## Next Generation Melter(s) for Vitrification of Hanford Waste: Status and Direction - 11049

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## ABSTRACT

Vitrification technology has been selected to treat high-level waste (HLW) at the Hanford Site, the West Valley Demonstration Project and the Savannah River Site (SRS), and low activity waste (LAW) at Hanford. In addition, it may potentially be applied to other defense waste streams such as sodium bearing tank waste or calcine. Joule-heated melters (already in service at SRS) will initially be used at the Hanford Site's Waste Treatment and Immobilization Plant (WTP) to vitrify tank waste fractions. The glass waste content and melt/production rates at WTP are limited by the current melter technology. Significant reductions in glass volumes and mission life are only possible with advancements in melter technology coupled with new glass formulations.

The Next Generation Melter (NGM) program has been established by the U.S. Department of Energy's (DOE's), Environmental Management Office of Waste Processing (EM-31) to develop melters with greater production capacity (absolute glass throughput rate) and the ability to process melts with higher waste fractions. Advanced systems based on Joule-Heated Ceramic Melter (JHCM) and Cold Crucible Induction Melter (CCIM) technologies will be evaluated for HLW and LAW processing.

Washington River Protection Solutions (WRPS), DOE's tank waste contractor, is developing and evaluating these systems in cooperation with EM-31, national and university laboratories, and corporate partners. A primary NGM program goal is to develop the systems (and associated flowsheets) to Technology Readiness Level 6 by 2016. Design and testing are being performed to optimize waste glass process envelopes with melter and balance of plant requirements. A structured decision analysis program will be utilized to assess the performance of the competing melter technologies. Criteria selected for the decision analysis program will include physical process operations, melter performance, system compatibility and other parameters.

## **INTRODUCTION**

The National Research Council's Committee on Waste Forms Technology and Performance has identified technical advances that could allow the U.S. DOE to accelerate nuclear waste immobilization. In response, DOE's EM-31 has created a research program to develop and (where possible and cost effective) implement these transformational technologies in the DOE

complex. In particular, iron phosphate glass formulations and CCIM were identified as offering potential advantages over the current glass formulations and melter technology used to vitrify HLW and LAW. The NGM program is a portion of the effort to improve nuclear waste vitrification capability. The primary goal is to reduce the cost/schedule required for nuclear waste vitrification by increasing the rate of glass production and/or increasing the fraction of waste immobilized in a given package volume. [1,2]

The NGM program will evaluate the potential of replacing the first generation WTP melters with higher capacity melters at the first scheduled change-out in 2024. The Hanford Site goal is to establish melter systems leading to immobilization rates of eight metric tons per day HLW glass and 42 metric tons per day LAW glass, respectively. The CCIM is to be compared with an upgraded JHCM. Programmatic activities include glass formulation (borosilicate, silicate, and iron phosphate), bench-scale melter testing, NMG design, and prototypic test platform development, installation and operations. [2-6]

## WTP Melters and Potential NGM Designs

The WTP is currently designed to pre-treat waste into suitable feeds and vitrify the wastes in separate LAW and HLW facilities. Each facility uses two JHCMs. Melters are commonly rated by throughput per unit surface per unit time, a direct function of melt surface area. The WTP HLW melters have  $\approx 3.7m^2$  of melt surface and are rated at 800-1,000 kg/m<sup>2</sup>/day. The WTP LAW melters are significantly larger and have a higher throughput of just under 10m<sup>2</sup> of melt surface, rated at 1,500 kg/m<sup>2</sup>/day. [7,8]

Preliminary design concepts prepared by Energy *Solutions* for next generation JHCMs (LAW and HLW) target increased production by increasing melt surface and temperature capability, and optimizing air agitation of the melt. Specific to HLW vitrification, a system would have approximately 25 percent more available surface and an additional 50-100°C temperature capability, but still have a similar external footprint. The air agitation (bubblers) would be optimized to correspond with the increased surface area and operating conditions. The preliminary LAW design has up to 50 percent more surface area and similarly upgraded agitation and temperature capability. The JHCM designs are essentially based on a well established production capability, with the potential to approximately double throughput and take advantage of glass formulation improvements. Additional enhancements such as microwave boosting and feed pre-heating are being explored. Figure 1 illustrates the cross section of a JHCM melter. [9,10]

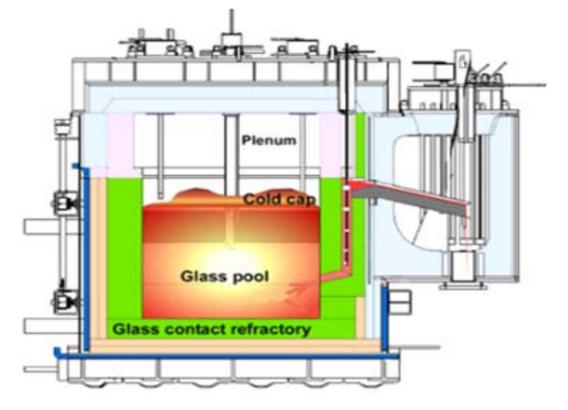


Fig. 1. Cross-Sectional Representation of a Joule-Heated Ceramic-Lined Melter. Courtesy Energy *Solutions*.

The CCIM (as developed by AREVA) has been initially optimized for vitrifying HLW generated by power reactors. The technology is based on electrically heating glass (with an induced current) inside a water-cooled metal shell. The water cooling forms a "skull" of highly viscous glass that protects the metal shell from thermal corrosion/erosion. The absence of refractory and electrodes implies the CCIM's operating temperature is not directly limited by contact materials issues, as is the case with the JHCM. A CCIM with a melt surface area of approximately 0.3 m<sup>2</sup> has been installed at La Hague, France, and is vitrifying radioactive waste. Throughput testing of SRS HLW simulant feed in a pilot melter (located in Marcoule, France) indicates a melt rate as high as  $\approx$ 2,800 kg/m<sup>2</sup>·day at 1,250 °C<sup>1</sup>. A view of the CCIM installed at Marcoule is shown in Figure 2. [11-13]

<sup>&</sup>lt;sup>1</sup> Simulant based on the SRS Defense Waste Processing Facility sludge batch #4.



## Fig. 2. Exterior View of Cold Crucible Induction Melter. Courtesy AREVA

## Next Generation Melter Programmatic Goals

To improve on the baseline mission estimates, the NGM systems will have to process more waste per unit time, to the extent practical within the WTP operating envelope. Table I denotes the expected production capacity demonstrated for HLW and LAW immobilization from cold/hot commissioning through normal operations, and provides the programmatic targets for the LAW and HLW NGM systems.

Table I, which is consistent with WTP project guidance, does not explicitly imply or relate to a waste loading. This table, while useful in establishing relative throughputs of a given melter type, may not be adequate to differentiate between dissimilar melters processing distinct, unique glasses – even those derived from the same waste formulation. Production improvement can be accomplished by increasing either the rate of glass production and/or waste loading.

The original WTP production goals were based on joule-heated melters of given temperature capability. Upgraded JHCMs or CCIMs are expected to have enhanced temperature capabilities to be fully exploited by the melter designer/glass chemist. Glass formulation activities for the CCIM or upgraded JHCM are targeted to maximize waste loading. Currently, the nominal waste loading projected for WTP HLW ranges from  $\approx$ 28-38 weight percent (oxide basis). Glass formulations with increased loading (40<sup>+</sup> weight percent for HLW – maximizing sulfur loading for LAW) lead to reduced glass production (fewer canisters) requirements, effectively increasing the productivity of a given melter without increasing "stress" on other facility systems.

Likewise, the higher waste loading (or different glass family) formulations may yield glasses with higher density – which may indirectly increase effective productivity. The density of a melt directly impacts the amount of waste in a given package (i.e. canister). Iron phosphate glasses are on the order of 10-15 percent denser than corresponding borosilicate glasses. The NGM program may eventually compare dissimilar melter systems optimized for processing glass compositions with significantly different physical and chemical properties. Effectively then, to compare the relative performance of the melter systems, their "throughput" will need to be based on waste oxide mass processed per unit time (into an acceptable product). [7,8,14-17]

Table I. Relative Production Targets Established for WTP Operations and Potential Upgrade Targets for the Next Generation Melter Program.

HLW	Unit Melter Design Capacity <sup>2</sup>	# Units	Design Basis HLW Capacity <sup>3</sup>	HLW Throughput Rate(70% TOE) <sup>4</sup>
Cold/Hot Comm. <sup>5</sup>	3	2	6.0	4.2
Plant Capacity per contract	3.75	2	7.5	5.25
NGM Program	5 - 5.75	2	10 - 11.5	$7 - 8^{(6)}$

LAW	Unit Melter Design Capacity	# Units	Design Basis LAW Capacity	LAW Throughput Rate(70% TOE)
Cold/Hot Comm.	15	2	30.0	24
Plant Capacity per contract	15	2	30	21
NGM Program	25 - 30	2	50 - 60	$35 - 42^{(7)}$

<sup>&</sup>lt;sup>2</sup> Metric tons glass per day (per melter glass throughput capacity)

<sup>&</sup>lt;sup>3</sup> Metric tons glass per day (design basis facility throughput – feed unlimited)

<sup>&</sup>lt;sup>4</sup> Metric tons glass per day (target WTP throughput)

<sup>&</sup>lt;sup>5</sup> Cold/Hot Commissioning. Activities performed during initial start-up through to nominal radioactive operations.

<sup>&</sup>lt;sup>6</sup> Target as per DOE – EM Tank Waste Research and Development Plan

<sup>&</sup>lt;sup>7</sup> Target as per DOE – EM Tank Waste Research and Development Plan

# **Project Status**

The NGM program and associated glass formulation and bench scale testing efforts have been initiated. Preliminary design concepts for the JHCM and CCIM indicate it will be possible to significantly increase HLW throughput beyond current WTP design criteria. The JHCM design effort likewise indicates that LAW immobilization can be at least doubled (feed unlimited melter basis). The NGM development project aims to develop a JHCM and CCIM to a point where a decision to deploy can be rendered in the 2013-2014 timeframe. Figure 3 represents the concerted effort required to develop those systems (and associated chemical flowsheets) up through and including deployment at WTP. In conjunction, the DOE's critical decision path and project phases are represented. [9,10,18,19]

A key date in the development and deployment timeline is April 2015. A proposed milestone (M-062-45) in the Hanford Federal Facility Agreement and Consent Order, the pact governing cleanup, requires a formal decision on the specific actions that will constitute supplemental treatment by this date. Supplemental treatment actions and options include enhancing WTP melter production rates. The NGM development project, therefore, is structured to support the April 2015 milestone. As a result, a decision as to which melter technologies will be further developed for deployment will likely be required in the 2013/2014 timeframe. Development of JHCM and CCIM technologies to a Technology Readiness Level (TRL) 6<sup>(8)</sup> for both LAW and HLW applications may not be possible within this timeframe. The NGM development will advance as far as possible given funding and schedule constraints, but additional NGM development will likely be required during the deployment phase. [6,7,18,20]

At this initial stage of project execution, it is not possible to predict the exact form that NGM development and deployment will entail. Planning to date includes pilot-scale melter and integrated melter system testing to provide the information required to confidently proceed with design and fabrication of NGMs during the deployment phase. Operating these test systems is expected to be completed by 2017 to 2018. If development yields an essentially "like kind" NGM (i.e., same space envelope, interface connections, and facility services as the first generation melters), deployment would encompass a relatively straightforward procurement and equipment replacement action. Conversely, if the NGM requires substantial modifications to the WTP, i.e., new or upgraded facility services or reconfigured interface connections, an engineering, procurement, and construction project as shown in Figure 4 will be required. Regardless, the structure of this project will be guided by DOE-STD-1189, *Integration of Safety into the Design Process*. [6,21]

<sup>&</sup>lt;sup>8</sup> Technical Readiness Level 6 definition: Engineering-scale models or prototypes are tested in a relevant environment. This represents a major step up in a technology's demonstrated readiness. Examples include testing an engineering-scale prototypical system with a range of simulants. Supporting information includes results from the engineering-scale testing and analysis of the differences between the engineering-scale, prototypical system/environment, and analysis of what the experimental results mean for the eventual operating system/environment. TRL 6 begins true engineering development of the technology as an operational system. The major difference between TRL 5 and 6 is the step up from laboratory scale to engineering scale and the determination of scaling factors that will enable design of the operating system. The prototype should be capable of performing all the functions that will be required of the operational system. The operating environment for the testing should closely represent the actual operating environment. *Technology Readiness Assessment (TRA) Deskbook, U.S. Department of Defense, Washington, D.C. (2009).* 

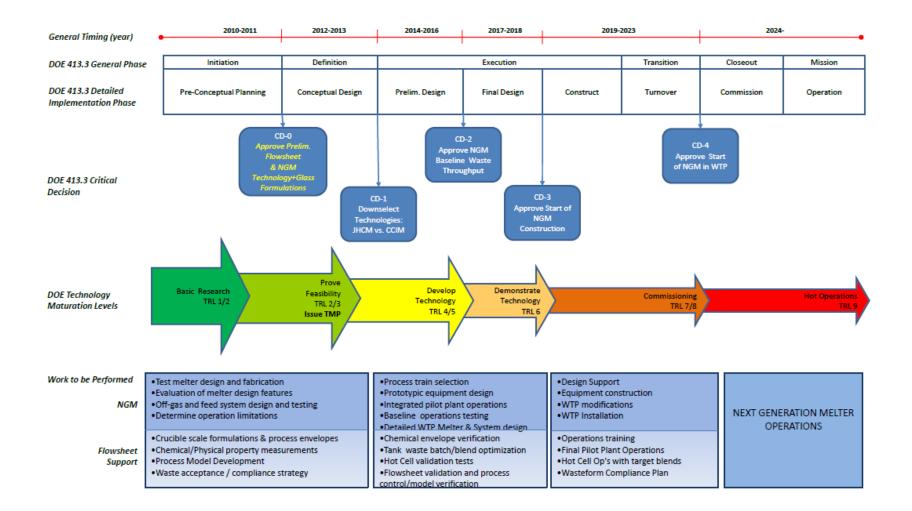
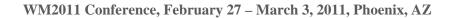


Figure 3. Overlay Representation of NGM Technology Development, Maturation Levels, and Critical Decision Path.



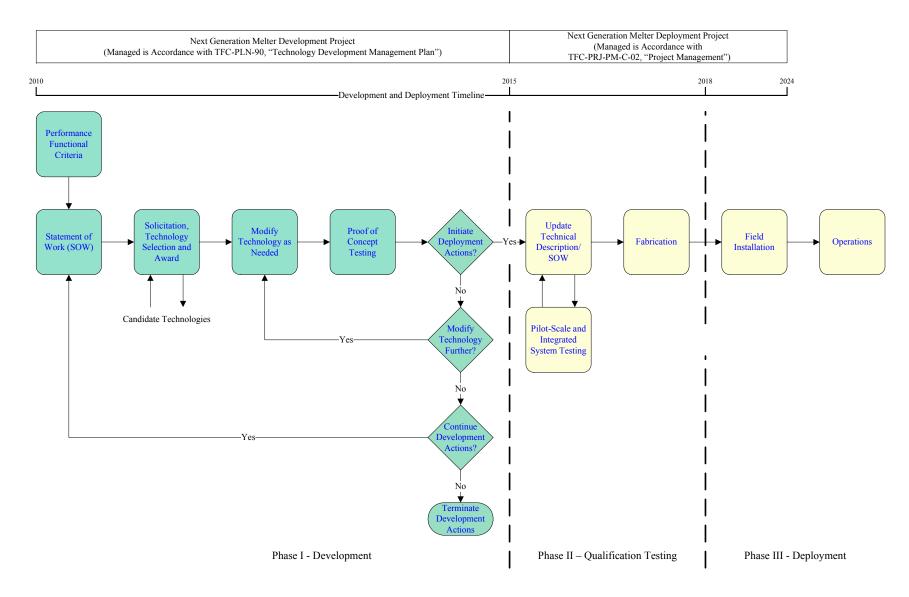


Figure 4. Phased Approach for Development, Qualification and Deployment of Next Generation Melter.

## **Project Direction – Near Term**

To support the DOE-EM Tank Waste Research and Development Plan objectives, a downselection process has been initiated. The process is expected to provide for a rigorous, defensible, and documented decision framework. Initially, selection criteria and decision information needs are established. These factors provide expectations for the technology providers, organizations responsible for coordination of tasks, and the decision-making entities. Periodic assessment of the progress of melter development will be performed and interim decision analysis may be warranted. The decision analysis process is assumed to be used at the end of each year until the final decision is made. In conjunction with the technology provider, a preliminary list of primary selection criteria was developed. A sub-set of this list is provided in Table II. Table II focuses on the technical development criteria but also demonstrates the facets of development that must be included to ensure the technology can be deployed at scale in a radioactive environment. While the process is directed toward technology development, it is essential to evaluate safety, environmental and facility considerations, and other factors to transition to a successful deployment project. [22]

Table II. Preliminary Criteria for the HLW NGM Down-Selection Process.

- 1) Safety
  - a. Operational Safety
- 2) Environmental
  - a. Liquid Waste Generation
  - b. Off-gas Generation
  - c. HLW Glass Mass Immobilization<sup>9</sup>
- 3) Impacts to Existing Facilities
  - a. Modifications to WTP HLW Facility
  - b. Modifications to WTP Outside of HLW
  - c. Impact on Facilities External to WTP
- 4) Melter Technology
  - a. Nameplate Waste Throughout<sup>10</sup>
  - b. Targeted Waste Throughput<sup>11</sup>
  - c. Melter Availability
  - d. Operability Issues
  - e. Maintainability Issues
  - f. Melter Failure (non-replacement) Recovery,
  - g. Melter Lifetime
  - h. Ease of Melter Replacement

<sup>&</sup>lt;sup>9</sup> It is possible non-borosilicate glasses will be utilized by competing NGM technologies and a simple waste loading comparison may not fully reflect the positive or negative factors involved in selection.

<sup>&</sup>lt;sup>10</sup> Waste throughput of a melter (as used for the NGM project) is a function of specific melter throughput (kg glass per meter square melt surface per day), melt surface area (square meters), and waste oxide mass fraction in glass. "Nameplate" waste throughput reflects the NGM design capacity for processing high-iron (AZ-101) waste.

<sup>&</sup>lt;sup>11</sup> Targeted waste throughput reflects the NGM capacity for processing high-chromium, high-aluminum, or other specific Hanford waste types – as identified in the NGM program guidance for glass formulation.

Table II. Preliminary Criteria for the HLW NGM Down Selection Process, continued.

- 5) Technical Maturity
  - a. Confidence to reach TRL-6 by 2016
- 6) Schedule
  - a. Confidence to meet WTP insertion date (2025 operations)
- 7) Economic
  - a. WTP development costs up to design and commissioning
  - b. WTP design and commissioning costs up to operations
  - c. WTP operations costs

## **Project Direction – Long Term**

The NGM program is a development project in concert with glass formulation and other associated programs (including technetium management, cold cap phenomena, and others) intended to increase the capability to safely manage and immobilize legacy nuclear waste. Coordination between these programs is essential from start to finish. For instance, as melter technology diverges, the optimal glass formulation for each melter may likewise diverge. Operating temperature, materials compatibility, routine maintenance schedules, and retention of semi-volatile radionuclides are examples of likely process differentials, and operational and/or system performance issues that will be impacted by programmatic decisions. As such, programmatic linkage is essential to ensure that optimal glass formulations/flowsheets are provided for specific systems. Ultimately, the insertion of a new technology into either WTP LAW or HLW processing will only occur in concert with process control and waste acceptance methodologies developed, demonstrated, and qualified for said technology. [2,6]

## CONCLUSIONS

Development and design activities (leading to an NGM for processing Hanford LAW and HLW) have commenced. Concepts for CCIM and JHCM systems have been refined and translated into pre-conceptual designs. These designs reflect the temperature and physical limitations of each melter as applied to the specific requirements for processing Hanford LAW and HLW. In concert, however, the NGM systems are designed to have greater melt throughput than the existing WTP LAW and HLW melters.

## JHCM Upgrades (HLW)

- Increased temperature capability
- Increased melt surface
- Improved agitation

# **CCIM Features (HLW)**

- Increased temperature capability
- Increased specific throughput
- Improved agitation

## **Change versus Current WTP**

50-100°C higher operating temperatures 25% greater surface area Optimized bubbler package

# Change versus Current WTP

100-200°C higher operating temperatures  $\approx 2^+$  times greater Mechanical stirring The melter systems are intended to accommodate higher waste loading glass formulations, primarily as a result of the greater thermal energy capability. Likewise, processing silicate and/or phosphate glasses (intended to have significantly higher waste loading) is a design consideration. Melter performance testing and WTP compatibility evaluations will include not only the melter but the type of glass to be processed, canister generation, impacts to facilities, pre-treatment and analytical requirements, and waste acceptance considerations. The melter test and ranking criteria and the down-selection process will entail considerable effort beyond glass production. As a result, melter design and process flowsheet development will be revisited periodically to ensure that down-selection criteria adequately frame the potential for successful NGM operations in the WTP.

## REFERENCES

- 1. <u>Waste Forms Technology and Performance Interim Report</u>, National Research Council, National Academies Press, Washington D.C. (2010).
- 2. <u>Tank Waste Research and Development Plan</u>, United States Department of Energy, Office of Environmental Management, Washington, D.C. (2010).
- 3. "*Glass Formulation for Next Generation Waste Glass Melters*," WP 4.1.2-2010-001, in DOE-EM 31 Technology Development & Deployment (TDD) Program Task Plan, U.S. Department of Energy, Washington, D.C. (2010).
- 4. "*Next Generation Advanced Joule-Heated Melter Bench Scale Testing*," WP 4.1.3-2010-001 Revision A , in DOE-EM 31 Technology Development & Deployment (TDD) Program Task Plan, U.S. Department of Energy, Washington, D.C. (2010).
- "Phosphate Glass Development and Demonstration," WP 5.1.1-2010-001, in DOE-EM 31 Technology Development & Deployment (TDD) Program Task Plan, U.S. Department of Energy, Washington, D.C. (2010).
- 6. RPP-PLAN47114, "Next Generation Melter and Glass Formulation Development Project Execution Plan," Washington River Protection Solutions, Richland, WA (2010).
- 7. ORP-11242, "*River Protection Project System Plan*," Revision 5, U.S. Department of Energy, Office of River Protection, Richland, WA (2010).
- 8. T. W. Crawford, P. J. Certa, and M. N. Wells, "*System Planning with the Hanford Tank Waste Operations Simulator*," Proceedings of the Waste Management Conference, Phoenix, AZ (2010)
- 9. *Next Generation Melter HLW Pre-Conceptual Design*, to be published in Proceedings of the Waste Management Conference, Phoenix, AZ (2011)
- 10. Next Generation Melter LAW Pre-Conceptual Design, to be published in Proceedings of the Waste Management Conference, Phoenix, AZ (2011)
- 11. A. Bonnetier, "Nuclear Waste Immobilization by Vitrification in a Cold Crucible Melter: 3D Magnetic Model of a Melter," Materials Research Society Symposium Proceedings, Volume 1107, Materials Research Society, Warrendale, PA (2008).

- 12. A. B. Barnes, D. C. Iverson, B. J. Adkins, E. Tchemitcheff, "Feasibility Evaluation and Retrofit Plan for Cold Crucible Induction Melter Deployment in the Defense Waste Processing Facility at Savannah River Site," Proceedings of the Waste Management Conference, Phoenix, AZ (2008)
- 13. CEA/DTCD/2009/02, "Vitrification of a Representative Simulated SB4 Sludge in a CCIM Pilot Platform at Marcoule – Demonstration Report."
- W. Huang, D. E. Day, C. S. Ray, C. W. Kim, "High Temperature Properties of an Iron Phosphate Melt Containing High Chrome Nuclear Waste," Journal of Nuclear Materials, 346, pages 298-305 (2005).
- 15. A. Karamanov and M. Pelino, "Crystallization Phenonema In Iron Rich Glasses," Journal of Non-Crystalline Solids, 281, pp. 139-151 (2001).
- W. K. Kot, H. Gan, and I.L. Pegg, "Sulfur Incorporation in Waste Glass Melts of Various Compositions," Ceramic Transactions, Volume 107, pp 44, Eds. G. T. Chandler and X. Feng, American Ceramic Society, Westerville, OH (2000).
- 17. PNNL-18501, J. D. Vienna et al, "*Glass Property Data and Models for Estimating High-Level Waste Glass Volume*," Revision 0, Pacific Northwest National Laboratory, Richland WA (2009).
- 18. *Next Generation Melter CCIM Configuration for HLW*, to be published in Proceedings of the Waste Management Conference, Phoenix, AZ (2011)
- 19. USDOE Order 413.3A, *Program and Project Management for the Acquisition of Capital Assets*, U. S. Department of Energy, Washington D.C. (2008).
- 20. Technology Readiness Assessment (TRA) Deskbook, U.S. Department of Defense, Washington, D.C. (2009).
- 21. USDOE Standard-1189, *Integration of Safety into the Design Process*, U. S. Department of Energy, Washington D.C. (2008).
- 22. RPP- RPT 48005, M. F. Gray, et al, "Next Generation Melter Optioneering Study Interim Report," Washington River Protection Solutions, Richland, WA (2010).