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EVALUATION OF NEW AND CONVENTIONAL THERMOLUMINESCENT PHOSPHORS FOR ENVIRONMENTAL MONITORING USING AUTOMATED THERMOLUMINESCENT DOSIMETER READERS

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In recent years, there has been considerable interest in a new generation of super-sensitive thermoluminescent (TL) phosphors for potential use in routine personnel and environmental monitoring (Prokic and Botter-Jensen 1993; Akselrod et al. 1993; Zha et al. 1993; Bacci et al. 1993; Horowitz 1993). Two of these phosphors, $\alpha\text{-Al}_2\text{O}_3\text{:C}$ and LiF:Mg,Cu,P , are evaluated in this paper for selected characteristics relevant to environmental monitoring, along with two conventional phosphors widely used in environmental monitoring, LiF:Mg,Ti and $\text{CaF}_2\text{:Dy}$. The characteristics evaluated are light-induced fading, light-induced background, linearity and variability at low dose, and the minimum measurable dose. These characteristics were determined using an automated commercial dosimetry system (Harshaw System 8800) and routine processing protocols. Annealing and readout protocols for each phosphor were optimized for use in a large-scale environmental monitoring program.

The use of TL phosphors in the Harshaw system requires mounting of the phosphors on a substrate such as polytetrafluoroethylene (PTFE) or polyimide (e.g., Kapton) film and suspension of the film in a rigid card structure with bar code label. The mounting of phosphors in cards poses limits to the oven-annealing and readout temperatures that may be applied. The inability to clear high-temperature traps in LiF:Mg,Ti , in LiF:Mg,Cu,P , and in $\text{CaF}_2\text{:Dy}$ can result in significant residual. The inability to clear high-temperature traps in $\alpha\text{-Al}_2\text{O}_3\text{:C}$ can result in significant light-induced background in chips with large dose histories (Akselrod et al. 1993). The manual handling of cards required during loading and unloading of dosimeter holders and the loading and unloading of the reader introduces the potential for contamination and requires some exposure to light. Significant light-induced fading has been reported for $\text{Al}_2\text{O}_3\text{:C}$ (Musk 1993; Mosovitch et al. 1993).

MATERIALS AND METHODS

All thermoluminescent dosimeter (TLD) cards used in this study were manufactured by Bicron for use in the Harshaw Model 8800 TLD reader. The $\alpha\text{-Al}_2\text{O}_3\text{:C}$ TL phosphor used in these cards is manufactured as single crystal chips by Urals Polytechnical Institute, Ekaterinburg, Russia, and marketed by Bicron under the product name TLD 500. For convenience, this phosphor will be referred to as TLD 500. The LiF:Mg,Cu,P phosphor used in these cards is manufactured as hot-pressed chips by Solid Dosimetric and Method Laboratory, Beijing, Peoples Republic of China, under the product name GR 207 and marketed by Bicron as TLD 700H. For convenience, this phosphore will be referred to as TLD 700H. Both the TLD 700H phosphor and TLD 700 have a Li^6 abundance of 0.007%. The $\text{CaF}_2\text{:Dy}$ and LiF:Mg,Ti phosphors used in these cards are manufactured and marketed by Bicron under the product names TLD 200 and TLD 700, respectively, and will be referred to by these names for convenience.

The TLD 200 and TLD 700 chips (3 mm x 3 mm x 0.9 mm) were mounted between two layers of PTFE film to form TL elements on Bicron's standard type 8807 Environmental TLD card, having two TLD 200 and two TLD 700 elements per card. The TLD 500 chips (5 mm x 0.5 mm) were adhesively bonded to Kapton substrate to form TL elements, with four elements mounted per card. The TLD 700H chips (3 mm x 3 mm x 0.5 mm) were also adhesively bonded to Kapton to form TL elements with four elements per card. Thus, the TLD 500 and TLD 700H phosphor types were represented by their own card types.

The readout equipment used was the Harshaw Model 8800 TLD reader. This is a large-capacity automated reader that uses heated nitrogen gas to heat the chips according to a user-programmable linear time-temperature profile (TTP).

The anneal and readout protocols used for all experiments described in this paper were designed to optimize dosimetry performance while maintaining reasonable throughput and handling convenience in a large-scale program. For each phosphor type, the same anneal and readout protocols were used for all experiments. The time-temperature profiles used for each phosphor type are shown in Table 1. For the TLD 200 and TLD 700 chips, separate TTPs were used for annealing and for dosimetric readout. For these phosphors, the anneal procedure consisted of readout under the anneal TTP followed by a 16-hour oven anneal at 80°C. Oven annealing reduces the traps available for peaks 2 and 3 in TLD 700, thereby improving fade characteristics but has no effect on the TLD 200 chips, which are on the same card. For TLD 500, annealing consisted of one reading using the TTP for dosimetric readout, with no oven anneal. For TLD 700H, annealing consisted of two readings with the dosimetric TTP and no oven anneal. This TTP was chosen after our own studies on sensitivity loss with reuse as a function of maximum readout temperature showed no significant loss of sensitivity with up to 100 readouts at 270°C. This readout temperature gives a residual of less than 1%, which was considered adequate for environmental dosimetry. These results are in agreement with those of other investigators (Oster et al. 1993).

The TLD 500 and TLD 700H elements were calibrated using a ^{90}Sr card irradiator. The TLD 200 and TLD 700 elements were calibrated in air using a ^{60}Co source. The calibration process provides an element correction coefficient (ECC) for each chip that normalizes its sensitivity to the mean sensitivity of the population of calibration chips in the same position. These ECCs were applied to all readings.

For each experiment, the reader was calibrated to provide reader calibration factors (RCFs) for each TTP used. These RCFs were based on the response of calibration cards exposed in the ^{90}Sr card irradiator. This irradiator is calibrated to provide dose in generic units (gU), where one generic unit is approximately equivalent to one millirad or 10 μGy . The RCFs based on this irradiator, therefore, have units of nC/gU. These RCFs were applied by the reader to all card readings.

TABLE 1. Time-Temperature Profiles

TLD Phosphor Type	TLD 200	TLD 200 (anneal)	TLD 700	TLD 700 (anneal)	TLD 500	TLD 700H
Preheat temperature (°C)	160	50	50	50	50	160
Preheat time (sec)	25	0	0	0	0	5
Temperature rate (°C/sec)	20	20	10	10	5	20
Maximum temperature (°C)	300	300	300	300	300	270
Acquire time (sec)	20	40	33.3	33.3	60	16.6
Anneal temperature (°C)	0	0	0	300	0	0
Anneal time (sec)	0	0	0	6	0	0

Light-Induced Fading

White, yellow, and red lighting environments were constructed in the laboratory to produce uniform lighting at a level of 300 lux on designated work surfaces. The white lighting was provided by standard 40-watt warm white fluorescent tubes; the yellow lighting was provided by General Electric F40G0 gold fluorescent tubes; the red lighting was provided by incandescent red bulbs. All lighting was filtered by transparent ultraviolet (UV) blocking film. Cards containing each chip type were annealed, then exposed to 100 gU (1 mGy) using a ^{90}Sr card irradiator. Ten cards of each type were exposed for durations of 1, 3, 10, 30, 100, and 300 minutes to the warm white source. Ten cards of each type were also exposed to the yellow and red light for the same durations. The same day, cards of each type were read together along with a set of controls of each card type exposed to the ^{90}Sr source but not to light.

Light-Induced Background

Groups of 10 TLD 500 cards were annealed, then exposed to red, yellow, and warm white light at an illumination level of 300 lux for durations of 1, 3, 10, 30, 100, and 300 minutes and read the same day with unexposed controls. To the author's knowledge, each of these chips had a cumulative radiation dose history of 1 Gy or less before performing this experiment. Throughout each of these light experiments, great care was taken to avoid unnecessary exposure to light.

Linearity and Variability at Low Dose

After annealing, cards of each type were loaded into Harshaw 8807 Environmental Dosimeter holders to minimize exposure to light. The holders used for the TLD 500 cards and for the TLD 700H cards provided 80 mg/cm² of ABS plastic filtration. The holders used for the 8807 card (TLD 200 and TLD 700) provided 422 mg/cm² Ta + 58 mg/cm² Pb filtration over front and back sides of the TLD 200 elements in addition to the 80 mg/cm² ABS over all elements. Groups containing 10 dosimeters of each type (8807, TLD 500, TLD 700H) were exposed to 0.1, 0.3, 1.0, 3.0, 10.0, 30.0, and 100.0 mR in air from a ¹³⁷Cs source. The estimated error in the applied exposure in all cases was less than 5%. A group containing 10 dosimeters of each type was designated as a control and accompanied the experimental dosimeters to and from the irradiation facility. Card annealing was performed the day before irradiations and readout was conducted on the same day as the irradiations.

Minimum Measurable Dose

The standard deviation of control dosimeters in the linearity test was used to calculate the minimum measurable dose for each phosphor type.

RESULTS AND DISCUSSION

Light-Induced Fading

The results of the light-induced fading experiments are summarized in Table 2. All readings were obtained with ECC and RCF (nC/gU) applied. To obtain the fade factor, the mean result for each chip position in an exposure group was normalized to the mean of exposed control chips in the same position. The fade factors for all chip positions were averaged to obtain the single fade factor shown in Table 2. Thus, for TLD 500 and TLD 700H, the fade factor was based on 40 data points per exposure condition. For TLD 200 and TLD 700, the average was based on 20 chips per exposure condition. The relative standard deviation was calculated for each chip position and the average reported in Table 2. In general, the results were as expected. The only phosphor to show significant fading was the TLD 500. The data for this phosphor are plotted in Figure 1. Other investigators have reported strong light-induced fading in TLD 500 for white and yellow light. It is interesting to note the superior performance of red lighting at the same level of illumination as the other sources. Other investigators have reported significant UV light-induced fading in TLD 200. The fact that none was observed in the present experiments attests to the effectiveness of the UV filter employed. Minimal sensitivity to visible light has been reported in the literature for both TLD 700 and TLD 700H, which is consistent with the data in Table 2.

TABLE 2. Light-Induced Fading

Time, min	Red		Yellow		White	
	Avg	%Std	Avg	%Std	Avg	%Std
TLD 200						
1	0.99	2.8	1.00	2.7	1.00	2.5
3	0.99	1.9	1.01	3.5	0.99	1.5
10	1.01	2.3	1.00	2.2	1.00	2.0
30	0.99	3.2	0.99	2.0	0.98	2.5
100	1.00	1.1	1.01	2.2	1.00	1.6
300	1.00	1.9	1.00	2.1	1.01	1.3
TLD 700						
1	1.00	1.2	1.00	1.3	0.99	1.3
3	0.99	1.0	1.00	1.1	1.00	1.4
10	1.00	1.0	1.00	0.6	0.99	1.8
30	0.99	1.4	1.00	1.2	1.00	1.6
100	1.00	1.3	1.00	2.3	1.00	1.7
300	1.00	0.9	1.03	2.3	1.01	1.7
TLD 500						
1	1.01	0.6	1.00	0.6	1.00	0.6
3	1.00	0.4	1.00	0.5	1.00	1.5
10	1.00	0.5	0.98	0.5	0.97	0.9
30	0.99	0.8	0.94	1.4	0.91	2.6
100	0.93	1.7	0.82	4.0	0.73	6.2
300	0.80	5.1	0.58	12.1	0.42	25.6
TLD 700H						
1	0.99	1.3	1.01	1.3	0.98	1.8
3	0.98	2.0	0.99	2.4	0.98	2.8
10	0.98	1.9	0.99	2.4	0.98	1.7
30	0.99	1.8	0.99	2.0	0.99	2.3
100	0.99	1.9	0.97	2.6	0.99	2.9
300	1.00	2.4	0.96	2.0	0.98	1.3

FIGURE 1. Light-Induced Fading in TLD 500

Light-Induced Background

The cards were read on a reader previously calibrated with cards exposed to 100 μGy with the ^{90}Sr card irradiator. The ECCs and RCFs were applied to all chip readings. For each exposure group (five cards) the mean and standard deviation for each chip position were calculated and the results averaged to obtain a single mean and standard deviation value for the group. The results are shown in Table 3 and Figure 2. Even though the cards had a prior dose history of less than 0.01 Gy, a small background effect is evident. It is believed that the light-induced background in TLD 500 results from light-transferred sensitivity from higher temperature traps and that the background effect will be greater with larger cumulative doses on the chip.

TABLE 3. Light-Induced Background at 300 lux, μGy

Exposure, min	Red		Yellow		White	
	Avg	%Std	Avg	%Std	Avg	%Std
0	2.90	0.27	2.90	0.27	2.90	0.27
1	3.24	0.29	2.90	0.19	3.09	0.36
3	3.17	0.53	3.00	0.27	3.07	0.28
10	3.20	0.32	3.04	0.43	3.78	0.86
30	2.94	0.38	3.05	0.38	4.48	1.38
100	3.27	0.38	3.28	0.43	5.50	2.13
300	3.43	0.33	3.78	0.64	9.25	4.27

FIGURE 2. Light-Induced Background in TLD 500

Linearity and Variability at Low Dose

Cards were read on a reader calibrated using the ^{90}Sr irradiator, and ECCs and RCFs were applied to all chip readings by the reader. For each chip type, the zero dose reading from the set of control dosimeters was subtracted from the reading of each experimental dosimeter and the net reading adjusted by a one-point calibration factor (gU/mR) derived from the response of the dosimeters exposed to 100 mR. For each group, the mean and standard deviation of these net adjusted readings were calculated from all chips in an exposure group without regard to chip position to obtain a single value for the exposure level and chip types. The results are shown in Tables 4 and 5. Linear regression analysis was performed on the mean adjusted readings in Table 4; the results are shown in Table 6. These results show excellent linearity for all phosphor types. The relative standard deviation in percent as a function of exposure is plotted in Figure 3. The standard deviation values in Table 5 and Figure 3 are for net responses. It should be noted that the TLD 500 and TLD 700H chips were calibrated using a ^{90}Sr card irradiator having close geometry. Because of the field gradient, and the nonpenetrating nature of beta radiation, small variations in chip positioning and chip thickness on the card can result in measurable random error in the ECC. This error is evident in the fact that the standard deviation for TLD 500 is on the order of 0.5% (Table 2) when irradiated with the ^{90}Sr irradiator used for ECC generation. The true variability for TLD 500 at exposures above 1 mR is probably less than 1%.

TABLE 4. Mean Adjusted Readings

Exposure Level, mR	TLD 200	TLD 700	TLD 500	TLD 700H
0.1	0.099	0.096	0.105	0.100
0.3	0.289	0.246	0.299	0.298
1.0	0.975	1.002	1.004	1.005
3.0	2.957	2.972	2.997	2.943
10.0	9.879	9.979	9.978	10.032
30.0	29.929	30.031	29.894	30.464
100.0	100.000	100.000	100.000	100.000

TABLE 5. Percent Coefficient of Variation for Adjusted Readings

Exposure Level, mR	TLD 200	TLD 700	TLD 500	TLD 700H
0.1	17.801	307.348	20.622	111.102
0.3	6.110	33.337	6.659	37.820
1.0	1.968	16.983	3.202	16.962
3.0	2.336	5.477	2.678	4.328
10.0	2.025	1.371	2.055	3.148
30.0	1.510	0.927	1.754	2.576
100.0	0.605	1.274	1.104	2.008

FIGURE 3. Variability at Low Dose for TLD 200, TLD 500,

TABLE 6. Phosphor Regression Data

	TLD 200	TLD 500	TLD 700	TLD 700H
Constant	-0.04468	-0.01515	-0.01586	0.050512
Standard Error of Y Est.	0.047256	0.043896	0.027233	0.194348
R ²	0.999999	0.999999	1	0.999977
Number of Observations	7	7	7	7
Degrees of Freedom	5	5	5	5
X Coefficients	1.000267	0.999888	1.000264	1.000617
Standard Error of Coefficient	0.000527	0.00049	0.000304	0.002169

Minimum Measurable Dose

The standard deviation of unexposed control dosimeter readings expressed in mR was used to calculate the minimum measurable dose (MMD) for each phosphor type. Using the relationship $MMD = 3\sigma_B$, where σ_B is the standard deviation of the zero dose reading, the MMDs were calculated for Table 7. For the anneal and readout protocols used, the sensitivity of each phosphor relative to 15-mil TLD 700 (3 mm x 3 mm x 0.4 mm) is shown in Table 7. These sensitivities are based on the phosphor response to ¹³⁷Cs gamma radiation.

TABLE 7. Sensitivity and Minimum Dose

Phosphor Type	Dimensions, mm	Reader Calibration Factor, nC/mR	Relative Sensitivity	Minimum Measurable Dose
TLD 200	3 x 3 x 0.9	8.0	40	0.051
TLD 700	3 x 3 x 0.9	0.45	2.25	0.645
TLD 700	5 x 0.5	9.8	50	0.063
TLD 700H	3 x 3 x 0.5	1.5	7.5	0.306

CONCLUSIONS

All phosphors exhibited excellent linearity down to 0.1 mR. All phosphors exhibited good reproducibility above 10 mR while TLD 200 and TLD 500 appear to be better choices for exposures below 10 mR (monthly or weekly monitoring). Only TLD 500 showed significant light-induced background or fading under laboratory lighting. Special lighting and handling will be necessary for use of this phosphor in a routine program. Yellow and red

lighting appear to offer adequate handling time for loading and unloading of dosimeter holders and TLD readers with the TLD 500 card.

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