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Contents

Articles

Conference Reports

News Briefs

Volume 95, Number 2, March–April 1990
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Calibration of Radon-222 Reference Instrument in Sweden

Volume 95 Number 2 March–April 1990

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A simple procedure to calibrate and characterize a recently developed radon-222 reference instrument is described. The system, which is now used as the official national Swedish reference, is quick and easy to use. Systematic as well as random errors are smaller than in an earlier system and compare well with other systems, as has been shown in a number of international intercomparison measurements.

Key words: alpha-spectrometry; calibration; intercomparison; radon measurement; reference instrument; surface barrier detector.

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1. Introduction

In the early 1970s it was recognized that radon in indoor air in Sweden was the source of a significant radiation dose to the population. As the dose assessment is based on measurements of radon and its decay products the need for coordinated calibration of measuring instruments became obvious.

A technique for calibration of a reference instrument for radon measurement was developed and adopted. The reference instrument used at that time was an ionization chamber with a measuring volume of 18 L [1]. In 1987 the ionization chamber was replaced by an instrument with higher sensitivity and accuracy. This instrument is based on α-spectrometric measurements of radon daughters electrostatically collected on a surface barrier detector. The instrument is designed according to the principles described by Wicke [2], and has a measuring volume of 10.8 L.

The calibration of both instruments is traceable directly to the National Institute of Standards and Technology (NIST) through the use of its Standard Reference Material Radium-226. The reference source consists of the NIST radium solution in a set of glass containers.

To collect and transport radon samples for calibration or measurement, a steel container of volume 4.74 L is used. The same transfer and measuring procedure is used both for calibration and for the determination of radon concentrations in air samples. A description of the procedures used, together with estimates of the uncertainties associated with the calibration technique, measuring method and instrument used are given below.

2. Materials and Methods

2.1 The Reference Source

Figure 1 is a schematic diagram of the radon reference bubbler. The three gas washing bottles are made of glass and fused to form a single unit. Also the stoppers and the valves are made of glass to ensure a radon-tight system. The first gas washing

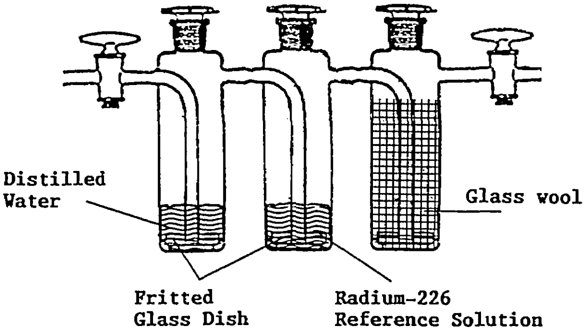

Figure 1. The reference source.

bottle contains distilled water, the second the NIST Radium-226 Standard Reference Material and the third glass wool. The total volume of the system is approximately 500 cm^3 and the volume of the reference solution is 54 cm^3.

When flushing the system with air, the air is first humified in the water bottle, then passes through the reference radium solution and finally through the glass wool, where water droplets will be removed from the radon-laden air stream.

Radon-free air, from a cylinder of aged air kept for the purpose, is used to flush the reference bubbler to remove accumulated radon. A volume of aged air of 4.74 L is used and complete removal of radon is achieved. A second flushing, immediately after the first, with the same volume of aged air removes less than 0.06% of the radon removed initially.

The build-up of radon-222 in the system is determined by the radium-226 activity of the reference solution and the decay constant of radon-222. For calibration purposes build-up times between 7 h and 14 d have been used. Shorter build-up times can be used but this increases the uncertainty in the radon-222 activity accumulated. At build-up times longer than two weeks, any small leakage of radon from the glass bottles will also influence the uncertainty of the calibration. Leakage of radon from the reference source has been checked and the uncertainty in the accumulated radon-222 due to leakage is less than 0.2%.

The NIST Radium-226 Standard Reference Material is certified to contain 31.33 Bq as of April 1978, with an uncertainty of 1.53%. The uncertainty in the radon activity removed from the bubbler is calculated to be 1.6%, which is the sum of the uncertainty in the reference activity and the removal error.

2.2 The Transfer Container

A commercially available steel container normally used for propane gas is used for sampling and calibration. Its volume has been determined by two independent methods.

The first method is based on comparison with the known volume of another metal container. The design of this second container is chosen so that its volume can be calculated accurately from its dimensions. The comparison of the two volumes is by an air-pressure measurement. One of the containers is evacuated to a very low pressure and then connected to the other container which is held at atmospheric pressure. The gas pressure in the containers are monitored before and after the connection. The volume of the "sample and transfer" container from repeated measurements was found to be 4734±3 cm^3 (1 S.D.). The systematic uncertainty is estimated to be 9 cm^3 (0.2%).

The second method used for determination of the volume is based on a water displacement technique. The container's outer volume is determined by measuring the volume of water displaced by the container when it is submerged. The volume of the metal in the container is assessed from the weight of the container and the density of the metal. In this way the volume is found to be 4753±19 cm^3 (±0.4%), total overall error.

From these two determinations the volume of the sample and transfer container is estimated to be 4740 cm^3 at 23 °C, with a maximum error of 0.5%.

2.3 The Reference Instrument

The reference instrument for the determination of radon-222 in air is based on α-spectrometric measurements of polonium-218 and polonium-214 electrostatically collected on a surface barrier detector. The instrument is built according to the principles and experiences described by Wicke [2].

Figure 2 is a schematic diagram of the instrument. The sensitive volume of the instrument is a sphere of aluminum with a volume of 10 810 cm^3 that contains the radon sample to be analyzed. In the surface of the sphere is a surface barrier detector of 150 mm^2 active area which is electrically isolated from the metal sphere.

A potential of 8 kV is applied between the detector surface and the metal sphere generating an electrostatic field that moves the charged polonium-218 to the detector surface. The decay of

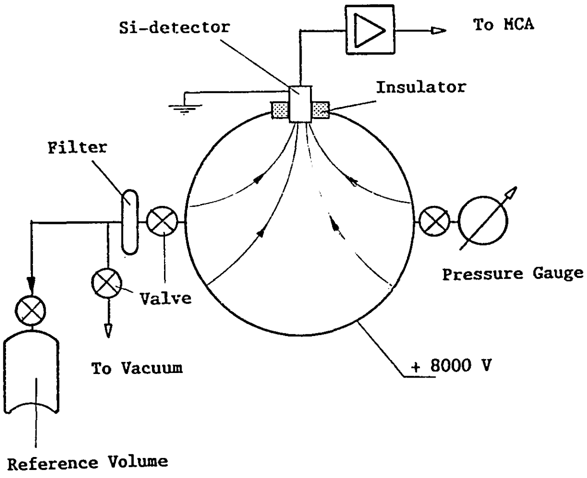

Figure 2. The reference instrument.

radon-222 within the sphere will generate polonium-218 at a rate proportional to the radon-222 activity. A build-up of polonium-218 activity on the detector surface takes place and after approximately 20 min collection a steady state is reached. The polonium-218 activity detected by the α-detector is then proportional to the radon-222 activity in the sphere.

To obtain accurate results from the instrument, the collection efficiency of polonium-218 on the detector surface and the α-counting efficiency must be stable.

The volume of the detector sphere is of no importance for the calibration and measurements but must be constant. Great efforts have been made to ensure the air tightness of the sphere.

To estimate the fraction of the polonium-218 atoms formed in the sphere that are collected on the detector surface, however, the volume must be known. From the geometrical dimensions, the volume has been determined as 10 790 cm^3 and from pressure comparison measurements with a known volume as 10 820 cm^3. A value of 10 810 cm^3 is adopted with a maximum error of ± 100 cm^3 ($\pm 1\%$).

The collection of polonium-218 on the detector surface and the counting efficiency of the α-particles from polonium-218 depend on a number of parameters such as the geometry and strength of the electric field, and the air pressure and humidity within the detector volume.

Parameters have been chosen so as to keep the parameter sensitivity as low as possible. The following apply to the most important parameters.

- The geometrical position of the detector affects the form and strength of the electric field between the chamber walls and the detector surface.

- The voltage generating the electric field and the air pressure inside the counting volume are also of concern. We have chosen 600 mbar working pressure and a high voltage of 8 kV. This gives a good margin against electrical breakdown while the collection efficiency as a function of the voltage applied is well within a flat plateau.

- The collection efficiency is affected by the humidity within the counting volume. To avoid this problem the air sample is dried during collection into the sample or transfer container. Different techniques have been tested but sampling through a drying agent (magnesium perclorate) fulfills the requirements. The sample has a humidity of less than 1 $\mu g/L$, corresponding to a dew point of less than -30 °C.

The surface barrier detector is connected to a multi-channel analyzer. Figure 3 shows a typical α-spectrum. The energy resolution of the detector separates almost completely the α-particles from polonium-218 and polonium-214. The α-particle count-rate within the energy band 4.34 to 6.16 MeV is used for evaluation. The affect of electronic instability has been studied and is negligible. For measurement of low radon-222 concentrations, correction for background counts is needed. The background with radon-free air corresponds to a radon-222 concentration of 0.6 Bq/m^3.

Adsorption of radon onto the inner surface of the sphere has been observed. As the sphere is made of aluminum the metal surface will be covered with a thin layer of aluminum oxide. It is reasonable to assume that the oxide layer causes this

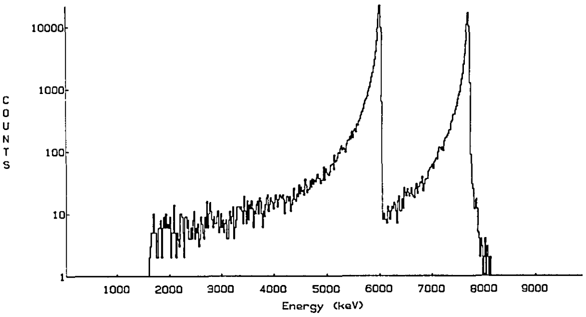

Figure 3. The d-spectrum from the reference instrument.

effect, as it is known that aluminum oxide adsorbs other gases. The effect is small and reproducible. A reduction in the radon content in the sphere of 0.1% per h is found and corrected for.

2.4 Calibration Procedure

2.4.1 Preparation of the Radon-222 Reference Concentration

- The reference source is flushed with 4.74 L of radon-free air and the valves of the reference source are then closed. The date and time are noted and the source is left undisturbed for the build-up of radon.

- The transfer container and the detector volume of the reference instrument are evacuated to less than 1 mbar.

- The transfer container is connected to the outlet of the reference source via a short tube containing the drying agent. A plastic bag filled with radon free air is connected to the inlet of the reference source. When the valves of the container and the reference source are opened, the radon is sucked into the container. The procedure is standardized to take 5 min.

- The valves are closed and time, temperature and ambient air pressure are recorded.

- As the volume of the transfer container and the radon-222 activity are known, the air concentration of radon-222 in Bq/m^3 of dry air in the container can be calculated.

2.4.2 The Transfer of Radon to the Reference Instrument

- The transfer container is connected to the reference instrument by a tube. A particle filter is inline at the entrance to the detector. An additional tube, provided with a valve, is connected by a T-piece to the tube between the container and the instrument.

- The above mentioned tubes are evacuated and the valve on the tube is closed leaving the connection between container and instrument under vacuum.

- The valve at the instrument inlet is opened and when the valve on the transfer container is opened the pressure difference will transfer about 70% of the radon in the container to the detector volume.

- The two volumes should be at the same temperature during the transfer procedure.

- The procedure takes about 10 min. The last 5 min establishes pressure equilibrium between the two connected volumes. The air pressure inside the detector will be approximately 300 mbar.

- There is no need to know exactly the amount of radon transferred. The relative amount, however, must be the same on every occasion. This depends only on the volumes of the transfer container and the detector.

- Time and temperature are recorded when the transfer is completed.

- Radon-free air is then added to the detector until the air pressure reaches 600 mbar. The valve at the detector is then closed.

- The electrostatic field for the detector is switched on and 30 min allowed to elapse to establish a steady state activity of polonium-218 on the detector surface.

2.4.3 Counting and Evaluation

- α-spectrometric counting is carried out for a sufficient time to ensure that adequate counting statistics are achieved.

- The mean net count rate of α-particles from polonium-218 within the energy band 4.34 to 6.16 MeV is calculated. Corrections for radioactive decay and adsorption of radon are applied and the calibration factor in Bq/m^3 of radon in dry air per count/s is evaluated.

2.5 Procedure for Determination of Radon Concentration in Air Samples

2.5.1 Sample Collection

- The same, or identical, transfer containers are used for sample collection and calibration purposes.

- The container is evacuated to less than 1 mbar and a short tube containing the drying agent is connected to the inlet of the container.

- The sample is taken by opening the valve on the container. The filling takes 5 min. The valve is left fully open for another 5 min to ensure that pressure equilibrium is reached. The valve is then closed and the temperature, pressure, and humidity of the ambient air as well as the date and time are recorded.

- The sample container now contains dry air at the ambient air pressure. Thus the radon concentration in the container is slightly higher than in the ambient air.

2.5.2 Transfer of Radon The transfer of radon to the reference instrument is done by the same procedure as is used for calibrations; see section 2.4.2.

2.5.3 Counting and Evaluation Counting and the evaluation of the spectrum is carried out with the same procedure as is used for calibrations; see section 2.4.3. The radon concentration in dry air is then calculated using the calibration factor, and a correction is applied to compensate for the low humidity in the container. The corrected value will then be the radon concentration in Bq/m^3 in air, but only for the temperature, pressure, and humidity existing during sampling.

3. Calibration Results and Error Calculations

Calibration factors from 30 independent calibrations with radon concentrations between 300 to 6500 Bq/m^3 are summarized in figures 4 and 5. The mean value of the calibration factor is 944 Bq/m^3 per count/s with a standard deviation (S.D.) of 1.2% and a standard error (S.E.) of 0.2%, which indicates the precision of the calibration factor. The uncertainty arising from counting statistics is 0.6% (1σ).

Known systematic errors in the calibration factor stem from the uncertainty in radon activity removed from the bubbler, 1.6%, and the volume of the transfer container 0.5%. Systematic errors due to thermal and pressure disequilibrium during the calibration procedure and uncertainties related to temperature and pressure measurements are estimated to be less than 1%. Thus the total systematic error in the calibration factor is less than 3.1%.

The uncertainty in the calibration factor, calculated as the linear sum of the standard error at 99% confidence level and the maximum likely systematic error, is 0.6%+3.1%=3.7%.

The precision of the measurement of radon samples is calculated from the sum of the uncertainty arising from counting statistics and the precision of repeated measurement of calibration samples. The sum of the variances is used to calculate the precision. The precision from repeated measurements of calibration samples is 1.0% excluding counting statistics. Assuming an uncertainty arising from counting statistics of 1.0% (1σ), the precision of a measurement will be 1.4% (1σ).

For radon sample measurements, systematic errors other than those in the calibration factor are estimated to be at most 0.5%—thus there is a maximum systematic error of 4.2% in these measurements.

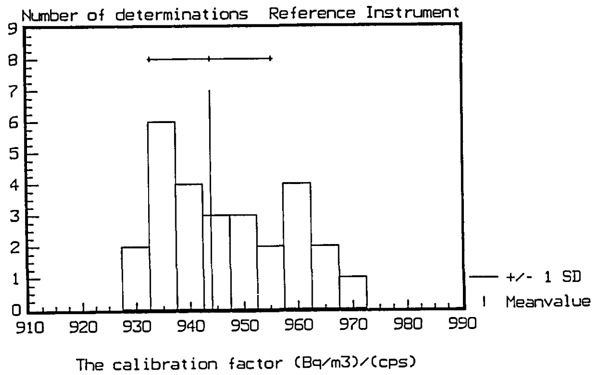

Figure 4. The distribution of the calibration factor.

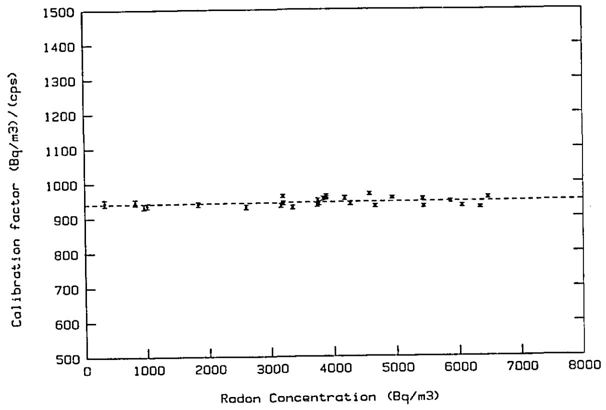

Figure 5. Calibration of radon reference instrument.

4. Results from Intercomparison Measurements

The results from the third Commission of the European Communities (CEC) intercomparison of active and passive detectors for the measurement of radon and radon decay products at the National Radiation Protection Board (NRPB), England (1987) [3] and from intercomparison measurements with the Environmental Measurement Laboratory (EML) in New York (1984) [4] are summarized in table 1.

Table 1. Intercomparison results

Ratio SSI[a]/NRPB	0.95
Ratio SSI/EML	0.96

[a] SSI=National Institute for Radiation Protection, Stockholm, Sweden.

Before 1987 an ionization chamber was used as the reference instrument [1]. The calibration procedure used was in principle the same as described above, but the uncertainty obtained with this instrument was larger. Figure 6 shows the results from calibration measurements during the years that the previous instrument was used. Intercomparisons with EML have been performed on seven occasions in the period 1982 to 1987 [4]. The results are summarized in figure 7. Compared to EML our ionization chamber gave as a mean 9% lower values. The origin of this difference may be due to larger uncertainties of the instrument used previously and the different radium-226 reference solutions used for calibration at EML and SSI.

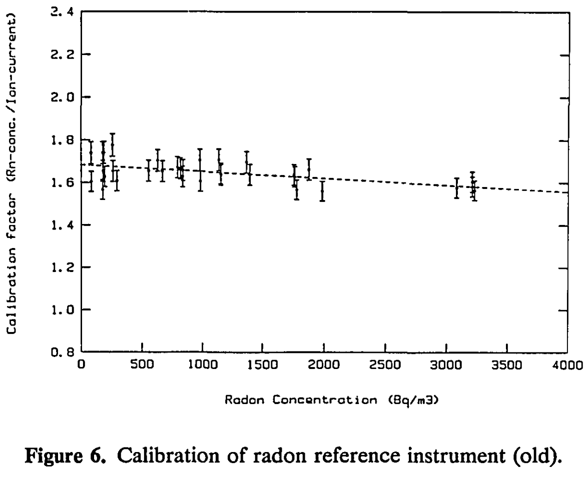

Figure 6. Calibration of radon reference instrument (old).

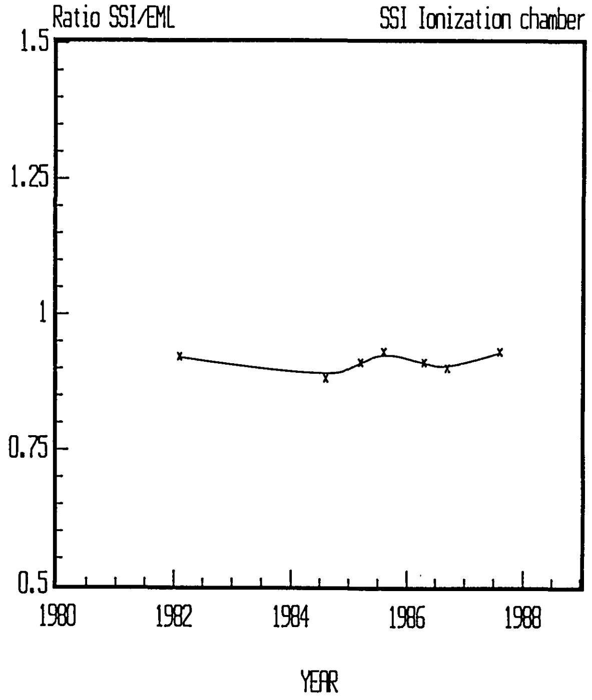

Figure 7. Intercomparison measurements of radon with EML, New York.

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Bureau of Mines Method of Calibrating a Primary Radon Measuring Apparatus

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One important requirement for accurate monitoring of radon in working environments, dwellings, and outdoors is to ensure that the measurement instrumentation is properly calibrated against a recognized standard. To achieve this goal, the U.S. Department of Interior Bureau of Mines (BoM) Radiation Laboratory has participated since 1983 in a program to establish international radon measurement standards. Originally sponsored by the Organization for Economic Cooperation and Development (OECD), the program is also sponsored by the International Atomic Energy Agency. While the National Institute of Standards and Technology (NIST) radium solution ampules are acceptable to all participating laboratories as a primary standard, a method of transferring radon from the NIST source into each laboratory's primary counting apparatus is a critical problem. The Bureau's method transfers radon from the primary solution by bubbling 3 L of air through it into a steel cylinder. After homogenizing the radon concentrations in the cylinder, eight alpha-scintillation cells are filled consecutively and measured in a standard counting system. The resulting efficiency is 81.7±1.2%.

Key words: calibration; measurement; radon.

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1. Introduction

A major part of the U.S. Department of Interior Bureau of Mines (BoM) radiation hazard program has been the establishment of standards for the measurement of radon and radon daughters, both for the Mine Safety Health Administration and private industry. Consistent with this objective, the Bureau was invited to participate in a cooperative effort to establish international measurement standards. The first publication of the results of this program [1], Part 1, Radon Measurement, pointed out that there is a 7% disagreement among the four participating laboratories and that further work is needed. These discrepancies were also reported by the U.S. Department of Energy (DoE) Environmental Measurements Laboratory [2]. Surprisingly, this disagreement was largely unnoticed even as late as 1988 [3].

2. Description of the Method

Figure 1 shows the schematic of the Bureau's system. Figure 2 is a photograph of the system comprising the de-emanation flask, the connecting tubing and a steel cylinder for the transferred radon.

The first step in the procedure is to evacuate the 3052±1 cm^3 steel cylinder and connect it to the radium solution/frit system. The inlet to this system has a flowmeter to monitor the flow rate. Control of the flow is accomplished by carefully operating valve #1, upstream from the solution. The second valve is opened all the way for the transfer operation.

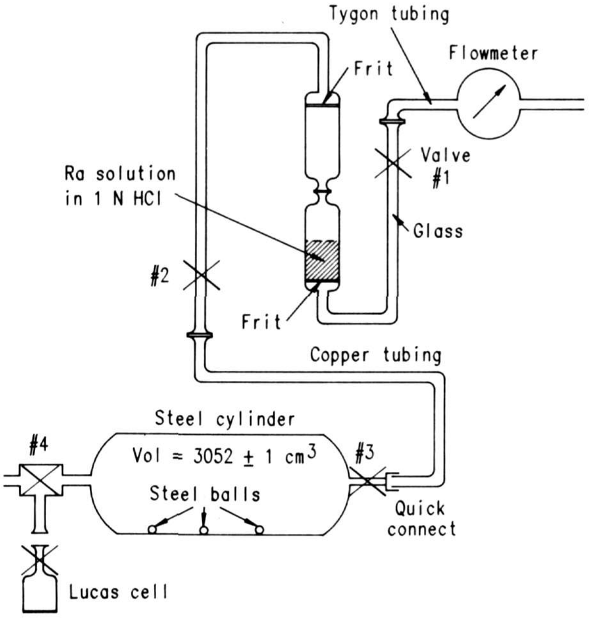

Figure 1. Schematic of the BoM method of calibrating a primary radon measuring apparatus.

After the radon gas is collected in the cylinder, the cylinder is disconnected and shaken vigorously for several minutes so that five steel balls inside homogenize the gases, ensuring a uniform concentration. It has been shown that failure to homogenize results in large errors.

Evacuated alpha-scintillation cells (commonly known as Lucas cells) are then connected to the cylinder and samples taken of the radon-containing air using valve #4; each consecutive sampling is corrected for the diminishing concentration of radon in the steel cylinder. Cells are then pressurized to 800 Torr, a standard method adopted at the Bureau to avoid the need to make pressure corrections. The magnitude of this correction is shown in figure 3. The results for four consecutive runs (32 hourly measurements in total), the efficiencies, and the calibration factors are given in table 1 and figure 4. In addition to the BoM results, the results reported by Lucas [4] from Argonne National Laboratory (ANL) are also shown in figure 4. Note that the BoM cell volume is 102 cm^3, whereas that used by Lucas is 92 cm^3.

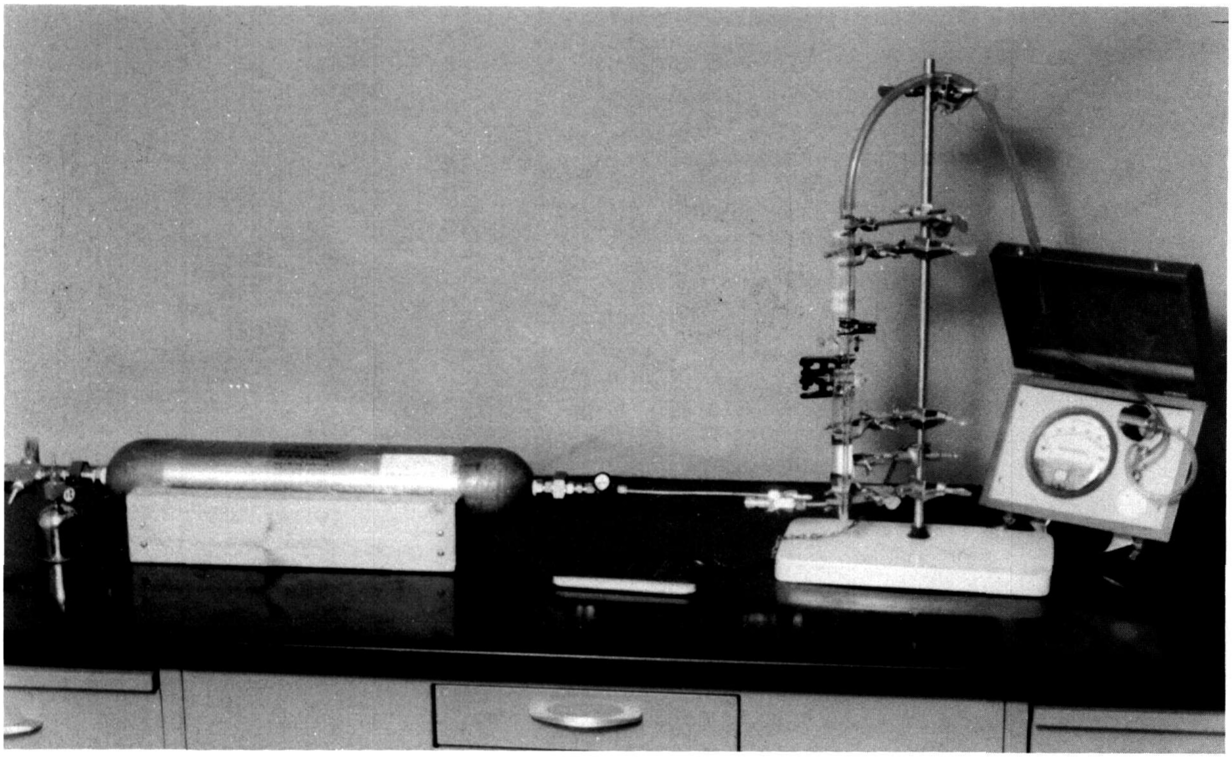

Figure 2. Photograph of the BoM system.

Table 1. Efficiencies for two runs, each repeated twice

Cell					Average	%SD
1	0.8097	0.8168	0.8165	0.8139	0.8142	0.40
2	0.8053	0.8201	0.8118	0.8094	0.8117	0.76
3	0.8228	0.8342	0.8299	0.8260	0.8282	0.60
4	0.8025	0.8064	0.8118	0.8088	0.8074	0.49
7	0.8050	0.8122	0.8077	0.8186	0.8109	0.73
8	0.8277	0.8378	0.8279	0.8352	0.8322	0.62
9	0.8196	0.8298	0.8223	0.8295	0.8253	0.62
10	0.8027	0.8156	0.8053	0.8126	0.8091	0.75
Total average=					0.817	±1.2%

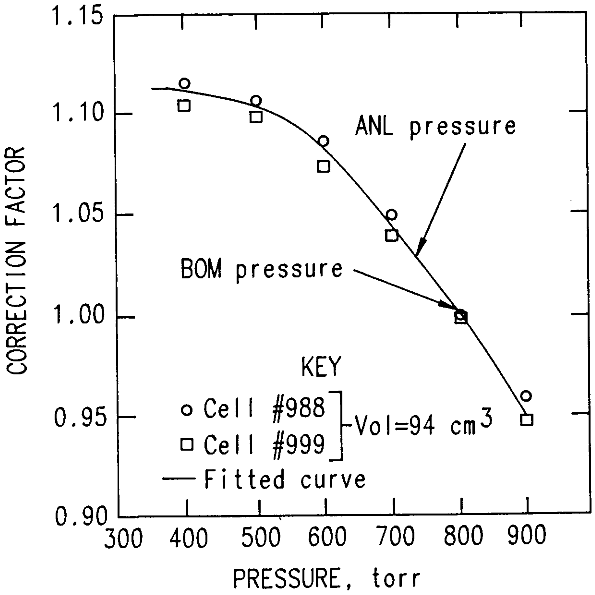

Figure 3. Pressure correction as a function of pressure inside the cell. The cells have a Ra source embedded in the window.

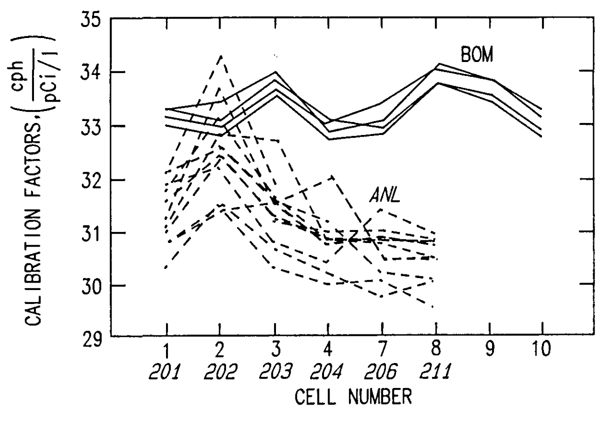

Figure 4. Plot of the BoM and ANL results. BoM and ANL cells have 102 and 92 cm^3 volumes, respectively.

3. Experimental

3.1 Determination of Cylinder Volume

A stainless steel cylinder approximately 53 cm in length and 10 cm in diameter was fitted at both ends with high-vacuum valves, as shown in figure 1. The five small steel balls of approximately 1.5-cm diameter were placed in the cylinder to facilitate mixing of the radon/air mixture. Volume was determined from valve seat to valve seat at opposite ends of the cylinder.

All procedures in this report were carried out at the normal ambient laboratory temperature of 21 °C. Accordingly, a nominal 1,000 mL burette was calibrated at 21 °C by weight of water delivered. Three successive determinations gave an average value of 996 mL delivered water per 1,000 mL indicated. Maximum deviation between the three calibrations was 1 mL. Water density at 21 °C was assumed to be 0.998 g/mL.

The stainless steel cylinder was then evacuated, the three-way #4 valve was closed, and the cylinder placed vertically beneath the calibrated burette. A length of 3/8-in Tygon[1] tubing was attached to the burette tip. The stopcock of the filled burette was opened, and water allowed to flow through the tubing until no air bubbles remained. The water-filled tube was then connected to the nipple of the valve. The unevacuated volume of this nipple was measured to be less than 1 mL. The burette was then filled to the 1,000 mL-mark, and the entire system allowed to equilibrate to 21 °C.

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[1] Certain commercial equipment, instruments, or materials are identified in this paper to specify adequately the experimental procedure. Such identification does not imply recommendation or endorsement by the National Institute of Standards and Technology, nor does it imply that the materials or equipment identified are necessarily the best available for the purpose.

The three-way valve #4 and valve #3 were opened, and the evacuated cylinder allowed to fill with water. Transfer was made in increments of 1,000 mL. No change on the final burette reading was observed over a period of 1 h. Two successive determinations resulted in cylinder volumes of 3,051 and 3,052 mL. The final accepted value was 3,052 ml at 21 °C.

3.2 Transfer of Standard Reference Material

A Standard Reference Material, No. 4953D, was obtained from NIST. This source consisted of approximately 5 g of $BaCl_2$ carrier solution in 1.4 *N* HCl. The certified Ra-226 concentration of the solution was 3.984×10^{-9} g of Ra-226 per g of solution. Transfer of the reference material to the de-emanation flask was greatly facilitated by "doll bottles" provided by Dr. Isabelle Fisenne of the DoE Environmental Measurements Laboratory. The doll bottles were small polyethylene flasks of some 7-mL capacity. The tips of these bottles were heated and drawn out to form capillary tubes some 10 cm in length.

The glass ampule containing the Ra-226 standard was opened, and the contents aspirated into a doll bottle via the capillary neck. Bottle and solution were then weighed to 0.01 mg, and the weight recorded. The Ra-226 solution was transferred quantitatively to a 100-mL de-emanation flask containing some 50 mL of 1 *N* HCl. The empty doll bottle was then reweighed to 0.01 mg. The actual amount of solution transferred was 4.95229 g, resulting in a Ra-226 activity of 19.50×10^{-9} Ci. The de-emanation flask was then sealed, with the date and time noted.

3.3 De-emanation and Transfer to the Cylinder

Air volume in the sealed de-emanation flask had previously been determined to be about 100 mL. Filling of the 3,052-mL evacuated cylinder then represented roughly 30 transfers of equilibrium vapor. Using a solubility coefficient, k, of 0.25 [5], and n equal to 30, the fraction of radon remaining in the liquid phase after filling the cylinder is given by the expression

$$Rn(\text{liquid})/Rn(\text{vapor})=(1-k)^n$$

where n equals the number of transfers of the vapor phase. Passage of some 3,000 mL of air through the de-emanation flask represents a transfer of over 99% of the equilibrium radon atoms. Previous experiments had determined that a flow rate of 100 mL/min should not be exceeded in order to prevent any possible liquid phase transfer to the steel cylinder. To further minimize any possibility of this occurrence, high efficiency glass filters were placed in the fittings at both ends of the cylinder. Accordingly, the steel cylinder was evacuated to 630 Torr, and the three-way valve closed. The cylinder was then connected to the de-emanation flask as shown in figure 1. The cylinder valve and the de-emanation flask stopcock were opened and the date and time recorded. Flow rate was monitored by means of the flowmeter at the entry port of the de-emanation flask. At the end of 43 min, a small, positive flow rate of about 3 mL/min was still observed. At this time, both valve and stopcock were closed, date and time recorded, and the cylinder disconnected from the de-emanation flask.

3.4 Loading and Counting the Cells

Evacuated cells were loaded through the three-way valve #4 (fig. 1) and pressurized to 800 Torr. The radon-free air used for pressurization was taken from an ordinary steel cylinder determined to be free of Ra contamination. The pressurization was measured in each cell to ±1 Torr. The pressurized cells were counted overnight at 1-h intervals using conventional photomultiplier tubes and counting equipment. The measured activity was extrapolated to time zero using a radon half life of 3.8235 days. Results were recorded and the standard deviation δ was calculated.

4. Discussion

Results, summarized in table 1, show the overall efficiency to be 81.7±1.2% in contrast to the 84.6±1.9% that ANL reported. Theoretical counting errors were calculated to range between 0.34% to 0.48%, as opposed to the observed values appearing in the last column of table 1.

A list of possible errors, summarized in table 2, includes:

1) Loss of material during the transfer from the doll bottle into the de-emanation flask. This error would result in a lower calibration coefficient.
2) Weighing errors during transfer of material. This error could result in either positive or negative deviation.

Table 2. Possible errors

	Effect on calibration coefficients		Remarks
	Higher	Lower	
Incomplete transfer of radium into flask		×	Not observed
Weighing error	×	×	
Incomplete transfer of Rn into cylinder		×	Back flow eliminated by having positive flow at all times
Pressure deviations in Lucas cell	×		Overpressurizing minimizes this error
Errors in cylinder volume	×	×	Successive experiments show ±1 cm^3 repeatability
Timing error	×	×	Considered insignificant
ZnS sulfide coating differences	×	×	Difficult to determine and eliminate
Changing plateout	×	×	Difficult to determine and eliminate

3) Incomplete transfer of radon into the steel cylinder. It seems that this could happen only if radon in the cylinder streamed against the flow into the system—an unlikely case. Both this error and error number 1 would lead to smaller calibration factors and efficiencies—exactly the opposite direction from that needed to bring the Bureau's results into agreement with the other laboratories [1]. On the other hand, a 2% to 4% loss could explain the Bureau's difference from Argonne National Laboratory results [4].
4) Pressure deviations in the Lucas cell could cause errors. The pressure correction curve, shown in figure 3, was obtained by using two cells that had a radon source incorporated into their window. Note that at Denver's elevation, the error is about +6%.
5) Errors in determination of the volume of the steel cylinder.

   There are two cases:
   a) The assumed volume would be greater than the true volume, leading to a lower calculated efficiency. However, the error would have to be about +60 cm^3 compared to our stated error of ±1 cm^3, thereby excluding this possibility.
   b) The assumed volume would be smaller than the true value. This is more likely than 5a. The presence of an unrecognized volume somewhere in the system might explain, for instance, why the ANL intercomparison results are lower than the Bureau's.
6) Timing error is not very significant because even a 20 min error in timing corresponds to only 0.25% of the time-zero count rate.
7) Differences among cells because of variations in photon yields from alpha-particle bombardment of the ZnS layer. This error probably accounts for the systematic variations shown in figure 4. It is not clear how to deal with these errors without spending a great deal of effort in perfecting the Lucas cells.
8) Different plateout characteristics on the ZnS-covered surfaces vs the window surface of Lucas cells. The role of natural convection due to temperature differences between inside air and the walls was investigated [6] and found to be a possible source of error. To determine the magnitude of this error would require a great deal of effort, also perhaps not justified in view of the fact the error is believed to be relatively small.

5. Conclusions

The Bureau's method of transferring radon from NIST solution into Lucas cells provides a reliable and relatively permanent system for primary calibrations. The recognized systematic errors in this method appear to be in the 1%–2% range.

6. Acknowledgments

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A Calibration and Quality Assurance Program for Environmental Radon Measurements

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The ideal facility for assessing the quality of radon measurements at environmental levels consists of: (1) an instrument whose response to radon and its progeny is determined from measurements of a certified or standard ^{226}Ra source, and (2) a calibration room with a known radon concentration.

The linkage between these two elements and additional quality control requirements are discussed here for some Environmental Measurements Laboratory radon measurements programs.

Key words: calibration; fast pulse ionization chambers; quality assurance; radon; radon calibration facility.

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1. Introduction

The Environmental Measurements Laboratory (EML) of the U.S. Department of Energy (DoE) has considerable experience in the measurement of environmental levels of ^{222}Rn. The mainstay of radon measurement programs at EML has been fast pulse ionization chambers. Originally designed and constructed in 1948 by the Health and Safety Division of the U.S. Atomic Energy Commission, New York Operations Office, the chamber systems incorporated a number of innovations which greatly simplified and improved the measurement of radon in breath and air samples.

The original set of four pulse ionization chambers has increased to nine over the intervening 40 years. The development of a reliable and flexible radon calibration chamber at EML completed the requirements for a facility to assess the quality of environmental radon measurements.

2. The EML Pulse Ionization Chamber Systems

An extensive report [1] has been published which describes the design of the EML fast pulse ionization chamber systems, the methods of calibrating these chambers and the programs dependent on measurements performed in these chambers. The unique features of the chamber systems will be briefly reviewed in this report.

An EML cylindrical stainless steel ionization chamber has a volume of 1.78 L and a nominal total volume of 2 L, including the purification system. Each chamber is constructed with a plug in the baseplate to accommodate an electrodeposited standard for determination of the alpha counting plateau. The purification system for the removal of oxygen and water vapor from air and breath samples has three advantages over other fast pulse detection systems: simplicity; a small dead space to chamber volume ratio; and dichotomy, i.e., simultaneous access to one or two ionization chamber.

To perform a radon measurement, the air or breath sample container is overpressured with hydrogen gas. The sample is transferred to the evacuated system in which the gas flow rate is regulated by capillary orifices. The oxygen in the sample and the added excess hydrogen combine to form water after passing through cartridges containing platinum black catalyst. The water vapor is removed with a calcium chloride column. The transfer of the sample to an ionization chamber is completed by the flow of forming gas (85% nitrogen, 15% hydrogen) through the sample vessel and purification system into the chamber. The chamber is pressurized to 35 kPa gauge, and then sealed and the sample is measured for a minimum of 17 h.

2.1 Variables in the Calibration of the Pulse Ionization Chamber Systems

The EML pulse ionization chamber systems have been calibrated with several different series of National Institute of Standards and Technology (NIST) Standard Reference Material (SRM) ^{226}Ra solutions by the radon emanation method since 1961. During the ensuing 28 years, a number of variables have been checked empirically to determine any effect on the calibration factor.

Two types of bubblers or gas washing towers have been used and both have been tested to determine the transfer efficiency from solution to the chambers, the possible effect of acidity or alkalinity on the gas transfer, the rate of loss of radon from the bubblers, and the quantity of ^{226}Ra in the bubbler. A significant finding was that the addition of a few drops of wetting agent such as aerosol OT to a radium-bearing acid solution in the bubbler and refrigeration of the bubbler prior to emanation yielded an efficient and reproducible transfer of radon (see reference [1]).

The effect of pressure within the pulse ionization chamber on the calibration factor was checked by measuring the radon emanated from a solution at both atmospheric and 35 kPa gauge pressure. No difference was found in the calibration factor.

The effect of the gas mixture in the ionization chamber was checked to determine whether this would affect the calibration factor. First a 0.16 L room air sample to which hydrogen had been added was transferred to the chamber, then a ^{226}Ra standard solution was emanated into the same chamber. No difference was found in the calibration factor from that determined by emanating radon from a ^{226}Ra standard solution with forming gas.

To determine whether substantial shifts occurred because of the gas filling, tests were made using the pulse size spectra of the forming gas background counts, an emanated NIST SRM ^{226}Ra solution and air samples. Only one ionization chamber was used in this series of measurements. For this chamber, the tripping level, that is, the pulse size expressed in volts below which pulses are not recorded, was 0.8 V. For the forming gas background measurements, 20% of the pulses were below 3.0 V with the median pulse size of 6.4 V and a maximum of 10.3 V. The NIST emanated ^{226}Ra solution spectrum showed 20% of the pulses below 4.2 V with a median pulse size of 6.4 V and a maximum of 10.3 V. Two 0.08-L air samples were collected in the EML radon calibration chamber and measured on consecutive days. The first sample was transferred to the ionization chamber and measured at atmospheric pressure. The second sample was transferred to the ionization chamber and measured at 35-kPa gauge. The sample spectra were identical and showed that 20% of the pulses were below 4.8 V, the median pulse size was 6.0 V and the maximum was 9.5 V. Lastly, a 0.16-L ^{220}Rn (thoron) sample was collected in the EML radon calibration chamber and measured in the ionization chamber at a pressure of 35 kPa gauge. The thoron sample spectrum showed that 20% of the pulses were below 4.0 V, the median pulse size was 6.4 V and the maximum was 10.3 V.

It was concluded from these series of measurements that the ionization chambers are insensitive to the gas mixture as long as oxygen and water vapor are removed and also that there is no difference in the calibration factor for the chambers determined at atmospheric or 35 kPa gauge.

3. Summary of NIST SRM ^{226}Ra Solution Certification

The calibration factors for the EML pulse ionization chambers are determined from measurements of NIST SRM ^{226}Ra solutions. Because the estimation of the systematic error for measurements in the ionization chambers includes the total uncertainty ascribed by NIST to SRM ^{226}Ra solutions, it is important to review the practices used in the certification process. Table 1 summarizes the

Table 1. NIST ^{226}Ra SRM series, 1940–1984

NIST series	Series units	Calibration instruments	Total uncertainty (%)
1940	μg per 5 mL	Electroscope	?
1947	μg per 5 mL	Electroscope	0.8
1957	ng per 100 mL	Radiation balance (electroscope)	0.3 (1967–changed to 1.5)
1965	μg per 5 mL	Radiation balance 4 π-γ ionization chamber	0.5
1967	μg per 5 mL	Radiation balance 4 π-γ ionization chamber	3.6
1978	pg per g	4 π-γ ionization chamber compared to 1957 SRM	1.34
1984	pg per g	4 π-γ ionization chamber calibrated with national radium standards	1.30

evolution of the NIST certification process from 1940 through 1984. Over this period, significant changes have been made in the choice of the primary instrument used in the NIST measurements and in the way in which the ^{226}Ra value is certified. Except for the 1940 series, the information in the table was taken directly from certificates of ^{226}Ra solution used to calibrate the EML pulse ionization chambers. Historically, the ^{226}Ra solutions have been certified in mass units. Until 1978, the certification was made on a mass per ampoule basis. Since that time the certification has been given as a concentration, but still as mass of ^{226}Ra g^{-1} of solution. However, a suggested half-life for ^{226}Ra did not appear on the certificate until 1984.

The 1940 and 1947 ^{226}Ra series were certified on the basis of comparative measurements against the national primary radium standard using a gold-leaf electroscope [2]. The total uncertainty for these measurements was less than 1%. In 1957, the primary instrument for the ^{226}Ra measurements was the radiation balance [3] with the electroscopic measurements used for confirmation purposes. The standardization was by direct comparison with the national primary radium standards. A reevaluation of this series a decade later resulted in an increased uncertainty. The 1965 series "was calibrated by comparing its γ ray emission rate with those of a series of standards prepared for material that was compared, in the NIST radiation balance, with the national radium standards. The γ-ray emission rates were compared in the NIST 4 π-γ ionization chamber." The estimated overall uncertainty was given as 0.50%. The 1967 series was calibrated by comparison with the 1957 series in the radiation balance and 4 π-γ ionization chamber. The uncertainty was quoted as 3.6%. The 1978 series marked the first time that ^{226}Ra was certified on a concentration basis, that is, the mass of ^{226}Ra g^{-1} of solution. Once again the material was certified by comparison of its γ-ray emission rate with the 1957 series of standards. Only the 4 π-γ ionization chamber was used in this certification process. The estimated total uncertainty for this series was 1.34% for the concentration value and 1.53% for the ampoule value. The series was certified from measurements made in the pressurized 4 π-γ ionization chamber calibrated with the national radium standards. The uncertainty in the ^{226}Ra concentration was assessed as 1.30%.

The total uncertainty in NIST SRM ^{226}Ra solution values has generally been on the order of 1%, but it is only one of several possible systematic errors which must be considered in the estimation of total uncertainty by an SRM user.

4. EML Pulse Ionization Chamber Calibration Factors and Estimated Total Uncertainty in Radon Measurements

The EML pulse ionization chambers have been rigorously calibrated five times between 1960 and 1984 using NIST SRM ^{226}Ra solutions. In each case, the SRM ^{226}Ra was from a different series. The determination of the calibration factors for the ionization chambers requires 2 to 3 months. The 1984 calibration of the ionization chambers will be used to illustrate the process at EML. The details of previous calibrations of the chambers are documented in reference [1].

When the 1984 NIST SRM ^{226}Ra series became available, EML purchased several ampoules. One ampoule of SRM 4950 E having a ^{226}Ra concentration of 7.566×10^{-11} g per g of solution was selected for the program. Nine emanation flasks or bubblers, which had been checked and found to be free of contamination, were needed for the measurements. A measured amount of 1*N*HC1 was added to each flask. The NIST SRM ^{226}Ra solution was transferred to a plastic ampoule used for weighing and delivery of solutions. A weighed aliquot of the ^{226}Ra solution was delivered to each of the nine bubblers. The bubblers were de-emanated with forming gas to establish time=0 for the build up of radon. Over the course of the program build up periods ranged from 2 to 10 d. The measurement protocol was devised as a 9×9 matrix, that is, the radon from each of the nine bubblers is emanated once into each of the nine chambers. This allows us to detect any bias in a particular bubbler or chamber. The redetermined calibration factor for the chambers was calculated from the 81 measurements. Only a single calibration factor need be used for the nine chambers since the mean value obtained for each chamber agrees with the remaining eight chambers within the standard deviation of the measurements. The 81 measurements have been shown to be normally distributed.

Table 2 summarizes the results of the major calibrations of the EML ionization chambers and gives an estimation of the magnitudes of the random and systematic errors associated with the measurements. The EML total uncertainty is the linear sum of the errors.

5. Quality Control and Quality Assurance Program for the EML Pulse Ionization Chambers

As with any instrument, routine checks and maintenance are required for proper performance of the pulse ionization chambers. The background count rate of each chamber is measured with forming gas every weekend and occasionally during the work week to ensure against temporal bias. The background count rates for the nine chambers in service at present range for 12 to 20 counts h^{-1}. A control chart of the weekend background count

Table 2. EML pulse ionization chamber calibrations by the radon emanation method

Year	NIST series	EML calibration factor (cph pg^{-1})
1960	1947	221 (1.8%)
1961	1957	225 (2.2%)
1975	1965	236 (3.0%)
1978	1978	236 (3.0%)
1984	1984	229 (2.2%)
Mean and SD	229 (3.1%)	
Random errors: 3.1% SD from ionization chamber measurements	Systematic errors: 1.30% NIST uncertainty 0.10% Rn half-life 0.44% Ra half-life 0.25% gravimetric measurements	
Estimated Total Uncertainty:	5.2%	

rates is maintained for each chamber. At the beginning of the work week, the average background count rates for the current year and the running average including the results from previous years are calculated for each chamber. The weekend background information is used to initiate corrective actions if deemed necessary. Over a period of years, the background count rate of a chamber increases due to the build up of long-lived radon progeny on the interior surfaces. The increase is a direct result of exposure in terms of Bq h^{-1}. The background count rate may be reduced by dismantling and electropolishing the chamber. This procedure requires a lengthy recalibration of each chamber and is therefore undertaken only after a period of several years.

Occasionally the ionization chambers systems are checked for electrically generated noise by filling the chambers with room air. The oxygen in the air effectively reduces the pulse size below the tripping level of the electronics and only electrically generated pulses are registered during the overnight measurement period. The electronic "noise" in the chamber systems is less than 1 count in 4 h.

The platinum black catalyst and calcium chloride columns are kept free of water vapor by maintaining these cartridges under vacuum except during sample introduction.

The calibration factor for the chambers is checked at least quarterly by emanating radon from a standard radium bubbler. It is important to emphasize that a bubbler containing radium for calibration purposes is only used for 1 year at EML.

EML participates in the Environmental Protection Agency Water Cross-Check Program for ^{226}Ra analyses. The results over the last 4 years show agreement within ±5% of the expected value. EML also administers the Quality Assessment Program for the DOE Office of Environment, Safety and Health. The ^{226}Ra analyses for soil, vegetation, and tissue are performed by radon emanation and the results are compared with those of DOE contractor laboratories. When possible, EML participates in the development of consensus standards for ^{226}Ra in natural matrix standards. These exercises are usually initiated by organizations such as the International Atomic Energy Agency and NIST.

Quality assurance for measurements of radon in air samples is provided through a series of national and international intercomparison exercises in which EML is both a sponsor and a participant. The facility used in these exercises is described in the next section.

6. The EML Radon, Thoron, and Progeny Calibration Facility

The EML radon, thoron, and progeny calibration facility was constructed to provide a range of well-controlled environmental conditions. Experiments are conducted in this calibration facility to assess the accuracy of the methods and instruments used to measure radon and thoron and their progeny in occupational and non-occupational settings. Research studies are conducted on the environmental factors which affect the physical behavior of the progeny.

The calibration facility consists of seven principal components:

1. radon and thoron source generation systems,
2. anteroom to the exposure chamber,
3. exposure chamber,
4. environmental chamber and air flow control system,
5. radon, thoron, and progeny monitoring instruments,
6. aerosol generating system,
7. environmental control panel and data acquisition system.

For the purpose of this paper our discussion will be restricted to the generation and measurement of a controlled radon atmosphere.

6.1 The Radon Calibration Chamber

6.6.1 The Physical Layout of the Exposure Chamber The EML radon calibration chamber is a 2.82×2.82×2.4 m aluminum clad chamber with a separated anteroom entering into the main room. The volume of the main room is 19.4 m^3. Entry to the exposure room through the double door system in the anteroom serves to minimize the influx of adjacent room air into the main room. There are six access ports located on one wall of the exposure room through which small instruments can be introduced or samples can be taken without entering the exposure room. The exposure room is equipped with multiple electrical outlets, fluorescent lighting and a two-way communications system. The exposure room is viewed from the outside through two large glass windows. A schematic diagram of the essential features of the calibration room is shown in figure 1.

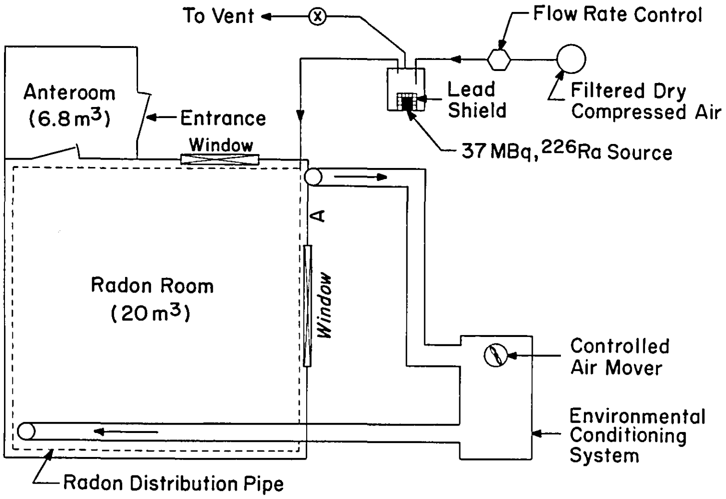

Figure 1. Schematic diagram of EML's radon calibration chamber.

6.1.2 Radon Delivery System The radon source is a 37 MBq (1 mCi) dry radium bromide source housed in a shielded 0.2 m^3 metal drum. The drum and its three input/output ports are hermetically sealed. Compressed air is filtered and dried prior to introduction into the drum. The air flow through the drum is maintained at a constant rate of 0.01 $m^3\ min^{-1}$. The outflow from the drum is introduced into a 19-mm diameter stainless steel tube extending around the perimeter of the exposure room at floor level. The tube has a series of 1100 holes spaced 1 cm apart to insure uniform gas delivery into the room. The generation drum is provided with exhaust valves to vent the radon outdoors when not in use. The radon concentration in the chamber can be controlled over a range of 37 to 3700 Bq m^{-3}.

6.1.3 Environmental Conditioning System Two 0.2-m diameter ducts with control dampers are the input/output connections to an externally located environmental conditioning system forming a closed loop with the exposure room. The conditioning system contains a refrigeration unit for temperature control and a steam generation unit for humidity control. Sensors for monitoring temperature and humidity are located in the exposure room and are displayed on the controller panel. Air flow through the system can be varied from 1.9 to 16 $m^3\ min^{-1}$ corresponding to air exchange rates of 0.09 to 0.8 min^{-1} in the exposure chamber. The exposure chamber is maintained at a slight positive pressure relative to the prevailing atmospheric pressure. This eliminates the infiltration of outside room air into the exposure chamber. The temperature and humidity in the chamber can be controlled over the range of 2 to 45 °C and 15% to 100%, respectively.

6.1.4 Continuous Monitoring of the Radon Concentration The radon concentration in the exposure chamber is monitored continuously from four different locations inside the chamber. Each monitoring system is a 2-L plastic scintillation cell mounted on a 12.5-cm diameter photomultiplier tube. The radon concentration measurements are logged into the data acquisition system at preset time intervals. Typically, hourly concentration results from each system are averaged every 3 h. The radon concentration measurements obtained with the four independent systems are usually within 5% of each other over the entire operating range. The accuracy of the monitors is checked periodically by collecting samples in the exposure chamber and measuring them in the pulse ionization chambers.

7. EML Participation in National Intercomparison Exercises

The DOE Office of Health and Environmental Research has funded a number of programs in the radon research area. In an effort to assure the quality of the results obtained from these programs as well as those sponsored by other agencies, EML developed a national radon intercomparison program which is open to any group in the public or private sector which conducts surveys or research programs on the indoor concentration of radon.

EML has conducted 16 radon intercomparison exercises since April 1981. The most recent in a series of reports that describe these exercises summarizes the philosophy and protocol for participation, as well as potential and actual problems in the collection and measurement of radon with various devices [4]. The EML staff has gained experience in the handling and filling of a variety of samplers from over 50 facilities including representation from Canada, the Federal Republic of Germany and Sweden.

At radon concentrations ranging from 220 to 3040 Bq m^{-3} it has been found that 60% to 90% of the participants report results within ±10% of the value obtained from measurements with the EML pulse ionization chambers. Virtually all participants agree with the nominal EML value within ±25%. These findings serve as an independent check on the validity of EML radon measurements.

8. EML Participation in International Intercomparisons

Since 1983, EML has expanded its efforts to include formal participation in an international quality assurance program aimed at the collection and correlation of valid information on the concentration of radon and progeny in the indoor environment assembled for epidemiological and dosimetric risk modeling purposes. Under the auspices of the Nuclear Energy Agency of the Organization for Economic Cooperation and Development (OECD) and the Radiation Protection Programme of the Commission of the European Communities (CEC), the International Intercomparison and Intercalibration Programme for Radon, Thoron, and Daughters Measuring Equipment (IIIP) was conceived and became operational in 1983. The principal feature of the IIIP is its designation of four laboratories expert in these measurements to act as regional reference centers for quality assurance activities. The four laboratories are: EML (environmental concentrations); the USDI Bureau of Mines, Denver, CO, USA (occupational concentrations); the National Radiological Protection Board, Didcot, Oxfordshire, UK; the Australian Radiation Laboratory, Yallambie, Victoria, Australia. Samplers containing radon from each regional laboratory's exposure chamber were exchanged and measured at the four laboratories. Of the four laboratories, three perform their radon measurements with scintillation cells, while EML's primary radon measurements are performed in the fast pulse ionization chambers. Three rounds of sample exchanges were completed over an 18 month period. The results of these international radon intercomparisons have been the subject of two reports [5,6]. At present, radon measurements performed at three of the laboratories agree within the replication error, while the fourth laboratory differs significantly (approximately 9% lower) from the others. The cause of this difference is under investigation and may be resolved through the measurement of a series of special radon samples prepared by NIST.

The IIIP is currently sponsored by the CEC and the International Atomic Energy Agency (IAEA), Vienna, Austria. Its activities are expected to expand in response to the development of the new IAEA-sponsored radon research program. EML will continue to sponsor regional radon and progeny intercomparisons as part of this effort as well as the U.S. Department of Energy's radon quality assurance program.

9. Summary

The EML pulse ionization chambers for radon measurements and the radon calibration room for exposures, while no longer unique, have been instrumental in alerting the scientific community to the needs, requirements, and the subsequent development of appropriate tools for quality assessment. Valuable guidance for the development of a program for the calibration, standardization, and quality assurance for measurements of radon and radon progeny in air has been given by the National Council on Radiation Protection and Measurements [7].

With the present state of technology, no suitable radon gas standard is available to calibrate instruments and assess the quality of measurements produced in measurement programs. In the absence of such a standard, calibration of an instrument must be based on a laboratory standard derived from a NIST SRM ^{226}Ra solution. As a practical approach for research and quality assurance purposes, exposure chambers with a fixed radon concentration have been constructed at a number of facilities. This represents a best effort approach to provide some form of quality assurance for a variety of purposes.

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U.K. National Radiological Protection Board Radon Calibration Procedures

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A procedure for the calibration of instruments for the detection of ^{222}Rn in air is described. The method is based on the alpha-spectrometric determination of the concentration in air of ^{218}Po in the calibration chamber. The calibration chamber is described, together with the method of maintaining a high aerosol concentration. The ^{218}Po concentration at steady state in the chamber is found to be 98% of the ^{222}Rn concentration typically. An assessment of the sources of uncertainty in the method presented indicate that the ^{222}Rn concentration in the chamber can be determined with an overall uncertainty of about 7% at the 95% confidence level.

Key words: aerosol generation; alpha-spectrometry; calibration chamber; radon daughter measurement; radon gas measurement; statistical uncertainty.

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1. The Calibration Chamber

The calibration of radon (^{222}Rn) equipment at the U.K. National Radiological Protection Board (NRPB) is carried out in a sealed chamber of 43 m^3 volume. The chamber has a double door (air-lock) allowing entry and exit with the minimum disturbance of the contained atmosphere. The ventilation rate of the chamber can be controlled by a variable speed exhaust fan and adjustable inlet apertures. When used for the calibration of instruments for the detection of radon, however, the exhaust fan is removed and all leakage apertures sealed. Under these conditions, the inherent infiltration is 0.006 to 0.008 air changes per hour as determined by the release of nitrous oxide and by monitoring its concentration as a function of time with a Miran-101[1] infra-red spectrophotometer (Fox Wilboro Inc., Norwalk, Connecticut).

Connected to the chamber is a temperature-controlled tube furnace in which carnauba wax is heated, while air from the chamber is circulated through the furnace and returned to the chamber. The furnace is operated at a temperature of 200 °C with a flow rate over the wax of 3 L min^{-1}, while a flow rate of 0.15 L min^{-1} is maintained through the annulus surrounding the boat containing the wax. The resulting aerosol has a nominal count median diameter of 0.1 μm with geometric standard deviation of 1.6. At the steady state this aerosol generator maintains an aerosol concentration in the chamber of 3×10^4 particles cm^{-3}.

Radon is generated in the chamber by bubbling air through solutions of radium-226 chloride. These liquid radon sources alone provide a steady-state radon concentration of 2500 Bq m^{-3}; with the addition of dry radon sources, about 8000 Bq m^{-3} is attained.

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[1] Certain commercial equipment, instruments, or materials are identified in this paper to specify adequately the experimental procedure. Such identification does not imply recommendation or endorsement by the National Institute of Standards and Technology, nor does it imply that the materials or equipment identified are necessarily the best available for the purpose.

2. Radon Daughter Measurement

Measurement of the radon concentration in the chamber is principally by alpha-spectrometric determination of the airborne radon daughters by the method proposed by Nazaroff et al. [1]. The decay scheme for radon is shown in table 1, together with that for ^{220}Rn (thoron). A sample of the air in the chamber is drawn through a membrane filter (type AA, 0.8 μm pore diameter, Millipore U.K. Ltd., Watford) at a nominal flow rate of 20 L min^{-1} for a period of 10 min by means of a high-vacuum pump (type EB3A, Edwards, Bristol). The total volume sampled is measured with a dry gas meter (type G4 Mk2s1, IGI Meters Ltd., London) attached to the exhaust of the pump. The gas meter reading is corrected for the temperature of the exhaust air; the systematic uncertainty in the volume measured has a semi-range of 2%. Repeated measurements with a number of clean filters has shown that the flow rate does not vary over a 10 min sampling period by more than 0.2% of the mean value. Any change in flow rate occurs during the first 3 min of the sampling period; thereafter it is constant. The control of all timing periods is effected by a personal computer (PC) as is the analysis of the measured spectra; timing errors are negligible. The membrane filter is transferred to the counting system during a 1 min interval between the end of the sampling period and the start of the first spectrum acquisition. The counting system comprises a drawer counter, to hold the filter, with a 450 mm^2 silicon surface barrier detector (type A-025-450-100, EG&G Ortec, Oak Ridge, Tennessee), a preamplifier (type 124, EG&G Ortec), a spectrometry amplifier (type 451, EG&G Ortec), and a multi-channel analyzer (MCA) (type 7500, EG&G Ortec).

Table 1. Decay schemes for ^{222}Rn (radon) and ^{220}Rn (thoron), short-lived daughters only

Nuclide	Half-life	Mode of decay	α-Energy MeV
^{222}Rn	3.823 d	α	5.48
^{218}Po	3.11 min	α	6.00
^{214}Pb	26.8 min	β,γ	
^{214}Bi	19.7 min	β,γ	
^{214}Po[a]	164 μs	α	7.69
^{220}Rn	55 s	α	6.28
^{216}Po	0.16 s	α	6.78
^{212}Pb	10.6 h	β,γ	
^{212}Bi	60.5 min	α,β,γ	6.94 25% 6.08 10% other 1%
^{212}Po	0.3 μs	α	8.78 64%

[a] ^{214}Po is always in equilibrium with ^{214}Bi.

As soon as the sample filter is in position beneath the detector, the space between the filter and the detector is partially evacuated to 8.7×10^4 Pa below atmospheric pressure by a small pump (type XX55, Millipore U.K. Ltd., Watford). When the 1 min interval following the end of the sampling period has elapsed, an alpha particle energy spectrum is accumulated for 10 min. A delay of 10 min follows before a second spectrum is accumulated over a period of 19 min. At the end of each spectrum accumulation period, the integral counts from the channels of the MCA corresponding to alpha particle energy ranges of 1.0 to 6.15 MeV, 6.15 to 8.0 MeV, and 8.0 to 9.8 MeV are acquired by the PC. These energy ranges encompass the peaks from ^{218}Po (6.00 MeV), ^{214}Po (7.69 MeV), and ^{212}Po (8.78 MeV). The data are corrected for the fraction of counts in the ^{218}Po channels due to alpha particles from ^{214}Po that are degraded in energy in their passage from the site of decay to the detector. Similarly, corrections are applied for the fractions of alpha particles from ^{212}Po that occur in the ^{214}Po channels and from ^{212}Bi that occur in the ^{218}Po channels; these last corrections are necessary as the chamber is used occasionally for the measurement of ^{220}Rn daughters. Finally, the volume of air sampled and the alpha particle detection efficiency are input to the PC, and the results are calculated to give the activity concentrations of ^{218}Po, ^{214}Pb, ^{214}Bi/^{214}Po, and potential alpha energy concentration (PAEC).

The detection efficiency for the spectrometry system has been determined by comparison with a laboratory standard alpha counter, the efficiency of which was established with a reference ^{241}Am source (Amersham International Ltd., Amersham). The alpha particle emission rate for this reference source is known with a semi-range uncertainty of 2%. A filter sample of ^{220}Rn daughters that had been taken some 16 h earlier was counted alternately on the spectrometry system and the laboratory alpha counter; this was repeated several times, corrections being made for the decay of ^{212}Pb with a half-life of 10.6 h. The efficiency of the spectrometry system was also determined using the reference ^{241}Am source directly with an appropriate correction for source-to-detector geometry. The efficiency found by the two methods differed by less than 1%.

Radon calibrations are always carried out under virtual steady state conditions. These conditions are achieved if the chamber has been undisturbed for at least 20 d (beyond 20 d a constant radon concentration exists in the chamber) or by removing all radon sources at least 3 h prior to the calibration (beyond 3 h, the state of equilibrium between radon and its short-lived daughters is constant and all decay is determined by the half-life of radon). A radon concentration in the chamber of not less than 2000 Bq m^{-3} is also used. Under these conditions, the uncertainty in the measurement of the ^{218}Po activity concentration has a standard deviation arising from counting statistics alone of less than 2%.

3. Radon Gas Related to ^{218}Po

The state of equilibrium between radon gas and radon daughters in the sealed chamber was calculated using the room model devised by Jacob [2] and refined by Porstendorfer et al. [3]. Repeated measurements of radon daughter concentrations in the chamber using the spectrometry system when the ^{218}Po concentration was assessed to be in the range 2000–2400 Bq m^{-3} yielded mean activity ratios ^{218}Po : ^{214}Pb : ^{214}Bi of 1 : 0.960 : 0.925. These data were fitted to the room model under the assumption that the deposition (plate-out) constant for radon daughters attached to the aerosol was two orders of magnitude smaller than that for unattached radon daughters. The model is quite insensitive to the value of the deposition rate for attached radon daughters: for example, changing the ratio between the deposition rate constant for attached and unattached daughters from 0.01 to 0.1 typically changes the predicted ratio of total ^{218}Po activity to that of radon gas by less than 11%. The probability that a ^{214}Pb atom formed by the decay of ^{218}Po attached to an aerosol particle will be detached by recoil [4] is taken as 0.83. Table 2 gives the range of parameter values for the room model that provide an adequate fit to the data for the chamber. The range of the ratio total ^{218}Po to radon is from 0.967 to 0.990. To account for modelling uncertainties, this ratio is taken to be 0.98 with a semi-range systematic uncertainty of 2%.

The range of the rate constant for attachment of radon daughters to aerosols implies, for an aerosol concentration of 3×10^4 particles cm^{-3}, a range in average attachment rate coefficient from 2.7×10^{-3} to 6.3×10^{-3} cm^3 h^{-1}. The attachment rate coefficient is the coefficient of proportionality relating the aerosol concentration to the rate constant for the attachment of radon daughters to aerosols. Reported values for this coefficient [5] vary from 1.8×10^{-3} to 6.8×10^{-3} cm^3 h^{-1}, and the values calculated here lie comfortably within this interval. The range of values found for the deposition rate constant for unattached radon daughters is rather lower than the range of 10 to 40 h^{-1} considered typical [6] in dwellings. Air movement within the chamber is low, however, and most surfaces are smooth and electrically conducting. The surface area to volume ratio for the chamber is about 2.8 m^{-1}. The corresponding deposition velocities for unattached radon daughters required to fit the data are in the range 1.07 to 1.79 m h^{-1}, similar to those found by Knutson et al. of 1.19 to 2.38 m h^{-1} in another metal exposure chamber [7]. It is concluded that the derived parameters for the room model are good and that the radon concentration may be taken to be 102% of the concentration of ^{218}Po in the chamber as measured by the spectrometry system, with a semi-range systematic uncertainty of 2%.

Table 2. Range of parameter values that account for the radon daughter conditions in the exposure chamber

Detachment probability for ^{214}Pb on formation	0.83
Ratio of deposition rate constants for attached and unattached radon daughters	0.01
Ventilation rate	0.006–0.008 h^{-1}
Rate constant for attachment of radon daughters to aerosols	80–190 h^{-1}
Deposition rate constant for unattached radon daughters	3–5 h^{-1}
Ratio of activity concentrations of ^{218}Po to radon	0.967–0.990
Assumed mean activity ratio, ^{218}Po to radon	0.98
Assumed range of activity ratio, ^{218}Po to radon	0.96–1.00

4. Summary of Uncertainties

All sources of uncertainty in the determination of radon concentration by the spectrometric measurement of ^{218}Po concentration are shown in table 3. There are unaccounted uncertainties, such as linearity of the spectrometry system, and it has been estimated that these produce a maximum standard deviation of 2%. The overall systematic uncer-

Table 3. Assessed uncertainties in the measurement of radon activity concentration

Systematic uncertainties		Per cent
Flow rate:	Semi-range	2
Reference ^{241}Am source:	Semi-range	2
Modelling:	Semi-range	2
Overall systematic, 95% confidence interval:		±3.9%
Random uncertainties		**Standard deviation %**
Counting statistics, determination of ^{218}Po:		2
Counting statistics, efficiency calibration:		1
Overall random, 95% confidence interval:		±4.48%
Estimated unaccounted uncertainty, 95% confidence interval		±3.96%
95% interval overall:		±7.1%

tainty, as a standard deviation, is found [8] by adding the individual semi-ranges in quadrature and dividing the result by the square root of 3. This is derived by considering each systematic uncertainty to have a rectangular distribution, i.e., all values over the range of uncertainty are deemed to be equally likely. The overall random uncertainty is obtained by the addition of each uncertainty in quadrature. The final overall uncertainty is simply the sum in quadrature of the systematic and random uncertainties derived for the same confidence level.

It is concluded that determination of radon concentration at NRPB by the alpha spectrometric measurement of the concentration of ^{218}Po provides a result that has an uncertainty of around 7% at the 95% confidence level.

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ENEA Reference Atmosphere Facility for Testing Radon and Daughters Measuring Equipment

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This paper gives the technical characteristics of the Italian Committee for Research and Development of Nuclear Energy and Alternative Energy (ENEA) radon chamber and the operational procedures developed for testing radon and daughters measuring equipment. Runs were carried out under different experimental conditions defined in terms of radon and daughter concentrations, equilibrium ratio (*F*-factor), and aerosol concentration and size distribution. Stable radon reference atmospheres with known equilibrium factors were obtained using standard aerosols.

Key words: calibration; radon and daughters measuring equipment; radon chamber; radon reference atmospheres.

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1. Introduction

Starting in 1970, the Italian Committee for Research and Development of Nuclear Energy and Alternative Energy (ENEA) developed a research program on radon test facilities [1]. Experimental radon chambers were designed to produce reference exposure atmospheres containing traces of radon and thoron progeny.

A radon test chamber of 1 m^3 was placed in operation in 1985 in the ENEA laboratory. The device allows experimental tests and calibration of radon and daughter measuring equipment under controlled climatic and aerosol conditions.

The testing of instruments used to measure radon decay products presents special problems because their accuracy of response is influenced by several factors, e.g., plate-out on surfaces, equilibrium factor, aerosol size distribution and concentration, and geometrical design and aerodynamical properties of the sampling devices. Instrument calibrations are usually carried out using reference alpha particle sources and air flow measurement standards.

The chamber that has been designed at ENEA provides reference radioactive atmospheres which tests the performance of both the sampling and detection subassembly of the instrument. Special requirements have to be fulfilled to maintain a stable particle concentration and size distribution. Investigations have been carried out to provide a range of radon daughter concentrations at different disequilibrium ratios in the chamber atmosphere. This paper briefly outlines the laboratory facilities, measuring methods, and technical equipment.

2. Description of ENEA Radon Test Facility

2.1 Criteria of Design and Applications

Procedural and technical aspects concerning laboratory calibration of radon and daughters measuring equipment have been discussed in the report of a group of experts of the Nuclear Energy Agency-of the Organization for Economic Cooperation and Development (OECD) [2]. Instruments used for the measurement of radon daughters need to be periodically calibrated against radioactive atmospheres of known characteristics. The following main parameters are used to characterize reference atmospheres:

- radon and daughters concentration;
- radon daughter disequilibrium ratios;
- unattached fraction;
- aerosol concentration and particle size distribution;
- climatic factors: relative humidity, pressure, and temperature.

The above parameters are also used to characterize atmospheres for thoron and its decay products as well as atmospheres for mixtures of radon and thoron decay products.

2.2 Technical Characteristics

The ENEA facility consists of the following main components (fig. 1):

- cylindrical stainless steel chamber;
- climatization apparatus with automatic and manual control system;
- climatic parameters monitoring and data acquisition system;
- aerosol system;
- radon sources;
- radon and daughters monitoring devices.

A characteristic of this facility is that it can be operated in a static state and under two different dynamic conditions—recirculation and dynamic open circuit. During recirculation the air mass is taken from the front side of the chamber and reintroduced through the rear wall. Under dynamic open circuit conditions, the air mass taken from the chamber is continuously exhausted outside.

2.3 Chamber Climatic System

A special climatization system[1,2] allows control of the following parameters without influencing the aerosol concentration and size distribution of the reference atmosphere:

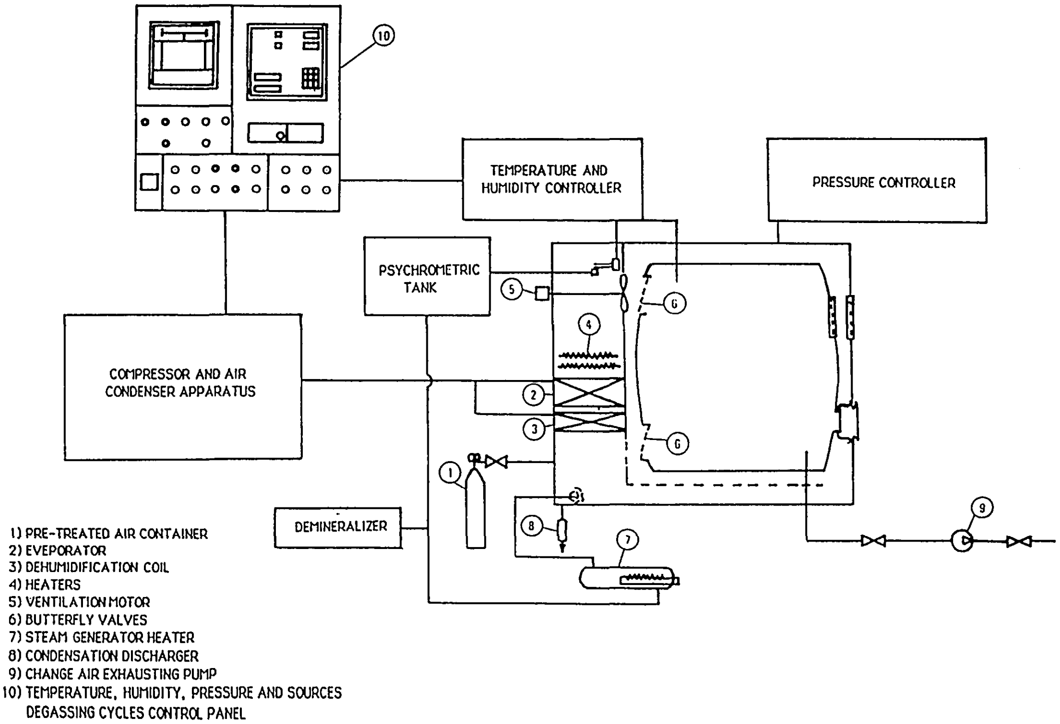

Figure 1. Diagram of the climatization apparatus of the radon test facility.

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[1] The system was developed with the technical cooperation of Angelantoni Company (Perugia, Italy).

[2] Certain commercial equipment, instruments, or materials are identified in this paper to specify adequately the experimental procedure. Such identification does not imply recommendation or endorsement by the National Institute of Standards and Technology, nor does it imply that the materials or equipment identified are necessarily the best available for the purpose.

- temperature range: from −35 to +60 °C;
- relative humidity range: from 10 to 95% in the temperature range from +5 to +60 °C with +4 °C dew point limitation;
- pressure range: from 700 to 1100 mBar.

The control of the operating conditions is fully automated. A microprocessor (programmer-controller) controls experimental cycles. A control panel is operated via a 10-key keyboard and displays set points or actual values (fig. 1).

3. Aerosol System

The aerosol system used is the Model Mage condensation nuclei generator for solid monodispersed particles.[3] This device consists of a condensation nuclei source, a thermostated bubbler containing the substance to be vaporized, and a reheater. A carnauba wax solid aerosol is used as the test [3].

4. Particle Concentration and Size Distribution Monitor

The aerosol concentration and size distribution are determined using the TSI Aerodynamic Particle Sizer type APS 33B. The instrument operates at both high and very low particle concentrations and measures the aerodynamic equivalent diameter in the 0.5 to 15-μm range. The instrument may be used for continuously monitoring the chamber atmosphere.

5. Experimental Tests at Dynamic Conditions

One of the objectives of the first experimental phase was to produce controlled atmospheres with a stable radon and daughters activity per unit volume at well defined concentration and size distribution of aerosols.

Tests were carried out under dynamic conditions using the experimental apparatus shown in figure 2. A radium solution was used as the radon source. Pylon sources were also routinely used. The radon concentration was maintained constant by continuously bubbling air through the solution during tests. A monodispersed carnauba wax aerosol was continuously injected to maintain a stable concentration of condensation nuclei. Radon and radon daughters concentrations were monitored using both commercially available monitors and monitors designed at the ENEA laboratory.

6. Test Results

Runs were carried out using the radon chamber in the open circuit dynamic operating mode under the following experimental conditions:

- flow rates ranging from 11 to 31 L/min;
- particle size distribution with a maximum frequency of about 0.6 μm.

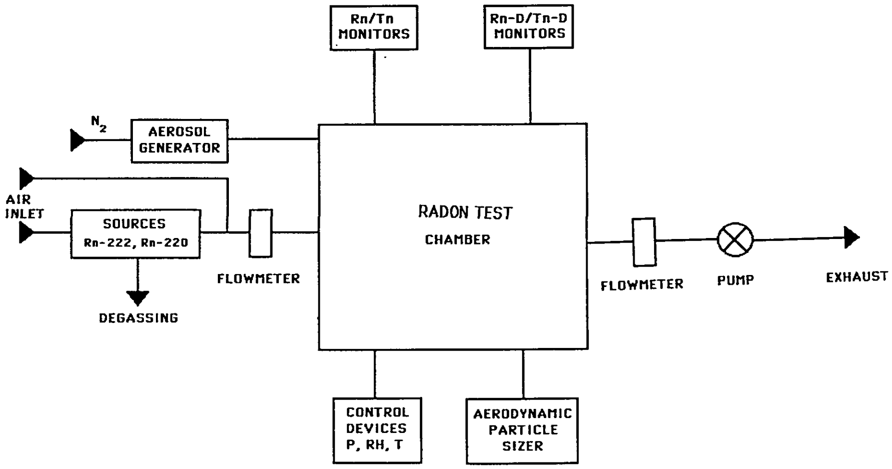

Figure 2. Diagram of the radon test facility.

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[3] Produced by the Lavoro Ambiente Company, Bologna, Italy.

Tests showed also that radon and daughters concentrations were uniform around the axis of the cylindrical volume of the chamber (useful volume). In the experimental runs, air samples were collected at a fixed point on the axis of the cylinder.

A first set of measurements was made in reference atmospheres using calibrated radon sources and monodispersed aerosols at controlled climatic conditions (temperature, relative humidity, and pressure). In the 11 to 31 L/min flow-rate range the measured radon concentrations varied from about 370 to 1110 Bq/m^3 (10 to 30 pCi/L) and potential alpha energy concentrations from about 0.83 to 4.78 μJ/m^3 (40 to 230 mWL).

The equilibrium factor (F-factor)[4] values were in the 0.5 to 0.7 range. During experimental runs, stable conditions were obtained after a delay time ranging from 2 to 6 h, at flow rates, respectively, of 31 and 11 L/min.

Further experimental tests are in progress to obtain a larger range of equilibrium factor values at well defined aerosol concentrations and particle size distributions.

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[4] The F-factor is the ratio of the equilibrium equivalent radon concentration (EEC) to the actual radon concentration (c) in the air: $F=EEC/c$.

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Calibration of Scintillation Cells for Radon-222 Measurements at the U.S. Environmental Protection Agency

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Zinc sulfide coated scintillation cells are the primary method for measuring radon-222 at the U.S. Environmental Protection Agency (EPA), Office of Radiation Programs (ORP), Eastern Environmental Radiation Facility (EERF). These cells are used to measure concentrations of radon in exposure chambers that are used to calibrate or test other devices or instruments. Individual cells are calibrated by analyzing samples of air with known concentrations of radon produced by emanation of radon from standard radium-226 solutions obtained from the National Institute of Standards and Technology. The calibration procedure includes ingrowth of radon-222 into equilibrium with the radium in the standard solution, transfer from the solution into an evacuated container, and dilution with a measured volume of air. Samples of the radon in air mixture are transferred to evacuated scintillation cells and sealed for 4 h prior to counting, which allows secular equilibrium to be established between the radon and its decay products.

Calibration factors for each individual cell are computed by decay correcting the radon to the time of collection and calculating the ratio of count rate (cpm), corrected for background, to radon activity (Bq) for the specific volume of the cell. Four or more calibration factors are determined for each cell and averaged to provide the calibration factor used for measurements. Calibrations are repeated at 6-mo intervals, and the results of each calibration are compared to the previous averages. When calibration factors fall outside the 95% confidence interval, they are rejected and the cell is checked for defects prior to recalibration.

Key words: calibration; radon measurement; scintillation cell; uncertainty.

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1. Introduction

The Environmental Protection Agency (EPA) recently has recognized radon-222 in indoor air as a significant threat to the health of the public, which has resulted in very rapid growth of the radon measurement industry in the United States. The EPA has initiated programs, such as the Radon Measurement Proficiency Program and individual testing and exposure programs, to help other federal and state agencies assure accurate measurements. The principal measuring device used by EPA to establish radon concentrations for these tests is the scintillation cell. Measurements made with commercially available scintillation cells with a volume of 0.125×10^{-3} m^3 (see fig. 1) are used as the basis for determining radon concentrations in two radon exposure chambers. These chambers are used to calibrate and test other measurement devices, so it is very important that they be accurate. Consequently, the cells must be accurately calibrated with reliable sources of radon-222.

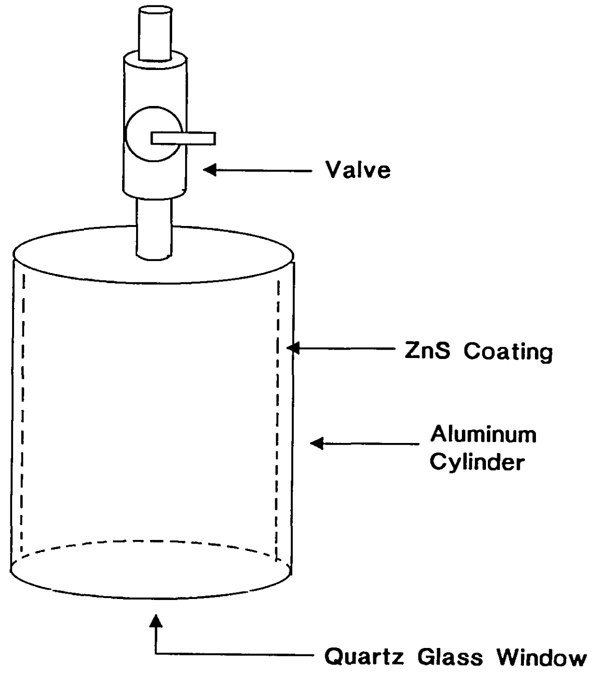

Figure 1. Scintillation cell.

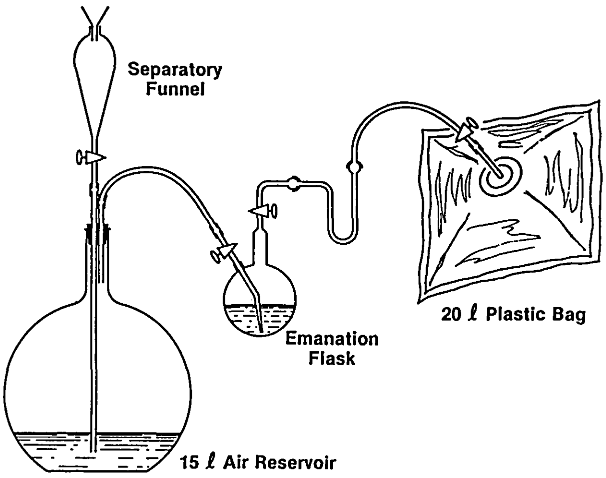

Figure 2. Radon emanation system.

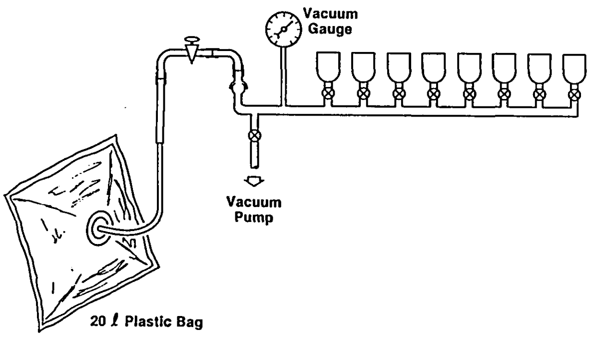

Figure 3. Radon transfer manifold.

2. Calibration

Scintillation cells are calibrated by filling evacuated cells with radon that has emanated from a radium-226 solution obtained from the National Institute of Standards and Technology. The procedure used, which has been described by Lucas [1], employs water displacement to force a measured amount of air through an emanation flask that contains the standard radium solution (see fig. 2). The 5–10 L of air required to purge the radon from the flask flows into a previously evacuated plastic bag. Although the bag has a usable volume of approximately 0.02 m^3, when the input is limited to 0.01 m^3 or less, the pressure inside the bag is negligible and no correction for pressure is required. The radon concentration is determined by dividing the activity of radon in becquerels (Bq) emanated from the radium solution by the volume of air in cubic meters (m^3) transferred to the bag.

Scintillation cells are filled with the radon and air mixture through a manifold and vacuum pump assembly as shown in figure 3. The cells and manifold are evacuated with a vacuum pump and then filled with the radon mixture from the bag. This technique allows several cells to be filled from a single emanation procedure. After filling, the cells are set aside for at least 4 h to allow ingrowth of radon decay products. The cells are analyzed for radon content by counting light pulses on a 2-in photomultiplier for 400 min.

Calibration factors are calculated for each cell using eq (1):

$$CF=\frac{(R-B)}{Rn}\cdot\frac{C}{A}, \tag{1}$$

where CF = Calibration factor (cpm/Bq),
R = Total count rate (cpm),
B = Background count rate (cpm),
Rn = Radon-222 concentration (Bq m^{-3}),
V = Volume of cell (m^3),
C = Correction for decay during counting,

$$C=\frac{\lambda t}{1-e^{-\lambda t}},$$

A = Correction for decay prior to counting,

Table 1. Example cell calibration factors

Cell No.	CF_1	CF_2	CF_3	CF_4	$\overline{CF}$	SD	SD/CF(%)
4	135	148	139	143	141	6	3.9%
9	122	138	130	120	128	8	6.4%
10	140	144	142	146	143	3	1.9%
14	133	142	130	139	136	6	4.0%
18	145	143	141	142	143	1	1.2%
25	143	145	139	148	144	4	2.6%
31	139	149	141	139	142	5	3.4%
41	131	141	134	139	136	4	3.4%
43	133	144	136	143	139	5	3.9%
44	136	144	142	145	142	4	2.8%

$A=e^{-\lambda T}$,
λ=decay constant for radon-222 (min^{-1}),
t=counting time (min), and
T=elapsed time between collection of sample and counting (min).

A typical value for calibration factors for our 0.125×10^{-3} m^3 scintillation cells is 135 cpm/Bq. Calibrations are repeated a minimum of four times and the average calibration factors are calculated. Typical data for 10 scintillation cells are shown in table 1. Calibrations are also repeated periodically and checked against previous data for indications of cell damage or aging. If the calculated calibration factor falls within the 95% confidence interval for that cell, it is added to the data base for that cell. If the factor falls outside the 95% confidence interval, the cell is inspected for damage or leakage and repaired if necessary.

3. Uncertainty of the Calibration

The relative systematic standard deviation for a single determination when using this method is estimated, by Lucas [1], to be 1.4%. The total uncertainty associated with a single radon measurement using our 0.125×10^{-3} m^3 scintillation cells calibrated with this method is described by Pohl [2] with the following relationship:

$$\%U=[(G+b)/(G-b)^2+(0.0175)^2\times(1/J)]^{1/2}\times 100 \qquad (2)$$

where $\%U$=Total uncertainty expressed as a percent,
G=Gross counts in time t,
b=Background counts in time t, and
J=Number of calibrations for the scintillation cell.

Ar 148 Bq m^{-3} (4 pCi/L) with counting times of 400 min, typical values would be

G=1200,
b=200, and
J=4.

Using eq (2),

$$\%U=[(1200+200)/(1200-200)^2+(0.0175)^2\times 1/4)]^{1/2}\times 100$$
$$\%U=3.8\%\ (1\ \text{sigma})$$

4. Conclusions

This procedure has proven to be an easy and reliable method that allows calibration of multiple scintillation cells for radon-222 measurements. Intercomparisons have shown that EERF radon-222 measurements are comparable to those obtained by other laboratories.

About the authors: E. L. Sensintaffar and S. T. Windham are U.S. Public Health Officers assigned to the U.S. Environmental Protection Agency, Office of Radiation Programs, Eastern Environmental Radiation Facility, located in Montgomery, AL.

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ICARE Radon Calibration Device

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An aerodynamic calibration device called ICARE is briefly described. ICARE is currently being used at the Nuclear Research Center of Saclay in France for the certification of instruments employed in measurement of artificial radioactive particulate airborne contamination. ICARE is essentially a blower. Aerosols calibrated in size and labelled with ^{137}Cs or ^{239}Pu are injected in the mainstream of the blower upstream of the test section.

To extend ICARE's field of application to the case of instruments employed in measurement of ^{222}Rn and of its decay products, a new line of injection has been designed and is under construction, in the frame of a contract financed by the European Community Commission. The main effort is presently oriented toward the development of a reliable ^{222}Rn source, based on a solid deposit of ^{226}Ra on organic fibers, and of a reference method for the measurement of ^{222}Rn activity per unit volume of air. This reference is based on a modified version of standard containers being used in the French reference method for calibrated measurements of ^{133}Xe and ^{85}Kr by gamma spectroscopy.

Key words: calibration; decay products; metrology; ^{222}Rn; standard.

Accepted: November 6, 1989

1. Introduction

In France, the Technical Centre for Ratification of Radiation Protection Instruments is responsible for approval of all instruments used by radiation protection officers for surveillance of ambient radiation or for dosimetry of ionizing radiation.

In the case of equipment designed for measurement of gaseous or particulate contamination of the atmosphere, special test methods are needed for aerodynamic certification and calibration.

An aerodynamic calibration device using standard radioactive aerosols called ICARE (Installation de Calibration à l'Aide d'Aérosols Radioactifs Etalons) has been designed for this purpose by J. Charuau and M. Ammerich [1]. It is currently being used for certification of instruments employed in measurement of artificial, radioactive, particulate contamination (alpha and beta emitters).

This paper describes the new procedures that have recently been used to adapt ICARE to ^{222}Rn and decay products measurements.

2. Description of ICARE

The essential characteristic of this device is that it operates under dynamic conditions. The device is a blower (figs. 1, 2, and 3) linked to the peripheral air-conditioning units. ICARE offers test conditions that are perfectly designed, both in terms of aerodynamics and of aerosol physics for the char-

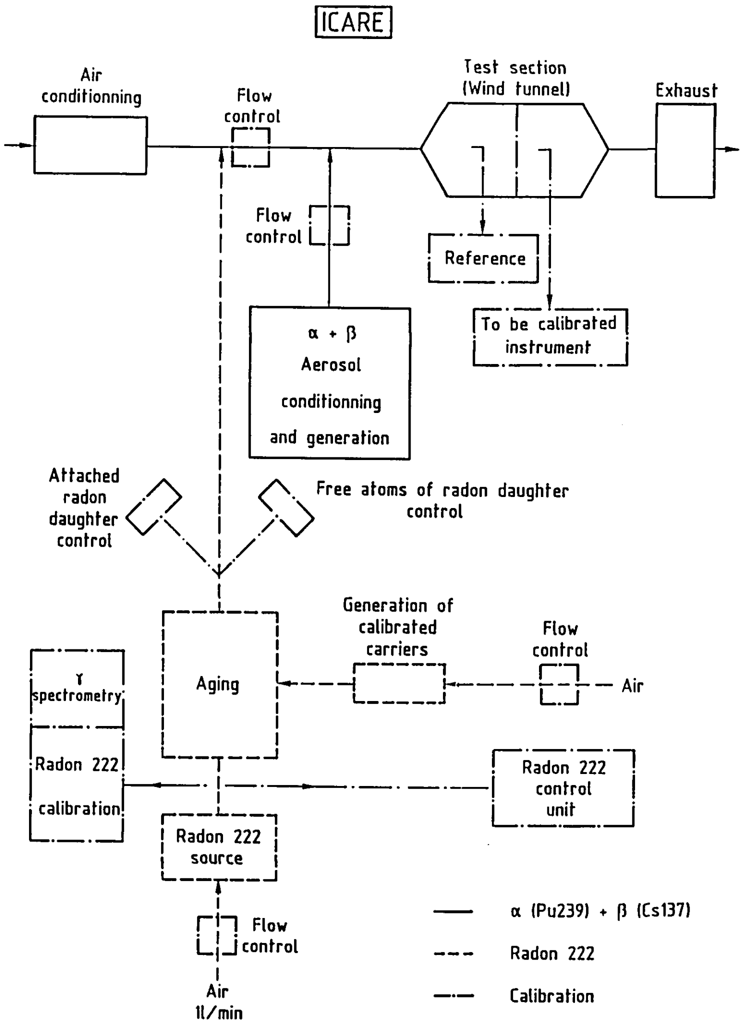


Figure 1. Description of ICARE.

acterization of radioactive aerosols monitoring systems. Calibrated aerosols of CsCl with size distribution centered on 0.4 and 4 μm (fig. 4), and radioactively labelled with ^{137}Cs and ^{239}Pu are generated and injected in the mainstream of the blower. The dilution factor of the injection line flow by the mainstream is kept constant. The blower concept ensures that exactly the same atmosphere is delivered by the sampling line used for the apparatus and by the sampling line used for the apparatus to be certificated.

In the blower concept, the final precision of the calibration provided depends mainly on the precision of the various instruments used to measure air mass flow (fig. 1) linked to the sampling and injection lines and to the mainstream, and also on the measurement of artificial radioactivity by alpha and beta reference spectrometry.

ICARE was initially equipped with an injection line for ^{222}Rn and its decay products, which may or may not attach to natural atmospheric condensation nuclei or to test aerosols. The aim was only to verify that, with low-level airborne ^{239}Pu contamination, the measurement instruments were able to discriminate between alpha emissions from ^{239}Pu and those of short-lived polonium isotopes naturally present in the laboratories. As the aim was not to calibrate instruments for the measurement of ^{222}Rn and its decay products, there was no need for stable radon generation. In fact, the stability of ^{222}Rn emission from the source (uranium ore) was large and inadequate for calibrating instruments for the measurement of ^{22}Rn. In addition there was no control over the concentration and particle size of the aerosols bearing radon decay products, and hence no possibility of free choice of the equi-

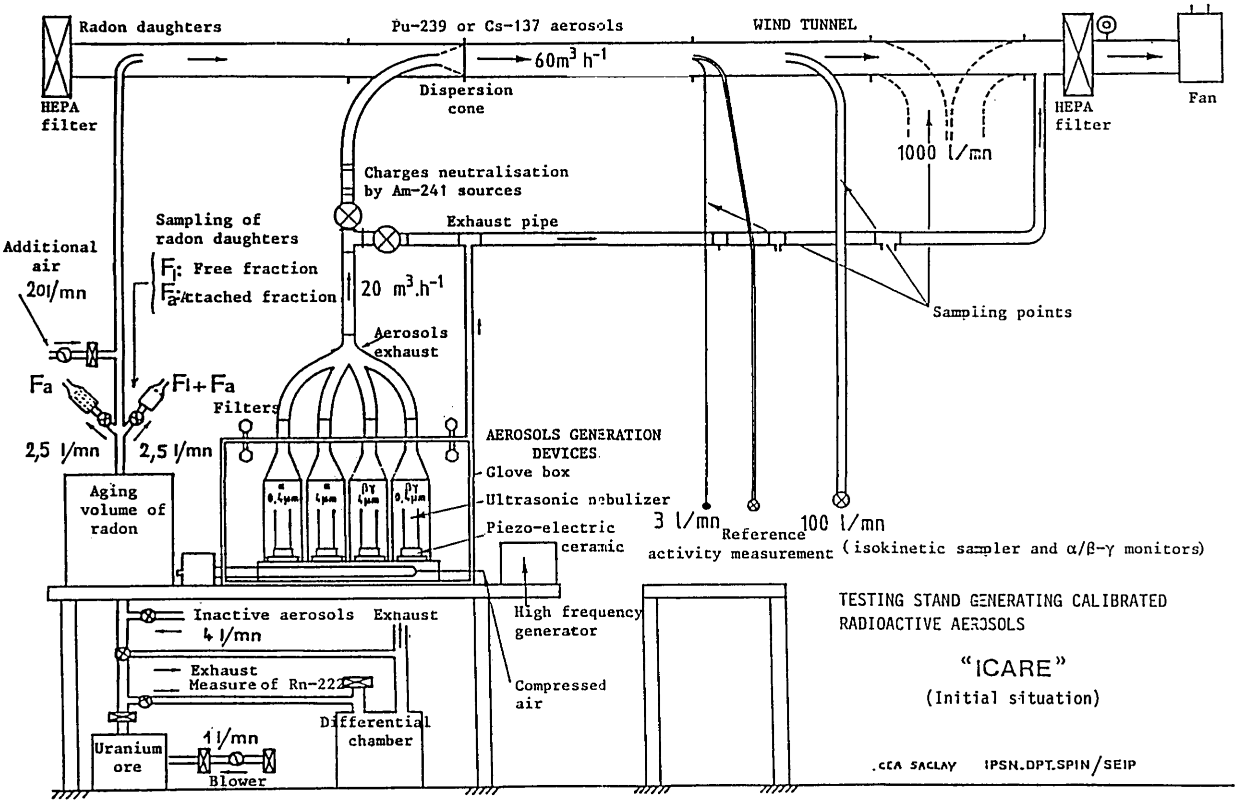


Figure 2. Schematic diagram of ICARE.

librium factor and of the unattached fraction of the decay products.

This is why, in a separate contract financed by the European Community Commission, a new line for injection of radon and its decay products has been designed and is being developed (ICARE Radon). The aim of the first phase (1988–1989) is to produce an atmosphere with a perfectly defined and stable radon 222 activity per unit volume. The second phase (1989–1990) will be devoted to development of an atmosphere in which the activities per unit volume of short-lived radon decay products and the particle size distribution will also be perfectly defined and stable. A description will only be given of the first phase, which is the subject of the thesis by M. Guelin [2].

3. The Radon Source

To obtain a stable, reliable radon source we prefer to package the radium in the form of a solid deposit fixed to an appropriate support, rather than solubilize ions in a liquid. Sources in which ^{226}Ra is adsorbed by ion-exchange resins have been described and are commercially available. Following a laboratory study of the binding of soluble radium to organic fibers impregnated with manganese oxides, we found that these fibers constitute the ideal support for a solid ^{222}Rn source. Indeed, we found that the emanation factor of radon emitted by such a source was about 100%, and was stable for scanning air humidity ratios greater than 80% fibers (fig. 5) [4]. The pressure loss in the fibers is negligible.

A diagram of the ^{222}Rn source is shown in figure 6. This device comprises an air-supply circuit with an on-line humidifier, flow rate controls on all air inlets, and an ionization chamber used to control the stability of generation and scanning. The radon injection line ends at the main pipeline of the blower upline from the test unit and from the unit for injection of artificial aerosols. Stirring devices ensure good mixing of the air from the radon injection line and the air from the blower.

The activity of the solid source of ^{226}Ra is measured by gamma spectrometry, after establishment of equilibrium between the ^{226}Ra and ^{214}Bi. Emission of ^{222}Rn by the source expressed in atom·s^{-1} is equal to the activity of the ^{226}Ra of the source expressed in Bq. The activity per unit volume of radon in the air delivered is equal to the product of the decay constant of ^{222}Rn and the source intensity

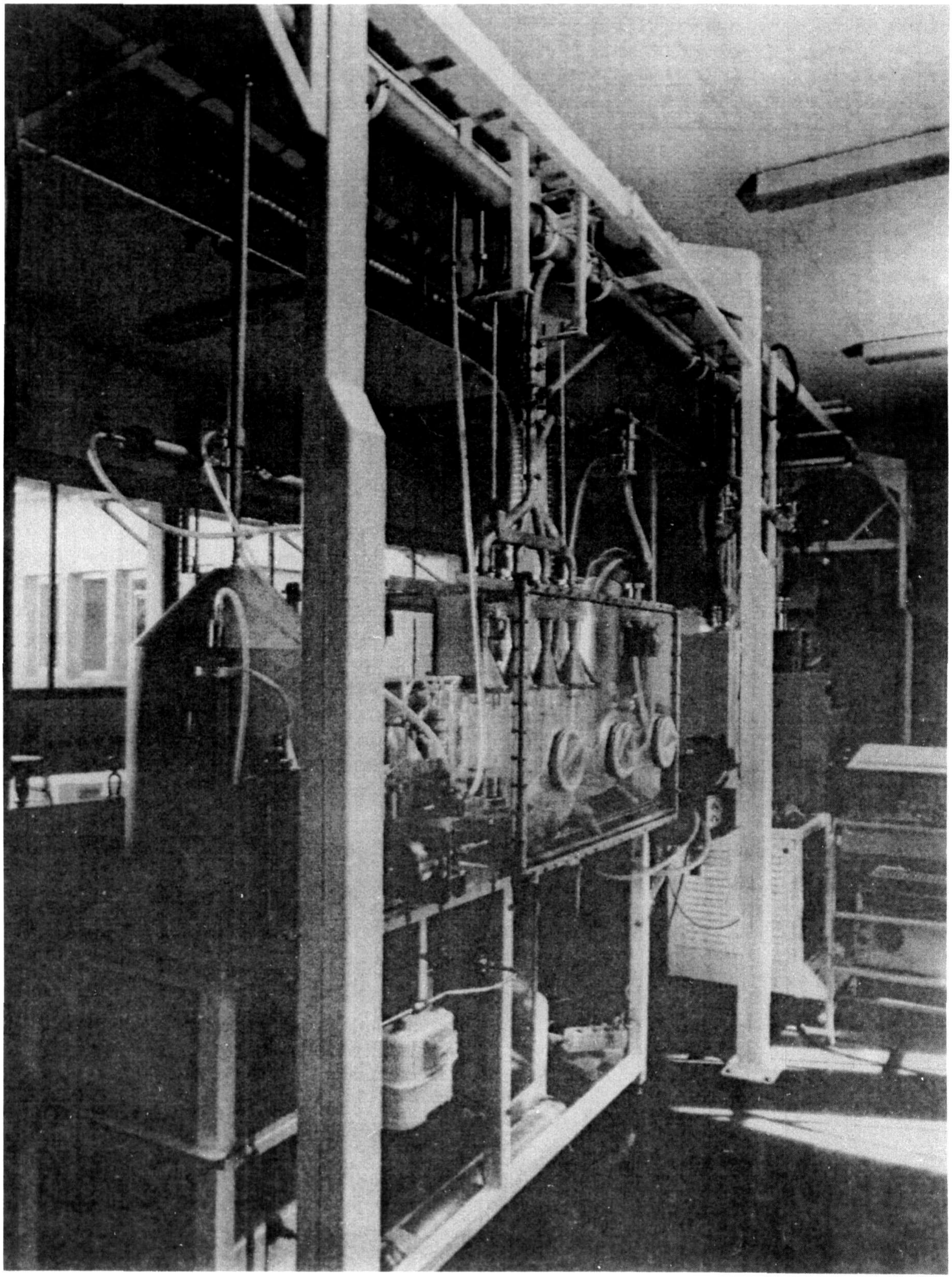
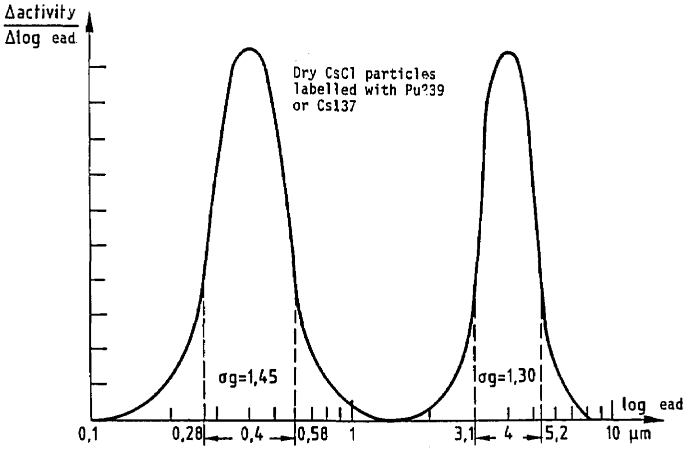
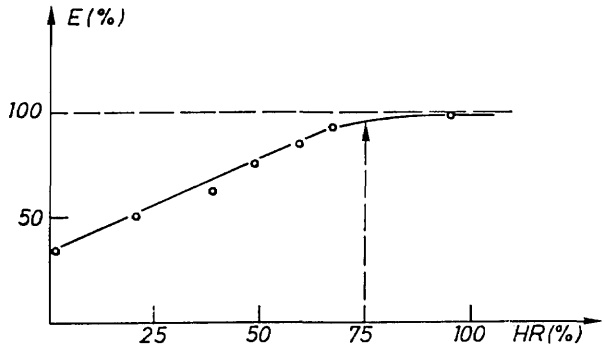


Figure 3. General view of device.



ead : equivalent aerodynamic diameter

Figure 4. Particle size distribution of test aerosols.



E : Emanation factor of the source
HR : Humidity ratio of the scanning air

Figure 5. Emanation factor of the source as a function of the scanning air humidity ratio.

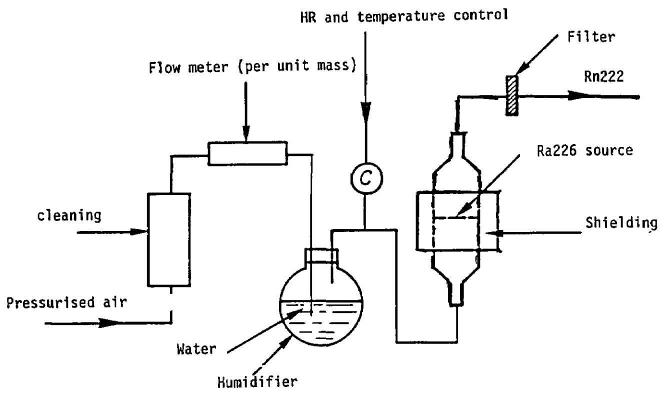


Figure 6. Diagram showing radon source.

expressed in atom·s^{-1}, divided by the scanning air flow rate, which is measured with a reference flow meter. Hence:

$$A^{v}_{Rn}=\frac{\lambda 222\ A_{ra}}{Q_{v}} \tag{1}$$

where:

A^{v}_{Rn} = activity of ^{222}Rn per unit volume (Bq·m^{-3}) in the scanning air
$\lambda 222 = 2.1\times 10^{-6}$s^{-1}
A_{ra} = activity of the radium source (Bq)
Q_{v} = flow rate of scanning air (m^3·s^{-1}).

4. Development of a Reference Method for ^{222}Rn Activity per Unit Volume Measurement

It is possible in theory to apply eq (1) in order to define A^{v}_{Rn} in the injection line. It has seemed to us unwise to rely uniquely on eq (1) for definition of ^{222}Rn activity per unit volume in the injection line, since this would involve the assumption that the radon emanation factor is strictly equal to 100%. This is why we have sought to develop a reference method for measurement of the airborne activity per unit volume, which may also by used in other situations. We took as our starting point the fact that there already exist standard containers for the measurement by gamma spectroscopy of the activity of ^{133}Xe and ^{85}Kr per unit volume of atmosphere (fig. 7).

Direct use of this type of container involves the difficulty that gamma spectrometry measurement of ^{222}Rn can only be achieved by examining gamma rays from one of the short-lived decay products, ^{214}Bi, considered to be in equilibrium with ^{222}Rn after 3 h. The difficulty is that since a non-gaseous decay product is involved at room temperature, the atoms formed by decay bind to the walls of the container and hence distribution of activity in the container volume is no longer homogeneous; thus, it is not easy to assess. To remedy this situation, it was decided to fill the inside of the container with a metal mesh structure of low gamma radiation absorption. This structure comprises light alloy meshes which allow for deposition of ^{214}Bi within the container space to be uniform at least on a scale of the mesh size of the metal structure, i.e., a few millimeters (figs. 8 and 9). This new device has been patented [3].

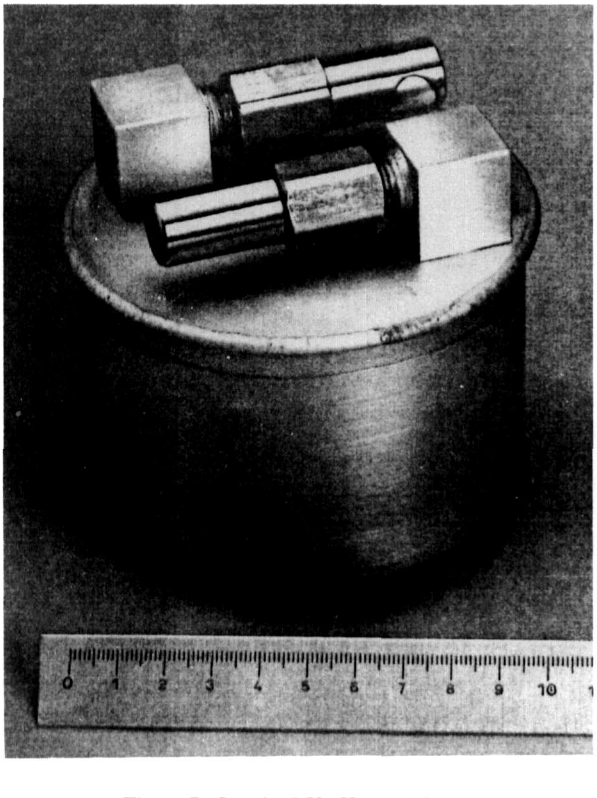


Figure 7. Standard Xe Kr containers.

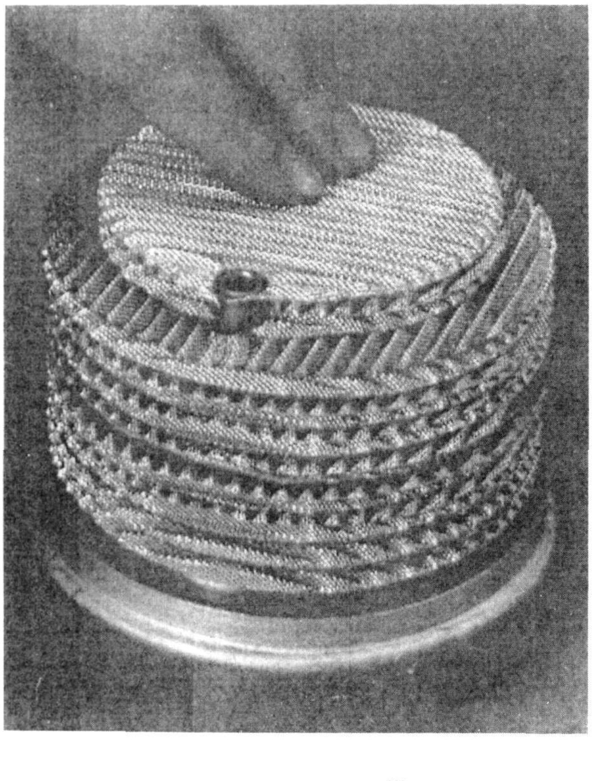


Figure 8. Internal structure of the ^{222}Rn container.

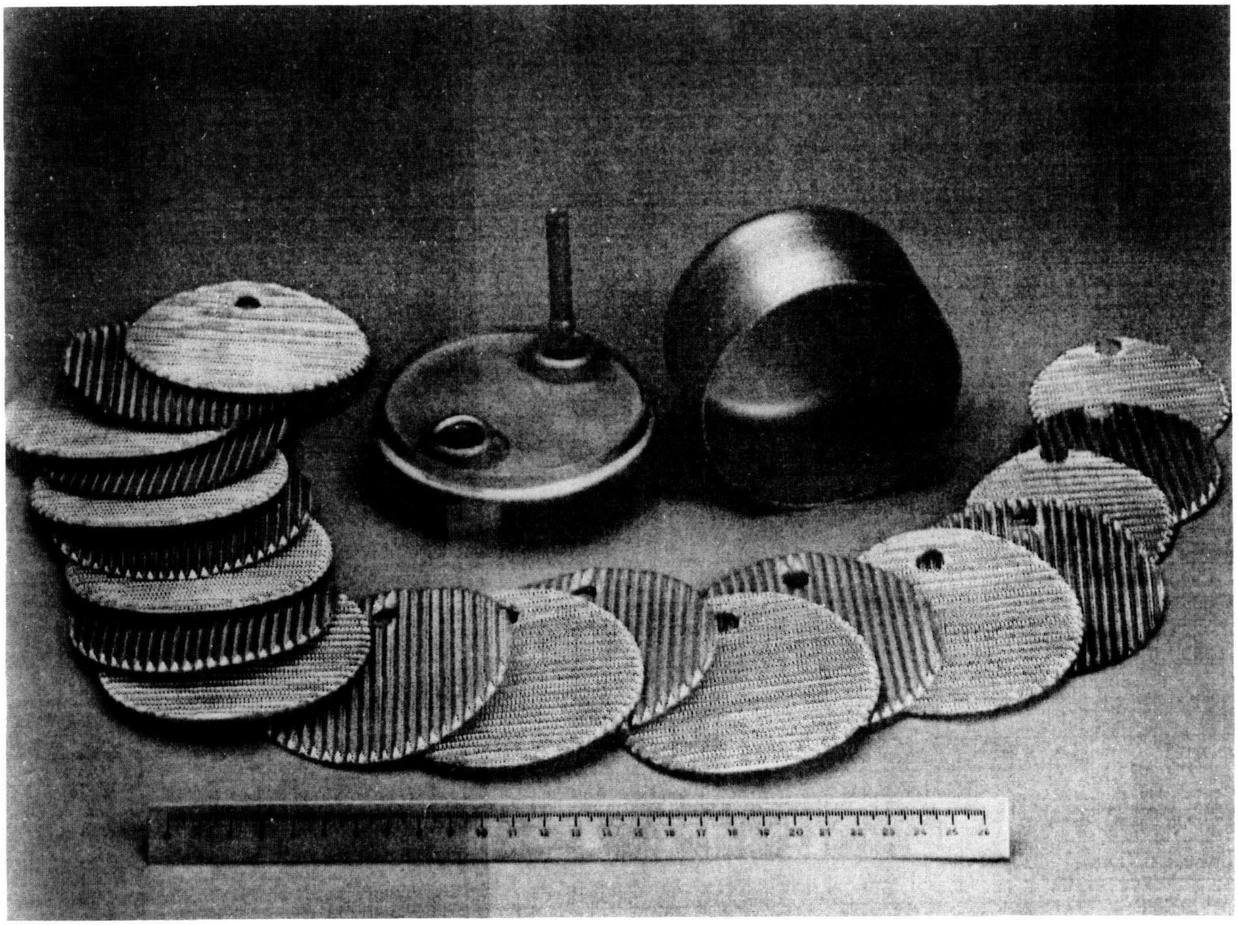


Figure 9. Display of the metal mesh structure.

The container intended for regular measurement of the airborne activity per unit volume in the radon injection line, the stability of which is guaranteed by continuous monitoring of the injection air flow rate and by the response of the ionization chamber serving as source monitor, will have a lower limit sensitivity of about 2,000 Bq·m^{-3} under laboratory conditions at the Nuclear Research Center of Saclay. With the gamma spectrometer used, the sampled air volume is about 500 cm^3.

The activity per unit volume in the test unit is obtained from that of the injection line by dividing the factor defining the dilution produced by the mainstream of the blower. The accuracy of this dilution factor is that obtained with reference flow meters. The homogeneity of the mixture is checked beforehand by injecting helium as a tracer in the radon injection line and examining the spatial distribution of the helium concentration in the test section with a mass spectrometer. In the reference sampling line, an ionization chamber, which constitutes a secondary standard, is used to verify the values provided by the test unit and their stability. Its lower sensitivity limit is a few Bq·m^{-3} for ^{222}Rn.

About the author: *P. Zettwoog has been measuring and studying radon for 20 years in underground mines, in the soil, and in the environment.*

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The NIST Primary Radon-222 Measurement System

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Within the United States, the national standard for radon measurements is embodied in a primary radon measurement system that has been maintained for over 50 years to accurately measure radon (^{222}Rn) against international and national radium (^{226}Ra) standards. In turn, all of the radon measurements made at the National Institute of Standards and Technology (NIST) and the radon transfer calibration standards and calibration services provided by NIST are directly related to this national radon standard. This primary radon measurement system consists of pulse ionization chambers and ancillary gas handling and gas purification equipment. The system is currently undergoing a significant upgrading and expansion which will replace the extant outdated system.

Key words: calibration; ionization chambers; measurement; radium-226; radon-222; standards.

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1. Introduction

Unlike the calibration for many other radionuclides, there is no currently available or demonstrated procedure for fundamentally or directly calibrating radon (^{222}Rn) without recourse to an indirect or comparative measurement. As a result, at the present time, all measurements of radon are ultimately related back to some radium (^{226}Ra) standard. Within the United States, the National Institute of Standards and Technology (NIST), formerly the National Bureau of Standards (NBS), maintains both national and international radium standards that can be directly related to the international primary mass standards of radium prepared by Marie Curie in 1911 and Otto Honigschmid in 1934 [1].

A primary measurement and calibration system for ^{226}Ra and ^{222}Rn has been maintained by NIST/NBS since the early 1940s [2]. This system was and still is one of the principal tools used to compare and trace radium standards to each other, as well as to relate radon measurements to radium standards.

The system consists of pulse ionization chambers which are used in conjunction with radium solution standards and ancillary gas handling and gas purification equipment. Use of the system relies upon a methodology that was originally developed for the purpose of assaying and standardizing radium solutions, such as comparing various preparations with each other or with national or international radium standards [2,3,4].

In this method, often referred to as the "radon method of analysis of radium," the radon generated from the decay of radium in a sample or standard is physically separated from the radium, and quantitatively transferred to the ionization chambers. Alpha particles resulting from the decay of the radon and the radon decay products are then detected and counted in the chambers. In addition to using the method for comparing laboratory-prepared solutions and standards of radium, laboratories here employed it for radium assay of a variety of samples (e.g., water and wastewater samples and various solutions of solid samples such as soils or uranium ores) [2,3,4]. Similarly, the method also has been used to measure the radon content of whole air samples. Such measurements were extensively performed by this laboratory in the late 1940s to assay mine atmospheres and breath samples from uranium-mine workers and radium-dial painters [2].

Inasmuch as this measurement system and methodology provides a quantitative link between a radon measurement and the radium content of a sample or standard, it also has served and continues to serve as a national calibration standard for radon measurements. In fact, this national standard, embodied in the primary measurement system, has been maintained for over the past 50 years for this purpose of accurately measuring radon against international and national radium standards. In turn, all radon measurements made by NIST and the radon transfer calibration standards and calibration services provided by NIST are directly relatable to this national radon standard [5,6].

2. Requirements for a Primary Calibration Standard

Before directly addressing the topic of the NIST primary radon measurement system and its application to other NIST radon transfer standards and calibration services, it may be useful and insightful to summarize what we believe are the necessary requirements for a primary calibration standard. The requirements are that the standard should: (i) have long-term stability; (ii) be sufficiently accurate to meet the calibration and subsequent measurement needs; and (iii) be efficacious for cross calibration purposes, allowing the transfer of calibrations to other measuring instruments and methods.

One of the great advantages of a calibration system based on ionization chambers over other measurement instruments (e.g., scintillation cells) is its ability to maintain a constant and stable calibration over very long time periods. Calibration results from this laboratory, as well as from the U.S. Department of Energy Environmental Measurements Laboratory [7], have demonstrated that radon calibration factors for ionization chambers have remained constant within a few percent for decades. This level of stability provides a useful independent check on the joint consistency of both the use of a radium standard and the instrument performance. With other instruments such as scintillation cells which are easily subject to physical degradation and subsequent changes in detection efficiency, one must assume, without verification, that the radium standard has remained constant and that the use of the standard has not changed.

In regard to the second requirement, overall calibration accuracies in the range of 1 to 2 percent are possible with ionization-chamber-based calibration systems. This appears to be more than adequate for presently envisaged radon measurement needs (see the workshop discussion in the Preface to this issue).

The third requirement is that the primary calibration standard must be capable of handling samples in a way that allows for a sufficiently accurate cross calibration to secondary calibration systems. The ancillary gas-handling and gas-purification manifold incorporated into the NIST primary system is very efficacious for transferring identical radon-in-air samples measured with one system to the ionization chambers for a direct calibration. The manifold also can be used to prepare replicate calibrated samples from radium sources.

3. The Pulse-Ionization-Chamber-Based Primary System

The NIST primary radon measurement system comprises three major components: (i) a manifold containing a radium standard source and a sampling location for processing gas sample bulbs; (ii) gas purification equipment; and (iii) the ionization chamber and appropriate electronics. A simplified schematic illustrating these components and the basis of their operation is shown in figure 1.

The system employs an internal gas counting technique in which a radon sample is introduced, together with a suitable filling gas, directly into a chamber. The chambers consist of a nominal 4-L volume cylindrical cathode with a central wire anode. Their operating voltage is maintained in the ionization region (typically 1200 V); and they are operated in a pulse counting mode (see fig. 2). The advantages of counting current pulses over a continuous direct current mode of operation has been described previously [3].

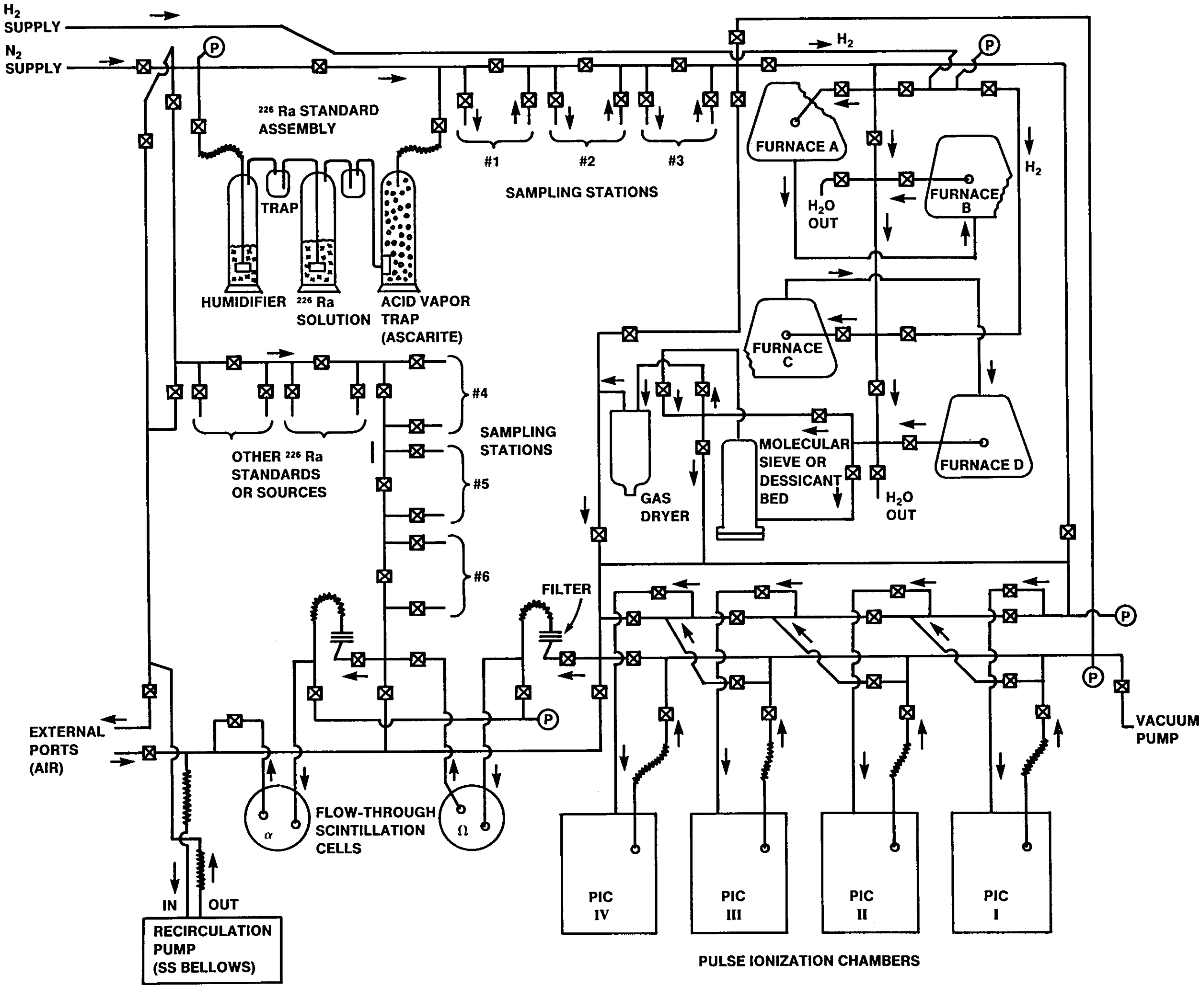


Figure 1. A simplified schematic of the NIST primary radon measurement system illustrating its major components and principle of operation.

The principles of operation are straightforward. Alpha particles resulting from the decay of radon and its decay progeny ionize the gas contained in the chamber. Electrons and positive ions liberated in the ionized gas are collected by means of the electric field maintained by the high voltage between the central electrode and chamber wall. The collection of the electrons on the central electrode produces a pulse of current to flow in an output resistor R. Since the voltage drop across R is proportional to the current flowing, the voltage resulting from the passage of a single alpha particle will increase and then decrease again as the current pulse fades away. This voltage pulse is amplified and then recorded by an electronic counting circuit.

Pure nitrogen is used as the filling gas since it has a very low electron affinity which allows for a sufficiently sharp and distinct pulse with a fast collection time of the electrons on the electrodes. Resolving times of less than 100 μs are readily achievable [3]. The performance of the chambers is very adversely affected by the presence of ions and condensable vapors in the gas sample, as well as by the presence of electronegative gases (such as oxygen). Therefore, an ancillary gas-handling and gas-purification capability is necessary.

The gas-handling manifold is used for processing samples i.e., separating and purifying the radon from whole air samples, mixing it with the filling gas, and transferring the gas into counting chambers that have been previously evacuated. This type of radon gas-handling system, commonly associated with methods for assaying radium samples, has been developed for many applications by Evans [8], by Curtiss and Davis [3], and by

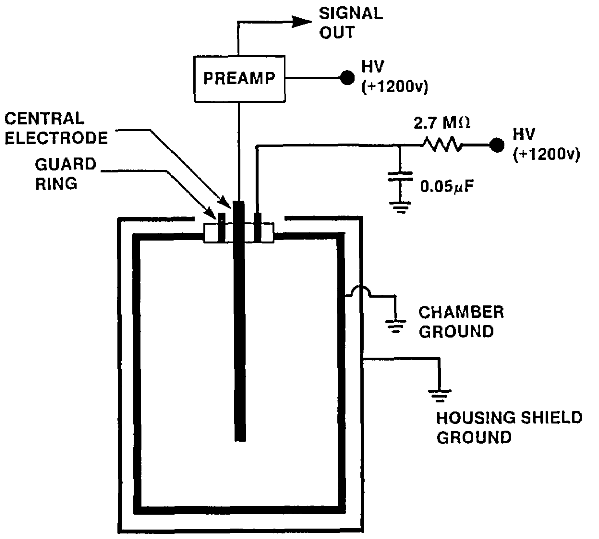


Figure 2. A block diagram of a pulse ionization chamber showing its electrical connections for operating in a pulse counting mode.

Harding, et al. [4]. Similar systems also have been described by Blanchard [9], by Harley [10], and by Kreiger and Jacobs [11].

With the system illustrated in figure 1, radon samples (either connected at the sampling location or obtained by generation from the radium standard) are transferred with a slow stream of nitrogen carrier gas through the purification equipment that removes water vapor and oxygen from the gas stream. Oxidative purification is achieved with a quartz combustion tube that is filled with tightly wound copper gauze and surrounded by a furnace operated at 500 °C. The heated copper in the combustion tubes is previously reduced by a stream of hydrogen before it is used to purify the samples. Samples transferred to the ionization chamber are counted after the radon and its short-lived decay products reach secular equilibrium.

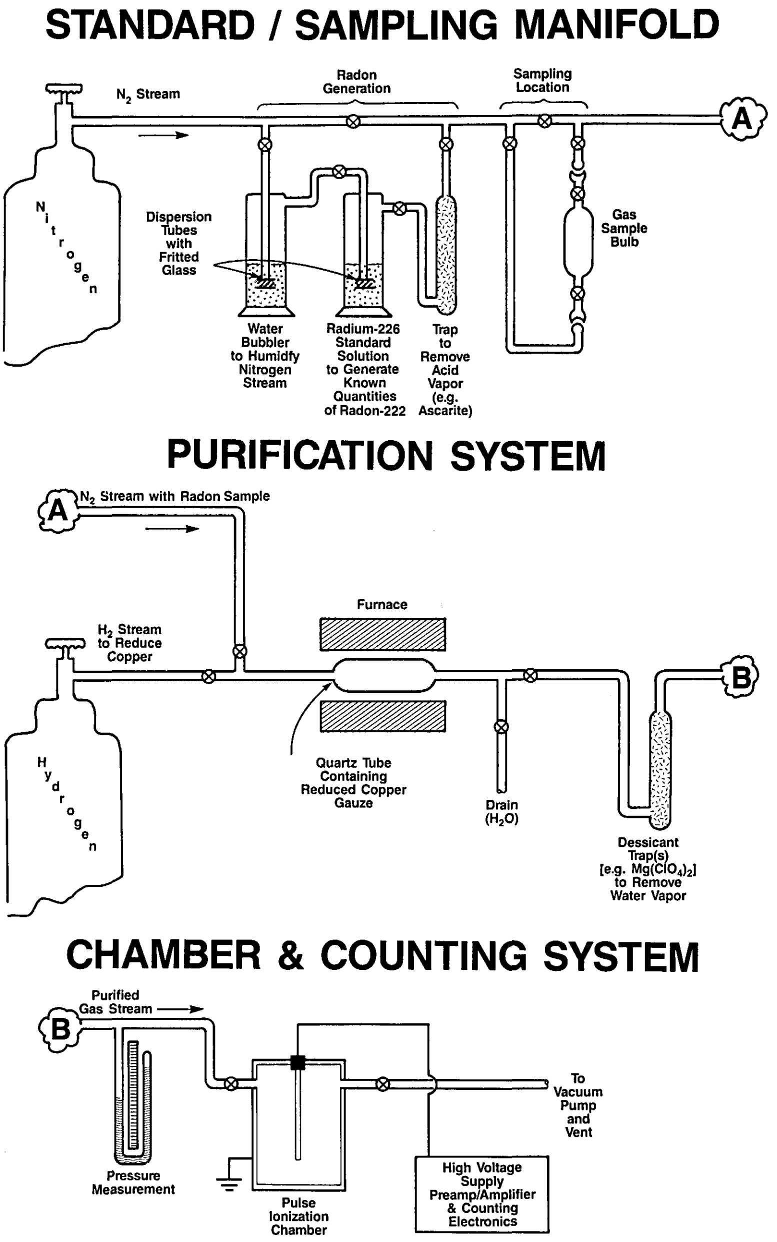
The ionization chambers are calibrated against ^{226}Ra standards (sec. 5.1) by quantitatively transferring known accumulated amounts of radon into the chambers. This transfer is performed by slowly bubbling the nitrogen carrier gas stream through a standardized radium solution which is contained in a modified gas-washing bottle. As in the procedure used for measuring radon samples, the gas stream passes through the purification system and fills the pre-evacuated chambers. Great care must be taken to insure that the transfer of radon from the radium solution to the chamber is 100% efficient, and that it replicates as closely as possible the conditions of the gas transfer for a radon sample [12].

The original NIST/NBS system, built in the 1940s, consisted of a bank of over 20 chambers. It was used for many years to perform routine measurements of air and breath samples—up to over 1200 samples per year throughout the late 1940s and 1950s. Although the system underwent many modifications over the intervening years (particularly changes in the gas-handling manifold, modernization in the electronics and signal processing, and taking some of the chambers out of service), some of the original chambers always have remained in continual use. These original chambers were constructed of silver-soldered brass with rather obsolete vacuum seals and electrical insulators, and were wax sealed to an all-glass gas-handling manifold. In recent years, it became increasingly apparent that this ancient system was rapidly becoming inoperable. Of the four remaining chambers, only two are usable, and both of these have leaks that are increasing in seriousness. The chamber backgrounds, because of the 40-year accumulation of deposited ^{210}Pb, preclude measurements at typical environmental concentration levels. As a result of the system's deteriorating condition and the reemphasized importance of this national standard, the design and construction of a replacement system was initiated in 1988.

4. The New Primary System

The new primary system is a modernized and expanded version that is based on the design of the original system. At the time this new system was being designed, consideration was given to possibly selecting an alternative measurement method for use as the primary standard. Despite the passage of 50 years, no other available method or instrumentation could be found that was more advantageous or superior to the original pulse ionization chamber.

A schematic representation of the new system is shown in figure 3. Despite the seeming complexity of the extensive gas handling and purification manifold, its principle of operation is virtually the same as the simple system illustrated in figure 1. The new manifold incorporates a similar radium standard assembly, various sampling stations, a gas purification network based on similar heated copper combustion tubes, and the necessary traps, desiccant beds, pressure and flow rate measurement devices, gas supplies, and vacuum lines. As part of


STANDARD / SAMPLING MANIFOLD

PURIFICATION SYSTEM

CHAMBER & COUNTING SYSTEM

Figure 3. Schematic of the new NIST primary radon measurement system showing the complete gas-handling and gas-purification manifold used in conjunction with the pulse ionization chambers.

its increased versatility, it also incorporates a recirculation pump for possible experiments requiring recirculated flow, as well as two flow-through ZnS(Ag) scintillation cells which can be used for less exacting radon assay measurements. All of the piping and valves for the manifold are of stainless steel high-vacuum construction with the exception of the modified gas-washing bottles for the radium standard and the quartz combustion tubes which are connected via glass-to-metal transition tubes.

For the system, four new custom-made ionization chambers were fabricated by a commercial vendor (Reuter-Stokes[1]) based on the original

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[1] Certain commercial equipment, instruments, or materials are identified in this paper to specify adequately the experimental procedure. Such identification does not imply recommendation or endorsement by the National Institute of Standards and Technology, nor does it imply that the materials or equipment identified are necessarily the best available for the purpose.

chamber design [3,4] and according to specifications provided by our laboratory. It was intended that these replacement chambers be of the same volume to obtain approximately the same detection efficiency and that they perform as nearly identically as possible as the originals, and that intercomparisons between the chambers be performed before the old ones are taken out of service. The new chambers, illustrated in figure 4, are fabricated out of stainless steel with completely sealed welded seams, unlike the originals which had O-ring gasket seals that were susceptible to leaks over time. All of the interior surfaces are electro-polished to minimize electrostatic field irregularities and to minimize surface deposition of the radon decay products. Electrical insulation of the central electrode and guard ring is made with the vendor's proprietary triaxial ceramic-to-metal seal that was welded as a complete assembly to the chamber top. The volume of one of the original chambers was measured by filling with water and was found to be 4.248 L. Based on dimensional tolerance limits, the new chambers have a minimum volume of 4.234 L and a maximum volume of 4.261 L.

Electrical coupling of the chambers for power and signal processing is as shown in figure 2. The resolving time of the output signal pulses is approximately 10 μs. These pulses are amplified and pulse height analyzed in the system's own dedicated computer.

Typical spectra obtained with the new chambers are shown in figure 5. They depict the alpha-particle spectrum shapes as they develop in time after the introduction of radon into the chamber, followed by the almost immediate removal of about 99% of the introduced radon. Figure 5a shows the

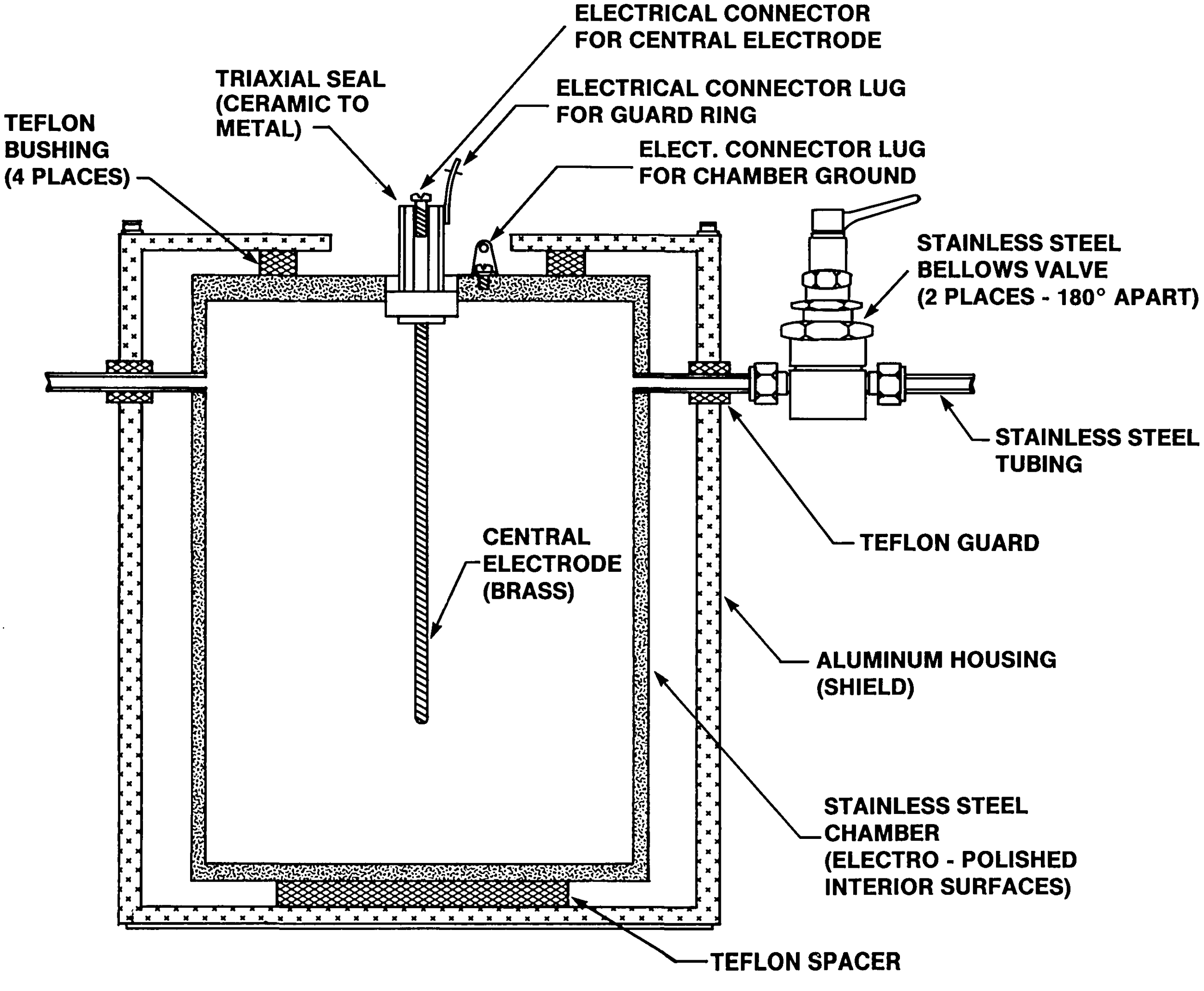

Figure 4. Cross section of the new NIST pulse ionization chamber.

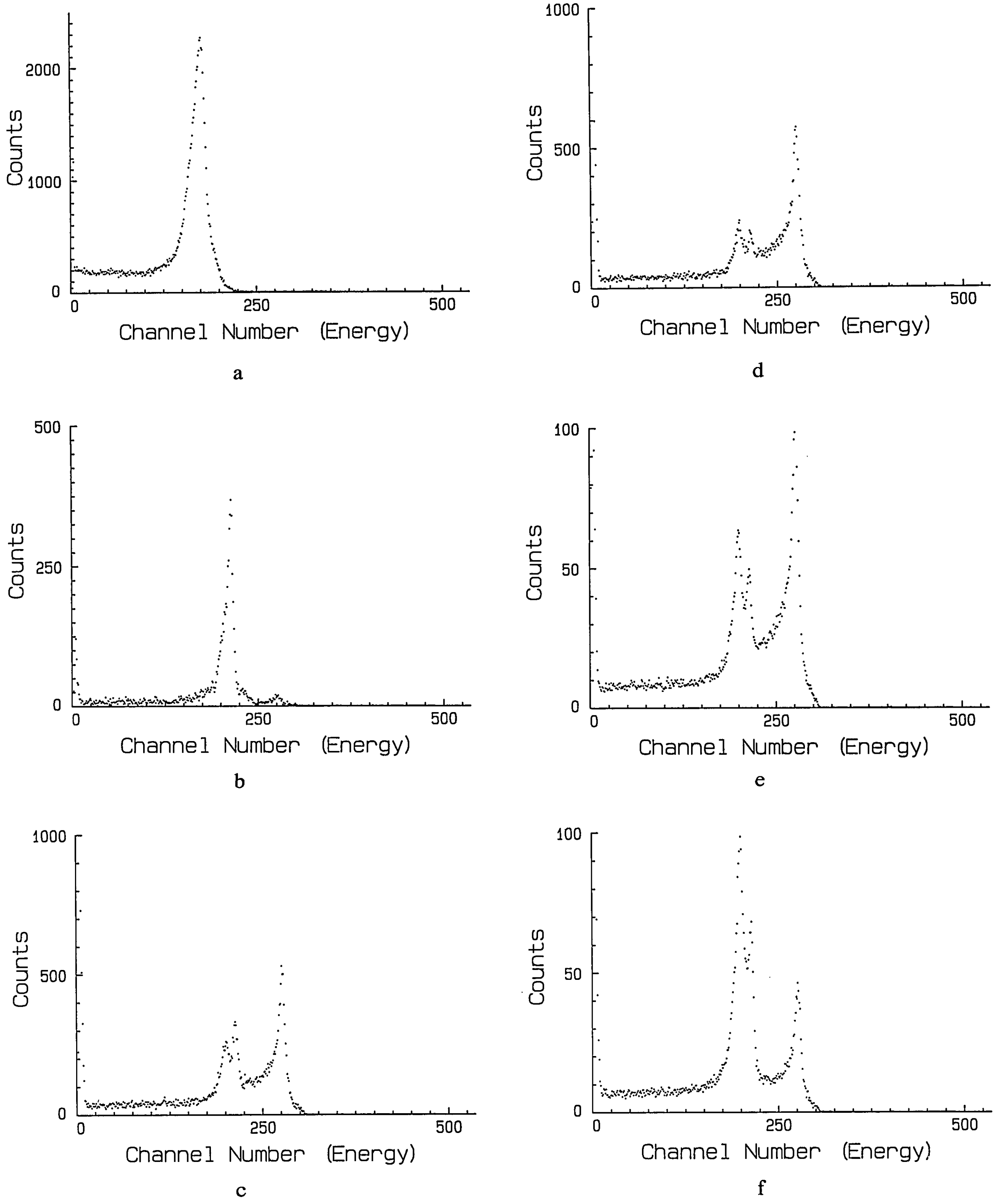

Figure 5. Typical spectra of ^{222}Rn and its decay products obtained with a pulse ionization chamber. (a) 100-s count immediately after filling; (b) 100-s count immediately after flushing; (c) 1000-s count 10 min after flushing; (d) 1000-s count 35 min after flushing; (e) 3000-s count 60 min after flushing; (f) 5000-s count 150 min after flushing. See text for details.

5.49 MeV peak of the monoenergetic spectrum of almost pure ^{222}Rn before the 3-min ^{218}Po and subsequent progeny have had time to grow to significant proportions. Shortly thereafter, a significant amount of ^{218}Po "grows in" from decay of the radon so that when most of the radon is flushed out of the chamber, the 6.00-MeV alpha particle peak of ^{218}Po dominates the spectrum, as shown in figure 5b. A small ingrowth of ^{214}Po is also evident as seen by the small peak at 7.69 MeV. The spectrum in figure 5c, taken 10 min after flushing, shows that the ^{218}Po (6.00 MeV peak) has decayed to a level comparable to the residual radon (5.49 MeV peak). At the same time, the 7.69 Mev peak from ^{214}Po has begun to more rapidly "grow in" at a rate genetically determined by the half-lives of the earlier members of the radon decay chain (26.8-m ^{214}Pb and 19.9-m ^{214}Bi). Figure 5d, obtained 35 minutes later, shows the ^{218}Po in secular equilibrium with the radon and the continued growth of the ^{214}Po. Although they are in equilibrium, note that the ^{218}Po count rate is lower than that for ^{222}Rn. Presumably this is because the detection efficiency for the gaseous radon is near 100 percent whereas the polonium, which attaches to the surfaces within the chamber, has a significant fraction of the emitted alpha particles absorbed by these surfaces without detection. Lastly, figures 5e and 5f show the gradual establishment of secular equilibrium between all members of the decay chain beginning with ^{222}Rn when the ^{222}Rn is not removed.

Calibrations with radium standards for the new chambers are not complete, but preliminary results indicate that the overall detection efficiency for radon in secular equilibrium with its decay products is very near the expected value of 2.00 counts per s per Bq of ^{222}Rn. The four remaining old chambers had efficiencies which ranged from 1.96 to 2.01 cps Bq^{-1}. The value of approximately 2 arises from the ^{222}Rn decay being detected with an efficiency of approximately 100% while the ^{218}Po and ^{214}Po are detected with an efficiency of about 50%. Losses in efficiency and spectrum peak shifts are very noticeable with incomplete oxygen removal or the presence of other impurities in the chambers. Typical calibration reproducibilities in terms of a standard error of the mean are a few tenths of one percent for counting time intervals of 8 to 10 h at radon activity concentrations of about 0.5 to 1 Bq L^{-1}. A substantial fraction of this uncertainty arises from the statistical "counting error" imprecision which could be further reduced and minimized with longer counting time intervals.

Background count rates for the new chambers, which primarily determine the measurement sensitivity, are approximately 0.008 counts per s. For a 4-L chamber volume, this corresponds to a background equivalent radon activity concentration of 0.001 Bq L^{-1} (or 0.03 pCi L^{-1}).

The new system presently is undergoing extensive testing and evaluation. Many additional tests, such as for adsorption losses of radon in traps, for the degree of gas purification, and for the transfer efficiencies as a function of flow rate and pressure differences, will have to be completed before the system is fully evaluated and operable. Conceptually, an overall accuracy (more correctly an *in*accuracy) for this primary measurement system of plus or minus several percent is achievable. However, uncertainties of this magnitude will be achieved only if the gas samples are carefully and quantitatively processed and transferred to the chamber, and if the chamber is accurately calibrated. Inasmuch as the system relies on comparative measurements, i.e., the assay is performed by comparing the output from an unknown radon sample with that from a known radium standard, the uncertainty in the radium standard and in its use is of paramount importance. In addition, it is clear that additional major sources of inaccuracy arise from errors in the gas-handling procedures used during both the sample processing and in the chamber calibration. These gas-handling procedures and subsequent counting techniques are a very complicated sequence of many individual operations or steps. It must be recognized that virtually every step in this sequence introduces a conceivable source of inaccuracy. The earlier evaluations performed in characterizing the original system [12] may no longer be completely applicable, and some of the present procedures used with that system may not withstand complete critical scrutiny. Therefore, it is necessary that the new system undergo a detailed evaluation for each step in order to obtain a complete uncertainty assessment.

5. Application to Other NIST Radon Transfer Standards and Calibration Services

As indicated, the pulse-ionization-chamber-based primary measurement system serves as the national standard for radon measurements and as such serves as an underpin to the entire NIST radon program. Its central role in linking all NIST radon and radium measurements, transfer standards, and calibration services is diagrammatically illustrated in figure 6. Brief descriptions of these transfer calibration standards and services, as well as some of the NIST activities and facilities used to support them are described below.

5.1 Radium-226 Standard Reference Materials

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

Most laboratories maintaining an independent radon calibration capability use these radium transfer standards as their "primary" standard for radon. Although more capable laboratories can be expected to utilize this standard with success, it is not a very practical transfer standard for laboratories which do not maintain a large internal calibration capability. The use of these radium solution standards requires an indirect radon calibration based on a very careful quantitative extraction, transfer, and collection of radon from a solution.

5.2 Radon Calibrations and Measurement Intercomparisons

Radon calibrations for other laboratories can be performed at NIST 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 NIST or in the sample bulb filled at NIST is directly relatable to the primary radon measurement system.

Approximately 2 years ago, a new secondary measurement system for these calibrations was introduced at NIST. A NaI(Tl) well counter was cross calibrated against the primary system. This detector used in conjunction with spherical 25-mL glass ampoules containing radon samples was shown to be very reproducible and fairly independent of geometry effects. The method has a wide dynamic range and can be used for radon-in-air samples from the picocurie level to 100 nanocurie level.

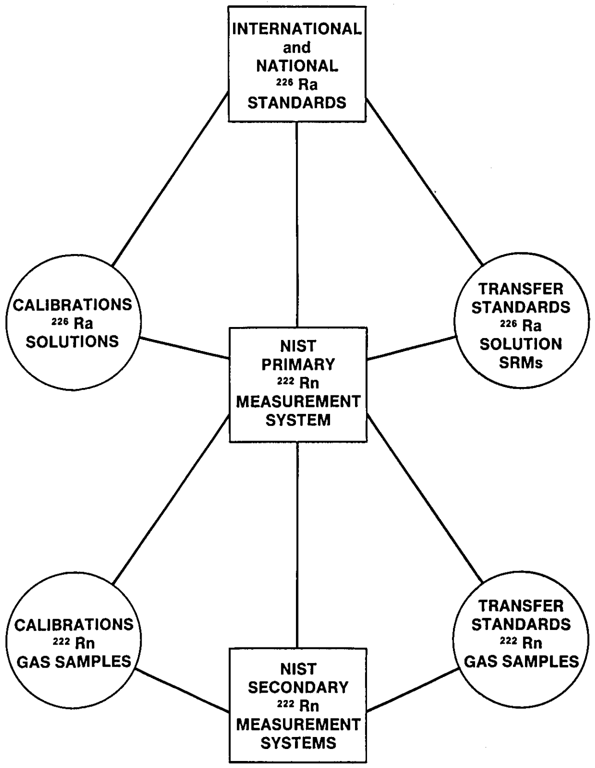

Figure 6. Illustration of the relationship of the NIST primary radon measurement system to other NIST radium and radon standards and calibrations.

This system was used for a recent measurement intercomparison of five principal laboratories within the United States that maintain an independent radon calibration capability based on radium solution standards. An unsatisfactorily large, systematic measurement discrepancy of over 7 percent among these laboratories was found despite the fact that all of the laboratories' calibrations were based on similar NIST radium solution standards. Our laboratory is continuing to work with these other laboratories to try to understand and resolve this discrepancy.

An international radon measurement intercomparison among international national metrology laboratories is also being planned for 1990.

5.3 Radon-in-Water Standard Generator

In 1986, NIST completed the development of a transfer standard for radon-in-water measurements. This work was initiated at the request of the U.S. Environmental Protection Agency to support the extensive national survey of radon concentration in drinking water supplies.

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. As in all other NIST transfer standards for radon, the generator calibration is directly relatable to the primary radon measurement system. The overall uncertainty in the radon calibration for a sample aliquot dispensed from the generator (such as into liquid scintillation vials) is estimated to be approximately 4%.

Additional details on the development, operation, and performance of the standard generator may be found in Hutchinson, et al. [13,14].

5.4 Radon Flux Density Standard

A project to develop a large surface area flux density standard (i.e., the radon exhalation rate per unit area) has been underway for several years [15].

A few attempts have been made by other laboratories to obtain a radon flux density calibration facility based on the characterization of a fabricated site of either natural or enriched radiumbearing soils, sands, or mill tailings. The novel, alternative approach considered for the NIST standard consists of incorporating an aqueous solution of radium into a sealed, shallow, large-surface-area container which is covered with a thin, but rigid, polyethylene sheet which is permeable to radon. The use of a radium solution eliminates possible spatial dependencies and minimizes the effects of ambient and meteorological variations. 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 movement in solid, porous media. A steady-state diffusion model for such a source configuration has been derived by Rubin [16] as an extension of the various radon transport cases treated in Collé et al. [17].

A 40-cm diameter prototype has been operational for the past 3 years. Preliminary evaluations for the constrained and unconstrained flux density have been completed. Although the efficacy of the flow-dependent extrapolation procedures used for calibrating the source were verified, problems arising from transpiration and evaporation of water through the polyethylene surface precludes the development of the prototype in its present configuration. Alternative designs incorporating small encapsulated radium sources under a rigid metal screen are under consideration.

5.5 Radon Transfer Standards Based on Polyethylene-Encapsulated Radium Sources

Based on our studies for the radon-in-water standard generator and the flux density standard prototype (secs. 5.3 and 5.4), it is believed that small polyethylene-encapsulated radium solutions would act as purely diffusive sources of radon that could be successfully used as transfer standards in a variety of configurations and measurement applications. The efficacy of such sources has been demonstrated in the development of the radon-in-water standard generator. These capsules would be an alternative or replacement for the present radium solution SRMs (see sec. 5.1).

Studies for their production and calibration have recently been initiated. At the present time, it is envisaged that the sources may be encapsulated using techniques originally developed for trace gas "permeation" tube calibration standards [18]. The development of suitable protocols for their use as SRMs also would be performed.

5.6 A Primary Calibration Based on Liquid Scintillation Counting

NIST also has work underway on a new initiative to perform a primary radon calibration which is not based on an indirect or comparative measurement against a ^{226}Ra standard. This may serve not only as an alternative primary radon calibration, but also as an independent verification of the primary radium standards. This calibration, expected to have an overall uncertainty of 1 to 2 percent, is based on liquid-scintillation-counting techniques using radium-free radon solutions obtained from the radon-in-water standard generator (see sec. 5.3). To date, extensive liquid scintillation measurements of radon-in-water samples have been made as a function of total sample volume and cocktail/water ratio. Comparative measurements against similarly prepared tritium (^{3}H) water samples have been initiated to fix a parameter in the model used to calculate the efficiency for counting radon progeny decaying by beta-particle emission. This primary calibration, if successful, will be a significant achievement in providing the first independent confirmation of the radium-based calibrations used over the past 50 years.

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The Closed-Can Exhalation Method for Measuring Radon

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Results from closed-can radon exhalation experiments must be interpreted bearing the time-dependent radon diffusion theory in mind. A rapid change from the free to final steady-state exhalation rate will take place for all samples that are thin compared with the radon diffusion length. The radon gas accumulating in a closed can corresponds to a free exhalation rate only if the outer volume of air is much larger than the pore volume of the enclosed sample, or the thickness of the sample is much larger than the radon diffusion length.

Key words: closed-can method; exhalation; radon; radon diffusion.

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1. Background

The closed-can method is a well-known and established way of measuring radon outgassing from porous samples. Unfortunately, the interpretation of experimental results has frequently been based on assumptions that are in conflict with the basic laws of diffusion. The erroneous and, one could say, classical approach is to fit the radon build-up in the accumulator to the equation

$$A=\frac{E}{\lambda}(1-e^{-\lambda t}) \tag{1}$$

where A = radon activity in enclosure (Bq),
λ = decay constant of radon-222 (s^{-1}),
E = exhalation rate (Bq s^{-1}),
t = duration of enclosure (s).

It is assumed without evidence that the exhalation E is initially constant, typically for several hours after closing the accumulator. Diffusion theory calculations show that this is not true for samples which are thin compared with the diffusion length. Including effects of leakage and the so-called "back diffusion" in eq (1) by incorporating an "effective" decay constant λ^* instead of λ is even more suspect. Equation (1) is typical for first-order kinetics and cannot successfully be imposed on pure diffusive transportation processes.

2. Results and Discussion

A 20-cm thick sample in an accumulator of height 30 cm will typically exhibit the exhalation rate illustrated in figure 1 if the thickness, d, is less than the diffusion length, L. The corresponding cumulated concentration curve [fig. 1(b)] will, in an experimental situation, falsely give the impression of being linear.

In order to reveal the rapid initial change in exhalation rate experimentally one has to follow the radon concentration build-up in the enclosed air volume more or less continuously. In figure 2 theoretical and experimental radon values for a 26-cm

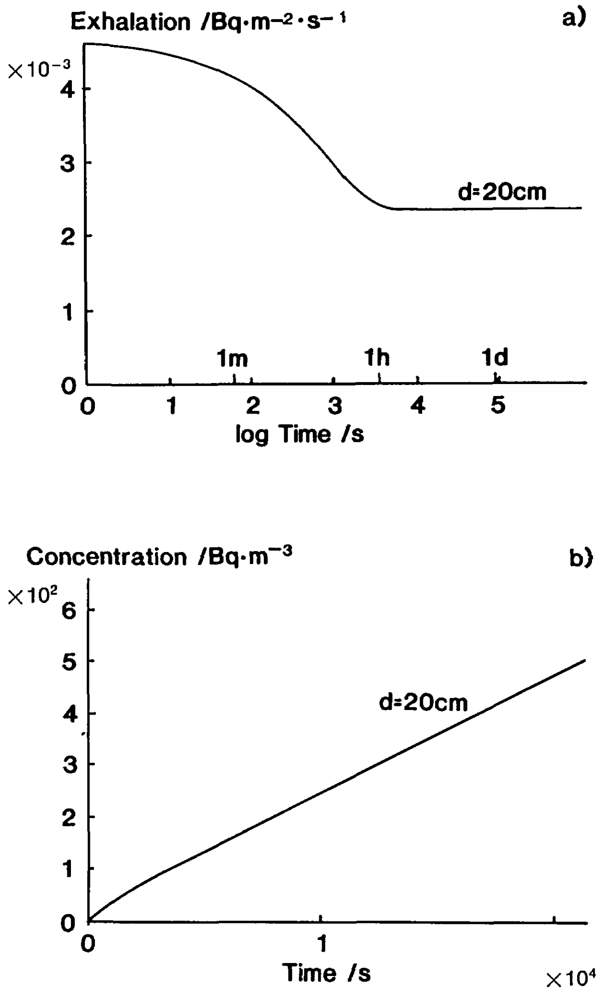

Figure 1. The temporal evolution of a) radon exhalation rate and b) the corresponding radon growth in the outer volume after closing a radon-tight accumulator. Diffusion length=2 m. (Theory).

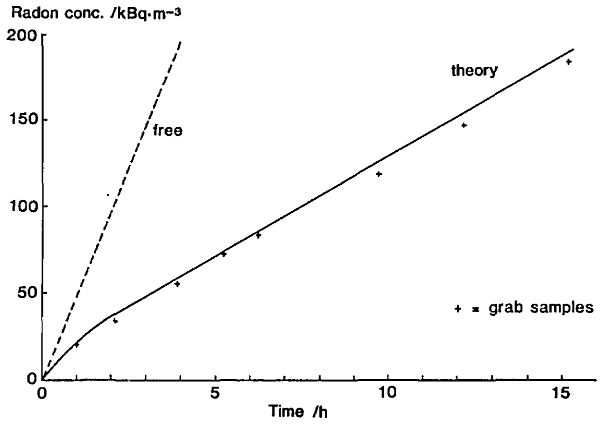

Figure 2. Radon concentration growth from enclosed dry sand mixed with 11% ground uranium ore by weight. Radon diffusion length=1.4 m, emanation fraction=0.33, radium-226 concentration =1.2 kBq kg^{-1}, and bulk density=1710 kg m^{-3}.

thick sample in an accumulator of height 30 cm are compared. The slope of the long linear part of the experimental curve in figure 2 corresponds to the final steady state exhalation rate, in this case about four times less than the free undisturbed exhalation rate.

Thin samples ($d<0.5L$) exhibit a final exhalation rate E_∞ that is related to the free exhalation rate E_f by the formula

$$\frac{E_f}{E_\infty}=1+\frac{1}{\alpha\gamma} \qquad (2)$$

where α=volume of the air above the sample divided by the pore volume of the sample,
γ=relative leakage factor [$\gamma=(\nu+\lambda)/\lambda$],
ν=leakage rate constant of enclosure (s^{-1}).

Choosing an outer air volume at least 10 times larger than the pore volume of the sample means that the depression of the free exhalation rate after closing the accumulator is less than 10%, a value acceptable in most experimental situations. If for reasons of sensitivity one has to work with small outer volumes, the initial drop in exhalation rate can be significantly reduced, but not completely avoided, if the outgassing radon is trapped in a charcoal trap or by other means in a flow-through system.

In cases where the change from free exhalation rate to final steady state exhalation rate is rapid enough (cf. fig. 1), eq (2) can be used to calculate the free exhalation rate because the slope of the radon growth curve is linear and corresponds to the steady state exhalation rate E_∞.

3. Conclusions

In general it is not relevant to fit simple exponential formulas to results from closed-can exhalation measurements.

Certain sample/accumulator geometries are definitely unsuitable for free exhalation determination unless the radon growth is monitored more or less continuously and the results are interpreted by means of a time-dependent diffusion theory.

The mean exhalation rate during the first hours is approximately equal to the free and undisturbed exhalation only if the outer volume of air is much larger than the pore volume or the sample thickness is much larger than the radon diffusion length.

4. Acknowledgment

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Standardization of Rn-222 at the Australian Radiation Laboratory

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The standardization of Rn-222 at the Australian Radiation Laboratory involves the calibration of scintillation cells by two methods using standard Ra-226 solutions traceable to the National Institute of Standards and Technology. One of these methods, namely the injection method, involves direct transfer of Rn-222 into a scintillation cell. In the other method, known as the volumetric method, the Rn-222 is flushed into a large container and the scintillation cell calibrated by sampling from this container. A comparison of the two methods showed that similar results were obtained, with the overall random uncertainty being 3.4% for one standard deviation. Using better estimates of the true calibration factors, the overall random uncertainty was reduced to 1.8% for one standard deviation.

Key words: Rn-222 calibration; scintillation cell; uncertainty components.

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1. Introduction

In 1983 the Nuclear Energy Agency (NEA) of the Organization for Economic Cooperation and Development (OECD) and the Commission of European Communities (CEC) jointly organized an international intercomparison program for measurements of radon, thoron, and daughter products [1]. The Australian Radiation Laboratory (ARL) was designated as the reference laboratory for the Pacific region. The primary method for Rn-222 measurements at ARL is based on scintillation cells which have been calibrated by Ra-226 solutions traceable to the National Institute of Standards and Technology (NIST). This report describes the calibration methodologies used in the Asian/Australasian Region Intercomparison for Radon which was completed in September 1988 [2].

2. Methodology

2.1 Injection Technique

The scintillation cells were calibrated at ARL using two complementary techniques, namely the direct or injection method and the volumetric method. In the injection method the scintillation cells were filled with a known activity of Rn-222 by bubbling radon-free air through a glass vessel containing a standard solution of Ra-226. The amount of Rn-222 present was calculated knowing the time of build-up since the solution was last flushed. The transfer of Ra-222 was effected by evacuating a scintillation cell (nominally of volume 0.14 L) and attaching it to the outlet valve on the standard solution container. Aged compressed air or nitrogen admitted from the inlet port was used to transfer the Rn-222 by bubbling through the source until the scintillation cell was at atmospheric pressure. The pressure was monitored at the outlet valve with an electronic pressure sensor (Druck[1]). Typical periods of approximately 20 min were required for completion of the transfer procedure. The scintillation cells were then counted using 30 repeated measurements of 10 min duration and a calibration factor to convert counts to concentration (Bq m^{-3}) was obtained from the known activity and cell volume. All 16 ARL cells were individually calibrated in this manner. However, repeat calibrations were obtained for some cells, in which case, the mean calibration factor was used as the individual calibration factor for the particular cell, with the standard deviation providing a measure of the random uncertainty due to the variability in the calibration procedure.

2.2 Volumetric Method

In the volumetric method the Rn-222 is transferred into a partially evacuated large bottle of volume about 1 L. Prior to calibration, the bottle was flushed with compressed air or nitrogen. On completion of Rn-222 transfer the Rn-222 to be sampled was slighty over pressure. After sampling, the scintillation cell pressure was equalized to atmospheric pressure. This method has two distinct advantages compared to the injection method. The first is that the volume of the bottle is known more accurately than the cell volume. The second advantage is that a much larger volume of air can be bubbled through the source, thus ensuring a more complete transfer of Rn-222. A disadvantage is that an additional transfer vessel is required, increasing the possibility of Rn-222 leakage, particularly since the vessel is overpressurized. No indications of system leakage were observed.

2.3 Reduction of Calibration Variability

The determination of the individual calibration factors of the set of 16 ARL cells using the above methods were influenced by physical differences between cells. These differences were attributable to two factors. The first factor is nonuniformities in the phosphor coating among the cells. The component of random uncertainty due to the resultant variability in scintillation response was estimated by filling each cell with the same radon concentration from a large Rn-222 chamber and analyzing the results of each cell using a calibration factor of unity for all cells.

The second factor is systematic differences in counting response in the bank of four photomultipliers used for simultaneous counting of cells in groups of up to four cells. During counting of the calibration samples, the 16 cells were divided into four separate groups, labelled N1-N4, with the individual cells of any group being considered as having a particular photomultiplier response. The appropriate relative response for each of the four photomultiplier tubes was determined by counting the one cell containing a known sample of radon gas in each of the four photomultiplier counting assemblies. In this manner, the effect on individual calibration factors so obtained was minimized.

However, the combined effect of the two responses resulted in small systematic biases in the calibration factors. The biases were observed as Rn-222 meaurement variability which was larger than the random uncertainty expected from counting statistics only. This variability was reduced by using individual calibration factors which had been adjusted for the photomultiplier response of the cell relative to the mean response, assuming that the mean calibration factor was the best estimate of the true calibration factor. These new calibration factors, Y_{rel}, based on the relative sensitivities of individual cells, were calculated from

$$Y_{\mathrm{rel}}=\frac{\text{Cell Counts}}{\text{Mean Cell Counts}}\cdot Y_{\mathrm{mean}} \qquad (1)$$

and were experimentally determined by filling each scintillation cell with the same concentration of gas from a large Rn-222 chamber.

3. Results

3.1 Injection Technique

The results for one cell calibrated using this technique are shown in table 1. The numbers in the columns refer to replicate calibrations of the same cell and the mean and standard deviation ($1s_i$) were obtained from the repeat determinations. In this case, for the same cell having a nominal volume of 0.14 L, the mean of four calibrations was determined to be 2260×10^{-6} counts s^{-1} (Bq $m^{-3})^{-1}$ [0.699 counts min^{-1} (pCi 0.14 $L^{-1})^{-1}$] with the variability in the calibration procedure being 2.3% for $1s_i$.

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[1] Certain commercial equipment, instruments, or materials are identified in this paper to specify adequately the experimental procedure. Such identification does not imply recommendation or endorsement by the National Institute of Standards and Technology, nor does it imply that the materials or equipment identified are necessarily the best available for the purpose.

Table 1. Values of calibration factors for one ARL cell using injection and volumetric methods counts$\times 10^{-6}$ s^{-1}(Bq m^{-3})$^{-1}$

Method	(1)	(2)	(3)	(4)	Mean$\pm 1s_i$
Injection[a]	2230	2200	2310	2300	2260±52[b]
Volumetric[a]	2240	2340	2240		2273±55[c]

[a] Difference in Means=0.3%.
[b] Injection $1s_i$ =2.3%.
[c] Volumetric $1s_i$ =2.4%.

3.2 Volumetric Method

The calibration factors of a single scintillation cell obtained by using the volumetric method are also shown in table 1. The mean of three replicate determinations was 2270×10^{-6} counts s^{-1} (Bq m^{-3})$^{-1}$ [0.701 counts min^{-1} (pCi 0.14 L^{-1})$^{-1}$], with a random uncertainty of 2.4% for $1s_i$. Both the mean and uncertainty compare favorably with the values obtained from the injection technique, the difference in means being 0.3%; clearly both techniques are comparable in accuracy and precision. However, as discussed below, due to the small volumes involved in the injection method, there was incomplete transfer of Rn-222 in some cases, leading to the rejection of some erroneous results. This action was unnecessary in the volumetric method, indicating a more reliable transfer of Rn-222 into the larger volume.

Since the individual measurements of Rn-222 using scintillation cells were also influenced by physical differences between cells, this component of variability was estimated (sec. 2.3) using results from cells which were exposed to the same Rn-222 concentration and analyzed using a calibration factor of unity for all cells. These results are listed in table 2, and exclude the component of systematic differences due to individual counting response of the four photomultipliers.

Table 2. Variability in Rn-222 measurements between ARL scintillation cells exposed to same Rn-222 concentration and analyzed using a calibration factor of unity (Bq m^{-3})

Group	(1)	(2)	(3)	(4)
N1	3290	3350	3430	3470
N2	3480	3570	3450	3470
N3	3350	3500	3480	3500
N4	3410	3520	3690	3470

Grand mean=3460±90 Bq m^{-3} or 2.6% at $1s_i$.

3.3 Reduction In Calibration Variability

The measurements of Rn-222 concentration using the original calibration factors determined by the volumetric method are shown in table 3 and are to be compared with the measurements of the same Rn-222 samples using the adjusted calibration factors as shown in table 4. The numbers in the columns are the measured concentrations with each group (N1-N4) comprising four replicate measurements each. The grand means and grand s_i were determined from the 16 replicate measurements. It can be seen from tables 3 and 4 that the grand means were virtually identical, the difference being 0.4%. However, the overall random uncertainty or grand s_i obtained using the old calibration factors was 3.4%, which is significantly larger than the value of 1.8% using the adjusted calibration factors.

Table 3. Rn-222 concentrations calculated using original calibration factors (Bq m^{-3})

Group	(1)	(2)	(3)	(4)
N1	2580	2610	2700	2690
N2	2600	2690	2690	2640
N3	2560	2660	2870	2510
N4	2660	2670	2780	2680

Grand mean=2660±90 Bq m^{-3} or 3.4% for $1s_i$.

Table 4. Rn-222 concentrations determined using adjusted calibration factors (Bq m^{-3})

Group	(1)	(2)	(3)	(4)
N1	2690	2630	2680	2650
N2	2590	2630	2660	2690
N3	2620	2600	2730	2690
N4	2540	2680	2670	2690

Grand mean=2650±50 Bq m^{-3} or 1.8% for $1s_i$

4. Discussion

4.1 Injection Technique

The sources of random uncertainty applicable to the technique include components which have been described in the previous section. Since calibrations at ARL are now standardized on the volumetric method, further discussion is confined to components of interest.

4.1.1 Variability In Calibration Procedure This was determined (table 1) from four repeat measurements on the same scintillation cell to be 2.3% for 1s_i. However this value was obtained from measurements in which erroneous results were excluded.

4.1.2 Variability Between Scintillation Cells From table 2, the determination of this component from 16 replicate measurements was 2.6% for 1s_i.

4.1.3 Efficiency of Rn-222 Removal by Bubbling The potential for a less reliable transfer of Rn-222 mainly associated with the injection technique arises from the small volume (0.14 L) of air passing through the Ra-226 solution, hence the possibility that some Rn-222 may not have been transferred to the cell. Measurements were carried out to determine the fraction not transferred. A second scintillation cell was used to measure the amount left behind in the solution by flushing with air immediately after a calibration transfer. A small percentage (0.87%) of Rn-222 was found in this second cell. While this amount may be a small percentage of the present transfer in absolute terms, it may constitute a substantial fraction of the next transfer. For example, if the Ra-226 standard solution is bubbled when the Rn-222 is close to full strength, the small remaining fraction will contribute up to 14% if the source is reused within 8 h. In practice, these calibration results were rejected, but the potential for small systematic biases of the order of a few percent still exists. These biases are then observed as Rn-222 concentration variability between cells and variability between repeat calibrations for any individual cell. The approach based on mean and relative sensitivity adjustment of individual calibration factors should minimize the effect of such biases.

4.1.4 Estimate of Volume of Cell This quantity applies to the injection technique only. Since the cell calibration is intended to provide the user with an estimate of the cell response per unit concentration of Rn-222 in the environment the injection method is only valid providing the volume of the scintillation cell is known accurately. The volume of the cells was measured by filling a sample cell with water and weighing the water to determine the volume of the cell. The cell volume was determined with an accuracy of 0.3% from nine repeat measurements. It was assumed this cell was representative of the batch of 16 cells used. Cell volume variability is believed to be less than 1%.

4.2 Volumetric Method

4.2.1 Variability in Calibration Procedure The random uncertainty of 2.4% for 1s_i was in good agreement with that found for the injection technique. Since the Rn-222 transfer was more reliable using the volumetric method, other sources contributing to this variability should be investigated. The most likely contributor is the variability in residual Rn-222 in the solution and dead space, which was caused by the variability in effective volume transferred due to the partial evacuation of the bottle. Perhaps complete evacuation of the bottle is necessary.

4.2.2 Variability Between Scintillation Cells As is shown in table 2, the variabilty due to real differences between cells amounts to 2.6% for 1s_i. These differences are attributable to variability in scintillation response and exclude the effect of photomultiplier scintillation counting response, as described in section 2.3.

4.2.3 Statistical Uncertainty Due to Counting This is normally small due to the large activity transferred and the length of time used to count the sample. For the counting periods and concentrations of Rn-222 used in our laboratory, this uncertainty is usually less than 1%.

4.2.4 Estimate of Volume in Volumetric Method In this case, the volume was determined in the same manner as for the cell, and the uncertainty was estimated to be 1 cm^3 in 1300 cm^3, or 0.08%.

4.2.5 Leakage of Rn-222 From the Vessel Holding the Standard To minimize this possibility, the vessel was held at negative pressure relative to atmospheric after transfer.

4.2.6 The Pressure of the Air in the Scintillation Cell Any change in pressure within the scintillation cell will change the range of alpha particles and so alter the number striking the zinc sulphide coating on the cells and hence the cell calibration. Therefore all cells were brought to the same pressure on completion of the transfer of the radon.

4.2.7 Humidity of the Air This was reduced using a dry ice trap in early work, but did not appear to affect the calibration results and so the practice was discontinued.

4.2.8 Overall Random Uncertainty In the estimation of overall random uncertainty in the determination of Rn-222 concentration by any one ARL scintillation cell calibrated by the volumetric method, the pertinent components which should be considered were

1) Variability in repeat calibration factor for one cell $=2.3\%$ (s_i with $n=4$) for any cell

2) Variability due to cell differences $=2.6\%$ (s_i with $n=16$) for any cell

3) Counting statistics during Rn-222 measurement $=0.9\%$ (SEOM with $n=30$)

where SEOM = standard error of the mean. The overall random uncertainty for the determination of Rn-222 concentration for any cell can be calculated by adding the components as follows:

$$(0.023)^2+(0.026)^2+(0.009)^2=(0.036)^2,$$

which is 3.6% and in good agreement with the measured value of 3.4% shown in table 3. The good agreement also indicates that most of the relevant sources of random uncertainty have been considered.

The systematic uncertainty was mainly derived from the total uncertainty of the Ra-226 solution, ignoring the contributions of other minor components. This solution was supplied by Amersham (UK) and traceable to NIST, and from the certificate, the random uncertainty applicable to the source was 0.7% at 3 standard deviations, or 0.2% at one standard deviation, with the systematic uncertainty estimated to be 3.0%. Using the recommendations of Collé et al. [3], the systematic uncertainty in the Rn-222 measurement due to the total uncertainty of the source was calculated as follows:

$$(0.002)^2+(0.030)^2\cong(0.030)^2$$

which is 3.0%. Hence the total uncertainty for any ARL scintillation cell was calculated from,

$$(0.036)^2+(0.030)^2=(0.047)^2$$

which is 4.7%, and both the random and systematic uncertainties are contributing comparable amounts to the total uncertainty.

4.3 Reduction in Calibration Variability

The results of Rn-222 concentration determined from the new individual calibration factors are shown in table 4; it is clear that the grand s_i has been reduced from 3.4% to 1.8%. Correspondingly, the variability due to differences between cells was reduced from 2.6% to 0.9%, which is comparable with the counting statistics.

5. Conclusions

The results of standardization of Rn-222 by ARL using two techniques clearly shows there is no difference between calibrations using the injection or volumetric methods, with the latter providing the reliable transfer of Rn-222. Calibration factors based on the volumetric method resulted in a reduction from 3.4% to 1.8% in the measurement uncertainty for Rn-222 by any ARL scintillation cell.

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Standardization of Radon Measurements: II. Accuracy and Proficiency Testing

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The accuracy of *in situ* environmental radon measurement techniques is reviewed and new data for charcoal canister, alpha-track (track-etch) and electret detectors are presented. Deficiencies reported at the 1987 meeting in Wurenlingen, Federal Republic of Germany, for measurements using charcoal detectors are confirmed by the new results. Accuracy and precision of the alpha-track measurements laboratory were better than in 1987. Electret detectors appear to provide a convenient, accurate, and precise system for the measurement of radon concentration. The need for comprehensive, "blind" proficiency-testing programs is discussed.

Key words: alpha-track detector; calibration; charcoal detector; electret detector; proficiency test; quality assurance; radon measurement; track-etch detector; uncertainty.

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1. Introduction

Since the 1987 meeting of the International Committee for Radionuclide Metrology (ICRM) Low Level Measurements Group in Wurenlingen, Federal Republic of Germany, the New York State Department of Health (NYSDH) has continued to evaluate the performance of *in situ* radon measurement devices available to homeowners and other users: charcoal canisters, alpha-track detectors (ATD) and electret. We have been concerned with the potential accuracies of the available measurement techniques, as well as the effectiveness of ongoing proficiency testing programs that aim to provide a link between these measurements and national standards.

2. Commercial Radon Measurements in the United States

Most of the routine radon measurements performed in the United States are made by commercial laboratories in an extremely competitive market which has caused price to be a major factor in the selection of a radon-measuring system. Unfortunately, there are many uncertainties which can contribute to the quality of data generated by various radon-measurement systems.

2.1 Charcoal Detectors

Charcoal detectors have been found to be unreliable for measurements, because of the very short integrating period [1] as well as the high degree of variability in radon adsorption efficiency caused by displacement of adsorbed radon from the charcoal by water which is also adsorbed during the collection period [1, 2].

Since the Wurenlingen meeting, we have tested four more sets of charcoal detectors using the radon calibration chamber operated by the Environmental Measurements Laboratory (EML) of the U.S. Department of Energy [3]. Three of the detector sets were obtained from two commercial laboratories: 7-cm diameter by 2.5-cm thick charcoal canisters from Laboratory A[1] and two sets of 10-cm diameter by 3-cm thick charcoal canisters, differing by the type of charcoal used as adsorbent, from Laboratory B. A set of 10-cm diameter by 3-cm thick canisters prepared in the Wadsworth Center for Laboratories and Research (WCLR), NYSDH, were also tested. All expose a screened, fully open face for adsorption of radon. Sets of three detectors of each type were exposed in the EML radon chamber for 1,3,4,5, and 7 d, respectively—a total of 15 detectors of each type. Exposed detectors were measured gamma-spectrometrically on a calibrated Ge(Li) system. Also, results published by Ronca-Battista [2] for 10-cm diameter by 3-cm thick charcoal detectors were compared to the data we obtained.

Two of the tested detector sets, Laboratory A and WCLR, use Calgon charcoal, as does the U.S. Environmental Protection Agency (EPA) [2]. Radon adsorption onto the WCLR detectors correlates well with that obtained by Ronca-Battista. The Laboratory A detectors, with approximately half the amount of charcoal, adsorb approximately half as much radon as do the WCLR and EPA detectors for each exposure period. The amount of radon adsorbed per gram of charcoal is approximately the same for each of the three sets of detectors, but the Calgon charcoal used by the WCLR adsorbs approximately twice as much water per gram as do the other two.

The two sets of Laboratory B detectors which use charcoal of unspecified origin showed markedly differing behavior from one another for both radon and water adsorption, apparently due to different brands of charcoal used as adsorbent. The charcoal in the newest Laboratory B detectors has more ^{137}Cs activity than other charcoals we have tested. Although the ^{137}Cs activity could interfere with radon-daughter activity measurements of exposed canisters, Laboratory B uses high-resolution gamma-spectrometric analysis measurements and hence ^{137}Cs presumably does not interfere. The new type of charcoal used by Laboratory B resulted in radon and water adsorption similar to that for the WCLR Calgon- loaded detectors. The charcoal used in the canisters available earlier from Laboratory B showed very poor radon adsorption and very high water uptake—performance very much like that exhibited by the bag type of detector described previously [1]. The new Laboratory B detectors, therefore, are superior to the old ones and approximately equivalent to those used by Laboratory A, EPA, and WCLR in the time over which they can be exposed.

These studies on charcoal detectors add to our previous evaluation [1] that the exposure time for charcoal detectors (at most, 2 to 4 d) is too short to provide a representative measurement of radon concentration in a home. Furthermore, it appears that selection of the wrong type or quantity of charcoal or container may compound the problem of short exposure time such that measurements using certain types of charcoal detectors may be in error by much more than tenfold from the actual radon concentration. (Since publication of the Wurenlingen proceedings [1], a representative of a particular company telephoned to state that the company has ceased using the bag-type detectors because they are considered unreliable. Other companies still are providing bag-type detectors for commercial measurements, however.)

2.2 Alpha-Track (Track-Etch) Detectors

The State of New York also continues to contract for radon measurements using alpha-track detectors (ATD), also called track-etch detectors. Although the price per measurement is greater for an ATD than for one using a charcoal detector, recognition of the need for confirmatory measurements using a long-term integrating device requires that the State purchase the more expensive ATDs. So far, our tests of ATDs have been limited. As reported previously [1], when Laboratory C bought out one of its competitors, the competitor laboratory's customers were required to submit exposed ATDs to Laboratory C for reading. The results were unreliable, with values ranging over an order of magnitude from the low value to the high and differing from the reference value by × 1.5 and ÷8. Laboratory C ATDs read by Laboratory C were tested for this study and the results are listed in table 1. Exposures at EML were as previously described [1]. Various combinations of the exposed ATDs were mailed to Laboratory C on four separate days, because of processing deficiencies noted previously [1]. This time, however, the date of

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[1] Certain commercial equipment, instruments, or materials are identified in this paper to specify adequately the experimental procedure. Such identification does not imply recommendation or endorsement by the National Institute of Standards and Technology, nor does it imply that the materials or equipment identified are necessarily the best available for the purpose. The views expressed in this article are solely those of the author and do not necessarily represent the views of the National Institute of Standards and Technology or the Department of Commerce. Neither NIST nor the DoC endorses or in any way approves the products or services of any company.

Table 1. Radon concentrations obtained using Laboratory C alpha-track detectors. Uncertainty values have been calculated from Laboratory C's reported percentage uncertainties, but the significant figures are consistent with those reported by Laboratory C. Reference values are listed as received from EML.

	Radon concentration (pCi/L±1 SD) by exposure time					
Exposure Time	1 d	2 d	3 d	4 d	5 d	7 d
	40.4±7.6	27.0±4.5	41.1±4.7	35.9±3.8	35.8±3.4	39.8±3.1
	37.0±7.3	35.8±5.2	35.4±4.3	38.8±4.0	29.7±3.1	32.8±2.9
	38.7±7.4	38.8±5.5	31.5±4.1	39.2±4.0	38.5±3.5	35.4±2.9
Mean	38.7±1.7	33.9±6.1	36.0±4.8	38.0±1.8	34.5±4.5	36.0±3.5
Reference	41.1	41.8	42.5	42.0	44.4	43.6

mailing did not produce a statistically significant variation in results, although we do not have information as to the actual date that Laboratory C processed each of the ATDs. There appears to be an overall negative bias in the ATD results of approximately 15%. The lowest values were only 65% of the reference value and none exceeded it.

We have not tested whether other, smaller laboratories which use other types of ATDs, can produce equally or more reliable results.

2.3 Electret Detectors

Other types of radon-measuring devices are for the most part either non-integrating, too bulky or fragile to mail, or too expensive to compete in cost with charcoal detectors or ATDs. The possible exception is the electret detectors.

An electret detector system, manufactured by a commercial company [4], was made available to us just one month prior to this symposium. The system is based on an electret detector made of a thin Teflon disc to which a charge of approximately 700 to 750 V is applied and which is then placed into a specially designed chamber. A separate reader is used to measure the charge on the disc prior to exposure and again after exposure to determine the loss of charge due to ionizations caused by impinging alpha particles. Charge loss is then equated to radon concentration using previously developed calibration factors and other correction factors. The results of exposing 15 short-term (typically exposed in a home for no more than 30 d) and 15 long-term (usually exposed for a few months to a year) electret detectors are summarized in table 2.

The electret detectors are convenient to use. Within 2 h of beginning to process the electret detectors for the first time in our laboratory, measurements were completed, results calculated, and data tabulated. In this instance we used a reader, calibration equation and calculational program provided by the manufacturer.

Table 2. Radon concentrations measured using short- and long-term electret detectors. Reference values are reported as received from EML

Exposure time (days)	Radon concentration (pCi/L±1 SD)[a]		
	Reference value	Short-term detectors	Long-term detectors
1	41.1	35.6±2.0	30.3±7.5
3	42.5	39.7±3.8	42.8±1.2[b]
4	42.0	40.3±0.9	41.0±2.3
5	44.4	43.6±3.3	42.1±2.1
7	43.6	42.6±0.9	41.9±0.6

[a] Mean of 3 measurements except as noted.
[b] Mean of 2 measurements.

One of the long-term detectors exposed for 3 d exhibited an abnormally high rate of discharge. Although it was suggested by the manufacturer that this may have occurred from someone inadvertently touching the disc when it was removed for reading, further charge measurements indicate that the high discharge rate continued during the following month. This potential source of error remains to be resolved, but one poor measurement out of 30 is a relatively low incidence of inaccurate data compared to the other systems studied. Also, from the standpoint of risk evaluation or remediation requirements, the false positive result created by excess loss of charge is not as serious as a false negative measurement. Presumably, a false positive value would be detected by follow-up measurements. Kotrappa et al. [4] provide a detailed discussion of the advantages and disadvantages of electret detectors.

Uncertainties (expressed as the standard deviation for replicate measurements) are small for the short-term electrets (table 2) even to exposures of 300 pCi d/L. There does not appear to be any significant bias in the short-term results. Although many values in table 2 are slightly negative relative to the corresponding reference value, they mostly seem to be within the statistical uncertainties [4] to be expected for this system.

The results for the long-term electret detectors were initially biased low by approximately 10%. When questioned, the manufacturer found from other users as well that the supplied calibration equation produced low results. A new calibration equation, which increased the long-term results by 10%, was sent to users of the system. The corrected results are the ones listed in table 2.

All of the results of the long-term electret detectors, except those for the shortest exposure time of 1 d, produced small uncertainties (again expressed as standard deviation among replicate measurements). Voltage loss is 5 to 8 V when the long-term electret detectors are exposed for 1 d at a radon concentration of 41 pCi/L. Since the reader variability is expected to be ±2 V [4], the observed standard deviation at this low exposure level is consistent with that predicted by the manufacturer. At a greater concentration of radon, or a longer exposure time, reader variations become small relative to voltage changes, so uncertainties become correspondingly small. There was insufficient time prior to the meeting to test the long-term electret to its maximum integrated-exposure level.

3. Discussion

Our earlier conclusion [1] that charcoal detectors are unreliable for anything but the crudest of radon measurements appears to remain valid. Charcoal detectors cannot be recommended even for screening measurements, because any particular measurement may be nonrepresentative. Especially critical would be a situation where the measured value turns out to be lower than the actual concentration by such a large amount that the homeowner fails to take corrective action even though mitigation is warranted (see discussion in [1]).

It appears that Laboratory C has markedly improved its performance with ATDs from that reported previously [1]. However, the overall negative bias, the fact that 4 of 18 measurements were biased low by 25% or more, and the overall range in values found in our most recent study raise questions about using these detectors for recommendations of remedial action. The generally poor performance of ATD vendors in EPA's proficiency testing program [5], coupled with Laboratory C's performance in our studies, indicate that ATD vendors should be tested more comprehensively using double-blind proficiency samples.

The electret detectors impressed us with their ease of handling and rapidity of the measurement process as well as their generally excellent precision. The fact that we obtained meaningful results even when we tested the long- and short-term detectors at their limits for low exposure and high exposures, respectively, suggests that the electret detectors can provide reliable measurements when used properly. Especially important is that in most cases when errors occur, such as that resulting from a high rate of discharge, the result is a false positive and would likely be corrected when follow-up measurements are performed.

One important source of error with the electret detectors can occur if the electrets are used outside their respective exposure ranges. False-negative values could develop if care is not taken at very high radon concentrations or over excessively long exposure times. Since the electret detectors (as also the ATDs) actually measure the time-integrated exposure (pCi d/L), radon concentration is derived from the ratio of integrated exposure and the time of exposure. Unlike an ATD, which continues recording alpha tracks regardless of how long it is in place, an electret detector left in place after it is fully discharged could produce a false negative value.

Another important potential error in using the electret detectors would be to use them in a home for very short-term measurements, say of the order of a few days. Any short-term measurement with the electret detectors would suffer from short-term fluctuations in radon concentration, much as described for charcoal detectors [1]. The large day-to-day fluctuations of radon concentration in a home require reasonably long-term, integrated measurements regardless of which type of detector is used.

This discussion concerning electret detectors highlights in another way the fallacy of relying on charcoal detectors which begin losing some of their adsorbed radon immediately. Some, like the bag and the old Laboratory B detectors, fail after only 1 d of exposure; others after 2 or 3 d. At least with the electret detectors, a false negative would occur only after an excessive exposure, and the potential error would be easily recognized by the low charge reading at the end of exposure. With charcoal detectors there is no easy way to identify a false negative result.

Some other incidental concerns for the electret detectors, such as discharge during storage, were raised during the discussion following this presentation. However, these issues involve cost management and are not necessarily critical to risk management. An excessive discharge rate during measurement because of a high gamma-radiation field could produce a false- positive result if not properly corrected, but this is preferable to a false negative and can be corrected by simply measuring the gamma-dose rate when the detector is installed in a home [4].

The sources of error, especially for charcoal detectors and ATDs, apparently are not being corrected by the U.S. EPA proficiency-testing program, the deficiencies of which have been noted previously [1]. Separate "double-blind" studies performed by *The Patriot News*, a Harrisburg, Pennsylvania newspaper, and by Public Citizen—Buyers Up, a Washington, DC consumer-interest group, found that many companies certified by EPA reported results which were inconsistent and contradictory. Some laboratories failed even to duplicate their own results. Unfortunately, this supplemental information is in the popular press rather than the scientific literature. It does support our contention, however, that a much more comprehensive proficiency testing program is necessary in the United States.

Part of the reason for this symposium, especially the workshops, has been to explore various means for transferring the technology of radon measurements to any country desiring the technology. The four IPPP laboratories have successfully exhibited that standards can be made traceable across continents as well as across borders. However, the U.S. situation indicates that traceability alone may not be sufficient to assure the quality of radon measurements. The problem should not be dismissed as a peculiarity of an entrepreneurial economy. Whenever and wherever cost is a major consideration, measurement quality is likely to suffer. A comprehensive international quality assurance program needs to be developed.

There appears to be an important role to be served by the ICRM. It can provide a system to assure the quality of radon measurements by specifying which systems or detectors are acceptable. Also, it can develop or approve protocols for proficiency testing of measurement laboratories and/or systems. Most importantly, the ICRM can provide a framework for organizing intercomparisons wherein the tested laboratories are not aware of the significance of the submitted detector (i.e., by setting up a system for performing double-blind proficiency tests).

4. Acknowledgments

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Conference Reports

CONFERENCE ON FIFTY YEARS WITH NUCLEAR FISSION Washington, DC and Gaithersburg, MD April 25–28, 1989

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The National Institute of Standards and Technology (NIST) and the American Nuclear Society jointly sponsored this unique conference to celebrate the fiftieth anniversary of the discovery of nuclear fission. An international audience of over 400 scientists and engineers joined the distinguished early pioneers to reminisce about the past and to study recent developments of this important technological discovery. The conference highlighted the early pioneers of the nuclear community by dedicating a full day of plenary presentations at the National Academy of Sciences in Washington, DC and later at the conference banquet. More recent developments in fission science and technology in addition to historical reflections were topics for two full days of sessions at NIST in Gaithersburg, MD. The meeting was extremely fortunate in having Professors John A. Wheeler and Edoardo Amaldi serve as Honorary Co-Chairmen and in having Professors Glenn T. Seaborg and Emilio Segrè serve as General Co-Chairmen. The conference attendees were saddened by the death of Professor Segrè on April 22, 1989, just three days before the start of the conference.

1. Opening the Conference

The conference was opened by welcoming remarks by Raymond G. Kammer, acting director of NIST and Gail de Planque, president of the American Nuclear Society. In his remarks, Kammer noted the relation of the National Bureau of Standards to the development of fission energy. Nuclear fission was discovered in Germany in late 1938 by Hahn and Strassmann. The news was brought to the United States in January of 1939 by Neils Bohr. Within days the discovery was announced in Washington, DC at a conference on theoretical physics at George Washington University. The results were soon repeated at the Department of Terrestrial Magnetism and at other laboratories throughout the nation.

The announcement at the Washington conference made the front page of the January 26, 1939 edition of the Washington Evening Star newspaper under the headline "Power of New Atomic Blast Greatest Achieved on Earth" with the subtitle "Physicists Here Hail Discovery Greatest Since Radium." After an extensive discussion of the physics of the process, the author stated that "the practical uses of the discovery remain vague. It may place hitherto undreamed-of resources in the hands of physicists for experimental purposes and help clear up some of the outstanding mysteries of creation." And finally the article concluded with the statement "As a practical power source, the new finding has at present no significance."

It is interesting that the director of the National Bureau of Standards, Lyman Briggs, was appointed by President Roosevelt to chair the first federal government committee to study the possible uses of this new development in nuclear science. The first meeting of the Advisory Committee on Uranium met at the Department of Commerce on October 21, 1939. The meeting included Professor Teller who was the author of a contribution to this conference. The report, dated November 1, 1939, stated that a chain reaction was a possibility. It speculated on the possibility of a new explosive for the military as well as a new source of energy for submarines for the navy. The report recommended that 4 tons of pure graphite be obtained at once for research and that later 50 tons of uranium ore be acquired. Three months later $6000 was made available to purchase a small quantity of graphite for experiments. Thus began the government support for the start of the immense enterprise of nuclear energy.

2. Conference Topics

The wide range of topics covered by this meeting included plenary sessions entitled:

Prelude to the First Chain Reaction—1932 to 1942
Early Fission Research—Nuclear Structure and Spontaneous Fission
50 Years of Fission, Science, and Technology
Nuclear Reactors, Secure Energy for the Future
Nuclear Fission—A Prospective,

invited sessions entitled:

Reactors
Fission Science
Safeguards and Space Applications
Fission Data
Nuclear Fission—Its Various Aspects
Medical and Industrial Applications of By-Products,

and contributed (both oral and poster) sessions on:

Theory and Experiments in Support of Theory
Reactors and Safeguards
General Research, Instrumentation, and By-Products
Fission Data, Astrophysics, and Space Applications.

3. Summary of Presentations on the Early Work on Fission

The titles and authors, along with a summary of several of the papers presented on the early work on the discovery of fission and applications of nuclear energy are given in this section.

1. "The Prelude to Fission, Italy," Edoardo Amaldi (University of Rome-Italy)

After the discovery by Irene Curie and Frederic Joliot of artificial radioactivity induced by alpha particles, Fermi thought that similar effects could be observed also by using neutrons. In 1934 Fermi and co-workers (Amaldi, D'Agostino, Rasetti, Segrè) irradiated 60 elements with neutrons, found 44 new radioactive bodies, and in 16 cases, identified the chemical nature of the product of the reaction. They demonstrated that two artificial radioactive bodies produced in U, one of 13 min (later 15 min) half-life, the other about 100 min half-life, were not due to elements with atomic number between 82 and 92. Fermi et al. suggested that these bodies could be radioisotopes of transuranic elements of $Z=93$ and 94 produced by neutron capture in ^{238}U followed by two successive beta
decays.

2. "The Prelude to Fission, France," P. Savić (Serbian Academy of Sciences and Arts-Yugoslavia)

A personal account of the events leading to the 1938 discovery, by Irene Joliot Curie and the author, of the unidentified element $R_{3.5h}$ under neutron bombardment of uranium was presented, as well as the experimental methods proving that $R_{3.5h}$ had chemical properties similar to lanthanum. This in turn led to Hahn and Strassmann's discovery of fission in 1939.

3. "How Fission Was Discovered," Siegfried Fluegge (University of Freiberg-Federal Republic of Germany)

After the great survey of neutron-induced radioactivity by Fermi and coworkers, the laboratories in Paris and Berlin-Dahlen tried to disentangle the complex results found in uranium. At that time neutron sources were small, activities low, and equipment very simple. Chemistry beyond uranium still was unknown. Hahn and Meitner believed they had observed three transuranic isomeric chains, a doubtful result even then. Early in 1938, Curie and Savić in Paris found an activity interpreted to be actinium, and Hahn and Meitner another to be radium. Both interpretations seemed impossible from energy considerations. Hahn and Strassmann, therefore, continued this work and succeeded in separating the new activity from radium. There remained no doubt that a barium isotope had been produced, the uranium nucleus splitting in the yet-unknown process we now call fission.

4. "The Early French Program," Bertrand Goldschmidt (Commission L' Energie Atomic, Paris-France)

The work of Joliot's team from 1939 to mid-1940, a physical proof of fission, the reaction to Szilard's proposal of secrecy, the detection of the secondary neutrons and their quantitative measurements, the taking-out of secret patents, the tentative agreement with Union Minière du Haut Katanga and the procurement of uranium, the search for a moderator, the purchase of the world-wide stock of heavy water from Norway and its transfer to England in June 1940 were highlighted.

5. "Early Work in Copenhagen and in England," Rudolf Peierls (Nuclear Physics Laboratory-England)

Starting from the insight by O. R. Frisch and Lise Meitner, the talk reviewed Frisch's first observation of fission fragments, which caused him to propose the term "fission." This was followed by an account of the argument which led Niels Bohr to the realization that the slow-neutron fission was due to ^{235}U, and his conclusion that no explosive reaction was possible in natural uranium. The Frisch-Peierls memorandum, which suggested that the critical mass of ^{235}U was much less than suspected and that the assembly of a supercritical amount of ^{235}U would lead to an explosive reaction with a high yield, was presented.

6. "Experiments at Columbia and The University of Chicago which Led to the First Chain Reaction," Walter Zinn (GNEC-retired)

The author offered his recollections of the experimental efforts beginning in 1939 which culminated in the Chain Reaction in the squash court on December 2, 1942. Recalled were Columbia University experiments which did much to establish the feasibility of the chain reaction in natural uranium and which stimulated the creation of the Manhattan District. The Columbia group moved to the University of Chicago, where, in early summer of 1942, construction and analysis of a number of subcritical reactors (piles) gave assurance with a high probability that only a reasonable amount of uranium and moderator would be required.

7. "Fission in 1939: The Puzzle and the Promise," John Archibald Wheeler (Princeton University and University of Texas at Austin)

How come fission? Above all, how does it come about that thermal neutrons and neutrons of energies above a couple of MeV are good at inducing fission in uranium, but not neutrons of intermediate energy? Bohr's 1935-1937 compound-nucleus concept of nuclear reactions proved itself the key to this 1939 puzzle. To turn the key in the lock it was necessary to establish and exploit the concept of "fission barrier," an idea contested initially by more than one colleague. To add a slow neutron to the even-neutron nucleus ^{238}U does not produce enough excitation to surmount the barrier, but addition to the odd neutron ^{235}U does. The barrier theory proposed this, and his colleagues confirmed it a year later, which signalled a new world of activity. The same argument said that ^{239}Pu must be fissile, a circumstance of which Louis A. Turner was the first to point out the fantastic alchemical promise, to be released in Du Pont's 1944-1945 deliveries of plutonium in many-kilogram amounts to Los Alamos.

8. "Soviet Research into Nuclear Fission Before 1941," G. N. Flerov (Joint Institute for Nuclear Research-USSR)

Some material pertaining to research aimed at clarifying the possibility of inducing the chain reaction was presented. Both theoretical and experimental studies were carried out to test different methods of producing the reaction and to analyze its kinetics. The high-sensitivity technique designed for detecting uranium fission permitted the discovery of the spontaneous fission of uranium and the later searches for the spontaneous fission of thorium. Subsequently this made it possible to assess the role of spontaneously fissioning nuclides in the occurrence of the uncontrolled chain reaction.

9. "The Early Japanese Program," Paul Kuroda (University of Nevada, Las Vegas)

Aston and Bohr visited and lectured in Japan in 1936 and 1937, respectively. Their visits to Japan shortly before WWII had a profound effect on Japanese scientists. The early Japanese program was led by Yoshio Nishina at the Institute of Physical and Chemical Research and Kenjiro Kimura at the Tokyo Imperial University during the period between 1938 and 1942. This work resulted in the discoveries of symmetric fission and a member of the "missing" radioactive family 4n+1, ^{237}U. In addition to the research carried out by the Nishina group, a physicist at the Kyoto Imperial University named Tokutaro Hagiwara proposed, as early as in May 1941, an extremely interesting idea of using a nuclear chain reaction to initiate a thermonuclear reaction.

10. "Reminiscences of Berlin and Chalk River," Leslie G. Cook (ERE-retired)

Personal reminiscences from the Kaiser Wilhelm Institute in Berlin, 1937-1938 were given. The course of experimental events and the continuing search for interpretations that would stand up, the guiding influence of the work of Irene Curie and Pavle Savič on the experimental work of Hahn and Strassmann, the influence and effects of Nazi political pressures within the Institute, the shadows of war, and how uranium fission finally got itself discovered were explained. The talk continued with personal reminiscences from the Nuclear project in Canada.

11. "Spontaneous Fission of the Heaviest Elements," Darleane C. Hoffmann (Lawrence Berkeley Laboratory, California)

Although spontaneous fission was discovered in ^{238}U by Petrzhak and Flerov in 1940, detailed studies of the process were first made possible in the 1960s with the availability of milligram quantities of ^{252}Cf. However, until 1971 it was believed that the main features of the mass and kinetic-energy distributions were essentially the same as those for thermal neutron-induced fission and that all low-energy fission proceeded via asymmetric mass division with total kinetic energies which could be derived by linear extrapolation from those of lighter elements. Measurements for the heavier elements have shown symmetric mass distributions with both high and low total kinetic energies. Recent results for spontaneous fission properties of the heaviest elements were reviewed and compared with theory.

12. "Nuclear Fission and the Transuranium Elements," Glenn T. Seaborg (Lawrence Berkeley Laboratory, California)

Neutron irradiation of uranium in the 1930s led to the reported discovery of such transuranium elements as ekarhenium, etc. After a few years of investigation these were correctly identified as fission products. Not until it was recognized that the first four of the real transuranium elements should be part of a 14-member "actinide series" could elements 95 and 96 (americium and curium) be chemically identified following their nuclear synthesis. This new view was the key to the synthesis and identification of the next seven transuranium elements, resulting in the completion of the "actinide series" at element 103 in 1961. The "transactinide elements" could, according to the actinide concept, also be correctly placed in the Periodic Table and the chemical properties of the first transactinide elements, elements 104 and 105 (rutherfordium and hahnium), confirm this point of view. Transuranium elements through atomic number 109 are now known. There are possibilities for the discovery of more transactinide elements.

13. "Fission Technology: Retrospect and Prospect," Alvin M. Weinberg (Institute for Energy Analysis)

Forty-five years have passed since the "New Piles Committee" at Chicago first deliberated on the future of nuclear power. The light water reactor, first conceived at that time, now dominates the world's fleet of 500-odd civilian power reactors. Though nuclear power now accounts for almost 8% of the world's primary energy, its future hangs in doubt in many countries because of the public's apprehensions over reactor safety and waste disposal. Yet the greenhouse threat adds great urgency to widespread deployment of nuclear power. Nuclear technologists are therefore challenged to develop nuclear systems that are fully acceptable to a skeptical public.

14. "The Future of Nuclear Reactors," Edward Teller (Hoover Institute and Lawrence Livermore National Laboratory)

Measures to assure the safety of nuclear reactors in the United States began more than 40 years ago, and they have produced an unrivaled record of safe energy generation. While many nations are rapidly converting to nuclear-generated electricity, usage is declining in the United States, where misplaced public concerns and their political consequences have produced counterproductive and economically harmful regulations. Three innovative reactor designs—the modular high-temperature gas reactor, the PIUS, and the Geyser—offer smaller size and increased simplicity. Those factors, possibly combined with underground location, would provide unmistakable and convincing evidence of safety and could lead to replacing current requirements with practical, safety-enhancing regulations.

15. "Radioiodine: The Atomic Cocktail," Rosalyn S. Yalow (Veterans Administration Medical Center)

The use of artificial radionuclides in medicine has continued to increase in importance resulting in the growth of a new medical specialty, Nuclear Medicine. The availability of very low cost radionuclides from Oak Ridge, beginning in 1946, initiated a revolution that led to widespread use of ^{131}I in the understanding and management of thyroid disease and to extensive use of ^{131}I-labeled dyes, fats, drugs, proteins, and other substances in diverse areas of medicine. While the role of the "atomic cocktail" in cancer therapy has diminished greatly, *in vivo* and *in vitro* radionuclide procedures in medical diagnosis are employed in over one-third of hospital admissions.

7. Publication

The proceedings of the conference were published in a two-volume set under the title:

FIFTY YEARS WITH NUCLEAR FISSION
Edited by James W. Behrens and Allan D. Carlson
Published by the American Nuclear Society, Inc.
La Grange Park, IL 50525

Also available from the publisher are video tapes of the presentations of the invited speakers as well as the talks of the banquet speakers.

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Conference Reports

TWELFTH NATIONAL COMPUTER SECURITY CONFERENCE Baltimore, MD October 10–13, 1989

Report prepared by

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The National Computer Systems Laboratory (NCSL) of the National Institute of Standards and Technology (NIST) and the National Computer Security Center (NCSC) of the Department of Defense (DoD) co-sponsored the Twelfth National Computer Security Conference held in Baltimore, Maryland on October 10-13, 1989. The theme of the conference, "Information Systems Security: Solutions for Today—Concepts for Tomorrow," highlighted the broader focus of information systems security which now challenges the user community, vendors, system developers, and administrators. Major areas addressed included advanced research developments and emerging technologies, network security architectures, risk management, management and administration issues in computer security, and an expanded focus on education and ethics. More than 2,000 attendees from government, industry, and academia participated in the 4-day conference, which was co-chaired by NCSL's Irene Gilbert and NCSC's George Mitchell.

1. History of the Conference

The National Computer Systems Laboratory has played a vital role in protecting the security and integrity of information in government computer systems through its Computer Security Program since the passage of the Brooks Act in 1965. Since 1972, the Laboratory has issued standards and guidelines on the cost-effective protection of unclassified information in computer systems. NCSL works with organizations from government, industry, academia, and voluntary standards groups to develop standards and guidance, produce tests to evaluate conformance to standards, transfer technology to users, and provide technical assistance to government and industry in computer security applications. The Computer Security Act of 1987 strengthened NCSL's role in protecting unclassified sensitive information in federal computer systems.

The Department of Defense also has pursued an active computer security program over many years. In 1978, the Assistant Secretary of Defense for Communications, Command, Control and Intelligence established a Computer Security Initiative to ensure the widespread availability of trusted ADP systems within the DoD community. In 1981, the National Computer Security Center was created to administer the activities of the initiative. NCSC advances the development of trusted computer systems and publishes guidelines on computer security. In response to the increased emphasis on computer security in recent years, NCSC expanded its efforts to support basic research for the development of additional trusted systems. NCSC's technology transfer program ensures that other federal agencies and industry benefit from technological advances in computer security.

Sharing common goals and mutual challenges, NCSL and NCSC joined forces in 1979 to co-sponsor the first National Computer Security Conference. Dr. Dennis Branstad, NCSL (now a NIST Fellow) and Mr. Stephen Walker, then Chairman, Computer Security Technical Consortium, organized the first seminar as an information exchange on computer security issues. Since its inception, the meeting has expanded significantly in size and scope to become a large, comprehensive computer security conference. This development parallels an increased national awareness of the need for computer systems security and an expanded interest in existing and emerging technologies available to protect vital information resources.

2. The Twelfth National Computer Security Conference

The Twelfth National Computer Security Conference was organized into four tracks: research and development; systems; management and administration; and education and ethics. Of particular interest was the expanded focus on computer security education and awareness resulting from requirements of the Computer Security Act of 1987 and the in-depth concentration on computer security ethics in the workplace. The first-day "Overture" sessions offered an introduction to basic computer security subjects including an overview of agency security plans submitted in response to the Computer Security Act, ethical conflicts in computer science, NCSL/National Security Agency (NSA) joint efforts, the Secure Data Network System, DoD Trusted Computer System Evaluation Criteria, and training guidelines.

The second day of the conference opened with welcoming remarks from NCSC Director Patrick R. Gallagher and NCSL Director James H. Burrows. Representative Tim Valentine of North Carolina presented the keynote address on the role of Congress in computer security. Valentine cited the progress of federal agencies in implementing the Computer Security Act and called for expediting federal progress in computer security technology and standards, including international standards. An opening plenary on "Information Systems Security—A Year in Review," followed. Participants included James Burrows, NCSL; Patrick Gallagher, NCSC; Clive Blatchford, ICL, United Kingdom; Steve Kent, Bolt, Baranek, and Newman; Stephen Walker, Trusted Information Systems; Stuart Katzke, NCSL; Eliot Sohmer, NCSC; and Harold Segal, Office of Personnel Management. Among the subjects addressed were legislation and policy; the international standardization effort; electronic data interchange; trusted systems, virus response centers; international trusted criteria; and computer security training and awareness.

Following this overview, speakers from government and industry presented concurrent sessions in the four tracks described above. Selected presentations are summarized below.

2.1 Track A: Research and Development

2.1.1 Database Management Achieving multilevel database security was the focus of the database management session chaired by Teresa F. Lunt of SRI International. Gary W. Smith of George Mason University presented a paper describing a balanced approach which uses good system design, management controls, and procedural security as well as technical solutions to achieve multilevel database security.

Tim Wood of Sybase, Inc. gave an overview of the system architecture of the Sybase Trusted SQL Server, targeted at the B2 level of trust. The Trusted SQL Server is a physical machine control program that is a hybrid of a secure, high-performance DBMS server with a dedicated kernel of original design. A third paper by R. Alan Whitehurst, University of Illinois at Urbana-Champaign, and Teresa F. Lunt, SRI International, discussed the verification of the SeaView formal top-level specifications and the benefits gained from formally specifying and verifying selected database operations. The SeaView project was a 3-year program to create the design of a multilevel secure relational database system that met the criteria for trusted system Class A1.

2.1.2 Verification Methodology Joshua Guttman, MITRE Corporation, chaired this session. Carla Marceau and C. Douglas Harper, Odyssey Research Associates, presented a paper describing an interactive approach to Ada verification. Penelope is a prototype Ada verification editor whose user interactively and concurrently develops specifications of programs, their Ada text, and proofs of their verification conditions. Adding CASE technologies to formal verification was the subject of the next paper by J. V. A. Janeri, J. S. Barlas, and L. L. Chang of the MITRE Corporation. The authors used CASE technology to further automate the labor-intensive task of formal verification, by integrating the process of formal software design verification with the software engineering life cycle. The result is a Specification Browser which serves as a verification aid. In the final paper of this series, Timothy E. Levin, Steven J. Padilla, and Roger R. Schell, Gemini Computers, Inc., presented engineering results from the trusted system level A1 formal verification process. The authors reviewed the formal methodology used to verify the security of the GEMSOS TCB, currently under development and targeted for the TCSEC Class A1 level.

2.1.3 Verification Marvin Schaefer, Trusted Information Systems, chaired the verification session. Barbara A. Mayer and Monica M. McGill, NCSC, presented an overview and rationale of recently published guidelines for formal verification systems. The paper described the history and status of the guidelines, the endorsement process, the evaluation approach, and possible future directions for verification systems.

A second paper by William D. Young, Computational Logic, Inc., compared and contrasted the Gypsy and Z specification languages. The authors suggested refinements to the two languages and pointed a direction for future language designs. The final paper in this group evaluated security model rule bases; John Page, Jody Heaney, Marc Adkins, and Gary Dolsen, Planning Research Corporation, are the authors of the paper. The evaluation viewed three different security models from the common point of reference provided by the Security Model Development Environment prototype.

2.1.4 Models The session on models was chaired by D. Elliot Bell, Trusted Information Systems. In the first presentation, Robert S. Lubarsky, Franklin and Marshall College, described a mathematical approach to hook-up security and generalized restrictiveness. A second presentation explained the Argus computer security model.

2.1.5 Security Architecture NCSL's Lisa Carnahan and NCSC's Mario Tinto chaired these sessions. Papers focused on a broad range of products and systems: the design of trusted workstations using a total "information security" (INFOSEC) solution; Formal Top Level Specification (FTLS) security testing for the Honeywell LOCK project; the formal specification of security aspects of a messaging system architecture; a Secure Distributed Operating System (SDOS) prototype; a high B level security architecture for the IBM ES/3090 Processor Resource Systems Manager; and a TRW security engineering effort to define an architecture for a MLS communications processor. An R&D panel and an ethics plenary session concluded the track.

2.2 Track B: Systems

Network security architectures received indepth coverage, focusing on privacy and access control issues. Recent incidents involving malicious code have drawn increased attention to the need for prevention and remedies in this area.

2.2.1 EMail and Authentication Ruth Nelson, GTE, chaired this session. John Linn, DEC, and Stephen Kent, BBN Communications Corporation, presented a paper on privacy for DARPA-Internet mail. The facilities discussed provide privacy enhancements on an end-to-end basis between originator and recipient User Agent processes, which may be implemented on heterogeneous systems. The authors defined and recommended a cryptographic key management approach employing RSA-based public-key certificates.

Key management and access control for an electronic mail system was the subject of the next paper by Martha Branstad, W. Curtis Barker, Pamela Cochrane, and David Balenson of Trusted Information Systems, Inc. The authors examined key management and access control services associated with the Embedded Network Security (ENS) Trusted Mail system, indicating how both encryption and trusted system functionality provide protection. Miles Smid, James Dray, and Robert Warnar, NCSL, described a token-based access control system for computer networks. In this system, a user's access is mediated by a smart token implementing a transparent cryptographic three-way handshake with the target computer.

2.2.2 Local Area Networks The increasing use of local area networks (LANs) has driven the search for cost-effective security solutions. Dennis Branstad, NCSL, chaired the session on LANs. Gary Stoneburner and Dean Snow, Boeing Aerospace and Electronics, described how and why the Boeing Multilevel Secure local area network (MLS LAN) is migrating towards an Information Security (INFOSEC) solution. Significant design issues were presented, as well as an overview of how encryption might be embedded into the MLS LAN. L. Kirk Barker, Datotek, and Kimberly Kirkpatrick, MITRE, next described the

Standard for Interoperable LAN Security (SILS) model which would provide a standard protocol for protecting LAN traffic; the IEEE 802.10 is basing its security protocols and services for LANs on this model. Peter Loscocco, NCSC, presented the last paper in this series on a dynamic network labeling scheme for a MLS LAN.

2.2.3 Networks Donna Dodson, NCSL, chaired the networks session addressing access control; Dennis Grayson, NCSC, chaired the second networks session. Extending mandatory access controls to a networked multilevel secure (MLS) environment was the subject of the first presentation by Ron Arbo, Eric Johnson, and Ron Sharp, AT&T Bell Laboratories. They introduced a software package design that permits MLS systems to securely communicate without modifying or trusting the existing network applications. Richard Graubart, MITRE, reexamined the traditional access control policies and proposed a new type of access control policy. Four DEC researchers next described a digital distributed system security architecture. Other papers in this series outlined guidelines for specifying security guards and the security of embedded tactical systems.

2.2.4 Computer Viruses and Related Threats Protecting information systems from threats of all kinds was the focus of these sessions; Jack Holleran, NCSC, and James Anderson, J. P. Anderson Co., were session chairs. Martha Brothers, AT&T, gave a "how to" guide for virus protection in MS-DOS, while Ronald Tencati, Goddard Space Flight Center, and Patricia Sisson, Science Applications Research, described the "Father Christmas Worm" of December 1988 which invaded a large DECnet network and reached 6,000 computer nodes worldwide. Cliff Stoll, Harvard—Smithsonian Center for Astrophysics, gave an epidemiology of viruses and network worms. Other talks summarized computer crime techniques and gave potential solutions.

The systems track concluded with a discussion of vendor activities by Dennis Sirbaugh, NCSC, and a plenary session on ethics.

2.3 Track C: Management and Administration

2.3.1 Management Irene Gilbert was session chair for management. William Norvell, Hughes Aircraft Company, gave the first presentation on integrating accreditation activities into the acquisition process, ensuring that all security requirements are specified in the functional baseline for design and test. David Juitt, Digital Equipment Corporation (DEC), proposed a security approach through system management; he detailed an ongoing advanced development effort within DEC to study security issues of computing across a worldwide distributed environment and how they relate to conducting business safely. A third paper described a systematic approach to software security evaluations.

2.3.2 Accreditation In this session chaired by Grant Wagner, NCSC, the first presentation by Toni Fish, Information Systems Security Association, and Corey Schou, Idaho State University, addressed the issue of the certification of computer security professionals. Darryl Song, MEI Associates, spoke on the accreditation of information systems and networks, followed by a talk by Horace Peele, Electronic Security Command (ESC), USAF, on the development of the ESC Accreditation Package. Peele recommended automated accreditation packages as effective security tools throughout the Department of Defense and the federal government.

2.3.3 Risk Management These sessions were led by Sylvan Pinsky, NCSC, and Irene Gilbert, NCSL. Jennie Stevens and Richard Weiner, Booz, Allen & Hamilton, Inc., presented an innovative concept for computer security risk assessment developed in 1988 and 1989 by their company. The concept provides a framework upon which an individual organization's customized guideline can be built. Next, Suzanne Smith, Los Alamos National Laboratory, introduced LAVA, the Los Alamos Vulnerability/Risk Assessment system, a three-part systemic approach to risk management that can be used to model a variety of application systems. A third paper focused on the purpose and framework of anomaly detection; G. Liepins, Oak Ridge National Laboratory, and H. Vaccaro, Los Alamos National Laboratory, placed anomaly detection of computer use in the framework of overall computer security. NCSL's Stuart Katzke gave an update of progress in the federal government's risk management activities.

The remainder of Track C dealt with Air Force customer programs, a "speak out" session for the informal exchange of ideas and opinions, an outline of NCSL programs by Miles Smid, and a concluding plenary session on ethics.

2.4 Track D: Education and Ethics

This new track on computer security education and ethics attracted much interest.

2.4.1 Ethics Overview Charles Pfleeger, Trusted Information Systems, chaired the overview session on ethics. In the first presentation, Larry Martin, Subcommittee on Automated Information Systems Security, addressed the issue of responsibility for unethical computer behavior. Martin concluded that while the ultimate responsibility to behave in an acceptable manner belongs to the user, all of us share in the responsibility for and the consequences of unethical computer use. Glenn D. Watt, NCSC, spoke next on the ethical dilemma of malicious code; his solution combines technology and ethics. A panel discussion followed.

2.4.2 Management Responsibility vs. Individual Rights Management responsibility versus individual rights was the focus of the next session, chaired by Lance Hoffman, George Washington University. The first speaker was Robert Veeder, Office of Management and Budget, whose topic was the Computer Matching and Privacy Protection Act of 1988. Anna Patrick, U.S. Department of Agriculture, spoke next on public access to government databases. A panel discussion followed on the role of management versus the prerogatives of individuals; panel members included session speakers plus Brian Hyland, U.S. Department of Labor; Jan Goldman, ACLU; and Marc Rotenberg, Computer Professionals for Social Responsibility.

2.4.3 Criminalization of Computer Abuse/Misuse William Murray, Ernst & Whinney, chaired the next session on the criminalization of computer abuse and misuse. Jay BloomBecker, National Institute on Computer Crime Data, discussed trends in computer misuse, followed by James Miller, University of Southern Mississippi, who gave an academic perspective on computer abuse. A prosecutor's point of view was presented by William Cook, U.S. Attorney's Office; he successfully prosecuted the first case under the Computer Fraud and Abuse Act of 1986. A panel discussion concluded the session.

2.4.4 Ethics in the Workplace Who is responsible for ethics in the workplace? James P. Anderson, James P. Anderson Co., led the session addressing this issue. Karen Forcht, James Madison University, presented a talk on the ethical use of computers, followed by a panel discussion.

2.4.5 Education, Training and Awareness At this point, the focus of Track D shifted to education, training, and awareness issues in computer security. Lauresa Stillwell, Department of State, chaired the session. W. V. Maconachy, NSA, discussed turning a philosophical orientation of computer security education into a practical reality. He challenged government personnel to develop and implement a well-orchestrated government-wide information systems security awareness, training, and education model. A panel then compared and contrasted the education, training, and awareness continuum, followed by a talk by John Higgins, Brigham Young University, on training computer science undergraduates in information security.

Harold Segal, Office of Personnel Management (OPM), led the session on computer security training in the federal government. Following his talk on OPM training modules, Anne Todd, NCSL, presented the training guidelines which she coauthored. A federal training panel concluded the session.

The computer security awareness session was chaired by Harry DeMaio, Deloitte, Haskins & Sells. His talk on employee awareness was followed by one on executive awareness by Joan Foreman, Bureau of Engraving and Printing. An executive awareness panel preceded the closing talk on mandated versus voluntary ethics given by Marlene Campbell, Murray State University (Kentucky).

3. Summary of the Conference

The Twelfth National Computer Security Conference covered a broad range of issues and emerging technologies of value to those charged with the responsibility of protecting vital information resources in computer systems. "Information Systems Security: Solutions for Today—Concepts for Tomorrow" offered guidance for the present and planning strategies for the future. In the rapidly changing world of information systems technology, the importance of planning for the security of tomorrow's information systems is critical to all who understand that an organization's information is its most valuable asset.

Conference proceedings are available from conference co-chair Irene Gilbert, National Computer Systems Laboratory, A216 Technology Building, National Institute of Standards and Technology, Gaithersburg, MD 20899, or you may call (301) 975-3360.

Conference Reports

SECOND INTERNATIONAL CONFERENCE ON CHEMICAL KINETICS Gaithersburg, MD July 24–27, 1989

Report prepared by

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The conference, chaired by John T. Herron of the Chemical Kinetics Division of NIST, was attended by 240 scientists from 14 countries representing academic, government and private sector institutions.

The conference was sponsored by the National Institute of Standards and Technology, the National Science Foundation, the National Aeronautics and Space Administration, the Environmental Protection Agency, the Gas Research Institute, the Petroleum Research Fund administered by the American Chemical Society, the Ford Motor Company, and the Exxon Research and Engineering Company. The range of the sponsors is indicative of the broad range of interests and applications which were the subjects of the conference.

1. The Conference

The conference followed the tradition of previous conferences held at the National Institute of Standards and Technology (formerly National Bureau of Standards) on important issues in chemical kinetics. The focus of these conferences has been on the theory and practice of chemical kinetics with emphasis on its application to important practical and societal problems of national and international scope such as photochemical smog, the antarctic ozone hole, and the use of plasmas to fabricate electronic devices. Thus, this year's conference had as its subtitle, "Application of Fundamental Data." That theme was developed in a series of seven conference sessions each focusing on a different aspect of chemical kinetics and its application.

The format of the conference was designed to encourage the broadest possible participation of the kinetics community, while at the same time allowing for a detailed discussion of both oral and poster presentations.

2. Introductory Session

The first session was devoted to a series of invited lectures on selected topics in basic and applied chemical kinetics. It was chaired by S. H. Bauer of Cornell University. The speakers and their topics were: S. R. Leone, National Institute of Standards and Technology, "Time-Resolved FTIR Emission Studies of Photochemical Kinetics"; P. Gray, Gonville and Caius College, Cambridge University, "Modeling Complex Behavior by Simple Chemical Schemes"; D. G. Truhlar, University of Minnesota, "Recent Advances in the

Calculation of Bimolecular Reaction Dynamics"; S. E. Stein, National Institute of Standards and Technology, "Chemical Databases for Kinetics." This session illustrated the great advances made in theory and experiment in the past decade, the complexity of chemical systems now under study, and the promise of new means to compile and distribute chemical kinetic data.

3. Topical Sessions

Six sessions followed, each opening with three invited talks followed by a poster session and an extended discussion period devoted to all aspects of the materials presented at the session. The discussion periods were led by the session chairmen with the invited speakers playing a major contributory role. The session titles, chairmen, and speakers were:

Session 2, Environmental Chemistry—The Stratosphere

Chairman, M. J. Kurylo, National Institute of Standards and Technology; F.S. Rowland, University of California Irvine, "The Chemistry of Halocarbons and Hydrocarbons in the Atmosphere"; J. G. Anderson, Harvard University, "Free Radicals in the Antarctic and Arctic Vortices: Ozone Destruction in the Polar Stratosphere"; M. Tolbert, SRI International, "Laboratory Studies of Heterogeneous Processes in the Stratosphere." This session provided a remarkable overview of the current status of our knowledge of the consequences of introducing halogen containing substances into the atmosphere. Laboratory studies are now beginning to provide a basis for understanding and ultimately predicting the anomalous ozone levels observed in the antarctic and arctic atmospheres. The recognition that heterogeneous chemistry plays a major role has been supported by recent experimental work.

Session 3, Environmental Chemistry—The Troposphere

Chairman, D. M. Golden, SRI International; R. A. Cox, Harwell Laboratory, "Laboratory Studies of Peroxy Radical Reactions of Importance for Tropospheric Chemistry"; H. Niki, York University, "Fourier Transform Infrared Spectroscopic Studies of Atmospheric Reactions involving Hydrocarbons"; W. L. Chameides, Georgia Institute of Technology, "Urban Photochemical Smog: The Problems, Uncertainties, and Possible Solutions." As this session illustrated, there remain many fundamental unresolved problems in tropospheric chemistry with very broad societal implications. The chemical kinetics mechanisms are still not well understood, nor is the role of natural hydrocarbon sources. The basis of current control strategies was also challenged.

Session 4, Ions, Clusters and Interfaces

Chairman, P. Davidovits, Boston College; A. W. Castleman, Jr., Pennsylvania State University, "Cluster Reactions"; C. E. Kolb, Aerodyne Research, Inc., "Sticking of Gaseous Molecules on Liquid Droplets—Applications to Atmospheric Acid Deposition and Ozone Depletion." This session further emphasized the recognition that interfacial chemistry plays a crucial role in many aspects of atmospheric chemistry, ranging from the depletion of ozone in the antarctic to tropospheric processes of acid deposition. The advances in measurement science are very apparent in these areas.

Session 5, Plasma Chemistry

Chairman, G. Hancock, Oxford University; D. L. Flamm, University of California and AT&T Bell Laboratories, "Kinetics of Silicon Etching in Fluorine and Chlorine Containing Plasmas"; J. W. Hudgens, National Institute of Standards and Technology, "The Observation of Silicon Centered Free Radicals Using Multiphoton Ionization Spectroscopy"; J. M. Jasinski, The Thomas J. Watson Research Center, IBM, "Silane Plasma Chemistry One Step at a Time: Silicon Hydride Radical-Molecule Kinetics." Plasma chemistry is now entering a new era in which modeling of complex processes on the basis of elementary chemical reactions is being actively pursued. This session emphasized the range of plasma chemistry under study, some of the highly specific diagnostic approaches being developed, and basic chemical kinetic measurement and modeling activities being carried out. The potential for advances in this previously unexplored area of chemical kinetics promises to be of great practical importance and almost certainly will provide new insights into fundamental aspects of chemical kinetics.

Session 6, Oxidation and Combustion

Chairman, A. Lifshitz, Hebrew University; J. Wolfrum, University of Heidelberg, "Elementary Chemical Reactions and Their Interaction with Transport Processes: Experiments and Models"; F. L. Dryer, Princeton University, "Chemical Kinetic Modeling for Combustion Processes"; A. M. Dean, Exxon Research and Engineering

Company, "The Role of Chemical Activation in Combustion Chemistry." Chemical kinetic modeling plays a dominant role in the elucidation of combustion chemistry. In this session the connection was strongly drawn between measurement of the kinetics of elementary reactions at the most elementary level, and processes occurring in practical combustion devices.

Session 7, Propellants and High Energy Chemistry

Chairman, Th. Just, DFVLR; C. F. Melius, Sandia National Laboratories, "The Chemistry of Nitramine Propellant Combustion"; M. C. Lin, Emory University, "Elementary Processes of Relevance to Nitroalkane, Nitramine and Hydrocarbon Combustion Reactions," J. Troe, University of Gottingen, "From Laser Photolysis Experiments to Thermal Dissociation Rates." A theoretical basis now exists for the general chemical processes relevant to propellent chemistry. This session outlined that basic conceptual approach, and showed how new laboratory programs are providing the chemical kinetic data base required to corroborate, modify, or extend our knowledge of propellent and high energy chemistry.

4. Summary

The emphasis on application meant that the main focus of the conference was on the measurement, estimation, and evaluation of thermal rate constants.The nature of the topics chosen for the technical sessions illustrates the direct contribution that fundamental chemical data can make to the solution of important national issues. In the case of stratospheric chemistry this is particularly well understood and chemical kinetic data are used with great effect. This successful area of application serves to illustrate the enormous potential for applying fundamental data to other practical problems. The tenor of the invited and contributed papers, which provide a catalogue of needs and a demonstration of capabilities, clearly indicates that other areas are ripe for such an organized approach to scientific problem solving. Furthermore, the fundamental nature of chemical kinetics makes advances in one subfield, whether in instrumentation, method development, or theory, immediately applicable to other problem areas.

Looking back at the conference held 4 years ago, it is apparent that there has been a great deal of progress. Nevertheless, as capabilities have improved, so have the needs for reliable kinetic data. There is a sense that many opportunities are not being fully exploited. Part of the problem is undoubtedly due to the quantity of data needed to simulate accurately a complex system. Although advances and applications in theory have made notable contributions during the past 4 years, the need for high quality experimental data will continue to dominate modeling efforts. The experience gained from the stratospheric ozone problem clearly indicates how such measurements contribute not only to the problem at hand, but also to all aspects of usable chemical kinetics. Certainly some of the most exciting developments in chemical kinetics in recent years have sprung from such studies, and in the next four years, we expect to see great advances in the understanding and control of other complex systems in which chemical kinetics plays a controlling role.

Copies of the meeting abstract book are available from the conference chairman. In addition, a special issue of the Journal of Physical Chemistry will be devoted to the conference proceedings.

News Briefs

General Developments

MAKING SOUND BUSINESS DECISIONS ON AUTOMATION

AutoMan, a microcomputer program developed at NIST, applies modern mathematical techniques to make factory automation decisions easier. Choosing whether or not to invest in automated manufacturing equipment is a complex problem for shop and factory owners. There are established criteria for making capital investment decisions, but more and more managers are questioning whether these measures alone are adequate for evaluating automation options. AutoMan, developed for the U.S. Navy's Manufacturing Technology Program, allows managers to account for important gains realized by automation, including engineering performance improvements, better control over manufacturing, and improved product quality, as well as harder-to-handle intangible benefits like technology advancement, plant modernization, flexibility, and competitive position. The program for MS-DOS-based machines is available from the National Technical Information Service, Springfield, VA 22161 for $40 plus $3 for shipping and handling. Orders may be placed by phone: (703) 487-4650. Order by PB389-221741. Please specify diskette size 5.25″ or 3.5″.

SOFT-DRINK GLASS BOTTLE STANDARD REVISED

NIST has published a replacement voluntary product standard on glass bottles for carbonated soft drinks. The standard, which supersedes PS73-77, is for manufacturing requirements to improve the safety performance of refillable and nonrefillable glass bottles made from soda-lime-silica glass with nominal capacities up to 36 fluid ounces. It was sponsored by the Glass Packaging Institute Inc., and developed by a standing committee of representatives from soft-drink bottle manufacturers, bottlers, consumers, and others interested in performance requirements, inspection, and testing procedures for soft-drink bottles. NIST, a nonregulatory agency, administers the U.S. Dept. of Commerce Voluntary Products Standards Program in support of private sector standards development. Specific activities are initiated in response to requests from interested parties and subsequent approval by the Secretary of Commerce. To obtain a copy of Voluntary Product Standard PS 73-89, Glass Bottles for Carbonated Soft Drinks, send a self-addressed mailing label to the Office of Standards Management, NIST, A625 Administration Building, Gaithersburg, MD 20899; telephone: 301/975-4023.

EFFECTIVELY DESIGNING COMPUTERIZED WORK STATIONS

The computerized work station often is considered the key to increased office productivity. Yet, work station furnishings and layout seldom are given the same attention and resources as the technology. Instead, space and furniture decisions too often are based on "status" and tradition. To help managers make intelligent choices, researchers at NIST have developed a process for designing work stations based on office activities including reading and writing, talking on the telephone, drafting and drawing, or using a video display terminal. Work station dimensions and configurations then are developed depending on the importance of the activity and the time spent on it. A publication describing this process, Guideline for Work Station Design (NISTIR 89-4163), is available by sending a self-addressed mailing label to Arthur Rubin, NIST, A309 Building Research Building, Gaithersburg, MD 20899. This report is the latest in a series prepared by NIST for the U.S. General Services Administration.

FLUID PROPERTIES OF LENNARD-JONES MIXTURES

The design of chemical process unit operations equipment requires detailed knowledge of the thermophysical properties of fluids and mixtures, often based on mathematical models of their properties and behavior. Many emerging technologies, such as enhanced oil recovery and biotechnology, typically involve materials and fluids not well represented in the current database or which are used in areas of fluid behavior difficult or impossible to study in the laboratory. Computerized models of these systems often benefit from comparison to a model of an idealized mixture. NIST has simulated an idealized mixture (one that obeys the Lennard-Jones intermolecular potential function and has a size ratio of 2:1) over a broad range of temperatures, composition, and energy ratios, and has published the data to be used as a point of comparison for similar real-fluid models. A report of this work, Properties of Lennard-Jones Mixtures at Various Temperatures and Energy Ratios with a Size Ratio of Two (TN 1331), is available from the Superintendent of Documents, U.S. Government Printing Office, Washington, DC 20402-9325. Order by stock no. 003-003-02952-1 for $20 prepaid. A computer listing of the data is available on DOS diskette; contact Dr. Marcia Huber, NIST, Div. 584.03, Boulder, CO 80303; telephone: 303/497-5252.

RESIDUAL STRESS MEASUREMENT AND CONTROL DURING FABRICATION OF CERAMIC COMPOSITES

The magnitude of stresses which can inhibit the sintering of ceramic matrix composites can be assessed using a model composite system of unidirectionally oriented fibers. The stress is measured by means of a sandwich compact consisting of a layer of SiC fibers sandwiched between two layers of ceramic powder of different thicknesses. Upon sintering, this configuration produces an asymmetric stress field across the thickness of the specimen, resulting in bending of the compact, which can be characterized by the curvature of the fibers.

Control of the stresses can be achieved by using a colloidal processing route for coating fibers with alumina or other suitable oxides. Coated fibers with small ($d=0.2$ μm) and large ($d=0.8$-1.0 μm) particle-size alumina powders have been shown to alter significantly the temperature at which stress initiation begins. For uncoated fibers, bending starts at 1300 °C and increases with increasing temperature. Coating with a 4-μm-thick layer of fine particle-size alumina powder lowered the temperature at which the stress initiates to 1265 °C, and a coating thickness of 16 μm lowered the initiation temperature to 1200 °C. In contrast, the coarse particle coating achieved the desired result of delaying stress initiation to higher temperatures. For a coating thickness of 10 μm, bending of the composite was reduced considerably and, most importantly, stress initiation was raised to 1325 °C. The coarse particle-size coatings appear to reduce the stresses by acting as a buffer layer between the matrix and the fiber. These experiments reflect the importance of the fiber/matrix interface with respect to stress enhancement (small particle-size powder) and stress retardation (coarse particle-size powder) during the sintering of fiber-reinforced composites. Even small interfacial layers of few microns considerably influence the stress state during sintering.

CHARACTERIZATION OF THICK-SECTION COMPOSITES

NIST has completed a survey of methods with potential to characterize the through-thickness variation of properties of thick-section composites. The purpose of the survey is to guide industry's response to the Navy's program to manufacture submarine hulls made of polymer composites. Owing to the performance requirements of submarine hulls, the cross-sectional thickness will be much larger than usually encountered in composites manufacture. Methods are therefore needed which can interrogate the interior of thick specimens nondestructively.

Drawing on a wide range of expertise, NIST scientists identified a wide variety of techniques and the technical barriers to their implementation. The level of development required to bring the techniques to fruition was also evaluated. The most promising methods rely on imbedded sensors. These range from miniaturized dielectric probes to fiber optic sensors. The types of sensor best suited for characterization of properties will also depend on whether thermosets of thermoplastics are selected as the polymer resin.

Industry will use the survey to formulate a research plan that encompasses both the manufacture and inspection of thick-section composites.

SMITHSONIAN AND NIST COLLABORATE ON NEUTRON STUDIES OF PAINTINGS

The Smithsonian Institution (SI), through its Conservation Analytical Laboratory, is collaborating with NIST in applying nondestructive neutron autoradiography to the study of works of art. This is the only facility available in the United States for neutron-induced autoradiography (NIA) with a large, uniform, highly moderated thermal neutron field.

In this project, the work of art is activated at the thermal column of the NIST reactor in a flux of 3×10^9 n/cm^2/s for 20 min. It is then placed in contact with film, which is removed and replaced to obtain a series of images created by nuclides with different half-lives. Examples of autoradiographs of paintings by Albert Pinkham Ryder (1847-1917), will be included in a 1990 exhibit of that artist's work at the National Museum of American Art. Until the paintings were studied by autoradiography, it was known only that Ryder had a reputation for continually reworking his paintings and not that he was actually engaged in serious reconsiderations of the paintings' compositions. These changes became apparent in the autoradiographs and are now available for study.

MODEL USED TO CONFIRM NEW EXPERIMENTAL METHOD

The NIST-developed ideal mixing of complex components (IMCC) model has been used by Russian high-temperature researchers to confirm experimental data obtained by their new ion-molecule equilibria technique for measuring low O_2 and alkali partial pressures. The IMCC model includes a large database of Gibbs energy of formation functions for complex liquid oxides, including Al-, Ca-, Fe-, K-, Li-, Mg-, Mn-, Na-, and Si-oxides typically found in glass, steel slags, combustion, and other high-temperature processing industrial applications. The model process provides predictive estimates of detailed solid, liquid, and gas phase compositions.

PROCESS CONTROL SENSOR TECHNOLOGY SURVEYED

NIST scientists recently reported to the National Center for Manufacturing Science (NCMS) on the status of sensor technology for monitoring polymer composites processing. NCMS commissioned the report in order to identify areas in which further research and development would lead to significant advances in sensor technology. The polymer composites industry would benefit greatly from the availability of sensors for process monitoring since many problems with polymer composites can be related to inadequate control during processing.

The report identifies three generic sensor technologies—dielectrics, ultrasonics, and spectroscopy—with the capabilities to monitor polymer composites processing. The report further identifies resin viscosity and degree of cure as the most important properties to be measured. Background information for the report was gathered from literature sources and from review of current work in industrial, university, and government laboratories. Several industrial visits were made to identify process control needs and the current state of implementation of process control sensors.

NCMS, whose members represent a broad range of industries, will use the report to design a research and development program to hasten the availability of sensors for polymer composites manufacture.

URANIUM AND THORIUM ANALYSIS OF POWDERED MILK PRODUCTS

In a NIST study, IAEA (International Atomic Energy Agency, Vienna, Austria), powdered milk and whey samples were assayed for sub-parts-per-billion quantities of uranium and thorium. There is a strong interest in the IAEA powdered milk and whey materials since they were collected shortly after the Chernobyl incident and were known to contain many orders of magnitude higher levels of fission products than pre-Chernobyl dairy products. The uranium isotopic composition was natural and both the thorium and uranium concentrations in these IAEA materials were found to be near ambient levels. The powdered IAEA milk had thorium and uranium levels comparable to the NIST powdered milk (SRM 1549, <500 and 170 parts-per-trillion, respectively), while the concentrations in the powdered IAEA whey (10 and 22 parts-per-billion, respectively) were about two orders of magnitude higher. Studies on locally available fresh milk and on fresh milk collected near the U.S. DOE plant at Fernald, OH, are currently being pursued to determine the distribution of the uranium and thorium between curds and whey.

DISTRIBUTION OF THORIUM DECAY PRODUCTS IN THOROTRAST PATIENT

NIST scientists are collaborating with the research staff at the National Cancer Institute to measure the distribution of short-lived thorium decay products in a variety of organs donated by a former thorotrast patient. This problem is being attacked by measuring the gamma-ray emissions from the radioactive thorium decay products in the donated organs. Thorotrast is colloidal thorium dioxide formerly used to enhance diagnostic medical x-ray images. Patients who were injected with the full dose of thorotrast (equivalent to 12 g of thorium) have suffered a marked increase in the incidence of cancer to the liver, pancreas, larynx, and esophagus. They also have shown excess leukemia and bone marrow failure. The objective of the study is to quantitate the distribution of the short-lived thorium decay products among organs to further refine human health and safety radionuclide biokinetic models and risk coefficients. Actinium-228, ^{224}Ra, ^{212}Pb, ^{212}Bi, and ^{208}Tl were sought among organs which include eye, larynx, esophagus, breast, kidney, liver, spleen, ribs, red blood cells, and blood serum. Of the materials measured at NIST, it is estimated that over 90 percent of the thorium-derived radioactivity is accumulated in the liver, spleen, and bone.

OPTIMUM FLOW CONDITIONING FOR ORIFICE METERS

A common way to measure flow rates of gases and liquids is to insert an orifice plate flowmeter into a length of pipe and measure the pressure drop across the plate. The accuracy of the measurement, however, is dependent on the condition of the flow, which is adjusted using flow conditioners upstream from the meter. European and U.S. specifications for the location of these conditioners differ, and the location has a substantial impact on measurement accuracy. In work sponsored by the Gas Research Institute, NIST analyzed the effect of varying the distance from conditioner to orifice meter. The results showed that, in general, the conditioner should be located 17 pipe diameters upstream from the orifice plate for tube bundle conditioners. More details are presented in Optimum Location of Flow Conditioners in a 4-inch Orifice Meter (TN 1330), available from the Superintendent of Documents, U.S. Government Printing Office, Washington, DC 20402-9325. Order by stock no. 003-003-02961-1 for $4.25 prepaid.

NEW TEST CHAMBER DEVELOPED

Electromagnetic Interference (EMI) has caused a range of problems in military electronic systems—from the inability of pilots to communicate with air-traffic controllers to catastrophic crashes. Researchers at NIST are developing a new type of electomagnetic test chamber to evaluate the ability of operational systems to withstand the effects of radiated EMI. The new chamber combines features of a transverse electromagnetic cell and a broadband reverberating chamber to provide an electromagnetic compatibility test capability for the frequency range 10 kHz to 40 GHz, a frequency range not offered previously by any single facility. A small prototype chamber, which measures $1.3 \times 2.4 \times 3.9$ m, has been constructed by NIST and delivered to another government agency. Initial tests with the chamber have shown the potential for significant cost and time savings. A paper, no. 52-89, describing the chamber is available from Jo Emery, NIST, Div. 104, Boulder, CO 80803; telephone: 303/497-3237.

FRACTURE MECHANICS MODEL FOR WEAR OF ADVANCED CERAMICS

Advanced ceramics materials are being considered increasingly for applications requiring wear resistance, corrosion resistance, and retention of mechanical properties at elevated temperatures. Prediction of wear performance is extremely important for design and selection of ceramic materials for these applications. It has been reported in the literature that the rate of wear suddenly increases by several orders of magnitude as a result of a slight increase in one of the test variables, such as contact load, sliding distance, or sliding speed. A simple contact mechanics model based on linear elastic fracture mechanics was developed to account for this transition. The model predicts that the transition from low to high wear is controlled by crack propagation in the tensile zone at the trailing edge of the contact circle. It also predicts that the wear transition can be prevented by the reduction of either the contact stress or the coefficient of friction, or by increasing the fracture toughness of the ceramic material. Experimental results have been obtained for α-alumina under various test conditions and temperatures ranging from room temperature to 800 °C in confirmation of the model.

NEW MODEL PREDICTS MICROSTRUCTURE DEVELOPMENT IN HIGH-T_c SUPERCONDUCTORS

The compound $Ba_2YCu_3O_7$, the prototype high-T_c material (often denoted the "123" superconductor), contains Cu atoms in "chain" and "plane" sites. Defects such as vacancies and substituents can be introduced in the structure with profound effect on the superconducting properties. The knowledge on a microscopic scale of the atomic configuration of the defects created by metal substitution and/or variation of oxygen concentration is vital not only for understanding the mechanism of superconductivity but also for understanding the evolution of critical microstructures during processing. The latter have a direct effect on mechanical properties;

for example, the density and type of twin boundaries in these materials are directly associated with oxygen content and migration of defects.

NIST neutron diffraction research on high-T_c materials has been extended recently to modeling the motion of vacancies and the configuration of the oxygen atoms and metal impurities in 123-type superconductors. In $Ba_2YCu_3O_{6+x}$ materials, the diffusion of oxygen in the plane of the chains is viewed as a correlated series of jumps of the oxygen atoms into vacant sites in the vicinity of twin boundaries. In $Ba_2YCu_{3-y}M_yO_x$ compounds, the model predicts a rearrangement of the oxygen atoms in order to satisfy the coordination requirements of the substituent metal atoms.

These models qualitatively explain and predict the observed motion and rearrangement of the twin boundaries during diffusion and the symmetry and stoichiometry of the substituted materials. From this, the details of the formation of microstructures which directly affect electronic and mechanical properties may be elucidated. This predictive capability is expected to have a direct bearing on processing strategies to improve materials for technological applications.

BASIC STUDIES OF CHLOROSILANE REACTIONS WITH SURFACES AID UNDERSTANDING OF SILICON FILM GROWTH

NIST and industry scientists have recently observed for the first time how the chlorosilanes $SiCl_4$ and Si_2Cl_6 interact with single-crystal silicon and germanium surfaces. The research, performed primarily at the National Synchrotron Light Source at Brookhaven National Laboratory, and in part at NIST, should further the understanding of how best to use these chemicals as safer alternatives to explosive silane gas during the growth of silicon films for microelectronic applications.

The scientists conducted experiments in ultrahigh vacuum with a variety of surface-sensitive spectroscopies including soft x-ray photoemission and thermal desorption. They discovered that the reactivity of the different chlorosilanes depends on how adsorption occurs on the surface. There is a very low probability that the $SiCl_4$ will stick directly in a single decomposition reaction. In contrast, Si_2Cl_6 first adsorbs in a weakly bound molecular state with near unit probability, and then fragments on the surface. When these surfaces are heated after adsorption, further reactions occur that produce gas-phase $SiCl_2$ and $SiCl_4$, but not Cl_2, indicating that when used as the sole reactant both $SiCl_4$ and Si_2Cl_6 will etch these surfaces. These results explain why an additional reactant that will remove chlorine from the surface is necessary for film growth, and indicate that Si_2Cl_6, with its larger sticking probability, should be the superior source material.

NEW CENTER FOR DENTAL MATERIALS RESEARCH ANNOUNCED

A new Center of Excellence for Materials Science Research has been established to expand the internationally recognized dental and medical materials program at NIST. The center is funded by a 5-year grant from the National Institute of Dental Research (NIDR), one of the National Institutes of Health. The NIDR grant was made to the American Dental Association Health Foundation (ADAHF), which maintains the Paffenbarger Research Center at NIST. Initial research projects will include: developing glass ceramic inserts to strengthen and stabilize composite dental restorations; improving the properties of resin-based composite dental materials to reduce shrinkage; developing protective coatings for both enamel and dentin to prevent caries on exposed roots and to seal restorations; formulating synthetic dentin and bony replacement materials by combining multifunctional monomers with calcium phosphates; and developing new materials that can be readily fabricated into improved shields to protect patients undergoing radiation treatments.

NIST PROPOSES FIPS FOR GOSIP TESTING

NIST is proposing a Federal Information Processing Standard (FIPS) to establish policies, procedures, and criteria for testing networking products for interoperability and conformance with the Government Open Systems Interconnection Profile—GOSIP. After August 1990, federal agencies must use GOSIP (FIPS 146) specifications in procuring networking products. GOSIP defines a set of data communication rules called protocols, which enables computer systems developed by different vendors to communicate, and enables the users of different applications on these systems to exchange information. For further information, contact Jean Philippe Favreau at NIST, telephone: 301/975-3634.

INDEX TO WEIGHTS AND MEASURES PROCEEDINGS REVISED

A new index to the proceedings of the National Conference on Weights and Measures (NCWM) has been published. NCWM is an organization of state, county, and city weights and measures enforcement officials that was established in 1905. NIST, which is a nonregulatory agency, provides technical support to NCWM through its Office of Weights and Measures. NIST Special Publication SP 769, Index to Reports of the National Conference on Weights and Measures from the First to the Seventy-Third (1905-1988), is a subject and speaker index covering the proceedings of the annual meetings of NCWM. SP 769 replaces NBS Special Publication 691, which covered the proceedings from NCWM annual meetings from the first to the sixty-ninth. For a copy of SP 769, send a self-addressed mailing label to the Office of Weights and Measures, NIST, A617 Administration Bldg., Gaithersburg, MD 20899.

TECHNIQUE DEVELOPED TO MEASURE INTERFACE STRENGTH IN POLYMER COMPOSITES

NIST researchers have developed a new technique to estimate the strength of the bonding between polymer matrix resin and reinforcements in polymer composites. Bonding of the resin to the reinforcements is an important engineering property as it strongly influences the mechanical properties of polymer composites. The previously available methods to evaluate the strength of this bond require highly trained personnel, are tedious to perform, and require special test specimens or destruction of the composite. The new test procedure is easy to carry out and correlates well with interfacial strength.

The new method uses a laser to heat a very small, localized region of the sample. The acoustic emission which results from interface debonding is measured. The debonding is a consequence of the widely different thermal expansion of the resin and reinforcement. Since thermal expansion is anisotropic in each ply, the technique also provides a measure of interlaminar bonding in multidirectional laminates. Contacts with industry are being made to seek assistance in adaptation of the new method to industrial test protocols.

ISO WORKING GROUP ON FRACTURE OF HIGH-PRESSURE GAS CYLINDERS

The first meeting of the Working Group on Fracture that was formed under the jurisdiction of International Standards Organization (ISO) Committee TC-58 on Gas Cylinders (Subcommittee SC-3 on Cylinder Design) was held at NIST.

The objective of the working group is to establish test methods and acceptance criteria for high-strength steel cylinders used for transporting compressed gases.

Agreement on a proposed new ISO specification for seamless steel gas cylinders (ISO-DP 9809) was reached at the meeting except for the requirements for tests and acceptance criteria for fracture resistance of cylinders. Significant differences exist between the fracture tests proposed by the U.S. cylinder manufacturers and those proposed by the European cylinder manufacturers.

A test program to evaluate the fracture performance of high-strength steel cylinders was developed by the working group and is to be completed by the working group's next meeting.

SUBMICRON RESOLUTION TWO-DIMENSIONAL X-RAY IMAGING NOW READY FOR MATERIALS SCIENCE STUDIES

An x-ray imaging system with submicron spatial resolution has been developed by the synchrotron radiation group at NIST. Installed at beam line X23A3 of the National Synchrotron Light Source at Brookhaven National Laboratory, the system is used to study the interiors of solids by the techniques of non-destructive x-ray diffraction imaging and microradiography. Important advantages for in-situ imaging accrue from the use of x rays (as opposed to electrons), such as no requirement for vacuum and much less heating of the sample.

The system performance has been evaluated by radiographically imaging palladium lines, as narrow as 0.6 μm, deposited on a silicon wafer. The results give an estimated spatial resolution of 0.5 μm and suggest that the resolution is limited by Fresnel diffraction. Diffraction images of device features and multilayer interfaces have also been observed with this new imaging system. These images are more sensitive to microstructures than microradiography, and are important to detailed understanding of the performance of devices designed from the band-structure engineering approach.

IMPROVED METHOD FOR MEASURING CHEMICAL BONDING ON SURFACES

A knowledge of the geometric structure of a molecule adsorbed on a solid surface is of great importance in understanding the chemistry and reactivity of the molecule with the solid. A technique called ESDIAD (Electron Stimulated Desorption Ion Angular Distributions), developed at NIST in the 1970s, allows for the direct determination of the local bonding geometry of adsorbed molecules. The technique takes advantage of the fact that atomic and molecular fragments created during electron bombardment may leave or desorb from the surface in the same direction as the chemical bond that was ruptured by the electronic excitation. A major limitation of the original apparatus was that only positively charged desorbed ions could be detected.

Recently, NIST scientists modified the ESDIAD technique to allow for the detection of negatively charged ions and electronically excited neutral species. This has opened up a new direction in the study of the structure of adsorbed molecules and provides new insights into how charged particles desorb from surfaces. For example, in experiments on fluorinated molecules adsorbed on surfaces, they found negative ions came primarily from molecularly intact species whereas the positive ions came mainly from dissociated species. The ability to measure this phenomenon offers the opportunity to study the local structure of complex chemical systems by a comparison of negative and positive ion signals.

MULTILAYERS FOR SOFT X-RAY MONOCHROMATORS

Multilayer structures consisting of alternating layers of ultrathin materials can be efficient, selective reflectors in the soft x-ray region. NIST scientists have recently reported one of the first applications of multilayer optics as a replacement for more conventional (and inefficient) soft x-ray monochromators on a synchrotron radiation beamline. For this application, multilayers were used instead of the usual diffraction elements in soft x-ray monochromators—natural crystals and ruled gratings. In the wavelength range of interest, 6-30 Å, gratings are inefficient, and suitable, robust, natural crystals are scarce. Multilayers are a good alternative because they can provide efficient diffraction with moderate resolution under relatively high power loads, such as produced at the latest generation of synchrotron-radiation facilities.

Operational tests have been performed at the NIST beamline at the National Synchrotron Light Source at Brookhaven National Laboratory. For these studies, multilayers made of tungsten and carbon (40 Å per layer-pair) replaced the two natural crystals in the beamline's primary monochromator. High efficiency was obtained for the multilayer diffraction ($\approx$ 6 percent per multilayer), yielding a flux of $\approx 10^{11}$ photons/s in a bandwidth of $E/\Delta E \approx 100$. No degradation of the multilayer structure with beam exposure was evident. The flux and resolution attained permitted new experiments to be done in a photon-energy range previously inaccessible at the NIST facility.

IONOSPHERIC CALIBRATOR FOR MEASUREMENT OF VARIATIONS IN RADIO SIGNAL DELAY

NIST scientists have recently developed a prototype ionospheric calibrator. The calibrator, which uses GPS (Global Positioning System) satellite signals, provides information on propagation delays through the ionosphere. It monitors the propagation delay for signals at 1.2276 and 1.5754 GHz broadcast from the orbiting GPS satellites. When fit to a $1/f^2$ model of propagation variation, the receiver calculates corrections to the delay of the signal. The added signal delay caused by the ionosphere can be more than 100 ns. The calibrator reduces the uncertainty in that delay to a few ns.

The receiver is codeless, that is, it does not require knowledge of the complex codes used by GPS. The receiver can track up to 12 satellites at once, and it produces a value for the ionospheric correction every 15 s. Each value accounts for the total electron content along the line between the satellite and the receiver. Eventually, the GPS constellation will include 21 satellites providing continuous access to four or more satellites at any location on the Earth.

With rising interest in using GPS for accurate surveying and geodesy (observation of motion of land masses), the ionospheric calibrator promises a simple approach to dealing with one of the key ranging uncertainties. This is especially important now that we are near a maximum in solar-radiation-activity level. Time transfer using GPS will also benefit from this advance. The method may also offer a particularly simple means for studying variations in electron content of the ionosphere itself.

NIST RESEARCHER DEVELOPS ROUTING PROTOCOL WITHIN OPEN SYSTEMS INTERCONNECTION (OSI) REFERENCE MODEL

A NIST researcher has developed a model for interdomain or policy-based routing within the OSI reference model for computer communications. Policy-based routing is a key infrastructure element for deploying significant, operational networks based on OSI.

The model provides the necessary structure to address administrative policy issues, such as selective acceptance of transit traffic across a routing domain or the creation of cooperative, private bilateral routing agreements between independent organizations. A technical report describes the model for policy-based routing, which has been adopted by the European Computer Manufacturers Association (ECMA) and is under consideration by the International Standards Organization (ISO).

GUIDELINE FOR QUALITY CONTROL OF IMAGE SCANNERS PUBLISHED

Federal Information Processing Standard (FIPS) 157, Guideline for Quality Control of Image Scanners, announces the adoption of the American National Standard for Information and Image Management—Recommended Practice for Quality Control of Image Scanners, ANSI/AIIM MS44-1988, as a FIPS guideline. MS44 provides procedures and physical test objects for use by document processing system analysts, designers, and operators in calibrating monochrome, digital image scanners.

NIST ANNOUNCES AVAILABILITY OF STANDARD FLAWS FOR EDDY-CURRENT NDE

NIST has announced the availability of Research Material (RM) 8458, a well-characterized artificial flaw used as an artifact standard in eddy-current nondestructive evaluation (NDE). The RM is the outcome of work carried out to address the need for calibration standards for eddy-current NDE, for example, as used to detect fatigue cracks in aircraft structures. The RM flaw is produced in an annealed aluminum alloy (7075) block by first indenting the block and then compressively deforming the resulting notch until it is tightly closed. The next operation is to restore a flat finish to the block face, after which the block is heat treated to the T6 temper. The problem has been to manufacture artificial flaws that closely simulate the eddy-current response of fatigue cracks. Currently used artifacts include electrical-discharge-machined and saw-cut notches, both of which are relatively poor representations of fatigue cracks as their widths are too great. The new method provides notches that can be made controllably in a variety of geometries, have known dimensions, and with widths that are narrow enough to provide an acceptable representation of fatigue cracks.

DECREASED PERFORMANCE FOR NEW REFRIGERANT DOCUMENTED

NIST researchers completed fundamental calorimetric experiments to document a 20 percent decrease in the heat transfer coefficient for a leading new environmentally safe refrigerant while boiling inside a single tube and over the evaporative boiling regime found in present day commercial office building centrifugal chillers. This decrease closely matches an 18 percent decrease in chiller cooling capacity found by leading manufacturers in field tests of the new refrigerant. Consequently, a cooperative research program has been planned with NIST, universities, and industry to understand and enhance the heat transfer during evaporative boiling of this refrigerant for example, NIST will complete nucleate pool and flow boiling visualization studies of this refrigerant on both smooth and special surfaces designed to enhance the heat transfer rates. High-speed photography will be used to understand and document the components of the heat flux at the interface between the solid and fluid surfaces.

NEW MODEL DEVELOPED FOR SORPTION OF FORMALDEHYDE

NIST researchers have developed a new mathematical model that correctly predicts the adsorption and desorption of formaldehyde gas in the presence of gypsum wallboard. Pressed-wood products are a major source of formaldehyde in indoor environments. Current models correctly predict the equilibrium indoor formaldehyde levels from knowledge of the emission rate of the pressed-wood products. However, the formaldehyde concentration takes days to achieve equilibrium because gypsum wallboard acts as a sink for the pollutant. The new model predicts the nonequilibrium concentration in the presence of wallboard. The model was developed on the basis of mass balances in a building and accounts for building air exchange rates and press-wood product and wallboard areas.

NIST MEASURES PERFORMANCE OF THERMAL BRIDGES IN NEW MOBILE TEST FACILITY

NIST completed its first series of measurements on the performance of thermal bridges in a new mobile test facility housed inside a NIST environmental chamber. Thermal bridges are highly conductive paths for energy flow into or out of a building, usually along structural supports that penetrate insulated exterior walls in office buildings.

These heat flow paths can account for as much as 35 percent of the total heat flow through a wall. Most existing computer codes used by building designers do not account for these thermal bridges. The new facility was built to gather basic data under controlled laboratory conditions on the heat flow along such penetrations. The test room measures 6×2.5×2.5 m with removable walls and roof and is mounted on a gooseneck trailer for moving. Heat flow transducers and specially built flat and corner calorimeters are used for measuring heat flow rates. To date, measurements have been made on a metal-framed wall typical of use in industrial buildings. The results indicated that measured heat flow rates were as much as 18 percent larger than those predicted due to the multidimensional heat flow paths that exist in metallic framing members.

NIST DEVELOPS NEW SIMULATION MODEL FOR INDOOR AIR MOTION

NIST has collaborated with researchers in Japan to develop a new computer model for predicting indoor airflow distribution. This model will be used to determine effective ventilation distribution for the design of more efficient manufacturing clean rooms and large building air-conditioning systems. The model called "EXACT" solves a nonlinear system of momentum, energy, and turbulence equations to predict three-dimensional nonisothermal flow in either steady-state or transient conditions. The model handles a variety of boundary conditions including air inflows, air outflows, and heat flux or temperature wall conditions. The model has the ability to handle an arbitrary number of obstacles in the flow region to account for furniture and partitions. The computer code for "EXACT" exists in vectorized and nonvectorized versions.

NEW METHOD TO DETECT ALUMINUM IN BLOOD SUBSTITUTE

Patients needing emergency blood transfusions can be given infusions of albumin as an initial replacement for blood loss. This practice eliminates the need for blood typing and can reduce the possibility of disease transfer. However recent studies have uncovered a problem with albumin that could prompt other adverse health effects: trace levels of aluminum. In the past, aluminum was difficult to measure at these low concentrations. But now, at the request of the Food and Drug Administration (FDA), NIST researchers have developed effective methods for measuring aluminum in albumin that combine several chemical analysis techniques. Laboratories in pharmaceutical companies and hospitals can adopt these methods to screen albumin products before they are marketed or used. NIST also has measured aluminum levels in a number of albumin samples the FDA and manufacturers will use to ensure measurement reliability. Apparently a byproduct of the albumin manufacturing process, aluminum at trace levels has been correlated to diseases affecting nerve tissue and bone. Aluminum exposure is a problem for patients with kidney impairment, because they have no way of excreting excess aluminum, and for patients receiving large or frequent albumin doses.

NIST/NASA WORKING ON FIRE SAFETY IN SPACECRAFT

In the not-too-distant future, people may be living in spacecraft for months or even years. Currently, the National Aeronautics and Space Administration strictly limits the type and amount of flammable material in a NASA spacecraft. But, in large spacecraft housing many people over long periods of time, these limitations may not be feasible. To help the space agency have more flexibility in selecting materials, researchers from NIST are assessing the test methods currently used by NASA for determining the fire hazard of materials. One goal of the project will be to help NASA develop broader test methods which can determine a level of hazard for a variety of materials. Current NASA screening tests are limited to either passing or failing a material. The NIST fire researchers also will work with NASA to develop ways to assess on Earth the flammability of materials in an environment with little or no gravity.

NDE EVALUATION OF SHEET METAL FORMABILITY

NIST and private industry have successfully demonstrated a nondestructive, ultrasonic system for evaluating the formability of steel sheet. Consistent formability in the sheet metal stock is necessary in a metal stamping plant to produce reliable, consistent shapes. This is even more important in a highly automated plant, because the automated equipment has difficulty dealing with inconsistent sheets. Current practice is a time-consuming process of cutting a sample from the sheet metal roll, preparing several specimens, and testing them. The new system, designed by NIST in collaboration with a guest researcher from the University of Belgrade, was installed at an industrial plant for evaluation. Tests show that the system, based on non-contact electromagnetic acoustic transducers and the analysis of ultrasonic wave speeds, can make reliable measurements of the formability directly on the sheet prior to stamping. Various training aids, including a videotaped tutorial, operator's manual, and software, were also developed for the system.

NIST UPDATING COMPUTER SECURITY GLOSSARY

In computer parlance, terms like virus and worm refer to malicious software intended to damage or destroy computerized information. But even the experts often disagree on their definition. To help clear up confusion about these and other computer-security related terms, NIST is evaluating existing glossaries for inclusion in a bibliography. The bibliography is to be issued as a Federal Information Processing Standard in 1990. (FIPS are developed by NIST for use by the federal government.) Glossaries can be sent to Samuel P. McCrea, NIST, A216 Technology Bldg., Gaithersburg, MD 20899; telephone: 301/975-5237. If you know of a glossary but do not have a copy to send, please contact McCrea with the title and publisher.

FIRST FIELD TESTS OF RAILROAD WHEEL INSPECTION SYSTEM

The first field trials of a system for inspecting railroad wheels as they roll over a sensor installed in the rail were recently completed by NIST in a research program sponsored by the Federal Railroad Administration (FRA) to improve railway reliability and safety. Over the past 4 years, according to the FRA, railroad wheel failures caused 134 accidents totaling $27.5 million in damages. In the NIST system, electromagnetic acoustic transducer (EMAT) sensors were mounted in rails at the American Association of Railroads' test track in Pueblo, CO, where loaded freight trains were repeatedly driven over them. As the wheels passed over the sensors, ultrasonic pulses were launched into the wheel, and the echoes reflected from cracks in the wheel were detected and analyzed by computer-controlled instrumentation. The track and EMAT system survived the stress of operation and provided clear signals at train speeds up to 15 mph. Further testing will be conducted to calibrate the system and make it more robust, and to evaluate the on-line digital signal processing system. For more details, contact Ray Schramm, NIST, Div. 430, Boulder, CO 80303-3328; telephone: 303/497-3232.

PRIVATE SECTOR PRODUCT CERTIFICATION PROGRAMS

The Office of Standards Code and Information has published an updated directory of private sector product certification programs, replacing NBS SP 703, Private Sector Product Certification Programs in the United States, which was published in 1985. This new edition of the directory lists 132 organizations, 23 more than were listed in the 1985 edition. The product certification activities of these organizations are described by the type and purpose of each organization, the nature of the activity, products certified, standards used, certification requirements, any accreditation or recognition by a U.S. or foreign government agency or by the private sector, availability of services, methods of cost determination, and other relevant details. The information is based primarily on submissions by the organizations and therefore documents the organization's description of its own activities.

This volume is designed to serve the needs of federal agencies and standards writers for information on U.S. private sector certification programs. Manufacturers, engineers, purchasing agents, and others may also find this volume of value. NIST SP 774, Directory of U.S. Private Sector Product Certification Programs, is available for purchase from NTIS and the Government Printing Office.

EXPRESSION OF MEASUREMENT UNCERTAINTY

A draft International Organization for Standardization (ISO) Guide to the Expression of Uncertainty of Measurements was reviewed at a meeting of ISO/Technical Advisory Group (TAG) 4/Working Group (WG) 3 at the Deutsches Institut fur Normung (DIN) in Berlin, West Germany, November 14-17, 1989. The guide is based on a 1981 recommendation by the International Committee of Weights and Measures (CIPM), and the effort is jointly sponsored by ISO, the International Electrotechnical Commission, the International Organization of Legal Metrology, and the International Bureau of Weights and Measures. In addition, ISO/TAG4/WG3 met in Berlin with a counterpart working group of ISO/Technical Committee (TC) 69 on Statistics that is drafting a related standard. The two working groups agreed to collaborate in completing the guide. NIST is coordinating the U.S. input and participation in this work on behalf of the American National Standards Institute. This guide is expected to enhance significantly international harmonization of the expression of uncertainty associated with measurement standards used by testing and certifying laboratories. When completed, it will promote confidence in mutual acceptance among nations of test reports and certificates for affected products in international trade.

DIAMOND FILMS DESIRABILITY

A unique combination of superior properties makes diamond a desirable material for a large number of optical, electrical, optoelectronic, mechanical, thermal, and chemical applications. NIST scientists are obtaining important data relating diamond film processing and structure to properties important to optical and optoelectronic applications. In particular, the group is examining the role of deposition conditions on morphology and the identity and distribution of optically active defects in the films. The films are grown by chemical vapor deposition in a hot-filament reactor. Researchers have recently studied the effects of feed gas composition on growth and properties of the films. The growth rate increases almost linearly with increasing methane content, and the surface morphology becomes smoother as the methane fraction in the feed gas increases. However, the quality of the diamond, as determined by Raman spectroscopy, decreases due to the increased presence of graphitic bonding. Examination of diamond films and particles by cathodoluminescence and photoluminescence reveals the absorption edge at the electronic bandgap, and the presence of dislocations and nitrogen related point defects. The dislocations and point defects are found to be inhomogeneously distributed in the films.

NIST/NIH NANOMETER ANALYSIS FACILITY: IMPLEMENTATION OF PARALLEL DETECTION ELECTRON ENERGY LOSS SPECTROMETRY

The analytical capabilities of the NIST/NIH (National Institutes of Health) Nanometer Analysis Facility, which is based upon a field emission scanning transmission electron microscope located at NIH, have been augmented by the implementation of a parallel detection electron energy loss spectrometer. In electron energy loss spectrometry (EELS), the energy of electrons transmitted through a thin (<50 nm) specimen is measured to reveal characteristic core level energy losses, which can be used to detect and quantify elemental constituents. Generally EELS is more sensitive than energy dispersive x-ray spectrometry for the analysis of thin specimens because the inelastic scattering is strongly peaked in the forward direction, which permits efficient collection of the signal, and because the signal is not partitioned among two or more de-excitation processes as is the case for x-ray emission.

Spectral information present in the fine structure at core edges can be used to determine nearest neighbor atom distributions. Previous EELS systems relied on a scanning single-channel spectrometer. The recent development of suitable spatially resolved detectors for EELS has led to the development of parallel detection. Parallel detection EELS provides an increase in collection efficiency of a factor of approximately 1,000 over single channel collection. The PEELS system is currently being applied by NIST scientists to the study of high T_c superconductors.

DEMONSTRATION OF THE QUANTUM ZENO EFFECT

Quantum mechanics predicts that the act of observing a system influences the behavior of that system. NIST scientists have recently completed the first unambiguous confirmation of one effect of this type—namely, that measurements of the state of a system tend to prevent that system from changing its state. The more frequent the measurements, the stronger the effect. Their particular experiment confirms one aspect of predictions by University of Texas theorists that a continuously observed quantum state can never decay. They referred to this as the quantum Zeno effect, after the paradox conceived by the ancient Greek philosopher, Zeno.

The NIST experiments involved observation of a few thousand $^9Be^+$ ions contained in an electromagnetic trap. Having placed all of the atoms in a particular, long-lifetime state, they noted that the transitions to a second state were heavily inhibited by brief observations made during the period of the transition. All of their ions moved to the second quantum level if they waited 256 ms before making an observation, whereas rapid observations every 4 ms allowed only a few ions to make the transition. The interpretation of the experiments involves a basic prediction of quantum mechanics, namely, that the act of observation projects the system into a particular quantum state. As a given atom progresses toward the second state, it can be described as being in an admixture of the initial and final states. If the system is observed during this process, it must be localized in either the final or initial state. Thus, early observation in the transition process yields a high likelihood of projecting the ion into the initial state where it must start through the transition process all over again.

HIGHEST "Q" VALUE ACHIEVED FOR OPTICAL FREQUENCY MEASUREMENTS

Scientists at NIST have recently reported the highest "Q" ever observed in atomic or molecular spectroscopy. In their experiments a single, laser-cooled mercury ion was stored in a Paul electromagnetic trap and radiated by a probe laser. The natural linewidth of the probed transition, which lies in the ultraviolet region of the spectrum, is extremely narrow, about 2 Hz. Several factors normally prevent one from measuring such a narrow width. A special dye laser (563 nm) was developed to improve the measurement of this narrow transition. This laser, stabilized to a carefully constructed Fabry-Perot cavity, was narrower than any visible laser ever reported. The frequency of this laser is doubled to probe the optical "clock" transition in mercury. The published resonance width (full width at half maximum) of 180 Hz was limited only by the laser linewidth. The most recent results indicate a linewidth of less than 80 Hz, giving a quality factor for the frequency measurement of $Q = \nu_0/\Delta\nu = 10^{13}$.

The systematic uncertainties in the mercury transition are estimated to be controllable to about one part in 10^{18}, making it an excellent candidate for an optical frequency standard.

COMPUTER SECURITY GUIDES PUBLISHED

NIST has published three guides on improving the awareness of executives, managers, and users in computer security requirements and one on establishing computer security training programs. The guides are NIST SP 500-169, Executive Guide to the Protection of Information Resources; NIST SP 500-170, Management Guide to the Protection of Information Resources; NIST SP 500-171, Computer Users' Guide to the Protection of Information Resources; and NIST SP 500-172, Computer Security Training Guidelines. The publications will assist federal agencies in complying with the requirements of the Computer Security Act of 1987 by establishing training programs to improve computer security practice and by improving employee awareness of the threats to computer systems.

NIST ISSUES SOFTWARE GUIDES

NIST Special Publication 500-173, Guide to Data Administration, provides a reference model for the various activities performed by data administration, information resources administration, data modeling tools administration, and database administration. Data administration is the management of information describing the data, functions, operations, and structure of automatic data processing systems and databases. The guide stresses the features of certain computing tools useful for data administration, such as data dictionary systems and computer-aided software engineering tools. NIST Special Publication 500-174, Guide for Selecting Automated Risk Analysis Tools, describes important considerations for developing selection criteria for acquiring risk analysis software and recommends a process for selecting automated risk analysis tools. The document recommends selecting a group of personnel with special skills to participate in the risk analysis studies and describes three essential elements of a risk analysis tool: data collection, analysis, and output results.

MAGNETIC FORCE MICROSCOPE FOR IMAGING MAGNETIC STORAGE MEDIA AND SUPERCONDUCTORS UNDER DEVELOPMENT

NIST recently adapted a scanning tunneling microscope (STM) for imaging the variations in the magnetic field above the surface of a computer disk as a magnetic tip is translated over the surface. The work is aimed at developing new high-resolution methods for imaging very small magnetic domains in materials—such as the "bit" patterns stored in magnetic media or the surface manifestation of the magnetic flux pattern known as an Abrikosov vortex lattice of type II superconductors. In contrast to recent developments reported elsewhere, to date with confirmed resolution of 100 nm, the NIST work has been based on simple force detectors scanned with an STM and is expected to result in a much less costly instrument having a potential resolution of 1 nm.

NIST uses a very-high-compliance magnetic STM tip that is in direct tunneling contact with the sample. The tunneling signal is used to keep the tip from physical contact with the sample. The resulting image is a convolution of surface topography and magnetic field. If the compliance of the magnetic tunneling tip is high enough, then the contribution of topography to the total image becomes negligible. This method has the potential of pushing the limit of magnetic image resolution to below 1 nm because of the close proximity of the magnetic tip to the sample surface. Tips were produced from pieces of a 1-μm Fe film having a spring constant smaller than 0.01 N/m, in order to detect typical tip sample magnetic forces, and a resonant frequency above 10 kHz, to be immune from instrument vibration. NIST is constructing a cryogenic version for superconductor flux lattice studies.

NIST DELIVERS HIGHLY ACCURATE SENSOR SYSTEM

NIST recently delivered a sensor system to a government manufacturing facility which produces precision turned metal parts. The sensor system requirements were that it measure part thickness from one side, in process, rapidly (about 30,000 measurement locations per part), and with high accuracy (better than 100 μin). The system has been demonstrated at NIST to meet all the measurement requirements. The system uses pulsed ultrasound and a measurement of transit time between the front and back surface of the part to determine local part thickness. The ultrasound is coupled to the part by a laminar stream of coolant/lubricant already in use in the process. The measurements are made in process so that any necessary corrections to the machining path can be made before dimensional control is lost. Plans are under way to enhance the system so that surface finish and shape profile can also be determined in process.

STANDARD E1321 TEST METHOD FOR DETERMINING MATERIAL IGNITION AND FLAME SPREAD PROPERTIES

A new test method for ignition and flame spread successfully passed the American Society for Testing and Materials (ASTM) society ballot. This is the last step to becoming an official standard. The acceptance was formalized at a recent meeting of ASTM E-5. This is the first fire test standard to provide measurement data consistent with theories of ignition and flame spread. Specifically, it provides data for radiative ignition on a vertical surface and data for lateral flame spread in terms of radiant heating, and surface temperature. Material properties can be deduced which can aid manufacturers in assessing the performance of their products in terms of specific components or additives. NIST scientists developed the test method and chaired the ASTM task group that guided the development of the standard. The test is also being considered by the International Standards Organization.

SHEARING APPARATUS FOR SMALL-ANGLE NEUTRON SCATTERING

A couette-type concentric cylindrical apparatus to investigate liquids at equilibrium and under shear by small-angle neutron scattering (SANS) has been developed by NIST scientists. This apparatus is the first of its type designed and built to be a general-purpose tool available to all researchers at a user-oriented neutron scattering facility in the United States. Design features include: a versatile yet rugged construction; a wide range of shear rates (up to 2000 Hz) and operating temperatures; and fully automated operation via a dedicated computer control system.

The cell consists of a stationary cylindrical quartz stator surrounded by a cylindrical quartz rotor with a 0.5 mm gap for the sample. Thermostating fluid is circulated within the stator for temperature control. The outer cylinder is directly coupled to a direct current brushless servo motor whose direction, velocity, and acceleration can be programmed to execute any combination of motion changes.

The cell has been used successfully to observe changes in the interparticle structure associated with the shear melting of charge stabilized colloids of spherical polystyrene latex particles. Other anticipated uses for the apparatus include: the alignment of anisotropic macromolecules by shear flow in order to determine their aspect ratio and orientational relaxation rates, tests of non-Newtonian behavior in both simple and complex fluids, and measurements to relate macro-molecular interactions to bulk rheological properties. Recently, a shear cell has also been developed by NIST scientists for polymer applications.

NIST AND MIT DEVELOP NEW METHOD FOR MEASURING SKIN DOSE FROM HOT PARTICLE EXPOSURE

In a joint project between NIST and the Massachusetts Institute of Technology (MIT), a new method for measuring skin dose from radioactive "hot" particles has been developed. Particles which are found in and around nuclear reactors, and those identified in fallout from the Chernobyl nuclear accident, have resulted in an increased need for more accurate measurement of skin dose. These particles are very small (less than 200 μm in diameter), have high beta-particle activities, and deliver high-dose rates to very small areas of the skin. In the past, dose estimates have been based on radionuclide identification and calculations.

Well-characterized spherical metal particles were fabricated and irradiated at the MIT reactor (to produce mainly ^{60}Co) and sent to NIST for shallow dose rate measurements using radiochromic dye films. A series of exposures was made with each particle, and the films were read on a laser scanning densitometer with a resolution of 40 μm. By combining the film images, dose rate information can be obtained for areas as wide as several mm in radius, which allows dose averaging over the 1-cm^2 area recommended for dose reporting. Preliminary results show good agreement with other experimental methods and point up shortcomings in currently used dose calculation algorithms.

FLUID-STRUCTURE INTERACTION STUDY COMPLETED IN CCT WATER TUNNEL

An experimental study of the interaction between a high Reynolds number boundary layer and a passive compliant surface has, for the first time, succeeded in producing flow velocity and surface displacement measurements for small amplitude, stable surface waves. This 3-year program was a collaborative effort involving the Office of Naval Research, NIST, and Johns Hopkins University. The program included flow measurement surveys for both noncompliant and two compliant surfaces. Measurement techniques included single component hot-film anemometry and laser-based surface displacement detection. Still photography and videotapes were used to make visual records of the compliant surface motions. Spectral analyses of these results were used to quantify the characteristics of the flow-structure interaction. Based on the results described above, it was concluded that actively controlled surfaces could play a decisive role in fluid-structure interactions. If fluid-structure interactions can be altered in such ways, it may be feasible to modify drag characteristics, acoustic properties, and heat and mass transfer rates. Potential application areas would include mixing processes, drag reduction for surface and underwater vehicles, sound absorption, vibration reduction, and noise shielding.

NEW COMPUTER SIMULATION DESIGN TOOL

Computer simulation of the dynamics of molecules is a powerful and widely used technique for studying the thermal properties of a wide range of materials, including liquids, crystals, and polymers. An important question about these simulations is: "How long must they be run so that reliable results are obtained?" The problem is knowing when the simulation has fully sampled the possible states of the system. To date, the methods used to estimate the time needed for the sampling to converge to the correct answer have all been empirical. Recently a quantitative method to determine convergence times for specific properties has been developed at NIST. The method predicts convergence times using the equivalence of long time interval averages and statistical (ensemble) averages for systems in thermal equilibrium (or in stationary, nonequilibrium states) along with the results of a short time interval simulation. The method is quite general and will be extremely useful in a wide variety of simulations of complex systems.

NIST SPONSORS HYPERTEXT STANDARDIZATION WORKSHOP

A Hypertext standardization workshop to address formally standardization issues was sponsored by NIST in January. Hypertext technology is an approach to information management in which data is stored in a network of nodes connected by links. The nodes, which may contain text, graphics, audio and video, source code, and other data, are viewed through an interactive browser and manipulated through a structure editor.

Papers presented at the workshop focused on requirements, reference models, and candidate specifications. Several well-known researchers and different industry groups already informally involved in some aspect of Hypertext standardization contributed their results to the workshop with the expectation that it could evolve into a more public forum for the planning and development of Hypertext standards.

FUNCTIONAL REQUIREMENTS OF NETWORK MANAGEMENT BASED ON OPEN SYSTEMS INTERCONNECTION (OSI) PUBLISHED

To provide for the management of future interoperable multivendor networks, the International Organization for Standardization (ISO) and other international organizations are developing management standards for communications networks based on the OSI reference model. NIST Special Publication 500-175, Management of Networks Based on Open Systems Interconnection (OSI) Standards: Functional Requirements and Analysis, examines current and proposed network management systems to determine both user and functional requirements for network management.

The report compares the derived functional requirements to emerging standards to determine where and how requirements are being met. Deficiencies are noted in cases where requirements are not being met. The examination of requirements focuses on those necessary for interoperability in the following broad areas: architecture, configuration management, fault management, security management, performance management, and accounting management.

FEDERAL INFORMATION PROCESSING STANDARD (FIPS) 127, DATABASE LANGUAGE STRUCTURED QUERY LANGUAGE (SQL), REVISED

A revision to FIPS 127 was recently approved. The revision incorporates Database Language Embedded SQL (ANSI X3.168-1989), recently published by the American National Standards Institute (ANSI). Embedded SQL adds programming language access alternatives to existing FIPS 127 specifications. Revised FIPS 127 also consists of Database Language SQL (ANSI X3.135-1989), which ANSI will soon publish. In addition, NIST released version 2.0 of the SQL test suite, which helps users and vendors determine compliance with FIPS 127, Database Language SQL. This version of the test suite includes tests for features to be available in the revision of FIPS 127, as well as tests for direct invocation of SQL commands (interactive SQL) and utility software to support the validation process. Version 2.0 replaces version 1.2 of the SQL test suite released in May 1989 and will be used in the SQL testing service that will start in April 1990. Over 40 organizations presently utilize the SQL test suite.

NIST CONTRIBUTES TO ASSESSMENT OF "LEAD IN PAINT" DETECTORS

The Department of Housing and Urban Development (HUD) is in the process of implementing a lead abatement program in public housing. To decide whether abatement is needed, they must determine the lead concentration in paint on walls, rails, cabinets, and so forth. HUD requested NIST to assess methods for measuring lead concentration in paint. NIST has provided the necessary statistical guidance. As part of the overall project, NIST scientists examined x-ray fluorescent (XRF) lead detectors because they are nondestructive and relatively simple to use. In order to assess the measurement quality of two types of commonly used lead-specific XRF detectors under a variety of conditions typically seen in the field, NIST proposed running a statistically planned experiment. This ensured that a balanced set of conditions were studied for each XRF detector and allowed independent evaluation of each source of uncertainty. After running the experiment, graphical and analytical statistical techniques were used to analyze the data. Results indicated that the nondestructive detectors examined were not capable of quantitatively measuring lead concentrations near the regulatory limit of 1 mg/cm^2. The study clearly established that improved techniques are needed.

NIST-MIT RESEARCH IDENTIFIES SOURCE OF WEAK-LINK BEHAVIOR IN CERAMIC SUPERCONDUCTOR

NIST and Massachusetts Institute of Technology scientists have identified the source of weak links in a ceramic superconductor. The superconductor in question is $BaPb_{1-x}Bi_xO_3$, which has a cubic structure and a relatively low critical temperature of 13 K. In spite of its isotropic structure and long coherence length of about 75 Å, this material's grain boundaries behave as weak links. The presence of weak links (unintentional junctions of superconductor-normal material-superconductor or superconductor-insulator-superconductor exhibiting Josephson-effect behavior) in a superconductor restricts its usefulness, as weak links are thought to limit the transport current even at low-magnetic fields. The team found that the weak links are due to Pb-Bi-O rich phases at the grain boundaries which result from melting during annealing. Transport critical current density measurements on samples prepared at low temperatures (below 600 °C) show that weak links are still present, and Auger spectroscopy and scanning transmission electron microscopy reveal a corresponding composition variation at the boundaries. Currently, work is under way to establish the solidus temperature, below which no liquid phase is formed, in this system. The team also is carrying out transport critical current density measurements, as a function of both temperature and applied field, in order to show that the grain boundaries containing second phase behave as superconductor-insulator-superconductor junctions.

NIST REDUCES UNCERTAINTIES IN MEASUREMENTS OF PEAK IMPULSE VOLTAGE

NIST scientists have reduced the uncertainties to ±0.1 percent in measurements of impulse voltage peaks based on the use of analog oscilloscopes. NIST needs the improved measurement methods for the calibration of voltage dividers to respond to needs of the pulse-power community. Typical oscilloscope uncertainties in the measurement of peak voltages of microsecond impulses have been of the order of ±1 percent. Researchers previously developed measurement methods that employed dc reference levels and offsets to facilitate measurement on more sensitive oscilloscope scales, with resulting uncertainties of ±0.3 percent. Building on this work, they have achieved the new reduction by using dc reference voltages applied in the form of voltage steps to reduce the measurement errors arising from differences in the oscilloscope amplifier's response to pulsed and to dc inputs. The researchers have verified the method using a 5-V standard voltage step maintained by the division and have already applied the method to the calibration of high-voltage impulse dividers.

NIST PROVIDES EPA PRELIMINARY EVALUATION OF THE PERFORMANCE OF VARIOUS REFRIGERANT MIXTURES FOR CONVENTIONAL REFRIGERATORS

At the request of the Environmental Protection Agency (EPA), researchers at NIST developed a vapor compression refrigeration cycle model which was used to evaluate the performance of conventional refrigerators using various refrigerant mixtures. Mixtures of refrigerants that do not damage the ozone layer of the upper atmosphere are considered viable replacements to the refrigerants currently used in household refrigerators. NIST developed CYCLE11, which models the thermodynamic cycle of a household refrigerator and is a modification of the existing CYCLE7 model developed previously by NIST to simulate the cycle in air conditioners and heat pumps. Eleven different binary refrigerant mixtures were evaluated, and the results were presented to EPA. Several of the mixtures improved the steady-state efficiency of the refrigerators between 5 and 10 percent compared to the use of R-12. Future research will focus on modification of the basic refrigeration cycle to get further enhanced performance with refrigerant mixtures.

Standard Reference Materials

NIST ANNOUNCES NEW PORTLAND CEMENT STANDARDS

A new series of Standard Reference Materials (SRMs) has been developed by NIST for the producers and users of Portland cement. SRMs 1884-1889 are used in analyzing the composition of Portland cement by instrumental and chemical analysis. The new series of SRMs replaces the 630 series of materials and covers the same nominal range of concentrations of certified constituents. Information on each of the new SRMs and their certified constituent properties and percent by weight is available from the Office of Standard Reference Materials (OSRM). Available for $98 a unit, each SRM consists of three sealed vials, each containing approximately 5 g of material. To order SRMs 1884-1889, contact: OSRM, NIST, Rm. 204, Bldg. 202, Gaithersburg, MD 20899; telephone: 301/975-6776, FAX: 301/948-3730.

ACCURATE EMISSIONS ANALYSES ARE AIM OF GAS STANDARDS

Enforcement agencies at the federal, state, and local levels regularly survey outdoor pollution levels by measuring exhaust emissions from cars, trucks, and other vehicles. Environmental concentrations of the gases propane, carbon monoxide, and carbon dioxide are key components of such a survey. The reliability of these studies depends on how well laboratory instruments work and how accurate analytical methods are. NIST is now offering four different standard reference materials (SRMs) that can help ensure measurement reliability. Each certified to contain a specified concentration of the three gases in nitrogen, these SRMs can be used to calibrate analytical instruments and to check laboratory methods. By running a sample from one of the SRMs through an instrument, a chemist can compare the results with the concentrations listed for the SRM to determine instrument accuracy. The SRMs, numbers 2725-2728, are available for $870 each from the Office of Standard Reference Materials, NIST, Rm. 204, Bldg. 202, Gaithersburg, MD 20899; telephone: 301/975-6776.

NIST HELPS INDUSTRY WITH VEHICLE EMISSION MEASUREMENTS

With federal limits on vehicle emissions more stringent than ever, car manufacturers need to ensure that measurements are reliable and comparable between in-house labs and with regulatory agencies. Currently, some standards are available for certain emission pollutants that allow chemists to check the accuracy of analytical instruments. Now NIST, in partnership with the Motor Vehicle Manufacturers Association, is developing seven previously unavailable standard reference materials (SRMs) intended to be national standards that will add to the accuracy base of emission measurements. These bottled gas mixtures—nitric oxide in nitrogen and methane in air among them—represent levels of these pollutants typically found in auto exhaust and have been rigorously certified for their concentrations. Since the amount of a given component in these SRMs will be known, chemists can run a sample through their equipment as a reference to calibrate instruments and methods. The materials also will be useful to specialty gas manufacturers who produce the day-to-day standards used by the motor vehicle industry. NIST expects the SRMs to go on sale by fall of 1990.

MICROPOROUS FUMED-SILICA INSULATION

Microporous fumed-silica insulation consists principally of extremely fine particles (~10 nm) of amorphous silica and fine ceramic fibers, forming a light rigid board of extremely low thermal conductivity [of the order of 20 mW/(m×K]. The low conductivity is a result of the very small pore size and tortuous conduction paths of both solid and gaseous components of the bulk material. This material has been previously issued as a NIST standard reference material for thermal resistance at temperatures below 330 K, and will be certified for its apparent thermal conductivity at temperatures from 318 to 733 K (45 to 460 °C) and air pressures from 26.7 to 83.5 kPa (200 to 626 Torr). A report describing these studies, Microporous Fumed-Silica Insulation as a Standard Reference Material of Thermal Resistance (NISTIR 89-3919), is available from the National Technical Information Service, Springfield, VA 22161. Order by PB #90-130311/AS for $17 ($8 for microfiche).

MATERIALS DESIGNED TO HELP MONITOR WATER POLLUTION

Environmental agencies, as well as others studying pollution in the nation's waterways, need materials containing an accurate composition of various compounds as a check to verify the reliability of laboratory instruments and methods. Now NIST has developed a bottled standard reference material (SRM) for this purpose. It contains marine sediment with a wide range of pollutant compounds of interest to environmental scientists. The sediment material, which has certified values for 11 polycyclic aromatic hydrocarbons (PAHs), was collected from the Chesapeake Bay area near Baltimore harbor. It is in dry powdered form which can be reconstituted into wet form so that the compounds can be extracted by solvents for organic analysis. The material also contains noncertified values for other PAHs, polychlorinated biphenyls, and chlorinated pesticides. It is available for $241 from the Office of Standard Reference Materials, NIST, Room 204, Building 202, Gaithersburg, MD 20899; telephone: 301/975-6776.

NIST INVESTIGATES RELATIONSHIP BETWEEN BONDED PHASE MORPHOLOGY AND CHROMATOGRAPHIC SELECTIVITY IN REVERSED-PHASE LIQUID CHROMATOGRAPHY

A major problem confronted by liquid chromatographers worldwide is the unexplained differences in retention behavior among columns that are generically identical, but prepared by different manufacturers or by the same manufacturer at different times. As a result of careful studies of the reversed-phase separation process, scientists at NIST have developed a model that explains the effect of subtle differences in bonded phase morphology on relative retention (selectivity) for structurally similar compounds.

Three molecular probes have been identified whose retention behavior provides the basis for classifying columns according to bonding phase morphology (i.e., those exhibiting behavior characteristic of monomerically or polymerically bound ligands). The elution order for these probes is useful in predicting the efficacy of columns for the difficult separation of isomers. Isomeric separations are becoming increasingly important in nutrition, cancer chemoprevention, and environmental/human health studies.

These efforts have provided the first evidence that temperature influences other parameters besides absolute retention and efficiency (the sharpness of bands). By performing separations of key probe molecules at various temperatures (−30 to 100 °C), it was observed that elution orders can change with temperature and that "monomerically bound" reversed-phase columns that normally provide little selectivity for the separation of isomeric compounds could be made to acquire "polymerically bound" behavior and provide separations of isomers as the temperature is lowered to the appropriate point. Conversely, columns with "polymerically bound" ligands can be induced to acquire "monomerically bound" character by increasing the temperature to the appropriate point.

Columns using "monomerically bound" ligands are used for the vast majority of separations in liquid chromatography. These results provide chromatographers with a tool for fine tuning the selectivity of columns obtained from various sources to meet specific separations needs. The three component probe mixture is being issued as SRM 869 and will be used by both manufacturers and bench scientists worldwide for column classification and quality control.

RADIOCHROMIC DYE STANDARD REFERENCE MATERIAL FOR HIGH-DOSE DOSIMETRY

NIST announces the availability of a new Standard Reference Material (SRM 4500) for the radiation processing industry. SRM 4500 is a radiochromic dye solution which can contribute to the standardization of measurements of absorbed dose in many radiation processing applications, including sterilizing medical devices, curing polymers and elastomers, testing electronics, and extending the shelf life of foods.

These standards can be used to calibrate cobalt-60 or cesium-137 sources of gamma radiation for absorbed dose in the range 50 Gy to 5 kGy (5-500 krad). Once the solution is irradiated, a blue chromophore is formed whose color intensity is linearly related to absorbed dose. The intensity is measured at 600 nm and the corresponding absorbed dose is calculated from the well-established values of the radiation chemical yield of the dye molecules in solution and their linear molar absorption coefficient at 600 nm.

SRM 4500 consists of a set of six flame-sealed amber glass ampoules. Each ampoule contains 5 mL of solution of a radiochromic dye in a solvent composed of 15 percent dimethyl sulfoxide (DMSO) and 85 percent n-propanol. The notes on the use of the SRM provide details on the characterization of the material for stability, dose rate dependence, dose range, and temperature coefficient.

CERTIFICATION OF COCAINE IN URINE SRM COMPLETED

NIST has an ongoing program in cooperation with the College of American Pathologists to provide the drugs- of-abuse testing community with urine-based reference materials. The first such material was SRM 1507, a freeze-dried urine pool certified for the principal urinary metabolite of marijuana (20±2 ng/mL). Scientists at NIST have recently completed work on SRM 1508, a freeze-dried human urine pool with certified concentrations of cocaine and its principal urinary metabolite, benzoylecgonine (BE). Each unit of this SRM consists of four bottles of freeze-dried urine: one bottle for each of three levels of cocaine and BE, plus one bottle of blank urine.

The SRM was certified using two independent methods that agreed within a statistical tolerance. One of the methods was based on a gas chromatography/mass spectrometry (GC/MS) procedure and was similar to those used in drug testing laboratories to confirm positive results from preliminary screening analyses. Information values are provided for the methyl ester of ecgonine, another important metabolite of cocaine in each of the three levels.

The certified concentrations (in ng/mL) in SRM 1508 are as follows: Level 1—cocaine 90±10, BE 104±7; Level 2—cocaine 254±10, BE 261±16; and Level 3—cocaine 425±10, BE 515±32. These concentrations bracket the cutoff level set by the National Institute on Drug Abuse (NIDA) for establishing cocaine abuse, thereby providing laboratories a basis for validating the accuracy of their measurement methods throughout the legally critical concentration range. This SRM should also be of benefit to laboratories making measurements concerning scientific and health-related questions regarding cocaine abuse.

Standard Reference Data

NEW DATABASE ON ELECTRON AND POSITRON STOPPING POWERS AND RANGES AVAILABLE FOR PC USERS

Medical and health physicists, laboratory researchers, and industrial users of high-energy radiation equipment will benefit from a new personal computer (PC) version of an important database developed by NIST. The database also provides for rapid calculations on the collision, radiative, and total stopping powers of materials, their continuous-slowing-down approximation (CSDA) ranges, and the radiation yields for electrons or positrons with kinetic energies from 1 keV to 10 GeV.

The new PC resource, NIST Standard Reference Database 7A, Electron and Positron Stopping Powers and Ranges, was developed from a database that is available for lease on magnetic tape. Unlike the tape version, which can only supply information on the performance of materials in tables of stopping powers and ranges, the PC program gives these quantities for any material that can be described by chemical formula using standard symbols.

The new PC version of the NIST database is available on 3-1/2 in or 5-1/4 in disks for $360. The program can be stored on a hard disk of any AT or XT-Class PC.

To order NIST Standard Reference Database 7A, contact the Office of Standard Reference Data Programs, A323 Physics Bldg., National Institute of Standards and Technology, Gaithersburg, MD 20899; telephone: 301/975-2208.

Calendar

October 1–4, 1990

13th NATIONAL COMPUTER SECURITY CONFERENCE INFORMATION SYSTEM SECURITY: STANDARDS—THE KEY TO THE FUTURE

Location: Omni Shoreham Hotel
Washington, DC

This conference provides a forum for the government and the private sector to share current information that is useful and of general interest to the conference participants on technologies, present and future, that are designed to meet the ever-growing challenge of telecommunications and automated information systems security. The conference will offer multiple tracks for the needs of users, vendors, and the research and development communities. The focus of the conference will be on: systems application guidance; awareness, training and education; ethics, evaluation and certification; innovations and new products; management and administration; and disaster prevention and recovery. Papers on the above topics are being solicited. Sponsored by NIST and the National Computer Security Center.

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