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Evaluation of Radiation Security of Coal-Mining and Thermal Power Waste Products

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Abstract

The concentrators of natural radionuclides (NR) include waste products from coal mining and thermal power industries. The aim of the work was to determine the radionuclide (^{226}Ra, ^{232}Th, and ^{40}K) composition of fractions of coal mining waste and fuel ash and slag and their compliance with the radiation safety standards of Ukraine and international radiological indicators. All wastes belong to the I class of radiation hazard (C_{ef} <370 Bq/kg) and can be used in construction without restrictions. According to international radiological indicators, the value of the activity utilization index is exceeded for almost all wastes under study. The values of the internal hazard index and gamma index of the Olkhovatska mine burnt rock (fraction <0.63 mm) exceed the recommended limits. The values of radium equivalent activity Ra_{eq} and the alpha index indicate that the studied ash-slag and dump rock do not constitute a danger of increased emanation of radon and its progeny into the room air. The absorbed dose rate in the open air for the studied wastes and the annual effective equivalent dose are higher than the world average, respectively: 58 nGy/h and 0.07 mSv, but lower than the value recommended by the IAEA for the population, 1 mSv/year.

***Keywords**: Coal waste; Effective specific activity; Fuel slag; Radiation hazard class; Radiation hazard indices; Radionuclides.*

1. Introduction

Disposal of recoverable industrial waste can contaminate the resulting product and affect negatively the organisms of the workers involved in the process. The problem of obtaining environmentally friendly materials is especially important when using waste products that concentrate natural radionuclides (NR), dangerous for human health and the environment. Such concentrators of NR also include waste from coal mining and thermal power industries.

Radioecological monitoring of raw materials and industrial waste is currently topical. Identification of factors that determine the content of NR (^{238}U, ^{226}Ra, ^{232}Th and ^{40}K) in the raw materials, their behaviour during processing, will allow us to predict their content in the final material, make adjustments and avoid additional public exposure. Systematic radiological monitoring of coal and its combustion products is required. According to the Scientific Committee on Action of Atomic Radiation at the UN (UNSCEAR) [1] the average specific activities of NR in coal are, Bq/kg: ^{238}U – 37, ^{226}Ra – 35, ^{232}Th – 30, ^{40}K – 400. A relatively high content of NR was recorded at some coal deposits of the USA, China, Russia and Serbia [2]. In Nigerian coal, the average specific activities were, Bq/kg: ^{226}Ra – 8.2, ^{232}Th – 7.0, ^{40}K – 27.4 with average values of radium equivalent activity of Ra_{eq} 20.26 Bq/kg and external hazard index I_{ex} 0.05, internal hazard index I_{in} 0.08 and gamma index 0.14 [3]. The authors from China [4] determined the average specific activity of ^{238}U, ^{232}Th, ^{226}Ra, ^{210}Pb, ^{210}Po and ^{40}K, Bq/kg: 46.6; 87; 38.8; 42.6; 23.8 and 540 for coal; 85.8; 139; 72; 122; 78.5 and 758 for fly ash; 93.2; 144; 76.3; 636; 386 and 747 for solid flue gas particles. NR in coal strata are associated with both inorganic (clayey, various sedimentary rocks) and organic substances. Th binds with zircon $ZrSiO_4$. Based on the analysis of the distribution of natural gamma radiation of coal and roofing rocks of 7 coal mines in China, the principle of recognition of coal mines using the method of natural gamma radiation is developed [5].

The results of studying the radioactivity of coal waste are of interest [6–8]. The radioactivity of dump rocks in Donbass mines exceeds background values; the variation in the intensity of gamma radiation at dumps is 11–25 μR/h. In weakly metamorphosed rocks typical of Donbass, the ^{40}K and ^{226}Ra isotopes are present [8]. The dust blown out by the wind from the surface of the dumps may contain NRs, which are inhaled into the human body and produce internal irradiation. The measured average specific activities of NR in the dust of South Africa heaps are equal to, Bq/kg: ^{226}Ra – 53.6, ^{238}U – 15.2, ^{232}Th – 6.6, ^{40}K – 278.5 [9]. In the dumps around the Maigang coal mine in the north-eastern Nigeria, the average specific activities were, Bq/kg: ^{226}Ra – 11.90, ^{232}Th – 17.7, ^{40}K – 70.4 [10]. With such activity values, the waste does not pose a radiological risk and can be utilized in other industries. By the values of the indices of radiation, external and internal hazard and absorbed dose rate, the suitability of rocks and deposits of coal mines of Silesia for construction is justified [11]. The danger of radon emanation arises when the concentration coefficient of NR is 2–10 compared to coal [12]. The lack of regulatory documents causes significant difficulties in organizing the systems for monitoring the content of NR in coal. The implementation of coal radiation monitoring systems at the stage of field development will minimize the occurrence of NR in the fuel cycle [13].

During combustion of the organic part of coal, NR is concentrated in the combustion products – waste from thermal power plants (TPP): ashes, slags, fly ash emitted into the atmosphere. The creation of an effective coal quality control system taking into account the radiation-hygiene factor and ensuring the protection of the environment and public health is relevant [14]. The maximum concentration of NR is recorded for fly ash; the activity of the NR of the uranium series in fly ash is 1442–12641 Bq/kg compared to coal 813–2609 Bq/kg [15]. In the soil near the TPP, where fly ash lodges, NR: ^{226}Ra, ^{232}Th, ^{40}K, ^{235}U, ^{238}U and ^{137}Cs were found [16]. The radionuclide composition of ash and slag correlates with their chemical composition; therefore, it is necessary to determine the elemental and mineral composition of thermal power waste [17]. The most likely mechanism for accumulation of NR is a heterovalent isomorphic substitution in the structures of slag minerals in a crystalline and amorphous state [18].

Coal mining waste can be used as raw materials for the construction industry and production of cement [19-20]. Producing fuel and energy from both renewable sources and various types of waste are prospective directions for development [21]. The chemical composition of coke varies depending on the particle size [22]. Therefore, it is necessary to study not only the average waste samples, but also their fractions.

The purpose of the research is to determine the radionuclide composition of coal waste fractions and fuel ash and slag, and their compliance with the radiation safety standards of Ukraine and international radiological indicators.

2. Research methodology

Gamma spectrometric analysis of slag was performed using a SEG-001 "AKP-S" scintillation gamma spectrometer with an error in measuring activity of ≤25 %. The measurement results were processes with the help of Akwin software. The results of gamma-ray spectrometric analysis are presented in Table 1.

3. Results and discussion

3.1. Compliance of coal-mining and thermal power waste products with the radiation safety standards of Ukraine

The radiation hazard criterion for construction, technical materials, by-products and industrial wastes according to the norms of radiation safety of Ukraine [23] is the effective specific activity of NR – C_{ef}, which is defined as the weighted sum of the specific activities of radium-226 (C_{Ra}), thorium-232 (C_{Th}) and potassium-40 (C_K) according to the formula

$$C_{ef} = C_{Ra} + 1.31C_{Th} + 0.085C_K, \text{ Bq/kg}, \quad (1)$$

where 1.34 and 0.09 are respectively weighted coefficients for thorium-232 and potassium-40 relative to radium-226.

By the value of C_{ef}<370 Bq/kg (Table 1) all wastes under study belong to the first class of radiation hazard and can be used in construction without restrictions.

Table 1. The results of gamma-spectrometric analysis of coal waste and fuel ash-slag

Fraction, mm	C_{ef}, Bq/kg	C_{ef}, Bq/kg (contribution to summary activity, %)		
		^{40}K	^{226}Ra	^{232}Th
Ash-slag, Slavianska TPP, Donetsk region				
Average sample	259	785 (82.2)	101 (10.6)	69.5 (7.3)
10–20	264	792 (82.0)	100 (10.4)	73.5 (7.6)
5–10	269	807 (82.0)	104 (10.6)	72.9 (7.4)
< 5	237	745 (83.0)	83.4 (9.3)	68.6 (7.6)
Ash-slag, Eskharovska HPP, Kharkiv region				
Ash	238	686 (81.1)	95.0 (11.2)	64.4 (7.6)
Ash-slag	244	732 (81.9)	97.8 (10.9)	64.0 (7.2)
Slag	236	750 (83.1)	89.5 (9.9)	62.8 (7.0)
Ash-slag, Zmiivska HPP, Kharkiv region				
Ash-slag	254	761 (81.9)	101 (10.9)	67.3 (7.2)
Coal-mining rocks Burnt rock of Olkhovatska mine, Donetsk region				
Average sample	251	1050 (88.1)	73.7 (6.2)	67.7 (5.7)
> 20	240	968 (87.7)	67.4 (6.1)	68.9 (6.2)
10–20	257	1010 (87.2)	77.2 (6.6)	71.8 (6.2)
5–10	258	1110 (88.7)	69.4 (5.5)	71.7 (5.7)
2.5–5	264	1130 (88.5)	74.5 (5.8)	71.7 (5.6)
1.25–2.5	278	1110 (87.3)	84.1 (6.6)	76.4 (6.0)
0.63–1.25	270	1070 (87.2)	84.4 (6.9)	72.3 (5.9)
< 0.63	305	912 (82.0)	111 (10.0)	89.3 (8.0)
Burnt rock of Tcheluskintsev mine, Donetsk region				
Average sample	225	610 (80.0)	92.1 (12.0)	61.6 (8.0)
Non-burnt rock of Khmelnytska mine, Luhansk region				
Average sample	172	815 (90.1)	45.9 (5.1)	43.7 (4.8)
Non-burnt rock of Sverdlova mine, Luhansk region				
Average sample	121	470 (86.8)	40.8 (7.5)	30.6 (5.7)
Non-burnt rock of Frunze mine, Luhansk region				
Average sample	176	839 (90.2)	46.1 (5.0)	44.8 (4.8)
Non-burnt rock of Vodianska mine, Donetsk region				
Average sample	193	640 (84.3)	56.8 (7.5)	62.7 (8.2)
Non-burnt rock of Belytska mine, Donetsk region				
Average sample	210	746 (85.4)	68.6 (7.8)	59.2 (6.8)
Non-burnt rock of Pavlohradska mine, Dnipropetrovsk region				
Average sample	184	171 (53.4)	82.3 (25.7)	66.6 (20.8)

3.2. Activity of the studied samples

The radionuclide composition of the samples under study has some peculiarities. When organics and carbon particles are burned out, NR concentrates in the remaining inorganic part. This applies to fuel ash and slag and burnt coal.

Fuel ash-slag. The values of the effective specific activity of the studied fractions of ash-slag are nearly the same. The values of the contributions of individual NR in C_{ef} are close (Table 1). C_{ef} value of investigated ash-slag waste exceeds the average C_{ef} for fuel slag (194 Bq/kg) and ash (204 Bq/kg) in the Community of Independent States (CIS) [24], but lower than the same value for ash- slag of the Prydneprovska hydro power plant (HPP) (366 Bq/kg) and Kryvorozhska HPP (352 Bq/kg) [25]. The investigated ash and slag have an increased C_{ef}

compared with the average value for construction materials in Ukraine (106 Bq/kg) and the CIS (93 Bq/kg), however, the range of its variation is much narrower than for construction materials [24].

The ash and slag fractions of the Slavianska TPP can be characterized by the content of individual NR. In order to reduce the ratio (%) of the maximum variation in the specific activities of individual NRs in ash-slag fractions from C_i of the average sample, the radionuclides can be arranged in a series: $^{226}Ra > {}^{40}K > {}^{232}Th$.

Coal-mining rocks. The distribution of NR by the volume of the heaps may depend on a number of factors. One of the main factors is water leaching, which is determined by the wind rose of the area. Thus, the distribution of NR in the surface layer of coal rock should vary depending on the compass point. Subsequent migration of NR to the depth is determined by the solubility of the formed compounds, the anionic composition of the coal rock, pH, redox properties, and other factors.

Coal-mining burnt rocks of Olkhovatska mine. A fraction >20 mm, is the most radiation-friendly. A fraction of <0.63 mm is characterized by the maximum value of C_{ef}. The increase of C_{ef} of rock particles with a size of <2.5 mm and especially <0.63 mm are associated with an increase in the specific activity of ^{226}Ra (Table 1). The study of the activity of individual fractions of the Olkhovatska mine burnt coal rock determines a number of decreases in the ratio (%) of the maximum dispersion of NR specific activities for ash-slag fractions from C_i of an average sample: $^{226}Ra > {}^{232}Th > {}^{40}K$.

Non-burnt coal-mining rocks have an organic component in their composition, hence the smaller values of C_i of individual NR and C_{ef}. The contribution of NR to the summary activity (C_{sum}) of samples is different. The heap rock of Pavlohradska mine, for which the maximum contributions of ^{226}Ra and ^{232}Th to the summary activity are recorded, is the most dangerous in terms of the emanations of the ^{222}Rn and ^{220}Rn isotopes. Hence the need for radiation monitoring of heaps, pre-empting their development.

3.3. Correspondence of coal-mining and thermal power waste products to international radiological indicators

Numerous criteria for assessing the radiological load on the population from exposure to radiation in various environmental components are presented in the literature [1, 26–30].

Hazard indices. Radium equivalent activity Ra_{eq} is used to summarize the hazards associated with individual NR when they are not uniformly distributed in the environment. Ra_{eq} is expressed as the weighted sum of activities ^{226}Ra, ^{232}Th and ^{40}K in Bq/kg, based on the assumption that 370 Bq/kg of ^{226}Ra, 259 Bq/kg of ^{232}Th, and 4810 Bq/kg of ^{40}K are characterized by the same gamma dose rate [1]:

$$Ra_{eq} = C_{Ra} + 1.43\,C_{Th} + 0.077C_K,\ \text{Bq/kg}. \qquad (2)$$

The value of Ra_{eq} should not exceed 370 Bq/kg [27].

The external hazard index I_{ex} evaluates the hazard at the expense of external exposure from ^{226}Ra, ^{232}Th and ^{40}K in the samples under study [28]:

$$I_{ex} = \frac{C_{Ra}}{370} + \frac{C_{Th}}{259} + \frac{C_K}{4810}. \qquad (3)$$

The internal hazard index I_{in} evaluates the risk for the respiration organs from the internal action of radon and its short-living progeny [28]:

$$I_{in} = \frac{C_{Ra}}{185} + \frac{C_{Th}}{259} + \frac{C_K}{4810}. \qquad (4)$$

Gamma index I_γ is calculated by the equation [28]:

$$I_\gamma = \frac{C_{Ra}}{300} + \frac{C_{Th}}{200} + \frac{C_K}{3000}. \qquad (5)$$

I_γ is used to identify materials that are of interest for construction. For the materials used in large volumes, for example, for concrete $I_\gamma \leq 1$, which corresponds to an annual effective dose of less than or equal to 1 mSv, $I_\gamma \leq 0.5$ corresponds to an annual effective dose of less than or equal to 0.3 mSv [28].

Alfa index I_α evaluates emanation of radon from materials [28]:

$$I_\alpha = \frac{C_{Ra}}{200}. \qquad (6)$$

When the activity of ^{226}Ra in the material is above 200 Bq/kg, the concentration of radon entering the room air can be equal to 200 Bq/m^3. $I_\alpha \leq 1$ corresponds to activity of ^{226}Ra not exceeding 200 Bq/kg.

Activity utilization index AUI determines the dose rate in the air for various combinations of ^{226}Ra, ^{232}Th and ^{40}K in industrial waste products. The index is calculated on the basis of the specific activities of NR C_i and fractional contributions of NR f_i to the total dose rate of gamma radiation in the air by the ratio [10]:

$$AUI = \frac{C_{Ra} \cdot f_U}{50} + \frac{C_{Th} \cdot f_{Th}}{50} + \frac{C_K \cdot f_K}{500}. \quad (7)$$

The fractional contributions of f_U, f_{Th} and f_K to the total dose rate of gamma radiation in the air from ^{226}Ra, ^{232}Th, and ^{40}K are 0.462, 0.604, and 0.041; the average specific activities of ^{40}K, ^{232}Th, and ^{226}Ra in soils are, Bq/kg: 500, 50, and 50 [27].

According to the UNSCEAR recommendations [1] the values of I_{ex}, I_{in}, I_γ, I_α and *AUI* should not exceed 1, in this case materials can be used in construction without a radiological threat to the population. The results of calculations of the radiation hazard indices are shown in Table 2.

The Ra_{eq} value of all samples does not exceed 370 Bq/kg, which corresponds to the external dose of 1.5 mSv/year [27]. The external hazard index of all samples is $I_{ex} \leq 1$. The internal hazard index for two samples subjected to heat treatment slightly exceeds 1 (highlighted in Table 2). The gamma index is $I_\gamma \leq 1$ for the samples excluding the fraction <0.63 of the Olkhovatska mine burnt rock. The alpha index is $I_\alpha \leq 1$ for all samples, which corresponds to activity of ^{226}Ra not exceeding 200 Bq/kg. A significant excess was found for the *AUI* activity utilization index practically for all studied rocks and ash-slag. For the fraction <0.63 of the Olkhovatska mine burnt rock *AUI* is >2.

Radiation doses. Air-absorbed dose rate D_R is calculated by the following formula at the expense of gamma radiation of the environmental NR [29]:

$$D_R = 0.462\, C_{Ra} + 0.604 C_{Th} + 0.0417 C_K, \text{ nGy/h} \quad (8)$$

The conversion factors of 0.462, 0.604 and 0.0417 Bq/kg respectively for ^{226}Ra, ^{232}Th and ^{40}K, determined by UNSCEAR [29], are used to estimate D_R for 1 m above ground level as a result of gamma radiation of ^{226}Ra, ^{232}Th and ^{40}K contained in the soil.

Annual effective dose equivalent AEDE determines the risk of exposure of absorbed dose rate to a person. Conversion factors are: 0.70 Sv/Gy converts D_R into an effective dose; the coefficient of stay in the open air is 0.2 [1]. *AEDE,* while staying in the open air, is estimated by the formula:

$$AEDE = 1.21 \cdot 10^{-3}\, D_R, \text{ mSv/y} \quad (9)$$

The D_R values of the production waste under study are higher than the world average of 58 nGy/h, presented in [29]. Similarly, the *AEDE* value for most waste samples is higher than the global average effective dose obtained in the open air, 0.07 mSv [1]. However, the calculated *AEDE* values are less than the annual effective dose recommended by the International Atomic Energy Agency (IAEA) for the population, 1 mSv/year [31].

Excess lifetime cancer risk ELCR at radiation is calculated on the basis of *AEDE* value by the equation [30]:

$$ELCR = AEDE \cdot DL \cdot RF, \quad (10)$$

where *DL* – is life expectancy of 70 years; *RF* – is a factor of risk, equal to 0.05 1/Sv for stochastic effects for any population [30].

The *ELCR* values for some samples of the studied waste are higher than the world average of $0.29.10^{-3}$ [1], but below the limit of 0.05 established by the International Commission on Radiation Protection (ICRP) [30]. Thus, the likelihood of cancer for the population utilizing the studied coal mining and thermal power waste products is altogether insignificant.

Table 2. Indices of radiation hazard, radiation dozes and carcinogenic risk coal-mining and thermal power waste products

Fraction, mm	Radiation hazard indices						Doses		ELCR ·10⁻³
	≤370 Bq/kg	(≤1)							
	Ra $_{eq}$, Bq/kg	I_{ex}	I_{in}	I_{γ}	I_{α}	*AUI*	*AEDE*, mSv/year	D_R, nGy/h	
Ash and slag, Slavianska TPP, Donetsk region									
Average sample	260.83	0.70	0.98	0.95	0.51	**1.84**	0.15	121.4	0.53
10–20	266.09	0.72	0.99	0.96	0.50	**1.88**	0.15	123.6	0.53
5–10	270.4	0.56	**1.01**	0.98	0.52	**1.91**	0.15	125.7	0.53
< 5	238.86	0.65	0.87	0.87	0.42	**1.66**	0.13	111.0	0.46
Ash and slag, Eskharovska HPP-2, Kharkiv region									
Ash	239.91	0.65	0.90	0.87	0.48	**1.71**	0.13	111.4	0.46
Ash and slag	245.68	0.66	0.93	0.89	0.49	**1.74**	0.14	114.4	0.49
Slag	237.05	0.64	0.88	0.86	0.45	**1.65**	0.13	110.6	0.46
Ash and slag, Zmiivska HPP, Kharkiv region									
Ash and slag	255.84	0.69	0.96	0.93	0.51	**1.81**	0.14	119.0	0.49
Coal mining rocks. Burnt rock of Olkhovatska mine, Donetsk region									
Average sample	251.36	0.68	0.88	0.93	0.37	**1.58**	0.14	118.7	0.49
> 20	240.46	0.65	0.83	0.89	0.34	**1.53**	0.14	113.1	0.49
10–20	257.64	0.70	0.90	0.95	0.39	**1.66**	0.15	121.2	0.53
5–10	257.40	0.70	0.88	0.96	0.35	**1.60**	0.15	121.7	0.53
2.5–5	264.04	0.71	0.91	0.98	0.37	**1.65**	0.15	124.8	0.53
1.25–2.5	278.82	0.75	0.98	1.00	0.42	**1.79**	0.16	131.3	0.56
0.63–1.25	269.78	0.73	0.96	0.88	0.42	**1.74**	0.15	127.3	0.53
< 0.63	308.92	0.83	**1.13**	**1.12**	0.56	**2.18**	0.17	143.2	0.60
Burnt rock, Tcheluskintsev mine, Donetsk region									
Average sample	227.16	0.61	0.86	0.82	0.46	**1.65**	0.13	105.2	0.46
Non-burnt rock of Khmelnytska mine, Luhansk region									
Average sample	171.15	0.46	0.59	0.64	0.23	**1.02**	0.10	81.6	0.35
Non-burnt rock of Sverdlova mine, Luhansk region									
Average sample	120.75	0.33	0.44	0.45	0.20	0.79	0.07	56.9	0.25
Non-burnt rock of Frunze mine, Luhansk region									
Average sample	174.77	0.47	0.60	0.66	0.23	**1.04**	0.11	88.3	0.39
Non-burnt rock of Vodianska mine, Donetsk region									
Average sample	195.74	0.53	0.68	0.72	0.28	**1.33**	0.11	90.8	0.39
Non-burnt rock of Belytska mine, Donetsk region									
Average sample	210.70	0.57	0.75	0.77	0.34	**1.41**	0.12	98.6	0.42
Non-burnt rock of Pavlohradska mine, Dnipropetrovsk region									
Average sample	190.71	0.52	0.74	0.66	0.41	**1.58**	0.10	85.4	0.35

4. Conclusion

The presence of NR was found in coal-mining and thermal power waste products: ^{226}Ra, ^{232}Th, and ^{40}K, as well as variation in their activity by particle size fractions. The main contribution to the C_{ef} value is made by radionuclides ^{226}Ra and ^{232}Th. The largest variation in specific activities by fractions of fuel ash and slag and burnt coal is characteristic of ^{226}Ra. According to C_{ef} value all coal-mining and thermal power waste products under study belong to the I class of radiation hazard (C_{ef} < 370 Bq/kg) and can be used in construction without restrictions.

Clarification of the characteristics of the waste gamma radiation when calculating the radiation hazard indices showed that the gamma radiation of the burnt rock from the Olkhovatska mine (fraction <0.63 mm) exceeds the recommended limits and can cause irradiation with an effective dose of more than 1 mSv/year. An excess of the activity utilization index was found practically for all studied rocks and ash-slags. According to the value of the equivalent activity of ^{226}Ra and the alpha index, the studied ash-slags and rocks do not constitute a danger of increased emanation of radon and its progeny into the room air. The absorbed dose rate in the open air for the wastes under study and the annual effective equivalent dose are higher than the world average, respectively: 58 nGy/h and 0.07 mSv, but lower than the value recommended by the IAEA for the population, 1 mSv/year. The excess lifetime carcinogenic risk is below the 0.05 limit established by the ICRP.

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