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Activities to Establish Technical Confidence for HTGR Fuel Processing

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June 2019

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EXECUTIVE SUMMARY

The Savannah River National Laboratory (SRNL) and Jülicher Entsorgungsgesellschaft für Nuklearanlagen mbH (JEN) are partnering in the demonstration of a digestion technology for the processing of graphite-based nuclear fuels. The purpose of this partnership is to identify, demonstrate, and mature a technology for processing high temperature gas-cooled reactor (HTGR) fuel currently stored at the Jülich and Ahaus facilities in Germany. To enable a decision on receipt of the fuel by the U.S. Department of Energy (DOE), the technology for dispositioning the fuel must achieve an acceptable level of technical maturity, what is referred to as "technical confidence."

Technical confidence represents a state of the technology development such that all high-risk technical issues have been either resolved or are understood sufficiently to chart a reasonable and affordable course for their resolution. There are five overlapping activities identified to achieve this confidence. The technologies and unit operations which precede these activities (cask handling, cask opening, pebble feeding) or succeed these activities (dissolution, immobilization) are of sufficient maturity to support the attainment of technical confidence.

- Activity 1: Establish engineering-scale baseline process conditions and system design
- Activity 2: Demonstrate engineering-scale process rates and sustainment conditions
- Activity 3: Demonstrate critical engineering functions with unirradiated fuel
- Activity 4: Validate fuel residue and fission product assumptions with irradiated fuel kernels
- Activity 5: Perform pilot-scale and integrated engineering-scale demonstrations

In general terms, technical confidence will be achieved when:

- The fuel digestion system, fuel preparation equipment, and off-gas system interface (including analytical monitoring) can be operated and maintained at the engineering scale without "fatal flaws" as part of an integrated system with minimal hands-on interaction while digesting representative fuel simulants.
- The fuel digestion system and its critical engineering features can be successfully operated at the pilot scale without fatal flaws using representative fuel simulants
- Critical engineering features of the fuel digestion system are adequately demonstrated without fatal flaws using unirradiated fuel.
- The behaviors of radioactive and simulated fission products are sufficiently understood to satisfy critical design and modeling inputs.
- For the L-Area Option specified in the HTGR Environmental Assessment, the feasibility of the reductive melt-dilute process is demonstrated as a disposition pathway for Th-U oxide fuel kernels.

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LIST OF ABBREVIATIONS

CTE	Critical Technology Elements
DOE	Department of Energy
EDL	Engineering Development Laboratory
EDS	energy dispersive X-ray spectroscopy
HTGR	high temperature gas-cooled reactor
JEN	Jülicher Entsorgungsgesellschaft für Nuklearanlagen mbH
SEM	scanning electron microscopy
SRNL	Savannah River National Laboratory
SRP	Standard Review Plan
SRS	Savannah River Site
TE	Technology Elements
TRA	Technical Readiness Assessment
TRL	Technical Readiness Level

1.0 Program Overview

The Savannah River National Laboratory (SRNL) and Jülicher Entsorgungsgesellschaft für Nuklearanlagen mbH (JEN) are partnering in the demonstration of a digestion technology for the processing of graphite-based nuclear fuels.[1] The purpose of this partnership is to identify, demonstrate, and mature a technology for processing high temperature gas reactor (HTGR) fuel currently stored at the Jülich and Ahaus facilities in Germany.

The fuel is in the form of graphite spheres, called pebbles, each of which contains thousands of small kernels containing oxides or carbides of uranium and thorium. SRNL developed a patented vapor-phase oxidation process for digestion of the graphite and recovery of the U and Th for disposition.[2] Flow diagrams are provided in Attachment 1. Feasibility of the process and an improved understanding of the chemistry have been documented.[3][4] However, additional development and demonstration activities are required to mature the technology to a level sufficient for the Department of Energy (DOE) to take ownership of the fuel and have it transported to the Savannah River Site (SRS).[5]

This document discusses the development and engineering tasks that will be performed to establish "technical confidence". Technical confidence differs from technical readiness in that technical confidence is not directly tied to stages toward full-facility design. The different technology maturation scales are depicted in Attachment 2, and critical process requirements are listed in Attachment 3. Technical confidence represents a state of the technology development such that all high-risk technical issues have been either resolved or are understood sufficiently to chart a reasonable and affordable course for their resolution. It recognizes that additional testing may be warranted to reach maturity for a high confidence engineering and construction project. A list of the technical confidence areas and their associated risk levels is provided in Attachment 4. There are five overlapping activities.

- Activity 1: Establish engineering-scale baseline process conditions and system design
- Activity 2: Demonstrate engineering-scale process rates and sustainment conditions
- Activity 3: Demonstrate critical engineering functions with unirradiated fuel
- Activity 4: Validate fuel residue and fission product assumptions with irradiated fuel kernels
- Activity 5: Perform pilot-scale and integrated engineering-scale demonstrations

2.0 Background

In January 2016, a Technology Readiness Assessment (TRA) team determined that five Technology Elements (TEs) constitute the major components of the baseline flowsheet: 1) the Primary Vapor Digester/Kernel Polisher, 2) the Roller Mill/Crusher, 3) the Secondary Crushed-Kernel Vapor Digester, 4) the Off-Gas System, and 5) the Pneumatic Transfer System. Of these five TEs, the Primary Vapor Digester/Kernel Polisher, the Secondary Crushed-Kernel Vapor Digester, and the Off-Gas Exhaust System were determined to be Critical Technology Elements (CTEs).[5][6] In all cases, the three CTEs were determined to be at Technical Readiness Level (TRL) 4; however, the integrated flowsheet was determined to be at TRL 3. See the following table for explanation of the readiness levels. To advance the technical readiness outlined by that assessment, important technology maturation activities and engineering-scale demonstrations occurred as authorized by Revision 4 of the Work for Others.[1]

The previous TRA did not evaluate two other TEs: 6) Cask opening, unloading, and pebble transfer, and 7) fuel dissolution and disposition. Cask opening, unloading, and pebble transfer is also a CTE. Further development of these to TEs is not needed at this time because neither are considered high-risk items (Attachment 4). It was also concluded that the pneumatic transfer systems developed by General Atomics and the DWPF-type off-gas system are mature technologies, and do not require further development at this time.

TRL 3	Equipment and process analysis and proof of concept have been demonstrated in a simulated environment.
TRL 4	Laboratory/bench scale testing, at a minimum, of similar equipment systems has been completed in a simulated environment.
TRL 5	Laboratory/bench scale equipment/process testing has been demonstrated in a relevant environment.
TRL 6	Engineering-scale equipment/process testing has been demonstrated in a relevant environment.
The TRL levels identified above follow the same pattern and general principles but differ from the formal TRL definitions in the Standard Review Plan (SRP) (DOE-EM-SRP-2010, Rev. 2), which assumes development in parallel with development of the project scope such that the TRL 6 level corresponds to CD-3 (ready to construct). There are three key differences. First, since the design is not being developed in parallel; hence, the tasks in DOE STD 1189-2008 do not formally apply. Second, the requirements for similarity matching the "final application in almost all respects" as required by the SRP would by better stated as matching the postulated rather than the "final" application. Third, SRP defines engineering scale as 1/10 scale or above. We use 1/30.	

Efforts to achieve Technical Confidence involve five core activities, each with supporting tasks.

- Activities 1 and 2 complete the baseline design and operating conditions. They also incorporate design features for maintenance and sustainment of operations. Hence, they provide critical input for Activities 3 and 5.
- Activity 3 entails the design and operation of a primary digester using unirradiated fuel that has similar features as the Engineering Development Laboratory (EDL) unit, especially the critical interfaces, but uses a short-length (~36 in.) digestion vessel.
- Activity 4, testing with irradiated fuel, provides data on the behavior of fission products from actual HTGR fuel to correlate with previously-measured simulant fission product data.
- Activity 5 demonstrates the design and operation of a non-radioactive, integrated, engineering-scale system in EDL which has a production-length digestion vessel, and successful operation of a primary digester with critical engineering features at pilot scale.

Flowsheet block diagrams for the two principle processing options, with deployment at either H-Canyon or in L-Area, are contained in Attachment 1. The orange blocks represent fuel digestion and are the principal areas of technology maturation. The blue blocks (fuel preparation for disposition), yellow blocks (fuel disposition), and green blocks (off-gas handling) are demonstrated technologies for this or similar processes. The orange and blue blocks are identical technologies for either processing option. Although the technologies associated with the blue boxes are established technologies, these activities will demonstrate an integrated system that combines the unit operations represented by the orange blocks, the blue blocks, and the first green block.

3.0 Defining Technical Confidence

This document identifies a series of tasks (described in subsequent paragraphs) which, when completed, establish the HTGR fuel processing technology as sufficiently mature to initiate a design project for full-scale processing. The tasks must be either successfully completed or an analysis completed to provide a clear, feasible, affordable path forward for successful completion.

Critical "technical confidence" assumptions are:

- Casks can be opened, and the pebbles transferred to a feed hopper for the primary digester. This is a medium-risk technology element that will be developed as part of subsequent technology maturation activities.
- Fuel disposition and solution disposition via H-Canyon for these types of fuels are existing, mature technologies.[8][9]

- For the L-Area Option specified in the HTGR Environmental Assessment, the baseline melt-dilute process for irradiated uranium fuels is a feasible disposition pathway for HTGR fuel kernels, and has been demonstrated at pilot-scale.[10][11][12] The use of a DWPF-type off-gas system in the HTGR approach reduces off-gas risks for the melt-dilute disposition path.

In general terms, technical confidence will be achieved when:

- The fuel digestion system (orange blocks in Attachment 1), fuel preparation equipment (blue blocks), and off-gas system interface (first green block), including analytical monitoring, can be operated and maintained at the engineering scale without fatal flaws as part of an integrated system with minimal hands-on interaction while digesting representative fuel simulants.
- The fuel digestion system (orange blocks) and its critical engineering features can be successfully operated at the pilot scale without fatal flaws using representative fuel simulants.
- Critical engineering features of the fuel digestion system (orange blocks) are adequately demonstrated without fatal flaws using unirradiated fuel.
- The behaviors of radioactive and simulated fission products are sufficiently understood to satisfy critical design and modeling inputs.
- For the L-Area Option specified in the HTGR Environmental Assessment, the feasibility of the reductive melt-dilute process is demonstrated as a disposition pathway for Th-U oxide fuel kernels.

In specific terms, technical confidence will be achieved when the following tasks are completed successfully or completed with sufficient accumulated knowledge to define a clear, feasible, affordable path forward for successful completion.

- Measure scrubber performance parameters and establish acceptable contact times for absorption of NOx gases in the off-gas system using hydrogen peroxide as the absorbent.
- Evaluate the feasibility of melt-dilute technology for the disposition of HTGR fuel kernels and fission product simulants to confirm the viability of the L-Area option described in the HTGR Environmental Assessment.
- Provide an evaluation by subject matter experts of the design with respect to remote operation in either H-Canyon or L-Area.
- Demonstrate particulate removal from top vent screen and bottom screen to mill feed.
- At the engineering-scale, demonstrate and validate fundamental interface design features (e.g., top filter vent, bottom screen, multi-zone control) against critical process requirements (Attachment 3) using representative fuel simulants.
- At the engineering scale, establish the preliminary throughput rate and validate against critical process requirements (Attachment 3). This effort includes extended duration testing where all subsystems are at steady state.
- Characterize the digester off-gas (particulate graphite, fission products, inorganic gases) for baseline process conditions, and validate these against critical process requirements (Attachment 3).
- Test the entrainment behavior of U-Th carbide and UO_2 kernel types and validate that their behavior does not represent a fatal flaw that prevents successful operation of the process. Data available from recent German testing will be combined with simulant particle testing and computer modeling to establish design properties for managing the kernels and particulate graphite in the digester.
- Evaluate off-normal process conditions and validate that such conditions do not represent a fatal flaw with regards to safety, security, operability, or maintenance.
- Validate maintenance and clean-out concepts of the digester system against critical process requirements (Attachment 3).
- Validate maintenance and clean-out concepts of the digester off-gas system against critical process requirements (Attachment 3).

- Demonstrate satisfactory digestion and engineering performance with unirradiated BISO U-Th oxide and unirradiated BISO U oxide fuel pebbles. This scope includes characterization of the partitioning (i.e., migration, fuel residues) of fuel to different regions of the process.
- Operate an integrated engineering-scale system and validate against critical process requirements (Attachment 3) using representative fuel simulants. Integration includes fuel digestion, first stage of the off-gas system, milling, and secondary digestion. This effort includes extended duration testing where all subsystems are at steady state.
- Demonstrate satisfactory digestion and engineering performance at the pilot-scale with representative fuel simulants.

4.0 Activities and Tasks

A conceptual schedule of the primary activities is provided in Attachment 5. A full schedule will be developed upon authorization of the work scope.

4.1 Activity 1: Establish Engineering-Scale Baseline Process Conditions and System Design

This activity matures the technology at the engineering-scale (1/30th scale, based on 1400 pebbles/day) to finalize the engineering technical baseline. To better understand engineering-scale, it is necessary to understand full-scale. The proposed full-scale primary digestion vessel is an annular vessel (for criticality control reasons) approximately 36 inches diameter, 8 feet tall, with an annular width of 3 inches (to accommodate pebbles that are 2.36 inches diameter). Engineering-scale reflects full-scale in two dimensions – it is about 8 feet tall and has a reaction cross-section of 3 inches. There are two approaches under consideration for the full-scale design. One approach is to use a 36-inch diameter annular vessel with internal compartmentalization to channel reaction over the required cross section, while maintaining a common top and bottom sections similar to the original design. Another approach is modified full-scale design that uses an array of cylindrical vessels (5.3 to 6.4 inches diameter). The modified full-scale design complicates material handling issues (pebble feeding and product removal) while simplifying the geometry of fundamental engineering components (top vent screen and bottom screen). Pilot-scale (1/10th scale) could be a single cylindrical vessel, 5.5 to 6.5 inches diameter or with a slab section of equivalent capacity. The report assumes the modified design, but leaves open the choice until Task 5. Attachment 2 provides a comparison of the different scales plus a picture of the engineering-scale primary digestion vessel. Attachment 2 compares modified full-scale configurations to the baseline configuration.

Engineering-scale is deemed adequate for advancing technical confidence for the full-scale design because it digests full pebbles in a system that reflects full-scale in two spatial dimensions. As can be seen in Attachment 2, pilot-scale approximates an array of three or four pebbles side-by-side; full-scale approximates ~30 engineering-scale vessels side-by-side. If the modified full-scale design with an array of smaller digestion vessels is adopted, the engineering-scale digestion vessel design is 1/3rd to 1/4th scale of an individual digestion vessel, and the pilot-scale digestion vessel design is at or near full-scale of an individual digestion vessel.

4.1.1 Technology Development Status

An engineering-scale system has been built and tested. The use of multi-zone heating to improve oxidation efficiency and reduce digestion temperature has been validated at the engineering-scale, although the report for those results is still in draft. Testing was completed at a range of process temperatures and a digestion temperature range has been identified for operations. Those results were consistent with more-extensive laboratory-scale system tests.[13] One refinement to the oxidant delivery system will be implemented to distribute oxygen more-uniformly in each feed zone – a perforated oxygen feed line will be installed inside the digester to allow multiple-point oxygen injection within each zone.

To date, the project has designed, fabricated, and validated the fundamental design feature of the top vent screen which transitions from the primary digester to the off-gas system.[4][14] Maturation activities also involved design, fabrication, and testing of the fundamental design feature of the bottom screen which transitions from the primary digester to milling.[14] Additional refinements to the types and sizes of screens are required to enable the bottom screen to function effectively. A variety of experimental data show no negative impacts of packing or channeling of reactant gases in the system, although fines accumulation remains a concern.[4][15][16] However, the longest digestion demonstration to date was 18 hours.

SRNL completed experiments to quantify two types of off-gas components. First, there are the reaction byproducts from the reaction of HNO_3 and O_2 with graphite.[17] The measured off-gas concentrations and generation rates were then used in a series of calculations to show that need for additional NOx abatement in H-Canyon is not likely needed.[13][17] A draft document with the calculations has been written and reviewed with the H-Canyon facility, but its conclusions have not yet been finalized.

Testing with radioactive fission products is needed. Small-scale nonradioactive simulant testing and sample analyses are complete, but the results have not yet been formally documented. Engineering-scale testing is complete using non-radioactive surrogates and sample analyses of archives samples are pending the availability of funding.

4.1.2 Technology Maturation Gap

Minor modifications to the existing engineering-scale design are recommended to finalize the baseline design and process conditions. These changes include modifications to the multi-zone oxidant feeding system and the bottom screen. The modification to the oxidant feeding will optimize the digester design and control. Modifications to the bottom screen are required to enable the system to adequately transition fuel from the bottom of the digester without transfer of excessive quantities of particulate graphite.

Finalization of analysis and reporting from simulant fission product tests is needed to provide vital data to modeling efforts and the off-gas exhaust design. Laboratory-scale tests were completed and samples analyzed. A report of the results has been drafted but not issued. Engineering-scale tests with simulant fission products were also completed, but the samples have not been analyzed. Those sample must be analyzed, and the results documented. Any concerns identified when analyses are complete will be addressed in subsequent tasks.

4.1.3 Tasks to Achieve Technical Confidence:

Task 1A: Complete engineering-scale testing with modifications to the oxidant feed system and the bottom screen design. Perform baseline testing on the entrainment behavior of simulant heavy metal fines (stainless steel powder) in the digester. Establish baseline process conditions (oxidant feed rates and digester temperatures) for subsequent demonstrations. Identify the bottom screen design that supports the need to remove fuel kernels from the bottom of the digester without excessive particulate graphite transfer. The sizes of materials that must pass through the bottom screen represent a tri-modal distribution – fines (10-100 μm), 400-600 μm, and 900 μm. Demonstration of adequate handling of bi-modal distribution (fines and 400-600 μm) is adequate to achieve technical confidence.

Task 1B: Complete analysis and documentation of laboratory-scale and engineering-scale simulant fission product studies.

Task 1C: Test the entrainment behavior of U-Th carbide and UO_2 kernel types and validate that their behavior does not represent a fatal flaw that prevents successful operation of the process. JEN has provided data on the decomposition behavior of U-Th carbide BISO particles during vapor digestion at 550 to 700 °C. The data include particle sizes for the digested fuel. The JEN results will be combined with particle fluidization data in the literature and expanded with nonradioactive simulant particle testing. The body of data will be evaluated using commercially-available computational fluid dynamics software combined with vapor equilibrium software to establish that the equipment design is adequate to manage the kernels and particulate graphite in the digester.

Task 1D: Complete a series of laboratory-scale tests to quantify the absorption of NOx gases in varying concentrations of hydrogen peroxide (H_2O_2). Assumptions have been made about the absorption of NOx prior to its emission into the H-Canyon ventilation system. The assumptions of the quantities of NOx gases absorbed by H_2O_2 must be validated to confirm that the digestion off-gas stream will not have a significant negative impact on the H-Canyon ventilation tunnel. Based on the available assumptions, current calculations show the impact to be acceptable.

Task 1E: Evaluate the feasibility of melt-dilute technology as a method for HTGR fuel final disposition. To decouple fuel-digestion from the Tank Farm, a non-aqueous method must be identified for dispositioning the fuel once the graphite has been removed. Actinide alloying has been demonstrated at INL and SRNL for disposition of metal actinides.[18] This technology was shown to be feasible at the bench scale for U-Th oxide HTGR kernels using Y, Ca, and Y-Nd with stainless steel.[19] Reductive melts of aluminum-calcium have been proposed for the conversion of U oxide to metal and incorporation into the larger Al-Ca matrix.[10] Based on the tests completed with stainless steel and U-Th oxide conversion to metal, it is expected that similar behavior will be accomplished with an Al-Ca, Al-Y, or Al-Y-Nd system.

Laboratory-scale alloys will be prepared for all three chemical systems (Al-Ca, Al-Y, Al-Y-Nd) using U-Th oxide fuel kernels. The best of the three chemical systems will be the baseline system for subsequent testing. Heavy-metal loading studies will be performed to establish the maximum concentration of U-Th that can be alloyed with the Al matrix. The U-Th oxide tests will be repeated with the presence of elemental carbon mixed with the U-Th oxide. After the potential waste forms are prepared, some will be cut open to determine extent of U-Th distribution and immobilization. Depending on the results, one or more of the samples will be analyzed using scanning electron microscopy (SEM) or energy-dispersive X-ray spectroscopy (EDS). Samples of any oxide slag will be sampled for U and Th. Samples of the aluminum matrix will be analyzed for leachability of the U and Th. The results will be documented in a technical report showing that the U and Th oxides are converted to metal and dispersed into the Al matrix adequately to support the material attractiveness requirements.

Additional tests will include nonradioactive evaluations of the impact of adding silicotitanates, silicon carbide, and zeolite (aspect of melt-dilute off-gas system) on the melt-dilute waste form. Silicotitanates are used to remove radioactive components in the off-gas scrubber solutions, zeolites remove radioactive off gas components from the melt-dilute off-gas system, and silicon carbide is a component of the HTGR fuel. Reactivity of the final Al matrix will be evaluated because of the presence of residual metal reductant alloyed with the Al. The results will be documented in a technical report showing that these compounds do not negatively impact the formation of the Al metal matrix.

4.2 Activity 2: Demonstrate Engineering-Scale Process Rates and Sustainment Conditions

Using the engineering-scale design and process conditions established in Activity 1, this activity begins to simulate production and maintenance activities. Modifications to the engineering-scale system will be made to periodically add fresh feed pebbles, remove particulate graphite and fuel kernel simulants, and regenerate the top screen. Testing will address extended-duration operations and off-normal process conditions. The need and means for conducting system maintenance will be evaluated, including strategies for periodic clean-out of digester residues.

4.2.1 Technology Development Status:

An engineering-scale digestion system exists in SRNL. The system has a valve-free design for remote operation and operates under slight negative pressure to prevent system leaks to the atmosphere. The system operates from a computer control interface, has real-time off-gas monitoring, and offers extensive data collection. It has been operated safely during a series of short-duration activities at high process efficiencies and acceptable pebble digestion rates.

4.2.2 Technology Maturation Gap

An engineering-scale pebble digestion system was fabricated and installed with a nominal process throughput of 30-50 pebbles per day. Tests to date have demonstrated the equivalent of 35 pebbles per day. Although the engineering system has been successfully designed and operated, it lacks the features and capabilities to be operated in a sustainable manner. The system must be modified for material addition and removal, screen regeneration, and periodic maintenance. With those capabilities in place, it will be possible to perform extended-duration operations and establish a more-representative throughput rate.

4.2.3 Tasks to Achieve Technical Confidence:

Task 2A: Procure simulant HTGR pebbles from U.S. vendor. This material will include pebbles that have ZrO_2 kernels instead of U-Th oxide or carbide. It may include graphite-only pebbles, depending on the inventory remaining in the SRNL stock and the relative cost of the simulant pebbles with ZrO_2 kernels. The behavior of the pebbles will be compared against graphite-only pebbles previously tested. Data already exist showing that the unirradiated fuel pebbles and graphite-only pebbles behave comparably.[15][16] If the vendor pebbles are fabricated from A3 graphite, it is highly likely that their digestion characteristics will be similar.

Task 2B: Modify the engineering-scale system – bottom product collection vessel and top-screen collection vessel – with a feature to routinely remove particulate graphite and fuel particle simulants from the primary digester. General Atomics successfully demonstrated a valve design during its HTGR fuel process program for regular removal of fuel without breaching containment.[20] A similar design will be installed and demonstrated. As needed, modifications will be made.

Task 2C: The engineering-scale system has demonstrated the successful operation of a top vent screen to regulate pressure in the digester and retain particulate graphite exiting the digester.[13] Under similar conditions for its HTGR processing systems, General Atomic demonstrated a sintered-metal backpulse filter which could be routinely regenerated with a pulse of air.[21] The engineering-scale system will be modified to regenerate the transition from the primary digester to the off-gas exhaust system. The first approach will use the existing screen design and demonstrate that it is sufficiently durable for in situ regeneration. If necessary, it will be replaced with a sintered-metal filter and demonstrated to function adequately.

TASK 2D: After the system design has been shown to adequately allow for removal of materials from the digester and regeneration of the top vent screen, extended-duration demonstrations will be successfully completed. This testing must include at least two successful 12-hour demonstrations. Longer-duration testing will be completed as part of Activity 5. Success will be based on process throughput, system performance, and operation sustainment.

Task 2E: It is expected that particulate graphite, fission products, and digested fuel will accumulate in the primary digester and off-gas system. It is incumbent upon the design to allow for periodic emptying or flushing of the primary digestion vessel and off-gas system. Clean-out strategies will be identified and at least one demonstrated. This demonstration will be completed using the engineering-scale system and graphite feeds that include pressed graphite pellets which contain fission product simulants and residual fuel particulate simulants. The goal will be to limit digester clean-out to no more than one time per month.

TASK 2F: In addition to establishing steady-state operating data, it is essential to evaluate off-normal conditions and events. These evaluations will include operating temperatures 100 °C above and below the baseline temperature range, extreme decreases or loss of HNO_3 or O_2 feed rates, surges in HNO_3 or O_2 feed rates, loss of heating, and either excessive or diminished ventilation. The off-normal conditions and events will define the operating range required for safe and secure operation, and they will demonstrate that expected off-normal conditions and events will not represent a fatal flaw with regards to operability and maintenance.

TASK 2G: At the conclusion of Tasks 2A through 2F, a team of experts will be enlisted to evaluate the equipment designs, operating methods, and potential maintenance issues from the perspective of operating and maintaining the system in a remote environment. If critical flaws in the design are noted, the engineering-scale design will be modified and shown to successfully correct the critical flaws.

4.3 Activity 3: Demonstration Critical Engineering Functions with Unirradiated Fuel

Using the engineering-scale design and process conditions established in Activities 1 and 2, this activity simulates process conditions during the digestion of actual fuel, albeit unirradiated. The activities will be completed in a radioactive chemical hood.

Vapor-phase digestion studies have been completed with full pebbles of unirradiated U-Th oxide BISO fuel.[15] These tests included collection of fuel kernels. Despite the favorable results, the results were limited in scope, and the digestion vessel design has changed significantly. Therefore, additional demonstrations are required with bounding types of full-size unirradiated fuel pebbles exposed to process conditions closer to those of the current baseline process. The results will provide baseline U-Th partitioning data and compare actual fuel behavior with the behavior of simulants.

4.3.1 Technology Development Status

Vapor-phase digestion testing with U-Th oxide BISO fuel was completed in 3Q FY2015. The results showed some recovery of fuel kernels from digested pebbles, some pyrolytic carbon layers still covering fuel kernels, some fractured fuel kernels, and minor entrainment of U-Th to the off-gas system. However, the fuel type tested is highly resistant to fuel kernel degradation, the quantities of U-Th fuel retained in the digester were not acceptable, and the digester design and conditions were significantly different from baseline conditions.

4.3.2 Technology Maturation Gap

Tests with representative fuel simulants require comparisons with actual fuel. The amount of relevant vapor digestion data for unirradiated fuel in baseline system designs is minimal. Previous data is helpful, but not enough to establish technical confidence. Although the availability of irradiated fuel is limited, some unirradiated fuel pebbles are available. Of interest are those fuel types which represent best-case and worst-case conditions. The U-Th oxide BISO fuel previously tested (with some pebbles still available at SRNL) is close to the best-case condition because the fuel kernels resist reacting with the oxidants and thus maintain their shape and integrity. Either U-Th carbide BISO or U oxide BISO pebbles are worst-case because the fuel kernels react with the oxidant and break down to smaller particulates and powders. Germany possesses some unirradiated U oxide BISO pebbles available for transport to SRNL. Successful testing with pebbles of best-case and worst-case fuel types is needed to validate critical design features of the engineering-scale system and provide data comparisons for other simulant studies.

4.3.3 Tasks to Achieve Technical Confidence:

Task 3A: SRNL will receive unirradiated fuel pebbles from Germany. The fuel pebbles must be either U-Th carbide BISO or U oxide BISO pebbles to establish engineering performance under worst-case fuel degradation.

Task 3B: Based on the design input of Activities 1 and 2, SRNL will design, fabricate, install, and operate a primary digester system that is 3 inches diameter. The system will be the same diameter as the engineering-scale system in EDL, but shorter due to the constraints of the test location. The system will include fundamental design features for venting gases from the system without excessive fuel entrainment and for removal of fuel through the bottom screens. Best-case and worst-case fuels will be tested separately and together. For each test, several graphite-only or graphite-ZrO_2 pebbles will be mixed with the unirradiated fuel pebbles to fill the digester.

The digester will be operated at or near baseline digestion conditions for sufficient time to digest all or almost all of the pebbles containing unirradiated fuel. The distribution of fuel in the digester and at various collection points outside of the digester will be quantified. The condition of fuel collected from the digester and at various collection points outside of the digester will be characterized with digital photography and SEM. Testing must establish that the performance of the critical engineering features is adequate to establish technical confidence.

4.4 Activity 4: Validate Fuel Residue and Fission Product Assumption with Irradiated Fuel Kernels

All work to date with HNO_3-O_2 vapor-phase digestion to establish fission product behavior is based on simulant studies or irradiated fuel digestion with different chemical systems. Thus far, the simulant studies with HNO_3-O_2 have significant overlap with the irradiated fuel digestion studies of other chemical systems. However, there is a high risk associated with designing the off-gas system based on simulant studies without any irradiated fuel digestion studies with HNO_3-O_2 to validate the simulant data. Achieving technical confidence requires the availability of HNO_3-O_2 digestion data with irradiated fuel. The activities to achieve technical confidence will be accomplished in the SRNL Shielded Cells facility.

4.4.1 Technology Development Status

Simulant studies with Cs, Sr, Eu (simulant for actinides), and Re (simulant for Tc) have been completed with HNO_3-O_2 vapor digestion at the laboratory[22] and engineering scales[4] at 550-850 °C. Small-scale studies with irradiated fuel have been performed at SRNL using a molten $NaNO_3$-NaOH at 700 °C[23] and at ORNL using oxygen at 700-850 °C with both carbide and oxide fuel kernels.[24][25] The correlation between the studies is good at 550-750 °C but poor at 800-850 °C.

Furthermore, vapor digestion studies with unirradiated U-Th carbide and oxide fuels have been completed in Germany and at SRNL, respectively.[15] These studies provide an understanding of the mechanical integrity of the fuel kernels during vapor digestion.

4.4.2 Technology Maturation Gap

Irradiated fuel pebbles will not be available for any stage of process development. Consequently, all engineering developments must be made with representative simulants and unirradiated fuel. Therefore, it is essential that some data be obtained with irradiated fuel to support the validity of decisions made based on studies with simulants and unirradiated fuel. The data can be obtained using irradiated fuel kernels. While not ideal, it is the best available material to help mitigate an area of high technical risk.

4.4.3 Tasks to Achieve Technical Confidence:

Task 4A. SRNL will receive two capsules of irradiated fuel kernels from Germany. One capsule will contain U-Th oxide BISO kernels and the other capsule will have U-Th carbide BISO kernels. These will be used for vapor digestion studies.

Task 4B. SRNL will design, fabricate, install, and operate a small-scale vapor digestion system at a range of temperatures (550 to 850 °C) with irradiated U-Th oxide and U-Th carbide fuel kernels. The distribution of fission products between the digestion vessel, downstream tubing, and the residual fuel will be quantified. The condition and characteristics of the final fuel will be evaluated using digital photography and SEM.

Vapor digestion of irradiated fuel kernels will correlate simulant data and validate fuel residue and fission product assumptions. The behaviors of actual and simulated fission products will be correlated to satisfy critical design and modeling inputs. Additional calculations are required to apply these data to the variety of fuel types and irradiation levels to obtain confidence that release fractions can be adequately estimated.

4.5 Activity 5: Perform Pilot-Scale and Integrated Engineering-Scale Demonstrations

The demonstration of individual technology components is a necessary aspect of technology development. The individual components of the HTGR process have been demonstrated, depending on the unit operation, at engineering scale (digester), pilot scale (fuel milling and secondary digestion), and full scale (off-gas system). However, a critical component of technical maturity is system integration. Using the primary digester design demonstrated in Activities 1 and 2, the digester will be operated in conjunction with milling and secondary digestion. Simulant pebbles with simulant fuel kernels will be digested in the SRNL EDL to demonstrate recovery of the simulated fuel kernels. Activities will include at least one 36-hour demonstration.

The selection of either a nominal 5-6" pipe or a small slab for subsequent pilot-scale testing remains open, but the pipe approach is the current assumption. Several aspects of the digester design warrant demonstration at the pilot scale ($1/10^{th}$ scale). Although it is not required for technical confidence to demonstrate pilot-scale capacity with an integrated system, it is important to validate critical engineering features at the pilot scale, such as the top vent cap, fuel collection, controlled digestion, and prototypic heating.

4.5.1 Technology Development Status

The individual components of the digestion process have been demonstrated at the engineering scale (or better) as part of this project or other related projects. The least mature of the technologies, graphite digestion, will have been matured as part of Activities 1 and 2. An engineering-scale digestion system resides in SRNL EDL for testing, although some modifications are expected based Activities 1 and 2. Milling and calcining technologies are well-established, but have not been demonstrated in conjunction with an operating digestion system.

A pilot-scale ($1/10^{th}$ Scale) type digester (5-inch Schedule 10 pipe) has been successfully operated with a properly designed top vent screen.[4] However, the process conditions were limited by heat removal from the system due to being heated by a furnace. A digester of similar size can be used to demonstrate the key engineering components, although it will be equipped with a different heating mechanism and chemical feed configuration.

4.5.2 Technology Maturation Gap

At the end of Activities 1 and 2, which will mature the engineering interfaces, the unit operations (Attachment 1) must be shown to work together at least at the engineering scale. This effort will require that SRNL obtain a milling and secondary digestion capability to evaluate the interfaces. Following Activities 1 and 2, it is also important for achieving technical confidence that the digestion process be demonstrated at pilot scale with prototypic heaters. Although an integrated pilot will be required as part of future maturation activities, it is not required for achieving technical confidence.

4.5.3 Tasks to Achieve Technical Confidence:

Task 5A: SRNL will procure a laboratory disc mill for continuous size reduction of fuel kernel simulants to less than 150-200 μm – the DWPF target is 170 μm. The mill will be combined with a transfer system to move fuel discharged from the primary digester to the mill feed hopper. The transfer and mill will be demonstrated independently and as part of the integrated engineering-scale demonstration.

Task 5B: SRNL will design and build or procure a system for continuous digestion of the graphite associated with the material exiting the disc mill. A fully-functional engineering-scale system will be demonstrated in a laboratory. Depending on the chemical digestion used in the secondary digester (e.g., if HNO_3 is used), it might happen that only the mechanical aspects of secondary digestion are active as part of the integrated engineering-scale demonstration.

Task 5C: An integrated engineering-scale demonstration will be performed using representative fuel simulants. The demonstration will include primary digestion, the first stages of the off-gas system, transfer of digested fuel to the mill, fuel milling, and at least the mechanical components of secondary digestion. Included in the testing will be an extended duration operation in which steady-state conditions are achieved in all unit operations and fuel digestion occurs.

Task 5D: A pilot-scale digester will be fabricated, likely from 5-inch pipe because three pebbles can set side-by-side in a 5-inch pipe. It will be similar to the digester previously tested but will differ in heating mechanism (prototypic), engineering features, and throughput. The digester will be operated at steady-state to demonstrate that the critical engineering components function successfully at pilot-scale. It is not a requirement that the pilot-scale system operate at full capacity or that it be incorporated as part of an integrated demonstration.

5.0 Additional High-Risk Items

Although not explicitly a part of the technology maturation, there are several high-risk technical and programmatic issues that must be resolved concurrently to achieve technical confidence.

- Complete calculation to demonstrate that C-14 release rates comply with site permits
- Calculation/modeling to predict the release of fission products if the fuel kernels are significantly size reduced by the digestion process
- H-Canyon throughput calculations using the Mod-Sim software
- Mass balance calculations and impact on site environmental permits if only AVR fuel is processed
- Confirm with high level waste contractor that proposed waste stream is compatible with their allowed chemical and physical properties, and that the waste is compatible with the waste process System Plan

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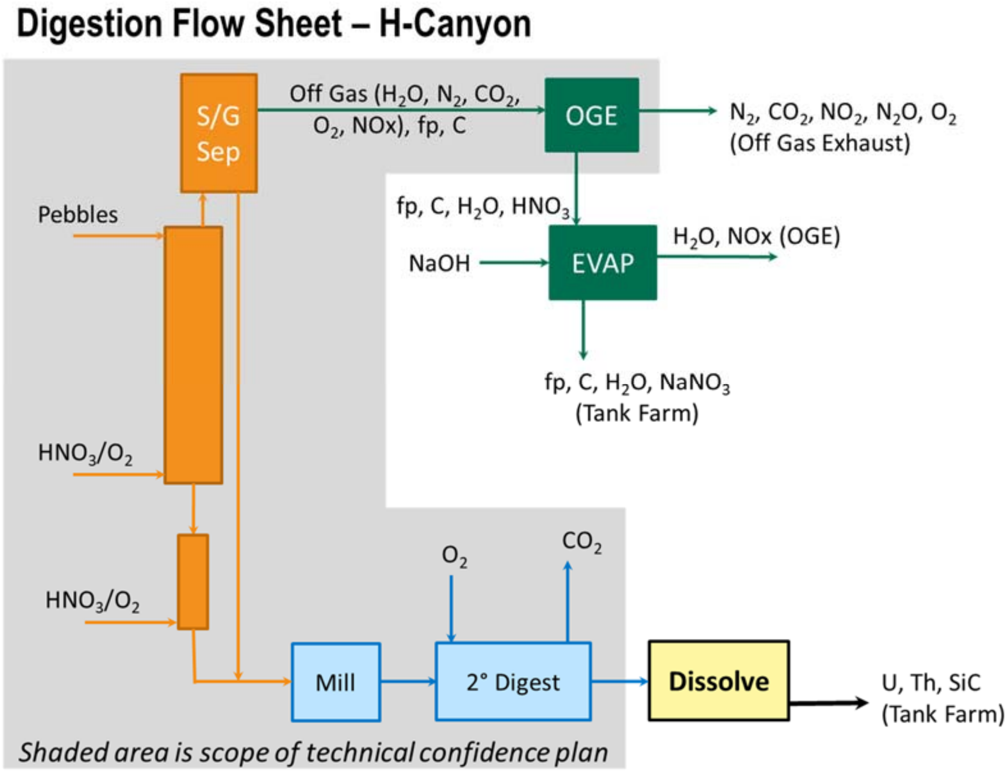
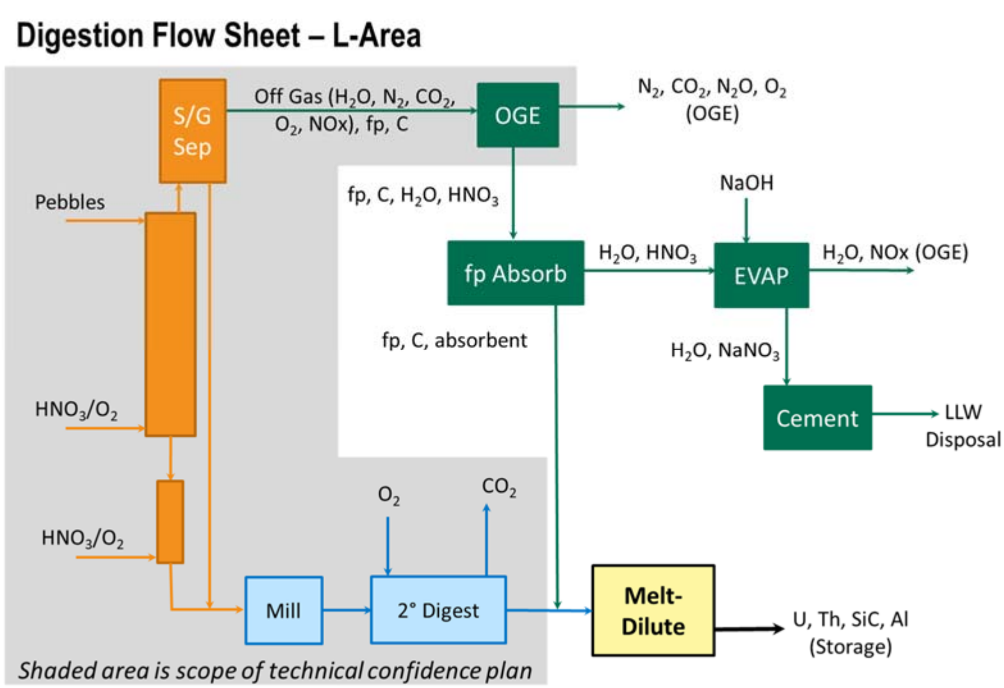
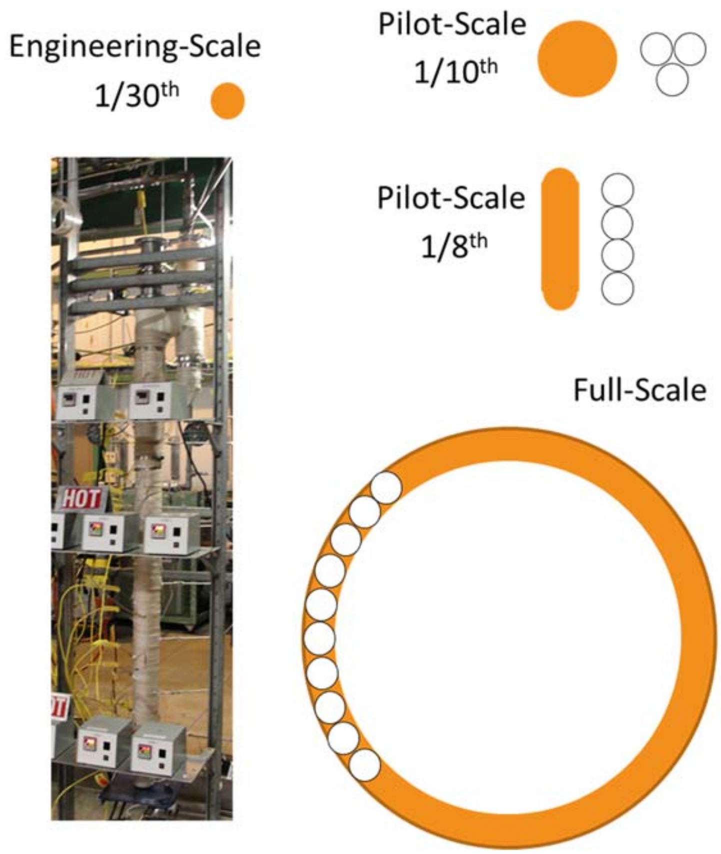
7.0 Attachments

7.1 ATTACHMENT 1 – Flowsheet Diagrams

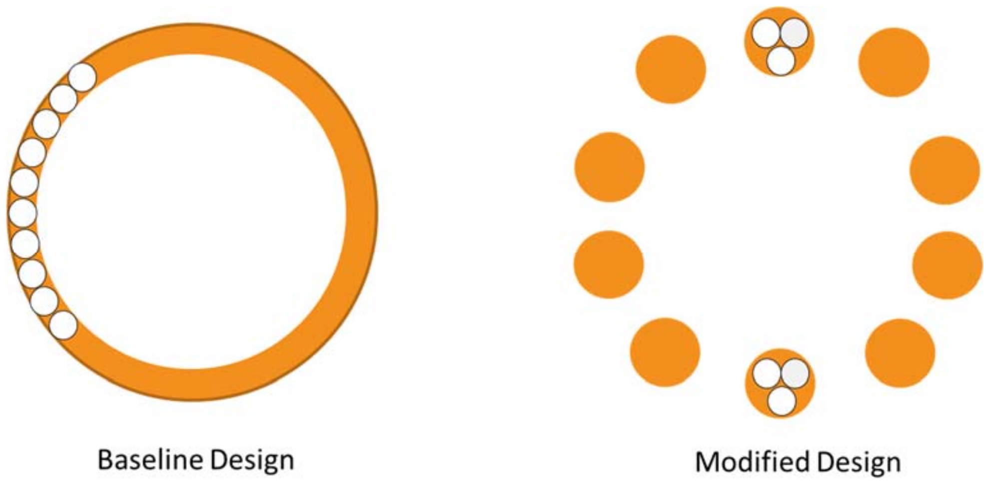




7.2 ATTACHMENT 2. SCALE OF DEMONSTRATION



BASELINE AND MODIFIED FULL-SCALE DESIGNS



7.3 ATTACHMENT 3: CRITICAL PROCESS REQUIREMENTS

Primary Digestion

- Minimization of remote equipment, especially mechanical devices.
- Heater design requirements are understood, and feasibility of the heating method is demonstrated.
- Compatible with remote operation, remote maintenance, and standard Canyon operating principles
- Minimizes reliance on the H-Canyon crane.
- System can be installed and maintained in H-Canyon.
- Graphite digestion nominal temperature maintained between 625 and 750 °C.
- Validate digestion thermal properties, including reactions and heat transfer, to support thermal modeling.
- Graphite digestion and off-gas equipment operate under negative pressure, to the extent possible, with operational stability under surge conditions.
- Adequately characterize the chemical, physical, and radioactive properties of the various fuel feed materials.
- Develop material balances and high-level modeling that predict system mass and concentration impacts associated with process baseline assumptions.
- Design digester system needs to attain nominal 1,400 pebbles per day throughput to account for ~70% operating utility.
- Primary digester design is compatible with the variety of AVR and THTR BISO and TRISO fuel types
- Develop an inventory of gaseous and particulate reaction products and fission products
- Design approaches must be compatible with geometrically favorable design regarding nuclear criticality safety.
- Confirm that selected materials of construction are compatible with the process.
- Provide features for monthly clean-out of the digester system and verify that the need for clean-out is infrequent (i.e., no more than once per month).
- Confirm that semi-volatile fission product releases from the process have similar particulate distribution and properties to those assumed in DWPF (since the proposed offgas system resembles that of DWPF).
- Limits releases of Cs and other semi-volatile fission products.
- Avoids carryover of heavy metal oxides (U and Th) to less than 0.15% of element basis.
- Provide digester top screen/filter design so that graphite releases from the digester are less than 0.2% of original carbon.
- Provide ability for in situ regeneration of the top screen/filter.
- Design allows for continuous pebble addition.
- Design bottom perforated plate to allow fuel kernels to pass without having excessive fines pass through the screen or build up on the screen.
- Operate above the auto-ignition temperature for potential flammable gases or maintain system below 25% of LFL for CO.
- Provide instrumentation to control process parameters and to detect when the system is operating outside normal operating parameters.
- Identify bounding conditions for normal operation of the system and validate safe and reliable operation of the system there within.
- Provides necessary data to develop mass balances, energy balances, and high-level modeling.
- Develop a design with heat removal that does not exceed the cell temperature limits for H-Canyon.

Fuel Milling

- Provide system for transfer of either BISO kernels or TRISO coated particles from the primary digester to the fuel mill.
- Crush TRISO and BISO fuel kernels to a target distribution with minimal quantities above 150-200 μm and with minimal fines less than 10 μm.

- Demonstrate mill that can continuously receive fuel kernels from the primary digester and continuously deliver milled fuel to the secondary digester.
- Fuel milling method can readily achieve crushing target rate of 12.5 kg/h.
- Fuel milling compatible with Canyon operation and maintenance requirements.
- Design to accommodate potential pyrophoricity of milled carbide fuel kernels.

Secondary Digestion

- Demonstrate method for secondary digestion that meets the interface requirements with fuel milling.
- Secondary digester design is compatible with the variety of AVR and THTR BISO and TRISO fuel types
- Demonstrate method for secondary digestion that minimizes material losses.
- Identify and validate reaction mechanisms and rates for secondary digestion.
- Secondary digestion is compatible with Canyon operation and maintenance requirements.
- Demonstrate secondary digestion that operates continuously or semi-continuously

Off-Gas System

- The off-gas system must use the same operating principles and unit operations as the DWPF off-gas system to benefit from its high technical maturity.
- Demonstrate interface between the primary digester and the off-gas system.
- Demonstrate an off-gas system approach that meets acid, liquid, and particulate removal requirements and that presents a low potential for mis-operation and pluggage.
- Demonstrate an off-gas system that uses the same operating principles and unit operations as the DWPF off-gas system.
- Provide features for continuous or periodic clean-out of the off-gas system.
- Off-gas system meets the requirement of <125 ppm nitric acid to the H-Canyon tunnel.

7.4 ATTACHMENT 4: TECHNICAL CONFIDENCE AREAS

Preliminary Technical Risks HTGR Project – Based on Option 1	Risk Level H/M/L	WFO Actions to Address	Contract Phase I Actions to Address Prior to Authorizing Shipment	Delta Estimate and Notes
1) Design Challenges	High			
a. Offgas – NOx/acid abatement, release limits	High			
i. New risk of acid interaction with tunnel rebar	High		Yes – Integrated Test	Delta – Add cost for caustic addition/acid scrubbing in Offgas system plus added waste volume. Note: Mitigate with Integrated (1/30 scale) System Test
ii. Assume DWPF size system	High		Yes – Integrated Test	Note: Mitigate with Integrated (1/30 scale) System Test
iii. C-14 Release limit compliance	High	Yes – AVR – only Calc	Yes – Integrated Test Yes – AVR – only Calc	Note: Could impact throughput for AVR only case. Note: Need to confirm data includes graphite <u>and</u> burnup generated C-14 Note: Mitigate with Integrated (1/30 scale) System Test and calculation for AVR-only.
b. Digestion to offgas Interfaces (Address kernel flow, pluggage, etc.)	High			
i. Chem and particle motion in airflow	High	Yes – Calc	Yes – Calc Yes – Integrated Test	Note: Calculate model for integrated test then validate/adjust after test. Identifies physical sizing and restrictions in system.
ii. Fission Product Release if kernels disintegrate in digester	High	Yes – Calc/Model	Yes – Calc/Model	Note: Near Term - Mitigate by calculation only. Model offgas FP offgas release vs. burnup. Long Term – Test with irradiated pebbles after authorized.
c. Digestion to kernel removal interfaces	High			
i. Kernel behavior and flows		Yes – Calc	Yes – Calc Yes – Integrated Test	Note: Calc to predict particles and FP flows in digester and interface then validate/adjust after test.
d. Periodic clean out of digestion system – method and frequency	High		Yes – Integrated Test	Note: Mitigate with Integrated (1/30 scale) System Test

Preliminary Technical Risks HTGR Project – Based on Option 1	Risk Level H/M/L	WFO Actions to Address	Contract Phase I Actions to Address Prior to Authorizing Shipment	Delta Estimate and Notes
e. Address behavior of U-Th Carbide & UO_2 kernel types	High	Yes – U-Th Carbide Test in Germany	Yes – UO_2 pebbles in Integrated Test	Note: Need UO_2 pebbles shipped Note: Closely related to b, c, and d. Note: U-Th Carbide to be tested in Germany ($25K). UO_2 kernels to be tested at SRNL with unirradiated pebbles in Integrated Test. Note: Could turn to powder & plug digester Note: Heavy particle behavior in offgas not well understood
2) Safety Analysis – Storage – Long term behavior in Castors in storage	High	Yes - PCHAP	Yes – CHAP/DSA	Delta – Develop Surveillance Program or equivalent & Mitigation Plan for leakage. Note: Evaluate BAM/GNS elastomer seal papers Note: Mitigation Plan for seal leak
3) DOT approval/revalidation of CASTORs	High	Yes	Yes	DOT CAC needed to ship from Germany
4) Throughput (Process scale-up, crane availability, cask handling)	High	Yes – Mod-Sim Runs	Yes – Mod-Sim Runs	Delta: Could extend lifecycle assumption to mitigate Note: Near term crane time-motion vs. COIN & Mod-Sim
5) Canyon Availability for processing (lifecycle risk – **Accepted by DOE)**	High	X	X	
6) Tank Farm/HLW Availability – HLW transfers to HTF end FY30 – Sludge Batch window for FGE is limited	High		Yes – Determine Loading Limitations (AVR and combined) Yes – Decide and estimate HLW Strategy	Delta: Requires HLW Decoupling or potential SB/DWPF extension and cost increase. Consider impact if FCA is run in HC.
7) Review Canyon Impact Assumptions from outage, installation, and cell cleanup/prep. Also includes operations impact on LC.	High		Yes – Validate estimate based on new information	Note: Check validity of 2-year cost/schedule impact estimate. Also address section 5 hi-rad waste box disposal.
8) Mass balance if AVR – only is processed (higher burnup – more FP)	High	Yes - Calculation		Delta: Mitigated by calculation Note: Original mass balance used average of AVR & THTR Note: Could extend LC
9) Approve/Confirm Defense Determination	High			Note: DOE action needed to confirm or develop. Needed for DWPF processing.

Preliminary Technical Risks HTGR Project – Based on Option 1	**Risk Level H/M/L**	**WFO Actions to Address**	**Contract Phase I Actions to Address**	**Delta Estimate and Notes**
10) Approve/Confirm WIR approval	High			Note: DOE action to confirm for any LLW generated
11) Design Challenges – Medium	Medium			
12) Cask handling, offloading to pad, movement within H-Canyon and removal of canisters, cutting off canister tops, maintainability	Medium		Yes – Initiate conceptual Design	Note: Engineering Design challenges but not "high risk". Concepts developed in 2015 need review with HC Ops/Eng.
a. Pebble failures (removal of bonded and/or broken pebbles from the canisters)	Medium		Yes – Initiate conceptual Design	Note: Engineering Design challenges but not "high risk"
b. Ability to heat and regulate digester temperature	Medium		Yes – Vendor and concept only	Note: Research available vendor specs and hardware for this application
c. HLW Waste Acceptance – crit, particulate, solubility, corrosion	Medium		Yes – SRR to review	
13) Interfaces – Treatment System and H-Canyon (ARU, solution storage, concurrent with SNF processing)	Medium		Yes – Review with Mod-Sim	Note: Could impact LC or cost for more hardware. Evaluate with Mod-Sim
14) Security/MC&A Strategy – Casks and kernel processing (revisit 2014)	Low			Note: MC&A strategy not mature in processing. Instruments, P&ID needed. Note: Classification of technology and cask attractiveness confirmed in meeting (Houck, Baker, Maxted, Amos, Bates) on 8/30.
15) Design Challenges - Low	Low			
16) Crusher/Mill – Material holdup, cleaning, maintaining	Low			Note: Review GA design
a. Safety Analysis – Processing	Low		Yes - PCHAP	
17) General Project Risks (Resources, Commodities, Procurement, craft)	Low			
18) Airborne Canister Unloading - Fission Products (Degraded kernels, unloading could create particulate dust, air monitoring)	Low			

7.5 ATTACHMENT 5: CONCEPTUAL SCHEDULE OF PRIMARY ACTIVITIES

Activity	Description	Months																	
		1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18
1	Baseline conditions and design																		
2	Process rates and sustainment																		
3	Unirradiated fuel testing																		
4	Irradiated kernel testing																		
5	Integrated demo / pilot demo																		

Key Assumptions:

3. Unirradiated fuel pebbles received with first 5 months
5. Irradiated kernels received in first 10 months

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