

We put science to work.™



**Savannah River
National Laboratory™**

OPERATED BY SAVANNAH RIVER NUCLEAR SOLUTIONS

A U.S. DEPARTMENT OF ENERGY NATIONAL LABORATORY • SAVANNAH RIVER SITE • AIKEN, SC

Scale-Up Maturation Plan for Digestion of Graphite Fuel Pebbles

Official Use Only
~~Pre-decisional Draft~~

R. A. Pierce

K. M. Fox

June 2014

SRNL-RP-2014-00464, Revision 0

SRNL.DOE.GOV

DISCLAIMER

This work was prepared under an agreement with and funded by the U.S. Government. Neither the U.S. Government or its employees, nor any of its contractors, subcontractors or their employees, makes any express or implied:

1. warranty or assumes any legal liability for the accuracy, completeness, or for the use or results of such use of any information, product, or process disclosed; or
2. representation that such use or results of such use would not infringe privately owned rights; or
3. endorsement or recommendation of any specifically identified commercial product, process, or service.

Any views and opinions of authors expressed in this work do not necessarily state or reflect those of the United States Government, or its contractors, or subcontractors.

Printed in the United States of America

**Prepared for
U.S. Department of Energy**

Keywords: *HTGR, graphite, molten salt*

Retention: *Permanent*

Scale-Up Maturation Plan for Digestion of Graphite Fuel Pebbles

R. A. Pierce
K. M. Fox


June 2014

Prepared for the U.S. Department of Energy under
contract number DE-AC09-08SR22470.

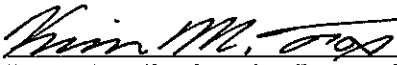


REVIEWS AND APPROVALS

AUTHORS:




R. A. Pierce, Separation & Actinide Science
6/24/2014
Date

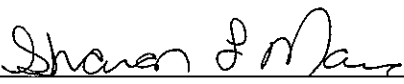


K. M. Fox, Engineering Process Development
6/24/2014
Date

APPROVAL:



T. B. Brown, Manager
Separation & Actinide Science
6/24/14
Date



S. L. Marra, Manager
Environmental & Chemical Process Technology Research Programs
6/24/14
Date



W. F. Bates, Manager
Nuclear Materials Program Integration
6/26/14
Date

EXECUTIVE SUMMARY

This scale-up maturation plan describes a multi-year program addressing technical considerations related to the processing of HTGR fuel. The objectives of this plan are to provide technical justification for the selected approach to HTGR fuel processing, review the progress of development efforts to date, identify technology needs and risks, and prioritize a plan for addressing those technology needs and mitigating risks. The focus of this plan is specifically on the removal of graphite and SiC from the fuel pebbles and kernels. Integration with other aspects of the HTGR fuel processing program is considered, with recognition that those other aspects are under concurrent development.

A review of the status of knowledge, technology needs, and risks associated with several aspects of the molten salt graphite digestion concept is provided. This review is used to generate a prioritized list of near term tasks that will address technology needs and reduce risks. Specific tasks to be addressed by SRNL in fiscal year 2014 are:

1. Determine the expected inventory of gaseous reaction products and fission products
2. Determine the viability of salt regeneration
3. Develop conceptual off gas system
4. Develop a bench scale, integrated demonstration platform
5. Demonstrate methods of fuel recovery from the molten salt
6. Demonstrate SiC digestion in “spent salt” using unirradiated fuel kernels
7. Develop targeted salt compositions at completion of graphite digestion
8. Determine ^{(b)(3), (4)} salt rheology and thermal properties
9. Develop a thermal profile for the overall graphite digestion process cycle
10. Complete a thermal model for the overall graphite digestion process cycle
11. Develop mass and energy balances

Detailed planning for each task will be addressed via task technical and quality assurance plans, as appropriate. It is premature to address some of the identified technology needs and risks at this point in the development process, given the conceptual state of the fuel processing program. Therefore, this scale-up maturation plan will be reevaluated after the completion of the near term tasks, and more frequently if necessitated by the outcomes of those tasks. The direction and prioritization of the remaining tasks will be reevaluated based on newly developed knowledge, as well as developments in other aspects of the overall fuel processing program. A multi-year commitment of resources and funding is needed for complete maturation of this technology.

TABLE OF CONTENTS

| | |
|---|------|
| LIST OF ABBREVIATIONS..... | viii |
| 1.0 Overview..... | 1 |
| 2.0 Background..... | 2 |
| 2.1 Basic Assumptions and Expectations..... | 2 |
| 2.1.1 Technology Needs..... | 2 |
| 2.1.2 Risks Associated with Assumptions and Technology Needs..... | 2 |
| 2.2 Pebble Bed Fuel Recovery Concepts..... | 3 |
| 3.0 Baseline Design Basis..... | 4 |
| 3.1 Graphite-Digestion Chemistry..... | 5 |
| 3.1.1 State of Knowledge..... | 5 |
| 3.1.2 Technology Needs..... | 7 |
| 3.1.3 Risks..... | 8 |
| 3.2 Salt Regeneration..... | 8 |
| 3.2.1 State of Knowledge..... | 9 |
| 3.2.2 Technology Needs..... | 9 |
| 3.2.3 Risks..... | 10 |
| 3.3 Fuel Recovery..... | 10 |
| 3.3.1 State of Knowledge..... | 10 |
| 3.3.2 Technology Needs..... | 10 |
| 3.3.3 Risks..... | 11 |
| 3.4 Off-Gas Generation and Handling..... | 11 |
| 3.4.1 State of Knowledge..... | 11 |
| 3.4.2 Technology Needs..... | 12 |
| 3.4.3 Risks..... | 12 |
| 3.5 Waste Generation and Disposition..... | 13 |
| 3.5.1 State of Knowledge..... | 13 |
| 3.5.2 Technology Needs..... | 13 |
| 3.5.3 Risks..... | 13 |
| 4.0 Equipment Design Parameters..... | 14 |
| 4.1 General Design Considerations..... | 14 |
| 4.1.1 State of Knowledge..... | 14 |
| 4.1.2 Technology Needs..... | 14 |
| 4.1.3 Risks..... | 15 |

4.2 Graphite Digestion Vessel and Basket 15

 4.2.1 State of Knowledge..... 15

 4.2.2 Technology Needs 15

 4.2.3 Risks 16

4.3 Digestion Vessel Heating Method..... 16

 4.3.1 State of Knowledge..... 16

 4.3.2 Technology Needs 17

 4.3.3 Risks 17

5.0 Scale-Up Maturation Plan for Molten Salt Graphite Digestion Technology 18

 5.1 Near Term Scale-Up Maturation Tasks..... 18

 5.2 Longer Term Scale-Up Maturation Tasks 19

 5.3 Prioritization of Scale-Up Maturation Tasks for Fiscal Year 2014..... 20

 5.4 Quality Assurance 21

6.0 Summary and Path Forward..... 22

7.0 References..... 23

LIST OF ABBREVIATIONS

| | |
|-------|---|
| DOE | U.S. Department of Energy |
| HEU | Highly Enriched Uranium |
| HLW | High Level Waste |
| HTGR | High Temperature Gas Cooled Reactor |
| LEU | Low Enriched Uranium |
| MC&A | Material Control and Accountability |
| OGE | Off Gas Exhaust |
| SRNL | Savannah River National Laboratory |
| SRS | Savannah River Site |
| TTQAP | Task Technical and Quality Assurance Plan |

1.0 Overview

This scale-up maturation plan describes a multi-year program addressing technical considerations related to the processing of high temperature gas cooled reactor (HTGR) fuel. The objectives of this plan are to provide technical justification for the selected approach to HTGR fuel processing, review the progress of development efforts to date, identify technology needs and risks, and prioritize a plan for addressing those technology needs and mitigating risks. The focus of this plan is specifically on the removal of graphite and silicon carbide (SiC) from the fuel pebbles and kernels. Integration with other aspects of the HTGR fuel processing program is considered, with recognition that those other aspects are under concurrent development.

2.0 Background

The United States Department of Energy (DOE) is considering accepting and processing used nuclear fuel from HTGR reactors in Germany. The fuel is in the form of graphite spheres, called pebbles, each of which contains thousands of small kernels containing uranium and thorium oxides. The fuel contains uranium enriched in the U.S. Repatriating this fuel would reduce the global amount of highly enriched uranium (HEU) in civil commerce, thereby reducing proliferation risks. After receiving the fuel, DOE has several potential options for reuse or disposal of the material:

- Dissolution, purification, and down blending of the highly enriched uranium to low enriched uranium (LEU) for reuse as a reactor fuel may be possible.
- Vitrification in the Savannah River Site Defense Waste Processing Facility may allow for disposal of the waste without down blending.
- Separating the uranium, down blending to LEU, and solidifying may allow for the LEU to be sent to an appropriate uranium disposal site.
- Direct disposal of undissolved kernels may be an option with development of an appropriate waste form.
- Other disposal options that do not require recovery of the fuel kernels from the graphite pebbles may be developed.

2.1 Basic Assumptions and Expectations

The following assumptions and expectations define the basis of this scale-up maturation plan:

- Recovery of irradiated fuel from the HTGR graphite pebbles is assumed to be the selected processing option.
- The full inventory of irradiated fuel from the German reactors is assumed to have been received at SRS prior to start of processing.
- The desired processing rate is assumed to be 1000 pebbles per day, for a graphite digestion campaign duration of about three years.
- The processing method developed is expected to be applicable or adaptable to other missions beyond irradiated fuel from the German AVR and THTR-300 reactors.

2.1.1 *Technology Needs*

There are several technology needs associated with the basis of this scale-up maturation plan. Some of these will be addressed through technical development efforts described later in this document, while others are dependent on later administrative decisions and on the progress of concurrent DOE missions.

- The fate of the recovered fuel has not yet been determined, therefore, the optimal form of the recovered material is undefined.
- The feasible or most efficient processing rate for the HTGR fuel is not known.
- The actual dates when the material and facilities are available to begin processing are not yet defined.
- The availability of SRS systems (e.g., H-Canyon, liquid waste processing, HLW immobilization, LAW disposal) when the processing of the HTGR fuel begins and their availability for the duration of the campaign is unknown.
- The optimum configuration for the process to digest graphite has not been established.

2.1.2 *Risks Associated with Assumptions and Technology Needs*

The development process will proceed with the acknowledgement of the assumptions and technology needs described above. There are risks associated with this approach, and an attempt has been made to identify some of those risks here. Decisions or discoveries that may change the direction of the maturation plan as it progresses were considered to be risks to the program. These include:

- Development efforts or administrative decisions may result in an alternative disposal method that does not require recovery of the fuel from the graphite pebbles.

- The feasible or most efficient throughput rate, and therefore the length of the processing campaign, will likely change as the process is further developed.
- The inventory of used fuel received may change, which could impact the process if compositions are different than expected.

2.2 Pebble Bed Fuel Recovery Concepts

Several methods and processes for the reprocessing of used HTGR fuel have been explored over the past six decades. The general objective is to remove the uranium and thorium from the used fuel for reprocessing or disposal. The process is challenging in that removal of graphite, pyrolytic carbon, and SiC from the fuel is difficult. These materials are utilized in the design of the fuel since they are resistant to mechanical, thermal, and chemical degradation. These properties, while beneficial for reactor operation, complicate reprocessing of the fuel.

Reprocessing methods for HTGR fuel generally involve removal of the graphite, pyrolytic carbon, and SiC layers from the fuel using mechanical or chemical methods, or a combination of both. A burning-leaching process has been demonstrated at the laboratory scale.^[1] A preliminary size reduction (crushing) step is followed by burning of the graphite and pyrolytic carbon using an oxygen-containing gas mixture in a fluidized bed of alumina powder. The burning rate is controlled by varying the composition of the gas. The off gas is treated to capture fission products and C-14. The alumina bed is acid leached after burning. The resulting solution is fed to a solvent extraction process to recover the uranium and thorium. The alumina bed remains contaminated with some amount of fuel and fission products and must be treated for disposal.

A grinding-leaching process has also been demonstrated in laboratory scale experiments.^[1-2] This process again begins with a crushing step to reduce the size of the material. Additional crushing and grinding steps are then used to fracture the kernel coatings, exposing the fuel for leaching. The fuel within the kernels must be exposed for proper leaching of the material; therefore, the grinding process becomes increasingly difficult when smaller sized kernels are used. Boiling HNO₃ is used to leach the uranium and thorium from the fractured kernels after grinding. The resulting solution is fed to a solvent extraction process to recover the uranium and thorium. This leaves a bed of graphitic residue contaminated with remaining fuel and fission products. Additional washes can be used to remove some of the remaining fuel, although full recovery is unlikely. The bed remains contaminated and must be treated for disposal.

Other novel methods for removal of fuel kernels from the graphite pebbles have been investigated more recently, such as electrochemical disintegration and high voltage discharge fragmentation. However, these methods are at very early stages of development and concepts for scaling beyond a single pebble are not yet available.^a

As mentioned above, reprocessing of HTGR fuel is challenging due to the nature of the materials used in the fuel. High level reviews of potential reprocessing routes have been provided in the past, for example by Lotts, et al.^[3] and by Del Cul.^[4] Methods that involve crushing and grinding are problematic due to the production of contaminated dust, particularly as particle size is further reduced. Equipment wear and maintenance in a remote environment can also be problematic due to the high hardness of graphite and SiC. The generation of contaminated residue and waste is a general issue for all reprocessing methods. Off gas generation is also a general issue, and is more so for high temperature processes. Scale up of the processes to a production level is a challenge for all of the methods, particularly those that are more novel. An ideal process would eliminate dusting, minimize mechanical wear and complexity, simplify off gas treatment, and minimize waste generation.

^a von Lensa, W., "Treatment and Disposal of Irradiated-Graphite & other Carbonaceous Waste," *slide presentation*, Forschungszentrum Jülich, December 6, 2012.

3.0 Baseline Design Basis

As stated earlier, the objectives of this scale-up maturation plan are to provide technical justification for the selected approach to HTGR fuel processing, review the progress of development efforts to date, identify technology needs and risks, and prioritize a plan for addressing those technology needs and mitigating risks. (b)(3), (4)

At a high level, selection of (b)(3), (4) is based on the following assumptions:

- (b)(3), (4) is the best available technology for graphite and silicon carbide removal from the irradiated HTGR fuel.
- Graphite and SiC are highly abrasive. Therefore, advantages of (b)(3), (4) over other methods include fewer tribological concerns associated with machinery in a remote environment (as compared to grinding and crushing), and less concern with dusting (as compared to crushing and thermal oxidation).
- DOE experience with chemical processes (e.g., H-Canyon), and melting with complex off gas treatment (e.g., the Defense Waste Processing Facility) in high radiation, remotely handled environments is more adaptable to the molten salt graphite removal concept than to other methods.
- The irradiated pebbles in their current form can be processed directly, without any prior preparation (e.g., size reduction).
- The product produced by (b)(3), (4) (i.e., fuel kernels) is in a form that is compatible with downstream processing or disposal.
- (b)(3), (4) can be scaled to the desired processing rate of 1000 pebbles per day.

SRNL has developed a conceptual process for graphite digestion for HTGR fuel reprocessing. This concept is considered to be the baseline design for the purposes of scale-up maturation. It is acknowledged that this baseline may change as the technology is matured, and that the full scale process may differ significantly from this baseline. The baseline process is described in the following paragraphs.

Prior to graphite digestion, canisters of fuel elements must be brought into the facility, the canisters cut open, and the contents unloaded into a feed system. The feed system meters the fuel elements into a charge basket and the charge basket is inserted into the digestion vessel. The basket design for the HTGR pebbles will probably not be designed to retain the individual fuel kernels, which can be as small as 0.20 mm. Rather, the fuel would be recovered by dissolving the salt in an aqueous medium and filtering the fuel. Prior to the digestion vessel being ready to digest the fuel elements, salt must be loaded into the process vessel and brought to temperature. Other process operations, such as off gas exhaust (OGE), must be connected to the process vessel.

Digestion of graphite addresses approximately 97% of the mass associated with HTGR fuel. While graphite digestion is the principal process under development, it is not independent of other unit operations and activities. In a non-radioactive, hands-on operation, graphite digestion would likely be the rate-limiting operation. However, in a remote-handle operation, other factors could greatly affect overall processing rates.

At the conclusion of graphite digestion, the salt must be removed from the process vessel. It is proposed that the vessel be cooled first and then the salt dissolved in an aqueous solution. The salt dissolution step is likely to be lengthy step. After the salt has been dissolved, the solution and all residues are filtered to retain any U-Th. The filtering mechanism is an approach that will likely be flexible enough for the recovery of intact fuel kernels, fractured fuel kernels, and digested UO₂ kernels. The filter becomes a

candidate for further drying and storage. (b)(3), (4)
[Redacted]
[Redacted] Afterward, the salt solution pH is adjusted back to caustic for transfer to the SRS liquid waste system. The process vessel must be prepared for a subsequent process cycle.

There are several risks associated with the baseline process and its associated assumptions, the most significant being that the molten salt concept is unable to meet the needs of the program. A detailed discussion is provided in the following subsections.

3.1 Graphite-Digestion Chemistry

A detailed knowledge of the chemical reactions among graphite, SiC, fission products, impurities, (b)(3), (4) [Redacted] required for design of the process. The reactions and their rates as a function of salt composition, temperature, salt viscosity, and graphite surface area must be understood. The required heat input and the heat generated as a function of the reactions must be known. Scaling factors for transitioning the process from the lab scale to the pilot and full scale need to be determined.

3.1.1 *State of Knowledge*

SRNL is developing a (b)(3), (4) [Redacted] process for the removal of graphite from HTGR fuel.^[5] (b)(3), (4) [Redacted]

[Redacted]

(b)(3), (4) [Redacted]. The occurrence of these two reactions has been verified using TGA-MS, small-scale off-gas experiments, and salt analyses from larger-scale experiments. Based on calculations using the standard heats of formation, the two reactions (Equations 1 and 2) and the overall reaction (Equation 3) are exothermic.



The addition of NaOH (b)(3), (4) [Redacted] allows for the oxidation of SiC layers in TRISO fuels. Removal of the SiC layers can occur in parallel with removal of graphite if NaOH is added to the salt, or can be performed as a separate step later in the process. (b)(3), (4) [Redacted]

(b)(3), (4) [Redacted]

(b)(3), (4)

(b)(3), (4)

It is speculated that graphite fines occasionally accumulate at the surface of the molten salt without reacting immediately. The accumulated fines occasionally react in a short time frame causing an accelerated release of gas, a brief temperature increase, and entrainment of salt and graphite fines in the off gas. The graphite fines have been positively identified as part of the salt in the off gas.^[10]

In a later report, SRNL demonstrated the digestion of individual, full graphite pebbles at the laboratory scale.^[10] Their findings are summarized below:

- (b)(3), (4)
 - (b)(3), (4)
- Dissolution of the pyrolytic carbon layers of the kernels was demonstrated in testing of pebbles containing unirradiated fuel.
- (b)(3), (4)
- (b)(3), (4)
- The need for a mechanism of dissipating the heat of reaction was demonstrated.

In general, the reaction scaled without issue from gram-scale tests to full-pebble digestion. It is believed that the effects of further scaling for the reaction can be minimized through equipment design. Each HTGR pebble has a volume of ~103 mL but requires a salt volume of ~1100 mL for complete graphite digestion. As a result, the charge basket can be designed such that multiple pebbles can be spaced within the molten salt to behave somewhat independently of each other. This would not be the case if pebbles were loaded indiscriminately into a large, single-chamber basket.

Initial studies digesting irradiated fuel kernels provided insight into the partitioning of fission products among the kernels, salt, and off gas.^[11] (b)(3), (4)

In all four kernel digestion tests, the kernels were intact after the molten salt digestion portion of the test protocol.

As expected, Cs-137 and Sr-90 comprise the great majority of the activity (99.7%) and a most of the mass (67.6%) of the radioisotopes tracked. Appreciable quantities of Np-237 (20.1%), Cs-135 (10.5%), Tc-99 (2.96%), and Am-241 (0.80%) were also detected, although the specific activity of these isotopes is small compared to Cs-137 and Sr-90. All of the other measureable radioisotopes were present at less than 0.05% of the mass.^[11]

Other than Cs-137 and Cs-135, the fission products were retained almost completely in the digestion salt or the undigested portion of the particle. Approximately 60% of Cs-137, 25% of Sr-90, and 5-30% of U were leached from the fuel kernels into the molten salt, as were appreciable quantities of other non-volatile fission products. Only Cs-137, Cs-135, and Ra-224 were positively measured in the OGE scrubber solution at greater than 0.1 wt % of the total estimated radioisotope inventory for each specific radioisotope.

3.1.2 Technology Needs

There are several remaining technology needs associated with the graphite and SiC reaction (b)(3), (4) including:

- The complete reaction chemistry has not yet been determined.
 - Selection of the salt chemistry and reaction conditions is dependent on other aspects of the process design (e.g., batch or continuous processing, BISO or TRISO kernels, etc.).
 - As will be discussed below, there is the potential for regeneration of the molten salt during or after the graphite digestion. If this process is shown to be successful, the appropriate time of regeneration and degree of regeneration need to be determined.
 - As will be discussed below, off gas requirements have not yet been defined. It may be appropriate (b)(3), (4) rather than send it to the off gas system, although the expectation is that Saltstone requirements greatly limit the acceptable quantity of C-14 retention.
- No testing has been conducted with full pebbles containing irradiated fuel.
 - Potential reactions involving the irradiated fuel and fission products have not yet been identified.
- The complete reaction temperature profile has not yet been determined.
 - In a batch-reaction approach, testing starts at the lower reaction temperature when the graphite surface area is highest and increases the temperature over time to accelerate the digestion rate as the graphite surface area diminishes. Another approach may be to feed the reaction vessel continuously such that a consistent surface area of graphite is maintained.
- A thermal model for the pebble digestion has not been completed. This model is needed to support scale-up and system design.
 - The graphite digestion reaction is exothermic. It is expected that the system must be capable of releasing significant heat in order to control the reaction temperature.
 - The reaction has been measured as a function of temperature and appears to increase linearly as a function of temperature.^[5]
 - The heat loss from laboratory scale tests was limited and thermal spikes (b)(3), (4) were common (although not instantaneous) due to the heat of reaction. It is not known whether a design feature can be developed that will mitigate the transient surface reactions observed in bench scale testing.^[10]
 - Heat transfer properties of the pebbles, kernels, and molten salt need to be determined as inputs to the model.
 - The necessary degree of temperature uniformity needs to be defined.
- Mass and energy balances for the conceptual process have not yet been developed.
- SiC dissolution from TRISO kernels in a graphite pebble has not yet been demonstrated.
 - SiC dissolution has been demonstrated at the lab scale only with monolithic pieces of SiC.^[5]

- The SiC in the pebbles containing TRISO kernels must be removed to completely digest all graphite and expose the fuel for later dissolution. (b)(3), (4)
- If NaOH is added earlier in the reaction, CO₂ generated by the graphite digestion reaction will be absorbed by NaOH to form Na₂CO₃, which is likely to decrease the effectiveness of the graphite oxidation.
- The mass of NaOH required to dissolve a given mass of SiC has not been established.
 - Salt rheology as a function of composition and temperature needs to be determined. These data will help define the expected reaction rates, serve as inputs to the thermal model, and impact equipment design.
 - Potential effects of impurities in bulk salts on the digestion reactions have not been evaluated. Testing to date has utilized reagent grade salts.
 - The potential for foaming of the melt due to gas generation and the changing viscosity (b)(3), (4) needs to be better determined.
 - Preliminary studies using dip rod measurements offered no indication of foaming within the vessel during single pebble dissolution tests.^[10] However, single pebble digestion may not accurately predict the behavior of multiple pebble digestion.
 - The lack of foaming thus far may be attributed to two factors. First, the pebbles digest in a relatively slow (several hours for a single pebble), uniform manner as a function of their surface area. (b)(3), (4)

3.1.3 Risks

(b)(3), (4)

- (b)(3), (4)
- Thermal control of the reaction may limit the ultimate throughput of the process. Control of thermal transients due to surface reactions could become a significant concern in scaling up the reaction.
- Thermal control of the reaction at full scale may not be practical without active cooling.
- (b)(3), (4)
- Development work using irradiated pebbles may identify unforeseen reactions that significantly impact the conceptual process.
- It may be impractical to digest SiC concurrently with graphite.
- It may not be possible to keep the SiC layers intact during graphite digestion of TRISO-containing pebbles. Studies at Jülich indicate that the SiC layers remain intact during digestion, but their tests were limited to the processing of a small amount of unirradiated coated fuel particles.^[8] Digestion of full pebbles of irradiated fuel is likely to include the presence of damaged TRISO particles. However, for the processing of German HTGR fuel, this is not considered an issue as the SiC layer will be breached during later operations. Furthermore, for the German fuel, because the TRISO particles will be mixed with BISO particles, fission products will already contaminate the salt and the positive integrity of TRISO particles will have no considerable benefit.
- Foaming could potentially limit process scale up or throughput rates. However, salt samples collected during full-pebble digestion experiments do not indicate the presence of foaming.

3.2 Salt Regeneration

Salt regeneration is not currently part of the baseline process. Regeneration of the salt reactants, as a process intensification, has the potential to transform the conceptual flow sheet. The recognition of the

potential for regenerating the salt is a recent development. Consequently, the knowledge base for this aspect of the process is small.

It is expected that the salt regeneration system will have the following requirements:

- (b)(3), (4)
- Provide a fine dispersion of gas to facilitate good gas-liquid contact
- Distribute gas throughout the vessel to promote uniform salt concentrations in the vessel
- Consist of adequate materials of construction

3.2.1 State of Knowledge

(b)(3), (4)

(b)(3), (4)

(b)(3), (4) After the salt has been dissolved, and after the fuel and fission products have been removed, the potential exists for recrystallization of the salt from the dissolved salt solution. No evaluations have been conducted relative to the viability of this approach.

3.2.2 Technology Needs

As mentioned above, the potential for regeneration of (b)(3), (4) was only recently recognized. Therefore, there are several technology needs to be addressed:

- (b)(3), (4)
 - (b)(3), (4)
 - (b)(3), (4)
 - The impact of this additional reaction on the materials of construction needs to be determined.
 - *In-situ* reactant regeneration will increase the amount of reaction heat generated and needs to be accounted for in thermal modeling and equipment design.
 - (b)(3), (4)
- . Additional testing is required.

3.2.3 Risks

Risks associated with the state of knowledge of the regeneration of reactants include:

- If the regeneration of reactants is shown not to be feasible, the opportunity to reduce the volume of contaminated salt waste will be significantly diminished.
- The reactant regeneration process could limit the ultimate throughput of the process.
- The complexity of the off gas system may increase if a reactant regeneration process is implemented.
- It may be appropriate to retain carbon (b)(3), (4) rather than send it to the off gas system. In this case, salt regeneration may not be feasible.
- Regeneration will increase the relative off gas flow rate and may increase salt entrainment.

3.3 Fuel Recovery

As described in Section 2.0, there are several options available for the end state of the irradiated fuel. It is assumed that for any of these options, the fuel will need to be recovered from the salt after the graphite and SiC are digested. Several alternatives are being explored for final disposition of the fuel. These include aqueous dissolution, molten salt reduction to metal, and crush/blend of powders.

3.3.1 State of Knowledge

The behavior of exposed fuel kernels (b)(3), (4) has been studied.^[5] Uranium-thorium oxide kernels, which represent the vast majority of the German HTGR fuel, do not dissolve in the molten salt. However, once the layers of graphite have been removed from the kernel, the kernel is sufficiently fragile that some chipping or fracturing may occur during the graphite digestion process. A number of the uranium only kernels (UO₂, UC₂, and UCO) will digest and be reduced to powder.^[8] Uranium only kernels comprise ~76,000 of the AVR pebbles, of which only 4000 are BISO coated and the rest are TRISO coated.

Early concepts for fuel recovery involved using a basket to retain the fuel particles (b)(3), (4). However, the use of a basket was found to be impractical due to several factors.^[10] Basket design was complicated by the variety of particle sizes resulting from the chipped and fractured kernels, and the very small particle size of those kernels that are reduced to powder. Issues also occurred with performance of the basket materials of construction (b)(3), (4).

Potential options for recovering the fuel from (b)(3), (4) include 1) pumping (b)(3), (4) through a filter, 2) using an electrochemical means to separate the fuel from (b)(3), (4), 3) mechanically separating the fuel from the cooled, solidified salt, and 4) dissolving the solidified salt with an aqueous solution and then filtering the fuel from the resulting liquid. If the aqueous solution has a pH greater than 9, the uranium powder from the uranium-only kernels is likely to be insoluble and filterable. Filtration of the fuel material has the potential to capture the fuel in a form suitable for direct charging to a dissolution vessel for U/Th recovery.

If salt regeneration (discussed in Section 3.2) shows promise, it may justify a scenario where the fuel is filtered from (b)(3), (4) salt is then returned to the reaction vessel. The reduction or elimination (b)(3), (4) through the regeneration process could provide a reduction in viscosity and (b)(3), (4) that would make filtering of the melt more attractive.

3.3.2 Technology Needs

There are several technology needs associated with fuel recovery from (b)(3), (4) including:

- The optimum form of the irradiated fuel after graphite and SiC digestion has not yet been defined, since the end state of the irradiated fuel is not yet known.
- The type of full scale process to be employed (e.g., batch or continuous processing) has not yet been determined. This will largely influence the method of fuel recovery.

- No tests have yet been performed with irradiated fuel pebbles. Thus, the behavior of irradiated kernels (b)(3), (4) has not been determined.
- Erosion of the fuel particles during the graphite digestion has not yet been well characterized and may complicate fuel recovery.
- Material control and accountability (MC&A) concerns have not yet been evaluated, and will influence the method of fuel recovery.
 - A method for measuring nuclear material hold up in the salt has not been identified.
 - Criticality control after graphite digestion has not yet been addressed.

3.3.3 Risks

Risks associated with the state of knowledge of fuel recovery include:

- A change in the desired end state of the irradiated fuel could significantly impact the conceptual flow sheet.
- Equipment design to address MC&A concerns could limit the ultimate throughput of the process.
- Filtering of (b)(3), (4) could prove to be impractical. If dissolution of the salt is required prior to fuel recovery, a larger volume of waste will be generated.
- The generation of fine particulates due to erosion of the fuel kernels could complicate a filtering process.
- Irradiated fuel could react differently than expected with (b)(3), (4), resulting in significant changes to the conceptual flow sheet.

3.4 Off-Gas Generation and Handling

An understanding of the generation of off gas species and their appropriate handling are important aspects of (b)(3), (4) digestion concept. As described below, preliminary testing has identified several of these species, originating both from the molten salt and the fuel pebbles.

3.4.1 State of Knowledge

Graphite digestion studies have consistently observed the presence of fine salt particulate in the off gas.^[5,10] The salt particles are small enough to be readily suspended in air. Some evaporation of the salt likely occurs at the reaction temperature. Graphite particulates are also likely to be entrained in the off gas. Some fraction of the salt has been shown to deposit in locations where there is low linear flow or a change of flow direction. The deposition can plug process lines. If the deposition occurs close enough to the heating source, the salt will subsequently melt and form a hardened plug in the process lines. The salt concentration has been confirmed as a mixture of NaNO₃, NaNO₂, and Na₂CO₃, suggesting (b)(3), (4) (b)(3), (4). A preliminary, laboratory scale scrubber design was successful in removing entrained salts from the off gas based on visual observations.^[10]

It is believed that the salt particulate originates principally from reaction transients in the salt melt. It is speculated that some graphite fines accumulate at the surface of the melt without reacting immediately.^[10] The accumulated fines occasionally react in a short time frame causing an accelerated release of gas as well as entrainment of salt and graphite fines. The graphite fines have been detected as part of the salt in the off gas. The off gas system will have to be designed to handle these transient flows, or the digestion reactions will have to be controlled in order to avoid them. Surface transients require the off gas system to have a higher throughput capacity than needed for steady-state operations. The higher throughput could have negative impacts on salt volatility.

Partitioning of radioactive material to the off gas stream needs to be well understood. Testing (b)(3), (4) with irradiated BISO fuel kernels showed that the only significant, non-gaseous fission products in the off

gas were Cs-135/137 (7.5% of initial), Sr-90 (0.03% of initial), and potentially Tc-99. It is unclear due to measurement uncertainty whether any Tc-99 was present. All three radioisotopes leach appreciably from the fuel kernels into the salt. Europium-154/155 and Am-241 also leach appreciably into (b)(3), (4), but were not entrained in the off gas at measureable levels. Other fission products also leach into the salt melt, but they were not detected in the off gas.^[11]

(b)(3), (4)

(b)(3), (4) SRS air permitting may allow for this material to be released to the atmosphere. Alternatively, a wet-scrubber off gas system employing NaOH as the scrubber solution (b)(3), (4), which would be disposed of as waste. Iodine gas containing I-129 will also be released into the off gas system from the reaction and is expected to pass through the scrubber system. If necessary, iodine gas could be absorbed as a separate step in the off gas treatment. A small fraction of I-129 may be present as CsI; if the CsI evaporates, it is expected to deposit in the off gas system along with the other particulate salt.

At the current state of knowledge, it is expected that the design of the OGE system will need to incorporate the following items:

- Designed to handle volatile and fine particulate salts
- Avoids salt accumulation at the exit point from the digestion vessel
- Designed to handle entrained graphite fines
- Captures volatile fission products such as Cs, Sr, Tc, and alkali-iodide salt
- (b)(3), (4)
- System capacity must be able to handle increased gas flow from reaction transients

3.4.2 Technology Needs

There are several technology needs associated with off gas generation and handling, including:

- A complete off gas system has not yet been designed for the conceptual process. However, SRNL will draw on the knowledge available from other SRS processes with off gas systems that are able to successfully operate with high temperatures, entrained particulates, and radioactive species.
- The requirements for off gas handling, retention of volatile fission products, and control of salt volatility have not yet been explored or defined.
- The expected inventory, and associated release, of gaseous species during the digestion reactions needs to be developed so that it can be compared with air permitting allowances and used in design of the off gas system.
- The potential for flammable gas generation and the required controls have not yet been investigated.
- No off gas testing has been conducted with full pebbles containing irradiated fuel. The generation of species that are not currently handled by SRS systems or authorized for release could impact the feasibility of the process.
- The type of digestion system developed (batch system, multiple batch systems, continuous system, etc.) will impact the design of the off gas system.
- Engineering solutions to reduce salt entrainment and evaporation (reaction vessel design, reduction of exposed surface area, etc.) may be available.

3.4.3 Risks

Risks associated with the state of knowledge of off gas generation and handling include:

- Success of (b)(3), (4) process is dependent upon the development and operation of a viable off gas treatment system since it relies on gas generation for the digestion of graphite and SiC.

- Processing complexity will increase and solid waste volume will increase if carbon cannot be released to the atmosphere.
- Some of the off gas species may require capture and disposition using a separate waste form.
- Changing environmental regulations may impact the design and operation of the off gas system.
- Generation rates for certain off gas species could limit production rates (e.g., flammable gas generation).
- No testing has been conducted with full pebbles containing irradiated fuel.
- Dependence upon an aqueous method for the off gas treatment could pose a safety risk.^[12] The potential for large quantities of water to be injected directly into the molten salt presents a mechanism for rapid gas generation and vessel pressurization.

3.5 Waste Generation and Disposition

Processing of the irradiated HTGR fuel pebbles will generate waste streams that will need to be treated for disposal.

3.5.1 *State of Knowledge*

Digestion of the irradiated pebbles will result in leaching of actinides and fission products into the molten salt. Testing at 700 °C with irradiated BISO fuel kernels showed that Cs-135/137, Sr-90, Tc-99, Eu-154/155, and Am-241 leach appreciably from the fuel kernels into the molten salt. Additional fission products also leach [REDACTED] (b)(3), (4) [11]. The salts will therefore have to be treated for disposal after use. The salts are water soluble and have been dissolved as part of recent studies.^[5,10] The fundamental chemistry for waste disposal is expected to be based on processes developed or in use at SRS. Removal of Cs, Sr, and actinides from contaminated salt streams is a well demonstrated technology.^[13]

Other stages of the conceptual process, particularly off gas treatment, will generate additional waste streams for treatment and disposal. Further development of these stages is required before the waste streams can be defined.

3.5.2 *Technology Needs*

There are several technology needs associated with waste generation and disposition, including:

- Treatment of the contaminated salts resulting from digestion of irradiated fuel pebbles has not been demonstrated.
- Disposal paths for decontaminated salt solutions and spent ion exchange resins have not been identified.
- Waste streams resulting from other process stages (e.g., off gas treatment), and their appropriate treatment and disposition, have not been defined since the conceptual process is in early stages of development.

3.5.3 *Risks*

Risks associated with the state of knowledge of waste generation and disposition include:

- The availability of current waste treatment systems at SRS at the time when the HTGR fuel will be processed is not known. The inability to leverage these facilities could add considerable complexity to the process, or require innovative changes to waste disposition.
- Waste inventory and treatment may become process limiting factors.

4.0 Equipment Design Parameters

Knowledge of equipment design parameters for the baseline process is limited given the developmental state of the graphite and SiC digestion concept. The development work completed to date, along with the baseline process concept, is used to develop the scope of an equipment design program in this section.

4.1 General Design Considerations

4.1.1 *State of Knowledge*

Studies to date have identified the necessity for a system that can react and control the temperature of the reactions appropriately.^[5,10] The temperature profile may vary as a function of time during the digestion process. Direct control of the melt temperature is desired as previous testing with indirect control of (b)(3), (4) was not optimal for responding to transient reaction behavior.^[10] Control of the reaction is improved when the pebbles are physically restrained from (b)(3), (4).^[10] A mechanism for controlling the distribution of the pebbles (b)(3), (4) will likely be needed to control reaction rates and temperatures. Therefore, a basket for containing the pebbles is a component of the baseline design. The process will occur in a remotely handled environment due to radiological concerns. Equipment design will have to be specifically tailored for operation and maintenance in a remote environment.

Addressing the technology needs associated with the other aspects of the program discussed earlier will provide further insight into equipment design parameters. For example, digestion of SiC may be effective in parallel with digestion of graphite, or may be more appropriate as a separate process stage. Regeneration of (b)(3), (4) may be possible in parallel with the digestion reactions, may be more appropriate as a separate process stage, or may be infeasible. Further progress in these areas will allow for better definition of equipment design parameters.

4.1.2 *Technology Needs*

There are several technology needs associated with equipment design parameters, including:

- The impacts of the introduction of new equipment and new modes of processing on safety basis documentation for SRS facilities have not yet been determined. It is not yet known whether the necessary approvals can be obtained in the present regulatory climate.
- Throughput needs are assumed to be 1000 pebbles per day, although the practicality of this rate has not yet been determined.
- It is not known whether a continuous process will be viable for the graphite and SiC digestion, or whether a batch process or batch processes will be more appropriate.
 - Materials handling methods can be defined once the process model is better understood.
- Thermal modeling and energy balances will determine heating and cooling needs of the process equipment.
- Materials compatibility issues can be addressed once the complete reaction scheme and thermal profile are developed.
- MC&A concerns will influence the design parameters (e.g., vessel volumes, etc.), and will need to be incorporated at an early stage.
- Mixing needs have not yet been identified, particularly since testing to date has been only up to the single pebble scale.
 - Thermal gradients and gas generation are likely to contribute to mixing of the system and will impact reaction rates. It is not known whether additional agitation will be needed (b)(3), (4)

(b)(3), (4)

4.1.3 Risks

Risks associated with the state of knowledge of equipment design include:


- The ultimate process throughput rate may not meet current expectations.
- It will be difficult or impossible for equipment design to proceed in parallel with development of the process until several of the process parameters are better defined.
- The currently proposed scale-up maturation tasks all involve hands-on activities. The final design will require that the process be performed in a remotely handled facility.

4.2 Graphite Digestion Vessel and Basket

4.2.1 State of Knowledge

A nominal process scale of 60 pebbles (~12 kg of graphite) has been identified as sufficient to evaluate the interactions among unit operations of the baseline process design. Based on the current expectations, processing 60 pebbles concurrently would be one-fifth to one-tenth of the full scale for operations without salt regeneration and approximately one-half scale with salt regeneration.

(b)(3), (4)



However, single-pebble digestion may not accurately predict the behavior of multiple-pebble digestion.

Batch feeding of the pebbles in a basket which completely immerses the pebbles (b)(3), (4) is currently the baseline approach based on testing completed in the early stages of the SRNL studies. The volume of salt is sufficiently large to make it possible to design a retractable basket that distributes the pebbles uniformly throughout the salt such that the interaction between pebbles is minimized. This will allow for more uniform reaction of each pebble and should lead to better temperature control, uniform mixing, and shorter process cycles. The basket will not be designed to retain fuel kernels. The digestion behavior of the fuel kernels and the stringent material compatibility issues associated with a fine-mesh screen for the basket currently preclude the design of a basket that is able to retain the undigested fuel while permitting salt migration through the basket wall.^[10]

The vessel design should include insulation, as necessary to provide the appropriate balance between heating efficiency and cooling efficiency. The vessel will be equipped with the ability to add NaOH (presumably as a solid) near the end of the cycle to digest SiC coatings associated with TRISO kernels.

4.2.2 Technology Needs

There are several technology needs associated with graphite digestion vessel and basket design, including:

- No testing has been conducted with full pebbles containing TRISO fuel kernels.
- The baseline heating method (as described in the following section) could change, which may impact the vessel design.
- MC&A and criticality reviews have not yet been incorporated into the equipment design.
- It is known that the salt rheology changes as a function of temperature and composition, but those changes have not been quantified and could have a significant effect on vessel design.

- (b)(3), (4)
- It is not known whether the design should include a mechanism to prevent temperature and OGE transients.
- Materials of construction have not been thoroughly evaluated, although stainless steel and Alloy 600 are considered to be viable candidates. The success of the salt regeneration process may further impact materials selection.
- The baseline design has not yet been thoroughly evaluated for use in a remotely handled environment.

4.2.3 Risks

Risks associated with the state of knowledge of graphite digestion vessel and basket design include:

- Final digestion of TRISO kernels could be more difficult than expected and require changes to the digestion vessel requirement.
- Demonstration of salt regeneration would likely affect various aspects of the vessel and basket designs.
- Demonstration of salt regeneration could require more robust materials of construction.
- The final design of the process vessel and basket could differ significantly from the baseline.

4.3 Digestion Vessel Heating Method

4.3.1 State of Knowledge

There are three principal ways to control the graphite digestion reaction (i.e., to control the heat generated by the reaction): control of graphite surface area, control of temperature, and control of oxidant. Control of graphite surface area means that the salt melt is maintained at a high enough temperature so that when a controlled amount of graphite is added (b)(3), (4)

(b)(3), (4) Control of temperature means that a large amount of graphite surface area is charged (b)(3), (4), and the overall digestion rate is controlled by regulating (b)(3), (4). As the reaction progresses and the graphite surface area decreases, the temperature can be increased to sustain the overall digestion rate. (b)(3), (4) means that a large volume of graphite is maintained at (b)(3), (4) reactant in the system.

It was found that the first two methods – control of surface area and temperature – yielded essentially the same total cycle time.^[5] While it may be possible to run the reaction at a constant temperature and surface area, it is likely that such an approach will only be viable if salt recycle can be developed because (b)(3), (4). The third method – (b)(3), (4) – was attempted and was unsuccessful. Therefore, direct control of the melt temperature is the baseline control method.

The reaction has been measured as a function of temperature and appears to increase linearly as a function of temperature. (b)(3), (4)

(b)(3), (4) In the baseline approach, digestion starts at the lower reaction temperature when the graphite surface area is highest and the temperature is increased over time to accelerate the digestion rate as the graphite surface area diminishes. (b)(3), (4)

The graphite digestion reaction is exothermic. To achieve the digestion rates required for a full-scale process, it is expected that the system must be capable of free-radiating enough heat to the atmosphere to

be passively cooled. To date, laboratory scale salt melts have been contained within insulated furnaces: a muffle furnace for small-scale testing and a tube furnace for full-pebble testing. Such designs do not offer adequate means for removal of the heat of reaction, which is required for elevated reaction rates at controlled temperatures. The heat loss from those tests was limited (b)(3), (4)

(b)(3), (4)

SRNL experience considers induction heating to be a viable candidate technology for the baseline design. Since induction heating will heat the side walls of the digestion vessel, which will then conduct heat into the salt, the heat should be distributed as evenly as possible along the walls of the vessel to prevent localized surface hot spots where salt would decompose more rapidly. Temperature uniformity (b)(3), (4) is not viewed as a critical requirement. Gases generated by the graphite digestion reaction should promote mixing and temperature uniformity. Heating equipment must be capable of allowing salt to be adequately removed from the digestion vessel. It is not known whether the salt will be removed *in situ* or at an alternate process station. The ability to regenerate the salt *in situ* may allow for the use of alternative heating technologies.

4.3.2 Technology Needs

There are several technology needs associated with the graphite digestion vessel heating method, including:

- Successful demonstration of salt regeneration could significantly change the vessel heating requirements.
- (b)(3), (4)
- Safety requirements associated with large quantities of high temperature molten salts have not yet been thoroughly reviewed.

4.3.3 Risks

Risks associated with the state of knowledge of the graphite digestion vessel heating method include:

- Spot heating could cause excessive salt decomposition.
- Temperature control could limit the ultimate scale of the digestion vessel or dictate the practical process throughput rate.
- Near-term schedule constraints may delay demonstration of induction heating for the graphite digestion process.

5.0 Scale-Up Maturation Plan for Molten Salt Graphite Digestion Technology

This section will present a maturation plan for the (b)(3), (4) digestion technology based on the state of knowledge and technology needs described previously. Near term tasks are listed in order of priority below. A description of each task is provided to demonstrate how the results of the activity will address technology needs.

5.1 Near Term Scale-Up Maturation Tasks

1. Determine the expected inventory of gaseous reaction products and fission products
The current graphite and SiC digestion concept is dependent on the release of species, (b)(3), (4) containing C-14, to the atmosphere. An inventory of the expected carbon and other gaseous reaction products, including fission products, will be developed. This inventory will then be compared with SRS air permitting allowances to determine whether the current graphite digestion concept is viable, or whether additional capture of the gaseous reaction products will be required. Further development of the graphite digestion concept is dependent on the outcome of this study because the disposition of C-14 through the SRS liquid waste system is unlikely.
2. Determine the viability of salt regeneration
The ability to regenerate (b)(3), (4) used in the graphite digestion process has the potential to significantly reduce the amount of waste generated, reduce the size of the reaction vessel, positively impact material handling, reduce the number of heating-cooling cycles, and enable continuous processing. Salt regeneration has not yet been studied, but should be explored immediately due to its potential benefits. Studies will include potential reactions with NaOH, fuel kernels, and fission products, and will determine whether salt regeneration can occur in parallel with graphite digestion.
3. Develop conceptual off gas system
A conceptual off gas treatment system will be developed through the adaptation of demonstrated SRS technologies. The conceptual off gas system will support further development of process equipment, and will define the additional waste streams resulting from off gas treatment.
4. Develop a bench scale, integrated demonstration platform
Demonstration of a small scale, integrated processing system is necessary for identifying interactive effects among the multiple conceptual process reactions and providing data for scale up and process equipment design. This integrated system will combine the ability to digest graphite from multiple unirradiated pebbles simultaneously, digest SiC, monitor and control reaction temperatures, and characterize off gas species (and potentially demonstrate their capture and treatment). This system should also be equipped to demonstrate salt regeneration, assuming salt regeneration is feasible.
5. Demonstrate methods of fuel recovery from the molten salt
Recovery of fuel kernels (b)(3), (4) has not yet been attempted. Several potential methods for recovering the fuel from the salt were described earlier in this document. A down selection process will be used to identify the most appropriate method, with the incorporation of knowledge gained through the full pebble digestion tests described above. Data gathered in these studies will support development of the overall process flow sheet, and will impact the downstream processing of the fuel kernels. Treatment and disposal of waste salt is also impacted by the fuel recovery method.

6. Demonstrate SiC digestion with unirradiated TRISO particles
Digestion of unirradiated TRISO fuel particles at the conclusion of bulk graphite digestion has not yet been demonstrated. Demonstration of graphite digestion followed by SiC digestion will provide important process information, including necessary NaOH concentrations, reaction times and temperatures, and interactive effects. These data are needed to develop the process flow sheet.
7. Develop targeted salt compositions
If SiC digestion or salt regeneration is feasible in parallel with graphite digestion, the optimized salt composition will be developed. (b)(3), (4)
. These data are needed to develop the process flow sheet.
8. Determine (b)(3), (4) rheology and thermal properties
The rheological and thermal properties of (b)(3), (4) as a function of its temperature and composition, will impact the rate of reaction and heat transfer in the system. Some of these data may be available in the literature, while other properties may need to be determined experimentally. These data are needed to support thermal modeling and later equipment design.
9. Develop thermal profile for reactions
Once data on SiC digestion and salt regeneration are available, the complete thermal profile for the reactions will be developed.
10. Complete a thermal model for the process reactions
A model is needed that describes the heat consumed and generated by the various stages of the reactions, including digestion and salt regeneration. This model will be critical in scale up of the digestion process and process equipment design, particularly for defining heating and cooling requirements. The thermal model will also support later energy balance calculations and process intensification.
11. Develop mass and energy balances
Complete mass and energy balances for the overall process are needed to support equipment design, waste generation and treatment planning, and process intensification.

5.2 Longer Term Scale-Up Maturation Tasks

Over the longer term, maturation of waste treatment and disposal technology (Section 3.5), and process equipment design (Section 4.0) will occur. It is premature to address these tasks in the near term given the conceptual state of the fuel processing program. Integration with other aspects of the HTGR fuel processing program will need to be addressed as well.

This scale-up maturation plan will be reevaluated after the completion of the near term tasks, and more frequently if necessitated by the outcomes of those tasks. The direction and prioritization of the remaining tasks will be reevaluated based on newly developed knowledge. Specific tasks that should be considered for longer term technology maturation include:

1. Operation of an integrated pilot-scale system
Decisions concerning a full-scale design should not be made until an integrated pilot-scale system has been constructed and operated. The purpose of the integrated pilot-scale system is to demonstrate that all subsystems work together and can be designed to operate in a remotely-

handled environment. Testing in fiscal year 2014 will advance the understanding of the various subsystems, but there will not be sufficient time to evaluate them as an integrated unit.

2. Demonstrate digestion of a full, irradiated fuel pebble
Digestion of a complete, irradiated fuel pebble has not yet been demonstrated. This demonstration will be used to further characterize off gas generation and composition, fuel and fission product leaching into (b)(3), (4) and the condition of the fuel kernels at the completion of the digestion reactions. These data will support further development of the process flow sheet, the off gas system, the fuel recovery method, and treatment of the contaminated salt.

It is noted, however, that the collection, transport, and receipt of an irradiated pebble is likely to be problematic. Therefore, this task is considered to be longer term both because of the challenges associated with obtaining the irradiated pebble, and more significantly, because the near term tasks described above are likely to provide much of the data needed to support maturation of the technology. The additional knowledge gained by digesting an irradiated pebble is likely to be minimal given that other aspects of this plan will provide considerable data on the digestion of unirradiated pebbles and irradiated kernels. When a review of data from recent fuel kernel tests^[1] is complete and compared with what can be discerned from the literature, the magnitude of risk associated with deferring this task to the longer term can be adequately assessed.

3. Complete a nuclear materials accountability assessment
The conceptual graphite digestion process needs to be evaluated from a nuclear materials control perspective, in terms of maintaining an accurate inventory of material, maintaining the security of the material, and minimizing or eliminating criticality concerns. Integration of this assessment with the scale-up maturation process is important as these concerns could have a strong influence on the ultimate design of the process.
4. Demonstrate decontamination of used salt
The salt remaining after fuel recovery will likely need to be decontaminated prior to disposal. This task will demonstrate the appropriateness of the spent molten salt for removing Cs, Sr, and actinides from contaminated salt solutions. The data developed will be used to support waste treatment and disposal planning.

5.3 Prioritization of Scale-Up Maturation Tasks for Fiscal Year 2014

The following tasks, as described in detail in Section 5.1, will be addressed by SRNL in fiscal year 2014. Several of these tasks may be approached concurrently. Task technical and quality assurance plans will be issued for individual or combined tasks, as appropriate.

1. Determine the expected inventory of gaseous reaction products and fission products
2. Determine the viability of salt regeneration
3. Develop conceptual off gas system
4. Develop a bench scale, integrated demonstration platform
5. Demonstrate methods of fuel recovery from the molten salt
6. Demonstrate SiC digestion in "spent salt" using unirradiated fuel kernels
7. Develop targeted salt compositions at completion of graphite digestion
8. Determine molten salt rheology and thermal properties
9. Develop a thermal profile for the overall graphite digestion process cycle
10. Complete a thermal model for the overall graphite digestion process cycle

11. Develop mass and energy balances

5.4 Quality Assurance

Studies performed in support of this scale-up maturation plan will be controlled via a Task Technical and Quality Assurance Plan (TTQAP). The TTQAP will include a procedure matrix identifying the applicable sections of the Savannah River Site Quality Assurance Manual (1Q) and associated implementing procedures for the SRNL E&CPT Research Programs Section. Work performed in other SRNL organizations, such as Analytical Development, will be controlled via those organizations' procedures and quality assurance programs. The TTQAP will be reviewed and approved by SRNL Quality Assurance personnel. Additional study plans may be issued, as appropriate, to provide further details of experiments. These study plans will be controlled via the TTQAP. Technical documents produced as a result of these studies will undergo a review prior to issuance. Requirements for performing reviews of technical reports and the extent of review are established in manual E7 2.60. SRNL documents the extent and type of review using the SRNL Technical Report Design Checklist contained in WSRC-IM-2002-00011, Rev. 2.

6.0 Summary and Path Forward

This scale-up maturation plan describes a multi-year program addressing technical considerations related to the processing of HTGR fuel. The objectives of this plan are to provide technical justification for the selected approach to HTGR fuel processing, review the progress of development efforts to date, identify technology needs and risks, and prioritize a plan for addressing those technology needs and mitigating risks. The focus of this plan is specifically on the removal of graphite and SiC from the fuel pebbles and kernels. Integration with other aspects of the HTGR fuel processing program is considered, with recognition that those other aspects are under concurrent development.

A review of the status of knowledge, technology needs, and risks associated with several aspects of the (b)(3), (4) digestion concept is provided. This review is used to generate a prioritized list of near term tasks that will address technology needs and reduce risks. Specific tasks to be addressed by SRNL in fiscal year 2014 are identified. Detailed planning for each task will be addressed via task technical and quality assurance plans, as appropriate.

It is premature to address some of the identified technology needs and risks at this point in the development process, given the conceptual state of the fuel processing program. Therefore, this scale-up maturation plan will be reevaluated after the completion of the near term tasks, and more frequently if necessitated by the outcomes of those tasks. The direction and prioritization of the remaining tasks will be reevaluated based on newly developed knowledge, as well as developments in other aspects of the overall fuel processing program. A multi-year commitment of resources and funding is needed for complete maturation of this technology.

7.0 References

1. "Oak Ridge National Laboratory Status and Progress Report", ORNL-3998, July 1966.
2. L. M. Ferris, "Grind-Leach Process for Graphite-Base Reactor Fuels that Contain Coated Particles: Laboratory Development", ORNL-4110, June 1967.
3. Letts, A. L., W. D. Bond, C. W. Forsberg, R. W. Glass, F. E. Harrington, G. E. Michaels, K. J. Notz, and R. G. Wymer, "Options for Treating High-Temperature Gas-Cooled Reactor Fuel for Repository Disposal," *U.S. Department of Energy Report ORNL/TM-12027 UC-522, -810, -811*, Oak Ridge National Laboratory, Oak Ridge, TN (1992).
4. Del Cul, G. D., B. B. Spencer, C. W. Forsberg, E. D. Collins, and W. S. Rickman, "Triso-Coated Fuel Processing to Support High-Temperature Gas-Cooled Reactors," *U.S. Department of Energy Report ORNL/TM-2002/156*, Oak Ridge National Laboratory, Oak Ridge, TN (2002).
5. R. A. Pierce, T. C. Shehee, D. J. Pak, and K. J. Imrich, "Digestion of Graphite-Based Materials", SRNL-STI-2013-00392, July 2013.
6. R. A. Pierce and D. C. Castiglione, Radioactive Studies of Sodium Peroxide Fusion for Accelerated Dissolution of 3013 DE Materials, SRNL-STI-2010-00425, July 2010.
7. (b)(3), (4)
8. A. Wilden, F. Sadowski, M. Klinkenberg, C. Schreinemachers, J. Holthausen, J. Dellen, E. Müller, G. Modolo, H. Schlenz, H. Curtius, D. Bosbach, G. Mank, and G. Damm, "HTR-WG Summary Report FZL-IEK-6", Forschungszentrum Jülich GmbH, August 2013.
9. (b)(3), (4)
10. R. A. Pierce, D. J. Pak, and K. J. Kalbaugh, "Digestion of Graphite-Based Fuel Materials: HTGR Pebble Digestion", SRNL-STI-2013-00598, September 2013.
11. R. A. Pierce, C. J. Coleman, D. P. Diprete, and D. J. Pak, "Digestion of Irradiated HTGR Fuel Kernels", SRNL-STI-2014-00266, June 2014.
12. (b)(3), (4)
13. D. T. Hobbs, T. B. Peters, M. R. Poirier, F. F. Fondeur, C. A. Nash, and S. D. Fink, "Separation of Fission Products and Actinides from Savannah River Site High Level Nuclear Wastes," Proceedings of the 12th International Conference on Environmental Remediation and Radioactive Waste Management, Liverpool, United Kingdom, October 11-15, 2009, pages 601-610, doi:10.1115/ICEM2009-16174

Distribution:

T. M. Adams, 773-41A
P. M. Almond, 773-A
W. F. Bates, 703-H
N. J. Bridges, 773-A
T. B. Brown, 773-A
T. B. Calloway, 999-2W
M. A. Creed, 703-42A
A. O. Delley, 707-24B
W. G. Dyer, 704-2H
S. D. Fink, 773-A
K. M. Fox, 999-W
K. S. Fuller, 703-H
E. K. Hansen, 999-W
C. C. Herman, 773-A
D. T. Herman, 735-11A
D. T. Hobbs, 773-A
E. N. Hoffman, 999-W
R. H. Jones, 707-24B
J. E. Laurinat, 703-41A
S. L. Marra, 773-A

J. C. Marra, 999-2W
G. M. Maxted, 703-H
D. H. McGuire, 999-W
E. N. Moore, 707-24B
J. R. Murphy, 703-H
F. M. Pennebaker, 773-42A
R. A. Pierce, 773-A
M. L. Restivo, 773-42A
T. S. Rudisill, 773-A
E. T. Sadowski, 773-A
T. F. Severynse, 707-24B
T. C. Shehee, 773-A
R. M. Sprague, 773-A
W. D. Stephens, 707-58B
K. M. Taylor-Pashow, 773-A
D. E. Welliver, 704-2H
B. M. Williamson, 704-2H
W. R. Wilmarth, 773-A
J. R. Zamecnik, 999-W
Records Administration (EDWS)