessels are sealed with epoxy resins or other caulking agents. Many different types and sizes of accumulators have been used including large barrel drums, large surface area parallelepipeds, and shallow cylindrical disks. A number of efforts have also been made to control the conditions within the accumulator. This includes the use of fans to mix the accumulator atmosphere and insure homogeneous sampling, maintaining constant pressure in the accumulator by drawing air in during sampling or using pressure-equalizing orifices, and monitoring and controlling internal temperatures.

Some early deployments of the method relied upon a long accumulation period followed by a single sampling and assay of the radon. The method is now commonly performed by periodic sampling and assaying to follow the growth of radon concentration in the accumulator. This is done because with materials of high porosity, such as most soils, the radon concentration in the accumulator will rapidly become comparable to the concentration of radon in the pores of the soil. "Back diffusion" of radon into the pores will then occur, resulting in marked departures from the growth

curve which is governed by the simple radioactive growth equation. The exhalation rate is determined from the initial slope of the observed radon concentration growth curve using a well-known and developed methodology.

The accumulation method is based on three important assumptions: (i) the accumulation time is short compared to the radon half-life; (ii) the concentration of radon in the accumulator is significantly less than the soil gas concentration to prevent appreciable back diffusion; and (iii) the presence of the accumulator does not seriously perturb the exhalation process. The third assumption can be very difficult to verify, and its implications will be addressed further in Section 4.5.

Sampling and assay of the radon from the accumulator is similar to many of the techniques used for laboratory measurements of radon. The techniques employed, however, are usually those more compatible to the demands of field deployment. As a result, the most common variation is the collection of radon grab samples directly into evacuated scintillation flasks, although other sampling, transfer, collection and gas-counting techniques have been employed.

The adsorption method for the measurement of radon flux density is a variation of the same charcoal canister technique used to measure radon concentration in air (Section 3.2). In this method, charcoal-filled canisters, very similar to those used for the radon-in-air measurements, are sealed to the surface to be measured. After a deployment period of several days, the canisters are sealed and returned to the laboratory for analysis by gamma-ray spectrometry. Several types of canisters have been used for a variety of purposes. The method has been used indoors to locate and measure radon sources and routes of entry into buildings. Various adapters have been employed for mounting canisters in corners, across wall-wall or wall-floor joints and over cracks in concrete foundations.

The charcoal canister method is now the most widely used method for determining radon flux density. In large part, this is because it is relatively simple, inexpensive, and much less labor intensive than the other methods. The extent to which the adsorption method, however, by the direct placement of the charcoal on the exhaling surface, perturbs the exhalation rate is often unclear. Of the radon flux density methods, it probably most poorly simulates natural conditions. Furthermore, the method is primarily calibrated by comparison against the accumulation method, and therefore any perturbing effect that might be observed by comparing the method to other methods will be masked.

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4. PRIMARY AND TRANSFER CALIBRATION STANDARDS

Unlike other radionuclides, radon can not be fundamentally or directly calibrated without recourse to an indirect or comparative measurement. As a result, all measurements of radon are ultimately related back to some ^{226}Ra standard. Within the U.S., NBS maintains both international and national radium standards that can be directly traced to the international primary standards of radium prepared by Mme. Curie in 1911 and Otto Hönigschmid in 1934^[21]. The national standard for radon measurements is embodied in a primary radon measurement system that has been maintained for nearly 50 years to accurately measure radon against these international and national standards. In turn, all of the radon measurements made by NBS and the radon transfer calibration standards and calibration services provided by NBS are directly relatable to this national radon standard. These primary and transfer calibration standards and services, as well as some of the facilities used to support them, are described below.

4.1 Radium-226 Standard Reference Materials

NBS ^{226}Ra solution Standard Reference Materials (SRMs) are one of the principle transfer standards for radon measurements^[22]. Various series of these standards have been available since the mid-1940s. The current series of four SRMs contain approximately 10 or 20 grams of solution in a flame-sealed ampoule and have a ^{226}Ra content ranging from 10^{-14} to 10^{-8} grams. A few standards in an additional series of nine ^{226}Ra gamma-ray solution standards are also available. They contain 5 grams of solution and have a radium content ranging from 10^{-7} to 10^{-4} grams. They can also be used for radon analysis at higher concentrations.

These transfer standards are used by some laboratories as their "primary" standard for radon. Although better laboratories can be expected to utilize this standard with success, it is not a practical standard for laboratories which do not maintain a large internal calibration capability. Their use requires an indirect calibration based on a very careful quantitative extraction and collection of radon from a solution. More directly usable standards or calibrations are needed by the hundreds of commercial firms presently making radon measurements in the U.S. and work in this area is planned by NBS.

4.2 Primary Radon Measurement System

The primary radon measurement system, which constitutes the national radon measurement standard, consists of pulse ionization chambers and ancillary gas-handling and -purification equipment^{23,24,25].} With this system, radon samples are transferred with a slow stream of nitrogen carrier gas through a purification system which removes water vapor and oxygen from the gas stream. Transferred samples are counted after the radon and its short-lived progeny reach secular equilibrium.

The ionization chambers are calibrated against ^{226}Ra standards (Section 4.1) by quantitatively transferring known accumulated amounts of radon into the chambers. This transfer is performed by slowly bubbling the nitrogen carrier gas stream through a standardized radium solution which is contained in a modified gas-washing bottle. As in the measurement procedure, the gas stream passes through the purification system and fills the chambers. Great care must be taken to insure that the transfer of radon from the radium solution to the chamber is 100% efficient^{24]}.

4.3 Radon Calibrations

Radon calibrations for other laboratories can be performed at NBS by either assaying the radon concentration in a gas sample bulb which is sent by the laboratory, or by sending a sample bulb filled with a known radon concentration to the laboratory for their assay. In either case, the radon concentration in the assay measurement made at NBS or in the sample bulb filled at NBS is directly relatable to the primary radon measurement system (Section 4.2).

Recently, a NaI(Tl) well counter was cross calibrated against the primary system. This detector used in conjunction with spherical glass ampoules containing radon samples was shown to be very reproducible and fairly independent of geometry. It has a wide dynamic range and can be used for radon samples from the picocurie level to a 100 nCi level. This system was used for a recent measurement intercomparison of all laboratories within the U.S. that maintain an independent calibration capability based on radium solution standards (see Section 4.1).

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4.4 Radon-in-Water Standard Generator

In 1986, NBS completed the development of a transfer standard for radon-in-water measurements^{20,26]}. This work was initiated at the request of the U.S. EPA to support the extensive national survey of radon concentration in drinking water supplies (see Section 3.3).

This standard consists of a polyethylene-encapsulated radium solution source in a small-volume accumulation chamber, and an ancillary mixing and dispensing system. It generates aqueous solutions of radium-free radon of which multiple aliquots may be dispensed and used as standardized solutions for calibrating radon-in-water assay procedures. The overall uncertainty in the radon concentration for a sample aliquot dispensed from the generator (such as into LS vials) is estimated to be approximately 4%.

The standard generator is illustrated schematically in Figure 2. The small source consists of a known quantity of ^{226}Ra sealed into a small thin-walled polyethylene capsule. The radon generated in the capsule from the decay of radon diffuses through the polyethylene and enters the water contained in the accumulation chamber. After a suitable accumulation period, the radon solution in the chamber is diluted and quantitatively transferred into syringe S1. The second syringe S2 is used for mixing by passing the solution between the two syringes. This mixing and dispensing system was partially automated with motor-driven syringes. There are several novel aspects to this standard which derive from the solubility of radon in both water and polyethylene. These aspects control not only the operating principles behind the standard, but also on how it may be used (e.g., the required flushing procedure). Additional details on the development, operation, and performance of the standard may be found in Hutchinson, et al.^{26]}.

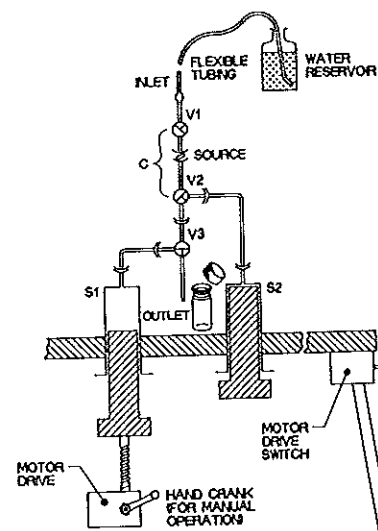


Fig. 2. Schematic diagram of the radon-in-water standard generator.

4.5 Radon Flux Density Standard

The two most popular methods for the measurement of radon exhalation rate or radon flux density (i.e., the exhalation rate per unit area) are the accumulation and charcoal canister methods (see Section 3.4). At the present time, in the absence of a direct radon flux density transfer standard, both methods require an indirect calibration of radon either sampled from the accumulation vessel or absorbed in the charcoal canister. In addition, the flux density calibration must also rely on additional assumptions such as the diffusion model for the accumulation of the radon in the accumulator can or the efficiency for the adsorption of radon in the charcoal canister. A direct transfer standard that provides a constant and known exhalation rate from a relatively large surface would be useful, and a significant calibration standard for users.

In the past two years, NBS has been working on the development of such a transfer standard. A few attempts have been made by other laboratories to obtain a radon flux density calibration facility which are based on the characterization of a fabricated site of either natural or enriched radium-bearing soils, sands, or mill tailings. The novel, alternative approach considered for the NBS standard consists of incorporating an aqueous solution of radium in a sealed, shallow, large-surface area container which is covered with a thin, but rigid, polyethylene sheet which radon may diffuse through. The use of the radium solution eliminates possible spatial and inhomogeneity dependencies in the source production rate of radon. The source configuration, illustrated schematically in Figure 3, consists of a water-polyethylene interface, a polyethylene-air interface, and all other surfaces are impervious to the transport of radon. This source configuration is expected to exhibit purely diffusive (Fick's Law) radon transport properties without a permeation (Darcy's Law) transport component which is one of the troublesome and unpredictable aspects of radon transport in solid, porous media. This will tend to minimize the effects of ambient and meteorological variations. The source configuration, in fact, can be described in terms of a classical physicochemical three-phase system of water, polyethylene, and air. The generated radon in the water will diffuse into the polyethylene

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Radon Flux Density Standard

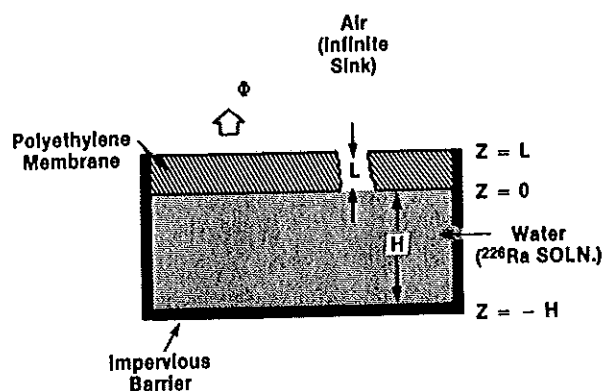


Fig. 3. Illustration of the configuration for a Radon Flux Density Standard under development at NBS.

and form a true radon in polyethylene solution. The radon, as for any two-phase solution system, will partition between the water and polyethylene in a predictable way. Similarly, the radon will exhale from the polyethylene surface while maintaining a constant partition coefficient between the polyethylene and air. The requirement that the partition coefficients be maintained should result in a very constant exhalation rate from the surface. The source configuration may be characterized in terms of a steady-state diffusion model. This model is an extension of the various radon transport cases treated in Collé, et al.^{12]}, and is based on a derivation by Rubin^{27]}.

A 40-cm diameter prototype, based on design calculations from the model, has been constructed (with partial support from the U.S. EPA). It consists of a stainless steel right circular cylinder with a radium solution depth of 6 cm which is covered and sealed with a 0.6 mm polyethylene

sheet. It was designed for a flux density of approximately 5 pCi per m^2 sec which is about 10 atoms per cm^2 per sec. The prototype is also equipped with an ancillary circulation system which is driven with a seal-less, magnetic-drive pump and contains appropriate valves, inlet port, and drain line. This plumbing system is used for loading the radium solution, for possible makeup of water transpiration losses, and for mixing to ensure that the solution remains homogeneous. In developing the prototype many engineering details and technical problems, such as in providing impervious seals between the polyethylene and stainless steel and in devising a filling procedure that minimized the occlusion of air within the source chamber, had to be resolved.

The prototype has been operational for over a year. The first preliminary calibrations have been completed. These include evaluations for the constrained flux density (i.e., when the radon is allowed to accumulate in a chamber above the source which constrains the release rate of radon from the source) and for an unconstrained case (i.e., when a negligible radon concentration in the air above the polyethylene surface is maintained). Unless the unconstrained flux density is measured directly, one must rely on an assumed mathematical model to calculate it from a measured constrained flux density. As mentioned in Section 3.4, this is one of the underlying assumptions and difficulties inherent in all field measurements of flux density. The demonstrated ability to measure the unconstrained flux density directly and its rough agreement with the constrained flux density is significant in implying that the final transfer standard may not have to rely on the adequacy of a diffusion model for the source configuration. Many additional calibrations and performance evaluations will have to be performed, of course, before we are satisfied with the efficacy of the standard. To evaluate these various test cases, the modelling of the transfer standard was, however, also extended to include time-dependent behavior^{27]}. It may be of interest to note that each of the evaluation measurements on the prototype transfer standard requires nearly a month-long duration because of the long time required to re-establish a steady-state after a change in the boundary conditions for the source.

4.6 Exposure Chamber

NBS has the capability of providing known and well-characterized atmospheres of various gaseous radionuclides including radon in a relatively large exposure chamber. The chamber was designed and built for use in calibrating instruments and for use in exposing passive monitors or dosimeters. At the time of the Three Mile Island nuclear power plant accident, for example, the chamber was used with known ^{133}Xe atmospheres to calibrate various beta-detection instruments that were deployed at the accident site. Although the chamber has never been used for radon exposures or calibrations, it easily could be employed for such purposes. As part of some standard development work, the chamber was equipped to continuously monitor radon concentrations and used to measure the exhalation of radon from materials placed inside it.

The chamber, shown in Figure 4, is a 650-L stainless steel (SS) vessel which is provided with sampling ports and equipped with a SS recirculation bellows pump and an all SS sampling and recirculation plumbing system. The system has the capability for measuring air mass flow rate at two locations and for monitoring ambient temperature, relative humidity and pressure. The sampling and radon monitoring manifold used with the chamber was shown in Figure 1. The chamber and operating system could be adapted to incorporate additional controls and monitors to obtain environmentally controlled and regulated aerosol atmospheres.

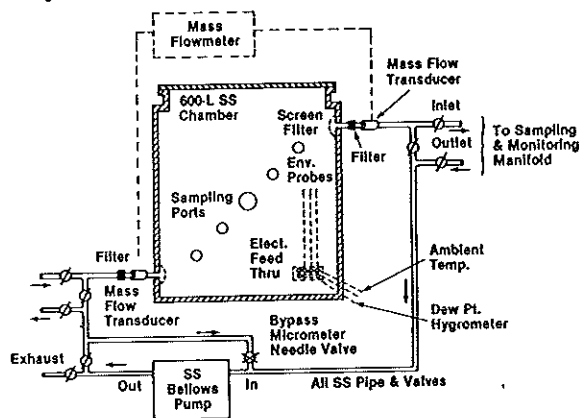


Fig. 4. Schematic diagram of the NBS Exposure Chamber.

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