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 performedwith a professional Alpha GUARD system. The mean values of the activity concentration of the exhaled radon of investigated building materials varied from 10 Bqm^{-3} to 101 Bqm^{-3}, radon exhalation rate values ranged from 77.0 mBqm^{-2}h^{-1} to 777.7 mBqm^{-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 earthhave 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 sourcesof radiaton, 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 majorcontributionto 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 especiallyimportantfor 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 assesment of risk exposure andthe development of adequate protection from radonrequire 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 γ -radiation, which originates from the disintegration of radioactive elements presentin 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.

Table 1. Activity concentration of^{226}Ra, ^{232}Th and ^{40}K in various structuralbuilding materials [7]

Type of construction material	A (Bq kg^{-1})		
	^{226}Ra	^{232}Th	^{40}K
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

[222]Rn atoms are generated from [226]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 significantlyaffect 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]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]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]:

$$E_{Rn} = \frac{C_{Rn}V\lambda}{S} \tag{1}$$

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), λ is the decay constant of radon ($2.1 \cdot 10^{-6}$ 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 α-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 2×10^6 Bqm[-3], while the temperature range is from -10 to 50 °C. The calibration error of this measurement system for [222]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).
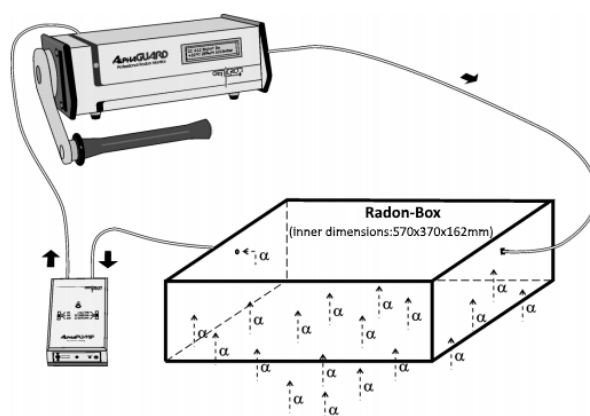

Figure 1. Experimental set-up for radon exhalation rate measurements (adopted from Manual AlphaGuard, Genitron Instruments) [12]

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 samplesare 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 hoursat 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.

Table 2. Radon activity concentration(C_{Rn}), radon exhalation rate (E_{Rn}) and gamma dose rate (H_γ) in buildingmaterials

| No.	Type of material	Commercial name or producer	C_{Rn} (Bq m^{-3})	E_{Rn} (mBq m^{-2} h^{-1})	H_γ (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	Kalvarija, Vitez	29	223.3	112
11	Block	Shamotte block, IGM Busovača	14	107.8	110
12	Block	Black block, Cazin	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	Vijenac, Lukavac	14	107.8	109
16	Limestone	Mrvelji, Posušje	33	254.1	111
17	Limestone	Sokolica, Zavidovići	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, Divič	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
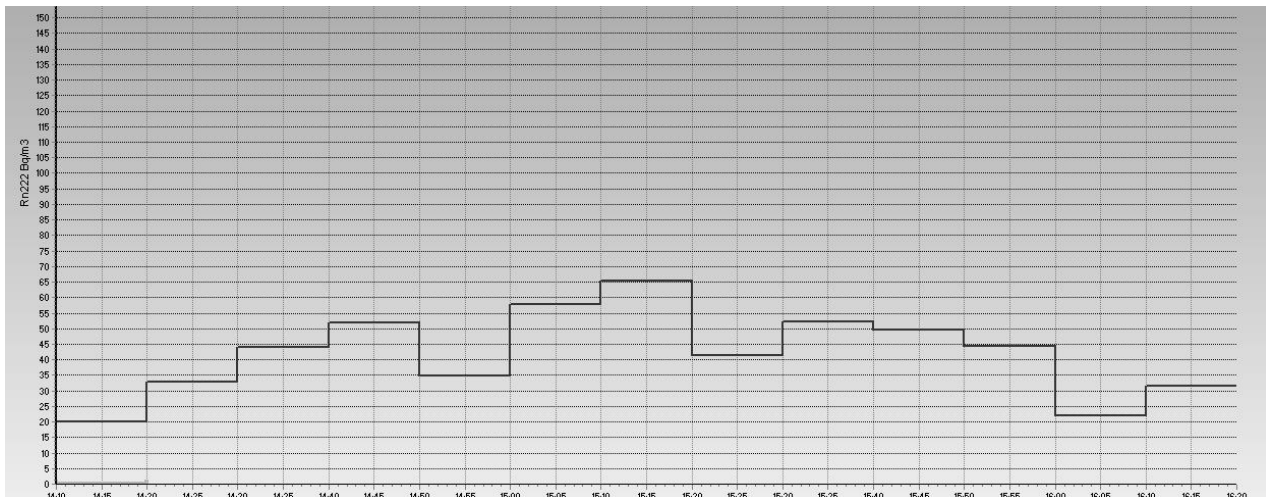
Figure 2. Activity concentration of exhaled radon from limestone (five minute intervals of time)- Jostanica, Divič

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 Bqm^{-3} to 132 Bqm^{-3}.

The radon exhalation rate for the investigated building materials samples was in the range from 77.0 mBqm^{-2}h^{-1} to 777.7 mBqm^{-2}h^{-1}. The minimum value was found for the sample No. 4, white cement, Spain, 77.0 mBqm^{-2}h^{-1}, whereas the maximum value was for sample No. 25, clay TOS Sarajevo, 777.7 mBqm^{-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 Bqm^{-3} to 52 Bqm^{-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 Bqm^{-3} to 44 Bqm^{-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, feldspart 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, ischaracterized by a low concentration of radon activity, 36 Bqm^{-3}. All investigated types of cement samples are characterized by low radon activity concentration values, which range from 10 Bqm^{-3} to 22 Bqm^{-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 Bqm^{-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 assesed 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]:

$$C_{\mathrm{Rn}} = \frac{E_{Rn} \cdot S_r}{V_r \cdot \lambda_v} \tag{2}$$

where C_{Rn} is the radon concentration (Bq m^{-3}), E_{Rn} is the radon exhalation rate (Bqm^{-2}h^{-1}), V_r is the room volume (m^3), λ_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}.

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

$$C_{\text{Rn}} = \frac{E_{Rn}}{0.9(\lambda_0 + \lambda_v)H} \qquad (3)$$

where H is the room height in meters (2.5 m), λ_0 is the radon decay constant (0.181 d^{-1}) and λ_v the air removal rate due to ventilation [10].

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

$$E = \frac{8760 \cdot 0.8 \cdot F \cdot C_{Rn}}{170 \cdot 3700} \qquad (4)$$

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 mSvWLM^{-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 μSv, 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 μSv, 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 aconsequence 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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МЈЕРЕЊЕ КОНЦЕНТРАЦИЈЕ АКТИВНОСТИ РАДОНА У ГРАЂЕВИНСКИМ МАТЕРИЈАЛИМА КОЈИ СЕ КОРИСТЕ У БОСНИ И ХЕРЦЕГОВИНИ

Сажетак: Човјек је перманентно изложен јонизирајућем зрачењу због присуства радиоактивних материјала, који се природно јављају у околини (НОРМ). Разним технолошким поступцима прераде и коришћења материјала који садрже природне радионуклиде добијају се материјали повећане природне радиоактивности (ТЕНОРМ). Највећи допринос озрачености природним изворима јонизирајућег зрачења, представља излагање становништва радону унутар затвореног простора, које потиче од дезинтеграције ^{226}Ra и ^{224}Ra, који су садржани у тлу испод зграда и грађевинским материјалима. У зависности од врсте грађевинских материјала, изложеност радону у затвореном радном и боравишном простору може бити и преко хиљаду пута већа него у отвореном простору. У Босни и Херцеговини нису вршена валидна и свеобухватна радиолошка истраживања грађевинских материјала, која би гарантовала њихову сигурност и дозиметријску исправност за уградњу у стамбене и индустријске грађевине, ауто-путеве, као и њихову примјену у друге сврхе. Квантификација нивоа радона, који долази из грађевинских материјала, представља неопходни и веома битни сегмент глобалне заштите становништва од јонизирајућег зрачења. У овом раду приказани су први резултати истраживања концентрације активности радона у грађевинским материјалима, који се користе у Босни и Херцеговини. Мјерења су вршена помоћу професионалног система Alpha GUARD. Средње вриједности концентрације активности ексхалираног радона истраживаних грађевинских материјала, варирају у интервалу од 10 Bq m^{-3}до 101 Bq m^{-3}, јачина ексхалације радона има вриједности у опсегу од 77.0 mBqm^{-2}h^{-1} до 777.7 mBqm^{-2}h^{-1}. Јачина дозе гама зрачења налази се у интервалу од 57–112 nSv h^{-1}.

Кључне ријечи: грађевински материјали, концентрација активности радона, јачина ексхалације радона, јачина дозе гама зрачења.

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