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Some Characteristics of Teflon-Thermoluminescent Dosimeters

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Abstract

The characteristic thermoluminescence responses of Teflon thermoluminescent dosimeters to radiations have been studied by the variation of radiation qualities as well as the high dose radiations. The change in the sensitivity of TLDs for different radiation qualities were studied through not only the photon energy dependence but also the change of supralinearity on the photon energy dependence, by exposing ^{60}Co gamma rays, the effective X-rays of 44keV, 69keV, 108keV, and thermal neutron of 0.04eV. The results were as the following:

The TL response of T-$CaSO_4$: Dy as a function of absorbed dose was linear up to about 5 Gy, and the response beyond 5Gy was supralinear for ^{60}Co gamma rays. The supralinearity of T-LiF-7 became noticeably apparent more than that of T-$CaSO_4$:Dy and also the lower the LET of radiation became the higher the supralinear effects were. No supralinearity appeared for the thermal neutron irradiations equivalent to 10Gy of ^{60}Co gamma rays.

The relative sensitivities (Rs), which depended on the doses of ^{60}Co gamma rays to the TLDs of T-LiF-7 and T-$CaSO_4$:Dy could be, respectively, approximated to the following empirical formula fitted by the least square method:

$R_{LiF}=1.021-0.04581\ \log D+0.402(\log D)^2-0.405(\log D)^3,\ \ 5\times10^3\geq D\geq1(Gy)$

$R_{CaSO_4}=0.976-0.03241\ \log D+0.262(\log D)^2-0.298(\log D)^3,\ \ 5\times10^3\geq D\geq1(Gy).$

I. Introduction

Recently, thermoluminescent dosimeters(TLDs) have been widely used for radiation dosimeters. Many kinds of phosphor powder for TLDs have been developed and studied by a number of investigators.

Many phenomena related to the thermoluminescence (TL) have been known since 1940's, and then the possible application of TL in the field of radiation dosimeter was suggested by Daniels[1,2] for the first time. From then on, numerous investigations on the thermoluminescence characteristics of various phosphors have been made.

Randall-Wilkins[3] were the first to investigate the thermoluminescence theoretically. They used a model in which electrons in metastable states were thermally raised to an excited state, from which the electrons returned to the ground state with emission of luminescence.

Halperin and Branner[4] suggested a model which was recombined or retrapped by electrons of the luminescence centers excited thermally.

This model was, therefore, introduced and compensated for the concepts of competing traps by N. Kristianpoller. And also Cameron et al.[6] have studied the characteristic response of the supralinearity of thermoluminescent dosimeters, TLD-100 and H13A, for high dose, and proposed its mathematical model.

In order to compensate the imperfection in this model, they proposed a competing trap model. On the other hand, Attix et al.[7] have suggested the track interaction model to testify to the LET dependence of supralinearity. However, Cameron et al. have carried out an experiment for the energy dependence of the thermoluminescence on TLD-100.

Nakajima[9] has investigated the changes of the sensitization on the annealing and reuseability ot the TLDs, Carlsson[10] has done the changes of the glow curves depending on the LET of the radiation, assuming that the TLD might serve as a LET meter, and Watanabe et al.[11] have studied the dependence of TLD-100 response in temperature during exposure.

As shown above, many researches have been done in powdered form TLDs such as TLD-100, TLD-600, TLD-700, CaF_2 and $CaSO_4$: Mn., whereas researches for LiF and $CaSO_4$: Dy embedded in Teflon matrix were very few.

Therefore, in this research the various behaviors of thermoluminescence of Teflon dosimeters for high dose radiations have been investigated. Especially, in order to study the characteristic responses of the supralinearity of Teflon-TLDs, the radiations of the different qualities of ^{60}Co gamma rays and the effective X-rays of 44 keV, 69 keV and 108 keV were exposed to the dosimeters, T-LiF-7 and T-$CaSO_4$: Dy, respectively, but the dosimeters of T-LiF-6 were irradiated by thermal neutrons.

An attempt was made to see the phenomena of the sensitization effects by the repeated irradiation method for the same dose, and the empirical formula by the method of least squares[12] were attempted to derive from the relative sensitivities of the experimental results as a function of absorbed dose to the T-LiF-7 and T-$CaSO_4$: Dy.

II. Experimental methods and procedures

1. The repeated irradiation method

Three kinds of TLDs used in this study were T-LiF-7 (12.5mm×0.4mm). T-$CaSO_4$: Dy (12.0mm ×0.4mm), and T-LiF-6 (11.3mm×0.4mm) of Teflon matrix discs containing 30% of phosphors by its weight.

In order to eliminate the background of these dosimeters, all of the TLDs were initially given the standard annealing prior to the first irradiation of high doses with different radiation qualities. T-LiF-6 were annealed for 8 hours at 300°C followed by 16 hours at 80°C, and the TLD, T-$CaSO_4$: Dy was annealed for 4 hours at 300°C and followed by 8 hours at 80°C.

The annealed T-LiF-7 and T-$CaSO_4$: Dy were used not only to study the determination of the TL versus Gy response curve but also to measure the sensitization effects happened by exposing ^{60}Co gamma rays in the range of 1Gy to 5×10^3Gy, and all the annealed T-LiF-7 and T-$CaSO_4$: Dy were irradiated to the effective X-rays of 44keV, 69keV and 108keV in the range of 1Gy to 10^2Gy.

The TLD, T-LiF-6 was sensitized by ^{60}Co gamma rays in the range of 1Gy to 3×10^2Gy, and then irradiated by the thermal neutron beam flux with energy of 0.04eV as a test dose equivalent to ^{60}Co gamma rays. All the irradiated samples were annealed for 10 min. at 100°C prior to the measurement of TL to remove any influence of the low temperature peaks from the glow curve. And then all TLDs were read out within 24 hours after the first irradiation. The TL from each sample was measured in order to obtain the glow curve and the integrated total light output, and then the total process of the measurement was called the first reading. After the first reading, all of the samples were annealed again for one hour at 280°C. This annealing apparently removed all the backgrounds of TL in the main glow curve peaks but gave little effect on the sensitivity. After these procedures each group of TLDs was exposed to different radiation qualities with the same doses, respectively.

According to the first irradiation method given by the above procedures, the dosimeters were annealed at 100°C for 10 min. to ensure the emptying of all electron traps prior to the second reading, and then the same method as done previously. This process that has been done in the above was called the repeated irradiation method.

2. The heating cycles

Fig. 1 showed the glow curve and the associated heating cycles for the TLD-7300 reader used in this study. These heating cycles were the most sensitive temperature ranges in operating of this work. In the Fig. 1, the preheat cycle showed the temperature of about 135°C in the heating rate of approximately 34°C/sec. and was maintained for 8 sec. The glow peak (A) followed the preheat cycle. The purpose of preheat was made to bleach out any shallow traps in the TLDs. The readout temperature was raised from 135°C to pre-selected readout temperature 285°C, and then the total time of readout was 12 seconds.

3. Thermal Neutron irradiations

The responses of T-LiF-6 to thermal neutron have been studied by exposing samples to the neutron beam from 2MW TRIGA Mark III Reactor installed at the Korea Advanced Energy Research Institute (KAERI). For thermal neutron exposure, samples of a thin plate in rectangular area of 36cm^2 were attached uniformly to neutron beam port which was the main source of thermal neutron.

At 1.2Mw reactor power the thermal neutron beam flux with energy of 0.04eV was 1.8×10^5n/cm^2-sec. at the sample position. Thermal neutron dose was measured in terms of SI unit equivalent to ^{60}Co gamma rays, which was defined as the thermal neutron dose incurred by the phosphor which yielded an amount of TL equivalent to 1Gy of ^{60}Co gamma rays.

T-LiF-6 samples in the present investigation were irradiated a test dose of 1Gy to 10^2Gy. The thermal neutron fluence equivalent to 1Gy of ^{60}Co gamma rays and the time required irradiation of thermal neutron were 2.77×10^9n/cm^2 and 256min., respectively

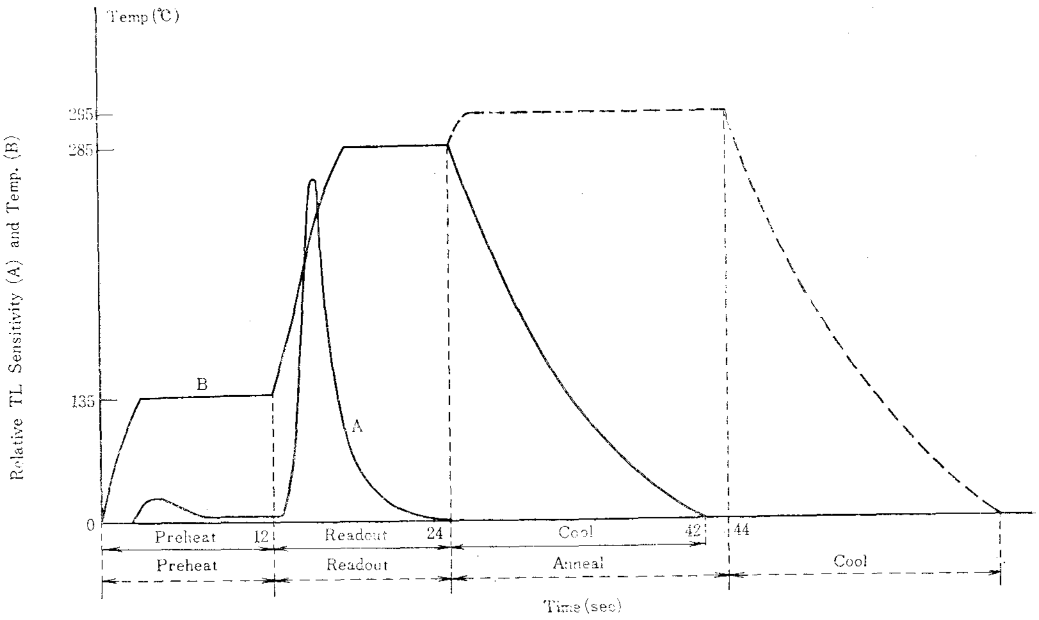

Fig. 1. Heating Cycles

4. Gamma-ray irradiations

Gamma-ray irradiations were carried out by using a ^{60}Co irradiation facility at the KAERI. The source strength was about 1.85×10^{15}Bq. The output was determined by using a Victoreen-550 ionization chamber which was calibrated at the Health Physics Department at the KAERI. Some of the standards annealed samples were enclosed in the thin paper bags of 3.2cm×4.1cm, and these samples were attached to a sample holder which was located 28cm high from the ground during the irradiation. At a distance of 282cm from the source the dose rate was 0.1Gy/min and it was 0.5Gy/min. at 116cm.

5. X-ray irradiations

X-ray irradiations were carried out by using a X-ray apparatus (Model SHT 250M-2type, 250KVP, 25mA) installed in the X-ray irradiation room at the KAERI. The dose rates at the distance of 47cm apart from the focal spot were 0.88Gy/min (250KVP, 19mA), 1.73Gy/min (150KVP, 25mA) and 1.27Gy/min (100KVP, 25mA), respectively, and the output was also determined by using a Victoreen-550 ionization chamber as in the gamma ray irradiations.

The dose ranges in this study were 1Gy to 10^2Gy and they were exposed to the two types of the TLD Teflon dosimeters, T-LiF-7 and T-$CaSO_4$: Dy, respectively.

A specially made probe holder was fixed to the position of the dosimeters accurately so that it might coincide with the center of the collimator of X-ray apparatus, and then the radiation exposure was carried out.

Table 1 shows the results of the experimental determination of the effective photon energies of X-ray beams through the filters, and the results of the experimental determination were compared with those of the theoretical computation by the method in the literatures.[13~16]

Throughout the experimentation each group of the batches contained five dosimeters for exposing the different radiations.

III. Results and discussions

The glow curves of TLDs probably best characterized the distribution of trap depth, and so the knowledge of the glow curves including temperature was essential in resolving anomalies of operation.

As has been pointed out earlier, one of the principal aim of this study was to investigate the response of TLD to different radiations.

All of the heating cycles were constantly maintained throughout the experiments. Therefore, the slight variations in the glow curves didn't appear due to the constancy of the TLD reader.

As shown in Fig. 2, these glow curves showed the temperature including the heating cycle when the TLDs, T-$CaSO_4$: Dy, 12.0mm diameter by 0.4 mm thick were exposed to 5Gy and 10^2Gy of gamma radiations, respectively.

The sharp peaks as shown in Fig. 2 were contracted by one-tenth when the currents of the glow curves overflowed in the range of the reader by the automatic relay. When these two glow peaks were compared each other the phenomena of the low temperature peaks were obviously same in the range of preheat cycle, but at higher irradiation level(10^2 Gy) the main glow peak showed a broad peak which was not observed from the other TLDs used in this investigation. The observation of this peak seemed, however, to be different markedly from the

Table 1. Results of Experimental Determination of the Effective Energies of X-ray Beam Through the Filters.

Filter material (foils)	Thickness(mm)	Max. energy of X-ray(KVP)	Appro. theoretical energy(keV)	Experimental results, K_{eff}(keV)
Al	1.992Al	100(25mA)	41	44
Cu · Al	0.284Cu+0.987Al	150(25mA)	69	69
Cu · Al	1.004Cu+0.982Al	250(19mA)	106	108

glow peak at 220°C for low doses, but the observed peak shape in the exposure more than 10^2Gy explained the existence of another glow peak at 250°C.

That is to say, the electron traps of the TLD Teflon dosimeters, T-$CaSO_4$: Dy, seemed to be different from those of the afterglows for higher doses to the LiF-Teflon dosimeters.

As shown in Fig. 2, even though the position of the main peak seemed to appear after the temperature reached to 285°C, this phenomena caused short lapse of the time between the temperature due to the rapid heating rate and the internal temperature of the TLD depending on the thickness of 0.4mm which was rather thick. According to the effect of heating rates on the glow curves, the faster heating rates gave narrowed peaks with greater peak heights,[17] so that the slowness of heating rates might have calibrated the main glow peak.

The response of the TL per absorbed dose was an important characteristic of TLDs, that is, as the absorbed doses got out of constant proportional region, the response of the TL, eventually, led to its supralinearity. Several models have been proposed to explain supralinearity on the basis of the following reasons: an increase in number or competitiveness of electron traps,[18,19] that of luminescence centers,[20~24] and a combination of the models in the above. As previously explained, such a supralinearity characteristic of the powdered form of TLD-100 has been reported in many other papers, but few works have been done for characteristic of the dosimeters embedded in Teflon matrix except Gorbics et al.[25]

Figures 3, 4, 5 and 6 showed the experimental results of the characteristic responses of the X-and

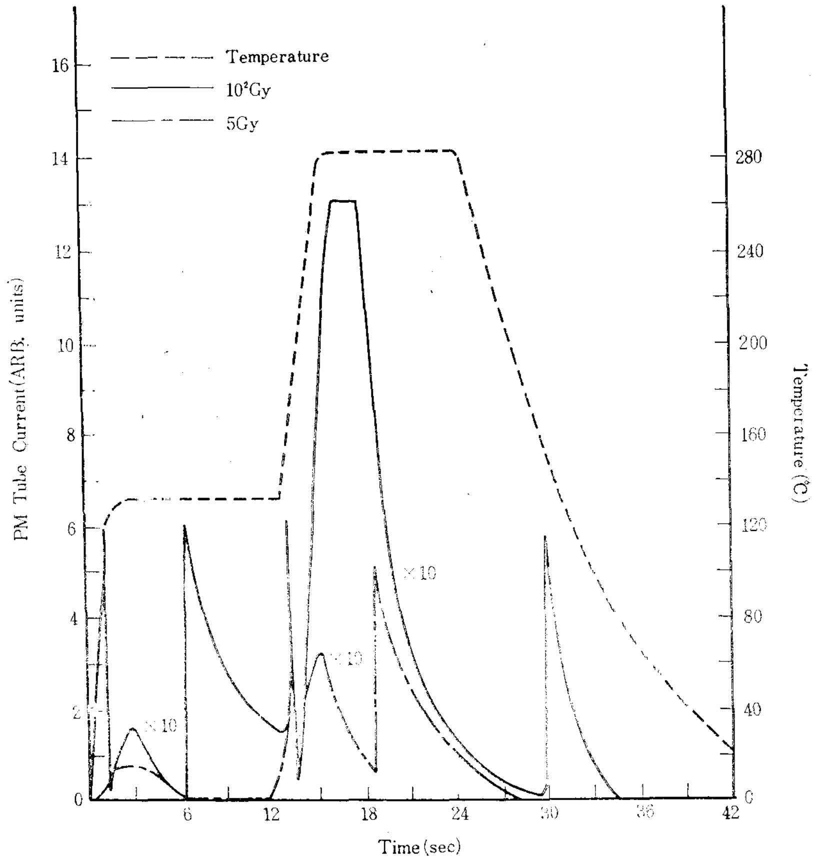

Fig. 2. Thermoluminescence Glow Curves of T-$CaSO_4$:Dy at 5Gy and 10^2Gy Exposure Level

gamma radiations. Thus the Figures explained that as the effective photon energies of the X-and gamma radiations increased, the greater amount of supralinearities appeared apparently.

When the energies of the incident radiations increased, the average energy of the electrons, which was created by Compton scattering within TLDs and/or Photoelectric effects, increased, while LET decreased. Thus, the higher the linear energy transfer of the radiation became the less the supralinearity was. Conversely, it seemed that a relatively small increase of LET caused the thermoluminescence efficiency of the TLDs, T-LiF-7 and T-Ca SO_4: Dy, to increase in the low dose linear region.

A particular characteristic response of the second irradiations in the experiments was found to be almost 1.2 to 1.5 times of the sensitivity for low doses when compared with the dosimeters of the first radiations which were pre-exposing history. The identity of sensitivities of the two TLDs, T-$CaSO_4$: Dy and T-LiF-7, for ^{60}Co gamma rays happened when the absorbed dose was over 10^2Gy, and then the sensitivities of the first and second irradiations became identical in each for about 3×10^2Gy in the case of T-LiF-7 and for about 5×10^2Gy in the case of T-$CaSO_4$: Dy.

On the other hand, several investigations have reported the measurements of neutrons with LiF.[26-28] The most obvious application of LiF for neutron measurements arose from the fact that the ^{6}Li(n,α)^3 H reaction had a thermal neutron cross-section of about 950 barns.[29-30] So the fission products of the ^{6}Li-neutron were a 2.07MeV alpha particle with an average LET of 160keV/μ and a 2.74MeV triton with an average LET 120keV/μ, and these particles were the source of high LET radiation.

The average LET value for these particles, as reported by Wingate et al.[28] from earlier calculations of Brownell and Sweet, was 140keV/μ, which was some 246 times of that for ^{60}Co gamma radiation. As the results of this study, it was found that there were no supralinearities from the thermal neutron irradiations and the response was linear from 1Gy to 10^3Gy as in Fig. 7.

In order to study the cause of the supralinearity response for the radiation of low LET, the relative total reading(TL) per absorbed dose was normalized to unity at 1Gy for all of the photon energies. The TL/Gy values obtained for the irradiations of the effective x-rays were seen in Figure 5 and those obtained for the ^{60}Co in Fig. 6, because the log-log graphs in the Figures 3 and 4 were partially insu-

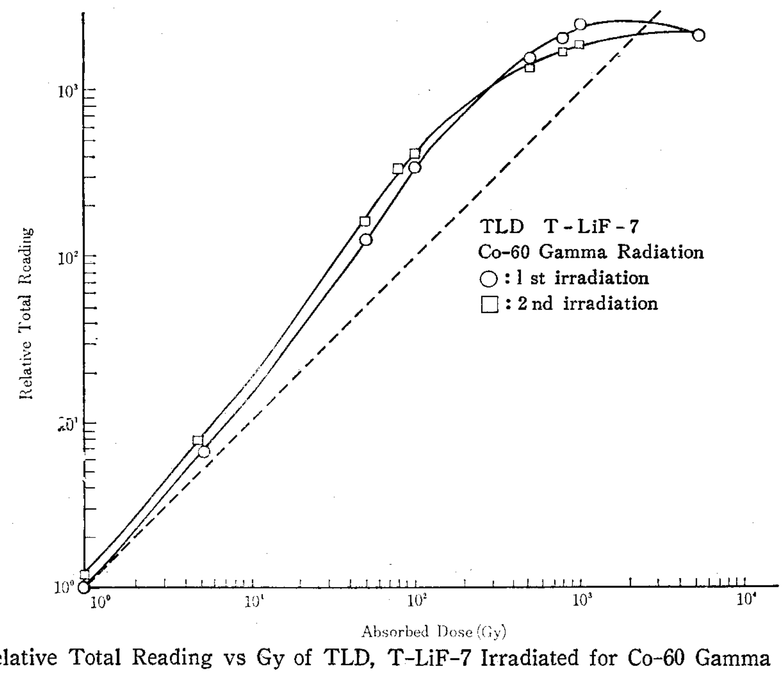

Fig. 3. Relative Total Reading vs Gy of TLD, T-LiF-7 Irradiated for Co-60 Gamma Radiation

fficient for the characteristic responses of TL.

Comparing these two values in the Figures each other, the T-LiF-7 dosimeters showed more non-linearity than T-$CaSO_4$: Dy for ^{60}Co gamma irradiation. But the sensitivities of the former dosimeter showed a maximum sensitivity at about 10^2Gy and the sensitivity beyond 10^2Gy began to decrease again, while those of the later dosimeters decreased nearly at about 80Gy. The fact that the sensitivity decreased again for very high dose could be described as the reduction of thermoluminesence behavior in the crystal of the TLDs effected by radiation damage.

The repeated irradiation seemed to be considerable drop in sensitivity compared with the first irradiation, and this result was almost the same as those of Marrone and Attix[31] and the others.[32]

In order to determine the mathematical formula of the changes in the sensitivities of Teflon TLDs, the data based on Figures 3 and 4 were shown as a function of absorbed dose, and then the response of thermoluminescence for the absorbed dose in terms of 1Gy was approximated to the third-order equation form as the following:

$$R=a+b\ \log D+c(\log D)^2+d(\log D)^3,$$

where $a,b,c,$ and d were constants to be determined, and D was the absorbed dose in terms of Gray (Gy). The calculation was carried out by the method of least squares[12] using the LINREG Code developed by P.S. Inglish at Arizona University to modify the RINREG Code in accordance with the computer CYBER-73, and the following resuls were obtained:

for T-LiF-7,

$$R_{LiF}=1.021-0.04581\ \log D+0.402(\log D)^2 -0.405(\log D)^3,\ 5\times10^3\geq D\geq 1(\text{Gy}),$$

and for T-$CaSO_4$: Dy,

$$R_{CaSO_4}=0.976-0.03241\ \log D+0.262(\log D)^2 -0.298(\log D)^3,\ 5\times10^3\geq D\geq 1(\text{Gy}),$$

respectively.

The results obtained with these procedures were identical within the experimental errors of $\pm 8\%$. The photon energy dependence of thermoluminescent

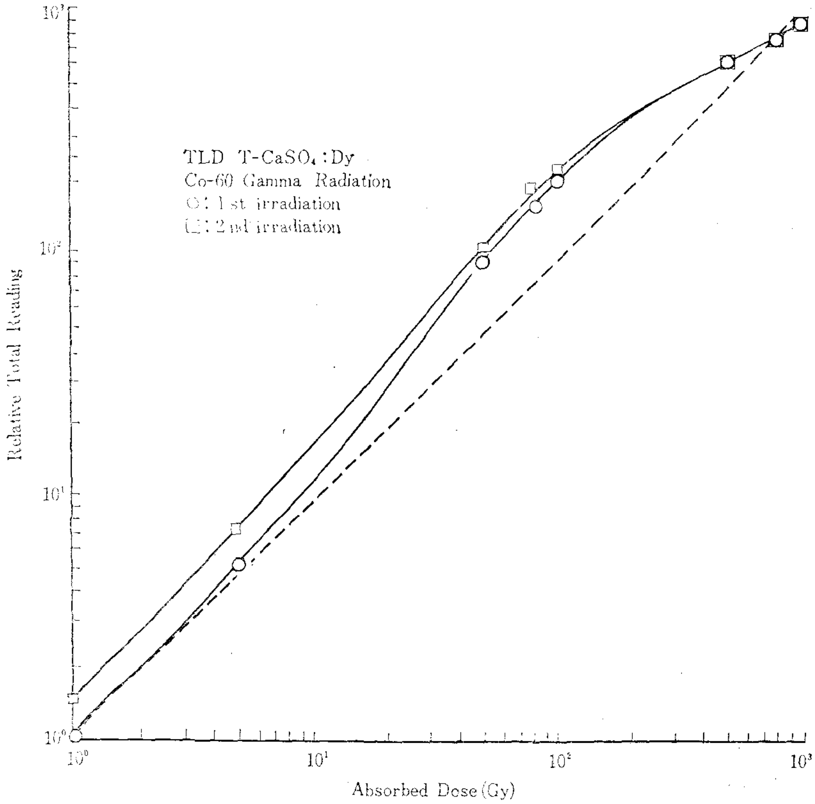

Fig. 4. Relative Total Reading vs Gy of TLD, T-$CaSO_4$: Dy Irradiated for Co-60 Gamma Radiation

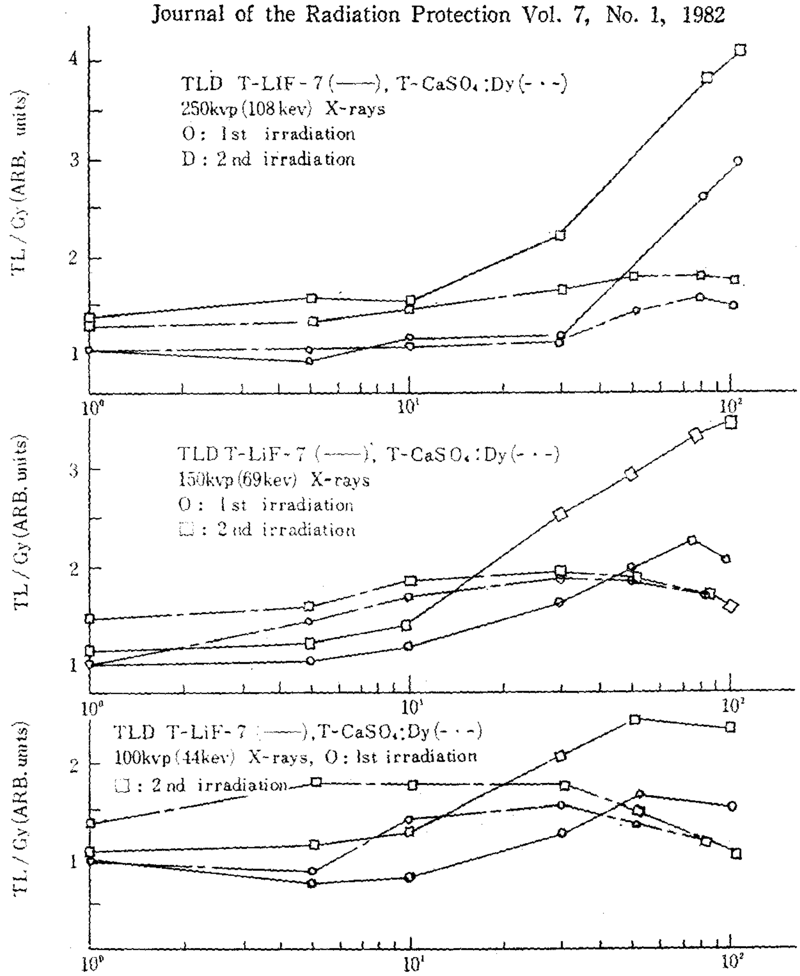

Fig. 5. The Relative TL Response (TL/Gy) of TLD T-LiF-7(——) and T-$CaSO_4$:Dy(— · —) Irradiated for the Effective X-rays of 44kev, 69kev and 108kev, Respectively.

dosimeters for low dose was not so significant, but as described early in this study the TL response showed considerable amount of difference by the energy of radiation quality for high dose. When compared with the results of the LiF dosimeters in powdered form and/or in crystal form performed by other investigators, the Teflon LiF-7 dosimeters showed the supralinearity for low doses.

Ⅳ. Conclusion

The characteristic thermoluminescence responses of TL-Teflon dosimeters to radiations have been studied by the variation of radiation qualities as well as the high dose radiations. The conclusion was as the following:

(1) The TL response of T-$CaSO_4$:Dy was linear up to 5Gy, and that of T-$CaSO_4$:Dy supralinear to 15 Gy for ^{60}Co gamma radiation.

(2) The supralinearity of T-LiF-7 became noticeably apparent more than that of T-$CaSO_4$:Dy.

(3) The lower the LET of radiation became the higher the supralinear effects were seen to 30Gy.

(4) No supralinearity appeared for the thermal neutron irradiations equivalent to 10Gy of ^{60}Co gamma radiations.

(5) The relative sensitivities (Rs) which depended on the doses of ^{60}Co gamma radiations to the TLDs, T-LiF-7 and T-$CaSO_4$: Dy could be respectively approximated to the following empirical formula fitted by the least square method:

$$R_{LiF}=1.021-0.04581\ \log D+0.402(\log D)^2$$

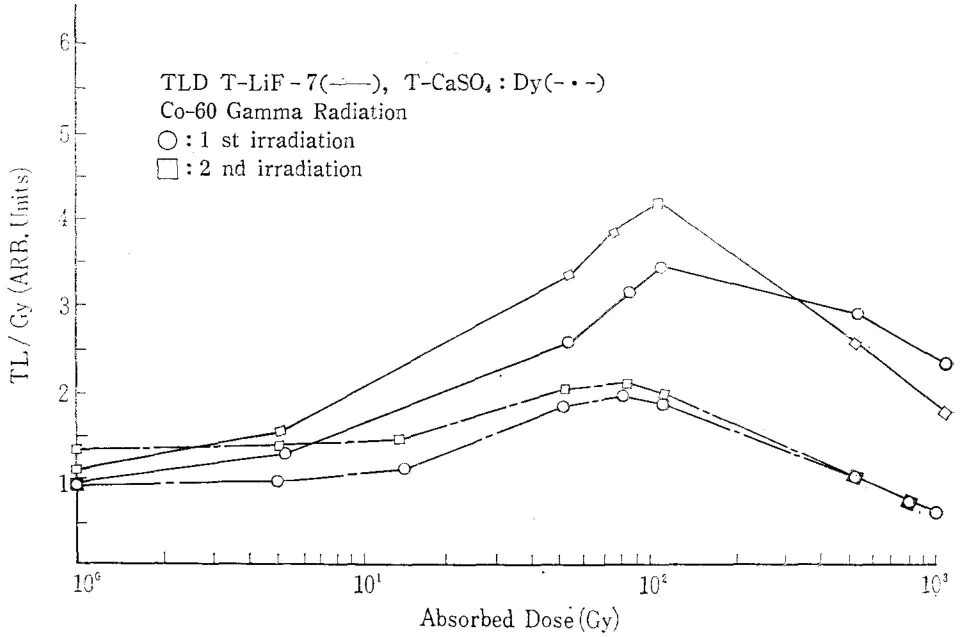

Fig. 6. TL/Gy vs Gy of TLD, T-LiF-7and T-CaSO$_4$:Dy Irradiated for ^{60}Co Gamma Radiation.

$$-0.405(\log D)^3, \quad 5\times10^3 \geq D \geq 1(Gy)$$

and

$$R_{CaSO_4}=0.976-0.03241 \log D+0.262(\log D)^2$$
$$-0.298(\log D)^3, \quad 5\times10^3 \geq D \geq 1(Gy)$$

respectively.

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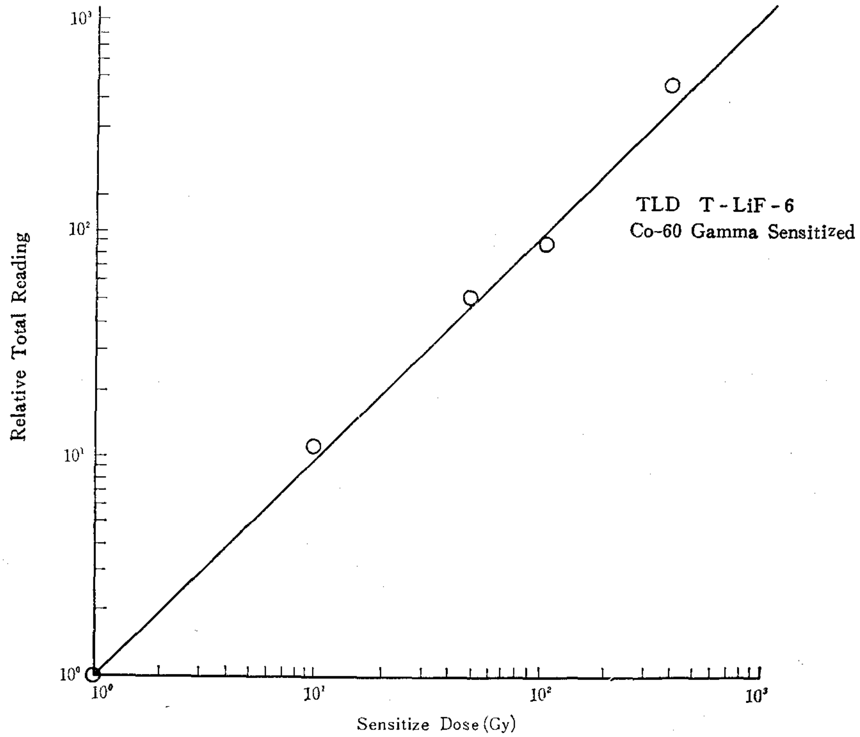

Fig. 7. Variation of Relative Total Reading of TLD T-LiF-6 Irradiated for Thermal Neutron of 0.04ev after Sensitized with Co-60 Gamma Radiation.

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테프론 熱螢光線量計의 特性

李 秀 容

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抄 錄

放射線의 線質 및 高線量의 變化에 따른 테프론 熱螢光線量計의 熱螢光感應特性을 硏究하였다. 相異한 放射線의 線質에 대한 TLD感度의 變化는 ^{60}Co감마線, 有效에너지가 44keV, 69keV, 108keV인 X線 및 0.04eV의 熱中性子에 照射시킴으로서, 光子에너지 依存性 뿐만 아니라 이에 따른 超線形特性의 變化도 조사하였다. 그 結果는 다음과 같다.

^{60}Co감마線에 대해서 吸收線量函數로서의 T-$CaSO_4$:Dy의 TL感應은 5Gy까지는 線形的이고, 5Gy 이상에 대해서 이 感應은 超線形的이었다. T-LiF-7 線量計의 超線形特性이 T-$CaSO_4$:Dy에 비해 顯著하였으며, 放射線의 LET가 낮으면 낮을수록 超線形效果가 더욱더 커짐을 알 수 있었다.

^{60}Co 감마線 10Gy에 等價하는 熱中性子 照射로 부터는 超線形特性이 觀察되지 아니하였다. TLD, T-LiF-7 및 T-$CaSO_4$:Dy의 ^{60}Co 감마線의 線量에 따른 相對感度(Rs)는 最小自乘法으로 다음과 같은 實驗式.

卽, $R_{LiF}=1.021-0.04581\ \log D+0.402(\log D)^2-0.405(\log D)^3,\ 5\times10^3\geq D\geq1(Gy)$

및

$R_{CaSO}=0.976-0.03241\ \log D+0.262(\log D)^2-0.298(\log D)^3,\ 5\times10^3\geq D\geq1(Gy)$

으로 各各 近似시킬 수 있었다.