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THE RADIOLOGICAL RISK ASSESSMENT DUE TO THE RADIOACTIVITY OF THERMAL POWER STATION ASHES ADDED IN BUILDING MATERIALS

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The paper presents the results of radioactivity measurements for the main components from building materials and thermal power station ashes. Thus, the radioactivity of respectively materials is calculated, versus their composition and compared with the experimental radioactivity. The accuracy of those radioactivity measurements is estimated using the mathematical statistics methods. Then, from measurements results is determined the radiation risk of population which lives or works in houses build from such materials. The obtained results are compared with the maximal values accepted by Standards of Radiological Protection recommended by European Union and also by the Roumanian regulations. The natural radionuclides take into account for calculations and experimental measurements are: ^{226}Ra, ^{232}Th and ^{40}K. The obtaining results leads to the conclusion that the electrofilter ash from thermal power station gives the higher contribution to total radioactivity of building materials in which is used as addition and, accordingly, must not exceed 30%.

INTRODUCTION

The radioactive pollution of environment (nuclear or non-nuclear related) is now a problem which concerns the scientifically community and also the political factors from entire world. Radioactive materials can be classified under two broad headings: man-made and naturally occurring radioactive materials (NORM). TENORM is produced when radionuclides that occur naturally in ores, soils, water, or other natural materials are concentrated or exposed to the environment by activities, such as uranium mining, energy products, water and waste treatment, etc.[1-4] The coals radioactivity (NORM) burns in thermal power station is a special concern and is concentrated on flying ash, being carry along into burning gases and water retain by electrofilter for this purpose. This ash represent a polluting radioactive source by its using as addition in cements component and other building materials and also by its evacuation at chimney; used as addition in building materials determine an supplementary radiation hazard towards the natural radioactivity background, which cannot be neglect (TENORM).

The raw materials radioactivity which entering into cements component: chalks, sands, gypsum and electrofilter ashes from thermal power station, especially based on coals, is given on 90% proportion by natural radionuclides from radioactive decay chains of ^{238}U and ^{232}Th, and also by ^{40}K radionuclide, which is also natural.[5] The abundance of these elements in ground is the following:[6] U: 3.0 g/tone (metamorphosis sedimentary rocks); Th: 12.4 g/tone (metamorphosis sedimentary rocks) and $K_{natural}$: 3.0% (metamorphosis sedimentary rocks). ^{40}K radionuclide represents, in weight percents, 0.0118% of the natural potassium. In coals, the concentration of these radionuclides varies from one deposit to another, depending on the time in which vegetal substance fixed the radioactive isotope. These natural radionuclides wielded over human body an external and internal irradiation. The external irradiation is mainly determined by ^{214}Pb(RaB) and ^{214}Bi(RaC)

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radionuclides from ^{238}U decay chain, and also by ^{228}Ac (Mesothorium II), ^{212}Pb(ThB), ^{208}Tl(ThC) radionuclides from ^{232}Th radioactive series, as well as ^{40}K radionuclide from natural potassium. The internal irradiation is determined especially by radon and its short life daughters.[7-9]

This paper consider the radiation risk assessment of population based on these radionuclides if it is used the electrofilter ashes from thermal power station as addition at cements and concretes fabrication, the measurements being made on coal-ash-cement-concrete chain.

RESULTS AND DISCUSSION

To evaluate the radioactivity of 20 pit coal samples, the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K, radioisotopes was determined considering ^{226}Ra as the most important radionuclide from ^{238}U radioactive decay chain. Also, is measured the debit of the equivalent gamma dose for 20 coal samples at 1 m distance from coal surface. The measurements results are presented in table 1, together with the calculated values for radioactive index (RI) using the relation recommended by the European Union and taken from previous work:[1,10]

$$IR = \frac{C_{^{226}Ra}}{300} + \frac{C_{^{232}Th}}{200} + \frac{C_{^{40}K}}{3000} \quad (1)$$

in which: $C_{^{226}Ra}$, $C_{^{232}Th}$ *and* $C_{^{40}K}$ represent the activity concentrations of respective radionuclides, in $Bq \cdot kg^{-1}$. The adequate constants 300, 200 and 3000 are also given in $Bq \cdot kg^{-1}$ which made that the radioactive index being dimensionless.

Table 1

The natural radioactivity of coal samples

No. sample	The debit dose equivalent, H ($\mu Sv \cdot h^{-1}$)	The activity concentration, C ($Bq \cdot kg^{-1}$)			RI	Observations
		^{226}Ra	^{232}Th	^{40}K		
1	0.18	40	30	82	0.31	Maximum value
2	0.15	31	24	80	0.25	
3	0.18	35	30	92	0.30	
4	0.16	33	26	87	0.27	
5	0.15	30	26	88	0.26	
6	0.14	26	18	80	0.20	Minimum value
7	0.14	28	18	78	0.21	
8	0.17	38	25	83	0.28	
9	0.16	34	23	81	0.26	
10	0.15	29	24	78	0.24	
11	0.19	32	27	82	0.27	
12	0.16	35	25	84	0.27	
13	0.18	37	29	86	0.30	
14	0.17	38	26	79	0.28	
15	0.15	33	31	81	0.29	
16	0.19	39	29	84	0.30	
17	0.15	28	27	87	0.26	
18	0.14	27	17	79	0.20	Minimum value
19	0.16	31	25	89	0.26	
20	0.17	39	24	85	0.28	
					$RI_{average} = 0.26$	

From these data we can conclude that in Jiu Valley carboniferous basin, the average radioactivity level is low. The average radioactive index $RI_{average} = 0.26$ is sensible situated under the value of 0.5, considered by national standards being as the maximum admitted. The electrofilter ash is sampled from Mintia-Deva thermal power station on several days in different burn coals lots, being analyzed 27 ash samples, which radioactivity is maintain constant. The radioactive index is calculated from values of activity concentrations based on relation (1) ranging between $RI_{min} = 0.89$ and $RI_{max} = 1.08$. The results of these measurements are presented in table 2.

Data from table 2 shows that at coals burning on thermal power station occurs a pronounced concentration of radioactivity in ash result (TENORM – technologically enhanced NORM), the average radioactive index of ash being four times higher than in coals (0.97 for ash, towards 0.26 for coals).

Table 2

The radioactivity of electrofilter ash samples from Mintia-Deva thermal power station

No. sample	The activity concentration (Bq·kg^{-1})			RI	RI in ascending order	$X_i - \overline{X}$	$(X_i - \overline{X})^2$
	^{226}Ra	^{232}Th	^{40}K				
1	118.3	58.6	613.5	0.96	0.88	-0.086	0.0074
2	146.7	73.2	600.9	1.05	0.89	-0.076	0.0058
3	132.5	66.4	755.5	1.03	0.91	-0.056	0.0031
4	150.1	60.1	662.5	1.02	0.92	-0.046	0.0021
5	136.6	50.0	629.5	0.92	0.92	-0.046	0.0021
6	126.3	55.3	712.2	0.93	0.92	-0.046	0.0021
7	146.5	63.4	626.4	1.01	0.93	-0.036	0.0013
8	128.4	51.7	747.2	0.94	0.93	-0.036	0.0013
9	136.1	66.9	812.0	1.06	0.94	-0.026	0.0008
10	137.7	62.4	655.3	0.99	0.95	-0.016	0.0003
11	138.2	65.7	603.8	0.99	0.95	-0.016	0.0003
12	139.3	53.9	816.1	1.01	0.95	-0.016	0.0003
13	127.7	56.8	747.6	0.95	0.96	-0.006	≈ 0
14	121.8	59.7	748.8	0.95	0.96	-0.006	≈ 0
15	123.4	62.4	695.4	0.96	0.96	-0.006	≈ 0
16	126.0	66.2	692.5	0.98	0.96	-0.006	≈ 0
17	131.3	56.2	700.3	0.95	0.98	+0.014	0.0002
18	147.6	58.3	622.8	0.99	0.99	+0.024	0.0006
19	126.7	70.7	555.6	0.96	0.99	+0.024	0.0006
20	132.3	48.0	607.0	0.88	0.99	+0.024	0.0006
21	132.7	65.4	574.5	0.96	1.01	+0.044	0.0019
22	140.0	47.7	639.1	0.92	1.01	+0.044	0.0019
23	119.3	55.5	638.5	0.89	1.01	+0.044	0.0019
24	135.0	58.8	805.3	1.01	1.02	+0.054	0.0029
25	114.4	63.3	660.0	0.92	1.03	+0.064	0.0041
26	118.1	60.9	703.7	0.93	1.05	+0.084	0.0071
27	125.9	56.5	629.3	0.91	1.06	+0.094	0.0088
							$RI_{average} = 0.97$

The ash used as additive in building materials brings an important supply to radioactivity of these materials and also to radiologic hazard of population. For the precise assessment of this radiological risk whereon the population is exposed it must be known the value of radioactive index for ash, and by calculus we establish the maximum value of percent in which the ash is added in building material. For this, we made the statistic processing of data presented in table 2.

$$\sum_{i=1}^{27} X_i = 26{,}07\,; \qquad \sum_{i=1}^{27} (X_i - \overline{X}) \approx 0\,; \qquad \sum_{i=1}^{27} (X_i - \overline{X})^2 = 0.0575$$

The statistic parameters of the measurements series are: number of measurements: n=27; the average value of radioactive index: $RI_{average} = \overline{X} = \frac{\sum_{i=1}^{27} X_i}{n} = 0{,}966$; the median of the measurements series: $\frac{1}{2}(X_{13} + X_{14}) = \frac{0.96 + 0.96}{2} = 0.96$; dispersion: $S^2 = \frac{1}{n}\sum_{i=1}^{n}(X_i - \overline{X})^2 = \frac{1}{27} \cdot 0.0575 = 0.00213$; root-mean-square deviation: $S = \sqrt{\frac{1}{n}(X_i - \overline{X})^2} = 0.0461$; the experimental standard deviation of measurement: $\sigma_i = \sqrt{\frac{\sum_{i=1}^{27}(X_i - \overline{X})^2}{n-1}} = \sqrt{\frac{0.0575}{26}} = 0.0470$; the standard deviation of average: $\sigma_m = \sqrt{\frac{\sum_{i=1}^{n}(X_i - \overline{X})^2}{n(n-1)}} = \sqrt{\frac{0.0575}{27 \cdot 26}} = 0.0091$ and the

relative standard deviation (in percent):

$$ASR = \frac{\sigma_m}{\overline{X}} \cdot 100 = \frac{0.0091}{0.966} \cdot 100 = 0.942\% .$$

To evaluate the precision of the radioactive index RI determination, we applied Grubbs-Smirnov[11,12] test to eliminate from series of measurement the "aberrant" values if these exist. For an experimental set of values X_1, X_2,...X_n we could determine, with a certain probability P, the theoretical maximal value $X_{n(P)}$ which can be exceed only with certain small probability 1-P:

$$X_{n(P)} = \overline{X} + Z_{n(P)} \cdot \sigma_i \qquad (2)$$

in which: $\overline{X}$ is average value of radioactive index from n measurements;

σ_i is experimental standard deviation of measurement;

$Z_{n(P)}$ is given in table 3, being dependent on the probability P (name degree of reliability) and also on the number of experimental determination n.

In table 3 are given some values of variable $Z_{n(P)}$ for degree of reliability P=0.95 and the number of determination between n=15 and n=30.[13]

From relation (2) it results the theoretical maximal value of radioactive index which can be exceed only with probability smaller than 0.05.

Table 3

The values of variable $Z_{n(P)}$ for P=0.95 and 15≤n≤30

n	15	17	19	21	23	25	27	29	30
$Z_{n(P)}$	2.493	2.548	2.587	2.617	2.638	2.652	2.664	2.671	2.673

$$X_{max.teor.} = \overline{X} + 2.664\sigma_i = 0.966 + 2.664x0.047 = 1.0912$$

Because the highest value of RI from 27 series of measurement is $RI_{max.}$=1.06, which is below $RI_{max.theor.}$, it results that the series of 27 experimental values is homogenous due to maximal value. Similarly, the theoretical minimal value is given by relation:

$$X_{\min.theor.} = \overline{X} - Z_{n(P)} \cdot \sigma_i \qquad (3)$$

that is: $X_{min.theor.}$ = 0.966-2.664x0.047 = 0.8408

The smallest value from 27 experimental values being X_{min} = 0.88, over $X_{min.teor.}$, we conclude that the set of 27 values is homogenous and this series constitute a homogenous population due to minimal value. Consequently it is not necessary to remove any data from those 27 obtaining experimentally. After this validation of the series homogeneity of experimental values we could estimate the precision of radioactive index determination. If we admitted that the experimental errors are submitted to normal distribution law, we could calculated the probability that error of determination being under limits $\pm\nabla$. Usually, ∇ is given under standard deviation multiple form of determinations method, σ_m:

$$\nabla = k \cdot \sigma_m \quad \text{namely} \quad X = \overline{X} \pm k \cdot \sigma_m \qquad (4)$$

This expression form of measurement precision is not completed, because not specified with what probability the measurement value will found in interval $X = \overline{X} \pm k \cdot \sigma_m$. This probability or degree of reliability[10] calculated by function of normal distribution for different k values is given in table 4:

Table 4

The values of probability P (degree of reliability) for different values of k

k	0.50	0.67	1.00	1.96	2.00	2.58	3.00
P	0.383 (38.3%)	0.500 (50%)	0.683 (68.3%)	0.950 (95%)	0.954 (95.4%)	0.990 (99%)	0.997 (99.7%)

From these reasons, the radioactive index of electrofilter ash from Mintia-Deva thermal power station is:

$$RI = 0.966 \pm 3 \cdot 0.0091 = 0.966 \pm 0.027$$

In order to determine in what measure the electrofilter ash radioactivity is found in the additive cement, 3 cement samples with ash addition in different quantities were prepared. For these cement samples are measured the activity concentrations of 3 reference radionuclides (^{226}Ra, ^{232}Th and ^{40}K) and from these are calculated experimental radioactive index of cements who were then compared with theoretic index obtaining from radioactive index of precursors and each weight in respective cement. As precursors for

cements obtaining were used the clinkerized raw materials (burn in furnace) with RI= 0.16, gypsum with RI = 0.19 and electrofilter ash with RI = 0.96. The clinkerized raw materials are used mixtures of chalks, clays and pyrite ash. The obtaining results are presented in table 5.

Table 5

The experimental and theoretic radioactive index for cements with different electrofilter ash content

Sample	Composition (% weight)			$RI_{exp.}$	$IR_{theor.}$
	Raw materials RI=0.16	Gypsum with RI = 0.19	Electrofilter ash with RI =0.96		
1	70%	5%	25%	0.38	0.36
2	75%	5%	20%	0.33	0.32
3	80%	5%	15%	0.27	0.28

Data from this table shows that the radioactivity of electrofilter ash used as addition at cement preparation influence critical the cement radioactivity, having the determinant role to radiological risk estimation for population who lives or works in houses that structure is used the cement with electrofilter ash addition. Also, there is a good agreement between experimental and theoretical values of radioactive index, differences from these values being in limits of measure errors.

According with the European regulation on the Radiological Protection Norms concerning building materials,[11] the supplementary exposure of the population due to the radioactivity of these materials must not exceed an equivalent of absorbed dose higher than 0.3 $mSv \cdot year^{-1}$ and in special cases this limit can be 1 $mSv \cdot year^{-1}$. To estimate this radiological risk for population we must know the activity concentrations of the reference radionuclides (^{226}Ra, ^{232}Th and ^{40}K) in building materials used, contribution of other radionuclides to supplementary exposure being insignificant.

A simple calculation allows establishing the correlation between activity concentration and the equivalent of yearly absorbed dose for each reference radionuclide mentioned.

Considering a model room from concrete with 4mx5mx2.8m sizes, 20 cm walls thickness and 2350 $kg \cdot m^{-3}$ concrete density to natural background of radiations of 50 $nGy \cdot h^{-1}$ medium value and conversion factor absorbed dose-equivalent dose of 0.7 $Sv \cdot Gy^{-1}$,[15] results for activity concentration unity, 1 $Bq \cdot kg^{-1}$, the following values of dose equivalent:[16] 0.92 $nGy \cdot h^{-1}$ or 0.644 $nSv \cdot h^{-1}$ for radionuclide ^{226}Ra; 1.10 $nGy \cdot h^{-1}$ or 0.77 $nSv \cdot h^{-1}$ for radionuclide ^{232}Th; 0.08 $nGy \cdot h^{-1}$ or 0.056 $nSv \cdot h^{-1}$ for radionuclide ^{40}K.

If we supposed that in this room a person spend every day 16 hours, 5840 hours yearly, the equivalent of the yearly absorbed dose at the same activity concentration unit, (1 $Bq \cdot kg^{-1}$), will be: 3.76 $\mu Sv \cdot year^{-1}$ for ^{226}Ra; 4.5 $\mu Sv \cdot year^{-1}$ for ^{232}Th and 0.327 $\mu Sv \cdot year^{-1}$ for ^{40}K. Evidently, in rare cases, when concrete contains all 3 reference radionuclides with known activity concentration, these values of equivalent absorbed dose will be summed leading to total value of dose equivalent experienced by person in one year, over the value determined by natural radiation background. For example, for the model room above-mentioned, made by concrete, having the activity concentrations 80, 70 and 800 $Bq \cdot kg^{-1}$ for ^{226}Ra, ^{232}Th and ^{40}K, respectively the absorbed debit dose will be:

$D = (0.92x80+1.10x70+0.08x800)\ nGy \cdot h^{-1} \approx 215\ nGy \cdot h^{-1}$

The yearly absorbed dose equivalent became:

$H = 0.7\ Sv \cdot Gy^{-1} \cdot 215x10^{-9}\ Gy \cdot h^{-1} \cdot 5840\ h \cdot year^{-1} = 0.88x10^{-3}\ Sv \cdot year^{-1} = 0.88\ mSv \cdot year^{-1}$

Therefore, the numeric value of radioactive index (dimensionless) coincide with absorbed dose equivalent generated by this material, if it is given in $mSv \cdot year^{-1}$.

EXPERIMENTAL

To evaluate the radiological risk due to building materials, the activity concentrations of reference radionuclides: ^{226}Ra, ^{232}Th and ^{40}K, in coal samples from Jiu Valley, electrofilter ash from Mintia-Deva thermal power station and cement with electrofilter ash addition was measured. The activity concentrations were measured by gamma spectrometry by using a multichannel analyzer ORTEC type (SUA) equipped with a high purity germanium, detector semiconductor HPGe. For ^{226}Ra radionuclide is used energetic picks from 352 keV (RaB) and 609 keV (RaC); for ^{232}Th: 910 keV and 970 keV (Mesothorium II), and for ^{40}K the line from 1460 keV.

The activity concentrations of each radionuclide were determined by relative method, referring the gamma pick intensity from sample to the same pick intensity from standard source with known activity concentration: $C_{pr.} = \frac{A_{pr}}{A_{et}} \cdot C_{et}$. As peak intensity was considered the peak surface area. All samples were powders form with circa 200 mesh granulation (about 0.075 mm particles diameter). The detection was made at constant volume (100 cm^3); the measure time was between

80,000-100,000 seconds. For each measured values are applied weight corrections, measuring time and natural background. For coal samples, was measured also the debit gamma dose equivalent by using a debitmeter EBERLINE (Germany), with a 0.01 $\mu Sv \cdot h^{-1}$ sensibility, according with standard legislation from Roumania. The measurements were made at 1 m distance from coal surface. The activity concentration of ^{222}Rn radionuclide was measured with radon-meter RAD-7 with 0.01 $Bq \cdot m^{-3}$ sensibility, the measurements made directly in coal mine in ventilation conditions.

CONCLUSIONS

The coals from Jiu Valley coal basin have low radioactivity, the radioactive index values ranging between 0.20-0.31, with an average value of 0.26, lower than value of 0.5, considered by national standard being maximum admitted. The coal burning in thermal power station leads to radioactivity concentration in resulted ash, the average radioactive index of this being approximately four times higher than coals (TENORM). The statistic data processing of radioactivity measurement for electrofilter ash shows that the obtained values series leads to homogenous population and do not contain ''aberrant values''. Consequently, the measurements are precise enough and the ash radioactivity is uniformly distributed in all ash mass. The analyzed cement samples with electrofilter ash addition shows that the cement radioactivity is due firstly by ash quantity added; this is the main factor which determine the radiological risk due to building materials. It was established a procedure to correlate the activity concentration and radioactive index with the equivalent absorbed dose made by reference radionuclides from building materials.

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