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Radioactive Content of Lantern Gas Mantles Used in Night Food Stalls and Camping

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Abstract: Incandescent gas mantles are used for illumination by nocturnal food stalls, camping and other activities. In some countries, thorium-containing gas mantles are available without any indication of the possible radioactive content, resulting in a potential increase of exposure to consumers. In this work, the radioactive content of gas mantles of several brands from Mexico and Spain is studied. Scanning electron microscopy, gamma-ray and alpha-particle spectrometry were used for morphological and radio metrical characterization of gas mantles. The results indicate that some of the analyzed gas mantles contain ^{232}Th and their descendants, with values ranging from detection limits up to 683, 345 and 277 Bq/g for ^{232}Th, ^{228}Ra and ^{228}Th, respectively. The levels of activity concentration of radio nuclides in thorium-containing gas mantles exceed the limits set by International Basic Safety Standards, but still, gas mantles with radioactive content are available in some countries to consumers without any radiological control.

Keywords:NORM, Thorium, Incandescent gas mantles.

1 Introduction

Incandescent mantles for gas lanterns have been used for interior and exterior lighting from the late nineteenth century. Gas lantern mantles may be classified as radioactive and non-radioactive. The presence of radioactivity in gas mantles is due to the content of thorium isotopes as well as of their buildup descendants [1]. The levels of radioactivity, radiological hazards and estimation of radiation dose associated with the manufacture, handling, routine use and disposal of thorium-containing gas mantles have been studied in different countries [2-5].

Radioactive gasmantles are manufactured by dipping inert rayon fibers into a nitrate solution of thorium(99%) and cerium (1%); and for different purposes, traces of beryllium, magnesium, among other elements are also present in the mantles. After pre-burning of the mantle, Th and Ce changes to thorium oxide and cerium oxide, emitting strong visible light when heated to a temperature between1870 and 2370°Cby using gas or kerosene as heat source. During the manufacture of gas mantles, thorium isotopes are separated from their daughters present in the initial ore (for example, ^{228}Ra) and for this reason disequilibrium occurs between members of the same series. Under certain conditions, the age of gas mantles could be determined using the buildup and decay of the daughters in thorium decay series [1]. On the other hand, because thorium is not indispensable for a bright light, non-radioactive gas mantles replace this radioactive element for yttrium or other rare earth elements, however thorium-containing gas mantles are believed to have the brightest incandescence by consumers [6].

Radioactive gas mantles are considered NORM(Naturally Occurring Radioactive Materials), because the radioactive content in these products represents a potential increase in exposure to ionizing radiation to humans [7].In the majority of developed countries, radioactive gas mantles are prohibited or subject to licensing by regulatory bodies, because the radioactive content overpass threshold levels [8].

Despite these regulations, gas mantles declared as non-radioactive have been found in Europe containing ^{232}Th and their descendants, accounting for fraud to consumers and serious environmental pollution problems [9]. In other countries over the world, the use of radioactive gas mantles is performed without control and no special measures are being taken about their handling and disposal after

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consumption. In Mexico, gas lantern mantles are commercially available without any restrictions and have an extensive use in different types of lighting, including illumination of night food stalls and camping activities. Due to the lack of Mexican regulations on NORM, gas mantles are sold without any information of the radioactive content. In this way, gas mantles are handled with low security measures and are discarded as household waste after the useful life, resulting in a potential radiological risk for users, exposed population and environment.

The aim of this work is to characterize radio metrically gas mantles used for Mexican consumers, studying the radiological implications about the use of radioactive gas mantles by night food stalls. In addition, the analysis of the radioactive content in ashes of expended gas mantles used by night food stalls is performed. For comparison purposes, the radiometric characterization of gas mantles commercially available in Spain has been also performed.

2 Materials and Methods

Gas mantles of different brands were collected in different regions; León and Veracruz in Central Mexico, and Seville in southern Spain. In total 9 samples of 5 different brands were collected. The net mass of the samples varies in the range of 0.9 to 3.0 g, as gas mantles presentations come in different sizes for gas lanterns.

In some cases, gas mantles obtained in Mexico do not have any packing and are sold in bulk. Also, the information in the gas mantles obtained in Mexico with packing does not warn about the potential content of radioactive materials (it is only indicated that gas mantles are made 100% silk and recommends throwing ashes directly to household waste).

On the other hand, in the etiquettes of the packing from all the gas mantles acquired in Spain, it is clearly noticed that the products are non-radioactive. More over, ashes of spent gasmantles from several night food stalls in Leon city were collected randomly. Due to the small quantity of sample generated after the consumption of one gas mantle, all the ashes were collected as one composite sample (A-MxFS).

Radiometric and morphological characterization of gasmantles was conducted by three different and complementary techniques: gamma-ray spectrometry, alpha-particle spectrometry and scanning electron microscopy with energy-dispersive X-ray analysis (SEM-EDX).

2.1 Gamma Spectrometry

Several gamma-emitters from the ^{232}Th natural series were measured by non-destructive gamma-ray spectrometry, allowing the determination of ^{228}Th and ^{228}Ra activity concentrations. To determine the activity concentration of ^{228}Ra, the maingamma ^{228}Ac(338 and 911keV) emissions were used. Furthermore, the concentration of ^{228}Thactivity was determined using the weighted average emissions of ^{212}Pb(238.6 keV), ^{212}Bi(727.3 keV) and ^{208}Tl(583 and2614keV), taking into consideration the corresponding branching ratios. The activity concentrations of ^{228}Th and ^{228}Ra obtained through the segamma emissions were determined within1σuncertainties.

The gamma measurements of gas mantles and ashes samples were performed in two different gamma spectrometric systems. The gas mantles samples were measured in aspectrometer system consisting of a Canberra HPGedetector(Hyper Pure Germanium) XtRa (eXtended Range)type, with a relative efficiency of37% and resolution of1.77keVfor the ^{60}Co1.33MeV photo peak.

The XtRa detector has a passive shielding of 10 cm of ancient lead and a 5 mm inner layer of cooper. Additionally, an active shielding formed by a plastic scintillator detector (Bicron B 418) is working in anti-coincidence mode with the XtRa detector. These features make the whole system a very useful tool with lower Minimun Detectable Activity (MDA) than usual HPGe detectors. Measurement times were higher than 3000s for a 95% confidence level and MDA for this system and conditions were in the range 0.15 – 0.25 Bq/g for ^{228}Ra and 0.04 - 0.43 Bq/g for ^{228}Th.

Gas mantles samples were compressed and vacuum sealed in polypropylene containers with cylindrical geometry. The dimensions of the cylindrical containerswere0.92cm height and 3.45 cm in diameter. The hermetically sealed gas mantles were stored for more than 30days before measurements. This was done to prevent the possible escape of radon and ensure secular equilibrium between ^{224}Raand their short half-life descendants.

On the other hand, a second gamma spectrometric system was used to measure ash samples. The system consists of a Canberra well-type HPGe detector, surrounded by a detector of NaI(Tl)anti-Compton of5-inch, connected to six photomultiplier tubes for anti-coincidence counting. The well-type HPGe detector has a relative efficiency of 30% and resolution of 2.3 keV for ^{60}Co 1.33 MeV photo peak. The whole system is in a 10 cm lead shielding to reduce background noise.

The samples analyzed in the well-type detector correspond to ashes of spent gas mantles collected from night food stalls as well as ashes of selected gas mantles burnt out in the laboratory. Because very small quantities of ash samples were obtained after gas mantels consumption (250 mg) special cylindrical vials of 1 cm of diameter were used. The samples were sealed for 30 days before being measured. Measurement times were higher than 90,000 s

for a 95% confidence level. MDA for this system and conditions were in the range 0.12 – 0.19 Bq/g for ^{228}Ra and 0.05 – 0.40 Bq/g for ^{228}Th.

The efficiency calibration in both spectrometric systems was performed by measuring the reference materialIAEA-RGTH1 under the same analytical conditions; measured in the same type of container and same source-detector distance. This reference material is a mixture of thorium ore with floated silicon powder whose activity concentration is well known [10]. Coincidence summing effects were despised due to the use of reference materials with the same radio nuclides of interest.

The bulk densities of the samples showvaluesfrom0.10 to 0.33g/cm^3. The bulk density of the reference material was 1.33g/cm^3.Although there is a significant difference between the composition and bulk densities of samples and reference material, no self-attenuation corrections were required due to the small sample size with little thickness of the cylindrical geometry and also because the energy range of interest ranged from 238 to 2614 keV, what make negligible this matrix effect [11].

2.2 Alpha Spectrometry

The activity concentration of^{232}Thand^{230}Th in gas mantles and ashes samples were determined by alpha-particle spectrometry, after radiochemical treatment and isolation of the thorium isotopes from the sample. Aliquots in the range of 10 -200 mg were taken, depending on the values obtained by gamma spectrometry.

Gas mantles samples were burnt out followed by the application of an alkali fusion process described in [12] using $KHSO_4$(800°Cfor 20 minutes). After waiting a period of cooling, samples were placed in a solution of8MHNO_3. Then,10 mg ofFe^{+3} carrier was added and precipitation of actinides took place by adding drops of concentrated ammonia rising pH to 9. This topes were concentrated in the precipitate, which was separated from the supernatant by centrifugation. The fraction was then dilutedin8MHNO_3 and subjected to the separation process described below.

A process of liquid-liquid solvent extraction using tributyl phosphate (TBP) and xylene was used for the isolation of Th. This Th isolated fraction was the npurified using the ionic exchange resinAG1-X8in order to remove remaining impurities of Uin the Th fraction. Full details of the Th isolation process can be found in [13]

To prepare the alpha particle sources for measurement, the solutions with the isolated Th fraction were electrode positedinstainless steel discs with an area of 15.21cm^2. Electrode position was carried out in cells of polytetra

fluorethylene with a platinum anode. The pH of the solution was adjusted to a value of 2.2 using ammonia vapor. Electrodepositionparameterswere1.5 A of current for 2 hours.

The measurements were carried out using a Alpha Analyst(Canberra) system, equipped with PIPS (passivated implanted planarsilicon) detectors with 450mm^2 activearea. The MDA of the alpha-particle system, considering a95%confidence leveland250,000smeasurement timewasless than0.01Bq/g for ^{232}Th and ^{230}Th.

2.3 SEM-EDX

JEOL 6460LV scanning electron microscope (SEM) with acquisition of digital images in both secondary (SEI) and backscattered (BEI) electron imaging modes (maximum resolution 3.5 nm) has been used for analysis of gas mantles and ashes.

This device was coupled to an EDX microprobe and fitted with an ATW2 beryllium window (resolution 137 eVat 5.9 keV). The semi-quantitative analysis was performed using the Oxford INCA software.

3 Results and Discussion

The activity concentrations of radio nuclides from the ^{232}Th natural series (^{232}Th, ^{228}Ra, ^{228}Th)and ^{238}U natural series (^{230}Th)found in gasmantless old in Mexico(GMx1-6) and Spain(GMEs1-3) are shown in Table 1. Because significant levels of natural uranium are also present in the ores used in the manufacture of the gas mantles, ^{230}Th from the ^{238}U decay series is also concentrated in the original thorium nitrate solution and ended in the gas mantles.

Focusing on the results of the analyzed gas mantles from Mexico, the measurements made by gamma spectrometry and alpha spectrometry show that activity concentrationof^{232}Th, ^{228}Thand^{228}Ra are present in the samples, with ranges482-683, 224-345and240-277Bq/g, respectively.

By computing the activity ratios R_1=^{228}Ra/^{232}Th and R_2=^{228}Ra/^{228}Th, an imbalance between the natural radio nuclides of the ^{232}Thseries can be observed that is due to the separation/isolation of thorium during the manufacture process of gas mantles. Activity ratio R2 = ^{228}Ra/^{228}Th of gas mantles present an average value of 0.80±0.01 indicating that the age since the formation of the gas mantles is higher than 4 years (the activity ratios are near the threshold of the method to determine the age of gas mantles and for that reason the ages cannot be estimated with enough accuracy [1]).

Using a conversion factor for ^{232}Th of 1 Bq/kg = 246 ng/g, an average concentration of 144 ± 21 μg/g of ^{232}Th is estimated in the Mexican gas mantles. With respect to the natural series of ^{238}U, the activity concentration of^{230}Thcoversthe range of62-97Bq/g, fact that implies that

the activity ratios R3 = $^{232}Th/^{230}Th$ of the analyzed gas mantles in Mexico are in the range 6.2 – 8.3.

The activity concentration values reported in Table 1 are in good agreement with those reported in previous studies: for example, activity concentrations of ^{232}Th, ^{230}Th and ^{228}Th in radioactive gas mantles have been determined with ranges 495-697, 50-105 and 219-308 Bq/g, respectively [14]. Considering an average mass of 2.9 g per gas mantle, the activity of ^{232}Th determined in this paper is in the range 1386-1963 Bq/mantle, very similar to the values reported by [5]. A compilation of values of ^{232}Th contained in gas lantern from several countries, may be found in [6] and [9].

Table1. Activity concentration(Bq/g) of radio nuclides from the natural series of 232Thand230 Thin gasmantles of different brands from Mexico and Spain. The uncertain tiesare expressed in brackets for1σ.

Sample code	^{232}Th	^{228}Ra	^{228}Th	Ratio R1	Ratio R2	^{230}Th	Ratio R3
GMx1	500 (24)	310 (6)	249 (10)	0.62	0.80	62 (3)	8.1
GMx2	554 (15)	224 (6)	259 (14)	0.59	0.80	90 (5)	6.2
GMx3	683 (22)	320 (5)	260 (15)	0.47	0.81	85 (3)	8.0
GMx4	665 (26)	345 (4)	277 (15)	0.52	0.80	97 (5)	6.8
GMx5	641 (29)	343 (8)	275 (15)	0.53	0.80	77 (4)	8.3
GMx6	482 (20)	302 (7)	240 (18)	0.63	0.79	71 (4)	6.3
Mean	587 (87)	324 (17)	260 (14)	0.56	0.80	81 (12)	7.3
GMEs1	MDA	MDA	MDA			MDA	
GMEs2	1.37 (0.01)	1.0 (0.1)	0.9 (0.1)	0.55	0.97	0.41 (0.01)	3.4
GMEs3	MDA	MDA	MDA			MDA	
Ratio R1= $^{228}Ra/^{232}Th$, R2= $^{228}Ra/^{228Th}$, R3= $^{232}Th/^{230}Th$							

In Mexico, nuclear regulations about the criteria and limits to consider a solid waste as a radioactive waste, and specifications for the exemption of practices which uses sources of ionizing radiation, clearly establish that naturally occurring radioactive materials (NORM) are excluded from the application of these regulations [15,16]. For this reason, there is not threshold of activity concentration for radio nuclides from the ^{232}Th and ^{238}U natural series contained in products, by-products or wastes classified as NORM.

Nevertheless, if gas mantles are not considered as NORM but as "consumer products", i.e. items that contain radioactive material commercially available to the population without radioactive restrictions, an activity concentration value of 10 Bq/g is established as an exemption level for most of the natural radio nuclides of interest. The results presented in Table 1 are clearly higher than this exemption level so that thorium-containing gas mantles must be subject to regulation or at least declare the radioactive content to consumers.

According to International Basic Standards, exposure due to materials containing radio nuclides of natural origin at an activity concentration greater than 1 Bq/g for any radionuclide in the ^{238}U or ^{232}Th natural series is considered a planned exposure situation, and actions should be taken to control exposure of population and prioritize the use of the material when an exemption level of 10 Bq/g is exceeded [8].

In this context, the values of activity concentration to all radio nuclides from the uranium and thorium natural series found in the Mexican gas mantles are much higher than this value, and the requirements set by these standards for consumer products should be applied. In this case, the gas mantles must provide a label establishing that contains radioactive substances, the corresponding authorization by the regulatory body and options for disposal after use to optimize protection and safety of public and environment.

On the other hand, the analyzed gas mantles from Spain present values of activity concentration of ^{232}Th, ^{228}Ra and ^{228}Th in the range of MDA-1.37, MDA-1.0 and MDA-0.9 Bq/g, respectively. The activity concentration of ^{230}Th covers the range MDA-0.41 Bq/g. Although the values of activity concentration of ^{232}Th are close to zero in the majority of the Spanish gas mantles, in one of them (GMEs2), the activity concentration of ^{232}Th is slightly above the threshold value of 1 Bq/g. Also, the packing of this sample clearly claims that is non-radioactive, but some radioactive content is found in the gas mantle.

In Table 2, the activity concentration of natural radio nuclides found in ashes of gas mantles collected from the night food stalls in Mexico (A-MxFs) as well as ashes obtained after combustion in the laboratory of selected gas mantles from Mexico (A-GMx6) and Spain (A-GMEs1) are presented.

The values obtained from the ashes collected directly from the food stalls have lower values than those obtained in the ash after combustion in the laboratory. One possible explanation for this result is that the ashes collected from

the stalls correspond to a mixture of different brands that include both thoriated and non-thoriated gas mantles, therefore, thorium contained in the ashes corresponding to radioactive gas mantles is diluted in inert ashes from non-radioactive gas mantles. The ashes of the Mexican mantle burnt in the laboratory poses higher activity concentrations of Th and Ra isotopes than the original raw material, indicating its concentration due to burning process.

Table 2. Activity concentration(Bq/g) of radio nuclides of the natural series of 232Thin ashes of gasmantles collected in night food stalls from Mexico, as well as ashes obtained after combustion in the laboratory of gasmantles from Mexico and Spain. The uncertainties are presented in parenthesesto1σ.

Sample	Description	^{232}Th	^{228}Ra	^{228}Th	^{230}Th
A-MxFS	Ashes of gas mantles collected in Mexican late night food stands	1.12 (0.04)	0.38 (0.05)	0.97 (0.1)	0.22 (0.01)
A-GMx6	Ashes of sample GMx6	1071 (72)	2014 (47)	1585 (37)	118 (11)
A-GMEs1	Ashes of sample GMEs1	MDA	MDA	MDA	MDA

Fig. 1: Gas mantles used in night food stalls without indication of radioactive content in the packing Potential radioactive contamination of food may pose a radiological risk to consumers.

The results of activity concentration of natural radio nuclides found in the ashes collected from the food stalls pose a potential radiological risk to exposed workers and food consumers. Exposure ways to radioactive content present in gas mantles are given by external irradiation or possible inhalation and ingestion, because radioactive ashes may be dispersed in food or heating plates (Figure 1)

In order to have information on the main elements as well as the morphological distribution of radioactive content in the different samples of gas mantles and ashes, scanning electron microscopy was applied. In Figure 2 are shown SEM images obtained for selected gas mantles (GMx5 and GMx6) and ashes of the same samples after combustion (A-GMx5 and A-GMx6). The semi quantitative analysis of characteristic X-rays provided by the microscope is used to identify the major elements in points of interest.

Brightest areas in the images are associated with high atomic number elements. In the BEI mode, images of the GMx5 and GMx6 samples, the fibers of gasmantles (less bright areas) can be observed covered by thorium fragments of variable sizes (brightest areas). Punctual X-ray analyses in less bright areas reveal that carbon is the major component(>50%), followed by O(>40%) and Th (<10%) (percentages correspond to relative weight). In the brightest areas of these samples, thorium is the major component(>60%) followed by O(<20%), C(<20%) and Fe(<5%), indicating that bright areas in these samples correspond to radioactive content (thorium oxides) in gas mantles.

After combustion of these samples, the SEM analyses to ashes (A-GMx5 and A-GMx6) shows that the bright areas are dominant, indicating that the Thorium (>60%) is concentrated in the ashes after combustion of the gas mantles, consistent with the results obtained by gamma-ray and alpha-particle spectrometry.

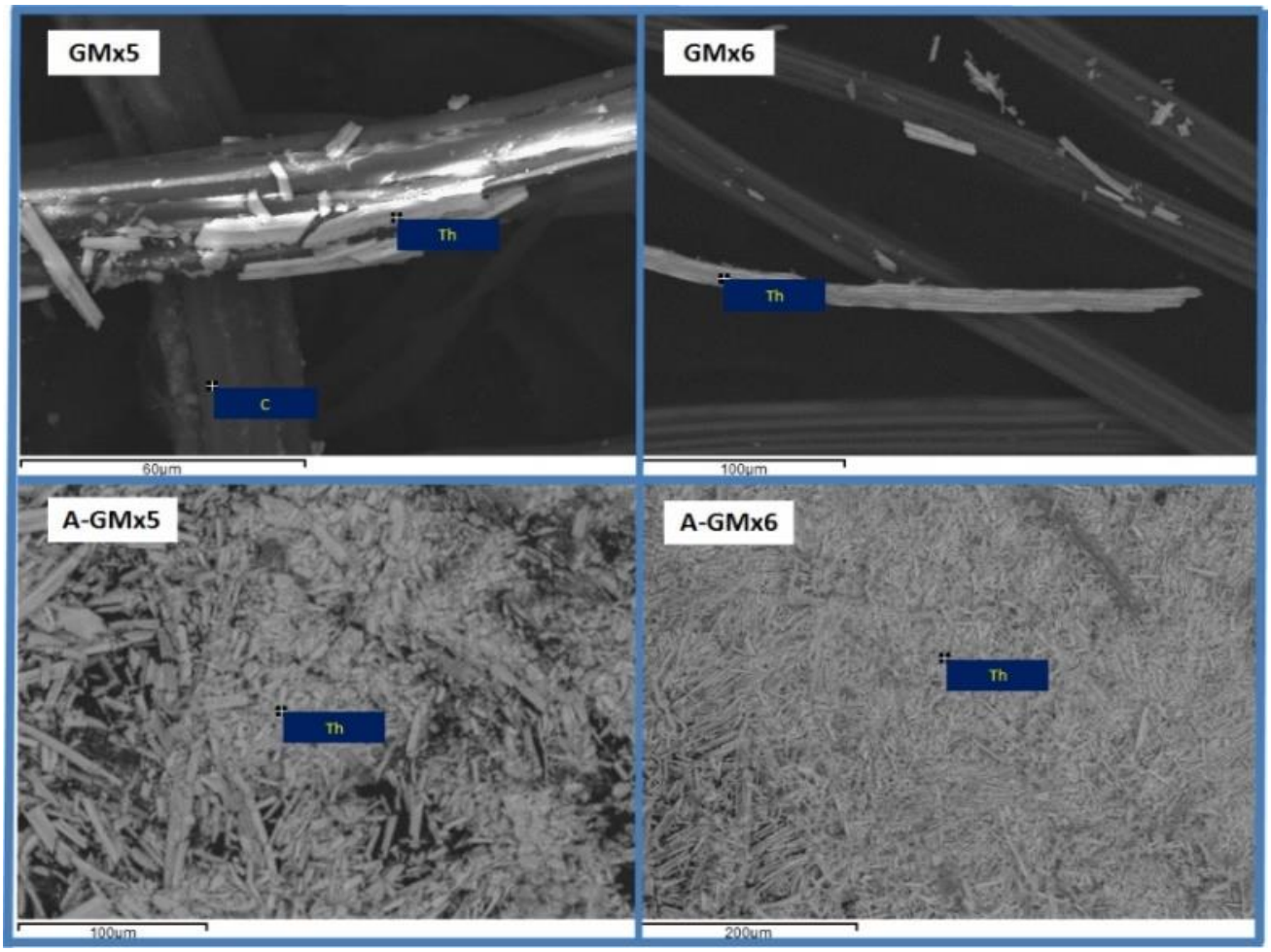

Fig.2: SEM images in backscattered electron imaging mode (BEI) ofGMx5andGMx6 samples and theirashes(A-GMx5 andA-GMx6).

A SEM analysis was performed also in a non-radioactive gas mantle (GMEs1). Semi-quantitative analysis of SEM-EDX in points of interest indicates that brightest areas correspond to Zr (>40%), Y (>18%) and Ce. In this case, rare-earth elements were dominant in the brightest areas, as expected in a non-radioactive gas mantle. In this sample, the Th does not appear as a major element in bright areas.

In Figure 3the images obtained from the ashes collected in night food stalls(A-MxFs) and the ashes obtained after combustion in the laboratory of one of the mantlesgas from Spain (A-GMEs1) are presented.

In the image corresponding to the sample A-MxFs several elements appear in bright areas. In the semi-quantitative graphics it is observed that O(>30%) and Y(>20%) are the major elements in addition to C(>10%) andZr(>10%), and in minor proportions, less than5%,appearLa, Sm, Mg, Si, P, Sand Zn. These results indicate that the ash collected in the night hawker food correspond to the ashes of gasmantles of different brands. The absence of Thas a major element in the seashes, indicates that some places used mostly gasmantles non-thoriated. However, the results obtained by gamma spectrometry (Table 2) reveal that gasmantl esthoriated are usually used in some night hawker.
The corresponding analysis of the sample A-GMEs1, indicates that Zr (>40%), O (>20%), Y (>15%) and C(>10%) appear as majority elements, and in smaller proportions, less than 5%, appear Ce and Fe. In this sample the Th also does not appear as a major element in bright areas.

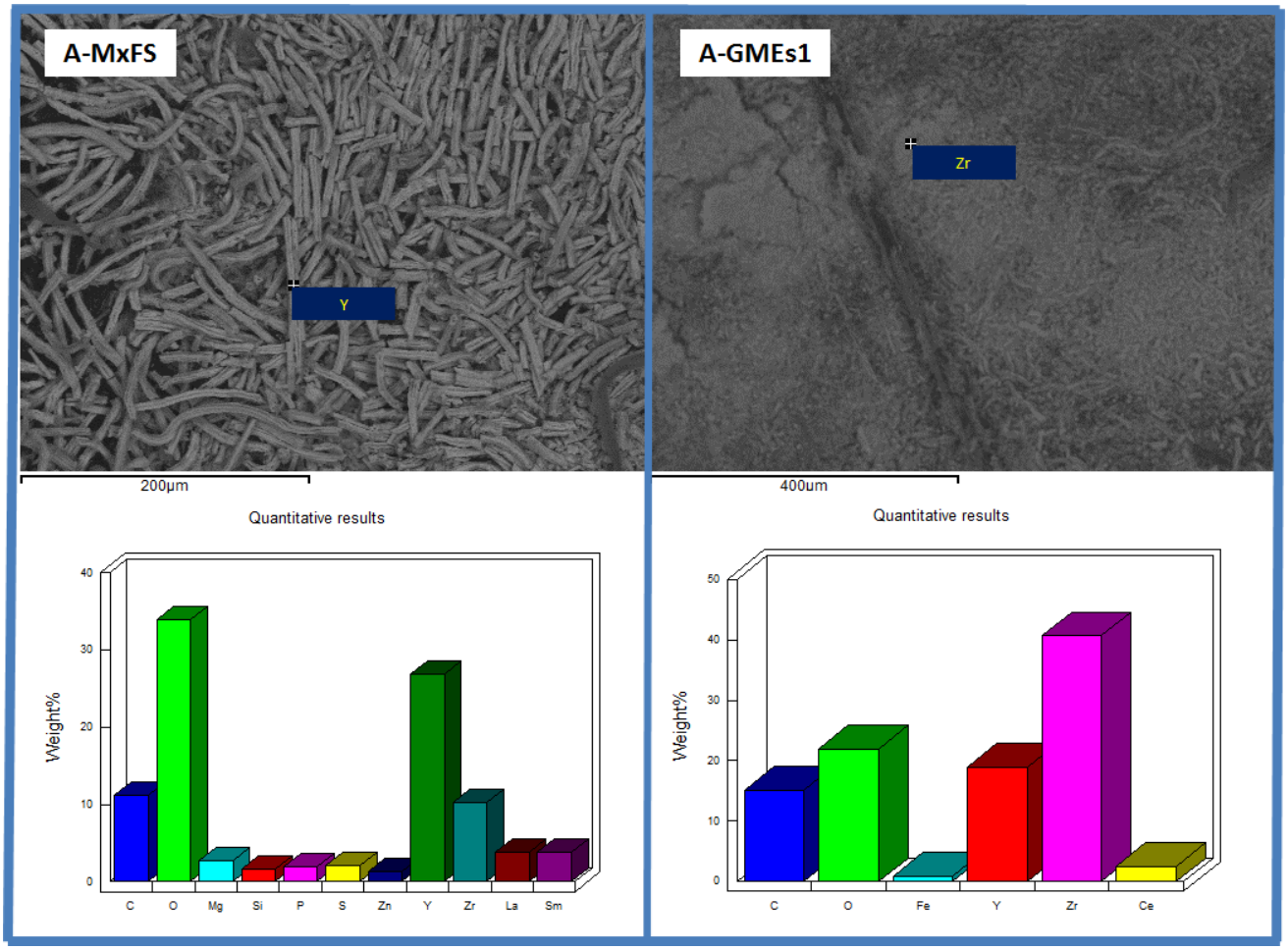

Fig.3: SEM images in BEI mode and semi quantitative analysis of SEM-EDX from samplesA-MxFsandA-GMEs1.

4 Conclusions

Radioactive content in gas mantles collected from Mexico and Spain was determined. Measurements of gamma and alpha spectrometry in analyzed samples, show radio nuclides from the ^{232}Th and ^{238}U natural series with activity concentration values from detection limit up to several hundred of Bq/g. Studied Mexican gas mantles presented very high activity concentration values of ^{232}Th (587 ± 87 Bq/g), in comparison with limits set by international regulations (1-10 Bq/g).

The analysis by SEM-EDX and alpha/gamma spectrometry revealed that in some cases gas mantles are been used by night food stalls without any information to consumers on their radioactive content. Likewise, it was observed that ashes obtained after combustion of radioactive gas mantles concentrate high amounts of thorium and their descendants, representing a potential increase to exposure, due to possible inhalation or ingestion of radioactive content in ashes of gas mantles.

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