CHARACTERISTICS OF NATURAL RADIOACTIVITY AT THE REICHE ZECHE MINE, GERMANY*

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Determination of the natural radiation background in underground localizations is necessary to describe them for possible use not only for physics experiments. The characteristics of natural radioactivity at the Reiche Zeche mine is presented and contains results from *in-situ* measurements, radon concentration in air, and α/β laboratory analyses of water and rock samples from the investigated localization. The measurements were performed in the Research and Education Mine "Reiche Zeche", Germany at the depth of 150 m (410 m w.e.).

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1. Introduction

Underground Laboratories (ULs) are nowadays widely used for experiments in physics, biology, environmental studies, and others. These localizations provide a necessary low radioactive background. The natural background in an underground laboratory depends on the depth and on the nature of the surrounding rocks [1, 2]. The cosmic rays muons flux decreases exponentially with increasing depth, while the muon spallation processes produce a depth-dependent flux of high-energy neutrons. Low-energy neutron flux originates mainly due to fission and alpha emission from U and Th contained in the rock. The gamma flux, including radon and its progeny, also depends on local geology and is practically depth-independent. The study presented in this paper is devoted to the characteristics of natural radioactivity in the Reiche Zeche mine. The Reiche Zeche is one out of six underground BSUIN (Baltic Sea Underground Innovation Network) laboratories [3]. The main purpose of the BSUIN project is to create a service offering for the

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Baltic Sea Region underground laboratories for business development and expand their capability to offer a technology transfer. The 6 Underground Laboratories from the Baltic Sea Region, the 14 Partners Institutions and 15 Association Organizations take part in the BSUIN project. Detailed information about the project, as well as the description of the participating underground laboratories, can be found on the project website [3].

In this paper, the results of following measurements performed in the Reiche Zeche mine: concentration of radioactive isotopes in the rock using *in-situ* and laboratory spectrometers, radon concentration in air and the content of radionuclides in water samples are presented.

2. Natural radioactivity measurements

The *in-situ* measurements were performed in the server room (dimensions $3\,\mathrm{m} \times 3\,\mathrm{m} \times 2.20\,\mathrm{m}$) at the Education and Research Mine "Reiche Zeche" of the Technical University of Freiberg, Germany at a depth of 150 m (410 m w.e.) located in the orthogneiss rock. In addition, the water samples from the mine water gullets and the rock samples from a newly blasted excavation were taken for laboratory analysis in the August Chełkowski Institute of Physics, University of Silesia, Katowice, Poland. The *in-situ* measurements were performed by using portable HPGe semiconductor spectrometer and RAD7 electronic radon detector. At the laboratory, analysis of the concentration of radioisotopes in water samples were performed using a liquid scintillation α/β LSC counter and alpha spectrometry technique. Moreover, rock samples collected in mine were analyzed for natural radioactivity by the use of alpha- and gamma-spectrometry techniques.

2.1. In-situ measurements

The *in-situ* measurements of natural background radiation in the Reiche Zeche mine (server room) were performed in December 2018. The GR4020 portable spectrometer (Canberra Industries, Inc., USA), a high purity n-type germanium coaxial detector of 40% relative efficiency with a resolution of 2.1 keV FWHM at the 1.33 MeV ^{60}Co line was used. Data were collected using the InSpector$^{\mathrm{TM}}$ 2000 multichannel analyser Canberra Industries, Inc., USA, while the gamma-ray spectra were analysed using the Genie$^{\mathrm{TM}}$ 2000 v.3.2.1 software package from Canberra Industries, Inc., USA. The following calibrations were performed: the energy calibration using 7 radioactive sealed sources (^{133}Ba, ^{137}Cs, ^{54}Mn, ^{57}Co, ^{109}Cd, ^{22}Na, ^{60}Co), and the efficiency calibration using the In Situ Object Counting System (ISOCS$^{\mathrm{TM}}$) — a mathematical calibration software (Canberra Industries, Inc., USA). During the measurement, the HPGe detector was placed in the horizontal position 1 m in front of the nearest wall (the center of the germanium crystal of the

detector was located 10 cm above floor). The measured integrated counts per second (cps) in the energy range of 7–3150 keV was 371.52 ± 0.05, the estimated gamma-ray flux density was 2.8 ± 0.8 [$cm^{-2}s^{-1}$] and the effective dose was equal to 0.036 ± 0.008 μSv/h. The radioisotopes that have main contributions in calculated effective dose are: ^{40}K (39%) and ^{214}Bi (27%) [4]. For comparison, in the three leading European Underground Laboratories: Gran Sasso (Italy), Modane (France) and Boulby (England), the integrated cps in energy range of 7–2734 keV are 49 ± 9 [5], 79 ± 23 [6], 24 ± 4 [7], respectively.

2.2. Measurements of radon concentration in air

The measurement of radon concentration in air was carried out in the server room (Reiche Zeche mine) by using RAD7 electronic radon detector (Durridge Company, Inc.). Radon concentration was obtained from 24 measurements, 1 h long each. The RAD7 detector was located near the gamma-ray spectrometer. The average radon concentration was equal to 805.1 ± 10.4 Bq/m^3, with the median value of 802.9 Bq/m^3 [4]. The obtained result of radon concentration in air is significantly higher than in leading European ULs: in Gran Sasso was in the range of 50–120 Bq/m^3, in Modane 5–15 Bq/m^3, whereas in Boulby was < 3 Bq/m^3 with ventilation on [1].

2.3. Alpha-spectroscopy measurements of uranium concentration in water and rock samples

The measurements of ^{234,238}U isotopes concentration in water samples were performed with the use of α-spectrometry technique (7401VR from Canberra — Packard). After collection, the samples were acidified with HNO_3 acid in order to avoid radionuclide precipitation as well as adsorption on the walls of the containers. Prior the chemical analysis, the standard ^{232}U of known activity was added to each water sample. The separation of uranium was performed with the use of the anion exchange resin Dowex 1×8 (Cl-type, 200–400 mesh) on the basis of the procedure worked out by Suomela [8]. A thin α source was prepared from uranium fraction by coprecipitation with NdF_3 and filtration [9]. Minimum Detectable Activity (MDA) was equal to 0.5 mBq/l for both ^{234,238}U isotopes and 0.5 l initial sample volume. Additionally, for rocks samples, the wet-mineralization of samples was performed with the use of hot acids: HF, HNO_3, HCl with H_3BO_3. Uranium was pre-concentrated with iron and co-precipitated at pH 9. Subsequently, the samples were separated from other radionuclides in the same way as described for water samples. The obtained concentrations of uranium in water and rock samples are presented in Table I. The ^{234}U/^{238}U

activity ratio for both samples is approximately equal to 1, which means the radioactive equilibrium was achieved in rock and water samples. Usually, the concentration of uranium in rock is about 20–30 Bq/kg [10]. The measured concentration of ^{238}U in rock in the Reiche Zeche mine is equal to 32.4 ± 2.3 Bq/kg and is significantly higher than concentrations in major European ULs: 1.8 ± 0.1 Bq/kg in Gran Sasso [5] and 11.8 ± 0.6 Bq/kg in Modane [6].

2.4. LSC technique measurements in water samples

The measurements of ^{226,228}Ra activity concentrations in water samples were performed with the use of LSC technique (WinSpectral α/β 1414 liquid scintillation counter from Wallac). The chemical procedure based on the Polish Norm PN-89 Z-70072 [11] was applied. The procedure involves preconcentration of radium by coprecipitation of Ra with $BaSO_4$ and separation of radium from other radionuclides. The samples were measured once per day with 1 hour counting time over a period of 25 days, until a secular equilibrium between ^{226}Ra and its daughters was reached. The results of radium concentration in water samples are presented in Table I. The activity concentration of radium isotopes were below MDA, equal to 0.015 Bq/l and 0.04 Bq/l for ^{226}Ra and ^{228}Ra, respectively, for 3600 s counting time and 1.5 l of water initial sample volume.

2.5. Measurements of radioisotopes in rock samples

Prior the measurements of the natural radioisotopes concentration (^{40}K, ^{226}Ra, ^{228}Ra(^{232}Th)), rock samples were dried, crushed, ground and stored in Marinelli container. In this form, the samples were left for the period of one month in order to achieve the secular equilibrium in thorium and uranium series. The measurements were performed using gamma-spectrometry method. The spectrometer was equipped with a lead-shielded HPGe detector (60.7 mm crystal diameter and a Cryo-Pulse 5 Plus, an electrically-powered cryostat (Canberra Industries, Inc., USA)) with a relative efficiency of 20%, an energy resolution FWHM of 1.8 keV, and a P/C ratio of 50 : 1 at 1.33 MeV ^{60}Co gamma line. The radioactivity concentrations were calculated based on a standard prepared from certificated materials obtained from the Central Laboratory for Radiological Protection in Poland. The activity of ^{226}Ra was calculated as the weighted mean of the values obtained from the ^{214}Pb (295.2, 351.9 keV) and ^{214}Bi (609.3, 1120.3 keV) isotopes, whereas the activity of ^{228}Ra was calculated from the gamma lines 338.3 keV and 911.1 keV originating from ^{228}Ac, while the activity of ^{40}K from the single 1460 keV line. The results of radium, thorium and potassium concentration in rock samples are presented in Table I. The obtained results of concentration of thorium and potassium in rock samples are higher than results

from Gran Sasso and Modane. For ^{232}Th, the concentration is equal to 31.5 ± 0.6 Bq/kg in Riche Zeche and for ^{40}K, is equal to 1049 ± 17 Bq/kg, while these concentrations are 1.5 ± 0.1 Bq/kg and 26 ± 2 Bq/kg in Gran Sasso [5] and 10.2 ± 0.5 Bq/kg and 182 ± 4 Bq/kg in Modane [6], respectively.

TABLE I

Results of the U, Ra, Th and K radioisotopes concentration in water and rock samples. The used method: *α-spectrometry; **LSC and ***gamma-spectrometry.

Sample	Uranium*	Radium	Potassium
Water sample [mBq/l]	^{238}U 150.4 ± 5.2 ^{234}U 142.4 ± 4.9 (^{234}U/^{238}U 0.95 ± 0.05; U 12.2 ± 0.4 μg/l)	^{226}Ra $< 15^{**}$ ^{228}Ra $< 40^{**}$	not determined
Rock sample [Bq/kg]	^{238}U 32.4 ± 2.3 ^{234}U 34.4 ± 2.4 (^{234}U/^{238}U 1.06 ± 0.11; U 2.6 ± 0.1 ppm)	^{226}Ra $43.8 \pm 0.4^{***}$ ^{228}Ra(^{232}Th) $31.5 \pm 0.6^{***}$	^{40}K $1049 \pm 17^{***}$

3. Summary

The results of the characterization of the natural background radiation measured in the server room, the Reiche Zeche mine, Germany have been presented including both *in-situ* measurements and laboratory analyses performed with the use of different techniques. The obtained results of concentration of radioisotopes in rock samples and radon concentration in air in the Reiche Zeche mine are higher than those measured in other European ULs (Gran Sasso, Modane and Boulby). The concentrations of radium isotopes ($^{226,228}Ra$) in water samples were below MDA. The gamma-ray flux density is 2.8 ± 0.8 [cm^{-2}s^{-1}] and the effective dose is 0.036 ± 0.008 μSv/h. In the studied localization, the neutron flux by using two proportional ^{3}He counters was also measured, resulting in value of $3.12 \pm 0.10 \times 10^{-6}$ [cm^{-2}s^{-1}] [4]. The detailed description of measurements performed in the Reiche Zeche mine can be found elsewhere [4].

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