GAMMA SPECTRUM MEASUREMENTS AND THE INTERPRETATION OF ABSORBED DOSE DURING PLUTONIUM FUEL FABRICATION

Roger D. Caldwell

and

William C. Judd

Nuclear Materials and Equipment Corporation

Apollo, Pennsylvania

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Nuclear Materials and Equipment Corporation
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Introduction

Plutonium is one of the most toxic materials known to man. This is because of its high specific alpha activity and its tenacious deposition in the soft tissues of the bone. Consequently, the greatest hazard in handling plutonium is the potential for internal contamination. Most radiation protection measures are directed toward eliminating the alpha problem by exhausted containment.

However, reactor produced plutonium gives off significant neutron and photon radiation. Plutonium produced in a reactor is not pure ^{239}Pu. This is shown in Figure 1. Continuous neutron irradiation of ^{238}U produces higher numbered isotopes of plutonium. Most presently available plutonium has undergone exposure to about 1000 MWD/Ton of uranium. This relatively low burnup plutonium has a typical ^{240}Pu content of 8.5% and ^{241}Pu content of 1.27%. 1000 MWD/T PuO_2 has a surface gamma dose rate of 1.7 Rad/hour (13). Future plutonium fuel which has been recycled will have a much higher percentage of ^{240}Pu and ^{241}Pu.

The neutron emission from plutonium metal is largely dependent on the ^{240}Pu content. ^{240}Pu has a spontaneous fission neutron emission rate of 10^3 n/sec per gram of ^{240}Pu. The neutron emission from light element compounds of plutonium is increased because of alpha-neutron reactions with the light element nuclei. The neutron exposure problem in handling plutonium has been adequately treated elsewhere.(2,3,4)

The spectral response of photographic film is well known.[6] The film density per unit exposure is constant above photon energies of 200 KeV. The density per unit exposure begins to increase below 200 KeV and reaches a maximum at about 45 KeV. Below this peak in the response, the density falls off rapidly. The peak is caused by the increased photo-electric absorption effect of the silver in the photographic emulsion.

Experience at Hanford Plutonium Facilities[7] had indicated that electromagnetic radiations from plutonium could be considered in three effective energy groups. The first group consisted of X-rays of low penetrating ability with an effective energy of 17 KeV. The second group with an effective energy of 60 KeV consisted of a number of gamma rays from various plutonium isotopes and daughters. The third group occurred at an effective energy of about 400 KeV.

Consideration of the isotopic composition and the age of the plutonium since separation indicated that the 17 KeV and 60 KeV groups should dominate. However, uncertainties in the geometry, self absorption and incidental shielding material made it difficult to predict with assurance what the exposure spectrum might be. In addition, NUMEC subscribes to commercial film badge service and the company supplying service to us at the time did not furnish the density data for the filters in their badge. If the spectra were to vary from location to location, one would need access to the filter ratios, for proper interpretation of the film data.

Because of these difficulties we decided to carry out a study of the gamma energy spectrum to which plutonium glove box operations were exposed.

Gamma Exposure Spectra From Plutonium Fuel

Gamma spectrum measurements were made at several locations in the Plutonium Laboratory. The electronic equipment used was a Baird Atomic single channel

analyzer with 100 channel automatic step-scanning capibility. The detector was a two by two inch NaI (Tl) crystal. The equipment was placed on a cart for convenience. Since measurements were made in areas where dose rates were several mr/hr, a lead shield was constructed with a pinhole to prevent photomultiplier swamping.

The study revealed that the measured spectrum was not a strong function of the orientation of the pinhole. If the pinhole was not pointed at the source, very little radiation could be detected. This was clarified in areas where the dose rate was less than a mr/hr and the detector was used unshielded. Here the spectra were quite similar to those measured through the pinhole.

Film badges and R-meters were exposed at the same location so that their data could be tied in with the spectrum measurement.

Figure 3 shows spectra obtained from the chemical processing line and the ceramics production line. The 60 KeV peak from ^{241}Am dominates both spectra. A peak at 26 KeV is also due to ^{241}Am gamma peaks at 208, 267 and 333 KeV are from ^{237}U. Peaks at 340 and 380 KeV are emitted by ^{239}Pu. The strong 100 KeV neptunium K X-ray peak was noted by Heath[8] in his published ^{237}U spectrum. Published data[2,3,4] on plutonium gamma emission does not include this peak. It is surely a strong contributor to plutonium exposure.

A careful attempt was made to show the 17 KeV X-ray component of the spectra. The detector was in a 40 mil aluminum can. 40 mil of Al should attenuate 17 KeV X-rays only 25%.[9] However, a 17 KeV peak could not be produced and it was assumed that the glove box walls were effective in absorbing the low energy X-rays.

An interesting correlation can be seen by overlapping the spectra from the two separate locations. The relative heights of the ^{239}Pu and ^{237}U peaks are the same for both spectra. But, the ^{241}Am peaks are a factor of six higher for the

chemical processing line. The plutonium in the chemical processing box at the time of the measurement was from a long term research contract and had been in the box for at least 2 years. On the other hand, the ceramics plutonium had just been received and was less than a year old since separation. This graphically illustrates the importance of rapid processing of plutonium if one wishes to reduce dose rates to personnel.

Figure 4 shows the ceramic line spectrum compared to the spectrum measured outside of a Pu-U-Mo alloy vacuum melting glove box. The plutonium at both locations was received at approximately the same time from the same source (Hanford) and had the same plutonium isotopic composition. In this case, Americium had preferentially vaporized on the sides of the vacuum chamber. Scrapings from the inside of the chamber were chemically analyzed and found enriched in Americium.

The fifth figure gives the gamma spectrum from an Americium glove box. The 26 and 60 KeV peaks are well defined. The slight peak at 660 KeV aroused our suspicions and a search uncovered a small ^{137}Cs source which had been lost in another section of the laboratory.

Film Badge Response to Pu Gamma Radiation

Since the plutonium fuel spectra are typically dominated by the 60 KeV peak of the ^{241}Am daughter of ^{241}Pu, the film response characteristics should be similar to that of pure ^{241}Am. The sixth figure illustrates clearly the reason for the overestimation of personnel exposure during Pu fuel fabrication. The open window response of Dupont 555 film was measured by Phillips for ^{241}Am and ^{60}Co. If ^{60}Co is the only isotope which is used to calibrate film badges and if filter ratios are not individually interpreted, as is the case with most commercial film badge vendors, then the dose will be grossly over estimated for ^{241}Am.

For instance, if the Open Window density is 1.0, the dose for ^{60}Co is 800 mr and the dose for ^{241}Am is only 60 milliroentgens.

Once we had confirmed that the energy dependence of the film was responsible for the over estimate of Pu fuel exposures, it would appear to be a simple matter to establish a constant correction factor and go on from there. But unfortunately, it was not that simple. We had noted that our previous discrepancies between thin-walled ion chamber and film badge measurements did not result in a constant factor. It varied all the way from no difference up to a factor of 7 or more. It was possible that shielding material had hardened the spectrum, preferentially filtering out the low energy components and leaving an exposure spectrum quite different from ^{241}Am. In addition, some of our Pu workers also worked in our Hot Cell Facility and vice versa. Clearly without the density data under individual film badge data. Our vendor at that time did not find it possible to furnish us this basic data. Consequently, in July of 1965, we contracted with Eberline, Inc. for film badge service. They furnished us with the individual densities under their open window and plastic, aluminum and cadmium filters as well as their interpretation of the exposure.

Figure 7 gives filter ratios for Eberline's badges exposed during the last half of 1965 at NUMEC's Plutonium Laboratory and Hot Cell facility. Data is given for all exposures where the open window density is greater than one. The striking thing about this data is the nearly constant ratio of ow to cd densities for hot cell workers as compared to the large scattering of ratio's for plutonium workers. The average ow/cd ratio for Pu workers is nearly 3 times that for hot cell workers. The difference in percentage of standard deviations is even greater. Our interpretation of this data is:

1. Hot cell workers are exposed to ^{60}Co and fission product activity. The

The eighth figure shows the relative contribution by energy group to personnel exposure during typical plutonium fuel fabrication from 1000 MWD/T plutonium. Sixty-five percent of the dose is from the 60 KeV group. Less than 7% comes from the two highest groups.

The last figure shows the calculated depth dose in soft tissue from plutonium fuel. The gonad dose is reduced by 23% and the blood forming organ dose by 65%.

A subsidary study showed that the gonads were shielded by the steel floors of the glove box and received exposures less than half of that to the chest, where the film badge is normally worn. This, together with the recent elimination of the lens of the eye as a critical organ for gamma radiation, would permit an increase in the administrative dose to which plutonium glove box operators were exposed.

The importance of calculating depth doses using the true rad dose was recently pointed out by Jones[12]. Presently, most dosimeters are calibrated in terms of exposure rate in roentgen per unit time. Using the exposure dose and the known energy spectrum, the absorbed dose must be calculated before making depth dose corrections on the administrative dose. This partially offsets the advantages one gains by setting the administrative dose as that to the critical organs, because the absorbed dose per roentgen of free-air exposure is greater than one rad at low energies.

Summary

Plutonium fuel fabrication presents a hazard because of the attendant gamma and neutron radiation which is in addition to the well known alpha hazard. Our study has shown that the gamma exposure comes primarily from the ^{241}Am

energy response of the film badge is constant above 200 KeV. The dose can be taken directly from a ^{60}Co calibration curve of dose versus film density. Our R-meter measurements have agreed with Eberline reported film data within 10% for exposures from high energy gamma sources. If the ow/cd ratio is close to 1.0, we accept the reported data.

2. Plutonium workers are exposed to a range of gamma energies. The upper end of the plutonium spectrum will produce ow/cd ratios close to 1.0. The lower end will produce a ratio of 3.0 or greater. One Pu worker may be exposed to an averaged spectrum which is quite different than another worker. This could occur because of shielding, different working distance, etc. Presently, if the ow/cd ratio is greater than 2.0 we use a calibration curve we have run with a typical plutonium gamma spectrum. If the ratio is less than 2.0 we accept the reported data.
3. The open window to plastic shield ratio was within a 6% standard deviation of 1.0. This means simply that the large ow/cd ratio is not due to beta radiation.
4. The ow/Al ratio is what might be expected from an X-ray exposure. However, we have not yet learned how to interpret the reading under the aluminium filter.

Dose to Critical Organs from Pu Gamma Radiation

ICRP recommendations and Federal and State regulations on limits for penetrating radiation are made in terms of the dose to certain specified organs (11). With a knowledge of the exposure dose and the energy spectrum, it is possible to calculate the dose to the so called critical organs.

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11. ICRP Publication 6, Pergamon Press, Oxford (1964).

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daughter of ^{241}Pu and that is a function of plutonium isotopic composition and age since chemical separation. Problems in the interpretation of film badge data can be overcome if the densities under filters are known. The practice by commercial film vendors of furnishing this data to the health physicist should be encouraged.

FORMATION OF PLUTONIUM RADIONUCLIDES AND DAUGHTERS

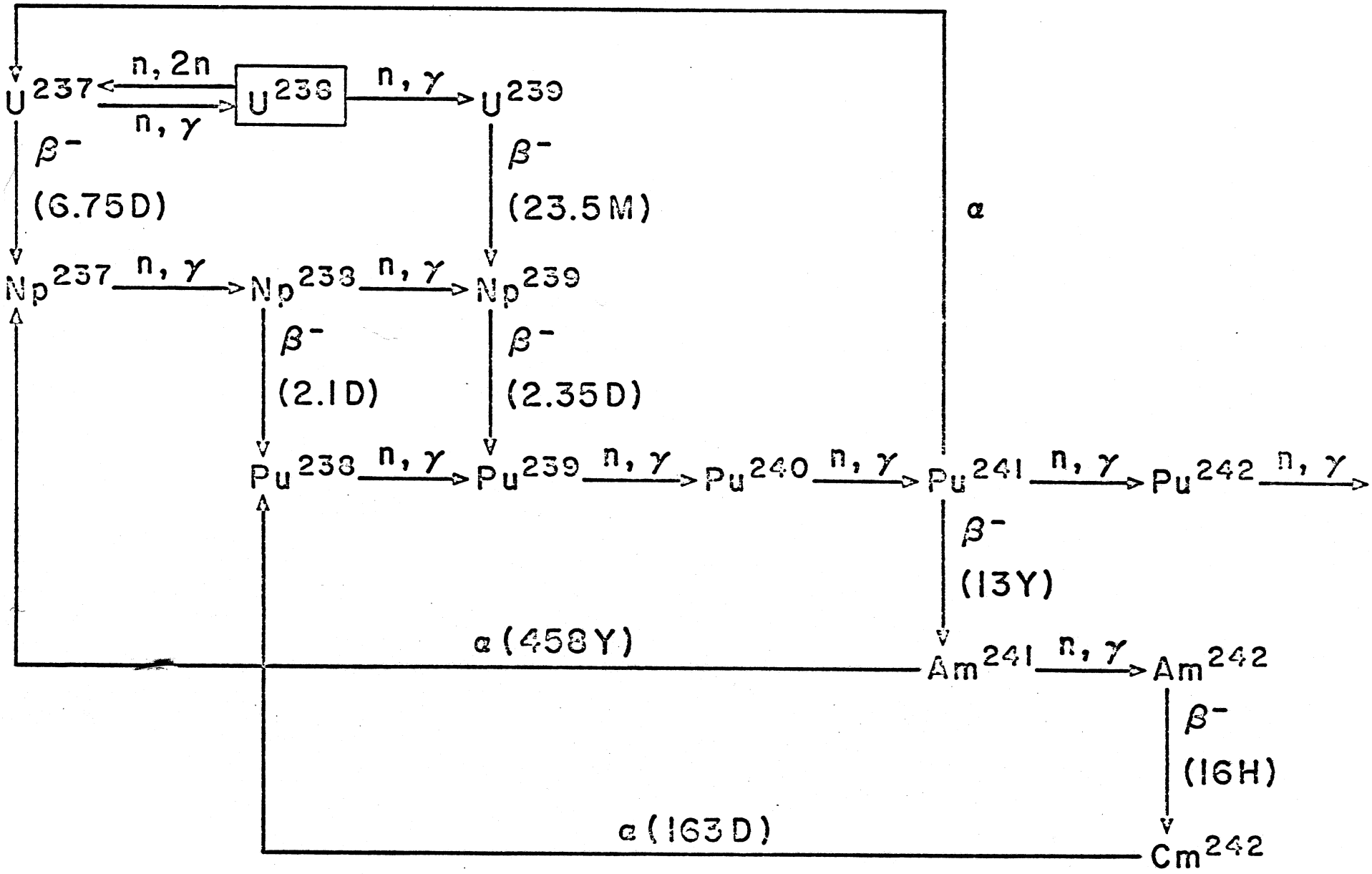


Figure 1

DOSE RATE CONTRIBUTIONS FROM Pu-241 DAUGHTERS

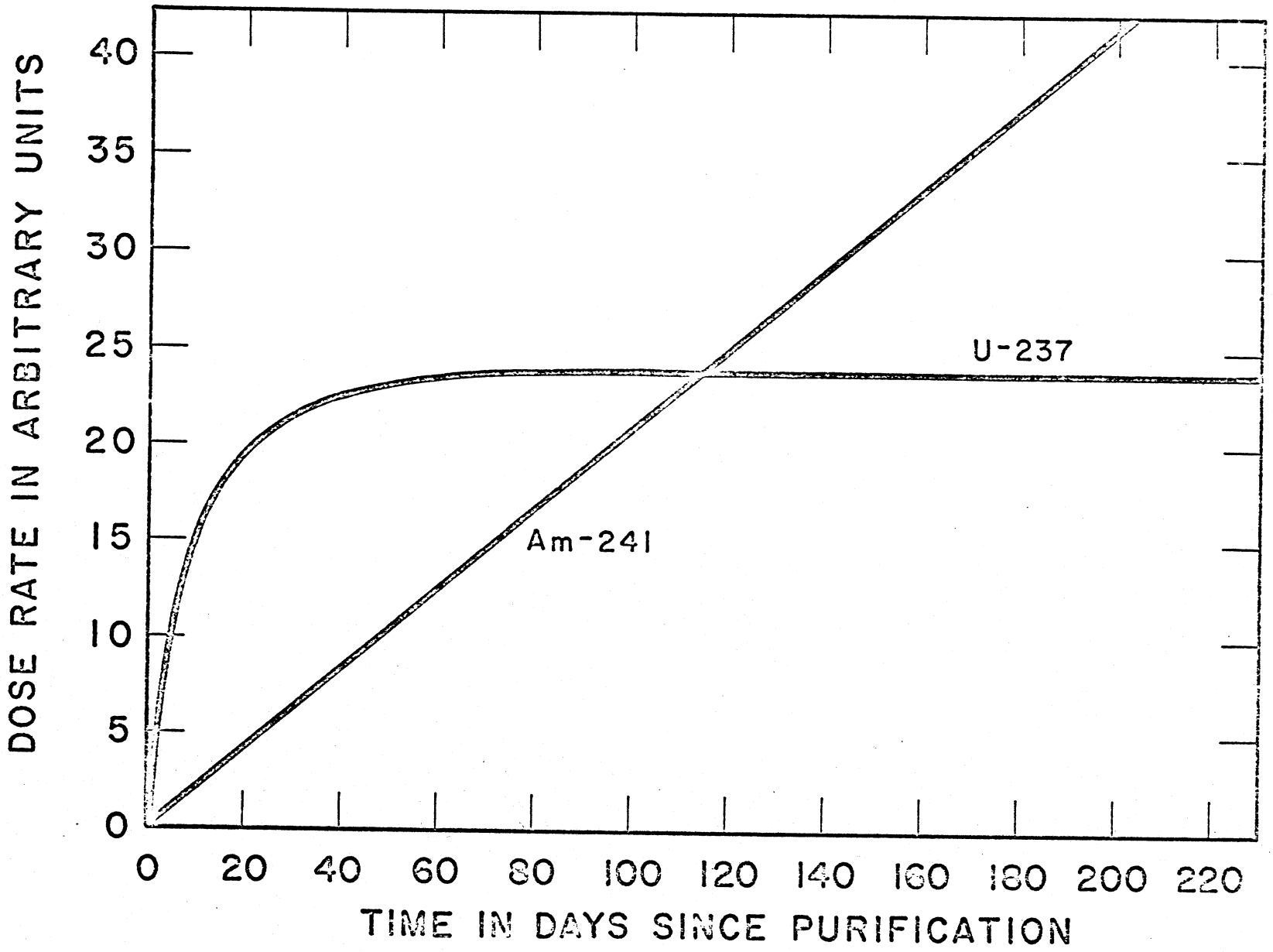


Figure 2

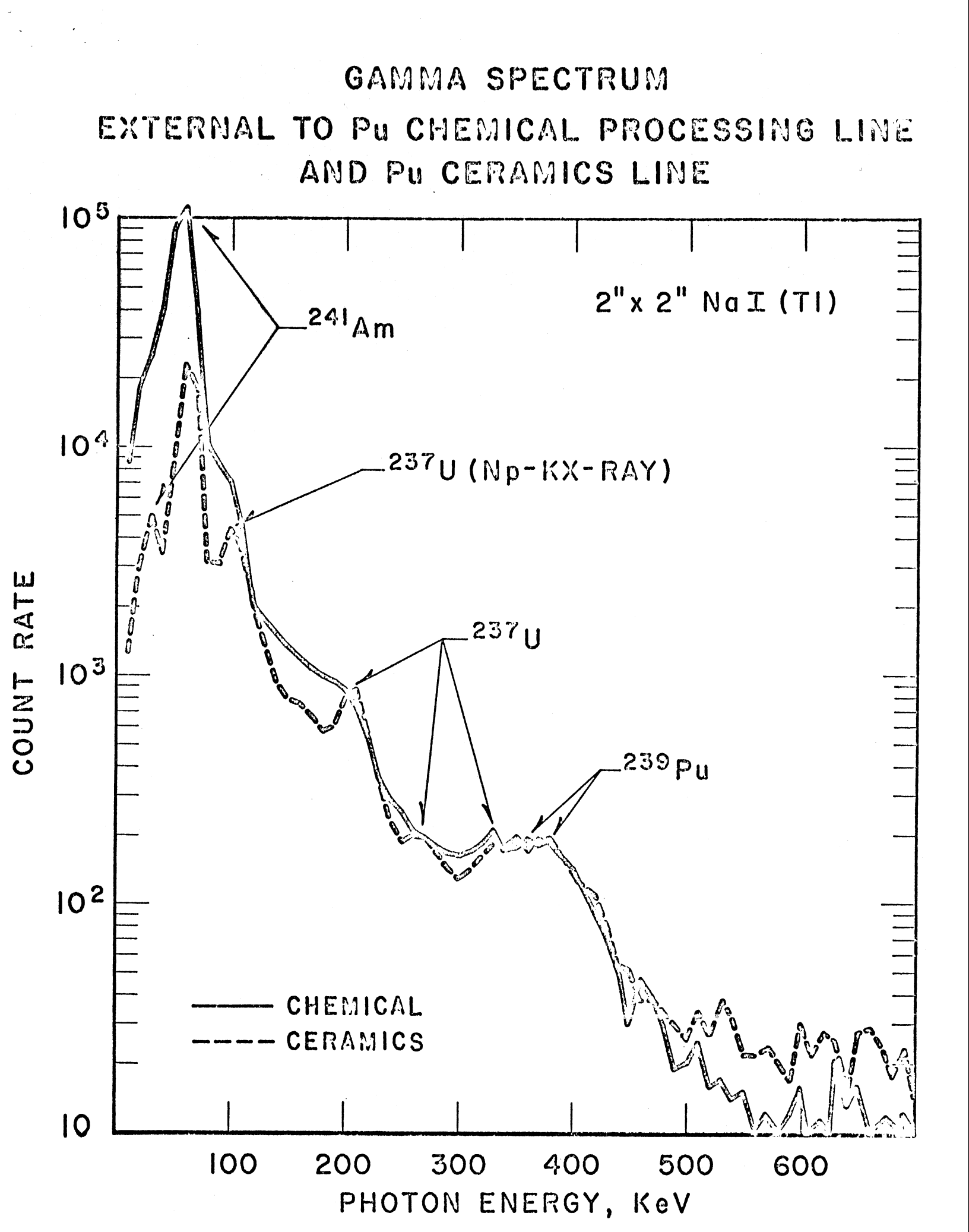

GAMMA SPECTRUM
EXTERNAL TO Pu CHEMICAL PROCESSING LINE
AND Pu CERAMICS LINE
2"x 2" NaI (Tl)
241Am
237U (Np-KX-RAY)
237U
239Pu
CHEMICAL
CERAMICS
COUNT RATE
PHOTON ENERGY, KeV
10
10²
10³
10⁴
10⁵
100
200
300
400
500
600

Figure 3

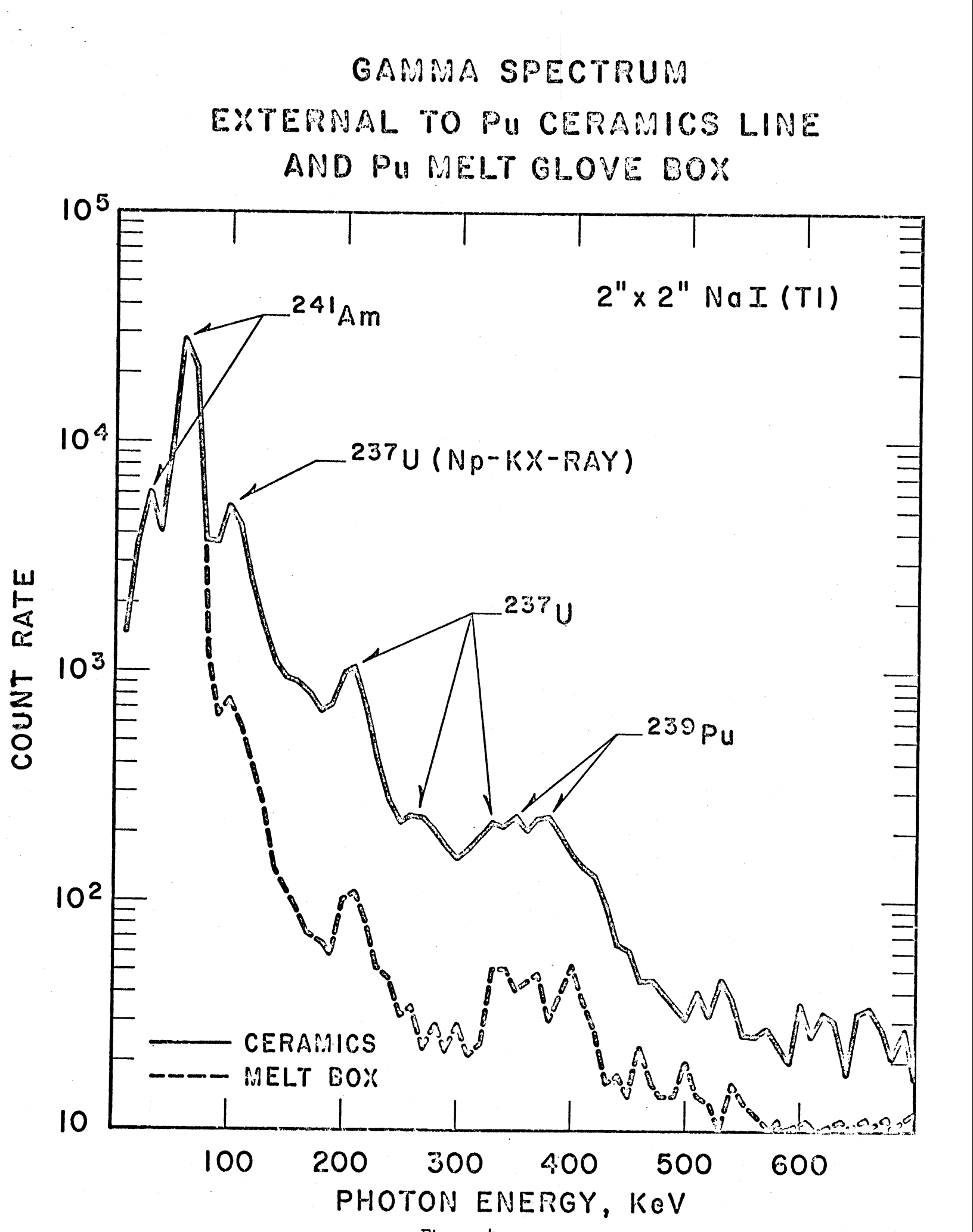

GAMMA SPECTRUM
EXTERNAL TO Pu CERAMICS LINE
AND Pu MELT GLOVE BOX
2" x 2" NaI (Tl)
241Am
237U (Np-KX-RAY)
237U
239Pu
CERAMICS
MELT BOX
COUNT RATE
10^5
10^4
10^3
10^2
10
100
200
300
400
500
600
PHOTON ENERGY, KeV

Figure 4

RESPONSE OF DuPONT TYPE 555 FILM
BNL FILM BADGE

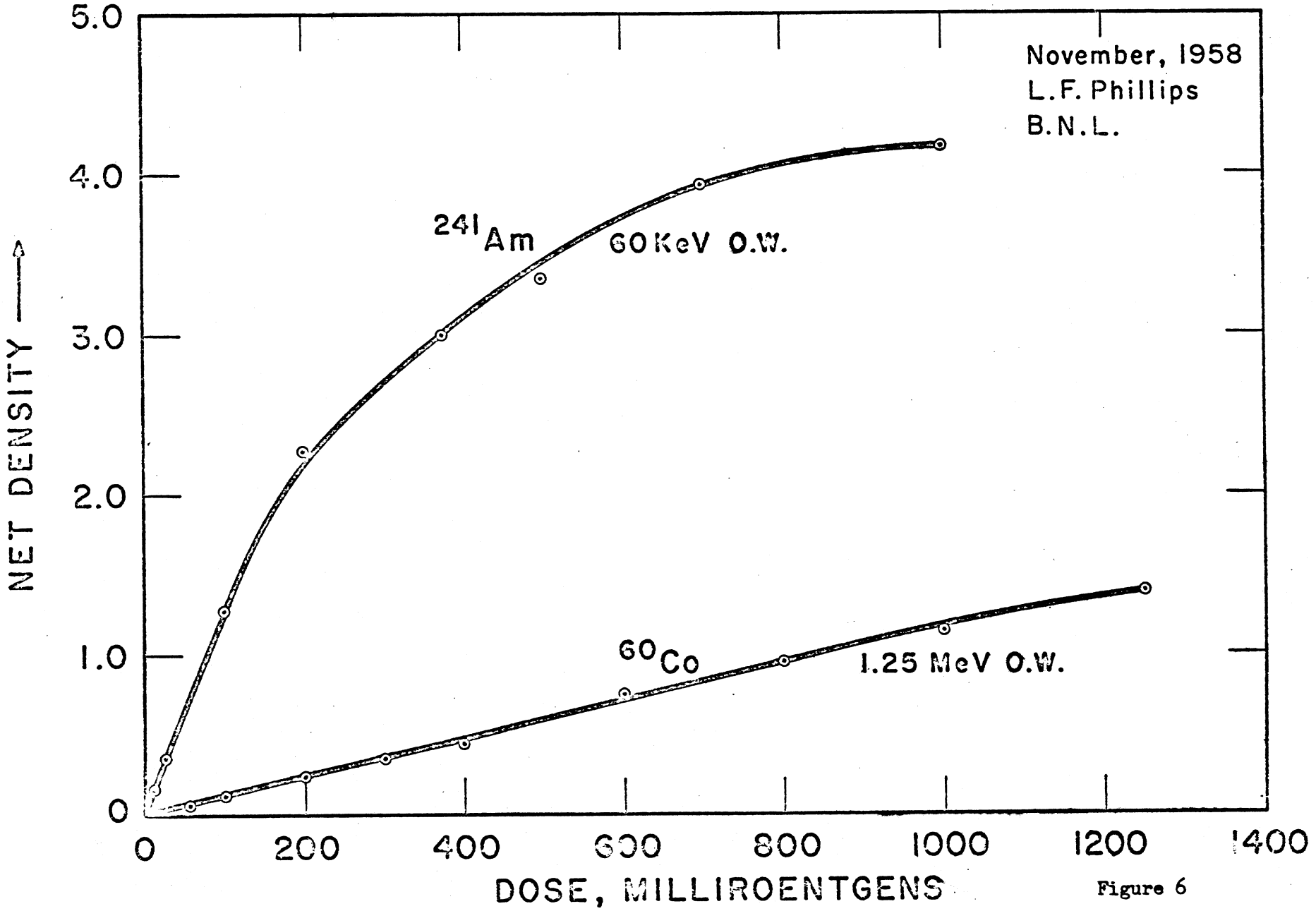


Figure 6

GAMMA SPECTRUM
OUTSIDE AMERICIUM BOX

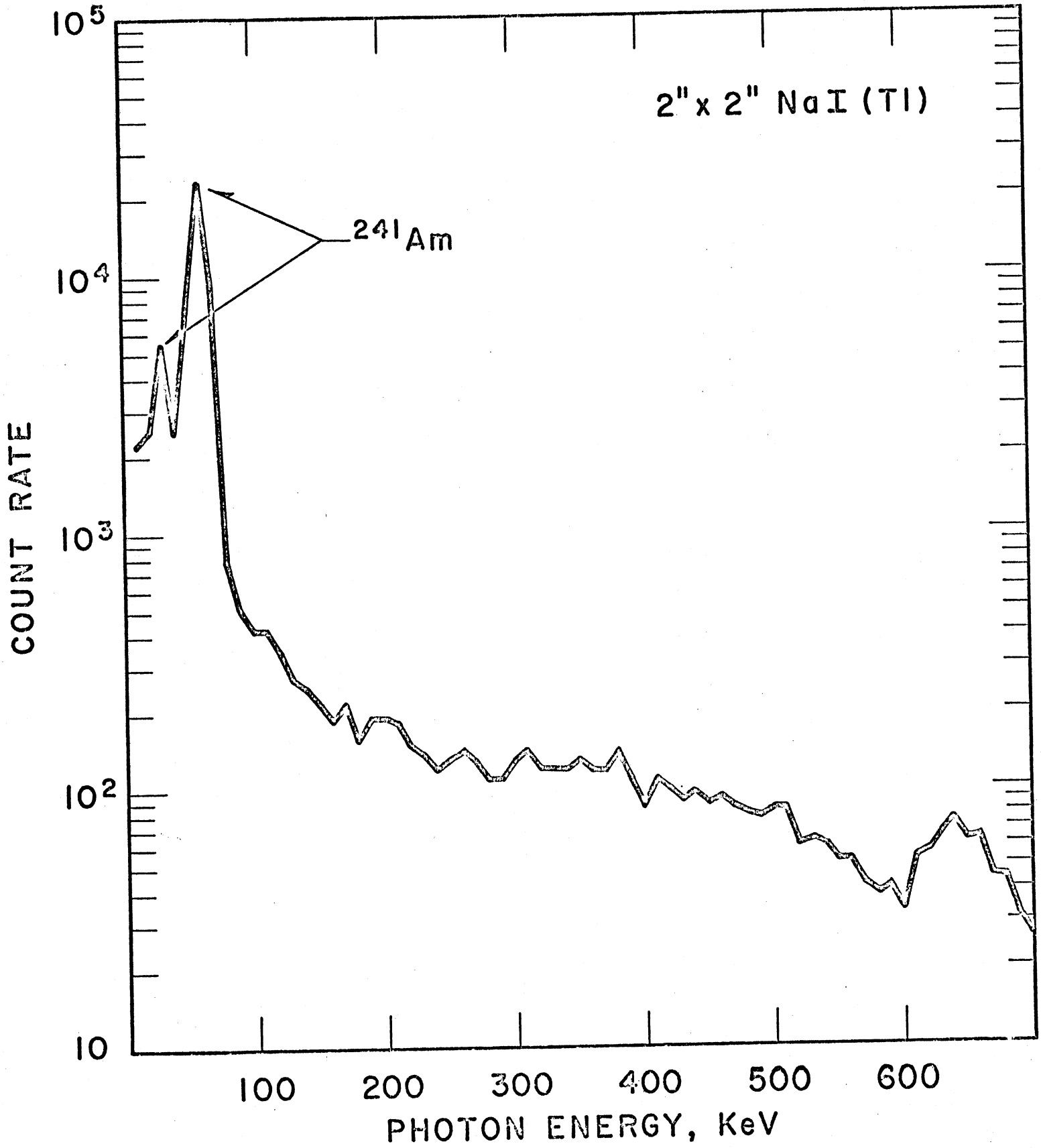
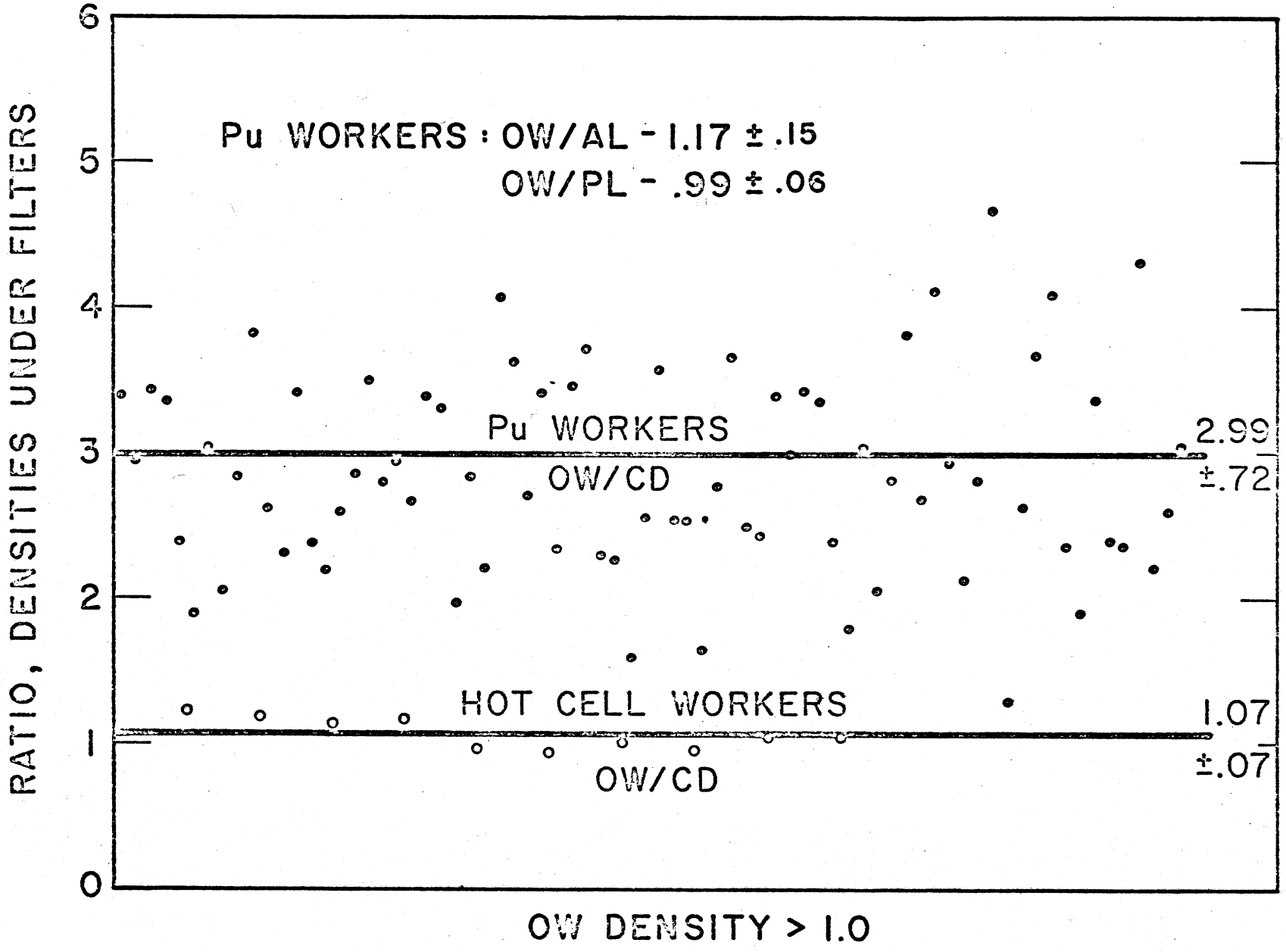


Figure 5

FILTER RATIOS FOR EBERLINE FILM BADGES



INDIVIDUAL DATA OBTAINED DURING 1965

Figure 7

RELATIVE CONTRIBUTION BY ENERGY GROUP TO PERSONNEL EXPOSURE DURING Pu FUEL FABRICATION FROM 1000 MWD/T PLUTONIUM

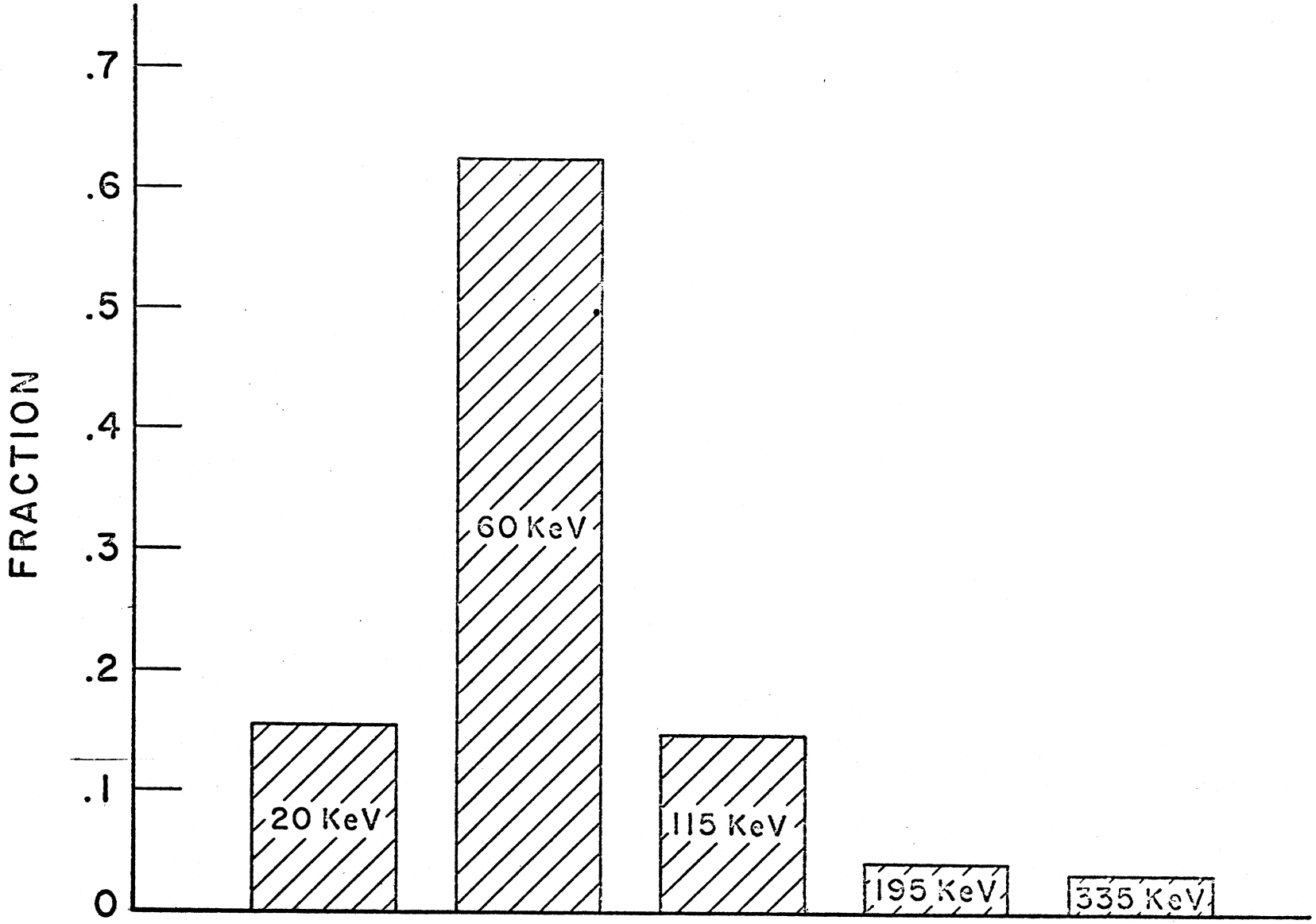


Figure 8

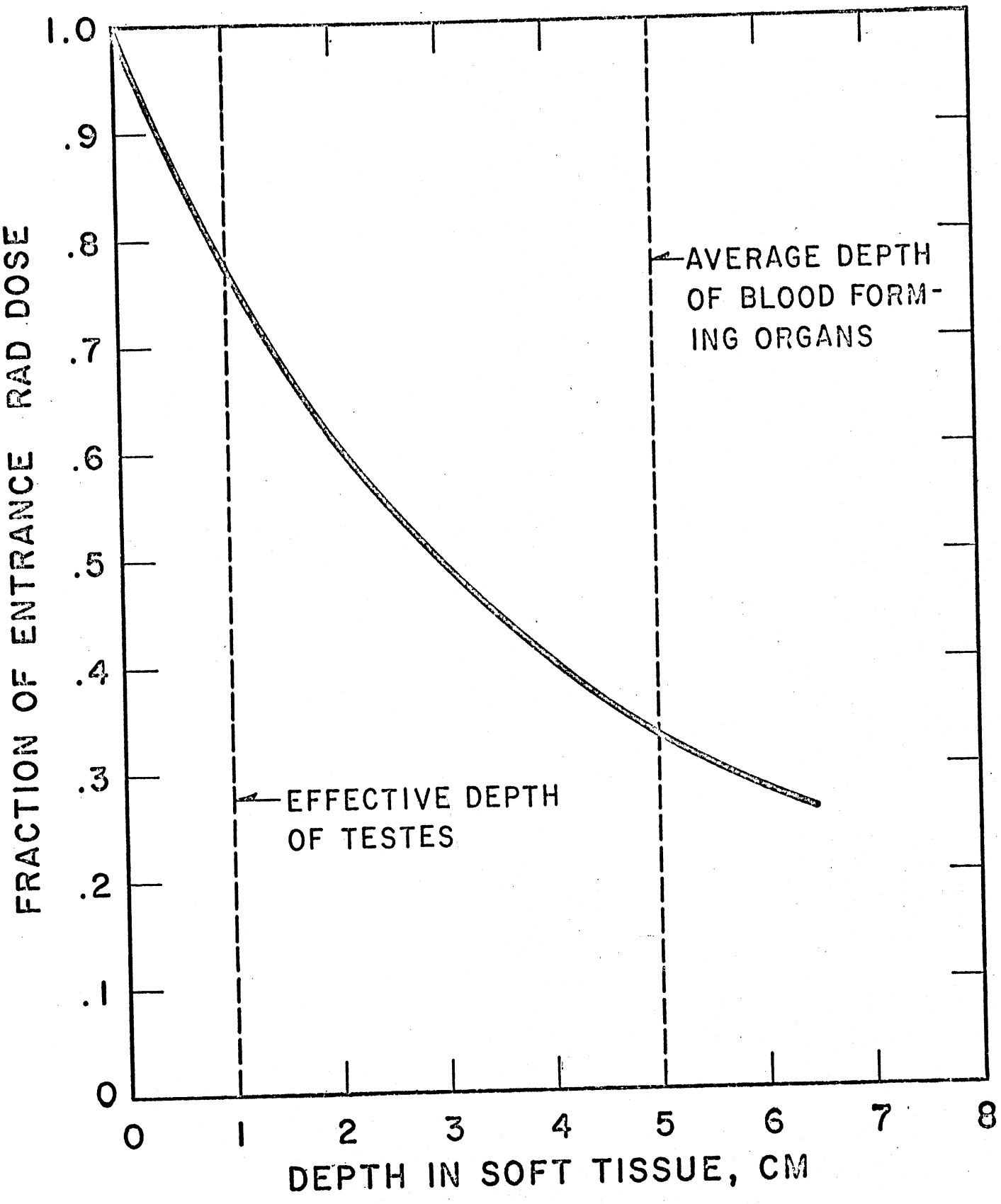

GAMMA DEPTH DOSE FROM Pu FUEL
FRACTION OF ENTRANCE RAD DOSE
1.0
.9
.8
.7
.6
.5
.4
.3
.2
.1
0
0
1
2
3
4
5
6
7
8
DEPTH IN SOFT TISSUE, CM
AVERAGE DEPTH OF BLOOD FORMING ORGANS
EFFECTIVE DEPTH OF TESTES

Figure 9