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Note

Study of Chemical Etching Conditions for Alpha-particle Detection and Visualization Using Solid State Nuclear Track Detectors

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We evaluate radon/thoron and its progeny concentration using passive-type monitors using CR-39 plates. After exposure, it is necessary to do chemical etching for CR-39 plates. In the present study, we considered shortening of chemical etching time for CR-39 and enlargement of the track diameter (i.e. etch pit diameter) aiming for introduction of automatic counting system in the future. Optimum conditions were determined by changing solution concentration, solution temperature and etching time. As a result, the optimized conditions (concentration, temperature and etching time) were determined to be 8 M NaOH solution, 75℃ and 10 hours. This result of etching time showed that the chemical etching was completed in less than half of conventional etching time. Furthermore, it was suggested that shorter etching time would be possible if we do not consider the enlargement of conventional track diameter.

Key words: solid state nuclear track detector, chemical etching, condition optimization, etching time, solution concentration/temperature, track diameter

1. Introduction

The character of solid-state nuclear track detector (SSNTD) was first discovered by Young[1)] in 1958. Young reported that tracking of fissile pieces in lithium fluoride crystals was enlarged by etching in glacial acetic acid hydrofluoric acid mixture and can be observed with an optical microscope. In 1962, Price and Walker clearly indicated that corrosion proceeded in a silicate mineral by chemical etching[2)]. After this report, enlargement of track in the SSNTD needs chemical etching. At the present time, the mainstream of the SSNTD is CR-39 which is a thermoset resin and is made by polymerization of diethylene glycol bis allyl carbonate. Although CR-39 is used as a lens material, it is also used as the SSNTD after Cartwright *et al.* reported that CR-39 plastics had a high sensitivity for charged particles and an excellent charge resolution, and easy to observe track (i.e. etch pit) compared to conventional plastics[3)]. CR-39 is used as the SSNTD in various fields such as measurement of radon/thoron concentration[4, 5)], measurement of cosmic ray[6)] and

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Table 1. Etching conditions in various research institutes

Country	Institute	Solution	Condition			Reference/Remarks
Australia	Australian Radiation Protection and Nuclear Safety Agency	KOH	6 h	6.25 M	70 ℃	Brown *et al.*, 2011[13]
Egypt	National Institute for Standard	NaOH	6 h	6.25 M	70 ℃	Mansy *et al.*, 2006[14]
Ireland	Environmental Protection Agency	NaOH	1 h	6.25 M	98 ℃	Dowdall *et al.*, 2016[15]
Italy	ENEA Radiation Protection Institute	NaOH	6 h	6.25 M	70 ℃	Calamosca *et al.*, 2003[16]
Japan	National Institute of Radiological Sciences	NaOH	6 h	6.25 M	90 ℃	Tokonami *et al.*, 2005[8]
Korea	Korea Institute of Nuclear Safety	NaOH	4.5 h	6.25 M	90 ℃	Yoon *et al.*, 2010[17]
Japan	Hirosaki University Institute of Radiation Emergency Medicine	NaOH	24 h	6 M	60 ℃	Our institute

biological cell irradiation[7].

For radon, thoron and thoron progeny measurements, passive radon-thoron discriminative monitors namely RADUET and thoron progeny monitors with their deposition rate measurements are used in our laboratory[8-10]. CR-39 plates (BARYOTRAK, Fukuvi Chemical Industry Co., Ltd., Japan) are installed inside both measuring devices. These CR-39 plates are the square of 10 mm × 10 mm and its thickness are 0.9 mm. After exposure, the CR-39 plates are chemically etched for 24 h in a 6 M NaOH solution (CAS No: 1310-73-2, NACALAI TESQUE, INC., Japan) at 60℃ in a beaker using a constant temperature bath (TM-3, AS ONE Corporation, Japan) and alpha tracks were counted by an optical microscope. Concretely, alpha tracks were photographed by setting a digital camera on the optical microscope, and then each track density was calculated using Image-J[11, 12] which is free image processing software. Generally, NaOH solution or KOH solution is used for etching of CR-39[13-17]. Since Green *et al.* reported that NaOH solution is more sensitive than KOH solution[18], and NaOH solution has been adopted as an etching solution in many research institutes[14-17], we adopted NaOH solution as an etching solution. Furthermore, stirring is not performed during etching. In the etching of CR-39, however, it has been found that magnetic stirring did not affect the bulk etch rate by Ho *et al.*[19]

Our chemical etching time is longer than other research institutes[8, 13-17] (Table 1). Furthermore, the current method is required a long time to read and cannot be analyzed promptly. Since several automatic counting systems of tracks have been developed[20, 21], we also considered their introduction to our evaluation system. When using one of the automatic counting systems, however, a larger track diameter than the current track diameter is required. In a present study, therefore, the following subjects are considered: (1) shortening of chemical etching time aiming for shortening the total analysis time, (2) enlargement of the track diameter aiming for introduction of automatic counting system in the future.

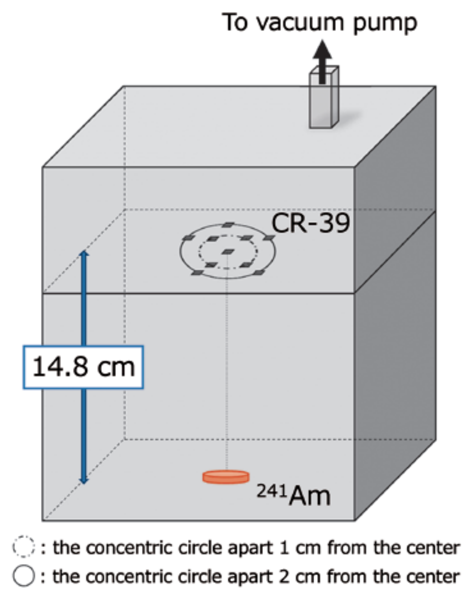

Fig. 1. The arrangement of the experiment for irradiation to CR-39 plates.

2. Material and Methods

2.1. Irradiation of CR-39 plates by an ^{241}Am source

A vacuum condition is set using a vacuum chamber (260 × 180 × 260 mm, 12 L) and a vacuum pump (OSP-90W, Fuji Medical Instruments Co., Ltd., Japan) so as to irradiate alpha particles to CR-39 plates[22]. A radioactive source of ^{241}Am (0.982 kBq, Calibration date: 2015.6.15) is used for irradiation of alpha particles. The irradiation time was adjusted to 40 min. This period was determined so that the track density calculated from an radioactivity and a geometric efficiency of the source was estimated to be 10 tracks mm^{-2}. The arrangement of the experiment for irradiation to CR-39 plates is shown in Figure 1. The arrangements of CR-39 plates are as follows: (1) on the central axis of the ^{241}Am source, (2) on the concentric circle apart 1 cm from the center, (3) on the concentric apart 2 cm from the center.

Table 2. The condition of the temperature and concentration

Run	Concentration (M)	Temperature (°C)	Etching time (h)
1	6	70	24
2	7	70	24
3	8	70	24
4	9	70	24
5	6	75	24
6	7	75	24
7	8	75	24
8	9	75	24

2.2. Relationship between solution concentration and track diameter at two different temperatures

It is well known that shortening of chemical etching time and enlargement of the track diameter can be achieved by raising the temperature and concentration of the NaOH solution[23]. Therefore, the variation of the track diameter is evaluated when the temperature and concentration of the NaOH solution are changed. The condition of the temperature and concentration is shown in Table 2. The highest allowable temperature of a constant temperature bath used in this study is 80℃. Since the temperature of NaOH solution might not be stabilized more than 80℃ with this a constant temperature bath, however, the conditions of temperature are set as 70℃ or 75℃. The number of CR-39 plates is five in each condition. The track diameter of each CR-39 plates are evaluated by measuring the diameter of 10 tracks which is randomly chosen using Image-J after photographing by setting a digital camera on an optical microscope. From the obtained results, the condition with high enlargement ratio of track diameter at the same etching time is determined on the optimized conditions of concentration and temperature.

2.3. Relationship between etching time and track diameter

Under the conditions of optimized concentration and temperature, the track diameter is evaluated when the etching time is varied. The etching time is set to 6, 8, 10, 12 and 24 hours. This etching time is decided to be shorter than the conventional time (i.e. 24 hours) and that of the other institutes[8, 13-17] is also taken into account. The number of CR-39 plates is five in each etching time. In this study, the track diameter required for introducing an automatic counting system is set to 40 μm from the previous study[24], and the etching time which can meet this requirement is taken as an optimized etching time. According to Csordás *et al.*[24], the ideal track diameter when evaluated by a commercially available scanner and a software developed by their research group is 40-60 μm. The same scanner and software used by Csordás *et al.*[24] are checked in our laboratory.

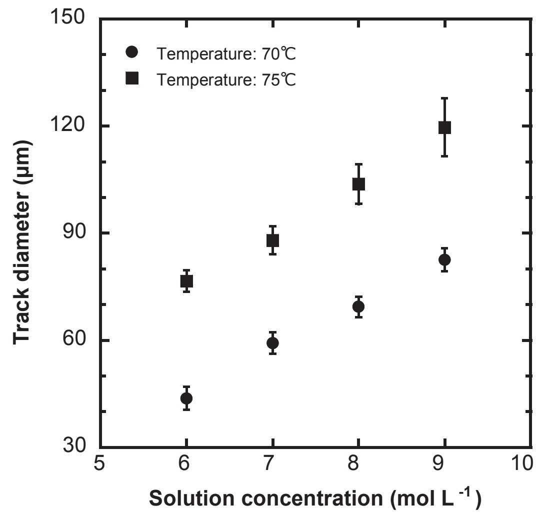

Fig. 2. The observed results of relationship between each condition of chemical etching (temperature and concentration) and etch pit diameter. The error bar means standard deviation.

3. Results and Discussion

3.1. Relationship between solution concentration and track diameter at two different temperatures

The observed results of relationship between each condition of chemical etching (temperature and concentration) and track diameter are shown in Figure 2. It is recognized that the higher the temperature and concentration the higher the enlargement ratio of track diameter. This result is the same as results of previous studies[23, 25]. Therefore, although it is reasonable to adopt the condition of 9 M NaOH solution at 75℃ which had the highest enlargement ratio, this condition was not suitable for measurement because the CR-39 plates under this condition were observed with their rough surface by visual observation. According to the previous study, it is appropriate that the concentration is around 7.5 M NaOH solution after considering an influence of the surface roughness[26]. As a result, the optimized concentration and temperature condition were determined to be 8 M NaOH solution at 75℃. This etching condition seems optimal in order to achieve the purpose using our system. According to Rana and Qureshi[27], however, the best etching condition for fission fragments and similar particles are 6 M NaOH solution at 60℃. It is also reported that the bulk etching rate (i.e. speed of removing surface thickness) of etching with 8 M NaOH solution at 70℃ is about twice as large as that of etching with 6 M NaOH solution at 70℃ by Al-Jubbori[28]. From such reports, it is necessary to investigate the validity of NaOH solution concentration in the future, but the new etching condition with an 8 M NaOH solution at 75℃ is optimal at the present time.

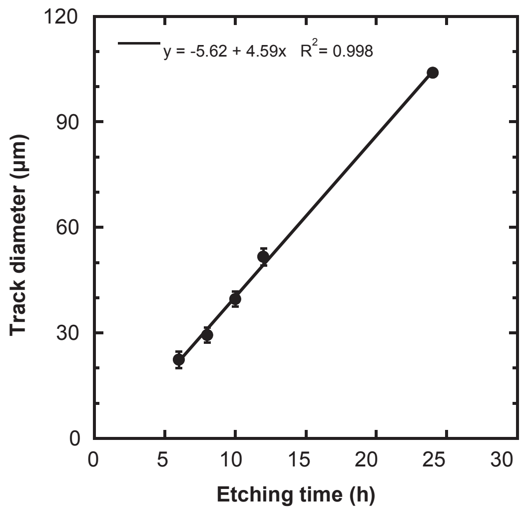

Fig. 3. The observed results of relationship between the etching time and etch pit diameter under 8 M NaOH solution at 75℃. The error bar means standard deviation.

3.2. Relationship between etching time and track diameter

The observed results of relationship between the etching time and etch pit diameter under 8 M NaOH solution at 75℃ is shown in Figure 3. A strong correlation was observed. This relation was similar to that reported by several previous studies[23, 25, 29]. Since the etching time of 10 hours was required to achieve 40 μm for the track diameter, this period was taken as an optimized etching time. In addition, this result suggested that chemical etching could be performed in much shorter time than 10 hours if the size of track diameter is the same as the conventional condition (i.e. 24 h in a 6 M NaOH solution at 60℃).

4. Conclusion

In this study, we considered shortening of chemical etching time for CR-39 and enlargement of the track diameter (i.e. etch pit diameter) aiming for introduction of automatic counting system in the future. As a result, the optimized conditions (concentration, temperature and etching time) were determined to be 8 M NaOH solution, 75℃ and 10 hours. The chemical etching time could be shortened to less than half compared with the conventional this time. Furthermore, it was suggested that shorter etching time which was similar to that of the other institutes would be possible if enlargement of the conventional track diameter is not considered in anticipation of introducing the automatic counting system. If an automatic counting system can be introduced, shorter time and simplicity of data analysis can be fulfilled.

Conflict of Interest Disclosure

The authors declare that they have no conflict of interest.

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