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PNNL Results from 2010 CALIBAN Criticality Accident Dosimeter Intercomparison Exercise

R. L. Hill
M. M. Conrady

October 2011

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Abstract

This document reports the results of testing of the Hanford Personnel Nuclear Accident Dosimeter (PNAD) during a criticality accident dosimeter intercomparison exercise at the CEA Valduc Center using the CALIBAN test reactor on September 20-23, 2010.

KEY WORDS: personnel nuclear accident dosimeter; PNAD; CALIBAN test Reactor; intercomparison.

Contents

1.0 INTRODUCTION

Pacific Northwest National Laboratory (PNNL) participated in a criticality accident dosimeter intercomparison exercise at the Commissariat a' Energie Atomique (CEA) Valduc Center near Dijon, France on September 20-23, 2010. The intercomparison exercise was funded by the U.S. Department of Energy, Nuclear Criticality Safety Program, with Lawrence Livermore National Laboratory as the lead Laboratory. PNNL was one of six invited DOE Laboratory participants. The other participating Laboratories were: Lawrence Livermore National Laboratory (LLNL), Los Alamos National Laboratory (LANL), Savannah River Site (SRS), the Y-12 National Security Complex at Oak Ridge, and Sandia National Laboratory (SNL).

The goals of PNNL's participation in the intercomparison exercise were to test and validate the procedures and algorithm currently used for the Hanford personnel nuclear accident dosimeters (PNADs) on the metallic reactor, CALIBAN, to test exposures to PNADs from the side and from behind a phantom, and to test PNADs that were taken from a historical batch of Hanford PNADs that had varying degrees of degradation of the bare indium foil. Similar testing of the PNADs was done on the Valduc SILENE test reactor in 2009 (Hill and Conrady, 2010). The CALIBAN results are reported here.

2.0 METHODS

2.1 PNNL Dosimeters Used

For this exercise, the Hanford PNAD was used and the results are reported here.

The Hanford PNAD is based on the outer dosimeter packet of the Hanford Fixed Nuclear Accident Dosimeter design and the PNAD used at the LANL (PNNL, 2010). The components of the Hanford PNAD are listed in Table 1 and illustrated in Figure 1-A and 1-B. Figure 1-A shows the Hanford PNAD that currently is in use. The PNAD shown in Figure 1-B shows the design used for the experiments at the Valduc center. The same specifications apply to the PNAD used in these experiments, except that, (1) the outer Plexiglas encasement was held together by nylon screws for easy disassembly instead of the glue used on the regular Hanford PNAD, (2), the sulfur pellet was put into heat-sealed plastic to facilitate easy handling when counting, as shown in Figure 1-B, and (3) two TLD-700 chips were added to this version of the PNAD instead of the one used in the currently used Hanford PNAD to improve counting statistics.

Table 1. Materials and Approximate Dimensions of the PNAD Components

Position in PNAD	Description	Diameter (cm)	Thickness (cm)[c]
1	Indium (Cd shielded)[a]	1.27	0.0127
2	Sulfur Pellet	1.27	0.292
3	Indium (Bare)	1.14	0.0127
4	Copper (Cd shielded)[a]	1.27	0.0127
--	TLD-700 Chip	0.32 x 0.32[b]	0.089

(a) The cadmium (Cd) shields covering the indium and copper foils are 0.051 cm thick.
(b) The TLD-700 chip measures 0.32 by 0.32 cm square by 0.089 cm thick.
(c) All dimensions are nominal.

A. **B.**

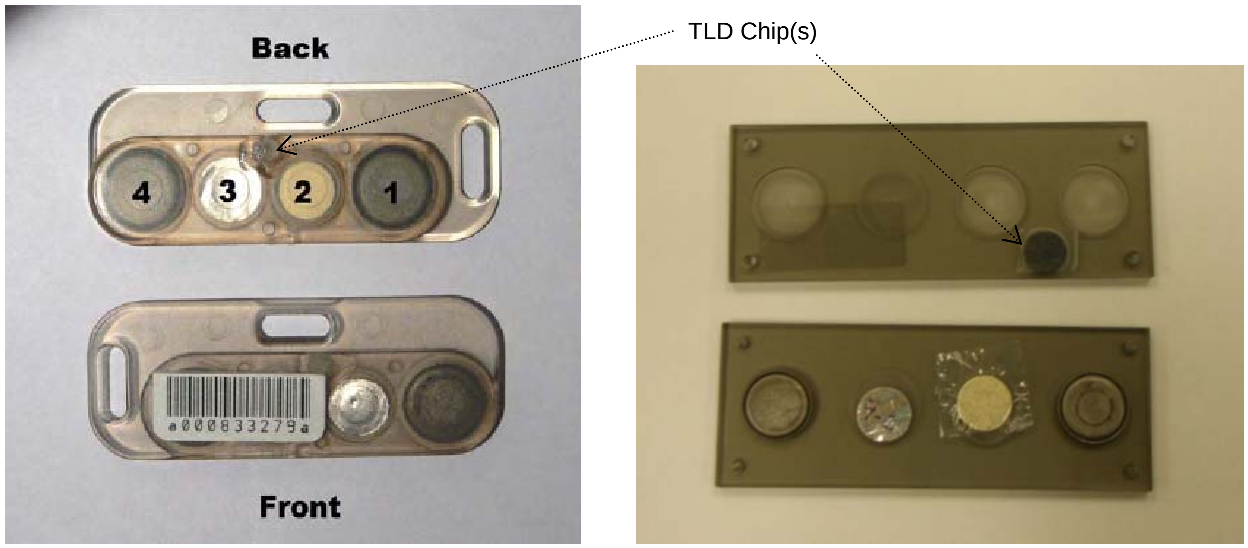

Figure 1. Hanford PNAD

2.2 CALIBAN Test Reactor at the Valduc Center

The CALIBAN test reactor is located at the CEA Valduc Center outside of Dijon, France. It is a metallic core cylinder with a diameter of 19.5 cm and height of 25 cm. It has a fixed block and moving block, each of which is made up of five metallic plates of molybdenum and highly enriched uranium. The axis of this reactor is vertical. The published neutron spectra for the CALIBAN reactor is shown in Figure 2.

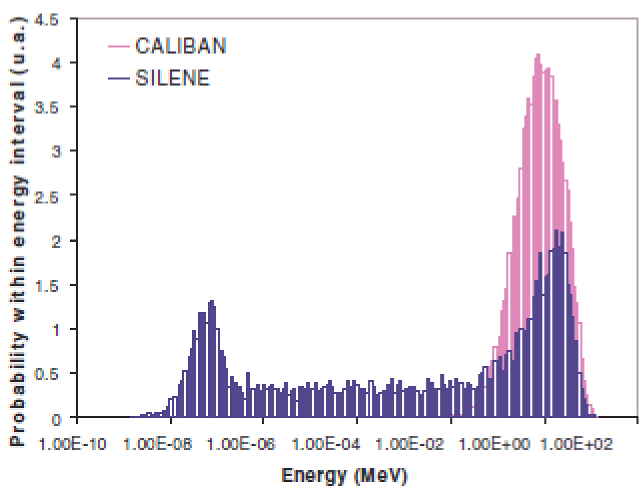

Figure 2. CALIBAN and SILENE Neutron Spectra[1]

[1] From Casoli, P. and N. Authier, Proceedings of the International Conference on Nuclear Data for for Science and Technology, April 22-27, 2007, Nice, France, editors O. Bersillon, et. al., EDP Sciences, 2008, pp 791-794.

2.3 Experimental Setup

2.3.1 Irradiations

The first experimental irradiation occurred on September 21, 2010 at 11:11:32 with the reactor unshielded for both irradiations.

For the first irradiation, a total of 18 PNADs were irradiated at 4 m from the reactor core at Positions 12 and 8 in Figure 3. Twelve PNADs were placed on a 10cm x 20cm x 30 cm polyethylene (PE) block at Position 12, with four on the front facing the reactor, two on the right side, two on the left side, and four on the back of the PE block. Six PNADs were placed in a plastic apron holder held in the air by a metal stand at Position 8. To test historical PNADs that were known to have various degrees of indium foil degradation, four PNADs having Excellent, Good, Fair, and Poor condition of the bare indium foil were disassembled and reloaded into the newer holder shown in Figure 2b. These PNADs were loaded in to apron holder at Position 8 along with two newly manufactured PNADs one of which was placed at a top position and the other at a bottom position on the apron holder.

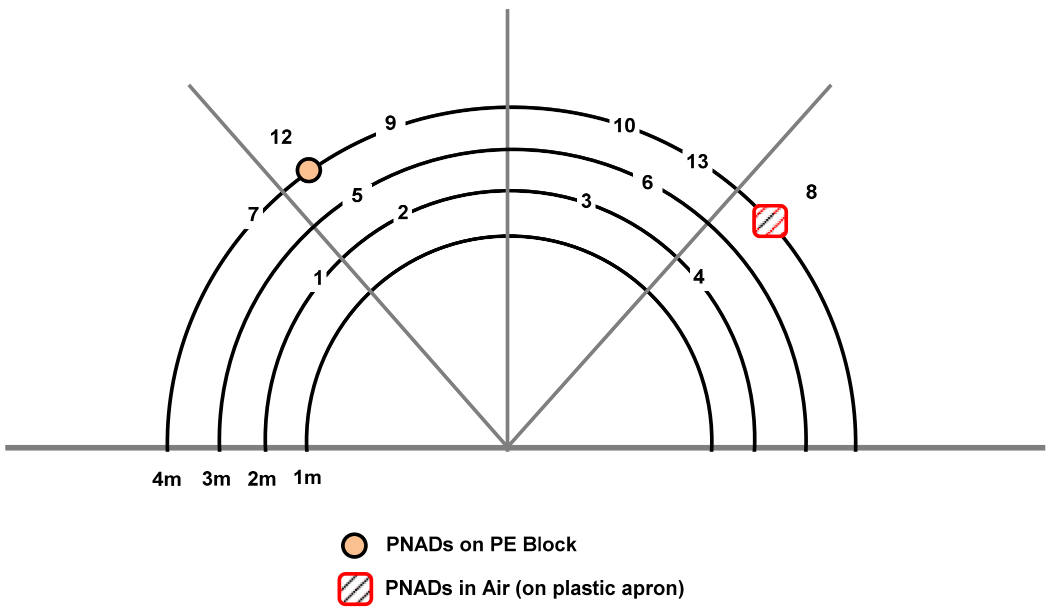

Figure 3. Map of PNAD Placement for Irradiation Experiment 1.

The second irradiation occurred at 11:13:02 on September 22, 2010. The PNADs number and placement for the second experiment were the same as for the first experiment with the addition of two additional PNADs. One PNAD each was added to the front and to the back of a sodium water phantom at Position 11, 2.5 m from the reactor core. The sodium (Na) phantom was 20 cm x 30 cm elliptical phantom filled with a sodium-water mixture on a stand about 80 cm above the floor. See Figure 4 for PNAD placement.

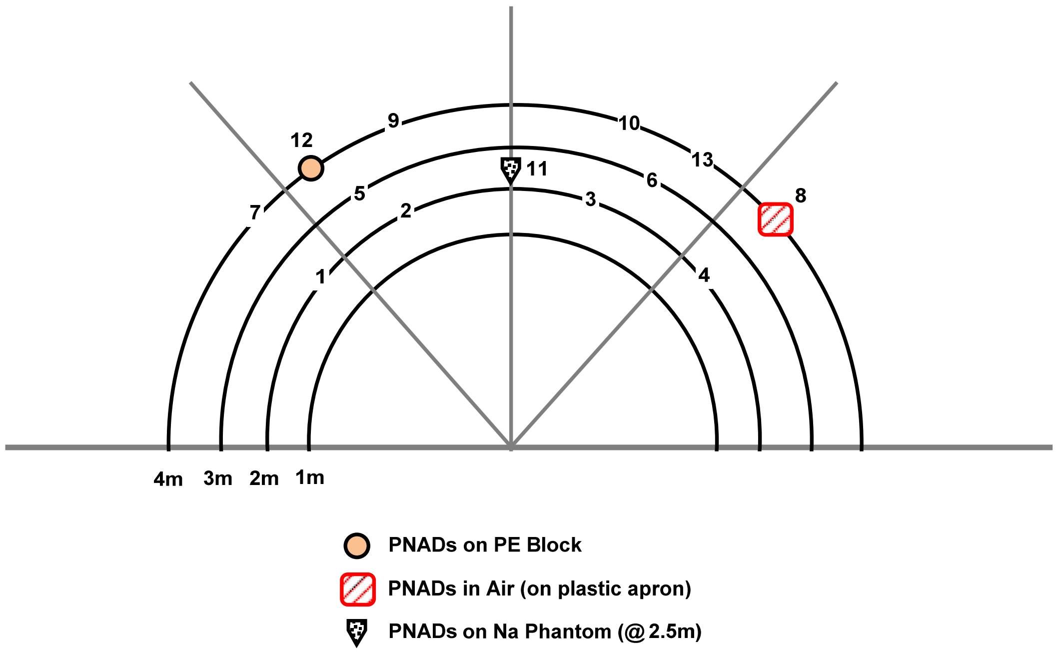

Figure 4. Map of PNAD Placement for Irradiation Experiment 2.

After each irradiation, the Valduc staff held the PNADs for 3-4 hours to ensure that the dose to the individuals handling them had decayed enough so that they would be within their organizational radiation limits for handling.

2.3.2 Counting Protocols

As the lead DOE Laboratory, LLNL offered to perform the counting of the foils and sulfur pellets. The indium and copper foils were measured using an electronically cooled high-purity germanium (HPGe) detector. The detector was calibrated for the average Hanford PNAD foil dimensions (Table 1) using the Canberra Industries ISOCS® technology. The indium foils were counted with a minimum of 2,000 counts in the 363 keV peak region and the copper foils were counted with a minimum of 500 counts in the 511 keV peak region. In some cases where total counting time was limited, the counting on some copper foils may have been terminated before 500 counts were reached.

For both Experiments 1 and 2, LLNL did not have adequate time to count all of the PNNL foil samples. SRS was able to count a large portion of the PNNL foils in which they used the same counting setup and geometry factors as LLNL. Because of time limitations in the foil counting phase of work, a number of the irradiated PNADs had to be eliminated from the counting scheme and, thus, do not have any data reported for them. See the data summary tables in Appendix A to see a listed of reported PNADs.

No capabilities were available at the Valduc Center for melting the sulfur pellets prior to counting. Therefore, the Hanford sulfur pellets were counted whole on a Canberra Industries iSolo alpha-beta counter by LLNL while still in the heat-sealed thin plastic. The LLNL iSolo instruments were calibrated using a 50 mm diameter standard beta source. , The counting geometry was adjusted to the 12.7 mm diameter of the PNNL sulfur pellets. The average mass of the Hanford sulfur pellets used was 0.451 g and were 99.9% pure sulfur. The counts were background corrected and used detector efficiencies of 25.98% for September 21, 2010 counting and 26.27% for counting on September 22, 2010. We were not able to ship the sulfur pellets back to PNNL. Therefore, the onsite count data of the whole pellet was all that was available for the analysis. A correction factor of 2 was used to estimate the count rate of crushed pellets (Hankins, 1969).

The TLD chips in the PNAD were used to obtain the dose from gammas, (D_{gamma}). The TLDs from the PNADs were brought back to PNNL for analysis. They were read on a Harshaw Model 5500 TLD reader using a standard process for PNAD TLD. Corrections for variations in individual chip sensitivity and for supralinearity were applied to the readings and the average of the two TLD chips from a given PNAD is reported.

2.4 Data Analysis

The data analysis approach used for the Hanford PNADs is discussed below. Half-life values used were taken from Nuclides and Isotopes 15th Edition, General Electric Co. and KAPL, Inc., 1996 (Table 2).

Table 2. Isotopic Half-lives Used

Isotope	Half-Life (units)	Half-Life (minutes)	Lambda
Cu-64	12.701 (h)	762.06	9.09E-04
S-35	87.2 (d)	125568	5.52E-06
P-32	14.29 (d)	2.06E+04	3.37E-05
In-116m	---	54.2	1.28E-02
In-115m	4.486 (h)	269.16	2.58E-03

The dose from neutrons, $D_{neutron}$, was calculated from the foil and sulfur pellet counting data. Table 3 provides the Hanford PNAD element information for the five neutron energy ranges evaluated.

$$C_X = \left(\frac{1}{\lambda n \sigma}\right)$$

where: C_X = abundance correction factor

λ = decay constant (min^{-1})

n = number of target atoms g^{-1} in foil/pellet

σ = activation cross section (barns).

Table 3. Activity to Fluence Factors for Hanford PNAD

Neutron Energy Range	Foil/Pellet Combination	Decay Constant λ (min^{-1})	Target Atom Abundance	Foil Atomic Weight (AMU)	Cross Section (barns)	C_X (min-g cm^{-2})
Thermal to 0.4 eV	In-116m (Bare –Cd)	1.28E-02	0.957	114.82	161	9.8E+01
Epithermal 0.4 eV to 2 eV	In-116m(Cd)	1.28E-02	0.957	114.82	2,600	6.00
2 eV to 0.5 MeV	Cu-64(Cd)	9.094E-04	0.692	63.54	0.341	4.92E+05
Above 1.2 MeV	In(Cd) (In-115m)	2.575E-03	0.957	114.82	0.188	4.11E+05
Above 2.9 MeV	Sulfur	3.368E-05	1.00	32.064	0.238	6.98E+06

2.4.1 Indium-116m Activity Determination

LLNL and SRS defined a pre-set number of photon energy peaks (Table 4) for determining the total activity of ^{116m}In. In the analysis report provided for every counted indium foil, each photopeak had an estimate of total activity for the sample associated with it. Any of the activity values provided for any one of the photopeaks could have been used for the total sample activity. PNNL chose to take each photopeak activity and normalize it to the total yield. A summation of these activity values was used to obtain the overall ^{116m}In activity for a given indium foil. This method was chosen to provide a better estimate of the total sample activity, based on a larger sample population.

Table 4. Photon Peaks Used for ^{116m}In

Photon Energy (keV)	Photon Yield (%)
416.86	27.7
818.72	11.5
1097.33	56.2
1293.56	84.4
1507.67	10.0
2112.31	15.5

$$Total\_Photon\_Yield = \sum_{i}^{n} Photon\_Yield_i$$

$$Normalized\_Activity_i = Activity_i * \left(\frac{Photon\_Yield_i}{Total\_\_Photon\_Yield} \right)$$

$$Total\_Activity = \sum_{i}^{n} Normalized\_Activity_i$$

2.4.2 Activity for Energy Group Above 1.2 MeV

For Experiments 1 and 2, the calculation for activity for the energy group above 1.2 MeV was calculated using data provided from photon data from bare indium foils using the results of the In-115m activity. For Experiment 2, there was a backlog of foils to be counted on both the LLNL and SRS counting systems, which lead to decay of some of the short-lived radionuclides before the foils could be counted. This was the case for several PNNL foils where no data was obtained for ^{115m}In using bare foils and/or cadmium-covered indium foils since this isotope had decayed before the foils were counted. As described in the PNNL procedure for processing PNAD results, it is acceptable to use ^{115m}In data from cadmium-covered indium foils as this energy is well above the neutron cutoff energy for cadmium when that data is available. Figure 7 shows the total neutron cross section for ^{113}Cd, which has the largest neutron cross section of all the stable cadmium isotopes. This figure shows that there is a very small cadmium cross section at energies around 1.2 MeV, thus allowing the use of either cadmium-covered indium foils or bare indium foils at this energy.

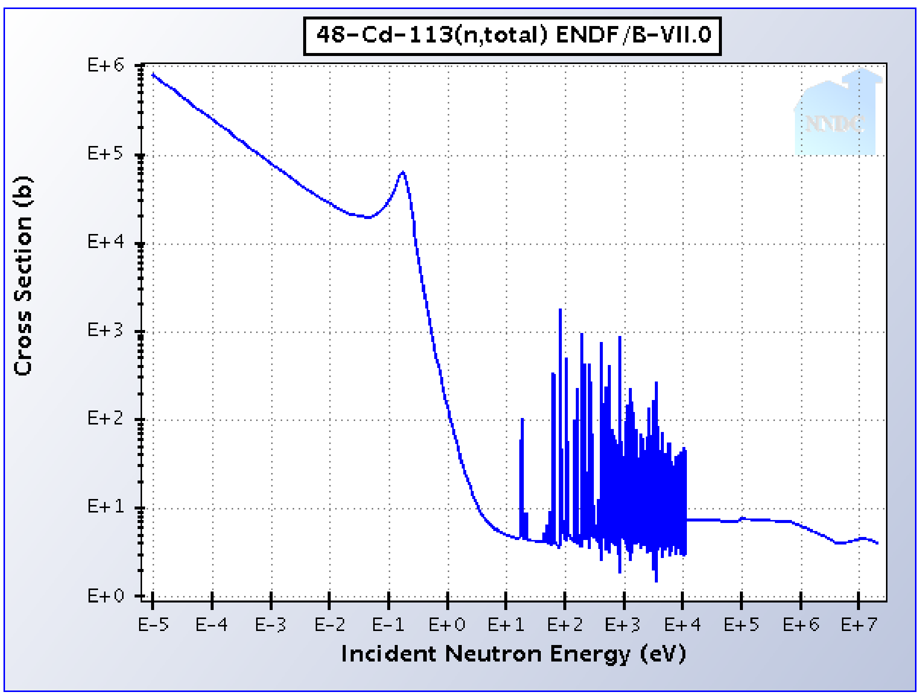

Figure 7. **Plot of Cd-113 (n, total) Cross Section**[2]

[2] Obtained from Sigma Plotting tool at National Nuclear Data Center, www.nndc.bnl.gov)

3.0 RESULTS

3.1 PNNL PNAD Results

The dose from neutrons ($D_{neutron}$) and gammas (D_{gamma}) was summed to get the total dose for each PNAD used in each of the two irradiation experiments. As mentioned above, not all exposed PNADs were able to be analyzed on site. The results of those PNADs that had all of their components analyzed are presented in Appendix A and summarized in Table 5.

Table 5. Summary of Estimated Doses and Number of Fissions for Hanford PNAD

Experiment	Setup Types (# PNADs)	$Dose_{neutron}$ (rad)	$Dose_{gamma}$ (rad)	$Dose_{Total}$ (rad)
1 at 4 m	Front of Small Phantom (3)	192	68	260
	In Air (1)	136	50	185
	Varying Indium Foils (4)	178	59	237
2 at 4 m	Small Phantom-Front (2)	164	93	257
	Phantom-Side (2)	95	93	189
	Phantom-Back (2)	92	109	201
	Varying Indium Foils (4)	150	74	224
2 at 2.5 m [(a)]	Sodium Phantom (1 in Front)	544	188	732
	Sodium Phantom (1 in Back)	36	108	143

(a) On Na phantom.

The neutron dose to gamma dose ratios for the PNNL PNADs are given in Table 6. The

Table 6. Preliminary PNAD Neutron Dose Related to Orientation

Experiment	Type or Orientation	Average n/g Ratio
1	Front of PE Block	2.5
	In Air	2.4
	Historic - Excellent	3.1
	Historic - Good	2.4
	Historic - Fair	3.3
	Historic - Poor	2.6
2	Front of PE Block	1.5
	Side of PE Block	0.82
	Back of PE Block	0.77
	Historic - Excellent	1.9
	Historic - Good	3.6
	Historic - Fair	0.96
	Historic - Poor	0.73
	Na Phantom - Front	2.5
	Na Phantom - Back	0.25

3.2 CALIBAN Reference Dose Results

The Valduc Staff provided a summary the CALIBAN reference dose results before we left the facility (Authier, 2010). The dose information that applied directly to the PNNL results is summarized in Table 7.

Table 7. Valduc CALIBAN Reference Neutron and Gamma Dose Results[(a)]

Experiment	Reported Reference Dose Information[(b)]			
	$Dose_{neutron}$ (rad)	$Dose_{gamma}$ (rad)	$Dose_{total}$ (rad)	n/g Ratio
1 @ 4 m	170	40	210	4.25
2 @ 4 m	240	60	300	4.00
2 @ 2.5 m	500	82	582	6.10

(a) Interpolated value.
(b) Valduc-supplied reference dose information for each experiment and distance from the CALIBAN reactor.

4.0 DISCUSSION

A total of 38 Hanford PNADs were tested in the two experimental irradiations at the CALIBAN reactor at the CEA Valduc Center. Of those 38 dosimeters, results were obtained for 20 exposed PNADs. PNNL relied on the LLNL and SRS teams to provide counting services of the foils and sulfur pellets. The foil counting systems for LLNL and SRS are essentially the same and used the same geometry factors[3].

Since water phantoms are generally not allowed in the CALIBAN exposure chamber, the phantom used was a block of polyethylene (PE). Dosimeters were placed on the front of the block facing the reactor, on the side, and on the back of the PE block that was placed 4 m from the reactor. For the first experimental irradiation, three PNADs that were on the front of the PE block were analyzed and the results are reported in Appendix A. For the second experimental irradiation, two PNADs each were analyzed which had been placed on the front, right side, and back of the PE block. Also for the second experiment, the Valduc staff was able to provide a sodium-water phantom at 2.5 m from the reactor upon which one PNAD each was placed on the front and back of the phantom.

The total dose measured on PNADs on the front of the PE block in Experiment 1 ranged from 3 to 43 percent over the Valduc reference doses reported. The in-air PNAD resulted in a total dose that was 12 percent below the reference dose at 4 m. The results of the Hanford PNADs on the PE block in Experiment 2 differed from the applicable total reference dose from -9 to -36 percent. The percentage difference was larger for those PNADs exposed on the sides and back of the PE block. The PNAD on the front of the Na phantom at 2.5 m from the reactor was 26 percent above the Valduc reference dose, while the PNAD on the back of the same phantom was 75 percent below the reference dose.

In addition, PNADs that had varying degrees of degradation of the bare indium foil from historic PNADs were also tested in air at 4 m from the reactor. The general breakdown of these PNADs

[3] See Table B.8 in LLNL Report (Hickman, et. al. 2011).

was given as excellent, good, fair, or poor. One of each type of PNADs was tested at 4 m from the reactors in both experimental irradiations. For Experiment 1 at 4 m from the reactor, these PNADs ranged from -5 to 34 percent of the Valduc reported total reference dose. For Experiment 2, also at 4 m, the total doses for the Hanford PNADs ranged from -50 to 3 percent of the reference dose.

The results from this intercomparison exercise using the CALIBAN reactor will be further studied to discern whether PNAD orientation corrections factors are needed to be put into place. In addition, the data for the PNADs made up of the constituents from historic PNADs that had varying degrees of degradation in the bare indium foil will be compared to other studies of this type of Hanford dosimeter.

5.0 ACKNOWLEDGEMENTS

The authors wish to thank Jerry McKamy and Jim Felty of the U.S. Department of Energy Nuclear Criticality Safety Program for financial support in order that we could participant in this intercomparison study. We wish to also thank Bruce Rathbone, Chandy Lindberg, and Randy Berg of PNNL for getting the test version of the PNAD designed and made to specifications in short order and providing analysis of the PNAD TLD chips; Dave Heinrichs, Dave Hickman, and Carolyn Wong of LLNL for setting up the DOE Laboratory participation in the Valduc exercises and providing a portion of the onsite counting of the foils and all of the sulfur pellets; to David Roberts of SRS for providing the remaining portion of the onsite counting of the foils; and to the CEA Valduc staff, without whose efforts and cooperation this work would not have been possible. Their staff were all most gracious and accommodating to our needs.

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APPENDIX A

INDIVIDUAL PNAD RESULTS

APPENDIX A. Individual PNAD Results

Table 7 contains the detailed results for each PNAD for each experimental irradiation. The average total doses were 910 rad, 834 rad, and 294 rad for the three experimental irradiations at the specified distance from the reactor.

Table A1. Experimental Irradiation #1 at 4 m

PNAD #	Description (a)	Thermal	Epithermal	2 eV to 0.5 MeV	> 1.2 MeV	>2.9 MeV	$Dose_{neutron}$ (rad)	$Dose_{gamma}$ (rad)	Total Dose (rad)	% Diff (b)
1	PE-12-F	1.2	0.0040	22	165	39	226	79	306	46
2	PE-12-F	1.3	0.0037	17	100	37	157	60	217	3
3	PE-12-F	0.039	0.017	19	136	36	192	66	257	23
15	A-8-B	0.66	0.0033	21	81	33	136	50	185	-12
16	A-8-EXCL	0.66	0.0032	18	150	27	195	58	252	20
17	A-8-GOOD	0.73	0.0025	19	111	25	156	59	215	2
18	A-8-FAIR	0.74	0.0022	17	114	25	156	44	199	-5
19	A-8-POOR	0	0.0098	17	158	32	207	74	281	34

(a) "PE-12-F" = On PE block, Position 12 around the reactor, front of the block
"A-8-B" = In Air, Position 8 around the reactor, bottom of the apron
EXCL = Excellent condition of the bare indium foil.
(b) "% DIFF" = Percent difference between PNNL measured dose and applicable Valduc reference dose.

Table A2. Experimental Irradiation #2 at 4 m and 2.5 m[(a)]

PNAD #	Description (b)	Thermal	Epithermal	2 eV to 0.5 MeV	> 1.2 MeV	>2.9 MeV	$Dose_{neutron}$ (rad)	$Dose_{gamma}$ (rad)	Total Dose (rad)	% Diff (c)
22	PE-12-F	1.8	0.0052	23	99	57	181	92	273	-9
23	PE-12-F	1.7	0.0054	20	74	50	146	94	240	-20
24	PE-12-RT	1.4	0.0043	20	0	44	65	93	158	-47
25	PE-12-RT	1.7	0.0005	2.5	90	31	126	93	219	-27
29	PE-12-B	1.1	0.0035	0	66	18	85	125	209	-30
30	PE-12-B	1.2	0.0034	20	56	21	99	94	192	-36
35	A-8-EXCL	1.0	0.0043	15	91	46	154	69	222	-26
36	A-8-GOOD	1.0	0.0043	27	174	44	246	63	309	3
37	A-8-FAIR	1.3	0.0002	21	0	69	91	60	151	-50
38	A-8-POOR	0.2	0.020	17	24	70	111	104	214	-29
13	Na-Front	2.1	0.0071	29	379	67	544	188	732	-50
34	Na-Back	1.2	0.0034	17	0	8.8	36	108	143	-29

(a) All irradiations were done at 4m from the reactor except the two PNADs on the Na phantom located at 2.5 m from the reactor.
“PE-12-F” = On PE block, Position 12 around the reactor, front of the block.
“A-8-B” = In Air, Position 8 around the reactor, bottom of the apron.
“Na-Front” = PNAD on front of the Na phantom located at 2.5 m from the reactor.

(b) EXCL = Excellent condition of the bare indium foil.

(c) “% DIFF” = Percent difference between PNNL total measured dose and applicable Valduc total reference dose.

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