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PAPER

Electret Ion Chambers for Passive Measurement of Airborne Tritium: Theory and Practice

P. Kotrappa, T. Hobbs*, and D. Brown*
Rad Elec, Inc.
*National Institute of Standards and Technology

Commercially-available electret ion chambers (EICs) are already in use for passive measurement of radon concentration in air. Modified versions of these are usable for the measurement of airborne tritium. Modification provides increased filtered area for rapid, passive exchange of airborne tritium or tritiated water vapor between the interior of an EIC to the outside air. Response factors are calculated from the first principles. These units are exposed to airborne tritiated water vapor inside an airtight chamber for a known period of time to deduce the experimental response factors. One-liter, vibrating-reed-electrometer tritium detectors calibrated by the National Institute of Standards and Technology (NIST) are used as reference detectors. EIC units with three different volumes (58, 210, and 960 mL) and electrets of two different sensitivities ("short-term" and "long-term") are studied. Using a "short-term" electret in a 210-mL-volume chamber, a 0.5 derived air concentration (12.5 µCi m^{-3} d) is measurable with an accuracy of about 10% in one day. Theoretically calculated and experimentally determined response factors agree fairly well, demonstrating the scientific validity of the theory. Field testing of these devices performed at Pickering Nuclear Facilities indicated satisfactory performance over a range of concentrations from 5 to 50 µCi m^{-3}. Advantages and disadvantages of using the EIC units for passive measurement of tritium are discussed.

Introduction

Electret ion chambers (EIC) have been used for the measurement of gamma radiation since 1978.[1-3] This method has not been widely used, however, because of the possible errors introduced by environmental factors (i.e., temperature and humidity) effecting the stability of the electrets. These problems have been recently solved.[4-6] The commercial availability of stable electrets of different thicknesses, suitable chambers, and a low-cost electret voltage reader have made it practical to have a viable EIC system. Field worthiness of these devices was demonstrated by their use in ambient radon measurement and in environmental gamma-radiation measurement.

Background

The EIC is an integrating ionization chamber wherein the electret (a charged Teflon™ disk) serves both as a source of an electrostatic field and as a sensor. It consists of an electret mounted inside a small chamber made out of conducting plastic. These are passive devices. The ions produced inside the chamber are collected by the electret causing a reduction of the charge on the electret. The reduction in charge is a measure of the total ionization occurring during that period in the volume of the chamber. The charge on the electret before and after the exposure is measured by a portable electret voltage reader. The sensitivity and the dynamic range depends upon the thickness of the electret, the area of the electret and the volume of the chamber. Several combinations of chamber volume and electret thicknesses are available. An optimum unit may be chosen for a particular application.

Theory and scientific basis

Figure 1 gives a schematic view of different commercially-available chambers. The volumes of these chambers vary from 50 mL to 1000 mL. The S-type chamber has an on/off mechanism which can be used to close and open the electret from outside.[5] Electrets of two different thicknesses[5] are commercially available. An electret with 1.542-mm thickness is usually referred to as a short-term (ST) electret and an electret with a

$$\frac{25 \times 0.4}{(10\,\mu\mathrm{Ci/m^3\text{-}day})}$$

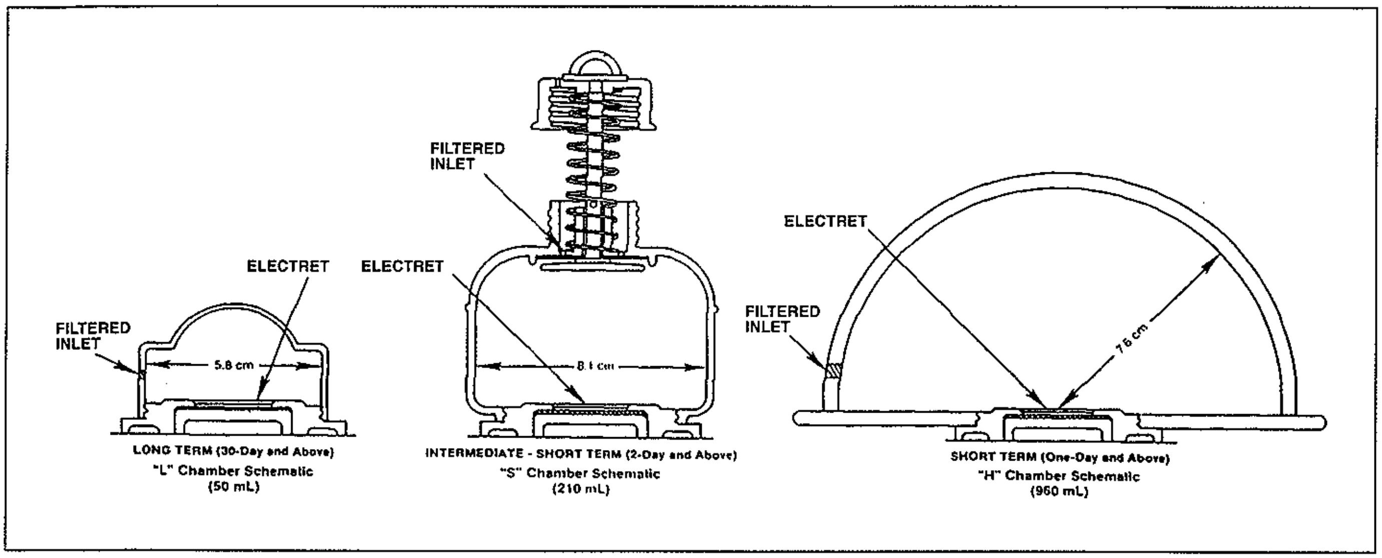

Figure 1 Schematic view of different commercially available EIC chambers.

thickness of 0.127-mm is referred to as a long-term (LT) electret.

An electret is used as a source of high voltage and also as a sensor. Therefore, the stability of the electret is very important. The discharge of the electret should occur only from the collection of ions and not from other environmental factors (i.e., humidity or temperature). Change in the surface voltage of the electret due to collection of ions should be permanent. It should be easy to measure the surface charge, and it should be a nondestructive method. These requirements are generally satisfied by the commercially-available electrets.[4-8]

Conventional ion chambers measure current, whereas EICs measure the collected charge which, when divided by the time of exposure, is also equivalent to an average ionization current. Since there is no limit for the exposure time, it is possible to have high sensitivity in small volume and at normal pressures.

Equation (1) gives a relationship between the surface charge, measurable surface voltage, the area of the electret and the thickness of the electret. This equation is the basis for controlling the EIC's response by choosing electrets of different thickness. For example, by holding the area constant, a 10-V change in a 1-mm-thick electret corresponds to the same change in electret charge as a 20-V change in a 2-mm-thick electret. Therefore, it is desirable to use thicker electrets when higher sensitivities are needed. This equation was verified recently for thicknesses ranging from 0.0015 to 0.1524 cm (two orders of magnitude).

$$\frac{Q}{V_s} = \frac{\varepsilon\, \varepsilon_0\, A}{d} = 9.400 \times 10^{-12}\, \frac{\mathrm{C}}{\mathrm{V}} \tag{1}$$

where:

Q = the total charge on the electret
V_s = the surface potential of the electret (V)
ε = the relative dielectric constant of the electret material (2.0 for Teflon)
ε_0 = the dielectric constant of free air (8.85×10^{-14} F/cm)
A = the area of the electret ($8.43\ \mathrm{cm}^2$)
d = the thickness of the electret (0.1588 cm)

Using the appropriate constants (given in parenthesis) for one of the commercially available electrets, equation (1) shows that a change of 1 V in the surface potential corresponds to 9.400×10^{-12} C of charge.

Another method of controlling the sensitivity is to change the volume of the chamber. For the same radiation dose, more ionization charge is created and more charge is collected on the electret in a chamber with a larger volume. However, recombination error starts appearing at larger volume limiting this direction of increasing the sensitivity.

Electret ion chamber for tritium measurement

Tritium, being a low-energy beta-particle emitter, has a very small range in air. If tritium gas or tritiated water vapor gets into the sensitive volume of the EIC, the energy is fully dissipated inside the volume. Therefore, it is possible to calculate the response of an EIC under such circumstances. The response factor (RF) of an EIC is defined as a drop in voltage per unit integrated concentration in one day. For tritium let us define it as electret voltage drop when exposed to 1 μCi m^{-3} for one day. The following analysis presents a theoretical method of calculating the RF for a tritium EIC with a chamber volume of 1 m^3.

The first step is to calculate the charge produced by 1 μCi of tritium in 1-m^3 volume in one day. It is assumed that the entire energy is dissipated inside the chamber and goes into causing ionization. Since 5685 eV is the initial average energy of a beta particle from tritium and 33.97 eV creates an ion pair, 8.5605×10^{-8} C of charge is produced per day. Dividing this value by the result from equation (1) yields a RF of 9107 V/d. This means that an EIC with an electret of 0.1588-cm thickness and a volume of 1 m^3 leads to a drop of 9107 V when exposed for one day to a tritium concentration of 1 μCi m^{-3}.

Table 1 gives the calculated response factors for chambers of different volumes and electret thicknesses. The table also reports experimentally deduced response factors. The differences between the calculated and measured values may be due to inadequate assumptions. Please refer to later sections.

Chamber volume (mL)	Electret thickness (cm) 'ST'	Calculated RF	Experimental RF (MPV of 500 V)
58	0.1588	0.53	0.68
210	0.1588	1.91	2.2
960	0.1588	8.80	8.9

Table 1 Theoretically calculated and experimentally determined response factors (RF) for EIC in V/(µCi m^{-3} d).

Practical EIC passive monitors for tritium

Electret ion chambers used in earlier studies[10,11] demonstrated the feasibility of this technology. The units studied were flow-through devices and the problems relating to the stability of electrets were not solved at that time, especially when used in very high humidity. These were far from being used as practical units. The present study uses practical and commercial devices that are available for the measurement of integrated average tritium concentrations.

Commercially-available EIC units for monitoring radon are not suitable for the measurement of tritium or tritiated water vapor because of the relatively small filtered inlet area that minimizes its response to the short-lived radionuclide of radon (^{220}Rn). These units were modified to increase the inlet filtered surface area by a factor of 100. The filter used was carbon-coated Tyvek™ which is known to be highly permeable to water vapor. Figures 2, 3 and 4 give cross-sectional views of the modified L (58 mL), S (210 mL) and H (960 mL) chambers respectively. These units are used in the current study as passive EIC tritium monitors.

Test chamber for calibration of EIC tritium monitors

The first step is to have a reproducible atmosphere of airborne tritium concentration for the calibration. Since the EIC units are known to perform well even at 100% relative humidity, the constant source of airborne tritiated water vapor in a sealed enclosure is considered to provide a satisfactory system.

A petri dish of 10-cm diameter containing approximately 150 mL of tritiated water was introduced at the bottom of a 10-L plastic vacuum desiccator. When the lid for this enclosure was shut, water vapor came into equilibrium with air rapidly. The moisture concentration in air depends upon the vapor pressure which in turn depends upon the temperature. A trial experiment indicated that the relative humidity went up from 40% at zero time, to 80% after 7.5 minutes, and to near saturation in less than 15 minutes. Table 2 lists the vapor density and the airborne concentration of tritiated water vapor expected from a water sample containing tritium at a concentration of 1 µCi mL^{-1} for temperatures between 15 and 30 °C. It is assumed that the isotopic ratio ($^{3}H/^{1}H$) essentially remains the same in water and in airborne water vapor. Table 3 summarizes the theoretically expected airborne tritium concentration and measured values under saturation conditions. After about an hour of equilibration, an air sample was drawn into an evacuated one-liter vibrating reed electrometer chamber (VREC)

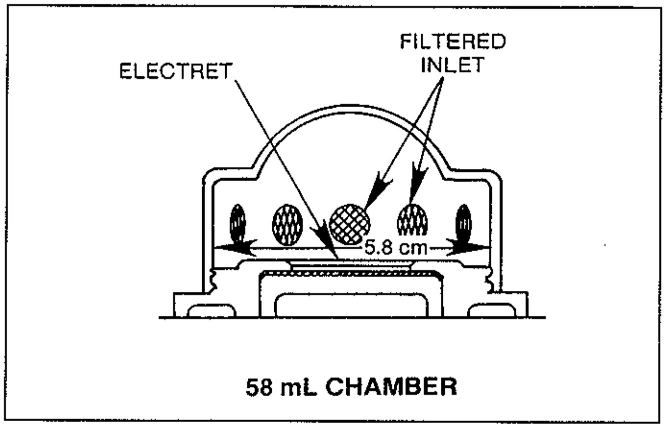

Figure 2 Schematic view of a 58-mL tritium EIC unit.

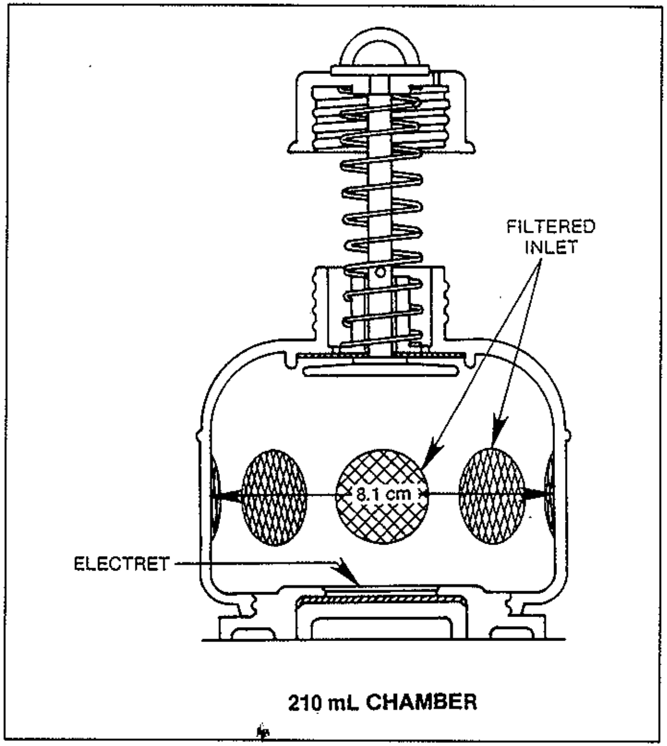

Figure 3 Schematic view of a 210-mL tritium EIC unit.

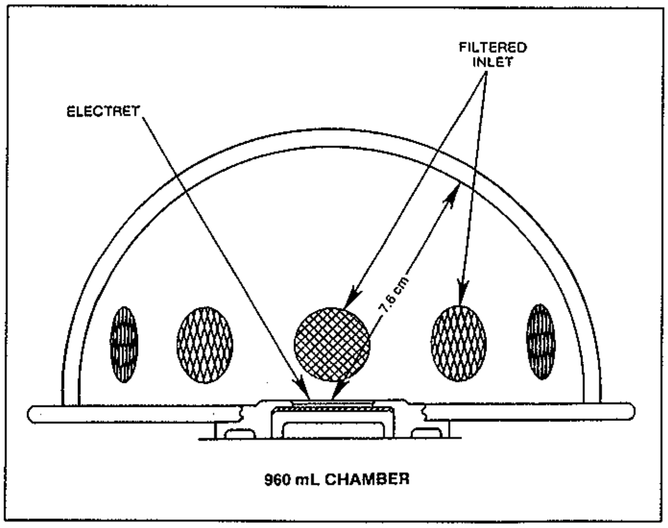

Figure 4 Schematic view of a 960-mL tritium EIC unit.

through a connecting spout. Suitable standard corrections were applied for the pressure differential and dilution in the VREC before arriving at the measured and corrected tritium concentration in the enclosure sampled. There was a consistent ratio of 0.80 between experimental values and theoretical values. However, this ratio was different from a published ratio of 0.90.[12,13] The reasons for the differences are not clear at present. The results were reproducible on the readings taken over 20 consecutive days from the same enclosure with the same source of tritiated water vapor. Such an enclosure gives a steady and reproducible atmosphere for calibrating different EIC units.

The data in Table 2 can be fitted by the analytical equation:

$$CV = -6.83 + 1.463 \times C \qquad (2)$$

where:

$CV = \mu Ci\ m^{-3}$ in air when water concentration is $1\ \mu Ci\ mL^{-1}$
C = the temperature in °C.

Equation (2) is useful in predicting airborne tritium concentration at a particular temperature from a water sample of known tritium concentration. It is also useful in calculating the tritium concentration in water by measuring the airborne tritium concentration. Determine tritium concentration in air phase ($\mu Ci\ m^{-3}$), divide it by 0.80 and further divide by CV to compute tritium concentration in water ($\mu Ci\ mL^{-1}$).

Experimental determination of response factors for EIC units

The response factor (RF) for EIC units does depend to some extent on the operating voltages. The RF can change[5] as much as 10 to 15% over electret voltages of 200 to 700 V. Therefore, for proper use of these units it is necessary to determine the RF as a function of operating voltage. The RF is different for EIC units of different volumes and for electrets of different sensitivities. The methodology used is similar to that used for determining the RF for measuring radon.[5] It is given by the general equation:

$$RF = \frac{(\text{ voltage drop })}{(\text{ cumulative concentration })} \qquad (3)$$

where cumulative concentration is given in the units of
$\mu Ci\ m^{-3}$ d.

The following procedures were adopted. Four units of the same type were loaded with appropriate electrets having a surface voltage of about 750 V. These were loaded into the test enclosure and the enclosure was sealed. After one day, the enclosure was sampled for tritium using the VREC and the enclosure was opened. The final electret voltages were measured. The voltage drop that was associated with a cumulative concentration ($\mu Ci\ m^{-3}$ d) was calculated. The average voltage (also called mid-point voltage or MPV) between the initial and final surface voltages of the electrets was calculated. The ratio of the voltage drop to the cumulative concentration was the response factor (RF) as shown in equation (3). This RF was associated with this particular MPV. The average RF and the respective average MPV of all four units was calculated. The units were placed back into the enclosure. This sequence was repeated on consecutive days and the experiment repeated for another day. The average RF and the average MPV was different for the second day and so on. The experiment was repeated until the final electret surface voltage was in the range of 100 to 150 V. A regression equation was derived between the average RF and the associated average MPV.

Table 4 gives typical calibration data for a 210-mL tritium EIC (S chamber) with an ST electret. Such a configuration is called an SST. The first column gives the electret serial numbers. Successive columns give the electret surface voltages after each exposure.

The following equation was derived from a regression fit to the data in Table 4. The correlation coefficient was 0.923 and the standard deviation of the estimate was 0.02817.

$$RF \frac{V}{\mu Ci\ m^{-3}\ d} = 1.34277 + 0.13476 \times \ln(MPV) \qquad (4)$$

Similarly, experimental response factors were derived for other chambers and electrets using appropriate tritium concentrations. These are given in equations (5) through (9).

$$RF(SST) = 1.3428 + 0.1348 \times \ln(MPV) \qquad (5)$$
$$RF(SLT) = -0.0289 + 0.0357 \times \ln(MPV) \qquad (6)$$
$$RF(LST) = 0.0059 + 0.1111 \times \ln(MPV) \qquad (7)$$
$$RF(LLT) = 0.0005130 + 0.009661 \times \ln(MPV) \qquad (8)$$
$$RF(HST) = -0.6270 + 1.5131 \times \ln(MPV) \qquad (9)$$

where:

RF = experimental response factor
MPV = mid-point voltage
SST = EIC unit with S chamber (210 mL) and ST electret
SLT = EIC unit with S chamber (210 mL) and LT electret
LST = EIC unit with L chamber (58 mL) and ST electret
LLT = EIC unit with L chamber (58 mL) and LT electret
HST = EIC unit with H chamber (960 mL) and ST electret

Figure 5 is a graphical representation of the experimental data.

Discussion of Table 5

Table 5 summarizes the response factors for the five different EIC configurations. Range is the range of EIC in units of $\mu Ci\ m^{-3}$ d. This corresponds to a discharge of fully charged electret from the initial voltage of 750 V to 200 V. A range of 250 means that such a unit can be used for only 10 days at a concentration of 25 $\mu Ci\ m^{-3}$. Sensitivity is the cumulative tritium concentration that gives a voltage drop of 10 V. Since the electret voltage reader reads only correct to 1 V, there is an uncertainty of ±1 V in the initial reading, and there is an uncertainty of ±1 V in final reading. Thus there is an uncertainty of about 15% when the voltage difference is about 10 V. This means that an SST unit can be used to measure a concentration of 4.6 $\mu Ci\ m^{-3}$ in one day with about 15% uncertainty. Range

Temperature (°C)	Vapor density (g m^{-3})	Tritium (µCi m^{-3})
15	12.8	12.8
20	17.3	17.3
25	23.0	23.0
30	30.4	30.4

* Jerry, Marion, Hornyak "General Physics". Wiley Publisher. Second Edition, 1985.

Table 2 Saturated vapor density in an enclosure and the corresponding tritium concentration when tritium concentration of water is 1 µCi mL^{-1} at different temperatures.

Temp. (°C)	Tritium concentration in water (µCi mL^{-1})	Theoretical tritium in air (µCi m^{-3})	Experimental tritium in air (µCi m^{-3})	Ratio
20.0	1.07	18.5	15.0	0.81
22.5	3.38	68.1	52.5	0.77
20.0	72.5	1460	1170	0.80

Table 3 Theoretical and experimentally measured (using VREC units) tritium concentrations in an enclosure with different tritium concentrations of tritiated water.

	Electret surface voltage						
Electret serial number	Exposure #1	2	3	4	5	6	7
SI4029	750	607	492	355	238	122	20
SI3967	750	607	495	361	248	137	40
SI4032	756	618	508	379	269	159	59
SI4006	749	613	505	379	269	161	65
Exposure duration (days)		1.1424	0.9063	1.0000	1.0000	1.0035	0.976
Average calculated RF		2.1749	2.1786	2.2082	2.1324	2.1008	1.915
Corresponding MPV		681	556	439	312	200	95
Measured tritium conc. (µCi m^{-3})		55.4	55.4	54.2	51.8	51.8	51.8

Table 4 Typical calibration data for 210-mL tritium EIC unit with ST electret.

and sensitivity can also be expressed in derived air concentration (DAC) units, assuming 1 DAC as 25 µCi m^{-3}.

An appropriate unit for making a short, routine laboratory measurement appears to be the SST unit. To cover wider range, an additional unit of low sensitivity such as the SLT or LLT can be placed adjacent to the SST unit. The LLT configuration is useful for measuring very high concentrations or covering very large ranges up to 10,000 µCi m^{-3} d. For measuring very low concentrations the HST is the most appropriate configuration.

Interferences

Because the EIC unit is an integrating ionization chamber, the gamma-radiation background and the airborne radon are the most frequently encountered interferences. Since the response of these devices for these interferences is precisely known, a suitable correction can be applied. A concentration of 1 pCi L^{-1} of radon is equivalent to 1 µCi m^{-3} and is also equivalent to 10 µR h^{-1}.

In a well-ventilated laboratory, the radon concentration is usually low. When the radon concentration is not known, a value of 1 pCi L^{-1} will closely approximate the true radon concentration. Subtraction of 1 µCi m^{-3} from the measured result for this radon interference may be appropriate. Similarly, if there is no known gamma radiation other than natural gamma-ray background which is usually 10 µR h^{-1}, a subtraction of another 1 µCi m^{-3} from the result for this interference may be appropriate. When measuring a tritium concentration of 1 DAC (25 µCi m^{-3}) the subtraction of 2 µCi m^{-3} may be an acceptable interference. However if more exact compensation is necessary for gamma radiation, a similar unit sealed in an airtight bag can be located side by side to subtract the signal from the gamma radiation. Such a compensation method is commonly used with other ion-chamber-based tritium monitors. More accurate compensation for radon requires additional procedures (e.g., measurement of an average voltage drop when tritium is not known to be present).

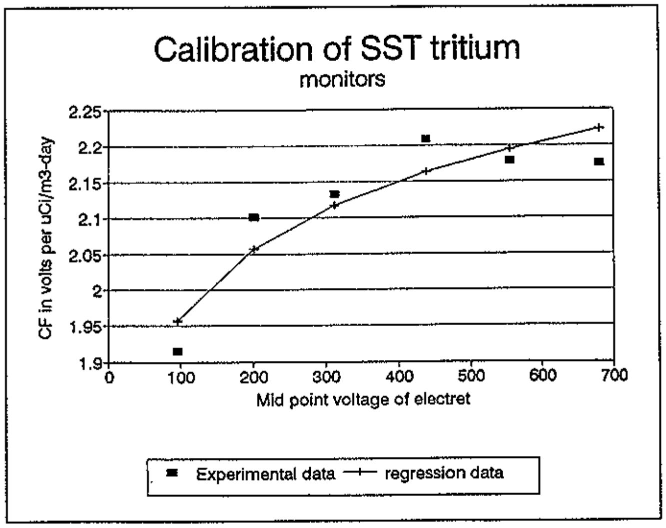

Figure 5 Typical calibration curve for a tritium EIC unit.

MPV (V)	RF(SST)	RF(SLT)	RF(LST)	RF(LLT)	RF(HST)
200	2.0570	0.2178	0.5945	0.05170	7.39
500	2.1805	0.2504	0.6963	0.06055	8.78
700	2.2259	0.2624	0.7337	0.0638	9.29
Range of EIC (μCi m^{-3} d)	250	2200	790	9085	63
Sensitivity (μCi m^{-3} d)	4.6	40	14	161	1.14

Range corresponds to a discharge of fully charged electret from 750 V to 200 V.

Sensitivity is the level that can be measured with an uncertainty of ±15%.

Sensitivity and range can be converted into DAC-day units by dividing the corresponding numbers by 25 (assuming 1 DAC = 25 μCi m^{-3} for tritiated water vapor).

Table 5 Calibration factors for different EIC configurations at MPV of 200, 500 and 700 V. Range and sensitivity.

Routine procedure for making a measurement

Example: using an SST unit to make a tritium measurement in a room.

$$T\left(\frac{\mu\text{Ci}}{\text{m}^3}\right) = \frac{I-F}{D \times \text{RF}} - (R) - (0.09 \times M) \qquad (10)$$

Where:

RF is determined by equations (5) through (9)
D = is the exposure period in days
R = the radon concentration in pCi L^{-1},
M = the gamma-radiation background in μR h^{-1}, and
T = the tritium concentration in μCi m^{-3}

For:

$I = 700$, $F = 650$, $M = 10$, $R = 1$, $D = 2$

Then:

RF = 2.2209
T = 9.4 μCi m^{-3}

Field testing at Pickering Nuclear Facility

Table 6 gives the results of measurements with SST units in comparison with the nominal tritium concentration of the test chamber. Two μCi m^{-3} is subtracted for background. Results generally agree with the expected chamber concentrations. These studies were meant for general verifications and not for rigorous tests. Recently, a further valuation of these devices was published.

Electret ID #	Initial voltage (V)	Final voltage (V)	Exposure period (d)	Measured tritium (μCi m^{-3})	Chamber tritium (μCi m^{-3})
SF7916	736	499	0.2083	513	500
SF9238	717	507	0.2083	454	500
SF9222	727	517	0.2083	454	500
SF7880	481	454	0.2083	57.7	50
SF8085	524	503	0.2083	44.0	50
SF7908	528	503	0.2083	52.1	50
SF7880	604	485	1.0	52.0	50
SF8085	653	529	1.0	54	50
SF7908	657	531	1.0	52.9	50
SF7880	672	609	3.0	7.5	5
SF8085	721	657	3.0	7.6	5
SF7908	715	659	3.0	6.4	5

Table 6 Results of field testing of tritium EIC units at Pickering Nuclear Facility, Ontario Hydro, Canada.

Discussions and conclusions

The EIC units and the associated electret voltage readers have proven to be fieldworthy and robust based on the large scale use of the system for measurement of indoor radon, ambient radon and environmental gamma radiation.[7,8] Nearly 30% of all the radon measuring companies in the United States are using these devices. The EIC units used for the tritium measurement are simply modified versions of the existing units. Therefore, these are expected to be equally fieldworthy and robust. One of the main limitations of the EIC unit is its range and sensitivity. If a higher sensitivity is desired, the range is reduced. If a larger range is desired, the sensitivity is limited. The results from Table 5 in the present work allow the user to choose an optimum device for a particular application.

The EIC units have many advantages which include shock resistance; robustness; humidity and temperature independence; low cost; repeated use of the same device, and simplicity in reading. The EIC units do not have any electronic components.

Electrets have to be handled with care. Their surfaces should never be touched. The surfaces of the electret and the interior of the chambers should be kept clean. This is achieved by cleaning with a jet of air or nitrogen. The manufacturer provides a quality assurance program that can be used with the system. While measuring tritium using EIC units, the interferences from the ambient gamma radiation and radon should be considered. This paper has provided a method of correcting for these interferences.

EIC units are likely to find use in the following applications:

1. Routine measurement of average tritium concentration over a desired period of time in laboratories handling tritium

compounds and in working areas of heavy-water-moderated and cooled nuclear reactors.

2. Routine measurement of stack discharges from reactor stacks and from the heavy water polishing plants.
3. Routine use as a personnel tritium dosimeter.
4. With further development, these can be used for the measurement of tritium in breath of exposed persons.

Acknowledgments

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Note

Certain commercial equipment, instruments, or materials are identified in this paper in order 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.

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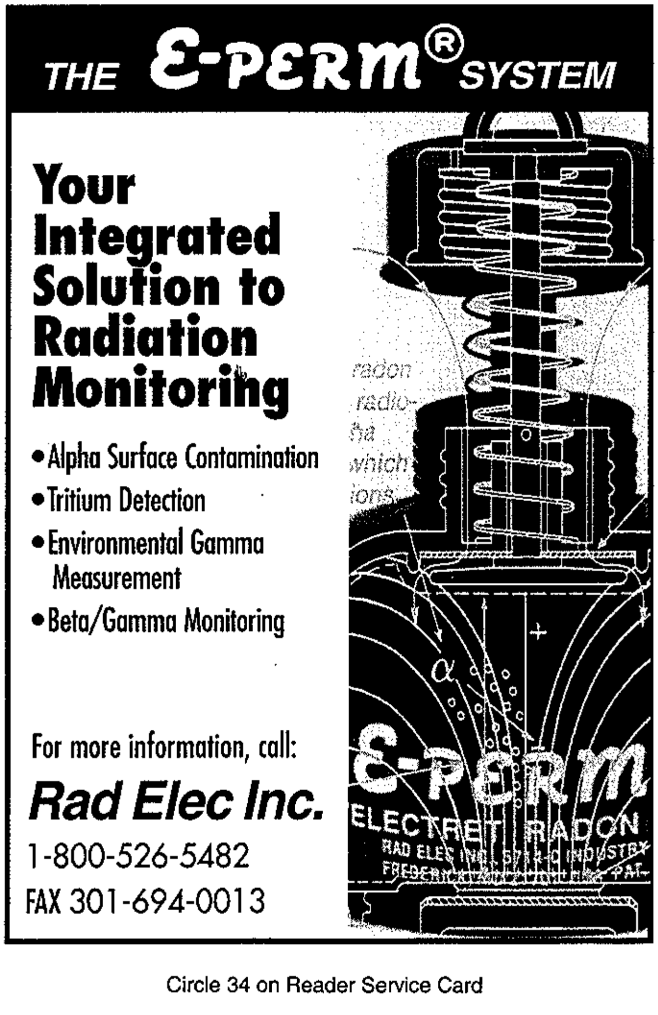

Biographies

Paul Kotrappa
is President of Rad Elec, Inc. He has been responsible for the development of numerous radon and radon progeny monitoring instruments based on electret ion chamber technology. He has served on several IAEA panels focusing on radiation protection and has published more than 100 scientific articles on radiometric measurements. He holds a Ph.D. in radiation biology and biophysics from the University of Rochester, NY, and a M.S. in physics from the University of Mysore, India. He received the 1989 Radiation Industry Award from the American Nuclear Society for designing practical electret ion chamber system.

Rad Elec, Inc.
5714-C Industry Lane
Frederick, Maryland 21701

Thomas G. Hobbs
is Chief of the Health Physics Group at the National Institute of Standards and Technology. He manages a radiation safety program that has encompassed nearly the full spectrum of sources, from high power circular and linear accelerators and research reactors through sealed source irradiators and radiochemistry laboratories, and including laser and microwave facilities. An applied health physicist for over 35 years, he received his M.S. degree in physics from American University and is an American Board of Health Physics Certified Health Physicist.

U. S. Department of Commerce
National Institute of Standards and Technology
Gaithersburg, Maryland 20899

David R. Brown
is a nuclear engineer with the Reactor Health Physics Group at the National Institute of Standards and Technology (NIST). His responsibilities include operational support of the NIST reactor encompassing maintenance, design, and surveillance activities. He is also primarily responsible for the oversight of the reactor health physics counting lab systems. He has over 10 years experience in reactor health physics and radiation detection and measurement and has co-published several papers relating to gamma-ray spectroscopy and radiation measurements. He holds a B.S. degree in Nuclear and Electrical Engineering from the University of Maryland and a Comprehensive Certification in Health Physics from the American Board of Health Physics.

National Institute of Standards and Technology
Reactor Radiation Division
Gaithersburg, Maryland 20899

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