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RADIOACTIVITY OF DRILLING CUTTINGS FROM SHALE RESOURCES OF THE LOWER PALEOZOIC BALTIC BASIN

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Abstract: Fractionated drilling wastes originating from shale gas exploration in the Baltic Basin in Pomerania were subjected to measurements of mean activity concentrations of naturally occurring radioactive materials (NORMs). X-ray diffraction (XRD) and scanning electron microscopy (SEM) analyses were used to understand the rock structure and texture. The activity concentration of radionuclides in bulk wastes and fractionated samples (latter obtained through a sieve analysis) was analyzed by using a gamma-ray spectrometer. After fractionation, three different size ranges were distinguished. Radiological indices were estimated by comparison with the levels recommended by International Association of Oil and Gas Producers (OGP) and an equivalent absorbed dose was determined with respect to appropriated disposal of cuttings. The results showed that the drilling cuttings from the Ordovician period have a natural radioactivity level comparable to other rocks with the same lithology (potassium K-40 800-992 Bq/kg and thorium Th-232 23.3-30.8 Bq/kg) and they did not exceed acceptable levels of the total absorbed dose rate. The average absorbed dose rate in outdoor air 1 m above the drilling cuttings was 54.1 nGy/h. The relation between particle size and natural radioactivity indicated that the concentration of radionuclides increased as the fractions size decreased.

Keywords: *radioactivity, radionuclides, sieve analysis, drilling cuttings, shale gas*

Introduction

Naturally occurring radioactive materials (NORM) are mostly nuclides with the half-life of hundreds millions of years and come from the Earth formation period. They are referred to as primordial (Ravisankar et al., 2014). Natural radioactivity is the major source of radiation to which humans are exposed and it represents more than 75% of all ionizing radiation (UNSCEAR, 2000). Gamma radiation is always associated with

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different lithological structures and can also be the factor used to distinct one type of rock from another. Shaliness, used in formation evaluation, is a good example of this phenomenon. Elevated gamma radiation emission characterizes shales and igneous rocks (e.g. granites) as well as lower sedimentary rocks (Mykowska and Hupka, 2014). The gamma ray logging data are valuable during well logging measurement, after the borehole has been drilled and also in the stage of drilling for real-time geosteering decisions. Another aspect of NORM accumulated in rocks, regards environmental issues. The radiological hazard from drilling wastes is especially important in cuttings management.

The drilling cuttings examined in the research originated from a shale gas exploration borehole in the Baltic Basin, which is one of the three prospective regions of unconventional gas resources (Baltic, Podlasie and Lublin Basins) located in the area of the East European Platform. Silurian and Ordovician deposits in sedimentary basins of Poland often contain a relatively large proportion of organic matter, so they could be considered as unconventional hydrocarbons reservoirs (Poprawa, 2010). They contain organic matter, which is correlated with the level of natural radioactivity of rocks. In this area, the gas-bearing shales can be found most frequently in the sediments of monotonous silty-sandstone. The advantage of Paleozoic sedimentary basins, such as the Baltic Basin, is their simple structure, conducive to exploration and exploitation of unconventional resources of oil and gas (Poprawa, 2010; Kiersnowski, 2014).

The natural radioactivity of oil and gas exploration wastes was investigated in some papers (Hamlat et al., 2001; El Afifi and Awwad, 2005; Atallah et al., 2012; Hrichi et al., 2013; Hilal et al., 2014). Recently, the research was focused on the impact of particle size on gamma radiation emission. The objective of this paper is to compare activity concentrations of radionuclides, in both bulk wastes and fractioned cuttings, before and after sieve analysis, to verify the relationship between particle size and emission of natural gamma radiation. Another aspect of the research is to estimate the radiological hazard based on the absorbed dose rate.

Origin of radioactivity in oil and gas exploration and production wastes

Radioactive isotopes occur naturally in the environment but can accumulate due to industrial activities. The natural radioactivity, greater than the background radiation, has been noticed in the oil and gas recovery waste in 1930s (Otto, 1989). At the beginning of 1980's the regulatory agencies started to pay attention to the NORM associated with hydrocarbons production (Smith, 1992). Gamma radiation from the waste has a very wide range. Radium-226, which is the primary isotope of concern, has activity concentration from undetectable to 1000 kBq/kg (Smith, 1992; Hilal et al., 2014). The primordial NORMs and decay chain radioisotopes in drilling wastes are presented in Table 1 (Atallah et al., 2012). Most of discussed radionuclides emit alpha radiation (U-238, Th-232, Ra-226 and Rn-222), potassium K-40 emits electrons.

Gamma radiation accompanies both types of decay and the energy of gamma rays is between 12 keV for thorium Th-232 to 1.46 MeV for potassium K-40.

Table 1. Selected natural radionuclides associated with oil and gas production (Atallah et al., 2012)

Nuclide	Type	Half-life	Decay energy
U-238	primordial	4.47×10^{9} y	α (4.2 Mev), γ (13 keV)
Th-232	primordial	1.4×10^{10} y	α (4.01 Mev), γ (12 keV)
K-40	primordial	1.28×10^{9} y	β-(1.31 Mev), γ (1.46 MeV)
Ra-226	U-238 decay series	1600 y	α (4.78 Mev), γ (186 keV)
Rn-222	U-238 decay series	3.8 days	α (5.49 Mev), γ (512 keV)

The radioactivity of oil and gas waste depends on the lithology of drilled reservoir, which determines the amount of radionuclides in the subsurface and chemical composition of formation fluid. The duration of production, extraction and treatment process (also hydraulic fracturing), changes pH, pressure, temperature and causes mobilization of the NORMs (Smith, 1992; Atallah et al., 2014). The radionuclides in the drilling wastes mostly come from different types of rocks. Selected, exemplary activity concentrations of radionuclides are presented in Table 2. It shows the range of natural radioactivity levels for the same lithological types of rocks.

Potassium K-40 has higher activity concentration than uranium U-238 and thorium Th-232. Gamma-ray spectrometry during well logging shows K-40 concentration in percentiles, while U-238 and Th-232 concentration in parts per million. The concentrations of the mentioned radionuclides provide information about producible zones and reservoir characteristics. They also allow to estimate the volume and type of clays.

The solid oil and gas wastes in the form of scale and sludge, tend to accumulate in the downhole casing and tubing as well as at the surface equipment (separators, pumps, vessels, pipelines, tanks). It could present some radiological hazard (Smith, 1992; Hamlat et al., 2001; Atallah et al.,2012; Hrichi et al., 2013; Hilal et al., 2014;). Radium Ra-226 and Ra-228 can co-precipitate in the scale made of BaSO4, SrSO4 or CaCO3 (Hamlat et al., 2001). The NORM can appear also in the liquid (produced water, formation water) and gas phase. The ways of radionuclides mobilization are shown in Fig. 1. Drilling cuttings and produced water are the major source of natural radioactivity in oil and gas production, also offshore (Bakke et al., 2013).

Table 2. A comparison of activity concentrations with standard deviation [Bq/kg] of radionuclides in rocks

Lithology	Activity concentration [Bq/kg]				Rn-222 [kBq/m^3] (*)	Country	Reference
	U-238	Th-232	K-40	Ra-226			
Sandstone	34±3	53.0±4	23236±32	-	-	Nigeria	Omeje et al., 2013
Clay	23±2	32.0±3	399±51	-	-	Nigeria	Omeje et al., 2013
Gneiss	-	38.0±4	1168±42	28.4±3	-	Egypt	Harb et al., 2012
Granite	-	90.5±7	2208±91	118.0±7	-	Egypt	Harb et al., 2012
Sandstone	-	12.5±3	264±11	7.5±1.5	-	Egypt	Harb et al., 2012
Limestone	-	-	53±2	35.7±2.4	-	Poland	Malczewski et al., 2006
Limestone	-	-	473±9	43.2±6.8	-	Poland	Malczewski et al., 2006
Granite-gneiss	-	-	1177±13	49.8±6.9	-	Poland	Malczewski et al., 2004
Hornfelses	-	-	494±6	30.7±3.7	-	Poland	Malczewski et al., 2004
Granite-gneiss	-	-	-	50.4±31.4	-	Poland	Przylibski, 2004
Sandstone	-	-	-	12.8±10.7	-	Poland	Przylibski, 2004
Hornfelses	-	-	-	35.7±1.2	-	Poland	Przylibski, 2004
Shell limestone	-	-	-	-	33±4	Germany	Kemski et al., 2001
Coloured standstone	-	-	-	-	24±3	Germany	Kemski et al., 2001
Carboniferous sediments	-	-	-	-	41±3	Germany	Kemski et al., 2001
Granites		-			106±8	Germany	Kemski et al., 2001
Gneiss	-	-	-	1.0-1800	-	Mean global value	IAEA, 2010
Limestone	-	-	-	0.4-340	-	Mean global value	IAEA, 2010
Clay/shale	-	-	-	1.0-990	-	Mean global value	IAEA, 2010
Sandstone	34±3	53.0±4	23236±32	-	-	Nigeria	Omeje et al., 2013
Clay	23±2	32.0±3	399±51	-	-	Nigeria	Omeje et al., 2013
Gneiss	-	38.0±4	1168±42	28.4±3	-	Egypt	Harb et al., 2012

(*) Radon activity concentration in soil gas above rocks

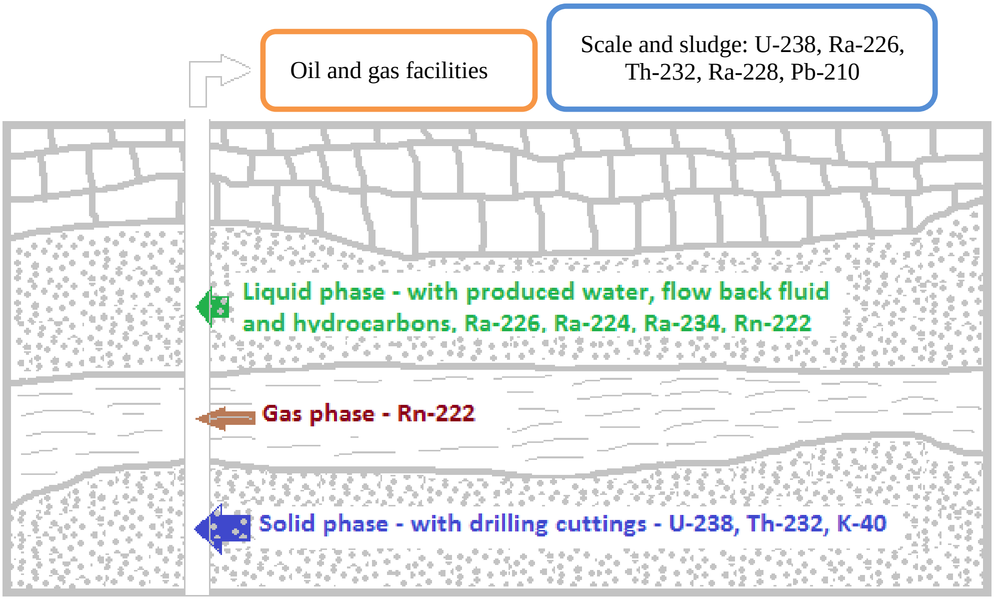


Fig. 1. NORM mobilization in oil and gas production (based on OGP, 2008)

The exemplary NORM exemptions levels taken from international regulations are provided in Table 3. The adhering to recommendations laid out in these recommendations, minimizes the risk of worker exposure to ionizing radiation.

Table 3. NORM exemption levels (OGP, 2008)

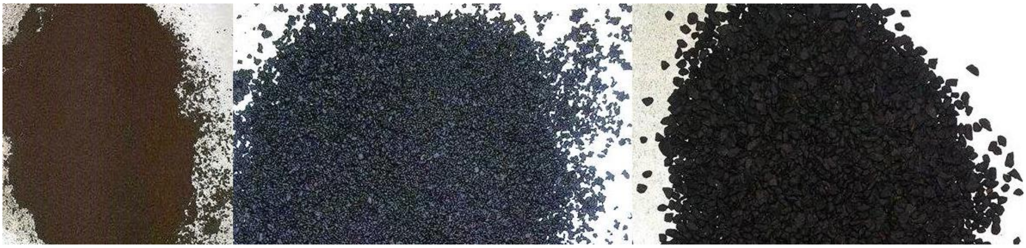
Nuclide	Type	Exemption level [Bq/kg]
U-238	primordial	5500
Pb-210	U-238 decay series	200
Po-210	U-238 decay series	200
Ra-226	U-238 decay series	1100
Ra-228	Th-232 decay series	1100

Experimental procedure

Sample preparation

The Ordovician drilling cuttings samples (Figs. 2a and 2b,) from exploration of shale gas reservoir in the Baltic Basin were collected. Two types of samples were measured: cuttings after sieve separation and centrifugation, which contain the residue drilling mud and cuttings after Dean-Stark extraction and sieve analysis. By means of sieve analysis three fractions were obtained (Fig. 2b). For activity concentration measurements 80 g of each sample was examined under the same conditions. The samples were closed tightly and stored for 1 month to obtain equilibrium between primordial and daughters radionuclides. The drilling mud did not contain potassium ions, therefore the value of K-40 activity concentration is related only to the rocks, which is an advantage since potassium chloride is frequently added to the drilling mud, thus disturbing well logging spectrometry measurement. It can also influence the laboratory experiment when the Dean-Stark extraction is not provided.

(a)



(b)

Fig. 2. Ordovician drilling cuttings. (a) original, (b) fractioned: from the left <0.425 mm, 0.425-1 mm, 1-1.6 mm

The bulk of drilling wastes originated from different depth of the borehole. One depth was chosen for a second part of experiment concerning measurement of drilling cuttings after the Dean-Stark extraction and sieve analysis.

Gamma radiation measurements

Each sample was non-destructively analyzed using a portable gamma-ray spectrometer InSpector1000 Canberra consisting of NaI(Tl), 2”x2” stabilized probe with sensitivity of Cs-137 ~3.5% and multichannel analyzer designed for environmental screening. Before measurement, the detector was calibrated using a cesium Cs-137 source. The activity concentrations were determined using decay energies provided in Table 1. The measurement was conducted in a lead shield to eliminate the influence of background radiation (Fig. 3). Each sample was measured three times. Eighty grams of drilling cuttings were placed in a 48 mm diameter container (corresponding to probes diameter).

Fig. 3. Scintillation stabilized probe in lead shield used in the research

Absorbed dose rate (Dγ)

An absorbed dose is a dose of ionizing radiation in the outdoor air 1m above the surface of drilling cuttings samples originating from the gamma radiation. The formula is:

$$D\gamma = 0.0417A_{\mathrm{K}} + 0.462A_{\mathrm{Ra}} + 0.604A_{\mathrm{Th}} \tag{1}$$

where A_{K}, A_{Ra}, A_{Th} are the activity concentrations of K-40, Ra-226 and Th-232, respectively. According to UNSCEAR Report (UNSCEAR, 1993) the absorbed dose rate should not exceed the average global value of 55 nGy/h. The absorbed dose consists mainly of uranium, thorium and potassium radioisotopes. Others are insignificant in the gamma radiation exposure.

Results and discussion

The X-ray diffraction (XRD) showed that the major component of the studied material is quartz, sodium aluminum dioxide, aluminum silicate hydrate and wustite. To elucidate the structure of examined samples, the SEM examination was performed (Fig. 4). It confirmed the presence of quartz, silicate and sulfur in the form of pyrite framboids. The particle size distribution after the Dean-Stark extraction and sieve analysis is provided in Table 4.

Table 4. Fractions of drilling cuttings after sieve analysis

No.	Size range [mm]	Weight percentage [%]
	Before Dean-Stark extraction	
1	<0.425	46.5
2	0.425-1.0	26.3
3	1.0-1.6	27.2
	After Dean-Stark extraction	
4	<0.425	44.9
5	0.425-1.0	28.0
6	1.0-1.6	27.1

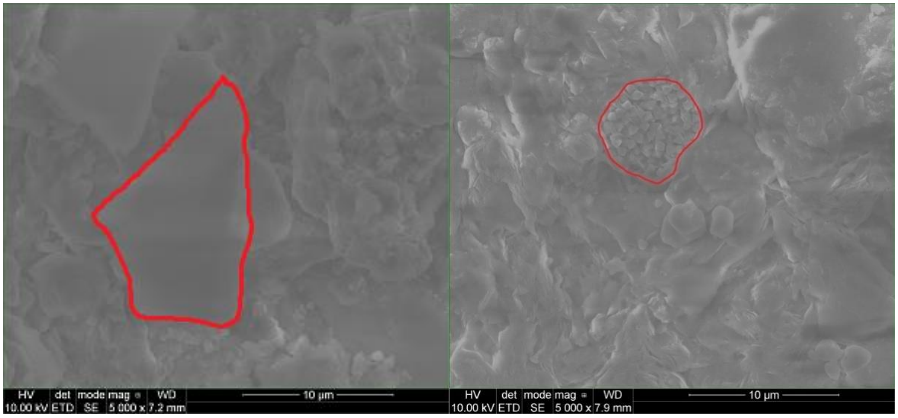


Fig. 4. SEM image of investigated material (quartz silt on the left side and pyrite framboid on the right side)

Radiation expressed in the activity concentration of radionuclides [Bq/kg] for analyzed samples is shown in Tables 5 (bulk wastes) and 6 (size fractions: BA and AA sample was also measured after sieve analysis). The average value from three measurements with standard deviation are provided. The activity concentration of Uranium U-238 was at an undetectable level, even during 24 h of exposure. Therefore, it does not pose any radiological hazard.

Table 5. Results of bulk drilling cuttings gamma radiation measurements (BX – before Dean-Stark extraction, AX – after Dean-Stark extraction, X -consecutive determination of samples SD – standard deviation)

No.	Sample	Mean concentration of K-40 [Bq/kg] with SD	Mean concentration of Th-232 [Bq/kg] with SD	Mean absorber dose rate *Dγ* [nGy/h]
		As received, before (B) Dean-Stark extraction		
1	BA	865±40	29.2±2.3	53.7
2	BB	838±39	29.5±2.4	52.7
3	BC	999±46	31.0±2.5	62.9
4	BD	905±42	29.2±2.3	57.0
5	BE	848±39	28.8±2.3	42.8
6	BF	680±31	28.8±2.3	53.4
		After (A) Dean-Stark extraction		
7	AA	834±38	23.3±1.9	48.9
8	AB	850±39	25.5±2.0	53.5
9	AC	867±40	26.3±2.1	54.6
10	AD	866±40	24.0±1.9	54.5

The absorbed dose associated with bulk drilling wastes was 54.1 nGy/h. Only two samples had higher absorbed dosages (BC 62.9 and BD 57.0 nGy/h) than the mean global value (55 nGy/h), however they did not constitute any radiological hazard. A diagram, comparing results from Table 5 is shown in Fig. 5. Fractioned samples are shown in Fig. 6.

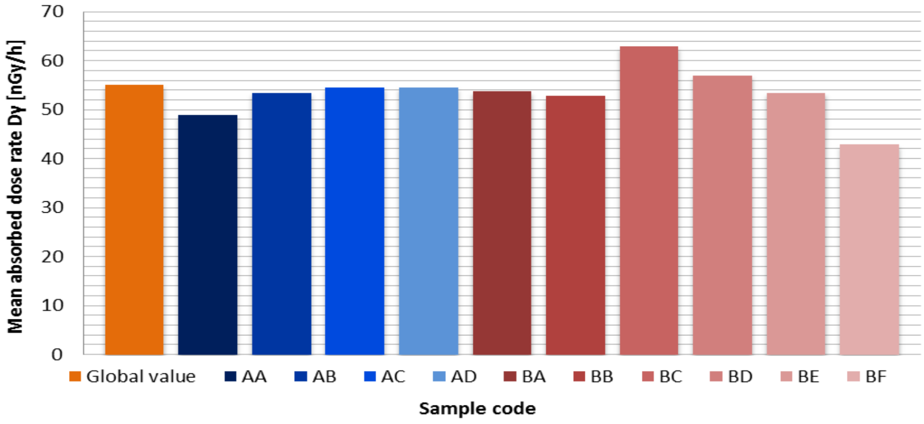


Fig. 5. Results of absorbed dose rate estimation in the bulk drilling cuttings samples (BX – before Dean-Stark extraction, AX – after Dean-Stark extraction, X -consecutive determination of samples**)** in relation to average global value (UNSCEAR, 1993)

The absorbed dose rate is greater for the drilling wastes before Dean-Stark extraction than after toluene extraction (53.9 nGy/h before Dean-Stark process and 53.8 nGy/h after Dean-Stark process). In the first case the sample from one depth had low level of natural radioactivity (680 Bq/kg for K-40 and 28.8 Bq/kg for Th-232). The range of activity concentration of potassium was 680-999 Bq/kg and of thorium 23.3-31.0 Bq/kg in the non-fractioned samples.

Table 6. Gamma radiation of drilling cuttings size fractions (BA before Dean-Stark extraction, AA after Dean-Stark extraction, 1-6 consecutive numbering of samples)

No.	Sample	Size range [mm]	K-40 [Bq/kg]	Th-232 [Bq/kg]	*D*γ [nGy/h]
		Before (B) Dean-Stark extraction			
1	BA	-	865±40	29.2±2.3	53.7
2	BA1	<0.425	992±46	30.8±2.5	60.0
3	BA2	0.425-1.0	905±42	28.3±2.3	54.9
4	BA3	1.0-1.6	825±38	26.7±2.1	50.5
		After (A)Dean-Stark extraction			
5	AA	-	834±38	23.3±1.9	48.9
6	AA4	<0.425	852±39	24.8±2.0	50.5
7	AA5	0.425-1.0	818±38	23.8±1.9	48.5
8	AA6	1.0-1.6	800±37	23.3±1.9	47.5

Table 6 shows that the dominating gamma radiation emitter in drilling cuttings is potassium K-40 (800-992 Bq/kg), which is in line with the data provided in Table 2. Thorium Th-232 was also observed and the results (23.3-30.8 Bq/kg) are again comparable to the literature values (Table 2). There is a difference in activity concentration of radionuclides before and after Dean-Stark extraction, because some of radionuclides can be leached out from the sample, and thus the concentration of K-40 and Th-232 decreases.

The analysis of specified fractions yielded different results. The elevated radionuclides concentration in smaller size fractions was observed and both factors seem to be negatively correlated. The level of gamma radiation in bulk wastes was not greater than in the level in the individual fractions of wastes. It was observed that radionuclides can be partially redistributed in fractioned drilling cuttings. Higher absorbed dose of gamma radiation could be associated with greater specific surface area of particles, which increases gamma-ray emission. On the other hand, gamma rays can penetrate rocks and it should be researched whether this correlation exists with poor penetrating radiation such as alpha or beta particles.

The comparison of analyzed samples is presented in Fig. 6. The absorbed dose criterion was selected to inform about the radiological hazard. The average absorbed dose rate in outdoor air 1 m above the drilling cuttings was 54.9 nGy/h. This result is lower than the global value of 55 nGy/h. In this regard, the drilling cuttings did not constitute any radiological hazard and can be utilized. Only the <0.425 mm size fraction before Dean-Stark extraction, exceeded the average global value and reached 62.4 nGy/h.

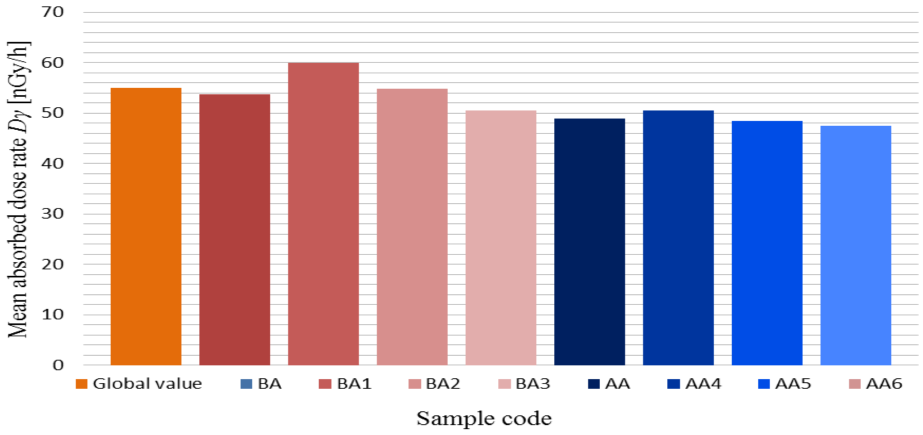


Fig. 6. Results of absorbed dose rate estimation in the fractioned drilling cuttings samples (BA – before Dean-Stark extraction, AA after Dean-Stark extraction, 1-6 consecutive numbering of samples) in relation to average global value (UNSCEAR, 1993)

The second part of the experiment aimed to check the relationship between the grain size and level of natural radioactivity. The radionuclides amount is negatively correlated with the size of grains in the drilling cuttings samples but on the other hand activity concentrations in the bulk wastes do not confirm the thesis. It can be stated with certainty, that Dean-Stark extraction leached some radioisotopes. The activity concentration and absorbed dose based on them, is higher in the samples before extraction. The difference is rather small – from a few Bq/kg for thorium to tens Bq/kg for potassium.

Final comments

The research concerned two aspects: relationship between natural radioactivity emission and grain size, and radioactivity of drilling wastes. It revealed that:

- the main radiological constituent in bulk and fractionated drilling cuttings is potassium K-40,
- the activity concentration is comparable with literature values and vary from 680 to 999 Bq/kg for potassium K-40 and from 23.3 to 31.0 Bq/kg for thorium Th-232. Other radioisotopes have undetectable concentrations,
- some of radionuclides were leached during Dean-Stark extraction,

- the examined drilling cuttings do not present any radiological hazard for humans and can be safely utilized. The absorbed dose (*Dγ*) is 54.1 nGy/h in respect to global average value 55 nGy/h,
- the radionuclides amount in the examined fractioned drilling cuttings increased, while the size of the grains decreased.

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