INVESTIGATION OF THE NATURE OF A CONTAMINATION CAUSED BY TRITIUM TARGETS USED FOR NEUTRON PRODUCTION

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1. INTRODUCTION

The purpose of this paper is to show that the risk of handling targets used for generating fast neutrons in a Van de Graaff accelerator is caused by metal-tritide particles which may be emitted from these targets. It is obvious that this fact, also described by Fehér and Biró (1) who arrive at a slightly different conclusion regarding the dosimetry, constitutes another type of risk than most other cases of handling tritium, where incorporation in the gaseous phase must be considered.

The targets consist of a thin layer of titanium with occluded tritium, on a backing of Cu, Al or Ag. Our investigations started in consequence of a widespread contamination in a Van de Graaff accelerator building, after 40 targets containing an average of 7×10^{10} Bq (varying from 4×10^{10} to 56×10^{10} Bq) of tritium per target had been stored, prepared, transported and applied over a period of 7 years. The total size of the area was 3000 m^2. The degree of contamination over this area varied and there were some 1000 hot spots on floors, work benches and walls. The high level of this contamination was only realized after windowless gas flow counters had been introduced in the area. Faecal and urine samples of persons concerned were taken. Samples of the contamination were collected from the surfaces and auto-radiographed. When microscopic examination of the samples showed solid particles a few were further examined microscopically and by X-ray spectroscope. The presence of titanium and tritium in the particles was established. The method to arrive at the radiation dose after incorporation of the particles is discussed. The procedure of removing the contamination and the steps taken to prevent further contamination is not mentioned here.

2. DETECTION METHODS AND RESULTS OF MEASUREMENTS

The contamination check of the area was carried out using windowless gas flow counters. The total activity found was 3.7×10^7 Bq, distributed unevenly over the contaminated surfaces, with maxima of up to 5.8×10^5 Bq measured over surfaces of 1 dm^2. The derived working limit for surface contamination by low energy beta emitters is 37 Bq/cm^2 for low activity areas (2). For more detailed testing some radioactive material was collected from the surface on adhesive tape by covering a contaminated spot with the tape, applying light pressure, then pulling the tape off. These samples were auto-radiographed (3) either with Ilford nuclear research plates (K-5) or with liquid emulsion (L-4). After an exposure time of up to 8 hours the developed films showed black spots, underneath which solid particles were located after examination under an optical microscope.

In order to test our assumption that the particles had been

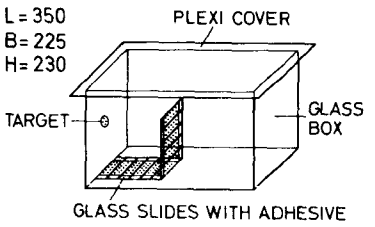

Fig.1

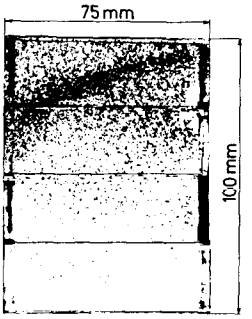

Fig.2a HORIZONTAL

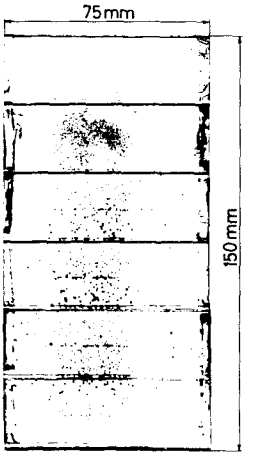

Fig.2b VERTICAL

expelled from the targets a special laboratory experiment was developed.

An unused target was attached to the wall of the box as shown in Fig.1. Microscopic slides covered with an adhesive were put in front of the target, in a horizontal and in a vertical position. After a period of 100 hrs the slides were removed and covered either with a liquid photographic emulsion or with nuclear research plates. The results of autoradiographs taken with nuclear research plates are shown in Fig.2a and b.

The maximum physical sizes of 300 particles, measured microscopically and plotted on logarithmic paper follow a log normal size distribution, with a median diameter by count (CMD) of 10μm and a standard deviation σ_g of 2.2μm (Fig.3). The smallest size detectable by this method is 1μm; no particles with sizes below 3 μm were found. The corresponding aerodynamic diameter (CMAD) is found from the relation CMAD=CDM$\sqrt{\text{density}}$. Using a density of titanium of 4.5 g.cm^{-3} we find CMAD=21 μm; the Mass Median Aerodynamic Diameter, MMAD, is calculated from the relation

$\log \text{CMAD} = \log \text{MMAD} - 6.91(\log \sigma_g)^2$.

We find MMAD = 140μm .

The presence of tritium in the particles was shown with the aid of a specially built, very small windowless gas flow counter with an opening of 2 mm diam. This was placed over a single particle by fixing the detector to the microscope, adjusting the co-ordinates of the window to that of a particular, isolated large particle. The β-spectrum thus measured corresponds to the spectrum of a tritium calibration source.

The presence of titanium was shown by measuring the characteristic line spectrum of the particles by X-ray spectroscopy as shown in Fig.4; the Ag, Cl and S lines are due to elements in the

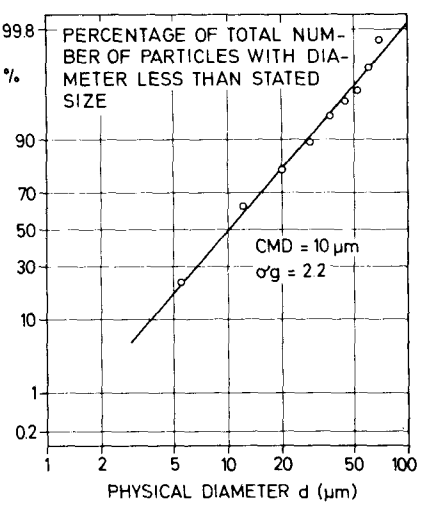

Fig. 3

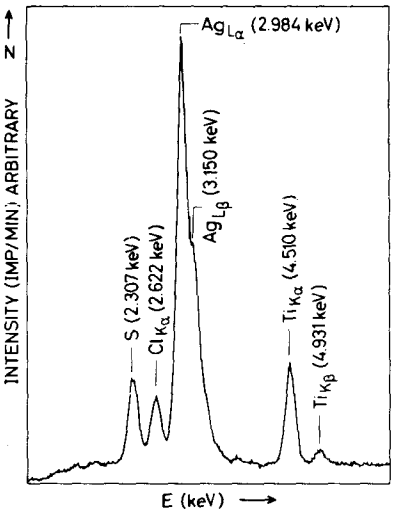

Fig. 4

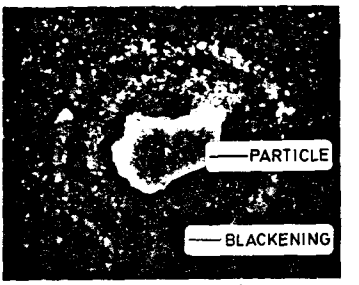

Fig. 5a

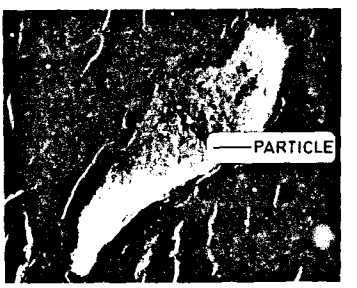

Fig. 5b

photographic emulsion. Fig.5a and 5b are scanning electron microscope pictures of a titanium particle with and without covering by emulsion.

The results of the urine sample analysis did not point to the presence of tritium in the urine. The faecal sample contents from 10 persons analysed during the period 1972-1979 ranged from 1.9×10^3 Bq - 0.4 Bq/24 hr sample. The tritium was unevenly distributed within the samples,which fact was found as each sample -for practical reasons- was divided into 4 parts before the analysis was carried out. The analyses were made by Dr.G.Koch of the S.C.K. laboratories at Mol. The fresh sample was dried under vacuum at room temperature in a disposable nickel crucible, the distillate being collected in a trap cooled with liquid nitrogen. The dry residu was burnt in a combustion train in an oxygen stream and the combustion water was collected in a trap cooled with dry ice. The radioactivity of the distillate and of the combustion water was measured in a liquid scintillation spectrometer. The detection limit for an aliquot of 10 cm^3 water is 1.9×10^{-2} Bq/cm^3.

An estimate at which the particles spontaneously detach from a target was obtained by applying the method described before (Fig.1) to a number of targets. An estimate of rate varied from 20 to 8000 particles per 24 hr.

3. DISCUSSION

Several results of our investigation point to the fact that the contamination consisted mainly of particles. The fact that

tritium was found in the faeces and not in the urine is the first indication. Tritium in gaseous form being inhaled or otherwise incorporated would have been distributed in the body fluids and excreted in the urine. The inhomogeneous distribution of the tritium in the faeces forms another indication. The results of the auto-radiographic analyses of the samples collected in the work as well as in the laboratory experiment confirm the "particle theory",together with the observation of particles by electron microscope. The X-ray spectroscopic analysis confirmed that the particles consist of titanium; the β-spectrum identified the tritium.

The distribution of inhaled tritium-titanium particles in the body organs can be estimated by applying the particle size data to the Dosimetric Model for the Respiratory System (4). According to the large size of the particles the initial deposition will exclusively take place in the naso-pharynx. Those deposited in the pharynx will subsequently be removed with the mucus into the mouth and then swallowed. The nose-deposited particles will either be removed by nose blowing or be swallowed.

The radiation dose to the naso-pharynx will be small because of the short passing time and the low energy of the β-radiation. The same applies to the gastro-intestinal (GI) tract through which the tritium will pass after having been swallowed. The surface cells of this tract are very insensitive for radiation (5). As metal tritides are not taken up into the body from the GI-tract (6) the material will be excreted after having passed this tract.

In order to determine the radiation dose quantitatively the relation between particle size and radioactivity must be better known than yet established. Work on this is going on. It can however tentatively be concluded that the dose must be considerably smaller than in cases where tritium is incorporated in comparitive quantities as a gas and that the dose to the lung in our case will be neglible.

ACKNOWLEDGEMENT

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