SPECTROMETRIC AND RADIOMETRIC TECHNIQUES OF NUCLEAR RADIATIONS IN THE ASSESSMENT OF ENVIRONMENTAL RADIOACTIVITY

Ion V. POPESCU^{1,2}, Petru MURSA³, Olimpiu-Razvan DUMITRESCU⁴

Abstract. The paper contains a synthesis of research results in the study of the level of environmental radioactivity obtained by using spectrometric technique, of high precision and high sensitivity, of atomic and nuclear radiations. Spectrometric technique were used to measure gamma nuclear radiation in determining the radioactivity of ash samples taken from the dumps of the ROMAG TERMO Drobeta Turnu Severin thermal power plant, Mehedinți county. By gamma spectrometry with high energy resolution semiconductor detector, high efficiency and protection for the cosmic radiation background (low radiation background) was determined the radioactivity of ash samples, taken from two coal dumps from ROMAG TERMO Drobeta Turnu Severin power plants, Romania. The results showed that the activities of the main radionuclides in the environmental samples including, ²²⁸Ac (7.5 ÷ 18.7 Bq/kg), ²²⁶Ra (6.2 ÷ 21.3 Bq/kg), ¹³⁷Cs (0.3 ÷ 1, 53(Bq/kg) and ⁴⁰K (169 ÷ 362 Bq/kg) are among the lowest values found compared to the maximum allowed limits.

Keywords: environmental radioactivity, spectrometric techniques, radionuclides.

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1. Introduction

In nuclear physics, radioactivity is defined by the totality of processes and phenomena of spontaneous emission of alpha, beta (corpuscular radiation) and gamma (electromagnetic) radiation by an unstable atomic nucleus, without external intervention [1, 2, 3].

Unstable nuclei also called natural or induced radionuclides (artificial radionuclides) can be dispersed in the environment in various forms, gases, liquids (radioactively contaminated precipitation) and solid materials (soils, ash resulting from burning coal in thermal power plants, etc.). atmosphere are driven by the processes of diffusion and transport due to air currents and temperature gradient. Atmospheric deposition of radionuclides on soil, surface water and vegetation is influenced by the

dynamics of air masses, gravity and precipitation[2, 3]. In surface and deep waters, through the diffusion and transport processes that take place, the spread over long distances of radionuclides is absorbed by the plants and the animal organisms encountered. Atmospheric deposition of radionuclides leads to an increase in the level of radioactivity of the soil at different depths depending on precipitation and the

¹ Professor, PhD, Valahia University of Targoviste (UVT), Academy of Romanian Scientists ² Multidisciplinarity Research Institute for Science and Technology of UVT(ICSTM-UVT), <u>ivpopes@yahoo.com</u>

³ Physicist, PhD, Center for Research and Innovation for Defense, CBRN and Ecology, (mursapetru_73@yahoo.com)

⁴ Chemist, PhD, Research and Innovation Center for CBRN Defence and Ecology.

granularity of the contaminating particles. By plants contaminated the radionuclides are transferred to animal organisms. Through air, water, plant and animal organisms, radionuclides reach the human body where radiation emission occurs at the cellular interaction and the occurrence of deterministic or stochastic effects [4-7]. The activity of radioactive materials with contain natural radionuclides can be artificially amplified by various technological processes, resulting in a new category of radionuclides called Technologically Enhanced Naturally Occurring Radioactive Materials (TENORM) [3].

Radioactivity is due to radionuclides existing in nature and those obtained artificially. For any of the components radioactivity is characterized quantitatively by the global radionuclid concentration in the substances that make it up, in Bq / kg, Bq / m^2 or Bq / m^3 [4-8].

2. Radioactivity. Types of radionuclides

2.1. The natural radioactivity is given by cosmogenic radionuclides (produced by cosmic radiation) and primordial radionuclides (since the formation of the Earth), which form the three natural radioactive series [6 - 8]. Cosmic radiation is made up of high-energy particles that originate in our galaxy, in distant galaxies, supernovae, pulsars and last but not least in the sun. Cosmic radiation is divided into primary and secondary cosmic radiation. Primary cosmic radiation consists of protons (85%), alpha particles (12%) and heavy nuclei. with Z> 30 (3%). The energy of these radiations reaches up to 1021 eV [10][8, 9]. The primary cosmic radiation interacts with the upper layer of the Earth's atmosphere with a frequency of about 0.02 cm² / s at the equator and 1 cm² / s at the poles. *https://ro.wikipedia.org >wiki> Radiation_cosmic*.

The difference in frequency is due to the shielding of electrically charged cosmic particles by the Earth's geomagnetic field [11d].[9-13]

Cosmic radiation has the highest intensity at the poles and the lowest at the equator.

Secondary cosmic radiation is produced by the interaction of primary cosmic radiation with the Earth's atmosphere and depends on latitude due to the Earth's uneven magnetic field. Secondary cosmic radiation consists mainly of: n, e +, e-, π , μ , ν , γ . Interactions of protons and neutrons with oxygen and nitrogen in the atmosphere can lead to spallation reactions or neutron capture[12]. *https://ro.wikipedia.org >wiki> Radiation cosmic*.

Most cosmogenic radionuclides occur as a result of the interaction of cosmic radiation with atmospheric constituents such as nitrogen, oxygen, argon, krypton and xenon. The rest appear through interactions with other elements in the biosphere and lithosphere (especially oxygen, magnesium, silicon, iron, aluminum, calcium and potassium) [9]. The main cosmogenic radionuclides resulting from the interaction of cosmic radiation with the Earth's atmosphere [11, 12] and their properties are given in Table 2.1.

Radionuclid	Half time	Type of emitted	The	Specific activity in the earth's crus				
type		radiation	nuclei	Air Rain water		Ocean		
						water		
¹⁰ Be	1.6·10 ⁶ y	β	N, O			2.0.10-8		
²⁶ Al	7.17·10⁵y	β^+	Ar			$2.0 \cdot 10^{-10}$		
³⁶ Cl	$3.01 \cdot 10^5 \mathrm{y}$	β	Ar			$1.0 \cdot 10^{-5}$		
¹⁴ C	5730 y	β	N, O			5.0·10 ⁻³		
³² Si	150 y	β	Ar			$4.0 \cdot 10^{-7}$		
³⁹ Ar	269 y	β	Ar			6.0·10 ⁻⁸		
³ H	12.33 y	β	N, O	$1.2 \cdot 10^{-3}$		$7.0 \cdot 10^{-4}$		
²² Na	2.60 y	β+	Ar	1.0.10-6	$2.8 \cdot 10^{-4}$			
³⁵ S	87.32 d	β	Ar	1.3.10-4	7.7.10-3			
⁷ Be	53.12d	γ	N, O	1.0.10-2	6.6.10-1			
³³ P	25.34 d	β	Ar	1.3.10-3				
^{32}P	14.26 d	β	Ar	$2.3 \cdot 10^{-4}$				
²⁸ Mg	21.0 h	β	Ar					
²⁴ Na	14.95 h	β	Ar		$3.0-5.9 \cdot 10^{-3}$			
³⁸ S	170.3 m	β	Ar		6.6-21.8·10 ⁻²			
³¹ Si	157.3 m	β	Ar					
¹⁸ F	109.77 m	β^+	Ar					
³⁹ Cl	55.6 m	β	Ar		$1.7 - 8.3 \cdot 10^{-1}$			
³⁸ Cl	37.24 m	β	Ar		$1.5 - 2.5 \cdot 10^{-1}$			
^{34m} Cl	32.00 m	β^+	Ar					

 Table 2.1.The main radionuclides resulting from the interaction cosmic radiation with the Earth's atmosphere [11,14]

A significant contribution to the radiation dose of the exposed population is mainly due the cosmogenic radionuclides, tritium, ¹⁴C, ³H, ²²Na and ⁷Be [10-12, 14]. Primordial radionuclides, appeared with the formation of the Earth, have a half-life of about 109 years, most belong to the three radioactive series ²³²Th (11 terms), ²³⁵U (16 terms) and ²³⁸U (18 terms) existing in nature [12, 14, 15]. In addition to the three natural radioactive series there are natural primordial radionuclides [10, 14] which are given in Table 2.2.

Table 2.2: Primordial radionuclides, în addition to the three
natural radioactive series [12 -14, 15]

Radionuclid type	Half time[years]	Type of emitted radiation	Specific activity in the earth's crust [Bq / kg]
40 K	$1.27 \cdot 10^{9}$	β,γ	630
⁵⁰ V	$1.4 \cdot 10^{17}$	γ	2.0.10-5
⁸⁷ Rb	$4.75 \cdot 10^{10}$	β	70
¹¹⁵ In	$4.41 \cdot 10^{14}$	β	2.0.10-5

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¹²³ Te	$1.0 \cdot 10^{13}$	raze X	2.0.10-7
¹³⁸ La	$1.05 \cdot 10^{11}$	β,γ	2.0.10-2
144 Nd	2.29·10 ¹⁵	α	3.0.10-4
¹⁴⁷ Sm	$1.06 \cdot 10^{11}$	α	0.7
152 Gd	$1.08 \cdot 10^{14}$	α	7.0.10-6
174 Hf	$2.0 \cdot 10^{15}$	α	2.0.10-7
¹⁷⁶ Lu	$3.78 \cdot 10^{10}$	β,γ	$4.0 \cdot 10^{-2}$
¹⁸⁷ Re	$4.35 \cdot 10^{10}$	β	1.0.10-3
¹⁹⁰ Pt	6.5.1011	α	$7.0 \cdot 10^{-8}$

The radionuclides from Table 2.2 are mainly found in some rocks in the lithosphere that can reach the earth's surface through the groundwater water circuit and the gaseous ones reach the atmosphere through cracks in the earth's crust. Most of these radionuclides are in very low concentrations in the environment [14].

Of course, we must add the primordial radionuclides, representative, ⁴⁰K, ³H ¹⁴C, ⁷Be and ²²Na, which also do not come from the 3 series of natural radioactive decay. ⁴⁰K is the only unstable isotope of potassium, with $T_{1/2} = 1.27 \cdot 10^9$ years and an abundance of 0.0118% in natural potassium [11-13].⁴⁰K is the most widespread primary radioactive isotope, and it disintegrates by electron capture (10.5%) in ⁴⁰Ar in steady state [11-13], present in the atmosphere. Tritium is formed in the stratosphere by the nuclear reaction ¹⁴N (¹n, ³H) ¹²C and by washing reactions. The ¹⁴C is formed in the upper layers of the atmosphere under the action of cosmic rays. ¹⁴C after being generated in the stratosphere, passes into the lower layers of the atmosphere from where it reaches living organisms through plant photosynthesis. Because $T_{1/2} = 5730$ years it is widely used in dating archaeological pieces of animal and plant origin. The ¹⁴C concentration is useful in archaeometry and geochronology. The nuclei of ⁷Be ($T_{1/2} = 53.4$ days) and ¹⁰Be ($T_{1/2} = 1.6 \cdot 10^6$ years) are produced by washing reactions in the upper layers of the atmosphere between protons and neutrons in cosmic radiation and the nuclei of Oxygen or Nitrogen in atmosphere. ¹⁰Be is a good monitor of the intensity of cosmic radiation [11 - 14].

Radionuclide ²²Na results from the interaction of neutrons produced by cosmic radiation, which have high energy, and Ar. ²²Na ($t_{1/2} = 2.6$ years) is converted to ²²Ne, by positron emission (90%) and electron capture (10%). A significant amount of ²²Na remains in the atmosphere where it is produced and the other side reaches the oceans (at the surface).ale[14].

2.2. The artificial radioactivity included in the anthropogenic factor which consists in the direct influence of human activity on the environment with negative effects on the relief, vegetation and fauna. This category also includes

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radioactivity resulting from experiments with nuclear weapons, the use of nuclear power plants and radionuclides obtained in the laboratory by nuclear reactions performed with particle accelerators and nuclear research reactors and which are used in various fields of applied research: archeology, metallurgy, medicine, environment, etc.). The first radioactive isotope, 30P, was artificially obtained in 1934, by Irene Curie and Piere Juliot, by bombarding thin sheets of aluminum with alpha particles [15].

In many activities, radioisotopes are used during the technological process in higher concentrations than those usually encountered (radioisotopes used in the preparation of luminescent paints, power supplies for pacemakers, fire detectors, analyzers). gases, radioactive tracers, etc. A significant amount of radionuclides also occurs in various non-nuclear fields such as coal, metal and geothermal mining or the disposal, storage and use of ash from thermal power plants 12-15].

3. Radioactivity of coal used in power plants

The use of coal for thermal energy purposes is one of the main options for obtaining electricity, although in recent decades there is "green energy" obtained by operating hydroelectric power plants, wind farms or the use of photovoltaic panels, of increasing importance. The technology of converting coal into energy has been applied for over a century, being justified both by the existence of world reserves and by the existence of technologies for extraction and generation of energy using this fuel [16].

The raw coal delivered from the extraction areas, is processed in the form of coal dust, dried at high temperatures and is introduced into the combustion process [16].

3.1. Radioactivity coal in Romania

Romanian lignite, extracted from the Oltenia basin, unlike other types of lignite extracted worldwide, is characterized by a moderate moisture content (40%) and a high ash content that can vary widely, reaching even maximums of 60%, characteristics that motivate the achievement of a lower caloric power, of 5.9 MJ / kg.[16-19].

In Romania, after 1990, the amount of electricity supplied by thermal power plants decreased, some power plants went bankrupt (the case of thermal power plants from Drobeta Turnu Severin, Doicești), others operate at breakdown parameters (Ișalnița thermal power plant), and others continued to operate at the initial parameters (Rovinari, Motru, Deva) investing in equipment to reduce the discharged pollutants to meet the requirements of environmental pollution wikipedia.ro.

The average annual quantities of ash and slag from a coal-fired power plant vary mainly with the installed capacity of the power plant, the quality and the degree of coal fragmentation. The main fraction resulting from combustion is ash $(70 \div 90\%)$ compared to slag and other resulting materials. In Romania, the ash dumps do not have high heights, but occupy large areas of land.

By the granulometric studies performed on the ash resulting from the main power plants in Romania, it results that the material in the composition of the dumps belongs to the category of fine sands and dust [17].

All coal assortments contain natural radioactive isotopes in certain concentrations. By burning them in the plant, a concentration of radioactive isotopes takes place in the combustion products, which are then discharged into the environment.

The table below presents the specific radioactive activities of the main radionuclides contained in the coal extracted from the mines in Romania [17].

Tab. 3.1 The specific radioactive activities of the main radionuclides contained in the coal extracted from the mines in Romania [17, 19].

Natural radioactive isotope	$\Lambda_{\rm s} ({\rm Bq/kg})$
⁴⁰ K	$30 \div 650$
²³⁸ U	$1 \div 100$
²³² Th	1 ÷ 50
²²⁶ Ra	1 ÷ 150

To produce 1 GW.year it consumes approx. 3 x 109 kg of lower coal. The coal contains 14C, 40K, uranium, thorium and all radionuclides in the decay chains. Also, radon from the disintegration chain of natural radionuclides is released by dislocation and granulation inside the combustion chamber, increasing the concentration of radon inside it.

The specific activity of the main radionuclides in fly ash emitted from coal-fired power plants in Romania is presented in the table below [17, 18, 19].

Tab. 3.2 The specific activity of natural radionuclides present in the fly ash emitted from coal-fired power plants in Romania [17, 18, 19]

Natural radioactive isotope	$\Lambda_{\rm s} ({\rm Bq/kg})$
40 K	160 ÷ 1200
238 U	2 ÷ 312
²²⁶ Ra	$3 \div 520$
²¹⁰ Pb	$10 \div 500$
²¹⁰ Po	$2 \div 170$
²³² Th	2 ÷ 170

It can be seen that the radioactivity of the ash resulting from the combustion of coal is $5 \div 10$ times higher than that of the primary fuel [17, 18, 19].

Worldwide, according to the literature [21 - 29], the average specific activity of 238U and 232Th of coal is approximately 20 Bq / kg, with values between 5 and 300 Bq / kg.[18]. During the combustion of coal, organic compounds are released which, due to the temperature in the furnaces, change to a gaseous state (water vapor and carbon

dioxide), while inorganic compounds are concentrated in the solid residue which also contains natural radionuclides existing in coal. Usually, the concentration of radionuclides in the ash increases about 10 times, (table 3.1 and 3.2 respectively).

3.2. Radioactivity of the ash samples resulting from the burning of coal in the ROMAG TERMO Drobeta Turnu Severin thermal power plant [20]

The ash samples were taken from the two existing ash dumps, arranged in the western part of ROMAG TERMO Drobeta Turnu Severin, from the surface of the dump to depths of $35 \div 40$ cm, in quantities of max. 1 kg per sample, noting the GPS coordinates of the locations of each sample. After drying, 100 grams of sample were prepared for spectrometric measurements.

13 samples were subjected to gamma spectrometric measurements (7 belonging to the first dump and 6 from the second) [17]. The measurements were made in the Gamma Spectrometry Laboratory of the Department of Life and Environmental Physics in IFIN-HH. The ORTEC gamma spectrometric system consists of a high-energy HPGe detector (1.9 keV FWHM at 1332 keV (Co-60)) and high efficiency (35%), introduced in a lead castle and associated electronics assisted by a computer running MAESTRO software, all of which make up a complex measurement system. For the identification and quantification of radioisotopes existing in the samples, the gamma spectrometer was calibrated in energy using isotopes, Cs-137, Co-60 and Co-57 and was calibrated in efficiency[20].

3.3. Experimental results and comments

The measured values of the specific radioactive activities for the samples taken from the two ash dumps arranged in the western part of ROMAG TERMO Drobeta Turnu Severin, for the identified radionuclides are given in tables 3.3 and 3.4.

Sampl. nr.	Sampl. depth. (cm)	Ac ²²⁸	Bi ²¹²	Pb ²¹²	Tl ²⁰⁸	Th ²³⁴	Ra ²²⁶	Pb ²¹⁴	Bi ²¹⁴	Pb ²¹⁰	K ⁴⁰	Cs ¹³⁷
1	$0 \div 8$	57	62	60	24	258	339	178	181	50	371	-
2	8-16	58	68	58	23	244	340	172	174	674	378	-
3	16-22	57	54	49	19	106	267	145	145	580	429	1.5
4	22-25	53	58	60	-	169	272	145	142	25	390	1.2
5	25-30	58	55	51	18	125	276	136	135	610	461	1.6
6	30-35	55	58	52	20	101	271	122	124	30	477	3.7
7	35-40	59	57	55	19	117	268	131	138	24	428	4
RSD^*		9 - 10	9-10	8-11	9-11	8-10	9-10	9-10	9-10	7-12	9-11	10-15
(%)												

Tab. 3.3.The results of the radioactivity measurements related to dump no.1. Specific activity of identified radionuclides (Bg/kg) [20].

RSD* - Relative Standard Deviation.

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Sample nr.	Sampling depth.	Ac ²²⁸	Bi ²¹²	Pb ²¹²	T1 ²⁰⁸	Th ²³⁴	Ra ²²⁶	Pb ²¹⁴	Bi ²¹⁴	Pb ²¹⁰	K ⁴⁰	Cs ¹³⁷
	(cm)											
1	0 ÷ 5	45	41	37	14	32	89	27	27	<30	578	15.7
2	5-10	44	54	36	16	32	32	57	23	<27	604	13
3	10-15	43	41	42	14	35	69	27	30	<30	604	9.7
4	15-20	55	47	49	18	45	55	36	41	<35	607	6.6
5	20-25	46	52	45	15	31	80	27	26	18	635	4.8
6	25-30	41	124	112	15	34	60	37	41	<35	600	<2.3
RSD*%		9 -10	9-10	8-11	9-11	8-10	9-10	9-10	9-10	7-12	9-11	10-15

Tab. 3.4 The results of the radioactivity measurements related to dump no. 2. Specific activity of identified radionuclides (Bq/kg) [20].

RSD* - Relative Standard Deviation.

The distribution of radioisotopes of interest (Ac228, Ra226 and K40) in the two ash dumps, depending on the sampling depth is shown in the graphs below:





In the literature [references 22-30] are reported values of specific activities (in Bq/kg), determined for ash samples from thermal power plants, worldwide, which fall into the following ranges: Ac^{228} (53 - 102), Ra^{226} (7 - 180), Th^{232} (34 - 290), U^{238} (10 - 79) and K^{40} (20 - 795).

4. Conclusions

From the experimental data obtained following the analysis of the radioactivity of the ash samples resulting from the combustion of lignite in the ROMAG TERMO plant, the following results resulted:- the specific activity values of the radionuclide Ra226 are higher in the ash dump no. 1, than the values related to the ash dump no. 2; the big difference between these values could be explained by the fact that the two ash dumps were formed in different periods of time from the ash resulting from different varieties of lignite that did not come only from the Husnicioara mine;

- the values of the specific activities of 40 K are higher than the values from the ash dumps no. 1 and 2; with two exceptions,

- the radionuclide concentrations in the ash samples analyzed, collected from the two dumps, are in the range of values reported in other scientific articles mentioned above (references 22 - 30) and in the publication UNSCEAR 2008 [21].

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