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## 1. INTRODUCTION

Building materials are materials which may contain to some extent naturally occurring radioisotopes <sup>226</sup>Ra, <sup>232</sup>Th (from uranium and thorium decay chain) and <sup>40</sup>K. Those materials are often referred to as NORM (Naturally Occurring Radioactive Materials) [1, 2]. Using NORM in buildings can present a potential health risk to the population, caused by gamma radiation and indoor radon (<sup>222</sup>Rn), leading to the necessary monitoring of radioactivity in used building materials [3]. Averaged values of activity concentration of <sup>226</sup>Ra, <sup>232</sup>Th and <sup>40</sup>K in building materials worldwide are 50 Bq kg<sup>-1</sup>, 50 Bq kg<sup>-1</sup> and 500 Bq kg<sup>-1</sup>, respectively [4]. In this paper, the level of radioactivity of some ceramic tiles produced in Serbia by two major manufacturers was examined. Ceramic tiles in Serbia are mainly used for covering floor and wall surfaces in domestics, as well in residential places.

According to the regulation in Republic Serbia allowed level of public exposure originated from building materials, as from the other sources, is 1 mSv on annual level [5]. Regulation in Republic Serbia is in consistency with one in the European Union [6].

The main goals of the paper were measuring the level of radioactivity of some ceramic tiles produced in Serbia and estimation of radiological, apropos health risk caused from the usage of these materials in domestic and residential spaces.

## 2. MATERIALS AND METHODS

### 2.1 Sample preparation

In order to analyze samples by gamma-spectrometric method, all the samples were crushed and homogenized in ball mill (Fig. 1a) around 45 minutes, after which they were sifting through the sieve granularity of 1.6 mm (Fig. 1b) and packed into plastic containers dimension diameter 67 mm and height 62 mm (Fig. 1c). Samples were taped with teflon tape to prevent diffusion of radon from the containers (Fig. 1d). Measurements started one month after sampling, after reaching the secular radioactive equilibrium between <sup>222</sup>Rn and <sup>226</sup>Ra. Samples were prepared at the Laboratory for sample preparation at the Faculty of Technology in Novi Sad. Typical weight of prepared sample was 220 g.

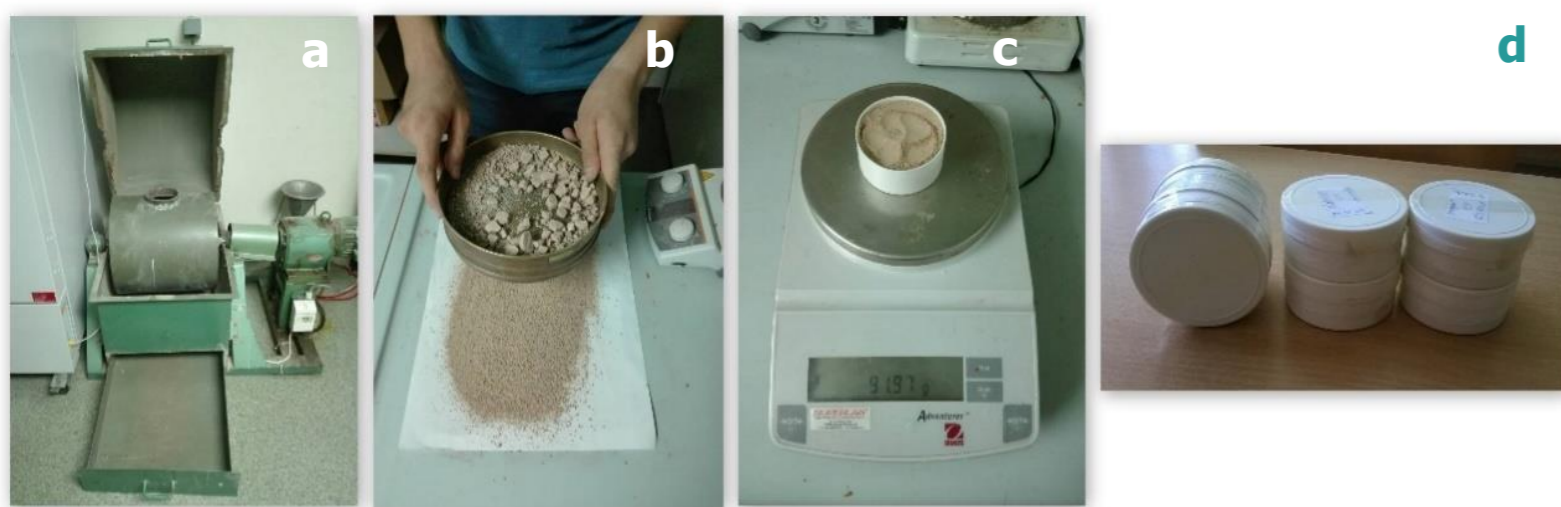


Fig. 1. Grinding of samples with a ball mill.



Fig. 2. ORTEC gamma spectrometry system for measuring radioactivity.

### 2.2 Measuring level of radioactivity

Activity concentration of <sup>226</sup>Ra, <sup>232</sup>Th and <sup>40</sup>K were determinate using method of low-level gamma spectrometry. The typical measurement time of samples was 72 000 s.

Activity concentration of <sup>226</sup>Ra was determinate from the gamma line of <sup>214</sup>Pb at 609.3 keV and <sup>214</sup>Pb at 351.9 keV, while for activity concentration of <sup>232</sup>Th gamma lines of <sup>228</sup>Ac at 911.2 and 969.1 keV and of <sup>212</sup>Pb at 238.6 keV were used. For determination of activity concentration of <sup>40</sup>K gamma line at 1460.8 keV was used. All the concentrations were measured by HPGe ORTEC GMX gamma spectrometer (Fig. 2), with nominal efficiency of 32% and energy resolution of 1.9 keV. HPGe detector was surrounded with 12-cm thick Pb-shield. Acquisition and analyzing of spectra were performed using Canberra Genie 2000 programme. Measurement uncertainties were given with a confidence level of 95% [8].

### 2.3 Radiation risk assessment

#### 2.3.1 Gamma index ( $I_\gamma$ ) [6]:

$$I_\gamma = \frac{C_{Ra}}{300 \text{ Bq kg}^{-1}} + \frac{C_{Th}}{200 \text{ Bq kg}^{-1}} + \frac{C_K}{3000 \text{ Bq kg}^{-1}} \quad (1)$$

#### 2.3.2 Radium equivalent activity index ( $Ra_{eq}$ ) [9-11]:

$$Ra_{eq} = C_{Ra} + 1.43C_{Th} + 0.077C_K \quad (2)$$

#### 2.3.3 Absorbed dose rate ( $D$ ) [6, 12,13]:

$$D(\text{nGy h}^{-1}) = 0.12 \cdot C_{Ra} + 0.14 \cdot C_{Th} + 0.0096 \cdot C_K \quad (3)$$

#### 2.3.4 Annual effective dose ( $E$ ) [6, 11, 12]:

$$E(\text{mSv y}^{-1}) = 0.7 \text{ Sv Gy}^{-1} \times 7000 (\text{or } 2000)h \times D \quad (4)$$

## 3. RESULTS AND DISCUSSION

Table 1 Type, manufacturer, color and activity concentration of <sup>226</sup>Ra, <sup>232</sup>Th and <sup>40</sup>K for ceramic tiles produced in Serbia.

Sample No.	Type	Producer	Color	Sample size (cm)	Activity concentration (Bq kg <sup>-1</sup> )		
					<sup>226</sup> Ra	<sup>232</sup> Th	<sup>40</sup> K
1	Floor	Zorka Keramika	Green	33 x 33	64.6±0.8	57.0±1.8	780±30
2	Floor	Zorka Keramika	Blue	33 x 33	65.3±1.0	69.1±1.9	710±30
3	Wall	Zorka Keramika	Beige	40 x 25	56.8±0.8	43.6±1.5	790±40
4	Floor	Zorka Keramika	Dark orange	33 x 33	96.8±1.5	69.8±1.9	1020±40
5	Wall	Toza Marković	Orange	33 x 25	52.3±0.6	47.5±1.6	740±30
Mean values ± SD					67.2±6.9	57.4±4.7	808±48
Average values in the world					50 <sup>1</sup>	50 <sup>1</sup>	500 <sup>1</sup>

<sup>1</sup>given in Ref. [4].

Table 2 Gamma index ( $I_\gamma$ ), radium equivalent activity index ( $Ra_{eq}$ ), absorbed gamma dose rate ( $D$ ) and annual effective dose ( $E$ ).

Sample No.	$I_\gamma$	$Ra_{eq}$ (Bq kg <sup>-1</sup> )	$D$ (nGy h <sup>-1</sup> )	80% time	20% time
				$E$ (μSv y <sup>-1</sup> )	$E$ (μSv y <sup>-1</sup> )
1	0.760±0.014	206±4	23.2±0.4	114±2	32.5±0.6
2	0.800±0.014	219±4	24.3±0.4	119±2	34.1±0.6
3	0.671±0.016	180±4	20.5±0.5	100±2	28.7±0.6
4	1.012±0.017	275±4	31.2±0.5	153±3	43.7±0.7
5	0.659±0.013	177±3	20.0±0.4	98±2	28.0±0.5
Mean values ± SD				117±9	34±2
Average or recommended values				≤1 <sup>2</sup>	370 <sup>3</sup>
				55 <sup>4</sup>	1000 <sup>5</sup>

<sup>2</sup>given in Ref. [6]; <sup>3</sup>given in Ref. [11]; <sup>4</sup>given in Ref. [13]; <sup>5</sup> given in Ref. [5].

Table 3 Activity concentration of <sup>226</sup>Ra, <sup>232</sup>Th and <sup>40</sup>K from earlier studies of the level of radioactivity of ceramic tiles in Serbia. – range, (mean values ± standard deviation).

<sup>226</sup> Ra	Activity concentration (Bq kg <sup>-1</sup> )		Reference
	<sup>232</sup> Th	<sup>40</sup> K	
43-75	49-65	556-729	[14]
93-133 (110±21)	66-83 (77±10)	883-1,053 (940±110)	[15]

## CONCLUSION

Building materials may contain a certain level of radioisotopes from uranium and thorium decay chain (<sup>226</sup>Ra and <sup>232</sup>Th), as well as primordial radioisotope <sup>40</sup>K. In this paper, the level of radioactivity for some ceramic tiles produced in Serbia by two leading manufacturers was analyzed. Activity concentration of <sup>226</sup>Ra, <sup>232</sup>Th and <sup>40</sup>K were measured by the low-level gamma spectrometry using HPGe detector. Based on the obtained activity concentrations, assessment of health risk potentially originated from the use of this kind of material for domestic and residential places was conducted.

Obtained values of activity concentration of <sup>226</sup>Ra, <sup>232</sup>Th and <sup>40</sup>K were higher than averaged values defined in Ref. [4], however, they are in consistency with the results from earlier studies in Serbia [14, 15]. Gamma indices and radium equivalent activity are below and around recommended values [6, 11]. Annual effective doses originated from ceramic tiles in domestic and residential places were also under limited value regulated in Serbia [5], leading to the conclusion there is no health risk in case of using analyzed ceramic tiles in building constructions, hence from the radiological standpoint they are safe and may be used without limitation.

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