ТЕХНІКА ТА МЕТОДИ ЕКСПЕРИМЕНТУ

УДК 621.039.638

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EVALUATION OF NATURAL RADIONUCLIDES FOR MICA AND QUARTZ IN EASTERN DESERT OF EGYPT, USING GAMMA-RAY SPECTROMETRY

By applying high–resolution γ-ray spectroscopic system, the various radionuclides of mica and quartz samples have been identified quantitatively. The specific activity of ^{226}Ra, ^{232}Th and ^{40}K in 25 mica samples collected from 5 locations and 15 quartz samples from 3 locations of geographical areas located in G. Kadabora in Central Eastern Desert of Egypt, were determined by gamma ray spectrometry with a high-purity germanium (HPGe) detector. This subject is important in environmental radiological protection, since mica and quartz are widely used as raw materials in different industries. The results of analysis for ^{238}U, ^{232}Th and ^{40}K specific activities were found to be higher than the permissible level for all mica and quartz samples. The radium equivalent activities in Bq/kg, dose rate in nGy/hr, external and internal hazards in nGy/yr and also (^{232}Th/^{238}U) ratios Clark's value s are calculated. From this study, it is clear that G. Kadabora, Central Eastern Desert, Egypt can be considered unsafe to use as raw materials.

Keywords: natural radioactivity, mica and quartz, external hazard index.

Introduction

Natural radioactive minerals deposits are found in suitable geological environment like unconformity contact, veins, surficial etc. [1]. There are few regions in the world that are known for high background radiation areas, where the local geological controls and geochemical effects cause enhanced levels of terrestrial radiation [2]. The greatest contribution to mankind's exposure comes from natural background radiation, and the worldwide average annual effective dose is 2.4 mSv. However, much higher levels of exposure are usual for inhabitants of natural high background radiation areas. High radiation above the earth is mainly due to naturally occurring radioactive elements in the earth's crust such as for ^{238}U, ^{232}Th and ^{40}K. Areas at high altitudes are also affected by cosmic radiations [3].

It is necessary to study the radiation and radioactivity to assess the dose to the population, in order to know the health risks, and to have a reference study to document changes in the environmental radioactivity in rock samples [4 - 6].

The present study initiates a radiological assessment program for the area of study to establish a database reference of radioactivity background levels in the Eastern Desert region.

The samples were collected from G. Kadabora, Central Eastern Desert of Egypt. About 66 km to the south of Quseir city, injected with numerous pegmatite's. This location at latitude 30º 25´ and longitude 34º 30´ (Fig. 1).

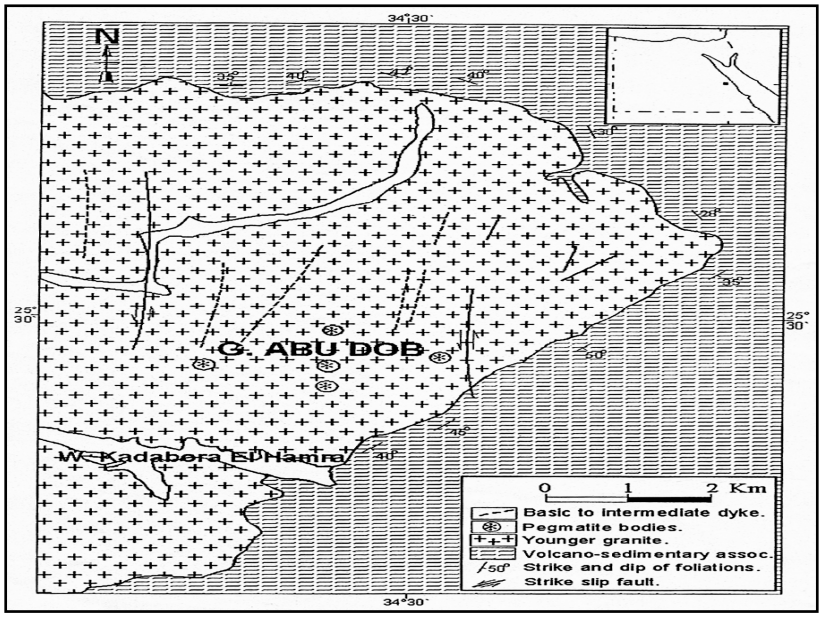

Fig. 1. Geological map of Gabal Abu Dob area.

The area is covered with numerous pegmatite's. Most of the pegmatite's are zoned, the zonation starts with milky quartz at the core followed by feldspars, which vary in color from rose to milky and in composition from K-feldspar to Na-feldspar. Mica flakes (up to 0.5 thicknesses) occur at the gradational contacts between K-feldspars and Na-feldspars.

In the present study, 25 mica samples were collected from 5 different locations, 15 quartz samples from 3 different locations, were analyzed using gamma- ray spectrometry to determine the specific activity of ^{226}Ra, ^{232}Th and ^{40}K, The results are used to assess the potential radiological hazards associated with these materials by computing the radium equivalent activity index, the indoor gamma absorbed dose rate, external hazard, internal hazard and radioactivity level index were also determined and evaluated for all samples under investigation.

Experimental procedure

Sampling and Sample Preparation

The samples under investigation were collected from G. Kadabora in Central Eastern Desert of Egypt. Mica samples were collected from five different locations, five samples from each one. Also quartz samples were collected from three locations, five samples from each one. A total of 40 rock samples, each about 1 kg in weight, were crushed, homogenized and sieved to about 100 mesh by a crushing machine. The samples were then placed for drying at 105 ºC for 24 h to ensure that moisture is completely removed. The samples were transferred to beakers polyethylene container of 250 cm^3 volume. Each sample was carefully sealed for four weeks to reach secular equilibrium between ^{232}Th and ^{238}U and its short lived daughter products [7].

Radiometric Analysis

Spectra for different samples were measured with a high-purity germanium (HPGe) detector of high-resolution gamma-ray spectrometer. The individual samples were placed on the detector manually and each sample was analyzed for a time of about 70000 sec. The detector has a resolution of 1.85 keV for the 1332.5 keV gamma-ray transition of ^{60}Co with relative efficiency 30 %. To reduce the gamma-ray background, a cylindrical lead shield with a fixed bottom and movable cover shield the detector. The lead shield contained two inner concentric cylinders of copper and cadmium.

The detection system was calibrated using multi-gamma ray standard source MGS-4 to perform the energy calibration of the detector [8]. The efficiency curve of the HPGe detector in the energy range from 186 to 2450 keV was obtained through two stages. In the first stage the relative efficiency curve of the detector was performed using a ^{226}Ra point source. In the second stage, the average relative curve of the detector was normalized to an absolute potassium chloride has been used [9].

Table 1 gives the energies used to determine the concentrations of different radionuclides and their yields [10]. In order to determine the background distribution in the environment around the detector, an empty bottle was counted in the same manner and in the same geometry as the samples. The background spectra were used to correct the areas of gamma rays of measured isotopes.

Table 1. **Photoenergies and branching ratios for different radionuclide's**

Nuclide	Energy, keV	Photon per disintegration, %
^{238}U series		
^{234}Th	63.3	3.8
^{226}Ra	186.1	3.3
	295.1	19.2
	352.1	37.1
	609.3	46.1
^{214}Pb	768.4	5.1
	934.1	3.2
^{214}Bi	1120.3	15
^{232}Th series		
^{228}Ac	209.4	4.1
	338.4	12.4
	462.1	4.6
	911.2	29.1
	966.6	23.2
^{212}Bi	727.3	6.7
	785.5	1.1
	1620	1.7
^{208}Ti	583.1	30.9
^{235}U series		
^{235}U	143.8	10.5
	163.3	4.7
	185.7	53.1
	205.3	4.7
^{40}K	1460.0	10.7

Results and Discussion

Detection limits

The lowest limits of detection (LLD) of the measuring system, which is required to estimate a minimum detection level for appropriate determination of radionuclides using analytical technique in each sample, were obtained following environmental measurement laboratory procedure using the expression [14, 15].

$$LLD = 4.66\ S_b / \zeta \cdot I_\gamma,$$

where S_b is the standard error of the net back-ground count rate in the spectrum of the radionuclide; ζ is the counting efficiency; I_γ is the abundance of gamma emission per radioactive decay.

The LLD of a measuring system measures its operating capability without the influence of the sample. The LLD values for ^{40}K, ^{232}Th and ^{238}U are obtained in Table 2.

Table 2. **The lowest limit of detection (LLD) for the radionuclide ^{40}K, ^{238}U and ^{232}Th**

Nuclide	Lowest detection limit (Bq/kg)
^{40}K	9.36
^{238}U	1.32
^{232}Th	1.35

The specific activity

The radioactivity concentration of naturally occurring radionuclides ^{238}U, ^{232}Th series and ^{40}K for mica and quartz samples were calculated and reported in Table 3.

From this table it is seen that the concentration of ^{238}U is higher than the concentration of ^{232}Th in mica samples, but in quartz samples the concentration of ^{232}Th is higher than the concentration of ^{238}U. It is clear that the average results of G. Kadabora in Central Eastern Desert samples are higher than the permissible levels (35, 35, 370 Bq/kg) for uranium, thorium and potassium.

Table 3. **The average specific activity of ^{238}U series, ^{232}Th series and ^{40}K (Bq/kg) for mica samples**

Kind	Lab Cod	N	Specific activity, Bq · kg^{-1}		
			A_U	A_{Th}	A_K
			Range mean		
Mica	K-2/14	5	2124 - 2131 2128	1116 - 1129 1122	1692 - 1700 1696
	K-2/3	5	1867 - 1874 1871	1055 - 1067 1061	1741 - 1754 1747.9
	K-2/2	5	2397 - 2406 2402	1328 - 1341 1334	1624 - 1634 1629
	K-2/7	5	3094 - 3105 3099	2054 - 2063 2058	1886 - 1894 1890
	K-2/1	5	190232 - 190249 190240	108929 - 108939 108934	11362 - 11378 11370

Radium equivalent activity

To compare the specific activity of the samples containing ^{226}Ra, ^{232}Th and ^{40}K, the radium equivalent index ($^{226}Ra_{eq}$) has been used to obtain the total amount of these activities [11, 12]

$$^{226}Ra_{eq} = 1.43C_{Th} + C_{Ra} + 0.077C_K,$$

where C_{Ra}, C_{Th} and C_K are the activities concentration in (Bq/kg) of ^{226}Ra (U series), ^{232}Th and ^{40}K respectively.

Estimation of the absorbed gamma dose rate and the effective dose rate

The absorbed dose rate at one meter above the ground due to the radioactivity in the samples are calculated using the following equation

$$D_R = k_kC_k + K_{Th}C_{Th} + K_{Ra}C_{Ra},$$

where D_R is the absorbed dose rate (nGy/h), also K_k, K_{Th} and K_{Ra} are the conversion factors (or dose rate coefficients) expressed in (nGy hr^{-1}per Bq/kg^{-1}) for potassium (0.043), Thorium (0.662) and Radium (0.427), respectively [13].

External and internal hazard index

To limit the annual external gamma-ray dose from materials to 1.5 mGy for the samples under investigation, the external hazard index H_{ex} is given by the following equation

$$H_{ex} = C_{Ra}/370 + C_{Th}/259 + C_K/4810 <1.$$

The internal exposure to ^{222}Rn and its radioactive progeny is controlled by internal hazard index, (H_{in}) which is given by

$$H_{in}= C_{Ra}/185 + C_{Th}/259 + C_K/4810 < 1.$$

The value of radium equivalent in Bq/kg, dose rate in nGy/hr, external hazard, internal hazards and also Clark's value ($^{232}Th/^{238}U$) in ppm for mica and quartz samples in G. Kadabora, Central Eastern Desert, Egypt are listed in Table 4. From the table it is clear that $^{232}Th/^{238}U$ ratio for mica samples are less than Clark's value, (U - enrichment). While ($^{232}Th/^{238}U$) ratio for quartz samples are more than Clark's value (Th - enrichment).

Table 4. **The average specific activity of ^{238}U series, ^{232}Th series and ^{40}K (Bq/kg), for quartz samples**

Kind	Lab Cod	N	Specific activity, Bq · kg^{-1}		
			A_U	A_{Th}	A_K
			Range mean		
Quartz	K-2/9		313 - 317 315	874 - 882 878	1691 - 1701 1696
	K-1/7		54 - 58 56	65 - 71 68	1818 - 1829 1824
	K-2/8		313 - 319 316	447 - 457 452	897 - 910 903

Table 5. **The values of radium equivalent (Bq/kg) dose rate (nGy/hr), external hazard, internal hazard and $^{232}Th/^{238}U$**

Sample No.	Ra_{eq}, Bq/kg	D_R, nGy/hr	H_{ex}	H in	$^{232}Th/^{238}U$
K-2/14	3864	1724	10.4	16.1	1.5
K-2/3	3523**	1576**	9.5**	14.5**	1.7
K-2/2	4436	1979	11.9	18.4	1.6
K-2/7	6189	2767	16.7	25.1	2.0
K-2/1	346893*	153836*	937*	1451*	1.7
K-2/9	907**	415**	2.5**	3.3**	8.4
K-1/7	3523*	1576*	9.5*	14.5*	3.6
K-2/8	1032	473	2.8	3.64	4.3

* The highest value.
** The lowest value.

Also we concluded that the average values of radium equivalent of all mica and quartz samples are higher than the recommended maximum value 370 Bq/kg and the values of dose rate for mica and quartz samples are higher than the international average mean value 55 nGy/h [16].

The calculate average external hazard index and internal hazard are more than unity. From the above results, it is noticed the level of radioactivity of mica and quartz samples are higher than the permissible levels, and cannot use as raw materials.

Conclusion

The results of analysis of ^{238}U, ^{232}Th and ^{40}K specific activities for mica and quartz samples were found to be higher than the permissible levels. The ratio of $^{232}Th/^{238}U$ (Clark's value = 3.5) for all mica samples are less than Clark's value, (U - enrichment), for all quartz samples are more than Clark's value, (Th - enrichment). Also dose rate, external hazard index and internal hazard index are higher than the international average mean value.

Finally, it is quite clear that G. Kadabora, Central Eastern Desert, Egypt can be considered unsafe to use raw materials (mica and quartz) in industry. The people working in that area have to be taking possible precautions and protection against the high radioactivity. Radiation protection must be used to reduce the risk.

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ОЦІНКА ВМІСТУ ПРИРОДНИХ РАДІОНУКЛІДІВ У СЛЮДІ ТА КВАРЦІ ІЗ СХІДНОЇ ПУСТЕЛІ ЄГИПТУ ЗА ДОПОМОГОЮ ГАММА-СПЕКТРОМЕТРІЇ

За допомогою гамма-спектроскопічної системи високої роздільної здатності було визначено кількісно вміст різних радіонуклідів у зразках слюди та кварцу. Методом спектрометрії гамма-випромінювання германієвим (HPGe) детектором високої чистоти була визначена питома активність ^{226}Ra, ^{232}Th і ^{40}K у 25 зразках слюди, зібраних у п'яти місцях і 15 зразках кварцу з трьох місць географічного району, розташованого в місцевості G. Kadabora в центральній частині Східної пустелі Єгипту. Ця проблема має важливе значення для радіологічної охорони навколишнього середовища, оскільки слюда та кварц широко використовуються як сировина в різних галузях промисловості. У результаті аналізу встановлено, що питома активність ^{238}U, ^{232}Th та ^{40}K виявилася вищою, ніж допустимий рівень для всіх зразків слюди та кварцу. Розраховано радієвий еквівалент активності в Бк/кг, потужність дози в нГр/год, небезпеку зовнішнього та внутрішнього опромінення в нГр/рік, а також значення відношення Кларка (^{232}Th/^{238}U). Із цього дослідження стає ясно, що сировину з місцевості G. Kadabora в центральній частині Східної пустелі Єгипту не можна вважати безпечною для використання.

Ключові слова: природна радіоактивність, слюда, кварц, індекс зовнішніх небезпек.

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ОЦЕНКА СОДЕРЖАНИЯ ЕСТЕСТВЕННЫХ РАДИОНУКЛИДОВ В СЛЮДЕ И КВАРЦЕ ИЗ ВОСТОЧНОЙ ПУСТЫНИ ЕГИПТА ПРИ ПОМОЩИ ГАММА-СПЕКТРОМЕТРИИ

При помощи гамма-спектроскопической системы высокого разрешения было определено количественно содержание различных радионуклидов в образцах слюды и кварца. Методом спектрометрии гамма-излучения германиевым (HPGe) детектором высокой чистоты была определена удельная активность ^{226}Ra, ^{232}Th и ^{40}K в 25 образцах слюды, собранных в пяти местах и 15 образцах кварца из трех мест географического района, расположенного в местности Г. Кадабора в центральной части Восточной пустыни Египта. Эта проблема имеет важное значение для радиологической охраны окружающей среды, поскольку слюда и кварц широко используются как сырье в различных отраслях промышленности. В результате анализа установлено, что удельная активность ^{238}U, ^{232}Th и ^{40}K оказалась выше, чем допустимый уровень для всех образцов слюды и кварца. Рассчитан радиевый эквивалент активности в Бк/кг, мощность дозы в нГр/ч, опасность внешнего и внутреннего облучения в нГр/год, а также значение отношения Кларка (^{232}Th/^{238}U). Из этого исследования становится ясно, что сырье из местности Г. Кадабора в центральной части Восточной пустыни Египта нельзя считать безопасным для использования.

Ключевые слова: естественная радиоактивность, слюда, кварц, индекс внешних опасностей.

Надійшла 25.11.2011
Received 25.11.2011