Contaminations of Radioactive Nuclides of ^{238}U, ^{232}Th, and ^{40}K in Tin Ore

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ABSTRACT

Problem statement: Tin ore plays a vital role in several industries. Tin ore could contain natural radioactive nuclides of ^{238}U, ^{232}Th, and ^{40}K with various concentrations which might be caused a significant exposure radiation levels to the workers who handle the Tin ore in the factory. Thus, the evaluation of natural radioactive nuclides ^{238}U, ^{232}Th, and ^{40}K in the Tin ore is important from the point of view of radiation protection to save the workers from the radiation hazards.

Approach: The natural radionuclides of ^{238}U, ^{232}Th, and their daughters and ^{40}K as well in Tin ore were measured using in gamma-ray spectrometry system.

The results: The activity concentrations of ^{238}U, ^{232}Th, and ^{40}K in Tin ore was 42.77, 21.9 and 145.33Bq kg^{-1}, respectively.

Conclusion: The natural radionuclides concentrations in Tin ore were lowered the exemption level recommended by IAEA. Moreover, all the radiological hazard indices were very low compared with the safe values recommended with UNSCEAR. Therefore, the obtained results imply that Tin ore can be used in industries with any risk of radiation.

Keywords:

Tin Ore; Natural radionuclides; hazard index; radiation dose; gamma spectrometer; NAA

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1- INTRODUCTION

Over 60 radionuclides can be found in the natural environment. They can be classified into three general categories, i.e. Primordial - formed before the creation of the Earth, Cosmogenic - formed as a result of cosmic ray interactions, and Human Produced - enhanced or formed due to human actions. Radionuclides that are found naturally in air, water, and soil, even in our bodies are natural radionuclides or naturally occurring radioactive material (NORM). Every day, we ingest and inhale radionuclides in our air, food and the water. Natural radioactivity is common in the rocks and soil that makes up our planet, in water and oceans, and in our building materials and homes [1]. There is no-where on Earth that we cannot find natural radioactivity [2]. Some radioactive nuclides are detectable in the soil. They belong to the natural radionuclides like the members of the uranium and thorium decay series. More specifically, natural radioactivity and the associated external exposure due to gamma radiation depends primarily on the geological and geographical conditions and appear at different levels in the soils of each region in the world [3,4]. The specific levels of terrestrial environmental radiation are related to the geological composition of each litho-logical separated area and to the content of the rock from which the soils originated in each area in the radioactive elements of thorium (^{232}Th), uranium (^{238}U) and potassium (^{40}K). It is well known, for instance, that igneous rocks of the granitic composition are enriched with thorium, uranium compared to other rocks of basaltic or ultramafic composition [3,10,11]. For that reason, higher radiation levels are associated with igneous rock and lower level with sedimentary rocks. There are exceptions, however, as some shales and phosphate rocks have a relatively high content of these radionuclides [3,4]. Uranium minerals are chemically weathered to soluble U(VI) complexes and carried by river water downstream to the oceans, while the primary mode for transport of thorium from the continents to the oceans follows the detrital phase [5,6]. The residence time for thorium in seawater is only approximately 300-350 years. Uranium remains soluble in the sea (as carbonate and other complexes) and has a residence time in seawater of some 500000 years [7]. Precipitation of uranium can occur easily by reduction to insoluble U(IV). Thus, environments in which carbonaceous and bituminous shales form are particularly favorable for U removal by reduction of U(VI) to U(IV). Lignites are also enriched in uranium, some as U(IV), as expected, and some as U(VI) because the latter form can easily be scavenged by coaly material without reduction. This accounts, in part, for the high U content of such rock. In the case of phosphorus rocks, co-precipitation of U(IV) with $Ca2^{+}$ is likely owing to their very similar ionic radii, but the exact mechanism for the reduction of U(VI) to U(IV) in these rocks is not known [7]. The Th/U ratio in nature varies widely. In rocks from which U has been removed, high Th/U ratio results; conversely, in rocks precipitated under chemically reducing environments far from suspected rock source, U is enriched over thorium. Thus, above average Th/U ratios are observed in continental sediments, especially in laterites and other residual deposits. Low Th/U ratios are found in chemically precipitated marine sedimentary rock, such as evaporate sand, limestone, and extremely low Th/U ratios are found in carbonaceous rock. [7]. Human beings have always been exposed to natural radiations from their surroundings. The exposure to ionizing radiations from natural sources occurs because of naturally occurring radioactive elements in the soil and rocks, cosmic rays entering the earth's atmosphere from outer space and the internal exposure from radioactive elements through food, water, and air. Therefore the assessment of gamma radiation dose from natural sources is of particular importance as natural radiation is the largest contributor to the external dose of the world population [8,9].

Wherever, Tin ore is used for many industrial purposes, such as Paints, Plastics industry, Container, Bronze alloys, Electroplating and various alloys of Tin. Consequently, there are specific measures to protect the working in Tin Ore related industry such as reducing exposure levels and the time of exposure and the use of exhaust ventilation. So that the aim of this work is to determine the concentration of natural radioactive uranium, thorium, and potassium in Tin Ore and to measure the surface radiation dose rate and the radium equivalent activity and radiation hazard index. Final determination of the elements presented in Tin ore samples by NAA using two identical isotopic radioactive Am-Be neutron Sources.

2- MATERIALS AND METHODS

Tin ore was collected from Nuweiba area in the Eastern Desert of Egypt, Egyptian General Authority for Mineral wealth Resources, Ministry of Petroleum. Tin was put into clean containers. Samples were sealed and stored for a 30 day period to achieve the secular equilibrium between radium and its products. Then, they were measured for 18000 s. The energy and intensity of various gamma-ray lines have been measured using High-Purity Germanium detector (HPGe, Canberra coaxial) which has a photo-peak efficiency of 70 %. The energy resolution of 2 KeV Full-Width at Half Maximum (FWHM) for the 1332 KeV gamma-ray line of ^{60}Co. A cylindrical lead shield of five cm thickness, which contains the inner concentric cylinder of Cu with a thickness of 10 mm, was used to shield the detector and to reduce the effect of the background. The detector was cooled to liquid nitrogen temperatures and coupled to a PC-based 8K multichannel analyzer and an ADC with Genie 2000 for data acquisition and analysis. The calibration of the detector was carried out by using standard point sources ^{60}Co (1173.2 and 1332.5 KeV), ^{133}Ba (356.1 KeV), ^{137}Cs (661.9 KeV) and ^{22}Na (1368.6 KeV) besides ^{226}Ra (186.2 KeV). Absolute efficiency calibration curves are calculated for activity determination of the sample by using standard ^{226}Ra, contained in the same cylindrical bottles as the samples. The samples were prepared with a uniform geometry. An empty bottle with the same geometry was measured for subtracting the background. The gamma-ray transitions of energies of 1120.3 KeV (^{214}Bi) and 1764 KeV (^{214}Bi) were used to determine the concentration of the ^{238}U series. The gamma-ray transitions of energies 911.1 KeV (^{228}Ac) and 2614 KeV (^{208}Tl) were used to determine the concentration of the (^{232}Th) series. The 1460 keV gamma-ray transition of ^{40}K was used to determine the concentration of ^{40}K in the samples as shown in Table (1) and their intensities. The spectra of the samples were analyzed using a special PC Genie 2000 software to calculate the concentrations of ^{238}U, ^{232}Th and ^{40}K and their decay products.

Table (1) the natural radionuclides, their gamma lines used and their intensities [12].

Parent Nuclide	Daughter Nuclide	ɣ-ray energy (keV)	Abundance (%)
^{226}Ra	^{214}Bi	1120.28	14.9
	^{214}Bi	1764.52	16.07
^{232}Th	^{228}Ac	911.16	29.0
	^{208}Tl	2614.7	36.0
^{40}K	^{40}K	1460	10.67

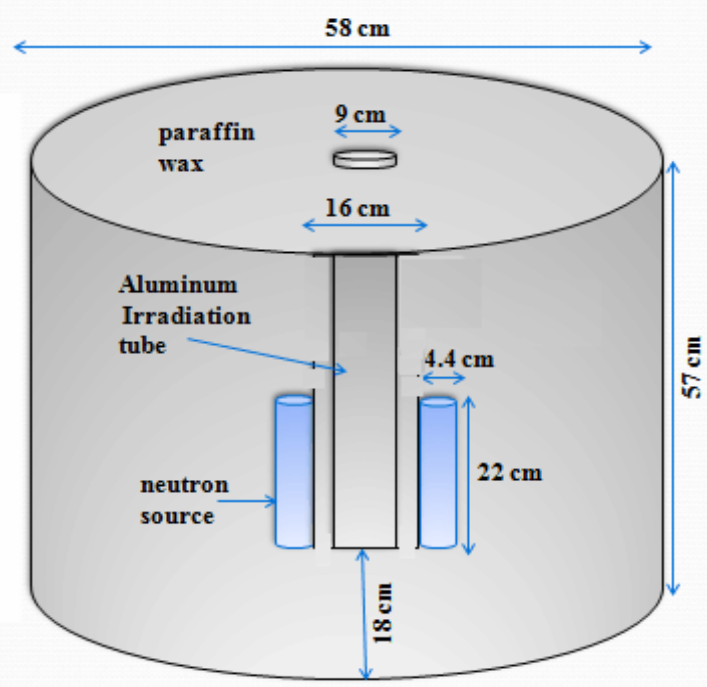

Figure (1-a): Describes the neutron irradiation facility used through the investigation.

The present facility plans to use two identical isotopic radioactive neutron sources of Am-Be type. Each source has a cylindrical shape with dimensions of about 22 cm long and 4.4 cm in diameter and has an activity of about 5 Curies (185 GBq). The sources are put in an iron cylindrical barrel, which filled with melted pure paraffin where the sources are isolated from the inner vertical central irradiation tube with the same paraffin material as shown in figure (1-a, b).

3- RESULTS AND DISCUSSION

3.1) Natural specific activity measurement

The activity levels for radionuclides in the measured samples are computed using the following equation [13] :-

$$A = C / I\, m\varepsilon \qquad (1)$$

Where : *A*=The specific activity level of a certain radionuclide (Bq/ kg)

C= The net count rate of the sample (counts/seconds)

ε= The detector peak efficiency for the specific gamma ray energy

I= The intensity of gamma-rays of each radionuclide

m=The mass of dried sample (kg).

^{238}U activity concentration was determined by measuring the 295.2 KeV (18.7 %) and 351.9 KeV(35.8.1%) gamma-rays from ^{214}Pb and the 609.3 KeV (45 %) and 1120.3 KeV (14.9 %) gamma-rays from ^{214}Bi. ^{232}Th activity was determined from the gamma-rays of 238.6 KeV (45 %) from ^{212}Pb and 338.4 KeV (12 %), 911.1 KeV (29 %) and 968.6 KeV (17.5 %) from ^{228}Ac and 583.1 KeV (30 %) gamma-rays from ^{208}Tl. ^{40}K concentration was measured from its 1460 KeV (10.67 %) gamma-ray line.

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 a sample is shown in Fig. 2, 3.

Activity concentration of ^{226}Ra, ^{232}Th, and ^{40}K radionuclides have been determined in the samples. The minimum, maximum and mean activity values of ^{226}Ra, ^{232}Th and ^{40}K found in these samples are listed in table 1. As may be seen in this table the measured values of activity in the samples due to ^{232}Th vary from 21.9 Bq kg^{-1} to 9.9 Bq kg^{-1}, ^{226}Ra activities vary from 42.77 Bq kg^{-1} to 21.43 Bq kg^{-1} and vary in ^{40}K activity ranges from 145.33 Bq kg^{-1} to 16.22 Bq kg^{-1}. The activity concentrations shown in table 2 were lower than world average except value recommended with IAEA, 2014

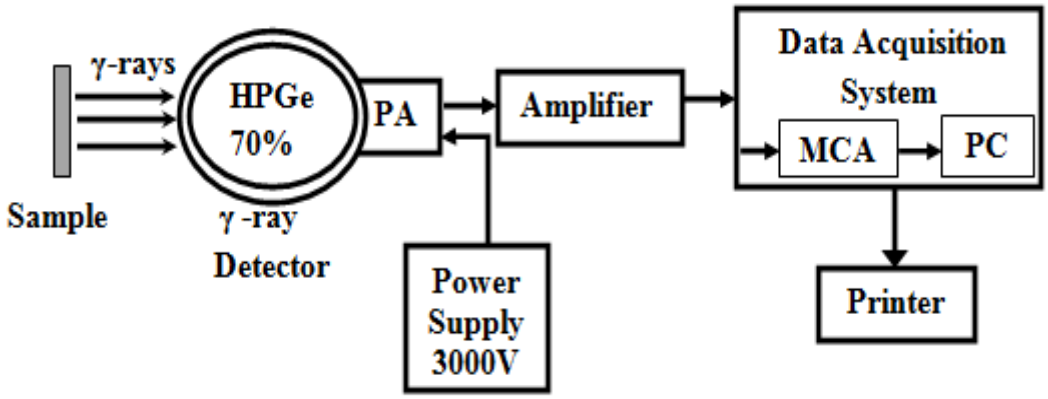

Fig. (1-b): Blocked diagram of HPGe γ-ray spectrometer system.

Table 2: The minimum, maximum and average values of the activities due to ^{226}Ra, ^{232}Th and ^{40}k in Tin ore sample.

Range	^{232}Th concentration ($Bqkg^{-1}$)	^{226}Ra concentration ($Bqkg^{-1}$)	^{40}K concentration ($Bqkg^{-1}$)
Minimum	9.9	21.43	16.22
Maximum	21.9	42.77	145.33
Mean	15.9	32.1	80.78

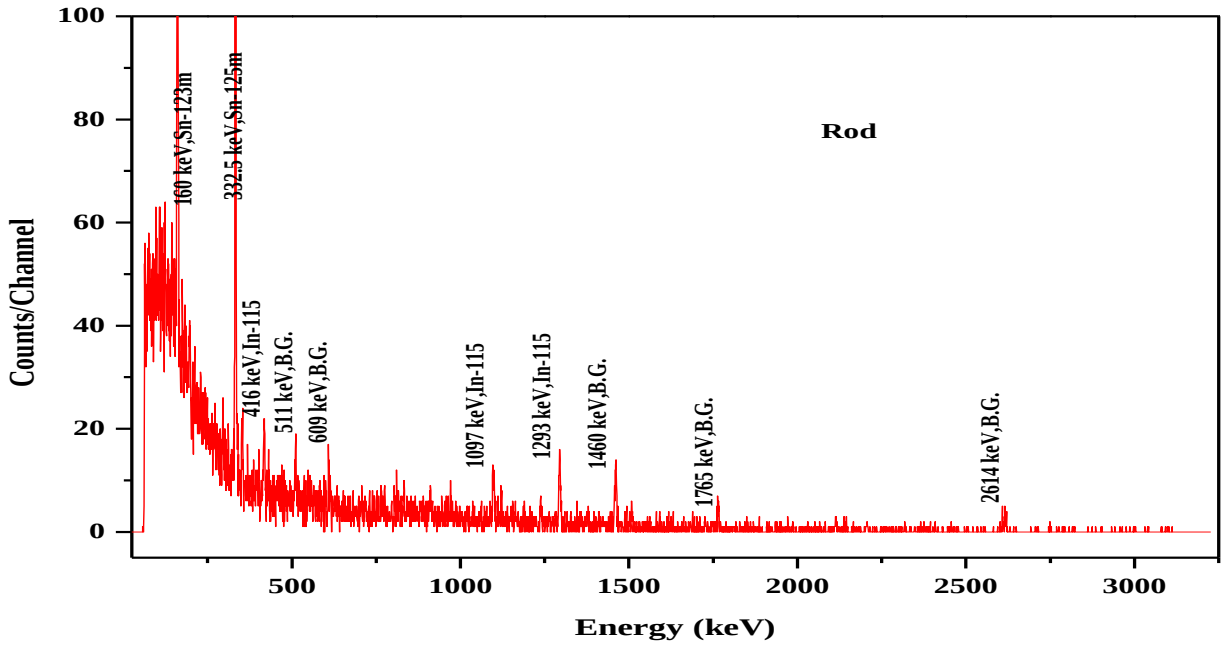

Fig.2: Portion of Gamma-ray spectrum irradiation 2days, decay 0 sec, 900 sec counting illustrates the analysis of cement sample by NAA Technique.

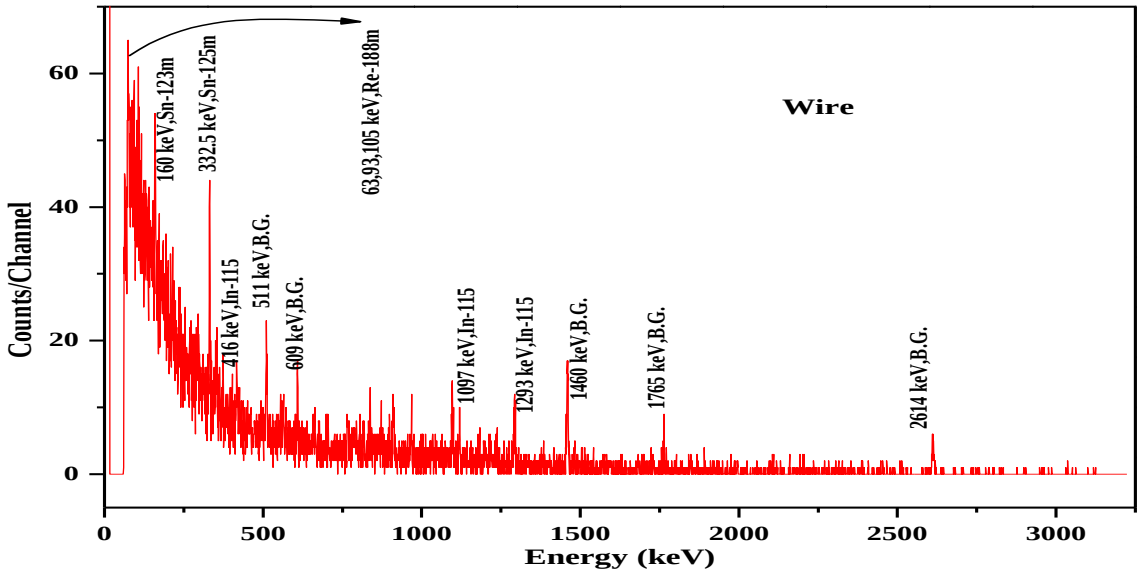

Fig. 3: Portion of Gamma-ray spectrum irradiation 2days, decay 900 sec, 900 sec counting illustrates the analysis of cement sample by NAA Technique.

To assess the radiological hazard of the Tin ore sample, it is useful to calculate an index called the radium equivalent activity, Ra_{eq}, defined according to the estimation that 1 Bq / kg of ^{226}Ra, 1.43 Bq / kg of ^{232}Th and 0.077 Bq / kg of ^{40}K produce the same γ-ray dose [13]. This index Ra_{eq} is given as:

$$\mathbf{R_{eq} = A_{Ra} + 1.43\ A_{Th} + 0.077\ A_K} \qquad \mathbf{(2)}$$

Where A_{Ra}, A_{Th}, and A_K are the activity concentration in Bq kg^{-1} of ^{226}Ra, ^{232}Th, and ^{40}K, respectively. The maximum value of Ra_{eq} in Tin ore samples must be less than 370 Bq kg^{-1} for safe use (UNSCEAR, 1993), i.e., to keep the external dose below 1.5 mSv y^{-1}. The values of Ra_{eq} are below this criterion limit. In Tin ore samples,

the Ra_{eq} activity is within the recommended safety limit when used in industry. The calculated values of the radium equivalent Ra_{eq} for the studied Tin ore samples are given in table 3.

Another radiation hazard index, the representative level index, $\mathbf{I_\gamma}$, used to estimate the levels of γ-radiation hazard associated with the natural radionuclides in specific Tin ore samples, is defined as [15].

$$\mathbf{I_\gamma = (A_{Ra} / 150) + (A_{Th} / 100) + (A_K / 1500)} \qquad \mathbf{(3)}$$

Where $\mathbf{A_{Ra}}$, $\mathbf{A_{Th}}$, and $\mathbf{A_K}$ are the activity concentrations in Bq/kg of ^{226}Ra, ^{232}Th and ^{40}K respectively. The values of $\mathbf{I_\gamma}$ for the studied samples are given in table 3. It is clear that the studied Tin ore samples do not exceed the upper limit for the representative level which is unity.

3.2) RADIATION HAZARD INDEX

This factor is used to estimate the level of gamma radiation hazard associated with natural radionuclides in specific Tin ore samples. The external hazard index is obtained from Ra_{eq} expression through the assumption that its maximum value allowed (equal to unity) corresponds to the upper limit of Ra_{eq} (370 Bq kg^{-1}) according to UNSCEAR, 1993. This index value must be less than unity in order to keep the radiation hazard insignificant; then, the external hazard index (H_{ex}) can be defined as [16],

$$\mathbf{H_{ex} = (A_{Ra} / 370) + (A_{Th} / 259) + (A_K / 4810)} \qquad \mathbf{(4)}$$

Where A_{Ra}, A_{Th}, and A_K are the specific activities of ^{226}Ra, ^{232}Th and ^{40}K (in Bq. kg^{-1}) were calculated for the investigated samples to indicate different levels of external γ-radiation due to different combinations of specific natural activities in specific Tin ore samples.

Table 3: The minimum, maximum and average values of the activities due to Radium equivalent activity (Bq kg^{-1}), External Annual dose (mSv/y) and Gamma-Radiation hazard (H_{ex})

Range	Radium equivalent activity (Bqkg^{-1})	External Annual dose (mSv/y)	H_{ex}
Minimum	35.94	0.16	0.10
Maximum	85.36	0.34	0.24
Mean	60.65	0.25	0.17

3.3) EXTERNAL ANNUAL DOSE

The external annual effective dose (EAD) is calculated. The equation used to calculate the annual effective dose may be defined as [17],

$$\mathbf{EAD = (0.92\ A_{Ra} + 1.1\ A_{Th} + 0.08\ A_K) \times 10^{-9}\ (Gy/\ h) \times (0.7\ Sv/\ Gy)\ (24 \times 365 \times 0.8\ /y)} \qquad \mathbf{(5)}$$

Where, 0.92, 1.1 and 0.08 are the specific dose rates of Ra, Th, and K, respectively; with an estimated indoor occupancy factor of 0.8.

4- CONCLUSION

It can be concluded that the measured values of activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K in Tin ore samples were lower than the recommended safe values by IAEA. Moreover, the radium equivalent activities calculated from measured values of ^{226}Ra, ^{232}Th, and ^{40}K activities in the Tin ore samples were lower than the

upper recommended limit of 370Bqkg^{-1}. These data serve as a basis for the assessment of radiological hazard to the workers involved in Tin ore mining, transportation and industrial applications in Egypt. The calculated external hazard indices are less than unity. Therefore, the Tin ore is acceptable for use in industries. The obtained results of this assessment indicated that the levels of natural radioactivity were lower than the international recommended limits, Thus, Tin ore materials have radiation risk to the workers who handle it in factories.

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