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Sucrose radical-production cross section regarding heavy-ion irradiation

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Abstract: We investigated the sucrose radical-production cross section induced by heavy-ion irradiation. L-alanine was also used in order to compare radical yield and cross section. The stable free radicals after irradiation were analyzed by EPR (electron paramagnetic resonance). The radical yield obtained by the irradiated samples had a logarithmic correlation with the LET (linear energy transfer). Quantitative EPR analyses showed that radical productions for sucrose and L-alanine vary both by different particle irradiation and the LET under the same absorbed dose. Furthermore, the cross sections of radical productions for samples were calculated. Both cross sections for C ions irradiation under LET 30 [keV/μm] at 50 Gy dose were ~3.0 x 10^{-9} [μm^2], taking account of the molecular areas of the samples. The values of the cross sections imply that multiple ionizing particles involve producing stable radicals.

1. Introduction

Intermediates (radicals) induced by particle-material interactions are of fundamental importance for radiological sciences. The interactions of ionizing particles with matter cause excitations, ionizations, and production by secondary electrons. Different types of ionizing radiation have different track structures when passing through material [1]. Secondary electrons are mainly thought to produce free radicals at the target sites. The difference in the radical production can be attributed to the difference in the track structure. The radicals produced as a result of particle-matter interactions can be investigated by Electron Paramagnetic Resonance (EPR) spectroscopy.

It is known that sucrose irradiated with various types of irradiation becomes stable (long-lived) free radicals. Studies on the fundamental and practical aspects of sucrose radicals have been made by various research groups [2-10]. EPR investigations of irradiated monosaccharides showed multi-radical productions [2-5]. Multi-radical productions were also experimentally proved by Nakagawa and co-workers [6]. The detailed structures of fructose radicals were further investigated by DFT (density functional theory) [3]. An original EPR investigation of sucrose irradiated with heavy ions showed that the radical yields (sucrose radicals) correlated with the logarithmic LET (linear energy transfer) [7-11]. It was also found that the production of sucrose radicals with helium ions was the most sensitive to the LET among several heavy-ion species examined. Moreover, there have been limited EPR investigations of sucrose irradiated with heavy ions under low-dose conditions [9, 10].

We studied polycrystalline sucrose and L-alanine in order to examine the detailed interaction with ionizing particles. The radicals produced by heavy-ion irradiation were obtained as a function of the LET and the absorbed dose. L-alanine was also used in order to compare radical yield and cross section for different heavy-ion irradiation. Furthermore, the yields of sucrose and alanine radicals were considered in terms of the radical-production cross section, which will be useful parameter for further investigations of the stable radicals.

2. Experimental section

2.1. Sample preparations

Sucrose (Nacalai Tesque grade) and L-alanine (99.0%) were purchased from Nacalai Tesque Inc., Japan and used as received. The molecular wights of sucrose and L-alanine are 342.3 and 89.09, respectively. Each polycrystalline sample (0.5 g weighted) was placed on an acrylic plate (4 x 4 cm^2) and wrapped with a thin plastic sheet. The typical thichness of the sample was ~1.0 mm. The wrapped samples were mounted on a sample holder for irradiation [9].

2.2. Irradiations

Sample irradiation by various heavy ions was performed in a biology experiment room of the Heavy Ion Medical Accelerator in Chiba (HIMAC) at the National Institute of Radiological Sciences (NIRS). Both samples were placed inside 10 cm diameter of the irradiation field, and uniformly and simultaneously irradiated. Heavy-ion beams from HIMAC with the extracted energy of carbon [290 MeV/nucleon: MeV/u], silicon [490 MeV/u], neon [230 MeV/u], and argon [500 MeV/u] were used for the present EPR study. A binary filter, made of poly(methyl methacrylate) with various thicknesses, was used to reduce the beam energy within the sample. The radiation dose was 50 Gy at the sample. The details of the sample irradiation are described elsewhere [6-10]. It is noteworthy that the experimental errors were described in the previous repots [8, 10].

2.3. EPR measurements

After irradiation, the samples were put into standard EPR tubes (o.d., 4.7 mm, i.d. 3.6 mm, JEOL Datum Co., Japan). The produced radicals were measured using a JEOL FE 1X 9 GHz EPR spectrometer. A sample quantity of 0.5 g in the EPR tube filled the TE_{011} cavity. The EPR measurements were performed at least one day after irradiation. The identical EPR conditions were used for both samples. The EPR settings were the following: sweep rate, 12.5 G per min; time constant, 0.1 s; modulation, 0.32 G; receiver gain, 1000; microwave power, 5 mW. The resonance frequency was measured using a microwave frequency counter, EMC-14 (Echo Electronics Co., Ltd., Japan). All processes of irradiation and measurements were carried out at ambient temperature. The details of the EPR measurements are also described elsewhere [6, 7-10].

3. Results and discussion

3.1. EPR of sucrose and L-alanine irradiated with particles

Figure 1 shows the EPR spectra of sucrose irradiated with Si ions under LET values of 60 and 80 [keV/μm] at a dose of 50 Gy. The experimental EPR conditions were the same for both values. The observed EPR intensity decreased as the LET increased. The obtained spectral patterns were identical for those previously reported [6, 9, 10]. The EPR pattern suggests that the radical sites are the same for the different LET. The EPR spectrum of the irradiated alanine also showed a similar tendency as a function of LET.

Figure 2 and 3 represent the spin yields for sucrose and alanine produced by heavy-ion irradiation as a function of LET, respectively. We found that sucrose and alanine show a LET dependence as well as particle dependence [6-10]. The number of the spin yields in the irradiated samples decreased with increasing LET value for each heavy ion. It is noted that the number of alanine molecule was ~3.8 times higher than that of sucrose.

The spin-yield for various heavy ions was analysed in the following manner. An empirical relation between the spin-yield and LET was expressed by [6]

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

where Y is the relative spin-yield [spins/g], A is the slope, and B is the interception. The steeper slope indicates more sensitive radical production. Moreover, the slope gradient for sucrose is not the same for both irradiation-particle types. Ne ions produce more stable radicals than C ions under the same LET and dose. This tendency is consistent with the previous result [9, 10]. Considering the resultant radicals for a single molecule, the production of sucrose radicals might be similar sensitivity to that of alanine radicals. Under the same conditions, a higher particle velocity produces a large radial distribution of energy deposition because the secondary electron increases with an increase in the particle velocity. The particle velocity (v) can play an important role for the radical production since the LET is proportional to (z^2/v^2) as previously reported [10]. It is notable that the charge (z) of the ionizing particle is also important for the impact of heavy ions on the organic solid sample. However, there are too many difficulties in estimating the effective charge of the particle [1]. Hence, sucrose molecules can be sensitive to the irradiation-particle type for radical productions.

3.2. Radical-production cross section

In order to further analyze the details of the radical production induced by the particle-material interaction, we can examine the interaction using a newly introduced parameter of radical-production cross section (σ).

The yields (Y) by ionizing particle irradiation can be empirically obtained using the radical-production cross section. The yields can be expressed by

$$Y = \left[1 - \exp(-\sigma N t)\right] \bullet \Phi, \tag{2}$$

where Y is the yield produced by the ionizing particle interaction, σ is the radical-production cross section, N is the number of molecules per unit volume, t is the sample thickness, and Φ is the number of trajectories. For a single particle, one can reduce the equation (2) to

$$P \cong \sigma N t, \tag{3}$$

where P is the number of radicals produced by a single traversing particle. The equation explains the number of radicals produced by a single particle. The σ-values obtained for sucrose and alanine were ~3.0 x 10^{-9} [μm^2] for C ion irradiation under LET 30 [keV/μm] at a dose of 50 Gy.

Next, radical-production cross section (σ) considering a target molecule and an ion track for the heavy-ion irradiation can be theoretically calculated by

$$\sigma = \pi (R + t)^2, \tag{4}$$

where R is the radius of a target molecule and t is the radius of an ion track. Figure 4 shows a schematic description of a target molecule and an ion track for the particle irradiation. The σ -value of this model calculation can predict possibility of the stable radical production by heavy-ion irradiation.

The molecular areas for sucrose and alanine are on the order of 10^{-19} [m^2]. The empirical σ-value of ~3.0 x 10^{-9} [μm^2] for the samples is smaller than those of the molecular areas. Both σ-values are smaller than the molecular areas by one order of magnitude, suggesting that stable radicals are produced via ionizations of a single molecule by several particles. Taking the difference in the size of the molecules into account, one can find that the sensitivity is roughly the same for these two compounds, regarding the production of stable radicals.

We examined σ as LET increased. Figure 5 shows the σ obtained by C ion irradiation as a function of LET. The σ values increase as the particle LET increases. The result indicates that the cross-section increases when the energy deposition increases in the material at the target sites. It is notable that the finding is valid under low LET values of ~100 [keV/μm]. The analysis implies that the radical-production reaction undergoes in proportion to the energy deposition of incoming particles.

4. Conclusions

The current EPR results show that sucrose is a sensitive material for heavy-ion irradiations. Both sucrose and alanine showed satisfactory dependences for ionizing particle and LET. The newly introduced parameter, the so-called radical cross section, showed that both cross sections obtained for C ions were ~3.0 x 10^{-9} [μm^2] for LET 30 [keV/μm]. The present results provided that the cross-section increases as LET increases and several particles involve to produce the stable radicals.

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Figure Captions

Figure 1. EPR spectra of polycrystalline sucrose irradiated by Si ions under two different LET values of 60 and 80 [keV/μm]. Each spectrum was obtained for a dose of 50 Gy. The EPR conditions were the same.

Figure 2. Plot of the relative spin-yield for sucrose as a function of the particle LET. The yield was obtained by the EPR spectra of irradiated polycrystalline sucrose sample using various particles. The C (filled circles), Si (filled diamonds), Ne (open circles), and Ar (open squares) ions were used as particle irradiations. Each data point corresponds to a dose of 50 Gy.

Figure 3. Plot of the relative spin-yield for alanine as a function of the particle LET. The yield was obtained by the EPR spectra of irradiated polycrystalline L-alanine using various particles. The C (filled circles), Si (filled diamonds), Ne (open circles), and Ar (open squares) ions were used as particle irradiations. Each data point corresponds to a dose of 50 Gy.

Figure 4. Schematic illustration of a target molecule and an ion track for particle irradiation. The radical-production cross section (σ) can be calculated by $\pi(R + t)^2$, where R is the radius of a target molecule and t is the radius of an ion track.

Figure 5. Plot of the radical-production cross section (σ) as a function of the particle LET. The C ions were used as particle irradiation. Each data point corresponds to a dose of 50 Gy.

Figure 1.

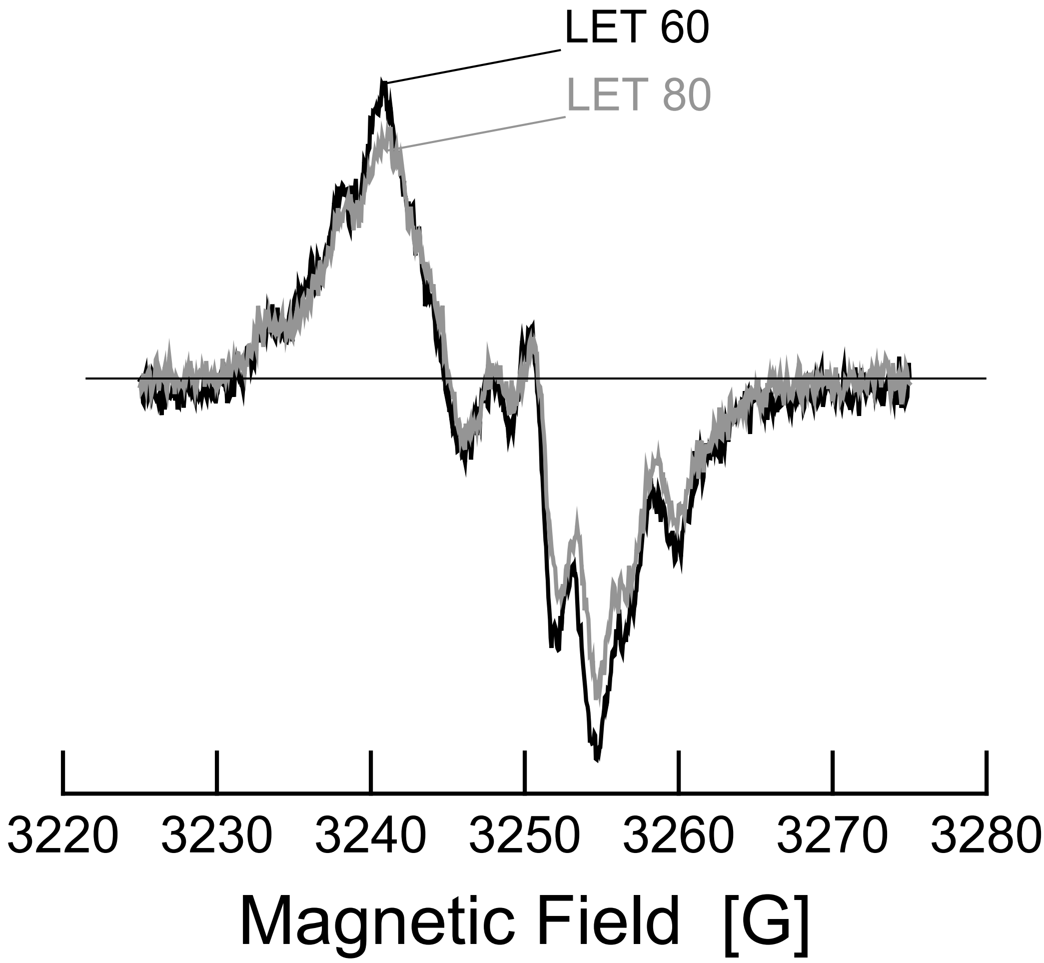

LET 60
LET 80
3220
3230
3240
3250
3260
3270
3280
Magnetic Field [G]

Figure 2.

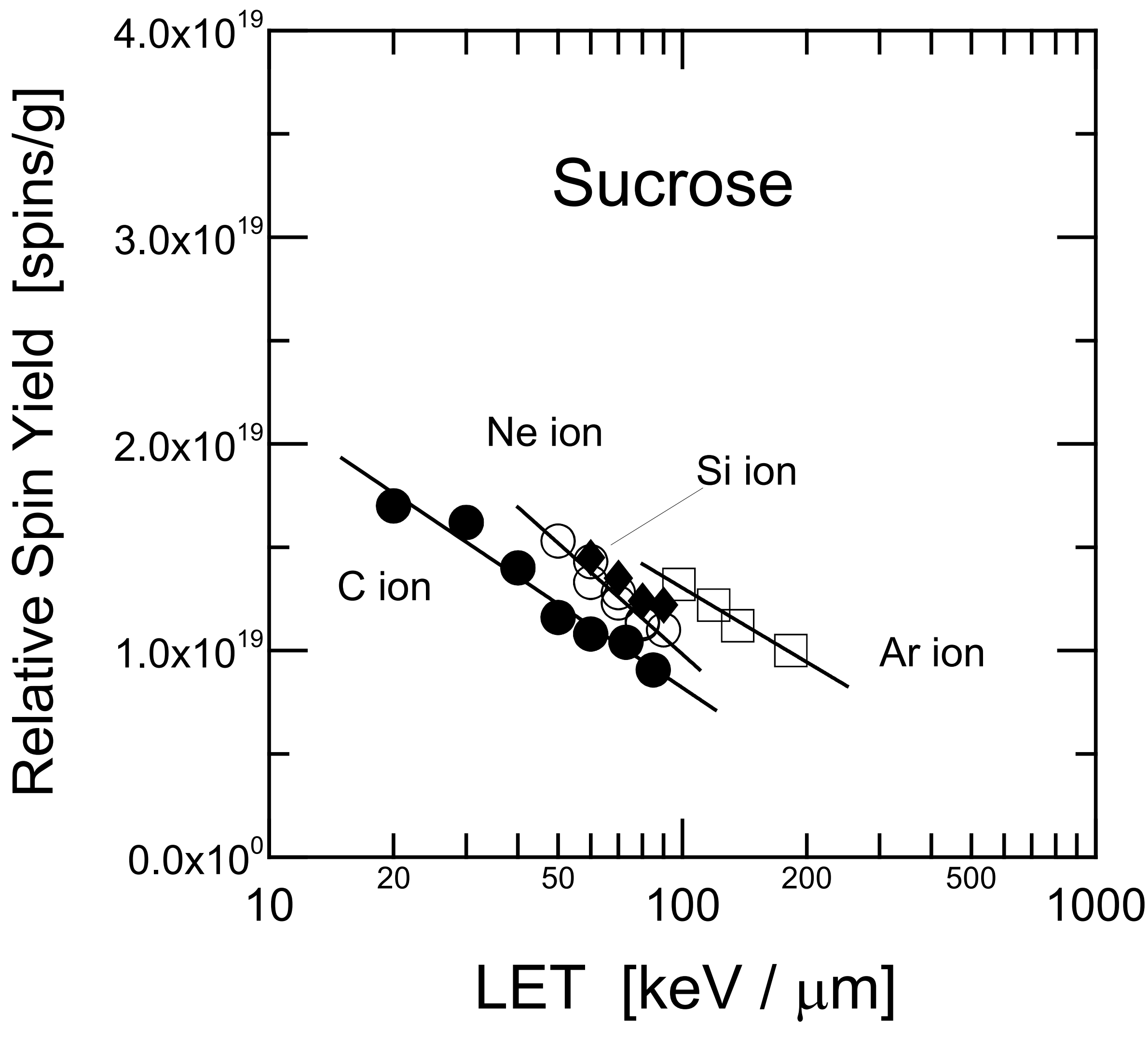
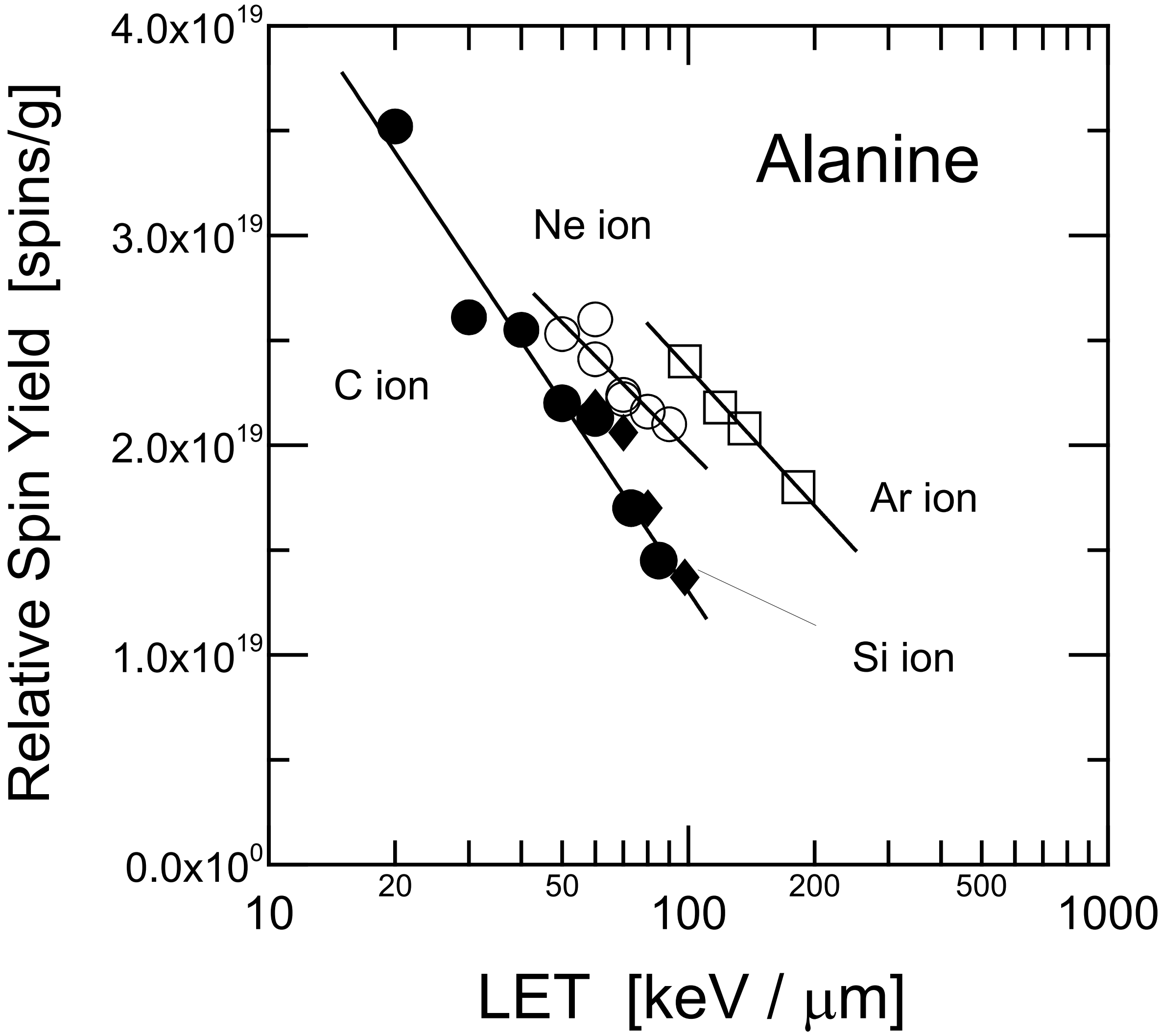

Sucrose
Relative Spin Yield [spins/g]
4.0x10^19
3.0x10^19
2.0x10^19
1.0x10^19
0.0x10^0
Ne ion
Si ion
C ion
Ar ion
20
50
200
500
10
100
1000
LET [keV / μm]

Figure 3.



Alanine
Ne ion
C ion
Ar ion
Si ion
4.0x10^19
3.0x10^19
2.0x10^19
1.0x10^19
0.0x10^0
20
50
200
500
10
100
1000
Relative Spin Yield [spins/g]
LET [keV / μm]

Figure 4.

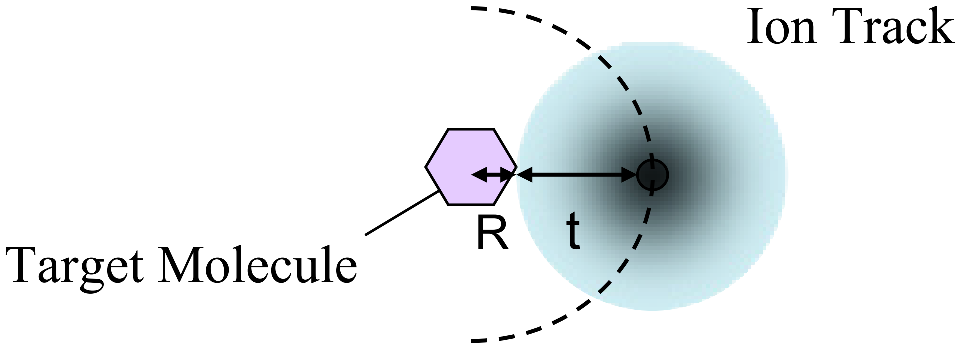
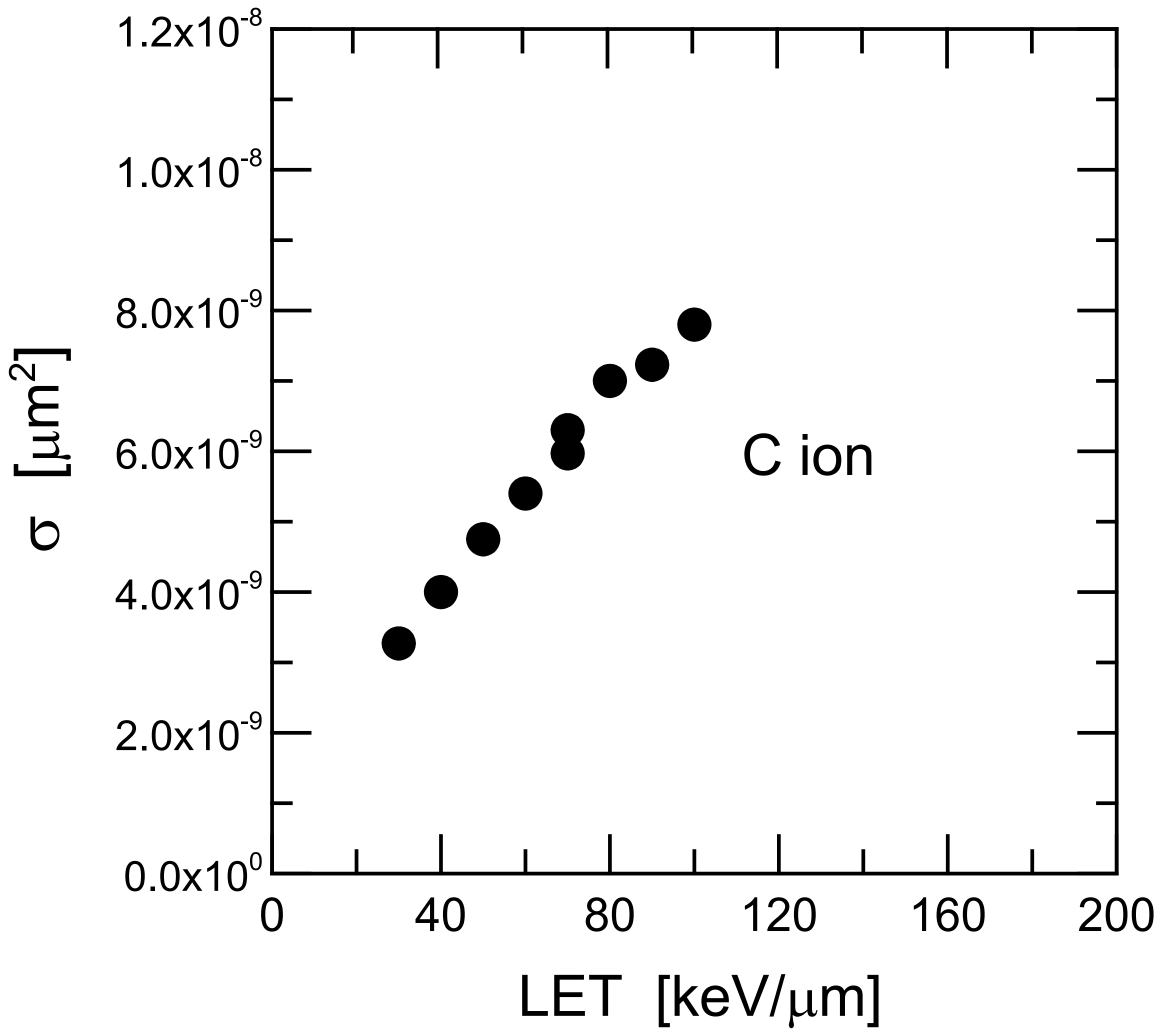


Figure 5.



1.2x10-8
1.0x10-8
8.0x10-9
6.0x10-9
4.0x10-9
2.0x10-9
0.0x100
σ [μm2]
C ion
0
40
80
120
160
200
LET [keV/μm]