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THE ESTIMATION OF CEMENTS RADIOACTIVITY OBTAINED BY ELECTROFILTER ASHES ADDITION DUE TO THERMAL POWER STATION BASED ON COAL FROM OLTENIA COALFIELD, ROMANIA

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abstract: For cements radioactivity estimation obtained by refinement of electrofilter ashes due to thermal power station from Oltenia coalfield was elaborated an estimation method. The method is based on radioactive index (RI) calculation for every component used at cement fabrication. The radioactive index was obtained by measurements of activities concentrations for ^{226}Ra, ^{232}Th and ^{40}K radionuclides, using gamma spectrometry, according with EU recommendations. It is found that for cements obtained with addition of thermal power station ash RI varies between 0.16 and 0.38, with an average value by RI_{med}=0.24, namely smaller than maximum value admitted by Romanian's legislation (RI≤0.5).

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Introduction

The building materials contain different quantities of natural radionuclides. Those obtained from rocks and soil contains at least radionuclides descendant of ^{238}U and ^{232}Th and also ^{40}K radionuclide [1÷3]. From uranium radioactive cycle, the ^{226}Ra radionuclide is radiological most important and so, frequently, in practice is determined the activity concentration of this and also the activity concentrations of thorium (^{232}Th) and potassium (^{40}K). The contribution of other radionuclides at building materials is insignificant and it is omitted. The medium values in the ground of activity concentrations for ^{226}Ra, ^{232}Th and ^{40}K are around 40 $Bq\cdot kg^{-1}$ for ^{226}Ra and ^{232}Th and 400 $Bq\cdot kg^{-1}$ for ^{40}K. The radiations exposure due to building materials can be classified in external and internal exposure. The external exposure is due to gamma radiation of all 3 radionuclides (^{226}Ra, ^{232}Th and ^{40}K) and internal

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exposure is due to radon inhalation (^{222}Rn and ^{220}Rn) which is formed as a result of radioactive disintegration of uranium and thorium descendants.

The cements with different composition constitute a base component of concrete and other materials used frequently to house construction or building in which some of population is working, so that the population suffers an external and internal exposure. According European regulations, this exposure cannot determine an equivalent of absorbed dose bigger than 0.3 $mSv \cdot year^{-1}$ and only in special situations this limit can reach 1 $mSv \cdot year^{-1}$ [2]. To respect these limits it must be known the radioactivity of all the building materials components and especially the radioactivity of cement which must be used.

The objective followed in this paper is to evaluate the cements radioactivity in which is added the ashes result from coals burning into electrical thermal power station. For this, we measured the activity concentration (in $Bq \cdot kg^{-1}$) of radionuclides ^{226}Ra, ^{232}Th and ^{40}K of all the components from which is made the respective cement.

Experimental

To evaluate the cement radioactivity, we investigated the used components (raw materials): chalks, clays, pyrite ashes, gypsum, coal from Oltenia coalfield, electrofilter ashes from thermal power stations. Additionally, we measured the activity concentration for radionuclides Ra, Th and K and also the equivalent debit of absorbed dose at 1 m distance. The activity concentration was measured by gamma spectrometry with multichannel analyzer [4] type ORTEC (USA), jacketed with semi-conductor detector with high purity HPGe. For radionuclide ^{226}Ra are used the gamma lines (picks): 352 keV (RaB) and 609 keV (RaC); for radionuclide ^{232}Th: 910 keV and 970 keV (Mezothoriu II) and for ^{40}K: 1460 keV line. The activity concentration of each radionuclide was obtained by relative method, relating the intensity of gamma pick (from sample) to intensity of gamma pick (from standard source) with known concentration: $C_{pr} = \frac{A_{pr}}{A_{et}} \cdot C_{et}$. As pick intensity is considered the surface area under this. All samples were powdery with 200 mesh granulation (circa 0.075 mm particles diameter), the detection is made at constant volume (100 cm^3), and the measure time is between 20.000÷100.000 seconds. The equivalent debit of absorbed dose [5] was measured with radiometer EBERLINE (Germany) with 0.01 $\mu Sv \cdot h^{-1}$, according with standard conditions of radioactive measure in Romania.

Results and discussions

From values of activity concentrations measured by gamma spectrometry for radionuclides ^{226}Ra, ^{232}Th and ^{40}K was calculated the radioactive index (RI), using calculus formula recommended by European Commission [1]:

$$\mathrm{RI} = \frac{C_{^{226}\mathrm{Ra}}}{300\,\mathrm{Bq}\cdot\mathrm{kg}^{-1}} + \frac{C_{^{232}\mathrm{Th}}}{200\,\mathrm{Bq}\cdot\mathrm{kg}^{-1}} + \frac{C_{^{40}\mathrm{K}}}{3000\,\mathrm{Bq}\cdot\mathrm{kg}^{-1}}$$

in which $C_{^{226}Ra}, C_{^{232}Th}, \text{and} C_{^{40}K}$ represent the activity concentrations values of respectively radionuclides, in terms of Bq·kg^{-1}.

The raw materials used to cements obtaining and for which were calculated the radioactive index are the following: chalks, clays, pyrite ashes, gypsum, electrofilter ashes from the thermal power station based on coals from Oltenia coalfield [6-7]. For each raw materials were measured many samples, indicating the medium values and RI_{med} for respectively radioactive index. The obtained results are presented in Table 1.

Table 1 The medium values of radioactive index for raw materials used to cements preparation

No.	The raw materials used	Number of measured samples	RI_{med}
1	Chalks	30	0.09
2	Clays	20	0.46
3	Pyrite ashes	20	0.29
4	Gypsum	20	0.18
5	Electrofilter ashes from thermal power station	31	0.98

The data from the Table 1 shows that the electrofilter ashes represent the component with higher supply to total radioactivity of cements whom composition enters these ashes.

To establish the contribution of each radionuclide to coals radioactivity used in thermal power station and also the radioactivity of electrofilter ashes result at these coals burning in thermal power station were measured 42 coal samples rise from 7 mining exploitations from Oltenia coalfield and also 31 electrofilter ash samples from 3 thermal power stations which working with coal from this coalfield [4].

From activity concentrations values measured for radionuclides Ra, Th and K were calculated the radioactive index maximum RI_{max}, minimum RI_{min} and medium RI_{med}.

The obtained results were presented in Table 2.

Table 2 The coals radioactivity and electrofilter ashes results at coals burning in thermal power station

The material	The activity concentration (Bq·kg^{-1})									RI		
	^{226}Ra			^{232}Th			^{40}K					
	min.	max.	med.	min.	max.	med.	min.	max.	med.	min.	max.	med.
Coals	85	101	92	28	38	33	229	316	268	0.50	0.63	0.56
Electrofilter ashes	75	287	144	28	112	66	216	873	505	0.46	1.81	0.98

The data from this table shows, on the one hand, that the natural radioactivity of coals is concentrated in ashes result and which is driven in combustion gases and then is retained by electrofilter, and on the other hand, although ^{40}K radionuclide have the higher activity concentration in coals and in electrofilter ashes, this radionuclide have the smaller contribution to global value of radioactive index for respectively material. The maximum supply to total value of RI is due to ^{226}Ra radionuclide for coals and also for electrofilter ashes.

The radioactive index calculus for electrofilter ashes independently for each thermal power station studied shows that the values of these indexes are identical, being 0.98 with deviation which not exceed the experimental measured errors.

The cements radioactivity will depend visibly on proportion in which is added the electrofilter ashes because the radioactive index of ashes are substantially higher than the others components hence the cement is obtained.

In technological process of cements preparation, first is obtained the clinker formed by mixture of chalk, clay and pyrite ash, burn in furnace, and then is added the electrofilter ash in variable proportions and gypsum (5%). To establish this proportion it is necessary to know the radioactive index values of clinker (calculated from RI of components introduced into known proportion), the electrofilter ash and gypsum. The condition is that the RI of cement not exceed 0.50, this value correspond to valid legislation in Romania.

In this paper, for cements obtaining is used a clinker with RI=0.16 and were obtained and analyzed 7 cements using ashes from 3 thermal power station with RI values of 0.97, 1.33 and 1.56 and variable proportions, calculated such as to respect the condition RI~0.45.

For example, if x is mass percent of clinker, y is mass percent of ashes with RI=1.33 and gypsum have 5%, then the conditions lead to equation system:

$$\frac{x}{100}\cdot 0.16+\frac{y}{100}\cdot 1.33+\frac{5}{100}0.18=0.45$$
$$x+y+5=100$$

By resolving this system results the mass composition of cement:

70% clinker with RI=0.16

25% thermal power station ash with RI=1.33

5% gypsum with RI=0.18

For prepared and analyzed cements is observed a good concordance between the radioactive index values determined experimental by measuring the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K radionuclides and calculated values for same cements from radioactive index of components in which are those obtain. Thus, for cement with maximum content of thermal power station ashes with RI=1.33, the experimental value of RI is 0.39, while the calculated value is RI_{max}=0.38. Also, for cement sample with minimum ash content, the calculated value is 0.22, instead 0.26 obtained experimentally.

Conclusions

1. The coals from Oltenia carboniferous basin (7 mining exploitations) have a radioactivity, indicated by radioactive index RI=0.51÷0.62.

2. The ashes obtained by coals burning are characterized through RI=0.59÷1.78, these values are found at 3 thermal power stations that uses coal from Oltenia.

3. The proposed method for cements radioactive index estimation from radioactive index values of those components leads to good results in concordance with the experimental data obtained by measured activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K radionuclides.

4. By proposed method, we can calculate the maximum admissible percent of thermal power station ashes such as not exceed the maximum value of cement radioactivity provided by valid standards, RI≤0.5.

REFERENCES

1. ****Radiological Protection Principles concerning the Natural Radioactivity of Building Materials*, European Commission, Radiation Protection 112 (1999), Luxemburg.
2. ***IAEA – TEC DOC – *1363 Guide* (2003)
3. ****Practical Use the Concepts of Clearance and Exception – Part II*, European Commission, Radiation Protection 122 (2001)
4. ***INCDMRR – CEEX 2006: „The radiological risk estimation over the population health by using of structures cementation at building construction based on new binder composite systems''
5. Nedelcu, G.M., Zaharescu, T., Podina, C. and Mihalcea, I. (2008) *Revista de Chimie*, (Bucuresti) **59**(1), 33-6.
6. Petrescu, I. et al. (1986) **The geology of coal deposits**, vol. I, Ed. Tehnica Bucharest, 183.
7. Petrescu, I. et al. (1987) **The geology of coal deposits**, vol. II, Ed. Tehnica Bucharest, 214.