Study of the use of Electret Ion chambers as radon dosimeters in South Africa

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KEYWORDS

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Radon Gas Monitors

Diffusion theory

Dose

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Polymers

ABSTRACT

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Radon is a naturally occurring radioactive gas which has been shown to be a major contribution to background dose. The underground mines in South Africa present a major occupational hazard. A need for cheap personal monitoring has been identified. In this thesis, commercially available electret ion chambers(EICs) are studied, with a view to use them as personnel monitors. These detectors were developed to measure the radon activity concentrations in houses. Their use in the harsh underground mining environments has shown some complications such as water entering the detectors and their response time to a rapid change in radon concentratiopn has been questioned. Several measurements are discussed in this thesis relevant to solve these problems. Firstly, the EIC's are checked for accuracy in the laboratory using a radon source, as well as in the radon chamber at the CSIR in Pretoria. Comparisons with alpha track detectors are also carried out.

The next aspect that is investigated, is the diffusion of radon through different materials. This is needed in order to make sure that covering the detectors with a bag does not cause a reduction in the radon entering the detector in a reasonable time. This work includes the mathematical development of radon diffusion for the experimental set-up which has been adapted for the purpose of measuring the diffusion coefficient of different materials. These results make it possible to discuss change in the response time due to the enclosure of the EIC's in a bag.

The results of this thesis indicate that the EIC's can be used in the mines,if they are protected with material with high diffusion coefficients.

January 2002

Declaration:

I, the undersigned, declare that the work contained in this thesis,**Study of the use of Electret Ion chambers as radon dosimeters in South Africa,** is my own original work and has not previously in its entirety or in part been submitted at any university for a degree.

Humphrey Monwabisi Vuza January 2002

Signature:...............................

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Finally to **my ancestors** for all the glory, power and honour, for they have carried me every step of the way!

Contents

Chapter 1

Background and outline of this thesis

1.1 Introduction

The main aim of this work is to study passive radon detectors for use as dosimeters in the mines. The Electret Ion Chambers (EIC), manufactured by Rad Elec Inc. in the U.S., have been tested in the mines [Bot99]. Some of these detectors got wet and gave unreliable readings. This thesis proposes that these detectors be covered with materials that keep water away but allow gases to pass through. Experiments to check on the diffusion coefficients of these materials and the response time of these detectors are performed.

1.2 What is radon?

In certain parts of the world radon gas poses a big threat to the health and everyday lives of human beings. Radon gas is radioactive, and in tightly insulated houses it can accumulate to concentrations that pose a health threat. If the gas is inhaled into the lungs, its decay and the decay of the radon daughters can increase the chance of getting lung cancer [Bei99]. Radon gas, like carbon-14 gas, is completely natural. It forms during the decay series of uranium-238, an element with a fairly interesting decay series. The radon atom is a gas atom with a half-life of only 3.82 days. Accumulation of radon atoms from the natural nuclear decay of uranium-238 is the source of radon . That means that radon gas concentrations are higher where uranium is plentiful in the soil and rocks. In the mines, where the uranium levels are very high, the radon concentrations can be extremely high.

1.2.1 Physical and chemical properties of radon

Radon gas is colourless, odourless and tasteless. It is the heaviest of the noble gases and is radioactive. It has all the properties of other noble gases. Being a noble gas, radon gas is extremely unreactive and is therefore free to bubble up out of the ground without becoming trapped chemically along the way. It decays through alpha particle emission into a radioisotope of polonium

$$^{222}\mathrm{Rn} \longrightarrow {}^{218}\mathrm{Po} + {}^{4}\mathrm{He}$$

Because radon has a short half-life and alpha particles have a high relative biological effectiveness, abbreviated RBE (a factor that measures the relative biological damage caused by the radiation), inhaled radon and especially its decay products are considered a probable cause of lung cancer [Bei99]. The fact that the decay product ^{218}Po is an alpha-emitting solid that has an even shorter half-life (3.11 minutes) than ^{222}Rn, makes it even worse.

1.3 Radon Measurements

Studies to measure radon concentrations in houses in large buildings and from tailing dumps in South Africa and other countries, like Canada; the U.S.; Sweden; Hong Kong etc. have been conducted [Dud96, Naz85, Tso94, Hol85, Wat93]. Active and passive detectors [Geo96] are the two main methods used in the detection of radon gas in houses and underground mines. The **active devices** need electrical power and include monitoring devices (continuous radon monitors, and continuous working level monitors). Active devices detect and record radon or its decay products continuously. These devices are generally more expensive and require professionally trained personnel for their operation. In contrast to active devices, **passive detectors** require no electrical power and generally trap radon or its decay products for later analysis in the laboratory. Passive detectors include charcoal canisters, liquid scintillation detectors, alpha track detectors and electret ion detectors.

In this thesis the Electret Ion Chambers (EIC) are employed and tested for use under different conditions. The type of EIC used in this study is commonly called, by the manufacturer, the Electret Passive Environmental Radon Monitor (E-PERM). The E-PERM system comes in different configurations, characterised by either the electret used, namely the short term "ST" or long term "LT" electret and the type of ion chamber used [Rad94, Kot92(a)].

One purpose is to study experimentally the diffusion characteristics of various materials that are proposed as E-PERM enclosures for use of E-PERMs under wet conditions in underground mines. Two lines of research may be distinguished: the first is the measurement of diffusion coefficients of the various materials, the second is the measurements of the E-PERM's response time. The approach in the experiments is to find the material enclosure that has the highest diffusion coefficient and give recommendations as to which materials are suitable for use. The effect of the enclosures on the response time of the detectors will then be discussed.

1.4 Outline of this thesis

At present, in South Africa, only alpha track radon gas dosimeters have been approved by the regulators as personnel dosimeters in the mines. Some problems with the E-PERM system have been pointed out, such as their reliability when used under wet conditions and the response time under rapidly changing radon concentrations.

This study looks into these two questions so that the E-PERM system can be recommended for use as personnel radon gas dosimeters in homes and underground mines in South Africa.

- In chapter 2, the origins of ionising radiation in general and its biological effects on human beings are discussed. The decay schemes leading to the three most abundant radon isotopes are also presented. Also in chapter 2 the properties of the different types of ionising radiation are touched on. The characteristics and behaviour of the short-lived radon progeny have been added in this chapter.
- The technical descriptions of all the materials used in this study are given in chapter 3.
- Chapter 4 gives the theory and mathematics of diffusion. Also, the determination of the diffusion coefficient using a source of known activity in the jar is explained in chapter 4.

- In chapter 5 all the experimental methods used in this study are explained and the results of these experiments are given and discussed.
- The conclusions and recommendations are presented in chapter 6.

Chapter 2

An introduction to the radon study

2.1 The origin of radon

As early as 1896 the French physicist Antoine Henri Becquerel (1852 - 1908) discovered that uranium and its compounds gave off a radiation that could cause a photographic plate to turn black even when the plate was separated from the source of radiation by thick paper [Ebb90]. The rays emitted were concluded to be an atomic phenomenon characteristic of the element.

This newly discovered phenomenon of spontaneous radiation from certain unstable elements was called radioactivity, for the reason stated above. Immediately after this the radioactive element causing this phenomenon, now known as radium, was discovered by the Curies (Pierre and Marie) in the uraninite (pitchblende) of the Eastern European metal mines of Schneeberg and Joachimstahl [Ebb90]. Friedrich Dorn in 1902 discovered the release of a radioactive gas from radium and called it radium emanation. It was found in 1908 by Gray and Ramsay that this radioactive gas was the heaviest member of the noble gases family [Tay71]. After about 1920 the name radon was attached to this gas.

The radiation from uranium is separable by an electric field into two electrically charged rays, one called alpha (α) rays and the other called beta (β) rays. A third type of radiation, called gamma (γ) rays, is unaffected by the field. Gamma rays are like visible light but can easily pass through a layer of matter.

Ernest Rutherford (1871 - 1937) and others showed in 1903 that alpha rays were a stream of helium ions, He^{2+}. While, on the other hand, beta rays proved to be a stream of high-speed

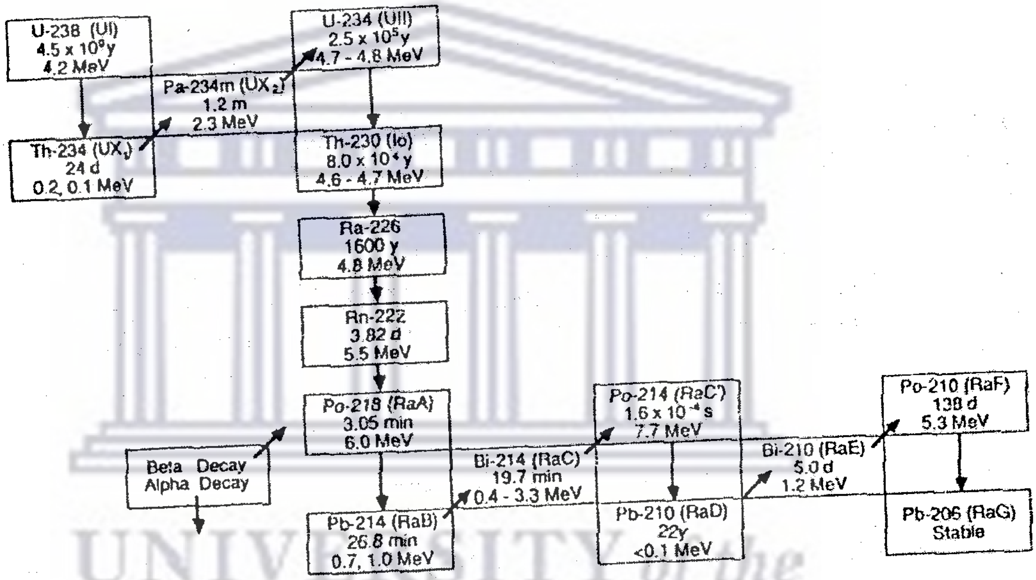


Figure 2-1: Decay scheme of ^{238}U including ^{222}Rn and its decay products.

electrons. There was clear evidence that uranium atoms had disintegrated to give off helium. For example the radioactive decay of ^{238}U by alpha-particle emission is written as :

$$^{238}U \rightarrow ^{234}Th + ^{4}He \tag{2.1}$$

There are twenty isotopes of radon known so far, which are all radioactive. Radon is a noble gas and is inert with no large sinks in the environment. Radon is relatively soluble in water and, for this reason, water transport can be a significant mechanism for bringing radon into some underground mines and into some homes where well water (ground water) is used instead of surface water. Like all noble gases, radon does not readily form chemical compounds although several compounds have been produced and used for experimental purposes [NCRP88]. Of these twenty isotopes, the most abundant are ^{222}Rn (radon),^{220}Rn (thoron) and ^{219}Rn (actinon). The origin of these three isotopes can be traced to the radioactive decay of radium, of which the isotopes are members of the naturally occurring decay series starting with ^{238}U, ^{232}Th and ^{235}U, respectively (Figs. 2.1 , 2.2 , and 2.3).

The radionuclides from which these series originate have half-lives that are longer or comparable to the age of the earth. The half-life, $T_{\frac{1}{2}}$, is the time it takes statistically until only half of the atoms are left :

$$T_{\frac{1}{2}} = \frac{\ln 2}{\lambda} \tag{2.2}$$

where λ is the decay constant, which is the probability of radioactive decay per second, and is characteristic of that particular nuclide. The decay constant, λ, of ^{222}Rn for example is $2.1\text{x}10^{-6}$ s^{-1}.

That is, for a number N of atoms of a radionuclide, the activity $\Lambda = N\lambda$ atoms will decay per second. Λ is expressed in Becquerels (Bq), defined so that 1Bq = 1 decay per second.

The half-life of ^{238}U is 4.5 x 10^{9}years while ^{226}Ra , the precursor of ^{222}Rn , has half-life of about 1600 years and ^{222}Rn one of 3.82 days. The subsequent short-lived polonium -(^{218}Po,^{214}Po), lead - (^{214}Pb), and bismuth - (^{214}Bi) isotopes, have half-lives of less than 30 minutes.

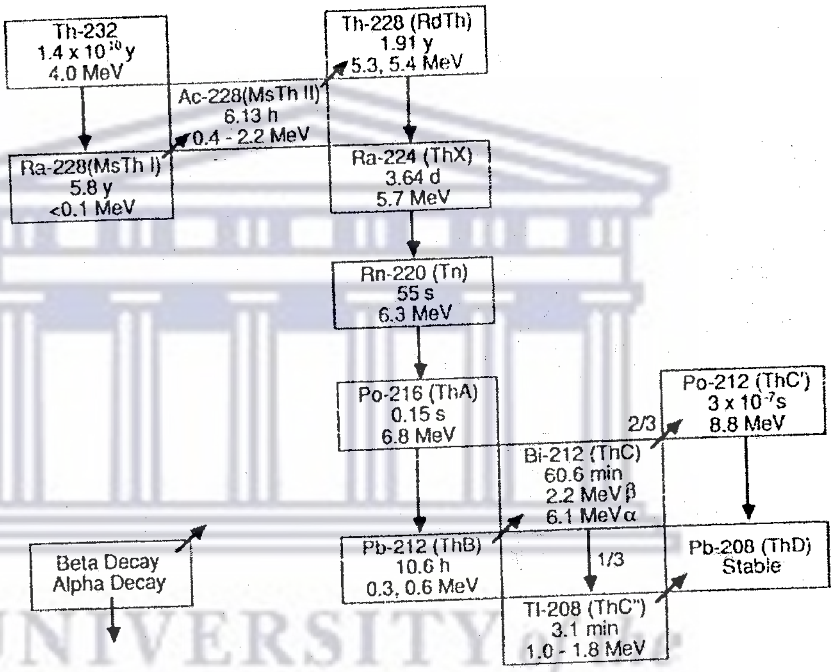


Figure 2-2: Part of the decay scheme of ^{232}Th.

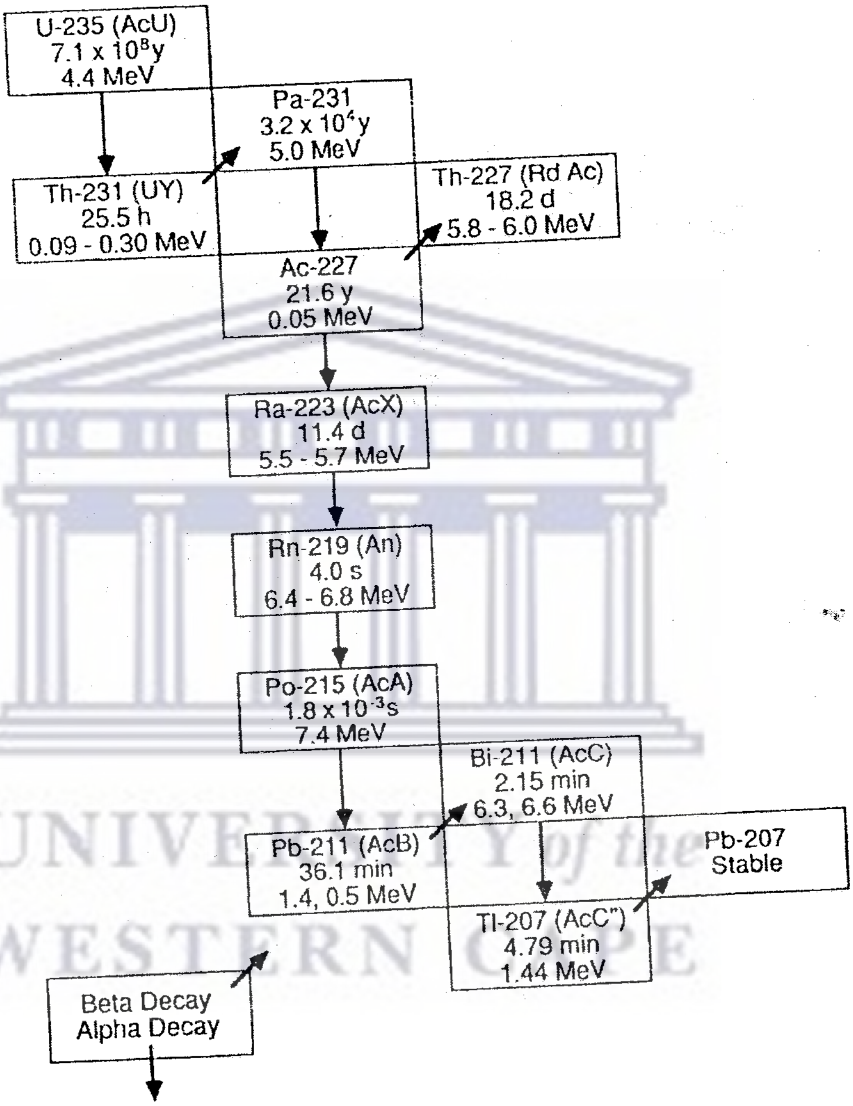


Figure 2-3: Decay series of 235U

2.2 Biological effects and radiation dosage

In this thesis whenever radiation is mentioned we mean ionising radiation, i.e. radiation which can ionise atoms. Electromagnetic radiation is energy travelling through space and it usually comes from atoms. Sunshine is one of the most familiar forms of electromagnetic radiation which delivers light, heat and suntans. To preserve life on earth we need radiation, but too much of it may be dangerous, especially at shorter wavelengths. Sunshine consists of radiation in a range of wavelengths from long-wave infra-red to shorter wavelength ultraviolet. For even shorter wavelengths we have X-rays and gamma rays which can be identical except that X-rays come from high energy electron transitions in the atomic nucleus and gamma rays come from the nucleus. These are also electromagnetic radiations (they belong to the same family of electromagnetic radiations such as visible light and radio waves). Apart from electromagnetic radiations, there are other kinds of ionising radiation which are used in medicine and which we all get in low doses from space, from the air, and from the earth. We have alpha (α) particles, beta (β) particles (electrons), and protons - all are electrically charged, and neutrons which have no electric charge. These are called corpuscular radiations (rays and particles) [WHO72]. Collectively we refer to these kinds of radiation as ionising radiation and it can cause damage to matter, particularly living tissue. At high levels it is therefore dangerous, so it is necessary to control our exposure. In this thesis we are more interested in the alpha radiation because radon-222 is an alpha emitter.

The earliest X-ray injury on record appears to have been observed in January 1896 by Grubbe and again in March 1896 when Edison reported that both he and his assistant were troubled by severe smarting of the eyes after several exposures to a "discharge tube" [Tay71].

Immediately after the discovery of radium, the possibility of radiation damage to human beings was recognised. The people who worked with radium ore were the first to be reported to have suffered injuries with skin burns [Ald94]. It was also discovered during the same era that not only did workers suffer skin burns but radiation could induce effects like cataracts (a gradual clouding of an eye lens which may skew the trajectory of incoming light rays and if the lens becomes totally opaque, light cannot enter the eye at all [Sta92]), sterility, damage to blood-forming tissues (in bone marrow) or genetic mutations.

The quantity of energy dissipated in a biological organism from a radiation dose can either be small or large. Even in the case where it is small the effects can be quite adverse because important chemical bonds may be broken. It is necessary to have a measure of radiation dosage to be able to monitor the effect of nuclear radiation on biological tissues. The type of radiation together with the energy deposited in the tissue determine the extent of the biological effect of radiation. This is the reason the neutrons are more harmful (destructive) than gamma rays of the same radiation dosage. Sources of alpha radiation outside the body are harmless, because the radiation is absorbed by the skin that consists of dead tissue, whereas internal sources are very destructive. The length of time in which the dose was received is also important to the effects of radiation. A series of minor doses has less overall effect than these doses given at once. Continuous exposure to such minor doses of radiation may result in cancer or leukemia. Hereditable defects are possible because radiation causes chromosome damage. Safe limits of radiation dose are being debated. It is important to have in mind the magnitude of the radiation humans may be subjected to even if there may not be strictly safe limits. We are all subjected to background radiation (that which is naturally and inevitably present in our environment) from the radiation from outer space (cosmic rays) and natural radioactivity which both vary considerably with location. Uranium and its daughters (decay products) in the soil are an important source of this background radiation. Soils contain varying amounts of uranium-238 (^{238}U), which decays in several steps to radium-226 (^{226}Ra), then to radon-222 (^{222}Rn), a gas (see Figure 2-1). Some dwellings situated in areas of high uranium content have been found to accumulate high concentrations of radon gas. Another source is potassium-40 (^{40}K), a radioactive isotope with a natural abundance of 0.012%.

In addition to natural background radiation, we may receive radiation from other fairly common sources. The most important of these are X rays used in medical diagnosis. The average person receives a radiation dose from this source that is about equal to that of the natural background. Very small radiation sources include consumer items such as television sets, luminous watches and now recently cellular telephones. The background radiation to which we are all subjected to has increased slightly since the advent of nuclear technology. Fallout from atmospheric testing of nuclear weapons increased this background by several percent, but it has decreased since atmospheric testing was banned. The radiation contributed by nuclear power plants is only a fraction of a percent of the natural background [Ebb90].

The radioactive isotope ^{222}Rn with a half-life of 3.82 days, has been identified to be a potential health risk due to its accumulation in especially poorly ventilated areas in the cold climates of Europe and North America. Radon-222 decays by alpha emission to radioactive lead, bismuth, and polonium.

Due to the high concentrations of radium and the low ventilation levels in underground mines, relatively high ^{222}Rn concentrations occur. When high levels of lung cancer were observed in the mines, it was attributed to ^{222}Rn. Studies were conducted in the 1950s which concluded that the radiation dose delivered to the lung tissue was not the result of ^{222}Rn itself, but of its short-lived radioactive decay products, called ^{222}Rn progeny or ^{222}Rn daughters. ^{222}Rn gas is inhaled and decays inside the lungs so that the daughters settle inside the lungs. Or, more importantly, ^{222}Rn decays in the atmosphere and the daughters attach to dust particles since they are solid. Decay of these ^{222}Rn daughters can cause damage to the living cells and ultimately lead to lung cancer [Fie00].

In South Africa, the main concern is the inhalation of radon and its daughters by workers in the mines. Since South Africa has more underground miners than any other western country, radon measurements are of particular significance for study here.

2.3 Properties of several types of ionising radiation

X-rays and gamma rays, like light, represent energy transmitted in a wave without the movement of material, just as heat and light from a fire or the sun travels through space. Unlike light, they both have great penetrating power and can pass through the human body. Thick barriers of concrete, lead or water are used as protection from them.

Alpha particles have a positive electrical charge and are emitted from naturally occurring heavy elements such as uranium and radium, as well as from some man-made elements. Because of their relatively large size and charge, alpha particles collide readily with matter and lose their energy within no time. Because of this, they have been shown to have little penetrating power and can be stopped by the first layer of skin or a sheet of paper. As a result, alpha particles do not contribute greatly to the external dose received.

This is not the case when an alpha emitter is taken into the body. By breathing or swal-

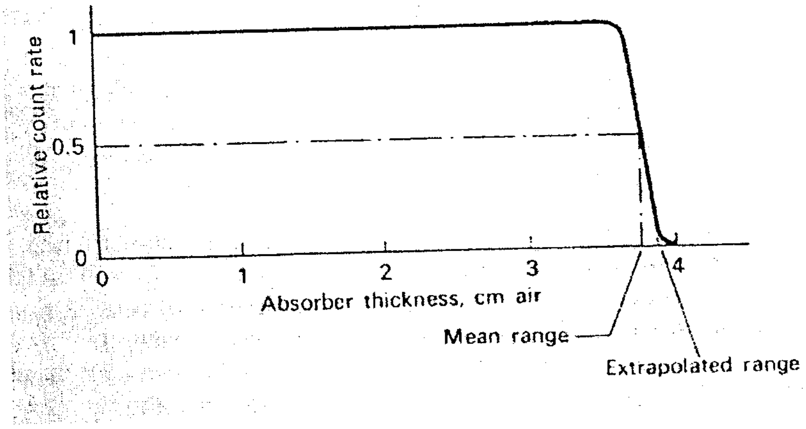


Figure 2-4: Alpha particle absorption curve [Cem96].

lowing, an alpha emitter, alpha particles can affect the body's cells. Because they give up their energy over a relatively short range, alpha particles can cause more biological damage than other radiations inside the body.

An alpha particle absorption curve (Fig. 2-4) is flat because alpha radiation is essentially monoenergetic (about 4 - 7 MeV) for ^{222}Rn and its progeny. If the thickness of absorbers is increased, then the energy of the alphas that pass through the absorbers will be reduced; the number of alphas is not reduced until the approximate range is reached. It is at this point that we find a sharp decrease in the number of alphas that pass through the absorber. It can be seen that near the very end of the curve the absorption rate decreases. This is due to straggling, or the combined effects of the statistical distribution of the "mean" energy loss per ion and the interaction with the electrons of the atoms of the absorber [Cem96].

Beta particles are fast-moving electrons ejected from the nuclei of atoms. Compared to alpha particles, they are much smaller and can penetrate up to 1 to 2 cm of human flesh for typical energies from naturally occurring beta sources. Beta particles are emitted from many radioactive elements. A few millimeters thick of aluminium sheet will be able to stop them.

Cosmic radiations consist of a variety of very energetic particles including protons which bombard the earth from outer space. These are more intense at higher altitudes than at sea level where the earth's atmosphere is most dense and gives the greatest protection.

Neutrons are particles which are also very penetrating. On earth, they mostly come from the splitting, or fissioning, of certain atoms inside a nuclear reactor. Water and concrete are the most commonly used shields against neutron radiation from the core of the nuclear reactor.

2.3.1 Radiation quantities and units

Radiation Exposure

This is a quantity that is roughly analogous to the strength of the electric field created by a point charge. Defined only for sources of X- or gamma rays, exposure is the quantity that expresses the ionisation produced by X- or gamma rays in a volume element [Sha72]. Alternatively, it is the sum of the electrical charges of the ions of identical sign produced in unit mass of air under defined conditions. It can also be defined in terms of the effect of a given flux of gamma rays on a test volume of air and is a function only of the intensity of the source, the geometry between the source and the test volume, and any attenuation of the gamma rays which may take place between the two [Kno89, WHO72, Sha72]. The basic unit of gamma-ray exposure is defined in terms of the incremental charge dQ due to ionisation created by the secondary electrons (negative electrons and positrons) formed within a volume element of air and incremental mass dm, when these secondary electrons are completely stopped in air. The exposure value will then be $\frac{dQ}{dm}$ and the SI unit is thus the Coulomb per kilogram (C.kg^{-1}), which has not been given a special name [Kno89]. The historical unit in which exposure is expressed is the roentgen (R), defined as the exposure that results in the generation of one electrostatic unit of charge per 1.293 x 10^{-3} g (1 cm^{3}at STP) of air.

The SI and the historical unit are related by

1 R = 2.58 x 10^{-4} C.kg^{-1}

Absorbed Dose

The absorbed dose of any ionising radiation in a material of interest is defined as the amount of energy imparted to that matter by ionising particles per unit mass of irradiated material. The historical unit of absorbed dose has been the rad, defined as the dosage of radiation that deposits 1 x 10^{-2} J of energy per kilogram of tissue. This unit is gradually being substituted by its SI equivalent, the gray (Gy) defined as 1 joule/kilogram.

The relationship between the two units is given by

1 Gy = 100 rad

The Dose Equivalent

Although the injury inflicted by a given type of ionising radiation depends on the amount of energy imparted to matter, some types of particle produce more significant effects than others for the same amount of energy imparted (equal exposure to different types of radiation do not however necessarily produce equal biological effects). The damage caused increases as the distance over which a given amount of energy imparted decreases. The biological damage inflicted by ionising radiation is attributed to the chemical alteration of the biological molecules that are influenced by the ionisation or excitation caused by the radiation. The intensity and duration of these alterations are directly related to the local rate of energy deposition along the particle track, known as the linear energy transfer L. The higher the L value (such as for heavy charged particles), the greater the injury produced for a given absorbed dose. To quantify the probable biological effect of a given radiation exposure, the concept of dose equivalent has been introduced.

Table 1-1 Quality Factors for Different Radiations

(from ICRU Report # 19)

L in water (keV/mm)	Quality Factor Q
3.5	1
7.0	2
23	5
53	10
175	20

The factor expressing the relative effectiveness of a given particle based on its linear energy transfer is known as the quality factor, Q. The dose equivalent H (radiation effect) is a quantity which gives a direct relation with the probability of radiation damage by stochastic effects:

$$H = D \times Q \tag{2.3}$$

If D is expressed in rad, H is defined to be in units of the rem. Under the SI system, D is expressed in grays, and a corresponding unit for H is called the sievert (Sv). Table 1-1 shows the quality factors as given by ICRU #19. (Later reports have replaced the quality factors with more complicated functions - the ICRU #19 are given as illustration of the importance of different radiation.)

1 Sv = 100 rem

One sievert of radiation produces a constant biological effect regardless of the type of radiation. The dose equivalent H can give an unclear picture of the radiation risk, because of the differences in sensitivity to radiation of organs and tissues and also because often exposure is not always uniform. To correct for this, weight factors, w_T, are introduced: these are defined as the fraction of the total stochastic risk due to the exposure of the organ T (see Table 2-2)[Gol88].

Table 2-2 : Fraction of the total stochastic risk which is due to the exposure of a specific organ T [Gol88].

Organ or Tissue (T)	Weight Factor w_T
Breast	0.15
Bone marrow	0.12
Lungs	0.12
Thyroid	0.03
Bone surface	0.03
All others	0.30
Gonads	0.25

We define the effective dose-equivalent, H_e (Sv), as the absorbed dose weighted to the radiation type and tissue or organ sensitivity :

$$H_e = \sum w_T H_T$$

The largest part of the airborne radiation exposure is due to the short-lived ^{222}Rn progeny filtered out of the gases respired by the lung. If the lung can be roughly divided into two regions, the alveolar and the bronchial regions, the tissue found to be most at risk is the bronchial epithelium, the site of most lung cancers believed to be ^{222}Rn progeny exposure induced.

2.4 Characteristics and Behaviour of the Short-Lived Radon Progeny

2.4.1 Introduction

In South Africa there are more miners than any other Western country. The levels of ^{238}U are very high in the underground gold mines. As radon gas belongs to the decay series of ^{238}U, then the radon levels are expected to be extremely high in the mines. Radon is the heaviest of the noble gases and is colourless, odourless, tasteless and radioactive. Radon does not easily interact chemically with other elements. The three most abundant radon isotopes are ^{222}Rn (radon), ^{220}Rn (Thoron) and ^{219}Rn (Actinon). Their half lives are 3.82 days, 56 s and 4 s respectively.

The particular ionising radiation that will be studied in this thesis is ^{222}Rn and its progeny. The short-lived ^{222}Rn progeny, ^{218}Po, ^{214}Pb, ^{214}Bi, and ^{214}Po, when inhaled by human beings, are radionuclides that deliver the alpha radiation dose to the tissue of the bronchial region that is implicated in radiogenic lung cancer. This section will seek to describe their behaviour in the three environments that are of interest; outdoors, indoors and in underground mines.

The four radionuclides in question do not exist in the environments in aggregates sufficient to be distinguished as the chemical elements polonium, lead, and bismuth. What is important in them is their radiometric properties which are listed in Table 2-3.

Table 2-3 Radiometric properties of ^{222}Rn and its short-lived progeny (NCRP, 1985)

Radionuclide	Half-life	Decay Constant (s^{-1})	Approximate No. of Atoms	
			Bq^{-1}	pCi^{-1}
^{222}Rn (α)	3.82 d	2.1×10^{-6}	476000	18000
^{218}Po (α)	3.05 min	3.78×10^{-3}	260	10
^{214}Pb (β)	26.8 min	4.17×10^{-4}	2300	85
^{214}Bi (β)	19.7 min	5.86×10^{-4}	1700	63
^{214}Po (α)	164μ s	4.22×10^{3}	2.4x10^{-4}	9x10^{-6}

It is important to always know the characteristics of each of the daughters in the decay series; for example ^{214}Pb is a beta emitter of little dosimetric significance, but it decays to

^{214}Po, an alpha emitter.

2.4.2 Units

Historically the airborne radioactivity of ^{222}Rn was expressed in the quantity Potential Alpha Energy Concentration (PAEC) in the unit Working Level (WL) which was introduced in the 1950s to introduce safety standards for ^{222}Rn levels in uranium mines [NCRP88]. Working Level (WL) is the common unit for expressing radon exposure rates, originally in uranium mines but at present in environmental exposures as well. By definition, WL is any combination of short-lived ^{222}Rn daughters in one litre of air that will result in an ultimate release of 1.3x10^{5}MeV of potential alpha energy (2.08 x 10^{-5} J.m^{-3}). A PAEC of 1WL is approximately the potential alpha energy concentration of the ^{222}Rn progeny in radioactive equilibrium with a ^{222}Rn concentration of approximately 3750 Bq.m^{-3}(= 100pCi.l^{-1}):

$$1Wl \sim E_{\alpha tot} C_0 = 2.08 \times 10^{-5} J.m^{-3} \tag{2.4}$$

where, C_o = 3750 Bq.m^{-3} and $E_{\alpha tot} = \sum E_{\alpha i}$ and $E_{\alpha i}$ is the potential alpha energy due to the decay of the individual decay products, where i = 1, 2, 3 indicates ^{218}Po, ^{214}Pb, and ^{214}Bi, respectively. Table 2-4 gives the potential alpha energies, $E_{\alpha i}$, per unit of activity (Bq) of each of the daughter products, that are based on the alpha-decay energies and half-lives of the ^{222}Rn decay series.

Table 2-4: Total potential alpha energy, $E_{\alpha i}$ (J Bq^{-1}), of each of the short-lived ^{222}Rn progeny released in decay to ^{210}Pb.

Radionuclide	Atoms per Bq	$E_{\alpha i}$(J Bq^{-1})
^{222}Rn	4.76×10^5	excluded
^{218}Po	2.65×10^2	0.58×10^{-9}
^{214}Pb	2.32×10^3	2.85×10^{-9}
^{214}Bi	1.72×10^3	2.11×10^{-9}
^{214}Po	2.37×10^{-4}	2.93×10^{-16}

with the potential alpha energies given in Table 2-4 the PAEC (Jm^{-3}) may be expressed in terms of the individual decay product concentrations:

$$PAEC = \sum E_{\alpha i} C_i \tag{2.5}$$

where C_i is the individual decay-product concentrations (Bq.m^{-3}).

The radioactivity of the mixture may be described in terms of the Equilibrium Equivalent Concentration (EEC). This is defined as that radon activity concentration in radioactive equilibrium with its short-lived daughters, that would have the same potential alpha energy per unit volume as the non-equilibrium mixture of progeny actually present.

Numerically, it is calculated from measured concentrations of the progeny,

$$EEC = 0.105[A] + 0.516[B] + 0.379[C] \tag{2.6}$$

where EEC is expressed in Bqm^{-3}or pCiL^{-1} and [A], [B], and [C] are concentrations of ^{218}Po, ^{214}Pb, and ^{214}Bi, respectively in the same units [NCRP88, Ner88].

The conversion between PAEC and EEC is then given by:

$$EEC = PAEC\left(\frac{C_0}{1WL}\right) \tag{2.7}$$

We can describe the activity concentration of the ^{222}Rn progeny in terms of the ^{222}Rn concentrations for a non-equilibrium case, by introducing an equilibrium factor F :

$$F = \frac{EEC}{C_{Rn}} \tag{2.8}$$

where C_{Rn} is the ^{222}Rn activity concentration. In the case where $C_i = C_{Rn}$, i.e. when ^{222}Rn and all its progeny are in radioactive equilibrium, F = 1 [NCRP88].

2.4.3 Behavior of the decay products

The concentration of the radon progeny is determined by removal processes and deposition on surfaces. The decay products are removed from the air, not only by radioactive decay, but also by ventilation and by reactions with the structure and with the equipment inside the structure. The decay products can form small airborne agglomerates and can attach to previously existing particles which is an additional and important manifestation of their chemical activity.

The alpha decays imparting the most significant radiation dose are those of ^{218}Po and ^{214}Po, for the ^{222}Rn series. We also have comparable isotopes for the ^{220}Rn series, which are ^{212}Bi and ^{212}Po. The concentrations of decay products combined, given by PAEC or EEC, depends on the concentrations of the first three decay products (^{218}Po, ^{214}Pb, and ^{214}Bi) for the ^{222}Rn series (and on^{212}Pb and - to a lesser extent ^{212}Bi - for the ^{220}Rn series) and on the amount of polonium alpha energy that each will yield [Ner88].

Some General Considerations

In an atmosphere with no ventilation where the concentration of radon is stable and where each decay product, once formed, is removed only by radioactive decay, radon and its progeny would be in a state of equilibrium, all having the same radioactivity concentration. In atmospheres of houses and buildings where air is exchanged at some fixed ventilation rates, not only is the radon concentration less than it would otherwise be, but the concentrations of the decay products are reduced relative to that of radon because of removal by ventilation and plateout as discussed below.

However, the chemical activity of the progeny further complicates their behaviour as suggested above. The fact that the progeny can attach to particles or surfaces, and that these attachment rates can vary with conditions, makes general characterisation of the state of the progeny - and of its dependence on ventilation rate, particle concentrations, and other factors - exceedingly complex. However, it is still possible to specify a relatively straightforward framework for looking at the behaviour of the progeny, since we are dealing with only a few species whose rate of production from early parents of the decay series is determined solely by known half-lives.

The various mechanisms for changing the state or presence of the progeny, other than radioactive decay itself, for an unspecified decay product (progeny) can be illustrated by Fig 2-5 [Ner88].

2.4.4 Formation and Behaviour of Unattached Radon Progeny

The first daughter product of radon, ^{218}Po, becomes a positively charged ion, after an atom of ^{222}Rn has decayed in the atmosphere. This happens by alpha emission and there will be excess

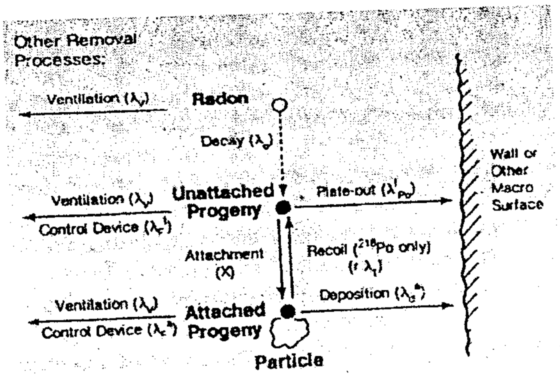


Figure 2-5: Removal processes of Radon and its decay products [Ner88].

electrons remaining which in turn are stripped away by recoil. Usually one or more of the orbital electrons are also lost. This leads to the charged ion becoming neutral and beginning to grow in size from the initial atomic dimensions.

There are many mechanisms by which the polonium cluster grows, one is the adsorption of water vapour molecules and trace gases. The polonium atom can also combine with oxygen or other substances such as nitrate or sulphate to become a simple compound [NCRP88]. This compound with its cover of water vapour becomes attached to a particle in the ambient aerosol within a relatively short time. These small clusters are continually being created by the decay of atmospheric radon and a small percent of the ^{218}Po is always present not attached to the ambient aerosol. The ^{218}Po that is not attached to the ambient aerosol is referred to as the unattached fraction of ^{218}Po in literature. The majority of the ^{218}Po and later progeny exist in the atmosphere in the attached form. ^{218}Po with a half-life of 3 minutes will decay by alpha emission and imparts recoil energy to its daughter ^{214}Pb. This will result in a fraction of ^{214}Pb atoms becoming detached from particles and following the general behaviour pattern of the original unattached ^{218}Po [NCRP88]. The concentration ratio of unattached ^{214}Pb: ^{218}Po is thought to be about 1:10 [NCRP88]. ^{214}Pb and ^{214}Bi both decay by beta emission which do not release sufficient recoil energy for detachment. The alpha emission from ^{214}Po can act to free some ^{210}Pb atoms, but these have long half lives and therefore are of no interest in this study.

The environmental unattached fraction diffuse rapidly, due to their extremely small particle size, and the diffusion process plays an important role in the physical process for their removal from the atmosphere and for their deposition in the respiratory tract. The diffusion coefficient of the unattached daughter clusters has been considered to be about $5 \times 10^{-6} \mathrm{m}^2\mathrm{s}^{-1}$ for many years, which is their most interesting characteristic [NCRP88]. This diffusion coefficient corresponds to the behaviour of a particle with size of about 1 nm.

The short mean life of a particular ^{218}Po cluster and the number of chemical and physical processes going on, make it difficult for various investigators to come up with a constant value for the diffusion coefficient.

The attachment rate to the ambient aerosol is directly proportional to the number concentration of the aerosol, which can easily be expressed as particles per cubic meter. The proportionality factor usually called the attachment coefficient is thought to be about $1.4 \times 10^{-12} m^3 s^{-1}$.

$$\text{Attachment rate} = (\text{attachment coefficient}) \times (\text{particle concentration})$$

Some studies to measure indoor deposition velocities of unattached radon progeny have yielded results ranging from 7 mh^{-1} to 36 mh^{-1}. Plateout rates for the unattached daughters can be found by combining these deposition velocities with the room characteristic of surface-to-volume ratio [NCRP88].

$$\text{Plateout Rate} = (\text{deposition velocity}) \times (\text{surface area})/\text{volume}$$

For the removal of the unattached daughters from the atmosphere, there are three main competing processes; radioactive decay, deposition on surfaces and attachment to the ambient aerosol.

2.5 Formation and Behaviour of Attached Radon Daughters

Some of the unattached ^{218}Po remains in the air for an appreciable time and a fraction is deposited on surfaces, while the rest becomes attached to the ambient aerosol. The relative distribution depends on the aerosol concentration and, to a lesser extent, on the size distribution of the aerosol.

Radioactive decay of ^{222}Rn in the atmospheric volume under study produces the majority of the attached progeny. In the mines, older air with higher concentrations of the daughters may be brought in by the ventilation systems.

The removal of the attached radon progeny from the air is primarily by radioactive decay. The mixture of the daughters has an effective half-life of about 30 minutes while none of the removal processes for aerosols is so rapid.

In this thesis methods to measure ^{222}Rn, rather than its progeny, are discussed.

Chapter 3

Equipment for radon measurements used in this work

3.1 Introduction

The assessment of radiation dose to the workers and the general public is done world-wide. One way of doing this is by measuring radon and radon daughters. Several methods have been developed to measure radon or its progeny [Geo96], such as pulse ionisation chambers, scintillation detectors with zinc sulfide ZnS(Ag), alpha particle spectrometers with silicon diodes, surface barrier or diffused junction detectors, registration of nuclear tracks in solid-state materials, gamma-ray spectrometry with NaI(Tl) scintillation crystals or germanium lithium (GeLi) semiconductors, and electret ionisation chambers. The instruments and methods mentioned depend on whether radon or radon progeny are being measured, on the type of radiation being monitored, and the duration of the measurement. The other criteria that are important are convenience, applicability, reliability, portability, and cost considerations. In this chapter the method of electret ionisation chambers (EIC) will be described, since this is the method mainly used in the work presented in this thesis. The work in this thesis uses the Electret Ion Chamber, E-PERM system, provided by Rad Elec Inc. and the description below is applicable to the systems provided by them.

The EIC has the following characteristics:

1) The EIC is a small and inexpensive device.

2) It operates by ion pair creation from alpha particle interactions with the air inside the chamber.

3)There is a correlation between the electret voltage drop versus the number of ions formed and radon concentration.

4) The measurement phase is passive, that is the EIC is simply deployed and it does the detection.

5) The relationship between electret voltage change and the measured radon activity concentration is not linear but is predictable and corrected using an appropriate algorithm.

The above characteristics make it easy to use and worth evaluating for use as a personal dosimeter.

3.2 What is an electret?

This consists of a piece of dielectric material that carries a quasi- permanent charge and it can be charged by a special process so that it retains the charge nearly permanently. Electrets are made from high dielectric fluorocarbon polymers such as Teflon which can maintain constant electrostatic fields under extreme temperature and humidity conditions. It is the charge on the electret that produces the strong electrostatic field that can collect ions of opposite sign. When ions are collected the surface charge is partly neutralized which will result in the surface voltage decreasing proportionally.

The drop in surface potential behaves according to established ion chamber theory and is not sensitive to humidity change.

Electrets are constructed such that they are stable at high humidities and this depends on the following factors :

i) the method used in preparing the electrets

ii) the method of processing the electrets such as annealing at high temperatures.

iii) The quality of the material from which it is constructed and

iv) on the way electrets are handled and stored.

There are different techniques of preparation and processing of the electrets, one being the simultaneous application of heat and electric field to orient the dipoles within the Teflon disks.

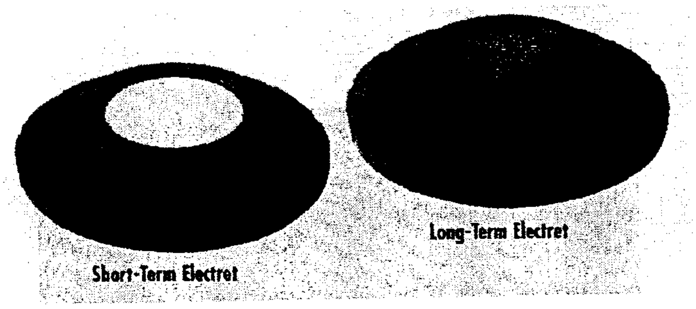


Figure 3-1: Long-term and short-term electrets.

The disks are then cooled to "freeze" the dipoles in place, while the electric field is still on. One other technique is by the electrical breakdown field technique in glass.

The disks are then loaded into holders such that exactly the same surface area of each electret is exposed. These holders are electrically conductive plastic materials whose purpose is to secure the Teflon and they are threaded so that they can be screwed into the electrically conductive ion chamber. This electret assembly (see Fig. 3-1) is then screwed into a plastic lid (protective cover) with a small volume to maintain a small air gap between the electret and the lid. A small air gap is maintained so that little or no ionisation occurs. The holders are also designed to fit smoothly into the measurement opening (recessed circular receptacle) in an electret voltage reader to ascertain reproducible voltage readings. The chamber lid has a small hole which is covered with filter paper [Rad94] .

3.2.1 Dynamic Range of the electrets

The various EIC system calibration factors only cover an electret voltage range of 200 V to 750 V. If electrets have voltages less than 200 V, then ion collection inside the chamber by electrets will be less efficient, while with electret voltages above 750 V ion multiplication (that is, freed electrons will have enough energy to cause further ionisation) can occur. This 200 V to 750 V range establishes the dynamic range of integrated radon concentration (expressed in pCi.l^{-1}.days or Bq.m^{-3}.days) which each EIC system can measure.

3.3 The Electret Ion Chamber (EIC)

The commonly used and simplest types of radiation detectors are based on the effects produced when charged particles pass through a gas [Kno89].

In general, all nuclear radiation when passing through air builds up ions in air. Ion chambers are gas-filled detectors based on sensing the direct ionisation created by the passage of the radiation. These detectors operate by collecting all charges created by direct ionisation within the gas through the influence of an electric field. In this sense an ionisation chamber can be regarded as a parallel-plate capacitor in which the space between the plates is filled with a gas. The electric field in the space between the plates keeps the ions from recombining with the electrons. In the parallel-plate model, one can think of a cloud of electrons drifting towards the plate with positive surface potential, while the positively charged ions drift toward the plate of opposite charge [Kra88].

There is a difference in response of the ionization chambers depending on the density of the air. At higher elevations (or at lower pressures), fewer air molecules are available for the radiation to produce ion pairs in a given path length compared to the ions produced at lower elevations (or at higher pressures). Under lower atmospheric pressures (or at higher elevations), the ionization chambers are expected to give a lower response.

An Electret Ion Chamber (EIC) consists of an electret screwed into a (threaded) conducting chamber, with the electret serving as one electrode and the inside wall of the chamber as the other. The radon gas (^{222}Rn) enters the chamber through filtered inlets on the sides and decays inside the volume of the chamber generating ions which are then drawn to and collected on a charged electret as a means of measuring the amount of radiation. The E-PERM® type of chambers are conductive plastic materials of different shapes and volumes. The three common types of chambers are [Rad94]:

i) The 200 ml standard or "S" chamber (Fig. 3-2) with an on/off mechanism. It consists of a conductive plastic and stainless steel spring. The spring-loaded assembly acts as a means to cover the electret and turn the collection of ions on and off. The "S" chamber is designed to allow radon gas to enter the chamber by diffusion while radon progeny, are filtered or baffled out. This set up allows serial exposure and multiple measurements with the same electrets.

The integrated exposure range (the integrated exposure which causes the electret to lose its

Figure 3-2: "S"-chamber in off and on positions.

potential) is:

- with short term "ST" electret : 9000 Bq.m^{-3} .days
- with long term "LT" electret : 92500Bq.m^{-3}.days
- The dimensions with electret :
- Height: open - 13.97 cm , closed - 10.16 cm
- Diameter : 8.255 cm
- Volume : 200 ml

ii)The 50 ml "dome-top" "L" chamber without an no on/off mechanism is a device which permits making multiple measurements. This is the device which is being compared to the alpha track device (RGM) for use as an environmental radon monitor in mines. The "L" chamber is made of conductive polypropylene material.

The integrated exposure range is:

- with "LT" electret : 462500Bq.m^{-3}.days
- with "ST" electret : 46250Bq.m^{-3}.days

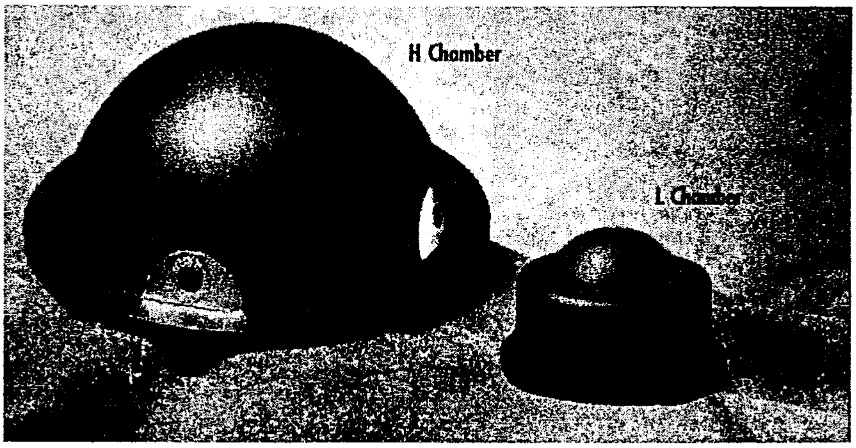


Figure 3-3: "H"-chamber and "L"-chamber.

The lower limit of detection (the minimum integrated exposure which causes the electret to lose enough volts to give a result with reasonable uncertainty) is 9250Bq.m^{-3}.days

The dimensions of the "L" chamber are:

- 6.6 cm diameter and 4.3 cm in height (without an electret)
- 7.62 cm diameter and 4.8 cm in height (with an electret)

iii)The 1000 ml "H" hemispherical chamber made of electrically conducting plastic which uses a short-term electret only. This has high sensitivity and can be used for measurements requiring prompt results. It has a sensitivity six times that of the "S" chamber. The "HST" configuration can measure low levels of radon, such as ambient outdoor radon levels with very high precision. This configuration has a precision of better than 10% for a one-day exposure period at 37 Bq.m^{-3}

The integrated exposure range is 1480 Bq.m^{-3}.days

In all E-PERM® chambers, ^{222}Rn decays and emits alpha particles in the chamber volume and the daughters formed get repelled to the walls of the chamber where they undergo further decay causing alpha emission. The daughters are repelled to the walls (plate-out) because the electret is positively charged and therefore repels the positively charged progeny to the walls of the chamber, which have an induced negative charge. Only a small fraction of the daughters will remain neutral and will soon diffuse and deposit on the walls. The range of energies of the emitted alpha radiation is about 4.15 MeV to 7.7 MeV with their corresponding ranges of 3.0 cm to 6.5 cm in air at STP [Kot92].

To be able to provide a qualitative explanation for the experimental results obtained, three considerations apply:

1. Ion chamber dimensions smaller than the range of alpha radiation. This will mean that only a portion of the energy is spent in the air volume and all the alpha particles terminate at the inner surface of the chamber. Under this condition the total ions produced depend directly on the air density or on the pressure.

2. The other case is where the chamber dimensions are much larger than the range of alpha radiation at sea level. In this case at lower pressures (higher elevations) alpha radiation will dissipate all their energy inside the ion chamber and there may not be any significant pressure effects at all under these conditions.

3. The intermediate case is when the chamber dimensions are slightly larger than the range of the alpha radiation at sea level. Here pressure effects may be manifested at somewhat lower pressure when the chamber dimensions become smaller than the increased range of the alpha particles at that lower pressure.

The manufacturer has supplied correction factors for the different cases at different pressures.

3.4 The radon accumulator (JAR)

This is a simple glass bottle which is used for the accumulation of radon inside it. When there are no E-PERMs inside, its air volume is about 4 litres and the air volume will depend upon the type and number of E-PERMS used in the accumulator. For different configurations see Table 3-1 below [Kot94]:

Table 3-1 Air volume of the accumulator for different type and number of E-PERMS.

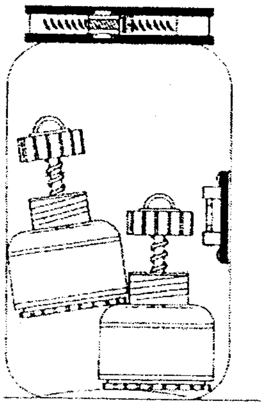


Figure 3-4: Accumulator Jar with two S-chambers and a source.

E-PERM Type	Number of units	Air volume In ml
SST or SLT	1	3843
SST or SLT	2	3720
SST or SLT	3	3597
LST or LLT	1	3920
LST or LLT	2	3873
LST or LLT	3	3827
LST or LLT	4	3780

The values in table 3-1 above were determined experimentally by carefully measuring the volume of the empty accumulator and its air volume when the different E-PERM configurations were loaded into the jar, were calculated by subtracting the known volume of the chamber material.

A number of the empty jars were filled with distilled water and the water needed to fill each jar was weighed. Now, with the different types and numbers of E-PERMs loaded into the jar, measurements were taken and the difference in the weight of the water between the two measurements gives the air volume of the jars. This process was repeated many times and the values in Table 3-1 have an error of less than 3% using the procedure of [Kot94].

The manufacturer of the jars claims that they are leak-tight when closed with the rubber seal provided.

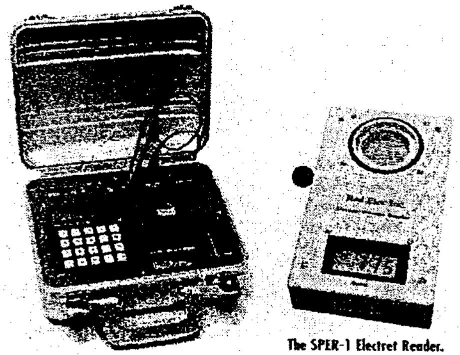
3.5 The Surface Potential Electret Reader (SPER-1)

SPER-1 is a non-contact, battery operated, voltmeter which reads electret surface potential with the E-PERM system with high precision.

The internal electronic components consist of an ultra - low leakage apparatus and specially designed integrated circuit board which will provide accurate, stable voltage measurements consistently. It operates by simply placing an electret face down on the recessed circular receptacle (shuttered window), and a precision machined aluminium/stainless steel shutter mechanism. The shutter mechanism is activated by drawing the shutter knob to its downward limit and the reader will read the electret voltage. SPER-1 is stored in its case which has a desiccant canister which must be periodically inspected and redried when necessary. If proper care of this desiccant is exercised, this will ensure that the reader is stored in a relatively dry environment as required for its proper use. The reader's voltage response is checked for stability using "reference" electrets which are used as part of quality assurance program. To ascertain that the reader functions properly regular checks with "reference" electrets for constancy should be performed [Rad94].

SPER-1 will not reproduce readings or they will fluctuate if subjected to very humid summer conditions or when moisture condenses inside the reader.

SPER-1 can measure an electrostatic field potential of up to 1999 Volts and the precision is ±1 Volt. The ultra-low leakage electrode has a humidity protective coating applied to a high dielectric substrate. The advantage with this instrument is that it gives readings immediately (no sending away of electrets) and no daily calibration of the instrument is necessary.



The SPER-2 Microprocessor Electret Reader.

The SPER-1 Electret Reader.

Figure 3-5: Sper readers.

3.6 The National Institute of Standards and Technology (NIST) Polyethylene - Encapsulated ^{226}Ra/^{222}Rn Emanation (PERE) Standards

Polyethylene Encapsulated ^{222}Rn sources developed by NIST in the USA have been used in this work. These sources consist of amounts ($\sim$0.08g) of calibrated ^{226}Ra solutions having known activity concentrations that are contained in polyethylene capsules. These capsules are circular cylinders of polyethylene with $\sim$0.32 cm outside diameter and a 1.0 cm effective length along the emanating surface with ends covered with two stopper - like 0.5 cm teflon plugs [Col95]. The ^{222}Rn diffuses through the thin polyethylene at a constant rate. This new emanation standard source has been demonstrated to emanate a well-characterized and known quantity of ^{222}Rn activity when employed in an accumulator. This source should be employed in a suitable accumulation chamber that allows quantitative transfer of the accumulated ^{222}Rn activity such as the jar described in section 3.4. When used in such an accumulation chamber, it will allow calculation of ^{222}Rn activity accumulated in the chamber and the standards are certified by NIST in terms of two parameters. These parameters are :

A_{Ra} = total ^{226}Ra activity contained in the capsule at some reference time t_r; and f = ^{222}Rn emanation fraction (i.e. the fraction of the total ^{222}Rn generated by decay of ^{226}Ra that is released from the capsule and contained within the volume of the accumulator).

These parameters $\Lambda_{Ra}$and f are certified with overall uncertainties of $2u_{A_{Ra}}$and $2u_f$, where $u_{A_{Ra}} = 0.93\%$ is the uncertainty in Λ_{Ra} and $u_f = 4.0\%$ is the uncertainty in the emanation factor f [NIST4968]. The uncertainty contributions from other components are negligible compared to $u_{A_{Ra}}$ and u_f so that the relative uncertainty in the accumulated ^{222}Rn activity, A_{Rn}, may be approximated by [TEC94]:

$$2u_{A_{Rn}} = 2\sqrt{(u_{A_{Ra}}^2 + u_f^2)} \tag{3.1}$$

These two parameters are calibrated in terms of measurements that can be directly related to the U.S. national radon measurement calibration standard (which is known as the pulse-ionization-chamber-based primary radon measurement system [Col90, Hut92]) and to national and international radium standards maintained by NIST [Col95].

The source must go through what NIST calls the "pre-conditioning requirement" before and between each accumulation. This is done by simply taking the source out in an open space for a minimum of 24 hours and this process is called open-air "equilibration" which should be performed between any sequential accumulations so that each accumulation begins under as identical as possible diffusion boundary conditions. This "equilibration" is done so as to "restore" the emanation fraction f and makes sure there has been no build-up of radon in the source [Col95].

3.6.1 Theoretical Calculations of ^{222}Rn in an Accumulator

If we have a capsule with radium (^{226}Ra) activity content of A^0_{Rn} at the beginning of an accumulation (t = 0) and a constant emanation fraction f, the build up of ^{222}Rn activity as a function of time t within a leak - tight accumulation jar is approximated as [Col95]:

$$A_{Rn} = A^0_{Ra}\exp[-\lambda_{Ra}(t+T_D)]f(1-exp[-\lambda_{Rn}t]) + A^0_{Rn}exp[-\lambda_{Rn}t] \tag{3.2}$$

In Eq. (3.2), A^0_{Rn} is the initially present ^{222}Rn activity in the accumulation jar (i.e. , $A_{Rn} = A^0_{Rn}$ at t = 0)

$A^0_{Ra(r)}$ = activity of ^{226}Ra contained in the capsule at some reference time t_r

λ_{Ra} = decay constant for ^{226}Ra

λ_{Rn} = decay constant for ^{222}Rn

$T_D = (t_0 - t_r)$, the decay-time interval

t_r = reference time for A_{Ra}

Since the half-life of radium (1600 years) is so much longer than the times relevant to the measurements in this thesis, the exponential involving the radium decay can be neglected. If equation (3.2) is integrated, we get the total integrated ^{222}Rn activity I_{Rn} accumulated over some total accumulation time T_A. This is necessary for encapsulated standards used in an accumulation mode with electret ion chambers.

Integrating equation (3.2) from $t = 0$ to $t = T_A$ will give

$$I_{Rn} = \int_0^{T_A} A_{Rn} dt = f A^0_{Rn} \int_0^{T_A} (1 - \exp[-\lambda_{Rn} t]) + A^0_{Rn} \int_0^{T_A} \exp[-\lambda_{Rn} t]$$

$$= f A^0_{Ra} \{ T_A - (1/\lambda_{Rn})(1 - \exp[-\lambda_{Rn} T_A])\} + A^0_{Rn} (1/\lambda_{Rn})(1 - \exp[-\lambda_{Rn} T_A])$$

In the case where there was no ^{222}Rn initially present in the accumulation jar (i.e., $A^0_{Rn} = 0$), then the integrated activity will just be equal to the first term on the right hand side

$$I_{Rn} = f A^0_{Ra} \{ T_A - (1/\lambda_{Rn})(1 - exp[-\lambda_{Rn} T_A]). \tag{3.3}$$

3.7 Technical Description of the Materials used to cover E-PERMS

The diffusion coefficient for radon of various materials will be investigated in this thesis. These materials are described below:

3.7.1 Polyethylene

These are polymers and can be described as chemical species of very high molecular weight. It is made up from many repeating units of low molecular weight. A compound used to make a polymer and from which the polymer's repeating units arises is called a monomer. Polyethylene is a simple polymer, which consists of many ethylene units $-CH_2CH_2-$, chemically bonded to one another to form long-chain molecules consisting of thousands of ethylene units. Polymers are classified as addition polymers or condensation polymers, depending on the type of reaction used in forming them [Ebb90]. An addition polymer is one that is formed by linking together many molecules by addition reactions. Polyethylene belongs to this category. These come in different thicknesses.

3.7.2 Polytetrafluoroethylene (PTFE)

Like polyethylene, PTFE is also an example of a polymer. PTFE is also a simple polymer but made up of many fluoroethylene units -CF_2CF_2-, chemically bonded to one another forming long-chain molecules consisting of thousands of such units. It is also an example of an addition polymer [Kotz91]. These come in different thicknesses. It is often called by the tradename TEFLON®.

PTFE is made into breathable, waterproof membranes manufactured from polyether urethane with high performance stretch and abrasion resistance. Polyether is just a polymer formed by many units of ether, which is also formed by replacing both hydrogen atoms of H_2O by hydrocarbon groups R and R [Ebb90].

H-O-H	R-O-H	R-O-R
Water	an alcohol	an ether

PTFE, or PORELLE®, as it is called by the manufacturer, may be laminated to a wide range of textiles including knitted and non-woven for a wide variety of applications [Sea00]. These also come in different thicknesses. These membranes are investigated in this work as a possible covering for the EICs, since they are waterproof but allow gasses to diffuse through fairly easily. This is so, because these materials have pores that are smaller than water molecules but bigger than gas molecules.

Chapter 4

Response Time and Diffusion Studies for the E-PERM Electret Ionisation Chambers (EIC)

4.1 Introduction

In South Africa, where there are larger numbers of miners working underground than anywhere else in the Western world and miners are exposed to ^{222}Rn concentrations in the range of 1000-2000 Bqm^{-3} [Bot99, Ntw00]. In some mines values much higher than these values are experienced. These miners are exposed to varying concentrations because in different parts of the mine we experience great differences in concentration levels. This necessitates that individual or at least a fraction of the mining population be monitored according to their movements around the mine.

This study seeks to address two problems that have been identified in provisional tests using E-PERM Electret Ionisation Chambers (EIC):

1. The ability of the EIC to respond to the variations in radon concentration.

2. These detectors give unreliable readings when used under wet conditions. So, this study looks at ways of protecting the electrets from being discharged by water. The obvious solution to this problem is to cover them with materials that allow radon (gases) through but keep

water away. The suggested materials are polyethylene, polytetrafluoroethylene (PTFE) and microporous membranes. A method for simple measurement of the diffusion coefficients is described which allows for the effect on the response time to variations in radon concentrations to be considered.

4.2 Response time

The EIC was designed primarily to measure the integrated radon concentration in places where the concentration does not change significantly over the period of exposure such as a week or a month [Rad94]. The situation in the mines is completely different. If a miner were to move around from area to area where the radon concentration could easily change by an amount greater than, say 500 Bqm^{-3} within a period of minutes, then the response time of this detector becomes important.

If it takes a long period for the radon concentration inside the electret ion chamber to be in equilibrium with the concentration outside, then this will cause an underestimate of the integrated radon concentration that the miner has been exposed to, especially if the time spent in an area with very high radon concentration is short compared to this equilibration time.

Consider for example an increase in the ^{222}Rn concentration for a time, t, from t_1 to t_2 as shown in Fig. 4-1.

We wish to study for what periods of $T = t_2 - t_1$, the EIC will respond correctly to the increase in concentration. This problem has been studied by Houle and Brodhead [Hou95], but they considered a repeated change in concentration only, which only considers the average ^{222}Rn concentration. This method does not check if the detectors keep up with the changing concentration.

4.3 Diffusion Studies

As mentioned, some EIC detectors have given unreliable readings when they were used under wet conditions in the mines. The proposed solution is to cover them with materials that keep water out but allow gases such as ^{222}Rn to pass through, as radon is also a gas. This necessitates that a study to find the best material to use be conducted. For different materials the time it

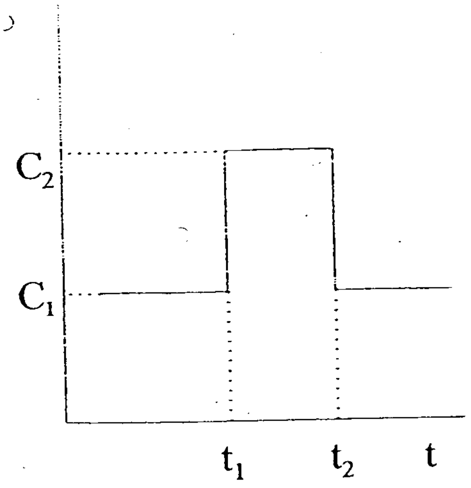


Figure 4-1: Increase in radon concentration for a short period. The question as to whether the E-Perm responds correctly to this change is studied in this work.

takes for radon gas to pass through will be different and will depend on the diffusion coefficient, D, of that particular material.

The study was conducted under laboratory and controlled conditions. The aim is to find the material, which is water-proof, with the highest D value comparable to that of the air. In South Africa and in other countries, a lot of work has been done to measure diffusion through concrete slabs [Bri00, Cul76, Lan82, Zap83].

The common definition of diffusion is that it is the movement of particles from a region of high concentration to a region of low concentration. This process leads to an equalization of concentrations within a single phase.

We also define as usual, the diffusion flow or current, J, of a substance in a mixture with other substances as the amount of this substance passing perpendicularly through a reference surface of unit area during unit time[Jos60]. The definition refers to any mixture, regardless of its state of aggregation. The unit of J is the quantity of substance per area per second.

4.4 The Mathematics of Diffusion

In this study we consider one-dimensional diffusion along the z axes of the Cartesian co-ordinate system.

Let z be the coordinate chosen perpendicular to the reference surface, and C the concentration of the diffusing substance, given as the amount of diffusing substance per volume in the same units as those chosen for the diffusion current. Then Fick's first law of diffusion may be stated in the form [Cra75, Jos60]:

$$J = -D\frac{\partial C}{\partial z} \tag{4.1}$$

where D is the diffusion coefficient for the material under consideration. This mathematical formulation is based on the hypothesis that the rate of transfer of diffusing substance through unit area of a section is proportional to the concentration gradient measured normal to the section.

If we wish to determine the diffusion coefficient by means of equation (4.1) we must find an arrangement in which both, J and $\frac{\partial C}{\partial z}$ are accessible to measurement.

Generally, however, it is not possible to study diffusion under conditions of constant concentration gradient, which necessitates the establishment of a steady state.

We, therefore, have to determine the change of concentration with respect to time caused by diffusion of the radon gas. This requires the use of time-dependent diffusion theory, see below.

In the case where the diffusion is in one direction only, and when we observe the increase in the concentration of the gas within a volume element bounded by two close parallel planes of unit area at z and z + dz, this increase is :

$$\frac{\partial C}{\partial t} = D\frac{\partial^2 C}{\partial z^2} \tag{4.2}$$

which is Fick's second law of diffusion [Cra75, Jos60].

Solutions for equation (4.2) can be found if given adequate initial and boundary conditions. These two equations were first formulated by Fick (1855) by direct analogy with the equations of heat conduction [Cra75].

Now, in a steady state there is no change of concentration with respect to time, that is

$$\frac{\partial C}{\partial t} = 0 \tag{4.3}$$

and it follows from equations (4.2) and (4.3) that,

$$\frac{\partial^2 C}{\partial z^2} = 0 \tag{4.4}$$

and

$$\frac{\partial C}{\partial z} = \text{constant} = c_1 \tag{4.5}$$

from which Fick's first law, equation (4.1), can be justified. From equation (4.5), it follows that

$$C = c_0 + c_1 z \tag{4.6}$$

This is a situation of constant concentration gradient and a linear distribution of concentration exist if D can always be treated as constant.

Figure 4-2: Set-up to measure the diffusion coefficient. The accumulation jar contains three S-chambers and a source. A microporous material covers the jar instead of the usual lid.

4.5 The determination of the Diffusion coefficient using a source of known activity

Consider now a diffusion experiment (which will be described in more detail in section 5.7) where the accumulation jar opening is covered with a material of which the diffusion coefficient is to be measured (see Fig. 4-2). In this problem, we first determine the average concentration of radon in the laboratory in which all diffusion experiments were performed. The average concentration was found to be 40 Bq.m^{-3}. This value will be treated as being the radon concentration outside the accumulation jar, which was described in section 3.4.

4.5.1 Time independent solution

The derivation that follows assumes that we can use Fick's first law, i.e. a steady state exists. This is only correct once a steady state has been established, but it does give more insight into the problem than the time dependent solution which will be discussed later in section 4.5.2.

If we let C_1 be the activity concentration inside the jar and C_2 be the concentration outside the jar, then

$$C_1 = \frac{A(t)}{V_{jar}} \tag{4.7}$$

where $A(t)$ is the radon activity in the jar at time t and V_{jar} is the volume of the jar.

Now, from Fick's first law, the activity diffusing through the material enclosure, Q, is given by,

$$Q = Ja = Da\frac{\triangle C}{\triangle z} \tag{4.8}$$

where a is the area of the effective opening of the jar, $\triangle z$ is the thickness of the material used, D is the diffusion coefficient and $\triangle$C is C_1- C_2, the change in activity concentration between the inside and outside of the jar.

The activity, A_{Rn}, is the number of ^{222}Rn atoms decaying in a second and is given by,

$$A_{Rn} = \lambda_{Rn}N \tag{4.9}$$

where λ_{Rn} is the decay constant of ^{222}Rn which has a value of 2.10 x $10^{-6}s^{-1}$ and N is the number of radon atoms at any given time. The activity concentration outside is

$$C_2 = \frac{A_{Rn}}{V_{out}} \approx 40Bqm^{-3} = \text{constant} \tag{4.10}$$

Now, we define C(t) to be the radon activity concentration in the jar at any time t, so that

$$\triangle C = \lambda_{Rn} S \triangle t / V - Q \triangle t / V - \lambda_{Rn} C \triangle t \tag{4.11}$$

where S is the source strength in Bq. The first term in equation (4.11) describes the creation of new radon atoms by the source, the second term describes the loss of radon atoms by diffusion and the third term the usual loss by radioactive decay.

Substituting for Q from equation (4.8), we get

$$\triangle C = \lambda_{Rn} S \triangle t / V - \frac{Da}{V \triangle z}(C_1 - C_2)\triangle t - \lambda_{Rn} C \triangle t. \tag{4.12}$$

Using equations (4.7) and (4.10) in the above, we get

$$\triangle C = \lambda_{Rn} S \triangle t / V - \frac{Da}{V \triangle z}(C(t) - 40)\triangle t - \lambda_{Rn} C \triangle t. \tag{4.13}$$

From equation (4.13), we get

$$\frac{dC}{dt} = S' - BC \tag{4.14}$$

where $S' = \frac{D'}{V}40 + \lambda S/V$ and $B = \frac{D'}{V} + \lambda_{Rn}$ where $D' = \frac{Da}{\triangle z}$

For practical purposes the activity of the source is constant, so that this equation after rearrangement and integrating becomes

$$\int \frac{dC}{S' - BC} = \int dt \tag{4.15}$$

After integrating and assuming $C = C_0 = 0$ at $t = 0$, we get

$$C(t) = \frac{S'}{B}[1 - e^{-Bt}] \tag{4.16}$$

or

$$C_1(t) = \frac{\lambda S' V}{D' + \lambda V}\left[1 - e^{-\left(\frac{D'+\lambda V}{V}\right)t}\right] \tag{4.17}$$

which gives the concentration of ^{222}Rn atoms as a function of time.

We can arrive at the average activity concentration, C_{a1}, of ^{222}Rn inside the jar by using the fact that it is given by the average activity concentration, that is

$$C_{a1} = \int \frac{C(t)dt}{T} \tag{4.18}$$

which is

$$\begin{aligned} C_{a1} &= \tfrac{S'}{BT}\textstyle\int_0^T \left[1 - e^{-BT}\right] dt \\ &= \tfrac{S'}{BT}\left[t + \tfrac{e^{-Bt}}{B}\right]_0^T \\ &= \tfrac{S'}{BT}\left[T + \tfrac{1}{B}(e^{-BT} - 1)\right] \\ &= \tfrac{S'}{B}\left[1 + \tfrac{1}{BT}(e^{-BT} - 1)\right] \end{aligned}$$

and after substituting for B, we obtain the final expression for the average concentration in the jar:

$$C_{a1} = \frac{S'V}{(D' + \lambda V)}\left[1 + \frac{V}{(D' + \lambda V)T}\{e^{-(D'+\lambda V)T/V} - 1\}\right] \tag{4.19}$$

Let us look at what happens to the expression if we assume C = C_0 at t = 0. In practice C_0 will correspond to the constant outside concentration C_2.

We start with equation (4.15) and integrate from C = C_0 to C and from t = 0 to t.

$$\int_{C_0}^{C} \tfrac{dC}{S'-BC} = \int_0^T dt$$

$$-\tfrac{1}{B}[\ln{(S'-BC)}]_{C_0}^{C} = t$$

$$\ln\left(\tfrac{S'-BC}{S'-BC_0}\right) = -Bt$$

$$S' - BC = (S' - BC_0)e^{-Bt}$$

$$C = \tfrac{S'}{B} - \left(\tfrac{S'}{B} - C_0\right) e^{-Bt}$$

and finally

$$C(t) = C_0 e^{-Bt} + \frac{S'}{B}\left(1 - e^{-Bt}\right) \tag{4.20}$$

From equation (4.20) we can work out the average radon activity concentration, C_{a1}, in the jar:

$$\begin{aligned} C_{a1} &= \tfrac{\int C(t)dt}{T} \\ &= \{\tfrac{C_0}{T}\int_0^T e^{-Bt}dt + \tfrac{S'}{BT}\int_0^T (1 - e^{-Bt})dt\} \\ &= \tfrac{C_0}{T}[-\tfrac{1}{B}e^{-Bt}]_0^T + \tfrac{S'}{B}\left[1 + \tfrac{1}{BT}(e^{-BT} - 1)\right] \\ &= -\tfrac{C_0}{TB}\left[e^{-BT} - 1\right] + \tfrac{S'}{B} + \tfrac{S'}{B^2T}\left[e^{-BT} - 1\right] \\ &= \tfrac{1}{TB}\left(\tfrac{S'}{B} - C_0\right)\left(e^{-BT} - 1\right) + \tfrac{S'}{B} \end{aligned}$$

If we substitute for S$'$and B in the above equation we get, for the average radon activity concentration in the jar

$$C_{a1} = \frac{1}{(D'/V + \lambda)\,T}\left[\frac{(\lambda S + D'40)}{D' + \lambda V} - C_0\right]\left[e^{\frac{-(D'+\lambda V)T}{V}} - 1\right] + \frac{\lambda S + 40D'}{D' + \lambda V} \tag{4.21}$$

Equation (4.21) gives the final form for the average radon concentration in the jar, if we do not ignore the radon already inside the jar at time t =0.

Fig. 4-3 shows the value of the activity concentration as a function of time in the jar for different D$'$ values, varying between 0 and 8 $\times 10^{-8}$ s^{-1}, as calculated from eq. (4.20).

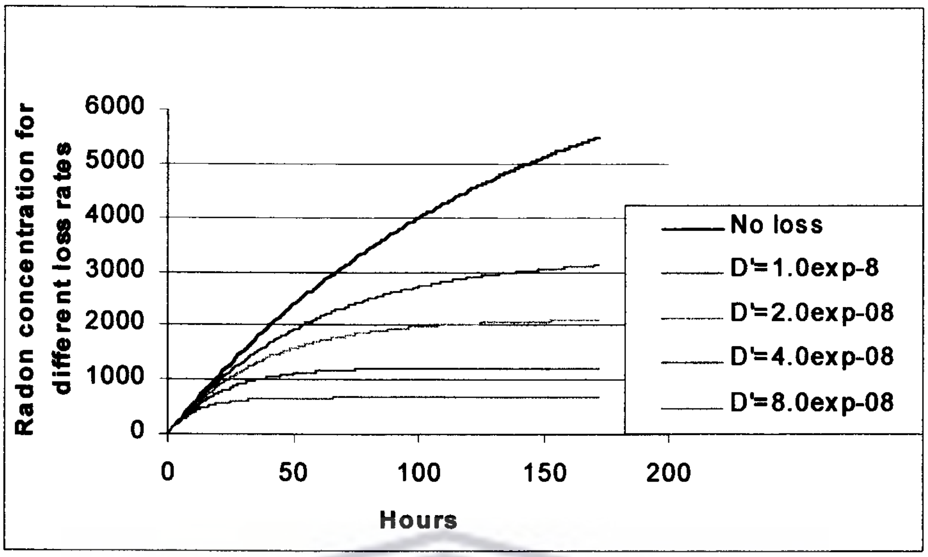


Figure 4-3: The build-up of radon in the jar (Bqm^{-3}) for different values of the diffusion constant (m^2s^{-1}) for a 30 Bq Ra source as calculated from eq. (4.20).

4.5.2 Time dependent Diffusion calculation.

The discussion in the previous section treated the problem of diffusion from the jar in a time independent way. In this section the full time dependent solution will be found. The solution follows the method used in [Van98] based on the methods in [Cra75]. The result shows that the discussion above is accurate for the dimensions used in this experiment and much easier for understanding the diffusion.

Fig. 4-2 shows a jar of volume 4 litre, normally used with the NIST/PERE system to be used as a radon chamber. The jar contains a source of radon(shown) and will contain some radon measurement system, E-Perm's in our case which will measure the average radon concentration over a period. From this average concentration, we wish to calculate the diffusion from the chamber and hence the diffusion coefficient.

Assume that the diffusion of the radon is given by the one-dimensional diffusion equation:

$$\frac{\partial C}{\partial t} = D\frac{\partial^2 C}{\partial z^2} - \lambda C \tag{4.22}$$

This is the usual diffusion equation with a term to account for the decay of the radon. Make the following assumptions.:

1. The concentration on the outside is constant $= 0$. Actually, it will be some small value C_2, but this is an unnecessary complication.
2. Equilibration in the chamber is so fast that we can assume that the concentration is the same everywhere inside. This leads to a boundary condition which links the flux at $z = 0$ to the concentration inside.

We wish to calculate the diffusion through the material which we approximate as a one-dimensional system with a z-axis chosen perpendicular to the surface through which diffusion occurs with $z = 0$ at the bottom of the material and $z = d$ at the top.

Boundary conditions follow from the above assumptions:

At $z = d$

$$C = C_2 \sim 0 \tag{4.23}$$

At $z = 0$

$$\frac{\partial C}{\partial t} = D \frac{A}{V} \frac{\partial C}{\partial z} - \lambda C + S/V \tag{4.24}$$

where

$V =$ the volume of the jar

$A =$ the area of the material on the top of the jar.

$S =$ the strength of the source inside the jar.

This boundary condition follows from the diffusion equation where the second term becomes a first derivative when using the fact that there is no diffusion through the bottom of the jar. the last term needs to be added since there is now a source in the jar which creates an *activity concentration* of S Bq.

Take Laplace transforms of the three equations:

$$p\overline{C} = D\frac{d^2\overline{C}}{d^2z} - \lambda\overline{C} \tag{4.25}$$

$$p\overline{C} = D\frac{A}{V}\frac{d\overline{C}}{dz} - \lambda\overline{C} + \frac{S}{pV} \quad \text{at} \quad z = 0 \tag{4.26}$$

$$\overline{C} = 0 \quad \text{at } z = d \tag{4.27}$$

The solution of the first of the three equations is

$$\overline{C} = F\cos kz + G\sin kz \tag{4.28}$$

where $k = \sqrt{-\frac{(p+\lambda)}{D}}$. Note that this uses the fact **yet to be determined** that p (the parameter in the Laplace transform) will be mostly negative.

We now need to find the constants, F and G, by using the boundary conditions:

From equation (4.22) we find that at $z = d$

$$F\cos kd + G\sin kd = 0 \tag{4.29}$$

$$\therefore G = -F\cot kd \tag{4.30}$$

and at $z = 0$

$$p\overline{C} = D\frac{A}{V}\frac{\partial\overline{C}}{\partial z} - \lambda\overline{C} + \frac{S}{pV} \quad \text{at} \quad z = 0 \tag{4.31}$$

$$pF = D\frac{A}{V}Gk - \lambda F + \frac{S}{pV} \quad \text{at} \quad z = 0 \tag{4.32}$$

Eliminate G :

$$pF = -D\frac{A}{V}kF\cot kd - \lambda F + \frac{S}{pV} \quad \text{at} \quad z = 0 \tag{4.33}$$

$$\therefore F\{p + D\frac{A}{V}k\cot kd + \lambda\} = \frac{S}{pV} \quad \text{at} \quad z = 0 \tag{4.34}$$

Therefore

$$F = \frac{S}{pV\{p + D\frac{A}{V}k\cot kd + \lambda\}} \tag{4.35}$$

or

$$F = \frac{S}{pV\{D\frac{A}{V}k\cot kd - Dk^2\}} \tag{4.36}$$

Therefore

$$\overline{C} = \frac{S}{pV\{p + D\frac{A}{V}k\cot kd + \lambda\}}[\cos kz - \cot kd \sin kz] \tag{4.37}$$

The inverse Laplace transform can now be obtained in the usual way [Cra75]

$$C = \sum_{n=1}^{\infty} f(a_n)/g'(a_n)\ e^{a_n t} \tag{4.38}$$

where the a_n's are the roots of $g(p) = 0$, where $g(p)$ is the denominator and $f(p)$ the numerator of $\overline{C}$.

The roots are $p_1 = 0$ and solutions of $\{p + D\frac{A}{V}k\cot kd + \lambda\} = 0$.

Replace from eq. for $k = \sqrt{-\frac{(p+\lambda)}{D}}$:

$$-Dk^2 + D\frac{A}{V}k\cot kd = 0 \tag{4.39}$$

$$\therefore \cot kd = \frac{kV}{A} \tag{4.40}$$

$$\text{or } \tan kd = \frac{A}{kV} \tag{4.41}$$

The roots of this equation tend to the values where cot → ∞ but these make the numerical method tricky to find the zeros since the function is changing so rapidly. It is therefore easier to use the equation in the form:

$$\tan k_n d = A/(k_n V)$$

where $n = 2, 3, 4...$

We can now find C as follows:

First find g': Remember that $p_n = -Dk_n^2 - \lambda$,

therefore

$$\frac{dk}{dp} = -\frac{1}{2Dk} \tag{4.42}$$

and

$$g = pV\{p + D\frac{A}{V}k\cot kd + \lambda\} \tag{4.43}$$

Therefore

$$g' = \frac{dg}{dp} = V\{\} + pV\{1 - \frac{A}{2Vk}\cot kd + \frac{Ad}{2V\sin^2 kd}\} \tag{4.44}$$

$$\therefore g' = pV + DAk\cot kd + \lambda V + pV - \frac{pA}{2k}\cot kd + \frac{pAd}{2\sin^2 kd} \tag{4.45}$$

but at the zeros we have that $\cot kd = \frac{kV}{A}$ and $\frac{1}{\sin^2} = \cot^2 + 1$

$$\therefore g' = pV + DkkV + \lambda V + pV - \frac{p}{2k}kV + \frac{pAd}{2}(\cot^2 kd + 1) \tag{4.46}$$

$$\therefore g' = \frac{3}{2}pV + Dk^2V + \lambda V + \frac{pAd}{2}(\frac{k^2V^2}{A^2} + 1) \tag{4.47}$$

But $p + \lambda = -Dk^2$

$$\therefore g' = \frac{1}{2}pV + \frac{pAd}{2}(\frac{k^2V^2}{A^2} + 1) \tag{4.48}$$

Thus

$$C = \{p_1\text{term}\} + \sum \frac{\cos kz - \frac{kV}{A}\sin kz}{\frac{1}{2}p + \frac{pAd}{2V}(\frac{k^2V^2}{A^2} + 1)} e^{-Dk^2t} e^{-\lambda t} \tag{4.49}$$

For the $p_1 = 0$ term:

$$g'(p) = pV + DAk\cot kd + \lambda V + pV - \frac{pA}{2k}\cot kd + \frac{pAd}{2\sin^2 kd} \tag{4.50}$$

At $p = 0$, this equals:

$$g'(0) = +DAk_1 \cot k_1 d + \lambda V \tag{4.51}$$

since $k_1 = \sqrt{-\frac{\lambda}{D}}$ at $p = 0$

$$\therefore C = \frac{S}{V}[\frac{\cos k_1 z - \cot k_1 d \sin k_1 z}{\frac{DA}{V} k_1 \cot k_1 d + \lambda} + \sum_{n=2}^{\infty} \frac{\cos k_n z - \frac{k_n V}{A}\sin k_n z}{\frac{1}{2}p_n + \frac{p_n A d}{2V}(\frac{k_n^2 V^2}{A^2} + 1)} e^{-Dk_n^2 t} e^{-\lambda t}] \tag{4.52}$$

where $n = 2, 3, 4 ...$and $k_1^2 = -\lambda/D$

The functions in the first term becomes hyperbolic functions due to the fact that $k_1$is imaginary.

Thus

$$\therefore C = \frac{S}{V}[\frac{\cosh\sqrt{\lambda/D}z - \coth\sqrt{\lambda/D}d \sinh k_1 z}{\frac{DA}{V}\sqrt{\lambda/D}\coth\sqrt{\lambda/D}d + \lambda} + \sum_{n=2}^{\infty} \frac{\cos k_n z - \frac{k_n V}{A}\sin k_n z}{\frac{1}{2}p_n + \frac{p_n A d}{2V}(\frac{k_n^2 V^2}{A^2} + 1)} e^{-Dk_n^2 t} e^{-\lambda t}] \tag{4.53}$$

(using $\sin ix = i \sinh x$ and $\cos ix = \cosh x$ etc.)

We need to look at the first two terms. The $n = 2$ term dominates the rest due to the denominator becoming larger and larger and the numerator has a maximum value.

The $n = 2$ term is found from the solution of $\tan k_2 d = A/(k_2 V)$

Since d is very small ($10^{-4} m$) and $A/V = 72 \times 10^{-4} m^2 / 4.7 \times 10^{-3} m^3 = 1.531\,9 m^{-1}$

Numerical calculation shows that $k_2 \cong 122$ m^{-1}and therefore $k_2 d = 0.012$.

Hence kd is small and $\tan kd \cong kd$.

Thus $k_2^2 = -(p_2 + \lambda)/D = \frac{A}{Vd}$ or

$$p_2 = -\frac{DA}{Vd} - \lambda = -\frac{1}{V}\{\frac{DA}{d} + \lambda V\} = -\frac{1}{V}\{D' + \lambda V\} \tag{4.54}$$

in the notation of the previous section.

For comparison to the time *independent* solution, we should compare this expression at $z = 0$ with equation (4.16), since the concentration in the jar is the same everywhere from our assumptions.

From the above good approximations, we get:

$$\begin{aligned}
C(0,t) &= \frac{S}{V}[\frac{1}{\frac{DA}{V}\sqrt{\lambda/D}\coth\sqrt{\lambda/D}d + \lambda} + & (4.55)\\
&\quad + \frac{1}{\frac{1}{2}p_2 + \frac{p_2 Ad}{2V}(\frac{V}{Ad} + 1)}\exp(-\frac{DA}{Vd} - \lambda)t] & (4.56)\\
&= \frac{S}{V}[\frac{1}{\frac{DA}{V}\sqrt{\lambda/D}\coth\sqrt{\lambda/D}d + \lambda} + \frac{1}{p_2 + \frac{p_2 Ad}{2V}}\exp(-\frac{DA}{Vd} - \lambda)t] & (4.57)\\
&\simeq \frac{S}{V}[\frac{1}{\frac{DA}{Vd} + \lambda} + \frac{1}{p_2}\exp(-\frac{DA}{Vd} - \lambda)] = \frac{S}{D' + \lambda V}[1 - \exp(-\frac{DA}{Vd} - \lambda)t] & (4.58)
\end{aligned}$$

if we use $\coth x \simeq 1/x$ when x is small, substitution of p_2 and using $D' = DA/d$. Inspection shows that this equation is identical to 4.18. Hence the result obtained using 4.18 is perfectly acceptable.

Chapter 5

Experimental Methods and Results for Radon Experiments.

5.1 Introduction

The purpose of this study is to evaluate E-PERMs for personal dosimetry in the mines. We needed to perform several different experiments such as to check ways to keep the E-PERM detectors dry in wet conditions.

In this chapter we describe the experiments that were performed to check on the accuracy of E-PERMs, their use under different conditions and experiments on diffusion.

The following experiments will be described :

1) Use of E-PERMs to measure ^{222}Rn levels in the laboratory.
2) Check on their accuracy in a calibration jar and comparison with RGMs.
3) Calibration check at the CSIR Radon chamber.
4) Experiment at the CSIR to check on L-chambers, including their response time.
5) Diffusion experiments.
6) Response time experiments.

In any radiation study it is vital that the background radiation be considered so that the values of the radiation being measured is measured accurately. This is why it is important that the radon levels in the laboratory be known and compared to the radon and gamma levels that are accumulated during the experiments to see the extent of their influence on the actual measurements. This background radiation, if it is significant, will then be subtracted when doing the calculations.

5.2 Experiments to determine the average radon concentrations in the laboratory

For this study the standard "S" chambers which are described in chapter 3 were used. These chambers were used with short term (ST) electrets and the combination is called SST meaning an "S" chamber and a short term electret. This combination was used because it can measure an integrated radon concentration as low as 300Bq.m^{-3}.day, meaning a shorter time interval was sufficient for collection. The high sensitivity of the SST means that a short exposure time will be able to give a large enough signal to give good sensitivity.

These electrets were read for the initial voltage before the start of each experiment using the Surface Potential Electret Reader (SPER-1). The initial voltage readings were then recorded. The electrets would be screwed onto the "S" chambers and the on/off switch would be set to the on position. Three detectors would be strategically deployed at different positions apart from one another. The reason for this is that radon is not necessarily uniform throughout the laboratory so that the three would give a fair representation of the average radon concentration.

The start time and the temperature of the room was recorded.

The laboratory does not house very strong sources of radon except for the 30 and 5 Bq ^{226}Ra sources which in most instances were in use in the accumulation jar for other experiments. The volume of the laboratory is so large ($\sim 180m^3$) that these sources make a very small contribution to the radon concentration in the laboratory.

The process was repeated for different number of collection times to confirm the consistency of the E-PERM detectors. After a calculated collection period, the experiment would be stopped and the detectors read for the final voltages using the electret voltage reader. Calculations for the integrated ^{222}Rn concentration were done either immediately or at a later stage depending on whether a value was needed immediately or not. The electrets are then screwed back to their cover lids for use later in other experiments.

5.2.1 Results of the Laboratory Measurements

In this section the procedure for calculating the radon concentration is shown for one particular experiment and the results for other experiments are given in a table because the same procedure is used.

In an experiment to measure radon in the room using the SST arrangement, the initial voltage reading, V_I, was 236 V, the final voltage reading, V_F, was 209 V and the time of accumulation, T_A was 7 days. To calculate the radon concentration the procedure below is used.

We first calculate the calibration factor, C.F, using the formula below [Rad94]:

$$C.F. = 1.69776 + 0.0005742 \times \left(\frac{V_I + V_F}{2}\right) \tag{5.1}$$

which will only apply to the SST arrangement.

We then calculate the radon concentration using the formula below:

$$C_{Rn}(\text{in } Bqm^{-3}) = 37 \times [\left(\frac{V_I - V_F}{C.F. \times T_A}\right) - 0.87] \tag{5.2}$$

where 0.87 is the gamma background correction which is only applicable to the SST arrangement, T_A is always in units of days and C_{Rn} is the radon concentration in units of Bq.m^{-3}. The gamma background value is based on the default value of 10 μR, which has been used because experiments in the laboratory have shown this to be a reasonable value for the laboratory that was used. In experiments where the source is used in a jar, the radon concentrations are so high that the gamma correction becomes negligible.

In the above example, the Calibration Factor is found to be 1.8255 and $C_{Rn} = 45.4$ Bq.m^{-3}.

Experimental results for radon in the laboratory are given in Table 5-1.

Table 5-1 Experimental Results for radon concentration in the room.

No of days	V_{in}	V_{final}	concentration in Bq.m^{-3}
4.0	208	195	34 ± 4
7.06	236	209	45 ± 4
8	273	242	45 ± 4
34	687	533	50 ±5
Weighted Average			43±7

The uncertainties in Table 5-1 represent one standard deviation which includes the uncertainty in the initial and final voltages (1V) and the systematic uncertainty in the manufacturing process (estimated at 5%)

The results shown in Table 5-1 show that the concentration in the laboratory does not exceed 50 Bq.m^{-3}. Although these results do not show any correlation between the number of days of exposure and the radon concentration, they do show that the radon in the laboratory is in the range of 30-50 Bq.m^{-3}. This discrepancy may be due to changing conditions in the laboratory, like at times all windows are either closed or open. When windows are open radon will escape and the concentration seen by the detectors will be lower. For convenience throughout this study we have assumed the value 40 Bq.m^{-3}. This value is negligible compared to the values obtained from the measurements in the jar.

5.3 Experiments in the Leak-Tight Jar using the 5 and 30 Bq ^{226}Ra NIST sources

To check whether the radon concentration inside the leak-tight jar does build up with time until it equilibrates and that this set-up could be used for diffusion studies described below, a few experiments with two NIST sources of different activity were performed. The accumulation time (in days) ranged from 1 to 11 days. Three "S" chambers or 5 "L" chambers were used in the jar each experiment.

5.3.1 Results from the jar measurements using the 5 Bq NIST source

The first experiments in the accumulation jar to test reliability of the E-PERMs were done with a NIST source whose activity is 5 Bq. The results obtained are summarised in Table 5-2.

Table 5-2: Results of radon concentrations when the 5 Bq source was used. The calculated values have an uncertainty of 4% as explained in section 3.6. and the experimental uncertainties represent one standard deviation.

Time in days	concentration in Bq.m^{-3}	Calculated values (eq. 3.3) in Bq.m^{-3}
3	290 ± 20	287
4	470 ± 20	361
5	490 ± 20	429
7	550 ± 20	543
9	570 ± 20	617
11	660 ± 20	710

Table 5-2 shows that the radon concentration inside the jar when there is the NIST source builds up with time. The calculated values have an uncertainty of 4% due to uncertainty in the source strength. The measurements are based on exposing 3–5 EIC's in the jar- see the discussion after Table 5-3 for an example of readings.

5.3.2 Results from the jar measurements using the 30 Bq NIST source.

The method to calculate the concentrations is illustrated by the following data for a 2 day exposure:

$V_{initial}$	V_{final}	Rn concentration (Bq m^{-3})
269	248	1260
245	225	1210
360	340	1150
377	356	1200
242	223	1150
Average		1200 ± 50

The results for the experiments performed are presented in Table 5-3 below.

Table 5-3: The results for the radon concentration in the jar when the 30 Bq source was used for different times. The calculated values have an uncertainty of 4% as explained in section 3.6.

Time in days	Concentration in Bq.m^{-3}	Calculated (eq. 3-3) values in Bq.m^{-3}
0.85	680 ± 50	562
2	1200 ± 50	1237
3	1890 ± 90	1755
4.24	2570 ± 110	2320
5.8	2880 ± 120	2926
7.75	3330 ± 130	3550

Five electrets in L-chambers with short term electrets were placed in the jar. The quoted uncertainty represents one standard deviation.

The results in Table 5-3 follow the same pattern as those of Table 5-2. This demonstrates that the E-PERMs give repeatable and reliable measurements with about 5% uncertainty for a 3 day measurement. This indicates that the source and jar system provides a reliable set-up to test the diffusion coefficients as described below.

5.4 Experiments to compare E-PERMs with alpha track devices (RGM)

Presently, in South African mines, the most widely used passive radon monitors are alpha track devices, which go under the name RGM (an acronym for Radon Gas Monitor). These devices are manufactured in South Africa [Par00]. The manufacturer supplies the mines with these for use as passive radon dosimeters. After a certain period of exposure the dosimeters are returned to the manufacturer for reading and then the results are sent to the mines.

In the United States of America and in other parts of the world, the E-PERMs and alpha track devices are widely used for measuring radon in homes.

The experiments to compare the two types of detectors were undertaken to see if the E-PERMs and RGM's give similar readings.

These experiments were conducted under controlled laboratory conditions. A different type of E-PERM, the LST type, was used in these experiments. The L stands for the L chamber described in chapter 3. Its volume is smaller than that of an S chamber. The lowest measurable integrated radon level for this type of arrangement is 2.22 kBq.m^{-3}.day.

The detectors were exposed for varying periods in a calibration jar with a NIST ^{226}Ra standard source of known activity [Col95]. At least five of each type of detector were simultaneously exposed in three experiments for periods of approximately 2, 3 and 6 days. The electrets were read 3 hours after the end of each exposure and final voltage readings were recorded. The RGMs were stored in a spare calibration jar with activated charcoal until all three experiments were complete. The exposed RGMs together with two unexposed detectors were sent away for reading. The unexposed ones were used as control devices and their readings were subtracted from the results to account for radon exposure outside the accumulation jar. This was not needed in the case of the E-PERMs, but a small gamma correction has to be made.

The RGM results were returned for comparison with those of the E-PERMs.

5.4.1 Results of comparison between E-PERMs and RGMs

The results for the three experiments to compare the E-PERMs with RGMs are shown in Table 5-4 below.

Table 5-4: Radon concentration when the two types of detector were simultaneously tested in the accumulation jar.

Time	Delivered value	Electrets	% deviation	RGMs	% deviation
in hours	In Bq.m^{-3}.h	in Bq.m^{-3}.h		in Bq.m^{-3}.h	
46	5.65 $\times 10^4$	5.76 $\times 10^4$	2%	7.08 $\times 10^4$	13%
	unc = 4%	unc = 4%		unc = 4.5%	
72	1.31 $\times 10^5$	1.32 $\times 10^5$	1%	1.48 $\times 10^5$	25%
	unc = 4%	unc = 4%		unc = 3.2	
140	4.31 $\times 10^5$	4.01 $\times 10^5$	-7%	4.70 $\times 10^5$	9%
	unc = 4%	unc = 3%		unc = 8.3%	

The uncertainties in Table 5-4 indicates the statistical uncertainty of one standard deviation based on our measurements using 5 "L"-chambers and 5 RGM's, e.g. for the 72 h exposure:

Electrets				RGM's
V$_i$	V$_f$	Rn C (Bqm^{-3})	Integrated Rn conc.($Bq.m^{-3}.h$)	Int Rn C ($Bqm^{-3}.h$)
453	406	1760	125000	1.34E+05
452	403	1840	131000	1.42E+05
416	367	1870	133000	1.47E+05
460	412	1800	128000	1.49E+05
450	398	1961	140000	1.54E+05
Average		1850±80	132000±5000	148000±7000

The value in Table 5-4 is based on these values multiplied by the exposure time in hours.

Both these radon detectors give values which do not differ too much from the NIST source values. The electret values are closer to the NIST source values. This may be due to a difference in calibration or possibly a background subtraction problem.

5.5 Calibration Experiment in the Radon Chamber at the CSIR's National Metrology Laboratory (NML)

The E-PERMs were mostly tested in the laboratory at the University of the Western Cape (UWC) inside the calibration jar and they proved to be remarkably consistent and sensitive under these conditions. To further confirm their consistency and sensitivity, they were taken to the national radon chamber at CSIR. The pressure there is about $8.68\times10^4 Pa$. This pressure corresponds to a height of about 1330 m above sea level. This radon chamber has a volume of about 60 m^3. Radon is pumped into this chamber up to the desired concentration. The radon concentration inside the chamber does not stay constant, but depends on how often and for how long its doors are opened. This chamber is equipped with a computer facility which keeps track of the fluctuations of the concentration with time by using ML-98 detectors [Mos99]. On the outside walls of the chamber, passive radon dosimeters are placed strategically to monitor leakages from the chamber.

Experiments with a 7 hour exposure and a 2 hour exposure were performed.

For the first part of the experiment, five S chambers with short term electrets (SST) were deployed inside the radon chamber for 7 hours. Throughout that period the average radon concentration inside the chamber was kept at around 6740 Bq.m^{-3}. The E-PERMs were then taken out of the chamber after the planned 7 hours. The electrets were then read after 3 hours.

The second part of the experiment was planned for 2 hours and six S chambers with short term electrets were deployed inside the radon chamber. The average radon concentration in the chamber was about 6440 Bq.m^{-3}. The E-PERMs were taken out after 2 hours.

5.5.1 First CSIR results

The 7 hours experiment was intended for checking if E-PERMs agree with the NML radon chamber . The SST arrangement was used as well here. The following results were obtained:

Initial voltage V_i	Final voltageVf	Callibr. factor	Rn conc. (Bqm^{-3})
669	557	2.050	6710
676	565	2.054	6630
659	547	2.044	6730
668	544	2.046	7440
661	547	2.045	6845
Average (with one standard deviation unc.)			6900 ± 300

The average radon concentration measured was: $C_{Rn} = 6900 \pm 300$ Bq.m^{-3}.After pressure correction which is the factor 1.03 we get 7100 ± 300 Bq.m^{-3}. The chamber radon concentration was 6740 Bq.m^{-3} as given by the chamber reading system. The percentage difference is about 5%.

Another experiment was done for 2 hours. The average radon concentration in the chamber was about 6440 Bq.m^{-3} and the average radon concentration obtained from the electrets was 6800 ±400 Bq.m^{-3}. If the pressure correction is done using the factor 1.03 we obtain 7000±400 Bq.m^{-3}. This value differs from the chamber value by about 9%.

5.6 Further Experiments at the CSIR's National Metrology Laboratory Radon Chamber

The second series of experiments had two different purposes compared to the first:

1) To check on the response time of the electrets using S chambers.
2) To check the response time for the L chambers with short term electrets.

For the response time experiment, ten S chambers with short term electrets were deployed inside the radon chamber. After exactly 30 minutes, all of them were taken out of the chamber.

The times for the experiment to check the sensitivity of the L chambers was performed based on the calculations of the lowest sensitivity of the LST arrangement, which is about 2.22 kBq.m^{-3}.day. For this sensitivity about 128 minutes were needed. For experimental purposes times lesser than this were also used, namely 115 minutes and 86 minutes. In the first, ten LSTs that were deployed, five were intended for 115 minutes and five for 128 minutes. After 115 minutes five were taken out and the last five were taken out after 128 minutes. At the same time that this set was taken out five LSTs were deployed intended for 86 minutes. This set was taken out of the radon chamber exactly after 86 minutes.

All electrets were read after 3 hours of their respective end of exposure time. The calculations for the radon concentrations were not done at the CSIR but in the laboratory at the University of the Western Cape.

5.6.1 Second CSIR results

The experiment consisted of checking the sensitivity of the LST arrangement. In this experiment it was assumed that the radon concentration in the NML radon chamber would be 25000 Bq.m^{-3}. The measured average radon concentration for 128 minutes was 24300 ±1300 Bq.m^{-3}. The percentage difference between this value and the average chamber value of 24900 Bq.m^{-3} was 2.5%. For the 115 minutes exposure the average radon concentration was measured as 24500 ± 800 Bq.m^{-3}. This value differs from the average chamber value by 2%. The 86 minutes exposure gave an average reading of 23100 ± 800 Bq.m^{-3}. During this period the average chamber value was 23000 Bq.m^{-3}. The electret reading differs from the average chamber value by only about 0.4%. These results confirm that the LST configuration gives reliable results.

5.6.2 Final experiment at the CSIR's National Metrology Laboratory Radon Chamber

Further experiments were done at the CSIR at a much higher concentration to look at the response time of L-chambers for 30 minutes and 15 minutes. L-chambers with the standard filter holes plus ones with a modified larger hole were used to see if there is a difference in response time. The 30 minutes exposure for the standard L-chamber gave an average concentration of 231000 ± 14000 Bq.m^{-3} and for the ones with a bigger hole gave an average reading of 216000 ± 16000 Bq.m^{-3}. The 15 minutes exposure gave an average reading for the standard L-chamber of 197000 ± 12000 Bq.m^{-3}, while for the ones with one big hole the average reading was 196000 ± 13000 Bq.m^{-3}. These two types of L-chambers give repeatable and reliable measurements with about 6-7% uncertainty over such short periods of exposure.

These results indicate that the response of the electret ion chambers is fast enough to respond accurately when exposed for the short periods used in these experiments as discussed in section 5.8.

5.7 Diffusion Experiments in the calibration jar

Some studies with E-PERMs revealed that they do not give consistent results when subjected to wet conditions [Bot99]. Water enters through the filtered inlets of the ion chambers and touches the electret surface to cause a voltage drop. The solution to this problem was to enclose the EICs in bags made of materials that will stop water from coming in but allow gases to pass through.

To be able to know which materials would be suitable for use, diffusion through these materials had to be studied in order to see which material had a high diffusion coefficient.

The method used was to cover the calibration jar's opening with the different materials with the EICs and the ^{226}Ra NIST source inside the jar. The accumulation time for each sample of material used was different. Some materials used, allowed radon to escape so fast that there was no measurable reading. For such materials, the thickness had to be increased in order to get a fair reading.

In all the experiments the SST arrangement was used with three chambers at each accumulation because the jar's volume allows for three only. The detectors would be taken out of the jar after some accumulation time. These electrets would also be read after three hours of end of exposure time.

5.7.1 Results of the diffusion studies for the various materials

In this section the results of the diffusion studies for various materials are presented and discussed. There are only a few earlier studies on radon diffusion through polymers, e.g. [Jha82], [Lab92] and [Van98]. Most of the other studies to determine the diffusion coefficient of ^{222}Rn concentrated on building materials such as concrete [Rog94, Gad95].

Polyethylene

In this study three different thicknesses, 25μm; 50μm and 75μm, of polyethylene were tested.

The average radon concentration in the jar was obtained from the electret readings as in section 5-2. These values were compared to the calculated values by creating an EXCEL spreadsheet which uses the formula given in eq. (4.21):

$$C_{a1} = \frac{1}{(D'/V + \lambda)T}\left[\frac{(S + D'40)}{D' + \lambda V} - C_0\right]\left[e^{\frac{-(D'+\lambda V)T}{V}} - 1\right] + \frac{\lambda S + 40D'}{D' + \lambda V} \qquad (5.3)$$

This equation cannot be solved explicitly for D', hence different values for D' were used until a result close to the measured one was obtained.

The results obtained for the diffusion coefficients of the 25μm polyethylene ranged from 1.7 × 10^{-11} to 5.9 × 10^{-11}m^2.s^{-1}. The value obtained by van der Spoel, for his polyethylene foil whose thickness was (2.33 ± 0.02) × 10^{-4}m, was

(1.02 ± 0.04) × 10^{-11}m^2.s^{-1} [Van98]. The ratio of these two thicknesses is 9.3 : 1.

The measured values for the 50μm polyethylene sheet were in the range 1.2 × 10^{-10} to 9.0 × 10^{-11}m^2.s^{-1}. The results obtained for the 75μm polyethylene range from 2.2 × 10^{-10} to 3.0 × 10^{-10}m^2.s^{-1}.

These discrepancies could be due to many factors, like the effective area used in this study was about 0.0072m^2 and that used by van der Spoel was 0.0314m^2. The other factor could be that the materials used had pinholes in them, which would increase the value for the diffusion coefficient, D. All the experiments in this study were performed under temperatures maintained at 18^{0}C, while van der Spoel's experiments were performed under similar conditions at temperatures of 20 ± 2 ^{0}C. There is also no indication as to whether the same type of polyethylene was used.

Microporous membranes (PTFE)

The measured values for these materials were in the range of 3.6×10^{-8} to 4.6×10^{-8}m^2.s^{-1}. Values for the average radon concentrations were obtained and substituted in the formula, which will then give the D$'$ value. From the D$'$ value the D value was calculated using this relationship:

$$D' = \frac{DA}{\triangle z}$$

where a is the area and $\triangle z$ the thickness which was established in section 4.5.

The diffusion coefficient, D, for the microporous membranes is about a thousand times larger than that of polyethylene. This value and the added advantage that they can be used under wet conditions, especially in the underground mines, makes the microporous materials a very good candidate for selection.

The results for our diffusion coefficients have large uncertainties, but are accurate enough to evaluate the response time in the next sections.

5.8 Discussion of the response time of Electret Ion Chambers.

To study the response time of the EIC when inside a bag, we need to consider the diffusion time of the EIC itself, that is the time needed for the concentration inside the chamber to equilibrate to the concentration on the outside, especially if it is taken to or from an area with a very much higher (or lower) concentration. The standard L-chamber has a filtered hole with area about

1.5 mm^2 and the thickness of the wall of the chamber is about 2 mm. We need to calculate the diffusion through this hole.

Consider the case where a miner moves from the surface where the Radon concentration is low, to an underground stope where the Radon (activity) concentration is very high, C_{out} say, where C_{out} is often around 1000 $Bq\ m^{-3}$.

Using Fick's first law, one sees that the number of Radon atoms inside the chamber will change according to

$$\Delta N = -\lambda N \Delta t + DA\frac{\Delta C}{d}\Delta t \tag{5.4}$$

where the first term corresponds to the usual decay part and the second term describes the diffusion into the chamber where

$D =$ the diffusion constant for radon in air in m^2s^{-1},

$A =$ the area of the hole(m^2) through which the radon is diffusing,

ΔC is the change in the concentration (m^{-3}) between the inside and the outside and

$d =$ the thickness of the hole in m.

Hence

$$\frac{dN}{dt} = -\lambda N + DA\frac{C_{out} - C_{in}}{d} \tag{5.5}$$

where

C_{out} is the (high) radon concentration outside the chamber and

C_{in} is the radon concentration inside the chamber.

But $C_{in} = \frac{N}{V}$ thus

$$\frac{dC_{in}}{dt} = -\lambda C_{in} + DA\frac{C_{out} - C_{in}}{Vd} \tag{5.6}$$

(Note that here, as in most of the thesis the radon concentration can be interpreted as *activity* concentration.)

This first order differential equation can be solved by separation of variables in the usual way; to give

$$C_{in} \approx C_{out}(1 - e^{-t/\tau}) \tag{5.7}$$

where

$$\tau = \frac{Vd}{DA}. \tag{5.8}$$

if we use the fact that $\lambda \ll \frac{Vd}{DA}$ as is borne out by the values in practice. (If this were not so, the response time of the electret would have been of the order of the 3.8 days half life of radon)

The value of τ can be used as an indication of the response time of the EIC, since it corresponds to the time during which the concentration inside becomes $C_{out}(1 - 1/e)$

For the dimensions of the L-chamber given above,

$$\begin{aligned} \tau &= \frac{Vd}{DA} \approx \frac{50cm^3 \times 2mm}{10^{-5}m^2s^{-1} \times \pi \times (0.7mm)^2} && (5.9) \\ &= \frac{50 \times 10^{-6} \times 2 \times 10^{-3}}{10^{-5} \times \pi \times (0.7)^2 \times 10^{-6}} && (5.10) \\ &\approx 6500s \approx 100 \text{ minutes} && (5.11) \end{aligned}$$

This would imply that the chamber *itself* has a diffusion time which is relatively long compared to times a miner would spend in a certain area. This could lead to an underestimate of the dose if a miner spent only an hour in a high radon concentration area. However, the experiment in the radon chamber at the CSIR where the L-chambers with small and large holes were compared (see section 5.6.2) , showed essentially NO difference between the standard L-chamber and one with larger holes. This implies that the change in radon concentration inside the hole and hence inside the chamber, is not driven by the diffusion through the hole, but much more strongly by the flow of air currents due to small pressure differences, often called advective transport [Rog91].

The influence on the response time of the L-chamber by covering it with a bag, leads to a similar derivation as for the one above, except that the response time τ_{bag} will depend on the time for the radon to diffuse through the bag to the space between the bag and the L-chamber. The relevant quantities are now V, the volume of this space,d, the thickness of the bag material, A, the effective area through which the radon can diffuse and D is the diffusion constant for the bag material. Measurement of the physical situation, gives the following very approximate quantities for the case where the EIC is tightly wrapped in a bag of thickness 0.1 mm and made out of the microporous material for which we measured a diffusion constant for radon of about $4 \times 10^{-8} m^2 s^{-1}$

$$\tau = \frac{Vd}{DA} \approx \frac{10^{-5} \times 10^{-4}}{4 \times 10^{-8} \times 20 \times 10^{-4}} \approx 12.5s$$

Thus the diffusion time through the barrier is much smaller than the pure diffusion through the hole, and will not have a large impact if the bag is made of a material such as in the example above. Note, however, that polyethylene with a diffusion constant which is 1000 times smaller, WILL have a noticeable impact.

One further aspect which needs to be investigated, is the effect of the bag on the flow of air inside the hole, but that is beyond the scope of this thesis.

Chapter 6

Conclusions and Recommendations

In this study methods to measure 1) radon concentration in the laboratory, 2) the difference between the EICs and the RGMs readings, 3) the response time of the EICs to rapidly changing radon concentrations, 4) the reliability of the EICs and 5) the diffusion coefficients for various materials, were developed and used.

From the results of the experiments described in this thesis, one can come to some conclusions about the EIC method:

1. The EIC system is accurate and sensitive to radon exposure.
2. The EIC system provides consistent, repeatable results that have, on average, a 5% error.
3. The response time of the EIC's is fast enough for practical use in the mines. The experiments at the CSIR showed that the response time for "L" chambers is less than 15 minutes, which means that the concentration inside the EIC will reach the outside value if the miner spends 15 minutes in the area. Further experiments are required to find the time for which the response time is not adequate.
4. The problem of the electrets getting wet can be overcome.
   The diffusion coefficient of a particular material depends largely on properties of that material. The response time of the detector depends on the volume of the bag, the area through which diffusion occurs, the diffusion coefficient of the bag and the thickness of the bag. If either the volume or the thickness are increased, then the EIC will respond

after a longer period. On the other hand using a material with a large diffusion coefficient will shorten the EIC's response time. The microporous material investigated in this work provides a practical material with the desired diffusion coefficient that does not affect the response time adversely.

This system can be recommended for use in the mines as passive radon dosimeters, where mine workers are reported to be exposed to extremely high and rapidly changing radon concentrations. The EICs can be enclosed in bags made from the microporous breathable fabrics, which in this study have been tested to have high diffusion coefficients. The EIC's, will then perform well even under wet conditions.

Appendix A

Calibration Equations and Correction For Background Gamma Radiation

A.1 Calculation Of Radon Concentration

To obtain the average radon concentration, a two-step calculation is required to convert the initial and final electret readings and the exposure period into a radon concentration value. First, the calibration factor (CF) must be calculated using equation A.1 below. The $\left(\frac{V_I+V_F}{2}\right)$ factor in the equation represents the average of the initial and final voltage readings of the electret for a particular radon measurement. This factor is also called the midpoint voltage (MPV).

$$CF = A + B \times \left(\frac{V_I + V_F}{2}\right) \tag{A.1}$$

where A and B are constants for a particular configuration, and V$_I$ and V$_F$ are the initial and final electret voltages. The second step is to calculate the radon concentration using the calculated calibration factor in equation A.2 below.

$$C_{Rn} = \frac{V_I - V_F}{CF \times T_D} - BG \tag{A.2}$$

where C_{Rn} , which when multiplied by 37, gives the average radon concentration given in units of Bq.m^{-3}, T_D is the exposure period in units of days, BG is the environmental gamma background radiation, which when multiplied by 37, is given in units of radon concentration Bq.m^{-3}. The environmental gamma background radiation can be measured using the E-PERMs. This is done by enclosing the E-PERMs in either the mylar bags or aluminium cans to keep radon away. In the US, the Environmental Protection Agency (EPA) has published a listing of average gamma radiation levels in each of the states there.

A.2 Calibration Equations and Corrections for Background Gamma Radiation for the Various E-PERM Types

In this section the proper calibration equations for calculating average radon concentrations for the various combinations of chamber and electret types are given. The manufacturer of the E-PERMs, Rad Elec Inc., has made available a software package, called RADSYS, which does these calculations automatically when the input data (V_I, V_F, altitude and exposure period) are entered. Rad Elec Inc. also provides the E-PERM users with hand-held pocket computer to use in the field for computing the calculations. The radon concentration for the different arrangements are given by eq. 5.2

$$C_{Rn}(\text{in } Bqm^{-3}) = 37 \times [\left(\frac{V_I - V_F}{C.F. \times T_A}\right) - \text{Gamma correction}] \tag{A.3}$$

where the calibration factors are given below.

Short-Term (ST) Electret in Standard "S" Chamber (SST)

$$CF = 1.69776 + 0.0005742 \times \left(\frac{V_I + V_F}{2}\right) \tag{A.4}$$

and gamma correction of .0.87

Long-Term (LT) Electret in Standard "S" Chamber (SLT)

$$CF = 0.14 + 0.0000525 \times \left(\frac{V_I + V_F}{2}\right) \tag{A.5}$$

and gamma correction of 0.87 .

Short-Term Electret in "L" Chamber (LST)

$$CF = 0.26127 + 0.0001386 \times \left(\frac{V_I + V_F}{2} \right) \tag{A.6}$$

and a gamma correction of 1.2.

Long-Term Electret in "L" Chamber (LLT)

$$CF = 0.02382625 + 0.00001120 \times \left(\frac{V_I + V_F}{2} \right) \tag{A.7}$$

and a gamma correction of 1.2.

Short-Term Electret in "H" Chamber (HST)

$$CF = 7.2954 + 0.004293 \times \left(\frac{V_I + V_F}{2} \right) \tag{A.8}$$

and the gamma correction here is 0.7 .

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