Quantitative Evaluation of LiF: Mg, Ti (TLD-100) Long-Term Sensitivity Stability

Mianji F.[1,2]*, Baradaran S.[1,2]

[1]Nuclear Safety and Radiological Protection Research Department, Nuclear Science and Technology Research Institute, End of North Kargar, Tehran, Iran
[2]Radiation Protection Department, Iran Nuclear Regulatory Authority, End of North Kargar, Tehran, Iran

*Corresponding author: F. Mianji, Ph.D. of Medical Radiation Engineering, Assistant Professor of NSTRI. E-mail: fmianji@aeoi.org.ir

ABSTRACT

Background: Sensitivity is an important dosimetric issue particularly in personal dosimetry where the range of doses is often very low to medium. In-service aging is assumed to be of negligible effect on the response of thermoluminescent dosimeters (TLD), consequently, some service providers may mix fresh TLDs to in-use ones and simply normalize the readings of TLDs through batch calibration. This may lead to an increase in the coefficient of variation of the batch.

Objective: The correlation between the sensitivity of LiF(Mg:Ti) and its in-service life is quantitatively studied in this paper.

Method: Three groups of dosimeters with different service lives were chosen from the inventory of the national TLD service provider. The TLDs were then exposed to different doses and their TL responses were measured and compared.

Result: By analyzing the results obtained from groups with different in-service ages, it was shown that the sensitivity of the dosimeters degrades with age in long term, and the amount of this sensitivity loss was measured. The reading standard deviation of the dosimeters degrades with the service life for low doses but seems to be insensitive to the service life in medium and high doses.

Conclusion: A more efficient annealing approach is recommended time to time for TLDs. Moreover, the calibration dosimeters must be updated whenever a big difference between the sensitivity of new and used TLDs is seen.

Keywords

Aging, LiF(Mg:Ti), Personal Dosimetry, Sensitivity, TLD-100

Introduction

Thermoluminescent dosimeters (TLD) are the most commonly used devices for passive personal and environmental monitoring purposes [1]. As an important part of dosimetry systems in nuclear facilities and radiation practices, their reliability has been proven in many aspects including a very low detectable dose (about 0.05 mGy), a wide recordable dose range (up to 10 Gy), good energy independency, etc.

One of the most important characteristics of a reusable passive dosimeter is its aging behavior, i.e., stability of sensitivity over time. For TLDs, according to ISO 21909, the performance and the test requirement for passive personal neutron dosimeters, aging of TLD is not a matter of concern [1, 2]; however, according to RS-G-1.3, one of the criteria to be assessed as part of the type testing of TLD is the stability of its response over time [3].

The most widely used material in TLDs is LiF with added magnesium to increase the number of traps in the lattice, and titanium to increase the number of luminescence centers. LiF(Mg,Ti) with the average atomic number of 8.2 consists of 92.5% of ^{7}Li and 7.5% of ^{6}Li and is known as TLD-100. Owing to its similar atomic number to the human tissue (7.4), it is widely used for personnel monitoring. TLD-100 is available in many forms such as pellets, single crystals, powders, rods and even gel. It is often used in the form of LiF crystal doped with magnesium and titanium. The common production process is melting lithium fluoride, lithium cryolite, magnesium fluoride and lithium titanium fluoride together. The emission peak of TLD-100 is at 400nm which matches the response of the photomultiplier tube's (PMT) photocathode [4]. Experimental study of intrinsic self-dosing effects of TLD-100 has shown very small self-dosing effects in the material [5]. It is also shown that fading is not a significant problem with TLD-100. Its reasonable resistance to storage temperature after irradiation with both gamma-rays and thermal neutrons is reported too [6, 7].

When a center, e.g. a hospital, uses TLD dosimeters like TLD-100 and an appropriate TLD reader for various applications of radiation dosimetry, the possible drift of the dosimeters from their primary sensitivity after a certain number of exposures or period of time can be compensated by recalibration. In this case owing to the fact that the batch of dosimeters is not very big and, that they all have almost the same working age, the standard deviation around the mean (coefficient of variation) is not very big and a recalibration suffices. This reasonably prevents under/overestimation of the doses in medical or occupational terms; however, for large facilities and service providers which time to time add new TLDs to their inventory such presumed stability cannot be assured beforehand. A considerable decrease in sensitivity of the TLD means that its minimum detectable dose is also decreased. Thus, mixing aged TLDs with fresh dosimeters may not be probably a sound idea. A wider sensitivity range of the batch causes a big coefficient of variation which is not statistically desirable. In this work, several experiments were carried out on long-term used, mean-term used and fresh TLDs to investigate the effects of service life on the performance of dosimeters. The results proved that the service life has a considerable effect on the performance of dosimeters even if they are not exposed to high doses.

The rest of the manuscript is organized as follows. Section Two presents the materials and methods including the experimental setup for this work. Section Three presents the results and discusses the findings. Conclusion is given in Section Four.

Materials and Methods

The main property of TLD that may reduce the sensitivity after frequent uses is its characteristic regarding to the electron-hole trapping centers. For instance, the glow curve of TLD-100 shows at least 13 peaks. This is due to the numerous traps of varying depth (or temperature) in the structure of TLD-100. Each trap represents a potentially different energy observable in the glow curve; however, the major peak commonly used in routine photon dosimetry reaches its maximum at about 210^{o} C [8]. Nevertheless, the trapping centers of TLD-100 are stable; availability, distribution and frequency of them in routine use are affected by various factors. Moreover, some of them are not thermally independent. This fact may cause simultaneous depletion in an energy trap and enhancement in another trap due to transition among traps. Therefore, the annealing history of the TLD is important. Other factors affecting the sensitivity include the dose level, physical damage and in-service age. The crystalline structure of the TLD exposed to very high doses (>100 Gy) may get damaged. Regarding the in-service age, there is no agreement on the recommended service life of TLDs. Limiting it to 50 using cycles has been suggested by some users; however seemingly, this strongly depends on the required level of dose uncertainty, the dose levels being measured (application type), the adequacy of the annealing process, the physical damages to the TLDs (can usually be minimized by careful handling, storage and cleaning), etc.

Even fresh TLDs made by a manufacturer, due to minor structural and fabricating variations, may have different responses, i.e., TL efficiency (hereinafter, TL refers to integrated TL glow curve), when they are irradiated to the same dose from the same source and are read out by the same reader under the same conditions. This is the case even for TLDs of the same production batch. The TL efficiency is expressed as the thermoluminescent light emitted per unit of absorbed dose. There is always a sensitivity standard deviation among TLDs of a typical batch which is normally expected not to exceed 10-15%. Such a standard deviation can, however, be greatly reduced, say to 1-2% by batch calibration. To narrow the range, which is mainly an intrinsic property of TLD rather than the errors caused by the reader, Element Correction Coefficient (ECC) is defined for any TLDs of the batch through batch calibration process. ECC relates the TL efficiency of a specific dosimeter to the average TL efficiency (TLE) of the Calibration Dosimeters (CD). To designate CDs, firstly, all TLDs of the batch are irradiated to the same uniform dose at the same geometrical conditions. Then, TLs are read out and the corresponding TLDs whose TLs lay in a reasonable distance from the average TL value, e.g. 0.95 to 1.05 of the average TL, are chosen as CDs. The average value of all CDs, i.e., average TLE, is calculated and compared with the efficiency of each one of dosimeters called Field Dosimeters (FD) to calculate the ECC for each FD.

ECC of TLD is given by:

$$ECC_i = TLE_{av} / TLE_i \quad (1)$$

where ECC_i is the ECC of the i_{th} FD of the batch and TLE_{av} is the average TL of the CDs.

When new dosimeters are added, the ECCs are re-evaluated with the aim of normalizing their efficiency with the existing ones. In order to achieve this, the sensitivity of the calibration dosimeters must remain constant [7]. The results of this research show that adding fresh TLDs to the used batch, particularly without updating CDs, will considerably affect the effectiveness of ECC in minimizing the coefficient of variation of the TLD responses. It is worth mentioning that updating CDs is not a trivial task for service providers who may possess thousands or tens of thousands TLD cards.

The aim of this research is to quantitatively assess the effects of in-service age on the sensitivity of TLD-100. To this end, TLDs of various in-service ages, but with similar annealing and exposure history and physical conditions, were chosen and irradiated to several doses. Then, their integrated TL glow curve signals were read out by the same reader and compared to each other.

Sample Selection

Iran Nuclear Regulatory Authority (INRA) had been the main TLD service provider (with a bimonthly period) for almost all the facilities and radiation practices from 2000 to 2012 in Iran. All used TLD cards were thus available for this research by INRA. Moreover, all used TLD cards have been from the same type and brand purchased at different times. They are Harshaw two-chip TLD-100, i.e., each card has two small TLD-100 pellets packed in an alominium frame and shielded by a wear and heat resistive plastic sheet. Therefore, each card has two identical TLDs which will be referred to by indices 1 and 2 hereinafter. Three groups of TLDs were selected for these experiments. Group one included 8 TLD cards (16 TLDs), randomly chosen from the TLDs that had been in service for 10-12 years. This group is referred to as G10. The next group (8 cards) was picked out from the TLDs that were in service for 5-6 years (G5). The last group was from the TLDs which have never been used before (fresh ones: G0). Attention was paid to ensure that the used TLDs had been under continuous service and to ensure all the TLDs were physically damage-free. None of used TLD cards had a record of extreme high dose.

Annealing Samples

Long period of service life and different annealing cycles may result in changes in dosimeter sensitivity. An ideal annealing for LiF(Mg,Ti) is 400° C for one hour followed by a longer annealing at a lower temperature, e.g. two hours at 100° C. This is obviously a time consuming and costly process, particularly when many dosimeters are supposed to be annealed. Furthermore, such a high temperature annealing is not practically applicable for TLD cards constructed of aluminum and plastic. Therefore, many service providers adopt TLD reader cycle to this end. This annealing approach is neither perfect nor guaranteeing preserving the sensitivity of TLDs in long term; nonetheless, consistency in the employed annealing method is more important. Other important factors are the duration of cooling period (slower cooling down results in less thermal defects in crystalline material and less trapping centers) and the annealing temperature in air (elevated temperature causes oxidative damage at the surface of TLDs). All these may gradually reduce the sensitivity particularly for the measurement of low penetrating radiations.

For annealing of the selected samples, the same procedure was routinely applied during their service life. These TLDs were read out three times at the temperature of 250° C to become depleted of the residual energy (before every new exposure).

Exposing Samples

For every experiment, after annealing TLD cards of each group, they were put in a flat and thin holder very close to each other. All groups were exposed by a standard Cs-137 source traceable to the national standard institute to different dose levels: 1, 25, 100, 500 mGy at a distance of 50 centimeters to eliminate the effect of angle. The cards were then immediately transferred to the reading room for preheating and reading.

Reading Samples

The glow curve of TLD-100 has 6 important peaks at different temperatures [9]. The main peak used for the dose measurement is the 5^{th} peak which reaches its maximum at around 210° C. Reading TLDs was carried out through the same reader under the same conditions. Harshaw reader model 6600 was used as the reader. TLD cards were first preheated to 100° C for 20 minutes for relaxation from the background peaks (peaks No. 1 to 4) in the oven. After letting them cool down to the room temperature, they were read out (the integrated TL glow curve) at the temperature of 250° C. The two TLDs of every card was read simultaneously by two separate PMTs of the reader, the efficiency of PMTs must be thus normalized using their reading of the system reference light. TL of chip 2 of each card was corrected to chip 1 of the same card using the related PMTs responses to the reader's reference light. The typical glow curve (after preheat) of the used reader and the cards are shown in Figure 1.

The within group standard deviations were then calculated and compared with other groups and also with the overall standard deviation (for all groups together). Moreover, the average sensitivity levels of the groups were compared to define the relationship between the sensitivity of TLD-100 and its in-service age in quantitative terms.

Results and Discussion

The results for the three groups exposed to the dose of 1 mGy are presented in Table 1.

For 1 mGy dose, the mean TL for G10 and G5 are less than G0. G10 shows a higher mean TL than G5, though its std/mean is about 83% which is very unreasonable. The high mean sensitivity of G10 is mainly owing to the two

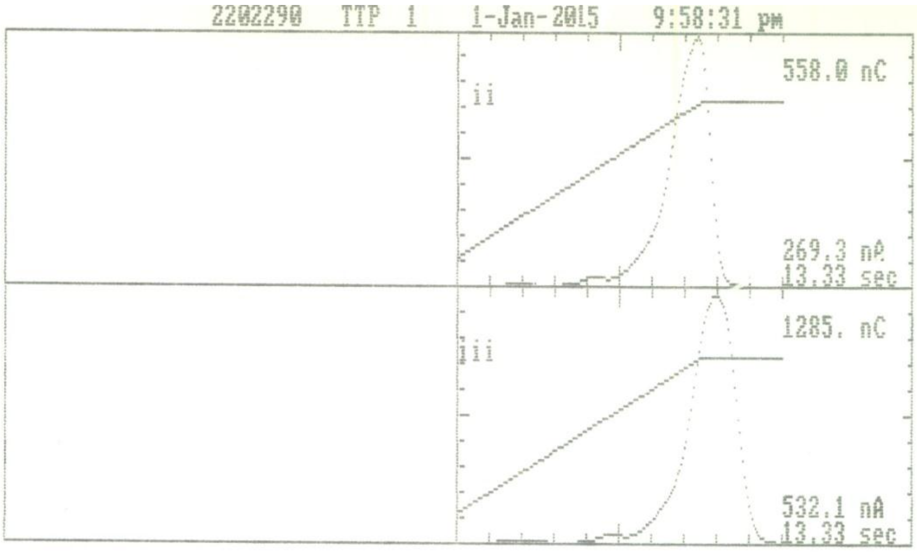

Figure 1: Glow Curves of Used TLDs after Pre-heating

Table 1: Integrated TL Glow Curve of TLD Cards G10, G5 and G0 after Receiving 1 mGy Dose

G10								
Card ID	1	2	3	4	5	6	7	8
TLD 1	2.41	2.96	20.03	3.33	14.68	2.7	3.20	4.62
TLD 2	3.73	3.40	8.38	7.71	13.08	6.15	4.81	4.72
Mean TL	6							
Std	5							
G5								
Card ID	1	2	3	4	5	6	7	8
TLD 1	2.14	2.21	3.55	1.86	2.71	1.89	2.73	1.85
TLD 2	2.22	2.58	3.09	1.56	2.42	1.81	4.69	1.84
Mean TL	2.4							
Std	0.8							
G0								
Card ID	1	2	3	4	5	6	7	8
TLD 1	10.11	5.21	6.67	5.10	7.87	3.25	3.65	9.02
TLD 2	13.7	4.50	4.80	4.00	5.00	7.50	6.10	11.90
Mean TL	6.8							
Std	3.1							

out of range readings (cards No. 3 and 5). The std/mean for G5 and G0 are 33% and 45% which are much better than the rate for G10. Overall, for this low dose level, G0 has the highest mean sensitivity with an acceptable std, G5 has the lowest mean sensitivity but with the best std, and G10 shows a high mean sensitivity with the poorest std. The std of all the TLDs together (including all the three groups) is 3.85.

The results for dose of 25 mGy are presented in Table 2. For the medium dose level of 25 mGy, the fresher is the group of TLDs, the higher is the mean sensitivity. G0 shows 23%

Table 2: Integrated TL Glow Curve of TLD Cards G10, G5 and G0 after Receiving 25 mGy Dose

G10								
Card ID	1	2	3	4	5	6	7	8
TLD 1	202.8	216.1	212.5	191.5	217.5	177.8	181.7	193
TLD 2	206.5	194.4	245.6	213.5	209.6	192.8	215.1	178.4
Mean TL	203.3							
Std	17.6							
G5								
Card ID	1	2	3	4	5	6	7	8
TLD 1	223.3	233.8	231.8	206.9	225	241.4	228.9	269.5
TLD 2	209.7	258.6	223.4	211.2	262.4	227.1	295	215.8
Mean TL	235.2							
Std	24.5							
G0								
Card ID	1	2	3	4	5	6	7	8
TLD 1	297.9	300.9	251.2	261.5	306.8	281.8	298.1	261.7
TLD 2	286	329.2	320.	336.8	299.7	272.8	291.5	256.5
Mean TL	290.8							
Std	25.6							

and 43% higher sensitivity than G5 and G10. In terms of std, all groups are comparable with std/mean values equal to 8.6%, 10.4%, and 8.6% for G10, G5, and G0, respectively. The std of all TLDs together (including all three groups) is 42.8 which is about 2.5 times greater than G10 and near 1.8 times G5 and G0.

The results for dose of 100 mGy are presented in Table 3. For the relatively high dose level of 100 mGy, the relationship between the responses of TLD groups is similar to the dose of 25 mGy. G0 shows 30% and 45% higher sensitivity than G5 and G10. In terms of std, all groups are comparable in this dose level too. TL std/mean ratio is 10.7%, 10.9%, and 8.8% for G10, G5, and G0, respectively. The std of all TLDs together (including all three groups) is 201.6 which is about 2.9 times G10 and near 1.9 times G5 and G0.

The results of 500 mGy are presented in Table 4. The situation in the high dose level of 500 mGy is similar to doses of 25 and 100 mGy. The mean TL for G0 is 25% and 47% higher than G5 and G10. TL std/mean ratios are respectively 9.5%, 11% and 9.6% for G10, G5 and G0, which are comparable again. The std of all TLDs together (including all three groups) is 899.3 which is about 2.5 times G10 and near 1.7 times G5 and G0.

Figure 2 shows the trend of mean TL response of G0, G5 and G10 versus dose level.

Conclusion

Dependency of the sensitivity of TLD100, the most commonly used passive dosimeter for personal dosimetry, to the in-service age is studied in this paper. Harshaw TLD100 fresh cards and used cards with continuous in-service life of about 10 and 5 years (used for personal dosimetry purpose with a bimonthly period) were chosen and exposed to doses of 1, 25, 100 and 500 mGy. The reading results showed that fresh TLDs are about 50% more sensitive than the TLDs that have been in service for about 10 years. Fresh TLDs are 25 to 30% more sensitive than mid-aged TLDs too (about 5 year in-service life). According to these findings, when new TLDs are to be

Table 3: Integrated TL Glow Curve of TLD Cards G10, G5 and G0 after Receiving 100 mGy Dose

G10								
Card ID	1	2	3	4	5	6	7	8
TLD 1	962	936.3	856.6	849.2	888.6	761.6	812.9	883.4
TLD 2	981	916.1	1031.9	973.6	907.9	838	988.4	883.2
Mean TL	904.4							
Std	72.2							
G5								
Card ID	1	2	3	4	5	6	7	8
TLD 1	934.6	974.3	967.1	855.9	987.3	1081	982.9	1145
TLD 2	909.5	1086.2	961.3	922.7	1144.5	1031.9	1285.8	919.4
Mean TL	1011.8							
Std	111.3							
G0								
Card ID	1	2	3	4	5	6	7	8
TLD 1	1311	1387	1173	1170	1301	1273	1296	1208
TLD 2	1221.7	1533.1	1504.3	1485.5	1349.9	1298.9	1335.9	1177.4
Mean TL	1314.1							
Std	116.1							

Table 4: Integrated TL Glow Curve of TLD Cards G10, G5 and G0 after Receiving 500 mGy Dose

G10								
Card ID	1	2	3	4	5	6	7	8
TLD 1	3924	3803	3882	3724	3910	3209	3621	3816
TLD 2	4143.5	3797.6	4789.1	4326	3896.4	3677.9	4484.3	3685.4
Mean TL	3918.1							
Std	372.1							
G5								
Card ID	1	2	3	4	5	6	7	8
TLD 1	4175	4525	4396	3884	4254	4585	4396	5166
TLD 2	4181.1	4939.8	4390.5	4206.3	5285.7	4810.9	5829.1	4307.6
Mean TL	4583.2							
Std	504.6							
G0								
Card ID	1	2	3	4	5	6	7	8
TLD 1	5603	6049	5194	4978	5680	5560	5553	5168
TLD 2	5606.4	6862.5	6610.4	6633	5952.2	5614.8	5744.6	5203.6
Mean TL	5750.8							
Std	552							

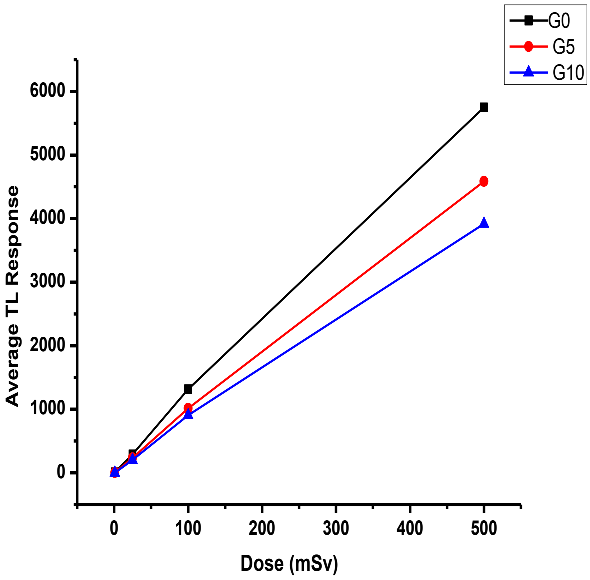

Figure 2: Mean Sensitivity vs. Dose for three TLD Groups with Different In-service Lives

added to the group of TLDs which have been in use by a personal dosimetry service provider, a preliminary evaluation to compare the sensitivity level of these two groups are recommended. Particularly, for low doses (less than few mSv) that are often the values of real occupational doses, the std of long- term used TLDs are unacceptable. This is likely due to the residual energy of TLDs that cannot be released through the commonly used annealing method of applying multiple TLD reader cycles. Therefore, a more efficient annealing approach is recommended time to time for personal dosimetry by TLD.

A sensitivity comparison of the new and in-service dosimeters will ensure that the variance of the whole group (the used and the fresh dosimeters together) will not exceed a reasonable level. In case a big difference between the new and used TLDs is seen, the calibration factors must be updated. The results showed that the coefficients of variation, i.e., the ratios of the standard deviations of TLD groups to their mean TL values, were comparable for the fresh, mid-aged and long-aged groups.

Acknowledgment

The authors would like to thank Mr. Bardia Hajizadeh and Mr. Arash Enferadi for their technical co-operations.

Conflict of Interest

None

References

1. HosseiniAliabadi SJ, Hosseini Pooya SM, Afarideh H, Mianji F. Improvement of Accuracy in Environmental Dosimetry by TLD Cards Using Three-dimensional Calibration Method. *J Biomed Phys Eng*. 2015;**5**:49-52. PubMed PMID: 26157729. PubMed PMCID: 4479385.
2. In: ISO: International Organisation for Standardisation. Passive personal neutron dosimeters – Performance and test requirements. 2005. Available from: https://www.iso.org/obp/ui/#iso:std:iso:21909:ed-1:v1:en.
3. Guide IS. Assessment of occupational exposure due to external sources of radiation. IAEA Safety Guides, No. RS-G-1.3, 18, 1999.
4. Freire L, Calado A, Cardoso J, Santos L, Alves J. Comparison of LiF (TLD-100 and TLD-100H) detectors for extremity monitoring. *Radiation Measurements*. 2008;**43**:646-50. doi.org/10.1016/j.radmeas.2007.12.013.
5. Moor D, Horspool B, Stokes R. Performance of the Harshaw DXT-RAD (TLD-100) dosemeter. *Radiation Measurements*. 2008;**43**:533-7. doi.org/10.1016/j.radmeas.2007.11.035.
6. Delgado A, Vergara JS, Ros JG, Romero A. Intrinsic Self-Dosing and Long Term Stability of LiF TLD-100 and GR-200 TL Detectors. *Radiation protection dosimetry*. 1995;**58**:211-6.
7. Bilski P, Berger T, Hajek M, Twardak A, Koerner C, Reitz G. Thermoluminescence fading studies: Implications for long-duration space measurements in Low Earth Orbit. *Radiation Measurements*. 2013;**56**:303-6. doi.org/10.1016/j.radmeas.2013.01.045.
8. Gilvin PJ, Burkett RA, Baker ST, Garratt NJ. Long-term stability of Harshaw LiF:Mg,Cu,P TLDS. *Radiat Prot Dosimetry*. 2011;**144**:192-4. doi.org/10.1093/rpd/ncq559. PubMed PMID: 21216732.
9. Luo LZ, Velbeck KJ, Rotunda JE. Evaluating two extremity dosemeters based on LiF:Mg,Ti or LiF:Mg,Cu,P. *Radiat Prot Dosimetry*. 2002;**101**:211-6. doi.org/10.1093/oxfordjournals.rpd.a005969. PubMed PMID: 12382737.
10. Savva A. Personnel TLD monitors, their calibration and response: University of Surrey; 2010.