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January 23, 2012

Reply to Attn of: Q

U.S. Nuclear Regulatory Commission
Attn: Document Control Desk
Washington, D.C. 20555

Subject: Plum Brook Reactor Facility, Licenses Nos. TR-3, Docket No. 50-30 and R-93, Docket No, 50-185, Technical Basis for Reevaluation of Structural DCGLs and Uranium Activity Fractions

An Independent Confirmation Survey performed during the week of July 15, 2011, by the Oak Ridge Institute for Science and Education (ORISE) identified a potential issue regarding the calculation of gross beta DCGLs when Uranium-234 was present in the radionuclide mixture.

In a telephone conference call with NRC Staff on November 1, 2001, NASA agreed that ORISE was correct on the calculation error identified in their report dated October 31, 2011. NASA agreed to evaluate the error and assess it's affect on all areas already surveyed under our Final Status Survey Program and to address any impacts on the conclusions stated in Final Status Survey Report Attachments previously submitted.

Technical Basis Document, PBRF-TBD-11-002, "Re-evaluation of Structure DCGLs and Uranium Activity Fractions", Revision 0, dated January 4, 2012, is enclosed for your information. It addresses the issue with erroneous surrogate DCGL calculation, the impact of the presence of Uranium-234 in the radionuclide mixture, and the impact of the error on our conclusions documented in previous submittals.

Should you have any questions or need additional information, please contact me a NASA Plum Brook Station, 6100 Columbus Avenue, Sandusky, Ohio 44870, or by telephone at (419) 621-3242.

Peter C. Kolb
NASA Decommissioning Program Manager

FSME20
FSME

Enclosures

1. Technical Basis Document PBRF-TBD-11-002, "Re-evaluation of Structure DCGLs and Uranium Activity Fractions", Revision 0, dated January 4, 2012.

cc:
USNRC/C. J. Glenn (FSME)
USNRC/J. Webb (FSME)
USNRC/J. Tapp RIII/DNMS/DB
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Plum Brook Reactor Facility

Technical Basis Document

Re-evaluation of Structure DCGLs and Uranium Activity Fractions

PBRF-TBD-11-002
Revision No. 0

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RSO Approval:	W. Stoner [signature]	Date:	1/4/12

Form
AD-04/02
Rev 0

NASA PBRF DECOMMISSIONING PROJECT CHANGE/CANCELLATION RECORD		
DOCUMENT TITLE: Re-evaluation of Structure DCGLs and Uranium Activity Fractions	**DOCUMENT NO: PBRF-TBD-11-002**	**REVISION NO: 0**
Revision 0: Initial issue of document.		

Form AD-01/3 Rev 1

LIST OF EFFECTIVE PAGES

DOCUMENT NO: PBRF-TBD-11-002 REVISION NO.: 0

Page No.	Revision Level	Page No.	Revision Level	Page No.	Revision Level
Cover Page	0				
Change/Cancellation Record	0				
LOEP	0				
TOC (1 page)	0				
Text 18 pages	0				
Appendix A (3 pages)	0				
Appendix B (4 pages)	0				
Appendix C (2 pages)	0				

Form
AD-01/ 5
Rev 2

TABLE OF CONTENTS

LIST OF TABLES

LIST OF FIGURES

Executive Summary

Concurrent with an NRC inspection at the Plum Brook Reactor Facility (PBRF) Decommissioning Project in July 2011, radiological surveys and records review were performed by the NRC independent verification contractor, the Oak Ridge Institute for Science and Education (ORISE). The ORISE review included a review of gross beta activity derived concentration guideline (DCGL) values used by the PBRF to evaluate final status survey (FSS) measurements for compliance with the 25 mrem/y release criterion. The gross beta activity DCGL values and the radionuclide mixtures used to calculate them are published in a PBRF Technical Basis Document (TBD), PBRF-TBD-07-001.

The ORISE report, issued in October 2011, identified an error in the gross beta activity DCGL used for the -15 ft. elevation (Area 20) of the Reactor Building, 35,296 dpm/100-cm^2. The ORISE report determined that this value did not correctly account for the use of beta surface activity measurements in areas such as the Reactor Building -15 ft. elevation where the radionuclide mixture reported in TBD-07-001 included alpha emitting U-234. According to the ORISE report, the correct DCGL value for this area is 17,084 dpm/100-cm^2.

In subsequent discussions with the NRC, it was agreed that NASA would re-evaluate the method used to calculate DCGLs, with particular attention given to areas where uranium was included in the radionuclide mixture. This evaluation would also include a review of the basis for determining that uranium was present in the radionuclide mixtures. As a result of this re-evaluation it was confirmed that the DCGLs published in TBD-07-001 for areas where uranium was included in the radionuclide mixture were indeed incorrect and they are corrected. The concrete core sample results used to establish radionuclide mixtures in TBD-07-001 were found not to be representative of residual contamination from enriched uranium, for which the activity is predominately U-234. The core sample results are consistent with natural uranium where U-234 and U-238 activities are equal and are at levels indistinguishable from the natural uranium in concrete. Thus, the radionuclide mixtures used to calculate DCGLs for the PBRF areas are revised by removing uranium (U-234 and U-235) from the mixture in all areas except the New Fuel Storage Vault (where the activity fractions are based on smear sample results and are indicative of enriched uranium).

The gross beta activity DCGLs for all 21 areas in the PBRF structures were recalculated using the revised radionuclide mixtures and the corrected method of calculating DCGLs. The changes in DCGL values which result are minor, except for the New Fuel Vault (a decrease from 30,831 to 11, 475 dpm/100-cm^2, or -62.8%) and the Reactor Building -15 ft. elevation, Area 20 (an increase from 35,296 to 40,500 dpm/100-cm^2, or 14.7%). Note that the latter DCGL is different from the value calculated by ORISE (17,084 dpm/100-cm^2) due to removal of U-234 from the mixture. The DCGL for the two areas where the "default" radionuclide mixture is used is also changed (from 27,166 to 23,146 dpm/100-cm^2, a decrease of 14.8%).

1.0 Introduction

An independent verification survey of several buildings and land areas undergoing final status survey at the NASA Plum Brook Reactor Facility was conducted in July 2011. The survey was conducted by ORISE, the independent verification contractor for the US Nuclear Regulatory Commission (NRC). The survey included total surface activity measurements (alpha plus beta) in the Reactor Building inner bioshield, Quads A through D and the -15 and -25 ft. elevations of the Reactor Building. The ORISE final report identified a potential issue regarding the manner in which the gross beta activity DCGL was determined where alpha-emitting U-234 was identified in the assigned radionuclide mixture [ORISE 2011].

The ORISE Report stated that the gross beta activity DCGL for the Reactor Building -15 ft. elevation (also identified as Area 20), 35,296 dpm/100-cm^2, was not adjusted to account for the use of Cs-137 as a surrogate for hard-to-detect (HTD) radionuclides, including U-234. The ORISE report indicated that the correct DCGL for this situation is 17,084 dpm/100-cm^2 [ORISE 2011]. The matter was discussed on a conference call with the NRC Headquarters and Region III Staff on Nov 1, 2011. It was agreed that the issue would be investigated, resolved by NASA/PBRF and the results provided to the NRC. In accordance with the PBRF QA procedure for corrective actions, a problem report was prepared for entry into the PBRF corrective action database (called the C/A-5 system) [PBRF 2011]. [1]

This document is prepared to address the technical issues identified in the ORISE Report in a manner that is responsive to the corrective actions identified in PBRF Problem Detail Report # 0421. Specific objectives of this report are to:

1. Review the method used in TBD-07-001 [PBRF 2007] to calculate DCGLs, and revise as necessary the DCGLs calculated for radionuclide mixtures in the PBRF structures.

2. Re-evaluate the radionuclide mixtures used in TBD-07-001 to develop DCGLs in areas identified as containing uranium. This is in light of the limited sample information used to develop the mixtures and the low activity concentration of uranium in the samples relative to the natural uranium content of concrete.

Background information is provided in Section 2.0. The method for calculating gross activity DCGLs in the PBRF FSS Plan is described. The information used to calculate DCGLs and to develop radionuclide activity fractions in TBD-07-001 is summarized. References are provided in Section 3.0. In Section 4.0, revised DCGLs are presented and radionuclide mixtures and activity fractions in uranium areas are re-evaluated. Section 5.0 presents the conclusions reached from the review and evaluations. Supporting information is provided in Appendices. Appendix A contains a summary of special nuclear material (SNM) and source material inventories at the PBRF and a history of handling and storage of these materials in the various PBRF buildings. Appendix B provides information on uranium characteristics to aid in interpreting measurement results. Appendix C provides details of the revised DCGL calculations.

[1] The corrective action program is directed by the PBRF Procedure, QA-03, Audits and Corrective Actions, Rev. 2, February 22, 2006.

2.0 Background

The methods used in TBD-07-001 to calculate DCGLs for the FSS and to establish radionuclide activity fractions are summarized in this section. Background information is provided to lay the ground work for re-calculation of DCGLs as may be necessary in view of the ORISE finding regarding the gross beta activity DCGL for the Reactor Building Area 20. Many of the concrete core characterization samples used to establish radionuclide mixtures and activity fractions contained low activity concentrations of uranium. This raises the question of whether the core sample uranium activity is due to natural background and not of PBRF origin.

2.1 DCGL Development

Gross activity DCGLs used in survey designs for the FSS of PBRF structures were developed in TBD-07-001. The DCGLs were developed from radionuclide mixtures established for specified PBRF areas. A default DCGL was established for areas where insufficient sample results were available to determine radionuclide mixtures and activity fractions. Table 1 shows the single radionuclide structure DCGLs for the eight principal radionuclides established in the FSS Plan [NASA 2007].

Table 1, Single Radionuclide Structure DCGLs from the FSS Plan

Structure DCGLs (dpm/100-cm^2)							
H-3	Co-60	Sr-90	I-129	Cs-137	Eu-154	U-234	U-235
9.10E+06	11,000	33,100	14,900	40,500	4,500	31,500	27,100

In TBD-07-001, these DCGLs and the radionuclide mixtures were used to calculate gross beta activity DCGLs for 19 areas in PBRF structures. The radionuclide activity ratios and the DCGLs are shown in Table 2. The method described in TBD-07-001 for calculating the DCGLs is obtained from the FSS Plan. The description of the method which follows is excerpted from Section 3.6 of the Plan.

> "For structural surfaces, the final DCGL for FSS design and implementation will be a gross beta DCGL that represents the 25 mrem/yr unrestricted use criterion. There are two steps required to determine the gross beta DCGL:
>
> 1. perform a surrogate calculation to account for radionuclides that cannot readily be measured by a beta-sensitive detector (e.g., plastic scintillation, gas proportional) with typical efficiency and
>
> 2. perform a gross activity DCGL calculation on the surrogate DCGL values and other beta-emitting radionuclides determined to be present in significant fractions.

The surrogate DCGL is computed based on the distribution ratio between the hard-to-detect radionuclides and the easy-to-detect radionuclides. The surrogate DCGL is calculated using the following equation: [2]

$$DCGL_{Sur} = \frac{1}{\left[\left(\frac{1}{DCGL_1}\right)+\left(\frac{R_2}{DCGL_2}\right)+\left(\frac{R_3}{DCGL_3}\right)+....+\left(\frac{R_n}{DCGL_n}\right)\right]} \quad \text{(Equation 1)}$$

Where: $DCGL_{Sur}$ = Surrogate radionuclide DCGL,
$DCGL_1$ = DCGL for the radionuclide to be used as the surrogate for the other radionuclides,
$DCGL_2$, $DCGL_3$,... $DCGL_n$, = DCGL for each radionuclide to be represented by the surrogate and
R_2 ....R_n = Ratio of concentration (or nuclide mixture fraction) of radionuclides 2 through n to surrogate radionuclide.

Where multiple radionuclides are present, a gross beta DCGL can be developed. This approach enables field measurements of gross activity (in the present application, gross beta activity) rather than a determination of individual radionuclide activities for comparison to the radionuclide-specific DCGLs. The gross beta DCGL is calculated using the following equation: [3]

$$DCGL_{GB} = \frac{1}{\left[\left(\frac{f_1}{DCGL_1}\right)+\left(\frac{f_2}{DCGL_2}\right)+\left(\frac{f_3}{DCGL_3}\right)+.....+\left(\frac{f_n}{DCGL_n}\right)\right]} \quad \text{(Equation 2)}$$

Where: $DCGL_{GB}$ = gross beta DCGL
f_1, f_2, f_3, f_n = activity fraction of each radionuclide
$DCGL_n$ = DCGL of each radionuclide to be represented by the surrogate.

Note 1: The gross beta equation may also be used to calculate a gross alpha DCGL for application in areas where DCGL values are established for alpha emitters.
Note 2: If a surrogate radionuclide is used, the "f_n" is equal to the surrogate radionuclide fraction (and the surrogate DCGL is inserted in the denominator of the term).[4]
Note 3: The value of 1 in the numerator is replaced by the actual fraction of beta (or alpha) emitters if less than 100% of the mixture."

[2] Surrogate DCGL equation from Section 3.6.1 of the FSS Plan.

[3] Gross beta activity DCGL equation from Section 3.6.2 of the FSS Plan.

[4] Parenthetical comment inserted by author.

Table 2, Radionuclide Activity Fractions and DCGLs from TBD-07-001 Table 5-3

Bldg. [1]	Location	Activity Fractions								$DCGL_{GB}$
		H-3	Co-60	Sr-90	I-129	Cs-137	Eu-154	U-234	U-235	(dpm/100-cm^2)
Rx Bldg	Area 2 & Lily Pad	0.9757	0.021	0.0013	0	0.002	0	0	0	11563
Rx Bldg	Area 17, -25 ft.	0.882	0	0	0	0.053	0	0.065	0	37235
Rx Bldg	Quad A	0	0.7499	0	0	0.2501	0	0	0	13450
Rx Bldg	Quads B, C, & D	0	0.3305	0	0	0.6695	0	0	0	21566
Rx Bldg	Canals E, F, G & H	0.0116	0.1169	0	0	0.8699	0	0.0016	0	31711
Rx Bldg	0 ft, & 12 ft el.	0.5405	0.053	0.1145	0.0009	0.2872	0.0039	0	0	29060
Rx Bldg	-15 ft. el. (Area 20)	0	0	0	0	0.484	0	0.516	0	35296 [2]
Rx Bldg	-25 ft. el.	0.0729	0.5389	0.2222	0	0.1205	0.0171	0.0242	0.0042	14600
Hot Lab	All Other Areas	0	0.0058	0.1577	0	0.8347	0	0.0017	0.0001	38647
Hot Lab	Hot Cells	0.1045	0.0255	0.3302	0.0003	0.53787	0.0012	0.0004	0.00003	34404
Hot Lab	Hot Pipe Tunnel	0.006	0.0203	0.3444	0	0.624	0	0.0048	0.0005	35781
SEB	Areas O/S CPT	0.2707	0.0965	0.0788	0.0142	0.4671	0.0012	0.0698	0.0017	27166 [3]
SEB	CPT	0	1	0	0	0	0	0	0	11000
FH	All Areas	0.7769	0	0	0	0.0563	0	0.1668	0	36857
WHB	All Areas	0.0052	0.0016	0.026	0	0.9634	0	0.0036	0.0002	40051
PPH	Room 4	0.4291	0.0956	0.0256	0.2255	0.1892	0	0.0334	0.0016	11186
PPH	All other Rooms	0.5422	0.0866	0.0939	0	0.1881	0	0.0805	0.0087	26348
ROLB	New Fuel Vault	0.3902	0.0076	0.0421	0.0287	0.1755	0	0.3438	0.0121	30831
ROLB	All other Rooms	0.2707	0.0965	0.0788	0.0142	0.4671	0.0012	0.0698	0.0017	27166 [3]
HRA	Vault & pipe tunnel	0.1362	0.0522	0.0601	0	0.7346	0	0.0148	0.0021	34213
WEMS	All Areas	0	0.0756	0	0	0.9244	0	0	0	33834

Table 2 Note:

1. See Table 3 Notes for identification of building abbreviations.
2. DCGL questioned by ORISE.
3. Default DCGL.

2.2 Radionuclide Mixtures and Activity Fractions

Sample analysis results and supporting documentation from characterization samples collected from PBRF structures by decommissioning contractors from 2003 through 2006 were assembled and reviewed in TBD-07-001. These samples were screened and a set of samples was obtained that were determined to best represent the radionuclide composition of residual contamination in the PBRF structures. Samples were eliminated which:

- did not have sufficient activity,
- were subjected to limited analysis (gamma spectroscopy only), or
- did not have sufficient supporting documentation.

Table 3 summarizes the sample information used to obtain radionuclide mixtures and activity fractions for calculation of the DCGLs presented in TBD-07-001 Table 5-3. As seen in Table 3, the sample set included 67 samples comprised of 32 concrete cores, 30 smears and five of other media (concrete chips, pipe debris and sludge). Most of these samples were subjected to comprehensive radionuclide analysis comprising gamma spectroscopy and radiochemical analysis for specified non-gamma emitting radionuclides including uranium (and transuranics). Areas where uranium was identified in the radionuclide mixture are noted.

Table 3, Sample Basis for Activity Fractions in TBD-07-001 Table 5-3

Building	Location	Type of Samples	Number of Samples	Analysis Performed (1)	Comments/Information Source (2)
Rx Bldg (3)	Area 2 & Lily Pad	Smears	2	10CFR61	Summary in TBD-07-001, Table 5-5. Sample results and DCGL calculations in App. F. No U identified in the mix.
Rx Bldg (3)	Area 17, -25 ft.	Concrete Cores	1	10CFR61	Summary in TBD-07-001, Table 5-6. Sample results and DCGL calculations in App. D. U-234 only identified in the mix.
Rx Bldg (3)	Quad A	Concrete Cores	1	10CFR61	Summary in TBD-07-001, Table 5-7. Sample results and DCGL calculations in App. D. No U identified in the mix.
Rx Bldg (3)	Quads B, C, & D	Concrete Cores	2	10CFR61	Summary in TBD-07-001, Table 5-8. Sample results and DCGL calculations in App. D. No U identified in the mix.
Rx Bldg (3)	Canals E, F, G & H	Concrete Cores	4	10CFR61	Summary in TBD-07-001, Table 5-9. Sample results and DCGL calculations in App. D. U-234 only identified in the mix.
Rx Bldg (3)	0 ft, & 12 ft el.	Smears	2	10CFR61	Summary in TBD-07-001, Table 5-10. Sample results and DCGL calculations in App. F. No U identified in the mix.
Rx Bldg (3)	-15 ft. el.	Concrete Cores	1	10CFR61	Summary in TBD-07-001, Table 5-11. Sample results and DCGL calculations in App. D. U-234 only identified.
Rx Bldg (3)	-25 ft. el.	Concrete	2	10CFR61	Summary in TBD-07-001, Table 5-12.

Table 3, Sample Basis for Activity Fractions in TBD-07-001 Table 5-3

Building	Location	Type of Samples	Number of Samples	Analysis Performed (1)	Comments/Information Source (2)
		Cores			Sample results and DCGL calculations in App. D. Both U-234 & U-235 identified.
Hot Lab	All Other Areas	Concrete Cores	4	10CFR61	Summary in TBD-07-001, Table 5-13. Sample results and DCGL calculations in App. D. Both U-234 & U-235 identified.
Hot Lab	Hot Cells	Smears	14	10CFR61	Summary in TBD-07-001, Table 5-14. Sample results and DCGL calculations in App. F. Both U-234 & U-235 identified.
Hot Lab	Hot Pipe Tunnel	Concrete Cores	5	10CFR61	Summary in TBD-07-001, Table 5-15. Sample results and DCGL calculations in App. D. Both U-234 & U-235 identified.
SEB (4)	Areas O/S CPT	Smears	6	N/A	Samples not believed to be representative. Default DCGL was assigned.
SEB (4)	CPT	Concrete Cores	1	N/A	Sample results not believed to be representative. The Co-60 DCGL was assigned for conservatism. No U identified.
FH (5)	All Areas	Concrete Cores	1	10CFR61	Summary in TBD-07-001, Table 5-18. Sample results and DCGL calculations in App. D. Both U-234 & U-235 identified.
WHB (6)	All Areas	Concrete Cores	3	10CFR61	Summary in TBD-07-001, Table 5-19. Sample results and DCGL calculations in App. D. Only U-234 identified.
PPH (7)	Room 4	Concrete Cores	2	10CFR61	Summary in TBD-07-001, Table 5-20. Both U-234 & U-235 identified.
PPH (7)	All other Rooms	Concrete Cores	5	10CFR61	Summary in TBD-07-001, Table 5-21. Both U-234 & U-235 identified.
ROLB (8)	New Fuel Vault	Smears	4	10CFR61	Summary in TBD-07-001, Table 5-23. Sample results and DCGL calculations in App. F. Both U-234 & U-235 identified.
ROLB (8)	All other Rooms	Smears	2	N/A	Summary in TBD-07-001, Table 5-22. Insufficient sample supporting information to determine representative mixture and DCGL. Default DCGL assigned.
HRA (9)	Vault & pipe tunnel	Other	2	10CFR61	Concrete chips & pipe debris. Summary in TBD-07-001, Table 5-24. Sample results in App I. Both U-234 & U-235 identified.
WEMS (10)	All Areas	Other	3	Gamma spec.	Sludge samples. Summary in TBD-07-001, Table 5-25. No U identified.

Table 3 Notes:

1. Analysis for a broad suite of radionuclides is typically referred to as "10CFR61 Analysis". The commonly used term "10CFR61 analysis" derives from the 10CFR61 regulations of the NRC, specifically 10CFR61.55, Waste Classification. This regulation lists specific radionuclides whose concentration in waste streams must be determined for waste classification purposes. These include specified activation and fission products, alpha emitting uranium and transuranic radionuclides with half-lives greater than 5 years, Pu-241 and Cm-242.

2. When it is stated that a U isotope was identified, it means that it was included in the mixture and the activity fraction was calculated.
3. Rx Bldg - Reactor Building (Building 1111), also denoted as RB.
4. SEB - Service Equipment Building (Building 1131).
5. FH - Fan House (Building 1132).
6. WHB - Waste Handling Building (Building 1133).
7. PPH – Primary Pump House (Building 1134).
8. ROLB – Reactor Office and Laboratory Building (Building 1141).
9. HRA – Hot Retention Area (Building 1155).
10. WEMS – Water Effluent Monitoring Station/System (Building 1192).

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4.0 *Analysis*

In this section, the method for calculating gross beta activity DCGLs is reviewed in light of the error noted in the ORISE report and the method is corrected. Next, the activity fractions assigned to uranium in Table 5-3 of TBD-07-001 are re-evaluated considering uranium use at the PBRF, uranium radioactivity properties and measured background concentrations in concrete. Then the gross beta activity DCGLs for the areas listed in Table 5-3 of TBD-07-001 are re-calculated taking into account revised activity fractions and the corrected DCGL calculation method.

4.1 Calculation of DCGLs

As discussed in Section 2.0, the ORISE report identified a problem with the gross beta activity DCGL for the Reactor Building -15 ft. elevation, Area 20. In this area of the

Reactor Building, the assigned radionuclide mixture comprised only Cs-137 and U-234 with 48.4% and 51.6% of the total activity, respectively. In TBD-07-001, the calculated DCGLs did not consistently incorporate the surrogate DCGL into the gross beta activity DCGL calculation.

The calculation is corrected here. Using Equation 1 (in Section 2.1) and the activity fractions assigned in Table 5-3 of TBD-07-001, the surrogate DCGL is calculated as:

$$DCGL_{sur} = \left(\frac{1}{\frac{1}{40{,}500} + \frac{0.516/0.484}{31{,}500}} \right) = 17{,}083 \text{ dpm/100-cm}^2,$$

Where: 0.484 = Cs-137 activity fraction; 40,500 = Cs-137 DCGL and
0.516 = U-234 activity fraction; 31,500 = U-234 DCGL.

The gross beta DCGL is calculated per note 3 following Equation 2, by replacing the value "1" in the numerator of Equation 2 with the activity fraction of the detectable radionuclides (in this case only Cs-137) and inserting the surrogate DCGL calculated above into the denominator as follows:

$$DCGL_{GB} = \left(\frac{0.484}{\frac{0.484}{17{,}083} + \frac{0}{DCGL_{Co}} + \frac{0}{DCGL_{Sr}}} \right) = 17{,}083 \text{ dpm/100-cm}^2.$$

In the present case, the calculation shown above is not necessary as there is only one non-zero term in the denominator; thus $DCGL_{GB} = DCGL_{sur}$. The second and third terms in the denominator are shown to note that in the general case, the quotients of activity fraction and DCGL for other detectable radionuclides are included in the calculation.

All the DCGLs listed in Table 5-3 of TBD-07-001 were re-calculated using an EXCEL template which incorporates the method described above. Significant differences were found between the re-calculated gross beta activity DCGL values and those listed in Table 5-3 of TBD-07-001 for several of the areas. The differences are due to two causes:

a. Failure to properly incorporate the surrogate DCGL into the gross beta activity DCGL calculation.
b. The DCGLs in TBD-07-001 were obtained as the average of DCGLs calculated for each characterization sample radionuclide mixture in areas where multiple sample results were available. In this approach, an accumulation of round-off and truncation errors in the many ratios of small numbers appears to have contributed to the differences noted.

The largest differences are seen in those areas where significant uranium activity fractions were assigned in TBD-07-001. The revised DCGLs are shown in Section 4.3 below. They include the incorporation of revised activity fractions developed in the next section.

4.2 Review of Uranium Activity Fractions

A review was conducted of the information used to obtain radionuclide activity fractions for calculation of DCGLs in TBD-07-001. This review was essentially a revisit of the evaluation in Section 5.3.2 of the TBD and the supporting sample information in the TBD appendices. The sample analysis results were reviewed to determine if inclusion of uranium in each of the mixtures is reasonable considering that the activity in the characterization core samples was very low. Also considered are the process history, uranium activity characteristics and uranium background concentrations in concrete. Table 4 identifies those areas in the PBRF buildings where uranium was included in the radionuclide mixtures for calculation of DCGLs reported in TBD-07-001. This table is derived from Table 3 in this report.

Table 4, Areas with Uranium in the Nuclide Mixtures Established in TBD-07-001

Building	Location	Number of Samples	Type of Samples	Uranium in Mixture	U Activity Fractions (%) (1) (2)	
					U-234	U-235
Rx Bldg	Area 17, -25 ft.	1	Cores	Yes	6.5	0
Rx Bldg	Canals E, F, G & H	4	Cores	Yes	0.16	0
Rx Bldg	-15 ft. el.	1	Cores	Yes	51.6	0
Rx Bldg	-25 ft. el.	2	Cores	Yes	2.42	0.42
Hot Lab	All Other Areas	4	Cores	Yes	0.17	0.01
Hot Lab	Hot Cells	14	Smears	Yes	0.040	0.003
Hot Lab	Hot Pipe Tunnel	5	Cores	Yes	0.48	0.05
FH	All Areas	1	Cores	Yes	16.68	0
WHB	All Areas	3	Cores	Yes	0.36	0.02
PPH	Room 4	2	Cores	Yes	3.34	0.16
PPH	All other Rooms	5	Cores	Yes	8.05	0.87
ROLB	New Fuel Vault	4	Smears	Yes	34.38	1.21
HRA	Vault & pipe Tunnel	2	Other	Yes	1.48	0.21

Table 4 Notes:

1. These are the uranium activity fractions assigned in TBD-07-001, Table 5-3.
2. The SEB Areas O/S the CPT and ROLB, All Other Rooms, are not included in the table. These areas were assigned the default mixture. The activity fraction of each radionuclide in the default mixture is a global average of the activity fractions of each radionuclide in the characterization samples from the 19 areas identified in Table 5-3 of TBD-07-001.

In view of the history of uranium storage, use and process waste handling in the PBRF buildings described in Appendix A, uranium could have been present (before remediation) in the areas listed in Table 4. During PBRF operations, uranium materials were present in significant quantities in the Reactor Building Quads and Canals, the PPH, the Hot Lab and the ROLB New Fuel Vault and Radiochemistry Laboratories. Waste streams containing uranium were present in the RB -15 ft. and -25 ft., the Fan House, WHB and HRA. Considering that most of the uranium in the PBRF was enriched, residual uranium contamination in PBRF structures would be expected to be characteristic of enriched uranium. As shown in Appendix B, the activity of enriched uranium activity is mostly due to U-234. The U-234 activity ranges from 85% to over 95% of total uranium activity, depending on the enrichment. The U-238 activity fraction is about 12% (in 4% enriched U) and less than 1% (in 93% enriched U). Thus, the activity composition of enriched uranium is distinctly different from natural uranium where the U-234 and U-238 activity fractions are equal.

In 10 of the 13 areas listed in Table 4, the uranium activity fractions in TBD-07-001, Table 5-3 were established using concrete core sample results. In one area, the HRA, the samples consisted of concrete chips and sludge. The uranium activity characteristics of individual samples used to obtain activity fractions in each of the areas (exclusive of the areas where smear samples were collected) are shown in Table 5. The information in Table 5 is obtained from vendor laboratory reports; only samples are included for which uranium activity was reported.

Table 5 includes U-238 and U-234 results (pCi/g) for 26 samples representing the 12 areas (a duplicate analysis result is included for the PPH Rm 8 sample). The table also lists the two-sigma analytical uncertainties for each result, the U-234/U-238 activity ratio and the ratio uncertainty. The activity ratios all are strongly indicative of natural uranium, i. e., a U-234/U238 ratio of one.

Table 5, Sample Uranium Analysis Results

Location	Sample ID	Data Source [1]	U-238 (pCi/g)	U-238 Uncert. (pCi/g) [2]	U-234 (pCi/g)	U-234 Uncert. (pCi/g) [2]	234/238 Act. Ratio	234/238 Uncert. [3]
CV, -25 ft. Area 17	Core Loc # 9	STL 5/14/04A	7.00E-01	2.40E-01	7.00E-01	2.40E-01	1.00	0.48
Rx Bldg Canal E	Loc # 6	STL 5/14/04A	8.80E-01	2.50E-01	7.50E-01	2.30E-01	0.85	0.36
Rx Bldg -15 Area 20	Loc # 7	STL 5/14/04	7.80E-01	2.60E-01	7.20E-01	2.40E-01	0.92	0.44
Rx Bldg -25 Area 22	Loc # 5	STL 5/14/04	6.20E-01	2.20E-01	6.40E-01	2.20E-01	1.03	0.51
Hot Lab 0 ft.	Loc # 11	STL 5/14/04A	4.40E-01	1.70E-01	3.60E-01	1.50E-01	0.82	0.46
Hot Lab 0 ft.	Loc # 12	STL 5/14/04A	8.50E-01	2.60E-01	1.04E+00	3.00E-01	1.22	0.51
Hot Lab 0 ft.	Loc # 13	STL 5/14/04A	8.20E-01	2.70E-01	9.60E-01	3.00E-01	1.17	0.53
Hot Lab 0 ft.	Loc # 14	STL 5/14/04A	6.40E-01	2.20E-01	8.10E-01	2.60E-01	1.27	0.60
Hot Lab HPT	Loc # 15	STL 5/14/04A	9.10E-01	2.80E-01	1.28E+00	3.60E-01	1.41	0.59
Hot Lab HPT	Loc # 16	STL 5/14/04A	9.10E-01	2.70E-01	9.10E-01	2.80E-01	1.00	0.43
Hot Lab HPT	Loc # SR 24-8	STL 9/7/06	5.80E-01	1.70E-01	6.30E-01	1.80E-01	1.09	0.44
Hot Lab HPT	Loc # SR 24-18	STL 9/7/06	7.00E-01	1.90E-01	8.00E-01	2.10E-01	1.14	0.43
Fan House -12 ft.	Loc # 17	STL 5/14/04A	1.02E+00	3.10E-01	1.14E+00	3.40E-01	1.12	0.48
WHB -13 ft.	Loc # 10	STL 5/14/04A	5.00E-01	1.90E-01	5.50E-01	2.10E-01	1.10	0.59
WHB -14.5 Evap. Pit	Loc SR 24-2	STL 9/7/06	6.30E-01	1.83E-01	7.00E-01	1.97E-01	1.11	0.45
PPH 0 ft. Rm 4	Loc SR 24-12	STL 9/7/06	7.70E-01	2.10E-01	7.00E-01	2.00E-01	0.91	0.36
PPH 0 ft. Rm 4	Loc # SR 24-16	STL 9/7/06	9.10E-01	1.80E-01	9.40E-01	1.80E-01	1.03	0.28
ROLB Fuel Vault	Loc # 21	STL 5/14/04	6.70E-01	2.30E-01	6.00E-01	2.10E-01	0.90	0.44
PPH 0 ft. Rm 4	SR-24-16	STL 9/7/06	9.10E-01	1.80E-01	9.40E-01	1.80E-01	1.03	0.28
PPH 0 ft. Rm 4	SR-24-15 R4 SP1-SL1	STL 5/4/07	9.70E-01	2.50E-07	9.80E-01	2.50E-01	1.01	0.26
PPH 0 ft. Rm 6	SR-24-12 R6-SP1-SL1	STL 5/4/07	9.70E-01	2.40E-01	1.00E+00	2.50E-01	1.03	0.36
PPH 0 ft. Rm 8	SR-24-11 R8-SP1-SL4	STL 5/4/07	1.06E+00	2.80E-01	8.30E-01	2.30E-01	0.78	0.30
PPH 0 ft. Rm 8	SR-24-11 R8-SP1-SL4	STL 5/4/07 DUP	6.60E-01	1.90E-01	8.50E-01	2.30E-01	1.29	0.51
PPH 0 ft. Rm 5	SR-24-13 R5-SP1-SL1	STL 5/4/07	8.50E-01	2.50E-01	7.30E-01	2.30E-01	0.86	0.37
PPH 0 ft. Rm 3	SR-24-14 R3-SP1-SL1	STL 5/4/07	1.00E+00	2.50E-01	8.00E-01	2.20E-01	0.80	0.30
HRA Vault & Pipe Tunnel	Concrete Chips	Eberline 9/13/05	5.33E-01	2.70E-01	8.26E-01	2.71E-01	1.55	0.94
HRA Vault & Pipe Tunnel	Drain Pipe Debris	Eberline 9/13/05	2.14E-01	1.20E-01	3.23E-01	1.50E-01	1.51	1.10

Table 5 Notes:

1. The vendor laboratory reports are identified by the vendor name (STL is Severn Trent Laboratories) and report date.
2. Reported as the total analytical 2-sigma uncertainty.
3. The U-234/U-238 ratio uncertainties are calculated by propagation of the sample analytical uncertainties. This underestimates the total, but unknown uncertainty, as sampling and other process errors are not accounted for.

The question is raised whether the subject concrete core uranium concentrations are different from background. Clearly the measured uranium activities (and that of other radionuclides) in smear samples were indicative of contamination of PBRF origin; this is not questioned here. The TBD-07-001 report indicates that concrete core samples are the preferred source of information for determining composition of residual contamination in the PBRF concrete structures under the assumption that surface contamination is removed during remediation. To enable comparison of the subject PBRF core sample results to concrete background uranium concentrations, Table 6 presents results from analysis of three core samples collected from Plum Brook Station reinforced concrete structures located well away from the PBRF and constructed in the early 1960s (see Appendix B for a description of the background samples).

Table 6, Uranium Background Core Sample Results

Sample ID	Sample Location	Concentration (pCi/g) (1) (2)		
		U-234 (3)	U-235	U-238 (4)
SR-363-1	Bldg. B-1	0.723 ± 0.186	<MDA (5)	0.725 ± 0.187
SR-363-2	Bldg B-3	0.965 ± 0.241	<MDA (5)	0.863 ± 0.232
SR-363-3	C-site (Bldg 2211)	0.895 ± 0.290	<MDA (5)	0.928 ± 0.296

Table 6 Notes:
1. Sample analysis results from Teledyne Brown Engineering, Inc. Nov. 18, 2011 Report for Work Order # L48463.
2. Results ± 2-sigma analytical uncertainty.
3. U-234 MDAs ranged from 2.08 to 6.40 E-02 pCi/g.
4. U-238 MDAs ranged from 3.06 to 6.40 E-02 pCi/g.
5. U-235 MDAs ranged from 1.38 to 3.34 E-02 pCi/g.

Comparison of the results in Tables 5 and 6 leads to the conclusion that the Table 5 concrete core sample uranium concentrations are not distinguishable from background, taking the uncertainties into account. Ninety-fifth % confidence intervals for the Table 5 results clearly encompass the Table 6 background sample U-234 and U-238 concentrations.

From the information presented here, it is apparent that the uranium in the Table 5 samples is natural uranium and the concentrations are indistinguishable from uranium background levels in Plum Brook Station structure concrete. It is thus concluded that uranium in the subject samples is highly unlikely to be of PBRF origin and uranium is to be removed from the radionuclide mixtures used to develop DCGLs for all areas except the ROLB New Fuel Vault.

It is noted that in the PBRF FSS Plan, U-238 was not included in the determination of activity fractions for residual contamination in structures. It was eliminated from consideration because the activity concentrations measured were generally at levels equivalent to background and it was among a group of radionuclides eliminated from further consideration because their combined dose contribution was less than 10% of the

25 mrem/y dose criterion.[5] Also, U-234 and U-235 were eliminated for the same reason from the mixtures for all areas except the ROLB; thus they are removed in the present re-evaluation from the Hot Lab, Hot Cells (see Tables A-4 and A-5, in Appendix A of the FSS Plan).

4.3 Revised Gross Activity DCGLs

As a result of the forgoing re-evaluation of radionuclide mixtures in the PBRF structure areas, revised mixtures and activity fractions have been developed. They are shown in Table 7.

Table 7, Revised Radionuclide Activity Fractions

Bldg	Location	Activity Fractions							
		H-3	Co-60	Sr-90	I-129	Cs-137	Eu-154	U-234	U-235
Rx Bldg	Area 2 & Lily Pad	0.9757	0.021	0.0013	0	0.002	0		
Rx Bldg	Area 17, -25 ft. Revised	0.943	0	0	0	0.057	0		
Rx Bldg	Quad A	0	0.7499	0	0	0.2501	0		
Rx Bldg	Quads B, C, & D	0	0.3305	0	0	0.6695	0		
Rx Bldg	Canals E, F, G & H Revised	0.012	0.117	0	0	0.871	0		
Rx Bldg	0 ft, & 12 ft el.	0.5405	0.053	0.1145	0.0009	0.2872	0.0039		
Rx Bldg	-15 ft. el. Revised	0	0	0	0	1.0	0		
Rx Bldg	-25 ft. el. Revised	0.075	0.555	0.229	0	0.124	0.018		
Hot Lab	All Other Areas Revised	0.000	0.006	0.158	0	0.836	0.000		
Hot Lab	Hot Cells Revised	0.105	0.026	0.330	0.0003	0.538	0.001		
Hot Lab	Hot Pipe Tunnel Revised	0.006	0.020	0.346	0	0.627	0.000		
SEB	Areas O/S CPT Revised (default mixture)	0.265	0.178	0.078	0.013	0.465	0.0013		
SEB	CPT	0	1	0	0	0	0		
FH	All Areas Revised	0.932	0	0	0	0.068	0		
WHB	All Areas Revised	0.005	0.002	0.026	0	0.967	0		
PPH	Room 4 Revised	0.445	0.099	0.027	0.234	0.196	0		
PPH	All other Rooms Revised	0.595	0.095	0.103	0.000	0.207	0		
ROLB	New Fuel Vault	0.3902	0.0076	0.0421	0.0287	0.1755	0	0.3438	0.0121
ROLB	All other Rooms Revised (default mixture)	0.265	0.178	0.078	0.013	0.465	0.0013		
HRA	Vault & Pipe Tunnel Revised	0.139	0.053	0.061	0	0.747	0		
WEMS	All Areas	0	0.0756	0	0	0.9244	0		

[5] In the FSS Plan, U-234 was also "deselected" from radionuclide mixtures in all areas except the ROLB. The radionuclide distribution for the ROLB was based on characterization smear samples. See FSS Plan Section 3.0 [NASA 2007].

The location column identifies those areas for which the activity fractions are revised. The revised activity fractions are calculated by removing the uranium activity fractions from all areas except the New Fuel Vault and "renormalizing" the remaining nuclide activity fractions by calculating the ratio of the "old" activity fraction to the sum of the activity fractions of the remaining radionuclides. Then the sum of the "new" activity fractions for each location then totals to one. The two locations in Table 7 identified as having default radionuclide mixtures are the SEB, Areas Outside of the Cold Pipe Tunnel and the ROLB, All Other Rooms. The activity fraction for each radionuclide in the default mixture is calculated as the average of the radionuclide fractions in the other areas, exclusive of the New Fuel Vault. The default mixture (and DCGL) are also assigned in TBD-07-001, Appendix L, to PBRF building exteriors, outbuildings and other miscellaneous structures and pads.

Determination of the revised radionuclide mixtures and activity fractions allows revision of the gross beta activity DCGLs using the corrected method of calculation as outlined in Section 4.1. The results are presented in Table 8.

Table 8, Revised PBRF Structure Gross Beta Activity DCGLs

Bldg	Location	Detectable Act. Fraction	TBD-07-001 Table 5-3 $DCGL_{GB}$	Revised $DCGL_{GB}$	% change
			(dpm/100-cm^2)		
Rx Bldg	Area 2 & Lily Pad	0.0243	11563	11544	-0.2%
Rx Bldg	Area 17, -25 ft. Revised	0.0567	37235	37707	1.3%
Rx Bldg	Quad A	1.0000	13450	13450	0.0%
Rx Bldg	Quads B, C, & D	1.0000	21566	21470	-0.4%
Rx Bldg	Canals E, F, G & H Revised	0.9884	31711	30734	-3.1%
Rx Bldg	0 ft, & 12 ft el.	0.4586	29060	28040	-3.5%
Rx Bldg	-15 ft. el. Revised	1.0000	35296	40500	14.7%
Rx Bldg	-25 ft. el. Revised	0.9250	14600	14382	-1.5%
Hot Lab	All Other Areas Revised	1.0000	38647	38538	-0.3%
Hot Lab	Hot Cells Revised	0.8952	34404	34583	0.5%
Hot Lab	Hot Pipe Tunnel Revised	0.9940	35781	35747	-0.1%
SEB	Areas O/S CPT Revised (default mixture & DCGL)	0.7054	27166	23146	-14.8%
SEB	CPT	1.0000	11000	11000	0.0%
FH	All Areas Revised	0.0676	36857	38157	3.5%
WHB	All Areas Revised	0.9948	40051	40090	0.1%
PPH	Room 4 Revised	0.3217	11186	10588	-5.3%
PPH	All other Rooms Revised	0.4047	26348	23914	-9.2%
ROLB	New Fuel Vault	0.2252	30831	11475	-62.8%
ROLB	All other Rooms Revised (default mixture & DCGL)	0.7054	27166	23146	-14.8%

Table 8, Revised PBRF Structure Gross Beta Activity DCGLs

Bldg	Location	Detectable Act. Fraction	TBD-07-001 Table 5-3 $DCGL_{GB}$	Revised $DCGL_{GB}$	% change
			(dpm/100-cm^2)		
HRA	Vault & pipe Tunnel Revised	0.8615	34213	34267	0.2%
WEMS	All Areas	1.0000	33834	33673	-0.5%

It is seen in Table 8 that the major changes in gross beta activity DCGL values from the TBD-07-001 values occur in the Reactor Building -15 ft. (14.7%) and the New Fuel Vault (-62.8%). The effect of the corrected DCGL calculation is not seen in the new value for the Reactor Building -15 ft. because the removal of uranium from the mixture leaves only Cs-137 in the mixture and the DCGL reverts to the single nuclide Cs-137 DCGL, 40,500 dpm/100-cm^2. The new DCGL value for the New Fuel Vault is entirely due to the corrected method of calculating the gross beta activity DCGL, as the mixture and activity fractions are not changed.

5.0 Conclusions

The method for calculating the DCGLs presented in TBD-07-001 Table 5-3 was re-evaluated in response to an ORISE finding following their July 2011 confirmatory survey and review of PBRF FSS records. The ORISE finding was that the gross beta activity DCGL calculated for the Reactor Building -15 ft. elevation did not properly incorporate the surrogate DCGL where Cs-137 is used as the surrogate for U-234. The recalculated DCGLs are presented in Table 8. The revised DCGLs also incorporated changes in the radionuclide mixtures and activity fractions which resulted from a re-evaluation of the concrete core sample uranium analysis results used in TBD-07-001 to establish the activity fractions.

From this re-evaluation, it is concluded that:

1. Considering that most of the uranium in the PBRF was enriched uranium, the isotopic composition of characterization samples collected from "uranium areas" should be consistent with that of enriched uranium. There is no evidence of enriched uranium in concrete core samples used to establish radionuclide mixtures for determination of structure DCGLs. The core sample results all are indicative of natural uranium at levels consistent with concrete uranium background concentrations.

2. The exception to this is the smear samples collected from the ROLB New Fuel Storage Vault. The U-235 to U-234 activity ratio calculated from these sample results is 0.0349 (3.49%); this is consistent with enriched uranium: a U-235/U-234 activity ratio of 3.5%.

6.0 Appendices

Appendix A – Source Material and SNM at PBRF

Appendix B – Uranium Primer

Appendix C – Revised DCGL Calculations

Appendix A –Source Material and SNM at PBRF

Summary

During the operational history of the PBRF, from 1961 through 1973, a wide variety of source and special nuclear materials (SNM) were handled and stored at the site. Areas of the PBRF where these materials were stored and handled could be expected to have radionuclide contamination profiles different from those in the balance of plant (BOP). These materials included uranium in several forms and other alpha-emitting radionuclides. A review was conducted of PBRF radioactive material inventory and transfer records to identify quantities of source and special nuclear materials on the site at the time of permanent shutdown of the facility in January 1973 [PBRF 2011a]. The principal source material and SNM are identified and quantities summarized:

- New fuel elements (93% enriched in U-235) stored in New Fuel Vault in ROLB.
- Spent fuel elements – in Canal G storage racks, 60 MW Test Reactor and MUR cores (328 elements).
- Neutron monitoring fission chambers (encapsulated U-235) – 16 items including three different fission chamber models.
- Uranium metal foils – 1 mil thick sheets (enriched in U-235).
- 100 milligrams of U-233 in uranyl nitrate solution.
- Ten UO_2 source material experimental pellets.
- Several Pu-239 sources (in encapsulated Pu-Be startup sources).
- Approximately one kg of depleted uranium (in UO_2 encapsulated pellets).
- Uranium fuel pellets, foils and pins; numerous items of various enrichments (from 5 to 98% - most were highly enriched) for experimental or analytical use.
- Thorium-232 foils (15 g).

These materials were routinely stored and handled in the Hot Laboratory and the ROLB. Irradiated fuel elements were stored in Canal G in the Reactor Building. Sources were also temporarily handled (and stored) in the Reactor Building and Containment Vessel (CV) experimental areas. New fuel elements were stored in the ROLB New Fuel Vault and new fuel elements were tested in a test loop located in the Primary Pump House, Room 7. Summaries of materials handled and stored in these areas follow. In addition to the discrete sources identified above, uranium and transuranics isotopes were released from leaking fuel elements to the primary cooling water.

Hot Laboratory

Irradiated and non-irradiated source and special nuclear materials were processed in the Hot Laboratory. Two irradiated PBRF Reactor fuel elements (each containing 168 grams of U-235) were placed in the hot cells and the Hot Dry Storage Area to verify the integrity of the shielding walls in 1964. The results showed that a fuel element source, producing a dose rate of 10^6 R/hr at one foot, could be handled in any of the hot cells and the Hot Dry Storage Area without exceeding dose rate limits in the operating areas. The Hot Laboratory was then utilized for its intended purposes. In 1968, several 240 gram (U-235) irradiated fuel elements were disassembled in Hot Cell No. 1 to identify leaking fuel plates.

During the operating period from 1963-1973, an estimated 384 grams of U-235 were included in the large quantity of radioactive material processed through the Hot Laboratory. Source and special nuclear materials were associated with various PBRF experiment programs; the majority of the material was experimental fuel specimens. About half was consumed in examination and about 10 per cent was lost in cutting and drilling. Of the cuttings and drillings (approximately 19 grams), at least half was vacuumed up, filtered out or processed as waste. Less than 10 grams of U-235 were calculated as lost down the drains. As the Hot Laboratory sump was cleaned several times over the operating period, only a small amount of U-235 remained at shutdown [PBRF 2009].

Reactor Office and Laboratory Building

A wide variety of source and special nuclear materials in the form of liquids, gases and solids were stored, handled in the ROLB and analyzed in the ROLB laboratories during the operations period. Typically, radioactive materials were transported from the Reactor Building, Containment Vessel, Hot Laboratory, Primary Pump House and other PBRF locations to the radiochemistry and metallurgy laboratories in the ROLB. Items such as primary reactor coolant samples, flux wires from the Plum Brook Test Reactor and the Mock-up Reactor (MUR), PBRF reactor vessel material specimens, small experiment specimens (such as irradiated space-related experiment samples, moon rocks, corn, coal, petroleum samples, etc.) were routinely carried in lead casks or other shielded containers for counting and analysis in the ROLB labs.

Radionuclides recorded on ROLB inventory sheets included Po-210, mixed fission products, Radium, Uranium (natural, depleted and enriched), Am-Be neutron sources and Np-237. Most of the source inventory was disposed as radioactive waste during operations (1961-73) and during the transition to "mothball" status in 1973. The remainder was transferred to other licensees, returned to the AEC, or stored on-site for potential future use [PBRF 2009a].

Reactor Building

Radioactive materials in the Reactor Building were those associated with PBRF operations, tests and experiments, including irradiated highly enriched uranium fuel elements stored in Canal G. Handling of radioactive materials in the Reactor Building usually involved the use of shielding casks and underwater manipulations. Long handling tools were used to move spent nuclear fuel elements and to load and unload fuel elements and experiments in both the 60 MW Test Reactor and the MUR. These tasks included cleaning hot sumps, decontamination work and maintenance on both reactors. Many tasks required safe work permits due to the potential for contamination and radiation exposure, e.g., refueling and loading spent fuel for shipment (most of these required the use of personnel protective clothing and respiratory protection equipment). Frequent shielded transfers of primary cooling water samples were made from the PPH to the ROLB through the Reactor Building. Shielded transfers of irradiated rabbits or specimens such as flux wires were made to the ROLB laboratories through the RB from the MUR, CV or HL. Such activities did occasionally result in unintended spills of radioactively contaminated materials in RB areas [PBRF 2009b].

Reactor Containment Vessel

Radioactive contamination in the CV was of two primary origins: 1) irradiated test specimens and associated hardware and 2) releases of airborne radioactive materials and contaminated liquids from the Reactor Tank and process systems. Most of the irradiated test specimens did not remain in the CV; they were transferred to other buildings in the PBRF, primarily the Hot Laboratory. Experiment transfers were occasionally conducted overland significantly increasing general radiation levels in the CV.

Leaking valves or contaminated water dripping from handling tools led to occasional surface contamination incidents. Experimental work led to gaseous and particulate releases into the CV which included activation products, fission products including Sr-90 and Cs-137 and U-235 [PBRF 2009c].

Primary Pump House

Radioactive materials (primary cooling water, resins and other samples) were routinely transported from the PPH to the radiochemistry and metallurgy laboratories in the ROLB. For example, primary reactor coolant samples were routinely carried from Room 8 in lead casks or other shielded containers for counting and analysis in the ROLB radiochemistry labs. This process occasionally led to minor spills. In addition, contamination occurred in various locations from primary cooling water (PCW) valve leakage, PCW main and auxiliary pump seal leakage or failure, and maintenance activities such as PCW strainer cleanout and pump replacement. The PPH was designed and operated to confine and control leaks and spills from potential sources (PCW water leakage, ventilating air, spent resins, etc.). Wastes from leaks and spills were contained and directed to hot storage facilities and systems where they were sampled, monitored and treated to ensure compliance with AEC/NRC regulations prior to release [PBRF 2009d].

New fuel elements were handled and tested in Room 7 of the PPH. The Fuel Element Test Loop (FETL) was designed to test the mechanical integrity of new fuel elements and shim rod fuel sections. This was accomplished by placing individual elements in a test section of the loop and subjecting them to the simulated thermal and hydraulic environment of the 60 MW Test Reactor. During the period of PBRF operations, hundreds of fuel elements were transported to the PPH and tested in the FETL.[6] Two pathways are identified for contamination from new fuel element handling and testing in PPH Room 7. Initially, liquid wastes generated in fuel handling and testing were collected in 55 gallon drums which were then emptied into the PPH Room 8 floor drain. This drain was converted into a hot drain which fed to the -15 ft. Reactor Building sump. Liquid wastes also entered the Room 7 floor drain which was also routed to the -15 ft. RB sump.

[6] Approximately 10 to 11 fuel elements were replaced at each operating cycle and 152 cycles were completed during the period of PBRF operations. This does not include MUR fuel elements which were changed infrequently.

Appendix B – Uranium Primer

This Appendix provides information to aid in interpreting radiological survey measurements and sample analysis results from areas in the PBRF where isotopes of uranium are potentially present in residual contamination. As shown in Appendix A, a wide variety of radioactive materials containing uranium and other long-lived high-atomic number radionuclides were stored and handled at the PBRF. Information is presented on:

- Uranium Isotopes and decay chain,
- Activity fractions of natural uranium, low enriched, high enriched and depleted uranium,
- Radionuclides in irradiated fuel and
- Uranium background concentration in concrete construction materials.

Uranium Isotopes and Decay Chain

Uranium in nature consists mostly of U-238 and its decay series members and a small fraction of U-235. The principal uranium isotopes are shown in Figure B-1. The U-238 decay series is shown in Figure B-2. When uranium ore is processed for preparation of reactor fuel, weapons or other uses, the decay series daughters are removed and only the principal uranium isotopes remain. However, after processing, in-growth of the U-238 decay series members begins anew and after about 9 months, the first two members, 24 day Th-234 and 1.2 minute Pa-234m, both beta emitters, are in secular equilibrium with U-238 [Abel 1997].

Figure B-1, Principal Uranium Isotopes

Summary of Uranium Isotopes (1)					
Isotope	**Percent in natural uranium (wt%)**	**No. of Protons**	**No. of Neutrons**	**Half-Life (years)**	**Specific Activity (Ci/g) (3)**
Uranium-238 (2)	99.284	92	146	4.46 billion	3.35E-07
Uranium-235 (2)	0.711	92	143	704 million	2.16E-06
Uranium-234 (2)	0.0055	92	142	245,000	6.24E-03

Figure B-1 Notes:

1. Adapted from IEER Factsheet|Uranium, http://www.ieer.org.
2. The percent (by weight) of the isotopes in natural uranium reported in various references varies slightly.
3. Specific activities from Handbook of Health Physics and Radiological Health [Schleien 1997].

Figure B-2, Uranium Series Decay Chain

URANIUM DECAY CHAIN -- Main Branch [1] Read from left to right. Arrows indicate decay		
Uranium-238 ==> (half-life: 4.46 billion years) alpha decay [2]	Thorium-234 ==> (half-life: 24.1 days) beta decay	Protactinium-234m ==> (half-life: 1.17 minutes) beta decay [3]
Uranium-234 ==> (half-life: 245,000 years) alpha decay [2]	Thorium-230 ==> (half-life: 75,400 years) alpha decay [2]	Radium-226 ==> (half-life: 1,600 years) alpha decay
Radon-222 ==> (half-life: 3.82 days) alpha decay	Polonium-218 ==> (half-life: 3.11 minutes) alpha decay	Lead-214 ==> (half-life: 26.8 minutes) beta decay
Bismuth-214 ==> (half-life: 19.9 minutes) beta decay	Polonium-214 ==> (half-life: 163 microseconds) alpha decay	Lead-210 ==> (half-life: 22.3 years) beta decay
Bismuth-210 ==> (half-life: 5.01 days) beta decay	Polonium-210 ==> (half-life: 138 days) alpha decay	Lead-206 (stable)

Figure B-2 Notes:

1. From IEER Factsheet|Uranium, http://www.ieer.org.
2. The half-lives of the long-lived members vary slightly as reported in other references.
3. Not shown is the 0.13% decay branch via 6.7 h Pa-234 (also a beta emitter).

Uranium Activity Fractions

As shown in Figure B-1, natural uranium is comprised of over 99% U-238, with less than 1% U-235 and about 0.5% U-234. However, when considering radioactivity fractions instead of mass fractions, a distinctly different picture emerges. This is because the specific activity of U-234 is 18,600 times that of U-238 and 2,890 times that of U-235. Thus, even though the wt% of U-234 in natural uranium is only about 0.005%, the activity fraction is 48.9%. And while U-238 comprises over 99 wt% of natural uranium, it comprises only 48.9% of the activity.

Table B-1 compares mass and activity fractions for natural uranium, enriched uranium and depleted uranium. The table includes mass and activity fractions for enrichment percentages: 4%, 5% and 93%. It is noted that while the uranium inventory at the PBRF included natural, low enriched, highly enriched and depleted uranium, most was highly enriched uranium. It is seen from the table, given that the majority of uranium in the PBRF was highly enriched, the activity is mostly U-234. Only when natural or depleted uranium are the principal contaminants, would U-238 activity be equal or greater than that of U-234. In all cases, U-235 constitutes 3.4% or less of the total uranium activity.

Table B-1, Uranium Mass and Activity Fractions

Uranium Description	U-234 [1]		U-235 [1]		U-238 [1]	
	Weight %	Activity %	Weight %	Activity %	Weight %	Activity %
Natural U [2]	0.0053	48.9	0.72	2.20	99.28	48.9
4% Enriched [3]	0.036	84.6	4.009	3.26	95.96	12.13
5% Enriched [3]	0.047	87.3	5.01	3.21	94.94	9.49
93% Enriched [3]	0.924	96.6	93.24	3.37	5.83	0.003
Depleted U [4]	0.0017	23.7	0.3	1.45	99.7	74.8

Table B-1 Notes:

1. Weight and activity fractions are calculated using the WISE Uranium Project calculators (http://www.wise-uranium.org).
2. The isotopic composition of natural uranium varies slightly depending on the origin of the uranium ore.
3. Isotopic weight (and activity) fractions in enriched uranium vary slightly depending on the uranium feedstock (natural ore or recycled uranium), the method of enrichment and other variables. See the WISE Project web site for additional details.
4. Isotopic composition and weight fractions of depleted uranium can vary significantly (within a range) depending on the original feedstock (multiple feed stocks are possible) and enrichment process parameters. See Note 3.

It is useful to show the data in Table B-1 as ratios of U-234 and U-235 to U-238 activity and U-235 to U-234 activity. This is done in Table B-2; these activity ratios are sometimes referred to as the "uranium signature" of a uranium sample or material. The origin of the uranium in a sample can usually be readily determined by examination of these activity ratios. In natural uranium the U-234 and U-238 activities are equal. Whereas in enriched uranium, the U-234 activity predominates; 7 to 9 times the U-238 activity in low-enriched uranium and over 32,000 times in highly enriched uranium. Only in depleted uranium is the U-234 activity significantly less than the U-238 activity.

Table B-2, Uranium Signatures

Uranium Description	U-Isotopic Activity Ratios		
	234/238	235/238	235/234
Natural U	1.00	0.04	0.045
4% Enriched	6.97	0.27	0.039
5% Enriched	9.20	0.34	0.037
93% Enriched	32200	1123	0.035
Depleted U	0.32	0.02	0.061

Radionuclides in Irradiated Fuel

Alpha emitting radionuclides in irradiated fuel at the PBRF are produced by neutron capture reactions in U-238 and to a lesser extent in U-235 contained in the fuel. Even though irradiation times for a typical operating cycle were relatively short, typically about two weeks, transuranic radionuclides were produced. While information on assay of transuranics in irradiated PBRF 60MW Test Reactor fuel is not available, information in the open literature indicates that in addition to the original uranium isotopes in the fuel, transuranic activity 30 years after shutdown includes Np-237, Pu-238, Pu-239, Pu-240 and Pu-242 with lesser amounts of Am-241 and Cm-242. Uranium-236 is also present, with activity comparable to U-238 [Pigford 1981].

Natural Uranium in Concrete

It is well known that natural uranium is a constituent of concrete, where the U-238 daughters, and K-40, contribute most of the background counts to beta detectors commonly used for FSS surface activity measurements. To provide information for comparison of uranium analysis results from concrete cores collected in the PBRF, concrete core samples were recently collected from three Plum Brook Station facilities. The facilities selected were located on the Plum Brook Station, but well away from the PBRF and were constructed in approximately the same time frame as the PBRF (late 1950's through the 1960's). The core samples were sent to a qualified vendor laboratory for analysis. The analysis request was for gamma spectroscopy and uranium isotopic: U-234, 235 and 238. The results are presented in Table B-3 [Tele 2011]. From the table it is seen that the measured Plum Brook Station concrete uranium background activity is constituted of U-234 with average concentration of 0.86 ± 0.26 pCi/g and U-238 with average concentration of 0.84 ± 0.20 pCi/g (two standard deviations) and U-235 with concentration < 0.03 pCi/g. As shown in Tables B-1 and B-2, the U-234 and U-238 activity concentrations in natural uranium are equal; this is confirmed by the sample results shown in Table B-3. The U-235 concentration in natural uranium is 2.2 % of the total uranium activity or 4.5 % of the U-234 or U-238 activity concentration. This is equivalent to about 0.038 pCi/g in the concrete background samples. This is approximately equal to the laboratory analysis method detection limit reported for these samples.

Table B-3, Background Uranium Sample Results

Sample ID	Sample Location	Concentration (pCi/g) (1) (2)		
		U-234 (3)	U-235	U-238 (4)
SR-363-1	Bldg. B-1	0.723 ± 0.186	<MDA (5)	0.725 ± 0.187
SR-363-2	Bldg B-3	0.965 ± 0.241	<MDA (5)	0.863 ± 0.232
SR-363-3	C-site (Bldg 2211)	0.895 ± 0.290	<MDA (5)	0.928 ± 0.296

Table B-3 Notes:

1. Sample analysis results from Teledyne Brown Engineering, Inc. Nov. 18, 2011 Report for Work Order # L48463.
2. Results ± 2-sigma analytical uncertainty.
3. U-234 MDAs ranged from 2.08 to 6.40 E-02 pCi/g.
4. U-238 MDAs ranged from 3.06 to 6.40 E-02 pCi/g.
5. U-235 MDAs ranged from 1.38 to 3.34 E-02 pCi/g.

Appendix C – DCGL Calculation Template

This appendix contains an image of the EXCEL template used to calculate gross beta activity DCGLs. It uses the method described in Section 2.0 of the body of this report. The data shown in the template range C8:J28 are the revised radionuclide activity fractions shown in Table 7 of this report. The "new" default activity fractions are shown in Row 19 (C19:H19) and Row 26 (C26:H26). They are calculated in this template as the average of the individual activity fractions assigned to all the other areas, excluding the ROLB New Fuel Vault (see the example cell formula in C19). The original gross beta activity DCGLs from TBD-07-001, Table 5-3 are shown in Column M (M8:M28). The Column N cells N8:N28 contain the EXCEL formula for calculating the surrogate DCGL, wherein Cs-137 is used as the surrogate for H-3, I-19, U-234 and U-235. This is the EXCEL formula for Equation 1 in Section 2.0. An example formula is shown for cell N8. The Column O cells O8:O28 contain the EXCEL formula for calculating the gross beta activity DCGL. This is the EXCEL formula for Equation 2 in Section 2.0. An example formula is shown for cell O8. The cell formulas in Column P (P8:P28) calculate the difference (% change) between the revised gross beta DCGLs and the original TBD-07-001, Table 5-3 values. An example cell formula is shown for cell P8.

	A	B	C	D	E	F	G	H	I	J	K	L	M	N	O	P
5																
6	Bldg	Location	Activity Fractions								Sum Check	Detectable Act. Fraction	Table 5-3 $DCGL_{GB}$	Revised DCGL	Revised $DCGL_{GB}$	% change
7			H-3	Co-60	Sr-90	I-129	Cs-137	Eu-154	U-234	U-235			(dpm/100-cm²)			
8	Rx Bldg	Area 2 & Lily Pad	0.9757	0.021	0.0013	0	0.002	0			1.00	0.0243	11563	12771	11544	-0.2%
9	Rx Bldg	Area 17, -25 ft. Revised	0.943	0	0	0	0.057	0			1.00	0.0567	37235	37707	37707	1.3%
10	Rx Bldg	Quad A	0	0.7499	0	0	0.2501	0			1.00	1.0000	13450	40500	13450	0.0%
11	Rx Bldg	Quads B, C, & D	0	0.3305	0	0	0.6695	0			1.00	1.0000	21566	40500	21470	-0.4%
12	Rx Bldg	Canals E, F, G & H Revised	0.012	0.117	0	0	0.871	0			1.00	0.9884	31711	40498	30734	-3.1%
13	Rx Bldg	0 ft, & 12 ft el.	0.5405	0.053	0.1145	0.0009	0.2872	0.0039			1.00	0.4586	29060	39827	28040	-3.5%
14	Rx Bldg	-15 ft. el. Revised	0	0	0	0	1.000	0			1.00	1.0000	35296	40500	40500	14.7%
15	Rx Bldg	-25 ft. el. Revised	0.075	0.555	0.229	0	0.124	0.018			1.00	0.9250	14600	40391	14382	-1.5%
16	Hot Lab	All Other Areas Revised	0	0.006	0.158	0	0.836	0			1.00	1.0000	38647	40500	38538	-0.3%
17	Hot Lab	Hot Cells Revised	0.105	0.026	0.330	0.0003	0.538	0.001			1.00	0.8952	34404	40404	34583	0.5%
18	Hot Lab	Hot Pipe Tunnel Revised	0.006	0.020	0.346	0	0.627	0			1.00	0.9940	35781	40498	35747	-0.1%
19	SEB	Areas O/S CPT Revised, Default	0.265	0.178	0.078	0.013	0.465	0.0013			1.00	0.7218	27166	37542	23146	-14.8%
20	SEB	CPT	0	1	0	0	0	0			1.00	1.0000	11000	11000	11000	0.0%
21	FH	All Areas Revised	0.932	0	0	0	0.068	0			1.00	0.0676	36857	38157	38157	3.5%
22	WHB	All Areas Revised	0.005	0.002	0.026	0	0.967	0			1.00	0.9948	40051	40499	40090	0.1%
23	PPH	Room 4 Revised	0.445	0.099	0.027	0.234	0.196	0			1.00	0.3217	11186	9530	10588	-5.3%
24	PPH	All other Rooms Revised	0.595	0.095	0.103	0	0.207	0			1.00	0.4047	26348	39987	23914	-9.2%
25	ROLB	New Fuel Vault	0.3902	0.0076	0.0421	0.0287	0.1755	0	0.3438	0.0121	1.00	0.2252	30831	9936	11475	-62.8%
26	ROLB	All other Rooms Revised, Default	0.265	0.178	0.078	0.013	0.465	0.0013			1.00	0.7218	27166	37542	23146	-14.8%
27	HRA	Vault & pipe tunn Revised	0.139	0.053	0.061	0	0.747	0			1.00	0.8615	34213	40467	34267	0.2%
28	WEMS	All Areas	0	0.0756	0	0	0.9244	0			1.00	1.0000	33834	40500	33673	-0.5%

29			
30	**Example Cell Formulas**		
31	Cell	Formula	Comment
32	C19	=AVERAGE(C8,C9,C10,C11,C12,C13,C14,C15,C16,C17,C18,C20,C21,C22,C23,C24,C2	Calculates activity fractions for default radionuclide mixture.
33	L8	=(D8+E8+G8+H8)/SUM(C8:J8)	Detectable activity fraction; sum of Co-60, Sr-90, Cs-137 & Eu-154 ÷ sum of all activity fractions.
34	N8	=1/((1/$G$4)+((C8/G8)/$C$4)+((F8/G8)/$F$4)+((I8/G8)/$I$4)+((J8/G8)/$J$4))	Surrogate DCGL calculation; Equation 1 in Section 2.1 of this report.
35	O8	=L8/((G8/N8)+(D8/$D$4)+(E8/$E$4)+(H8/$H$4))	Gross activity DCGL calculation; Equation 2 in Section 2.1 of this report.
36	P8	=(O8-M8)/M8	Calculates % diffference between revised gross activity DCGL and TBD-07-001 value.