

RADON EXHALATION FROM BUILDING MATERIALS AND SOIL

Halina Pieńkowska

Chair of Agricultural Engineering and Natural Raw Materials
University of Warmia and Mazury in Olsztyn

Key words: radon, radon isotope, radionuclide, alpha particles, synergy effect, epidemiological research, relative risk.

Abstract

Radon is one of many natural sources of ionizing radiation on earth. The study presents radon properties and specifies its content in the natural environment and building materials. Trace-detectors were used to measure radon concentration. Measurement results for the city of Olsztyn are presented along with procedures aimed at reducing its concentration. Opinions on health hazards are also quoted.

EKSHALACJA RADONU Z MATERIAŁÓW BUDOWLANYCH I GRUNTU

Halina Pieńkowska

Katedra Inżynierii Rolniczej i Surowców Naturalnych
Uniwersytet Warmińsko-Mazurski w Olsztynie

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Streszczenie

Radon jest jednym z wielu naturalnych źródeł promieniowania jonizującego na Ziemi. Opisano właściwości radonu i określono jego zawartość w środowisku naturalnym i materiałach budowlanych. Zastosowano detektory śladowe do pomiaru jego stężenia. Podano wyniki pomiarów dla Olsztyna oraz tok postępowania w celu zredukowania stężenia tego karcynogenu. Przytoczono opinie na temat jego szkodliwości dla zdrowia.

Introduction

We have always been living in the presence of radon, but only thanks to the development of science are we aware of this. Deaths caused by lung illnesses among underground miners were already reported around 450 years ago. Upon describing mining techniques in the Rudawy region in 1556, dr. Agricola observed an unusually high mortality rate among young miners caused by respiratory illnesses (LINECKI 1997).

Cases of illness were so high at the beginning of the 16th century in and around Schneeberg, that the illness was named Schneeberger Lungenkrankheit and only in 1879 was it identified as lung cancer by Haerting and Hesse, which stated in their publication that around 75% of miners died as a result of this illness (ZAGÓRSKI 1997).

The opinion that the cause of illness was toxic metals in ore was maintained until the 1940s. It was research carried out by RAJEWSKY in 1944 which revealed that the most probable cause of illness was high radon concentration. Harley and Bale reached the conclusion that the dose from short-lived radon decay product is a source of hazard for the bronchus epithelium and lung parenchyma (LINECKI 1997).

The parameter defined by Holaday in 1957 as "Working Level", signifying the degree of exposure as the product of the average potential energy concentration of alpha particles in the air of the working environment during exposure time, is applied in order to assess the exposure dose (LINECKI 1997). In 1988, the International Agency for Research on Cancer (IARC 1988) recognized radon as a class 1 carcinogen.

Epidemiological research on the consequences of exposure to radon at the workplace are unusually difficult since it is influenced by too many interfering factors (smoking cigarettes, lifestyle, diet etc). This research has so far not generated a definite answer (ZAGÓRSKI 2000).

However, numerous radon concentration measurements carried out in residential homes have demonstrated that high concentration levels exist in a significant percentage of homes (LUBIN 1997). The authors of the publication stated that over a period of 25 years the relative risk amounted to 1,13¹ for concentration equal to 150 Bq/m³ ².

¹ Relative risk (odds ratio) – risk of occurrence of illness in group of exposed individuals (subject to the studied factor) compared to individuals not exposed.

² Bq reflects the substance activeness, in which one nucleus conversion (decay) takes place in 1 second. Radon concentration is measured in Bq/m³.

Radon in the natural environment

Among 27 radon isotopes (from Rn – 200 to Rn – 226) only Rn – 222 has significance with respect to contaminating the environment and due to its individual migrating possibilities. It belongs to the U-238 radioactive series forming through the decay of radium Ra-226. Radon's half-life $T^{1/2}$ amounts to 3.8 days. Radon's atomic radius amounts to $1.55 \cdot 10^{-10}$ m. The monoatomic structure of Radon facilitates its diffusion. It is a colorless, tasteless and odorless gas. Its density under normal conditions amounts to $9.82 \text{ kg} \cdot \text{m}^{-3}$ and is 7.6 fold heavier than air. It is easily soluble in water and even more so in alcohol. Its solubility decreases as the temperature rises. Since it is less soluble in the aqueous solutions of certain salts, sea water contains less radon than water in rivers and lakes (MILESZKIEWICZ 1978). As a radioactive isotope, radon is further subject to disintegration into four short-lived products (Po-218, Pb-214, Bi-214, Po-214) out of which two (Po-218 and Po-214) are emitters of alpha particles. Thoron (Rn-220) may also contaminate the environment due to alpha particles emitted by derivative radionuclides resulting from it.

The concentration of radon in the environment depends on the geological and geochemical composition (SOLECKI 1997). The formation of radon atoms depends on the content of radionuclides directly preceding the Ra-226 radioactive series. The radon atoms of interest to us are those released into the hydrosphere and atmosphere, while this accounts for a small percentage of formed radon.

Radon trapped in mineral structures is indifferent to us since it can travel 20–70 nm within the mineral structure. In most cases, radium – radon's parent element – is present in minerals.

The radon present in the hydrosphere and atmosphere was originally formed in the lithosphere, out of which it is ejected. The ejecting process from minerals is facilitated by the fact that upon radium disintegration, alpha particles are emitted throwing out radon with energy greater than that of chemical bond energy. Radon's atom thrown outside the mineral grain enters pore space. This is the first stage of migration in the environment. Its fate is decided by the state of the pores. The presence of radon in soil air enables the detection of porous fault zones. The migration of radon in highly porous soil is accelerated by convection caused by temperature fluctuation (CIEŻKOWSKI 1992).

According to ZAGÓRSKI (1997), uranium is distributed in the earth's crust in the amount of around 2 ppm and for this reason we register radon almost everywhere, but in various concentration levels. The stress forming in the earth's crust influence radon migration, whose concentration fluctuation is a signal of foreseen earthquakes. An earthquake occurred in northern Poland on 21 September 2004. Such crust movements may change radon concentration, which is established as natural background in northern Poland.

Rocks with pores filled with water lengthens radon migration, since radon dissolves in the flowing water (RAMA, MOORE 1984). The concentration of radon in water bodies depends on geological and atmospheric conditions. Radon migration under surface water conditions may reach over 1000 m. In continental surface water, radon is present due to emanation from the bottom, where the radium origin is located. Radon in sea water originates from dissolved radium. The average radon concentration in sea water amounts to 7 Bq/100 kg.

Radon concentration in atmospheric air is significantly lower than in soil air and in caves of natural or artificial origin. Radon concentration in the air at the Jachymov mine (Czech Republic) fluctuates between 12 and 330 kBq/m³ while in Colorado and Utah mines between 94 and 190 kBq/m³ (WHITTLESTONE et al. 1992).

Radon concentration in atmospheric air depends on the geographical location, e.g. in Great Britain it amounts to around 2.6 Bq/m³, while in Poland to 4.4 Bq/m³ on average. The difference is caused by the impact of oceanic atmosphere, which is clearly observed in Alaska and Hawaii (SOLECKI 1997).

The emanation and migration mechanism of radon into the atmosphere depends on various factors.

The following regularities are observed:

- I Radon concentration decrease in sea region.
- II Radon concentration decrease with altitude.
- III Seasonal fluctuation of radon concentration.

Radon concentration in human environment forms as a result of emanation from the lithosphere and building materials of mineral origin. An important source of radon migration is underground water, and its local degassing intensifies its concentration in the air. What must be noticed is that the concentration of radium, from which radon is formed, is variable, as are rock emanation cofactors, which results in variable radon concentration levels in soil. Radon concentration in atmospheric air, as well as in our homes, depends on this.

Radon from building materials

According to current observations and research, the source of radon in residential buildings are rocks and soil that form the foundation. According to UNSCEAR (1988), this accounts for 70–77.9% of radon emanation. Building materials (e.g. concrete) believed to be one of the safest materials also increase radon concentration since they are sometimes produced from waste aggregate or industrial waste.

They generate two types of hazards:

- I Gamma radiation caused by the cumulative concentration of elements from the uranium, thoron and K-40 potassium.
- II Alpha radiation, which is associated with the liberation of radon and thoron decay products.

According to norms of most European countries, the highest concentration for newly constructed buildings amounts to 200 Bq/m³, while for existing buildings from 200 to 800 Bq/m³ (for example in Canada). The permissible maximum concentration in Poland in existing buildings amounts to 400 Bq/m³, and 200 Bq/m³ in newly constructed ones (*Directive and amendment* 1995, RAWIŃSKI 1997).

Radon concentration measurements are carried out in order to locate areas of higher risk, verify the effectiveness of applied reduction methods as well as satisfy the curiosity of interested residents. The measurement results inform us whether old buildings must be modernized through sealing and new ventilation systems as well as of what materials the new ones are built.

The dependence in accordance with instruction ITB no. 234/95 was applied to determine the radon exhalation value from building materials:

$$e_{Rn-222} = S_{Ra} \cdot \eta \cdot \rho \cdot \lambda_{Rn-222} \cdot d/2,$$

where:

- e_{Rn-222} – radon exhalation in Bq/m²·h,
- S_{Ra} – radium concentration in material, in Bq/kg,
- η – radon emanation cofactors depending on the type of building material in %,
- ρ – density of building material in kg/m³,
- λ_{Rn-222} – radon decay constant³ $\lambda = 7,56 \cdot 10^{-3} \text{ h}^{-1}$,
- d – thickness of structural partition in meters.

For example, the exhalation value for concrete and natural sandstone amounts to around 12.78 Bq/m²·h, despite the fact that the concentration of radium in concrete is three times greater than in sandstone:

$$S_{Ra} (\text{concrete}) = 153.77 \text{ Bq/kg} \quad S_{Ra} (\text{sandstone}) = 52 \text{ Bq/kg}.$$

An eightfold radon exhalation difference is observed at similar radium concentration levels in ceramic brick and keremsite-slag concrete. It amounts to 0.95 Bq/m²·h for the brick and 7.98 Bq/m²·h for the concrete.

The density of these materials as well as the emanation cofactor (η w %), depend on their internal structure, impact the exhalation value of radon from building materials. For natural sandstone $\eta = 10\%$, for granite 8.3%, for lime-sand brick 9%, for concrete 2.2% and for natural gypsum 4%.

³ Decay Constant $\lambda = \ln 2 / T_{1/2}$.

Radon and health

Radon and its derivatives enter the human organism through the inhalation of air. The air includes dust measuring from around 0.01 μm to the first decimal place of mm.

Short-lived radon decay products in the air are deposited on the dust. While breathing, the dust particles, along with radon decay products deposited on them, enter the lungs. The air with radon is exhaled but the dust particles with radon decay products remain in the lungs. Dust with a diameter over 1 μm sets down in the nose and larynx. While dust with a diameter measuring from 0.1 to 1 μm is deposited in the bronchi.

They remain longer in the lungs with decay products than its half-life (around 50 minutes) and are then naturally expelled from the body (NIEWIADOMSKI 1994). It is the decay of radon derivatives that is the source of the dose received through the respiratory system, since radon increases the dose insignificantly seeing as it is exhaled with air. It does not attach itself to walls in gas form, so the possibility of decay in the lungs is then small. The inhalation of radon derivatives leads to heterogeneous irradiation of the respiratory system. The highest doses are taken by basic epithelium cells of the upper bronchus in the trachea and bronchi area of the lungs.

No severe and immediate effects are observed as a result of inhaling air with radon decay products, since the effects are distant in time and as such difficult to study. The distant effects are effectively overshadowed by other factors such as smoking cigarettes, diet, lifestyle etc. It seems that epidemiological research could help in assessing the situation. Such synthesis was carried out by an international team under the supervision of LUBIN in the USA (1994).

A team of underground miners, who spent time working in air highly concentrated with radon, was selected from a mine of uranium and other materials, as well as a group of residents subject to low concentrations of radon in air (statistically insignificant).

This team attempted to establish a link between lung cancer and radon. The fact that the results were not statistically significant testifies to the difficulty of such a study, but the final conclusions are interesting.

It was found that around 40% of deaths caused by lung cancer may be associated with exposure to radon derivative decay. Epidemiological research taking into account other factors than radon has shown that radon is a trival of tobacco smoking. According to Lublin, the impact of radon on US residents is as follows: radon caused 11% of deaths among non-smokers and 30% of deaths caused by lung cancer among smokers. A synergy effect was observed at small radon doses, i.e. the mutual action and intensification of two harmful factors.

Lublin worked out a risk model according to which reducing radon concentrations in US residential homes to the level recommended by the EPA (Environmental Protection Agency) could lower mortality by 2 to 4%.

The remote in time harmful effects of radon have resulted in the use of radon water in the form of baths, mud baths etc.

At inhalatoriums, the sick inhale dry air enriched with radon, while water with radon is sprayed from inhalers. Radon therapy is applied by balneology specialists. Worked out and closed uranium mines are used for this purpose, where the concentration of radon -222 amounts to a few hundred Bq/m³.

The largest radon therapy center in the world is the Bad Gastein health resort located in Austria, in the area of Central Alps, 1280 m over sea level.

Radon inhalatorium is located in the side drift of the worked out gold mine. The main geological structures in this region are grejsy and mica-schist.

The Rn-222 concentration in the air in side drifts reaches 170 kBq/m³, with air humidity reaching 80%. According to the doctors of the health resort, the medicinal effect is based on the mutual action of three factors, i.e. air with radon, air temperature (37–42°C) and high humidity. In Bad Gastein, a lot of radon is diffused into the atmosphere. Radon concentration in residential buildings amounts from 74 to 558 Bq/m³, while outside the building from 19 to 130 Bq/m³ (PACHOCKI 1997).

There are three known health resorts in Poland that apply radon therapy. These include: Łądek Zdrój, Świeradów and Cieplice. In 1975 – 1990, the Cieplice health resort applied radon inhalation on a larger scale using for this purpose the closed side drift of the Kowary uranium ore mine. Around a 300,000 patients were subject to such treatment, out of which in around 6,000 patients the treatment was repeated.

According to the latest research, radon may induce lung tumors (LUBIN 1994), but balneology specialists are not taking these effects into consideration. They assume that radon decay products are a small radiation load for short-term patients. However, the risk does exist and requires research on the inhalatorium's personnel, but also residents living near the health resort which are continuously subject to radon in the air and water.

The Americans closed the radon-active waters just in case. Water treatment also does not provide clear and fast therapeutic results, same as radon does not provide immediate harmful effects (ZAGÓRSKI 2000). For this reason, radiation effects may be examined only as subsequent cause and effect changes in a time function (PRZYBYTNIK 1997) – Figure 1.

Deoxyribonucleic acids (DNA) fulfill the role of conveying hereditary traits to derivative cells and account for cell genetic material.

From chemistry's perspective, they are high-molecular compounds as polymers. Double-strand breaks are damages that are subject to enzymatic

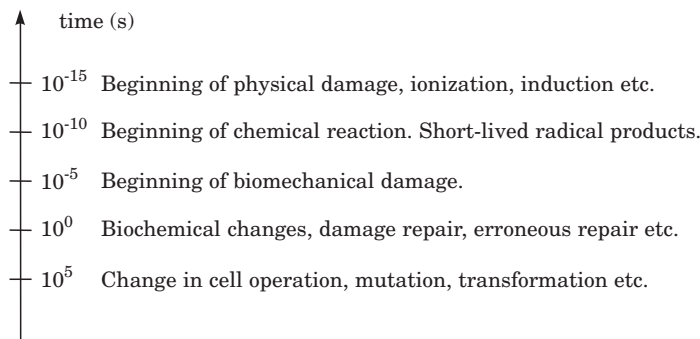


Fig. 1. Diagram sequence of times (PRZYBYTNIK 1997)

repairs. The ineffectiveness of repairs results from multiple damage to the DNA strand at the molecular level. This is no easy task since the subject of research is a chemically highly complex compound, whose properties depend on small changes in its surroundings.

Research on DNA damage is highly interdisciplinary research, starting from polymer radiation chemistry, ending with molecular biology and biochemistry (ZAGÓRSKI 1997).

Radon measuring methods

The basic characteristic of a measuring technique should be its sensitivity. The techniques are divided into passive and active. Passive techniques are usually much less expensive and more convenient since the device operates without any power supply or servicing personnel. A classic measuring method is the use of Lussac's chambers. A sample of the tested air is captured in a standard cylindrical glass container, whose walls are covered with scintillating material, after which a photomultiplier counts the scintillations in a laboratory.

Another passive device is a container with active carbon, used to collect radon over a period from one day to one week. It is a tightly sealed container 10 cm in diameter filled with active carbon. After opening, the radon present in the air is deposited in carbon pores via natural diffusion. Following the exposure, the container is closed and radon activity is assessed in a laboratory, which is proportional to the average concentration of Rn-222 in the air (GORZKOWSKI 1994). Such a container may be used several times after it is heated for around 3–4 hours in 150°C, in order to remove deposited radon atoms.

Another passive detector is a trace-detector. The phenomena applied here is that of solid body structure disarrangement by alpha particles, since

other types of radiation (beta and gamma) do not generate such damage. Special plastics are currently used (TUSZYŃSKI et al. 1998). A 4 cm² plate is placed inside a container of a given volume and covered by film through which radon atoms cross. The plate is then etched in 6M NaOH solution at temperatures of 20–90°C for a few hours, while the visible traces are then counted under a microscope.

Radon concentration measurements were performed in Olsztyn with the use of a "TASTRAK" trace detector. Since radon concentration is subject to significant fluctuations over day, week and annual periods (NIEWIADOMSKI 1994), measurements are carried out by two methods. The short-term method on a wide scale as well as the method lasting over a period of several months at selected locations.

Small containers with active carbon are used in short-term measurements, while trace detectors are usually used for long-term measurements. Measurements of radon concentration in the air are carried out by Radiation Protection Laboratories and Centers which are located in larger district towns in Poland, e.g. Warsaw or Łódź.

Radon concentration in selected residential buildings in the city of Olsztyn

Radon concentration tests in Olsztyn were carried out with the use of a "TASTRAK" trace detector, which was calibrated at H.H. Wills Physics Laboratory, Bristol, G.B.

Following the set six-day period, the plates were etched in 6M NaOH for 7 hours at temperatures of 20–90°C, after which the number of pits (traces) were counted under a microscope according to the model provided by the above-mentioned laboratory.

$$\text{Radon concentration (Bq/m}^3\text{)} = (5,3 \pm 0,5) \cdot N/D,$$

where:

N – number of traces per cm² of the plate,

D – number of days.

The measurements were taken in the city of Olsztyn. Most measurements were taken in homes located between Lake Długie and Lake Krzywe. The results are presented in Table 1.

According to the generated results, radon concentration in certain cellar rooms exceeded the Polish Standard several times and amounted to a maximum of around 689 Bq/m³, while the minimum radon concentration in these rooms amounted to around 11 Bq/m³. The average concentration level in buildings over five stories high amounted to around 100 Bq/m³, with a minimum value of around 27 Bq/m³; radon concentration was higher in the kit-

Table 1

Radon concentration in residential buildings

Concentration range R_n (Bq/m ³)	Number of homes	Concentration range R_n (Bq/m ³)	Number of homes	Concentration range R_n (Bq/m ³)	Number of homes
0-10	1	141-150	5	281-290	1
11-20	6	151-160	2	291-300	1
21-30	2	161-170	1	301-310	3
31-40	14	171-180	2	311-320	0
41-50	7	181-190	5	321-330	0
51-60	10	191-200	6	331-340	1
61-70	5	201-210	3	341-350	3
71-80	8	211-220	0	...	0
81-90	3	221-230	2	551-560	1
91-100	2	231-240	2	...	0
101-110	4	241-250	4	631-640	1
111-120	3	251-260	2	...	0
121-130	5	261-270	1	681-690	1
131-140	3	271-280	2		

chen and bathroom. Despite the fact that water and natural gas are secondary sources of radon emission (UNSCEAR 1985, 1988), under certain conditions they may increase its concentration. These conditions include: quality of utilities and method of their use. Aside chemical contamination, natural gas may exhibit a higher radioactive level when heavy radionuclides of geological origin are present in it (PAWUŁA 1996). While radon is easily dissolved in water (MIKLASZEWICZ 1978) which may result in increased radon concentration in bathrooms. Radon concentration was lower in rooms that were often aired. The location of apartments within buildings also impacted radon concentration and decreased the higher the apartment was located.

Methods of reducing radon concentration in homes

The precise identification of locations of radon requires many dosimetric tests. Once radon's paths are identified, they can be simply closed (ENNEMOSER 1997, PACHOCKI 1997).

Different methods for lowering radon concentration are proposed for homes already built and different for homes that are in the design stage. Many

countries have standard procedures that architects and developers must follow. Radon-tight membranes or floor ventilation are most often used in already built homes (NIEWIADOMSKI 1994).

Sealing gaps and cracks in the floor using a polythene membrane does not radically reduce the amount of radon.

Raising the pressure in the residential section prevents the suction of radon out of the foundation. This type of effect is achieved by placing a ventilator in the attic, which forces air in from top to bottom increasing pressure and diminishing the so-called "chimney effect".

This method lowers radon concentration in the air by threefold. Floor ventilation is applied where the so-called suspended floor is used, under which natural or forced ventilation may be implemented. Respective ventilators of appropriate power lower radon levels by up to fourfold.

The most effective method of lowering radon concentration in the air is the so-called floor depression method (radon trap). It is a hole in the ground under the house or in the cellar with an installed ventilator sucking air out of the house, resulting in negative pressure in the hole. Radon concentration levels may be lowered by as much as eightfold by using this method. Such traps were used in Great Britain.

The listed methods are costly and require energy and material use. Their use depends on the construction of the house. For this reason, one of the best and most effective methods of protecting against radon is the frequent airing of homes, including cellars. In order to lower radon values, the home must be sealed.

It is best to use silicon to fill gaps surrounding the installation, small cracks in the concrete floor. A new layer separating from the foundation should be applied in a cellar that does not have a tight floor. The effectiveness of applying the listed methods depends on the route taken by radon to enter the home, the home's construction etc.

In many countries, after assessing that radon concentration levels were exceeded, actions are recommended to reduce these levels. However, these actions are not compulsory. There are countries where financial support is provided upon implementing the listed methods. These countries are wealthy and have radon intervention levels established, while actions aimed at lowering radon levels are carried out immediately. Based on measurements carried out so far in Poland, there is no premise to set intervention levels (NIEWIADOMSKI 1994).

Summary

Until recently there was no radon problem in homes. Radon was only an issue addressed in mines. It was only the synergy effect (i.e. the mutual

effect of two harmful factors, e.g. low radon concentration assisted by cigarette smoke, dust etc.) that led to establishing radon levels in homes.

Precisely identifying locations where radon is present in cellars, water pipes etc. enables eliminating radon traveling paths. Frequent airing is the simplest method for removing radon from homes.

The most practical methods of measuring radon concentration are passive methods, e.g. "TASTRAK" plates or others.

Electric meters used in mining provide the value of momentary concentration, which may result in reaching false conclusions by either exaggerating or underestimating the value of radon concentration.

Epidemiological research on large human populations are difficult to carry out since the population's response to ionizing radiation is complicated by external factors as well as differences in individual reactions.

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