UCRL--53341

DE83 003289

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Manuscript date: October 11, 1982

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Comparison of Measured and Calculated Radiation Doses in Granite Around Emplacement Holes in the Spent Fuel Test—Climax, Nevada Test Site

Abstract

Lawrence Livermore National Laboratory (LLNL) has emplaced eleven spent nuclear-reactor fuel assemblies in the Climax granite at the Nevada Test Site as part of the DOE Nevada Nuclear-Waste Storage Investigations. One of our objectives is to study radiation effects on the rock. The neutron and gamma-ray doses to the rock have been determined by MORSE-L Monte Carlo calculations and measurements using optical absorption and thermoluminescence dosimeters and metal foils. We compare the results to date. Generally, good agreement is found in the spatial and time dependence of the doses, but some of the absolute dose results appear to differ by more than the expected uncertainties. Although the agreement is judged to be adequate for radiation effects studies, suggestions for improving the precision of the calculations and measurements are made.

Introduction

In April-May, 1980, Lawrence Livermore National Laboratory (LLNL) emplaced eleven canisters of spent nuclear fuel from a commercial power reactor into holes drilled in an underground granite formation. This project, called the Spent Fuel Test—Climax (SFT-C),¹ is being carried out at the Nevada Test Site as part of the DOE Nevada Nuclear Waste Storage Investigations, managed by the Nevada Operations Office of the DOE. LLNL is responsible for technical direction of the test. The overall objective of this project is to evaluate the feasibility of safe and reliable short-term storage of spent reactor-fuel assemblies at a plausible repository depth in a typical granite rock, and to retrieve the fuel afterward.

As part of this project, we want to evaluate the effects of the radiation emitted by the spent fuel on the rock. Also of interest are the radiation effects on the temperature profiles, corrosion of the steel liners, and radiolysis of any water which is present.

A plan has been written to study these radiation effects which calls for thermoluminescence measurements and identifying changes in the rock's thermal, mechanical, and structural properties.² Preliminary work on rock thermoluminescence has been reported,³ as have mechanical properties measurements on the irradiated rock.⁴

A knowledge of the radiation doses in the rock of the SFT-C is essential for understanding the effects of radiation and for relating this work to other radiation effects studies. The doses are being determined in two ways: radiation transport calculations and *in situ* measurements. Results have now been reported from both approaches, and we compare them here.

Radiation Transport Calculations

Monte Carlo calculations⁵ using the MORSE-L code⁶ were carried out at LLNL, using source terms provided by the Hanford Engineering Development Laboratory (HEDL).⁷ These source terms had been calculated using the ORIGEN2 code,⁸ and were the same source terms as those used by HEDL in conjunction with the calorimetry they performed which provided the heat source term for the Climax thermal calculations.⁹

For the radiation transport calculations, only the four principal gamma-emitting radionuclides were considered. Neutron emission was assumed to have a spontaneous fission spectrum. The fuel-element geometry was modeled as shown in Fig. 1. We assumed the emplacement geometry to be perfectly concentric.

The results of the calculations showed the neutron dose, out to a radius of 0.76 m, to be negligible compared with the gamma-ray dose. Within the statistical uncertainty of the calculation, the dose was independent of vertical location over the central 2.44 m of the fuel assembly.

The absorbed dose-rate results, as a function of radius in the rock for the central 2.44 m of the fuel assembly, turned out to be essentially straight lines on semilog plots. Excellent agreement was found with measurements reported by HEDL for the dose rate at the surface of a spent fuel assembly in a hot cell.¹⁰ We found that less than 3% of the total heat output of the assemblies was deposited in the rock as gamma rays.

The dose rates were found to decrease with time as a result of radioactive decay, and to exhibit more negative slopes with radius as time progressed, presumably as a result of the softening of the spectrum as the shorter-lived emitters of higher energy gamma rays decayed more rapidly.

The highest dose rate calculated for the rock was at the surface of the emplacement hole at a time of 2.45 years, and amounted to 42.5 Gy/h in granite (4,250 rad/h in granite). The dose over a 5-year period at this location was calculated to be 2.2 MGy in granite (2.2×10^8 rad in granite). The dose on a granite basis came out about 8% higher than the dose on a LiF basis because of the differences in gamma-ray absorption cross sections. The overall uncertainty of the calculated results was estimated at $\pm 25\%$.

In Situ Dosimetry

Gamma-ray and neutron dosimetry at SFT-C site is being performed by E.G.&G., Inc., of Goleta, California.¹¹⁻¹³ This work has included optical-absorption measurements on ⁷LiF, thermoluminescence measurements on CaF₂ and MgB₄O₇, and neutron-activation measurements on metal foils and capsule parts.

The ⁷LiF dosimeters and neutron-activation foils are encapsulated and inserted into stainless-steel tubes located on the inside surface of the holes in the rock (radius = .30 m), as well as at radii of 0.51 and 0.66 m in the rock. Those to be considered here were left in position for periods ranging from about 7.5 to 9.5 months, and were then removed. The ⁷LiF chips were measured for optical absorption at four wavelengths, corresponding to peaks produced by its various color centers. Since the peaks grow sequentially as the absorbed dose is increased, the plan was to obtain continuous coverage over the relevant range of doses to be observed by using different peaks for different dose ranges.

Since it was anticipated that fading of the peaks would occur as the dosimeters were held at elevated temperatures over these long time periods, E.G.&G. experimenters performed a series of laboratory experiments which involved furnace anneals of previously room-temperature-irradiated dosimeters and periodic optical-absorption measurements. They hoped that this procedure would provide fade-correction factors which could be applied to the results from the *in situ* dosimeters once their temperatures had been measured. We recognized that laboratory experiments would not provide an exact simulation of the *in situ* conditions, since the laboratory dose was delivered beforehand at room temperature rather than continuously at rock temperature, but this was not thought to be important based on published information, which indicated the dosimeter response to be independent of irradiation temperature.¹⁴

However, since the initial *in situ* results were found to be self-inconsistent (i.e., different peaks indicated different doses for the same dosimeter), this assumption had to be re-examined. Laboratory experiments were then performed which indicated that the peak height for a given dose and color center was indeed sensitive to the irradiation temperature. This led to the laboratory generation of 60°C calibration curves and the elimination of all but two absorption peaks from consideration. Improvements to the spectrophotometry equipment made it possible to obtain continuous coverage over the relevant dose range with these two peaks, and it was hoped that use of the 60°C calibration curves, combined with the fade corrections determined earlier, would provide a means for compensating for the time-temperature history of the *in situ* dosimeters.

Figure 1. Approximate geometry of spent-fuel emplacement.

As another approach to circumventing the problems of time-temperature induced fade of the optical absorption dosimeters, we decided to do a short exposure of thermoluminescent dosimeters (TLDs). CaF_2 and MgB_2O_7 were selected, and chips were calibrated at 60°C over the dose range expected for a one-hour exposure. They were then exposed for 59–60 minutes at the SFT-C site and the thermoluminescence was measured. We anticipated that the shorter annealing times, as well as the lower observed temperature sensitivities of these dosimeters, would produce more reliable results.

The neutron-activation dosimetry involves measurement of four isotopes: ^{60}Co in cobalt foils, $^{110\text{m}}\text{Ag}$ in silver sealing rings, and ^{54}Mn and ^{58}Co in stainless-steel parts. Neutron activation does not suffer from temperature effects, but in this case, there were overall uncertainties of at least $\pm 30\%$ because of statistical uncertainties resulting from low counting rates and spectrum unfolding.

Comparison of Measurements and Calculations

At the time of writing, there were two sets of long-term ^7LiF optical-absorption data and two sets of neutron-activation data, representing the periods from April, 1980 to January 12, 1981 and from January 12, 1981 to October 23, 1981.^{11,12} There was also a set of short-term CaF_2 and MgB_2O_7 thermoluminescence data taken for the 59–60 minute irradiation on August 13, 1982.¹³ We compared these results with the calculations that had been performed earlier.

Since the calculated dose-rate results had not been integrated over the same time periods as those for which the measurements were made, it was necessary to perform the appropriate integrations before comparisons could be made. On the basis of the theoretical work on the temporal dependence of mixed fission product decay by Way and Wigner,¹⁵ as well as our empirical observation that the results for a single location formed essentially straight lines when plotted versus time on a log-log plot, we decided to fit the dose-rate results at each location to analytical functions of the form $D = D_0 t^{-n}$. The parameter, n , was found to have values between 1.03 and 1.67 for the various time periods and locations of interest. These functions were then integrated analytically over the time periods of interest.

Since the dosimeters were inserted into 0.5-mm (wall thickness) stainless-steel tubes which had not been modelled in the calculations, the additional gamma-ray absorption had to be estimated and was found to be about 1%. The measured data should be increased by this amount for comparison with the calculated results, but this correction has been ignored here because of its small size.

The comparisons between the measurements and the calculations are shown in Tables 1 through 3 for all locations where comparisons could be made. In addition, the comparative results as a function of radius are plotted for Hole CEH3 in Figs. 2 through 4. In these figures, the dashed lines indicate the $\pm 25\%$ -error limits estimated for the calculations. Error bars are not shown for the measured data because the uncertainties in these data are not yet well-known, in the opinion of the author, for the reasons described below. Only the data points for the central 2.44 m of the fuel-assembly length have been plotted, since the calculations indicated constant dose rates as a function of height only over this section. The data plotted in Fig. 4 were obtained by using straight lines connecting the two bordering calibration points, rather than the straight lines which the experimenters fitted to all the calibration data. This was done because the calibration data showed obvious curvature, and straight lines over the entire dose range did not appear to represent the data adequately.

Table 1. Comparison of calculated doses and doses measured by optical absorption for the first period (April 1980 to 12 January 1981).

Hole number, fuel assembly number, dosi- meter location	Vertical distance from midplane (m)	Measured absorbed dose (Gy in LiF)		Calculated absorbed dose (Gy in LiF)	Ratio Meas. dose Calc. dose
		247 nm	374 nm		
CEH1, D34, 0.30 m (wall)	+1.22		2.9×10^5	4.79×10^5	0.60
	0		3.2×10^5	4.79×10^5	0.67
	-1.22		2.9×10^5	4.79×10^5	0.60
CEH3, D40, 0.30 m (wall)	+1.22		3.2×10^5	5.08×10^5	0.63
	+0.61		3.4×10^5	5.08×10^5	0.67
	0		3.6×10^5	5.08×10^5	0.71
	-0.61		3.6×10^5	5.08×10^5	0.71
	-1.22		3.8×10^5	5.08×10^5	0.75
CEH3, D40, 0.51 m	+1.22	1.6×10^4	1.3×10^4	1.69×10^4	0.95, 0.77
	0	1.3×10^4		1.69×10^4	0.77
	-1.22	1.1×10^4	1.2×10^4	1.69×10^4	0.65, 0.71
CEH3, D40, 0.66 m	+1.22	1.0×10^3		1.40×10^3	0.71
	0	1.3×10^3		1.40×10^3	0.93
	-1.22	1.4×10^3		1.40×10^3	1.00
CEH7, D06, 0.30 m (wall)	+1.22		3.0×10^5	4.53×10^5	0.66
	0		3.0×10^5	4.53×10^5	0.66
	-1.22		3.7×10^5	4.53×10^5	0.82
CEH11, D04, 0.30 m (wall)	+1.22		2.8×10^5	4.24×10^5	0.66
	0		3.0×10^5	4.24×10^5	0.71
	-1.22		2.7×10^5	4.24×10^5	0.64

Table 2. Comparison of calculated doses and doses measured by optical absorption for the second period (12 January 1981 to 23 October 1981).

Hole number, fuel assembly number, dosi- meter location	Vertical distance from midplane (m)	Measured absorbed dose (Gy in LiF)		Calculated absorbed dose (Gy in LiF)	Ratio Meas. dose Calc. dose
		247 nm	374 nm		
CEH1, D34, 0.30 m (wall)	+1.22		2.8×10^5	4.06×10^5	0.69
	0		2.9×10^5	4.06×10^5	0.71
	-1.22		2.9×10^5	4.06×10^5	0.71
CEH3, D40, 0.30 m (wall)	+1.22		2.8×10^5	4.18×10^5	0.67
	+0.61		2.9×10^5	4.18×10^5	0.69
	0		3.1×10^5	4.18×10^5	0.74
	-0.61		3.2×10^5	4.18×10^5	0.76
	-1.22		3.1×10^5	4.18×10^5	0.74
CEH3, D40, 0.51 m	+1.22	1.6×10^4	1.2×10^4	1.31×10^4	1.22, 0.92
	0	1.5×10^4		1.31×10^4	1.14
	-1.22	1.4×10^4		1.31×10^4	1.07
CEH3 D40 0.66 m	+1.22	9.5×10^3		1.00×10^3	0.95
	0	1.0×10^3		1.00×10^3	1.00
	-1.22	7.5×10^2		1.00×10^3	0.75
CEH7, D06, 0.30 m (wall)	+1.22		2.9×10^5	4.16×10^5	0.70
	0		2.7×10^5	4.16×10^5	0.65
	-1.22		3.0×10^5	4.16×10^5	0.72
CEH11, D04 0.30 m (wall)	+1.22		2.7×10^5	4.18×10^5	0.64
	0		2.8×10^5	4.18×10^5	0.64
	-1.22		2.8×10^5	4.18×10^5	0.67

Table 3. Comparison of calculated dose rates and dose rates measured by TLDs for hole CEH3 on 13 August 1982.

Dosimeter location	Vertical distance from mid- plane (m)	MgB ₂ O ₇ data (Gy in LiF/h)	CaF ₂ data (Gy in LiF/h)	Calculated dose rates (Gy in LiF/h)	Ratios Meas. Calc.
0.30 m (wall)	+1.22	52.0	52.0	44.1	1.18, 1.18
		64.0	49.0	44.1	1.45, 1.11
	0	69.0	57.5	44.1	1.56, 1.30
		68.0	57.0	44.1	1.54, 1.29
	-1.22	64.0	70.0	44.1	1.45, 1.59
		64.0	64.5	44.1	1.45, 1.46
0.51 m	+1.22	1.28	1.16	1.29	0.99, 0.90
		1.19	1.23	1.29	0.92, 0.95
	0	1.10	1.02	1.29	0.85, 0.79
		1.06	1.05	1.29	0.82, 0.81
	-1.22	1.19	1.16	1.29	0.92, 0.90
		1.11	1.15	1.29	0.86, 0.89
0.66 m	+1.22	0.141	0.113	0.0901	1.56, 1.25
		0.131	0.132	0.0901	1.45, 1.46
	0	0.096	0.090	0.0901	1.06, 1.00
			0.087	0.0901	- 0.96
	-1.22	0.123	0.100	0.0901	1.36, 1.11
		0.123	0.104	0.0901	1.36, 1.15

Figure 2. Comparison of calculated and optical-absorption measured doses for Hole CEH3 for the first period from April 1980 to 12 January 1981. The measured data are from W. Quam and T. DeVore of E.G.&G. (see Table 1 for actual data points).

Figure 3. Comparison of calculated and optical-absorption measured doses for Hole CEH3 for the second period from 12 January to 23 October 1981. The measured data are from W. Quam and T. DeVore of E.G.&G. (see Table 2 for actual data points).

Figure 4. Comparison of calculated and thermoluminescence-measured doses for Hole CEH3 for Hole CEH3 on 13 August 1982. Data measured by Quam, DeVore, and Sever of E.G.&G. (see Table 3 for actual data points).

Discussion

Looking first at the optical-absorption data for the first irradiation period, (Table 1 and Fig. 2), we can see that the measured data are generally below the calculated doses, in many cases below the lower error limit. In the case of the data taken at the wall of the holes in the granite, using the 374 nm optical-absorption peak in ^7LiF , we suspect that the data are systematically low because the calibration curve used was for 25–30°C, rather than for 60°C. The experimenters were unable to extend the 60°C calibration curve to high enough doses with the calibration source used. We plan to extend this curve during FY 83. It appears, based upon extrapolation of the available 60°C calibration data, that this could bring the measurements at the wall into better agreement with the calculations.

The measurements at the 0.51- and 0.66-m radii, using the 247 nm optical-absorption peak in ^7LiF , are in better agreement, but some data points are still outside the error limit. These data also show considerable scatter. One is tempted to attribute the lack of agreement and the scatter to inhomogeneities in the rock or lack of alignment of the canisters with the holes, except that a comparison with the data in Tables 2 and 3 does not show a systematic pattern in the variations of dose and dose rate with location. It seems probable that at least some of the lack of agreement can be attributed to the fact that the measurements of the *in situ* doses were done with a Beckman 5270 spectrophotometer, while the calibration doses were measured with a mercury lamp and an IL500 photometer.¹² The use of two different instruments probably added some uncertainty to the results. (This is supported by the better agreement of the second period data, in which a single instrument was used.) In addition, the experimenters have attempted to separate the processes of color-center formation at the exposure temperature and color-center fading during the ensuing period that the dosimeter is held at elevated temperature, and make corrections for each. Unfortunately, this cannot be done in a clear-cut manner, since some fading undoubtedly takes place during the short-term calibration measurements.

Turning now to the optical-absorption data for the second irradiation period, the same explanations are offered for the differences between measurements and calculations as for the first period, with the exception that the better agreement at 0.51 and 0.66 m may have resulted partly from the use of a single instrument, as noted (Table 2 and Fig. 3).

Next, we examine the thermoluminescence dose-rate data for August 13, 1982, (Table 3 and Fig. 4). Since there do not appear to be significant differences between the MgB_4O_7 data and the CaF_2 data, they have been plotted together. The data at the 0.51-m radius is below the calculated dose rate but within the error limit, while the data at the wall position and at the 0.66-m radius are above the calculated values, have greater spread, and lie partially outside the error limit. It is difficult to find a ready explanation for these observations. The fact that the deviations of the wall data appear to be in the opposite direction from those of the optical-absorption wall data may not be significant, since the latter may follow suit when a 60°C calibration curve is used, as discussed above. Unaccountable temperature and dose-rate effects appear to be unlikely with the TLD data, since the temperatures and dose rates of the calibration and measurement runs were not very different. Fading errors should not be significant, since the waiting times and temperatures between exposure and readout were similar for the calibrations and the in-hole measurements.

It is interesting to note that the TLD data taken for the midplane of the fuel assembly at radii of 0.51 and 0.66 m appear to be systematically lower than the corresponding data for vertical locations of ± 1.22 m; this is not true for the optical-absorption data or for the calculations. There is no explanation for this at present.

Finally, considering the neutron data, the E.G.&G. measurements showed the neutron dose rate to be about five orders of magnitude less than the gamma dose rate at the wall of the hole. This is in general agreement with the calculations, which indicated that the neutron dose rates were negligible compared to the gamma dose rates. Precise calculations are not available for comparison.

Conclusions

In general, the spatial and time dependences of the measured and calculated dose rates are in good agreement. The absolute values in some cases are in good agreement and in other cases are outside of

expected error limits ($\pm 25\%$). We feel that the agreement between the optical-absorption results and the calculations might be improved if the 60°C calibration curve were extended into the relevant dose range. Agreement between the thermoluminescence measurements and the calculations might be improved by even more careful duplication of dose rate, dose, temperature, and waiting-time histories of the calibration and down-hole measurement runs.

This work demonstrates our ability to calculate and measure gamma-ray doses due to emplaced spent fuel in rock. The degree of agreement appears to be adequate for study of the radiation effects on the rock. Should more precise determinations be necessary, the most promising approaches would probably be better characterization of the time-temperature dose-rate behavior of the dosimeters, more precise measurement of the alignment of the fuel canisters and the emplacement holes, and additional calorimetric work and gamma-ray spectrometry to specify the source term more accurately.

References

1. L. D. Ramspott, L. B. Ballou, R. C. Carlson, D. N. Montan, T. R. Butkovich, J. E. Duncan, W. C. Patrick, D. G. Wilder, W. G. Brough, and M. C. Mayr, *Technical Concept for a Test of Geologic Storage of Spent Reactor Fuel in the Climax Granite, Nevada Test Site*, Lawrence Livermore National Laboratory, Livermore, CA, UCRL-52796 (1979).
2. R. Carlson, R. Van Konynenburg, W. Durham, and L. Page, *Plan for the Study of Radiation Effects on Climax Stock Quartz Monzonite Due to Emplaced Spent Nuclear Fuel*, Lawrence Livermore National Laboratory internal document, SFT 80-61 (1980).
3. R. Carlson, L. Page, L. Koons, and C. Sundbeck, *Preliminary Report on Radiation-Induced Thermoluminescence in Climax Stock Quartz Monzonite*, Lawrence Livermore National Laboratory, Livermore, CA, UCID-19229 (1981).
4. W. B. Durham, *The Effect of Gamma Irradiation on the Strength of Climax Stock Quartz Monzonite*, Lawrence Livermore National Laboratory, Livermore, CA, UCRL-87475 (1982).
5. T. P. Wilcox and R. A. Van Konynenburg, *Radiation Dose Calculations for Geologic Media Around Spent Fuel Emplacement Holes in the Climax Granite, Nevada Test Site*, Lawrence Livermore National Laboratory, Livermore, CA, UCRL-53159 (1981).
6. T. P. Wilcox, MORSE-L, *A Special Version of the Morse Program Designed to Solve Neutron, Gamma and Coupled Neutron-Gamma Penetration Problems*, Lawrence Livermore National Laboratory, Livermore, CA, UCID-16680 (1972).
7. F. Schmittroth, Hanford Engineering Development Laboratory, Richland, WA, private communication (October 2, 1980).
8. A. Croff, *A User's Manual for the ORIGEN2 Computer Code*, Oak Ridge National Laboratory, Oak Ridge, TN, ORNL-TM-7175 (1979).
9. F. Schmittroth, G. J. Neeley, and J. C. Krogness, *A Comparison of Measurements and Calculated Decay Heat for Spent Fuel Near 2.5 Years Cooling Time*, Hanford Engineering Development Laboratory, Richland, WA, TC-1759 (1980).
10. R. B. Davis, *Pre-Test Non-destructive Examination Data Summary Report on Turkey Point Spent Fuel Assemblies D01, D04, and D06 for the Climax-Spent Fuel Test*, Hanford Engineering Development Laboratory, Richland, WA TC-1693 (1980) and private communication with Sharon Atkins of Hanford Engineering Development Laboratory, Richland, WA (1981).
11. W. Quam and T. DeVore, *Climax Spent Fuel Dosimetry Progress Report: September, 1980-September, 1981*, E.G.&G. Inc., Goleta, CA, EGG 1183-2432 (UCRL-15419, Contract No. SANL 909-034, October, 1981).
12. W. Quam and T. DeVore, *Climax Spent Fuel Dosimetry Progress Report: September, 1981-September, 1982*, E.G.&G. Inc., Goleta, CA, EGG 1183-2455 (UCRL-15419-1, Contract No. SANL 909-034, August, 1982).
13. W. Quam, T. DeVore, and D. Sever, *Short-Term TLD Measurements at the Climax Spent Fuel Test Facility*, E.G.&G. Inc., Goleta, CA, Informal report (August, 1982).
14. W. L. McLaughlin, A. C. Lucas, B. M. Kapsar, and A. Miller, "Electron and Gamma-Ray Dosimetry Using Radiation-Induced Color Centers in LiF," *Radiat. Phys. Chem.* **14**, 467-480 (1979).
15. K. Way and E. P. Wigner, "The Rate of Decay of Fission Products," *Phys. Rev.* **73**, 1318-1330 (1947).