



ETCHABLE TRACK LENGTHS OF 3.56 GeV ^{208}Pb IN VARIOUS DIELECTRIC SOLIDS

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(Received 29 April 1994; revised 22 September 1994; in final form 10 November 1994)

Abstract—Etchable track lengths of 3.56 GeV ^{208}Pb ions in nine different dielectric materials, namely CR-39 cellulose nitrate, Lexan, Mica, Makrofol-E, Makrofol-N, Triafof-BN, Triafof-TN and X-ray film base, have been determined. The accuracy of the etchable track lengths has been determined and the values are compared with theoretical data obtained from computer codes (1) RANGE (2) TRIM and (3) the program of Henke and Benton.

1. INTRODUCTION

The nuclear track technique has come a long way since its inception three decades ago (Young, 1958). It has been successfully applied to many fields like the development of microfilters (Fleischer *et al.*, 1963; Vater, 1988 and references therein), the study of nuclear reactions (Price and Walker, 1962; Khan, 1985; Gottschalk *et al.*, 1979), fusion-fission phenomenon (Debeauvais and Ralarosy, 1983; Dwivedi and Fiedler, 1988; Dwivedi *et al.*, 1993a) and to areas such as archaeological, geological and cosmological sciences (Fleischer *et al.*, 1975). Newly emerged technological applications involving microstructuring and heavy ion lithography (Spohr, 1990) have great applied value. In a number of applications involving the track technique, a knowledge of the track length of different heavy ions in elemental and complex media is essential.

The track length is that portion of the range of the heavy ion over which the actual energy loss rate remains above the critical value $(dE/dx)_c$, which is characteristic of a particular SSNTD. For a given energy a given particle has a fairly definite range in a particular track detector provided there are no nuclear reactions before it reaches the end of its trajectory.

Nine different detectors; eight plastics and one crystalline detector were used for this study. The maximum etchable track length of 3.56 GeV ^{208}Pb was determined in the different detectors. These experimental results were then compared with theoretical values obtained from three different computer codes: (1) RANGE (Dwivedi, 1988); (2) TRIM (Biersack and Haagmark, 1980); and (3) the program of Henke and Benton (HB) (Henke and Benton, 1968). The significant observations of these works are highlighted.

2. EXPERIMENTAL

The experimental technique is discussed in the following sections.

2.1. The track detectors

In the present work nine track detectors were used to register tracks of 3.56 GeV ^{208}Pb ions. These include a muscovite mica and eight plastic detectors of the following specifications:

Muscovite Mica: Circular sheets (diameter = 50 mm and thickness 400 μm) of muscovite mica $[\text{KA}_1\text{Si}_3\text{O}_{10}(\text{OH})_2]$ were used for irradiation. The density of the material was found to be 2.81 g/ml.

Triafof-TN: It is a blue coloured cellulose triacetate $[\text{C}_3\text{H}_4\text{O}_2]$ manufactured by Bayer AG, Germany. A 100 μm foil was cut into rectangular pieces (2 cm \times 3 cm) and a stack of two foils was prepared.

The density of Triafof-TN was found to be 1.15 g/ml. Triafof-BN: This plastic material is chemically characterized as cellulose acetate butyrate $[\text{C}_{20}\text{H}_{32}\text{O}_5]$ and is manufactured by Bayer AG, Germany. Rectangular piece (2 cm \times 3 cm) of 200 μm thick foil was cut for the purpose of irradiation. The material density was determined to be 1.20 g/ml.

Makrofol-N and Makrofol-E: These commercial polycarbonates $[\text{C}_{16}\text{H}_{14}\text{O}_3]$ are manufactured by Bayer AG, Germany. Makrofol-N (100 μm) is yellow coloured film having a density 1.23 g/ml whereas the colourless Makrofol-E (200 μm) has a density 1.14 g/ml. These materials were cut into rectangular pieces for irradiation.

Lexan: A non-UV sensitized polycarbonate film manufactured by General Electric Co., U.S.A. and available under the trade name Lexan $[\text{C}_{16}\text{H}_{14}\text{O}_3]$ was used in the present work. The transparent Lexan film (thickness: 500 μm and density: 1.20 g/ml)

was obtained by removing protective polythene layers.

CR-39: Square pieces (2 cm × 2 cm) of CR-39 plates were used for exposure to lead ions. The material is manufactured by Homalite Corp., Delaware, U.S.A. and has a thickness of about 1.5 mm. It is one of the most widely used track detectors and is characterized by the chemical formula $C_{12}H_{18}O_7$ and density 1.32 g/ml.

X-ray film base: This material is obtained from commercial screen type X-ray film (INDU, Hindustan Photo Film Co., Ootacamund, India) by dissolving the sensitive gelatin layer in warm NaOH solution. It is then washed in running water for 5 minutes and dried in air. The light blue coloured plastic sheet (thickness: 200 μ m) was then characterized by elemental analysis and was found to be a cellulose acetate with chemical formula $C_{10}H_{14}O_7$. The density of the material was found to be 1.30 g/ml. **Cellulose nitrate:** Small pieces of a 500 μ m thick film of cellulose nitrate [$C_6H_8O_9N_2$] from Nixon-Baldwin, U.S.A. were used. The density of the material was found to be 1.49 g/ml.

The detector characteristics and the suitable etching conditions are given in Table 1.

2.2. Irradiation

The detectors were irradiated by a well-collimated beam of ^{208}Pb ions with an energy of 3.56 GeV. The irradiations were done at the XO channel of UNILAC, GSI, Darmstadt at an angle of 45° to the detector surface. The irradiations were done at a fluence of $\approx 10^4$ ions/cm 2 .

2.3. Chemical etching of detectors

The first step in etching was the optimization of the etching conditions. While 6 N NaOH appeared to be the best etchant for most plastics, some plastics like Triafol-TN, Triafol-BN, Makrofol-N and cellulose nitrate were etched in 3 N NaOH in order to minimize surface roughness. The X-ray film base was

etched in 2 N NaOH solution. In the case of muscovite Mica we used 20% HF as a suitable etchant instead of 40% or 48% HF. This resulted in slow etching, which allowed enough time for the etchant to diffuse along the trajectory of the particle and also for removal of etch products. A uniform and homogeneous etching was the outcome, which is essential for track studies. The different etchants used for the various detectors are listed in Table 1, along with the etching conditions. The tracks were etched until their tips became rounded, ensuring complete etching.

2.4. Measurement of track parameters

The projected track lengths were measured from the centre of track face to the tip and the track diameters were taken as the minor axis of the ellipse formed by the track face. The measurements were done using an optical microscope (Leitz, Laborlux D). The true track lengths or the maximum etchable track lengths were derived from the projected track lengths (l) using the formulation given by Dwivedi and Mukherji (1979):

$$L = l/\cos \theta + V_G t/\sin \theta - V_G(t - t_c), \quad (1)$$

where V_G is the bulk etch rate, θ is the angle of incidence (in this case 45°), t is the total etching time and t_c the time taken to etch the track completely. As the tracks were etched for a time $t = t_c$ the over-etching factor $V_G(t - t_c)$ vanished. For Mica, the thickness of surface removed due to bulk etching is negligible ($< 1 \mu\text{m}$), hence the maximum etchable track length was determined from the projected track length using the formula (Dwivedi and Mukherji, 1979):

$$L = l \sec \theta. \quad (2)$$

About 300 tracks were measured in each foil. From the measured data a track length distribution curve was plotted and the most probable track length was obtained from the peak position of the distribution curve.

Table 1. Chemical composition, colour, density and etching conditions for different track detectors

SSNTD	Chemical composition	Colour	Density* (g/ml)	Etching conditions
Triafol-TN	$C_3H_4O_2$	Blue	1.15	3 N NaOH/55°C/120 min
Triafol-BN	$C_{20}H_{32}O_5$	Violet	1.20	3 N NaOH/55°C/480 min
Makrofol-N	$C_{16}H_{14}O_3$	Yellow	1.23	3 N NaOH/55°C/215 min
Makrofol-E	$C_{16}H_{14}O_3$	Colourless	1.14	6 N NaOH/55°C/100 min
Lexan	$C_{16}H_{14}O_3$	Colourless	1.20	6 N NaOH/55°C/110 min
CR-39	$C_{12}H_{18}O_7$	Colourless	1.32	6 N NaOH/55°C/120 min
X-ray film Base	$C_{10}H_{14}O_7$	Light Blue	1.30	2 N NaOH/55°C/225 min
Cellulose Nitrate	$C_6H_8O_9N_2$	Colourless	1.49	3 N NaOH/35°C/180 min
Muscovite Mica	$KAl_3Si_3O_{10}(OH)_2$	Brown	2.81	20% HF/30°C/120 min

*The densities were experimentally determined for all the detectors except in the case of CR-39 for which the value is derived from the literature (Durrani and Bull, 1987).

2.5. Experimental errors

The energy of ²⁰⁸Pb ions impinging on the detectors was measured accurately (within 0.1%) with the help of a TOF system at UNILAC, GSI, Darmstadt. The angle of incidence for collimated ions was accurate to $\pm 1^\circ$. The overall errors in track length measurements were $\pm 1.2 \mu\text{m}$ while that of the track diameter was $\pm 0.56 \mu\text{m}$. The standard deviations obtained from the track length distribution curves vary between $2.2 \mu\text{m}$ to $5.2 \mu\text{m}$ and are given in Table 2 along with the most probable track lengths. The spread of experimental ranges in all the nine solids is within 1.7% of the mean values.

3. THEORETICAL ASPECTS OF THE COMPUTER CODES

The penetration of energetic ions in matter has been the subject of theoretical and experimental investigations and evinced a lot of interest as shown by the availability of a vast amount of literature in this field. Here, we have used the computer codes 'RANGE', and 'TRIM' and the program of Henke and Benton to compare the calculated ranges with our measured values. Brief outlines of the theoretical basis of these computer programs are given below.

3.1. Computer code 'RANGE'

The computer code 'RANGE' developed by Dwivedi (1988) is based on the universal stopping-power equations of Mukherji and co-workers (Mukherji and Srivastava, 1974; Srivastava and Mukherji, 1976; Mukherji and Nayak, 1979). It is written in Fortran with a subroutine HEART which contains all the thirteen stopping-power equations valid for specific velocity regions of stopping ions. For complex media, the stopping-powers $[(dE/dx)_E]_k$ have been obtained using Bragg's additivity rule.

The total penetration depth or range R is calculated as

$$R(\text{mg}/\text{cm}^2) = \sum_{E_i}^{E_0} \frac{\delta E}{[(dE/dx)_E]_k},$$

where δE is a small but finite energy interval (0.01 MeV/u) over which the stopping-power remains virtually constant, E_i is the initial ion energy in MeV and E_0 is the final energy corresponding to the ion velocity V_0 ($=c^2/\hbar$). This code requires a small area ($< 15 \text{ kb}$) and can be executed on any personal computer for calculating ranges, track lengths, total energy loss rate for both accelerating and stopping ions in any medium of known chemical composition. The output may be obtained in the form of either a plot or a printed data table.

3.2. The computer code 'TRIM'

A high efficiency computer code TRIM (TRANSPORT of Ions in Matter) has been developed by Biersack and Haagmark (1980) that is based on the Monte Carlo simulation of slowing down and scattering of energetic ions in amorphous targets. The calculations for electronic energy loss at low energies are based on the velocity dependent treatment of Lindhard and Scharff (1961); at high energy regions the Bethe-Bloch equation is used. In order to bridge the gap between low energy and high energy regions, the interpolation scheme of Biersack and Fink (1974, 1975) has been employed. The basic code TRIM and its several advanced and modified versions are very widely used by laboratories engaged in ion implantation, radiation damage, heavy ion research and semiconductor technology.

3.3. Henke and Benton's program

Henke and Benton have used a general method for the calculation of range, energy and energy loss relations for a number of ions in several dielectric

Table 2. Etchable track lengths of 3.56 GeV ²⁰⁸Pb in nine detectors along with corresponding theoretical values from three different computer codes

SSNTD	Track Length (μm)			
	Experimental (Present work)	Theoretical		
		RANGE*	TRIM†	HB‡
Triafol-TN	253.60 ± 3.6	254.2	272.0	246.0
Triafol-BN	219.70 ± 5.2	245.5	251.0	222.0
Makrofol-N	234.60 ± 4.0	235.0	257.0	225.0
Makrofol-E	265.00 ± 4.5	253.0	278.0	243.0
Lexan	229.30 ± 3.3	240.0	264.0	230.0
CR-39	213.20 ± 2.4	217.0	237.0	210.0
X-ray Film Base	212.50 ± 2.6	225.0	249.0	205.0
Cellulose Nitrate	202.20 ± 4.5	210.0	224.0	200.0
Muscovite Mica	125.40 ± 2.2	131.8	138.0	130.4

*Computer code 'RANGE' (Dwivedi, 1988).

†Computer code 'TRIM' (Biersack and Haagmark, 1980).

‡Computer code 'HB' (Henke and Benton, 1968).

track detectors (Henke and Benton, 1968). For covering a broad spectrum of particles, energies and stopping materials, the methods of Barkas and Berger (1964) have been used after suitable modifications and extensions to include lower energies.

In this program the range of a charged particle in matter is expressed in relation to that of an 'ideal' proton. An ideal proton has been defined as a particle having protonic mass and charge which does not capture electrons or interact strongly with nuclei. The range extension in the material is dependent on the charge of the ion and is caused by the neutralization of the ion towards the end of the trajectory. This program requires the input of the following five parameters:

- A/Z : atomic mass per electron of the stopping material in amu/electron;
- I_{adj} : adjusted mean ionization potential in eV;
- Z : atomic number of the ion;
- M : mass of the ion;
- E : energy of the ion.

The rate of energy loss is obtained by differentiating the range equation. Henke and Benton's program calculates range and energy loss of heavy ions in any stopping material in the energy region of 0.1–2000 MeV/u.

4. RESULTS AND DISCUSSION

The quality of etched tracks of ^{208}Pb registered in different detectors varies from one material to another. While Mica and CR-39 were found to be the best at registering well-defined tracks, the polycarbonates (namely Lexan, Makrofol-N and Makrofol-E) also register tracks very well. The cellulose acetates (namely Triafol-TN, Triafol-BN and X-ray film base) were, however, not good at registering smooth tracks. The tracks formed in these detectors were neither narrow nor well-defined, unlike those formed in the other detectors. The average etchable track lengths of ^{208}Pb in different detectors were determined from the maxima of the track length distribution curves. The

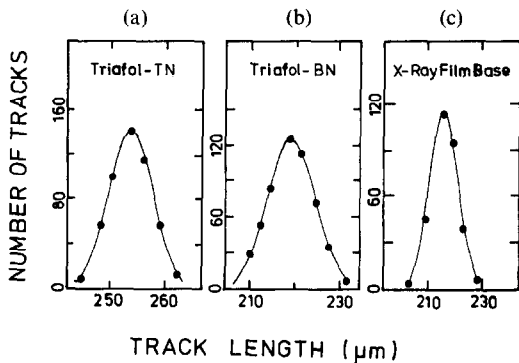


Fig. 1. Plot showing the track length distributions of 3.56 GeV ^{208}Pb in (a) Triafol-TN, (b) Triafol-BN and (c) X-ray film base.

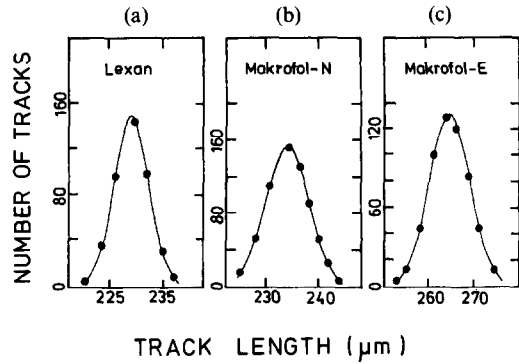


Fig. 2. Plot showing the track length distributions of 3.56 GeV ^{208}Pb in (a) Lexan, (b) Makrofol-N and (c) Makrofol-E.

standard deviations were found to vary between 2.2 and 5.2 μm . The track length distribution curves for these detectors are shown in Figs 1, 2 and 3. Table 2 gives a list of the experimental values along with the standard deviations and three sets of theoretical values obtained from computer codes RANGE (Dwivedi, 1988), TRIM (Biersack and Haagmark, 1980) and the program (HB) of Henke and Benton (1968).

While comparing the theoretical and experimental ranges it has been observed that the values derived from the code TRIM are consistently higher than the corresponding experimental ranges by a margin of 3σ or more for all the detector solids used in this work. Theoretical ranges from the code HB agree within 2σ for six solids but for remaining three solids the values are underestimated by 2σ or more. For Triafol-TN, Makrofol-N, CR-39 and cellulose nitrate the calculated ranges from the code RANGE agree with experimental values within 2σ , for Makrofol-E, Triafol-BN, Lexan, X-ray film base and Mica the RANGE values deviate by more than $\pm 2\sigma$. On the basis of the 'minimum sum of squares of deviation' between the experimental and three sets of the theoretical ranges, it has been found that for 3.56 GeV ^{208}Pb ranges in nine solids, the computer code HB (Henke and Benton, 1968) predicts the most reliable

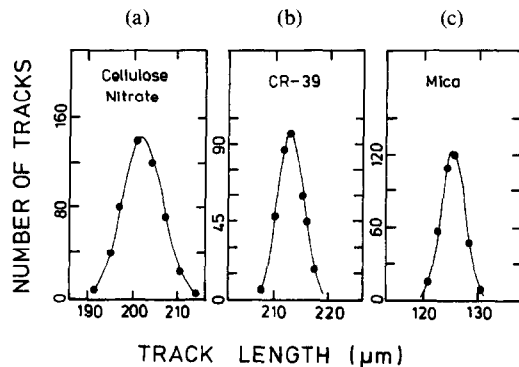


Fig. 3. Plot showing the track length distributions of 3.56 GeV ^{208}Pb in (a) Cellulose nitrate, (b) CR-39 and (c) Mica.

data. The values obtained from the code RANGE (Dwivedi, 1988) have shown limited applicability while the code TRIM (Biersack and Haagmark, 1980) predicted the least reliable data in the present case. From the standard deviations, the spread of experimental ranges around the mean values was determined for all nine solids and was found to be 1.7%. The mean of deviations between experimental and theoretical ranges obtained from three computer codes HB, RANGE and TRIM was evaluated and found to be 3.0%, 4.2% and 11.1%, respectively.

This work has qualitatively supported some earlier findings (Dwivedi, 1991; Dwivedi *et al.*, 1993b; Ghosh *et al.*, 1988, 1994; Jain *et al.*, 1989, 1991; Kocsis *et al.*, 1993; Saxena and Dwivedi, 1990) barring a few cases (Ghosh *et al.*, 1994; Kocsis *et al.*, 1993) that the computer code 'RANGE' may be used to derive fairly reliable values of heavy ion ranges in complex solids. Though the validity of theoretical ranges of heavy ions from computer code 'RANGE' has been tested below 20 MeV/u, the program from Henke and Benton is applicable up to 500 MeV/u. Recently Kocsis *et al.* (1993) reported that theoretical ranges of ^{20}Ne in CR-39 from code HB agree fairly well with the experimental values. The program TRIM is frequently used by groups engaged in ion implantation and heavy ion interactions due to its reliability at low energies but for higher energies (> 10 MeV/u) it has shown large discrepancies in a number of cases (Ghosh, 1990; Ghosh *et al.*, 1994; Kocsis *et al.*, 1993). For some ion-detector pairs (Dwivedi, 1991; Dwivedi *et al.*, 1993b) the TRIM ranges are in good agreement with experimental ranges. More experimental range data, both in elemental and complex media, are needed before making valid conclusions on the general reliability of these computer codes.

Acknowledgements—We thank the staff of UNILAC, GSI, Darmstadt for providing ^{208}Pb irradiations. We acknowledge the receipt of an equipment grant from the German Agency for Technical Cooperation (DGTZ), Germany.

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