Effect of Heavy Ion Irradiation on Sucrose

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Heavy ion irradiation of sucrose produces stable free radicals. The relative spin concentration obtained after the heavy ion irradiation increases linearly as the absorbed dose increases. It decreases logarithmically as LET (linear energy transfer) increases. Production of the relative spin concentration of He (helium) ion irradiated specimen was two times more dependent on LET than that for C (carbon) ion irradiation.

Irradiation effects induced by various irradiations on materials are important subjects of radiation research. The results will lead to a wide range of applications, such as clinical usage, a dosimeter for cosmic rays, etc. Especially, effects caused by heavy ion particles are of interest. Heavy ion particles exert much larger impulses on the orbital electrons of molecules of a material, causing excitations and ionizations. Free radical production could be used as an indicator of heavy ion exposure.

Sucrose is household sugar and has been known to form stable radicals by γ- and X-ray irradiations at ambient temperature.[1] Electron paramagnetic resonance (EPR) spectroscopic technique is the most reliable method to measure free radicals. Irradiated sucrose has been studied by various EPR techniques in order to specify the radical sites.[2-4] These studies were focused on single crystal irradiation at low temperature. Recently, EPR measurements with high doses were reported (0.01 - 4 [kGy]).[5] However, there have been no reports regarding heavy ion irradiated sucrose at ambient temperature.

We studied sucrose in order to examine the effects of heavy ion irradiation. EPR was used to estimate (or yield) a number of unpaired spins in sucrose after the irradiation. Sucrose radicals produced by the irradiation as a function of linear energy transfer (LET) were analyzed. LET and heavy ion particle dependence for production of the relative spin concentration was also examined.

Sucrose was purchased from Nacalai Tesque Inc. Glucose and fructose were obtained from Tokyo Chemical Industry Co. Ltd. Spin label, 2,2,6,6-tetramethylpiperidine-1-oxyl (TEMPO) was obtained from Aldrich, USA. All chemicals were highest grades and were used as received.

Irradiation by heavy ion beams was performed in a biology experiment room of Heavy Ion Medical Accelerator in Chiba (HIMAC) at the National Institute of Radiological Sciences (NIRS). Heavy ion beams of helium (150 MeV/u) or carbon (290 MeV/u) from HIMAC were used for the present study. A binaryfilter, composed of plates made of poly (methyl methacrylate) with various thicknesses from 0.5 mm to 128 mm, was used to adjust LET.

Amorphous solid sucrose was attached on an acrylic plate (4 x 4 cm^2) and wrapped with a plastic wrap. The wrapped samples were mounted in the sample holder. In addition to heavy ion irradiation, γ-ray irradiation (Co-60) of sucrose, glucose and fructose were performed at the Tohoku University irradiation facility for comparison.

Sucrose radicals produced by heavy ion irradiation were measured using an X-band EPR spectrometer. The resonance frequency was measured using X-band microwave frequency counter CEM-14 (Echo Electronics Co. Ltd., Japan). Relative spin concentrations were estimated using a calibration curve for known concentration of TEMPO in benzene.[6] All processes of irradiation and measurements were carried out at ambient temperature.

The EPR spectrum of sucrose radicals formed by helium (He) ion irradiation is presented in Figure 1. The identical spectra for various irradiated samples were measured after one year, but the intensities decreased a few percentages of them when the samples were kept in EPR tubes with the caps. The EPR spectrum is relatively broad (~7 mT wide) with asymmetry at the center. In general, powder EPR spectra of radicals show a symmetrical pattern. The asymmetric pattern suggests that there are several radical sites in the irradiated sucrose.

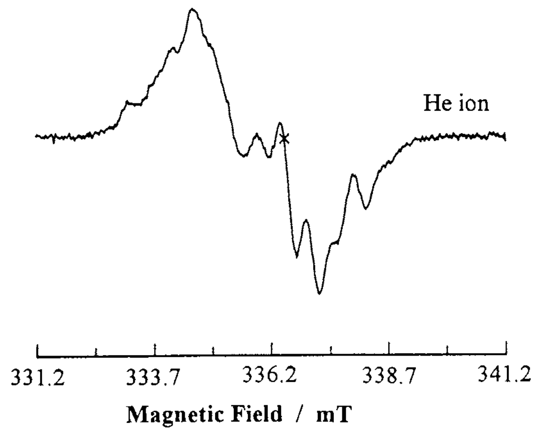

Figure 1. EPR spectrum of He ion [150 MeV/u] irradiated sucrose. The cross indicates g = 2.003. The typical EPR settings were the following: sweep width, 10 mT; time constant, 0.1 s; modulation, 0.03 mT; receiver gain, 1000; microwave power, 5 mW.

In order to examine the detailed effects on sucrose of various heavy ion particles and LETs, we performed experiments on He and carbon (C) ion irradiation as a function of LET. First, similar EPR spectra were observed for both particles and various LETs. Although the EPR spectra were similar for both He and C ion irradiation, the kinetics would be different to produce the stable radicals in the sucrose. Second, the relative spin concentration decreased logarithmically as LET increased as shown in Figure 2. The following empirical relation was obtained:

$$Y = -A \ln(\mathrm{LET}) + B \quad (1)$$

where Y is relative spin yield, A is slope, and B is interception.

The slope for He ion is –5.2 x 10^{18} and –2.4 x 10^{18} for C irradiation. Production of the stable radicals for He irradiation was strongly dependent on LET in relation to C ion irradiation even though the particle energy was about half of the C ion. Sucrose responded well to various LETs of heavy ion irradiations in terms of the steady state spin concentration. This is the first clear effect regarding heavy ion irradiation of the solid organic sample.

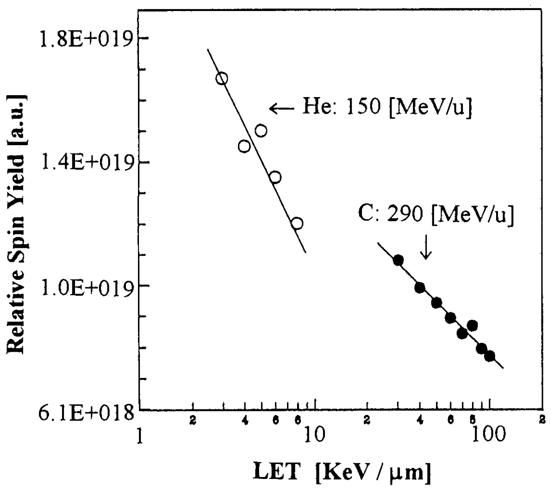

Figure 2. The relative spin concentration versus LET for He ion [150 MeV/u] and C ion [290 MeV/u] irradiation. The dose was about 50 Gy.

Next, we examined the EPR intensity change for different doses. The EPR intensity at various doses of He ion irradiation linearly changed. The slope for the irradiation at LET 2.2 [KeV/µm] is ~4.8. A number of spins (or EPR intensity for the sucrose) increases linearly as the dose increases. Also, linear dependence was obtained for C ion irradiation. Thus, sucrose can be a good candidate to monitor the effects of heavy ion irradiation at a certain dose.

In order to specify the radical sites in the sucrose, we investigated glucose and fructose irradiated by γ-rays. Although glucose and fructose do not fully explain the chemical nature of sucrose, both compounds will provide information regarding hyperfine structures obtained. Preliminary analyses showed that the added spectrum of irradiated glucose and fructose was reasonable agreement with that for sucrose. The noticeable difference is less intense at the central region. There may be conformation differences that are changing hyperfine coupling. Reproduction of the EPR spectrum shown in Figure 1 suggests that the sucrose radicals are composed of glucose and fructose radical sites. The EPR results suggest that a possible radical site can be carbon-centered radicals. These radicals have normally large hyperfine couplings in irradiated organic solids.[7] Two carbon-centered radicals were also suggested by the single crystal studies of irradiated sucrose.[2,4]

In conclusion, the present results concerning the heavy ion irradiated sucrose showed the following: First, the sucrose radicals are very stable at ambient temperature. Second, the relative spin concentration has the logarithmic relation to LET at a certain dose. Third, production of sucrose radicals was dependent on LET and heavy ion particles. The spin yield for He ion irradiation showed strong LET dependence in comparison with that for C irradiation. The results are the first clear effects with respect to heavy ion irradiation of sucrose. Fourth, the EPR intensity increases linearly as the corresponding dose increases. We further investigated the radical sites in the sucrose by analyzing structural constituents: glucose and fructose.

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