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THE RADIOACTIVITY OF BRICKS PRODUCED IN SERBIA†

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Abstract. *Natural radioactivity of the brick, one of the most used building materials was investigated. The content of natural radionuclides in the samples was measured by HPGe gamma-spectrometry. The average values of specific activities of ^{226}Ra, ^{232}Th and ^{40}K in the samples are: 31 ± 9, 40 ± 11 and 460 ± 118 in Bq kg^{-1}, respectively. The absorbed dose in the air was computed by the method of buildup factors for the standard model of the room. The obtained values for the absorbed dose are in the range of 9 – 47 (nGy h^{-1}), with the average of 36 ± 9 nGy h^{-1}. The radiation hazard was estimated calculating the corresponding gamma index and the effective dose. The gamma index belongs to the range of 0.11 to 0.56, and the effective dose is in the range of 0.044 – 0.23 (mSv), far below the reference limit of 1 mSv.*

Key words: *natural radioactivity, brick, dose, buildup factor*

1. INTRODUCTION

The structural building materials are obtained by processing rocks and minerals, so they usually contain natural radionuclides of terrestrial origin: ^{238}U (^{226}Ra), ^{232}Th and ^{40}K. High adsorption capability of the layers of clay minerals in sedimentary deposits - raw materials for brick production - can significantly contribute to the uranium and thorium concentration (Egidi and Hull, 1999) in the brick and enhance indoor exposure to ionizing radiation.

In the legislation of many countries, including Serbia (Off. Gazette RS, 2009), the prescribed limit of the individual dose above background radiation in the course of one year is 1 mSv. For this value, dose criterion (EC, 1999) was derived for the feasibility of building materials – the index of activity concentrations, i.e. the gamma index:

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$$I = \frac{A_{\mathrm{Ra}}}{300} + \frac{A_{\mathrm{Th}}}{200} + \frac{A_{\mathrm{K}}}{3000} \leq 1 \tag{1}$$

where the A_{Ra}, A_{Th} and A_{K} denote specific activity of ^{226}Ra, ^{232}Th and ^{40}K (in Bq kg^{-1}) in the material. This formula applies to the building materials used in full in the interior.
The aim of this study is to examine the fulfillment of dosimetric criterion, so the content of natural radionuclides in samples of brick from different producers from Serbia was measured in the work and corresponding gamma indices were determined. For a more accurate determination of the fulfillment of dose limit condition, for each of investigated brick samples the corresponding effective dose was calculated.

2. METHOD

Samples preparation and measurement

Samples of brick from different manufacturers were acquired on the market, and two products were sampled on the site. After a manual crushing, the samples were homogenized in a mill to a granule size of 3 mm. By exposure to the temperature of 105° C in an oven, the residual moisture was removed from the samples, and after that they were placed in Marinelli beakers of 1l volume. About 1.6 kg of the samples was used for measurement (density of sample was 1.6 g cm^{-1}). The measurements were carried out 30 days after the sealing, when radioactive equilibrium between ^{226}Ra and its progenies ^{214}Pb and ^{214}Bi was reached.

The content of radionuclides in the samples was explored by the method of gamma-spectrometry with a Canberra HPGe detector calibrated in the energy range 40 keV - 3000 keV, with relative efficiency 26% and *FWHM* (full width at half of maximum) = 1.8 at 1332 keV (^{60}Co), using the software Genie 2000, Canberra, USA. The energy calibration was carried out using the gamma standard of Czech Metrological Institute, Praha, Czech Republic (Cert. No: 931-OL-004/04, 07. 01. 2004) in the geometry of Marinelli beaker. The following lines were used: 59.5 keV (^{241}Am), 661.7 keV (^{137}Cs), 1173.2 keV (^{60}Co) and 1332.5 keV (^{60}Co).

For measurements of ^{226}Ra, the photopeaks ^{214}Pb (295 keV and 351 keV) and ^{214}Bi (609 keV and 1120 keV) were used. The specific activity of ^{232}Th was determined on the basis of energy lines in the spectrum of ^{228}Ac (338 keV and 911 keV), while the activity of ^{40}K was determined using the photopeak at energy of 1461 keV. The acquisition time of the spectral data was at least one hour. The specific activities, A (Bq kg^{-1}), of ^{226}Ra, ^{232}Th and ^{40}K in the samples were measured with the associated combined standard uncertainty at 1σ confidence level of 10%.

Calculation

The effective dose, E (Sv), corresponding to the gamma radiation of a building material was determined from the expression:

$$E(\mathrm{mSv}) = D(\mathrm{nGy\,h^{-1}}) \cdot 8760(\mathrm{h}) \cdot 0.8 \cdot 0.7(\mathrm{Sv\,Gy^{-1}}) \cdot 10^{-6} \tag{2}$$

In this relation D represents the absorbed dose in the air:

$$D = q_{\mathrm{Ra}} A_{\mathrm{Ra}} + q_{\mathrm{Th}} A_{\mathrm{Th}} + q_{\mathrm{K}} A_{K}, \tag{3}$$

produced by the activities of ^{226}Ra, ^{232}Th and ^{40}K in building material (EC, 1999; UNSCEAR, 2000).

In this paper, specific absorbed dose rates, q_{Ra}, q_{Th} and q_{K}, were computed for the detection point in the center of the standard room (Koblinger, 1978), taking into account only the walls' contribution (floor and ceiling are excluded), as well as for the whole room (with floor and ceiling). The calculation was performed assuming the brick density of $\rho = 1.6$ g cm^{-3} and its chemical composition: $_{8}$O (51.35%), $_{14}$Si (32.25%), $_{13}$Al (14.30%), $_{22}$Ti (1.20%), $_{26}$Fe (0.90%) (Sheppard, 1986).

The method of buildup factors was used for dose calculation, as in the papers by Manic et al. (2012, 2014), applying the parametric (G-P) i.e. Harima form (Harima, 1991):

$$\begin{aligned} &B(\mu r, E) = 1 + (b-1)\mu r \text{, for } K = 1 \\ &B(\mu r, E) = 1 + (b-1)(K^{\mu r} - 1)/(K-1) \text{, for } K \neq 1 \\ &K(\mu r, E) = c(\mu r)^{a} + d\,\frac{Tanh(\mu r / X_{K} - 2) - Tanh(-2)}{1 - Tanh(-2)} \end{aligned} \tag{4}$$

which is the most precise functional form of the buildup factors. In this expression, μ represents the coefficient of attenuation of gamma radiation in a material, E is the energy of photons, r denotes the distance which gamma ray travels in the material, and a, b, c, d and X_{K} are the parameters of buildup factors B.

The parameters of the exposure buildup factors for brick were determined by the interpolation by atomic number, with prior calculation of equivalent atomic number by interpolation on the logarithm of the coefficient of attenuation. For equivalent atomic number Z_{eq}, (G-P) parameters for certain integer energies were computed by interpolation by logarithm of atomic number (Singh et al., 2010). Then the energy interpolation was carried out, where the parameters a, b, c, d and X_{K} for ^{226}Ra, ^{232}Th and ^{40}K energy groups were calculated by the parabolic interpolation (Yoshida, 2006), based on (G-P) parameters for the corresponding neighboring energies, published in the ANSI/ANS (1991) standard.

The energy transfer coefficients were determined using the appropriate tabulated values (Hubel, 1969), by interpolation on logarithm of energies. The attenuation coefficients for brick composition were obtained using the program XCOM (Berger et al., 2010).

3. Results and Discussion

The equivalent atomic number, Z_{eq}, for brick, which was determined by the interpolation on attenuation coefficients is presented in Table 1. The parameters of (G-P) exposure factors calculated by interpolation on atomic number, are also shown.

Table 1 Equivalent atomic number and parameters of exposure buildup factors for brick

E (MeV)	Z_{eq}	b	c	a	X_k	d
0.03	12.07	1.283	0.447	0.190	16.24	-0.1022
0.04	12.13	1.598	0.550	0.146	13.85	-0.0775
0.05	12.18	2.037	0.639	0.120	13.68	-0.0626
0.06	12.21	2.407	0.773	0.078	13.65	-0.0567
0.08	12.24	2.873	1.011	0.012	13.69	-0.0238
0.10	12.28	3.028	1.195	-0.028	13.99	-0.0080
0.15	12.32	2.981	1.422	-0.069	14.56	0.0116
0.20	12.35	2.822	1.508	-0.083	14.25	0.0169
0.30	12.39	2.578	1.542	-0.090	14.17	0.0206
0.40	12.44	2.422	1.522	-0.089	14.60	0.0215
0.50	12.49	2.301	1.490	-0.086	14.53	0.0215
0.60	12.53	2.208	1.460	-0.083	14.91	0.0236
0.80	12.56	2.089	1.391	-0.074	13.69	0.0215
1.00	11.99	2.004	1.350	-0.069	15.69	0.0235
1.50	11.29	1.875	1.234	-0.049	15.00	0.0168
2.00	11.13	1.794	1.154	-0.034	14.66	0.0120
3.00	11.01	1.684	1.056	-0.011	10.53	0.0009
4.00	11.00	1.606	0.992	0.006	12.86	-0.0089

A similarity can be noticed between the values of Z_{eq} and the Z_{eq} for concrete given in Harima et al., 1991 and Mann et al., 2012. Also, Z_{eq} mostly belongs to the lower part of the range of atomic numbers characteristic for most rocks and minerals (IAEA, 2003).

Table 2 Specific absorbed dose rates in the room of bricks and of concrete

	q (nGy h^{-1}/ Bq kg^{-1})		
	^{226}Ra	^{232}Th	^{40}K
Walls of the standard room			
Brick	0.291	0.364	0.0281
Whole standard room			
Brick	0.613	0.770	0.0598
Concrete	0.757	0.912	0.0700

Table 2 shows the results of calculation of specific absorbed doses in the center of the room 5 × 4 × 2.8 m^3 with walls 20 cm thick (parameters of the standard room), originating from the radiation of ^{226}Ra, ^{232}Th and ^{40}K only from the walls. For comparison, the results are given for q_{Ra}, q_{Th} and q_K if the whole room is made of bricks, as well as for the standard room made from concrete.

It is obvious that in the standard room the coefficients q have higher values for concrete with respect to brick, which results primarily from the higher density of this material (2.35 g cm^{-3}). The influence of the chemical composition is not great, it is around 5%. Namely, the mass attenuation coefficients, (μ/ρ), and buildup factors corresponding to the chemical composition of brick evidently differ from the data for concrete only in the range of low energy, where the contribution of the photoelectric effect (with a marked dependence on Z_{eq}) in the total attenuation coefficient is significant.

The measured values of radionuclide specific activities in 24 samples of bricks (solid bricks and blocks) are shown in Table 3. In this table we also list the calculated values of gamma index, absorbed dose rate, as well as the effective dose rate for the investigated samples, implying their use only in the walls of the standard room.

Table 3 Specific activities, *A* (Bq kg^{-1}), of natural radionuclides in brick samples; the associated standard uncertainty is 10%. The corresponding gamma indices *I*, absorbed doses *D* and effective doses *E*

Sample	*A* (Bq kg^{-1})			*I*	*D* (nGy h^{-1})	*E* (mSv)
	^{226}Ra	^{232}Th	^{40}K			
Solid bricks						
1.	32	40	532	0.48	39	0.19
2.	28	39	465	0.44	35	0.17
3.	40	45	580	0.55	44	0.22
4.	34	41	547	0.50	40	0.20
5.	35	48	530	0.53	43	0.21
6.	33	29	418	0.39	32	0.16
7.	42	52	519	0.57	46	0.22
8.	30	41	488	0.47	37	0.18
9.	34	45	462	0.49	39	0.19
10.	31	39	544	0.48	38	0.19
Blocks						
1.	29	44	479	0.48	38	0.19
2.	27	39	464	0.44	35	0.17
3.	24	41	460	0.44	35	0.17
4.	30	41	485	0.48	37	0.18
5.	29	38	429	0.43	34	0.17
6.	7	11	108	0.11	9	0.04
7.	6	11	112	0.11	9	0.04
8.	26	39	412	0.42	33	0.16
9.	37	50	462	0.53	42	0.21
10.	39	54	583	0.59	47	0.23
11.	29	33	453	0.41	33	0.16
12.	36	47	456	0.51	40	0.20
13.	41	48	536	0.56	44	0.22
14.	35	46	506	0.52	41	0.20

The average values of specific activities of ^{226}Ra, ^{232}Th and ^{40}K in the samples and the corresponding standard deviations are: 31 ± 9, 40 ± 11 and 460 ± 118 in Bq kg^{-1}, respectively. These values slightly differ from the contents of radionuclides in brick published in (Krstić et al., 2007) – the mean values in that paper are: A_{Ra} = 34, A_{Th} = 43 and A_{K} = 579 in Bq kg^{-1}.

It can be observed that the results are grouped into relatively narrow bands – the standard deviations are about 30%. The exceptions are two blocks (6. and 7.) by a manufacturer from Eastern Serbia. For them, significantly lower activities of all three radionuclides in comparison to other samples were measured, which are characteristic of “sandy clay“ bricks

(El-Tahawy and Higgy, 1995). The similarity of the results reflects a similar composition of the main raw materials used in the products from different manufacturers, while the measured activities are largely determined by mechanisms of radionuclide concentrating in the layers of the clay (^{238}U and ^{232}Th), as well as clay composite properties.

The mean value and the standard deviation of the index of activity concentrations, determined by the measured activities, are 0.46 ± 0.12. Otherwise, the gamma index belongs to the range of 0.11 to 0.56, i.e. each brick sample fulfills the dosimetric criterion $I < 1$.

The values of the absorbed dose are in the range of 9 – 47 (nGy h^{-1}), with the average and standard deviation of 36 ± 9 nGy h^{-1}. These values are lower than the data for the absorbed dose, which corresponds to the use of bricks in many EU countries (Nuccetelli et al., 2012). The effective dose belongs to the range of 0.044 – 0.23 (mSv), with a mean of 0.18 ± 0.05 mSv, far below the dose limit of 1 mSv.

3. CONCLUSIONS

The application of dosimetry criterion in the protection against ionizing radiation implies the restriction of population exposure by limiting the individual annual effective dose to the value of 1 mSv. Since dose limit takes into account the irradiation which is a result of human activities (work), exposure to gamma radiation of building materials refers to the increase in dose level compared to the NORM background (STUK, 2010), for which external irradiation is 0.41 mSv (UNSCEAR, 2000). The results for the effective dose produced by use of bricks are lower than this value, so the dosimetry criterion is fulfilled for all investigated brick samples.

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RADIOAKTIVNOST CIGLE U SRBIJI

U radu je istražena prirodna radioaktivnost cigle, jednog od najčešće korišćenih građevinskih materijala u Srbiji. Sadržaj prirodnih radionuklida u uzorcima izmeren je metodom HPGe gama-spektrometrije. Srednja vrednost specifičnih aktivnosti ^{226}Ra*,* ^{232}Th *i* ^{40}K *u uzorcima je* 31 ± 9*,* 40 ± 11 *i* 460 ± 118 *u* $Bq\ kg^{-1}$*, respektivno. Apsorbovana doza u vazduhu izračunata je primenom metode faktora nagomilavanja za standardni model prostorije. Dobijene su vrednosti apsorbovane doze u opsegu 9 – 47 (*$nGy\ h^{-1}$*), sa srednjom vrednošću* 36 ± 9 $nGy\ h^{-1}$*. Za procenu radijacionog rizika, za svaki uzorak cigle određena je efektivna doza i odgovarajući gama indeks. Gama indeks pripada opsegu od 0,11 do 0,56, a efektivna doza je u opsegu od 0,044 – 0,23 (mSv), daleko ispod limita doze od 1 mSv.*

Ključne reči: *prirodna radioaktivnost, cigla, doza, faktor nagomilavanja*