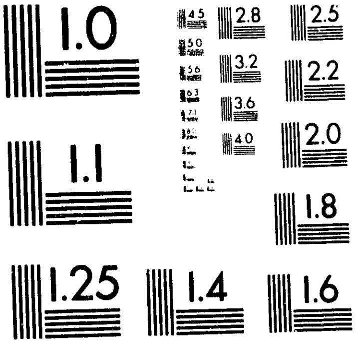
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REUSABILITY OF CaF_2:Mn TLDS FOR PHOTON IRRADIATIONS AT HIGH ABSORBED-DOSE LEVELS[2]

REFERENCE: Vehar, D. W., "**Reusability of CaF_2:Mn TLDs for Photon Irradiations at High Absorbed-Dose Levels,**" Reactor Dosimetry, ASTM STP 1228, Harry Farrar IV, E. Parvin Lippincott, and John G. Williams, Eds., American Society for Testing and Materials, Philadelphia, 1994.

ABSTRACT: The effects of high absorbed dose on the reusability of CaF_2:Mn thermoluminescence dosimeters (TLDs) are investigated to determine a recommended upper limit on absorbed dose for TLDs that are to be reused. This investigation examines degradation in the uniformity of response and changes in sensitivity of a batch of TLDs when exposed to gamma-radiation doses ranging from 1 Gy to 1000 Gy, and confirms earlier work suggesting that CaF_2:Mn TLDs should not be reused in applications where cumulative absorbed doses are likely to exceed 100 Gy.

KEYWORDS: thermoluminescence dosimeter, thermoluminescent dosimeter, CaF_2:Mn, TLD, reusability, gamma radiation, photon irradiations, high absorbed dose.

Introduction

The Radiation Metrology Laboratory (RML) has previously investigated the effects of high radiation dose on CaF_2:Mn thermoluminescence dosimeters (TLDs) to determine conditions under which the dosimeters could be reliably reused. This earlier study involved a sample of several tens of thousands of TLDs and demonstrated a broadening in batch standard deviation, as well as changes in sensitivity, for TLDs that had been exposed to cumulative absorbed doses in excess of 200 Gy (20 krad). Based on this study, a decision was made by the RML not to reuse CaF_2:Mn TLDs in applications where cumulative absorbed doses were likely to exceed 100 Gy. The difference between the 100 Gy recommended limit and the 200 Gy level at which these effects had been noted was intended to provide a margin of safety, because the precise level to which a TLD will be exposed during a test is not usually known before the test is conducted.

There were several reasons for repeating the earlier study. Because the study was informal and intended only for internal use, it was never formally documented. Second, although the experiment was fundamentally sound, the TLDs that formed the basis for this test came from those actually used for irradiations of other experiments. TLDs were separated into groups according to absorbed dose received, resulting in relatively wide groupings. Thus, correlation of broadening of batch standard deviation with cumulative absorbed dose could only be done in general terms. Finally, the TLDs used in this test were obtained from a variety of experiments at several facilities. These facilities included fast-pulse and TRIGA-type nuclear reactors as well

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as high-energy bremsstrahlung and ^{60}Co irradiators. It was therefore possible that some of the observed broadening might have been caused by other factors, such as exposure of the TLDs to neutron environments.

The purpose of the present experiment was to examine the broadening of batch standard deviation with absorbed dose under much more rigidly controlled conditions so that limitations in reusability of CaF_2:Mn TLDs could be more precisely defined. Irradiations were performed at specific, well-defined levels of absorbed dose, and exposures were restricted to a single, gamma-ray-only irradiation facility. Irradiations were done at several absorbed-dose levels from 1 to 1000 Gy, spanning the 200 Gy level at which broadening effects had previously been noted.

Initial Irradiation and Sorting

A total of 18 200 TLDs was divided into 13 groups of 1400 TLDs each. Each group of 1400 TLDs was placed in a 25 by 56 TLD array in a rectangular fixture that provided 2.2 mm (0.090 in) thickness of aluminum to the front and back of the TLDs for electron equilibration. Each group of 1400 TLDs was irradiated separately at Sandia's Gamma Irradiation Facility (GIF) in a gamma field previously demonstrated to be uniform to within ±0.5% (1σ). TLDs were irradiated to initial absorbed-dose levels of 1, 2, 4, 7, 10, 20, 40, 70, 100, 200, 400, 700 and 1000 Gy.

To ensure that effects of fading were negligible for the purposes of this study, each group of 1400 irradiated TLDs was held for a minimum of 24 hours before being read. All TLDs within each group were read with the same TLD reader, over a time period of approximately 8 hours. Fading corrections were then applied, based on fading data previously obtained for TLDs from this same batch.

In addition to providing a baseline absorbed dose, the initial irradiation was also used to identify any TLDs that exhibited abnormal behavior with respect to batch sensitivity. All of the TLDs used in this study were new TLDs produced from the same batch of starting material in the manufacturing process. They were, however, TLDs from several different slugs, and had previously been rejected for routine use by the RML because they 1) did not as a group have sensitivity matching that for other TLDs received from the same batch, or 2) exhibited the presence of TLDs having anomalous response, based on random sampling of the TLDs. They were, however, completely suitable for use in an isolated study such as this, providing that the anomalous TLDs were identified and removed from the study.

The identity of each TLD within a group was maintained until all TLDs from the group had been read. Based on the mean and standard deviation for this group of readings, 1300 TLDs were selected for subsequent irradiations and a new standard deviation calculated for the group. The remaining 100 TLDs were kept in reserve in case a TLD from the 1300 was lost or broken and thus required replacement. If needed, a replacement TLD was chosen with a response that matched most closely that of the group of 1300 TLDs. Following this sorting, the group of 1300 TLDs was randomized, but the group identity maintained. This process was repeated for each of the 13 absorbed-dose levels.

Table 1 shows the results of the sorting process. For each of the initial absorbed-dose levels, batch standard deviation is shown before and after sorting. As can be seen from the table, removal of the 100 outlying TLDs from each group improved the uniformity of the batch from an average of 11.59% to 6.30%, an acceptable value for the present experiment.

Annealing and Second Irradiation

After all 13 groups of TLDs were irradiated and read, each group was annealed for 1 hour at 400°C and allowed to cool at room temperature. This is the annealing procedure normally followed by the RML during TLD preparation, and is in accordance with manufacture's recommendations and ASTM standards **[1]**. This annealing was essential for removing any residual thermoluminescence from the initial irradiation prior to subsequent irradiations. The TLDs were not cleaned.

Each group of 1300 TLDs was divided into 13 subgroups of 100 TLDs each. Maintaining the identity of each subgroup, a subgroup was taken from each absorbed-dose level from the first irradiation to form a new group of 1300 TLDs. To each set were added 100 TLDs that had not been previously irradiated. In this manner, 13 new groups of 1400 TLDs were formed. These groups of TLDs were irradiated as before to levels of 1, 2, 4, 7, 10, 20, 40, 70, 100, 200, 400, 700 and 1000 Gy.

Each group of TLDs was read as before, maintaining the identity of each subgroup. In general, the TLDs from each group were read in the order of lowest to highest initial dose. As a check on the validity of the fading corrections, and to insure that no biases were generated as a result of the order in which TLDs were read, three of the groups were read from highest to lowest initial dose. No systematic differences were identified in the groups that were read in reverse order.

Table 1. Batch Standard Deviation

Initial Dose (Gy)	Standard Deviation (%)	
	Pre-sort	Post-sort
1	13.36	8.20
2	13.13	5.79
4	12.25	5.43
7	11.70	5.36
10	10.73	5.46
20	13.70	7.93
40	12.61	5.86
70	12.34	6.56
100	12.32	7.41
200	11.67	6.41
400	9.23	5.45
700	8.28	5.96
1000	9.45	6.14
Mean	11.59	6.30

Results

Figure 1 shows normalized mean response for each of the subgroups of 100 TLDs. Each data point in Figure 1 represents the mean response of 100 TLDs, while each curve represents the data for a second irradiation at a given absorbed-dose level as identified in the legend. The data for each curve have been normalized to the mean response for the 1400 TLDs at that absorbed-dose level. Data for initial absorbed doses of 1, 100 and 400 Gy were invalidated because of reader malfunctions during the course of the readings, and are not included in Figure 1 or any subsequent analysis. There is an anomaly in the data shown in Figure 1 that must be addressed. A spike occurs in several of the curves at an initial irradiation of 200 Gy. This spike appears largest in the data for the second irradiation at 2 Gy, and becomes progressive smaller with increasing second dose. At absorbed doses larger than 20 Gy, the effect can no longer be seen. This behavior strongly suggests that there was a slight residual response from the initial irradiation at 200 Gy that was not completely eliminated during annealing.

In Figure 2 each data point represents the mean of the data points in Figure 1 for each initial absorbed dose. For the point at 200 Gy, however, only data for the second irradiations above 20 Gy are included. This eliminates the effects of the anomaly previously discussed, without changing the conclusions that can be drawn from the data. As can be seen from the data in Figure 2, there is a radical change in the slope of the curve, beginning at an initial dose of 100 Gy, but becoming significant above an initial dose of 200 Gy. This represents a decrease in sensitivity in the TLDs with increasing radiation dose. For TLDs used and tracked individually, these changes in sensitivity would not necessarily result in worsening of statistics, as illustrated in Table 2. The entries in Table 2 are the percent standard deviations of each of the subgroups of 100 TLDs. As can be seen from the table, there is no significant effect on the standard deviations of the individual subgroups. Thus, TLDs might successfully be reused if irradiation histories are maintained, and TLDs grouped into batches having similar irradiation histories. However, this can quickly result in a large number of very small batches, and the task of tracking them would become impractical if not impossible. Typically, such tracking is not done. Rather, all TLDs in a batch are used once, the entire batch is annealed and recalibrated as a new batch for the next use, and so on. A method is required for examining batch statistics that takes into account this mixing process. This is done in Table 3. Each row in Table 3 represents data for one initial dose level, while each column represent data for a second dose. Table entries are the normalized means as displayed in Figure 1. The rightmost column in Table 3 represents the standard deviation of the means for that initial dose and all initial doses less than that dose. This is equivalent to selecting an upper limit to initial dose, above which a TLD would not be reused, but below which no attempt is made to keep track of initial dose. Figure 3 shows a dramatic increase in percent standard deviation for initial doses above 100 Gy. Note also the slight increase in percent standard deviation from approximately 1.1 for an initial dose of zero (previously unirradiated) to approximately 1.5 for the smaller, but nonzero, initial doses. This suggests that there is a benefit from not reusing TLDs at all. Note that the percent standard deviations in Table 3 and Figure 3 are for the means of a large number of TLDs, and are therefore considerably smaller than what would be seen for TLDs in normal use. Furthermore, the data should be typical of uniformly distributed initial doses, and could be considerably different for applications in which a larger or smaller fraction of the irradiations is above 100 Gy.

Recommendations

Based on the data obtained in this study, it is clear that CaF_2:Mn TLDs do in fact suffer from sensitivity changes and degradation of batch statistics as a result of gamma-radiation doses in excess of 100 Gy. This result is entirely consistent with the earlier unpublished experiment even though the earlier experiment included data that were not as well controlled. The absence of effects at lower doses indicates that the effects are not due to readout or annealing procedures. Furthermore, these effects are seen even in the absence of neutrons. It is therefore recommended that CaF_2:Mn TLDs having received an initial gamma dose exceeding 100 Gy not be reused. In addition, they should not be used more than twice in high-dose applications where they are likely to receive cumulative absorbed doses exceeding 100 Gy unless individual TLD histories are maintained. In any case, a batch of TLDs to be reused must undergo stringent acceptance testing to verify suitability for the intended application.

This work does not address two aspects of CaF_2:Mn TLD reuse that might relax the severe restrictions recommended here. The first is the possibility that the sensitivity changes are not the result of cumulative doses, but rather the result of extreme doses, i.e., absorbed dose greater than 100 Gy. This would allow multiple uses with only those TLDs receiving any single dose greater than 100 Gy being discarded. It must be emphasized that such a result is unlikely, but only further irradiations can confirm this. A second possibility is that pretreatment with an extreme dose, e.g., 1000 Gy, might stabilize TLD sensitivity for all subsequent doses. Again, this experiment has not been performed, nor is it likely to be because of the very high costs associated with a study of this type.

Acknowledgments

The author wishes to express his gratitude to William Reed (deceased) and Hans Snyder for their careful and tireless efforts in the reading of TLDs for this study.

References

[1] American Society for Testing and Materials, "Standard Practice for Application of Thermoluminescence-Dosimetry (TLD) Systems for Determining Absorbed Dose in Radiation-Hardness Testing of Electronic Devices," ASTM E668-93, June, 1993.

Table 2. Subgroup Percent Standard Deviation

Initial Dose (Gy)	Second Dose (Gy)										Row Mean
	2	4	7	10	20	40	70	200	700	1000	
0	9.5	9.3	5.6	9.2	9.4	11.1	12.3	7.2	7.5	6.9	9.1
1	7.2	6.9	7.0	7.4	6.7	6.6	6.6	5.6	6.2	5.7	7.0
2	5.5	6.3	5.2	5.5	6.3	6.6	8.1	6.7	5.8	7.2	6.2
4	5.6	5.2	5.0	4.7	6.8	6.5	5.1	5.5	5.9	5.8	6.0
7	6.2	6.7	4.6	5.7	5.4	6.1	10.8	5.9	4.9	6.4	6.2
10	6.3	5.9	6.2	6.1	5.4	5.0	6.0	5.1	5.7	6.2	6.0
20	7.9	5.5	5.4	4.5	6.0	7.5	6.4	6.2	5.9	5.6	6.1
40	6.1	5.5	5.1	7.7	4.9	5.8	7.9	5.6	5.9	6.6	6.5
70	4.4	5.2	5.1	5.5	6.0	5.3	5.6	5.2	4.9	5.4	5.4
100	5.9	6.4	7.2	5.7	6.1	6.2	5.5	6.5	5.5	5.6	6.2
200	5.8	6.0	5.1	4.9	4.7	4.7	4.3	5.8	5.6	6.1	5.9
400	6.2	5.1	5.5	5.5	4.4	6.9	4.6	5.0	5.0	5.0	5.6
700	7.5	5.8	6.3	6.4	5.8	6.6	6.5	5.3	4.8	5.3	6.2
1000	9.4	7.3	6.2	6.3	7.3	6.5	6.9	6.7	5.9	5.6	7.0

Table 3. Normalized Subgroup Mean Dose

Initial Dose (Gy)	Second Dose (Gy)										Composite Standard Deviation (%)
	2	4	7	10	20	40	70	200	700	1000	
0	1.008	0.998	1.022	1.029	1.030	1.023	1.025	1.006	1.007	1.020	1.09
1	1.006	1.005	1.012	1.036	1.039	1.056	1.047	1.033	1.030	1.027	1.49
2	1.018	1.011	1.021	1.048	1.022	1.060	1.034	1.036	1.023	1.025	1.48
4	1.019	1.013	1.008	1.047	1.021	1.047	1.037	1.042	1.019	1.019	1.45
7	1.024	1.002	1.013	1.038	1.045	1.051	1.030	1.023	1.019	1.030	1.44
10	1.020	1.024	1.009	1.028	1.033	1.065	1.050	1.032	1.028	1.029	1.45
20	1.020	1.020	0.995	1.017	1.016	1.046	1.030	1.000	1.006	1.019	1.48
40	0.985	1.005	1.020	0.987	1.012	1.046	1.025	1.041	1.025	1.014	1.57
70	1.040	1.020	1.020	1.013	1.016	1.027	1.020	1.035	1.027	1.015	1.50
100	1.046	1.013	1.014	1.015	1.002	1.000	1.013	1.011	1.008	1.013	1.51
200	1.125	1.079	1.092	1.063	1.052	1.010	1.013	1.000	0.982	0.984	2.05
400	0.991	0.963	0.961	0.929	0.946	0.894	0.933	0.946	0.962	0.958	2.95
700	0.878	0.952	0.941	0.904	0.901	0.860	0.890	0.915	0.942	0.943	4.08
1000	0.820	0.894	0.872	0.845	0.863	0.816	0.853	0.880	0.921	0.904	5.51

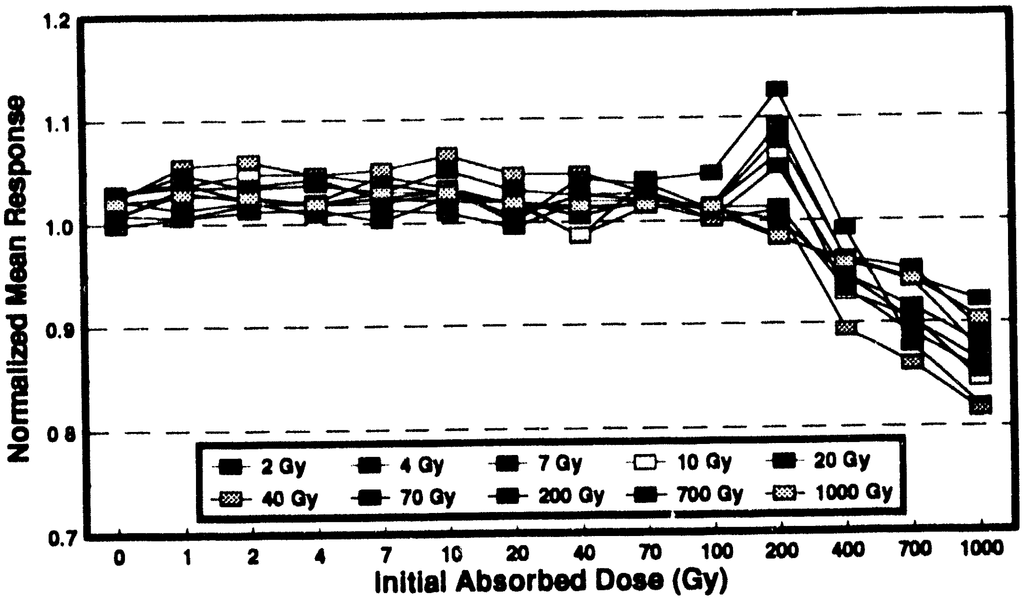

Figure 1. Normalized Mean Response vs. Initial Absorbed Dose

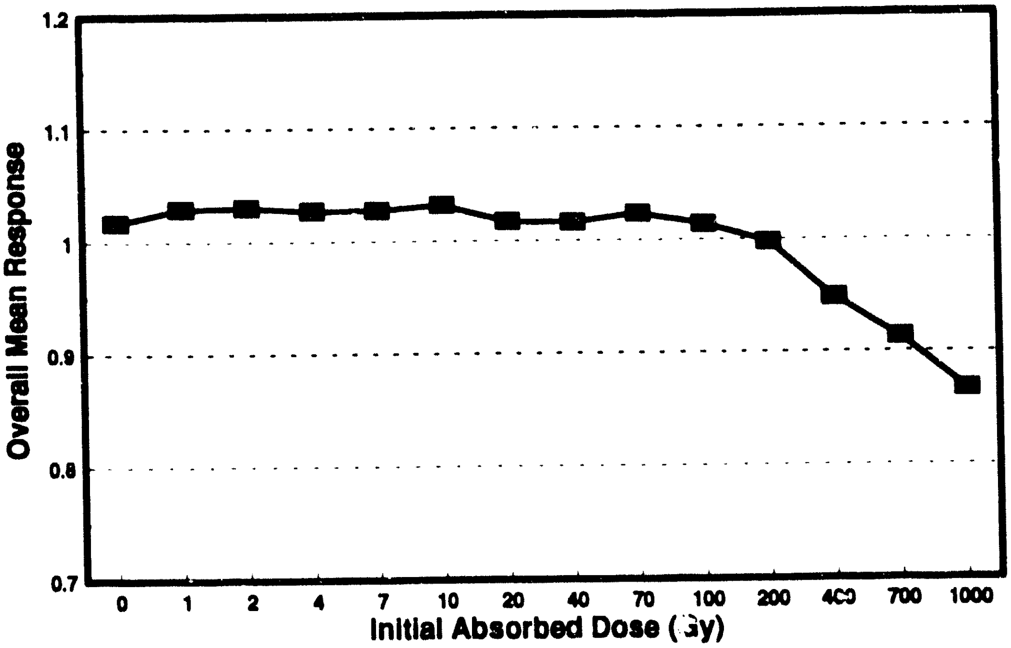

Figure 2. Overall Mean Response vs. Absorbed Dose

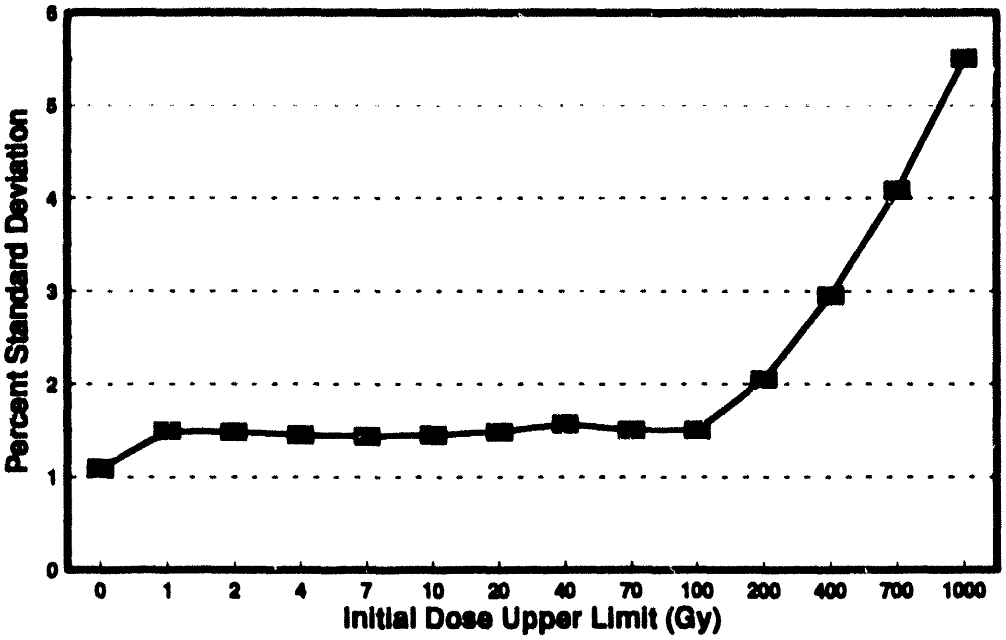

Figure 3. Percent Standard Deviation vs. Initial Dose Upper Limit

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