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FT-IR spectroscopy of carbon dioxide in CR-39 and SR-90 track detectors irradiated with ions and gamma-rays at different energies and fluences

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Abstract

Two types of polymeric track detectors CR-39 and SR-90 were irradiated with protons, alpha particles, heavy ions and gamma-rays at different energies and fluences. After irradiation these detectors were analyzed with an FT-IR spectrometer of Jasco type 5300 in transmission and ATR modes. We have found that CO_2 is produced not only by irradiation but also by polymerization. The amount of CO_2 in the detector material is closely related to the latent track formation.

Keywords: Polymeric track detectors; FT-IR spectroscopy; CO_2; Latent tracks

1. Introduction

Ionizing particles passing through polymeric track detectors produce latent tracks that are trails of radiation damage. These tracks are usually revealed as etch pits through chemical etching. However, some information about the structure of radiation damage will be lost after etching. As a new tool for our study, we introduced Fourier transform infrared spectroscopy (FT-IR spectroscopy) in order to make direct analyses of the change in the molecular structure along latent tracks in CR-39 and SR-90. It should be noticed that FT-IR spectroscopy has already been applied to studies on nuclear tracks in solids by several research groups (Gagnadre et al., 1993; Chong et al., 1997; Barillon et al., 1999; Malek, 1999).

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2. Experimental

Two kinds of polymeric track detectors CR-39 and SR-90 (Fujii et al., 1993) were irradiated with protons, alpha particles, heavy ions and gamma-rays at different energies and fluences. Proton irradiation was performed at the Tandem van de Graaff accelerator in Kobe University of Mercantile Marine, Japan, and Heavy ion irradiation at NIRS (National Institute of Radiological Science)—HIMAC (Heavy Ion Medical Accelerator in Chiba). Alpha- and gamma-ray irradiations were performed with radioactive sources, ^{241}Am, ^{252}Cf and ^{60}Co.

In the experiment, we used several types of detectors: CR-39 (IC: Intercast) made by the Intercast Europe Co. of Parma, Italy, CR-39 (BT: Baryotrak) supplied by Fukuvi Chemical Industry Co. Ltd., Japan, CR-39 (HM: Homemade) and SR-90 (HM: Homemade) polymerized in our laboratory. The thickness of CR-39 (IC), CR-39 (BT), CR-39 (HM) and SR-90 (HM) were about 660, 850, 930 and 930 μm, respectively. Conditions of the irradiation are summarized in Tables 1 and 2.

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Table 1
List of irradiations with accelerators

Sample code	Ion	Energy (MeV/n)	Fluence (ions/cm^2)	Detector
ICp1	p	3.4	1×10^{13}	CR-39 (IC)
ICp2	p	3.4	5×10^{13}	CR-39 (IC)
ICXe1	Xe	290	1×10^{6}	CR-39 (IC)
ICXe2	Xe	290	1×10^{8}	CR-39 (IC)
ICXe3	Xe	290	1×10^{9}	CR-39 (IC)
BTAr	Ar	550	1×10^{9}	CR-39 (BT)
BTFe	Fe	6	1×10^{11}	CR-39 (BT)
ICFe1	Fe	6	1×10^{10}	CR-39 (IC)
ICFe2	Fe	6	5×10^{10}	CR-39 (IC)
ICFe3	Fe	6	1×10^{11}	CR-39 (IC)

Table 2
List of irradiations with radioactive sources

Sample code	Ion	Irradiation source	Exposure (days)	Detector
SHM1	α, f.f.	^{252}Cf (37 kBq)	0–52	SR-90 (HM)
SHM2	α	^{241}Am (37 kBq)	0–52	SR-90 (HM)
ICα1	α	^{241}Am (37 kBq)	43	CR-39 (IC)
ICα2	α	^{241}Am (2.8 MBq)	51	CR-39 (IC)
ICγ	γ	^{60}Co (3.7 MBq)	14	CR-39 (IC)

An FT-IR spectrometer of Jasco type 5300 was operated in both transmission and ATR (Attenuated Total Reflection) mode because our CR-39 and SR-90 detectors were too thick for the majority of IR absorption bands in transmission mode. ATR-500/M, an attachment equipped with a KRS-5 prism, was used for ATR mode measurements. The working range of the spectrometer is 4600–400 cm^{-1}. A resolution of 4 cm^{-1}, scan numbers of 16 and apodization of cosine were used.

3. Results and discussion

As an example, an FT-IR spectrum in ATR mode for an unirradiated sample of CR-39 (IC) is shown in Fig. 1. It was hard to find differences in spectra between irradiated and unirradiated samples of CR-39 (IC) because changes in the molecular structure due to our irradiation condition were too small to be detected in ATR mode. No difference between spectra of CR-39 (IC) and SR-90 (HM) was seen because the molecular structure of these detectors was almost the same except for the chain length between two adjacent carbonate groups (Fujii et al., 1993). Some of the absorption bands are identified with the help of a structure analysis program, IR Mentor Pro of Sadtler. The broad absorption around 2920 cm^{-1} is due to C–H stretching. The strong absorption at 1738 cm^{-1} is due to C = O stretching in carbonate groups. The absorption band at 874 cm^{-1} is identified as that of ether groups.

An FT-IR spectrum in transmission mode is shown in Fig. 2 for a sample of CR-39 (IC) that was exposed to 3.4 MeV/n protons at a fluence of 5×10^{13} ions/cm^2. In this mode, the majority of IR absorption bands is saturated but two absorption bands of CO_2 at 2340 and 655 cm^{-1} are detected. In order to see the larger absorption at 2340 cm^{-1} in an enlarged scale, a part of this spectrum in the wave number range from 3000 to 2000 cm^{-1} is shown in Fig. 3. In this figure, a spectrum of CR-39 (IC), which was exposed to protons of the same energy at a lower fluence of

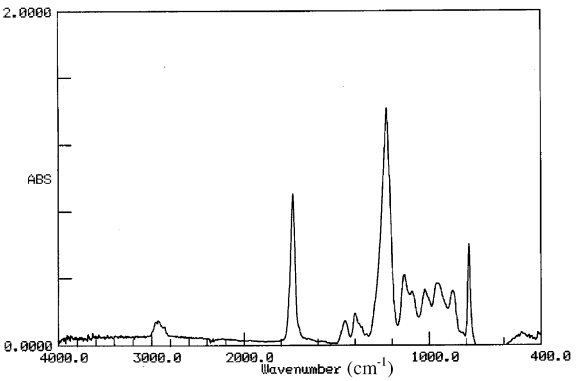

Fig. 1. FT-IR spectrum of unirradiated CR-39 (IC) in ATR mode.

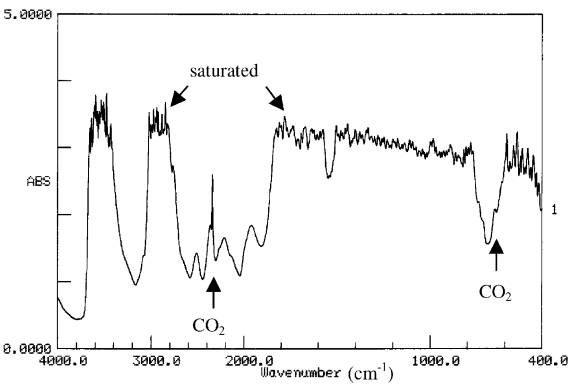

Fig. 2. FT-IR spectrum of irradiated CR-39 (IC) in transmission mode. Irradiation was performed with 3.4 MeV/n protons at a fluence of 5×10^{13} ions/cm^2.

1×10^{13} ions/cm^2, is also shown. From these two spectra, it is clear that the CO_2 absorption increases on increasing the fluence of protons. Here, it should be mentioned that atmospheric CO_2 background was carefully subtracted with the spectrometer.

In Fig. 3, spectra of other samples measured in transmission mode are shown. Small absorption bands of CO_2 at 2340 cm^{-1} are detected in all spectra of CR-39 (IC) samples irradiated with 6 MeV/n Fe at a fluence of 1×10^{11} ions/cm^2,

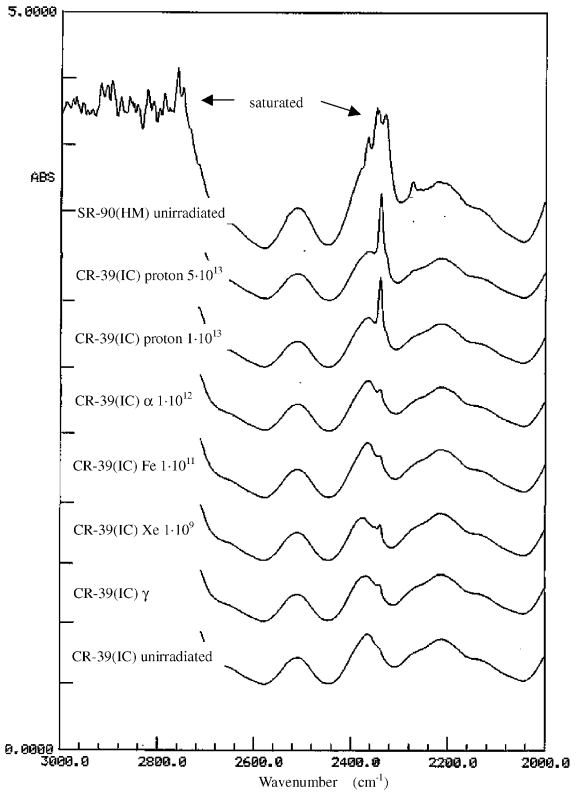

Fig. 3. FT-IR spectra of unirradiated SR-90 (HM), unirradiated CR-39 (IC) and irradiated CR-39 (IC) in transmission mode. Irradiations were performed with protons, alpha particles, Fe, Xe and gamma-rays at different energies and fluences as shown in Tables 1 and 2.

290 MeV/n Xe at a fluence of 1×10^9 ions/cm^2, alpha particles from 2.8 MBq ^{241}Am for 51 d and gamma-rays from 3.7 MBq ^{60}Co for 14 d. The estimated fluence of alpha particles on the CR-39 (IC) is about 1×10^{12} ions/cm^2. We suppose that CO_2 in these samples is mainly produced by the irradiation, because absorption at 2340 and 655 cm^{-1} is very small in unirradiated CR-39 (IC).

On the other hand, in the case of an unirradiated sample of SR-90 (HM) which was polymerized in our laboratory and stored in the freezer, absorption bands of CO_2 at 2340 cm^{-1} were very large and saturated in transmission mode as is seen in Fig. 3. In order to study the effect of aging after polymerization, we measured spectra for different batches of CR-39 (BT), CR-39 (HM) and SR-90 (HM). From these measurements we have found that CR-39 and SR-90 contain a lot of CO_2 just after polymerization and that CO_2 escapes from the detector material by diffusion if the detector is stored at room temperature.

In the case of CR-39 (BT) samples which were exposed to 550 MeV/n Ar at a fluence of 1×10^9 ions/cm^2 and to 6 MeV/n Fe at a fluence of 1×10^{11} ions/cm^2, it was difficult to separate CO_2 produced by polymerization from CO_2 produced by irradiation. Consequently, it is worth mentioning that CR-39 or SR-90, which has been sufficiently aged in the air, should be used to detect a small absorption signal of CO_2 produced by irradiation.

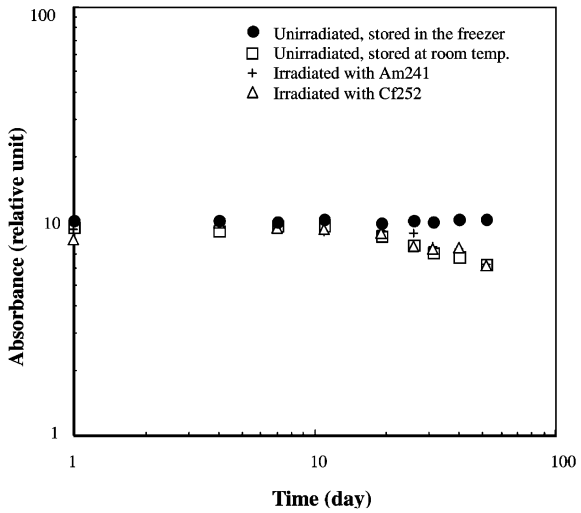

Fig. 4. Time variation of CO_2 absorption for four samples of SR-90 (HM):unirradiated and stored at room temperature, unirradiated and stored in the freezer, irradiated with alpha particles from 37 kBq ^{241}Am source, irradiated with alpha particles and fission fragments from 37 kBq ^{252}Cf source.

Fig. 4 presents the time variation of CO_2 absorption measured intermittently up to 52 days for four samples of SR-90 (HM): unirradiated and stored at room temperature, unirradiated and stored in the freezer, irradiated with alpha particles from 37 kBq ^{241}Am source, irradiated with alpha particles and fission fragments from 37 kBq ^{252}Cf source. In this figure, the sample stored in the freezer shows no change of CO_2 absorbance but absorbance for other samples shows similar decrease with time. This means that the amount of CO_2 decreases at room temperature even if the sample is irradiated with a comparatively weak radioactive source of 37 kBq. In order to accumulate CO_2 in the detector by reducing CO_2 diffusion, we tried to irradiate the sample CR-39 (IC) with alpha particles from 37 kBq ^{241}Am source in the freezer for 43 days, but 37 kBq was not strong enough to obtain CO_2 absorption signal.

One of the authors (M.F) has reported that the sensitivity of SR-90 slowly decreases if it is stored at room temperature (Fujii et al., 1993) and that CR-39 and SR-90 can be sensitized with an aging in CO_2 (Fujii et al., 1995, 1997). A graph in the reference (Fujii et al., 1993) that shows the aging effect on SR-90 is reproduced in Fig. 5. The time variation of sensitivity shown in Fig. 5 together with the variation of CO_2 absorbance shown in Fig. 4 suggests that the amount of CO_2 in the detector is closely related to the latent track formation. The fact that SR-90 contains much more CO_2

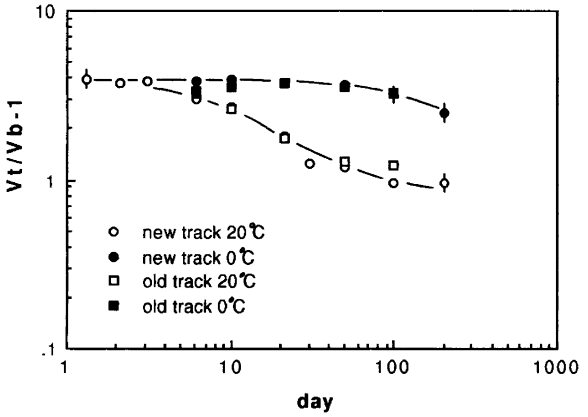

Fig. 5. Aging effects on SR-90 (Fujii et al., 1993). In this figure, the new track means that the test sample was irradiated with 6 MeV alpha particles just before etching and the old track, just after the polymerization.

than CR-39 just after polymerization is consistent with the fact that both the sensitivity and the aging effect of SR-90 are much larger than that of CR-39.

4. Conclusion

The results of our measurements can be summarized as follows: (1) In both CR-39 and SR-90, certain amount of CO_2 is produced in the polymer through the polymerization process. (2) The CO_2 escapes from the polymer by diffusion if the detector is stored in the air at room temperature. (3) Additional CO_2 is produced by radiation damage through the irradiation process. (4) The amount of CO_2 in the detector is closely related to the latent track formation and to the track detection sensitivity.

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