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GAMMA SPECTROMETRIC METHOD FOR MEASURING NATURAL RADIOACTIVITY OF BUILDING MATERIALS

Hungarian Academy of Sciences

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GAMMA SPECTROMETRIC METHOD FOR MEASURING NATURAL RADIOACTIVITY OF BUILDING MATERIALS

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ABSTRACT

^{232}Th, ^{226}Ra and ^{40}K concentrations determined by gamma spectrometric analysis of 121 building material samples collected from all over Hungary are presented and compared with the relating data from abroad. The expected radon and thoron concentrations as well as external gamma dose rate in an average brick house is estimated.

АННОТАЦИЯ

В работе сообщаются результаты гамма-спектрометрического анализа содержания ^{232}Th, ^{226}Ra и ^{40}K в 121 образце строительных материалов, собранных со всей территории Венгрии. Результаты сопоставляются с соответствующими международными данными. Оцениваются ожидаемая концентрация радона и торона в обыкновенных отечественных жилых домах с кирпичными стенами, а также интенсивность дозы внешнего гамма-излучения.

KIVONAT

Magyarország egész területéről begyüjtött összesen 121 épitőanyagminta gammaspektrometriás ^{232}Th,- ^{226}Ra,- és ^{40}K- tartalmának elemzési eredményeit közöljük és összehasonlitjuk ezeket a megfelelő külföldi adatokkal. Becsüljük az átlagos téglafalazatú hazai lakóházakban várható radon és toron koncentrációt, valamint a külső gammadózisintenzitást.

INTRODUCTION

The natural external radiation burden of the population is mainly due to the ^{232}Th, ^{226}Ra and ^{40}K concentrations of building materials. The total gamma radioactivity [1-6, 10, 11, 12, 14] and the ^{232}Th, ^{226}Ra and ^{40}K concentrations [7, 8, 9, 13-25, 28] of building materials has already been examined in several countries. From the latter data more information can be gained since knowing the ^{232}Th, ^{226}Ra and ^{40}K concentrations of the building materials and having appropriate models [30], [32] the estimation of the expected gamma dose rate in living rooms and a preliminary qualification of the building materials can be carried out from the point of view of health physics.

The minimum, maximum and average natural radioisotope concentrations of building materials used in several countries are summarized in Table I. Regarding the arithmetical means the highest (3.1 pCi/g) ^{232}Th concentration was measured in the FRG in slag brick, the lowest (0.3 pCi/g) in gypsum in the FRG and UK. The maximum ^{226}Ra concentration (18 pCi/g) was found in parget samples, the minimum value (0.3 pCi/g) in limestone both from the FRG. Some wall and floor covering materials had the maximum (31 pCi/g) ^{40}K concentration while the gypsum and parget the minimum (3 pCi/g). Accordingly, the ^{232}Th, ^{226}Ra and ^{40}K concentrations vary remarkably even within a given material from different geographical sites. This "variance" is characterized by the $(C_{max} - C_{min})/C_{average}$ ratio, the ^{232}Th concentration has an average "variance" of 218 per cent while the ^{226}Ra 171 per cent and the ^{40}K 165 per cent which can be attributed to local geological factors.

No information is yet available on the relative frequency distribution of the concentrations because of the insufficient number of analyses.

MEASURING METHODS

The natural ^{232}Th, ^{226}Ra and ^{40}K concentrations of building materials in Hungary were determined by gamma spectrometry [28, 29]. The measuring method and the calibration of the instrument as well as the spectrum evaluation have been detailed previously [28, 29]. Here we only mention that the pulverized samples of 700-2000 g were placed into airproof metal cans of 1075 cm^3 and the gamma spectra of the samples were determined behind the shielding of the HY. 3.2. whole body counter generally after 20 days of storage. Both the individual samples and their background were measured for 40 minutes each by using an 8"x4" NaI(Tl) detector and a multichannel analyser. The spectra were evaluated partly manually [28] partly by a computerized weight-iteration routine TORKA-I developed for this purpose [29].

Table I. The natural radioisotope concentration of building materials of several countries

Serial number	Building material	Number of samples	Country (number of regions)	Radioisotope concentration, pCi/g									Reference
				^{232}Th			^{226}Ra			^{40}K			
				min.	max.	mean	min.	max.	mean	min.	max.	mean	
1.	Concrete *	87	USSR (12)	0.4	3.7	0.8	0.6	2.9	0.9	7	24	15	[15, 21]
1/A	Light weight concr.	16	USSR (6)	0.6	2.1	0.9	1.4	3.3	2.0	5	26	14	[15, 21]
1/B	Sand, coarse (ballast)	36	USSR, UK, FRG	0.2	1.0	0.5	0.2	0.7	0.5	1	18	6	[15,21,14,23]
2.	Blast furnace slag	29	USSR (9)	0.3	1.2	0.5	1.0	3.9	1.8	1	12	7	[15, 21]
3.	Concrete made by fly ash	3	UK	0.9	1.2	1.0	0.2	5.7	1.8	6	16	12	[14]
4.	Red brick	86	USSR(5), FRG	0.2	2.0	1.1	0.6	3.0	1.5	1	30	17	[15, 21, 23]
5.	Lime-sand brick	50	UK, USSR (7)	0.1	1.2	0.7	0.2	1.4	[illegible]	2	22	12	[14, 15]
6.	Building brick	?	The Netherlands	-	-	1.3	-	-	0.7	-	-	18	[11]
7.	Adobe brick	?	Taiwan	-	-	-	-	-	1.2	-	-	14	[22]
8.	Wall- and floor covering materials (paving blocks, floor tiles)	49	USSR (7) , FRG	0.4	4.7	2.2	0.4	3.4	1.8	5	45	31	[15, 21, 23]
9.	Cement	12	USSR (3) , FRG	0.4	5.2	0.5	0.4	5.3	0.8	1	11	[illegible]	[15, 21, 23]
10.	Portland-cement (with 15% blast furnace slag)	3	USSR (1)	0.4	0.5	0.5	0.9	0.9	0.9	3	11	6	[15]
11.	Gypsum, natural	83	UK, FRG	-	-	0.3	-	-	0.6	-	-	3	[14, 23]
12.	Limestone	?	FRG	-	-	0.4	-	-	0.3	-	-	11	[13]
13.	Stone, coarse, crushed	41	USSR (2) , FRG	0.2	5.0	1.4	0.2	1.8	0.8	1	41	17	[15, 21, 23]
14.	Phosphorous slag	15	USSR (1)	-	-	0.6	-	-	6.1	-	-	4	[15, 21]
15.	Slag brick	7	FRG	0.6	7.6	3.1	0.6	4.1	2.0	8	32	17	[23]
16.	Slag- pumice	6	USSR (2)	0.5	0.5	0.5	2.8	6.0	5.5	4	5	5	[15]
17.	Pumice	20	FRG	1.0	2.8	1.7	1.6	3.0	2.4	13	28	20	[23]
18.	Sandstone	?	USSR, FRG	-	-	0.5	-	-	0.4	-	-	10	[13, 27]
19.	Artificial stones	10	FRG	0.2	4.6	1.6	0.2	2.7	1.0	2	31	12	[23]
20.	Aggregates	7	FRG	0.1	1.7	0.7	0.1	1.5	0.6	1	17	6	[23]
21.	Parget	5	FRG	-	-	0.5	7.0	29.7	18.0	1	7	3	[23]
22.	Soils with normal radioactivity	?	USSR	-	-	0.6	-	-	0.7	-	-	21	[26]
23.	Sands, natural	18	USSR (6)	0.4	0.8	0.5	0.4	1.0	0.5	2	16	7	[15, 21]

* This is known as "heavy concrete" in the USSR.

ERRORS

The statistical errors of the ^{232}Th, ^{226}Ra and ^{40}K determination (which did not exceed ± 10% even in the case of low radioactivity cement and concrete) and the χ^2 values characterizing the goodness of spectrum fitting (which in most cases did not exceed the acceptable figure of 4.0) were obtained by the computerized evaluation.

Examining the random and systematic errors of our measurements the total uncertainty of our data is estimated to be 20%.

ESTIMATION OF THE LOWEST MEASURABLE CONCENTRATIONS

The concentrations and σ_i statistical errors obtained by the computerized analysis of concrete and cement samples having the lowest radioactivity - and hence the highest statistical error - together with the 5% error of first-kind and 5% error of second-kind lead to the definition of the lower measuring limit [31]

$$LLD_i = 3.29 \cdot \sigma_i$$

where i = 1, 2 and 3 denote the elements ^{232}Th, ^{226}Ra and ^{40}K, respectively. This means that the concentrations of about

0.07 pCi $^{232}Th/g$,
0.1 pCi $^{226}Ra/g$ and
0.8 pCi $^{40}K/g$

could be measured approximately with a statistical error of ± 10%.

All the concentrations measured in building material samples exceeded these threshold values.

RESULTS

Altogether 121 samples from all over Hungary, one from each factory producing building materials, were examined (Table II).

The ^{232}Th concentration of cement made of fly ash from Pécs or Beremend is three times higher than that of similar materials made of fly ash from other Hungarian power plants due to the 5-7 pCi/g [28] ^{232}Th concentration of the filter fly ash from Pécs (Table II). The ^{232}Th, ^{226}Ra and ^{40}K concentrations in the fly ash concrete produced of fly ash from Power Plant II in Tatabánya are, however, only slightly lower than those in the fly ash concrete from Pécs. It is well known [28], that the fly ash from Power Plant I in Ajka - Tatabánya has a high (40-60 pCi/g) ^{226}Ra concentration. It is for this reason that the ^{226}Ra concentration in the fly ash concrete produced from the fly ash of the Power Plant Tatabánya I is so extremely high (23.8 pCi/g).

Table II Natural radioisotope concentrations of building materials of Hungary

Serial number	Building material	Number of samples	Radioisotope concentration, pCi/g									Notes
			^{232}Th			^{226}Ra			^{40}K			
			min.	max.	mean	min.	max.	mean	min.	max.	mean	
1.	Concrete	30	0.2	0.6	0.4	0.1a	0.6	0.3	3	9	5	a: in one case
1/B	Danubian ballast	1	-		0.3	-	-	0.2	-	-	7	as aggregate for concrete
2.	Blast furnace foamed slag concrete	2	0.7	0.8	0.8	2.8	3.3	3.0	4	5	5	
3.	Fly ash concrete	5	4.3	5.0	4.7	5.1	6.7	6.0	17	19	18	with ca.75% filter fly ash from Pécs
	"	2	1.1	1.6	1.4	1.6	1.7	1.6	7	7	7	with ca.75% fly ash from Gyöngyösvisonta
	"	1	-	-	1.7	-	-	23.8	-	-	13	with ca.75% fly ash from Power Station I Tatabánya
	"	1	-	-	3.8	-	-	4.6	-	-	16	with ca.75% fly ash from Power St. II Tatabánya
	"	1	-	-	1.9	-	-	4.3	-	-	7	with ca.75% fly ash from Power Station Csepel
4.	Red brick	15	1.1	2.0	1.4	0.9	1.5	1.2	13	21	16	burnt, small of clay
5.	Yellow brick	5	1.1	1.4	1.2	0.9	5.7b	1.4	15	18	17	burnt, small of clay b:from Devecser, not included in average
6.	Bricks for curtain walls	26	1.2	1.7	1.4	1.0	1.9	1.3	12	22	17	
7.	Adobe brick	4	1.1	1.4	1.2	0.7	1.2	0.9	12	21	16	
8.	Covering materials:											
	a/ fireclay-brick	1	-	-	2.1	-	-	1.2	-	-	18	for lining of cockle stoves
	b/ marble tile	2	0.2	0.3	0.3	0.4	0.6	0.5	1	3	2	with limestone crushing
	c/ wall- and floor covering materials (paving blocks, floor tiles)	4	1.4	2.1	1.6	1.1	2.1	1.4	6	21	15	tile, stoneware plate, brick
9.	Cement	12	0.3	0.6	0.5	0.3	1.9c	0.7	1	6	4	c:cement № 600 from Tatabánya, not included in the average
10.	Cement with fly ash											
	a/ BCM	6	1.0	1.5	1.2	1.4	1.9	1.5	6	9	8	with 10% fly ash from Pécs
	b/ DCM	1	-	-	0.6	-	-	1.2	-	..	4	DCM pc № 500
11.	"Keramzit" balls	1	-	-	1.7	-	-	1.1	-	-	17	additive agent for thermal insulation in concrete
12.	Roof-tile	1	-	-	1.5	-	-	2.0	-	-	21	for roofing, from Bátaszék

It is presumably the high ^{226}Ra concentration in the geological environment of Ajka-Tatabánya which explains that the small, yellow bricks produced in the Devecser Brick Factory II 10 km westwards of Ajka show a ^{226}Ra concentration of 5.7 pCi/g - a value four times higher than the average for bricks. The same can be said of the 1.9 pCi/g value of the ^{226}Ra concentration measured in the Tatabánya Nº 600 cement, which is nearly three times higher than the average for cements. On the other hand, the natural radioactivity of the Danubian ballast is rather low (Table II).

The concentrations in red brick, yellow brick and in bricks for curtain walls are practically equal.

In accordance with the well known low activity of limestone, a quite low radioactive concentration was measured in marble tiles made with crushed limestone.

Comparing values of Hungarian materials with those found for foreign ones the following <u>conclusions</u> can be drawn:

a/ The natural radioactivity of the Hungarian concretes amounts to the half or third of that of the Soviet ones.

b/ Red bricks from Hungary contain practically the same ^{232}Th, ^{226}Ra and ^{40}K concentrations as those of other countries.

c/ The ^{226}Ra concentration of the Dutch building brick is half as high as that of the curtain wall brick used in Hungary while their ^{232}Th and ^{40}K concentrations are practically equal.

d/ The ^{226}Ra and ^{40}K concentrations in adobes from Hungary and from Taiwan do not differ very much.

e/ The wall covering materials used in Hungary show lower natural radioactivity than those of other countries.

f/ The natural radioactivity of cements from Hungary and of other countries are practically equal.

As we think that the number of samples analysed (121) is not yet sufficient for further conclusions, we intend to continue our investigations for one or two years by analysing every month two samples from each building material factory.

Our data presented here are considered to be of preliminary character.

Finally we mention the expected values which were calculated for a living room built of exclusively red brick on the basis of our approximate model of radon and thoron concentration as well as gamma dose rate [30]:

$C^{e}_{Rn} \approx$ 1 pCi/litre equilibrium radon concentration

$C^{e}_{Tn} \gtrsim$ 0.26 pCi/litre equilibrium thoron concentration

$D_{\gamma} \approx$ 105.7 mrad/year air dose rate.

For the calculation we used the average values from lines 4, 5, 6 and columns "means" of Table II (1.3 pCi ^{232}Th/g, 1.3 pCi ^{226}Ra/g, 17 pCi ^{40}K/g), an emanation factor of $K_{Rn}=K_{Tn}=0.04$ and the $k=0.13$ air change per hour. These expected values are in good agreement with the data measured in Hungary [33] and other countries.

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