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Monitoring Of Natural Radionuclides In Oil And Gas Enterprises

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Abstract.
In paper the results of radiation monitoring of natural radionuclides on objects of the territory, technological equipment, raw materials samples, ready products of oil and gas production enterprises is date. The results of the study show that natural radionuclides ^{226}Ra, ^{232}Th and their fission products ^{214}Bi, ^{214}Pb, ^{228}Ac are extracted from the earth's interior together with oil and gas condensate and accumulate in the form of radioactive salt deposits in the internal surfaces of closed technological equipment us tanks, separators, pipelines and metal scraps with total specific activity up to 45449 Bq/kg. On basis of radiation monitoring, recommendations were developed to reduce environmental risks for the personnel of gas-producing enterprises. Two methods was been developed for accurate calibration of radioisotope level gauges under conditions of receipt with gas condensate and accumulation of natural radionuclides in closed process tanks during gas condensate processing.
Background*: Radiation safety is one of the main components of the safety of oil and gas companies and an important aspect of the sanitary and epidemiological well-being of the population. At present, natural radionuclides are significant radioactive pollutants of the environment. As a result, of the extraction, processing and storage of oil and gas, natural radionuclides extracted from the bowels of the earth pollute not only the environment, but also the personnel of the oil and gas enterprise to radiation hazard are exposed. In this work we carried out radiation monitoring of natural radionuclides and spectrometric analysis of the content of natural radionuclides in various soil samples, reservoir water, pipeline solids, finished products of enterprises oil and gas producing enterprises of LLC "Gazlineftegazdobycha" (Bukhara, Uzbekistan) and JV "Uz-Kor Gas Chemical" (Akshalak Karakalpacstan) A phenomenon has been revealed that radionuclides ^{214}Bi, ^{214}Pb, ^{228}Ac, present in gas condensate, gradually and continuously accumulate in closed containers and during processing they are found together with a free-flowing powdery intermediate product. The radioisotope level gauge (Berthold Technologies GmbH & Co. KG) is not working properly and it is impossible to accurately calibrate the radioisotope level gauge.*
Materials and Methods*: In this study the dosimetry devices for measuring gamma radiation were used, us digital spectrometer Identi Fiender Target (Thermo Fisher Scientific, Inc., USA), personal portable dosimeter and Radiagem 2000 dosimeter (Canberra, USA), Polymaster DKG-RM (Scientific Technical Center "Expert", Republic of Belarus). The measurement of the exposure dose rate of gamma radiation was carried out at a distance of 5.0 mm from the surface of controlled objects (solid deposits from the inner surface of process pipes, soil samples from reservoir drainage water, scrap metal from previously used well pipes, oil, gas condensate, primary free-flowing powder product gas condensate, end products in the form of ethylene and propylene granules). Spectrometry analysis of investigated samples was performed with a Radek gamma-beta spectrometer, MKGB-01 (Russia) with Ascinti software. To measure the specific activity of ^{226}Ra and ^{232}Th radionuclides, a Canberra gamma spectrometer with a germanium detector and Genie-2000 software was used.*
Results*: Radiating monitoring results showed that no radioactive contamination with natural radionuclides was observed on the territory, rooms and buildings, where the exposure dose of gamma radiation is at the level of the natural gamma background of 0.86 C/kg·s (12 μR/h). The results of gamma spectrometry analysis showed that there are no natural radionuclides ^{226}Ra and ^{232}Th in the ready refined products of oil and gas condensate. In some samples, the specific activity of the ^{226}Ra radionuclide exceeds by 4.3 times the allowable standard values and the specific activity of the ^{232}Th radionuclide exceeds by 19.5 times the allowable standard values. It was found that radionuclides ^{214}Bi, ^{214}Pb, ^{228}Ac, present in gas condensate, gradually accumulate in closed containers with a free-flowing intermediate powder product, the total gamma background increases and the level of the intermediate product is incorrectly measured by a radioisotope level gauge. In particular, in radioisotope level gauges (Berthold Technologies GmbH & Co. KG) in the radiation source block with a charged source of ionizing radiation Cs-137, an additional increase in the gamma radiation dose rate is occurred, resulting in an increase or decrease of the analog signal output current of radioisotope level gauge It was found that the ^{214}Bi, ^{214}Pb, ^{228}Ac radionuclides present in the gas condensate constantly and continuously accumulate in closed containers with a loose powdery intermediate product, the overall gamma background increases, and as a result of this phenomenon, the level of the loose powdery intermediate product in the closed container is incorrectly measured*

by a radioisotope level gauge. . In radioisotope level gauges in the radiation source block with a source of ionizing radiation Cs-137, an additional increase in the dose rate of gamma radiation occurs, as a result of which the output current of the analog signal of the radioisotope level gauge increases or decreases.

Conclusion: *Based on the results of radiation monitoring and spectrometric analysis in the studied samples of the content of natural radionuclides, recommendations were developed to reduce the environmental risks of overexposure to gamma radiation by the personnel of gas and oil producing enterprises. Methods for accurate calibration of radioisotope level gauges under emergency conditions of accumulation of natural radionuclides in closed process tanks during the processing of initial gas condensate have been developed.*

Key Word*: Natural Radionuclides; Radiating Monitoring; Formation Water; Gas Condensate; Gamma-Spectrometry Method; Gamma-radiation Exposure Dose; Radiation Safety; Radioisotope Level Gauges; Calibration*

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I. Introduction

Formation waters are the main source of contamination of oil fields with radioactive isotopes of natural radionuclides [1]. Formation waters contain lot of alkaline earth elements, including radioactive radium. The primordial radioisotopes ^{235}U, ^{238}U and ^{232}Th and their decay daughters comprise the naturally occurring radioactive materials that may be present in oil and gas bearing formations and are released as by-products of oil and gas production [2] and the total content of dissolved compounds in reservoir waters varies from 5 to 300 g/l [3]. The main dose-forming radionuclides in natural waters, along with uranium isotopes ^{234}U, ^{238}U are isotopes of radium: ^{226}Ra, ^{228}Ra, ^{224}Ra. They have a high index of radiotoxicity, which is associated with their long-term deposition in the body in the case of high-energy alpha and beta radiation [4]. The concentrations of radionuclide ^{226}Ra can exceed the background values by 100–1000 times [5]. In the case of soil contamination with formation waters, radioactive territories of technogenic contamination are formed, and as a result, natural radionuclides can enter surface, ground and underground waters.

At the current technological level of oil and gas production, it is impossible to avoid such a negative phenomenon, and therefore, during the operation of oil and gas producing enterprises, it becomes necessary to constantly monitor natural radionuclides. Currently, topical tasks are radiation monitoring of natural radionuclides at technological facilities of oil and gas producing enterprises and assessment of the degree of environmental pollution by natural radionuclides.

The purpose of the study is to conduct radiation monitoring of territories, buildings and structures, wells, technological facilities of oil and gas producing enterprises LLC "Gazlineftegazdobycha" and JV "Uz-Kor Gaz Chemical", as well as to investigation of them by method spectrometric analysis quantuty content in a various samples us formation water, gas condensate, solid deposits from the internal part pipelines, scrap metal, gas condensate processing products.

II. Material And Methods

This prospective comparative study was carried out on objects of dosimetry control were the territory, rooms, buildings of oil and gas processing enterprises, boreholes, scrap metal of spent borehole pipes, inactive and operating gas condensate pipelines, process equipment. The materials for gamma spectrometry analysis are formation waters, initial and purified gas condensate, crude and refined oil, finished products (ethylene and polypropylene granules), solid sediments from the inner surface of process pipelines and process equipment, tanks, separators, and others. This prospective comparative study of content of natural radionucledes was carried out on objects of oil and gas production enerprises were the territory, rooms, buildings, boreholes, scrap metal, spent borehole pipes, inactive and operating gas condensate pipelines, active and inactive process equipment. The materials for gamma spectrometry analysis are formation waters, initial and purified gas condensate, crude and refined oil, finished products (ethylene and polypropylene granules), solid sediments from the inner surface of process pipelines and process equipment, tanks, separators, and others.

Table 1 shows the main characteristics of natural radionuclides ^{226}Ra, ^{228}Ra, ^{224}Ra [4].

Table no 1: Shows main characteristics of natural radionuclides ^{226}Ra, ^{228}Ra, ^{224}Ra.

Radionuclide	Half-life $T_{1/2}$	Type of radiation and energy, keV
^{226}Ra and its short-lived daughter decay products. Establishment of radioactive equilibrium in the group - 23 days		
^{226}Ra	1600 y	α: 4780
^{222}Rn	3.824 d	α: 5490
^{218}Po	3.05 m	α: 6000
^{214}Pb	26.8 m	β: 1040

^{214}Bi	19.9 m	β: 3280
^{214}Po	$1.64 \cdot 10^{-4}$ s	α: 7687
^{228}Ra and its short-lived daughter decay products. Establishment of radioactive equilibrium in the group - 1.5 days		
^{228}Ra	5.75 y	β: 55
^{228}Ac	6.13 h	β: 2087
^{224}Ra and its short-lived daughter decay products. Establishment of radioactive equilibrium in the group - 2.7 days		
^{228}Ra	5.75 y	β: 55
^{212}Pb	10.64 h	β: 569
^{216}Po	0.15 s	α: 6780
^{212}Po	$2.98 \cdot 10^{-7}$ s	α: 8780
^{212}Bi	66.55 m	α(0,36): 6050 β(0,64): 2246

Identification of ^{214}Pb radionuclide on analytical lines gamma-radiation by analytical peaks energy of 609 keV, 296 keV, 352 keV, ^{241}Bi radionuclide on analytical lines gamma-radiation with analytical peaks energy of 1120 keV and 1764 keV, ^{228}Ac radionuclide on analytical lines gamma-radiation with analytical peaks energy of 338 keV, 911 keV, 965 keV, and ^{212} Pb radionuclide on analytical lines with analytical peaks energy of gamma-radiation 239 keV was spent. Table 2 Shows basic energies of gamma quanta of daughter decay products of radium isotopes.

Table no 2: Shows basic energies of gamma quanta of daughter decay products of radium isotopes [6].

Determined radionuclide	Energy of the main analytical peaks, keV	Decay yield of gamma quanta, %
	Daughter radionuclide of the radioactive series ^{226}Ra	
^{214} Pb	295.6	18.5
	351.9	35.8
	609.3	44.8
^{214} Bi	1120.3	14,8
	1764.5	15.4
	Daughter radionuclide of the radioactive series ^{228}Ra	
^{228} Ac	338.3	11.3
	911.2	26.6
	964.8+968.9	Σ= 21,3

III. Result

Table 3 shows the results of radiation monitoring of the territory and technological facilities of LLC «Gazlineftegazdobycha».

Table no 3: Shows the results of radiation monitoring of the territory and technological facilities of LLC «Gazlineftegazdobycha».

№	**Object, Process Equipment**	**Gamma-radiation Exposure Dose Rate (EDR), C /kg·s**
1	Territory, rooms and premises*	0.86
2	Pipeline gas distribution line (acting) *	10.8
3	Pipeline gas distribution line (inactive) *	15.8
4	Scrap metal and metal waste*	11.8
5	Territory, rooms and premises**	0.86
6	Receiver**	53.8
7	Capacities (E-1, E-2, E-3)**	52.5
8	Heat exchanger**	14.3
9	Territory, rooms and premises***	0.86
10	Gas condensate waste ***	5.9
11	Ball valve***	129
12	Territory, rooms and premises****	0.86
13	Scrap metal and metal waste****	19
14	Territory, rooms and premises*****	0.86
15	Scrap metal and metal waste*****	86

*Remark: * Gas processing plant «Uchkur»; ** Gas processing plant «Daykhatun»;*** Gas processing plant «Kulbeshkek»;**** Gas processing plant «Mullakhol»; ***** Well «Yangikazgan»*

Table 4 Shows the results of gamma-spectrometer analysis of investigated samples received from objects of LLC «Gazlineftegazdobycha».

Table no 4: Shows results of gamma-spectrometer analysis of investigated samples.

№	Samples	Natural Radionuclide Specific Activity, Bk/kg	
		^{226}Ra	^{232}Th
1	Firm deposit from internal surface of cut off inactive pipeline №2*	43 492.5 ±410	19 515.6 ±1870
2	Gas distribution line №4*	32 194.5 ±310	9 412.5 ±870
3	Ground soil from plum formation waters*	5 331.3 ±525	2 452.7 ±230
4	Soil sample in the area around*	2 859.6 ±275	1 071.9 ± 98
5	Smear from internal surface of scrap metal*	1 923.4 ± 125	1 846 ± 110
6	Soil sample in the area around**	1 810 ±126	1 200 ±84
7	Contaminated Priming***	17 923.4±1625	4 984.6 ±420
8	Formation water*	292.7 ±150	152.4 ±150
9	Oil*	–	–
10	Gas condensate*	–	–

Remark: Gas processing plant «Uchkur»;** Oil well No. 374; ***well "Yangilangan"*

It has been established, that at the technological facilities of the JV «Uz-Kor Gas Chemical» there is an excess of the level of the natural gamma background to a maximum level of 810 times higher than the background (Table 5, No. 5).

Table no 5: Shows of radiating monitoring of objects and process equipment of gas production enterprise of JV «Uz-Kor Gas Chemical».

№	Object, process equipment	Gamma-radiation exposure dose on equipment surface, C /kg·s	Gamma-radiation exposure dose on distance 1.0 m, C /kg·s
1	Territory, rooms, premises*	0. 86	0. 86
2	Heat exchanger*	330	32
3	Spherical tank*	122	7.9
4	Filter equipment F-5201 A*	90	8.6
5	Filter equipment F-5201 B*	697	50
6	Entrance thread block*	79	3.6
7	Territory**	0. 86	0. 86
8	Formation water**	1.0	0. 86
9	Territory***	0. 86	0. 86
10	Buffer capacity***	11	0. 86

Table 6 shows the gamma-spectrometer analysis of investigated samples received from objects of the JV «Uz-Kor Gas Chemical» (wells "Urga" and "Surgil"). It was been found that in the formation waters, the gas condensate and the powdered product of gas condensate contains natural radionuclide ^{226}Ra, ^{228}Ra and their radioactive decay products ^{214}Bi, ^{214}Pb, ^{228}Ac, but all natural radionuclides are within the limits of permissible concentrations established in sanitary standards [7].

Table no 6: Shows the results gamma-spectrometer of the analysis of investigated samples.

№	Sample	Natural radionuclide specific activity, Bk/kg						Remark
		^{226}Ra	^{214}Pb	^{214}Bi	^{228}Ac	^{212}Pb	^{212}Bi	
1	Gas condensate from separator № C 101	-	10±3	12±1	-	-	-	clean
2	Gas condensate from ziro separator	-	-	-	-	-	-	clean
3	Formation water buffer tank	153±42	33±5	52±8	28±8	8±2.5	-	clean
4	Formation water from separator	162±40	30±5	35±6	32±9	7.5±2	-	clean
5	Powdery bulk intermediate product	-	-	-	-	-	-	clean
6	Granules ethylene	-	-	-	-	-	-	clean
7	Granules polypropylene	-	-	-	-	-	-	clean

In a Fig. 1 shows gamma spectrum of the hard coating of the internal inactive slotted pipe of line No. 2 from gas processing plant «Uchkur» (LLC «Gazlineftegazdobycha»).

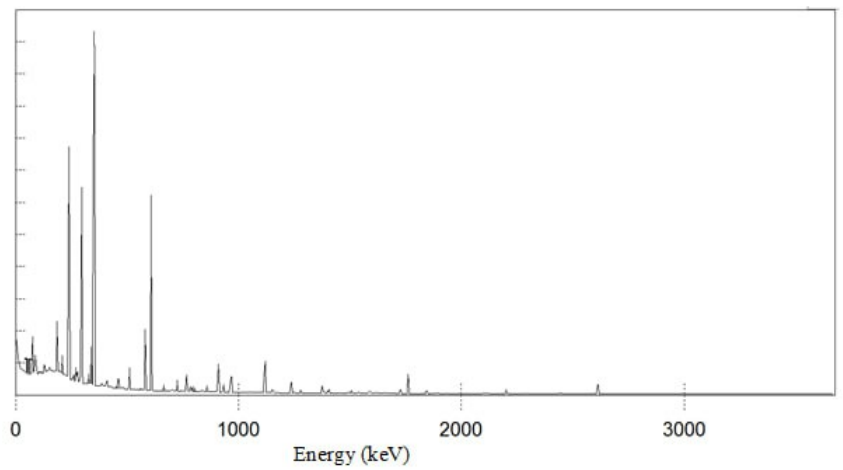

Fig. 1. Gamma spectrum of the hard coating of the internal inactive slotted pipe of line No. 2 from gas processing plant «Uchkur» (LLC «Gazlineftegazdobycha»).

IV. Discussion

It has been found (Table 3) that natural radionuclides mainly accumulated in the form of salt deposits on the internal surfaces of closed equipment, including in scrap metal from spent drill pipes, in places where formation waters are discharged and on lines for transporting raw gas condensate. Content radium in formation water $C_{Ra,f.w.}$ is equal to [8]:

$$C_{Ra\ f.w.} = C_{Ra\ a.o.} / K_r \quad (1)$$

where $C_{Ra\ a.o.}$ is the content of radium in anhydrous oil (approximately $2.8 \cdot 10^2$ Bq/m^3); K_r – balanced distribution coefficient of radium between oil and water ($K_r = 2.9\ 10^{-4}$).

When formation water rises along the wellbore, radioactive salts precipitate in electric centrifugal pumps, tubing and surface tanks. Specific activity precipitation reaches up to $4 \cdot 10^8$ Bq/kg. Basically, sediments by 90…95% consist of radiobarite – Ba(^{226}Ra)SO_4 with minor impurities of other compounds [9].

The results of gamma spectrometry analysis showed (Table 4) that there are no natural radionuclides ^{226}Ra and ^{232}Th in the ready refined products of oil and gas condensate. However, in some samples, the specific activity of the ^{226}Ra radionuclide exceeds by 4.3 times the allowable standard values of the minimum significant specific activity (MSSA) of the ^{226}Ra radionuclide at the workplace (MSSA is $1 \cdot 10^4$Bk) and the specific activity of the ^{232}Th radionuclide exceeds by 19.5 times the allowable standard values of the minimum significant specific activity of the ^{232}Th radionuclide at the workplace (MSSA is $1 \cdot 10^3$Bk) [7]. During gamma-activation analysis of a solid deposit obtained from the surface of an inactive sheared pipe (line No. 2, Gas processing plant «Uchkur»), a ^{214}Pb radionuclide was determined having analytical gamma peaks of 242 keV, 295 keV, 352 keV, as well as a ^{214}Bi radionuclide having analytical gamma peaks at 609 keV, 1120 keV, and 1764 keV (Fig. 1).

It was been found (Table 6) that, no radioactive contamination with natural radionuclides on the territory, rooms, buildings, gas condensate, ready products (granules ethylene and polypropylene) are absent, where the exposure dose of gamma radiation is at the level of the gamma background (0.86 C/kg·s or 12 μR/h).

A phenomenon has been revealed that radionuclides ^{214}Bi, ^{214}Pb, ^{228}Ac, present in gas condensate, gradually and continuously accumulate in closed containers and during processing they are found together with a free-flowing powdery intermediate product. As a result of this, the general gamma background increases many times and, as a result, an incorrect measurement of the level of a powdered bulk product in a closed container occurs. In this non-standard situation in radioisotope level gauges (Berthold Technologies GmbH & Co. KG) in the radiation source bloch with a source of ionizing radiation Cs-137, an additional increase in the dose rate of gamma radiation occurs, and in the detection bloch of the radioisotope level gauge, an increase or decrease in the output current of the analog signal occurs, those. The radioisotope level gauge is not working properly and it is impossible to accurately calibrate the radioisotope level gauge.

Fig. 2 shows dependencies signal current (I, mA) from level (H, m) intermediate powdered loose product in close tank

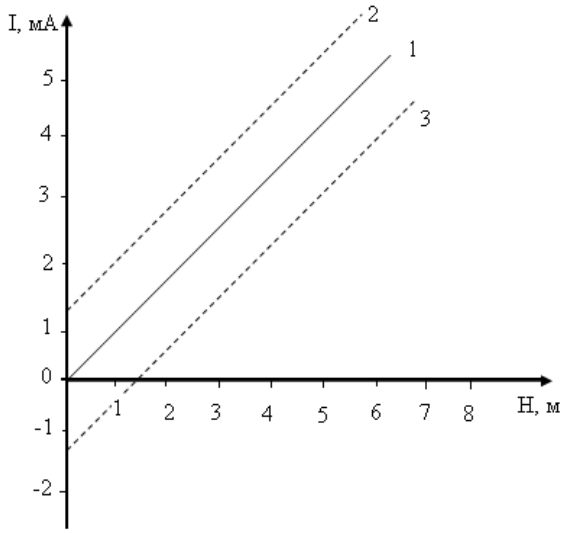

Fig. 2. Dependencies signal current (I, mA) from level (H, m) intermediate powdered loose product in close tank:
1 - normal dependencies of level intermediate powdered loose product (no natural radionuclides); 2, 3 - abnormal dependencies of level intermediate powdered loose product (natural radionuclides are present).

To overcome the emergency and accurately calibrate radioisotope level gauges, we used 2 methods:

The first method. For precise calibration of the radioisotope level gauge, an alarm current correction unit was developed, which was built into the detection unit of the radioisotope level gauge (Fig. 3). The current correction unit is designed for rectify (increase or decrease) the analog signal current and supply it to the information collection and analysis unit in the 0-5.0 mA, 4.0-20.0 mA connectors [10].

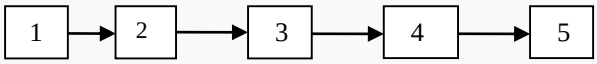

Fig. 3. Functional diagram of the detectors of LB440 level gauge with a block for correcting the current signal:
1 – radiation source block with Cs-137 radiation ionization source; 2 - closed tank with a intermediate powdered loose product; 3 - detection block; 4 – signal current correction block; 5 - electronic switchboard block.

The second method. The outer surface of the closed tank with the accumulated intermediate powdery bulk product was sheathed with a special protective screen made of sheet lead (Pb) 5 mm thick and 25 mm high, which was installed in front of the collimator in the path of radiation of ^{137}Cs radionuclide gamma rays, which ensured complete absorption of additional radioactivity of natural radionuclides. As a result of the installation of a lead shield in the radiation source bloch, a signal current was formed in the detection block in the 0-5.0 mA connectors in the case of the minimum product level and in the 4.0-20.0 mA connectors at the maximum product level
i.e. an accurate calibration of the radioisotope level gauge has been achieved.

V. Conclusion

According to the results of radiation monitoring of oil and gas industry enterprises and spectrometric analysis of the studied samples obtained from these enterprises, the quantitative content of natural radionuclides was determined, on the basis of which recommendations were developed to reduce environmental risks for the personnel of gas and oil producing enterprises LLC Gazliftegazdobycha and JV LLC Uz-Kor Gas Chemical . Two methods have been developed for accurate calibration of radioisotope level gauges under conditions of receipt with gas condensate and accumulation of natural radionuclides in closed process tanks during gas condensate processing.

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