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Sensitization of PADC track detector in carbon dioxide gas

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Abstract

The fading of carbon dioxide treatment on PADC detectors after alpha-particle irradiations has been confirmed to relate to the diffusion loss of carbon dioxide. Diffusion coefficient of carbon dioxide in the film is found to be $D = 1.1\mathrm{x}10^{-9}$ cm^2/s at room temperature. The effect of the post-irradiation treatment has strong relation with the dissolving process of carbon dioxide into the etching solution through the renewing surface of the detector during etching. Absorbed carbon dioxide can modify the network structure of PADC with higher permeability for etching solution.

PACS: 61.80.Ed; 61.62.Pv
Keywords: Latent track; CR-39; Carbon dioxide; FT-IR; Sensitization

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1. Introduction

The sensitization effects on PADC, poly(allyl diglycol carbonate), type track detectors with carbon dioxide gas at several atmospheres was reported firstly by Fujii et al. (1995; 1997), which was soon followed by a comprehensive examination by Csige (1997). The post-irradiation sensitizing was found to be effective and convenient so that has been applied for neutron and Radon dosimetry (Hulber et al., 2005), and also micromachining process using proton beams (Baradács et al., 2008). Nevertheless the fundamental process of the sensitivity enhancement by the treatment has not yet been resolved, from which we could learn the way to control the response of PADC detectors.

The present study is attempted to confirm the sensitization effects once again and obtain fundamental information to understand in what way the absorbed carbon dioxide acts directly or indirectly to increase the sensitivity of PADC detectors. Effect of the sensitization has been studied on the pressure of the carbon dioxide up to 0.6 MPa and the treatment time up to two weeks for alpha-particles with a several incident energies, for thicker PADC detectors with 0.9 mm. The fading of the sensitization with time between the treatment and etching has been examined for 6.1 MeV alpha-particles for thinner detector films with 100 μm. Releasing behavior of carbon dioxide from the treated films was also examined by means of FT-IR spectrometry.

2. Experimental

PADC detectors utilized throughout this study were BARYOTRAK sheets (Fukuvi Chemical Industry, Japan), which were produced from the highly purified monomer, with different two nominal thicknesses of 0.9 mm and 100 μm. To estimate the effect of the carbon dioxide treatment, the thick samples cut in 2.5×2.5 cm^2 size were exposed to normal incident alpha particles with energies of 0.52, 1.68, 2.78, 3.83 and 4.94 MeV from Am-241 source, by changing the distance between the samples and the source in air. Every sample was also irradiated with fission fragments, as well as 6.1 MeV alpha particles, from Cf-252 source in vacuum, to determine the amount of layer removed during the following etching. To evaluate the fading behavior of the treatment, the other thin samples with 2.5×2.5 cm^2 were irradiated using the Cf-252 source.

Carbon dioxide treatment was carried out in a stainless steel chamber, which was connected to the carbon dioxide cylinder with a pressure regulator, and also to a rotary pump. Prior to the introduction of carbon dioxide gas, the chamber room with samples was evacuated to below a few Pa. Then the samples were exposed to the introduced gas at a room temperature with the various pressures ranging from 0.1 to 0.6 MPa for different durations up to 2 weeks. The following chemical etchings were started within 5 min after the exposure to evaluate the treatment effect for the thick samples. Fading behavior of the treated thin films has been estimated by changing the interval between the gas exposure; at 0.6 MPa for 24 h, and the etching.

Chemical etching was performed in 6 M KOH solution kept at 70ºC for one and 2 h, for thick and thin samples, respectively. Pit observation was made under an optical microscope (OLYMPUS BX60-F3), which was operated on PC with WinROOF Ver 5.04 image processor (MITANI Corporation, Japan). FT-IR measurements were made to estimate the amount of absorbed carbon dioxide in the thin films (FTIR-8400S, SHIMADZU, Japan).

3. Results and discussion

3.1. Carbon dioxide treatment effect

Bulk etch rate increased slightly but clearly by the treatment in 0.6 MPa carbon dioxide gas with the treatment time, as shown in Fig. 1. The relative etch rate was determined as the ratio of the averaged pit radius of fission fragments on the treated PADC sheets to that of un-treated ones. The both sheets were cut as neighbors from the same lot and etched simultaneously in the same etching solution. Sensitivity, namely, the reduced etch rate ratio of treated samples to alpha-particles was also found to increase under 0.6 MPa with the treatment time, as shown in Fig. 2, resulted in the same trend to that previously reported by Csige for a different kind of PADC (1997). This implies that the track etch rate was enhanced more significantly by the present treatment than the bulk etch rate.

Figure 3 indicates dependence of the sensitivity on the pressure of carbon dioxide gas for three days storage, showing almost monotonous increment with the pressure. It shows saturation behavior above 0.2 MPa, especially for lower incident energies. Csige has reported relatively higher sensitivity at 0.9 MPa (1997).

Figure 4 shows the increase of sensitivity with storage duration in carbon dioxide as a function of stopping power of the alpha-particle in PADC. Plotted data are equivalent to those in Fig. 2. It is hard to find significant dependence of the present carbon dioxide treatment on the stopping power of incident alpha-particles.

3.2. Fading of the treatment effect

Figure 5 shows clear decrease of the sensitivity after the treatment with time. Csige did the similar experiment in order to study the effect of aging of latent tracks after the treatment up to 1440 min (1997). He found only slight decrease in the sensitivity, on the contrary to the present result. This might be the result that he utilized normal thick PADC sheets in the experiment. The present authors used the thin samples with a thickness of 100 µm, to make it possible for carbon dioxide gas to escape from the films more rapidly. We think the releasing behavior should be governed by diffusion mechanism as discussed in the later.

We can observe the carbon dioxide absorbed in PADC films by means of FT-IR spectrometer, as shown in Fig. 6. The absorption peaks around 2340 cm^{-1} and 660 cm^{-1} are assigned to anti-symmetric vibration and bending vibration, respectively. The IR spectra of carbon dioxide in PADC films were different in the shape from that in the atmosphere, due to the suppression of rotational moving in the films. As shown in Fig. 7, the height of peaks decreases with time after the treatment. This indicates that carbon dioxide was diffusing out from the PADC film with passing the time. After 1440 min, there exists no detectable carbon dioxide in it. The fading of the treatment should relate to the diffusion loss of carbon dioxide as observed in the present work.

The concentration of carbon dioxide, $c(x,t)$, in a PADC sheet with a thickness of *d* kept in air after the treatment should be expressed by the following one dimensional diffusion equation with sufficient initial and boundary conditions;

$$\frac{\partial c(x,t)}{\partial t} = D\frac{\partial^2 c(x,t)}{\partial x^2}, \tag{1}$$

$$t = 0 : c(x,0) = c_0, \quad 0 \le x \le d,$$

$$t > 0 : c(0,t) = c(d,t) = 0,$$

where D is diffusion coefficient and c_0 is equilibrium concentration in the treatment chamber. The chamber is opened at $t = 0$. By solving this, we obtain the followings;

$$c(x,t) = \frac{4c_0}{\pi}\sum_{n=0}^{\infty}\frac{1}{2n+1} sin\left\{\frac{(2n+1)\pi}{d}x\right\}\exp\left[-\left\{\frac{(2n+1)\pi}{d}\right\}^2 Dt\right]. \tag{2}$$

After long time storage in air, $Dt/d^2 > 0.025$, it becomes simply as,

$$c(x,t) = \frac{4c_0}{\pi} sin\left(\frac{\pi}{d}x\right)\exp\left\{-\left(\frac{\pi}{d}\right)^2 Dt\right\}. \tag{3}$$

Then the averaged concentration in the sheet, $\bar{c}$, is described like,

$$\bar{c} = \frac{1}{d}\int_0^d c(x,t)dx = \frac{8c_0}{\pi^2}\exp\left\{-\left(\frac{\pi}{d}\right)^2 Dt\right\}. \tag{4}$$

Therefore, IR absorbance, A, should be written as,

$$A = \varepsilon\bar{c}d = \frac{8c_0}{\pi^2}\varepsilon d\exp\left\{-\left(\frac{\pi}{d}\right)^2 Dt\right\}, \tag{5}$$

where ε is the molar absorbance coefficient of carbon dioxide. Obtaining the natural logarithm of both sides of equation (5);

$$\ln A = \ln\left(\frac{8c_0}{\pi^2}\varepsilon d\right) - \left(\frac{\pi}{d}\right)^2 Dt. \tag{6}$$

This relation indicates that the slope of the plot of $\ln A$ against $(\pi/d)^2 t$ gives the diffusion coefficient. We have obtained $D = 1.1\text{x}10^{-9}$ cm^2/s at room temperature as a tentative value.

3.3. Escape of carbon dioxide during etching

Finally, the behavior of carbon dioxide during etching has been examined. Within 5 min after the treatment, a sample was etched for 2 h. Then FT-IR measurement was made for this sheet, at 3.5 h after the treatment. As shown in Fig. 8, carbon dioxide is released away totally during the etching, on the other hand the treated film kept in air for 3.5 h contains it clearly. This can be explained by the increase of the diffusion coefficient at elevated temperatures. In addition, we observed the bubbles of carbon dioxide on the surface of the treated PADC film, which was immersed in distilled water about 80°C. This is the other proof that carbon dioxide will diffuse out from the surface during the etching.

When etchant is penetrating into PADC, carbon dioxide is passing through the opposing direction from PADC to the solution. Carbon dioxide can modify the polymer network structure by plasticization where it is absorbed (Bos et al., 1999). It can also react with K^+ ions in the etching solution, for an example, forming K_2CO_3 (Tse et al., 2007). We need to construct a sufficient model for the present sensitizing effect.

4. Conclusion

The effect of carbon dioxide treatment has been confirmed on PADC detectors after alpha-particle irradiations. The fading of the treatment was found to have relation with the diffusion loss of carbon dioxide in PADC films. As a tentative value, diffusion coefficient of carbon dioxide in the film was determined to be D = 1.1x10^{-9} cm^2/s at room temperature.

During the etching, carbon dioxide was found to escape into the etchant. This implies that the effect of the post-irradiation treatment relates to the dissolving process of carbon dioxide into the etching solution through the renewing surface of PADC detector. It is true that carbon dioxide can modify the network structure of PADC and also some property of the solution. The treatment may alter the condition of the interface. We need to construct a realistic model for the carbon dioxide treatment based on the present work and recent studies on the etching process.

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Figure Captions

Figure 1 Changes in bulk etch rate caused by the carbon dioxide treatment at 0.6 MPa with the treatment duration. The longitudinal bar at the end datum indicates the statistical deviation.

Figure 2 Increase of reduced etch rate ratio for alpha-particles with incident energies of 0.52, 1.68, 2.78, 3.83 and 4.94 MeV due to 0.6 MPa carbon dioxide treatment against the treatment time. Plots for 6.1 MeV are according to Csige (1997).

Figure 3 Changes in sensitivity with treatment gas pressure. Time of treatment was for 3 days.

Figure 4 Response of PADC detectors for alpha-particles with and without carbon dioxide treatment, at 0.6 MPa for various times.

Figure 5 Decrease of the sensitivity after the treatment with time.

Figure 6 FT-IR spectra of PADC films after the treatment.

Figure 7 FT-IR spectra of carbon dioxide in PADC films after the treatment.

Figure 8 FT-IR spectra of treated PADC films stored in air and after the chemical etching.

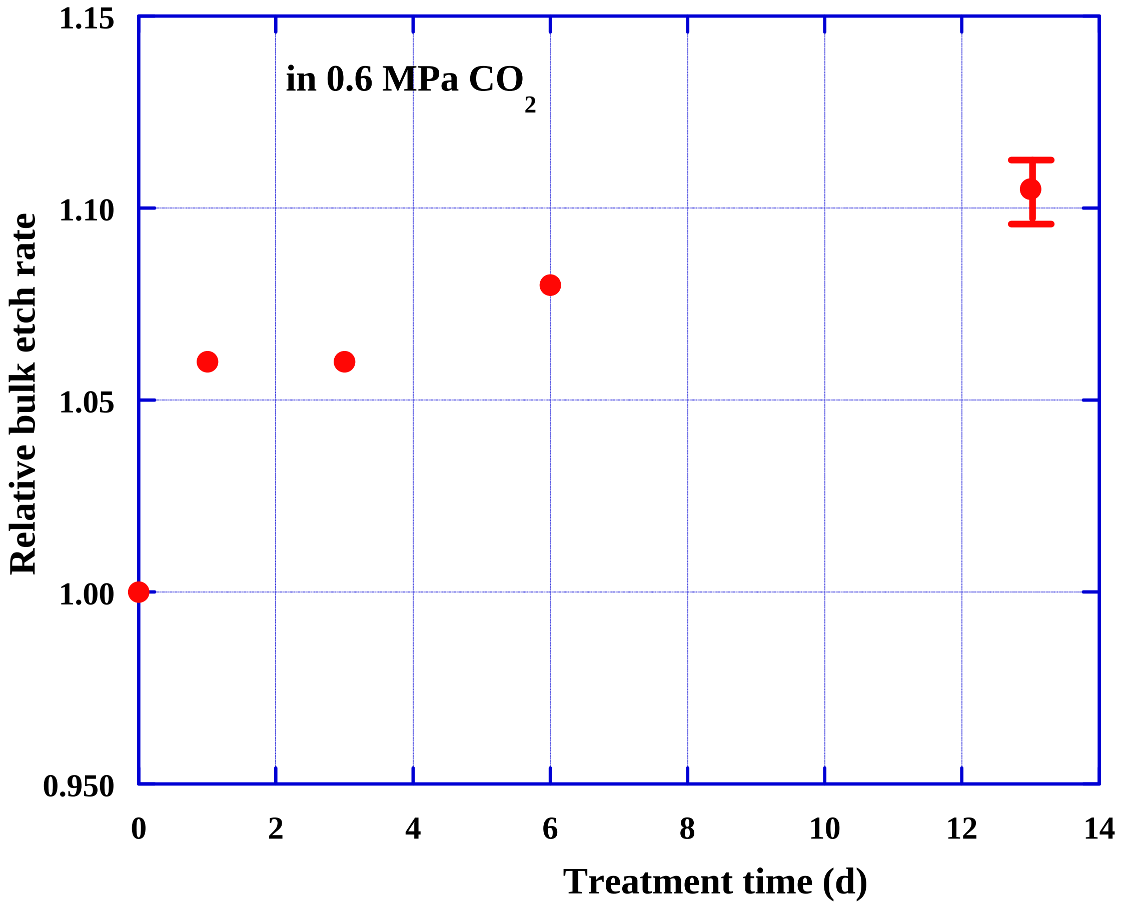
in 0.6 MPa CO_2
Relative bulk etch rate
1.15
1.10
1.05
1.00
0.950
0
2
4
6
8
10
12
14
Treatment time (d)

Fig. 1

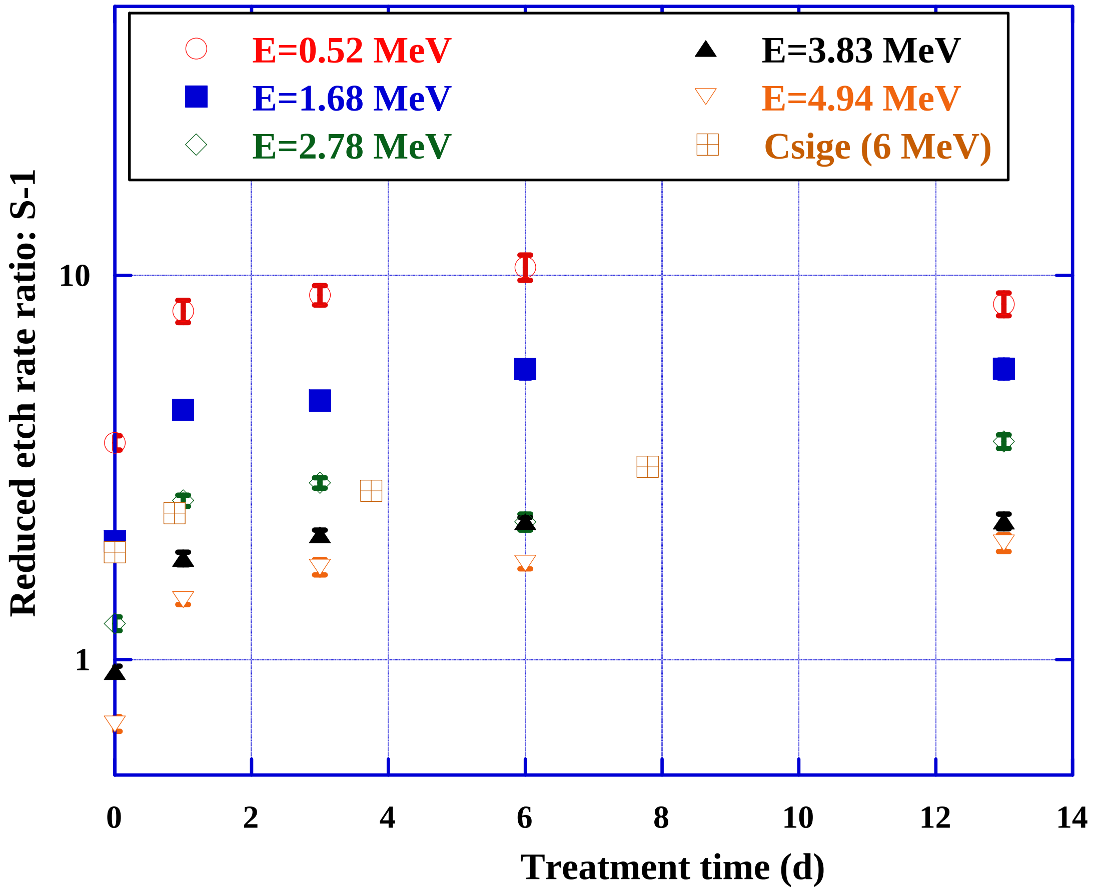
○ E=0.52 MeV
■ E=1.68 MeV
◇ E=2.78 MeV
▲ E=3.83 MeV
▽ E=4.94 MeV
⊞ Csige (6 MeV)
Reduced etch rate ratio: S-1
10
1
0
2
4
6
8
10
12
14
Treatment time (d)

Fig. 2

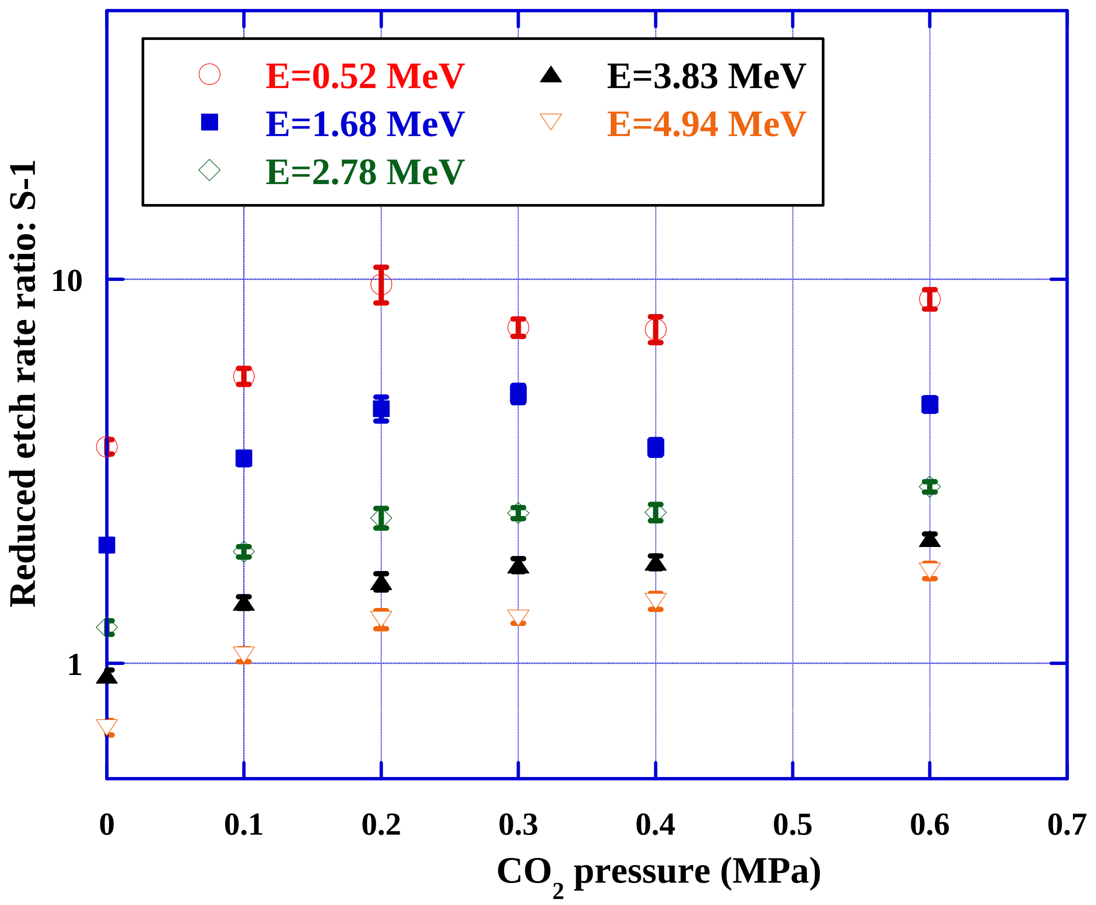
○ E=0.52 MeV
■ E=1.68 MeV
◇ E=2.78 MeV
▲ E=3.83 MeV
▽ E=4.94 MeV
Reduced etch rate ratio: S-1
10
1
0
0.1
0.2
0.3
0.4
0.5
0.6
0.7
CO$_2$ pressure (MPa)

Fig. 3

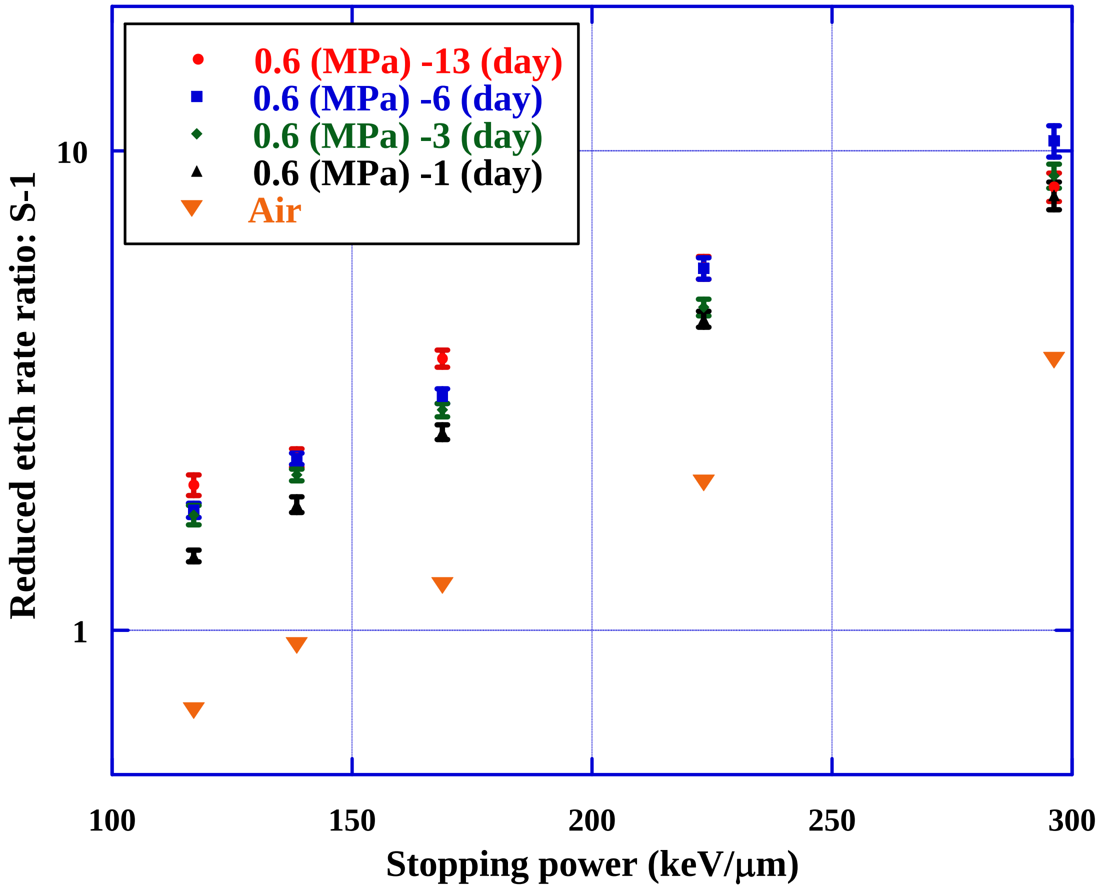
0.6 (MPa) -13 (day)
0.6 (MPa) -6 (day)
0.6 (MPa) -3 (day)
0.6 (MPa) -1 (day)
Air
Reduced etch rate ratio: S-1
10
1
100
150
200
250
300
Stopping power (keV/μm)

Fig. 4

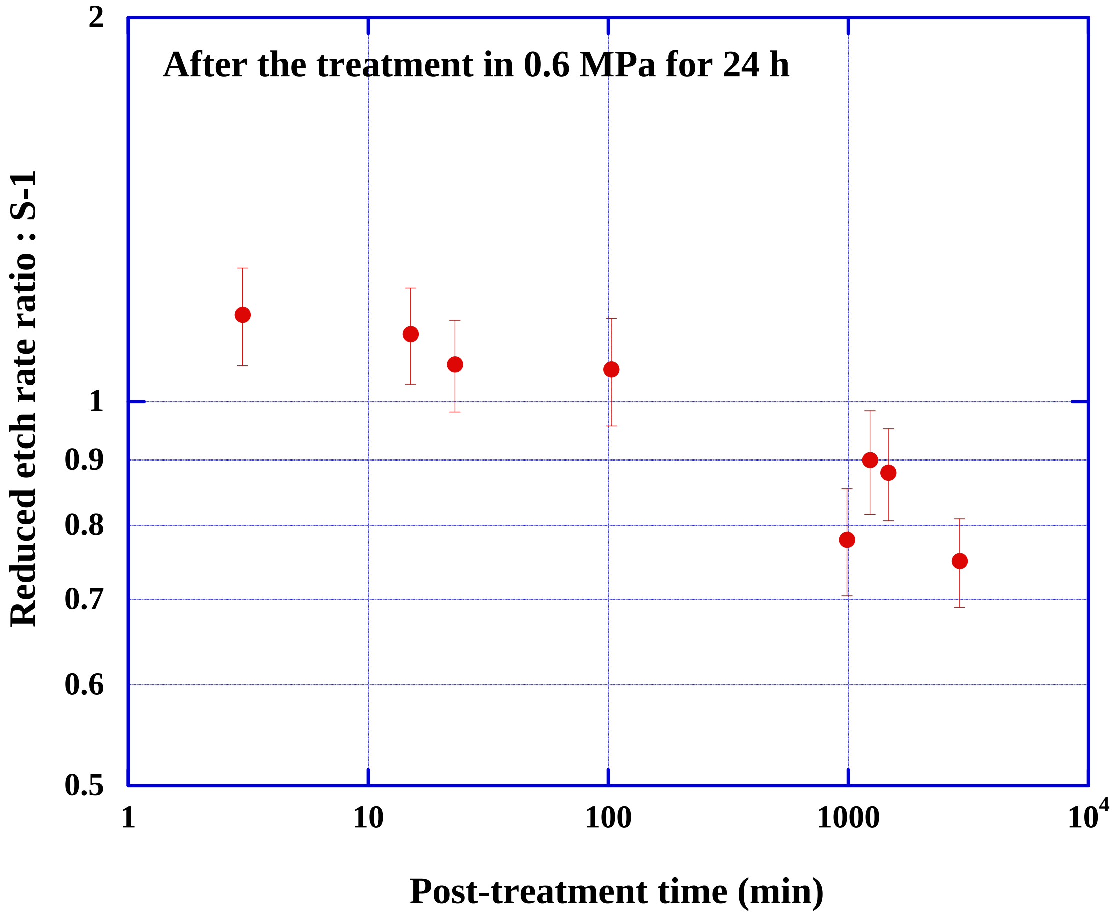
After the treatment in 0.6 MPa for 24 h
Reduced etch rate ratio : S-1
2
1
0.9
0.8
0.7
0.6
0.5
1
10
100
1000
10^4
Post-treatment time (min)

Fig. 5

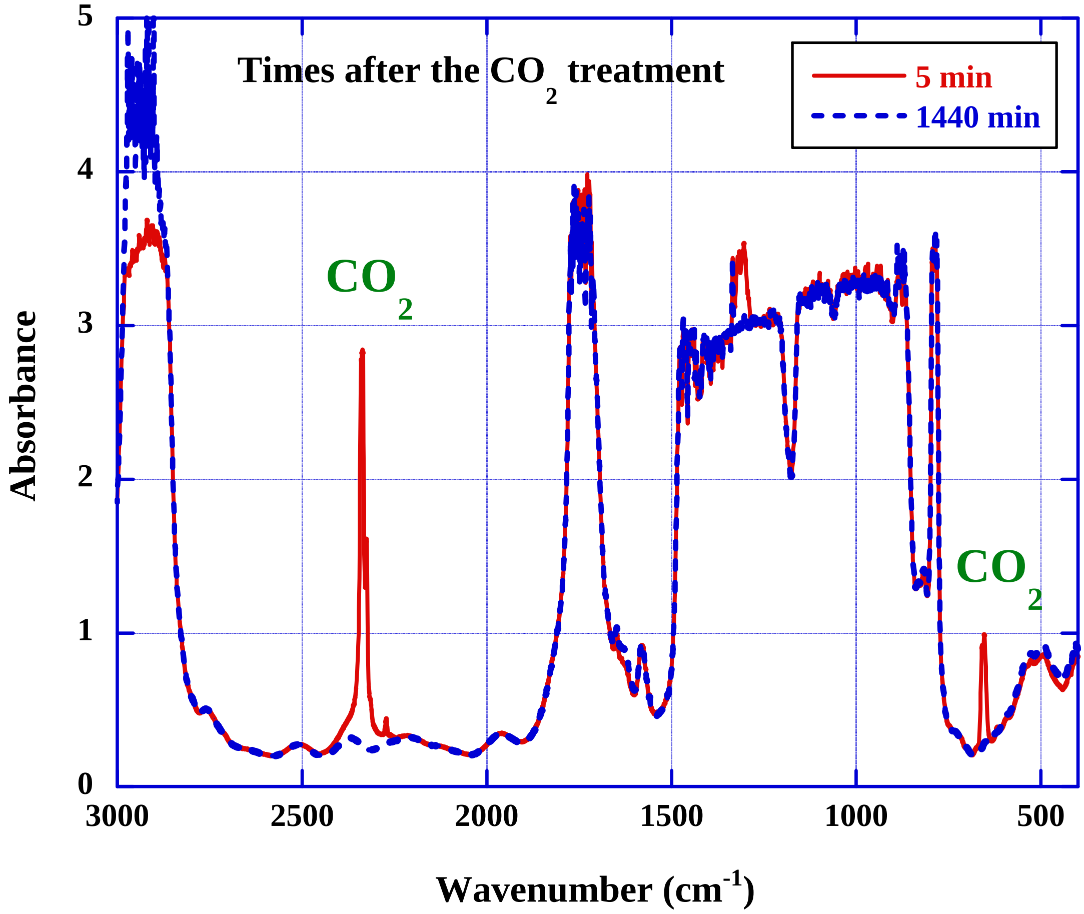
Times after the CO_2 treatment
5 min
1440 min
CO_2
CO_2
Absorbance
0
1
2
3
4
5
3000
2500
2000
1500
1000
500
Wavenumber (cm^{-1})

Fig. 6

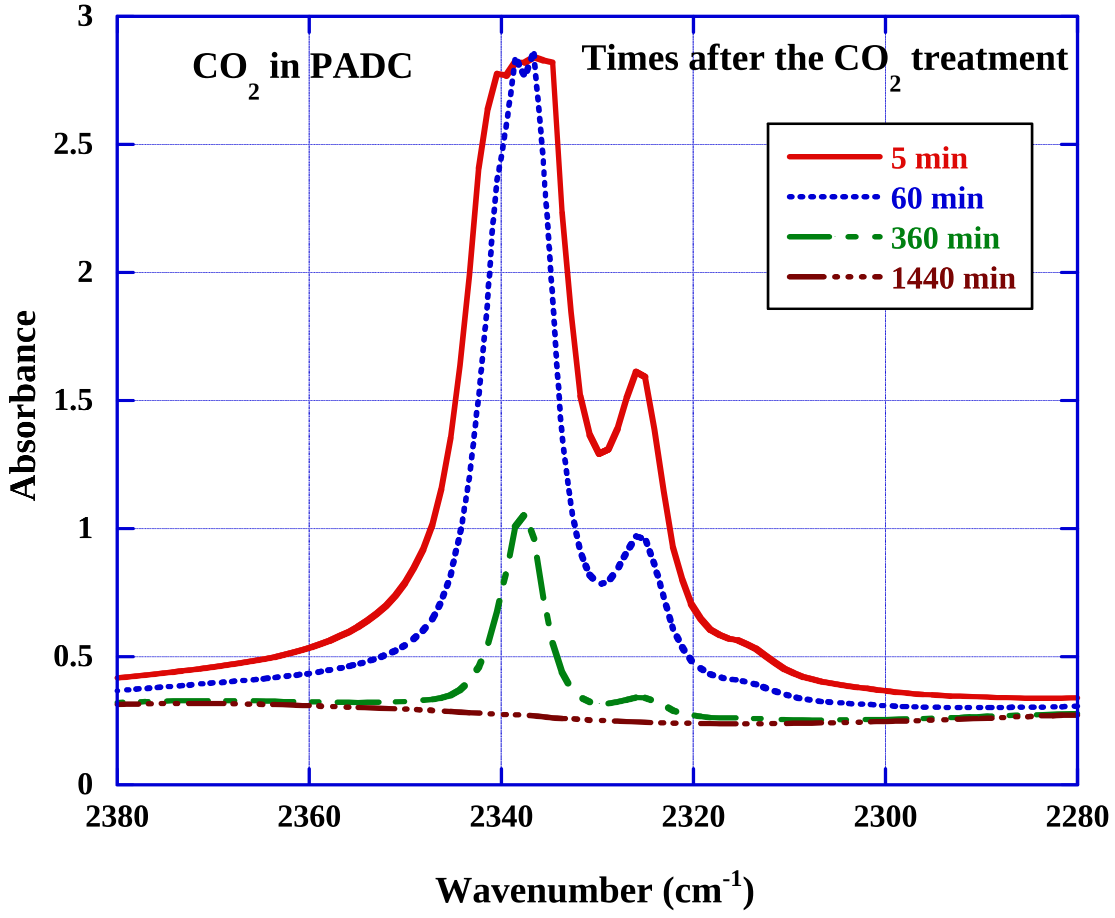
CO$_2$ in PADC
Times after the CO$_2$ treatment
5 min
60 min
360 min
1440 min
Absorbance
0
0.5
1
1.5
2
2.5
3
2380
2360
2340
2320
2300
2280
Wavenumber (cm^{-1})

Fig. 7

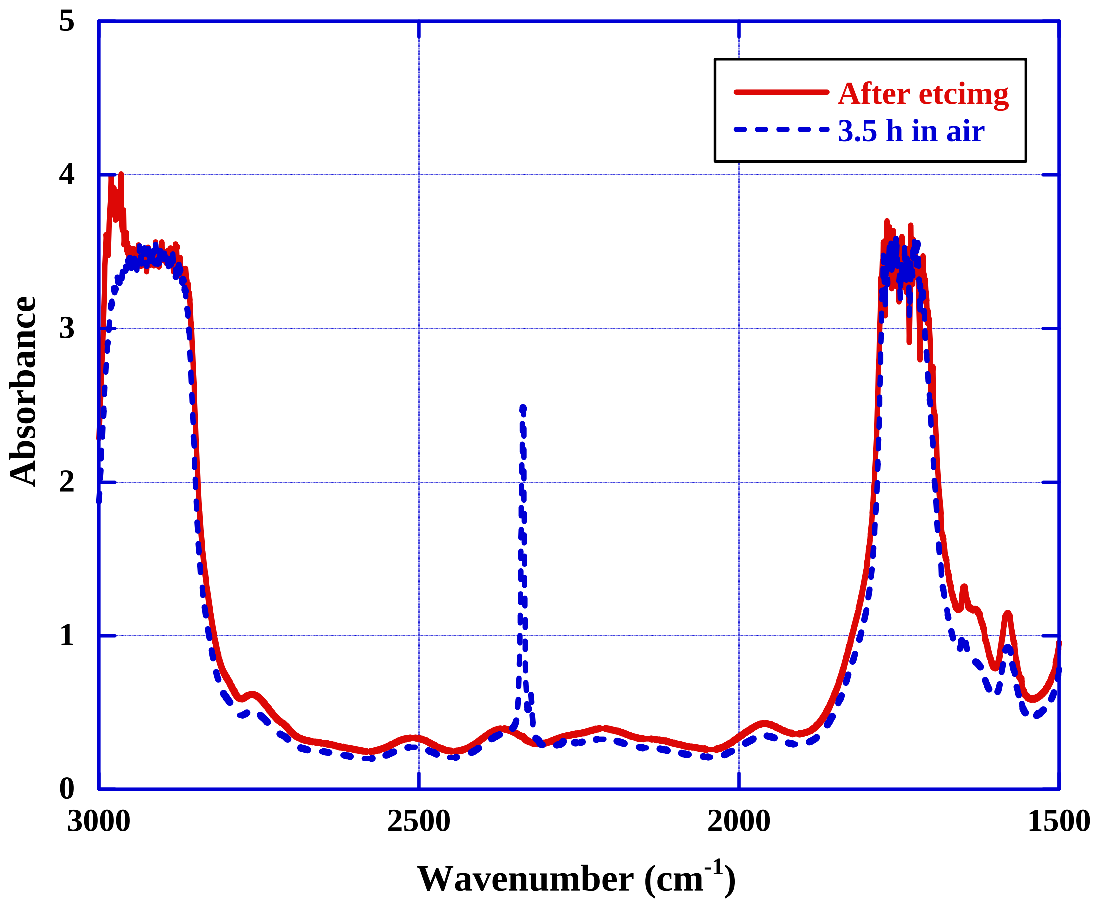
After etcimg
3.5 h in air
Absorbance
0
1
2
3
4
5
3000
2500
2000
1500
Wavenumber (cm^{-1})

Fig. 8