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Radioactivity of buildings materials available in Slovakia

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Abstract. In the last decades building materials, both of natural origin and containing industrial by-products, have been shown to significantly contribute to the exposure of the population to natural radioactivity. As a matter of fact, neither the absorbed dose rate in air due to gamma radiation nor the radon activity concentration are negligible in closed environments. The soil and rocks of the earth contains substances which are naturally radioactive and provide natural radiation exposures. The most important radioactive elements which occur in the soil and in rocks are the long lived primordial isotopes of potassium (40K), uranium (238U) and thorium (232Th). Therefore, additional exposures have to be measured and compared with respect to the natural radiation exposure. Further, it is important to estimate the potential risk from radiation from the environment. The paper presents the results of mass activities of 226Ra, 232Th a 40K radionuclides in cement mortars with addition of silica fume. The gamma index was calculated as well.

1. Introduction
The most important radioactive elements which occur in the environment are the long lived primordial isotopes of potassium (40K), uranium (238U) and thorium (232Th). These radioisotopes are occurring in almost all the building materials but in different concentration. About 85% of dose received by man is due to natural radioactivity, of these 55.8% of dose comes from 238U series, 14% from 232Th series and 13.8% from 40K [1]. The radiation level due to natural radioactivity is about 2.4 mSv/y and the estimated worldwide average activity of 238U, 232Th and 40K in the earth’s crust to be 32 B/kg and 45 Bq/kg respectively [2]. Additional exposure have to be measured and compared with respect to the natural radiation environment. It is important to estimate the potential risk from radiation from environment. Man is continuously exposed to ionizing radiation from naturally occurring radioactive materials (NORM). The origin of these materials is the Earth’s crust, but they find their way into building materials, air, water, food, and the human body itself. Measurement of activity concentrations of radionuclides in building materials is important in the assessment of population exposures, as most individuals spend 80% of their time indoors [3]. Building materials cause direct radiation exposure, because most of them contain naturally occurring radioactive materials, mainly radionuclides from the 226Ra and 232Th decay chains and 40K. The population-weighted average of indoor absorbed dose rate in air from terrestrial sources of radioactivity is estimated to be 84 nGy h⁻¹ [4]. The worldwide average indoor effective dose due to gamma rays from building materials is estimated to be about 0.4 mSv per year [1]. Elevated indoor external dose rates may arise from high radionuclide content in building materials [5,6,7]. Generally, natural building materials reflect the geology of their site of
origin. The average activity concentrations of $^{226}$Ra, $^{232}$Th, and $^{40}$K in the Earth’s crust are 35, 30, and 400 Bq kg$^{-1}$, respectively. However, elevated levels of natural radionuclides causing annual doses of several mSv have been identified in some regions around the world, e.g. in Brazil, France, India, Nigeria, Iran [1,4]. This external radiation exposure, caused by gamma emitting radionuclides in building materials, can be assessed either by direct exposure measurements in the existing buildings or by radionuclide analyses of building materials with the dose rate modelling. Moreover, the trend of incorporation of various wastes into cement composites may result in increased radiological activity of concretes and cement-based materials depending on the radioactivity of waste utilized. The aim of the paper is to study the radiological parameters (mass activity of radionuclides and gamma index) of cement mortars prepared with various portions of silica fume. The activity of natural radionuclides in building materials have to be in accordance with the legislative requirements in the Slovak republic [8,9,10]

2. Material and methods

2.1. Cement samples

Research on radioactivity of building materials was performed using 4 mixtures of cement mortars with different portions of silica fume as follows: 0 wt.% (KU 0 – reference sample), 10 wt.% (KU 10 sample), 20 wt.% (KU 20 sample) and 30 wt.% (KU 30 sample) as cement replacement. Table 1 describes the recipe of mixtures in more detail.

| The components of mortar mix | Composition |
|------------------------------|-------------|
| [kg] Cement CEM 42.5 R | KU 0 | KU 10 | KU 20 | KU 30 |
| Cement replacement [wt.%] | 0% | 10% | 20% | 30% |
| Silica fume [kg] | 0 | 45 | 90 | 135 |
| Fine aggregate [kg] | 1350 | | | |
| Water [L] | 225 | | | |

Silica fume, also known as silica or microsilica is a by-product of manufacture of silicon or various silicon alloys by reducing quartz to silicon in an induction are furnace at temperature up to 2000°C. Gassified SiO at high temperatures condenses in the low-temperature zone to tiny spherical particles consisting of noncrystalline silica. The chemical composition of silica fume depends not only upon the raw materials used, but also upon the quality of the electrodes and the purity of the silicon product. Generally speaking, the impurities in condensed silica fume decrease as the amount of silicon increases in the final products. The by-products from silicon metal and ferrosilicon alloy industries, producing alloys with 75% or higher silicon content, contain up to 95% noncrystalline silica. Minor components in silica fume are aluminium oxide, iron (III) oxide, carbon, sulphur, calcium oxide, titanium (IV) oxide phosphorous(V) oxide and less than 1% alkalis [11]. The chemical composition of the silica fume, incorporated into the cement mortars in our experiments, is given in table 2.

| Oxide | SiO$_2$ | CaO | Al$_2$O$_3$ | Fe$_2$O$_3$ | MgO | K$_2$O | Na$_2$O | Cl | SO$_3$ | C | LOI |
|-------|---------|-----|------------|------------|-----|--------|---------|----|--------|---|-----|
| [%]   | 71.65   | 18.17 | 0.70       | 1.19       | 1.95 | 1.14   | 0.52    | 0.14| 2.39   | 1.21 |      |

It is well known that use of silica fume in concrete decreases or eliminates the free lime content, increases the strength and decreases or eliminates concrete. Silica fume showed an effect on the released lime 8 hour after the hydration of Portland cement, which is significantly faster than slag or fly ashes due to its small particle size. Laboratory results have indicated that the use of silica fume decreases the diffusion coefficient of contaminants very significantly [12].
2.2. Testing methods
The mass activities of radionuclides in cement mortars were measured using gamma ray spectrometry. Gamma ray spectrometry has been used for many years to measure the specific radionuclides activity in a given sample. This technique is an important tool in field of environmental radioactivity measurements due to its high resolution, large photo peak efficiency and it can measure different radionuclides in a single spectrum. Measurement of radioactivity was carried out using an EMS-1A SH (Empos, Prague, Czech Republic) detection system equipped with a NaI/Tl scintillation detection probe and a MC4K multichannel analyser with optimized resolution of 818 V, 4,096 channel and with 9 cm of lead shielding and internal lining of 2 mm tinned copper.

The background of the detection system plays a vital role in the measurement of low-level activity as typically found in construction materials. The counting system must have a background as low as attainable with a minimum number of spectral lines originating from natural radionuclides which may be present in the system components and in the surrounding environment of the counting facility. In the presented study, routine measurements of the background count rates for natural radionuclides were carried out before each set of measurements, each for a counting time of 43,200 s (i.e., 12 h). The Cement specimens were crushed and milled to prepare the powder samples and consequently filled into Marinelli containers and weighed. The filled containers were sealed and kept for 30 days to achieve the radioactive equilibrium. The emphasis was on the determination of specific activity concentration of $^{226}$Ra, $^{232}$Th and $^{40}$K. The radioactivity of $^{40}$K was measured directly through its gamma ray energy peak at 1461 keV, while activities of $^{226}$Ra and $^{232}$Th were calculated based on the mean value of their respective decay products. Activity of $^{226}$Ra was measured using the 351.9 keV gamma rays from $^{214}$Pb and the activity of $^{232}$Th was measured using the 238.6 keV gamma rays of $^{212}$Pb. Every sample was measured for 86,400 s.

Radiation hazard index, called the gamma activity concentration index, $I_\gamma$, has been defined by the European Commission [13] and equation (1) is given below:

$$I_\gamma = \frac{A_R}{300} + \frac{A_{Th}}{200} + \frac{A_K}{3000}$$

(1)

The index $I_\gamma$ is correlated with the annual dose rate due to the excess external gamma radiation caused by superficial material. The table 3 shows a dose rate criterion [14].

| $I_\gamma$ | Dose rate criterion |
|-----------|---------------------|
| [-1]      | [mSv·y$^{-1}$]      |
| $I_\gamma \leq 2$ | 0.3  |
| 2$<I_\gamma \leq 6$ | 1   |
| $I_\gamma > 6$ | >1   |

3. Results and discussion
The table 4 presents the results of measured mass activities of radionuclides $^{226}$Ra, $^{232}$Th and $^{40}$K. Comparing the radium activities measured, the limit value of 120 Bq·kg$^{-1}$ was not exceed for any composite even silica fume itself. As presented in our previous work [15], the radionuclide activities in CEM I cements ranged from 3.69 – 36.8 Bq·kg$^{-1}$, 11.8 – 24.9 Bq·kg$^{-1}$ and 36.98 – 331.4 Bq·kg$^{-1}$ for $^{226}$Ra, $^{232}$Th and $^{40}$K, respectively. According to Terpakova (2000) [16], the aggregates radioactivity strongly depends on the geological sources and ranges from 21.5 to 29.9 Bq·kg$^{-1}$for $^{226}$Ra, 5.0 to 11.4 Bq·kg$^{-1}$ for $^{232}$Th and 106.5 to 270.6 Bq·kg$^{-1}$ for $^{40}$K. Cabanekova (1996) [17] presents a very wide range of radionuclides activities regarding concretes. As seen in Table 4, the silica fume’s radioactivity is comparable to the values presented for cement and aggregates as majority components of cement mortars. The highest values were measured for reference sample KU 0 without any silica fume. The hypothesis that the silica fume does not increase the radioactivity levels of
concretes was confirmed. On the other hand, the hypothesis that silica fume can play a role of a dilution agent regarding the radioactivity levels in concretes was not quite confirmed when compared the mass activities of silica fume-based samples.

Table 4. Mass activities of radionuclides in studied cement mortars.

| Sample   | Mass activity [Bq·kg⁻¹] |
|----------|-------------------------|
|          | \(^{226}\text{Ra}\) | \(^{232}\text{Th}\) | \(^{40}\text{K}\) |
| KU 0     | 5.72                   | 20.74               | 80.45             |
| KU 10    | 2.67                   | 11.50               | 5.02              |
| KU 20    | 2.18                   | 12.74               | 18.17             |
| KU 30    | 3.47                   | 9.06                | 2.06              |
| Silica fume | 3.91                 | 6.11               | 297.13            |

The mean values of index of natural radionuclide mass activity (gamma index, \(I_\gamma\)) calculated from the measured activity concentration of \(^{226}\text{Ra}, {^{232}\text{Th}}\) and \(^{40}\text{K}\) are presented in figure 1.

![Gamma index of cement samples (\(I_\gamma\)).](image)

Gamma index ranged from \(I = 0.149\) to \(I = 0.057\) for the sample with addition of silica fume. Silica fume had gamma index with value \(I = 0.14\). The lowest value of was calculated for KU 30 cement sample with 30 wt.% silica fume. The highest value was found for sample KU 0 without any silica fume addition. The gamma index should also take into account typical ways and amounts in which the material is used in a building. The limit values depend on the dose criteria, the way and amount of the material and the manner in which it was used in a building and construction. The limit value of \(I = 1.0\) for building materials was not exceeded in any cement mortar sample.

4. Conclusion

The natural level of radioactivity in building materials is one of the major causes of external exposure to γ-rays. By the determination of the radioactivity level in building materials, the indoor radiological hazard to human health can be assessed. Knowledge of basic radiological parameters and radioactive contents in the construction materials is important since it allows us to calculate the exposure of the population of radiation from natural sources. It plays an important role in the protection measurement, geoscientific research and guidelines for the use and management of these materials. During the last decades, there has been an increasing interest in the study of radioactivity in various building materials.

In this study, the commonly used composites based on silica fume addition as partial replacement of cement - in the amount of 0% (reference), 10%, 20% and 30% were investigated to assess the radiological hazards. The values obtained in the study are within the recommended safety limits, showing that these building materials do not pose any significant radiation hazard and hence the use of these materials in the construction for dwelling purpose can be considered to be safe for the
inhabitants. This study can be used as a reference for more extensive studies of the same subject in future.

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