



**University of Benghazi  
Faculty of Science- Department of Physics**

**Study of radon exhalation from building materials used in  
Libya with the alpha track detector**

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قال تعالى: □ **قَالُوا سُبْحَانَكَ لَا عِلْمَ لَنَا  
إِلَّا مَا عَلَّمْتَنَا إِنَّكَ أَنْتَ الْعَلِيمُ الْحَكِيمُ** □

**اية 32 من سورة البقرة**

## **Dedication**

To my parents.....

My brothers and sisters.....

My husband and my son.....

My dear friends.....

My teachers and souls of martyrs Benghazi...

## Acknowledgement

*My great thankful to my guide Dr. Saad A. Abdallah for his advices and directions to this work. My supper thanks to my supervisor Dr. Nagi A. Hussien for his assistance support and useful notes to complete my project. A huge thankful to both helped me directly or indirectly during my scholarship journey. Last but not least my thanks to my husband for his patience and kind support as well as to my parents.*

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### *List of abbreviations*

NCRP.....	National Council for Radon Protection
Bq.....	Becquerel
SSNTDs.....	Solid State Nuclear Track Detectors
USEPA.....	US Environmental Protection Agency
Ra.....	Radium
Rn.....	Radon
UNSCEAR.....	United Nation Scientific Committee on the Effect of Atomic Radiation
ICRP.....	International committee on Radiological Protection
WLM.....	Work Level Month
LPG.....	Liquid Petroleum Gas
PAEC.....	Potential Alpha Energy Concentration
$E_x$ .....	Exhalation rate
$E_m$ .....	Emanation rate
MPY.....	Million Person per Year
WHO.....	World Health Organization
TLDs.....	Thermo luminescent Dosimeters
EERC.....	Equilibrium Equivalent Radon Concentration
EF.....	Equilibrium Factor
IOF.....	Indoor Occupancy Factor
$C_{Rn}$ .....	Radon concentration
REC.....	Radium Equivalent Content

## Abstract

Radon concentration and radon exhalation rates have been determined for various different samples of domestic and imported building materials available on the Libyan market for home construction and interior decoration. Radon exhalation rates were measured by the sealed-can technique based on CR-39 NTDs. The results show that radon exhalation rates from some imported building materials used as foundations and for decoration are extremely high, and these samples are the main sources of indoor radon emanation. Radium contents and annual effective doses were also estimated.

The radon concentration levels in the non-dried, dried and wet solid samples were found varying from  $91.5 \pm 3.2$  to  $2265.7 \pm 79.2$ ,  $264.6 \pm 9.3$  to  $2283.3 \pm 79.8$ , and  $90.1 \pm 3.2$  to  $1533.4 \pm 53.6$   $\text{Bq.m}^{-3}$  with a mean value of  $515.7 \pm 18.0$ ,  $698.6 \pm 24.4$  and  $497.3 \pm 17.4$   $\text{Bq.m}^{-3}$  respectively. For dried and wet porous powdery samples those collected from Benghazi, the radon concentration levels were found ranging from  $441.1 \pm 3.5$  to  $906.5 \pm 31.7$  and  $1113.0 \pm 38.9$  to  $3252.5 \pm 113.6$   $\text{Bq.m}^{-3}$  with mean value of  $635.5 \pm 22.2$  and  $1921.1 \pm 67.1$   $\text{Bq.m}^{-3}$ , respectively. Radon exhalation rate in terms of area for non-dried solid samples was varying from  $69.5 \pm 2.4$  to  $1721.2 \pm 60.1$   $\text{mBq.m}^{-2}.\text{h}^{-1}$  with mean value  $391.8 \pm 13.7$   $\text{mBq.m}^{-2}.\text{h}^{-1}$ . In the dried solid samples was found ranging from  $123.7 \pm 4.3$  to  $2847.9 \pm 99.5$   $\text{mBq.m}^{-2}.\text{h}^{-1}$  with mean of  $728.7 \pm 25.5$   $\text{mBq.m}^{-2}.\text{h}^{-1}$ , for the wet solid samples the areal exhalation rates was varied from  $55.3 \pm 1.9$  to  $2258.7 \pm 62.3$   $\text{mBq.m}^{-2}.\text{h}^{-1}$  with mean value of  $546.3 \pm 19.1$   $\text{mBq.m}^{-2}.\text{h}^{-1}$ . In the dried and wet porous powdery materials, the exhalation rates in terms of area was found to varying from  $433.0 \pm 15.1$  to  $889.9 \pm 31.1$ ,  $1092.5 \pm 38.2$  to  $3192.8 \pm 111.5$   $\text{mBq.m}^{-2}.\text{h}^{-1}$  with mean value of  $623.8 \pm 21.8$  and  $1885.8 \pm 65.9$   $\text{mBq.m}^{-2}.\text{h}^{-1}$ , respectively. The radon exhalation rate in terms of mass for dried samples was ranging from  $7.7 \pm 0.3$  to  $85.0 \pm 29.7$   $\text{mBq.kg}^{-1}.\text{h}^{-1}$  with mean of  $42.4 \pm 1.5$   $\text{mBq.kg}^{-1}.\text{h}^{-1}$ . And for wet solid samples was ranging from  $1.5 \pm 0.1$  to  $188.5 \pm 6.6$   $\text{mBq.kg}^{-1}.\text{h}^{-1}$  with mean value of  $33.4 \pm 1.2$   $\text{mBq.kg}^{-1}.\text{h}^{-1}$ , whereas, for powdery samples dried and wet the mass exhalation was varied from  $16.5 \pm 0.6$  to  $33.7 \pm 1.2$ ,  $21.3 \pm 0.7$  to  $61.3 \pm 2.1$   $\text{mBq.kg}^{-1}.\text{h}^{-1}$  the mean value of  $23.8 \pm 0.8$  and  $36.3 \pm 1.3$   $\text{mBq.kg}^{-1}.\text{h}^{-1}$ , respectively.

From the current results, can conclude that the exhalation rates from the wet samples were higher than those from non-dried and dried building material samples. These values do not exceed the reported values of  $57,600 \text{ mBq.m}^{-2}.\text{h}^{-1}$  (UNSCEAR, 2000). The radium equivalent content (REC) values are also lower than the allowed value of  $370 \text{ Bq.kg}^{-1}$  (OECD, 1979). The annual effective dose for non-dried, dried and wet solid samples were in the range of  $194.8 \pm 6.8$ ,  $330.6 \pm 11.6$  and  $259.8 \pm 9.1 \text{ } \mu\text{Svy}^{-1}$ , and with mean value of  $46.2 \pm 1.6$ ,  $86.6 \pm 3.0$ , and  $64.4 \pm 2.3 \text{ } \mu\text{Svy}^{-1}$ , respectively. Also, for the dried powdery samples the annual effective dose was varied from  $51.1 \pm 1.8$  to  $104.9 \pm 3.7 \text{ } \mu\text{Svy}^{-1}$  with mean value of  $73.6 \pm 2.6 \text{ } \mu\text{Svy}^{-1}$  and for the wet powdery samples the annual effective dose was varied from  $128.8 \pm 4.5$  to  $376.5 \pm 13.1 \text{ } \mu\text{Svy}^{-1}$  with mean value of  $222.4 \pm 7.8 \text{ } \mu\text{Svy}^{-1}$ .

## دراسة معدل تحرر غاز الرادون من مواد البناء المستخدمة في ليبيا باستخدام كواشف أثر الفا النووي

### الملخص

غاز الرادون هو غاز مشع حيث ان التعرض لفترات طويلة لهذا الغاز يزيد من خطورة الإصابة بسرطان الرئة. لهذا السبب هناك قلق واهتمام عام عن قياسات الرادون في مواد البناء والترتبة. لقد تم تحديد تركيز ومعدل تحرر الرادون لعينات مختلفة ومتعددة محلية ومستوردة لمواد البناء المتاحة في السوق الليبي المستخدمة في بناء المنازل والتزيين الداخلي. تم قياس معدلات تحرر الرادون باستخدام تقنية الكأس المغلقة التي تعتمد على CR-39 وهي كواشف الاثر النووي للحالة الصلبة. لقد أظهرت النتائج ان معدلات تحرر الرادون من بعض مواد البناء المستوردة والمستخدمة كأساسات بناء وديكورات للتزيين الداخلي عالية جدا , وهذه العينات هي مصادر اساسية لانبعاث الرادون الداخلي. ايضا تم تقييم محتوى الراديوم ومعدل الجرعة السنوية من هذه العينات.

مستويات تركيز الرادون في العينات الغير مجففة والمجففة والمبلولة وجدت تتراوح من  $91.32 \pm 3.2 \text{ Bq.m}^{-3}$  الى  $22265.7 \pm 79.8 \text{ Bq.m}^{-3}$  ,  $264.6 \pm 9.3 \text{ Bq.m}^{-3}$  الى  $264.6 \pm 9.3 \text{ Bq.m}^{-3}$  مع قيمة متوسطة  $2283.3 \pm 79.8$  و من  $90.1 \pm 3.2 \text{ Bq.m}^{-3}$  الى  $1533.4 \pm 53.6 \text{ Bq.m}^{-3}$  مع قيمة متوسطة  $698.6 \pm 18.0 \pm 24.4$  ,  $515.7 \pm 18.0 \pm 24.4$  و  $497.3 \pm 17.4 \text{ Bq.m}^{-3}$  على التوالي. بالنسبة للعينات المسحوقة المجففة والمبلولة التي تم جمعها من مدينة بنغازي فإن مستويات تركيز الرادون وجدت تتراوح من  $441.1 \pm 3.5$  الى  $906.5 \pm 31.7$  و من  $1113.0 \pm 38.9$  الى  $3252.5 \pm 113.6$   $\text{Bq.m}^{-3}$  مع قيمة متوسطة  $635.5 \pm 22.2$  و  $1921.1 \pm 67.1 \text{ Bq.m}^{-3}$  على التوالي. ان معدل تحرر الرادون في حدود المساحة للعينات الغير مجففة تتراوح من  $69.5 \pm 2.4$  الى  $1721.2 \pm 60.1 \text{ mBq.m}^{-2} \cdot \text{h}^{-1}$  مع قيمة متوسطة  $60.1 \text{ mBq.m}^{-2} \cdot \text{h}^{-1}$  مع قيمة متوسطة  $391.8 \pm 13.7 \text{ mBq.m}^{-2} \cdot \text{h}^{-1}$  بينما في العينات الصلبة المجففة كانت تتراوح ما بين  $4.3 \pm 123.7$  الى  $2847.9 \pm 99.5 \text{ mBq.m}^{-2} \cdot \text{h}^{-1}$  مع قيمة متوسطة  $728.7 \pm 25.5 \text{ mBq.m}^{-2} \cdot \text{h}^{-1}$  , بالنسبة للعينات الصلبة المبلولة فإن معدل تحرر الرادون المساحي يتراوح ما بين  $1.9 \pm 55.3$  الى  $2258.7 \pm 62.3 \text{ mBq.m}^{-2} \cdot \text{h}^{-1}$  مع قيمة متوسطة  $546.3 \pm 19.1 \text{ mBq.m}^{-2} \cdot \text{h}^{-1}$  في المواد المسحوقة المجففة والمبلولة , فإن معدل تحرر

الرادون المساحي وجد يتراوح من  $433.0 \pm 15.1$  الى  $889.9 \pm 31.1$  ,  $1092.5 \pm 38.2$  الى  $1885.8 \pm 65.9$  و  $623.8 \pm 21.8$  مع قيم متوسطة  $3192.8 \pm 111.5 \text{mBq.m}^{-2}.\text{h}^{-1}$  و  $1885.8 \pm 65.9$  على التوالي. معدل تحرر الرادون في حدود الكتلة للعينات الصلبة المجففة وجدت تتراوح من  $7.7 \pm 0.3$  الى  $85.0 \pm 29.7 \text{mBq.kg}^{-1}.\text{h}^{-1}$  بقيمة متوسطة  $42.4 \pm$  و  $1.5 \text{mBq.kg}^{-1}.\text{h}^{-1}$  وبالنسبة للعينات الصلبة المبلولة وجدت تتراوح من  $1.5 \pm 0.1$  الى  $188.5$  و  $6.6 \text{mBq.kg}^{-1}.\text{h}^{-1}$  بقيمة متوسطة  $33.4 \pm 1.2 \text{mBq.kg}^{-1}.\text{h}^{-1}$  , بينما للعينات المسحوقة المجففة والمبلولة فإن معدل تحرر الرادون في حدود الكتلة وجد يتغير من  $16.5 \pm 0.6$  الى  $33.7$  و  $1.2$  ,  $21.3 \pm 0.7$  الى  $61.3 \pm 2.1 \text{mBq.kg}^{-1}.\text{h}^{-1}$  بقيم متوسطة  $23.8 \pm 0.8$  و  $36.3 \pm$  الى  $1.3 \text{mBq.kg}^{-1}.\text{h}^{-1}$  على التوالي.

بالنسبة للنتائج الحالية يمكننا ان نستنتج ان معدلات التحرر من العينات المبلولة كانت هي الاعلى من بين كل عينات مواد البناء الغير مجففة والمجففة. هذه القيم لا تتجاوز القيم المسجلة دوليا وهي (UNSCEAR, 2000)  $57,600 \text{mBq.m}^{-2}.\text{h}^{-1}$  . وكذلك بالنسبة لقيم محتوى مكافئ الراديوم (REC) فهي اقل من المسموح به وهي (OECD, 1979)  $370 \text{Bq.kg}^{-1}$  . الجرعة السنوية المؤثرة بالنسبة للعينات الغير مجففة والمجففة والمبلولة وجدت في المدى  $194.8 \pm 6.8$  ,  $330.6 \pm$  و  $11.6$  و  $259.8 \pm 9.1 \mu\text{Svy}^{-1}$  , و بقيم متوسطة  $46.2 \pm 1.6$  ,  $86.6 \pm 3.0$  و  $64.4 \pm 2.3$  ,  $\mu\text{Svy}^{-1}$  على التوالي . كذلك بالنسبة للعينات المسحوقة المجففة فإن الجرعة المؤثرة السنوية كانت تتراوح من  $51.1 \pm 1.8$  الى  $104.9 \pm 3.7 \mu\text{Svy}^{-1}$  بقيمة متوسطة  $73.6 \pm 2.6 \mu\text{Svy}^{-1}$  وبالنسبة للعينات المسحوقة المبلولة فإن الجرعة السنوية المؤثرة كانت تتغير من  $128.8 \pm 4.5$  الى  $376.5 \pm 13.1 \mu\text{Svy}^{-1}$  بقيمة متوسطة  $222.4 \pm 7.8 \mu\text{Svy}^{-1}$ .

# Chapter 1

## Introduction

The individual exposure to ionizing radiation from several natural sources exceeds all manufactured sources. This exposure is continuous and considered as one of the features of life on earth. The two main contributors to natural radiation exposure are the radioactive nuclides those created in the earth's crust and they present everywhere in the environment, the high-energy cosmic ray particles bombarded the earth's atmosphere. Both external and internal exposures can arise from these sources. These exposures reviewed in previous reports of the United Nation Scientific Committee on the Effect of Atomic Radiation (UNSCEAR 1993).

External exposure arise from earthly radionuclides existing at trace levels in all soils. The specific levels related to the type of rocks from which the soil originated. Higher radiation levels connected to igneous rocks, such as granite, and lower levels with sedimentary rocks. There are exceptions, however, as some shales and phosphate rocks have relatively high content of radionuclides. Many studies to determine the background levels of radionuclides in soils have been done, which they related to the absorbed dose rates in air. All of these spectrometric measurements show that the three components of the external radiation, clearly from the gamma-emitting radionuclides in the  $^{238}\text{U}$  and  $^{232}\text{Th}$  series and  $^{40}\text{K}$ , make almost equal contributions to the externally incident gamma radiation dose to individuals (UNSCEAR 2000).

Internal exposures occurs from the intake of earthly radionuclides by inhalation and ingestion. Doses by inhalation result from the presence in ambient air of dust particles carrying radionuclides of the  $^{238}\text{U}$  and  $^{232}\text{Th}$  decay chains. The dominant component of inhalation exposure is the radon gas. Doses by ingestion are mostly due to  $^{40}\text{K}$  and to the  $^{238}\text{U}$  and  $^{232}\text{Th}$  series radionuclides present in foods and drinking water (UNSCEAR, 2000). The internal exposure due to radon and its radioactive daughters, results in the largest contribution to the average effective dose received by human beings (UNSCEAR, 1982). The worldwide average annual effective dose from ionizing radiation from natural sources is estimated to be 2.4 mSv, of which about 1.0 mSv is

due to radon exposure (UNSCEAR, 2000) and can contribute to occurrence of lung cancer. Therefore, it is important to measure the radon levels in dwellings to assess the radiological consequences. Even more important is the knowledge of the amount of natural radioactivity present in the materials, which are used in the construction of these dwellings. The amount of activity present in building materials will decide its use in the construction of homes (Senthilkumar et al., 2014).

The most exposure levels to radon are not due to technology or some other human activities. Radon is everywhere in our natural environment and its presence in the environment is older than life itself. Even human exposure to indoor radon is not new, early humans living in caves most certainly experienced elevated levels of indoor radon. Recognition of the possible health consequences associated with exposure to radon is also not new, as evidence by the studies of underground miners. From such studies, various estimates of the number of lung cancer deaths that might be attributable to radon exposure have been made (Cothorn and Smith, 1987).

The radioactive decay products of  $^{238}\text{U}$ ,  $^{232}\text{Th}$  and  $^{235}\text{U}$  are ( $^{222}\text{Rn}$ ,  $^{220}\text{Rn}$  and  $^{219}\text{Rn}$ ) respectively, they exist in various concentrations in earth's crust.  $^{220}\text{Rn}$  or thoron from the  $^{232}\text{Th}$  series, and  $^{219}\text{Rn}$  from  $^{235}\text{U}$  have very short half-lives 55.61 s and 3.96 s, respectively, and they are negligible significance compared to  $^{222}\text{Rn}$  in the  $^{238}\text{U}$  series. Uranium has a half-life of 4.5 billion years and its intermediate transformation products,  $^{230}\text{Th}$  and  $^{226}\text{Ra}$ , the immediate parent of  $^{222}\text{Rn}$ , have half-lives of 75,380 and 1600 y, respectively. In natural soils,  $^{230}\text{Th}$  and  $^{226}\text{Ra}$  are in radioactive equilibrium with uranium, thus a perpetual source of radon exists naturally even though its half-life is only 3.82 d. whereas, uranium and its intermediate products are solids and remain in the soils and rocks where they originate.  $^{222}\text{Rn}$  is a radioactive noble gas, which diffuse through soil to zones of low pressure such as homes. Its 3.82-day half-life is long enough for it to diffuse into and build up in homes unless they are constructed in ways that prevent entry of radon gas, or provisions are made to remove the radon. Once radon accumulates in a home it will undergo radioactive transformation, however, the resulting transformation products are no longer gases but are solid particles, which due to an electrostatic charge, become attached to dust particles that are inhaled by

occupants, or the particles can be inhaled directly. Because they are electrically charged, the particles readily deposit in the lung, and since they have half-lives of the order of minutes or seconds their transformation energy is almost certain to be deposited in lung tissue (Martin, 2013).

Radon transformation products commonly called daughters emit alpha particles with energies ranging from 6 to 7.69 MeV. Because these alpha particles (helium nuclei) are massive and highly charged, this energy is delivered in a huge jolt to the cellular structure of the surface of the bronchi and the lung, which damages and kills these cells. Unlike many pollutants that were control by standards, lung cancer has observed directly in humans exposed to radon. Over 300 years ago, before radioactivity was known to science, it was common for feldspar miners in Germany and Czechoslovakia to contract and die from a mysterious disease; it is now known that this mysterious disease was lung cancer caused by the short-lived alpha-emitting transformation products of radon continuously emanating from the uranium in the rocks (Martin, 2013). Similarly, in the USA and Canada there is a direct epidemiological association between uranium miner exposure to radon and lung cancer, evidence so compelling that vigorous controls have been used to reduce the concentrations of radon and its transformation products in uranium mines ( Martin, 2013).

The noble gas isotope  $^{222}\text{Rn}$  was formed by the decay of  $^{226}\text{Ra}$ . As  $^{226}\text{Ra}$  is one of the nuclides formed in the disintegration series from  $^{238}\text{U}$ , the amount of radon formed in rocks and soils depends on their uranium content. But that alone does not decisive in determining the radon concentration of air or water in the pores of the soil. It was determined also, by the extent to which the radon atoms formed actually emanate from the mineral grains and whether radon can leave the pore space either by diffusion or together with flow of air or water. In addition, radon concentration in the soil air is significantly affected by the occurrence of water in the pores (Durrani and Ili'c, 1997).

Building materials cannot always be clearly isolated as a source of radon, because often-local building materials are derived from similar rock to that underlying the house. However, particularly in high-rise buildings and those with granite facing, radium- bearing structural materials can be identified as a source of human exposure.

Initially, this was seen primarily as a gamma-ray exposure problem, because the gamma-emitting radon progeny are trapped in solid, concern has been expressed that tight sealing of wall surfaces would result in a significant, and possibly hazardous, increase in external gamma exposure from the trapped radionuclides (Culot et al., 1976).

The natural radiation exposure of the general population is the dominant component of radiation dose inhaled from decay products of  $^{222}\text{Rn}$ . Monitoring in various countries yields average residential  $^{222}\text{Rn}$  concentration ranging from 10 to 100  $\text{Bq}\cdot\text{m}^{-3}$ . For a country such as the United States, with an average of about 40  $\text{Bq}\cdot\text{m}^{-3}$ , the average lifetime risk of lung cancer caused by exposure to radon decay products were estimated to be about 0.3%, causing about 10,000 cases of lung cancer annually among the U.S. population of 235 million. This average risk is more significant than that received on the average from all other natural radiation sources or from medical exposures. The radon dose exceeds by a factor of 10 to 100 the average doses from nuclear power or weapons testing (Nazaroff and Nero, 1988).

Moreover, in any country indoor levels a factor of 10 or higher than the average sometimes occur. In fact, it is the common experience of the radon research community that  $^{222}\text{Rn}$  concentrations in the range of 200-2000  $\text{Bq}\cdot\text{m}^{-3}$  are found with startling frequency. Although the lung cancer risk associated even with an ordinary concentration such as 40  $\text{Bq}\cdot\text{m}^{-3}$  is very large compared with many environmental problems of concern, living for prolonged period at concentration above 200  $\text{Bq}\cdot\text{m}^{-3}$  leads to estimated individual lifetime risks exceeding 1%. The highest values found - more than 200  $\text{Bq}\cdot\text{m}^{-3}$  - have risks even larger than those from cigarette smoking do. However, unlike smokers, those living in unusually high radon concentration are rarely aware of the large risks they may thereby be suffering (Nazaroff and Nero, 1988).

The later work on the developing of methods of controlling the entry of radon into houses from the underlying ground drew attention to the potential for radon progeny buildup in poorly ventilated spaces. It became gradually more obvious that similar conditions could develop in any building that was exposed to significant radon inflow (Schiager and Olson, 1971; Auxier, 1976; NCRP, 1984). Since then it has been overwhelmingly evident that progeny exposure inside houses, particularly well-

insulated ones, may constitute a major portion may involve frequently much higher population exposures than those so carefully monitored and controlled due to effluents from nuclear power plants (Hursh et al., 1965).

When  $^{226}\text{Ra}$  disintegrates, a radon atom and an alpha particle is formed. When alpha particle was eject, a recoil effect arises. This removes the radon atom from the place in mineral lattice or molecular where the radium atom was. The distance the radon atom can then move in mineral grain of normal density has been, from kinematics, put at (0.02-0.07  $\mu\text{m}$ ). It is this very movement of the radon atom that enables radon to emanate from a mineral grain (Fleischer, 1980; Kigoshi, 1971; Tanner, 1980). Radon concentrations in soil air or ground water are determined by the radium concentration in surrounding soil and bedrock, how many of the radon atoms emanate to the pore space from mineral grain in which they are formed, and the porosity and water content of the soil. In addition, the radon concentration is affected by how much of the radon removed or supplied by diffusion and transported by the movement of the soil air or water or of ascending gases such as  $\text{CO}_2$  and  $\text{CH}_4$ . The permeability of the soil and rock are of decisive importance for transporting radon. If the radium concentration, porosity and emanation for type of soil are known, the maximum radon concentration that can occur in the pore air can be calculated (Durrani and Ili'c, 1997).

The radon concentration in air is three time greater than in water in closed volume with equal volume at temperature of  $+15^\circ\text{C}$ , while, at  $0^\circ\text{C}$  the radon concentration in air is twice that in water (Allen, 1976; Tanner, 1991). Radon concentration measurements in situ require detectors to be put in place and left for a sufficient length of time. Measurements can be continuous or discrete by carrying out grab sampling. It can be passive when the radon concentration was measure under natural conditions, radon entering the detection volume by mere diffusion; or active, which involves the pumping of gas into or through a detecting instrument (Durrani and Ili'c, 1997).

$^{222}\text{Rn}$  exhalation from the ground surface depends upon many variables such as the  $^{226}\text{Ra}$  activity concentration and its distribution in soil grains, soil porosity, temperature and moisture, atmospheric pressure, rainfall. Radon from building materials is introduced to the indoors mainly due to diffusion. Generally, there is relation between

indoor radon concentration and the radium content of building materials. The contribution of building materials to the indoor radon concentration is determined with radon exhalation rates measurements. The radon exhalation rate is the amount of radon activity released per unit area per unit time. The radon exhalation rate depends on the radium content, the radon emanation factor and the radon diffusion coefficient in the given material. The emanation factor and the radon diffusion coefficient depend in turn on the physical properties of building materials such as spatial distribution of radium atoms in the mineral grain, texture humidity content, porosity, density and ambient temperature (Stranden and Kolstad, 1984; Nazaroff and Nero, 1988; Durrani and Ili'c, 1997).

When radon decays, it produces radon daughters or radon progeny. The detection and measurements of radon can thus be performed directly on radon itself called radon alone measurements or indirectly on radon and/or its daughters. Radon and several of its daughters decay away by emitting  $\alpha$ -particles.  $^{214}\text{Pb}$ ,  $^{214}\text{Bi}$ ,  $^{210}\text{Pb}$  and  $^{210}\text{Bi}$  are also  $\beta$  emitters and  $\gamma$ -rays accompany their  $\beta$ -emissions. Accordingly, radon detection and measurements can be performed through  $\alpha$ ,  $\beta$ ,  $\gamma$  radiation detection. Therefore, the understanding of the detection and measurement techniques requires a basic knowledge of the interaction of those radiations with matter. There are many techniques available, and they demonstrate the ingenuity of a large number of investigators. One of the following techniques can now be used for detection; solid and liquid scintillation, gamma spectroscopy, Electret, ionization chambers and others (Durrani and Ili'c, 1997).

## 1.1 Literature Review

A few studies of radon exhalation from building materials have been investigated in Libya using the passive techniques based on solid-state nuclear track detectors. Radon exhalation rates were measured from building materials on the university of Benghazi campus by using the can technique, containing CR-39, to estimate the radiation exposure in the atmosphere. It was found that the radon exhalation rates per unit area from some types of building materials such as brick walls, marble ledges and ceramic floor varied from 82.0 to 210  $\text{Bq}\cdot\text{m}^{-2}\cdot\text{h}^{-1}$ , 76.0 to 226.0  $\text{Bq}\cdot\text{m}^{-2}\cdot\text{h}^{-1}$  and 66.0 to 208  $\text{Bq}\cdot\text{m}^{-2}\cdot\text{h}^{-1}$ , respectively. In addition, the average exhalation rate from the brick

walls and marble ledge showed approximately the same value, which was higher than that of the ceramic floors by more than 15% (Saad et al., 2010).

Another study of the radon exhalation rates from soil samples collected from the cities of Benghazi and Al-Marj, located in northeast of Libya. Mass and areal radon exhalation rates, radium content and radon concentration contribute to indoor radon, and annual effective doses were determined. The results indicated mostly normal rates, where, the exhalation rate in Benghazi varied from 30.60 to 460.94  $\text{mBq.m}^{-2} \cdot \text{h}^{-1}$ , with a mean of 216.49  $\text{mBq.m}^{-2} \cdot \text{h}^{-1}$ , whereas in Al-Marj it varied from 58.25 to 506.94  $\text{mBq.m}^{-2} \cdot \text{h}^{-1}$ , with mean of 316.5  $\text{mBq.m}^{-2} \cdot \text{h}^{-1}$ . The radon exhalation rate in terms of mass in Benghazi varied from 1.16 to 17.40  $\text{mBq.kg}^{-1} \cdot \text{h}^{-1}$ , with a mean of 8.08  $\text{mBq.kg}^{-1} \cdot \text{h}^{-1}$ , whereas in Al-Marj it varies from 2.20 to 19.13  $\text{mBq.kg}^{-1} \cdot \text{h}^{-1}$ , with a mean of 12.07  $\text{mBq.kg}^{-1} \cdot \text{h}^{-1}$ . The radium activity in Benghazi also varied from 1.5 to 23.0  $\text{Bq.kg}^{-1}$ , whereas in Al-Marj it varied from 2.9 to 25.3  $\text{Bq.kg}^{-1}$  (Saad et al., 2013).

Measurements of radon exhalation for a total of 50 selected samples of construction materials used in Saudi Arabia were take using radon gas analyzer. These materials included sand, cement, gypsum, hydrated lime, ceramics and granite. It were find that the granite samples were the main source of radon emanation. Thirty-two local and imported granite samples were tested. It was found that the radon exhalation rates per unit area from these granites varied from not detectable to 10.6  $\text{Bqm}^{-2} \cdot \text{h}^{-1}$  with an average of 1.3  $\text{Bq.m}^{-2} \cdot \text{h}^{-1}$  (Al-Jarallah., 2001).

Natural and fabricated building materials commonly used in Egypt were survey for both natural radionuclides content and radon exhalation rate. These included raw as well as construction products. Concentration of natural radionuclides in all samples were determine by  $\gamma$ -ray spectroscopy with HPGe detector. Various samples were fabricate for radon exhalation rates measurements. The seal can-technique has been applied using CR-39 plastic track detectors. The hazard indices of the total natural radioactivity in the studied samples were estimate. The results were compare with corresponding results of different countries and were find to be lying within the average world values. Radon exhalation rate in the studied samples varied between 197 (cement brick) and 907  $\text{mBq.m}^{-2} \cdot \text{h}^{-1}$  (blast furnace slag cement). The results of this survey suggested that, using blast furnace slag cement for pre-coating the internal walls of

buildings in the urban region of Egypt is discouraged and the replacement of clay brick by cement brick will be healthier and safer for the public (Sharaf et al., 1999).

In Tanta University, Egypt, another survey was done to control the natural radiation exposure for the residents of dwelling. In that work, Uranium  $^{238}\text{U}$  and thorium  $^{232}\text{Th}$  content were measured in different building materials samples using two different techniques: first, using Joel high resolution inductively coupled plasma mass spectrometer HR-ICP-MS (plasmamax2); and, secondly, using solid-state nuclear track detectors SSNTDs. There was a good agreement between the measurements of uranium and thorium with both methods. Exhalation rate and emanation coefficient of radon in several building material samples were determined using LR-115 polymeric nuclear track detectors. Radon exhalation rate varied from  $11.83 \text{ mBq}\cdot\text{m}^{-2}\cdot\text{h}^{-1}$  in red clay brick to  $36.86 \text{ mBq}\cdot\text{m}^{-2}\cdot\text{h}^{-1}$  in granite (Moharram et al., 2012).

Fifty-three different samples of drywall, tile and granite available in Canadian market for interior home decoration were collected to measure the radon exhalation rates of those samples. The measured radon exhalation rate was found to be ranging from non-detectable to  $13,000 \text{ mBq}\cdot\text{m}^{-2}\cdot\text{h}^{-1}$ . Slate tiles and granite slabs had relatively higher radon exhalation rates than other decorative materials, such as ceramic or porcelain tiles. The average radon exhalation were  $1250 \text{ mBq}\cdot\text{m}^{-2}\cdot\text{h}^{-1}$  for slate tiles and  $1750 \text{ mBq}\cdot\text{m}^{-2}\cdot\text{h}^{-1}$  for granite slabs of various types and origins. Analysis showed that even if an entire floor was covered with a material having a radon exhalation rate of  $300 \text{ Bq}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$  ( $12500 \text{ mBq}\cdot\text{m}^{-2}\cdot\text{h}^{-1}$ ), it would contribute only  $18 \text{ Bq}\cdot\text{m}^{-3}$  to a tightly sealed house with an air exchange rate of 0.3 per hour (Jing et al., 2010).

In China, using the RAD7 radon monitor, selecting one-story cottage in Shenzhen city area as research object. The range values of soil surface radon exhalation rate was found to vary from 21960 to 100080  $\text{mBq}\cdot\text{m}^{-2}\cdot\text{h}^{-1}$  and it is higher and was about two times that of cement floor of the room that had values ranging from 13320 to 36720  $\text{mBq}\cdot\text{m}^{-2}\cdot\text{h}^{-1}$ , which indicated that the soil was the largest source of indoor radon. The surface radon exhalation rate of cement floor was higher than that of ceramic tile floor that had values ranging from 3960 to 8280  $\text{mBq}\cdot\text{m}^{-2}\cdot\text{h}^{-1}$  and was about 4 times that of the ceramic tile floor (Xiaofeng and Guosheng., 2011).

In India, the radon exhalation rate was measured in number of building materials (bricks, plastered bricks, marble, etc.) commonly used for building construction in India. The effect of internal wall covering (i.e. paints, primer, snocem and wallpapers, etc.) on radon emanation had also been investigated. It could have been seen from the results of that investigation that the exhalation varied from 52.39 to 157.42  $\text{mBq}\cdot\text{m}^{-2}\cdot\text{h}^{-1}$ . The results showed highest radon emanation from unfired bricks while marble gives minimum value (Rawat et al., 1991).

In Pakistan, experimental results concerning the radon exhalation rate from samples of building materials, which were collected from the districts of Muzaffarabad and Neelum Valley, Azad Kashmir. The study aimed at assessing the contribution of building materials towards the total indoor radon exposure to the inhabitants of the studied area. Samples of building materials, namely, soil, sand, gravel aggregates, and bricks were collected from different parts of the districts of Muzaffarabad and Neelum Valley. The results of radon exhalation rate from soil, gravel aggregates, sands, and bricks varied from  $171 \pm 11$  to  $344 \pm 11$ ,  $168 \pm 17$  to  $322 \pm 11$ ,  $366 \pm 8$  to  $649 \pm 8$  and  $184 \pm 14$  to  $231 \pm 14$   $\text{mBq}\cdot\text{m}^{-2}\cdot\text{h}^{-1}$ , respectively (Rafique et al., 2011).

In Iran, thirty-five building material samples were collected from different areas of Ramsar (19 local materials and 16 market available materials). The commonly used market available building materials were marble, granite, brick, travertine, block, concrete, gypsum and mosaic, which were collected randomly from sites where housing and other buildings were under construction and from building material suppliers. Radon exhalation rates from these samples varied from below the lower detection limit up to  $384 \text{ Bq}\cdot\text{m}^{-2}\cdot\text{h}^{-1}$  (Bavarnegin et al., 2012).

In Lebanon, by using CR-39 detector, some building materials such as cement, gypsum and gravel were studied to measure the radon exhaled from these samples. The average areal and mass exhalation rates found to be between  $44 \pm 7$  and  $226 \pm 267$   $\text{mBq}\cdot\text{m}^{-2}\cdot\text{h}^{-1}$  and between  $0.40 \pm 0.07$  and  $20.0 \pm 0.3$   $\text{mBq}\cdot\text{m}^{-2}\cdot\text{h}^{-1}$ , respectively (Kobeissi et al., 2008).

## **1.2 Present Work**

Because of the high risk of the radon gas exhalation and its daughter products from building materials. In this present work, the radon exhalation rates from 39 different types of building materials that used as basic construction and decoration of these houses were measured. The solid state nuclear track detector SSNTD CR-39 was used in this study to measure alpha track and then radon activity concentration, radon exhalation rates and the annual effective dose results from the long term exposure to radon gas from those building materials, were also determined.

## Chapter 2

### Natural Radioactivity and Isotopes

#### 2.1 Nuclear Stability

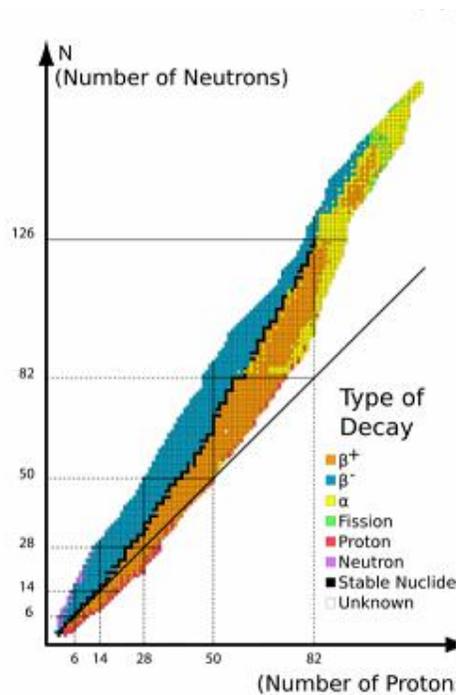
If the number of neutrons in each nucleus of the 270 or so known stable nuclides is plotted as ordinate against the corresponding number of protons as abscissa, a series of points is obtained, as shown in Fig. 2.1, forming a diagonal that represent equal numbers of neutrons and protons. It is seen that, in many stable nuclei of low mass number, the numbers of neutrons and protons are equal or approximately so. In other words, the neutron/proton ratio is exactly or slightly larger than unity. With increasing mass (or atomic) number, however, a nucleus is stable only if it contains more neutrons than protons. Thus, for the heaviest stable nuclei, with atomic numbers of 80 or more, the neutron/proton ratio has increased to about 1.5 (Glasstone and Sesonske, 1986).

Before attempting an interpretation of the foregoing results, it should be noted that certain nuclei exhibit exceptional stability. These so-called magic nuclei contain the magic numbers of 2, 8, 20, 50, or 82 protons or 2, 8, 20, 50, 82, or 126 neutrons, as indicated in Fig. 2.1. Nuclei having magic numbers of both protons and neutrons are said to be "doubly magic". In illustration of the stability associated with the magic numbers, it may be noted that tin, which has 50 protons in its nucleus, exists in ten stable isotopic forms (Glasstone and Sesonske, 1986).

The special stability of the magic nuclei is attributed to the presence of completed (or closed) shells of neutrons or protons (or both). Just as the inert elements helium, neon, argon, etc., which contain closed shells of electrons have great chemical stability. In addition to the magic numbers, which represented closed main shells, there are semi magic numbers, e.g., 6, 14, and 28, which correspond to one or more closed shells plus a completed subshell. The isotopes of the abundant elements carbon and silicon fall in this category (Glasstone and Sesonske, 1986).

A part from the details associated with magic numbers, the general character of Fig.2.1 could be account for by postulating that two types of forces exist between

nucleons. First, there are attractive forces of approximately equal magnitude among the nucleons, i.e., protons attract other protons, neutrons attract neutrons, and protons attract neutrons to about the same extent. These are characteristic intranuclear forces operative over very short distances, about  $10^{-15}$  m only. One consequence of the short range is that total attractive force in a nucleus is roughly proportional to the number of nucleons present. This is the basic reason for the approximate constancy of the binding energy per nucleon over a large range of mass numbers. In addition to the short-range (attractive) forces, there are the conventional electrostatic (or coulomb) repulsive forces between the positively charged protons that are capable of acting over relatively large distances. The total electrostatic repulsive force between all protons in the nucleus is proportional to the square of their number (Glasstone and Sesonske., 1986).



**Fig. 2.1** Neutron-proton plot of the nuclides.

## 2.2 Radioactivity

In 1896, the French scientist Henri Becquerel took note of Roentgen's discovery of x-rays and studied a variety of fluorescent materials. He took a pure double salt, potassium uranyl sulphate, and placed it on a photographic plate wrapped in black paper and exposed it to sunlight. When the photographic plate was developed, it was found to be darkened indicating the uranium salt emitted radiation, which could penetrate the black paper. He repeated the experiment and discovered that the uranium gave off rays like x-rays. He therefore concluded that these invisible radiations were emitted by phosphorescence, which persisted for much longer time than that of the luminous radiation. These radiations were like x-rays, able to produce ionization in the gases through which they passed. These rays do not require any electric potential, while x-rays appear only in the presence of an electric discharge. After many experiments in Becquerel's laboratory, Marie Curie, found that thorium salts were found to give off these rays. In 1898, she was able to isolate a material much more active than uranium from pitchblende by chemical separation. It was named polonium. The next element that has been discovered was radium. Radioactivity is spontaneous and self-disruptive activity exhibited by several of the heavy elements like uranium. The elements of atomic number 84 (polonium) through 92 (uranium) exist in nature, but they are unstable and exhibit the phenomenon of radioactivity (D. K. JHA, 2004).

Radioactive nuclides undergo spontaneous change at a definite rate, which varies with the nature of the nuclide. The unstable nucleus emits a characteristic particle or radiation and is thereby transformed into a different nucleus, which may or may not also be radioactive. In 1899, Rutherford showed that radiations from a radioactive material had a complex composition, consisting of two types of rays, which he called  $\alpha$ -rays and  $\beta$ -rays, which differed in their penetrating powers. In 1909, Villard discovered a third type of radiation, which could not be deflected in a magnetic field, which nevertheless had considerable penetrating power and marked effect on a photographic plate. These radiations were named as  $\gamma$ -rays. Further investigations have shown that  $\alpha$ -rays and  $\beta$ -rays are not so strictly rays. They are corpuscles in nature and so they are described as  $\alpha$ -particles and  $\beta$ -particles. From the direction of the

deflections in magnetic field it appears that  $\alpha$ -particles carry a positive charge, the  $\beta$ -particles carry a negative charge and  $\gamma$ -rays are uncharged. The radiations ( $\alpha, \beta, \gamma$ ) emitted from radioactive sources are also known as Becquerel rays (D. K. JHA, 2004).

## 2.3 Types of Radiation Decay

### 2.3.1 Alpha Decay

Alpha decay is disintegration of massive and unstable nuclei by emission of 2 protons and 2 neutrons as a single alpha (nucleus of helium). Alpha emission is confined to relatively heavy nuclei such as  $^{226}\text{Ra}$ :



Where  $\alpha$  represents the alpha particles. The sum of mass numbers and the sum of atomic numbers after the transition equal the mass and atomic numbers of the parent before the transition. In alpha decay, energy is released as kinetic energy of the alpha particles, and is sometimes followed by energy released during an isomeric transition resulting in emission of gamma ray or conversion electron. Alpha particles are always ejected with energy characteristic of the particular nuclear transition (Hendee et al., 2005).

An alpha transition is depicted in Fig.2.2, in which the parent  $^{226}\text{Ra}$  decays directly to the final energy state (ground state) of the progeny  $^{222}\text{Rn}$  in 94% of all transitions. In 6% of the transitions,  $^{226}\text{Ra}$  decays to an intermediate higher energy state of  $^{222}\text{Rn}$ , which then decays to the ground state by isomeric transition. For each of the transition pathways, the transition energy between parent and ground state of the progeny is constant. In the example of  $^{226}\text{Ra}$ , the transition energy is 4.78 MeV.

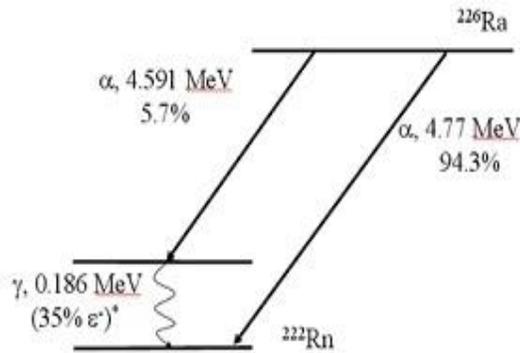


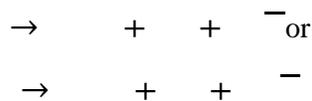
Fig. 2.2 Radioactive decay scheme:  $\alpha$  decay of  $^{226}\text{Ra}$ .

Alpha particles because of positively charge can be reflected in electric and magnetic field. The velocities range from about  $1.4 \times 10^9$  to  $2.2 \times 10^9$  cm/sec, or less than — of the velocity of light (Hendee et al., 2005).

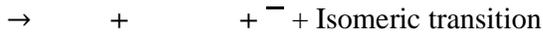
### 2.3.2 Beta Decay

Nuclei with  $n/p$  ratio that is above the line of stability tend to decay by a form of beta ( $\beta^-$ ) decay known as negative beta emission ( $\beta^-$ ). In this mode of decay, a neutron is transformed into a proton, and the  $Z$  of the nucleus is increased by 1 with no change in  $A$ . In this manner, the  $n/p$  ratio is reduced, and the product nucleus is nearer the line of stability. Simultaneously an electron (termed a negative beta particle or electron) is ejected from the nucleus together with natural massless particle, termed a neutrino (actually an "antineutrino" electron decay), that carries away the remainder of the released energy that is not accounted for by the negative beta emission. The neutrino (or antineutrino) seldom interacts with matter and is not important to applications of radioactivity in medicine (Hendee et al., 2005).

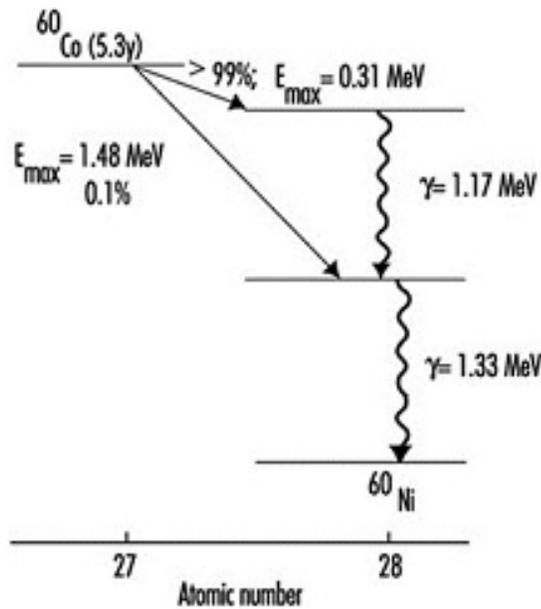
The process of  $\beta^-$  emission may be written as



where  $e^-$  depicts the ejected electron (negative beta particles) and  $\bar{\nu}_e$  reflect the nuclear origin of the  $e^-$ . The symbol  $\bar{\nu}_e$  represents the antineutrino. An example for emission is beta decay equation of  $^{60}\text{Co}$ :



The isomeric transition often accomplished by release of cascading gamma rays of 1.17 and 1.33 MeV. A decay scheme for  $^{60}\text{Co}$  is shown in Fig. 2.3. The transition energy for decay of  $^{60}\text{Co}$  is 2.81 MeV.



**Fig. 2.3** Radioactivity decay scheme: decay of  $^{60}\text{Co}$ .

A discrete amount of energy is released when a  $\beta^-$  is emitted from the nucleus. This energy is depicted as the maximum energy  $E_{\text{max}}$  of  $e^-$ . However, usually are emitted with some fraction of this energy, and the remainder is carried from the nucleus by the antineutrino. The mean energy of the negatron is  $E_{\text{max}}/3$ . An energy spectrum of 0.31 MeV  $E_{\text{max}}$  emitted from  $^{60}\text{Co}$  is shown in Fig. 2.4. Electron energy spectra are specific for each  $\beta^-$  transition in every nuclide by this mode of nuclear transformation (Hendee et al., 2005).

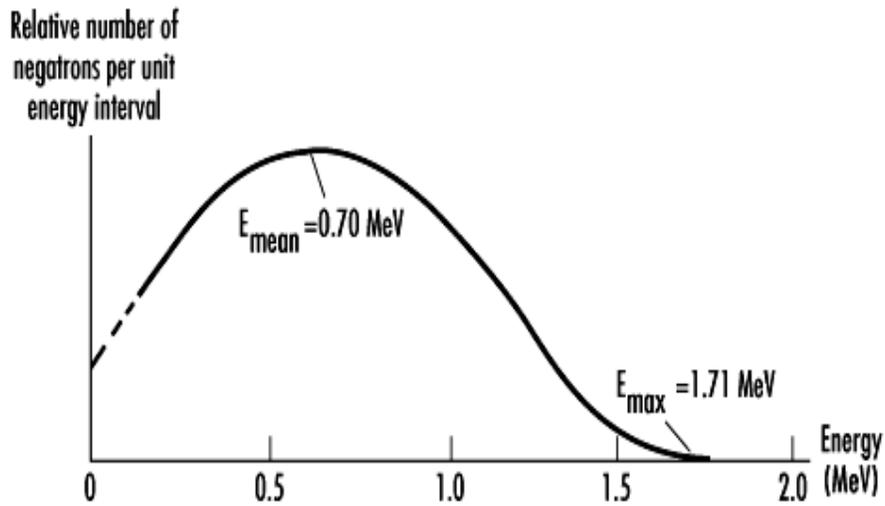


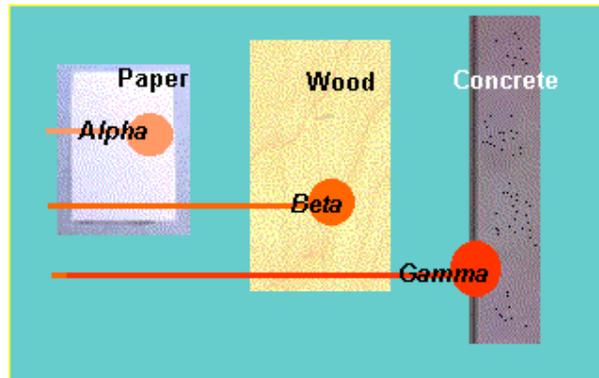
Fig. 2.4 Energy spectra of  $^{60}\text{Co}$  decay from  $^{60}\text{Co}$ .

Nuclei below the line stability are unstable because they have too few neutrons compared to the number of protons in the nuclei. These nuclei tend to gain stability by decay process in which a proton is transformed into a neutron, resulting in a unit decrease in  $Z$  with no change in  $A$ . One possibility for this transformation is positron decay:



Where  $\beta^+$  represent the nuclear origin of the emitted positive electron (positron).

The beta particles could be reflected in magnetic or electric field and much more penetrating than alpha particles, being able to pass through one millimeter of aluminum, whereas the alpha particle are completely stopped by 0.006 cm of aluminum or by a sheet of ordinary writing paper as shown in Fig.2.5. The ionization density produced in a gas by beta particles is much less intense, however, than that produced by alpha particles.



**Fig.2.5** The penetrating power of alpha, beta particles and gamma radiation.

### 2.3.3 Gamma Radiation

Gamma radiation is one of electromagnetic radiations, first detected as emissions from natural radioactive substances such as uranium, radium, and thorium. Gamma radiation has no charge or mass but it is a penetrating radiation. Its properties are similar to x-rays but only differs in origin i.e. while, the sources of gamma rays are nuclear processes, those of x-rays are atomic. There are many different sources of gamma radiation. After the emission of an alpha or beta particle from a parent nucleus, the daughter nucleus formed may have more energy than it would have in its normal state. The nucleus then de-excites by the emission of gamma rays carrying the excitation energy. Gamma radiation was also produced in a nuclear reaction such as the combination of a neutron with a proton to form a deuteron. When a particle like an electron combines with its antiparticle, in this case a positron, they give rise to gamma radiation in processes known as radiation inhalation.

Gamma radiation undergoes many diverse interactions with matter at different energy ranges. Low-energy gamma radiation may be very absorbed by an atomic electron that is then emitted. The ejected electron is known as a photoelectron, and the process is known as the photoelectric effect. Gamma radiation can also interact with an atomic electron, sharing its energy and giving rise to the Compton Effect, in which the original gamma radiation is scattered away with reduced energy and the electron is ejected. This electron is known as a Compton electron. Gamma radiation of sufficiently

high energy can also interact with the electric field of the positively charged nucleus producing an electron and a positron. This phenomenon is known as pair production.

When a beam of gamma radiation passes through matter, its intensity after emergence has diminished, principally as a result of the above three processes. Very high-energy gamma radiation can also cause nuclear disintegration and can eject a nuclear particle such as a neutron or a proton. Various types of mesons can also be produced by gamma radiation of extremely high energy in its interaction with atomic nuclei. Gamma radiation is emitted as photons, or discrete quanta of energy (Solomon, 2005).

## **2.4 Radioactive Isotopes**

The discovery of isotopes was one of the results of the work on the radioactive elements. It was found that generally the products of radioactive decay is itself radioactive, and that each of the decay products behaves chemically in different manner from its immediate parent and its daughter product. In searching for the element that transforms into radium, Boltwood 1907 found that a radioactive element with half-life about  $10^5$  years is the parent element, but onium, as it was named, could not be chemically separated from thorium. The product of the radioactive uranium and thorium series were identified as lead but the two forms of lead were found to have atomic weight of 206 and 208 respectively. Soddy coined the word *isotope* for atomic species with different atomic weight but, which are chemically inseparable.

## **2.5 Radioactive Decay Law**

The nuclei of a given radioactive species have a definite probability of decaying in unit time; this decay probability has a constant value characteristic of the particular nuclide. It remains the same irrespective of the chemical or physical state of the element at all readily accessible temperature and pressures. In a given specimen, the rate of decay at any instant is always directly proportional to the number of radioactive atoms of the nuclide under consideration present at that instant. Thus, if  $N$  is number of the

particular radioactive atoms (or nuclei) present at any time  $t$ , the decay rate is given by (Glasstone and Sesonske., 1986). :

$$\frac{dN}{dt} = -\lambda N \quad (2.1)$$

Where  $\lambda$ , called the *decay constant* of the radioactive nuclide, is a measure of its decay probability in unit time. Upon integration between any arbitrary zero time, when the number of radioactive nuclei of the specified kind present is  $N_0$ , and a time  $t$  later,

$$\ln \frac{N}{N_0} = -\lambda t,$$

$$\text{or } N = N_0 e^{-\lambda t} \quad (2.2)$$

Radioactive decay is seen to be an exponential process, the actual decay rate being determined by the decay constant  $\lambda$  and by the number of the particular nuclei present. The reciprocal of the decay constant, represented by  $\tau$ , is called the mean life (or average life) of the radioactive species and given by:

$$\tau = \frac{1}{\lambda} \quad (2.3)$$

It can be shown that the mean life is equal to the average life expectancy of the nuclei present at any time. The most widely used method for representing the rate of radioactive decay is by means of the *half-life*. It is defined as the time required for the number of radioactive nuclei of a given kind (or for their activity) to decay to half its initial value. Because of the exponential nature of the decay, this time is independent of the amount of the radionuclide present. Thus, if  $N$  is set equal to  $\frac{1}{2} N_0$  in equation (2.2), the corresponding time  $t_{1/2}$ , which is the half-life, is given by:

$$\frac{1}{2} = \frac{N_0 e^{-\lambda t_{1/2}}}{N_0}$$

$$\text{or } \ln \frac{1}{2} = -\lambda t_{1/2} \quad (2.4)$$

The half life is thus inversely proportional to the decay constant or, by equation (2.3), directly proportional to the mean life, i.e.,

$$N = N_0 e^{-\lambda t} = 0.639 N_0 \quad (2.5)$$

The half-lives of known radioactive nuclides range from a small fraction, e.g., about a millionth of a second to billions of years. Since the number of nuclei (or their activity) decays to half its initial value in a half-life period, the number (or activity) will fall to one-fourth by the end of two period, and so on (Fig. 2.6). In general, the fraction of the initial number of nuclei (or activity) remaining after  $n$  half-life period is  $(\frac{1}{2})^n$ . After seven half-life periods, the amount of radioactive material has decreased to less than 1 percent of its initial value.

**Table 2.1** Characteristic of some common radioactive nuclides.

Natural occurring			Artificial		
Species	Activity	Half-life, yr.	Species	Activity	Half-life
Thorium-232	Alpha	$1.40 \times 10^{10}$	Thorium-233	Beta	22.2 min
Uranium-238	Alpha	$4.47 \times 10^9$	Protactinium-233	Beta	27.0 days
Uranium-235	Alpha	$7.04 \times 10^8$	Uranium-233	Alpha	$1.58 \times 10^5$ yr
			Uranium-239	Beta	23.5 min
			Neptunium-239	Beta	2.35 days
			Plutonium-239	Alpha	$2.44 \times 10^4$ yr

The half-lives of number of substances of interest in the nuclear energy field are quoted in Table 2.1. On the left side of the table are given the half-lives of species which exist in nature, whereas on the right side are those of artificial nuclides. Thorium-233 is

formed from thorium-232 by neutron capture; it then decays to protactinium-233, which in turn yields uranium-233. Similarly, uranium-239 results when uranium-238 captures a neutron, the successive decay products are then neptunium-239 and plutonium-239 (Glasstone and Sesonske., 1986)

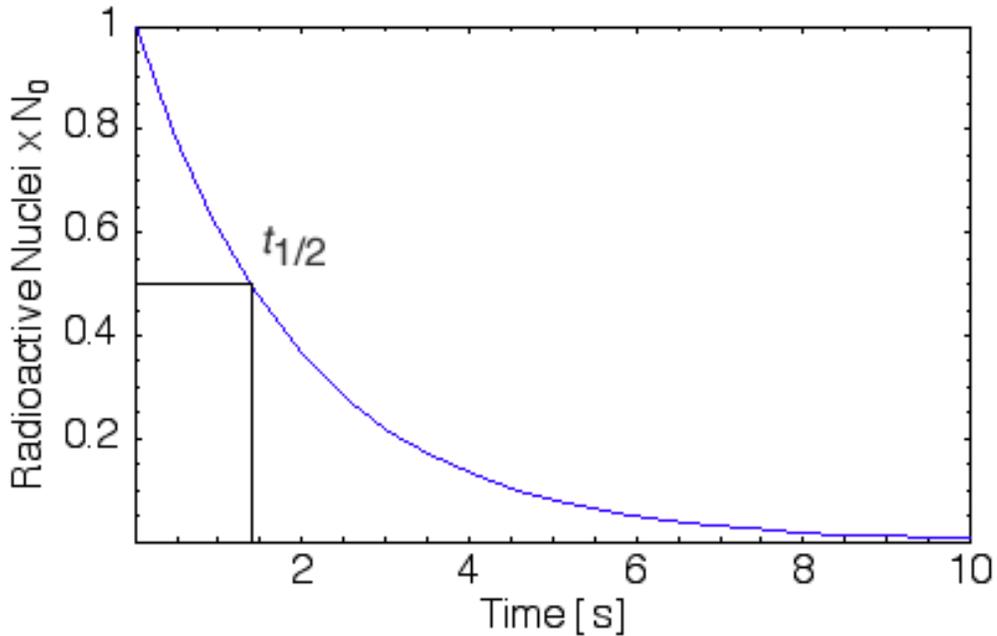


Fig .2.6 Representation of exponential radioactive decay in terms of half-life periods.

## 2.6 Radioactive Equilibrium

In a series of decay stage, such as those, which follow nonfission neutron capture by thorium-232 or uranium-238, or as represented by the members of a natural radioactive series, or as exhibited by many fission products, each radioactive member of the series decays in accordance with equation (2.1) with its own specific value for the decay constant. Such a series may be represented by:

... .. ,

where A may be the parent of a natural radioactive series, or the species formed when a neutron is captured, e.g., thorium-232 or uranium-239, or it may be a radioactive fission product; the stable end-product of series is represented by X. Consider any member of the series such as B, for example. The rate of decay is given by equation (2.1), but it must be remembered that, at the same time, B is being formed by decay of A; hence, at

any specified time, the net rate of change of B with time is given by (Glasstone and Sesonke., 1986).

$$\frac{dB}{dt} = \lambda_A A - \lambda_B B \quad (2.6)$$

The first term on the right representing the rate of formation of B by decay of A, and the second term the rate of decay of B.

Upon rearrangement, this yields

$$\frac{dB}{dt} + \lambda_B B = \lambda_A A,$$

Which is a linear differential equation of the first order. If both sides multiplied by the integrating factor  $e^{\lambda_B t}$ , the left side becomes a complete differential; thus,

$$\frac{d}{dt} (B e^{\lambda_B t}) = \lambda_A A e^{\lambda_B t}$$

So that

$$\left( B e^{\lambda_B t} \right) = \int \lambda_A A e^{\lambda_B t} dt \quad (2.7)$$

by equation (2.2),

$$B e^{\lambda_B t} = \lambda_A A_0 \frac{e^{\lambda_B t} - 1}{\lambda_B - \lambda_A} + C \quad (2.8)$$

Where  $C$  is the integration constant? If  $B_0$  is the initial amount of B, then setting  $t = 0$  yields?

$$B_0 e^{\lambda_B \cdot 0} = \lambda_A A_0 \frac{e^{\lambda_B \cdot 0} - 1}{\lambda_B - \lambda_A} + C, \quad (2.9)$$

Where  $C$  is the integration constant? If  $B_0$  is the initial amount of B, then setting  $t = 0$  in equation (2.9) gives the value of  $C$  as

$$C = B_0 - \lambda_A A_0 \frac{1 - 1}{\lambda_B - \lambda_A},$$

Hence, equation (2.9) leads to the expression

$$B e^{\lambda_B t} = \lambda_A A_0 \frac{e^{\lambda_B t} - 1}{\lambda_B - \lambda_A} + B_0 - \lambda_A A_0 \frac{1 - 1}{\lambda_B - \lambda_A} \quad (2.10)$$

Thus, equations (2.8) and (2.10) give the amounts of the parent A and daughter B present at any time  $t$ , in terms of their initial amounts and the respective decay constant.

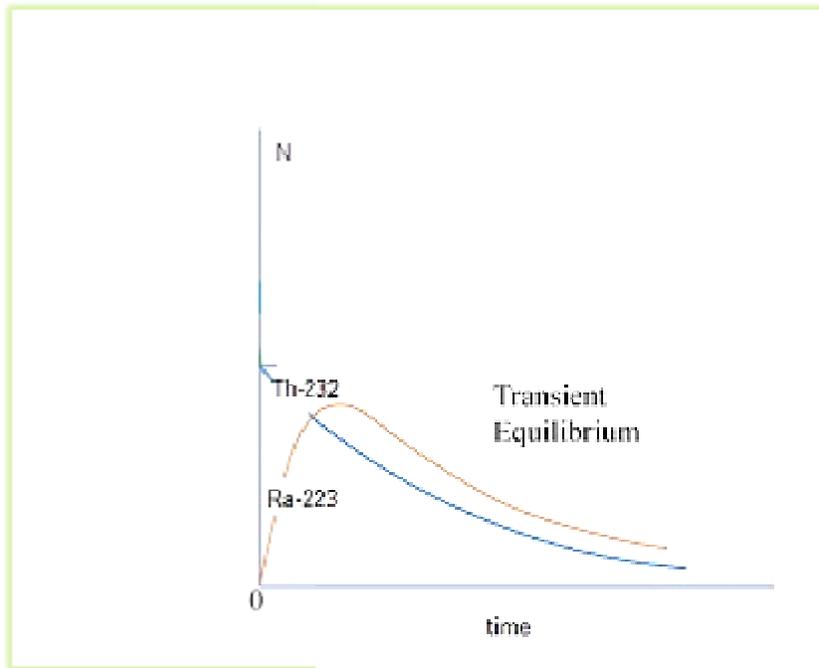
If the parent has an appreciably longer half-life than the daughter does nuclide, i.e.,  $\lambda_A$  is less than  $\lambda_B$ , and  $N_B$  is zero, equation (2.10) reduces to the approximate form

$$N_B \approx \frac{\lambda_A N_A}{\lambda_B} \quad (2.11)$$

Since, after a sufficient time,  $N_B$  can be neglected in comparison with  $N_A$ . Upon introducing equation (2.8), it follows that

$$\frac{N_B}{N_A} \approx \frac{\lambda_A}{\lambda_B} \quad (2.12)$$

i.e., the ratio of the amount of daughter B to that of the parent A becomes constant. This expression describes the condition of *transient equilibrium* in which the absolute amounts of A and B are changing but their ratio remains the same, this case illustrated in F.g.2.7.



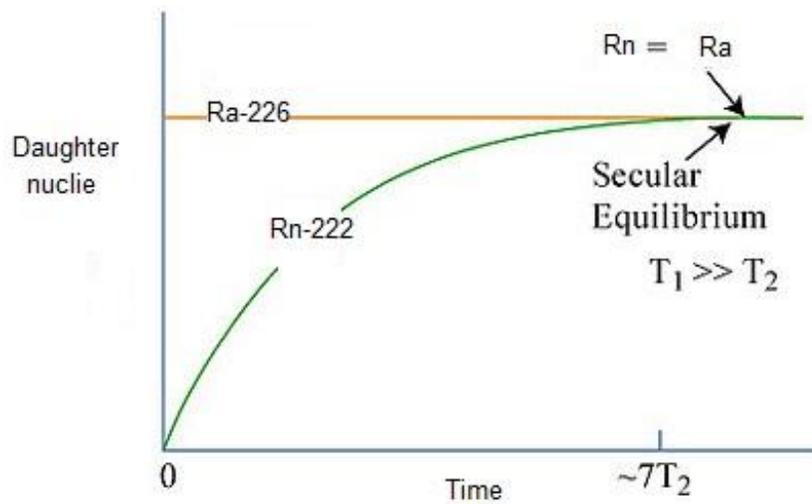
**Fig.2.7** Buildup of  $^{226}\text{Ra}$  from  $^{232}\text{Th}$ .

In the case that the parent has a very long half-life, so that  $\lambda_A$  is very small in comparison with  $\lambda_B$ , the  $N_B$  terms in equation (2.10) may be disregarded and,

further,  $\lambda_B - \lambda_A \approx \lambda_B$ . Consequently, it can be shown from equation (2.10) that, after a period of time,

$$N_B \approx N_A \quad \text{or} \quad \lambda_B N_B \approx \lambda_A N_A$$

which represents the state of *secular equilibrium*. The accumulation of radon from radium is typical for this case and the growth of  $^{222}\text{Rn}$  from  $^{226}\text{Ra}$  is illustrated in Fig .2.8. Not only is the ratio of B to A fixed, but the absolute amount of B also remains constant. This follows since A is decaying to form B at the same rate as B is decaying into C, so that the net amount of B is unchanged. If C is radioactive, it will also attain a condition of secular (or radioactive) equilibrium in the course of time and its amount will become constant. Thus, all the members of a radioactive series, except the last, will ultimately be in radioactive equilibrium with one another, provided that parent of the series has a very long half-life (Glasstone and Sesonske ., 1986).



**Fig. 2.8** Build-up of Ra-222 from Ra-226.

## 2.7 Radioactivity Units

In the SI system, a sample of radionuclide decaying at the rate of 1 disintegration per sec (1 dis/s) is defined as having an activity of 1 *Becquerel* (1Bq). Traditionally, activities have been expressed in terms of *curie*, which was based on the estimated rate of decay of 1 gram ( $10^{-3}$ kg) of radium-226; one curie (1Ci) is defined as the activity of radionuclide decaying at the rate of  $3.7 \times 10^{10}$  dis/s. Hence,

$$\text{Activity in Ci} = \frac{\text{Activity in Bq}}{3.7 \times 10^{10}}$$

According to equation (2.1) above, a sample containing  $N$  atoms (or nuclei) of a given radionuclide decays at the rate of  $\lambda N$ ; hence, if  $\lambda$  is expressed in  $\text{s}^{-1}$  units, the activity is  $\lambda N$  dis/s or  $\lambda N / 3.7 \times 10^{10}$  Bq. Since  $\lambda$  is then equal to  $0.693 / t_{1/2}$  is the half-life in seconds, it follows that

$$\text{Activity} = \frac{\lambda N}{3.7 \times 10^{10}} \text{ Bq} \quad (2.13)$$

$$= \frac{\lambda N}{3.7 \times 10^{10}} \text{ Ci} \quad (2.14)$$

The number of atoms  $N$  in a sample of a certain mass is inversely related to the isotopic mass (or roughly the mass number) of the species. Hence, the activity in Becquerels or curies, of a certain mass of a radionuclide is inversely related to both its mass number and its half-life (Glasstone and Sesonske., 1986).

## 2.8 Natural Radioactive Series

### 2.8.1 Radioactive Nuclides

The naturally occurring forms of the great majority of elements are stable, but a few of high atomic mass, from polonium (atomic number 84) onward, e.g., radium (88), thorium (90), and uranium (92), consist entirely of unstable, radioactive nuclides. In addition, the elements thallium (81), lead (82), and bismuth (83) exist in nature largely as stable isotopes, but also to some extent as radioactive species. The unstable

substances undergo spontaneous change, i.e., radioactive disintegration or radioactive decay, at definite rates. This decay is associated with the emission from the atomic nucleus of an either electrically charged particles, an *alpha particle*, i.e., a helium nucleus, *or beta particles*, i.e., an electron. In many instance, gamma radiation accompanies the particles emission. Frequently, the products of decay are themselves radioactive, expelling either an *alpha or beta particle*. After a number of stages of disintegrations, an species with a stable nucleus is formed (Glasstone and Sesonske., 1986).

Because of physical and chemical researches on the naturally occurring radioactive elements, it was proved that each radioactive nuclide is a member of one of three long chains, or radioactive series, stretching through the last part of the periodic system. These series are named the uranium, actinium, and thorium series, respectively, after elements at, or near, the head of the series. In the uranium series, the mass number of each member can be expressed in the form  $(4n + 2)$ , where  $n$  is an integer, and the uranium series is sometimes called the  $(4n + 2)$  series. In the actinium and thorium series, the mass number are given by the expressions  $(4n + 3)$  and  $4n$ , respectively. There is no natural radioactive series of nuclides whose mass number are represented by  $(4n + 1)$  (Cothorn and Smith, Jr., 1987).

**Table 2.2** Properties of natural radioactive series.

<b>Radioactive Species</b>	<b>Nuclide</b>	<b>Half-Life (yr)</b>	<b>Final Stable Element of the Series</b>	<b>Radionuclide</b>
Uranium	$^{238}\text{U}$	$4.47 \times 10^9$	$^{206}\text{Pb}$	$^{222}\text{Rn}$
Actinium	$^{235}\text{U}$	$8.04 \times 10^8$	$^{207}\text{Pb}$	$^{219}\text{Rn}$
Thorium	$^{232}\text{Th}$	$1.41 \times 10^{10}$	$^{207}\text{Pb}$	$^{220}\text{Rn}$

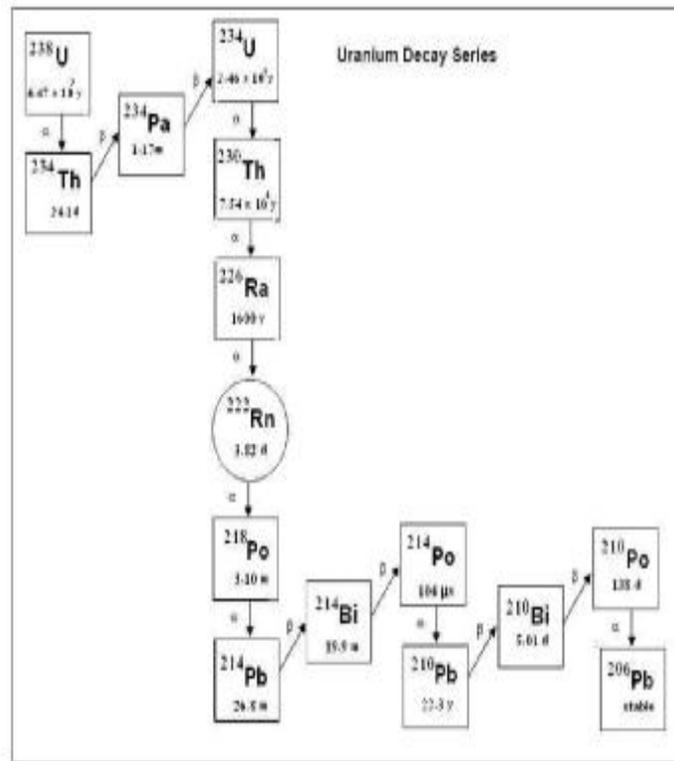
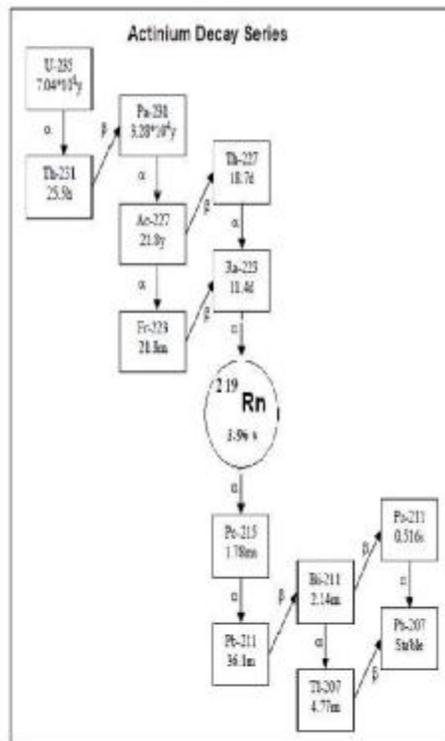
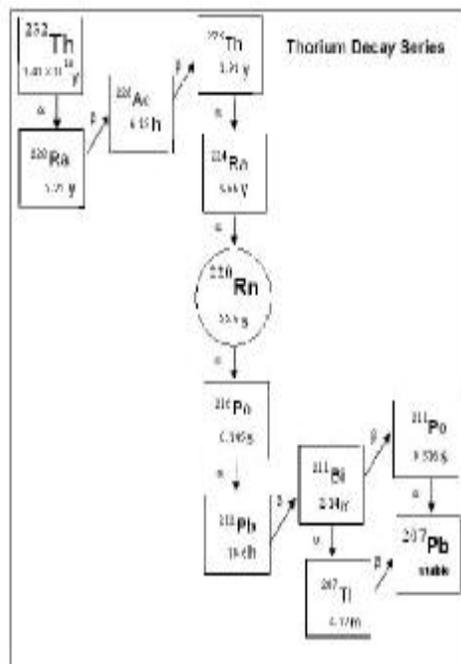


Fig.2.9 <sup>238</sup>U decay series



**Fig.2.10.**  $^{235}\text{U}$  decay series.



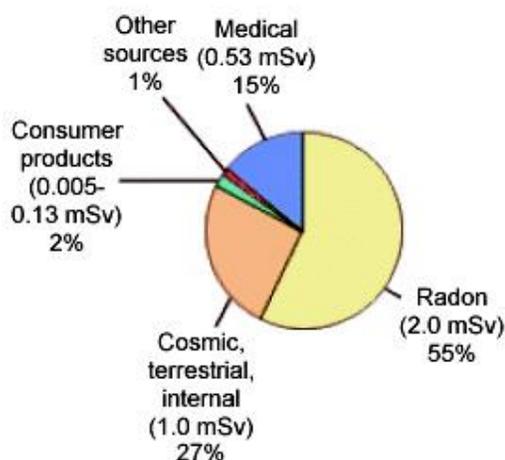
**Fig.2.11**  $^{232}\text{Th}$  decay series.

## Chapter 3

### Radon: discovery, properties, generation, and transport mechanism

#### 3.1 Introduction

Radon was identified at the beginning of the 20<sup>th</sup> century; it was not until late in 1960 that a correlation between radon and its progenies exposure and lung cancer among uranium miners was established (Snihs, 1992; Webb, 1992). In our environment, there are four most important decay series, specifically uranium ( $^{238}\text{U}$ ), thorium ( $^{232}\text{Th}$ ), actinium ( $^{235}\text{U}$ ) as well as single isotopes such as potassium ( $^{40}\text{K}$ ) and the series of neptunium ( $^{237}\text{Np}$ ). These elements occur in nature in trace amounts and in different concentrations diffusive across the earth crust. These concentrations depend on the mineral composition of rocks, (Emsley, 1989). All the products of these series are metals but the radon is a gas daughter due to the alpha decay of radium. We have to think about radon as isolated atoms, which diffuse within fluid that may itself be moving. Radon is a noble gas; it is mobile and can move over significant distance within the earth and in the atmosphere. Inhalation of radon can be considered as a problem to health. In 1899, the British scientist, Owens. R. B. and the New Zealand scientist, Ernest Rutherford were discovered  $^{222}\text{Rn}$ , which was know at this time as thoron. In 1900, the German chemist Friedrich E. Dorn observed  $^{222}\text{Rn}$  and called it as radium emanation. Radon exposure (actually radon progeny) has been link to lung cancer in numerous case-control studies performed in the United States, Europe and China. However, the health hazard because of exposure to radon and its progenies is sometimes so significant. For example there are approximately 21,000 deaths per year in the USA due to radon-induced lung cancers (EPA., 2003). Fig.3.1 shows the radiation dose from radon exposure compared to other radiation source exposures (NCRP, 1987).



**Fig.3.1** Radiation dose from different radiation sources.

### 3.2 Radon Characteristics

There are 27 known isotopes of radon ranging from  $^{200}\text{Rn}$  to  $^{226}\text{Rn}$  (Cothem, 1987).  $^{222}\text{Rn}$  (true radon),  $^{220}\text{Rn}$  (thoron) and  $^{219}\text{Rn}$  (actinon) are the most three common isotopes of radon. The atomic number of radon is 82 and like any other noble gas, radon is a toxic, colorless, odorless radioactive gas, is invisible and intangible. Its melting point is  $71\text{ }^{\circ}\text{C}$  and boiling point is  $61.8\text{ }^{\circ}\text{C}$ . Radon has highest density of  $\sim 9.96\text{ kg}\cdot\text{m}^{-3}$  and it is about ten times heavier than air. Each series of natural radioactive nuclides produce an isotope of radon,  $^{238}\text{U}$  decays to give  $^{226}\text{Ra}$  which is the direct parent nuclide of  $^{222}\text{Rn}$ ,  $^{235}\text{U}$  is the grand parent of  $^{219}\text{Rn}$  and finally  $^{220}\text{Rn}$  which it comes from the decay series of  $^{232}\text{Th}$  (NCRP-97,1998).  $^{222}\text{Rn}$  has a half-life of 3.82 days, therefore it is considered as major concern of the radiation pollution and human health hazard (Kunz et al., 1981; UNSCEAR, 1993) while  $\text{Rn}^{220}$  has a half-life of 55.6 seconds and  $^{219}\text{Rn}$  has a half-life of 3.96 seconds ( Sumner., Wheldon., and Watson., 1991).  $^{219}\text{Rn}$  and  $^{220}\text{Rn}$  do not exist long enough to migrate a significant distance to become health hazard (USEPA, 1987).

Radon gas can diffuse easily into indoor environment from construction materials that originate from soils and rocks, which contain traces of uranium and

thorium. Radon enters the body through breathing; consequently, it can cause the risk of lung cancer to the inhabitant of buildings with poor ventilation and old houses (Hassan, 1996). Hence, it is important to evaluate the long exposure and concentration levels of radon and its progenies in buildings. A large number of investigations have been conducted in different countries all over the world in order to monitor radon level before action can be taken. The recommended indoor action level for the population ranges between 200 and 600 Bq/m<sup>3</sup> (ICRP, 1994). An indoor concentration of 200 Bq/m<sup>3</sup> has been assigned as a reference for radon level in most European countries (Popvic et al., 1996). A radon level of 150 Bq/m<sup>3</sup> and an average effective dose of 7.5 mSv/y have been also adopted in the USA as a reference level for <sup>222</sup>Rn concentration and as an acceptable dose before taking any action (Hassan; 1996).

### 3.3 Sources of Radon

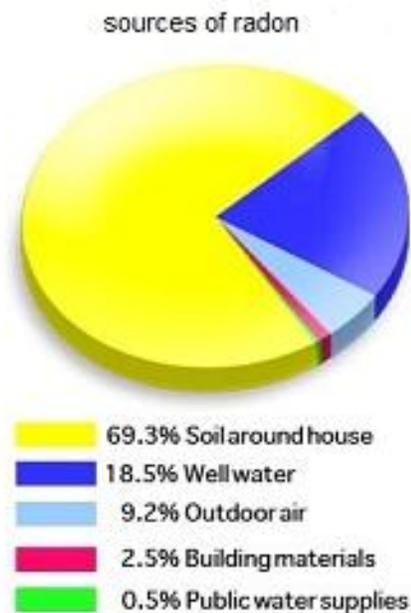


Fig.3.2. Radon sources.

As shown in Fig. 3.2 above, radon is in everywhere around the world in materials containing radium, soil and rocks,..etc, and there are many forms of transporting radon such as, water, ambient air,..etc. The most widely radon sources are described in following subsections.

### 3.3.1 Soil

Soil and rocks contain amount concentration of uranium and thorium. These concentrations depend upon the location and geologic features of soil.  $^{222}\text{Rn}$  and  $^{220}\text{Rn}$  are produced in the soil and rocks from the decaying process of uranium and thorium, respectively. They were continuously transport to the surface layer of the earth and then mix with the air. The rate of exhalation of radon from the surface area depends on the amount of uranium and thorium present in the soil (Fleischer, and Morigo-Compero, 1978). On the average, 80% of radon enters atmosphere from the top layers of ground (Knuth; 1981). Transport by molecular diffusion of radon in soils is limited because of its short half-life. Migration by diffusion ranges from about 5 m in gravel to about 2 cm in mud or clay, and distances greater than 1 m are probably unusual (Cothorn and Smith, 1987).

Radon enters to the soil pores due to recoil of radon atoms and decay of  $^{226}\text{Ra}$ . The fraction of radon enters pores called "emanating power" and *it is defined as the fraction of the radon generated in the materials that can enter into the pore volume of the material*. Reported values ranges from about 1– 80 % depending on the soil type, pore space, water content (Fleischer, and Morigo-Compero, 1978). The emanation rate varies from place to another due to different radium concentration and soil permeability. The areal exhalation rates of  $^{222}\text{Rn}$  from soil varies from 0.2 –7.0 ( $\text{mBq}\cdot\text{m}^{-2}\cdot\text{s}^{-1}$ ) or 720 – 25,200 ( $\text{mBq}\cdot\text{m}^{-2}\cdot\text{h}^{-1}$ ).

For people whose dwells in floors close to the ground, the most important radon source is radium in the ground. The radium concentration in soil usually lies

in the range 10 – 50 Bq/kg, but it can reach values of hundred Bq/kg, with an estimated average of 40 Bq/kg (UNSCEAR.,1993). In dry soil with radium activity approximately 50 Bq/kg, the maximum radon activity concentration in the pore air or also called radon gas concentration in soil (for porosity = 0.3) is thus about 80,000 Bq.m<sup>-3</sup> with no ventilation. In soil consisting largely of granite rich in uranium and with a radium concentration of 250 Bq/kg, the maximum radon concentration is 400,000 Bq.m<sup>-3</sup> and in sand consisting of fragments of alum shale with a radium concentration of 1,250 Bq/kg, it is about 2x10<sup>6</sup> Bq.m<sup>-3</sup>. Concentrations in air-filled pores will not be as high as in horizons close to the surface as each form of ventilation of the soil, and diffusion from it, reduces the radon levels in the pores. Thus, the normal radon concentration in the soil air i.e., under Swedish conditions is 10,000-50,000 Bqm<sup>-3</sup>; but levels are significantly higher in soils enriched with radium concentrations: in granite rich in uranium, it is up to 250,000 Bq.m<sup>-3</sup>, and in alum shales up to more than 1 million Bq.m<sup>-3</sup>. If emanation is great, .e.g., as from gravel and clay, radon concentrations can be relatively high: 50,000-100,000 Bq.m<sup>-3</sup>, without radium concentration being above normal for the soil air. Table. 3.1 gives the normal concentration of <sup>226</sup>Ra in the soil and <sup>222</sup>Rn in the soil air, in various soils (Durrani and Ilić, 1996).

**Table. 3.1.** Normal concentrations of <sup>226</sup>Ra and <sup>222</sup>Rn in soils, measured at a depth of 1 m (Durrani and Ilić., 1996).

Soil type	<sup>226</sup> Ra (Bq.Kg <sup>-1</sup> )	<sup>222</sup> Rn (Bq.m <sup>-3</sup> )
<b>Till or residual soils with normal radium content</b>	15-65	5,000-30,000
<b>Till or residual soils with fragments of granite</b>	130-125	10,000-60,000
<b>Till with fragments of uranium-rich granite</b>	125-360	10,000-200,000
<b>Gravel</b>	30-75	10,000-150,000

### 3.3.2 Building Materials

Building materials are used as construction and decoration for buildings, which are considered as one of the important sources of radon and its progenies. These building materials which are derived from soil and rocks contain trace amounts of radioactive nuclides that occur in the earth's crust, such as  $^{238}\text{U}$ , the grand parent of  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$ . The building materials become a source of radon especially if they contain radium (more than  $37 \text{ Bq.kg}^{-1}$ ). Typical uranium-bearing natural materials are certain granites, but also can find uranium-rich bedrocks of different types used as building materials. Soils, clays, sands, marbles, etc., used as building materials may have enhanced uranium content (Durrani and Ilić, 1996).

Moreover, the production rate of radon in dwellings depends on the concentration of radium content in the subsoil and building materials and other soil parameters such as porosity, density of the wall material and permeability (Cartwright et al., 1987; Camplin et al., 1988). The emission of radon from building materials was found to depend on the ventilation of buildings as well as of the radium content in building materials. The nongaseous  $^{222}\text{Rn}$  decay products are partially suspended in air as a mixture of attached and unattached fractions and partially deposited on walls and furniture (Brenner, 1989). There are many types of bedrocks containing uranium which is a significant source of radon when uranium concentration should be over 50 ppm ( $615 \text{ Bq.m}^{-3}$ ), especially if the bedrock is deep down (deeper than 100 m). The radon gas moves through along the layers and cracks of the bedrock (Durrani and Ilić, 1996). The radon emanation rate from building depends on the pressure and temperature and even the moisture content. Radon emanation increases with moisture content in building materials.

The USA National Bureau of Standards (Gesell and Prichard, 1983) conducted a comprehensive analysis and assessment of radon transport and exhalation from building material. This work includes a detailed analysis of transport mechanism, and how the microstructural properties and internal

characteristics of building materials may affect the transport and exhalation of radon. The radon emanation from most building materials would not generate radon levels above 0.0015 WL ( $4.2 \times 10^{-8} \text{ Jm}^{-3}$ ) for an average home. As an example, table. 3.2 shows the radium and thorium content for some German building materials.

**Table.3.2**  $^{226}\text{Ra}$  and  $^{232}\text{Th}$  content for some German building materials (Winter and Wicke, 1993).

<b>Material</b>	<b><math>^{226}\text{Ra}</math> ( Bq.kg<sup>-1</sup>)</b>	<b><math>^{232}\text{Th}</math> ( Bq.kg<sup>-1</sup>)</b>
Granite	30-50	17-311
Brick	10-200	12-200
Gypsum	2-70	2-100
Limestone	4-41	2-20
Concrete	7-92	4-71
Light –weight concrete	6-80	1-60

### 3.3.3 Ground Water

Although, the occurrence of radon in groundwater was first reported in 1902 by Thompson. J. at Cambridge University in England, it was "rediscovered" in Maine in the 1950s. Since that time, there have been numerous studies of radon in groundwater in specific areas. The concentration of radon in groundwater is directly related to the composition and rock type of the aquifer. Radon concentration will be higher in the groundwater coming from metamorphic rocks such as granite than from all other aquifer types. In every study where the average radon content in ground water is reported by rock type, granites always have the highest levels. Average levels in water from granite are usually  $100,000 \text{ Bq.m}^{-3}$  or

greater. Concentrations in individual domestic wells frequently be as high  $3.7 \times 10^6$  Bq.m<sup>-3</sup> (Cothorn and Smith, Jr, 1987). Water in the ground can dissolve the radon at all temperature, the solubility of radon varies from 51 ml of radon per 100 ml of water at 0 °C to 13 ml of radon per 100 ml of water at 50 °C (Chemical Rubber Publishing Company; 1977). Since the underground radon pressure is high, the water passing through rock pores dissolved radon in sufficient amounts.

The occurrence of radon in water is feature of the concentration of radium in host rock. The physical factors such as grain size distribution, porosity, etc., of rocks play a basic role in radon transport into water. The radon movement in water is governed by water transport rather than the radium content. Radon released from water when the pressure is reduced, temperature is increased, and water is aerated.

### **3.3.4 Natural Gas**

It is another source of radon entry to buildings and dwellings, whereas the emanated radon in the porous geological formations will mix with the natural gas. In particular, using the natural gas immediately after underground storage, radon will move with natural gas to the places of use of natural gas such as kitchens and heating rooms (Tufail; 1992). However, this source is considered usually as a very small contributor in comparison with other sources.

The concentration of radon at the places of natural gas usage depends on the production rates of natural gas, natural gas processing, pipeline dilution, pipeline transmission time and storage time (Handbook of radon in buildings, 1988). Radon also spread in the liquid petroleum gas (LPG) and the concentration of radon in the (LPG) is measured to be in the range from 70.3 Bq.m<sup>-3</sup> to  $4.8 \times 10^4$  Bq.m<sup>-3</sup> (Johnson et al, 1973). The concentration of radon from burning natural gas depends on the amount of the used gas.

The united nation scientific committee on the effects of atomic radiation (UNSCEAR) has made a very simple model to try to estimate the relative

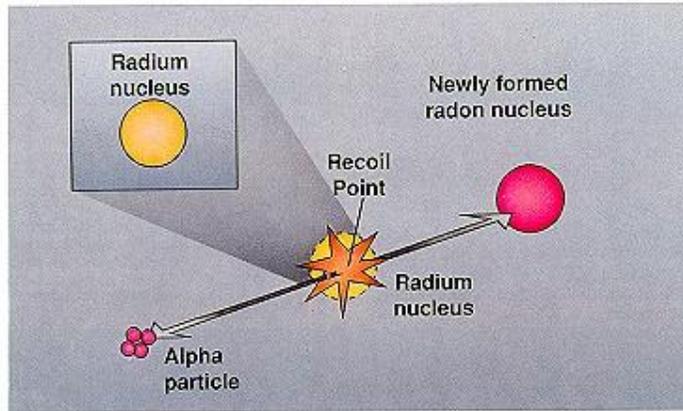
contribution of some radon sources such as soil, building materials and outdoor air at the ground floor. The results were respectively, ~ 60%, ~ 20%, and 20%, while for the upper floors in high-rise buildings, where the radon concentration is estimated to be "typically" 20 Bq/m<sup>3</sup>, these values become ~ 0%, ~ 50% and 50% respectively, (UNSCEAR, 1993).

### 3.4 Radon Generation

<sup>222</sup>Rn, is the most abundant in mineral grains, is 25 times lower than that in soil gas in the ground, a further 1000 times less in homes, still lower in the air over the ground by 5 times, and another 100 times lower over surface of ocean. <sup>226</sup>Ra is a product decay from <sup>238</sup>U chain reaction. Radium always appears as trace element in soil, with other elements such as magnesium, calcium and barium. Radium emanation in the soil earth's surface is considered as the major source of the radon in atmosphere. The global emanation from soil is estimated at about 2 billion curies per year. The ground water potential is about 500 million curie per year (Harley., 1974 and NCRP Report No.97., 1988).

Radon is fixed in the soil grains and the soil can be treated as porous medium where these pores are filled with water or gas. Radon atoms created in the grains, because of radium decay as shown in Fig.3.3, have kinetic energy of 86 keV. This energy may help the atoms to reach the pore volume in the soil. If the pore volume is filled with gas, the atoms may transport to the next grain, but if there is water then the atoms will trapped in the soil fluid.

Most building materials also generate radon, caused by the uranium and/or radium containing natural minerals in their components. The radon generation depends on the age of the building material (Roelofs and Scholten., 1994), the relative humidity (Stranden et al., 1984) and of course on the sort and the amount of building material. However, also between similar building material samples, the amount of generated radon can differ by an order of magnitude (Cozmuta and Graaf., 1999). This makes comparison of radon concentrations between "identical" dwellings more difficult.



**Fig. 3. 3**The recoil radon from radium.

### 3.5 Radon Emanation

One of main parameters influencing the radon concentration is the radon emanation coefficient of the material. By definition, the emanation coefficient is dimensionless and has values between 0 and 1. Radon emanated in grains when the parent nuclide  $^{226}\text{Ra}$  and its isotopes decay with alpha emission. The fraction of produced radon can escape through the pore spaces of the sample. Using of the recoil energy of its atoms during decaying process, the recoil energy ranges from 86 to 100 KeV. Afterward it can migrate and then exhale to the air of atmosphere through cracks and fissures by transport mechanism such as diffusion, fluid convection and advection (The movement of radon due to pressure gradient between the air in pore space and ground surface). This escape depends on the grain size and the site of  $^{226}\text{Ra}$  in the grain. The rest of the radon is trapped in the grains. *The ratio of the escaped radon to the radon created in the grain is defined as emanation coefficient, denoted by ( $e$ ), and is known to be in the order of 0.2-0.3 (Durrani and Ilic, 1996).* In soil and building materials, the diffusion and advection are responsible for radon transport. The transfer of  $^{222}\text{Rn}$  atoms from one place to other due to the concentration gradient is called diffusion (Nazaroff and Nero, 1988). The diffusion distance for wet soil is some centimeters while in

dry soil can be up to 1.5m (Fleischer et al, 1980).  $^{222}\text{Ra}$  Based on advection transport the range of a transport in soil can reach up to 100 m or more (Fleischer, 1998). The cracks and fissures can be considered as pores that increase the grain surface area leading to increase in radon emanation. Many research groups have made many interesting studies on the physical factors that affect the emanation of radon such as soil moisture and porosity, pore size, amount of radium and its distribution in the grain, grain shape and micro climatological parameters (humidity, pressure and temperature of soil). The moisture of the soil is affect the emanation of radon because of the strong ability of water in the pore spaces to prevent the recoiling atoms from entering the neighboring grain (Garver and Baskaran 2004). These studies show that the emanation coefficient is enhanced with increasing of moisture. Maximum radon emanation occurs when soil is between 15% and 30% moisture (Saad et al., 2013). The increase or decrease of moisture value can reduce the emanation coefficient close to zero. Experimental results showed that radon emanation is proportional to porosity (Sun and Furbish 1995; Misdaq et al. 1998; Maraziotis 1996).

Porosity is defined as the volume of pore space to the volume of the solid material. This porosity factor significantly influences the emanation process of radon. It should be noted that any increase in the pore space followed by increase in the porosity makes a great chance for radon to migrate from the host material. The process of emanation for the  $^{222}\text{Rn}$  is somewhat similar to that for the isotopes of radon ( $^{220}\text{Rn}$  and  $^{219}\text{Rn}$ ) but with different recoil ranges and diffusion lengths. The recoil ranges of  $^{222}\text{Rn}$  are given as 30-50 nm for solids, 95 nm for water and 64000 nm in air (Tanner 1980; Greeman and Rose 1995). These short ranges refer only to radium atoms that lie so close to the surface, which can be the main source of the emanated radon. The diffusion coefficient of radon through the solid material is of the order of  $10^{-20} \text{ cm}^2 \cdot \text{s}^{-1}$  (Lawrence., 2005) . Overall, the emanation process of radon from natural materials such as soil, rocks and construction materials as shown in Fig.3.4 are more or less the same. The radium distribution is considered as an important factor that affects the radon emanation. In addition,

assume that, the radium distribution is homogenous inside grains and close to the surface, which distribution led to the high radon emanation. Furthermore it was found that the high emanation radon was observed from soil samples more than the solid ones (Sakoda and Nishiyama, 2010).

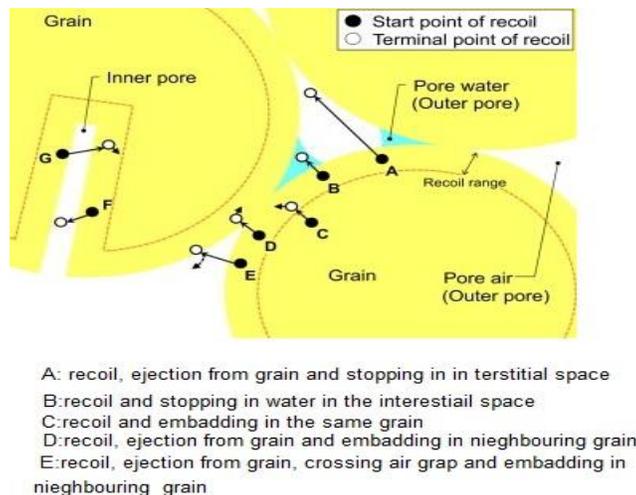


Fig. 3.4 The radon emanation process for different type of alpha recoil.

### 3.6 Radon Migration

Radon migration explains the track of the radon atoms through rock, soil, water and even the air, but usually this expression is used to describe transport within the soil. There are two important transport mechanisms of radon movement from its origin to the atmosphere: diffusion and convection. Most radon produced from radium never escapes from the mineral in which it is born, it is usually stuck in the crystal lattice until it decays. There are two possible ways for escape of radon. The first possibility is the direct ejection of radon by recoil from alpha emission (Kigoshi 1971). The emission of alpha particles with 4.78 MeV by  $^{226}\text{Ra}$  gives the residual  $^{222}\text{Rn}$  recoil energy of 86 KeV. If the radium within the 26 nm from the surface of the mineral the recoiled radon will enter the interstitial space and if the pore space contains water the radon atoms then will be free to diffuse

from the water or transported by it. The second possibility, if the interstitial space is dry or filled of gas, in this case the recoiled radon will move to neighboring grain.

Radon can migrate within soil by diffusion and/or with air gases like CO<sub>2</sub> or flowing water in the soil. Radon diffusion is result of radon atoms migration from high to low concentration as described by Fick's law. Radon diffusion rates in air and water are vastly different. As in Table 3.3 the diffusion constant, D, in water is lower by factor 10,000. From that fair, thus, the average <sup>222</sup>Rn atom will moves about 1.6 m in dry soil, while in saturated water will move about 1.6 cm.

**Table3.3**Mean diffusion distances of radon isotopes in different media (Tanner 1964)

Medium	Mean distance		Diffusion constant (cm <sup>2</sup> .s <sup>-1</sup> )
	<sup>222</sup> Rn (cm)	<sup>220</sup> Rn (cm)	
Air	220	2.85	10 <sup>-1</sup>
Porous soil	155	2.0	5x10 <sup>-2</sup>
Water	2.2	0.0285	10 <sup>-5</sup>

### 3.7 Radon Exhalation Rates

The transport flux of radon from previous sources to the indoor environment is called radon exhalation. The exhalation rates of <sup>222</sup>Rn from many sources, such as soils, sands, rocks, construction materials, uranium tailings and uranium ores have been the researches' aim for many studies (Fleischer et al.1980; Kvasnicka 1990; Todd 1998; Bollhøfer et al.2003; Saad, 2008; Saad et al., 2010; Saad et al., 2013). This exhalation can be measured by exhalation rate that defined as the number of escaped atoms from material per unit surface area per unit time

and expressed in ( $\text{Bq}\cdot\text{m}^{-2}\cdot\text{h}^{-1}$ ) (Saad et al., 2013). It depends on the radioactive content of material, emanation factor, and diffusion coefficient of the radon in the material, porosity and density of the material. The parameters that also affect the exhalation rates are atmospheric pressure, rainfall, humidity, and temperature. The exhaled radon from soil depth into atmosphere depends on the soil type (Saad et al., 2002) its moisture content, isotopes half-life and underlying geology. This depth is about 1-2 m in the low soil moisture content, deeper for sands and shorter for high moisture content and compacted soil (Tanner 1964; Holdsworth and Akber 2004). The small amount of moisture can increase the exhalation of radon, while the rising of moisture can reduce that exhalation. The growth of water content can trap further radon atoms and reduce the exhalation accordingly. A global average value for  $^{222}\text{Rn}$  exhalation rates from natural soils is given (UNSCEAR 2000) as  $16 (\text{mBq}\cdot\text{m}^{-2}\cdot\text{s}^{-1})$  or  $57,600 (\text{mBq}\cdot\text{m}^{-2}\cdot\text{h}^{-1})$  taken from the work performed by (Wilkening et al. 1974). For building materials, the two main parameters that the exhalation process depend on, is the porosity and water content. It has been proved that the presence of water does not involve the same variations in the concentration on the surface of the medium, according to its porosity. For porous media with low porosity like concrete or granite, ( $e < 0.2$ ), the radon concentration and radon exhalation rate sharply increase with water until the volumetric water content becomes higher than 30%. At this point, radon emanation plays an important role, in relation to the molecular diffusion process (Fournier et al., 2005). Various studies showed that the increasing of atmospheric pressure is decrease the exhalation rates (Wilkening et al., 1974; Clement and Wilkening, 1974; Janssens et al. 1988). As a result, the increasing of pressure is press on the air into the soil and interrupted the exhalation (Kraner et al., 1964). Decrease of the pressure will drag the air away from the soil surface and increase the exhalation. The dependence of radon exhalation rates on the ambient temperature has been reported with low correlation coefficients (Jha et al. 2000; Strandén et al. 1984; Schery and Petschek 1983a). The wind speed also affect the radon exhalation (Kraner et al; 1964) reported that increased wind speed would reduce the upper layer of soil and increase the radon exhalation.

Calculation of the contribution of radon from building materials requires knowledge of the radon flux density or exhalation rates from the various components of the structure, which did not quantitatively known. The exhalation process is very complex and dependent on many factors, such as radium content, microscopic configuration of the material, moisture content, pressure changes, temperature, and relative humidity (Gesell and Prichard; 1983). These casual internal and external variables did only qualitatively understood.

The contribution of building materials of indoor radon can be estimated by summing the products of the radon flux density for each type of material, multiply by its surface area in the structure, as the following equation:

$$Rn_{bm} = \sum \frac{\dot{F}_i A_i}{V} \quad (3.1)$$

Where,  $Rn_{bm}$  = Indoor radon concentration from building materials in  $Bq.m^{-3}$

$F$  = Flux density for material  $i$ , in  $Bq.m^{-2}.h^{-1}$ ,

$A$  = Area of material  $i$ , in  $m^2$ ,

$\lambda$  = Ventilation rate,  $s^{-1}$ ,

$V$  = Volume of structure, in  $m^3$ ,

### 3.8 The Decay Products of Radon-222

The estimated contribution of radon and its products is about 68.8% of the total dose received from the natural radiation sources (Khan et al., 1990). Radon products or radon progenies are radioactive isotopes of polonium, bismuth, lead, and thallium. These daughters of the radioactive gases are isotopes of heavy metals and are easily fixed to existing aerosol particles in the atoms. They decay by alpha particles and beta /gamma emission. Radon decay products are divided into two groups, the "**short-lived**" radon daughters  $^{218}Po$  (3.05 min),  $^{214}Pb$  (36.8

min),  $^{214}\text{Bi}$  (19.7 min),  $^{214}\text{Po}$  with half – lives below 30 min, and the **"long-lived"** radondecay products  $^{218}\text{Po}$  (22.3 years),  $^{210}\text{Bi}$  (5.01 days),  $^{210}\text{Po}$  (138.4 days). Radon has no immediate health risk, its short-lived metal products have the main effect on the human risk.

The radon gas is have a long lifetime enough to be inhaled after its exhalation and before it decaying. Radon is chemically inert gas; therefore, a small fraction of radon will inter the blood by absorption. In this case, a small amount of  $^{222}\text{Ra}$  will decay while it is in the lung tissue. The concentrations of radon daughters are represented by the potential alpha energy concentration (PAEC). Most of the direct product of radon is electrically charged (~88%), and a large proportion (~99%) become attached aerosol particles (Pagelkopf and Porstendorfer 2003).

The annual dose equivalent in lung and in tranche bronchial cells amounts to approximately 100  $\mu\text{Sv}$ , the contribution of outdoor exposure being 70  $\mu\text{Sv}$  and that of indoor exposure 1000  $\mu\text{Sv}$ .

### **3.9 Lung Cancer Risk**

It was reported that the radon and its direct products are the main source of radiation hazard in the environment. Hazard can be defined as activity that involves events with undesirable consequences of partially or completely unknown degree and uncertain occurrence in the future. The major risk associated with radon is the lung cancer. The lung cancer risk is defined as the occurrence of excess deaths per million person per year (MPY) due to lung cancer because of exposure to radon and its progeny. The risk coefficient, defined as the number of lung cancer cases per MPY per working level month (WLM), is determined from the epidemiological data of the occupationally exposed mine workers. A limited data is available for lung cancer cases due to the indoor radon exposure (Field., 2001; Field et al., 2000; ICRP-60, 1990; Field and Becker, 2001).

The health impact of radon is based upon the amount of dose received and the number of lung cancer cases due to that dose. The estimates of relation of lung cancer and radon exposure rely upon the extrapolation of lung cancer cases in miners. Fig. 3.5 illustrates the excess lung cancer risk as a function of indoor radon level.

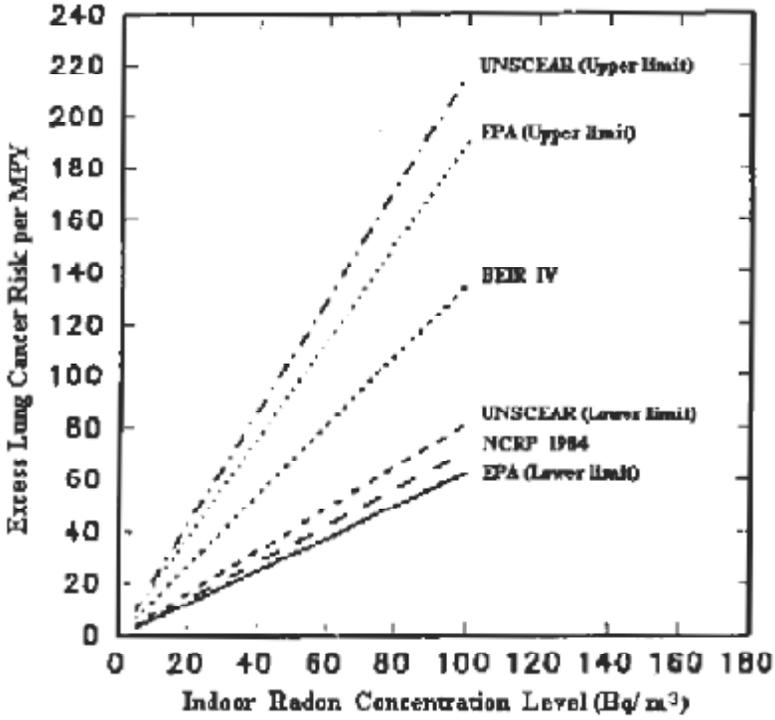


Fig.3.5 Excess lung cancer risk as function in indoor radon levels

### 3.10 Action and Reference level

World Health Organization (WHO) presented in 2009 a recommended reference level (the national reference level),  $100 \text{ Bq/m}^3$ , for radon in dwellings. The recommendation also says that where this is not possible,  $300 \text{ Bq/m}^3$  should be selected as the highest level. A national reference level should not be a limit, but should represent the maximum acceptable annual average radon concentration in a dwelling as recommended by World Health Organization. The actionable concentration of radon in a home varies depending on the organization doing the recommendation. For example, the United States Environmental Protection Agency encourages that action be taken at concentrations as low as  $74 \text{ Bq/m}^3$ , and the European Union recommends action be taken when concentrations reach  $400 \text{ Bq/m}^3$  for old houses and  $200 \text{ Bq/m}^3$  for new one. On 8 July 2010, the UK's Health Protection Agency issued new advice setting a "Target Level" of  $100 \text{ Bq/m}^3$  whilst retaining an "Action Level" of  $200 \text{ Bq/m}^3$ .

### 3.11 Radon Measurement Techniques

As was mentioned before, radon is the gas daughter of  $^{226}\text{Ra}$  from the natural series of uranium and thorium. Radon has three isotopes  $^{222}\text{Rn}$ ,  $^{220}\text{Rn}$  and  $^{219}\text{Rn}$ . Because of the short half-lives of  $^{220}\text{Rn}$  and  $^{219}\text{Rn}$ , they travel a lesser extent and decay before they can be detectable.

Accordingly, in the following text the word and measurement techniques will refer to  $^{222}\text{Rn}$  exclusively, because of the long half-life and diffusion length extent. Radon decaying with alpha particle emission in such case the measurements can thus be performed directly on radon itself "called radon alone measurements" or indirectly on radon and/or its daughters. Radon and its progeny decay by  $\alpha$ - particles, while these radon products of  $^{214}\text{Pb}$ ,  $^{214}\text{Bi}$ ,  $^{210}\text{Pb}$  and  $^{210}\text{Bi}$  are also  $\beta$ - emitters as well as  $\gamma$ -ray accompany their  $\beta$ -emission. Therefore, radon detection and measurement can be performed through the detection and measurements of these radiations by understanding basic knowledge of interaction of these radiations with matter. Many techniques are available for measurements

of radon. These are classified into two main categories passive and active detectors techniques.

### **3.11.1 Passive Detector Techniques**

In the long term measurements of radon, the passive detectors is the most widely used technique, particularly the nuclear track detectors such as CR-39 equipped with the sealed can technique, which incorporate the effect of seasonal, weather and environmental conditions on radon concentration in dwelling. It is very important to carry out measurements over a long period. Passive devices are the most widely used and practical way to determinate radon concentration, where, the detection sensor usually placed inside a container. The container is meant to protect the detector and make room around the detector for sensitive volume large enough so as to have many alpha particles produced and detected in as short time as possible (Durrani and Ilic', 1996). There are other types of passive detectors such as TLD, Electrets, Charcoal canister technique.

- **Thermoluminescent Detectors.**

Based upon the principle of some of substances, which have ability to release an energy in the form of light, when those heated to a high temperature. A several thermoluminescent dosimeters (TLDs) are developed. The incident ion radiations interact with the matter of the detector, some of it will eject electrons from their position to the conduction band and leaving holes in the valence band. After heating the detectors those electrons will release and recombined with holes in the valence band, light is emitted. In the case of the radon detection, one must rely on the activity of the radon daughters. The TLDs are sensitive to the alpha, beta and gamma radiations. To determine the alpha radiation, two TL detectors will be mounted in an inverted cup and placed in the ground. One of the TLDs is wrapped in a foil that will exclude the alpha particles, but not exclude gamma and beta particles, the other one will be exposed directly to the alpha, beta and gamma

radiations. After a period of exposure to the radon, the TLDs recovered, heated to 300 °C and read in the TLDs apparatuses. The calculation of the alpha contribute from radon and radon daughters is by subtracting the intensity of energy in first TLD that exposed to only beta and gamma radiation from the second TLD that exposed to alpha , beta and gamma radiation (Hussein, 1997).

- **Electrets**

Electret radon monitor is a technique to measuring the radon concentration levels (Kotrappa et al., 1988). The detectors of the Electret ions are a piece of dielectric material that are exhibit almost a permanent electric charge and are widely used for long-term radon measurements. Ions produced by radon decay are strike and reduce the surface voltage of the electrets. By measuring the voltage reduction, the radon concentration can be calculated.

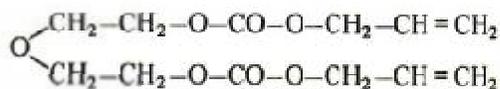
- **Charcoal Canister Technique**

This technique is also called radon on activated charcoal (ROAC) (Durrani and Ili'c., 1997) and it is used for rapid radon survey. The charcoal canister technique uses for adsorption the radon gas, where a few grams of charcoal have deposited inside the plastic can and exposed to the air for few days; Typically, from 4 to 15 days, radon easily enter to the canister. The plastic can be retrieved and then determined the gamma activity of the radon daughters  $^{214}\text{Pb}$  and  $^{214}\text{Bi}$  those trapped in the charcoal by the gamma spectroscopy or the liquid scintillation counter. But this technique has problem with the need to sophisticated electronics for analyses.

- **Nuclear Track Detectors ( Detector Material)**

Plastic track detectors provide the most common method of radon measurements, because the detectors are small, easy to use, inexpensive and simple to process and read. CR-39 (Columbia Resin-1939 from Pittsburg Glass Co.). CR's chemical name is Polyallyli di-glycol carbonate (PADC) and the chemical structure of CR-39 is shown in Fig. 3.6.

Its element composition is (C<sub>12</sub> H<sub>18</sub>O<sub>7</sub> ) with appearance clear, colorless, rigid plastic its density is 1.32 g.cm<sup>-3</sup>



**Fig 3.6** Chemical structure of CR-39.

The plastic track detector used in the current study is Tas-track, which is manufactured by Track Analysis System Ltd., Bristol, UK. The detector comes in sheets of thickness 750 μm. The detector was kept away from background radon using plastic adhesive coat. It has been found that these materials are excellent for alpha particles detection and radon measurements because they are by far the most sensitive material, and capable of recording protons and alpha particles with a wide range of energies. Another advantage of these detectors is the clarity of their surface after etching and their low background. In order to distinguish each detector, an inscription code was engraved on top corner of its surface.

### • Methodology and Track Formation

When an ionizing charged particle passes through a dielectric material, the transfer of energy to electrons results in a trail of damage molecules along the particle trajectory. In some materials, the track can be made visible upon etching in strong acid or base solution. The entire surface of the material is attacked, but those points at which particle tracks have entered are etched about 10 times faster. The tracks can thus be made to form pits on the surface that are large enough to be easily visible through conventional microscope.

In common with other passive detectors, track-etch detectors have the advantage of simplicity and low cost. They also possess a very useful inherent threshold, in that there is a minimum value of the specific energy loss (— — ) required of the particle before the damage is severe enough to lead to an etchable track. The threshold is always well above the specific energy loss of an electron track, so that track-etch material are

inherently insensitive to fast electron or gamma ray interactions. The damage created by the incident particle can be through collision of the particle itself or from the energetic delta rays created along its track. The range of the delta rays may extend approximately 5 nm in any direction away from the particle position, so that the radius of the primary damage track is thought to be about this dimension (Fleischer et al., 1975).

- **Track Etching Geometry**

Nuclear particles incident on track detector may be characterized by their charge or effective charge  $Z$ , mass  $m$ , and energy  $E$  or relative velocity,  $v = \beta c$ . Differences in these particle parameters manifest themselves as changes in the track length  $R$  and the track-etching rate  $v_e$  as well as in the variation of  $v_e$  with position along the track. The most easily measurable parameters of an etched track are the etched-cone length  $L$  or the length  $L_p$  projected onto the detector surface and the major and minor axes  $a$ ,  $b$  of the etch pit opening. The track etching geometry also determines quantities such as the efficiency of a given detector which is defined as the ratio of the number of the observed etched tracks to the number of latent damage tracks crossing a unit area of the original surface of the detector where the etching first starts. The calculation of the etched-track parameters is comparatively simple when the track etch rate  $v_e$  can be taken to be constant along the portion of the latent damage track, which is etched out. This condition will apply in many instances where the ionization rate of the particle is not varying rapidly. Furthermore, the constant  $v_e$  model allows a number of features of the track etching process to be established fairly simply.

Initially, we shall make a further simplification by concentrating on a track, which is normally incident upon the detector surface. Now, the linear rate of attack down the track is  $v_e$  the radial extent of the enhanced etchability is assumed to be very small compared with the final dimensions of the etched track, so that in an etching time  $t$  the etch pit will extend to a distance  $L$  from the point of origin, where  $L = v_e t$ . However, the surface is also being removed at a rate  $v_s$ ; so that the length of the etch pit is  $L_p = L - v_s t$ . (3.2)

At etch point along the track, the etchant moves outwards at a rate  $v_B$ . Any point at a distance  $r$  from the beginning of the track is reached by the etchant at a time  $(t) = r/v_B$ ; and there is a residual time  $(t - r/v_B)$  available for the etchant to attack radially outwards from the point at  $r$  to a distance  $r \cdot (t - r/v_B)$  in the medium. The three-dimensional pit wall is then formed by the locus of all the spheres of radius  $r \cdot (t - r/v_B)$ , where  $r$  is the variable. It will be seen from Fig. 3.6 that this leads to the formation of a cone with semi-cone angle  $\theta_c$  given by:

$$\sin \theta_c = \frac{v_B}{v_T} = \frac{v_B \cdot t}{v_T \cdot t} = \frac{V_B \cdot t}{V_T \cdot t} \quad (3.3)$$

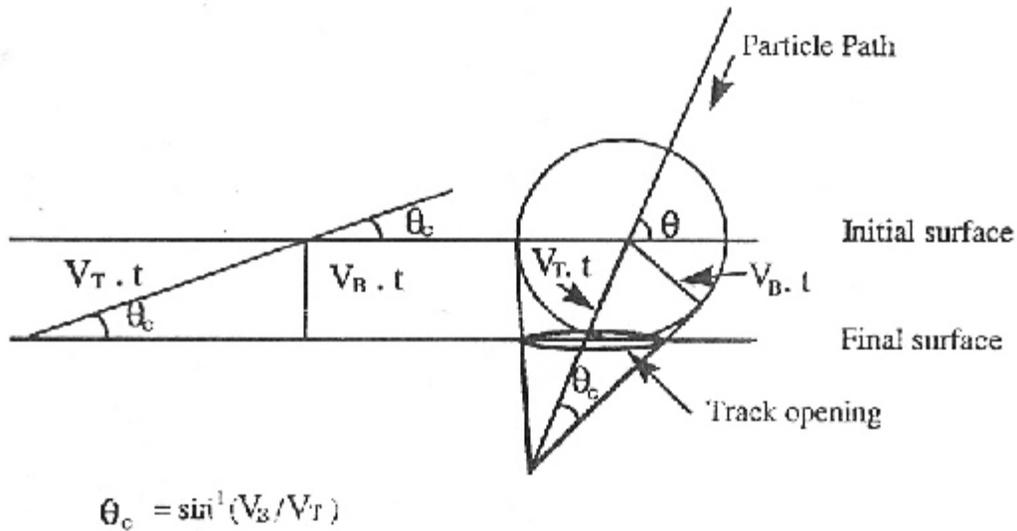


Fig. 3.7 The critical angle of etching

This angle  $\theta_c = \sin^{-1}(v_B/v_T)$  is also known as the critical angle of etching from the triangle O'PT in Fig. 3.7 it is apparent that

$$\frac{v_B}{v_T} = \sin \theta_c \quad (3.4)$$

From Eq. (3.2), it follows that

$$\tan \theta_c = \frac{v_B}{v_T} \quad (3.5)$$



$$= \frac{D_1}{D_2} = 2 \frac{V_1}{V_2} \quad (3.8)$$

Where,  $V = V_1 / V_2$ . Since for most plastics and etching conditions used the track etch rate is very much higher than the bulk etch rate so that  $V > 1$ , we have

$$= \frac{D_1}{D_2} \cong 2$$

or

$$= \frac{D_1}{2D_2} \quad (3.9)$$

Thus a measurement of the diameter of the normally incident alpha particle tracks at known etching time yields a value for the bulk etch rate. If the ratio of the diameters of some tracks having an etch rate ratio  $V$  to those of alphas is denoted by  $R$ , then we write Eq. 3.8 and 3.9

$$= \frac{D_1}{D_2} = \frac{1}{(V - 1)/(V + 1)}$$

and upon rearranging, we get the useful formula

$$= \frac{(1 + R)}{(1 - R)} \quad (3.10)$$

Most treatments of track geometry are formulated directly in terms of the thickness of the removed surface layer  $h = \dots$ .

### 3.11.2 Active Technique

These devices are used normally for short-term measurements. The active technique is based on method in which grab sample of air is collected at instant time followed by measurements of radon concentration through  $\alpha$ -particles activities. The active measurements techniques are not so beneficial to accurately measure the radon levels because of temporal variation in radon level due to the temperature and pressure gradients. Examples of these techniques are Lucas cell and Ionization chamber.

- **Lucas Cell (Scintillation Method).**

The detection of ionizing radiation by the scintillation light produced in certain materials is one of the oldest techniques to measuring the radon in air. The device consist of a glass vessel coated internally with scintillating material such as ZnS, except the bottom and the surface which is transparent and coupled with photomultiplier tube (PMT) employed to count the scintillation produced. The alpha particles produced from radon decay from the sample of air inside the cell will interact with the ZnS. Light emitted from the ZnS, is detected by (PMT) and generate an electric pulse. The efficiency of these cells is typically 70 to 80%.

- **Ionization Chamber**

The ionization chambers are not widely used as scintillation counters, since the ionization chambers are expensive. Alpha particles produced from the radon decay and its daughters can be detected in the ionization chambers, whereas the alpha particles ionize air in the chambers. The resulted current is measure of quantity of decayed radon atoms.

Many of active devices are use in radon measurements, but in this present work, where, the passive technique was based on nuclear track detectors (NTDs) and more details will be given on the fourth chapter.

### **3.12 Theoretical Considerations for Radon Exhalation Rates**

In order to measure the radon exhalation rate from samples such as soil or building materials, which considered as source of radon atoms, passive or active techniques are widely used for that purpose. In a closed chamber, ( ) is the number of radon atoms that exhaled from the material samples in the emanation container? The variation rate of ( ) with time is given by equation:

$$\frac{dN}{dt} = -\lambda N \quad (3.11)$$

Where

$(N)$  : Total number of radon atoms in closed chamber at time  $t$ ,

$(E)$  : Exhalation rates ( $\text{Bq}\cdot\text{m}^{-2}\cdot\text{h}^{-1}$ ) or ( $\text{Bq}\cdot\text{kg}^{-1}\cdot\text{h}^{-1}$ ),

$(A)$  : Area of sample  $\text{m}^2$ ,

$\lambda$ : decay constant of radon  $\text{h}^{-1}$ ,

the solution of the equation (3.11) with initial condition  $(N)_0=0$  is given by:

$$(N) = \frac{EA}{\lambda} (1 - e^{-\lambda t}) \quad (3.12)$$

If  $V$  is the air volume ( $\text{m}^3$ ) then, the radon concentration in air is  $= \frac{N}{V}$ , by using equation( 3.2) we get

$$\left(\frac{N}{V}\right) = \frac{EA}{\lambda V} (1 - e^{-\lambda t}) \quad (3.13)$$

The track density increases with the increasing radon concentration, and is given by:

$$D = K \left(\frac{N}{V}\right) \quad (3.14)$$

Where

$(D)$  : track density (number of tracks/ $\text{cm}^2$ ),

$K$ : detector efficiency,

From (3.13) and (3.14) we get:

$$D = \frac{KEA}{\lambda V} (1 - e^{-\lambda t}) \quad (3.15)$$

Applying the initial condition  $(D)_0 = 0$  will get the following solution:

$$= \frac{V}{V_0} - (1 - \frac{V}{V_0}) \quad (3.16)$$

The volume of air in this study is the volume of air content in the emanation container and it is equal to the free space in the container (V).

$$= h \quad (3.17)$$

Where  $h$  : the height of container

By using the equation (3.17) in the equation (3.16) we get the radon exhalation rate according to the following equation;

$$= \frac{V}{V_0} - (1 - \frac{V}{V_0}) \quad (3.18)$$

### 3.13 Radon Concentration Units

As already mentioned before, radon concentration refers to  $^{222}\text{Rn}$  concentration because it has a long half-life of 3.82 days compared to  $^{220}\text{Rn}$  which has short half-life of about 55.6 s. Radon concentration is usually measured in the atmospheric air, in Becquerel per cubic meter ( $\text{Bq}/\text{m}^3$ ), the SI derived unit. It is often measured in picocuries per liter ( $\text{pCi}/\text{l}$ ) in the USA, with  $1\text{pCi}/\text{l} = 37 \text{Bq}/\text{m}^3$ . In the mining industry, the exposure is traditionally measured in working level (WL), which defined as any level of concentration or burden of radioactivity in a given air environment, which produced a prespecified potential alpha energy concentration. The cumulative exposition in working level month (WLM): 1 WL equals any combination of short-lived  $^{222}\text{Rn}$  progeny ( $^{218}\text{Po}$ ,  $^{214}\text{Pb}$ ,  $^{214}\text{Bi}$ , and  $^{214}\text{Po}$ ) in one liter of air that releases  $1.3 \times 10^5 \text{MeV}$  of potential alpha energy. One WL is equivalent to  $2.08 \times 10^{-5}$  joules per cubic meter of air ( $\text{J}/\text{m}^3$ ) (Toxicological profile for radon; 1990). The SI unit of cumulative exposure is expressed in joule-hours per cubic meter ( $\text{J}\cdot\text{h}/\text{m}^3$ ). One WLM is equivalent to  $3.6 \times 10^{-3} (\text{J}\cdot\text{h}\cdot\text{m}^{-3})$ . An exposure to one WL for one working month (170 hours) equals one WLM cumulative exposure. A cumulative exposition of one WLM is

roughly equivalent to living one year in an atmosphere with a radon concentration of  $230 \text{ Bq}\cdot\text{m}^{-3}$  (CEA; 2005).

$^{222}\text{Rn}$ , when released into the air, decays to  $^{210}\text{Pb}$  and other radioisotopes. The levels of  $^{210}\text{Pb}$  can be measured. The rate of deposition of this radioisotope is weather dependent. Radon concentrations found in natural environments are much too low to be detected by chemical means. A  $1000 \text{ Bq}/\text{m}^3$  (relatively high) concentration corresponds to  $0.17$  pictogram per cubic meter ( $\text{pg}/\text{m}^3$ ). The average concentration of radon in the atmosphere is about  $6 \times 10^{-20}$  atoms of radon for each molecule in the air, or about  $150$  atoms in each ml of air. The radon activity of the earth atmosphere originates from some tens of grams of radon, consistently replaced by decay of larger amounts of radium and uranium.

### 3.14 Special Quantities and Units for Radon and Radon Decay Products

#### 3.14.1 Units of Activity

The activity of radioactive source is defined by:

$$= \text{---} \quad (3.19)$$

Where  $N$  is the number of decays in time  $t$ . The SI unit of activity is the Becquerel (Bq), equal to one nuclear transformation per second.

$$1 \text{ Curie (Ci) (traditional unit)} = 3.7 \times 10^{10} \text{ Bq.}$$

#### 3.14.2 Units of Activity Concentration

It is activity per unit of volume or mass, whose SI unit is the Becquerel per cubic meter ( $\text{Bq}/\text{m}^3$ ) or the Becquerel per kilogram ( $\text{Bq}/\text{kg}$ ).

$$1 \text{ pCi}/\text{l (traditional unit)} = 37 \text{ Bq}/\text{m}^3.$$

#### 3.14.3 Absorbed Dose

It is defined by  $D = E / m$ , where  $E$  is the mean energy imparted to mass of matter by ionizing radiation. For radiation protection purposes, the average absorbed dose by a tissue or organ is defined as  $D = E / m$ , where  $E$  is the total energy imparted to a tissue or organ of mass  $m$ .

The SI unit of absorbed dose is the Gray (Gy), equal to one Joule per kilogram (J/kg). 1Gy = 100 rad (traditional unit).

### 3.14.4 Equivalent Dose

The equivalent dose is the product of absorbed dose and the weighting factor ( $W_R$ ) for specific type of radiation. The weighting factor  $W_R$  accounts for the different ability of the different type of radiation to cause biological damage. For beta particles, gamma-rays and x-rays  $W_R$  is taken to be unity. For alpha particles  $W_R$  is 20, whereas for neutrons it is ranging from 5 to 20, depending on its energy (ICRP., 1991).

The SI unit of equivalent dose is the Sievert (Sv), equal to one Joule per kilogram. 1Sv = 100 rem (traditional unit).

### 3.14.5 Effective Dose

This is the sum of the products obtained by multiplying the equivalent doses to various organs and tissues by appropriate risk weighting factor  $W_R$  for each. This quantity is considered proportional to the total probability of stochastic effects. The most recent estimation of risk of stochastic effects for general population, expressed as probability of fatal cancer per unit of effective dose, is  $5 \times 10^{-2} \text{ Sv}^{-1}$  (ICRP., 1991).

### 3.14.6 Radon Progeny Concentration

The concentrations of the four short-lived radon daughters are usually expressed in terms of potential alpha energy concentration (PAEC) or equilibrium equivalent radon concentration (EERC or EER). These two quantities combine the four concentrations in a single value, using weighting factors that take into

account the relative importance for the health effects to the lung, which are due to the alpha particles emitted by the radon daughters along the decay chain down to  $^{210}\text{Pb}$ .

The potential alpha energy concentration (PAEC) of any mixture of radon daughters is the sum of the potential alpha energy of all the short-lived radon daughters atoms present per unit of air volume. Its SI unit is the Joules per cubic meter ( $\text{J}/\text{m}^3$ ), but the traditional unit of the working level (WL) is still widely used, representing a PAEC of  $1.3 \times 10^5$  MeV per liter, where  $1 \text{ WL} = 2.08 \times 10^{-5} \text{ J}/\text{m}^3$ .

The equilibrium equivalent radon concentration (EERC) is the fictitious activity concentration of radon in equilibrium with its daughters (i.e. all daughters having the same concentration as radon; giving the same potential alpha energy concentration (PAEC) of the actual non- equilibrium mixture of radon daughters. Its SI unit is the Becquerel per cubic meter ( $\text{Bq}/\text{m}^3$ ). A potential alpha energy concentration (PAEC) of 1WL correspond to about  $3700 \text{ Bq}/\text{m}^3$  ( $=100 \text{ pCi}/\text{l}$ ) of EERC.

### **3.14.7 Equilibrium Factor**

The ratio of EERC to actual radon concentration is called equilibrium factor. It is usually represented by the symbol "EF". Due to the actual no-equilibrium conditions, its value in indoor air is usually in the range 0.2-0.7.

### **3.14.8 Cumulated Exposure to Radon Progeny**

Its practical measurement unit is the working level month(WLM). It corresponds to an exposure to about  $3700 \text{ Bq}/\text{m}^3$  of radon progeny concentration in equilibrium with range radon gas (i.e. all progeny having the same concentration as radon; actually there is always a certain degree of disequilibrium), during the working period of one month (170 hours).

This unit was historically used for occupational exposure in mines, while for indoor exposure of the public in dwellings it is preferable to express the

cumulative exposure in terms of mean radon gas concentration during a one-year period, since this is the quantity that is usually measured in dwellings. The following conversion formula can be used:

$$1\text{WLM} \cong 72/(\text{EF} \times \text{IOF}) \text{ Bq/m}^3.$$

Where EF is the equilibrium factor between radon and its progeny, and IOF is the indoor occupancy factor, i.e. the fraction of time spend indoor. Using EF = 0.5 and IOF = 0.8, a cumulated exposure of 1 WLM corresponds to an exposure to about 180 Bq/m<sup>3</sup> of radon gas concentration during the period of 1 year, or 90 Bq/m<sup>3</sup> for period of 2 years, and so on. If EF= 0.4 is measured, then 1 WLM  $\cong$  220 Bq/m<sup>3</sup>.

## Chapter 4

### Materials and Methods

#### 4.1 Samples Collection

Various types of building materials, which are used, for construction and decoration in typical houses in Libya specifically in Benghazi are shown in Fig 4.1. were collected for testing. The commonly used market available building materials were granite, marble, ceramic, block, gypsum and cement which were collected randomly from market stores. Some of these building materials are domestics and other ones are imported. Some of these materials are solid that have different surface areas and different thicknesses while others are porous powder. Each sample material was coded to distinguish the samples from each other, where the first letter of code refers to the type of sample material, second letter refers to the origin of the sample material and the third letter refers to the state of sample material; normal, dry or wet, respectively. As an example; (CF-N), C; refers to ceramic, F; refers to France and N refers to normal material.

Eighteen normal (non-dried) solid samples consisting of six marbles, four gypsums, three ceramics, three blocks and two granites have been investigate and listed in table 5.1. The samples were taken directly from the construction material stores, and then put where the emanation container was inversely mounted on the surface of these tile samples.

Another group from the same solid sample materials was dried in an oven at temperature of  $105 \pm 1^{\circ}\text{C}$  for 24 hrs. to remove the moisture content. These samples were loaded into the emanation container directly after the dryness procedure and the container was tightly sealed. The same procedure was applied for five powder materials of 0.5 Kg consisting of black and white cement, gypsum, sand and soil. These dried powder samples were gently pressed to form a disk-like shape, which permit radon to diffuse out of this host material with a high degree of homogeneity.

All solid and powder samples were introduced into water in a vessel. Each sample was then weighed once every week until the weight reached the saturated-steady state and the sample was considered to be in wet condition. After about 6 weeks the samples were removed from water vessel and all water allowed to drip off them and these the wet samples were then directly mounted in the emanation container.



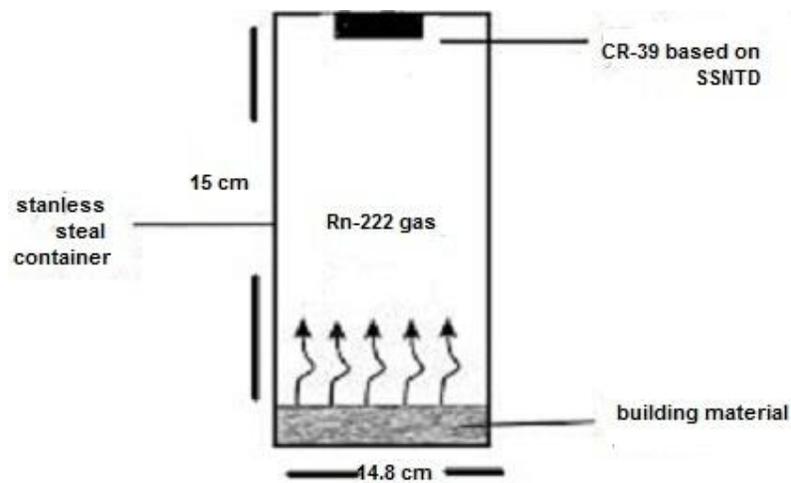
**Fig. 4.1** Solid building material samples.

## 4.2 Radon Dosimeter

Radon dosimeter is a device used to assess the radon dose. Radon concentration levels were measured using passive-technique based on solid state nuclear track detectors (SSNTDs) type of CR-39 which record the average radon concentrations over long-term measurements.

The radon dosimeter consists of cylindrical cup made of stainless-steel of diameter 14.7 cm, length 14.8 cm and volume  $2.513 \times 10^{-3} \text{ m}^3$  (2.513 ) as shown in Fig. 4.2 (a, b). The CR-39 detector, which were cut into  $1.5 \times 1.5 \text{ cm}^2$  pieces, was fixed on the inner lid surface of the cup; the distant from the sample to the top of cup is about 15 cm. This distant was chosen to detect  $\text{Rn}^{222}$  and keep its daughters and  $\text{Rn}^{220}$  away from

detection. By taking advantage of the difference between half-life of  $^{222}\text{Rn}$  (3.83d) and that for  $^{220}\text{Rn}$  (55.6s), it takes too long for  $^{222}\text{Rn}$ 's daughters and  $\text{Rn}^{220}$  to pass through this distance or diffusion barrier before decaying away, while  $\text{Rn}^{222}$  can go through without appreciable loss. A double-adhesive tape onto the bottom surface of the container fixed the CR-39 detector. This container was sealed tightly with silicon to prevent the escape of radon. Long radon exposure period of 90 days was chosen in order to obtain good statistics.



**Fig 4.2** (a; Schematic diagram showing detailed structure of the CR-39 dosimeter, b; Radon dosimeter (emanation container).

### 4.3 Chemical Etching Process

Chemical etching is the most widely used method of fixing and enlarging the image of the latent damage trail in a solid-state track detector. Essentially, etching takes place via rapid dissolution of the disordered region of then undamaged bulk material. For the plastics, the most frequently used etchant is the aqueous solution of NaOH with concentrations typically within the range 1 to 12 mol. The etching is done at constant temperature using digital water bath. The temperature is usually set in the range 40 – 70°C.

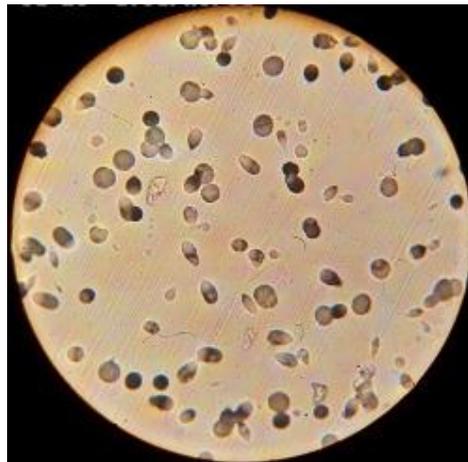
In the present work, directly after exposure, the detectors were chemically etched in 6.25-M NaOH solution as shown in Fig 4.4 at  $60 \pm 1$  °C for different etching periods. The detectors were then washed in distilled water in order to remove contamination by the etching solution on the surface of the detector material.



**Fig 4.3.** The chemical etching solution of NaOH.

#### 4.4 Counting of Radon Tracks under Optical Microscope

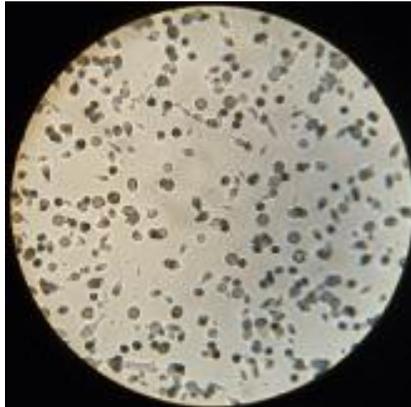
Since track detector materials are transparent, optical transmission microscopes are the easiest and most widely used counting tool. Most chemically etched track range in length from less than  $1\mu\text{m}$  to perhaps 10 or 15  $\mu\text{m}$ . Therefore, the microscope must have an overall magnification in the range 100x to 1000x. Information other than track density, such as track size or orientation, can also be obtained using an optical microscope. In the case of radon monitoring, it is often required to count tracks in many microscopic field for a large number of samples. The latent damage trails resulted from alpha particles due to radon exposure in the emanation container become permanently fixed as "tracks", as seen in Fig 4.4 (a, b and c), after a chemical etching process. These etched tracks are also visible under an ordinary optical microscope with magnification of 400 x that used for counting the tracks as shown in Fig 4.5. Hundred fields are scanned for each detector under theocular lens of microscope. However, it is necessary to count background tracks using an unexposed detector. The next step is to calculate the arithmetic mean for the hundred fields, which is the number of tracks per field, and finely the number of tracks per  $\text{cm}^2$  were calculated.



a. medium density  $\alpha$  particles tracks.



b. Low density of  $\alpha$  particles tracks.



C. high density of  $\alpha$  particles tracks.

**Fig 4.4 a, b, c;** Track of alpha particles under optical microscope.



**Fig 4.5** The optical microscope used in the measurements.

## 4.5 Experimental Procedures

After counting the number of tracks per field using optical microscopic, the following parameters; were determined, calculated, and estimated, respectively; radon activity concentration, radon exhalation rates, annual effective dose and the radium content.

### 4.5.1 Determination of Radon Activity Concentration, $C_{Rn}$

After counting tracks procedure, the determination of radon concentration in the volume air of container was done by calculation of alpha track density. The track density ( $\rho$ ) in the samples was calculated according to the following formula:

Track density ( $\rho$ ) = average number of total tracks/ area of field view

$$\rho = \text{average number of total tracks} \times 2973.3\text{cm}^{-2}$$

The exposure time or effective exposure time in the emanation container was measured according to equation

$$= -1/ \quad (1 - \quad ) \quad (4.1)$$

Where

: exposure time to reached the equilibrium build up between radon and radium in the closed can.

: decay constant of <sup>222</sup>Rn which is equal to:

$$= \frac{\quad}{\quad} = \frac{\quad}{\quad} = 7.56 \times 10^{-3} \text{ h}^{-1} \quad (4.2)$$

Using the effective time equation, the radon activity concentration was calculated using following equation

$$= (1 - \quad ) \quad (4.3)$$

where

$$= \quad (4.4)$$

whereas

: The decisive value of radon activity concentration

: Track density per unit area

: Calibration factor of CR-39 track detector

The radon activity concentrations and other parameters were measured and then used to calculate the radon exhalation rates from building material samples, and were tabulated in the next chapter.

#### 4.5.2 Calculation of Radon Exhalation Rates, $E_x$

Radon level of building materials in an emanation container can be monitored with passive radon dosimeter based on the CR-39 solid-state nuclear track detector SSNTD. The radon concentration in the emanation container was measured hourly for a long period (90 days). The radon concentration in the closed can reached secular equilibrium within several half-lives of radon. It should be noted that the radon exhalation rates are constant, whereas the radon concentration reaches a maximum value that depends on the exhalation rate. The passive radon detector can be used to measure the accumulated activity and then to evaluate the average radon exhalation rate from the building materials using the following equations (Saad, 2008; 2010, Fleischer and Mogro-Campero, 1978; Khan et al., 1992; Kumar et al., 2005, 2008; Mahur et al., 2008a, 2008b; Singh et al., 2008; Rafique et al., 2011):

$$= \frac{\text{—————}}{\text{( —————)}} \quad (4.5)$$

$$= \frac{\text{—————}}{\text{( —————)}}$$

(4.6)

where

- : The areal exhalation rate ( $\text{mBq.m}^{-2}.\text{h}^{-1}$ ),
- : The mass exhalation rate ( $\text{mBq.kg}^{-1}.\text{h}^{-1}$ ),
- : The volume of emanation container ( $\text{m}^3$ ),
- : The surface area of building material sample ( $\text{m}^2$ ),
- : The sample mass (kg),

- : Integrated radon exposure (Bq.m<sup>-3</sup>),
- : Decay constant (h<sup>-1</sup>),
- : Time elapsed since sealing ( h),

### 4.5.3 Estimation of the Annual Effective Dose, E<sub>p</sub>

The radiation hazard of people's prolonged exposure to high concentration of radon and its daughters in a room with an air ventilation rate of 0.5 h<sup>-1</sup> were estimated. The risk of lung cancer from domestic exposure to radon and its daughters can be estimated directly from the effective dose equivalent. The radiation hazard of radon and its decay products were estimated from the radon exhalation rate of the building material samples. The contribution of indoor radon concentration from building material samples can be calculate using the following expression (Nazaroff and Nero, 1988).

$$= \frac{\cdot}{\cdot} \tag{4.7}$$

where

- : Radon concentration (Bq.m<sup>-3</sup>),
- : Radon exhalation rate (Bq.m<sup>-2</sup>.h<sup>-1</sup>),
- : Room volume (m<sup>3</sup>),
- : Air exchange rate ( h<sup>-1</sup>),

In these calculations, the maximum radon concentration from the building materials was assessed by assuming the room to be a cavity with — = 2.0 m<sup>-1</sup> and an air exchange rate of 0.5h<sup>-1</sup>. The annual effective dose equivalent, , is then related to the average radon concentration by the following expression (Saad et al., 2010):

$$[ \cdot ] = \frac{(\cdot) \cdot (\cdot)}{(\cdot) \cdot (\cdot)} \tag{4.8}$$

Where  $C_{Ra}$  is in  $Bq.m^{-3}$ ,  $n$  is the fraction of time spent indoors,  $F$  is the equilibrium factor, 8760 is the number of hours per year and 170 is the number of hours per working level month. The values of  $F = 0.8$  and  $n = 0.42$  were used to calculate  $C_{Ra}$ . For radon exposure, the effective dose equivalents were estimated by using a conversion factor of 6.3 mSv/WLM (ICRP, 1987).

#### 4.5.4 Estimation of The Effective Radium Content, $C_{Ra}$

The effective radium content in the soil samples was determined using a track detection method (Somogyi, 1986). Once radioactive equilibrium between radium and radon is established, one may use the analysis of the radon alpha activity for the estimation of the steady-state activity concentration of radium. After the sealing of the can, the activity concentration of radon begins to increase with time  $T$  as follows:

$$C_{Ra} = \frac{C_{Rn}}{F} \left( 1 - e^{-\lambda T} \right) \quad (4.9)$$

Where  $C_{Ra}$  is the effective radium content of the building material samples? This effective radium content of a specimen in a sealed-can can be estimated as:

$$C_{Ra} (Bq.kg^{-1}) = \frac{C_{Rn}}{F} \left( 1 - e^{-\lambda T} \right) \quad (4.10)$$

Where  $h$  is the distance between the detector and the top of the building material sample in m,  $C_{Rn}$  is the counted track density,  $k$  is the calibration factor of the CR-39 track detector, and  $T$  denotes the effective exposure time. The exposure time in these measurements was 90 days.

## Chapter 5

### Results and Discussion

#### 5.1 Radon Exhalation from Non-Dried and Dried Building Materials

In fact, all building materials used in construction and decoration of houses are originated from Earth's crust. Marble, gypsum, sand, granite, blocks and cement are the basic materials used in the building of typical houses. In Benghazi, Libya, all houses are built using these materials. All of our experimental results are summarized in Tables (5.1, 5.2, and 5.3). These tables show the radon activity concentrations, radon contribution to indoor radon, the areal and mass exhalation rates, the radium equivalent contents, and the annual effective dose equivalents for the building material samples, (i.e, non-dried tiles, dried slabs, and dried porous powdery materials). The variations (minimum, maximum, range, and mean values) of radon activity concentrations (in  $\text{Bq m}^{-3}$ ) and radon exhalation rates in terms of area and mass, together with the statistical uncertainties ( $1\sigma$ ) and standard deviations (S.D.) for these building materials, are also provided in Tables (5.1, 5.2, and 5.3). The radon activity concentrations in the non-dried samples were found to vary from  $91.5 \pm 3.2$  to  $2265.7 \pm 79.2 \text{ Bq m}^{-3}$ , with a mean, standard deviation, and range of  $515.7 \pm 18.0$ ,  $529.6 \pm 18.5$ , and  $2174.2 \pm 76.0 \text{ Bq m}^{-3}$ , respectively. Those of the dry materials varied from  $264.6 \pm 9.3$  to  $2283.3 \pm 79.8 \text{ Bq m}^{-3}$ , with a mean, standard deviation, and range of  $698.6 \pm 24.4$ ,  $548.4 \pm 19.1$ , and  $2018.6 \pm 70.5 \text{ Bq m}^{-3}$ , respectively. In general, the porous powdery material samples showed insignificant radon exhalation, except for one sample of soil and another of sand. In addition to being fine grained, they could be expected to have low radon emanations, as seen in Table 5.3. The radon activity concentrations of these powdery materials varied from  $441.1 \pm 3.5$  to  $906.5 \pm 31.7 \text{ Bq m}^{-3}$ , with a mean, standard deviation, and range of  $635.5 \pm 22.2$ ,  $165.7 \pm 5.8$ , and  $465.4 \pm 16.3 \text{ Bq m}^{-3}$ , respectively.

The results obtained for the non-dried tile samples showed a maximum radon concentration of  $2265.7 \pm 79.2 \text{ Bq m}^{-3}$  for sample  $G_n\text{In-N}$ , which was an Indian granite.

Sample M<sub>2</sub>It-N, an Italian black marble, showed the second largest value,  $1277.1 \pm 44.6 \text{ Bq m}^{-3}$ . The third largest radon value,  $965.1 \pm 33.7 \text{ Bq m}^{-3}$ , was for sample G<sub>n</sub>E-N, an Egyptian granite. The results for the dried solid slab samples showed a maximum radon concentration of  $2283.3 \pm 79.8 \text{ Bq m}^{-3}$  in sample G<sub>n</sub>In-D, an Indian granite. The next five largest radon concentrations were  $1456.4 \pm 50.9$ ,  $1293.1 \pm 45.2$ ,  $810.8 \pm 28.3$ ,  $716.9 \pm 25.5$ , and  $600.6 \pm 21.0 \text{ Bq m}^{-3}$  for samples M<sub>2</sub>It-D, G<sub>n</sub>E-D, G<sub>Y2</sub>E-D, BIt-D, and MG<sub>r</sub>-D, denoting Italian marble, Egyptian granite, Egyptian gypsum, Italian brick, and Greek marble, respectively. The results for the dried porous powdery building materials showed a maximum radon concentration of  $906.5 \pm 31.7 \text{ Bq m}^{-3}$  in sample S<sub>o</sub>L, a Libyan soil sample. Sample S<sub>a</sub>L, Libyan sand, showed the second largest value,  $735.7 \pm 25.7 \text{ Bq m}^{-3}$ .

The areal radon exhalation rates from the non-dried samples varied from  $69.5 \pm 2.4$  to  $1721.2 \pm 60.1 \text{ mBq m}^{-2} \text{ h}^{-1}$ , with a mean of  $391.8 \pm 13.7 \text{ mBq m}^{-2} \text{ h}^{-1}$ . The exhalation rates from the dry samples varied from  $123.7 \pm 4.3$  to  $2847.9 \pm 99.5 \text{ mBq m}^{-2} \text{ h}^{-1}$ , with a mean of  $728.7 \pm 25.5 \text{ mBq m}^{-2} \text{ h}^{-1}$ . For the porous powdery samples, the exhalation rates varied from  $433.0 \pm 15.1$  to  $889.9 \pm 31.1 \text{ mBq m}^{-2} \text{ h}^{-1}$  with a mean of  $623.8 \pm 21.8 \text{ mBq m}^{-2} \text{ h}^{-1}$ . The mass radon exhalation rates from the dried samples varied from  $7.7 \pm 0.3$  to  $85.0 \pm 3.0 \text{ mBq kg}^{-1} \text{ h}^{-1}$  with a mean of  $42.4 \pm 1.5 \text{ mBq kg}^{-1} \text{ h}^{-1}$  for the solid slabs. From  $16.5 \pm 0.6$  to  $33.7 \pm 1.2 \text{ mBq kg}^{-1} \text{ h}^{-1}$  with a mean of  $23.8 \pm 0.8 \text{ mBq kg}^{-1} \text{ h}^{-1}$  for the porous powdery materials.

To assess the contribution of building materials to indoor radon concentrations, a specific scenario was considered. In this case, a room with an air exchange rate of  $0.5 \text{ h}^{-1}$  was assumed as mentioned above. The radon concentration contribution to indoor radon in the room due to exhalation from the thermally untreated building materials was found to vary from 0.3 to  $6.9 \text{ Bq m}^{-3}$ . Samples CF-N and CS-N, ceramics made in France and Spain, respectively, showed the same minimum value, and the accumulated radon concentration per day in a kitchen or bathroom with its entire floor and walls decorated with ceramic tiles would be  $7.2 \text{ Bq m}^{-3}$ . Thus, on average, such ceramic tiles would contribute only slightly to the radon concentration in Libyan homes. On the other hand, granite imported from India would give an accumulated radon

concentration per day of  $165.6 \text{ Bq m}^{-3}$ . For the dried solid slab samples, radon concentrations varied from  $0.5$  to  $11.4 \text{ Bq m}^{-3}$  and for the porous powdery materials varied from  $1.7$  to  $3.6 \text{ Bq m}^{-3}$ . The BL-D sample, a Libyan brick, showed the lowest value, and the daily-accumulated radon concentration in a room with its entire walls built completely of such brick blocks was estimated as  $12 \text{ Bq m}^{-3}$ . On the other hand, the Italian brick BIt-D sample would contribute  $156 \text{ Bq m}^{-3}$ , an exhalation rate 13 times higher than that with the Libyan bricks. The Indian granite sample,  $G_n\text{In-D}$ , gave the highest value observed in this study. In a room such as a kitchen with its entire walls decorated with these granite slabs, a daily-accumulated radon concentration of  $273.6 \text{ Bq m}^{-3}$  would be contributed. It can be concluded that under normal ventilation in a kitchen or living room, granite countertops would make a large contribution to the radon concentration at the range of radon exhalation rates reported here, whereas the effect would be much less with ceramics and brick. Similar findings have been reported in the literature (Kitto and Green, 2005; Allen et al., 2009; Alvarcz et al., 2009; Kitto et al., 2009; Chen et al., 2010).

The radium equivalent content (REC) in the dry material samples was also found to vary from  $0.5 \pm 0.0$  to  $17.0 \pm 0.6 \text{ Bq kg}^{-1}$ , with a mean of  $5.7 \pm 0.2 \text{ Bq kg}^{-1}$ . And even for the wet material it was found to vary from  $0.6 \pm 0.0$  to  $28.5 \pm 1.0 \text{ Bq kg}^{-1}$  with mean value of  $4.7 \pm 0.2 \text{ Bq kg}^{-1}$ , whereas in the non-dried samples this parameter could not be determined. It can be seen from the results that the radon concentration varied significantly between the samples under the two conditions. The differences were due to the high moisture contents of the samples, which resulted in lower exhalation rates from the non-dried samples compared with the dry ones. In general, the radon concentrations of dry samples are higher than those of non-dried ones and lower than those of wet ones. Thus, radon exhalation rates of dry samples are found to be higher than those of non-dry samples and less than those of wet ones. It should be noted that some samples of granite and marble showed very considerable radon exhalation, as shown in Tables 5.1, 5.2 and 5.3. These materials could be significant sources of radon emanation, indicating that they contained high amounts of uranium or radium (Durrani and Ilic, 1997). Most of non-dried samples did not show significant radon exhalation,

suggesting that they contained little radium or uranium, as shown in Table 5.1. As previously mentioned, it is expected that exhalation of radon should depend on the uranium and radium concentrations in the samples, although it also depends on many other factors, such as moisture content, grain size, permeability, porosity, density, and texture.

Finally, one can see that it is necessary to assess the radon emanation potential from building materials to evaluate the radiation risk to the inhabitants. Thus, the assessment of the annual effective dose expected to be received by populations due to radon and its progenies was based on the calculations of radon exhalation rates. The respective minimum, maximum, and mean annual effective doses were found to be  $8.2 \pm 0.3$ ,  $203.0 \pm 7.1$ , and  $46.2 \pm 1.6 \mu\text{Sv y}^{-1}$  for the non-dried tiles,  $14.60 \pm 0.5$ ,  $345.2 \pm 12.1$ , and  $86.6 \pm 3.0 \mu\text{Sv y}^{-1}$  for the dried slabs, and  $6.5 \pm 0.2$ ,  $266.4 \pm 9.3$ , and  $64.4 \pm 2.3 \mu\text{Sv y}^{-1}$  for wet slab materials, and  $51.1 \pm 1.8$ ,  $104.9 \pm 3.7$ , and  $73.6 \pm 2.6 \mu\text{Sv y}^{-1}$  for the dried porous powdery material samples and finally  $128.8 \pm 4.5$ ,  $376.5 \pm 13.1$ , and  $222.4 \pm 7.8 \mu\text{Sv y}^{-1}$  for wet porous powdery material samples. The non-dried samples giving the lowest values of the annual effective dose were ceramic tiles imported from Spain and France, the lowest dose value from the dried and wet slab samples was attributed to the domestic Libyan brick and Egyptian brick, respectively. The minimum value from the dried powdery samples was from Egyptian gypsum and the wet powdery Libyan sand has the minimum value of annual effective dose. The highest dose from the non-dried and dried samples came from granite imported from India, and the second highest dose was from marble from Italy. It can be concluded that the annual effective doses from brick samples were almost equal to those from ceramics, and both of them are somewhat lower than those for other brick and ceramic samples reported in the literature (Moharram et al., 2012). Finally, the values of annual effective dose reported in the present study conform to the worldwide range, except for those of granite, marble imported from India and Italy, respectively and for those wet porous powdery attribute to black and white cement from Egypt.

**Table 5.1:** The values of radon gas measured from various non-dried solid tiles building materials by

Sample number	Sample code	Radon concentration (Bq.m <sup>-3</sup> )	Radon concentration contributing to the indoor radon (Bq.m <sup>-3</sup> )	Areal exhalation rates (mBq.m <sup>-2</sup> .h <sup>-1</sup> )	Annual effective dose (μSv.y <sup>-1</sup> )
1	CF-N	97.1 ± 3.4	0.3 ± 0.0	73.7 ± 2.6	8.7 ± 0.3
2	CE-N	420.1 ± 14.7	1.3 ± 0.1	319.2 ± 11.2	37.6 ± 1.3
3	CS-N	91.5 ± 3.2	0.3 ± 0.0	69.5 ± 2.4	8.2 ± 0.3
4	BIt-N	622.6 ± 21.8	1.9 ± 0.1	473.0 ± 16.5	55.8 ± 2.0
5	BE-N	402.1 ± 14.1	1.2 ± 0.0	305.5 ± 10.7	36.0 ± 1.3
6	G <sub>Y1</sub> E-N	381.3 ± 13.3	1.2 ± 0.0	289.7 ± 10.1	34.2 ± 1.2
7	G <sub>Y2</sub> E-N	481.2 ± 16.8	1.5 ± 0.1	365.5 ± 12.8	43.1 ± 1.5
8	G <sub>Y</sub> T-N	194.1 ± 6.8	0.6 ± 0.0	147.5 ± 5.2	17.4 ± 0.6
9	G <sub>Y</sub> L-N	320.3 ± 11.2	1.0 ± 0.0	243.3 ± 8.5	28.7 ± 1.0
10	M <sub>1</sub> It-N	241.3 ± 8.4	0.7 ± 0.0	183.3 ± 6.4	21.6 ± 0.8
11	MG <sub>r</sub> -N	249.60 ± 8.7	0.8 ± 0.0	189.7 ± 6.6	22.3 ± 0.8
12	MIn-N	187.2 ± 6.5	0.6 ± 0.0	142.3 ± 5.0	16.8 ± 0.6
13	M <sub>2</sub> It-N	1277.1 ± 44.6	3.9 ± 0.1	970.6 ± 33.9	114.5 ± 3.1
14	ML-N	203.8 ± 7.1	0.6 ± 0.0	154.9 ± 5.4	18.3 ± 0.6
15	ME-N	366.1 ± 12.8	1.1 ± 0.0	278.2 ± 9.7	32.8 ± 1.1
16	G <sub>n</sub> E-N	965.1 ± 33.7	2.9 ± 0.1	733.2 ± 25.6	86.5 ± 3.0
17	G <sub>n</sub> In-N	2265.7 ± 79.2	6.9 ± 0.2	1721.2 ± 60.1	203.0 ± 7.1
Max		2265.7 ± 79.2	6.9 ± 0.2	1721.2 ± 60.1	203.0 ± 7.1
Range		2174.2 ± 76.0	6.6 ± 0.2	1652.0 ± 57.7	194.8 ± 6.8
Mean		515.7 ± 18.0	1.5 ± 0.1	391.8 ± 13.7	46.2 ± 1.6
S.D		529.6 ± 18.5	1.6 ± 0.1	402.3 ± 14.1	47.4 ± 1.7

**Table 5.2:** The values of radon gas measured from various dried solid slabs building materials by using CR-39 SSNTDs.

Sample number	Sample code	Sample weight (kg x 10 <sup>-3</sup> )	Radon concentration (Bq.m <sup>-3</sup> )	Radon concentration contributing to the indoor radon (Bq.m <sup>-3</sup> )	Areal Exhalation rates (mBq.m <sup>-2</sup> .h <sup>-1</sup> )	Mass exhalation rates (mBq.kg <sup>-1</sup> .h <sup>-1</sup> )	Annual effective dose (μSv.y <sup>-1</sup> )	Radium content (Bq.kg <sup>-1</sup> )
1	CF-D	152.1	351.0 ± 12.3	3.3 ± 0.1	814.7 ± 28.5	42.7 ± 1.5	96.1 ± 3.4	2.4 ± 0.1
2	CE-D	130.2	270.3 ± 9.4	1.0 ± 0.0	248.2 ± 8.7	38.5 ± 1.3	29.3 ± 1.0	5.8 ± 0.2
3	CS-D	173.9	381.0 ± 13.31	1.3 ± 0.1	322.5 ± 11.3	40.6 ± 1.4	38.0 ± 1.3	6.7 ± 0.2
4	BIt-D	368.1	716.9 ± 25.5	6.5 ± 0.2	1625.1 ± 56.8	36.1 ± 1.2	191.6 ± 6.7	1.5 ± 0.1
5	BL-D	1094.7	442.9 ± 15.5	0.5 ± 0.0	123.7 ± 4.3	7.7 ± 0.3	14.6 ± 0.5	3.6 ± 0.1
6	G <sub>Y1</sub> E-D	100.1	275.9 ± 9.6	0.8 ± 0.0	206.0 ± 7.2	51.1 ± 1.8	24.3 ± 0.9	9.2 ± 0.3
7	G <sub>Y2</sub> E-D	178.0	810.8 ± 28.3	2.1 ± 0.1	529.8 ± 18.5	84.4 ± 3.0	62.5 ± 2.2	17.0 ± 0.6
8	M <sub>1</sub> It-D	688.3	441.1 ± 15.4	1.5 ± 0.1	361.2 ± 12.6	11.9 ± 0.4	42.6 ± 1.5	1.7 ± 0.1
9	MG <sub>r</sub> -D	360.2	600.6 ± 21.0	2.5 ± 0.1	617.2 ± 21.6	30.9 ± 1.1	72.8 ± 2.5	3.6 ± 0.1
10	MIn-D	498.3	264.6 ± 9.3	0.9 ± 0.0	212.7 ± 7.4	9.8 ± 0.3	25.1 ± 0.9	3.7 ± 0.1
11	M <sub>2</sub> It-D	317.4	1456.4 ± 50.9	6.1 ± 0.2	1528.7 ± 53.4	85.0 ± 3.0	180.3 ± 6.3	2.6 ± 0.1
12	ML-D	433.9	542.4 ± 19.0	2.0 ± 0.1	487.1 ± 17.0	23.2 ± 0.8	57.4 ± 2.0	1.4 ± 0.1
13	ME-D	311.3	349.1 ± 12.2	1.4 ± 0.1	352.7 ± 12.3	20.8 ± 0.7	41.6 ± 1.5	10.9 ± 0.4
14	G <sub>n</sub> E-D	350.9	1293.1 ± 45.2	2.6 ± 0.1	653.2 ± 22.8	68.3 ± 2.4	77.0 ± 2.7	15.2 ± 0.5
15	G <sub>n</sub> In-D	500.8	2283.3 ± 79.8	11.4 ± 0.4	2847.9 ± 99.5	84.5 ± 3.0	345.2 ± 12.1	10.8 ± 0.4
Min			264.6 ± 9.3	0.5 ± 0.0	123.7 ± 4.3	7.7 ± 0.3	14.60 ± 0.5	0.5 ± 0.0
Max			2283.3 ± 79.8	11.4 ± 0.4	2847.9 ± 99.5	85.0 ± 29.7	345.2 ± 12.1	17.0 ± 0.6
Range			2018.7 ± 70.5	10.9 ± 0.4	2724.2 ± 95.2	77.3 ± 2.7	330.6 ± 11.6	16.5 ± 0.6
Mean			698.6 ± 24.4	2.9 ± 0.1	728.7 ± 25.5	42.4 ± 1.5	86.6 ± 3.0	5.7 ± 0.2
S.D			548.4 ± 19.1	2.7 ± 0.1	713.5 ± 24.9	26.2 ± 0.9	86.0 ± 3.0	5.0 ± 0.2

**Table 5.3:** The values of radon gas measured from various dried porous powder building materials by using CR-39 SSNTDs.

Sample number	Sample code	Radon concentration (Bq.m <sup>-3</sup> )	Radon concentration contributing to indoor air (Bq.m <sup>-3</sup> )	Areal exhalation rate (mBq.m <sup>-2</sup> .h <sup>-1</sup> )	Mass exhalation rate (mBq.kg <sup>-1</sup> .h <sup>-1</sup> )	Annual effective dose (μSv.y <sup>-1</sup> )	Radium content (Bq.kg <sup>-1</sup> )
1	S <sub>a</sub> L	735.7 ± 25.7	2.9 ± 0.1	722.2 ± 25.2	27.4 ± 1.0	61.7 ± 2.2	3.8 ± 0.1
2	C <sub>B</sub> E	533.0 ± 18.6	2.1 ± 0.1	523.2 ± 25.2	20.4 ± 0.7	85.2 ± 3.0	2.2 ± 0.1
3	C <sub>w</sub> E	561.2 ± 19.6	2.2 ± 0.1	550.9 ± 19.2	20.8 ± 0.7	65 ± 2.3	2.3 ± 0.1
4	G <sub>y</sub> E	441.1 ± 15.4	1.7 ± 0.1	433.0 ± 15.1	16.5 ± 0.6	51.1 ± 1.8	1.9 ± 0.1
5	S <sub>o</sub> L	906.5 ± 31.7	3.6 ± 0.1	889.9 ± 31.1	33.7 ± 1.2	104.9 ± 3.7	4.4 ± 0.2
	Min	441.1 ± 3.5	1.7 ± 0.1	433.0 ± 15.1	16.5 ± 0.6	51.1 ± 1.8	1.9 ± 0.1
	Max	906.5 ± 31.7	3.6 ± 0.1	889.9 ± 31.1	33.7 ± 1.2	104.9 ± 3.7	4.4 ± 0.2
	Range	465.4 ± 16.3	1.9 ± 0.1	456.9 ± 16.0	17.2 ± 0.6	53.8 ± 1.9	2.5 ± 0.1
	Mean	635.5 ± 22.2	2.5 ± 0.1	623.8 ± 21.8	23.8 ± 0.8	73.6 ± 2.6	2.9 ± 0.1
	S.D	165.7 ± 5.8	0.7 ± 0.0	162.7 ± 5.7	6.1 ± 0.2	19.2 ± 0.7	1.0 ± 0.0

## **5.2 Effect of Moisture Content on Radon Exhalation Rates from Building Materials**

Some solid and porous powdery samples were exposed to water, as previously mentioned in Chapter 4, until they have reached a steady state; namely wet samples. The results of wet samples are summarized in Tables 5.4 and 5.5. These results show a significant increase in the exhalation rates of radon in these wet samples. This enhancement of the exhalation rates occur more noticeably in the powdery samples. Tables 5.4 and 5.5 show the results of different parameters; the radon activity concentration, the areal and mass exhalation rates, annual effective dose, the contribution of indoor radon and the radium content for all wet solid and powdery building materials under study. The minimum, maximum, range, arithmetic mean and the standard deviation for those materials are also shown in the same tables.

The radon activity concentrations from the wet solid samples were found to vary from  $90.1 \pm 3.2$  to  $1533.4 \pm 53.6$  Bq.m<sup>-3</sup> with mean and standard deviation of  $497.3 \pm 17.4$  and  $445.9 \pm 15.6$  Bq.m<sup>-3</sup>, respectively. In addition, the range of these values are  $1443.26 \pm 50.4$  Bq.m<sup>-3</sup>. The radon activity concentrations for wet porous powdery samples were found to vary from  $1113.0 \pm 38.9$  to  $3252.5 \pm 113.6$  Bq.m<sup>-3</sup> with mean value  $1921.1 \pm 67.1$  Bq.m<sup>-3</sup> and standard deviation  $841.3 \pm 29.4$  Bq.m<sup>-3</sup>, respectively. As well as the range of these values are  $2139.5 \pm 74.7$  Bq.m<sup>-3</sup>. It is clear to see the significant radon exhalation of these wet powder samples over solid ones.

It can be noted that the wet solid slab samples showing maximum radon concentration of  $1533.4 \pm 53.6$  Bq.m<sup>-3</sup> was from the CE-W sample, which was an Egyptian ceramic. A value of  $1277.8 \pm 44.6$  Bq.m<sup>-3</sup> was the second largest concentration value obtained from M<sub>2</sub>It-W, which was an Italian black marble. The third largest radon concentration value of  $257.6 \pm 43.9$  Bq.m<sup>-3</sup>, was from the sample G<sub>n</sub>In-W, an Indian granite. The results for the wet porous powdery samples showed maximum radon concentration of  $3252.5 \pm 113.6$  Bq.m<sup>-3</sup> from the sample C<sub>B</sub>E-W, an

Egyptian black cement. The next fourth largest radon concentration for wet porous powdery samples were  $2556.2 \pm 89.3$ ,  $1480.8 \pm 51.7$ ,  $1203.0 \pm 42.0$ ,  $1113.0 \pm 38.9$  Bq.m<sup>-3</sup> for samples C<sub>WE</sub>, G<sub>YE</sub>, S<sub>OL</sub>, and S<sub>aL</sub>, denoting Egyptian white cement, Egyptian white gypsum, Libyan soil, and Libyan sand, respectively.

The areal exhalation rates from wet solid slab building material samples ranged from  $55.3 \pm 1.9$  to  $2258.7 \pm 62.3$  mBq.m<sup>-2</sup>.h<sup>-1</sup> with mean value  $546.3 \pm 19.1$  mBq.m<sup>-2</sup>.h<sup>-1</sup> and standard deviation of  $635.4 \pm 22.2$  mBq.m<sup>-2</sup>.h<sup>-1</sup>, whereas the exhalation rates from the wetly porous powdery materials were found to vary from  $1092.5 \pm 38.2$  to  $3192.8 \pm 111.5$  mBq.m<sup>-2</sup>.h<sup>-1</sup> with mean value of  $1885.8 \pm 65.9$  Bq.m<sup>-2</sup>.h<sup>-1</sup> and standard deviation of  $825.8 \pm 28.9$  mBq.m<sup>-2</sup>.h<sup>-1</sup>. The mass exhalation rates from the wet solid slab building materials were found to vary from  $1.5 \pm 0.1$  to  $188.5 \pm 6.6$  mBq.kg<sup>-2</sup>.h<sup>-1</sup> with mean and standard deviation of  $33.4 \pm 1.2$  and  $45.4 \pm 1.6$  mBq.kg<sup>-2</sup>.h<sup>-1</sup>, respectively. The mass exhalation rates from wet porous powdery building materials varied from  $21.3 \pm 0.7$  to  $61.3 \pm 2.1$  mBq.kg<sup>-1</sup>.h<sup>-1</sup> with mean value of  $36.3 \pm 1.3$  mBq.kg<sup>-1</sup>.h<sup>-1</sup> and standard deviation of  $15.8 \pm 0.6$  mBq.kg<sup>-1</sup>.h<sup>-1</sup>.

In order to evaluate the contribution of wet building materials to indoor radon concentration, as previously mentioned it is assumed that the room has an air exchange of  $0.5 \text{ h}^{-1}$ . The exhaled radon from the wet solid slab materials contribute to the indoor radon with values varied from  $0.2 \pm 0.0$  to  $9.0 \pm 0.3$  Bq.m<sup>-3</sup>. The BL-W sample, a Libyan brick, showed the lowest value, and the daily accumulated radon concentration in a room with entire walls built entirely from such brick blocks was assessed to be equal to  $4.8 \text{ Bq.m}^{-3}$ . This result was then compared with the resulting value from the BI-W sample, an Italian brick, which gave a value of  $129.6 \text{ Bq.m}^{-3}$ , an exhalation rate 27 times higher than that with the Libyan bricks. On the other hand, the imported Indian granite, which showed the largest value would give an accumulated radon concentration per day of  $216 \text{ Bq.m}^{-3}$ . Samples CF-W, CS-W, and ML-W, which are French and Spanish ceramics, and Libyan marble, respectively, showed the same contribution of indoor radon, and the accumulated radon concentration per day in a house with its entire floors and walls decorated with ceramic and marble of  $21.6 \text{ Bq.m}^{-3}$ . As for the wet

porous powdery materials, radon concentration contributions varied from  $4.4 \pm 0.2$  to  $12.8 \pm 0.5 \text{ Bq.m}^{-3}$ .

The radium equivalent content (REC) of the wet solid slab material samples was found to vary from  $0.6 \pm 0.0$  to  $28.5 \pm 1.0 \text{ Bq.kg}^{-1}$ , with mean of  $4.7 \pm 0.2 \text{ Bq.kg}^{-1}$ , whereas in the wet porous powdery ones varied from  $2.9 \pm 0.1$  to  $6.6 \pm 0.2 \text{ Bq.kg}^{-1}$  with mean value of  $4.2 \pm 0.2 \text{ Bq.kg}^{-1}$ . From the current results, one can see that the radon exhalation significantly enhanced in samples with high moisture content. This enhancement was clearer in the wet porous powdery materials, and can be attributed to the fact that particles of powdery materials are crumbly (grains), the grains have many opening with large surface areas. The radon recoils out of the walls of these opening into the internal porous and then diffuses out to the intergranular pores. The elevation for wet porous powdery samples could be a result of the initiation of a thin film of water around grain surfaces. This thin film of water prevents radon atoms from embedding into grains, and therefore the number of radon atoms in the pore space increases (Sasaki et al., 2004). It can be noted from the above results that the high radium content was concentrated with high amount in the Egyptian ceramic followed by the Egyptian marble and granite and then Indian granite, while the French ceramic has the lowest REC value and therefore may considered to be a safe material for indoor decoration use. As well as Libyan brick, Italian marble contained small amount of radium. For the powdery building materials, Egyptian black cement contained the highest amount of radium followed by Egyptian white cement while the lowest content of the radium has been found to be from Libyan sand sample. All the results obtained have values within the international safe levels, except for those of Egyptian ceramic and all of powdery building materials.

To evaluate the radiation risk to the inhabitants from wet building materials, the assessment of the annual effective dose expected to be received by populations due to radon and its progenies was based on the calculations of radon exhalation rates. The respective minimum, maximum, and mean annual effective doses were found to be  $6.5 \pm 0.2$ ,  $266.4 \pm 9.3$ , and  $4.7 \pm 0.2 \mu\text{Sv.y}^{-1}$  for wet solid slabs,  $128.8 \pm 4.5$ ,  $376.5 \pm 13.1$ , and  $222.4 \pm 7.8 \mu\text{Sv.y}^{-1}$  for wet porous powdery samples. Solid slabs samples

contributed the lowest annual effective dose compared to those from porous powdery samples. On other hand, Egyptian brick slabs gave the minimum value of effective dose whereas, an Indian granite slabs give the highest value of effective dose. The highest effective dose from wet porous powdery samples came from Egyptian black cement and the lowest came from Libyan sand samples.

**Table 5.4** The values of radon gas measured from wetly solid building materials by using CR-39 SSNTDs.

Sample Code	Sample weight (kgx10 <sup>-3</sup> )	Radon concentration (Bq.m <sup>-3</sup> )	Radon concentration contribute to indoor radon (Bq.m <sup>-3</sup> )	Areal exhalation rates (mBq.m <sup>-2</sup> .h <sup>-1</sup> )	Mass exhalation rates (mBq.kg <sup>-1</sup> .h <sup>-1</sup> )	Annual effective dose (μSv.y <sup>-1</sup> )	Radium content (Bq.kg <sup>-1</sup> )
CF-W	161.3	95.7 ± 3.3	0.9 ± 0.0	222.2± 7.8	11.0 ± 0.4	26.2 ± 0.9	0.6 ± 0.0
CE-W	150.7	1533.35 ± 53.6	5.6 ± 0.2	1408.3± 49.2	188.5 ± 6.6	166.1 ± 5.8	28.5 ± 1.0
CS-W	186.2	252.8 ± 8.8	0.9 ± 0.0	214.0 ± 7.5	25.2 ± 0.9	25.2 ± 0.9	4.1 ± 0.1
BIt-W	401.1	593.1 ± 20.7	5.4 ± 0.2	1344.2 ± 47.0	27.4 ± 1.0	158.5 ± 5.5	1.1 ± 0.0
BL-W	1104.8	90.1 ± 3.2	0.2 ± 0.0	155.6 ± 5.4	1.5 ± 0.1	18.4 ± 0.6	0.7 ± 0.0
BE-W	1191.4	557.4 ± 19.5	0.6 ± 0.0	55.3 ± 1.9	8.7 ± 0.3	6.5 ± 0.2	1.4 ± 0.1
G <sub>Y1</sub> E-W	170.2	144.5 ± 5.1	0.4 ± 0.0	107.9 ± 3.8	15.7 ± 0.6	12.7 ± 0.4	2.8 ± 0.1
G <sub>Y2</sub> E-W	296.0	364.1 ± 12.7	1.0 ± 0.0	237.9 ± 8.3	22.8 ± 0.8	28.1 ± 1.0	4.6 ± 0.2
M <sub>1</sub> ItW	689.7	182.1 ± 6.4	0.6 ± 0.0	149.1 ± 5.2	4.9 ± 0.2	17.6 ± 0.6	0.7 ± 0.0
MG <sub>r</sub> -W	499.1	230.9 ± 8.1	1.0 ± 0.0	237.2 ± 8.3	8.6 ± 0.3	28.0 ± 0.98	1.0 ± 0.0
MIn-W	360.5	177.7 ± 6.2	0.6 ± 0.0	142.8 ± 5.0	9.1 ± 0.3	16.8 ± 0.6	1.7 ± 0.1
M <sub>2</sub> It-W	317.7	1277.8 ± 44.6	5.4 ± 0.2	1341.2 ± 46.9	74.5 ± 2.6	158.2 ± 5.5	2.2 ± 0.1
ML-W	442.8	259.0 ± 9.1	0.9 ± 0.0	232.6 ± 8.1	10.8 ± 0.4	27.4 ± 1.0	1.3 ± 0.1
ME-W	323.1	315.3 ± 11.0	1.3 ± 0.1	318.5 ± 11.1	18.1 ± 0.6	37.6 ± 1.3	9.6 ± 0.3
G <sub>n</sub> E-W	351.6	625.0 ± 21.8	1.3 ± 0.1	315.7 ± 11.0	32.9 ± 1.2	37.2 ± 1.3	9.1 ± 0.3
G <sub>n</sub> In-W	309.7	1257.6 ± 43.9	9.0 ± 0.3	2258.7 ± 78.9	75.2 ± 2.6	266.4 ± 9.3	5.0 ± 0.2
Min		90.1 ± 3.2	0.2 ± 0.0	55.3 ± 1.9	1.5 ± 0.1	6.5 ± 0.2	0.6 ± 0.0
Max		1533.4 ± 53.6	9.0 ± 0.3	2258.7 ± 62.3	188.5 ± 6.6	266.4 ± 9.3	28.5 ± 1.0
Range		1443.26 ± 50.4	7.0 ± 0.2	2203.4 ± 77	187.0 ± 6.5	259.8 ± 9.1	27.9 ± 1.0
Mean		497.3 ± 17.4	1.7 ± 0.1	546.3 ± 19.1	33.4 ± 1.2	64.4 ± 2.3	4.7 ± 0.2
S.D		445.9 ± 15.6	2.6 ± 0.1	635.4 ± 22.2	45.4 ± 1.6	74.9 ± 2.6	6.7 ± 0.2

**Table 5.5:** The values of radon gas measured from wetly porous powdery building materials by using CR-39 SSNTDs.

Sample Code	Radon Concentration (Bq.m <sup>-3</sup> )	Radon Concentration Contribute to Indoor Radon (Bq.m <sup>-3</sup> )	Areal Exhalation Rates (mBq.m <sup>-2</sup> .h <sup>-1</sup> )	Mass Exhalation Rates (mBq.kg <sup>-1</sup> .h <sup>-1</sup> )	Annual Affective Dose (μSv.y <sup>-1</sup> )	Radium Content (Bq.kg <sup>-1</sup> )
S <sub>a</sub> L	1113.0 ± 38.9	4.4 ± 0.2	1092.5 ± 38.2	21.3 ± 0.8	128.8 ± 4.5	3.0 ± 0.1
C <sub>B</sub> E	3252.5 ± 113.6	12.8 ± 0.5	3192.8 ± 111.5	61.3 ± 0.8	376.5 ± 13.2	6.6 ± 0.2
C <sub>w</sub> E	2556.2 ± 89.3	10.0 ± 0.4	2509.3 ± 87.7	48.1 ± 1.7	295.9 ± 10.3	5.2 ± 0.2
G <sub>y</sub> E	1480.8 ± 51.7	5.8 ± 0.2	1453.6 ± 50.8	28.0 ± 1.0	171.4 ± 6.0	3.2 ± 0.1
S <sub>0</sub> L	1203.0 ± 42.0	4.7 ± 0.2	1180.9 ± 41.3	22.7 ± 0.8	139.3 ± 4.9	2.9 ± 0.1
Min	1113.0 ± 38.9	4.4 ± 0.2	1092.5 ± 38.2	21.3 ± 0.7	128.8 ± 4.5	2.9 ± 0.1
Max	3252.5 ± 113.6	12.8 ± 0.5	3192.8 ± 111.5	61.3 ± 2.1	376.5 ± 13.1	6.6 ± 0.2
Range	2139.5 ± 74.7	8.4 ± 0.3	2100.3 ± 73.4	40.0 ± 1.4	247.7 ± 8.6	3.7 ± 0.1
Mean	1921.1 ± 67.1	7.5 ± 0.3	1885.8 ± 65.9	36.3 ± 1.3	222.4 ± 7.8	4.2 ± 0.2
S.D	841.3 ± 29.4	3.3 ± 0.1	825.8 ± 28.9	15.8 ± 0.6	97.4 ± 3.4	1.5 ± 0.1

### 5.3 Inter-Comparison between Radon Exhalation from Non-Dried, Dried and Wet Building Material Samples.

Table 5.6 shows the comparison between the minimum, maximum, range, mean and standard deviation of the areal and mass exhalation rates of radon from the non-dried, dried and wet solid building materials. Results of the calculated effective dose from these building material samples are also listed in the Table 5.6. It can be seen from this table: among of all material samples, the dried ones have the maximum areal exhalation of  $2847.9 \pm 99.5 \text{ mBq.m}^{-2}.\text{h}^{-1}$  with range, mean and standard deviation of  $2724.2 \pm 95.2$ ,  $728.7 \pm 25.5$  and  $713.5 \pm 24.9 \text{ mBq.m}^{-2}.\text{h}^{-1}$ , respectively. The second maximum value of areal exhalation belong to wetly samples and was found to be  $2258.7 \pm 62.3 \text{ mBq.m}^{-2}.\text{h}^{-1}$  with range, mean and standard deviation of  $2203.4 \pm 77$ ,  $546.3 \pm 19.1$  and  $635.4 \pm 22.2$ , respectively. The last maximum value of areal exhalation and that the lower among previous results belong to the non-dried sample materials were found to be  $1721.2 \pm 60.1 \text{ mBq.m}^{-2}.\text{h}^{-1}$  with range, mean and standard deviation of  $1652.0 \pm 57.7$ ,  $391.8 \pm 13.7$  and  $402.3 \pm 14.1 \text{ mBq.m}^{-2}.\text{h}^{-1}$ , respectively. The maximum values of mass exhalation rates gained from wetly samples were found to be  $188.5 \pm 6.6 \text{ mBq.kg}^{-1}.\text{h}^{-1}$  with range, mean and standard deviation of  $187.0 \pm 6.5$ ,  $33.4 \pm 1.2$  and  $45.4 \pm 1.6 \text{ mBq.kg}^{-1}.\text{h}^{-1}$ , respectively. The dried samples had the second maximum mass exhalation of  $85.0 \pm 29.7 \text{ mBq.kg}^{-1}.\text{h}^{-1}$  with range, mean and standard deviation of  $77.3 \pm 2.7$ ,  $42.4 \pm 1.5$  and  $26.2 \pm 0.9 \text{ mBq.kg}^{-1}.\text{h}^{-1}$ , respectively. For the doses received from the non-dried, dried and wetly building materials, the maximum value were found to be  $345.2 \pm 12.1 \mu\text{Sv.y}^{-1}$  coming from dried samples and the second maximum dose received from wetly samples was found to be  $266.4 \pm 9.3 \mu\text{Sv.y}^{-1}$ , the non-dried samples contribute to indoor with maximum annual dose of  $203.0 \pm 7.1 \mu\text{Sv.y}^{-1}$ .

Table 5.7 shows the maximum results of areal, mass exhalation rates and the annual effective dose from dried and wetly porous powdery building materials. The maximum values of the areal and mass exhalation rates came from wetly samples which were  $3192.8 \pm 111.5 \text{ mBq.m}^{-2}.\text{h}^{-1}$  and  $61.3 \pm 2.1 \text{ mBq.kg}^{-1}.\text{h}^{-1}$  with range, mean and standard deviation of  $2100.3 \pm 73.4$ ,  $1885.8 \pm 65.9$  and  $825.8 \pm 28.9 \text{ mBq.m}^{-2}.\text{h}^{-1}$ ;  $40.0 \pm 1.4$ ,  $36.3 \pm 1.3$  and  $15.8 \pm 0.6 \text{ mBq.kg}^{-1}.\text{h}^{-1}$ , respectively. The second maximum values

belonged to the dried samples which were  $889.9 \pm 31.1 \text{ mBq.m}^{-2}.\text{h}^{-1}$  and  $33.7 \pm 1.2 \text{ mBq.kg}^{-1}.\text{h}^{-1}$  with range, mean and standard deviation of  $456.9 \pm 16.0$ ,  $623.8 \pm 21.8$  and  $162.7 \pm 5.7 \text{ mBq.m}^{-2}.\text{h}^{-1}$ ;  $17.2 \pm 0.6$ ,  $23.8 \pm 0.8$  and  $6.1 \pm 0.2 \text{ mBq.kg}^{-1}.\text{h}^{-1}$ , respectively.

From the results obtained for the annual effective dos, of the porous powdery building materials the maximum value came from wetly ones which was  $376.5 \pm 13.1 \text{ }\mu\text{Sv.y}^{-1}$  with range, mean and standard deviation of  $247.7 \pm 8.6$ ,  $222.4 \pm 7.8$  and  $97.4 \pm 3.4 \text{ }\mu\text{Sv.y}^{-1}$ , respectively. The maximum annual effective dose received from the dried powdery samples was found to be  $104.9 \pm 3.7 \text{ }\mu\text{Sv.y}^{-1}$  with range, mean and standard deviation of  $53.8 \pm 1.9$ ,  $73.6 \pm 2.6$  and  $19.2 \pm 0.7 \text{ }\mu\text{Sv.y}^{-1}$ , respectively.

A comparison of all these results collectively, it can be concluded that for solid slabs and tile samples, the areal exhalation rates for dried samples found to have the maximum value followed by the wet samples and then lastly the non-dried samples. On other hand, the minimum values were found to be due to non-dried samples followed by dried samples and lastly by wet samples. As for mass exhalation rates, wet samples gave maximum values more than those of dried and non-dried samples, whereas, the minimum values came from dried and wet samples, respectively. Maximum Effective doses from solid samples belonged to the dried samples, whereas the minimum belonged to the non-dried samples. The variation of results between dried, non-dried and wet samples need more investigations and deeper studies. For the powdery samples, shown in table 5.5, it was found that all maximum values of areal, mass exhalation rates and annual effective doses were due to wet samples while the minimum values were due to dried ones. This result has confirmed that the water content has a strong effect on the radon exhalation from building materials

**Table 5.6** comparison between radon exhalation and annual effective dose from non-dried, dried and wet solid building materials.

	Areal Exhalation Rates (mBq.m <sup>-2</sup> .h <sup>-1</sup> )			Mass Exhalation Rates (mBq.kg <sup>-1</sup> .h <sup>-1</sup> )			Annual Effective Dose (μSv.y <sup>-1</sup> )		
	Normal	Dry	Wet	Normal	Dry	Wet	Normal	Dry	Wet
<b>Min</b>	69.5 ± 2.4	123.7 ± 4.3	55.3 ± 1.9	-	7.7 ± 0.3	1.5 ± 0.1	8.2 ± 0.3	14.60 ± 0.5	6.5 ± 0.2
<b>Max</b>	1721.2 ± 60.1	2847.9 ± 99.5	2258.7 ± 62.3	-	85.0 ± 29.7	188.5 ± 6.6	203.0 ± 7.1	345.2 ± 12.1	266.4 ± 9.3
<b>Range</b>	1652.0 ± 57.7	2724.2 ± 95.2	2203.4 ± 77	-	77.3 ± 2.7	187.0 ± 6.5	194.8 ± 6.8	330.6 ± 11.6	259.8 ± 9.1
<b>Mean</b>	391.8 ± 13.7	728.7 ± 25.5	546.3 ± 19.1	-	42.4 ± 1.5	33.4 ± 1.2	46.2 ± 1.6	86.6 ± 3.0	64.4 ± 2.3
<b>S.D</b>	402.3 ± 14.1	713.5 ± 24.9	635.4 ± 22.2	-	26.2 ± 0.9	45.4 ± 1.6	47.4 ± 1.7	86.0 ± 3.0	74.9 ± 2.6

**Table 5.7** comparison between dried and wetly powdery samples.

Sample code	Areal Exhalation Rates (mBq.m <sup>-2</sup> .h <sup>-1</sup> )		Mass Exhalation Rates (mBq.kg <sup>-1</sup> .h <sup>-1</sup> )		Annual Effective Dose (μSv.y <sup>-1</sup> )	
	Dry	Wet	Dry	Wet	Dry	Wet
	<b>S<sub>a</sub>L</b>	722.2 ± 25.2	1092.5 ± 38.2	27.4 ± 1.0	21.3 ± 0.8	61.7 ± 2.2
<b>C<sub>B</sub>E</b>	523.2 ± 25.2	3192.8 ± 111.5	20.4 ± 0.7	61.3 ± 0.8	85.2 ± 3.0	376.5 ± 13.2
<b>C<sub>W</sub>E</b>	550.9 ± 19.2	2509.3 ± 87.7	20.8 ± 0.7	48.1 ± 1.7	65 ± 2.3	295.9 ± 10.3
<b>G<sub>Y</sub>E</b>	433.0 ± 15.1	1453.6 ± 50.8	16.5 ± 0.6	28.0 ± 1.0	51.1 ± 1.8	171.4 ± 6.0
<b>S<sub>O</sub>L</b>	889.9 ± 31.1	1180.9 ± 41.3	33.7 ± 1.2	22.7 ± 0.8	104.9 ± 3.7	139.3 ± 4.9
<b>Min</b>	433.0 ± 15.1	1092.5 ± 38.2	16.5 ± 0.6	21.3 ± 0.7	51.1 ± 1.8	128.8 ± 4.5
<b>Max</b>	889.9 ± 31.1	3192.8 ± 111.5	33.7 ± 1.2	61.3 ± 2.1	104.9 ± 3.7	376.5 ± 13.1
<b>Range</b>	456.9 ± 16.0	2100.3 ± 73.4	17.2 ± 0.6	40.0 ± 1.4	53.8 ± 1.9	247.7 ± 8.6
<b>Mean</b>	623.8 ± 21.8	1885.8 ± 65.9	23.8 ± 0.8	36.3 ± 1.3	73.6 ± 2.6	222.4 ± 7.8
<b>S.D</b>	162.7 ± 5.7	825.8 ± 28.9	6.1 ± 0.2	15.8 ± 0.6	19.2 ± 0.7	97.4 ± 3.4

**Table 5.8** International results of Radon exhalation rates form some building materials.

<b>Building Material</b>	<b>Country</b>	<b>Areal Exhalation Rates (mBq.m<sup>-2</sup>.h<sup>-1</sup>)</b>	<b>References</b>
Ceramic	Present work Canada	69.5 – 319.2 ND – 100	Jing chen et al. 2010
Marble	Present work Canada Algeria Iran Egypt	212.7 – 1528.7 ND -25 35 – 66 0.17 – 384 333.3 – 125.8	Jing chen et al., 2010 Amrani and cherouti, 1999 Bavaregin et al , 2012 Walley El- Dine et al., 2001
Brick	Present work Pakistan Algeria	123.7 – 1625.1 245 – 365 78 – 117	Rahman et al. 2007 Amrani and cherouti, 1999
Granite	Present work Brazil Egypt Saudi Arabia	733.7 – 1721.2 470.8 – 212.5 287.1-1074.6 119-10,600	Jing chen et al. 2010 Walley El- Dine et al., 2001 Al-jarallah 2001
Gypsum	Present work Egypt Iran	107.9 - 529.8 19 - 74 ND- 610	Maged and Borham, 1997 Bavarnegin et al., 2012
Soil	Present work Pakistan China  Syria Morocco India	889.9 114 - 416 366 - 1668  72000 - 32400000 3 – 45 246.63 - 1100	Rahman et al. 2007 He xiaofeng and wangGuosheng 2011 Shweikhani and hushari, 2005 Oufni (2003) Singh et al. 2008
Cement	Present work Algeria Egypt Egypt	523.2 – 550.9 83 - 140 2549 - 7557 64 - 982	Amrani and cherouti, 1999 El – Bahi, 2004 Maged and Borham, 1997
Sand	Present work Lebanon Pakistan India	722.2 51.5 – 884.5 205 - 291 529,000 – 1685,000	Kobessi et at., 2008 Rahman et al. 2007 Rajesh et al , 2008

## **Chapter 6**

### **Conclusion**

The closed can technique that depends on the CR-39-NTDs detector for radon measurements, these techniques has many advantages such as a precise and simplicity as any other techniques, and it is more appropriate other than the electronic devices in the long term measurements of radon.

In spite of the concern about radon exhalation from building materials, this issue has not hitherto been paid a great deal of attention by the public community or scientific institutions in Libya. Thus, our motivation was to assess the radon exhalation rates for 39 samples of different building materials available on the Libyan market for constructive and decorative use, we had used the can technique. The current results are mostly within the worldwide range of values found in building materials, and this range is within the safe limits given in the report issued by the United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR, 2000). Two high values of radon concentration have been observed in the studied building material samples, from Indian granite and Italian marble. For this reason, these two materials are not favorable for decorative use and should be replaced by alternatives. It is necessary that radium contents and annual effective dose equivalents are within the safe limits given in the reports issued by the ICRP (1993), ICRP (1987), and UNSCEAR (2000). With poor ventilation and using materials with radon exhalation rates higher than the average, elevated radon levels could arise. In general, however, building materials used in home construction and decoration make no considerable contribution to indoor radon for a house with sufficient air exchange.

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