

# Calibration of CR-39 regarding the Track Diameter Using the Spontaneous Fission Spectrum of $Cf^{252}$

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## Abstract

The aim of this study was to calibrate the Solid State Nuclear Track Detector (SSNTD), CR-39 for the identification of spontaneous fission produced from the  $Cf^{252}$ . The calibration of detectors was carried out with Solid State Surface Barrier Detector and the Radio Chemical Technique. A number of CR-39 detectors were exposed to the fission fragments of  $Cf^{252}$  in vacuum at  $45^\circ$  and  $90^\circ$ . The detectors were etched in 6N NAOH at  $40^\circ$  C to reveal the tracks of spontaneous fission. The etched tracks recorded by CR-39 were scanned using the optical microscope at the magnification of 40x (Leitz-orthoplan). The actual diameters of the tracks were measured using the projected lengths and depths. The graphs of the lengths and diameters were drawn as a function of % yield from the obtained data. Now this data was correlated with the results obtained by Shmitt. He used a solid state surface barrier detector. Our detectors provide us an offline technique. The calibration curve between the track diameters vs the ffs energies was drawn. It was observed that the accuracy in measuring the masses and energies of the ffs is preserved in our case.

## 1. INTRODUCTION OF NUCLEAR FISSION

Shortly after the discovery of neutron, Fermi bombarded uranium with neutrons in an effort to produce transuranic elements (elements with nuclear charge greater than 92). He found that the neutron irradiated Uranium was radioactive. They were not able to pursue the process.

It was left to Hahn and Strassman (1939) to clarify the issue. They proposed that the Uranium nucleus, after capturing a neutron, breaks up into large fragments. This process is called Fission (Rehman, 1977).

## 2. MECHANISM OF NUCLEAR FISSION

### 2.1 Bohr and Wheeler Theory

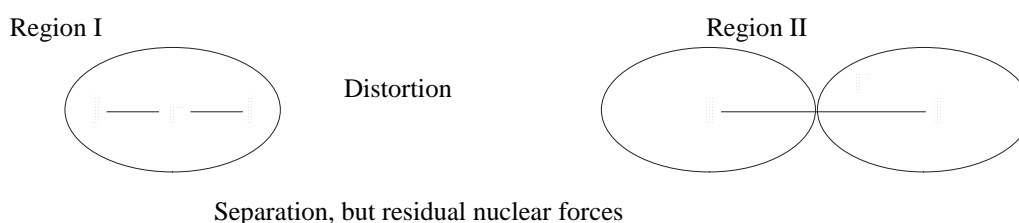
Bohr and Wheeler explained the Fission phenomenon with definite quantitative results. They assumed that the Atomic Nucleus is analogous to a liquid drop. Each Nuclei, like article in the liquid drop, interacts equally with its nearest neighbors therefore the (B.E) is proportional to  $A$  (mass number).

Nucleus, like liquid drop, has close packing of particles, constant density and short range forces. As in the liquid drop, particles on the surface will have smaller number of nearest neighbors than in the interior. It is therefore necessary to decrease the B.E e.g. volume energy reduced by an amount proportional to the surface area of the Nucleus,  $A^{2/3}$ .

### 2.2 Liquid Drop Model

In the case of liquid drop, the dissipative forces acting between the constituent molecules are counteracted by the surface tension forces. Very large drops are impossible since the surface tension forces are feeble and therefore there will be a certain critical size of drop which cannot exist permanently.

In nucleus, short range forces keep the nucleus together, while the electrostatic repulsive forces between the protons tend to disrupt the stability. If the nucleus exceeds to that of a critical size, it will show a tendency to break spontaneously.



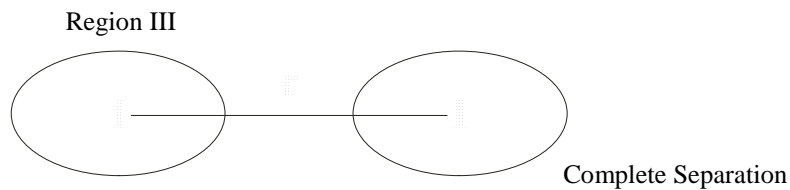


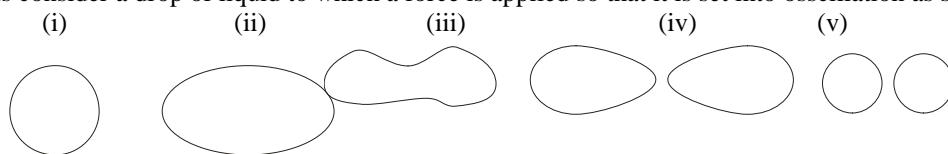
Fig.1 Potential energy of a deformed nucleus vs Separation between the ff's.

As indicated in the Fig1, if the nucleus is deformed, the surface energy will increase, but the coulomb energy will decrease because the repulsive charges move apart (Herald, 1966). ( $C_f$  the terms in the main formula i.e.)

$$E = 4\pi R^2 S + \frac{3}{5} \frac{(c)^2}{4\lambda} \leftarrow R \quad 2.1$$

Hence S is the surface tension, the surface energy per unit area and R is the nuclear radius.

Let us consider a drop of liquid to which a force is applied so that it is set into oscillation as shown stag-wise.



In the steps (ii) and (iii) the volume energy remains constant but the surface area has been increased and if the volume energy exceeds the surface energy, the drop will again return to its original state. Here the surface energy is greater than the volume energy which provides cohesive force. The steps (iv) and (v) occurs only if the deforming forces are sufficiently large. Consequently the liquid drop breaks up and yields two droplets. The Nuclear Fission is similar to the breakup of a liquid drop.

### 2.3 Critical Energy for Fission

According to calculations based on the liquid drop model, the critical energy for fission should decrease as the value of  $Z^{2/A}$  increases (Z and A are atomic number and mass number respectively). So that fission should occur more easily as  $Z^{2/A}$  increases. When  $Z^{2/A}$  is less than about 35, the critical energy is so large that neutrons of very high energy would be required to cause fission but for nuclear having  $Z^{2/A}$  values more than 35, the critical energy is down to 6 MeV, which is of the order of binding energy of a neutron (Samuel & Alexendar, 1967).

It is seen that for the fission nuclides Uranium 235 and Plutonium 239, the neutron binding energy exceeds the critical energy for fission. Hence the capture of a neutron of zero energy would provide sufficient excitation energy to permit the compound nucleus to undergo fission. This explains why these nuclides are fissionable by neutrons of all energies.

To make fission occur we must supply an amount of energy.

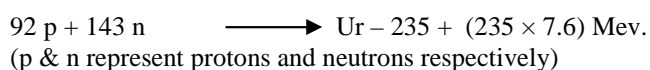
$$E_a = E_b - E_f \quad (\text{see Fig. 2})$$

Where  $E_a$  is critical energy for fission,  $E_f$  is the energy released in fission and  $E_b$  is known as the potential barrier height against fission. For the nuclei of very high mass number ( $A \geq 260$ ), these nuclei would undergo the spontaneous fission within an interval of  $10^{-20}$  sec.

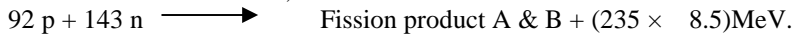
### 2.4 Energy Released in Fission

The amount of energy released in the Nuclear fission can be calculated by considering the net decrease in mass, from the known isotope masses and utilizing the Einstein mass – energy relationship. The fission reaction may be represented (Approx) by:

Uranium – 235  $\rightarrow$  Fission Product A + Fission product B + Energy  
 In Uranium 235, the mean B.E per nucleon is about 7.6 Mev, so it is possible to write



(The mass numbers of the two fission products are mostly in the range of 95 to 140, where the B.E per nucleon is about 8.5 MeV; hence



So upon subtracting the two B.E expressions, the result is 200 MeV (Approx:)

The energy released during fission is the sum of the K.E of fission fragments, K.E of the emitted neutrons, the K.E of the gamma rays etc.

The major proportion – over 80 percent of the energy of fission appears as a kinetic energy of the fission fragments and this immediately manifests itself as heat.

Actually the whole decay energy appears in the form of heat, it may be stated that if pound of fissile material should be capable of producing the same amount of energy as 1400 tons of 13000 Btu/lb of coal. The total energy, (200 Mev) available per fission is about  $3.2 \times 10^{-11}$  watt sec, so that  $3.1 \times 10^{-10}$  fissions are required to release 1 watt-sec of energy. In other words fissions at the rate of  $3.1 \times 10^{-10}$  per sec produce 1 watt of power.

### 2.5 Mass Yield in a Nuclear Fission

A detailed study of the slow neutron fission of Uranium – 235 has shown that the compound nucleus splits up in more than 40 different ways, yielding over 80 primary fission products (or fission fragments). The range of mass numbers of the products is from 72 to 160.

The fission yield is defined as the proportion (or percentage) of the total nuclear fission that form products of a given mass number. The masses of nearly all the fission products fall into two groups, a “light group” with mass no: from 80 to 110, and a “heavy group” with mass no: from 125 to 155 [5]. The most probable type of fission, comprising nearly 6.4 percent of the total, gives products with mass numbers 95 and 139. It is apparent that the

slow-neutron or the thermal neutron  $\frac{1}{40}$  eV fission of Ur – 235 is asymmetrical in great majority of cases (Schmitt et al., 1966). As the energy of the neutron increases, the probability of symmetrical fission also increases.

Corresponding to the distribution of mass numbers, there has been observed a distribution of Kinetic Energy. Two distinct K.E groups have been detected. The K.Es are 67 MeV (Approx:) for the heavy group and 98 MeV for the light group. The ratio of 98 to 67 is about 1.46 and this is very close, as it should be if the momentum is conserved, to the ratio of the mass numbers for the maximum yield i.e. 139 to 95 or 1.46.

During the fission process many of the orbital electrons of the atom undergoing fission are ejected, with the result that the fission fragments are highly charged. The lighter fragments carry an average positive charge of about 20 units, whereas the heavy fragments carry some 22 positive charges. Such particles, moving at the speeds of order of  $10^9$  cm/sec are able to produce considerable ionization in their passage through matter. Because of their large mass and charge, the specific ionization is high and their range is therefore relatively short.

### 2.6 Critical Energy as Function of Distance between Fission Fragments and Mass No: a.

If the mutual potential energy of the two fission fragments is plotted against their separation “r”, a curve is obtained (Figure attached). This curve manifests how the potential energy is changed as a function of separation between the ffs. It is evident from the curve that at the point E, the two ffs are far apart and eventually the P.E is zero.

- i. As the fragments are brought closer, there is an increase in P.E due to the electrostatic repulsion of the positively charged nuclei. When the ffs reach the point “C”, the attractive forces become dominant and the P.E decreases towards A. the figure (2) (attached with), shows and two ffs  $\text{La}^{148}$  and  $\text{Br}^{87}$ . It exhibits the P.E as a function of Distance between the two ffs. in the Fig (1).  
 “ $E_f$ ” is the excess energy of the compound nucleus in its ground state.  
 “ $E_b$ ” is known as the potential barrier height against fission.  
 “ $E_a$ ” is known as the “CRITICAL ENERGY” or “ACTIVATION ENERGY” for fission.

In the figure (2.4) (attached with), the critical energy  $E_a$  is shown as a function of mass number A.  $E_a$  is at first very large, but decreases with increasing mass number. For the nuclei of very high mass number,  $E_f$  and  $E_b$

curves cross, showing that no critical energy is required for mass numbers exceeding about 260. These nuclei would undergo "Spontaneous fission within an interval of  $10^{-20}$  sec.

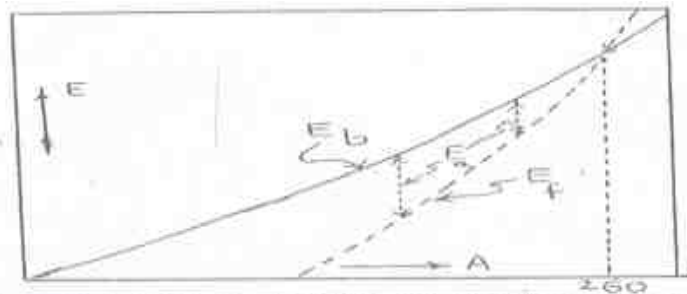


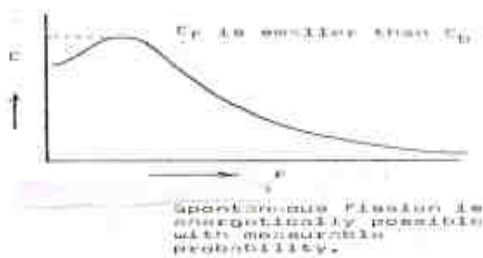
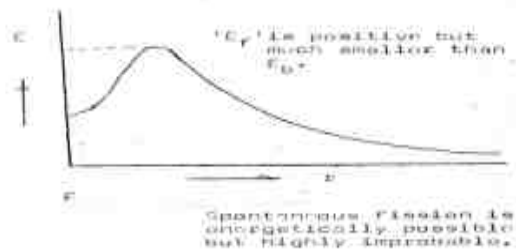
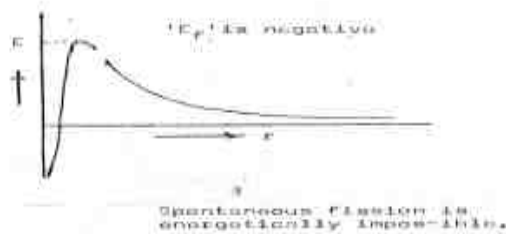
FIG. 2.4 Critical Energy as a function of Mass Number.



FIG. 2.5 Potential energy as a function of distance between fission fragments.

Fig. 3.2

Figures 3.2a through 3.2d show nuclear potential energy as a function of the separation between two fission fragments during a fission process of nuclei of atomic mass numbers 75, 150, 225, 300.



### 3. EXPERIMENTAL METHODOLOGY

#### 3.1 Preparation of Samples

##### 3.1.1. Exposure (Irradiation)

The Solid State nuclear Track Detectors (SSNTDs) are particularly well suited in the field of nuclear physics, especially in the study of nuclear fission reactions. As they are capable of registering low fluxes of fission fragments so, remaining unaffected by very large fluxes of lightly ionizing particles. In their ability to withstand high temperatures and being able to discriminate against high backgrounds they produce less ionizing radiation

(gammas and alphas). They have chief advantages due to their small and flexible geometry (Ashenbach, 1973). In our project we have used the plastic detectors, CR-39 (Columbia Rasin, comp: Homopolymer of alloy diglycol carbonate,  $C_{12}H_{18}O_7$ ) of thickness 600 micron. A number of these detectors were irradiated in vacuum, both perpendicularly and angularly (at  $45^\circ$ ) by the fission fragments of  $Cf^{252}$  (a source of spontaneous fission). In this way samples were irradiated. These detectors were developed by Intercost Europe Co. of Pharma, Italy.

The kinematical analysis of events is based upon the measurement of geometrical parameters. The depth of a track is measured by focusing the microscope on the start and at the end point of track with the help of linear displacement transducer attached with the stage of the microscope. Whereas the projected length of the track on the surface of the detector is measured with the help of a tracing optical tube attached to the microscope. Afterwards, simple geometrical calculations give the actual lengths of the tracks.

### 3.1.2. Etching

The irradiated CR-39 detectors, after the exposure, were etched in 6N NaOH at the temperature of  $40^\circ$  C for one hour to reveal the tracks. It is worth mentioning here that the successful etching of the tracks depends upon the fact that the velocity  $V_t$  of the etchant along the damage trail is higher than the general (bulk) velocity  $V_o$  of the etchant in the material. Once the etchant has reached the end point of the track, the velocity of the etchant along that direction reverts to its general value  $V_o$ . It is expected that the more inclined is a track w.r.t the etched surface, the track would be lost and consequently at a smaller depth would be resulting etch pit become too shallow to be recognized. A steeper (more vertical) track, however, should continue to be recognized for much longer period of etching.

The process of etching introduces the important phenomena. This consists in the fact that there now exist a critical angle of etching below which the tracks fail to be registered. The reason that track becomes visible at all is that the damage trails are etched preferably by the etchants, in other words the velocity  $v_t$  off the etchants along the track is greater than the general bulk velocity  $V_g$  along any other direction in the medium.

An important consequence of the existence of the critical angle, and of the fact that smaller it is, the greater the efficiency of registration of a given particle, is that it over-rides the ordinary solid angle considerations in detection efficiency.

The ffs tracks are made visible through etching and for long etching times, a linear dependence between the surface area of pits and the particle velocity was observed. The diameter of the etched tracks thus are related to the energy of the ffs (Khan & Durani, 1972).

### 3.2. Scanning and Measurements

The etched samples were scanned with an optical microscope (Leitz-orthoplan) at the magnification of about 40X. Hundreds of events were scanned from three samples. Two samples were irradiated angularly ( $45^\circ$ ) and one sample was irradiated perpendicularly. We measured the track diameter of the tracks.

### 3.3 Track Diameters

The sample used in measuring the track diameters was exposed at  $90^\circ$ . Firstly the events were scanned with a compatible grid (Patterns of numbered squares). The data of projected diameter was traced by using a tracing tube. The actual diameters were obtained by multiplying the projected diameter with the calibration factor.

Actual Diameter = Measured D x CF. The measured values of the track diameters are given below in the Table.

## 4. EXPERIMENTAL FINDINGS

### 4.1 THE PLOT OF TRACK DIAMETERS

Firstly the track diameters were plotted against the frequency of occurrence, Fig.4.1. The figure depicts the relationship between the diameters of the etched tracks to the energy of the ffs. This yielded two peaks. The data containing information about the energy (MeV) of ffs, obtained by Schmitt et al, was correlated with our experimental data, fig 4.2. Some points on the said graph were selected with the error bars. The Gaussian has been fitted to the data to obtain the mean value and the standard deviation. The result clearly demonstrates that the energy of the ffs increases as the track diameter increases. The energies of the ffs can be determined if we have a data of track diameters, fig 4.3.

Figure 4.1. Frequency distribution of the track diameters ( $\mu\text{m}$ ) of the ffs

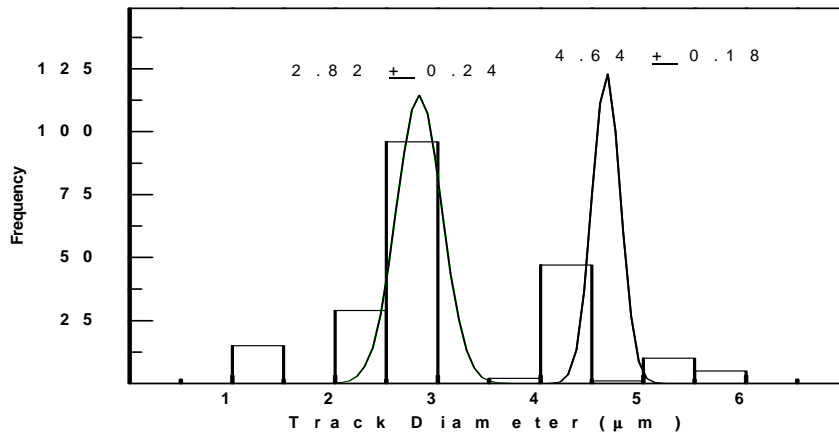


Figure 4.2 Distribution of the sizes of the etch pit diameter of the ffs of the spontaneous fission of Cf-252. The dashed lines show the pulse height distribution as obtained from Schmitt et al [1966].

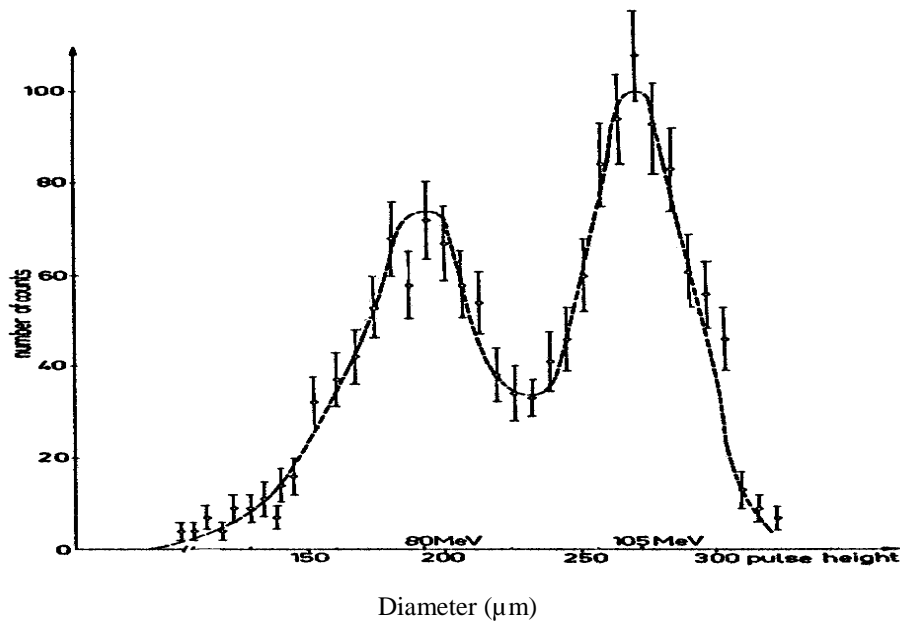


Figure 4.3 Experimental data correlated with the data obtained by Schmitt et al [1966].

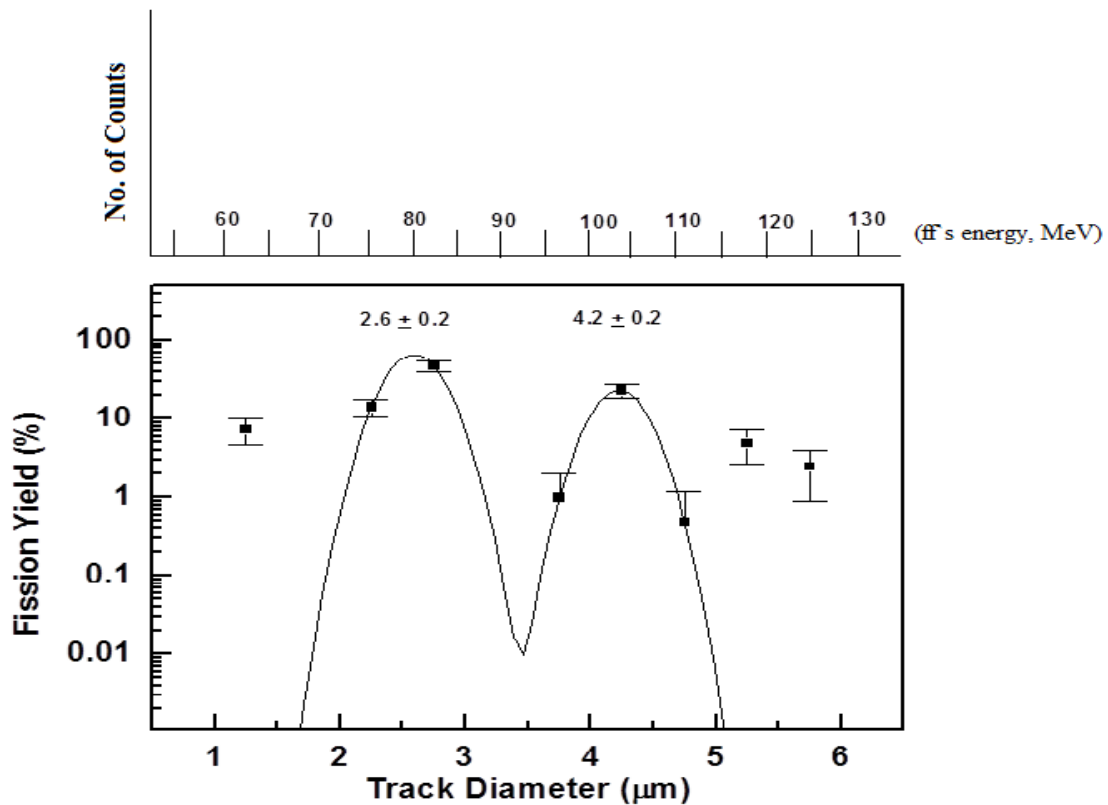
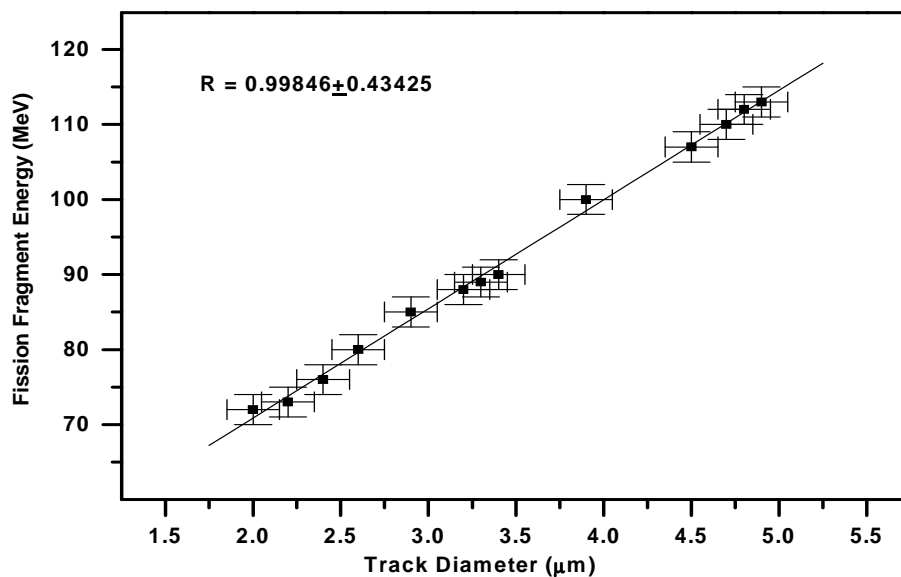


Figure 4.4 Calibration graph between the track diameters ( $\mu\text{m}$ ) and the ffs energies (MeV)



## 5. RESULTS AND DISCUSSIONS

The mass distribution is probably the most important characteristic of the fission process. Large number of experimental data on the mass distribution in the fission process for various nuclei under variety of conditions has been carried out by various scientists, using Solid State Nuclear Track Detectors (SSNTDs). The knowledge about the fission phenomena was repeatedly enhanced using the SSNTDs. The fission fragments (ffs) and energetic heavy ions create tracks in the insulators, which can be made visible with an optical microscope of high resolving power. This can be made only possible when proper etching is carried out.

In our project the etching process was done by 6N NaOH for one hour at 40 °C to reveal the tracks. Further information's about the related parameters of the tracks viz track lengths, track diameters, track depth and corresponding energies and masses are obtained. It has been shown earlier that the diameters of the etched tracks ffs on the glass detectors are related to the energy of fission fragments (Ashenbach, 1973). The quality of the energy resolution thus obtained depends upon the nature of the glass and etching time. In this work we have represented some of the experimental representation through the graphical data using the plastic detector CR-39. This data was correlated with the data obtained by Flynn et al and Schmitt et al, those who used radiochemical technique and the solid state surface barrier detector. The results were utilized to calibrate our detector with these detectors.

## 6. CONCLUSIONS

The main objective of this study is to calibrate the CR-39 detector by computing the masses and energies of the ffs yielded during the spontaneous fission. The detector was standardized by correlating our results with; Schmitt et al results who found the relationship between the pulse heights (Energy of ffs) and the number of counts. He used the solid state surface barrier detector as said before.

To fulfill this purpose, fission fragments of  $\text{Cf}^{252}$  registered on CR-39, which is solid state detector, was used. A number of CR-39 detectors were exposed to the fission fragments of  $\text{Cf}^{252}$  in vacuum at 45° and 90°. The detectors were etched in 6N NAOH at 40° C to reveal the tracks of spontaneous fission. The etched tracks were scanned using optical microscope 40x (Leitz-orthoplan). It is worth mentioning here that the actual lengths and the diameters of the tracks were measured using the projected lengths and depths. A graph of the track lengths was drawn as a function of percentage fission yield from the obtained data. Consequently the mass of the fission fragments was obtained. A graph of the track diameters was drawn as a function of % age fission yield from the obtained data. Consequently the energy of the fission fragments was obtained. Finally a calibration curve was drawn to verify the accuracy of our detector. After correlation, the reliability of the plastic detector was calibrated with the detector used by Schmitt as mentioned above. On the basis of this study it is concluded that the plastic detectors can also be used successfully for the study of fission phenomena to calculate the fission fragment masses and energies. It is also concluded that the masses ffs is inversely proportional to the track length and the energy of ffs is directly proportional to the track diameter.

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