

THE DOSE FROM RADIOACTIVITY OF COVERING CONSTRUCTION MATERIALS IN SERBIA

by

Vesna M. MANIĆ¹, Goran J. MANIĆ², Dragoslav R. NIKEZIĆ³, and Dragana Ž. KRSTIĆ^{3*}

¹ Department of Physics, University of Niš, Niš, Serbia

² Institute of Occupational Health, Niš, Serbia

³ Faculty of Science, University of Kragujevac, Kragujevac, Serbia

Scientific paper

DOI: 10.2298/NTRP1504287M

The indoor dose due to the radiation of ceramic and granite tiles, marble, granite and travertine plates, as well as some components of covering materials, produced in Serbia or imported from other countries, was estimated in the work. Activity concentrations of ²²⁶Ra, ²³²Th, and ⁴⁰K were measured by the standard gamma-spectrometry system. The lowest content of the radionuclides was found in white marble ($A_{Ra} = 2$ Bq/kg, $A_{Th} < 2$ Bq/kg, and $A_K < 3$ Bq/kg), while the highest activities were in some granite samples (Balmoral red: $A_{Ra} = 200$ Bq/kg, $A_{Th} = 378$ Bq/kg, and $A_K = 1679$, and Madura Gold: $A_{Ra} = 273$ Bq/kg, $A_{Th} = 20$ Bq/kg, and $A_K = 1456$ Bq/kg). The indoor absorbed dose rate in air due to the gamma radiation from covering materials was determined based on the specific absorbed dose rate computed in this work. Concentration of ²²²Rn that emanated into the indoor space was also calculated from the known ²²⁶Ra content. The radiation hazard estimated from the usage of each sample was expressed through the evaluated effective dose. Almost all samples, except one, fulfil the dosimetric criterion for safe use.

Key words: dose, activity concentration, natural radionuclide, covering material

INTRODUCTION

The radiation protection principle of dose limits implies the restriction of the individual effective dose to the value of 1 mSv per year above the typical natural background [1]. According to the UNSCEAR 2008 [2] data for indoor doses received from gamma radiation (0.41 mSv per year) and by inhalation of radon (1.15 mSv per year), the value of 1.56 mSv per year can be considered as the average background level for the exposure to building materials radiation.

Covering building materials may contain significant amounts of natural radionuclides and thus increase the indoor dose. In papers [3-11], the activity concentrations of ²²⁶Ra, ²³²Th, and ⁴⁰K (A_{Ra} , A_{Th} , and A_K) in covering materials used in different countries have been shown. The results vary in a wide range, *i. e.*, measured values for tiles are A_{Ra} : 28-105 Bq/kg, A_{Th} : 33-88 Bq/kg, and A_K : 24-1049 Bq/kg, for granite is A_{Ra} : 1-588 Bq/kg, A_{Th} : 1-906 Bq/kg, and A_K : 58-1606 Bq/kg, while in the case of marble: A_{Ra} : 2-37 Bq/kg, A_{Th} : 3-19 Bq/kg, and A_K : 4-310 Bq/kg. The values of the activity concentration for the materials in Serbia, published in the papers [12-14] are also in these ranges.

In estimation of the dose originating from the radioactivity of ²³⁸U (²²⁶Ra), ²³²Th, and ⁴⁰K in a covering material, in addition to the radiation from the material itself, the effect of the covering of a structural material (concrete) also has to be taken into account [15, 16]. To determine the dose that originates from the cover layer, it is necessary to know the concentration of radionuclide activities in the structural material. In the document EC 1999 [16] the hypothetical values of the activities, typical for EU materials, have been used, so the absorbed dose of covering material is expressed through unique conversion coefficients. For a material with thickness of 3 cm and a density of 2.6 g/cm³, placed over all the walls of a concrete room, for the specific absorbed dose rates of ²²⁶Ra, ²³²Th, and ⁴⁰K it was obtained: $q_{Ra} = 0.12$, $q_{Th} = 0.14$, and $q_K = 0.0096$, in nGy/h per Bq/kg.

However, covering materials for common indoor usage have various thicknesses, often less than 3 cm. Also, depending on the composition, material density varies within the same species. In this paper, therefore, the doses originated from natural radionuclides in covering materials of different thickness and density were evaluated. The results of the calculation of specific absorbed doses and exhalation rate of ²²²Rn (per unit concentration of ²²⁶Ra) were applied to certain materials produced or imported in Serbia, for which the activity concentrations of ²²⁶Ra, ²³²Th, and ⁴⁰K were known.

* Corresponding author; e-mail: dragana@kg.ac.rs

MATERIALS AND METHODS

Measurement

Measurement of radionuclide content were made for 35 samples of the covering building materials, final construction products and material components, originating from Serbia or imported. The types of investigated materials and their producers are listed in tab. 1, in an overview of the results of measurement of radionuclide activities.

The activity concentrations of ^{226}Ra , ^{232}Th , and ^{40}K in the materials (powdered and hermetically closed in 1 L Marinelli beakers) were determined by standard gamma ray spectrometry. The Canberra HPGe detector with relative efficiency 26 % and FWHM = 1.8 at 1332 keV (^{60}Co) was used. Efficiency calibration of the detector was performed in the energy range 40-3000 keV.

Calculation

The total effective dose E from a covering material can be represented as a sum of the contributions of external gamma ($E_{\text{cov}}_{\text{ext}}$) and internal (^{222}Rn) irradiation ($E_{\text{cov}}_{\text{int}}$) [2], reduced by the amounts corresponding to the shielding of radiation from the structural material, ($\Delta E_{\text{str}}_{\text{ext}}$) and ($\Delta E_{\text{str}}_{\text{int}}$), respectively

$$E = E_{\text{ext}} + E_{\text{int}} - (\Delta E_{\text{str}})_{\text{ext}} - (\Delta E_{\text{str}})_{\text{int}} \quad (1)$$

(indices cov and str refer to the covering and structural material).

The effective dose of gamma radiation is usually calculated from the absorbed dose rate in the air, D , which, for covering building materials, can be represented in the form [16]

$$D = D_{\text{cov}} + \Delta D_{\text{str}} = (q_{\text{U}} A_{\text{U}} + q_{\text{Th}} A_{\text{Th}} + q_{\text{K}} A_{\text{K}})_{\text{cov}} - (\Delta q_{\text{U}} A_{\text{U}} + \Delta q_{\text{Th}} A_{\text{Th}} + \Delta q_{\text{K}} A_{\text{K}})_{\text{str}} \quad (2)$$

where A_{U} , A_{Th} , and A_{K} , are the activity concentrations of ^{238}U , ^{232}Th , and ^{40}K in a material. Coefficients q_{U} , q_{Th} , and q_{K} denote the specific absorbed dose rates of these radionuclides for covering material and Δq_{U} , Δq_{Th} , and Δq_{K} are the appropriate amounts of reduction of these parameters for structural material. Using the conversion factor $9 \text{ nSv m}^3/(\text{Bq h})$, according to the UNSCEAR 2006 [17], E_{int} is determined by the concentration of ^{222}Rn exhaled from the material

$$C_{\text{Rn}} = \frac{1}{\lambda_{\text{v}}} \frac{S}{V} (J_{\text{cov}} - \Delta J_{\text{str}}) \quad (3)$$

where the exhalation rate, J_{cov} , and the flux attenuation, ΔJ_{str} , are proportional to the concentration of ^{226}Ra in individual materials [18].

Specific absorbed doses for ^{238}U , ^{232}Th , and ^{40}K radiation were computed for a detection point at the centre of the room with walls of concrete 20 cm thick, with standard room dimensions: 5 m \times 4 m \times 2.8 m [19]. Uniform covering of surface (all the walls, floor, and ceiling) with the layer of covering material with thickness of 0.5-3 cm and density of 1.5-3 g/cm^3 was assumed.

The calculation of q_{U} , q_{Th} , and q_{K} was performed in KERMA approximation, for NBS air composition [20], using the (G-P) build-up factors [21]. The Kalos form of build-up factors for the combination of layers was applied in the assessment of the effect of covering to the reduction of dose of structural material (concrete,) [22]. Further, for determination of the dose from a very thin surface layer (glaze) only the non-collided component of the radiation was considered. The computation was done by software Mathematica 8, applying the Maduar and Hiromoto [23] method of integration over the volumetric source of radiation.

The concentration of ^{222}Rn in the room was calculated from the expression for the intensity of the exhalation from the wall [18], where the effect of the attenuation of ^{222}Rn flux from concrete, caused by the presence of the covering layer, has been estimated using the solution of the transport equation, given in NUREG [24]. Typical parameters were used – the coefficient of the emanation: 0.15 [25], the diffusion length for ^{222}Rn in granite: 16 cm [26], and in concrete: 40 cm [27]. The value of the intensity of ventilation was 0.5 per hour, the equilibrium factor between radon and its progenies was 0.5, while possible differences in the moisture content and porosity of the materials were not taken into account.

The chemical content of each of explored building materials was approximated with the elemental composition of ceramics, published in Turtiainen *et al.* [28]. The attenuation coefficients for gamma radiation of radionuclides in this composite were obtained using the program XCOM [29]. In addition to the interpolation on the energy [30], the parameters of (G-P) build-up factors for ceramics were determined by the Lagrange interpolation of the ANSI/ANS [31] data on the atomic number [32]. The effective atomic number, Z_{eff} , was evaluated, which does not vary significantly with energy: $Z_{\text{eff}} = 11.5-13.5$, due to the dominant participation of the Compton scattering in the attenuation of radiation. Otherwise, these values of Z_{eff} belong to the range characteristic for most of the rocks and minerals [33] and are close to the results for concrete [21, 34].

RESULTS

Measurement

The activity concentrations, A (Bq/kg), of natural radionuclides: ^{226}Ra , ^{232}Th , and ^{40}K (with associated

Table 1. The activity concentrations A of natural radionuclides in the covering building materials; the average values are in the brackets

Material (species, manufacturer)	Activity concentrations A [Bqkg ⁻¹]		
	²²⁶ Ra	²³² Th	⁴⁰ K
Ceramic tiles			
“Polet”, Bečej, Serbia	50 6	57 6	696 73
“Toza Marković”, Kikinda, Serbia	43 5	49 6	556 58
“Keramika Kanjiža”, Kanjiža, Serbia	66 7	65 7	729 75
“Zorka keramika”, Šabac, Serbia	75 8	58 6	720 81
“Keramika Vojnić”, Croatia	41 5	35 4	619 62
“Martex”, Slovenia	30 3	38 4	423 44
“KAI Group”, Bulgaria	32 4	52 6	195 21
“Prima”, Spain	54 6	59 6	940 100
“Pamisa”, Spain	48 5	55 6	870 90
“Gemma”, Egypt	32 4	47 5	607 62
	[47 5]	[52 6]	[636 67]
Granite tiles			
“Emil Ceramica Group”, Italy	34 4	38 5	403 45
“Gres Ceramico Porcellanato”, Italy	56 6	51 6	641 65
(black) “Ergon”, Italy	32 4	55 6	633 65
(beige) “Ergon”, Italy	93 9	59 7	491 50
(gray) “Ergon”, Italy	37 4	62 7	721 73
“Keramin”, Belarus	44 5	65 7	324 34
	[50 5]	[55 6]	[536 55]
Granite			
“Granit”, Jablanica, Bosnia and Herzegovina	<2	<2	70 8
“Mermeren kombinat”, Prilep, Macedonia	23 3	33 4	560 59
“Krin”, Prilep, Macedonia	61 7	67 9	1143 133
“Pirin Mramor”, Sandanski, Bulgaria	31 3	47 5	812 83
“Pavlidis”, Drama, Greece	12 1	19 2	997 100
“Asan Insaat”, Ankara, Turkey	16 2	15 2	327 35
“Blockgandara”, Spain	61 7	106 12	1130 118
Royal White “Marmor Kamin”, Greece	10 1	12 1	234 9
Absolute Black “Marmor Kamin”, Greece	10 1	11 1	219 9
Madura Gold “Marmor Kamin”, Greece	273 14	20 2	1456 30
Balmoral Red “Pavlidis”, Drama, Greece	200 22	378 40	1679 172
New Imperial Red, China	89 9	110 10	730 75
	[65 6]	[68 8]	[780 70]
Marble			
White marble Arandjelovac, Serbia	2 0.2	<2	<3
“Mermeren kombinat”, Prilep, Macedonia	<2	<2	6 0.7
“Predsednik”, Vratsa, Bulgaria	11 1	2 0.3	33 4
“Komuru Cuoglu Mermer”, Turkey	3 0.4	<1	3 0.3
	[4 0.5]	[2 0.3]	[11 2]
Travertine			
“Feranex”, Vaduz, Turkey	10 1	28 3	208 22
Perlite			
“S & B Industrial Minerals”, Athens, Greece	58 3	69 4	1068 52
Zircobit			
“Industrie Bitoss”, Sovigliana, Italy	2891 310	493 51	47 5

Table 2. Specific absorbed doses q_{cov} from the covering layer of thickness d and density ρ ; the lowering of specific absorbed doses for concrete, Δq_{str} , with respect to the values of q_{str} in the standard room [the contribution of the floor is: 26.5 2.3 %]

d [cm]; ρ [gcm ⁻³]	q_{cov} [nGyh ⁻¹ per Bqkg ⁻¹]			q_{str} [nGyh ⁻¹ per Bqkg ⁻¹]		
	²³⁸ U	²³² Th	⁴⁰ K	²³⁸ U	²³² Th	⁴⁰ K
0.7; 2.4	0.0549	0.0649	0.00486	0.0520	0.0570	0.00390
1; 2.8	0.0905	0.107	0.00800	0.0860	0.0940	0.00640
2; 1.7	0.109	0.129	0.00932	0.107	0.117	0.00810
3; 2.6	0.241	0.289	0.0209	0.227	0.248	0.0173

combined standard uncertainties at 1σ confidence level), measured in covering building materials used in Serbia, are presented in tab. 1. Obviously, the lowest content of the radionuclides was determined for white marble: $A_{Ra} = 2$ Bq/kg, $A_{Th} < 2$ Bq/kg, and $A_K < 3$ Bq/kg; while the highest activities were exhibited by samples of granite: Balmoral red ($A_{Ra} = 200$ Bq/kg, $A_{Th} = 378$ Bq/kg and, $A_K = 1679$ Bq/kg) and Madura Gold: $A_{Ra} = 273$ Bq/kg, $A_{Th} = 20$ Bq/kg, and $A_K = 1456$ Bq/kg.

The variations of the content of radioactive elements in the samples reflect the different geological origin of the constituent minerals and rocks. Thus, travertine, typical for limestone sediments, has low concentrations of ²²⁶Ra, ²³²Th, and ⁴⁰K: 10 Bq/kg, 28 Bq/kg, and 208 Bq/kg, respectively, for the sample studied in this paper. The values of activity concentration of ²²⁶Ra and ²³²Th in granite materials are mainly determined by the abundance of the actinides in accessory minerals: zircon, monazite, apatite [33]. The activities of natural radionuclides, measured in the samples of granite plates were within following ranges: $A_{Ra} = 2-273$ Bq/kg, $A_{Th} = 2-378$ Bq/kg and $A_K = 70-1679$ Bq/kg, and for granite tiles: $A_{Ra} = 32-93$ Bq/kg, $A_{Th} = 38-65$ Bq/kg, and $A_K = 324-721$ Bq/kg. Accessory minerals are likely those which determine the radioactivity of perlite ($A_{Ra} = 58$ Bq/kg, $A_{Th} = 69$ Bq/kg, $A_K = 1068$ Bq/kg), the material obtained from the aluminosilicate ore of volcanic origin. The similar ²²⁶Ra and ²³²Th content in this material (observed also in most other samples) may indicate the coherent geochemistry of ²³⁸U and ²³²Th in constitutive minerals, which could be caused by similar crystallochemical properties of these radioelements in igneous rocks [33].

The radionuclide content in the layers of raw clay material and pigment minerals in the glaze contributes to the radioactivity of ceramic tiles $A_{Ra} = 30-75$ Bq/kg, $A_{Th} = 35-65$ Bq/kg, and $A_K = 423-940$ Bq/kg. For one of them, produced on the basis of zircon minerals – zircobit, unusually high activity was measured: $A_{Ra} = 3000$ Bq/kg. The apparent difference between the content of ²²⁶Ra and ²³²Th in this sample (as in granite Madura gold) can point out to a disequilibrium among the ²³⁸U and ²³²Th series, because of a migration of uranium in raw rock material or due to a separation during a production process.

The activities in tab. 1 are in the range of activity concentrations of natural radionuclides in the samples

of the same kind worldwide [3-11]. Also, these results do not differ significantly from the previously measured values for covering materials in Serbia [12-14].

Calculation

The results of the calculation of specific absorbed dose rates, q_U , q_{Th} , and q_K , in the centre of the room 5 m 4 m 2.8 m, originating from ²³⁸U, ²³²Th and ⁴⁰K in the materials of certain densities (ρ) and thicknesses (d), which approximate the usage parameters of most of the covering materials, are given in tab. 2. The table also lists the amounts of the reduction of specific absorbed doses of covered structural material – concrete, q_U , q_{Th} , and q_K , with respect to the values of q_U , q_{Th} , and q_K corresponding to the radiation of concrete in a standard room: $q_U = 0.757$, $q_{Th} = 0.912$, and $q_K = 0.0700$, in (nGy/h per Bq/kg) [35].

For different combinations of the thickness and density of the covering material in the interval d : 0.5-3 cm and ρ : 1.5-3 g/cm³, a simple fitting formula was derived in *Mathematica* for determination of the absorbed dose of gamma radiation in the air

$$D = \rho d [(0.0311A_U \quad 0.0371A_{Th} \quad 0.00271A_K)_{cov} \quad (0.0298A_U \quad 0.0326A_{Th} \quad 0.00225A_K)_{str}] \quad (4)$$

with D [nGyh⁻¹], for d expressed in cm and ρ in g/cm³. The coefficient of determination of particular elements of the fit, i. e., q_U , q_{Th} , q_K , q_U , q_{Th} , and q_K , has the value: $R^2 = 0.999$.

For very thin covering layers (glazes, varnishes, coatings) using parameters: $d = 0.1$ cm, $\rho = 1.25$ g/cm³, it was calculated: $q_U = 0.00417$, $q_{Th} = 0.00490$, and $q_K = 0.000368$ (nGy/h per Bq/kg), while the reduction of irradiation by the structural material is slight: $D_{str} = 1\%$. In addition, the dose [nGyh⁻¹] is preferably expressed through the mass of a material m [kg], used for covering the surface S [m²], so that

$$D_{ext} = \frac{m}{S} (0.00278A_U \quad 0.00327A_{Th} \quad 0.000245A_K)_{cov} \quad (5)$$

Table 3 shows the results of calculating the concentration of ²²²Rn in the room, C_{Rn} , exhaled from the covering material of thickness d and density ρ and the corresponding reduction, ΔC_{Rn} , for structural material (20 cm concrete), due to the presence of the cover

Table 3. Specific activity concentrations of ^{222}Rn [C_{Rn} (Bq/m³)/ A_{Ra} (Bq/kg)]_{cov}, for the covering materials, and associated reductions for structural material [ΔC_{Rn} (Bq/m³)/ A_{Ra} (Bq/kg)]_{str}

d [cm]; ρ [gcm ⁻³]	$[C_{\text{Rn}} \text{ (Bq/m}^3\text{)}/A_{\text{Ra}} \text{ (Bq/kg)}]_{\text{cov}}$	$[\Delta C_{\text{Rn}} \text{ (Bq/m}^3\text{)}/A_{\text{Ra}} \text{ (Bq/kg)}]_{\text{str}}$
0.7; 2.4	0.061	0.0228
1; 2.8	0.102	0.0326
2; 1.7	0.124	0.0659
3; 2.6	0.282	0.0991

Table 4. The ranges and the average values of absorbed gamma dose rate D , radon concentration C_{Rn} , the corresponding effective dose rates, as well as the total effective dose E per year, calculated for the explored covering materials. The error of approximately 10 % from measurement uncertainties are associated to all results. The values of doses for floor and walls (without the covering of the ceiling) are (26.5 – 2.3) % lower

Material species	$(D_{\text{Min}} - D_{\text{Max}}) / \bar{D}$ [nGyh ⁻¹]	$(E_{\text{Min}} - E_{\text{Max}})_{\text{ext}}$ [mSv]	$(C_{\text{RnMin}} - C_{\text{RnMax}}) / \bar{C}_{\text{Rn}}$ [Bqm ⁻³]	$(E_{\text{Min}} - E_{\text{Max}})_{\text{int}}$ [mSv]	$(E_{\text{Min}} - E_{\text{Max}}) / \bar{E}$ [mSv]
Ceramic tiles	(<2-7.82); 3.95	(<0.01-0.04); 0.02	(1.03-3.78); 2.07	(0.033-0.12); 0.065	(0.04-0.16); 0.085
Granite tiles	(2-10.3); 6.31	(0.01-0.05); 0.03	(2.12-8.35); 3.69	(0.067-0.26); 0.12	(0.08-0.31); 0.15
Granite	(<2-170); 29.3	(<0.01-0.83); 0.14	(<1-73.5); 14.9	(<0.01-2.31); 0.47	(<0.01-2.70); 0.61
Marble	<2	<0.01	<1	<0.01	<0.01
Travertine	<2	<0.01	<1	<0.01	<0.01
Perlite	14.7	0.072	4.88	0.15	0.22
Zircobit	14.5	0.071	13.2	0.42	0.49

layer. The data are given per unit activity concentration of ^{226}Ra in the overlay, or structural material. For very thin layers it was computed: $[C_{\text{Rn}} \text{ (Bq/m}^3\text{)}/A_{\text{Ra}} \text{ (Bq/kg)}]_{\text{cov}} = 0.00549$, while the effect of the covering of the structural material obtained was negligible. Still, on the basis of their composition and structure, a certain lowering of ^{222}Rn permeability is likely.

In calculating the indoor dose for each type of the explored materials, the appropriate conversion factors from tab. 2 and specific concentrations of radon (tab. 3) were applied, *i. e.*, those referring to: $d = 0.7 \text{ cm}$, $\rho = 2.4 \text{ g/cm}^3$, for ceramic tiles, $d = 1 \text{ cm}$, $\rho = 2.8 \text{ g/cm}^3$ – granite tiles and $d = 3 \text{ cm}$, $\rho = 2.6 \text{ g/cm}^3$, for granite, marble, and travertine. The dose corresponding to the radiation of perlite was estimated assuming its maximum presence (100 %) in the layer of mortar with thickness of 2 cm and density of 1.7 g/cm^3 , while zircobit was treated as the only component of the glaze layer.

To quantify the effect of covering to the reduction of the dose of structural material, the activity concentrations: $A_{\text{Ra}} = 35$, $A_{\text{Th}} = 30$, and $A_{\text{K}} = 400$ in Bq/kg in structural material were assumed, which are equal to the world average for ^{226}Ra , ^{232}Th , and ^{40}K content in soil. Namely, these activities produce the dose of uncovered structural material – concrete in the standard room: 0.40 mSv per year, for gamma exposure (using the conversion factors [35]) and 0.93 mSv per year, for irradiation by radon, that are close to the average level of indoor natural background [2, 36].

The minimum, the maximum and the average values of indoor absorbed gamma dose, ^{222}Rn concentration and associated effective doses, determined on

the basis of the activity concentrations of the radionuclides in the samples of covering materials (tab. 1), are presented in tab. 4. Because of the negligible irradiation effect, the doses lower than 10 Sv per year are not presented.

The lowest doses were calculated for marble, travertine, and granite sample. The maximum value of the total effective dose, 2.70 mSv per year, was obtained for granite Balmoral Red, which is the only one of the investigated materials that exceeds the dosimetry limit of 1 mSv per year by about 10 % (compared to the indoor background of 1.56 mSv per year). However, since it is the result corresponding to the covering of all the walls, floor and ceiling, in common usage of this and other materials (on the floor and walls) the dose is lower by about 27 %.

CONCLUSIONS

The indoor effective doses due to the gamma radiation of covering materials in Serbia, evaluated in this work, are lower than most of the results for the materials of the same species, found in the literature. In addition to the variations in the measured content of radionuclides, these differences are caused by different conversion factors used to calculate the absorbed dose of gamma radiation. Specifically, EC values of specific absorbed dose for ^{226}Ra , ^{232}Th , and ^{40}K [16], were computed for covering thickness of 3 cm and density of 2.6 g/cm^3 . In this paper, the dose was calculated for each sample by applying the conversion factors which are appropriate to the corresponding type of

the material. Variation from published internal dose data originate from differences in ^{226}Ra activity concentration in the samples and the model of dose evaluation.

Considering the covering of all the surfaces of the standard room, all of the investigated materials fulfil the dosimetric criterion of 1 mSv per year, except one sample of granite (Balmoral red). The average effective dose determined for ceramic tiles, granite tiles, and granite are: 0.085, 0.15, and 0.61 (mSv per year), respectively, while the irradiation by marble and travertine samples is minor, *i. e.*, less than 10 μSv per year. For application restricted to the floor and walls, the dose values are about 27 % lower than those shown in the paper. Lowering of dose should be also taken into account when materials take part only as a component in covering material, for example, in the case of mortar and glaze.

ACKNOWLEDGEMENT

This work was supported by the Ministry of Education, Science and Technological Development of the Republic of Serbia; projects number 171021 and 171025.

AUTHORS' CONTRIBUTIONS

The experimental work was carried out by G. M. Manić. The computation of the specific absorbed dose for ^{226}Ra , ^{232}Th , and ^{40}K by means of the Mathematica 8 software, was done by V. M. Manić. All the authors were involved in preparation of the text of the manuscript, analysis and discussion of the presented results.

REFERENCES

- [1] ***, International Commission on Radiological Protection, The 2007 Recommendations of the Commission on Radiological Protection, ICRP Publication 103, Annals of ICRP, 2007
- [2] ***, Sources and Effects of Ionizing Radiation United Nations Scientific Committee on Effects of Atomic Radiation, 2008 Report, United Nations, New York, 2008
- [3] Kumar, A., *et al.*, Natural Activities of ^{238}U , ^{232}Th and ^{40}K in Some Indian Building Materials, *Radiat. Measur.*, 36 (2003), 1-6, pp. 465-469
- [4] Tzortzis, M., *et al.*, Gamma Radiation Measurements and Dose Rates in Commercially-Used Natural Tiling Rocks (Granites), *J. Environ. Radioact.*, 70 (2003), 3, pp. 223-235
- [5] Pavlidou, S., *et al.*, Natural Radioactivity of Granites Used as Building Materials, *J. Environ. Radioact.*, 89 (2006), 1, pp. 48-60
- [6] Sahoo, B. K., *et al.*, Estimation of Radon Emanation Factor in Indian Building Materials, *Radiat. Measur.* 42 (2007), 8, pp. 1422-1425
- [7] Medhat, M. E., Assessment of Radiation Hazards Due to Natural Radioactivity in Some Building Materials Used in Egyptian Dwellings, *Radiat. Prot. Dosim.*, 133 (2009), 3, pp. 177-185
- [8] Righi, S., *et al.*, Natural Radioactivity in Italian Ceramic Tiles, *Radioprotection*, 44 (2009), 5, pp. 413-419
- [9] Kovler, K., *et al.*, Natural Radionuclides in Building Materials Available in Israel, *Build. Environ.*, 37 (2002), 5, pp. 531-537
- [10] Moharram, B. M., *et al.*, External Exposure Doses Due to Gamma Emitting Natural Radionuclides in Some Egyptian Building Materials, *Appl. Radiat. Isot.*, 70 (2012), 1, pp. 241-248
- [11] Turhan, S., Baykan, U. N., Sen, K., Measurement of the Natural Radioactivity in Building Materials Used in Ankara and Assessment of External Doses, *J. Radiol. Prot.*, 28 (2008), 1, pp. 83-91
- [12] Popović, D., Todorović, D., Radon Indoor Concentrations and Activity of Radionuclides in Building Materials in Serbia. Facta Universitatis, Series: Physics, Chemistry and Technology, 4 (2006), 1, pp. 11-20
- [13] Krstić, D., *et al.*, Radioactivity of Some Domestic and Imported Building Materials from South Eastern Europe, *Radiat. Measur.*, 42 (2007), 10, pp. 1731-1736
- [14] Ujić, P., *et al.*, Internal Exposure from Building Materials Exhaling ^{222}Rn and ^{220}Rn as Compared to External Exposure Due to Their Natural Radioactivity Content, *Appl. Radiat. Isot.*, 68 (2010), 1, pp. 201-206
- [15] Markkanen, M., Radiation Dose Assessments for Materials with Elevated Natural Radioactivity, Finnish Centre for Radiation and Nuclear Safety, Helsinki, Report STUK-B-STO 32, 1995
- [16] ***, EC, European Commission Radiological Protection Principles Concerning the Natural Radioactivity of Building Materials, Radiation Protection 112, Office for Official Publications of the European Communities, Luxembourg, 1999
- [17] ***, Report: Sources to Effects Assessment for Radon in Homes and Workplaces, UNSCEAR, 2006
- [18] Nero, A. V., Nazaroff, W. W., Characterising the Source of Radon Indoors, *Radiat. Prot. Dosim.*, 7 (1984), 1-4, pp. 23-29
- [19] Koblinger, L., Calculation of Exposure Rates from Gamma Sources in Walls of Dwelling Rooms, *Health Phys.*, 34 (1978), 5, pp. 459-463
- [20] Hubbell, J. H., Photon Cross-Sections, Attenuation Coefficients, and Energy Absorption Coefficients from 10 keV to 100 GeV, NSRDS-NBS 29, National Bureau of Standards, Washington DC, USA, 1969
- [21] Harima, Y., *et al.*, Development of New Gamma-Ray Buildup Factor and Application to Shielding Calculations, *J. Nucl. Sci. Technol.*, 28 (1991), 1, pp. 74-84
- [22] Goldstein, H., Fundamental Aspects of Reactor Shielding, Addison-Wesley Publishing Co 1959, Reprinted by Johnson Reprint Corp., 1971
- [23] Maduar, M. F., Hiromoto, G., Evaluation of Indoor Gamma Radiation Dose in Dwellings, *Radiat. Prot. Dosim.*, 111 (2004), 2, pp. 221-228
- [24] ***, Regulatory Guide 3.64 (Task WM 503-4) Calculation of Radon Flux Attenuation by Earthen Uranium Mill Tailings Covers, U. S. Nuclear Regulatory Commission NUREG, 1989
- [25] Haquin, G., Natural Radioactivity and Radon in Building Material, *Proceedings*, 12th Int. Congress of the Int. Rad. Prot. Assn., Buenos Aires, October 19-24, 2008, pp. 3187-3200
- [26] Keller, G., Hoffmann, B., The Radon Diffusion Length as a Criterion for the Radon Tightness, 10th In-

- ternational Congress of the International Radiation Protection, Hiroshima, Japan, 2000
- [27] Stoulos, S., Manolopoulou, M., Papastefanou, C., Assessment of Natural Radiation Exposure and Radon Exhalation from Building Materials in Greece, *J. Environ. Radioact.*, 69 (2003), 3, pp. 225-240
- [28] Turtiainen, T., *et al.*, Collective Effective Dose Received by the Population of Egypt from Building Materials, *J. Radiol. Prot.*, 28 (2008), 2, pp. 223-232
- [29] Berger, M. J., *et al.*, XCOM: Photon Cross Section Database (version 1.5). Available: <http://physics.nist.gov/xcom>. National Institute of Standards and Technology, Gaithersburg, Md., USA, 2010
- [30] Yoshida, Y., Development of Fitting Methods Using Geometric Progression Formulae of Gamma-Ray Buildup Factors, *J. Nucl. Sci. Technol.*, 43 (2006), 12, pp. 1446-1457
- [31] ***, Gamma-Ray Attenuation Coefficients and Buildup Factors for Engineering Materials, ANSI/ANS-6.4.3-1991, American National Standards Institute, American Nuclear Society, La Grange Park, Ill., USA, 1991
- [32] Singh, S., *et al.*, Buildup of Gamma Ray Photons in Flyash Concretes: A Study, *Ann. Nucl. Energy*, 37 (2010), 5, pp. 681-684
- [33] ***, International Atomic Energy Agency Guidelines for Radioelement Mapping Using Gamma Ray Spectrometry Data, Vienna, IAEA-TECDOC-1363, 2003
- [34] Mann, K. S., *et al.*, Investigations of Mass Attenuation Coefficients and Exposure Buildup Factors of Some Low-Z Building Materials, *Ann. Nucl. Energy* 43 (2012), pp. 157-166
- [35] Manić, V., *et al.*, Calculation of Dose Rate Conversion Factors for ^{238}U , ^{232}Th and ^{40}K in Concrete Structures of Various Dimensions, with Application to Niš, Serbia, *Radiat. Prot. Dosim.*, 152 (2012), 4, pp. 361-368
- [36] ***, Report: Sources, Effects and Risks of Ionizing Radiation United Nations Scientific Committee on Effects of Atomic Radiation, 2000 Report, UNSCEAR, United Nations, New York, 2000
- Received on August 27, 2015
Accepted on November 27, 2015

Весна М. МАНИЋ, Горан Ј. МАНИЋ, Драгослав Р. НИКЕЗИЋ, Драгана Ж. КРСТИЋ

**ДОЗЕ ОД РАДИОАКТИВНОСТИ ГРАЂЕВИНСКИХ МАТЕРИЈАЛА КОЈИ СЕ
КОРИСТЕ ЗА ПОКРИВАЊЕ ЗГРАДА У СРБИЈИ**

Доза зрачења унутар просторија потиче углавном од керамичких и гранитних плочица, мермера, гранита, као и неких компоненти у материјалима за облагање, произведеним у Србији или увезеним из других земаља. Активносне концентрације ^{226}Ra , ^{232}Th , и ^{40}K мерене су стандардном гама спектрометријском методом помоћу германијумског полупроводничког HPGe детектора. Јачина апсорбоване дозе у ваздуху услед гама зрачења ових радионуклида одређена је помоћу специфичне јачине дозе рачунате за керамику. На основу садржаја ^{226}Ra рачуната је концентрација ^{222}Rn унутар просторије. Процењена је средња годишња ефективна доза за поједине материјале.

Кључне речи: доза, густина активности, природни радионуклид, материјал за облагање