MEASUREMENTS OF THORIUM CONTENT AND RADIOACTIVITY IN GAS MANTLES

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الخلاصـة :

تم قياس إشعاع جاما الطبيعي للثوريوم-٢٣٢ وولائده في أنواع مختلفة من فتائل فوانيس "التريك" المستوردة والمستخدمة في المملكة العربية السعودية باستخدام تجهيزات تحليل أطياف جاما الموجودة في جامعة الملك فهد للبترول والمعادن. وتم تحديد اشعاع جاما الطبيعي من هذه الفتائل باستخدام كاشف من الجرمانيوم عالي النقاوة معزول تماماً. كذلك تم قياس تركيز النظير المشع يورانيوم-٢٣٨ - باستخدام كاشف يوديد الصوديوم NaI(TI)، متصل بحاسب آلي لجمع المعلومات ومعالجتها. وقيست إشعاعية عينتين معياريتين معلومتي النسب من الثوريوم بنفس الأجهزة وذلك لحساب الكمية المطلقة من الثوريم-٢٣٨ في كل فتيل لغرض المقارنة. ووجد أن نسب كتلة الثوريوم في الفتائل تتراوح بين ٣,٦٪ و ٩,٢٪ بينما تتراوح اشعاعية جاما في الفتائل بين ٧ و ٢٥ نانوكوري علماً بأن كتلة الفتيل الواحد تبلغ عدة غرامات. وتم كذلك دراسة توزيع الثوريوم في الفتيل باستخدام كواشف الآثار النووية من نوع CR-39. وقيست الجرعة الخارجية المكافئة لأشعة جاما من أحد الفتائل بلصقها على سطح الكاشف فكانت أربعة مايكروسيفرت بالساعة. وتم حساب الجرعة الاشعاعية المكافئة الملزمة التي يمكن أن تنتج عن ابتلاع (امتصاص) ١٠٪ من ثوريوم الفتيل الأقل والأكثر إشعاعية فوجد أنها تبلغ (٠,٥) ملي سيفرت و (١,٨) ملي سيفرت على التوالي.

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ABSTRACT

Natural gamma rays from the thorium (^{232}Th) isotope and its daughters in various imported gas lantern mantles were measured at the KFUPM gamma-ray spectroscopy setup. The origin of the natural gamma rays was qualitatively determined by a well shielded Hyper Pure Germanium (HPGe) detector, and then the concentration of the radioactive element was quantitatively measured using a 5″×5″ NaI(Tl) detector and a PC-based data acquisition and analysis system. The mantle samples, of a few grams each, were studied for thorium concentration and its gamma ray radioactivity. In order to determine the absolute concentrations, the sample activity was compared with the activities of standards. The concentration of Th was determined in weight percent (wt.%) and ranged from 3.6 to 9.2% while the activity per mantle ranged from 7 to 25 nCi. Thorium distribution measurement in the mantle was carried out using CR-39 nuclear track detectors. The external gamma rays dose equivalent rate from one mantle sample was measured as 4 μSv/h when the mantle was placed on the detector surface. The calculated committed dose equivalent for ingestion of 10% of each of the lowest and the highest concentration of thorium mantles was found to be 0.5 mSv and 1.8 mSv respectively.

MEASUREMENTS OF THORIUM CONTENT AND RADIOACTIVITY IN GAS MANTLES

1. INTRODUCTION

Thorium is used for producing luminescence in gas lantern mantles. It is introduced into the mantle by impregnating the fabric mesh with thorium nitrate which changes to thorium oxide when the mantle is burned in air [1]. Thorium oxide glows with a brilliant white light when heated in fuel. Gas mantles containing natural radioactive thorium present a radiation health hazard [2]. Many research groups have studied the thorium content and radioactivity of gas lantern mantles [1–4]. A study by Doretti *et al.* [3] involved the determination of thorium isotopes in mantles by alpha-spectroscopy, while Mohammadi and Mehdizadeh [2] and Luetzelschwab and Googins [1] determined thorium activities by comparing the gamma-counts of ^{228}Th daughters with the gamma-counts of the ^{228}Th daughters in thorium reference samples of known ^{232}Th content in which the entire thorium family was in equilibrium (see Figure 1). Couch and Vaughn [4] identified thorium in a mantle by gamma rays emitted by lead-212 (^{212}Pb), in the thorium decay series. They also assessed the internal and external exposure hazard of a mantle. Commercial thorium is produced mainly from ores such as monazite or bastnaesite in which ^{232}Th and ^{228}Th are in secular equilibrium [3]. When thorium is extracted and purified, one may assume that no daughters are present at a time $t = 0$, and the ^{232}Th and ^{228}Th activities are equal. However ^{228}Th, because of its relatively very short half-life compared with ^{232}Th, decays faster than its production by ^{232}Th at the beginning reaching its minimum activity at 4.58 years (42% that of ^{232}Th) after extraction, then starts growing with time to be in secular equilibrium with ^{232}Th again [3]. However, ^{228}Th daughters, because of the shortness of their half-lives (see Figure 1) build up and reach equilibrium with ^{228}Th about twenty days after extraction. Therefore when ^{208}Tl activity is measured it represents that of ^{228}Th, and to find the activity of ^{232}Th one also needs to know the time elapsed after thorium extraction [3].

Gas lantern mantles are usually used in rural areas and in camping as a source of light. Many types of mantles are in use in Saudi Arabia. It is important to measure the thorium content in samples of these mantles to asses the risk to users and sellers of these items. The gamma-radiation emitted from the mantles and the possible inhalation or ingestion of the fine thorium oxide powder during replacement may become a health hazard for regular users.

A low level gamma-ray spectroscopy setup has been established at the Energy Research Laboratory of the Research Institute to measure the natural gamma-ray activity in various samples [5]. The setup was used to measure the thorium content of different types of gas lantern mantles. In this paper the results of these measurements are presented. Thorium distribution in the mantle was carried out using CR-39 nuclear track detectors. The external gamma dose from the mantle was measured using a Cavity Ionization Chamber. An assessment of the risk arising from the use of these mantles is presented. It is thought that this study is the first of its kind in Saudi Arabia.

2. EXPERIMENTAL METHOD

Six different types of gas mantles were collected from local commercial shops. The mantles were carefully weighed to a sensitivity of 1 mg, then the natural gamma rays of the mantle samples were measured using the low level gamma-ray spectroscopy setup at KFUPM/ERL [5]. The setup consists of a high purity germanium HPGe detector and a thallium-activated sodium iodide NaI(Tl) detector together with signal processing equipment and a data acquisition and analysis system. A schematic diagram of the setup is shown in Figure 2. The high energy resolution of HPGe allows precise identification of the element while the higher efficiency of NaI(Tl) detector allows the determination of the element's concentration. Therefore a gamma ray setup consisting of a HPGe and a NaI(Tl) detector allows the determination of the element's concentration with a good level of accuracy [5].

To identify the elements, the sample activity was first measured using the HPGe detector with 2 keV resolution at 1333 keV of ^{60}Co and with a relative efficiency of 33%. Then a 5″×5″ NaI(Tl) detector with 7.3% resolution was used to determine the concentrations of the elements. After preamplification, the signal is processed through a Spectroscopy Amplifier and an ADCAM Multichannel Buffer Model 919 of EG&G Ortec hooked to an IBM-PC computer. The spectra were acquired in 4096 channels and calibrated with standard gamma-ray sources (^{60}Co and ^{137}Cs). The energy range of the spectrum covered up to 3000 keV which cover the 1460 keV line from ^{40}K, the 1765 keV line from ^{214}Bi, and the 2615 keV line from ^{208}Tl [5, 6].

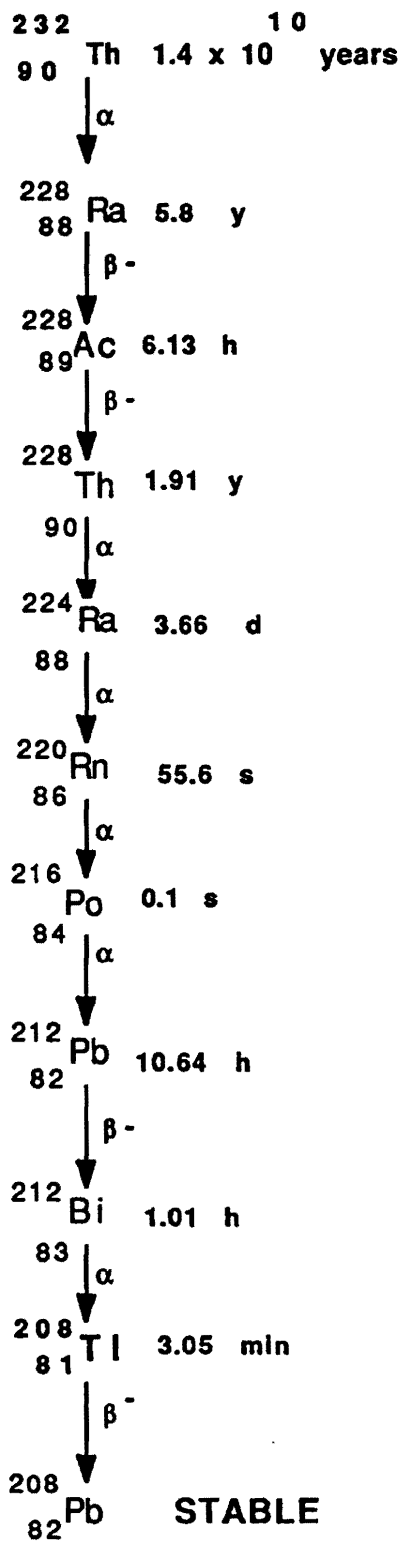

Figure 1. Natural Radioactivity Decay Series of Thorium-232 (^{232}Th).

The detector is well shielded, thus the room background is very low and the dead time of the system is negligibly small. In order to obtain better statistics, the gamma-ray activity of each sample was measured for 24 hours, and the acquired spectra were stored. Room background measurements were taken several times for the same period as the sample and the average value from the background runs was used in the data analysis. Figures 3*a* and 3*b* show a typical spectrum of gamma-rays of low and high energies, respectively, from a sample using the HPGe detector while Figure 3*c* shows the high energy side of this spectrum acquired using the NaI detector. The HPGe spectra did not show any interference under the thorium gamma ray peaks. This means that the radioactivity in the mantle sample comes only from the gamma lines of thorium daughters as shown in Figures 3*a* and 3*b*. In other words, no uranium or considerable amount of potassium is present in the sample. The collected spectra from the NaI(Tl) detector were then analyzed off-line by putting the Region of Interest (ROI) around the 2615 keV peak of ^{208}Tl. The interval of the gate was 390 channels which covers the whole peak (see Figure 3*c*). The integrated counts under the peak were determined. The background subtraction and dead time correction were also done. Later on, the sample mass normalization was performed.

Similar measurements were done for two standard thorium ore samples. The standards were provided by the U.S. Department of Energy, New Brunswick Lab. (NBL), USA. They have certified values of thorium in percentage by weight (wt.%). The two thorium standards were NBL 79-A and NBL 80-A with thorium ore concentrations of 1% and 0.1005% respectively. Thorium standards with a mass of up to 103 gram/sample were carefully weighed with a balance having a sensitivity down to the 1 mg range. They were packed inside plastic containers of about 3″ diameter and 0.5″ height. The standard containers were then air-tight sealed and placed in heat sealed thin plastic bags to isolate the standards from ambient conditions. Similarly, a quartz sample (SiO_2), free of natural radioactivity [7, 8] was prepared with a geometry identical to that of the standards. The quartz sample was used for sample related background measurements.

Finally, from the comparison of corrected net counts from samples and standards, the concentrations of Th in wt.% in gas lantern mantle samples were determined.

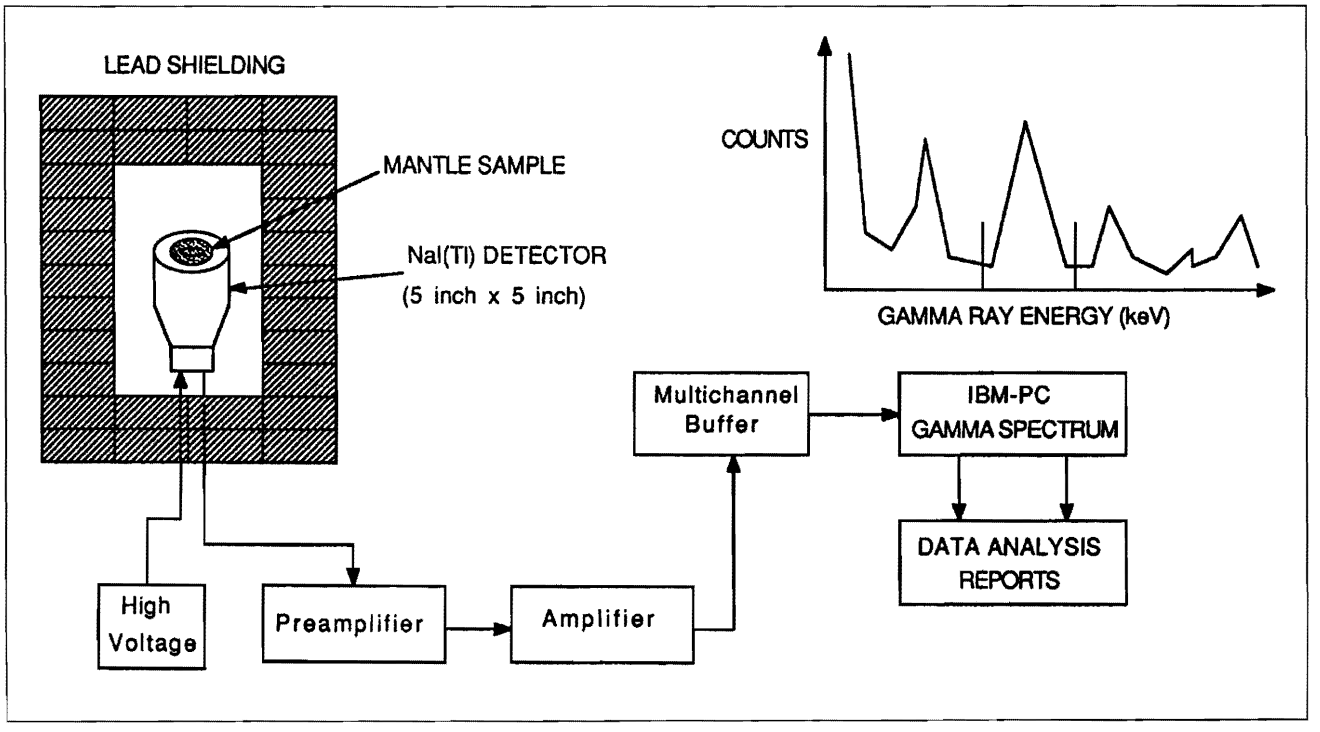

Figure 2. A Schematic Diagram of the Natural Gamma Ray Spectroscopy Setup at the KFUPM/ERL.

The total activity from each mantle sample was calculated using the following equation of specific activity for natural thorium [9]:

$$A_{specific} = 4.0 \times 10^3 \text{ Bq/g of thorium } (1.11 \times 10^{-7} \text{ Ci/g of thorium}) \quad (1)$$

where it was assumed that ^{232}Th and ^{228}Th are in secular equilibrium.

The distribution of the thorium content in a mantle sample was studied using alpha emitted by thorium and its daughters using nuclear track detectors of the type CR-39, 2×2 cm^2. The detectors were placed over a gas mantle for different times (1–7 days), then the detectors were removed one by one at different periods. The first detector was removed after one day, the second after two days, and so on until 7 days had elapsed. The CR-39 detectors were chemically etched under the following conditions: 30% KOH, 60°C, and 4 hour etching. The detectors were counted under an optical microscope and counting results are shown in Figure 4 which shows clearly that there is a linear relationship between exposure time and alpha tracks counted per unit area on the different detectors.

To study the distribution of the mantle thorium content, 5 CR-39 detectors were placed on different locations on one mantle for the same period of time (4 days). The alpha tracks on each detector were then counted.

The total external gamma dose equivalent rate of the mantles was measured using a Cavity Ionization Chamber "Victoreen Reader Model 570 and Chamber Model 227". Seven readings were taken over a period of three weeks and the average dose equivalent rate (μSv per hour) was obtained. Finally an internal risk assessment was performed assuming 10% ingestion of the ash from two mantles.

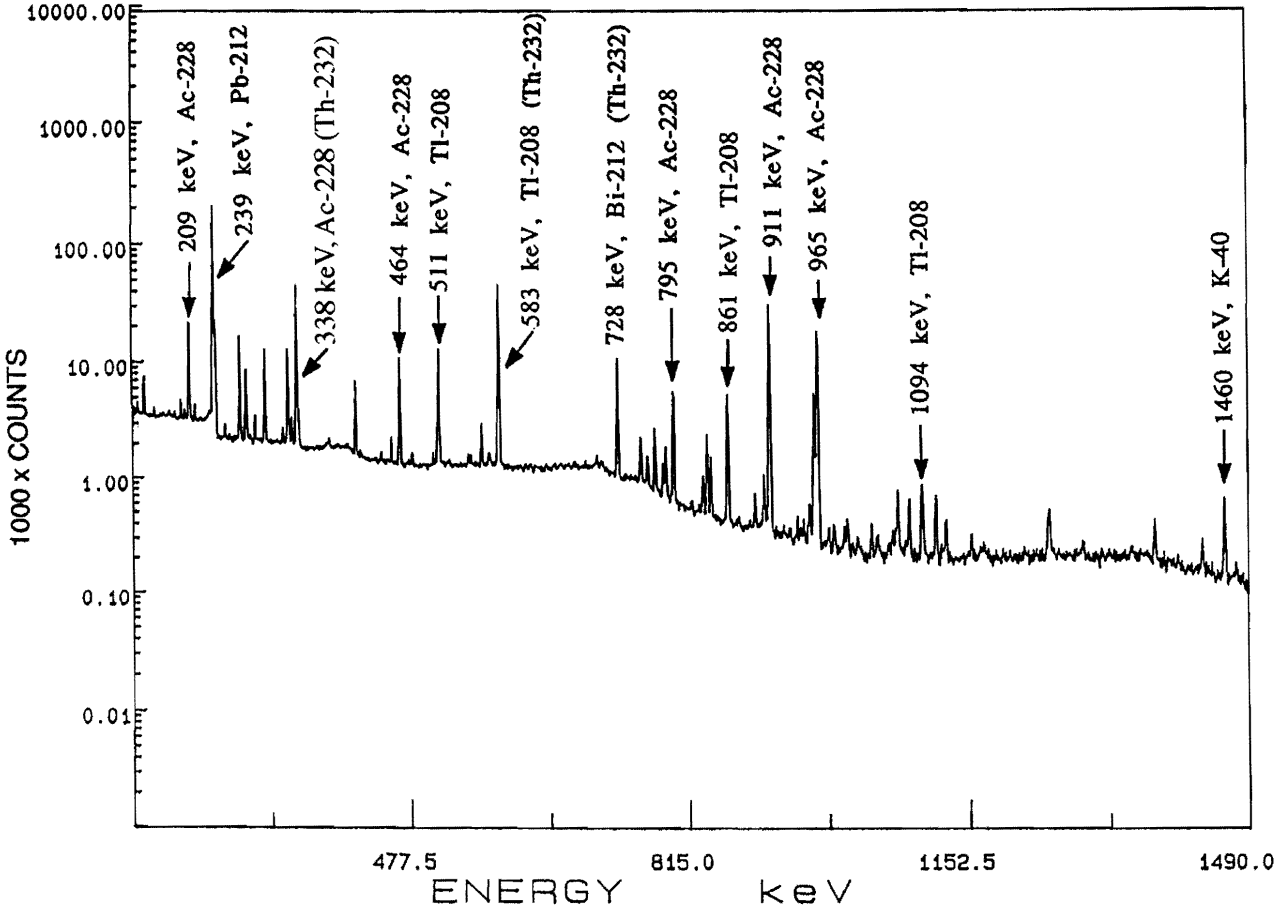

Figure 3a. A Typical Spectrum of Natural Gamma Rays of Low Energies from Thorium Daughters in a Gas Lantern Mantle. Measurement was taken by HPGe detector.

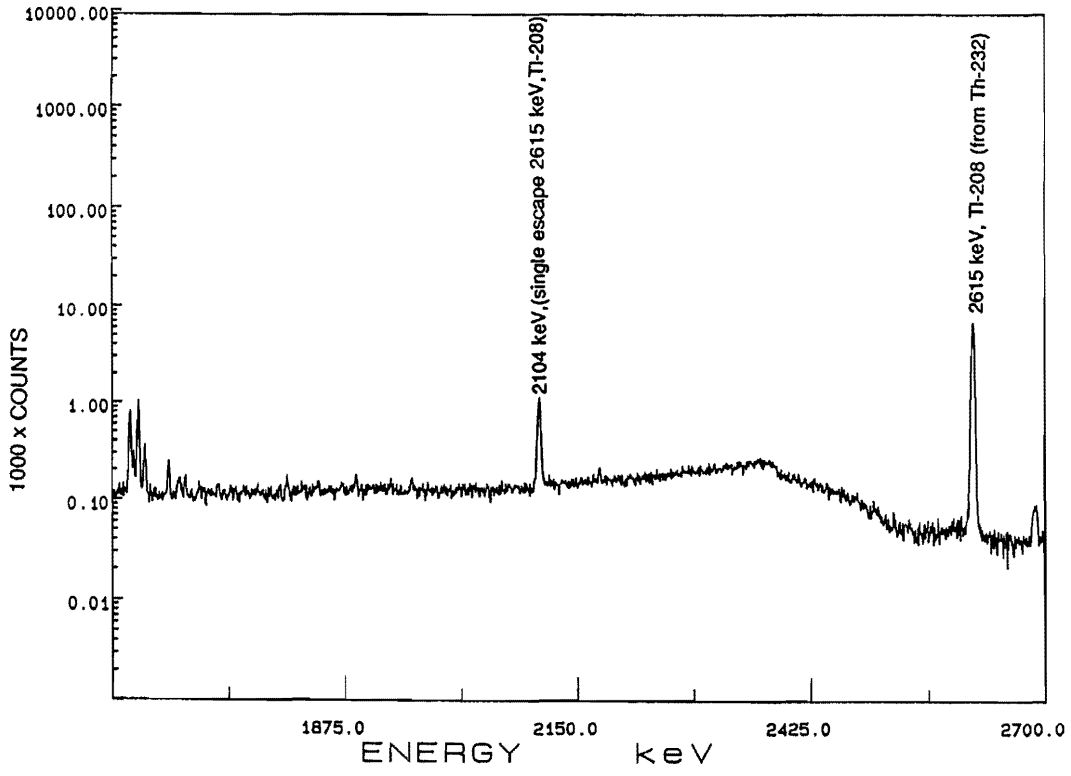

Figure 3b. A Typical Spectrum of Natural Gamma Rays of High Energies from Thorium Daughters in a Gas Lantern Mantle. Measurement taken by a HPGe detector.

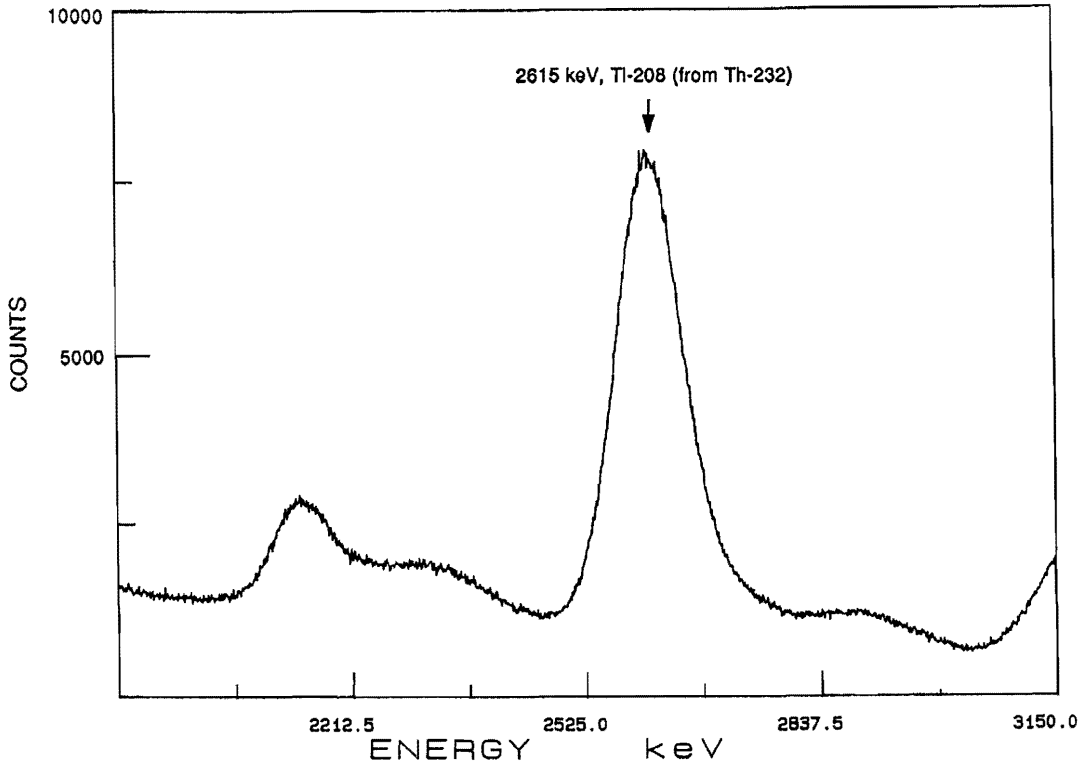

Figure 3c. A Typical Spectrum of Natural Gamma Rays from Thorium Daughters in a Gas Lantern Mantle. Measurement taken by a NaI(Tl) detector.

To determine the concentration of Th in the sample the following equation was used [11]:

$$\mathrm{Th} = (1/F_{\mathrm{th}})\,(A_1 - B_1) = (1/F_{\mathrm{th}})\,A_{\mathrm{th}} \tag{2}$$

where,

A_1 represents the total integrated counts/day under the peak in the ^{208}Tl region of interest (ROI); F_{th} is the calibration factor for the thorium series in the ^{208}Tl ROI given by:

$$F_{\mathrm{th}} = \frac{\text{Net counts/day in the Tl ROI of the standard of 1g}}{\text{The concentration of actual thorium in the standard}}$$

B_1 represents background counts over the ^{208}Tl ROI measured for the same period of time;

A_{th} are net counts for ^{208}Tl over the ^{208}Tl ROI normalized to sample weight of 1 g.

The net counts under each peak were obtained by subtracting the background from the measured counts in the same ROI of the spectra. The calibration factors were determined from the measurements of the standards. The weights of the samples were determined with a 1 mg uncertainty. Therefore, by inserting these measured values into the above equations, the concentrations of Th in wt.% can be calculated. The uncertainties of the measurements are the statistical uncertainties of the measurements with those of the background counts folded in.

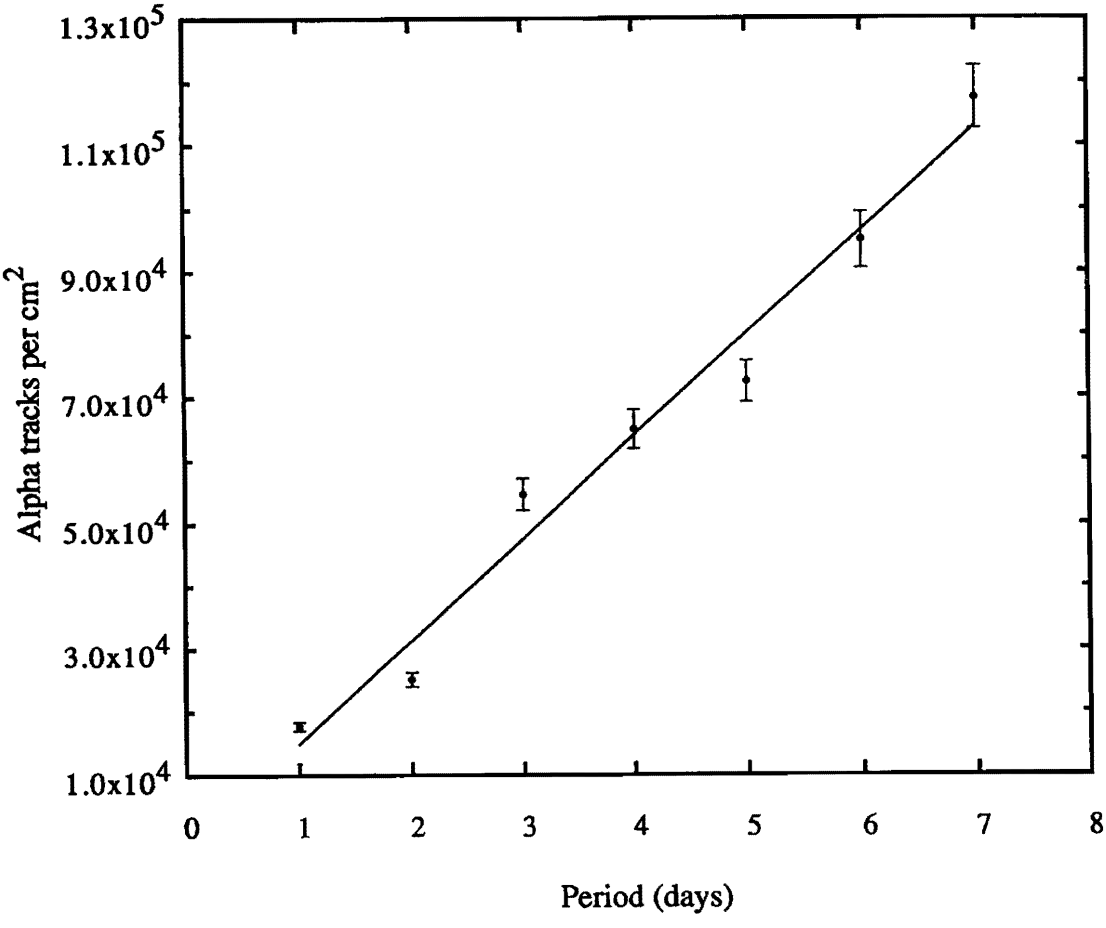

Figure 4. Variation of Alpha Tracks Densities on CR-39 Detectors with Exposure Time on a Gas Lantern Mantle, Showing Linear Relationship.

3. RESULTS AND DISCUSSION

Table 1 lists the results of the measurements for 9 studied mantle samples. As shown in Table 1, the Th concentration in the mantles ranges from 3.6% to 9.2% which is much more than the 0.25% quantity stated by ERDAs CFR 40.13 (c) Rules and Regulations to be unimportant [2]. The thorium content in the mantles studied varied from a minimum of 61 mg to a maximum of 220 mg of Th. This is almost one third of the ^{232}Th contents in the work by Doretti *et al.* [3] which varied from 200 mg to 450 mg. This difference could be due to the fact that ^{228}Th and ^{232}Th might not be in secular equilibrium and that the types of mantles studied might be different.

The total activity calculated by Equation 1 for these mantles is also shown in Table 1. The activity per mantle ranges between (2.4 to 8.8) $\times 10^2$ Bq, which corresponds to 6.8 and 24.5 nCi, depending on the type of the sample. Leutzelschwab and Googins [1] reported activities ranging from 45 to 75 nCi per mantle. The activities reported by Mohammadi and Mehdizadeh [2] ranged from 14 to 37 pCi/gm. Their mantle masses were similar to ours. Comparing the activities found with others [1, 4] it is clear that the mantle activities reported by Mohammadi and Mehdizadeh [2] should be nCi/gm rather than pCi/gm.

The external dose equivalent rate of one mantle sample (No. 4, with 151 mg of Th, see Table 1) was 4 μSv/h when the mantle stuck to the detector surface, which is equivalent to 8 mSv per year. The dose equivalent rate limit for the general public, in the latest ICRP recommendation of 1990, is 1 mSv per year. The dose equivalent rate will be much lower if quantities of mantles are stored at a distance of, say 1 m. Therefore it is recommended that the mantles be stored at a distance of 1 m or more from the working area.

The calculated committed dose equivalent for ingestion of 10% of each of the lowest and the highest concentration of thorium mantles (7 to 25 nCi) in Table 1 is 0.5 mSv and 1.8 mSv respectively [4]. The dose limit per year for the general public is 1 mSv, according to the new ICRP recommendation. For the committed dose based on the assumption of 10% ingestion of a mantle with 300 mg of ^{232}Th, Couch and Vaughn [4] report 2.4 mSv which is about 30% higher than our highest value of 1.8 mSv. This difference is due to the lower mass of thorium content in our sample, namely 220 mg. This difference is also seen in the different activity of 33 nCi in Couch and Vaughn [4] work and 25 nCi in this work.

The distribution of thorium in a mantle sample was measured by counting alpha tracks on each CR-39 nuclear track detector. The results of the counting are shown in Figure 5. These results show that thorium is uniformly distributed in the mantle because the track density is nearly equal within one standard deviation from the average.

Table 1. Thorium Content and the Gamma Radioactivity of Various Imported Mantles Arranged According to Their Activity.

No.	Type of Mantle	Mass of mantle (g)	Th% per mantle	Activity (nCi) of origin	Country
1	Butterfly 300 – 400 C.P. No.197	3.283	6.71 ± 0.24	24.5	India
2	Jing Brand 500 – 600 C.P.*	2.888	6.13 ± 0.23	19.7	China
3	Comet Concorde 300 – 400 C.P.	1.789	9.16 ± 0.55	18.2	Italy
4	Comet Concorde 300 – 400 C.P.	1.646	9.16 ± 0.55	16.7	Italy
5	Original Patromax 500 C.P.	2.754	4.22 ± 0.13	12.9	Germany
6	Original Patromax 500 C.P.	2.874	3.58 ± 0.11	11.4	Germany
7	Butterfly 500 C.P.	1.778	3.86 ± 0.14	7.6	China
8	Butterfly 300 – 400 C.P.	1.628	3.85 ± 0.14	7.0	India
9	Butterfly 500 C.P.	1.695	3.62 ± 0.14	6.8	China

*C.P.: Candle Power of the mantle

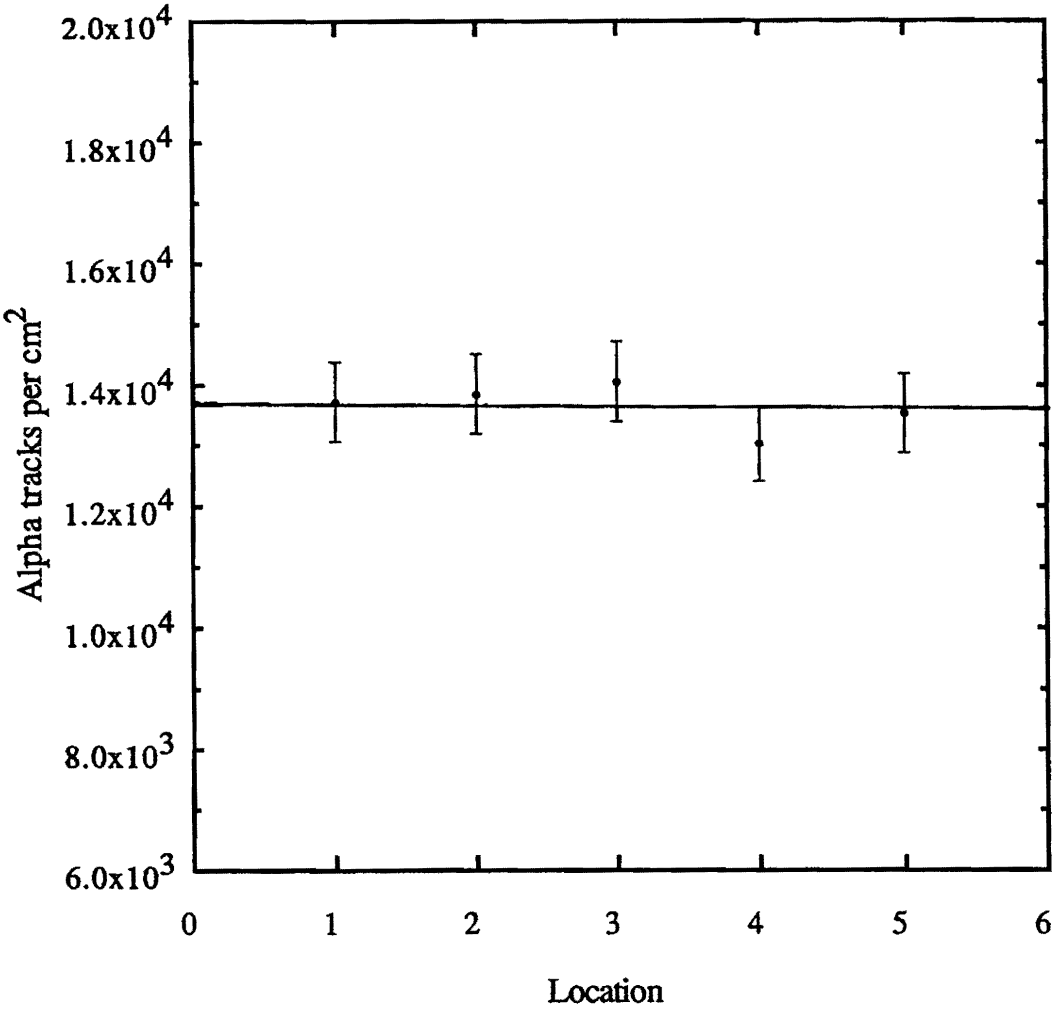

Figure 5. Variation of the Alpha Tracks versus *Position on a Gas Lantern Mantle, Showing Uniform Distribution of Thorium within the Mantle Materials.*

4. SUMMARY

Thorium content and its radioactivity in different types of imported gas lantern mantles in Saudi Arabia were measured using the KFUPM Natural Gamma Rays setup. Similar measurements were performed for thorium standards. The concentrations of Th in weight percent varied from 3.6 to 9.2% depending on the type of mantle. The distribution of thorium in a mantle was studied from the alpha tracks on nuclear track detectors type CR-39. The results showed that thorium was uniformly distributed in the mantle. The total gamma ray radioactivity of each sample was calculated and found to be between (2.4 to 8.8) $\times 10^2$ Bq which corresponds to 6.8 and 24.5 nCi, per mantle. The external gamma dose equivalent rate was measured for one mantle (No. 4 in Table 1) and was found to be 4 μSv/h while it was on the detector surface. The internal hazard of the studied mantles, assuming ingestion of 10% of the ash from one mantle, will give a committed dose equivalent to between 0.5 mSv and 1.8 mSv.

These studies indicate that gas lantern mantles contain natural thorium radioactivity, and therefore present an external and internal hazard for regular users in specific circumstances.

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