MEASUREMENT OF RADON ACTIVITY CONCENTRATION IN BUILDING MATERIALS USED IN BOSNIA AND HERCEGOVINA

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Abstract: Man is continuously exposed to ionizing radiation because of the presence of naturally occurring radioactive materials (NORM) in the environment. Various technological processes of processing and using of materials that contain natural radionuclides generate materials of enhanced natural radioactivity (TENORM). The largest contribution to irradiance with natural sources of ionizing radiation is the exposure of the population to indoor radon. This gas originates from the radioactive decay of $^{226}$Ra and $^{224}$Ra that are present in the soil under houses and building materials. Depending on the type of building materials, indoor exposure to radon at dwellings and workplaces can be over a thousand times greater than in outdoor space. In Bosnia and Herzegovina, no valid and comprehensive radiological studies on the building materials have been performed that would guarantee for their dosimetric safety use for installation in residential and industrial buildings, highways, as well as their application for other purposes. The quantification of the radon levels that comes from building materials is a necessary and very important part of the global protection of the population from ionizing radiation. This paper presents the first results of a study on the radon activity concentrations in building materials used in Bosnia and Herzegovina. Measurements were performed with a professional Alpha GUARD system. The mean values of the activity concentration of the exhaled radon of investigated building materials varied from 10 Bq m$^{-3}$ to 101 Bq m$^{-3}$, radon exhalation rate values range from 77.0 mBq m$^{-2}$ h$^{-1}$ to 777.7 mBq m$^{-2}$ h$^{-1}$. Gamma dose rate was in the range 57–112 nSv h$^{-1}$.

Keywords: building materials, radon activity concentration, radon exhalation rate, gamma dose rate.

1. INTRODUCTION

Living beings on the earth have always been exposed to ionizing radiation from natural sources and for the last 100 years from artificial sources as well. Over 80% of global exposure comes from natural sources of radiation, and about 20% comes from artificial sources, mainly from radiation used in medicine. Natural radioactive sources contribute with 2.4 mSv to the average annual effective dose, and it varies from 1 mSv to more than 10 mSv, depending on where people live. The average annual effective dose from inhalation of isotope $^{220}$Rn and its decay products is about 70 μSv and about 1.2 mSv from inhalation of $^{222}$Rn and its decay products [1].

The main components of natural radiation are cosmic rays, terrestrial gamma rays, ingestion and inhalation of natural radionuclides. Under normal conditions, the terrestrial radionuclides are responsible for most of human exposure to natural radiation. These primarily include the radionuclides that are members of three natural radioactive decay series: uranium radium ($^{238}$U), uranium actinium ($^{235}$U) and thorium series ($^{232}$Th). Generally, the radionuclides at the beginning of these radioactive decay chains are called primordial or primary natural radionuclides. All members of these series are genetically related and formed by disintegration of the first member of the series, which is explained by the law of radioactive decay.

It is known that the major contribution to natural radiation exposure comes from radioactive gas radon. Radon and its decay products are constantly present in the environment due to the radioactive decay of isotopes of radium, natural minerals contained in rocks and soil. The largest amount of radon in the closed

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structures comes from ground, through direct contact of the structure with soil below the basement or foundation, but also from the used building materials. Problems associated with radioactive gas radon are very complex and being investigated in a various field of scientific research. Radon is especially important for human health, and this is a motive, among others, why there is a permanent interest in it. The study of the biological effects of radon on humans is a very demanding scientific problem. The human body is a very complex system with many organs of different chemical properties, functions and radiosensitivity, so the reliable assessment of risk exposure and the development of adequate protection from radon require a multidisciplinary scientific approach to these problems [2].

1.1 Radioactivity of building materials

The largest contribution to the external irradiation of natural sources of ionizing radiation is indoor exposure to $\gamma$-radiation, which originates from the disintegration of radioactive elements present in all terrestrial formations [3]. Raw materials of natural origin containing different amounts of primordial radionuclide are natural radioactive materials (NORM - naturally occurring radioactive materials). These materials are generated during the geological processes in the formation of minerals and rocks from which these materials are produced. People spend about 80% of their time indoors. Therefore, a large contribution to the dose from the natural sources of ionizing radiation is from the natural radionuclides that are present in the building materials and soil [4,5,6]. Gas radon has a dominant role in this contribution.

Table 1 shows typical values of the content of natural radionuclides in certain types of construction materials, which are most commonly used in different countries of the world.

| Type of construction material       | $^{226}\text{Ra}$ (Bq kg$^{-1}$) | $^{232}\text{Th}$ (Bq kg$^{-1}$) | $^{40}\text{K}$ (Bq kg$^{-1}$) |
|-----------------------------------|----------------------------------|----------------------------------|-------------------------------|
| Structural materials              |                                  |                                  |                               |
| Concrete                          | 18-67                            | 3-43                             | 16-1100                       |
| Brick                             | 7-140                            | 8-127                            | 227-1140                      |
| Gas-concrete                      | 10-60                            | 6-66                             | 51-870                        |
| Gypsum                            | 1-67                             | 0.5-190                          | 22-804                        |
| Cement                            | 13-107                           | 7-62                             | 48-564                        |
| Covering materials                |                                  |                                  |                               |
| Construction ceramics             | 25-193                           | 29-66                            | 320-1049                      |
| Ceramic tiles                     | 33-61                            | 45-66                            | 476-788                       |
| Marble                            | 1-63                             | 0.4-142                          | 9-986                         |

The concentration of radionuclides in rocks, soils and sand depends on the local geology of each region in the world. Various technological and physical processing methods, as well as certain uses of materials that contain natural radionuclides (NORM) create materials of increased natural radioactivity (TENORM - Technologically Enhanced Naturally Occurring Radioactive Materials). These secondary products, mainly waste products, are the result of industrial activities related to: mineral processing, exploitation and chemical processing of ores, production of phosphates, use of fossil fuels to produce electricity, exploitation of oil and gas, etc. Because of the suitable exploitation, the reduction of production costs, as well as the structure which enables obtaining the final products with the appropriate characteristics, TENORM materials are used primarily in the construction industry but also in other industries. This is one way of conserving natural resources as well as addressing the problem of industrial waste disposal. However, with those applications of TENORM materials, the level of population exposure to ionizing radiation is increased.

Depending on the type of building materials, indoor exposure to radiation in dwellings and workplaces can be up to a thousand times greater than in outdoor space mainly because of the exhalation of radioactive gas radon from building materials and poor ventilation. The measurement of the radiation levels that comes from building materials presents a necessary and very important part of the global protection against ionizing radiation. In order to minimize radiation risk, a
system of protection against ionizing radiation which is related to building materials has been recommended by leading international organizations, and has been implemented in the legislation of many countries.

1.2. Radon exhalation from building materials

226\(^{226}\text{Ra}\) atoms are generated from 226\(^{226}\text{Ra}\) alpha decay in solid building material grains when radon atoms move to the pore void (this is called emanation). Radon diffuses through the system pore, whereby a certain amount of radon atoms is emitted from the surface of the material (this is called exhalation). The significant characteristics of building materials radiation are emanation power and exhalation power (or radon flux). The emanation power is the activity of radon obtained from a material divided by the mass of that material in a unit of time. The ratio of the radon amount that enters the pore spaces and the total amount of produced radon is called the emanation coefficient. The water (moisture) content in the pore space of the material can significantly affect the radon emanation coefficient for materials [8]. In addition to this, the radon emanation coefficient is also affected by other material characteristics, such as the size of the material granules, the distribution of 226\(^{226}\text{Ra}\) in the granules, the microscopic structure of the material, the part of the space occupied by the cavities-pores where radon is generated (material porosity).

The radon exhalation rate of any sample \(E_{Rn}\) is defined as the flux of radon released from the surface of a material. Radon exhalation rate depends upon a number of parameters that are stochastic and independent, such as the radioactive disintegration of 226\(^{226}\text{Ra}\) which produce radon, the direction of recoil of radon in the grain, the water content in the pore space and diffusion of atoms in the pore space [9]. The radon exhalation rate of any sample \(E_{Rn}\), is calculated from the formula [10, 11]:

\[
E_{Rn} = \frac{C_{Rn} V \lambda}{S}
\]  

(1)

where \(E_{Rn}\) is the radon exhalation rate in Bqm\(^{-2}\)s\(^{-1}\), \(C_{Rn}\) is radon concentration (Bq m\(^{-3}\)), \(V\) is the volume of accumulation box in m\(^3\) (corrected for sample volume), \(\lambda\) is the decay constant of radon (2.1 \(\times\) 10\(^{-6}\) s\(^{-1}\)) and \(S\) is the surface area covered with exhalation box.

2. MATERIALS AND METHODS

In this study the radon activity concentration, radon exhalation rate and gamma dose rate from samples of various building materials and raw materials for their production, such as: brick, clay, trowel, glue for ceramics, sand, diabase, dolomite, limestone was measured. The most investigated building materials originate from Bosnia and Herzegovina. But, some of these materials, like tiles and ceramic tiles, which are used in Bosnia and Herzegovina, originate from other countries.

Measurements of the radon activity concentration were performed by means of AlphaGUARD PQ2000 PRO measuring system (Genitron Instruments, Germany). The main part of the device, AlphaGUARD PQ2000 PRO, is the ionization chamber of the active volume of 0.56 dm\(^3\). The device can operate in two operating modes: with diffusion and with pump. Radon penetrates into the inside of appliance-ionisation chamber through a glass fiber filter, where \(\alpha\)-particles resulting from radioactive decay ionize the air within. Electrical pulses generated in the chamber are correspondent to the number of atoms of decaying radon. The measurement results can be read on the display. Measuring range of this device for radon activity concentration is from 2 to 2 \(\times\) 10\(^6\) Bqm\(^{-3}\), while the temperature range is from -10 to 50 °C. The calibration error of this measurement system for 222\(^{222}\text{Rn}\) is 3%. The system besides radon simultaneously measures air temperature, atmospheric pressure and relative humidity that are meteorological parameters deep correlated with the distribution of radon.

For measurement of the radon exhalation rate \(E_{Rn}\) (Bqm\(^{-2}\)s\(^{-1}\)) from building materials the Exhalation Box in combination with the AlphaGUARD PQ2000PRO and AlphaPUMP electronic pumping unit was used (Figure 1).

Figure 1. Experimental set-up for radon exhalation rate measurements (adopted from Manual AlphaGuard, Genitron Instruments) [12]

All the investigated samples of building materials were washed to remove any impurities.
After washing, the samples of building materials were dried at 105 °C, until inhomogeneities of the samples are eliminated. All the samples were crushed to a fine powder form. All these methods of the samples preparation of low activity and tracer samples are based on the concentration of radionuclides in the samples. This is obtained by reducing the larger sample volumes to less, achieved by grinding. Except the milling and drying, the samples were not exposed to any other physical or chemical treatment. After homogenization, the samples were weighed and placed in a Radon-Box. During the measurement the Radon box was hermetically sealed with adhesive tape to the base. In a closed circulating system gas radon is pumped with the AlphaPUMP at a flow rate of 0.3 dm$^3$ min$^{-1}$. Every sample was measured approximately of two hours at one minute intervals. The contribution of thoron ($^{220}$Rn, half-life 55 s) was negligible at this flow rate.

3. RESULTS AND DISCUSSION

Table 2 shows the radon activity concentration, radon exhalation rate and gamma dose rate in investigated building materials.

| No. | Type of material | Commercial name or producer | $C_{Rn}$ (Bq m$^{-3}$) | $E_{Rn}$ (mBq m$^{-2}$ h$^{-1}$) | $H_{\gamma}$ (nSv h$^{-1}$) |
|-----|------------------|-----------------------------|------------------------|-------------------------------|--------------------------|
| 1   | Glue for ceramics | EurofixFx, Gradačac         | 12                     | 92.4                          | 100                      |
| 2   | Glue for ceramics | Eurofix K, Gradačac         | 13                     | 100.1                         | 104                      |
| 3   | Skim coat mass   | Domal, Gradačac             | 14                     | 107.8                         | 101                      |
| 4   | Cement           | White cement, Spain         | 10                     | 77.0                          | 105                      |
| 5   | Cement           | White cement, CRH Slovakia  | 22                     | 169.4                         | 108                      |
| 6   | Cement           | Berement, Hungary           | 18                     | 138.6                         | 104                      |
| 7   | Cement           | Larfage BFC, Beočin, Serbia | 15                     | 115.5                         | 100                      |
| 8   | Cement           | Holcim, Koromačno, Croatian | 22                     | 169.4                         | 108                      |
| 9   | Diabase          | Ribnica                     | 14                     | 107.8                         | 103                      |
| 10  | Dolomite         | Kalvariija, Vitez           | 29                     | 223.3                         | 112                      |
| 11  | Block            | Shamotte block, IGM Busovača| 14                     | 107.8                         | 110                      |
| 12  | Block            | Black block, Cažin          | 56                     | 431.2                         | 106                      |
| 13  | Limestone        | MG „MND“, Mrkonjić Grad     | 22                     | 169.4                         | 104                      |
| 14  | Limestone        | Teko Mining, Lapišnica     | 26                     | 200.2                         | 108                      |
| 15  | Limestone        | Vjenac, Lukavac             | 14                     | 107.8                         | 109                      |
| 16  | Limestone        | Mrvelji, Posušje            | 33                     | 254.1                         | 111                      |
| 17  | Limestone        | Sokolica, Zavidoviči       | 19                     | 146.3                         | 108                      |
| 18  | Limestone        | Zaklopača, Milići           | 52                     | 400.4                         | 111                      |
| 19  | Limestone        | Podorašac, Konjic          | 12                     | 92.4                          | 109                      |
| 20  | Limestone        | Perutina, Čapljina          | 15                     | 115.5                         | 68                       |
| 21  | Limestone        | Joštanica, Divić           | 42                     | 323.4                         | 69                       |
| 22  | Limestone        | Miljevina, Mostar          | 32                     | 246.4                         | 70                       |
| 23  | Gravel           | Jasenica, Mostar           | 10                     | 77.0                          | 104                      |
| 24  | Gabbro           | Granit, Jablanica          | 36                     | 277.2                         | 65                       |
| 25  | Clay             | Clay TOS Sarajevo          | 101                    | 777.7                         | 59                       |
| 26  | Brick            | Full brick, IGM            | 63                     | 485.1                         | 63                       |
| 27  | Tile             | Tondach -73, Constant Plus | 51                     | 392.7                         | 58                       |
| 28  | Tile             | Trend, Polet               | 55                     | 423.5                         | 62                       |
| 29  | Tile             | Oktavijan, Nexegrupa, Croatian | 33                 | 254.1                         | 62                       |
| 30  | Tile             | Concrete, Bramac           | 39                     | 300.3                         | 62                       |
| 31  | Tile             | Cezar, Nexegrupa, Croatian | 26                     | 200.2                         | 64                       |
| 32  | Ceramic tiles    | Idea Ceramica, Italy       | 44                     | 338.8                         | 58                       |
| 33  | Ceramic tiles    | Undefasa, Castellón, Spain | 40                     | 308.0                         | 68                       |
| 34  | Ceramic tiles    | Keramika Kanjiža-1, Serbia | 43                     | 331.1                         | 66                       |
| 35  | Ceramic tiles    | Keramika Kanjiža-2, Serbia | 29                     | 223.3                         | 57                       |
The results in the Table 2 show that the mean value of the activity concentration of radon exhaled of investigated building materials was in the range of 10 Bq m$^{-3}$ to 101 Bq m$^{-3}$. The maximum value of the radon activity concentration during the measurements of the investigated building materials was in the range from below 16 Bq m$^{-3}$ to 132 Bq m$^{-3}$.

The radon exhalation rate for the investigated building materials samples was in the range from 77.0 mBq m$^{-2}$h$^{-1}$ to 777.7 mBq m$^{-2}$h$^{-1}$. The minimum value was found for the sample No. 4, white cement, Spain, 77.0 mBq m$^{-2}$h$^{-1}$, whereas the maximum value was for sample No. 25, clay TOS Sarajevo, 777.7 mBq m$^{-2}$h$^{-1}$. Gamma dose rate was in the range 57–112 nSv h$^{-1}$.

The activity concentration of radon exhaled of investigated ten limestone samples was in the range of 12 Bq m$^{-3}$ to 52 Bq m$^{-3}$, which confirms the fact that the carbonate rocks of limestone, dolomite and rock salt have the lowest radionuclide content [13], because the primary carbonate mineral in limestone - calcite, has a low content of radioactive elements. Increased content in this building material can be found due to the presence of non-carbonate accessory minerals clay or calcium phosphate. That refers to the sample 18, which has a higher value than others of the same type of construction material. The activity concentration of radon exhaled for coating materials, such as building ceramics and ceramic tiles, was in the range of 29 Bq m$^{-3}$ to 44 Bq m$^{-3}$. The basic mass for the production of construction ceramics is made of brick and ceramic clay, and in addition contains the most common minerals kaolinite, quartz, calcite, pyrite, feldspar and muscovite, which are of interest from the radiological point of view.

The highest value of concentration of radioactive elements was found in granite, a magmatic material composed of quartz, feldspar, mica and other petrogenic minerals. The famous Jablanica building stone, Gabbro (jablanite), that is magmatic originating, is characterized by a low concentration of radon activity, 36 Bq m$^{-3}$. All investigated types of cement samples are characterized by low radon activity concentration values, which range from 10 Bq m$^{-3}$ to 22 Bq m$^{-3}$.

Among other investigated samples of building materials, the highest value of the activity concentration of radon exhaled was found in the clay sample from the TOS Sarajevo Clay, 101 Bq m$^{-3}$. Because of its compact structure, the clay retained and trapped emanated radon gas. By grinding clay, radon is released and contributes to increased levels of radiation.

The risk of lung cancer from domestic exposure to radon and its daughters can be estimated directly from the effective dose equivalents. The contribution of indoor radon concentration from building material samples can be assessed with different scenarios, according to the way of using those materials. If assumed that radon is exhaling from walls, ceiling and floor of the room where same building material is used for an entire room, the contribution of indoor radon concentration from building material samples can be calculated with the following expression [14, 15]:

$$C_{Rn} = \frac{E_{Rn} \cdot S_r}{V_r \cdot \lambda_v}$$  \hspace{1cm} (2)

where $C_{Rn}$ is the radon concentration (Bq m$^{-3}$), $E_{Rn}$ is the radon exhalation rate (Bq m$^{-2}$h$^{-1}$), $V_r$ is the room volume (m$^3$), $\lambda_v$ is the air exchange rate (h$^{-1}$). In these calculations, the maximum radon concentration from the building materials was assessed by assuming the room to be a cavity with $E_{Rn}$, $V_r = 2.0$ m$^{-1}$ and an air exchange rate of 0.5 h$^{-1}$. 

![Figure 2. Activity concentration of exhaled radon from limestone (five minute intervals of time)- Jostanica, Divić](image)

This figure shows the activity concentration of exhaled radon from limestone (five minute intervals of time) from Jostanica, Divić.
If a room has its entire floor decorated with investigated building material and 10% of the room volume is occupied by furniture, the radon concentration, due to radon exhalation from the floor can be determined by

\[
C_{Rn} = \frac{E_{Rn}}{0.9(\lambda_0 + \lambda_v)H}
\]

where \(H\) is the room height in meters (2.5 m), \(\lambda_0\) is the radon decay constant (0.181 d\(^{-1}\)) and \(\lambda_v\) the air removal rate due to ventilation [10].

The annual effective dose equivalent, \(E\) in WLM \(\gamma\)\(^{-1}\), is then related to the average radon concentration by the following expression:

\[
E = \frac{8760 \cdot 0.8 \cdot F \cdot C_{Rn}}{170 - 3700}
\]

where 0.8 is the fraction of time spent indoors, \(F\) is the equilibrium factor (0.42), 8760 is the number of hours per year and 170 is the number of hours per working month. For radon exposure, the effective dose equivalents were estimated by using a conversion factor of 6.3 mSv/WLM \(^{-1}\) [15, 16].

Based on the contribution of indoor radon concentration from building materials the annual effective dose equivalents were estimated. Minimum, maximum, and average annual effective doses were found to be 1.69, 14.25 and 5.68 \(\mu\mathrm{Sv}\), respectively, for materials that are used for covering floor of the room. When bulding materials are used for covering entire room then minimum, maximum, and average annual effective doses were found to be 8.65, 54.48 and 22.09 \(\mu\mathrm{Sv}\), respectively.

The values of annual effective doses reported in the present study are comparable or below then results from other studies [15, 17]. Results obtained from the current study show that the average annual effective doses are below the world average value of 1.1 mSv [3].

4. CONCLUSIONS

In this study the measurements of radon activity concentration, radon exhalation rate, gamma dose rate and annual effective dose in commonly used building materials from the territory of Bosnia and Herzegovina, as well as imported construction materials are presented. Most of the investigated building materials contain small amounts of NORM as a consequence of the geological variation of the site. The measurements of the radon activity concentration of exhaled radon from building materials are the first step when taking measures to protect the health of the population. The results showed that the glue for cement (No.4) and gravel (No.23) contain the lowest value of radon concentrations, while the other samples, except the clay (No.25) contain approximately a similar value. The radon concentration levels and the annual effective doses are, on average, below the action level recommended by the ICRP [16, 18].

5. ACKNOWLEDGMENTS

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