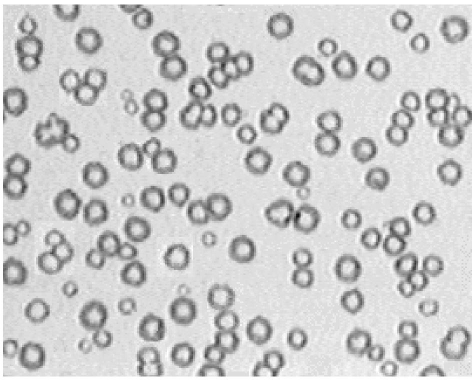
Universitat Autònoma de Barcelona
Grup de Física de les Radiacions

Long-Term Measurements of Indoor Radon and its Progeny in the Presence of Thoron Using Nuclear Track Detectors: A Novel Approach



Khalil Amgarou

PhD Thesis

April, 2002

Abstract

In this work, we establish a novel approach for long-term determination of indoor ^{222}Rn progeny equilibrium factor, even in the presence of ^{220}Rn, using a passive, integrating and multi-component system of nuclear track detectors. The 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 nuclear track detectors depends on the electrochemical etching conditions used. The new dosimeter set up for this purpose 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 sensitivities of each Makrofol detector in front of ^{222}Rn, ^{220}Rn and their decay products have been theoretically obtained, using Monte-Carlo technique 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 computer code used for the calculation has been validated by reproducing the response of an ideal detector, both in the free air and enclosed within a diffusion chamber.

A detailed experimental study has been carried out in which we have determined the optimum filters for ^{222}Rn/^{220}Rn separation and the appropriate electrochemical etching conditions for each of the Makrofol detector used. Otherwise, we have calibrated the detectors A and B in pure ^{222}Rn atmospheres and we have confirmed experimentally that the equilibrium factor values determined with our system agree with those obtained by active methods.

Finally, as a pilot test, several dosimeters were exposed in an inhabited Swedish single-family house. The results of this exposure suggest the usefulness of this method to perform routine surveys for ^{222}Rn level measurements in private homes and in workplaces in order to estimate the annual effective dose received by the general public and the workers.

Resumen

En este trabajo, hemos establecido una nueva aproximación para determinar el factor de equilibrio de los descendientes de ^{222}Rn, durante periodos largos de tiempo y en presencia de ^{220}Rn, utilizando un sistema pasivo, integrador y multicomponente de detectores de trazas nucleares. El método se basa en el hecho de que los periodos de semi-desintegración del ^{222}Rn y del ^{220}Rn son diferentes, de que ambos isótopos tienen el mismo coeficiente de difusión en un medio dado, y de que la respuesta de los detectores de trazas nucleares depende de las condiciones del revelado electroquímico utilizadas. El dosímetro puesto a punto consta de: i) dos detectores de Makrofol, designados como detectores A y B, que están situados en el interior de dos cámaras de difusión — cada una con un filtro diferente — para medir ^{222}Rn+^{220}Rn y ^{222}Rn, además de ii) dos detectores de Makrofol (C y D) que están en contacto directo con el aire y a los cuales se les aplica diferentes condiciones de revelado electroquímico para obtener las concentraciones del ^{218}Po y del ^{214}Po.

Las sensibilidades de cada detector de Makrofol con respecto al ^{222}Rn, ^{220}Rn y progenie se han calculado teóricamente utilizando la técnica de Monte Carlo, teniendo en cuenta: (1) la ecuación de Bethe-Bloch para determinar el poder de frenado de partículas cargadas pesadas en un medio material, (2) las propiedades y el comportamiento del ^{222}Rn, ^{220}Rn y descendientes en el aire libre y dentro de una cámara de difusión, y (3) el rango de energías de partículas α que registra cada detector. El código desarrollado para estos cálculos se ha validado reproduciendo la respuesta de un detector ideal, tanto en el aire libre como dentro de una cámara de difusión.

Se ha llevado a cabo un estudio experimental detallado en el que se han determinado los filtros óptimos para la separación ^{222}Rn/^{220}Rn y las condiciones de revelado electroquímico apropiadas para cada detector. Asimismo, hemos calibrado los detectores A y B en atmósferas puras de ^{222}Rn y hemos confirmado experimentalmente que los valores del factor de equilibrio determinados con nuestro dosímetro están en buen acuerdo con los obtenidos mediante sistemas activos.

Finalmente, como experiencia piloto, varios dosímetros han sido expuestos en una casa unifamiliar habitada de Suecia. Los resultados de esta exposición indican la utilidad de nuestro método para llevar a cabo campañas de medida de los niveles de ^{222}Rn, en lugares de trabajo y en casas privadas, a fin de estimar la dosis efectiva anual recibida por los trabajadores y por el público en general.

Acknowledgements

Desitjo expressar el meu sincer reconeixement a la Dra. Carmen Baixeras i al Dr. Lluís Font, directors d'aquesta Tesi, per la seva inesgotable paciència, per les seves encertades orientacions i, sobre tot, per l'estímul diari tant a nivell científic com a nivell personal.

Al Prof. Francisco Fernández per fer-me aquella trucada telefònica a Tanger, per haver-me acollit dins el Grup de Física de les Radiacions i pel seu suport en molts aspectes.

Al Prof. Agustin Alonso, de la Universidad Complutense de Madrid, por haber aceptado ser presidente del tribunal en el cual han aceptado, también, formar parte los Doctores Eugenio Gil, del Consejo de Seguridad Nuclear, y Joan Albert Sánchez Cabeza, del Grupo de Física de las Radiaciones.

Egalement, je remercie le Prof. Jean Gasiot, de l'Université de Montpellier, pour vouloir prendre part avec les autres membres du juré.

My gratitude to Dr. Peter Yu from the National Institute of Radiological Sciences (Japan), to Dr. Shinji Tokonami from the Department of Physics and Materials Science (Hong Kong) and to Dr. Jon Miles from the National Radiological Protecction Board (England) for their helps in making the detector irradiation at their exposure facilities.

Als Doctors i compatriotes Tayeb Bouassoule i Moaad Bakali per compartir amb mi els bons i mals moments. També, m'agradaria agrair al Dr. Carles Domingo per ajudar-me sempre sobre tot en temes informàtics.

A la resta dels meus companys, Javier, Raquel, Jordi Garcia, Joan Font, Pere, Joan Manel, Eduard, Ainhoa, Anna, Alex, Andreu, Raúl, Arnau i altres més que han estat al nostre grup per haver creat tan bon ambient de treball.

A Fermi, Maite, Cèlia i Mercè per estar sempre a disposició i per la simpatia que m'han mostrat. També, a Jordi, Manel, Rafa i Pepe pels seus ajuts tècnics.

A Mohamed, Mohssine, Tarik, Omar et le reste de mes compatriotes de la UAB pour leur amitié et support inconditionnels.

Finalement, mes très vives salutations à mes amis de Tanger (Rachid, Hamid, Jaafar, Jawhar, Hassan, ...) pour les merveilleux et inoubliables années qu'on a passé ensemble (merci infiniment).

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إلى عائلتي ...

Contents

1. Introduction

Nowadays, it is well-known that airborne short-lived radon (^{222}Rn) and thoron (^{220}Rn) progeny inhalation has a large contribution to radiation exposure of the public (ICRP, 1987; Nero, 1988; NRC, 1991; NRC, 1999; ICRP, 1994; UNSCEAR, 2000). In recent decades, a growing body of evidence has linked the lung cancer incidence to exposure to high levels of ^{222}Rn and/or ^{220}Rn regardless the cigarette smoking habit. As some of ^{222}Rn and ^{220}Rn daughters are isotopes of polonium, which are predominantly α-emitting radionuclides with energy above 5 MeV, the relatively high biological effectiveness of this radiation has to be taken into account. When these radionuclides are inhaled and release all their α-particles within the bronchial epithelium, the cells lining the airways may be damaged and lung cancer may ultimately result (James, 1988; Steinhäusler, 1988; Cross, 1988; NRC, 1991).

In the past, elevated ^{222}Rn and ^{220}Rn concentrations were thought to be found only in the underground mines. However, unusual high concentrations of these natural elements could occur in many closed environments (private homes and workplaces); so that, subsequent measurements are required for evaluation and control. The large number of people potentially exposed to elevated ^{222}Rn and ^{220}Rn progeny concentrations has led to recommendations, now being implemented by different countries, for national studies to assess the magnitude of the problem, for adopting remedial action levels, and for introducing mitigation procedures to reduce population exposures. In fact, the Commission of the European Communities, in its recommendation 90/143/EURATOM (OJEC, 1990), sets a reference level of 400 Bq m^{-3} for the annual average radon concentration to take remedial action in the exiting dwellings and a preventive upper level of 200 Bq m^{-3} for future constructions.

The dependence of the exposure risk from inhalation of indoor short-lived ^{222}Rn decay products on their airborne concentrations suggests that the time-integrated measurements of their equilibrium factor are necessary to well assess the associated annual radiation dose. To date, because of numerous difficulties found, there is not a well-established technique for a direct measurement of indoor ^{222}Rn daughter concentrations over long periods of time (i.e., from months to a year). The most common procedure used is based only on the knowledge of long-term concentration of the parent (^{222}Rn) assuming a mean equilibrium factor of 0.4 for its decay products (ICRP, 1994; UNSCEAR, 2000). However, due to local and temporal fluctuations of the processes influencing the indoor levels of ^{222}Rn and its progeny (ventilation, aerosol concentration, surface deposition, etc.), the assumption of an unique equilibrium factor for any real situation of indoor exposures can result in considerable misinterpretations of the inhalation dose actually accumulated.

The main goal of this PhD dissertation is to set up a new passive integrating and multi-component system of Nuclear Track Detectors (NTDs), for simultaneous measurement of indoor ^{222}Rn and its progeny concentrations even in the presence of ^{220}Rn. For this purpose, it is necessary to accomplish the following partial objectives:

i) To establish a mathematical approach for long-term equilibrium factor determination from the measurement of ^{222}Rn and its α-emitter progeny (^{218}Po and ^{214}Po).

ii) To construct a new dosimeter for obtaining the individual airborne concentration of ^{222}Rn, ^{220}Rn, ^{218}Po and ^{214}Po.

iii) To develop a Monte-Carlo computer code for numerical simulation of α-particle emission, propagation and detection with NTDs.

iv) To investigate the optimal experimental conditions for the detector evaluation.

v) To perform experimental calibration of the dosimeter as well as its subsequent comparison with active methods.

vi) To carry out a first application indoors.

Conceptually, the core of this report is structured into three parts:

1. The first part comprises a literature review and includes Chapters 2, 3 and 4. Chapter 2 is devoted to the ^{222}Rn and ^{220}Rn fundamentals and basic features making special emphasis on their generation and transport processes within medium materials as well as their accumulation indoors. Chapter 3 deals with the complex behaviour of airborne ^{222}Rn and ^{220}Rn decay products explaining in detail all the factors influencing their concentration indoors. Finally, Chapter 4 gives a brief description of the main measurement methods used for ^{222}Rn, ^{220}Rn and their daughters. Particular attention is paid to the NTDs as this is the type of detectors used in this work.

2. The second part constitutes the theoretical study and is divided into two chapters (Chapters 5 and 6). In Chapter 5, we comment the problematic of radiation dose

evaluation from indoor exposure to ^{222}Rn progeny by means of NTDs and we propose a new method based on the determination of a new parameter defined in this work as the reduced equilibrium factor. This method also contemplates the possibility of a separate measurement of ^{222}Rn and ^{220}Rn concentration indoors taking into account the fact that they have different half-lives but identical diffusion coefficient in a given medium. Chapter 6 describes the theoretical calculation of NTD response to ^{222}Rn, ^{220}Rn and their α-emitter progeny using Monte-Carlo techniques. The sensitivities estimated for the Makrofol detectors, both facing directly the atmosphere to be measured and enclosed within a diffusion chamber, are presented and validated with respect to an ideal detector.

3. The Chapters 7 and 8, being the third part of this volume, are fully centered to the experimental work carried out in this study and include the following sections:

- The description of the electrochemical etching system developed in our laboratory.
- The improvement of the existing semi-automatic track evaluation system.
- The performance of an initial phase of constructing a small exposure chamber for both ^{222}Rn and ^{220}Rn.
- The set-up of an irradiation device to generate mono-energetic α-particles from 2 MeV up to 8 MeV.
- The determination of the appropriate filters for ^{222}Rn/^{220}Rn separation.
- The optimisation of the electrochemical etching conditions to obtain the required α-energy response of the NTDs used.
- The partial calibration of our dosimeter in pure ^{222}Rn atmospheres.
- The evaluation of the measurement uncertainties as well as the associated detection limits.
- The comparison of our dosimeter with active methods using well-controled exposures in a reference laboratory.
- The obtention of the very first annual effective dose results due to inhalation of ^{222}Rn progeny in an inhabited Swedish house.

This dissertation finishes with the conclusions obtained and outlines the main perspectives left open for future works.