Abstract

I will present a method to measure radon concentration using compact discs (CDs) and digital video discs (DVDs). Radon is considered to be the second leading cause for lung cancer after smoking. Due to this long-term retrospective radon exposure estimates are highly desirable for risk estimations. There are many active and passive methods for measuring radon concentration. The best methods for long-term measurement are passive, which CDs and DVDs are a part of. A large laboratory study presented in this seminar shows less than 25% deviation of radon concentration measurements from their reference values, which means that this method can be used for large scale surveys. The method is also capable of monitoring thoron concentration, which most other methods are not.
1 Introduction

According to International Commission on Radiological Protection, radon exposure is recognised as the second leading cause for lung cancer after smoking. The risk comes from years, even decades of exposure. Radon concentration varies a lot from location to location. The biggest concentrations can be found in locations containing uranium ores, phosphate rock and metamorphic rocks such as granite and to a lesser degree, in common rocks such as limestone. Despite its short half-life (∼3.8 days), radon gas can accumulate to concentrations far higher than normal in buildings, especially in low areas such as basements due to its larger than air density. Safe radon activity concentration in buildings in Slovenia is below 500 Bq/m³. However, in some regions the activity concentration can reach above 2000 Bq/m³ due to the environment. Radon can also occur in water which comes from a ground source, e.g. in some spring waters and hot springs.

Therefore, long-term retrospective radon exposure estimates are highly desirable for risk estimations. Radon concentration analysis in buildings such as kindergartens, schools, homes, is important for determining whether a building requires a radon mitigation.

In this seminar I will present a method for measuring radon concentration using home stored CDs and DVDs. Almost every home has some CDs or DVDs stored in their home, which can be used for radon concentration measurement. If not, blank CDs or DVDs are inexpensive. The concentration measurement of exposed discs is performed in special designed laboratory. This method is a passive one, contrary to the active measurement methods which use scintillation cells, ionisation chambers, etc.

2 Radon

Radon is a chemical element with the symbol Rn and atomic number 86. It is a radioactive, colorless, odorless, tasteless noble gas occurring naturally. Radon is one of the densest substances that remains a gas under normal conditions. It is also the only gas that has no stable isotopes under normal conditions.
Radon is formed as one of the products in the radioactive decay chain, through which uranium and thorium decay into the stable element lead. The decay chain is presented in figure [1]. Thorium and uranium are the two most common radioactive elements on Earth. Their naturally occurring isotopes have very long half-lives ($\sim 10^9$ years), which is why isotopes will continue to exist for tens of millions of years at almost the same concentrations as they do now.

Figure 1: Radioactive decay chain of uranium and thorium through which radon and thoron are produced. The color of a circle represents the half-life of the element. The branching ration is presented as the thickness of the arrow of decay. Decay chain data was taken from [1].

The most stable isotope of radon $^{222}$Rn has a half-life of 3.8 days. The second important isotope for radiation analysis is thoron ($^{220}$Rn) with a half-life of 55.6 s. As radon decays, it produces other radioactive elements called radon progenies (also known as radon daughters) or decay products. Radon and some of its daughters ($^{218}$Po and $^{214}$Po) decay by emitting alpha particles, which have a travel distance in orders of centimetres before absorption in air. This means that alpha particles are harmless in air, but can cause damage in the body upon ingestion. Unlike radon, its decay products are solids and stick to surfaces, such as dust particles in air, which can be inhaled and stick to the airways of the lung. [2]

2.1 Methods for radon measuring

There are many methods and instruments available for measuring radon and its decay products. Every method has its own advantages and disadvantages depending on the situation. For many purposes we want to know the long-term average exposure, but for diagnostic purposes a continuous measurement may be appropriate. The decision for the method used normally depends on the feasibility and the cost of the measurement, as well as its accuracy and applicability of the technique. Measurement methods are divided into two groups: active and passive.

Active measurement methods use scintillation cells, ionisation chambers, etc. A scintillation cell is a hermetically sealed metal container filled with the air of interest. Usually the air that enters the cell is filtered, thus preventing radon decay products from entering the cell. This way, there will only be three alpha particles emitted for every radon decay, which come from radon decay and decay of its decay products. The three alpha particles are detected via the light pulses
they produce by interaction with the internal surface of the cell, which is covered in a scintillator. The light pulses are detected by a photomultiplier tube and its associated electronics.

Passive measurement methods use electret ion chambers, etched traced detectors, etc. Electrets are the electrostatic equivalent of permanent magnets, having a permanent surface charge. The electret is placed at the bottom of a conducting plastic chamber. Radon diffuses into the chamber volume and the electret loses charge because of the general air ionisation produced by radon and its decay products.

Etched track detectors rely on the use of plastic material to record the tracks of alpha particles from radon and its progenies. There are three materials which are generally used for radon measurements: (1) a thin film of coloured cellulose on an inert backing (LR-115), (2) polycarbonate and (3) poly allyl diglycol carbonate (CR-39). These etched track detectors can be in a container or exposed to ambient atmosphere. In the container the detectors are shielded from radon progenies, because only radon can diffuse into the container and thus only radon decay is detected. However, open detectors record alpha particles from both radon and thoron. Etched track detectors are generally used over periods from months to years.

The method of radon concentration measurement using CDs and DVDs falls in the group of etched track detectors. CDs and DVDs are made out of a material called Makrofol®, which is a bisphenol-A based polycarbonate. The material has a special property of high radon absorption. Because to this, the CDs and DVDs are considered to be good radon detectors. They can also be used to measure radon concentration in water.

3 Theoretical model of sorption and desorption

Radon concentration in the exposed polycarbonate depends on the ambient radon concentration, its half-life and the kinetics of the process of absorption. After the termination of the exposure, the radon concentration in the polycarbonate decreases due to decay and outgassing.

Once the radon atom is captured in the polycarbonate matrix, its further transport is ruled by diffusion and radioactive decay. The equation describing the transport is [5]:

$$\frac{\partial n}{\partial t} = D \left( \frac{\partial^2 n}{\partial x^2} + \frac{\partial^2 n}{\partial y^2} + \frac{\partial^2 n}{\partial z^2} \right) - \lambda n,$$

where $n$ is the number of radon atoms per unit volume, $D$ is the diffusion coefficient and $\lambda$ is the decay constant. Equation (1) is considered with initial condition $n_0(x, y, z)$ in the specimen and boundary condition $c(t)$ (boundary concentration). Following ref. [5] the general solution is given as:

$$n(x, y, z, t) = - D \int_S \int_0^t \frac{\partial G(x, y, z, t, \xi, \eta, \zeta, \tau)}{\partial \nu|_{\xi=\eta=\zeta}} c(\tau) dS_{\xi=\eta=\zeta} d\tau +$$

$$+ \int \int \int G(x, y, z, t, \xi, \eta, \zeta, \tau = 0) n_0(\xi, \eta, \zeta) d\xi d\eta d\zeta,$$

where $S$ is the surface in $\xi, \eta, \zeta$ coordinates, $G$ is the Green function and $\partial G/\partial \nu$ is the directional derivative to the normal of $S$. $c(\tau)$ is the radon concentration at the boundary of the sample.

CDs and DVDs are cylinders with radius $R$ and height $H$. Equation (1) in cylindrical coordinates takes the form [5]:

$$\frac{\partial n}{\partial t} = D \left( \frac{1}{r} \frac{\partial}{\partial r} \left( r \frac{\partial n}{\partial r} \right) + \frac{\partial^2 n}{\partial z^2} \right) - \lambda n.$$

For sorption of radon in disc with zero initial concentration in it, the second term in equation (2) is zero and the solution is denoted as $n_S$. For desorption of exposed disc in atmosphere free...
of radon after previous sorption for time $T$, the first term in equation (2) is zero and the solution is denoted as $n_D$.

The initial and boundary conditions are as follows:

- **Sorption:** $n_S(r, z, t = 0) = 0$, $n_S(r = R, z, t) = n_S(r, z = 0, t) = n_S(r, z = H, t) = c(t)$.
- **Desorption:** $n_D(r, z, t = 0) = n_S(r, z, T)$, $n_D(r = R, z, t) = n_D(r, z = 0, t) = n_D(r, z = H, t) = 0$. \[5\]

In sorption experiments two cases of exposure are used:

- **Case A:** exposure to constant concentrations. This condition is normally fulfilled when measuring radon concentration in the field. In this case $c(t) = c$.
- **Case B:** exposure to concentrations that decreases due to radioactive decay. This case is used in a posteriori calibration exposure, which is going to be presented in the next section. In this case $c(t) = ce^{-\lambda t}$.

The comparison of experimental data for sorption and theoretical solution under case A is presented in figure 2. The experiment was done using radioactive isotope $^{85}$Kr. The behaviour of krypton in plastic is similar to radon. The half-life of krypton is longer compared to radon’s half-life (≈ 10.7 y). From the comparison we see a good matching between experimental data and the theoretical model.

![Figure 2](image)

Figure 2: Sorption of $^{85}$Kr in plate plastics with thickness 0.7 mm exposed in air. The signal is gamma rays counts emitted in the decay. The graph in figure presents sorption under case A. The curve is the theoretical dependence. The figure is taken from [5].

Figure 3 presents the comparison between experimental data and the theoretical model of desorption after sorption of plastic in water with radon under case B. From the comparison we see a very good matching between experimental data and the theoretical model. Figure 3 also shows the difference between the time dependence of the signal from radioactive decay with degassing and just radioactive decay of radon in the plastic. Without degassing we would need to wait longer before processing the detectors, because radon concentration in the plastic would decrease slower. Degassing is an important step in processing CDs and DVDs, which is presented in the next section.
4 Processing of detectors

After the CDs and DVDs were exposed to the radon (months or years of exposure), the discs are collected and processed using following steps in the laboratory:

- A posteriori calibration exposure,
- Chemical pre-etching,
- Electrochemical etching,
- Counting of alpha tracks.

4.1 A posteriori calibration exposure

First the discs are cut into 2 parts. A three quarter part of each disk is stored in radon-free air and the other quarter is used for a posteriori calibration exposure. The piece of disk is placed in a hermetic exposure vessel, connected in a closed loop to a certified radon source, a pump and a commercially available reference radon-meter. At the beginning of the exposure the pump is started and radon activity is delivered to the vessel. When homogenous activity is reached in the system, the initial radon concentration is measured. After that the vessel is isolated by hermetic valves. Thus, the disk piece is exposed to radon concentration exponentially decreasing due to radioactive decay. At the end of the exposure the loop is restored (without the source) and the radon concentration is measured, in order to ensure that there was no activity leakage from the exposure vessel. After the exposure the disk is left for 2 weeks to degas in a room with low radon activity. [4]

4.2 Chemical pre-etching

After the disc parts are degassed, they are chemical pre-etched. This process removes the outer layer of the discs, which was exposed to not only radon, but also to radon progenies and other radioactive materials in air. Thus, the only tracks left on the etched surface of the disc

Figure 3: Time dependence of the signal from a plastic plate after exposure in water with radon for 48 h under case B. The signal is gamma rays counts emitted in the decay. The curve is the theoretical model and the dashed line represents only radioactive decay. The figure is taken from [5].
are from radon decay inside the disc. The disc is etched with a solution of KOH, water and methanol at 30°C. The mass ratio of the solution is 1:0.937:0.702. The thickness of the remove layer is controlled by the pre-etching duration. The desired removed thickness is a least 80 μm to remove all the tracks caused by the radon progenies in the air around the discs during the exposure time. This is presented in figure 4. The removed thickness is always determined by measurements with a micrometer before and after pre-etching. [6]

Figure 4: The reason for removing the top 80 μm of plastic, is to remove tracks caused by radon progenies on the surface of the disc. The whole lines represent the range of the alpha particles. The picture was provided by Dr. Ivelina Dimitrova from Sofia University.

4.3 Electrochemical etching

After electrochemical etching the alpha tracks in the disc are easier to spot and count with a microscope. Due to this, the disc parts are electrochemically etched. They are placed in a solution, through which a 3 kV/mm effective high voltage current is passed at 6 kHz frequency. The temperature of the solution is 25°C and it takes 3 h to develop the tracks. One developed track is presented in figure 5. After the process, the developed tracks are counted automatically by means of a computer scanner and a special computer algorithm. [6]

Figure 5: Developed track after electrochemical etching. The picture was provided by Dr. Ivelina Dimitrova from Sofia University.

However, after electrochemical etching we lose some information regarding the tracks. Before the etching the size of the track in the plastic determines the particle, that caused it. This way, we can distinguish between tracks caused by alpha particles from other particles. Electrochemical etching enlarges the tracks to the similar size and thus make it impossible to distinguish between
the origin of the track. Thus we count all the tracks to be caused by alpha particle from radon and its daughters decay.

4.4 Counting and analysis of alpha tracks

After the disc parts are processed the number of developed tracks is counted. The track density \( n_0 \) is proportional to \( ^{222}\text{Rn} \) activity concentration \( C_A \) integrated over the time of exposure \( t_{\text{exp}} \) \(^4\):

\[
n_0 = \text{CF} \cdot I_{C_A} = \text{CF} \cdot \int_0^{t_{\text{exp}}} C_A \text{d}t, \tag{4}
\]

where CF is the calibration factor. Thus, the average \( ^{222}\text{Rn} \) activity concentration \( \overline{C_A} \) could be estimated if the CF and \( t_{\text{exp}} \) are known and the net track density is determined.

The calibration factor CF is determined using the a posteriori exposed disc part. The CF is estimated as \(^4\):

\[
\text{CF} = \frac{\Delta n}{\Delta I_{C_A}}, \tag{5}
\]

where \( \Delta n \) is the increment in the track density formed by the a posteriori exposure and \( \Delta I_{C_A} \) is the integrated \( ^{222}\text{Rn} \) activity concentration during the a posteriori exposure. From the obtained CF the average activity concentration can be determined:

\[
\overline{C_A} = \frac{n_0}{\text{CF} \cdot t_{\text{exp}}}, \tag{6}
\]

The relative uncertainty in \( \overline{C_A} \) at the level of one standard deviation is:

\[
\delta(\overline{C_A}) = \sqrt{\delta^2(n_0) + \delta^2(\text{CF})}. \tag{7}
\]

The uncertainty in the exposure time is neglected. It should be taken into account that \( n_0 \) and CF are correlated, since \( n_0 \) is used to determine CF. However, in the case where the value of CF is estimated using 5 or more disks, this correlation can be neglected. \(^4\)

5 Results of method testing

The method was tested by using 50 discs (CDs and DVDs) which were exposed to four different \( ^{222}\text{Rn} \) reference atmospheres for a defined period of time. The exposures were carried out at the Radon Calibration Service Laboratory at the Federal Office for Radiation Protection Bundesamt für Strahlenschutz (BfS) in Berlin, Germany. The exposure duration and the mean \( ^{222}\text{Rn} \) activity concentration were chosen to cover a wide interval of integrated activity concentrations (corresponding to ten-year exposure to activity concentrations of about 70 - 550 Bq/m\(^3\)). The reference atmospheres for the discs exposures were created in a stainless steel calibration container with a volume of about 0.4 m\(^3\). The \( ^{222}\text{Rn} \) activity concentration was monitored every 10 min using scintillation cells and kept constant during the exposure time. Temperature and humidity were also monitored.

Before the beginning and after the end of exposure the discs were stored in an automatically ventilated room with low \( ^{222}\text{Rn} \) activity concentration. It was guaranteed that after the exposure the disks were kept there for at least 10 days, so that \( ^{222}\text{Rn} \) absorbed in them outgassed before analysing. The discs were analysed in the laboratory at the Sofia University using steps described in previous section. The results are presented in figure 6.
As seen from figure 6 the differences from the reference values are greater for CDs than for DVDs. For CDs they are between 11.1% and 23.4% and for DVDs they are between 3.4% and 9.6%. The uncertainties are systematic and tend to be higher at high concentrations. Part of this can be due to a bias in the reference concentrations during the posteriori exposure. In all cases the results were within ±25% interval around the reference values. It should be mentioned however, that most of the CDs/DVDs analysed in practice give a signal that corresponds to the lowest exposure group. There the bias from the reference value was 11.1% for CDs and 3.4% for DVDs.

![Graph showing average results for the 4 exposure groups.](image)

Figure 6: Average results for the 4 exposure groups. The reference exposures are given by horizontal lines. The uncertainties for CDs/DVDs are presented as error bars and the dashed lines correspond to uncertainties of the reference exposures. The graph was taken from [4].

6 Thoron

In recent years the scientific community became increasingly more interested in the thoron (\(^{220}\text{Rn}\)). Although, in some cases, the thoron exposure is non-negligible, the primary concern is the influence of thoron presence on radon measurements. In addition, until recently no methods for retrospective \(^{220}\text{Rn}\) measurements have been developed. One possibility to expand the CD

![Diagram showing the concept of the method for measuring thoron concentration using CDs and DVDs.](image)

Figure 7: The concept of the method for measuring thoron concentration using CDs and DVDs. The picture was provided by Dr. Ivelina Dimitrova from Sofia University.
and DVD method for retrospective thoron measurements, is to study the signal at two depths beneath the surface. The signal in the first (which should be in the interval 64-76 µm) is created by sources related to $^{220}$Rn and the absorbed $^{222}$Rn. The signal in the second depth (that should be $>76$ µm) is due only to the absorbed $^{222}$Rn and its progenies. For the purpose of measuring $^{220}$Rn the signal at the second depth is used to determine the $^{222}$Rn contribution to the signal in the first depth. The remaining signal is from $^{220}$Rn. Figure 7 illustrates the concept. This direction of studies is brand new and the future will reveal its potential for practical application.

7 Conclusion

Radon is a big hazard to health and thus needs to be monitored. There are many methods for measuring radon concentration, but for homes and large scale surveys, the method with small detectors, which do not need a constant electrical power, is the best to obtain long-term retrospective radon exposure estimates. The method using CDs and DVDs falls in this category of methods. Almost every home has somewhere stored some CDs and DVDs. If not, new CDs and DVDs are cheap. They do not take up a lot of space and can be placed almost everywhere in the monitored building. The method also allows measuring radon concentration in water. For the whole country like Slovenia only one laboratory would be enough to process CDs and DVDs.

The method was tested in a laboratory in Bulgaria which included a wide interval of radon concentrations. The results show, that the method is accurate within 25% interval around the reference value and thus is acceptable for large scale surveys. The method is also capable of monitoring thoron concentration, which most other methods are not. Because the method is still in the development, till now there were no studies on home stored CDs and DVDs, but just with new CDs and DVDs.

References


