NATURAL RADIOACTIVITY IN EGYPTIAN AND INDUSTRIALLY USED AUSTRALIAN BAUXITES AND ITS TAILING RED MUD.

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Abstract

Red mud is produced in considerable masses as a waste product in the production of aluminum from bauxite. It may be used for industrial or agricultural purposes. According to it's genesis by weathering and sedimentation bauxites contain high concentrations of uranium and thorium. Three Egyptian bauxites , Australian industry used buaxite and its red mud tailing were analyzed by a high resolution gamma spectrometer, with a hyper pure germanium detector. The three Egyptian bauxites show high concentrations in uranium series, and around 120 Bq kg^{-1} for uranium –235. K-40 concentrations for these samples ranged from 289 to 575 Bq kg^{-1}. Thorium series concentrations show lower values. The industrially used bauxite shows very low concentrations for all radioactive nuclides. Its tailing red mud as a low level radioactive waste LLRW, shows low concentrations for uranium – series, thorium – series and also ^{40}K, so it is recommended to be used in industrial and agricultural purposes, which is not permissible for the normal red mud.

Introduction

Laterites vary widely in composition, but most contain aluminum hydroxides and iron hydroxides and oxides, mixed with a little residual quartz. A rare variety called bauxite is almost pure hydrous aluminum oxide Al_2O_3 nH_2O, and hence valuable as an ore of aluminum production. [1]. Uranium and thorium are present in bauxite mineral as an accessory, which means that it present in small amounts. Also bauxites contain uranium and thorium in appreciable amounts according to their genesis by weathering and sedimentation [1,2].

For sedimentary rocks as bauxite ores many geochemical processes occur which cause isotopic and elemental fractionation, which produce a state of disequilibrium between parent and daughter nuclides in the series. [3]. Measurements of concentrations of specific radionuclides, and the activity ratios of uranium and thorium daughters, give geochemists knowledge to study the origin and the history of these deposits. [3].

Red mud is produced in huge amounts as a waste product in the production of aluminium from bauxite by the Bayer's method. It may be used as a raw material in tile production or in the heavy clay industry. Also it can be used for cesium adsorption from the contaminated agricultural lands or contaminated water.[4] Red mud as a naturally occurring radioactive material, (NORM) waste, produced in such great amounts has to be considered with particular care, from the environmental and regulatory view points. Health physicists have concern about the exposure from NORM despite of their low to medium specific activity owing to the large quantities produced [5].

In this work three different Egyptian bauxite samples, from Abu Thor Trench , Sinai, and an industrially Australian , used bauxite and its tailing red mud where analyzed by a high resolution gamma spectrometer with high purity germanium detector (HpGe). Radioactivity concentrations of different nuclides in the uranium and thorium series were measured, also the ^{40}K concentrations in the three Egyptian bauxites, the imported industrially used bauxite and its tailing red mud were measured in Bq kg^{-1}.

Samples and Experimental Setup

The Egyptian bauxites were provided from three different shafts in Abu Thor Trenches, Sinai. The industrially used bauxite and its tailing red mud were provided from the Egyptian Aluminum Company in Naga Hammadi, Egypt. The industrially used bauxite is a mixture of many Australian bauxites. For technical and industrial reasons they were mixed and chemically processed before aluminum production. So the red mud tailing can not be specified to certain bauxite.

Samples were crashed then sieved by 0.8 x 0.8 mm sieve and homogenized , mixed and transferred to a 750 ml or a 500 ml capacity Marinelli, beakers for gamma measurements. Each sample was carefully weighed.

Gamma spectrum for each sample were measured for 2 – to – 24 hours, by a high resolution gamma spectrometer, based on a hyper pure germanium coaxial detector from EG & G Ortec. The Hp Ge detector has a FWHM of 1.9 keV at the1.33 MeV ^{60}Co gamma energy, and relative efficiency of about 50%. A lead cylindrical shield with fixed bottom and moving cover, and a concentric copper cylinder was used to decrease the gamma background.

Efficiency calibration for the Hp Ge gamma spectrometer system were carried out by a mixed radioactive source (^{109}Cd, ^{137}Cs, ^{57}Co, ^{123m}Te, ^{113}Sn, ^{88}Y and ^{60}Co) in a model 138 G Marinelli beaker 1000 ml capacity. Also a ^{226}Ra point source was used to obtain broader energy range. The relative efficiency curve was normalized to the used 750 ml and 500 ml capacity Marinelli beakers by three different concentrations of a chemically pure potassium chloride solutions in distilled water.

K-40, ^{238}U, ^{226}Ra series, ^{232}Th series and ^{235}U concentrations for the three Egyptian bauxite samples, the industrially used imported Australian bauxite and its tailing red mud were measured in Bq kg^{-1} for each sample for each nuclei.

For the ^{238}uranium series gamma energies of 63.3 keV (3.8%)^{234}Th 186.1 keV (3.3%) ^{226}Ra (after the subtraction of the 185.7 keV 53% of ^{235}U), the 295.1 keV (19.2%) , and 352.0 keV (37.1%) ^{214}Pb, and 609.3 keV (46.1%), 768.4 keV (5%), 934.0 keV (3.4%) and 1120.3 keV (15%) ^{214}Bi, were taken to determine the concentrations of the assigned nuclides in the series [6].

For the ^{232}Th series gamma energies of 463.1 keV (4.6%) , 911.2 keV (29%) and 966.0 keV (2.3%) ^{228}Ac, 727.3 keV (6.7%), 1620.7 keV (1.5%), ^{212}Bi and the 583.0 keV (30.9%) ^{208}Tl were taken to represent the concentrations of the assigned nuclides in the series [6].

For the actinium series gamma energies of 143.8 keV (10.5%), 163.4 keV (4.7%) and 205.3 keV (4.7%) were taken to represent the ^{235}U concentrations [6].

The 1460.8 keV (10.7%) gamma energy was taken to determine the ^{40}K concentration. [6].

Table I : **Activity concentration for different nuclides in (Bqkg^{-1}).**

Nuclide	Concentration (Bqkg –1)				
	Sample(1)	Sample(2)	Sample(3)	Sample(4)	Red mud
Uranium series					
Th-234	1814±165	8415± 79	8089± 82.3		
Ra-226*	2576± 7	3059±7	2830± 9	3.5 ± 0.6	10.3± 0.5
Pb-214	2340± 5	2432± 2.5	1692± 2.5	0.8 ± 0.1	3.5 ±0.01
Bi-214	2005± 5	2059± 5	1426± 4	0.9 ± 0.1	4 ± 0.2
Thorium series					
Ac-228	107.3±4	35 ± 1	55.6 ± 1.2		5.7 ± 0.2
Bi-212	328 ± 2	43.3± 3	71.3 ± 3.3	0.8 ± 0.34	4.6 ± 0.3
Tl-208	17.9 ± 1			0.7 ± 0.06	4.1 ± 0.1
Actinium Series					
U-235	132 ± 2	119± 2.2	120 ± 3		
K-40	289 ± 4	346± 5	575 ± 6	6.1 ± 0.35	9.3 ± 0.4

* ^{226}Ra concentration was calculated after the subtraction of the 185.7 keV(53%) gamma of ^{235}U

- sample 1, Abu Thor Trench 1.1
- sample 2, Abu Thor Trench 1.2
- sample 3, Abu Thor Trench 2
- sample 4, the industrially used bauxite.

Table II: **The activity concentrations for ^{226} Ra, ^{232} Th , and ^{40} K in Bqkg^{-1} and the radium equivalent for red mud compared to that obtained by Beretka et al[9]**

	Ra - 226	Th - 232	K - 40	Ra eq.
Present work	10.3	5.7	9.3	19.2
Beretka et a1[9]	326.6	1128.0	29.6	1942.5

Table1. represents the concentrations in Bq kg^{-1} for three bauxite ores taken from Sinai and an industrially used unspecified Australian bauxite and its tailing red mud. Activities of individual nuclides in the ^{238}U – ^{226}Ra series, ^{232}Th series, ^{40}K and ^{235}U are shown in Bq kg^{-1}. The concentrations in the industrially used bauxite and its tailing red mud are very low compared with the three Egyptian ore samples due to the chemical processing for the mixed bauxites.

For the three bauxite samples a state of disequilibrium in both the ^{238}U-series and the ^{232}Th series is clear. [3,7 & 8] . Ivanovich & Harmon, [1992] , Ivanovich [1991] and Philipsborn & Kuhnast [1992] studied in details uranium series and thorium series disequilibrium, which can be explained by :

1) Due to weathering followed by sedimentation, most surface and near surface nuclides in the radioactive series, geological environments are subject to migration of nuclides.

2) Radon nuclides are intermediate product nuclides in the three natural series, which may diffuse out as the system is not closed. So radioactive disequilibrium induced between nuclides below radon in the decay series.

3) The chemical state and the chemical environment of each naturally occurring isotope of uranium (^{238}U , ^{234}U, and ^{235}U), thorium isotopes (^{234}Th, ^{232}Th, ^{230}Th and ^{228}Th), and radium isotopes (^{228}Ra, ^{226}Ra, ^{224}Ra and ^{223}Ra) can be regarded as geochemical analogues of divalent radionuclides and multivalent actinides [8] .

4) Mobility of each radionuclei depends on the chemical parameters as solubility, adsorption, precipitation, complexation and physical impulses generated by alpha and beta radioactive decay.

5) Uranium and thorium isotopes are often associated with other elements and metals as iron, aluminum , copper and manganese, which affect the organic and inorganic groundwater collision . This indicates that iron, manganese oxyhydroxied either as coatings or inorganic particles or in association with humic fulvic acides are binding heavier metals such as the actinides into stable transportable colloidal complexes. [8].

The activity concentrations of red mud and the radium equivalent present in this work is very low compared with the results obtained by [3,9], which is shown in table II.

Concentrations of natural radioactivity in the present work for red mud are very much lower than that of the exempt activity concentrations, for the most relevant radionuclides plus their progeny as a NORM waste product, which established by the Basic Safety Standards (IAEA, 1994) [5].

As a conclusion about red mud produced from the Egyptian Aluminum industry, red mud will not cause any radiological risks when used for the production of tile,or heavy clay industry, also, with this low concentrations of radionuclides naturally occurred, it can be used for the removal of radiocesium from the contaminated water [4].

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