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RADIOACTIVITY IN RAW MATERIALS USED IN CERAMICS INDUSTRY

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Abstract

The natural radioactivity in zirconium samples used in the ceramics industry has been measured. One of the most important use of zircon is as opacifier for ceramic tiles. The measurements were made by gamma spectrometry with a high purity germanium detector (HPGe). The gamma index, *I*, the external hazard index, H_{ex} and the internal hazard index, H_{in}, were calculated. The investigated samples can be used as one component in the recipe for the production of ceramics only if 3% of the samples used.

Introduction

The investigation of zirconium ores as sources of TENORM (technologically enhanced natural radioactivity) began in the late 1970s and early 1980s [1,2]. These works highlighted the relatively high concentrations of natural radionuclides in zirconium ores. At present Australia, South Africa, Ukraine, India, China, Brazil and Sri Lanka are the largest exporters of zirconium minerals [2]. The aim of this study was to measure the radioactivity in zirconium used as raw material in ceramics industry for production of ceramic colors, glazes, tiles and sanitary and to estimate the radiological hazards. These materials are tested continuously in Laboratory for Radiation and Environmental Protection Department, Vinča Institute, Belgrade, as part of regular inspection of imported goods from the customs.

Experimental

The investigated material was zirconium glaze. Before measurements, the samples were crushed, sieved and placed in the plastic 500 cm^3 Marinelli beakers. All samples were measured immediately after preparation because of the rapid assessment of whether the activity in a given sample is in accordance with applicable regulations or not. The spectra of samples were acquired using a high purity germanium detector (HPGe) with relative efficiency of 23 % and energy resolution of 1.8 keV for the 1332 keV ^{60}Co peak. The activity of ^{226}Ra and ^{232}Th was determined by measuring activity of their decay products: ^{214}Bi (609 keV, 1120 keV and also 1764 keV), ^{214}Pb (295 keV and 352 keV) and ^{228}Ac (338 keV and 911 keV), respectively. The activities of ^{40}K and ^{137}Cs were determined from 1460 keV and 661 keV γ-lines, respectively. ^{235}U was determined via its lines on 143 keV and 163 keV. ^{238}U was determined by measuring activity of their decay products: ^{234}Th (63 keV) or by ^{234}Pa (1000 keV).

Results and discussion

The measured specific activities of ^{226}Ra, ^{232}Th, ^{235}U and ^{238}U are presented in Table 1. The ^{238}U concentration ranging from 672-6524 Bq kg^{-1} was higher than concentration of ^{232}Th (187-885 Bq kg^{-1}), ^{235}U (42-279), ^{40}K (<MDC (16 Bq kg^{-1}) and ^{137}Cs (<2 Bq kg^{-1}) in all samples. On the other hand activity ratio $^{238}U/^{226}Ra$ was approximatelly 1, and can be assumed that these two radionuclides are in the radioactive equilibrium. In order to estimate the radiological hazards the gamma index, the external hazard index and the internal hazard index must be calculated. Due to the high activity of ^{226}Ra and ^{232}Th in the samples, it must be determined what percentage of the sample can be used in the recipe for the production of ceramics. Gamma index can be calculated using the following relation [3]:

$$I = \frac{C_{Ra}}{200} + \frac{C_{Th}}{300} + \frac{C_K}{3000} \qquad (1)$$

where C_{Ra}, C_{Th} and C_K are the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in Bq kg^{-1} in the building material, respectively. Gamma index must be less than 1 that the material could be used in high construction for interior, exterior and low construction. This applies to any building material. Gamma index for the investigated samples will be less than 1 only if 3% of the sample used. The obtained values for gamma index are presented in Table 1. For all investigated samples $I < 1$ (in the account, 3% of the activity of ^{226}Ra, ^{232}Th and ^{40}K was used).

Table 1. Activity concentrations of radionuclides in zirconium and appropriate gamma index

№	imported	Activity concentrations of radionuclides / Bq kg^{-1}				I
		^{226}Ra	^{232}Th	^{235}U	^{238}U	
1	Italy	2312 ± 347	367 ± 73	194 ± 58	2236 ± 894	0.38
2		2969 ± 297	816 ± 106	78 ± 33	3540 ± 1133	0.53
3		4147 ± 415	665 ± 80	266 ± 27	4522 ± 588	0.69
4		3967 ± 397	509 ± 61	230 ± 44	3855 ± 694	0.65
5		4350 ± 434	885 ± 88	178 ± 53	3853 ± 462	0.75
6		3201 ± 320	643 ± 64	191 ± 34	3948 ± 592	0.54
7		2732 ± 137	526 ± 68	164 ± 16	2425 ± 1336	0.46
8		3031 ± 303	630 ± 94	198 ± 20	3055 ± 611	0.52
9		3993 ± 399	555 ± 72	203 ± 32	3517 ± 914	0.66
10		3661 ± 366	658 ± 79	279 ± 28	3410 ± 682	0.62
11		2961 ± 296	570 ± 74	214 ± 21	2630 ± 736	0.50
12	Great Britain	2021 ± 202	396 ± 59	55 ± 26	1867 ± 672	0.35
13		3511 ± 351	454 ± 54	162 ± 41	2958 ± 740	0.57
14		3778 ± 378	584 ± 64	215 ± 54	4037 ± 1009	0.63
15	Slovenia	2603 ± 234	460 ± 60	42 ± 24	2266 ± 748	0.44
16		2106 ± 210	428 ± 51	54 ± 19	2687 ± 564	0.36
17		814 ± 163	187 ± 47	71 ± 14	672 ± 138	0.14
18	Sweden	2941 ± 294	565 ± 85	225 ± 56	2859 ± 715	0.50
19	Spain	3008 ± 301	596 ± 89	206 ± 21	3808 ± 762	0.51
20	Slovakia	4090 ± 1227	550 ± 165	193 ± 58	6524 ± 1174	0.67
21	Germany	4092 ± 409	624 ± 69	258 ± 52	3570 ± 714	0.68

The external hazard index, H_{ex}, is defined as [1]:

$$H_{ex} = C_{Ra}/370 + C_{Th}/259 + C_K/4810 \qquad (2)$$

where C_{Ra}, C_{Th} and C_K have the same meaning as in equation (1). The value of this index must be less than unity in order to keep the radiation hazard insignificant. The obtained values of H_{ex} for the zirconium ranged from 0.09 to 0.46 as shown in Fig. 1, values which are indeed less than unity (results obtained with 3% of the activity of ^{226}Ra, ^{232}Th and ^{40}K). In addition to the external hazard, radon and its short-lived products are also hazardous to the respiratory organs. The internal exposure to radon and its decay products is quantified by the internal hazard index, H_{in}, which is given by the equation [4]:

$$H_{in} = C_{Ra}/185 + C_{Th}/259 + C_K/4810 \quad (3)$$

For the safe use of a material in the ceramics industry, H_{in} should be less than unity. The calculated values of H_{in} ranged from 0.15 to 0.81 as shown in Fig. 1.

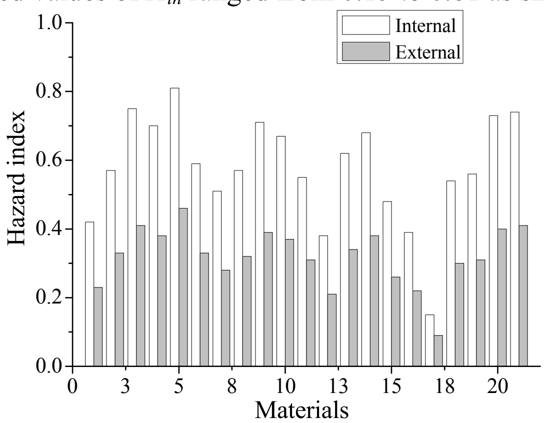

Fig.1. The measured values of both the external and the internal hazard indices.

Conclusion

Based on the results of measurement it can be concluded that the investigated materials are not radioactive in the context of the law [5]. Keeping in mind that the investigated samples used as one component in the recipe for the production of ceramics it can be concluded that there are no legal restrictions in terms of radioactivity in circulation and use of the investigated materials only if 3% of the samples used.

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