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Inter-comparison of geometrical track parameters and depth dependent track etch rates measured for Li-7 ions in two types of CR-39

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Abstract

The depth dependent track etch rates of two types of CR-39, TASTRAK and BARYOTRAK, for Li-7 ions with incident energies of 4.82, 6.75 and 10.77 MeV have been evaluated from track length measurements. The pit lengths versus the etching time obtained by three different laboratories have the same trend, but did not agree completely. These differences can be attributed to an error margin of less than one decimal point in the etching temperature. Significant deviations were also present for the assessed track etch rates. Most of the deviation was attributed to the derivation step of the growth curves. A sufficient amount of data and reasonably smooth growth curves were required near the Bragg peak in order to make a precise estimation.

Keywords: CR-39, Li-7, Track length, Depth-dependent response

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1. Introduction

During the past several years, we have been working on the development of reliable methods to obtain the depth-dependent response of the CR-39 detector for light ions using different approaches. The group from the Universite de Franche-Comte (UFC) has created a comprehensive computer code to simulate the track's evolution under chemical etching, using an adjusted function for the track etch rate (Fromm et al., 1991, 1993, 1996; Membrey et al., 1993; Meyer et al., 1997, 1998). Such functions have a number of adjustable free parameters. Here, these parameters were determined so that the simulation matched the experimental values of track diameters. On the other hand, the group from the Technische Universitaet Dresden (TUD) has developed a parameter-free method based on the track length measurements versus the etching time (Doerschel et al., 1999, 1998, 1997a, 1997b, 1996; Hermsdorf et al., 1999). This can be called the direct V_t measurement. At the 19th ICNTS held in Besançon, in August 1998, the responses obtained by these two methods were compared for alpha-particles. The UFC method gave lower track etch rate values around the Bragg peak than the TUD method (Brun et al., 1999). We concluded that the direct, no free parameter TUD method appears to be more accurate.

The group from the Kobe University of Mercantile Marine (KUMM) has worked from a slightly different point of view, based on the measurements of both the track radius and the thickness of layer removed without the assumption of the constant track etch rate (Oda et al., 1992, 1993). For the latter, we discovered that under certain conditions of incident energy and angle, the response near the Bragg peak doesn't reflect on the pit radius growth curves in principle, «missing track effect» (Yamauchi et al., 1995a, 1995b). This effect is caused by the characteristics of the pit's geometry. In other words, significantly different responses around the Bragg peak could generate a similar etch pit radius at the same etching time. The previously mentioned discordance between the UFC and TUD methods on the alpha-response most likely corresponds to this effect. The UFC method could be improved by adjusting certain parameters such as the track length or other suitable parameters, while leaving the pit diameters as they are. For the KUMM method, the difficulties brought by the missing track effect was could be to overcome, in principle, by choosing the incident energy to avoid the effect (Yamauchi et al., 1997, 1999). Namely, for the response estimation around the Bragg peak, the growth curves with low incident energy is available. In addition to these three methods, the shape of the pit profile can also provide perfect information on the response when the front of the etch pit cone slightly passes the track's end point (Al-Najjar et al., 1984; Yamauchi et al., 1997). In this study we apply the TUD method, also known as the direct method.

Therefore, in this study, the response for Li-7 ions has been evaluated from length measurements independently in the three groups. The selected irradiation and chemical etching conditions were the same, but the results of pit lengths, L, versus the etching time, t, were different from each other. In the TUD method, it is inevitable to derive the growth curves of track length, $L(t)$, with respect to t. The derivation of $L(t)$ was also found to be a critical step in the response estimation. Through the inter-comparison among the results from three teams, we have extracted the practical problem in applying the TUD method for Li-7 ions.

2. Experimentals

Two types of CR-39 detectors, TASTRAK (Track Analysis Systems Ltd., UK) and BARYOTRAK (Fukuvi Chemical Ltd., Japan), were used as samples with dimensions of 1.0 x 1.0 x 0.1 cm^3. Irradiations with Li-7 ions were carried out using the tandem accelerator at the Rossendorf Research Center, Germany. A reduced ion beam in flux was obtained by using a Rutherford forward scattering target of a thin Ta foil with a thickness of 1.1 μm, which was bombarded on the sample perpendicularly. The fluence was adjusted to the desired amount ranging from 2.0×10^4 to 4.5×10^4 cm^{-2}. Three different irradiation series with energies at the sample surface of $W = 4.82 \pm 0.16$, 6.75 ± 0.17 and 10.77 ± 0.11 MeV were made (with one exception of TASTRAK for TUD: $W = 10.96$ MeV). The values of energies and their spread were attained using Si(Li) drifted-detector. As calculated using the program STOPOW, the corresponding Li-7 ion ranges in the samples were $R = 13.22 \pm 0.85$, 19.15 ± 1.14 and 34.88 ± 0.91 μm, respectively (Doerschel et al., 2000). More than 60 pieces of irradiated samples were used for each energy and type of CR-39. The TUD group divided the samples into three batches and supplied them to the other teams.

The subsequent etching and track size measurements were made, independently by the three teams. Chemical etching was performed in a stirred 7.25 N NaOH solution kept at 70 °C. To measure the track lengths the etched detectors were broken into several pieces perpendicular to the surface. Since the material has an excellent optical transparency, tracks that were situated some micrometers below the edge's surface were visible under the transmission optical microscope. This makes it possible to evaluate the etch-pit longitudinal sections (Doerschel et al., 1996, 1997). In that way, the track lengths could be determined as a function of the etching time. The values of V_b obtained by the fission fragment method at the UFC, TUD and KUMM were $V_b = 1.95$, 1.73 and 1.80 μm/h for TASTRAK, and $V_b = 1.79$, 1.64 and 1.70 μm/h for BARYOTRAK, respectively. This indicates that the real chemical etching conditions in our respective laboratories were slightly different from each other. In general TASTRAK had a higher bulk etch rate than BARYOTRAK. The individual values of V_b were used in the following analysis.

3. Results and Discussions

3.1. Pit-length versus etching time

In Fig. 1, two examples of Li-7 etch pit profiles are shown. There is a bend on the right side, probably caused by the nuclear collision between a Li-7 ion and an atom belonging to the detector material (C or O). This is a very rare case and most of the etch pits are straight as that of on the left side. As shown in this photograph, the Li-7 etch pits are very sharp compared to those of the alpha-particles or hydrogen isotopes (Fromm et al., 1991; Doerschel et al., 1996; Yamauchi et al., 1997).

Figures 2, 3 and 4 show the pit lengths versus the etching time for both types of CR-39 detectors, (a) TASTRAK and (b) BARYOTRAK, at indicating incident energies. As expected from the basic considerations at every obtained growth curves, the pit lengths growth rates increase gradually reaching a maximum just before the pit length's saturation. The highest growth rates correspond to the Bragg peak. Pits of ions with lower incident energies are larger in the early stages of etching, and they have shorter saturated lengths. The error bars represent the standard deviation of more than 10 measurements. In general they are largest at the steepest parts of the curves just before saturation, as a result of range straggling. In some of the figures, there are plots with smaller symbols. These were obtained by following up the growth of individual pits by repeating the KUMM team's etching and sizing. These pit lengths differ somewhat from the average ones, especially as shown in Figure 3(b).

Although these trends in track length evolution can be found in the data from all three teams, the values do not exactly agree with each other. It is not surprising that some variations exist, since the etching conditions were not equivalent, judging from the individual values of the bulk etch rates. For TASTRAK, the highest bulk etch rate of 1.95 μm/h was attained by the UFC, and the lowest one of 1.73 μm/h was obtained by the TUD. Simple calculations revealed that the differences in the layer's thickness removed between them is 0.22, 0.37 and 0.93 μm at the total etching times of 60, 100 and 250 minutes, respectively. These etching times correspond to the regions where the growth curves became steepest for the adopted incident energies. Furthermore the difference in the track length is several times larger. Such difference in etching condition could produce the discrepancies in the growth curves that were observed. We have checked our etching conditions; for example, the impurities in NaOH agents, the effect of accumulation of poly-allylalcohol, the aging effect by the absorption of CO_2 in the air, the used distilled water and so on. The remaining possibility is that the observed difference in the bulk etch rate was mainly caused by the different etching temperature below the decimal point. The confirmed temperature dependence on V_b showed that the observed difference of V_b between TUD and UFC was possible if the temperature difference was about 1 degree. In the following, we call this as the « temperature disaster ». As long as the bulk etch rate is constant throughout each series of experiments, however, the « temperature disaster » generates few significant track etch rate errors. Because the main difference of

$L(t)$ and each V_b could be canceled out in the following analyses.

About the sizing step, it should be pointed out that the pit length measurement is not as easy to make as for the pit diameter. Because the Li-7 track has a very sharp conical angle, special attention was needed to determine the tip position. We used our own optical microscopic systems to observe the track and its different properties. It is also probable that the three teams used different criteria to obtain the pit length. We call the dependencies on the microscopic systems and operators the « sizing disaster ». Round robin measurements on the same samples would be useful in checking this disaster.

To finish this subsection, the growth curves should be examined from the viewpoints of the sizing and temperature disasters mentioned above. At first it is noteworthy that three series of pit lengths for TASTRAK at 6.75 MeV had quite good agreement (Fig. 3(a)), despite the different V_b. On the BARYOTRAK in Figure 3(b), the data, with the exception of those that were followed up, also agree with each other. The temperature disaster should not seriously influence the results in these shorter etching times below about 100 minutes, which was as expected.

In Fig. 4, with an incident energy of W = 10.77 MeV, the deviation among the three series of data is very significant in the steep regions. At the same etching time, the groups with the larger bulk etch rate obtained the larger pit length. This is concordant with the effects of the temperature disaster. There is an additional factor in Figure 4(a), that the incident energy of TUD data was W = 10.96 MeV, which has a longer range than that of W = 10.77 MeV by about 0.83 μm. Indeed, the TUD saturated track length is slightly larger than those of UFC and KUMM. When only considering the temperature disaster, it is difficult to understand the data differences in Fig. 2. It should be mentioned that the UFC measurements shown in this figure were made using the TUD microscopic system. The values of UFC and TUD are almost the same. The KUMM data were greater for the TASTRAK and smaller for the BARYOTRAK than those of the other two groups. These deviations should be caused by the sizing disaster.

3.2. Track etch rate vs. depth

The track etch rate, $V_t(x)$, at a depth x, along the ion trajectory was obtained using the following relations:

$$V_t(t) = dL(t)/dt + V_b, \qquad (1)$$

$$x = L(t) + V_b t, \qquad (2)$$

where V_b is the bulk etch rate and $V_b t$ is equivalent to the thickness of the layer removed, G. The derivation of *L(t)* expressed in equation (1) was graphically made by each team using their own data shown in Figures 2, 3, and 4 (Doerschel et al., 1996). We obtained the track etch rate, which is represented in Figures 5, 6 and 7 for the incident energies of 4.82, 6.75 and 10.77 MeV, respectively.

It was difficult to obtain concordant results of the track etch rates among laboratories. However, well-matched results can be found in Figure 6(a), between the three teams (TASTRAK at W = 6.75 MeV). It is noteworthy that the measured *L(t)* are also very consistent with each other, as shown in Figure 3(a). The UFC's and TUD's BARYOTRAK were also in strong accordance at W = 6.75 MeV (Fig. 6(b)). The KUMM results for BARYOTRAK, however, are lower and the peaks' width is narrower than for the others. It should be mentioned that the KUMM team's BARYOTRAK analyzed data were not averaged but represent the intermediate values of the followed up data. As indicated in Figure 3(b), the followed up data are lower than the averaged data.

As shown in Figure 4, the UFC's and TUD's measured *L(t)* at W = 10.77 MeV was quite different from each other. The analytical results from the track etch rates, however, were in good agreement (Figure 7). This implies that the « temperature disaster » does not have much of an effect on bringing out the significant error in the final results. On the other hand, the UFC and TUD obtained the almost equivalent *L(t)* at W = 4.82 MeV as indicated in Figure 2. However, the track etch rates at this energy are not in strong accordance (Figure 5). This indicates that this disagreement was produced in the following analytical step. It probably occurred during the derivation step.

For the response estimation around the Bragg peak, the amount of data related to the peak is important. If the track etch rate were 22 μm/h around the peak, the pit-tip position would progress 5.5, 3.7 and 1.8 μm during the etching time intervals of TI = 15, 10 and 5 minutes, respectively. From the viewpoint of discrete mathematics, it is apparent that we need a sufficient amount of data to obtain a reasonably smooth growth curve for the derivation using the discrete data. For the precise estimation, namely, an appropriate number of points are required around the Bragg peak, because it would be difficult to detect a rapid change in track etch rate using longer etching time intervals.

3.3. Follow up measurements

We do not know whether TI = 5 minutes is short enough to obtain the precise response. Even if this is a sufficient value, we are soon faced with the other problem that the measured lengths have comparable standard deviations to the distance of 1.8 μm. The calculated range straggling is also in the same magnitude. By adopting the averaged pit length, the pit length's growth rate, *dL(t)/dt*, can be underestimated. In order to examine this, the track etch rates and average rates obtained from the follow up growth curves are shown in Figure 8. As shown in this figure, one of the followed up curves (F-1) gives a larger and sharper peak for the track etch rate at a greater depth than the averaged track etch rate. The other followed up response (F-2) has the same peak value of $V_t(x)$, but the position is somewhat deeper. On the other hand, the track etch rate at a depth below about 8 μm are in strong agreement. Since the data examples are limited, more studies should be conducted on this in the future. The effect of repeating the etching and sizing on the pit growth behavior around the Bragg peak should be checked in detail.

4. Conclusions

The track etch rates of two types of CR-39, TASTRAK and BARYOTRAK, for Li-7 ions for incident energies of 4.82, 6.75 and 10.77 MeV have been evaluated versus depth directly from track length measurements. The selected irradiation and chemical etching conditions were the same (7.25 N NaOH, 70 °C), but the pit lengths versus the etching time obtained by the three teams do not agree. The causes in deviation were discussed from the viewpoints of the etching « temperature disaster » as well as the « sizing disasters ». Even if the difference in the temperature was less than a decimal point, it could cause significant differences in the shape of the growth curves after long etching times of more than about 200 minutes. The subsequent derivation was also found to produce deviations in the track etch rates. For the precise estimation of the depth dependent response, a higher amount of data was required around the Bragg peak region to characterize the rapid changes that occurred in the track etch rate. Although the shortest etching time interval in this study was five minutes, we were unable to conclude that this time was short enough to evaluate the Li ion response. We do know, however, that a shorter interval would be needed to assess the response of heavier ions. Comparisons should also be made between the averaged data points and the follow up method to draw the growth curves.

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Figure Captions

Fig. 1. Side view of Li-7 etch pits in BARYOTRAK (W: 6.75 MeV, etching time: 120 min).

Fig. 2. Track length against etching time, L(t), for UFC, TUD and KUMM for 4.82 MeV, (a) TASTRAK, (b) BARYOTRAK, F-1 and F-2 were obtained by following up the evolution of individual etch pits by KUMM.

Fig. 3. Track length against etching time, L(t), for UFC, TUD and KUMM for 6.75 MeV, (a) TASTRAK, (b) BARYOTRAK, F-1 and F-2 were obtained by following up the evolution of individual etch pits by KUMM.

Fig. 4. Track length against etching time, L(t), for UFC, TUD and KUMM for 10.77 MeV (a) TASTRAK, (b) BARYOTRAK, F-1 and F-2 were obtained by following up the evolution of individual etch pits by KUMM.

Fig. 5. Track etch rate against the depth, $V_t(x)$, for UFC, TUD and KUMM for 4.82 MeV, (a) TASTRAK, (b) BARYOTRAK.

Fig. 6. Track etch rate against the depth, $V_t(x)$, for UFC, TUD and KUMM for 6.75 MeV, (a) TASTRAK, (b) BARYOTRAK.

Fig. 7. Track etch rate against the depth, $V_t(x)$, for UFC, TUD and KUMM for 10.77 MeV, (a) TASTRAK, (b) BARYOTRAK.

Fig. 8. Track etch rate against the depth: Comparison between the averaged and the followed up data by KUMM, (BARYOTRAK: W = 4.82 MeV).

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by TUD, UFC, KUMM

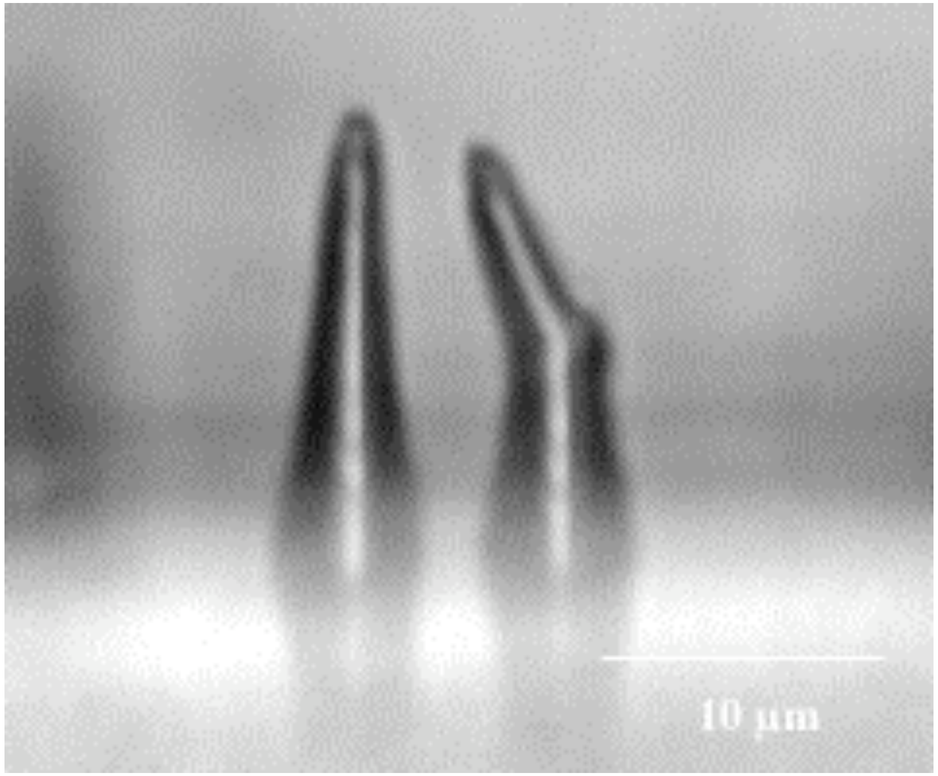


Fig. 1 #107

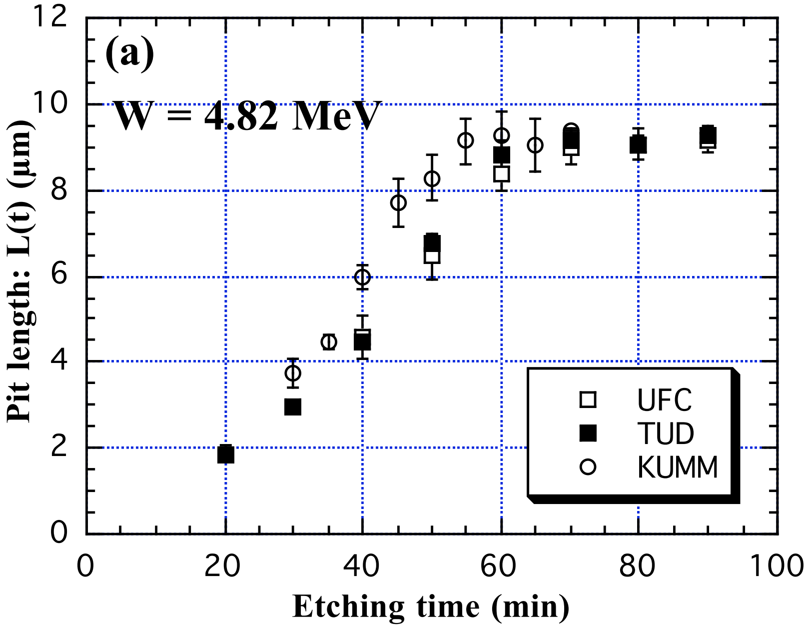
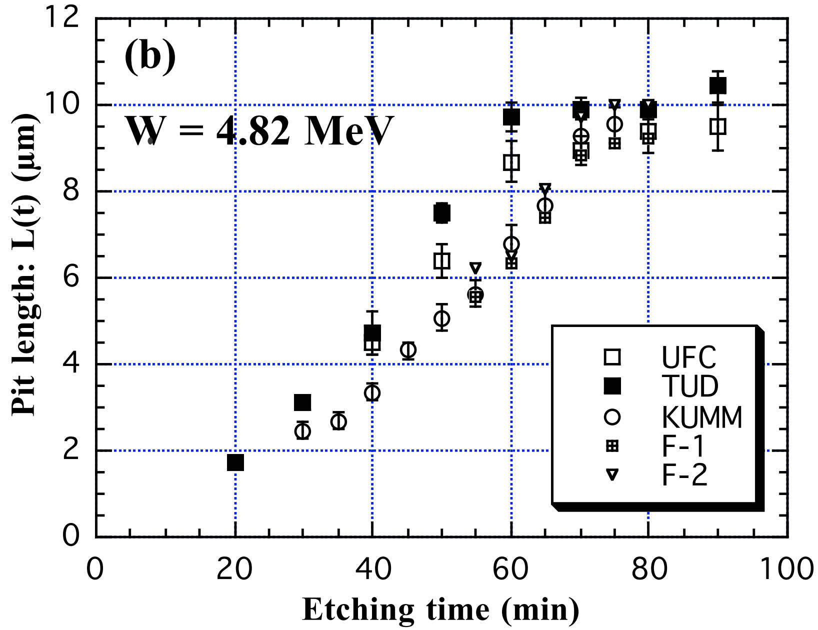


Fig. 2(a)#107



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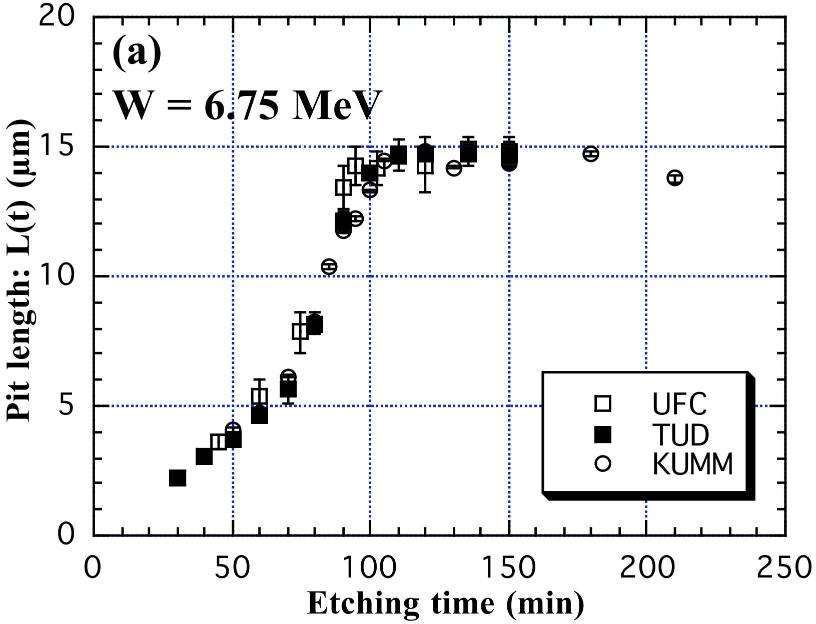

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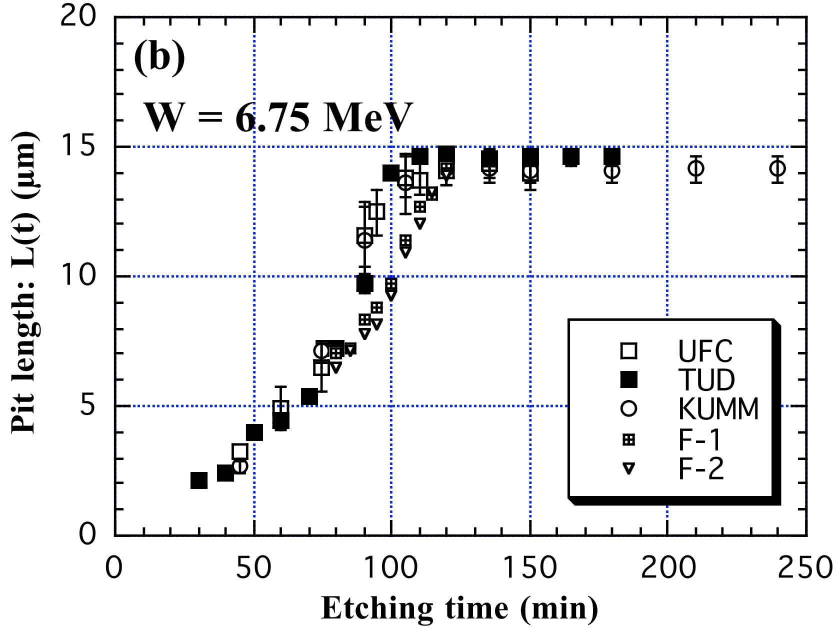

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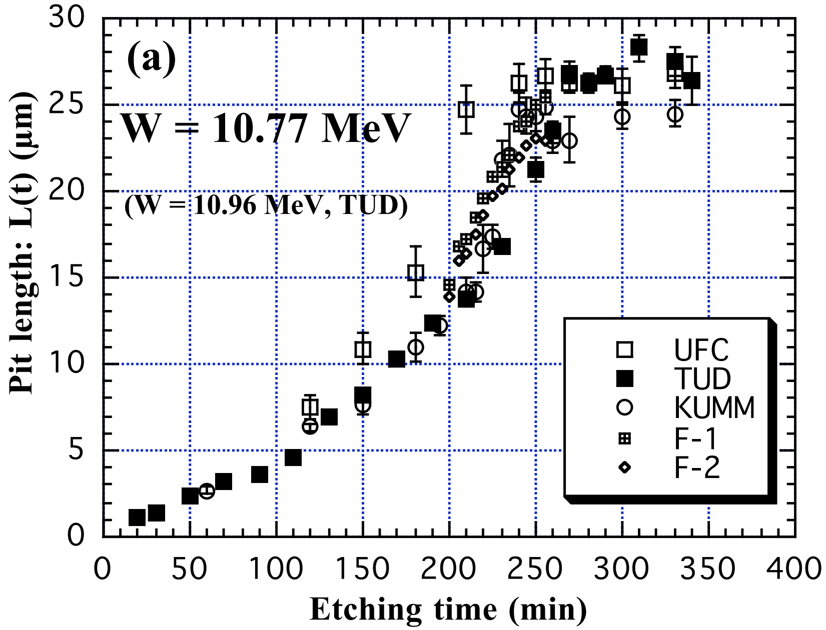

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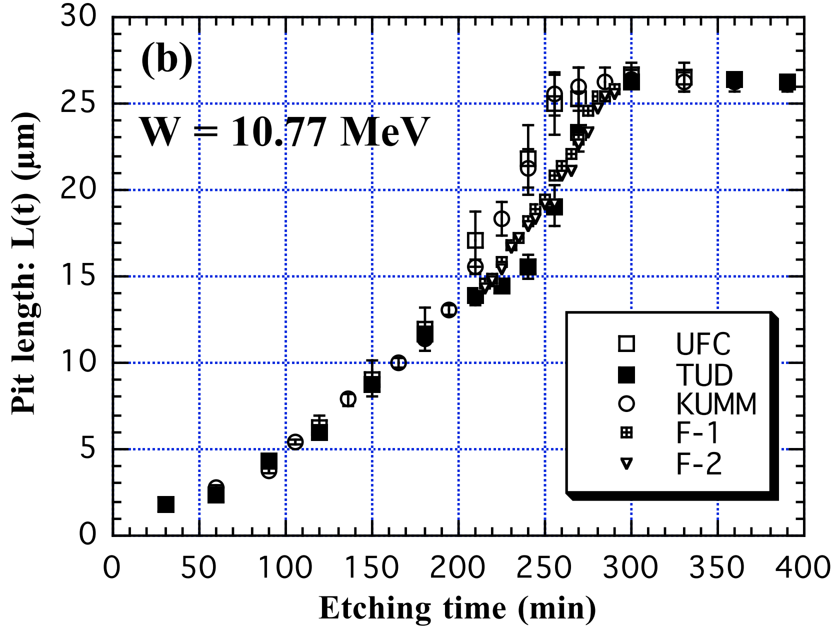

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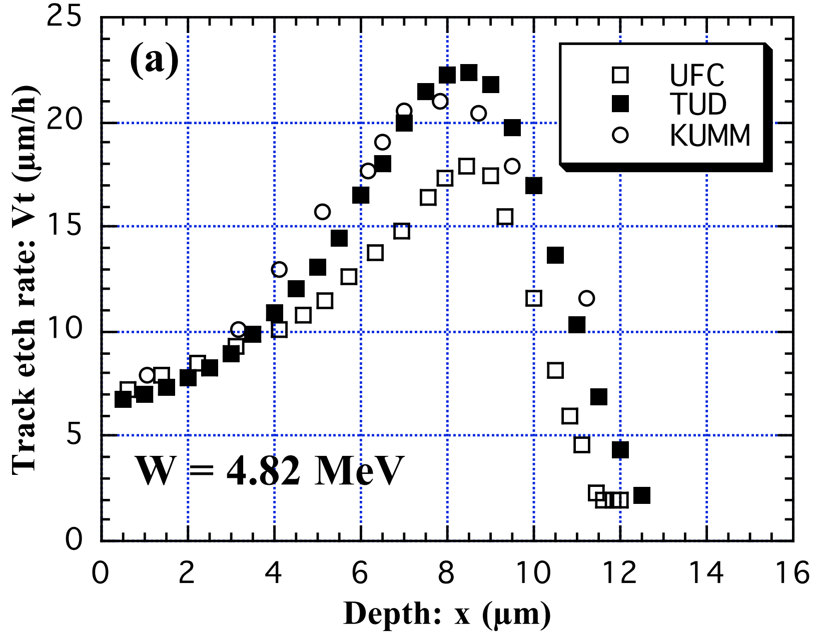
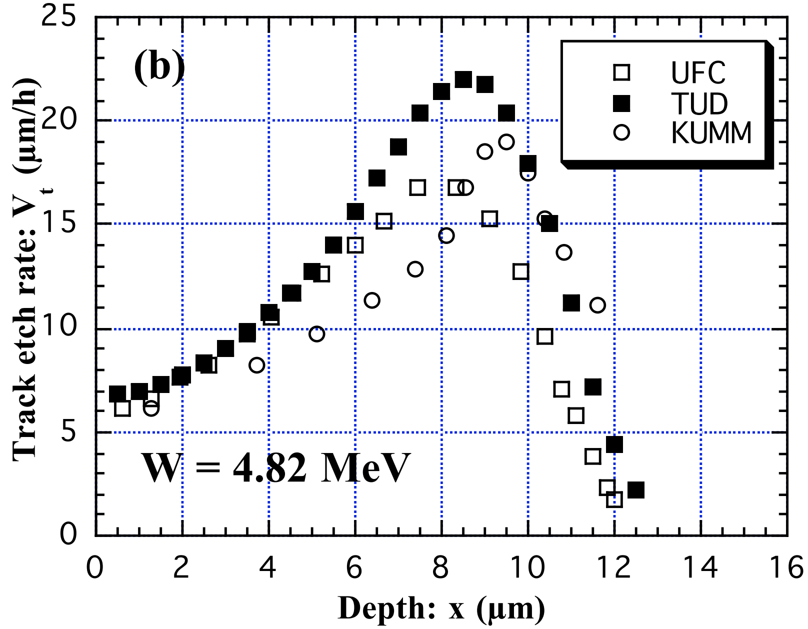


Fig. 5(a) #107



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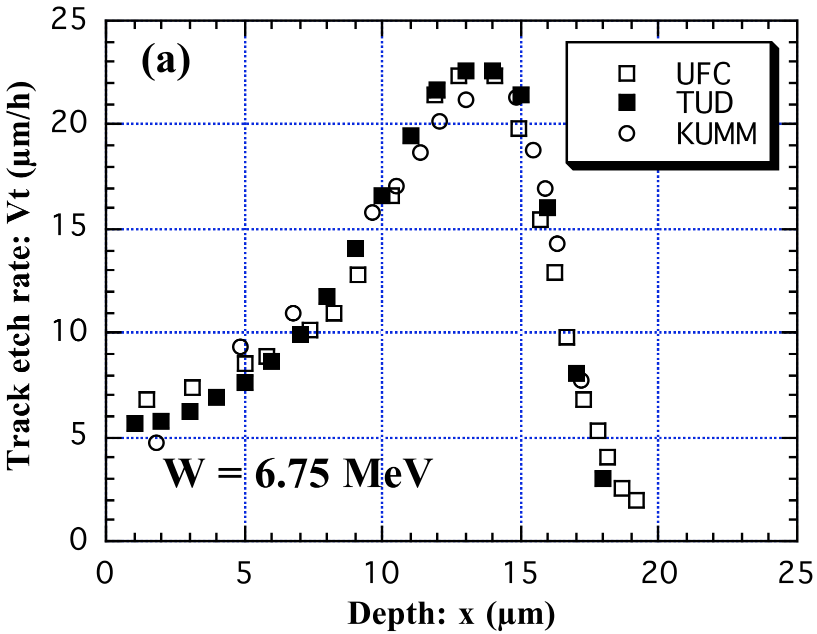
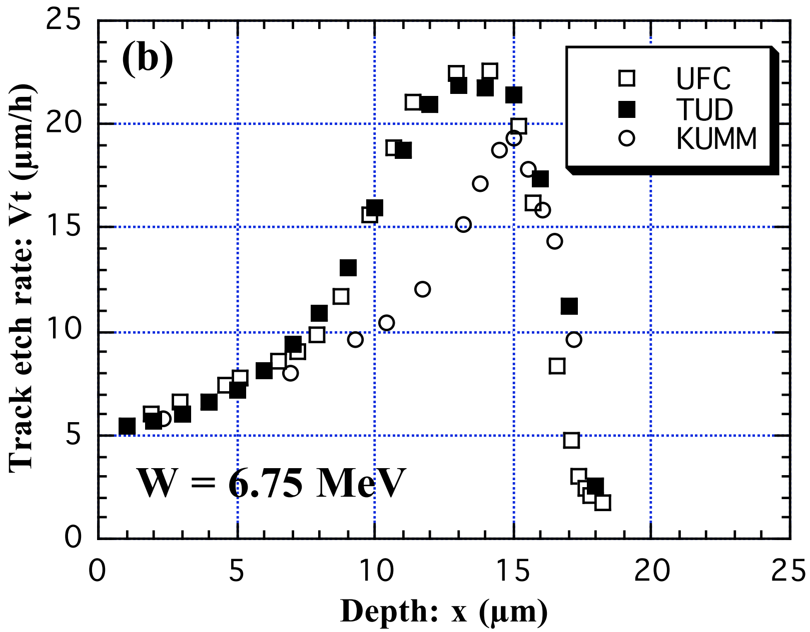


Fig. 6(a) #107



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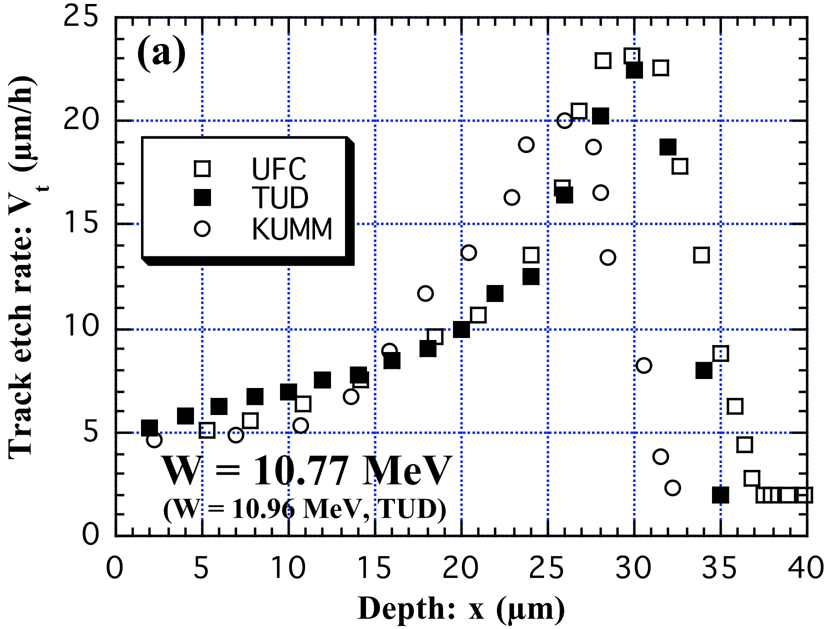
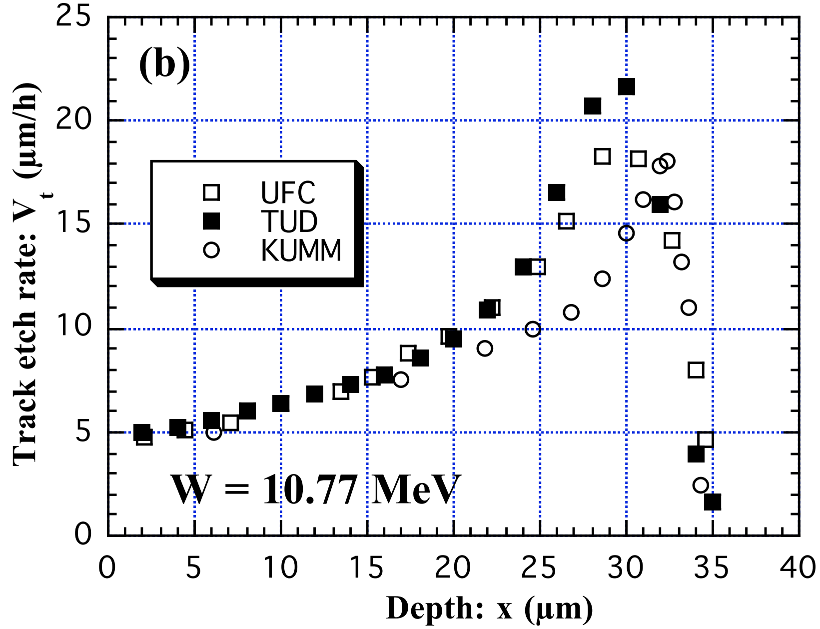


Fig. 7(a) #107



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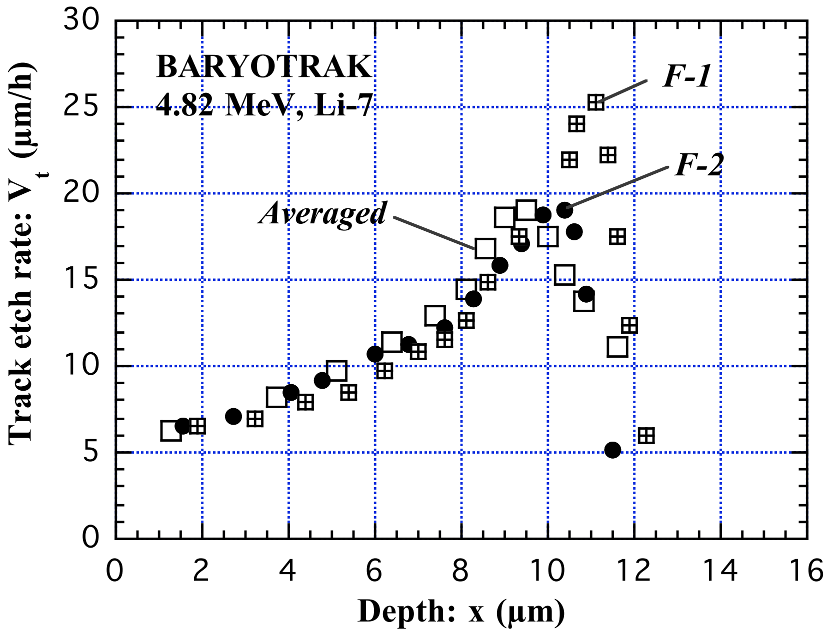
BARYOTRAK
4.82 MeV, Li-7
F-1
F-2
Averaged
Track etch rate: V_t (μm/h)
Depth: x (μm)
0
5
10
15
20
25
30
0
2
4
6
8
10
12
14
16

Fig. 8 #107