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Measurement of the Indoor Radon Concentration from Different Building Materials Using CR-39 Nuclear Track Detectors and the Associated Annual Effective Dose Determination

Abstract: This work presents experimental results concerning the radon concentration from different building materials used for construction of houses in the municipality of Bihac. The passive technique using nuclear track detectors C-39 was used for a period for three months. The highest and lowest radon concentration was found in concrete brick buildings 280±5 Bqm^{-3} and in stone buildings 122±1 Bqm^{-3}. It depends on the radioactive content of the materials, emanation coefficient and diffusion coefficient of radon in that material, porosity and density of the material. The mean annual effective dose was 3.26 mSv/y. The results obtained also give a correlation between indoor radon levels and the associated level of risk.

Keywords: radon, building materials, nuclear track detectors, passive techniques, annual effective dose

1. INTRODUCTION

Radon (^{222}Rn) is a decay product of radium (^{222}Ra) which in turn is a decay product from the natural decay series of ^{238}U in most soils. Since radon is produced by decay of radium isotopes in rocks and minerals, there are three natural isotopes of the element -^{222}Rn, ^{220}Rn i ^{219}Rn resulting in the so-called radioactive decay series (decay chain), uranium series ^{238}U, thorium series ^{232}Th and actinium series ^{235}U, respectively. In its decay, radon is transformed into stable lead ^{206}Pb over its short-lived daughters, emitting 5 alpha particles of energies of 7.7 MeV beta radiation energy of 2.8 MeV gamma-ray photon energy up to 2.4 MeV. Short-lived radon daughters, ^{218}Po, ^{214}Pb, ^{214}B and ^{214}P represent a greater threat to human health than radon, as they can easily bind to aerosols, and then with air enter the respiratory tract where they accumulate and ionized radiosensitive lung tissue.

In each row of the disintegration an isotope of radon is the result from of alpha emissions of radium, so it can be rigorously concluded that the production of radon is in proportion to the presence of radium in the soil. As the parent nuclides of radium are found in the condensed solid materials, radon is produced one locations with uranium, however, there are exceptions. So, there are places where radium was dispersed, transferred and re-deposited elsewhere, with the result that the radon generated far away from the parent uranium deposits [1].

Measurements of radon play a key role in monitoring the safety of people, both in their homes and in mines. Radon is a widespread danger and problems with radon are heightened where it is naturally concentrated in certain geologic materials or sociological intensified undesirable practices in the construction of houses. The most important fact when it comes to radon and it is dangerous to human health is that it is responsible for over 50% of natural radioactivity received by a person in a year.

Radioactive atoms which are decomposition products of radon, are collected on the aerosol particles, dust and moisture in the room air. If there is also tobacco smoke, radon can be increased almost twice. Most of the radiation dose a person gets through radionuclides enters the body through inhaled air. Lungs are exposed to the biggest impact of the decay products of radon. Experts from the International Agency for Cancer Research conclusively determined that radon and products and its decay products are carcinogenic for humans, but an increased risk of developing lung cancer related to exposure to radon in residential areas are not convincing, although a number of epidemiological studies showed an increase in the relative risk for the occurrence of lung cancer in conjunction with a high level of radon. The hypothesis of carcinogenic effects of radon on humans is not new, but this problem has attracted attention when it was found that the concentration of radon in the premises varies more than a hundred times and depends on the place where the building is situated and the type of construction materials used in the construction. Radon enters the body system during inhalation, which results in an increase in the exposure dose that can result in the development of lung cancer [2]. Building materials like sand, soil, bricks, and gravel aggregates contain a trace amount of ^{226}Ra which generates radon. However, only a fraction of radon from radioactive material is able to escape to the atmosphere so that it can be transported to an indoor environment through diffusion and convective flow.

The concentration of radon varies only according to the geological nature of the soil. It primarily emanates from granite and volcanic soil layers. After leaving the ground it diffuses through the air and water or dissolves. Its concentration in free air is very small. Radon is a natural part of our environment. Its concentration depends not only on geological and climatic factors but also of architectural and building solutions and building our way of life. It is noted that its concentrations are higher in the winter then in the summer, and there are also variations in the level during the day, which obviously depends on opening doors and windows.

The quantity of radon in soil and rocks, therefore, depends on the content of uranium and thorium in them. Uranium is often found in the bedrock as an ingredient in very fine particles of minerals coffinite [$U(SiO_4)_{1-x}(OH)_{4X}$] and uraninite (U_3O_8) or absorbed into the crystal structure of the mineral zircon ($ZrSiO_4$), apatite [$Ca_5(F,C1)(PO_4)_3$], titanite ($CaTiSiO_5$) and allanite [$H(Ca,Fe)(Al,Fe)_3Si_3O_{13}$]. Uranium-rich mineral grains are released during uranium erosion. Soft grains of minerals in contact with water create particles of clay while firmer grains, such as zircon, monazite and titanite, link the sandy parts which results in a huge variety in the concentration of uranium and radium when creating many types of soil.

Indoor radon concentration mainly depends on the number of atoms of radium, and the possibility of penetration of radon into the interior of buildings. Consequently, the soil and rock beneath or in the environment of residential buildings are a major source of internal radon. The second most important source of indoor radon is building materials.

The presence of radium in building materials depends on the type. Since these two elements are not in a natural balance to their chemical properties and are different, they need to be discussed separately. The amount of the radium produced from a γ-emission of radon daughters (^{214}Pb i ^{214}Bi) after it was sealed and kept long enough ensures a balance between radium and radon. The amount of time needed for this is determined by the γ-decay of its first short-lived daughters. So, we see that for the determination of the amount of uranium and radium in the soil and building materialist is necessary to include γ-spectrometric methods.

The concentration of radon will usually be largest in the basement and ground floor. Concentration on the first floor will be halved, while the concentration of radon above the first floor is usually negligible. The concentration of radon within residential buildings is typically two to three times higher than in the open.

Typical natural materials carrying the uranium are certain granites, but it is always possible to find stone deposits of various types of rich uranium, which is used locally for construction. Certain types of soil, for example, some types of clay, which is used as a construction material may have an increased content of uranium. Typical artificial materials are different types of concrete or lightweight concrete composed of ash, aluminum slate or uranium-rich materials derived from mining activities.

Radon concentration from building materials depends on factors such as: the radium concentration, the fraction of radon produced which is released from the materials grain to its interstitial space, the porosity of the material, the surface preparation and building material covering [3].

This work studies the radon concentration in some building material in the municipality of Bihac using passive techniques type CR-39 nuclear track detectors [4].

2. MATERIALS AND METHODS

2.1. Study area

The study area is located in the municipality of Bihac, Una-Sana Canton, Bosnia and Herzegovina. Detailed diagnostic measurements were carried out in 9 selected buildings made of different building materials.

2.2. Methodology

The method of measuring indoor radon concentration using nuclear track detectors CR-39 is a passive method for radon concentration measuring. Passive devices are represented by nuclear track detectors based on CR-39 chips, provided by Radosys Ltd. Budapest, namely RSK, used to measure radon concentration [5].

Detectors were placed onto present furniture within 25 to 50 cm from the wall. The detectors are generally placed on the first floor of the house in order to avoid as much of the possible impact of radon from the soil. Detectors are placed in rooms that are rarely used in the household and are not often ventilated. The nuclear track detector consists of a cylindrical plastic bottle provided with an appropriate cap. The 1 cm^2 CR-39 chip is attached to the bottom of the plastic bottle with an adhesive. The radon present indoor is able to enter the bottle through the space created between the cap and the bottle body. Thus inside, the alpha particles from radon decay hit the CR-39 chip living black tracks. After the exposure, all devices were collected and brought to the laboratory where they were chemically etched in 25% NaOH (sodium hydroxide solution) for 4.5 hours at 90°C in RadoBath device.

Track-counting was performed by using an automatic counting device Radosys system 2003. This device includes an optical microscope reconnected to a CCD (Charge-Coupled Device) camera controlled by a personal computer and RADOSYS software. The track densities found on the films were automatically converted into radon concentrations (Bq m^{-3}) by this software.

2.3. Annual Effective Dose

For the calculating of the annual effective dose for the population, the equation below is used ICRP, 1993 [6]. To calculate annual effective dose it is necessary to first calculate the annual absorbed dose. The annual absorbed dose, is: $D_{Rn} = C_{Rn} \cdot D \cdot H \cdot F \cdot T$ *[mSv/y]* (1) where,

C_{Rn} is the measured ^{222}Rn concentration (in Bq/m^3),

D is the dose conversion factor ($9.0 \cdot 10^{-6}$ mSv/h per Bq/m^3),

H is the indoor occupancy factor (0.4),

T is the ^{222}Rn equilibrium factor indoors (0.4), and

F is the indoor occupancy time 24·365 = 8760 h/y.

Now to calculate the annual effective dose, one has to apply tissue and radiation weighting factors according to ICRP, 1991 [7]. The radiation weighting factor for alpha particles is 20 as recommended by ICRP, 1991. With the effective dose, a tissue weighting factor is applied. According to ICRP 1991, the tissue weighting factor for lung is 0.12. The annual effective dose is then calculated according to the equation below: $H_E = D_{Rn} \cdot W_R \cdot W_I \cdot$ *[mSv/y]* (2) where,

D_{Rn} is annual absorbed dose,

W_R is radiation weighting factor for alpha particles, 20

W_R is tissue weighting factor for the lung, 0.12

3. RESULTS AND DISCUSSION

The concentrations of naturally occurring radionuclides in building materials have been reported in several publications which can vary according to the type and origin of the building material. The most common structural building materials used in this area are concrete, red bricks, concrete bricks, siporex bricks, adobe, gypsum and cement. The results of the radon concentration of these building materials are shown in Table 1.

Table 1. Radon concentration for different types of building materials

Building materials	Radon concentration (Bq/m3)
Concrete	108±2
Red bricks	133±3
Concrete bricks	140±3
Adobe	92±2
Adobe and mortar	89±2
Siporex bricks	104±2
Gypsum board	111±2
Red bricks and gypsum board	132±3
Stone building	61±1

The radon concentration varies from 140±3 Bqm^{-3} in the concrete bricks building to 61±1 Bqm_{-3} in the stone building. We noticed from the data given in Table1 that radon alpha-activities per unit of volume in concrete bricks samples were higher than those of the other samples. This is due to the fact that these samples contain more uranium than the others. This difference also depends on emanation factor and diffusion coefficient of radon in that material, porosity and density of the material. Figure 1 shows the variation ratio of building materials to indoor radon concentration.

The annual effective dose from the corresponding measured radon concentration in the different houses has been calculated using Equation 2, which varies from (1.85 to 4.24) mSv/y with a mean value 3.26 mSv/y which is above the recommended ICRP intervention level of (3-10) mSv/y (ICRP, 1993).

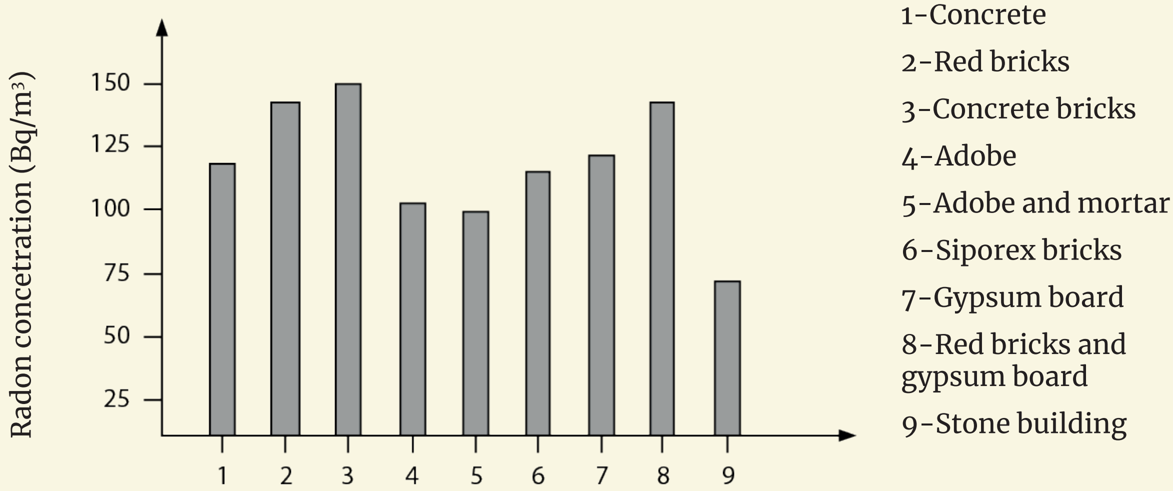

Fig.1 Radon concentration in different building materials

Table 2 gives a summary of the annual effective dose measured in nine different houses built from diverse building materials in Bihac for the present study.

Table 2. Annual effective dose from different building materials

Building materials	Annual effective dose (mSV/y)
Concrete	3.28
Red bricks	4.01
Concrete bricks	4.24
Adobe	2.78
Adobe and mortar	2.69
Siporex bricks	3.17
Gypsum board	3.35
Red bricks and gypsum board	3.99
Stone building	1.85

The highest value was observed in the house made of concrete bricks with an indoor radon concentration of 140±3 Bq/m^3 and an annual effective dose of 4.24 mSv/y. The high radon concentration level in concrete bricks is probably caused because red bricks contain more uranium than others, and higher emanation coefficient and diffusion coefficient of radon in that material. The lowest value was found in the house made of stone with an indoor radon concentration of 61±1 Bq/m^3 and an annual effective dose of 1.85 mSv/y, which is probably caused by poor porosity and high density stone blocks from which the house was built.

The results of the study indicate that radon concentrations in most of the houses investigated were slightly high. Although all of the indoor radon concentration is within the ICRP action level (200 Bq/m^3). One reason for the increased concentration of indoor radon is the increased concentration of uranium in the form of building materials and a highly porous material. Another factor explaining the high levels of radon in these compartments are the poor ventilation, which is required when placing the detector due to reduction of the impact of radon from the soil.

4. CONCLUSIONS

The highest concentration of radon was detected in concrete bricks and the lowest concentration of radon was detected in a stone building.

It is recommended to use stone for the construction of houses and it is also not recommended to use concrete bricks extensively. The results obtained from the current study show that the radium content in concrete bricks is higher than other building materials, this depends on the radioactive content of the materials, emanation factor and diffusion coefficient of radon in that material, porosity and density of the material. Figure 1 shows that the concrete, red bricks, concreate bricks, siporex bricks, gypsum board, red bricks and gypsum board have a radon concentration higher than the global permissibility limit of exposure to radon for the population according to (200 Bqm^{-3}) ICRP, 1993.

The mean annual effective dose in the studied area were 3.26 mSv/y. All studied houses recorded concentrations below the recommended action

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Mjerenje koncentracije radona u zatvorenom prostoru iz različitih građevinskih materijala korištenjem CR-39 nuklearnih detektora i odgovarajuće godišnje određivanje efektivne doze

U ovome radu su prikazani eksperimentalni rezultati koji se odnose na koncentraciju radona iz različitih građevinskih materijala koji se koriste za izgradnju kuća u općini Bihać. Korištena je pasivna metoda za mjerenje koncentracije unutrašnjeg radona pomoću nuklearnih tragova detektora C-39 u trajanju od tri mjeseca. Najviša i najniža koncentracija radona pronađena je u kućama izgrađenim od betonskih blokova 280±5 Bqm^{-3} i kućama izgrađenim od kamenih blokova 122±1 Bqm^{-3}. To ovisi o sadržaju radioaktivnog materijala, koeficijenta emanacije i koeficijenta difuzije radona u tom materijalu, poroznosti i gustoći materijala. Srednja godišnja efektivna doza iznosi 3.26 mSv/y. Dobiveni rezultati također daju vezu između koncentracije unutrašnjeg radona i pridruženog nivoa rizika.

Ključne riječi: radon, građevinski materijal, Bihać, koeficijent emanacije, difuzija