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Reference Fuel Assembly for Dry Storage Demonstration of L Basin Spent Fuel

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1.0 SUMMARY

A “Reference Fuel Assembly – Dry” has been constructed for planning and designing of a system for a dry storage demonstration and its safety evaluation at the Savannah River Site. This “RFA-Dry” is based on fuel with the Materials Test Reactor Equivalent design presently in, or expected to be received in the L Basin. The RFA-Dry is a hypothetical fuel based on fuel in L Basin bundled storage with composite attributes including:

- the present L-Area Reference Fuel Assembly (RFA) that bounds the radiological hazard for incoming fuel to the site and that is considered in the safety basis for fuel in wet storage [1, 2];
- additional bounding features of the spent fuel, on a single assembly basis, from the Appendix A information for the fuel received at the site [3]; and
- additional bounding features of the spent fuel not listed in the Appendix A information including oxyhydroxides on the fuel, the heat generation rate, and the areal fraction of exposed fuel core of failed fuel.

The RFA-Dry attributes would be considered in designing the system(s) and in evaluating the safety of fuel that would be handled, dried, and loaded in sealed canister storage, and would remain retrievable from a dry storage demonstration that would be located at the site. Fuel-specific attributes, whole or partial, could be used in lieu of the attributes of the RFA-Dry assembly, if justified, for placement in a dry storage system.

Table 1 below contains the bounding attributes of the RFA-Dry with full consideration of degradation from reactor irradiation and wet storage. The metrics are not necessarily congruent, and the value used is dependent on the application. For example, the heat source term is not derived from the isotopic contents that is used for the radiological hazard source term, but rather from an analysis of actual fuel [4]. Similarly, the Beginning of Life fissile material content of the fuel is the attribute suggested for nuclear criticality safety analysis.

It is emphasized that no one assembly has the composite attributes of the RFA-Dry. It may not be feasible to design a sealed canister for a dry storage demonstration that would account for load of fuel at the bounding metrics of the RFA-Dry. Nevertheless, the RFA-Dry would allow for fuel selection and overall design considerations in the development of the system(s) for a dry storage demonstration for L Basin fuel.

An inventory of non-aluminum clad fuel presently stored in L Basin is also available for the dry storage demonstration. The radiological characteristics and corrosion resistance of this fuel are expected to be well within the bounding attributes of the RFA-Dry. It is recommended that attributes of specific fuel selected from this inventory for use in the dry storage demonstration should be evaluated against the design features and safety basis assumptions at the time of fuel selection.

Table 1 Reference Fuel Assembly for Dry Storage of Fuel from L Basin – MTRE Design

Attribute Category	Application to SRS Dry Storage Demonstration Project	Metric
Fuel Design[a]	**General fuel characteristics for basket design and canister loading**	**Materials Test Reactor Equivalent -with 19 fuel plates**
	Dimensions	Bounded cropped: 65 cm L x 8.5 cm x 8.5 cm (typical is 63.5 cm L x 7.73 cm W x 7.73 cm W, with a total (aluminum) surface area (typical) of 1.746 m^2 [5])
	Weight	Bounding 10kg (typical 3-7kg for MTRE design fuel)
	Materials	Aluminum alloys for plate cladding and end plates. Stainless steel screws for fuel assembly. Fuel core materials are dispersoids of UAl_x, U_3Si_2, or U_3O_8 in aluminum.
Fuel Isotopic Contents[b]	**Radioisotopes for: accident source term; shielding; heat load; NCS**	**Radioisotope content - dependent on application**
	Radiological hazard (source term) for accident analysis	Listed under the L Basin RFA [1, 2]
	Shielding hazard for handling and shielding design	HFBR fuel for gamma flux and spontaneous neutron flux [6]
	Heat load for handling, drying, and dry storage system design	25 watts [4] (3-years post-discharge of bounding fuel)
	Nuclear criticality safety analysis	410 grams U-235 per assembly with 93% enriched uranium (bounds MTRE fuel per Appendix A information [3]) at beginning of life
Fuel Condition Post-Discharge and Storage[c]	**Describes spent fuel and challenges to interim dry storage in terms of confinement, retrievability, and other safety considerations of radiolytic gas generation from chemically-bound water**	**Degree of corrosion - in terms of metal consumption, including exposed fuel core (meat) material, and oxide corrosion product presence**
	Oxyhydroxide Film - Boehmite	50 μm thick adherent layer on all aluminum surface area (typical) for a total mass per assembly of 284 grams
	Oxyhydroxide Deposits – Gibbsite	Total mass of Gibbsite per assembly of 5 grams
	Cladding Average Thickness (Post-irradiation)	Average initial (design) clad thickness per Appendix A data with is 0.038 cm. An average metal loss of 0.0038 cm (corresponds to a film of Boehmite of 50 μm) is a bounding estimate for spent fuel
	Total Exposed Fuel Meat	5% total fuel meat

[a]The metrics of the fuel design can be changed with cropping and disassembly of the MTRE. The dimension is nominal for a cropped assembly. The dimensions, weight, materials of the MTRE are available in Appendix A information [3]
[b]The L Basin RFA [1, 2] is the present reference fuel in the L-Area DSA and provides the Radiological Hazard. The HFBR remaining metric are bounding attributes of the fuel in L Basin used in shielding analysis of the Shielded Transfer System
[c]The MTRE aluminum materials are corroded and altered from the design condition by virtue of reactor operation and also post-discharge storage where oxide formation is dependent on the water quality.

2.0 INTRODUCTION

Fuel from the L Basin at the Savannah River Site is being considered for a dry storage demonstration project in which the fuel would be placed in canisters, dried, sealed, and potentially backfilled with helium. The isotopic and physical condition of the fuel must be considered in the handling, drying, and dry storage. Site safety and general safety functions defined for dry storage must be maintained.

The predominant fuel design in L Basin is the Materials Test Reactor Equivalent. Figure 1 shows the MTRE sketch. An assemblage of the bounding characteristics or attributes of an MTRE fuel is developed in this report.

The isotopic content of a fuel assembly is part of the consideration in accident scenarios, in shielding during both handling and in a dry storage cask system, and in nuclear criticality safety analysis. Further, the heat load from an assembly is a critical parameter for dry storage system design to limit temperatures to within acceptable for safe storage.

The physical condition is important since fuel cladding breaches will impact the source term for confinement in dry storage. Nevertheless, it is feasible that failed fuel can be directly placed in a dry storage demonstration. Also, aluminum oxyhydroxides present on even fully dried fuel would present a challenge to safety of the storage system through radiolytic gas production.

Other non-aluminum fuels including stainless steel-clad and zircaloy-clad fuel are in bundled storage in L Basin and would be available for a dry storage demonstration. These fuels are expected to be well within the envelope of attributes of the aluminum fuel in terms isotopics, and they are intrinsically much more resistant to corrosion degradation. Verification of the bounding attributes of the MTRE against this inventory was not performed in the scope of this report. It is suggested that non-aluminum fuel selected from the L Basin inventory be evaluated to identify the attributes against the design and safety basis at the time of fuel selection.

The water associated with a fuel assembly would be considered on the canister basis to be dried, and is outside the scope of this report.

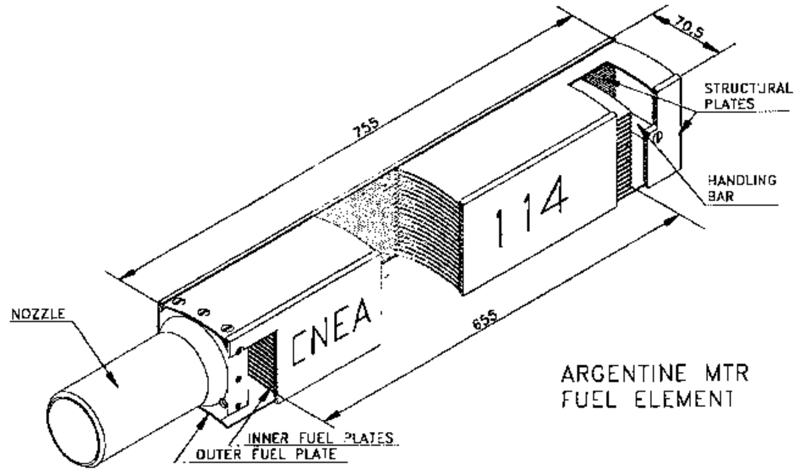

Figure 1 Material Test Reactor Equivalent Design

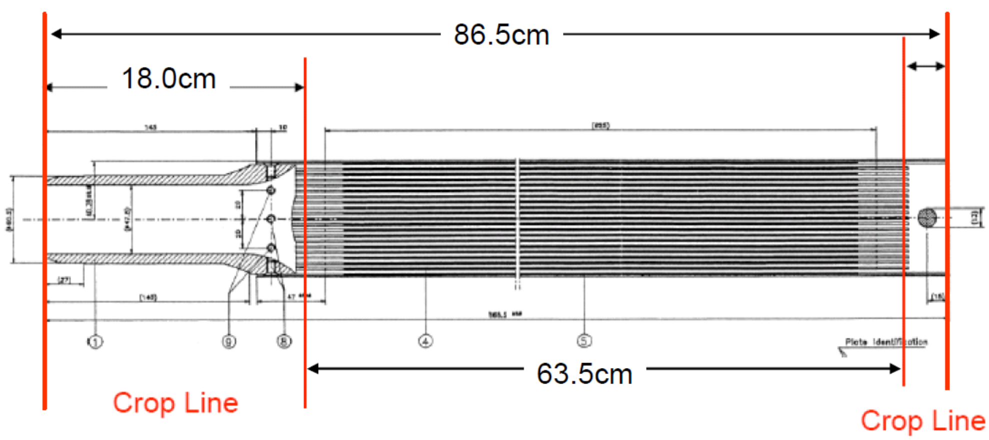

Figure 2 Dimensions of MTRE showing the cropped length (63.5 cm) for L Basin bundled fuel storage

3.0 CHARACTERISTICS OF FUEL RECEIVED AT SRS – APPENDIX A

Spent Nuclear Fuel (SNF) characteristics information is needed for transport, storage and disposal systems for evaluation of system operation, safety, and environmental impact. Characteristics information for each spent nuclear fuel assembly to be received at and stored in pool storage at the U.S. Department of Energy (DOE) Savannah River Site (SRS) is compiled in an "Appendix A" document for the fuel [3]. The Appendix A document for a fuel assembly (collection of the fuel elements, e.g., plates into a fuel unit or assembly) aims to provide the accurate and pertinent information concerning the post-reactor-operation characteristics of the fuel assembly.

The Appendix A document includes a detailed description of each element, rod or plate, that makes up a fuel assembly as well as a detailed description of the assembly as a whole. This includes geometric and mass information including fuel meat mass, composition, and geometry for each element that comprise a full assembly. Cladding material mass, composition, and geometry are also included for each element. The full assembly description includes geometric and mass information on additional materials, such as spacers, dummy plates, thermocouples, fittings, etc., that are included in the full assembly.

Additional information provided in a typical Appendix A document includes fuel identification numbers, the fuel irradiation history, and some limited post irradiation information. The irradiation history information includes assembly power level, exposure duration, burnup (in percent), and cooling time. The post irradiation information includes mass of several actinides including the isotopes of uranium and plutonium. The assembly activity and decay heat levels are also listed in a typical Appendix A document.

There are several sources of information that may be available to provide the background information that is required to develop an Appendix A document. These sources include, but may not be limited to, historical documents including monthly reports, Materials Control and Accountability (MC&A) reports, and test authorization and conclusion documents, fuel and reactor design documents, operator log reports, and safety analysis/evaluation reports for the irradiating facility. These and other sources together provide all of the information available on the subject fuel assemblies.

The MTRE fuel dimensions, physical conditions, and fissile isotopic loadings that are reported in the Appendix A documentation were reviewed, and the bounding conditions are listed in table 1.

4.0 RFA FOR BASIN STORAGE

Receipt of spent nuclear fuel at the L Basin for continued storage is contingent that the fuel is within the safety authorization basis that is provided in the L-MSF DSA [1]. An important aspect of the safety authorization basis is the dose to onsite and off-site personnel under design basis accidents. A Reference Fuel Assembly that bounds the radiological hazard for incoming fuel was previously constructed [2]. The RFA is a fictitious assembly with its radionuclide inventory and activity of each of its radionuclides obtained by selecting the maximum activity of radioisotopes from a broad range of fuels. The RFA provides, on a single assembly basis, the set of radioisotopes in fuel subject to design basis accident scenarios.

Table 2 below lists the RFA for fuel received and handled at the SRS L Basin. This isotopic inventory is used for accidents and can be used for shielding considerations in handling and storage.

Table 2 Material Contents (Ci) of the Reference Fuel Assembly (RFA) for L-Materials Storage Facility [1, 2]

	DSA RFA		DSA RFA
H-3	5.16E+01	**Po-212**	1.51E+01
Kr-85	1.05E+03	**Po-213**	8.55E-03
Sr-89	4.92E+01	**Po-215**	1.66E-02
Sr-90	8.08E+03	**Po-216**	2.36E+01
Y-90	8.08E+03	**At-217**	8.74E-03
Y-91	2.13E+02	**Rn-219**	1.66E-02
Zr-95	4.54E+02	**Rn-220**	2.36E+01
Nb-95	1.01E+03	**Fr-221**	8.74E-03
Nb-95m	3.37E+00	**Ra-223**	1.66E-02
Tc-99	1.03E+00	**Ra-224**	2.36E+01
Ru-103	2.17E+00	**Ra-225**	8.74E-03
Rh-103m[†]	1.96E+00	**Ac-225**	8.74E-03
Ru-106	2.11E+04	**Ac-227**	1.71E-02
Rh-106	2.11E+04	**Th-227**	1.64E-02
Ag-110	2.32E+00	**Th-228**	2.35E+01
Ag-110m	1.74E+02	**Th-229**	8.74E-03
Cd-113m	6.95E+00	**Th-231**	1.14E-02
Sn-119m	3.93E+00	**Th-232**	1.72E-02
Sn-123	1.45E+01	**Th-234**	2.16E-04
Sb-125	8.70E+02	**Pa-231**	2.28E-01
Te-125m	2.12E+02	**Pa-233**	8.59E-02
Te-127	3.47E+01	**Pa-234m**[†]	2.16E-04[†]
Te-127m	3.54E+01	**U-232**	4.09E+01
Te-129	1.20E-03	**U-233**	3.93E+01
Te-129m	1.85E-03	**U-234**	1.92E+00
Cs-134	1.03E+04	**U-235**	1.14E-02
Cs-137	9.28E+03	**U-236**	3.29E-02
Ba-137m	8.78E+03	**U-237**	2.59E-01
Ce-141	6.46E-02	**U-238**	8.42E-02
Ce-144	4.78E+04	**Np-237**	8.81E-03
Pr-144	4.78E+04	**Np-239**	9.62E+00
Pr-144m	5.74E+02	**Pu-236**	1.12E+02
Pm-147[†]	1.88E+04[†]	**Pu-238**	5.19E+01
Pm-148m	8.93E-03	**Pu-239**	5.80E+01
Sm-151	6.94E+01	**Pu-240**	9.78E+03
Eu-154	7.27E+02	**Pu-241**	1.06E+04
Eu-155	3.81E+02	**Am-241**	5.17E+01
Tl-208	8.46E+00	**Am-242m**	3.41E-01
Pb-209	8.74E-03	**Am-242**	3.40E-01
Pb-211	1.66E-02	**Am-243**	9.62E+00
Pb-212	2.36E+01	**Cm-242**	4.90E+02
Bi-211	1.66E-02	**Cm-243**	4.90E+00
Bi-212	2.36E+01	**Cm-244**	2.75E+03
Bi-213	8.74E-03	**Cm-246**	2.15E-01
		Total	2.31E+05

[†]from N-CLC-H-00084, Not in DSA

5.0 HEAT LOAD OF FUEL

Heat load evaluation, not based on radiological content per RFA, but on actuals was previously evaluated. The results are in Table 3 below. A value of 25 Watts is the bounding heat load for fuel that has been received at SRS. It is selected as design heat load, recognizing the decay with time.

Table 3 Decay Heat (watts) for an Aluminum MTRE Assembly [table reproduced from reference 4]

Decay Time (yr) from Reactor Discharge	Aluminum Fuel Heat Generation (watts)			
	Bounding Fuel		Nominal Fuel	
	HEU	LEU	HEU	LEU
1	105.23	108.89	45.18	46.61
2	45.64	47.67	20.05	20.88
3	26.14	27.24	11.55	12.00
6	11.30	11.52	5.200	5.260
10	8.52	8.580	4.010	4.030
20	6.367	6.530	3.023	3.073
30	4.997	5.243	2.371	2.453
60	2.480	2.830	1.163	1.284
100	1.010	1.382	0.461	0.594
200	0.144	0.487	0.055	0.179
300	0.0438	0.3442	0.0128	0.1224
600	0.0133	0.2218	0.0036	0.0826
1,000	0.0078	0.1468	0.0023	0.0574
2,000	0.0043	0.0794	0.0015	0.0345
3,000	0.0035	0.0630	0.0012	0.0285
6,000	0.0028	0.0505	0.0010	0.0232
10,000	0.0024	0.0410	0.0009	0.0187
20,000	0.0017	0.0265	0.0006	0.0120
50,000	0.0011	0.0103	0.0004	0.0046
100,000	0.0009	0.0034	0.0003	0.0014

6.0 CORROSION-DEGRADED CONDITION

Corrosion on aluminum fuel in water-cooled nuclear reactors typically produces two (2) primary aluminum oxyhydroxides. They are boehmite ($Al_2O_3 \bullet H_2O$) and gibbsite or bayerite ($Al_2O_3 \bullet 3H_2O$). An example of a corroded aluminum fuel assembly with both boehmite and gibbsite is shown in the photograph in Figure 1.

Investigations have shown that the formation of a protective oxide film on the aluminum surface at moderate temperatures occurs in three distinct stages [7], which are a function of time and temperature. The chemical precursor of the crystalline oxyhydroxide film structures is a gelatinous pseudo-boehmite [8]. The gelatinous film ages to form a tri-hydroxide with the structure of Gibbsite (hydrargillite) [γ -Al(OH)3] if the pH is lower than 5.8 or higher than 9, and Bayerite [α -Al(OH)3] if the pH is between 5.8 and 9 at temperatures below about 70–80°C. A pH of 5.2 is the value to limit Gibbsite formation on aluminum at 25°C [9, 10].

Between 100 and 400°C, crystalline boehmite will form. This is formed during heat flux conditions of reactor operation. The boehmite films grown on the fuel cladding may be 20–50 μm thick [11, 12].

The mature hydroxides are normally white color but other hues have been reported and may stem from absorption of Fe, Cr, Ni, or other metal ions leached from steels in the reactors.

Full removal of these corrosion products, especially boehmite, would be difficult, and therefore transitioning this fuel into dry storage containers requires knowledge of their characteristics and behavior under the conditions of storage.

In a dry storage system, the fuel surfaces, including the attached corrosion products, would be subject to an attendant gamma radiation field from the fuel itself and neighboring fuel in the storage system.

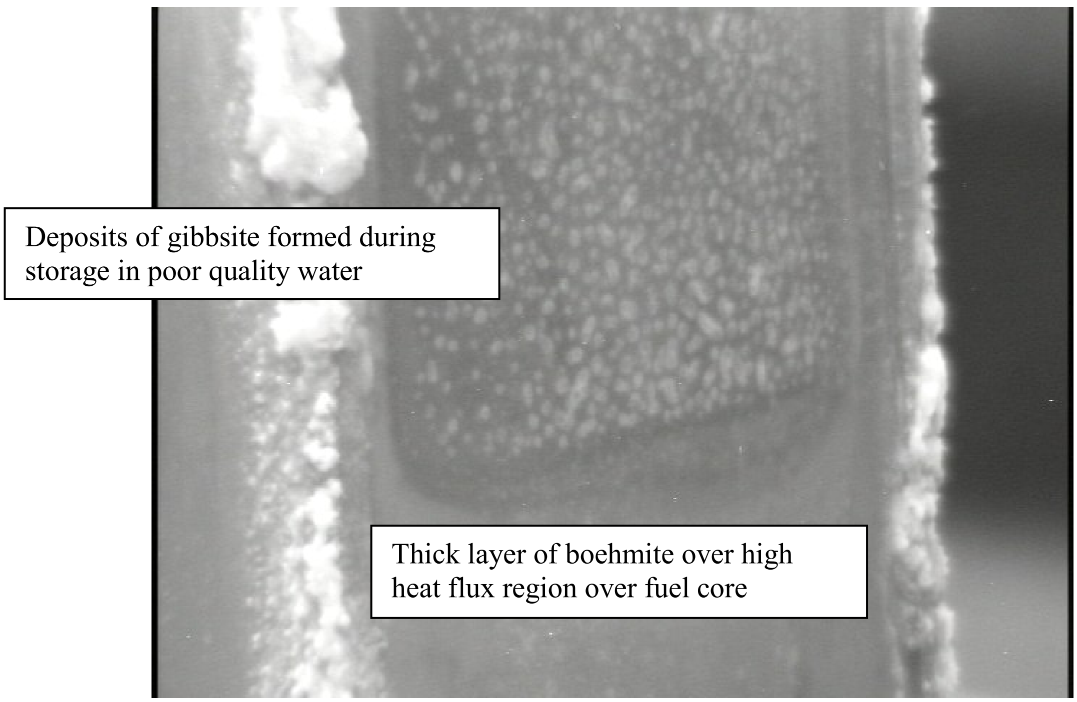

Figure 3 Aluminum-clad fuel assembly post-discharge and in basin storage

7.0 PITTING AND EXPOSED SURFACE AREA

Research reactor facilities primarily use wet storage for their spent fuel. Storage of fuel in poor quality water prior to receipt at SRS has caused pitting attack and, in some cases, areas of exposed fuel meat. This fuel was tested prior to shipment to SRS to verify acceptable C-137 release rates. The fuel is in L Basin bundle storage since the release rate is well within basin clean up capability.

As described above, the corrosion produced at low temperatures (< 77°C) leads to gibbsite production. An estimated of approximately 2 cm^3 is the bounding volume of Gibbsite on a single assembly. With a density of 2.4 g/cm^3, a total of approximately 5 grams Gibbsite bounds its content on a single assembly in the L Basin inventory. Further, this corrosion product can be removed, to an extent, by scraping the fuel.

L Basin inventory for fuel in direct basin storage is bounded by an assembly with 5% total exposed fuel meat [1].

8.0 RESULTS AND CONCLUSIONS

Table 1 summarizes the attributes for the RFA-Dry. No one assembly in L Basin has the composite attributes of the RFA-Dry, and it may not be feasible to design a sealed canister for a dry storage demonstration that would account for load of fuel at the bounding metrics of the RFA-Dry.

The RFA-Dry would allow for fuel selection and overall design considerations in the development of the dry storage demonstration for L Basin fuel.

9.0 REFERENCES

[1] WSRC-SA-2004-00002, Rev. 7, L-Area Materials Storage Facility Documented Safety Analysis, March 2011.

[2] N-CLC-H-00084, Revision A, Samer D. Kahook, "Radionuclide Activity of the Bounding Fuel Assembly in RBOF (U)," Westinghouse Savannah River Company, Aiken, SC 29802 (June 27, 1994).

[3] M.D. Dunsmuir, T.J. Spieker, "Return of research reactor spent fuel to the country of origin: requirements for technical and administrative preparations and national experiences," International Atomic Energy Agency. IAEA-TECDOC-1593, 2008.

[4] Si Y. Lee, R.L. Sindelar, and D.C. Losey, "Thermal Modeling and Performance Analysis of Interim Dry Storage and Geologic Disposal Facilities for Spent Nuclear Fuel," Nuclear Technology, Vol. 131, July 2000, pp. 124-151.

[5] WSRC-TR-97-0075, W.S. Large and R.L Sindelar, "Review of Drying Methods for Spent Nuclear Fuel," Westinghouse Savannah River Company, April 1997.

[6] WSRC-TR-2006-00111, D.W. Vinson, "Evaluation of Neutron and Gamma Source Spectra from Spent Nuclear fuel from HFR-Petten," Washington Savannah River Company (March 27, 2006).

[7] HART, R.K., Formation of films on aluminium, Trans. Faraday Society **53** (7) (1957) 1020–1027

[8] ROBERGE, P.R., Handbook of Corrosion Engineering, McGraw Hill, New York (2000).

[9] http://complex.gmu.edu/people/peixoto/subpages/ece590s08/Pourbaix.pdf

[10] ALWITT, R.S., The Aluminium-water system, Chapter 3 in Oxides and Oxide Films (DIGGLE, J.W., Ed.), Vol. 4, Marcel Dekker, New York (1976) 169–254.

[11] M. L. Griebenow, G. H. Hanson, A. P. Larrick, TRA Oxide Film Control and Surveillance; RE&C Report RE-A-77-059; Idaho National Laboratory: Idaho Falls, ID, Oct 1977.

[12] IAEA Nuclear Energy Series, NP-T-5.2, Good Practices for Water Quality Management in Research Reactors and Spent Fuel Storage Facilities, 2011