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SETUP OF AN 8 INCH SANDWICH DETECTOR FOR IN-VIVO MEASUREMENT OF TRANSURANIUM NUCLIDES IN THE HUMAN LUNG

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Abstract

At the Health Physics Division of the Karlsruhe Nuclear Research Center a NaI(T1)-CsI(T1) sandwich scintillation detector system was set in operation for monitoring of persons with regard to incorporation of transuranium nuclides. The report discribes the calibration of the device, for which a makeshift lung and chest phantom as well as point sources were used. In addition several background measurements with and without persons have been carried out. Based on the results of these measruements the lower detection limit for 239 Pu was found to be 6.5 nCi within a measuring time of 50 minutes.

INBETRIEBNAHME EINES 8 ZOLL-SANDWICH-DETEKTORS ZUR IN VIVO-MESSUNG VON TRANSURANEN IN DER LUNGE

Zusammenfassung

In der Abteilung Strahlenschutz und Sicherheit des Kernforschungszentrums Karlsruhe wurde ein NaJ(Tl)-CsJ(Tl) Sandwich-Scintillationszähleranordnung zur Überwachung von Personen auf eine Inkorporation von Transurannukliden in Betrieb genommen. Der Bericht beschreibt die Kalibrierung dieser Anordnung, wozu ein selbstgefertigtes Lungen- und Brustphantom sowie Punktquellen benutzt wurden. Zusätzlich wurden einige Nulleffektmessungen mit und ohne Personen durchgeführt. Unter Benutzung dieser Meßergebnisse ergab sich als untere Nachweisgrenze für ²³⁹Pu ein Wert von 6,5 nCi bei einer Meßzeit von 50 min.

1. Introduction

For incorporation monitoring of the personal at the Karlsruhe Nuclear Research Center measuring devices with high sensitivity have to be available for in-vivo detection of transuranium nuclides in the lung. The in-vivomeasurement of these transuranium elements is difficult because, on the one hand, the maximum permissible concentration levels are very low and, on the other hand, the quantum radiation emitted has a rather low energy, which gives rise to a marked dependence of the counting result on the dimensions of the body. So far, commercial $Ar-CH_4$ proportional counters have been used for this purpose whose sensitivity was not sufficient and which, in addition, did not allow any distinction to be made between ²³⁹Pu and ²⁴¹Am. Although improved $Ar-CH_4$ proportional counters are now available, the excellent experience of many authors with NaI(T1)-CsI(T1)-sandwich detectors and additional pulse shape discrimination has resulted in such a system being commissioned at the Karlsruhe Center. Compared with proportional counters it above all offers the advantage of allowing ²⁴¹Am to be determined separately.

Several methods has been published (10,16) which were used to calibrate such devices by means of volunteers having incorperated suitable K-X-ray emitting isotopes. Nevertheless the use of an makeshift lung- and chest phantom has been prefered for the investigations, because of its simple handling.

2. Description of the Method of Measurement and the Equipment

2.1 Method of Measurement

It is well known that a sufficiently sensitive in-vivo measurement of incorporated transuranium elements requires the use of detectors which have a very low background rate in the energy range of interest while offering the highest possible detection probability. NaI(Tl)-CsI(Tl) sandwich detectors with pulse shape discrimination have turned out to be quite useful units, despite their unfavourable resolution. When using a NaI(Tl) crystal covered with a Be-window of 0.25 mm thickness, the following

E _{X-ray}	Detection Probability n			
keV	0.5 mm NaI(T1)	1 mm NaI(Tl)	2 mm NaI(T1)	
13.6	0.984	0.985	0.985	
17.2	0.981	0.989	0.989	
20.2	0.945	0.988	0.990	
59.6	0.671	0.888	0.982	

absorption factor for vertically incident quantum radiation can be expected:

with $n = \frac{\text{quanta absorbed in NaI(T1)}}{\text{quanta impinging upon Be-window}}$

Tab. 1: Calculated detection probability of a NaI(Tl) crystal

The background of such detector can be reduced to a very large extent by means of an additional detector for Compton quanta arising from the NaI(T1). A CsI(T1) crystal of large thickness optically coupled to the sodium iodide crystal is rather satisfactory for this purpose if the electrical pulses of this sandwich detector are analyzed not only by their magnitudes but also by their rise times. Because of the different optical decay times of NaI(T1) (0.25 usec) and CsI(T1) (1.1 usec) it is possible to distinguish photo-absorption events in NaI(T1) rather satisfactorily from absorption events in CsI(T1) only and from Compton effects in the NaI followed by secondary absorption in CsI.

2.2 Equipment

The sandwich detector used at the Karlsruhe Nuclear Research Center is made up of an 8" dia. x 1 mm NaI(T1) behind 0.25 mm of Be, optically connected with 8" dia. x 2" CsI(T1). This device is contained in a lowradioactivity stainless steel casing and is attached to an RCA 4525 photo-





Fig. 2: BLOCK SCHEME OF THE ELEKTRONIK EQUIPMENT

multiplier by means of a light conductor of non-activated NaI (Fig. 1).

The electrical pulses of this detector, which have different rise times depending on the detection of quanta in NaI(T1) or CsI(T1) and in the two scintillator layers, are fed to a preamplifier (Ortec 113) and then to an amplifier with delay line clipping (Ortec model 460) (Fig. 2). If pulse shaping is used with a delay line, the decay time of the output-pulses is approximately equal to their rise time. These pulses are now converted by a pulse shape analyzer (Ortec model 458) into pulses whose amplitude is proportional to the decay time of the input pulses. This module in addition contains a single channel analyzer which allows the selection from the spectrum of converted pulses offered of those pulses which correspond to a quantum detected in the NaI(Tl) crystal (window-output). In addition it has an output for pulses exceeding the upper threshold of the single channel analyzer (U.L. output), i.e., which have longer decay times than would correspond to NaI(T1) pulses. These two output pulses are used to trigger linear gates, one of which is operated in coincidence with the window output and the other in anticoincidence with the U.L. output. The output pulses of the delay line amplifier (model 460), which are correspondingly delayed (Ortec model 427 A), will pass through the two linear gates only if their decay times correspond to those of NaI(T1) pulses. In this case they are fed to a multichannel analyzer which is setted to analyze an energy range between 10 and 120 keV.

2.3 Pulseshape Analyzer Adjustment

The pulse rise-time of the NaI(T1) detector is independent from the absorbed energy and therefore also from the obtained pulse height. So the adjustment of the pulse shape analyzer, expecially the walk-adjustment, is optimal if the position of the maximum figure of merit obtained with a very small window does not vary with quantum energy. Only in this case it is useful to performe the next step by varying the window width (window middle position as parameter) as shown in Fig. 3.



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3. Lung and Chest Phantom

A lung phantom was prepared on the basis of a model by Ishihara (7) which consists of 1384 plutonium point sources sealed by lucite platelets and arranged one above the other in several rows of plates so as to simulate the shape of the two labes of the human lung.

The activity of each source is 1.047 nCi. In order to take into account even the most adverse case in a measurement of plutonium in the lung, a plutonium solution was selected whose fraction of 241 Pu and hence also 241 Am was as low as possible. This solution had the following isotopic composition on February 2, 1970:

%	wt	< 0.002	Pu-238
%	wt	98.995	Pu-239
%	wt	0.986	Pu-240
%	wt	0.015	Pu-241
%	wt	< 0.002	Pu-242

According to the construction of the lobes by 12 parallel lucite plates each of 4 mm thickness the average density is to high compared to the average density of the human lung.

Since no anthropomorphic phantom was available the ribs, breastbone and vertebratewere simulated anatomically correct by aluminium of 3 mm thickness, and tissue by an uniform layer of paraffin of 2 cm thickness.

Although several Authors (3,8,12,13,18) have recommended these materials to use them for the construction of a human size lung chest phantom additionally absorption measurement have been performed with an 10μ Ci ²⁴¹Am source (see for example Fig. 4).

The obtained half-layers of $0.1559/\text{cm}^2$ for aluminum, 1.05 g/cm^2 for and 1.65 g/cm^2 for paraffin show good agreement with results abtained from Yavin (19).



4. Results

4.1 Detection Properties

All measurements have been carried out inside an steel chamber with a wall thickness of 15 cm, commonly used for body counting. A typical pulse height spectrum of the ²³⁹Pu lung phantom obtained with the sandwich detector (Fig. 5) showes that there is only a single peak at an average energy of 17 keV. The energy resolution was estimated to be 51 % at 17 keV and 20 % at the 59.6 keV peak of ²⁴¹Am. Based on this spectrum and on ²⁴¹Am spectrum and due to the resolutions of the detector the energy range of interest was found to be 14-24 keV for ²³⁹Pu and 40-80 keV for ²⁴¹Am. It can be seen in Fig. ⁵ that the major fraction of the total background rate is in the lower energy range of interest. There is alsoabackground peak at range of about 80 keV due to fluorescence K-X-rays from a thin lead-layer inside the steel shielding. The reduction factor of the background rate (sandwich detector inside the steel chamber) was found to be 3,8 for the energy range. 14-24 keV and 47 in the energy range 40-80 keV. The background rate is given in Table 2.

4.2 Point Source Calibration

The sandwich detector was calibrated to obtain efficiencies and the minimum detectable activities for plutonium content in lung.

The first calibration was made with point sources of 239 Pu (pure) and 241 Am. The sources were placed at a distance 15 cm from the detector surface. For the 239 Pu source (0.5µCi) we obtained an efficiency of 6 cpm/nCi (14-24 keV) for the 241 Am source 57 cpm/nCi 40-80 keV. The distance of 15 cm was chosen to be the real distance between detector surface and middle of the human lung.

The detector efficiency was also derived for a aluminum sealed americium source placed at the rear side of the phantom. In this case we obtained an efficiency of 23 cpm/nCi for 59.6 keV X-rays suitable for a rough estimation of 241 Am content in the case of an incorporation .



PHOSWICH DETECTOR (INSIDE STEEL ROOM)

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The phantom described above was employed to obtain the counting efficiency for ²³⁹Pu LX-rays in lungs. The measurements were carried out in the following positions of the detector above chest:

- a) middline of the complete phantom above the sternum,
- b) sideways over right and left lung, respectively.

For position a) above the sternum we obtained a efficiency of 0.32 cpm/nCi ²³⁹Pu for position sideways over right and left lungs an detector efficiency of 0 38 cpm/nCi ²³⁹Pu. The ratio between position sideways and at the middle line is 1.22. These results show that the position of the detector is more favourable to be sideways over human chest.

Finally we used this system for in-vivo-measurements of the body background from several subjects. Their weigth to height ratio varies between 0.37 and 0.52. The value obtained for energy band 14-24 keV (17 keV average energy of 239 Pu and 241 Am) is 0.226 \pm 0.03 cpm/cm (2) and for energy band 40-80 keV 0.382 \pm 0.08 cpm/cm (2) in the left lung. The right lung values are 0.230 \pm 0.04 cpm/cm for energy band 14-24 keV and 0.406 \pm 0.09 cpm/cm for energy band 40-80 keV, respectively. One can see that higher background counting rates are measured abouve the right lungs of the subjects independent of their body size. These our results have good agreement with the dates form other investigators (2). The subject background for different ratios W/H is shown in Fig. 6. All measurements are carried out with position of arms behind the head. Therefore the comparison between the "in vivo" and phantom efficiencies is based for such a detector-subject arrangement only. The following Table 2 gives a survey about the detection characteristics for 239 Pu and 241 Am.

4.4 Minimum Detectable Activity

Using the lung-chest phantom we calculated the minimum detectable amounts of activity which is possible to estimate with our sandwich system. The minimum detectable activity defined as 2σ of the background rate was 6.5 nCi 239 Pu for a counting time of 50 min inside the steel chamber. Naturally it should be considered, that the minimum detectable 239 Pu lung content is dependent on the body size of the measured subject.



Fig. 6: SUBJECT BACKGROUND DEPENDING FROM RATIO WEIGHT / HEIGHT IN kg/cm MEAS. TIME - 50 min INSIDE STEELROOM (PHOSWICH DETECTOR)

	²³⁹ Pu	²⁴¹ Am
energy region (keV)	14-24	40-80
room background (cpm/cm ² +2 ₀)	0,200 <u>+</u> 0.006	0 .191 <u>+</u> 0 .004
detector area (cm²)	324	324
efficiency with source in air at 15 cm distance from the detector (cpm/nCi)	6.0	57.0
efficiency with source in air at 1 mm distance from the detector (cpm/nCi)	28.0	
efficiency with source of ²⁴¹ Am placedat the rear of phantom (cpm/nCi)		.14-24 keV: 1.83 40-80 keV: 23.00
subject background (50 min counting time (cpm/cm ² +2σ)	left lung 0.226+0.03 right lung 0.230+0.04	left lung 0.382 <u>+</u> 0.08 right lung 0.406 <u>+</u> 0.09
counting efficiency from phantom (cpm/nCi)	0.38	14-24 keV: 1.83 40-80 keV: 23.00
minimum detectable activity (nCi)	6.5	0.2

Table 2: Detection characteristics for ^{239}Pu and ^{241}Am .

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