PNNL-20239

Prepared for the U.S. Department of Energy
under Contract DE-AC05-76RL01830

Report on First Activations with the Lead Slowing Down Spectrometer

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March 2011

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Pacific Northwest National Laboratory
Richland, Washington 99352

Summary

On Feb. 17 and 18 2011, six items were irradiated with neutrons using the Lead Slowing Down Spectrometer (LSDS). After irradiation, dose measurements and gamma-spectrometry measurements were completed on all of the samples. No contamination was found on the samples, and all but one provided no dose. Gamma-spectroscopy measurements qualitatively agreed with expectations based on the materials, with the exception of silver. We observed activation in the room in general, mostly due to ^{56}Mn and ^{24}Na. Most of the activation was short lived, with half-lives on the scale of hours, except for ^{198}Au which has a half-life of 2.7 d.

Acronyms and Abbreviations

WSU	Washington State University
PNNL	Pacific Northwest National Laboratory
RPT	Radiation Protection Technician
HPGe	High-purity germanium
LSDS	Lead slowing down spectrometer/spectroscopy
BGO	Bismuth Germanate

Contents

1.0 Introduction

The research activities related to the Lead Slowing Down Spectrometer (LSDS) are not typically done at PNNL. As such, the project management has taken a step-wise approach to build a strong safety basis for these activities. The mid-term objective is to determine the correlation of time the neutrons enter the stack and their energy through a series of measurements. These measurements will be conducted by placing metal foils outside of a scintillator and measuring the time dependence of various (n,γ) processes. However, before these measurements can begin, we need to understand the potential radiological safety concerns generated by placing a variety of materials in the neutron field of the LSDS.

As a first step, we irradiated six items in the LSDS and then conducted dose and gamma-spectroscopy measurements to develop a better understanding of the risks. This report describes those activations and the ensuing characterization measurements.

2.0 Experimental Setup

The LSDS is a 1.5-m cube of lead that has been built to test concepts for use in direct measurements of Pu mass in spent fuel. A deuterium-tritium neutron generator is used to create 14-MeV neutrons inside the lead cube. The generator can produce up to 3×10^8 neutrons per second. One millimeter of cadmium is placed on the outside of the lead stack to absorb thermal neutrons, both from within the lead and those that escaped outside of the lead and then scatter back inside ("room return"). The generator is placed in the horizontal hole on the side of the stack, while the samples were put down the 4 1/8″ diameter vertical hole that is 45″ deep. A photo of the lead stack is shown in Figure 1.

The six irradiated items are summarized in Table 1. They are empty nylon bag and foils of silver, gold, iron, manganese and aluminum. The metal foils were placed inside of nylon bags to reduce removable contamination risks. The samples were lowered down the vertical hole of the LSDS using fishing line. All of the samples except gold were irradiated for an hour with DC beam with settings of 60 μA and 90 kV on the generator. The gold sample was irradiated with a pulsed beam with a pulse width of 20 μs and a repetition rate of 5 kHz. For the pulsed beam, the neutron room monitor read about 80% of the neutron rate as for DC beam.

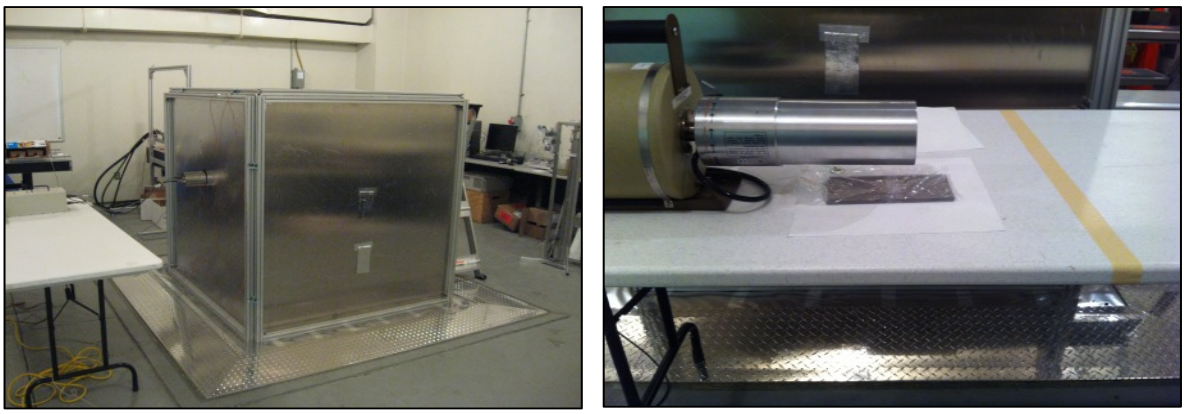

Figure 1: Photo of the lead stack with the DT generator in place (left) and photo of the HPGe detector measuring the manganese foil (right). Note that the table was moved between photos.

Table 1: Summary of irradiated samples. Weight includes the nylon bag, except for aluminum. ^{24}Na, with signatures at 1368.6 and 2754.0 keV with a half-life of 14.7 h, and ^{56}Mn, with signatures at 846.8, 1810.8 and 2113.2 keV with a half-life of 2.6 h, appear in most of the spectra. ^{56}Mn was expected to be observed from the iron foil; however it is difficult to separate the ^{56}Mn from the room versus from the foil with the measurements performed.

Sample	Weight (g)	Isotopes Observed	$T_{1/2}$	γ Signature (keV)
Nylon Bag	~3-6	none		
Silver	58.0	Al^{28} V^{52} I^{128}	2.24 m 3.74 m 25.0 m	1779.0 1434.1 442,9
Iron	385.9	Mn^{56}	2.6 h	846.8, 1810.8, 2113.2, …
Aluminum	145.9	none		
Manganese	793.5	Mn^{56}	2.6 h	846.8, 1810.8, 2113.2, …
Gold	8.35	Au^{198}	2.7 d	411.8

After irradiation, the samples were retrieved by a Radiation Protection Technician (RPT). The samples were swiped to test for removable contamination and also checked for gamma and beta dose and counts. No contamination was ever found. The count rates were below detection limits (1000 cpm) of the hand-held meters for all samples except for the manganese sample. The silver had 0.8 mR/h dose rate for betas, but no dose for gammas, when it was initially pulled out of the stack. Minutes later the dose from the silver was below minimum detectable activity. The RPT report is provided in Appendix A.

Two detectors were used for these measurements. A 130% relative efficiency HPGe detector was used to conduct the gamma spectroscopy measurements. The samples were placed on the table 4.5" below the center of the detector, except for the gold foil which was taped to the outside of the foil. The HPGe detector was removed from the vault while the neutron generator was running. The preamp of the HPGe was not healthy, resulting in dead times of 50% for background measurements. The resolution appeared to be reasonable, however. The BGO scintillator was operational while the neutrons were being generated.

3.0 Results

3.1 Room Background

Once the neutrons are generated in the room, many materials in the room can become activated. As a check on this possibility, background runs were completed before the neutron generator was turned on and after it had been run for an hour. The spectrum in Figure 2 shows strong Mn^{56} lines, which likely result from either the activation of iron or manganese. There are also lines at 239, 511 and 1238 keV that have not been identified. Manganese-56 has a half-life of 2.6 h, so that this incidental activation of the room will disappear over the period of roughly a day.

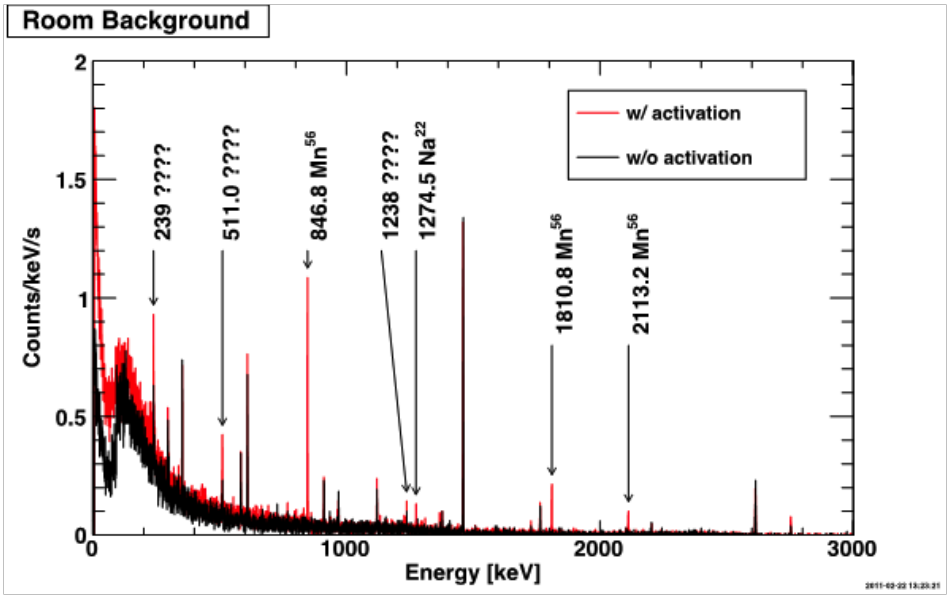

Figure 2: Comparison of room background before and after activation

3.2 Nylon Bag

The HPGe spectrum for the nylon bag was equivalent to the post-activation background spectrum.

3.3 Silver

The silver spectrum proved to be the most interesting as it has three gamma lines that cannot be attributed to expected isotopes. After an hour of activation, it was expected that the most active isotopes would be Ag^{110}, Ag^{108}, Ag^{110m} and Ag^{106} with half-lives of 24.6 s, 2.37 m, 250 d and 24.0, respectively. These isotopes are not strong gamma emitters, except for Ag^{110m} which has a 95% chance of emitting a 657.8-keV gamma with each decay, and Ag^{106} which has a 16.7% chance of emitting a 511.8-keV gamma ray with each decay. Neither of these were observed. The lines at 443, 1434 and 1779 keV were attributed to I^{128}, V^{52} and A^{l28} respectively. Each of these isotopes have their strongest gamma signature at their respective energies, and the relatively short half-lives (2.24 m, 3.74 m, 25.0 m, respectively), agree qualitatively with the observed time dependence of the gamma spectrum.

The silver isotopes are strong beta emitters, resulting in a slight increase in the continuum spectrum as seen in Figure 3. This could result from bremsstrahlung photons created by the emitted betas.

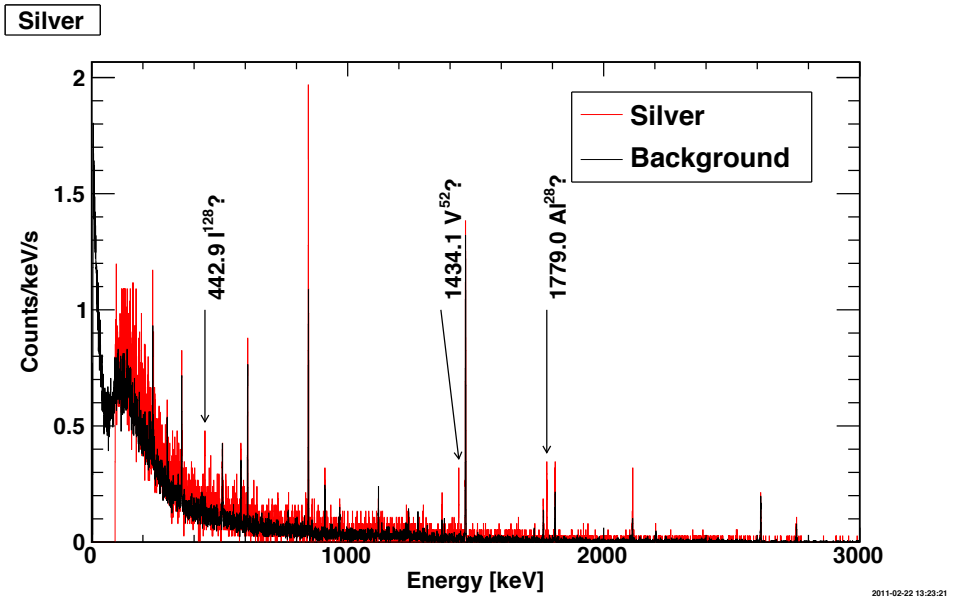

Figure 3: HPGe spectrum from silver plate

3.4 Iron

The iron spectrum, shown in Figure 4, is dominated by contributions from Mn^{56}. These peaks also appear in the activated background, so that the increased heights of the Mn^{56} could be a result of either Mn^{56} activation in the iron plate or from Mn^{56} activation from the material in the room. Measurements were not taken that would enable us to distinguish the two possibilities.

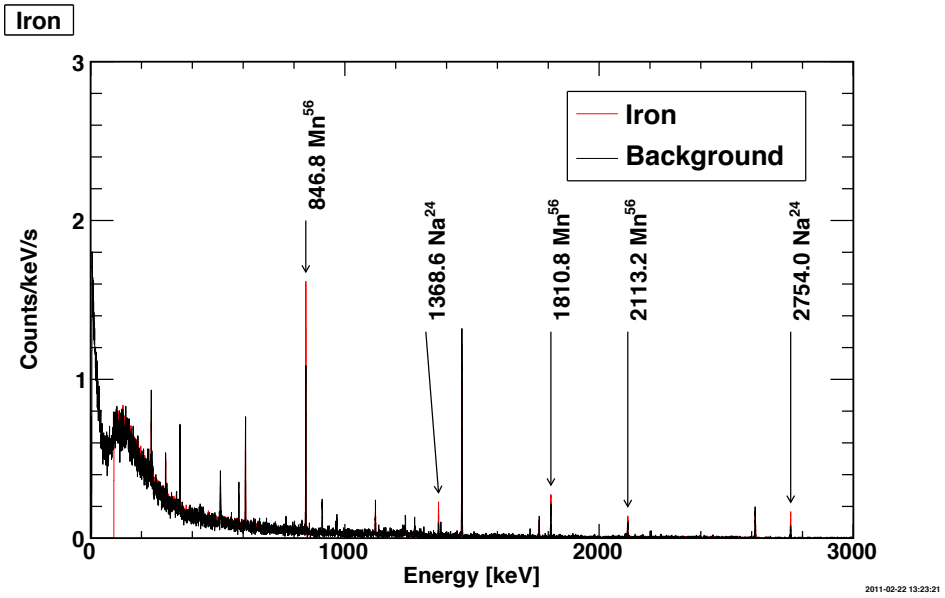

Figure 4: HPGe spectrum from iron plate

3.5 Aluminum

The aluminum piece was a support piece taken from the target ladder assembly constructed by the WSU mechanical engineering students. The HPGe spectrum is shown in Figure 5. No lines are observed besides 1368.6 and 2754.0 keV from Na^{24} (14.7 h half-life). The Na^{24} lines probably originate from a bank of NaI detectors stored near the HPGe detector and metal plates. These lines grow steadily stronger as more activations are completed. The most likely activation related line to observe would have been the

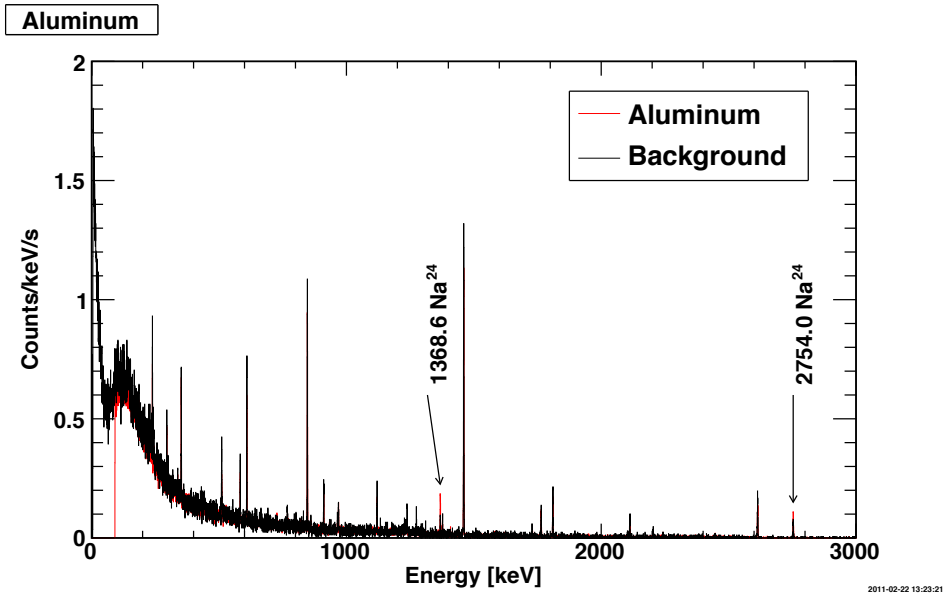

Figure 5: HPGe spectrum of Al plate.

1779.0 line from Al^{28} (2.24-m half-life). There is a hint of this line in the first spectrum taken on the aluminum plate, but it is not present in the longer aluminum measurement, which is shown in Figure 5.

3.6 Manganese

The manganese was the most active of the plates after the irradiation. The spectrum is shown in Figure 6. Most of the lines are attributable to ^{56}Mn. However, lines at 1091, 1300 and 1602 keV could not be easily attributed to an isotope. The half-life of each of these lines varies. The 1091-keV peak is not in the initial manganese spectrum, but appears in the later one suggesting that the peak is a result of an isotope growing in. The 1300-keV line is very long-lived, showing essentially no decrease over the 1 hour over which the two spectrum were taken. The 1602-keV line drops to about 60% of the original rate over the two measurements separated by roughly an hour, suggesting that the half-life is approximately 1-3 h.

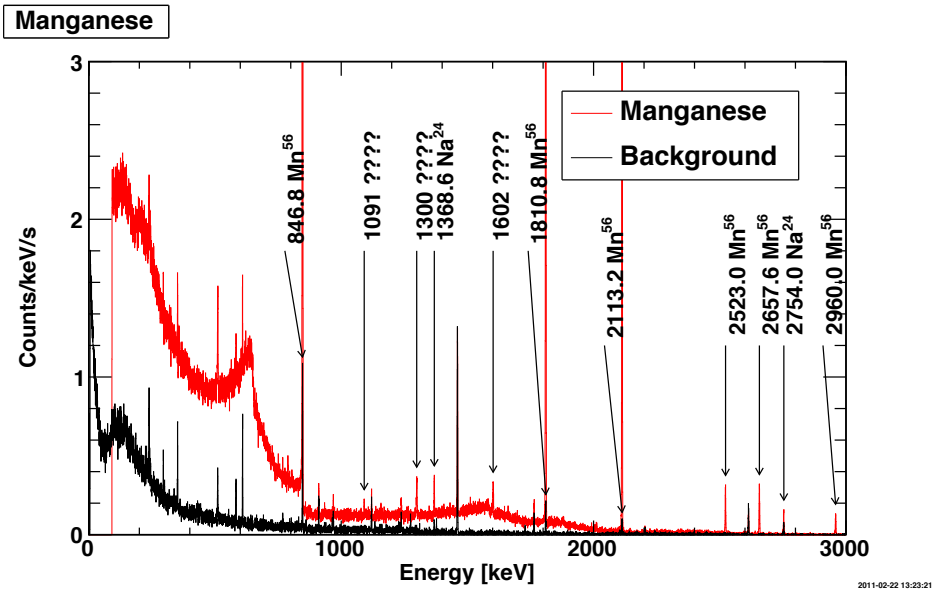

Figure 6: HPGe spectrum of activated manganese. Note that the peaks for 846.8, 1810.8 and 2113.2 keV are significantly off scale.

3.7 Gold

The gold foil, seen in Figure 7, show the 411.8-keV line of Au^{198} (2.7-d half-life) as well as strong Na^{24} lines. Gold was the last activated foil, so the Na^{24} lines should be the strongest for this plate. Gold-198 was the only activation product expected.

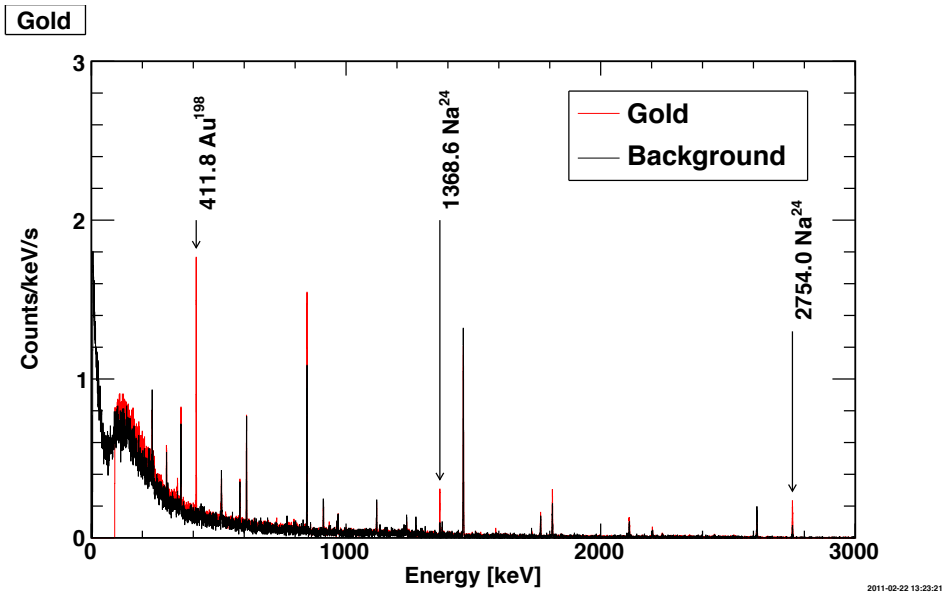

Figure 7: HPGe spectrum of gold foil

4.0 Conclusions

The activated samples present minimal radiological risk. Only the silver foil provided dose above background, 0.8 mR/h of betas on contact. However, that dose rapidly decayed with the relevant parent isotope half-lives on the scale of minutes. Gamma-rays from the silver, manganese and gold plates where observed using the HPGe detector. All of the gamma-rays for the silver and gold plates were identified, whereas 8 out of 11 gamma-rays from the activated manganese plate were identified. The nylon bag and aluminum plate appeared not to be activated. The iron plate may have gamma-rays related to ^{56}Mn, but it is difficult to distinguish whether those lines are from the activated iron plate or the activated room background.

Future efforts include running similar tests on other plates as well as a Bismuth-Germanate (BGO) scintillator. Once those tests have been completed, we will conduct a safety review of the intended (n,γ) experiments. Those measurements involve detecting the gamma rays emitted during the radiative neutron capture process from the foils. The metal foils were picked because they have a strong resonance in the energy region of interest for the LSDS for this process. Through these measurements, we will be able to determine the time and energy correlation of the neutrons in the LSDS.

Appendix A: RPT Report

Below is the initial survey report covering the two days when the activations were completed. There is a second report, 331-11-02-064, when the foils were sampled the following week in which all direct readings were below the minimum detectable activity.

Pacific Northwest NATIONAL LABORATORY

Radiological Control Record
Radiological Survey Report

Survey Report Number: **331-11-02-050**

Date	Time	Purpose of Survey: [ ] Routine [X] Demand [ ] RCHP FOIL ACTIVATIONS	H43238
02/18/2011	10:00-16:00		

Room(s) / Item(s)	Building	TWD(s)#	RWP Number
171	331	LSDS Meas.	171A-Foils

SMEAR	DATE	TIME	SAMPLE ID/METAL	DIRECT READING (INITIAL CPM)	DIRECT READING (RECOUNT CPM)	RECOUNT TIME
1	02/18	10:00	Al cap	NA	<1000	1545
2	02/18	13:00	MN plate	3,500	2,000	1545
3	02/18	15:30	Gold foil	<1000	<1000	1545
4	02/17	12:00	Iron plate	NA	<1000	1545
5	02/17	12:00	Silver plate	NA	<1000	1545

Dose Rate Measurements

Item Description	Inst. #	Distance	OW	CW	CF_{Beta}	mrem/h β	CF_{Gamma}	mrem/h γ	CF_{other}	Smear #
Misc Metals	4	C/30cm	<0.5	<0.5	NA	<0.5	NA	<0.5		

Contamination Measurements

#	Location	β-γ	Inst. #	β-γ CF	α	Inst. #	α CF
1-3	Items coming out of the RGD	<MDA	1,2	10	<MDA	3,4,5	7/2.8

Instruments Used	1.	CMEBB-0227	2.	DTHNC-1120	3.	ACBC1-0045	4.	ICEB3-0199
5.	CSEB3-0032	6.		7.		8.		9.	

Signature on this survey form indicates that 1) the instruments above have been source checked in accordance with *RCP-5.5.06, Portable Radiological Survey Instruments*, and 2) RCT actions in the applicable TWDs related to this job have been reviewed in accordance with *RCP-3.4.09, Radiological Job Coverage and Emergency Response*.

= mrem/h gamma ### β = mrem/h beta ### η= mrem/h whole body neutron ✱ = mrem/h Contact E = mrem/h Extremity
D# = Direct Survey (dpm/100cm²) M# = Smear (dpm/smear) ## = Air Sample (##) = Technical Smear (dpm/100 cm²) ~(##)~ = Special Smear (dpm/100 cm²)
<MDA: MDAs for portable survey instruments and field counters (i.e., SAC-4, BC-4, and Ludlum 2929) are found in RCP-5.5.11, Radiological Surveys, Exhibit 10.
Ludlum 2200 = < _____dpm/β–γ

RCT Name and Signature: Angie Hall / Date: 02/18/2011
Reviewed By RCT Supervisor: Holly Black-Kania / Date: FEB 18 2011

5511ex1.dot RCP-5.5.11 (11/08)

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