Journal of Alloys and Compounds, 213/214 (1994) 484–485
JALCOM 4110

The measurement of incorporated radioactive actinides in the body by direct methods

J.L. Genicot
CEN/SCK, Boeretang 200, B-2400 Mol (Belgium)

Abstract

From the beginning of the nuclear industry the assessment of incorporated radionuclides has been an absolute necessity. Although this assessment is easy for many nuclides (fission or activation products, etc.), the problem of the detection and quantification of some of the actinides is still a challenge. Several of these, e.g. uranium, are easy to detect, but thorium, americium and plutonium are very difficult to assess.

The necessity for the quantification of these elements is dictated by new recommendations of the International Commission for Radiological Protection which propose lower Annual Limit of Intake values, often less than the detection limits of the present measurement systems (proportional counters and "Phoswich" scintillators).

The new technologies are based on semiconductor detectors with large detection areas. Presently HPGe detectors are most appropriate for this application, but surface barrier detectors or room temperature systems (CdTe, HgI_2) are being investigated.

This paper describes the state of the art and the trends of Belgian Nuclear Research Centre in this field. Some examples of measurements are given.

1. Introduction

From the early years of the nuclear industry the International Commission for Radiological Protection (ICRP) has regularly published recommendations for the protection of workers. Among the concepts defined by the ICRP, the Annual Limit of Intake (ALI) specifies the amount of any radionuclide which, when ingested or inhaled, develops in the body the maximum annual permissible dose. These ALI values are frequently revised and have been reduced recently for the actinides. The actinides have played a major role in radioprotection, especially in the direct assessment of body contamination.

First, most of them are α emitters and so the ALI is small. Second, they are very difficult to measure because the γ rays are emitted with very low intensities. Third, the X-rays emitted by their daughters have low energies and are strongly attenuated even by soft tissues. Fourth, these photon emissions are of low abundance relative to the α decays.

It is very important to know the physical properties and metabolism of these elements, which are very different from each other.

2. *In vivo* measurement techniques used at CEN/SCK

In over 35 years the Belgian Nuclear Research Centre CEN/SCK has undertaken more than 60 000 body burden assessments by direct methods. Among these, 2500 lung countings for the detection of plutonium and americium have been made with the aid of a twin proportional counter positioned on the torso. The detection limits are between 300 and 3000 Bq for an acquisition time of 2000 s.

Measurements of wounds contaminated with Pu and Am have been made with NaI(Tl) detectors (130 measurements so far). Since last year a GeHP detector placed near the wound has been used, which allows the identification of isotopes.

3. *In vivo* measurement techniques for the assessment of actinides

The isotopes of plutonium are very difficult to detect. The α decay of ^{239}Pu, for example is detected by the L X-rays of its daughter ^{235}U: U Lα at 13.6 keV, U Lβ at 17.22 keV and U Lγ at 20.2 keV. The total yield of these photons is 3.6% per emitted α and the fractions of photons emerging from the body in the case of a lung contamination are 0.1% for Lα, 1% for Lβ and 4% for Lγ [1].

An important improvement appears when the plutonium is associated with ^{241}Am. The daughter of this isotope (^{237}Np) emits an Np Lβ_1 X-ray (17.75 keV) with an intensity which is an order of magnitude greater. Besides, a γ ray at 59.6 keV is emitted with an intensity of 40%.

SSDI 0925-8388(94)04110-P

Various types of detectors have been developed for the assessment of Pu and Am in the lungs.

Proportional counters are characterized by a good resolution but are not very sensitive to ^{241}Am.

Thin NaI(Tl) scintillators and "Phoswich" detectors [2] have better efficiencies but their resolution is worse than that of proportional counters. A more recently developed system is the gas scintillation type of proportional counter, which shows an improvement in the peak resolution.

All these detectors could be replaced in the near future by semiconductor detectors arrays (HPGe), which present a much better resolution and also a higher counting efficiency in the region of ^{241}Am photons.

A last point to emphasize in the in vivo counting of Pu is that only the lung or liver burden can be detected and then assessed because of the strong attenuation of the photon intensity in the tissues. ^{241}Am, thanks to its γ ray of 60 keV, can also be counted in certain parts of the skeleton (the knee and the skull).

Another group of actinides more easy to detect is uranium. The isotopes of this element or its daughters (63.3 and 92.5 keV emitted by ^{234}Th; 144, 163, 185.7 and 205 keV emitted by ^{235}U) can be detected in the lungs with NaI(Tl) scintillators. However, the acquisition made with a GeHP detector array shows much better results.

The thorium isotopes (^{232}Th and ^{228}Th) have been detected till now with an NaI(Tl) detector by measuring ^{208}Tl (an isotope at the end of the decay chain and emitting a γ ray at 2.61 MeV). The assessment of ^{232}Th and ^{228}Th in the body in this way implies a knowledge of the equilibrium state of the decay chain. The use of a semiconductor detector allows the precise quantification of ^{232}Th and ^{228}Th independently [3, 4].

4. New types of detectors

For many years new types of detectors have been developed and tested for the detection of low energy photons. Silicon surface barriers work well and can be operated on thermoelectric cooling devices. The problem is the number of detectors necessary to reach a low detection limit. The same problem occurs for HPGe [5].

New semiconductor detectors, HgI_2 and CdTe, are now commercially available and have a very good resolution at room temperature [6]. Their surface area is still very small so that the number of diodes necessary for a good detection limit is very high. The types of HPGe detectors best adapted for the detection of Pu and Am isotopes are the large area planar crystals associated in arrays to reach a total area of about 8000 mm^2 (four detectors of 2000 mm^2). These detectors are cooled to liquid nitrogen temperature and their signals must be treated together. In spite of these technical problems, the HPGe array is the most sensitive system today.

5. Metabolism of the actinides

Many observations of accidental ingestion and inhalation of radioactive actinides show that the isotopes of uranium, plutonium, americium and thorium behave in a very complex way [7]. Knowledge of the chemical properties of these elements is necessary to explain the differences in the localization of the nuclides and their translocation in the body.

Thorium oxide, for example, can be detected in the lungs by direct methods without finding any trace in the urine.

6. Conclusions

Many problems in the detection and assessment of actinides in the body are being overcome with semiconductor diodes. Among these, the HPGe array seems to be the best solution for the detection of nuclides for which the new recommendations of the ICRP have reduced the ALI.

References

1 R.E. Toohey, A.T. Keane and J. Rundo, Measurement techniques for radium and the actinides in man at the Center for Human Radiobiology, *Health Phys., 44* (1983) 323–341.

2 G.R. Laurer and M. Eisenbud, In-vivo measurement of nuclides emitting soft penetrating radiations, in *Diagnosis and Treatment of Deposited Radionuclides, Proc. Symp., Richland, WA, 15–17 May, 1967,* Excerpta Medica Foundation, 1968, pp. 189–207.

3 J.F. Colard, J.P. Culot and E. Di Ferrante, Observation de quelques cas de contamination interne par du 75Se, 175Hf et du Th naturel, *Colloq. Int. sur l'Estimation de la Contamination Radioactive chez l'Homme, IAEA-SM-0276/37, Paris, 19–23 November 1984,* pp. 421–432.

4 L. Holmstock, *Thorium and Nuclear Power: Aspects about the Metabolic Behaviour of Thorium,* SCK/CEN, Internal Report, Mol, 1985, pp. 77–81.

5 K.L. Swinth, Status and trends in the external counting of inhaled heavy elements deposited in vivo, *Health Phys., 37* (1976) 641–657.

6 D. Hernandez, M. Righetti and J. Chararay, Medicion de 239Pu y 241Am con detectores de CsI(Tl) y de HgI2, *Colloq. Int. sur l'Estimation de la Contamination Radioactive chez l'Homme, IAEA-SM-276/37, Paris, 19–23 November 1984,* pp. 233–244.

7 N.P. Singh and McD.E. Wrenn, Plutonium concentration in human tissues: comparison to thorium, *Health Phys., 44* (1983) 469–476.