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TITLE: APPLICATION OF LINEAR ACCELERATOR TECHNOLOGY TO THE DETECTION OF TRACE AMOUNTS OF TRANSURANICS IN WASTE BARRELS

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APPLICATION OF LINEAR ACCELERATOR TECHNOLOGY TO THE DETECTION OF TRACE AMOUNTS OF TRANSURANICS IN WASTE BARRELS

ABSTRACT

Electron linear accelerators (linacs), as sources of photons and neutrons, can produce a significant number of fissions in transuranic isotopes contained in large barrels of waste material. Both photons and thermal neutrons have been used to detect about 1 mg of plutonium in 105-kg matrices. A sequential interrogation with neutrons and photons, easily possible with linacs, can show both fertile and fissile constituents among the heavy-mass isotopes. The advantages of linacs in solving existing assay problems include: (1) high available beam current; (2) variable beam current, beam energy, pulse width, and pulse repetition frequency; and (3) beam-scanning ability. They also are compatible with passive assay instruments. Their versatility makes it likely that they will remain useful as assay technology advances.

1. Introduction

The upper limit of specific activity of transuranic isotopes (TRU) allowed in solid wastes that are put into non-retrievable storage in Department of Energy (DOE) facilities is 10 nCi/g.[1] This amount, for example, corresponds to 0.16 µg of ^{239}Pu, 1.1 µg of ^{233}U, or 0.003 µg of ^{241}Am per gram of matrix. In high-density matrices, those about 0.3 g/cm^3 or greater, the natural photon emission from TRU is severely attenuated and natural neutron emission, such as that from spontaneous fission of ^{240}Pu found with the ^{239}Pu, is produced at low rates that can be dominated by natural background counts.[2,3] Passive assay systems that measure spontaneous emissions not only require a reasonable count rate, but are limited to those isotopes that have natural neutron emissions, unless the package sizes and densities are small.

Most of the problems associated with passive assay are eliminated when a source of nuclear particles is introduced into the waste matrix at sufficiently high energies to produce fissions in the TRU that act as "signatures" of their presence. Such techniques, called active assay, can use natural sources, nuclear reactors, or particle beams from accelerators as interrogation mechanisms.

Accelerators offer by far the greatest versatility among choices of interrogation methods because they produce the largest available flux densities of fast neutrons and photons and have appreciable adjustability in beam current, energy, and duty cycle. The major disadvantages of accelerators are their

size and cost, although both of these have been reduced in recent years, particularly because of the development of special-purpose machines for medicine or materials testing. Our research group, made up of scientists and engineers from the Los Alamos Scientific Laboratory (LASL) and EG&G Incorporated, decided to pursue the use of electron linear accelerators (linacs) as waste assay tools. In this presentation we will show data and provide further details to demonstrate the viability and usefulness of linac technology in TRU waste assay.

2. Basic Parameters of Linacs in Waste-Assay Applications

Electron linacs can produce hundreds of milliamperes of peak electron current at energies from a few MeV to hundreds of MeV. Energies from about 8 to 20 MeV lie in the most practical range for waste assay purposes. The electrons are used to produce bremsstrahlung photon radiation by impinging them on targets of high-Z material. Neutrons can also be made by allowing the bremsstrahlung photons to generate (γ,n) and (γ,f) reactions in materials such as beryllium or uranium. Figure 1 shows bremsstrahlung spectra produced as a function of electron energy for a water-cooled tungsten target.[4] The data are for the DOE/EG&G linac in Santa Barbara, California, shown in Fig. 2. It was this machine that we have used for all the experimental work we shall describe. Figure 3 shows neutron spectra produced by this linac using a beryllium target. The spectra shown are calculations normalized to data obtained at a machine energy of 27 MeV.

Another important feature of linacs is their pulse structure. In the Santa Barbara linac, the pulse width can be varied from about 50 ps to about 4 μs, and the pulse repetition rate can go from 300 Hz to 1 Hz or less. Special-purpose linacs, built for waste assay, can be designed for the optimum pulse structure needed in their application.

3. Dynamics of Photofission and Delayed Neutron Production

The basic approach for this assay technique is to bombard the waste barrel with either photons or neutrons produced by the linac. Let us first deal with the photofission approach. Because of the high photon flux, linacs can induce a large number of fissions among the TRU in the matrix. Figure 4 shows photofission cross sections versus photon energy for ^{239}Pu, ^{233}U, ^{237}Np, ^{241}Am, ^{238}U, and ^{232}Th.[5] In each case the curve reaches a peak around 14 MeV. Figure 5 shows the product of the bremsstrahlung spectra from the linac with the cross section for ^{239}Pu as a function of photon energy for different electron energies. Here we can note that the fission yield continues to go up significantly as the electron energy is increased, at least to about 18 MeV. In fact, this increase continues more or less indefinitely, but it is accompanied by a

corresponding increase in photoneutron production from materials in the matrix itself. Figure 6 shows this effect on ^{208}Pb and $^{27}A\ell$. Other common materials, like deuterium in hydrogenous matrices and ^{13}C in carbonaceous matrices, have yields similar to those for lead. Even the most neutron-stable materials, like iron, silicon, etc. will, like aluminum, produce a large number of photoneutrons when the beam exceeds about 18 MeV.

The effect of these photoneutrons is to generate a large prompt neutron flux that cannot be separated practically from prompt fission neutrons. Fortunately, however, TRU fissions produce a significant number of delayed neutrons from particle decays of some of the fission products. There are six prominent delayed groups with emission half lives varying from about 0.2 to 55 seconds.[6] These groups, their decay constants (β) and yields from fission of ^{239}Pu, are shown in Table I. The emission rate of delayed neutrons from photofission is shown in Fig. 7. Two other expressions are also shown; these are the important ones for our consideration. The second equation shown is the delayed neutron yield counted during and long after the irradiation. In other words, this expression represents the maximum delayed neutron yield and is a function of the total irradiation time. The third expression gives the delayed neutron yield when the counting occurs after the radiation has ceased. This value is not a function of the irradiation time after about 200 seconds of irradiation because the production-decay rate reaches a steady state value. The importance to waste assay is that if a reasonable fraction of the delayed neutrons can be counted between beam pulses without interference from the prompt events, an assay can be continued for as long as is necessary to count a statistically sufficient number of neutrons.

Another approach that could be used with photofission assay is to lower the electron energy such that few photoneutrons are produced in the matrix, and attempt to count the prompt fission events in coincidence. This method, while possible, is less practical if sufficient beam energy is available, because the delayed neutron yield per beam electron at high energies, such as 12 or 14 MeV, is comparable with the prompt fission yield at low energies, such as 7 or 8 MeV.

4. Photofission Experimental Results

Figure 8 is a photograph of the experimental arrangement we used for delayed neutron counting after photofission. A polyethylene chamber for neutron moderation contained a 55-gallon (208 liter) barrel that is a standard waste container throughout the DOE and American industrial complexes. Pressurized ^{3}He gas proportional counters were placed in the moderator to count the thermalized neutrons present at any time. A typical detector is shown in Fig. 9.

In our first complete photofission assay study, we took advantage of existing electronics for amplification and processing of the detector signals, and we were constrained to locate the detection geometry in the high room background conditions of the linac accelerator chamber itself. Because of these limitations we chose to count neutrons emitted and thermalized after the radiation ended. A typical detector signal is shown in Fig. 10 for a 1-g plutonium sample at 14 MeV electron energy. The total counts are the sum under this curve. Figure 11 shows a set of 18-MeV data for a 105-kg matrix of aluminum scrap containing various masses of plutonium. Here we can see the detection limit, where the signal to background becomes difficult to discern, is about 1 mg of plutonium, or less than 1 nCi/g of matrix.

We have begun a similar study with better electronics in which we will count delayed neutrons between linac pulses. We expect improvements in total counts of about a factor of 10 for 5-min irradiations. Some preliminary data indicate that the prompt events require about 1 ms after each pulse to die away to background levels. If we assume, then, a steady state production of delayed neutrons and a 300-Hz pulsing rate, we can expect to count about 70 percent of the available neutrons (weighted by the detection efficiency, which is typically about 0.2).

5. Thermal Neutron Fission Experimental Results

Figure 12 is a photograph of the detection geometry used for thermal-neutron activation of the TRU in the waste barrel. The detection method is the "differential die away" technique[7] discussed in the previous paper.[8] A set of data, similar to those from the photofission study, are shown in Fig. 13. The electron beam energy was 8 MeV. Here again, the detection limit is well below 10 nCi/g; in fact, it is less than 1 nCi/g.

6. Matrix Effects for Photon and Neutron Interrogation

Inducing fissions by either photons or neutrons is complicated somewhat by the effect of the matrix which contains the TRU. The matrix effect, fortunately, is not too severe for typical materials for either type of interrogation, and is different for photons than for neutrons. For neutrons the two most serious problems are the presence in the matrix of good absorbing materials, such as boron and cadmium, and the masking effect of TRU masses that absorb neutrons in their outer surfaces, thereby preventing access to their interiors. Only in the case where a large amount of cadmium and boron was added to the matrix did the results vary much. From the shape of the plutonium oxide in our sample capsules, we estimated the effect of surface absorption of neutrons by the plutonium and calculated effective masses of plutonium for the various irradiations. Not only did the curve of counts versus

plutonium mass become a linear function, showing the basic validity of the correction, but the necessary corrections are small for amounts corresponding to about 10 nCi/g or less.

Figure 14 shows results from a matrix study we did for photofission. The chamber used was smaller than the large barrel counter, but the results can be scaled with reasonable certainty. The data indicate that none of the matrix effects are particularly large for common materials. Some effect of neutron absorption can be surmised from the pyrex glass (containing boron) data, since it gives a somewhat lower yield than expected. In a converse manner, the polyethylene matrix probably gives a higher yield because it increases the amount of moderating material in the whole assembly.

When both photon and thermal neutron interrogations are used for the same matrix, we can expect that the photons will penetrate to any TRU that is masked to neutrons by itself or some other material. The neutrons, conversely, will help compensate for any photon losses due to matrix variations by their lack of strong dependence on the electron density of the matrix atoms.

7. Photon and Neutron Interrogation in the Same Assay Geometry

One of the most powerful ways in which a linac can be used for waste assay is in a mode where thermal neutron and photon irradiation of the matrix can be performed in sequence with little or no change in the detecting geometry. The main reasons for this method are that thermal neutron interrogation can clearly identify the presence of fissile materials and photofission can identify any heavy mass fertile constitutents. In combination the fraction fissile and the fraction fertile can be determined. This type of assay is very important for certain classes of waste, such a soil samples which contain large amounts of natural uranium and thorium, waste containing large amounts of americium and neptunium, and waste with large uncertainties on the TRU species in it. In this last case it may also be necessary to use additional assay techniques, such as an analysis of the fission product gases, [9] to gather further information on the species constituents.

We have begun a study of the sequential interrogation method (SIM) using a beryllium target that can be moved into or out of the beam of photons from the linac bremsstrahlung target. In some of the work we have counted delayed neutrons from both types of fission; in other cases we have counted neutrons in the differential dieaway method and delayed neutrons from photofission. Although the study is not complete, we have seen large increases in the photofission signal with the presence of ^{238}U, as an example, in the same matrix with plutonium, while the neutron-induced signal was virtually unchanged. Figure 15 shows a schematic layout of a possible detector geometry for SIM assay. The features of this geometry are these: first,

sufficient moderator and reflecting material to give the thermal neutrons a long persistence (around 0.5 ms or longer half life) in the chamber; second, a reasonably large counting efficiency for neutrons from the TRU fissions, and third, reliable electronics that can recover in a few tens of microseconds after each linac burst. Notice that the detectors are shielded from thermalized room neutrons and are shielded from thermalized interrogation neutrons. The outer shielding is to lower the background in the photofission mode; the inner shielding is to allow the differential dieaway technique to function. Since both thermal neutrons and photons from linacs independently can assay TRU at well below 10 nCi/g, we expect a SIM approach to have at least 10 nCi/g sensitivity as well.

8. Other Advantages of Linac Interrogation

Besides the demonstrated sensitivity and the potential for SIM assay in linac methods, these sytems offer other advantages in waste management applications. First of all, they are compatible with passive counting systems. They can use the same or similar detection systems and electronic processing, since most passive systems assaying large containers will also utilize neutron detection. Second, they have energy variability and control of beam current. Both can be important in shortening assay time. Energy variability could be useful in species identification because of variations in fission thresholds and cross sections among TRU species. Third, they are capable of assaying large packages, such as "six-packs" of waste barrels or large crates. This is both because of the high intensities available from the interrogating beams and because of the potential for sweeping the beam across the package. Fourth, the beam sweeping can be used to scan containers to locate contaminated portions of the volume that can sometimes be removed for recycling or to reduce the overall volume to below 10 nCi/g. Fifth, SIM assay, which is practical only with electron accelerators, will minimize the effects of the waste matrix on the assay. Sixth, matrices with high β-γ backgrounds can be assayed with no changes in geometry other than additional lead shielding. Increased cosmic-ray backgrounds, significant for passive systems or those with marginal count rates, would have little effect on the assay. Seventh, linacs can be used in counting and coincidence assay techniques that take advantage of their pulse structure, which can be varied in frequency and pulse width. The linac, then has versatility that can be used effectively when future assay problems begin to be identified. It is not likely to become obsolete as an assay instrument.

The continuing LASL program is undertaking applications research in these seven areas as well as in advanced detection systems that can exploit the linac active interrogation approach to waste assay.

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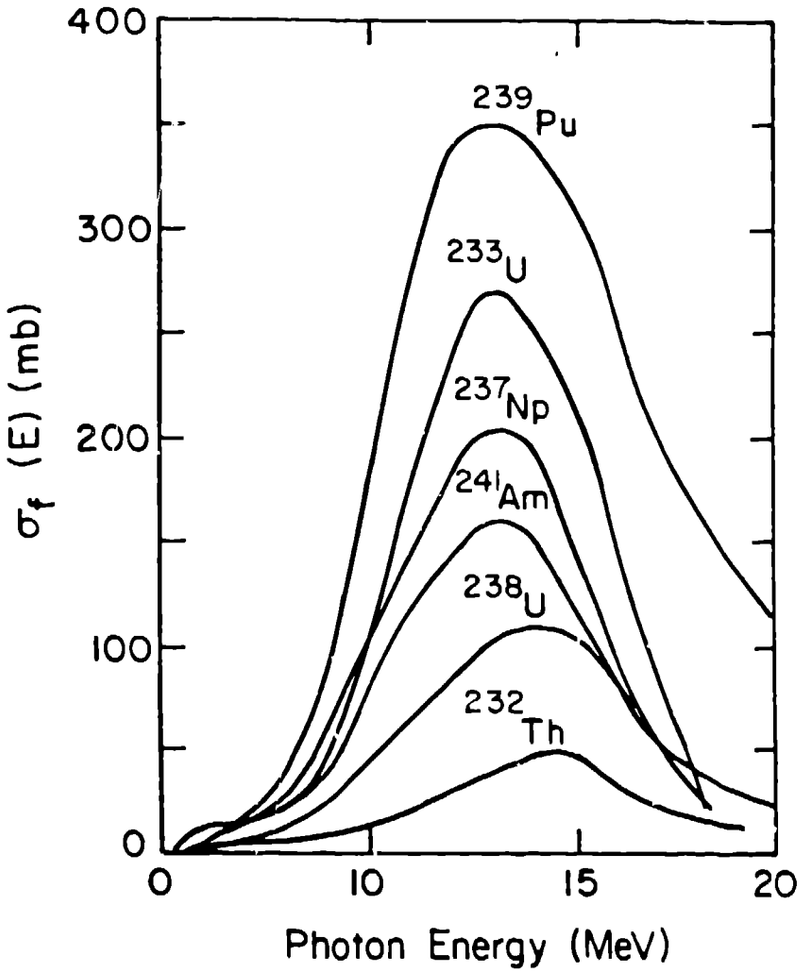

Fig. 4. Photofission cross sections versus energy for various TRU species.

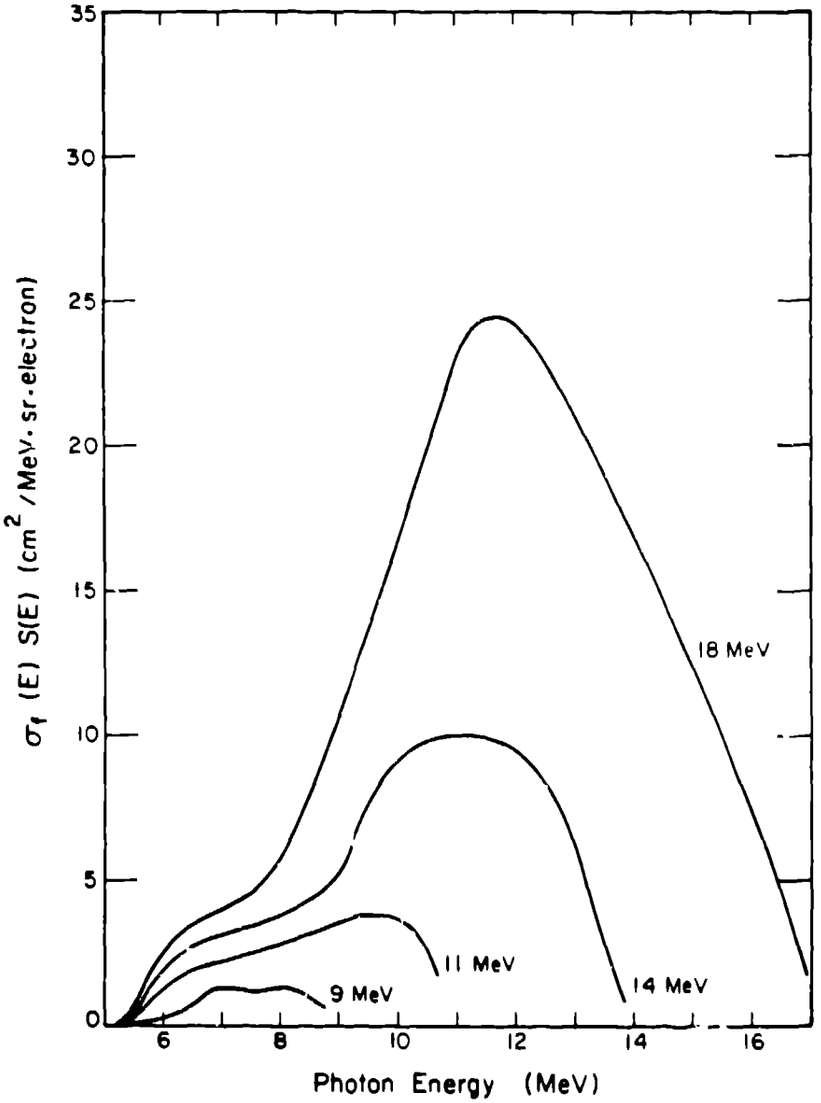

Fig. 5. Product of the bremsstrahlung photon spectra and photofission cross section for ^{239}Pu at various electron energies.

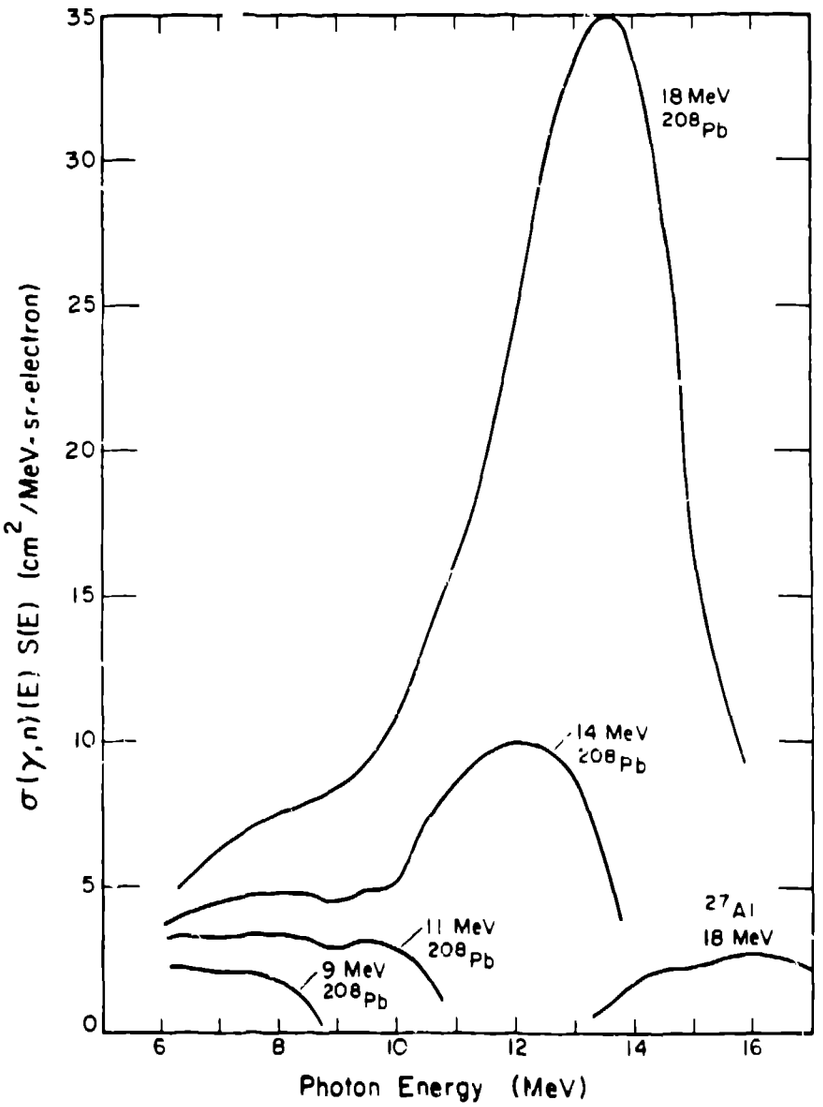

Fig. 6. Product of the bremsstrahlung photon spectra and photoneutron cross section for ^{208}Pb and ^{27}Al at various electron energies.

TABLE I

DELAYED NEUTRON DECAY CONSTANTS AND YIELDS FROM FAST (FISSION SPECTRUM) NEUTRON-INDUCED FISSION OF ^{239}Pu

Delayed Neutron Group	β_i (1/s)	γ_i (neutron/fission)
1	0.0129	0.00024
2	0.0311	0.00176
3	0.134	0.00136
4	0.331	0.00207
5	1.26	0.00065
6	3.21	0.00022

$$R_d = R_P \sum_{i=1}^{6} \beta_i (1 - e^{-\lambda_i t_r}) e^{-\lambda_i t_d}$$

R_d = The emission rate of delayed neutrons (n/s)

R_P = Fission rate (fissions/s)

where

β_i = absolute yield of the ith delayed neutron group (n/fission)

λ_i = decay constant of the ith group (1/s)

t_r = irradiation time (s), and

t_d = decay time (s)

$$N_T = \sum_{i=1}^{6} \beta_i R_P t_r = 0.00631 R_P t_r$$

N_T = The total yield of delayed neutrons

$$N_A = R_P \sum_{i=1}^{6} \beta_i / \lambda_i = 0.0922 R_P$$

N_A = Delayed neutrons after irradiation

Fig. 7. Emission rate of delayed neutrons from photofission.

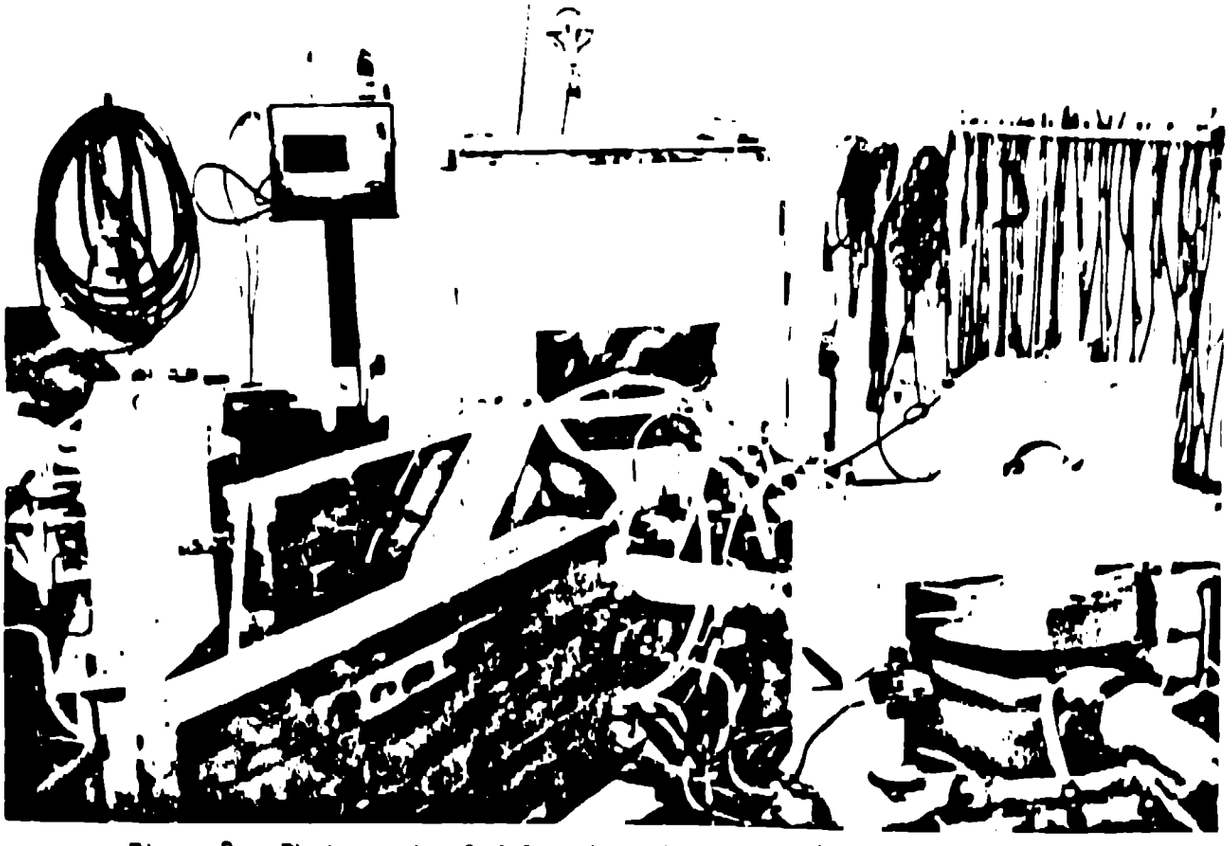

Fig. 8. Photograph of delayed neutron counting geometry.

Fig. 9. A ^{3}He gas proportional counter.

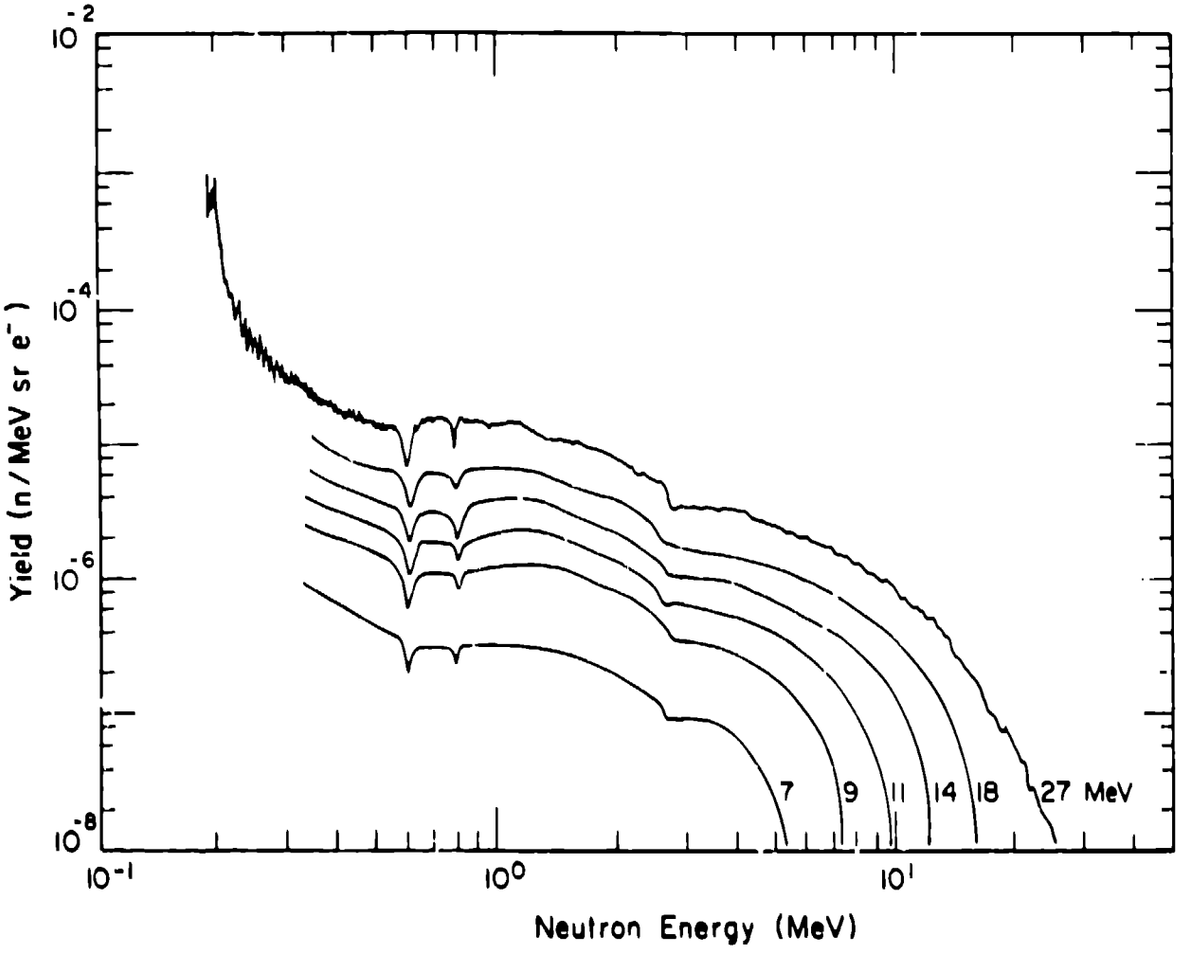

Fig. 3. Neutron spectra inferred from the 27-MeV spectrum of the DOE/EG&G linac.

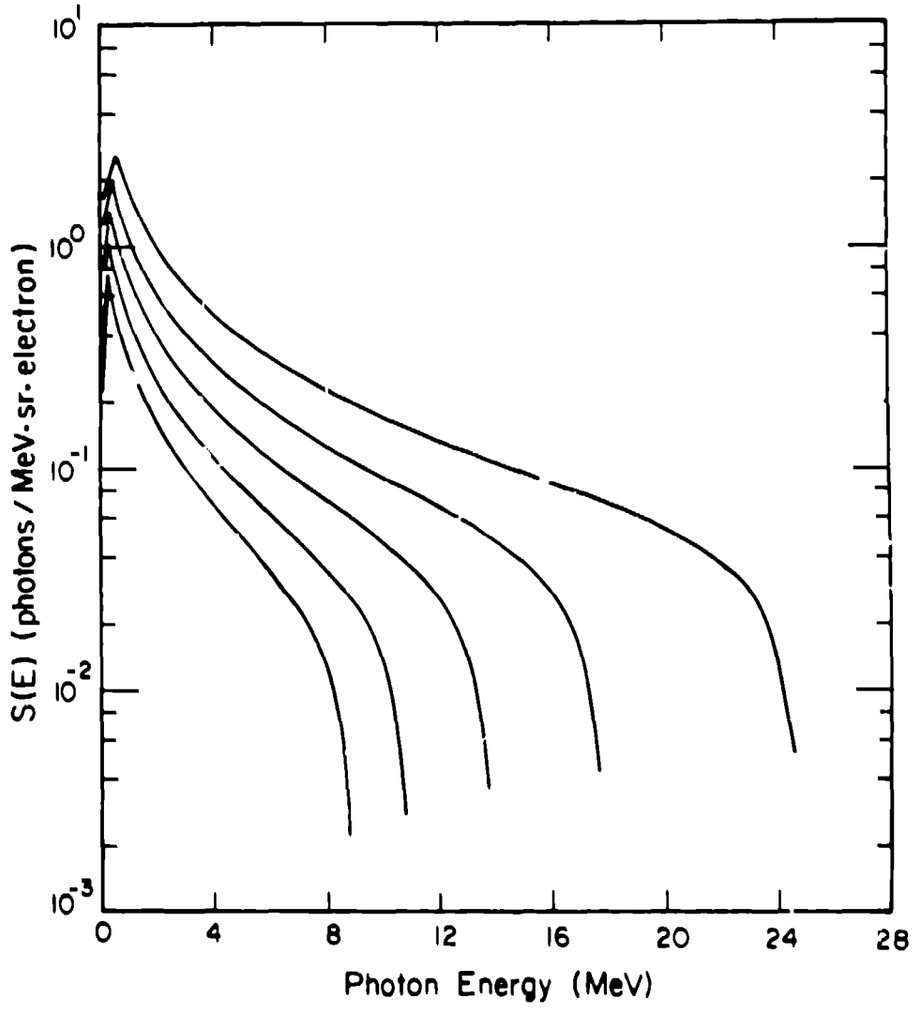

Fig. 1. Bremsstrahlung spectra as a function of electron energy from the Santa Barbara linac.

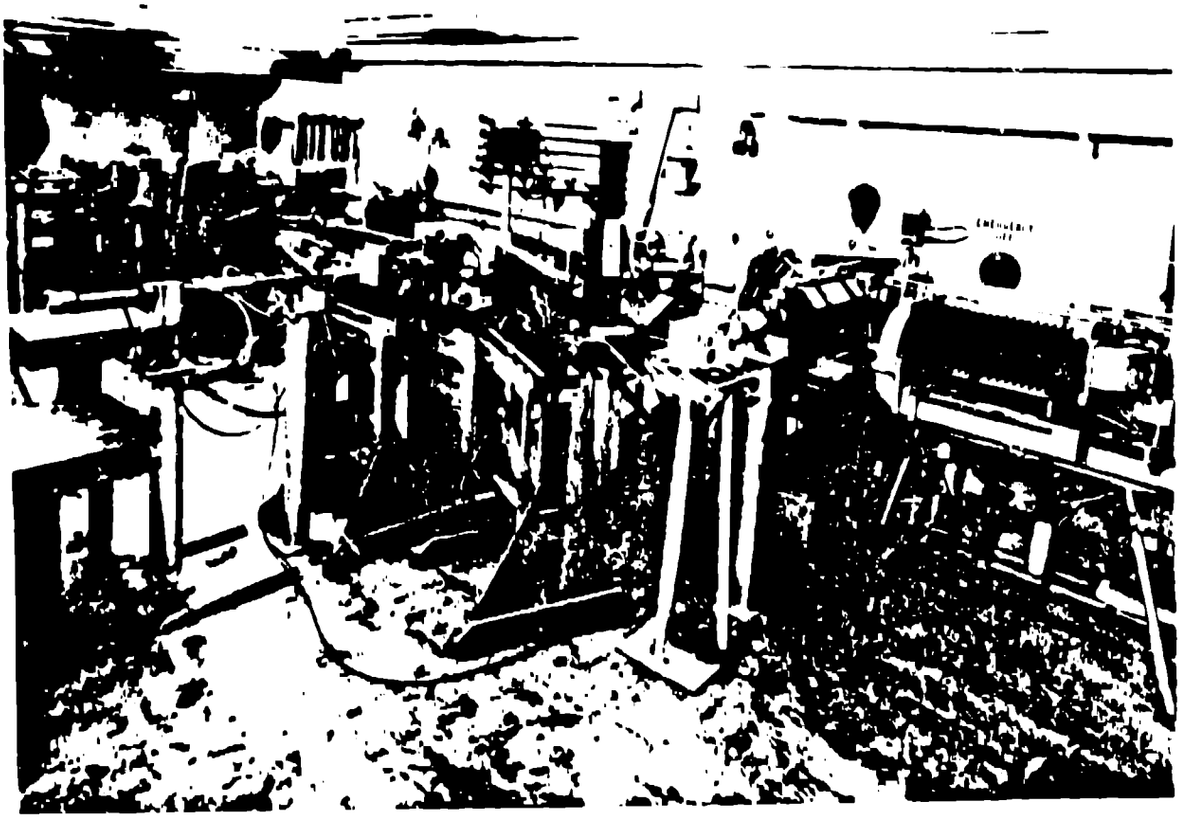

Fig. 2. Photograph of the DOE/EG&G linac.

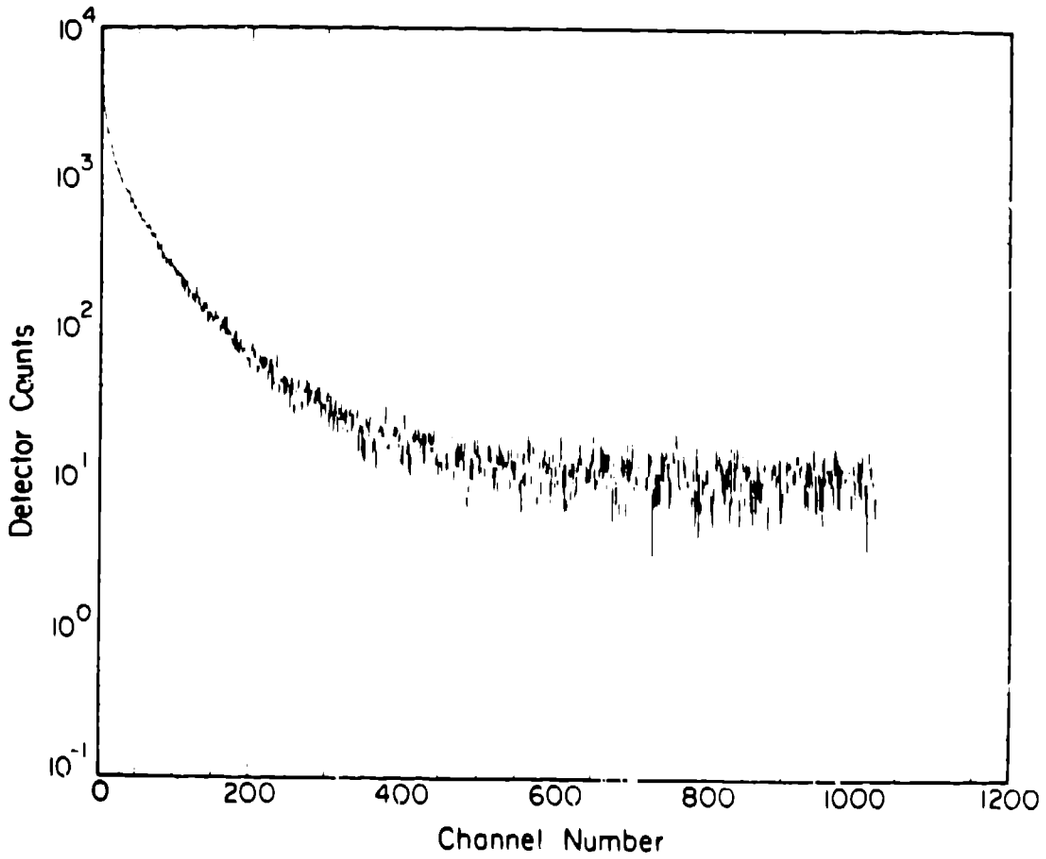

Fig. 10. Delayed neutron signal from 1 g Pu in a waste matrix.

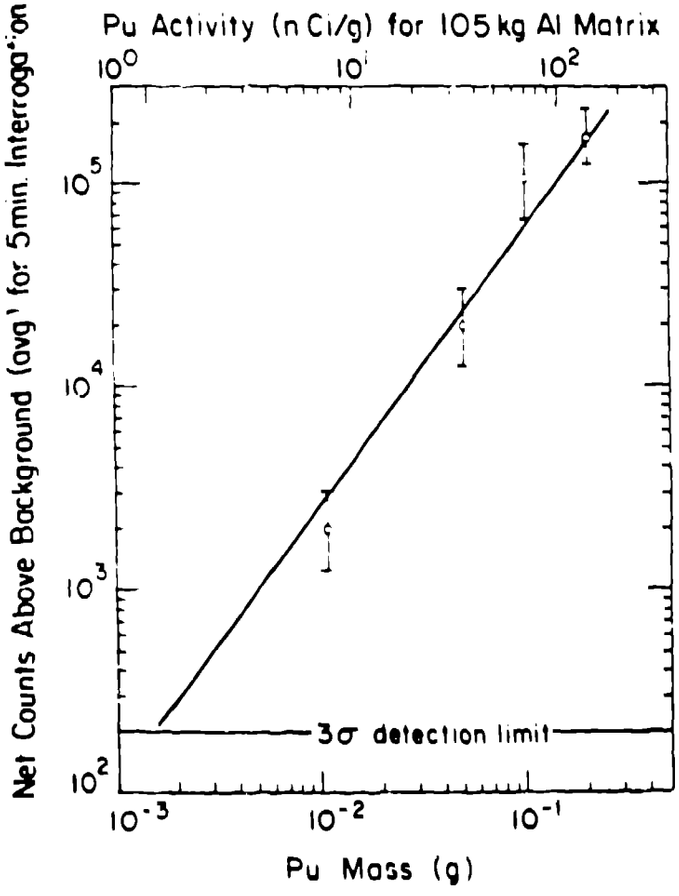

Fig. 11. Delayed neutron counts versus Pu mass in a 105-kg Al matrix.

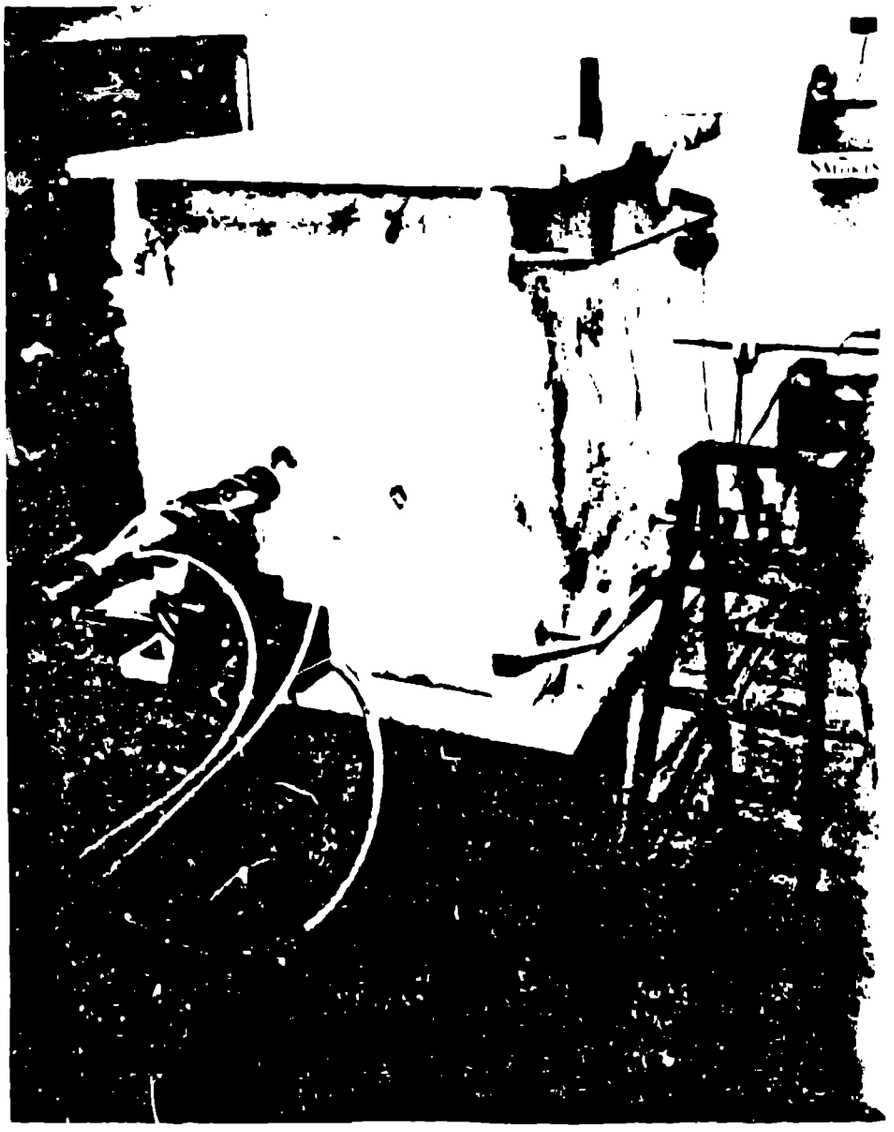

Fig. 12. Differential dieaway detection geometry.

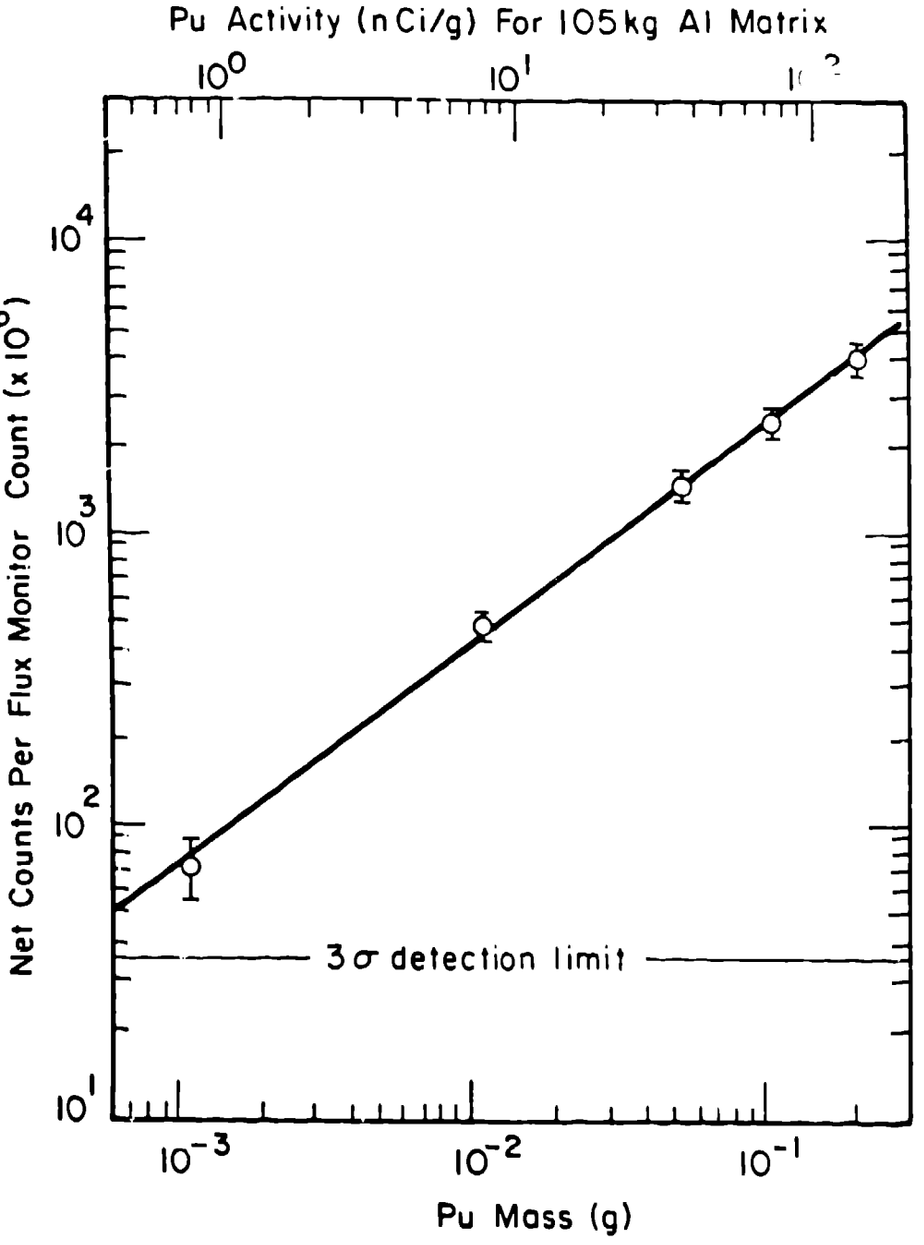

Fig. 13. Neutron counts per flux monitor count versus Pu mass in a 105-kg Al matrix.

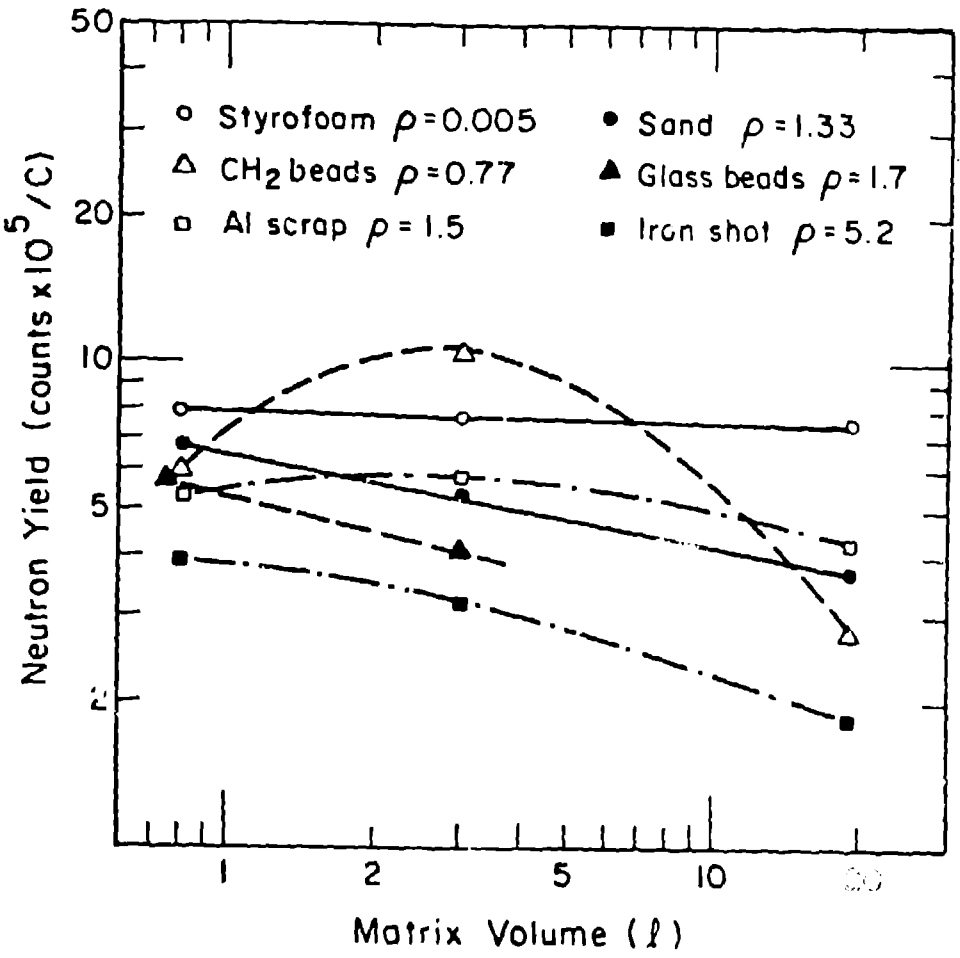

Fig. 14. Relative delayed neutron counts versus mass from Pu for various matrices, beam energies, and matrix masses.

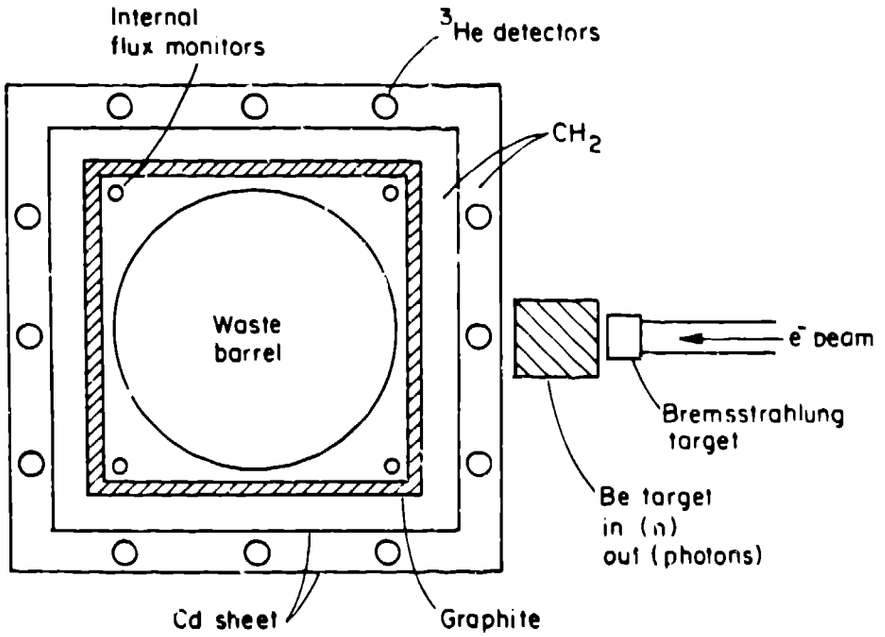

Fig. 15. Schematic layout of an SIM assay detector geometry.