CHEMICAL ANALYSIS OF THORIUM CONTENT IN GAS MANTLES

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الخلاصـة :

تمت دراسة محتوى الثوريوم في عدة أصناف من فتائل التريك المستوردة بطريقة كيميائية، كما قيست النسب الوزنية المئوية للثوريوم فيها باستخدام طريقة البلازما الحثية الازدواجية بموجة طولها (٢٨٣,٧٣) نانوميتر، فوجد أنّ تركيز الثوريوم في هذه الفتائل يتروا ح بين (٦,٢٪) و (١٥,٢٪) ويتروا ح نشاطها الإشعاعي بين (٤٧٠) و (١٩٨٠) بيكريل [١٣,٢ الى ٥٥,٦ نانوكوري]. وحسبت الجرعة الاشعاعية المكافئة في ابتلاع ١٠٪ من إشعاعية الفتيل الأقل والأكثر نشاطاً إشعاعياً فوجد انهما (٠,٩) و (٤,٠) ملي سيفرت على التوالي. كما وجُد أنّ كتلة رماد الفتيل المحترقة تنخفض إلى عُشر كتلة الفتيل قبل الاحتراق مع نقصان مماثل في كتلة الثوريوم الذي يَحتويه الرماد.

ABSTRACT

The thorium content of various gas lantern mantles was studied by a chemical method. In this study, the weight percentages of thorium were measured by inductively coupled plasma analysis at a wavelength of 283.73 nm. The thorium concentration in mantles was found to range from 6.2% to 15.2%. Accordingly, the activity per mantle ranges from (4.7 to 19.8) 10^2 Bq (13.2 to 55.6 nCi), and the calculated committed dose equivalent for ingestion of 10% of the lowest and the highest concentration of thorium mantles was found to be 0.9 mSv and 4.0 mSv respectively. The mass of the mantle ash was found to be one tenth the original mass of the mantle with a similar reduction in the thorium mass.

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1. INTRODUCTION

Thorium is used for producing luminescence in gas lantern mantles. Being naturally radioactive, thorium presents a radiation health hazard [1]. Many research groups have studied the thorium content and radioactivity of gas lantern mantles [1–5]. When thorium is extracted and purified from the ore, the ^{232}Th and ^{228}Th activities are in secular equilibrium but none of the daughters remains. However, ^{228}Th, because of its relatively very short half-life (1.9y) compared with ^{232}Th (1.4×10^{10} y), initially decays faster than its production from ^{232}Th, 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], as shown in Figure 1. However, ^{228}Th daughters, because of the shortness of their half-lives, build up and reach equilibrium with ^{228}Th about twenty days after extraction. Therefore, when ^{208}Tl activity is measured it represents that of the daughter ^{228}Th.

In a previous study by Aksoy *et al.* [5], the thorium content in gas lantern mantles was determined by gamma spectroscopy measurements. The natural γ-rays of ^{208}Tl were measured in both mantles and standards. The thorium content was then calculated assuming that ^{228}Th is in secular equilibrium with the parent thorium ^{232}Th. Although γ-spectroscopy is a well established method and the measurements were accurately done, the results of the thorium content were found to be lower than some of the reported data [1, 3]. This discrepancy could be due to the case that ^{232}Th and ^{228}Th were not in secular equilibrium as indicated in the previous study [5]. This triggered the idea of measuring the parent nucleus (^{232}Th) by another method, since it cannot be directly measured by γ-spectroscopy. Measuring total thorium represents ^{232}Th due to the fact that the ratio of ^{228}Th/^{232}Th in secular equilibrium is equivalent to the ratio of their half lives (*i.e.* 1.4×10^{-10}) which is negligible. Therefore, in order to determine the total thorium content in the mantles, inductively coupled plasma analysis method was used. The reduction of thorium in the mantle ash was also determined to assess the risk of the ash and the mantle smoke during initial burning.

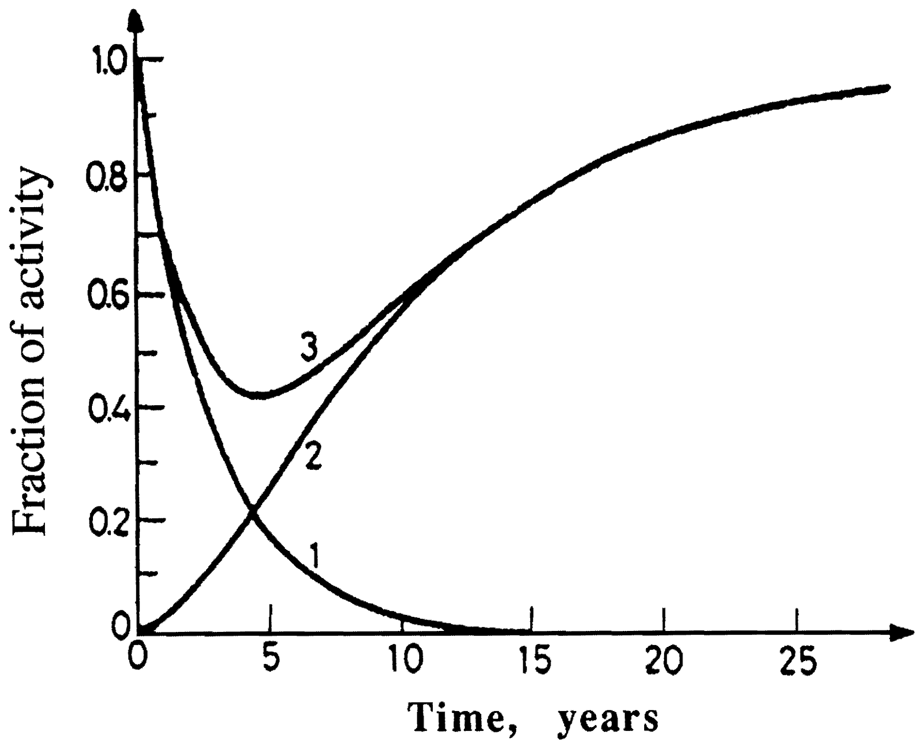

Figure 1. Fraction of ^{228}Th Activities as a Function of Time after Thorium Extraction. Curves: (1) decay of initially present ^{228}Th; (2) growth of ^{228}Th produced by decay of ^{232}Th; (3) effective activity of ^{228}Th [3].

2. EXPERIMENTAL METHOD

Six different brands of gas lantern mantles were collected from local commercial shops. As preliminary experiments, pieces of around 0.2 g each were cut from three mantles of the same brand and carefully weighed using an analytical balance of a sensitivity of 0.1 mg. Each sample was placed in a long neck round bottomed flask and 100 ml of concentrated nitric acid was added. The flask was heated gently until the sample dissolved completely and the solution became clear. After dissolution, the flasks were left to cool and the contents of each flask were transferred quantitatively to a 500 ml volumetric flask and made up to the mark using deionized water. However, after analyzing these samples for thorium, the results showed that thorium is not evenly distributed over the whole mantle. Samples from the same mantle gave incompatible results although the analyses were made by the same method under the same conditions. Another sampling method was then followed in which the whole mantle was treated as one sample, which was weighed carefully, digested in concentrated nitric acid and the resulting solution treated in the same manner as above. After the analysis of four mantles from the same brand it was found that the percentage of thorium in the mantles was consistent with each other, and so this sampling method was adopted for all the mantles collected. Mantles from the same brand were burnt in a lantern for 5 minutes and the remaining ash was collected, accurately weighed, transferred into a 100 ml beaker and boiled with 40 ml of 5% nitric acid. After digestion was complete, the solution was filtered into a 100 ml volumetric flask using Whatman filter paper and made up to the mark using deionized water.

The final solution of each sample was analyzed using a Perkin Elmer inductively coupled plasma emission spectrophotometer model (400 ICP). The spectra were measured at an observation height of 11 mm with an integration time of 100 ms and a photomultiplier voltage of 600 V. A standard solution of 1000 ppm was prepared by dissolving 0.2380 g of thorium nitrate, $Th(NO_3)_4.4H_2O$, in 100 ml deionized water and kept as the stock solution. A 100 ppm working standard solution was then prepared by dilution of 10 ml of the stock solution using deionized water. A series of working standards were prepared from the 100 ppm Th^{4+} solutions by successive dilutions. A calibration curve was constructed using the standard Th^{4+} solutions and emission intensity was measured at 283.73 nm. The solutions of the gas mantle samples were then analyzed for thorium at the same wavelength by taking three replicate measurements for each emission intensity reading. The weight percentage of thorium in each sample was computed from the following formula:

$$\%\mathrm{Th} = (\mathrm{ppm} \times \mathrm{sample\ volume/sample\ weight}) \times 10^{-4},$$

where ppm represents micrograms/milliliter.

3. RESULTS AND DISCUSSION

Table 1 lists the results of the measurements for eight studied mantles. As shown in Table 1, the thorium (^{232}Th) concentration in the mantles ranges from 6.24% to 15.21%. Therefore, the thorium content in the mantles studied varied from a minimum of 119 mg to a maximum of 499 mg. This is almost the same as the thorium (^{232}Th) content reported by Doretti *et al.* [3] which varies from 200 mg to 450 mg.

As shown in Table 1, the concentrations of thorium (^{232}Th) are, on average, double the estimated values of the previous study [5], determined assuming a secular equilibrium of ^{232}Th and ^{228}Th. This shows clearly that the two isotopes are far from secular equilibrium, which explains the reason for the low estimation of thorium (^{232}Th) in the Aksoy *et al.* study [5].

The total activity of thorium per mantle ranges from (4.7 to 19.8) × 10^2 Bq, which corresponds to 13.2 and 55.6 nCi, depending on the type of the mantle. Leutzelschwad and Googins [1] reported activities ranges from 45 to 75 nCi per mantle.

The calculated committed dose equivalent for ingestion of 10% of each of the lowest and the highest content of thorium (13.2 to 55.6 nCi) in Table 1 is 0.9 mSv and 4.0 mSv respectively [4] while the dose limit per year for the general public is 1 mSv, according to the new ICRP recommendations [6]. For the committed dose equivalent based on the assumption of 10% ingestion of a mantle with 300 mg of ^{232}Th, Coach and Vaughn [4] reported 2.4 mSv, which is equivalent to the average dose equivalent of the lowest and the highest content of thorium in this work.

The reduction in the mass of the mantle after 5 minutes actual burning in a lantern was found to be 90%, with a similar reduction of thorium (^{232}Th) of about 86% as shown in Table 2. This means that most of thorium activity is carried out with the air and smoke during the initial burning process. Therefore, it is advisable to take precautions during the initial burning of the mantles to avoid gas inhalation or smoke ingestion.

Table 1. Comparisons of the Thorium Content in Various Imported Gas Mantles Determined by ICP and Gamma Spectroscopy Methods.

Type of Mantle	Mass of Mantle (g)	Th% This Study [*a*]	Th% Previous Study [5] [*b*]	Ratio % [*b*][*a*]
Butterfly 300-400 C.P*. No. 197, India	3.283	15.21 ± 0.16	6.71 ± 0.24	44
Jing Brand 500-600 C.P., China	2.888	11.75 ± 0.16	6.13 ± 0.23	52
'Comet' Concorde 300-400 C.P., Italy	1.789	14.90 ± 0.16	9.16 ± 0.55	61
Original 'Petromax' 500 C.P., Germany	2.754	7.57 ± 0.17	4.22 ± 0.13	56
Original 'Petromax' 500 C.P., Germany	2.874	6.24 ± 0.18	3.58 ± 0.11	57
Butterfly 500 C.P., China	1.695	7.69 ± 0.20	3.6 ± 0.14	47
Butterfly 500 C.P., China	1.778	7.18 ± 0.20	3.86 ± 0.14	54
Butterfly 300-400 C.P., India	1.628	7.28 ± 0.20	3.85 ± 0.14	53

(*a*) ICP analysis in this study; (*b*) gamma-spectroscopy in the previous study [5]

*C.P.: candle power of the mantle

Table 2. Comparisons of Masses and Thorium Content Measured by ICP Method in Various Imported Gas Mantles Before and After Burning.

Mass of mantle (g) [A]	Mass of ash (g) [B]	Thorium content (g) Before burning [C]	Thorium content (g) in the ash [D]	Mass ratio% [B] / [A]	Average thorium ratio % [D] / [C]
2.555		0.172			
2.470		0.149			
2.598		0.159			14
2.547	0.230		0.022	09	
2.490	0.261		0.023	10	

Column A: mass of the mantles

Column B: mass of the ash of the last two burnt mantles

Column C: thorium content of the first three mantles chemically analyzed

Column D: thorium content of the ash of the last two mantles chemically analyzed

4. SUMMARY

The thorium content in different types of imported gas lantern mantles in Saudi Arabia was measured using inductively couple plasma analysis. The concentrations of thorium in weight percent varied from 6.2 to 15.2% depending on the type of mantle. A comparison of these results with a previous study [5] shows clearly that the two thorium isotopes (^{232}Th and ^{228}Th) are far from secular equilibrium. The total gamma ray activity of each mantle was calculated and found to be between (4.7 to 19.8) × 10^2 Bq which corresponds to 13.2 and 55.6 nCi, per mantle. The internal hazard of the studied mantles, assuming ingestion of 10% of the ash from one mantle, would give a committed dose equivalent to between 0.9 mSv and 4.0 mSv.

The reduction in mantle mass during actual burning in a lantern was found to be 90%, with a similar reduction of thorium. Precaution should be taken during the initial burning process to avoid gas inhalation or smoke ingestion from the burnt mantles, as this gas and smoke contains the major thorium activity of the mantle.

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REFERENCES

[1] H. Mohammadi and S. Mehdizadeh, "Re-Identification of ^{232}Th Content and Relative Radioactivity Measurements in a Number of Imported Gas Mantles", *Health Phys.*, **44(6)** (1983), pp. 649–653.

[2] J.W. Luetzelschwab and S.W. Googins, "Radioactivity Released from Burning Gas Lantern Mantles", *Health Phys.*, **46(4)** (1984), pp. 873–881.

[3] L. Doretti, D. Ferrara, and G. Barison, "Determination of Thorium Isotopes in Gas Lantern Mantles by Alpha-Spectrometry", *J. Radioanal. Nucl. Chem. Articles*, **141(1)** (1990), pp. 203–208.

[4] J.G. Couch and K.L. Vaughn, "Radioactive Consumer Products in the Classroom", *The Physics Teacher*, **33** (1995), pp. 18–22.

[5] A. Aksoy, M. Al-Jarallah, and F. Abu-Jarad, "Measurement of Thorium Content and Radioactivity in Gas Mantles", *AJSE*, **22(1A)** (1997), pp. 25–35.

[6] *1990 Recommendations of the International Commission on Radiological Protection*, ICRP Publication 60.

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