ASSESSMENT OF PLUTONIUM IN HUMAN LUNGS WITH THIN NaI(Tl) DETECTOR SYSTEMS

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Abstract

The development of detector systems for assessment of inhaled Plutonium-Americium dust deposited in human lungs by direct detection of externally emitted low-energy photons, is a continuing programme at Trombay. This paper describes the work done with thin NaI(Tl) scintillation detector systems. The background data inside Trombay steel room in different low-energy bands for crystals of thicknesses 1, 2 and 5 mm are presented. To study the capabilities of three detector systems, each consisting of a set of crystals of the same thickness, a realistic chest phantom of an Indian adult was designed and employed. The chest phantom was constructed from a rib cage of an Indian adult enclosed in a hard polythene cover provided to simulate the chest profile. Measurements on absorption and scattering of low-energy photon (17, 22 and 60 keV) by four constructional materials were made to verify their degree of equivalence to human tissue and granular sugar was used as tissue equivalent material in phantom construction. The counting efficiencies and limits of detection of three detection systems for point sources of plutonium distributed in the central plane of each simulated lung of the designed phantom are reported.

A few normal subjects were counted with one detector system and the increase in background in low-energy region was investigated. The natural radioactivity of the subjects was monitored with a (20.32 cm dia. x 10.16 cm thick) NaI(Tl) crystal in a 50 cm Arc Chair. Finally, the effect of body build of a subject on the counting efficiency of plutonium is commented and our future programme is briefly indicated.

Introduction

At the Bhabha Atomic Research Centre, Trombay, we have an on-going programme of design and development of systems for *in vivo* assessment of plutonium and other transactinide elements deposited in the lungs. This paper (1) presents the results of our studies on the suitability of Trombay-produced thin NaI(Tl) crystals for this application, (2) describes the design and construction of a chest phantom and its use for calibration of the Pu lung monitor, and (3) compares our results with those of other laboratories using NaI(Tl) detectors, as reported in the literature.

Evaluation of Trombay-Produced NaI(Tl) Crystals

The Trombay-produced NaI(Tl) crystals, used in the present study, are of 7.6 cm diameter and 2 and 1 mm thick. They are canned in Al and

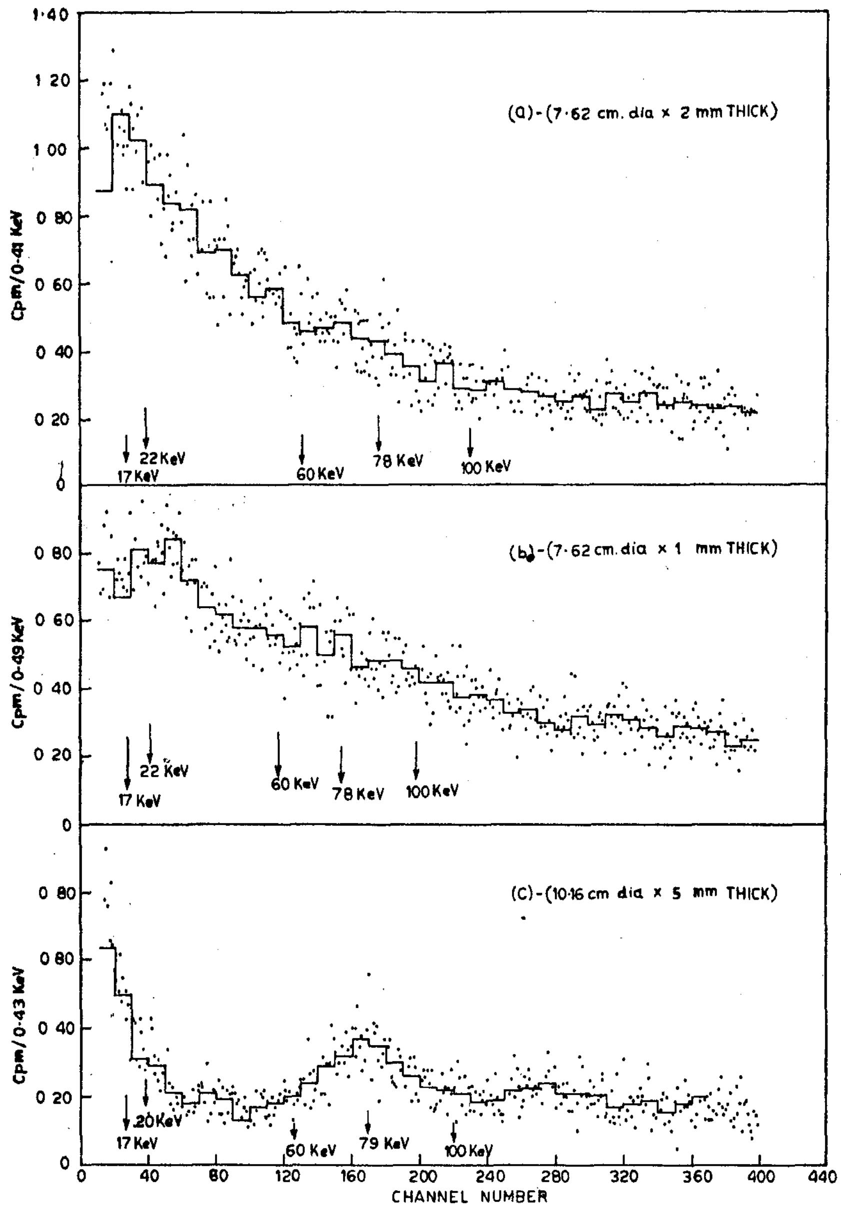

FIG. 1 - A COMPARISON OF THE BACKGROUND SPECTRA OF LOW ENERGY RADIATION INSIDE TROMBAY STEEL ROOM FOR THREE DIFFERENT THIN CRYSTAL DETECTORS. HISTOGRAMS REPRESENT TEN POINT AVERAGES.

are provided with 1 mil thick Al foil radiation entrance window and 3 mm thick glass optical window. They were coupled to selected low-noise Dumont 6363 phototubes and the output from the phototube was fed through an FET pre-amplifier to a 400-channel pulse height analyser fabricated in our laboratory.

A typical pulse height spectrum of a Trombay-produced Pu source, obtained with these crystals, showed that the L X-rays of U are not resolved and exhibited a peak at an average energy of 17 keV. A small peak at 60 keV pointed to the presence of ^{241}Am in the source, the ^{241}Am concentration being about 0.3% of the total Pu alpha activity. The energy resolution at 17 keV was estimated to be about 50%, compared to 46% obtained with a Harshaw integral assembly Model No. 16 MBS 5M/5A Q X (10.16 cm dia x 5 mm thick NaI(Tl) crystal, having 1 mm Be radiation entrance window and 2.54 cm thick quartz optical window) Fig.1 shows the background spectra of low-energy radiations inside the Trombay steel room[1], observed with the three detectors. The histogram depicted represents ten point averages. The Harshaw assembly showed the lowest background rates in the energy bands of interest. These spectral measurements have indicated the aspects in which Trombay-produced crystals require improvement to be suitable for Pu detection[2].

Construction of Chest Phantom

Speight et al[3] had suggested some tissue-equivalent materials for construction of chest phantoms. Due to non-availability of Lincolnshire bolus and Mix D and the handling problems of water, it was decided to test three probable constructional materials, viz. masonite, perspex and sugar, for their degree of tissue-equivalence. The effective atomic number ($\bar{Z}$) and electron density (n) for tissue are quoted as 7.33 and 3.32×10^{23} respectively and for water as 7.42 and 3.36×10^{23} respectively[2]. Perspex and masonite have much higher values of $\bar{Z}$ and n than tissue or water. The half-value thicknesses for water, sugar, perspex and masonite were found as 6.0, 6.0, 7.0 and 5.25 mm respectively at 17 keV, and 14.0, 14.0, 16.75 and 12 mm respectively at 22 keV[2]. The effects of forward scattering by almost equal thicknesses of the four materials as measured by energy shift, degradation of spectral resolution and variation of the ratio of count-rate in the 24-43 keV energy band to that in the 60 keV peak (43.5 - 76 keV) were all found to indicate a similarity of behaviour between water and sugar[2]. Sugar was, therefore, chosen in the construction of the phantom. No attempt was made to simulate the presence of fat in the chest region.

The chest phantom was constructed from a thoracic cage with calavicles, scapulae and shoulder blades taken out of the cadaver of an Indian adult. The whole rib cage fixed at the bottom to a perspex sheet was first enclosed in a frame of thin perspex strips. A hard polyethylene sheet was used to cover the frame to get the chest profile. All measurements were matched to a subject. The vertebrae contained a copper rod to keep them firmly attached. This, however, would not affect the calibration for Pu, since the bone is essentially a dark body for low-energy photons. Two symmetric polyethylene bags were filled with saw dust (density 0.3 g/cm^3), each weighing about 450 g. These were shaped to a human lung and inserted inside the rib cage. Prior to this insertion, point sources of Pu deposited on perspex (1 mm thick both sides) were stuck to a filter paper which was spread longitudinally in the central plane of each simulated lung. The presence of other parts of the respiratory tract and the heart were not simulated. Granular sugar was filled inside the polyethylene enclosure surrounding the rib cage, to provide simulation of tissue. The chest phantom thus constructed had a circumference of 83 cm. The total activity of Pu incorporated in the simulated lungs was 2.5 uCi.

Multi-Crystal Arrays

The phantom described above was employed to obtain the counting efficiencies and the minimum detectable activities for Pu in lungs for a multi-crystal array geometry simulated with a single detector. The multi-crystal array was selected with a view to achieve a large coverage of the frontal area of the chest phantom. The six crystal array chosen is shown in Fig. 2 together with the phantom outline. Each crystal array consisted of detectors of the same thickness. Thus, three multi-crystal arrays were studied to assess the capabilities of these systems and to evolve an array system of Trombay-produced crystals to count suspected cases of internal contamination by Pu and Am.

Table 1 gives the results with our systems and compares them with data of various Pu-lung monitors employing thin NaI(Tl) scintillation detectors as reported in the literature. The systems compared are those of Swinth & Griffins[4], Ishihara et al[5]. The first column lists the various monitor parameters. In the net spectrum of radiation from point sources of plutonium distributed in the central plane of each lung longitudinally of the chest phantom we observed that attenuation of 13.6 and 16.9 keV results in shifting of the 17 keV peak to about 20 keV. The presence of ^{241}Am was indicated by peaks at 29 keV (26 keV + escape) and then at about 57.5 keV. A major conclusion drawn from Table 1 is that Pu lung monitors employing thin NaI(Tl) crystals in different configurations are not capable of achieving the MDA for plutonium desirable for routine monitoring. We achieved the best results when ^{241}Am is used as a tracer for Pu. The use of ^{241}Am is certainly not valid for soluble Pu. Nevertheless an immediate estimate of lung burden of Pu for a subject involved in an accident may be derived on this basis if isotopic composition of the contaminant is known. The MDA quoted for our systems are for very low ^{241}Am (0.5% of total alpha activity) content in Pu.

The MDA quoted for the multi-crystal arrays are all based on the statistical criterion only, i.e. three times the standard deviation in background rate for a given time of counting. Each subject would increase the background in the low energy region of the array, depending upon the level of internal contamination by other radionuclides and the thickness of the detectors employed. For the 5 mm crystal array, we observed that a subject with 125 gm K and 1 nCi ^{137}Cs increased the background by 20 cpm (12-25 keV) region and by 120 cpm in (43.5 - 76 keV) band. ^{137}Cs and K contents of the subjects are determined by whole body counting in 50 cm arc chair using a 20.32 cm x 10.16 cm NaI(Tl) detector. Collection of these data is continuing with a view to derive some useful correlations.

The counting efficiencies and MDA for Pu for the three six crystal arrays reported in Table 1 are thought to be valid for humans having body build and distribution of Pu in lungs similar to that of phantom. Several methods to correct the calibration factor for differing body builds have been proposed[6,7]. We have found it advantageous to use the concept of

Table 1

COMPARATIVE DATA OF VARIOUS Pu-LUNG MONITORS EMPLOYING THIN NaI(Tl) SCINTILLATION DETECTORS

Sl.No.	Monitor Parameters	Swinth and Griffin	Ishihara et al	Our Multi-Crystal Array
1.	Detector size employed and geometry used	Array of 32 NaI(Tl) (2" dia x 1 mm each) four boxes of 13 detectors each. Two boxes positioned over the chest of the reclining subject. [illegible] box edge 2" below the top of [illegible]. The other one on each side [illegible] the armpits	Single (8" x 4 mm) NaI(Tl) on the centre axis of the phantom and at a distance of 1.4 cm from the top plate to the crystal face	(10.16 cm x 5 mm); (7.6 cm x 2 mm); (7.6 cm x 1 mm) Three multi-crystal arrays studied each consisting of a set of six crystals of the same thickness; simulated with a single detector. Detector arrangement relative to phantom is shown in Fig.2. Crystal face in every position remains horizontal with the minimum distance from phantom surface being 1 cm.
2.	Phototubes coupled	Low noise 2" d[illegible] crystal separately	Single 7" dia VMP/II/170 20th Century Electronics without light guide	(10.16 cm x 5 mm) NaI(Tl) : RCA-8055 through 2.54 cm of quartz pipe (7.6 cm x 2 mm) NaI(Tl) : Dumont-6363 through 0.30 cm of glass (7.6 cm x 1 mm) NaI(Tl) : Dumont-6363 through 0.30 cm of glass
3.	Phototube Noise predominance	Below 10 KeV	Below 13 KeV with no light guide	Below 12 KeV for all
4.	Total Sensitive Detector Area	157π cm^2	103.2π cm^2	154.8π cm^2 for 5 mm crystal 57.1 cm^2 for the other two.
5.	Energy Bands used to cover (a) Pu- X-ray 17 KeV photopeak (b) Am-241 gamma	(a) (13.6 to 25.6 KeV) (b) (13.6 to 66.5 KeV) both X-ray and gamma covered.	(a) (16 to 31 KeV) (b) (35 to 73 KeV)	(a) (12 to 25 KeV) (b) (43.5 to 76 KeV)
6.	Energy Resolution at 17 KeV level	-	67.5% with no light guide	*46%, 50%, 48%
7.	Background counts per min	(a) Pu Ch.- 56.2±2.9 (b) Pu+Am Ch.- 204.8±2.8 (In 4" thick Pb shield lined with Cd and Cu)	(a) Pu Ch.- 51.2 (b) Am " - 135.0 (With no light guide) (In shielded chamber 20 Iron + 3 mm of Pb)	*(a) Pu Ch.- 84.0, 204.0, 132.0 *(b) Am Ch.- 105.0, 246.0, 234.0 For six crystal array In shielded chamber 20 cm Iron + 3 mm of Pb.
	With distilled water phantom	(a) 74.5 cpm (b) 388.0 cpm	-	
8.	Calibration phantom and source position	Alderson Remab phantom filled with water. Plutonium (720 ppm Am) mixed with lung equivalent material kept in lung cavities	Phantom of Lucite sheets cluster of pt. sources placed in the model of human lungs	Chest phantom from human rib cage. Tissue equivalent material used is granular sugar. Point sources distributed longitudinally in the central plane of each simulated lung.
9.	Counting efficiency for Pu-239 in lungs counts per sec/nCi	(a) 0.641×10^{-3} (Pu ch) (b) 4.38×10^{-2} (X-ray and Am gammas)	(a) 1.34×10^{-2} (b) -	*(a) Pu ch. 3.83×10^{-3}; 3.46×10^{-3}; 1.65×10^{-3} For six crystal array each.
10.	Minimum Detectable amount of Pu	(a) 49 nCi, (b) 67 nCi — 20 min counting time	(a) 6.2 nCi of Pu-239 (b) - 100 min counting time.	*(a) Pu Ch. 12 nCi; 28 nCi; 34 nCi and 2.44, 10, 8 nCi if Am is used as a tracer. (For Trombay Pu) For six crystal array each and counting time of 100 min.
11.	Contribution from degraded Cs-137 and K-40 gammas	Human subjects add in (a) Pu Ch.- 63.2 to 130.2 cpm (b) Am Ch.- 458 to 798 cpm	A subject with 130 gK and 4 nCi of Cs-137 adds in (a) Pu Ch.- 12.6 cpm (b) Am Ch.- 75.6 cpm	For 5 mm detector array only; a subject with 125 gK and 1 nCi of Cs-137 contributes: (a) Pu Ch.- 20.0 cpm (b) Am Ch.- 120.0 cpm.

*The three successive values are for 5 mm, 2 mm and 1 mm thick detectors respectively.

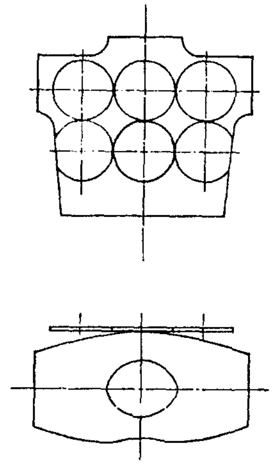

FIG.2-THE MULTI-CRYSTAL ARRAY AND THE PHANTOM OUTLINE

effective tissue thickness (ETT) which is defined as the thickness of a tissue equivalent material that produces for a point source of Pu, the same overall attenuation of X-rays as would occur in a subject having Pu deposited in lungs. ETT takes account of self-absorption of Pu X-rays in the lungs and the attenuation in tissue overlying the ribs. Since our Pu contains traces of ^{241}Am, we determined the variations in the ratio of count-rates in Am and Pu peaks with different thicknesses of overlying water for a point source on the axis of a thin NaI(Tl) crystal at a fixed distance of 10 cm. Calculating the same ratio for the phantom, we found ETT for the phantom as 6.3 cm. This value was also confirmed by the observed shift of the 60 KeV peak of ^{241}Am.

Our future programme of work in this field will include development of phoswich detectors and improvements in the methods of calibration.

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