CRITICAL RATE OF ENERGY LOSS FOR REGISTRATION OF CHARGED PARTICLES IN CELLULOSE NITRATE*

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Cellulose nitrate films LR-115 type II (Kodak-Pathé) have been exposed, at right angles, to alpha-particle beams in the energy range 2.5 – 5.5 MeV. From measurements of both through-etched track diameter and through-etched track density, a critical rate of energy loss for track registration of (0.85 ± 0.05) MeV cm^2/mg has been derived, which corresponds to a critical alpha-particle energy of (4.6 ± 0.4) MeV. These results are compatible with those obtained by other authors whenever similar etching conditions are used. The concepts of a threshold rate of energy loss and a threshold energy for etched-track formation are introduced, and their values are obtained from the experiment as being (0.80 ± 0.05) MeV cm^2/mg and (5.1 ± 0.4) MeV, respectively. In addition, the present work provides a suitable set of useful, reference data for further applications of such plastic nuclear track detector in problems concerned with the detection of low-energy alpha particles.

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1. Introduction

It is well known that the average rate of energy loss of a charged particle by electronic excitation and ionization, $-dE/dx$, determines whether a particle track can be chemically etched or not in a solid-state nuclear track detector. Early in 1963, Fleischer *et al.*[1,2]) established in a series of experiments the critical energy for registration of charged particles in muscovite mica, Lexan polycarbonate and cellulose nitrate detectors. In the case of cellulose nitrate then investigated ($C_{12}H_6O_{18}N_4$), the amount of $-dE/dx$ below which tracks were not observed at optical microscope, i.e. the critical rate of energy loss, $(-dE/dx)_c$, was that corresponding to a ~4-MeV alpha-particle. Since then, several investigations have been undertaken[3-7]) aiming to determine the critical energy for registration of charged particle tracks in cellulose nitrate detectors of varied chemical composition. In addition, plastic track detectors have shown to be of high resolution in identifying heavily, energetic ionizing particles[8-10]).

The high sensitivity of polymers to track registration of low-energy alpha particles and protons, particularly the cellulose nitrate, makes feasible numerous applications of such track detectors to several branches of nuclear science[11]). In alpha-particle dosimetry, for instance, it turns out important to know, under definite experimental conditions, the track registration characteristics as well as the critical rate of energy loss in order to account for the quantitative aspects of the response of the detector to low-energy incident alpha particles. During the last years, cellulose nitrate detectors

have been widely tested with very encouraging results as personnel dosimeters in radiological protection programmes[12-17].

In the present paper we describe a general method which allows one to determine the experimental $(-dE/dx)_c$ for registration of charged particles in solid-state track detectors. Although the method can in principle be applied to any plastic track detector, we will concern ourselves to the determination of $(-dE/dx)_c$ for charged particle etched-track formation in the special red cellulose nitrate film LR-115, type II, supplied by Kodak-Pathé[5,18]. This particular track detector consists of a thin reddish layer ($\sim$ 13 μm) of cellulose nitrate covering a $\sim$ 100 μm thick polyester sheet. Such a configuration has the advantage of enhancing track contrast when tracks perforate the residual cellulose nitrate layer after chemical etching, thus allowing for a better determination of $(-dE/dx)_c$. Alpha-particles from sources for routine use in laboratory (^{241}Am for instance) can be used in experiments aimed to determine $(-dE/dx)_c$ for etched-track formation, provided the variation of $-dE/dx$ with alpha-particle energy is well established for the cellulose nitrate compound under investigation. Results will be compared with those obtained for cellulose nitrate detectors from other suppliers and under different etching conditions as well.

2. Fundamentals of the Method

Consider a collimated beam of alpha particles of energy E_o which impinges on the detector surface at right angles.

If E_o is less than or equal to the threshold energy value E_{th} the preferential attack along the latent particle track by the etching solution begins at the original surface of the detector, since in this case the density of radiation damage is high enough to start the preferential solvent action along the track. However, if $E_o > E_{th}$ the track formation will begin only when the etched surface reaches the point of the particle trajectory in which $E_o = E_{th}$. Fig. 1-a shows schematically the effect of increasing etching time on the evolution of the shape of the etched particle track. There is a particular etching time from which the developed particle track perforates the residual cellulose nitrate layer, thus giving high-contrast circular holes on the polyester surface, whose diameters increase with etching time. On the other hand, at a given etching time the through-etched track diameter decreases when the incident particle energy increases. From these considerations the following method was developed in order to determine the threshold rate of energy loss for particle track formation.

Let $v_T = dx/dt$ be the rate of chemical attack in the detector along the track and $S(E) = -dE/dx$ the rate of energy loss of the particle with an energy E along its trajectory. The track length dx formed by etching during the time dt will be given by

$$v_T dt = - \frac{dE}{S(E)} \quad . \tag{1}$$

By multiplying both sides of eq. (1) by v_G/v_T, where v_G denotes the general rate of chemical etching in the undamaged material, we obtain

$$v_G dt = - \frac{v_G}{v_T} \frac{dE}{S(E)} \quad . \qquad (2)$$

Let us indicate by t the etching time the track needs to reach a length equal to the thickness of the residual cellulose nitrate layer (see fig. 1-b). The integration of eq. (2) can be written as

$$\ell = \int_o^t v_G dt = - \int_{E_o}^{E(\ell_o)} \frac{v_G}{v_T} \frac{dE}{S(E)} \quad , \qquad (3)$$

where ℓ represents the detector thickness removed during the time t by the chemical etchant, $E(\ell_o)$ the particle energy corresponding to the rate of energy loss at the polyester surface, and ℓ_o the original detector thickness. Noting that $v_G = v_T$ at points of the particle trajectory in which the particle energy was greater than or equal to the threshold value, i.e. $E \geq E_{th}$, eq. (3) can be written as

$$\ell = - \int_{E_o}^{E_{th}} \frac{dE}{S(E)} - \int_{E_{th}}^{E(\ell_o)} \frac{v_G}{v_T} \frac{dE}{S(E)} \quad . \qquad (4)$$

With gradually increasing incident energy, more prolonged etching times will be needed for the etched track length to remain equal to the thickness of the residual detector layer. But in this case, the point of the particle trajectory where the energy reaches the threshold value approaches the etched surface. In the limiting case, when the detector material gets completely removed and the incident particle energy reaches the value E_o^*, the point of threshold energy would be just on the etched surface, which in turn coincides with the polyester surface. This means $E_{th} = E(\ell_o)$, and from eq. (4) results

$$\ell_o = - \int_{E_o^*}^{E_{th}} \frac{dE}{S(E)} \quad . \tag{5}$$

The result expressed by eq. (5) defines an *ideal* critical rate of energy loss, $(-dE/dx)_{th}$, in the sense that it is obtained as a limiting condition. Experimentally, in order to obtain this ideal S_{th} value, it suffices to determine by extrapolation the incident energy E_o^* to which the through-etched track diameter reduces to zero when the detector material tickness ℓ_o is completely removed by etching. The above procedure leads to values of S_{th} and E_{th} which are independent of etching conditions, as can be readily concluded from inspection of eq. (5).

Actually, through-etched track diameters less than about 3 μm cannot be recognized unambiguously with the magnification of optical microscopes commonly used in laboratory practice. Thus, it turns out important to know, under specific experimental conditions, the critical rate of energy loss, $(-dE/dx)_c$, from which through-etched particle tracks of a minimal measurable diameter can be formed by etching. This situation is represented in fig. 1-b. For an incident particle of energy $E_o > E_{th}$, an increase of the etching time by a small amount Δt gives, from eq. (3),

$$\ell + \Delta\ell = - \int_{E_o}^{E_{th}} \frac{dE}{S(E)} - \int_{E_{th}}^{E(\ell_o)} \frac{v_G}{v_T} \frac{dE}{S(E)} - \int_{E(\ell_o)}^{E(\ell_o+\Delta\ell_o)} \frac{v_G}{v_T} \frac{dE}{S(E)} \quad . \tag{6}$$

The shape of the etched track that would be formed if the cellulose nitrate layer were increased by an amount $\Delta\ell_o$ is a conical

surface, provided that, starting from the polyester surface, the ratio v_G/v_T remain constant when covering the path length $\Delta\ell_o$. For an observed through-etched track diameter D small enough, the following approximate relation holds

$$\frac{v_G}{v_T} = \frac{D}{2\Delta\ell_o} \quad , \tag{7}$$

which combined with eq. (6) gives

$$\ell + \Delta\ell = -\int_{E_o}^{E_{th}} \frac{dE}{S(E)} - \int_{E_{th}}^{E(\ell_o)} \frac{v_G}{v_T} \frac{dE}{S(E)} + \frac{D}{2} \quad , \tag{8}$$

for $E_o > E_{th}$ and D small. Under definite etching conditions, there is a particular value of ℓ, say ℓ_{th}, given by

$$\ell_{th} = -\int_{E_{th}}^{E(\ell_o)} \frac{v_G}{v_T} \frac{dE}{S(E)} \tag{9}$$

for which $D = 0$ when $E_o = E_{th}$. The smallest through-etched track diameter actually observed after removing the thickness ℓ_{th} will correspond to a maximum incident particle energy E_c. This critical incident energy will correspond to a S_c value which can be defined as the *observed* critical rate of energy loss for charged particle track formation. The $(-dE/dx)_c$ value obtained as described above will depend on both etching conditions and the optical system used. Nevertheless, for the actual situation we are concerned with, the observed critical rate of energy loss for alpha-particle track formation in cellulose nitrate does not differ substantially from the ideal S_{th} value defined by eq. (5), as we shall see in Section 4.

Fig. 1-b also suggests a simple method to obtain the variation of the ratio v_T/v_G with particle energy, and thus the condition $v_T(E_{th}) = v_G$ can be used as an alternative way in evaluating the threshold energy value E_{th}. We have:

$$v_T(E) = v_G \lim_{\Delta\ell \to o} \frac{\Delta\ell_o}{\Delta\ell} = v_G \lim_{\Delta\ell \to o} \frac{R\left[E(\ell_o)\right] - R\left[E(\ell_o+\Delta\ell_o)\right]}{\Delta\ell} =$$

$$= v_G \lim_{\Delta\ell \to o} \frac{\Delta R}{\Delta\ell} = v_G \frac{dR}{d\ell} \quad , \qquad (10)$$

where $R(E)$ denotes the residual range of the particle with an energy E (recall that the function $\ell(E_o)$, which can be obtained from the experiment, gives, for each incident energy E_o, the removed detector thickness in order to obtain zero through-etched track diameter). Knowledge of the function $v_T(E)/v_G$ is important since information about the registration efficiency can be gained for alpha particles of oblique incidence on the detector surface.

3. Experimental

Clean sheets of 1cm × 2cm reddish, double-layer, cellulose nitrate ~ 13 μm thick were exposed perpendicularly to collimated beams of alpha particles from an intense ^{241}Am source (main peak energy of 5.49 MeV). Irradiations were conducted inside a chamber containing argon, connected with a manometer able to measure gas pressure up to about 200 Torr with a mean deviation of ± 2 Torr (fig. 2). By changing the gas pressure, alpha-particle fluxes of energy ranging between 2.5 MeV and 5.5 MeV could

be obtained. The incident energy, as well as the alpha-particle beam intensity, were previously calibrated by using a conventional alpha-spectrometry line (a surface-barrier detector connected with a multi-channel pulse-height analyser). Within the energy range considered, a mean energy deviation of about ± 0.1 MeV could be estimated. At each fixed incident energy, a total number of 6×10^4 alpha particles per square centimetre hit the detectors which were positioned at a distance of 12 cm from the source.

The detector samples were etched with a 4.0 N NaOH solution at a constant temperature of 60°C without stirring. Several etching times ranging from 15 min up to 3.5 h were chosen in order to investigate the evolution of through-etched track diameter for each group of samples irradiated at different incident energies.

The thickness of the removed cellulose nitrate layer was determined from the measured mass-difference of the samples prior and after chemical etching. Measurements of optical density in a large number of etched samples showed quite good overall thickness uniformity of the residual detector layer. Under the etching conditions described above, the amount of detector thickness removed with time was obtained as $\ell = 0.46(t-0.34)$, $t \geq 0.34$, with a maximum uncertainty of 5% (ℓ is expressed in mg/cm^2 and t in h). In this way, a general rate of chemical etching $v_G = (0.46 \pm 0.02)$ mg/cm^2h has been derived. An induction time of about 20 min was apparent, which may be interpreted as a very slow dissolution of the chemical species during the early stages of etching. Also, it has been observed that for an etching time of about 4.7 h the detector layer dissolves

$R(E)$ for alpha particles in cellulose nitrate is essential. By making use of the CHN gas analysis we found out for the plastic detector LR-115, type II, a composition very similar to that of cellulose trinitrate ($C_{12}H_{17}N_3O_{16}$), which in turn agrees quite well with the composition of the cellulose nitrate detector studied by Anno and Commanay[4]). Then, taking into account the rate-of-energy-loss-curve and the range-curve as those reported by these authors, we found for the threshold energy the value E_{th} = (5.1 ± 0.4) MeV, which corresponds to a threshold rate of energy loss of $(-dE/dx)_{th}$ = (0.80 ± 0.05) MeV cm^2/mg. On the other hand, the values of the observed critical energy E_c and the corresponding critical rate of energy loss $(-dE/dx)_c$ turn out to be slightly different from the threshold values obtained above, viz. ∿ 4.6 MeV and ∿ 0.85 MeV cm^2/mg, respectively (see Section 2). For the sake of comparison, table 1 reports some results regarding E_c previously obtained by other authors. In despite of the differences in chemical composition of the detector materials as well as the etching conditions employed, it is seen that almost cellulose nitrate detectors listed in the table have similar responses for registration of alpha-particle tracks.

An alternative procedure we have used to determine $(-dE/dx)_{th}$ from the present data was to study the variation of the ratio v_T/v_G with $-dE/dx$. According to eq. (10), this function could be obtained from numerical calculations by making use of the curve plotted in fig. 4 and, again, the curves of $R(E)$ and $S(E) = -dE/dx$ reported by Anno and Commanay[4]). The trend of v_T/v_G with $-dE/dx$ under the specified experimental conditions is shown in fig. 5. The large majority of the calcul-

ated v_T/v_G values lies within the shaded area which indicates an average deviation of ± 10% from the calculated trend (full line). By back-extrapolating down to $v_T/v_G = 1$ the value $(-dE/dx)_{th} = (0.80 \pm 0.02)$ MeV cm^2/mg is obtained which is in a very good agreement with the value obtained by using the previous method. In addition, fig. 5 shows a sharp increase of v_T/v_G from threshold up to about 1 MeV cm^2/mg (the ratio v_T/v_G reaching the value $\sim$ 3 at this point), and a slight increase for larger values of -dE/dx, perhaps towards a constant value in the range under consideration. The strong proportional-like relationship between v_T/v_G and $S - S_{th}$ within the interval 0.8 – 1 MeV cm^2/mg does not remain the same as S increases further. The observed trend suggests a possible mechanism of saturation, i.e. an inhibition of the preferential rate of chemical attack due to a non-complete diffusion of etching products at points of particle trajectory of larger radiation damage densities. These results agree satisfactorily with the observations of Tanti-Wipawin[6]).

As regards charged particles impinging on the detector surface at oblique angles, the critical angle θ_c = arc sin (v_G/v_T) (measured from the surface) for track registration, and therefore the registration efficiency, can be inferred from the v_T/v_G-curve of fig. 5. As an example, let us consider alpha particles of 2.7 MeV. From the -dE/dx-curve by Anno and Commanay[4]) we have a linear energy transfer of 1.15 MeV cm^2/mg which corresponds to $v_T/v_G = 3.1 \pm 0.3$, and finally to a critical angle of incidence for track registration of $\theta_c = 19^o \pm 2^o$. This result compares favourably with $\theta_\ell = 65^o$ reported by Anno and Commany ($\theta_\ell = 90^o - \theta_c$ in their notation), if we take into account the

large errors involved in both θ_ℓ and θ_c measurements. For alpha particles which strike the detector surface at random, the v_T/v_G-curve is of utmost importance on evaluating the registration efficiency, making it possible to use the cellulose nitrate LR-115, type II, as a suitable tool in detecting alpha particles from contaminated atmospheres of radon and its daughters, as has been already tested from other laboratories. Such an application will be the subject of a future work.

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TABLE 1

Critical energies for registration of alpha particles tracks in various cellulose nitrate detectors.

Type of cellulose nitrate detector	Etching conditions	Critical energy E_c (MeV)	Ref.
Anyl acetate plus nitrocellulose ($C_{12}H_6O_{18}N_4$)	6.25 N NaOH 55°C	∿ 4	2
Clear Nixon--Baldwin	6.25 N NaOH 40°C	∿ 3	3
Orange Nixon--Baldwin	6.25 N NaOH 40°C	∿ 5	3
$C_{7.35}H_{9.87}O_{7.31}N_{1.76}$	6.25 N NaOH, 60°C without stirring	∿ 4	4
LR-115, type II, Kodak-Pathé	2.5 N NaOH, 25°C and 60°C, without stirring	∿ 4	5
LR-115, type II, Kodak-Pathé	25% NaOH 55°C	∿ 5*	6
LR-115, type II, Kodak-Pathé	2.5 N NaOH 40°C	1.9-2.5	7
LR-115, type II, Kodak-Pathé	4.0 N NaOH, 60°C without stirring	∿ 4.6	this work

*Threshold energy value deduced taking into account the $-dE/dx$-curve by Anno and Commanay[4]).

Figure Captions

Fig. 1. Schematical representation of etched-track formation in a double-layer cellulose nitrate detector for an alpha particle of right angle of entrance. a) Sequencial stages of track etching showing the track evolution until a measurable track diameter is formed on the polyester surface. b) Track profile and its parameters as used in the text.

Fig. 2. Experimental arrangement for detection of low-energy alpha particles in plastic nuclear track detectors.

Fig. 3. Through-etched track diameter (a) and through-etched track density (b) measured as a function of the incident alpha-particle energy. Different symbols for experimental points refer to different etching times as indicated in a). All curves are eye-fits through the experimental points.

Fig. 4. Dependence of removed detector thickness on incident alpha-particle energy in order to obtain zero through-etched track diameter. For E_o > 4 MeV, points were obtained by extrapolating the curves of fig. 3-a; for E_o < 4 MeV points were obtained by taking the incident energy at half maximum of the through-etched track density plotted in fig. 3-b. The full line is a least-squares fit of the "experimental" points. From the scale on the right side, a bulk etching velocity can

be deduced as $v_G = (0.46 \pm 0.02)$ mg/cm^{2}h, with a period of latency of about 20 min.

Fig. 5. Relative etching velocity v_T/v_G plotted against the rate of energy loss. The full line is the result obtained from the data of the present experiment as described in the text. The shaded area indicates the uncertainty of the calculated curve.

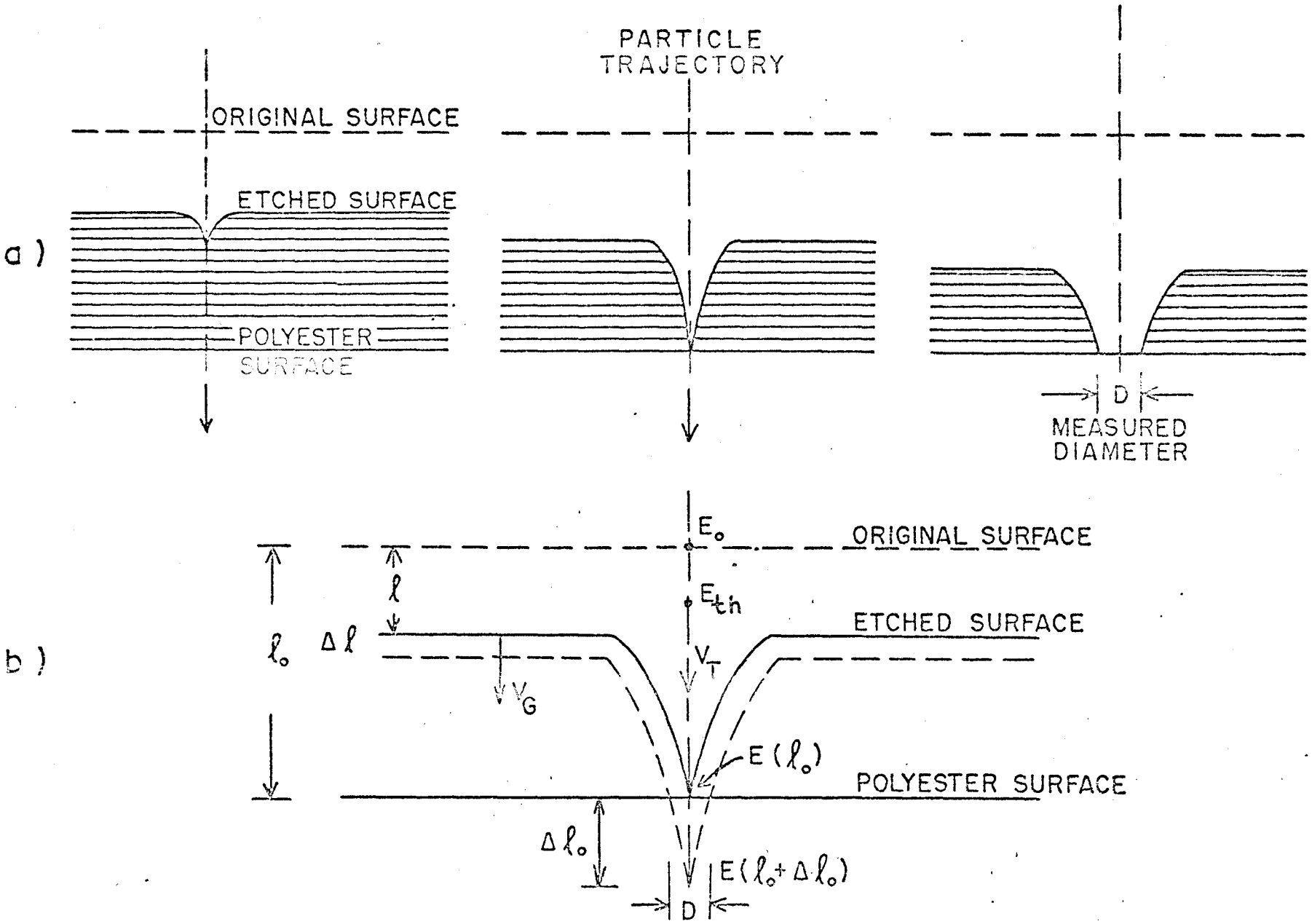
a)
PARTICLE TRAJECTORY
ORIGINAL SURFACE
ETCHED SURFACE
POLYESTER SURFACE
D
MEASURED DIAMETER
b)
E_o
ORIGINAL SURFACE
ℓ
E_{th}
ETCHED SURFACE
ℓ_o
$\Delta\ell$
V_T
V_G
$E(\ell_o)$
POLYESTER SURFACE
$\Delta\ell_o$
$E(\ell_o+\Delta\ell_o)$
D

Figure 1

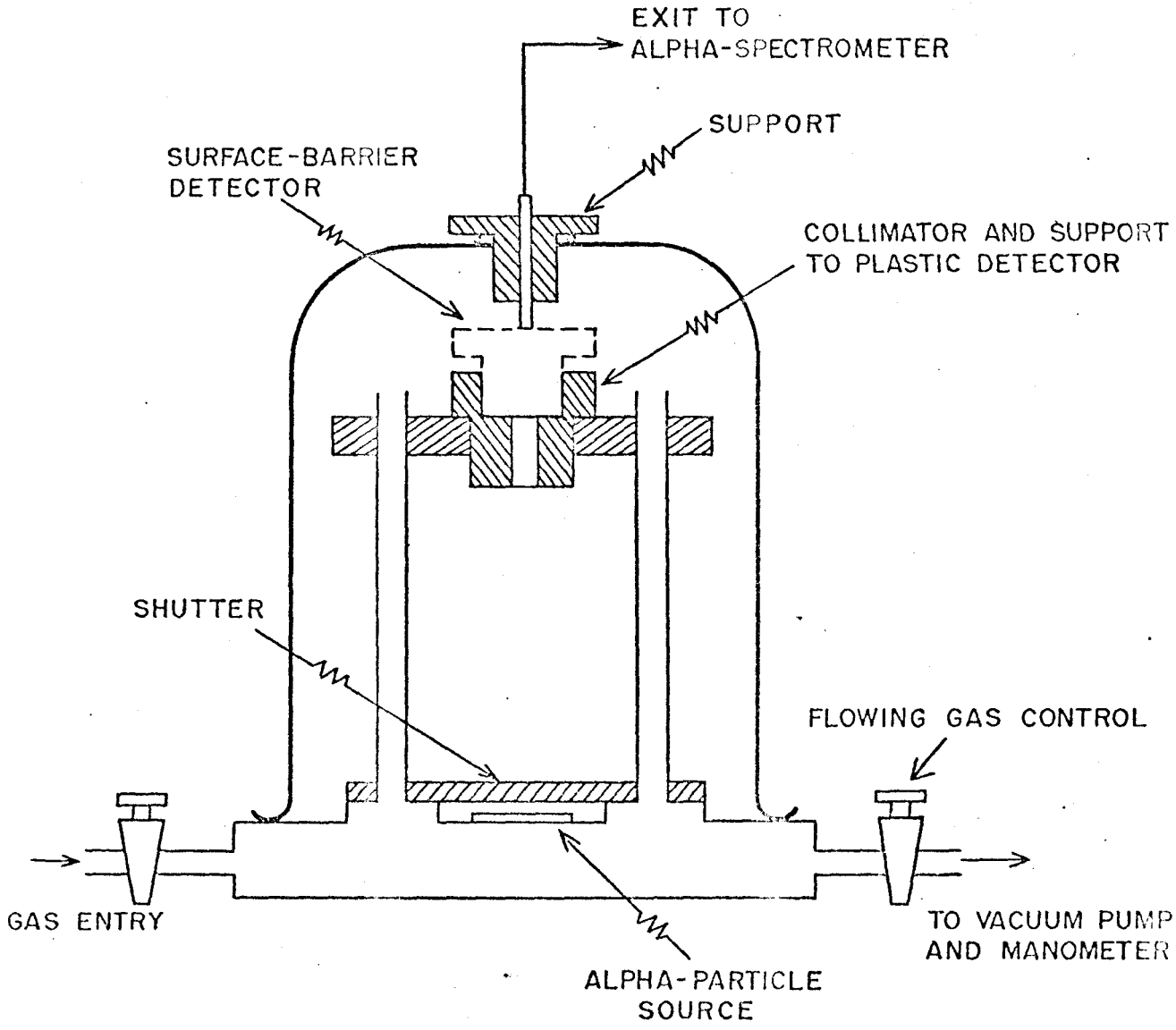
EXIT TO
ALPHA-SPECTROMETER
SUPPORT
SURFACE-BARRIER
DETECTOR
COLLIMATOR AND SUPPORT
TO PLASTIC DETECTOR
SHUTTER
FLOWING GAS CONTROL
GAS ENTRY
TO VACUUM PUMP
AND MANOMETER
ALPHA-PARTICLE
SOURCE

Figure 2

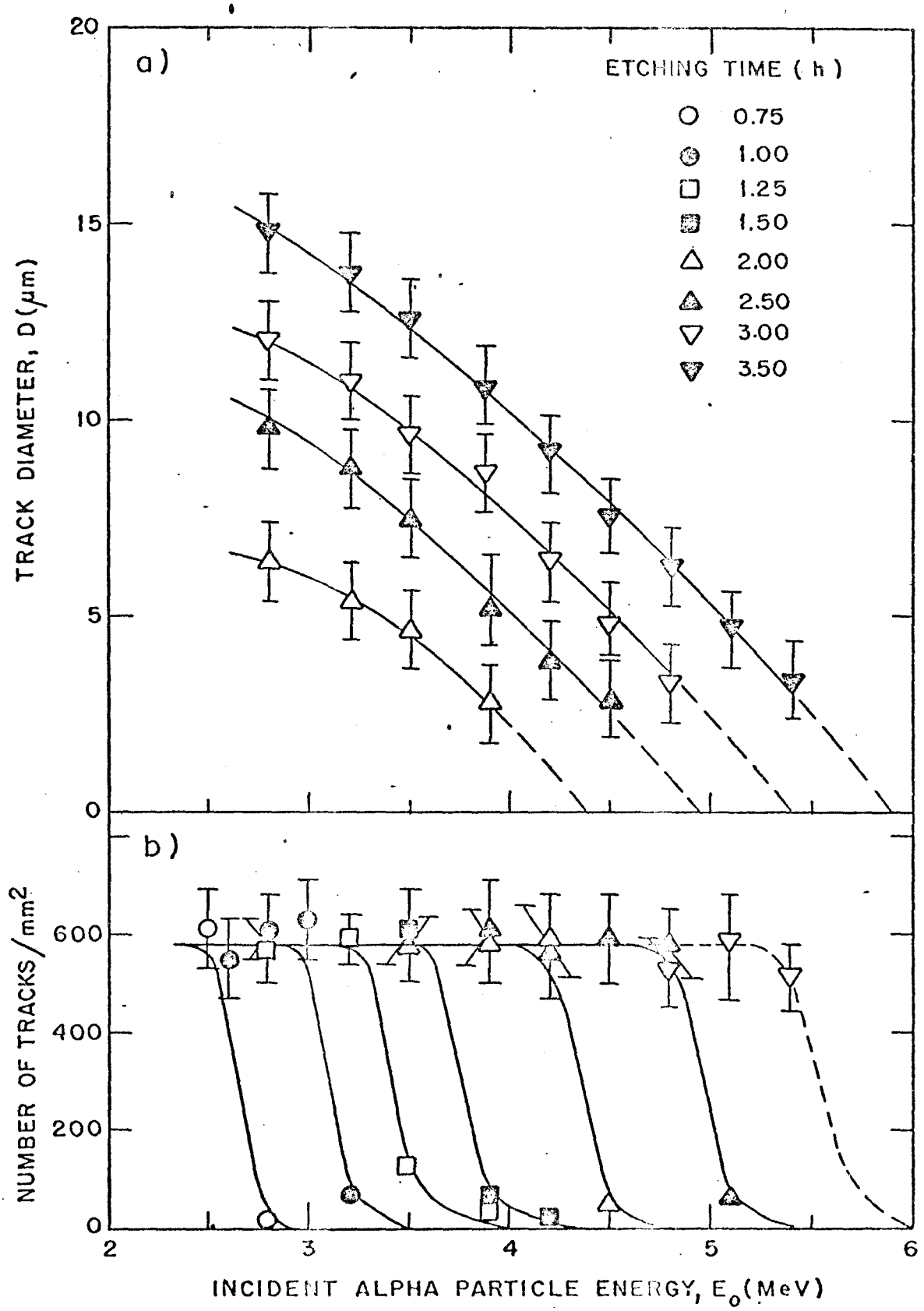
a)
ETCHING TIME (h)
0.75
1.00
1.25
1.50
2.00
2.50
3.00
3.50
TRACK DIAMETER, D(μm)
20
15
10
5
0
b)
NUMBER OF TRACKS/mm²
600
400
200
0
2
3
4
5
6
INCIDENT ALPHA PARTICLE ENERGY, E₀(MeV)

Figure 3

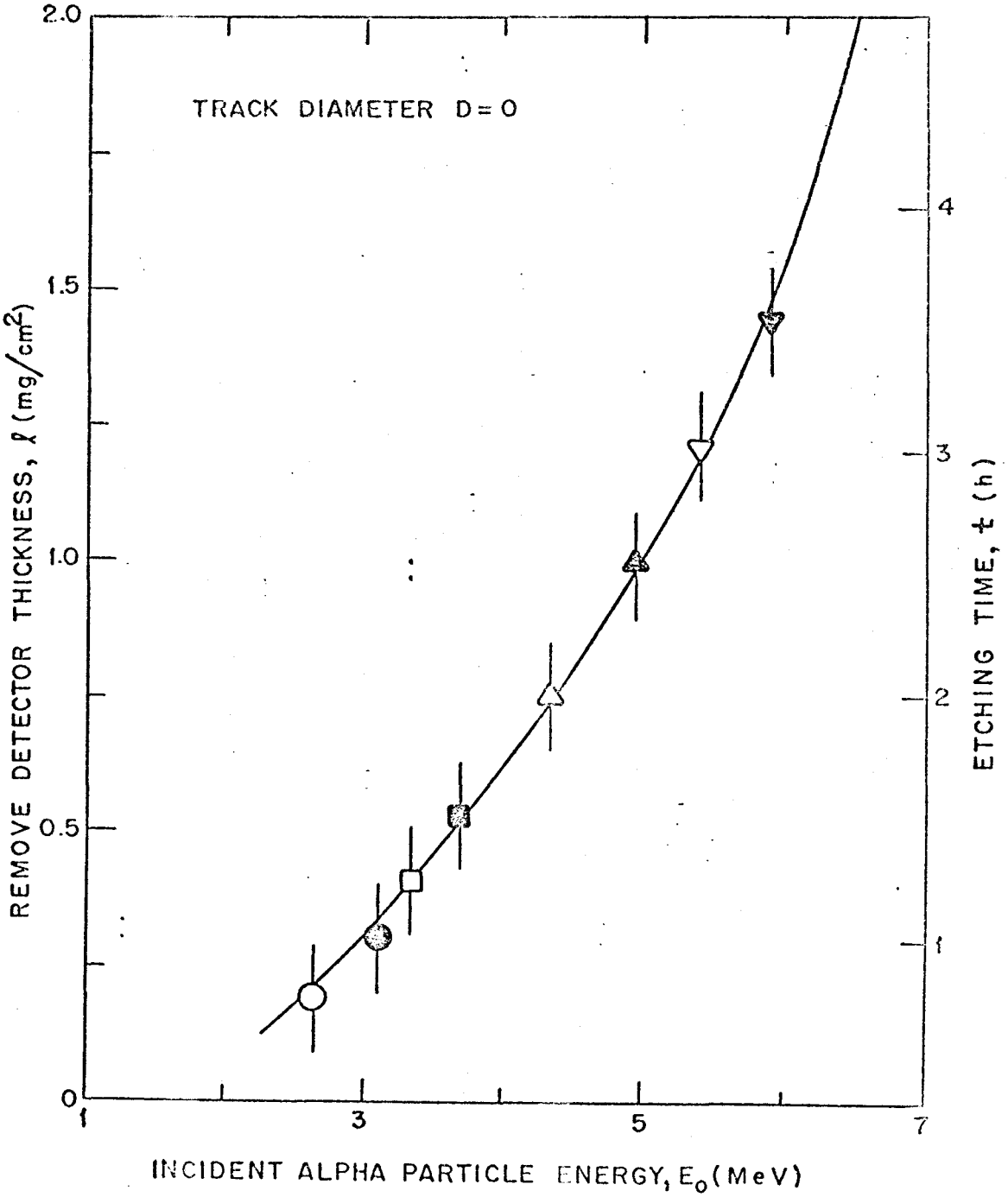
TRACK DIAMETER D=0
REMOVE DETECTOR THICKNESS, ℓ (mg/cm²)
ETCHING TIME, t (h)
INCIDENT ALPHA PARTICLE ENERGY, E₀ (MeV)
2.0
1.5
1.0
0.5
0
1
3
5
7
4
3
2
1

Figure 4

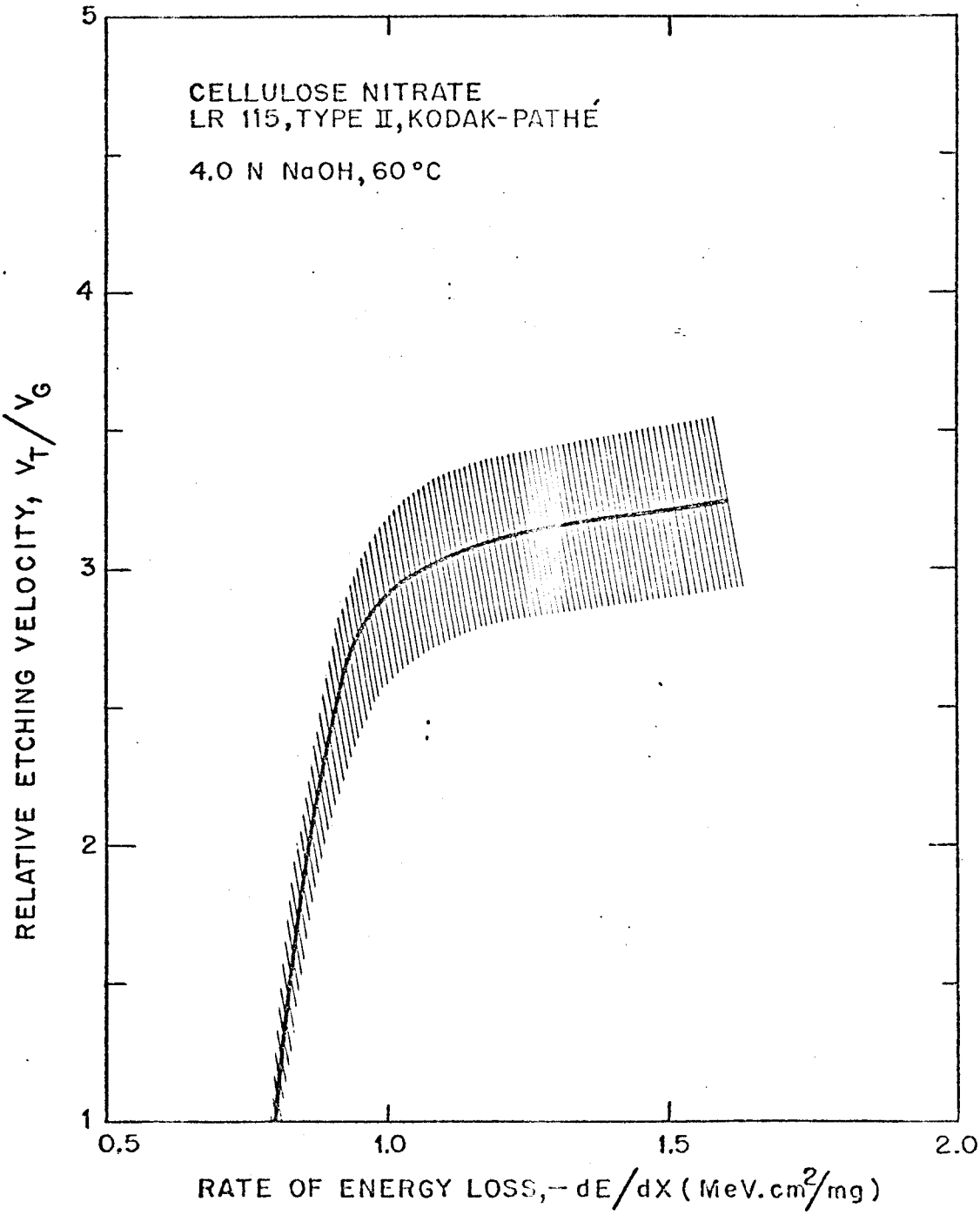
CELLULOSE NITRATE
LR 115, TYPE II, KODAK-PATHÉ
4.0 N NaOH, 60 °C
RELATIVE ETCHING VELOCITY, V_T/V_G
RATE OF ENERGY LOSS, $-dE/dX$ (MeV.cm^2/mg)
1
2
3
4
5
0.5
1.0
1.5
2.0

Figure 5