MONITORING OF NATURAL RADIOACTIVITY IN MANGANESE ORE

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The natural radionuclides ($^{238}\text{U}$, $^{232}\text{Th}$, and $^{40}\text{K}$) contents of Manganese ore collected by Sinai Manganese Company in Egypt-Cairo have been determined by low background spectroscopy using hyper-pure germanium (HPGe) detector. The mean activities due to the three radionuclides ($^{238}\text{U}$, $^{232}\text{Th}$, and $^{40}\text{K}$) were found to be $3543 \pm 106$, $222 \pm 6.6$ and $3483 \pm 104$ Bq/kg, respectively. The absorbed dose rates due to the natural radioactivity in samples under investigation ranged from $1522 \pm 45$ to $1796 \pm 53$ nGy/h. The radium equivalent activity varied from $3807 \pm 114$ to $4446 \pm 133$ Bq/kg. Also, the representative external hazard index values for the corresponding samples were estimated.

**Keywords:** radionuclides, $^{238}\text{U}$, $^{232}\text{Th}$, $^{40}\text{K}$, manganese ore, absorbed dose rates, radium equivalent activity, external hazard index.

**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 [1]. Natural radiation is usually classified as either cosmic or terrestrial radiation [2]. Large variations in dose rates of both cosmic and terrestrial radiation are found depending on where the measurements are made [3]. Measurements of natural radioactivity in environmental samples, especially 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 [4]. Emanation of radon ($^{222}\text{Rn}$), for example is associated with the presence of radium and its ultimate precursor uranium in the ground [5]. The inhalation of its short-lived daughter produces is a major contributor to the total radiation dose to exposed subjects [6]. Many studies have investigated the radioactive elements in different ore samples [7], 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 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 [8]. Also, manganese intoxication has been described in children on long term parenteral nutrition presenting with liver and nervous system disorders [9]. In adults, together with occasional oral intake and product contamination, the element can lead to brain accumulation and neurotoxicity [10]. Manganese exposure usually occurs via inhalation where the risk varies 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. 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 and dry battery industry. 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 by Sinai Manganese Company. Samples were dried at 105 °C to eliminate any traces of water. The representative powdered samples were placed in polyethylene bottles of 350 cm$^3$ volume. Each sample was fixed in its airtight 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 $^{60}\text{Co}$-1332 keV gamma-ray line. The detection array was energy calibrated using $^{60}\text{Co}$ (1173.2 and 1332.5 keV), $^{133}\text{Ba}$ (356.1 keV) and $^{137}\text{Cs}$ (661.9 keV). Also, efficiency calibration was made using different energy peaks covering the range up to 2050 keV. The spectra of
all samples were precisely analyzed using a special PC software program to calculate the concentrations of $^{238}\text{U}$, $^{232}\text{Th}$ and $^{40}\text{K}$. Calculations of count rates for each detected radionuclides depend on the establishment of secular equilibrium reached between $^{238}\text{U}$ and $^{232}\text{Th}$ and for their decay products. Radioactivity concentrations of each sample were measured for about 20 h. Since the detection system gives only the count rate which is proportional to the amount of radioactivity in the samples, the radioactivity concentration in the environmental samples was obtained using the following formula:

$$A = \frac{(\text{cps})_{\text{net}}}{I} \cdot \varepsilon \cdot \frac{m}{10^3},$$

where $A$ - concentration in Bq/kg; $I$ - intensity of gamma-line in a radionuclide, $\varepsilon$ is the measured efficiency for each gamma-ray line observed for the same number of channels either for the sample or the background and $m$ - mass of the sample in kilograms. $^{238}\text{U}$ activity concentration was determined by measuring the 295.1 (19.2 %) and 352 (37.1 %) keV gamma-rays from $^{214}\text{Pb}$ and the 609.3 (46.1 %) and 1120.3 (15 %) keV gamma-rays from $^{214}\text{Bi}$. $^{232}\text{Th}$ activity was determined from the gamma-rays of 238.6 (43.6 %) keV from $^{212}\text{Pb}$ and 338.4 (12 %), 911.2 (29 %) and 969 (17 %) keV from $^{228}\text{Ac}$ and 583.0 (86 %) keV gamma-rays from $^{208}\text{Tl}$. $^{40}\text{K}$ concentration was measured from its 1460 (10.7 %) keV gamma-ray line.

Results and 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. At least 34 million tons of ore being mined annually and invested industrially in a long list of industries. It is used 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 such as iron, silicon, calcium, phosphorus, and other.

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

![Fig. 1. Portion of Gamma ray spectrum for sample 1.](image1)

![Fig. 2. The activity concentrations of $^{228}\text{Ac}$ and $^{214}\text{Bi}$ in the investigated samples.](image2)
As there is 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. Fig. 2 shows the activity concentrations of $^{228}$Ac, $^{214}$Bi in the collected 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 $3263 \pm 97$ to $3819 \pm 114$ Bq/kg, with an average of $3543 \pm 106$ Bq/kg; $^{232}$Th concentrations ranged from $193 \pm 5.7$ to $247 \pm 7.4$ Bq/kg, with an average of $222 \pm 6.6$ Bq/kg; and $^{40}$K concentrations ranged from $3330 \pm 99$ to $3556 \pm 106$ Bq/kg, with an average of $3483 \pm 104$ Bq/kg, respectively.

To assess the radiological hazard, the radium equivalent activity $R_{a_{eq}}$, defined by the estimate that 1 Bq/kg of $^{226}$Ra, 0.7 Bq/kg of $^{232}$Th and 13 Bq/kg of $^{40}$K produce the same gamma-ray dose rate can be calculated (Table). The $R_{a_{eq}}$ is given by the equation [11]

$$R_{a_{eq}} = C_U + A C_{Th} + B C_K,$$

where $C_U$, $C_{Th}$ and $C_K$ - activity concentrations of U, Th and K in Bq/kg, respectively; $A$ and $B$ - constants [12]. The highest value of radium equivalent in manganese ore was $4446 \pm 133$ Bq/kg. Also, it was observed that the calculated radium equivalent was higher than the recommended maximum value 370 Bq/kg [13]. The dose rates in n Gy/h were calculated using the equation [14]

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

where $R_K$, $R_{Th}$ and $R_U$ - constants. As shown in the Table 1 the lowest dose rate was $1522 \pm 45$ nGy/h and the highest dose rates ($1796 \pm 53$) was higher than the international limit (55nGy/h) recommended by UNSCEAR data. Effective dose rates varied from 1.86 to 2.20 mSv/yr, with an average value (±SD) of 2.03 ± 0.05 mSv/yr. The external hazard index (Hex) was calculated from the following formula:

$$H_{ex} = C_U/370 + C_{th}/259 + C_K/4810 < 1,$$

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 [15].

<table>
<thead>
<tr>
<th>Samples</th>
<th>$R_{a_{eq}}$, Bq/kg</th>
<th>$H_{ex}$</th>
<th>$D$, nGy/h</th>
<th>Effective dose, mSv/yr</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>$4446 \pm 133$</td>
<td>$12.01 \pm 0.35$</td>
<td>$1796 \pm 53$</td>
<td>$2.02 \pm 0.06$</td>
</tr>
<tr>
<td>2</td>
<td>$4225 \pm 126$</td>
<td>$11.40 \pm 0.34$</td>
<td>$1701 \pm 51$</td>
<td>$2.08 \pm 0.06$</td>
</tr>
<tr>
<td>3</td>
<td>$4259 \pm 127$</td>
<td>$11.50 \pm 0.34$</td>
<td>$1718 \pm 51$</td>
<td>$2.10 \pm 0.06$</td>
</tr>
<tr>
<td>4</td>
<td>$3951 \pm 118$</td>
<td>$10.67 \pm 0.31$</td>
<td>$1586 \pm 47$</td>
<td>$1.94 \pm 0.05$</td>
</tr>
<tr>
<td>5</td>
<td>$4314 \pm 129$</td>
<td>$11.64 \pm 0.34$</td>
<td>$1738 \pm 52$</td>
<td>$2.13 \pm 0.06$</td>
</tr>
<tr>
<td>6</td>
<td>$4049 \pm 121$</td>
<td>$10.93 \pm 0.32$</td>
<td>$1633 \pm 48$</td>
<td>$2 \pm 0.05$</td>
</tr>
<tr>
<td>7</td>
<td>$3876 \pm 116$</td>
<td>$10.47 \pm 0.31$</td>
<td>$1552 \pm 46$</td>
<td>$1.90 \pm 0.05$</td>
</tr>
<tr>
<td>8</td>
<td>$3807 \pm 114$</td>
<td>$10.27 \pm 0.30$</td>
<td>$1522 \pm 45$</td>
<td>$1.86 \pm 0.05$</td>
</tr>
<tr>
<td>9</td>
<td>$4424 \pm 132$</td>
<td>$11.95 \pm 0.35$</td>
<td>$1785 \pm 53$</td>
<td>$2.18 \pm 0.06$</td>
</tr>
<tr>
<td>10</td>
<td>$3945 \pm 118$</td>
<td>$10.65 \pm 0.31$</td>
<td>$1584 \pm 47$</td>
<td>$1.94 \pm 0.05$</td>
</tr>
</tbody>
</table>

Conclusions

The data obtained in this work 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 $1522 \pm 45$ to $1796 \pm 53$ nGy/h, with an average value of $1661 \pm 49$ nGy/h, and the average effective dose rate was $2.03 \pm 0.05$ mSv/yr in the investigated samples. Therefore, safety rules and precautions should be applied for those working in these fields.
I would like to thank Sinai Manganese Company for releasing the samples for this study. Also I would like to thank Dr. Abdel Monam, M. Hassan prof. of nuclear physics, Atomic Energy Authority of Egypt, for his useful comments and assistance.

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ДОСЛІДЖЕННЯ ПРИРОДНОЇ РАДІОАКТИВНОСТІ МАРГАНЦЕВОЇ РУДИ

X. A. Абдель Хани

Вміст природних радіонуклідів (238U, 232Th та 40K) у марганцевій руді, що видобувається Сінайською марганцевою компанією (Каїр, Єгипет), визначався методом низькофонової спектроскопії з використанням детектора з надчистого германію. Установлено, що середнє значення, зумовлене трьома радіонуклідами (238U, 232Th та 40K), становить 3543 ± 106, 222 ± 6.6 та 3483 ± 104 Бк/кг відповідно. Установлено, що потужність поглинутої дози, зумовленої природною радіоактивністю зразків, становить від 1522 ± 45 до 1796 ± 53 мГр/год. Радієвий еквівалент активності змінюється від 3807 ± 114 до 4446 ± 133 Бк/кг. Також було оцінено типове значення індексу зовнішньої небезпеки відповідних зразків.

Ключові слова: радіонукліди, 238U, 232Th, 40K, марганцева руда, потужність поглинутої дози, радієвий еквівалент активності, індекс зовнішньої небезпеки.

ИССЛЕДОВАНИЕ ПРИРОДНОЙ РАДИОАКТИВНОСТИ МАРГАНЦЕВОЙ РУДЫ

X. A. Абдель Хани

Содержание природных радононуклидов (238U, 232Th и 40K) в марганцевой руде, которая добывается Синайской марганцевой компанией (Каир, Египет), определялось методом низкофоновой спектрометрии с использованием детектора из сверхчистого германия. Установлено, что средние активности, которые обусловлены тремя радионуклидами (238U, 232Th и 40K), составляют 3543 ± 106, 222 ± 6.6 и 3483 ± 104 Бк/кг соответственно. Установлено, что мощность поглощенной дозы, обусловленной природной радиоактивностью образцов, составляет от 1522 ± 45 до 1796 ± 53 мГр/ч. Радиевый эквивалент активности изменяется от 3807 ± 114 до 4446 ± 133 Бк/кг. Также были оценены типовые значения индекса внешней безопасности соответствующих образцов.

Ключевые слова: радионуклиды, 238U, 232Th, 40K, марганцевая руда, мощность поглощенной дозы, радиевый эквивалент активности, индекс внешней безопасности.

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