Advances in Physics Theories and Applications
ISSN 2224-719X (Paper) ISSN 2225-0638 (Online)
Vol.37, 2014
www.iiste.org

Calibration of CR-39 regarding the Track Length using the Spontaneous Fission Spectrum of Cf^{252}

Bakht Yar Akhtar
Department of Physics, Gomal University, Dera Ismail Khan, Khyber Pakhtukhwa, Pakistan

Abstract
The objective of our studies 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^o and 90^o. The detectors were etched in 6N NAOH at 40^o 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 lengths of the tracks were measured using the projected lengths and depths. The graphs of the lengths were drawn as a function of % yield from the obtained data. Now this data was correlated with the results obtained by Flynn. He used the radiochemical technique (offline technique). Our detectors provide us an offline technique. The calibration curve between the track lengths vs the ffs masses was drawn. It was observed that the accuracy in measuring the masses and energies of the ffs is preserved in our case.
Keywords: Calibration, CR-39, Track Length, Spontaneous Fission Spectrum

1. INTRODUCTION

Solid State Nuclear Track Detectors are the members of the nuclear particle detector family. They have been existing on earth and the cold planets in the form of crystalline minerals and glassy matter for the billions of years. However, they were discovered recently (Durrani & Bull, 1987). The process of track formation is older than the creation of man. However the story of the detection of charged particle tracks began in Young (1958), when D.A Young working at the atomic energy held in contact with a uranium foil irradiated with thermal neutrons, revealed a number of etch-pits after treatment with a chemical regent. After words in 1959, Skill and Barnes (1959) working in the same laboratory observed hair like tracks using electron microscope in thin sheets of mica. In 1961, Price and Walker of the general electric company research and development Schenectady carried on from the stage where Silk and Barnes had left. After words in 1963, Fleischer joined the team of Price and Walker and for several years three scientists carried out almost all the work on SSNTD's.

When an ionizing charged particle passes through a dielectric material, the transfer of energy to electrons results in a trail of damaged molecules along the particle track. In some materials, the track can be made visible upon etching in a strong acid or base solution. When these exposed detectors are immersed in the etchant (the acid or the base solution) the entire surface of the material is attacked but those points at which particle tracks have entered are etched at a faster rate. The tracks can thus be made to form pits on the surface which are large enough to be easily visible through a conventional micro scope. Materials used to detect particles in this process are called track–etch detectors, Knol, 1989 and their properties and applications have been thoroughly reviewed in a comprehensive book by (Fleischer, 1975; Durrani & Bull, 1987). In common with other passive detectors such as photographic emulsions or neutron activation foils, the track-etch detectors have the advantages of simplicity and low cost. They also possess a very useful inherent threshold in that there is a minimum value of the specific energy loss (- dE/dx) required by the particle before the damage is severe enough to lead to an etchable track.

The threshold is always well above the specific energy loss of an electron track, so that track-etch materials are inherently insensitive to fast electron or gamma-ray interactions. Most materials also do not respond to lightly ionizing charged particles, such as protons or deuterons and will therefore also be insensitive to the recoil protons produced by fast neutron interactions. However, some dielectrics will register proton tracks (CR-39, a plastic detector) Matiullah, 1987 over a certain range of energy. Track etch detectors share with nuclear emulsions the disadvantage of requiring individual track counting. When done manually, the counting step is tedious, laborious and is very time consuming if a large number of track detectors are to be counted. Automatic counting systems have been developed but involve a sizable investment in cost and complication. Although some differentiation can be made of the particle type and energy based on appearance of the etched pit, many of the original tracks are lost in the etching process.

The damage created by the incident particle can be through collisions of the particle itself or form the energetic delta rays created along its track. The range of the delta rays may extend approximately 5mm in any direction

away from the particle position, so that the radius of the primary damage track is thought to be about this dimension (Fliescher et al., 1975). After etching, the tracks are greatly enlarged to a diameter up to 10-20 mm. Etchable tracks are formed in a variety of materials. All are electrical insulators, although some wide-band-gap semiconductors are also known to record tracks. These materials fall into two main categories: inorganic solids, such as crystals and glasses, and organic solids, such as polymers. In the first category, the most popular materials are mica and flint glass, whereas polycarbonate and polyester films are the most common organic track-etch detectors.

A completely satisfactory mode for the mechanisms that lead to the registration of tracks does not yet exist. Past attempts have been made to link the etchability of tracks to the specific energy loss (-dE / dx), primary ionization, restricted energy loss, the density of atomic displacements of the creation of a "thermal spike" of molten material along the particles track. Another hypothesis, known as the particle track by chart imbalance, caused by the ejection of many electrons from the immediate region of the track. Some of these models are briefly discussed below. None of these approaches can fully account for some of the observed differences in etching behavior. The threshold of track etching is best correlated with the restricted energy loss, which is tied to the energy deposition rate only along the primary particle path, excluding that of long-range delta rays (Ramli, 1983). Because the energy imparted to the delta rays is one component of the more traditional specific energy loss dE/dx, the restricted value is somewhat smaller, particularly at high particle energies.

2. TRACK FORMATION MECHANISM

As mentioned above, attempts have been made in past, to correlate the etching behavior of different track materials to the physical phenomena occurring in the detector materials due to the passage of ionizing radiation through different detector materials. Based on these efforts following are the proposed criteria which must fit to the observed behavior of plastics and crystals.

2.1 Total Rate of Energy Loss de/dx

The earliest and perhaps the most natural explanation of track formation was that it depends on the total amount of energy deposited per unit path length by the incident ion. This criterion was proposed by Fleischer, Price, Walker & Hubbard (1967). The proposition was that tracks are formed when *dE/dx* exceeds some critical value $(dE/dx)_C$. This approach became untenable when results of more extensive irradiations, particularly in the high-energy region, became available and was rejected by Fleischer (1967) in favor of the primary-ionization criterion described below.

2.2 Primary Ionization J

In this model, the formation of etchable tracks is related to the number of primary ionizations produced close to the ion path. A relationship for the primary ionization, J based on the work of Bethe is usually employed:

$$J = C' \frac{Zeff^2}{\beta^2} \left[in\left(\frac{\beta^2}{1-\beta^2}\right) - \beta^2 - \delta + K \right] \qquad 1.1$$

Where *Zeff* and β are, respectively, the effective charge and the velocity of the ion relative to the velocity of light c. The terms *K* and C' are constants for a given stopping medium and δ is a relativistic correction term which is related to the polarization of the stopping medium. It has not yet proved possible to calculate *K* from first principles or to make an absolute determination of J for most stopping media. *K* is frequently used as a fitting parameter and *J* is calculated in arbitrary units. This procedure does not affect correlations, since the fitting of the data depends on the shape of the *J* versus β curves, rather than on the absolute values of "*J*".

It was found that primary ionization did indeed give a good fit to the result of heavy-ion irradiation experiments and that a consistent value of primary-ionization threshold, *J* could be obtained for a given stopping medium. The reason of the success of the primary-ionization criterion in comparison to *dE/dx* is that the higher-energy δ-rays carry off an appreciable energy outside the central track region ($\approx 50^o$-100^o A), and this energy is unlikely to play a part in track formation. In the formula for the total rate of energy loss, the energy of these δ-rays makes a significant contribution to *dE/dx*; where as in the primary ionization formula such high-energy events are given the same weighting factors as low-energy δ-rays and thus assume comparatively less importance.

The primary-ionization criterion is open to the criticism that it takes no account of the ionization produced by even the low energy δ-rays and that the ionization potentials implied by the values of *K* used in obtaining the best fit to experimental data are physically unrealistic. With regard to the second of these criticisms it must be said that Fleischer (1967) regard Eq. (1.1) as phenomenological useful rather than of deep theoretical significance. Moreover, since in crystals the radial extent of unetched tracks is only a few tens of angstroms, it seems reasonable that primary processes should predominate over secondary ones as the basis for track formation although this may not be true for polymers, or generally in the cases of low-energy ions. (< 1 Mev/nucleon).

The primary ionization criterion has proved highly successful in practice, and has survived to the present day except that the concept of a threshold *J* is regarded nowadays as being less decisive. Thus, for both plastics and minerals it has been found that the track etch velocity V_T is a continuous function of *J*. typically, V_T versus *J* curves generally rise very steeply and particularly for minerals, the concept of a threshold remains a reasonable approximation to reality (Fleischer, 1967).

2.2.1 Restricted Energy Loss

This criterion, proposed by Benton (1967), makes allowance for the energy removed from the track core by high-energy δ-rays. This is done by imposing a cutoff ceiling W_o on the energy of delta rays, such that only that part of the energy loss is assumed to be relevant to track formation where the energy carried by the δ-rays electrons is less than W_o Restricted energy loss can be calculated from the following expression:

$$\left(\frac{dE}{dx}\right)W < W_O = \frac{n_O e^4}{8\pi\varepsilon^3{}_0 m_0 c^2}\cdot\frac{z^2\ eff}{\beta^2}\left(In\ \frac{W\max W_O}{I^2} - \beta^2 - \delta - U\right) \quad 1.2$$

Where *U* is the term which takes account of the non-participation of inner electron shells and all the other symbols are as defined earlier. For the purposes of this equation to experimental data, W_O is variously taken as 200, 350, or 1000 eV. This quantity, *(dE/dx)* $W_< W_O$, varies with ion energy *E* in a manner rather similar to primary ionization. It gives a good fit to mist experimental data Siegmon (1978). Have found that it fits track etch rate data for **Fe** isotopes in plastics equally as well as does primary ionization. It may be noted, however, that Ahlen has recently shown Ahlen (1980) that some cosmic ray track data are less well fitted by REL than by primary ionization.

The choice of W_o fitting parameter introduces a certain arbitrariness into REL calculations, a feature the REL shares with the primary-ionization criterion. Also, even electrons of energy $\approx$350 eV will deposit some of their energy far from the track core; so the physical basis of W_o is not clearly understood. The aforementioned models appear to describe track formation in polymers reasonably well. However, in inorganic crystals it seems that appeal must be made to some more specific mechanism for track formation, which is triggered only by the passage of very heavily ionizing particles through a stopping medium. Some mechanisms of this kind are considered below.

2.3 Thermal Spike Model

Bonfoglioli have concluded [13] that track in solids are formed by a thermal spike process. According to this model the passage of energetic particles is assumed to produce intense heating of the localized region of the lattice. This region is therefore raised to very high temperature and if the state of this high temperature remained for appreciable time, the processes of melting, re-crystallization surface evaporation or point defect creation and migration would be highly probable. This model imposes two conditions for the production of etchable tracks.

1- The state of high temperature should exist for appreciable times.
2- Normal heat conductivity of the medium should be low.

From these conditions it is obvious that there is a greater probability of track formation in insulators and semiconductors. That is in agreement with general observation.

2.4 The Ion Explosion Spike Model

This model was put forward by Fleischer (1959) after the failure of thermal spike model. According to this model a charged particle while passing through a medium produces a narrow positively charged cylindrical region around its trajectory. The positive ions exert coulomb repulsion upon each other as a result some of them are ejected into the interstitial positions leaving behind a vacancy rich cylindrical core. Thus a track is formed. This model requires the fulfillment of following conditions for the track. Electrical stress must be greater than

mechanical strength of the medium. There exist a maximum permissible density of free electrons and the materials having greater electron density are unable to store tracks of the charged particles.
The tracks will not be produced in the materials having high holes mobility. Because of the rapid outward diffusion of the holes will neutralize the core atoms as a result the tracks will be erased.

2.5 Track Formation in Plastics

The models discussed above explain the mechanism of the track formation in inorganic crystals and glasses and plastics in general. As we are more concerned with CR-39 plastic detector, therefore in this section, track formation mechanism in CR-39 (a polymeric nuclear track detector) in particular, will be discussed.

Ionizing radiations directly produce ionized and excited molecules, and electrons. Some excited molecules may de-excite through the emission of radiation or through non-radiative transitions. Excitation energy can also be transferred from one molecule to another. Electrons are trapped at various sites or can combine with molecules to form negative ions or recombine with positive ions yielding excited molecules. Ions may participate in charge transfer reactions (Durrani & Bull, 1987). Both ions and excited molecules may acquire considerable vibrational energy and undergo bond rupture to form a complex carry of stable molecules, free radicals, ionized molecules and radical ions. Further reactions between these ions, radicals and molecules will then take place.

The net effect on the plastic will be the production of many broken molecular chains, leading to the reduction of the average molecular weight of the substance (radiation can, in fact, also initiate cross linking and thus produce an increase in the average molecular weight of the substance; but this process is probably not related to the track formation). The mechanism of track formation in plastics is represented by Ahlen (1980) who has reported that the rate of chemical attack on a plastic increase as the average molecular weight decreases. A number of authors have also shown that the rate of reaction of a given etchant with a plastic increases as a function of the dose of radiation absorbed by the plastic.
Environmental factors also play an important role in determining the magnitude of the effects of the radiation on plastics. For example, the presence of oxygen either before during or after the irradiation tends to increase the final chemical reactivity generated by the irradiation. It is thought that oxygen tend to combine with ions and radicals, thus preventing their subsequent recombination. Bonfigiioli, Ferro & Nojoni (1961), Benton and Henke (1965) have discussed the effects such as track length increase.

2.5.1 Track Lengths

Firstly the track lengths were traced by using the tracing tube attached with the microscope**.** We measured the actual length of the track after measuring the projected lengths and the depths of the end of the tracks in the detector. For this purpose we used the formula.
$L = [(\text{Projected length x calibration factor})^2 + (\text{Depth x Refractive index of CR-39})^2]^{1/2}$
Where refractive index of CR-39 = 1.50 and calibration factor, 1mm=1.47µm. The depth was also measured by using the linear transducer attached with the microscope.

The widely used plastic detectors are Cellulose Nitrate, Diallyldiglicol carbonate (CR-39), Lexan and Microtel etc.

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 corbonate, $C_{12}H_{18}O_7$) of thickness 600 micron. A number of these detectors were irradiated in vacuum, both perpendicularly and angularly (at 45^o) 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^{o} 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 $_{Vt}$ 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

3.2.1. Scanning

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^{o}) and one sample was irradiated perpendicularly. We measured the track diameter and the lengths of the tracks given in the tables.

4. EXPERIMENTAL FINDINGS

4.1 Plot of the Track Length

The actual track lengths (projected track length x CF) measured in micrometers are plotted against the frequency of occurrence. The graphs thus obtained yielded two peaks (Fig.2.1). Again, the data obtained by Flynn et al from the graph plotted between the fission yield and masses of the fission fragments was utilized (Fig.4.2). The mean value and the standard deviation have been obtained by the Gaussian's fit to the experimental plot of the track lengths of the ffs. Thus we obtained a complete information and correlation between the track lengths and corresponding masses. This result clearly demonstrates the fact that as the mass of the fission fragments increases, the length of the track decreases. The masses of the ffs can be determined if we have a data of track lengths, fig 4.4.

Figure 4.1 Frequency distribution of the Track Lengths (μm) of the ffs

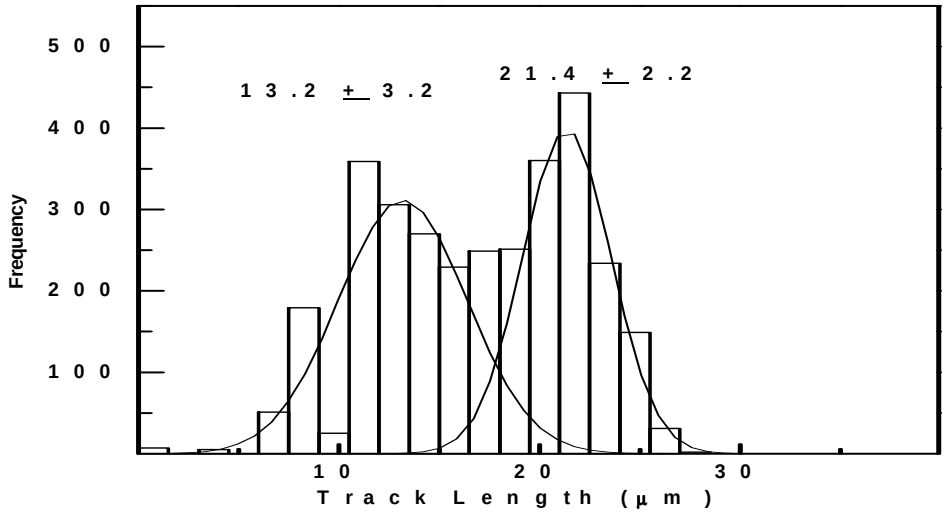


Figure 4.2 Experimental data correlated with the results obtained by Flynn et al,1972.

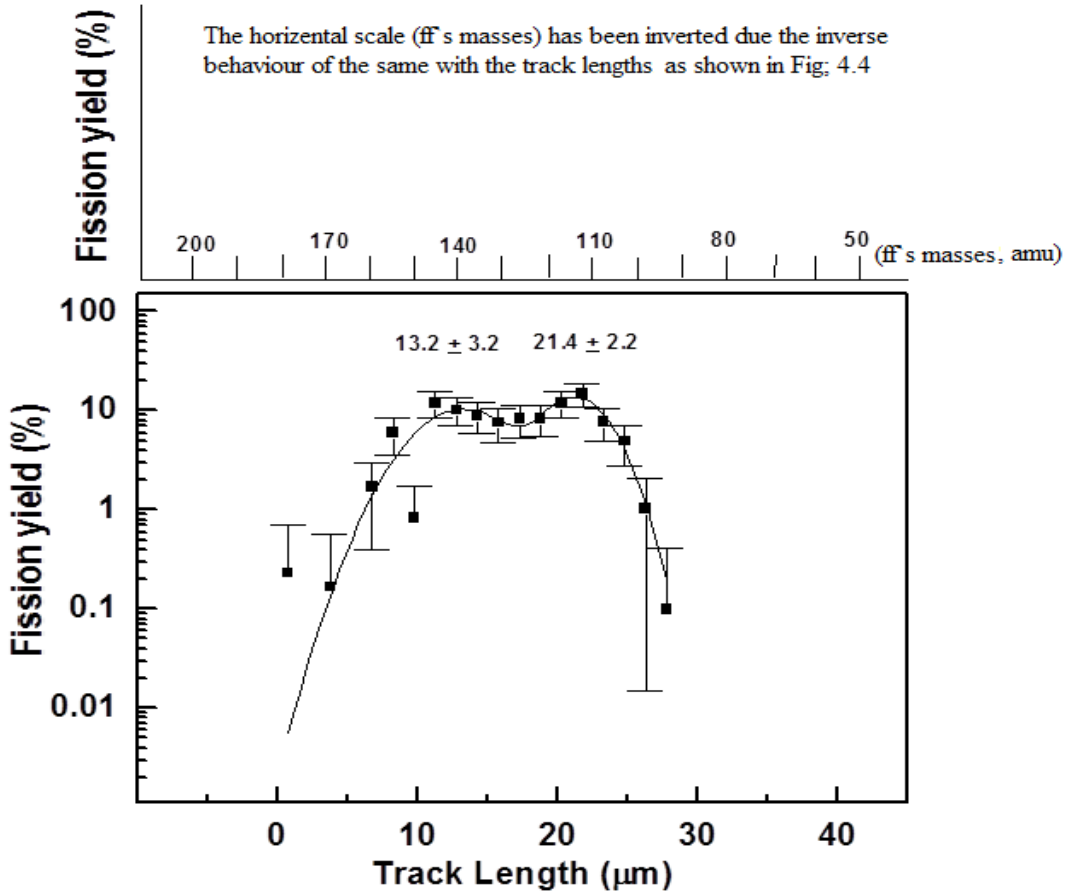


Figure 4.3 % Fission yield as a function of mass number for the spontaneous fission of Cf-252[Flynn, et al, 1972].

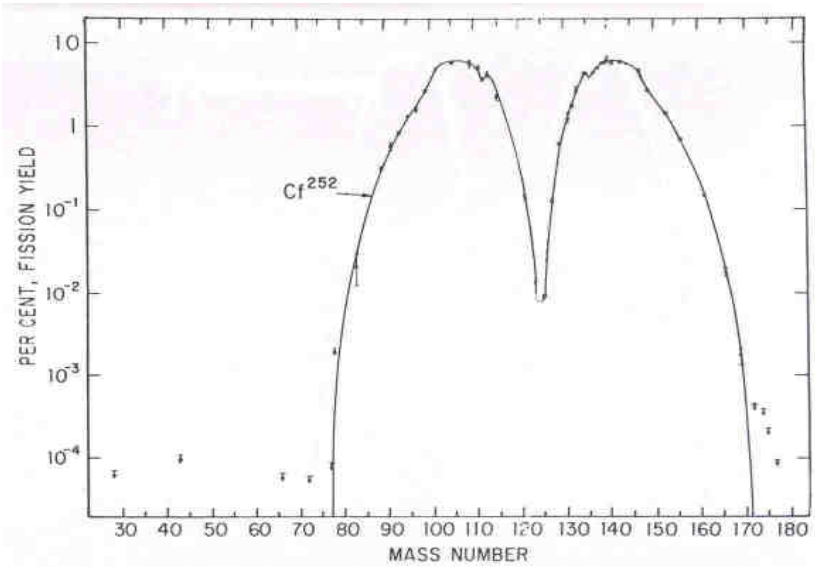
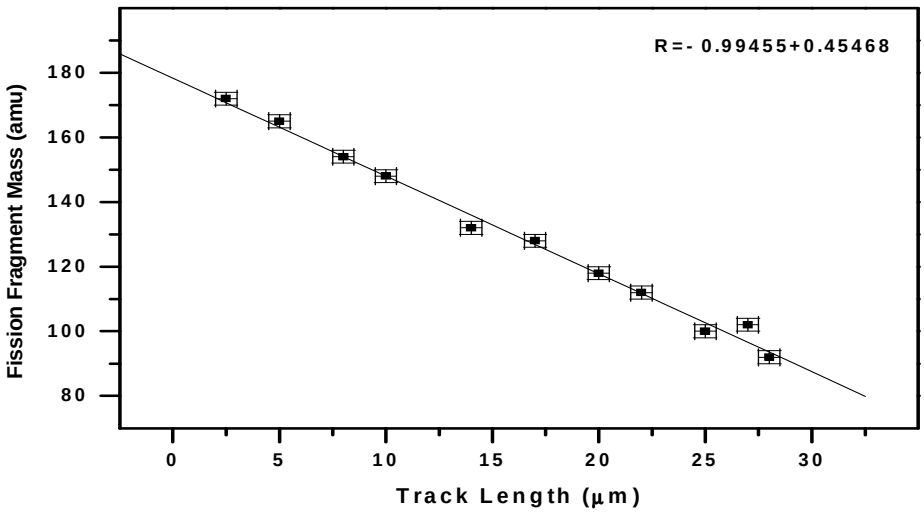


Figure 4.4 Calibration graph between the track lengths (µm) and the ffs masses (amu).



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 ^{o}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; Flynn et al results, who found the relationship between mass numbers and % age fission yields of the ffs of Cf^{252}. The radiochemical technique which is an off line technique, was used to collect the information about the product energies and momentum transfer in the reaction.

To fulfill this purpose, fission fragments of 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 Cf^{252} in vacuum at 45^o and 90^o. The detectors were etched in 6N NAOH at 40^o 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 of the tracks were measured using the projected lengths. 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. 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 Flynn 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 lengths.

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