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ENERGY MEASUREMENTS

CLIMAX SPENT FUEL DOSIMETRY

SHORT TERM EXPOSURE, 8 MARCH 1983

by

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CONTENTS

TABLES

1. INTRODUCTION

This work describes the second short-term exposure (performed 8 Mar 1983) in Hole CEH3 at the Climax Spent Fuel Test site.[1] These short-term (1 hour long) exposures are intended to provide an independent measurement of the exposure rate at the wall and the 0.51-m and 0.66-m locations. The previous short-term exposure (done on 13 Aug 1982 and reported on 19 August 1982)[2] used MgB_4O_7 and CaF_2 thermoluminescent dosimeters (TLD's) cut to fit the usual stainless steel holders. The present exposure used only CaF_2 TLD's.

Several changes were made in the second short-term exposure procedures compared to the first. Listed below (in no particular order) are detailed descriptions of the various procedures, many of which were developed during discussions with R. Van Konynenburg preceding the experimental work.

1.1 DOSIMETERS

Use of the MgB_4O_7 TLD's, a low-Z detector, together with CaF_2 TLD's in the first short-term exposure[2] had shown little, if any, differences. This is taken to mean that the stainless steel holder provided a sufficiently uniform energy response for both TLD types. Calibration data at 60°C compared to 25°C data showed no changes that could be attributed to temperature for either TLD; thus either type was satisfactory. However, we found that continued work in the laboratory with the MgB_4O_7 TLD's yielded data of steadily decreasing quality. Eventually some of the chips suffered large changes in sensitivity and no ministrations were helpful. We therefore chose to use only CaF_2 TLD's in the second short-term exposure. Harshaw chips were cut to $0.32 \times 0.18 \times 0.09$ cm size and aged by several exposure/readout/bakeout cycles until all odd chips were weeded out and the remaining chips exhibited stable sensitivities.

These chips were sorted into sensitivity groups and assembled into the stainless steel holders, each of which had four chips. Individual chip numbers were assigned and each chip was followed throughout the experiment.

1.2 EXPOSURE

Exposure at Climax was done by removing the existing long-term dosimetry strings and inserting identical strings using the CaF_2 TLD's in the stainless steel holders. Timing was done with a stopwatch and was within ±10 seconds of 60 minutes. The stopwatches used were checked for accuracy by comparison to WWV for 16-hour periods the preceding week. This overall procedure was identical to that used for the first short-term exposure.

1.3 READOUT

Careful attention was paid to timing of the readout cycle, i.e., the chips were exposed at ~10 a.m. on 8 Mar 83 and the readout was started at

~10:30 a.m. on 9 Mar 83. A 1-h calibration exposure, closely matched to the total exposure and exposure rate just determined, was given on 10 Mar 83 at ~10 a.m. (following a bakeout the previous day). These chips were then read out on 11 Mar 83 at ~9:30 a.m., which resulted in a delay between exposure and readout comparable to that used for the field exposure.

The TLD reader used was a Harshaw Model 2000 A/B, which produces an integrated charge proportional to the thermoluminescence (TL) from each chip. A chart recorder output was also used to examine each TL peak. No anomolous behavior was found.

The TLD reader was set to integrate between 150 and 350°C with a 10°C per second temperature ramp. The maximum temperature was limited at 395°C. A continuous N_2 purge was used. The heater strip was platinum. The heater temperature controller had been calibrated by use of a miniature Type E thermocouple, and the reader temperature set-points were verified to be within ±2°C. These set-points are not very critical however, since the main TL peak occurs between them and the upper point is positioned in the valley preceding the heater glow.

1.4 CALIBRATION EXPOSURES

The chips were calibrated by exposing them to ^{60}Co and ^{137}Cs gammas while they were heated to ~60°C in an aluminum heater/holder. This aluminum device was thick enough so that electron equilibrium was established for the in-air irradiation.

The exposure rate from each source was determined with one or more NBS traceable ionization (ion) chambers. These three-terminal, tissue-equivalent ion chambers are routinely used in our facility for this purpose. Their output in coulombs per unit time was measured with a Keithley Model 616 electrometer; its coulomb scales were calibrated with the aid of a set of Hi-Meg Victoreen resistors and an NBS traceable voltage source plus one of the stopwatches mentioned earlier. The Hi-Meg resistors are periodically calibrated at the LVAO Standards Laboratory and thus also have NBS traceability. This calibration chain has been shown to be within 1% of the NBS values (at 1 sigma). It should be noted however that the NBS exposure standards are themselves ±1½% as given by the Bureau. Thus we expect our overall technique to yield a ^{137}Cs or ^{60}Co exposure in air to ±5% (with some allowance for positioning error).

This so-called post-exposure calibration, a routine procedure in our facility, has the advantage that an individual chip's sensitivity is determined immediately after exposure, and hence a group or average sensitivity is neither needed nor determined. For this system to be viable the individual chips must be followed throughout the procedures. This is done by means of silicon rubber pads which hold 50 chips in numbered holes. This method has another advantage in that an individual chip's sensitivity history can be followed and thus any deviant behavior identified and corrective action taken (usually summary dismissal from the experiment).

The use of both ^{137}Cs and ^{60}Co sources for calibration is not routine. It was done for a significant number of the TLD's to determine any energy dependence over this energy range. None was expected and none was found.

The calibration data in coulombs per R exposure are intended of course to be used to convert the field data in coulombs to absorbed dose in LiF, the medium used for passive dosimetry in the long-term irradiations. This conversion must take into account 1) the dosimeter material, CaF_2 in this case, 2) the calibration environment, an aluminum holder, and 3) the field environment, a stainless steel holder. Lastly, the dose deposited in a LiF dosimeter must be calculated from the measured dose deposited in the CaF_2 TLD. This process is documented in the Appendix and yields:

$$\text{Rads - LiF} = \left(\frac{\text{coulombs from SS exposure}}{\text{coulombs from A}\ell\text{ exposure}}\right) (0.820) \left(R_{\text{calibration}}\right)$$

where the SS exposure is that found in the field experiment, and the aluminum exposure is that found in the laboratory exposure to $R_{\text{calibration}}$ roentgen. For ^{137}Cs, this constant is 0.818.

1.5 LINEARITY CORRECTIONS

The first short-term exposure produced absorbed doses as high as ~6000 rads-LiF. The linearity corrections determined for the CaF_2 TLD's at these exposure levels were ~12%. At the exposure levels encountered in this experiment (3000 rads-LiF), this correction at worst is ~7% if the correction is 0 at 1000 rads-LiF and the deviation is linear. It is known however that the deviation is not linear but rather a complex function dependent upon the TLD material, the readout equipment, and other factors. In a letter report[2] on the first short-term exposure data, we showed a few data points to illustrate the general trends in the absorbed dose region of interest. By chance, one of the CaF_2 data points was very near 3000 rads-LiF and it exhibited essentially no nonlinear behavior.

The present post-exposure calibration method used calibration doses very close to those encountered in the field. Thus nonlinearity corrections would be the same for each irradiation and would tend to cancel one another. This fact, together with the small correction indicated above at a comparable dose level, led us to use no nonlinearity corrections in the present data set.

2. DATA ACQUISITION

Table 1 presents field data from all the exposed CaF_2 TLD's together with their ^{60}Co calibration exposures. Table 2 contains the same data for dosimeters 1 through 12, but associated with ^{137}Cs calibration exposures.

Dosimeters were exposed for three separate one hour periods at the wall position (0.31 m radially outward from the spent-fuel centerline). They were also exposed simultaneously at positions 0.51 and 0.66 m radially outward from the spent-fuel centerline. Measurements of the wall position were repeated to establish system reproducibility.

The calibration exposures were arranged to provide conditions close to those encountered in the field exposure. Thus dosimeters 1 through 35, which were at the wall position, were held at 60°C and received 3850 R in one hour as a calibration exposure from ^{60}Co. After readout, dosimeters 1 through 12 were exposed to 3900 R in one hour using a ^{137}Cs source, which yielded the data given in Table 2. Dosimeters 37 through 48 were given a 96.2-R ^{60}Co exposure in one hour, and dosimeters 49 through 60 were given a 9.61-R ^{60}Co exposure in one hour, for their respective calibrations. These latter two exposures were also done at 60°C. All dosimeters seemed to function properly. Note that the TLD reader had an offset that varied with the coulombs range in use. This offset (given in the Table 1 notes) must be subtracted from each reading, and is due to a combination of photomultiplier dark current not completely bucked out and a small amount ($\sim$0.0015 microcoulombs, or μC) of heater glow.

Table 1. Short-term TLD exposure (one hour) in Climax Hole CEH3, ^{60}Co calibration at 60°C

Hole Number, Dosimeter Location	Vertical Distance From Midplane (m)	Dosimeter Number	Raw Field Data (μC)* †	^{60}Co Calibration (μC)† ††	Rads-LiF** ‡
				Exposure 3850 R	
First Exposure	+1.22	9	30.00	23.99	
CEH3 (wall)		10	36.61	28.91	3904 ±96
		11	38.27	31.02	
		12	33.05	27.65	
	0	5	47.03	34.41	
		6	39.62	29.25	4236 ±79
		7	37.80	28.31	
		8	40.09	30.61	
	-1.22	1	48.08	33.05	
		2	50.47	35.53	4481 ±81
		3	45.47	32.41	
		4	48.11	34.41	
Second Exposure	+1.22	21	38.86	32.55	
CEH3 (wall)		22	37.92	31.38	3735 ±86
		23	39.11	34.17	
		24	38.92	32.84	
	0	17	45.74	32.80	
		18	42.85	30.17	4413 ±49
		19	49.23	35.43	
		20	45.60	32.90	
	-1.22	13	49.20	36.06	
		14	45.76	33.37	4246 ±222
		15	37.55	29.67	
		16	45.20	34.29	
Third Exposure	+1.22	33	33.84	29.48	
CEH3 (wall)		34	39.80	33.36	3649 ±98
		35	41.18	36.73	
		36	31.34	27.06	
	0	29	42.61	31.14	
		30	42.40	30.75	4266 ±84
		31	47.73	35.82	
		32	41.92	31.64	
	-1.22	25	48.16	35.40	
		26	37.89	27.85	4262 ±42
		27	38.26	28.44	
		28	46.06	34.55	

Table 1. (continued) Short-term TLD exposure (one hour) in Climax Hole CEH3, ^{60}Co calibration at 60°C

Hole Number, Dosimeter Location	Vertical Distance From Midplane (m)	Dosimeter Number	Raw Field Data (μC)* †	^{60}Co Calibration (μC)† ††	Rads-LiF** ‡
				Exposure 96.18 R	
CEH3 (0.51 m)	+1.22	45	0.820	0.610	104.1 ±1.7
		46	0.902	0.697	
		47	0.957	0.732	
		48	0.799	0.603	
	0	41	0.860	0.795	85.7 ±1.5
		42	0.661	0.602	
		43	0.748	0.678	
		44	0.702	0.661	
	-1.22	37	1.000	0.786	98.4 ±1.6
		38	0.726	0.581	
		39	0.720	0.587	
		40	0.755	0.612	
				Exposure 9.61 R	
CEH3 (0.66 m)	+1.22	57	0.1034	0.0911	9.44±0.42
		58	0.0939	0.0804	
		59	0.0658	0.0550	
		60	0.0674	0.0604	
	0	53	0.0472	0.0516	6.64±0.30
		54	0.0716	0.0806	
		55	0.0571	0.0682	
		56	0.0649	0.0745	
	-1.22	49	0.0683	0.0662	8.06±0.67
		50	0.0499	0.0524	
		51	0.0665	0.0604	
		52	0.0845	0.0852	

*Field exposure 8 Mar 1983; time of exposure 10:30 a.m.; exposure duration 60 minutes ±10 seconds.

**Plus or minus values are 1 sigma and refer to reproducibility only.

†Subtract TLD reader residual from all data: 0.018 microcoulombs (μC) from dosimeters 1 through 36; 0.005 μC from units 37 through 48; and 0.0015 μC from units 49 through 60.

††Calibration exposure rate and duration were varied to approximately match field exposure conditions.

‡Overall accuracy of absorbed dose data is expected to be within ±5%.

Table 2. Short-term TLD exposure (one hour) in Climax Hole CEH3, ^{137}Cs calibration at 60°C

Hole Number, Dosimeter Location	Vertical Distance From Midplane (m)	Dosimeter Number	Raw Field Data (μC)*	^{137}Cs Calibration 3900 R (μC)*	Rads-LiF
CEH3 (wall)	+1.22	9	30.00	23.05	4119 ± 47
		10	36.61	28.05	
		11	38.27	30.03	
		12	33.05	25.77	
	0	5	47.03	33.33	4455 ± 67
		6	39.02	27.85	
		7	37.90	26.92	
		8	40.09	29.36	
	-1.22	1	48.08	33.83	4677 ± 101
		2	50.47	34.43	
		3	45.47	30.54	
		4	48.11	32.35	

*Subtract TLD reader residual of 0.018 microcoulombs (μC) from each reading.

3. DISCUSSION

The average absorbed dose in rads-LiF found during this short-term exposure is given in Table 3. The wall data incorporate the three separate exposures, but they exclude the ^{137}Cs calibration since it is not a different exposure. Data calculated from page 14 of report No. UCRL-53159[3] are provided for comparison. This later data set was evaluated at 5.37 years out of core and represents the average over the central 2.44 m of the fuel.

The experimental data at the wall are similar to those obtained in the first short-term exposure in that a maximum is indicated somewhat below the center line. Both sets of data show an exposure rate change with distance above the center line that is approximately the same as the long-term exposure data. Below the center line both short-term exposure data sets are identical in shape to the exposure versus distance curve, but differ from the long-term exposure curve shapes at the wall position.

The exposure rates calculated from UCRL-53159 are lower than the measured values at the wall position, but are higher at the 0.51-m and 0.66-m positions. This may reflect a difference in the source linear activity distribution, or small changes in the absorption coefficients of the granite and other construction materials.

Table 3. Average exposure rates

Dosimeter Location	Vertical Distance From Midplane (m)	Rads-LiF per Hour	
		Present Data	From UCRL-53159
Wall	+1.22	3760 ± 130	3890
	0	4300 ± 96	
	-1.22	4330 ± 130	
0.51 m	+1.22	104.1 ± 1.7	111
	0	85.7 ± 1.5	
	-1.22	98.4 ± 1.6	
0.66 m	+1.22	9.44 ± 0.42	7.53
	0	6.64 ± 0.30	
	-1.22	8.06 ± 0.67	

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APPENDIX: CONVERSION OF RADS-CaF_2 TO RADS-LiF

A1. INTRODUCTION

The calculation of absorbed dose in various media when exposed to a gamma ray fluence is usually assisted by placement of an imaginary non-absorbing and non-perturbing small cavity in the medium. The calculated energy deposition in this cavity is then proportional to the kerma in the cavity wall, since the postulated conditions result in charged particle equilibrium in the cavity vicinity. Calculation of the energy deposited in this cavity is relatively simple and depends upon electron stopping powers of the cavity material (since the cavity is small compared to the electron range) and mass energy absorption coefficients of the wall material. Detailed descriptions of these methods are given in NBS Handbooks 78 and 79.[A1, A2]

In the case at hand however, the "cavity" used is a TLD which is not a non-absorbing detector in the usual Bragg-Gray sense treated in the NBS handbooks. The CaF_2 TLD's used were approximately $0.3 \times 0.2 \times 0.09$ cm. The maximum electron range in CaF_2 from electrons produced by 1.25-MeV gammas is approximately 420 mg/cm^2 or 0.13 cm. Hence the TLD severely attenuates any electrons produced in the wall of its holder or "medium." The attenuation is worse for a 0.5-MeV gamma exposure, which results in a maximum electron range of 0.03 cm in CaF_2. The TLD is thus not a Bragg-Gray cavity/medium situation, but more nearly one treated by Burlin.[A3]

Burlin's cavity theory enables calculation of the ratio, f, of the average dose in the cavity $\bar{D}_c$ (i.e., the CaF_2 TLD) to the wall kerma, assuming the incident gammas are not attenuated significantly in the wall or cavity. This was given by Burlin as

$$f = \frac{\bar{D}_c}{D_{med}} = d\,\overline{\left(\frac{(S/\rho)_c}{(S/\rho)_{med}}\right)} + (1-d)\left(\frac{(\mu_{en}/\rho)_c}{(\mu_{en}/\rho)_{med}}\right) \qquad \text{(A1)}$$

where Burlin assumed $K_{med} = D_{med}$. This latter assumption is true only where charged particle equilibrium exists. D_{med} is not the dose in the cavity wall because the absorbing cavity perturbs the charged particle equilibrium there. It should be interpreted as the dose in the medium at least one electron range away from the cavity.

The first term in Eq. (A1) is the mean value of the mass collision stopping power ratios averaged over the electron spectrum in the cavity. The second term is the ratio of mass energy absorption coefficients for the gamma rays near the cavity. The dimensionless weighting factor d depends upon the absorption characteristics of the cavity (i.e., TLD) for the electrons entering the cavity. When $d = 0$, the contribution of wall electrons to $\bar{D}_c$ is negligible.

An example of this is a cavity large compared to the electron range. This is nearly the case for the CaF_2 TLD's irradiated with 0.50-MeV gammas and is an adequate approximation for 1.25-MeV gammas. Ogunleye, Attix, and Paliwal[A4] recently examined this theory experimentally using LiF TLD's variety of holder materials from LiF to lead. Reasonable agreement (1 to 3%) between theory and experiment was found for polystyrene, aluminum, and copper holders. F. H. Attix[A5] suggested use of $d = 0$ for CaF_2 for the present case based on his experience with Burlin cavity theory. This implies that all the dose deposited in the CaF_2 TLD's is due to electrons generated within the TLD by the incident gamma rays. Presumably the incident electrons from the wall of the holder are balanced by an exiting fluence of electrons from the TLD.

A2. CALIBRATION EXPOSURE

The CaF_2 TLD's were irradiated in an aluminum holder with ^{60}Co gamma rays. The gamma exposure was measured with an NBS traceable ion chamber, resulting in R_{cal} roentgens. Each chip received the same exposure, but due to individual chip to chip variations the TLD readings obtained, coulombs (cal), were unique to each chip. Individual chip calibration data have been carried throughout the procedure.

The rads-CaF_2 will then be:

$$\text{rads-CaF}_2 = \left(R_{cal}\right) (0.877) \frac{\mu_{en}^{CaF_2}}{\mu_{en}^{air}} e^{-\mu_{A\ell} X_{A\ell}} \quad \text{(A2)}$$

where the (0.877) (R_{cal}) is the exposure in rads-air, and the various absorption coefficients[A6] are to be evaluated at 1.25 MeV. The aluminum holder thickness was 0.229 cm. Entering the numbers we have:

$$\text{rads-CaF}_2 = \left(R_{cal}\right) (0.877) \frac{0.0259}{0.0267} e^{-(0.150)\,(0.229)} = 0.820\ R_{cal}$$

For the ^{137}Cs calibration we have:

$$\text{rads-CaF}_2 = \left(R_{cal}\right) (0.851) \frac{0.02851}{0.0292} e^{-(0.0643)\,(0.229)} = 0.818\ R_{cal}$$

Since the coulombs generated during the readout of the TLD are proportional to rads-CaF_2 we find

$$\frac{\text{rads-CaF}_2}{\text{coulomb (cal)}} = \frac{0.820\ R_{cal}}{\text{coulomb (cal)}} \quad \text{(A3)}$$

as the calibration factor for each chip for ^{60}Co.

A3. FIELD EXPOSURE

The same chips were exposed in the field but in a stainless steel holder. The effective gamma energy was 0.5 MeV.[A7] Processing of the chips resulted in coulomb (field) from each chip. This was converted to rads-CaF_2 by means of the calibration factor given in Eq. (A3):

$$\text{rads-}CaF_2 = \text{coulomb (field)}\ \frac{(0.820)\left(R_{cal}\right)}{\text{coulomb (cal)}}$$

The field exposure was done in 0.0914-cm-thick stainless steel holders, and thus the exposure at the exterior of this holder but within the permanently installed dosimeter tubes is:

$$\begin{matrix}\text{rads-}CaF_2\text{ within} \\ \text{the dosimeter tubes}\end{matrix} = \frac{\text{coulomb (field)}}{\text{coulomb (cal)}}\ (0.820)\ \left(R_{cal}\right)\ e^{+\mu_{SS}\ X_{SS}} \qquad \text{(A4)}$$

where μ_{SS} is evaluated at 0.5 MeV and $X_{SS} = 0.0914$ cm.

A4. CaF_2 TO LiF CONVERSION

Burlin's cavity theory as interpreted by Ogunleye, et al., as explained in the Introduction, implies that only the ratios of absorption coefficients (evaluated at 0.5 MeV) would be needed to calculate the dose in LiF from a measured dose in CaF_2 because both TLD's are "thick" with respect to the electron range. This argument yields:

$$\begin{matrix}\text{rads-LiF within} \\ \text{the dosimeter tubes}\end{matrix} = (\text{rads-}CaF_2)\left(\frac{\mu_{en}^{LiF}}{\mu_{en}^{CaF_2}}\right)$$

Combining this equation with Eq. (A4) and inserting numbers we have

$$\begin{matrix}\text{rads-LiF within} \\ \text{the dosimeter tubes}\end{matrix} = \frac{\text{coulomb (field)}}{\text{coulomb (cal)}}\left(R_{cal}\right)\ (0.820)\ \left(e^{+(0.665)(0.0914)}\right)\left(\frac{0.0275}{0.0292}\right)$$

$$= \frac{\text{coulomb (field)}}{\text{coulomb (cal)}}\left(R_{cal}\right)\ (0.820) \qquad \text{(A5)}$$

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