

Energy calibration of a CR-39 nuclear-track detector irradiated by charged particles

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Abstract Charged particle diagnosis is an important aspect of laser–plasma experiments conducted at super-intense laser facilities. In recent years, Columbia Resin #39 (CR-39) detectors have been widely employed for detecting charged particles in laser–plasma experiments. This is because the CR-39 polymer does not respond to electromagnetic pulses or X-rays. This study presents a method for calibrating the relationship between particle energy and track diameter in a CR-39 detector (TasTrak[®]) using 3–8 MeV protons, 6–30 MeV carbon ions, and 1–5 MeV alpha particles. The particle tracks were compared under the manufacturer’s recommended etching conditions of 6.25 mol/l NaOH at 98 °C and under the widely adopted experimental conditions of 6.25 mol/l NaOH at 70 °C. The results show that if the NaOH solution concentration is 6.25 mol/l, then the temperature of 70 °C is more suitable for etching proton tracks than 98 °C and employing a temperature of 98 °C to etch alpha-particle and carbon-ion tracks can significantly reduce the etching time. Moreover,

this result implies that C³⁺ ion or alpha-particle tracks can be distinguished from proton tracks with energy above 3 MeV by controlling the etching time. This calibration method for the CR-39 detector can be applied to the diagnosis of reaction products in laser–plasma experiments.

Keywords CR-39 detector · Energy calibration · Bulk etch rate · Etching temperature

1 Introduction

Charged particle identification is an important method for observing laser–plasma experiments, because the spectral characteristics of reaction products can provide direct information concerning the plasma conditions in the experiment [1–4]. With the extreme laser intensities currently utilized at terawatt facilities for charged particle acceleration, the demand for discriminating charged particles is also increasing [5]. Laser–plasma experiments involve considerably more difficult particle discrimination tasks than conventional nuclear reaction experiments, as the reaction products contain discrete lines and continua from protons, deuterons, tritons, ³He, carbon ions, and alpha particles [1, 2, 6]. The requirements for detectors that can discriminate between these charged particles include a high detection efficiency, large dynamic range, and good energy resolution. Owing to the strong electromagnetic radiation of X-rays produced by the laser interacting with the target, detectors must also be insensitive to background radiation and electromagnetic noise.

Columbia Resin #39 (CR-39) polymer has been widely employed as a track detector in recent laser–plasma

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experiments [7]. CR-39 detectors do not respond to electromagnetic pulses (EMPs) and X-rays and achieve 100% detection efficiency for protons up to approximately 6–8 MeV if the proton incidence is normal to the detector surface. In addition, 100% detection efficiency is possible for alpha particles and carbon ions. In laser–plasma experiments, CR-39 combined with a fixed-thickness filter can be utilized to identify protons and other charged particles. CR-39 has been used in conjunction with a wedge-range filter (WRF) [8], which can deliver an accurate reconstruction of the proton spectrum over a wide range of incident energies at the OMEGA laser facility [2] and can be covered with aluminum foil of various thicknesses to measure the numbers of charged particles [1, 9]. Furthermore, a CR-39 detector can also be placed downstream of a Thomson parabola spectrometer (TPS), which separates particles of different ionic species and charge states, to form a magnetic charged particle spectrometer that can detect particles of different charges and masses [6]. In addition, CR-39 detectors have been employed in neutron spectrometers using the recoil technique at the OMEGA laser facility [10] and have also been widely utilized in the field of laser-ion acceleration diagnostics [5, 11].

CR-39 is a clear and rigid plastic polymer whose chemical formula is $C_{12}H_{18}O_7$ (polyallyldiglycol carbonate). When a charged particle enters CR-39, it breaks the material's polymeric bonds and leaves a trail of damage along its path, forming a so-called latent track [12]. The amount of local damage along the track is indirectly related to the loss of energy per unit path length, and the length of the latent track indicates the range of the particle in the plastic. These latent tracks can be observed under an optical microscope after etching the material in NaOH solution because the damaged parts of the material react more intensely with the etching agent than the undamaged parts. During the etching process, the bulk etch rate (V_B) and track etch rate (V_T) are used to describe the growth of the track. Here, V_B and V_T are the etch rates of the undamaged and damaged regions of the material, respectively. V_B is constant under a given temperature and etching solution concentration, whereas V_T is closely related with the particle energy lost along the latent track. When particles enter normally to the surface of the CR-39, the track forms a conical pit after etching in NaOH solution. As long as the etching depth is smaller than the particle range, the diameter and depth of the track increase with the increase in etching time. The track position shows where the particles were incident, and the average track diameter will depend on the mean incident energy and how the CR-39 is processed.

To use these parameters to discriminate the particles produced by a high-intensity laser experiment, the CR-39 detector must be carefully calibrated in experiments with well-known particles, to determine the relationship

between the measured dimensions and particle characteristics. The etching process depends on the etching time and the temperature and molarity of the NaOH solution, which can be controlled in each experiment. These parameters should be chosen to ensure the optimal detectability of tracks, because larger values of the ratio V_T/V_B will generally result in larger track diameters and depths. Previous work has generally adopted molar concentrations of NaOH of 6 or 6.25 mol/l, because V_T/V_B is maximized at molar concentrations of approximately 6.25 mol/l at temperatures over 60 °C [13]. Azooz's experiment measured the value of V_B in 6.25 mol/l NaOH solution over a temperature range from 65 to 85 °C [14], finding that V_B increases monotonically with temperature. We measured the value of V_B in 6.25 mol/l NaOH solution at 98 °C and fitted the bulk etch-rate curve for temperatures from 25 to 98 °C, in combination with other experimental data [12, 15]. Previous studies have investigated the response of CR-39 to charged particles at low etching temperatures [6, 16–22]. However, little research has addressed how high etching temperatures can affect CR-39's usefulness for discriminating charged particles [5]. Seimetz's experiment measured the proton track diameter in 6.25 mol/l NaOH solution at 90 °C [5]. In addition, because 6.25 mol/l NaOH at a temperature of 70 °C is widely adopted in many experiments, we also etched CR-39 under this etching condition, for the sake of comparison with experimental data from other laboratories.

We tested different etching temperatures to collect calibration data for CR-39 irradiated by protons, carbon ions, and alpha particles. Protons and carbon ions were produced by an electrostatic accelerator, and alpha particles were emitted by an ^{241}Am isotope. A solution of 6.25 mol/l NaOH at temperatures of 70 °C and 98 °C was used to etch the CR-39. We tested etching times of 6 h, 12 h, and 18 h at 70 °C, and from 1 to 6 h at 98 °C. These tests were performed to determine the optimal conditions for distinguishing particle tracks. Our calibration results show that the sizes of alpha-particle and carbon-ion track diameters etched for 1 h at 98 °C are approximately the same as those etched for 6 h at 70 °C, and C^{3+} ion or alpha-particle tracks can be distinguished from proton tracks by varying the etching time.

2 Experimental section

CR-39 detectors were irradiated by 3–8 MeV mono-energetic protons, 6–30 MeV mono-energetic carbon ions, and 1.1–4.9 MeV mono-energetic alpha particles. Protons and carbon ions were produced by the 2×6 MV tandem accelerator at Peking University's Institute of Heavy Ion Physics.

As shown in Fig. 1, the target holder has four working faces. One is a fluorescent target, which is used to adjust the particle beam, and the other three are used for installing CR-39 detectors. As shown in the top view, the beam will scan back and forth on the target with an intensity of $10^4 \text{ cm}^{-2} \text{ s}^{-1}$. A plastic scintillator detector is available to monitor the number of particles that reach the target. However, its accuracy is not high, and the scintillator pulse counts can only be used as a rough estimate. By controlling the irradiation time, the number of particles incident on the CR-39 was kept at less than 10^6 cm^{-2} . Table 1 lists the particle types, energies, and irradiation times used in our calibration tests.

As shown in Fig. 2, to generate alpha particles with different energies, we employed an ^{241}Am isotope emitting 5.4 MeV alpha particles with an intensity of 7.197 kBq. We varied the distance between the isotope source and detector, to vary the incident energies of the alpha particles. The alpha-particle energy was first recorded using a silicon carbide (SiC) detector, and then, the CR-39 detector was placed at the position of the SiC detector so that it could be irradiated with alpha particles of known energy. By controlling the irradiation time, the number of particles incident to the CR-39 was controlled to be less than 10^6 cm^{-2} . The air gap distance between the detector and source, resulting alpha energy, and irradiation time is listed in Table 2.

For comparison, we tested the etching conditions of 6.25 mol/l NaOH at 98 °C and 6.25 mol/l NaOH at 70 °C. The manufacturer's recommended alpha etching conditions are 6.25 mol/l NaOH at 98 °C for 60 min. The conditions of 6.25 mol/l NaOH at 70 °C have been adopted in many previous studies. The CR-39 detectors used in our experiments were manufactured by Track Analysis Systems Ltd.,

(TASL) in Bristol, UK, and are of size 25 mm \times 25 mm \times 1.5 mm. Information about the track position, track diameter, gray level, and other track information was recorded using the TASLIMAGE radon dosimetry system from the same manufacturer. As mentioned in the introduction, the track diameter is often utilized to identify the particle type, and this diameter mainly depends on the ion energy, ion mass, etching time, and etching conditions (solution concentration and temperature). By default, the TASLIMAGE software filters out tracks with diameters less than 3 microns or greater than 40 microns, to remove background noise.

Figure 3 presents an example of 4.42 MeV alpha-particle tracks after etching for 1 h in 6.25 mol/l NaOH at 98 °C. Figure 4 presents the histogram distribution of the above alpha-particle track diameters on one square centimeter of the CR-39 sample. Gaussian fitting was employed to obtain the mean and standard deviation of the track diameters, and the standard deviation was used as the error. The means and errors of the other charged particle track diameters were also calculated by the same method.

3 Results and discussion

3.1 Bulk etch rate in 6.25 mol/l NaOH

The bulk etch rate V_B is the rate at which the undamaged surface of the detector is removed [12]. In this study, gravimetric measurements were used to measure the bulk

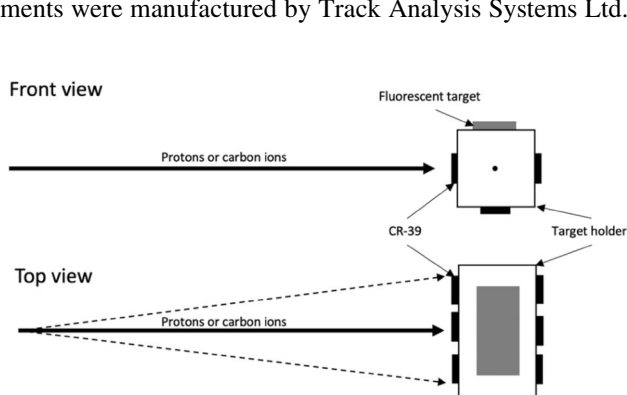


Fig. 1 Experimental setup used to irradiate CR-39 samples by mono-energetic protons and carbon ions

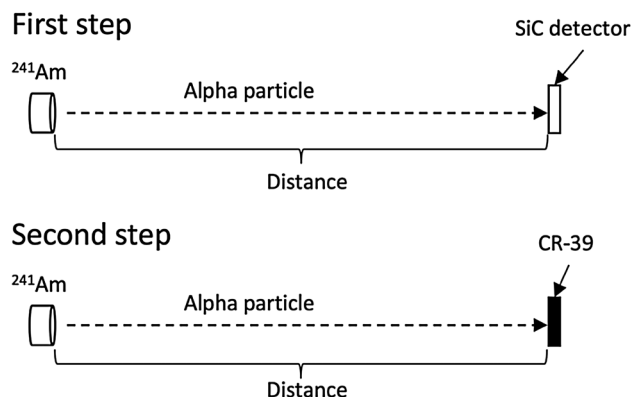


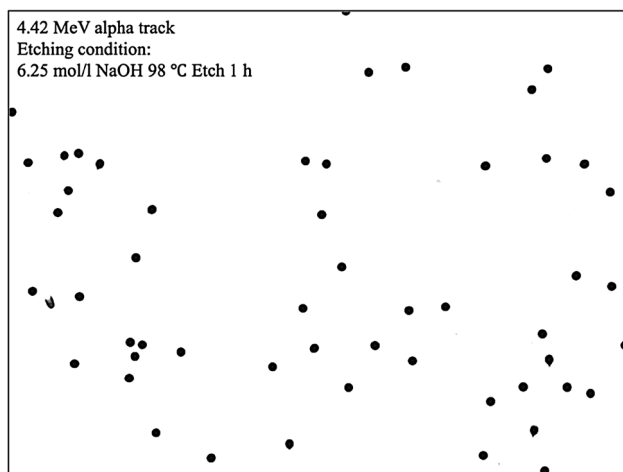
Fig. 2 Experimental setup used to irradiate CR-39 samples with mono-energetic alpha particles

Table 1 Protons and carbon ions produced by the accelerator

Particle type	H ⁺	H ⁺	H ⁺	H ⁺	H ⁺	C ³⁺	C ³⁺	C ³⁺	C ³⁺	C ⁴⁺	C ⁵⁺
Energy (MeV)	3	4	5	6	8	6	10	15	20	25	30
Irradiation time (s)	89.6	50.1	93.9	62.4	80.7	41.2	92.9	46.5	68.8	98.4	76
Scintillator count	261	263	261	261	262	263	261	261	261	261	261

Table 2 ^{241}Am isotope alpha-particle source and energy–distance correlations

Particle type	He	He	He	He	He	He	He	He	He	He
Distance (mm)	35	30	28	25	23	20	15	13	10	5
Energy (keV)	1105	2114	2361	2807	3061	3446	3940	4160	4420	4915
Irradiation time (s)	40	40	30	30	30	20	20	20	20	20

**Fig. 3** Microscope images ($\times 20$ magnification and pixel size of 0.8 microns) of 4.42 MeV alpha-particle tracks etched for 1 h in 6.25 mol/l NaOH at 98 °C

etch rate of CR-39 in 6.25 mol/l NaOH at 98 °C. The etch rate is given by $V_B = \Delta m / 2\rho h^2 t$, where Δm is the difference in the mass of the CR-39 before and after etching, ρ is the density, h is the side length of the CR-39 square, and t is the etching time. Gaussian fitting returns a bulk etch rate of $9.95 \pm 1.33 \mu\text{m/h}$ from the data measured from 30 CR-39 samples in our experiments.

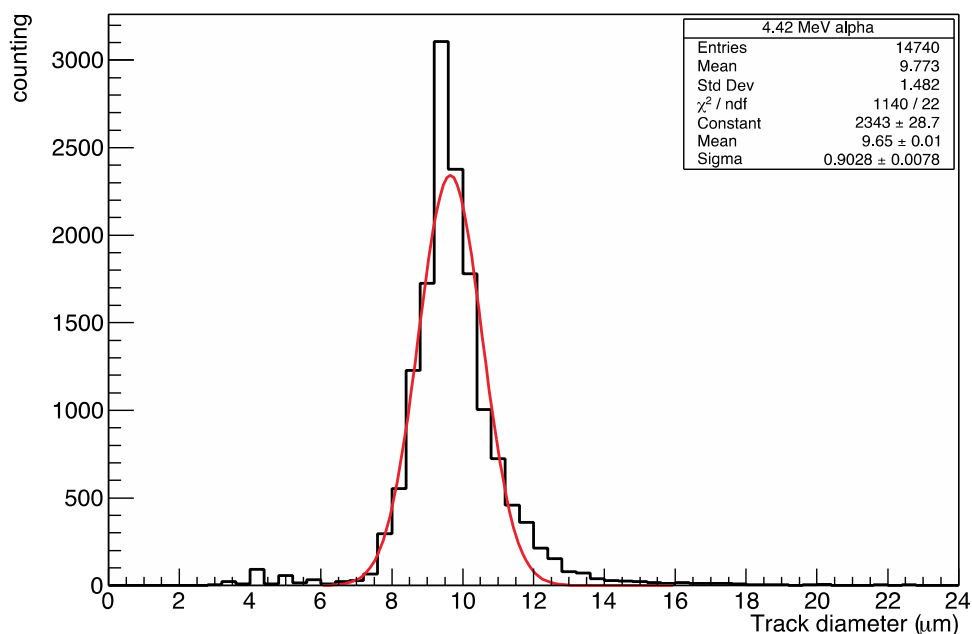
Fig. 4 Histogram distribution of 4.42 MeV alpha-particle track diameters

Figure 5 plots the bulk etch rate in 6.25 mol/l NaOH solution at different etching temperatures. The CR-39 detectors used by Azooz [14], Ho [15], and Fowler [23] were manufactured by Page Mouldings (Pershore) Limited (Worcestershire, UK). As the plot shows, the bulk etch rate curve increases exponentially with temperature, and the bulk etch rate is almost the same for the two types of CR-39 at 75 °C. Therefore, supplementing the bulk etch rate in 6.25 mol/l NaOH solution at 98 °C can optimize the bulk etch rate. The error is explained by discrepancies in the CR-39 samples' masses and sizes. The bulk etch-rate curve function is $V_B = ae^{bT}$, where $a = 0.00669 \pm 0.00107$, $b = 0.07461 \pm 0.00213$, and T is the etching temperature (°C).

According to the bulk etch-rate curve, when the temperature is lower than 80 °C, the bulk etch rate is less than $2.61 \mu\text{m/h}$, explaining why many previous experiments choose to etch CR-39 at a temperature of 80 °C. When the etching temperature is 98 °C, the bulk etch rate is large, helping to reduce the etching time so that tracks appear quickly. Our results show that tracks of alpha particles and carbon ions can appear within 1 h and that the etching effect is equivalent to etching at a temperature of 70 °C for 6 h.

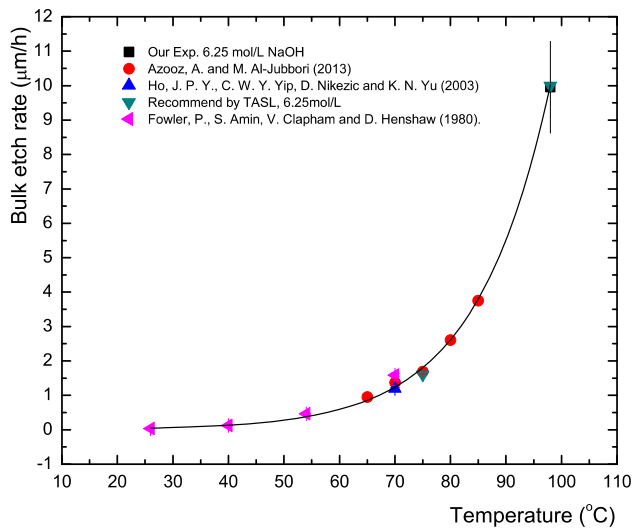


Fig. 5 Bulk etch-rate curve in 6.25 mol/l NaOH solution at different etching temperatures

3.2 Track diameters for protons in 6.25 mol/l and 7.25 mol/l NaOH at 98 °C and 70 °C

The calibration of proton track diameters for 1500 μ thick CR-39 was studied after determining the optimal etching time. As shown in Fig. 6, the proton track diameter is inversely related to the energy in the range of 3 to 5 MeV in 6.25 mol/l NaOH at a temperature of 98 °C. For 5 MeV proton tracks, increasing the etching time does not increase the size of the proton track. Moreover, tracks from 6 to 8 MeV, protons were not apparent after etching for 6 h. Figure 7 depicts the proton track diameter data after etching in 6.25 mol/l NaOH and 7.25 mol/l NaOH at 70 °C. The proton track diameter decreases as the incident energy increases from 3 to 8 MeV. Furthermore, the proton track diameter at energies below 5 MeV etched for 18 h in 7.25 mol/l NaOH is clearly larger than that in 6.25 mol/l NaOH etched for 18 h, showing that a molarity of 7.25 NaOH increases the proton track diameter at energies below 5 MeV than a molarity of 6.25 NaOH.

Comparing the results for the proton track diameter etched for 6 h at a temperature of 98 °C in Fig. 6 with those etched for 12 h at a temperature of 70 °C in Fig. 7, the former data do not include any 6 or 8 MeV proton tracks, while the latter does. Because V_B increases exponentially with the temperature and is approximately six times greater at 98 °C than at 70 °C, etching for 2 h at 98 °C is equivalent to etching for 12 h at 70 °C. However, the result of etching for 2 h at 98 °C is less precise than that of etching for 12 h at 70 °C. Therefore, 70 °C is more suitable than 98 °C for etching proton tracks.

Previous studies have addressed the track diameter response curves of protons in the energy range of 1–9 MeV

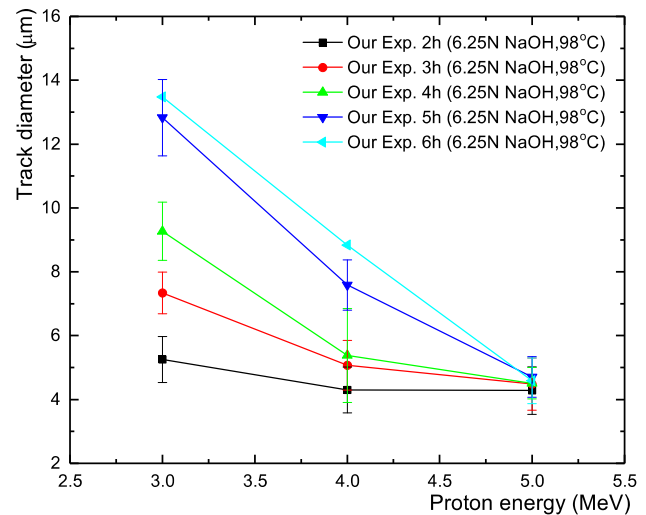


Fig. 6 Proton track diameter versus energy in 6.25 mol/l NaOH at 98 °C

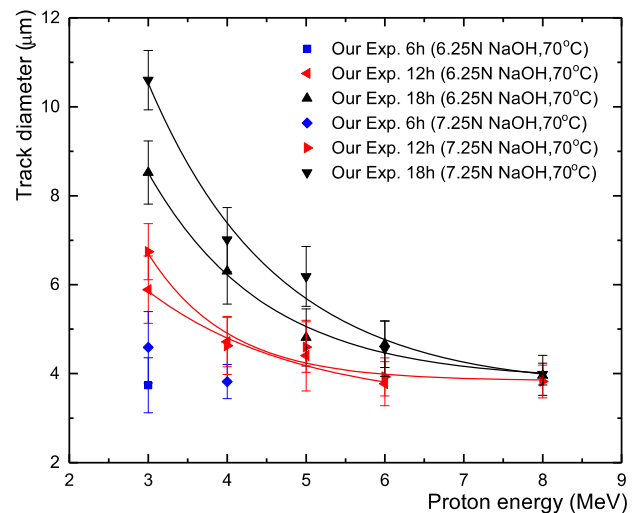


Fig. 7 Proton track diameter versus energy in 6.25 mol/l NaOH at 70 °C

with various etchant temperatures (Fig. 8). Baccou (2015) [6] etched CR-39 manufactured by TASL for 6 h in a 6 mol/l NaOH solution at a temperature of 70 °C. The CR-39 used by Xiaojiao Duan (2009) was produced by Fukuvi Chemical Industry, Tokyo, Japan [18]. By comparing the experiment results of Baccou with those of Xiaojiao Duan, it was found that the track diameter of Baccou's experiment was significantly larger than that of Xiaojiao Duan's results with a proton energy from 0.2 to 1 MeV at an etching temperature of 70 °C. The track diameter of our results is consistent with the results of Baccou. Based on comparing the above results, it is concluded that the CR-39 of different manufacturers respond differently to proton tracks. To compare the results for the proton track diameter

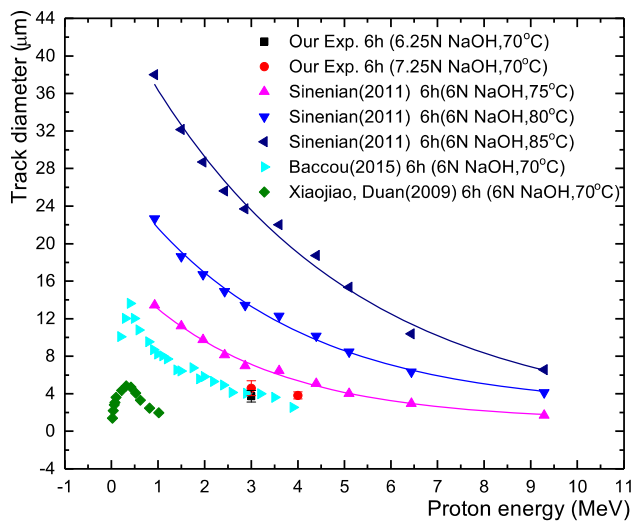


Fig. 8 Proton track diameter versus energy for different CR-39 manufacturers

at different temperatures, we added the results of Sinenian (2011), whose CR-39 was produced by TASL and etched for 6 h in a 6 mol/l NaOH solution at temperatures of 75 °C, 80 °C, and 85 °C [16]. As shown in Fig. 8, the variation in the track diameters for low-energy protons is larger than that for high-energy protons as the etching temperature increases.

3.3 Track diameters for alpha particles in 6.25 mol/l NaOH at 98 °C and 70 °C

As mentioned in the introduction, the V_T/V_B ratio peaks at concentrations of approximately 6.25 mol/l NaOH at a temperature over 60 °C, and other similar studies have also tended to employ a molar concentration of 6 or 6.25. Sadowski (1997) [24] and Baccou (2015) [6] published alpha-track results that are comparable to ours. The CR-39 used for Sadowski's experiment was produced by Pershore Mouldings Ltd., unlike for Baccou's experiment and ours.

Figure 9 illustrates the track diameter after etching for 2 h and 4 h in the two NaOH solutions with a small concentration difference, at an etching temperature of 70 °C. After etching for 2 h, the track diameters of the three experiments are almost the same size. After etching for 4 h, our and Sadowski's results show almost the same size, but the track diameters reported by Baccou are significantly larger than those of our and Sadowski's results. Baccou's CR-39 was manufactured by the same manufacturer as ours, whereas Sadowski's CR-39 was from a different manufacturer. However, Sadowski's results are almost consistent with ours, whereas Baccou's are very different, and so the difference in results must be due to differences in the detector storage history and age, and differences in the production series.

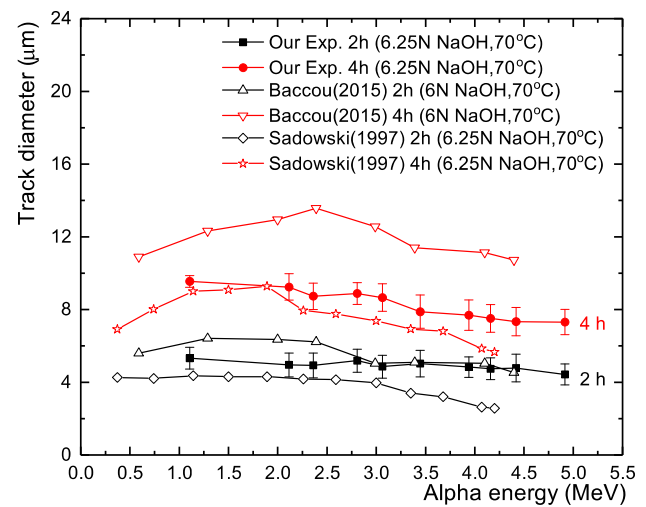


Fig. 9 Alpha-particle track diameter versus energy in 6.25 mol/l and 6 mol/l NaOH at 70 °C

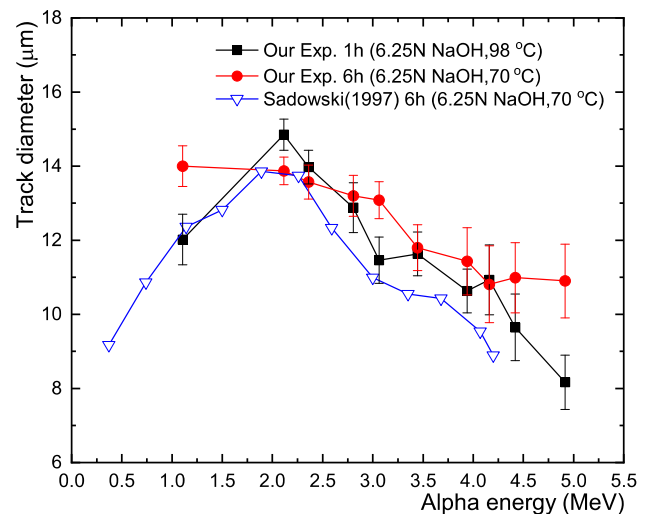


Fig. 10 Alpha-particle track diameter versus energy in 6.25 mol/l NaOH at 98 °C and 70 °C

As shown in Fig. 10, comparing the alpha-particle tracks of our results etched for 1 h in 6.25 mol/l NaOH at 98 °C with those of Sadowski's experiment etched for 6 h in 6.25 mol/l NaOH at 70 °C, the alpha-particle track diameters are maximized at an energy of approximately 2 MeV, and the trend of the track diameters with increasing energy is also consistent. In addition, the alpha-particle track diameters vary little with energy when etched for 6 h in 6.25 mol/l NaOH at 70 °C in our experiment. Therefore, if the track diameter is between 10 and 15 microns, then the alpha-particle energy may be less than or over 2 MeV. Thus, it is very difficult to derive the alpha-particle energy at two etching temperatures by considering the track diameter only. Comparing the track diameters etched for

1 h at 98 °C and for 6 h at 70 °C, they are observed to be almost the same, and hence etching alpha-particle tracks at 98 °C can significantly reduce the etching time. Therefore, 98 °C is better for etching alpha particles than 70 °C from the viewpoint of saving etching time.

3.4 Track diameter from carbon ions in 6.25 mol/l NaOH at 98 °C and 70 °C

Carbon-ion tracks in the energy range of 10–30 MeV were also investigated, and Baccou (2015) [6] studied C^{3+} ion tracks in the energy range of 0.6–4 MeV. As shown in Fig. 11a, the C^{3+} ion track diameters increase almost linearly with an increasing energy after etching for 6 h in 6 mol/l NaOH at 70 °C. As shown in Fig. 11b, the C^{3+} ion track diameter has almost no correlation with energy in the range of 10 to 20 MeV after etching for 6 h in 6.25 mol/l NaOH at 70 °C and for 1 h in 6.25 mol/l NaOH at 98 °C. Furthermore, we found that the track diameters of C^{4+} ions at 25 MeV and C^{5+} ions at 30 MeV have the same size as those of C^{3+} ions at 10 to 20 MeV. Thus, it is possible to infer the energy of C^{3+} ions from the track diameter in the energy range of 0.6 to 4 MeV. However, the C^{3+} ion energy cannot be inferred from the track diameter from 10 to 20 MeV. Furthermore, the C^{3+} , C^{4+} , and C^{5+} ion track diameters after etching for 1 h at 98 °C are approximately the same as those etched for 6 h at 70 °C, and so etching carbon-ion tracks at 98 °C can significantly reduce the etching time. Therefore, 98 °C is also better than 70 °C for etching carbon-ion tracks.

3.5 Comparison of track diameters

In laser–plasma experiments, CR-39 detectors are utilized to distinguish charged particles, which are generally mixed together. Comparing our measured data with other experimental data from the literature, we find that alpha particles, C^{3+} ions, and protons can be identified by controlling the etching time.

According to Fig. 12, the alpha-particle and C^{3+} ion track diameters are greater than 8 μm after etching for 1 h in 6.25 mol/l NaOH at 98 °C. However, the proton tracks with energies greater than 3 MeV are not revealed after etching for 1 h. Figure 13 illustrates the results for alpha-

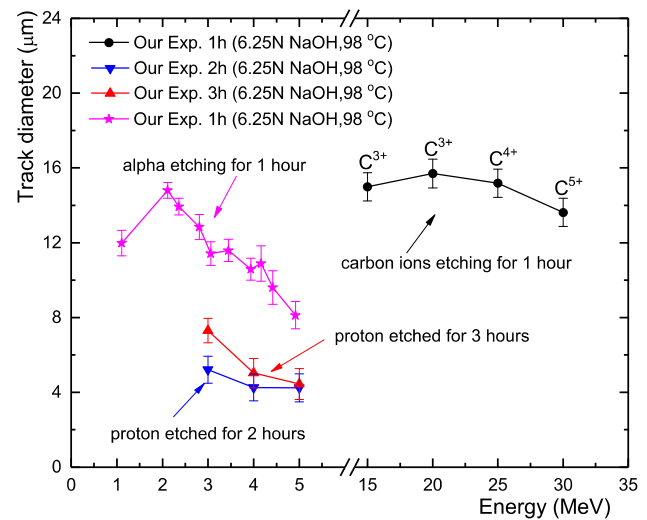


Fig. 12 Track diameter versus energy for carbon ions, alpha particles, and protons in 6.25 mol/l NaOH at 98 °C

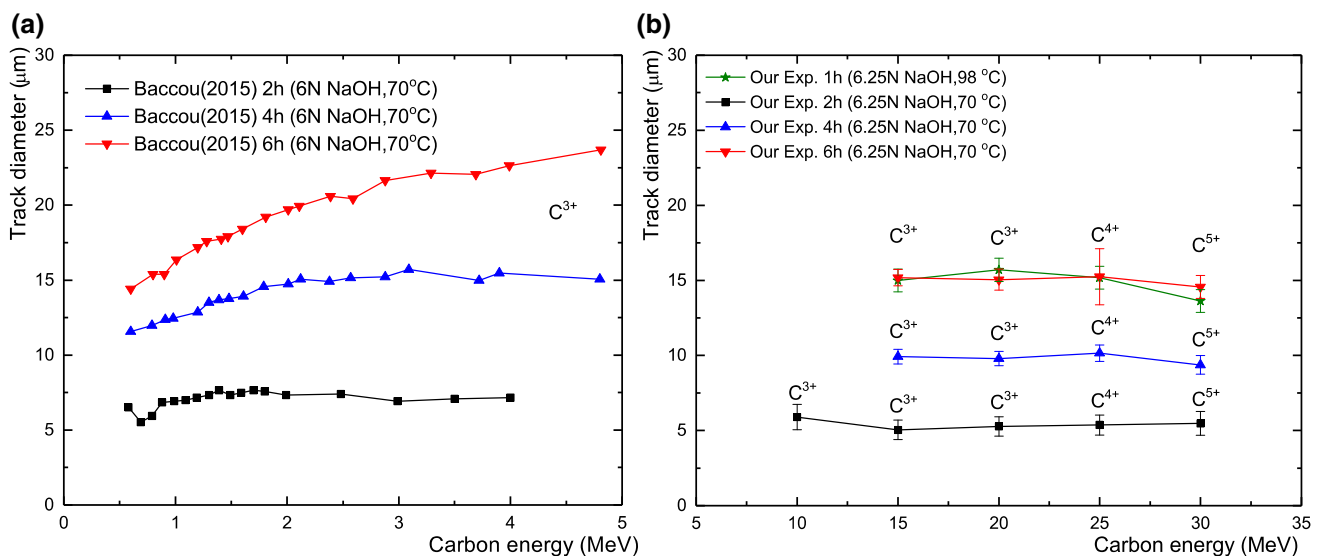


Fig. 11 Carbon-ion track diameter versus energy

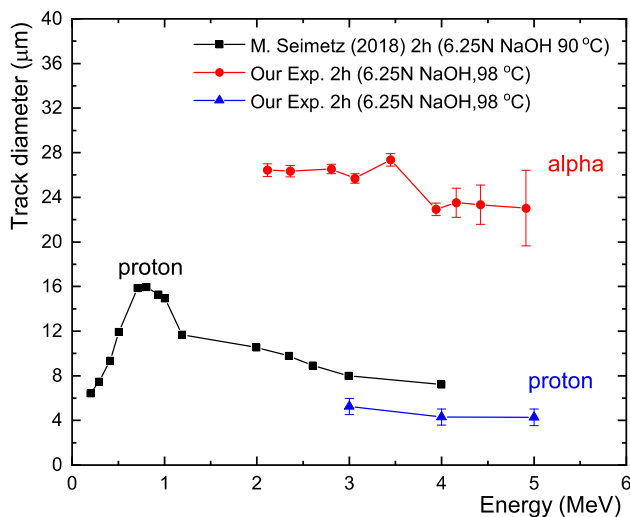


Fig. 13 Track diameter versus energy for alpha particles and protons etched for 2 h in 6.25 mol/l NaOH at high temperature

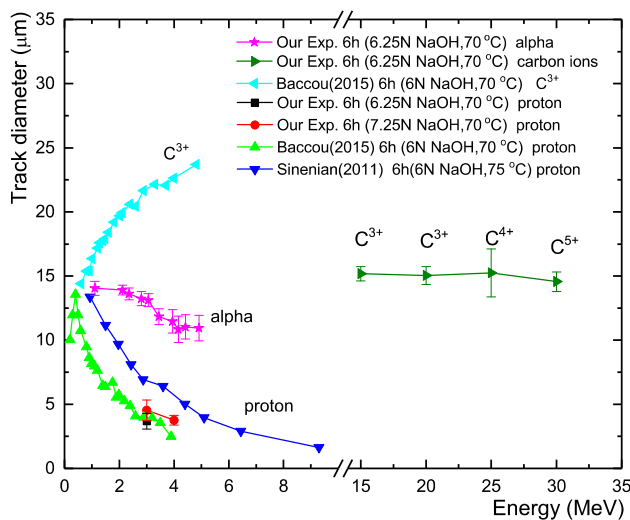


Fig. 14 Track diameter versus energy for carbon ions, alpha particles, and protons in 6.25 mol/l NaOH at 70 °C

particle and proton tracks after etching for 2 h at high etching temperature over 90 °C. Seimetz [5] etched proton tracks for 2 h in 6.25 mol/l NaOH solution at 90 °C. Comparing with our results etched at 98 °C, the track diameter of alpha particles is significantly larger than for proton tracks. Thus, if we need to distinguish alpha-particle tracks from proton tracks with energies over 3 MeV while etching in 6.25 mol/l NaOH solutions at 98 °C, we should first etch for 1 h and mark the alpha-particle tracks and then continue to etch the sample for another hour to reveal the proton tracks. Using this two-stage etching process, alpha particles, C^{3+} ions, and protons with energies over 3 MeV can be distinguished by controlling the etching time, but alpha-particle and C^{3+} ion tracks are

indistinguishable based on the track diameter, because their track diameters are almost the same.

Figure 14 shows the diameters of charged particle tracks after etching for 6 h in 6.25 mol/l NaOH at 70 °C. The track diameters of the three types of particle were almost the same near an energy of 1 MeV, and so it is difficult to distinguish low-energy proton tracks from alpha-particle and C^{3+} ion tracks. However, the proton track diameters at energies over 3 MeV are very small, so we can only distinguish proton tracks of energies over 3 MeV from alpha-particle or C^{3+} ion tracks based on the track diameter.

4 Conclusion

The goals of this study were twofold: calibrating a CR-39 detector to the energies of protons, carbon ions, and alpha particles under two etching conditions; testing the bulk etch rate in 6.25 mol/l NaOH at 98 °C.

The mean of the bulk etch rate in 6.25 mol/l NaOH at 98 °C is 9.95 $\mu\text{m}/\text{h}$, which is significantly larger than at 70 °C. For this reason, an etching temperature of 98 °C can reduce the etching time. The bulk etch rate increases exponentially with the temperature.

The proton track diameter is inversely related to energy between 3 and 8 MeV. Because an etching temperature of 70 °C can reveal high-energy proton tracks, the etching temperature of 70 °C is more suitable than 98 °C for etching proton tracks. At an etching temperature of 98 °C, the maximum alpha-particle track diameter occurs with an energy of approximately 2 MeV, and it is very difficult to derive the alpha-particle energy from the track diameter only. C^{3+} ion track diameters are almost independent of the incident energy in the range from 10 to 20 MeV. When comparing the track diameter and etching time, an etching temperature of 98 °C can significantly reduce the etching time for alpha particles and carbon ions. C^{3+} ion or alpha-particle tracks can be distinguished from proton tracks by controlling the etching time.

These data on CR-39 detectors will be useful for identifying reaction products in laser-plasma experiments with complex conditions. For instance, our results can be used to formulate models that researchers will need to differentiate between 3.02 MeV DD-protons, 14.7 MeV $D^3\text{He}$ -protons, and alpha particles that are produced from laser-induced proton-boron reactions.

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