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A DISCUSSION OF AN EXPERIMENTAL EVALUATION OF THE USE OF THE FISSION FRAGMENT TRACK REGISTRATION METHOD FOR PARTICLE FLUENCE MEASUREMENTS

Introduction

Heavy charged particles, such as fission fragments, produce linear trails of radiation damages (tracks) in many solids including insulators like plastic and minerals like mica. The electron microscope can be used to directly observe these tracks which have a diameter on the order of 50 Å. However, a much more useful technique is first to enlarge and fix these tracks by suitable etching procedures; then, the tracks may be counted by optical microscope or in the special case of thin foil detectors by physical counting methods to be described in this report.

The application of these materials to be studied in this report is in the measurement of particle fluence by using the fission reaction. In this method a fissile material (Th in this study) is placed next to the track registration material during irradiation. The number of tracks produced in the insulator is then a function of the fission cross section and total fluence. If the cross section is known, fluence may then be determined.

Track Formation Mechanisms

Any valid model for the production of etchable particle tracks in solids must account for the following characteristics: a) the damage region is narrow (less than 50 Å in diameter) and continuous atomically, or nearly so; b) tracks are not formed in metals and good semiconductors; c) differential materials have different critical rates of energy loss for track formation; and d) tracks are highly stable, and their fading is controlled by the motion of atomic, not electronic defects.[1]

An energetic fission fragment moving through a solid loses energy in two primary ways; exciting and ionizing atoms and at lower energies (at the end of its range) elastic nuclear scattering. Many authors have attributed the tracks either to the direct production of displaced atoms in elastic collisions or to thermal effects (thermal spike model[1]).

Atomic displacements due to these elastic collisions are not the cause of the tracks because they would be (a) expected to occur equally in conductors or insulators, and (b) become more prevalent near the end of the range of a charged particle because of the larger elastic nuclear scattering cross section. However, experiments show that tracks do not appear in conductors and often do not register near the end of the particle range even in semiconductors.

The other possibility, the thermal spike, has also led to invalid predictions and fails to account quantitatively for certain track features. For example, Chadderton and Montagu-Pollock have used the thermal spike (See Ref. 1) model to predict that intrinsic semiconductors should be the best track registering materials -- in contradiction to experimental results.

One other alternative source of tracks can be shown not to apply generally -- the direct effect of displaced electrons. If tracks consisted merely of displaced electrons, it would be expected that the subsequent migration of electrons through the solid would allow them either to diffuse away or to return to their vacated sites. Erasing the track in either case. Note that at 25°C tracks are stored longer than the time necessary for electrons to diffuse over a distance of more than 1 mm, which is at least 4×10^5 track radii.

The ion explosion spike model[2] is currently the model showing the most promise. It postulates that the electrostatic repulsion of the positive ions created by the removal of electrons ejects these ions into displaced positions, thus producing high concentrations of interstitials and vacancies along the particle trajectory. Figure 1 is a schematic depicting the process; (a) indicates the ionized trail, and (b) the subsequent production of lattice defects. If the time between (a) and (b) is insufficient to neutralize the ionized trail, either by arriving electrons or departing holes, and if the forces are sufficient to produce about one atomic displacement per atom plane, tracks can be formed by this mechanism.

Semiquantitative development of this model has led to the following comparisons with observed track characteristics:

(1) The strained and chemically reactive nature of a track is a natural consequence of a region with high concentrations of vacancies and displaced atoms (interstitials).

(2) The size of tracks (<50Å for fission fragments) is compatible with the theory.

(3) The finding of tracks only in materials of low hole mobility is consistent with the requirement that the ionization created by a passing charged particle must exist long enough allow ions to seek out interstitial positions.

(4) The measured energy loss thresholds for track production corresponds approximately to that calculated by the model when assuming one ionization produced per atomic plane crossed by the track producing particle.[2]

Thus, the ion explosion spike model appears to be consistent with known properties of charged particle tracks. More recent work has shown several faults in this model and more comprehensive models have been developed.[3] However, the track description offered by this model is sufficient for the purposes of this report.

Track Development Process

Because of the large free energy associated with the disordered structure, radiation damage trails are more chemically reactive than normal material. For this reason, if a substance containing damage tracks is immersed in a solvent, the track area will dissolve much more rapidly than the undamaged material resulting in a greatly enlarged track region easily visible in an optical microscope. In thin foil detectors this etching can result in actual holes in the detector material. Electric sparks, when passed through the holes, enlarge them and darken the area around them so they are visible to the naked eye. In addition, the sparks open up new holes corresponding to partly etched-through tracks. The size of the tracks after etching is greatly dependent upon the type of solvent used, the etching time and the temperature.

The etching process has been noted to take place in three stages:

(1) Immediately upon immersion the solvent penetrates each track and reacts with the track material producing a hollow well-defined channel.

(2) With increased time the rate of increase of channel width is at first negligibly small. This incubation time is probably the time it takes to remove the reaction products from the cores of the damaged regions and to supply fresh solvent to the sides of the channels.

(3) In the third stage the channels widen at a nearly constant rate.

Since these stages involve diffusion it is clear that the process should be temperature dependent.[4]

The procedure for developing tracks followed in all experimental results presented in this study was as follows. First, the foils were submerged in a 5 normal solution of KOH for 60 minutes at $60^{\circ}C$; the solution was stirred all the while. Then the foils were washed in water and then allowed to dry. Finally, the foils were placed in the counter at 1500 volts where sparks were passed through the damage regions.

Track Detection Methods

The method of counting tracks used most often in the past was scanning with an optical microscope. However, this method requires a long time to count hundreds of tracks and high track densities for statistical accuracy since only small areas can be scanned in a practical time period. The practical limit for microscope counting is 10^3 to 10^6 tracks/cm^2. Several faster methods of track registration have been tried; most of these involve thin film detectors where the tracks extend through the film so that actual etched holes are what must be detected.

The first attempts at measuring the hole densities in these thin film detectors involved physical measurements such as gas flow or ionic permeability. Bean, Doyle and Entine[1] have obtained reproducible curves of cell conductivity as a function of time for several irradiated mica specimens used as a barrier between two halves of a cell containing a HF solution. An alternate method which works well if the density of holes is very small is to coat the film with Al on one side and etch with hydroxide solution from the other. Wherever a track is present, the hydroxide flows through and attacks the aluminum coating, making a hole in the Al large enough to be seen and counted by the naked eye. One other physical method is to place a membrane filter against the film and force a dye solution through the holes from the other side. The dye spreads out sideways in the filter, making spots that are large enough to be seen.[5]

A different approach is to place the etched insulating film against a grounded, plane electrode and scan across it with a knife edge at high potential. As the knife edge passes each hole a spark goies through the hole, enlarging and arkening the area around it. These sparks can be counted electrically but the density of holes that can be counted is limited because of the difficulties of resolving holes that are close together.

A similar method which is extremely fast and appears to be capable of counting higher densities of holes than any of the above methods has been developed by Cross and Tommasino[5] and is the subject of the experimental investigation presented in this report.

Figure 2 shows the electrical circuitry. The etched insulating film is placed between a plane electrode and a thin layer of Al evaporated on a Mylar backing. When a high voltage is applied a spark passes through one of the holes in the film, discharging the capacitor and evaporating a hole in the Al layer. By the time the capacitor has recharged (RC = 2msec) a second spark cannot discharge through the same hole because the path to the edge of the evaporated hole in the Al is too long. Therefore, the next spark must pass through another hole repeating the process until sparks have passed through all holes, then the discharge stops. A scaler can be used to directly count the current pulses of the sparks. The resulting holes in the Al provide a pattern of the holes in the etched film which can easily be seen by the naked eye.

To ensure that a second spark does not discharge through the same hole it is necessary to match the voltage and capacitance to the thickness of the Al layer. The required hole size to be evaporated in the Al is determined by the operating voltage since the distance a spark may gap is a function of voltage. The actual size of the hole evaporated in the Al is dependent both upon the rate of the energy transfer ($P = CV^2t^{-1}$) and the duration of the spark which are determined by the resistance (100Ω), capacitance and high voltage. A third consideration is that the duration of the spark is long enough to be counted by the scaler. Thus, to operate successfully the circuit must provide a spark which is powerful enough to evaporate a hole in the Al, large enough to prevent further sparking in that same hole and of long enough duration to be counted by the scaler. In addition, the holes in Al should not be too large or high track densities could not be accurately counted due to the overlapping of the Al holes.

Figure 3 shows two typical plateau curves for different track densities. Between 450 volts and 500 volts the variation of the number of counts is about 10% or less. One complication is the fact that the plateau as illustrated in Figure 3 is dependent on the capacitance. For example, we found that increasing capacitance from 2nF to 4 nF gave a 10% variation of counts over 25 volts instead of the above mentioned 50 volts.

As can be seen a delicate balance between voltage, capacitance and resistance is required for proper operation. The values given in Figure 2 were found experimentally to work satisfactorily.

Evaluation of Thin Film Electric Counter Technique

Theory: Consider a plane uniform source of fission fragments emitted isotropically. The source thickness is $\geq \bar{R}$,

where

$\bar{R}$ = average range of fission fragments g cm^{-2} in the source material. For thorium, $\bar{R} = 10.3 \times 10^{-3} gcm^{-2}$ has been chosen.[6]

$\bar{P}$ = average probability that one fission fragment created in the source escapes from one surface. By a simple geometrical consideration it can be shown that $\bar{P} = 0.25$. (As long as source thickness $\geq \bar{R}$, and fission event emits fragments isotropically.

$\phi(E)$ = differential particle fluence (cm^{-2}, MeV^{-1})

$\sigma(E)$ = fission cross section per nucleus (barn)

N = Avogadros number

A = atomic weight

B = number of fission fragments per cm^2 escaping from the semi-infinite fission foil of thickness $\geq \bar{R}$

ϵ = efficiency of fission fragment registration and counting

T = measured track density (cm^{-2})

The number of fissions per cm^2 of target material caused by neutrons in the energy range $(E, \Delta E)$ is

$$\frac{N}{A} \bar{R} \ \phi(E) \sigma(E) \ 10^{-24} \ dE \qquad (1)$$

and the number of fission fragments escaping one surface of the source is

$$B = \bar{P} \ \frac{2N \ \bar{R}}{A} \ 10^{-24} \int_{E_1}^{E_2} \phi(E) \sigma(E) \ dE \qquad (2)$$

$$T = B \epsilon = \epsilon \bar{P} \ \frac{2N \ \bar{R}}{A} \ 10^{-24} \int_{E_1}^{E_2} \phi(E) \sigma(E) \ dE \qquad (3)$$

Define the effective cross section of the fissile material as:

$$\sigma_{eff} = \frac{\int \phi(E) \ \sigma(E) \ dE}{\int \phi(E) \ dE} \qquad (4)$$

Also define the total fluence as:

$$\Phi = \int \phi(E) \ dE \qquad (5)$$

Then

$$T = \epsilon \bar{P} \ \frac{2N \ \bar{R}}{A} \ 10^{-24} \ \sigma_{eff} \Phi$$

$$= 1.33 \times 10^{-5} \epsilon \sigma_{eff} \Phi \qquad (6)$$

For isotropic fission fragment emission the value 1.33×10^{-5} is determined solely by the fissile material used and is a characteristic of the type of insulating film and track counter used. Therefore, for a given track registration - counter system we have T directly as a function of Φ .

Experimental Measurement of ϵ :

To measure ϵ a 10 Ci Pu-Be neutron source was used. For ^{232}Th and the Pu-Be spectrum σ_{eff} has been determined in reference 7 to be: $\sigma_{eff} = 0.14$ barn. Thin (10μm) polycarbonate insulating film detectors (area = 1.27 cm^2) coupled with ^{232}Th were placed on a device such that they rotated about the 10 Ci Pu-Be source at a radius of 40 cm. The yield of the source is given as $2.05 \times 10^7 \frac{n}{s}$. With this knowledge Φ is easily calculated for a given run time. Experimental results follow.

Theoretical No. of Tracks per cm^2 ($= 1.33 \times 10^{-5} \sigma_{eff} \Phi$)	No. of Counts per cm^2 (=T)	$\epsilon = \frac{T}{1.33 \times 10^{-5} \sigma_{eff} \Phi}$
41	17	0.423
72	37	0.434
113	49	0.434
291	124	0.425
643	266	0.414
1094	336	0.308
2547	794	0.312

These results are plotted in Figure 4. It is seen that ϵ remains essentially constant over a range below 650 counts/cm^2. It is quite convenient that this is the range over which this method works best since microscopic counting is applicable from the top of this range up to about 10^6 counts/cm^2. With a larger area detector ($\sim$10 cm^2) it appears feasible to count track densities as low as 0.5 tracks/cm^2. Thus, the applicable range of the fission fragment track registration method is seen to cover six orders of magnitude.

Minimum Detectable Fluence (Φ) For Personnel Dosimetry

It has already been shown that,

$$\Phi = \frac{T}{1.33 \times 10^{-5} \epsilon \sigma_{eff}} \quad \frac{\text{neutrons}}{\text{cm}^2} \qquad (7)$$

Plans are to design a detector system using 10 cm^2 of ^{232}Th together with 10μm thick polycarbonate (Kimfol). Experiments indicate that the background for one month's exposure in this sytem will be less than 1 track. (The half-life for spontaneous fission is about 2.10^{18} years, corresponding to about 1 fission/month, cm^2. With the discussed efficiency this will, of course, give a negligible background.)

Given a background that is essentially zero, it should be possible to detect and interpret as low as 5 tracks. This corresponds to T = 0.5 tracks/cm^2 for a 10 cm^2, ^{232}Th foil. The minimum detectable neutron fluence for Pu-Be spectrum neutrons is the, from equation 7

$$\Phi = \frac{0.5}{(.42)(1.33 \times 10^{-5})(.14)} = 6.1 \times 10^5 \text{ n/cm}^2$$

The absorbed dose in tissue to fluence conversion for 4.5 MeV neutrons is 4.5×10^{-9} rad cm^2, which yields

$$(6.1 \times 10^5)(4.5 \times 10^{-9}) = 2.75 \text{ mrad}$$

For Th, photofission is a competing process and the cross section is significant over a wide energy range and is estimated to be about 3 mb for 8 MeV photons[7]. Using this value, a minimum detectable fluence of photons can easily be calculated.

$$\Phi = \frac{T}{1.33 \times 10^{-5} \epsilon \sigma_{eff}}$$

$$\Phi = \frac{0.5}{(.43)(1.33 \times 10^{-5})(0.003)} = 2.9 \times 10^7 \text{ quanta/cm}^2$$

This corresponds to an energy fluence of

$$(8 \text{ MeV})(2.9 \times 10^7 \frac{1}{\text{cm}^2}) = (2.32 \times 10^8) \frac{\text{MeV}}{\text{cm}^2}$$

The energy absorption is now

$$(2.32 \times 10^8 \frac{\text{MeV}}{\text{cm}^2}) (\mu_{en} \frac{\text{cm}^2}{\text{gm}}) (1.6 \times 10^{-6} \frac{\text{erg}}{\text{MeV}}) (10^{-2} \frac{\text{rad-gm}}{\text{erg}})$$

For 8 MeV photons with water as the absorbing medium,

$$\mu_{en} = 0.018 \text{ cm}^2/\text{gm}$$

and the absorbed dose corresponding to the minimum detectable fluence is:

$$(2.32 \times 10^8)(0.018)(1.6 \times 10^{-6})(10^{-2}) = 67 \text{ mrad.}$$

Conclusion

The spark counter method of counting tracks in thin polycarbonate films is fast, uses simple equipment and has sufficient accuracy for many measurements. The ratio of the number of counts actually recorded to the number of fission fragments theoretically expected to hit the thin film was determined to be $\epsilon = 0.43$ for track densities below 650 tracks/cm^2. Optical counting under a microscope becomes a practical method above 10^3 tracks/cm^2 and gives an efficiency $\epsilon = 0.80$.

The difference in counting efficiency is due to the fact that only tracks that penetrate the thin film can be electrically counted.

For dosimetric application it was shown that the minimum detectable track density in a 10 cm^2 Th thin film detector corresponds to 2.8 mrad Pu-Be neutrons or 67 mrad 8 MeV photons.

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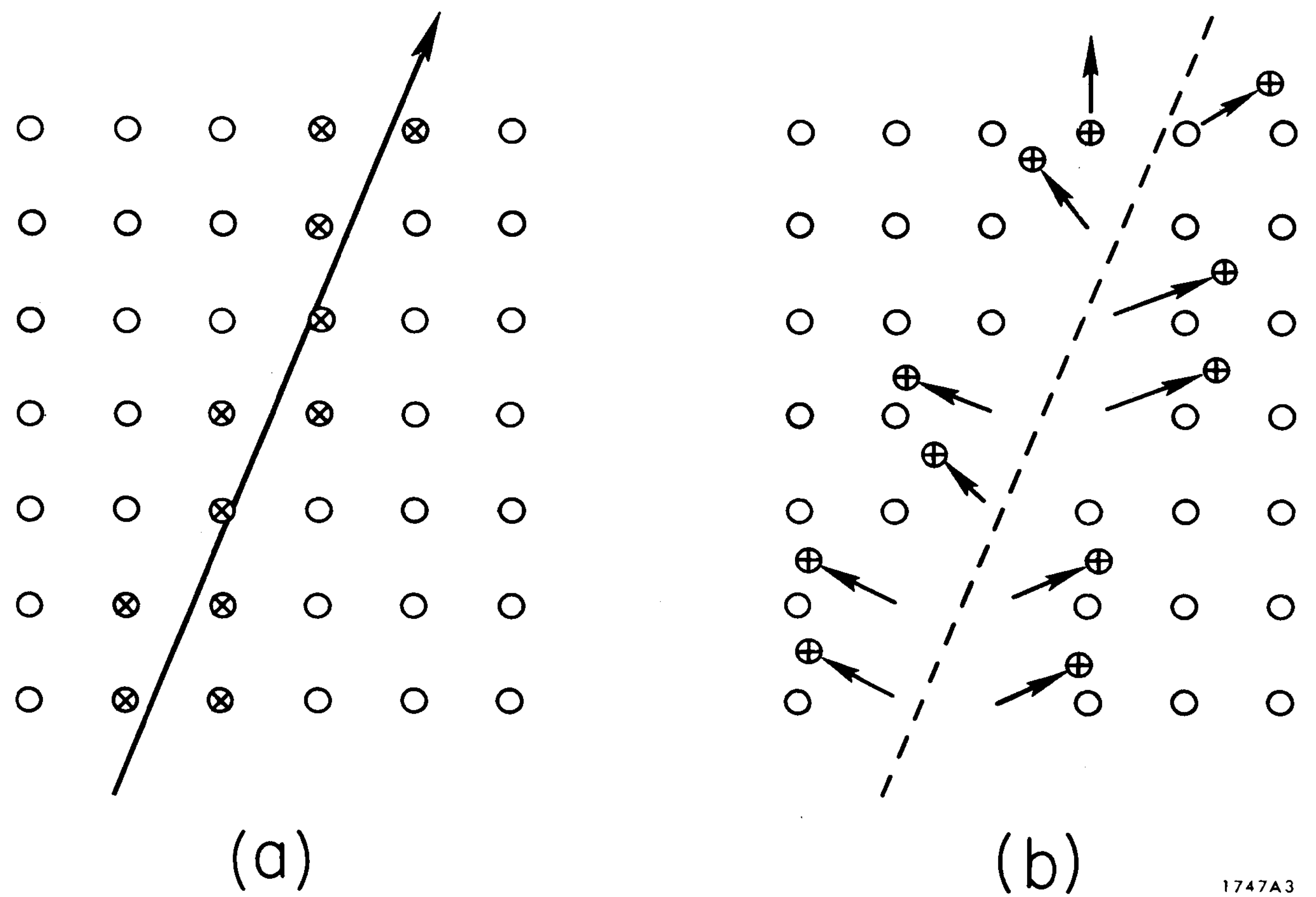

Fig. 1 - An illustration of the ion explosion spike process as described in ref. 2.

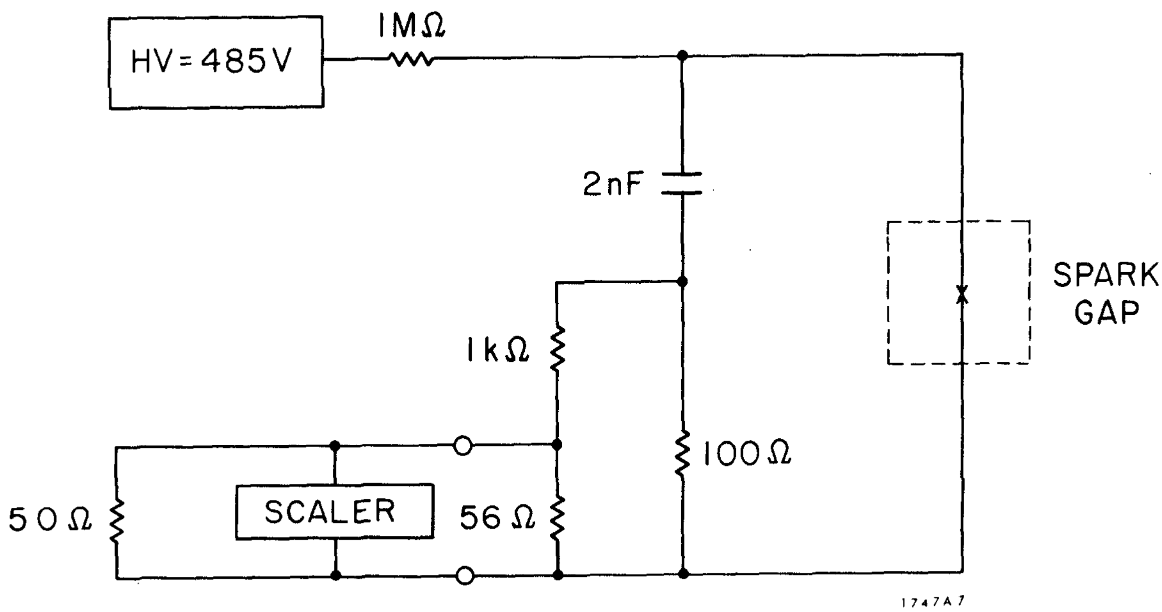

Fig. 2 - The electrical circuitry used in this study.

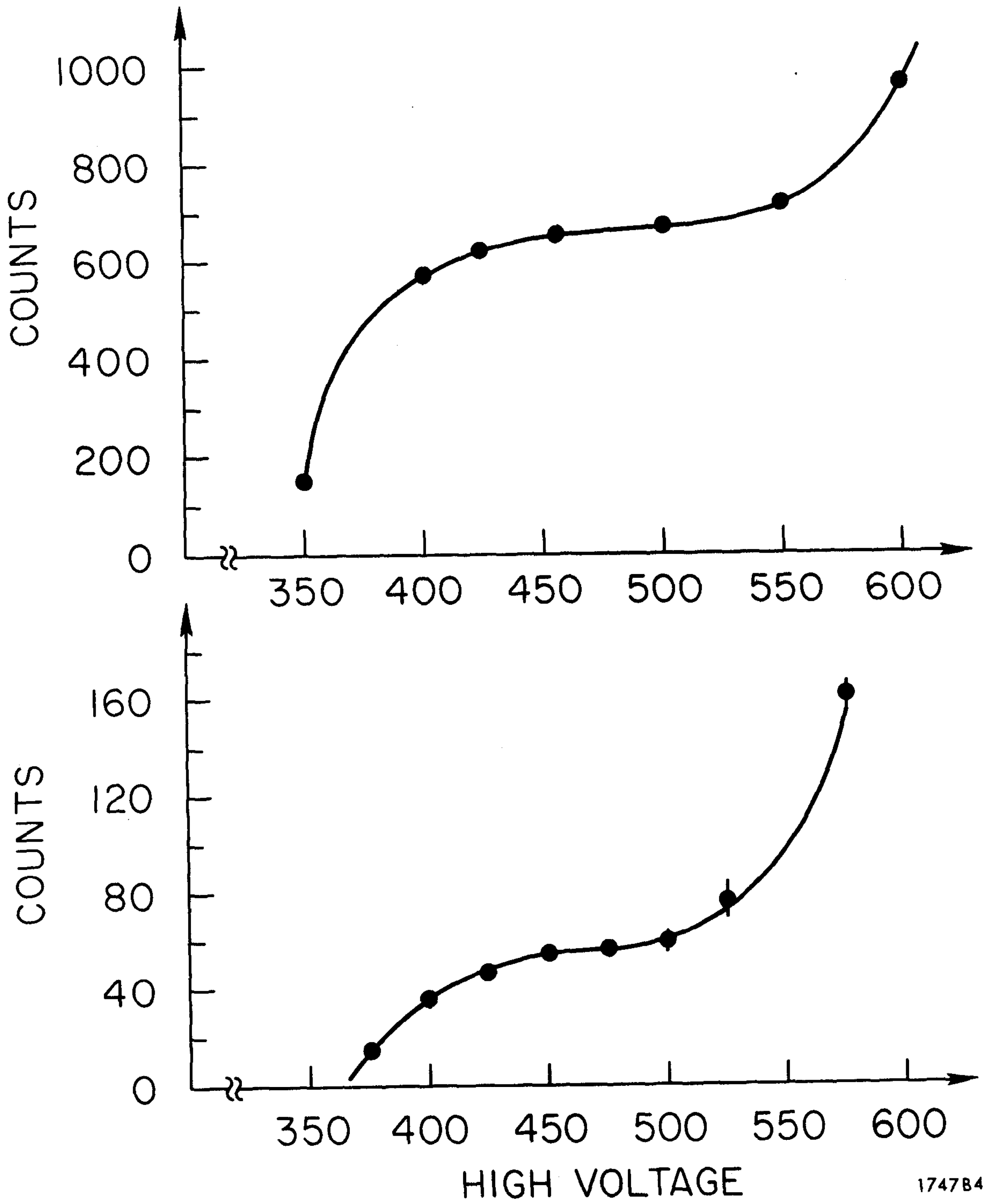

Fig. 3 - Typical plateau curves (counts vs. HV) for low and high track densities.

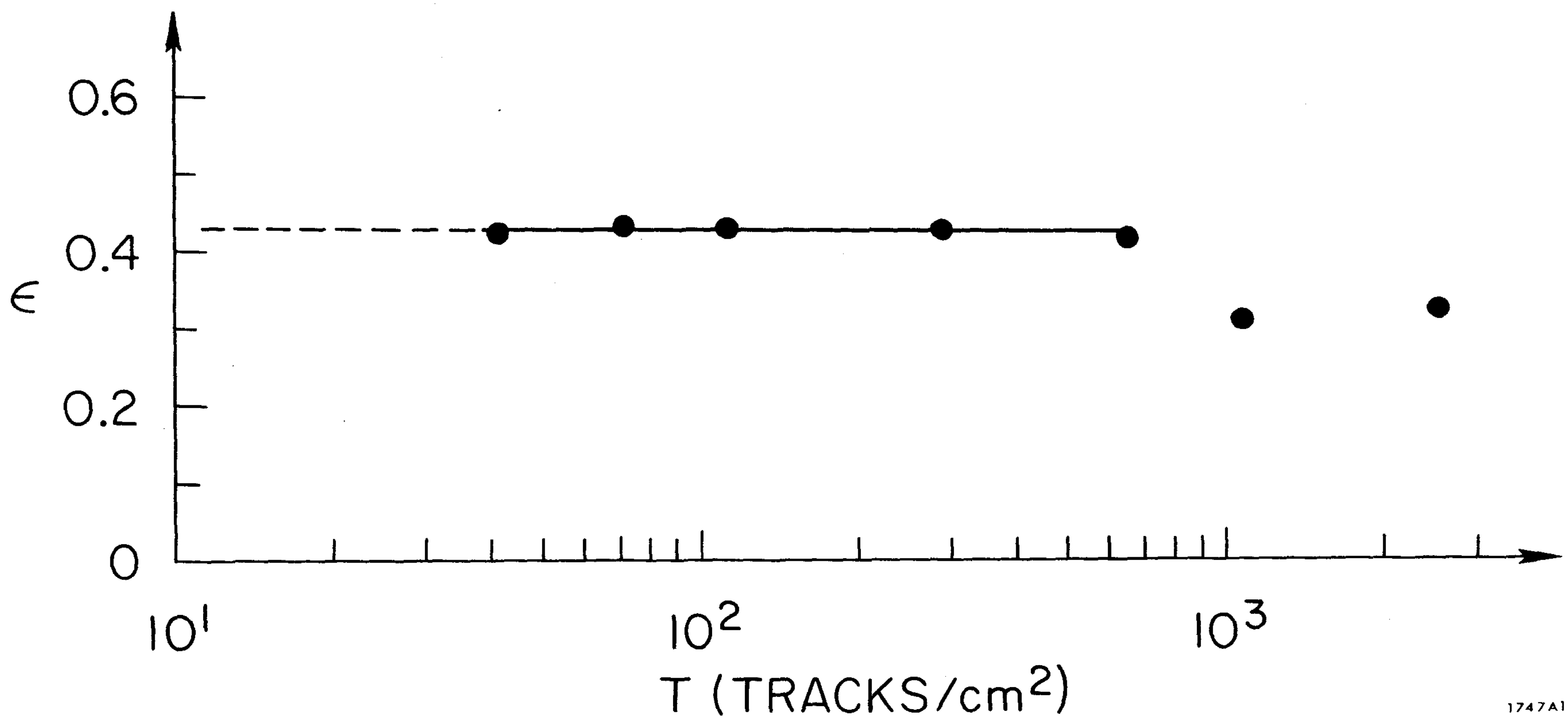

Fig. 4 - The fission fragment registration efficiency (ϵ) versus track density.

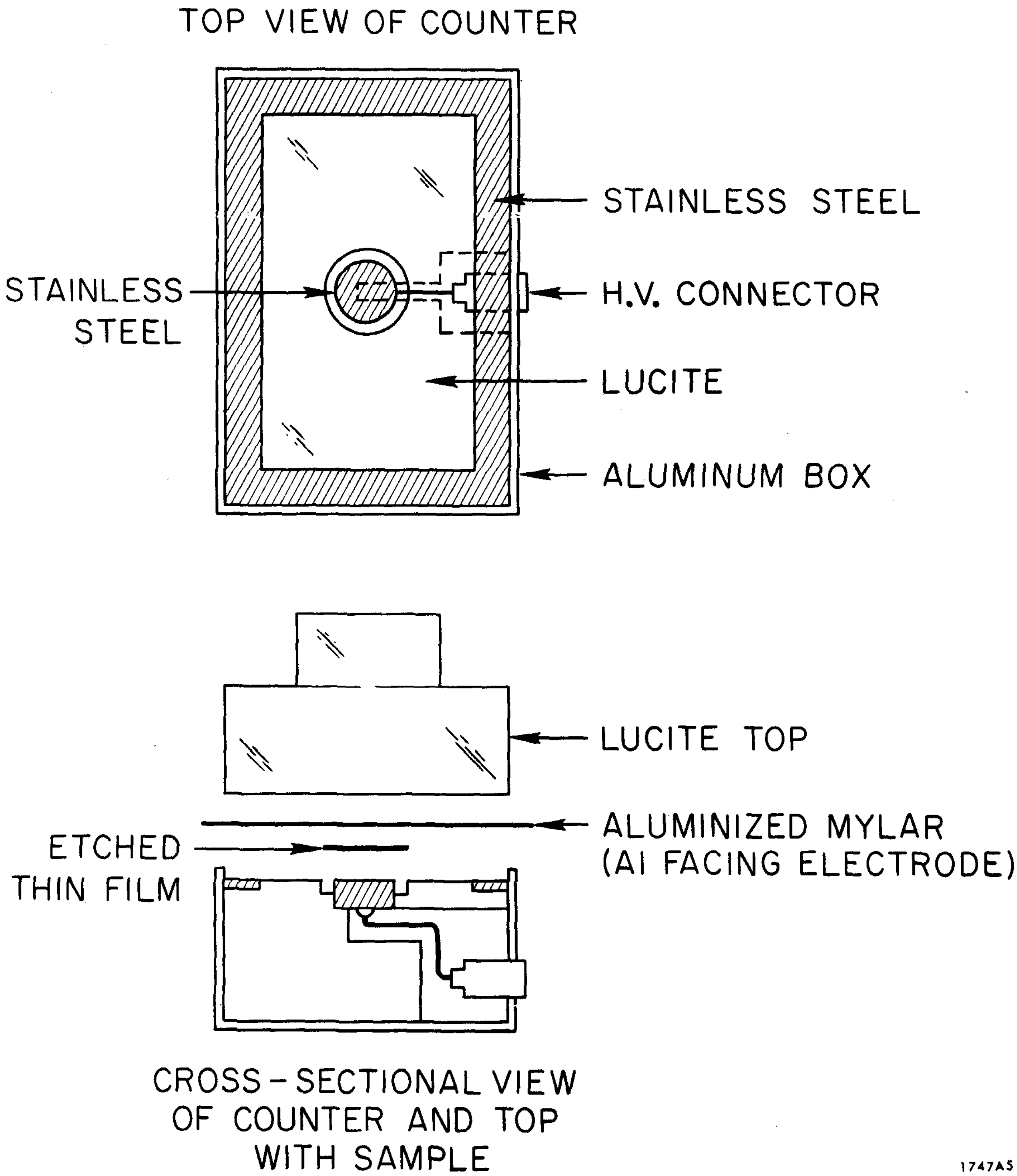

Fig. 5 - Schematic of the counter being used in this experiment.