

Natural Radioactivity Levels in Environmental Samples (Iron and Copper) in the Arabian Shield, the Western Part of Saudi Arabia

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Abstract: The Arabian shield, in the western part of Saudi Arabia has significant iron and copper ore deposits, they have promising economic potential with their reserves. Nine rock samples were collected from different areas of it and prepared for analysis. XRD was applied to determine the mineral composition of the samples, which revealed that the major minerals are QUARTZ (mostly in all samples), MAGNETITE, HEMATITE, CORUNDUM, GOETHITE, MONTMORILLONITE, PYRITE, ANKERITE, BOEHMITE, SPINEL and ALBITE, with additional minor and trace minerals. Samples were analyzed for concentrations of ^{238}U , ^{232}Th , and ^{40}K by the gamma spectrometer based on hyper pure germanium detector "HPGe" crystal. For ^{238}U concentrations, the values in Bq/kg dry weight ranged from 152.68 to 264.73 (for iron ore), 156.37 and 329.98 (for copper ore). For ^{226}Ra , the activities of ^{214}Pb and ^{214}Bi in equilibrium with parent (^{226}Ra) were used to calculate the concentrations in Bq/kg dry weight, the average values ranged from 2.50 to 386.30 (for iron ore), 57.41 and 1048.01 (for copper ore). While the activities of ^{232}Th series were calculated from daughters ^{228}Ac , ^{212}Bi , and ^{208}Tl , the average concentrations in Bq/kg dry weight ranged from 1.50 to 183.90 (for iron ore), 43.66 and 44.41 (for copper ore). ^{40}K concentration values in Bq/kg dry weight ranged from 2.70 to 186.99 (for iron ore), 48.92 and 191.33 (for copper ore), and the ^{235}U concentrations in Bq/kg dry weight ranged from 8.35 to 13.70 (for iron ore), ND and 18.37 (for copper ore). the Ra_{eq} Bq/kg dry weight which ranged from 10.88 to 333.59 (for samples 1,2,3, 6,7,9), they are less than 370 the permissible value adopted by EPA and UNSCEAR (2000), while for samples 4, 5 were 610.63 and 662.56 which they are higher than the value 370, for sample 8 (Jabal Sayid) has the highest value 1114.21 (Bq/kg).

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1. Introduction

Saudi Arabia has a significant iron ore (Wadi Sawawin and Wadi Fatima) and copper ore (Jabal Sayid) deposits. These locations in the west central Arabian Shield in the Kingdom have promising economic potential with their reserves, as shown by several studies. Hematite (iron ore) from Wadi Fatima was analyzed by instrumental neutron activation analysis using ^{59}Fe ($^{241}\text{Am} - \text{Be}$) neutron irradiation facility. More than 35 gamma-ray lines were identified as well as the concentration of Mn in hematite was obtained to be about 0.332 (Hassan *et al.*, 1994). Chemical analysis of the ore shows that it has an average content of 41.27% iron (corresponding to about 58.96% hematite). X-ray analysis shows that the ore contains goethite in addition to hematite, The upgrading process consists of several steps to raise the hematite content of the ore to about 87.11 weight percent (Manieh, 1986).

Measuring the activity concentrations due to ^{226}Ra and ^{232}Th for hematite samples from two different locations (Abu Aggag and Um Gereifat) areas in the Eastern Desert of Egypt. The obtained results indicated that; samples from Um Gereifat area

have activity concentrations higher than that of Abu Aggag area (Ahmed *et al.*, 2007).

The Palaeoproterozoic Murphy Inlier is situated at the southern end of the McArthur Basin in Northern Australia. The inlier contains over uranium, copper, tin and base metal occurrences (Mernagh, 2011).

The ironstones of Wadi Al shemysi are mainly enclosed within the middle part of the fluvio-lacustrine siliciclastic succession which is consisted mainly of conglomerates, sandstones, siltstones, muddy, sandy and glauconitic ironstones, fresh water carbonates, tuffaceous mudstone and basalts in descending order. The most important diagenetic process in ironstone is the dehydration and recrystallization of the amorphous Fe-clays and formation of goethite and hematite cement (Mesaed *et al.*, 2012). The trace element contents in two copper minerals (brochantite and native copper) were determined using k_0 -NAA before and after quantitative removal of copper by electrolysis. This work confirmed that the content of some trace elements (Na, K, Rb, Cs, Sb, Pt and Zn) was higher after Cu removal in chalcopyrite (CuFeS_2) and chalcantite ($\text{CuSO}_4 \cdot 5\text{H}_2\text{O}$) (Taseska *et al.*, 2012).

This study is aimed to determine the activity concentrations of naturally levels radionuclides in iron and copper ore samples that have been collected from different locations of the Arabian shield. The activity concentrations of ²³⁸U and ²²⁶Ra have been inferred from gamma-ray transitions associated with their decay progenies and measured using a hyper-pure germanium detector. Details of the samples' preparation are presented, the values of the activity concentrations and ²²⁶Ra equivalent are measured.

Geological setting

Many types of mineral deposits in Saudi Arabia are wide spread, the bulk of metallic mineral resources are contained in Precambrian rocks of the Arabian shield, in the western part of the country. For example deposits of base metals such as iron and copper. The largest known iron ore deposit in the Kingdom is Wadi Assawawin (extends over a belt measuring 15-20 by 25 km SW of Tabuk). This deposit holds reserves of 84 million tons at 42.5 % iron. Another iron ore deposits Wadi Fatima, which

is the largest NE-SW trending low topographic area in the west central Arabian Shield, Saudi Arabia (reserves 48.4 million tons at 45-48% iron). Also, Jabal Idsas (220 km SW of Riyadh) is an important iron ore deposit with reserves 105 million tons of 15 - 20% iron. The copper mineralization is the broader metals prevalent in rock belonging to the pre-Cambrian periods of Arabian Shield. Many reservoirs copper have been explored, with large reserves of copper metal. Jabal Sayid, (copper mine located at 315 km NE of Jeddah, about 40 km north of Mahd Althahab Mine) is the most important one discovered in the Kingdom, The mine under the surface contains proven reserves of up to 99 million tons of copper ore Of 2.68% copper metal. Also, there are many copper ore deposits such as Mibari at Alqassim (Saudi Geological Survey,2012).

Samples and measurements

Nine rock samples were collected from different areas of the Arabian shield, in the western part of the Saudi Arabia as shown in figure 1.



- ▲ Jabal Idsas-Alreyad (23^o.20, 45^o.10)
- ▲ Wadi Fadima-Makka (21^o.30 , 39^o.25).
- ▲ Alzobiera–Hail (25^o.80, 42^o.30).
- ▲ Wadi Sawawin –Tabouk (27^o.90, 35^o.40).
- ▲ Jabal Sayid- Al Madienah (23^o.90, 40^o.90).
- ▲ Mibari – Al Qassim (25^o.80,42^o.30).

Fig. 1 Map of the samples' locations

Table 1 shows the type of sample, its code, and sampling Location.

Table (1): Type of sample, its code and sampling Location.

Ore Name	Sample No.	Location (Lat. And Long.)
Iron Ore	1	Jabal Idsas-Alreyad (23 ^o .20 , 45 ^o .10)
	2	Wadi AsSawawin –Tabouk (27 ^o .90 , 35 ^o .40)
	3	Wadi AsSawawin –Tabouk (27 ^o .90 , 35 ^o .40)
	4	Wadi Fadima- east of Jeddah(Al shemysi) (21 ^o .30 , 39 ^o .25)
	5	Wadi Fadima- 45 km of Makkah) (20 ^o .30 , 39 ^o .25)
	6	Wadi Fadima- 45 km of Makkah

Copper Ore	7	(20 ^o .30 , 39 ^o .25) Alzobiera–Hail (25 ^o .80 , 42 ^o .30)
	8	Jabal Sayid- Al Madienah (23 ^o .90 , 40 ^o .90)
	9	Mibari – Al Qassim (25 ^o .80 , 42 ^o .30)

Samples were grounded, sieved by 1mm x 1mm, then dried to 95°C for 24 hours in order not to lose the volatile polonium or cesium. The dried fine grained samples were packed in polyethylene Marinelli beakers for gamma spectroscopy, and then stored for up to four months to reach secular equilibrium between ²³⁸U and ²³²Th and their progenies.

Ten grams of the dried sample were analyzed by XRD for the mineral constituents. Samples were analyzed for concentrations of U-238, Th-232 series and K-40 using the gamma spectrometer based on high pure germanium detector "HPGe" with relative efficiency of 20%, and FWHM 4.2 keV at 1461 keV, the measurements were done to twenty four hours. After analyzing the spectrum, count rates for each detected photopeak were used to calculate the specific activity (A) for each detected nuclide using the following equation:(Amrani and Tahtat,2001).

$$A = \frac{c}{M \beta \epsilon} \quad (1)$$

Where: c is the net counting rate of a specific gamma ray (count per second)

M is the mass of the samples (kg.)

β is the transition probability of gamma-decay energy.

ϵ is the detector efficiency at the specific gamma-ray energy.

Table 2 represents isotopes and photopeak energies used for gamma-ray measurements of ^{238}U decay series, ^{232}Th decay series, ^{235}U , and ^{40}K .

Table(2):Isotopes and photopeak energies used for gamma-ray measurements of ^{238}U , ^{226}Ra , ^{232}Th , ^{235}U and ^{40}K .

U-238, Ra-226 series		Th-232 series		U-35	K-40
Isotopes	E(keV)	Isotopes	E(keV)	E(keV)	EkeV
^{234}Th	63.29	^{228}Ac	338.42	143.80	1460.8
^{234}Th	92.78+ 92.35	^{228}Ac	911.16	185.70	
$^{234\text{m}}\text{p-}$	766.60	^{228}Ac	968.97		
$^{234\text{m}}\text{pa}$	1001.00	^{212}Bi	727.25		
$^{214}\text{Pb-}$	295.09	^{212}Bi	785.51		
^{214}Pb	351.87	^{208}Tl	583.10		
$^{214}\text{Bi-}$	609.31	^{208}Tl	860.40		
$^{214}\text{Bi-}$	1120.27	^{208}Tl	2614.5		
^{214}Bi	1764.49				

3. Results and Discussions

Table 3 gives the X-RD results, which revealed the major, minor and trace minerals. Results show that the major minerals are QUARTZ(mostly in all samples), MAGNETITE (Fe_2O_4 with 70% iron), HEMATITE(Fe_2O_3 with 70% iron), CORUNDUM, GOETHITE, MONTMORILLONITE, PYRITE, ANKERITE, BOEHMITE, SPINL, and ALBITE, with additional minor and trace minerals.

Table 4 represents the specific activity concentrations in Bq/kg dry weight for iron and copper ore samples.

There is disequilibrium in the ^{238}U - ^{226}Ra series, so for ^{238}U a 63.29KeV photopeak, which comes the decay of ^{234}Th , was used to find the concentrations in Bq/kg dry weight, the values ranged from 152.68 to 264.73 (for iron ore), 156.37and 329.98(for copper ore).

For ^{226}Ra , the activities of ^{214}Pb and ^{214}Bi in equilibrium with parent (^{226}Ra) were used to calculate the concentrations in Bq/kg dry weight, the average values ranged from 2.50 to 386.30 (for iron ore),57.41and 1048.01(for copper ore).

While the activities of ^{232}Th series were calculated from daughters ^{228}Ac , ^{212}Bi , and ^{208}Tl , the average concentrations in Bq/kg dry weight ranged from 1.50 to 183.90(for iron ore), 43.66 and 44.41(for copper ore).

^{40}K concentration values in Bq/kg dry weight ranged from 2.70 to 186.99 (for iron ore), 48.92 and 191.33 (for copper ore), and the ^{235}U concentrations in Bq/kg dry weight ranged from 8.35 to 13.70 (for iron ore), ND and 18.37 (for copper ore).

It is noticed that, copper – bearing samples (4,5,8) have ^{226}Ra much higher than ^{238}U (Tables 2,4) which means the leaching out of uranium during the processes of alterations. Samples with iron minerals show high uranium than radium, and these represent the disequilibrium in the ^{238}U -series (Table 4). Thorium was found to vary with variation of rock types and the same behavior of ^{40}K .

Exposure to radiation has been defined in terms of the radium equivalent Ra_{eq} which is calculated from equation (1) Tufail *et al.* (2006):

$$\text{Ra}_{\text{eq}} = A_{\text{Ra}} + (A_{\text{Th}} \times 1.43) + (A_{\text{K}} \times 0.077) \quad (2)$$

Where:

A_{Ra} , A_{Th} and A_{K} are concentrations Bq/kg for radium, thorium and potassium Bq/kg dry weight, respectively.

Table (5) represents the values of the Ra_{eq} Bq/kg dry weight which ranged from 10.88 to 333.59(for samples 1,2,3, 6,7,9), they are less than 370 the permissible value adopted by EPA and UNSCEAR (2000),while for samples 4, 5 were 610.63 and 662.56 which are higher than the value 370, for sample 8 (Jabal Sayid) has the highest value1114.21(Bq/kg) because it lies at 40 km of Mahd Althahab known with high radiation level (Saudi Geological Survey, 2012).

Table (3): The mineral constituents of iron and copper ore samples analyzed by XRD spectrometer (Leet *et al.*, 1982, and Mineral Data, 2012)

Sam.No.	Major	Minor	Trace
1	MAGNETITE($\text{Fe}^{3+}_2\text{Fe}^{2+}\text{O}_4$)	-----	GREENALITE ($(\text{Fe}^{3+}_3\text{Fe}^{3+})_2\text{Si}_2\text{O}_5(\text{OH})_4$)
2	QUARTZ(SiO_2) HEMATITE($\text{Fe}^{3+}_2\text{O}_3$)	CALCITE(CaCO_3)	-----
3	BOEHMITE ($\text{AlO}(\text{OH})$) HEMATITE($\text{Fe}^{3+}_2\text{O}_3$) GIBBSITE ($\text{Al}(\text{OH})_3$)	ANATASE (TiO_2)	-----
4	HEMATITE($\text{Fe}^{3+}_2\text{O}_3$) CORUNDUM (Al_2O_3) QUARTZ(SiO_2)	PERICLASE (MgO) LIME (CaO) KAOLINITE ($\text{Al}_2\text{Si}_2\text{O}_5(\text{OH})_4$) ILLITE($(\text{K}_2\text{H}_3\text{O})(\text{Al},\text{Mg},\text{Fe})_2(\text{Si},\text{Al})_4\text{O}_{10}(\text{OH})_{22}(\text{H}_2\text{O})$)	MANGANITE($\text{MnO}(\text{OH})$) ANATASE(TiO_2), ORSCHALLITE ($\text{Ca}_3(\text{SO}_3)_2(\text{SO}_4)\cdot 12(\text{H}_2\text{O})$)
5	GOETHITE ($\text{Fe}^{3+}\text{O}(\text{OH})$) MONTMORILLONITE ($\text{NaCaAl}_2\text{Si}_4\text{O}_{10}(\text{OH})_2(\text{H}_2\text{O})_{10}$) QUARTZ(SiO_2)	BEAVERITE($(\text{PbCu}^{2+}(\text{Fe}^{3+}\text{Al})_2(\text{SO}_4)_2(\text{OH})_6$) PARTHEITE ($\text{Ca}_2\text{Al}_4\text{Si}_4\text{O}_{15}(\text{OH})_2\cdot 4(\text{H}_2\text{O})$) BENAUITE($(\text{SrBaPbFe}^{3+}(\text{PO}_4)_2(\text{SO}_4)(\text{OH})_6$) SCHNEIDERHOEHNITE ($\text{Fe}^{2+}\text{Fe}^{3+}_3\text{As}_5\text{O}_{13}$) IRON (Fe) CHESTERITE ($(\text{MgFe}^{2+})_{17}\text{Si}_{20}\text{O}_{54}(\text{OH})_6$) MICROCLINE (KAlSi_3O_8) MARCASITE(Fe^{2+}S_2) PYRRHOTITE (Fe^{2+}S)	CHLORITE($(\text{MgFe}^{2+})_6(\text{AlFe}^{3+})\text{Si}_3\text{O}_{10}(\text{OH})_8$)
6	GOETHITE ($\text{Fe}^{3+}\text{O}(\text{OH})$) SPINEL (MgAl_2O_4) QUARTZ(SiO_2)	BEAVERITE ($\text{PbCu}^{2+}(\text{Fe}^{3+}\text{Al})_2(\text{SO}_4)_2(\text{OH})_6$) PARTHEITE ($\text{SrBaPbFe}^{3+}(\text{PO}_4)_2(\text{SO}_4)(\text{OH})_6$) BENAUITE ($\text{SrBaPbFe}^{3+}(\text{PO}_4)_2(\text{SO}_4)(\text{OH})_6$) SCHNEIDERHOEHNITE ($\text{Fe}^{2+}\text{Fe}^{3+}_3\text{As}_5\text{O}_{13}$) IRON (Fe) CHESTERITE ($(\text{MgFe}^{2+})_{17}\text{Si}_{20}\text{O}_{54}(\text{OH})_6$) MICROCLINE (KAlSi_3O_8) MARCASITE (Fe^{2+}S_2) PYRRHOTITE (Fe^{2+}S)	-----
7	QUARTZ(SiO_2) HEMATITE($\text{Fe}^{3+}_2\text{O}_3$)	CALCITE (CaCO_3)	-----
8	PYRITE (Fe^{2+}S_2) ANKERITE($\text{CaFe}^{2+}\text{MgMn}^{2+}(\text{CO}_3)_2$) QUARTZ(SiO_2)	CHABAZITE ($\text{CaNaKMgSrAlSiO}_{24}(\text{H}_2\text{O})$) CLINOCHLORE ($\text{MgFe}^{2+}\text{Si}_3\text{Al}_2\text{O}_{10}(\text{OH})_8$) CHALCOPYRITE ($\text{CuFe}^{2+}\text{S}_2$)	BIOTITE($(\text{K}(\text{MgFe}^{2+})_3\text{AlSi}_3\text{O}_{10}(\text{OH})_2$) HYDROTALCITE, MAGNETITE($\text{Fe}^{3+}_2\text{Fe}^{2+}\text{O}_4$) ROGGIANITE ($\text{Ca}_2\text{Be}(\text{OH})_2\text{Al}_2(\text{Si}_4\text{O}_{13})\cdot 2.4(\text{H}_2\text{O})$) NONTRONITE ($\text{NaFe}^{3+}\text{Si}_3\text{AlO}_{10}(\text{OH})_2\cdot 4(\text{H}_2\text{O})$) ARSENOPYRITE(Fe^{3+}AsS)
9	QUARTZ(SiO_2) ALBITE ($\text{Na}(\text{AlSi}_3\text{O}_8)$)	CLINOCHLORE ($\text{MgFe}^{2+}\text{Si}_3\text{Al}_2\text{O}_{10}(\text{OH})_8$) MICROCLINE (KAlSi_3O_8)	BIOTITE ($(\text{K}(\text{MgFe}^{2+})_3\text{AlSi}_3\text{O}_{10}(\text{OH})_2$) MAGNETITE($\text{Fe}^{3+}_2\text{Fe}^{2+}\text{O}_4$) ZIRCON(ZrSiO_4) SAPONITE($(\text{Ca}(\text{Mg},\text{Fe})_3((\text{Si},\text{Al})_4\text{O}_{10}(\text{OH})_2(\text{H}_2\text{O}))$) KOVDOORSKITE($(\text{Mg}_5(\text{PO}_4)_2(\text{CO}_3)(\text{OH})_2\cdot 4.5(\text{H}_2\text{O}))$)

Table (4): The specific activity concentrations in Bq/kg dry weight for iron and copper ore samples.**N D: not detected**

Sample Code	Sample 1	Sample2	Sample3	Sample4	Sample5	Sample6	Sample7	Sample8	Sample9
U-238 s Series Nuclei	183.46±0.8	167.46±0.77	ND	264.73±0.77	152.68±0.79	ND	ND	329.98±0.77	156.37±0.78
Th-234 Ra-226s Series	9.91±0.04	18.72±0.08	69.40±1.20	367.89±0.05	388.62±0.04	53.10±.90	LDL	1032.49±0.03	55.74±0.04
Pb-214	8.51±0.23	18.80±0.12	70.80 1.02	365.09±0.11	383.97±0.06	57.500.40	2.5 0±0.20	1064.65±0.81104	59.07±0.11
Bf-214 Average	9.21±0.24	18.76±0.10	70.10±1.11	366.49±0.22	386.300.05	55.30±0.65	2.5 0±0.20	8.0±0.42	57.41±0.08
Th-232 Series	1.04±0.11	14.24±0.11	181.20 ±2.00	167.65±0.14	184.47±0.12	27.50±1.00	6.9 0±1.20	ND	42.95±0.15
Ac-228	ND	14.86±0.34	184.70 ±6.00	164.61±0.71	181.76±0.71	28.20 ±2.90	LDL	ND	45.73±0.73
Bf-212	1.07±0.07	15.82±0.06	185.80 ±5.60	173.50±0.05	ND	30.40 ±2.60	4.30 ±0.90	41.57±0.05	44.54±0.08
TL-208 Average	1.50±0.09	14.97±0.25	183.90 ±3.20	168.7±0.59	183.12±0.41	28.70±1.30	5.60±0.90	43.66±0.11	44.41±0.32
U-235	9.67±0.80	ND	ND	13.70±0.08	8.35±0.09	ND	ND	18.37±0.07	ND
K-40	39.14±0.65	15.22±0.65	6.30 ±0.10	32.61±0.10	186.99±0.08	2.70 ±0.10	4.80 ±0.10	48.92±0.08	191.33±0.06

Table (5) The radium equivalent Bq/kg for the samples.

Ore Name	Sample Code	Ra _{eq} Bk/kg
Iron Ore	1	14.36 ±0.16
	2	41.34±0.34
	3	333.59±1.3
	4	610.36±0.07
	5	662.56±0.06
	6	96.55±2.51
	7	10.88±0.91
Copper Ore	8	1114.21±0.04
	9	135.65±0.03

Conclusion

The X-RD results show that the major minerals are QUARTZ (mostly in all samples), MAGNETITE (Fe₂O₄ with 70% iron), HEMATITE (Fe₂O₃ with 70% iron), CORUNDUM, GOETHITE, MONTMORILLONITE, PYRITE, ANKERITE, and ALBITE. A state of disequilibrium between ²³⁸U and ²²⁶Ra is clear. Most of the analyzed samples have radioactive concentrations within the accepted range which gives radium equivalent less than 370 Bq/kg dry weight for samples 1, 2, 3, 6 and 7, 9 except samples 4, 5, and 8 which have high concentrations. There for, we can conclude that the mining process of iron and copper ore in these area caused enhancement of the exposure from natural radiation.

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