



U.S. DEPARTMENT OF
ENERGY

PNNL-21352

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

Report on Second Activations with the Lead Slowing Down Spectrometer

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April 2012



Pacific Northwest
NATIONAL LABORATORY

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Pacific Northwest National Laboratory
Richland, Washington 99352

Summary

On August 18 and 19 2011, five 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. As during the first activation run, we observed activation in the room in general, mostly due to ^{56}Mn and ^{24}Na . Most of the activation of the samples was short lived, with half-lives on the scale of hours to days, except for ^{60}Co which has a half-life of 5.3 y.

Acronyms and Abbreviations

PNNL	Pacific Northwest National Laboratory
RPT	Radiation Protection Technician
HPGe	High-purity germanium
LSDS	Lead slowing down spectrometer/spectroscopy

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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. Using a series of measurements, the mid-term objective is to correlate the neutron energy with neutron slowing-down time as the neutrons progressively lose energy as they collide with the Pb nuclei in the LSDS. 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 in February 2011, and then conducted dose and gamma-spectroscopy measurements to develop a better understanding of the risks. The air release permit from the State of Washington did not allow the testing of additional foils until August, 2011. This report describes the activations and the ensuing characterization measurements of a second set of foils which completes the initial radiological safety tests.

1.0 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 neutron generator was 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 five irradiated items are summarized in Table 1, listed in order of decreasing expected activity. These items are foils of indium, copper, cadmium, cobalt, and tantalum. 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 were irradiated for an hour by a continuous source of D-T neutrons, with settings of 60 μ A and 90 kV on the neutron generator.

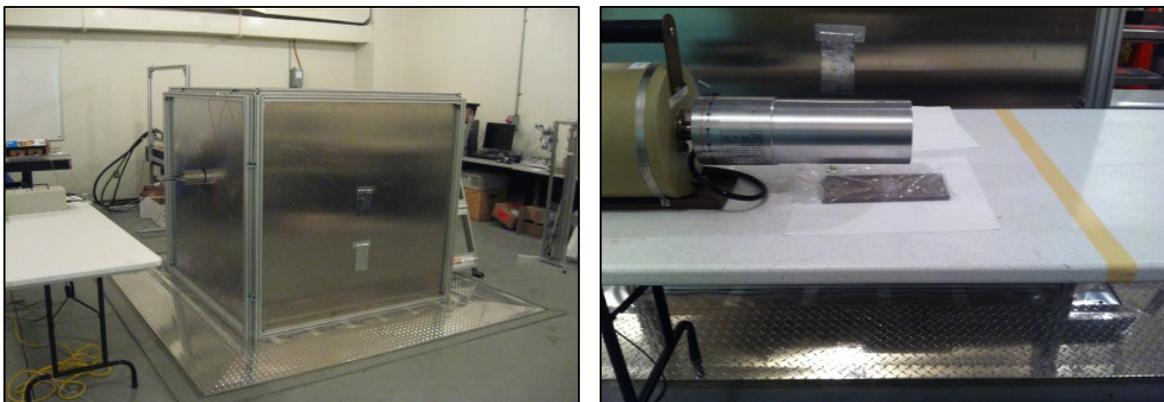


Figure 1: Photo of the lead stack with the DT generator in place (left) and photo of the HPGe detector measuring a foil in Feb., 2011(right). Note that the table was moved between photos.

Table 1: Summary of irradiated samples. Weight does not include the nylon bag. ^{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.

Sample	Weight (g)	Isotopes Observed	$T_{1/2}$	γ Signature (keV)
Indium	4.76	In-116m	54.2 m	417, 1097, 1294
Copper	1107.2	Cu-66	5.1 m	1039
Cadmium	521.4	Cd-115	53.5 h	336, 528
		Cd-117	2.49 h	273, 344, 1303, 1576
Cobalt	54.45	Co-60	5.3 y	1173, 1332
Tantalum	53.93	Ta-182	114 d	1121, 1189, 1221, 1231

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 found, but the samples were activated. When it was initially pulled out of the stack, the indium had 2.0 mR/h dose rate at 1 inch for betas, but no dose for gammas. All of the foils except indium registered less dose than the detector limit of 0.5 mR/h. The copper had a direct survey beta-gamma result of 5000 dpm/100cm². The RPT reports are provided in Appendix A.

A 120% relative efficiency HPGe detector was used to conduct the gamma spectroscopy measurements. The heavier copper and cadmium samples were placed on the table 4.375" below the center of the detector, while the lighter indium, cobalt and tantalum were placed on top of the detector. The HPGe detector was removed from the vault while the neutron generator was running. The HPGe was tested prior to the measurements and the resolution was determined to be acceptable, having about a 2.2

keV FWHM at the 1460 keV ^{40}K line. The data acquisition system had 16384 channels and a full range of about 3100 keV, yielding approximately 0.19 keV/channel.

2.0 Results

2.1 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 several hours. The spectrum in Figure 2 shows strong Mn^{56} lines, which likely result from either the activation of iron or manganese. Lines at 270, 336, 511, 528 and 806 keV also appear in the spectrum but have not been identified. Manganese-56 has a half-life of 2.6 h, so its activity will be negligible after roughly one day. This incidental activation of the room will disappear over the period of roughly a day.

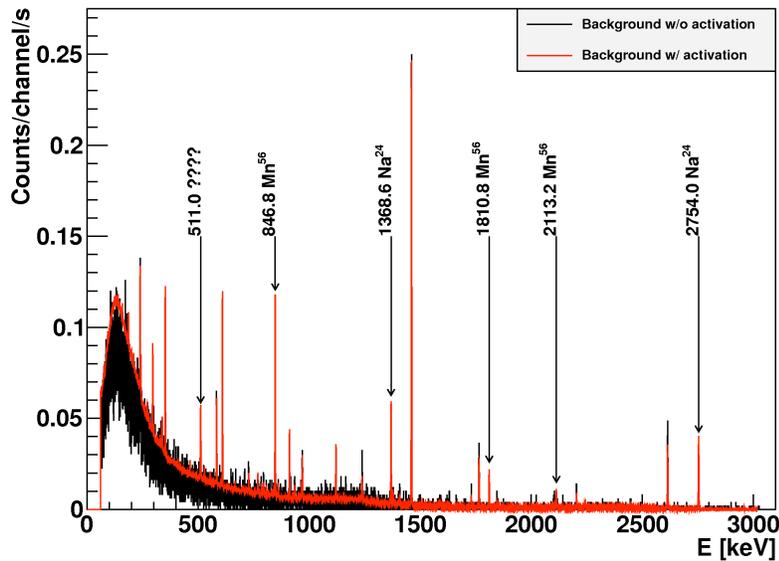


Figure 2: Comparison of room background before and after activation

2.2 Indium

Indium was expected to have the highest activity post-irradiation amongst the second set of foils. A significant deviation from the background was observed, and it was most likely due to the many Compton edges from the gamma-rays and the beta spectrum from the beta-emitters. The indium foil was placed directly on the HPGe detector (while still in the nylon bag), so there is a good chance that bremsstrahlung from the beta particles is being detected in the HPGe. Due to the elevated background, it is difficult to verify that there are no weak additional lines. However, the lines that are observed were strong and from $^{116\text{m}}\text{In}$: 417, 819, 1097 and 1294 keV. The weaker 138.3 keV line is also from $^{116\text{m}}\text{In}$. Lines at 2391, 2529, and 2808 keV are sum peaks of the strong $^{116\text{m}}\text{In}$. The thin indium foil was wrapped around the outside of the HPGe detector so there was a good probability of coincidences. There is also a 2112 keV line from $^{116\text{m}}\text{In}$, but it is difficult to resolve from an activated room background line from ^{56}Mn at 2113 keV.

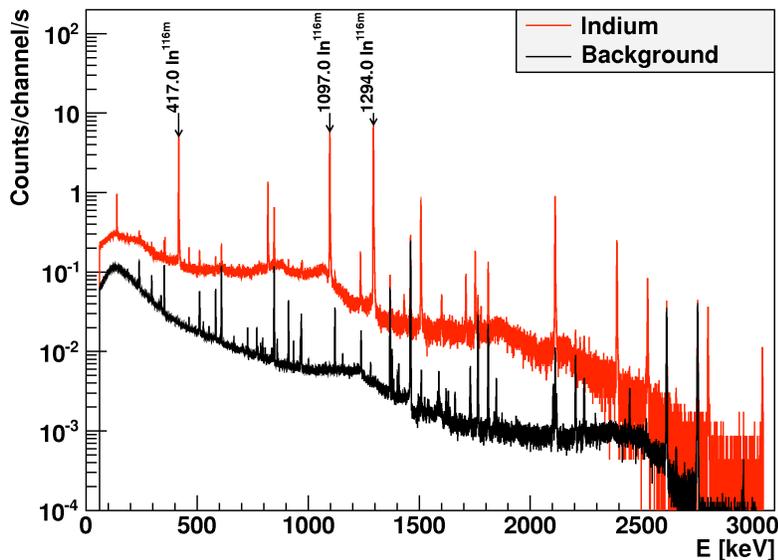


Figure 3: HPGe spectrum from indium foil.

2.3 Copper

The copper spectrum, shown in Figure 4, is dominated by contributions from Mn^{56} , likely from room background, as shown in Figure 2. The strongest gamma peak is the 1039 keV peak from ^{66}Cu . There is also a weak line at 1346 keV from ^{64}Cu . Most of the lines are from the background activation, but there are at least two, weak unidentified lines: 245 and 443 keV.

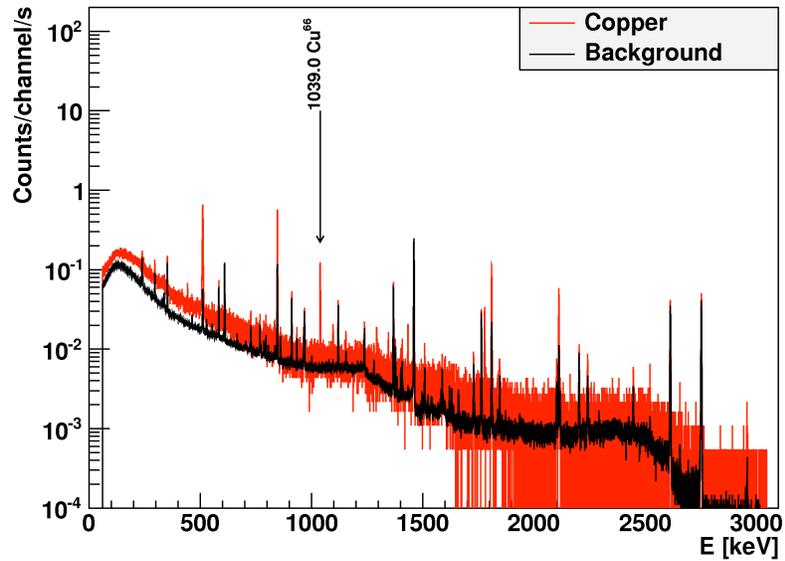


Figure 4: HPGe spectrum from copper.

2.4 Cadmium

Figure 5 shows the HPGe spectrum for activated cadmium out to 3 MeV while Figures 6 and 7 shows two selected energy regions. There are several naturally occurring isotopes of cadmium leading to several different excited nuclei. As a result, the gamma-ray spectrum is more complicated than for the other foils. Two energy regions were chosen where the known lines tended to cluster. Several lines from ^{115}Cd and ^{117}Cd are shown in Figures 6 and 7. There are additional lines which have not been identified. The cadmium also had an elevated counting rate of 312 cps versus the 194 cps of background. However, since the half-lives are in the several hours to days range, any residual ^{115}Cd and ^{117}Cd activity will decay away in a fairly short amount of time.

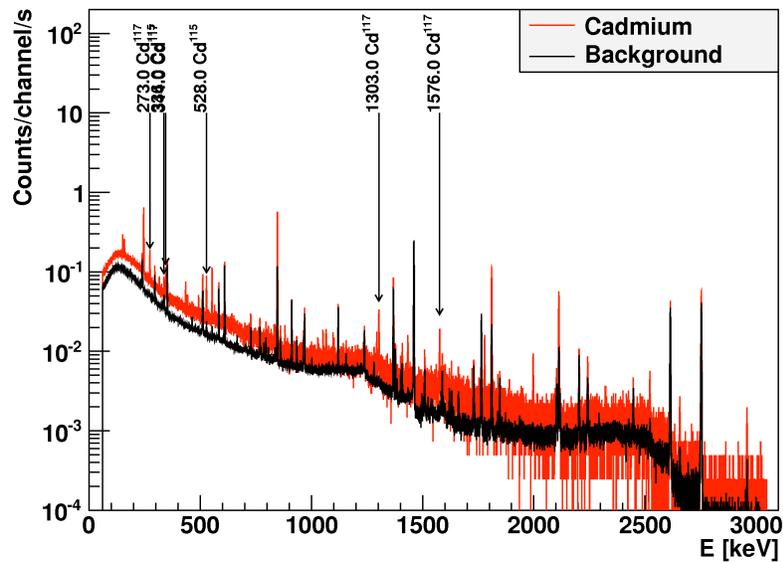


Figure 5: HPGe spectrum for cadmium.

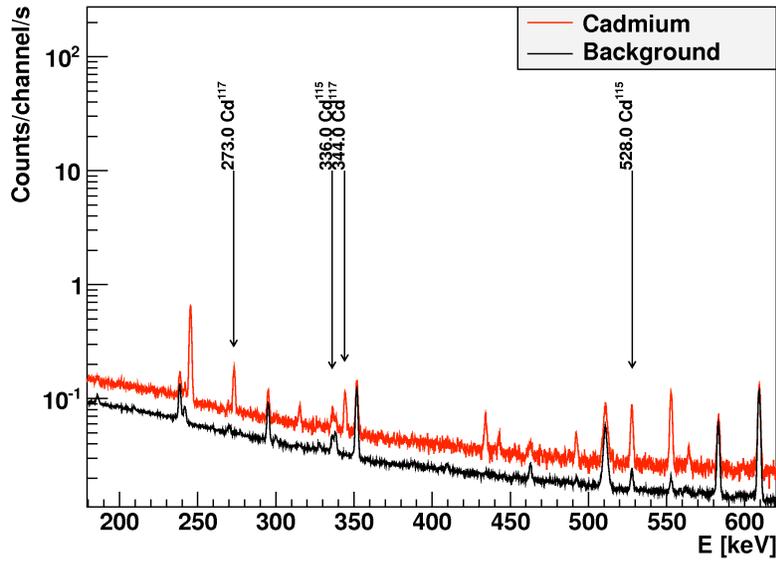


Figure 6: HPGe spectrum of cadmium in a lower energy region.

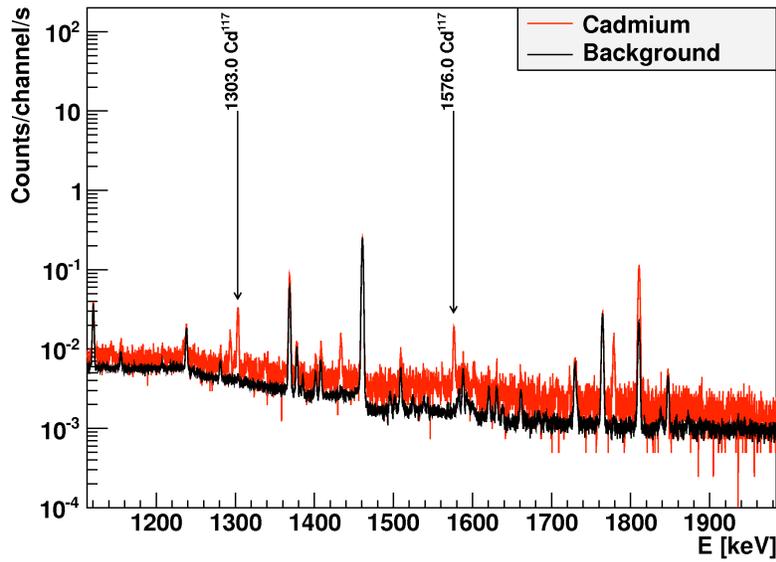


Figure 7: HPGe spectrum of cadmium in a higher energy region.

2.5 Cobalt

Cobalt was one of the weaker sources due to the much longer half-life of the main product, ^{60}Co . The two main lines from ^{60}Co (1173 and 1332 keV) are just able to be seen in Figure 8.

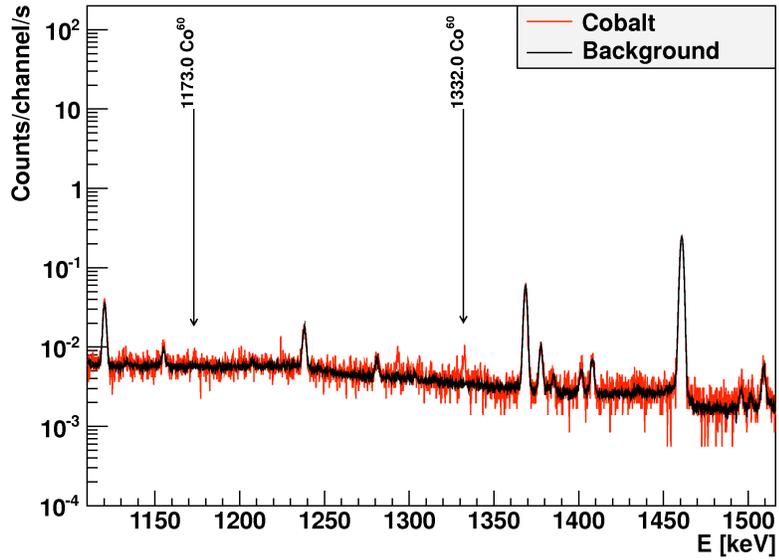


Figure 8: HPGe spectrum of activated cobalt.

2.6 Tantalum

The results from tantalum are shown in **Figure 9** and Figure 10. Even with the foil placed directly on the HPGe, the detector count rate started at 291 cps and fell to 241 cps after an hour (background rate was 194 cps). It is clear in **Figure 9** that there is some elevation of the level of the continuum, but there are not any prominent peaks. Figure 10 shows the results in a small energy range so that the individual peaks can be more easily seen. The peaks are much clearer but not particularly strong. All of the peaks listed in Table 1 were observed except for 1147 keV, which may be present but is weak, and 1121 keV which overlaps with a ^{214}Bi background line. The 1121 keV counts are elevated above background.

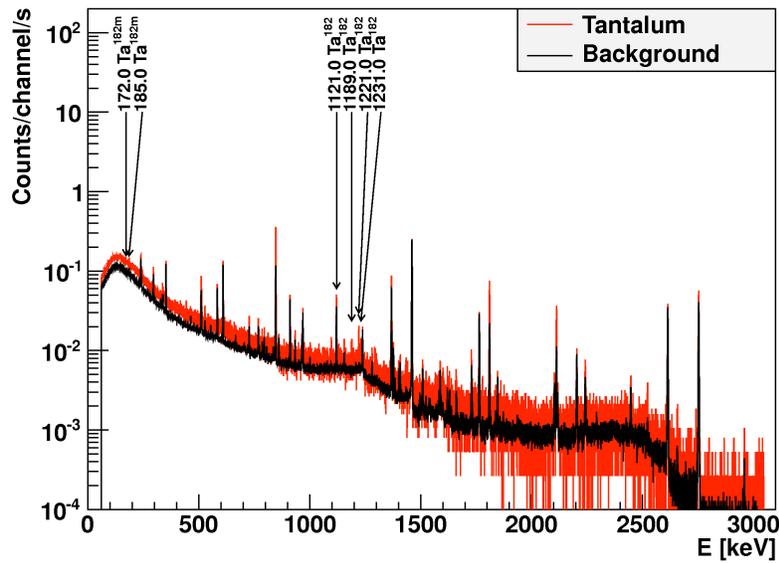


Figure 9: HPGe spectrum from activated tantalum.

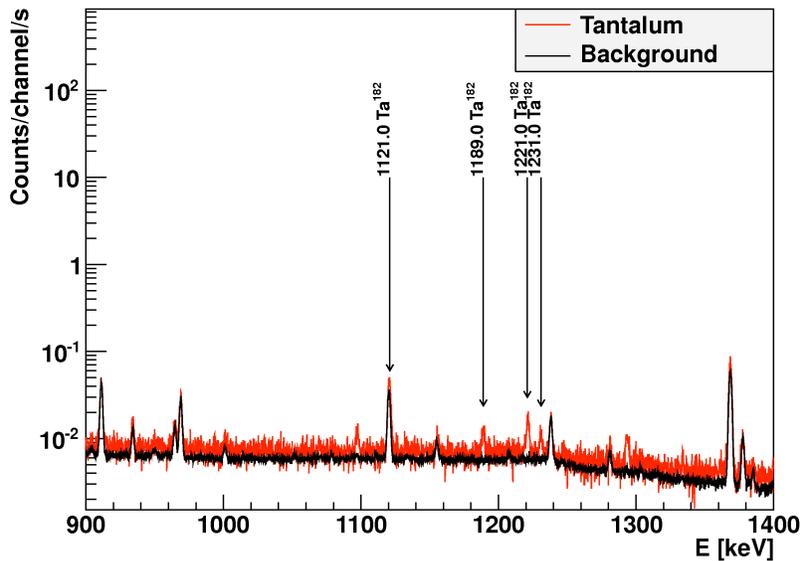


Figure 10: HPGe spectrum of tantalum in a small energy range.

3.0 Conclusions

The second set of activated samples presents minimal radiological risk. Only the indium foil registered a dose, 2.0 mR/h of betas at 1 inch. Activity was detected in each foil, but they are expected to rapidly decay with the relevant parent isotope half-lives on the scale of hours to days except for cobalt-60, which has a half-life of 5.3 years. Gamma rays from all the foils were observed using the HPGe detector. All of the expected gamma-rays were identified with only one exception in tantalum. The manganese background continues to make identification difficult when the activity is low.

Now that these additional foils have been found to present minimal radiological risk, future efforts will involve detecting the gamma rays emitted during the radiative neutron capture process. The metal foils were chosen because they have strong neutron absorption resonances in the neutron 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 are the initial survey reports covering the two days when the activations were completed.

Indium

Copper

Pacific Northwest Radiological Control Record										Survey Report Number		
Radiological Survey Report										331-11-08-49		
Date	Time	Purpose of Survey	Route	Demand	RCHP	Foal activation	Room(s) / Item(s)	Building	TWD(s)	RWP Number		
8/18/11	1300						171	331	5.5.11	RPT1		
Item Description	Inst. #	Distance	OW	CF _{max}	mrem/h	CF _{gross}	mrem/h	30cm	Smear #			
Foil	5	1'	1	< 0.5	2	-	-					
Contamination Measurements												
#	Location	Inst. #	CF	Inst. #	CF	Inst. #	CF					
1	Foil		< MDA	4	2.9		< MDA	4	2.8			
Instruments Used												
1. CMEB3-0050		2. DTHNC-0055		3. ACBC1-0097		4. SCLL4-0022						
5. ICEB4-1429		6.		7.		8.						
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.												
RPT Name and Signature: J. Zelazo, Date: August 18, 2011, Reviewed By: H. Black-Kania, Date: AUG 18 2011												

Pacific Northwest Radiological Control Record										Survey Report Number		
Radiological Survey Report										331-11-08-49		
Date	Time	Purpose of Survey	Route	Demand	RCHP	Foal activation	Room(s) / Item(s)	Building	TWD(s)	RWP Number		
8/18/11	1300						171	331	5.5.11	RPT1		
Item Description	Inst. #	Distance	OW	CF _{max}	mrem/h	CF _{gross}	mrem/h	30cm	Smear #			
Foil	5	1'	1	< 0.5	2	-	-					
Contamination Measurements												
#	Location	Inst. #	CF	Inst. #	CF	Inst. #	CF					
1	Foil		< MDA	4	2.9		< MDA	4	2.8			
D1	Foil		5000	1.2	10		< MDA	3	7			
Instruments Used												
1. CMEB3-0050		2. DTHNC-0055		3. ACBC1-0097		4. SCLL4-0022						
5. ICEB4-1429		6.		7.		8.						
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.												
RPT Name and Signature: J. Zelazo, Date: August 18, 2011, Reviewed By: H. Black-Kania, Date: AUG 18 2011												

Cadmium

Pacific Northwest NATIONAL LABORATORY		Radiological Control Record Radiological Survey Report			Survey Report Number: 331-11-08-051	
Date	Time	Purpose of Survey: <input type="checkbox"/> Routine <input type="checkbox"/> Demand <input type="checkbox"/> RCHP <input type="checkbox"/> Survey of samples				
Room(s) / Item(s)	Building	TWID(s)	RCP	RWP Number	RPT1	
171	331		5.5.11			
Foil sample post neutron exposure <0.5 @ * stored in RMA						
Dose Rate Measurements						
Item Description	Inst. #	Distance	OW	CF _{inst}	min/m ² β	CF _{area} min/h y
See Above	4	As	Noted			
Contamination Measurements						
#	Location	Inst. #	β-α	Inst. #	β-α	CF
1-2	Foil Sample		<mda	10	<mda	3/5 7/2.8
Instruments Used						
1. CMEBB-0001	2. DTEB9-0485	3. ACHN2-0114	4. ICEB4-1490	5. SCLL4-0022	6.	7.
Signature on this survey form indicates that 1) the instruments above have been source checked in accordance with RCP-3.5.06, Portable Radiological Survey Instruments, and 2) RPT actions in the applicable TWIDs related to this job have been reviewed in accordance with RCP-3.4.06, Radiological Job Coverage and Emergency Response.						
RPT Name and Signature: <i>[Signature]</i> Date: 8/18/11 Reviewed By: RPT Supervisor: <i>[Signature]</i> Date: AUG 19 2011 551 text dot RCP-3.5.11 (12/08)						

Cobalt and Tantalum

Pacific Northwest NATIONAL LABORATORY		Radiological Control Record Radiological Survey Report			Survey Report Number: 331-11-08-053	
Date	Time	Purpose of Survey: <input type="checkbox"/> Routine <input type="checkbox"/> Demand <input type="checkbox"/> RCHP <input type="checkbox"/> Survey of samples				
Room(s) / Item(s)	Building	TWID(s)	RCP	RWP Number	RPT1	
See below	331		3.4.04, RCP 5.5.11			
Room	Description	Dose Rate	Tech smear on sample/container/ special smears	Boundary Survey	Floor under hood or chamber	Disposition of samples
113	2 jugs of liquid	<0.5	1,2	3,4	5,6	Stored in RMA
195	Shipped rats to RPL, 2 boxes of samples containing Co60	<0.5	7-15	16	17	Stored in RMA
171	Foil samples	<0.5	18,19,20	NA	NA	Stored in RMA
166	Trash and samples	<0.5	21-25	26	27	Stored in RMA
Dose Rate Measurements						
Item Description	Inst. #	Distance	OW	CF _{inst}	min/m ² β	CF _{area} min/h y
See Above						
Contamination Measurements						
#	Location	Inst. #	β-α	Inst. #	β-α	CF
1-27	See above items in the chart		<MDA	1,2,5 10/2.9	<MDA	3/5 7/2.8
Instruments Used						
1. CMEBB-0077	2. DTHNC-0196	3. ACHN2-0425	4. LMBC1-0015	5. SCLL4-0022	6.	7.
Signature on this survey form indicates that 1) the instruments above have been source checked in accordance with RCP-3.5.06, Portable Radiological Survey Instruments, and 2) RPT actions in the applicable TWIDs related to this job have been reviewed in accordance with RCP-3.4.06, Radiological Job Coverage and Emergency Response.						
RPT Name and Signature: <i>[Signature]</i> Date: 08/18/2011 Reviewed By: RPT Supervisor: <i>[Signature]</i> Date: AUG 29 2011 551 text dot RCP-3.5.11 (11/08)						



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