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Investigation of sugar irradiated with He, Ne and C ions for dosimetric purposes

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Electron paramagnetic resonance (EPR) study on solid sugar irradiated by He, Ne and C ions and UV spectrometric study of its water solutions are presented. The samples are treated with different doses of radiation in the region of 50 - 300 Gy and several values of linear energy transfer (LET). All samples exhibit identical EPR spectra, due to radiation induced stable sugar radicals, and UV absorption at 267 nm of their water solutions. The EPR signal amplitude (peak-to-peak of the first derivative) and the UV absorption at 267 nm are used as dosimetric indexes. The dependence of these parameters as a function of LET and absorbed dose radiation is studied. The present results show that the EPR response with increase of LET of a given particle decreases, while the UV absorption at 267 nm increases. In addition, the new relation between LET of irradiating particles and UV absorbance is obtained.

Keywords*:* Sugar; irradiation; heavy ion; EPR dosimetry; UV spectrometry

INTRODUCTION

In the last decades, the extensive use of ionizing radiation in various fields of human activities requires effective radiation protection and reliable dosimetric systems to estimate the received radiation doses. On the other hand, it is well known that treatment of sugar with various types of radiation generates stable long-lived free radicals [1, 2, 3, 4, 5, 6, 7, 8, 9]. The application of heavy particles in medicine leads to the investigation of sugar in the last two decades as a dosimetric material for heavy particles irradiation [6, 10, 11, 12, 13, 14, 15, 16, 17, 18]. It is found that identical EPR spectra of radiation induced stable free radicals are obtained both with photons and heavy particles.

On the other hand, the irradiation of sugar creates products of radical recombination besides paramagnetic species. These recombination products are used in solid state/UV dosimetric system [19] and aqueous solutions/UV dosimetric system [4, 5, 20, 21, 22, 23]. A characteristic band at 267 nm is appears in aqueous solutions of irradiated sugar [4, 5, 21] or at 263 nm as reported by other authors [7].

This new application of UV spectrophotometry is very important because it is a calibrated method. Thus, before EPR dose estimations each spectrometer has to be calibrated by a separate calibration graph for each batch of samples, which is only valid for the appropriate EPR spectrometer and laboratory. In order to overcome the above disadvantage, an attempt was recently made to calibrate EPR using UV spectrophotometry [22]. By this approach it is possible to calibrate not only sugar, but also other, for example alanine dosimeters, if they are simultaneously irradiated.

In the present paper, are reported the first results on the effect of LET of the He, Ne and C ions on the UV response. The relation between UV absorption and LET of the irradiating particles is reported.

EXPERIMENTAL

Materials and sample preparation

Sucrose (sugar) of the highest grade is purchased from Nacalai Tesque Inc., Japan and used as received. Sugar sample weight of 0.50 g is used in order to fill the EPR cavity. The same sample is then used for preparation of 5% water solution for UV spectrophotometry.

Each sugar sample is placed before irradiation on an acrylic plate (40 mm x 40 mm) and wrapped with a thin plastic sheet. The thickness of the sample is approximately 1.0 mm. The wrapped samples are mounted on a sample holder for irradiation.

Heavy-ion irradiation

Heavy-ions sample irradiation is performed in a biology experiment room of a heavy ion medical accelerator in Chiba (HIMAC) at the National Institute of Radiological Sciences (NIRS). The biology room was equipped with an irradiation system similar to that found in a treatment room, including dose monitors, a binary filter, and a wobbler system. A target in the atmosphere was placed at a distance of 50 cm from a thin aluminum window that sealed the vacuum in the beam ducts. The wobbler system realized a 10 cm diameter uniform field with a uniformity of 2% or less. The beam intensity was measured using dose monitors

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installed in the beam course. A binary filter composed of poly (methyl methacrylate) plates with thicknesses ranging from 0.5 to 128 mm was used to adjust the LET. Residual ranges of the beams, when plates of various thicknesses were inserted, were measured precisely before the irradiation. LET values at the target position were estimated by a simulation code using the data of residual ranges [24]. The sugar samples were irradiated from 5 to 300 Gy with follow ions and LET: 1) with helium (150 MeV/u with LET 3, 4, 5, 7.9 and 10 keV/µm); 2) with neon (400 MeV/u with LET 30.9, 50, 60, 80, 100.9, 119.7, 120 and 139 keV/µm) and 3) with carbon (290 MeV/u with LET 10.9, 20.5, 30.2 and 49). All processes of irradiation and measurements are carried out at ambient temperature. A schematic illustration of the experimental setup for sample irradiation is presented in Figure 1.

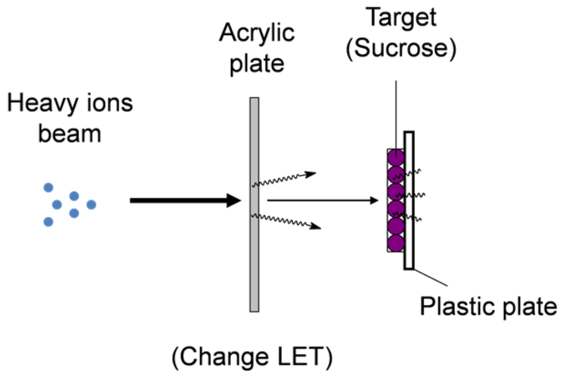

Fig. 1. Schematic illustration of the experimental setup for irradiation with heavy ions.

Instrumentation and procedures of measurements

The EPR spectra are recorded on a JEOL JES-FA 100 EPR spectrometer operating in the X–band with standard TE_{011} cylindrical resonator at room temperature. After irradiation the crystal sugar is directly transferred to EPR quartz sample tube with 5 mm o.d. and 4 mm i.d. and fixed in the cavity center. The EPR signal intensity i.e. peak-to-peak of the first derivative in the EPR spectrum is used as a measure for the quantity of radiation generated free radicals. Eight to twelve EPR measurements are performed for each irradiation dose and the results are averaged.

UV measurements are performed on a Specord UV-VIS (Carl Zeiss, Jena) spectrophotometer at room temperature. All UV spectra of water solutions of irradiated sugar are recorded versus distilled water. At the same conditions non-irradiated sugar has no absorption in the studied UV region (240-360 nm). All water solutions after preparation are heated up to 70°C for 1 h to reach stable with the time UV absorption [21]. Quartz sample cells with a path length of 5 cm are used. Five measurements were made for every sample.

RESULTS AND DISCUSSION

EPR investigation of crystal sugar irradiated with He, Ne and C ions.

All sugar samples are EPR silent before irradiation. A typical signal due to radiation induced free radicals is recorded in the EPR spectrum after sample irradiation. The signal is complex and is a superposition of the spectra of different paramagnetic species [25].

The influence of the instrumental parameters (microwave power and modulation amplitude) on the EPR response is examined in order to record undistorted and maximal intense signal. For this purpose, the EPR spectra of irradiated sugar with Ne ions of 50 Gy dose and LET 50 keV/µm is performed in the region of 0.1 to 20 mW microwave power and 0.05 to 1 mT modulation amplitude. The experiments show that the influence of the above parameters on the intensity and the shape of the EPR signal are avoided when all EPR spectra are recorded at 1 mW microwave power and modulation amplitude of 0.4 mT. The EPR spectra of sugar irradiated with He, Ne and C ions are identical with those described in the literature [14, 17]. There is no difference in the shape of the spectra of irradiated with these ions samples except their intensities (number of radiation induced free radicals). The intensity of the observed EPR spectrum depends on the concentration of the radiation induced radicals. As the dose response is of paramount importance for any dosimetry method, are providing raw results of the measurements (Table 1) to enable an interested reader to see the statistical significance of the results. Ten measurements for every sample were used for statistical study. The relative standard deviation of replicate signals was 2.4, 5.5 and 4.7% for lowest dose helium, carbon and neon ions, respectively. For highest dose it is 1.35, 1.52 and 2.09%, respectively. These data also characterize the reproducibility of measurements at various radiation doses and irradiating ions.

Table 1. The experimentally obtained values from the EPR measurements and standard deviation.

irradiation	He		Ne		C	
Dose, Gy	5	50	5	300	5	300

I_1	73.42	450	18.25	1323	20.74	1779
I_2	71.25	452	19.03	1358	21.56	1765
I_3	75.32	455	21.55	1369	19.88	1788
I_4	74.05	460	20.19	1322	23.54	1757
I_5	73.23	442	21.66	1378	20.11	1778
I_6	70.21	458	18.47	1351	22.36	1745
I_7	73.23	446	17.55	1364	20.85	1784
I_8	74.25	452	20.18	1353	18.78	1698
I_9	70.15	446	21.06	1388	24.53	1752
I_{10}	73.21	459	22.18	1297	19.77	1744
Mean I_{pp}	72.832	452	20.012	1350.3	21.212	1759
Std. deviation	1.73399	6.09189	1.6141	28.24516	1.80136	26.78723

The scatter of doses determined with replicate samples is due to variation of the density of the samples and usually very small of the geometry and the position of the sample. Although the uncertainty of around 4-5% in the low doses is not sufficiently low for primary calibration, sugar can be successfully used in dosimetry. Linear increase in the EPR signal intensity with increasing absorbed dose of C, He and Ne ions in the investigated dose range is found. This statement is already known [12] and will not be discuss here.

The relation between the EPR response and the atomic weight of the irradiating ions is confirmed [16], the heavier ions in the order He < C < Ne giving lower EPR response. The irradiation of sugar samples with He, Ne and C of different LET shows in the present study that EPR signal intensity increases with decreasing LET of every ion. This tendency is in a good agreement with previous observations [6, 10, 11, 12, 13, 26]. Therefore, EPR spectrum of sugar is sensitive both to the weight of the particles and their LET.

UV investigation of He, Ne and C ions irradiated crystal sugar

As was mentioned above, water solutions of non-irradiated crystal sugar do not exhibit UV absorbance, whereas water solutions of irradiated crystal sugar show an absorption band at 267 nm [4, 5, 21, 17]. Figure 2 shows UV spectra of Ne and C ions irradiated crystal sugar. Samples of sugar irradiated with He ions do not exhibit UV absorbance.

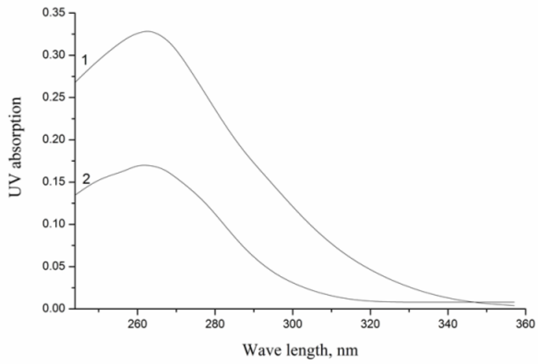

Fig. 2. UV spectra of 5% aqueous solutions of sugar samples irradiated with: 1) Ne (300 Gy, LET 50 keV/μm) and 2) C (300 Gy, LET 49 keV/μm) ions.

Previous UV spectrophotometric study of gamma irradiated sugar [21] shows that detection limit of c.a. 55 Gy may be obtained by using 5 cm sample cell and 20% solution. Having in mind that the highest He ion dose in the present study is 50 Gy with LET 10 keV/μm and in view of its relatively low atomic weight, the effect of He ion could be expected to be closely below that of gamma irradiation. Therefore, this explains absence of UV absorption in case of He ion irradiation. The appearance of 267 nm peak is due to recombination products of radiation induced free radicals in sugar. According to literature data [27, 28] absorption band in this spectral region is typically associated with carbonyl groups in organic compounds. It is assumed that glycoside bond breaks and the formation of carbonyl group may be common mechanism in the radiation chemistry of disaccharides [29].

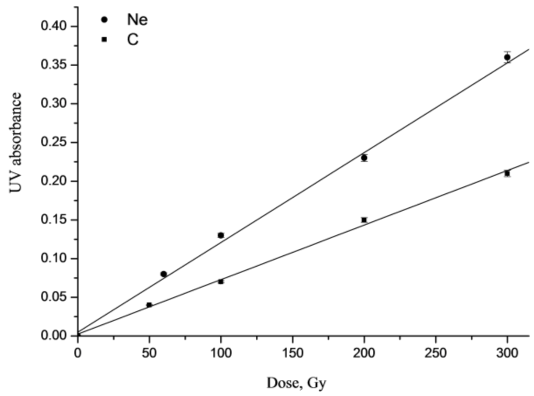

Fig. 3. The dependence of UV absorption of water solution of solid sucrose irradiated with Ne (LET 30.9 keV/µm) and C (LET 30.2 keV/µm) ions as a function of absorbed dose ionizing radiation.

The UV absorbance at 267 nm is used as a dosimetric index for the absorbed dose of photons and heavy particles [21, 14]. The dependence of the UV absorbance at 267 nm on the absorbed dose of C and Ne ions is shown in Fig. 3. As seen from Fig. 3, the UV absorbance intensity linearly increases, but with different slope for samples irradiated with Ne and C ions. In comparison with EPR UV results have better reproducibility and the standard deviation for low doses is 1%, whereas for high doses it is under 1%. The error bars represent a standard uncertainty in percent of five measurements for every sample.

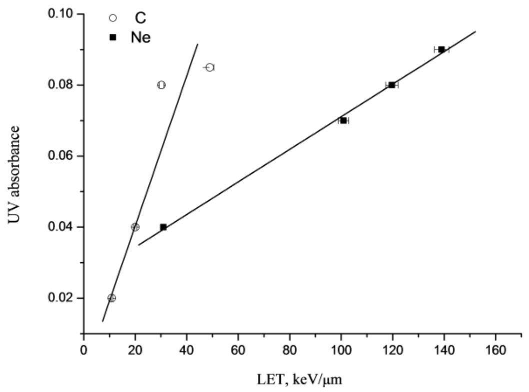

Fig. 4. Dependence of UV absorption maximum of aqueous solutions of solid sugar irradiated with dose of 50 Gy as a function of LET of Ne and C particles.

In addition, it is found that UV absorption intensity increases with increasing weight of the ions. In order to determine more deeply the effect of various LET of the particle on the UV absorption, the relation between UV absorption and LET was studied. The obtained results on Fig. 4 show that the UV absorption increases with increasing LET of the particle. This statement is contrary to the EPR result where EPR signal intensity decreased when LET of the particle increased. Since the absorption band depends on the particle species, their LET and the applied dose, to find the UV dose response of irradiated with heavy particles sugar have to take in account and LET.

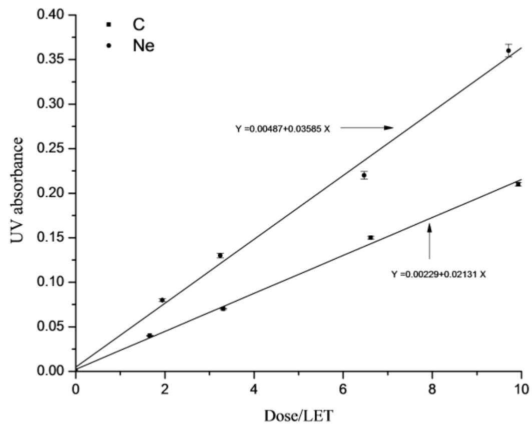

Fig. 5. UV absorbance at 267 nm as a function of the ratio of the absorbed dose and LET of the particles.

Figure 5 shows dependence of UV absorbance from the attitude of the dose and LET. As can see the linearity of the dose response is retained. Therefore in both cases with and without accounting LET the UV spectrometry may be used as a method for dosimetry of irradiated with heavy ions sugar because of the linear dose response.

Comparison between EPR and UV response of He, Ne and C ions irradiated sugar.

The relation between EPR and UV response for different heavy particles was discussed before [14, 16, 17]. However, the EPR and UV response of sugar irradiated with heavy particles with different LET is not discussed up to now. During irradiation, the effects that are measured with EPR and UV spectrometry are different. EPR measures free radicals whereas UV spectrometry measures some kinds of recombination products formed because of dissolving in water. Fact is that these effects are created from the irradiation with ionizing radiation. So this gives opportunity to compare both effects and methods. The dependence of EPR and UV response of sugar irradiated with Ne and C ions with LET 30 and 50 keV/µm shows a perfect linearity (Fig. 6).

Therefore, the linearity remains independently from LET and the particle species. Thus UV spectrometry could be used for calibration of EPR results when it is necessary.

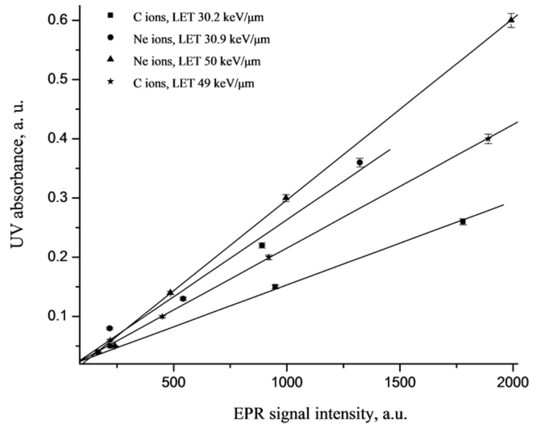

Fig. 6. Comparison between the results obtained by UV and EPR spectroscopy of sugar irradiated with Ne and C ions.

CONCLUSIONS

The UV spectra of water solution of irradiated with Ne and C ions solid sugar shows an absorption band at 267 nm. The intensity of this band linearly increases with increasing of the absorbed dose high energy radiation for samples irradiated with Ne and C. The samples irradiated with He ions do not exhibit UV spectrum due to the lower dose and LET of irradiation than the detection minimum. The relation between UV absorbance and LET of irradiating particles, studied for the first time in this paper, shows that the UV absorption increased with increasing LET of every particle. Taking in account the LET of the irradiating ions, the linear dependence of dose response do not change. Therefore, the effect of LET of the particle could be neglect.

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ИЗСЛЕДВАНЕ НА ЗАХАР ОБЛЪЧЕНА С He, Ne И С ЙОНИ ЗА ДОЗИМЕТРИЧНИ ЦЕЛИ

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(Резюме)

Представено е Електрон парамагнитен резонанс (ЕПР) изследване на захар облъчена с He, Ne и C йони и УВ изследване на водните и разтвори. Пробите са третирани с различни дози йонизиращо лъчение (50-300 Gy) и няколко стойности на линейно енергийния трансфер (ЛЕТ). Всички твърди проби показват идентични ЕПР спектри, дължащи се на радиационно индуцирани стабилни радикали в захар, а водните им разтвори - УВ абсорбционна ивица при 267 nm. Като дозиметричен индекс са използвани амплитудата на ЕПР сигнала (разстоянието от пик до пик в първата производна на ЕПР спектъра) и УВ абсорбцията при 267 nm. Изследвани са зависимостите на тези параметри като функция от ЛЕТ и абсорбираната доза радиационно лъчение. Представените резултати показват, че с нарастване на ЛЕТ на частицата, ЕПР отклика намалява, докато УВ абсорбцията при 267 nm нараства.