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A Tritium Research and Collaboration Plan for the NGNP Project

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ABSTRACT

Contamination of downstream hydrogen production plants or other users of high-temperature heat is a concern of the Next Generation Nuclear Plant (NGNP) Project. Due to the high operating temperatures of the NGNP (850-900ºC outlet temperature), tritium produced in the nuclear reactor can permeate through heat exchangers to reach the hydrogen production plant, where it can become incorporated into process chemicals or the hydrogen product. This document provides a plan and funding needs estimates for research and development that is directed towards reducing or mitigating the potential for downstream tritium contamination. Collaboration is sought between the INL, SRNL, and AECL that is directed towards reducing or eliminating the potential for tritium contamination of non-nuclear processes that are downstream of the nuclear reactor in the NGNP. Work will be pursued in the areas of permeability and barrier materials, high-temperature getters and separation systems, system modeling, instrumentation, waste handling, and disposal. With sufficient funding, the work plan can be completed within two to three years, which is sufficient to support NGNP permitting and licensing efforts.

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1. INTRODUCTION

1.1 Next Generation Nuclear Plant (NGNP) and Tritium

The Next Generation Nuclear Plant (NGNP) is envisioned as consisting of a high-temperature gas-cooled nuclear reactor (HTGR), gas-driven or steam-driven electrical generators, a high-temperature heat transfer loop, and thermal energy delivery systems capable of providing high temperature heat to a hydrogen production plant or other down-stream industrial process (e.g., coal-to-liquids plant, petroleum refinery, underground fossil fuel recovery). A basic schematic of the NGNP featuring a hydrogen production plant is shown in Figure 1. An artist's conception of this plant is shown in Figure 2.

The HTGR, or more specifically a VHTR (Very High Temperature Gas-Cooled Reactor), is a helium-cooled, graphite moderated, thermal neutron spectrum nuclear reactor that will operate at a nominal pressure between 5-9 MPa and an outlet temperature between 800-950°C [1]. The development of the HTGR for this application builds upon earlier gas-cooled nuclear reactor experiences including Dragon, Peach Bottom, Fort St. Vrain, PNP, and others [2, 3]. The reactor will employ tri-isotopic (TRISO)-coated uranium fuel and can be constructed using either a prismatic or pebble bed core. The reactor will contain passive safety features, and would be capable of withstanding a postulated long-term, depressurized, loss-of-forced-convection accident without damaging the fuel. The VHTR is one of six reactor concepts recommended by the Generation IV Technology Roadmap for further development [4] and the NGNP is the top priority in the U.S. Generation IV Implementation Strategy [5].

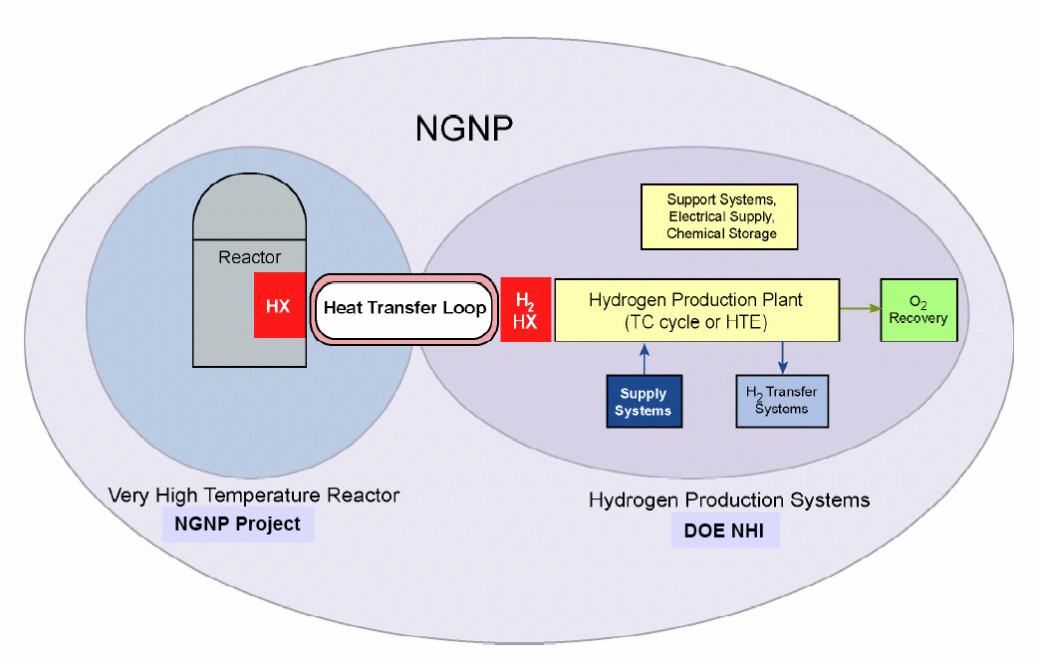

Figure 1. Schematic of Next Generation Nuclear Plant (NGNP) with hydrogen production plant.

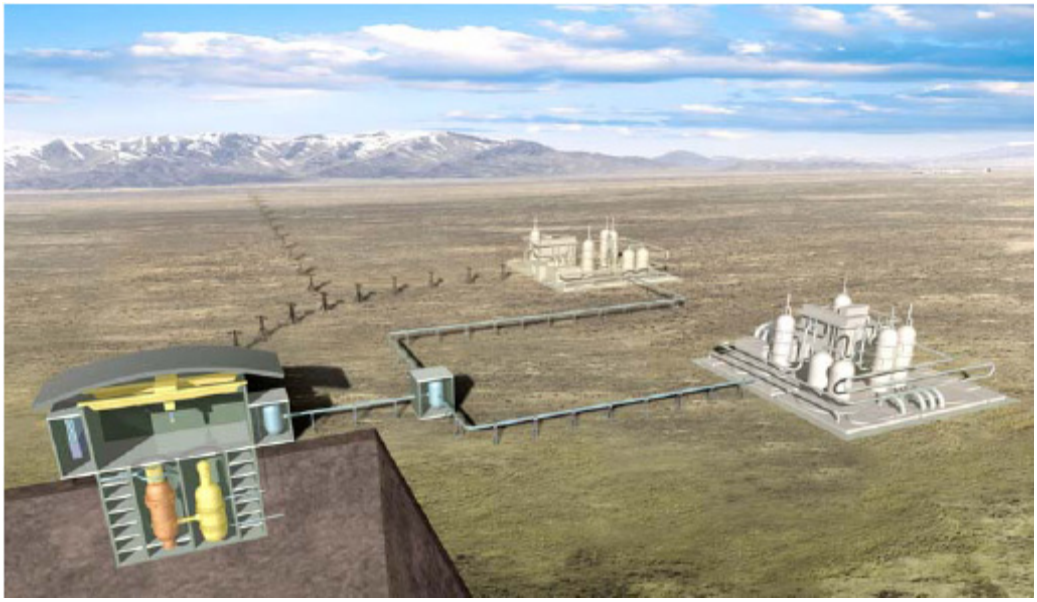

Figure 2. Artist's conception of the NGNP (left) featuring a hydrogen production plant (top) and a chemical plant (right).

In 2005, the U.S. Congress passed the Energy Policy Act (EPACT 2005 [6]) which authorized the creation of the NGNP Project and provided performance targets. In Section 641, the Act states that the Project shall consist of research, development, design, construction, and operation activities for a prototype plant that includes a nuclear reactor supported by the Generation IV Nuclear Energy Systems Initiative, that produces electricity or hydrogen or both, and that start-up of the prototype will occur no later than the end of FY 2021 (September 2021). The Act calls for placement of the NGNP at the Idaho National Laboratory (INL). Major program elements described in the Act include

- High-temperature hydrogen production technology development and validation
- Power conversion technology development and validation
- Nuclear fuel development, characterization and qualification
- Materials selection, development, testing and qualification
- Reactor and balance-of-plant (BOP) design, engineering, safety analysis, and qualification.

As a result of this Act, the U.S. Department of Energy (DOE) selected the INL to lead the development of the NGNP by integrating and coordinating all necessary research and development activities, and by coordinating project efforts with industry and other Project participants. The nuclear reactor will be licensed by the U.S. Nuclear Regulatory Commission (NRC) under a commercial license.

A distinguishing characteristic of the NGNP from established commercial nuclear plants is the presence of a thermal energy connection between the nuclear plant and a non-nuclear user of high-temperature heat. In the NGNP, thermal energy is delivered from the core of the nuclear reactor to a downstream user without the intermediate conversion of thermal energy into electrical energy, and this is accomplished by heat transfer through heat exchangers and the bulk movement of heat transfer fluids through conduits. While direct heat transfer is more efficient than using an intermediary electricity conversion step, a direct heat transfer pathway provides an

opportunity to transport fission products, particularly tritium, from the nuclear reactor into the downstream plant.

Tritium is a radioactive form of hydrogen. Its molecular weight is 3.016 amu and it has a half-life $t_{1/2}$ of 12.33 years. One gram of tritium is equivalent to approximately 9,600 Ci. Tritium is produced in the core of the reactor as a by-product of the uranium fission reactions, and acts as a contaminant in the nuclear reactor heat transfer streams. Tritium is a weak beta emitter (0 to 18.6 keV), and decays to ^{3}He upon disintegration. The primary route of tritium exposure in living things is through the ingestion or absorption of tritiated water. Apart from a difference in mass, tritium behaves chemically as hydrogen [7]. Tritium, like hydrogen, is highly mobile and can readily permeate into and through materials, especially metals, at higher temperatures, and this permeation is the cause of hydrogen embrittlement under some conditions. Due to the potential health hazard from the ingestion or absorption of tritium, tritium emissions to the environment are regulated by the NRC and the U.S. Environmental Protection Agency (EPA).

Since tritium behaves chemically as hydrogen, it can undergo isotope exchange reactions with hydrogen-containing chemicals (e.g., H_2O, H_2, CH_4, etc.) in downstream industrial processes and become incorporated into the plant's process chemicals or products, and incorporation of tritium in any significant amount may lead to a number of problems. Maintenance of the downstream facilities becomes more complicated because radiation dose to workers becomes a consideration. The downstream facilities may not be licensed or permitted to operate as a nuclear facility, and excessive contamination of the plant by tritium may push it into the nuclear-regulated regime. Also, customers of the downstream facilities may have little tolerance for a radioactive product, and so contamination of downstream facilities with tritium may lead to rejected product and loss of revenue for the plant.

Tritium control systems and technologies will be employed in the NGNP to reduce or eliminate downstream tritium contamination. These systems will consist of tritium permeation barriers, separation systems, and instrumentation capable of measuring tritium at very low concentrations. Development and operation of these systems will be assisted by dynamic system models and defined procedures for the handling and disposal of tritium-contaminated materials.

1.2 NGNP Tritium Workflow

The tritium control systems and technologies that will be used in the NGNP are not off-the-shelf items, and some research and development is needed to extend the capabilities of currently available technologies to operate at the NGNP's higher temperature range and at the very low tritium concentrations that are required to achieve tritium effluent limits. The performance of select tritium control technologies must be understood, quantified, and documented, so that the implications of their use can be understood by the NGNP Project's NRC licensing group, and detailed system designs can be developed by the NGNP vendor teams.

In an effort to organize the research and development effort, the NGNP Project has prepared a workflow diagram for tritium-related work, which is shown in Figure 3. There are three workflow pathways. The first pathway concerns development and validation of dynamic system models (particularly THYTAN [8] or its equivalent). The second pathway concerns the performance of experiments and the creation of conceptual system designs involving high-temperature tritium barrier materials, separation systems, and instrumentation. The third pathway is procedural, and involves the definition or determination of applicable tritium release limits, and

waste handling and disposal options. These pathways are pursued in a parallel fashion and information is exchanged between work teams to avoid duplication of effort and to achieve efficient execution of the work. The pathways converge once the significant research and development items associated with each pathway are addressed, and the data is used to support NGNP Project vendors and the NGNP Project's NRC licensing group. The workflow diagram is preliminary, and further updates to the diagram are anticipated.

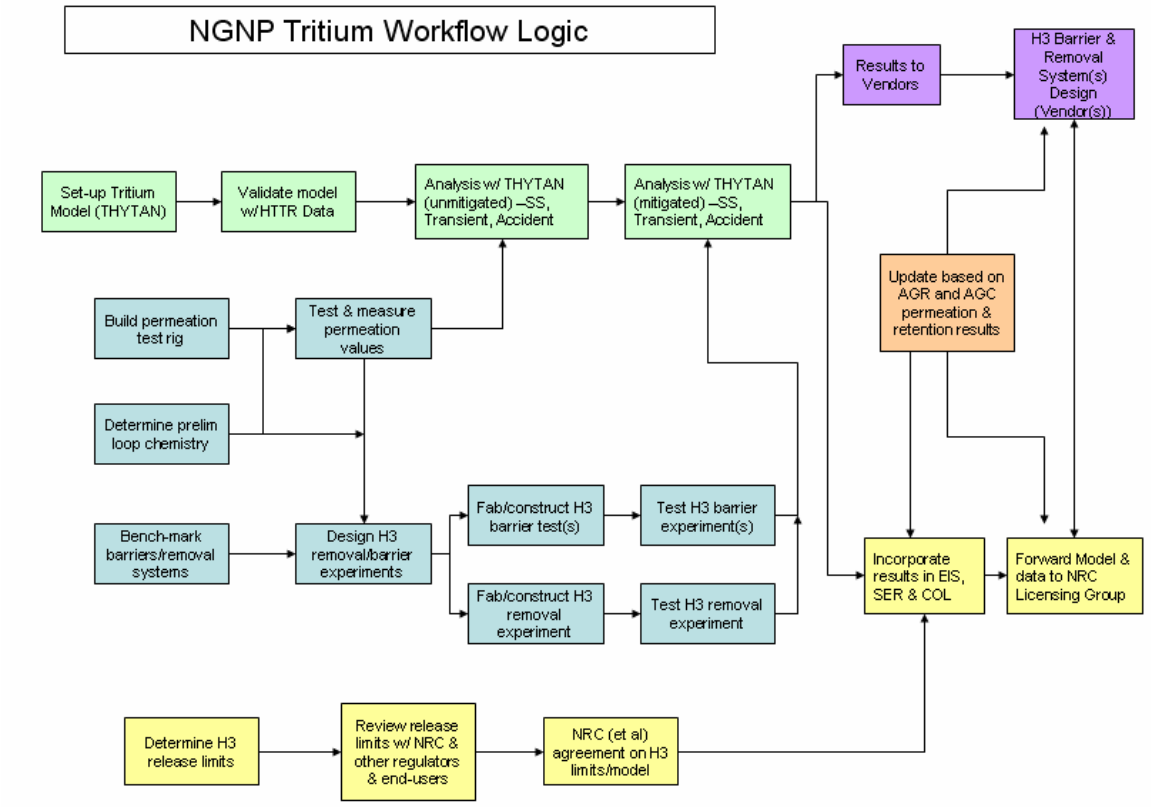

Figure 3. Modeling, experimental, and procedural pathways to support future licensing and vendor activities.

1.3 Overview

This document provides a work plan and funding needs estimates for research and development that is directed towards reducing or mitigating the potential for downstream tritium contamination. Work is recommended in the areas of permeability and barrier materials, high-temperature getters and separation systems, system modeling, instrumentation, waste handling, and disposal. These tasks are aligned with the NGNP Tritium Workflow Logic diagram shown in Figure 3, and involve collaboration between the Idaho National Laboratory (INL), Savannah River National Laboratory (SRNL), and Atomic Energy of Canada Limited (AECL). Detailed work plans, timelines, and funding requirements are provided. With sufficient funding, the work plan can be completed

within two to three years, which is sufficient to support NGNP permitting and licensing efforts. According to the latest NGNP project timelines, the NGNP combined construction and operating license application (COLA) will be submitted to the NRC in the latter half of 2012.

2. RESEARCH AND DEVELOPMENT PLAN

2.1 Summary

Work is proposed in five areas. These areas are:

- permeability measurements
- tritium getters and separation systems
- dynamic system modeling
- on-line instrumentation
- waste handling and disposal.

Preliminary rankings and funding requirements are assigned, and suggested sequences of activities are provided. The list of proposed projects is drawn from the NGNP report "Tritium Barrier Materials and Separation Systems for the NGNP" [9]. The locations where work might be performed are named, and it is recognized that some tasks could be carried out at more than one location if the capabilities are not unique to any one laboratory, or if independent validation of data is required.

Table 1 shows a prioritized project list. Priority 1 projects are ones that are recommended for execution in FY09. Priority 2 projects are ones that should be funded in FY09, but may be delayed in their start until FY10 if the funding is insufficient to support them in FY09.

Table 1. Prioritized List of Tritium Projects

Priority	Description	Year 1 Request ($K)	Year 2 Request ($K)
1	Tritium Permeation Barrier Development for NGNP Heat Exchangers (INL, Sharpe and Ebner)*	500	500
1	*Tritium Permeation Barrier Development for NGNP Heat Exchangers (SRNL, Korinko and Brinkman)*	*300*	*300*
1	Development of Tritium Sequestration System for NGNP (SRNL, Clark, Klein, Brinkman)	600	1,500
1	Dynamic Modeling for Tritium Migration (INL, Oh and Kim)	400	200
2	*Dynamic Modeling for Tritium Migration Using Aspen Custom Modeler (ACM) (SRNL, Gorensek)†*	*350*	*200*
3	Evaluation of Tritiated Species Cracking/Decomposition Agents (SRNL, Korinko, Klein, Fox)	400	200
2	On-line Sensors for Measurement of Dilute Tritium (SRNL, Ferrar, Cordaro and Sigg)	1,200	1,200
2	Development of Tritiated Materials/Components Disposal Strategies (SRNL, Clark)	105	0
	Priority 1 Funding Priority 2 Funding	1,800 2,055	2,500 1,600
	Total	3,755	4,100

*Funding to be provided by NGNP R&D Branch instead of NGNP Engineering Branch.
†Alternative modeling pathway allows direct use of current Aspen process flow sheets.

Requested funding is spread out over two years, and the amounts needed during each year of a task are shown under the heading "Year 1 Request ($K)" or "Year 2 Request ($K)." The first year is used to measure data and test systems in non-tritium environments, and the second year is used to confirm the performance of barriers, getters, separation systems, or instruments in tritium environments. If adequate funding is provided and all of the tasks are pursued in parallel, all of the recommended work could be completed in two years. With less funding, a staggered start to the projects may be required, and some or all of the Priority 2 projects may have to be delayed until FY10, in which case the list of projects could be completed in three years.

The work will be performed in a collaborative manner. The INL and SRNL will be the primary contributors, and AECL will be available to perform work or provide reviews of on-going work, as needed. U.S. DOE has determined that collaboration on tritium technologies between the U.S. and Canada be conducted under the Generation IV International Forum (GIF) framework and managed by the GIF Hydrogen Production Project Management Board. Under this agreement, information is shared among GIF participants but funding is not exchanged. This does not preclude the NGNP Project from writing a direct contract with AECL in order to perform a certain portion of the work, but the NGNP Project is not obligated to do so under the GIF framework.

2.2 Permeability and Barrier Materials

FY09 Priority: 1 and 2
Location: INL, *SRNL*
Funding Needs:

Priority	Description	Year 1 Request ($K)	Year 2 Request ($K)
1	Tritium Permeation Barrier Development for NGNP Heat Exchangers (INL, Sharpe and Ebner)*	500	500
2	*Tritium Permeation Barrier Development for NGNP Heat Exchangers (SRNL, Korinko and Brinkman)*†	*300*	*300*
	Total	800	800

*Task will be funded out of NGNP Project Fission Products Transport area
†Task would be funded out of NGNP System Interface area

Description:

If a downstream process becomes contaminated with tritium, it most likely will have occurred due to the permeation of tritium through the intermediate loop heat exchangers. Therefore, having data on the tritium permeability of heat exchanger materials and of candidate barrier materials is needed in order to develop more accurate models, choose the proper heat exchanger barrier material(s), and to aid in the design of tritium separation/sequestration systems.

Tritium barrier materials for heat exchanger surfaces have been studied extensively, but most of the work has been performed at temperatures lower than the operating temperatures of the NGNP. Some work has been done at higher temperatures in connection with the German gas-cooled reactor programs in the 1970's and 1980's, but that work is incomplete. The available data must be extended to more fully cover the temperature range 600-950ºC, so that design

decisions can be made in regard to the NGNP intermediate loop heat exchangers and the use of barrier coatings. The recent topical report by Sherman and Adams [9] recommends the use of in-situ oxide layers on structural metals, and the use of solid ceramics as the best tritium permeation barriers. Aluminide coatings are also suggested as a back-up technology because of their greater barrier potential and self-healing characteristics.

In FY09, the INL will be funded by the NGNP Project R&D branch to measure the tritium permeability of engineering materials of interest under natural and controlled atmospheric chemistry conditions. The assumed funding amount for this work is assigned as $500K, though this is a placeholder amount until the actual funding level is fixed by the Project. Following steady-state measurements, the effects of temperature changes on the permeability of samples with established coatings will be studied as well. P. Sharpe and M. Ebner are assigned as the P.I.'s on this work.

As an extension of the INL tasks, SRNL would be able to provide additional experimental capabilities and experienced personnel for work on barrier materials. SRNL has permeation test rigs that are currently used to support the SRS Tritium Facilities, and would be able to perform duplicate or additional measurements of tritium permeation through material samples of interest to the NGNP Project. Also, SRNL has much expertise in the use of tritium barrier coatings or layers, and would be able to expand the permeation studies to include alternative surface chemistry conditions, dynamic testing of barriers, and the use of aluminide layers as additional barriers. At SRNL, P. Korinko and K. Brinkman would be assigned as P.I.'s on this supporting work. This work is listed as optional at this time, because the basic measurements will be pursued by the INL.

The technical information that must be determined from these studies is shown in Table 2.

Table 2. Research Needs for High-Temperature Tritium Permeation Barriers

Priority	Area	R&D Tasks
1	Experimental: In-situ oxide tritium barriers	• Permeability versus temperature and H, D, T partial pressure • Determine oxide layer preparation recipe for Inconel 617/Haynes 230 • Measure permeability response after temperature transients • Determine method(s) of healing damaged barriers and study healing rates • Study effect of adding barrier layer on just one side of the base material or both sides of the base material • Surface layer characterization
2	Experimental: Aluminide-based tritium barriers	• Permeability versus temperature and H, D, T partial pressure • Determine aluminide/alumina layer preparation recipe for Inconel 617/Haynes 230 • Measure permeability response after temperature transients • Determine method(s) of healing damaged barriers and study healing rates • Surface layer characterization • Determine stability of aluminide layer at upper temperature range (use diffusion barrier layers if necessary to slow down dispersal of aluminide layer into base material)

2.3 Tritium Capture/Sequestration Systems

FY09 Priority: 1 and 2
Location: SRNL
Funding Needs:

Priority	Description	Year 1 Request ($K)	Year 2 Request ($K)
1	Development of Tritium Sequestration System for NGNP (SRNL, Clark, Klein, Brinkman)†	600	1,500
3	Evaluation of Tritiated Species Cracking/Decomposition Agents (SRNL, Korinko, Klein, Fox)†	400	200

†Task funded out of NGNP System Interface area.

Description:

SRNL will lead the effort to develop tritium sequestration systems for the multiple NGNP heat exchanger loops. The most appropriate tritium sequestration system for the NGNP is believed to be one that uses tritium getters. In such a system, a side stream of heat transfer fluid containing tritium is directly contact with gettering material in order to absorb tritium from the fluid stream. Tritium captured by the gettering material is then removed from the sequestration system along with the gettering material. Periodically, the gettering material along with absorbed tritium is removed from the sequestration system and replaced with fresh getter material, and the contaminated material is disposed as low-level radioactive waste.

Candidate high-temperature gettering materials (particularly Ti, Zr, Y and others) will be tested in the laboratory to determine their performance as tritium getters at high temperatures ($T>600^{o}C$) and low tritium partial pressures ($P<10^{-5}$ Pa). The side reactions of the getter materials with oxygen and water will also be measured in order to understand how adsorption of these materials affects the performance of the getter bed. From this data, operating scenarios can be developed, and this will help shape a design concept for a tritium sequestration system that is suitable for the NGNP. With the laboratory data, a conceptual design of a tritium sequestration system employing getters will be developed.

Gettering experiments will be performed in the laboratory in the first year, along with conceptual design work on a tritium sequestration system. In the second year, a lab-scale or pilot-scale tritium sequestration system will be constructed and tested in the SRS tritium facility in order to verify its operation and effectiveness in a nuclear industrial setting. If successful, the tests in the SRS Tritium Facility will validate the technology for use by the NGNP and will serve to advance the technology readiness levels (TRLs) of the sequestration technologies to the point where they can be adapted for full-scale use by the NGNP Project without additional lab-scale or pilot-scale testing.

In addition to the work with tritium getter materials, experiments are planned to examine the performance of tritium species cracking agents such as Ni-based and mixed metal catalysts and reactants. These cracking agents react with CH_4, NH_3 and other non-oxygenated contaminants to release tritiated hydrogen. Cracking of such species to form tritiated hydrogen is necessary because these species are not as readily absorbed as hydrogen on getter materials, and tritium capture efficiencies of getter systems are therefore reduced. Cracking agents, if used, would be employed upstream from a getter bed to improve tritium capture efficiencies. The non-hydrogen element of the contaminant is either absorbed by the cracking material or is released as a non-hydrogen-containing gas. The cracking experiments are listed as Priority 2 rather than Priority 1 because these experiments would provide an improvement to the function of a tritium sequestration system but is not absolutely essential to developing a basic tritium sequestration system.

A summary of recommended research topics in this area is shown in Table 3.

Table 3: Research Needs for High-Temperature Tritium Sequestration System

Sequence	Topic	Suggested Work
1	Experimental: Ti, Zr, Y as high-temp tritium getters (other getter materials are possible)	• Absorption kinetics • Getter capacity as function of temperature (bed breakthrough, storage capacity, etc.) • Effects of changing getter morphology (sponge, foil, etc.) • Effects of contaminant spikes on performance (excess water vapor, oxygen) • Saturated getter stability for storage purposes
1	Experimental: Ti, Zr, Y, Ni-Ti-Zr as high-temp moisture getters (other materials are possible)	• Absorption kinetics • Getter capacity as function of temperature (bed breakthrough, storage capacity, etc.) • Effects of changing getter morphology (sponge, foil, etc.) • Effect of excess oxygen on performance

2	Design: Conceptual Design of NGNP Tritium Barrier and Getter System	• Develop conceptual design of tritium barrier and capture system for NGNP System Interface
		  • Mass/energy balances
		  • Equipment sizing
		  • Operating schedules
		  • Capital and operating costs
		  • Disposal path for waste materials

2	Experimental: Non-oxygenated tritium cracking agents	• Reaction kinetics • Reaction conversion versus temperature, water vapor concentration • Bed stability • Effects of oxygen, water vapor on conversion reaction
3	Integrated testing of sequestration system using tritium instead of hydrogen or deuterium	• Intended to provide experimental testing and qualification of methods/equipment at higher Technology Readiness Levels (TRLs) • Goal is to reach TRL sufficient to install in NGNP facility

2.4 Dynamic Modeling of Tritium Migration

Priority: 1 and 2
Location: INL, *SRNL*
Funding Needs:

Priority	Description	Year 1 Request ($K)	Year 2 Request ($K)
1	Dynamic Modeling for Tritium Migration (INL, Oh and Kim)†	400	200
2	*Dynamic Modeling for Tritium Migration Using Aspen Custom Modeler (ACM) (SRNL, Gorensek)†*	*350*	*200*

† Task funded out of NGNP System Interface area.

Description:

There are several options for modeling the migration of tritium from the NGNP nuclear reactor to a downstream user of high-temperature heat, and a decision will need to be made in early FY09 to choose which option to use. Using THYTAN is one option. THYTAN is a code developed by JAEA to model the movement of tritium in the HTTR system, and has been adapted to model some configurations of the NGNP, including a simplified hydrogen plant layout. TMAP7 is another option. TMAP7 was developed by the INL for modeling the movements of tritium in fusion systems, and could be adapted to model the NGNP. Building a custom model in MATLAB is another option, and some work has been accomplished in FY08 to reproduce the capabilities of THYTAN in a non-proprietary MATLAB format. Yet another option, not previously considered, is to use Aspen Custom Modeler to perform the work. These models are described in more detail below, and the work needed in FY09 and beyond is described.

THYTAN is a Fortran 95-based code that uses a node-link structure to calculate the movement of tritium through systems of code-defined configurations. It is not a GUI-based code, and models of the system being studied must be programmed. JAEA owns the source code for THYTAN, and modifications to the source code cannot be performed outside of JAEA without express permission to do so by them. The NGNP Project is negotiating with JAEA for the use of the THYTAN code and supporting HTTR tritium data (which is needed to help validate the code), but other issues related to intellectual property rights and the use of the source code by the NGNP Project have prevented its use by the NGNP Project so far. If the NGNP Project gets the rights to the THYTAN source code, then THYTAN work by the NGNP Project can be resumed.

TMAP7 is a Fortran 77-based code that also uses a node-link structure to calculate the movement of tritium through systems of code-defined configurations. INL owns the rights to the code. Although written in Fortran 77, the obsolete structures in the code have been removed, so that it can be compiled using more modern compilers. The performance of TMAP7 is expected to be similar to THYTAN. Additional work would be necessary to build models of the NGNP in TMAP7, which has already been accomplished to some extent in THYTAN.

MATLAB with Simulink is a modern software package that reduces the burden of developing computer code in order to perform the detailed calculations needed to track the movement of tritium in the NGNP. Some tritium work has already been accomplished at the INL with MATLAB, and the results that were generated by THYTAN in Reference [2] have been reproduced using MATLAB. MATLAB is available commercially, and any model developed in MATLAB is entirely portable, as long as the user has the MATLAB software.

The last option, not previously considered, is to use Aspen Custom Modeler to model the movement of tritium in the NGNP. Aspen Custom Modeler is a dynamic software package sold

by AspenTech to model the time-dependent behaviors of batch, semi-batch and continuous chemical plants. The advantage of using Aspen Custom Modeler is that the full flow sheets for the three hydrogen plant technologies (S-I, Hybrid Sulfur, and high-temperature electrolysis) have already been implemented in Aspen, and those flow sheets could be used in this work with little modification. Also, adding models of new processes or tritium separation systems would be relatively easy. Use of an Aspen-based model requires the user to have access to Aspen software.

Among the first three options (THYTAN, TMAP7, MATLAB), the software chosen is exclusive, and only one of the three software models should be pursued for the NGNP. The fourth option, using Aspen Custom Modeler, may be pursued in parallel with one of the other three choices because it would provide additional flow sheeting capabilities that are harder to obtain using the other software models. Table 4 shows some specific items that need to be addressed by work in this area.

Table 4. Research Needs in Tritium System Modeling

Sequence	Topic	Suggested Work
1	Modeling: Dynamic modeling of tritium migration in NGNP	• Validate THYTAN code or other code/software for use with NGNP tritium migration work • Analyze NGNP tritium migration scenarios in dynamic and steady-state situations using updated NGNP configurations and physical data from the experiments when it becomes available.

2.5 Instrumentation

Priority: 2
Location: SRNL
Funding Needs:

Priority	Description	Year 1 Request ($K)	Year 2 Request ($K)
2	On-line Sensors for Measurement of Dilute Tritium (SRNL, Ferrar, Cordaro and Sigg)†	1,200	1,200

†Task funded out of NGNP System Interface area.

Description:

Tritium monitoring techniques have been developed for commercial and defense reactor liquid and gaseous effluents as well for tritium extractions plants located at the Savannah River Site(SRS) [10]. For gases, tritium is measured using ionization detectors which rely on the tritium beta decay (e^-) to setup a current which is directly proportional to the concentration of tritium in the gas stream. Sensitivity of the ionization chamber is primarily dependent on the chambers volume and the sensitivity of the electrical circuit that measures the resulting current. SRNL specifies and or builds all of the tritium monitoring systems used at SRS. Commercial instruments are incorporated into the design whenever possible but the unique process conditions

typically require designs that are not commercially available. In addition to ion chambers, SRNL has developed advanced electrometers for industrial use that measure at femtoamp resolution while providing state of the art reliability and maintainability features. For liquids, most applications use liquid scintillation detectors which rely on the interaction of ionizing radiation with liquid scintillation cocktails to emit light photons. The light photons are counted with photomultiplier-based counting systems. Other methods to measure tritium at the SRS and Japan Atomic Energy Research Institute rely on the low energy photons (Bremsstrahlung radiation) that are produced as tritium's beta decay particles lose energy from interaction with matter. The Bremsstrahlung radiation is detected using a low-energy photon spectrometer such as a sodium iodide detector having beryllium window and the radiation is counted by a photomultiplier [11, 12].

In the case of the SRS application, several technologies were evaluated to safely analyze the tritiated water samples including liquid scintillation counting (LSC), solid scintillation counting, mass spectrometry and ion chambers before deciding to pursue bremsstrahlung counting. LSC, the most commonly used tritium analysis technique, would require multiple dilutions to avoid excessively high count rates. The additional sample handling and dilutions required for LSC would increase exposure risk to personnel. Less dilution is needed for solid scintillants since they are less efficient for short-range tritium betas (most are stopped by water before reaching scintillant); however, their performance degrades as contaminants foul the scintillant surfaces. Mass spectrometry generally requires higher concentrations than those of concern for this application. Ionization chambers are capable of detecting low tritium concentration over a wide dynamic range but are very sensitive to impurities, easily contaminated, and difficult to decontaminate. Induced currents and charge leakage across insulators also pose problems in applying ion chambers [13, 14, 15].

Limits will be imposed on the amount of tritium that can be tolerated in chemical processes downstream from the nuclear reactor in the NGNP facility, and those limits are likely to be near the EPA and NRC limits of tritium in gaseous and water effluents. At such small concentrations of tritium, only counting techniques are useful, but such instruments tend to be bench-top equipment. Ideally, it is desired to adapt bench-top devices for use as on-line instruments for the NGNP heat transfer loops or for downstream chemical plants, so that information on tritium concentration is available on a periodic basis without the need for manual sampling. With such instrumentation, measured tritium concentrations that rise above certain action levels could trigger system alarms or other automatic control system options.

This task focuses on the development and testing of on-line instrumentation that could be used with high-temperature heat transfer lines. Figure 4 shows potential monitoring points for an NGNP facility using a high-temperature electrolysis hydrogen plant. Tasks in FY09 would include a topical report on instrumentation options, acquisition of two instruments (one for liquid streams, one for gas streams), modification of the acquired instruments (if necessary) for on-line service, and testing of the instruments using non-radioactive analogs. In FY10, these instruments would be modified for radioactive service and tested directly in the SRS Tritium Facilities using tritium standards. Table 5 shows some specific items that need to be addressed by work in this area.

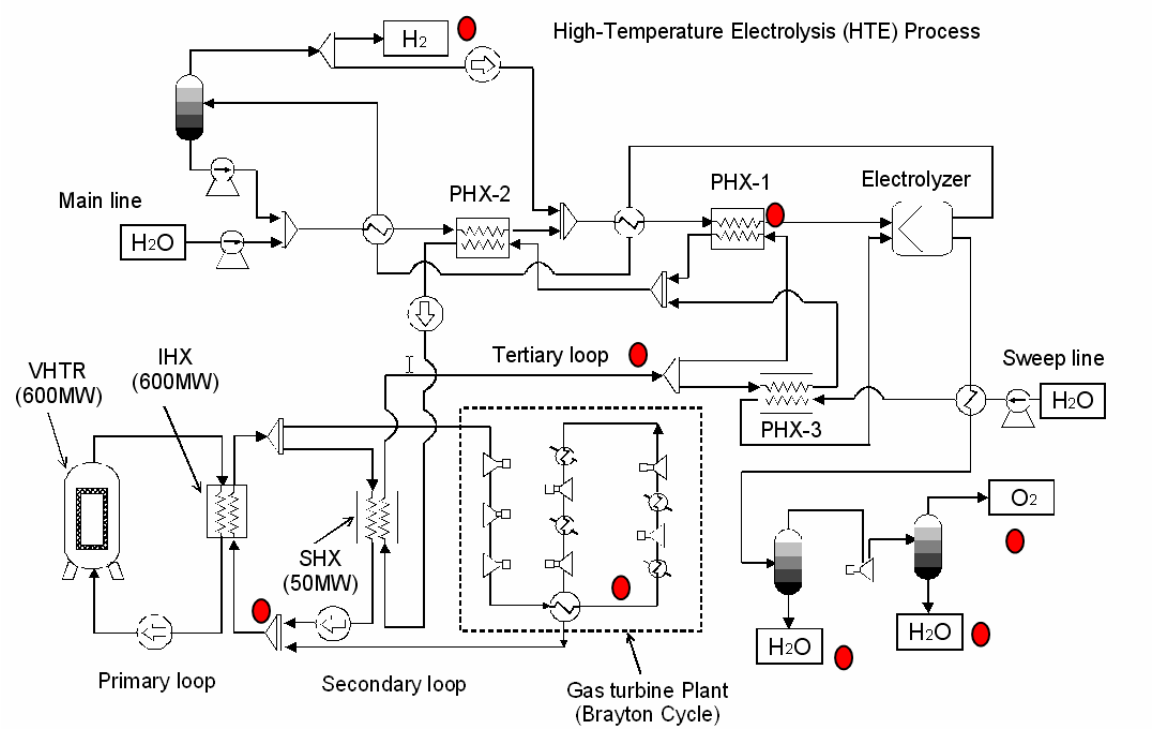

Figure 4. Potential monitoring points for tritium contamination.

Table 5. Research Needs in On-line Tritium Instrumentation

Sequence	Topic	Suggested Work
1	Literature Survey and Experimental: Tritium detection instrumentation	• Examine available instrumentation for quantification of very dilute tritium • Adopt/adapt/develop concept(s) for on-line measurements • Test proposed on-line tritium detection instrumentation

2.6 Waste Handling and Disposal

Priority: 2
Location: INL and/or SRNL
Funding Needs:

Priority	Description	Year 1 Request ($K)	Year 2 Request ($K)
2	Development of Tritiated Materials/Components Disposal Strategies (SRNL, Clark)†	105	0

† Task funded out of NGNP System Interface area.

Description:

A critical aspect to the management of tritium systems is the disposal of tritiated waste from the affected facilities. The INL and/or SRNL will develop a tritiated waste disposal strategy for the NGNP heat exchangers and other components. This analysis will consider periodic component and materials disposition as well as failed component disposal, and will make initial bounding assumptions as to the possible tritium concentration level to be disposed. This analysis will evaluate various disposal options will be evaluated and a hierarchical decision tree analysis will be performed to support the waste disposal strategy. Table 6 shows some specific items that must be addressed by this task.

Table 6. Analysis Needs in the Waste Handling and Disposal Area

Sequence	Topic	Suggested Work
1	Analyses: Waste Handling and Disposal	• Determine types, quantities of waste and projected tritium levels • Identify waste disposal optons • Develop decision tree analysis to support waste disposal strategy

3. COLLABORATION OF U.S. LABORATORIES AND AECL

The establishment of an international collaborative framework by the Generation IV International Forum (GIF) Hydrogen Project Management Board has allowed for the cooperation of U.S. national laboratories with laboratories in other GIF countries. In this case, the INL and SRNL plan to work with Atomic Energy of Canada Limited (AECL) Chalk River Laboratories in order to increase the capabilities for technical reviews and to open up the possibility for shared tritium-related experiments between the U.S. and Canada. AECL Chalk River Laboratories is a recognized authority in Canada on tritium, and their expertise and facilities can be made available to the NGNP Project.

On June 23-24, 2008, a kick-off meeting was held at AECL Chalk River Laboratories to start the collaboration. At the meeting, SRNL was represented by S.R. Sherman (steven.sherman@srnl.doe.gov) and T.M. Adams (thad.adams@srnl.doe.gov), and AECL was represented by Dr. Sam Suppiah (suppiahs@aecl.ca). The purpose of the visit was to discuss the development of a more comprehensive collaboration concerning tritium migration barrier materials and tritium separation systems for application to high-temperature gas-cooled reactor systems such as the NGNP. The results of the discussions were that collaboration is welcome, self-healing in-situ oxide surface layers would likely make the best barriers, and tritium getters such as Ti spong would be best for capturing dilute tritium in the NGNP heat transfer fluid streams. AECL has a tritium lab capable of working with up to one million Ci of tritium and it is available for use by the collaboration.

In FY09 and beyond, it is proposed that the sharing of technical information related to tritium be continued, and efforts be made in the U.S. to encourage AECL to provide data or pursue experiments that complement the work planned for execution in the U.S. If successful, this working relationship may be expanded to the conduct of joint experiments in future years. As a measure of good faith, SRNL will supply AECL with the tritium topical report that has recently been released [9]. The collaboration will continue to be managed under the current GIF agreement if the nature of the collaboration remains general and there are no issues related to intellectual property protection. If there arise certain items from the collaboration that require strong intellectual property protection and the U.S. and Canada are wary of sharing the information with other GIF members, then there exists the possibility of managing such research under other bilateral agreements (e.g., International Nuclear Energy Research Initiative, I-NERI). However, the GIF will be the primary path for managing the collaboration, as directed by the DOE Office of Gas Reactor Deployment (NE-33), and all efforts will be made to use the existing GIF agreement to manage the collaboration.

REFERENCES

[1] NGNP Project, *Next Generation Nuclear Plant Project: Preliminary Project Plan*, PLN-2489, Idaho National Laboratory, June 2007.

[2] H. Ohashi, S.R. Sherman, *Tritium Movement and Accumulation in the NGNP System Interface and Hydrogen Plant*, INL/EXT-07-12746, Idaho National Laboratory, June 2007.

[3] D. Greneche, W.J. Szymczak, "The AREVA HTR Fuel Cycle: An analysis of technical issues and potential industrial solutions," *Nucl. Eng. and Des.*, Vol 236, 2006, pp. 635-642.

[4] U.S. DOE, *A Technology Roadmap for Generation-IV Nuclear Energy Systems*, GIF-002-00, U.S. DOE Nuclear Energy Research Advisory Committee and the Generation IV International Forum, December 2002.

[5] U.S. DOE, *The U.S. Generation-IV Implementation Strategy*, DOE Office of Nuclear Energy, September 2003.

[6] 42 USC 14801, 2005, "Energy Policy Act of 2005," (EPACT 2005), Federal Energy Regulatory Commission, August 8, 2005.

[7] U.S. DOE, *DOE Handbook: Tritium Handling and Safe Storage*, DOE-HNBK-1129-99, Mar 1999.

[8] H. Ohashi, N. Sakaba, T. Nishihara, Y. Inagaki, K. Kunitomi, "Numerical Study on Tritium Behavior by Using Isotope Exchange Reactions in Thermochemical Water-Splitting Iodine-Sulfur Process," *J. Nucl. Sci. and Technology*, Vol 44, No 11, 2007, pp 1407-1420.

[9] S.R. Sherman, T. M. Adams, "Tritium Barrier Materials and Separation Systems for the NGNP," WSRC-STI-2008-00358, Washington Savannah River Company, Aiken, SC, August 2008.

[10] J.R. Devore, M.A. Buckner, "Tritium Monitoring Techniques," ORNL/TM-13132, Oak Ridge National Laboratory, Oak Ridge, TN, May 1996.

[11] R.A. Sigg, D.P. DiPrete, D.J. Pak, L.M. Arrigo, "Remote Analysis of High Tritium Water," WSRC-STI-2007-00417, Savannah River National Laboratory, Washington Savannah River Company, Aiken, SC, August 2007.

[12] W.M. Shu, M. Matsuyama, T. Suzuki, M.F. Nishi, "Characteristics of a promising tritium process monitor detecting bremmsstrhalung X-rays," *Nucl.Inst. and Meth. In Phys.Rev.*, Vol A521, 2004, pp 423-429.

[13] R. Sigg, J. McCarty, R. Livingston, M. Sanders, "Real-Time Aqueous Tritium Monitoring Using Liquid Scintillation Counting," *Nucl.Inst. and Meth. In Phys.Rev.*, Vol A353, 1994, p 494.

[14] K. Hofstetter, "Continuous Aqueous Tritium Monitoring," *Fusion Technology*, Vol 28, 1996, p 1527.

[15] G. Knoll, *Radiation Detection and Measurement*, Wiley, New York, 2000.