

《Original》 A Study of the Thermoluminescent Properties of  
Korean Natural Quartz for Possible  
Use in Gamma-Ray Dosimetry<sup>+</sup>

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Abstract

Various thermoluminescent properties of Korean natural quartz for possible use in  $\gamma$ -ray dosimetry has been studied. If the heating is exactly linear,  $\gamma$ -irradiated radiation sensitive (type 1)  $\alpha$ -quartz can yield a glow curve of single peak, hence glow peak height could be taken as a  $\gamma$ -dose for its dosimetry. Quartz crystal dosimeter exhibited the linearity of thermoluminescent intensity in the range from about  $2 \times 10^3$ R to  $2 \times 10^6$ R, and also had an advantage of low fading because of the high peak temperature ( $300 \pm 40^\circ\text{C}$ ). The pulverized quartz sample having the grain size of  $0.3 < \phi < 0.9$ mm showed the linearity of T. L. intensity in the range from 50R to  $2 \times 10^3$ R. Therapeutic application of the pulverized sample on the correct measurement of the absorbed dose in a body region of a cancer patient seems to be successful.

요 약

國內産 天然水晶의 熱螢光發生의 性質을 利用해서  $\gamma$ 線의 線量을 測定할 수 있는 可能한 여러가지 方法을 研究하였다. 加熱方法이 正確히 線形的일때는  $\gamma$ 線에 照射된 放射線에 敏感한  $\alpha$ 水晶은 單一尖頭의 熱螢光 發生曲線을 나타낼 수 있는고로 이 發生曲線의 尖頭의 높이는  $\alpha$ 水晶에 依해서  $\gamma$ 線의 線量을 測定하는 方法에 있어서  $\gamma$ 線의 線量을 나타낸다고 볼 수 있다. 이 水晶線量計는  $2 \times 10^3$ R에서  $2 \times 10^6$ R까지의 線量範圍內에서 熱螢光強度의 直線性을 나타내었으며, 또한 發生曲線의 尖頭 때의 溫度( $300 \pm 40^\circ\text{C}$ )가 높은 고로 熱螢光의 常溫에서의 自然衰退가 적다는 長點이 있다. 粒子的 크기가  $0.3 > \phi < 0.9$ mm인 粉末水晶은 50R에서  $2 \times 10^3$ R까지의  $\gamma$ 線의 線量範圍內에서 熱螢光強度의 直線性을 나타내었다. 癌患者의 身體一部에 照射된  $\gamma$ 線의 吸收線量을 正確히 測定해야하는 放射線治療上의 適用에 粉末水晶試料를 使用해본 試圖은 좋은 結果였다고 생각된다.

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## I. Introduction

Thermoluminescence dosimetry due to solid crystals has been extensively studied by many investigators<sup>(1-3)</sup> in recent years. From the results of these studies, synthetic crystals of  $\text{CaF}_2$  (Mn),  $\text{CaSO}_4$  (Mn), and  $\text{LiF}(\text{Mg})$  have been found to be especially suitable for this purpose, and they have the advantage of high sensitivity and excellent linearity to the absorbed dose.

On the other hand, some natural substances have been used to find the radiation absorbed by the samples in the past, such as limestone in the determination of the geologic age of carbonate sediments,<sup>(4)</sup> amorphous silica in ancient pottery for dating,<sup>(5)</sup> silica sands in roof tiles for the estimation of absorbed dose after twenty years from the drop of A-bombs in Hiroshima and Nagasaki.<sup>(6)</sup> Since silicates are ordinary substances which can easily be found everywhere on the earth, they can be used in case of an accident of a nuclear reactor, and so on.

Especially, natural quartz is very sensitive for  $\gamma$ -radiation in general. It has been known that,  $\gamma$ -irradiated radiation sensitive (type 1)<sup>(7)</sup>  $\alpha$ -quartz is a kind of thermoluminescence phosphor having the smoky color centers as well as luminophor centers of  $^{\text{Al}}\text{A}_+$  type and trapping centers of  $\text{xP}$ -type, and the population of these color centers is almost proportional to the absorbed dose of  $\gamma$ -rays within the extent of their saturation.

The present investigation was undertaken to examine the various thermoluminescence properties of Korean natural quartz for possible use in  $\gamma$ -ray dosimetry. Through this study, it has been clarified that, the crystal bulk sample of type 1  $\alpha$ -quartz can be used for the  $\gamma$ -ray dosimetry in the range from about 2000R to  $10^7$ R, and in case of pulverized sample of the quartz, sensitivity for  $\gamma$ -radiation can be greatly increased though it depends on a suitable grain size. The minimum detectability of the  $\gamma$ -dose due to the pulverized sample is about 50R.

It seems that the advantage of this quartz material for  $\gamma$ -dosimetry is in the following points

such as, easy acquiring of the material, easy handling of the material without wrapping it up with a black paper, easy reading of the  $\gamma$ -dose due to the glow peak height with a simple reader. The attempt on therapeutic application due to the pulverized sample seems to be successful.

## II. Experimental Procedures

### (1) Preparation of Quartz Crystal Samples

Several species of  $\gamma$ -sensitive (type 1) Korean natural quartz were cut perpendicular to c-axis in a uniform size of  $15 \times 15 \times 1$  mm, and polished them to be transparent.

In order to anneal out the effect of natural radiation, the samples were preheated at about  $350^\circ\text{C}$  for 5 minutes prior to their  $\gamma$ -irradiation. A sample case made of thin silver plate was used to aid the uniform heat transmission to the sample in the heating furnace.

### (2) Preparation of Pulverized Quartz Samples

A crystal bulk which is one of the best species of type 1  $\alpha$ -quartz was ground in an agate mortar after its rough crushing, and then the grains were sieved so that their sizes of  $\phi < 0.06$  mm,  $0.06 < \phi < 0.3$ ,  $0.3 < \phi < 0.9$ ,  $0.9 < \phi < 5.0$ , and  $5.0 < \phi$  were obtained.

Each pulverized sample was weighed to be 700 mg. A dish-formed silver container was used to aid the sample heating. After filling the sample in the container, the surface of the grains was pressed lightly by an iron mould to make better heat transmission among the grains.

### (3) The Method of Glow Curve Yielding

As has shown elsewhere<sup>(8)</sup> in detail, the heating method of the crystal samples for the glow curve measurements was based on a principle of the spontaneous linear rise in temperature due to the heat capacities of the furnace and the sample at a predetermined heater voltage under an evacuated environment, and the temperature of the sample rose gradually until it reaches thermal equilibrium.

The glow curve was yielded by K-point (a point near the saturation temperature) cut-off method (Ref.8), that is, the heater switch was cut off at point K, and the time was marked on the chart

paper by interrupting the PM tube power supply. By using this method, almost symmetrical glow curves were obtained. The glow peak height was taken as a  $\gamma$ -dose for the dosimetry.

The results of the glow curve measurements appeared in part III-1 were obtained by employing the reading system of abovementioned heating mechanism, in which the position of the heating pan was vertical in the bell jar. The advantage of this type is in alleviating unnecessary overheating of the quartz window glass as the heat convection in the bell jar takes place to the vertical direction.

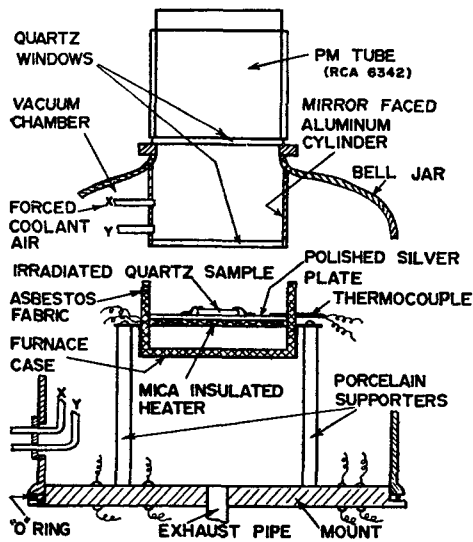


Fig. 1. T.L. reader system having a horizontal position of furnace for the quartz samples.

In order to facilitate the measurements of pulverized quartz samples, the position of the heating pan was changed to be horizontal, so that the PM tube was installed on the top of the bell jar as shown in Fig.1. The heating rate of the glow curve measurements in both of part III-1 and III-2 was  $100^{\circ}\text{C}/\text{min.}$

### III. Results and Discussion

#### 1. Properties of Quartz Crystal Dosimeter

##### (1) Linearity of Dose

The linearity of thermoluminescent intensity

with doses was measured in the mega-R range by using the type I quartz crystal samples, and the result is shown in Fig 2. Glow peak height was adjusted to avoid the overscaling on the recording paper by  $100\text{K}\Omega$  resistor connected in series with the high voltage source. The saturation of the color centers set in at the dose of about  $2 \times 10^6\text{R}$ , and up to  $1 \times 10^8\text{R}$ .

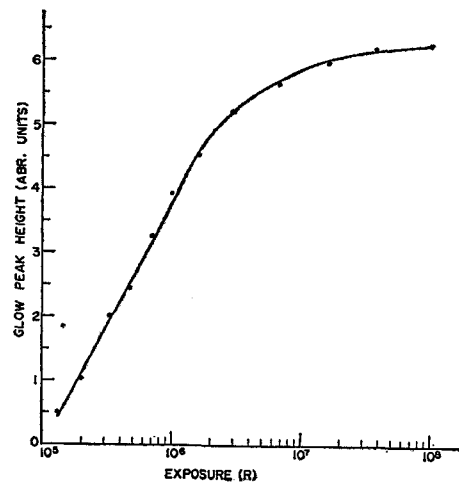


Fig. 2. Response of the quartz crystal dosimeter vs. exposure.

The linearity of doses ranging from  $10^3\text{R}$  to  $10^5\text{R}$  was examined by using the type 1 quartz crystal samples as shown in Fig.3. In these meas-

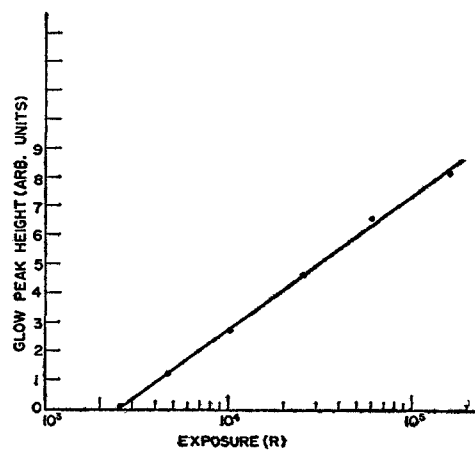


Fig. 3. Response of the quartz crystal dosimeter vs. exposure.

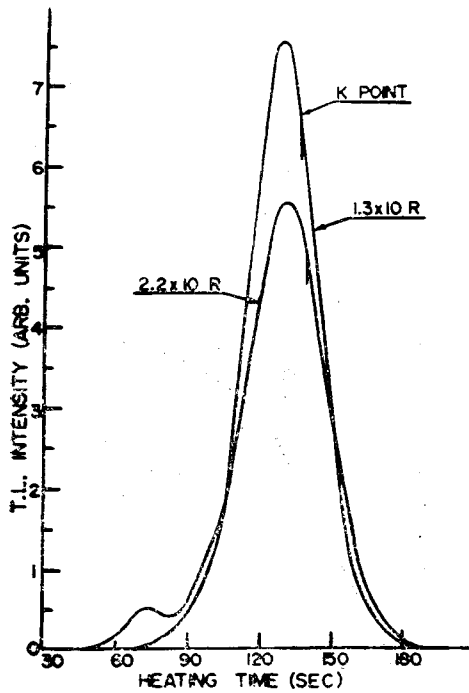


Fig. 4. Glow curves due to  $\gamma$  irradiated type 1 quartz samples showing a single peak and a "before glow".

urements, 10K $\Omega$  series resistor was used to adjust the photosensitivity. It is seen that relatively good linearity was obtained over the range of doses.

At these dosimetric glow curve measurements described above, K-point cut-off method was used for the heating in order to avoid unnecessary high temperature and long time constant of decay at the latter half of a glow curve as shown in Fig.4, and the method of peak height was taken for the dosimetry. As has shown in Fig.4, a "before glow" is appeared on the lower glow curve. It has been clarified<sup>(9)</sup> that the "before glow" was caused by a dominant retrapping of conduction electrons which were thermally excited from the shallowest traps and the height of "before glow" not only depends upon the dose rate but also the species of quartz and the frequency of the repeated use of the sample. Facey<sup>(10)</sup> suggested that both the population of the traps and the phosphorescence decay law should be function of dose rate. It has been proposed (Ref.9) that peak

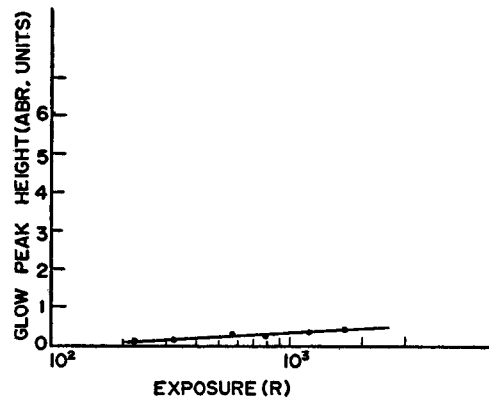


Fig. 5. Response of the quartz crystal dosimeter vs. exposure (low dose).

height of a glow curve could be reduced by yielding a "before glow", so that it is desirable to avoid its yielding by an accurate linear temperature control.

The linearity in the range from 200R to 10<sup>3</sup>R was examined by using a best species of type 1  $\alpha$ -quartz samples as shown in Fig.5. In this case, no series resistor was used. The results indicates that the present  $\gamma$ -sensitivity of the samples is not enough for the dosimetry in this dose range, though the samples were prepared for the improvement of TL emission by Ichigawa's method.<sup>(11)</sup> According to his method the susceptibility of natural quartz for radio-thermoluminescence can be fairly improved by  $\gamma$ -irradiation of high dose of  $1 \times 10^7$ R. He suggested that the gain comes from the radiation damage of crystal making a contribution to the production of trapped center. This kind of improvement also occurs by repeated use of the sample. Batrak<sup>(12)</sup> suggested that the quartz crystals repeatedly subjected to irradiation can exhibit stronger luminescence, i. e., they become "trained". The principal disadvantage of such low dose measurements are in the following points such as the difficulty of getting equally improved susceptibility of each sample for radio-thermoluminescence by the method, and the fact that difference among sample sizes gives considerable influence for such low dose measurements. The low dose measurements were attempted by pulverized quartz samples more successfully as shown in part III-2 later.

## (2) Fading Effect of Quartz Crystal Samples

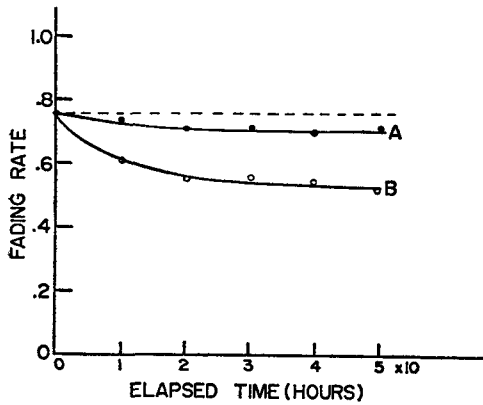


Fig. 6. Fading in T.L. intensity due to the quartz crystal dosimeters.

In ordinary thermoluminescent phosphor, the fading means the lowering of the amount of thermoluminescence which is concerned with the number of released electrons from the shallow traps rather than the fading of the color centers formed by exposure to radiation.

Fading in this sense has been measured for the  $\gamma$ -irradiated type 1  $\alpha$ -quartz samples under various conditions after exposure to  $10^7$ R of Co-60  $\gamma$ -ray. The results are presented in Fig. 6. The curve A shows the fading of the samples placed under the two fluorescent lamps of 860 lux, and the fading due to the sample is seen to be slight. The curve B shows the fading of the samples placed under the direct sunlight at clear day time, and the fading rate is fairly larger than that in the curve A.

In both curves, the fading occurs most prominently at the beginning of the curves, and it appears that these rapid fadings are probably due to the electrons trapped in the shallowest traps. Schulman<sup>(13)</sup> suggested that, in a glass dosimeter, early fading was largest and most rapid, indicating that some unstable centers involved in the coloration.

In case of curve B, the method can be regarded as the fading test of the color centers rather than as that of thermoluminescence as it was tested under direct sunlight, but in case of  $\gamma$ -irradiated type 1  $\alpha$ -quartz sample, this kind of fading test is inevitable on account of its deep depth.

Since the trap depth of  $\gamma$ -irradiated natural quartz is fairly deep in comparison with other thermoluminescent substances, the color centers as the stored radiation energy are not faded easily under ordinary environmental conditions. According to the Randal and Wilkins's<sup>(14)</sup> formula,  $E = 21kTg$ , the trap depth of the quartz was calculated to be 2.10eV if the glow peak temperature is 340°C.

For fading in a type 1  $\alpha$ -quartz sample, electrons trapped in the shallow trap levels tend to be released thermally, but in this case the excitation of electrons is not enough to maintain the necessary lifetime for the recombination, they are re-trapped immediately at the deeper levels of the shallow traps, hence the deepened trap depth of the trapped electrons will bring a reduction of the thermoluminescent intensity at a given temperature when the sample is bleached.

Another type of fading tested is shown in Fig. 7. Samples (2) and (B), and (3) and (C) in each

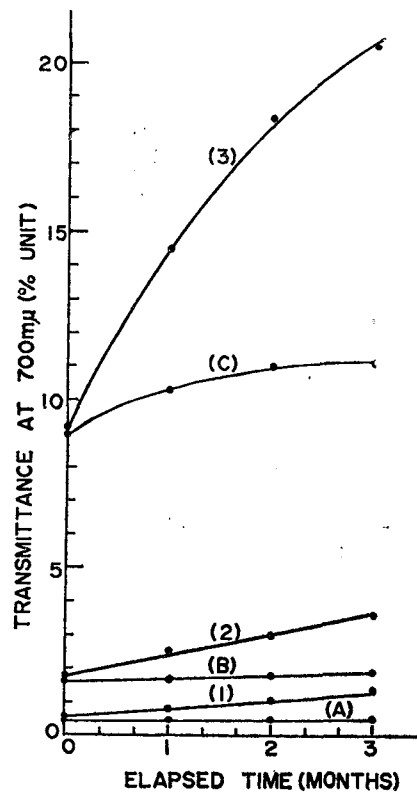


Fig. 7. Fading of the color centers in  $\gamma$  irradiated  $\alpha$ -quartz samples.

couple are  $\gamma$ -sensitive (type 1)  $\alpha$ -quartz while (1) and (A) belong to  $\gamma$ -insensitive (type 2)<sup>(15)</sup> one. Sample (A), (B), and (C) were placed in an illuminated study room, and (1), (2), and (3) were placed under the direct sunlight. Radiation dose for sample (3) and (C) were  $2.5 \times 10^6$ R; for (2) and (B),  $5.6 \times 10^6$ R; and for (1) and (A),  $9.4 \times 10^7$ R.

The transmittance of these samples was examined at  $700\text{m}\mu$  at one month interval including night time. The test due to  $700\text{m}\mu$  as an absorption-free visible band may be regarded as a method of examining the population of color centers and the related electron traps. According to the glow curve of the sample (3) and (C) measured after finishing the above fading test, the glow peak height of the sample (3) was considerably decreased compared to sample (C), and moreover a small "before glow" was observed for sample (C). It seems that, in case of the sample (C), color centers were faded a little and small amount of trap population remained in the sample.

### (3) Dose Rate Dependence.

High dose rate dependence was examined by exposing the crystal samples of type 1  $\alpha$ -quartz to a total dose of approximately  $10^5$ R at dose rates varying from  $2.0 \times 10^4$ R to  $2.0 \times 10^5$ R. Results of the measurements is as shown in Fig. 8. There appears to be no significant effect over the range of doses investigated, but it seems that a little weaker response at the lower side of the doses is due

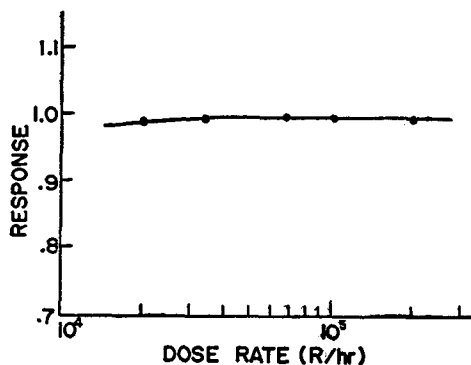


Fig. 8. Response of the quartz crystal dosimeters vs. dose rate.

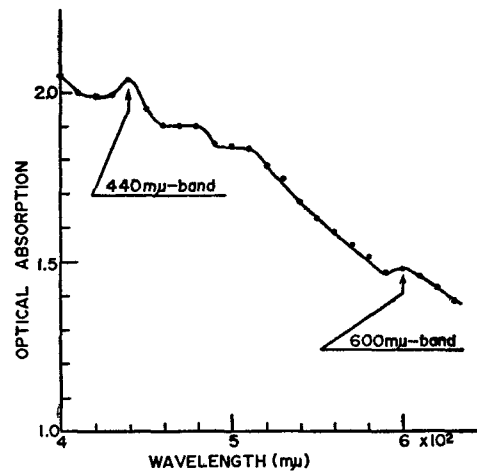


Fig. 9. Optical absorption vs. wavelength due to a quartz crystal dosimeter.

to the decrease in glow peak heights caused by the effect of "before glow" as described in part III-1-(1). The detailed description of the effect of "before glow" which is related to the dose rate is to be appeared in the Ref.9.

### (4) Thermal Bleaching of the Smoky Color Centers

One of the best type I samples exposed to  $3 \times 10^7$ R was used for the thermal bleaching measurements. The sample was heated in the furnace at the constant heating rate of  $100^\circ\text{C}/\text{min}$ , and taken out from it after forced cooling. The color centers showing the absorption maxima of  $440\text{m}\mu$  and  $600\text{m}\mu$  before bleaching are shown in Fig. 9.

Optical absorption of the sample bleached at

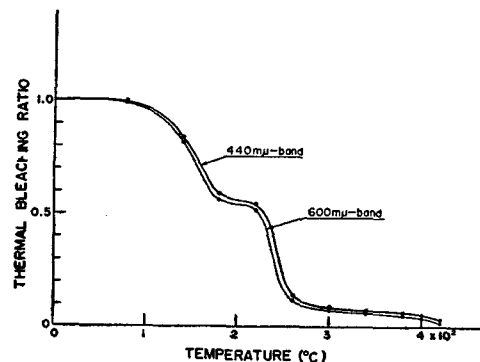


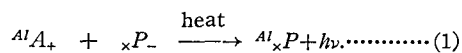
Fig. 10. Thermal bleaching ratio vs. temperature due to a quartz crystal dosimeter.

higher temperatures was measured in the range of 400-630m $\mu$ , and the thermal bleaching ratio of the color centers are shown in Fig. 10.

According to the optical absorption measurements of various species of  $\gamma$ -irradiated quartz samples, the absorption maxima appeared in the ranges of 440-455 m $\mu$  and 600-620m $\mu$ . Cohen<sup>(16)</sup> found natural and bleached X or  $\gamma$ -irradiated quartz to yield absorption maxima at 425m $\mu$  and 625m $\mu$  for orientation either parallel or perpendicular to the optic axis.

Usually, the absorption maxima appeared more distinctly when the color centers were in their saturated state, and the maximum of 600m $\mu$  appeared when the samples were irradiated to an exposure of more than 10<sup>7</sup>R. The bleaching action of the color centers occurred most prominently in the temperature range from 220°C to 260°C, and the absorption maxima decayed almost completely at about 260°C, and especially the decay of the maximum of 600m $\mu$  was more pronounced as the peak was tiny. At the beginning of the bleaching, the points of absorption maxima tended to shift toward the longer wavelength in accordance with the decrease of the saturated state of the color centers.

When a  $\gamma$ -irradiated quartz sample is heated, a trapped electron in a  $\times P_-$  defect is released to the conduction band,<sup>(17)</sup> thus it will recombine with a trapped hole on the  $^A A_+$  defect to yield the thermoluminescence, and eventually an original progenitor  $^A P$  defect is formed again, that is,



From the above explanation, the  $^A A_+$  defect is the luminophor center as well as the smoky color center which will cause the optical absorption, while the  $\times P_-$  defect is the trapping center of an electron. Strictly speaking, the  $\times P_-$  defect is closely related to the characteristics of the  $^A A_+$  defect.

Megla<sup>(18)</sup> suggested that thermal bleaching of the color centers in SiO<sub>2</sub> network was caused by thermal excitation of the trapped electrons, and

the optical bleaching was caused by an excitation of holes. And Lell<sup>(19)</sup> suggested that the absorption intensity varied with type of alkali though the wavelength of the absorption maximum is independent of the alkali. Bleaching due to the trapped electrons in the shallow traps of the  $\times P_-$  centers and the related  $^A A_+$  centers can be seen at around 100°C in Fig.8.

The number of  $^A A_+$  center should be equal to that of  $\times P_-$  center which can be regarded as a type of F center. An approximate density of F center has been calculated by Smakula<sup>(20)</sup> with the following formula,

$$n = A a m W, \dots \dots \dots (2)$$

where A is the constant of proportionality; am is the maximum factor of the absorption, and W is the half-width of the absorption band.

(5) Content of Luminescent Impurities in  $\alpha$ -Quartz Materials

Table 1. The result of activation analyses

| Sample Number | Al content (ppm) | Na content (ppm) | Al/Na   |
|---------------|------------------|------------------|---------|
| A-1           | 1000-1200        | 3.0-4.0          | 330-300 |
| B-1           | 100-120          | 0.9-1.1          | 110-100 |
| C-1           | 90-110           | 0.9-1.1          | 100-100 |
| D-1           | 45-55            | 2.-3.0           | 22-18   |
| E-1           | 65-95            | 7.0-9.0          | 9-8     |

The impurity content in several species of  $\alpha$ -quartz materials has been examined by the method of neutron activation analysis. It is known that the necessary impurities in  $\alpha$ -quartz for its thermoluminescence are the aluminum and alkali ions which are constituting of  $^A A_+$  and  $\times P_-$  defect centers. The determined contents of Al and Na are as shown in Table 1. The principal impurities in  $\alpha$ -quartz materials are Li and Na, and has been detected<sup>(21)</sup> by the colorimetric analysis that the concentration of Li is about 10 percent of that of Na in  $\alpha$ -quartz. The content of Li can not be detected by  $\gamma$ -ray spectrometry after neutron activation, because the nuclides which were formed by ( $n, \gamma$ ) or ( $n, \alpha$ ) reactions are pure  $\beta$  emitters. Though the exact concentration of Li in  $\alpha$ -quartz is not known by this analytical method,

the influence of Li content on the emitability of thermoluminescence in  $\alpha$ -quartz may be important. The sample A-1 is one of the the most  $\gamma$ -sensitive species of type 1  $\alpha$ -quartz, and the  $\gamma$ -sensitivity is gradually decreased in turn in the listed samples. It is shown in Table 1 that the Al/Na in A-1 sample is largest among the listed samples. This point can coincide with our former result (Ref. 7), but it seems that the various deeper discussions pertaining to the optimum concentrations of Al and Na ions in type 1  $\alpha$ -quartz for the best emitability of thermoluminescence need further research.

## 2. Properties of Pulverized Quartz Dosimeter

### (1) Linearity of Dose

The linearity of thermoluminescent intensity with comparatively low doses was measured in the range from 50R to about 2000R by using the pulverized samples of type 1  $\alpha$ -quartz, and a plot of the glow peak heights is shown in Fig. 11. It can be pointed out that the sensitivity for  $\gamma$ -radiation in the pulverized sample was greatly impro-

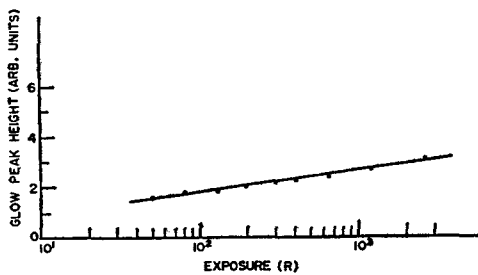


Fig. 11. Response of the pulverized quartz dosimeters vs. exposure (low dose).

ved in comparison with the case of bulk crystal samples though the effect of glow enhancement was also probably exercised partly by the preparation due to the Ichigawa's method (Ref. 11) which was done before its crushing, that is, the minimum detectability in the former was improved to be about 40 times as much as that in the latter. It is seen in Fig. 11 that each glow peak height responds linearly with dose of the measured range.

As for the DC high voltage at present device, 900 volts was applied to the PM tube circuit for the safe handling and its longer life time, but if the voltage is increased to the value of allowable rating, the minimum detectability of dose should become far lower, probably less than 1R, though it was not tested at present investigation.

### (2) Nonlinear Effect in T.L. with Doses

Nonlinear effect in thermoluminescent intensity with doses due to the three grain sizes of pulverized samples was observed at the region of high

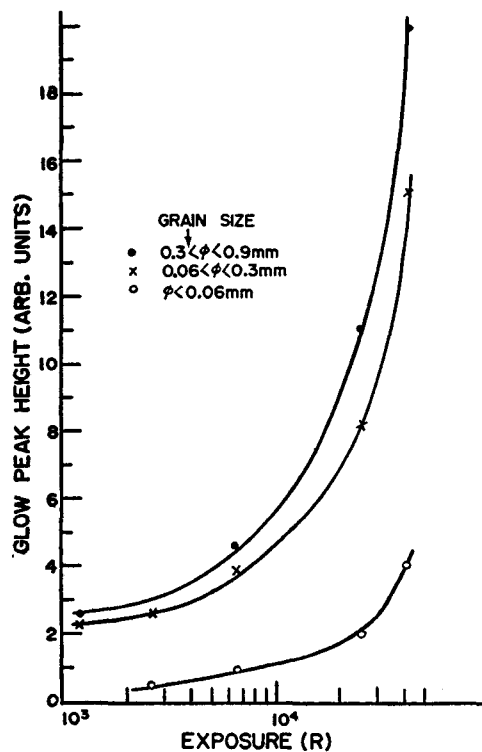


Fig. 12. Response of the pulverized quartz samposure showing the nonlinearity of dose and the effect of grain size.

dose rates as shown in Fig.12. It is seen in the figure that, in proportion as the dose decreases in each curve, T.L. intensity is exponentially decaying. It seems that this range of doses with high dose rates is not suitable to be detected by the pulverized samples. As has been described in part III-1-(1), this range of doses could be detected linearly by using crystal bulk samples. It may be inferred that such exponential decay of thermo-



luminescence with respect to the decrease of dose is probably caused by the nonlinear heat transmission in the pulverized samples. Especially, at such region of high dose rates, decaying rate of thermoluminescence should be pronounced on account of the nonlinear heat transmission among the grains though this effect is negligible at the region of dose rates as described in part III-2-(1). It is also seen in Fig. 12 that the  $\gamma$ -sensitivity is greatly decreased with diminishing grain size.

### (3) Effect of Grain Size

The size of the grain is an important factor to increase glow intensity in the present investigation. Five sizes of the grain as shown in part II-(2) were used for this measurement. The dependence of thermoluminescence intensity on the grain size is as shown in Fig. 13. It is shown in the figure that the glow intensity is decreased with diminishing grain size whereas the grains having the each size of  $0.3 < \phi < 0.9\text{mm}$  shows the best glow intensity.

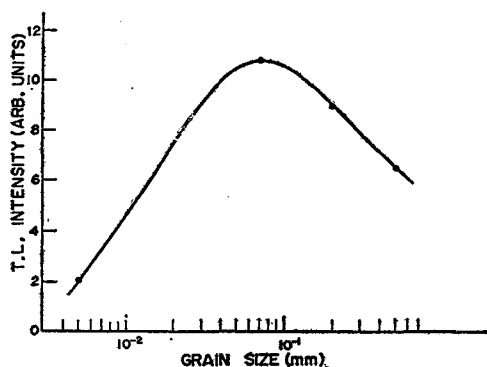


Fig. 13. The changes in T.L. intensity of pulverized quartz samples vs. grain size.

It seems that, the thermoluminescent light is more easily emitted from a grain having a suitable size than the same size of a crystal domain in a bulk crystal, and also a minimum possible crystal size may be necessary for the process of unit recombination between an electron and a hole. Another reason of decreasing the thermoluminescence at large grains having each size of  $5.0\text{mm} < \phi$  (rough crushing) may be in incomplete heat transmission among the grains. By repeated

use of the grains, the effect of glow enhancement due to the grains was reduced a little regardless of their grain size though the degree of reducing due to the grain size is different. It can be inferred that, in the first use of the grains, the effect of tribothermoluminescence<sup>(22)</sup> may be partly exercised in the glow intensity.

### (4) Effect of Heat Treatment

The effect of heat treatment on the pulverized quartz was examined. The pulverized quartz having the grain size of  $\phi < 0.06\text{mm}$  was used for the heat treatment. As the crystallinity can only be maintained without cracking within the transition temperature of  $573^\circ\text{C}$ , pulverized quartz of above-written grain size could only be used for the heat treatment of the temperature of higher than  $700^\circ\text{C}$ .

Table 2. Annealed Conditions of Pulverized Quartz Samples

| Sample Number | Grain size (mm) | Annealed time (min.) | Annealed temp. ( $^\circ\text{C}$ ) | Atmosphere |
|---------------|-----------------|----------------------|-------------------------------------|------------|
| A-30          | $\phi < 0.06$   | 30                   | 700                                 | Air        |
| V-30          | "               | 30                   | 850                                 | Vacuum     |
| V-60          | "               | 60                   | 850                                 | Vacuum     |
| H-30          | "               | 30                   | 900                                 | Hydrogen   |
| N-30          | "               | 30                   | 900                                 | Nitrogen   |

The annealed conditions of pulverized quartz samples are as shown in Table 2. Unexpectedly, no samples have shown the enhancement of glow intensity, and on the contrary, they have become quite insensitive even for a large dose.

This reason can be inferred as follows. Before  $\gamma$ -irradiation, an alkali ion is electrostatically combined to the  $A_1A_+$  defect to form a progenitor  $A_1P$  defect for its charge compensation, and is to be released into the crystal as a  $xP_-$  defect if the crystal subjected  $\gamma$ -irradiation. In proportion as the grain size becomes smaller, displacing scope of alkali ion through the interstitial channel is confined more and more. The thermal energy due to the heat treatment may urge the release of alkali ion from the progenitor, and the released alkali ion will be escaped from the surface of a crystal grain through the dangling bonds of  $\text{SiO}_2$ .

The heat treatment of pulverized quartz for the enhancement of thermoluminescence has been examined by Bayley and Holzapfel<sup>(23)</sup> Though they have had a slight improvement of thermoluminescence by mixing a few additives into the quartz powder, it seems that the description is not enough for the attempt of its reproducing. The detailed information of this point is expected to the further research.

#### (5) Therapeutic Application

Lethal doses for whole-body irradiation are generally much lower than for partial-body irradiation, and the demarcation of the part of body irradiation is of great importance. Successful radiotherapy is based on this circumstance. It should be very helpful to have detectors judiciously placed in body regions particularly liable to exposure. The pulverized quartz dosimeter can be used for such purpose.

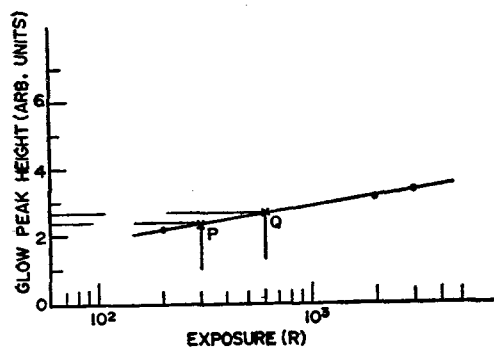


Fig. 14. Response of the pulverized quartz dosimeters vs. exposure (low dose) showing a therapeutic application.

A pulverized quartz dosimeter contained in a small polyvinyl thin package can be inserted in any part of body regions as the form of package can be changed freely, so that the actual absorbed dose at the body region can be exactly detected.

The measured examples of absorbed doses in body regions of two cancer patients are as shown in Fig. 14. First, the standard linear line was drawn by the values of three glow peak heights and their each corresponding  $\gamma$ -dose read by

Victoreen R meter. Next, the irradiated doses of 300R and 600R were read by the Victoreen R meter after placed the sample packages on body regions of two patients.

By measuring each glow peak height of two samples, P and Q points could be marked on the standard line. If these two doses are unknown or different from the irradiated dose, P and Q points will tell the correct value of the absorbed doses.

#### IV. Conclusion

The linear detectable range of unprepared type 1  $\alpha$ -quartz dosimeters was from  $2 \times 10^3$ R to  $2 \times 10^6$ R. When the samples were prepared by Ichigawa's method (Ref. 11), their  $\gamma$ -sensitivity was fairly increased, but they were not enough for low dose dosimetry. The pulverized quartz dosimeter having the grain size of  $0.3 < \phi < 0.9$ mm exhibited far greater enhancement of the glow intensity and enabled the linear measurement of doses in the range from 50R to 2000R. Such linear region of doses due to the pulverized samples was applied successfully to the measurement of absorbed dose in a body region of a cancer patient. Since the quartz dosimeters had the advantage of low fading, they could be used in an illuminated room with room temperature without wrapping them up by black papers.

The attempt of improving the thermoluminescent emitability in pulverized quartz sample due to the heat treatment under various atmosphere was not successful.

The influence of "before glow" on thermoluminescent glow curve measurements due to the quartz sample seems to be important in case that the glow peak height is taken for the  $\gamma$ -ray dosimetry, and the detailed description of this problem is to be appeared in the Ref. 9

Though the measurement of energy dependence of the quartz dosimeter was unable on account of the lack of the irradiation facilities, it can be inferred that their energy dependence should be akin to that of a glass dosimeter,

Judging from the various advantages of the pulverized quartz dosimeter such as, low fading,

abundant material, easy deformability in its packing, easy reading of the dose due to a single peak, and simple mechanism of the reader, it can be expected that it will be easily utilized in this country for therapeutic application to the cancer patients.

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