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FISSION YIELDS MEASURED WITH TARGET MATERIALS IN CONTACT WITH SOLID STATE TRACK DETECTORS

by

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Abstract - The configuration of the contact of fissionable target materials with dielectric track detectors is reviewed to obtain fission yields corrected for fragment self-absorption in fission sources of different thicknesses and for optical magnification for the observation of etched fission tracks. Total detection efficiency and "effective" thickness of the target sample are obtained in the case of formation of cone-shaped tracks by etching. A number of useful formulae for evaluation of fission yields in both induced and spontaneous fission experiments is reported. The method can be extended and applied also to fission-related problems, natural and induced emission of nuclear fragments, and nuclear reaction studies as well.

Key-words: Fission fragments; Track-etch detectors; Fission yields; Thin and thick targets; Detection efficiency.

1. INTRODUCTION

The procedure of contacting a target material with a dielectric track detector has been widely used in investigations of fission phenomena and in many applications to fission-related problems (Gold *et al.* , 1968; Roberts *et al.* , 1968; Fleischer *et al.*, 1975; Durrani and Bull, 1987). In these experiments fission fragments are recorded as trails of damage formed along the fragment path in the dielectric material, and after development by an appropriate etching procedure these latent images become visible at the level of ordinary optical microscopy as fission tracks. The fraction of latent tracks revealed by etching defines the etching efficiency, ε_e, which depends essentially upon the rates of chemical attack along the track (track etching velocity, v_T) and the undamaged detector material (general etching velocity, v_G). In addition, identification of etched fission tracks depends strongly upon the magnification of the optical system used for observation of tracks, in the sense that not all the tracks revealed by etching are actually observed. This makes it necessary to introduce an additional efficiency factor, ε_i, related to visualization (identification) of the etched fission tracks. The total, combined efficiency (detection efficiency, ε) is clearly given by $\varepsilon_e \varepsilon_i$.

Although fission fragments (or, alternatively, fission events) are produced in the whole volume of the target sample material, only a fraction of such fission events is visualized as fission tracks in the detector surface. Thus, there is an additional effect to be considered in obtaining fission yields, namely, the effect of fragment self-absorption in the fission source material. As fission fragments move towards the detector from a point-origin inside the sample, their kinetic energy is degraded along their trajectories and, thus, depending on sample thickness and fragment direction, only a fraction of the total number of fission fragments produced in the sample will be detected. Gold *et al.* (1968) were the first ones to place on a precise absolute basis fission rate measurements with solid-state track recorders, where the efficiency and sensitivity of their method were determined and discussed for both pre-etched mica and makrofol track detectors.

In a previous paper (Tavares, 1991) a method to obtain fission yields in experiments which use target materials in the configuration of contact with dielectric track detectors has been developed where the various effects mentioned above have been considered for detector materials of high etching efficiency for registration of fission fragments (micas and plastics in general), i.e., for the cases where the condition $v_T \gg v_G$ occurs. In the present work this method is extended to the cases of $v_T > v_G$. This condition is verified, for instance, for detection of fission fragments with glasses, or nuclear fragments of low rate of ionization in plastics. In these cases the geometrical aspect of the etched tracks under an optical microscope is of a cone, the etch pit opening being circular or elliptical, depending upon the normal or oblique incidence of the fragments on the detector surface. Thus, etch pits will be identified as etched nuclear tracks if their measured diameters (or minor-axes) are greater than a certain minimal etch pit opening allowed by the magnification of the optical system for observation of tracks. This limiting condition for visualization and identification of the etched tracks, associated with the effect of fragment self-absorption in the target material, introduces important corrections to be taken into account in deducing the appropriate yield formulae.

In the recent past, different methods have been developed to obtain corrections to fission yields measured by using the target-detector 2π-geometry (Dietz and Rassl, 1982; Guo *et al.* , 1982; Durrani and Bull, 1987; de Lima *et al.* , 1988). The most recent publication in these efforts is the ASTM (1990) Standard. In the present work an alternative approach is given for the various situations in which very thin, thin, and thick targets are used in intimate contact with dielectric detectors, and results apply to cases where fission tracks are identified from the etch pit opening.

2. BASIC QUANTITIES AND ASSUMPTIONS

In induced fission experiments the fission yield (or cross section, σ) is obtained by

$$\sigma = \frac{N_e}{Q\ N_a} \quad , \tag{1}$$

where N_e is the number of fission events produced per unit volume of the sample, Q is the number of particles perpendicularly incident to the sample surface per unit area, and N_a is the number of fissionable target nuclei per unit volume, provided the values of σ and N_a are such that they do not cause any significant attenuation of the particle beam. For spontaneous fission studies it suffices to replace the quantity σQ with the new one, $\lambda\tau$, where λ denotes the decay constant, and τ is the exposure time. If a dielectric track detector is used in close contact with a target sample (2π-geometry) only a fraction of the N_e fission events will produce a number of fission tracks, N_t, observed per unit area of the detector. The relationship between N_t and N_e will depend upon i) sample thickness, x_0; ii) rate of energy loss (essentially due to ionization) of the fission fragments in both the target and detector materials; iii) average residual range of full energy of the median fission fragment in both the target and detector materials (respectively, $\bar{a}_0$ and $\bar{r}_0$); iv) thickness of the surface layer of detector material removed by etching, $v_G t$ (t is the etching time); and v) minimal etch pit opening, d, capable of being observed under given optics. The following simplifying assumptions have been made in deducing the yield formulae: i) the target sample is homogeneous, and nuclear fragments are emitted isotropically from point-origins distributed uniformly inside the sample; ii) the distribution of fission fragments is replaced by its most probable (or median) fission mode; iii) latent fission tracks are revealed by etching for all those fission fragments which have crossed the target-detector interface at angles greater than their critical angles of etching; both v_G and v_T are assumed constant, with $v_T > v_G$; iv) the rate of energy-loss of fragments in both the target and detector materials is approximately described by a relationship of the form (Northcliffe and Schilling, 1970; Tripier *et al.*, 1974; Almási and Somogyi, 1981)

$$- \frac{dE}{dy} = \zeta E^{\beta} \quad , \tag{2}$$

where E is the kinetic energy of the fragment, and ζ and β are constant ($0 < \beta < 1$). The constant β may be considered as much the same for both media, and $\zeta_{target} > \zeta_{detector}$. The residual ranges result, therefore, proportional do $E^{1-\beta}$, and the average full residual ranges in the target and detector materials are given, respectively, by

$$\bar{a}_0 = \frac{E_0^{1-\beta}}{(1-\beta)\zeta_{\text{target}}} \quad \text{and} \quad \bar{r}_0 = \frac{E_0^{1-\beta}}{(1-\beta)\zeta_{\text{detector}}} \quad , \qquad (3)$$

where E_0 is the full kinetic energy of the median fission fragment.

3. FISSION YIELD FORMULAE FOR $v_T > v_G$

Figure 1 shows schematically the arrangement of a target material placed in close contact with a dielectric detector, where the various quantities used throughout the text have been defined. From equations (3) and Fig. 1(a) it follows that

$$x = \bar{a}_0 \left(1 - \frac{r}{\bar{r}_0}\right)\sin\phi \quad . \qquad (4)$$

The number of fission fragments produced in a sample layer of thickness dx and within the solid angle $d\Omega = 2\pi\cos\phi d\phi$ is given by

$$\frac{d^2N}{d\Omega dx} = \frac{N_e}{4\pi} \times 2 \ . \qquad (5)$$

Therefore, the number of etched fission tracks which are observed per unit area of the detector surface is obtained by

$$N_t = N_e \int_0^{x_M} \int_{\phi_1}^{\phi_2} \cos\phi d\phi dx = N_e \int_0^{x_M} (1-\sin\phi_1) dx \ , \qquad (6)$$

where $\phi_2 = \pi/2$, since tracks which are formed from normal incidence of fragments on the detector surface can be seen as circular-shaped tracks. ϕ_1 is the dip angle which allows for visualization of etched tracks down to the threshold etch pit opening d, and x_M

(the maximum of x) is such that a fission fragment which has entered the detector can still be revealed and observed.

The limiting values ϕ_1 and x_M can be evaluated from Fig. 1(b) and the track etching geometry for a track incident at an arbitrary dip angle with the detector surface, where the assumption is made of constant track etching velocity v_T (see e.g. Durrani and Bull, 1987). Thus, we have

$$r = v_T t \tag{7}$$

$$d = 2v_G t \left[\left(\frac{v_T \sin\phi_1}{v_G} - 1 \right) \Big/ \left(\frac{v_T \sin\phi_1}{v_G} + 1 \right) \right]^{1/2} , \tag{8}$$

which combine with equation (4) to give

$$\sin\phi_1 = \frac{x}{\bar{a}_0} + \frac{1+B}{A(1-B)} , \tag{9}$$

where

$$A = \frac{\bar{r}_0}{v_G t} , \quad B = \left(\frac{d}{2v_G t} \right)^2 . \tag{10}$$

The value of x_M is obtained from the extreme condition $\phi_1 = \pi/2$ to be inserted into equation (9), i.e.,

$$x_M = \bar{a}_0 \left[1 - \frac{1+B}{A(1-B)} \right] . \tag{11}$$

We can now integrate (6) by making use of the results expressed by equations (9) and (11), where the two different cases must be considered:

i) $x_0 \geq x_M$. In this case the maximum of x is x_M, and equation (6) gives

$$N_t = \frac{1}{2} N_e \frac{x_M^2}{\bar{a}_0} . \tag{12}$$

ii) $x_0 < x_M$. Here the maximum of x is obviously x_0, and thus equation (6) gives

$$N_t = \frac{1}{2} N_e \frac{x_0}{\bar{a}_0} (2x_M - x_0) \quad . \tag{13}$$

Inserting these two results into the general expression for the cross section (equation (1)), one obtains

$$\sigma = \frac{N_t}{QN_a \frac{1}{2} \frac{x_M^2}{\bar{a}_0}} , \quad x_0 \geq x_M \tag{14}$$

and

$$\sigma = \frac{N_t}{QN_a \frac{1}{2} \frac{x_0}{\bar{a}_0}(2x_M - x_0)} , \quad x_0 < x_M \tag{15}$$

It is straightforward to see that a target sample having a thickness $x_0 = x_M$ will produce the maximum number of tracks observed per unit area of the detector,

$$(N_t)_{max} = \frac{1}{2} \sigma QN_a \frac{x_M^2}{\bar{a}_0} \quad , \tag{16}$$

which is an important result for statistical consideration of low fission-yield experiments.

Finally, let us discuss the above results by considering the different cases of the actual sample thickness, x_0, defining in each case the "effective" thickness of the fission source and the total efficiency as well:

i) Very thin targets ($x_0 << x_M$). In this case, equation (15) gives

$$\sigma = \frac{N_t}{QN_a \; x_0 \frac{x_M}{\bar{a}_0}} \quad , \tag{17}$$

where x_0 is the "effective" thickness, and the quantity $x_M/\bar{a}_0$ is identified as the total efficiency, i.e.,

$$\varepsilon_{\text{very thin}} = \frac{x_M}{\bar{a}_0} = 1 - \frac{1+B}{A(1-B)} \quad . \tag{18}$$

Note that if the optics were not taken into account we would have $\varepsilon_{\text{very thin}} = 1-v_G t/\overline{r}_0$ (cf. equation (10)). Since for a full-range etched track $\overline{r}_0 \approx v_T t$, it follows that $\varepsilon_{\text{very thin}} \approx 1-v_G/v_T = = 1-\sin\phi_c$, which is the commonly used expression for efficiency of very thin target samples in contact with track detectors.

ii) Thin targets ($x_0 < x_M$). In this case we may interpret the quantity $x_0/2$ in equation (15) as the "effective" target thickness, and the quantity

$$\varepsilon_{\text{thin}} = (2x_M - x_0)/\overline{a}_0 \tag{19}$$

as the total efficiency.

iii) Thick targets ($x_0 \geq x_M$). Here, equation (14) is applicable, from which we may define the quantity $\overline{a}_0/2$ as the "effective" thickness of the target sample, and

$$\varepsilon_{\text{thick}} = (x_M/\overline{a}_0)^2, \tag{20}$$

as the total efficiency.

The best possible evaluation of the quantities $\overline{a}_0$, $\overline{r}_0$, $v_G t$, and d is essential for applications of the present method to specific fission cases of interest. The two first ones can be evaluated from available range-energy tables, or calculated from appropriate computer codes like the one reported in Durrani and Bull (1987). Kinetic energy of median fission fragments for different fission cases can be evaluated from empirical correlations such as those reported in Viola *et al.* (1985) and Tavares and Terranova (1992). The $v_G t$ is easily measured by current laboratory etching procedures, and the quantity d (minimal etch pit opening capable of being identified under given optics) can be estimated by using calibrated eye-pieces fitted to oculars of routine use in optical microscopy.

4. APPLICATIONS

A quantitative result based on the formulae developed above is presented by using the data taken from the work by de Carvalho *et al.* (1982). An extremely thin ^{252}Cf-source of known absolute fission activity was placed to direct contact with a pre-annealed glass plate for evaluation of the total efficiency. From the data reported in that work one can deduce the values $v_G t = 8.8 \pm 0.5 \mu m$ and $d = 3.2 \pm 0.3 \mu m$. An estimate of the quantity $\bar{r}_0$ from the range-energy tables (Northcliffe and Schilling, 1970) gives $\bar{r}_0 \approx 13 \pm 1 \mu m$ for ^{252}Cf-fission fragments in ordinary glass. Inserting these values into equation (18) one has $\varepsilon = 0.28 \pm 0.07$, which is comparable in magnitude with the result $\varepsilon = 0.263 \pm 0.009$ obtained by de Carvalho *et al.* (1982) from direct experimental evaluation.

An illustrative example of the general results discussed in the present work is shown in Fig.2, where the relative number of etched fission tracks observed per unit area of the detector is plotted as a function of d, for a range of d-values of routine use in ordinary optical microscopy. In this example, a film of uranium oxide is supposed to be in contact with an ordinary glass plate as a fission track detector, for which case $\bar{r}_0 \approx 13 \mu m$, $\bar{a}_0 \approx 7 \mu m$, and the value $v_G t \approx 8 \mu m$ may be considered from the usual etching conditions for fission fragments in glasses. The figure shows the number of fission tracks expected (normalized to $d = 1 \mu m$) in the case of a thick sample (equation (14), $x_0 \gtrsim 3 \mu m$), a thin sample (equation (15), $x_0 = 1\mu m$), and a very thin sample (equation (17), $x_0 \lesssim 0.1\mu m$). A significant decrease of the number of tracks is seen as the optical magnification diminishes (i.e., d increases), thus leading to poor statistics, especially in experiments of low fission yields.

Another example in this line can be appreciated by inspecting Fig.3, where the fission yield of the titanium nucleus (induced, for instance, by intermediate-energy incident photons) is supposed to be investigated by using a metallic titanium target placed in close contact with a makrofol sheet as a fission track detector. From an estimate of the fragment total kinetic energy release in this fission case (Tavares and Terranova, 1992), and by using the current range-energy tables we have $\overline{a}_0 = 4.6\mu m$ (Northcliffe and Schilling, 1970) and $\overline{r}_0 = 8.5\mu m$ (Tripier *et al.*, 1974). The appropriate etching conditions for revelation of nuclear fragments of low rate of ionization in makrofol provides $v_G t \approx 3\mu m$. In this way, equations (14) and (15) give the trends of the relative number of fission tracks expected as a function of d for thick and thin targets. As shown in Fig. 3, a more severe dropping of the number of fission tracks occurs as compared with the fission case depicted in Fig. 2. The illustrative examples discussed above are both typical examples of observation of fission tracks formed under the condition of $v_T > v_G$. They make clear the combined effects of the etching, optics, and target thickness on the final number of fission tracks capable of being observed.

Finally, the accuracy of the formulae deduced in the present work is briefly discussed. Since $x_M/\overline{a}_0$ is the basic quantity directly related to the evaluation of the total detection efficiency, let us calculate, therefore, the uncertainty associated with $x_M/\bar{a}_0$. From equation (18) one has

$$\delta_{x_M/\bar{a}_0} = \left(\frac{\bar{a}_0}{x_M} - 1\right)\left[\delta^2_{\bar{r}_0} + \delta^2_{v_G t} + \left(\frac{4B}{1-B^2}\right)^2\left(\delta^2_d + \delta^2_{v_G t}\right)\right]^{1/2} , \qquad (21)$$

where the δ's are the percent uncertainties associated with the various quantities indicated by the subscripts. These uncertainties vary typically in the range ∿ 5-8% for $\delta_{\bar{r}_0}$, ∿ 1-6% for $\delta_{v_G t}$, and ∿ 1-3% for δ_d. The final value for $\delta_{x_M/\bar{a}_0}$ will depend strongly upon the optics used (i.e., the d-values), as well as the actual, particular experimental conditions. Under a good optical magnification (say, $d \lesssim 2.5\mu m$) uncertainties in the range ∿ 3-10% for $x_M/\bar{a}_0$ can be attained as estimated by equation (21). Although the accuracy of the formulae may be not comparable with that obtained from absolute values based upon direct experimental determinations, the proposed formulae can be used to guide a number of experiments. In many instances they can be useful, and even unique, such as in fission experiments of very low fission yield, for which cases the evaluation of the total detection efficiency by the current experimental methods becomes difficult.

5. CONCLUSION

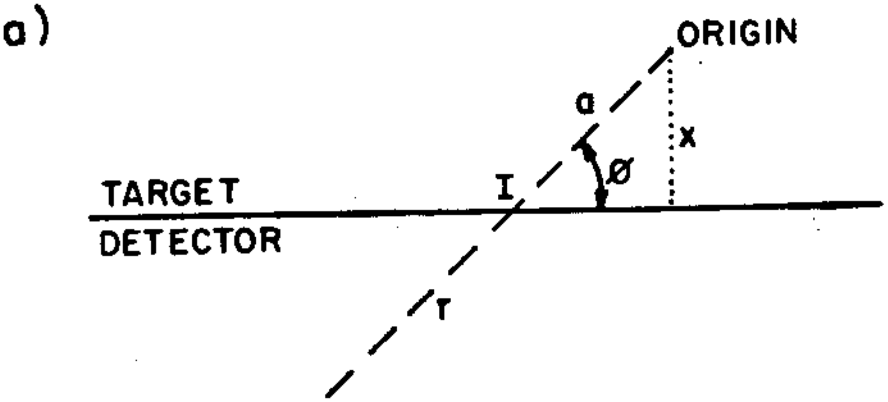
In the course of the present work the arrangement of target sample materials in close contact with dielectric track detectors has been considered to obtain fission yields corrected for the effect of fragment self-absorption in the sample and for the optical magnification for the observation of the etched fission tracks. Target samples have been classified according to thickness, and expressions for total efficiency (etching efficiency times observation efficiency) and "effective"sample as well have been derived for the cases where $v_T > v_G$, i.e., cone-shaped tracks formed by etching. It has been shown how the etching conditions (essentially $v_G t$) and the optics to be used in track analysis are related to the number of etched fission tracks expected, and such a relationship may be useful as a guide in choosing the most adequate (from a statistical point of view) target thickness to give the maximum density of etched fission tracks.

FIGURE CAPTIONS

FIG. 1. Schematic view of a fissionable target material in contact with a solid state nuclear track detector to measure fission yields. In a) the dashed line represents the fragment trajectory from a point-origin inside the target; it crosses the target-detector interface at I with a dip angle ϕ . In b) is shown a cone-shaped track of threshold etch pit opening d corresponding to the limiting dip angle ϕ_1; $v_G t$ is the thickness of detector material removed by etching.

FIG. 2. Relative number of tracks plotted against threshold etch pit opening of fission fragment tracks registered on a common glass plate. The curves represent the trends calculated for uranium oxide samples of different thicknesses as indicated (for details, see text).

FIG. 3. The same as in Fig.2 for metallic titanium targets with makrofol track recorder.


a)
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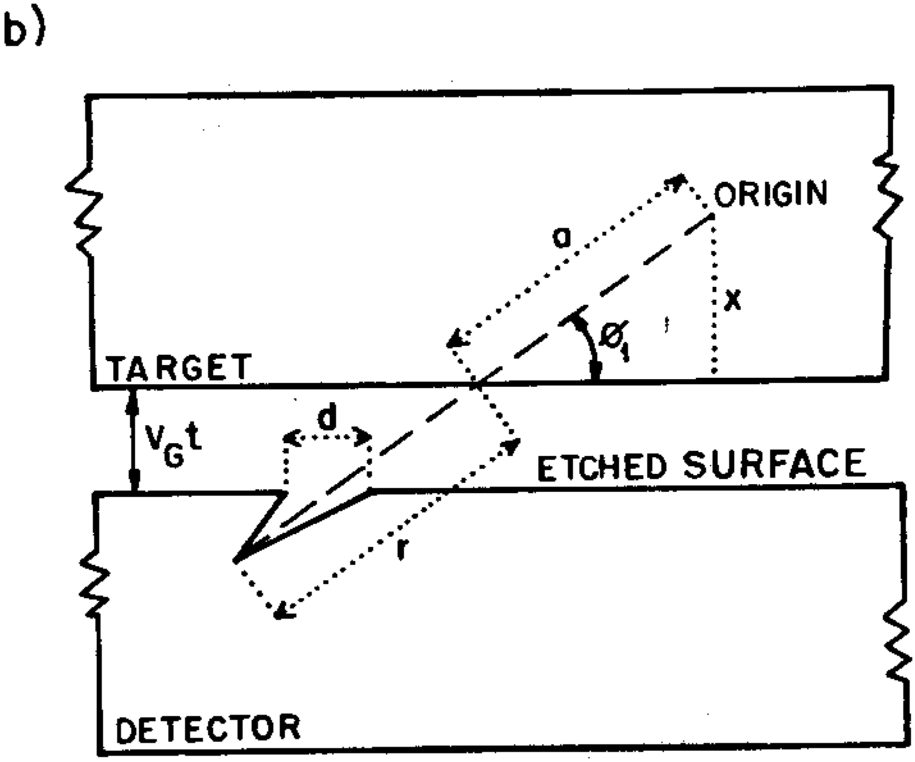
b)
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a
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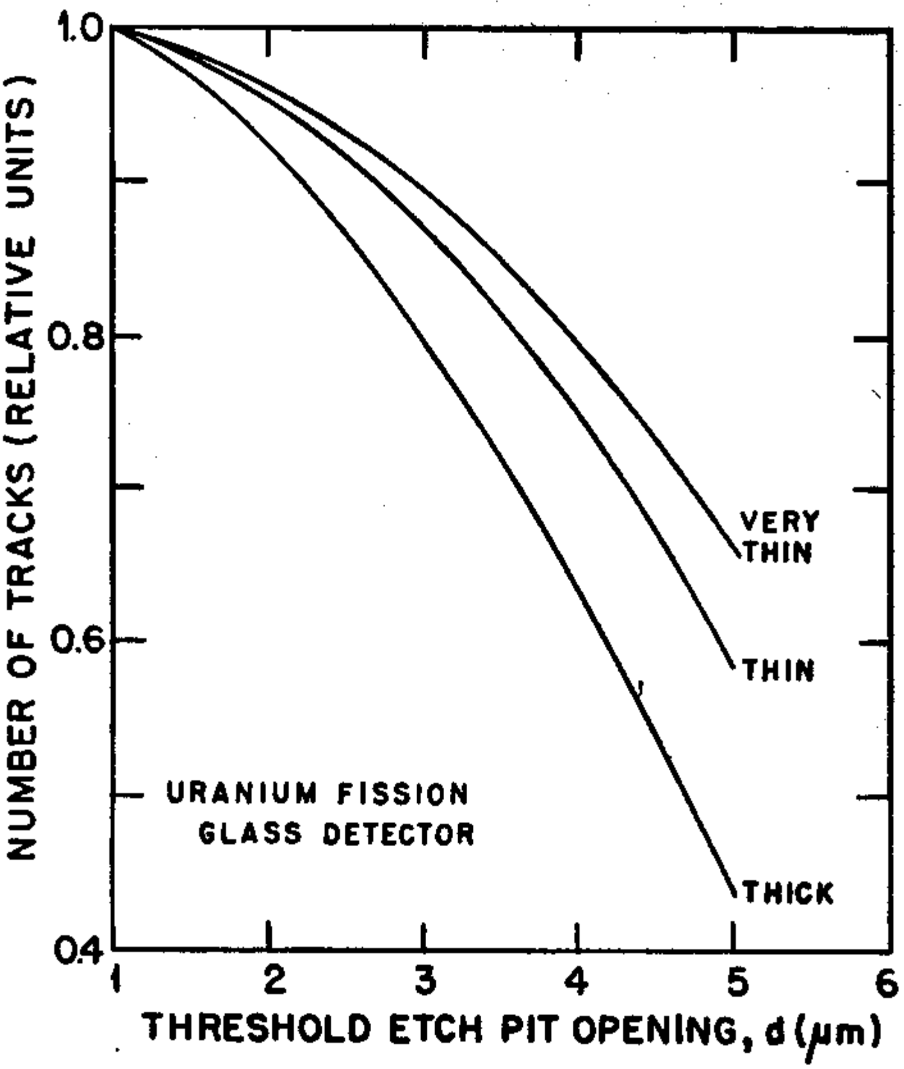
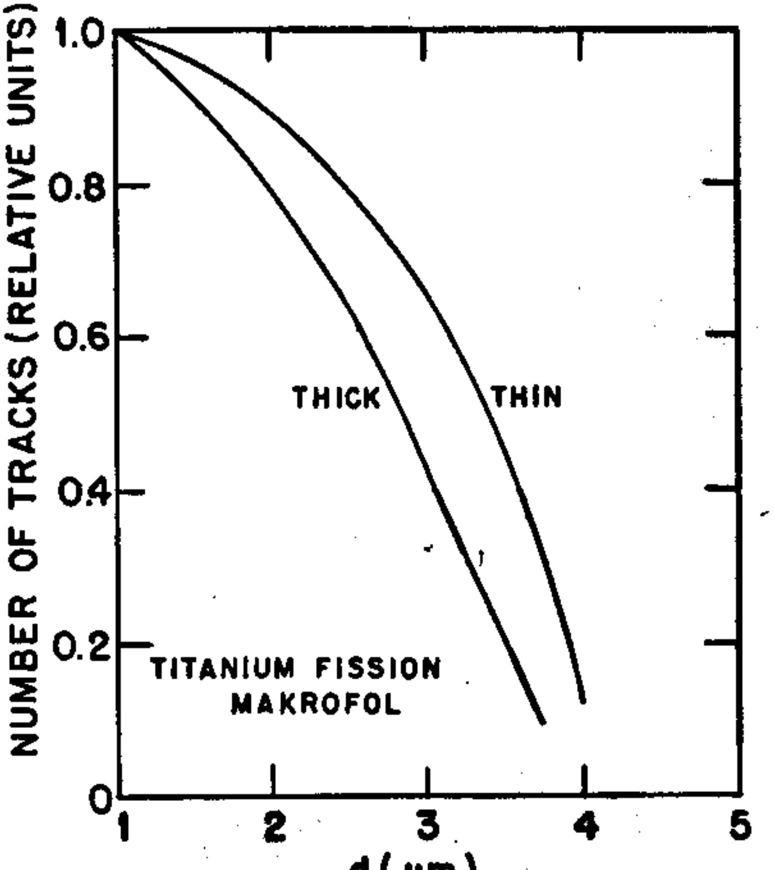
TARGET
$V_G t$
d
ETCHED SURFACE
r
DETECTOR


1.0
0.8
0.6
0.4
1
2
3
4
5
6
NUMBER OF TRACKS (RELATIVE UNITS)
THRESHOLD ETCH PIT OPENING, d (μm)
VERY THIN
THIN
THICK
URANIUM FISSION
GLASS DETECTOR


NUMBER OF TRACKS (RELATIVE UNITS)
1.0
0.8
0.6
0.4
0.2
0
1
2
3
4
5
d (μm)
THICK
THIN
TITANIUM FISSION
MAKROFOL

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