

Alpha Spectrum Degradation by PuO₂ Particles

alpha dosimetry

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Introduction

Alpha radiation, originating in the interior of a particle of radioactive material, loses energy before it emerges. The first slide illustrates this concept. Imagine a large PuO₂ particle immersed in water. This is similar to an aerosol particle in the soft tissue of the lung or contamination in a wound.

If an alpha ray is emitted from one of the particles surface atoms, it will travel about 47 microns⁽¹⁾. It deposits all of its energy in the water because it lost none in the particle. However, when the particle is larger than the range of the alpha radiation, some alpha rays will be totally absorbed in the particle. The range of ²³⁹Pu alphas in PuO₂ is about 9 microns. For particles smaller than 10 microns, the majority of the alpha rays will lose part of their energy in the particle and part in the water. This results in a degradation of the alpha spectrum from a PuO₂ particle.

This degradation of the alpha energy spectrum must be known for calculation of the local tissue dose around a particle of PuO₂. In addition, the measured activity of air samples, or other particle samples, can be in error, if the detection method is sensitive to alpha energy. For example, alpha counters, which have windows, are dependent on the energy spectrum, because the loss of counts will vary with the alpha energy.

Experimental Techniques

Rather than attempt calculation of the spectrum degradation, we decided to measure it directly with an alpha spectrometer.

Air samples were drawn from a glovebox where PuO_2 powder work was in progress. The second slide shows our technique. The samples were drawn through a HASL type cascade impactor⁽²⁾, where different particle size fractions were deposited on separate glass slides. The final stage was a membrane filter. The HASL impactor is conveniently used for this work, because the slides can be moved in discrete steps, thus avoiding particle pile up. A Brooks rotameter was used to regulate the air flow provided by a vacuum pump. An absolute filter on the pump exhaust turned out to be unnecessary. The membrane filter effectively stopped all activity.

We made an attempt to confirm the HASL cascade impactor calibration by electron microscopy. Our calibration agreed generally with the published data although we did not take enough electron micrographs for statistical verification. Electron micrography is expensive.

The third slide shows an electron micrograph of a almost spherical 3 micron particle of PuO_2 . The shadow at the right was made by evaporating platinum on the sample at an angle⁽³⁾. The shadow is about twice as long as the particle is high.

The fourth slide shows another 3 micron particle. The shadow on this particle, however, is quite short. The particle is shaped like a flake. It is interesting to note that both this particle and the preceding one were impacted on the same stage. While they have similar aerodynamic characteristics, the particles obviously have drastically different weights of plutonium.

Slide No. 5 gives an instance of two smaller particles agglomerating into a single particle. The individual particles are one and one and a half microns respectively.

A typical final membrane filter micrograph is shown on the next slide. All particles are smaller than 0.5 micron. Although the number of particles

collected on this final stage was greater, more total activity was found on the upper stages. This is typical of log-normal distributions.

Slides were collected under the same conditions as for the electron microscope examination. They were then placed in a vacuum bell chamber and the alpha energy spectrum measured with an Ortec silicon diode detector. The pulses were fed through an Ortec preamplifier and linear amplifier to a Nuclear Data 256 channel pulse height analyzer. Resultant spectrums for 0.5, 2 and 10 micron MMD size fractions are shown on the seventh slide.

Alpha Spectrum Degradation

The dotted line shows the spectrum from a so-called "weightless" source. These weightless sources are used to measure the resolution of alpha spectrometers. They are fabricated by evaporation from methanol. This produces a full width at half maximum peak height of 16 KeV for the ^{239}Pu spectrum.

The solid line shows the spectrum from the final membrane filter. All particles are less than 0.5 micron in diameter. The spectrum is only slightly degraded from the weightless source. ^{239}Pu alphas have 5.15 MeV of energy.

The two other spectra are for particles of 2 and 10 microns mass median diameter. The HASL impactor is calibrated for the MMD of the particle size distribution. This is appropriate for hazard analysis since half of the activity will come from particles larger than this size and half from those smaller.

The 2 micron fraction shows definite degradation of the spectrum. Fifty-five percent of the alphas have been degraded to energies below 5 MeV. About 25% of the alphas from the 10 micron fraction are degraded below 4.5 MeV.

These spectra are all normalized to represent identical activities and have been put into histogram form for ease in calculating local tissue doses.

This degradation of the alpha spectrum by the PuO_2 aerosol particles themselves must be evaluated when pulse alpha spectroscopy is used for air samples. In a recent article⁽⁴⁾, Lindeken and Petrock noted a displacement

of energy peaks to lower channels even when the sample was collected on a membrane filter. They ascribed this energy degradation as being inherent to the filter medium. The previous electron microscope pictures showed that the particles sat free and clear on the glass slides. We were careful to avoid pile up of the particles and believe that the observed degradation is caused by energy loss in the particles.

This energy loss significantly affects the efficiency of scintillation counters (or any other windowed counter). An energy degradation of 1 MeV will cause the same loss of counts as a centimeter of air or 1/4 mil mylar.

Most of the total activity in an air sample will be in the form of larger particles, because particle mass is proportional to the cube of the radius. If counts are lost because of the inherent energy degradation of large particles, the activity of an aerosol will be underestimated.

It should be possible to estimate the MMD of an aerosol sample by alpha spectroscopy. We have not yet explored this possibility. Care would have to be taken to limit degradation due to inert dust loading, particle pile up and particle burial in the filter media. Short period sampling with a membrane filter and long counting times may overcome these difficulties.

Material hangs up between stages in cascade impactors. The loss has been reported to be as high as 45% of the material drawn into the impactor (5). If material is lost at any point in an impactor, the following stages will have been robbed of material and the absolute efficiencies of the various stages will be in doubt. Alpha spectroscopy can be used to study this perturbation of stage efficiency.

An alpha emitting aerosol can be drawn through a prototype impactor, which is designed to be disassembled. The surfaces, which retain material, can be counted with an alpha spectrometer and the MMD determined. Knowing the MMD of the hung up articles, the impactor designer can modify the impactor

so that the particle size distribution on the stages represent the actual aerosol distribution.

Before more such sophisticated work can be done, the degradation of the alpha spectrum as a function of particle size will have to be known with more precision. The major weakness in our work is the poor particle size selection of available cascade impactors. Each stage has a distribution of particle sizes on it.

An improvement could be made by making use of a technique described recently by Couchman⁽⁵⁾. A sample is drawn through two identical stages in series. The size distribution on the second stage will be normally distributed and have a smaller standard deviation.

Local Tissue Dose From PuO₂ Particles

The hazard from a plutonium particle in the lung or a wound depends on the pattern of energy deposition in the surrounding tissue. The fraction of alpha rays escaping from an absorbing sphere and the average alpha energy emerging has been calculated by Sherwood and Stevens⁽⁶⁾. However, in order to give a detailed description of the pattern of energy deposition, one must know the energy spectrum of the emerging alpha radiation.

The next slide (No. 9) gives the model for the calculation of the local tissue dose from alpha emitting particles. This is similar to that previously described for beta emitters⁽⁷⁾. The energy deposited in a thin tissue shell at any distance from a spherical particle is divided by the tissue shell mass. For alpha radiation one needs to know the number and residual energy of alpha rays which travel a given distance from the particle. This can be calculated by using a histogram of the energy spectrum and the Bragg curve for alphas, shown on the next slide. The increase in specific ionization at the end of the alpha track, partially compensates for degraded alphas which have stopped short of the maximum alpha range.

The last slide shows the alpha dose rate versus the distance in tissue from spherical particles of $^{239}\text{PuO}_2$. The dose rate of interest in the lung is that below the viscous and serous layers of mucous lining the epithelium. These layers have a combined thickness of 15 microns. At thirty microns, dose rates from one micron particles are 100 Rads/hour. The dose rate drops to zero at 47 microns, the maximum range of 5.15 MeV alphas. We show a ^{239}Pu dose rate curve for comparison. ^{238}Pu alphas are more energetic and have an energy of 5.3 MeV.

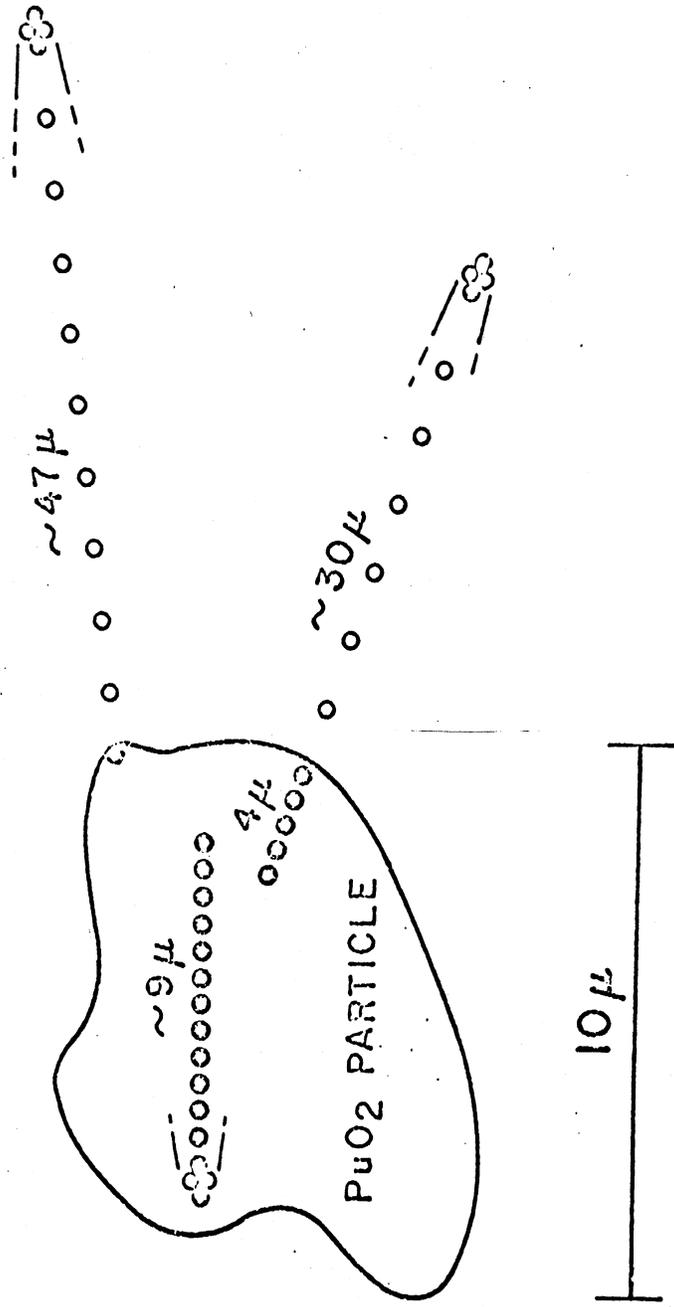
The biological interpretation of this very high local tissue dose needs study. If lung cancer is caused by irradiating an individual cell or small groups of cells to the threshold dose, then a single PuO_2 particle inhaled into the lung might cause cancer. If cancer incidence is a probability function of the average dose to a large number of lung cells then the present ICRP permissible organ burden is correct.

Bibliography

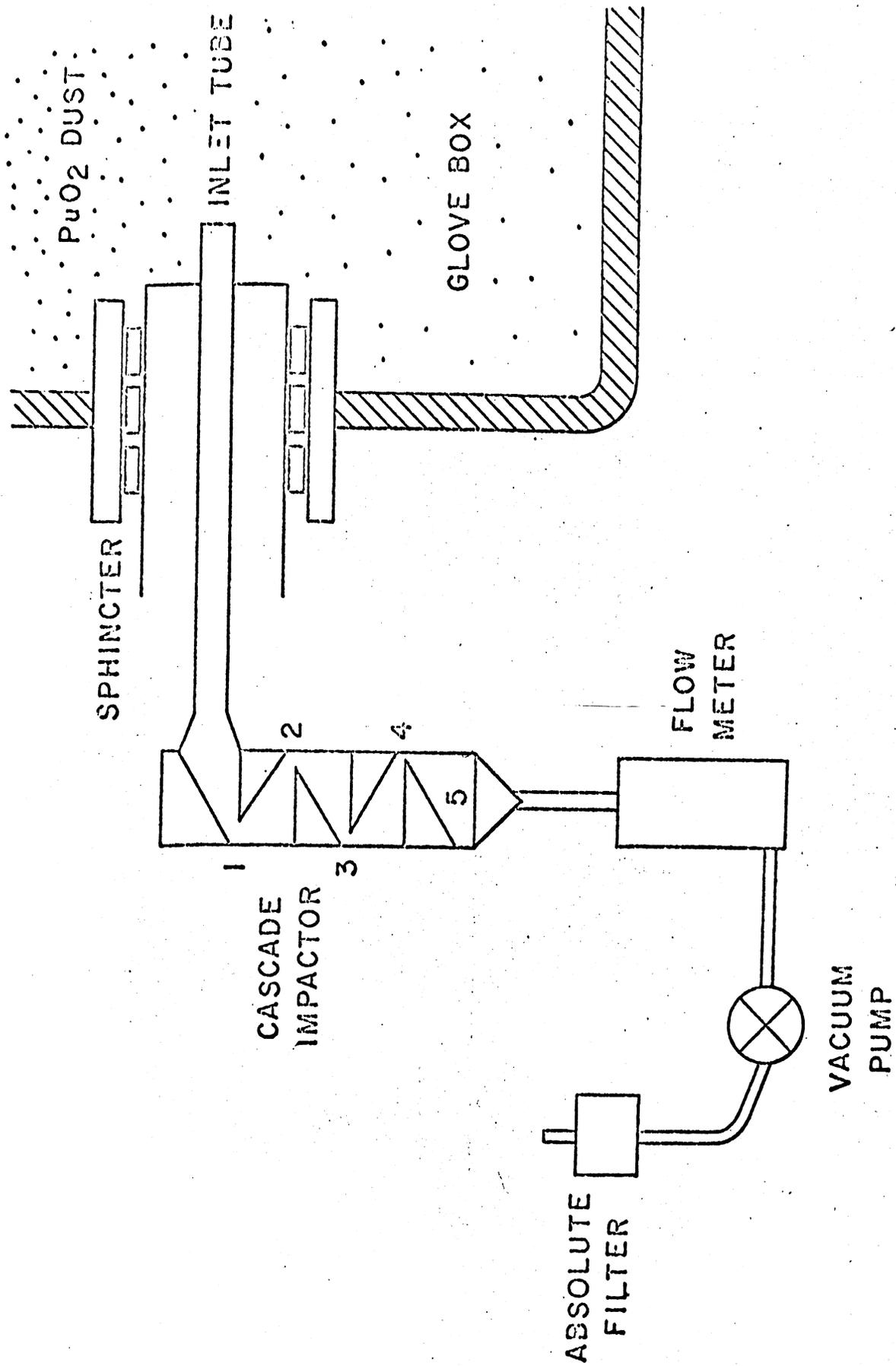
1. Evans, R. D., The Atomic Nucleus, McGraw-Hill (1955).
2. Lippman, M., American Industrial Hygiene Association Journal, 20 (5) 406-16 (1959).
3. Roth, J. and Halteman, E. K., Morphological Characteristics of Co-precipitation (U, Pu)O₂ Powders by Electron Microscopy, NUMEC-2389-5, Nuclear Materials and Equipment Corporation, Apollo, Pa. (1965).
4. Lindeken, C. L. and Petrock, K. F., Health Physics, Vol. 12, (683-91) (1966).
5. Couchman, J. C., "Simplified Method for Determining Cascade Impactor Stage Efficiencies", S-23-P, EG&G, Inc., Santa Barbara (1966).
6. Tyler, G. R., Health Physics, Vol. 12 (509) 1966.
7. Caldwell, R. D., "Local Tissue Dose From Insoluble Radioactive Particles", Proceedings of the 11th Annual AEC Bioassay and Analytical Chemistry Conference, Albuquerque, 1965.

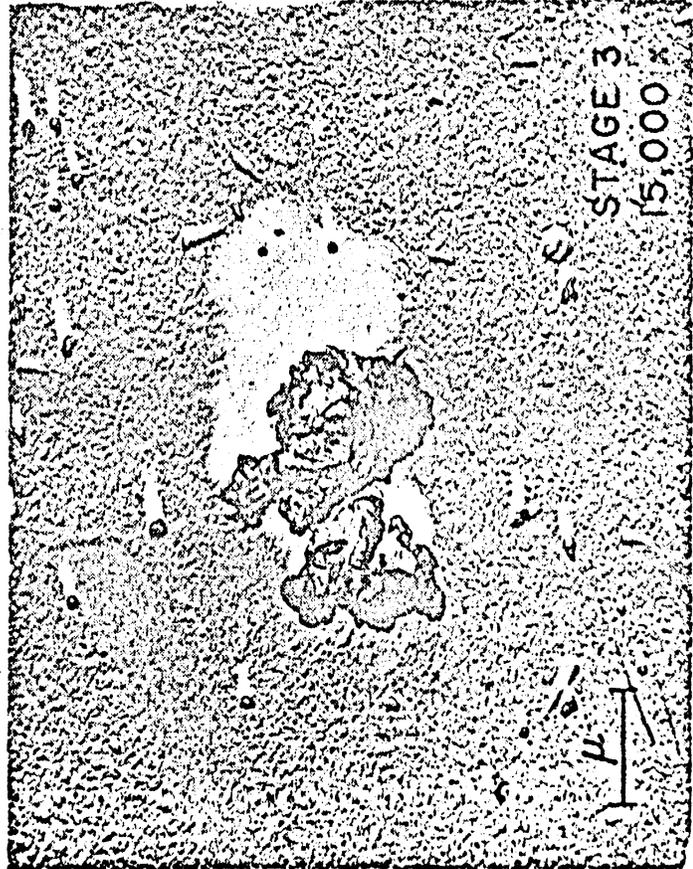
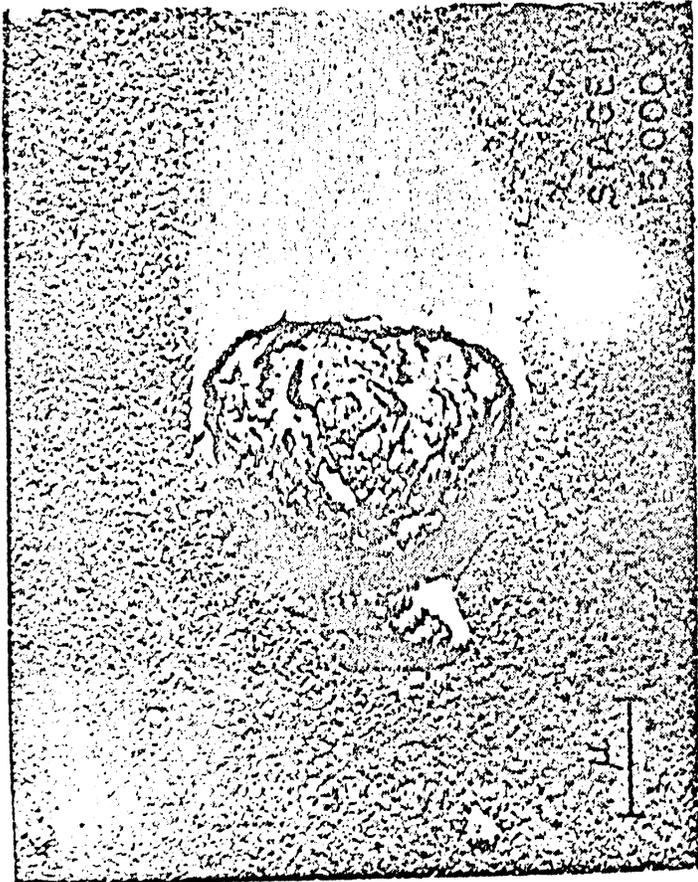
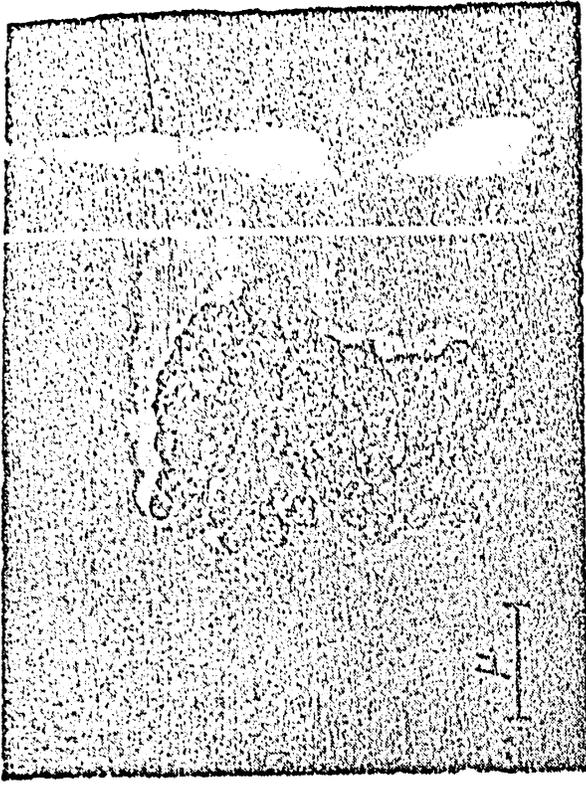
ALPHA ENERGY LOSS IN A PuO₂ PARTICLE

SOFT TISSUE (WATER)

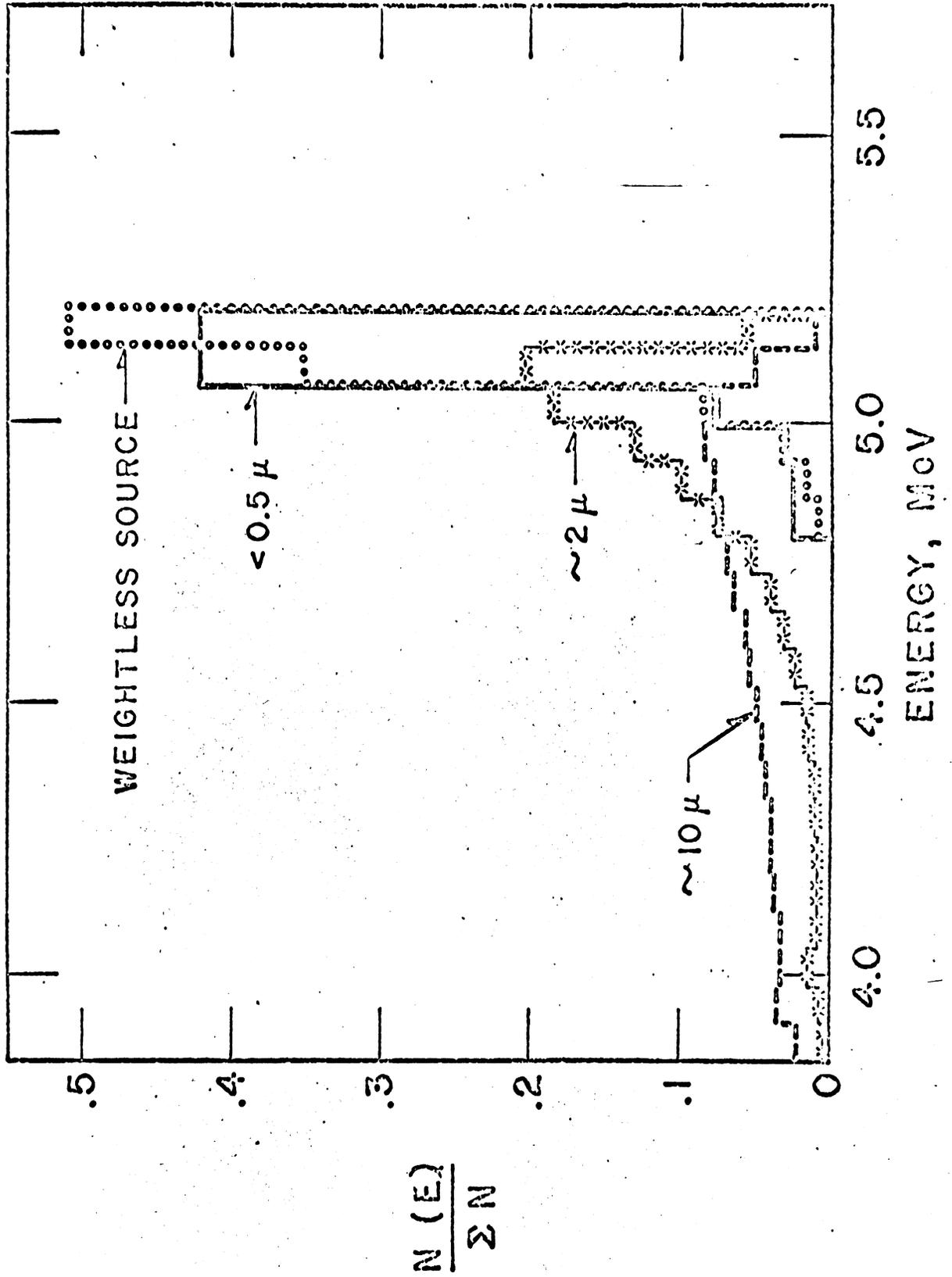


SAMPLING TECHNIQUE PuO₂ PARTICLE SIZE STUDY

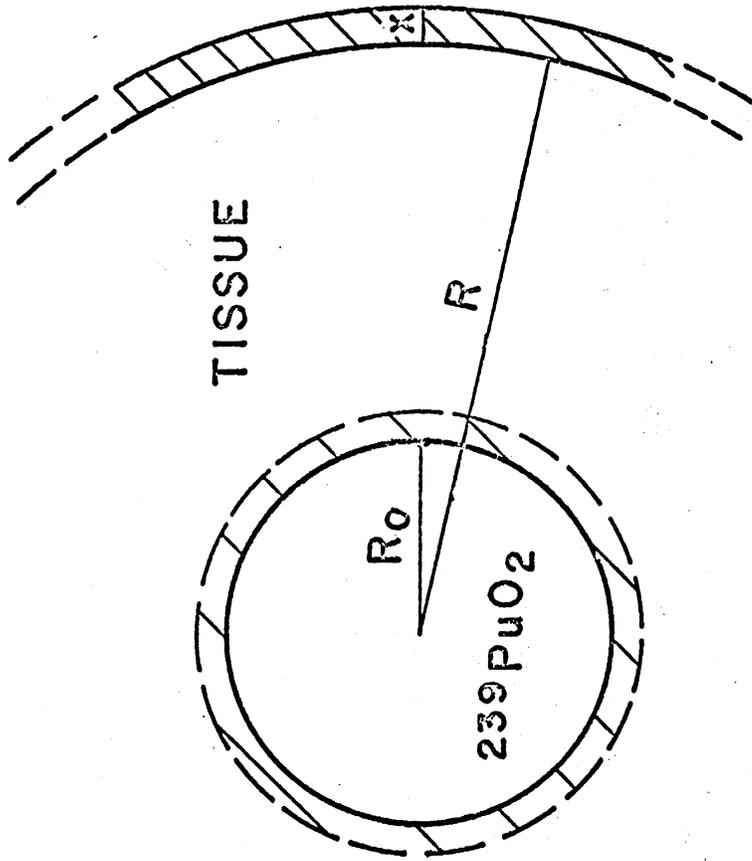




SPECTRUM DEGRADATION FOR Pu²³⁹O₂ PARTICLES



CALCULATION OF DOSE TO THIN TISSUE SHELL FROM
 SPHERICAL PARTICLE OF $^{239}\text{PuO}_2$

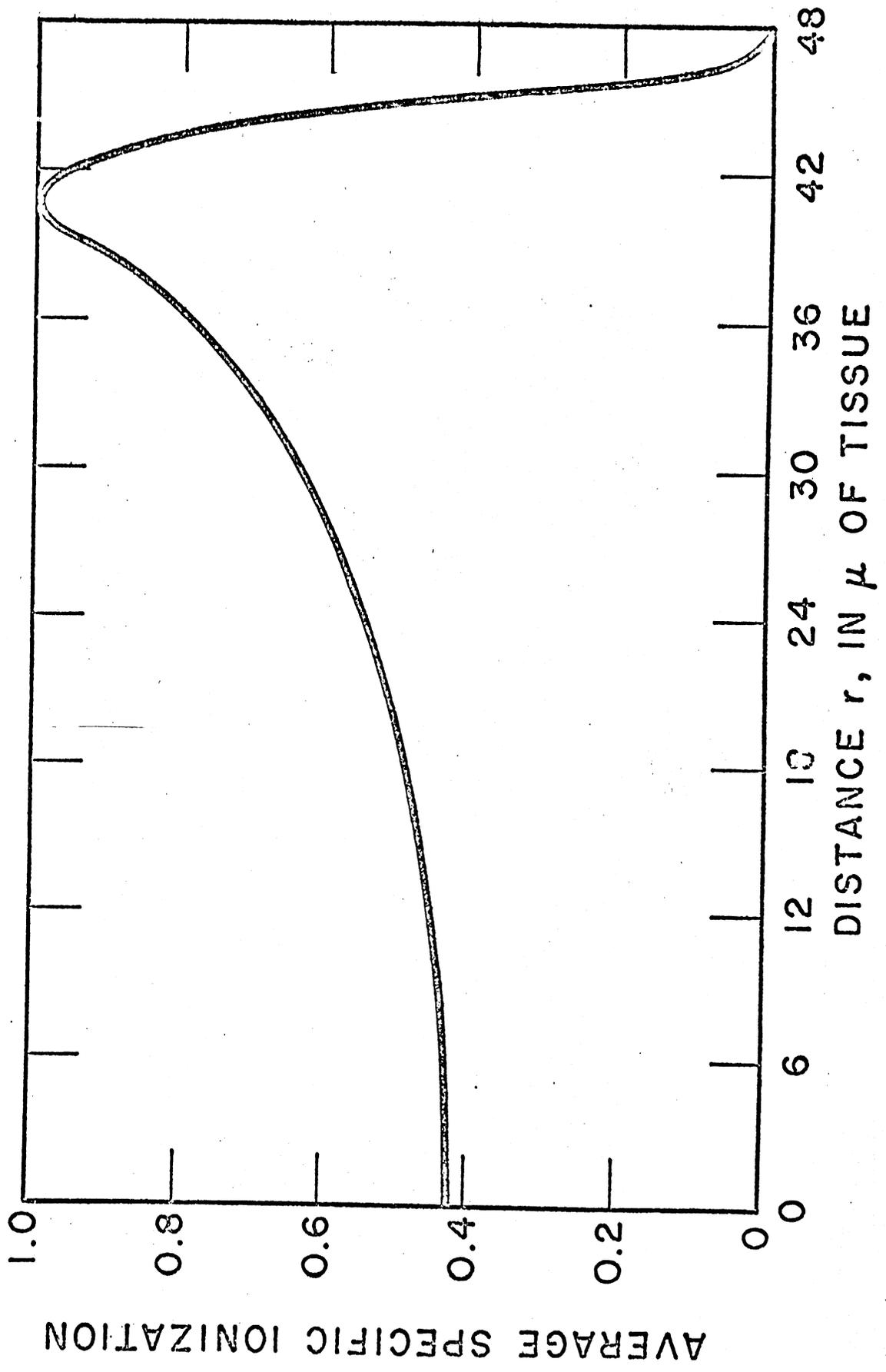


$$\text{DOSE RATE (DR)} = \frac{\text{ENERGY DEPOSITED IN TISSUE SHELL (ERGS/SEC)}}{\text{TISSUE SHELL MASS (GM) } 10^2} \frac{\text{ERGS}}{\text{GM-RAD}}$$

$$\text{ENERGY DEPOSITED / SECOND} = 4\pi R^2 \phi(a) \frac{dE}{dx} \times$$

$$\text{TISSUE SHELL MASS} = \frac{4}{3} \pi \rho_t [(R+x)^3 - R^3]$$

BRAGG CURVE FOR Pu^{239} ALPHAS IN TISSUE



DOSE RATE FROM $\text{Pu}^{239}\text{O}_2$ PARTICLES

