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Natural Activities of ^{238}U, ^{232}Th and ^{40}K in Manganese Ore

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Abstract: Problem statement: Manganese ore is widely used in much industries. Such as ore contain natural radioactive nuclides at various concentrations. If this ore contain high concentrations of natural radioactive nuclides, workers handling them might be exposed to significant levels of radiation. Therefore it is important to determine the radioactive nuclides in this ore. **Approach:** The natural radionuclides (^{238}U, ^{232}Th and ^{40}K) contents of Manganese ore samples collected by Siani Manganese Company in Egypt-Cairo have been determined by low background spectroscopy using hyper-pure germanium (HPGe) detector. **Results:** The mean activities due to the three radionuclides (^{238}U, ^{232}Th and ^{40}K) were found to be 3543±106, 222±6.6 and 3483±104 $Bqkg^{-1}$, respectively. The absorbed dose rate due to the natural radioactivity in samples under investigation ranged from 1522±45-1796±53 $nGyh^{-1}$. The radium equivalent activity varied from 3807±114-4446±133 $Bqkg^{-1}$. The representative external hazard index values for the corresponding samples are also estimated. **Conclusion:** The results of this assessment obtained by the gamma-ray spectroscopic analysis, have indicated that the levels of natural radioactivity were higher than the international recommended limits.

Key words: ^{238}U; ^{232}Th and ^{40}K estimation in manganese ore, manganese ore, radium equivalent

INTRODUCTION

Radionuclides are present always in the natural environment. The main natural contributors to external exposure from gamma-radiation are uranium and thorium series together with potassium (Luigi *et al*., 2000). Natural radiation is usually classified as either cosmic or terrestrial radiation (El-Zakla *et al*., 2007). Large variations in dose rates of both cosmic and terrestrial radiation are found depending on where the measurements are made (Shenber, 1997). Measurements of natural radioactivity in environmental samples, specially in raw materials produced by mining are very important to determine the amount of change of the natural background activity with time as a result of any radioactive release (Iwaoka *et al*., 2009). Emanation of radon (^{222}Rn), for example is associated with the presence of radium and its ultimate precursor uranium in the ground (Bossew and Lettner, 2007). The inhalation of its short-lived daughter produces is a major contributor to the total radiation dose to exposed subjects (UNSCEAR, 1993). Many studies have investigated the radioactive elements in different ore samples (Chau *et al*., 2008), however few studies have investigated the radioactive content of manganese (Mn) ore.

Manganese is an essential trace element in the metabolism of all living organisms, animals or plants. Normally it is found in human blood with concentration <320 nmol L^{-1} and functions as a cofactor for some enzymes. Exposure of man to high levels of manganese leads to hypermanganesaemia (high Mn levels in blood) and defect in its metabolism with its accumulation in the liver and the basal ganglia is lethal (Tuschl *et al*., 2008). Also, manganese intoxication has been described in children on long term parenteral nutrition presenting with liver and nervous system disorders (Kafritsa *et al*., 1998). In adults, together with occasional oral intake and product contamination with the element can lead to brain accumulation and neurotoxicity (Hardy *et al*., 2008).

Manganese exposure is usually via inhalation (the risk varying with the manganese species involved and with particle size). There are specific measures to protect those working in manganese-related industry (or mining) such as reducing exposure levels and time of exposure and the use of exhaust ventilation (Kavasi *et al*., 2009). In addition to the risk of exposure to high doses, manganese provides another risk factor if the ore contains residual radioactive elements. This arises the our interest to investigate the potential of existence of residual radioactive elements in manganese ore, particularly with the increasing demand of using this ore in many industries including the steel industry in Egypt. So the aim of this work is to determine the natural radioactivity in manganese ore produced by one of the local companies working in manganese mining.

MATERIALS AND METHODS

Sample preparation: Ten samples of manganese ore were used in this work. Samples were collected from Om Bogma near Cairo and from Sinai Manganese Egypt. Samples were dried at 105°C to eliminate any traces of water. The representative powdered samples were filled in a polyethylene bottles of 350 cm^3 volume. Each sample was fixed in its container, whose inner diameter was equal to the diameter of the detector in face to face geometry. Finally samples were stored for four weeks to reach the equilibrium state between radium and its decay products.

Detection system setup: The energy and intensity of various gamma-ray lines have been measured using an ORTEC coaxial (HPGe) detector of relative efficiency 50% coupled to 8096 channel analyzer. The Full Width at Half Maximum (FWHM) was found to be 1.9 keV for ^{60}Co-1332 keV gamma-ray line. The detection array was energy calibrated using ^{60}Co (1173.2 and 1332.5 keV), ^{133}Ba (356.1 keV) and ^{137}Cs (661.9 keV). Also, efficiency calibration was made using different energy peaks covering the range up to 2050 keV. An empty bottle with the same geometry was measured for subtracting the background in each measurement. The spectra of all samples were perfectly analyzed using a special PC software program to calculate the concentrations of ^{238}U, ^{232}Th and ^{40}K. Calculations of count rates for each detected radionuclides depend on the establishment of secular equilibrium was reached between ^{238}U and ^{232}Th and for their decay products. Radioactivity concentrations of each sample was measured for about 20 h. Since the detection system gives only the count rate that is proportional to the amount of radioactivity in the samples, the radioactivity concentration in the environmental samples was obtained using the following formula:

$$A = (cps)\ net/\ I\ \varepsilon_{ff}\ m$$

$$(cps)_{net} = (cps)_{sample} - (cps)_{B.G}$$

Where:

- A = The concentration in Bq kg^{-1}
- I = The intensity of gamma-line in a radionuclide
- ε_{ff} = The measured efficiency for each gamma-ray line observed for the same number of channels either for the sample or the background
- m = The mass of the sample in kilograms

^{238}U activity concentration was determined by measuring the 295.1 (19.2%) and 352 (37.1%) keV gamma-rays from ^{214}Pb and the 609.3 (46.1%) and 1120.3 (15%) keV gamma-rays from ^{214}Bi. ^{232}Th activity was determined from the gamma-rays of 238.6 (43.6%) keV from ^{212}Pb and 338.4 (12%), 911.2 (29%) and 969 (17%) keV from ^{228}Ac and 583.0 (86%) keV gamma-rays from ^{208}Tl. ^{40}K concentration was measured from its 1460 (10.7%) keV gamma-ray line.

RESULTS

The obtained spectrum of the background gamma radiation was subtracted from the measured gamma ray spectra of the samples. The characteristic gamma-ray emitters are marked above the corresponding peaks. A selected one of the obtained spectrum for sample number 1 is shown in Fig. 1.

As there is an equilibrium among members of each of the natural radioactive chains, the ^{228}Ac and ^{214}Bi isotopes that emit clear peaks of high intensity from ^{232}Th and ^{238}U series, respectively, have been selected from each chain. Figure 2 shows the activity concentrations of ^{228}Ac, ^{214}Bi in the collected samples.

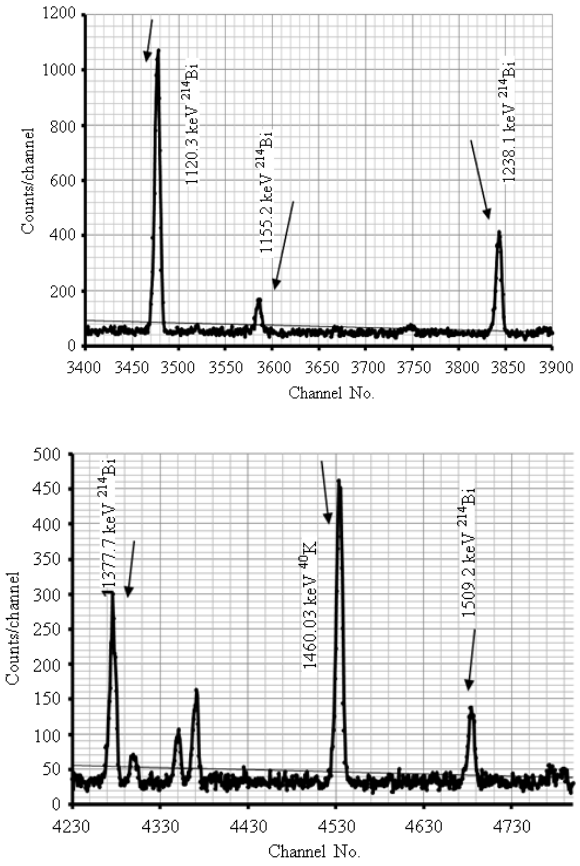

Fig. 1: Portion of Gamma ray spectrum for sample 1

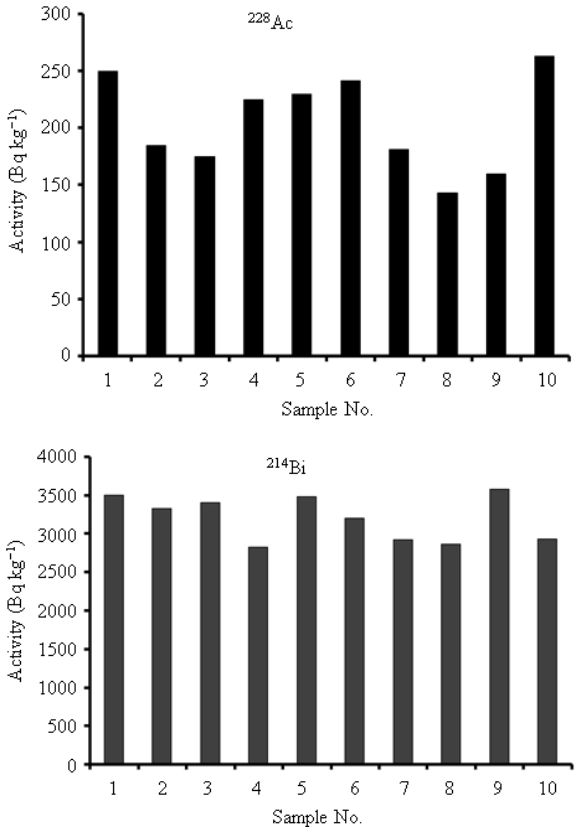

Fig. 2: The activity concentration of ^{228}Ac and ^{214}Bi in the collected samples

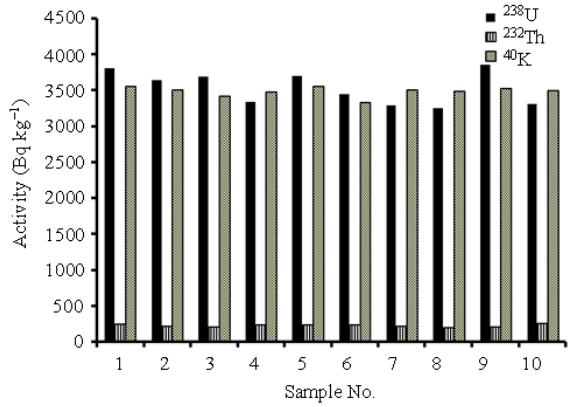

Fig. 3: U, Th and K concentrations in different samples

The activity concentrations for ^{238}U, ^{232}Th and^{40}K are shown in Fig. 3. The activity concentrations of ^{238}U ranged from 3263±97-3819±114 Bqkg^{-1}, with an average of 3543±106 Bqkg^{-1}, ^{232}Th ranged from 193±5.7-247±7.4 Bqkg^{-1}, with an average of 222±6.6 Bqkg^{-1} and ^{40}K ranged from 3330±99-3556±106 Bqkg^{-1}, with an average of 3483±104 Bqkg^{-1}, respectively.

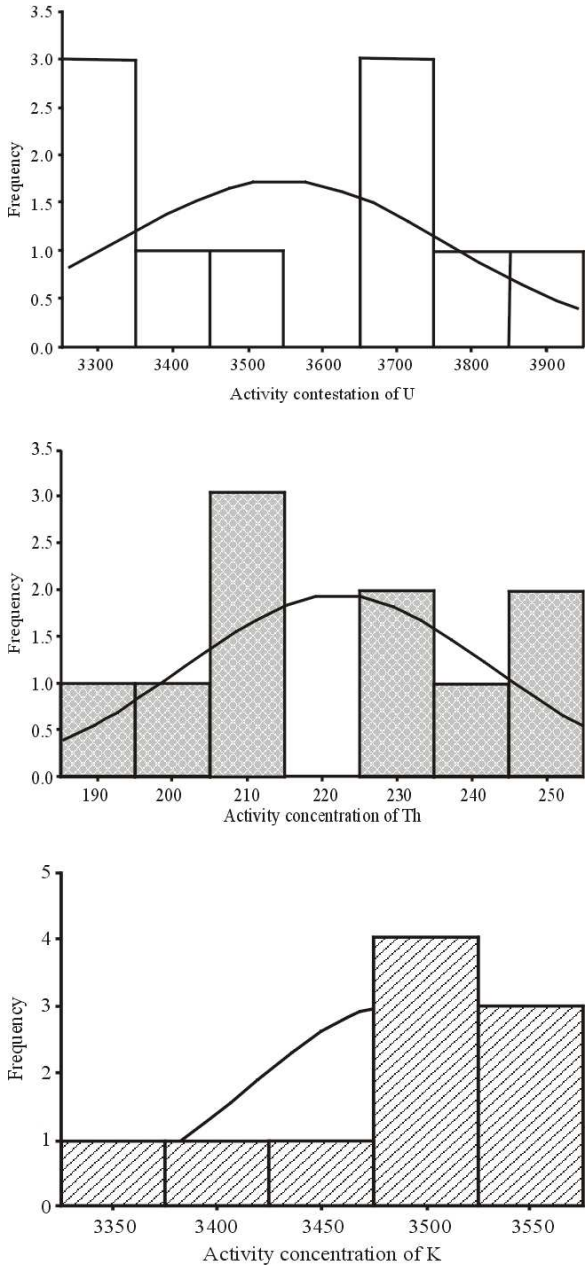

Fig. 4: Frequency distribution of activity concentration Bqkg^{-1} of U, Th and K in the investigated samples

Figure 4 shows the corresponding frequency distribution of activity concentrations of ^{238}U, ^{232}Th, and ^{40}K in the investigated samples. The activity concentrations corresponding to the ^{232}Th and ^{238}U radionuclides analyzed in the investigated samples were fitted to a normal curve. Only ^{40}K shows activity concentration fitted to a log-normal distribution.

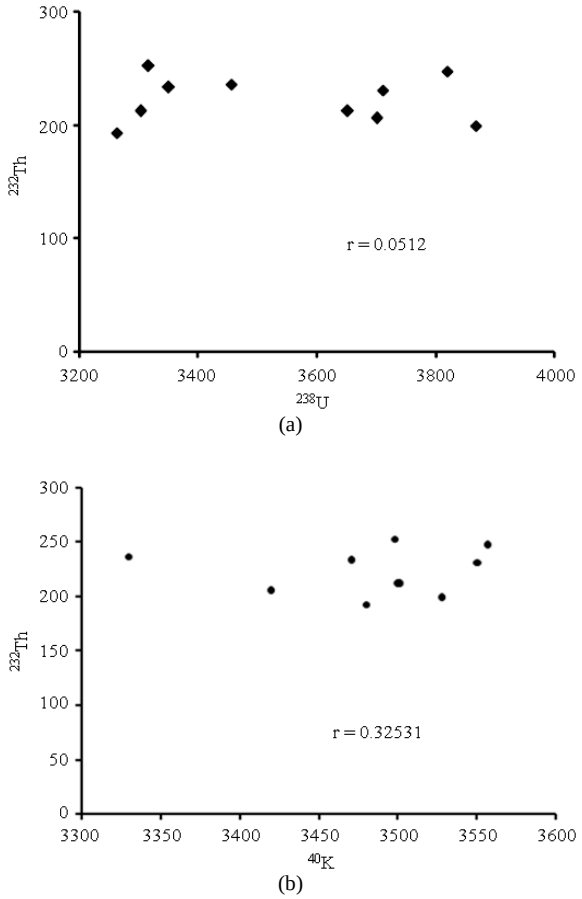

Fig. 5: The correlation between ^{238}U, ^{232}Th (a) and ^{40}K, ^{232}Th (b)

Figure 5a and b indicate a weak correlation between concentrations of ^{238}U, ^{232}Th where r = 0.0512 and concentrations of ^{40}K, ^{232}Th where r = 0.032, respectively in the samples under investigation, which suggests that individual results of either are independent.

DISCUSSION

According to the international manganese institute (UK), manganese is the fourth most used metal in terms of tonnage, being ranked behind iron, aluminum and copper, with in the order of 34 million tons of ore being mined annually. The manganese ore is invested industrially in a long list of industries including as additive in unleaded gasoline, making dry cell battery, steel industry and fertilizer industry. This gives the importance of screening of manganese ore for residual radioactive elements. Naturally, the element is found in combination with other elements.

Table 1: The radium equivalent in Bqkg^{-1}, the external hazard index, the dose rate in nGyh^{-1} and the effective dose in mSvy^{-1} for the studies samples

Samples	Ra_{eq}	H_{ex}	D (nGyh^{-1})	Effective dose (mSvy^{-1})
1	4446±133	12.01±0.35	1796±53	2.02±0.06
2	4225±126	11.40±0.34	1701±51	2.08±0.06
3	4259±127	11.50±0.34	1718±51	2.10±0.06
4	3951±118	10.67±0.31	1586±47	1.94±0.05
5	4314±129	11.64±0.34	1738±52	2.13±0.06
6	4049±121	10.93±0.32	1633±48	2.00±0.05
7	3876±116	10.47±0.31	1552±46	1.90±0.05
8	3807±114	10.27±0.30	1522±45	1.86±0.05
9	4424±132	11.95±0.35	1785±53	2.18±0.06
10	3945±118	10.65±0.31	1584±47	1.94±0.05

To assess the radiological hazard, the radium equivalent activity Ra_{eq}; defined by the estimate that 370 Bqkg^{-1} of ^{226}Ra, 259 Bqkg^{-1} of ^{232}Th and 4810 Bqkg^{-1} of ^{40}K produce the same gamma-ray dose rate can be calculated (Table 1). The Ra_{eq} given by the following equation (Van Dijk and De Jong, 1991):

$$Ra_{eq} = C_u + A\,C_{Th} + B\,C_K$$

where, C_U, C_{Th} and C_K are the activity concentrations of U, Th and K in Bqkg^{-1}, respectively and A; B are constants (El-Arabi, 2007). The highest value of radium equivalent in manganese ore is 4446±133 Bqkg^{-1}. Also, it is observed that the calculated radium equivalent is higher than the recommended maximum value 370 Bqkg^{-1} (OECD, 1997). The dose rates in nGyh^{-1} were calculated using the equation (Beretka and Mathew, 1985):

$$D = R_K C_K + R_U C_U + R_{Th}\,C_{Th}$$

where, R_K; R_U and R_{Th} are constants. As shown in Table 1 that the lowest dose rate was found 1522±45 nGyh^{-1} and the highest dose rates 1796±53 nGyh^{-1} are very high than the international limit (55 nGyh^{-1}) as given by UNSCEAR data. Effective dose rates varied from 1.86-2.20 mSvy^{-1}, with an average value (± SD) of 2.03±0.05 mSvy^{-1}. The external hazard index (Hex) was be calculated from the following criterion:

$$C_u/370 + C_{th}/259 + C_k/4810 < 1$$

where, C_U, C_{Th} and C_K are the concentrations of U, Th and K. The calculated external hazard value was higher than unity, which may cause harm to workers in this field (Freitas and Alencar, 2004).

CONCLUSION

These data serve as a basis for the assessment of radiological hazard to the workers involved in manganese mining, transportation and industrial applications in Egypt. The results of this assessment obtained by the gamma-ray spectroscopic analysis, have indicated that the levels of natural radioactivity were higher than the international recommended limits, where the absorbed gamma dose rates varied from 1522±45-1796±53 $nGyh^{-1}$, with an average value of 1661±49 $nGyh^{-1}$ and the average effective dose rate was 2.03±0.05 $mSvy^{-1}$ in the investigated samples. Therefore, safety rules and precautions should be applied for those working in this field.

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