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TRITIUM CONTROL and CAPTURE in SALT-COOLED FISSION AND FUSION REACTORS: EXPERIENTS, MODELS, and BENCHMARKING

Workshop Proceedings

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ABSTRACT

PROCEEDINGS of the WORKSHOP on TRITIUM CONTROL and CAPTURE in SALT-COOLED FISSION AND FUSION REACTORS: EXPERIENTS, MODELS, and BENCHMARKING

The *Workshop on Tritium Control and Capture in Salt-Cooled Fission and Fusion Reactors: Experiments, Models and Benchmarking* was held on October 27-28, 2015 in Salt Lake City. The workshop objectives were to bring together researchers involved in experiments, modeling and benchmarking for tritium control at ~700°C in liquid salts and related systems to (1) exchange information and enable the future exchange of information, (2) initiate an effort for benchmarking of experiments and models, and (3) encourage cooperation between different groups working on the same challenges.

The workshop was organized by 5 organizations with common interests in tritium in salt systems at high temperatures: the Department of Nuclear Science and Engineering (NSE) at the Massachusetts Institute of Technology (MIT), the Nuclear Reactor Laboratory (NRL) at MIT, the Plasma Fusion Center (PFC) at MIT, the University of Wisconsin at Madison (UW) and the Chinese Academy of Science (CAS).

These diverse organizations have a common interest because three advanced power systems use liquid salt coolants that generate tritium and thus face common challenges. The Fluoride-salt-cooled High-temperature Reactor (FHR) uses the same graphite-matrix coated-particle fuel as high-temperature gas-cooled reactors and fluoride salt coolants. Molten salt reactors (MSRs) dissolve the fuel in a fluoride or chloride salt with release of fission product tritium to the salt. In both systems, the base-line salts contain ^{7}Li. Isotopically separated lithium is used to minimize tritium production. The Chinese Academy of Science plans to start operation of a 10-MWt FHR and a 2-MWt MSR by 2020. High-magnetic field fusion machines proposed to use lithium enriched in ^{6}Li to maximize tritium generation—the fuel for a fusion machine. Advances in superconductors that enable higher power densities may require the use of lithium salts as coolants.

This proceedings summarize results from that workshop including descriptions of the power systems that use high-temperature salts, the common chemistry and tritium challenges, ongoing work removing tritium using carbon, other technologies for tritium control, and other tritium capabilities in the U.S. The appendixes include the workshop agenda, participants, and presentations from the workshop.

ACKNOWLEDGEMENTS

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1. INTRODUCTION

The *Workshop on Tritium Control and Capture in Salt-Cooled Fission and Fusion Reactors: Experiments, Models and Benchmarking* was held on October 27-28, 2015 in Salt Lake City. The workshop objectives were to bring together researchers involved in experiments, modeling and benchmarking for tritium control at ~700°C in liquid salts and related systems to:

- Exchange information and enable the future exchange of information
- Initiate an effort for benchmarking of experiments and models
- Encourage cooperation between different groups working on the same challenges.

The workshop was organized by five organizations with common interests in tritium control in salt systems at high temperatures

- Department of Nuclear Science and Engineering (NSE) at the Massachusetts Institute of Technology (MIT)
- Nuclear Reactor Laboratory (NRL) at MIT
- Plasma Fusion Center (PFC) at MIT
- University of Wisconsin at Madison (UW)
- Chinese Academy of Science (CAS).

These diverse organizations have a common interest because three advanced power systems use liquid salt coolants that generate tritium and thus the need to avoid tritium (a health hazard) from escaping to the environment. The Fluoride-salt-cooled High-temperature Reactor (FHR) uses the same graphite-matrix coated-particle fuel as high-temperature gas-cooled reactors and fluoride salt coolants. Molten salt reactors (MSRs) dissolve the fuel in a fluoride or chloride salt with release of fission product tritium to the salt. In both systems, the base-line salts contain ^{7}Li. Isotopically separated lithium is used to minimize tritium production. The Chinese Academy of Science plans to start operation of a 10-MWt FHR and a 2-MWt MSR by 2020.

The third power system are advanced fusion machines with liquid-salt coolants. In fusion machines tritium is produced and used as a fuel. There are the dual requirements of (1) avoiding release of tritium to the environment and (2) efficient recovery of tritium as the fuel. The tritium inventories are several orders of magnitude larger than in fission machines. High-magnetic field fusion machines proposed to use lithium enriched in ^{6}Li to maximize

tritium generation. Recent advances in superconductors that enable higher power densities may require the use of lithium salts as coolants.

We summarize results from that workshop including descriptions of the power systems that use high-temperature salts (Section II), the common chemistry and tritium challenges (Section III), ongoing work removing tritium using carbon (Section IV), other technologies for tritium control (Section V), and major tritium capabilities in the U.S. (Section VI). Table 1 lists the talks by general area. The workshop was organized so half the time was reserved for discussions

Appendix A contains the workshop agenda. Appendix B contains the list of participants and Appendix C are the presentations from the workshop.

Table 1: Workshop Talks by Subject Area

Fission and Fusion Reactor Systems with Salt Cooling and Tritium Challenges

C. Forsberg (MIT NSE): Fluoride-Salt-Cooled High-Temperature Reactor (FHR)
B. Sorbom (MIT PSFC): Liquid Immersion Blankets for Fusion Power Plants
W. Liu (CAS): Thorium Molten Salt Reactor (TMSR) Project in China
M. Laufer (UCB): Tritium and Chemistry Management for the Mark-1 PB-FHR

Tritium Generation, Corrosion Chemistry and Modeling

J. Stempien (INL) Tritium Transport and Corrosion Modeling in the Fluoride Salt-Cooled High-Temperature Reactor
T. Chrobak (UW): FLiBe Electrochemistry and Materials Corrosion Research at UW

Tritium Control and Carbon

C. Contescu and T. Burchell (ORNL): Hydrogen – Carbon Interactions: A Brief Literature Survey
T. Burchell and C. Contescu (ORNL): AGR Fuel Compact Development Program
H. Wu (UW): Experimental Work on Tritium Transport Analysis in Flibe-Graphite system
S. Lam (MIT NSE): Tritium Control Using Carbon Outside the Core
H. Wu (UW) Effect of Hydrogen on Tritium Control in Molten Salt System
D. Carpenter (MIT NRL) Planned FHR IRP-2 Tritium Experiments at the MIT NRL

Tritium Control with Other Technologies

W. Liu (CAS): Tritium-Control Technologies for TMSR System in CAS (gas sparging)
F. (UNM): Research on Techniques for Tritium Sequestration and Removal at UNM
B. Wallace (UNM): Investigation of Tritium Control and Release Mitigation Options in Double-Wall Twisted-Tube Heat Exchangers (DT-HXRs)
P. Humrickhouse (INL): Tritium Permeation Control and Extraction – Perspectives from Fusion System Studies

X. Sun (OSU): Tritium Management in FHRs: Ongoing and Planned Activities in Integrated Research Project Led by Georgia Tech

Tritium Control Experience

G. Stack (SRNL): An Overview of SRNL Tritium Activities

M. Shimada (INL): Overview of Tritium and Molten Salt FLiBe research at Safety and Tritium Applied Research (STAR) facility

D. Carpenter (MIT NRL): Experience with Tritium Evolution During Irradiation of MSRE Flibe in the MITR

D. Senor (PNNL): Irradiation Testing in Support of the Tritium Production Enterprise

2. SALT-COOLED FISSION AND FUSION POWER SYSTEMS

There is a rapidly growing interest in fission and fusion systems that use salt coolants that is driven by (1) separate developments in FHRs, MSRs, and fusion and (2) advances in gas turbines that can couple to salt-cooled reactor systems. Workshop presentations (Forsberg, Sorbom, Liu and Laufer) and discussions summarized the multiple basis for interest in developing these technologies.

2.1. Salt-Cooled Power Systems

2.1.1. Fluoride-salt-cooled High-temperature Reactors (FHRs)

The FHR uses salt coolant and the graphite-matrix coated-particle fuel developed for High-Temperature Gas-cooled Reactors (HTGRs). Advances in the fuel are enabling the development of the FHR. Because this reactor uses a proven fuel and a clean salt coolant, it is the near-term commercialization option for a salt-cooled reactor. Three different fuel designs are proposed by different groups (Fig. 1).

- *Pebble bed*. The pebble-bed FHR[1] uses 3-cm diameter graphite pebbles with embedded coated-particle fuel—the same basic fuel that was used in the German HTGRs and will be used in the Chinese HTGRs that are under construction. The pebbles are 3-cm rather than the traditional 6-cm diameters used in HTGRs to increase surface area per unit volume of the core to allow higher power densities. The pebble-bed FHR design is the most developed. The Chinese Academy of Sciences plans to complete a 10 MWt pebble-bed FHR test reactor by 2020. Like pebble-bed HTGRs, this design allows online refueling. It is the near-term option.
- *Plate fuel*. Oak Ridge National Laboratory[2] is developing a plate fuel where the hexagonal fuel assembly is similar in shape to a sodium-cooled reactor fuel assembly. The fuel plates are made of a carbon composite with the coated-particle fuel on the plate surfaces. It is a "traditional" type fuel assembly with a refueling strategy similar to a sodium fast reactor—another low-pressure reactor.
- *Fuel Inside Radial Moderator (FIRM).*[3] This FHR core design is somewhat similar to the operating British Advanced Gas-Cooled Reactors (AGRs) except for use of salt coolant, higher power densities and the details of the fuel design. The AGRs are graphite-moderated carbon-dioxide-cooled high-temperature reactors with exit gas temperatures of 650°C. The AGR fuel consists of UO_2 pellets in stainless steel pins with an assembly consisting of a circular array of pins inside an annular graphite shell. Fourteen AGRs have been operating for several decades. The FHR FIRM assembly replaces the AGR fuel assembly with a graphite cylinder containing liquid-salt cooling channels and fuel channels filled with coated-particle fuel in carbon-matrix pellets—a cylindrical variant of the prismatic fuel blocks used in some HTGRs. FIRM assemblies would be refueled using the same refueling strategies used by the AGR, pulling assemblies straight up through the vessel cover. AGRs refuel on-line at about 650°C, similar to FHR operating temperatures.

Fig. 1. Alternative FHR Fuel Designs

The base-line coolant is a lithium-beryllium-fluoride salt known as flibe ($^7Li_2BeF_4$). The characteristics of the flibe as well as other potential salts are listed in Table 2. The primary coolant system is a closed loop that operates at atmospheric pressure with nominal core coolant inlet and outlet temperatures of 600°C and 700°C respectively.

There are proposals for FHRs using advanced fuels including pin-type fuels with SiC clad. These are longer term options that do not change the need for tritium control strategies.

Table 2. FHR Coolant Options[1]

Coolant	T_{melt} (°C)	T_{boil} (°C)	ρ (kg/m^3)	ρC_p (kJ/m^3 °C)
66.7^{7}LiF-33.3BeF_2	**459**	**1430**	**1940**	**4670**
59.5 NaF-40.5 ZrF_4	500	1290	3140	3670
26 ^{7}LiF-37 NaF-37 ZrF_4	436		2790	3500
51 ^{7}LiF-49 ZrF_4	509		3090	3750
Water (7.5 MPa)	0	290	732	4040

[1]Compositions in mole percent. Salt properties at 700°C and 1 atmosphere. Pressurized water data shown at 290°C for comparison

2.1.2. Molten Salt Reactors (MSRs)

Molten salt reactors were first developed as part of the nuclear aircraft propulsion program in the 1950s and then as a thermal-neutron-spectrum breeder reactor using the thorium fuel cycle in the 1960s. The Molten Salt Reactor Experiment (MSRE), an 8-MWt reactor (Fig. 2), successfully demonstrated the technology in the late 1960s. This reactor used flibe ($^{7}Li_2BeF_4$) salt with fuel and fission products dissolved in the salt. The reactor used bare graphite as the neutron moderator. The program was cancelled in the early 1970s when the United States decided to focus its breeder reactor program on sodium-cooled fast reactors (SFRs). In the last decade[4, 5] there has been a renewed interest in MSR for several reasons.

Fig. 2. Molten Salt Reactor Experiment

- *Fuel cycle versatility*. MSRs can operate on a variety of fuel cycles including thorium breeder fuel cycles and various cycles that destroy actinides. This capability has been increased by recent work to develop fast spectrum MSRs.
- *Advancing technology*. Many of the technology challenges of the 1960s have been reduced or eliminated thanks to advances in other fields. Better high-temperature carbon forms can provide better materials for reactor internals. The development of high-temperature additive manufacturing enables fabrication of complex components including control rods, distillation columns, and other items out of molybdenum and other salt-compatible high-temperature materials. Advanced carbon and metal absorbers may enable efficient removal of noble metal fission products from the molten salt. Unless removed, these fission-

product metals plate out on heat exchanger surfaces, are a major source of short-term decay heat, and cause multiple challenges. Last, advances in metallurgy may enable development of alloys that allow higher temperature operations.

- *Safety*. MSRs enable alternative safety strategies relative to solid fuel reactors including the options of (1) dumping the liquid fuel to critically-safe passively cooled tanks under any accident scenario and (2) potentially the option to minimize the inventory of longer lived fission products such as cesium in the reactor that may dominate the accident source term and the potential for land contamination. The second option is enabled by the advancing separation technologies enabled by new materials and material fabrication methods as noted above, for components such as distillation columns.

The Chinese Academy of Science plans to build a small 2-MWt MSR by 2020 with an emphasis on the thorium fuel cycle for fuel sustainability. Several large western companies (Terrapower, Hatch) are examining MSRs as are many smaller companies.

2.1.3. High Magnetic-Field Fusion Reactors

Advances in magnetic fusion may drive fusion systems to use liquid salt coolants. The size of magnetic fusion devices for any given fusion power level is determined by the maximum feasible magnetic field with the size proportional to one over the magnetic field to the fourth power. Practical fusion machines require superconducting wire or tape to generate the magnetic fields to minimize electrical consumption by the magnets. However, standard superconductors lose their superconducting properties in high magnetic fields. In the last five years, methods have been developed to manufacture a new superconductor: Rare-Earth Barium Copper Oxide (REBCO). This new superconductor enables magnetic fields at the coil over 22 Tesla—more than twice the capability of older superconductors. It eliminates magnetic field strength as the primary design constraint in magnetic confinement fusion devices with the new limit being magnetic field induced stress in the coils. The REBCO is in the form of a steel tape that enables addressing the high stresses.

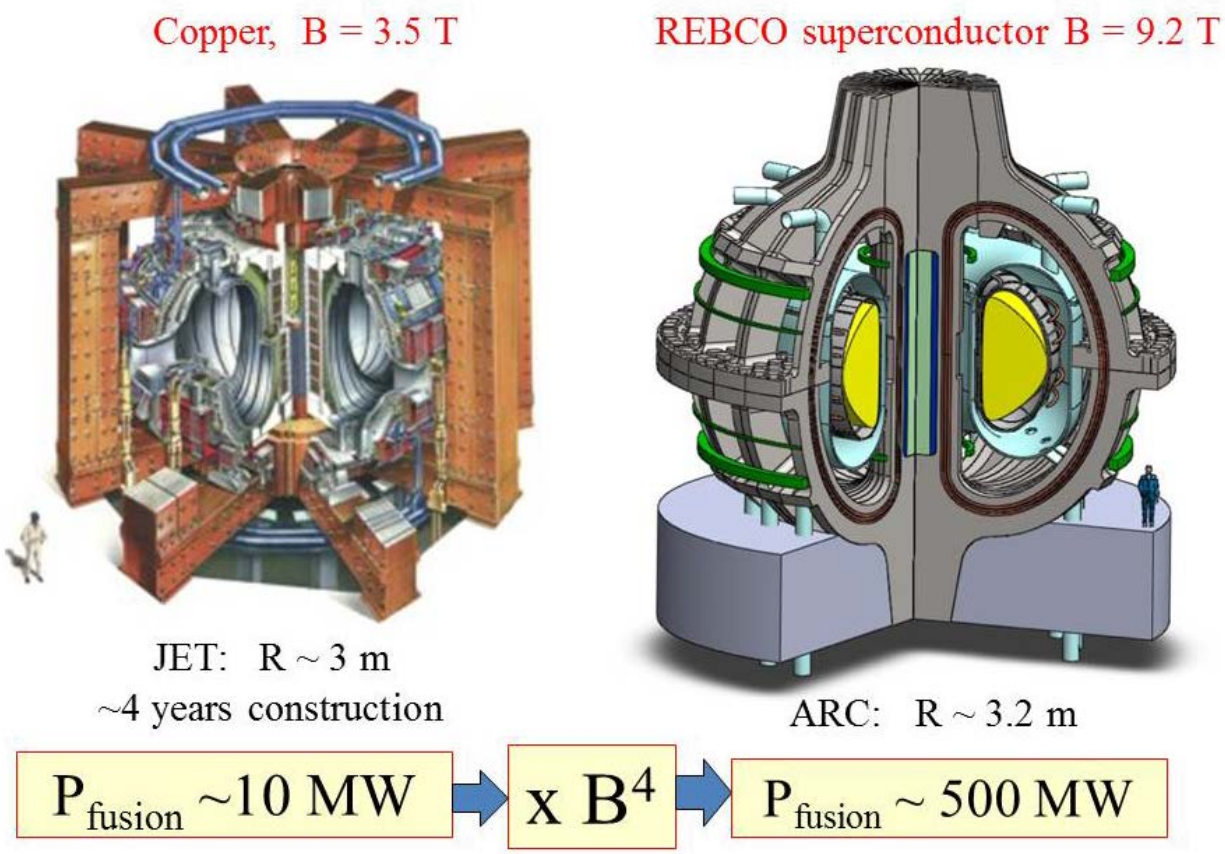

Fig. 3. Impact of Higher-Field Superconductors on the Size of Magnetic Fusion System

REBCO superconductors may enable doubling the practical peak magnetic field in a fusion machine and thus reduce the volume of fusion systems by an order of magnitude. The radius of a 500 MW plasma fusion system would be about 3 meters—the size of several magnetic fusion devices already built. Figure 3 shows JET (an existing fusion experimental device in the United Kingdom) and the proposed high magnetic field fusion system based on REBCO superconductors.[6] It is potentially a revolution in fusion.

Increasing fusion power density by an order of magnitude improves long-term economic viability. However, it imposes major changes in fusion blanket design because of the much higher power densities. Historically proposed blankets have been solid lithium-containing materials for production of tritium fuel ($^{6}Li + n \rightarrow {}^{3}H + {}^{4}He$). The higher power densities will likely require changing to a liquid blanket containing lithium—most likely flibe ($66.7^{6}LiF\text{-}33.3BeF_2$) for several reasons. A liquid blanket is needed for highly efficient neutron shielding with such high plasma fusion power densities. Fusion generates about 17 MeV per fusion of tritium and deuterium—most of this energy is in the form of 14 MeV neutrons that deposit their energy as heat in the liquid. The heat transfer challenges in solid fusion blankets become very difficult at these very high power densities. The coolant choices are (1) fluoride coolant salts—most likely flibe or (2) a liquid metal coolant containing lithium (lithium, lead-lithium, etc.). A low-electrical-conducting liquid salt rather than liquid lithium or a lead-lithium eutectic is preferred to ease magneto hydrodynamic issues such as coolant pumping and plasma control because the magnetic fields produced by external coils more rapidly penetrate through the blanket. These factors may drive magnetic fusion to liquid salt cooling.

2.2. Nuclear Air Brayton Combined Cycles (NACC)

Salt coolants were originally developed for the Aircraft Nuclear Propulsion Program in the 1950s with the goal of coupling a nuclear reactor to aircraft jet engines. They can transfer heat from the reactor to the power cycle at between 600 and 700°C. Recent advances in utility natural-gas combined-cycle technologies now enable coupling these reactors to a Nuclear Air-Brayton Combined Cycle (NACC) or potentially a Nuclear Helium Combined Cycle (NHCC). NACC can provide base-load electricity with additional variable peak electricity produced by using auxiliary natural gas, biofuels, hydrogen, or stored heat to (1) increase nuclear plant net revenue by 50 to100% relative to base-load nuclear plants and (2) enable a low-carbon nuclear renewable electricity system. These developments create large incentives to develop salt-cooled reactors.

During base-load operation of a NACC,[7, 8, 9] atmospheric air is filtered, the air is compressed, heat is added from the reactor through a coiled-tube heat exchanger (CTHX), the hot compressed air goes through turbines to produce electricity, the warm air exiting the gas turbine goes through a heat recovery steam generator to generate steam that is used to produce added electricity, and the air is exhausted to the stack. This power cycle is similar to that used in natural gas combined-cycle plants. If coupled to a salt-cooled reactor delivering heat between 600 and 700°C, heat-to-electricity efficiency is 42%. This specific example uses a modified General Electric 7FB gas turbine.

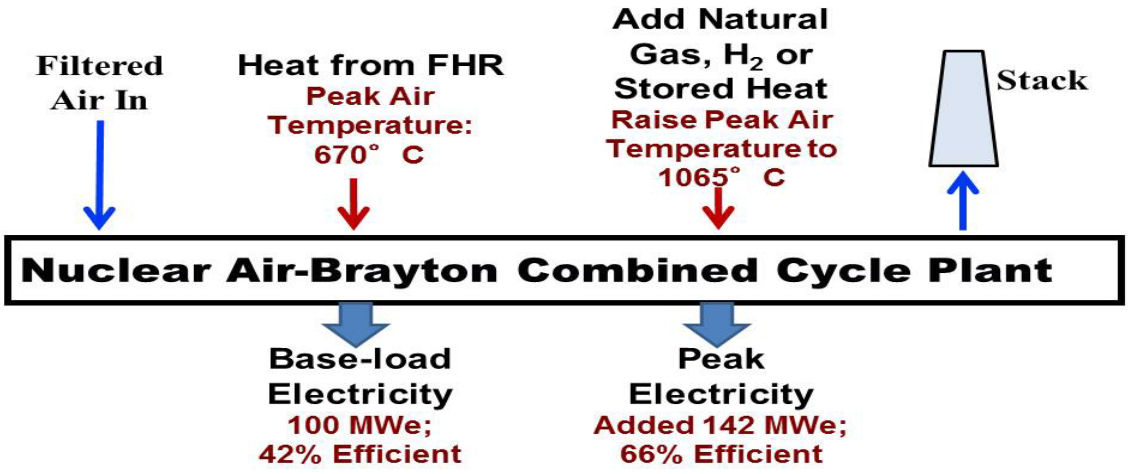

Fig. 4. Nuclear Air-Brayton Combined Cycle (NACC)

The base-load NACC temperatures, determined by heat-exchanger materials constraints, are far below maximum peak gas turbine temperatures. Thus, there is the option of adding heat after the nuclear heating to further raise compressed gas temperatures before entering a power turbine—a topping cycle. The incremental heat-to-electricity efficiency depends upon the design, ranging from 66 to 70%. This is the most efficient system known to convert heat to electricity based on existing technology.

An economic analysis[10] was done on the performance of an FHR with NACC in California and Texas using natural gas to produce peak electricity. These states have deregulated electricity markets. The peaking capability increased the plant yearly revenue by about 50% after subtracting the cost of the natural gas compared to a base-load nuclear plant. Because NACC is more efficient than a stand-alone natural-gas combined cycle plant in

converting natural gas to electricity (uses less natural gas), its electricity production costs for peak electricity are less than a stand-alone natural gas plant; thus, it earns large profits when electricity prices are set by natural gas plants.

The addition of wind and solar in some electricity grids has resulted in significant hours per year with very low electricity prices—near zero at times of high wind or solar input.[11, 12, 13] In such utility systems it is proposed that a Firebrick Resistance-Heated Energy Storage (FIRES) system replace the use of natural gas for providing heat to produce peak electricity. FIRES consists of high-temperature firebrick heated to high temperatures with electricity at times of low or negative electric prices. The firebrick, insulation systems, and most other storage system components are similar to high-temperature industrial recuperators. For peak electricity production, the compressed air after nuclear heating is sent through the firebrick to raise its temperature before going to the turbine. The round-trip storage efficiency from electricity to heat to electricity is ~66%, based on ~100% efficiency in resistance electric conversion of electricity to hot firebrick and 66% efficiency in conversion of incremental heat to electricity within NACC. FIRES enables the reactor to operate at base-load at all times while the station buys electricity from the grid at times of low prices to charge FIRES and sells electricity at times of high prices.

3. SALT CHEMISTRY AND TRITIUM GENERATION

Table 3 summarizes some of the differences and similarities in salt coolant requirements among the different reactor concepts. Carbon in the system can have a large impact on system behavior because carbon can absorb tritium and other impurities in the salt and has other chemical impacts.

Table 3. Salt Characteristics of Different Systems

Property	FHR	MSR	Fusion
Salt	Fluoride	Fluoride or Chloride (fast spectrum only)	Fluoride (Flibe)
Impurities	Corrosion impurities and possible fission product impurities	High concentrations of fission products and actinides	Corrosion impurities
Use lithium salts	Optional	Depends upon goals	Required
Tritium production	Small (^{7}Li in Coolant)	Small (^{7}Li in Coolant)	High (^{6}Li in Coolant)
Tritium value	Waste	Waste	Fuel
Carbon in system	Yes	Depends upon option	No
Redox control	Ce^{+2}/Ce^{+3}, other	U^{+3}/U^{+4}	Ce^{+2}/Ce^{+3}, Be, other

The choice of salt depends upon neutronic and thermal-hydraulic considerations. Most proposed salts contain lithium because of its ability to lower the melting points of these salts to a few hundred degrees C. Flibe (Li_2BeF_4) has the best overall properties and is the choice in most proposed designs of FHRs and MSRs. For the FHR and MSR, one wants low neutron absorption cross sections with minimum tritium production. If a lithium salt is used, isotopically-separated ^{7}Li must be used to minimize neutron absorption and tritium production. There are other salt choices for the FHR and MSR with all of the options involve complex tradeoffs. For fusion one wants to maximize tritium production (the fuel) and thus ^{6}Li is required to maximize tritium production. For fusion systems, flibe is clearly the preferred salt coolant because it maximizes tritium production.

Under neutron irradiation these salts generate tritium by multiple pathways.

$$^{6}\mathrm{LiF} + n \rightarrow {}^{4}_{2}\mathrm{He} + {}^{3}_{1}\mathrm{HF} \quad (1)$$

$$^{7}\mathrm{LiF} + n \rightarrow {}^{4}_{2}\mathrm{He} + {}^{3}_{1}\mathrm{HF} + n' \quad (2)$$

$${}^{19}_{9}\mathrm{F} + n \rightarrow {}^{17}_{8}\mathrm{O} + {}^{3}_{1}\mathrm{H} \quad (3)$$

$${}^{9}_{4}\mathrm{BeF_2} + n \rightarrow {}^{4}_{2}\mathrm{He} + {}^{6}_{2}\mathrm{He} + 2\mathrm{F} \quad (4)$$

$${}^{6}_{2}\mathrm{He} \rightarrow {}^{6}_{3}\mathrm{Li} + e^{+} + \overline{\nu}_e \quad \left(t_{1/2} = 0.8\mathrm{sec}\right) \quad (5)$$

Lithium-7 has a very small neutron cross section and ^{6}Li has a large neutron cross section that maximizes tritium generation rates. With ^{7}Li salts, the residual ^{6}Li will partly burn out but will not go to zero if the salt also contains beryllium. Neutron reactions with beryllium will generate ^{6}Li that is converted into tritium.

The nuclear reactions have important chemical implications. With appropriate materials of construction, clean salts have extremely low corrosion rates. This was demonstrated in the Molten Salt Reactor Experiment, an 8-MWt test reactor built and operated in the late 1960s. The secondary loop used flibe with very low corrosion rates. However, in a reactor LiF is converted to ^{3}HF—hydrogen fluoride. Hydrogen fluoride is corrosive. Corrosion in

salt-cooled reactors (fission or fusion) is directly tied to the production of tritium. *Tritium control and corrosion control can't be separated.*

If ^3HF is allowed to corrode metals of construction, tritium in its molecular form will be released. To avoid corrosion redox control agents can be added to the coolant that result in converting ^3HF to $^3H_2$. The redox potential determines the relative amounts ^3HF versus $^3H_2$. While the ^3HF can't escape the system, the $^3H_2$ diffuses through hot metals such as heat exchangers to the environment. Maintenance of long term system integrity by assuring low corrosion rates implies converting ^3HF to $^3H_2$, but $^3H_2$ can escape the system requiring methods for removal of $^3H_2$ and methods for slowing escape of $^3H_2$ from the system. A partial pressure of $^1H_2$ in the cover gas will lead to isotopic exchange with ^3HF, producing ^3H-^1H, thus affecting the tritium transport rates. At the same time, introduction of H_2 in the cover gas may also shift the redox potential of the salt, thus affecting corrosion control.[14, 15, 16]

Tritium generation rates depend not only upon the salt selection and whether the reactor is a FHR, MSR, or fusion machine but also on the specific design features. This is most evident in the design of FHRs where there are large variations in the fraction of the core that is salt with a significantly higher salt fraction in a pebble-bed reactor than one with a FIRM core design. For one design of pebble-bed FHR, it was estimated[17] that ~0.03% of the tritium produced could be allowed to escape in order to stay below the tritium emission rates of the current fleet[18] of pressurized water reactors (2.2 Ci/GWe/d), or 810 Ci/GWe/y.

Stempien has built a model[19] for FHRs that accounts for tritium production, corrosion, transport that can predict behavior and calculate the impact of different methods to limit tritium loses or capture tritium. The model has been validated with the limited experimental data that is available. More experimental data is required to validate results. The model features are shown in Figure 5.

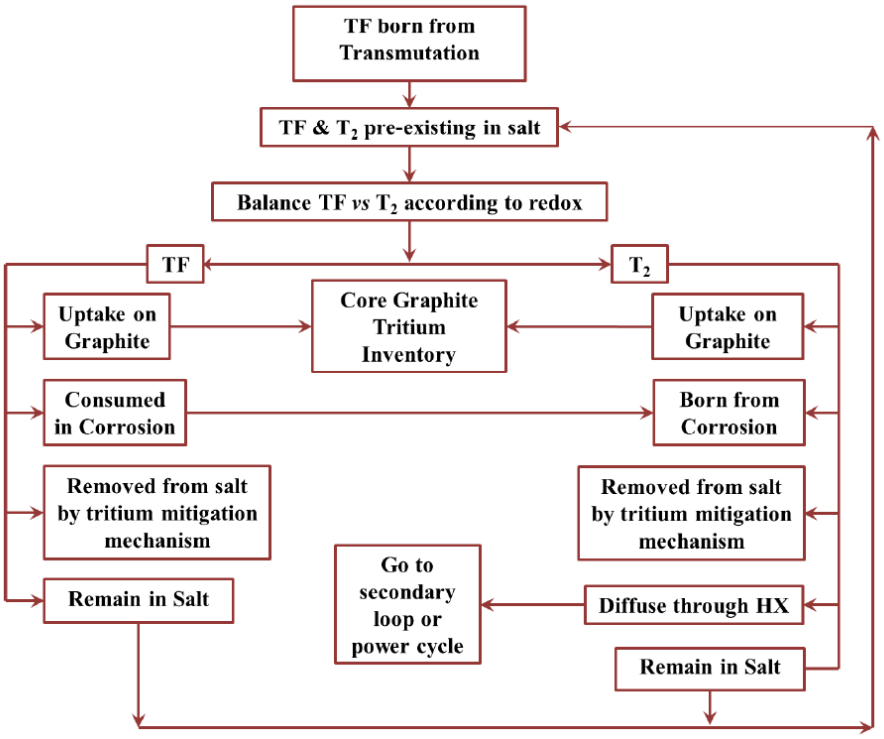

Figure 5. TRIDENT: Tritium Diffusion EvolutioN and Transport

TRIDENT has been used to model an FHR with a carbon bed for tritium removal as $^3H_2$. The model results are shown for one case in Table 4. In this case the carbon bed is designed to enable sufficient tritium removal to limit tritium releases to acceptable levels.

Table 4. TRIDENT Output for FHR with Tritium Carbon Absorber Bed

Temperatures*	
Coolant Freezing	459°C (FliBe)
Operating Core Outlet	700°C
ATWS	<800°C
Coolant Boiling	1400°C (FliBe)
Pressures (primary loop) *TRIDENT Simulation	
p_{T2} Unmitigated	3.3-20 Pa
p_{T2} with Graphite Capture	**0.03-0.08 Pa** (Peak release 7.5 Ci/GW/d)
p_{TF} Unmitigated	0.03-0.075 Pa
p_{TF} with Graphite Capture	**0.0027-0.0045 Pa** (Peak release 7.5 Ci/GW/d)

There are several observations from such modeling. The allowable tritium gas pressure in the primary system with metallic heat exchangers is on the order of magnitude of 0.05 Pa. If the tritium gas pressure is greater than this, the concentration gradients of tritium through hot heat exchangers may allow tritium to escape during operations in excess of releases from LWRs. The same limit would apply to a MSR or a fusion machine. The difference with a fusion machine is that the starting concentration of tritium in the salt is three orders of magnitude larger than in an FHR, and tritium is recovered from the salt for subsequent use as fuel. The calculated partial pressure of ^3HF is also given but HF can't diffuse through hot metal. Graphite absorbs both $^3H_2$ and ^3HF.

The requirements in Table 4 for allowable levels of tritium in the salt are strongly dependent upon permeation rates of tritium through hot heat exchanges to the environment. If high-performance coatings are incorporated into the heat exchangers to slow migration of tritium, much higher concentrations of tritium can remain in the salt. Consequently there is a tradeoff between high-efficiency removal of tritium from the salt and highly effective barriers to slow tritium transport through the heat exchangers. The CAS has a major effort to measure and develop better tritium barriers. The INL STAR facility for the fusion program has also had a major effort in measuring permeation rates through various materials.

4. TRITIUM CONTROL AND CARBON

The largest fraction of the workshop was devoted to carbon and tritium in salt-cooled systems. Carbon absorbs tritium and is chemically compatible with high-temperature salts. It is important in these systems in three different contexts.

4.1. Carbon in Reactor Cores

The FHR fuel is made of carbon and thermal spectrum MSRs use bare graphite. Tritium holdup in the carbon is significant. In the pebble-bed FHR with on-line refueling, there is the potential to use the pebbles as the main tritium capture system. In a pebble-bed reactor the pebbles circulate through the core typically once a month. There is the option of heating pebbles as they are circulated out of the core to remove the tritium before recirculating the pebbles back to the reactor core. This is not viable for other fuel forms because the carbon capability to pick up tritium will saturate due to the longer times between refuelings.

Tritium on carbon components is important in one other context. If carbon components in the nuclear system pick up a large inventory of tritium over time, this inventory could be partly released in a reactor over-temperature transient event. The equilibrium quantities of tritium in the core depend upon the average tritium levels in the coolant. The reactor core tritium inventories can be minimized by maintaining low levels of tritium in the coolant because there is an equilibrium between these tritium reservoirs.

Tritium uptake on carbon depends not only upon the carbon form but also on radiation damage in the graphite and potentially radiation flux levels. The MIT activities are primarily associated with experimental activities to understand these behaviors in 700°C salt in the MIT reactor. UW activities are investigating absorption onto previously-irradiated fuel matrix graphite.

4.2. Out-of-Core Tritium Removal with Carbon

Initial modeling indicated that a carbon bed out-of-core should be able to remove tritium from the clean salt in FHRs. The TRIDENT modeling used the limited tritium absorption data available on nuclear grade graphite for this analysis. However, non-nuclear-grade carbon forms have surface areas per unit of mass up to 1000 times larger than nuclear grade graphite—and potentially hydrogen sorption capacities a 1000 times larger. Nuclear grade graphite undergoes very high-temperature processing to produce a graphite with dimensional stability under high neutron radiation—a requirement that does not exist for any carbon absorber outside the reactor core. An out-of-core carbon bed can have the carbon and the bed optimized for tritium removal. This suggests the potential for relatively small carbon beds to efficiently remove tritium to very low concentrations. MIT (Lam) is investigating this option.

In the design of the MSRE great efforts were undertaken to avoid uptake of xenon and krypton in the graphite moderator because they are neutron absorbers. However, some uptake of xenon and krypton was observed. It is believed that these inert gases diffused into void spaces that do not fill with salt because of liquid surface tension. It is unknown at this time whether efficient carbon absorbers could be developed for MSRs to simultaneously remove tritium, krypton, and xenon—or potentially removal of the fission-product noble metals from the salt. Fission product noble metals that are generated insitu migrate to liquid-gas and liquid-solid surfaces because of the very low solubility in salts. Metals can be plated onto carbon. What is unknown is whether a practical system can be developed and what type of surface treatment of carbon may be required for noble metal fission products. That

will require good methods to generate noble metals in salts or a MSR test loop to test different carbon absorbers for noble metals.

4.3. Carbon and Corrosion

The presence of carbon in a salt system alters corrosion rates. Experiments at MIT and Wisconsin are underway to understand the various mechanisms and testing different materials with and without carbon in the system.

5. OTHER OPTIONS FOR TRITIUM REMOVAL FROM SALT

5.1. Gas Sparging

Tritium can be removed from high-temperature salt using gas sparging where an inert gas such as helium or argon is mixed with the liquid salt and tritium in different forms preferentially transfers to the gas phase and then to the off-gas system. The Chinese Academy of Science (CAS) has an ongoing experimental program to develop such a system is initially using water analogs to be followed by testing in salt systems. The CAS plans to complete by 2020 a 10 MWt FHR and a 2 MWt MSR. In a MSR, the off-gas system must remove tritium but also remove much larger quantities of fission product xenon and krypton. This creates an incentive for a common system to remove all volatile gases—a capability of a gas sparging system.

New Mexico is initiating a program using ultrasonic gas sparging. Ultrasonics can potentially create smaller gas bubbles with higher surface area and thus more efficient mass transfer of tritium to the inert gas phase. While ultrasonic gas dispersion has been used in a variety of systems, it has not been applied to very high temperature systems.

There is the option of using a spray tower or equivalent where the salt is in droplets or flowing over a high-surface area media and the purge gas is the continuous media—reverse of gas sparging. It is an option originally examined by LLNL for fusion machines. The complication with this strategy is the larger size of equipment when the purge gas is the continuous phase.

This option may be attractive for one class of advanced MSR designs that is being developed by Hatch of Canada where the liquid salt flows downward through the reactor core through constrictions to critically-safe passively-cooled dump tanks under the reactor core—a type of spray tower. The liquid salt is pumped from these tanks through the heat exchangers back to the reactor core. Any failure, including loss-of-power, results in the salt draining to a safe configuration. This configuration may allow tritium, xenon, and krypton removal options not available with other designs. The salt can be dispersed as droplets or flow over a metal mesh—both providing a high surface area for the liquid salt to allow the dissolved gases to diffuse over short distances from the liquid salt phase into the gas phase.

If a high-surface-area metal mesh is used, one would expect the noble metals to plate out on the surface of the mesh—addressing another salt cleanup challenge of MSRs. Alternatively the metal mesh could be salt pool at the bottom of the system for criticality control and noble metal plate out.

5.2. Permeators

Tritium can be removed by metallic permeators. Permeators are tubes designed for high rates of hydrogen transfer through the tube where salt would be on one side of the tube and a vacuum or hydrogen getter would be on the other side of the tube to provide a large hydrogen gradient to maximize hydrogen transport. Permeators are used in the laboratory and some process operations for tritium separation and isotopic separation of different hydrogen isotopes. Investigations are underway at Ohio State for tritium separation in salt-cooled systems using permeators.

5.3. Double-Wall Heat Exchangers

Double wall heat exchangers are used in the chemical industry where there are two fluids where violent reactions would occur if there was a tube failure. They have also been developed for sodium-cooled fast reactors for sodium water heat exchangers. Such heat exchangers can be used to block tritium transport by three mechanisms:

(1) vacuum between the tubes to capture tritium, (2) solid hydrogen getter between the tubes to sorb the tritium or (3) flowing fluid such as lithium that acts as a getter. The disadvantages of double wall heat exchangers is cost and added temperature drop across the heat exchanger. Oak Ridge National Laboratory, The Ohio State University and the University of New Mexico are investigating this option for liquid salt systems.

6. TRITIUM CONTROL EXPERIENCE

There is a massive experience base in tritium separations and control from national security, heavy water reactor, high-temperature gas-cooled reactor, fusion, and research programs. This experience provides the basis for research going forward and provides many of the required research tools. It also provides proven methods to handle tritium once captured for disposal as a waste or recycle as a fuel (fusion).

In the United States there are three organizations with much of this experience. Savannah River National Laboratory has the national security tritium handling facilities. Pacific Northwest National Laboratory has been responsible for targets to produce tritium in the Watts Bar Nuclear Power Plant. Last, Idaho National Laboratory[20] has facilities that conduct research on tritium for the fusion community. Oak Ridge National Laboratory has the expertise in carbon and done much of the work on hydrogen absorption on carbon. MIT laboratories using the MIT reactor are investigating tritium behavior in 700°C salt under neutron irradiation with the University of Wisconsin conducting similar work in a laboratory environment (no irradiation).

7. CONCLUSIONS

A confluence of events in three power technologies (FHR, MSR, and Fusion) in the last several years has created the need for control and removal of tritium from high-temperature coolant salts. While there is massive experience in tritium capture and control under many different environments, that experience has not been in 700°C salt where the tritium is in the forms of $^{3}H_2$ and ^{3}HF at partial pressures of fractions of a Pa. A first workshop has been held as a starting point for a larger international cooperative effort to develop the required technologies for tritium control. A basic understanding of requirements and options has been developed but there is a large need for experimental data to develop models and engineered systems. The appendices include the agenda, the list of participants, and the presentations.

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Appendix A:

Workshop Agenda

Final Agenda

Workshop on Tritium Control and Capture in Salt-Cooled Fission and Fusion Reactors: Experiments, Models and Benchmarking

Salt Lake City; October 27-28, 2015
DoubleTree by Hilton Hotel Salt Lake City Airport, Salt Creek Ballroom

The objectives of the workshop are to bring together researchers involved in experiments, modeling and benchmarking for tritium control at ~700°C in liquid salts and related system to (1) exchange information and enable future exchange of information, (2) initiate an effort for benchmarking of experiments and models, and (3) encourage cooperation between different groups working on the same challenges. The workshop includes tritium sorption behavior in carbon because (1) fluoride-salt-cooled high-temperature reactors (FHRs) and some types of molten-salt reactors (MSRs) have carbon matrix fuels and/or carbon moderators and (2) several carbon forms are leading candidates to remove tritium from high-temperature salts.

The workshop goals are information exchange between the participants to enable future cooperation. The agenda will be modified based input from participants. Abstracts, papers, and presentations will be posted on the website (*http://tcw15.mit.edu*).

Tuesday: October 27

7:30 – 8:30 Continental Breakfast – *Stillwater Poolside Atrium*

8:30 – 10:00 *Session 1. Welcome, Introductions, and Workshop Goals (MIT, CAS, Wisconsin)*

Welcome

Introductions

Workshop Goals

Brief summaries of different programs (U.S., China, Europe, etc.)

Charles Forsberg (MIT NSE)

Xiaodong Sun (OSU)

10:00 – 10:30 Break

10:30 – 12:00 *Session 2. Environment for Tritium Control*

This session will include descriptions of (1) the different tritium-generating energy systems that use high temperature (~700°C) salts, (2) the environments in which the tritium is generated, and

(3) constraints on tritium removal. The goal is to define the challenges—common challenges and unique challenges with tritium capture and control associated with each technology.

Fluoride-salt-cooled High-temperature Reactors (Tritium environment: clean salt, carbon in system, possibility of small quantities of fission products, tritium is waste or useful product, redox control strategy?)

Fusion ARC concept (Tritium environment: clean salt, no carbon in system, high-efficiency recovery of tritium because tritium is the fuel, very high tritium levels, redox control?)

Molten salt reactor (MSR) (Tritium environment: salt with fission products and actinides, carbon may or may not be in the system, redox controlled by U^{+3}/U^{+4}, tritium is waste or useful product)

This session will also include discussions on tritium generation rates, required recovery rates, and various limits (Goals) that differ for various technologies.

Charles Forsberg (MIT NSE) "Fluoride-salt-cooled High-Temperature Reactors"

Brandon Sorbom (MIT PSFC) "The Fusion ARC Concept"

Cristian Contescu and Tim Burchell (ORNL) "Hydrogen – Carbon Interactions"

Tim Burchell and Cristian Contescu (ORNL) "AGR Fuel Compact Development Program"

12:00 – 1:30 Lunch – *Stillwater Poolside Atrium*

Wei Liu (CAS) "TMSR Project in China"

Michael Laufer (UCB) "Tritium and Chemistry Management for the Mark-1 PB-FHR"

1:30 – 3:00 *Session 3. Tritium handling experience*

Greg Staack (SRNL) "An Overview of SRNL Tritium Activities"

Masashi Shimada (INL) "Overview of Tritium and molten salt FLiBe research at Safety and Tritium Applied Research (STAR) facility"

David Carpenter (MIT NRL) "Experience with Tritium Evolution During Irradiation of MSRE Flibe in the MITR"

3:00 – 3:30 Break

3:30 – 4:30 *Session 4. Characteristics of salt, corrosion, and redox on tritium control challenge*

Chemistry determines tritium chemical form ($^3H_2$ or ^3HF or ^3HCl) and thus transport of tritium to the environment, tritium holdup in the system, and tritium control options. Tritium control tightly coupled to corrosion control in most but not all cases. Options for redox control and implications of those options on tritium control and capture to be discussed. (Example, if hydrogen used as part of redox control or salt cleanup, may have significant to remove with tritium)

Thomas Chrobak (UW) "FLiBe Electrochemistry and Materials Corrosion Research at UW-Madison"

Huali Wu (UW) "Effect of hydrogen on tritium control"

Wednesday October 28

7:30 – 8:30 Continental Breakfast – *Stillwater Poolside Atrium*

8:30 – 9:30 *Session 5. Tritium and Carbon*

Carbon is a large sink for tritium and a potential capture method for tritium in multiple chemical forms. What is known and not known

Huali Wu (UW) "Experimental work on tritium transport analysis in Flibe-Graphite system"

Stephen Lam (MIT NSE) "Tritium Control Using Carbon Outside the Core"

9:30 – 10:00 Break

10:00 – 12:00 *Session 6. Tritium Capture*

Systems for tritium capture at high temperatures in salts from salts, the gas space above, and related environments.

Paul Humrickhouse (INL) "Tritium Permeation Control and Extraction – Perspectives from Fusion System Studies"

Wei Liu (CAS) "Tritium-Control Technologies for TMSR System in CAS"

Floren Rubio (UNM) "Research on Techniques for Tritium Sequestration and Removal at UNM"

12:00 – 1:00 Lunch – *Stillwater Poolside Atrium*

1:00 – 2:30 *Session 7. System models of tritium transport*

John Stempien (INL) "Tritium Transport and Corrosion Modeling in the Fluoride Salt-Cooled High-Temperature Reactor"

David Senor (PNNL) "Irradiation Testing in Support of the Tritium Production Enterprise"

2:30 – 3:00 Break

3:00 – 5:00 *Session 8. Closeout and path forward*

David Carpenter (MIT NRL) "Planned FHR IRP-2 Tritium Experiments"

Bryan Wallace (UNM) "Investigation of Tritium Control and Release Mitigation Options in Double-Wall Twisted-Tube Heat Exchangers (DT-HXRs)"

Charles Forsberg (MIT NSE)

Thursday October 29

7:30 – 8:30 Continental Breakfast – *Stillwater Poolside Atrium*

8:30 – 10:00 *Side Meetings as Appropriate*

10:00 – 10:30 Break

10:30 – 12:00 *Side Meetings as Appropriate*

Appendix B:

Workshop Participants

List of Participants

Name	Organization
Tim Flaspoehler	Georgia Tech
Carl Stoots	Idaho National Laboratory
John Stempien	Idaho National Laboratory
Masashi Shimada	Idaho National Laboratory
Paul Humrickhouse	Idaho National Laboratory
David Carpenter	Massachusetts Institute of Technology, Nuclear Reactor Laboratory
Guiqiu Zheng	Massachusetts Institute of Technology, Nuclear Reactor Laboratory
Charles Forsberg	Massachusetts Institute of Technology, Nuclear Science & Engineering
Stephen Lam	Massachusetts Institute of Technology, Nuclear Science & Engineering
Brandon Sorbom	Massachusetts Institute of Technology, Plasma Science & Fusion Center
Cristian Contescu	Oak Ridge National Laboratory
Nicholas Brown	Oak Ridge National Laboratory
Tim Burchell	Oak Ridge National Laboratory
Xiaodong Sun	Ohio State University
David Senor	Pacific Northwest National Laboratory
Wei Liu	Shanghai Institute of Applied Physics
Greg Staack	Savannah River National Laboratory
Michael Laufer	University of California Berkeley
Bryan Wallace	University of New Mexico
Floren Rubio	University of New Mexico
Guoping Cao	University of Wisconsin Madison
Huali Wu	University of Wisconsin Madison
Jee-Hoon Kim	University of Wisconsin Madison
Raluca Scarlat	University of Wisconsin Madison
Thomas Chrobak	University of Wisconsin Madison
Nisarg Patel	University of Wisconsin Madison
Yasir Arafat	Westinghouse

Appendix C:

Workshop Presentations

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218 David Senor (PNNL) "Irradiation Testing in Support of the Tritium Production Enterprise"

234 David Carpenter (MIT NRL) "Planned FHR IRP-2 Tritium Experiments at MIT NRL"

239 Bryan Wallace (UNM) "Investigation of Tritium Control and Release Mitigation Options in Double-Wall Twisted-Tube Heat Exchangers (DT-HXRs)"

TRITIUM WORKSHOP GOALS

Charles Forsberg

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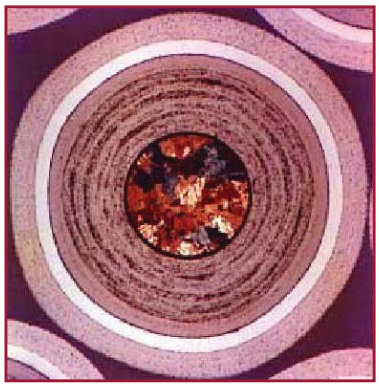
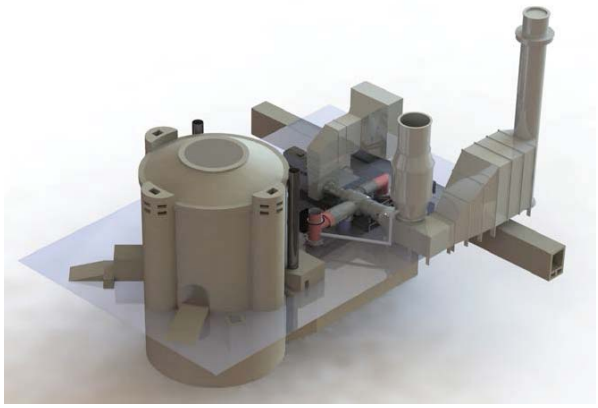
Workshop on Tritium Control and Capture in Salt-Cooled Fission and Fusion Reactors: Experiments, Models, and Benchmarking
Salt Lake City
October 27, 2015

Three Technologies Depending Upon Liquid Salt Coolants & Control of Tritium

Fluoride-salt-cooled High Temperature Reactors: Solid Fuel & Clean Salt

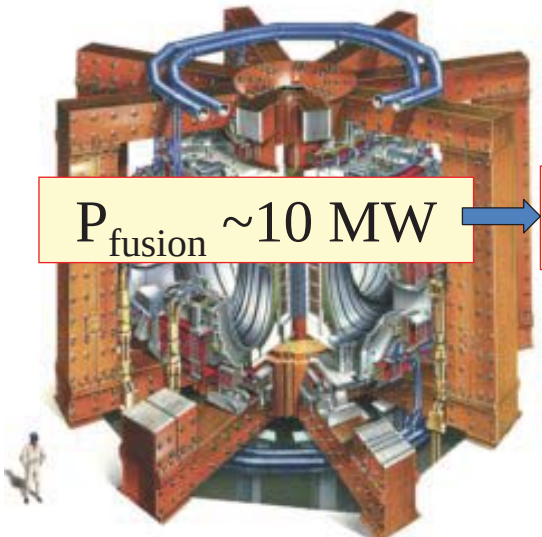




Enabled by Advances In Gas-cooled High-Graphite-Matrix Coated Particle Fuel

High-Magnetic Field Fusion

Copper, B = 3.5 T

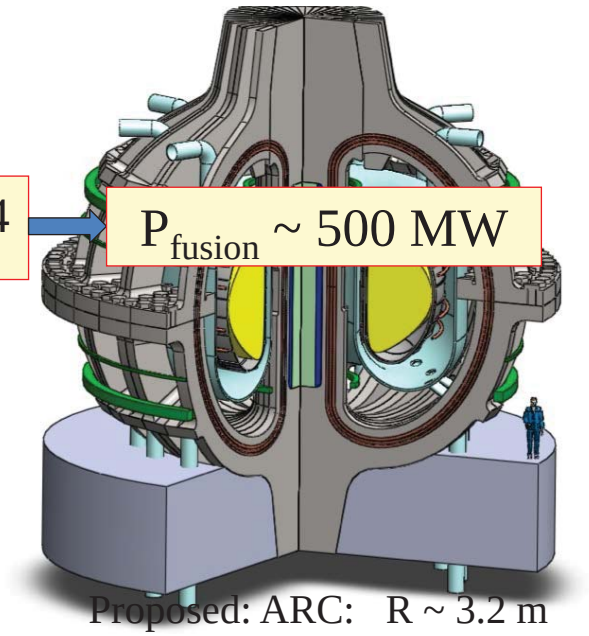


P_{fusion} ~10 MW

JET: R ~ 3 m
Operated in United Kingdom

x B^4

REBCO superconductor, B = 9.2 T



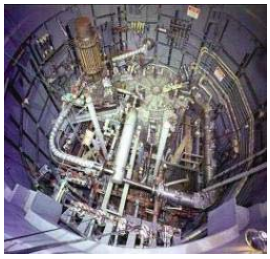
P_{fusion} ~ 500 MW

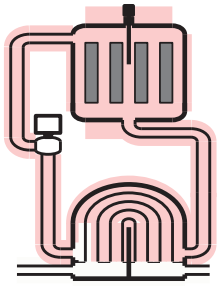
Proposed: ARC: R ~ 3.2 m

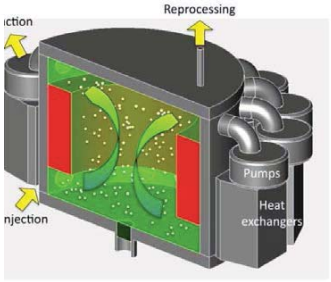
Enabled by REBCO Superconductors that Enable Doubling Magnetic Fields

Molten Salt Reactor

Fuel Dissolved in Salt







Many Options

Enabled By Multiple Technologies and Interest In Alternative Breeder Reactors and Fuel Cycles

Massachusetts Institute of Technology

Common and Different Salt Challenges for FHR, MSR, and Fusion

Property	FHR	MSR	Fusion
Salt	Fluoride	Fluoride or Chloride (fast spectrum only)	Fluoride
Impurities	Corrosion and possible fission product impurities	High concentrations of fission products and actinides	Corrosion impurities
Use lithium salts	Optional	Depends upon goals	Required
Tritium production	Small (^{7}Li in Coolant)	Small (^{7}Li in Coolant)	Very High (^{6}Li in Coolant)
Tritium value	Waste	Waste	Fuel
Carbon in system	Yes	Depends upon option	No
Redox control	Ce^{+2}/Ce^{+3}, other	U^{+3}/U^{+4}	Ce^{+2}/Ce^{+3}, Be, other

Massachusetts Institute of Technology

Common Technology Challenges for FHR, Fusion, and MSR

Common Challenges

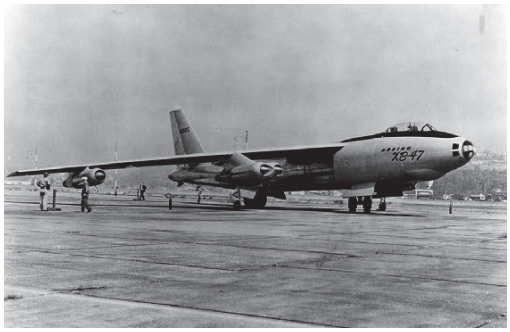
- **Power Cycles**
- Themohydraulics
- Mechanical Equipment
- Instrumentation
- Lithium Isotopic Separation (^{6}Li or ^{7}Li)
- **Tritium Generation**
- **Corrosion Control**
- **Tritium Control**

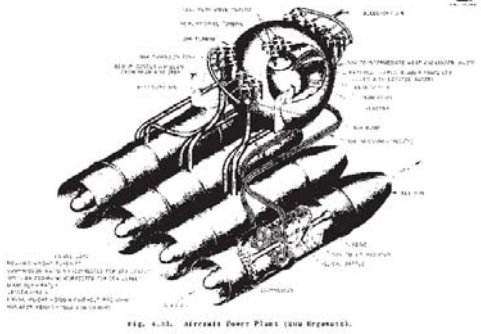
Common Technology Challenges for FHR, Fusion, and MSR

Unique Capability of All Salt-Cooled Fission and Fusion Systems to Couple to Air or Helium Brayton Cycles

Salt Coolants Were Developed for the Aircraft Nuclear Propulsion Program

Salt Coolants Designed to Couple Reactors to Jet Engines

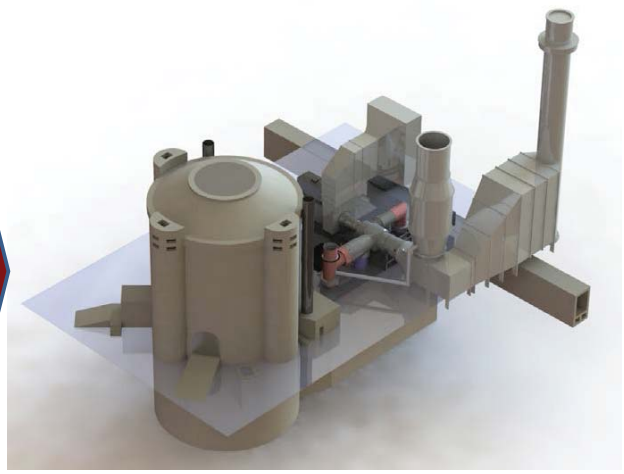




It Has Taken 50 Years for Utility Gas Turbine Technology to Mature Sufficiently to Enable Coupling with an FHR

Coupling Reactors to Gas Turbines is Transformational

Advances In Natural Gas Combined Cycles Enable Coupling Reactors to Gas Turbines



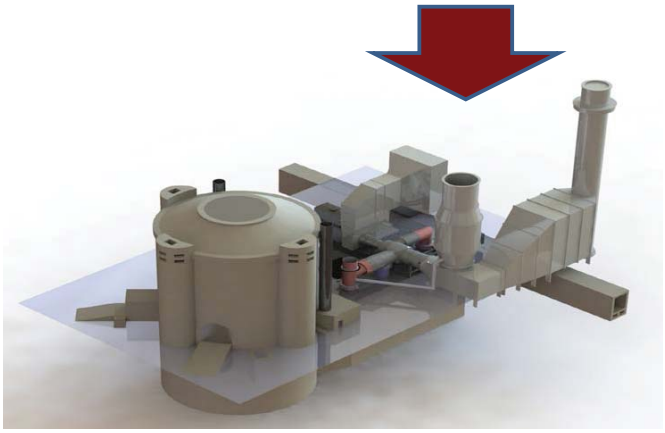
Gas-Turbine Technology Not Viable 15 Years Ago

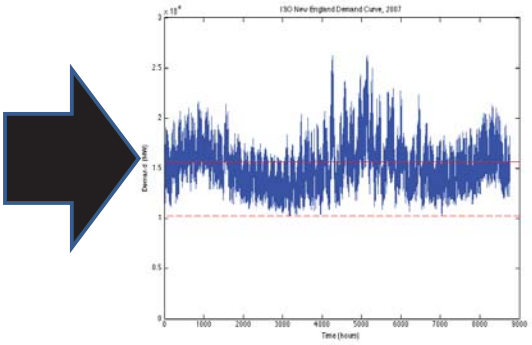
Only Salt-Cooled Reactors (Fission & Fusion) Couple Efficiently to Nuclear Air-Brayton Combined Cycles (NACC)

Commercial gas-turbine exit air compressor temperatures are between 400 and 500°C; thus, must deliver all heat above these temperatures

Coupling Salt Fission and Fusion Reactors Using NACC Enables Base-Load nuclear with Variable Electricity to the Grid

Stored Heat, Natural Gas, or Hydrogen





Base-Load Reactor **Gas Turbine** **Variable Electricity**

Boosts Revenue by 50 to 100% Relative to Base-load Nuclear Power Plants

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Power Cycle Choices May Impact Tritium Control Strategies

- Can trap tritium in some power cycles because cold side of power cycle prevents tritium releases
  - Supercritical carbon dioxide
  - Helium Bryaton cycles
- Tritium major challenge if enters some power cycles
  - Steam cycles
  - Air-Brayton power cycles

Common Technology Challenges for FHR, Fusion, and MSR

The Coupled Challenges of Tritium Generation, Corrosion and Tritium Control

Tritium Generation, Corrosion and Control Are Coupled

Neutrons + $^{7}Li/^{6}Li/Be \rightarrow {}^{3}HF$

^{3}HF + Metal → Corrosion + $^{3}H_2$

Can't Separate Tritium Generation, Corrosion and Control

Workshop Goals

Workshop Goals

- Exchange information and enable future exchange of information
- Initiate an effort for benchmarking of experiments and models
- Encourage cooperation between different groups working on the same challenges

Common Challenges for Multiple Power Systems

Tritium Management in FHRs

Ongoing and Planned Activities in Integrated Research Project Led by Georgia Tech

Workshop on Tritium Control and Capture in Salt-Cooled Fission and Fusion Reactors: Experiments, Models and Benchmarking

Timothy Flaspoehler and Bojan Petrovic
Georgia Tech

Xiao Wu, Sheng Zhang, Richard Christensen, and Xiaodong Sun
The Ohio State University

Work Supported by DOE Nuclear Energy University Programs

NEUP Nuclear Energy University Program
U.S. Department of Energy

Salt Lake City, UT
October 27-28, 2015

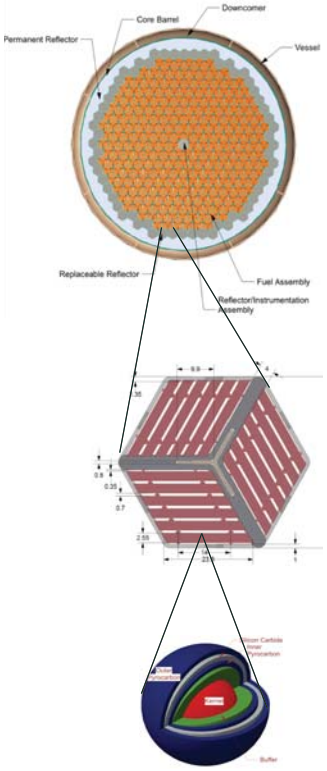
Outline

- Tritium Source Generation
  - Planned for Georgia Tech
- Design, Testing, Demonstration, and Modeling of Heat Exchangers for FHRs
  - Ongoing at Ohio State University
  - Heat Exchanger Design
  - Tritium Permeation Barrier Coating
- Tritium Control/Mitigation Strategy for FHRs
  - Redox Control Facility
  - Tritium Removal Facility
  - Planned Experiments

Tritium Source Generation

- INTRODUCTION
- Georgia Tech: Timothy Flaspoehler
  - Advisor (Bojan Petrovic)
- Use neutron transport to calculate accurate tritium source in FHR
  - Full-core
  - Time-dependent
- Funding doesn't start till 2nd fiscal year

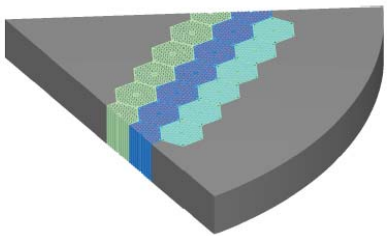
Pathway	MT #	Scale6.1
Ternary Fission	18 (x%yield)	YES
^{6}Li (n, α) ^{3}H or	107	NO
^{6}Li (n, t) ^{4}He	105	YES
^{7}Li (n, nα) ^{3}H or	22 / 105	NO / NO
^{7}Li (n, nt) ^{4}He or	33	NO
^{7}Li (n, Xt)		*YES
^{10}B (n, 2α) ^{3}H or	108	NO
^{10}B (n, t2α)	113	YES
^{10}B (n, α) ^{7}Li (n, nα) ^{3}H	107 & 22	YES & NO

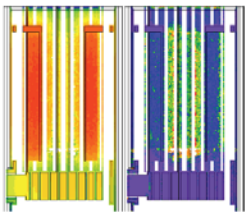


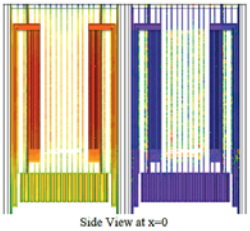
Tritium Source Generation

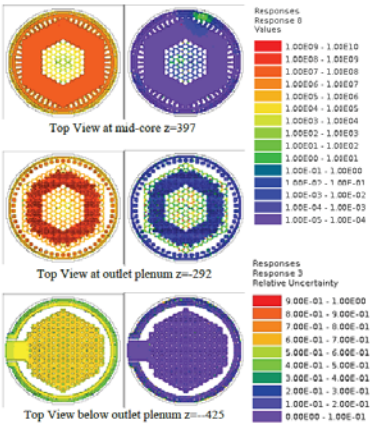
PREVIOUS WORK

- BACKGROUND: VHTR / NGNP goal to provide heat source for industrial applications
  - Also without NRC licensing secondary side
  - Tritium leakage to secondary must be below EPA limits
- RESULTS: Calculated different pathways in full-core model
  - Used MAVRIC shielding sequence
  - Globally converged MC tallies in reflector









Tritium production from ^{4}He impurities in coolant

Georgia Tech

Tritium Source Generation

PREVIOUS WORK

- RESULTS: Possible underestimate of tritium source in VHTR from impurities in graphite reflector

Table 3.1.1 Comparison of tritium generation rates in VHTR estimated in [3.1.1] and [3.1.2]

	Values based Ref. on [3.1.1]		New estimate in Ref. [3.1.2]		
Pathway	**Activity (Bq/y)**	**Production (t/s)**	**Activity (Bq/y)**	**Production (t/s)**	**Ratio (C/A)**
Ternary Fission	1.03E+14 (62.0%)	1.83E+15	1.03E+14 (29.8%)	1.83E+15	1.00
From ^{3}He	2.98E+13 (18.0%)	5.30E+14	1.43E+13 (4.1%)	2.53E+14	0.48
From ^{6}Li	2.32E+13 (14.0%)	4.12E+14	1.78E+14 (51.6%)	3.16E+15	7.67
Core Graphite	3.31E+12 (2.0%)	5.89E+13	5.45E+13 (15.8%)	9.68E+14	2.74
Core Matrix	1.66E+13 (10.0%)	2.94E+14			
Reflector	3.32E+12 (2.0%)	5.88E+13	1.23E+14 (35.8%)	2.19E+15	37.24
From ^{10}B	1.49E+13 (9.0%)	2.65E+14	5.00E+13 (14.5%)	8.89E+14	3.36
Control Rod	1.16E+13 (7.0%)	2.06E+14	4.35E+13 (12.6%)	7.74E+14	3.75
Absorber	1.66E+12 (1.0%)	2.94E+13	4.51E+12 (1.3%)	8.02E+13	2.72
Reflector	1.66E+12 (1.0%)	2.94E+13	2.00E+12 (.6%)	3.56E+13	1.21
Total	1.71E+14	3.03E+15	3.45E+14 (100.0%)	6.13E+15	2.02
Total (Bq/y/MWt)	2.84E+11		7.88+11		2.77

4

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Design, Testing, Demonstration, and Modeling of HXs for FHRs

- Design of Heat Exchangers (IHX, SHX, DHX, and NDHX) for AHTR, considering Tritium Management and Heat Transfer Effectiveness
  - Goal: To reduce tritium diffusion into the secondary (cold) side while maintaining heat transfer rate
- Double-wall Heat Exchangers
  - Fluted tube heat exchanger
  - Printed circuit heat exchanger
- Tritium Permeation Barrier
  - Located between the outer tube and the inner tube walls
  - Fluoride salt (FLiNaK/FLiBe)
  - Sweep gas
  - Tritium getter

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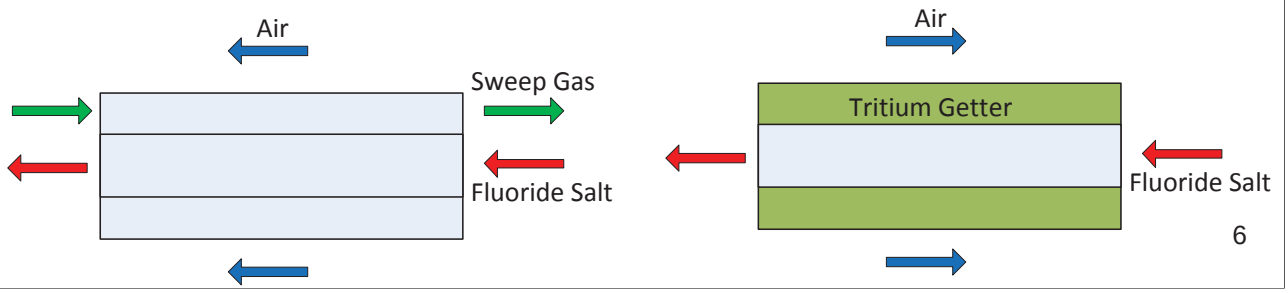
5

Heat Exchanger Design

- Ongoing Activity for NDHX in DRACS: Two Preliminary Designs being Considered
  - Option 1: Double-wall NDHX with sweep gas in the annulus
    - Inner tube: Allow tritium permeation
    - Outer tube: Inhibit tritium permeation (with surface treatment if necessary)
    - Sweep gas: Pressurized helium
    - Tritium: Trapped in the gap and taken away by sweep gas
  - Option 2: Double-wall NDHX with tritium getter in the annulus
    - Gap/annulus filled with a tritium getter (yttrium) to sequester tritium



Tritium Permeation Barrier Coating

Surface Treatment: Tritium Permeation Barrier

Barrier	Base Metal	PRF
Al_2O_3	SS316, MANET, TZM, Ni, Hastalloy-X	10 to >10,000
TiC, TiN, TiO_2	SS316, MANET, TZM, Ti	3 to >10,000
Cr_2O_3	SS316	10 to 100
Si	Steels	10
BN	304SS	100
N	Fe	10 to 20
Er_2O_3	Steels	40 to 700

	Al_2O_3	Cr_2O_3-SiO_2	ZrO_2	MSZAC	W
Thickness [μm]	0.03-1.4	50	50	50-100	10
PRF	100-10^4	292	50	3-4	300
References	Levchuk (2004); Yang (2011); Forcey (1991); Forcey (1989)	Nakamichi (2007) [13]	Nakamura (2010)	Nakamura (2010)	Moir (1984)

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Low Tritium Permeability Metal

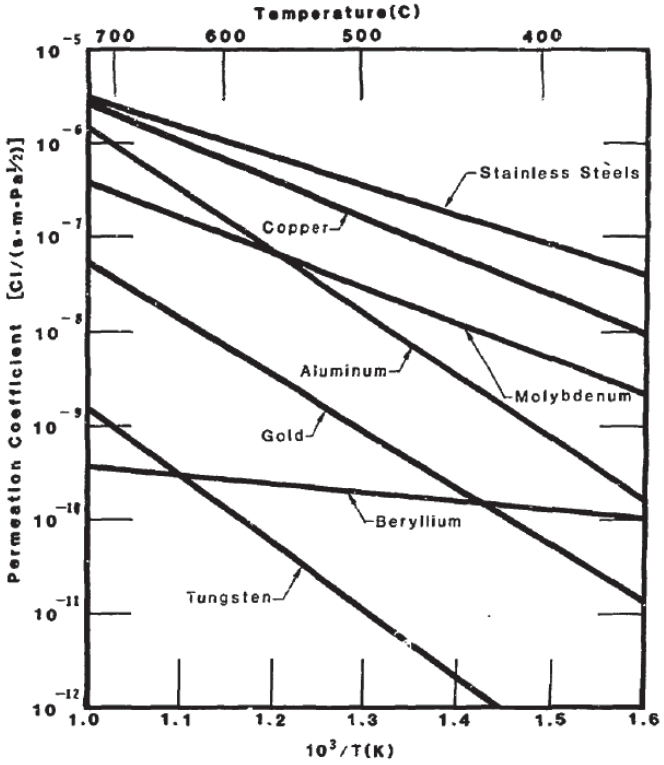

Figure 8.10-18
Permeation coefficient of tritium through metals.

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Tritium Permeation Barrier Coating (Cont'd)

- Tritium Permeation Reduction Factor (PRF) of candidate coatings

$$\text{PRF} = \frac{\text{Permeation flux without coating}}{\text{Permeation flux with coating}}$$

- Al_2O_3 Coating Methods
  - Hot-dip aluminazation
  - Chemical vapor deposition (CVD)
  - Sol-gel
- Potential Issues with Al_2O_3 Coating
  - Integrity is crucial to the surface coating
  - Cracks can lead to significant decrease in the PRF

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Tritium Control/Mitigation Strategy for FHRs

- Generation
  - Major form of tritium in the core: TF (corrosive)
- Redox Control
  - Beryllium metal is used to convert TF to T_2: Be + 2TF → T_2 + BeF_2
- Tritium Removal Facility
  - Goal: Removal rate similar to the production rate
  - Cross-flow plate-type T_2 removal facility
- Tritium Permeation Barrier
  - FLiNaK/FLiBe could be used as the barrier in intermediate heat exchanger (IHX)
  - Tritium permeation barrier used as the outer wall coating in necessary areas

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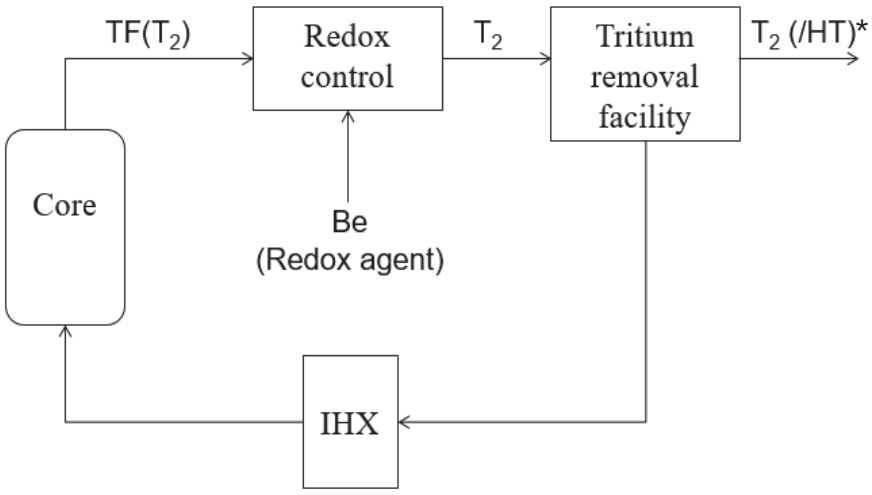
Schematic of Tritium Mitigation/ Control System for FHRs



* :1. HT exists if H_2 is used in the purging gas
2. Studies have shown that by adding H_2 in the purging gas, T_2 removal efficiency can be improved

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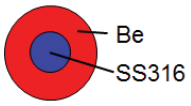
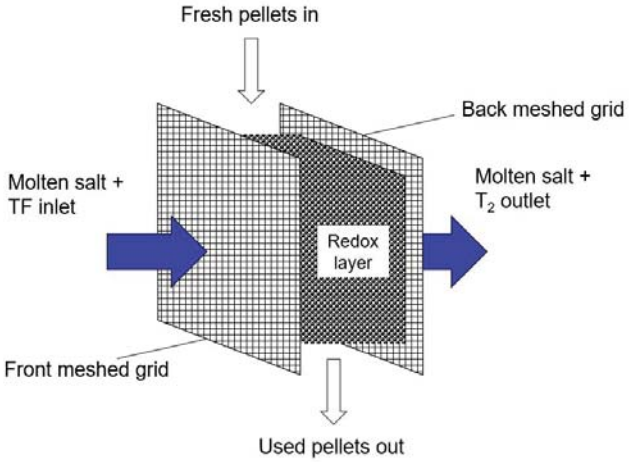
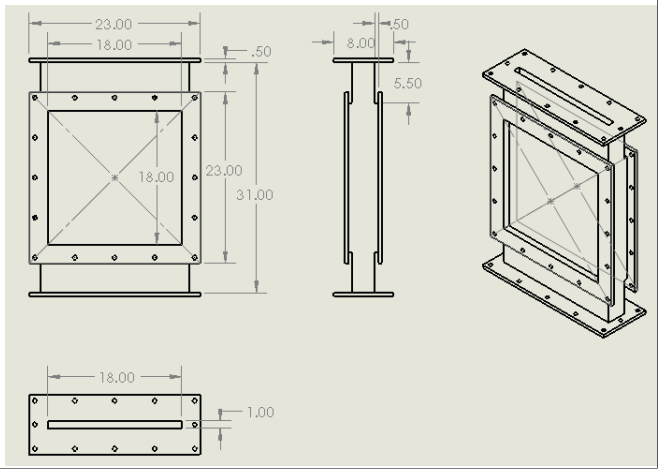
Redox Control Facility

- Easy Replenishment of Redox Pellets
- Modular Design
  - Located prior to the tritium removal module
- Pellet with SS316 Core
  - Beryllium pellets with a spherical SS316 core
  - To avoid used (smaller) pellets from being carried away by the salt with meshed grids







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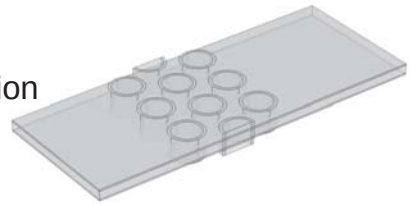
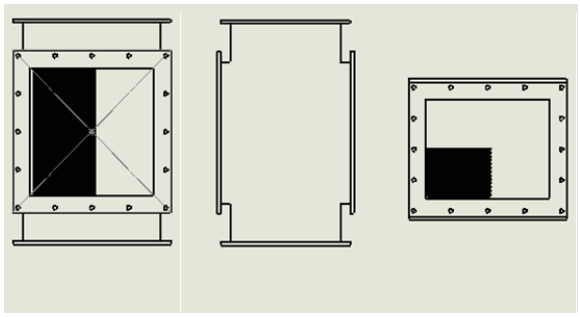
Tritium Removal Facility

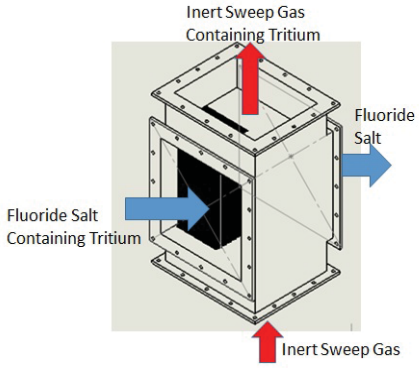
- Cross-flow Configuration
  - Purging gas flows in the tube bank
  - Molten salt flows in the perpendicular direction to the tube bank
  - Increase the salt flow turbulence level
- Modular Design
  - Located after the redox control facility
  - Flexibility for applications of different tritium removal rates



Cut view of a unit cell of the facility



Front view Side view Top view



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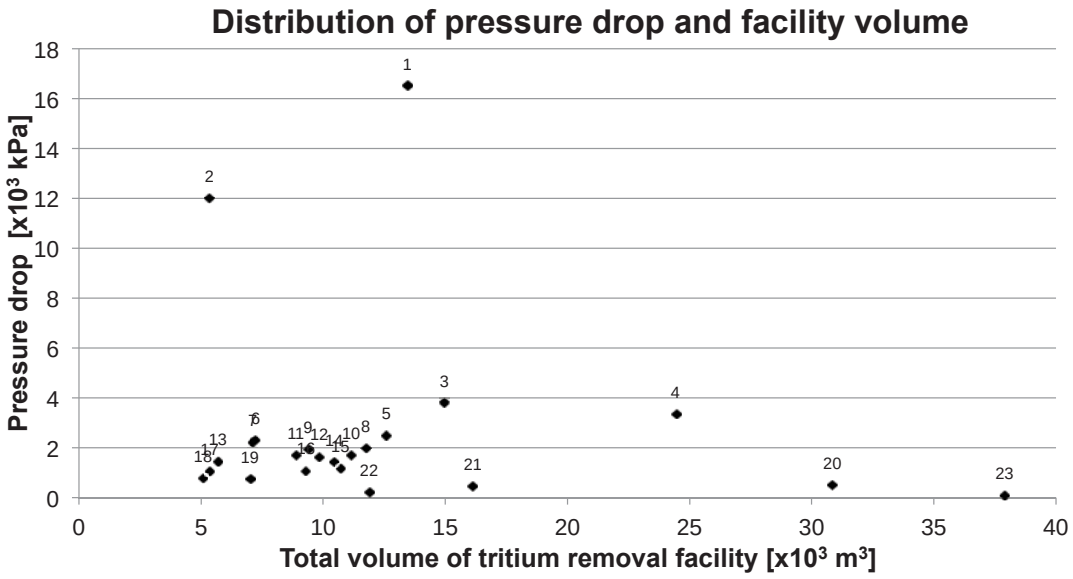
Results from COMSOL

- Comparison of Tritium Removal Facility Models
- Main Variables
  - Tube size
  - Tube pitch
  - Salt inlet flow velocity



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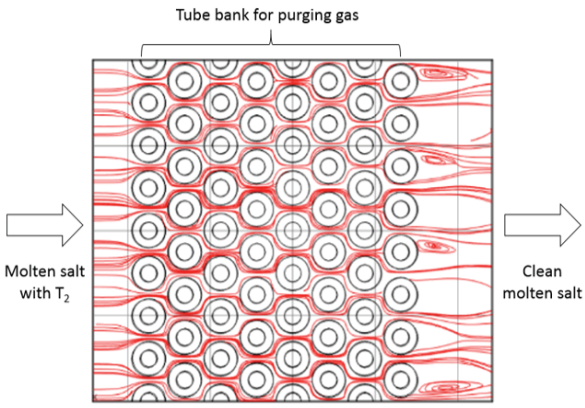
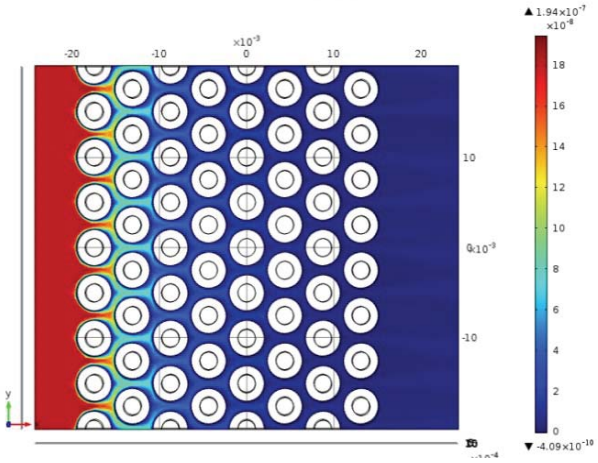
Computational Simulation Using COMSOL





- Plot of Salt Flow Streamlines
  - Main streamlines are splitted each time as they meet the next row of tubes
- Plot of H_2 Concentration Distribution in the Molten Salt
  - Transport coefficients of H_2 instead of T_2 in FLiBe used due to the lack of data
  - H_2 concentration decreases quickly along the salt flow path

Code Calculation Using MATLAB

- Overall Mass Transfer Coefficient k_o

$$\left(\frac{1}{k_o H_s}\right)^{\sqrt{2}} = \left(\frac{1}{k_s H_s}\right)^{\sqrt{2}} + \left[\frac{t_w}{K_w}\frac{(p_{1,in}^{0.5} - p_2^{0.5}) - (p_{1,out}^{0.5} - p_2^{0.5})}{\ln\left(\frac{p_{1,in}^{0.5} - p_{2,in}^{0.5}}{p_{1,out}^{0.5} - p_{2,in}^{0.5}}\right)}\right]^{\sqrt{2}}$$

- Corresponding Dimensionless Groups of Mass and Heat Transfer

No.	Mass transfer	Heat transfer
1	Reynolds number $\mathrm{Re} = \frac{\rho v D}{\mu}$	Reynolds number $\mathrm{Re} = \frac{\rho v D}{\mu}$
2	Schmidt number $\mathrm{Sc} = \frac{\mu}{\rho D_{AB}}$	Prandtl number $\mathrm{Pr} = \frac{c_p \mu}{k} = \frac{\nu}{\alpha}$
3	Sherwood number $\mathrm{Sh} = \frac{k_x D}{D_{AB}}$	Nusselt number $\mathrm{Nu} = \frac{hD}{k}$
4	Peclet number $\mathrm{Pe} = \mathrm{Re}\,\mathrm{Sc}$	Peclet number $\mathrm{Pe} = \mathrm{Re}\,\mathrm{Sc}$
5	Grashof number $\mathrm{Gr} = \frac{gL^3(\Delta\rho)}{\rho}\left(\frac{\rho}{\mu}\right)^2$	Grashof number $\mathrm{Gr} = \frac{gD^3\rho^2\beta(\Delta T)}{\mu^2}$ β=coefficient of expansion
6	Stanton number $\mathrm{St} = \frac{\mathrm{Sh}}{\mathrm{Re}\,Sc} = \frac{\mathrm{Sh}}{\mathrm{Pe}}$	Stanton number $\mathrm{St} = \frac{\mathrm{Nu}}{\mathrm{Re}\,\mathrm{Pr}} = \frac{\mathrm{Sh}}{\mathrm{Pe}}$

- Sherwood number is calculated using the correlations for Nusselt number: heat transfer coefficient *h* replaced with mass transfer coefficient k_x and thermal conductivity *k* replaced with diffusivity D_{AB}
- The concept of heat transfer resistance is applied to mass transfer using corresponding parameters

Facility Design Comparisons

	Dimension Set A	Dimension Set B
Mass flow rate of molten salt [kg/s]	11190.8	
Tritium inlet concentration [mol/m³]	1.8×10^{-6}	
Tritium outlet concentration [mol/m³]	1.62×10^{-6}	
Tritium removal rate [mol/s]	1.8×10^{-7}	
Tube OD [in]	1.050	1.315
Tube ID [in]	0.824	1.049
Tube wall thickness [in]	0.113	0.133
Tube bank pitch [in]	1.31 (Pitch/OD = 1.25)	1.64 (Pitch/OD = 1.25)
Tube length [in]	18	18
Tube number	49971	41365
Molten salt inlet frontal velocity [m/s]	1.0	1.0
Re	4.64×10^4	5.82×10^4
Molten salt inlet flow area [m²]	5.54 (2.35 × 2.35)	5.54 (2.35 × 2.35)
Total mass transfer area [m²]	9.85×10^3	1.02×10^4
Molten salt flow length estimated [m]	20.61 (in the direction normal to the tube bank)	26.72 (in the direction normal to the tube bank)
Molten salt frictional pressure loss [kPa]	197	189

- Fluoride salt flow rate from the AHTR preliminary design
- Tritium inlet concentration is raised to 10 times of that equivalent to the tritium production rate in the core

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FLUORIDE-SALT-COOLED HIGH-TEMPERATURE REACTORS (FHR)

Implications for Tritium Management

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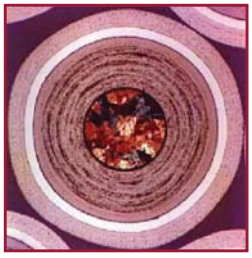
Workshop on Tritium Control and Capture in Salt-Cooled Fission and Fusion Reactors: Experiments, Models, and Benchmarking
Salt Lake City
October 27, 2015

FHR Combines Existing Technologies



Fuel: High-Temperature Coated-Particle Fuel Developed for High-Temperature Gas-Cooled Reactors (HTGRs)

Coolant: High-Temperature, Low-Pressure Liquid-Salt Coolant developed for the 1950s Aircraft Nuclear Propulsion Program

Power Cycle: Salt Cooling Creates New Options including Brayton Power Cycles

The FHR Is a Family of Reactors

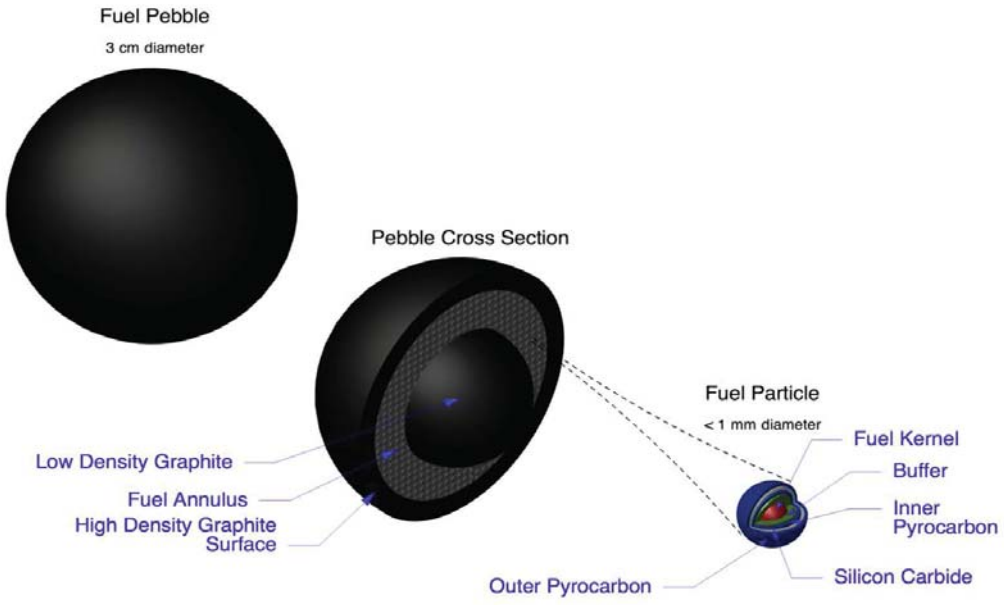
- Designs from 50 to 3000 MWt
- Different fuel geometries
- Different high-temperature salts
- Different power cycles

The Fuel

Advances in Carbon-Matrix Coated-Particle Fuel are the Enabling Technology for the FHR

FHR Uses Graphite-Matrix Coated-Particle Fuel

Same Fuel as High-Temperature Gas-Cooled Reactors in Several Different Geometric Forms



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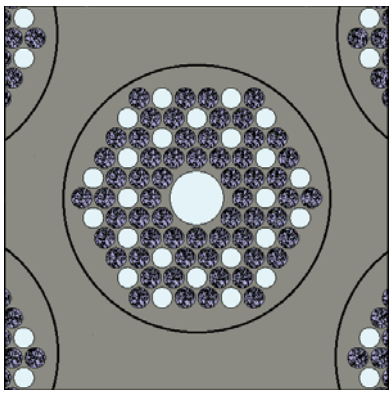
Many Fuel Options

Graphite-Matrix Coated-Particle Fuels

Pebble Bed

Fuel Plates in Hex Configuration



Fuel Inside Radial Moderator (FIRM)

- Pebble bed: Base-Case: Current technology
- Plate Fuel: Existing materials, New Design
- Fuel in Radial Moderator: Variant of HTGR Prismatic Block Fuel

Pebble-Bed FHR Reactor Built on Helium-Cooled Pebble-Bed Reactor Technology

- Most developed design and the near term option
- Similar to helium-cooled pebble bed reactors
  - FHR power density 4 to 10 times higher because liquids are better coolants than gases
  - On-line refueling (but pebbles float in salt so pebbles out top

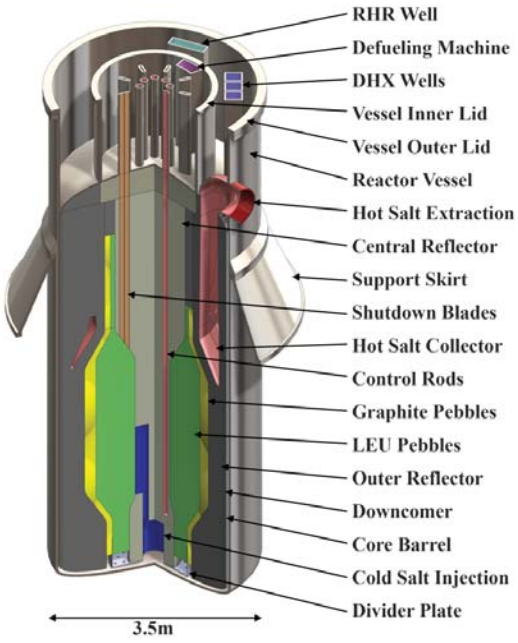
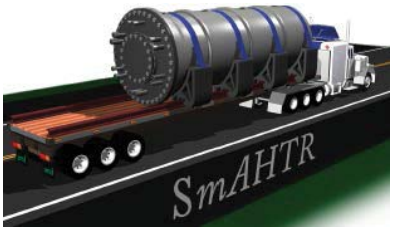


Plate-Fuel FHR Built Upon Sodium Fast Reactor Plant Designs

- Hexagonal fuel assembly
- Plant designs similar to sodium fast reactors (low pressure, high-temperature coolant)
- "New" fuel
  - Coated-particle fuel
  - Carbon composite plates



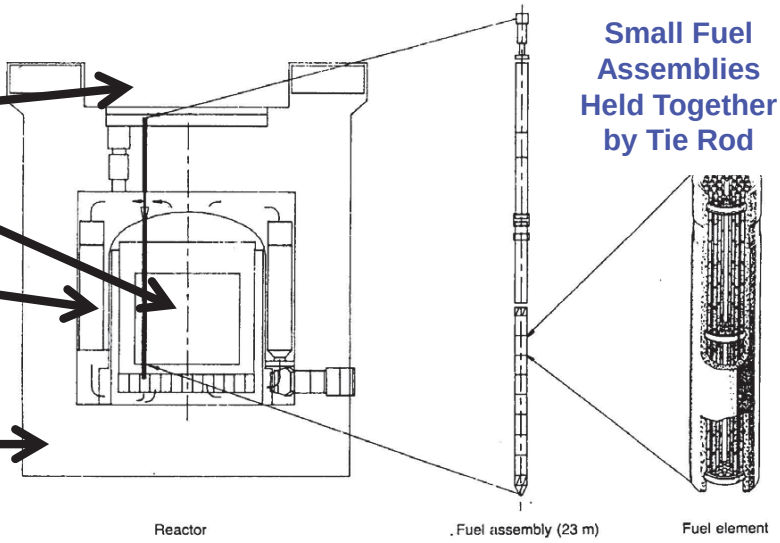
2010 125 MWt

2012 3600 MWt

FIRM FHR Built upon British Advanced Gas-Cooled High-Temperature Reactor (AGR)

14 AGRs Operating (2-Reactor Plants)
Graphite Moderated, Carbon-Dioxide-Cooled, Metal-Clad Pin Fuel

- Refueling Floor
- Graphite Core
- Boiler
- Pre-Stress Concrete Reactor Vessel



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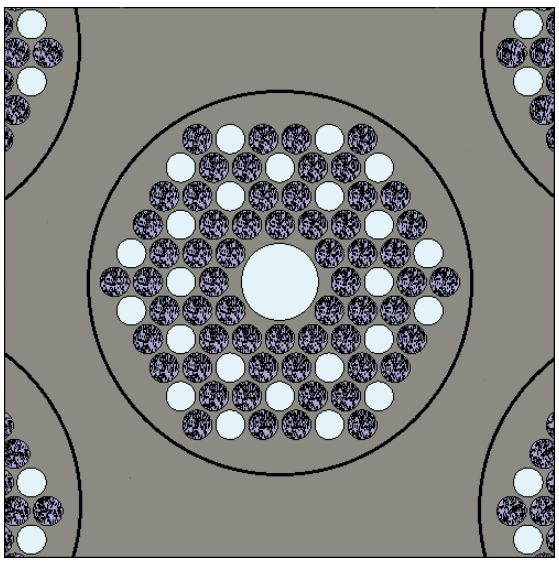
Use AGR Core, External Fuel Geometry and Refueling Designs

Fuel Inside Radial Moderator (FIRM) Assembly Design

- Surround fuel and coolant channels with solid graphite region
  - 54 fuel channels
  - 24 coolant channels
  - Central hole for handling and materials irradiations
- Introduces spatial resonance self-shielding:
  - Enhances resonance escape probability
  - Significantly increases fuel burnup



Fuel Design is Variant of Ft. St. Vrain Gas-Cooled High-Temperature Reactor Fuel

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Similar FHR and AGR FIRM Fuel Geometry →Similar Core Designs

- Similar refueling (AGR 650°C versus 700°C peak FHR coolant temperatures)
- Similar in-core graphite inspection / maintenance
- Similar instrumentation
- Similar control rod systems
- 50-year AGR operational experience base to build upon

But FHR is Low-Pressure with Liquid Cooling so Much Smaller Machine

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Advanced Fuel Option: Work at General Atomics and Elsewhere May Enable FHR Pin-Type Fuel Assemblies

- Lower fuel fabrication costs
- Lower enrichments with higher fuel loading
- Longer fuel cycle and higher burnup (less waste)
- Work in progress—being developed as part of LWR accident tolerant fuel program

The Salt Coolant

Massachusetts Institute of Technology

For Most Proposed FHRs The Base Case Salt is $^{7}Li_2BeF_4$ (Flibe)

There Are Alternative Coolant Salts

Coolant	T_{melt} (°C)	T_{boil} (°C)	ρ (kg/m^3)	ρC_p (kJ/m^3 °C)
$^{7}Li_2BeF_4$ (Flibe)	**459**	**1430**	**1940**	**4670**
59.5 NaF-40.5 ZrF_4	500	1290	3140	3670
26 ^{7}LiF-37 NaF-37 ZrF_4	436		2790	3500
51^{7} LiF-49 ZrF_4	509		3090	3750
Water (7.5 MPa)	0	290	732	4040

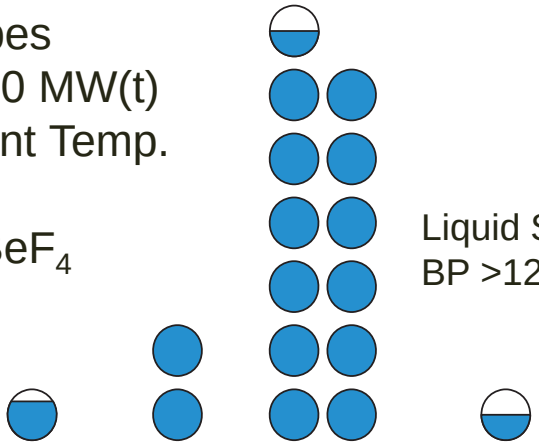
Salt compositions are shown in mole percent. Salt properties at 700ºC and 1 atm. Sodium-zirconium fluoride salt conductivity is estimated—not measured. Pressurized water data are shown at 290°C for comparison.

Liquid-Salt Coolant Properties Can Reduce Equipment Size and Thus Costs

(Determine Pipe, Valve, and Heat Exchanger Sizes)

Number of 1-m-diam. Pipes Needed to Transport 1000 MW(t) with 100ºC Rise in Coolant Temp.

Baseline salt: Flibe: $^{7}Li_2BeF_4$



	Water (PWR)	Sodium (LMR)	Helium	**Liquid Salt**
Pressure (MPa)	15.5	0.69	7.07	**0.69**
Outlet Temp (ºC)	320	540	1000	**To 1000**
Coolant Velocity (m/s)	6	6	75	**6**

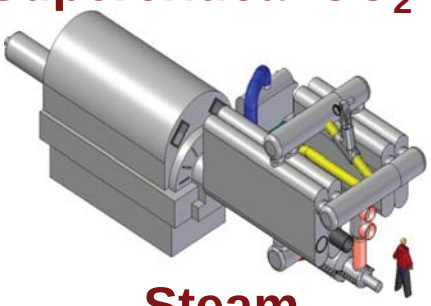
The Power Cycle

Power Cycle Options with 700°C Salt

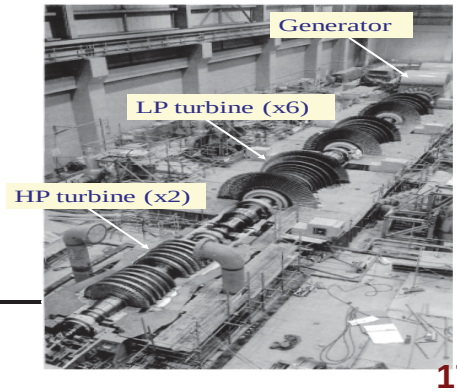
NACC or NHCC

Nuclear Air or Nuclear Helium Brayton Combined Cycle based on natural-gas plants

Supercritical CO_2



Steam



Power Cycle Choices May Impact Tritium Control Strategies

- Can trap tritium in some power cycles because cold side of power cycle prevents tritium releases
  - Supercritical carbon dioxide
  - Helium Bryaton cycles
- Tritium major challenge if enters some power cycles
  - Steam cycles
  - Air-Brayton power cycles

The FHR Tritium Challenge

FHR Tritium Challenge

- Reactor environment
  - Clean fluoride salt coolant containing Lithium-7
  - Tritium generation varies by order of magnitude depending upon FHR design
  - Carbon-based fuel that absorbs tritium and impacts chemistry
  - Potential for small quantities of fission products from leaking fuels
- Tritium challenge
  - FHRs produce tritium significantly above levels requiring controls
  - Tritium absorbed in graphite fuel in significant quantities
    - Possibility in pebble bed to capture tritium on pebbles and recycle pebbles with tritium removal during recycle for tritium control
    - Must consider off-normal events where tritium inventory in fuel may be released if increased core temperatures
- Tritium is a waste—can recover for use but not a requirement

Questions

Tritium Environment Below-Previous Slide

- Reactor environment
  - Clean fluoride salt coolant containing Lithium-7
  - Inventory varies by order of magnitude depending upon design
  - Carbon-based fuel
  - Potential for small quantities of fission products from leaking fuels
- Tritium challenge
  - FHRs produce tritium significantly above levels requiring controls
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- Tritium is a waste—can recover for use but not a requirements

Biography: Charles Forsberg

Dr. Charles Forsberg is the Director and principle investigator of the High-Temperature Salt-Cooled Reactor Project and University Lead for the Idaho National Laboratory Institute for Nuclear Energy and Science (INEST) Nuclear Hybrid Energy Systems program. He is one of several co-principle investigators for the Concentrated Solar Power on Demand (CSPonD) project. He earlier was the Executive Director of the MIT Nuclear Fuel Cycle Study. Before joining MIT, he was a Corporate Fellow at Oak Ridge National Laboratory. He is a Fellow of the American Nuclear Society, a Fellow of the American Association for the Advancement of Science, and recipient of the 2005 Robert E. Wilson Award from the American Institute of Chemical Engineers for outstanding chemical engineering contributions to nuclear energy, including his work in hydrogen production and nuclear-renewable energy futures. He received the American Nuclear Society special award for innovative nuclear reactor design on salt-cooled reactors and the 2014 Seaborg Award. Dr. Forsberg earned his bachelor's degree in chemical engineering from the University of Minnesota and his doctorate in Nuclear Engineering from MIT. He has been awarded 11 patents and has published over 200 papers.

Many Teams Working on the FHR

- MIT, UCB, UW, and UNM

Organization	PI	Area
MIT	Charles Forsberg Project Leader	Market case Severe Accidents
	Lin-wen Hu	Irradiation experiments
University of California, Berkeley	Per F. Peterson	Thermal-hydraulics, safety
	Massimiliano Fratoni	Neutronics
University of Wisconsin, Madison	Kumar Sridharan	Materials
University of New Mexico	Edward Blandford	Thermal-hydraulics, safety

- National Laboratories: ORNL, INL, etc.
- Georgia Tech Consortium
- Chinese Academy of Science (2020 FHR test reactor)
- Vendors

The Idea of a Fluoride-salt-cooled High-temperature Reactor (FHR) dates to 2002

- No FHR has been built
- Compelling reasons must exist to develop a new reactor type
  - Commercial: Improve economics
  - Government: Meet national goals
  - Public: Safety against major accidents

MOLTEN-SALT–COOLED ADVANCED HIGH-TEMPERATURE REACTOR FOR PRODUCTION OF HYDROGEN AND ELECTRICITY

FISSION REACTORS

KEYWORDS: molten salt, high-temperature reactor, hydrogen production

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The molten-salt–cooled Advanced High-Temperature Reactor (AHTR) is a new reactor concept designed to provide very high-temperature (750 to 1000°C) heat to enable efficient low-cost thermochemical production of hydrogen (H_2) or production of electricity. This paper provides an initial description and technical analysis of its key features. The proposed AHTR uses coated-particle graphite-matrix fuel similar to that used in high-temperature gas-cooled reactors (HTGRs), such as the General Atomics gas turbine–modular helium reactor. However, unlike the HTGRs, the AHTR uses a molten-salt coolant and a pool configuration, similar to that of the General Electric Super Power Reactor Inherently Safe Module (S-PRISM) liquid-metal reactor. Because the boiling points for molten fluoride salts are near ~1400°C, the reactor can operate at very high temperatures and atmospheric pressure. For thermochemical H_2 production, the heat is delivered at the required near-constant high temperature and low pressure. For electricity production, a multireheat helium Brayton (gas-turbine) cycle, with efficiencies >50%, is used. The low-pressure molten-salt coolant, with its high heat capacity and natural circulation heat transfer capability, creates the potential for robust safety (including fully passive decay-heat removal) and improved economics with passive safety systems that allow higher power densities and scaling to large reactor sizes [>1000 MW(electric)].

I. INTRODUCTION

The Advanced High-Temperature Reactor (AHTR) is a new reactor concept developed by the authors to produce high-temperature heat (750 to 1000 °C) for efficient production of electricity and thermochemical H_2. The AHTR is based on four technological developments:

1. high-temperature, low-pressure molten-fluoride-salt reactor coolants from the aircraft nuclear propulsion program[1] of the 1950s and the molten-salt breeder reactor program of the 1960s and 1970s.[2] Today, development continues on using these salts and associated advanced high-temperature materials for cooling fusion reactors.

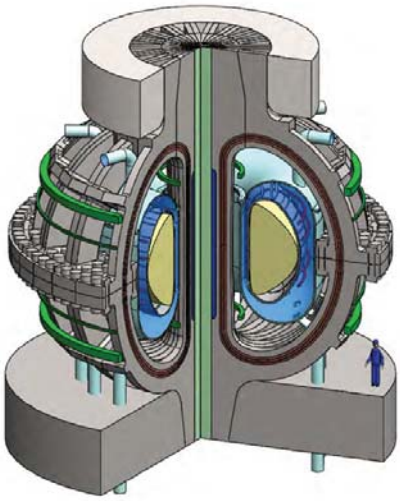
2. coated-particle graphite-matrix fuel developed for high-temperature gas-cooled reactors[3] (HTGRs) in the United States and Germany, starting in the 1960s.

3. passive safety systems for gas-cooled and liquid-metal reactors introduced in the 1980s.

4. advanced gas turbines—including commercialization in the last 5 yr of magnetic bearing systems that can permit these turbines to be used in closed helium cycles.

*E-mail: forsbergcw@ornl.gov

Liquid immersion blankets for fusion power plants



Brandon Nils Sorbom

Workshop on Tritium Control and Capture in Salt-Cooled Fission and Fusion Reactors, October 27, 2015

Thank you to all who contributed to ARC!

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Collaborators

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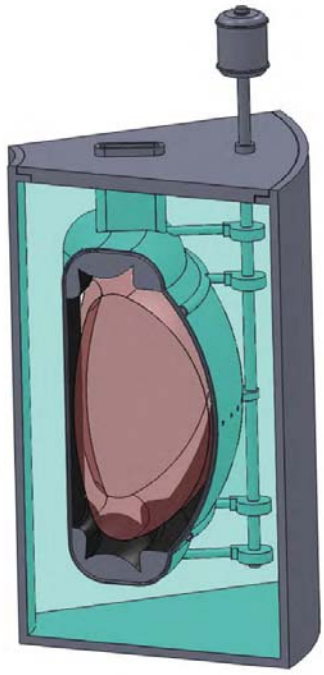
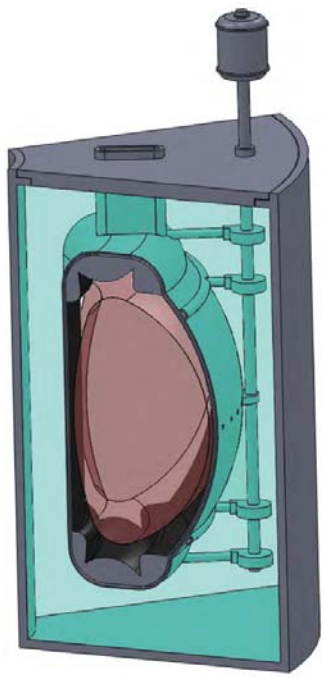
Outline



- What functions do a fusion blanket have to perform?
- Traditional blankets vs. the liquid blanket
- How well do liquid blankets perform?

Outline



- What functions do a fusion blanket have to perform?
- Traditional blankets vs. the liquid blanket
- How well do liquid blankets perform?

Fusion blanket systems must perform three main roles—two are different than for fission!

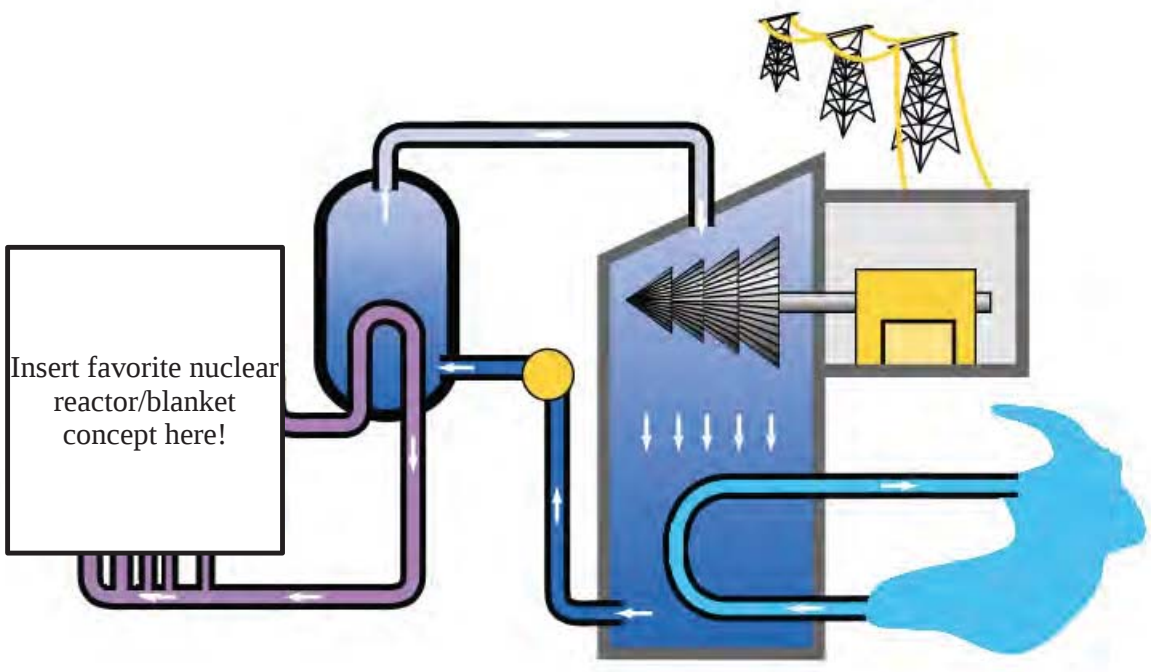
A blanket must:

1) Extract the energy from the fusion reactions in a useable form (same as fission)

2) Breed enough tritium to fuel the reactor

3) Shield the TF coils from high-energy neutrons

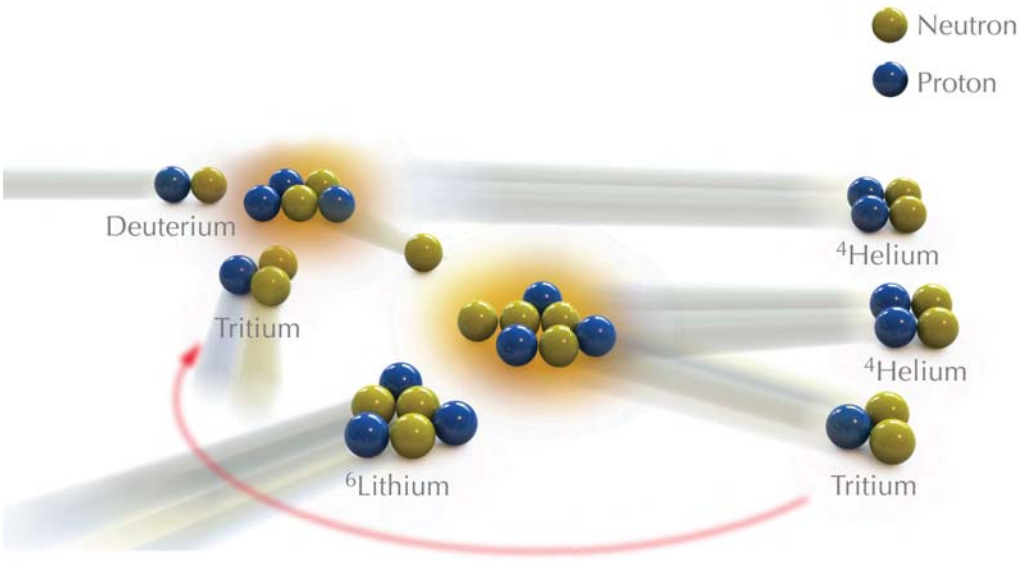
Blanket is used to extract useable energy from nuclear reactions



Energy extraction blanket requirements

- Should have favorable thermal-hydraulic properties
  - Low density and viscosity (easy to pump)
  - High heat capacity (efficient at removing heat)
  - High temperature operation (higher Carnot efficiency)
- Want something that looks like water, but at a higher temperature
- Only major difference from fission is that MHD effects in fluid become important (more on this later)

The blanket must breed fuel (tritium) for the reactor



https://www.euro-fusion.org/glossary/tritium-breeding/

Tritium breeding requirements

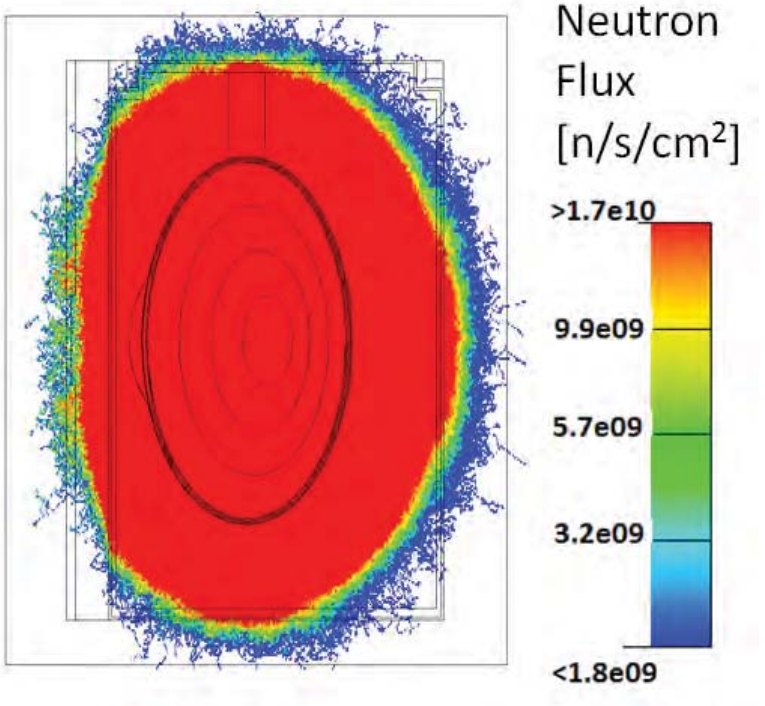
- 500 MWth ARC reactor consumes ~67 g of T *per day*
- Must provide a tritium breeding ratio (TBR) greater than 1, i.e. you get more tritium out of the blanket than you put into the reactor

$$TBR = \frac{Tritium\ Bred}{Tritium\ Consumed}$$

- We use the ARIES critera[1] to have TBR > 1.1 to account for deficiencies in nuclear data and uncertainties in exact reactor geometry
- Lithium is by far the best material to breed tritium, so blanket must have lithium in some form
- As a bonus, the blanket could have a neutron-multiplying isotope (such as beryllium) to increase the number of low-energy neutrons to interact with Li-6

1.) El-Guebaly, L. A., & Malang, S. (2009). Toward the ultimate goal of tritium self-sufficiency: Technical issues and requirements imposed on ARIES advanced power plants. Fusion Engineering and Design, 84(12), 2072-2083.

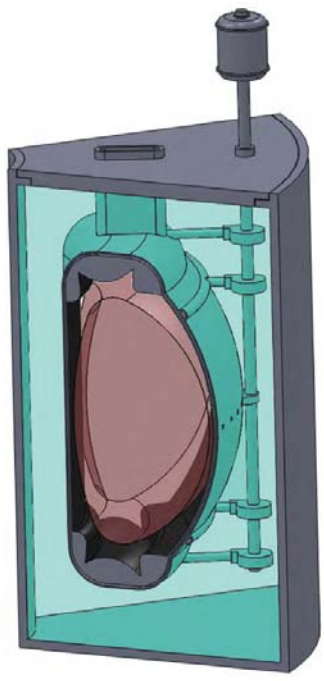
The blanket must effectively shield the TF coils from neutrons



Magnet Shielding Requirements

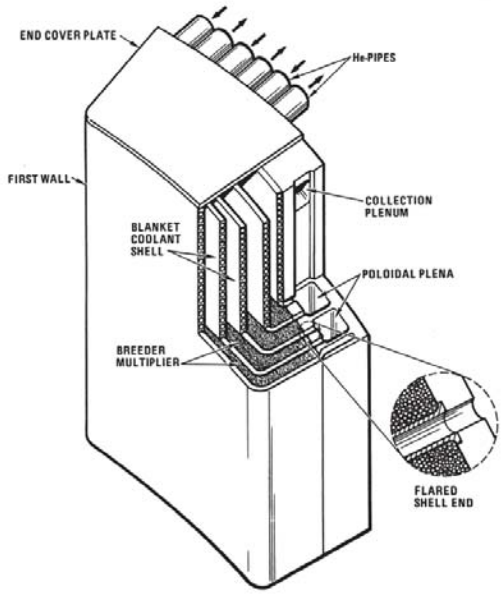
- All practical reactor designs utilize superconducting coils
- Critical current capability of superconductor degrades after a certain amount of damage from high-energy (> 0.1 MeV) neutrons
- REBCO high-temperature superconductors have not been tested to failure, but Nb_3Sn starts degrading around a neutron fluence of $3x10^{18}$ neutrons/cm^2 for high energy neutrons

Outline

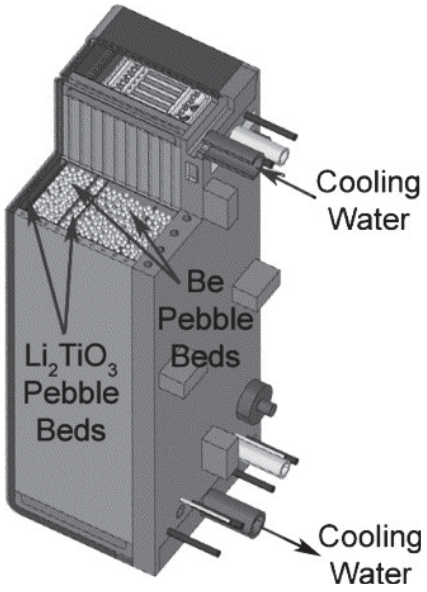


- What functions do a fusion blanket have to perform?
- Traditional blankets vs. the liquid blanket
- How well do liquid blankets perform?

"Traditional" blankets utilize a large amount of solid material



ARIES-I Blanket



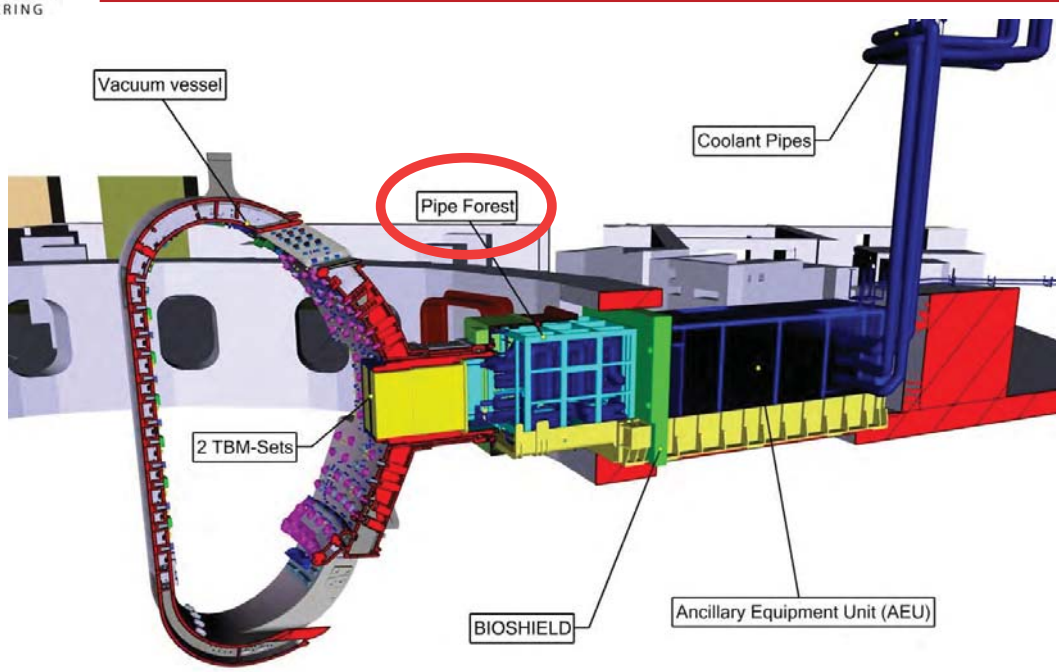
Japanese Tritium Breeding Module for ITER

13

Najmabadi, Farrokh, et al. "The ARIES-I Tokamak Reactor Study." Fusion Science and Technology 19.3P2A (1991): 783-790.

Giancarli, L., et al. "Breeding blanket modules testing in ITER: an international program on the way to DEMO." Fusion Engineering and Design 81.1 (2006): 393-405.

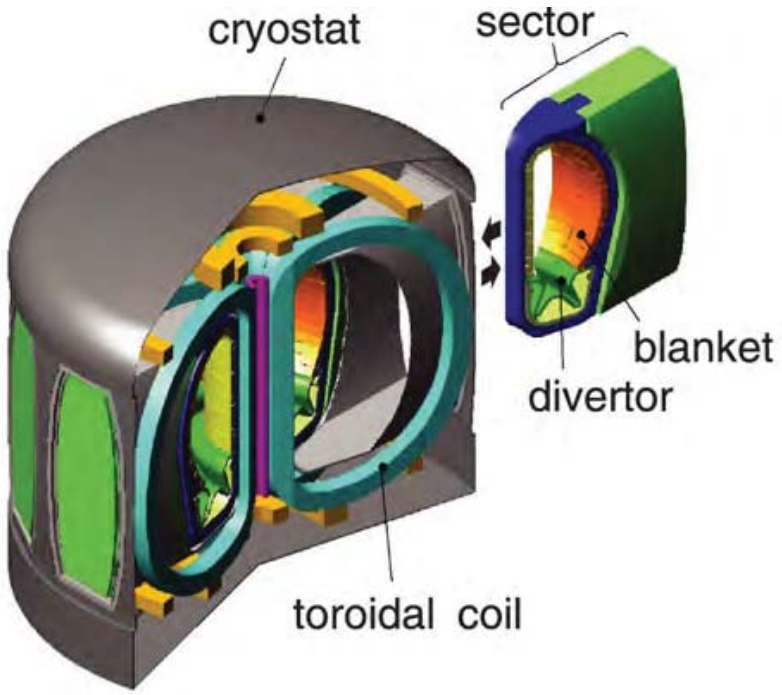
Solid blankets are complicated



https://www.iter.org/newsline/-/2207

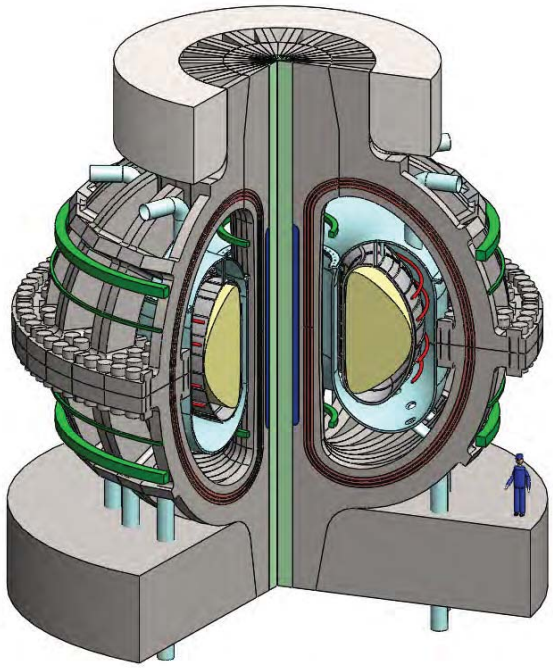
14

Sector maintenance is not ideal



http://jolisfukyu.tokai-sc.jaea.go.jp/fukyu/mirai-en/2008/3_10.html

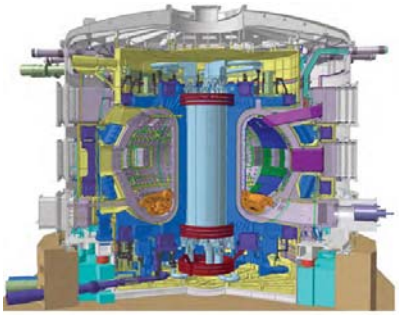
The ARC reactor is a high-field conceptual pilot plant design



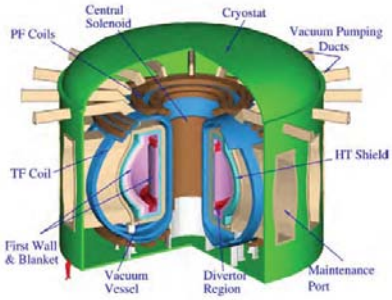
Key Design Parameter	Value
Fusion Power	525 MW
Total Thermal Power	708 MW
Net Electric Power	190 MW
Plasma/Electric Gain	13/3
Major Radius	3.3 m
Minor Radius	1.1 m
Toroidal Magnetic Field	9.2 T
Plasma Current	7.8 MA
Average Temperature	13.9 keV
Average Density	1.75 $x10^{20}$ m^{-3}
Tritium Breeding Ratio	1.10

What makes ARC different from other reactor designs?

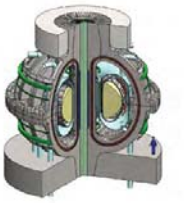
- ARC is compact and has a high magnetic field through the use of high-temperature superconductors
- ARC's magnets are demountable (no sector maintenance)
- **ARC has an all liquid, molten salt blanket**



ITER[2]

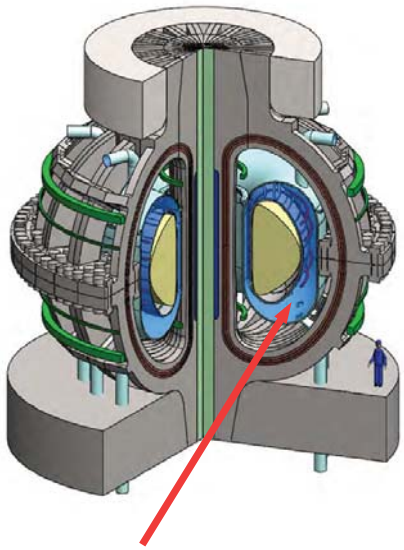


ARIES-AT[3]

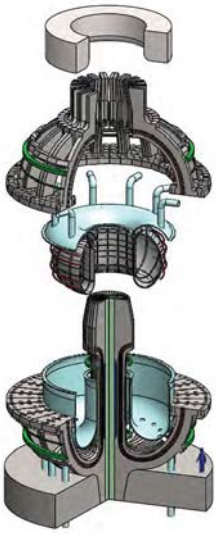


ARC

Liquid immersion blanket fully surrounds the vacuum vessel

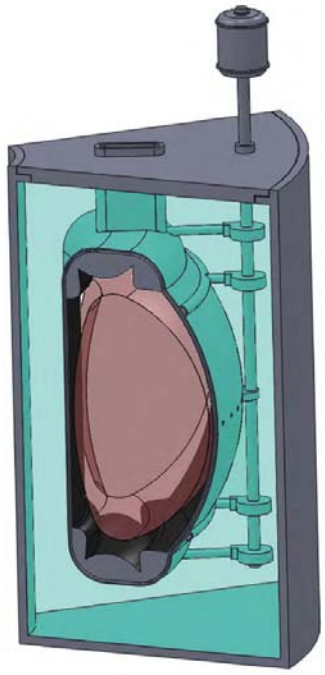


Liquid Blanket
(represented as blue liquid)



Liquid blanket not required for vertical maintenance scheme, but simplifies things considerably

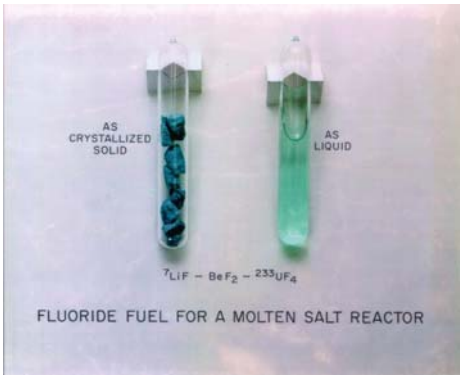
Outline



- What functions do a fusion blanket have to perform?
- Traditional blankets vs. the liquid blanket
- How well do liquid blankets perform?

Many liquid blanket candidates have been investigated by the fission community

- For fusion, tritium breeding sets first requirement—the liquid must have some element that can breed tritium
- As mentioned before, lithium is only practical tritium breeding material, so whatever liquid is chosen must contain lithium



- An (incomplete) list of liquid blanket materials
  - **$LiF\text{-}BeF_2$ (FLiBe)**
  - **LiF-NaF-KF (FLiNaK)**
  - $Kf\text{-}ZrF_4$
  - $Kcl\text{-}MgCl_2$
  - $NaNO_3\text{-}NaNO_2\text{-}KNO_3$
  - NaF-ZrF4
  - **PbLi**
  - **Liquid lithium**

How does a liquid blanket stack up? – Thermohydraulics

- FLiBe and FLiNaK have the closest heat transfer properties to water
- All liquid blankets look roughly as easy to pump as water...but what about MHD?

Property[2]	FLiBe	FLiNaK	Liquid Li	PbLi	Water
Melting Point (K)	732	727	453	507	273
Density (kg/m^3)	1940	2020	475	8940	1000
Specific Heat (kJ/kg K)	2.4	1.93	4.15	0.19	4.2
Thermal Conductivity (W/m K)	1	0.88	57.7	19.5	0.58
Viscosity (mPa s)	6	4.11	0.280	0.89	1
Reynolds Number[1] normalized to water	**0.32**	**0.49**	**1.7**	**1**	**1**
Prandtl Number normalized to water	**2.4**	**1.26**	**0.002**	**0.001**	**1**

1. Assumed characteristic length of 1m and flow velocity of 0.2 m/s

2. Zinkle, S. J. "Summary of Physical Properties for Lithium, Pb-17Li, and (LiF) n. BeF2 Coolants." APEX Study Meeting, Sandia National Lab. 1998.

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MHD effects on liquid metals lead to large pumping power requirement but molten salts are less affected

- We have a liquid moving through an extremely high magnetic field—if the liquid is conductive, this leads to MHD effects!
- Simple flow through a conducting pipe, transverse to a magnetic field leads to the relationship:

$$P_p \propto \sigma_f B_\perp^2$$

- Actual pumping power required is highly dependent on geometry and magnetic field structure, but calculations[1] for simple cooling systems indicate that pumping power for liquid Li is ~10% of the thermal power of fusion device
- **FLiBe and FLiNaK are both orders of magnitude less conductive and would have very small MHD effects**

Property[2]	FLiBe	FLiNaK	Liquid Li	PbLi
Electrical Conductivity, σ_f (S/m)	241	230	2.3x10^6	7.0x10^7

1. Kammash, Terry. "Fusion reactor physics: principles and technology." (1975).

2. Zinkle, S. J. "Summary of Physical Properties for Lithium, Pb-17Li, and (LiF) n. BeF2 Coolants." APEX Study Meeting, Sandia National Lab. 1998.

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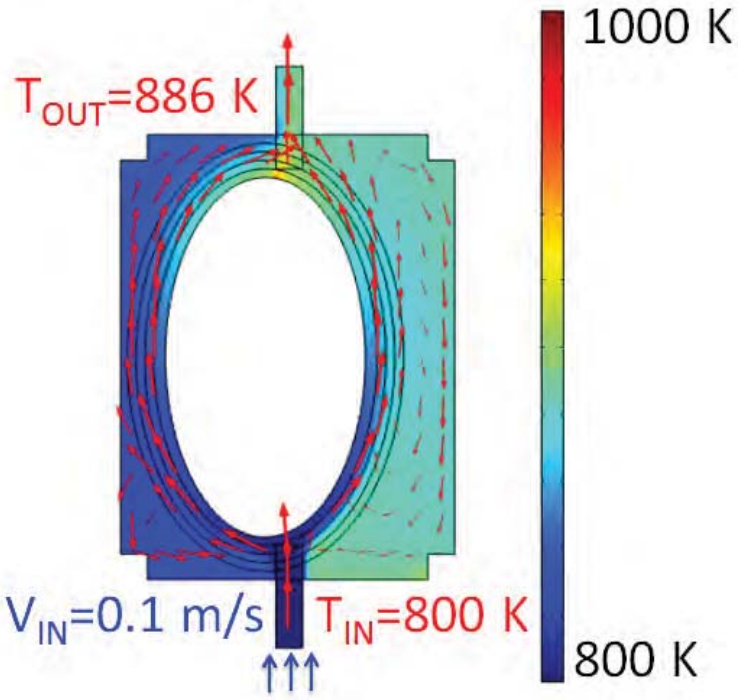
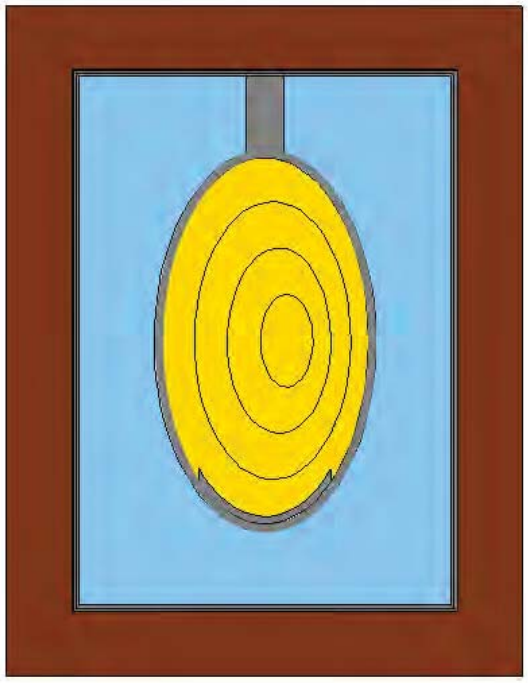
COMSOL turbulent flow simulation suggests minimal inlet velocity required to exhaust neutron heating

- FLiBe used as molten salt for COMSOL simulation, temperature-dependent properties (e.g. specific heat, thermal conductivity) manually input from literature
- MHD effects considered negligible for FLiBe and are not modeled
- Heating inputs are modeled as conduction through vacuum vessel from plasma heating and volumetric neutron heating assessed using MCNP
- Goal for ARC outlet temperature is approximately 900 K

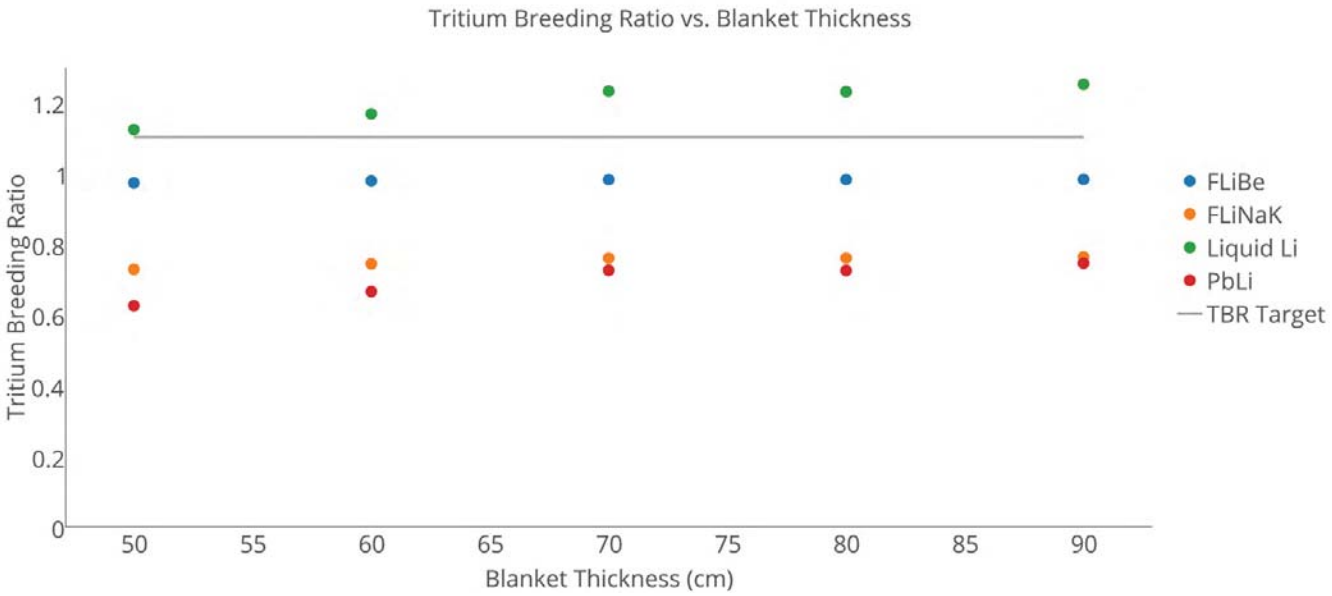


How does a liquid blanket stack up? – Tritium Breeding

- Unlike fission, we actually *want* to breed as much tritium as possible!
- For this assessment, I have used a simple MCNP model of the ARC reactor to compare our four candidates
- Neutron source is a four-volume approximation based on fusion plasma profiles, with most neutron production coming from core (also note that core shifted out due to plasma effects)
- Structural material in model is Inconel 617, a nickel-based alloy
- Vacuum vessel has internal cooling channel to model first wall cooling scheme



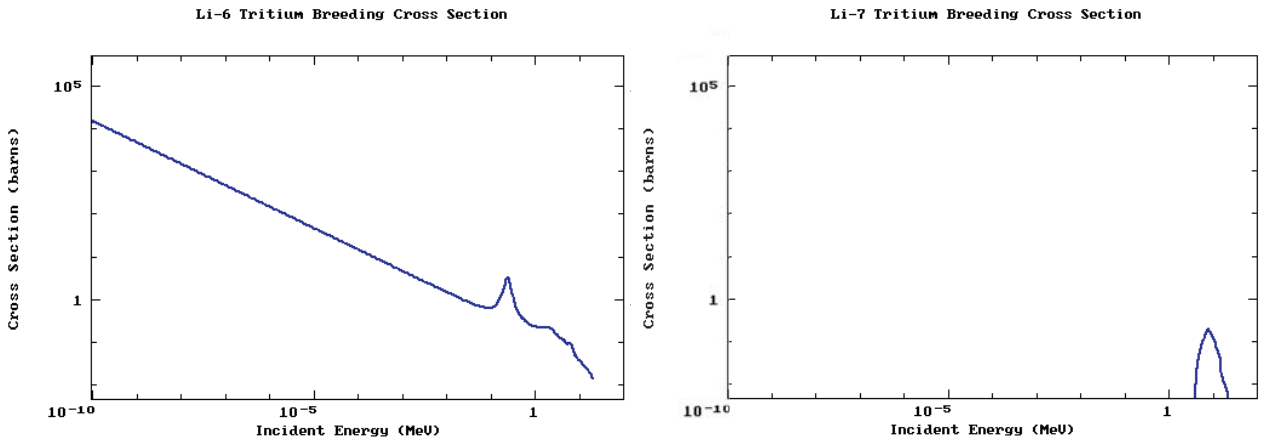
Natural abundance Li liquid blankets struggle to breed enough tritium



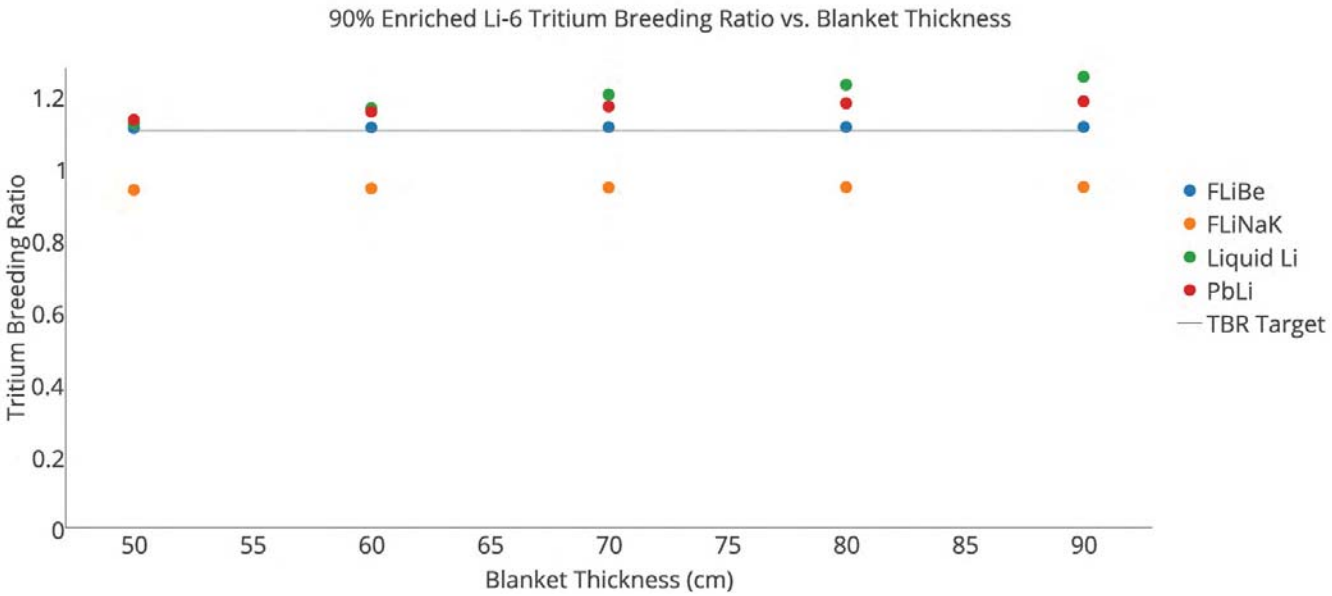
- Liquid lithium is the only blanket material which achieves TBR > 1.1
- Possible to boost breeding capacity by changing isotopic lithium fractions

Lithium must be enriched to provide adequate tritium breeding

- Li-6 has an enormously higher breeding cross section than Li-7, extending down to thermal energies
- Natural abundance is 7.5% Li-6 and 92.5% Li-7
- Enriching blanket with Li-6 will increase tritium breeding performance



Enriching to 90% Li-6 significantly improves breeding performance



- Enriching to 90% Li-6 allows FLiBe and PbLi to be used as tritium breeders
- Blanket thickness has much less of an effect on breeding for enriched blankets (this will be important later...)

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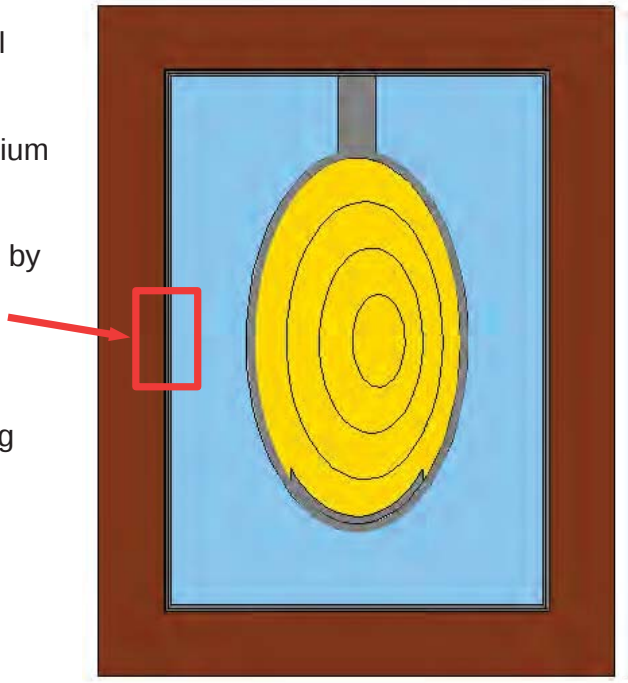
How does a liquid blanket stack up? – TF Shielding

- Also highly dependent on material and geometry
- Use same MCNP model as for tritium breeding calculations
- "Worst case" scenario considered by assessing neutron flux at inner midplane, where space is most limited
- TF lifetime calculated by assuming ARC power (525 MW) and using conservative neutron fluence limit from Nb_3Sn



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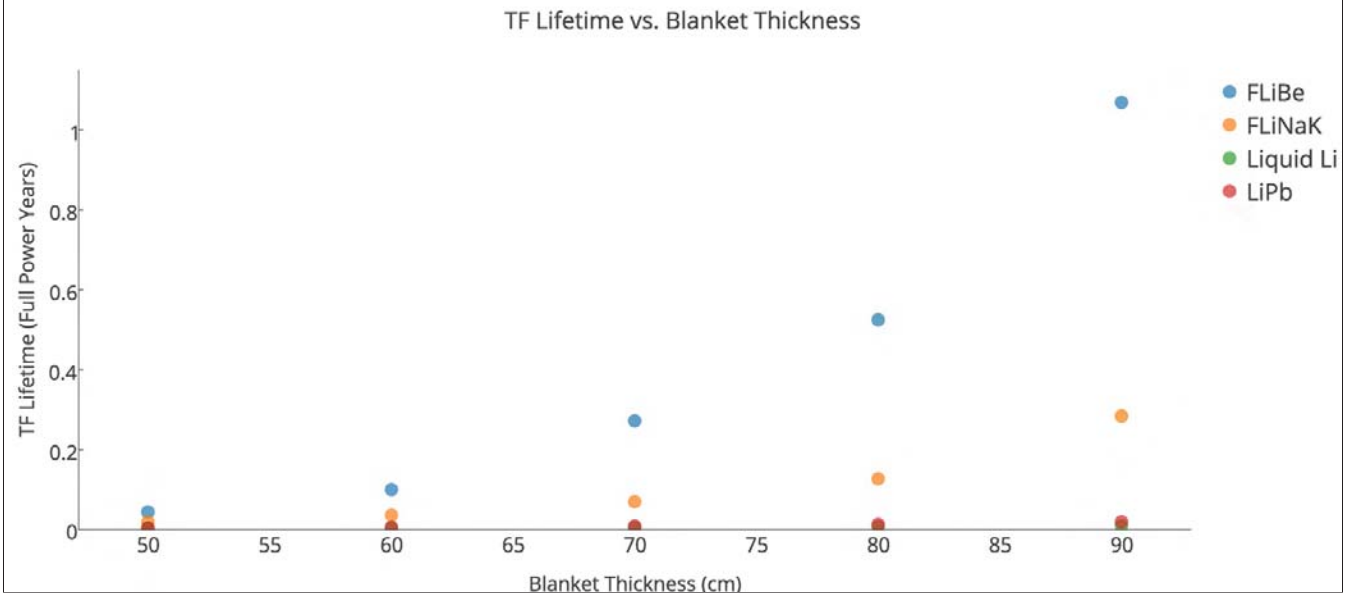
Liquid blankets are abysmal neutron shields

- Would require a lot of blanket to effectively shield TF, even with the best candidate material
- This requires us to use a secondary shielding material
- Hydrides work well (this was the solution for ARC)



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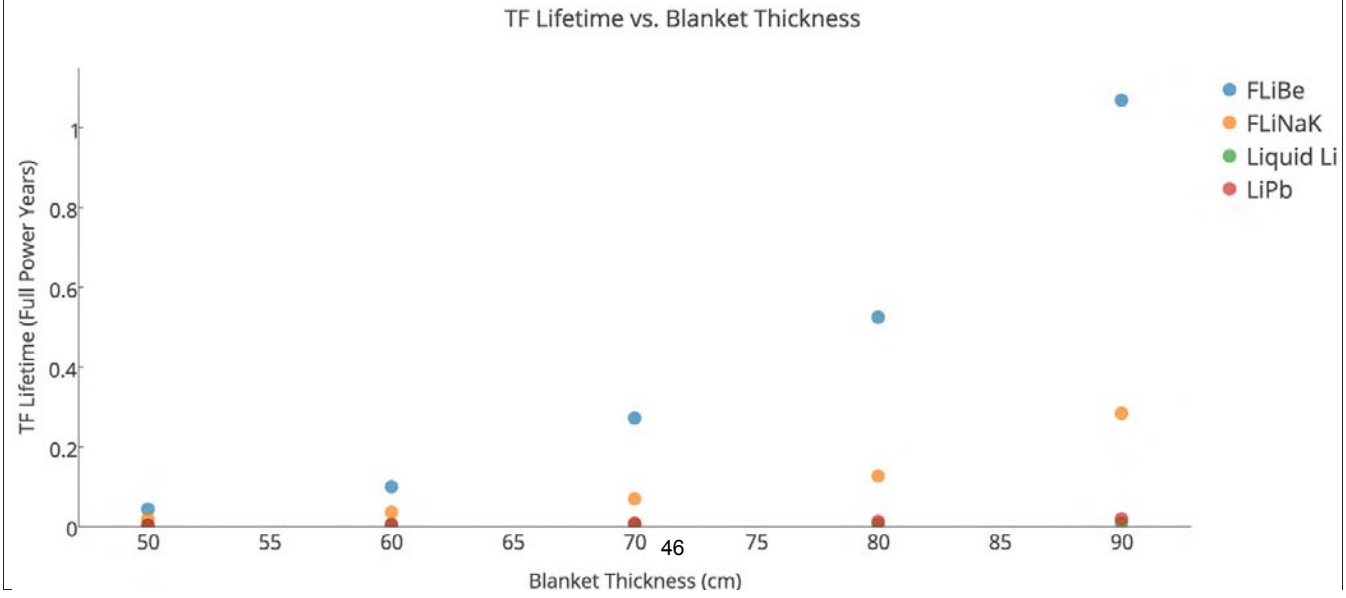
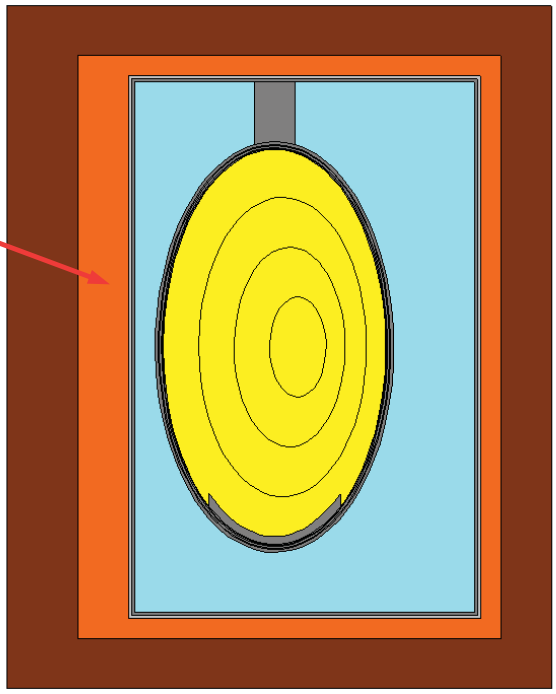


Solid shields on inboard side improve lifetime tremendously

- Solution to TF lifetime problem in ARC was to replace inboard blanket volume with TiH_2 shielding (represented in orange)
- For ARC, replacing 50cm of 70cm blanket with hydride shielding raised TF lifetime to 10 FPY (up from 0.4 FPY for pure FLiBe)
- Since most tritium breeding was found to occur within the first 30-40 cm of blanket, TBR was largely unaffected (1.095 from 1.110)



Other observations about liquid blankets...

- Less high-Z solid material in blanket means less activated waste and liquid is constantly circulating so less overall neutron exposure to blanket
- Exothermic nuclear reactions in blanket can have a large effect on power balance (ARC blanket reactions contribute ~100 MW on top of the 500 MW of fusion power)

Are we happy or sad about using liquid blankets in fusion?

- Energy conversion? – Happy, liquid blankets have favorable thermohydraulic properties
- Tritium breeding? – Happy, achieve TBR > 1.1
- Shielding? – Mostly happy, can supplement liquid blanket with additional shielding material
- Conclusion: Liquid immersion blankets are an attractive concept, worth pursuing in fusion reactor designs.

References

1. B. Sorbom et al. “ARC: A compact, high-field, fusion nuclear science facility and demonstration power plant with demountable magnets,” Submitted to Fusion Engineering and Design (2014)

2. Iter.org, http://www.iter.org/album/media/7%20-%20technical#2044

3. F. Najmabadi et al. "The ARIES-AT advanced tokamak, advanced technology fusion power plant." Fusion Engineering and Design 80.1 (2006): 3-23.

4. Zinkle, S. J. "Summary of Physical Properties for Lithium, Pb-17Li, and (LiF) n. BeF2 Coolants." APEX Study Meeting, Sandia National Lab. 1998.

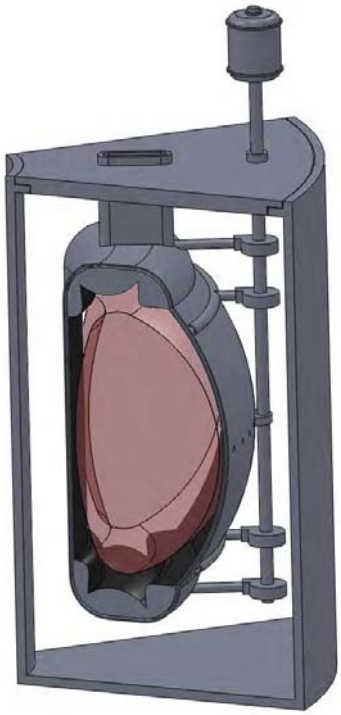
Backup Slides

An all-liquid FLiBe blanket provides magnet shielding, tritium breeding, and is a working fluid

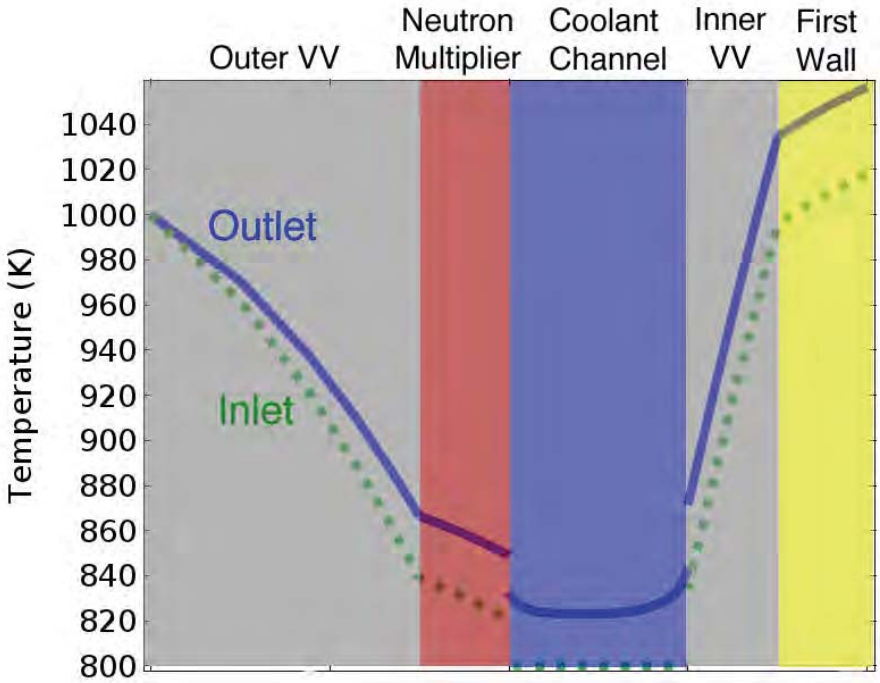
- Molten salt use in reactors is well-studied within fission community
- Fluorine Lithium Beryllium (FLiBe) molten salt has similar thermohydraulic properties to water—but at higher temperature (and operating window)

Property	FLiBe	Water
Melting Point (K)	732	273
Boiling Point (K)	1703	373
Density (kg/m^3)	1940	1000
Specific Heat (kJ/kg K)	2.4	4.2
Thermal Conductivity (W/m K)	1	0.58
Viscosity (mPa s)	6	1

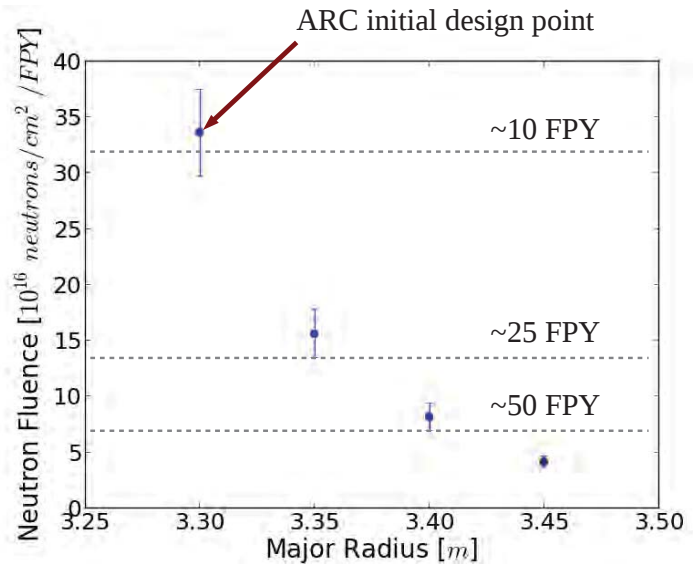
Double-walled Vacuum Vessel



Vacuum vessel has a cooling channel, with higher velocity FLiBe used as coolant

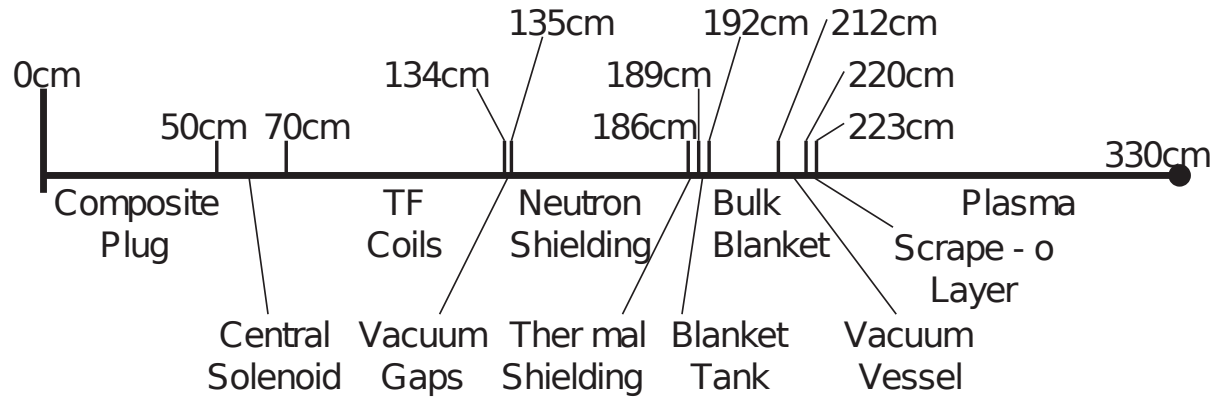


3% increase in R improves coil lifetime by a factor of 5

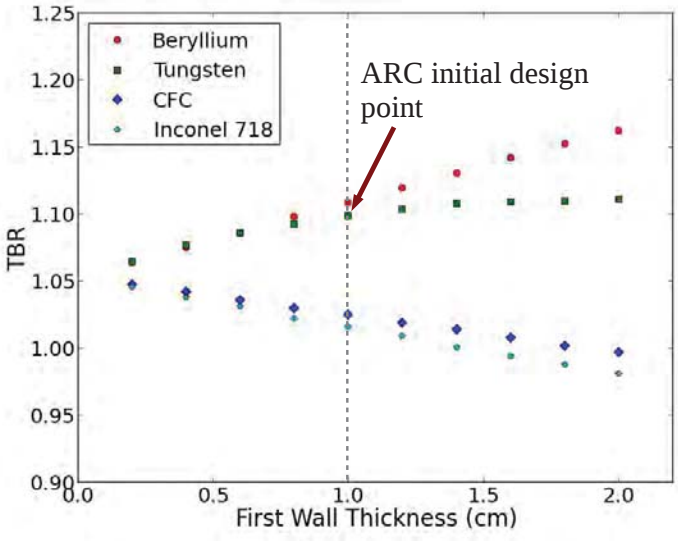


- Reactor lifetime limited by neutron fluence to superconducting coils
- ARC was optimized to provide the most compact reactor, so ~10 full-power year operation is acceptable
- Scaling up reactor size a small amount would allow for much longer lifetime, appropriate for a commercial power plant

ARC Inboard Radial Build



Neutronics simulations indicate range of possible first wall choices



- Tritium breeding ratio (TBR) must be above 1 to breed enough fuel to run reactor
- First wall material and thickness has a large effect on TBR
- ARC will allow multiple vacuum vessel/first wall configurations to be tested without building an entirely new device

Tritium Recovery System

- Full analysis beyond the scope of this conceptual design
- Through a literature search, found recent[1] Japanese studies on T extraction from FLiBe using "counter-current extraction tower"
- Basic concept:
  - Saturate FLiBe with Be to maintain TF concentration in FLiBe
  - Pass saturated FLiBe down through series of filters with He pumped up in opposite direction
  - TF diffuses in He, and T2 is pumped out with He and separated
  - According to study, achieves T recovery > 99.9%

1.) S. Fukada, A design for recovery of tritium from Flibe loop in FFHR-2 (2007)

TBR Uncertainty in Cross Sections for MCNP Calculation

- UCLA study found 2-6% uncertainty in TBR for various materials based on uncertainties in nuclear databases[1]
- Closest material combination to ours (FLiBe/He/FS/Be) had TBR predicted overestimate of ~4.3%
- Total uncertainty subtracted from our TBR still gives a TBR of 1.07

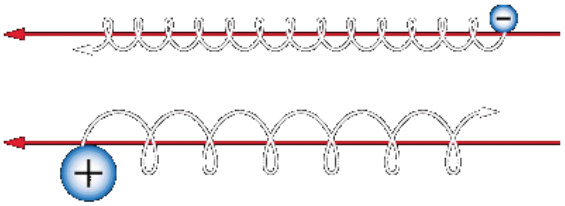
1.) Uncertainties in Prediction of Tritium Breeding in Candidate Blanket Designs Due to Present Uncertainties in Nuclear Data Base, M.Z. Youssef et al, (1986)

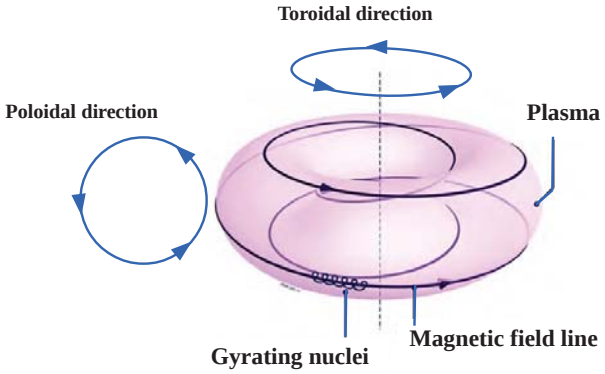
MCNP Validations

- Simple fluence validation – changed all cells to vacuum and made sure that all source neutrons accounted for
- TBR validated using simple toroidal model and comparing to UW results

Mainstream fusion community has accepted toroidal magnetic confinement as best candidate for fusion energy

$$\vec{F} = q(\vec{E} + \vec{v} \times \vec{B})$$





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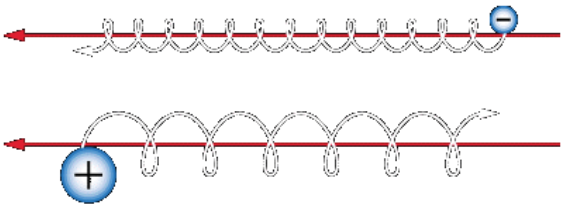
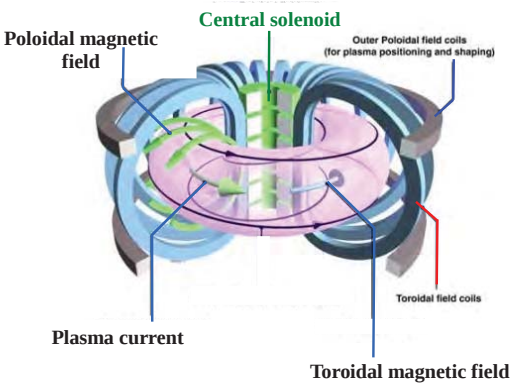
Mainstream fusion community has accepted tokamak magnetic confinement as best candidate for fusion energy

$$\vec{F} = q(\vec{E} + \vec{v} \times \vec{B})$$



- Wrap linear device around into a donut (torus)
- Add a central solenoid to inductively drive current in the plasma and give field lines a helical twist—now you have a tokamak!



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Fusion research is a serious, multinational effort, specifically tokamak research

Alcator C-Mod
Cambridge, MA, USA

ASDEX Upgade
Garching, Germany

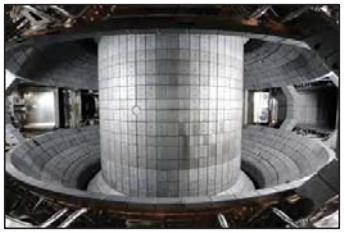
DIII-D, *San Diego, CA, USA*

EAST (HT-7)
Hefei, Anhui, China

Joint European Torus (JET)
Culham, Oxfordshire, UK

JT-60SA (under construction)
Naka, Japan

KSTAR, *Daejeon, Republic of Korea*



SST-1, *Gandhinagar, Gujarat, India*

Tore Supra, *Cadarache, France*

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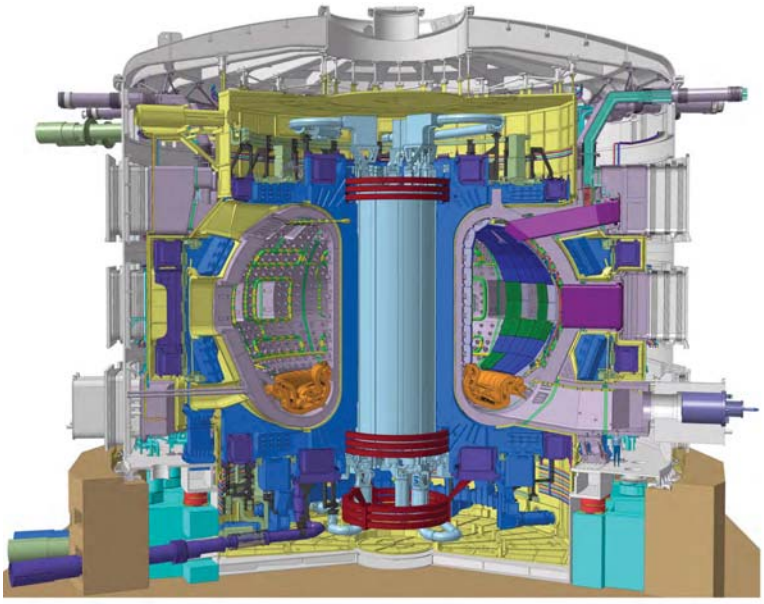
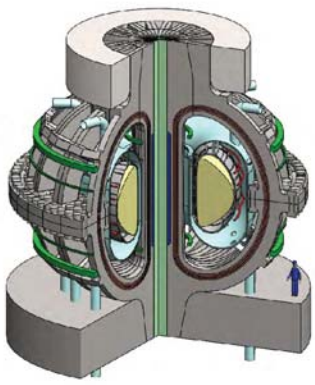
ARC is significantly smaller than ITER with the same fusion power



- Both machines produce ~ 500 MW of fusion power
- Engineering drawings are same scale



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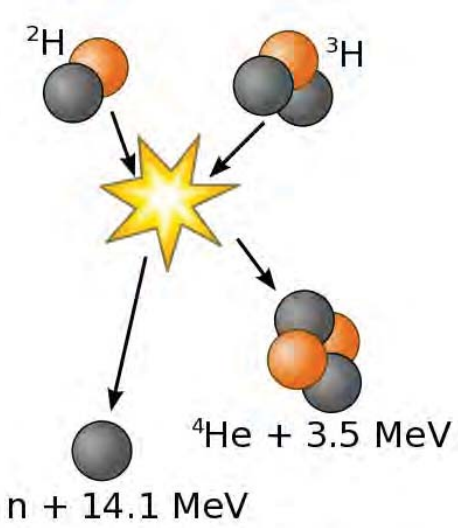
The DT reaction is most favorable for a fusion reactor

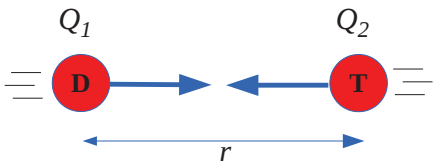


- Fusion reactions which release energy are possible for many light nuclei
- The most promising reaction is the reaction between deuterium and tritium (abbreviated DT), two isotopes of hydrogen
- The sun uses pure hydrogen fusion (really inefficient but the sun is big enough)

Nuclei must overcome Coulomb repulsion in order to fuse

- Potential energy between two charged particles is given by:

$$U = \frac{1}{4\pi\epsilon_0}\frac{Q_1 Q_2}{r}$$

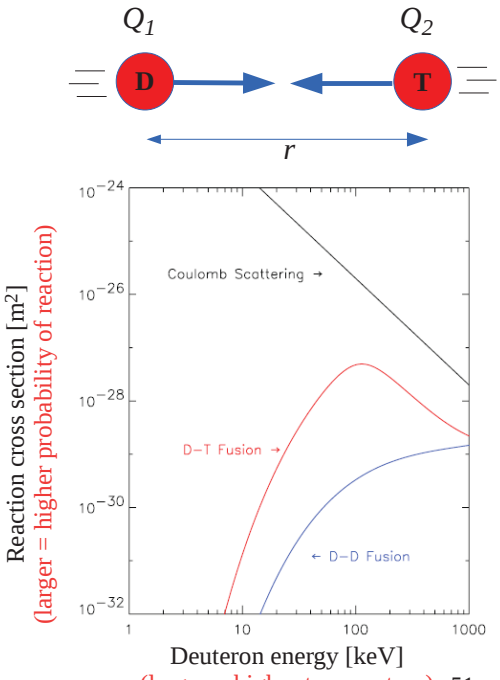


This requires particles to be moving really, really fast!

- Potential energy between two charged particles is given by:

$$U = \frac{1}{4\pi\epsilon_0}\frac{Q_1 Q_2}{r}$$

- As particles get close enough to fuse, potential energy "barrier" increases
  - Particles need to move fast → i.e. need high energy → i.e. need high heat
  - For D-T, minimum energy for fusion is 10 keV, roughly 100 million Kelvin*



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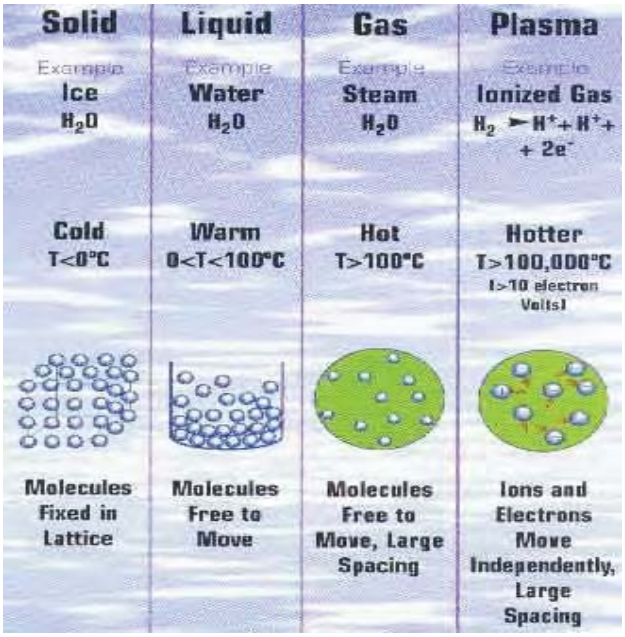
* or Celsius, it doesn't really matter at this high of a temperature...

Temperatures required for fusion necessitates confinement of plasma

- Matter at thermonuclear fusion temperatures only exists in a plasma state
- Plasma is the "fourth" state of matter where electrons are ripped off of nuclei and gas becomes ionized
- So how do you confine a superheated, charged gas?



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HYDROGEN – CARBON INTERACTIONS

A BRIEF LITERATURE SURVEY

Cristian Contescu
Tim Burchell

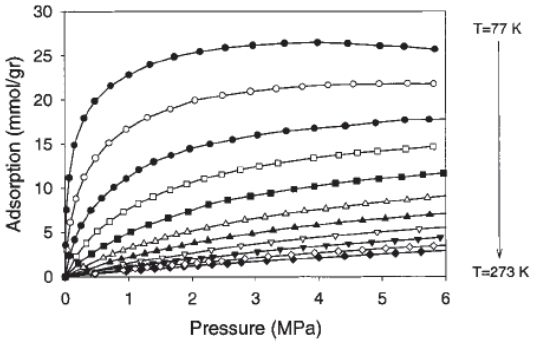
Oak Ridge National Laboratory
Materials Science and Technology Division
Oak Ridge, TN 37831 USA

Workshop on Tritium Control and Capture in Salt-Cooled and Fusion Reactors

Salt Lake City, October 27-28, 2015

Hydrogen retention mechanisms

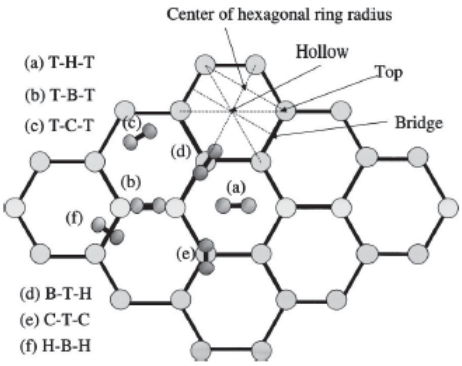
- Molecular H2 physisorption on porous carbons
  - Significant at cryogenic temperatures
  - Decreases with increase of temperature
  - Well documented from prior work on H2 storage
- Atomic H chemisorption
  - Lower saturation capacity
  - Not as much investigated as physisorption
  - Temperature dependence may be sensitive to structural factors and state of the surface
- Molecular H2 trapping / solubility
  - Mixed mechanism (molecular diffusion / chemisorption)
  - Sensitive to structural factors, irradiation



Benard, Chahine, *Langmuir* **173** (2001) 1950

Theory and modeling

- DFT calculations:
  - Dissociative chemisorption of H2 on armchair sites requires high activation energy
  - Dissociative chemisorption on zig-zag sites is easier (no activation)
  - H atom chemisorption on basal planes requires pocking of a C atom from its planar position and requires activation energy
  - The crucial step is H2 dissociation which is energetically demanding



Dino et al. Surf Sci Nanotech 2 (2004) 77; Solid State Commun 132 (2004) 713

H retention on graphite

- Retention and diffusion of H2 on isotropic graphite from Toyo Tanso
  - At 1000 C and 1 bar, H2 uptake leads to loading of up 20-600 ppm (based on pressure drop)
  - Diffusion and trapping in the graphite structure
  - Two types of trapping sites proposed (based on thermodesorption data)
  - Enhanced trapping on defective and irradiated graphite

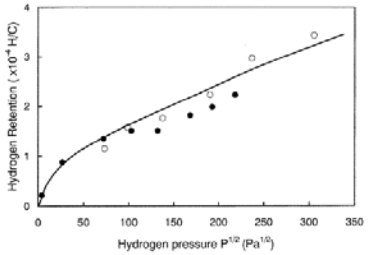


Fig. 3. Pressure dependence of hydrogen retention in graphite measured by absorption experiment (solid circles) and desorption experiment (open circles) [6] (sample: ISO-880U (absorption), ISO-88 (desorption), temperature: 1273 K) (ISO-880U is purified sample of ISO-88).

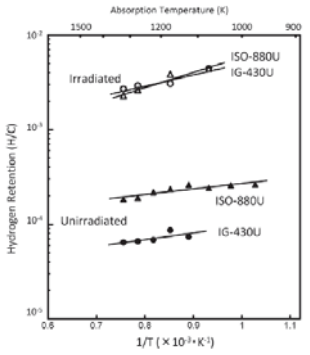


Fig. 2. Temperature dependence of hydrogen retention in graphite before and after neutron irradiation (irradiation fluence: 3.9×10^{25} n/m^2 (0.047 dpa), absorption temperature: 1273 K, equilibrium pressure: ~10 kPa).

Atsumi, Phys Scripta T103 (2003) 77; J. Alloys Compounds 356-357 (2003) 705

Trapping sites in graphite

- Two types of trapping sites
  - Dangling bonds at crystallite edges (2.6 eV)
  - Interstitial sites inside crystallites (4.4 eV)
  - The prevalence of each type of sites varies with irradiation fluence

Atsumi, J, Nucl. Mater. 313-316 (2003) 543

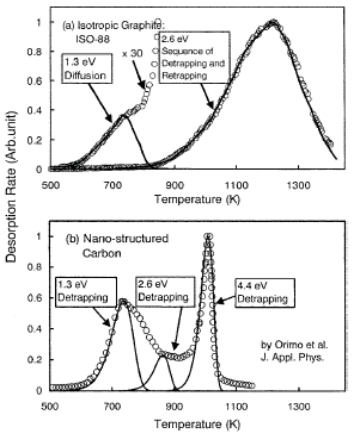


Fig. 4. Deuterium desorption spectra and the curve fittings for isotropic graphite (ISO-88: (a)) [6] and nano-structured carbon milled in hydrogen atmosphere [31] (heating rate: 10 K/min).

- Thermodesorption of H2 trapped in isotropic graphite is similar to that from graphite mechanically milled in a hydrogen atmosphere

Orimo, J Appl Phys 90 (2001) 1545

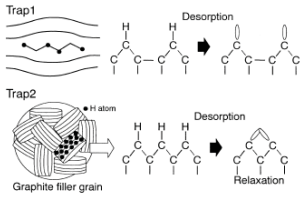


Fig. 3. Schematic illustration of the proposed model on hydrogen trapping in a graphite material. The chemical bond model was referred from Ref. [18].

- The nature of trapping sites in graphite is still debated

Miyabe, J Appl. Phys. 104 (2008) 044311

Deuterium in graphite

- D2 thermodesorption from isotropic graphite exposed to D2 at various temperatures
  - Solubility is proportional with $P^{1/2}$
  - Three main desorption peaks
  - Evidence of various trapping sites
  - Very small CD4 desorption was found

Atsumi, J, Nucl. Mater. 155-157 (1988) 241

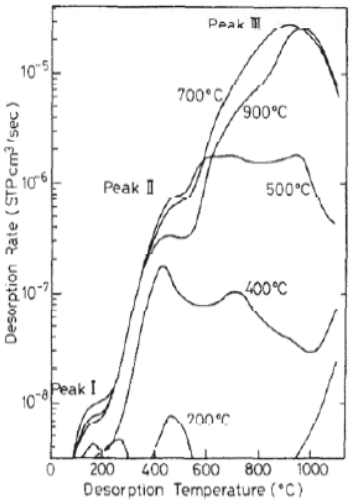


Fig. 2. Thermal desorption curves of deuterium for graphite exposed to D_2 at various temperatures (60 kPa, 5 h).

Tritium in graphite and PyC

- Diffusion and solubility in PyC
  - T2 diffusion in PyC is much lower than in metals, and activation energy is high, suggesting chemical bonding
  - D2 solubility in PyC suggests a dissociative mechanism (varies with $P^{1/2}$)
  - PyC suggested as an effective barrier for T implantation in fusion reactors

Causey, Carbon 17 (1949) 323

- Retention of D+ and T+ ions in POCO graphite
  - Plasma exposure to 100 eV ions below 500 K causes fast saturation
  - Between 500-1000 K diffusion along pore surfaces occurs deep in the sample
  - Above 1000 K the isotopes penetrate graphite and decorate high energy traps

Causey, J Vac Sci Technol 4 (1986) 1189

Thermal release of tritium

- Thermal release of T2 from irradiated graphite
  - T2 is released as HT, and also as HTO (if oxidation occurred)
  - The release temperature increases with the increase of neutron fluence received by graphite

Saeki, J Nucl Mater 99 (1981) 1019

- Thermal release of T2 implanted in fine grain isotropic graphite
  - Detrapping o implanted T starts at ~ 600 K, reaches maximum rate at 1100-1400 K, and is 95% completed at 1600 K
  - On single type of trapping sites was obserrved
  - High T retention up to high temperature suggests that graphite will retain T inventory after exposure to energetic T ions

Sawicki, J. Nucl Mater 162-164 (1989) 1019

Factors affecting H trapping

- Hydrogen trapping in neutron irradiated graphite
  - Neutron irradiation creates high energy trapping sites
  - H atoms diffusivity is 1-2 orders of magnitude lower after irradiation at only 0.047 dpa
  - Irradiation and subsequent annealing of graphite changes substantially the absorption rates

Atsumi, J Nucl Mater 386-388 (2009) 379; ibid 390-391 (2009) 581

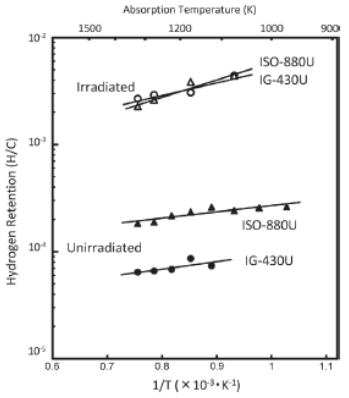


Fig. 2. Temperature dependence of hydrogen retention in graphite before and after neutron irradiation (irradiation fluence: 3.9×10^{25} n/m^2 (0.047 dpa), absorption temperature: 1273 K, equilibrium pressure: ~10 kPa).

- Hydrogen retention in graphite irradiated at high temperature
  - Irradiation causes damage in graphite structure
  - H retention is significantly reduced at high temperature

Yoshida, J. Nucl. Mater 386-388 (2009) 841

Hydrogen thermodesorption

- Thermodesorption spectroscopy from H-implanted graphite
  - Thermodesorption spectroscopy can be used to characterize the energy strength of trapping sites

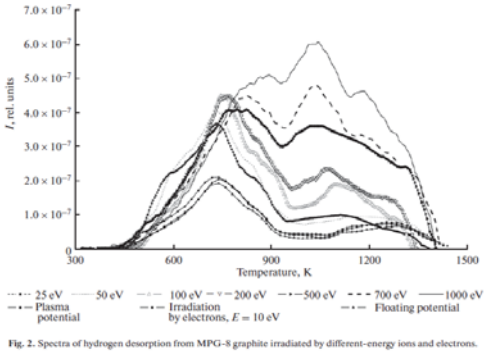


Fig. 2. Spectra of hydrogen desorption from MPG-8 graphite irradiated by different-energy ions and electrons.

Airapetov, J Surf Investigation 4 (2010) 537-571

- Modeling H2 thermodesorption from H+ implanted graphite
  - Study showed the effect of graphite porosity and crystallite size on the number and types of trapping sites

Liu, Nucl Instruments Methods Phys Rev B 269 (2011) 431

Other works

- Modeling of reactive-diffusive transport of hydrogen after ion implantation
  - Release and retention of H in porous graphite depends on the graphite internal structure and on the energy and flux of incident ion beam

Rai, J Nucl Mater 374 (2008) 304

- Hydrogen isotopes permeations through carbon materials
  - Measurements of gas pressure driven permeation of isotopes

Spitsyn, J Nucl. Mater 390-391 (2009) 701

Perspectives

- Physisorption on high surface area, porous carbons is far more efficient than chemisorption on graphite
  - Requires cryogenic temperatures
  - Can be used in conjunction with a sweep gas (He) to separate T2 from T2/He mixtures
- Retention by carbon elements in contact with the molten salt (pebbles) will be difficult to control given the irradiation effects on carbon
- Salt corrosion effects on carbon (pebbles) will change the hydrogen retention properties

AGR FUEL COMPACT DEVELOPMENT PROGRAM

TIM BURCHELL and CRISTIAN CONTESCU

METALS & CERAMICS DIVISION

PRESENTED TO TRITIUM &

MSR TECHNOLOGY WORKSHOP

Oct 27, 2015

OAK RIDGE NATIONAL LABORATORY

AGR FUEL COMPACT DEVELOPMENT

OVERVIEW OF PRESENTATION

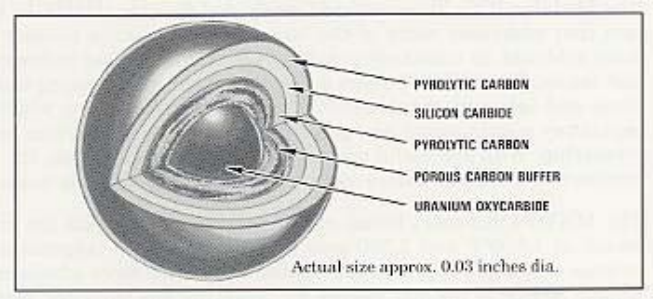
- **BACKGROUND**
  - **FUNCTION OF COMPACT**
  - **METHODS OF MANUFACTURE**
  - **ATTAINABLE FUEL PARTICLE VOLUME FRACTIONS**
  - **NEUTRON IRRADIATION DIMENSIONAL STABILITY**
- **RECOMMENDED APPROACH**
  - **METHOD & MATERIALS**

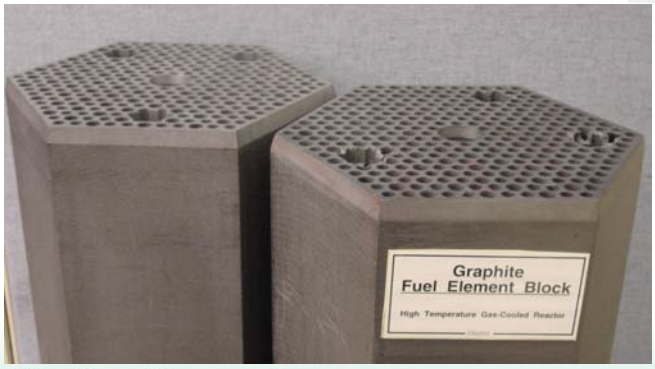
AGR FUEL COMPACT DEVELOPMENT

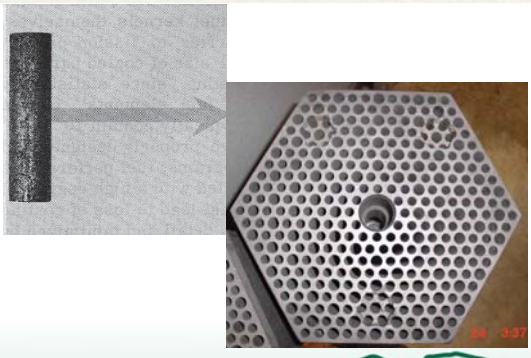
BACKGROUND

THE GAS TURBINE-MODULAR HELIUM REACTOR (GT-MHR) UTILIZES CERAMIC COATED PARTICLE FUEL

FUEL PARTICLES ARE FORMED INTO 12.5 mm DIAMETER FUEL STICKS AND INSERTED INTO GRAPHITE FUEL BLOCKS



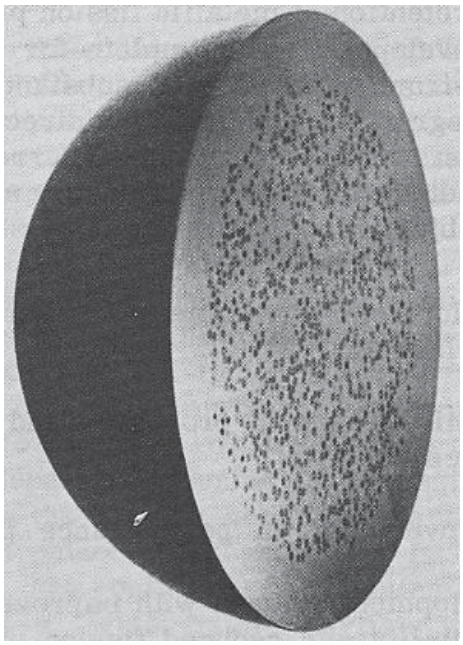




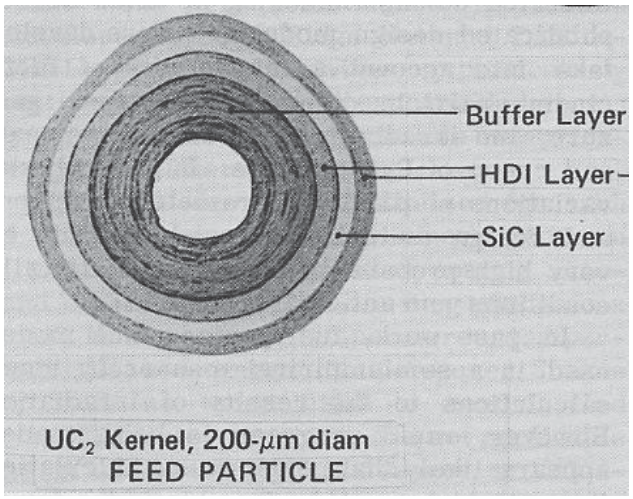
OAK RIDGE NATIONAL LABORATORY
U. S. DEPARTMENT OF ENERGY

UT-BATTELLE

THE PEBBLE BED REACTOR UTILIZES CERAMIC COATED PARTICLE FUEL



THE TRISO FUEL PARTICLES ARE COMBINED INTO A CARBON FUEL BALL (PEBBLE) 6 cm IN DIAMETER



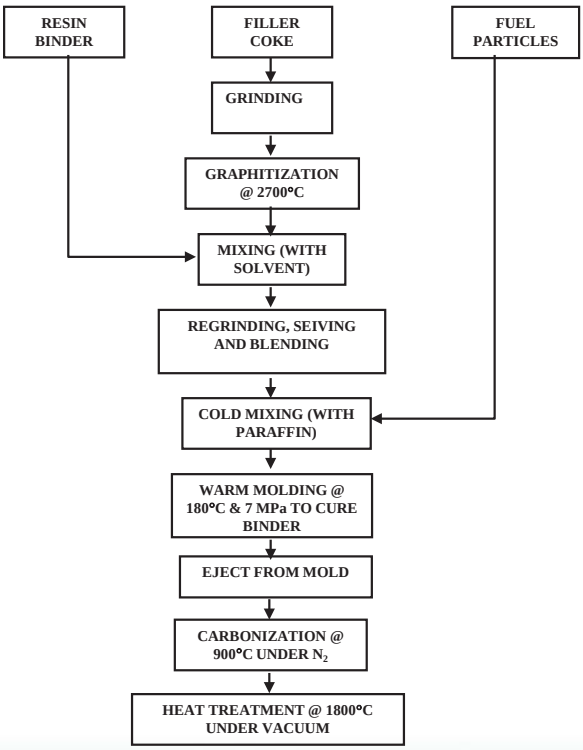
FUEL COMPACT: FUNCTION & TERMINOLOGY

- RENDERS FUEL PARTICLES INTO HANDLEABLE FORM (i.e., COMPACT OR FUEL PEBBLE)
- COMPACT COMPRISES OF FUEL PARTICLES, MATRIX AND GRAPHITE SHIM
- MATRIX CONSISTS OF FILLER (COKE OR GRAPHITE) AND BINDER (PITCH OR RESIN)
- THE MATRIX BINDS TOGETHER THE FUEL PARTICLES AND PROTECTS THEM FROM MECHANICAL DAMAGE BY FAILING PREFERENTIALLY SO AS TO AVIOD DAMAGE TO THE FUEL PARTICLE COATINGS
- ADDITION OF GRAPHITE FILLER TO THE BINDER INCREASES THE THERMAL CONDUCTIVITY OF THE FUEL COMPACT, AND INCREASES DIMENSIONAL STABILITY DURING HEAT-TREATMENT AND NEUTRON IRRADIATION
- AGR COMPACT IS 12.5 mm DIA & 49.3 mm LEN, AND HAS A FUEL PARTICLE VOLUME FRACTION IN THE RANGE 22-33.6%

AGR FUEL COMPACT DEVELOPMENT

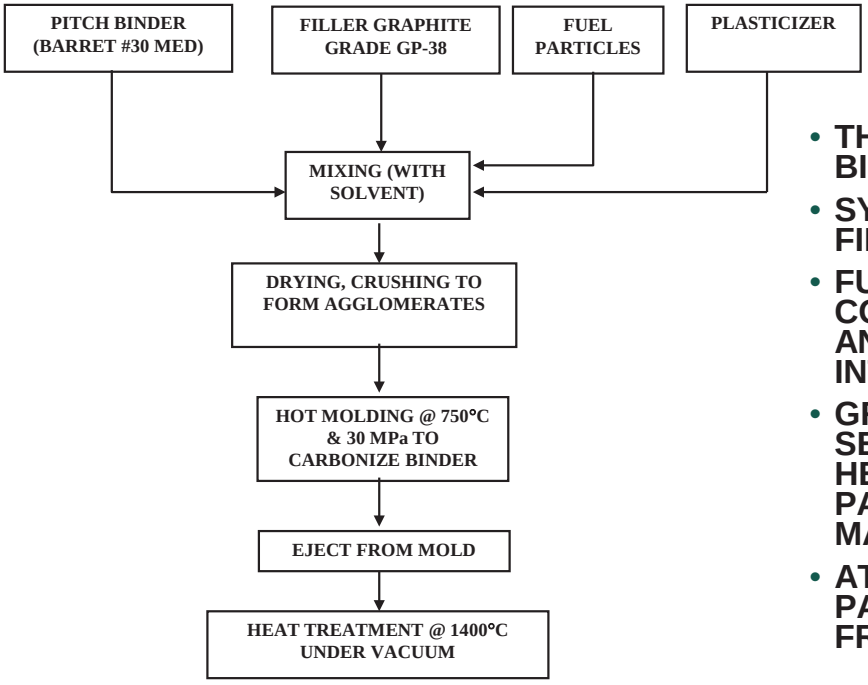
FUEL COMPACT METHODS OF MANUFACTURE

DRAGON PROJECT ADMIX METHOD



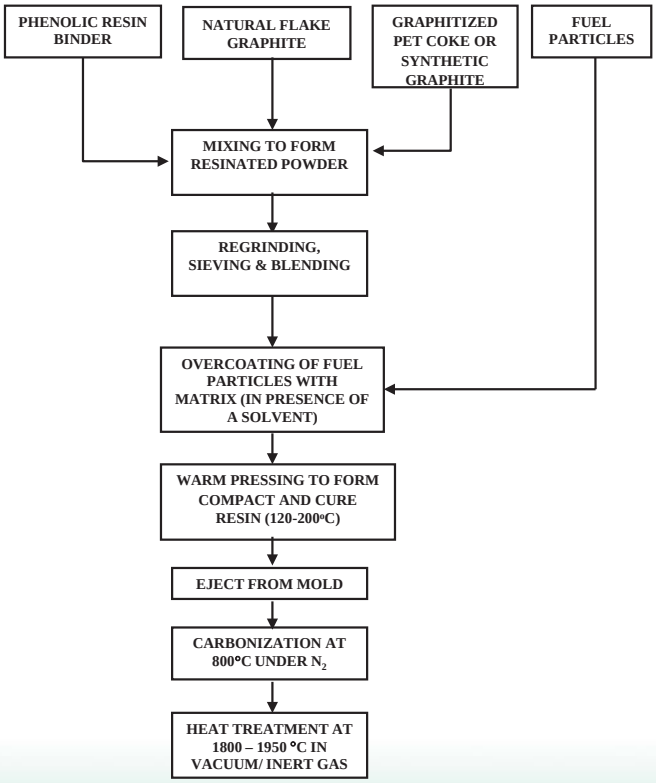
- HIGH CHAR YIELD RESIN (PHENOL FORMALDEHYDE) SOLVATED WITH ALCOHOL
- GRAPHITIZED PETROLEUM COKE (< 50 MICRON SIZE)
- PRODUCE A RESIN COATED GRANULATED POWDER (88% FILLER)
- SIEVED, BLENDED AND MIXED WITH COATED FUEL PARTICLES
- WARM MOLDED TO FORM AND CURE BINDER, & EJECTED FROM MOLD
- CARBONIZED @ 900°C & HEAT TREATED TO 1800°C
- ATTAINABLE FUEL PARTICLE VOLUME FRACTIONS TYPICALLY < 25%

PEACH BOTTOM PROCESS (GENERAL ATOMICS)



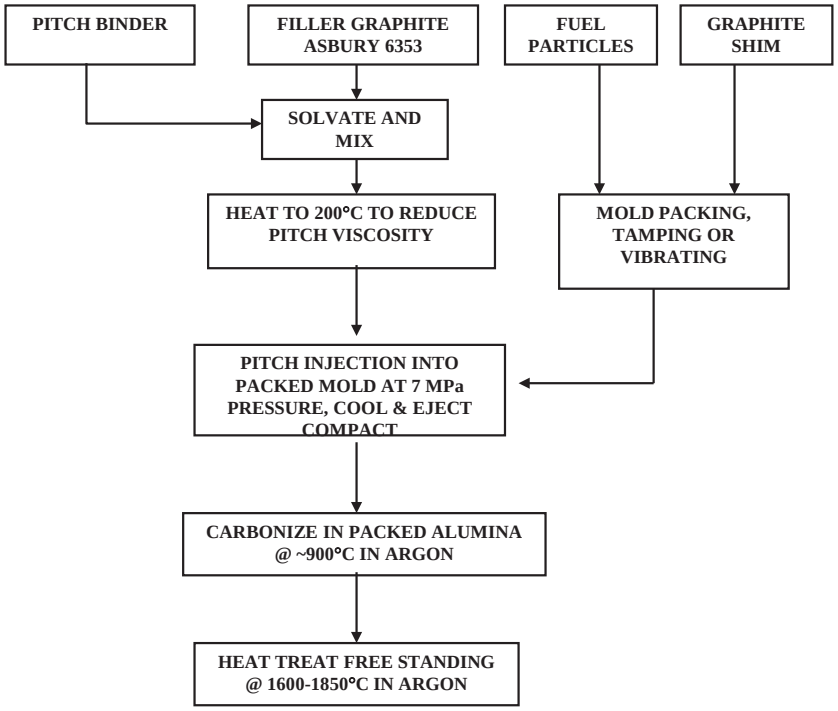
- THERMOPLASTIC (PITCH) BINDER
- SYNTHETIC GRAPHITE FILLER (90% OF MATRIX)
- FUEL AND MATRIX COMPONENTS MIXED AND THEN GRANULATED INTO 3-9 mm PELLETS
- GRANULIZATION AVIODS SEGREGATION OF THE HEAVIER FUEL PARTICLES & LIGHTER MATRIX COMPONENTS
- ATTAINABLE FUEL PARTICLE VOLUME FRACTION 25-35%

PARTICLE OVERCOATING PROCESS



- •THERMOSETTING RESIN (PHENOLIC) BINDER
- •MATRIX FILLER CONSISTS OF NATURAL FLAKE AND SYNTHETIC GRAPHITE OR GRAPHITIZED PET. COKE
- •RESINATED POWDER MATRIX MIX FORMED THE “A3” MATRIX (WITH 80 wt% GRAPHITE FILLER)
- •MATRIX MIX FED INTO ROTATING DRUM WITH FUEL PARTICLES AND SOLVENT (METHANOL) TO “OVERCOAT” THE FUEL PARTICLE
- •COMPACTS ARE WARM MOLDED TO CURE RESIN
- •FINAL HEAT TREATMENT 1800-1950°C
- •ATTAINABLE FUEL PARTICLE VOLUME FRACTION 5-50%

GENERAL ATOMICS MATRIX INJECTION PROCESS



- THERMOPLASTIC (PITCH) BINDER
- NATURAL GRAPHITE FILLER (~28 % OF MATRIX)
- CLOSE PACKED BED OF FUEL PARTICLES
- MATRIX HEATED ABOVE ITS SOFTENING POINT AND "INJECTED" INTO PARTICLE BED.
- COMPACT COOLED, EJECTED AND PACKED IN ALUMINA TO SUPPORT COMPACT DURING CARBONIZATION WHEN PITCH SOFTENS PRIOR TO PYROLYSIS
- ATTAINABLE FUEL PARTICLE VOLUME FRACTION < 60%

SUMMARY OF THE FUEL PARTICLE VOLUME FRACTIONS ATTINED FROM THE VARIOUS COMPACTING PROCESSES

FUEL COMPACTING PROCESS	FUEL PARTICLE VOLUME FRACTION (%)
ADMIX (DRAGON REACTOR)	< 25
ADMIX/AGGLOMERATE (PBR)	25-35
PARTICLE OVERCOATING (DRAGON, AVR, THTR, HTTR, HTR-10)	5-50
PITCH INJECTION (FSV)	< 60

AGR COMPACT FUEL PARTICLE VOL. FRACTION TARGET IS 22-33.6%

NEUTRON IRRADIATION DIMENSIONAL STABILITY OF COMPACTS

- THE FUEL COMPACT MATRIX MATERIAL SUSTAINS A SIGNIFICANT AMOUNT OF NEUTRON INDUCED DISPLACEMENT DAMAGE
- IRRADIATION BEHAVIOR OF CARBONS & GRAPHITES MARKEDLY AFFECETED BY THE DEGREE OF CRYSTALINITY OF THE MATERIAL
- AGR COMPACTS WILL HAVE A HIGH MATRIX CONTENT SO THE IRRADIATION BEHAVIOR OF THE MATRIX IS CRITICAL
- IT HAS BEEN ARGUED THAT PITCH PRECURSERS ARE MORE SUITED FOR BINDERS SINCE FOR A GIVEN FINAL HTT THEY ARE MORE CRYSTALINE THAN RESIN CHARS, ALTHOUGH PITCH IS A MAJOR SOURCE OF CHEMICAL CONTAMINATION
- LOADING THE THERMOSETTING RESIN (GLASSY CARBON) WITH A LARGE FRACTION OF HIGHLY GRAPHITIC FILLER MARKEDLY IMPROVES THE MATRIX IRRADIATIOIN BEHAVIOR, REDUCES THERMAL SHRINKAGE ON PYROLYSIS, AND INCREASES MATRIX THERMAL CONDUCTIVITY

AGR FUEL COMPACT DEVELOPMENT

RECOMMENDED APPROACH

THE FOLLOWING FACTORS MUST BE CONSIDERED IN RECOMMENDING AN APPROACH

1. A thermosetting resin binder has been selected for the production of AGR fuel compacts
2. The required fuel particle volume fraction for the AGR compacts is very modest (22-33.6%) and is within the attainable range of the admix/agglomerate, the overcoating, or injection processes.
3. The most stable matrix is one with a large fraction of graphite filler.
4. Highly filled (>40 vol.%) pitch or resin matrix materials cannot be injected into packed particle beds.
5. Injectable low graphite filler content thermosetting resin binder formulations with additions of low char yield (fugitive) resin (e.g., polystyrene) were developed, but were never adopted for the manufacture of large quantities of fuel compacts.
6. The overcoating method with resin binder was used for the manufacture of fuel compacts for the Dragon and HTTR, and for fuel pebbles for the AVR, THTR and HTR-10.

AGR FUEL COMPACT DEVELOPMENT

BASED UPON THE FORGOING DISCUSSION IT IS RECOMMENDED THAT THE PARTICLE OVERCOATING PROCESS BE ADOPTED AS THE REFERENCE METHOD FOR THE FABRICATION OF AGR FUEL COMPACTS

TMSR Project in China

Wei LIU， Zhimin DAI， Hongjie XU ，

TMSR Center，CAS

2015-10-27

SINAP 中国科学院上海应用物理研究所
Shanghai Institute of Applied Physics, Chinese Academy of Sciences

SINAP 中国科学院上海应用物理研究所
Shanghai Institute of Applied Physics, Chinese Academy of Sciences

Outline

1. Project Overview
2. Research Progress

1. Project Overview

- **January, 2011, Chinese Academy of Sciences (CAS) initiated the "Thorium Molten Salt Reactor Nuclear Energy System"(TMSR) project.**
- **August, 2013, TMSR has been chosed as one of the National-Energy Major R&D projects of Chinese National Energy Administration (CNEA)**
- **2014, Shanghai Local Government plans to start a major new-Energy project to support the TMSR project, including the manufacture of the special materials, devices, building&utilities.**

中国科学院上海应用物理研究所
Shanghai Institute of Applied Physics, Chinese Academy of Sciences

The Aims of TMSR Project

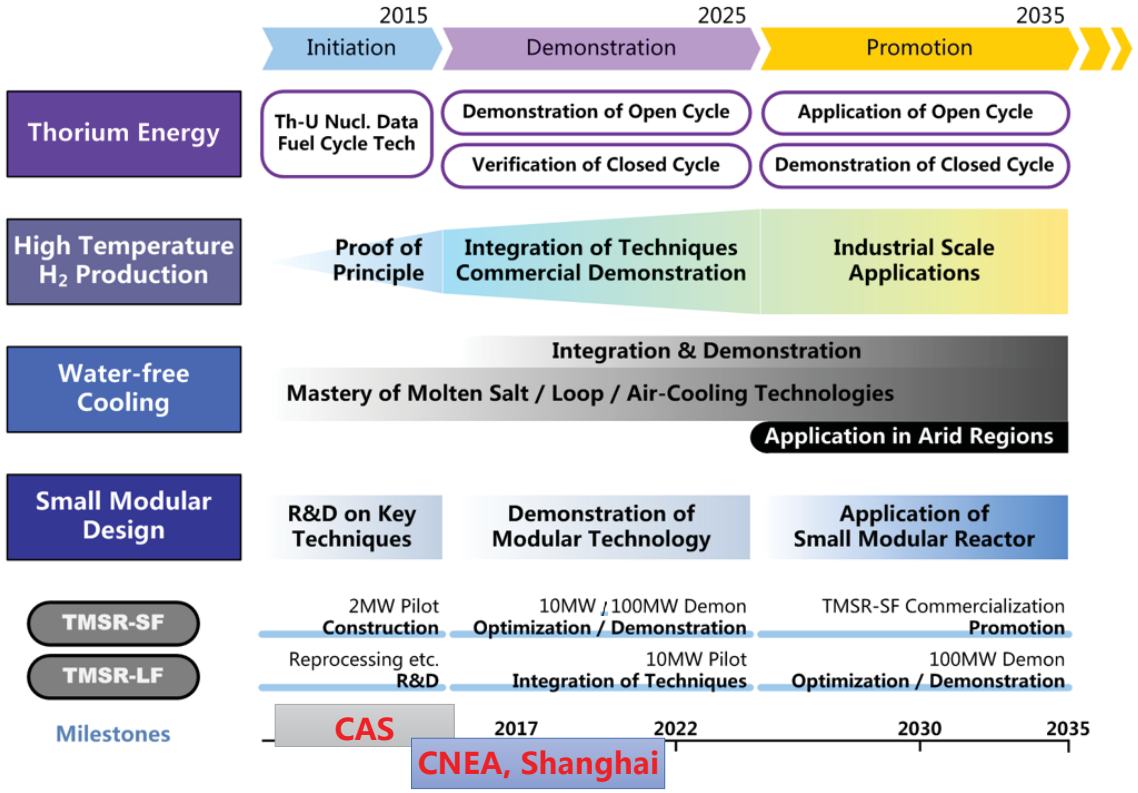
- The Aims of TMSR Project is to develop Th-Energy, Non-electric application of Nuclear Energy based on Liquid-Fuel TMSR and Solid-Fuel TMSR during coming 20-30 years.

  - Liquid-Fuel TMSR (TMSR-LF)--- MSRs
  - Solid-Fuel TMSR (TMSR-SF1)--- FHR s

TMSR-SF: Optimized for high-temperature based hybrid nuclear energy application (Non-electric application).

TMSR-LF: Optimized for utilization of Thorium.

TMSR Road Map



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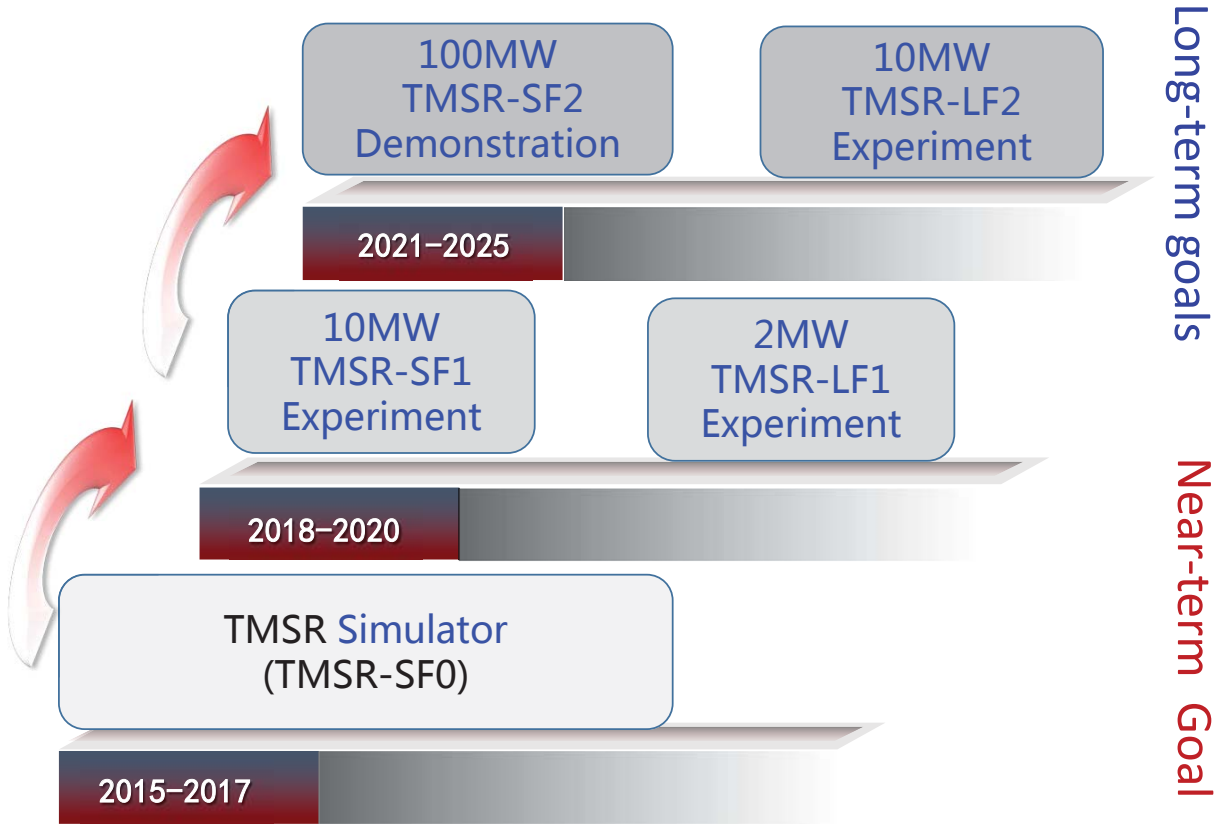
Shanghai Institute of Applied Physics, Chinese Academy of Sciences

TMSR Goal



TMSR Near-term Goal

Phase-I (~2017)

- Completion of TMSR-SF simulator.
- Start to construct 10MW TMSR-SF test Reactor (TMSR-SF1), 2MW TMSR-LF test Reactor (TMSR-LF1) & Pyro-Process Facility (PDF).
- Build up full capability of non-radioactive laboratories in Jiading.

Phase II-(~2020)

- Completion of the TMSR-SF1, TMSR-LF1 and PDF,
- Completion of the engineering design of 100MW TMSR-SF demonstration reactor (TMSR-SF2).
- Build up R&D abilities for future TMSR deveopment , including the TMSR Nuclear Park in DaFeng

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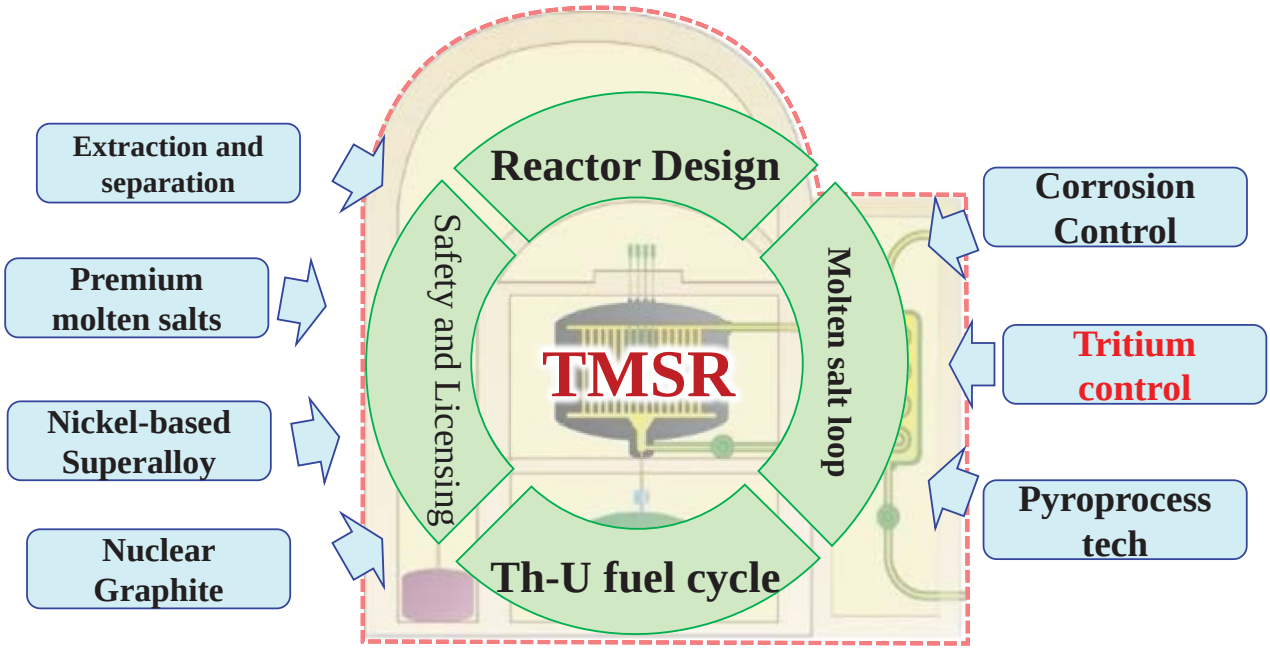
Shanghai Institute of Applied Physics, Chinese Academy of Sciences

Outline

1. Project Overview
2. Research Progress

4 Prototype Systems and 7 key technologies



Key technology : Tritium control

Master the key technologies of tritium control in the molten salt reactor such as tritium extraction with bubbling, tritium separation, tritium monitoring and son on .

(6) Tritium Monitoring

(1) Tritium Behavior → (2) Tritium Extraction → (3) Tritium Separation → (4) Tritium Storage → (5) Tritium Sampling

Chemical forms of Tritium in TMSR

Interaction between tritium with either of coolant or metal.

Extraction of tritium from the molten salt

Separation of tritium from other gaseous mixture

tritium storage with alloys after the recovery of tritium in TMSR

Sampling tritium with different chemical forms

TMSR International Cooperation

- Th Utilization, Reactor Tech.
- Material、Molten Salt Tech,
- Pyro-process
- Nuclear Safety Standards

Organizational Overview
The Chinese Academy of Sciences (CAS) and U.S. Department of Energy (DOE)
Nuclear Energy Cooperation Memorandum of Understanding (MOU)

MOU Executive Committee Co-Chairs
China – Mianheng Jiang (CAS)
U.S. – Pete Lyons (DOE)

GEN IV International Forum

Australia

ansto
Nuclear-based science benefiting all Australians

Future
- Russian
- EU
- Korea
- Japan

Collaboration with USA

FHR technology
Pyro-process

US DOE Lab

FHR (solid fuel)
Safety Standards
ANSI/ANS-20.1

High-tem. Materail
Standars (ASME)

America Society

US Industry

FHR technology

US University

CAS-DOE MOU on Nuclear Energy

Collaboration with USA-II

TMSR—ORNL FHR CRADA, Signed in July 2014

TMSR—MIT FHR MRA, signed in April 2015

Started to discuss the CRADA on Pyro-process between TMSR and INL & ANL.

Cooperation Workshop for R&D of Pyro-process Technology will be held in SINAP, 14-15, May, 2015. Stephen Kung, DOE; K. Michael Goff, INL & Mark A. Williamson, ANL will attend the workshop.

Tritium and Chemistry Management for the Mark-1 PB-FHR

Workshop on Tritium Control and Capture in Salt-Cooled Fission and Fusion Reactors: Experiments, Models, and Benchmarking

Salt Lake City

October 27, 2015

Michael Laufer

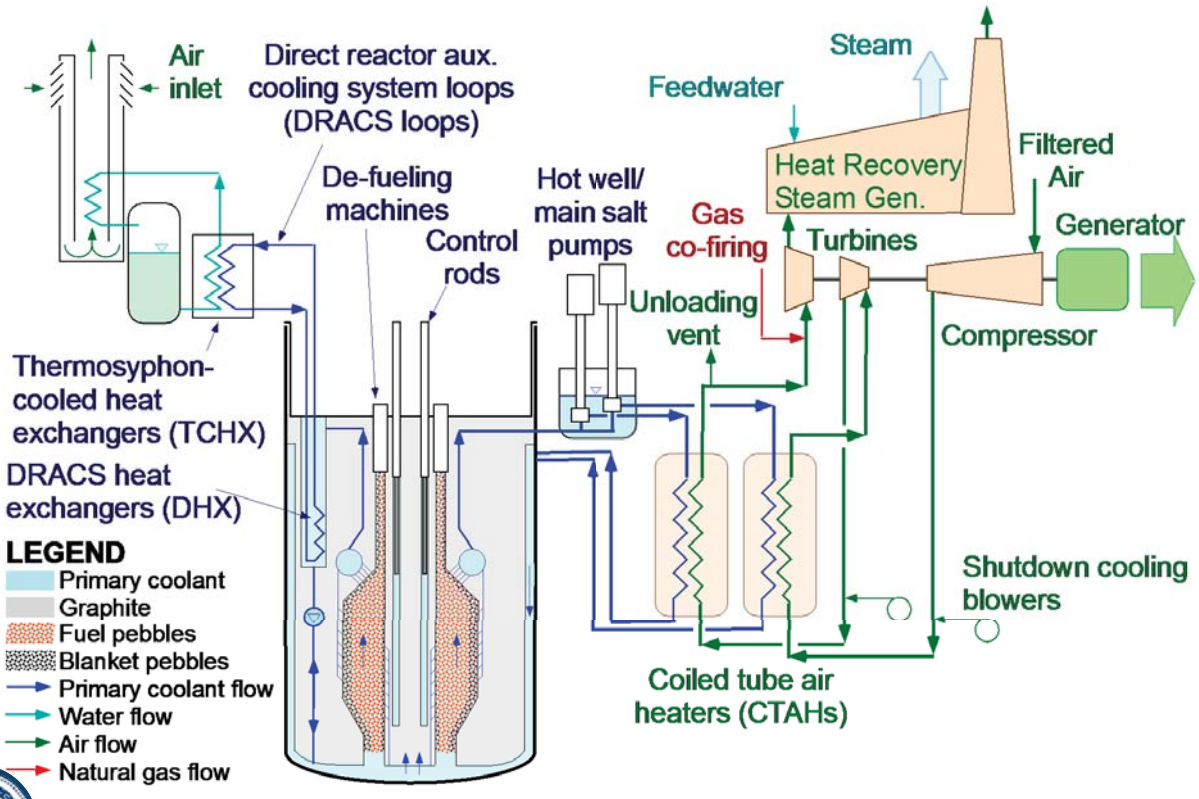
U.C. Berkeley

FHR Fluoride Salt Cooled High Temperature Reactor

NEUP Nuclear Energy University Programs

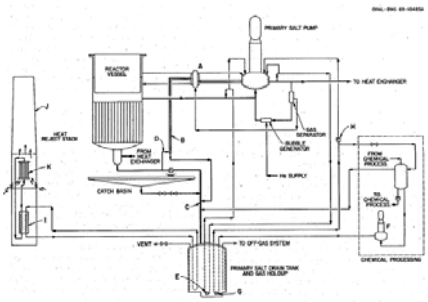
U.S. Department of Energy

Mk1 PB-FHR flow schematic



The recent UC Berkeley Mk1 PB-FHR design effort had 4 goals

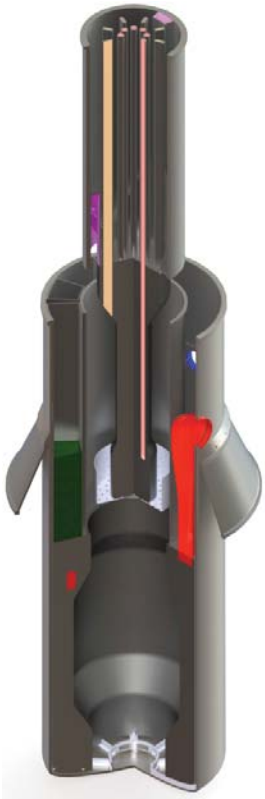
- Demonstrate a plausible, self-consistent Nuclear Air Combined Cycle (NACC) system design
  - Believable predictions for base-load and peaking power levels using an industry-standard design code (Thermoflex)
    - » 2 archival articles now published in the *ASME Journal of Engineering for Gas Turbines and Power*
  - Self-consistent approach to heat air directly with primary coolant
- Provide detailed design for decay heat management systems
  - Provide basis for establishing CIET experiment test matrix
  - Enable TH code validation and benchmarking exercises



MSBR drain tank cooling system

The Mk1 PB-FHR design had 4 goals (con't)

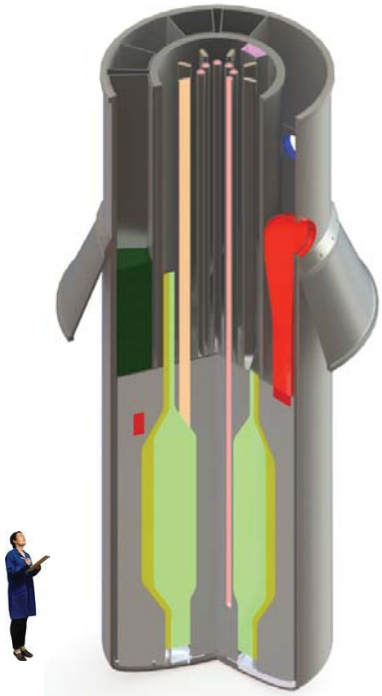
- Develop a credible, detailed annular FHR pebble core design
  - Inner and outer graphite reflector including assembly method
  - Pebble injection and defueling
  - Coolant flow distribution and pressure loss calculations
  - Provide basis for future FHR code benchmarking
  - Neutronics/depletion/control-rod worth calculations are documented in A.T. Cisneros doctoral dissertation
- Identify additional systems and develop notional reactor building arrangement
  - "Black-box" level of design for many of these systems
  - Includes beryllium and tritium management, and chemistry control strategies



Nominal Mk1 PB-FHR Design Parameters

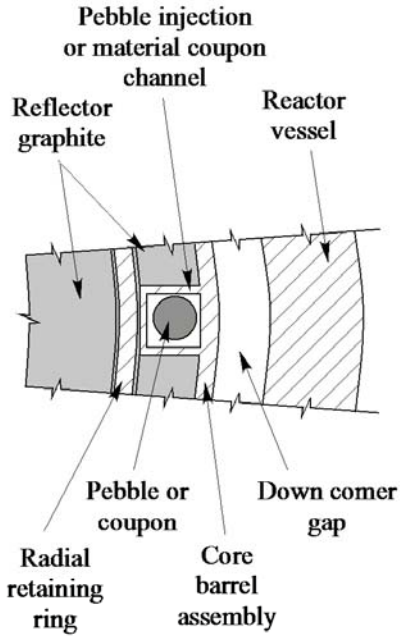
- Annular pebble bed core with center reflector (600/700° C Core Inlet/Outlet)
- Reactor vessel 3.5-m OD, 12.0-m high
- Power level: 236 MWth, 100 MWe (base load), 242 MWe (peak w/ gas co-fire)
- Power conversion: GE 7FB gas turbine w/ 3-pressure HRSG
- Air heaters: Two 3.5-m OD, 10.0-m high CTAHs, direct heating
- Tritium control and recovery
  - Recovery: Absorption in fuel and blanket pebbles and additional graphite media
  - Control: Diffusion barrier coating on air side of CTAHs

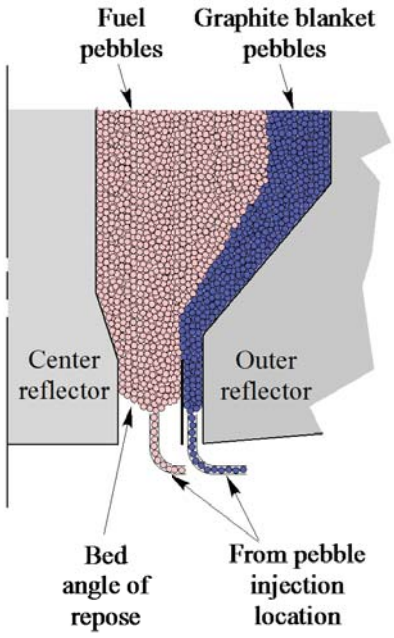
Equilibrium Tritium Production
~0.07 g (670 Ci) per EFPD
99.9% Target Recovery Rate



PB-FHR cross section

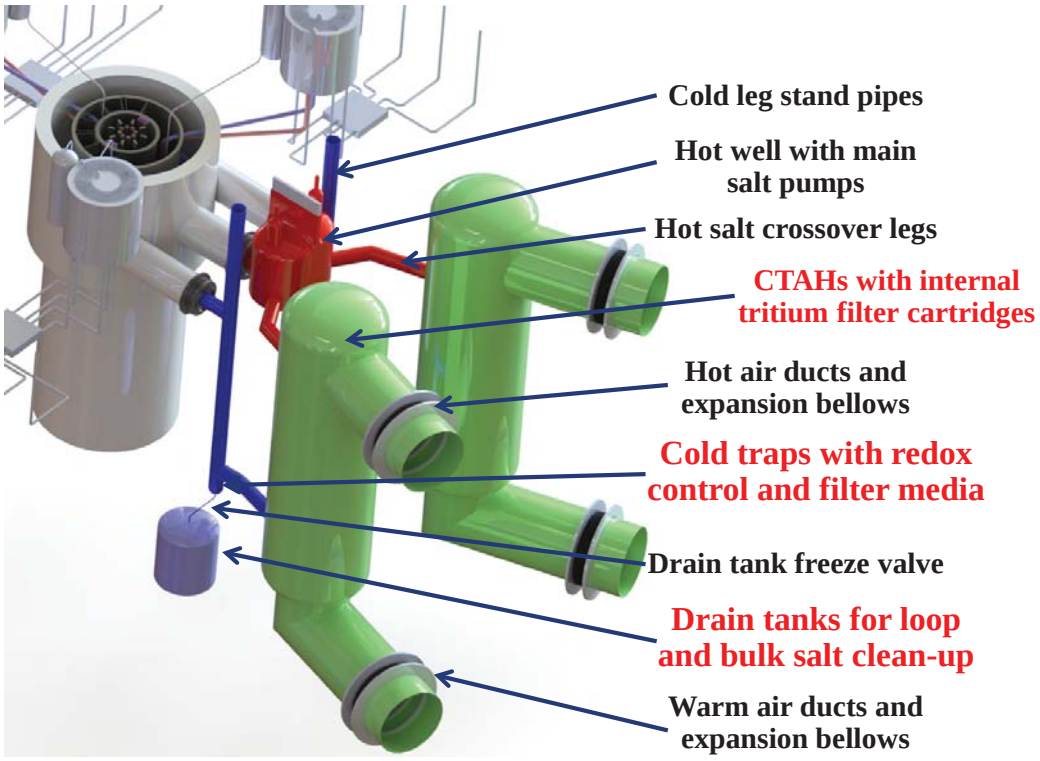
Mk1 pebble injection feeds pebbles to the bottom of the core at a controlled rate



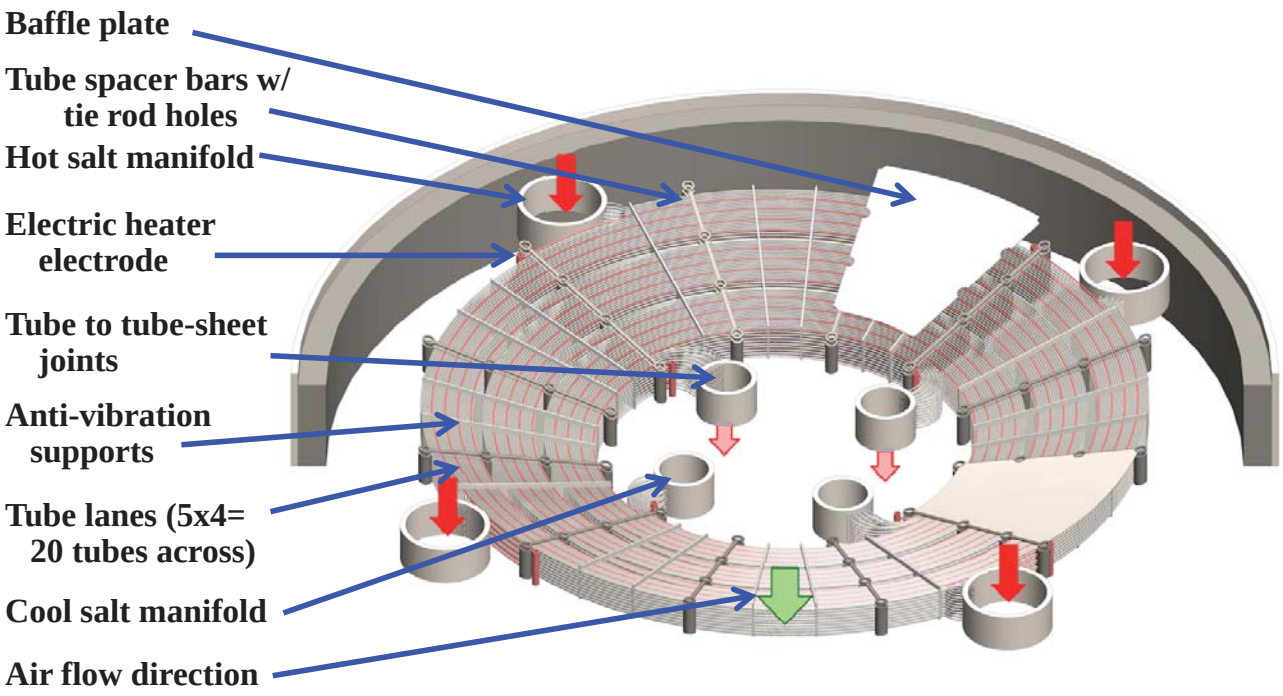


Mk1 blanket and fuel pebbles are expected to provide an important sink for tritium

The Mk1 heat transport system delivers heated salt to the two CTAHs

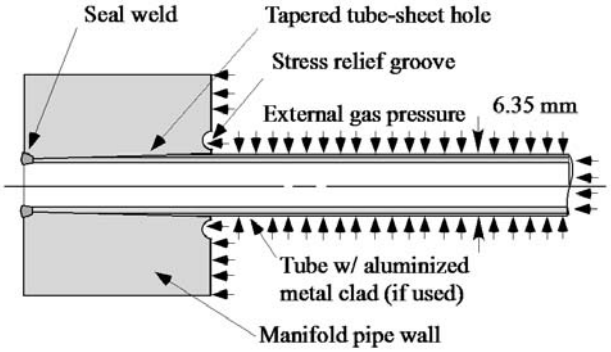


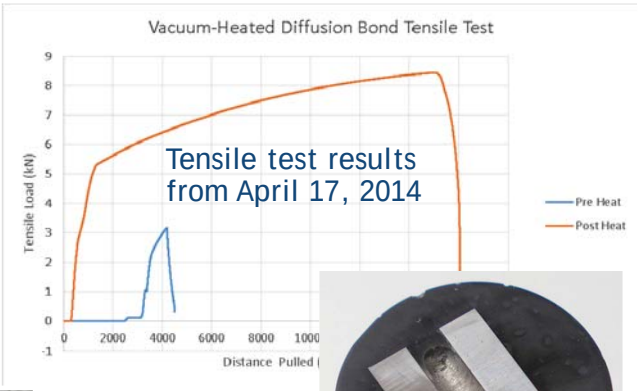
Mk1 CTAHs have 36 annular sub-bundles

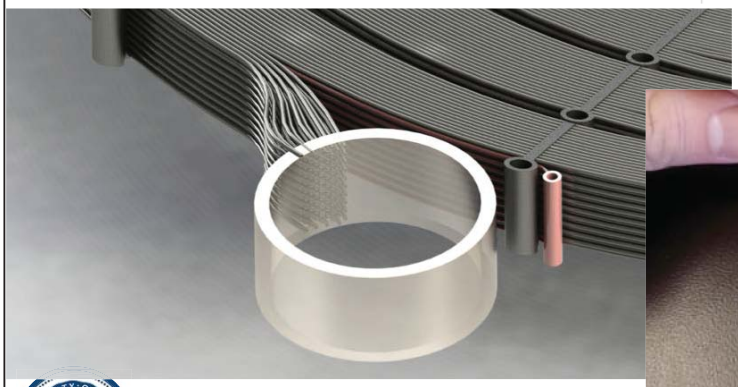


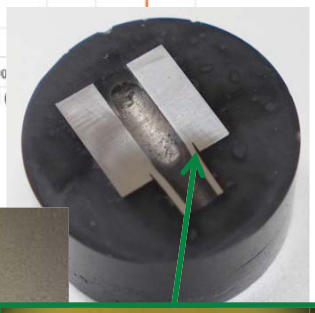
Mk1 CTAH Tube Sub-bundle Model

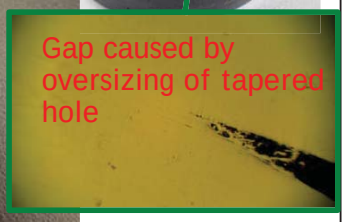
Tapered Mk1 CTAH tube to tube-sheet joints allow use of alumina-forming coating





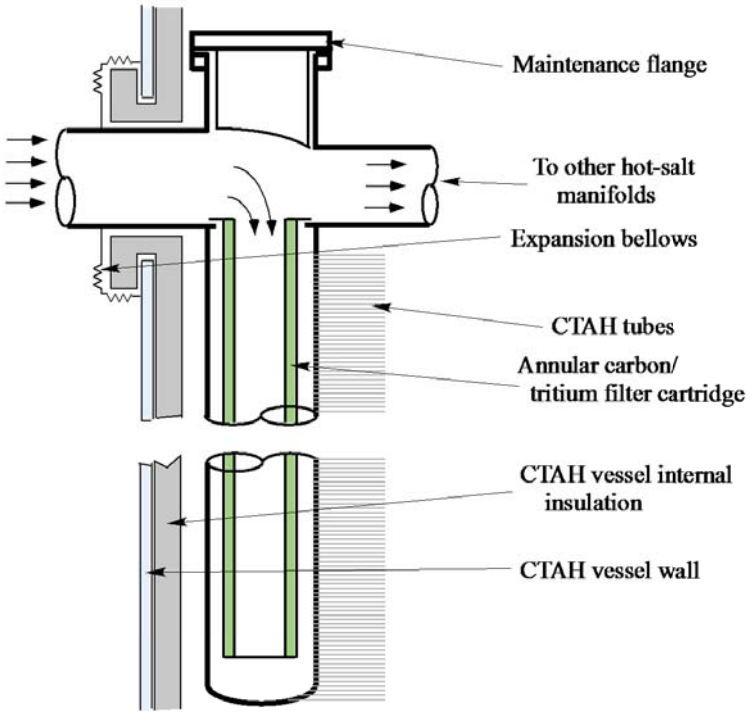






Mk1 CTAH hot leg stand pipes allow the use of an annular filter cartridge.

- 100% of hot salt flow can be treated before it enters the CTAH tubes
- Solid filter media can be used to remove tritium if absorption in fuel and blanket pebbles is not sufficient



Cold spray coating provides a scalable method to apply tritium diffusion barriers to tubes

- Cold spray coating uses a supersonic jet to deposit particles onto surfaces
  - Process performed at ambient pressure, so can be used for mass production of coated tubes and other primary coolant boundary external surfaces
  - Alumina forming compounds can be applied as well as other diffusion barriers
- Many possible alumina forming compounds are possible
  - Ti_2AlC ceramic has been demonstrated by UW Madison
  - Many ductile coating materials are also candidates
    - » Kanthal (Al_2O_3 forming alloy)
    - » 316SS or Alloy N alloyed with a few percent aluminum
    - » Many others

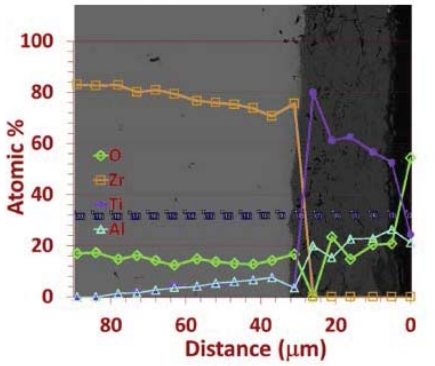
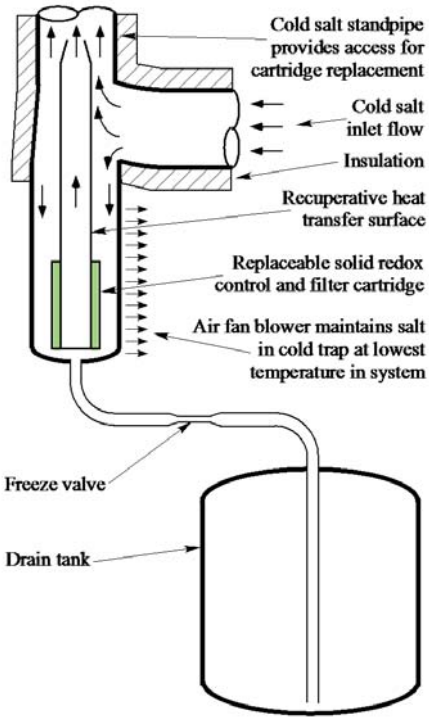


Fig. 11. Cross-sectional SEM image of Ti_2AlC MAX phase coated Zry-4 after simulated LOCA testing at 1005 °C for 20 min in an Ar/steam environment followed by quenching in boiling water. The EDS line scan is also overlaid on the SEM image.

UW Madison has demonstrated cold spray coating of Ti_2AlC on zirconium cladding

Mk1 Cold Traps and Drain Tanks aid coolant chemistry control

- Cold trap filters oxides and other contaminants that precipitate at low temperature
- Cold trap also provides location to continuously contact salt with reducing agent (if used)
- Drain tank allows CTAHs to be drained for inspection/maintenance
- Drain tank provides volume to perform bulk salt clean up (e.g., HF/H_2 sparging) if needed during shutdown and maintenance.



Questions?

Savannah River National Laboratory™
OPERATED BY SAVANNAH RIVER NUCLEAR SOLUTIONS

We put science to work.™

An Overview of SRNL Tritium Activities

Greg Staack

Hydrogen Processing Group

Workshop on Tritium Control and Capture
October 27-28, 2015

Savannah River NUCLEAR SOLUTIONS™
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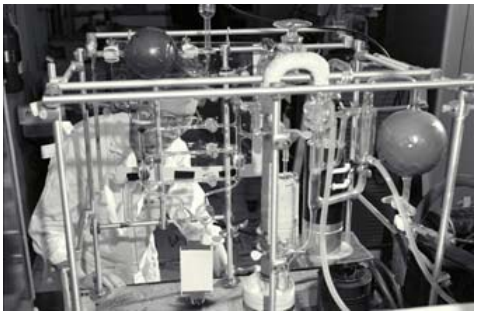
Savannah River Site

Savannah River Site - Production Years



SRS produced about 36 metric tons of plutonium from 1953-1988

- Began operation in 1953
- Produce and recover nuclear materials
  - Tritium
  - U, Pu
  - Cf, Np, Cm, Am,…
- Facilities
  - Heavy water extraction plant
  - Nuclear fuel and target fabrication facility
  - Five reactors
  - Two chemical separations plants
  - Naval Fuels
  - Waste management facilities
  - Tritium

Savannah River Tritium Enterprise Mission

Fill Tritium Reservoirs in support of the Nation's Nuclear Stockpile

- Extract new tritium from TPBARs irradiated in TVA reactors
- Recycle gas from returned bottles
  - Receive
  - Unload
  - Reprocess
  - Mix
  - Load
  - Ship
- Minimize environmental impact
- Provide a SAFE working environment
- One objective of the SRNL Hydrogen Processing Group is to provide technical support to the Facilities

Overview – Tritium Processing Research and Development Focus Areas

Remove impurities

- Impurity Removal
  - Hydrogen Separation from Impurities
  - He-3 cleanup

Separate hydrogen isotopes and Store

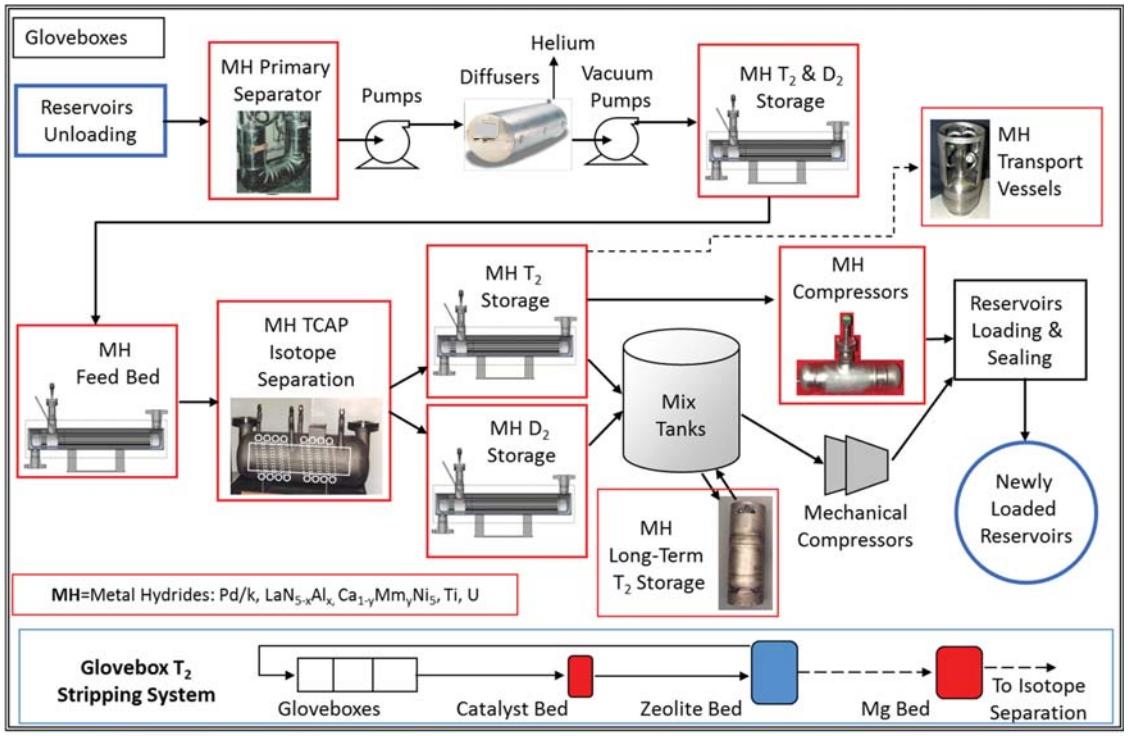
- Storage (Metal Hydride Beds / Tanks)
  - Materials options based on temperature/pressure/purity requirements
  - In-bed calorimetry
- Hydrogen Isotope Separation
  - Thermal Cycling Absorption Process (TCAP)

Clean up tritium-containing gases

- Secondary Confinement
- Tritiated Water Processing / Detritiation Systems
  - Glovebox Atmosphere Detritiation (Stripper Systems)
  - Tritium Recovery from water (Z-Bed Recovery)
  - Detritiation of process effluent gases (TPS)
- Pumping (Evacuation/Circulation)
- Tritium Effects on materials

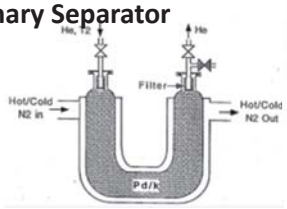
SRNL R&D provided current metal hydride-based Tritium Processes. Current research efforts are underway to improve efficiency, decrease operation costs, and to modularize processes

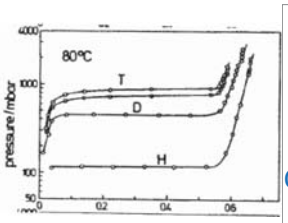
Tritium Processing at SRS – The Largest MH Based Facility in the World

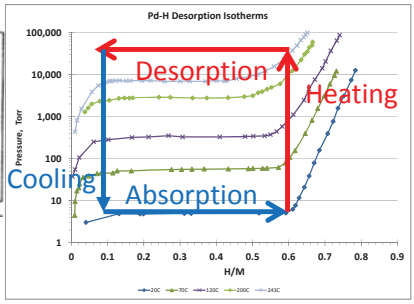


Impurity Removal by Bulk Separation

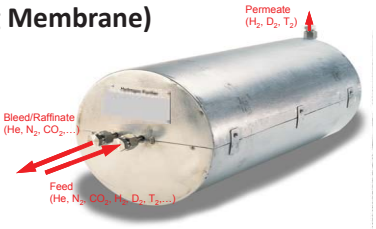
Flow-Through Bed/ Primary Separator

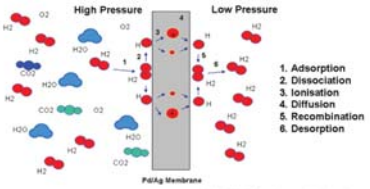


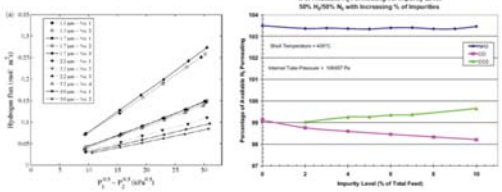




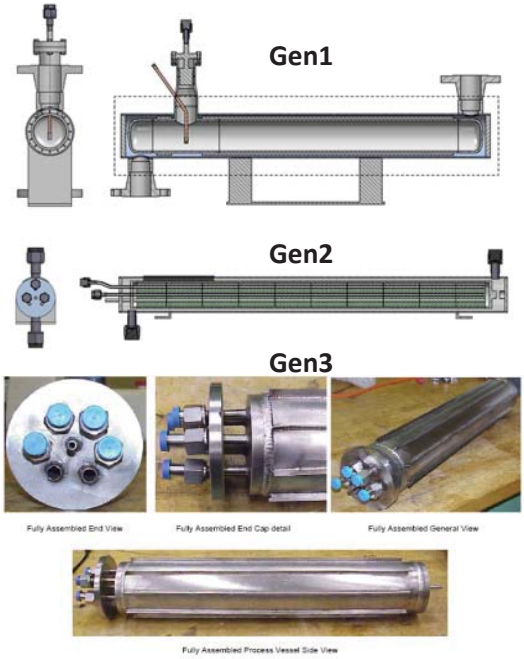
Diffuser/Permeator (Pd-Ag Membrane)







Tritium Storage: La-Ni-Al Metal Hydride for "He-3 Free" Gas Delivery

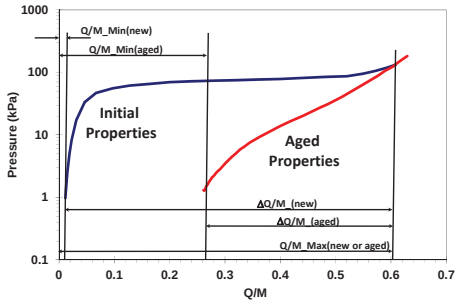


LANA properties change with tritium aging.

- Formation of "heel"
- Decreased plateau pressure
- Eventual loss of plateau
- Eventual weeping of He-3

Limited life components

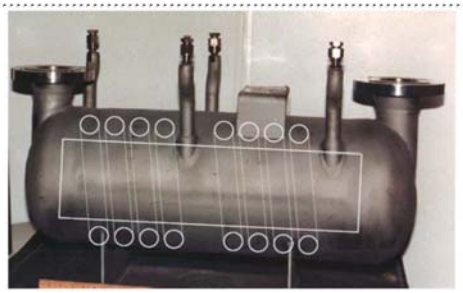
He-3 recovery from retired beds is being pursued.



Three Generations of Metal Hydride Storage Bed Development

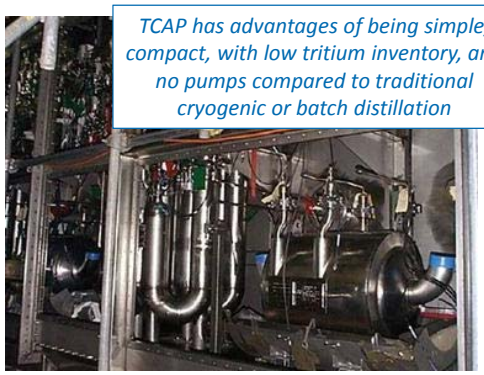
Thermal Cycling Absorption Process - TCAP

- "Heart" of the Tritium Facilities
- 4th generation of isotope separation at SRS
  - *Thermal Diffusion (1955-1986)*
  - *Fractional Absorption (1964-1968)*
  - *Cryogenic Distillation (1967- 2004)*
- Semi continuous TCAP invented at SRS in 1981. Originally coupled with a Plug Flow Reverser (PFR).
- Uses Pd/k to separate hydrogen isotopes
  - *Recover tritium (T_2)*
  - *Stack protium (H_2)*
- Hot and cold N_2 for thermal swing
- Feed, product, and raffinate gases stored on LANA beds



Original plant TCAP (1994)

TCAP has advantages of being simple, compact, with low tritium inventory, and no pumps compared to traditional cryogenic or batch distillation



HT-TCAP installed in Tritium Facility (2004)

Advances in Hydrogen Isotope Separation Technology

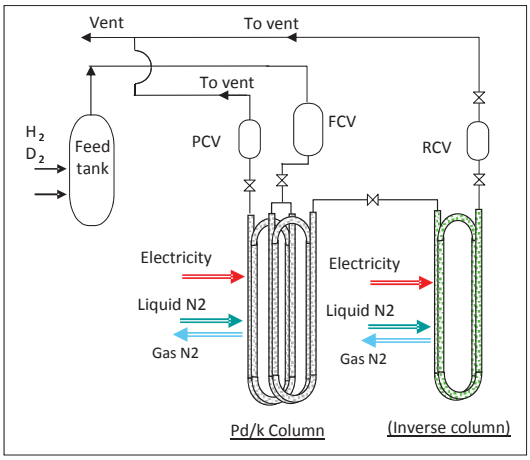
TCAP (Thermal Cycling Absorption Process) advanced from

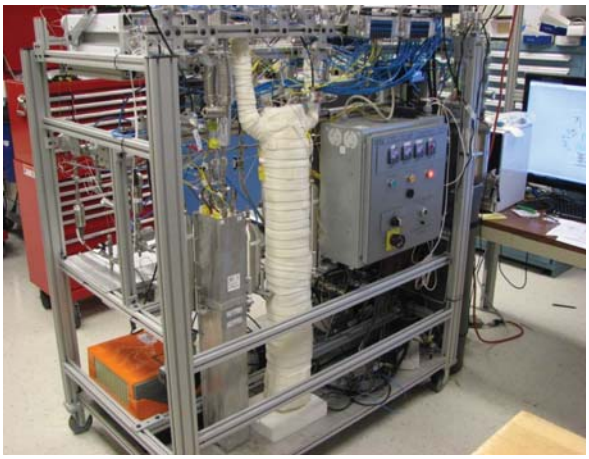
"Pd/k – passive Plug Flow Reverser" to "Pd/k – Active Inverse Column"

Results: Increased throughput and improved product purity

Thermal Cycling with Hot and Cold N_2 to Electric Heating and once through LN_2 Cooling

Results: Decreased cycle time

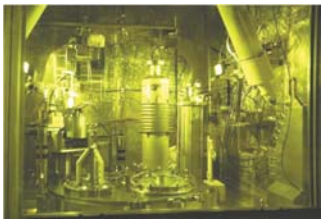


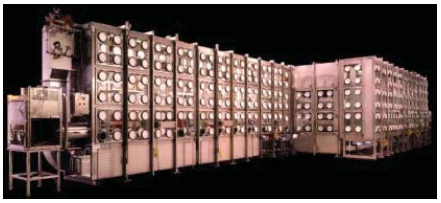


Glovebox Confinement Systems with Tritium Stripping

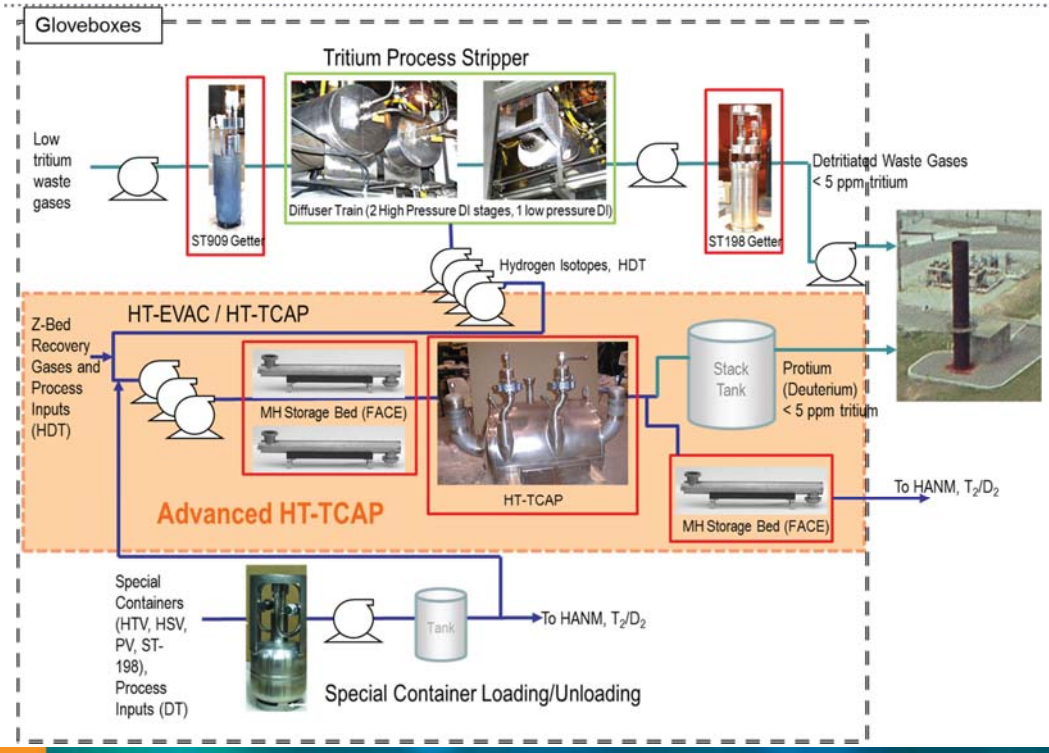
Glovebox/Secondary Confinement vs Fresh Air Hoods

- Legacy facilities used once through fresh air hoods – allowed product to escape
- In order to stem releases, closed loop glovebox system employed (RTF & TEF)
- Equipped with a secondary stripper for high activity
- Emissions were reduced from 100's kCi/year average (60's – 80's) to 10's of kCi/year average now
- Closed glovebox cleanup loop requires processing of byproducts – impact not fully realized during the early days of RTF (mid 90's)





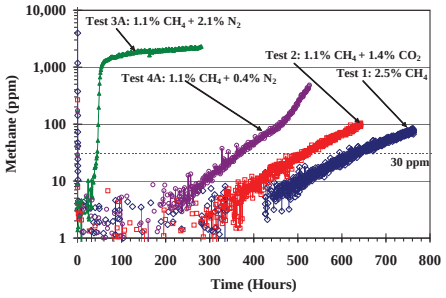
H-Area New Manufacturing – Tritiated Waste Gas Processing



Tritium Process Impurity Removal

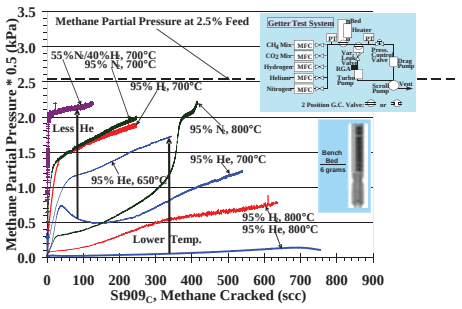
Molecular Sieve

- Large temperature swings
- May require upstream catalyst (e.g. water processing)
- Typically desorbed to regenerate bed, transferring impurities to a different process stream (Z-Bed Recovery or He-3 cleanup)



Non-Evaporative Getters

- Selected to "getter" various species, with or without decomposition
- Usually at elevated temperatures
- St909 crack methane, absorb carbon (not regenerated)
- St198 absorb tritium (regenerated)



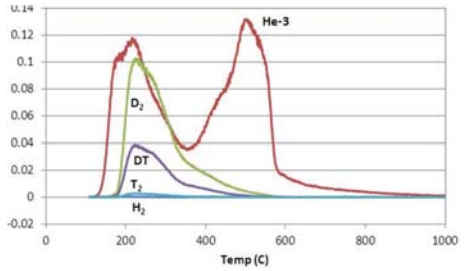
Tritium Effects on Materials –Hydrides & Polymers

Hydrides:

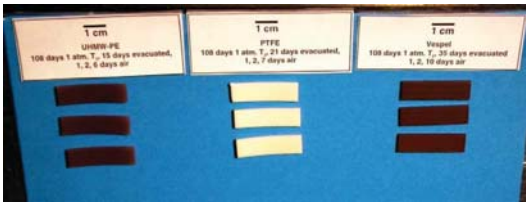
- Pd based operations – (TCAP) long service life with clean gas
- LANA operations – limited service life due to He-3 formation
- Getters – depend on service conditions

Polymers - a necessary compromise:

- Sometimes unavoidable in tritium systems (seals, lubrication, packing, etc.)
- Polyethylene can release hydrogen, methane and other hydrocarbons
- PTFE (Teflon®) can react to form TF
- EPDM and Vespel™ polyimide – OK



Gas release from tritium aged LANA.75



Tritium aged samples of UHMW-PE, PTFE, and Vespel

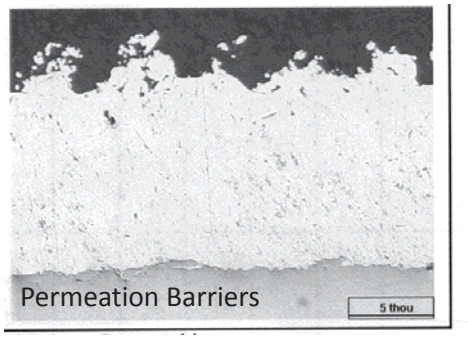
Tritium Effects on Materials –Steels & Coatings

Hydrogen embrittlement of containment materials exacerbated by decay to He-3:

- Use Austenitic SS
- Do not use Ferritic, Martensitic, or Precipitation Hardened SS
- Cleanliness and Surface Coatings Are Important

Permeation Barriers – must maintain integrity with temperature fluctuations:

- Tritium wetted face
  - Additional protection for equipment
  - Manufacturing challenges
  - Must be inert to process stream
- Glovebox face
  - Reduces stripper load
  - Easier to deposit



Intergranular Cracking -characteristic of hydrogen embrittlement

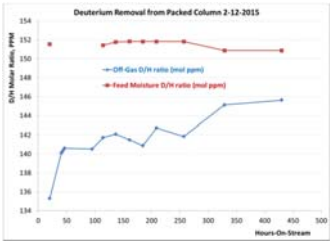
Application of 60+ Years of Tritium Experience to Other Areas

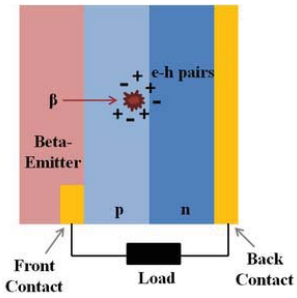
Current Endeavors:

- He-3 recovery from process materials
- Medical isotope production
- Fusion Support
  - ITER
  - LLE

Potential Endeavors:

- Water detritiation
- Betavoltaics
- Electron Capture Detectors
- Analytical capabilities
- T_2 Recovery from breeder materials





SRNL Support for SHINE

SHINE Medical Technologies, Inc. in Monona, WI

- Producer of medical isotopes, primarily Mo-99
- Currently no domestic producers
- NRU in Canada, the only North American producer and single largest producer in the world, is scheduled for shutdown in 2018
- Production technique favorable from a nonproliferation standpoint

SRNL leading design process for tritium system including:

- Accelerator interface
- Impurity removal
- Isotope separation
- Hydrogen storage and delivery
- Shutdown/startup protocols

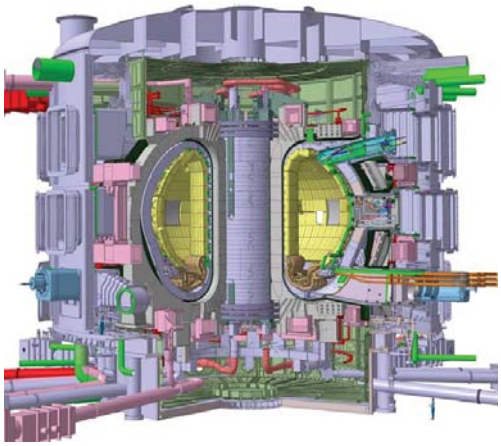
*Image from SHINE Medical Technologies™ website

SRNL Support for ITER

- US ITER Project Office

  SRNL/SRNS is the Design Authority/Design Agent for the Tokamak Exhaust Processing System which is one of 16 Procurement Packages within the ITER Fuel Cycle

- ITER International Organization Direct Support
  - Tritium Analytical System Conceptual Design
  - Highly Tritiated Water processing development and design verification
  - Tritium Process Control System Conceptual Design
  - Tritium Accountability and Tracking Program Development (MC&A)
  - Tritium Storage and Delivery System Hydride Storage Bed Design Support



SRNL Support for LLE

Laboratory for Laser Energetics in Rochester, NY

- NNSA Funded for Stockpile Stewardship
- "established in 1970 as a center for the investigation of the interaction of intense radiation with matter"
- Direct drive inertial confinement fusion
- ~1mm DT "ice" target placed within 10 μm of target

The OMEGA EP Laser System bathed in its own light as laser beams ignite during a target shot
*Image from LLE Website

SRNL involvement

- Fabricated micro -TCAP for batch isotope separation
- Provided technical guidance for startup
- Improved test results through better gas control of gas mixtures

Summary

- Tritium Processing is a complex chemical process – containment, separations, unexpected reactions, materials compatibility, and accountability.
- Tritium Processing R&D at SRNL is ongoing to ensure that both:
  - the needs of the Savannah River Tritium Enterprise are met, AND
  - the continuously evolving requirements of tomorrow's fusion machines are met.

Thank You

Backup Slides

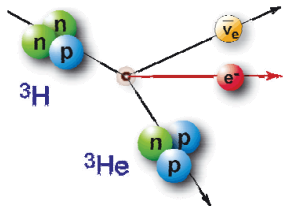
Radiochemistry of Tritium and Impurities (1/2) *

- Self-radiolysis of T_2

$$T_2 \rightarrow T_2^+ + others$$

$$T_2^+ + T_2 \rightarrow T_3^+ + T$$

$$T_3^+ + e^- \rightarrow 3T$$



- Self-radiolysis of T_2O (liquid T_2O is self heating)
  - β - radiation causes the formation of T_2O^+ ions, T_3O^+ ions, $T^\bullet$ radicals, $OT^\bullet$ radicals, and T_2O_2 via consecutive chemical reactions
    - *Due to the primary self-radiolysis products of T_2O it is highly corrosive*
  - Gas phase above T_2O becomes pressurized
    - *Radiochemical equilibrium above T_2O at 159 kPa (~1200 torr)*
      - Rate of gas production balanced by rate of (radiochemical) recombination

* Glugla, Fusion Reactor Fuel Cycle INSTN Lecture, February 6, 2009

Radiochemistry of Tritium and Impurities (2/2) *

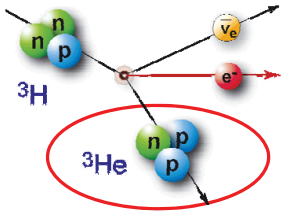
- Reaction of T_2 with oxygen, nitrogen and air

$$2T_2 + O_2 \rightarrow 2T_2O$$

$$3T_2 + N_2 \rightarrow 2NT_3$$

$$T_2 + air \rightarrow T_2O + T_2O_2 + NO + NO_2$$

$$CO_X + T_2 \rightarrow CT_4 + T_2O$$ as well as other heavier hydrocarbons



  - Reaction products are subject to (self-) radiolysis
  - Radiochemical equilibrium composition depends on tritium concentration (ratio of T/ΣH) and presence of metal catalysts (especially precious metals such as Pt, Pd)

- Reaction of T_2 with methane
  - If deuterium is present 15 different labeled methanes appear in gas phase

$$T_2 + CH_4 \rightarrow CH_3T + CH_2T_2 + CHT_3 + CT_4$$

* Glugla, Fusion Reactor Fuel Cycle INSTN Lecture, February 6, 2009

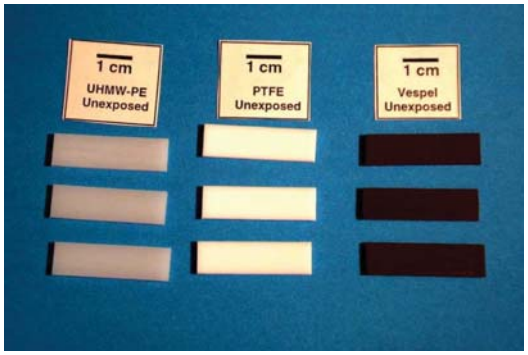
Tritium Effects on Polymeric Materials

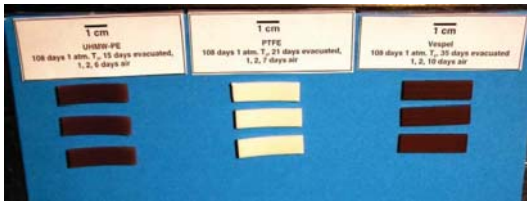
Polymers are a necessary compromise:

- Can't always avoid their use in tritium systems
- Needed for seals, lubrication, packing, etc.

SRNL and others have found that:

- When polymers are exposed to ionizing radiation (for example gamma rays, alpha particles, or beta particles from tritium decay), highly reactive "free radical" groups form when the energy of the ionizing photon or particle is absorbed *
- Polyethylene is known to emit hydrogen, methane and other hydrocarbon gases upon exposure to ionizing radiation *
- Tritium can react with PTFE (Teflon®) to form ^{3}HF (TF) *
- Crosslinked polymers contain sulfur which can be released in sulfur byproducts due to tritium decay – catalyst and permeation membrane poison





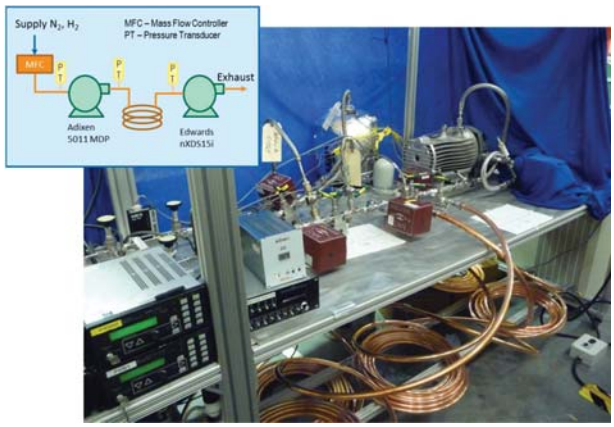
* Effects of Tritium Exposure on UHMW-PE, PTFE, and Vespel® (U) – Clark and Shanahan – May 2006

Tritium Fuel Cycle Technologies – Pumping (evacuation)

Evaluation of pumping technologies is necessary to move tritium-containing gases without the addition of impurities (from tritium exposure) – all metal construction is desired.

Candidate Technologies

- Normetex scroll pump and Metal Bellows pump
  - Normetex pumps unavailable
  - Alternate pump testing needed
- "Booster" pump to move gases
- High vacuum pumps (turbomolecular drag pumps)



Metal Bellows Pump MB-601

Edwards nXDS15i Dry Scroll Pump

Normetex (Eumeca) Scroll Pump

Adixen Molecular Drag Pump 5011

FHR and Tritium workshop, Salt Lake City, UT, Oct. 27

Overview of Tritium and molten salt FLiBe research at Safety and Tritium Applied Research (STAR) facility

Masashi Shimada
Fusion Safety Program, INL

www.inl.gov

Idaho National Laboratory

STIMS # INL/MIS-15-37109

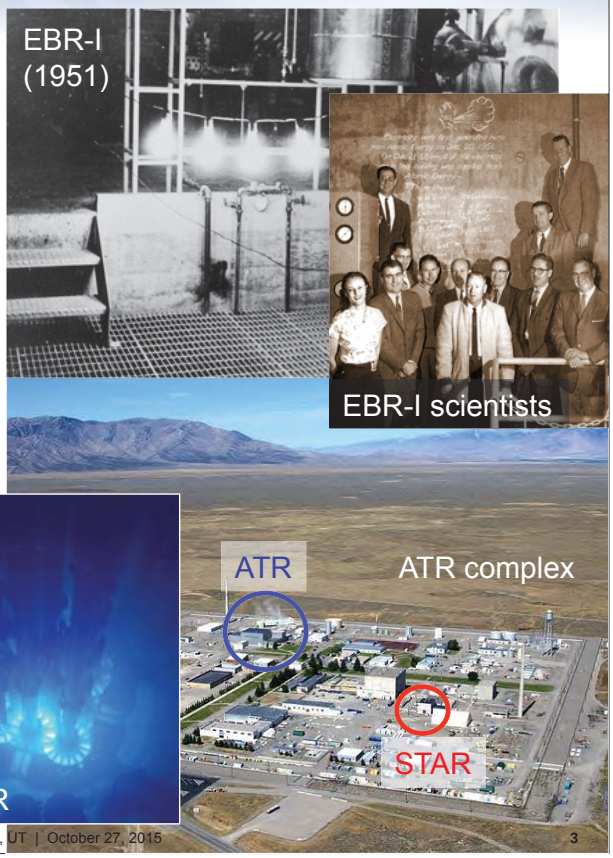
U.S. DEPARTMENT OF ENERGY | Office of Science

Outline

1. Motivation and overview of STAR research
2. Molten salt research at STAR during JUPITER-II project (April 2001 – March 2007)
3. Present capabilities

Idaho National Laboratory (INL) and fusion safety

- INL overview:
  - Geographically, the largest lab in 10 multi-program US national laboratories
  - 52 reactors were designed and built in Idaho
  - The world's first usable electricity from nuclear energy generated in EBR-I in 1951
  - The nation's lead laboratory for nuclear energy research and development
- Advanced Test Reactor (ATR)
  - Light water moderated/cooled with Be neutron reflector
  - Max: 250 MW_{th} and Four Leaf Clover" design
  - Materials and fuels testing, isotope production (e.g. ^{60}Co)
- Safety and Tritium Applied Research (STAR) Facility
  - Fusion safety and tritium research



STAR facility at Advanced Test Reactor Complex

Comparison of site size:

- INL
  - 890 sq. mile
- Rhode Island
  - 1214 sq. mile (0.73)
- Boston
  - 89.6 sq. mile (9.9)

NOTE: the number in the () is the ratio of the INL site to each site.

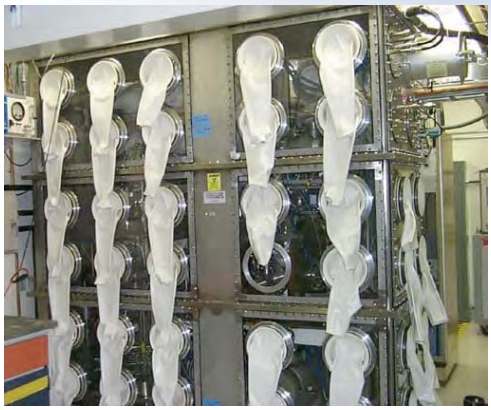
STAR facility is

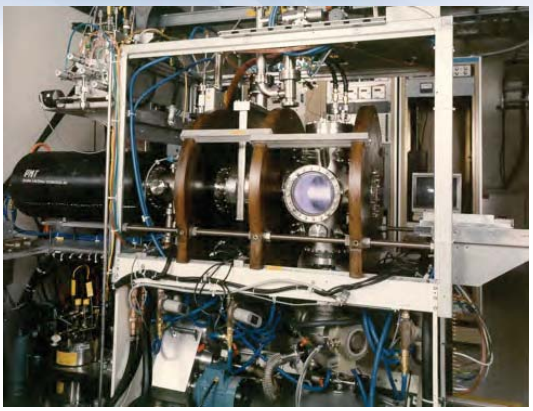
- Supported by DOE SC Office of Fusion Energy Sciences
- Restricted to a facility total tritium inventory of less than 1.6 gram (15,390 Ci), to remain below DOE Hazard Category 3 threshold
  - ➔ less than Hazard Category 3 Nuclear Facility
  - Current tritium inventory is ~ 0.35 gram (~3500 Ci)
- Specializes in:
  - Tritium (fusion fuel)
  - Activated materials (neutron-irradiated tungsten)
  - Advanced coolant (FLiBe, PbLi, He)
  - Toxic material (Be)
- Containment strategy for hazardous materials:
  - Ventilated enclosures and Laboratory hood for handling tritium
  - Gloveboxes for handling beryllium, Flibe etc..

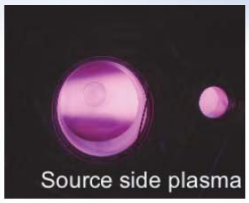
Motivation of STAR research

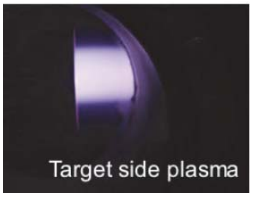
- Investigate tritium related and non-tritium related experimental fusion safety research
  1. **Tritium related fusion safety**
     - In-vessel tritium source term
       - Tritium retention in fusion materials
     - Ex-vessel tritium release term
       - Tritium permeation in fusion materials
  2. **Non-tritium related fusion safety**
     - Dust explosivity
       - Beryllium dust
     - Steam reactivity
  3. **Tritium related fission safety**
     - Tritium removal in molten salt ?

Tritium retention studies for fusion PFC development in Tritium Plasma Experiment









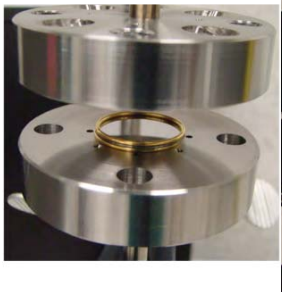
Unique capabilities

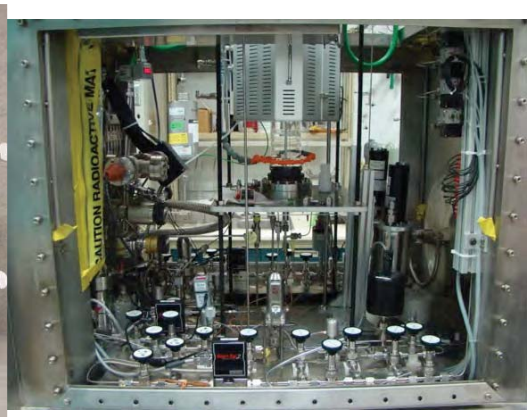
- *Designed to measure tritium behavior (e.g. retention and permeation) in activated materials with tritium plasma (e.g. tritium/deuterium ratio ~ (0.01-0.1) at divertor relevant ion flux condition*
- *Capable of testing neutron-irradiated (< 1 mSv/hr @ 30cm) specimens (W, RAFM steels, etc.)*
- *TPE is contained within double enclosure (PermaCon Box and Glovebox)*
- *Collaborations includes: US-JA TITAN collaboration (2007-2012), US-JA PHENIX collaboration (2013-2018), IAEA CRP on irradiated tungsten (2013-2018)*

Tritium absorption and permeation studies for fusion blanket development

Idaho National Laboratory

Test section for NFRI





Test section for PHENIX

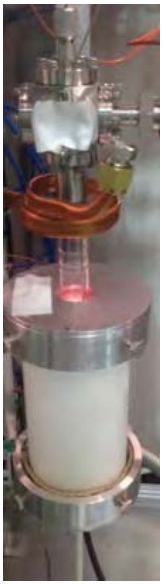
Unique capabilities

- Designed to measure tritium transport properties (e.g. diffusivity, solubility, and permeability) in activated materials at realistic fusion sweep gas conditions (e.g. low tritium <10 Pa & hydrogen partial pressures < 1000 Pa, moderate < 700 C)
- Capable of testing liquid or ceramic breeder materials (e.g. PbLi, Li_2TiO_3, etc.) and disc shaped metal specimens (W, RAFM steels, etc.)

Hydrogen/deuterium absorption studies for fusion blanket development

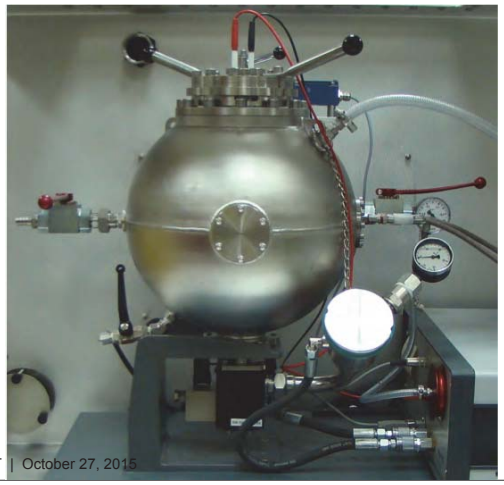
Idaho National Laboratory

- This static gas absorption system studies deuterium gas absorption in materials. It is located inside Laboratory fume hood for hydrogen safety.
- **Unique capabilities**
  - Sub atmospheric absorption (< 0.1 MPa) up to 950 C
  - Utilize three calibrated capacitance manometers (0.002, 1.3, 133 kPa)
  - Capable of testing liquid or ceramic breeder materials (e.g. PbLi, Li_2ZrO_3, KO functional materials etc.) and PFC/structural material (W, RAFM steels, etc.)
  - Capable of testing neutron-irradiated materials



Explosive dust evaluation for fusion vacuum vessel safety in ExCEED

- Developed for evaluating explosivity of mobilized beryllium dust for ITER VV safety (particularly in the presence of hydrogen generated by beryllium-steam reactions)
- **Unique capabilities**
  - One-of-a-kind dust explosion chamber for toxic and hazardous dusts
  - A 20 Kühner liter sphere, a standard device for dust explosion testing
  - A typical measurement test series varies the dust concentration to identify the maximum pressure and maximum rate of pressure rise and the concentration at which these occur
- Future work may include combined hydrogen/dust explosions for fusion safety



Outline

1. Motivation and overview of STAR research
2. Molten salt research at STAR during JUPITER-II project (April 2001 – March 2007)
3. Present capabilities

Overview of JUPITER-II program (Apr. 2001 – Mar. 2007)

- JUPITER-II
  - **J**apan-**USA** **P**rogram of **I**rradiation/**I**ntegration **Te**st for Fusion **R**esearch –**II**
  - Six years (2001-2006) under the collaboration implemented between MEXT (Ministry of Education, Culture, Sports, Science and Technology) and US DOE
- Task 1: Self-cooled liquid blanket
- **Task 1-1: FLiBe system**
- (Task 1-1-A) FLiBe Handling/Tritium Chemistry
  - Experimental work with FLiBe at STAR, INL for self-cooled liquid blanket of a fusion reactor.
  - Maintaining Flibe under a reducing atmosphere is a key issue to transform TF to T_2 with a faster reaction rate compared with the residence time in blanket.
  - The purpose of the task is to clarify whether or not the Redox control of Flibe can be achieved with Be through the following reaction.
    - Be + 2 TF ➔ BeF_2 + T_2
- (Task 1-1-B) FLiBe Thermofluid Flow Simulation
  - Simulation work at U. of Kyoto and UCLA

Reference:
K. Abe, A. Kohyama, S. Tanaka, T. Muroga, C. Namba, S.J. Zinkle, and D.K. Sze "Summary Report of Japan-US Joint Project (JUPITER-II)" ***NIFS- PROC-71 (2008)***

JUPITER-II (2001-2006)
Task 1-1-A: FLiBe Chemistry Control, Corrosion, and Tritium Behavior

- **Molten salt handling and purification**
  - Developed Material Handling Protocol with Safety Emphasis
  - Proved Effectiveness of Hydro-fluorination Purification
  - Demonstrated Capabilities for Impurity Quantification
  - **Experimental procedure:**
    - BeF_2 and LiF powders were dried and weighted to the mole ratio of 2:1
    - Melted with helium gas purge first, and then with gas mixture of He, H_2, HF at 520°C to reduce inherent oxides
    - The salt was transfered to another vessel through filtered through 60 μm metal mesh frit

Table 1 Impurities in ingredients and final salt

	O (ppm)	C (ppm)	N (ppm)	Fe (ppm)	Ni (ppm)	Cr (ppm)
BeF2	5700	<20	58	295	20	18
LiF	60	<2	78	100	30	4
Flibe	560	10	32	260	15	16

- **Mobilization studies**
  - Developed and Validated Transpiration System for Vapor Pressure Measurement of Molten Salts
  - Measured FLiBe Vapor Pressure at Low-temperature range Relevant to Fusion Blanket Designs
  - **Experimental procedure:**
    - Mobilization test was performed with Ar, air, and moist air in inert gas glove box.
    - (Ar test) conducted at 500, 600, 700, and 800°C with 25 sccm Ar flow
    - (Air test) conducted at 500, 600, 700, and 800°C with 25 and 50 sccm air flow
    - (Moist air test) conducted at 600, 700, and 800°C with 25 and 50 sccm moist air flow
    - Both Ni and glassy carbon crucibles were used

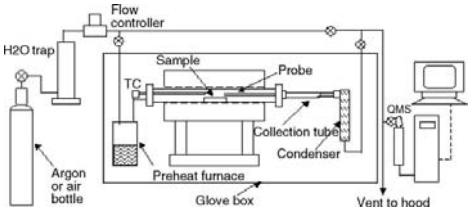


Fig. 1. Transpiration test setup.

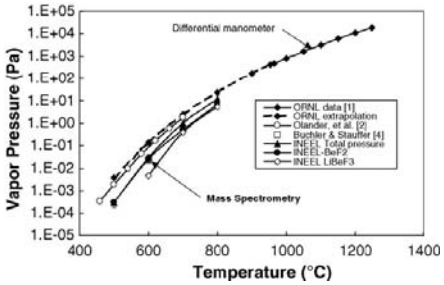


Fig. 2. Total pressure over Flibe.

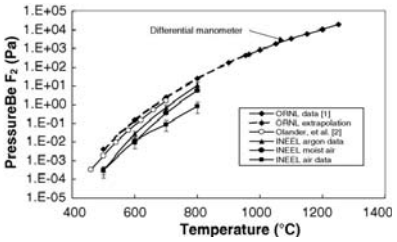


Fig. 3. BeF_2 pressure in various environments.

- **Redox control**
  - Demonstrated active control of the fluorine potential in FLiBe/Nickel systems using metallic Be
  - Proved the inhibition of FLiBe corrosion of Reduced Activation Ferritic Steel in static conditions
  - **Experimental procedure:**
    - The purpose of the task is to clarify whether or not the Redox control of Flibe can be achieved with Be through the Be + 2 TF ➔ BeF_2 + T_2 reaction
    - HF was bubbled with He and H_2 through FLiBe with various concentration of dissolved Be (cylindrical Be rod, 0.76 cm OD and 3 cm long) .
    - Ni crucible and Ni tubes were used and all the wet surface was Ni coated

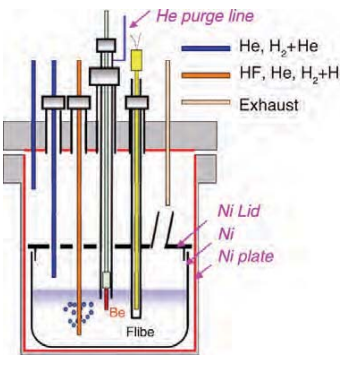


Fig. 5. Reactor for measuring redox

RAFS sample and metallic Be rods used for Redox and corrosion tests before and after immersion in FLiBe bath

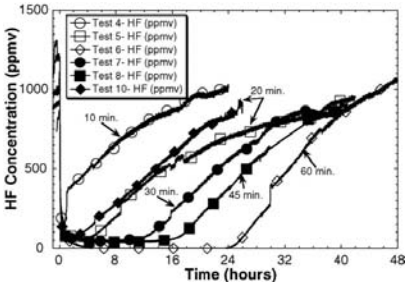


Fig. 6. HF concentration measured by QMS on the outlet of the REDOX experiment for several Be immersion times.

- **D_2 and T_2 permeation**
  - Measured transport properties (diffusivity and solubility) of D_2 and T_2 in FLiBe between 550 and 700 C
  - Investigated the effect of FLiBe Redox condition on T_2 transport
  - **Experimental procedure:**
    - (D_2 test) was conducted in a cylindrically symmetric dual probe permeation pot
      - Ni crucible and Ni tubes are used
      - at 600 and 650°C at $9.0x10^4$ Pa in NI Probe 1
    - (T_2 test) was conducted in a permeation pot with 2mm thick Ni membrane
    - at 550, 600, 700 and 800°C with 1 and 20 sccm (0.1 ppm-10 vo..% T_2/Ar)
    - Measured with QMS and GC

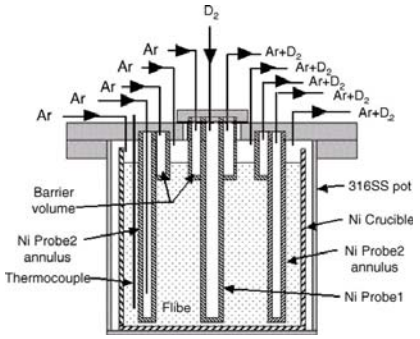


Fig. 10. Schematic illustration of cylindrically symmetric, permeation probe assembly.

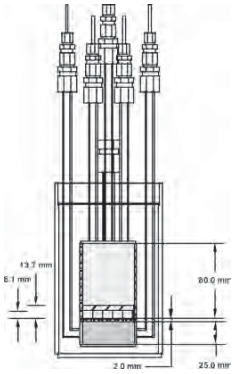


Fig. 1. Permeation cell scheme with relevant dimensions.

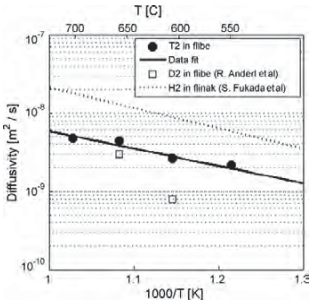


Fig. 3. Measured T diffusivity in flibe.

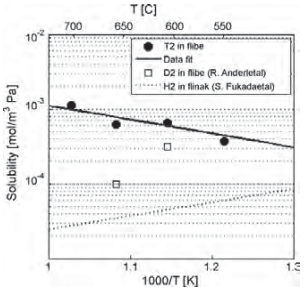


Fig. 4. Measured T solubility in flibe.

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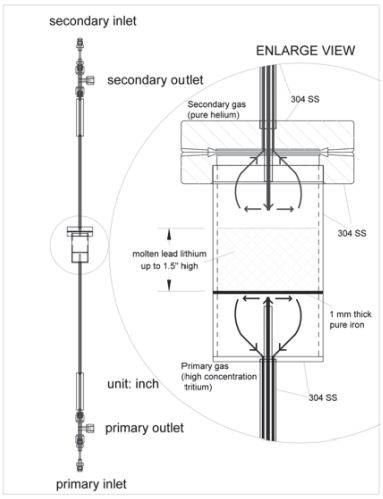
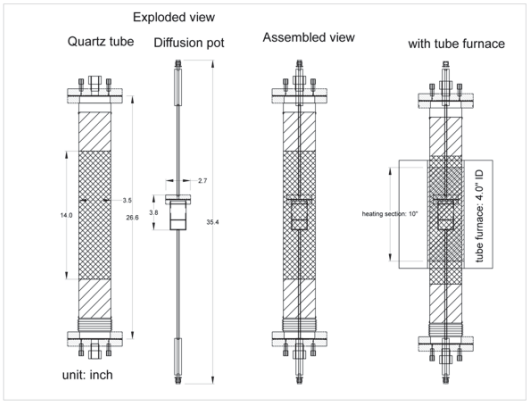
TGAP design is based on the FLiBe permeation pot

Tritium Gas Absorption Permeation experiment

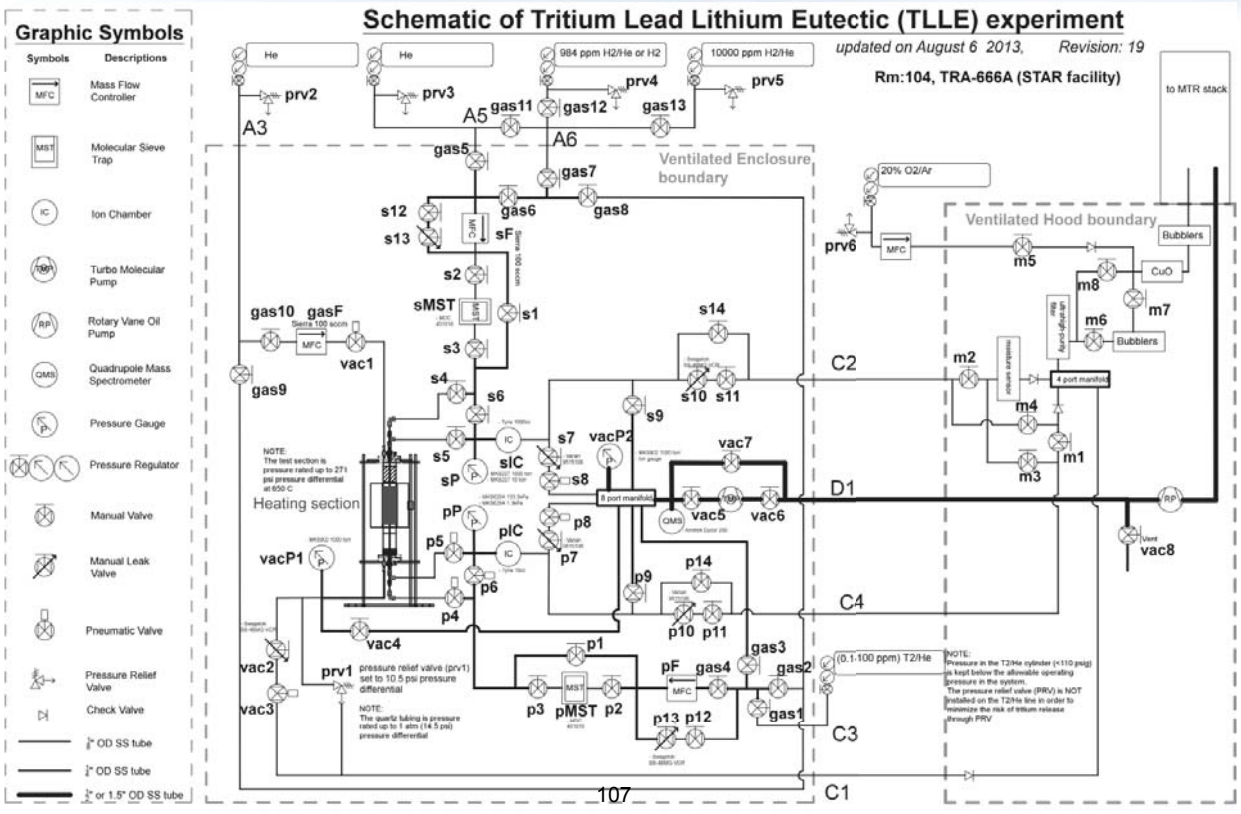




Unique capabilities

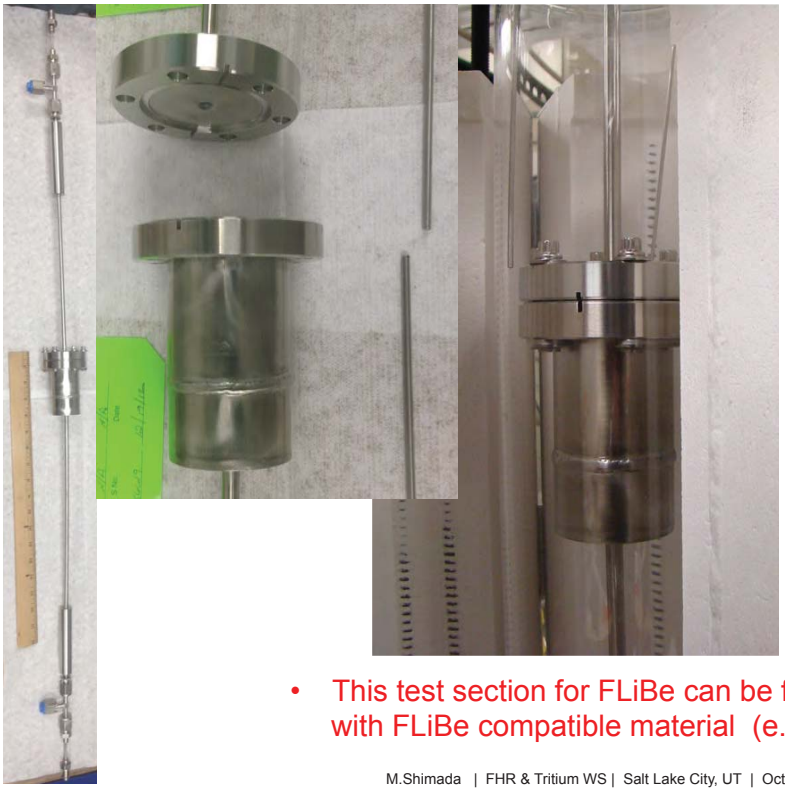
- Designed to measure transport properties (e.g. diffusivity, solubility, and permeability) of tritium at realistic blanket conditions (e.g. low tritium partial pressure < 1000 Pa) for disc geometry sample
- Capable of testing liquid breeder material (e.g. PbLi and FLiBe) and disc shaped metal
- Uniform temperature (+/- 10 C) within the test section utilizing 12” tube furnace

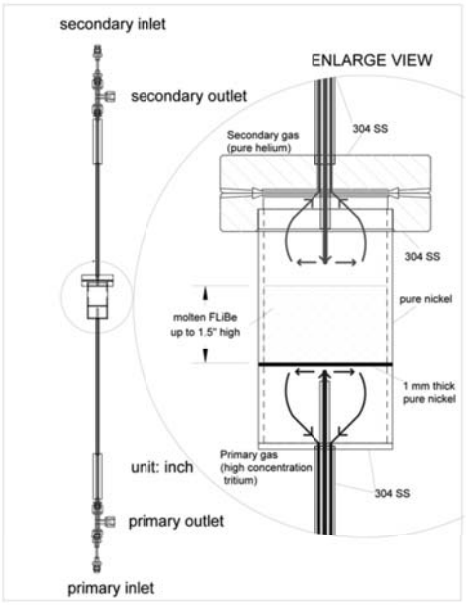
Schematic of tritium gas permeation system



Test section for Lead Lithium Eutectic (LLE)

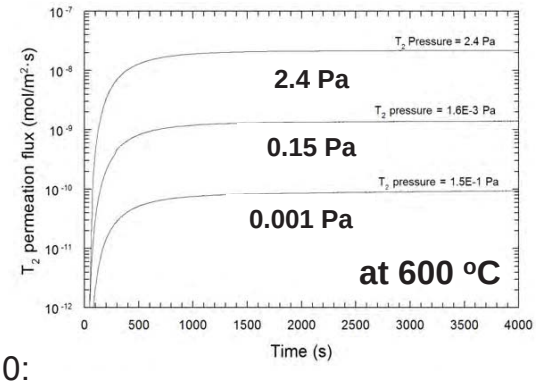
- **Photo of Test section for lead lithium eutectic**



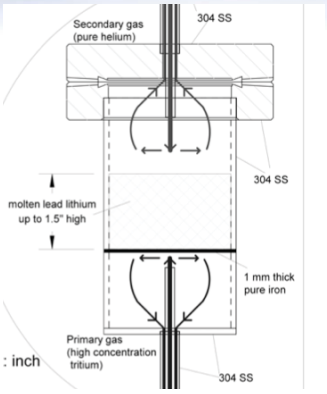


- This test section for FLiBe can be fabricated based on LLE drawing with FLiBe compatible material (e.g. nickel) on all wet surface

Experimental procedure (Fe/LLE)



H_2/He

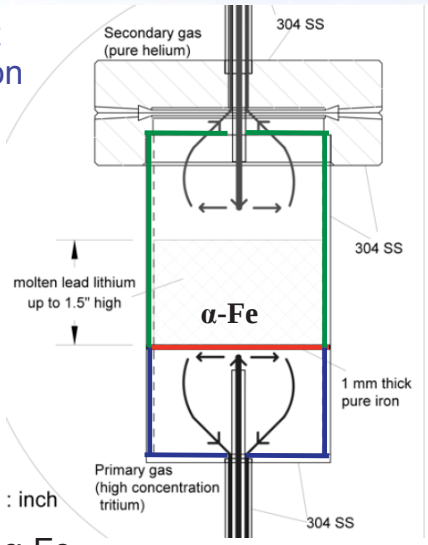


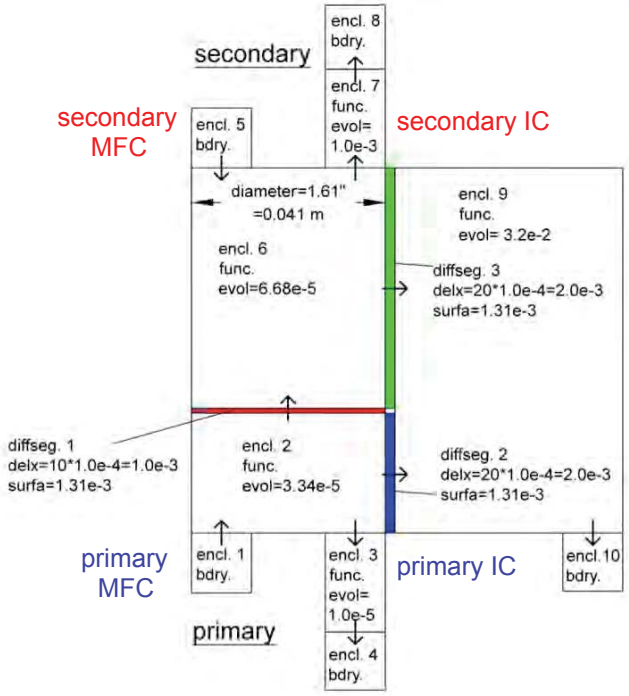
T_2/He

- t < 0:
  - Primary/Secondary was purged with **1 % H_2/He 100/200 sccm, p_t=10^5 Pa**
  - Bake out at 600 C with for 2 hours to remove oxide
  - Test section was kept at uniform temperature (+/- 10 C) for 1 hour at t<0.
  - Traps in the α-Fe were saturated by hydrogen.
- At t=0,
  - Tritium (0.001, 0.15, 2.4 Pa **T_2/He**) were introduced in the primary.
- t >0 :
  - Fast breakthrough time was obtained (within a minutes) and tritium equilibrates within 30 minutes

TMAP configuration for (1 mm) α-Fe

Experiment configuration

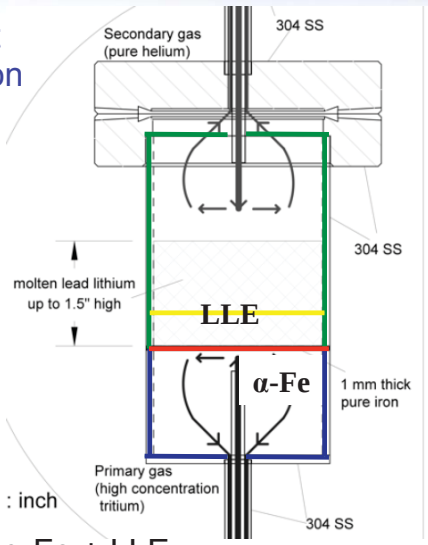


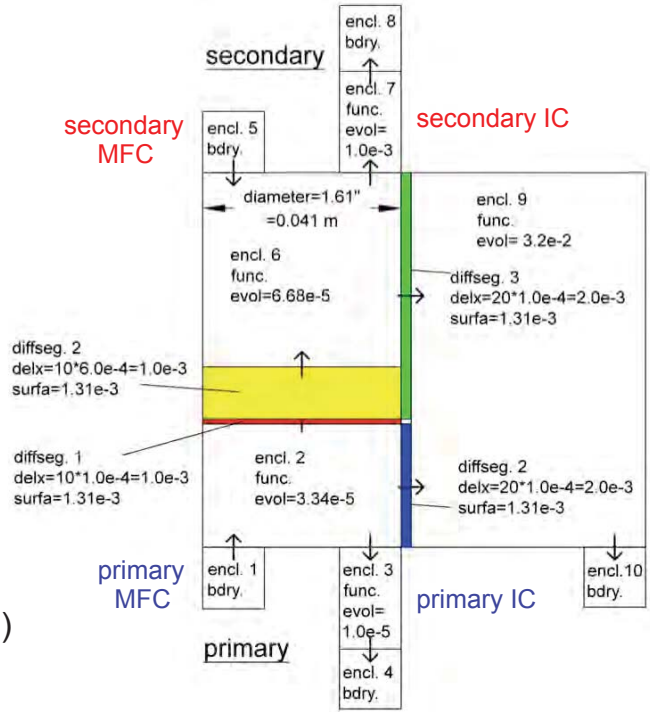


- TMAP for α-Fe
  - 1D geometry
  - 4 enclosure species (H_2, T_2, HT He)
  - 2 diffusion species (H, T)
  - 3 segments
  - 10 enclosures

TMAP configuration for (1 mm) α-Fe + (6mm) LLE

Experiment configuration



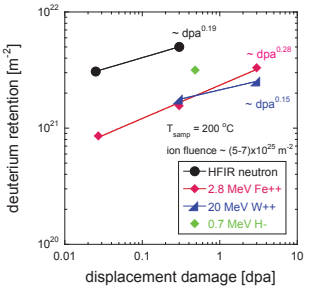


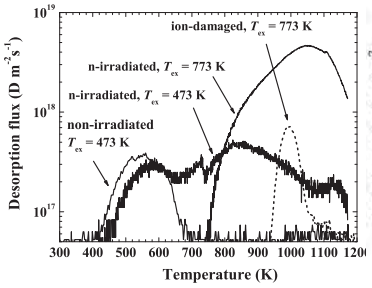
- TMAP for α-Fe + LLE
  - 1D geometry
  - 3 enclosure species (H_2, T_2, HT, He)
  - 2 diffusion species (H, T)
  - 4 segments
  - 10 enclosures

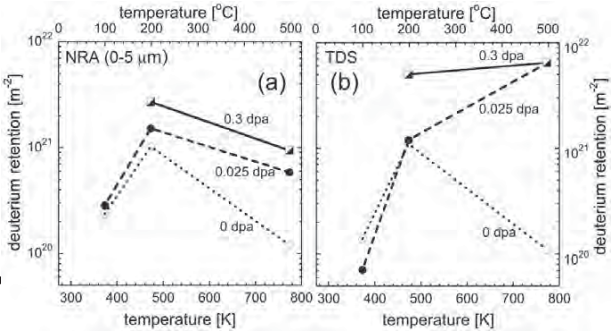
Idaho National Laboratory

Tritium retention in HFIR neutron-irradiated tungsten

- ✓ **US-Japan TITAN program (2007-2013):**
  - Low-temperature (<100 °C) low-dose (0.025 & 0.3 dpa) HFIR neutron-irradiated tungsten
  - Tritium was trapped in bulk (>10 μm), and retention increases at high temp. (500 °C)
  - Measurement of microstructural evolution and characterization of radiation defects and defect annealing before/after plasma exposure is required to reveal trapping mechanism.
- ✓ **US-Japan PHENIX program (2013-2018):**
  - High-temperature (500, 800, & 1200°C) medium-dose (0.3 &1.5 dpa) HFIR neutron-irradiated tungsten RB* irradiation with Gd thermal neutron shield
  - Positron annihilation spectroscopy (PAS) at ORNL and INL will characterize radiation defects and before/after plasma exposure to reveal defect annealing mechanism.
  - Nuclear reaction analysis, thermal desorption spectroscopy, and PAS is used to determine tritium migration depth and trap density for tritium retention assessment.







M. Shimada et.al. *Fus. Eng. Des.* **87**(2012) Y. Hatano et.al. *Nucl. Fusion* **53**(2013) M. Shimada et.al., *Nucl. Fusion* **55**(2015)

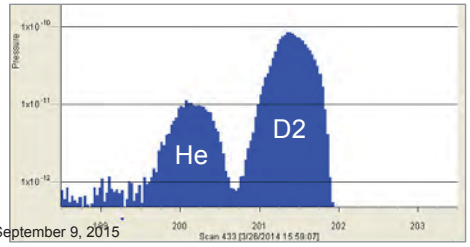
Idaho National Laboratory

Thermal desorption spectroscopy (TDS)

- Both TPE and NIMIIX study retention.
- To investigate retention,

$$R = \frac{\Phi_{out}}{\Phi_{in}}$$

- Heat samples up to 1100ºC
  - *Vacuum infrared tube furnace*
  - *Linear ramp rate*
  - *6 calibrated leaks (x3 He and x3 D2)*
- Analyze residual gases
  - *Two quadrupole mass spectrometers*
  - *High resolution QMS can distinguish:*
    - D2: 4.0282 amu
    - He: 4.0026 amu



Summary

- STAR at INL is DOE less than Hazard Category 3 Nuclear Facility for both fission and fusion safety R&D.
  - Maximum tritium inventory allowed 1.6 gram (15,390 Ci)
  - Capable of handling tritium, beryllium, FLiBe, and activated material
- FLiBe was extensively studied at the STAR facility during JUPITER-II program (2001-2006)
- STAR still operates 2x inert (Ar) gas gloveboxes for FLiBe use
- TGAP is capable of studying tritium permeation and absorption in FLiBe.
  - Design is based on the tritium permeation pot of FLiBe
- TPE, NIMIIX and SGAS can be used to investigate deuterium (tritium at TPE) retention in irradiated graphite for FHR.
- STAR can help develop tritium control and capture in FLiBe to enhance technical readiness level of FHR technology

Supporting slides

Tritium Migration Analysis Program (TMAP)

- The TMAP calculates the time-dependent response of a system of solid structures or walls (may be a composite layer), and a related gas filled enclosures or rooms by including
  - Movement of gaseous species through structures surfaces, governed by dissociation/recombination, or by solution law such as Sieverts' or Henry's Laws
  - Movement in the structure by Fick's-law of bulk diffusion with the possibility of specie trapping in material defects
  - Thermal response of structures to applied heat or boundary temperatures
  - Chemical reactions within the enclosures
- User specified convective flow between enclosures
- Equations governing these phenomenon are non-linear and a Newton solver is used to converge the equation set each time-step

NOTE: TMAP ver.4 can be obtained from DOE OSTI ENERGY SCIENCE AND TECHNOLOGY SOFTWARE CENTER

TMAP Capabilities (cont.)

- TMAP does not treat plasma surface physics, such as sputter or sputtered material re-deposition. TMAP's basic equations are:

Surface Movement

Non-equilibrium

$$J_m = K_d p_m - K_r C_m^2$$

Equilibrium

$$C_m = K_s \sqrt{p_{m_2}}$$

or

$$C_m = K_s \cdot p_{m_2}$$

Bulk Diffusion

$$\frac{\partial C_m}{\partial t} = \frac{\partial}{\partial x}\left(D \frac{\partial C_m}{\partial x}\right) + S_m - \sum_{i=1}^{K} \frac{\partial C_t^i}{\partial t}$$

Bulk Trapping

$$\frac{\partial C_t}{\partial t} = \alpha_t f_t C_m - \alpha_r C_t$$

Surface

J_m – Net specie surface flux (m^{-2}-s)
K_d – Molecular disassociation coefficient (m^2-s^{-1})
K_r – Atom recombination coefficient (m^2-s^{-1})
K_s – Sievert's solubility coefficient (m^2-s^{-1})

Bulk

C_m – Mobile specie concentration (m^{-3})
D – Specie diffusion coefficient (m^2-s^{-1})
K – Number of traps (-)
C_t – Trapped specie concentration (m^{-3})
α_t – Trapping rate coefficient (s^{-1})
f_t – Probability of landing in a trap site (-)
C_m – Mobile concentration (m^{-3})
α_r – Release rate coefficient (s^{-1})

$$\alpha_t = \frac{D}{\lambda^2};\ f_t = \frac{c_t^o - C_t}{N};\ \alpha_r = \nu_o \exp\left(-\frac{E_t}{kT}\right)$$

λ – jump distance or lattice constant (m)
c_t^o – Trap site concentration (m^{-3})
N – Bulk material atom density (m^{-3})
ν_o – Debye frequency (s^{-1})
E_t – Trap energy (eV)

Input parameter for TMAP modeling

- To apply TMAP to experimental data, property data for H diffusivity, solubility, and surface recombination/dissociation coefficients in α-Fe are required.
- Physical data of tritium partial pressure and sample temperatures are also required
- Mass transport property data used for H permeation in α-Fe are:
  - H diffusivity in alpha-iron [m^2/s]
    - y=4.43e-8*exp(-638.7/temp)
  - H solubility in alpha-iron [1/(m^3 Pa^0.5)]
    - y=4.2e23*exp(-2922.6/temp)
  - H/T recombination coefficient in alpha-iron [m^4/s]
    - y=4.6e-24/temp*exp(+4365.9/temp)
  - H/T dissociation coefficient in alpha-iron [m^4/s]

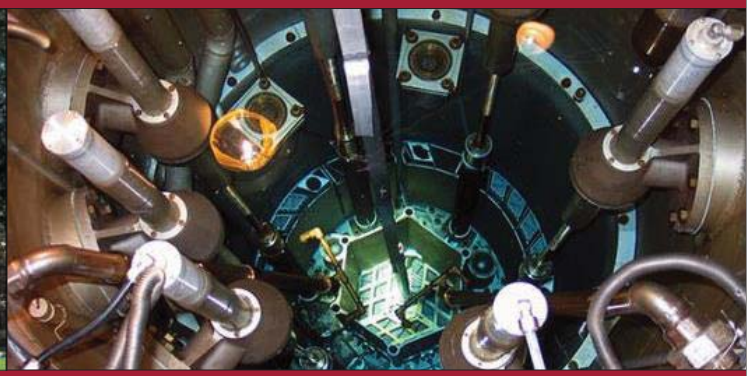
$$K_d = K_r \cdot K_s^2$$

- Mass transport property data used for H permeation in PbLi from Okitsu[5]
- Adjusted T diffusivity and T solubility to fit exp. data as two fitting parameters

[1] Yamanishi, et. al., Trans. Japan Institute of Metal (1983)
[2]Tahara and Hayashi, Trans. Japan Institute of Metal, v.26 (1985) 869
[3] Eichenauer et. a., Z. Metallkunde, v.49 (1958) 220
[4] Nagasaki et. al., JNM v.191-194 (1992) 258
[5] Okitsu et.al., FED v.87 (2012) 1324

MIT NUCLEAR REACTOR LABORATORY

AN MIT INTERDEPARTMENTAL CENTER



Experience with Tritium Evolution During Irradiation of MSRE Flibe in the MITR

David Carpenter

Group Leader, Reactor Experiments

10/27/15

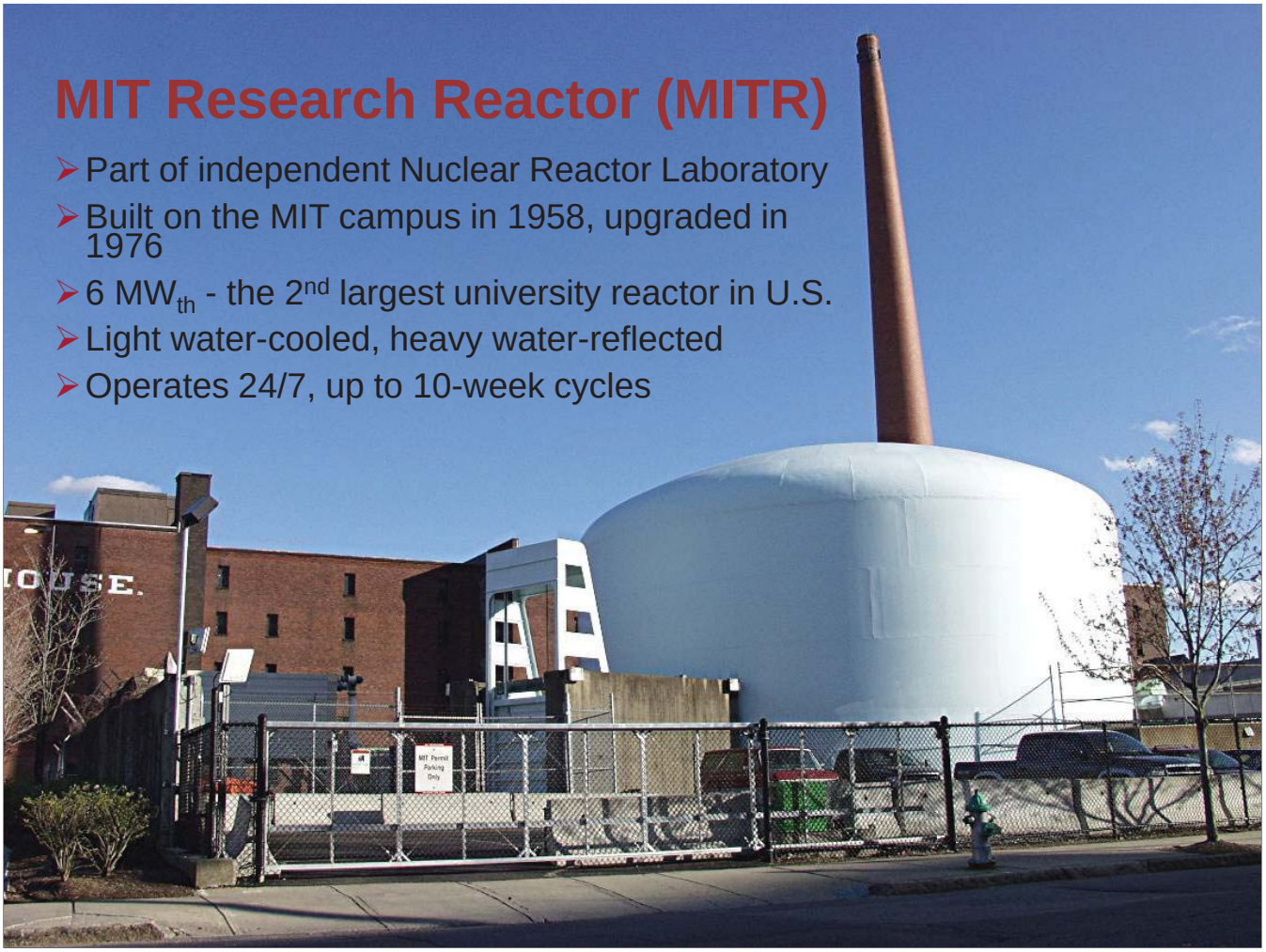
Outline

- MITR Introduction
- Tritium Experiments in the MITR
  - Goals
  - Limitations
- Tritium and Other Gas Release Measurements
  - FS-1
  - FS-2
- PIE Progress

- Please interrupt

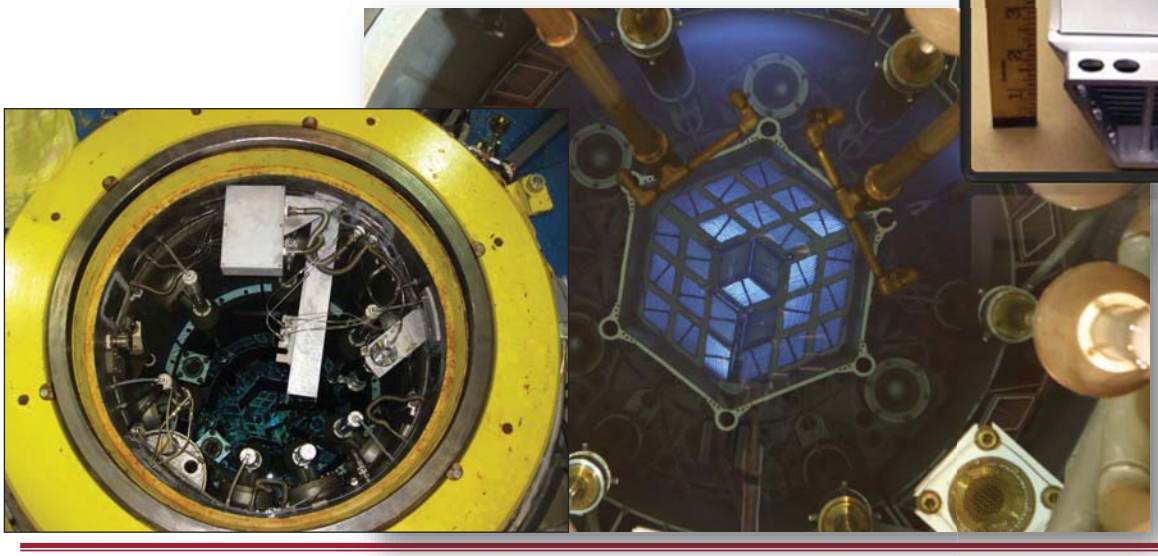
MIT Research Reactor (MITR)

- Part of independent Nuclear Reactor Laboratory
- Built on the MIT campus in 1958, upgraded in 1976
- 6 MW_{th} - the 2nd largest university reactor in U.S.
- Light water-cooled, heavy water-reflected
- Operates 24/7, up to 10-week cycles



MITR Core

- 24 HEU rhomboidal elements with UAl_x plate-type fuel
- 50°C outlet, atmospheric pressure
- 3 dedicated in-core experimental positions



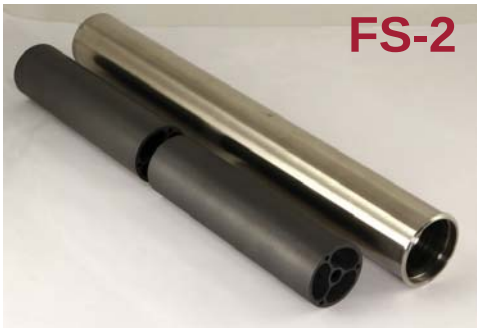
FHR IRP Irradiation Goals

- To test the interactions between the flibe and potential FHR fuel and structural materials
- To compare material test results with parallel test at University of Wisconsin (no irradiation)
- **To measure tritium production and partitioning among components**
- To evaluate the experimental components and methods for future FHR-related tests—the starting point that ultimately leads to larger experiments in HFIR and ATR

Initial Flibe Irradiations

- First in-core irradiation of MSRE flibe in 60 years
- Primary goal to identify potential safety and design issues for future experiments
- 1000 hours
- 700°C

- Study effect of different fluoride redox potentials on salt corrosion and tritium release
- Tritium and salt interaction with fuel compact graphite
- Cracking performance of TRISO particles
- 300 hours



Irradiation Design Progress

- Test conditions:
  - 700°C
  - Double encapsulation (Ni, Ti)
  - He cover gas, He/Ne control gas
  - 5.5MW reactor power

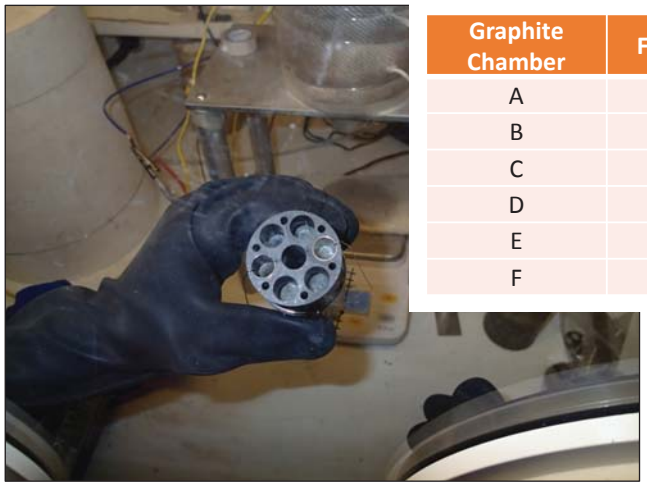
- FS-2 modifications:
  - Independent irradiation facility
  - 2.7x more salt volume
  - Two fluoride potentials
  - New C/C, graphite, and TRISO particles
  - Improved gas flow and exhaust

FS-1 Loading

Chamber	Container Material Combinations
A	Two Hastelloy-N samples
B	Two 316 Stainless Steel samples
C	Three types of SiC samples: R&H CVD SiC SiC/SiC Tyranno-SA3 CVI SiC composites SiC/SiC Hi-Nicalon type-S CVI SiC composite
D	300 surrogate TRISO particles
E	Two Hastelloy-N samples in nickel-lined hole
F	Two 316 Stainless Steel samples in 316SS-lined hole

FS-1 Capsule Assembly

Graphite sample holder filled with samples and flibe



Flibe Loading

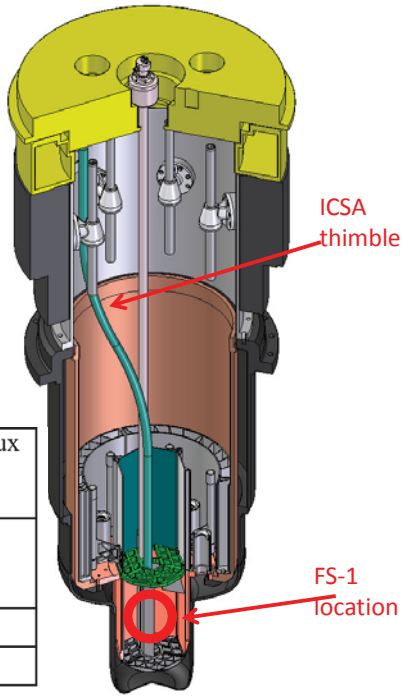
Graphite Chamber	Flibe Mass (g)
A	21.3
B	21.2
C	21.2
D	21.2
E	18.2
F	18.1

Capsule sealed with TCs and gas sampling lines

Samples in FS-1 are all identical to UW corrosion tests

FS-1 Capsule Irradiation

- The FS-1 capsule runs inside the MITR In-Core Sample Assembly (ICSA) thimble
  - The ICSA is an instrumented inert gas irradiation facility in a central core position
- Separate gas flow in capsule and thimble
- Thermocouples in graphite for temperature measurement

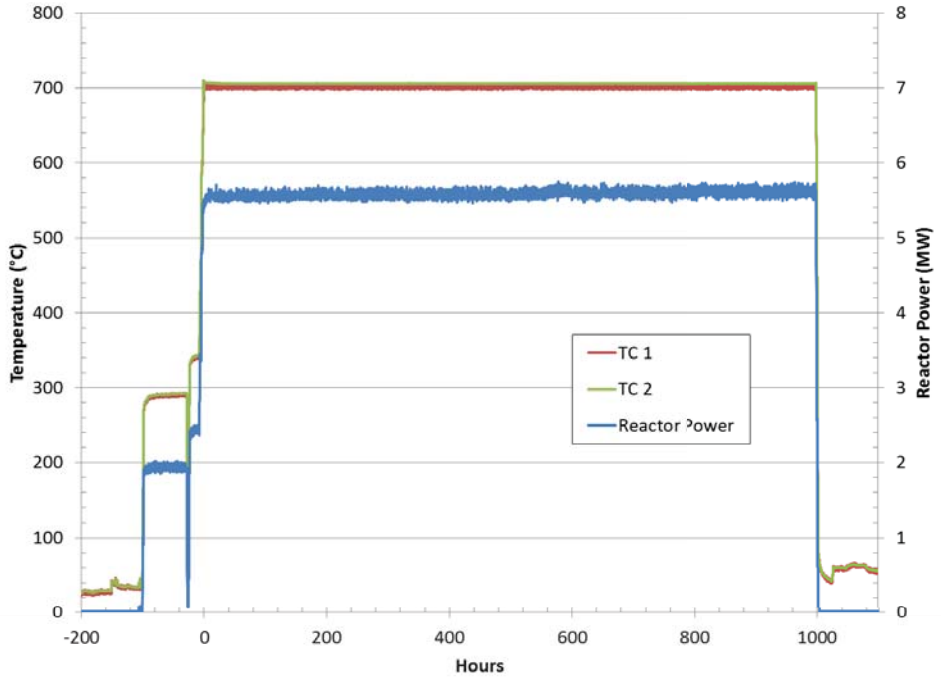


	Fast flux (n/cm^2-sec) (E > 0.1 MeV)	Total neutron flux (n/cm^2-sec)	Fast flux/Total flux ratio, %
FS-1 Flibe compartments (5.5 MW)	1.06×10^{14}	1.38×10^{14}	76.8
2009 PB-AHTR*	6.95×10^{13}	4.04×10^{14}	17.2
2011 AHTR*	6.18×10^{13}	3.64×10^{14}	17.0

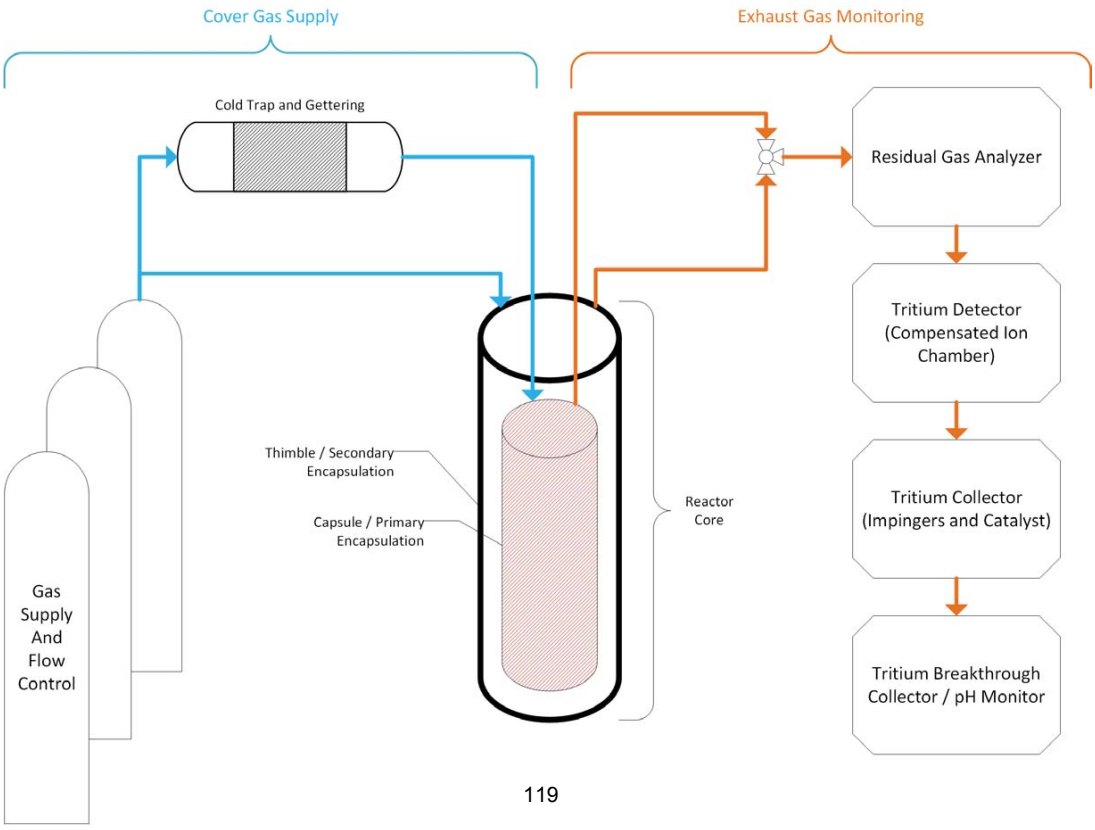
* FHR Materials, Fuels and Components White Paper, UCBTH-12-003, July 2013.

FS-1 Irradiation History

- Slow, stepped startup with fine control through melting
- Experimented operated at 700±3°C for 1000 hours



ICSA Irradiation Facility



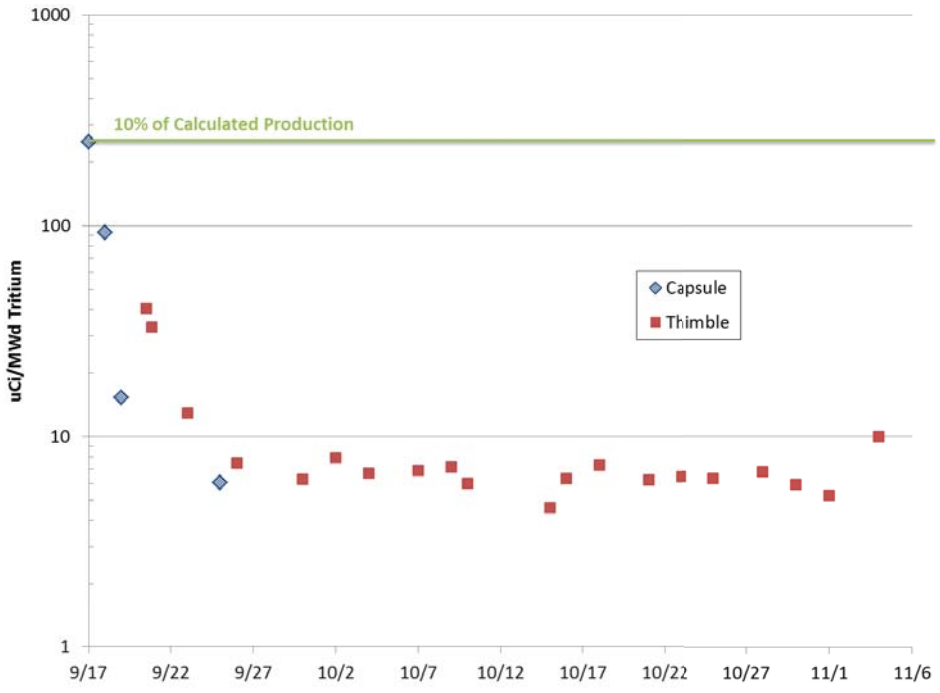
Tritium Measurements

- First irradiations attempted combination of real-time and integrated measurements
  - Dual compensated ion chambers used to measure real-time tritium activity
  - Six-pass water bubbler system with catalytic furnace to capture tritium in water for later liquid scintillation counting (LSC)
- This system should allow separate collection of soluble (TF, HTO, T_2O) and non-soluble (HT, T_2) species
- Bubbling with LSC has been is highly repeatable with good capture efficiency, and allows differentiation of H-3, C-14 activity

- Released activity below range of ion chamber sensitivity/background rejection

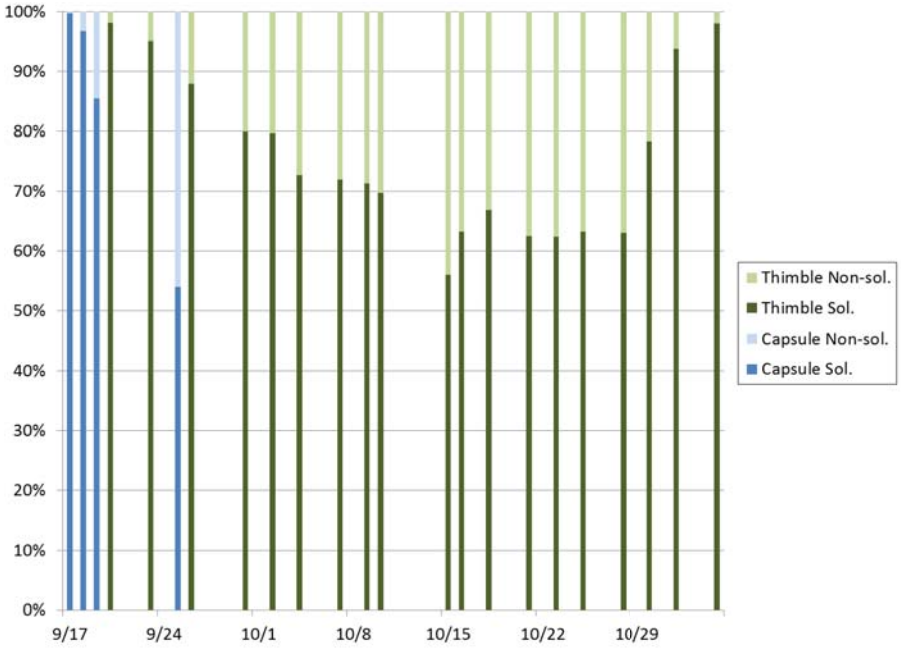
FS-1 Initial Findings – Tritium Activity

- Tritium counts integrated over 24-48 hour intervals
- Tritium collected during startup was ~10% of predicted production
- Subsequently, collected less than 1% of production
- Capsule and thimble tritium levels similar



FS-1 Initial Findings - Solubility

- Tritium collected in initial impingers expected to be only water-soluble form,
  - Post-catalyzing furnace the previously non-soluble species will be captured
- Only non-soluble species (T_2) should be released through diffusion
- Additional tests are required to identify species to explain apparent contradiction



FS-2 Loading

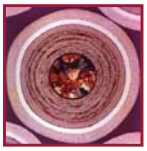
- Added C/C and matrix graphite, removed Hastelloy
- Redox potentials straddle the historical MSRE potential (-665 to -707 kJ/mol F_2)

Graphite Holder	Compart-ment	Material	Flibe F_2 Potential (kJ/mol)
Upper	A	Graphites	-632
	B	SiC/SiC, TRISO	-632
	C	SiC/SiC, TRISO	-711
Lower	A	C/C	-632
	B	SS316	-632
	C	SS316	-711

FS-2 Loading

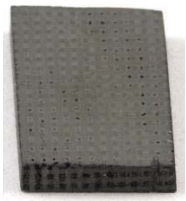
ARB-B1-Z009

IG-110U

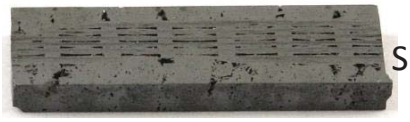


B&W TRISO
G73F-NF-93044

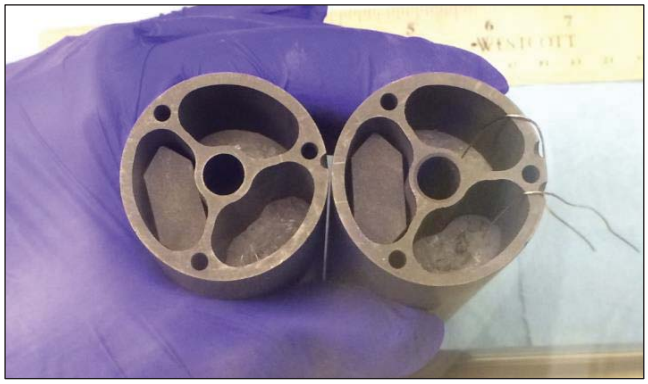
SS316



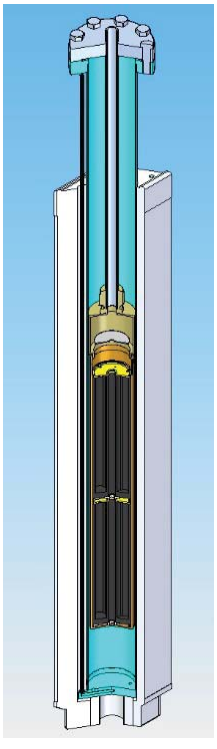
C/C



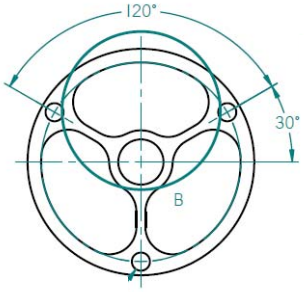
SiC/SiC



FS-2 In-Core Facility Design

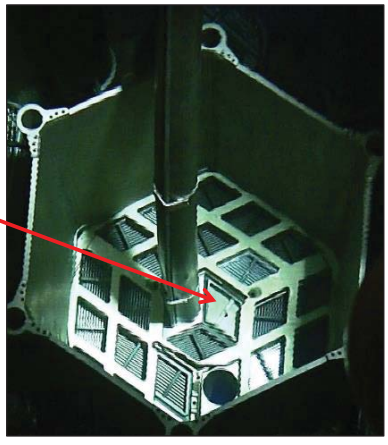


Legend:
White – Aluminum dummy element
Blue – Titanium thimble
Orange – Nickel capsule
Yellow – Internal nickel parts
Gray – Graphite sample/flibe holder



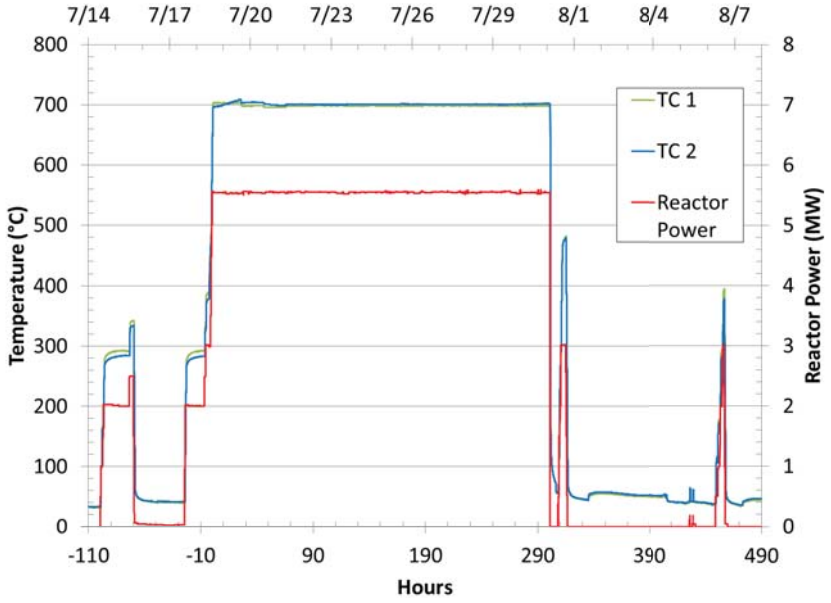
Graphite cross-section

FS-2 Position
(A-1)



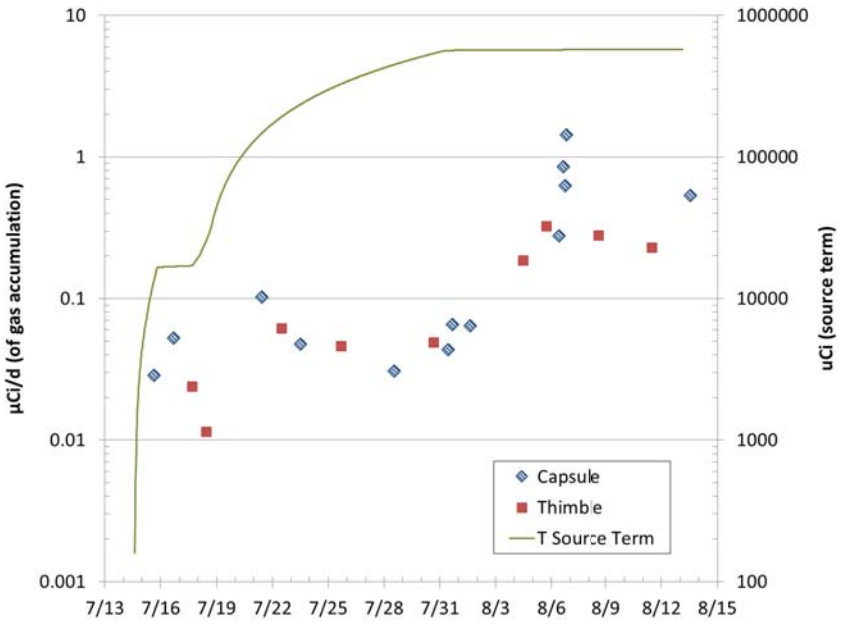
FS-2 Irradiation History

- First startup did not reach the flibe melting point
  - Reduction in power to 50kW for maintenance (~50°C)
- Eventual startup to 700°C was uneventful
- Unrelated reactor SCRAM at 300hrs, restart attempt
- After several days of troubleshooting, performed short test to 3MW, then removed experiment



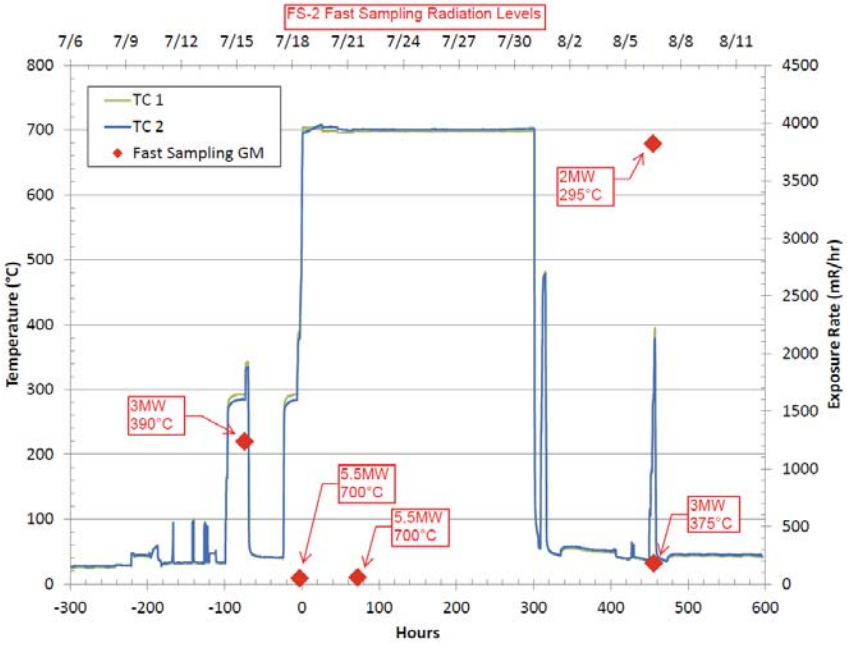
Tritium Release vs. Source Term

- Capsule and thimble gas tritium levels similar
  - Easy diffusion through capsule
- Majority of tritium is immediately soluble in water
  - Additional catalyzing had small effect
- Tritium release rate increases during temperature changes
- Tritium release during normal operation (5.5MW, 700°C) is same order of magnitude as FS-1 (<1% of generation)



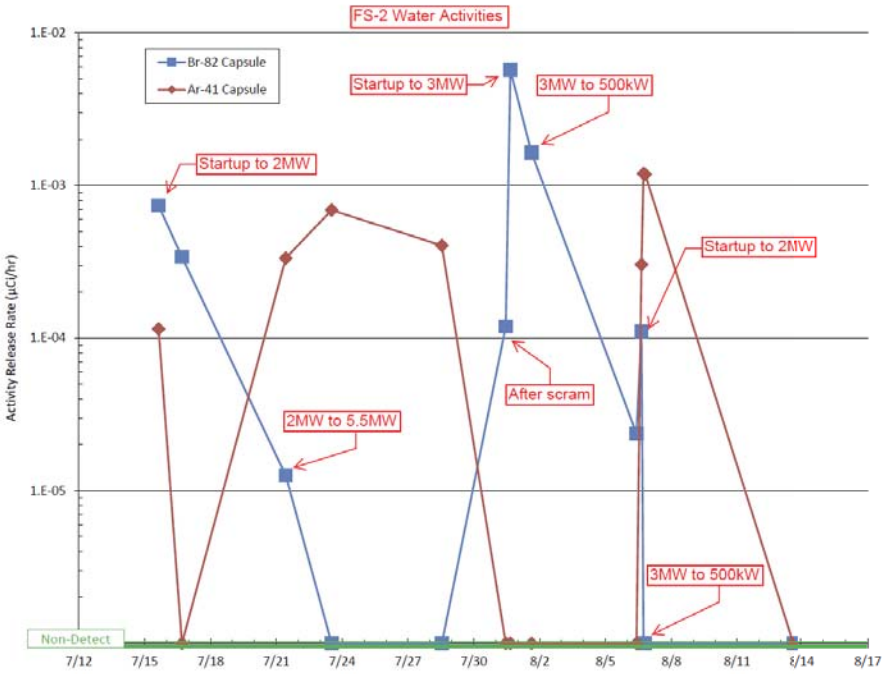
Short Half-Life Detection

- In FS-1, problem of short-lived activation products in capsule exhaust
  - Added special gas system for FS-2
- In FS-2, highest N-16 and O-19 release when flibe was solid
  - Produced by F-19 neutron activation
- Almost no detectable release at full reactor power, 700°C



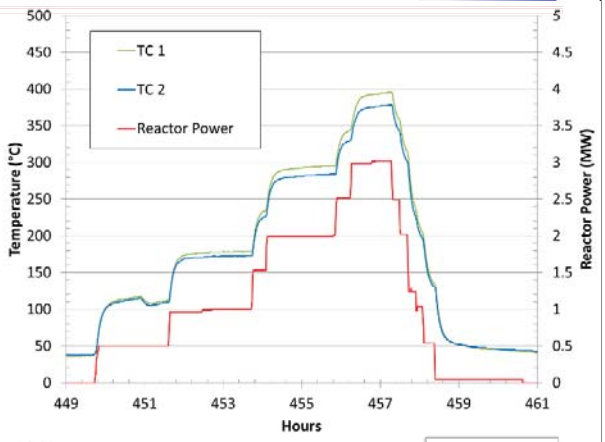
Bromine Release

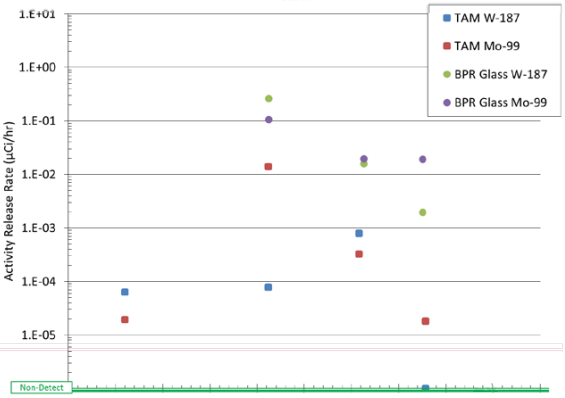
- Bromine was detected in tritium collection system
  - Only from capsule gas
  - Here contrasted with Ar-41, an expected He impurity
- Highest Br levels when at low power – flibe is solid
- Not detected at full power, 700°C



FS-2 Gaseous Release Test

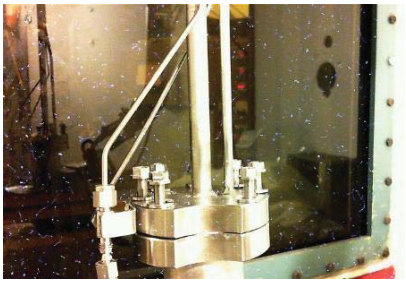
- Following the SCRAM, conducted a test to 3 MW to allow additional sampling
  - Added glass, paper, and charcoal filters prior to tritium capture bubblers
  - Activation products only seen from capsule
- Molybdenum release and capture on filters highest from 0-2MW (<200°C)
- W and Mo captured on glass and paper filters effectively, not on charcoal
- Br not captured on filters, minimal tritium capture on filters
- Experiment removed from reactor following the test

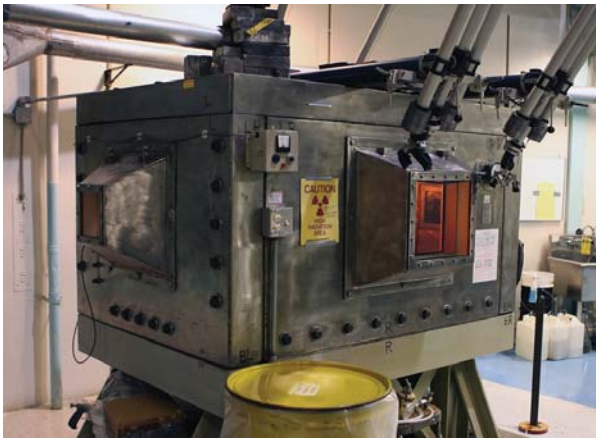




Post-Irradiation Examination

- FS-1 and FS-2 irradiation capsules extracted from in-core facility and transferred to Hot Box and Hot Cell, respectively
- Pressurized with helium to prevent moisture uptake
- Exterior condition of titanium pressure vessel and the nickel/Inconel capsule is excellent

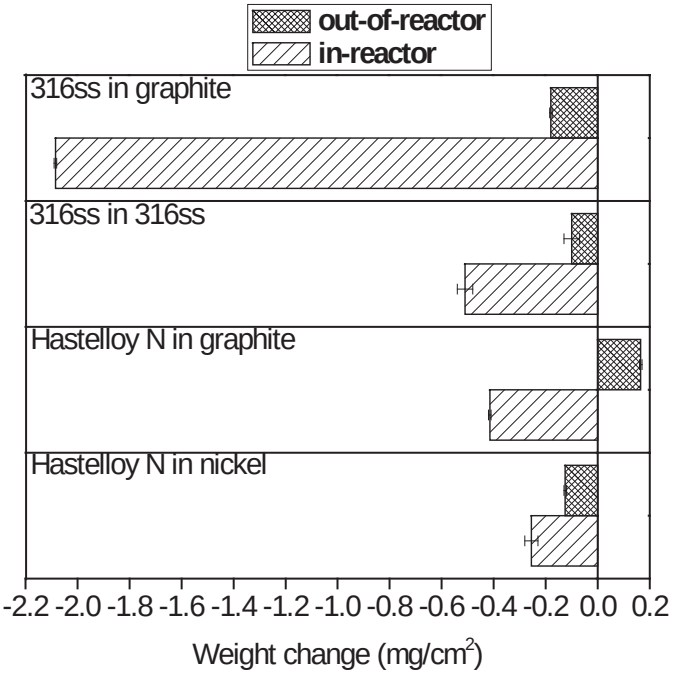


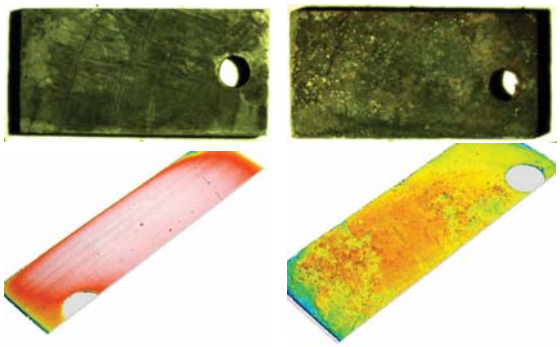


Specimen Extraction

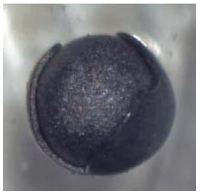
- All specimens have been removed from the FS-1 capsule and cleaned of flibe
  - Furnace operated in dry helium glove box to prevent HF generation and contain tritium
- Cleaning involved melting and collection of flibe ~500°C, followed by soaking in water at ambient conditions until weight decrease arrested
- Did not see evolution of other activation products or substantial tritium release during salt melting

Initial PIE – Corrosion & Cracking

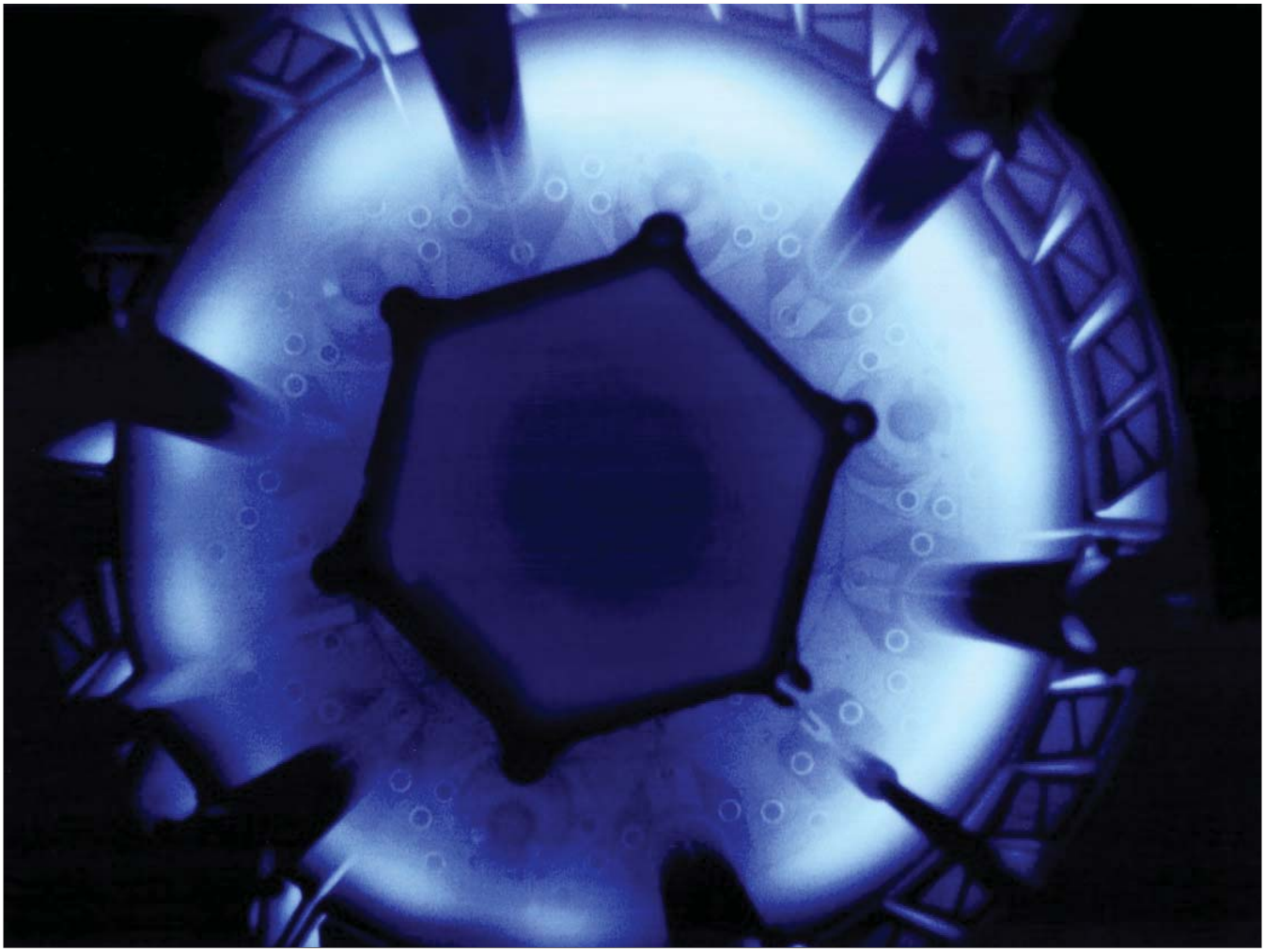




SS316 irradiated in lined (left) and unlined (right) chambers



Surrogate TRISO particle with OPyC damage



FLiBe Electrochemistry and Materials Corrosion Research at UW-Madison

Thomas Chrobak, Karl Britsch, Dr. Guoping Cao, Dr. Kumar Sridharan, Dr. Mark Anderson

Tritium Workshop, Salt Lake City, UT

10/27/2015

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Outline

- Introduction to LiF-BeF_2 (FLiBe)
  - Fluoride Salt Chemistry
  - Molten Salt Corrosion
- Electrochemistry Studies on Fluoride Salts
  - FLiBe Redox Measurements
- Static Corrosion Test
  - Experimental design and materials
- FLiBe Natural Circulation Flow Loop
  - Flow-assisted corrosion testing in FLiBe convection loop

Research Questions to be answered

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- Will a 0.3V difference in the **redox potential** of FLiBe salt cause a significant **increase in corrosion behavior**?
  - (-1.71V) As-purified vs. (-1.41V) Be-reduced
- Does the presence of **graphite** in the salt facilitate **corrosion**?
  - Liner vs. without liner
- What is the effect of **flow** on **corrosion?**
- Is there a significant difference in **corrosion behavior** of samples in the **cold or hot leg** of a natural circulation flow loop?
- What is the **compatibility** of new selected **materials**?

More Research Questions for Consideration

- What is the **optimum amount of Be** that should be added to FLiBe?
  - Balance between over and under-reduction → smallest quantity possible sequentially added

Stoichiometric vs. Excess ?




- Does the **over-abundance of Be metal** in the salt cause enhanced **corrosion** to **carbon-containing parts**?
  - Possible formation of a BeC passivation layer that can keep salt redox low while protecting C from further corrosion?
  - Increased wetting of glassy carbon crucible following Be reduction
    - Convex surface before reduction vs. concave surface after reduction.

Why Use FLiBe as Liquid Salt Coolant?

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- Higher outlet temperatures lead to:
  - More valuable process heat applications
  - Greater cycle efficiencies→ $\eta_{Carnot} = 1 - \frac{T_C}{T_H}$

	Operating Temperature [°C]	Operating Pressure [MPa]	Thermal Conductivity [W/m-K]	Volumetric Heat Capacity [kJ/m^3-K]	Viscosity [Pa-s]
Water	300	**15**	0.55	3970	8.8×10^{-5}
Helium	850	7.5	0.29	**20.9**	4.2×10^{-5}
Sodium	550	Atmospheric	**62**	1008	2.3×10^{-4}
Lead	550	Atmospheric	18.25	1499	1.67×10^{-3}
FLiBe	650	Atmospheric	1.0	**4683**	5.6×10^{-3}

- No ideal heat transfer fluid exists
- Molten fluoride salts offers a good compromise of properties

Properties of FLiBe meet most requirements for FHR Salt

- Molten Salt Primary Coolant Requirements
  1. ✓ Exhibit chemical stability at T > 800 °C
  2. ✓ Stable in an intense radiation field
  3. ✓ Consist of low thermal cross section elements
  4. ✓ Melt at useful temperature (<500 °C) without being volatile
  5. ? Compatible with high-temperature alloys and graphite

MSRE Salt. Blue tint from dissolved UF_4

- $LiF\text{-}BeF_2$ – FLiBe as primary coolant
  - \+ Atmospheric pressure operation
  - \+ Good heat transfer properties
  - \+ Neutron transparent
  - \+ Wealth of MSRE experience
  - – Tritium production from ^6Li
  - – Beryllium toxicity
  - – **Corrosive without chemistry control or proper materials**

Isotope	F	Be	Li-7	B-11	Zr	Rb	Na
Thermal Cross Section	0.009	0.010	0.033	0.05	0.18	0.37	0.53

Overall Corrosion Process

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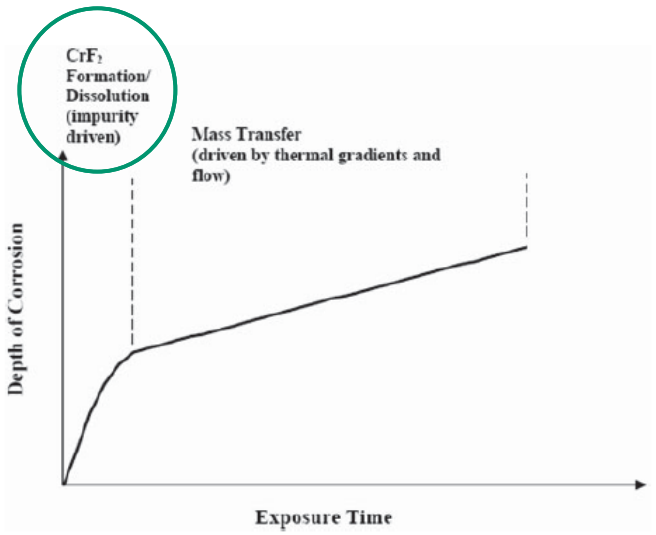
- Impurity-driven corrosion dominates initial phase
- Thermodynamically-driven leads to continuous corrosion



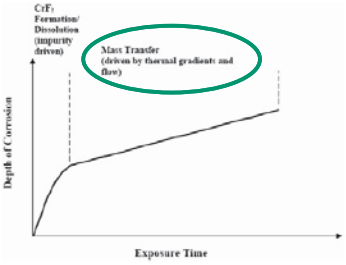
Possible Corrosion Solutions

- Minimize thermal gradients?
- Use high Ni, low Cr Alloys?

➢ Implement chemistry control of salt with **redox potential measurement** to maintain high salt quality

Thermodynamically Driven Corrosion

- Non-favorable reactions slowly occurring
- Assisted by a temperature gradient and mass flow

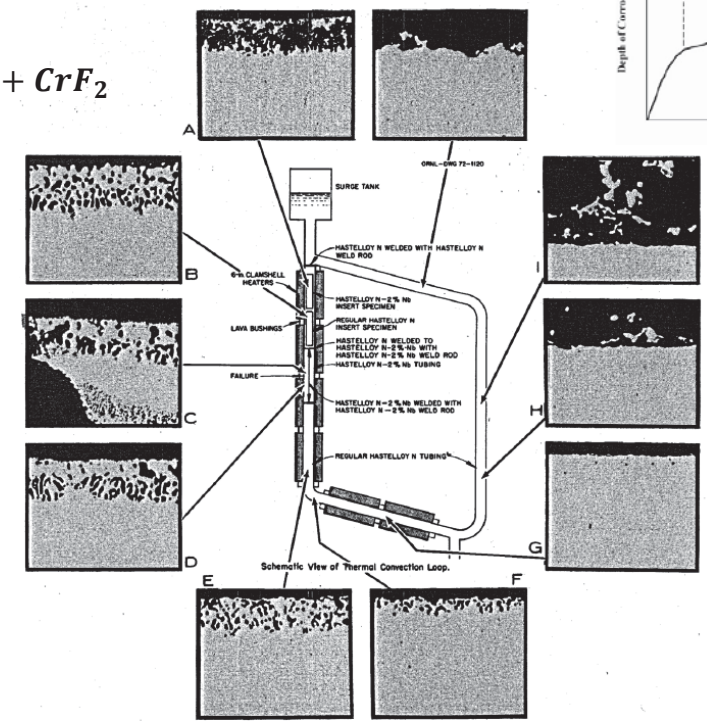


$$BeF_2 + Cr \leftrightarrow Be + CrF_2$$

$$K_{eq} = \frac{[Be][CrF_2]}{[BeF_2][Cr]}$$

K_{eq} = 8.66 x 10^{-13} at 700 °C

K_{eq} = 2.76 x 10^{-14} at 600 °C



Impurity Driven Corrosion

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- Thermodynamically favorable reactions due to unstable impurities
- Occurs quickly in initial corrosion stages

Metal Fluoride Impurity Reactions

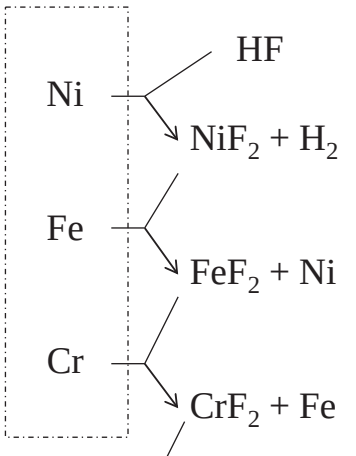
$$M_{alloy}(s) + M_{imp}F_x(d) \rightarrow M_{alloy}F_y(d) + M_{imp}(s)$$

Moisture Impurity Reactions

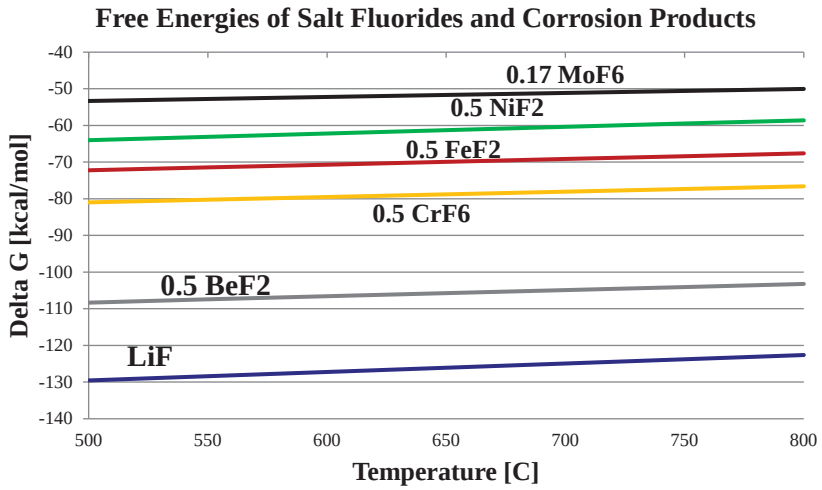
$$\frac{x}{2}H_2O + M_{salt}F_x \rightarrow M_{salt}O_{\frac{x}{2}} + xHF$$

$$xH_2O + M_{salt}F_x \rightarrow M_{salt}(OH)_x + xHF$$

$$M_{alloy}(s) + xHF \rightarrow M_{alloy}F_x(d) + H_2$$



Fluoride Salt Corrosion Mechanisms

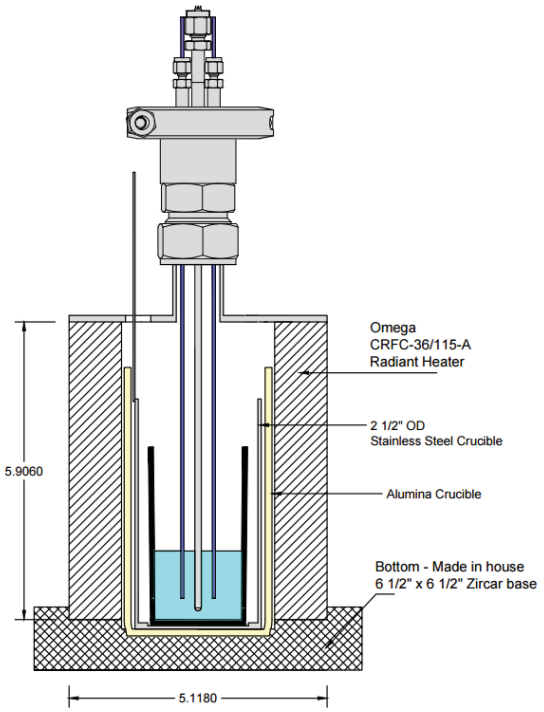


Corrosion Products
(less stable)

Salt Constituents
(more stable)

- Salt constituents are more stable than metal fluorides
- Almost no corrosion expected from pure FLiBe
- How can we ensure purity of FLiBe? → Redox potential

Experimental facilities for electrochemical testing of FLiBe



- HP 3616A Power Supply
- Ar glovebox. O_2 and Moisture <1 ppm
- Radiant heater, PID used to maintain 500±0.5°C

Dynamic reference probe design for compact redox potential testing of FLiBe

- O-Ring
- BN Spacer
- Glassy Carbon Anode
- Mo Cathode
- Mo Indicator
- Mo TC well
- K-Type TC

Dynamic Reference Electrode Measurements

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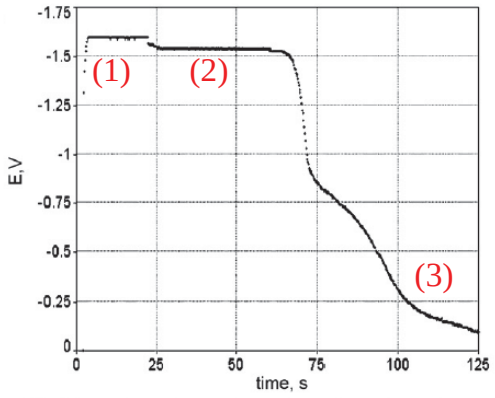
- Combination of Dynamic and Static Techniques

First Phase

- Beryllium is plated from the salt onto an electrode (1)

Second Phase

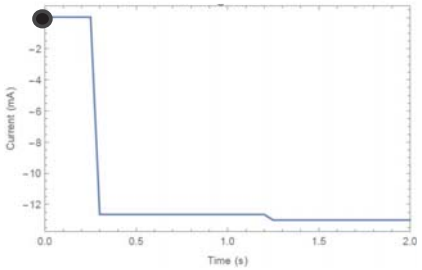
- Voltage is cut, beryllium allowed to redissolve back into the salt
- Be|BeF_2 reference voltage is formed from dissolution reaction (2)
- As plated products deplete, voltage relaxes back to zero (3)



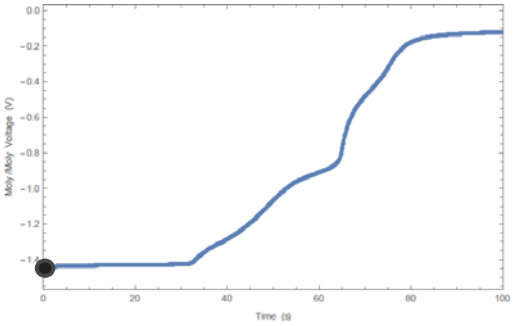
(Afonichkin, 2009)

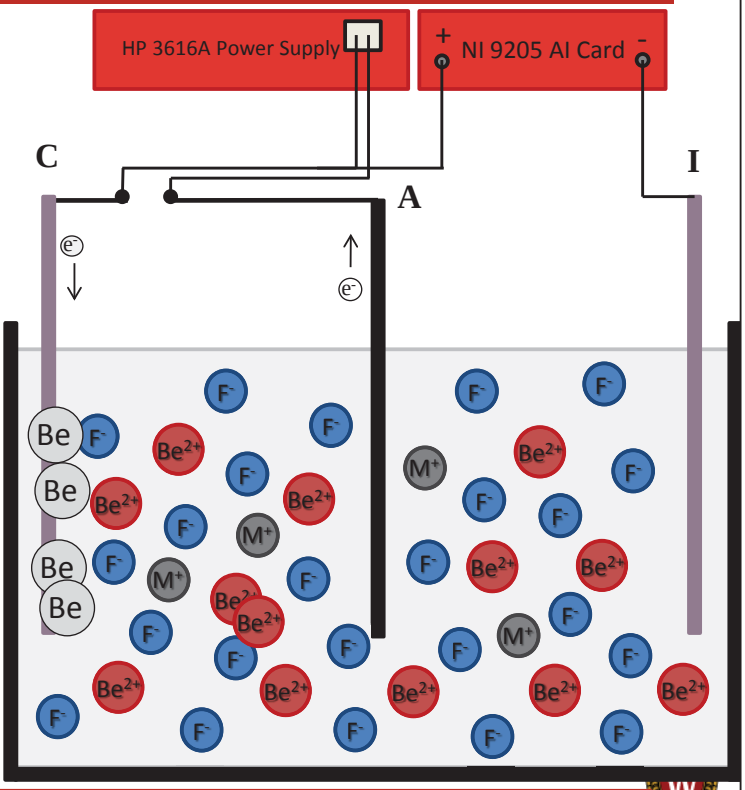
Redox Probe Measurement Process

Phase I: Plating



Phase II: Voltage Measurement





Plateau voltage of Be dissolution indicates redox potential of FLiBe

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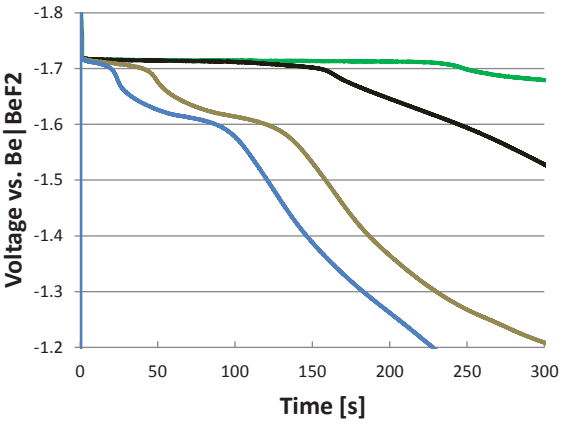

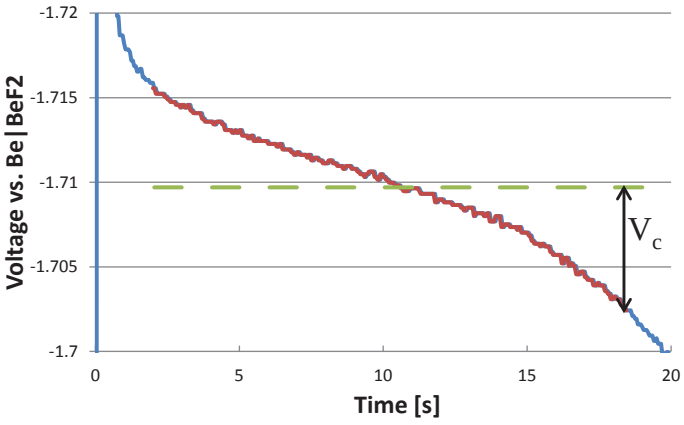

Procedure:

- Start after plating time + 1 second
- (current point – moving average) < V_c?
  - If true, move on to next point, update average

Blue: Original data
Red: Plateau data points
Green: Average of red points
-Used as Redox Voltage

- Points collected and averaged until end point exceeds a set cutoff voltage, V_c.

Redox potential testing of purified, unreduced UW-made FLiBe

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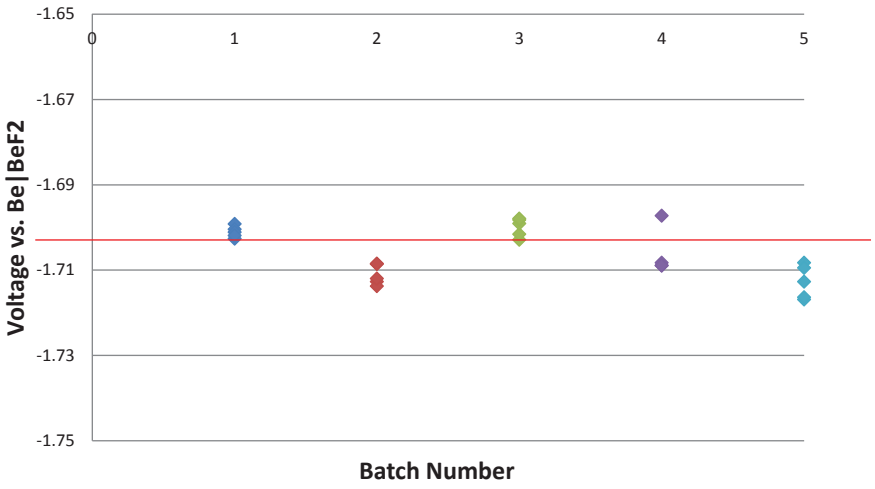

- 24 Measurements total, average of -1.708V with standard deviation of 6.2 mV
- Average standard deviation within each batch of 3.11 mV
- All batches will be mixed together prior to crucible loading

Production, purification and reduction of UW-made FLiBe

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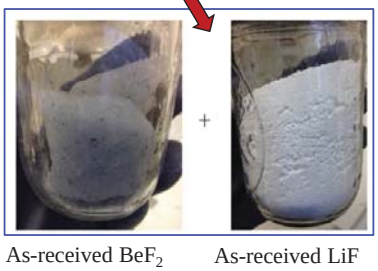
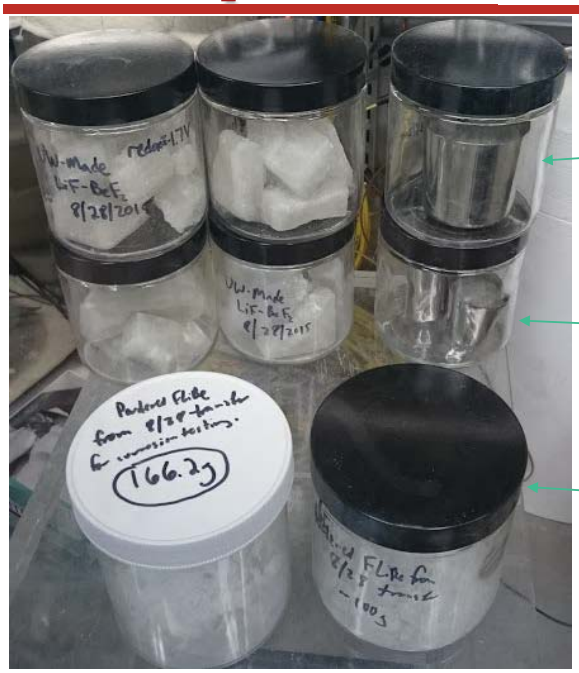
Raw Materials → $HF/H_2 \rightarrow H_2 \rightarrow$ Filtration, $600 - 630°C$ → UW Flibe → Beryllium and Filtration, $650°C$ → Beryllium Reduced



As-received BeF_2 As-received LiF

Video of FLiBe being poured from vessel into tray in glovebox

Glovebox inventory of FLiBe for all future experiments



Four nickel crucibles are fully filled and stored in jars.

Approximately 250 g of granulated salt was separated for one crucible in corrosion test.

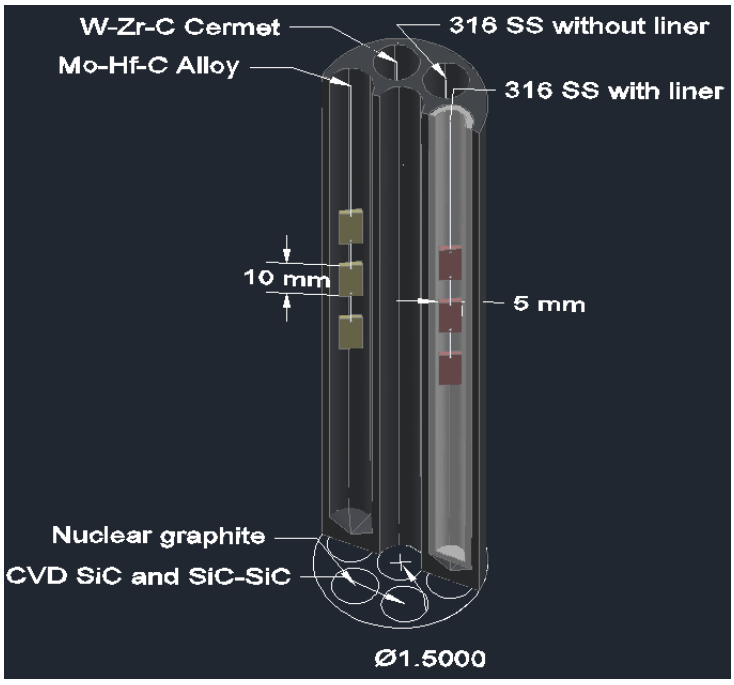
Total of 2.2 kg of UW-made FLiBe currently stored in glass jars in Ar glovebox.

Next static corrosion experiment will test multiple variables

- Metrics to test against corrosion:
  - **Redox potential effect**
    - HF/H_2 Purified salt (redox potential = -1.71V)
    - Beryllium Reduced salt (redox potential = -1.41V)
  - **Effect of carbon from IG-110 graphite crucible**
    - Corrosion test with or without liner for 316 SS
  - **New materials testing in FLiBe**
    - GA SiC-SiC
    - Mo-Hf-C alloy
    - Zr/C-W Cermet

Experimental Design of Static Corrosion Experiment in FLiBe

Crucible 1: -1.7V UW FLiBe	Crucible 2: -1.4V UW FLiBe
Hole 1: SiC → 3x SiC-SiC samples, 1x bulk SiC sample	
Hole 2: Nuclear Graphite → 3x matrix graphite samples, 1x IG-110 sample	
Hole 3: No liner. Mo-Hf-C → 3x Mo-Hf-C alloy samples with Mo wire suspension*	
Hole 4: No liner. W-Zr-C → 3x W-Zr-C Cermet samples with W wire suspension*	
Hole 5: No liner. 316 SS → 3x 316 samples with SS wire suspension*	
Hole 6: With 316 SS liner. 316 SS → 3x 316 samples with SS wire suspension*	



*Avoid dissimilar materials in contact in FLiBe wherever possible

Immediate Future Work

- Flow-assisted corrosion in natural circulation FLiBe loop
  - Corrosion samples in hot and cold legs of loop
  - Thermo-physical properties of FLiBe can be measured
  - Surge tank on top of loop for in-situ salt measurements and chemical control

FLiBe Natural Convection Loop 1/2

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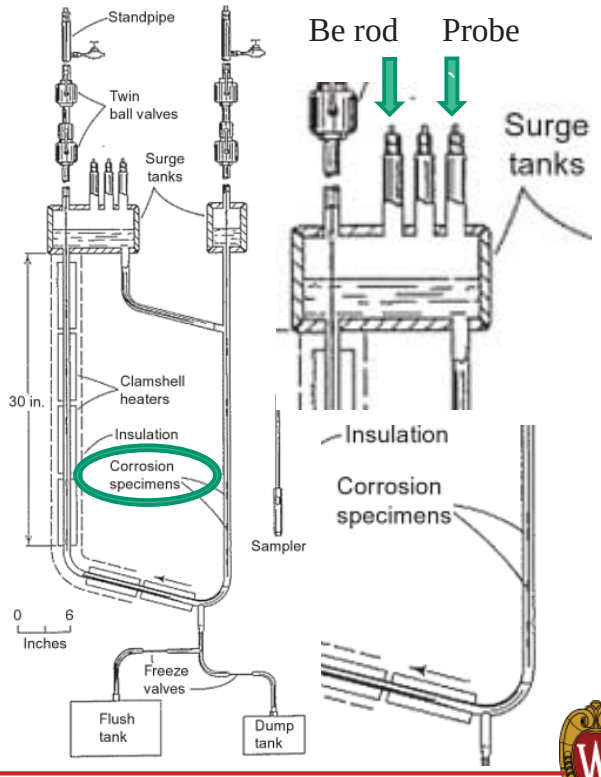
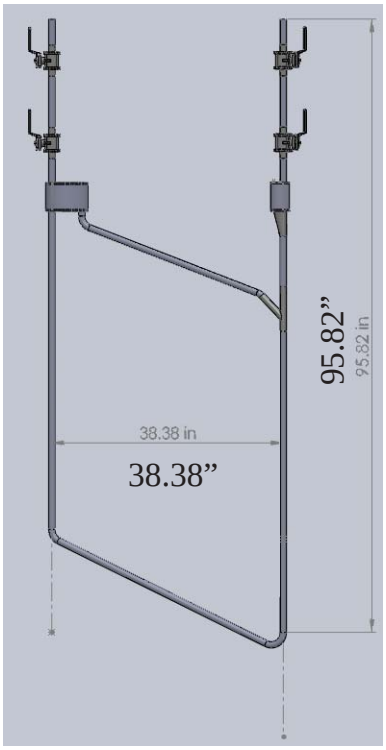
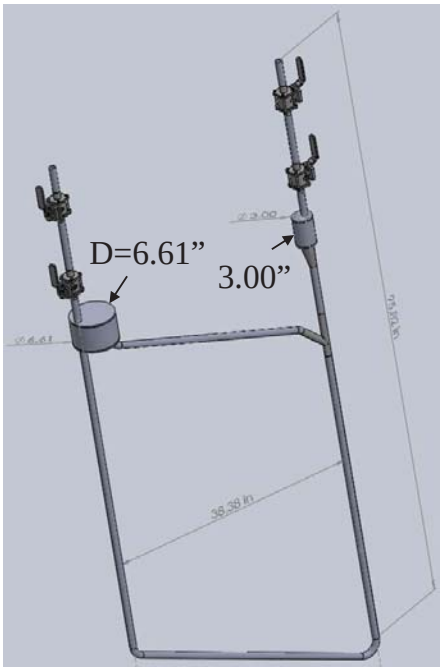
- FLiBe natural convection loop to be built
  - Incorporate Be-addition, redox measurement, and corrosion tests.
  - Use ports in surge tank for:
    1. Sacrificial Be rod with bellow
    2. 3-electrode redox probe
    3. Port-hole
    4. Anything else?



FLiBe Natural Convection Loop 2/2

- 1” OD Stainless Steel tubing
  - Composition matching important
- Two double ball valves to support in-loop corrosion tests.





Thank you for your attention!

Questions?

Workshop on Tritium Control and Capture in Salt Cooled Fission and Fusion Reactors

Salt Lake City
10.27.2015 & 10.28.2015

The Effect of Hydrogen on Tritium Control in Molten Salt System

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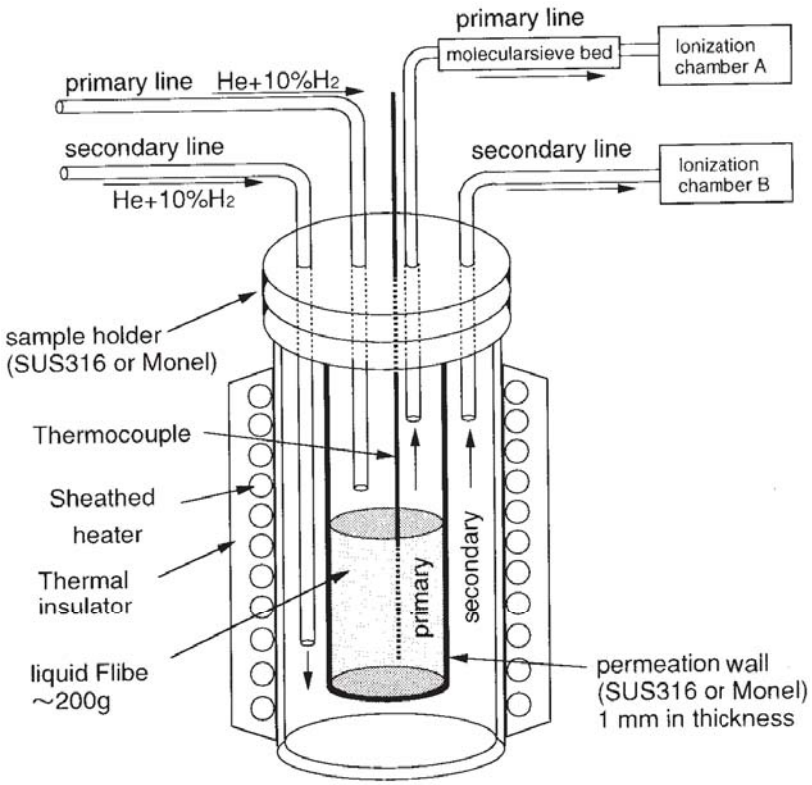
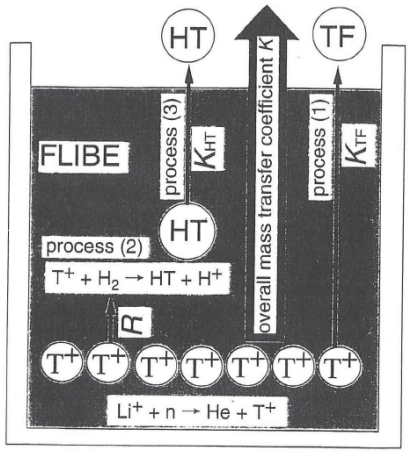
Tritium Chemical Form with He/H_2 Sparging



Chemical Form of Released Tritium with He/H_2 Sparging



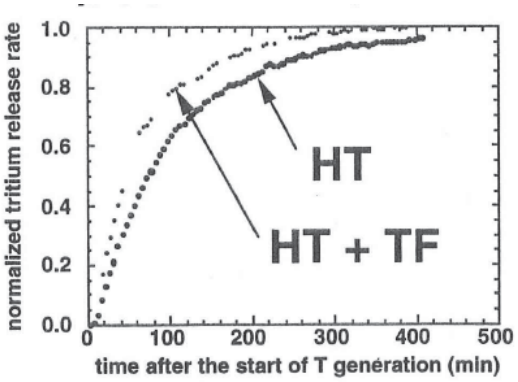


Fig.4(a) Tritium chemical form at 873K with H_2 purge gas.

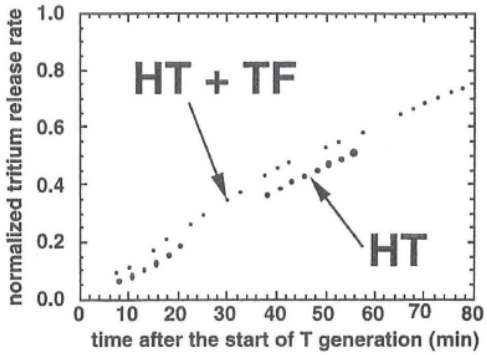


Fig.4(b) Tritium chemical form at 873K with He+1%H_2 purge gas.

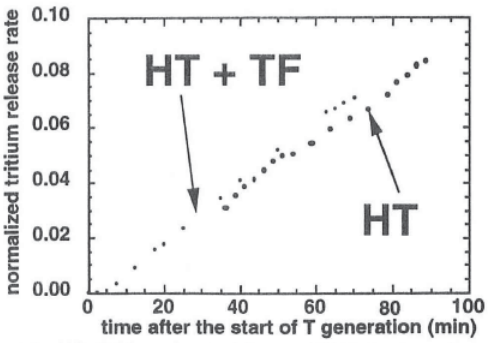


Fig.4(d) Tritium chemical form at 873K after long-time (24-26 days) dehumidification with He purge gas.

Permeation of Tritium through Structural Wall

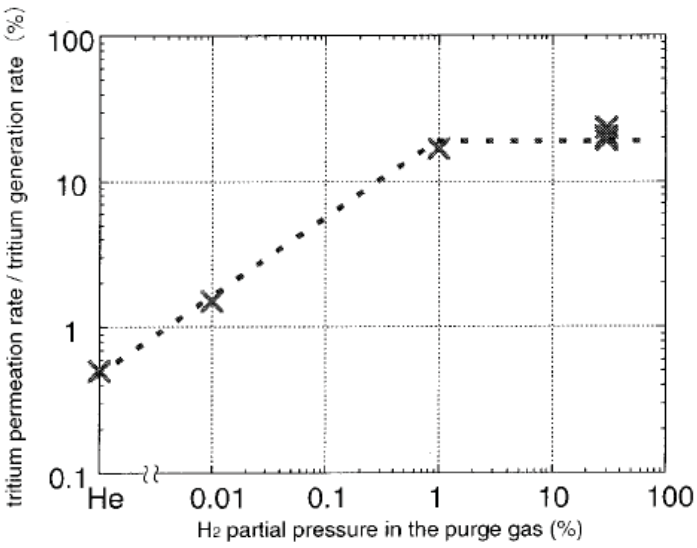


Fig. 3. The ratio of tritium permeation rate to tritium generation rate at steady state.

1. Would it be useful in the FHR system to sparge with He/H2, for tritium control?
2. How does the diffusion coefficient differ between TF and HT? Is it important to differentiate between the two?

H_2 Treatment of Graphite

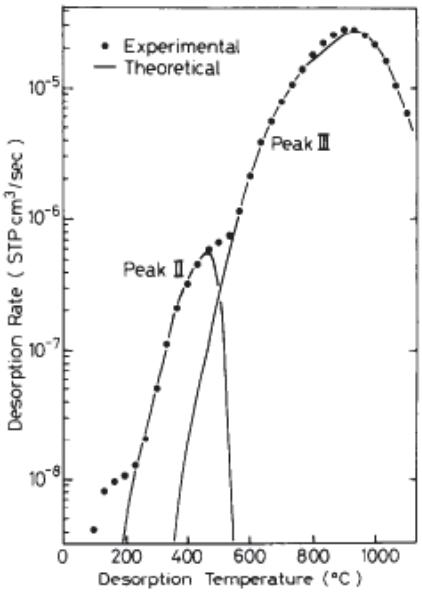
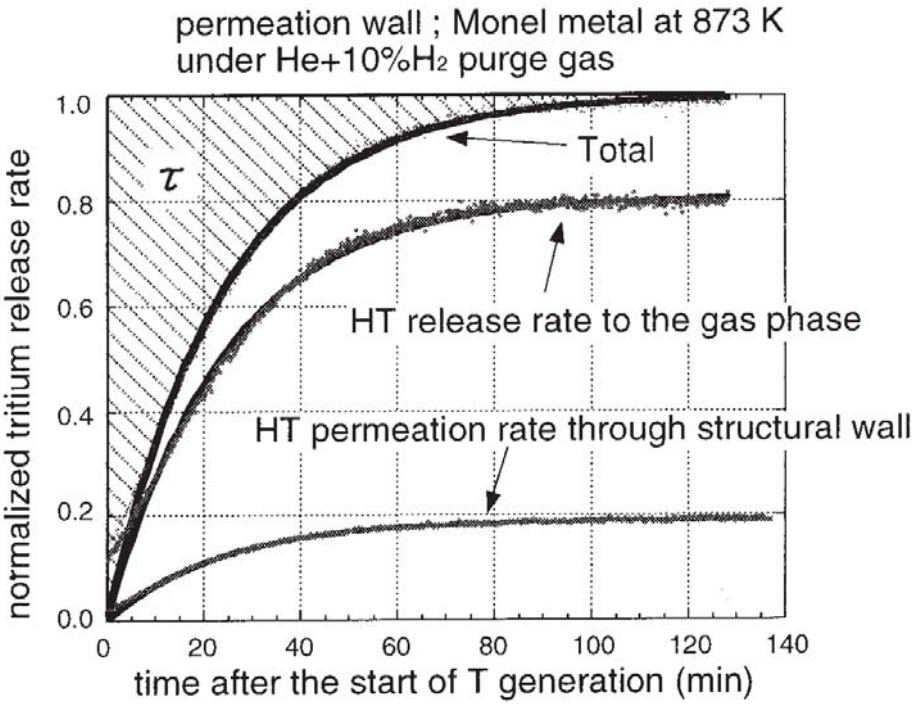


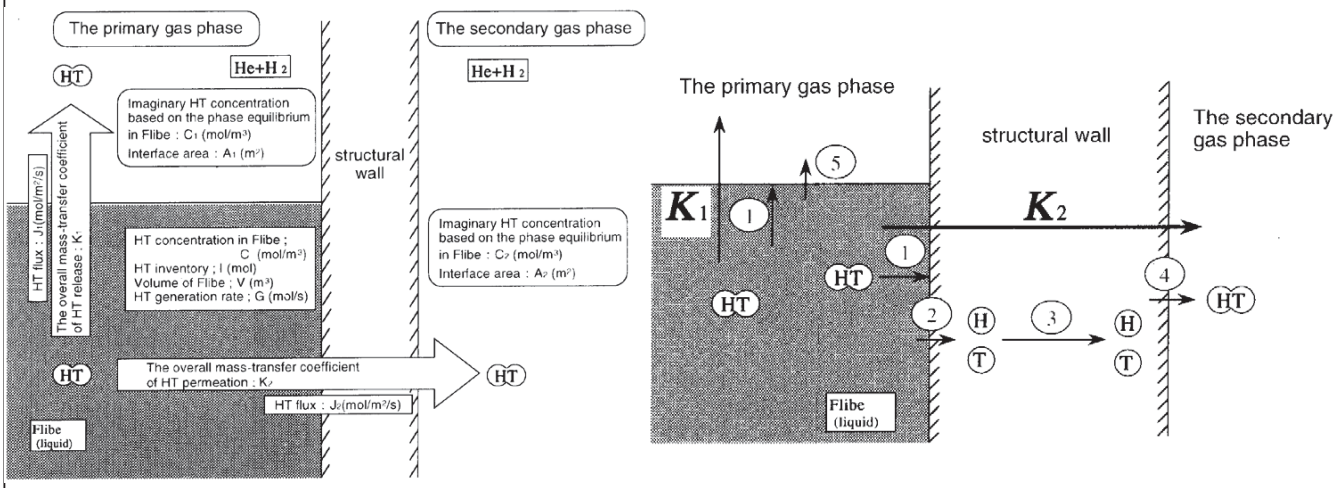
Fig. 6. Thermal desorption curve derived from diffusion analysis.

1. Would the fuel element be H2 treated to remove oxides before introducing in the salt?

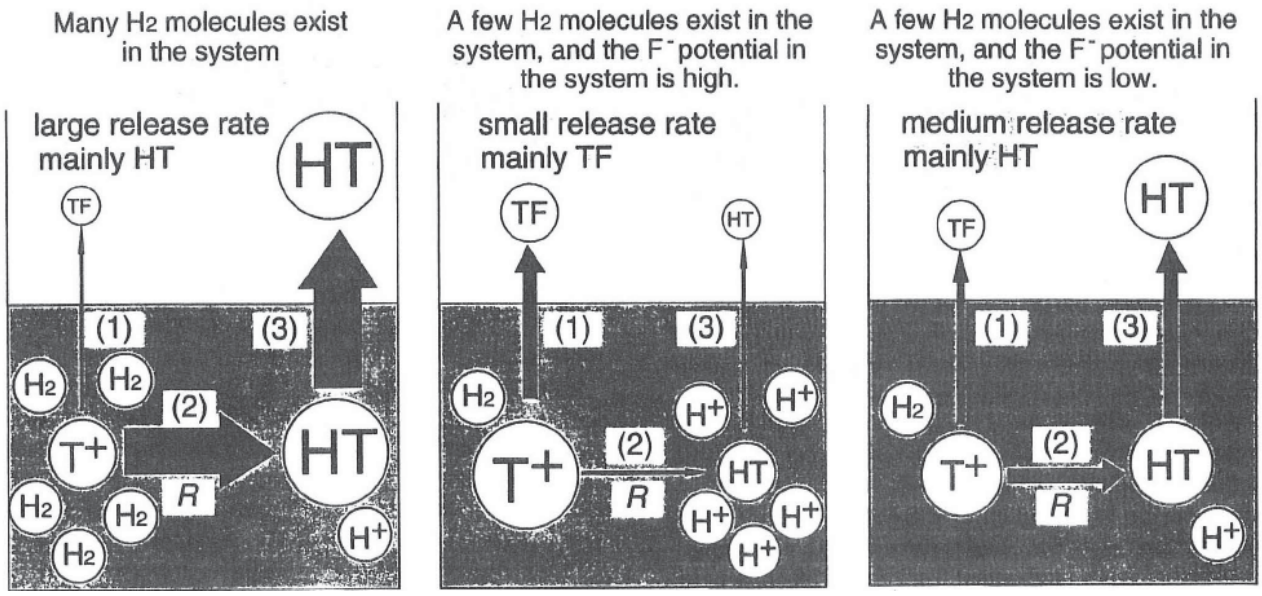
Backup Slides



Backup Slides – HT release and HT permeation



Mechanism of Tritium Release From Flibe



Workshop on Tritium Control and Capture in Salt Cooled Fission and Fusion Reactors

Salt Lake City
10.27.2015 & 10.28.2015

Experimental Work on Hydrogen Transport Analysis in Flibe-Graphite System

Huali Wu, Nisarg, Patel, Jee-hoon kim, Jayeesh Bakshi
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DEPARTMENT OF
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UNIVERSITY OF WISCONSIN-MADISON

Research Topic 1: Matrix Graphite Characterization

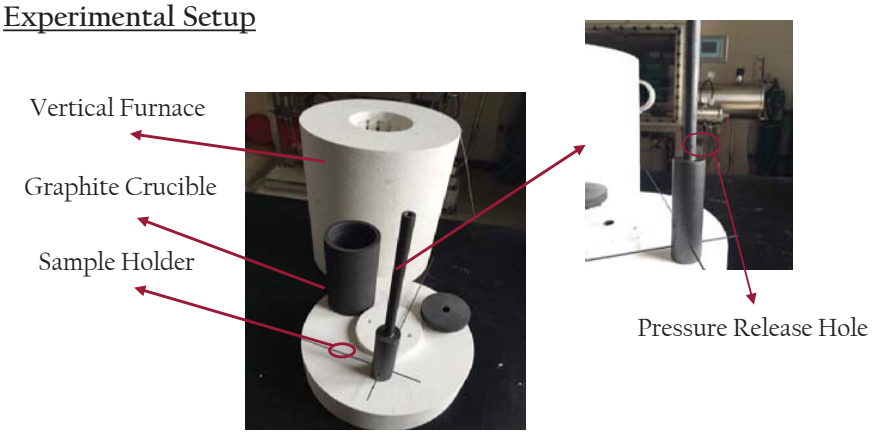
Goal: 1) To study matrix graphite microstructures– **density, porosity, pore distribution, specific surface area, graphitization, etc**; 2) To understand the difference between matrix graphite and nuclear graphite

Graphite Characterization	
Property	**Technique**
Density	Apparent Density
	Gas Pycnometer
Porosity	Numerical Calculation
	BET(Nitrogen)
	Matlab Image Analysis
Pore Distribution	Mercury Porosimetry
Surface Area	BET(Nitrogen)
Graphitization (d_{002})	X-ray Diffraction
In-plane Crystalline Size(nm)	Raman
Other Techniques	SEM
	Optical Microscopy

Research Topic 2: Static Salt Infiltration into Graphite

Goal: 1) to investigate salt infiltration process in graphite 2) to understand how graphite salt interaction will affect Fluoride-salt purity

Experimental Setup



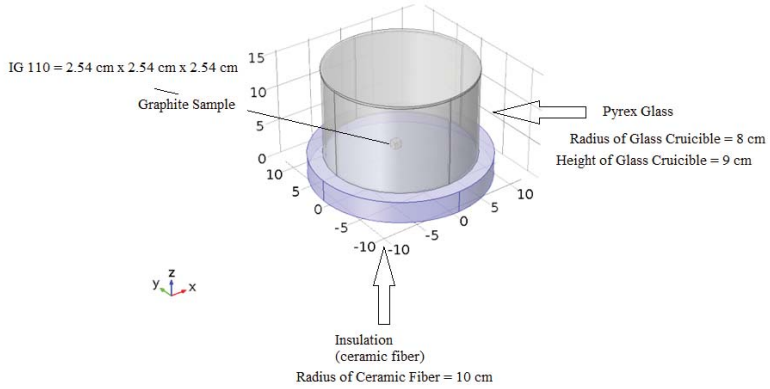
Sample (1200 grit polished & DI water ultrasonic cleaned)

NG: 0.0915g
MG: 0.0477g

Research Topic 3: Contact Angle Measurement-Jayeesh

Goal: 1) to investigate whether surface interaction between graphite and flibe will affect flibe intrusion in graphite

Experimental Setup



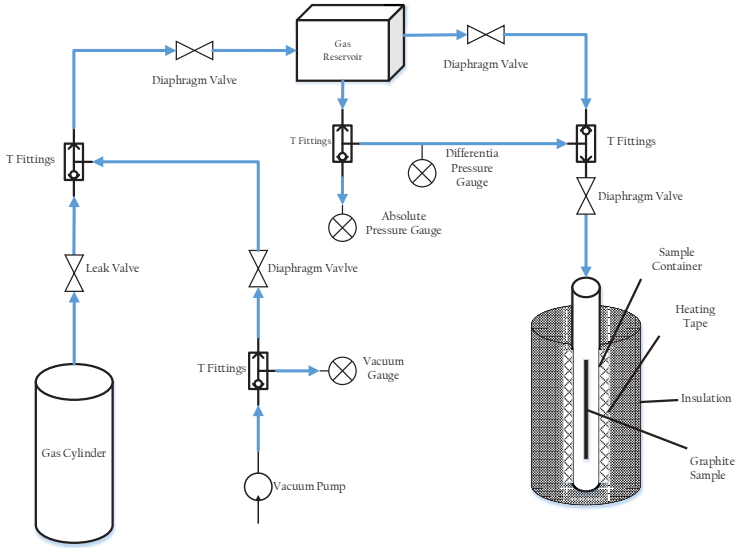
Resistance Analysis

Resistance Measurements of IG-110 Samples					
Sample No	Length (inches)	Width (inches)	Resistance (Ω)	Resistivity (Ω inch)	Resistivity (Ω meter)
1	1.297	1.3082	0.4	0.4328	1.099×10^{-2}
			0.4	0.541	1.374×10^{-2}

Research Topic 4: Hydrogen-Graphite Experiment (Constant Volume Method)

Goal: 1) to study hydrogen isotope transport phenomena in matrix graphite 2) to help to study hydrogen behavior in flibe-graphite system in the future (saturation-limited or diffusion-rate-limited)

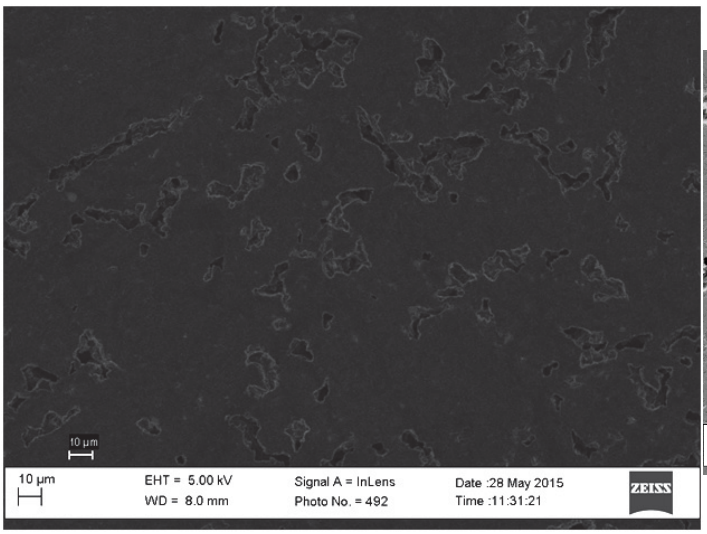
Constant Volume Method

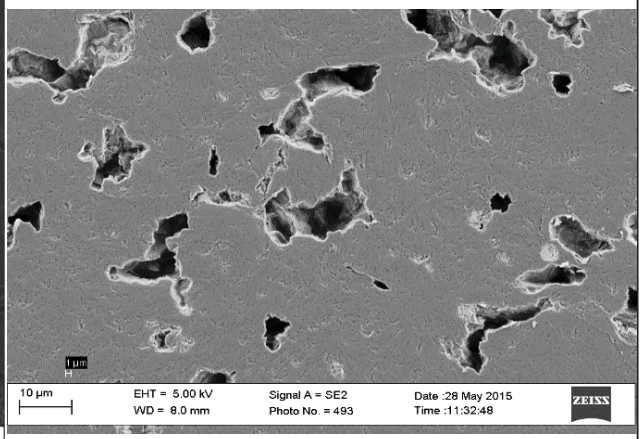


Questions to be Answered

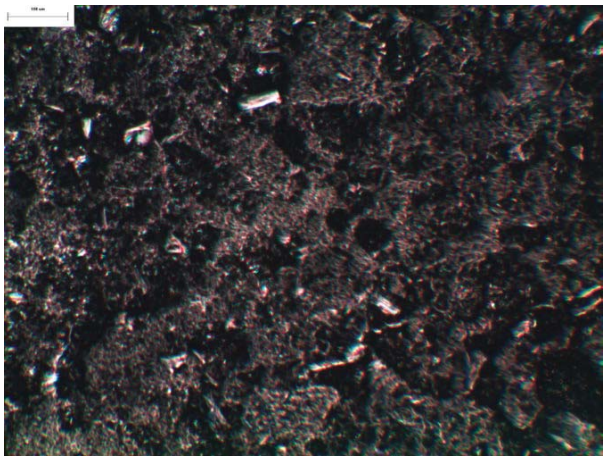
- Graphite Characterization:
  - Q 1: What physical properties are important for characterizing tritium transport?
  - Q 2: The effect of hydrogen baking & oxidization of graphite?
- Flibe Intrusion Experiment
  - Q1: Will salt have significant intrusion for modern graphite?
  - Q2: Any other post experiment measurements is recommended?
- Hydrogen-Graphite Experiment
  - Q 1: Hydrogen leak problem through the whole system?
  - Q 2: Total hydrogen and hydrogen depth profile measurement?
  - Q 3: How neutron irradiation will affect hydrogen transport in graphite?
- Modeling:
  - Q1: Appropriate software for pebble, core, system level simulation?

SEM Image of A3

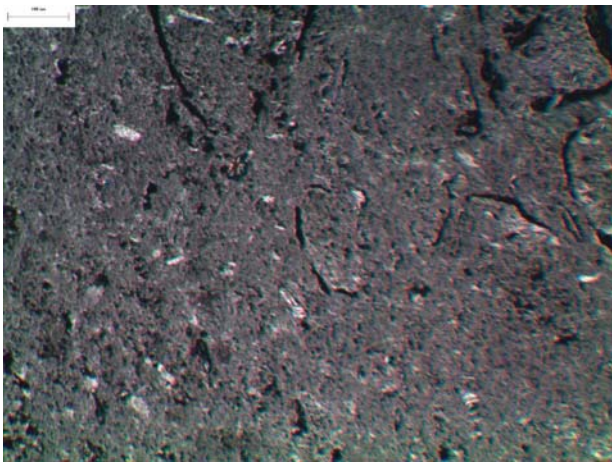




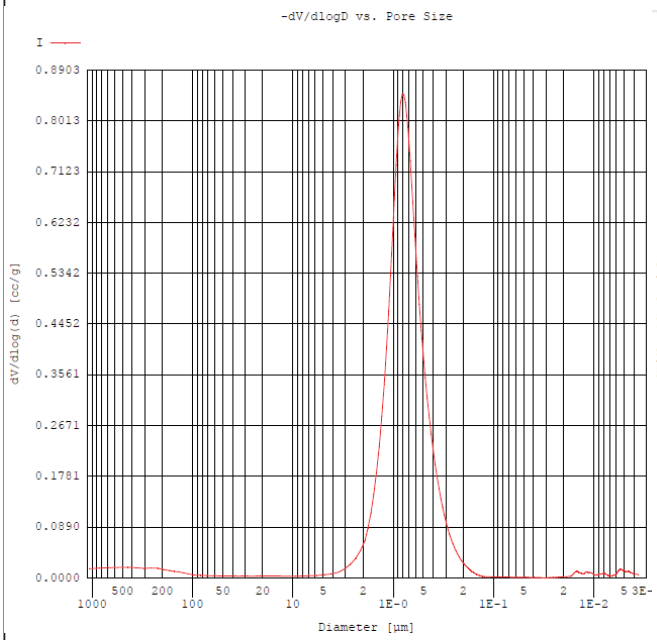
Optical Microscopy Visual Image



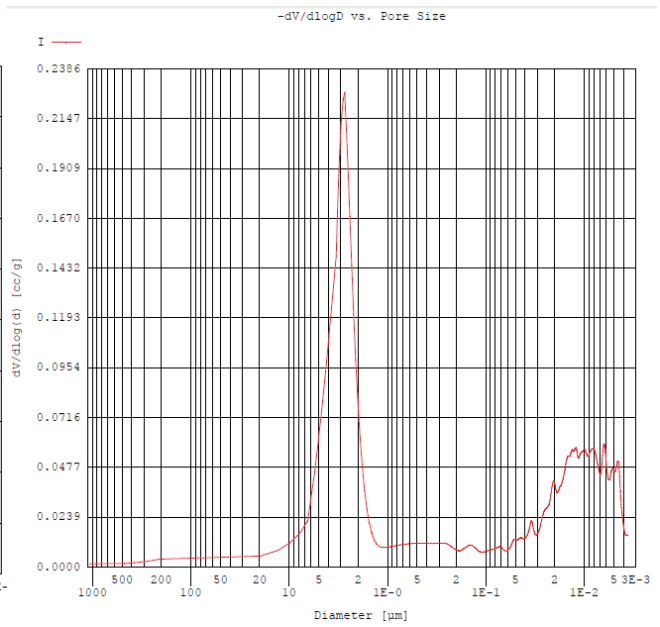
A3 Matrix Graphite



IG-110 Nuclear Graphite

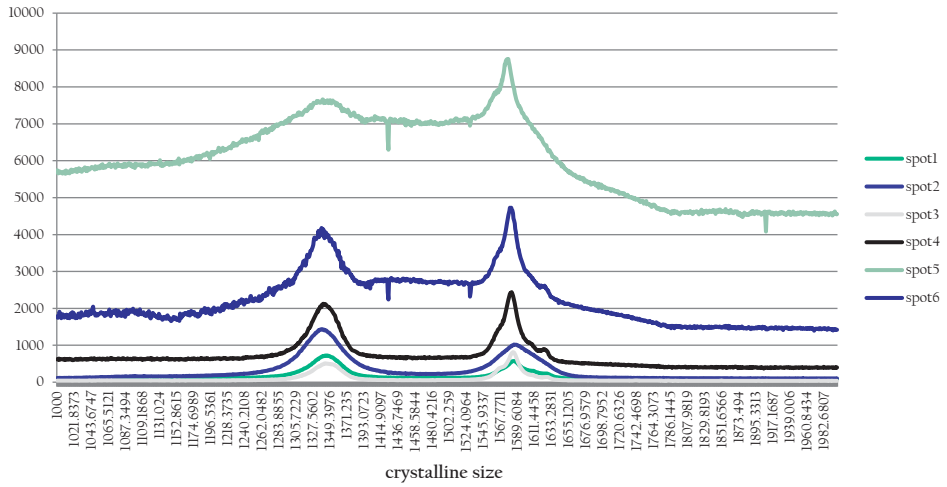


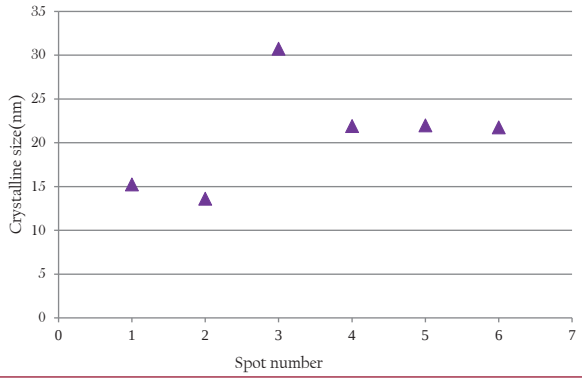
Pore Size Distribution of A3



Pore Size Distribution of IG-110

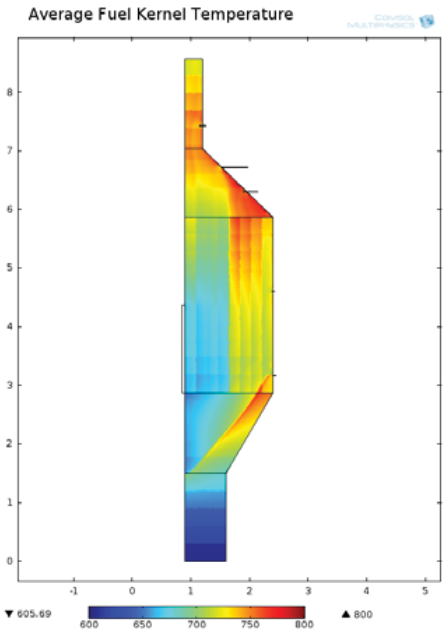
Raman Shifts





Research Topic 4: Hydrogen-Graphite Simulation on different Scale

- COMSOL is used for hydrogen transport simulation on **pebble** scale and also can be used for **core** scale, as shown in the Fig.1

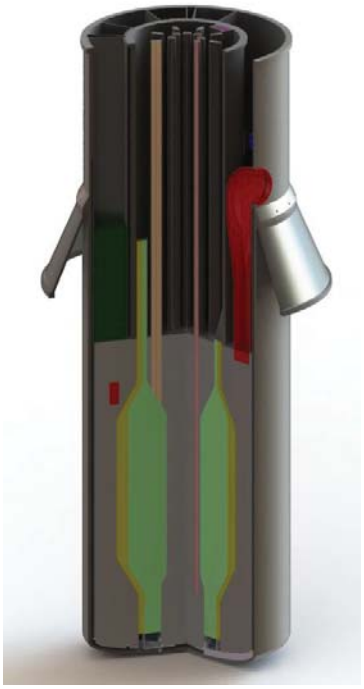


Q 1: Other modeling software for pebble, core, system scale?

Tritium Control Using Carbon Outside of Core

Stephen T Lam

Charles Forsberg
Ron Ballinger



Massachusetts Institute of Technology

Tritium Overview

Generation

- Thermal neutron transmutation of Li-6

$$^{6}\mathrm{LiF} + n \rightarrow {}^{4}_{2}\mathrm{He} + {}^{3}_{1}\mathrm{HF}$$

- Initially 0.005 wt. % Li-6 in Flibe consumed but is continually produced by Be-9 transmutation:

$$^{9}_{4}\mathrm{BeF_2} + n \rightarrow {}^{4}_{2}\mathrm{He} + {}^{6}_{2}\mathrm{He} + 2\mathrm{F}$$

$$^{6}_{2}\mathrm{He} \rightarrow {}^{6}_{3}\mathrm{Li} + e^{+} + \overline{\nu}_{e} \quad \left(t_{1/2} = 0.8\,\mathrm{sec}\right)$$

Concerns

- **Corrosion:** TF oxidizes chromium in stainless steel

$$2\mathrm{TF}_{(\mathrm{d})} + \mathrm{Cr}_{(\mathrm{s})} \rightarrow \mathrm{CrF}_{2(\mathrm{d})} + \mathrm{T}_{2(\mathrm{g})}$$

- **Release:** T_2 diffuses through piping and escapes to environment
- **Uncertainty:** Lack of industrial experience with FHR

Regulatory Tritium Limits

Limit: Concentration limits in Effluent

Target: ALARA. Similar magnitude to existing commercial reactors

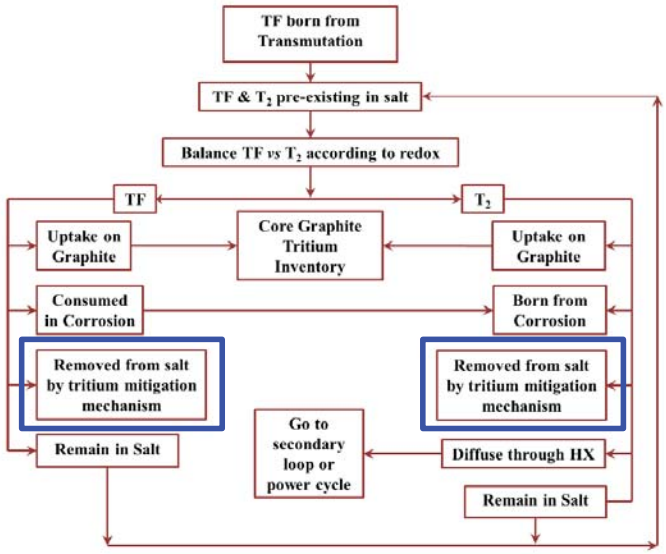
	Regulation	Annual Radiation Dose (mrem)	Annual Radiation Dose (mSv)	Effluent Concentration Air (µCi/ml)	Effluent Concentration Air (Bq/ml)	Effluent Concentration Water (µCi/ml)	Effluent Concentration Water (Bq/ml)
Limit	10 CFR 20.1301(a)1	100	1	-	-	-	-
	Table 2 of Appendix B to 10 CFR 20	50	0.5	1E-7	3.7E-3	1E-3	37
Standard	10 CFR 20.1301(e)	25	0.25	(5E-8)[a]	(1.85E-3)[a]	(5E-4)[a]	(18.5)[a]
ALARA	Appendix I to 10 CFR 50	20 (β,air)	0.20	(4E-8)[a]	(1.48E-3)[a]	-	-
		3 (water)	0.03	-	-	1.5E-5	0.56
Drinking Water	EPA standard	4	0.04	-	-	2E-5	0.74

a. Calculated by assuming the linear relationship between the annual dose of 50 mrem and the values in Table 2 of Appendix B of 10 CFR 20.
ALARA = as low as reasonably achievable
CFR = Code of Federal Regulations

"Sherman, S.R. Adams, T.M., "Tritium Barrier Materials and Separation Systems for the NGNP," WRSC-STI-2008000358, Rev.1, Savannah River National Laboratory, (2008)."

Tritium Modeling in FHR

Figure 1:TRIDENT Model Overview



*"J. Stempien, "A Model of Tritium Transport and Corrosion in Salt-Cooled Reactors," Cambridge, 2015."

TRIDENT

Tritium Diffusion EvolutioN and Transport

Time dependent tritium in FHR model developed at MIT*:

- Tritium generation in core
- TF and T_2 Speciation (Redox)
- In-core graphite Up-take
- Corrosion consumption & generation
- Diffusion in coolant, vessels, heat exchangers, reflectors
- **Mitigation mechanisms**
- Tritium release to environment

Tritium Production Rates

Estimated Base Case FHR Without Mitigation

	Tritium Production Rates [Ci/GWd]
BWR*	12.3
PWR*	13.9
HTGR*	18.5
FBR*	24.9
HWR*	1176
FHR	**Beginning of Life: 11,000** **Equilibrium: 2,900**

"J. Stempien, "Tritium Transport, Corrosion, and Fuel Performance Modeling in the Fluoride Salt-Cooled High-Temperature Reactor," MIT, Cambridge, 2015."
*Other reactor values calculated from data in:
"Management of waste containing tritium and carbon-14", International Atomic Energy Agency, Technical Reports Series No. 421, Vienna, 2004.

Mechanisms and Release Rates

Tritium Capture Evaluation

- Without mitigation, release to environment peak at **2410 Ci/EFPD**
- Three mitigation mechanisms were evaluated in TRIDENT

1. Stripping Column

- 10 stage counter current column with 20,000 L/hr STP stripping gas
- Release rate with column: **436 Ci/EFPD**

2. Permeation Window

- Shell with permeation tubes (Nickel) with 2x heat exchanger area
- Release rate with window: **800 Ci/EFPD**

3 . Carbon Absorber Bed

- 1.2(R)x3.85(H)m bed nuclear grade graphite ISO-88 with 1 regen/30 days
- Release rate with bed: **7.5 Ci/EFPD** ← **Similar to a PWR**

Simplified Tritium Removal Analysis

Numbers from "J. Stempien, "Tritium Transport, Corrosion, and Fuel Performance Modeling in the Fluoride Salt-Cooled High-Temperature Reactor," MIT, Cambridge, 2015."

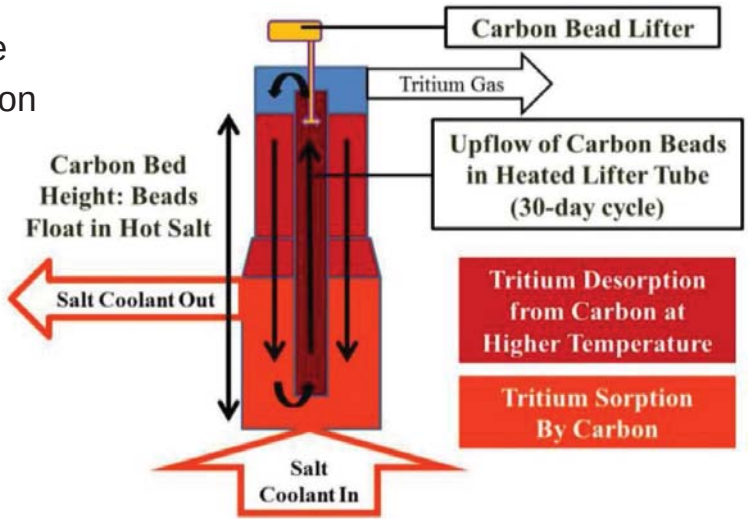
Carbon Absorber Bed

Concept

- Counter-current pebble bed absorber for coolant from core
- Continuous on-line regeneration
- Example: High temperature tritium de-gassing

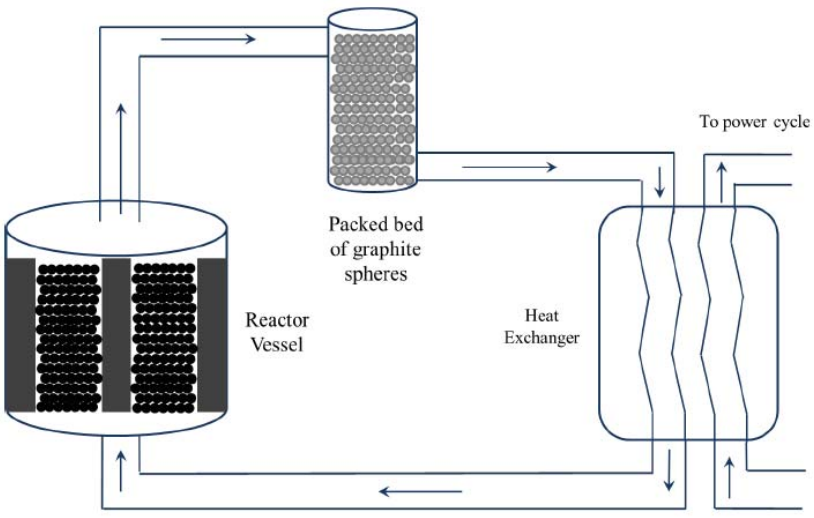
Specifications

- Bed size and flow rate (% of total primary molten salt flow)
- Operating temperatures
- Absorbent partial pressures
- Carbon type and area



"J. Stempien, "A Model of Tritium Transport and Corrosion in Salt-Cooled Reactors," Cambridge, 2015."

Graphite Bed Location



Schematic of a PB-FHR Continuously regenerated carbon bed

Location

- Primary system before heat exchanger
- Full or partial flow

Current Modelling

- 1-D tritium diffusion through molten salt
- Graphite capacity limited
- **Graphite ISO-88 (Nuclear grade)**

"J. Stempien, "Tritium Transport, Corrosion, and Fuel Performance Modeling in the Fluoride Salt-Cooled High-Temperature Reactor," MIT, Cambridge, 2015."

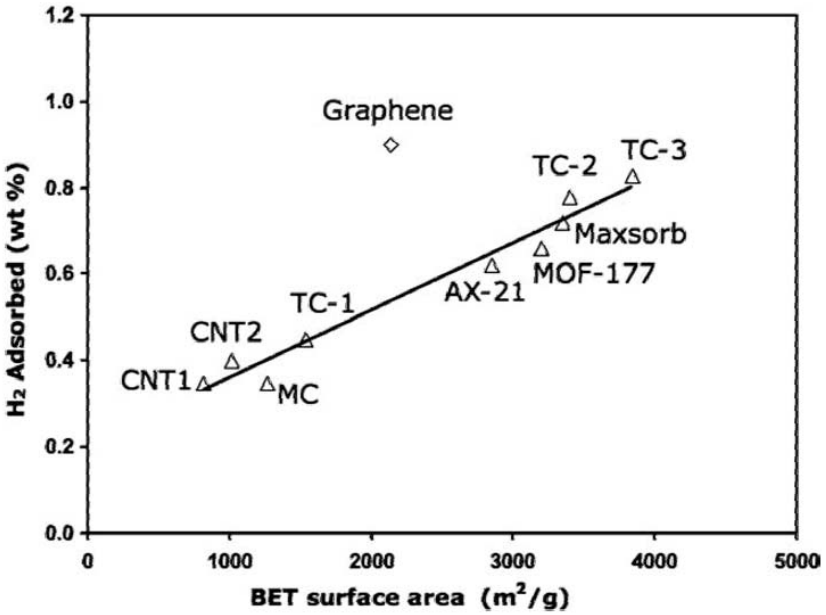
Choice of Absorbent

Considerations

- Non-nuclear grade (outside of core)
- Performance under operating conditions
- Long-term behavior in FHR

Carbon Properties

- Absorption rate
- Desorption rate
- Hydrogen Capacity:
  - BET Surface Area
  - **~1 vs. 3000 m²/g** (nuclear graphite vs. activated carbon)



L. Wang, N. R. Stuckert and R. T. Yang, "Unique Hydrogen Adsorption Properties of Graphene,"

H_2 Uptake Completion Time

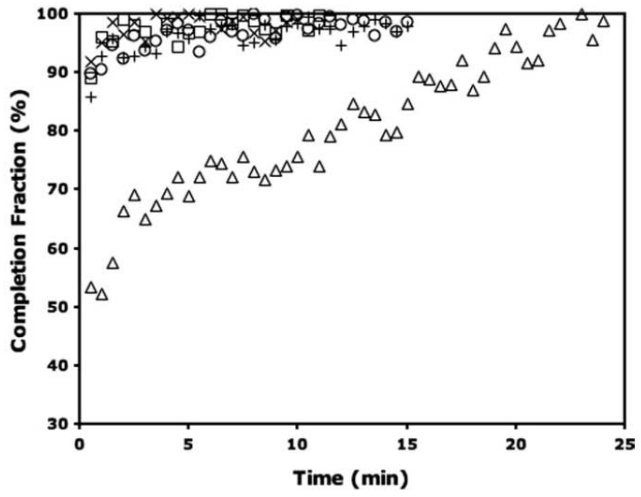


Figure 6. Fractional completion for hydrogen uptake at 298 K on AX-21 (×), Maxsorb (○), CNT (□), templated carbon (+), and graphene (Δ) at ~6 MPa end pressure.

L. Wang, N. R. Stuckert and R. T. Yang, "Unique Hydrogen Adsorption Properties of Graphene,"

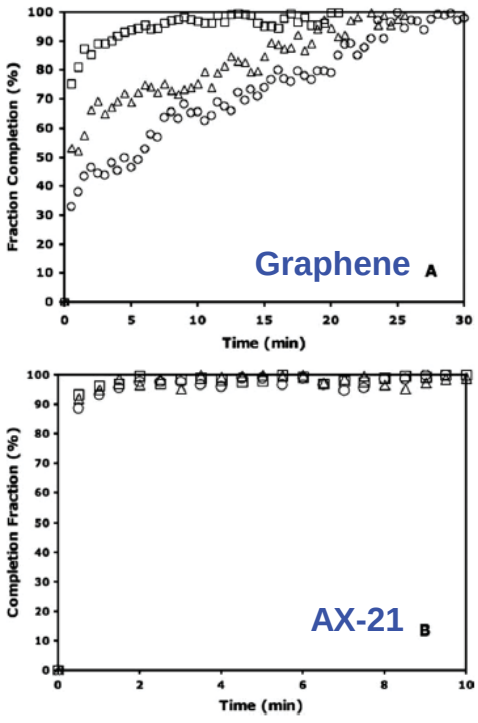
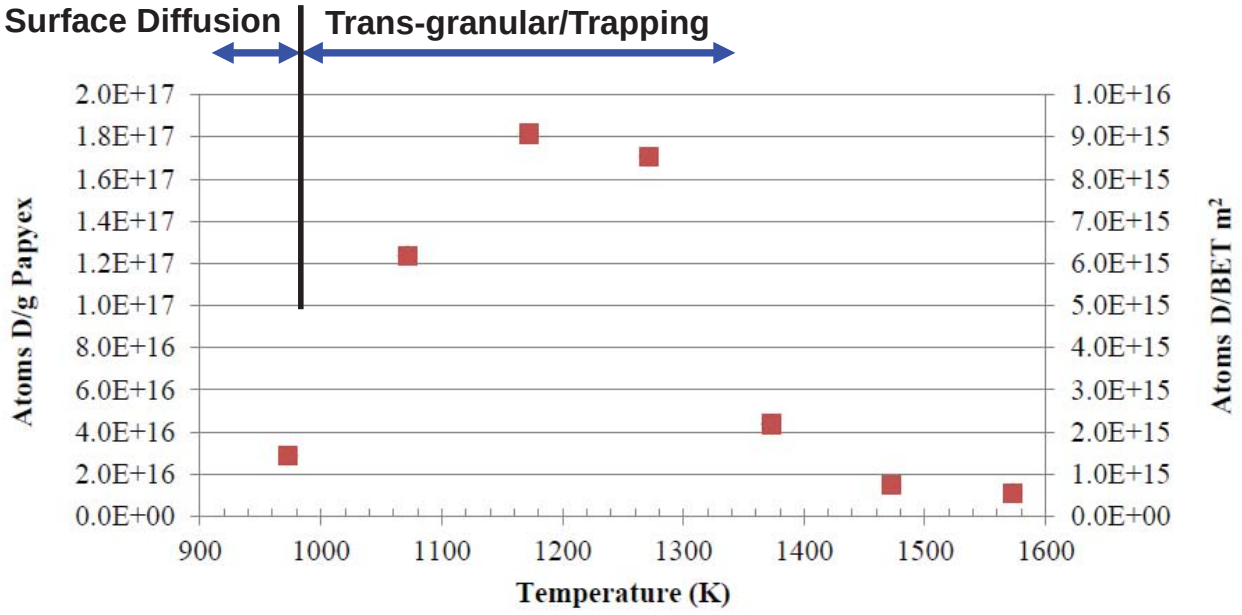


Figure 7. Adsorption fraction at 298 K on graphene (A) and AX-21 carbon (B) during each pressure ramp step at final pressures of: ~2 MPa (□), 6.2 MPa (Δ), and 7.9 MPa (○).

H_2 Capacity vs. Temperature



Specific capacity of Papyex graphite under deuterium gas only at 0.66 Pa.

Engineering implications: Column size, percent flow, regeneration

Massachusetts Institute of Technology

Data converted "J. Stempien, "Tritium Transport, Corrosion, and Fuel Performance Modeling in the Fluoride Salt-Cooled High-Temperature Reactor," MIT, Cambridge, 2015."
Original Data: CAUSEY, R.A., WILSON, K.L., "Retention of deuterium and tritium in Papyex graphite," *Journal of Nuclear Materials*. **138**, 57–64 (1986).

FHR Conditions

Temperatures*	
Coolant Freezing	459ºC (FliBe)
Operating Core Outlet	700ºC
ATWS	<800ºC
Coolant Boiling	**1400ºC (FliBe)**
Pressures (primary loop) *TRIDENT Simulation	
p_{T2} Unmitigated	3.3-20 Pa
p_{T2} with Graphite Capture	**0.03-0.08 Pa** (Peak release 7.5 Ci/GW/d)
p_{TF} Unmitigated	0.03-0.075 Pa
p_{TF} with Graphite Capture	**0.0027-0.0045 Pa** (Peak release 7.5 Ci/GW/d)

Challenge: Low pressure and high temperature data

* "C. Andreadas, et. Al. "Technical Description of the "Mark 1" Pebble-Bed Fluoride-Salt-Cooled High-Temperature Reactor (PB-FHR) Power Plant," University of California, Berkeley, Berkeley, 2014."

Massachusetts Institute of Technology

Next Steps

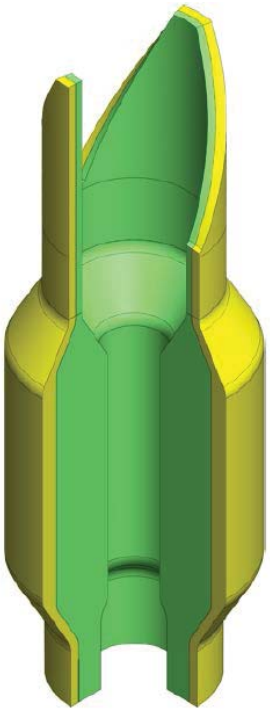
1. **Design and construct experiment for testing performance of different carbons in FHR conditions**
   - Vacuum chamber
   - Temperature and pressure controls, etc.

2. **Data collection to understand hydrogen uptake mechanics**
   - Absorption-Desorption kinetics (trapping, diffusion, etc.)
   - Behavior in high temperature exposure, cycling

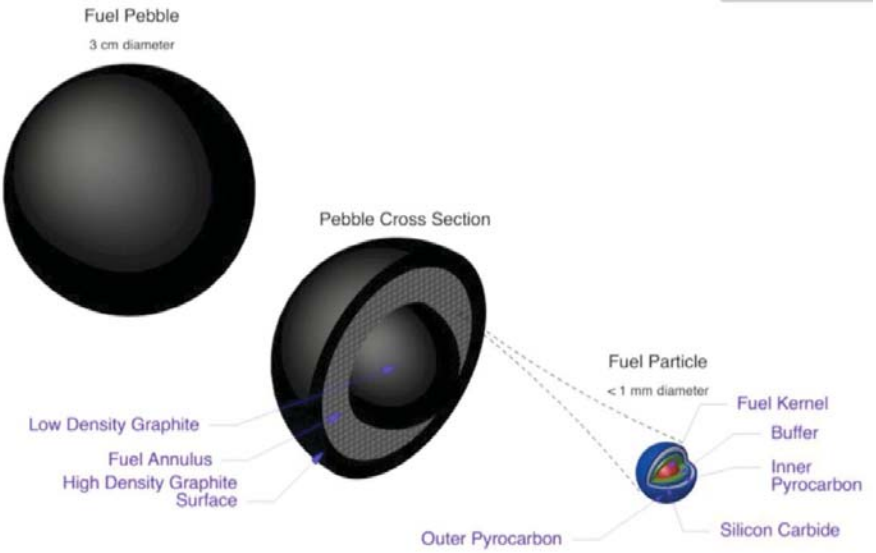
3. **Modeling and technology qualification**
   - Use new data to reduce model uncertainty
   - Improve predictive capability

Questions

In-Core Graphite



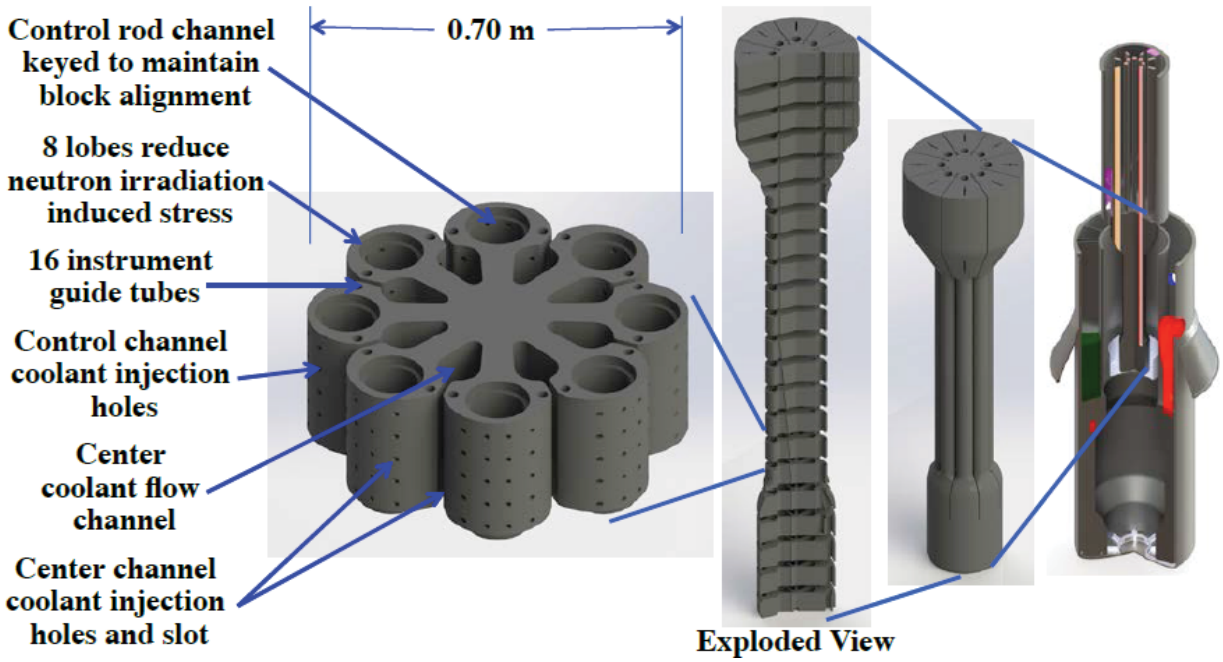
(Left) Mk1 pebble core geometry showing fuel pebble (green) and graphite reflector pebble (yellow) regions



(Above) A PB-FHR pebble fuel element

ANDREADES, C., CISNEROS, A.T., CHOI, J.K., CHONG, Y.K., FRATONI, M., HONG, S., ET AL., "Technical Description of the 'Mark 1' Pebble-Bed Fluoride-Salt-Cooled High-Temperature Reactor (PB-FHR) Power Plant," UCBTH-14-002, University of California-Berkeley, (2014).

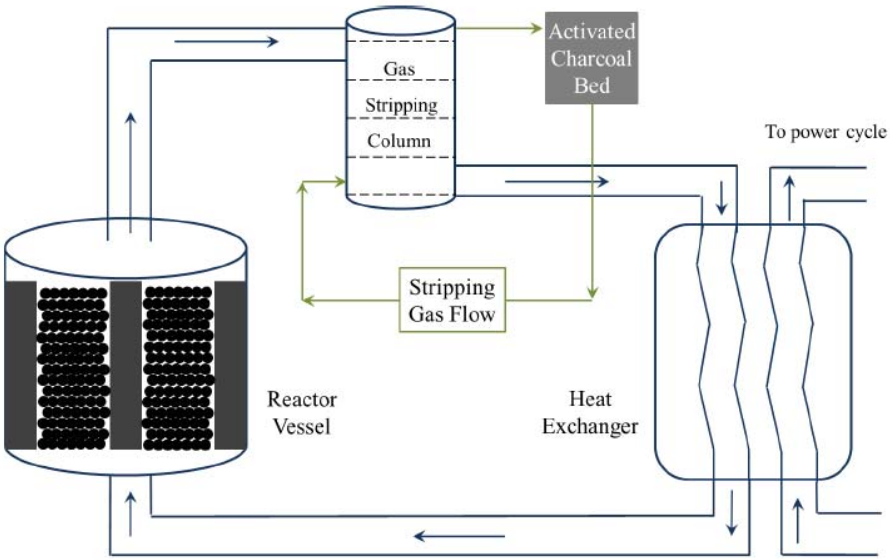
In-Core Graphite

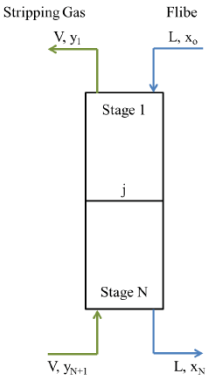


Replaceable Mk1 center graphite reflector

ANDREADES, C., CISNEROS, A.T., CHOI, J.K., CHONG, Y.K., FRATONI, M., HONG, S., ET AL., "Technical Description of the 'Mark 1' Pebble-Bed Fluoride-Salt-Cooled High-Temperature Reactor (PB-FHR) Power Plant," UCBTH-14-002, University of California-Berkeley, (2014).

Gas Stripping Column



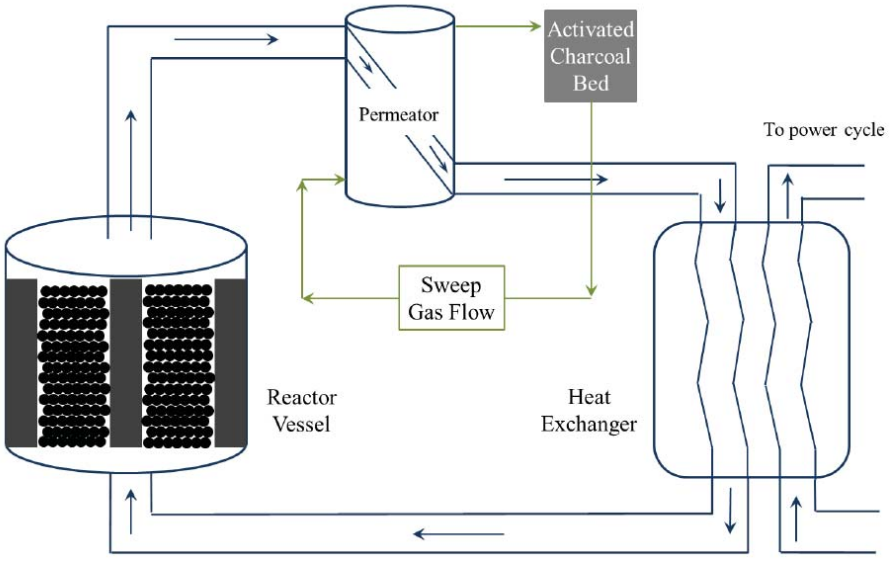


Equilibrium Stage

Schematic of a PB-FHR with multi-stage counter current gas stripper and charcoal bed

Numbers from "J. Stempien, "Tritium Transport, Corrosion, and Fuel Performance Modeling in the Fluoride Salt-Cooled High-Temperature Reactor," MIT, Cambridge, 2015."

Permeation Window

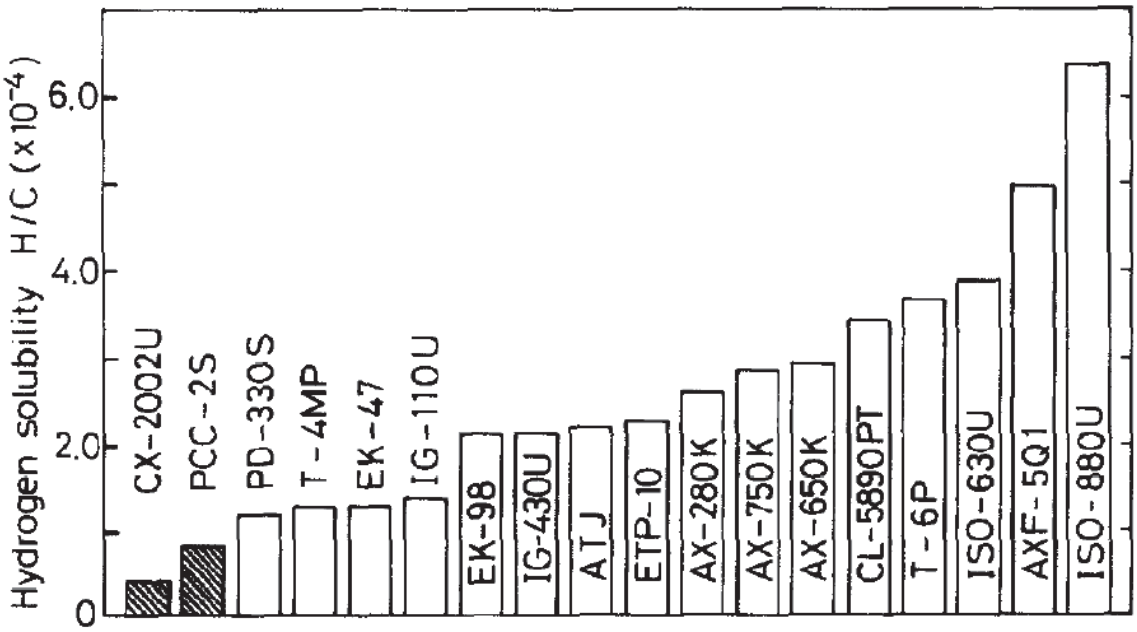


- **1-D diffusion through Ni Tubes**
- **20,000 m^2 surface area**
- **27,360 permeation tubes**
- **Permeation tubes OD: 0.00635**

Schematic of a PB-FHR Permeation Window and Activated Charcoal bed for gas recovery

"J. Stempien, "Tritium Transport, Corrosion, and Fuel Performance Modeling in the Fluoride Salt-Cooled High-Temperature Reactor," MIT, Cambridge, 2015."

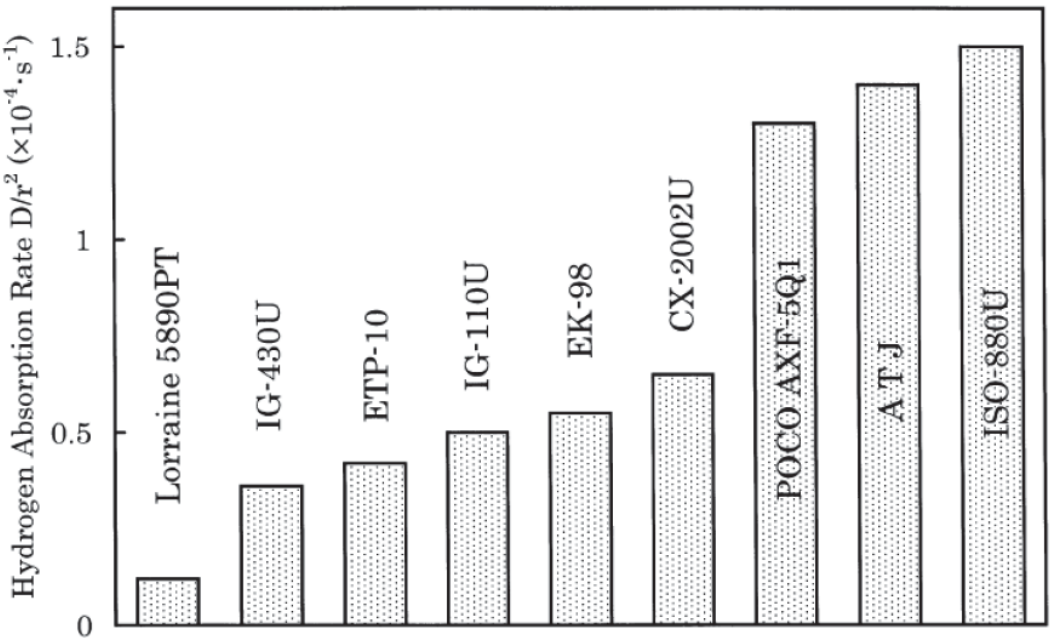
Hydrogen Solubility



Hydrogen solubility of different carbons at 1000 ^{o}C and 101 kPa

"Astumi, H., Iseki, M., Shikama, T., "Trapping and detrapping of hydrogen in carbon-based materials exposed to hydrogben gas,' Journal of Nuclera Materials. 212-215, 1478-1482 (1994) "

Hydrogen Absorption Rates

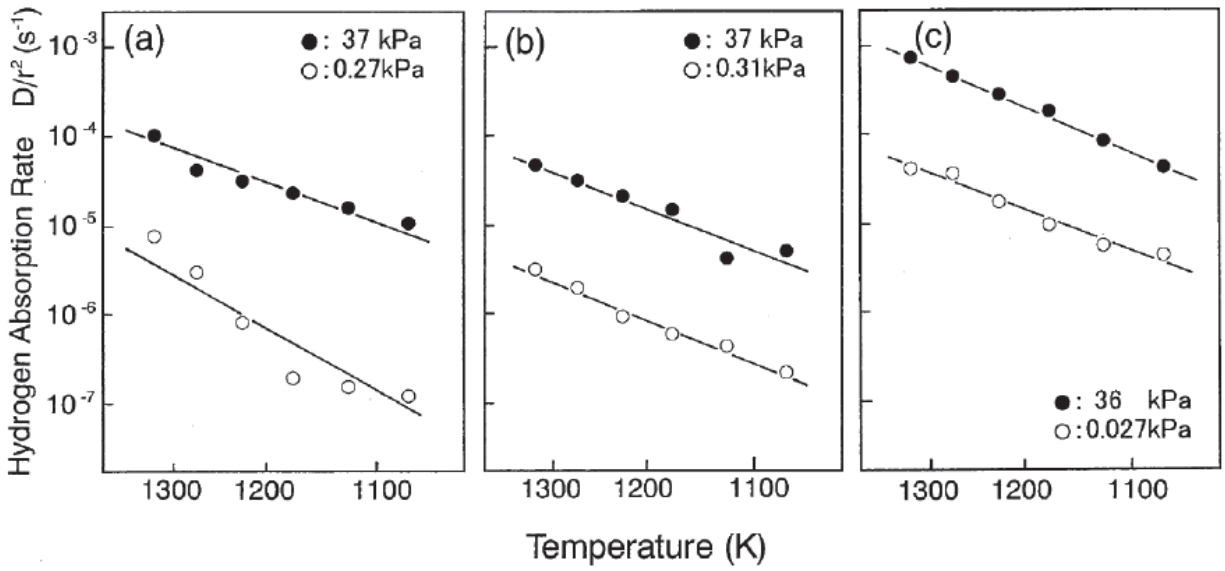


Hydrogen absorption rates in graphite and the CFC CX-2002U. T = 1273 K and P = 10 kPa.

ATSUMI, H., ISEKI, M., "Hydrogen absorption process into graphite and carbon materials," *Journal of Nuclear Materials*. **283–287, Part 2**, 1053–1056 (2000).

Absorption Rate vs. Temperature

- Increasing absorption **rate** with increasing temperature

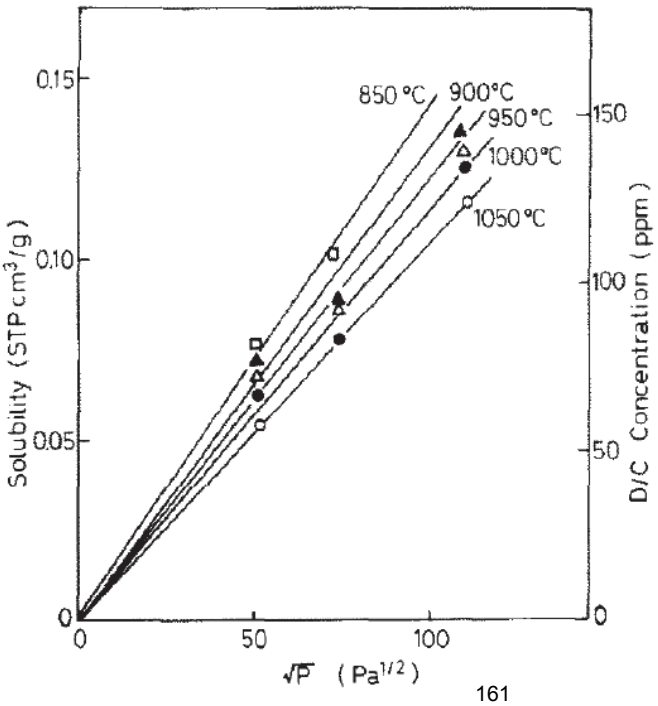


Hydrogen absorption rate in (a.) IG-110U, (b.) IG-430U, and (c.) ISO-880U.

ATSUMI, H., "Hydrogen bulk retention in graphiet and kinetics of diffusion," *Journal of Nuclear Materials*. **307-311**, 1466–1470 (2002).

Solubility vs. Temperature

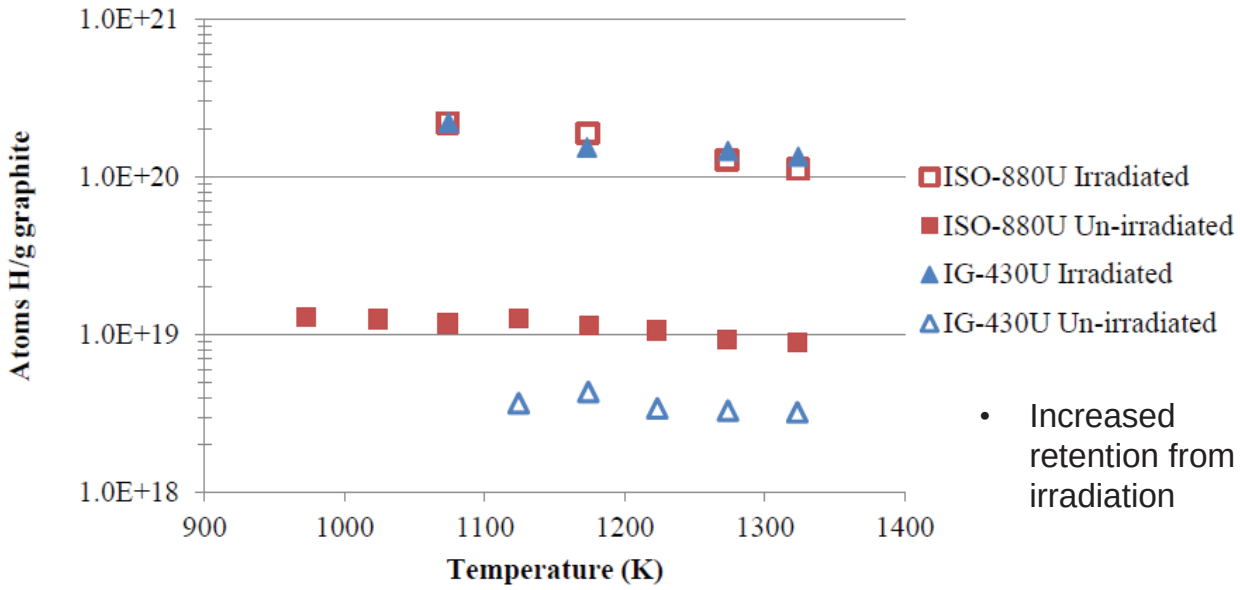
Solubility isotherms for deuterium adsorption on ISO-88 graphite



- Decreasing solubility with increasing temperature

ATSUMI, H., TOKURA, S., MIYAKE, M., "Absorption and desorption of deuterium on graphite at elevated temperatures," *Journal of Nuclear Materials*. **155–157, Part 1**, 241–245 (1988).

Tritium Retention vs. Irradiation



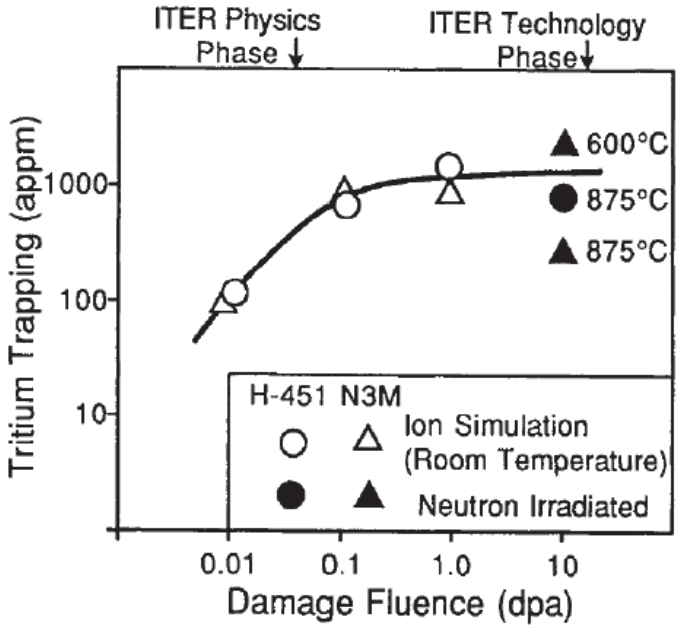
- Increased retention from irradiation

Hydrogen retention in ISO-880U and IG-430U. Irradiation was to 0.047 dpa.

Data converted "J. Stempien, "Tritium Transport, Corrosion, and Fuel Performance Modeling in the Fluoride Salt-Cooled High-Temperature Reactor," MIT, Cambridge, 2015."

Original: ATSUMI, H., TANABE, T., SHIKAMA, T., "Bulk hydrogen retention in neutron-irradiated graphite at elevated temperatures," *Journal of Nuclear Materials*. **390–391**, 581–584 (2009).

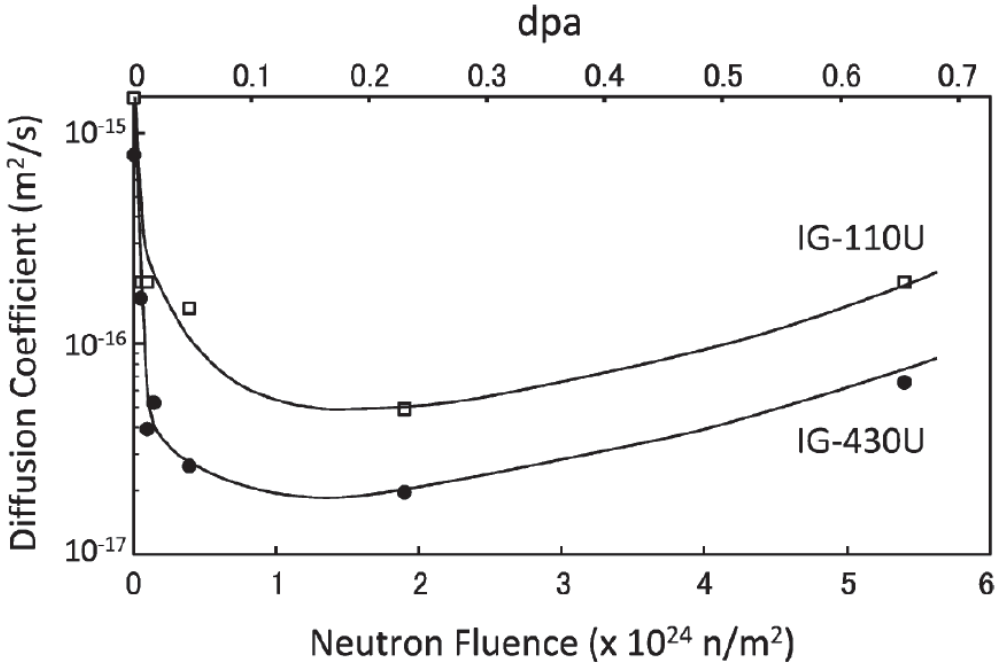
Tritium Trapping vs. Irradiation



Tritium Trap Concentration (atom ppm) for H-451 and N3M

CAUSEY, R.A., WILSON, K.L., WAMPLER, W.R., DOYLE, B.L., "The Effects of Neutron Irradiation on the Trapping of Tritium in Graphite," *Fusion Science and Technology*. **19**, 1585–1588 (1991).

Hydrogen Diffusion vs. Irradiation



Hydrogen Diffusion with Neutron Flux at 1273K and 10 kPa

Massachusetts Institute of Technology

ATSUMI, H., TANABE, T., SHIKAMA, T., "Hydrogen behavior in carbon and graphite before and after neutron irradiation – Trapping, diffusion and the simulation of bulk retention–," *Journal of Nuclear Materials*. **417**, 633–636 (2011).

Tritium permeation control and extraction- perspectives from fusion systems studies

Paul W. Humrickhouse

Workshop on Tritium Control and Capture in Salt-Cooled Fission and Fusion Reactors

Salt Lake City, Utah

October 28, 2015

www.inl.gov

Idaho National Laboratory

Outline

- Tritium in fission and fusion reactors: similarities and differences
- Overview of breeding concepts for fusion
- Tritium management in fusion
- Concepts for tritium extraction from liquids
- Permeation barriers
- Permeation scaling with pressure
- Conclusions and recommendations for future research

Tritium generation in fission and fusion reactors

- Tritium generation in salt-cooled fission reactors is large relative to other fission reactors, but several orders of magnitude less than fusion

	PWR[1]	CANDU[1]	Gas-cooled reactor[1]	Molten salt reactor[1]	ITER	FNSF	DEMO
T generated (kg/y)	0.000075	0.1	0.002	0.09	0.0042	1 - 10	100 - 167

- Fusion consumes ~55 kg of tritium per GW-year of fusion power, and must necessarily breed this amount from lithium
- Tritium is very mobile and will permeate through solids at high temperature; losses must be limited to < 20 Ci/day (very roughly 1 g/yr)
- A fusion reactor must recover and separate bred tritium for re-use as fuel; in a fission reactor is it a waste product
- Strategies for tritium permeation control and extraction investigated for fusion should apply to salt-cooled fission reactors

[1]H. Schmutz, INL/EXT-12-26758, 2012

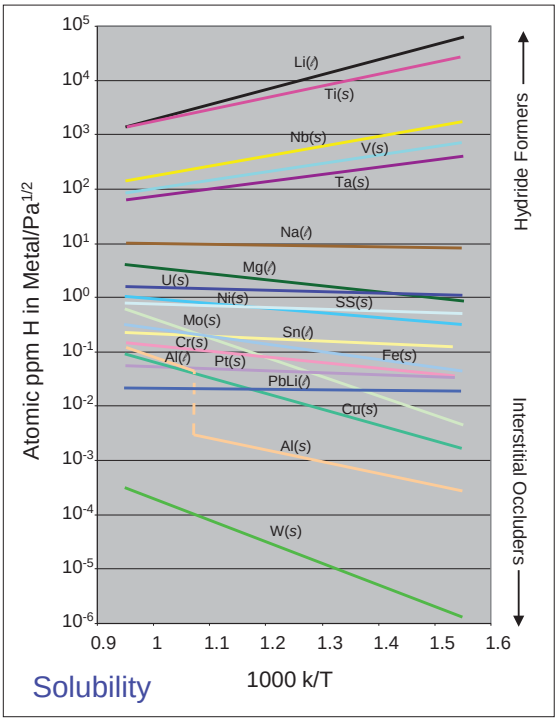
Tritium breeding materials in fusion

- Tritium is bred via neutron interactions with materials enriched in lithium-6
- Solid breeders (beds with ~1 mm diameter pebbles)
  - Li_4SiO_4 or Li_2TiO_3 ceramic breeder
  - Be or $Be_{12}Ti$ (lower chemical reactivity) neutron multiplier
- Liquid breeders
  - Li liquid metal
  - PbLi eutectic (Pb is a multiplier; less chemically reactive than Li)
  - FLiBe (requires additional Be multiplier)
- Current research is focused on solid ceramic (European, Japanese, Korean, Chinese, and Indian TBMs in ITER) or PbLi breeders (EU, Indian TBMs)
- The US does not have a TBM program, but our reference design is based on a PbLi breeder

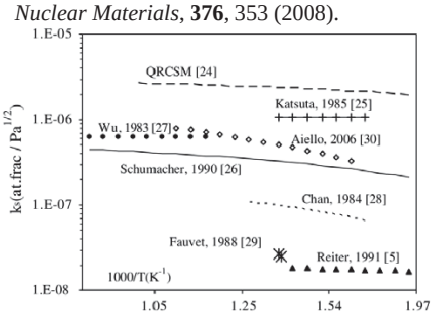
PbLi breeder concepts

- Structural material: reduced activation ferritic-martensitic (RAFM) steel
  - Limited to 550 °C operation, maybe lower (~480 °C) due to PbLi corrosion
- Helium-cooled lead-lithium (HCLL)
  - EU ITER TBM design and DEMO concept
  - PbLi breeder flows very slowly and serves no cooling function
    - High tritium partial pressure; permeation barriers required
  - Cooling is provided entirely by separate helium channels
- Dual-coolant lead-lithium (DCLL)
  - US TBM conceptual design (not pursued) and DEMO concept
  - Higher PbLi flow rates
    - Low tritium partial pressures *if extraction system is highly efficient*
  - ~50% of power extracted from PbLi, ~50% from separate helium coolant
  - SiC flow channel inserts for thermal (potential PbLi temp ~700 °C), and electrical (mitigate MHD forces) insulation, and corrosion barrier

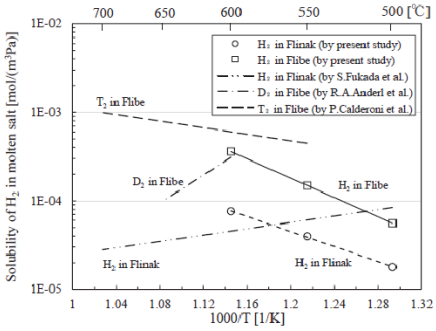
Tritium solubility in PbLi, FLiBe, and metals



PbLi: E. MAS DE LES VALLS et al., *Journal of Nuclear Materials*, **376**, 353 (2008).



Molten Salt: A. NAKAMURA al., *Journal of Plasma Fusion Research SERIES*, **11**, 25 (2015).



Tritium control in fusion

- Both PbLi and FLiBe have low tritium solubilities- this results in a higher tritium partial pressure and tends to drive permeation losses through solid structures
- Extraction concepts therefore attempt to do the following:
  - Provide a medium (purge gas, getter, etc.) where tritium will preferentially accumulate, relative to structural materials
  - Maximize the contact area of the breeder/coolant with this medium
  - Minimize the transport distance through the breeder/coolant to reach this medium
  - Maximize the residence time in the extraction system (i.e. reduce the flow rate)
- These same ideas for PbLi should be applicable to FLiBe or other molten salts (chemistry may complicate things somewhat)
- Additionally, one can apply permeation barriers (e.g. in the form of coatings) to structural materials- probably necessary for fusion, not for fission

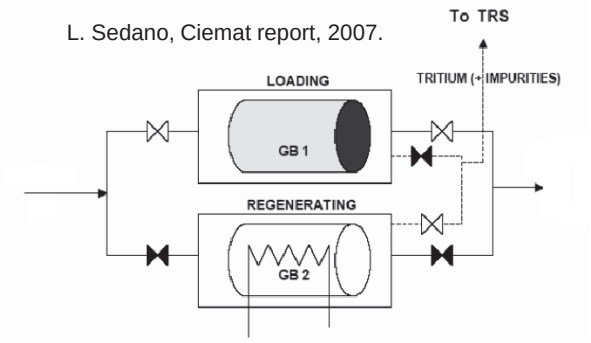
Tritium extraction concepts

- Immersed getters
- Liquid getter/cold trap
- Release to purge gas or vacuum
  - Bubblers, spray/droplets, extraction columns
- Vacuum permeator

Immersed Getters

- Getters such as U, Zr(Co), Pd, Ti, etc. are commonly used to remove tritium from gases (Example: a 60g U bed can hold 2g tritium)
- Immersed getters (V, Nb, Ta) have been proposed in the past to remove tritium from PbLi
- Principle demonstrated at small scale with V in PbLi, though not to saturation[1]

L. Sedano, Ciemat report, 2007.



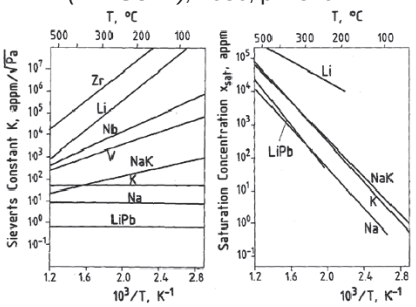
- Issues:
  - Integrity of material (getter beds for gases are reduced to fines and require ceramic filters)
  - Lifetime of beds under cyclic loading (e.g. daily)
  - Deleterious effects of impurities including oxygen

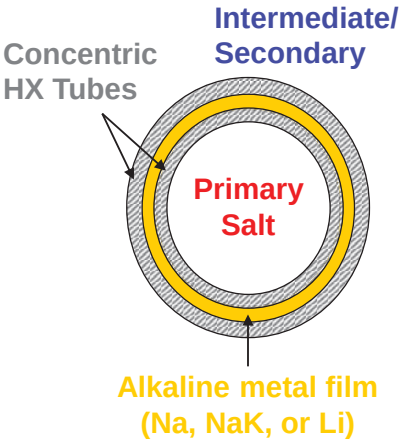
[1]H. Feuerstein, *Fusion Technology (14th SOFT),* 1986, p. 646

Liquid metal getter with cold trap

- Concept investigated at KIT in the 1980s
  - Intermediate NaK loop proposed, which acts as a tritium getter
  - NaK cooled so as to precipitate solid hydrides
  - Tritium removed from solid hydrides by vacuum pumping
- Might also take the form of a thin film between concentric HX tubes
  - Processing rate required for fusion may imply large heat loss
  - Salt-cooled fission reactors may require only infrequent batch processing and have minimal impact on HX performance
  - Li suitable for fission reactors where subsequent extraction is unnecessary
    - High saturated concentration
    - Less chemically reactive than Na/NaK

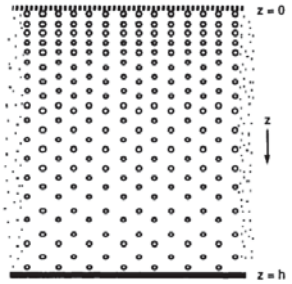
J. Reimann, *Fusion Technology (14th SOFT)*, 1986, p. 1579





Droplets in vacuum

- Concept: spray coolant as small droplets into a purge gas or vacuum
- Small droplets provide high surface area and small transport distance
- "Vacuum Disengager" proposed for HYLIFE-II IFE design study[1]
- Analytical solution and numerical models suggested 99.9% efficiency; no experiments performed
- Now under investigation for PbLi (as "Vacuum Sieve Tray")[2]
- Different analytical solution and numerical models suggest 70% efficiency achievable
- Measured extraction was lower than predicted by 10x, but models depend on (uncertain) solubility and diffusivity



[1]T. Dolan, *Fus. Tech.* **21** (1992) 1949-1954

[2]F. Okino, *FED* **87** (2012) 1014-1018

Compact Mass Extractor

- Gas/Liquid Contactor – planned for HCLL (TBM and DEMO)
- Structured packing disperses PbLi flow and creates a large interfacial area between PbLi and gas
- ≤ 30% efficiency for single column as tested in MELODIE loop[1]

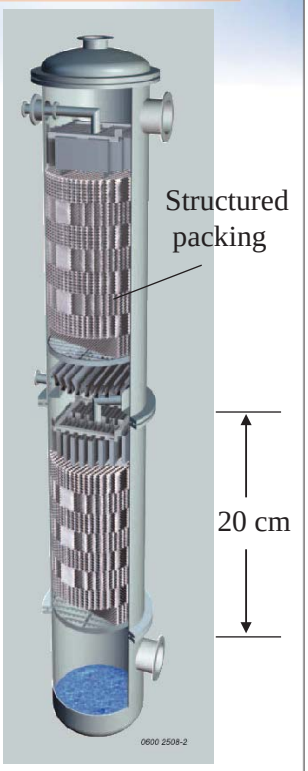
Extraction tests in a non-immersed hydraulic configuration ($T = 673$ K)

Test No.	L (l h^{-1})	G (Ncm^3 min^{-1})	$P_{H_2,IE}$ (Pa)	η (%)
1	70–90	100	1200–1350	20–22
2	30–50	100	1000–1100	29–31
3	30–50	500	975–1000	29–31
4	30–50	100	450–475	23–25
5	30–50	100	220–230	23–25

Extraction tests in an immersed hydraulic configuration ($T = 673$ K)

Test No.	L (l h^{-1})	G (Ncm^3 Min^{-1})	$P_{H_2,IE}$ (Pa)	η (%)
6	80–105	500	1300–1400	10–12
7	80–105	1000	1200	9–11
8	50–65	1000	1150–1200	>14

[1]N. Alpy et al. *FED* **49-50** (2000) 775-780



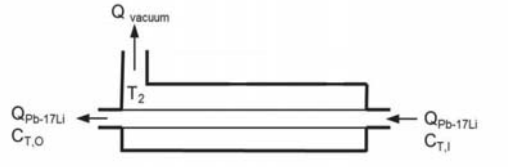
Sulzer Column

- HCLL DEMO (14 inventory re-circulations per day) requires at least 80% efficiency[2] even with permeation barriers (with ~100x reduction factor)
- Larger scale tests, optimization planned at TRIEX loop (ENEA) but no results as of yet

[2]O. Gastaldi et al. *FED* **83** (2008) 1340–1347

Vacuum permeator

- For a PbLi-cooled (e.g. DCLL) or salt-cooled fission or fusion reactor, higher flow rates must be processed
  - Simple scaling from most efficient MELODIE tests indicates that for a DCLL blanket (~470 inventory re-circulations/day), 240,000 extraction columns would be required[1] (!)
- DCLL flow rates are much higher (~470 inventory re-circulations/day required)
- Similar performance in a much smaller device is potentially achievable with a vacuum permeator
- Concept: a shell-and-tube mass exchanger with tritium-laden primary, vacuum secondary, and high-permeability, thin-walled tubes
- Required efficiency depends on the reactor design (losses), but:
  - What is achievable?
  - How does it scale?



[1]B. Merrill, 2005/06/15 ARIES meeting

Permeator extraction efficiency[1]

- Transport processes in a permeator tube:
  - Advection in PbLi in the axial direction
  - Convective mass transport in PbLi in the radial direction
  - Permeation (Diffusion) through solid in the radial direction
- Can be solved analytically: $\eta = 1 - \exp\left(-\frac{\tau\zeta}{1+\zeta}\right)$
- ζ and τ are dimensionless numbers that indicate the relative importance of three transport phenomena

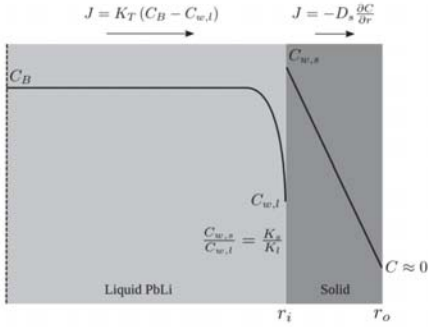
$$\zeta = \frac{\Phi_s}{K_T K_l r_i \ln(r_o/r_i)}$$

$$\tau = \frac{2K_T L}{v r_i}$$

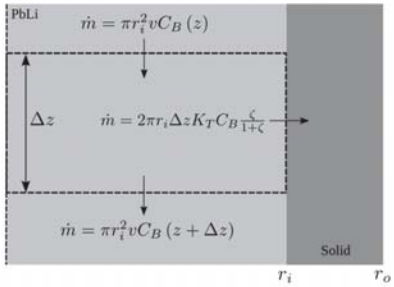
r_i – inner diameter of tubes
r_o – outer diameter of tubes
L – length of tubes
v – PbLi velocity
K_l – PbLi solubility
K_T – mass transport coefficient
Φ_s – permeability of solid tube

[1]P. Humrickhouse *Fusion Science and Technology* **68** (2015) 295-302

Radial Transport



Axial Transport



Significance of ζ and τ

- ζ indicates whether radial transport is limited by mass transport in PbLi, or by permeation through the solid tube wall
- When $\zeta << 1$: Diffusion in the solid is limiting; there is no dependence on the PbLi transport property K_T
- When $\zeta >> 1$: Mass transport in the PbLi is limiting; there is no dependence on the solid transport properties Φ_s, r_o or PbLi solubility K_l
- τ is a ratio of axial to radial transport times:

$$\tau = (L/v)/(r_i/2K_T)$$

- When $\tau >> 1$ tritium is swept through the length of the permeator tube before it has a chance to migrate radially
- Need to evaluate K_T…

Mass Transport Correlations

- K_T is defined by the Sherwood number: $\mathrm{Sh} = dK_T / D_l$
- Sherwood number correlations have the form $\mathrm{Sh} = \beta \mathrm{Re}^a \mathrm{Sc}^b$
- For PbLi at 470-700 ˚C, 10 < Sc < 150 $\left(\mathrm{Sc} = \mu / \rho D_l\right)$
- The correlations below are remarkably consistent with each other, and with the heat transfer analogy- this approach is valid
- The choice of correlation is not a significant source of uncertainty in this analysis relative to other parameters

β	a	b	Range	Reference	Notes
0.023	4/5	1/3		Colburn 1933	Heat transfer analogy
0.023	0.83	0.44	2000 < Re < 35000 0.6 < Sc < 2.5	Gilliland and Sherwood 1934	Vaporization of nine different liquids in air
0.0328	0.77	0.33	3000 < Re < 40000 0.5 < Sc < 3	Johnstone and Pigford 1942	Distillation of five different substances in a wetted-wall column
0.023	0.83	1/3	2000 < Re < 70000 1000 < Sc < 2260	Linton and Sherwood 1950	Solution of benzoic acid, cinnamic acid, and beta-naphthol in water
0.0163	0.83	0.44	Sc ~ 0.6	Kafesjian et al. 1961	Vaporization of water in a wetted-wall tower
0.0096	0.913	0.346	10000 < Re < 100000 430 < Sc < 100000	Harriott and Hamilton 1965	Benzoic acid in glycerin-water, and hydroxymethycellulose solutions

Permeator Optimization

- Regardless of the transport regime, permeator efficiency is always increased by:
  - Increasing the temperature, T
  - Increasing the tube length, L
  - Decreasing the permeator velocity, v (e.g. by increasing the number of permeator tubes)
  - Decreasing the tube diameter, d
- Using the analytical solution, we can optimize the design (minimize the total volume)
- The following slide does so for different materials and temperatures, subject to the following constraints (from ARIES-CS):

$$\eta \geq 0.7 \qquad \mathrm{d} \geq 0.01\mathrm{m} \qquad \dot{m} = \rho N \pi r_i^2 v = 26000\,\mathrm{kg/s} \qquad \Delta p = f \frac{L}{d} \frac{\rho v^2}{2} \leq 1\ \mathrm{MPa}$$

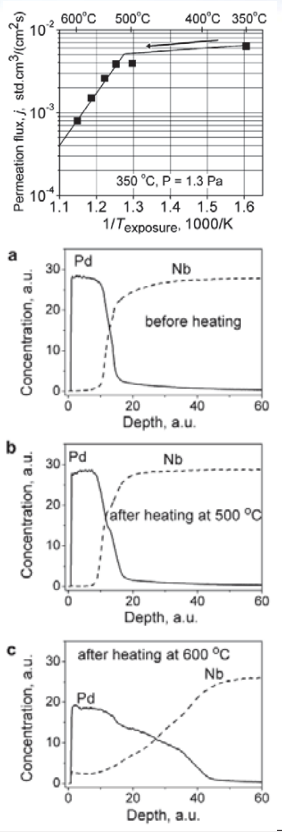
Tube material comparison

	B&W PWR steam generator	RAFM 470 ˚C	RAFM 470 ˚C	RAFM 470 ˚C	Vanadium 400 ˚C	Vanadium 500 ˚C	Vanadium 600 ˚C	Vanadium 700 ˚C
η (low solubility)		0.7	0.7	0.7	0.7	0.7	0.7	0.7
Tubes (#)	15,000	343,521	68,704	19,432	13,347	10,136	8,274	7,095
Tube length (m)	20.7	8.54	16.61	37.3	18.25	11.15	7.65	5.7
v (m/s)		0.1	0.5	1.77	2.55	3.4	4.22	4.98
Total volume (m^3)	61.8	278.7	108.42	69.0	23.15	10.74	6.01	3.84
ζ		4.85	1.27	0.45	1681	425	148	65
η (high solubility)		0.10	0.04	0.03	0.47	0.36	0.29	0.23

- There is a significant size/cost advantage to high-permeability materials

Group 5 Metals - oxidation

- Group 5 metals have very high tritium permeabilities and from that standpoint are promising tube materials
- They are compatible with PbLi, but the oxygen partial pressure on the vacuum side must be kept below 10^{-10} Pa to prevent oxidation[1]
- Application of a Pd coating can prevent this, and commercial hydrogen purifiers based on this concept are available
- They have a very narrow range of operation around 400 ˚C
  - At lower temperatures, hydrides form and embrittle the structure
  - At higher temperatures, Pd and substrate diffuse together, reducing (irreversibly) the tritium permeability



[1]R. Kurtz, 2005 ITER TBM meeting

V. Alimov, *International Journal of Hydrogen Energy* **36** (2011) 7737-7746

Potential solutions

- Inter-diffusion of Pd and substrate can be prevented by an intermediate layer that separates them
  - Such composites are being actively investigated in the hydrogen energy research community
  - These are typically ceramics with some porosity so as not to prevent tritium permeation
  - Al_2O_3, Nb_2C, HfN, YSZ, etc. mentioned in literature
- Alloys?
  - V-Ni, Pd-Cu, V-Ti, V-Cu, others mentioned in literature
- Other coatings- Pd is necessary for separation from other gases, but we only need to prevent oxidation

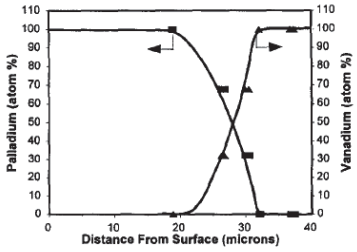


Fig. 6. Cross-sectional analysis of a palladium-coated vanadium membrane after 22 h under argon showing some diffusion of vanadium into the palladium layer. Test conditions: 700°C, membrane stored under 100-psig argon.

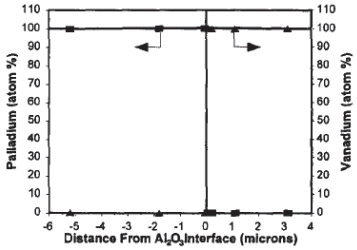
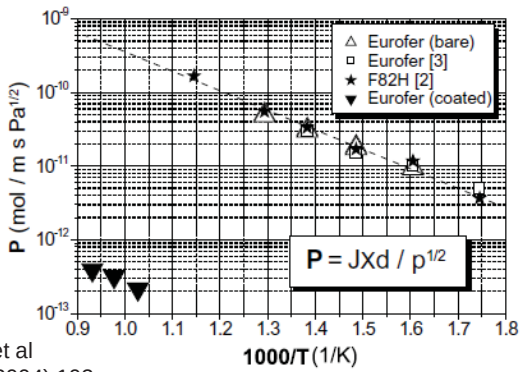


Fig. 7. Cross-sectional analysis of the palladium and vanadium layers of a three-component composite-metal membrane incorporating a porous aluminum oxide intermediate layer operated for 76 h. Test conditions: 700°C, 100-psig hydrogen feed pressure, permeate hydrogen at ambient pressure.

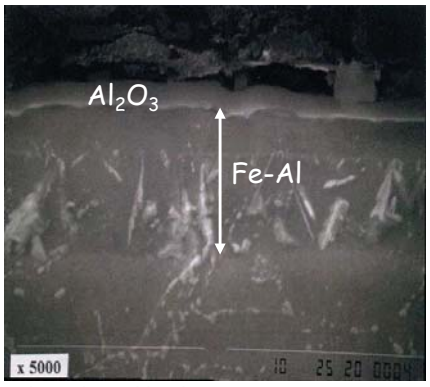
Edlund, *Journal of Membrane Science* **107** (1995) 147-153

Permeation Barriers

- Even under relatively optimistic assumptions for the extraction system, fusion systems studies usually find a permeation reduction factor (PRF) of 10-1000 on structures is necessary to meet release limits
- Many barriers have been investigated experimentally, such as low-permeability metals (e.g. aluminum) or ceramics such as Al_2O_3, Cr_2O_3, Er_2O_3
- These have achieved permeation reduction factors as high as 10,000 in the laboratory



Levchuck et al
JNM **328** (2004) 103



Al_2O_3 deposition by CVD - Jürgen Konys

Permeation Barriers in a Radiation Environment

- While permeation reduction factors up to 10,000 have been measured in the laboratory, reactor tests on the same materials have not achieved this

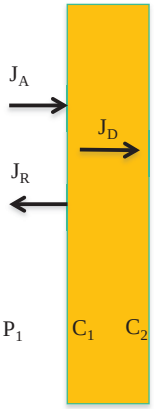
Irradiation testing of tritium/hydrogen barriers — Hollenberg et al *FED* **28** (1995) 190-208

Test	Tritium source	Tritium sink	Reactor	Barrier system[a]	Temperature (°C)	Effective PRF	Reference
LIBRETTO-2	Pb–17Li	$He + H_2$	HFR	Alum/316L	275–440	<80	[41, 42]
LIBRETTO-3	Pb-17Li	$He + H_2$	HFR	316L/TiC	280–450	3	[43]
				Al_2O_3/316L	280–450	3	
				316L/alum/Al_2O_3	280–450	15	
TREXMAN	Pb–17Li	$He + H_2$	YAYOI	Cr_2O_3/SS316	600	10	[14]
				SS316/Cr_2O_3	600	100	
Loop-1	$LiAlO_2$	H_2O	ATR	Alum/SS316/alum	318	150	[44]
WC-1	$LiAlO_2$	H_2O	ATR	Alum/SS316/alum	<330	150	[45]

- This reduction may result from damage (e.g. cracking) of the barrier, an increase in defects, or some other effect under irradiation

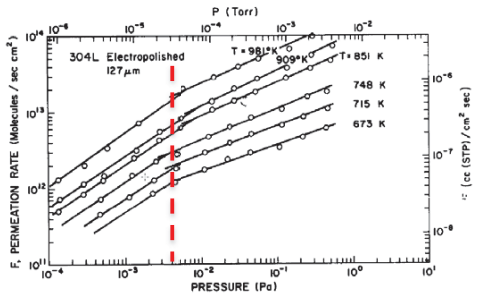
Permeation scaling with pressure

- Permeation usually scales with the square root of partial pressure, implying that it is limited by diffusion through the solid
- Low pressures or surface changes can result in surface-limiting and a change to linear dependence on partial pressure
- Systems designed to achieve low tritium partial pressures need to investigate this experimentally
  - Dimensionless number governs transition:

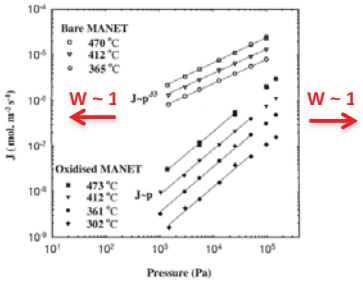


$$W = \frac{K_A x \sqrt{P}}{K_S D}$$

Ali-Kahn et al *JNM* **76/77** (1978) 337-343.



Perkins and Noda *JNM* **71** (1978) 349-364.



Serra and Perujo *JNM* **240** (1997) 215-220.

Conclusions

- A number of tritium capture/extraction concepts have been proposed for fusion over the last several decades
- Some of these have been investigated experimentally, but none on the scale (size, tritium inventory) or under the conditions (radiation, high temperatures, long times) necessary for fusion
- Because of the low solubility of tritium in both PbLi and molten salts (including FLiBe), extraction techniques developed for PbLi are likely applicable to salt-cooled fusion and fission reactors as well
- Tritium generation will be orders of magnitude lower in fission reactors, for which tritium is a much more manageable problem
  - Tritium must be captured, but not necessarily separated/purified
  - Extraction systems may only need to process a fraction of the coolant on each pass
  - Permeation barriers may be unnecessary; for fusion it will be difficult to keep losses sufficiently low without them

Tritium- Control Technologies for TMSR in CAS

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Center for TMSR, CAS

2015-10-28

钍基熔盐核能系统

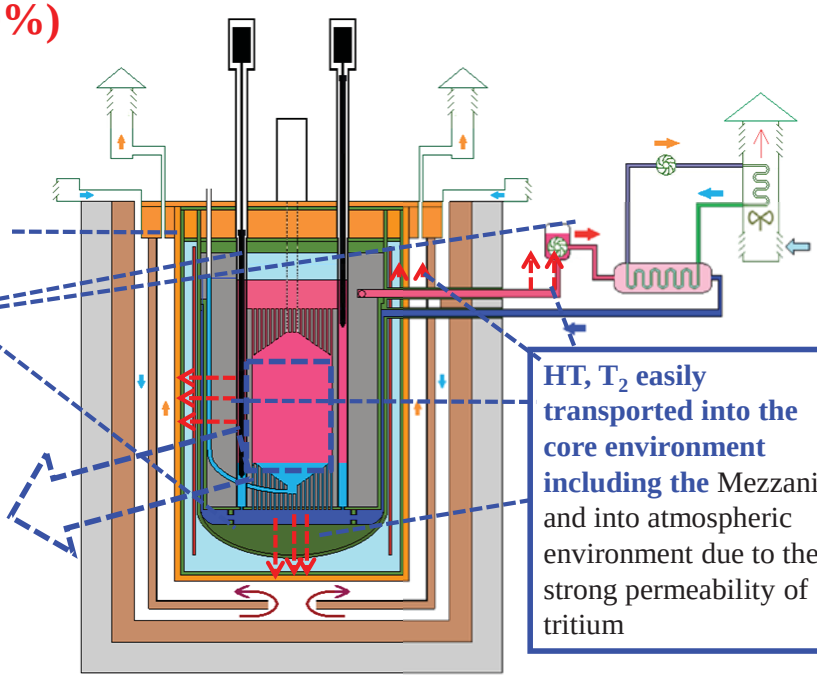
中国科学院上海应用物理研究所

Shanghai Institute of Applied Physics, Chinese Academy of Sciences

Outline

I. Background

1. Tritium production in TMSR (FLiBe as primary coolant, ^{7}Li abundance: 99.99%)



Mezzanine of reactor core

Cover gas（Ar/He）

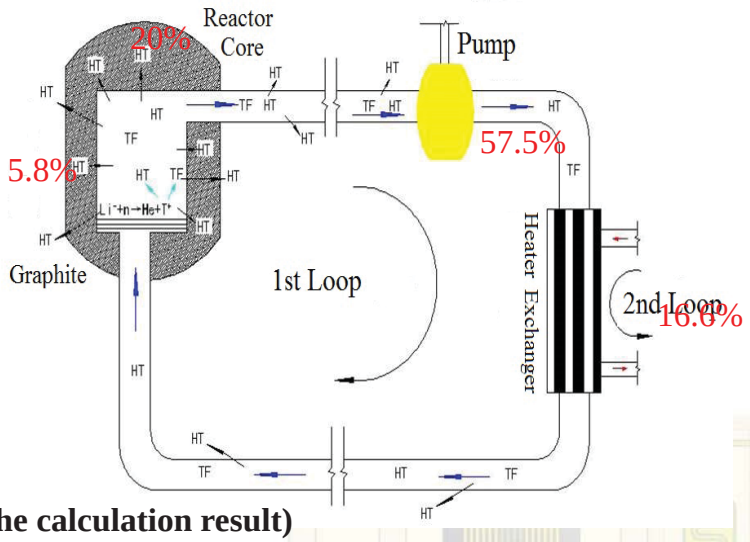
Main Gaseous radioactive Nuclides
① T (HT, T_2, TF)
② Inert gas (Kr, Xe

HT, T_2 easily transported into the core environment including the Mezzanine, and into atmospheric environment due to the strong permeability of tritium

2. Distribution of Tritium in TMSR

Tritium distribution in TMSR-SF1

Position	%	Bq/a*
Core Graphite	20 (Cited from MSRE)	5.5E14
Mezzanine	5.8	1.6E14
1st Loop	57.5	1.6E15
2nd Loop	16.6(Cited from MSRE)	4.6E14
Total	100	2.8E15



* $10MW_{th}$, ^{7}Li99.99%, 300-days run (**the calculation result**)

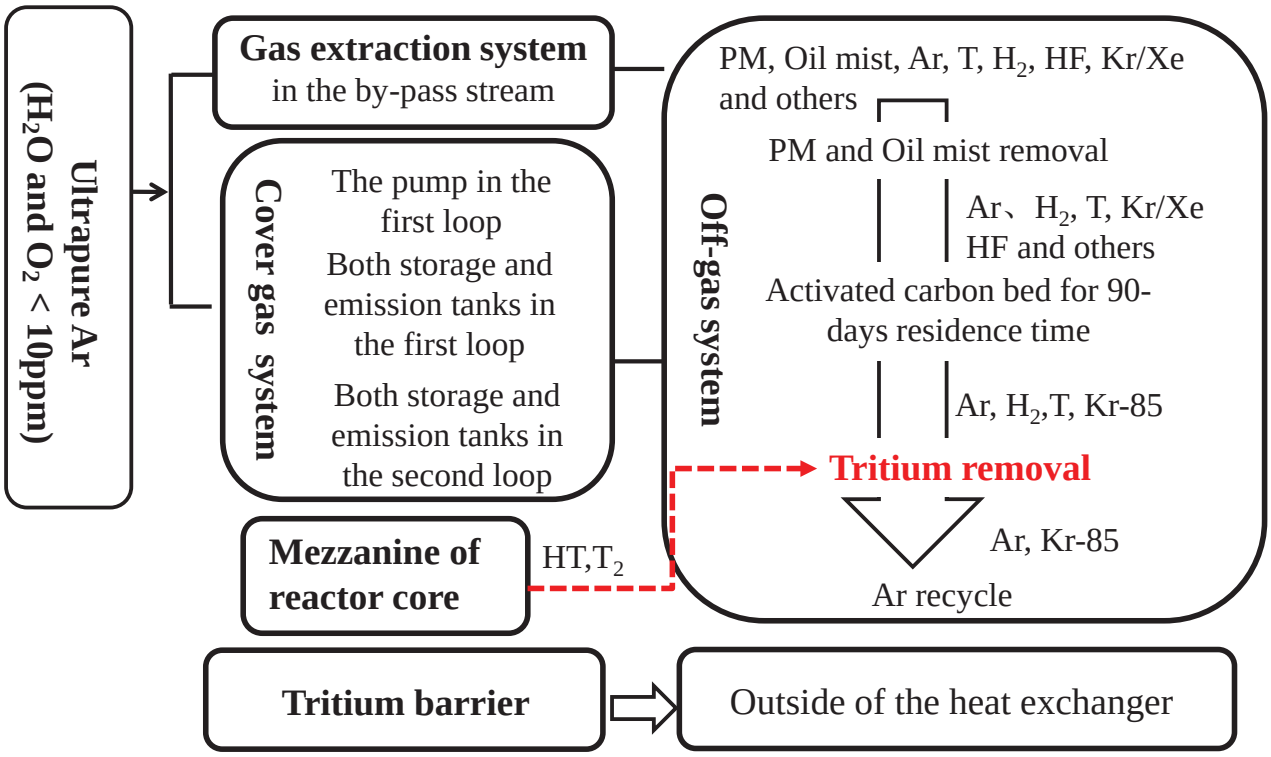
Emission limit of tritium in 3000MW nuclear power station in China:1.5E13Bq/a (LWR), 4.5E14 Bq/a (HLR)

Does tritium transport into the cover gas completely? What is the chemical form of tritium?

Tritium removal: necessary and difficult

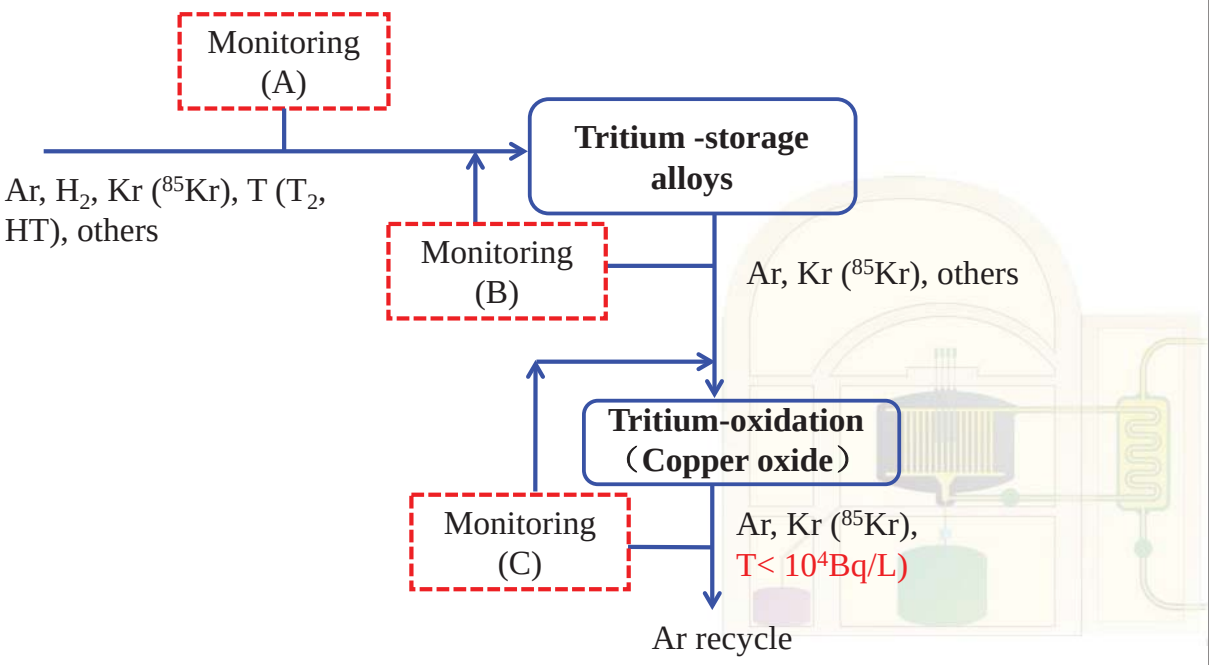
3. Considerations of Tritium control for TMSR

Remove tritium in the first loop as much as possible
Prevent tritium permeation into the second loop as much as possible



Tritium removal

Combination of **tritium -storage alloys** and **tritium-oxidation** with the **on-line tritium monitoring**



Outline

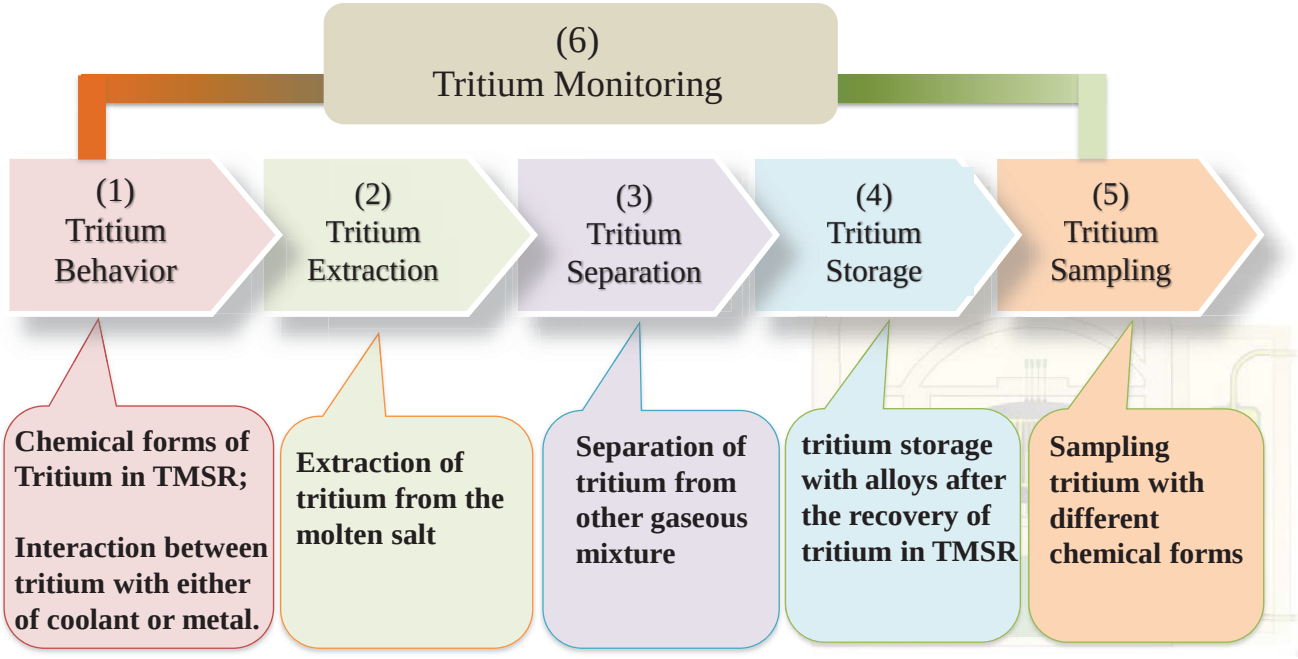
I. **Background**

II. **Roadmap** of Tritium-control technologies for TMSR in CAS

III. **Performances** at the tritium-control technologies for TMSR in CAS

IV. **Summary**

II. The Roadmap of tritium-control technologies for TMSR in CAS



Outline

I. **Background**

II. **Roadmap** of Tritium-control technologies for TMSR in CAS

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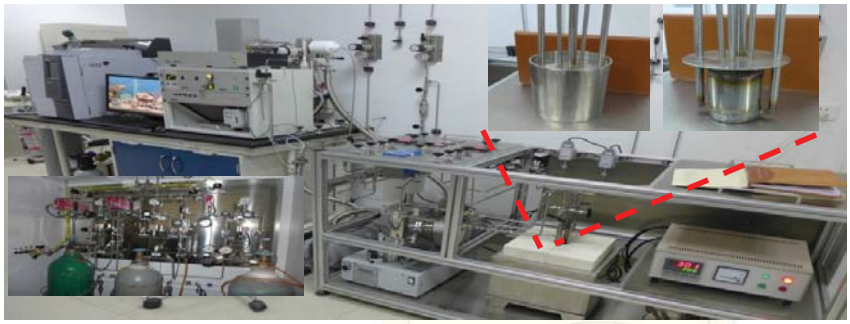
IV.Summary

1. Tritium behavior

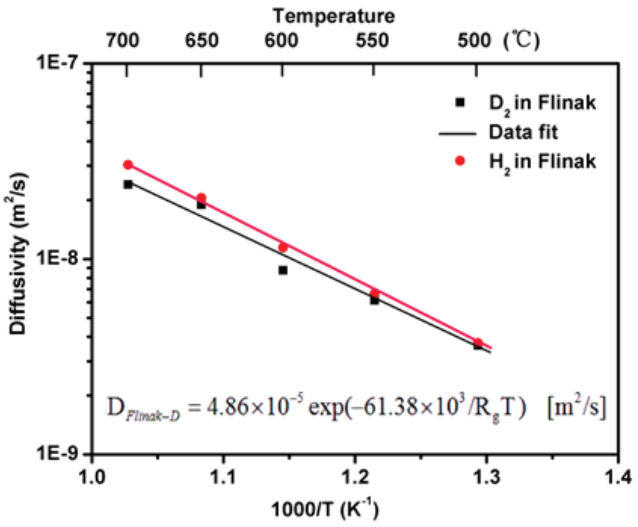
Interaction of hydrogen isotopes with either of high-temperature molten salt or structural materials

- Test apparatus for diffusion of hydrogen isotopes **in high-temperature molten salt**

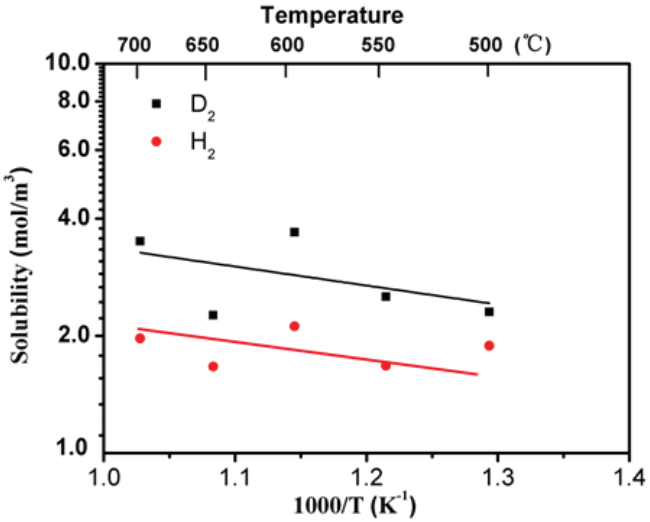


- Test apparatus for permeation of hydrogen isotopes **in structural materials**

➢ Diffusion of Hydrogen isotopes (H_2, D_2) in high-temperature FLiNaK

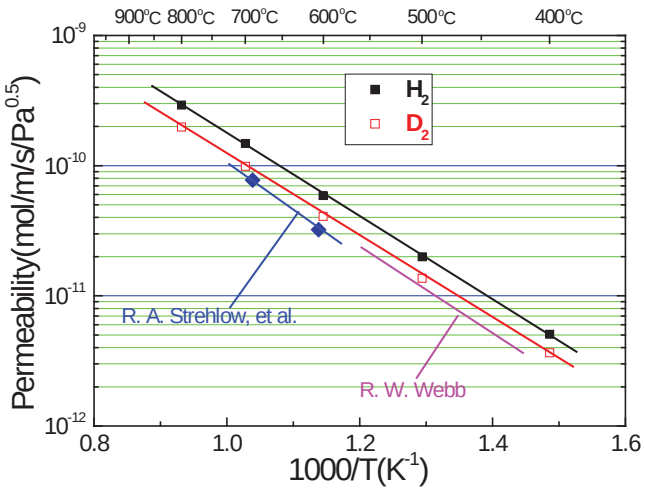


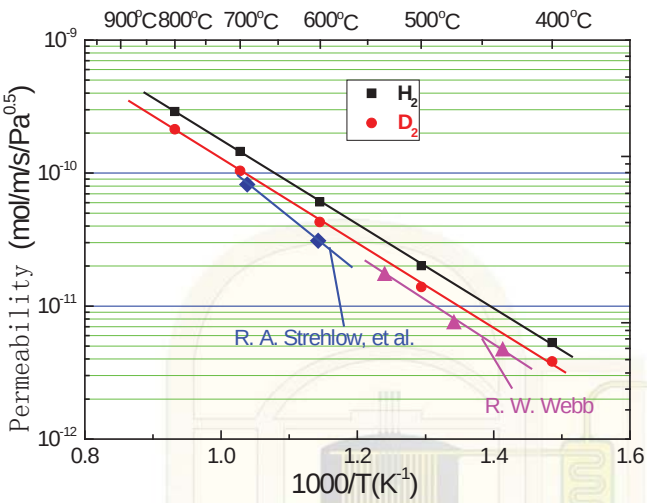
Little difference of diffusion coefficient between Deuterium and hydrogen in FLiNaK



Much more solubility of deuterium than hydrogen in FLiNaK at 500-700˚C

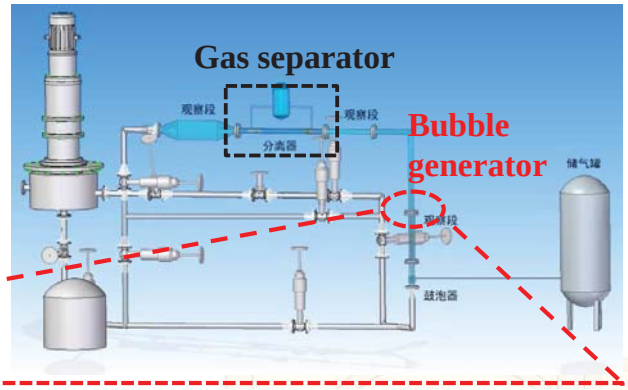
➢ Permeability of hydrogen isotopes in Hastelloy N (left) and GH 3535 (right) alloy

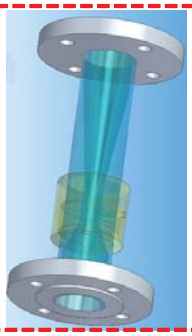


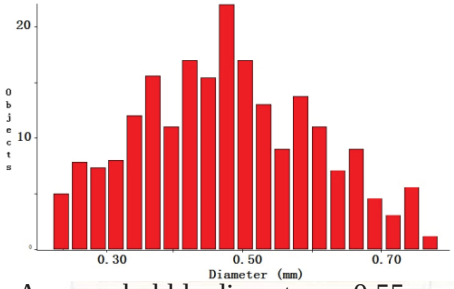


2. Tritium Extraction

Set up gas extraction system in water test loop and the separation efficiency was qualitatively analyzed







Average bubble diameter： 0.55mm

Bubble generator

1 hole

2 holes

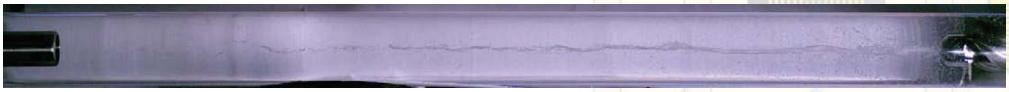
4 holes

Little effect of number of holes on the bubble size distribution at the flow rate greater than 15 m^3/h,

Gas separator

Negative Press 0.009MPa

Balance 0.012MPa



Positive Press 0.030MPa

Separation efficiency of separator relating with the outlet pressure

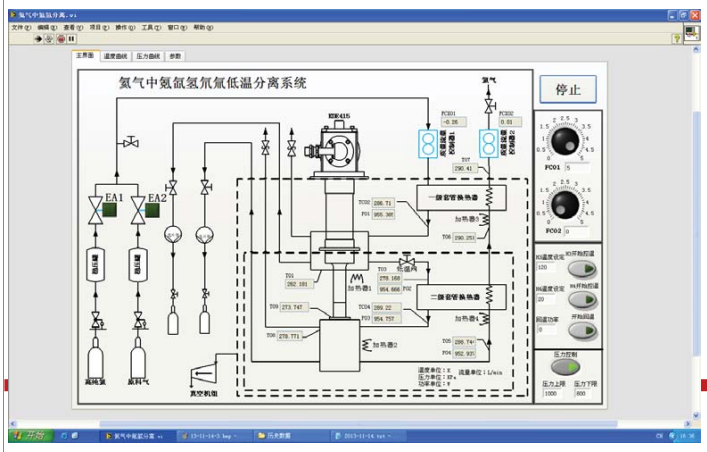
3. Tritium Separation

Successful separating xenon, krypton and hydrogen from helium environment using cryogenic technology for tritium separation

Kr、Xe <1PPb and H_2<0.1PPM after two-step frozen

Labview interface

(In-site monitoring 9-way temperature and 4-way pressure pressures as well as data acquisition)



examination of gaseous leak

Detection of temperature and pressure

4. Tritium Storage

Complete research platform for tritium storage using alloy

Testing system for selecting tritium-storage material

(Low pressure)

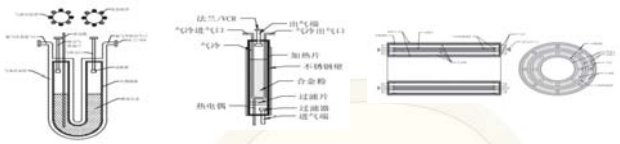
Pressure range：0.1Pa-50KPa

(High Pressure)

Pressure range：20KPa-10MPa

Testing system of tritium-reservoir performance

Tritium reservoir



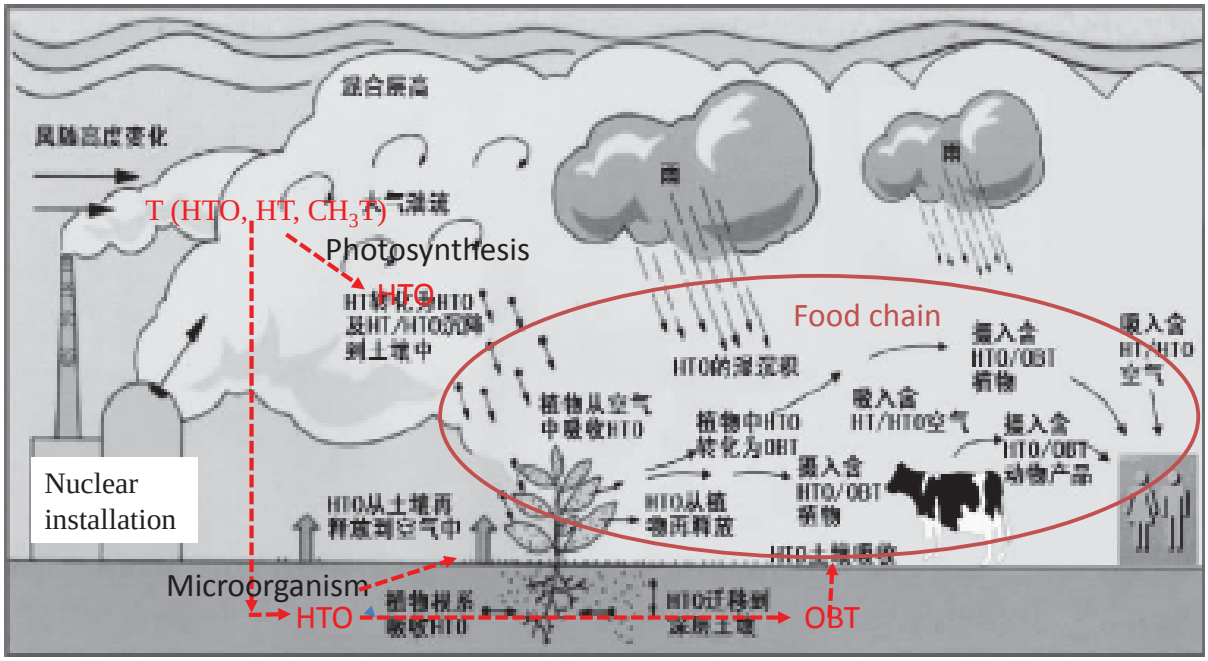
tritium-reservoir performance **Control**

Gas distribution

Detection

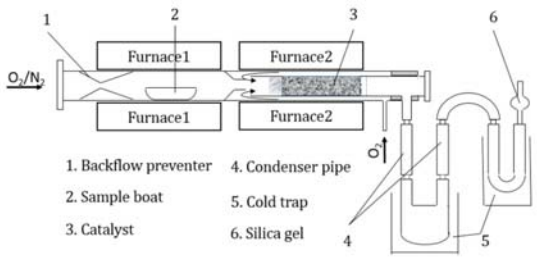
5. Tritium Sampling

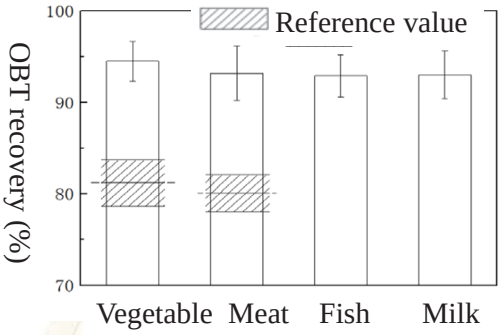
Tritium transport in environment

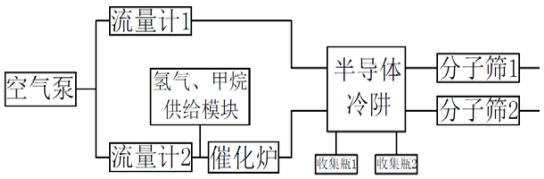


Sampling tritium (HTO, HT, CH3T, OBT): Necessary

Successful development of OBT-oxidation collection and tritium sampler







Sampling HTO, HT and CH_3T simultaneously;
Collecting efficiency >95%

6. Tritium Monitoring

➢ **Off-line Measurement**

Tritium measurement platform (Detected limit of HTO< 0.5Bq/L)

Sampling ⇒ **Pretreatment** ⇒ **Measurement**

OBT collection

Tritium sampler

Distillation

Electrolytic enrichment

LSC

➢ **Online monitoring**

On line monitoring of HTO, HT and Kr, Xe, simultaneously ;

HT,HTO： 1-10^7μCi/m^3

Kr and Xe: 1-10^6μCi/m^3

Other studies

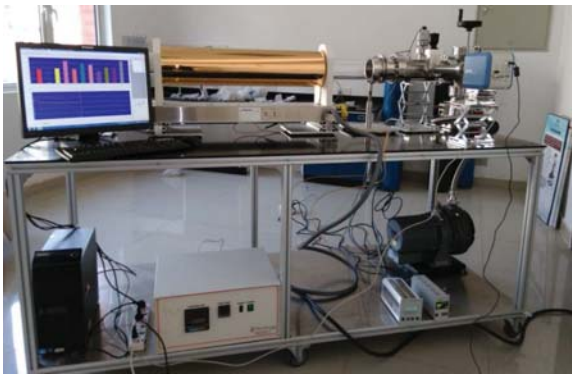
Tritium barrier

Selection of tritium barrier

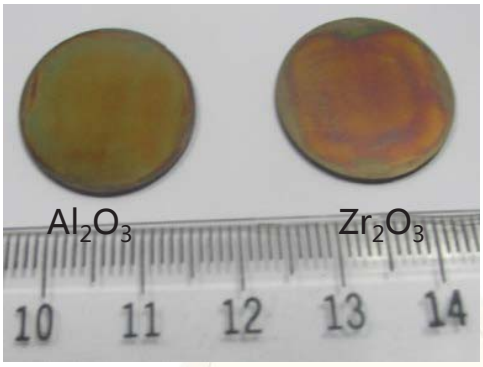
Barrier	Base Metal	PRF*
Al_2O_3	SS316, MANET, TZM, Ni, Hastalloy-X	10 to >10,000
TiC, TiN, TiO_2	SS316, MANET, TZM, Ti	3 to >10,000
Cr_2O_3	SS316	10 to 100
Si	Steels	10
BN	304SS	100
N	Fe	10 to 20
Er_2O_3	Steels	40 to 700

*permeation reduction factor

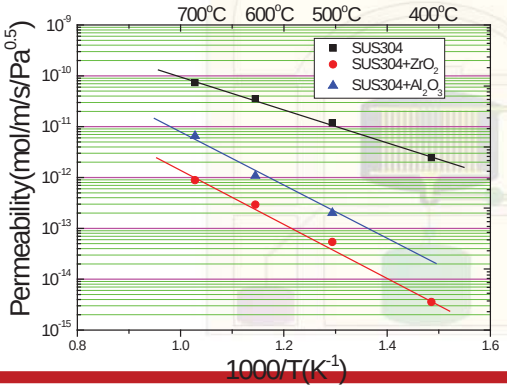
Experimental results of tritium barrier



Fabrication for tritium barrier by pack cementation



Measurement of FRF of tritium barrier



FRF
Al_2O_3: 200
ZrO_3:30

Outline

I. **Background**

II. **Roadmap** of Tritium-control technologies for TMSR in CAS

III. **Performances** at the tritium-control technologies for TMSR in CAS

IV.Summary

IV.Summary

1. Several test platforms for tritium control have been set up in CAS
2. Sampling and monitoring technologies have been mastered in CAS
3. Several experiments are conducting in CAS, those consist of interaction of hydrogen isotope with graphite, gas extraction in high-temperature molten salt loop
4. The chemical form of tritium in both irradiation high-temperature molten salt and graphite will be determined along with the cooperation between SINAP and MIT

Research on Techniques for Tritium Sequestration and Removal at UNM

Floren Rubio
Cole Mueller and Edward D. Blandford
University of New Mexico

Leonard J. Bond
Iowa State University
Center for Nondestructive Evaluation

Tritium Workshop
Salt Lake City, UT
October 28th, 2015

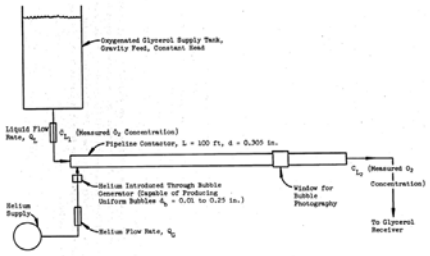
University of New Mexico – Department of Nuclear Engineering

Objectives

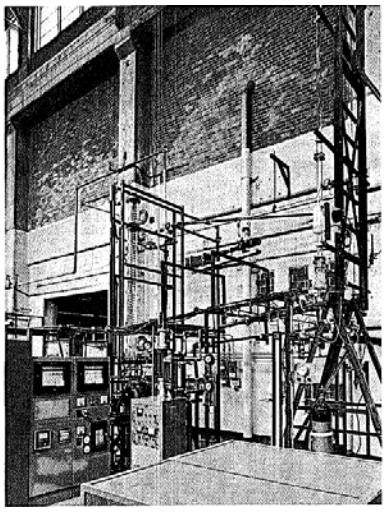
- Previous and concurrent work on inert gas sparging
- Introduction to ultrasonics
  - Theory
  - Current industrial uses
- Inert gas sparging enhancement for salt fueled and salt cooled reactors

Sparging History in the MSR

- In the past, ORNL had investigated the use of inert gas sparging for the MSR project.



Peebles, ORNL-TM-2245 1968



Kress, 1972

Focus of Sonomechanically Enhanced Sparging Research at UNM

- Use ultrasonic effects to increase the efficiency of inert gas sparging
  - Rectified diffusion to increase mass transfer of dissolved gasses into sparging bubbles.
  - Cavitation to reduce size of injected bubbles as well as take advantage of the degassing behavior.
  - Use water and dissolved oxygen as surrogates for molten salts and dissolved tritium.

Ultrasonic Theory

- Ultrasonic basics for process streams.
  - Sonomechanical effects
  - Sonochemical effects
- Industrial examples (non-exhaustive)
  - Metal melt degassing
  - Biological cell disruptors
  - Chemical catalysts

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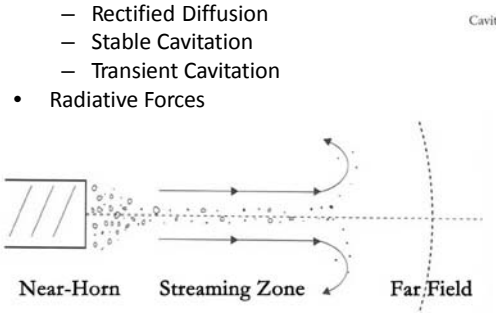
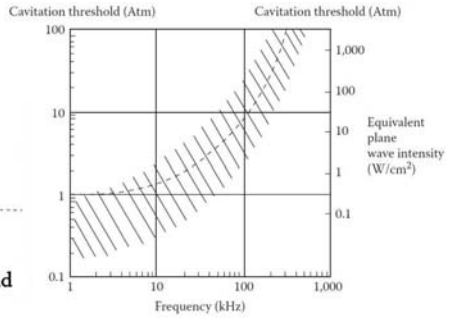
Sonomechanical Effects

- Rayleigh Plesset Equation

$$R\ddot{R} + \frac{3\dot{R}^2}{2} = \frac{1}{\rho}\left[\left(p_0 + \frac{2\sigma}{R_0} - p_v\right)\left(\frac{R_0}{R}^{3\kappa}\right) + p_v - \frac{2\sigma}{R} - \frac{2\eta\dot{R}}{R} - p_0 - p_a(t)\right]$$

  - Determines the cavitation thresholds and bubble dynamics.
  - Determines resonance frequency and size.
- Distinct regions of behavior
  - Rectified Diffusion
  - Stable Cavitation
  - Transient Cavitation
- Radiative Forces





Sonochemical Effects

- Uses high intensity ultrasonics to increase chemical activity by adding energy to the chemical system by cavitation.
  - Cavitation applies localized high temperatures (~5000 K) and pressures (~10MPa).
- General categories of processes:
  - Homogenous sonochemistry of liquids
  - Heterogenous sonochemistry of liquid-liquid systems
  - Heterogenous sonochemistry of liquid-solid systems
  - Sonocatalysis

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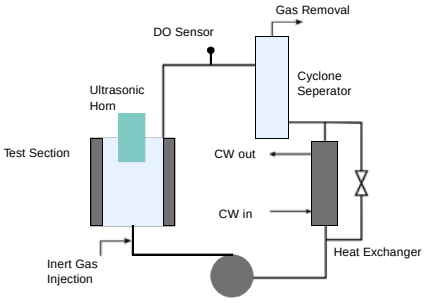
Initial Rectified Diffusion Studies

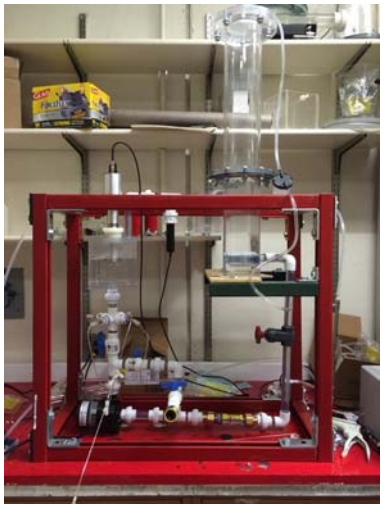
Experimental setup to capture the behavior of sparging bubbles in an ultrasonics field.
The goal is to oscillate sparging bubbles in the rectified diffusion regime.

Visualizations of sparging bubbles flowing into an ultrasonic field. Bubbles were observed oscillating at around the predicted diameter.

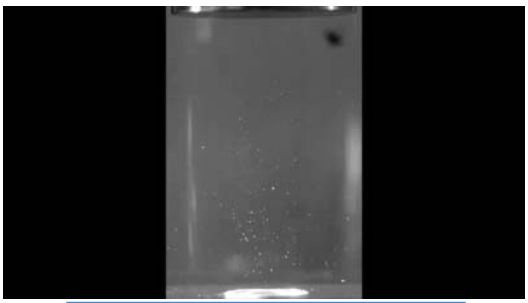
Experimental Conditions	
Sonic Field	20 kHz, Pulsing
Intensity	~0.085 W/cm^2

Small Scale Test Loop





Small Scale Loop Test Section



Experimental Conditions	
Sonic Field	20 kHz, Pulsing
Intensity	~0.085 W/cm^2
Flow Rate	1.25 GPM
Initial DO	8.0 mg/L
Final DO	3.0 mg/L

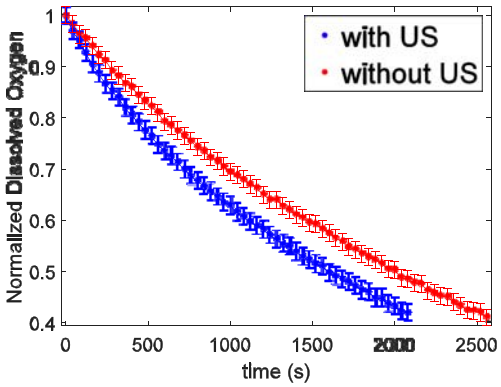
Preliminary Experimental Results for Small Scale Test Loop

Experimental Conditions	
Sonic Field	20 kHz, Pulsing
Intensity	~0.085 W/cm²
Flow Rate	1.25 GPM
Initial DO	8.0 mg/L
Final DO	3.0 mg/L

Results:

20% reduction in Degassing time.

Bubbles tend to avoid near field and oscillate in anti nodes and coalesce into bubbles of the resonant size for 20 kHz



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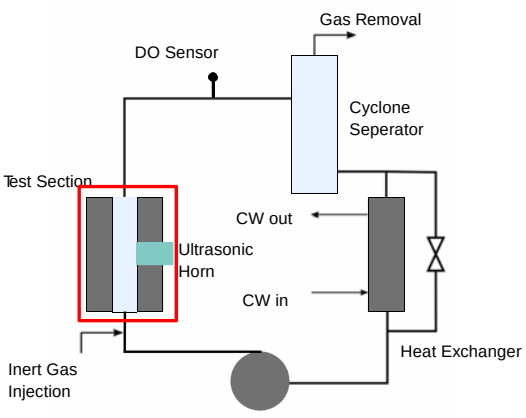
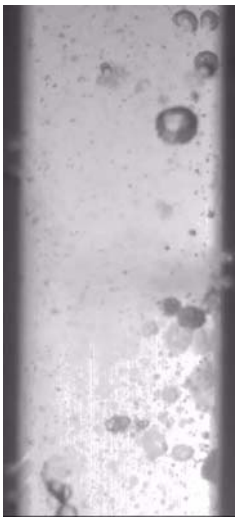
Acoustic Reflector and Cavitation Studies



Preliminary Cavitation Results

Narrow Channel Test Section

Experimental Conditions	
Sonic Field	20 kHz
Intensity	275 W/cm² continuous
Flow Rate	1.25 GPM
Initial DO	8.0 mg/L
Final DO	3.0 mg/L



Bubbles no longer avoid near field and can start breaking up into smaller bubbles.

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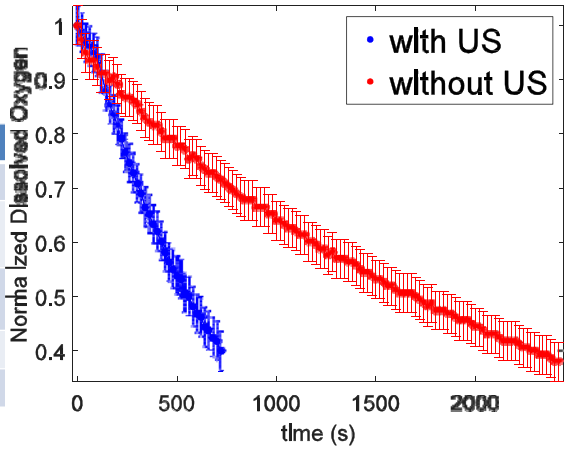
DO Results with Reflector and Cavitation

Result:

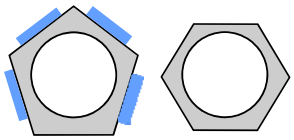
~70% reduction in time for the same DO reduction.

Experimental Conditions	
Sonic Field	20 kHz
Intensity	275 W/cm² continuous
Flow Rate	1.25 GPM
Initial DO	8.0 mg/L
Final DO	3.0 mg/L



Next Steps

- Different cell geometries
  - Ring Transducers
  - Indirect Sonication
- Scale up considerations



Challenges

- Materials challenges
  - Erosion
  - Salt Compatibility
  - Activiation
- High Temperatures
- High Radiation Doses

Conclusion

- Discussed the ultrasonic theory
- Ultrasonics has the potential of enhancing a known method of tritium removal for fluoride salt applications.
- Discussed some next steps and future challenges of ultrasonics with molten salts

University of New Mexico – Department of Nuclear Engineering

Questions?

Tritium Transport and Corrosion Modeling in the Fluoride Salt-Cooled High-Temperature Reactor

John D. Stempien, PhD

Content Based on Doctoral Thesis Defense

Workshop on Tritium Control
Salt Lake City, Utah

October 28, 2015

Presentation Outline

I. Tritium poses two problems

II. These problems are coupled

III. The TRIDENT model captures these dependencies

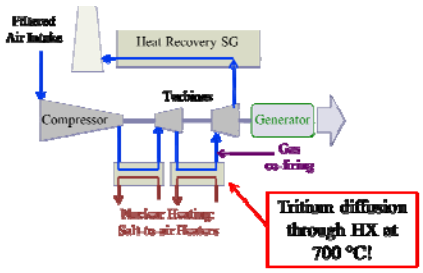
IV. Major elements of the TRIDENT model

V. Results of FHR simulations and comparison of tritium mitigation methods

Tritium Poses Two Problems

1. Corrosion - preferential attack of Cr in alloys by TF:
   - $2TF_{(d)} + Cr_{(s)} \rightarrow CrF_{2(d)} + T_{2(g)}$
   - Corrosion reaction consumes TF, generates T_2

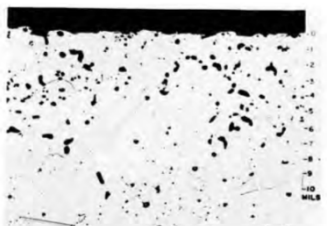
2. Radiological:
   - T_2 fast diffusion through metal
   - $T_{1/2}$ = 12.3 yr
   - β = 5.9 keV



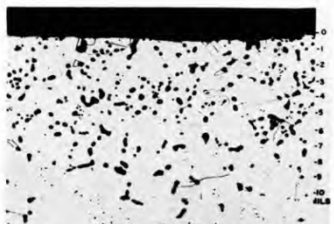
- Must control corrosion and manage tritium escape from system
- Modeling/simulation to help evaluate tritium control options

Tritium Behavior Couples to Coolant Chemistry and Corrosion Rates

- Knowing corrosion rates is important for long-term operation:
  - Rates are sensitive to coolant impurities
  - Corrosion couples to tritium behavior!



Fluoride Salt MODERATE in Fe and Ni



Fluoride Salt HIGH in HF

Images of Inconel from ORNL-2349

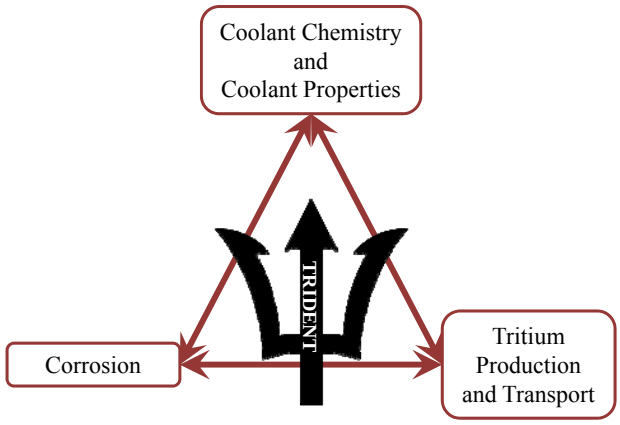
FHR Produces More Tritium than Other Fission Reactors

- Tritium behavior must be characterized to build licensing case:
  - Need to know release rates
  - Need to know distribution throughout reactor

	Tritium Production Rates [Ci/GWd]
BWR*	12.3
PWR*	13.9
HTGR*	18.5
FBR*	24.9
HWR*	1176
FHR	**11,000 (peak production rate calculated here)**

*Values calculated from data in: "Management of waste containing tritium and carbon-14", International Atomic Energy Agency, Technical Reports Series No. 421, Vienna, 2004.

TRIDENT (TRItium Diffusion EvolutioN and Transport) Was Developed to Link FHR Tritium Behavior to Coolant Chemistry



Basic Elements of TRIDENT: Tritium Generation in Flibe Coolant

Neutron Transmutation Generates Tritium in Flibe

$$^{6}LiF + n \rightarrow He + TF$$

$$^{7}LiF + n \rightarrow He + TF + n'$$

$$BeF_2 + n \rightarrow {}^{4}_{2}He + {}^{6}_{2}He + 2F^{-}$$

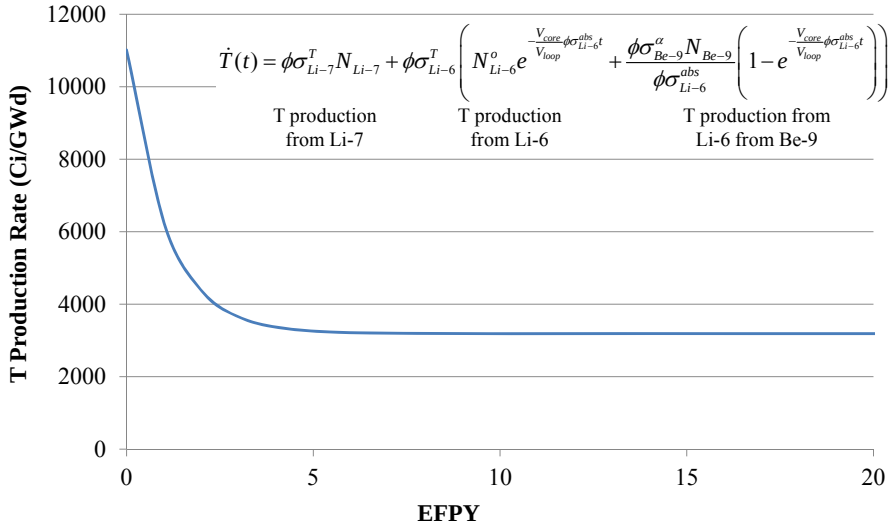
$${}^{6}_{2}He \rightarrow {}^{6}_{3}Li + e^{+} + \bar{\nu}_e \quad (t_{1/2} = 0.8 \text{ sec})$$

^{6}Li = 0.005 wt%

^{7}Li = 99.995 wt%

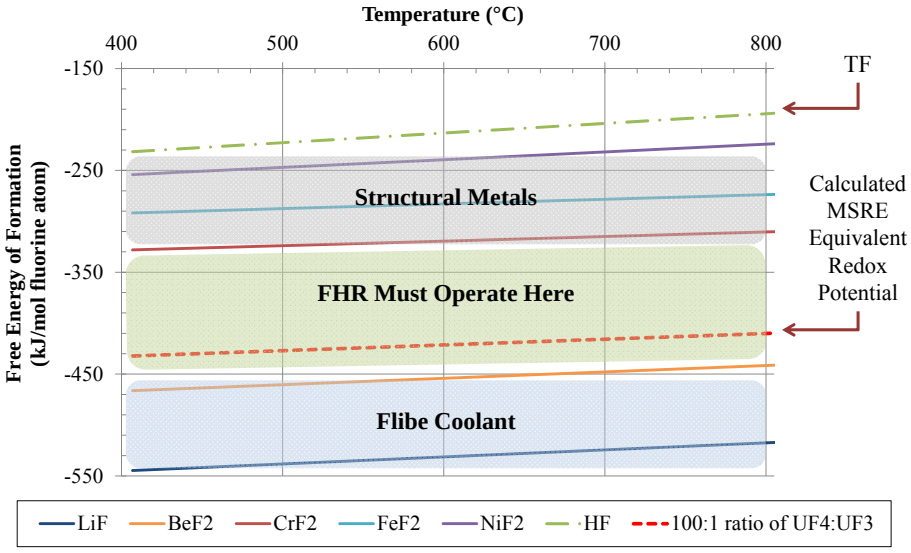
	One-group Cross section (b)
$\sigma^{T}_{Li\text{-}6}$	148
$\sigma^{\alpha}_{Be\text{-}9}$	3.63×10^{-3}
$\sigma^{T}_{Li\text{-}7}$	1.00×10^{-3}

FHR Tritium Production Rate is Not Constant

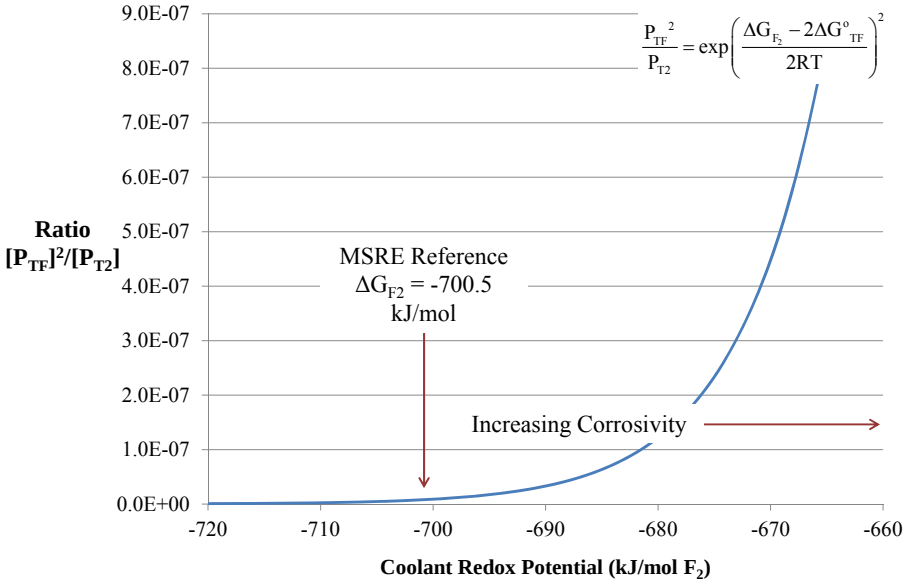


Basic Elements of TRIDENT: Effect of Redox on Corrosion and Tritium Behavior

Coolant Chemical Redox Potential Required as a Point of Reference for Modeling

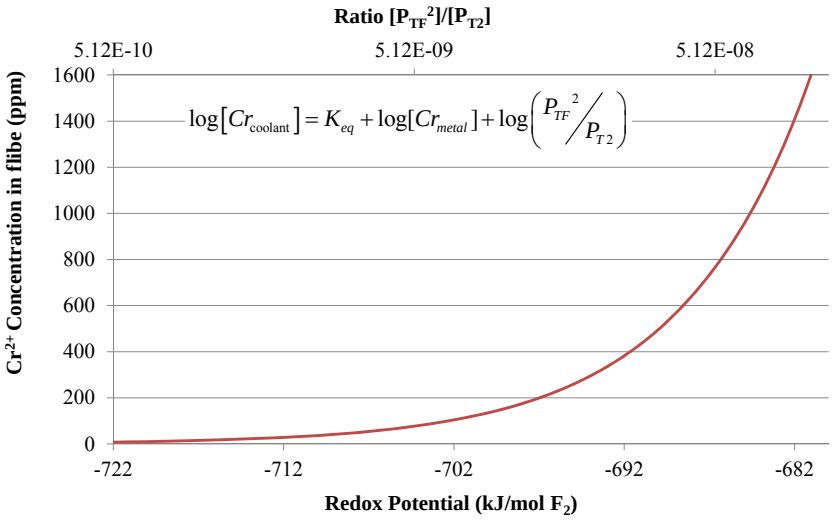


Redox Potential Dictates Relative Amounts of T_2 and TF



Redox Potential Determines Extent of Corrosion

- For Type 316L SS in flibe at 650 °C

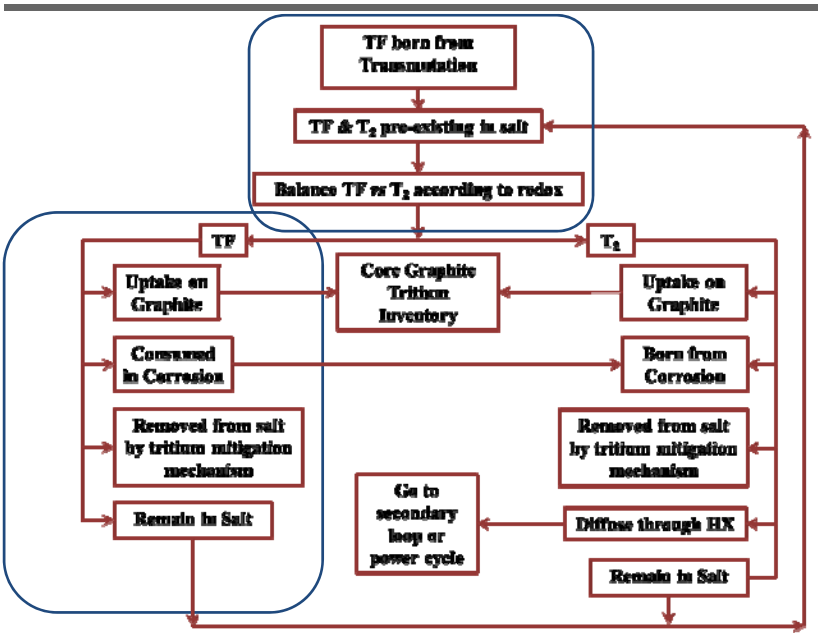


TRIDENT Tritium Diffusion and Corrosion Models Were Benchmarked Against Experiments

- Tritium diffusion in Nickel/Flibe and Nickel/Flinak systems
  - Experiment: Fukada, S., Morisaki, A., "Hydrogen permeability through a mixed molten salt of LiF, NaF and KF (Flinak) as a heat-transfer fluid," *Journal of Nuclear Materials*. **358**, 235–242 (2006).
  - Experiment: Calderoni, P., Sharpe, P., Hara, M., Oya, Y., "Measurement of tritium permeation in flibe (2LiF–BeF2)," *Fusion Engineering and Design*. **83**, 1331–1334 (2008).
- Corrosion and corrosion product mass transfer in flibe containing dissolved UF_3/UF_4
  - Experiment: Keiser, J.R., "Compatibility Studies of Potential Molten-Salt Breeder Reactor Materials in Molten Fluoride Salts," ORNL/TM-5783, Oak Ridge National Laboratory, (1977).

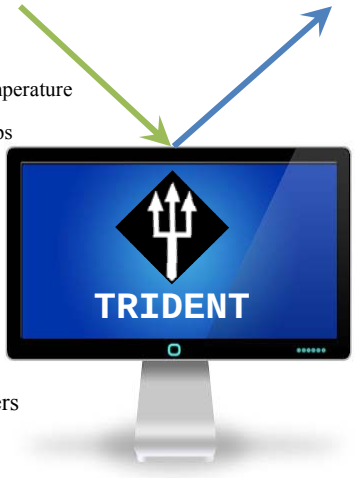
Modeling Tritium Behavior in the FHR: TRIDENT Code Description

TRIDENT – TRItium Diffusion EvolutioN and Transport



TRIDENT – Selected Input/Output

- Reactor Parameters
  - Thermal power
  - Coolant inlet/outlet temperature
  - Number of coolant loops
  - Core dimensions
  - One group flux
- Fuel parameters
  - Refueling rate
  - Pebble size
- Heat exchanger parameters
  - Tube diameter
  - Number of tubes
  - Total surface area

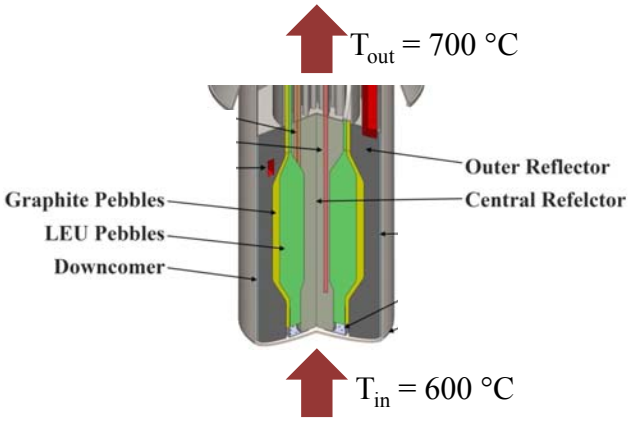


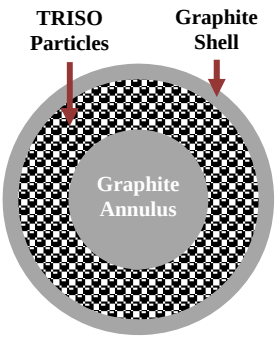
- Tritium distribution:
  - Coolant
  - HX tube walls
  - Graphite
  - Power cycle
- Tritium release rate
- Corrosion rates
- Options
  - Redox potential
  - Performance of tritium mitigation systems

TRIDENT – Materials Property Inputs

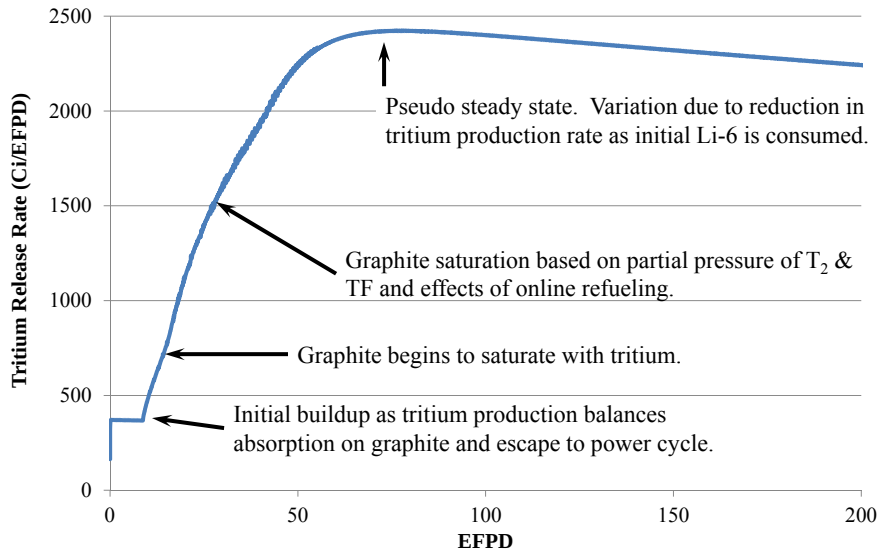
Material Properties
Baseline Redox Potential (specified as fluorine potential or ratio)
Flibe specific heat
Flibe density
Flibe viscosity
Henry's law constant for T_2 in flibe
Henry's law constant for TF in flibe
Diffusion coefficient for T_2 in flibe
Diffusion coefficient for TF in flibe
Diffusion coefficient for Cr^{2+} in flibe
Initial dissolved Cr concentration in the salt
Sieverts law constant for T_2 in 316 SS
Diffusivity of T_2 in 316 SS
Baseline permeation reduction factor
Cr grain boundary diffusion coefficient in 316 SS
316L SS elemental composition
316L SS density
316L SS lattice parameter
316L SS grain diameter
316 L grain boundary width
Graphite capacity for tritium
Graphite (IG-110) density
Nuclear cross sections relevant to tritium production in flibe

Results of TRIDENT Simulations of Baseline 236 MWt Mk1 PB-FHR

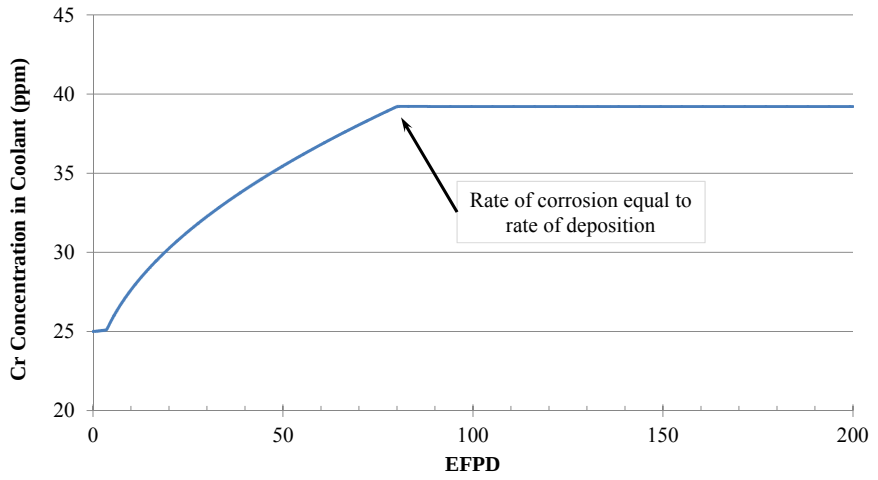




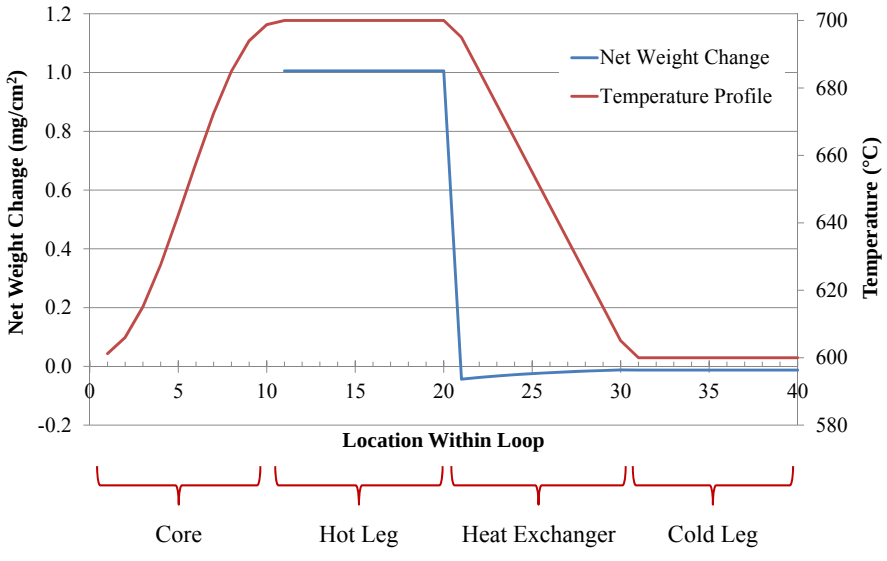
Tritium Release Rate (Ci/EFPD) to Power Cycle



Cr Coolant Concentration (ppm)



Net Weight Change After 200 EFPD Due to Cr Mass Transfer



FHR Release Rate Without Tritium Capture is High

- FHR tritium release rate with no engineered tritium mitigation systems:

  ~ 2500 Ci/EFPD for 236 MWt PB-FHR (10600 Ci/GWD)

- HWR tritium release rate:

  20 Ci/GWD

- LWR tritium release rate:

  < 1 Ci/GWD

TRIDENT Simulations of Proposed Tritium Mitigation Methods

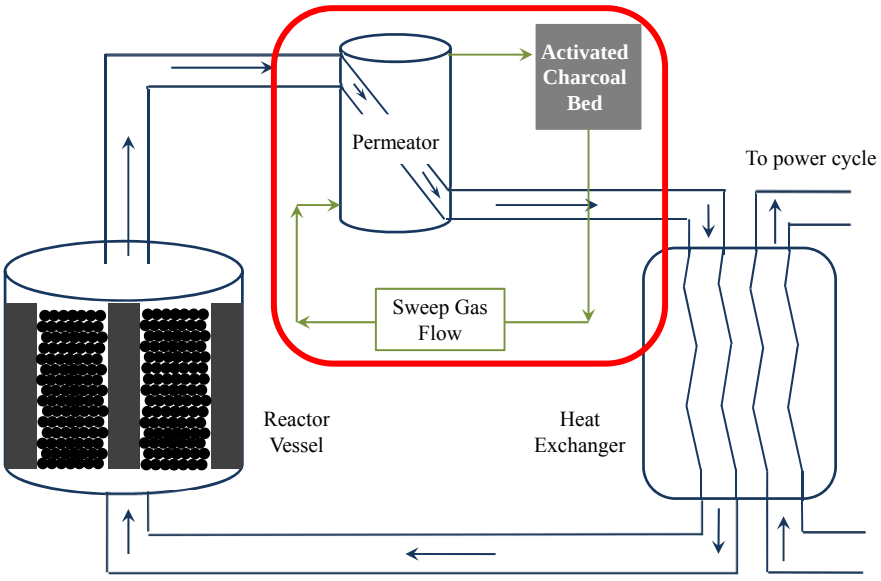
- Permeation windows
- Counter-current gas stripping
- Capture on graphite outside of core
- Oxide layer on air-facing side of heat exchanger
- Tungsten heat exchanger
- Increased Li-7 enrichment in flibe

Permeation Window

- Make a structure with a high surface area
- Construct the structure out of a metal with high hydrogen permeability

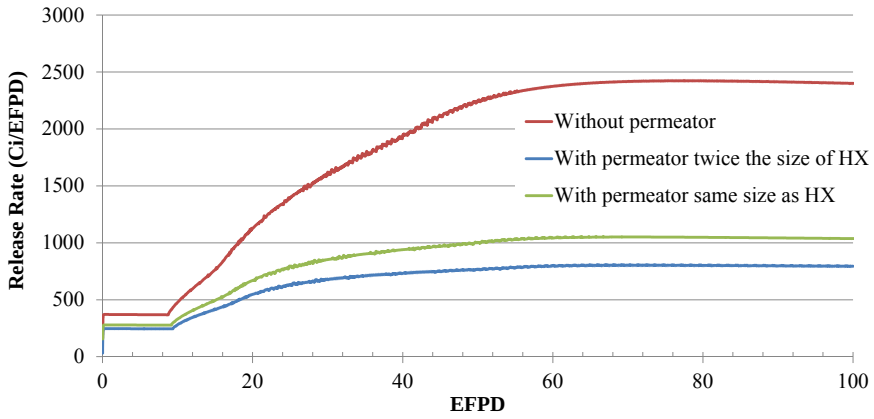
	Permeability (mol H_2/m-s-$MPa^{0.5}$) at 873 K	Reference
316 SS	5.5×10^{-8}	Tanabe, 1984
Ni	3.7×10^{-7}	Tanabe, 1984
Pd	2.5×10^{-5}	Steward, 1983

Permeation Window Schematic

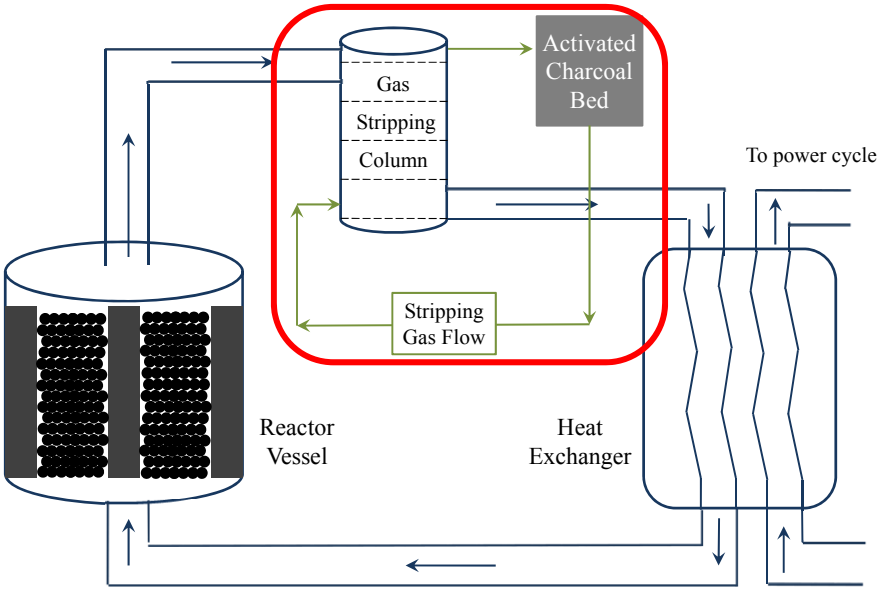


Release Rate with Permeation Window < 800 Ci/EFPD

- Ni permeation window, salt on one side, sweep gas on other side
- Permeator surface area = HX and 2 times HX
- 100% of coolant flow

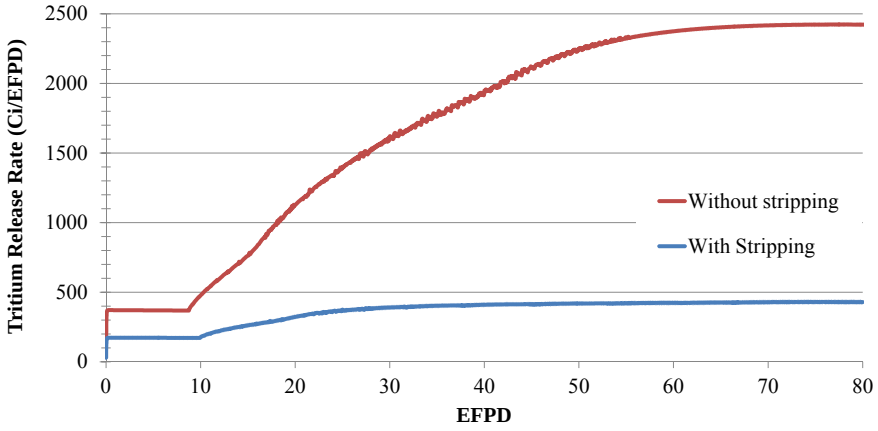


Counter-current Gas Stripping Schematic

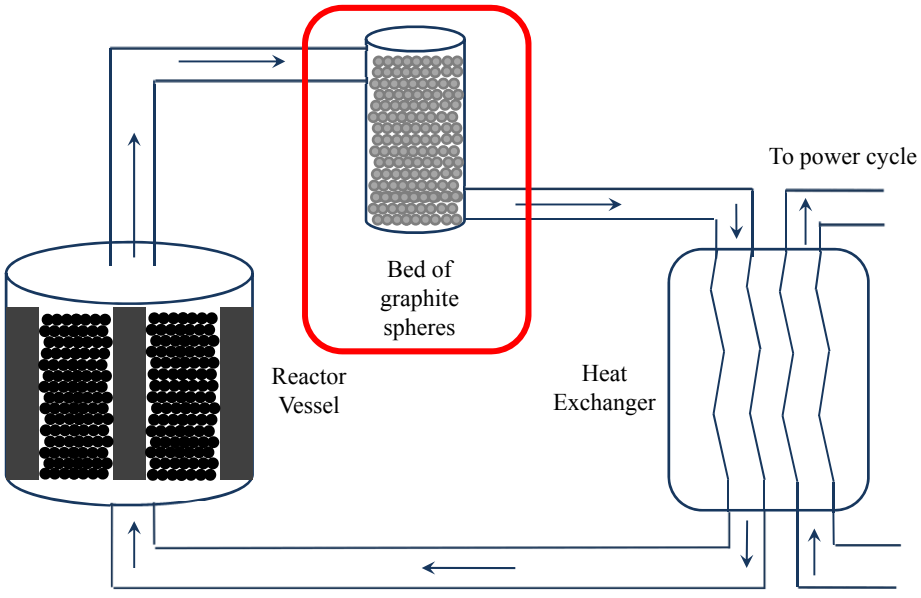


Release Rate with Gas Stripping < 500 Ci/EFPD

- 10 stripping stages
- 50% of primary coolant flow (total flow: 498 kg/s, 250 L/s)
- 2×10^4 L/hr stripping gas flow rate (5.6 L/s)

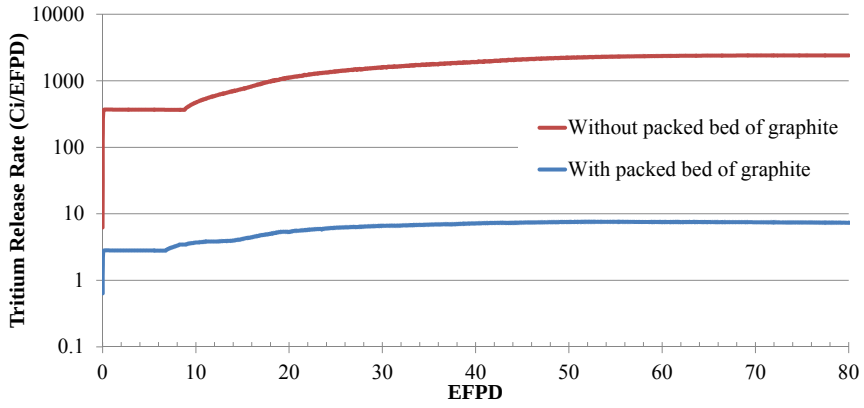


Mitigating Tritium Release: Graphite Capture



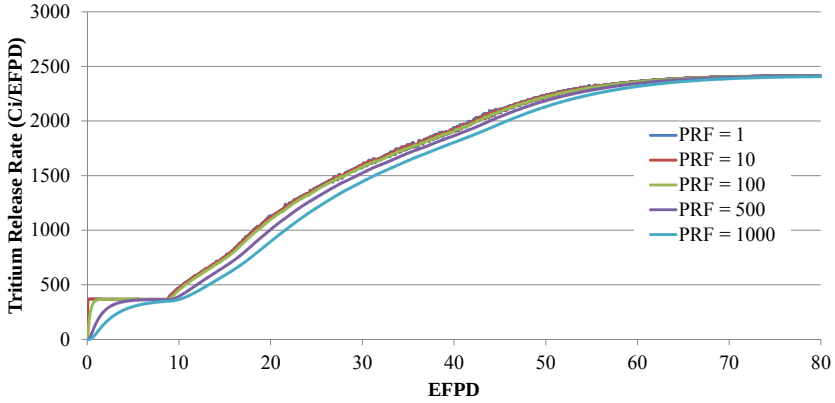
Release Rate with Capture on Graphite with Online Replacement < 10 Ci/EFPD

- Pebbles replacement rate same as core pebble refueling rate (1/30th per day)
- Total bed graphite surface area: 1945 m^2
- Friction pressure drop: 0.50 atm



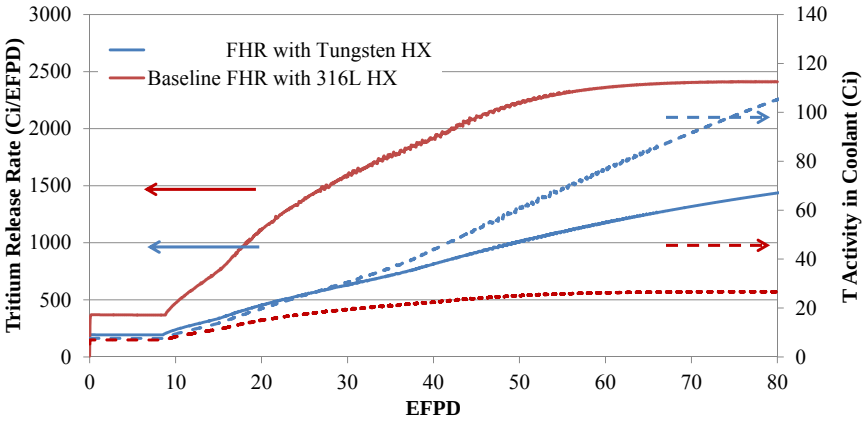
Release Rate with Variable PRF

- Baseline assumes permeation reduction factor (PRF) of 10 for 316 SS
- Increasing PRF causes a pile-up of tritium in the HX tube wall
- Increased PRF delays, but does not reduce tritium release

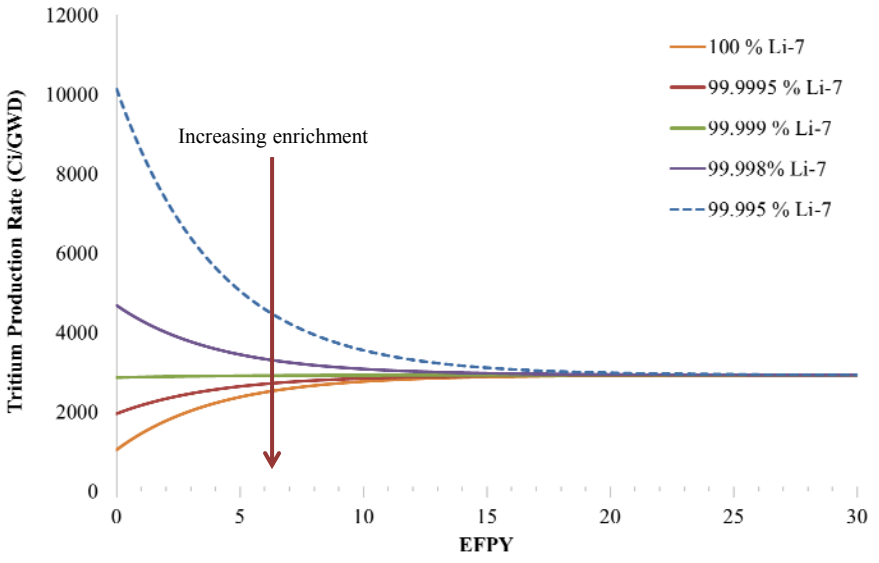


Tritium Transport with Tungsten Heat Exchanger

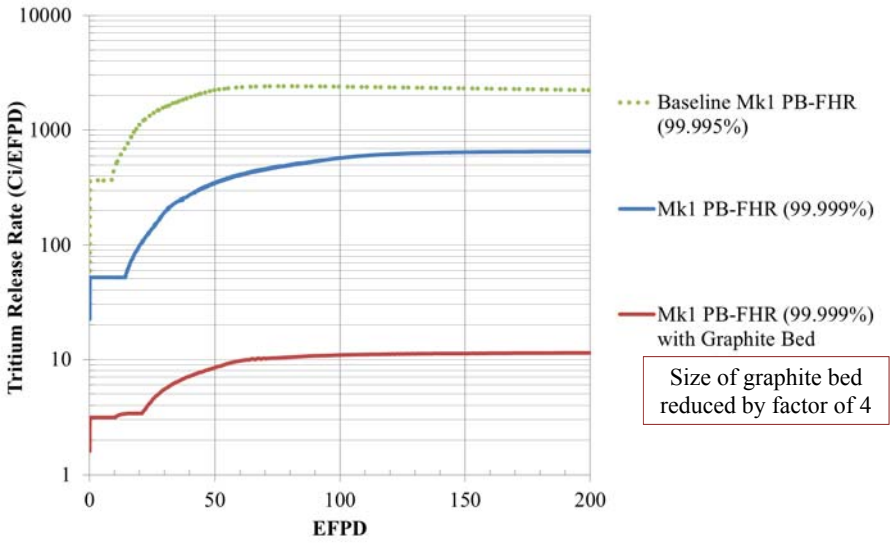
- Tungsten is stable in flibe
- W has low hydrogen solubility: 1.4E-3 mol H_2/m^3-$MPa^{0.5}$ compared to 62.9 for 316
- Could be applied as flibe facing coating on stainless steel (apply Ni between W and SS)
- Also would protect Cr from selective oxidation



Variation in Tritium PRODUCTION RATE with Li-7 Enrichment



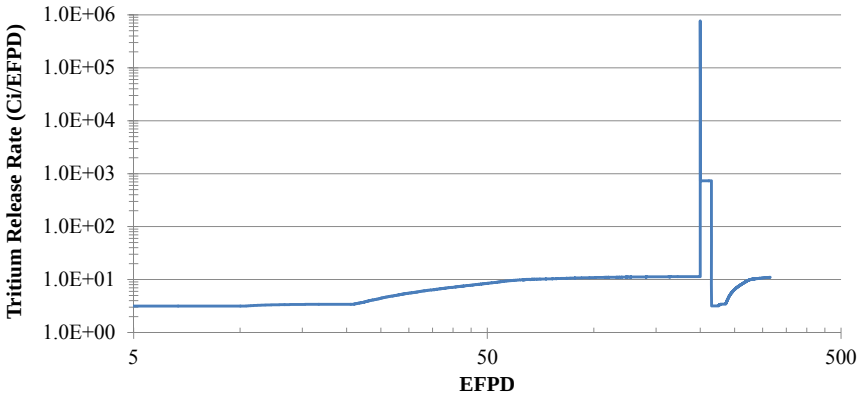
Variation in Tritium RELEASE RATE with Li-7 Enrichment



Analysis of tritium behavior at above-normal temperatures in the FHR for Beyond Design Basis Accidents (BDBA)

Tritium Desorption at High Temperatures Simulated with TRIDENT

- System allowed to reach steady state
- Coolant inlet and outlet temperatures increased by 300 °C and maintained for 15 days
- Normal operation resumed after 15 days at high temperature



Conclusions and Future Work

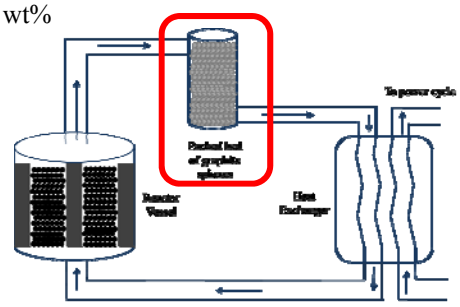
TRIDENT Provides FHR System Modeling Capabilities

- TRItium Diffusion EvolutioN and Transport (TRIDENT)
  - Corrosion and tritium behavior coupled
  - Predicted tritium distribution/release in FHR systems
  - Predicted/Compared effectiveness of tritium capture systems

Conclusions on Tritium and Corrosion

Simulations show:

- Corrosion rate with controlled redox: 0.08 mg/cm^2 per EFPY
- Tritium release rates without engineered solutions: 2500 Ci/d
- Proposed New Solutions:
  - Sorption on bed of graphite release rates < 10 Ci/EFPD
  - Increase Li-7 enrichment to 99.999 wt%
  - Use of W permeation barrier



Selected Future Work

- Wide option space for tritium control, need to optimize capture systems
- Explore use of graphite specifically engineered for tritium capture (outside of core)
- May include radiation effects on graphite for tritium absorption
- Tritium/Protium isotopic exchange reactions if H_2 deliberately added to system
- Add detail to corrosion model: currently 1D grain boundaries, in reality grain boundaries are 3D networks
- Highlights need for experimental work:
  - Tritium transport in flowing salt contacting metal membranes and graphite
  - Tritium uptake and desorption kinetics on graphite in salt at low partial pressures and relevant temperatures

Thank You

Questions
Comments

john.stempien@inl.gov

FHR

Massachusetts Institute of Technology

NSE Nuclear Science and Engineering

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Irradiation Testing in Support of the Tritium Production Enterprise

DJ SENOR

Pacific Northwest National Laboratory
NUEN 481/681, Texas A&M University

PNNL Roles in the Tritium Program

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- TPBAR Design Authority
- TPBAR and lead use rod design and analysis
- WBN1 coolant tritium analysis and operations support
- TPBAR component development and testing
- TPBAR component procurement and assembly support
- Tritium extraction development and support
- Basic and applied research and development
  - Post-irradiation examination (PIE)
  - Ex-reactor testing
  - In-reactor testing (design, fabrication, PIE)
  - TPBAR performance model development

Tritium Production Enterprise: Background

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- Tritium is required for US nuclear weapons stockpile
- Tritium has a 12.3 year half-life and must be replenished
- 1988: DOE ceased production of tritium at SRS
- 1988-1992: The US considered the use of dedicated reactors for tritium production
  - Heavy water reactors (HWRs)
  - High temperature gas-cooled reactors (HTGRs)
  - Light water reactors (LWRs)
- 1995-1998: The US considered dual-use facilities
  - Commercial LWRs
  - Accelerators
- 1995: PNNL selected by DOE to be Design Authority for Commercial Light Water Reactor irradiation demonstration

L Reactor at SRS

Tritium Production Enterprise: Background

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- 1995 – 1997: Lead Test Assembly (32 Tritium-Producing Burnable Absorber Rods, TPBARs) designed and built at PNNL for irradiation in TVA Watts Bar Nuclear Unit 1
- 1999: Post-irradiation examination of LTA
- 2000: The current Commercial Light Water Reactor tritium program was selected by DOE over accelerators for production
- 2001 – 2003: Design and manufacturing scale-up for production TPBARs
- 2003: First production core (240 TPBARs) irradiated at WBN1
- 2005 – 2008: TPBAR design modifications
- 2008: Modified TPBARs (Mark 9.2) first irradiated at WBN1

Watts Bar Nuclear Plant
Spring City, TN

Tritium Target Current Technology

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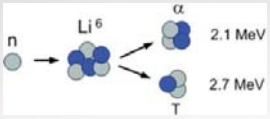
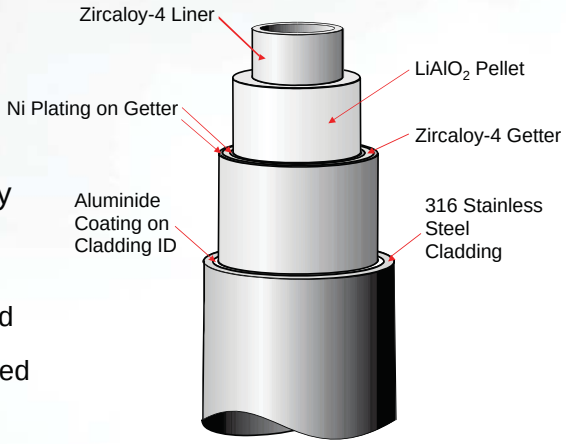
- TPBARs replace burnable absorber rods normally used in Westinghouse PWRs (WABAs)
  - WABA reaction:
    - $^{10}B + {}^{1}n_{th} \rightarrow {}^{4}He + {}^{7}Li$
  - TPBAR reaction:
    - $^{6}Li + {}^{1}n_{th} \rightarrow {}^{3}H + {}^{4}He$
- Reactivity worth of TPBARs is slightly greater than WABAs
- Because TPBARs provide reactivity hold-down, they are considered a safety-related component by the NRC
  - All irradiation testing work governed by QA requirements in 10 CFR 50, Appendix B so results can be applied to TPBAR modeling and design





Not to scale

TPBAR Irradiation Performance

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- In 2004, during the first production cycle at WBN1, it was determined that TPBAR tritium permeation was higher than predicted by performance models
  - Predicted ≈ 0.5 Ci/TPBAR/cycle
  - Actual ≈ 4 Ci/TPBAR/cycle
- Even 4 Ci/TPBAR/cycle represents only about 0.04% of the tritium produced
- TVA limited the number of TPBARs that could be irradiated because of current license limits on tritium release
- Subsequent irradiations have continued, but quantities are limited to <704 TPBARs/cycle
- An irradiation testing program was implemented in 2006 to provide a scientific basis for improving performance models and providing systematic, long-term TPBAR design evolution

Irradiation Testing Program Objectives

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- Overall goal is risk reduction through fundamental understanding of TPBAR performance
  - Accurately explain and predict existing permeation performance
  - Provide confidence in performance predictions to support
    - Operating condition changes
    - Supplier changes
    - Manufacturing process changes
  - Provide basis for evolutionary design changes
- The testing program was tailored to address these objectives in support of the tritium production mission

Irradiation Testing Program

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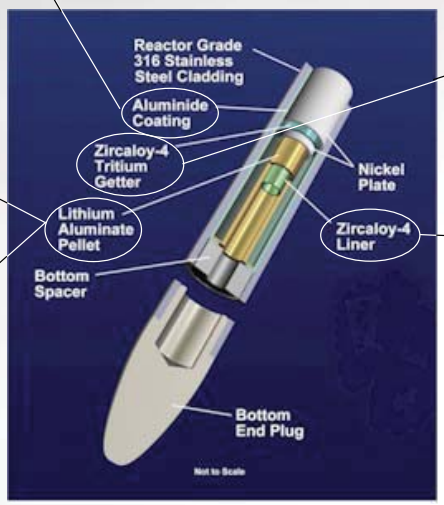
Cladding Permeation
TMIST-2
2006-2012

Getter Performance
TMED-4
2008-2010

Pellet Performance
TMIST-3
2009-2019

Advanced Pellet Mfg
TMED-3
2008-2011

Liner Oxidation
TMIST-1
TMED-1
2006-2010



Motivation

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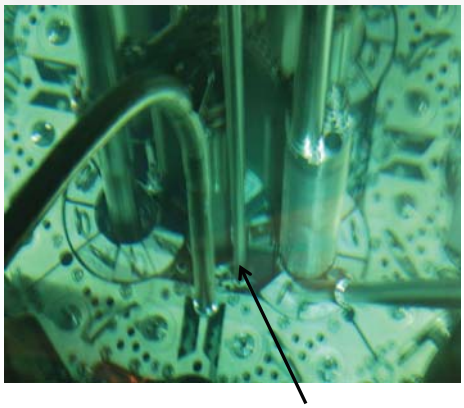
- TMIST-1/TMED-1
  - TPBAR liners are responsible for reducing T_2O released by pellets so that T_2 can be captured by getters
  - Nascent tritium uptake in liners is beneficial
  - In-reactor oxidation rates of liner materials at low water partial pressure needed for improved TPBAR performance modeling
  - Materials with higher oxidation rates may be needed to improve TPBAR performance
- TMIST-2
  - Hydrogen isotope permeation through stainless steel is enhanced by irradiation (Irradiation Enhancement Factor, IEF)
  - Ex-reactor permeation may have different rate-controlling mechanism than in-reactor permeation at very low pressures (i.e. surface decomposition versus diffusion)
  - In-situ measurements support TPBAR performance modeling



TMIST-1 leadout in the Advanced Test Reactor, Idaho National Laboratory

Objectives

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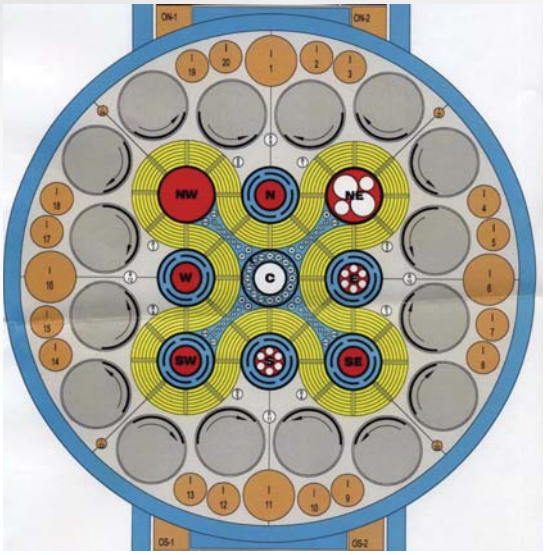
- TMIST-1/TMED-1
  - Quantify temperature and pressure dependence of in-reactor oxidation for liner materials
  - Quantify nascent fraction of hydrogen isotopes deposited in test specimens during oxidation
  - Evaluate irradiation performance of advanced liner materials
- TMIST-2
  - Quantify irradiation enhancement factor
  - Determine temperature/pressure dependence of tritium permeation through stainless steel
  - Estimate permeation contribution from triton recoil resulting from He-3 conversion



ATR Core Map

Test Specimens and Conditions TMIST-1/TMED-1

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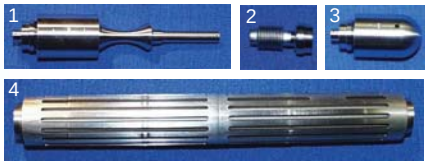
- Test Specimens
  - Zircaloy-4 (radial and uniform texture)
  - Zircaloy-2
  - Surface-Modified Zircaloy-4 with four different OD Ni plating thicknesses
    - 2.5, 5, 7.5, and 10 μm
  - Seamless tubing
    - 5.3 mm OD x 0.1 mm wall x 25 mm long
- Irradiation Conditions
  - Temperatures of 330 and 370˚C
  - He carrier gas at ~1 atm and 30 sccm
  - D_2O partial pressures of 300 and 1000 Pa
  - Irradiated for ~138 effective days at temperature (peak fast fluence, E>0.1 MeV, ~2 x 10^{21} n/cm^2)

Several TMIST-1 samples with fixturing



Selected capsule components showing (1) bulkhead, (2) bellows, (3) bottom end plug, and (4) capsule body

Materials for TMIST-1/TMED-1

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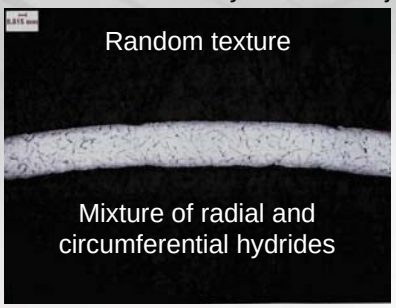
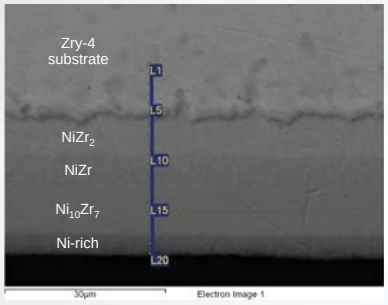
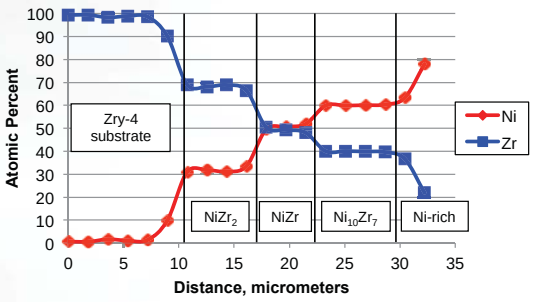
As-fabricated Zry-4 liners subjected to hydride orientation test







Post heat treatment microstructure of Surface-Modified Zry-4 with initial 10 μm Ni coating

Capsule Design TMIST-1/TMED-1

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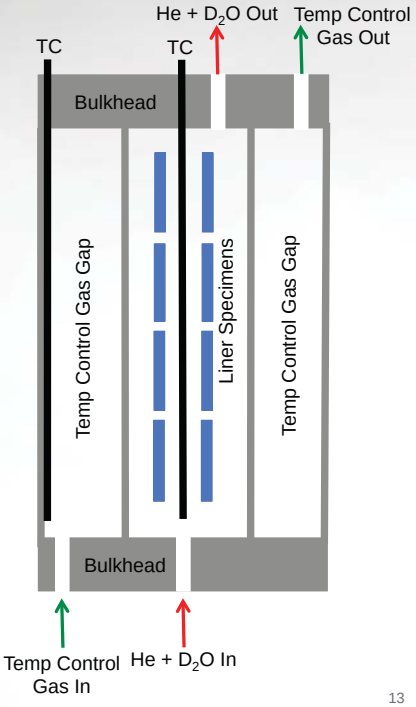
- Four specimens per capsule
- Four capsules with individual temperature control gas
  - Each specimen had unique gas gap dimensions within capsule
- Active temperature control using Type K thermocouples and He-Ne mixture in gas gap
- Fixturing to center specimens in capsule, minimize axial temperature gradients, and accommodate adjacent specimen degradation



Post-Exposure Specimen Condition

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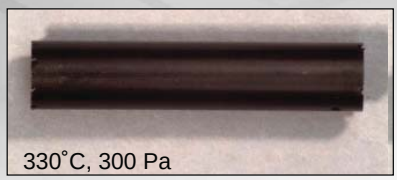
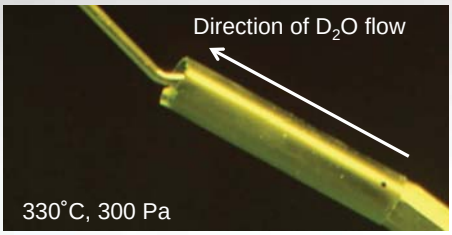
Ex-Reactor Exposure



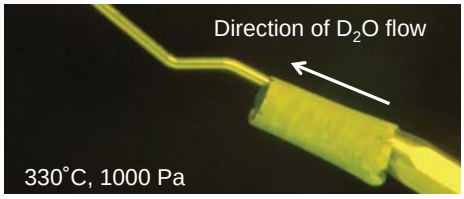
Zircaloy-4 – Fully Intact

SM-10 – Some Distortion and Cracking

In-Reactor Exposure



Zircaloy-4 – Fully Intact



SM-10 – Partially Consumed

Because of SM specimen degradation, mass measurements were only useful for the Zircaloy samples

Experiment Performance
TMIST-1/TMED-1

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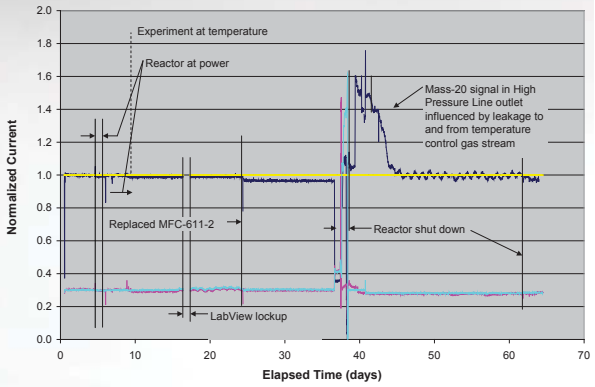
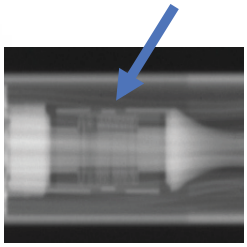
- No significant depletion of D_2O at either supply pressure
- D_2O leak observed during second ATR cycle in 1000 Pa/370°C capsule
  - Leak mitigated by differential pressure control in temperature control gas
  - Post-irradiation neutron radiography revealed a tear in the bellows as the cause of the leak
- Capsule temperature setpoints maintained to within ±5°C
- Four thermocouples failed during irradiation (one per capsule)
  - Temperature control maintained with redundant thermocouples





Results
TMIST-1/TMED-1

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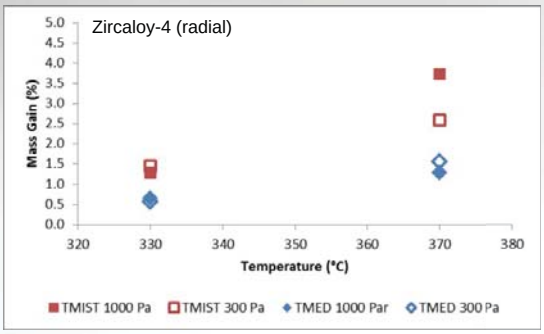
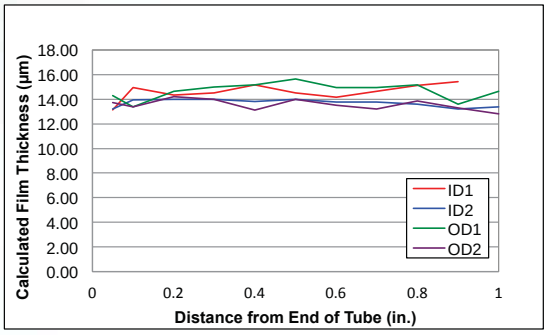
- Mass Gain - Zircaloy
  - Increased with temperature both ex-reactor and in-reactor
  - No obvious dependence on D_2O partial pressure
  - Irradiation enhancement by a factor of ~2-3X at both temperatures for Zircaloy specimens
- Fourier Transform Infrared Spectroscopy
  - FTIR oxide thickness measurements unreliable due to fine structure patterns in the diffraction spectra
  - FTIR data showed uniform oxidation along sample length on both inner and outer surfaces





Results
TMIST-1/TMED-1

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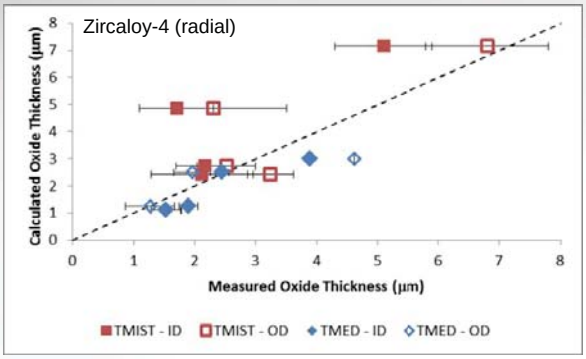
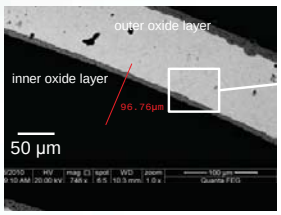
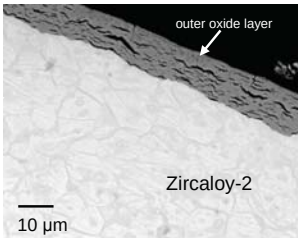
- Oxide Thickness - Zircaloy
  - Measured via optical and scanning electron microscopy
  - Mass gain measurements used to compare measured and calculated oxide thickness assuming uniform growth of tetragonal zirconia
  - In Zr-base alloys, a transition from dense tetragonal to porous tetragonal + monoclinic oxides occurs around 2 μm
    - Care must be taken when comparing pre- and post-transition oxides







Zircaloy-2
Irradiated at 370°C and 1000 Pa

Results
TMIST-1/TMED-1

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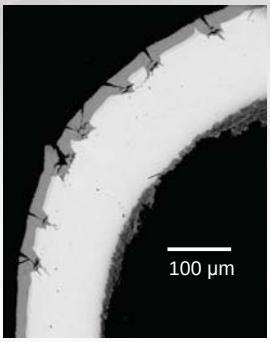
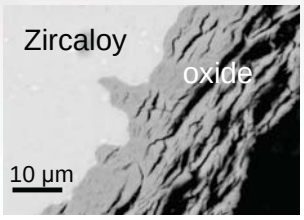
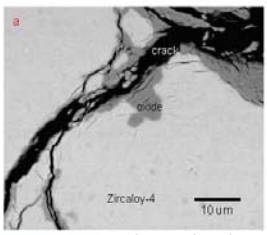
- Oxide Thickness – Surface Modified
  - Little irradiation enhancement is evident with thicker SM layers
  - Thicker SM layers were very prone to spalling
  - Dependence on both temperature and pressure
  - Inner surface of SM samples exhibited significantly more oxidation than Zry-4 specimens, indicating heat treatment affected corrosion resistance of substrate
- Large radial cracks observed in SM samples extending into substrate
- Prevalence of cracks higher for thicker SM samples
- SM layers thicker than 2.5 μm are probably not desirable due to extent of oxidation and deleterious impact on Zircaloy substrate

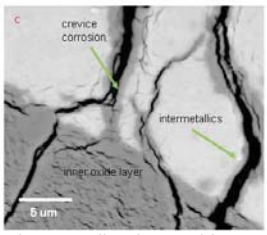


SM-7.5 specimen showing severe bending



SM-10 specimen showing significant delamination within oxide layer





SM-5 specimen showing cracks extending from oxide into substrate with substrate oxidation confirming that cracks formed during irradiation

Results
TMIST-1/TMED-1

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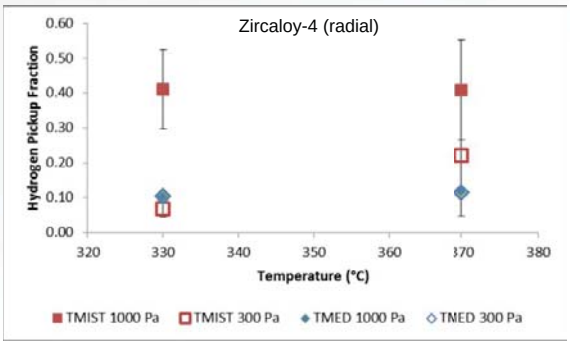
- Nascent Hydrogen Uptake - Zircaloy
  - Ex-reactor results appear insensitive to temperature and pressure
  - Temperature and pressure dependence of in-reactor results is unclear
  - Hydrogen uptake can be significantly enhanced by irradiation (1-4X)
- Nascent hydrogen Uptake – Surface Modified
  - Significantly more uptake than Zircaloy samples at same conditions
    - 2,000-30,000 ppm D
  - Irradiation enhancement of uptake (1-6X) is greater than irradiation enhancement in oxide thickness
  - Temperature and pressure trends are inconsistent
  - No significant dependence in D_2 uptake on SM layer thickness
  - Thicker SM layers had higher H_2 uptake (up to 10% of D_2 uptake)



Results
TMIST-1/TMED-1

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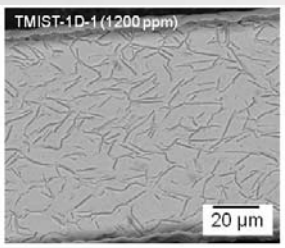
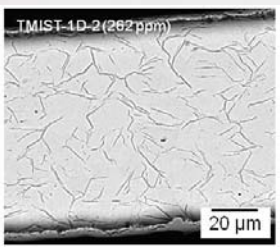
- Hydride Morphology
  - Appears to be similar for in-reactor and ex-reactor exposure
  - Irradiation does not seem to affect hydride morphology
  - Hydride morphology dictated by Zircaloy texture
    - Hydrides form parallel to basal planes

Hydride Orientation in Zry-4 Specimens Irradiated at 330˚C and 1000 Pa D_2O

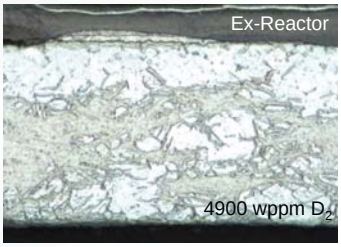


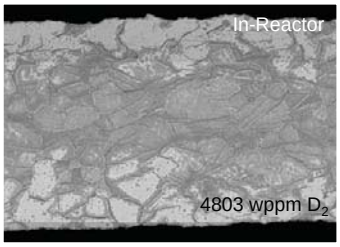
Radial Texture



Uniform Texture

Hydride Orientation in SM-5 Specimens Exposed at 330˚C and 300 Pa D_2O





Test Specimen and Conditions TMIST-2

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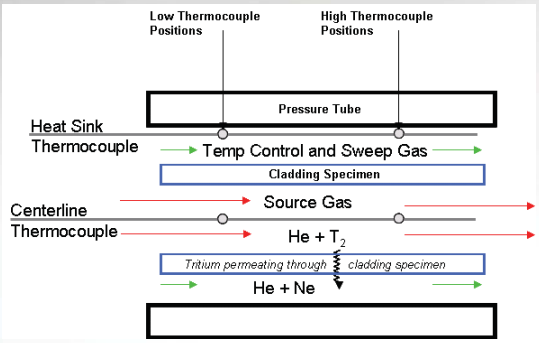

- Test Specimen
  - 316 stainless steel
  - 0.85 cm ID x 0.97 cm OD
- Irradiation Conditions
  - Temperatures of 292 and 330°C
  - He carrier gas at ~1 atm and 30 sccm
  - Tritium partial pressures of 0.1, 5, and 50 Pa, pre-mixed in carrier gas cylinders
  - Irradiated for ~200 effective days (peak fast fluence, E>0.1 MeV, est. ~3 x 10^{21} n/cm^2)
  - Temperature and pressure set points changed online after obtaining satisfactory permeation measurements on each experiment step
  - At least two separate measurements at each combination of temperature and pressure to evaluate possible fluence effects

TMIST-2 Test Capsule

Permeation Measurement System TMIST-2

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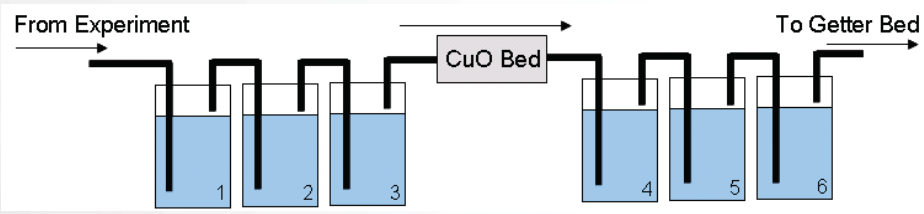
- Tritium Permeation Measurements
  - Real-time indication of tritium permeation rate provided by ion chambers
    - Used to determine when permeation reached steady-state (usually several days)
  - Quantitative tritium permeation results determined from periodic scintillation counter measurements of DI water bubbler vials
    - At least two collections of at least 24 hr each time a temperature/ pressure combination was tested
    - Two banks of three vials each to differentiate between T_2O/HTO and T_2/HT



Results
TMIST-2

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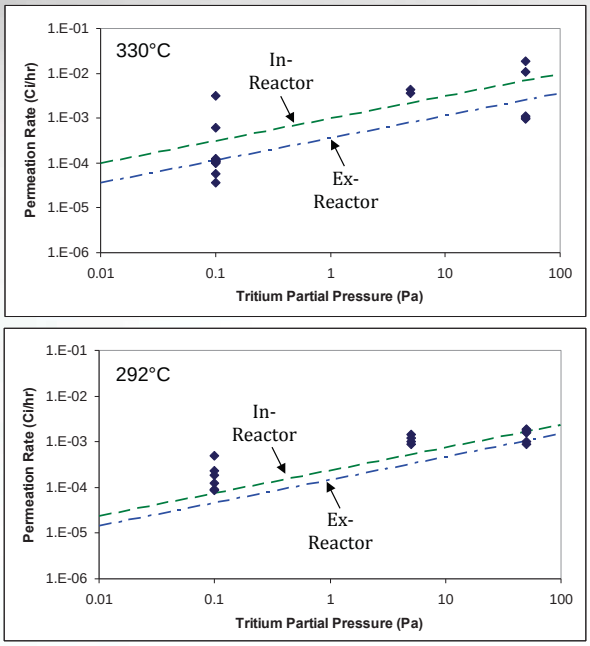
- In-reactor permeation rates through TPBAR cladding measured for the first time
- Permeation data show definite irradiation enhancement relative to ex-reactor behavior
  - Mechanism unclear
  - Currently evaluating microstructural evolution of TPBAR cladding during irradiation
- New TPBAR permeation model developed to address irradiation effect



TMIST-2 Data Showing Enhanced (~3X) In-Reactor Tritium Permeation Rate Through Uncoated Stainless Steel

Rate-Limiting Permeation Mechanisms

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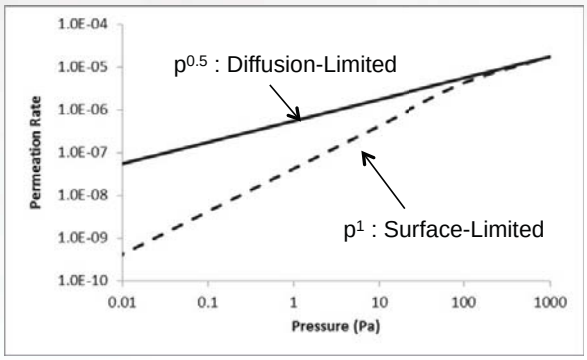
- Ex-reactor permeation measurements
  - > 100 Pa → Diffusion-limited → $p^{0.5}$
  - < 100 Pa → Surface-limited → p^{1}
- In-reactor permeation mechanism uncertain
  - Direct dissociative chemisorption
    - Molecules adsorb and readily dissociate upon contact
    - Disrupted ex-reactor at low pressure by surface impurities or oxide films
  - Radiation-enhanced dissociation
    - Radiolysis of T_2 in gas phase
    - Physical or chemical changes in surface in-reactor



Results
TMIST-2

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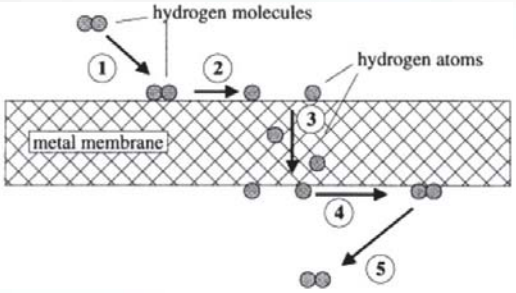
- Pressure dependence of $p^{0.5}$ observed
  - Suggests tritium permeation is rate-limited by diffusion, not surface decomposition (p^1)
- Temperature dependence consistent with diffusion
  - Scatter prevents determination of statistically significant trends in apparent activation energy
  - Nominal activation energies ~100 kJ/mol, consistent with diffusion-limited mechanism
  - Trend for increased permeation with temperature observed within the scatter, as expected



Processes involved in permeation of hydrogen through metals

Results
TMIST-2

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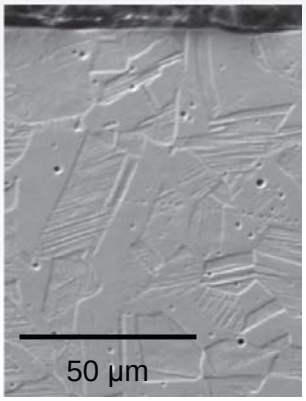
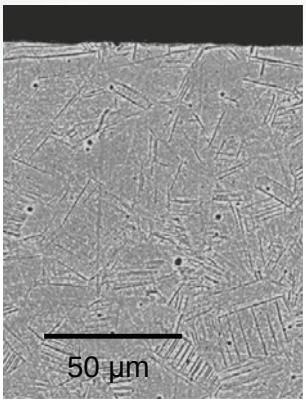
- Microscopy
  - After irradiation, grain boundaries are not as well defined
  - TEM necessary to evaluate extent of irradiation damage
- Auger Electron Spectroscopy
  - After irradiation, the inner surface of the specimen appears to be enriched in carbon and depleted in oxygen
    - Reducing atmosphere during test
    - Source of carbon?

Pre-Irradiation



SEM Image

Post-Irradiation



SEM Image

Results
TMIST-2

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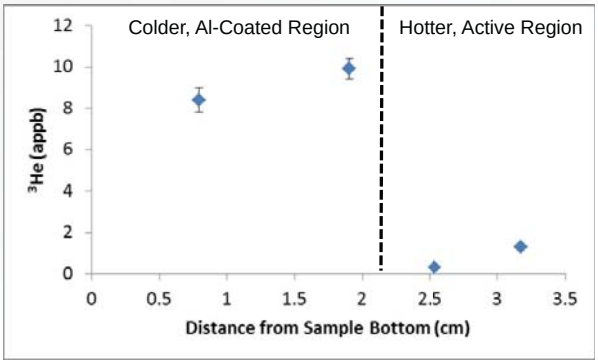
- Low-level ^{3}He Assay
  - Sample outer surface area below active region sputter-coated with Al to inhibit permeation
  - ^{3}He measurements made within and below active (hot) sample region
  - ^{3}He acts as a tracer for tritium
  - Higher concentration of ^{3}He in cold, sputter-coated region suggests slower tritium permeation



Results
TMIST-2

Pacific Northwest NATIONAL LABORATORY
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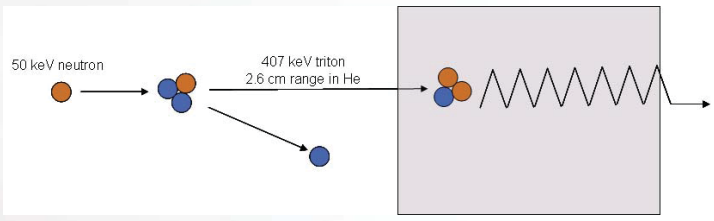
- Utilized source gas mixture of ^{4}He + 1% ^{3}He
- Calculations determined the conversion rate of He-3 to tritium
  - Used average neutron energy spectra for ATR
- Triton recoil distance large relative to inner diameter (2.6 vs. 0.85 cm)
- Implantation depth in stainless steel is ~ 2 μm
- Less than 4.1% (2σ confidence) of the tritium resulting from He-3 conversion permeated through the test specimen



Data from the Testing Program Has Improved TPBAR Performance Predictions

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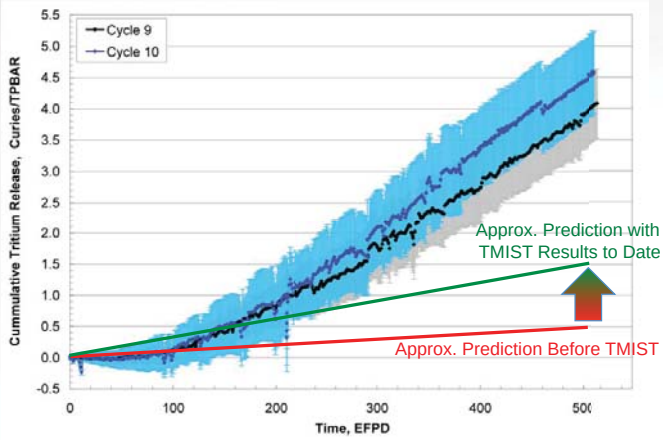
- TROD performance prediction code models updated with data from TMIST-1,TMED-1, TMIST-2, and TMED-4
- Discrepancy between predicted and observed permeation decreased by ~30%
- Time dependence still not correctly modeled
  - Will be improved by TMIST-3 data



Pellet Performance Irradiation Experiment TMIST-3

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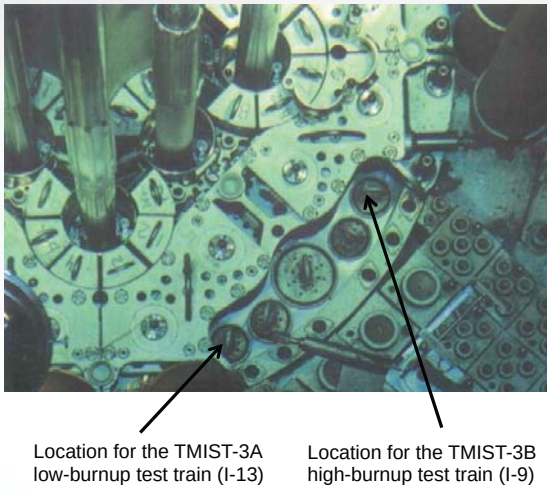
- Data from TMIST-3 will
  - Explain time dependence of pellet tritium release and its relationship to TPBAR permeation
  - Evaluate the speciation of tritium release as a function of burnup, burnup rate, and time (T_2O versus T_2)
  - Define relationships between pellet burnup, burnup rate, and tritium release to help define an acceptable TPBAR operational envelope
  - Improve fundamental understanding of pellet microstructure and its effects on performance
  - Provide a better definition of the pellet burnup limit
  - Determine whether modifications to the pellets could improve TPBAR performance
    - Increased tritium retention
    - Increased TPBAR void volume



Test Specimens TMIST-3

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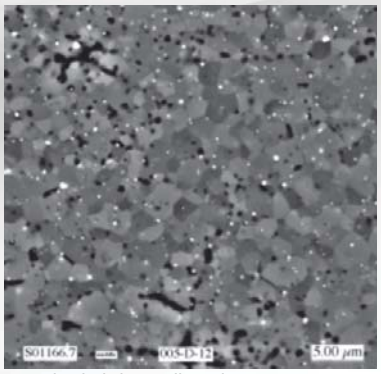
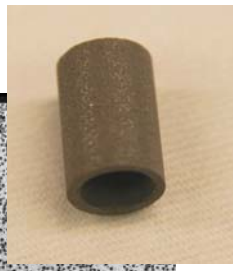
- Test specimens
  - Standard TPBAR $LiAlO_2$ pellets
    - 2 μm grain size
    - 97-98% TD
    - 1 mm wall thickness
  - Large grain $LiAlO_2$ pellets
    - 10 μm grain size
  - Porous $LiAlO_2$ pellets
    - Small pores (~90% TD)
    - Large pores (~85% TD)
  - Thin-wall $LiAlO_2$ pellets
    - 0.76 mm wall
  - Cermet pellets
    - $LiAlO_2$ particles in Zr matrix
    - Four ceramic particle loadings from 10-40 v/o



Standard $LiAlO_2$ pellet microstructure



Cermet pellet with 40 v/o LiAlO2

Capsule Design TMIST-3

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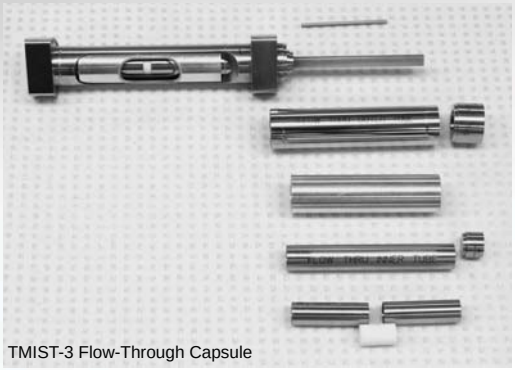
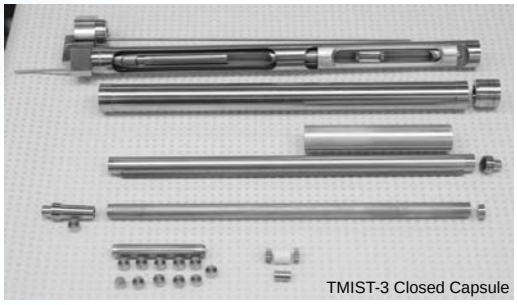
- Two test trains
  - TMIST-3A – Irradiate for ~1.5 yr
  - TMIST-3B – Irradiate for ~2.5 yr
- Two capsule types in each test train (41 total)
  - Flow-through – 15 total
  - Closed – 26 total
- All capsules have active He-Ne temperature control gas
  - One capsule designed to operate over a wide temperature range to evaluate temperature effects
- Flow-through capsules have He sweep gas to remove tritium for ex-reactor sampling
- 106 total leads for both test trains


TMIST-3 Flow-Through Capsule


TMIST-3 Closed Capsule

Capsule Design TMIST-3

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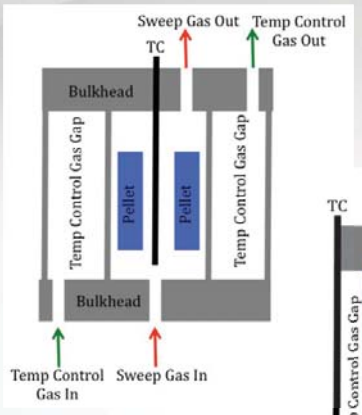
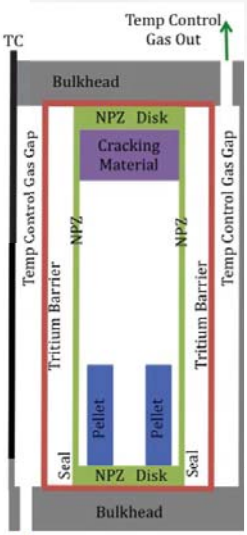
- Flow-through capsules
  - Used for time, burnup, burnup rate, and temperature dependent tritium release measurements
  - Tritium released from pellets is carried to ex-reactor measurement system for analysis
  - Total tritium measurement only
- Closed capsules
  - Used for speciation measurements and pellet integrity/retention tests
  - Tritium released from pellets as T_2 and T_2O is spatially segregated and gettered in-situ
  - Speciation data inferred from post-irradiation examination tritium assays



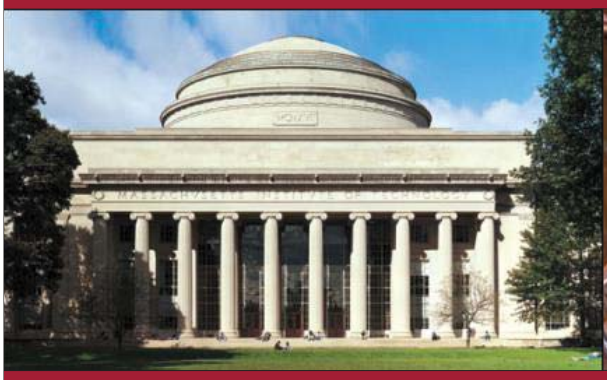
Flow-Through Capsule

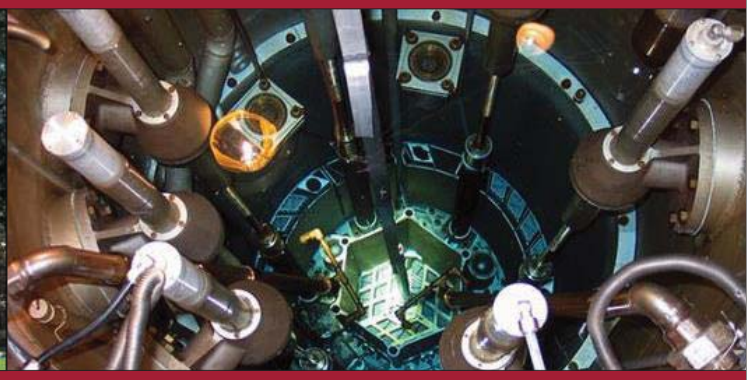


Closed Capsule

MIT NUCLEAR REACTOR LABORATORY

AN MIT INTERDEPARTMENTAL CENTER





Planned FHR IRP-2 Tritium Experiments at the MIT NRL

David Carpenter

Group Leader, Reactor Experiments

Outline

- Post-Irradiation Examination for FS-1/2
  - Thermal tritium release
  - Imaging
  - Other methods
- IRP-2 Experiments
  - Tritium uptake in graphite
  - Tritium and activation product release
  - Tritium diffusion from salt through metals
  - Ongoing PIE
- Suggestions, Lessons Learned?

Planned Post-Irradiation Tritium Tests

- Thermal release measurement of tritium uptake
  - Components of FS-1 and FS-2 will be progressively heated to above their irradiation temperatures to examine the tritium release
  - Important to have real-time and LSC measurement
- New methods for tritium measurement are being examined:
  - Graphite powdering and/or digestion followed by LSC
  - Pyrolysis followed by LSC
  - Proton or deuterium ion beam irradiation and gamma or fast neutron detection
  - Beta-plate imaging (concentration and gradients)

Upcoming MITR FHR Irradiations



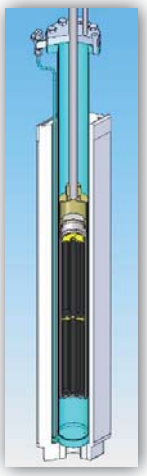

- Designing new experimental facilities based on lessons learned from the first two flibe irradiations
  - Control of salt condensates to prevent accumulation in gas lines
  - Off-gas holdup for short half-life decay (N^{16} and O^{19})
  - Avoiding <200°C radiolysis (fluorinated compound production)
- Collaboration with Chinese Academy of Sciences on irradiation of new graphite and SiC materials in flibe
- Three types of experiments planned for IRP-2
  - Release of tritium and activation product gasses from flibe
  - Tritium uptake on graphite
  - Tritium diffusion through metals

(1) Tritium and Activation Product Release

- Initial MITR irradiations highlighted (1) the effects of unmitigated activation and radiolysis product release, and (2) variability of that release with the salt temperature
- This test is dependent on simulating the FHR core environment because the proper neutron spectrum, neutron and gamma flux, and temperature is needed
- Planning new dedicated salt irradiation facility
  - Minimize tritium sinks
  - Maximize possible flibe volume
  - Variable cover gas (sparging?) with optional H injection up- and downstream
  - Improved “fast sampling”
  - Freedom to vary reactor power / temperature (electrical secondary heating)

(2) Tritium Uptake in Graphite

- The capture of tritium via adsorption onto or diffusion into graphite has the potential to be the primary method of tritium inventory management during FHR operation.
  - Tritium uptake into graphite observed during initial irradiations, but was not primary focus



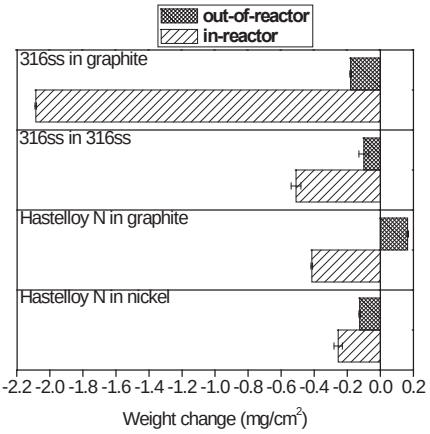
- Options for uptake facility
  - In-core, most representative environment but most limiting for size, controls, sampling
  - Independent facility using tritiated flibe generated in the MITR, gives flexibility
- Variables to investigate
  - Saturation
  - Temperature and thermal gradients
  - Radiation damage to materials, radiolysis, *in situ* generation
  - Salt chemistry, cover gas mixture
  - Material preparation (graphite types, surface preparation, etc.)

(3) Tritium Diffusion Through Metals

- Tritium transport out of the primary system is a critical phenomenon to understand and reliably control
  - Important to enable coupling to once-through gas turbine (i.e. salt-air heat exchangers)
- Work is underway (UNM) investigating double-walled heat exchangers
- Out-of-reactor facility with tritiated flibe to measure tritium transport from salt through metal surfaces
  - Primary piping
  - Heat exchangers
  - DRACS components
- Need to consider test matrix
  - Flow or static
  - Salt chemistry and temperature
  - Thermal gradients
  - Barrier coatings
  - Sweep gasses and secondary side media (air, helium, water, salt)

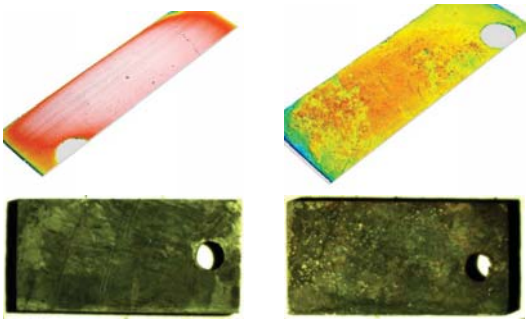
Post-Irradiation Examination

- Additional analysis on IRP-1 specimens continues – wealth of data is available
  - Weight, dimensions, spectroscopy, optical microscopy, and profilometry completed
  - Analysis of cracking in TRISO particles linked to combination of irradiation and salt freeze-thaw cycles



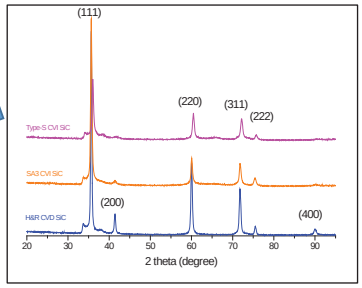
Accelerated corrosion of metals in-reactor compared to UW autoclave tests.

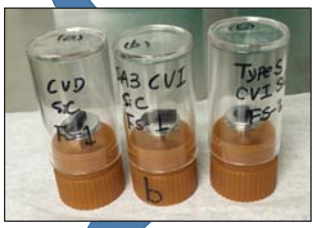
Observing effects of a flibe-only and a ternary environment.

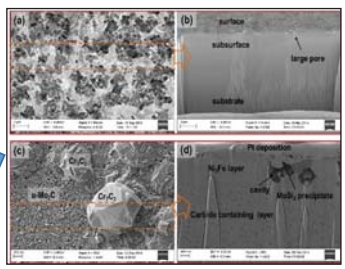


SS316 + flibe **SS316 + graphite + flibe**

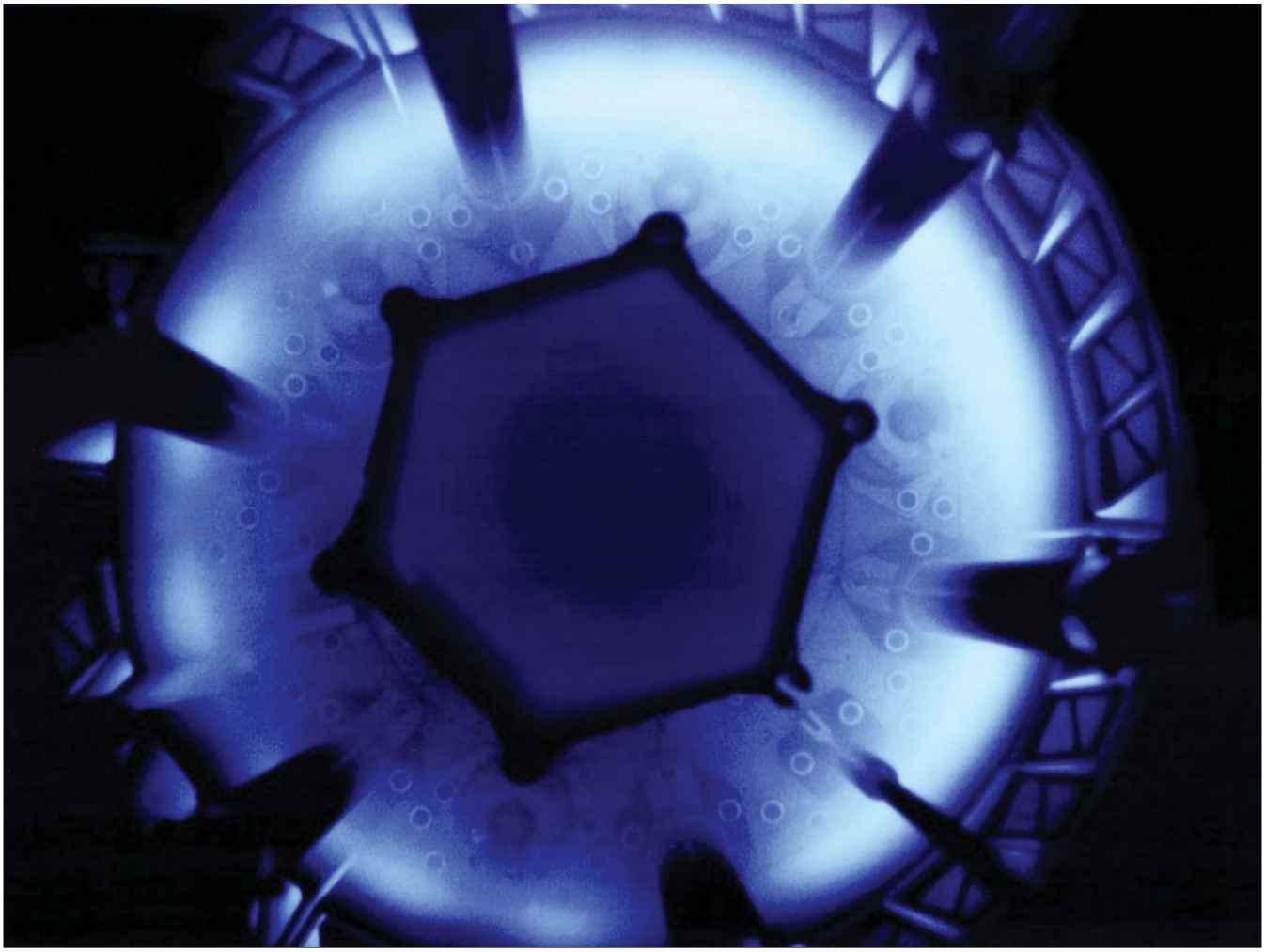
Irradiated Microstructure Analysis







- Have begun x-ray diffraction analysis of irradiated specimens
  - SiC has lower activity, good for initial measurements
  - Hastelloy N and stainless steel specimens planned
  - Probing changes in phases, swelling
- SEM/EDS starting soon
  - Identify microstructural features such as grain size, layer formation
  - Map depletion/infiltration of elements
  - Important for SiC/SiC and C/C fiber composites with matrix porosity, fiber-matrix interfaces
- Tritium thermal extraction
  - Have custom-designed furnace for tritium desorption from solids with re-capture for counting
  - Also capable of running out-of-pile tritium exposures at high temperature.



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Investigation of Tritium Control and Release Mitigation Options in Double-Wall Twisted-Tube Heat Exchangers (DT-HXRs)

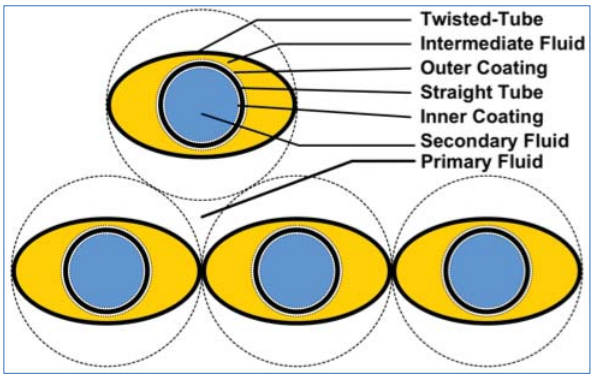
Bryan Wallace
Joel Hughes and Edward D. Blandford
University of New Mexico

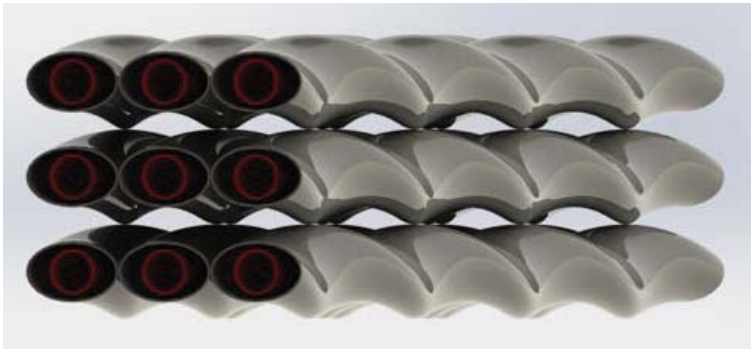
Tritium Workshop
October 2016
Salt Lake City, Utah

Double-Wall Twisted-Tube Heat Exchangers

Double-wall twisted-tube heat exchangers are being investigated as an option for the removal of heat from molten salt coolant.

- Double-wall tubing ideal for prevention of fluids mixing
- Twisted-tubes provide increased heat transfer between mediums





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Tritium Control in DT-HXRs

We will be investigating multiple methods for the control of tritium permeation throughout the DT-HXR system.

Intermediate Fluids	Surface Coatings
He	Carbides
He/O	Aluminides
Li	Titanium Ceramics
	Tungsten
	Yttrium (for Gettering)

Current Status of DOWTHERM Heat Trasnfer Loop

- Project start October 1, 2015
- Currently constructing experimental loop
- Low pressure heat exchangers purchased and fabricated
- Low pressure testing to begin in 2016

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Project Scope

- Scoping simulation and trade-off studies for intermediate fluids considering tritium recovery
- Low pressure testing with DT-HXR
- Work on optimizing high pressure heat exchanger design
- High pressure testing coupled to S-CO_2 loop at SNL
- Data reduction, validation, and inspection techniques

Questions?