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■ 論 文 ■

Anneal Characteristics of LiF:Mg,Cu,Na,Si Teflon TLDs

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LiF:Mg,Cu,Na,Si Teflon TLD의 열처리 특성

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Abstract—The study of anneal characteristics is important for TL dosimeter to reuse. To obtain the annealing condition of the recently developed, new TL dosimeter, LiF:Mg,Cu,Na,Si Teflon TLDs in a disk type (diameter 4.5 mm, thickness about 90 mg/cm^2), we studied for pre-irradiation annealing, readout procedure and post-readout annealing, in order. The gamma irradiations were carried out with a ^{60}Co, dose of 0.1 Gy. We have used the method that observe the variation of thermoluminescent(TL) intensity of these Teflon TLDs over repeated cycles by changing both anneal temperature and anneal time with the TLD reader and the oven. There is a 5% loss in sensitivity over the ten repeated readouts by the annealing condition:pre-irradiation annealing at 80℃ for one hour, readout to 280℃ and post-readout annealing at 270℃ for 20 seconds.

Key words:annealing, reuse, TLD, thermoluminescence dosimeter, LiF:Mg,Cu,Na,Si Teflon TLDs

요약—열처리 특성의 연구는 열형광선량계를 재사용하는데 있어서 중요하다. 최근 개발된 디스크 형태(직경 4.5 mm, 두께 약 90 mg/cm^2)의 LiF:Mg,Cu,Na,Si Teflon TLD의 열처리 조건을 구하기 위하여 조사전 열처리, 판독과정 및 판독후 열처리의 순서로 연구하였다. Teflon TLD의 감마선 조사는 ^{60}Co 0.1 Gy로 하였다. LiF:Mg,Cu,Na,Si Teflon TLD의 열처리 특성의 연구는 전기로와 판독장치를 이용하여 열처리 온도와 열처리 시간을 변화시키면서, 측정반복횟수에 따른 열형광강도 변화를 관찰하는 방법으로 수행하였다. LiF:Mg,Cu,Na, Si Teflon TLD의 열처리 조건은 조사전 열처리를 80℃에서 1시간 한 후 280℃까지 판독하고 판독후 열처리를 270℃에서 20초간 하는 것으로 결정되었고, 이 조건에서 10회 반복측정시 원래의 열형광강도는 5%의 감소를 보였다.

중심어:열처리, 재사용, TLD, 열형광선량계, LiF:Mg,Cu,Na,Si Teflon TLDs

INTRODUCTION

LiF TLDs have been developed by adding several activators to a host material for a few decades. LiF:Mg,Ti(TLD-100) in the middle of TLD materials has been most well studied and used, but it has a complex glow curve and low thermoluminescent sensitivity[1]. In order to improve these drawbacks, LiF:Mg,Cu,P (GR-200) has been developed[2-4] and commercialized but its annealing[5, 6] is cumbersome.

Recently LiF:Mg,Cu,Na,Si phosphor with a high sensitivity and good fading characteristics was developed [7]. LiF:Mg,Cu,Na,Si Teflon TLDs in a disk form were fabricated with LiF:Mg,Cu,Na,Si phosphor for the practical radiation dosimetry[8].

In the present study, we have investigated the annealing characteristics of LiF:Mg,Cu,Na,Si Teflon TLDs, which has an importance to reuse for TL dosimeter. To obtain the annealing condition of LiF:Mg,Cu,Na,Si Teflon TLDs, we study for pre-irradiation annealing, readout procedure and post-readout annealing, in order. First, pre-irradiation annealing at 80℃ is performed in order to restore the original shape of the glow curve and the TL intensity[1]. Second, the influence of readout procedure on the TL intensity is investigated as a function of the maximum readout temperature. Finally, post-readout annealing is studied in order to erase the residual TL signals completely prior to reuse.

MATERIAL AND METHODS

LiF:Mg,Cu,Na,Si Teflon TLDs were fabricated from a mixture of LiF:Mg,Cu,Na,Si phosphor and Teflon powder as an adhesive material at the liquid nitrogen temperature. The mixture was moulded in a disk form (diameter 4.5mm, thickness about 90mg/cm^2) by pressing in room temperature and the color was a light blue[8].

Gamma irradiations of all Teflon TLDs were carried out using a ^{60}Co facility (installed in Baek Hospital, Pusan) with a dose of 0.1 Gy. All TL response of these Teflon TLDs were measured with TLD reader (Teledyne System 310) in a constant nitrogen flow. All measurements were made 24 hours after irradiation to eliminate the influence of low temperature TL glow peaks. The TL intensity is taken from the total glow curve area at a linear heating rate of 5℃ · S^{-1}. Data points of TL intensities are estimated by the average of four experimental data. Relative TL intensity represents the ratio of the TL intensity of annealed Teflon TLDs to that of the original Teflon TLD. For a batch, % standard deviation in TL sensitivities of Teflon TLDs is found about 4.5% [8].

The TL sensitivities per 10mg of TLD-100 powder (Harshaw), GR-200 powder (Harshaw), LiF:Mg,Cu,Na,Si phosphor powder and LiF:Mg,Cu,Na,Si Teflon TLDs were simultaneously compared to each of the phosphors. At this time, prior to use, TLD-100 was annealed at 400℃ for 1 h and 80℃ for 24 h, GR-200 was annealed at 240℃ for 10 minutes prior to use.

To obtain the annealing condition of LiF:Mg,Cu,Na,Si Teflon TLDs we have used the method that observe the variation of TL intensity of these Teflon TLDs over repeated cycles by changing both anneal temperature and anneal time with the TLD reader and an oven. Anneal cycles are performed as follows.

Pre-irradiation annealing:Anneal cycles of pre-irradiation annealing are performed as follows. (1) The virgin Teflon TLDs are irradiated by ^{60}Co dose of 0.1 Gy, then readout to 280℃, the measured TL intensity set to original value. (2) Pre-irradiation annealing at 80℃ is performed and then 1st annealed Teflon TLDs are reirradiated by ^{60}Co 0.1 Gy. In pre-irradiation annealing, 1st anneal cycle means "1st pre-irradiation annealing-readout".

Readout procedure:The irradiation and readout processes are as follows. (1) Pre-irradiation annealing at 80℃ is done prior to irradiation. (2) The TLDs are irradiated with a ^{60}Co dose of 0.1 Gy. (3) The TLDs are readout to the maximum readout tempera-

tures of 280℃, 290℃ and 300℃ respectively. (4) The above processes are repeated at the same condition until the 9th readout.

Post-readout annealing:The used TLD reader has the restriction of post-readout anneal time (that is 1 minute). To do post-readout annealing for a long time than 1 minute, we may use an oven. Pre-irradiation annealing at 80℃ for one hour is done prior to irradiation. The Teflon TLDs are irradiated with ^{60}Co dose of 0.1 Gy. These Teflon TLDs are readout to 280℃, then successively annealed between 250℃ and 300℃ with 10℃ interval by the TLD reader, and consecutively repeated 9 times readout-post-readout annealing without irradiation. The above readout constitutes the dosimetric signal, and the sum of nine subsequent readouts determines the total residual signal. Relative TL intensity and total residual signal represent ratios relative to the original unannealed TLD.

In order to show the variation occurring for a long anneal time, Teflon TLDs are treated each for 5 minutes with 10℃ interval in the range between 250℃ and 360℃, in an oven. Anneal cycles of post-readout annealing are carried out the procedure:pre-irradiation annealing(80℃ 1h), readout procedure (up to 280℃), and post-readout annealing for 5 minutes.

RESULTS AND DISCUSSIONS

TL Glow Curve and Sensitivity

Fig. 1 shows the typical TL glow curves of (a) TLD-100 powder (Harshaw), (b) GR-200 powder (Harshaw), (c) LiF:Mg,Cu,Na,Si phosphor powder and (d) LiF:Mg,Cu,Na,Si Teflon TLDs. In the shapes of glow curve, we can show that both side peaks of main peak of LiF:Mg,Cu,Na,Si Teflon TLD have lower than those of GR-200. From this fact, we can see that Teflon TLDs have advantage to separate the main glow peak (dosimetric peak) from the other glow peaks, it is more easy and more accurate to estimate the dose by choosing the main peak area. The relative TL sensitivities (total area and peak height) and the main peak temperatures of the TLDs are compared in table 1. It is shown that the TL sensitivity of LiF:Mg,Cu,Na,Si Teflon TLDs is higher (11 and 15 times in area and peak height, respectively) than that of TLD-100 powder, and higher (1.1 and 1.4 times in area and peak height, respectively) than that of GR-200 powder, but lower than that of LiF:Mg,Cu,Na,Si phosphor. And it is shown that the main peak temperature of the Teflon TLD occurs at more higher temperature than that of the other TLDs.

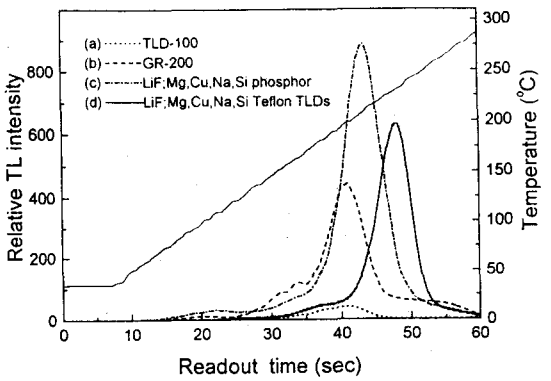

Fig. 1. Typical TL glow curves of TLDs for constant heating rate of 5℃ s^{-1}. (a) TLD-100 (LiF:Mg, Ti ; Harshaw), (b) GR-200 (LiF:Mg,Cu,P ; Harshaw),), (c) LiF:Mg,Cu,Na,Si powder (d) LiF:Mg,Cu,Na,Si Teflon TLD.

Table 1. Relative TL sensitivities of TLDs.

TLDs	Main peak temperature	TL Sensitivity	
		Peak height	Total area
TLD-100 (LiF:Mg,Ti)	211℃	1	1
GR-200 (LiF:Mg,Cu,P)	213℃	11	10
LiF:Mg,Cu,Na,Si powder	222℃	21	17
LiF:Mg,Cu,Na,Si Teflon TLDs	236℃	15	11

Pre-irradiation Annealing at 80℃

Fig. 2 shows the shapes of glow curves of Teflon TLDs as a function of anneal times of 0, 0.5, 1, 2, 3 hours after pre-irradiation annealing at 80℃, these are compared with that of the unannealed Teflon TLD. In pre-irradiation annealing, the TL intensities of peaks 1 and 2 was decreased with increasing anneal time. The relative ratios of peak heights of annealed Teflon TLDs are compared with the original Teflon TLD in table 2. In pre-irradiation annealing at 80℃, we can see that the original glow curve shape is well maintained for one hour as anneal time.

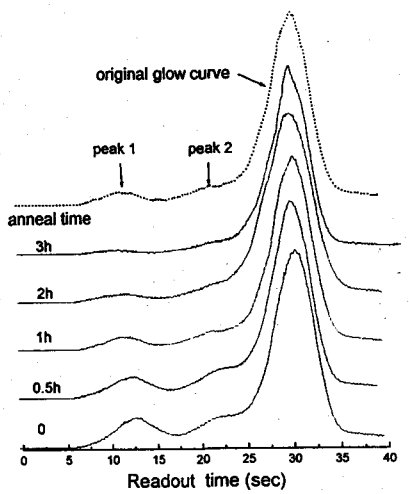

Fig. 2. Comparison of glow curves of LiF:Mg,Cu,Na, Si Teflon TLD used after pre-irradiation annealing at 80℃ for anneal times of 0(without anneal), 0.5, 1, 2 and 3 hours with the original glow curve.

Table 2. Relative ratios of the peak heights with pre-irradiation annealing at 80℃.

Anneal time	Peak 1	Peak 2
Original	1	1
0	2.23	1.62
0.5h	1.67	1.46
1h	1	1.02
2h	0.67	0.98
3h	0.27	0.78

Fig. 3 shows the relative TL intensities as a function of anneal time in pre-irradiation annealing at 80℃, according to anneal-readout cycles (a) 1st, (b) 2nd and (c) 3rd. From Fig. 3 (a), it is shown that the relative TL intensity of one hour annealing is highest. This tendency clearly appears with the increasing readout cycles. In Fig. 3 (c), TL intensity for one hour is 4% higher than that of the unannealed. The best anneal time in pre-irradiation annealing at 80℃ was determined to one hour that the original TL intensity restores well over anneal cycles.

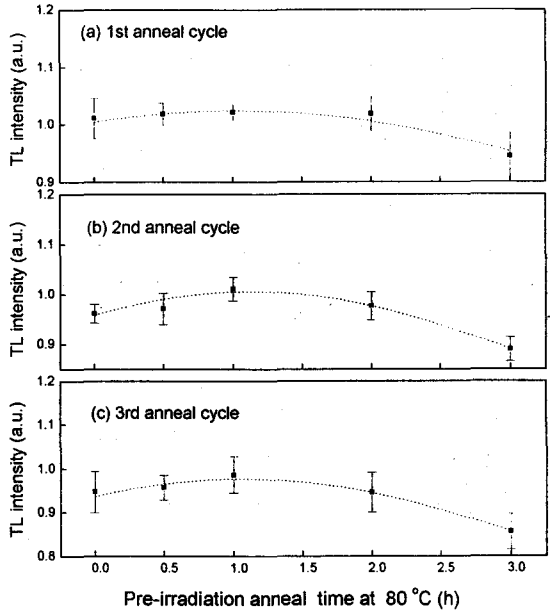

Fig. 3. Relative TL intensity changes due to pre-irradiation anneal time for the anneal cycles 1, 2 and 3 respectively.

Readout Procedure

Fig. 4 shows influence of readout procedure as a function of repeated readout cycles at the maximum readout temperatures of 280℃, 290℃ and 300℃. The shape of the glow curve is found to be varied a little over the readout cycles. The loss of TL sensitivities of Teflon TLDs after 9th readout cycle is 11%, 25% and 28% for the maximum readout temperatures,

280℃, 290℃ and 300℃, respectively. we can see that readout procedure of readout to 280℃ after the pre-irradiation annealing (80℃, 1h) has 11% loss in sensitivity over the 9th readout cycle. From the results, the maximum readout temperature is determined to 280℃.

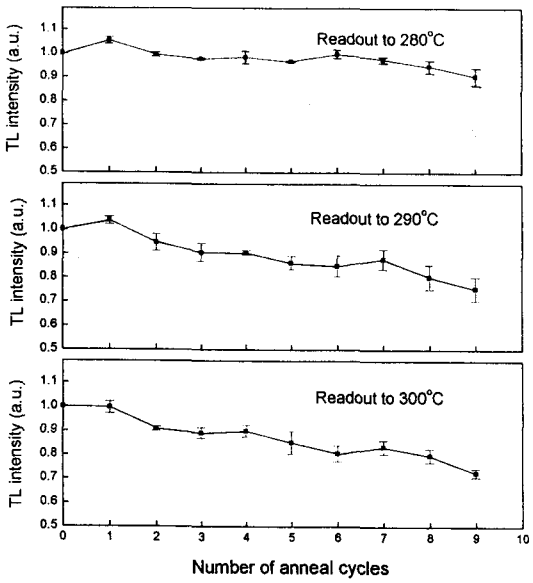

Fig. 4. Influence of maximum readout temperatures of 280℃, 290℃ and 300℃ at linear heating rate 5℃ s^{-1}.

Post-readout Annealing

To determine the anneal time of post-readout annealing, we change the anneal time in the TLD reader. Fig. 5 shows the relative TL intensities as a function of anneal times from 0 to 50 seconds in the post-readout annealing. The TL intensity at 20 second is highest, 5% higher than that of zero second annealing.

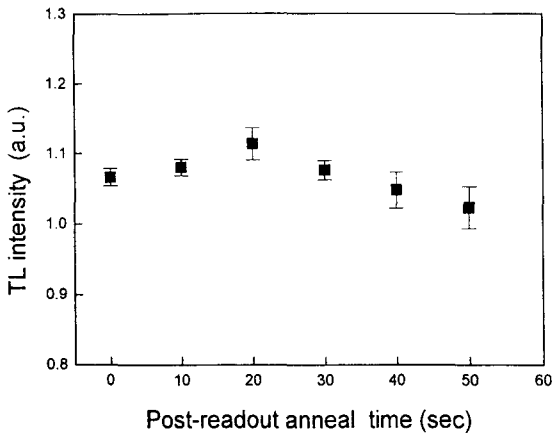

Fig. 5. Relative TL intensity as a function of post-readout anneal times 0, 10, 20, 30, 40 and 50 second.

The anneal time of 20 second is selected in the post-readout annealing by the TLD reader. Fig. 6 shows the relative TL intensity and the total residual signal of the post-readout annealing in reader as a function of the anneal temperatures from 250℃ to 300℃. The relative TL intensity of 11th readout is decreased by 7%, 5%, and 12% in post-readout annealing in reader for 20 seconds at the annealing temperatures 260℃, 270℃, and 280℃, respectively (in Fig. 6). It can be seen that the loss of the relative TL intensity is 5% over the ten readouts with the anneal condition that pre-irradiation annealing at 80℃ for one hour, readout to 280℃ and post-readout annealing in reader at 270℃ for 20 seconds. In this anneal condition, the total residual signal is slightly decreased with the anneal temperature (1.66% at 250℃ and 0.83% at 300℃).

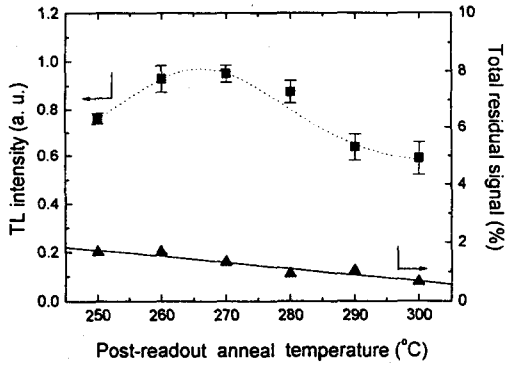

Fig. 6. Relative TL intensity and total residual dose as a function of the post-readout anneal temperatures in reader from 250℃ to 300℃ for 20 seconds.

Fig. 7 shows the variation of the relative TL intensities after 1st, 2nd anneal cycles as a function of the oven anneal temperature between 250℃ and 360℃ for 5 minutes in the post-readout annealing. We can see that the loss of relative TL intensity has a little bit in the range from between 260℃ and 270℃, and rapidly increased with 84% at 360℃ in the post-readout annealing. Comparing Fig. 6 and Fig. 7, the tendencies of TL intensities in both are similar but the relative TL intensity in Fig. 8 is lower than that of Fig. 7 between 280℃ and 300℃. Because post-readout annealing for 5 minutes in an oven does not contribute to restore the TL sensitivity, it is not selected.

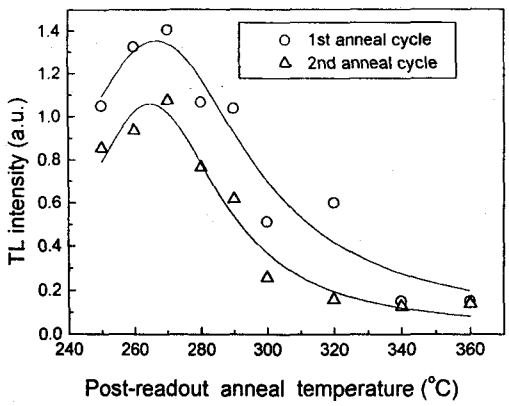

Fig. 7. Relative TL intensity for the post-readout annealing in oven as a function of anneal temperatures between 250℃ and 360℃ for 5 minutes.

CONCLUSION

The relative TL sensitivity of LiF:Mg,Cu,Na,Si Teflon TLDs is more higher than that of TLD-100 powder, and GR-200 powder, but lower than that of LiF: Mg,Cu,Na,Si phosphor. And the main peak temperature of the Teflon TLD occurs at more higher temperature than that of the other TLDs.

In pre-irradiation annealing, the TL sensitivity and the shape of the original glow curve are well restored at 80℃ for one hour. The relative TL intensity pre-irradiation annealing (at 80℃ for one hour) is 4% higher than that of unannealed Teflon TLD.

In readout procedure without post-readout annealing, we can see that readout procedure of readout to 280℃ after the pre-irradiation annealing (80℃, 1h) has 11% loss in sensitivity over the 9th readout cycles. When the post-readout annealing is performed, there is 5% loss in sensitivity over the ten readouts with the anneal condition that pre-irradiation annealing at 80℃ for one hour, readout to 280℃ and post-readout annealing in reader at 270℃ for 20 seconds. In this anneal condition, the total residual signal is slightly decreased with the anneal temperature (1.66 % at 250℃ and 0.83% at 300℃). Post-readout annealing for 5 minutes does not contribute to restore the TL sensitivity, it is not selected for future use.

From these results, we could determine the annealing condition:pre-irradiation annealing at 80℃ for one hour, readout to 280℃ and post-readout annealing at 270℃ for 20 seconds. that pre-irradiation annealing at 80℃ for one hour, readout 280℃ and post-readout annealing in reader at 270℃ for 20 seconds, there is a 5% loss in sensitivity over the ten repeated readouts.

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