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ACTIVITY OF RADIONUCLIDES IN BUILDING MATERIALS

Abstract:

Activity concentrations of ^{226}Ra , ^{232}Th and ^{40}K in some building materials used in Montenegro were measured by the gamma spectrometry (HPGe spectrometer and multidetector spectrometer PRIPYAT-2M), and the results are presented in this paper. Levels of the radionuclides in analyzed sand, cement, brick, gypsum, ceramic tile, as well as marble and granite, are found to be significantly below the permitted limits for radioactive contamination of building materials used in construction. Hazard indices related to building materials – radium equivalent activity and activity concentration index – have also been evaluated and found to be below 370 Bq/kg (equivalent to the dose rate of 1.05 mSv per year) and 1, respectively. This indicates that materials in question pose no health risk.

Keywords: terrestrial radionuclides, gamma spectrometry, building materials, hazard indices

АКТИВНОСТ РАДИОНУКЛИДА У ГРАЂЕВИНСКИМ МАТЕРИЈАЛИМА

Сажетак:

Концентрације активности ^{226}Ra , ^{232}Th и ^{40}K у појединим грађевинским материјалима који се користе у Црној Гори мјерене су гама-спектрометријски (HPGe спектрометар и вишедетекторски спектрометар ПРИПЈАТ-2М), а резултати су приказани у овом раду. Нивои радионуклида у анализованом пијеску, цементу, цигли, гипсу, керамичкој плочици, као и мермеру и граниту, знатно су испод прописаних граница за радиоактивну контаминацију материјала који се користе у грађевинарству. Индекси ризика везани за грађевински материјал – радијум еквивалентна активност и индекс концентрације активности – такође су разматрани и били су испод 370 Bq/kg (што је еквивалентно јачини дозе од 1.05 mSv годишње) и 1, респективно. Ово указује да предметни материјали не носе ризик по здравље.

Кључне ријечи: терестријални радионуклиди, гама спектрометрија, грађевински материјали, индекси ризика

1. INTRODUCTION

Exposure to ionizing radiation from natural sources is the most significant part of the total exposure to radiation for most individuals. The annual effective dose of 2.4 mSv represents an average worldwide exposure to natural radiation sources, with a large contribution given by the radon (^{222}Rn) inhalation (1.15 mSv). The dose from external terrestrial radiation is the second largest contributor, in particular the dose from exposure indoors (0.41 mSv), which is significantly higher than the dose from outdoors exposure (0.07 mSv) [1], [2].

Terrestrial radiation is mainly gamma radiation emitted during radioactive decays of terrestrial radionuclides such as ^{40}K and radionuclides from the ^{232}Th and ^{226}Ra (^{238}U) series, naturally occurring in various degree in all soils, rocks, all terrestrial materials, as well as building materials derived from them. The absorbed dose rate in air calculated from medians of average concentrations of ^{226}Ra , ^{232}Th and ^{40}K in soils worldwide was found to be 51 nGy/h, while median of average absorbed dose rates in outdoor air is 57 nGy/h [1]. In regard to indoor air, these doses are found to be 75 nGy/h and 84 nGy/h, respectively, with the dose ratio (indoors to outdoors) in the range from 0.6 to 2.3 [1]. It is important to note that the mentioned ratios are influenced by the buildings structural properties, e.g. materials used in the constructions (acting as shields from outdoor radiation and simultaneously as radiation source indoors), but also thickness etc.

As it is well-known, building materials can contribute to indoor radiation exposure through both – radon inhalation and gamma radiation. Namely, in addition to ground underlying the dwellings, building material can also be the source of radon in a closed place, since ^{222}Rn is the product of α -decay of ^{226}Ra which is present in different concentrations in all building materials. At the same time, exposure to gamma radiation indoors dominantly depends on building materials used for construction. As stated in the UNSCEAR report 2000, this exposure “is inherently greater than outdoor exposure if earth materials have been used” [1], but depends on the occupancy duration as well. Having in mind the portion of time humans spend indoors (80%), a consideration of exposure to ionizing radiation indoors becomes more and more important from the radiation protection standpoint.

In contrast to lightweight houses (including wooden ones), where the absorbed dose rate in indoor air can be even lower than outdoors, in buildings constructed of stone, brick, concrete and similarly – that dose mainly depends on natural radionuclides (i.e. their activity concentrations) contained in the building materials. Therefore, natural radionuclides activity concentrations in building materials are the subjects of national legislations, guidance, recommendations and standards of the relevant international organizations, but also the European Union (EU) directives. The EU Council Directive 2013/59/EURATOM defined the reference level for indoor external exposure to gamma radiation emitted by building materials as 1 mSv per year [3].

Any building material which was identified as being of concern from a radiation protection point of view should be controlled on the radionuclides content, preferably before its placing on the market and applying in constructions. It also involves an evaluation of some other indices and relevant factors, all together liable to give dose which exceeds the mentioned reference level [3], [4].

A list of materials which require special attention with regard to gamma radiation includes: fly ash, marl, anhydrate used in the cement industry, clay used in the ceramics industry, etc. [1], [2], although each material with increased activity concentration of natural and/or artificial radionuclides can pose a risk. The 2013/59/EURATOM directive introduced an indicative list of types of building materials that can be of radiological significance. That list includes natural materials, such as alum-shale, materials of natural igneous origin (granitoides, tuff, lava, etc.), but also materials incorporating residues from processing naturally occurring radioactive material (fly ash, phosphorus slag, red mud, residues from steel production etc.) [3]. The use of such materials requires appropriate measures, which may include restrictions on their envisaged use or specific requirements in relevant building codes [3].

This paper deals with natural radionuclides (^{40}K , ^{232}Th and ^{226}Ra) activity concentrations in some building materials used in Montenegro, as well as with associated radiation protection issues, i.e. hazard indices as indicators for the material safe use.

2. MATERIALS AND METHODS

Samples of building materials – ceramics (ceramic tile), granite, marble, gypsum, brick, cement and sand – were analyzed in the Centre for Ecotoxicological Research in Podgorica (as a part of regular monitoring of radioactivity in the environment in 2018); and additional four sand samples were analyzed at the Faculty of Natural Sciences and Mathematics in Podgorica (as a part of wider research carried out in 2019 and 2020).

2.1. SAMPLE PREPARATION AND MEASUREMENTS

The samples analyzed in the Centre for Ecotoxicological Research were taken from local supplier of building materials in Podgorica. Dry samples of brick and ceramic tile have been pulverized, ground with the grinder and ball mill, sieved, finally dried and sealed in 1L Marinelli beakers. The representative samples of sand (dried at natural conditions and sieved), cement and gypsum were also sealed in 1L Marinelli beakers. All the samples were kept for more of 30 days at airtight condition to allow for radioactive equilibrium between radium and its daughters to be established before measurements.

The measurements were conducted using standard gamma spectrometry (Figure 1), i.e. the HPGe detector ORTEC GEM-40190, with relative efficiency of 40%. The system was calibrated using the calibration standards (Czech Metrology Institute), i.e. mixtures of gamma emitting radionuclides in Marinelli beaker. The measuring time for all the samples individually was 24 h. Processed pieces of granite and marble cut in the shape of a cylinder were directly placed at the detector and measured for 24 h as well. Efficiency calibration was performed using the software Angle [5].



Figure 1. Laboratory for gamma spectrometry in the Centre for Ecotoxicological Research in Podgorica – two ORTEC HPGe spectrometers (photography by author).

In all the cases, background spectra acquired over the same measuring times were subtracted from the building material spectra before analysis performed by the software Gamma Vision. The activity concentrations (A_c , in Bq/kg) of ^{226}Ra , ^{232}Th and ^{40}K were determined in a standard procedure, using the total absorption peaks created by the intense gamma rays from radionuclide daughters:

$$A_c = \frac{N}{t \varepsilon I_\gamma m}, \quad (1)$$

where N represents the net counts under the selected peak, t is the live measuring time, ε is the detection efficiency, I_γ is the relative gamma ray intensity, and m is mass of the sample. In regard to intense peaks, for ^{40}K – it is the peak created by the 1461 keV gamma rays (10.7%) emitted after electron capture, from the excited state of ^{40}Ar ; for ^{232}Th – the peaks created by gamma rays from beta minus decay of ^{228}Ac (to ^{228}Th) with the energies of 338 keV (11.4%), 911 keV (26.2%) and 969 keV (15.9%) – where activity concentration is an average of the activities obtained from the three peaks; for ^{226}Ra – the peaks created by gamma rays from beta minus decay of ^{214}Pb (to ^{214}Bi) with the energies of 295 keV (18.4%) and 352 keV (35.6%), as well as the peaks created by gamma rays from beta minus decay of ^{214}Bi (to ^{214}Po) with the energies of 609 keV (45.5%), 1120 keV (14.9%) and 1764 keV (15.3%) – where activity concentration is again an average of the activities obtained from the given peaks. The coincidence summing corrections were carried out using the Efftran and Mefftran (when dealing with Marinelli beakers) [6]. Additionally, activity concentration

of artificial radionuclide (fission product) ^{137}Cs was also considered on the base of the peak created by gamma rays with the energy of 662 keV (85%) accompanying beta minus decay of ^{137}Cs to ^{137}Ba . It should be noted that ^{137}Cs in environmental samples in Montenegro originates from the Chernobyl accident [7].

The sand samples analyzed at the Faculty of Natural Sciences and Mathematics in Podgorica were taken from four locations of private houses building – two in Podgorica and two in Nikšić. The samples were dried, sieved and hermetically sealed in 1L Marinelli beakers, and measured after 40 days by the multidetector 4π spectrometer PRIPYAT-2M (Figure 2), consisting of six NaI(Tl) detectors [8]. This spectrometer (with its software PRIP) had been previously used for a development of the coincident measuring methods [9], [10], measurements of radionuclides in environmental samples [11], and radon measurements [12]. It is also used for coincidence registration of gamma rays emitted from decays of some artificial radionuclides, such as ^{133}Ba and ^{134}Cs [13]. The spectrometer is characterized with relatively high detection efficiency, high sensitivity and a short measuring time. Activity concentrations of ^{40}K , ^{232}Th , ^{226}Ra and ^{137}Cs in four sand samples have been obtained from the spectra acquired for 1 h (each), using the intense gamma peaks.



Figure 2. Spectrometer PRIPYAT-2M at the Faculty of Natural Sciences and Mathematics in Podgorica (photography by author).

2.2. HAZARD INDICES

In order to consider hazardous effects of building materials, activity concentrations of natural radionuclides (and some artificial, e.g. ^{137}Cs) can be compared with limits specified in a legal framework for radiation protection. In the actual legal framework in Montenegro, the limits of radioactive contamination of building materials used in construction for interior and exterior, as well as materials used for the civil engineering constructions as substructure (for roads, playgrounds etc.) are given through limits for ^{226}Ra , ^{232}Th , ^{40}K and sum of activities of all artificial radionuclides (Table 1), and the gamma index which should be (in all cases) not greater than 1 [14].

Table 1. Limits of radionuclide activity concentration in building materials in Montenegro [14]

Radionuclide	Limits for interior (Bq/kg)	Limits for exterior (Bq/kg)	Civil engineering constructions (Bq/kg)
^{226}Ra	200	400	700
^{232}Th	300	300	500
^{40}K	3000	5000	8000
All artificial	4000	4000	2000

Activity concentrations of natural radionuclides can also be used to evaluate the radium equivalent activity (Ra_{eq}), which allows comparing concentrations of radionuclides in a certain material and represents the weighted sum of the radionuclide activity concentrations. It is usually calculated by the equation [15]:

$$Ra_{eq} = A_c(^{226}\text{Ra}) + 1.43 A_c(^{232}\text{Th}) + 0.077 A_c(^{40}\text{K}), \quad (2)$$

where A_c is the activity concentration (in Bq/kg) of given radionuclide in the building material. As defining the Ra_{eq} in this way, it is assumed that different activity concentrations of ^{226}Ra , ^{232}Th and ^{40}K produce the same dose rate. The recommended maximum value of the Ra_{eq} is 370 Bq/kg (equivalent to the dose rate limit of 1.05 mSv per year [16]). If the Ra_{eq} exceeds 370 Bq/kg, that material is considered to be one which can pose a radiation hazard. Therefore, such material should be further investigated, and its use might be restricted, though any decision on restricting material usage should be based on a precise dose assessment for real scenario and typical use of material in question.

The activity concentration index (I) required in the 2013/59/EURATOM directive is given by the equation [3]:

$$I = \frac{A_c(^{226}\text{Ra})}{300} + \frac{A_c(^{232}\text{Th})}{200} + \frac{A_c(^{40}\text{K})}{3000}, \quad (3)$$

where 300, 200 and 3000 are in Bq/kg, and the A_c are also in Bq/kg. This index is related to gamma radiation doses (in addition to outdoor external exposure), and its value of 1 can be used for identifying building materials that may cause exceeding the dose reference level (1 mSv per year). It can be used as a conservative screening tool only, since a precise dose evaluation needs taking into consideration some other factors as well (density, thickness and the other factors related to the building type, but also the intended usage of certain material, etc.) [3].

3. RESULTS AND DISCUSSION

3.1. RADIONUCLIDE ACTIVITY CONCENTRATIONS

The results of gamma spectrometric measurements of building materials are given in Table 2.

Table 2. Natural radionuclide activity concentrations in the building materials

Material	^{226}Ra (Bq/kg)	^{232}Th (Bq/kg)	^{40}K (Bq/kg)
Ceramic tile	31.8 ± 2.9	22.5 ± 2.2	320 ± 22
Granite	48.2 ± 3.1	25.5 ± 2.1	489 ± 31
Marble	4.10 ± 0.50	1.50 ± 0.10	3.05 ± 0.55
Gypsum	4.80 ± 0.60	2.10 ± 0.20	38.5 ± 2.2
Brick	98.0 ± 9.0	58.3 ± 4.9	298 ± 21
Cement	31.2 ± 0.5	3.01 ± 0.30	184 ± 9
Sand-1*	11.8 ± 0.6	10.1 ± 0.9	123 ± 13
Sand-2	9.14 ± 0.74	16.0 ± 0.9	41.2 ± 9.6
Sand-3	6.93 ± 0.31	2.16 ± 0.18	116 ± 10
Sand-4	17.7 ± 0.5	3.81 ± 0.40	305 ± 17
Sand-5	38.5 ± 1.7	19.3 ± 0.8	83.9 ± 7.4

*Sand-1 is sand sample analyzed by the HPGe spectrometer; the other sand samples were analyzed by the PRIPYAT-2M (sand-2 and 3 – samples from Podgorica, sand-4 and 5 – samples from Nikšić).

Activity concentration of ^{137}Cs in ceramic tile, granite, marble, gypsum and brick was found to be below minimum detectable activity of the HPGe spectrometer for given measuring time, which ranged from 0.21 Bq/kg in the case of brick to 0.44 Bq/kg in the case of ceramic tile. In the sample sand-1, activity concentration of ^{137}Cs has also been below minimum detectable activity (≤ 0.19 Bq/kg), while in sand-2, 3, 4 and 5 it was found to be (1.52 ± 0.19) Bq/kg, (0.33 ± 0.10) Bq/kg, (0.65 ± 0.10) Bq/kg and (0.28 ± 0.09) Bq/kg, respectively.

The results presented in Table 2 clearly show that natural radionuclides activity concentrations in the studied building materials are significantly below the limits given in Table 1, and they are safe for use in the building industry. Many studies reported that materials standardly used in the building industry are safe and cannot pose a radiological risk, if ^{226}Ra , ^{232}Th and ^{40}K activity concentrations were determined to be below the permitted limits. From that point of view, the results presented here are in a good agreement with results obtained in research on radioactivity in building materials commonly used in neighbor countries Serbia and Bosnia and Herzegovina [17], [18].

The highest ^{40}K activity concentration of 489 Bq/kg was found in the sample of granite, and that is significantly lower than in granite analyzed in Serbia (an average of 1280 Bq/kg for two samples [17]), but also lower than in brick and ceramic tile also analyzed in Serbia (average values of 646 Bq/kg for 12 samples and 828 Bq/kg for five samples, respectively [17]). In addition, the granite sample showed relatively low ^{226}Ra and ^{232}Th activity concentrations. For a comparison, in the granite from Serbia – their activities have been found to have averages of 200 Bq/kg and 77 Bq/kg, respectively [17]. Moreover, for example in the granite from China (Beijing) – ^{226}Ra and ^{232}Th activity concentrations were found to have averages of 356 Bq/kg and 318 Bq/kg, respectively [19]. The highest ^{226}Ra and ^{232}Th concentrations of 98 Bq/kg and 58.3 Bq/kg, respectively, were found in the brick sample. These ^{226}Ra and ^{232}Th concentrations are higher than in brick from Serbia [17], some building materials from Iberian Peninsula [20], or in brick from Poland [21] and some other countries in the world [22], [23]. At the same time, ^{226}Ra and ^{232}Th concentrations in marble and/or gypsum are lower than those found in the same materials used in some other countries [23], [24], [25], [26].

Sand samples showed ^{226}Ra activity concentration from 6.93 Bq/kg to 38.5 Bq/kg, ^{232}Th activity concentration from 2.16 Bq/kg to 19.3 Bq/kg, and ^{40}K activity concentration from 41.2 Bq/kg to 305 Bq/kg. For a comparison, previous research on radionuclide activity concentrations in beach sands on the Coast of Montenegro showed ^{226}Ra activity in the range (2.1–15.5) Bq/kg, while ^{232}Th and ^{40}K activities ranged from 1.4 Bq/kg to 16.6 Bq/kg, and from 7.1 Bq/kg to 305 Bq/kg, respectively [27].

Looking at the results in Table 2, activity concentrations of ^{226}Ra , ^{232}Th and ^{40}K in all the samples are below typical level of the radionuclides in building materials used traditionally for construction, often taken as 50 Bq/kg, 50 Bq/kg and 500 Bq/kg, respectively [16].

3.2. RADIUM EQUIVALENT ACTIVITY AND ACTIVITY CONCENTRATION INDEX

The radium equivalent activity calculated by the Eq. (2) for all the samples showed the range from 6.48 Bq/kg (marble) to 204 Bq/kg (brick), with an average of 62.8 Bq/kg (median: 46.6 Bq/kg) and standard deviation of about 58.4 Bq/kg. The Ra_{eq} for individual building material is shown in Figure 3, confirming that every value is significantly below the upper level of 370 Bq/kg. Taking into account average activity concentration of ^{226}Ra , ^{232}Th , ^{40}K in ceramic tile, granite, marble, gypsum brick, cement and sand-1, the Ra_{eq} would be around 74 Bq/kg.

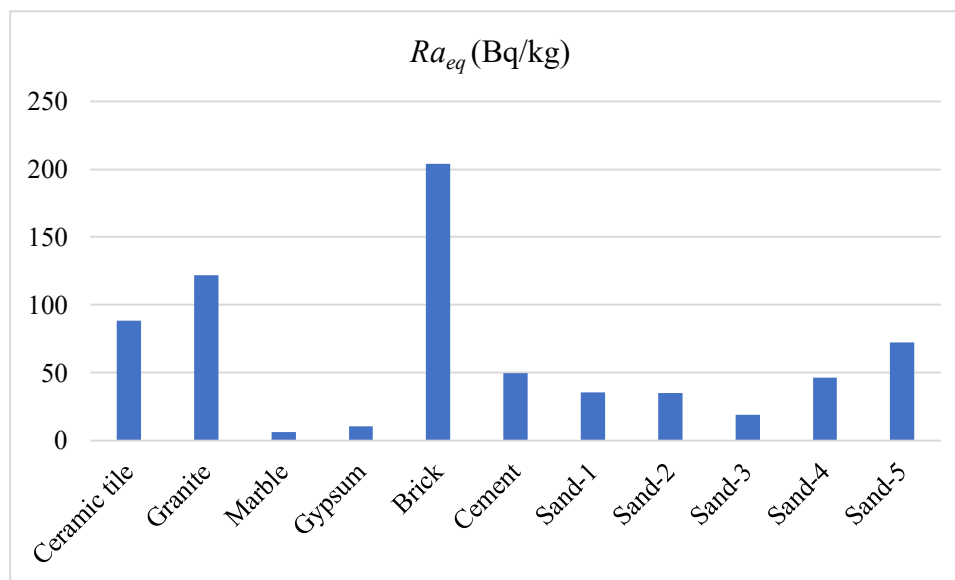


Figure 3. Radium equivalent activity for some building materials used in Montenegro (created by author).

The activity concentration index, calculated for all the samples by the Eq. (3), was found to have minimum value of 0.022 for marble and maximum of 0.718 for brick. An average value for all the samples is found to be 0.23. This hazard index also confirms that considered material is safe for use in construction. Taking into account average activity concentration of ^{226}Ra , ^{232}Th , ^{40}K in ceramic tile, granite, marble, gypsum brick, cement and sand-1, the activity concentration index is around 0.27.

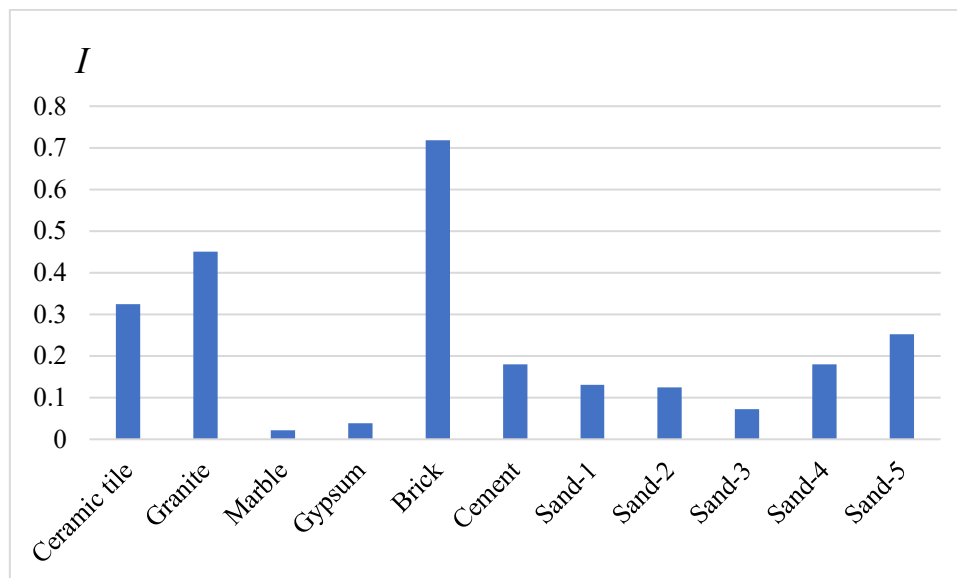


Figure 4. The activity concentration index for some building materials used in Montenegro (created by author).

It is worth noting that, when consider value of the activity concentration index for a certain material, the way and the amount material is used in construction, are very important. For instance, material used in bulk amounts (such as concrete) with $I \leq 0.5$ satisfies the dose criterion 0.3 mSv/y, and with $I \leq 1$ – dose criterion 1 mSv/y; whilst superficial materials (e.g. tiles) satisfy the same criterion with $I \leq 2$ and $I \leq 6$, respectively [28].

4. CONCLUSIONS

The building materials commonly used in Montenegro – sand, cement, brick, gypsum, ceramic tile, along with marble and granite – were analyzed in the present study in order to determine natural radionuclides activity level and consider potential health risk due to their use in a building construction.

Activity concentration of ^{226}Ra in the analyzed materials ranged from 4.10 Bq/kg (marble) to 98 Bq/kg (brick), which is significantly below the limits defined in the legal framework (the lowest for radioactive contamination of building materials used in construction for interior – 200 Bq/kg). The same conclusion can be done for ^{232}Th activity in the studied materials (from 1.50 Bq/kg in the marble sample to 58.3 Bq/kg in the brick sample), as well as ^{40}K activity (from 3.05 Bq/kg in the marble sample to 489 Bq/kg in the granite sample). Namely, the limit for ^{232}Th activity in building materials used in construction for interior and exterior is 300 Bq/kg, whilst for ^{40}K activity – 3000 Bq/kg and 5000 Bq/kg, respectively.

The radium equivalent activity and activity concentration index, as hazard indices for building materials, are found to be all within the safe limits – 370 Bq/kg and 1, respectively. However, a general conclusion regarding safety of building materials used in Montenegro cannot be done on the base of here presented results only. The main limitation of the present study is number of samples for certain type of building material (five sand samples and by one of the other material types). A detailed analysis should include significantly higher number of samples for each type of building material, to obtain representative highest and average values for both – radionuclide activity concentrations and hazard indices.

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