

THE MEASUREMENT OF ^{241}Am IN THE BODY WITH A DOUBLE LOW ENERGY GERMANIUM DETECTOR SYSTEM

J.L. Genicot¹, J.P. Culot², F. Hardeman¹, J.P. Alzetta¹, H. Vanmarcke¹

¹ SCK•CEN - Boeretang 200, 2400 Mol Belgium

² A.V. Nuclear Avenue du Roi, 157 1060 Bruxelles Belgium

ABSTRACT

A system based on two low energy germanium detectors has been calibrated for the investigation of internal contamination with ^{241}Am . The two detectors have a total active area of 5775 mm² and a resolution of 600 eV at the energy of 59.5 keV. The measurement system was calibrated with the realistic torso phantom of Livermore which was obtained from the IAEA. The detection efficiency was determined in real measurement conditions with the two detectors above the right lung. Assuming a standard tissue thickness of 25 mm, a detection efficiency of 0.083 cpm/Bq was found. For a counting time of 50 minutes this corresponds to a detection limit of 6 Bq of ^{241}Am . Applying the new ICRP lung model and assuming a typical Pu(MOX) mixture with 1.2 % (w) ^{241}Am , an inhalation corresponding to an internal dose of 20 mSv is measurable 30 days after the intake for a class M compound. In case the americium of the Pu(MOX) mixture can be classified as slow (S), a single intake corresponding to a dose of 20 mSv is detectable over a period of about six months. The subject specific background is evaluated from a background prediction region above the peak region. This system allows a fast response in case of accident. The detectors are also calibrated for the measurement of americium and plutonium in the liver, for uranium and americium in the tracheo-bronchial lymph nodes and for plutonium in the lungs.

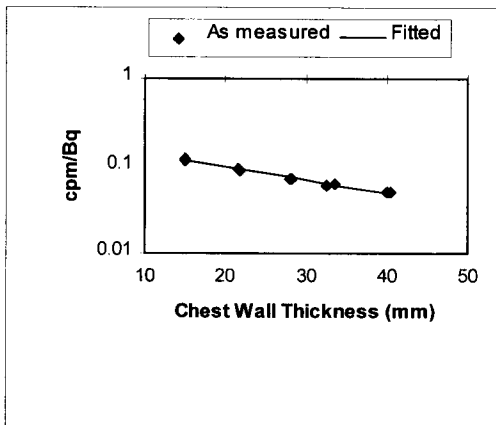
INTRODUCTION

The in-vivo measurement of lung contamination with actinides has always been a difficult task because the low energy X-rays emitted by most of the nuclides are strongly absorbed by the tissues. Since two years, a system using two low energy HPGe detectors has been used in the SCK•CEN for this type of measurements. This system greatly improves the capabilities for the quantitative assessment of actinides in the lungs, the liver and the tracheobronchial lymph nodes. The two crystals having a total active area of 5775 mm² are placed over the right lung and positioned with an angle between them of about 30 degrees. The technique is based on the quantification of ^{241}Am associated with plutonium in MOX fuel particles. The counting room is made of steel plates 200 mm thick lined with thin plates of Pb (3.2 mm), Sn (1. mm) and Fe (0.5 mm).

CALIBRATION

The efficiency of the counting system has been determined with the aid of a torso phantom of the Livermore type (lent by IAEA). The calibration is made for the assessment of ^{241}Am and of pure ^{239}Pu in lungs, in liver and in tracheo-bronchial lymph nodes. Different overlayers were used for the simulation of the different morphological types. These measurements show an influence of the tissue thickness, tissue composition and distance torso-detector on the total efficiency of the system. An increase of 20 mm in the distance torso-detector results in a decrease of the efficiency of about 20%. The efficiency of our system with two detectors for a man with a tissue thickness of 25 mm is 0.083 cpm/Bq. For a counting time of 50 minutes this corresponds to a detection limit of 6 Bq of ^{241}Am . The influence of the tissue thickness on the counting rate is shown in figure 1.

Figure 1. Decrease in counting rate with increasing chest wall thickness of a dual germanium system for the assessment of ^{241}Am in the lungs. The full line is obtained by a least square fitting including all observed values.



ANALYSIS PROCEDURE

A dispersion of 0.2 keV/channel is used for the counting. The examined zone between 59.2 and 60.0 keV, is compared to the zone between 61.4 and 64.2 keV used for the background calculation. The zone chosen for the background assessment is normally free of photopeaks in case of contamination by another radionuclide so that false negative results are minimized. Uranium, in case of contamination with mixed oxide fuel particles, because of the relative proportion of each element, will produce a negligible contribution in the 63 keV region as compared with ^{241}Am .

APPLICATION

A common bioassay tool for Pu(MOX) workers is chest counting for the ^{241}Am progeny of ^{241}Pu . An estimation of the intake requires working assumptions about the isotopic composition of the inhaled mixture and the lung's clearance dynamics. Many factors have to be considered:

- Particle size of the material: From a recent survey of the literature (1) an AMAD of $5\ \mu\text{m}$ and a median geometric standard deviation of 2.56 are realistic default values for occupational exposure when the particle size distribution is unknown.
- Chemical form of the source material: The ICRP-30 and ICRP-68 designate ^{241}Am as class W or class M material. However, a minor contaminant of a matrix may exhibit the behaviour characteristics of the matrix. It could be assumed that the ^{241}Am is of class Y or S such as the plutonium in the Pu(MOX) matrix.
- Metabolic retention and dosimetric model: The new ICRP-66 lung model and ICRP-67 systemic model for Pu and Am provide a basis for the retention and dose assessments. For the density, ICRP-68 uses a default value of 3. In this work, a more realistic density of 10 is used, keeping all the other parameters of ICRP-68 (e.g. breathing rate of $1.2\ \text{m}^3/\text{h}$, resting for 31.3 % of the time and light work for 61.7 %, size factor of 1.5).
- Isotopic composition: In this work, a standard isotopic composition (in weight) is assumed: 1.2% ^{238}Pu , 61.1% ^{239}Pu , 23.5 % ^{240}Pu , 8.8% ^{241}Pu , 4.2% ^{242}Pu and 1.2 % ^{241}Am . The doses due to all isotopes of the mixture vary with the solubility class when 1 Bq ^{241}Am is inhaled:

	$1\ \mu\text{m}$ (mSv/Bq(^{241}Am))	$5\ \mu\text{m}$ (mSv/Bq(^{241}Am))
Class M	0.23	0.13
Class S	0.20	0.11

Defining $Q(20)$ as the measured activity in the lung that gives an effective dose of 20 mSv when

we assume the above-mentioned isotopic composition of the mixture:

$$Q(20)(\text{Bq}) = (20 \text{ mSv/Dose coefficient (mSv/Bq)} (^{241}\text{Am})) \cdot \text{Fraction remaining in the lung.}$$

We found:

	²⁴¹ Am class S (Bq)	²⁴¹ Am class S (Bq)	²⁴¹ Am class M (Bq)	²⁴¹ Am class M (Bq)
Time (d)	1 μm	5 μm	1 μm	5 μm
0.5	16.4	12.9	12.8	9.8
1	16.0	12.5	12.4	9.5
2	15.6	12.2	12.1	9.2
3	15.5	12.1	11.9	9.1
4	15.3	11.9	11.8	8.9
5	15.2	11.8	11.6	8.8
6	15.1	11.7	11.5	8.6
7	14.9	11.6	11.3	8.5
15	14.0	10.7	10.2	7.6
30	12.6	9.4	8.5	6.2
45	11.6	8.5	7.3	5.2
90	9.8	7.1	4.9	3.5
180	8.3	6.0	2.7	1.9
360	6.9	5.0	0.9	0.6

This table shows that, for class S and an AMAD of 5 μm americium, an effective dose of 20 mSv could be assessed up to 180 days after an acute intake (inhalation), when the detection limit is 6 Bq. The situation is less favourable for a class M compound but better for an older Pu(MOX) mixture.

REFERENCE

1. M.-D. Dorrian and M.R. Bailey, Particle size distribution of radioactive aerosols measured in workplaces. *Radiation Protection and Dosimetry* Vol. 60, 119-133 (1995).