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Energy Response in Chemiluminescence Dosimetry with Sugar and Sorbite

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Abstract

A series of study on energy dependence in chemiluminescence dosimetry with sugar and sorbite produced in two different countries was carried out administering a dose of 5 Gy to the samples at six different mean photon energies of 30, 50, 80, 130, 662 and 1250 keV. The results revealed distinct energy dependence of chemiluminescence(CL) output of sugar and sorbite. Although the energy dependence, in general, could be fitted by a polynomial of *log E*, with *E* being radiation energy, up to cubic term, we reached a conclusion that the adoption of a fitting function, $y_R = a \cdot (1-e^{-b \cdot \log E})^c + d$, deduced from theoretical energy response curve calculated as the ratio of the mass energy absorption coefficients of the samples of interest to the soft tissue is more reasonable and rational. Here y_R is CL intensity, and *a, b, c* and *d* are constants to be determined in the fitting process. Energy dependence of relative sensitivities of one sample to the other, discrepancy in sensitivities of the samples from the two countries, and prominent grain size effect in Sorbitol were also shown.

Key Words : energy response, chemiluminescence dosimetry, sugar, sorbite, photon energy, retrospective dosimetry, dose reconstruction

1. Introduction

Many dosimetric studies on lyoluminescence (LL) and/or chemiluminescence (CL) performed previously can roughly be categorised as :

- LL and/or CL output and dose relationship studies for such substances as saccharides and sugar. [1~4]
- Studies for LL and/or CL enhancement used such sensitisers as terbium solution and eosin B or controlling solvent pH. [5~8]
- High dose dosimetry and its intercomparison

studies. [9,10]

In most of those studies emphases were basically put on LL(or CL)-dose relationship, regardless the quality of the radiation used. Excellent overview about the fundamentals in dose reconstruction using CL is given by Hammermeier et al.[11], Proschitzki et al.[12] and Koch et al..[13]

However, studies on the energy dependent characteristics of CL are hardly found in the published papers, except for those of Puite and Crebolder[14] and Ettinger et al. [15] , which briefly dealt with the energy response of LL from several saccharides as subsidiary subjects in their work.

Sugar and sorbite are well known to be suitable materials for CL as well as electron spin resonance (ESR) dosimetry, in particular, for retrospective dose evaluation in an emergency or accident situation mainly because of their tissue-equivalence in dosimetric characteristics. Due to the strong resemblance of these organic substances to that of human tissue in over-all chemical composition, the mechanisms of energy transfer and the energy dependence are very similar to that of the tissue. This study investigates the energy-dependent behavior of sugar and sorbite (sugar substitute) in CL dosimetry.

2. Materials and Methods

2.1. Chemiluminescence Device and Sample Material

The Berthold AutoLumat LB953 counter containing fully automated sample changer was used for CL analysis in this study. An ultra low noise photo multiplier (fast photon counter mode with 20 ns resolution) detects the CL intensity without a filter. The external computer was connected for controls and evaluation, and the software embedded is AutoLumat LB953 control version 1.04B.

Sarstedt polystyrol (PS) analysis test tube of 12 $\phi \times 55$ mm was used to put in CL readout sample.

The reagent solution for dissolving the sample was made of

Luminol (Sigma) : variable
Hemin (Sigma) : 1.25 mg (3.8 mM)
Na_2CO_3 (Merck) : 625 mg (11.8 mM)
Distilled water : ad 500 ml
1 M HCl was used to set pH value.

The analysis material was obtained in the retail or chemical trade. Sugar samples used are German sugar, white (GSW) (customary refined sugar in crystals, mean grain size 0.7 mm), and Korean sugar, white (KSW) (customary product). The trade names of the sorbite samples used are Sionon (trade name of Drufoga GmbH, Cologne, composition: sorbite 99.89%, saccharine 0.11%) and Sorbitol (D-Sorbite of Duksan Pharmaceutics, Korea, composition: sorbite 98.50%, sugar 1.28%, glucose 0.11%, sulfate + cloride 0.01%, other impurities 0.1%), respectively. Summation formula of sorbite is $C_6H_{14}O_6$ and molecular weight is 182.17.[16]

2.2. Irradiation Facilities

<u>X-ray unit</u>: X-ray unit of 420-keV Richard Seifert & Co was used in the mean energy range of 30 - 130 keV. Irradiation was carried out in round open polystyrol (PS) test tubes (diameter 5 cm, wall thickness 0.5 mm). The data referring to the individual irradiation mean energies are as follows;

30 keV : 48 kV, 19 mA, without filter, distance 51 cm, D = 0.1 Gy/min
50 keV : 100 kV, 25 mA, filter 0.2 mm Al, distance 65 cm, D = 0.4 Gy/min
80 keV : 180 kV, 17 mA, filter 0.25 mm Cu,

distance 65 cm, D = 0.4 Gy/min
130 keV : 250 kV, 15 mA, filter 1 mm Cu,
distance 65 cm, D = 0.4 Gy/min.

Gamma irradiation facilities: The gamma irradiation units used are located at the Research Centre of Environment and Health (GSF) in Neuherberg.

- A ^{137}Cs gamma irradiation facility of Hans Waglisch Muller provided the 662 keV irradiation energy at 0.8 Gy/min dose rate. During the irradiation process, two ^{137}Cs sources were conducted at the right and left side of the measurement chamber, in the center of which was previously positioned the sample material.
- The Gammacell 220 of Atomic Energy of Canada Ltd. has a ^{60}Co source with a mean irradiation energy of 1250 keV. When starting the test, the dose rate was 20 Gy/min. The measurement chamber containing the sample is moved into the homogeneous radiation field, being generated by cobalt sources arranged in ring-form. The samples were exposed to gamma radiation in glass vessels (diameter 2 cm, wall thickness 1 mm) and atmospheric oxygen could not be excluded during irradiation.

Administered dose: To each sample used in this study a fixed dose of 5 Gy of X- and gamma-rays mentioned above was given.

2.3. Chemiluminescence Measurement and Evaluation

The sample materials were used for irradiation tests without further pre-treatment. From the different materials, about 3 g each were irradiated and the same nonirradiated quantity was treated in parallel for control. In order to stabilize the samples due to the fading effect, the samples were kept at 60℃ for 48 hours. Heating of the sample material eliminates the unstable radicals in the irradiated material, so that a better constancy is to be expected in the measured value. Following this, the sample is weighed in PS test tubes which should be rinsed with methanol and well dried at 60℃. For each sample is made a 5-fold measurement, respectively. In the case of sorbitol, the grain size was separated in $\leq$0.4 mm and $>$0.4mm diameter by analytical sieve, because the inhomogeneity of the grain size leads to wide variations in the results. At the start of each measurement series, the zero value of the luminol solution was checked, i.e. 5 empty PS tubes are previously put into the sample chain before the series of the sample tubes. After they were inserted into the autoanalyser, 0.2 ml of luminol solution are injected and the analytical programme is started immediately. The light emission is measured for 4 seconds.

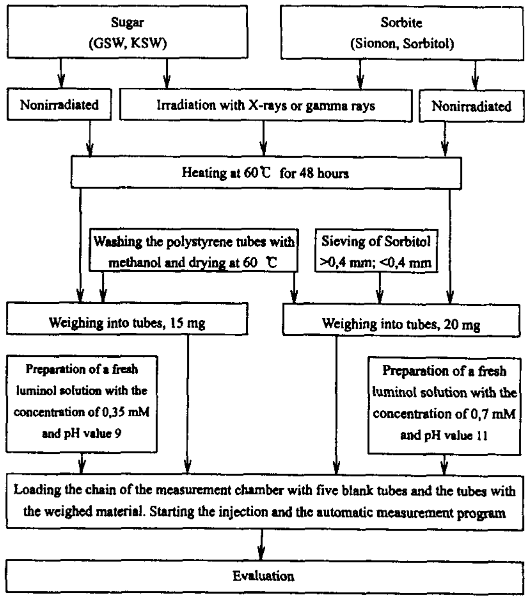

Fig. 1. Design for The CL Measurement Procedure Used in This Study

An essential factor in CL measurements is the preparation of the luminol solution. Its reaction is

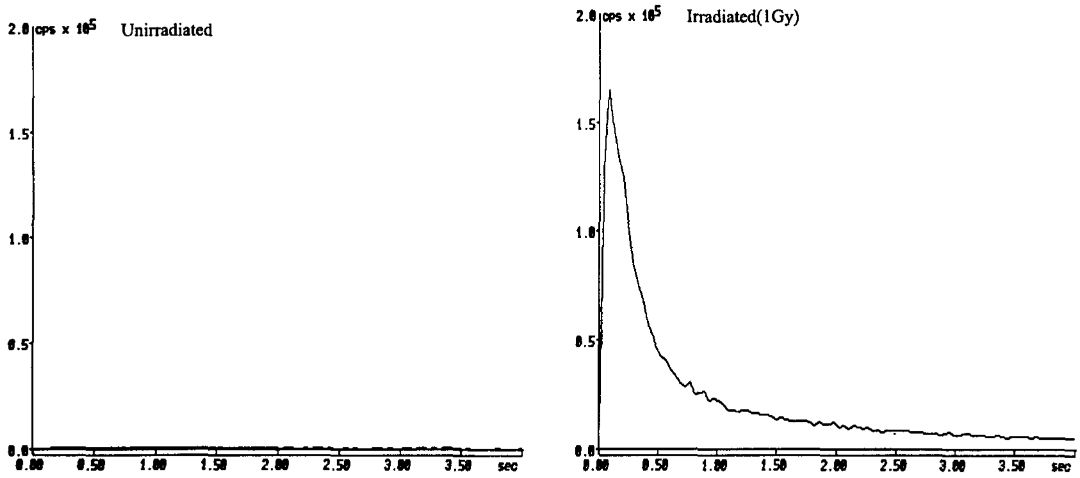

Fig. 2. CL Glow Curves of Nonirradiated and Irradiated (1 Gy) Sionon

very sensitive and to ensure reproducible results the solution have to be prepared freshly every day and the concentration should be kept constant and the storage conditions must be always the same.[13] The composition and the pH value for sugar and sorbite, as well as the weighed test portion, was optimized already in an earlier tests as follows ;

	Sugar	Sorbite (Sionon)
Weighed sample (mg)	15	20
Luminol concentration (mM)	0.35	0.7
pH value of the solution	9	11

The steps of the CL measurement procedure of this study are summarized in Fig. 1. The recorded light emission, projecting a glow curve due to the injection of Luminol to the sample, is the important measurement value. In Figure 2 is shown a typical glow curve of nonirradiated and irradiated (1 Gy) Sionon. The integral over the total or partial course of the glow curve is the measured CL intensity.

For sugar, the integral was evaluated from 0.4 to 4.0 seconds, because nonirradiated sugar shows a high CL intensity within the first 0.5 seconds. Integration of Sionon CL was made over the whole course of the curve until 4 seconds. From the CL intensity of the 5-fold-measurements was calculated the mean value and the single standard deviation from which result the indications of uncertainties.

3. Results and Discussion

3.1. Expression of Energy Response

Puite and Crebolder[14] and Ettinger et al.[15] were found to be the first who dealt briefly with radiation energy dependence of LL. The former authors have measured energy dependence of mannose at five different energies of X-rays down to 40 keV and one gamma-ray of ^{60}Co. They projected the results to the theoretical curve calculated as the ratio of the mass energy absorption coefficients of mannose and muscle. Ettinger et al. also have done similar work for several saccharides, including mannose and sucrose, and they pointed out the necessity of further study on the energy response.[15]

Among the samples used by the latter authors

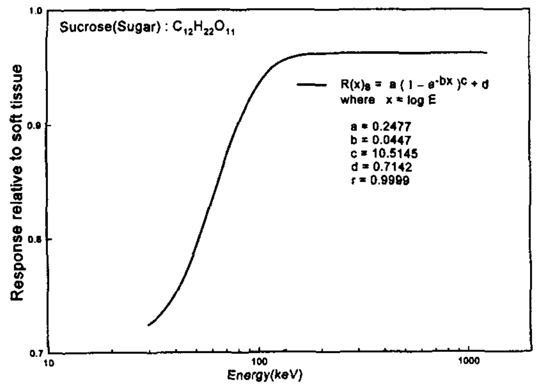

Fig. 3. **Energy Response Relative to Standard Striated Muscle Calculated as The Ratio of Mass Energy Absorption Coefficients of Sucrose(Sugar) and Standard.[15]**

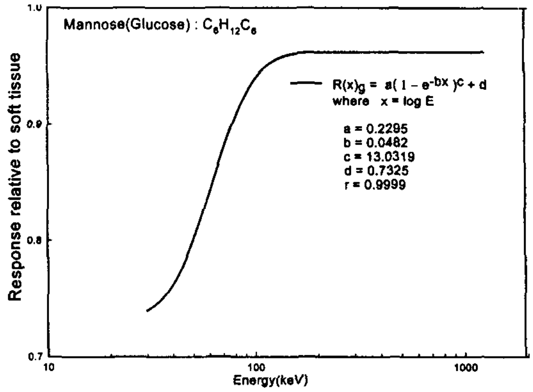

Fig. 4. **Energy Response Relative to Standard Striated Muscle Calculated as The Ratio of Mass Energy Absorption Coefficients of Mannose(Glucose) and Standard.[15]**

the sucrose (sugar) and mannose (glucose) are the subject of our particular interest because of their identity and similarity with our sugar and sorbite samples in atomic composition.

The theoretical energy dependence curves of sucrose and mannose were reproduced in this study and the best forms of fitting function found for both samples were as

$$R(x) = a \cdot (1 - e^{-bx})^c + d \quad (1)$$

with $x = \log E$,

where E is radiation energy in keV, and a, b, c and d are constants. As shown in Figs. 3 and 4, the functions $R(x)s$ and $R(x)g$ (suffix s and g represent sugar and glucose, respectively) are fitted with correlation coefficient r of 0.9999.

As described before, GSW, KSW, Sionon and Sorbitol were used in our energy response study. Sorbitol, in particular, was divided into two different categories of grain size, namely, smaller and larger than 0.4 mm in diameter in order to look for the grain size effect on the energy dependence as well as on the sensitivity. X-rays of four different mean energies, 30, 50, 80 and 130 keV, and the gamma-rays of ^{137}Cs (662 keV) and ^{60}Co (1250 keV) were used. Doses given are 5 Gy at each energy of photons. All the measured CL

Table 1. Energy Dependence of CL Intensities of Sugar and Sorbite*

Rad. Energy (keV)	GSW	KSW	Sionon	Sorbitol (<0.4mm)	Sorbitol (>0.4mm)
30	0.53 ± 0.04	0.50 ± 0.05	0.64 ± 0.03	0.63 ± 0.08	0.66 ± 0.06
50	0.54 ± 0.07	0.68 ± 0.07	0.66 ± 0.04	0.87 ± 0.10	0.96 ± 0.09
80	0.66 ± 0.07	0.69 ± 0.14	0.83 ± 0.03	0.89 ± 0.09	1.01 ± 0.13
130	0.66 ± 0.04	0.78 ± 0.05	0.95 ± 0.06	1.02 ± 0.10	1.07 ± 0.24
662	1.00 ± 0.05	1.00 ± 0.14	1.00 ± 0.03	1.00 ± 0.15	1.00 ± 0.10
1250	1.19 ± 0.13	1.10 ± 0.08	1.14 ± 0.03	1.28 ± 0.09	1.33 ± 0.13

* Irradiated dose : 5 Gy. CL intensities are normalized to unity at 662 keV.

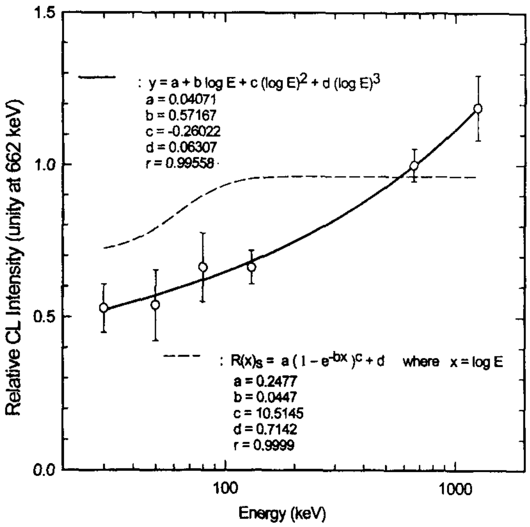

Fig. 5. Polynomial Fitted Energy Dependence of CL Intensity of GSW Irradiated to 5 Gy

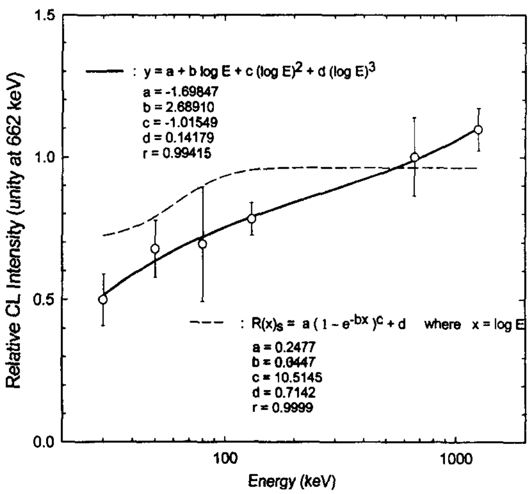

Fig. 6. Polynomial Fitted Energy Dependence of CL Intensity of KSW Irradiated to 5 Gy

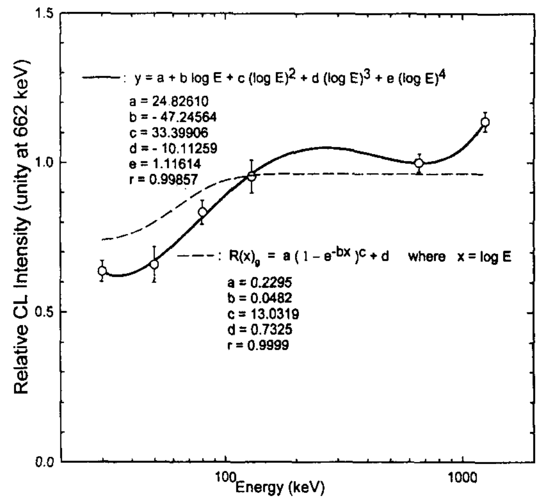

Fig. 7. Polynomial Fitted Energy Dependence of CL Intensity of Sionon Irradiated to 5 Gy

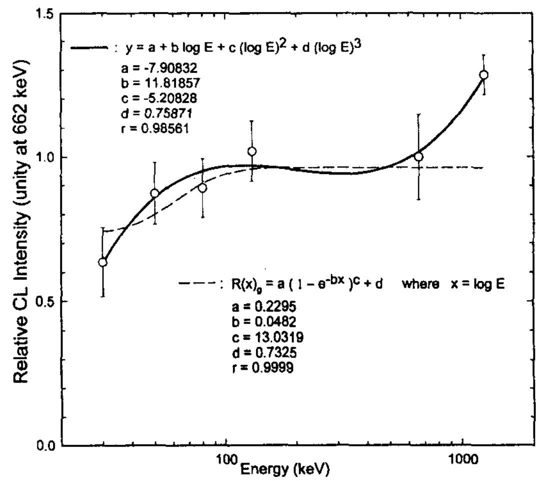

Fig. 8. Polynomial Fitted Energy Dependence of CL Intensity of Sorbitol (<0.4 mm) Irradiated to 5 Gy

output was normalized to unity at 662 keV. Results are summarized in Table 1 and graphically expressed in Figs. 5 through 14.

In the first five of those figures, the measured CL intensities were tentatively fitted to a polynomial of *log E* in order to make a comparison between these fitted to

$$y_l = \sum_{n=0,k=a} k \cdot (\log E)^n \qquad (2)$$

and those fitted to eq. (1).

Here $k = a, b, c, d \cdots$ are constants and E is

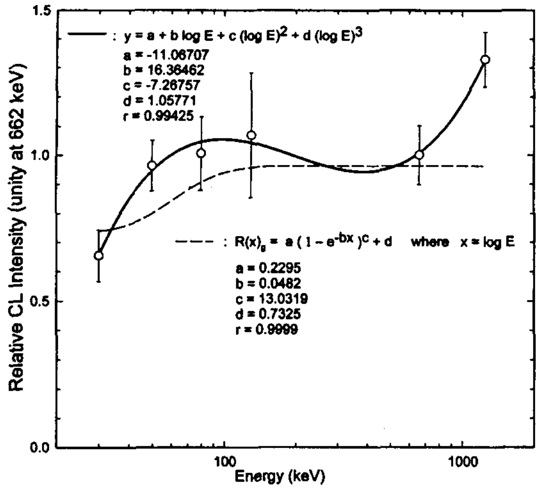

Fig. 9. Polynomial Fitted Energy Dependence of CL Intensity of Sorbitol (>0.4 mm) Irradiated to 5 Gy

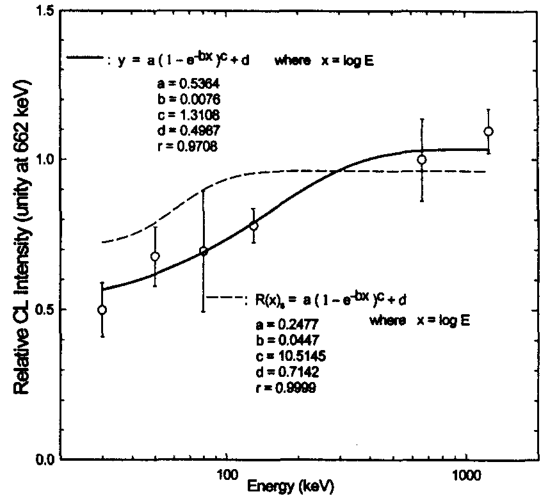

Fig. 11. Exponential Fitted Energy Dependence of CL Intensity of KSW Irradiated to 5 Gy

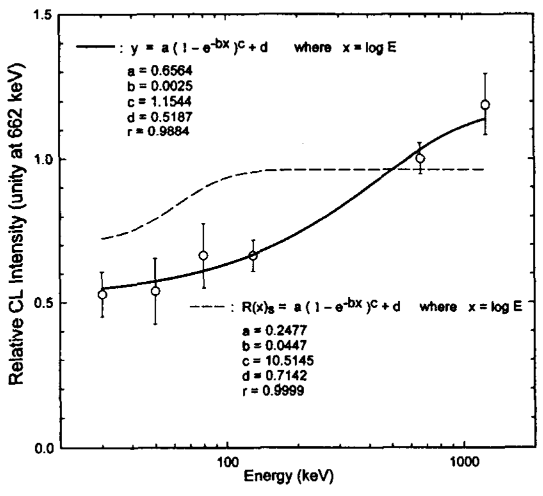

Fig. 10. Exponential Fitted Energy Dependence of CL Intensity of GSW Irradiated to 5 Gy

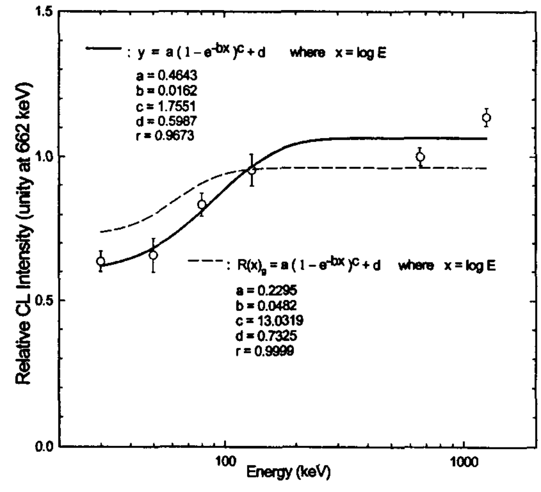

Fig. 12. Exponential Fitted Energy Dependence of CL Intensity of Sionon Irradiated to 5 Gy

photon energy in keV. The solid line in the figures indicates the polynomial fitting, while the dotted line shows $R(x)_s$ or $R(x)_g$ fitting. Numerical values of all relevant constants are indicated together with correlation coefficient r in each figure. All those fitting polynomials terminate at cubic term of *log E* with correlation coefficients close to or larger than 0.99, except for the case of Sionon of which fitting polynomial is extended to biquadratic term of *log E* with similar correlation

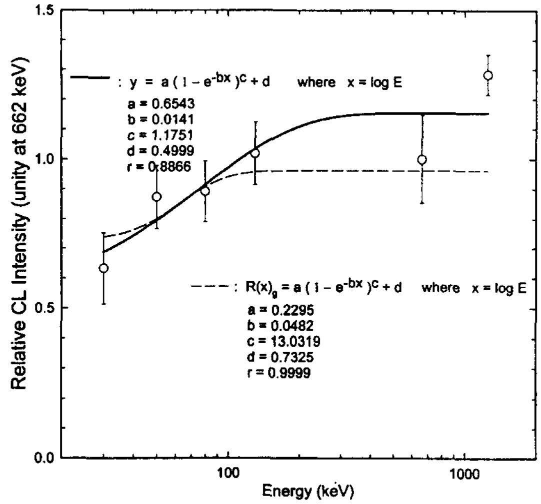

Fig. 13. Exponential Fitted Energy Dependence of CL Intensity of Sorbitol (<0.4 mm) Irradiated to 5 Gy

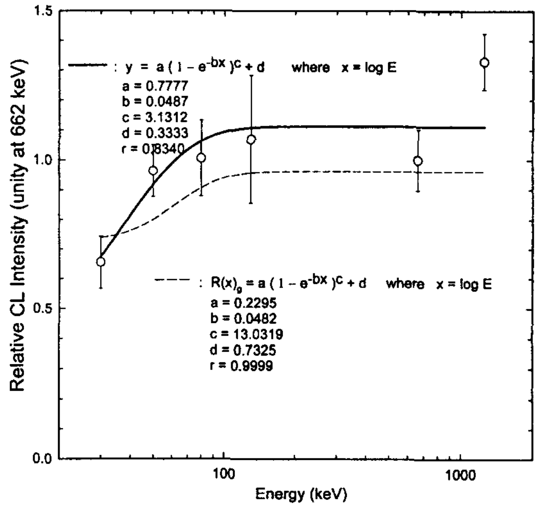

Fig. 14. Exponential Fitted Energy Dependence of CL Intensity of Sorbitol (>0.4 mm) Irradiated to 5 Gy

Table 2. Numerical Values of Constants in $y_R = a \cdot (1 - e^{-bx})^c + d$ Fitting

Sample	a	b	c	d	r
GSW	0.6564	0.0025	1.1544	0.5187	0.9884
KSW	0.5364	0.0076	1.3108	0.4987	0.9708
Sionon	0.4643	0.0162	1.7551	0.5987	0.9673
Sorbitol (<0.4mm)	0.6543	0.0141	1.1751	0.4999	0.8866
Sorbitol (>0.4mm)	0.7777	0.0487	3.1312	0.3333	0.8340
R(x)s	0.2477	0.0447	10.515	0.7142	0.9999
R(x)g	0.2295	0.0482	13.032	0.7325	0.9999

coefficient. In those Figs. 5 through 9, closeness of the polynomial fitting of experimental data to the theoretical response curve can estimably be visualised.

In the succeeding five figures, namely, Figs. 10 through 14, experimental data were fitted to the function

$$y_R = a \cdot (1 - e^{-bx})^c + d \qquad (3)$$

which is deduced from the theoretical energy response curve expressed as Eq. (1). The solid lines are fitted to experimental points (y_R fitted) and dotted lines are theoretical curves fitted to Eq. (1) ($R(x)_s$ or $R(x)_g$ fitted). Numerical values of relevant constants and correlation coefficient are expressed in each figure.

Although the contiguity between y_R fitted and $R(x)$ fitted curves does not seem to be satisfactory, particularly in the case of sugar, it is quite noteworthy to see the shape of the fitted curves are very closely resembled to the theoretical response curves, in particular, in the case of sorbite. Attaching importance to this point, we

Table 3. Relative Sensitivities of Sugar and Sorbite as a Function of Irradiated Photon Energy*

Energy (keV)	S_{KSW}/S_{GSW}	S_{Sion}/S_{GSW}	$S_{Sorb>0.4}/S_{Sorb<0.4}$	$S_{Sorb<0.4}/S_{Sion}$
30	0.68 ± 0.08	0.56 ± 0.05	0.34 ± 0.05	0.23 ± 0.03
50	0.90 ± 0.14	0.53 ± 0.03	0.37 ± 0.05	0.31 ± 0.04
80	0.75 ± 0.17	0.55 ± 0.07	0.38 ± 0.06	0.25 ± 0.03
130	0.85 ± 0.07	0.63 ± 0.05	0.35 ± 0.08	0.25 ± 0.03
Mean (30-130)	0.80 ± 0.12	0.57 ± 0.05	0.36 ± 0.06	0.26 ± 0.03
662	0.72 ± 0.11	0.44 ± 0.03	0.33 ± 0.06	0.23 ± 0.04
1250	0.66 ± 0.09	0.42 ± 0.05	0.35 ± 0.04	0.26 ± 0.02
Mean (662-1250)	0.69 ± 0.10	0.43 ± 0.04	0.34 ± 0.05	0.25 ± 0.03
Grand Mean	0.76 ± 0.12	0.52 ± 0.05	0.35 ± 0.06	0.26 ± 0.03

* CL intensities are normalised to unity at 662 keV.

came to a conclusion of taking Eq. (1) for fitting the energy response of CL intensity, rather than taking polynomial of log E for the fitting. For a comparative observation, the numerical values of the constants and correlation coefficients appeared in Figs. 10 through 14 are collectively tabulated together with those of $R(x)_s$ or $R(x)_g$ in Table 2.

Once the value of y_R is known one might be able to make correction for energy dependent CL output of sugar and/or sorbite irradiated to low energy photons, particularly below about 200 keV, to obtain normalised dose as it otherwise should be.

3.2. Energy Dependence of Relative Sensitivity

With the CL outputs obtained in the course of this energy response study, relative sensitivities of the samples at various radiation energies were estimated. The evaluated relative sensitivities are the sensitivity ratio of KSW to GSW (S_{KSW}/S_{GSW}), the ratio of Sionon to GSW (S_{Sion}/S_{GSW}), and that of Sorbitol of < 0.4 mm to Sionon ($S_{Sorb<0.4}/S_{Sion}$). The sensitivity ratio of Sorbitol of larger than 0.4 mm to that of smaller than 0.4 mm ($S_{Sorb>0.4}/S_{Sorb<0.4}$) was also made in anticipation of appearance of the grain size effect. The results are summarised in Table 3.

In the table, it is shown that the relative sensitivities tend to be higher in low energy region, although there appeared no prominent energy dependence in the whole region of radiation energy. As one can see in the table, mean relative sensitivities taken in the energy region of 30 to 130 keV is as high as 32.6 % more than the mean values taken for gamma-rays of 662 to 1250 keV in the case of S_{sion}/S_{GSW}. For the sugar, S_{KSW}/S_{GSW} in low energy region appeared to be about 16 % higher than that in 662 - 1250 keV region. In the case of sorbite, however, the discrepancies in the relative sensitivities in low (< 200 keV) and high (> 500 keV) energy regions did not come out to be as big as in the previous cases (Table 3), though the values in low energy region are yet somewhat higher. In Table 3, mean relative sensitivities in 30 to 130 keV and 662 to 1250 keV regions are indicated, respectively, together with grand mean values.

As a whole, we can say that even the samples like KSW and Sionon which are, in general, less sensitive compared to GSW could become more sensitive to the irradiation of low energy X-rays than to the gamma-rays of higher energy probably because of the relatively larger total absorption coefficient of the low energy photons.

Uncertainties of the grand mean values in terms of standard deviations of the CL counts were shown to range from about 10 % in the case of S_{Sion}/S_{GSW} to 17 % in $S_{Sorb>0.4}/S_{Sorb<0.4}$. Grand mean values of S_{KSW}/S_{GSW} and S_{Sion}/S_{GSW} came out to be 0.76 ± 0.12 and 0.52 ± 0.05, respectively.

3.3. Grain Size Effect

In the distribution of $S_{Sorb>0.4}/S_{Sorb<0.4}$ values in Table 3 we can easily see a remarkable grain size effect in CL sensitivity of irradiated Sorbitol samples. Discrepancy in relative sensitivities in low and high energy regions is, in this case, shown to be negligibly small. The grand mean of the ratio came out to be 0.35 ± 0.03, which means that Sorbitol of finer grain is about 2.9 times more sensitive, on average, than that of coarser grain in whole range of irradiated radiation energy. At the same time it is shown on the last right side column of the table that even finer Sorbitol is 0.26 times less sensitive, on average, than Sionon to radiation.

4. Conclusions

In accordance with the results obtained in this study, in which 5 Gy irradiation dose of different photons from 30 to 1250 keV in energy was administered to sugar and sorbite produced in Germany and Korea, respectively, it is concluded as follows :

(1) Distinct energy dependencies of CL of sugar and sorbite were found in the region of 30 to 1250 keV photons irradiated. This energy dependence, in common, can be fitted with a satisfactory correlation coefficient by a polynomial of *log E*, with *E* being radiation energy, up to cubic term of *log E*. According to a comparative investigation, however, with theoretical energy response curve calculated as the ratio of the mass energy absorption coefficients of the samples of interest to the soft tissue[14, 15], we reached a conclusion that adoption of Eq. (3) as a fitting function is more reasonable and rational. With satisfactory correlation coefficients of +0.83 to +0.99, the fitting function was applicable to all the samples used in this study with varying numerical values of the constants involved in the equation.

(2) No noticeable discrepancy was found in the degree of energy dependencies of German and Korean sugar. Contiguity between the fitted curves by Eq. (3) and theoretical curve (see Figs. 3 and 4) was shown to be closer in the case of sorbite (both Sionon and Sorbitol) than sugar. Should the value of fitted y_R, energy dependent CL intensity is known at a certain energy, one can make correction of CL output to obtain a normalized value corresponding to the irradiated radiation dose.

(3) It appeared that the relative sensitivity of less sensitive sample to the more sensitive one, in general, shows comparatively higher relative sensitivity in low energy X-ray region (<200 keV) than in high energy gamma-ray region (> 500 keV). It seems to indicate that in low energy photon field, even less sensitive samples can become more sensitive to the irradiated radiation due to relatively larger total absorption coefficient of the radiation than that of higher energy photons. Overall mean values of the relative sensitivities of KSW and Sionon to that of GSW came out to be 0.76 ± 0.12 and 0.52 ± 0.05, respectively, with slightly larger and smaller values in low and high energy region (see Table 3), which implies energy dependence of relative CL sensitivities.

(4) Prominent grain size effect of Sorbitol in radiation sensitivity was found. Sorbitol of < 0.4 mm is turned out to be about 2.9 times more sensitive to radiation, on average, than that of > 0.4 mm in grain size. A similar effect was reported in sugar EPR dosimetry.[17] Contiguity of fitted energy response curve to the theoretical curve appeared to be closer in the case of finer Sorbitol, though patterns of the fitted curves in both cases are similar to each other.

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