LAWRENCE LIVERMORE NATIONAL LABORATORY PLUTONIUM FACILITY PERSONAL NUCLEAR ACCIDENT DOSIMETER

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ABSTRACT

The LLNL Plutonium Facility contains significant quantities of fissile materials and thus must be able to provide dose estimates to effected individuals in the unlikely case of a nuclear criticality accident to meet the regulations set forth in the Code of Federal regulations, 10CFR835.1304, *Nuclear Accident Dosimetry*. In order to do this, the LLNL personal nuclear accident dosimeter (pNAD) design provides a robust capability for measuring fluence in four approximate energy ranges; thermal (0.025 eV); 1 eV – 1MeV; 1 – 3 MeV; and greater than 3 MeV. These ranges are investigated using metal foils (Au, Cu, and In) and a sulfur pellet for neutron activation analysis, and a Panasonic Thermoluminescent Dosimeter (TLD) for gamma dose measurement. Shielding materials are designed to remove thermal neutron contributions with minimal interference and chemical interactions with the detectors. Experiments were conducted at CEA-Valduc in October 2009 using the SILENE reactor and in September 2010 using the CALIBAN reactor to validate the dosimeter. The SILENE and CALIBAN reactors were operated in pulse mode providing brief power excursions simulating nuclear criticality accidents. Results from the experiments have demonstrated the LLNL pNAD design to be effective at evaluating the fluence dose in each of the four energy regions from this spectrum.

Key Words: Dosimetry, Accident Response, Foil Activation

1 INTRODUCTION

The US Code of Federal Regulations (10CFR835.1304[1]) requires facilities "possessing sufficient quantities of fissile material to potentially constitute a critical mass" to provide nuclear accident dosimetry. Additional guidance is given in the ANSI/HPS N13.3 *Dosimetry for Criticality Accidents*[2] which states that the dosimetry system should allow for accuracy "within ± 25 percent." The LLNL Plutonium Facility contains significant quantities of fissile mass and thus is obligated to provide neutron dose estimates to effected individuals in the unlikely case of a nuclear criticality accident.

Lawrence Livermore National Laboratory (LLNL) participated in two international nuclear accident dosimetry exercises hosted by CEA-Valduc in order to exercise the dosimetry system and confirm that the performance targets put forth in ANSI/HPS N13.3 are satisfied. In October 2009 the SILENE reactor was used in an experiment to represent a nuclear criticality accident and in September 2010 the experiment was repeated using the CALIBAN reactor.

2 LLNL PERSONNEL NUCLEAR ACCIDENT DOSIMETER

The LLNL personnel nuclear accident dosimeter (pNAD) consists of neutron activation elements which are placed around a Panasonic Thermoluminescent Dosimeter (TLD) as seen in Figure 1.

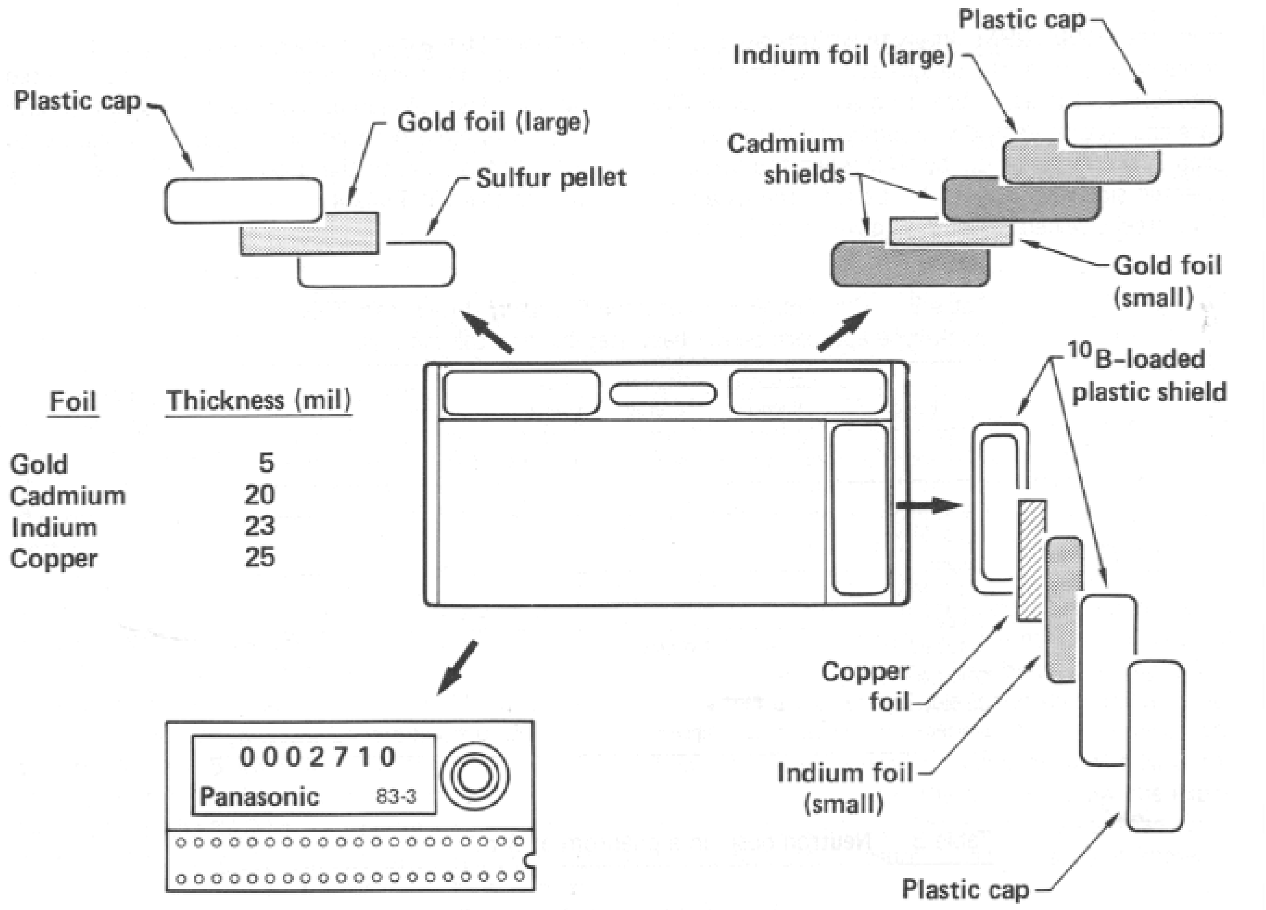

Figure 1. LLNL Personnel Nuclear Accident Dosimeter Design

The neutron activation elements consist of 5 metal foils (2 Au, 2 In, 1 Cu) and a sulfur pellet. Together with the appropriate shielding, the activation elements can be used to resolve the neutron spectrum through neutron activation analysis. The activation elements and corresponding energy regions are shown in Table I and explained in detail below.

Table I. Energy Range and Corresponding Activation Elements

Energy Range	Activation Elements
Thermal	Gold (unshielded) minus Gold (shielded)
1 eV to 1 MeV	Copper
1 MeV to 3 MeV	Indium minus Sulfur
> 3 MeV	Sulfur

The gold foils are used to determine the thermal neutron dose. The large gold foil is unshielded so it receives the full flux spectrum while the small gold is fully shielded by cadmium which removes the thermal portion of the flux. Using the difference of the two will yield the activation and thus fluence and dose of the thermal neutrons. The important reaction is the $^{197}\mathrm{Au}(n,\gamma)^{198}\mathrm{Au}$ gamma decay which generates a 0.411 MeV gamma ray. This gamma ray is produced 95.5% of the time the decay takes place.

The copper foil is used to determine the dose due to epithermal neutrons (1 eV to 1 MeV). Boron-10 shielding removes the thermal flux portion of the spectrum that reaches the copper and the small indium foil placed in front of it shifts any high energy neutrons into the epithermal range. The important reaction is the $^{63}\mathrm{Cu}(n,\gamma)^{64}\mathrm{Cu}$ gamma decay which generates a 0.511 MeV gamma ray. This photon is produced 35.8% of the time the decay takes place.

The small indium foil is used to determine the dose due to the lower energy region of fast neutrons (1 MeV to 3 MeV). It is also shielded by ^{10}B to remove the lower energy flux region. The important reaction is the $^{115}In(n,n')^{115m}In$ gamma decay which generates a 0.336 MeV gamma ray. This photon is produced 45.4% of the decays.

The sulfur pellet is used to determine the dose due to the higher energy region of fast neutrons (>3 MeV) and is not shielded at all. The important reaction is the $^{32}S(n,p)^{32}P$ beta decay which generates a 1.71 MeV beta.

The large indium is unshielded and is only used to determine personnel who were exposed to the radiation source. By quickly checking the activity level of the badge of each employee exiting the facility all people who were working in close proximity to the radiation source can be identified, questioned for useful information in emergency response, decontaminated, and placed under the proper medical care.

Using the measured activation in each of the elements, the neutron fluence can be calculated with constants and material properties such as: the time since irradiation, decay constant, atomic weight, Avogadro’s number, and cross section. Once the fluence is calculated a dose conversion factor is applied to each region and the total dose determined by summation.

Gamma doses are measured using the commercially available Panasonic TLD system which sits inside of the LLNL pNAD. The irradiated TLD are fed into a Panasonic reader which measures the gamma exposure seen by the TLD components and reports the results. Note that for these experiments the TLDs were not able to be read until the equipment was received back at LLNL.

3 EXPERIMENTAL SETUP

3.1 2009 Experiment using SILENE

The 2009 experiment was performed using the SILENE reactor at CEA-Valduc. The SILENE reactor is a solution (71 g/L of uranyl nitrate with 93% enriched uranium[3]) reactor which can be operated in three modes: pulse, free evolution, and steady state. The pulse mode results in a fission burst and spectrum that mimics what is created in a criticality accident. Due to this close approximation, the reactor was operated in pulse mode for the nuclear accident dosimetry experiments. The neutron spectrum of each pulse was changed by varying the yield and shielding the reactor core.

The LLNL pNADs were placed on phantoms in the reactor cell located at distances of 2 m, 4 m, and 6 m away from the reactor core. Three separate pulses were performed on the SILENE reactor.

3.2 2010 Experiment using CALIBAN

The 2010 experiment was performed using the CALIBAN reactor at CEA-Valduc. The CALIBAN reactor is an unreflected highly enriched uranium metal fast burst reactor which was operated in pulse mode to again represent a criticality accident. The CALIBAN core consists of ten fuel discs and 4 control rods composed of 93.5% enriched uranium metal alloyed with 10% molybdenum[4].

The LLNL pNADs were placed in the reactor cell at distances of 2 m, 3 m, and 4 m away from the reactor core. Two separate pulses were performed on the CALIBAN reactor.

3.3 Post Irradiation Experimental Procedure

Upon receipt of the dosimeters after irradiation by SILENE or CALIBAN they were disassembled into separate activation components and measured by the LLNL dosimetry team using gamma and beta detectors. The resulting measurements provided data to use in neutron activation analysis which allowed for the rapid determination of neutron dose. The gamma dose from the pNADs could not be calculated in the field as the Panasonic TLDs must be read by the Panasonic TLD reader which was located back at the home laboratory in Livermore, California. The gamma dose results were determined months after the reactor pulses when the equipment had been shipped back from France.

4 EXPERIMENTAL RESULTS

The results were calculated using the measured counts from the foil activation and the TLD readings. These results are compared to the given values from CEA Valduc to test the validity of the LLNL pNAD.

4.1 Neutron Dose Results

Table II shows the neutron dose results for both the 2009 and 2010 experiments.

Table II. LLNL Neutron Dose (Rad) Results[5,6]

Year	Pulse	Shield	Distance (m)	Valduc	LLNL
2009	1	Lead	2	690	791
			4	190	232
			6	110	109
	2	None	2	320	344
	3	None	6	150	159
2010	1	None	2	510	490
			3	260	290
			4	170	220
	2	None	2	720	707

To see which results fall inside of the ± 25% performance objective set by the ANSI/HPS N13.3 standard, the results will be compared to the given Valduc dose. Table III shows the ratios of the compared results where values that do meet the objective (outside of the range 0.75-1.25) are bolded.

Table III. LLNL Neutron Dose Ratios

Year	Pulse	Shield	Distance (m)	Valduc	LLNL
2009	1	Lead	2	1	1.15
			4	1	1.22
			6	1	0.99
	2	None	2	1	1.08
	3	None	6	1	1.06
2010	1	None	2	1	0.96
			3	1	1.12
			4	1	**1.29**
	2	None	2	1	0.98

4.2 Gamma Dose Results

Table IV shows the gamma dose results for both the 2009 and 2010 experiments.

Table IV. LLNL Gamma Dose (Rad) Results[5,6]

Year	Pulse	Shield	Distance (m)	Valduc	LLNL
2009	1	Lead	2	50	221
			4	30	46
			6	20	28
	2	None	2	380	432
	3	None	6	210	172
2010	1	None	2	70	64
			3	50	18
			4	40	18
	2	None	2	100	87

To see which results fall inside of the ± 25% performance objective set by the ANSI/HPS N13.3 standard, the results will be compared to the given Valduc dose. Table V shows the ratios of the compared results where values that do meet the objective (outside of the range 0.75-1.25) are bolded.

Table V. LLNL Gamma Dose Ratios

Year	Pulse	Shield	Distance (m)	Valduc	LLNL
2009	1	Lead	2	1	**4.42**
			4	1	**1.53**
			6	1	**1.40**
	2	None	2	1	1.14
	3	None	6	1	0.82
2010	1	None	2	1	0.91
			3	1	**0.36**
			4	1	**0.45**
	2	None	2	1	0.87

5 CONCLUSION

The CEA-Valduc exercises confirmed that the LLNL nuclear accident dosimetry system meets United States regulations and standards. The dosimetry team was able to rapidly disassemble and measure all dosimeters immediately following irradiation despite new personnel who had never participated in the procedure prior to the experiment.

As is seen in Section 4, only one of nine neutron dose predictions fell outside of the recommended window of ± 25%. The neutron dose is the dominant component in overall dose so there is an emphasis put on being able to accurately predict this component. The gamma dose measurements were not as successful as only four of nine gamma doses were predicted to within ± 25%. This inaccuracy demonstrates that more analysis and experimentation is necessary. However it is important to note the relatively small contribution of gamma radiation to overall dose. Since the neutron doses were predicted accurately, the overall dose estimates would still fall within the ± 25% objective in seven of 9 cases. Nonetheless future testing and research into improving the gamma dosimetry system is already underway.

6 ACKNOWLEDGMENTS

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