RESEARCH ARTICLE | MAY 31 2023

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AIP Conference Proceedings 2684, 040005 (2023)
https://doi.org/10.1063/5.0121540

Determination of Radioactivity of Natural Raw Materials for the Development of Radiation-Safe Construction Materials

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Abstract. An analysis of the current level of ecological safety of alarm materials showed that now in Ukraine, nutrition control and assessment of safety indicators are insufficient. In Ukraine, the main sources of natural radiation are concentrated within the Ukrainian Crystal Shield and to a lesser extent in the Donbass Shield and the Carpathian Shield. The results of the study of the radiation properties of natural raw materials of Ukraine are presented. The following quartz-feldspar rocks were studied in the work: pegmatite of the Lozuvatka deposit, as well as Anadol and Kremenovsky granites, which are located within the Priazovsky crystalline massif. Gamma-spectrometric analysis was used to measure natural radionuclides and determine the specific activities of natural radionuclides. It is determined that all experimental quartz-feldspar materials in terms of radiation safety belong to the 1st class, for which the condition Aef ≤ 370 Bq/kg is fulfilled. Since it is known that the activity of natural radionuclides in construction materials is 370 Bq kg-1 (equal to the average activity of the earth's crust), a radiation dose is about 1.0 mSv year-1. Living in houses with such a concentration of natural radionuclides in building structures creates a dose of human exposure that is approximately equal to the dose from the natural gamma background. Thus, the studied domestic raw materials can be used as raw materials in the manufacture of construction products without restrictions, as the number of natural radionuclides in them does not exceed the permissible norm. According to international requirements, the parameters of radiological danger to human health have been determined. According to international standards, the assessment of radiation hazard of construction materials was performed according to the criteria: equivalent radium activity (radiation hazard index) Raeq (Bq/kg), external hazard index Iex, gamma index Iγ, alpha index Iα. Recommendations for further use as a raw material in the development of non-combustible radiation-safe construction materials are provided, which guarantee protection of people from the harmful effects of radioactive elements and their sources in case of threat and emergencies.

PROBLEM STATEMENT

The greatest contribution to the value of the effective radiation dose (about 70%) is made by man-made sources of natural origin. Among these sources, ionizing irradiation of natural radionuclides in the construction materials of enclosing structures and in the soils under buildings are dominant. Since the impact of ionizing sources of construction production is exposed to almost the entire population, special attention is paid to the issue of radiation safety of construction materials and sites. Radiation control is an integral part of quality control of construction products in Ukraine. This ensures reliability, proper compliance with sanitary and hygienic standards in buildings and structures during their operation, as well as at the design stage.

Reliability and Durability of Railway Transport Engineering Structure and Buildings
AIP Conf. Proc. 2684, 040005-1–040005-7; https://doi.org/10.1063/5.0121540
Published by AIP Publishing. 978-0-7354-4501-7/$30.00

ANALYSIS OF RECENT RESEARCH AND PUBLICATION

The level of natural and man-made safety of Ukraine is largely due to excessive man-made loads on the environment [1]. Industrial areas are the ones with an extremely high risk of accidents and man-made disasters [2]. Man-made radioactive background increases from natural radionuclides and is formed due to the widespread use in construction of materials with high content of natural radionuclides [2, 3]. Another reason for creating the preconditions for accidents is the unsatisfactory technical condition of many buildings and structures [4]. Reliable assessment, forecasting of technical condition, rational use of fixed assets and the use of domestic natural raw materials can prevent structural accidents and associated losses [5, 6].

Today, more than 50% of all construction materials in the domestic market cannot be considered safe. Most construction materials are natural components of the ecosystem and therefore have their own specific radiation properties. For example, all construction materials of mineral composition contain different amounts of chemical elements, the isotopes of which are radioactive. The most dangerous ones in this regard may be construction materials made of natural stone and materials based on mineral binders [3, 7].

It is known that all quartz-feldspar rocks are characterized by a certain radioactivity due to the presence of minerals containing radioactive elements [8, 9]. White or pink feldspar contains 40K, black mica and sphalerite - 40K, U and Th, as well as small impurities of minerals such as zirconium, apatite and titanium. Since such natural raw materials contain these elements in greater quantities, considerable attention is paid to the study of its natural radioactivity [10]. In particular, using quartz feldspar as a promising material not only in the production of fine ceramics [11], but also in the production of non-combustible construction materials, it is necessary to determine the content of natural radionuclides, which will allow to establish their safety level at the design stage.

In the work [12] the results of researches on the possibility of using quartz-feldspar rocks of Ukraine in the manufacture of ceramic products are given. A theoretical assessment of the flux capacity of the rock is made and the necessity of using additives that reduce the viscosity of melts in pasty compositions is proved. No radiological evaluation of the experimental quartz-feldspar rocks was performed during the studies.

The authors in the work [13] used modern methods for the study of quartz-feldspar raw materials, such as X-ray scattering, Mössbauer spectroscopy, chemical analysis, UV-Vis-NIR spectrophotometry. Radiological analysis of natural raw materials using gamma spectrometry was not used.

The results of research of quartz-feldspar raw materials for the production of ceramic tiles using thermal, electron microscopic and X-ray methods are given by the authors in the work [14]. However, in this work there is no study of the radiological properties of the experimental raw materials.

The analysis of the studies presented by the authors in the work [15] showed that the concentrations of primary radionuclides (226Ra, 232Th and 40K) in the raw material for the production of binders slightly exceeded the typical average values. However, during the study of radiological properties of raw materials, the authors did not pay attention to determining the content of natural radionuclide 137Cs.

In works [16, 17], the authors studied the content of natural primary radionuclides (226Ra, 232Th and 40K) in raw materials used in construction materials. The average activity of the radium equivalent and the indices of external and internal danger for the experimental materials were also determined. But the content of the natural radionuclide 137Cs has not been studied.

During the review of literature sources [1–17] it has been revealed that there is no radiological assessment of natural raw materials that can be used as a construction component in full. This gives grounds to argue about the feasibility of more detailed radiation monitoring of natural quartz-feldspar raw materials that can be used in the construction industry.

PURPOSE AND OBJECTIVES OF RESEARCH

In order to develop radiation-safe construction materials, it is necessary to experimentally investigate the radiation properties of quartz-feldspar raw materials of Ukraine. Also, on the basis of experimental data to assess the radiological safety of experimental rocks and provide recommendations for their feasibility in the construction industry without any radiation threat to the population.

To achieve this purpose, the following problems should be solved:

- to determine the concentrations of primary radionuclides in natural raw materials;
- to determine the parameters of radiological danger to human health according to international requirements.

MATERIALS AND METHODS

According to the results of radiogeochemical studies [18], the territory of Ukraine is divided into three uranium provinces: Ukrainian Shield, Carpathian Shield and Donetsk Shield. In Ukraine, the main sources of natural radiation are concentrated within the Ukrainian Crystal Shield and to a lesser extent on the Azov Crystal Massif. In particular, using quartz-feldspar raw materials as the main component in the production of non-combustible construction materials, it is necessary to determine the content of natural radionuclides, which will allow at the design stage to establish their safety and determine their use in construction. The following quartz-feldspar rocks were studied in the work: pegmatite of the Lozuvatka deposit, as well as Anadol and Kremenovsky granites, which are located within the Priazovsky crystalline massif.

Studies of radiation properties are carried out by two methods: express (radiometric) or laboratory (spectrometric). The express method is intended for preliminary assessment of rocks and for periodic initial control over the production of construction materials and industrial waste. The radiation background of natural stone is determined at the stage of approval of the field reserves. At the same stage, mechanical characteristics such as abrasion, etc. are determined. As a result of definition in the passport of the deposit it is written down, to what group on radioactivity raw materials and recommendations on its application belong. To determine the level of radioactivity in the granite massif, wells are drilled into which the dosimeter is lowered. The received certificates are entered in the passport of the field.

The laboratory method is designed to establish and clarify the class of construction material or raw materials obtained by the express method, as well as for certification and sanitary-epidemiological examination. For the laboratory method, multi-channel gamma spectrometers with gamma radiation energy up to 3 MeV, the lower limit of measuring the specific activity of each PRN is not more than 50 Bq kg^{-1} and the relative measurement error is not more than 30% [19].

In this work, measurements of the effective specific activity of natural radionuclides of samples of quartz-feldspar materials were performed using gamma spectrometric analysis. Gamma spectrometric analysis was performed on a scintillation gamma spectrometer SEG-001 AKP-C with a range of energies of measured gamma radiation from 50 to 3000 keV. The gamma-ray energy spectrometer SEG-001 AKP-C is designed to determine the qualitative and quantitative composition of gamma-emitting radionuclides in food, the environment, agricultural products, construction materials, radioactive waste, etc.

RESEARCH RESULTS

1. The ecological safety of industrial products made of granite-containing raw materials is determined by the content of natural radionuclides in them, which is characterized by the value of the effective specific activity of A_{ef}. According to the Radiation Safety Standards of Ukraine (NRBU), the value of A_{ef} for quartz feldspar raw materials must be lower than or equal to 370 Bq • kg-1, and belong to the 1st class of radiation safety.

In this regard, the study of the radiation properties of the experimental quartz-feldspar raw material was carried out according to the method described above. Analysis of the obtained data shows that the presence of the following natural radionuclides was recorded in the composition of the materials: 232Th, 226Ra, 40K, 137Cs. Experimental and calculated data are presented in Table. 1.

Experimental and calculated data are given in Table1, it was found that the highest specific activity of ^{232}Th (A_{Th}) is characterized by granite of the Anadol deposit, while Lozuvatka pegmatite has a higher specific activity of ^{40}K (A_K) and ^{226}Ra (A_{Ra}). Flint granite is distinguished by the presence of the radionuclide ^{137}Cs (A_{Cs}) and the absence of ^{226}Ra (A_{Ra}).

The maximum rate of specific activity for Anadol granite and Lozuvatka pegmatite is almost the same. At the same time, the granites of the Anadol deposit have the highest effective specific activity, and the granites of the Kremenovske deposit have the lowest.

TABLE 1. The results of gamma spectrometric analysis of samples quartz feldspar raw materials.

Name of deposits	Sample code	Specific radioactivity, Bk kg^{-1} (contribution, %)				The sum of specific activities, Bk kg^{-1}
		^{232}Th	^{226}Ra	^{40}K	^{137}Cs	
Anadol granite	Sample1	81.3 (4.1)	15.2 (0.8)	1870 (95.1)	-	1967
Kremenovsky granite	Sample 2	4.67 (0.4)	-	1150 (99.5)	1.31 (0.1)	1156
Lozuvatka feldspar material	Sample 3	14.2 (0.72)	16,6 (0.85)	1930 (98.4)	-	1961

For comparative assessment of radioactivity of raw materials and raw materials the indicator of effective specific activity (A_{ef}) is used. The value of this indicator for the experimental quartz feldspar materials was calculated by the equation:

$$A_{ef} = A_{Ra-226} + 1.31A_{Th-232} + 0.085A_{K-40} + 0.31A_{Cs-137} + 0.874A_{Cs-134} \quad (1)$$

a)

where A_{Ra-226}, A_{Th-232}, A_{k-40}, A_{cs-137}, A_{cs-134}, (Bk·кg^{-1}) are specific activities ^{226}Ra, ^{232}Th, ^{40}K, ^{137}Cs, ^{134}Cs; 1.31, 0.085, 0.31 and 0.874 – coefficients for ^{232}Th, ^{40}K, ^{137}Cs, ^{134}Cs respectively in relation to ^{226}Ra.

Estimated data for experimental domestic raw materials are shown in Fig.1.

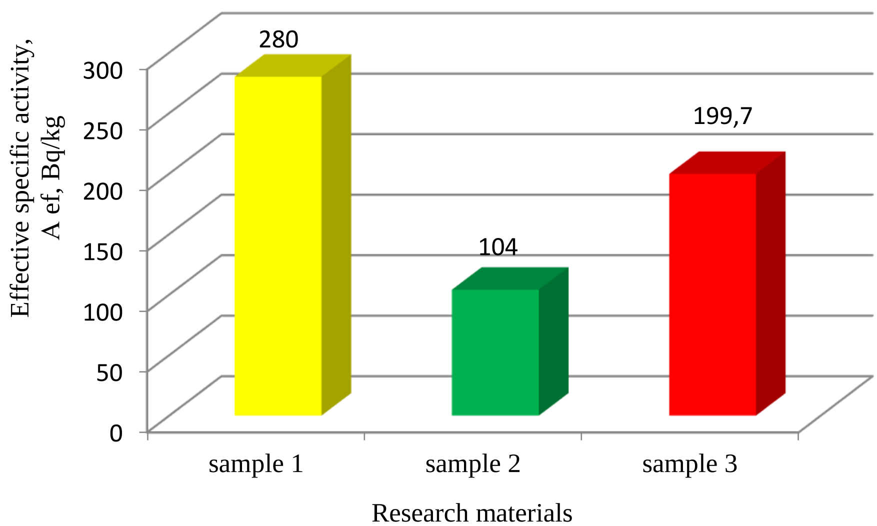

FIGURE 1. The results of studies of the effective specific activity of natural radionuclides in quartz feldspar raw materials.

It should be noted that all experimental quartz-feldspar materials for radiation safety belong to class 1, they meet the condition $A_{ef} \leq 370$ Bq/kg and can be used as raw material in the manufacture of construction products without restrictions, as the number of natural radionuclides in them does not exceed the permissible norm.

RESULTS AND DISCUSSION

According to international standards, the assessment of radiation hazard of construction materials is carried out according to the criteria: equivalent activity of radium (radiation hazard index) Raeq (Bq / kg), external hazard index Iex, gamma index Iγ and alpha index Iα [17].

The Radiation Hazard Index is used to compare the effective activities of construction materials containing different amounts of radium, thorium and potassium. Ra_{eq} is calculated by the equation:

$$Ra_{eq} = A_{Ra} + 1.43A_{Th} + 0.077A_K \tag{2}$$

Based on the assumption that 1 Bq/kg ^{226}Ra, 0.7 Bq/kg ^{232}Th or 13 Bq/kg ^{40}K give the same dose rate of γ-radiation as Ra_{eq}. The results of the calculation are shown in Fig. 2.

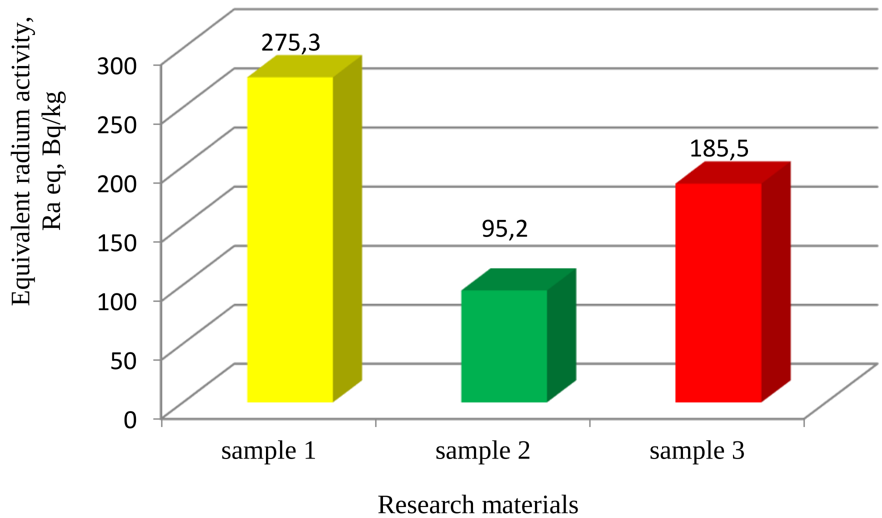

FIGURE 2. Study of the equivalent activity of radium in the experimental raw material.

According to the value of Ra_{eq}, all tested samples do not exceed the permissible conditions (370 Bq / kg). The external hazard index I_{ex} is calculated by the equation and must meet the condition and must be $I_{ex}<1$

$$I_{ex} = \frac{A_{Ra}}{370} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \tag{3}$$

This criterion is used to assess the level of γ-radiation hazard associated with the presence of natural radionuclides in specific construction materials. Another criterion that characterizes the γ-radiation of the construction material is the gamma index I_γ, calculated by the equation:

$$I_\gamma = \frac{A_{Ra}}{300} + \frac{A_{Th}}{200} + \frac{A_K}{3000} \tag{4}$$

The gamma index is used in screening to identify materials that could be of interest in construction and should correspond to $I_\gamma \leq 1$. Quantitative assessment of the exhalation of radon isotopes from construciton materials can be performed using the alpha index I_α, calculated by the equation:

$$I_\alpha = \frac{A_{Ra}}{200} \tag{5}$$

$I_\alpha \leq 1$ corresponds to an activity of ^{226}Ra and do not exceed 200 Bq / kg. The research results are shown in Fig. 3.

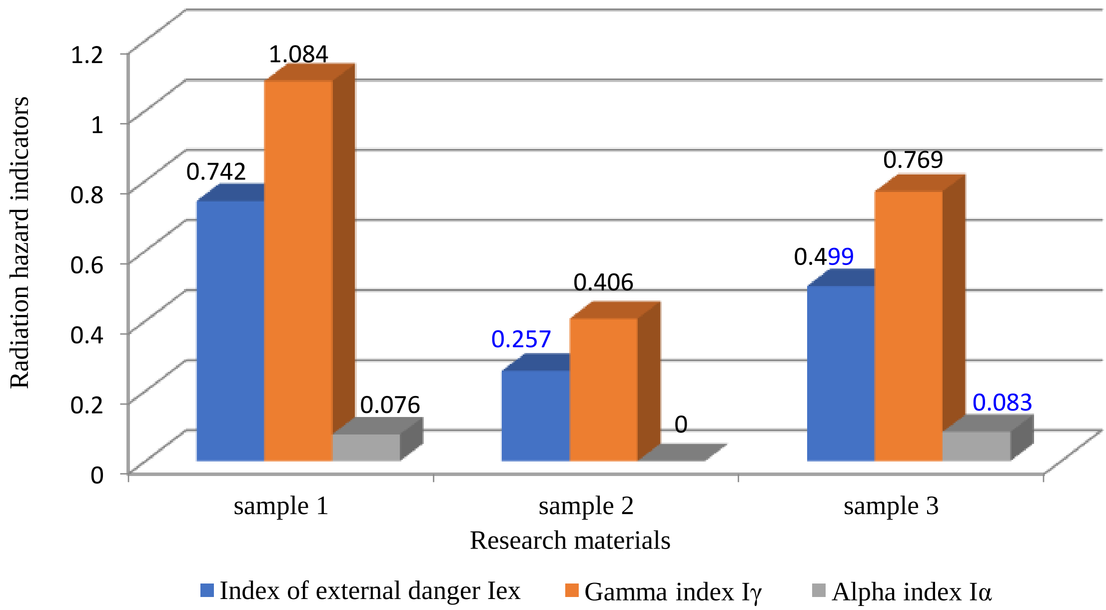

FIGURE 3. Parameters of radiological danger of quartz feldspar raw materials.

It should be noted that the calculated values of I_{ex} for most of the tested samples are in the allowable range and do not exceed 1. Regarding the gamma index I_γ, for materials used in large volumes, $I_\gamma \leq 1$ corresponds to an annual effective dose less than or equal to 1 mSv. $I_\gamma \leq 0.5$ corresponds to an annual effective dose of less than or equal to 0.5 mSv [15]. Only samples of the Kremenivka deposit belong to the second category. An exception to these categories is the sample of the Anadol deposit (I_γ = 1.084). Analysis of I_α values for the tested samples indicates no danger of radon inhalation from the construction material indoor.

CONCLUSIONS

The conducted research determined the concentration of primary radionuclides in natural raw materials. It is established that domestic quartz-feldspar raw material is characterized by the presence of the following natural radionuclides: 232Th, 226Ra, 40K, 137Cs. The number of natural radionuclides in them does not exceed the permissible norm ($A_{ef} \leq 370$ Bq · kg-1) and according to radiation safety belong to the 1st class. Analysis of the parameters of radiological danger to human health (Ra_{eq}, I_{ex}, I_γ, I_α) showed that the obtained values are within the limits recommended by the EU for construction materials. Therefore, the use of experimental quartz-feldspar materials of domestic deposits is recommended as a raw material in the manufacture of non-combustible radiation-safe materials without any significant radiological threat to the population.

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