Thorium and Uranium in the Rock Raw Materials Used For the Production of Building Materials

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Abstract. Thorium and uranium are constant components of all soils and most minerals thereby rock raw materials. They belong to the particularly dangerous elements because of their natural radioactivity. Evaluation of the content of the radioactive elements in the rock raw materials seems to be necessary in the early stage of the raw material evaluation. The rock formations operated from deposits often are accumulated in landfills and slag heaps where the concentration of the radioactive elements can be many times higher than under natural conditions. In addition, this phenomenon may refer to buildings where rock raw materials are often the main components of the construction materials. The global control system of construction products draws particular attention to the elimination of used construction products containing excessive quantities of the natural radioactive elements.

In the presented study were determined the content of thorium and uranium in rock raw materials coming from the Bełchatów lignite deposit. The Bełchatów lignite deposit extracts mainly lignite and secondary numerous accompanying minerals with the raw material importance. In the course of the field works within the framework of the carried out work has been tested 92 samples of rocks of varied petrographic composition. There were carried out analyses of the content of the radioactive elements for 50 samples of limestone of the Jurassic age, 18 samples of kaolinite clays, and 24 samples of siliceous raw materials, represented by opoka-rocks, diatomites, gaizes and clastic rocks. The measurement of content of the natural radioactive elements thorium and uranium based on measuring the frequency counts of gamma quantum, recorded separately in measuring channels. At the same time performed measurements on volume patterns radioactive: thorium and uranium. The studies were carried out in Mazar spectrometer on the powdered material. Standardly performed ten measuring cycles, after which were calculated the concentration of radioactive elements in the sample.

The highest concentration of thorium and uranium has been found in the clayey raw material. Their value was respectively from 8 to 12 mg/kg for thorium and from 2.3 to 3.5 mg/kg for uranium. In carbonate sediments the content of thorium was at the level from 0.5 to 2.1 mg/kg and uranium from 0.5-2.2 mg/kg. From a group of the siliceous raw materials the diatomite had a highest concentrations of radioactive elements where the content of thorium was from 1.5 to 1.8 mg/kg and uranium from 1.3 to 1.7 mg/kg.

1. Introduction

Planning of construction investments beyond basic information about the geotechnical conditions of the ground requires assessment of the rheological conditions. This assessment should include the soil substrate on which the building is constructed as well as building materials both natural and processed origin. Radiation hazard results from the type of building materials and rocks occurring in the soil substrate as well as the tectonic or reservoir conditions of the area. It is assumed that radioactivity of geological origin is a natural source and therefore harmless. In the assessment of the radiation threat, the most important thing is the total radiation dose, regardless of whether it is from a natural or artificial source. The rocks enriched with heavy radionuclides uranium U-238 and thorium Th-232 and their...
derivatives, among others. Rad Ra-226 are the natural sources of the radioactivity. In recent years, more and more attention has been paid to the radon emanations passing to the dwelling rooms as a result of complex geochemical interactions between rock, soil, vegetation, water, atmosphere and structural elements [1 - 4]. Geologists can estimate a radon potential for geological area, defined as an average threat of high radon emissions to buildings located in this area by having information about: the content of radioactive elements in rocks and the impact of tectonics on the ability of migration and penetration of radon.

1.1 Geology of thorium and uranium

Uranium and thorium are a constant components of all soils and most minerals (figure 1). The average content of these elements in the earth's crust is 2 mg / kg for uranium and 12 mg / kg for thorium. A distinctive feature of the uranium ion is its ability to rapidly oxidize and convert to uranyl (U⁶⁺). Hexavalent uranium can easily combine with oxygen and interact in complex compounds with carbonates, sulphates and fluorides. In addition, it has the ability to precipitate in rocks containing reducing substances. They usually are decaying organic remains [5].

The varieties of the igneous rocks are particularly rich in these elements, especially in uranium. They are also rich in alkali, and usually also in SiO₂. The alkaline granites belong to the richest in uranium rocks within which the concentration of uranium and thorium can reach one hundred of grams per ton of rock. The sedimentary rocks with the highest concentrations of uranium and thorium are characterized by clayey rocks. Their values are respectively from 3 to 4 mg / kg for uranium and 9.6 to 12 mg / kg for thorium. The uranium compound with organic matter causes to its accumulation in modern peats. The carbonate and clastic rocks are definitely poor in uranium. Its values for clastic rocks is from 0.45 to 0.59 mg / kg and for carbonate rocks from 2.2 to 2.5 mg / kg. The concentration of thorium in the clastic rocks is slightly higher from 1.7 to 3.8 mg / kg and from 1.7 to 2.9 mg / kg for limestones [6]. The aforementioned radionuclides damage the chromosomes and they are a cause of lung cancer. In addition, uranium causes disorders in the functioning of the kidneys [7].

![Figure 1. The concentration of thorium and uranium in European soils [8]](image-url)
2. Scope and research methodology

In this paper, the content of thorium and uranium in the rock raw materials coming from the Mesozoic - Neogene contact zone in the Belchatów lignite deposit, located in Central Poland (figure 2), was determined. Over 40 years of an opencast exploitation in the Belchatów lignite deposit from which more than 50% of extraction of this raw material in Poland originates, has led to forming within the deposit area a surface excavation of a depth of about 300 m and slag heaps about height of up to 170 m.

![Figure 2. The study area. The surface excavation of lignite from the Belchatów deposit with slag heap.](image_url)

The studied material was taken directly from the Belchatów opencast mine and from drilling cores preceding the exploitation front. During field work, 98 rock samples of different petrographic composition were tested. The radiological elements were analysed for 50 samples of Jurassic limestones, 18 samples of the kaolinite clays and 24 samples of the siliceous raw materials represented by opoka-rocks, diatomites, gaizes and clastic rocks as well as 6 samples of marls.

In earlier works a detailed mineralogical and petrographic analysis of the examined rocks was presented and it was found that the studied rocks were represented by:

- carbonate rocks: limestones
- transitional rocks: opoka- rocks, gaizes and marls
- siliceous rocks: diatomites, flints
- medium grained deposits : wackes and arenites;
- weathered deposits: kaolinite clays, decalcified opoka-rocks and poly and monomictic breccias [9].

In the works [10,11] furthermore it was showed the content of heavy metals in the transitional rocks, kaolinite clays and the influence of silica mineral phases on the physico-mechanical parameters of the studied rocks with raw material importance. None of the mentioned works does not include the study of the content of radioactive elements in rocks coming from the Mesozoic - Cenozoic era contact zone. From cognitive and practical point of view, rheological evaluation of these sediments seems necessary because part of the petrographically named rocks meet the criteria to classify them as natural mineral sorbents and aggregates as well as in the case of the opoka rocks for construction material rocks.

The studies of the content of natural radioactive elements of thorium and uranium were carried out in a Mazar spectrometer on powdered material. Standardly performed ten measuring cycles, after which were calculated the concentration of radioactive elements in the sample. The content of the determined radioactive elements was on the measurement of frequency counts of gamma quantum, recorded separately in the measuring channels. Simultaneous measurements were made on volumetric radioactive model: uranium and thorium.
3. The results and discussion

In all petrographic varieties of the rocks, the presence of radioactive elements of thorium and uranium was found. The highest concentrations of thorium and uranium were found in the clayey raw materials. Their values were respectively from 8 to 12 mg/kg, on the average values of 10.1 mg/kg for thorium and from 2.3 to 3.5 mg/kg on the average values of 2.93 mg/kg for uranium (tables 1, 2 and figures 3, 4). In the carbonate sediments the content of thorium was from 0.5 to 2.1 mg/kg and uranium from 0.5 to 2.2 mg/kg.

In the clastic rocks the maximum values of thorium were up to 3.2 mg/kg at an average of 1.95 mg/kg and for uranium the maximum was 0.9 mg/kg at an average of 0.5 mg/kg. From the group of the siliceous raw materials, the diatomites showed the highest concentrations where thorium content was from 1.5 to 1.8 mg/kg and uranium ranging from 1.3 to 1.7 mg/kg. It is worth noting that there is considerable variation in the content of the examined elements in the opoka-rocks. In the case of uranium, these contents range from 0.2 to 2.93 mg/kg, and for thorium from 0.06 to 2.8 mg/kg. These rocks belong to the group of the transitional rocks between carbonate and silica rocks, so it is difficult to assess permitted values of radioactive elements for this type of rock material.

The main carriers of thorium and uranium in the studied rock raw materials are accessory minerals. In previous research, it was found in them the presence of minerals such as monacite or zirconium (figure 5). The concentration of uranium (U) in the monacite is 0.05 - 0.3 [wt.%] and thorium (Th) 2-20 [wt.%]. In zirconium the content of the studied elements is slightly lower and is in the range of 0.01-0.6 [wt.%] for uranium and 0.01-2 [wt.%] for thorium.

| Uranium content [mg/kg] | Type of rock raw material |
|-------------------------|--------------------------|
|                         | Limestone | Kaolinite clay | Opoka rock | Decalcified opoka rock | Diatomite | Gaize | Marl | Clastic rock |
| Min.                    | 0.5       | 2.3            | 0.2        | 0.75                   | 1.3       | 0     | 1    | 0.5 |
| Max.                    | 2.2       | 3.5            | 2.9        | 1                      | 1.7       | 1.3   | 1.9  | 0.9 |
| Aver.                   | 1.06      | 2.93           | 1.45       | 0.91                   | 1.42      | 0.7   | 1.2  | 0.51|

| Thorium content [mg/kg] | Type of rock raw material |
|-------------------------|---------------------------|
|                         | Limestone | Kaolinite clay | Opoka rock | Decalcified opoka rock | Diatomite | Gaize | Marl | Clastic rock |
| Min.                    | 0.5       | 8              | 0.06       | 0.3                    | 1.5       | 0     | 0.5  | 0.5 |
| Max.                    | 2.1       | 11.9           | 2.8        | 0.6                    | 1.8       | 1     | 2    | 3.2 |
| Aver.                   | 1.07      | 10.1           | 1.56       | 0.46                   | 1.5       | 0.5   | 1.58 | 1.95|
Figure 3. Uranium (U) content in the studied rock raw materials

Figure 4. Thorium (Th) content in the studied rock raw materials
4. Conclusions
The open cast mining of the lignite plays an important role in the economy of the rock raw materials. During the mining process of the main mineral there are real conditions of the sale of recovered minerals in different directions of industry. Previous studies and current situation show that some of the studied rocks from the Mesozoic - Neogene contact zone in the Belchatów lignite deposit can be used in pro-ecological technologies as mineral sorbents, in the ceramic industry as a component for the production of glazed cladding tiles, coloured floor tiles, porcelainite, and sanitary porcelain as well as various stoneware or construction materials. There are real reasons for the expanding of these possibilities. That is why there is a need for realization of basic and experimental research to expand the raw material alternative and deepening of the knowledge of new directions for the use of the studied rocks for the production of building materials. The carried out study of the content of the radioactive elements in the studied rocks complements the gap in terms of such research. It should also be borne in mind that the carried out studies have an importance in assessing of the impact of the extraction of mineral resources on the natural environment. The open cast nature of exploitation works promotes the accumulation of heavy metals in the rock material, stored in the slagheaps. Even a slight decrease in pH caused by climatic fluctuations leads to their demobilization, sometimes on a mass scale.

In all rock raw materials coming from the Mesozoic - Neogene contact zone in the Belchatów lignite deposit, it was found the presence of the radioactive elements of thorium and uranium. The clayey raw materials had the highest concentrations of the radioactive elements with an average value of 10.1 mg / kg for thorium(Th) and 2.93 mg / kg for uranium (U). These concentrations do not exceed the permitted values for clayey rocks. A similar situation is in the case of carbonate and clastic raw materials.

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