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UNCERTAINTIES EVALUATION FOR ELECTRETS BASED DEVICES USED IN RADON DETECTION

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Received July 24 2004, amended September 15 2004, accepted October 26 2004

In recent years uncertainty evaluation in measurements has achieved great importance. National and international standards offer guidelines to evaluate uncertainties, but these procedures are, until now, not well understood by the operators. This is because of the fact that a detailed uncertainty evaluation is not an easy operation and a standard rule to apply in all cases is not available. Every measurement procedure has its own uncertainty evaluation. In this work, attention is focused upon the electret ion chambers (EIC), widely used in radon concentration measurements. Measurements of gamma radiation sensitivity are performed in a secondary standard calibration laboratory and measurement of radon concentration sensitivity is performed in a radon chamber 0.8 m³ in volume. Raw data are analysed to evaluate the calibration factors and the combined uncertainties are determined. The aim of the work is to give a practical method to assess the uncertainty of a radon measurement.

INTRODUCTION

Electret ion chambers (EIC)⁽¹⁻⁴⁾ can be used both as gamma dosemeters and radon concentration meters. In the first case, the radon contribution to the signal (electret potential lowering) can be suppressed simply by hermetically sealing the detector in a radon-proof bag. In the second case, there is no practical method of suppressing the gamma radiation contribution to the signal. So this contribution is usually transformed into a 'radon equivalent concentration' (REC) arising from background gamma radiation, and represents a noise affecting the signal due to radon concentration. A complete characterisation of these devices requires a measurement both of gamma radiation sensitivity and radon concentration sensitivity. The uncertainty associated with the gamma radiation and radon concentration calibration factors are evaluated according to ISO guidelines.⁽⁵⁾

MATERIALS AND METHODS

Electret ion chambers are supplied by Rad Elec Inc. They are available in eight different configurations. Two different charged Teflon discs, named short-term (ST), with high sensitivity, and long-term (LT) electrets, with low sensitivity, can be associated with four different chambers named D (10 cm³), L (50 cm³), S (200 cm³) and H (1000 cm³). Only the most widely used configurations were tested, that is, S chamber with short-term electret (SST), S chamber with long-term electret (SLT), chamber with long-term electret (LLT) L chamber

with short-term electret (LST) D chamber with short-term electret (DST) and H chamber with short-term electret (HST).

GAMMA IRRADIATION

Electrets, in the six configurations listed above, were irradiated to conventionally true air kerma values. Irradiations were performed in a secondary standard calibration laboratory using a ¹³⁷Cs source and a collimated beam. The standard uncertainty associated with the conventionally true air kerma values is 2%.

Irradiation was organised as shown in Table 1. The air kerma values are selected to obtain a voltage drop of ~40-50 V for each irradiation. The number of irradiations are selected in order to achieve a discharge down to ~180 V. Below this voltage value the ion chamber is out of the saturation range.

Table 1. Organisation of gamma irradiations.

| Configuration | Air kerma delivered (mGy) | Number of measurements useful for data analysis | Number of electrets used |
|---------------|---------------------------|---|--------------------------|
| SST | 0.07 | 33 | 3 |
| SLT | 1 | 27 | 3 |
| LLT | 2.5 | 36 | 3 |
| LST | 0.2 | 36 | 3 |
| DST | 1 | 62 | 5 |
| HST | 0.015 | 54 | 5 |

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1 mGy → 100 mR

Table 2. Organization of radon exposures.

| Configuration | Radon exposure range (kBq · h · m ⁻³) | Number of measurements useful for data analysis | Number of electrets used |
|---------------|---|---|--------------------------|
| SST | 10-100 | 20 | 5 |
| SLT | 80-900 | 45 | 5 |
| LLT | 160-4300 | 36 | 3 |
| LST | 24-1000 | 36 | 3 |
| DST | 440-4300 | 62 | 5 |
| HST | 10-40 | 54 | 5 |

RADON EXPOSURE

Radon exposure was performed in a radon chamber (RC). The radon source is a vial containing ²²⁶Ra salt. Radon is introduced in the RC through a volumetric dosage system that permits a raw assessment of radon concentration. Inside the RC an Alpha-guard PQ2000 works as reference instrument.

The reference value of radon exposure is given by

$$D = \sum_{i=1}^n CRn_i \cdot \Delta t_i, \quad (1)$$

where CRn_i represents the i th radon concentration sampling supplied by the reference instrument. Because the Alpha-guard PQ2000 samples the radon concentration every hour, Δt_i assumes the constant value of 1 h and D is expressed in Bq · h · m⁻³.

Radon exposure was organised as shown in Table 2. As in the gamma radiation case the electrets are discharged down to about 180 V. The voltage drop for each exposure is variable from about ten to about hundred volts, depending on the configuration sensitivity.

The uncertainty associated with a measurement of D is evaluated as the sum of three contributions. The first is a 3% uncertainty associated with the reference instruments and is obtained from the calibration certificate; the second is a statistical uncertainty supplied by the reference instrument together with the measurement; the third is a time uncertainty arising from the operation of electrets' insertion and extraction in the RC.

DATA ANALYSIS

The calibration factor (CF) is defined by

$$CF = \frac{\Delta V}{D}, \quad (2)$$

where ΔV represents the voltage discharge due to irradiation/exposure and D is the air kerma/radon exposure delivered.

It should be noted that CF is a function of the electric field inside the chamber. In order to assess a functional dependence, the following relation is used⁽⁶⁾.

$$CF = q + m \cdot \ln\left(\frac{V_i + V_f}{2}\right) = q + m \cdot X, \quad (3)$$

where V_i and V_f are the initial and final electret potentials; q and m are fitting parameters that will be determined experimentally.

The relative variance associated with CF given by (2) is expressed by the following:

$$u_{\text{rel}}^2(CF) = u_{\text{rel}}^2(\Delta V) + u_{\text{rel}}^2(D), \quad (4)$$

The first term on the right-hand side of (4) is associated only with the voltage measurement procedure and is evaluated as follows:

$$\Delta V = (V_i - V_0) - (V_f - V_0), \quad (5)$$

where V_0 is the reference potential that in our case is set equal to zero. It is our experience that repeated measurement of the same electret, made by different operators can give a spread of readings up to 3 V. For this reason, we assess the uncertainty assuming, for the stochastic variable, a rectangular distribution 3 V wide. The uncertainty is given as

$$u(V) = \frac{1.5}{\sqrt{3}}, \quad (6)$$

The uncertainty associated with V_0 is obtained following the same argument described above; the only difference is that the rectangular distribution is assumed to be 1 V wide, that is, the instrumental resolution of voltage reader. The uncertainty is obtained by

$$u(V_0) = \frac{0.5}{\sqrt{3}}, \quad (7)$$

The variance associated with ΔV is obtained by

$$u^2(\Delta V) = u^2(V_i) + u^2(V_f) + 2 \cdot u^2(V_0), \quad (8)$$

The second term on the right-hand side of (4) is associated only with the exposure procedures and represents an uncertainty contribution common to all measurements. The mathematical instrument used to perform the best fit of (3) is based on a weighed least squares method⁽⁷⁾. According to the method the vector a , containing the fitting parameters, is given by

$$a = \begin{bmatrix} q \\ m \end{bmatrix} = [T^t \cdot V^{-1} \cdot T] \cdot T^t \cdot V^{-1} \cdot C, \quad (9)$$

where T is a vector containing the experimental X values, C is a vector containing the experimental CF values and V is the variance-covariance matrix associated with C .

1000 Bq / m³ · h
= 1.13 Bq / m³ · day

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The variance-covariance matrix M associated with a is given by

$$M = \begin{bmatrix} u^2(q) & u(q, m) \\ u(m, q) & u^2(m) \end{bmatrix} = [T^t \cdot V^{-1} \cdot T]^{-1}, \quad (10)$$

The V matrix is built in the following way:

$$\begin{bmatrix} u^2(CF_1) + c^2 & u^2(CF_{1,2}) + u(D_{1,2}) & u(D_{1,3}) & \dots & u(D_{1,n}) \\ u^2(CF_{2,1}) + u(D_{2,1}) & u^2(CF_2) + c^2 & u^2(CF_{2,3}) + u(D_{2,3}) & \dots & \dots \\ u(D_{3,1}) & u^2(CF_{3,2}) + u^2(D_{3,2}) & \dots & \dots & u(D_{n-2,n}) \\ \dots & \dots & \dots & \dots & u^2(CF_{n-1,n}) + u(D_{n-1,n}) \\ u(D_{n,1}) & u(D_{n,2}) & \dots & u^2(CF_{n,n-1}) + u(D_{n,n-1}) & u^2(CF_n) + c^2 \end{bmatrix}.$$

In the main diagonal there are the variances associated with experimental CF values plus a constant c . The constant c enters in the calculation of χ^2 and is assessed to obtain a value of that is χ^2 lower than the number of degree of freedom. In the element out of the mean diagonal there are the covariance elements. In particular, the elements just beside the mean diagonal are evaluated as the sum of two components. The first reflects the correlation of two consecutive ΔV measurements. The argument employed to assess this correlation starts from the consideration that in consecutive exposures of the same electret the final voltage of n th exposure agrees with the initial voltage of $(n+1)$ th exposure. So the potential common to both exposures is considered as a correlation element. In order to quantify the correlation the following mathematical model is used:

$$CF_{n+1,n} = CF_{n,n+1} = \frac{V_{\text{common}} + V_0}{D}, \quad (11)$$

$CF_{n,n+1}$ has no physical meaning and is introduced only to have an analytical expression to calculate $u^2(CF_{n,n+1})$ that represents the correlation element:

$$\begin{aligned} u^2(CF_{n,n+1}) &= u^2(CF_{n+1,n}) \\ &= \frac{u^2(V_{\text{common}}) + u^2(V_0)}{D^2}, \end{aligned} \quad (12)$$

Note that in Equation 12 D is not considered a stochastic variable because its contribution is considered in the second term.

The second term in the elements beside the mean diagonal is equal to the terms in leftover elements of the matrix. These terms are introduced because the uncertainty associated with air kerma/radon exposure is a common uncertainty. They are evaluated, according to Equation 13 by taking the relative variance associated with air kerma/radon exposure multiplied by the corresponding CF. In the radon exposure, only the uncertainty associated with the

reference instruments is considered a common uncertainty.

$$\begin{aligned} u(D_{ij}) &= CF_i \cdot u_{\text{rel}}(D_i) \cdot CF_j \cdot u_{\text{rel}}(D_j) \\ &= \frac{\Delta V_i}{D_i^2} \cdot u(D_i) \cdot \frac{\Delta V_j}{D_j^2} \cdot u(D_j), \end{aligned} \quad (13)$$

The M matrix allows one to express the uncertainty associated with the calibration factor CF by the following analytical form:

$$\begin{aligned} u(\text{CF}) &= \sqrt{u^2(q) + X^2 \cdot u^2(m) + 2X \cdot u(q, m) + m^2 \cdot u^2(X)} \end{aligned} \quad (14)$$

RESULTS

The arguments explained above permit one to obtain a measurement of the calibration factor and the associated uncertainty, both for radon concentration and for gamma radiation. In Table 3 the values obtained for the electrets used as gamma dosimeters are listed. In Table 4 the same values are listed for electrets used as radon concentration meters.

In radon concentration measurements, a correction for gamma background must be introduced. For this purpose the following formula is used:

$$E_{Rn} = \frac{\Delta V_{Rn}}{CF_{Rn}} = \frac{\Delta V - \Delta V_\gamma}{CF_{Rn}}, \quad (15)$$

where E_{Rn} represents the integrated concentration, and the voltage drop (ΔV) is split into two contributions, the first due to radon (ΔV_{Rn}), the other due to gamma radiation (ΔV_γ). The second contribution can be assessed by (2). Equation (15) can thus be rewritten as follows:

$$E_{Rn} = \frac{\Delta V}{CF_{Rn}} - \frac{CF_\gamma}{CF_{Rn}} \cdot D = \frac{\Delta V}{CF_{Rn}} - C \cdot D, \quad (16)$$

where C represents the radon equivalent concentration of the gamma contribution. In Table 5 the values of C for the studied configurations are listed.

Table 3. Fitting parameters of the weighted least squares method for exposures to gamma radiation.

| | DST | HST | LLT | LST | SLT | SST |
|---|-------|--------|---------|--------|-------|--------|
| Degree of freedom | 60 | 52 | 34 | 34 | 25 | 31 |
| q (V Gy ⁻¹) | 27.65 | 941.72 | 10.2641 | 98.95 | 32.02 | 295.58 |
| m (Gy ⁻¹) | 1.7 | 392.49 | 1.2622 | 18.91 | 5.25 | 77.75 |
| $u^2(q)$ (V ² Gy ⁻²) | 5.84 | 65194 | 3.2997 | 214.11 | 23.02 | 2910.5 |
| $u^2(m)$ (Gy ⁻²) | 0.145 | 1813 | 0.0882 | 6.4 | 0.61 | 81.4 |
| $u(q, m)$ (V Gy ⁻²) | -0.87 | -10469 | -0.5271 | -35.4 | -3.59 | -464.8 |
| c (V Gy ⁻¹) | 0.66 | 95 | 0.45 | 0.2 | 1.35 | 18 |
| χ^2 | 59.4 | 51.69 | 31.4 | 32.49 | 23.57 | 30.9 |

Table 4. Fitting parameters of the weighted least squares method for exposures to radon concentration.

| | DST | HST | LLT | LST | SLT | SST |
|--|-----------|-----------|-----------|-----------|-----------|-----------|
| Degree of freedom | 46 | 18 | 54 | 42 | 43 | 18 |
| q (V Bq ⁻¹ h ⁻¹ m ³) | 2.47E-05 | -5.33E-03 | 8.40E-06 | 2.69E-04 | 7.16E-05 | 1.09E-03 |
| m (Bq ⁻¹ h ⁻¹ m ³) | 1.03E-06 | 2.73E-03 | 3.33E-06 | 1.76E-05 | 1.60E-05 | 1.82E-04 |
| $u^2(q)$ (V ² Bq ⁻² h ⁻² m ⁶) | 1.13E-11 | 8.72E-06 | 1.12E-11 | 3.02E-09 | 8.55E-10 | 1.96E-07 |
| $u^2(m)$ (Bq ⁻² h ⁻² m ⁶) | 2.98E-13 | 2.68E-07 | 3.04E-13 | 8.17E-11 | 2.38E-11 | 5.64E-09 |
| $u(q, m)$ (V Bq ⁻² h ⁻² m ⁶) | -1.76E-12 | -1.52E-06 | -1.78E-12 | -4.84E-10 | -1.40E-10 | -3.28E-08 |
| c (V Bq ⁻¹ h ⁻¹ m ³) | 1.03E-6 | 6.10E-4 | 1.00E-5 | 2.25E-5 | 9.2E-6 | 1.03E-4 |
| χ^2 | 45.5 | 17.49 | 52.6 | 41.2 | 41.6 | 17.1 |

Table 5. C values for gamma radon equivalent concentration.

| Configuration | C (Bq h m ⁻³ nGy ⁻¹) | Standard uncertainty |
|---------------|---|----------------------|
| SST | 0.335 ± 8% | 0.016 |
| SLT | | 0.018 |
| LST | | 0.024 |
| LLT | 0.595 ± 2.9% | 0.029 |
| HST | 0.27 ± 8.9% | 0.016 |
| DST | 1.20 ± ? | 0.050 |

Final agreement

associated with gamma background measurement are considered. Figure 2 shows, for the LLT configuration, the relative uncertainty as a function of integrated radon concentration. The relative standard uncertainty associated with gamma background is assessed as 20%. Different values of gamma background are considered. Figures 1 and 2 simply plot Equation 17.

CONCLUSION

Equations for an evaluation of the uncertainty can be used both for assessing an uncertainty of the measurements and to plan the measurement campaign. The operator can use the formula (17) without other consideration starting from the value of gamma background and its uncertainty. This information can be derived from the calibration certificate of the instrument used for gamma background measurement. In other cases they can be derived from published data of the mean gamma background in the site under study.

A prediction of expected uncertainty is useful to evaluate the degree of knowledge of background gamma radiation, and so to assist in using the short-cut method to perform the measurement without compromising the quality of results.

The final expression of uncertainty is given by (17). It can be noted that uncertainty is a function of the gamma background value, uncertainty associated with gamma background value and ΔV .

$$u^2(E_{Rn}) = \frac{u^2(\Delta V)}{CF^2} + \frac{\Delta V^2}{CF^4} \cdot u^2(CF_{Rn}) + D^2 \cdot u^2(C) + C^2 \cdot u^2(D), \quad (17)$$

Figure 1 shows, for the LLT configuration, the relative uncertainty as a function of integrated radon concentration. The gamma background is assessed as 150 nGy h⁻¹. Different uncertainties

It → 0.28
 0.28

$$1 \frac{Bq}{m^3} \cdot h \rightarrow \frac{1}{37} \frac{pCi}{L} \cdot \frac{1}{24} d^{-1} \\ \approx 0.001126 \frac{pCi}{L} \cdot d$$

4
 $D \rightarrow 0.28 \frac{pCi}{L} \cdot \frac{1}{24} d^{-1} \cdot 0.35 \frac{Bq}{m^3} \text{ per } \frac{nGy}{h}$
 for 100 nGy/h → 35 Bq/m³ (≈ 0.95 pCi/L)

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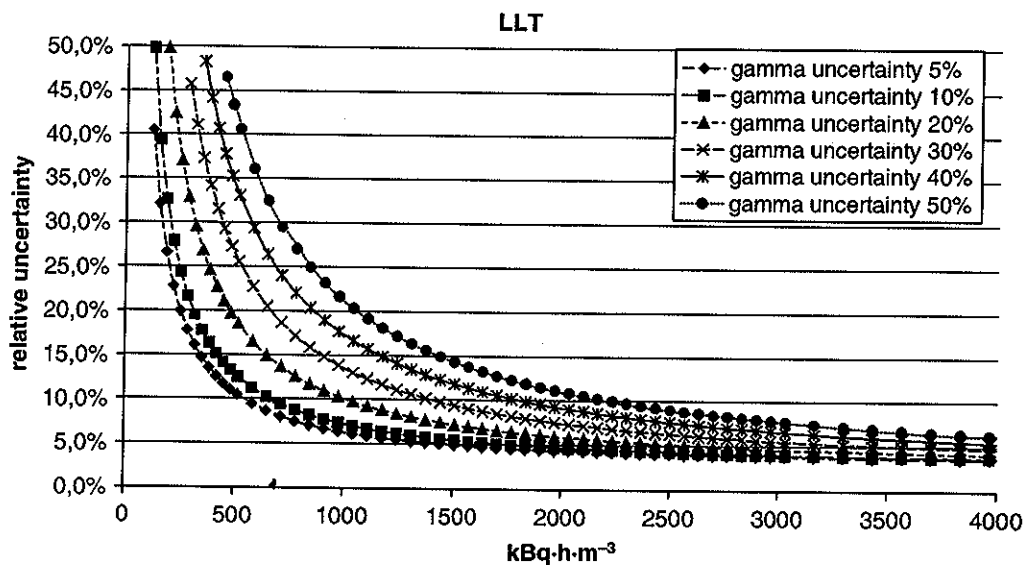


Figure 1. LLT configuration. Relative uncertainty associated with radon exposure measurement as a function of the measured radon exposure. The gamma background is assumed to be 150 nGy h^{-1} . The gamma background relative uncertainty is assumed to range from 5% to 50%.

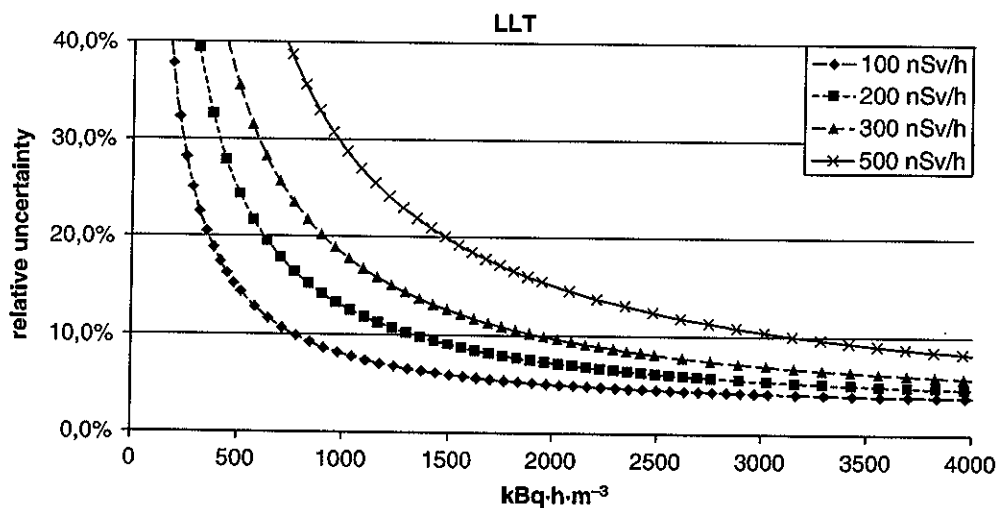


Figure 2. LLT configuration. Relative uncertainty associated with radon exposure measurement as a function of the measured radon exposure. The gamma background is assumed to range from 100 to 500 nGy h^{-1} . Uncertainties associated with gamma background are assumed to be 20%.

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