Change in the Sensitivity of CR-39 for Alpha-Tracks
after Storage at Different Temperatures

Hiroko ENOMOTO and Nobuhito ISHIGURE
National Institute of Radiological Sciences
4-9-1, Anagawa, Inage, Chiba JAPAN

INTRODUCTION

Solid state track detector CR-39 is used widely in the field of health physics, such as for radon monitoring or neutron dosimetry. Authors have used the CR-39 for α-particle measurement and α-particle autoradiography (1-4).

The authors have noticed that the diameter of etch pits observed in CR-39 stored for a long time became smaller than those observed in a fresh plate of CR-39. There are some reports about aging and/or fading effect of CR-39 (5-13). However, authors felt the necessity to examine more systematically the effect of aging and/or fading for the plates of CR-39 that were used by the authors themselves. In this study, the effect of storage on track registration property of CR-39 has been investigated. Pieces of CR-39 were irradiated with normally incident α-particles and fission fragments by using a ^{252}Cf source prior and posterior to the storage of them in air for one year at different temperatures of -80℃, -23℃, 4℃, 23℃ and 35℃.

The term "aging" is used for the change in the track registration property during storage until it is irradiated and "fading" means the change in the track registration property during storage after irradiation.

MATERIALS and METHODS

1. CR-39

Before the start of the experiment, the plate of CR-39 used had already been stored in a refrigerator of the manufacturer for 1 month and moreover for 2 months in a freezer of the author's laboratory at -23 °C. Therefore, three months had passed after it was manufactured at the time of the start of the experiment.

2. Storage of CR-39

Pieces of CR-39 were enclosed one by one in polyethylene bags with zipper so that background due to radon progeny might not increase during the storage. The outside of the polyethylene bag was wrapped in aluminum foil to prevent the effect of ultraviolet radiation on the property of the CR-39.

The pieces were stored (a) in a freezer of -80 °C, (b) in a freezer of -23 °C, (c) in a refrigerator of 4 °C, (d) in a laboratory, the humidity of which is kept 50% ± 10% and temperature is kept 23°C ± 2°C, and (e) in an incubator at 35 °C. The storage period was 1 year the longest. The etching was done from the start of the storage for 1 month, 2 months, 3 months, 5 months, 6 months, 9 months and one year

3. Irradiation method

An electro-deposited radioactive source of ^{252}Cf with 9 kBq was used for the irradiation of fission fragments and α-particles. The schematic diagram is shown in **Fig.1**.

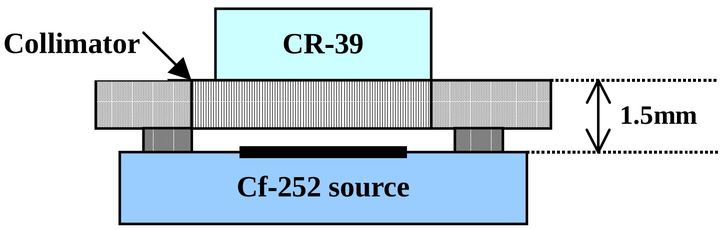


Fig.1 Schematic diagram of the experimental arrangement used to irradiate CR-39

A thin aluminum plate, 0.9 mm in thickness, was used as a collimator to minimize the source to CR-39 distance. Ten thousand holes, 0.22 mm in diameter spaced 0.3 mm apart were drilled in the aluminum plate. The collimator was put on the radiation source with a thin space. The resultant irradiation distance was 1.5 mm in the air. The irradiation time was 10 minutes.

4. Etching method

The sodium hydroxide aqueous solution of 7 mol•l^{-1} was used as the etching solution. The solution was prepared at every etching step on the previous day. The etching was done in a bottle with a tight lid to prevent change in the concentration of the etching solution due to vaporization of water and absorption of moisture. A water bath was used for heating the etching solution at 70 °C with the precision of ± 0.1 °C. The etching time was 4 hours.

5. Measurement of etch pit diameter

A picture of etch pit was taken by the video camera connected to a light microscope. The etch pit diameter was measured with a pair of calipers on the CRT screen. The total magnification was about 1,200 times and an instrumentation error was 0.15 μm.

RESULTS AND DISCUSSION

1. Change of the etch pit diameter

Photographs of CR-39 on which tracks were registered are shown in **Fig.2**. The series in the column (A) show the etch pits for the pieces of CR-39 stored at -80 °C, (B) at 23 °C and (C) at 35 °C.

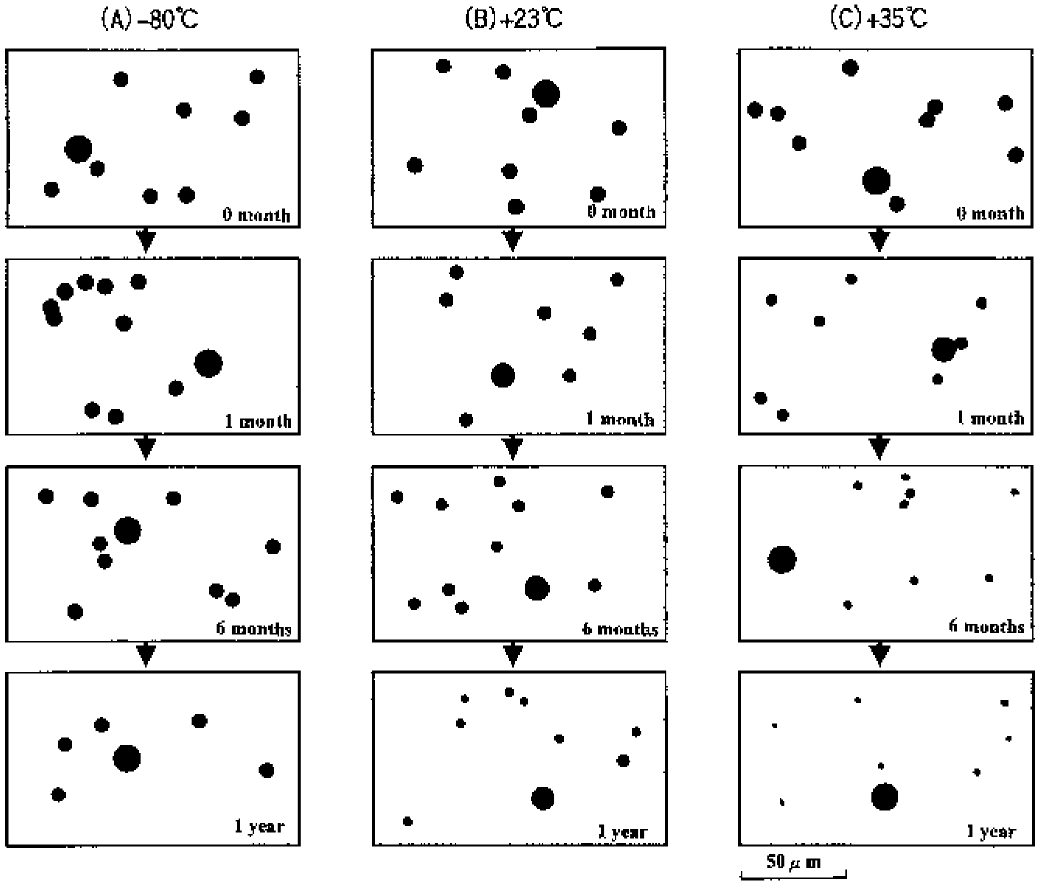


Fig.2 Photographs of CR–39 showing the changes of etch pit diameters during storage at the different temperatures.

The larger etch pits in each photograph are due to fission fragments and the smaller are due to α-particles.

These photographs clearly show that the etch pit diameters of α-particles deceased with the storage period and that the rate of the decrease depended markedly on the storage temperatures.

Figure 3 shows the results of the measured etch pit diameters; (a) is for the experiment of aging and

(b) for the experiment of fading.

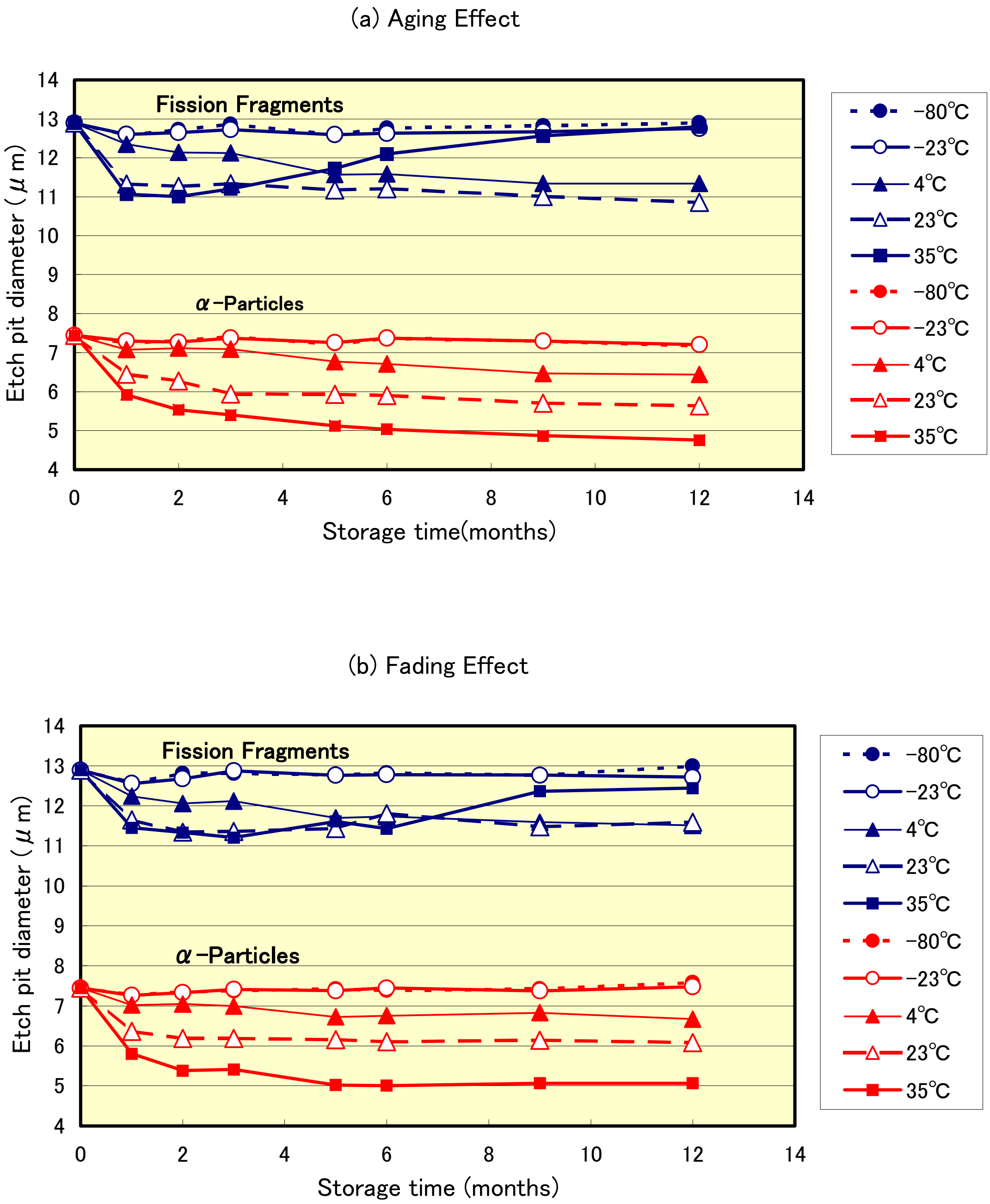


Fig.3 Diameters of etch pits of α-particles and fission fragments registered on CR-39 that were stored for one to 12 months at different temperatures

The changes of etch pit diameters are summarized as follows:

(1) After being stored at –80 °C and –23 °C, no change of etch pit diameters was observed.

(2) For the storage at the other temperatures, the etch pit diameters of α-particles decreased with the storage period. As to fission fragments, on the other hand, the etch pit diameters once decreased and then slowly increased again.

(3) The decrease rate of etch pit diameter of α-particles was as remarkable as storage temperature was high.

(4) Clear difference was not recognized between experiments for aging and fading.

2. Change of the bulk etch rate

The etch pit diameter D after etching for the time t is expressed by the equation

$$D = 2V_b t\sqrt{(V_t/V_b - 1)/(V_t/V_b + 1)} \quad (1)$$

where V_b is namely the bulk etch rate, that is the etching velocity of the CR-39 plate itself, and V_t is namely the track etch rate, that is the etching velocity along the track.

Because V_t/V_b>>1for fission fragments, the bulk etch rate is approximately expressed as a function of the etch pit diameter of the fission fragment D_f:

$$V_b = D_f/2t \quad (2)$$

The results for bulk etch rate obtained by this equation are shown in **Fig.4**.

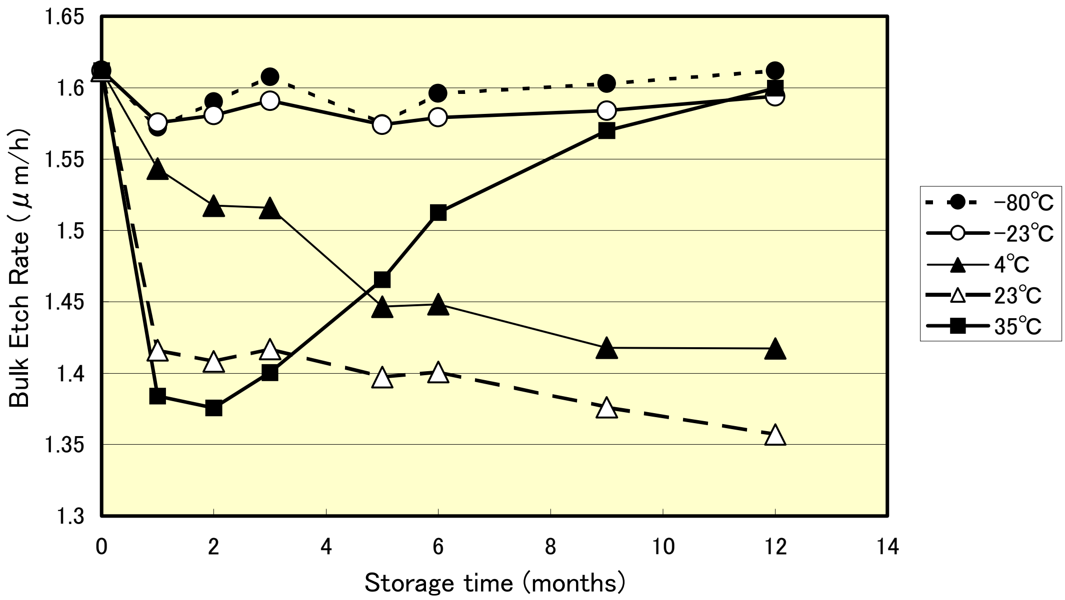


Fig.4 Changes in bulk etch rates of CR-39 during storage at different temperatures.

The changes of bulk etch rates of CR-39 observed in this study are summarized as follows.:

(1) The storage at –80 °C and –23 °C for one year did not bring any significant changes to the bulk etch rate.

(2) At 4 °C, the bulk etch rate decreased gradually with the storage period and reached down to 88 % at one year after the start of the experiment.

(3) At 23 °C, the bulk etch rate decreased down to 88% during one month. After that it continued to decrease slowly and reached to 84 % at one year after the start of the storage.

(4) At 35 °C, the bulk etch rate decreased during one month down to 86%. It turned off to the tendency of increase at 3 months. One year after, the bulk etch rate returned to almost the same as that at the start of the experiment

The change of bulk etch rate thus observed in this study is a new finding. Up to now the registration property of CR-39 has been investigated under an assumption that the bulk etch rate is almost constant even after storage for long time at room temperature.

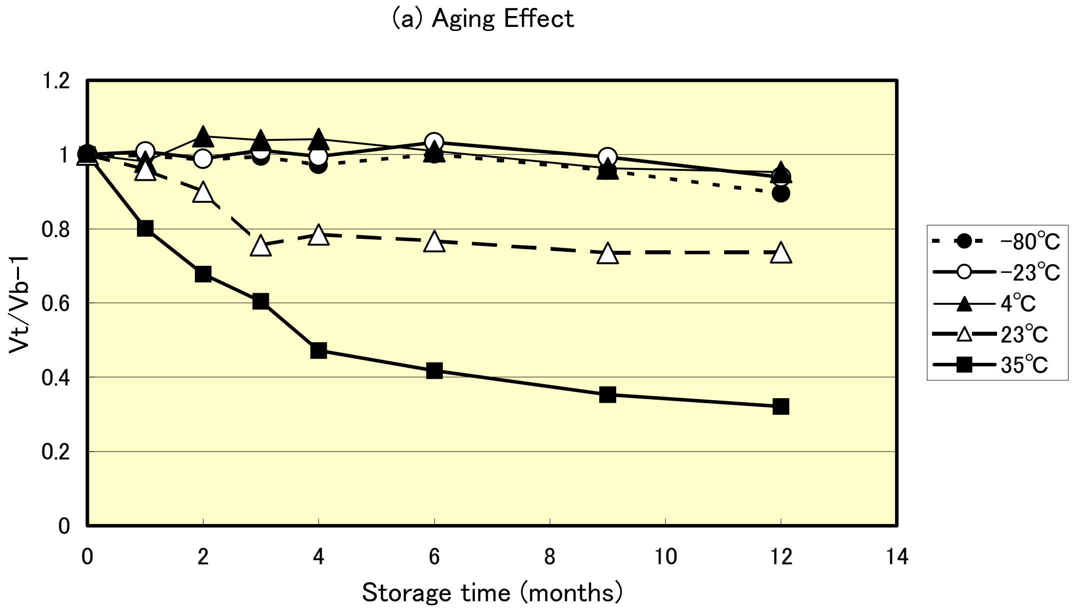
3. Change of sensitivity.

As an index of sensitivity of the solid state track detector, the value [V_t/V_b – 1] is often used. This value is derived from the equations (1) and (2) as the equation (3):

$$V_t/V_b - 1 = \left\{1 + (D_a/D_f)^2\right\} / \left\{1 - (D_a/D_f)^2\right\} - 1 \quad (3)$$

where D_a is the etch pit diameter of α-particles.

The sensitivity of CR-39 stored at different temperatures was calculated by using the equation (3) from the measured values of D_a and D_f. The results are shown in **Fig.5**; (a) is for the experiment of aging and



(b) for fading.

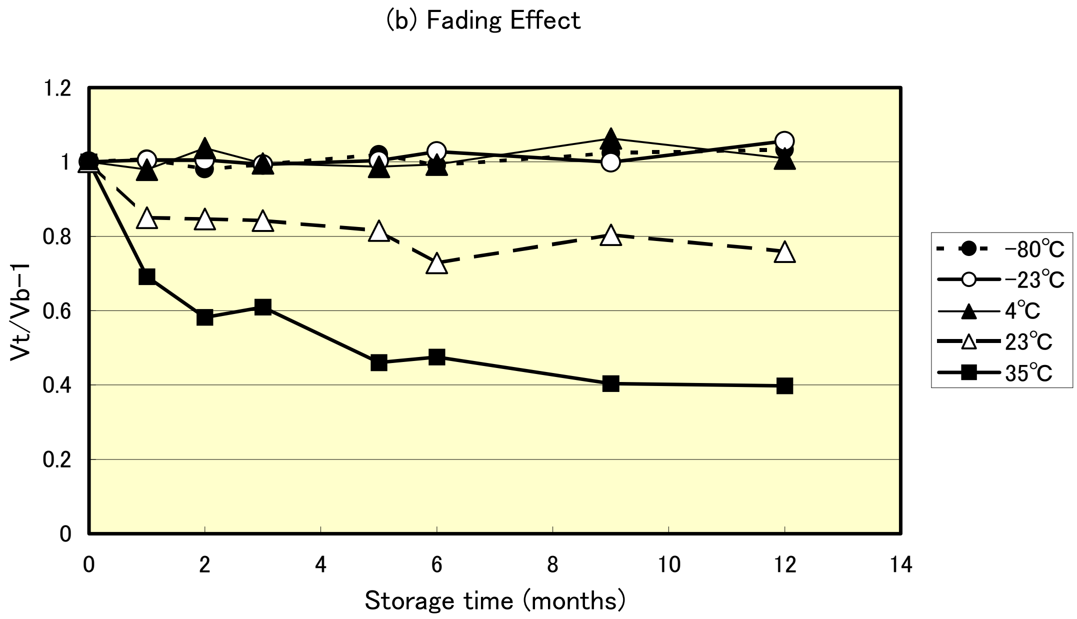


Fig.5 Changes in sensitivity of CR-39 during storage at different temperatures

The changes of sensitivity of CR-39 are summarized as follows.:

(1) The storage at the temperatures of –80 °C, –23 °C and 4 °C for one year did not bring any significant changes to the sensitivity. However, it should be noticed that the etch pit diameters themselves both for α-particles and fission fragments at 4 °C changed a little as shown in **Fig.3**.

(2) At 23 °C, the sensitivity decreased gradually with storage period. It reached down to 74% (aging) and 76% (fading) one year after the start of the experiment.

(3) At 35 °C, the sensitivity decreased largely with storage period and reached down to 32% (aging) and 40% (fading) for the storage of one year. The difference between the experiments of aging and fading is not necessarily meaningful because of the large uncertainty in the measurement of the sensitivity of solid state track detectors. This strongly suggests that the storage effect is attributed not to fading of latent tracks but mainly to some changes in the detector itself.

4. Change of the detection efficiency

In an application of CR-39 to radon monitoring or neutron dosimetry, the number of etch pits is important. Therefore, constancy of detection efficiency of CR-39 is a key factor determining the reliability of the measured results in these application.

The detection efficiency *f* of the solid state track detector is expressed by the equation

$$f = (1/2\pi)\int_{\theta_c}^{\pi/2} d\Omega = 1 - \sin\theta_c \qquad (4)$$

where θ_c, called as a critical angle, is an angle of incidence of α-particles from the surface of CR-39, over which the tracks are observed. The angle θ_c is expressed as

$$\theta_c = \arcsin(V_b/V_t) \qquad (5)$$

The value of the detection efficiency was calculated by using the equations (4) and (5) and the result for the experiment of aging is shown in **Fig.6**. The results for the experiment of fading are omitted, because they were nearly the same as those for the experiment of aging.

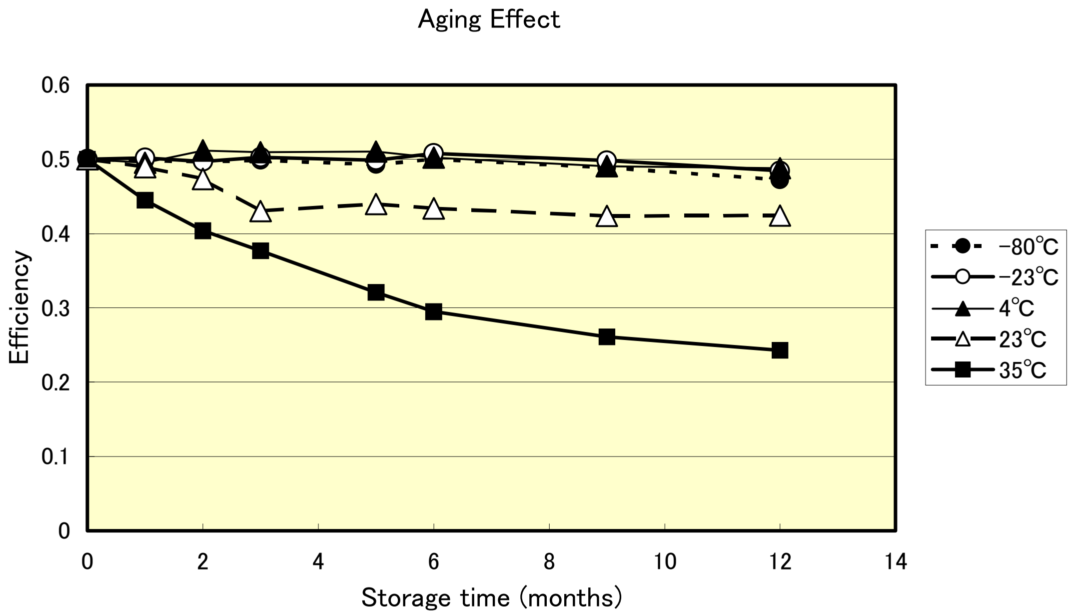


Fig.6 Changes in detection efficiency of CR-39 during storage at different temperatures

The storage at the temperatures of –80 °C, –23 °C and 4 °C for one year did not bring any significant changes to the detection efficiency, which is easily derived from the results for the sensitivity mentioned before. At 23 °C, the detection efficiency decreased from 0.5 to 0.42 by the storage of one year. At 35 °C , the detection efficiency decreased the most remarkably from 0.5 to 0.3.
quantitatively before the interpretation of the measurement fructification.

CONCLUSION

When CR-39 is used for radon monitoring or neutron dosimetry, the following points should be paid attention to: (1) the detector should be stored in a refrigerator before exposure and until etching after the exposure, (2) the change in the sensitivity between the time of calibration and the time of use should be evaluated and the counting efficiency at the measurement should be corrected and (3) for comparison or for interpretation of experimental results in different experiments the effect of storage should be carefully considered.

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