

NATURAL RADIOACTIVITY LEVEL IN MATERIALS USED FOR MEDIEVAL VAULTING IN THE TERRITORY OF THE CENTRAL BALKAN REGION

by

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This work presents the results of an investigation undertaken to determine the level of natural radioactivity in the traditional building materials used for medieval indoor vaulted constructions in the territory of the central Balkan region. Indoor radiation exposure varies appreciably if it comes from the earth building materials, hence the presence of natural radioisotopes of ²²⁶Ra, ²³²Th, and ⁴⁰K in masonry vaulted constructions was analyzed using gamma ray spectrometry. In addition, the internal health hazard index, the absorbed dose rates and the effective annual doses were calculated. The results were then compared both with the reported data from the previous studies concerning the territory of the Balkan Peninsula, as well as with the worldwide values for the materials of historic buildings. The results obtained from the materials examined in this paper all showed the radioactivity levels below the maximum permitted values.

Key words: natural radioactivity, building material, medieval vault, Balkan, gamma ray spectrometry

INTRODUCTION

The use of earth materials is widespread within the traditional Balkan architecture. As most of these materials have been recognized as ecologically friendly, their role is increasing also in modern architecture, which tends to be in equilibrium with energy efficiency. The implementation of traditional materials in the old buildings (of which a large number of examples survives in the old Balkan architecture) is extremely important for the existence of the traditional vaulted structures. Unlike façade walls, vaulted structures do not have a contact with the outside air due to roof covering constructions.

Traditional building materials (of natural origin) reflect the geological composition they are derived from. Most of building materials are of a natural origin containing varying amounts of naturally occurring radionuclides. Therefore, building structures like masonry vaults that are made of earthen materials can contain high levels of natural radioisotopes concentration. So far several studies dealt with concentration of

naturally occurring radionuclides in the composition of modern commercial building materials in the Balkan region, among which the most important are presented in [1-5].

The average worldwide concentrations of naturally occurring radionuclides (²²⁶Ra, ²³²Th, and ⁴⁰K) for the building materials are 50, 50, and 500 Bq/kg, respectively [6]. The exposure to gamma rays in the interior of buildings (indoor exposure) is mainly determined by materials of construction. Radium and its radioactive progenies are present in almost all buildings. According to the statements of the UN Scientific Committee on the Effects of Atomic Radiation the indoor exposure to gamma rays is inherently greater than outdoor exposure if earth materials have been used [7].

Indoor radiation exposures depend, among other things, on the user occupancy. According to UNSCEAR Report and European Commission Report, the radiation exposure inside buildings (*i. e.*, the air dose absorption of gamma radiation from the ground) is roughly 40 % higher than that of the exterior environment [7]. Due to these facts, there is a greater probability of radioactive materials presence in the vaulted structures, at least higher than that in the facade walls. However, traditional

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building materials have not been subjected to any systematic examination. Therefore, there is an evident need to investigate the radioactive isotopes activity in building materials of vaulted structures.

The regulations for protection of vaulted structures of the medieval monuments forbid the use of concrete during the restoration, but only the use of original natural materials. It is therefore very important to determine the concentration of radioisotopes in the vaulted structures, which are usually integral parts of the medieval interiors.

The aim of this paper was to determine the activity concentration of radioisotopes, their internal health hazard index, the absorbed dose rates and annual effective doses in certain earth materials of the vaulted structures in order to define the benefits of their use for the modern construction sector.

MATERIALS AND METHODS

Building materials from a large number of medieval monuments were analyzed for radioisotope presence in their chemical composition. These materials are the best for this kind of investigation as they have been integral parts of the vaulted structures for centuries. All examined materials originate from the arches and vaults of medieval monasteries and cities on the territory of central Balkan region, namely, from the monastery of Studenica (XII century), the monastery of Banjska (XIV century), the monastery of Gradac (XIII century), and the medieval towns of Kotor (XII century) and Novo Brdo (XIV century). The examined building materials included brick, carbonate tufa, breccia and limestone.

Four different kinds of building materials commonly used in medieval vaulted constructions (carbonate tufa, brick, limestone, and breccia) were examined for their natural radioactivity concentrations, using γ -ray spectrometry. Measurements of activity concentrations of radioisotopes were performed at the Laboratory of Radiation and Environmental Protection at the Vinča Institute for Nuclear Sciences in Belgrade. Eight samples of four different vaulting materials (originating from different monasteries) were subjected to natural ventilation and dried, and then crushed. The examined samples were packed in to the cylindrical plastic containers (of 125 ml and 250 ml of volume and in larger "Marinelli" beakers of 450 ml), which were then sealed. The measurement geometry depended on the available quantity of the samples.

Gamma spectrometry was performed with the Canberra HPGe (high purity germanium) detectors D-2 and D-3 with relative efficiency of 23 % and 50 %, respectively and with an energy resolution of 1.8 keV at 1332 keV of ^{60}Co . The measurement time for samples was set to 60000 s. Computer software GENIE2000 was used to record and analyse obtained spectrum. Back-

ground value was obtained immediately before the measurements of radioisotope activity of the samples. The measurement uncertainty of each procedure did not exceed 10% [8-11].

For ^{226}Ra activity concentration, the activity of the photopeak of ^{214}Pb (at energies of 295 keV and 352 keV) and ^{214}Bi (at 609 keV, 1120 keV, and 1764 keV) were used. To measure ^{232}Th , photopeaks from ^{228}Ac (at the energies of 338 keV and 911 keV) were used. The concentration of potassium ^{40}K activity is directly determined by using the photopeak at the energy of 1460 keV. Activity of ^{235}U was measured from its intensive line at the energy of 186 keV with the correction values for ^{226}Ra (at the energy of 186 keV). Activity of ^{238}U is determined by the presence of his progenies of ^{234}Th (at 63 keV) and ^{234}Pa (at 1000 keV).

The European Commission set guidelines on the protection concerning the natural radioactivity of building materials (RP-112 document) for the EU Member States [12]. Public doses in the member states should be at the lowest possible level. Within this set of principles, the control on the radioactivity of building materials is defined as well. Control is based on the dose criterion which is defined as the excess exposure caused by building materials. Often, it is determined considering the general national circumstances of one member state. Within the European Union, doses exceeding 1 mSv per year should be treated from the radiological protection point of view. Activity Index or the gamma-index, I , is determined by regulatory standards [12]. Gamma-index is defined as the activity of natural radioisotopes which are present in a certain building materials; primarily it is defined by the presence of ^{226}Ra , ^{232}Th , and ^{40}K . For the interior of the building, the gamma index is determined according to the expression

$$I_{\gamma} = \frac{C_{\text{Ra}}}{300 \text{ Bq/kg}} + \frac{C_{\text{Th}}}{200 \text{ Bq/kg}} + \frac{C_{\text{K}}}{300 \text{ Bq/kg}} \quad (1)$$

where C_{Ra} , C_{Th} , and C_{K} are the activity concentrations of radium, thorium, and potassium, respectively (in the unit of Bq/kg) in building materials [7]. Gamma index can only be used as a monitoring tool for identifying materials that can be used as building materials.

In relation to the exemption doses of 0.3 mSv per year and of 1 mSv per year, the gamma-index should not exceed the limit values given by the principles which are suggested by the European Commission, presented in tab. 1 [12, 13].

Excess alpha radiation is caused by the inhalation of radioisotopes originating from building materials.

Table 1. Limits of the activity concentrations index [12]

Dose criterion	0.3 mSv per year	1 mSv per year
Materials used in bulk amounts	$I \leq 0.5$	$I \leq 1$
Superficial and other materials	$I \leq 2$	$I \leq 6$

Radiation hazards for respiratory organs due to influence of the excess alpha radiation in the indoor space of one building are determined by the internal health hazard index. The value of the internal health hazard index is calculated according to the expression [14]

$$H_{in} = \frac{C_{Ra}}{185 \text{ Bq/kg}} + \frac{C_{Th}}{259 \text{ Bq/kg}} + \frac{C_K}{4810 \text{ Bq/kg}} \quad (2)$$

where C_{Ra} , C_{Th} , and C_K are the activity concentrations of radium, thorium, and potassium in building materials, respectively, in Bq/kg. Value of internal hazard index must be less than the unity for building materials.

An estimation of indoor absorbed dose rate due to gamma-ray emission in the building materials was obtained from the known concentrations of natural radioisotopes ^{226}Ra , ^{232}Th , and ^{40}K . The value of the specific indoor absorbed dose rate is expressed in Gy/h, and it is calculated using the equation proposed by [15]

$$D_R = 0.92A_{Ra} + 1.10A_{Th} + 0.08A_K \quad (3)$$

where A_{Ra} , A_{Th} , and A_K are the activity concentrations of natural radioisotopes in building materials.

The results calculated for the estimation of the absorbed dose rates are used for the calculation of the annual effective dose, which is obtained by using the following expression [15]

$$E_{eff} = D_R \cdot 0.7 \text{ Sv/Gy (conversion factor)} \cdot 7012.7 \text{ h} \cdot 10^{-6} \quad (4)$$

In the eq. (4), a value of 0.7 was used as the conversion factor from absorbed dose received by adults, while the value of 7012.7 h is determined as the annual exposure time (which means that 80 % of time is spent indoors) [15].

RESULTS AND DISCUSSION

The presence of natural radioisotopes ^{210}Pb , ^{238}U , and ^{235}U , ^{226}Ra , ^{232}Th , and ^{40}K was most noticeable in material samples which come from vaults and arches of medieval churches and fortifications, while

the activity concentration of man-made isotope ^{137}Cs is negligible. Obtained radioisotopes activity concentration along with the gamma index value is shown in the tab. 2.

The concentration of radioactive substances in the examined building materials corresponds to their concentration in the soil, *i. e.*, to the geological composition of the soil from which they derive. Certain small concentrations of the man-made radioisotope ^{137}Cs were registered in the samples which are caused by the exposure of certain samples of vaults and arches to the atmospheric influences for several decades. It is obvious that the small presence of the radioisotope ^{137}Cs in these samples is a result of the Chernobyl accident to the total radioactivity in the atmosphere above these geographic locations, as is the case at the other historic monuments in the world [16].

The average values of activity concentration for ^{226}Ra , ^{232}Th , and ^{40}K in the building materials which are used in the world should not exceed the values 50 Bq/kg (for radium), 50 Bq/kg (for thorium), and 500 Bq/kg (for potassium) [17]. By comparing these values with the results shown in tab. 2, it can be concluded that the concentration of radioisotopes in medieval materials does not exceed the average values prescribed by the recommendations of UNSCEAR, except when it comes to the concentration of ^{40}K in the medieval bricks. The presence of potassium ^{40}K does not represent any possible radiological risks to human health because the concentration of ^{40}K is in a homeostatic balance with other isotopes of potassium in nature. By comparing the value of gamma-index with recommended values, it can be seen from tab. 2, that all the analyzed materials meet the criteria of the recommended annual doses so they can be used as building materials in present time.

The comparison of the activity concentrations for the radioisotopes obtained in this paper and concentrations in the modern building materials which are applied throughout the Balkans is of specific importance. The comparison of activity concentrations in medieval and modern brick products shows the equal values for the concentration of natural radioisotopes ^{226}Ra , ^{232}Th , and ^{40}K [2, 16]. Comparing the radioisotope

Table 2. Activity concentration of the radioisotopes in the arch and vaults samples of the medieval churches and urban agglomerations in the territory of central Balkan region

	Material	Location	Activity concentration of the radioisotopes [Bqkg ⁻¹]									Gama-index I_γ
			²¹⁰ Pb	²³⁸ U	²³⁵ U (²³⁵ U/ ²³⁸ U)		²²⁶ Ra	²³² Th	⁴⁰ K	¹³⁷ Cs		
1	Brick	Banjaska	23 5	44 5	2.1	0.2 (0.048)	37 3	49 4	800 50	<0.08	0.635	
2	Carbonate tufa	Banjaska	4 1	5 1	0.22	0.06 (0.047)	2.3 0.5	1.7 0.4	31 3	<0.07	0.027	
3	Breccia	Novo Brdo	<8	27 7	1.4	0.2 (0.053)	22 2	33 3	460 30	<0.06	0.392	
4	Brick	Novo Brdo	36 5	45 5	2.0	0.2 (0.045)	42 3	40 3	840 50	<0.08	0.62	
5	Carbonate tufa	Studenica	19 5	7 2	0.31	0.05 (0.041)	1.9 0.4	1.7 0.4	15 2	0.18 0.06	0.02	
6	Brick	Studenica	31 6	37 8	1.6	0.3 (0.042)	36 4	42 4	540 40	<0.1	0.51	
7	Limestone	Kotor	<8	<4	<0.1		2.4 0.4	2.4 0.4	25 3	<0.06	0.03	
8	Carbonate tufa	Gradac	5 1	6 1	0.28	0.04 (0.047)	6.4 0.7	<0.4	7 1	0.32 0.09	0.314	

tope activity concentrations in stone used for the constructions of medieval arches and vaults to the concentration in stone used in the contemporary architecture [4, 16], shows that the activity concentration of ^{226}Ra , ^{232}Th , and ^{40}K in the medieval ages was significantly lower. The choice of a carbonate tufa and limestone for the constructions of arch and vault contributed to the reduced concentration of radioisotopes in the Middle Ages in relation to the contemporary vaulted surfaces where marble and granite are commonly used for this kind of constructions. In addition, all the medieval materials examined in this paper showed equal or lower concentration of radioisotope activity than is the case with the materials used for contemporary vaulted structures [3, 18].

Calculated results for the internal health hazard indexes of separate materials, as well as for the absorbed dose rates and for the annual effective dose rates are given in tab. 3, while in tab. 4, the activity concentration of the radioisotopes in the contemporary building materials is presented.

According to the results given in tab. 3, it can be seen that the values of the internal hazard index in medieval materials do not exceed the value of 1, so that all these materials can be used in the contemporary architecture as building materials. According to the UNSCEAR2000 report, the corresponding worldwide average values for the absorbed gamma dose rate in the interior of buildings range from 20 to 200 nGy/h with a population-weighted average of 84 nGy/h. By comparing these values with the results shown in tab. 4, it can be

seen that the values of the absorbed dose rates in the medieval materials do not exceed the recommended upper limit values given by the UNSCEAR report and European Commission report. The maximum values were obtained for the materials that are most similar to the chemical composition of the soil – as in the case of the bricks from the monasteries of Studenica, Banjska and the town of Novo Brdo, as well as with the breccias from Novo Brdo. High levels of the absorbed dose rates for these materials are caused by high concentrations of natural radioisotope ^{40}K in their composition.

The average values of the annual effective dose rates in most of medieval material are significantly lower than a value of 1 mSv which represents the upper limit for the basic materials used in building construction. This circumstance is particularly related to those types of stone that were used for the construction of arches and vaults. Higher levels of annual effective doses are recorded in the case of bricks from the monasteries of Studenica and Banjska and Novo Brdo as well as in breccia from Novo Brdo, which is again caused by the concentration of ^{40}K in their chemical composition.

The relationship between the activity concentration values and the gamma indexes of radioisotopes from the materials analyzed in this paper and the material samples from other historical sites in the world [16, 20] is as it has been expected. This comparison confirms that the activity concentrations of the natural radioisotopes in the traditional building are within the range or below the average values that can be registered in the contemporary worldwide building materials.

Table 3. The internal hazard index, the absorbed dose rates and the annual effective doses

	Material	Location	H_{in}	D_R [nGyh ⁻¹]	E_{eff} [mSv]
1	Brick	Banjska	0.555509	151.94	0.745857
2	Carbonate tufa	Banjska	0.025441	6.466	0.031741
3	Breccia	Novo Brdo	0.341966	93.34	0.458196
4	Brick	Novo Brdo	0.556103	149.84	0.735548
5	Carbonate tufa	Studenica	0.019952	4.818	0.023651
6	Brick	Studenica	0.469023	122.52	0.601437
7	Limestone	Kotor	0.027437	6.848	0.033616
8	Carbonate tufa	Gradac	0.037594	6.888	0.033812

Table 4. The activity concentration of the radioisotopes in the contemporary building materials for the constructions of the vaults and arches in the territory of the Balkan region

	Material	The country of origin	Activity concentration of the radioisotopes [Bqkg ⁻¹]					Ref.
			^{238}U	^{235}U	^{226}Ra	^{232}Th	^{40}K	
1	Brick	Greece	35 11	–	–	45 15	710 165	[17]
2	Ceramic elements	Greece	–	–	46 2	1.7 0.4	31 3	[2]
3	Marble	Macedonia	–	–	46 2	32 1	945 50	[2]
4	Marble	Greece	–	–	63 3	32 2	770 20	[2]
5	Marble	Bulgaria	–	–	26.1 0.2	24 1	1400 70	[2]
6	Limestone	Turkey	–	–	20.2	5.0	55.0	[4]
7	Limestone	Greece	–	–	14 6	1.6 0.5	–	[3]
8	Pumice stone	Greece	up to 715	–	50-874	54-60	1048-1158	[19]

CONCLUSIONS

Eight medieval building material samples, used for the vaulted structures of churches and for tifications, were examined in the paper. Activity concentrations in the analyzed materials do not exceed the world recommended average values, except in the case of ^{40}K , the concentration of which is in the homeostatic balance with other isotopes of potassium in the living organisms. The man-made radioisotope ^{137}Cs concentration is negligible in the results. The maximum values were registered for the materials which are most similar to the chemical composition of the soil from which they originate (which is especially the case with the medieval bricks and breccias from Novo Brdo), and which is also the consequence of high concentration of natural radioisotope ^{40}K . The minimum values for the above mentioned samples were recorded in the case of compact limestone from Kotor.

The calculated activity concentration index values for all the examined traditional material samples were lower than or equal to the recommended exception limits for exposure to indoor γ -radiation. It was concluded that all materials which had been used in the construction of arches and vaults inside the medieval objects do not pose a significant radiological hazard when used in modern vaulted constructions. Therefore, the traditional materials can be applied with a high degree of certainty even in the contemporary architecture. The maximum values of the internal hazard indexes, the intensity of the absorbed dose rates and of the annual effective doses do not exceed the recommended exception limits specified by the international UNSCEAR regulations.

Based on the analysis, it can be concluded that in comparison to the modern building materials, the traditional materials reduce the dose rate values that people receive. At the same time, the materials used for arches and vaults in medieval architecture can still be used in the contemporary architectural design and the restoration of older buildings in order to improve the environmentally sustainable architecture.

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AUTHORS' CONTRIBUTIONS

All authors performed theoretical analysis and literature research. Experiments were carried out by I. S. Bjelić, D. J. Todorović, and J. D. Krneta-Nikolić. The tables were prepared by I. S. Bjelić. All authors analyzed and discussed the results. The manuscript was written by all authors.

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**НИВО ПРИРОДНЕ РАДИОАКТИВНОСТИ У МАТЕРИЈАЛИМА
КОРИШЋЕНИМ ЗА ПРЕСВОЂАВАЊЕ СРЕДЊОВЕКОВНИХ
СПОМЕНИКА НА ТЕРИТОРИЈИ ЦЕНТРАЛНОГ БАЛКАНА**

У овом раду приказани су резултати испитивања нивоа природне радиоактивности у традиционалним грађевинским материјалима коришћеним за пресвођавање средњовековних манастира на територији централног Балкана. Излагање зрачењу у затвореном простору доста варира ако потиче из земљаних грађевинских материјала, тако да је у раду применом гама спектрометрије анализирано присуство природних радиоиотопа ^{226}Ra , ^{232}Th и ^{40}K у конструкцијама за пресвођавање. Поред тога, испитани су и унутрашњи хазард индекс, апсорбована доза и годишња ефективна доза. Добијени резултати упоређени су са подацима о материјалима историјских споменика територије централног Балкана и света. На основу тога изведени су закључци о нивоу радиоактивности у старим и модерним грађевинским материјалима централног Балкана.

Кључне речи: природна радиоактивност, грађевински материјал, средњовековни свод, Балкан, гама спектрометрија