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≪Original≫ *An Experimental Approach for Verifying the Effect of Scattered Gamma-rays on the "Before Glow" in a Thermoluminescent Glow Curve

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Abstract

In order to verify the contribution of scattered photons in a restricted gamma-cell as a cause of the "before glow" on a thermoluminescent glow curve of natural quartz, the ratio of the scattered to primary radiation contributions (S/P) in the cell is measured and the relationship between the effective "before glow" height (h_b) and S/P ratio is quantitatively investigated. The result shows quite good linear relationship between them with a correlation coefficient of +0.9, which possibly suggests that the electrons originally released by the photons of reduced energy are trapped in the shallower traps.

Moreover, the ratios of h_b to total glow area (A_t) and of effective "before glow" area (A_b) to A_t are also examined to see the relationships between S/P and each of them, respectively. The relationships are represented by exponential functions in the region of S/P greater than 0.035. Finally, the exposure limit for re-use of the natural quartz as a TLD was found to be approximately 10^5 R by analyzing total thermoluminescent output and corresponding exposure dose.

요　　약

수정의 영형광 glow curve 상, "before glow"의 생성원인중의 하나가 밀폐된 제한 공간내의 산란 감마선의 기여임을 확인하기 위하여 산란선대 일차선의 기여비(S/P)를 측정하였다.

이 S/P와 "before glow"의 유효높이 (h_b)와의 상관 관계를 고찰하였는바 상관계수+0.9라는 비교적 밀접한 일차 관계가 있음을 알았으며 이는 에너지가 감소된 산란선에 의하여 여기되었던 전자가 본래 일차선으로 여기되었던 전자보다 얕은 trap에 걸려 있었음을 입증하는 것으로 보인다.

한편 h_b와 glow curve의 전면적 (A_t)의 비 및 "before glow"의 유효면적(A_b)과 A_t와의 비가 S/P와 어떤 관계에 있는가도 조사하였는데 이들의 관계는 단순하지 않으며 다만 S/P 값이 0.035 보다 큰 영역에서는 간단한 대수함수로 표현되었다. 끝으로

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자연수정을 TLD로 사용할 경우 그 재사용을 위한 선량한계를 살펴본 결과 그것은 10^5 R 정도임을 알아내었다.

1. Introduction

In the previous work[1,2] it was pointed out that the occurance of "before glow" on thermoluminescent glow curves of gamma-ray irradiated type 1 α-quartz possibly stems from two main factors; 1) nonlinearity of heating and 2) the effect of scattered photons.

Generally, the thermoluminescent emission is caused by the radiative transitions of thermally released electrons from their trapped centers to their original position, and such energies of the conduction electrons are supplied by a continuous rise of temperature due to linear heating[3,4]. If, however, the constant supply of thermal energy is disrupted by nonlinear heating, conduction electrons wander about in the conduction band by exchange of energies, and at a certain energy level, retrapping suddenly becomes dominant with results similar to an avalanche of electrons. Such retrapping tends to occur at the initial stage of a glow curve because of the lower energies of the electrons released from the shallowest traps. Consequently, such an effect results in a "before glow" on the thermoluminescent glow curve.

Fig. 1 shows a "before glow" on a glow curve obtained in a type 1 α-quartz[5] which was irradiated at about maximum distance from the gamma source, that is, closest to the shielding wall of the cell among the irradiated samples[1]. In this case, electrons can be trapped in the shallowest traps, hence even a slight variation of the heating rate will greatly affect to the occurance of "before glow".

As the cause of this effect, the contribution of scattered photons originating in the restricted cell of the gamma-ray irradiation facility was suggested in the previous work.

In the present study, a series of experimental measurements of the ratio of scattered-to-primary radiation in the cell at various distances and in various directions from the source was carried out in order to support this explanation more clearly on the contribution of the scattered photons to the "before glow" occurance.

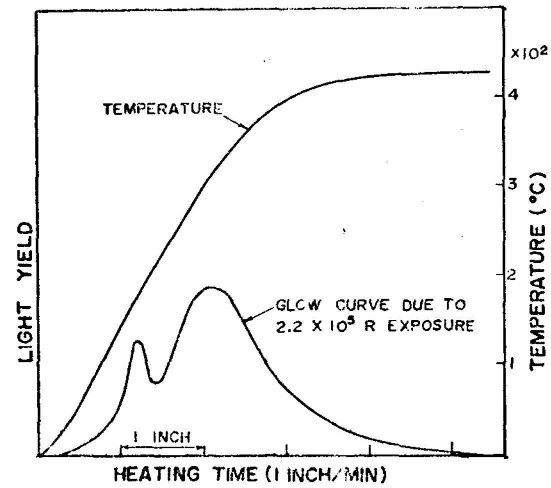

Fig. 1. A "Before Glow" on a glow curve with showing the uniformity of heating rate

2. Experiment

2-1) Measurement of Scattered Photon Ratio

The AERI Gamma-Cell used for the irradiation of type 1 α-quartz is a closed cell of ordinary concrete wall, ceiling and floor. Two Co-60 gamma sources are set in two aluminum guide pipes vertically installed at nearly center of the cell and the geometrical arrangement is as shown in Fig. 2.

During the course of irradiating the samples, source I (350 Ci in Jan. 1971) was operated with a height of nearly 10 cm above the floor keeping the source II shut down. Although there is about 10 cm difference in source height between in the quartz sample irradiation and in S/P measurement, it shows not any significant effect on this study according to careful examination on the geometries.

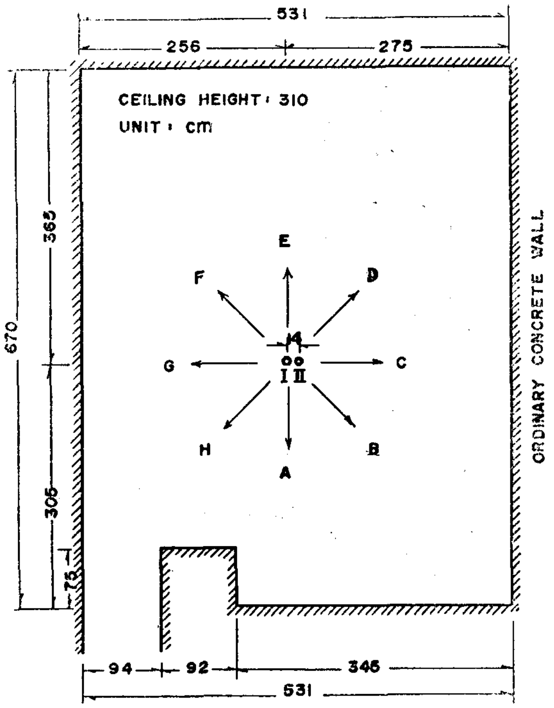

Fig. 2. Internal dimensions and directions of S/P measurement in Co^{-60} gamma-cell

The geometrical estimation in the worst possible case, in this study, which may yield the largest error due to the difference of source height shows that only a fraction~10^{-4} of all photons emitted by the source come to exist in the space. The difference in this order of all emitted photons is negligible comparing with 5 to 6% error involved in the measurement of S/P ratio.

The ratio of scattered radiation (photons of reduced energy) contribution to primary rays in eight directions shown in Fig. 2(A. B. C. ⋯ ⋯*etc.*) were measured at 10 to 15 points in every directions by means of Victoreen's RADOCON Ⅱ R-meter with a simple lead bar, of 20 cm in length and 3 cm in diameter, attached to the front of the detecting chamber in the arrangement shown as Fig. 3 assuming collimated beam condition for primary radiation between the source and detecting chamber.

Under this condition, primary gamma-ray of Co-60 is to be cut down to less than the order of 10^{-4} when the lead bar is attached to the front of the chamber. Thus in this case only scattered photons are to be detected almost without the contribution of the primary radiation. Let us denote this quantity as S. When the lead bar is removed, the chambar will detect the total amount of radiation consisted of primary plus scattered photons which interact with the detector at the position. This quantity is denoted as T. Then T-S will represent the contribution of primary radiation

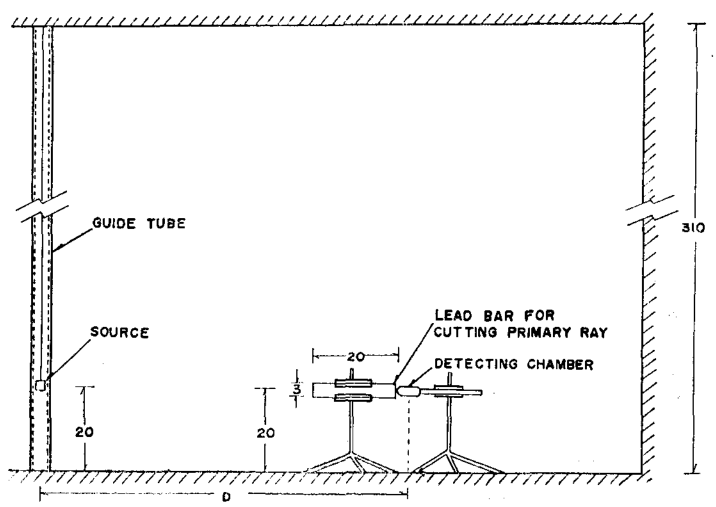

Fig. 3. Arrangement for S/P measurement

only (P) at any definite point. In this way, the ratio of the scatterd to primary radiation contributions(S/P or S/T-S) were measured twice for each point and the values were simply averaged.

2-2) Irradiation Conditions of the Quartz Samples

The type 1 α-quartz samples are irradiated in the direction of E shown in Fig. 2 and the distances from the source to each sample are indicated in table 2 with corresponding total exposure dose in R.

3. Results and Discussion

The results of the S/P ratio measurement along with the eight directions shown in Fig. 2 at intervals of 25 cm are summarized elsewhere[6). Here in this paper, the S/P ratio in E direction, which is directly related to the thermoluminescent "before glow" of type 1 α-quartz, is summarized numerically and graphically in table 1 and Fig. 4, respectively.

Although the graphic representations of the scattered to primary ratio along with the various directions and distances are very complicated[6) because of the non-symmetry of

Table 1. S/P ratio in E direction

Distance from the source (cm)	Total Exp. Dose* (P+S=T) (R)	Exp. Dose* due to Scattered Photons(S) (R)	S/P ratio (S/T-S)
30	160.0	5.6	0.0350
50	66.0	4.5	0.0682
75	32.0	2.2	0.0688
100	18.3	1.6	0.0877
125	12.0	1.33	0.1108
150	8.9	1.27	0.1428
175	6.7	0.96	0.1432
200	5.3	0.82	0.1547

* Dose measurement time was identically two minutes at every point.

the cell geometry with respect to the source, an overall or a general tendency indicates apparent increment of the scattered to primary ratio with distance from the source. Moreover, in the region of distances up to about 130 cm from the source, the curves coincide with each other within about ±10% in S/P value. This tendency is not exceptional for the direction of E as shown in Fig. 4.

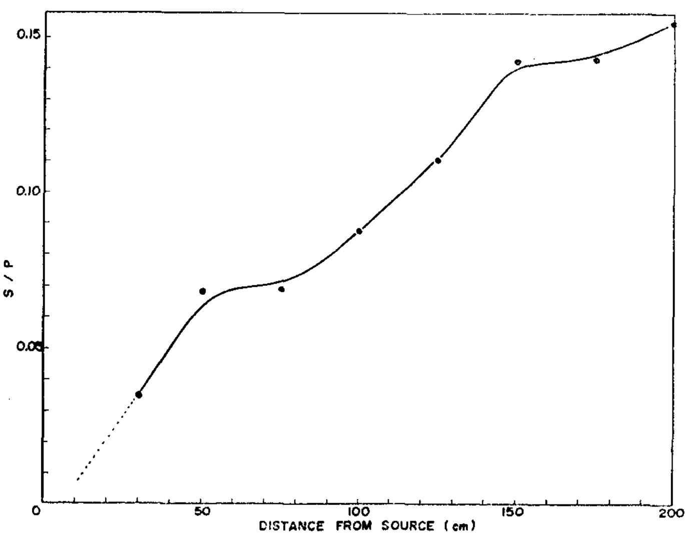

Fig. 4. Scattered to primary radiation ratio in E direction

[Table 2. **Values of h_b, h_b/A_t and A_b/A_t with corresponding S/P ratio**]

Sample	Distance from source (cm)	Corresponding S/P ratio	Total exposure (R)	Effective "before glow" height(h_b) (arb. unit)	Effective "before glow" area (A_b) (arb. unit)	Total glow area (A_t) (arb. unit)	h_b/A_t	A_b/A_t
a	6.5	—	1.3×10^{7}	—	—	931	—	—
b	16	0.015	2.2×10^{6}	1.0	16	803	0.125×10^{-3}	1.99×10^{-2}
c	32	0.038	5.5×10^{5}	1.2	11	534	0.225×10^{-3}	2.06×10^{-2}
d	47	0.060	2.5×10^{5}	5.1	37	259	1.97×10^{-3}	14.3×10^{-2}
e	70	0.070	1.2×10^{5}	6.3	27	77	8.18×10^{-3}	35.1×10^{-2}

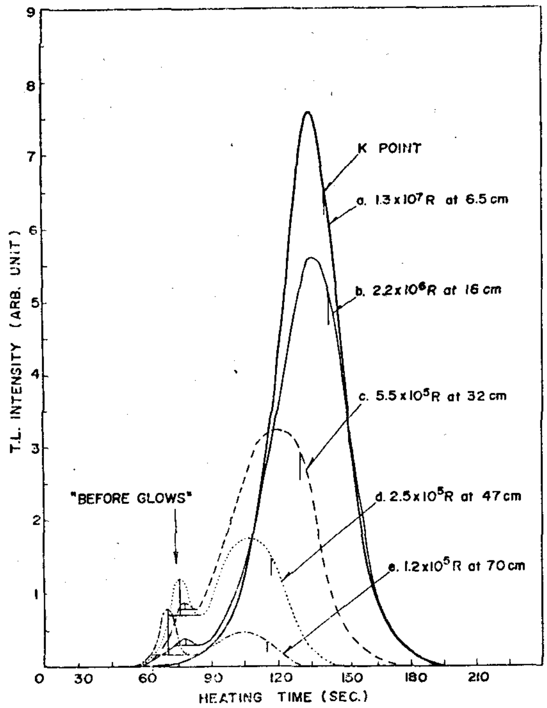

Fig. 5. Glow curves of γ-irradiated type 1 α-quartz showing various "Before Glow" heights

The thermoluminescent glow curves of gamma-irradiated type 1 α-quartz are shown in Fig. 5. As shown in the figure, the effective "before glow" height (h_b)[1,2] is varying with the distance from the gamma-ray source to the irradiated samples. In other words, it varies with S/P ratio in that direction. Numerical representations of these quantities are summarized in table 2.

Although the number of "before glow" samples is not sufficient to analyze the relationship in association with the S/P ratio on the statistical basis, it seems to have a linear relationship between them. According to the numerical calculations, lines of regression of $Y(h_b)$ on X(S/P) and of X on Y are passing very closely each other as shown in Fig. 6. Calculated regression equations are

$$Y=10.317X-0.132$$

$$X=0.085Y+0.017,$$

respectively. Correlation coefficient between them is estimated to be +0.9. These lines of regression, indicating fairly close quantitative relationship between h_b and S/P, are considered to imply that the occurance of "before glow" on thermoluminescent glow curve of gamma irradiated type 1 α-quartz is caused by the contribution of scattered photons whose energies were at least once reduced. It is not difficult to suppose that the electrons released due to gamma-ray of reduced energy will be trapped in shallower traps than the electrons released by the primary gamma-ray.

And generally the number of peaks in a glow curve of luminescent brightness vs. heating time is equal to the number of different types of traps in the material[7]. These facts are connected to the relation between the trap depth of the shallower traps which are originally occupied by the released electrons due to scattered photons and that of the ordinary traps which are occupied by the electrons liberated by primary radiations.

Furthermore, we note that typically once scattered photon carries about one-half of the

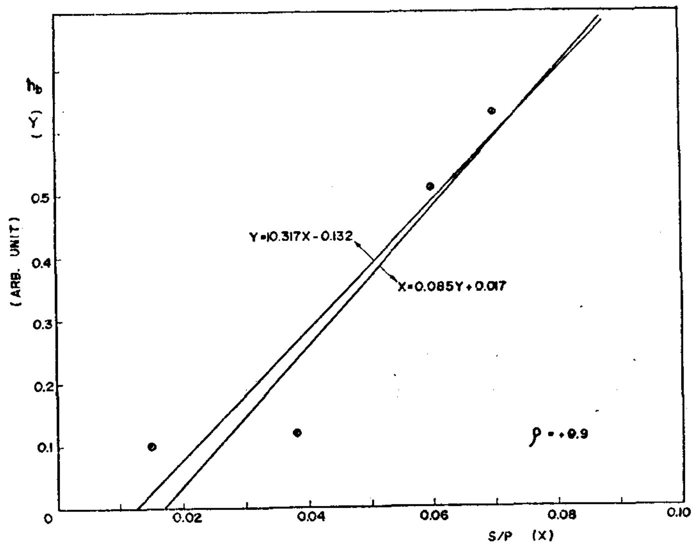

Fig. 6. Relationship between effective "Before Glow" height and S/P

primary photon energy although in general the energy of the scattered photon depends on the angle of scattered and the primary photon energy[8]. At the same time, it is easily found in Fig. 1 that the temperature corresponding to the "before glow" peak is nearly one-half of that corresponding to the main glow peak. This implies clearly that the origin of the "before glow" on a glow curve of the gamma irradiated type 1 α-quartz is possibly the contribution of the scattered photons as far as the rate of heating is kept constant during the glow reading.

On the other hand, it is thought that there may be a possibility of evaluating the contribution of primary and scattered radiations at a certain point in the restricted radiation field simultanously by the quantitative analysis of the "before glow" and "main glows" of thermoluminescent glow curve if it is true that there is a linear relationship between the individual peak height or area of the glow curve and the contributions of scattered and primary radiations.

In order to examine these relationships, the ratio of effective "before glow" height (h_b) to total area under the glow curve (A_t) and that of effective "before glow" area (A_b) and A_t are computed and summarized in table 2. Here, A_b is defined as the area under the "before glow" curve which is cut by the horizontal line crossing the down end point of the h_b (refer to Fig. 5). Defining A_b in this

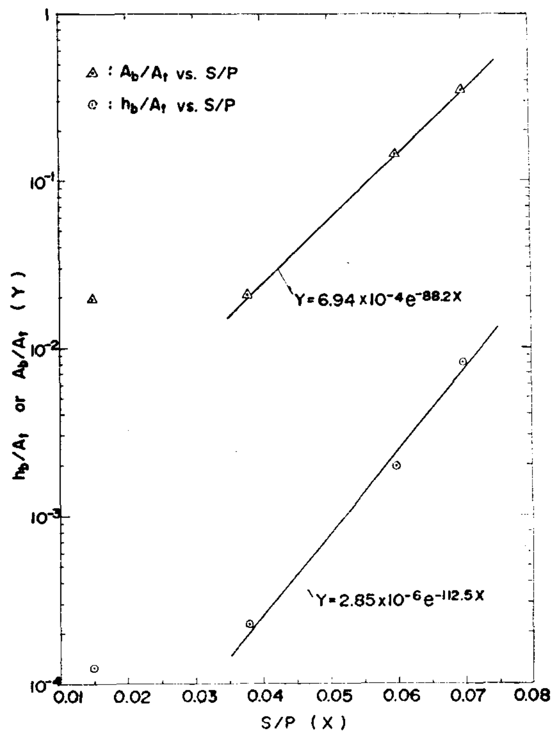

Fig. 7. h_b/A_t or A_b/A_t vs. S/P

simple way without analyzing two overlapping gaussian shapes is based on the definition of h_b where a particular phenomenon of electron retrapping was taken into account. In Fig. 7 h_b/A_t and A_b/A_t versus S/P are plotted, respectively.

As shown in this figure, it looks not easy, in either cases of h_b/A_t vs. S/P or A_b/A_t *vs.* S/P, to search for a proper form of simple function to fit all the points at once. If, however, the region of S/P greater than about 0.035 is taken into account for the sake of convenience as an approach, the relationships between them seem to be represented with a simple exponential function, $Y=be^{aX}$. According to least squares fitting, with an elimination of the points appeared in the region of S/P less than 0.035, the equations become;

for h_b/A_t *vs.* S/P,

$$Y=2.85\times10^{-6}e^{112.5X} \quad (X>0.035)$$

for A_b/A_t *vs.* S/P,

$$Y=6.94\times10^{-4}\, e^{88.2X} \quad (X>0.035),$$

respectively.

Finally, the relationship between the total thermoluminescent output and corresponding exposure dose is investigated. As is well known, dosimetrical utilization of thermoluminescent material is based on the assumption that the number of electrons excited and trapped, hence the amount of light given off when the material is heated, is proportional to the amount of absorbed radiation. Therefore, the relationship between the thermoluminescent output and the exposure dose is an essential factor in dosimetrical point of view. In this study, the total thermoluminescent output is evaluated by the light integration technique, *i.e.* the integration of the area under the entire glow curve.

In Fig. 8, which represents a curve of relative total thermoluminescent output vs. corresponding exposure dose in R, the relationship indicates a curvature rather than a line,

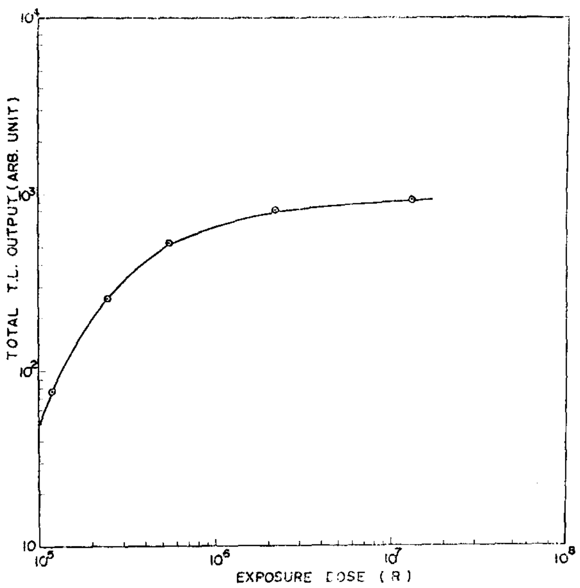

Fig. 8. Total thermoluminescent output *vs.* corresponding exposure dose

In the exposure range below 10^5 R down to 300 R, it shows fairly good linearity according to the study of one of the authors[9]. The curvature shown in the figure is quite similar with that in the case of LiF(Harshaw TLD-100) which was investigated by Cameron *et al.* with a proposed mathematical model concerning to the total thermoluminescent output *vs.* total exposure dose in R[10]. In the case of LiF, such a curvature in thermoluminescent output *vs.* R curve starts at the exposure of the order of 10^4 R, while in the case of present study (type 1 α-quartz) the exposure of 10^5 R seems to be the starting point of the curvature comparing with the shape of that of the LiF.

According to Marrone and Attix[7], this is the result of damage, so-called "permanent" damage, to the crystal rather than the filling of all available traps, that is, thermoluminescent material which is once exposed to the dose of above mentioned order cannot be reused indiscriminately. Consequently, for type 1 α-quartz, the exposure of 10^5 R may be the limitting dose in its gamma-ray responsiveness.

Summarizing the above mentioned results, it may be concluded that in evaluating gamma-ray exposure dose by means of quartz TLD, the "before glow" should not be neglected particularly when it is used in a restricted high-dose gamma-radiation field where the contribution of scattered radiation must be taken into account. This idea on the "before glow" occurance may be extended in investigation of various trap depths of a particular crystal, assuming the traps are occupied by electrons initially released due to gamma-rays of correspondingly various energies.

4. Conclusions

The conclusions derived through this study are as follows;

1) Contribution of scattered photons in a restricted radiation field is possibly the origin of "before glow" on a thermoluminescent glow curve of type 1 α-quartz as far as the rate of heating is kept constant during the glow reading.

2) It may be possible to evaluate the contribution of primary and scattered gamma-rays at a certain point in a restricted radiation field by analyzing the relationships between the effective "before glow" height (or it's area) and the total thermoluminescent output.

3) The exposure limit for re-use of type 1 α-quartz TLD is found to be the order of 10^5 R.

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