Natural radioactivity of wastes

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Abstract. By-products of the combustion of coal (wastes) are often used for various types of construction (dwellings, roads, etc.). The legal regulations (The Ordinance of the Council of Ministers of 2 January 2007 "On the requirements for the content of natural radioactive isotopes of potassium K-40, radium Ra-226 and thorium Th-228 in raws and materials used in buildings for the residence of people and livestock, as well as in the industrial by-products used in the construction, and the control of the content of the aforementioned isotopes" – Law Gazette no. 4/2007 item 29) are in force in Poland. The regulations permit the possibility of utilization of raws and by-products basing upon the level of the natural radioactivity of the examined raws and materials. The article is a survey of the results obtained during the measurements of many types of raws and building materials for almost 30 years by the network of the laboratories in Poland. It is based upon the results stored in the database of the Central Laboratory for Radiological Protection (CLRP), Warsaw. The article tends to outline the radioactivity of the waste materials with respect to other raws and materials used in the construction industry. The article shows the possibilities for the use of by-products originating in the power stations and heat- and power stations (mainly ashes, slag and hinter) in the construction of dwellings and roads.

Key words: natural radioactivity • waste • building materials • raw materials • radioactivity

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Introduction

The values of the concentration of natural radionuclides in the particular components of our environment can differ by the orders of magnitude depending on the natural or man-made local conditions. The considerable differences of the isotopic composition of the particular components are observed as well.

The soils in Poland contain the following scope of natural (potassium K-40, radium Ra-226 and thorium Th-228) radionuclide concentration [2]:
– K-40 scope 60–1028 Bq/kg, mean value 408 Bq/kg,
– Ra-226 scope 4.2–116 Bq/kg, mean value 25.0 Bq/kg,
– Th-228 scope 3.6–82.8 Bq/kg, mean value 23.4 Bq/kg.

The by-products that are further recycled in the various branches of the industry usually have increased natural radioactivity levels due to their concentration.

Basing upon the results of the measurements of the radioactivity of raws and building materials carried out by the CLRP and over 30 other institutions in Poland, the Polish database of the results of the measurements was established. The database contains the results of the measurements since 1980. Since the beginning of the network of such measurements in Poland all measurements (in all institutions) are carried out according to the methods elaborated in the CLRP and using unique instruments elaborated in the CLRP (three-channel gamma ray spectrometer AZAR/MAZAR).

Legal regulations

Wastes from a power plant (by-products of the combustion of coal) are used in the production of building materials. Methodology of testing raw materials and building materials (including secondary raw-materials), was well described in the Instruction 234/2003 by the Building Research Institute, titled "Testing of the natural radioactivity of raw materials and building materials".

The Instruction contains procedures connected with the protection against ionizing radiation originating from the natural radioactive elements occurring in raw materials and industrial by-products of mineral origin, used in the production of building materials and goods. Moreover, the Instruction gives an inventory of natural raw materials and industrial by-products of mineral origin, building materials and goods subject to control, basic requirements, methods of testing concentrations of natural radioactive elements in materials, as well as principles for interpreting the results of tests and for conformity assessment.

The Ordinance of the Council of Ministers of 2 February 2007, is a fundamental law defining requirements for raw materials and building materials in different construction fields. It is titled "On requirements concerning concentrations of natural radioactive isotopes of potassium K-40, radium Ra-226 and thorium Th-228 in raw materials and materials used in buildings for dwelling of people and livestock, and in industrial by-products used in construction, with control of content of these isotopes" and was published in the Polish State Gazetteer (Law Gazette no. 4/2007 item 29).

The ordinance classifies the possibility of the use of raws and building materials in various types of the constructions by the determination of two parameters:

I. Activity coefficient f_1, describing the content of natural radioactive isotopes (it indicates exposure of the entire body to gamma radiation), defined by the following formula:

$$f_1 = \frac{S_K}{3000} + \frac{S_{Ra}}{300} + \frac{S_{Th}}{200} \quad (1)$$

where: S_K, S_{Ra}, S_{Th} are the respective radioactive concentrations of potassium K-40, radium Ra-226 and thorium Th-228 expressed in Bq/kg.

II. Activity coefficient f_2, which describes the content of radium Ra-226 (it indicates the exposure of the lung tissue to alpha radiation emitted by radon decay products, which are absorbed through the respiratory tract into human body):

$$f_2 = S_{Ra} \quad (2)$$

Depending upon the values of the two coefficients, the investigated material can be used in various types of construction. Thus, the values of coefficients f_1 and f_2 should not exceed by more than 20%:

1. $f_1 = 1$ and $f_2 = 200$ Bq/kg regarding the raws materials and building materials used in buildings for dwelling of people or livestock.
2. $f_1 = 2$ and $f_2 = 400$ Bq/kg regarding the industrial wastes used in surface objects constructed on the built-up areas, or designed for built-up in the local urbanization plans, or for levelling of such areas.
3. $f_1 = 3.5$ and $f_2 = 1000$ Bq/kg regarding the industrial wastes used in surface parts of the objects not mentioned in point 2 and for levelling of the areas not mentioned in point 2.
4. $f_1 = 7$ and $f_2 = 2000$ Bq/kg regarding the industrial wastes used in the underground parts of the objects mentioned in point 3, and the underground constructions, including railway and road tunnels, excluding the industrial wastes used in underground mining pits.

Additionally, for the use of the industrial wastes for the levelling of the areas mentioned in points 2 and 3, and for the construction of roads, sport and recreational objects, it is ensured (taking into account the values of f_1 and f_2), the reduction of the absorbed dose rate at a height of 1 m above the area, road or object to the value not exceeding 0.3 mGy/h, in particular by adding the layer of other material.

In this paper it was analysed, if industrial by-products (ash and slag) could be used for the construction of houses and roads, in other words, if activity indexes f_1 and f_2 calculated for these by-products meet the criteria specified in the quoted Order of the Council of Ministers.

The results of the measurements of natural radioactivity in raws and wastes in the years 1980–2009

The hard coal is characterized by the concentration of the natural radionuclides close to the average concentration in soil in Poland.

The combustion of coal in power and heat plants effects in the multiple increase of the concentration of natural radionuclides in fly ashes and slag. The degree of the concentration depends upon the quality of the combusted coal, combustion efficiency, types of boilers and many other factors.

The lowest concentrations are found in slags from power plants, in which an average degree of enrichment in radioactive isotopes in relation to the content in coal is approx. 3.1. In turn, the highest values of concentrations are found in ashes from power plants and cenospheres, where the degree of enrichment is approx. 4.0. Only in gypsum from power plants the concentrations of natural radionuclides are lower than in the combusted coal (see Table 1).

Similarly, high concentrations of natural radionuclides occur in some by-products from the metal industry. For comparison, in Table 2 are given values of the concentration of natural radionuclides in blow furnace slags, and slags from copper and nickel smelting.

The highest concentrations are found in slags from copper smelting.

The possibilities for the use of wastes in the construction of dwellings

Because people spent in their dwellings approximately 80% of lifetime, legal regulation concerning parameters of suitability of raw materials and building materials used for housing construction is fully justified. Activity indexes f_1 and f_2 are calculated to evaluate the suitability of materials.

Table 1. Concentrations of natural radioactive isotopes: potassium K-40, radium Ra-226 and thorium Th-228 in coal combustion by-products in the years 1980–2009

Type of material or waste	Number of samples	Concentration (Bq/kg)					
		K-40		Ra-226		Th-228	
		Average	Scope (min–max)	Average	Scope (min–max)	Average	Scope (min–max)
Coal	469	198.95	0–776	28.19	2.0– 90.5	22.13	0–95
Power-plant ashes	11 730	702.65	6.6–1664	120.11	0.1–876	91.06	1.0–206
Power-plant slag	2641	583.77	4.9–1436	87.97	4.6–468.7	73.12	0.1–185
Ash-slag mixture	1319	597.21	29.8–1146	97.97	10.0–286	73.55	4.0–155
Alumino-silicaeous cenospheres	158	859.97	58.8–1589.5	107.91	17.5–255.9	84.18	11.2–140
Power-plant gypsum	86	41.40	2.2–147.4	9.40	0.1–72.0	3.70	0.1–30

Table 2. Concentrations of natural radioactive isotopes of: potassium K-40, radium Ra-226 and thorium Th-228 in metallurgical industry by-products

Type of material or waste	Number of samples	Concentration (Bq/kg)					
		K-40		Ra-226		Th-228	
		Average	Scope (min–max)	Average	Scope (min–max)	Average	Scope (min–max)
Copper-smelt slag	80	886.46	615.4–1250.6	316.78	236.6–517.8	54.16	25.7–183
Nickel-smelt slag	3	604.70	78.1–888	234.57	16.7–364	44.90	7.9–82
Blast-furnace (boiler) slag	368	192.33	18.0–1400	115.33	12.4–351.1	34.55	2.2–115

Based on tests of natural radioactivity of raw materials and building materials made to date, Table 3 provides information on a number of samples, whose activity indexes f_1 and f_2 conform to the standards for use in housing construction (according to the Order of the Council of Ministers from 2007). A percentage share was also given

Table 3. Numbers of samples of raw materials suitable for housing construction – activity indexes $f_1 < 1.2$ and $f_2 < 240$ Bq/kg

Natural or secondary raw material	Number of samples	Samples with $f_1 < 1.2$ and $f_2 < 240$ Bq/kg	
		Number	%
Raw materials of natural origin			
Gravel	17	17	100.0
Raw materials of industrial origin			
Ash from power generation	11 730	8905	75.9
Ash	666	570	85.6
Slag from power generation	2641	2571	97.3
Slag from copper smelting	80	1	1.3
Slag from nickel smelting	3	1	33.3
Blast furnace slag	368	365	99.2
Slag	1312	1211	92.3
Fly ash-bottom ash mixture	1319	1275	96.7
Alumino-silicaeous cenospheres	158	142	89.9
Gypsum from power generation	86	86	100.0
Gypsum	207	207	100.0
Aggregate	70	67	95.7
Doped aggregates	6	6	100.0
Aggregate from copper-smelting slag	35	1	2.9
Slag aggregate	14	14	100.0
Ash aggregate	2219	2184	98.4
Coal waste	40	40	100.0
Buildings materials			
Cement	135	135	100.0
Lightweight concrete	2934	2934	100.0
Traditional concrete	105	104	99.0
Brick	1383	1382	99.9
Roof tile	67	67	100.0
Styro-ash-concrete	3	3	100.0
Concrete hollow brick	340	340	100.0
Quick lime	104	104	100.0

for each type of raw material and building material, of samples that might be directly used in housing construction in concordance with Polish regulations.

As can be seen from the above table, almost 100% of natural origin and building materials can be used in the construction of dwellings. In the case of the raws of industrial origin the best activity coefficient values have: gypsum (100% of the investigated samples can be used in the construction of dwellings), slag aggregate (100%), doped aggregate (100%) and ash aggregate (98.4%), boiler slag (99.2%), power-plant slag (97.3%).

On the other hand, the lowest possibility of using as industrial by-products in housing is found for slags from copper smelting (only 1.3% of samples are suitable for housing construction) and aggregates from copper smelting slag (2.9%).

The use of wastes in road construction

The wastes of industrial origin are primarily used in the road construction. According to the regulations, the requirements for the activity coefficient values for road construction are less demanding and amount to $f_1 < 2.4$ and $f_2 < 480$ Bq/kg (taking into account 20% excess of the limit values given in the Ordinance of the Council of Ministers – 2007).

Based on the results of measurements collected in our data base, Table 4 provides a number of samples whose activity indexes are $f_1 < 2.4$ and $f_2 < 480$ Bq/kg and which are not suitable for housing construction (that is $f_1 > 1.2$ or $f_2 > 240$ Bq/kg).

Considering that waste suitable for housing construction ($f_1 < 1.2$ and $f_2 < 240$ Bq/kg) may of course be used in road construction, we see from the above table, that the majority of raw materials may be in 100% used in road construction.

Also, of course, materials such as slag from copper smelting (98.7% samples may be used only in road construction – cannot be used in housing construction), aggregate from copper smelting slag (97.1%) or slag from nickel smelting (66.7%) may be utilized, first of all, in road construction.

Estimation of the population exposure

According to the data published in the UNSCEAR (United Nations Scientific Committee on the Effects of Atomic Radiation) reports, a standard man spends, on the average, 20% of the year outdoors (ca. 1750 h).

If, resulting from the application of the material with enhanced natural radioactivity content, a person spends part of this time in places with elevated dose rate (over the natural background), on additional exposure to the whole body caused by the external ionizing radiation is possible. The radionuclides contained in such materials can also migrate to the human and animal alimentary chain, thus increasing the internal exposure.

The population exposed to ionizing radiation can be divided into three groups: road construction workers, professional drivers and other users of the roads. For calculations of exposure, it is assumed that for road construction workers and professional drivers the annual time of exposure is 2100 h per year, while for other users is not more than 750 h.

The gamma dose rate at a height of 1 m over the unlimited flat surface of the terrain of the average density of $r = 1.6$ g/cm^3 is determined semi-empirically by the formula [1]:

$$D = 0.043\,S_K + 0.43\,S_{Ra} + 0.66\,S_{Th} \qquad (3)$$

where S_K, S_{Ra}, S_{Th} – concentrations in Bq/kg of respectively K-40, Ra-226 (in radioactive equilibrium with daughter nuclides) and Th-228 (in radioactive equilibrium with daughter nuclides).

Based on measured values of concentrations of natural radioactive isotopes in wastes, taking into account the above described time of residence for three groups

Table 4. The number of samples of wastes, which can be used in road construction – the activity coefficients not exceeding the values $f_1 < 2.4$ and $f_2 < 480$ Bq/kg – not applicable for the construction of dwellings

Type of waste	Number of samples	Percentage of samples usable in the construction of dwellings	Samples for which $f_1 < 2.4$ and $f_2 < 480$ Bq/kg	
			Quantity	%
Power-plant ash	11 730	8905 (75.9%)	2822	24.06
Ash	666	570 (85.6%)	96	14.40
Power-plant slag	2641	2571 (97.3%)	70	2 > 7
Copper-smelt slag	80	1 (1.25%)	78	97.50
Nickel-smelt slag	3	1 (33.3%)	2	66.70
Blast-furnace slag	368	365 (99.2%)	3	0.8
Slag	1312	1211 (92.3%)	99	7.55
Ash-slag mixture	1319	1275 (96.7%)	44	3.3
Alumino-silicaeous cenospheres	158	142 (89.9%)	16	10.1
Power-plant gypsum	86	86 (100%)	0	0
Gypsum	207	207 (100%)	0	0
Aggregate	70	67 (95.7%)	3	4.3
Doped aggregate	6	6 (100%)	0	0
Copper-smelt slag aggregate	35	1 (2.9%)	34	97.1
Slag aggregate	14	14 (100%)	0	0
Ash aggregate	2219	2184 (98.4%)	35	1.6
Coal waste	40	40 (100%)	0	0

Table 5. Population exposure (calculated dose rate) from the use of wastes for road construction – average and maximum values

Type of raw or building material	Annual exposure time (h)			
	2100		750	
	Value of exposure (mSv/year)			
	Average	Maximum	Average	Maximum
Power-plant ash	0.203	0.985	0.072	0.352
Power-plant slag	0.159	0.650	0.056	0.232
Ash-slag mixture	0.166	0.463	0.059	0.165
Alumino-silicaeous cenospheres	0.199	0.456	0.070	0.163
Power-plant gypsum	0.012	0.096	0.004	0.034

Table 6. The excess number of the limit activity coefficient values f_1 > 2.4 or f_2 > 480 Bq/kg determined for the industrial wastes and buildings materials – these raws should not be used in surface objects in built-up areas or designed for built-up in the local urbanization plan in years 1980–2009

Raw or building material	Number of samples	Samples for which f_1 > 2.4 or f_2 > 480 Bq/kg	
		Quantity	(%)
Raw materials of industrial origin			
Power-plant ash	11 730	3	0.03
Copper-smelt slag	80	1	1.25
Slag	1312	2	0.15
Building materials			
Traditional concrete	105	1	0.95

of users, mean and maximum values of exposure dose were calculated for different industrial by-products. The values of exposure dose are presented in Table 5.

As can be seen from the above table, the mean calculated dose (that is exposure to the external gamma radiation) for road construction workers and professional drivers is in range between 0.012 and 0.203 mSv/a. A maximum value was calculated for power-plant ash and amounts to 0.985 mSv/a.

Mean values of exposure to gamma radiation for other users are smaller, and are within range between 0.004 and 0.072 mSv/a (the maximum calculated value being 0.352 mSv/a).

Conclusions

The by-products from combustion of coal are not radioactive wastes, but the products having the enhanced content of natural radioactive isotopes.

Maintaining the present measure of obligatory control of raw materials and building materials ensures the use of non-harmful (from radiological protection point of view) materials in dwellings.

Based on accessible results of measurements of natural radioactivity of building materials, in Table 6 there are presented numbers of particular secondary materials, which cannot be used in road construction that is whose activity indexes are f_1 > 2.4 or f_2 > 480 Bq/kg – according to the quoted Ordinance of the Council of Ministers – 2007).

As can be seen from the above table, practically all raw materials and building materials (also wastes) may be used in road construction. One should note that all values presented here concern 'pure' materials and raw materials. Though it is well known that, in practice, both housing and road construction, industrially-arising materials are mixed with other materials of much lower concentrations of natural radioactive isotopes, due to which it is not possible, that the resulting product activity indexes might be higher than those permitted in the quoted Ordinance of the Council of Ministers.

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