Chapter 4 Appendix 10

INTERLABORATORY CALIBRATION USING NBS-IRRADIATED Mg_2SiO_4:Tb

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Several interlaboratory comparisons have been undertaken in conjunction with the Hiroshima-Nagasaki thermoluminescence (TL) study with the aim of validating techniques or methodologies. Unfortunately none of these intercomparisons has been totally successful in providing precise data on differences in source calibration between all of the laboratories. To accomplish this and to avoid bias introduced by variations in equipment or techniques it was decided at the 2nd TL workshop in November 1983, that a single laboratory would perform the TL analyses on samples irradiated with known doses by each of the other laboratories. These irradiations were to be performed using sources directly tied to each laboratory's primary calibration. Analyses of these samples would provide a relative comparison of calibrations between the laboratories. An absolute calibration would be obtained by analyzing identical portions of sample irradiated at the National Bureau of Standards (NBS).

Materials and Methods

Commercially available Mg_2SiO_4:Tb TLD phosphor (Kasei Optonics, Tokyo) with grain diameter ranging from 75 to 250 μm was annealed for one hour at 400 °C and distributed to each of the laboratories involved in this study. The laboratories were asked to irradiate the annealed sample with a dose of less than 100 rad and to specify the dose given in terms of the units normally reported by that laboratory. Each laboratory also received control phosphor dosed prior to shipment from the Utah laboratory with a nominal but precisely measured dose of ^{137}Cs gamma radiation. This sample was to be treated identically to the experimental sample.

The samples were then returned to the Utah laboratory and all analyzed on the same equipment. The control phosphor was also analyzed as a check on induced or reduced

luminescence. The measurements of the irradiated Mg_2SiO_4:Tb from each laboratory were to provide a relative indication of calibration by the laboratories. To provide an absolute calibration, Mg_2SiO_4:Tb identical to that distributed to the laboratories was sent to Dr. Ellett of the National Research Council for irradiation in a ^{60}Co gamma-ray beam at NBS. The vials and the procedures used for irradiation are described below. Exposures to the samples were 100 R and 50 R corresponding to average doses to the magnesium orthosilicate phosphor of 82.6 rad and 41.3 rad, respectively, ±2 % (estimated standard deviation). The dose to quartz would be nearly identical to these doses. The samples were returned to the University of Utah (UU) for analysis. Controls identical to those distributed to the other laboratories were included with the NBS-irradiated samples.

Sample Irradiation at NBS

The TL calibration samples were irradiated at the vertical beam ^{60}Co gamma-ray facility described by Ehrlich and Seltzer.[1] The source, source holder, collimater, and beam catcher at this facility were designed to minimize scattered radiation. Eighty-six percent of the energy fluence incident on the phosphor samples was due to unscattered photons. The energy distribution of scattered photons has been accurately measured[1] so that the dose per unit exposure can be calculated by standard methods. Exposures (in roentgens) were certified as accurate to ± 1%.

The phosphors were exposed within quartz spectrophotometric cells having inside dimensions 1 × 1 × 1.87 cm with a wall thickness great enough to insure electron equilibrium, 0.25 cm. Samples were irradiated individually at 147.89 cm source-to-sample distance and rotated 180° halfway through their period of exposure so that the dose distribution was more nearly uniform throughout the phosphor. Sample attenuation was estimated three ways:

1. Calculation of the kerma at the midplane of the sample taking into account the transient electron equilibrium in an attenuated photon beam as outlined by Roesch.[2]
2. An empirical approach based on the measured dose distribution in water from a ^{60}Co source as reported by Johns.[3]
3. Monte Carlo calculations at NBS by Charles Eisenhauer.

Each of these methods yielded slightly different estimates of sample attenuation, i.e., the ratio of the dose at the center of the sample to the dose without attenuation. The first method yielded 0.941, the second 0.953, and the third 0.965.* The uncertainty in each of these estimates is about 1%. An average attenuation factor of 0.95 was used to estimate the dose received by the Mg_2SiO_4:Tb phosphor. The overall accuracy of the dose received by these samples, considering both the uncertainty in the exposure and the calculated sample attenuation, is 2%.

TL Analysis

The TL analyses were performed using two Daybreak TL readers with glow ovens capable of evacuation and nitrogen backfilling. EMI9635QA photomultipliers (quartz windows) with

*Scaled from NBS Monte Carlo results for SiO_2

Corning 4-69 and 7-59 filters were in water-cooled (10 °C) enclosures. Because of the high intensity of the phosphor, neutral density filters were also used. On-plate irradiations were carried out with a 40-mCi ^{90}Sr source (Isotope Products, Burbank, California). The source was housed in a Daybreak Model 740 irradiator with solenoid-controlled shutter activated by a Grey Lab Model 625 timer.

Mg_2SiO_4:Tb samples averaging 0.5 mg were weighed before and after TL analysis with a Mettler AE163 balance (reproducibility to ± 0.01 mg). Samples were transferred to and from the heating plate with a vacuum pipette. Photon counts were collected and displayed at every 2 °C over the full heating range of the phosphor, 20 to 400 °C. Integrated counts from 160 to 200 °C were used for the analysis.

Grain Sizes

Grains in the size range of 70 to 250 μm were included in the samples distributed to the laboratories. This was done to allow those laboratories using beta-particle sources for irradiations to irradiate the Mg_2SiO_4:Tb samples on-plate if desired using grain sizes for which calibration had been made previously. This was not carried out, however, since all laboratories used gamma-ray sources for irradiation. Ranges of grain size used for TL analysis included 75 to 250, 106 to 150, and 150 to 250 μm. The former were analyzed first to preclude the possibility of spurious effects from seiving.

The Glow Curve

A Glow curve of a sample receiving repeated doses of approximately 120 rad is shown in Figure 1. An increase in sensitivity of the trailing edge of the peak is clearly seen. To minimize this effect, integrated counts taken from 160 to 200 °C were used for the dose estimation. Nonetheless, small (1 to 2%) increases in sensitivity were seen with repeated heating.

To verify that the increase in sensitivity following the first heating was proportional to that following the second heating the following test was made. Fifty milligrams of the sample was irradiated on-plate in batches of 0.5 mg each. The sample was stored a minimum of 24 hours at room temperature and measured on the same TL reader. Nineteen portions of sample were analyzed. Estimated dose obtained assuming a linear sensitivity increase between the first and second heatings was 59.8 ± 0.7* seconds. Assuming no sensitivity increase following the first heating, the measurement was 58.2 ± 0.6 seconds. No indication was seen of a 5% increase in sensitivity reported to occur several hours after irradiation.[4]

Measurement of Samples Irradiated at NBS

Results of analysis of the NBS-irradiated samples are shown in Table 1. These measurements were used to calibrate the source used for analysis of the remaining samples. A difference in dose rate was seen between the two TL readers used. The dose rate for reader 1 averaged 1.93 ± 0.05** rad/s for all grain sizes while that of reader 2 was 2.07 ± 0.05 rad/s. A difference in dose rate for grain sizes of 75 to 106 μm (1.97 ± 0.03* and 2.13 ± 0.061 rad/s for readers 1 and 2, respectively) was seen relative to those of 106 to 150 μm

*Standard error of the mean. **Standard deviation.

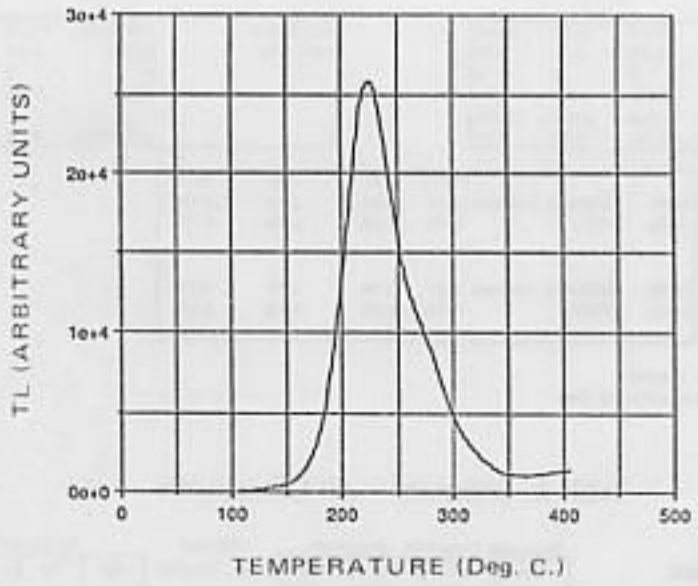

Figure 1. Glow curve of sample receiving repeated doses of approximately 120 rad

Table 1. Analysis of NBS-irradiated Samples

		†Extrp	1st	2nd	3rd
50R NBS	AVERAGE*	21.59	21.42	21.25	21.77
Reader #1	STDEV*	0.96	0.50	0.17	1.45
106-150µm	N	7	7	7	7
	S.E.M.	1.7%	0.9%	0.3%	2.5%
	NBS1/NBS3	0.490	0.496	0.502	0.521
	RADS/SEC	1.93	1.95	1.96	1.92
50R NBS	AVERAGE	22.49	22.23	21.96	21.81
Reader #1	STDEV	1.05	0.68	0.51	0.73
150-250µm	N	18	18	18	18
	S.E.M.	1.1%	0.7%	0.6%	0.8%
	NBS1/NBS3	0.522	0.529	0.535	0.538
	RADS/SEC	1.85	1.88	1.90	1.91
50R NBS	AVERAGE	20.88	20.58	20.30	20.21
Reader #1	STDEV	0.75	0.47	0.31	0.55
75-106µm	N	9	9	9	9
	S.E.M.	1.2%	0.8%	0.5%	0.9%
	NBS1/NBS3	0.485	0.494	0.504	0.503
	RADS/SEC	2.00	2.03	2.05	2.06
ALL RDR 1	AVERAGE	21.65	21.41	21.17	21.26
50R	STDEV	0.82	0.83	0.83	0.91
	N	3.00	3.00	3.00	3.00
	S.E.M.	2.2%	2.2%	2.3%	2.5%
	NBS1/NBS3	0.499	0.506	0.514	0.521
	RADS/SEC	1.93	1.95	1.97	1.94
50R NBS	AVERAGE	20.47	20.43	20.40	20.07
Reader #2	STDEV	1.31	0.72	1.04	1.25
106-150µm	N	19	19	19	19
	S.E.M.	1.5%	0.8%	1.2%	1.4%
	NBS1/NBS3	0.494	0.507	0.522	0.523
	RADS/SEC	2.04	2.04	2.05	2.08
50R	AVERAGE	20.51	20.62	20.73	20.77
Reader #2	STDEV	1.91	1.31	1.56	2.09
150-250µm	N	19	19	19	19
	S.E.M.	2.1%	1.5%	1.7%	2.3%
	NBS1/NBS3	0.508	0.521	0.535	0.536
	RADS/SEC	2.03	2.02	2.01	2.01
50R NBS	AVERAGE	19.92	20.25	20.59	20.35
Reader #2	STDEV	1.35	0.94	2.05	1.87
75-106µm	N	9	9	9	9
	S.E.M.	2.3%	1.5%	3.3%	3.1%
	NBS1/NBS3	0.515	0.530	0.545	0.533
	RADS/SEC	2.09	2.06	2.03	2.05

		†Extrp	1st	2nd	3rd
100R NBS	AVERAGE	44.07	43.20	42.32	41.82
Reader #1	STDEV	1.92	1.53	1.32	1.32
106-150µm	N	12	12	12	12
	S.E.M.	1.3%	1.0%	0.9%	0.9%
	RADS/SEC	1.89	1.93	1.97	1.99
100R NBS	AVERAGE	43.06	42.05	41.05	40.55
Reader #1	STDEV	1.51	1.19	1.09	1.40
150-250µm	N	19	19	19	19
	S.E.M.	0.8%	0.6%	0.5%	0.8%
	RADS/SEC	1.94	1.98	2.03	2.06
100R NBS	AVERAGE	42.98	41.63	40.28	40.18
Reader #1	STDEV	1.40	1.42	1.69	2.02
75-106µm	N	9	9	9	9
	S.E.M.	1.1%	1.1%	1.6%	1.9%
	RADS/SEC	1.94	2.00	2.07	2.08
ALL RDR 1	AVERAGE	43.37	42.29	41.22	40.85
100R NBS	STDEV	0.61	0.81	1.03	0.86
	N	3	3	3	3
	S.E.M.	0.8%	1.1%	1.4%	1.2%
	RADS/SEC	1.92	1.97	2.02	2.04
100R NBS	AVERAGE	41.47	40.28	39.09	38.34
Reader #2	STDEV	2.17	1.80	1.62	1.70
106-150µm	N	11	11	11	11
	S.E.M.	1.6%	1.3%	1.3%	1.3%
	RADS/SEC	2.01	2.07	2.13	2.18
100R NBS	AVERAGE	40.38	39.54	38.73	38.73
Reader #2	STDEV	2.61	2.38	2.42	2.65
150-250µm	N	21	21	21	21
	S.E.M.	1.5%	1.3%	1.4%	1.6%
	RADS/SEC	2.07	2.11	2.15	2.15
100R NBS	AVERAGE	38.64	38.22	37.79	38.18
Reader #2	STDEV	2.55	2.30	2.39	2.92
75-106µm	N	12	12	12	12
	S.E.M.	1.9%	1.7%	1.8%	2.2%
	RADS/SEC	2.16	2.18	2.21	2.19

→

Table 1. Continued

ALL RDR 2	AVERAGE	20.30	20.43	20.57	20.40
5GR NBS	STDEV	0.33	0.18	0.17	0.35
	N	3	3	3	3
	S.E.M.	0.9%	0.5%	0.5%	1.0%
	NBSI/NBS3	0.505	0.519	0.534	0.531
	RADS/SEC	2.06	2.04	2.03	2.04

ALL RDR 2	AVERAGE	40.18	39.35	38.54	38.42
10GR NBS	STDEV	1.42	1.05	0.67	0.29
	N	3	3	3	3
	S.E.M.	2.0%	1.5%	1.0%	0.4%
	RADS/SEC	2.08	2.12	2.16	2.17

		†Extrp	1st	2nd	3rd
RDR1	AVERAGE (rad/sec)	1.93	1.96	2.00	2.00
(n=6)	STDEV	2.6%	2.8%	3.3%	3.7%
RDR2	AVERAGE (rad/sec)	2.07	2.06	2.10	2.11
(n=6)	STDEV	2.6%	2.8%	3.8%	3.5%

* Seconds of Sr-90 beta irradiation (Src#3)
† change in sensitivity from 1st to 2nd calibrating dose

Table 2. Summary – All Laboratories

					Measured Dose (rad in quartz)				Applied	MEAS/APPL			
LAB	N	RDR	GRNSIZE		M0	M1	M2	M3	rad (quartz)	R0	R1	R2	R3
DUR	18	1	075-250		52.4	52.4	52.6	52.2					
DUR	14	2	075-250		51.0	51.8	52.9	52.4					
DUR	8	1	075-250		51.9	52.2	52.8	52.3					
DUR	22	2	075-250		52.6	53.0	53.6	56.7					
DUR	2	1	106-150		51.5	52.1	52.6	52.3					
DUR	4	2	106-150		52.7	52.2	51.4	52.9					
DUR	3	1	106-150		51.9	52.4	53.0	51.8					
DUR	10	2	106-150		51.1	53.0	54.6	54.3					
DUR	4	2	150-250		53.2	54.1	54.7	56.9					
DUR	8	1	150-250		52.2	51.9	51.7	50.7					
DUR	12	2	150-250		53.2	53.1	52.7	52.9					
				MEAN	52.2	52.6	53.0	53.2	47.8	1.09	1.10	1.11	1.11
				STDEV	0.8	0.7	1.0	2.0					
				N	11	11	11	11					
				STDEV%	1.5%	1.3%	2.0%	3.7%					
NRS	4	1	075-250		51.5	52.3	53.4	52.7					
NRS	14	2	075-250		51.3	51.8	52.2	52.4					
NRS	8	1	106-150		52.3	52.5	52.8	51.4					
NRS	14	2	106-150		51.6	52.6	53.2	54.8					
NRS	7	1	150-250		53.1	52.0	52.2	52.4					
NRS	7	2	150-250		51.5	50.9	51.3	51.9					
				MEAN	51.9	52.0	52.5	52.6	52.1	1.00	1.00	1.01	1.01
				STDEV	0.6	0.6	0.8	1.2					
				N	6	6	6	6					
				STDEV%	1.2%	1.2%	1.5%	2.3%					
NUE	21	1	075-250		83.1	83.1	83.4	82.7					
NUE	20	2	075-250		83.6	83.3	83.3	82.7					
NUE	12	1	106-150		86.3	85.8	85.2	83.7					
NUE	4	2	106-150		85.7	85.3	84.4	83.9					
NUE	12	1	150-250		87.9	87.5	87.1	86.5					
NUE	7	2	150-250		88.6	86.2	83.2	82.9					
				MEAN	85.9	85.2	84.4	83.7	87.2*	0.98	0.98	0.97	0.96
				STDEV	2.2	1.7	1.5	1.5					
				N	6	6	6	6					
				STDEV%	2.6%	2.0%	1.8%	1.7%					
OXF	4	1	106-150		81.7	81.5	81.2	80.3					
OXF	7	1	106-150		81.8	82.0	82.2	82.2					
OXF	3	2	106-150		82.1	80.5	78.5	77.0					
OXF	8	1	150-250		81.3	81.4	81.6	80.0					
OXF	11	1	150-250		81.7	81.9	82.1	81.0					
OXF	7	2	150-250		82.4	81.7	80.6	80.5					
				MEAN	81.8	81.5	81.0	80.2	77	1.06	1.06	1.05	1.04
				STDEV	0.4	0.5	1.4	1.7					
				N	6	6	6	6					
				STDEV%	0.5%	0.7%	1.7%	2.1%					
UOFU	17	1	075-150		34.5	34.8	35.3	35.2					
UOFU	10	2	075-150		35.0	35.3	35.7	35.1					
UOFU	11	1	075-250		35.8	35.8	36.0	35.4					
UOFU	4	1	106-150		34.8	34.8	34.9	34.6					
UOFU	7	2	106-150		33.9	34.4	34.6	35.6					
UOFU	19	1	150-250		35.2	35.4	35.6	35.3					
UOFU	12	1	150-250		34.7	35.3	35.7	35.8					
UOFU	10	2	150-250		35.5	35.6	35.5	35.5					
UOFU	14	2	150-250		35.4	36.9	38.2	39.5					
				MEAN	35.0	35.4	35.7	35.8	31.5	1.11	1.12	1.13	1.14
				STDEV	0.6	0.7	1.0	1.5					
				N	9	9	9	9					
				STDEV%	1.7%	2.0%	2.9%	4.1%					

* 60R + 35 rad (quartz)

grains (1.91 ± 0.04 and 2.02 ± 0.04 rad/s) and 150 to 250 μm grains (1.90 ± 0.03 and 2.05 ± 0.05 rad/s).

Measurement of Laboratory-dosed Samples

For the calculation of results of individual laboratory measurements, the dose rate corresponding to the appropriate grain size analyzed was used. For analysis of 75 to 250 μm sized grains the means quoted above were used (1.93 and 2.07), while for the 106 to 150 and 150 to 250 μm grains, the averages of the 106 to 150 and 150 to 250 μm dose-rate values were used (1.91. and 2.04). Results are shown in Table 2. The column M0 corresponds to the sensitization-corrected value and the remaining three values (M1 to M3) to those obtained by direct curve matching (three calibrating doses were applied to each sample). The column labeled "applied rad (quartz)" is the dose applied by each laboratory:

Durham, 57.6 R × 0.83 rad/R,* NIRS, 60 R × 0.87 rad/R,

NUE, 60 R × 0.87 rad/R plus 35.0 rad (the NUE control sample was mistakenly irradiated),

Oxford, 77 rad, and

UU, 40 rad × 0.79 rad/R.**

The UU irradiation used the same capsules used for the NBS irradiations and a ^{137}Cs source at a distance of 1 m. The values reported in Table 2 for UU include measurements of the control samples analyzed before and after shipment to the other laboratories.

Resulting calibration factors, the value by which a reported dose estimate in rad (in quartz) should be multiplied to correspond to the NBS calibration are: Durham, 1.10; NIRS, 1.00; NUE, 0.98; Oxford, 1.06; and UU, 1.11. The standard errors (one sigma) associated with these values for all laboratories other than NUE are approximately 2.5%. Those for NUE are approximately 5%.

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*Attenuation included **Standard Deviation