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EXPERIENCES IN THE DETERMINATION OF ACTIVITY IN THORIUM CONTAMINATED MATERIALS IN WORKING AREAS, USING GAMMA SPECTROMETER MEASUREMENTS, FROM THE VIEWPOINT OF PRACTICAL RADIATION PROTECTION

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Abstract — In industrial processes, classification and clearance of thorium contaminated material is often a time problem. Short-term decisions have to be made about acceptance limits, industrial safety as the workers are in contact with the contaminated material, and the environmental aspect regarding the disposal of the wastes. The use of gamma ray spectrometry measurements is an easy method to estimate the level of activity in materials. Under the precondition of equilibrium of activity within the thorium decay chain, which can be assumed only with knowledge of the materials' history, adequate results can be produced in a reasonable time. The advantages of gamma ray spectrometry are the low investment and operational costs, simple sample preparation, and a system that is relatively quick and easy to operate.

INTRODUCTION

When carrying out maintenance, dismantling of industrial plants, and the use and disposal of production materials (e.g. welding rods, and electric filaments etc.), it has become necessary to measure radiation in individual working areas caused by thorium and its decay products. According to EURATOM radiation protection recommendations(1), measurements within working areas are necessary if the dose due to natural radioactive materials exceeds 1 mSv.a^{-1}. The external dose exposure from thorium and its products can be classed as being trivial, but measurements of air in working areas (inhalation), and in effluents are of great importance. By measuring the activity of incoming materials, and from the knowledge of their use and processing, an estimate of the dose exposure in the individual working areas can be made. The accuracy of these measurements depends strongly upon a representative sample, its preparation, and the available analysis time. Costs and the number of samples to be measured have a practical influence on the analysing time. With the help of gamma ray spectrometry measurements of air filters and material samples, (dependant upon geometry and nuclides of the thorium decay chain) a minimal detectable activity can be achieved of approx. 3×10^{-3} Bq.m^{-3} for filters, 2×10^{-3} Bq.g^{-1} for incoming materials, and 10^{-2}Bq.cm^{-2} for surface contamination wipe tests. Analysis time is between 1800 and 54,000 s.

MEASURING METHOD

The current method used to identify radionuclides with gamma ray emission is the gamma ray spectrometer with a semiconductor detector, in particular high purity germanium (HPGe) detectors(2). In comparison with a scintillation detector, up to 100 times higher energy resolution can be achieved. The principle is to generate a voltage signal that is proportional to the energy produced by gamma-quant in the semiconductor. Fundamentally a semiconductor detector functions similarly to an ionisation chamber. The average energy necessary to create an electron–hole pair is only 3 eV(2,3). Detailed description of the physics, the calibration and the measuring methods of semiconductor radiation detectors can be found in the literature(4,5).

EQUIPMENT

The Siempelkamp Nuklear- und Umwelttechnik laboratory has two HPGe detectors (n-type and p-type) used for gamma ray spectrometer measurements. The n-type detector with a relative efficiency of 73% is mainly used for the detection of low activity, in gamma ray emitters of low energy (i.e. ^{210}Pb). This detector has a carbon window for gamma ray energy measurements down to approx. 5 keV. Figure 1 shows the gamma spectrometric equipment, the measuring chamber with the detector and the attached power supply with amplifier and AD converter. The software, Gammavision by EG&G Ortec is used for spectrum analysis. Depending upon the state of the sample (density,geometry etc.) and the measuring time, a minimal detectable activity for nuclides of the thorium decay progeny of between 5×10^{-2} and 5×10^{-3} Bq.g^{-1} can be achieved. Table 1 compares minimal detectable activity, in Bq, for wipe tests, air filters and material samples in beakers, for various measuring times. For clearance of contaminated material leaving a controlled area, or to classify con-

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taminated materials for transport, a minimum detectable activity of at least 6×10^{-3} Bq for wipe tests must be reached, which means a measuring time, using the above equipment, of approx. 90 min. An analysed wipe test can be used to calibrate a contamination monitor for direct measurements of large surfaces. If the sample contains not only nuclides of the thorium decay chain, but also nuclides of the uranium decay chain, which is usual with NORM (naturally occurring radioactive material), then the minimum detectable activity will be increased due to a high background. The counting uncertainty will also increase in the individual energy lines.

Table 1. Theoretical detection limits for several geometries calculated by using Reference 6.

Nuclide	Filter/wipe test		Beaker 100 ml 1800 s (Bq)
	54,000 s (Bq)	1800 s (Bq)	
^{228}Ac	2.0×10^{-1}	1.1	6.1×10^{-1}
^{228}Th	2.8	1.5×10	7.5
^{224}Ra	6.7×10^{-1}	3.7	1.9
^{212}Pb	6.8×10^{-2}	3.8×10^{-1}	1.9×10^{-1}
^{212}Bi	6.2×10^{-1}	3.4	1.9
^{208}Tl	1.1×10^{-1}	5.8×10^{-1}	3.2×10^{-1}

SAMPLE PREPARATION

The accuracy of the measurements depends a great deal upon how the sample has been prepared, and for an estimate of activity in a large quantity of material, upon a representative sample. The gamma ray spectrometer equipment is calibrated for various measurements with set sample shapes and sizes. Unfortunately contaminated material is not always in a shape or size that can easily be measured. Figure 2 shows three examples of thorium contaminated material. The left and middle pictures show two examples of material that can be poured into a beaker, giving a reasonably homogeneous and representative sample.

The right picture shows material used in the production of phosphate fertiliser, which is contaminated with a scale containing thorium and radium. For the measurement of activity this surface scale must be removed, milled and then filled into a calibrated geometry. To determine the total activity, it is necessary to estimate the surface area of contamination and thickness of scale. Often the analysis results have to be corrected because they differ in density or geometry from

Figure 1. Gamma spectrometry equipment; 73% n-type HPGe detector with carbon window for low energy γ ray measurement; power supply.

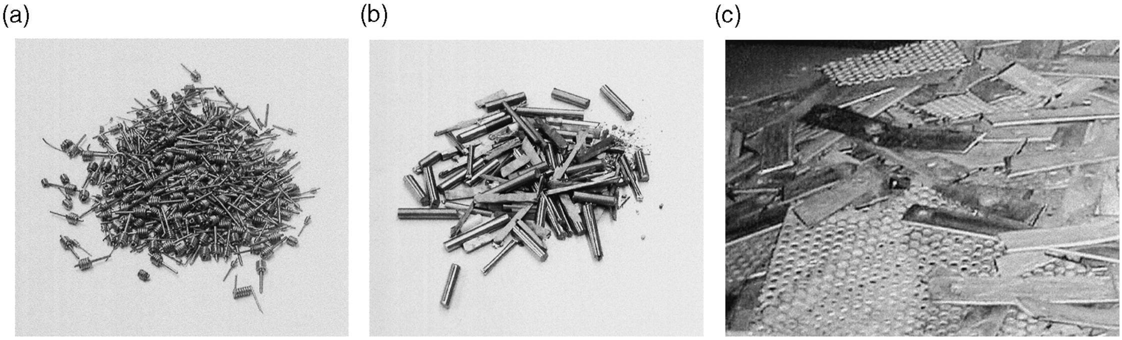

Figure 2. Examples of thorium-contaminated materials: filaments, welding rods, contaminated scrap.

the standard calibration. Insufficient sample material in a beaker must be geometry corrected, using a correction factor related to the filling height of the material. If Marinelli beakers are used for measurements they surround the detector, and the sample material must reach up to the filling mark. If not, a wedge shaped segment can be placed into the Marinelli beaker that reduces the volume of the sample material but not the filling height. This does not effect the efficiency, because there is no symmetrical change in the measurement arrangement. The density can be corrected before the measurement through dissolving the sample in water, or adding a specifically lighter material that is inactive (e.g. polystyrene particles), but the mixture must be homogeneous. Because of the costs and especially the intensive time needed for sample preparation, which is, in the industry, not always possible, the measurements can be corrected after analysing the sample. The software 'Gammatool' by Amersham Buchler (AEA Technology) makes this possible, and takes into consideration the self-absorption inside the sample. Using Gammatool, correction factors for mixtures consisting of different materials can be calculated. These factors are a ratio between the peak efficiency of the calibration source and the efficiency of the mixture at a given density.

Table 2 shows an example for density correction factors, for a sample containing tungsten welding rods. Table 3 shows the density correction factors for the energy lines of ^{228}Ac, ^{212}Pb and ^{212}Bi of a Thorium Network sample$^{(7)}$.

SUMMARY

Measurements of thorium in working areas using a gamma ray spectrometer are accurate enough as long as the thorium decay family is in equilibrium, if not, then determination of ^{232}Th is not directly possible. A prediction of the external dose for the workers can be made by measuring the specific activity and the nuclide distribution of the incoming material. Also it is possible to observe and maintain inhalation limits in working areas, with a high volume aerosol collecter (min. 50 $m^3.h^{-1}$) and measuring times for air filters as shown in Table 1.

Table 2. Density correction factors for tungsten welding rods.

Energy (keV)	Density correction factor*
74.8	24.51
87.2	21.40
238.6	2.61
300.1	1.84
583.2	1.2
795.0	1.12
860.5	1.11
911.2	1.10
967.0	1.10

*$\eta_{\text{calibration source}}/\eta_{(\rho)\text{tungsten}}$.

Table 3. Correction factors for liquid sample of 2nd intercomparison$^{(7)}$.

Nuclide	Energy	p_γ	$Bq.g^{-1}$ before correction	Density correction factor	$Bq.g^{-1}$ after correction	Average* weighted by p_γ
^{228}Ac	911.2	0.2900	10.12	1.085	9.33	
	967	0.1700	10.98	1.083	10.14	
	338.3	0.1200	11.64	1.131	10.29	
	57.7	0.0052	17.93	1.736	10.33	9.77
^{212}Pb	238.6	0.4340	13.48	1.152	11.70	
	77.1	0.1750	14.67	1.417	10.35	
	74.8	0.1180	14.58	1.438	10.14	
	87.2	0.0617	13.37	1.345	9.94	
	300.1	0.0324	11.58	1.137	10.18	11.00
^{212}Bi	727.3	0.0675	12.45	1.091	11.38	
	785.5	0.0109	8.49	1.091	7.79	
	39.9	0.0102	26.58	2.787	9.54	10.73

*average $= \sum_i a_i P_{\gamma,i} / \sum_i p_{\gamma,i}$; a_i = specific activity of nuclide i.

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