9. Conclusions and perspectives

9.1. Concluding remarks

This study, which deals with the problematical determination of long-term indoor ^{222}Rn progeny equilibrium factor using NTDs to well assess the annual effective dose in privates homes and in workplaces, has led to the following conclusions:

1. The review of the most relevant parameters and processes affecting indoor ^{222}Rn and ^{220}Rn progeny concentrations has shown that their behaviour is very complex and that the equilibrium factor may change significantly from one house to another, depending on the geometry of the house, on the aerosol concentration, on the air mass movement, on the ambient conditions and on the inhabitants habits. Therefore, there is the need of measuring the long-term equilibrium factor indoors.

2. A detailed study of the measurement principles of airborne ^{222}Rn, ^{220}Rn and their progeny by means of NTDs, taking into account the range of variation of the parameters influencing their concentration, has shown that it is not possible for the existing methods to obtain the long-term equilibrium factor with an appropriate accuracy.

3. A new approach for long-term equilibrium factor determination from the measurement of airborne ^{222}Rn and its α-emitter daughters is presented in this PhD dissertation. This approach is based on the new concept of reduced equilibrium factor (F_{red}), which is defined as $F_{\mathrm{Red}} = \frac{0.105\,C_{^{218}\mathrm{Po}}+0.380\;C_{^{214}\mathrm{Po}}}{C_{^{222}\mathrm{Rn}}}$. We have shown that the equilibrium factor can be obtained with the best precision if proper optimisation of experimental conditions for the F_{red} measurement by means of NTDs is performed. In this method, assumptions about ventilation, aerosol attachment and deposition (attached and unattached) rates are not necessary.

4. We have designed a new passive, integrating and multi-component dosimeter to measure simultaneously the individual airborne concentration of ^{222}Rn, ^{220}Rn, ^{218}Po and ^{214}Po. It consists of: i) two Makrofol detectors, namely detectors A and B, which are

enclosed within two diffusion chambers — each one with different filter membrane — to measure indoor ^{222}Rn+^{220}Rn and ^{222}Rn, together with ii) two Makrofol detectors (C and D) that are kept in direct contact with air and that are electrochemically etched at different conditions to obtain the airborne ^{218}Po and ^{214}Po concentrations. The measurement method is based on the fact that the half-lives of ^{222}Rn and ^{220}Rn are different, that both isotopes have the same diffusion coefficient in a given medium and that the response of the Makrofol detector depends on the electrochemical etching conditions used.

5. From the slowing down spectrum of α-particles emitted by the airborne ^{222}Rn, ^{220}Rn and their progeny, and in order to avoid the plate-out peaks of these last, two α-energy windows of interest are chosen, one from 3.0 to 5.0 MeV for the detector A, B and C and another one from 6.3 to 7.5 MeV for the detector D. With these α-energy windows, the detector B lets the measurement of ^{222}Rn concentration. The concentration of ^{220}Rn can be obtained as a response difference of the detectors A and B. The reading of detector D allows the determination of the airborne ^{214}Po concentration. From this quantity and the information given by the detector C the airborne ^{218}Po concentration can be determined.

6. We have developed a Monte-Carlo computer code, called SIMAR, to obtain the sensitivity of each Makrofol detector, taking into account: (1) the Bethe-Bloch expression for the stopping power of heavily charged particles in a medium, (2) the behaviour of ^{222}Rn, ^{220}Rn and their progeny in the open air and within the diffusion chamber, and (3) the α-energy window response of each detector. The estimated sensitivity values have been validated by reproducing the response of an ideal detector, both in the free air and enclosed within a diffusion chamber.

7. The semi-automatic track counting system has been improved without any excessive cost, by connecting a photo video camera of an optical field area of 8.4×6.3 mm^2 to a digital TV-graphic card and using a public domain Java image processing software, called ImageJ, for track analysis.

8. We have performed the initial phase of constructing a small exposure chamber, for both ^{222}Rn and ^{220}Rn, and we have set up an irradiation device to generate mono-energetic α-particles from 2 MeV up to 8 MeV with an α-energy resolution lower than 10%.

9. By studying the ^{222}Rn diffusion through some of the commercially available filters, we have shown that the glass fiber and the polyethylene are very appropriate for the

detectors A and B, respectively, to perform separate measurement of indoor ^{222}Rn and ^{220}Rn concentration.

10. We have confirmed experimentally using the irradiation device that the electrochemical etching conditions for the detectors A, B and C to generate an α-energy window response of [3.0 - 5.0] MeV are:

Etchant	:	KOH **6** M mixed with 50% ethanol
Temperature	:	40 ^{o}C
Pre-etching duration	:	**4** h
Frequency	:	3 kHz
Electric field strength	:	33 kV cm^{-1}
ECE duration	:	**1.5** h

11. A detailed study of the main parameters influencing the electrochemical etching process of the Makrofol detectors have shown that the optimal etching conditions for the detector D to generate an α-energy window response of [6.3 - 7.5] MeV are:

Etchant	:	KOH **7.5** M mixed with 50% ethanol
Temperature	:	40 ^{o}C
Pre-etching duration	:	**6** h
Frequency	:	3 kHz
Electric field strength	:	33 kV cm^{-1}
ECE duration	:	**1** h

12. The detectors A and B have been calibrated in pure ^{222}Rn atmospheres showing identical, consistent, and reproducible responses. The experimental sensitivity obtained for these detectors is very close to that given by the Monte-Carlo simulation.

13. With our passive, integrating and multi-component dosimeter, the a priori lower limit of detection can be estimated only for ^{222}Rn. The minimum detectable ^{222}Rn concentration is equal to 10 Bq m^{-3} for an eventual exposure time of 90 days.

14. By using well-control exposures in a reference laboratory, we have shown that the equilibrium factor values determined with our system agree with those obtained by active methods.

15. The results of an application indoors of our dosimeter in an inhabited Swedish single-family house suggest the usefulness of the method used in this study to carry out routine surveys for ^{222}Rn level measurements in private homes and in workplaces in order to estimate the associated annual effective dose received by the general public and the workers.

9.2. Future outlooks

In this PhD dissertation a novel approach has been proposed for long-term equilibrium factor determination from the measurement of ^{222}Rn and its α-emitter progeny (^{218}Po and ^{214}Po), and, therefore, new implications for future works have been opened. The main perspectives of this study are:

1. The sensitivity of the detector A in front of ^{220}Rn should be improved in order to extent its detectability to concentrations of the same order as those of ^{222}Rn. Further investigations of the parameters affecting the response of the other detectors should be also performed to optimise the system precision and to determine the sources or causes of errors.

2. A series of calibration exercices must be carried out in well-controlled ^{222}Rn and ^{220}Rn exposure facilities to complete the experimental determination of the sensitivities of the detectors A, B, C and D with respect to ^{222}Rn, ^{220}Rn and their α-emitter decay products. In addition, the response of our dosimeter at different equilibrium factors, ambient relative humidities and temperatures should be studied and evaluated.

3. The concept of the reduced equilibrium factor introduced in this work offers a lot of possibilities for the design and the development of new methods based on active or passive detectors for ^{222}Rn progeny equilibrium factor measurement.

4. With the passive integrating system set up in this study, it will be of great interest to carry out a national survey, in private homes and workplaces, in order to estimate the annual effective dose due to inhalation of indoor ^{222}Rn daughters and to identify those sites with high concentrations of ^{220}Rn.

A. Recoil energy determination from α-decay

Most of the heavy nuclei are energetically unstable against the spontaneous emission of a mono-energetic α-particle (or ^{4}He nucleus). The α-decay process can be written schematically as

$$^{A}_{Z}X \longrightarrow ^{A-4}_{Z-2} Y +^{4}_{2} \alpha \tag{A.1}$$

where X and Y are the initial and the final nuclear species, A is the mass number of the nuclei X and Z its atomic number. For each distinct transition between initial and final nucleus, a fixed energy difference or Q-value characterises the α-decay as follows

$$Q = (m_X - m_Y - m_\alpha)c^2 \tag{A.2}$$

where m_X, m_Y and m_α are the mass of the nuclei X, Y and the α-particle, respectively, and c is the light celerity.

In general, the kinetic energy of α-particle is usually lower than Q-value because of the recoil energy carried out by the residual nuclei Y. Thus, by applying the mass-conservation law, the kinetic energy of both α-particle and residual nuclei are given by

$$E_\alpha = \frac{m_Y}{m_Y + m_\alpha} Q = \frac{A-4}{A} Q \text{ and } E_Y = \frac{m_\alpha}{mY + m_\alpha} Q = \frac{4}{A} Q \tag{A.3}$$

As the α-particle energy is a well known quantity for all the α-emitter radionuclide, the recoil energy can be estimated by the following expression

$$E_Y = \frac{4}{A-4} E_\alpha \tag{A.4}$$

Table A.1 summarises the recoil energy of interest in the ^{222}Rn and ^{220}Rn chains.

Table A.1. The recoil energy of interest in the ^{222}Rn and ^{220}Rn chains.

X	E_α (MeV)	Y	E_Y (keV)
^{226}Ra	4.77	^{222}Rn	86
^{222}Rn	5.49	^{218}Po	101
^{218}Po	6.00	^{214}Pb	112
^{224}Ra	5.69	^{220}Rn	103
^{220}Rn	6.29	^{216}Po	117
^{216}Po	6.78	^{212}Pb	128
^{212}Bi	6.07	^{212}Po	115

B. Stopping power and range of α-particle in a medium

It is well known that the heavy ion (~8000 times the mass of the electron), when passing through matter loses, its energy predominantly via electronic interactions with the absorber atoms. The energy transferred in such collisions per unit of path length can be obtained from the well known Bethe-Bloch formula as follows (Bethe, 1930; Bloch, 1933)

$$(-\frac{\mathrm{d}E}{\mathrm{d}x})\ (\mathrm{MeV\ cm^{-1}}) = \frac{nZ_{\mathrm{eff}}^2 e^4}{4\pi\varepsilon_0^2 m_{\mathrm{e}} v^2}\left[\ln\frac{2m_{\mathrm{e}}v^2 w_{\mathrm{max}}}{I^2(1-\beta^2)} - 2\beta^2 - \delta - U\right] \tag{B.1}$$

where Z_{eff} is the effective charge of the heavy ion, v is its velocity, e is the electron charge, n is the electronic density of the absorber medium, ε_0 is the free space permittivity, m_{e} is the electron mass, I is the ionisation potential of the medium, β is the ion velocity relative to that of the light, w_{max} is the maximum energy transferred to the medium atoms, δ is a correction factor for polarisation of the medium, and U is a term that takes into account the participation of inner electron shells.

The most direct application of energy loss data is the determination of the ion ranges in medium materials. These last are regarded as having well defined ranges usually approximated to a straight line. The range of α-particles in a given medium can be calculated from the following integral

$$R(E) = \int_0^E (-\frac{dE}{dx})^{-1}\mathrm{d}E \tag{B.2}$$

where E (MeV) is the ion energy. According to this equation, theoretical calculation of ion ranges within medium materials is not trivial. Instead, semi-empirical expressions, based on experimental data and guided by theory, are usually used.

In this study, we used the Srim-2000 code as a reference for the stopping power and range calculation. The range-energy results obtained for a given medium are fitted using the least-square minimisation method to establish the range-energy relationship. Figure

B.1 presents the range-energy data obtained for α-particles in Makrofol and in air using the Srim-2000 code. As shown in this figure, the α-particle range-energy relationships in both Makrofol and air are polynomial and the corresponding least-square fitting parameters obtained are

$$R_{\text{Makrofol}}(E) = 1.05 + 2.50E + 0.666E^2 \ (\mathrm{R}^2 = 0.998) \tag{B.3}$$

$$R_{\text{air}}(E) = 0.12 + 0.297E + 0.07E^2 \ (\mathrm{R}^2 = 0.998) \tag{B.4}$$

where the α-energy, E, is given in MeV and its corresponding range in Makrofol and in air are respectively given in μm and in cm. The uncertainties introduced by the fit were found to be less than 1%. These results differ from those obtained in a previous work of our group (Amgarou et al., 2001b) in which the Makrofol and air range-energy data calculated by the Srim-2000 code were adjusted to the common power function of the form $R(E) = aE^b$, suggested by the Bragg's rule and denoted by a dot line in Figure B.1. Nevertheless, as can be clearly seen in this figure, the polynomial adjustment is much more better for the Srim-2000 energy-range data than the function $R(E) = aE^b$.

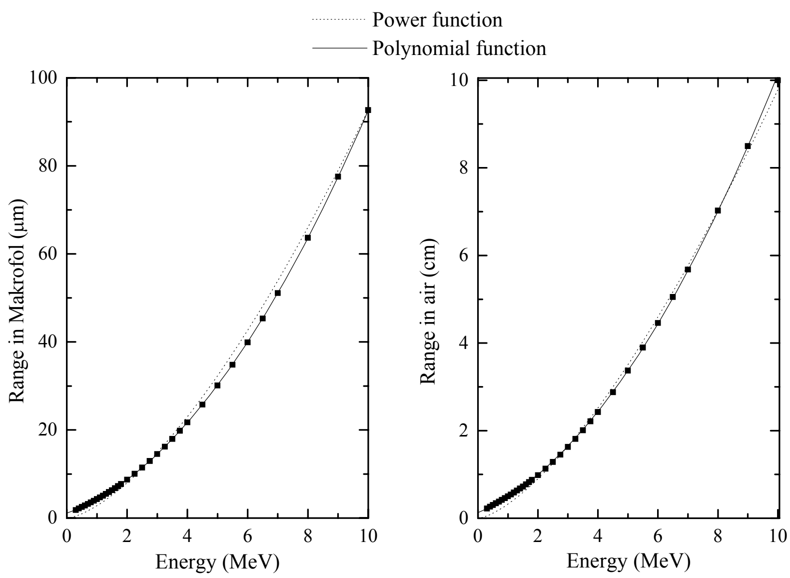

Figure B.1. Range-energy dependence for α-particles in Makrofol and in air.

C. A note on Monte-Carlo simulation

The name *Monte-Carlo* was applied for the first time by scientists working on the nuclear weapon project in Los Alamos, during the Second World War, to design a class of numerical methods based on the use of random numbers. A good review of the Monte-Carlo techniques and their application to simulate the physical systems could be found in Kalos and Whitlock (1986). The essential ingredient of the Monte-Carlo techniques is the numerical sampling of random variables with specified probability distribution functions (PDFs), which are positive function normalised to unity. Thus, considering a variable x that is randomly distributed in the interval (a, b) according to a given PDF, $p(x)$, we have

$$p(x) \geq 0 \text{ and } \int_a^b p(x)\ \mathrm{d}x = 1 \tag{C.1}$$

The cumulative distribution function of x is defined by

$$P(x) = \int_a^x p(x)\ \mathrm{d}x \tag{C.2}$$

This function increases monotonically from $P(a) = 0$ to $P(b) = 1$ and, therefore, has an (univaluate) inverse function. The transformation $\xi = P(x)$ defines a new random variable, which takes values uniformly distributed in the interval $(0, 1)$; so that,

$$\xi = \int_a^x p(x)\ \mathrm{d}x \tag{C.3}$$

This equation is referred to as the sampling equation of the variable x. This procedure for random sampling is known as the inverse transform method; it is particularly adequate for PDFs, given by simple analytical expressions, such that the sampling equation can be solved analytically.

In general, random sampling algorithms are based on the computer generation of a pseudo-random number sequence between zero and unity from a given *seed* using a linear congruential method. The name pseudo reflects the fact that the generated sequence is not truly random, since it is obtained from a deterministic algorithm. In this study, as a pseudo-random number generator algorithm we employed the subroutine RANDOM_NUMBER that is included in the Fortran 90 Scientific Function Package. This subroutine produces 32-bit floating point numbers uniformly distributed in the interval $(0, 1)$ and have a periodic sequence of the order of 10^{18}, which is virtually infinite for practical simulations.

Consider, for instance, the example of particle emission from a given radioactive source. The random numbers are then used to choose the coordinates and the direction of emission. By supposing that the variable Cartesian coordinate x is uniformly distributed in the interval (a, b), we can write

$$p(x) = \frac{1}{b-a} \tag{C.4}$$

Then, the sampling equation (C.3) leads to the well-known sampling formula

$$x = a + \xi(b-a) \tag{C.5}$$

On the other hand, in order to simulate an isotropic particle emission (i.e., in all directions) from a radioactive source, the following angular distribution probability density is adopted

$$p(\theta, \varphi)\ \mathrm{d}\theta\ \mathrm{d}\varphi = \frac{1}{4\pi} \sin\theta\ \mathrm{d}\theta\ \mathrm{d}\varphi \tag{C.6}$$

where θ and φ are, respectively, the zenith and azimuthal angles. Notice that $0 \leq \theta \leq \pi$ and $0 \leq \varphi \leq 2\pi$. Since these variables are independent, the term $p(\theta, \varphi)$ can be factorise as the product of two associated PDFs $f(\theta)$ and $g(\varphi)$, which are given by

$$f(\theta)\mathrm{d}\theta = \frac{1}{2} \sin\theta\ \mathrm{d}\theta \text{ and } g(\varphi)\ \mathrm{d}\varphi = \frac{1}{2\pi} \tag{C.7}$$

By choosing two independent random values, ξ_1 and ξ_2, with uniform probability between 0 and 1, the intial direction of emission of a particle from an isotropic source can be generated by imposing that

$$\xi_1 = \int_0^\theta f(\theta)\mathrm{d}\theta \text{ and } \xi_2 = \int_0^\varphi g(\varphi)\mathrm{d}\varphi \tag{C.8}$$

Solving these integrals, we finally obtain

$$\theta = \arccos(1 - 2\xi_1) \tag{C.9}$$

$$\varphi = 2\pi\xi_2 \tag{C.10}$$

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