

PRE-IRRADIATION FADE AND POST-IRRADIATION FADE FOR LiF:Mg,Ti, TLD-600 AND TLD-700, AS A FUNCTION OF TIME

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Abstract — Pre-irradiation fade and post-irradiation fade are two contributing factors that affect the response of a thermoluminescent (TL) phosphor as a function of time. These phenomena have been extensively studied, yet the results from both experimental and theoretical calculations vary considerably. The Naval Dosimetry Center conducted several studies examining pre-irradiation fade and post-irradiation fade in the Harshaw/Bicron Radiation Measurement Product's multi-element ^6LiF (TLD-600) and ^7LiF (TLD-700) 8801 TLD cards. Photon and neutron irradiations were conducted using ^{226}Ra and PuBe sources, respectively. Pre-irradiation fade and post-irradiation fade investigations were conducted for evaluation periods that varied from 1 to 40 weeks. Findings show that the fade factor following neutron exposure is greater than from photon exposure alone. Further a fade algorithm based on a multiple regression model using two independent variables, the number of weeks of pre-irradiation fade and post-irradiation fade, gives a more accurate fade correction factor than Bicron's (our current) algorithm. This is especially true for neutron exposures.

INTRODUCTION

A known disadvantage of the use of LiF:Mg,Ti phosphors for dosimetry is their pre-irradiation and post-irradiation fade as a function of time. These phenomena have been extensively studied, yet the results from both experimental and theoretical calculations vary considerably⁽¹⁾. This is not surprising since many factors affect these phenomena, including batch dependence.

Currently many facilities are using a dosimetry system similar to that of the Naval Dosimetry Center. The dosimeter consists of three TLD-700 phosphors and one TLD-600 phosphor. Phosphors are processed on a Harshaw model 8800 automated thermoluminescence dosimetry (TLD) system. This dosimetry system is described elsewhere⁽²⁾.

Like the Dosimetry Center, most facilities use a fade correction algorithm that was established by the dosimeter's manufacturer, Bicron, Harshaw/Bicron Radiation Measurement Products*. This fade algorithm was established by Bicron from a 14 week fade study that only evaluated the diminished response of the TLD-700 phosphor to photon exposure⁽²⁾.

The Naval Dosimetry Center supplies dosimetry to over 500 facilities worldwide. Large variations in elapsed time are encountered between preparation (reader anneal), issue, collection, and processing of the dosimeter, some exceeding 14 weeks. Dosimetry issue may be delayed at intermediate locations for several

weeks pending distribution to end users. Also, dosimetry return may be delayed pending termination of extended deployments or irregularities involving overseas mail delivery. Dosimeters can be 'in the field' for periods ranging from 8 to 29 weeks. This results in many combinations of TLD pre-irradiation and post-irradiation fade.

The feasibility of improving the accuracy of a collective fade term, taking into account both pre-irradiation and post-irradiation fade over an extended period, and to investigate fade resulting from mixed-field photon and neutron exposure was considered worthy of investigation.

METHODS AND MATERIALS

The design methodology for this study was prompted by current Navy dosimetry issue and collection policies. Most Navy commands are on a 6 week issue cycle, the exception being Fleet Ballistic Submarine crews which are on a 12 week issue cycle. In extreme circumstances, activities are authorised to issue dosimetry up to 150 days (21.4 weeks). In addition, periods of pre-issue and post-issue were considered. Collectively these issue periods define the magnitude and type of fade the dosimeter is experiencing. During the pre-issue period the dosimeter undergoes pre-irradiation fade. During the issue period the dosimeter experiences both pre-irradiation and post-irradiation fade. During the post-issue period the dosimeter sustains only post-irradiation fade.

A time utilisation analysis was conducted to examine the elapsed times between dosimeter preparation

* Formally Solon Technologies, Inc.; Englehard; Harshaw/Filtrol; and Harshaw.

(reader anneal) and issue, issue and collection, and collection and processing. The maximum and minimum elapsed time between each of these events was evaluated to ascertain the duration of the pre-issue, issue, and post-issue periods. These results are shown in Figure 1. From this information an experimental design was developed, see Table 1. This design actually presents two independent studies. The first study evaluated all possible combinations of pre-irradiation fade and post-irradiation fade, by week, for a 10 week period and the second study evaluated selective combinations of pre-irradiation and post-irradiation fade over a 40 week period. Each number combination represents a sample of 5 dosimeters and gives the sample's pre-irradiation fade and post-irradiation fade in weeks, i.e. the number combination 7, 10 represents a sample of 5 dosimeters with 7 weeks pre-irradiation fade and 10 weeks post-irradiation fade. Number combinations that are shaded represent samples that were common in both the 10 week and 40 week studies. Each sample had a matching set of 5 control dosimeters. Averaged control dosimeter results were subtracted from the sample means.

Dosimeters were exposed to photons from a ²²⁶Ra source and to neutrons from a moderated PuBe source. Dosimeters received a ¹³⁷Cs equivalent dose of 3 mSv from each source. The dosimeter's photon response was derived by averaging the three TLD-700 phosphors. The neutron response was determined by subtracting the mean response of the three TLD-700 elements from the TLD-600 response. Changes in response were determined by normalising each sample's mean response to the mean response of a group of dosimeters that had 0 time pre-irradiation fade and 1 h post-irradiation fade.

Dosimeters used in the study were maintained at room temperature in opaque containers. All phosphors were factory annealed and reader annealed prior to the study. These parameters are given in Table 2. The reader's time-temperature profile (TTP), Table 2, was calibrated with TLD cards with 3 h pre-irradiation fade and 24 h post-irradiation fade. This is important because Bicron/Harshaw's fade algorithm was developed with a slightly different TTP and calibrated with TLD cards that had 8 days pre-irradiation fade and 1 day post-

irradiation fade⁽³⁾. The response of a phosphor was the total integrated area of the glow curve.

RESULTS

As seen in Figure 2, the photon response of TLD-700 initially fades faster due to post-irradiation fade than pre-irradiation fade. This inequality ceases to exist at approximately 10 weeks. At that time both phenomena change similarly. This holds true until about week 25, after which pre-irradiation fade slightly dominates.

For neutron exposures, the TLD-600 minus TLD-700 response, pre-irradiation fade and post-irradiation fade never coincide. As seen in Figure 3, post-irradiation fade always dominates.

Fade due to combinations of pre-irradiation and post-irradiation fade, when compared to the corresponding fade due solely to pre-irradiation or post-irradiation, varies depending on the relative contribution of the two underlying fade processes. As seen in Table 3, these variations are small for photon fade since neither pre-irradiation fade nor post-irradiation fade dominates. However, as seen in Table 4, this is not true for neutron fade since post-irradiation fade is more dominant than pre-irradiation fade.

As shown in Figures 4 and 5, the response of the phosphors changes most during the first 10 weeks after preparation (reader anneal). At 10 weeks the photon fade ranged from 22% to 30%. By the end of 40 weeks these values increased by another 10%. For the neutron component these values ranged from 22% to 41% at 10 weeks and 27% to 51% at 40 weeks. The predominantly larger neutron fade resulted from combinations favouring post-irradiation fade.

The data were analysed using both simple and multiple regression models. The simple regression model related the photon and neutron responses to a single independent variable, weeks between preparation and processing. The multiple regression models related the response to two independent variables, the weeks of pre-irradiation fade and weeks of post-irradiation fade. To delineate distinct periods of pre- and post-irradiation fade for the multiple regression models, it was assumed

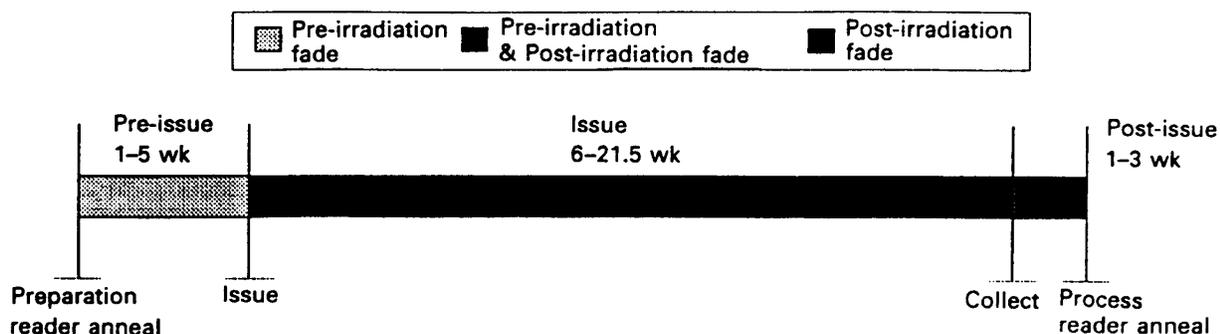


Figure 1. Duration of pre-issue, issue, and post-issue dosimetry periods.

that the exposure occurred half-way through the issue period. Both the simple and multiple regression models fit the data adequately for the photon responses with r^2 values of 0.93 and 0.82, respectively. However, for the neutron response the multiple regression model provided a much better fit than the simple regression model with r^2 values of 0.78 and 0.37, respectively.

DISCUSSION AND CONCLUSION

The exact mechanisms governing fade in LiF phosphors are unknown and are dependent on many factors. For neutron exposure, our data support the findings of

Table 2. Factory anneal and reader time-temperature profile (TTP) anneal parameters.

Factory oven anneal	
400°C for 1 h	
100°C for 2 h	
Room temperature quench	
Reader anneal (TTP)	
Preheat temperature:	100°C
Preheat time:	2 s
Heating rate:	30°C.s ⁻¹
Maximum temperature:	300°C
Acquire time:	13.3 s
Anneal temperature:	300°C
Anneal time:	3 s

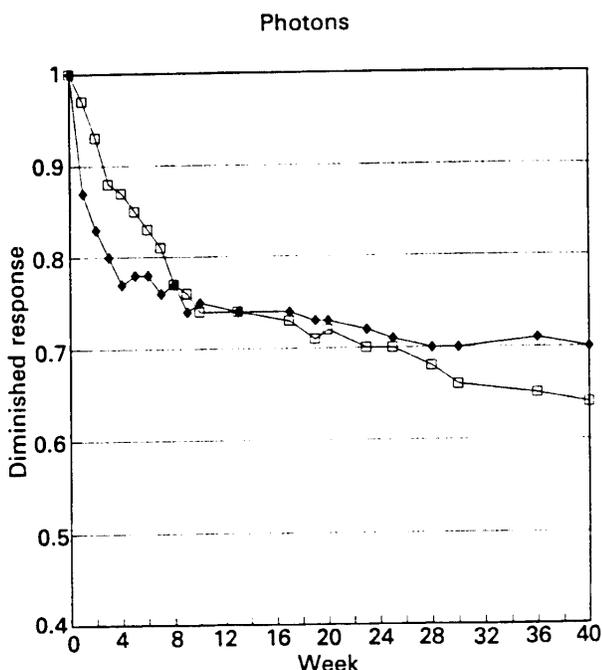


Figure 2. TLD-700 photon fade due solely to pre-irradiation (□) and post-irradiation (◆) fade.

Johnson and Luersen⁽⁴⁾ that show a greater amount of fade (post-irradiation fade) for thermal neutrons than for photons. We can speculate that the increased post-irradiation fade for neutrons is due to greater interactions between the higher concentration of charged carrier defects distributed around charged particle tracks. This leads to an increase in the number of thermal escape reactions between defects with trapped charge carriers and possibly by the conversion of peaks 4 and 5 to higher energy traps. However, Shachar and Horowitz saw no significant difference in the fading of LiF-100 between photon and high LET exposures⁽⁵⁾.

It must also be realised that since the TLD-600 phosphor responds to both neutrons and photons, its overall post-irradiation fade will not only be a function of time, but also a function of the relative contribution from the two types of radiation. This certainly complicates the use of a neutron fade factor unless the relative contribution is known, or is always the same.

If the relative contribution is known or is always the same, a neutron fade factor could be determined for that specific photon to neutron ratio. Otherwise a third independent variable, the photon to neutron ratio (the TLD-600 minus TLD-700 to TLD-700 ratio) is needed in the model. This ratio should be determined using the ratio of the ¹³⁷Cs equivalent* values, that is, the raw chip values. This gives an unbiased neutron to photon ratio that is independent of the energy spectrum of the two

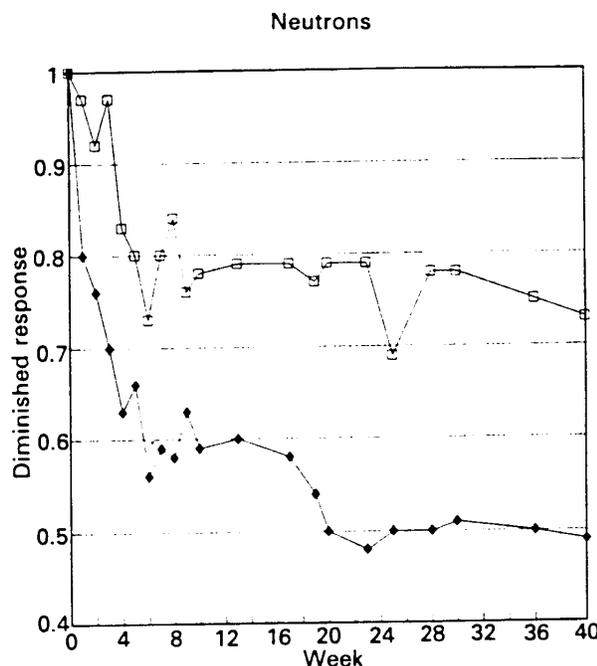


Figure 3. TLD-600 minus TLD-700 neutron fade due solely to pre-irradiation (□) and post-irradiation (◆) fade.

* Assumes TLD reader is calibrated with a ¹³⁷Cs source.

Table 3. Diminished photon response of TLD-700.

		Post-irradiation fade (wk)																																																		
Post-irradiation fade (wk)	0	Post-irradiation fade (wk)																																																		
		1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18	19	20	21	22	23	24	25	26	27	28	29	30	31	32	33	34	35	36	37	38	39	40											
0	1.00	0.87	0.83	0.80	0.77	0.78	0.78	0.76	0.77	0.74	0.75	-	0.74	-	-	0.74	-	0.74	-	0.73	0.73	-	0.72	-	0.71	-	0.71	-	0.70	-	0.70	-	0.70	-	0.70	-	0.70	-	0.70	-	0.70	-	0.70	-	0.70							
1	0.97	0.89	0.85	0.81	0.82	0.80	0.81	0.81	0.79	0.78	-	0.76	-	0.76	-	0.76	-	0.74	0.76	-	0.74	0.76	-	0.76	-	0.74	-	0.74	-	0.72	-	0.72	-	0.72	-	0.72	-	0.72	-	0.71	-	0.71	-	0.71	-	0.71						
2	0.93	0.85	0.80	0.81	0.78	0.79	0.79	0.78	0.77	-	0.75	-	0.75	-	0.75	-	0.74	0.75	-	0.74	0.75	-	0.73	-	0.73	-	0.73	-	0.71	-	0.71	-	0.71	-	0.71	-	0.71	-	0.71	-	0.71	-	0.71	-	0.71	-	0.71					
3	0.88	0.80	0.81	0.80	0.78	0.78	0.79	0.77	0.77	-	0.75	-	0.74	-	0.74	-	0.74	0.75	-	0.74	0.75	-	0.74	-	0.72	-	0.72	-	0.71	-	0.70	-	0.70	-	0.70	-	0.70	-	0.70	-	0.70	-	0.70	-	0.70	-	0.70					
4	0.87	0.83	0.80	0.79	0.79	0.79	0.76	0.74	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-			
5	0.85	0.80	0.78	0.77	0.75	0.73	-	-	0.71	-	-	0.71	-	0.71	0.73	-	0.71	0.70	-	0.71	0.70	-	0.68	0.68	-	0.68	0.68	-	0.68	-	0.67	-	0.67	-	0.67	-	0.67	-	0.67	-	0.67	-	0.67	-	0.67	-	0.67	-	0.67			
6	0.83	0.77	0.76	0.74	0.74	-	-	0.71	-	-	0.70	0.70	0.71	-	0.69	0.68	-	0.68	0.68	-	0.67	0.68	-	0.67	0.68	-	0.67	0.68	-	0.66	-	0.66	-	0.66	-	0.66	-	0.66	-	0.66	-	0.66	-	0.66	-	0.66	-	0.66	-	0.66		
7	0.81	0.76	0.72	0.71	-	-	0.71	-	-	0.70	0.70	0.71	-	0.69	0.68	-	0.68	0.68	-	0.67	0.68	-	0.67	0.68	-	0.67	0.68	-	0.66	-	0.66	-	0.66	-	0.66	-	0.66	-	0.66	-	0.66	-	0.66	-	0.66	-	0.66	-	0.66	-	0.66	
8	0.77	0.73	0.70	-	-	-	-	-	-	0.69	0.67	0.68	-	0.67	0.66	-	0.66	0.66	-	0.65	0.64	-	0.65	0.64	-	0.64	0.64	-	0.64	-	0.64	-	0.64	-	0.64	-	0.64	-	0.64	-	0.64	-	0.64	-	0.64	-	0.64	-	0.64	-	0.64	
9	0.76	0.72	-	-	-	-	-	-	-	0.69	0.67	0.68	-	0.67	0.66	-	0.66	0.66	-	0.65	0.64	-	0.65	0.64	-	0.64	0.64	-	0.64	-	0.64	-	0.64	-	0.64	-	0.64	-	0.64	-	0.64	-	0.64	-	0.64	-	0.64	-	0.64	-	0.64	
10	0.74	-	0.69	-	-	-	0.69	-	0.69	0.67	0.68	-	0.67	0.66	-	0.66	0.66	-	0.65	0.64	-	0.65	0.64	-	0.64	0.64	-	0.64	-	0.64	-	0.64	-	0.64	-	0.64	-	0.64	-	0.64	-	0.64	-	0.64	-	0.64	-	0.64	-	0.64		
11	-	-	-	-	-	-	-	-	-	0.69	0.67	0.68	-	0.67	0.66	-	0.66	0.66	-	0.65	0.64	-	0.65	0.64	-	0.64	0.64	-	0.64	-	0.64	-	0.64	-	0.64	-	0.64	-	0.64	-	0.64	-	0.64	-	0.64	-	0.64	-	0.64	-	0.64	
12	-	-	-	-	-	-	-	-	-	0.69	0.67	0.68	-	0.67	0.66	-	0.66	0.66	-	0.65	0.64	-	0.65	0.64	-	0.64	0.64	-	0.64	-	0.64	-	0.64	-	0.64	-	0.64	-	0.64	-	0.64	-	0.64	-	0.64	-	0.64	-	0.64	-	0.64	
13	0.74	-	-	0.68	-	0.68	0.68	0.68	-	0.66	0.66	0.68	-	0.65	0.65	-	0.64	0.64	-	0.64	0.64	-	0.63	0.63	-	0.63	0.63	-	0.63	-	0.63	-	0.63	-	0.63	-	0.63	-	0.63	-	0.63	-	0.63	-	0.63	-	0.63	-	0.63	-	0.63	
14	-	-	-	-	-	-	-	-	-	0.66	0.66	0.68	-	0.65	0.65	-	0.64	0.64	-	0.64	0.64	-	0.63	0.63	-	0.63	0.63	-	0.63	-	0.63	-	0.63	-	0.63	-	0.63	-	0.63	-	0.63	-	0.63	-	0.63	-	0.63	-	0.63	-	0.63	
15	-	-	-	-	-	-	-	-	-	0.66	0.66	0.68	-	0.65	0.65	-	0.64	0.64	-	0.64	0.64	-	0.63	0.63	-	0.63	0.63	-	0.63	-	0.63	-	0.63	-	0.63	-	0.63	-	0.63	-	0.63	-	0.63	-	0.63	-	0.63	-	0.63	-	0.63	
16	-	-	-	-	-	-	-	-	-	0.66	0.66	0.68	-	0.65	0.65	-	0.64	0.64	-	0.64	0.64	-	0.63	0.63	-	0.63	0.63	-	0.63	-	0.63	-	0.63	-	0.63	-	0.63	-	0.63	-	0.63	-	0.63	-	0.63	-	0.63	-	0.63	-	0.63	
17	0.73	-	0.67	0.69	-	0.65	-	0.65	0.64	-	0.63	0.62	-	0.62	0.62	-	0.62	0.62	-	0.62	0.62	-	0.62	0.62	-	0.62	0.62	-	0.62	-	0.62	-	0.62	-	0.62	-	0.62	-	0.62	-	0.62	-	0.62	-	0.62	-	0.62	-	0.62			
18	-	-	-	-	-	-	-	-	-	0.65	0.65	0.65	-	0.62	0.62	-	0.62	0.62	-	0.61	0.61	-	0.62	0.62	-	0.62	0.62	-	0.62	-	0.62	-	0.62	-	0.62	-	0.62	-	0.62	-	0.62	-	0.62	-	0.62	-	0.62	-	0.62	-	0.62	
19	0.71	0.69	-	0.65	-	0.65	-	0.65	0.65	-	0.62	0.62	-	0.62	0.62	-	0.62	0.62	-	0.61	0.61	-	0.62	0.62	-	0.62	0.62	-	0.62	-	0.62	-	0.62	-	0.62	-	0.62	-	0.62	-	0.62	-	0.62	-	0.62	-	0.62	-	0.62	-	0.62	
20	0.72	-	0.66	0.65	-	0.65	-	0.65	0.65	-	0.62	0.62	-	0.62	0.62	-	0.62	0.62	-	0.62	0.62	-	0.62	0.62	-	0.62	0.62	-	0.62	-	0.62	-	0.62	-	0.62	-	0.62	-	0.62	-	0.62	-	0.62	-	0.62	-	0.62	-	0.62	-	0.62	
21	-	-	-	-	-	-	-	-	-	0.65	0.65	0.65	-	0.62	0.62	-	0.62	0.62	-	0.62	0.62	-	0.62	0.62	-	0.62	0.62	-	0.62	-	0.62	-	0.62	-	0.62	-	0.62	-	0.62	-	0.62	-	0.62	-	0.62	-	0.62	-	0.62	-	0.62	
22	-	-	-	-	-	-	-	-	-	0.65	0.65	0.65	-	0.62	0.62	-	0.62	0.62	-	0.62	0.62	-	0.62	0.62	-	0.62	0.62	-	0.62	-	0.62	-	0.62	-	0.62	-	0.62	-	0.62	-	0.62	-	0.62	-	0.62	-	0.62	-	0.62	-	0.62	
23	0.70	-	0.66	-	0.63	-	0.63	0.62	-	0.62	0.62	-	0.62	0.62	-	0.62	0.62	-	0.62	0.62	-	0.62	0.62	-	0.62	0.62	-	0.62	-	0.62	-	0.62	-	0.62	-	0.62	-	0.62	-	0.62	-	0.62	-	0.62	-	0.62	-	0.62	-	0.62		
24	-	-	-	-	-	-	-	-	-	0.63	0.62	-	0.62	0.62	-	0.62	0.62	-	0.62	0.62	-	0.62	0.62	-	0.62	0.62	-	0.62	-	0.62	-	0.62	-	0.62	-	0.62	-	0.62	-	0.62	-	0.62	-	0.62	-	0.62	-	0.62	-	0.62		
25	0.70	-	0.63	-	0.63	0.62	-	0.62	0.62	-	0.62	0.62	-	0.62	0.62	-	0.62	0.62	-	0.62	0.62	-	0.62	0.62	-	0.62	0.62	-	0.62	-	0.62	-	0.62	-	0.62	-	0.62	-	0.62	-	0.62	-	0.62	-	0.62	-	0.62	-	0.62	-	0.62	
26	-	-	-	-	-	-	-	-	-	0.63	0.62	-	0.62	0.62	-	0.62	0.62	-	0.62	0.62	-	0.62	0.62	-	0.62	0.62	-	0.62	-	0.62	-	0.62	-	0.62	-	0.62	-	0.62	-	0.62	-	0.62	-	0.62	-	0.62	-	0.62	-	0.62		
27	-	-	-	-	-	-	-	-	-	0.63	0.62	-	0.62	0.62	-	0.62	0.62	-	0.62	0.62	-	0.62	0.62	-	0.62	0.62	-	0.62	-	0.62	-	0.62	-	0.62	-	0.62	-	0.62	-	0.62	-	0.62	-	0.62	-	0.62	-	0.62	-	0.62		
28	0.68	-	0.63	-	0.63	0.62	-	0.61	0.61	-	0.61	0.61	-	0.61	0.61	-	0.61	0.61	-	0.61	0.61	-	0.61	0.61	-	0.61	0.61	-	0.61	-	0.61	-	0.61	-	0.61	-	0.61	-	0.61	-	0.61	-	0.61	-	0.61	-	0.61	-	0.61	-	0.61	
29	-	-	-	-	-	-	-	-	-	0.61	0.61	-	0.61	0.61	-	0.61	0.61	-	0.61	0.61	-	0.61	0.61	-	0.61	0.61	-	0.61	-	0.61	-	0.61	-	0.61	-	0.61	-	0.61	-	0.61	-	0.61	-	0.61	-	0.61	-	0.61	-	0.61	-	0.61
30	0.66	-	-	-	-	0.61	-	0.60	0.60	-	0.60	0.60	-	0.60	0.60	-	0.60	0.60	-	0.60	0.60	-	0.60	0.60	-	0.60	0.60	-	0.60	-	0.60	-	0.60	-	0.60	-	0.60	-	0.60	-	0.60	-	0.60	-	0.60	-	0.60	-	0.60	-	0.60	
31	-	-	-	-	-	0.61	-	0.60	0.60	-	0.60	0.60	-	0.60	0.60	-	0.60	0.60	-	0.60	0.60	-	0.60	0.60	-	0.60	0.60	-	0.60	-	0.60	-	0.60	-	0.60	-	0.60	-	0.60	-	0.60	-	0.60	-	0.60	-	0.60	-	0.60	-	0.60	
32	-	-	-	-	-	0.61	-	0.60	0.60																																											

fields. This is important since both phosphors exhibit energy dependence.

For these studies the ^{137}Cs equivalent neutron to photon ratio was 1. This is the ratio the Navy's dosimeter exhibits after exposure to bare ^{252}Cf . However, the Navy's dosimeter is exposed to a wide variety of neutron spectra, many similar to moderated ^{252}Cf that gives a neutron to photon (^{137}Cs equivalent) ratio of 10 to 1. Therefore, we will continue our fade studies, evaluating different neutron to photon ratios.

The impetus behind our research was not to arrive at a universally accepted characterisation of the fading properties of TLD-600 and TLD-700. Instead it was to encourage dosimetrists using a LiF:Mg,Ti based system to develop their own fade algorithm based on their own operating procedures. To that end we have provided a fundamental experimental design and mathematical

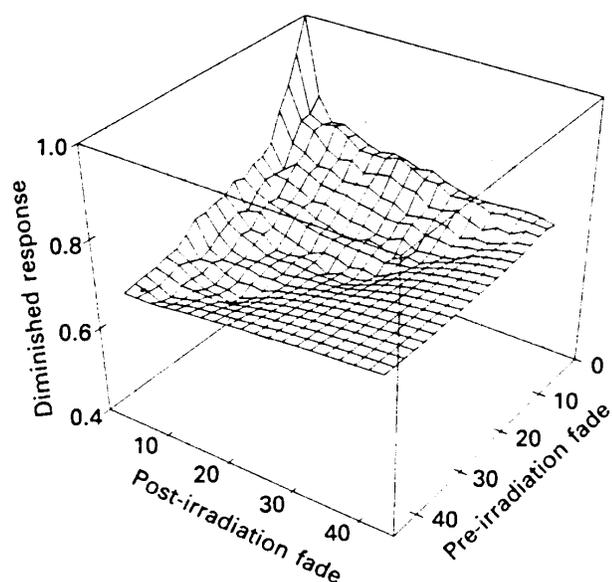


Figure 4. All combinations of TLD-700 photon pre-irradiation and post-irradiation fade.

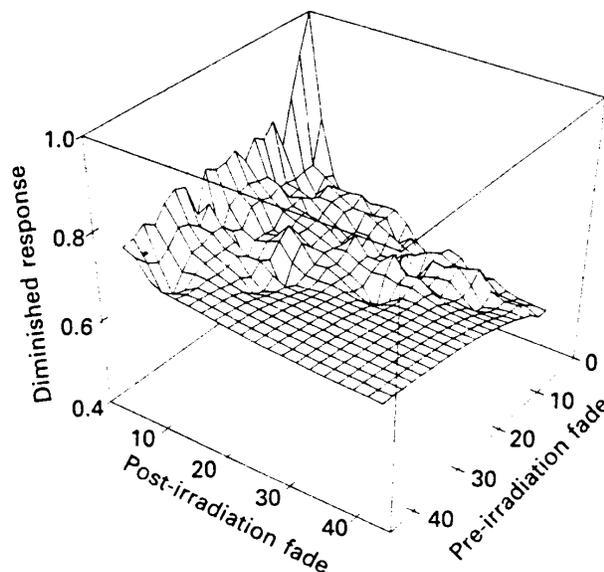


Figure 5. All combinations of TLD-600 minus TLD-700 neutron pre-irradiation and post-irradiation fade.

model. As aptly stated by Shachar and Horowitz⁽⁵⁾ 'inclusion of peaks 2 and 3 (or even peak 3 alone) in the dosimetric signal results, as expected, in very high and irregular fading rates due to the very rapid decay of these peaks'. This is quite evident by our research, which shows a photon and neutron fade of 40% and 50%, respectively, over 40 weeks. This is extremely important since most processors using a Bicon/Harshaw TLD-600/TLD-700 system include peaks 2 and 3 in the dosimetric signal for routine (not accident) dosimetry.

In conclusion, from our studies a 5% improvement in the accuracy of our dosimetry was achieved by using a simple or multiple regression model developed for our photon fade factor and a 10% improvement in our accuracy using a multiple regression model for our neutron fade factor. It is significant that the neutron component of TLD-600 loses signal faster than the photon response of TLD-700.

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