Tritium Production Analysis and Management Strategies for a Fluoride-salt-cooled High-temperature Test Reactor (FHTR)

By

Judy N. Rodriguez

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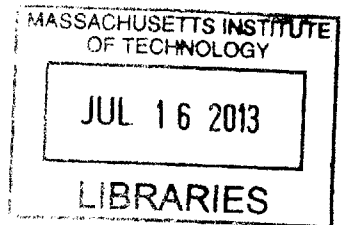
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ABSTRACT

The Fluoride-salt-cooled High-temperature Test Reactor (FHTR) is a test reactor concept that aims to demonstrate the neutronics, thermal-hydraulics, materials, tritium management, and to address other reactor operational and maintenance issues before a commercial Fluoride-salt-cooled High-temperature Reactor (FHR) can be deployed. The MIT Nuclear Systems Design class proposed a design for a 100 MW FHTR that uses enriched-^{7}Li flibe (Li_2BeF_4), has both thermal and fast flux testing positions for fuel and materials testing, and provides a neutron flux greater than 3E14 n/cm^2-s for accelerated irradiation testing. One of the key technical issues of the FHR and FHTR is tritium generation from the flibe coolant and its radiological control. The objectives of this study are: 1) to provide an overview of tritium production in various types of nuclear systems, 2) to estimate the tritium source term in the FHTR using the ORIGEN-S computer code, and 3) to propose a tritium management strategy for the FHTR. A review of existing nuclear systems shows that tritium is the primary radionuclide in liquid and gaseous tritium release. Light water reactors release up to several hundred curies per year for which various tritium removal and control strategies have been developed and implemented. Using the ORIGEN-S code analysis, tritium production for the MIT FHTR design at 20 MW is estimated to be about 2600 Ci per year (based on a 70% capacity factor and~10 Ci/day), with 99.99% enriched-^{7}Li flibe. Using this source term, a tritium removal rate of >90% is proposed as a design target for the tritium control system of the FHTR in order to maintain tritium release within the limits of existing nuclear reactors. Proposed tritium management strategies for the FHTR include increasing the ^{7}Li enrichment, carbon-based or metallic getters, and inert gas sparging with a high-temperature recombiner system.

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Table of Contents

1 Introduction

1.1 Overview of the Fluoride-salt-cooled High-temperature Reactor (FHR)

Research in the nuclear and materials engineering fields worldwide is currently investigating a new reactor concept called the Fluoride-salt-cooled High-temperature Reactor (FHR). The design features a low-pressure, high-temperature liquid fluoride-salt coolant, coated-particle fuel, increased efficiency for electricity generation, process heat production, and fully passive decay heat rejection. These qualities contribute to the generation of electricity at a low cost while maintaining passive safety features, overcoming current limitations of Light Water Reactors (LWRs) [1]. The concept of the FHR must undergo further design study, materials and fuel irradiation tests, and operational and safety evaluation before a full-scale power reactor can be built. The development of a Fluoride-salt-cooled High-temperature Test Reactor (FHTR) will allow for the testing of critical concepts in the FHR including the testing of materials, coolants, and the neutronics.

An interest in Molten Salt Reactors (MSRs) originated first in the 1950s and 1960s as highly experimental designs. The interest evolved around the potential ability of the MSR to burn actinides, operate as a breeding reactor for Thorium, provide a simplified fuel cycle, and make way for limited transport of radioactive materials. Advances in High-Temperature Gas-cooled Reactor (HTGR) fuel technology, materials, and MSR technology led to the development of the FHR concept. The MSR and FHR designs are mainly different in the fuel aspect: the fuel in MSRs was dissolved in the molten salt coolant whereas the fuel for the FHR is the same type of solid fuel developed for HTGRs.

The primary coolant candidate for the FHR is flibe (66.7%LiF-33.3%BeF$_2$) because it has excellent heat transfer and neutronics properties, with a high melting point of 460°C

and boiling point of 1430°C that allow reactor operation at very high temperatures [2]. As a result, flibe is also the primary coolant candidate for the FHTR. Tritium is a radioactive isotope of hydrogen with an atomic mass of three that may be produced from neutron capture reactions with the components of flibe, primary ^{6}Li. Tritium has a half-life of 12.3 years and is a pure beta emitter. At high temperatures, tritium behaves like hydrogen and may diffuse through materials and escape from the reactor.

1.2   Overview of the FHTR [3]

Before the FHR becomes commercially viable, a test reactor is necessary to study important aspects relevant to neutronics, thermal hydraulics, materials, and most relevant to this case, methods for managing tritium. A preliminary 100 MW FHTR has been designed by the 2012 MIT Nuclear Systems Design Class to prove commercial feasibility of and provide accelerated testing for the FHR. The MIT FHTR design has notable features that include: a large central thermal test position with a neutron flux greater than 3E14 n/cm^2-s for accelerated fuel and materials testing, numerous in/ex-core testing positions in contact with the primary coolant, a coated-particle-based fuel form, and high inlet (>600°C) and outlet (~700°C) temperatures to avoid salt freezing and provide the expected high-temperature environment of the commercial design. Numerous safety features have been suggested in the FHTR design for tritium control and decay heat removal. Figure 1 shows the core geometry layout of the MIT FHTR design and Table 1 shows the core's relevant dimensions [4]. Table 2 shows the flux levels in several parts of the FHTR.

Coolant
Graphite
TRISO Fuel
Hafnium
Outer Test Position
Inner Test Position
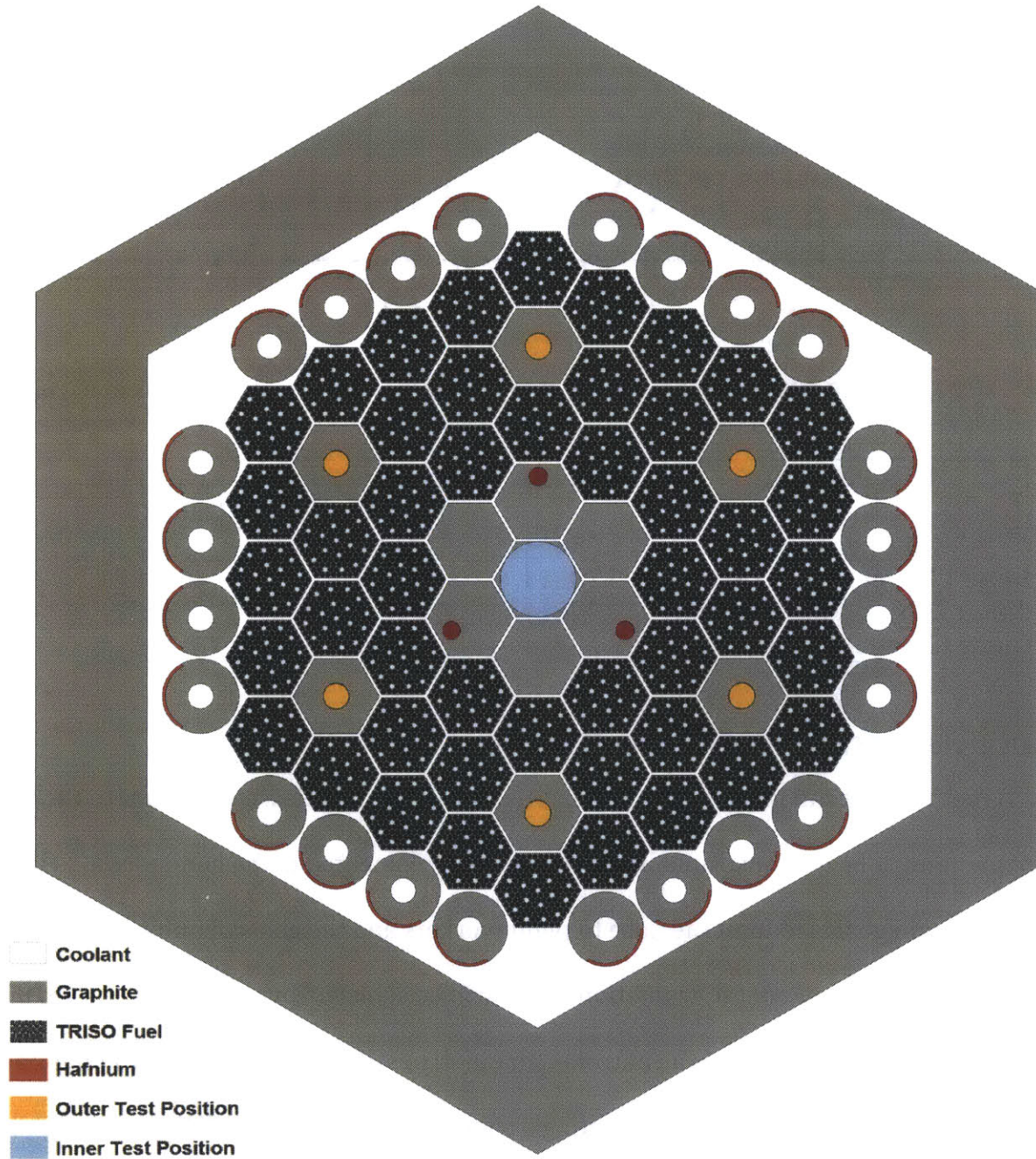
Figure 1 MIT FHTR core geometry layout [4]

Table 1 MIT FHTR core geometry parameters [4]

Dimension	Value (cm)
Short Radius of the Core	100
Core Height	120
Radial Reflector Thickness	40
Axial Reflector Thickness	30
Short Radius of Assembly	12
Assembly Pitch	25
Fuel and Coolant Channel Radius	0.8
Fuel and Coolant Channel Pitch	1.8
Inner Test Position Radius	12
Outer Test Position Radius	4

Table 2 MIT FHTR 100 MW flux values of the average core and core test positions in n/cm²-s [4]

Flux Position	Total Flux	Thermal Flux (<0.625 eV)	Fast Flux (>0.625 eV)
Average Core	2.85E14	2.28E13	2.62E14
Inner Channel Test Position	5.28E14	3.25E14	2.03E14
Inner Center 1000 cm³ Box	6.90E14	4.53E14	2.37E14
Outer Channel Test Position	2.79E14	2.00E13	2.59E14

1.3 Overview of Tritium Production in the FHTR

A method that can monitor the production, removal, and release of tritium to the environment must be demonstrated before it can be implemented in the commercial FHR design. The main goal is to limit the dose to plant workers and the general public as specified by the Code of Federal Regulations. This will be achieved by containing tritium in the reactor system, removing it in a controlled manner, possibly through a getter and/or a cover gas/recombiner system, and preventing a significant amount from diffusing through reactor materials by employing barriers in heat exchangers and other sensitive areas, or other methods.

Tritium in the flibe coolant is formed by neutron reactions with its constituents: [19]F, [6]Li, [7]Li, and [9]Be. The most prominent is the neutron reaction with [6]Li:

$$^6_3Li + n \rightarrow He + \, ^3_1H$$
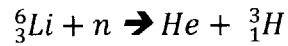
which has a cross-section of 940 b for thermal neutrons at 0.025eV [13]. The tritium ions formed in these reactions can combine with ^{19}F to form the corrosive TF, becoming a target of interest for removal from the coolant. When H_2 is introduced into the salt at high enough pressure, an isotopic exchange with TF can be induced to produce HT gas [5]. This HT gas is an attractive target for tritium removal systems, and can be involved in a proposed recombiner system.

In order to accurately determine the tritium production rate in the FHTR, variables such as neutron flux, coolant temperature, and core geometry must be known. An ORIGEN-S analysis is employed to estimate the tritium production rate for up to five years without removal. After determining the tritium production rate, retention and removal strategies can be proposed. Simulation of tritium transport is out of the scope of this study, but should be studied further to accurately determine the best management method and the potential for release in accident scenarios.

1.4   Overview of Tritium Production in Other Nuclear Reactor Systems

Tritium production and its control have also posed an issue for other nuclear systems mainly for radiological controls. There has been significant research in the MSRE project (1950-60s) in which flibe was used as the primary and secondary coolant, and studies of tritium breeding in flibe as fuel for fusion applications [6]. Currently, CANada Deuterium Uranium (CANDU) reactors, Light Water Reactors (LWRs), and some research

and test reactors employ tritium control methods. The tritium production in a PWR, CANDU, the MSRE, two FHR designs, and a fusion design are listed in Table 3[1]. It is important to note that the tritium production is highly based on the ^7Li enrichment in the molten salt; the higher the ^7Li enrichment, the less ^6Li available for the neutron capture reaction that contributes the most to tritium production[2].

Table 3 Tritium production in a PWR, CANDU, the MSRE, and two FHR designs.

Reactor	Reactor Power	Tritium Production (Ci/day)	^7Li enrichment
PWR [7]	1000 MWe	1.9	n/a
CANDU [8]	1000 Mwe	2740	n/a
MSRE [8]	1000 Mwe	2420	99.99%
FHR [9]	2400 MWt	5000 (initial) 500 (equilibrium)	99.995%
FHR Baseline from UC-Berkeley Design [8]	900 MWt	32.8 (equilibrium)	99.995%
Fusion Designs [8]	1000 MWe	4.2E6 [3]	92.5%

While many technologies have been developed to control tritium release into the environment in different reactor designs and fusion applications, there is a need to study the feasibility of these technologies for the FHTR design based on estimates of total tritium production and its behavior with the FHTR coolant and structural materials. This project will address this issue to determine the best tritium management strategies for the FHTR by estimating the MIT FHTR's long-term tritium production without removal, and assessing the feasibility of current tritium management in other reactors and nuclear applications.

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[1] Initial and equilibrium values exist due to the initial high production and then eventual burnout of ^6Li in the flibe salt.
[2] Natural abundance of ^7Li is 92.4%
[3] Order of magnitude is much greater since tritium is purposely bred for fuel.

1.5 Thesis Objectives

The objectives of this thesis are as follows:

1) To provide an overview of tritium radiological control and management strategies of the MIT reactor and other existing nuclear systems

2) To estimate the total tritium production expected in a reference FHTR using the ORIGEN-S computer code, and

3) To propose a tritium management strategy for the FHTR

2 Tritium Control and Management in Existing Nuclear Reactor Designs

2.1 Tritium at the MIT Reactor (MITR)

2.1.1 Overview of the MITR

The MIT Nuclear Reactor Laboratory (MIT-NRL) is a leading university laboratory that conducts interdisciplinary research. MIT-NRL operates a 6 MW light-water-cooled and moderated, heavy-water-reflected reactor, the MIT Research Reactor (MITR). It is the second largest university research reactor in the United States. The MITR utilizes flat, plate-type finned, aluminum-clad fuel elements. The fuel is 93%-enriched U-235. Its mission is to provide faculty and students from MIT and other institutions with both a state-of-the-art neutron source and infrastructure required to facilitate use of the reactor [10].

Because it uses heavy water as a reflector and it has a very high neutron flux rate, 6E13 n/cm²-s, the MITR has strict limits on the level of tritium in both the heavy water reflector and secondary (light water) cooling systems, as outlined in the MITR Technical Specifications [11]. Tritium is formed whenever a deuterium nucleus captures a neutron, as illustrated in the following reaction:

$$\,^2_1H + n \rightarrow\,^3_1H$$
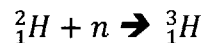
Although the absorption cross-section for thermal neutrons of deuterium is small (0.508 mb) [12] and relatively little tritium is produced, replacing the heavy water after several years of tritium buildup is desirable to reduce the risk of its escaping to the environment and causing radiological protection concerns to reactor workers and to the general public. The heavy water system is closed and hence the tritium that is contained does not pose a hazard during normal operation. Still, activity levels should be minimized

to reduce the radiological controls necessary to perform maintenance activities that require opening the system.

The secondary system, which cools the heavy water reflector, is also monitored in case of leakage through the intermediate heat exchangers. Although the secondary system is continuously monitored for activity by on-line detectors, these detectors do not sense tritium because it emits a very low energy beta particle (18.6 keV) [13]. A daily sample of secondary water is taken to analyze it for tritium anytime secondary coolant is supplied to a heavy water heat exchanger [14].

Stack effluents are also monitored to preclude the discharge of radioactivity. A radiation monitor that samples the stack effluent is operating at all times. The tritium concentration and radioactivity are monitored to measure the amount discharged to the environment. Tritium releases are rep[11]orted annually to the Nuclear Regulatory Commission (NRC).

2.1.2   MITR Tritium Production and Limits

2.1.2.1   Technical Specifications

Technical specifications of the MITR require that the heavy water reflector coolant and the secondary coolant be analyzed for tritium. The tritium radioactivity shall not exceed 5 Ci/liter in the heavy water system. If the radioactivity of the tritium in the heavy water approaches this guideline limit, preparations are initiated to replace the heavy water. Normal operation may continue while the preparations are being made, but the tritium radioactivity should never exceed 6 Ci/liter. On indication of $\geq$ 1μCi/liter of tritium in the secondary coolant, the cooling tower spray must be stopped, and the heavy water

heat exchangers shall be isolated until tritium leakage into the secondary coolant has been controlled.

2.1.2.2   Offsite Dose Limits

The Environmental Protection Agency (EPA) controls the regulations for residential areas while the Nuclear Regulatory Commission (NRC) controls the regulations of nuclear reactor facilities. No more than 10 mrem/yr from the reactor effluent release should reach the general public. While it is possible to exceed this limit, it is advised that the levels be maintained below this limit posed by the MIT Radiation Protection Office (RPO). The NRC limit is 100 mrem wherever the highest dose is monitored, which for the MITR is at the tip of the stack. A dilution factor of 50,000 is applied to the activity limit (5E-3 μCi/mL) to produce an effective concentration limit for stack effluents [15].

Part 20, Code of Federal Regulations (10CFR20) governs this limit for radioactive effluents released to the environment. The air and water effluent limits for tritium in 10CFR20 are 1E-7 μCi/mL and 1E-3 μCi/mL, respectively [36]. Although the heavy water system is intended to be a closed system, there is still unavoidable leakage through gaskets and seals in the system (pumps, penetrations) that are taken into account in the stack effluent releases.

Table 4 shows the values of the tritium activity released at the stack for the last 16 years [15]. In the past three years, the tritium release from the reactor through the stack averaged about 35.6 Ci/year, although it was as high as 180.6 Ci during which there were leakages in the system.

Table 4 Yearly stack tritium effluents for the MITR from 1996 to 2012.

Year	Tritium Activity in the Stack (Ci)
1996	9.8
1997	26.6
1998	36.7
1999	52.4
2000	71.2
2001	132.2
2002	62.4
2003	84
2004	180.6
2005	54.4
2006	39.7
2007	12.2
2008	10.2
2009	14.2
2010	35.3
2011	34.6
2012	37.0

2.1.3   MITR Tritium Management

2.1.3.1   Recombiner System

The main heavy water reflector tank contains a helium cover gas blanket that drives the recombiner system. The helium cover gas is cycled through a recombiner to control the concentration of deuterium in the system. The helium is circulated by penetrations through the core tank bottom section flange. A gasholder tank that is maintained at constant pressure (1" H_2O above atm) serves as a surge and storage tank for the system. Figure 2 shows an excerpt from the system. The recombiner has the input of helium gas containing the deuterium and oxygen that recombine, reforming into heavy water and dropping by gravity through pipes into the heavy water system storage tank.
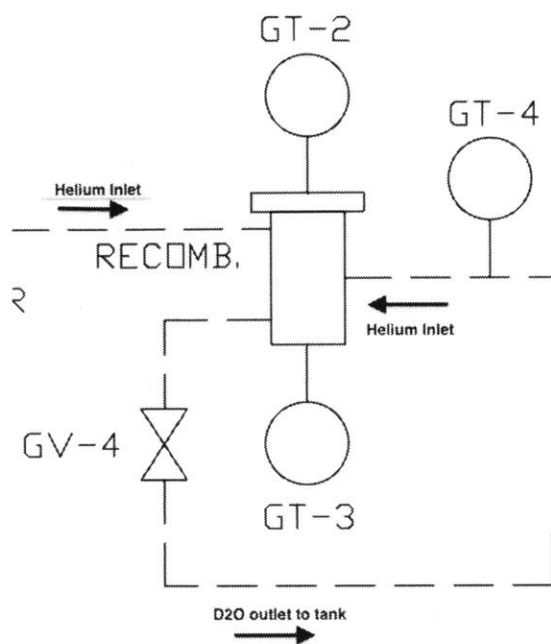
Figure 2 Recombiner system for the MITR. The recombiner has helium inlets containing the D_2 and O_2 that recombines with the help of a platinum catalyst. The GTs are thermocouples that monitor the temperatures at all times. The recombined D_2O returns to the main heavy water tank by gravity.

One of the main purposes of the recombiner system is to provide an inert, non-radioactive vehicle to circulate disassociated D_2 and O_2 from the reflector tank to a catalytic recombiner. The recombiner uses a platinum catalyst that operates around 80°C during normal reactor operations at 5.8 MW, and recombines D_2 and O_2 to reform D_2O. Once D_2O is reformed, it returns to the storage tank by gravity. This controls the concentration of deuterium in the system to prevent a flammable amount in the cover gas. It also helps in reducing the amount of tritium in the cover gas. The two other purposes of the recombiner system are to prevent air entrained with H_2O moisture from entering the system and degrading the D_2O and to prevent corrosion caused by nitrous-oxide formation from air in

the presence of high radiation fields [14]. Table 5 shows some operating specifications for the recombiner system.

Table 5 Operating Specifications for the MITR recombiner system [14]

Parameter	Value
Cover Gas Flow Rate	19.5 gpm
Catalyst	Platinum pellets
Recombiner Inlet Temperature	31.5°C
Recombiner Outlet Temperature	50°C
Recombiner Middle Temperature	80°C

2.1.3.2   Heavy Water Replacement

The Department of Energy (DOE) owns the heavy water that is used in the MITR. Whenever the tritium specific activity approaches the 5Ci/liter limit, the heavy water must be replaced to reduce the potential radiological hazard in the event there is a heavy water spill. High purity, fresh heavy water is provided by the DOE, and the tritiated heavy water is returned to the DOE as per a loan agreement. There are about 500 gallons of heavy water in the system at all times. Of the 500 gallons, about 170 are in the reflector system piping, 70 in the storage and dump tanks, and 260 in the reflector tank and medical shutter [16]. Once the shipment arrives, the tritiated heavy water is drained into stainless steel barrels, and replaced with the fresh heavy water. Shipment of heavy water is carefully monitored, especially during cold weather when the heavy water can freeze (D_2O's freezing point is higher than H_2O's) [17].

2.1.3.3   Radiological Control of Heavy Water System Maintenance Activities

The MITR RPO enforces guidelines whenever the heavy water system must be opened to perform maintenance activities. Precautions include: minimizing release of heavy water from the system, using proper ventilation, and wearing appropriate protective

clothing such as gloves and labcoats. A RPO officer must be present to monitor for tritium whenever any amount of heavy water will be removed from the system. Before any significant work on the system is to be done, those working have to have a urine analysis prior to and after the work as a means of ensuring that no tritium was ingested and absorbed into the body. Additional measures include setting up a portable air monitor; a bubbler that traps the heavy water that can be analyzed for activity afterwards.

Small amounts of tritiated heavy water that cannot be returned to the system due to potential contamination are evaporated through the stack as opposed to drained to the sewer. In this way, it becomes less of a consequence to the general public since the effluent releases are less limited than the sewer releases.

2.1.3.4 Stack Release

Before effluents exit the stack, the air is sampled for tritium through a fritted glass bubbler. After bubbling a known volume of air through a known volume of collecting water, a sample of the water is counted in a liquid scintillation counter to determine the tritium concentration in the sampled air [18]. The measurements using this method are shown in Table 4 above. The gas bubblers collect 98% of the tritium activity [15].

2.1.3.5 Sewage Release

Waste water is also sampled and monitored for radioactivity prior to being discharged to the sewage. Prior to discharge into the sewage, the water in the waste tanks is recirculated for at least six hours in order to mix the water, producing a representative sample of the activity present. A sample is then taken and analyzed to ensure that the total activity discharged will be less than the tritium 10 CFR 20 limit of 5 Ci/year. Once

approved, the water is continuously monitored as it is dumped into the sewage using a liquid scintillation detector [14].

From the operation experience at the MITR, it is shown that the tritium effluent release can be much higher (more than two orders of magnitude) than the sewage release. The effluent release is limited by the radiation dose to the general public, which depends on siting and other radionuclides released through the stack, while the tritium sewage release for the MITR is strictly limited at 5 Ci/year.

2.2 Tritium in Fusion Systems

Tritium is used as fuel in fusion reactors. Fusion applications of flibe have warranted extensive study of tritium behavior and control in molten salt blanket systems. Flibe is the material of interest because of its inherent advantages such as high temperature stability, low electrical conductivity, and lithium's high tritium breeding ratio. However, flibe presents similar issues as in fission systems: TF impurities in the salt produced from nuclear reactions with Li that can be very corrosive to structural materials, tritium permeability through materials, and safe handling practices and releases from flibe during an accidental spill. Different methods have been proposed to recover tritium from a fusion blanket system including: molten salt extraction, permeation windows, gettering, fractional distillation, and so on. Different fusion programs are presented here with their proposed tritium management strategies.

2.2.1 HYLIFE-II [19]

HYLIFE-II is a conceptual design for a molten-salt inertial fusion energy power plant designed in 1993. The design uses a neutronically thick array of flowing flibe jets, which does not burn, has a low tritium solubility and inventory, and protects the chamber walls,

giving a robust design with a 30-year lifetime. Issues have risen in relation to irradiation

behavior and chemical kinetics of flibe, as well as materials compatibility for the

containment vessel and primary loop, and most importantly, tritium-related issues. The

HYLIFE-II design tritium production rate is 3.7E6 Ci/day. Suggestions for the HYLIFE-II

tritium control has included holding tritium as LiT in molten salt by adding lithium or

beryllium into the salt [20]:

$$Li_2BeF_4 + Be \rightarrow 2BeF_2 + 2Li$$

$$Li + \frac{1}{2}TF \rightarrow \frac{1}{2}LiT + \frac{1}{2}LiF$$
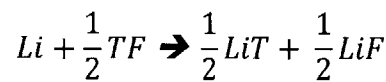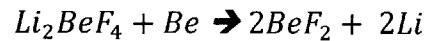
However, it has been pointed out that LiT will react with TF, such that LiT will not remain

in the salt [21]. Other suggestions to control tritium permeation involve using an

intermediate coolant for decreasing leakage into the secondary coolant and to add getter

materials to the salt [22].

2.2.2   Integrated Pool Fusion Reactor Concept (IPFR) [23]

The Integrated Pool Fusion Reactor Concept (IPFR) is a concept that was presented

by Argonne National Laboratory (ANL) suggesting to place a fusion reactor into a pool of

molten flibe. The main goal was to have flibe serve the multiple functions of breeding,

cooling, shielding, and moderating. The design expected to improve safety, reliability, and

maintainability aspects of the fusion system. The two most critical issues with the design

were tritium control and pool-wall cooling, and the design proposed a possible solution for

tritium and corrosion control.

The study suggests the formation of a molybdenum coating on the surfaces of

structural components, achieved by dissolving MoF_6 in the flibe. Assuming a V-alloy for the

first wall that comes in contact with the flibe, V, Ni, and tritium will react with MoF_6 to form

VF_4, NiF_2, and TF respectively. In the reaction with V or Ni, the Mo is precipitated out and

forms a continuous coating on the wall to prevent further reaction. The formation of TF will

greatly reduce the mobility of tritium and prevent it from permeation. The total tritium

inventory in the reference design is six grams, and the tritium leakage rate is expected to be

about 4 Ci/day.

2.2.3   JUPITER-II Program [24]

The Japan/US Program on Irradiation Tests for Fusion Research (JUPITER-II) in the

Idaho National Laboratory in the Safety and Tritium Applied Research (STAR) facility is the

second of its kind and focuses on studies of flibe for fusion applications. In particular, the

program discusses corrosion control and its relation to tritium.

Due to the inevitable combination of tritium with fluorine to form TF, the

reduction/oxidation reactions will determine the overall tritium behavior of a system.

Beryllium is suggested as a redox agent to control the free F in the system because it is also

used in fusion for neutron multiplication to ensure adequate tritium breeding. Experiments

were performed in which HF was bubbled through flibe with varying concentrations of

dissolved Be to investigate the viability of using Be as a redox agent in a molten flibe

blanket. If successful, it is expected that tritium will be present in the flibe fusion blanket

primarily as $T_2(g)$.

The JUPITER-II program has also studied flibe mobilization to determine influence

of different environments, i.e. dry air, moist air, steam, possibly encountered during an

accident. Accident scenario concerns involve handling of Be and knowledge about the

behavior of aerosols and vapors produced from a spill of irradiated flibe when it interacts with air or water. Reaction with moist air and water may produce HF, which could mobilize tritium, ^{18}F, and other radionuclides during irradiation.

In an effort to control the formation of TF in the blanket system and maintain it below a level that would lead to structural materials corrosion, hydrogen can be purged in through a gas stream in the space over and through the salt. It has been demonstrated that this promotes changes in the tritium chemical species to HT via exchange reactions and enhances release of tritium from the salt to the gas phase above the salt. Gas sparging is therefore a promising option that many other systems besides fusion systems have suggested.

2.3 Tritium in the Pressurized Water Reactor (PWR)

2.3.1 PWR Tritium Production [25]

The study of tritium in PWRs is important due its long-term buildup within the plant systems. Since the reactor coolant is recycled, tritium is retained within the plant as tritiated water and release may occur as liquid, water vapor or gaseous tritium. The primary sources of tritium in the reactor coolant system in a PWR are: diffusion of tritium from the fuel through the zircalloy cladding, neutron activation of boron in the burnable poison rods and subsequent tritium diffusion through the cladding, and neutron activation of boron, deuterium and ^{6}Li in the reactor coolant. The fission yield of tritium for U-238 is ~0.01%.

The major neutron reaction with boron resulting in tritium production is:

$$^{10}_{5}B + n \rightarrow 2\,^{4}_{2}He + \,^{3}_{1}H$$
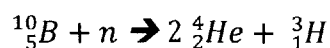
Another reaction that results in the production of tritium is the ^{6}Li $(n,\alpha)^3$H reaction in the coolant. This reaction is controlled by limiting the ^{6}Li impurity in the ^{7}LiOH used in the reactor coolant that controls the pH of the coolant, and in the lithium from demineralizers with 99.9%^{7}Li. The tritium produced by the neutron activation of deuterium in water (deuterium concentration makes up about ~0.015% of the coolant) is less than 5 Ci/year, a few percent of the total production rate. The tritium inventories in typical PWR plant components are given in Table 6. The rate of tritium production in a 1000 MW$_e$ PWR is 1.9 Ci/day [26].

Table 6 Tritium Inventories in Plant Components in a PWR

Component	Water Volume (g)	Tritium Concentration (μCi/g)	Tritium Inventory (Ci)
Reactor Coolant	1.17E8	0.416	48.7
CVCS Holdup Tank	9.5E7	0.315	33.3
Component Cooling System	1.1E7	0.240	2.6
Refueling Water Storage Tank	9.6E8	0.11	106.0
Spent Fuel Pit	2.26E9	0.09	204
Boric Acid Storage Tanks	7.56E6	0.184	1.4
Waste Holdup Tanks	1.09E7	0.132	1.4

2.3.2   PWR Tritium Management [27]

According to a 2008 report on radioactive effluents from nuclear power reactors, tritium exists in liquid effluents along with ^{55}Fe, ^{58}Co, ^{60}Co, ^{134}Cs, ^{137}Cs, ^{131}I. For PWRs, tritium liquid release ranges from about 160 Ci to 1670 Ci, with a median value of about 530 Ci. (BWRs tritium release ranges from 1E-3 to 127 Ci with median release of about 2.5 Ci.) Other radionuclides total about 0.1 Ci in liquid release, which is much lower than the tritium activity. Tritium gaseous release from PWRs ranges from 2.6 to 870 Ci with a median release of about 30 Ci.

The biggest concern with tritium in PWRs is that most of it is built up in liquid form as tritiated water and its release as a liquid effluent is more limiting than its release as a gas effluent due to NRC regulations. The most economical and relatively efficient way of restricting the negative impact of liquid tritium releases is to convert the liquid tritium to gas form. One possibility for achieving this is by evaporating tritiated water through the gas effluent to the atmosphere, a method employed in the MITR for small amounts of tritiated heavy water. Separation of tritium from the reactor coolant may also be looked at for possible ways of purposely breeding tritium for use in detection or weapons.

2.4 Tritium in CANDU Reactors

2.4.1 Overview of Tritium in CANDU Reactors

CANada Deuterium Uranium (CANDU) reactors are of particular interest because they use heavy water as their primary coolant and moderator with significant tritium production. Tritium is produced mainly by neutron activation of deuterium, as in the MITR. A typical production rate for 1000 MW_e CANDU reactor is 2740 Ci/day, and mostly in the form of tritiated heavy water [29][8]. A typical average concentration of tritium in the moderators has reached 16 Ci/kg and in the coolants 0.5 Ci/kg [28]. The large production rate and specific activity require extensive tritium management controls.

As a result from the need to establish technologies that control large amounts of tritium, Canada has built laboratories with significant research and development capabilities, specifically at the Kinetrics and Chalk River Laboratories. The Chalk River Tritium Facility can handle up to 1E6 Ci for dispensing operations and research and development activities.

Detritiation research has seen significant activity with the successful design, construction, and commissioning of the Wolsong Tritium Removal Facility (WTRF) in Korea. The WTRF uses a liquid phase catalytic exchange process. Atomic Energy of Canada Limited (AECL) has also successfully demonstrated the Combined Electrolysis and Catalytic Exchange (CECE) process for heavy water upgrading and detritiation in semi-industrial scale [28].

A CANDU reactor without detritiation will reach a steady-state tritium concentration of 84 and 2 Ci/kg in the moderator and the heat transport system (HTS), respectively, after about 60 years of operation [28]. The three main ways to avoid environmental emissions are by: detritiation to minimize tritium in heavy water, control of heavy water leaks, and recovery of any heavy water that leaks from the system. CANDU reactors currently employ detritiation measures as the primary way of preventing large build up of tritium in their systems.

2.4.2   Detritiation Processes

Although detritiation technologies have been implemented in the power reactor facilities themselves, tritium removal itself has the potential to emit both elemental tritium and tritiated water vapor during operation. The Darlington Nuclear Generating Stations (DNGS) have their own detritiation facility, DTRF, which is the largest in the world and is responsible for 30-35% of the total tritiated heavy water emissions from the DNGS [29].

The WTRF and DTRF facilities provide moderator detritiation to maintain a steady-state tritium level less than 10 Ci/kg D_2O. The DTRF employs a gas phase catalytic exchange process to transfer tritium from completely-vaporized D_2O to gaseous deuterium while the
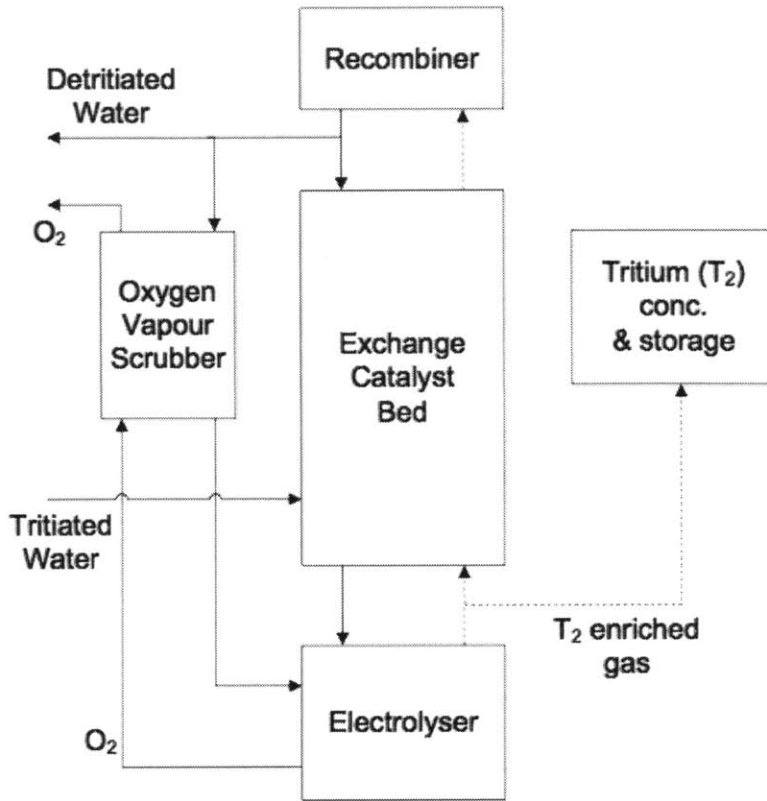
**Figure 3 Simplified CECE process schematic diagram
developed by AECL.**

WTRF uses a Liquid Phase Catalytic Exchange (LPCE) process in which the deuterium gas is

humidified/equilibrated in separate mass-transfer sections and then passed through

catalyst sections for the actual transfer of tritium from liquid D_2O to deuterium gas [29].

Cryogenic distillation is used in both facilities for final separation of elemental tritium from

deuterium for final storage of tritium in the form of gas. General parameters for the WTRF

are listed in Table 7.

Table 7 General parameters for the WTRF design. [35]

Parameter	Value
D_2O feed isotopic	$\geq$ 99.8 mol% D_2O
D_2O feed tritium concentration	10-60 Ci/kg
D_2O processing rate	100 kg/h
Minimum tritium extraction efficiency per pass	97%
Tritium by-product	$\geq$ 99.0 mol% T_2
Service life	40 years

AECL has been studying the CECE process as simple way of tritium removal while simultaneously concentrating tritium. Figure 3 demonstrates a simplified configuration for the CECE process. The CECE process uses an isotope exchange and a trickle-bed recombiner catalyst for the exchange of isotopes and recombination of deuterium to D_2O and an industrial electrolysis cell for splitting the heavy water into deuterium and oxygen.

Preventing release to the environment involves a multiple-barrier approach that includes: careful consideration for system design pressure and temperature to limit leakage, minimizing tritium inventory in the system to limit emission in case of leakage, use of special code class equipment and/or secondary confinements where needed to improve reliability and strength of barriers to leakage, incorporating chillers and absorbers to condense and/or absorb tritiated water from vented streams, to capture chronic leakage and emissions, and the use of an Air Cleanup System (ACS) to recover leaked and spilt tritium. Both the WTRF and DTRF facilities employ these methods.

3 ORIGEN-S Analysis of the Tritium Source Term in the FHTR

3.1 Tritium Production Estimate

The MIT FHTR was designed to operate at 100 MW, with a neutron flux roughly five times higher than the commercial FHR design in order to provide for accelerated testing. Due to costs and expected licensing restrictions, it is assumed in these calculations that the reactor will operate at a power of 20 MW. A simple calculation of the tritium production rate in Ci/day for the MIT FHTR can be made assuming the primary reaction producing tritium is the thermal neutron capture of ^{6}Li. The following equations are used to calculate the tritium production rate:

$$Target \; {}_{3}^{6}Li \; atoms \; = \; n = \frac{N_A m_{Li}}{M_{Li}}$$

$$Production \; Rate \; (\frac{atoms}{s}) \; = \; R_T = \; \varphi \sigma n$$

$$Activity \; Rate \; \left(\frac{Ci}{day}\right) = \frac{R_T \lambda_T (86400 \frac{s}{day})}{3.7E10 \frac{Bq}{Ci}}$$

where N_A is Avogadro's number, m_{Li} is the mass of ^{6}Li in the core, M_{Li} is the molar mass of ^{6}Li, φ is the thermal flux, σ is the thermal capture cross-section for ^{6}Li, and λ_T is the tritium decay constant. The values used for these variables are listed in Table 8. Note that the thermal neutron flux is the MIT FHTR core average neutron flux at 20 MW, and the mass of ^{6}Li in the core was calculated using the geometric values detailed in Table 1. The mass fraction of ^{6}Li assumes 99.99% enriched ^{7}Li flibe for the FHTR whereas other FHR designs consider 99.995% enriched ^{7}Li flibe, which limits the tritium production further.

Table 8 Values for the variables used to estimate the MIT FHTR tritium production rate

Variable	Value
N_A	6.022E23 atoms/mol
m_{Li}	8.85 g
M_{Li}	6.015 g/mol
φ	4.56E12 n/cm²-s
σ	940E-24 cm²
λ_T	1.786E-9 /s

The following assumptions were made for this calculation:

1) The amount of ^{6}Li target atoms remains constant and stagnant in the core for the total irradiation period

2) Tritium decay does not affect its production rate (half-life 12.3 years)

3) The total flibe coolant volume seen by the core is 4.56E5 cm³ and the ^{6}Li mass fraction in flibe is 1E-5 [30]

4) The thermal neutron capture cross-section is single-energy for 290 K neutrons

The resulting estimate for tritium production in the FHTR is 15.8 Ci/day. This production rate can be used to verify the calculations made in the ORIGEN-S analysis.

3.2   ORIGEN-S Analysis

3.2.1   ORIGEN-S Description [31]

The ORIGEN-S isotopic depletion and decay analysis system was used to determine tritium production in the FHTR. ORIGEN-S is part of the suite of SCALE programs developed and supported by the Oak Ridge National Laboratory. ORIGEN-S uses the matrix exponential method to solve the rate equations that describe nuclide generation, depletion, and decay processes. It contains cross-section libraries for several reactor types that include LWRs, MSBRs, and HTGRs. It has the capability of utilizing problem-dependent multi-group cross-section data for a burnup simulation process that uses reactor operating

conditions specified by the user. The calculation described here used the ORIGEN-S program associated with SCALE version 5.1.

Three neutron energy groups were used in the ORIGEN-S calculations: a thermal group below 0.625 eV, a resonance (epithermal) energy group extending up to 1 MeV, and a fast energy group above 1MeV. The user specifies the cross-section weighting factors THERM, RES, and FAST. THERM is used to adjust the 2200-m/s cross-sections in the library for a thermal neutron spectrum system, and can be determined using the moderator temperature. RES and FAST are used to weight the resonance and fast group cross-sections, with respect to the thermal group cross-section, in forming effective one-group values.

A thermal flux is provided along with time intervals to indicate the irradiation periods. The user can specify to use an elemental library, which applies natural isotopic abundances for the calculations, or an isotope-specific library. The program can analyze multiple isotopes and/or elements during the same irradiation period, taking into account secondary reactions. The output provides the mass of isotopes that are initially loaded, or are produced in the core. It is also possible to determine the activity in Curies and to model the decay of this activity after any specified time interval of irradiation.

3.2.2   FHTR Problem

The FHTR irradiation problem is solved using initial nuclide concentrations for each of the isotopes in 1000 grams of flibe. Table 9 shows the mass fractions and densities for each of the constituents of 99.99% [7]Li-enriched flibe. The results can then be extrapolated for the reactor inventory and adjusted for the amount seen by the core (8.85E5 g flibe which includes the flibe through the fuel flow channels and the gaps between the prismatic

block fuel elements, refer to Figure 1). The specific THERM, RES, and FAST ratios are input, along with the thermal flux in the core, 4.56E12 n/cm^2-s. The THERM term uses the average operating temperature expected in the core, 956.15K (683°C) [30]. The epithermal flux is assumed to be five times higher than the thermal flux, and the fast flux is adjusted such that the total flux remains the same. Table 10 shows the 20 MW flux values that were extrapolated from the 100 MW values of the MIT FHTR in the Romatoski et al. paper, and the resulting THERM, RES, and FAST factors [4].

Table 9 Mass fractions and densities of the constituents of 99.99% ^{7}Li-enriched flibe [30]

Isotope in flibe	Mass Fraction	Density (g/cm^3)
^{6}Li	1E-5	1.94E-5
^{7}Li	1.417E-1	2.74E-2
^{9}Be	9.1E-2	1.77E-1
^{19}F	7.673E-1	1.49

Table 10 MIT FHTR flux values adjusted for 20 MW and resulting THERM, RES, and FLUX parameter values input into ORIGEN-S

Parameter	Value
Thermal Flux	4.56E12 n/cm^2-s
Epithermal Flux	2.28E13 n/cm^2-s
Fast Flux	2.96E13 n/cm^2-s
THERM (683 C)	0.489
RES	5
FAST	6.5

The contribution from each flux energy can be determined by setting the RES and FAST terms to zero to determine the thermal flux contribution, and then setting FAST to zero to determine the epithermal flux contribution as the difference between the result and the thermal flux contribution. Similarly, the fast flux contribution can be determined by the difference between the total production rate and production rate from the thermal and epithermal fluxes. The irradiation is repeated over nine time intervals of 30, 60, 90, 180,

360, 700, 1080, 1500, and 1800 days to ensure that the behavior is accurately modeled. The ORIGEN-S input and outputs are shown in Appendix A and B, respectively.

3.2.3   Analysis of Tritium Production in the FHTR

The ORIGEN-S analysis results are illustrated in Figure 4 and detailed in Table 11. The graph shows that if the tritium is allowed to buildup over a long period of time without removal, it will increase approximately linearly with time. The average tritium activity based on the ORIGEN-S results is 10.2 Ci/day. This activity is less than the production estimate in Section 3.1 because of the shift in thermal cross-sections due to the higher operating temperature of the FHTR. It is important to note that the core's spectrum is largely fast, resulting in a slight increase of the tritium production as the production due to the fast flux increases over time. The increase in tritium production from the fast spectrum may be due to the increasing amounts of ^{16}O and ^{19}F in the system; these nuclides have relatively large fast-capture cross-sections [32]. These results show that tritium production is sufficiently high to warrant serious retention and removal strategies.
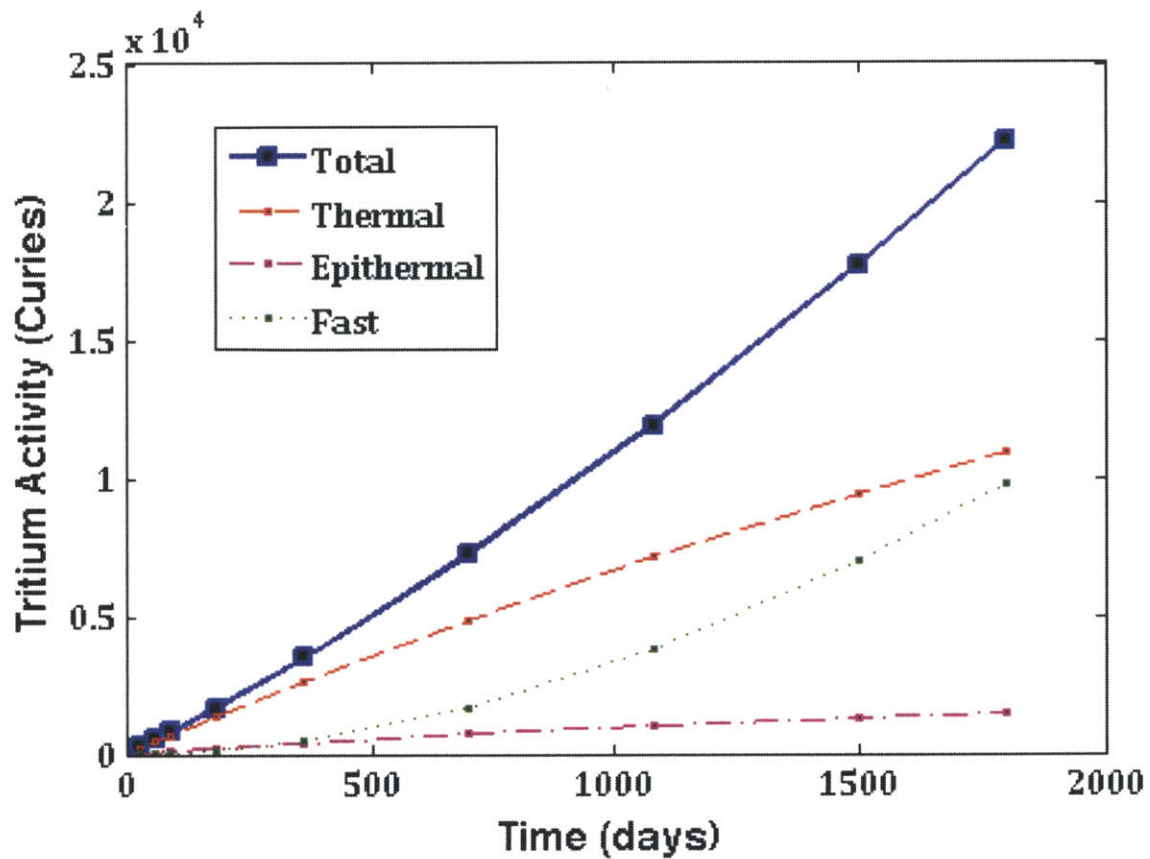
Figure 4 ORIGEN-S result. The total tritium activity and the contributions to the total activity from the thermal, epithermal, and fast flux

Table 11 ORIGEN-S result. The tritium activity (Ci) for the thermal, epithermal, fast, and total fluxes after specified specified time intervals measured in days

	30	60	90	180	360	700	1080	1500	1800
Thermal	232	463	688	1356	2637	4883	7165	9461	10965
Epithermal	37	72	108	211	402	724	1030	1310	1496
Fast	4	14	31	118	455	1645	3765	6974	9770
Total	272	549	826	1684	3494	7252	11960	17744	22231

4 Proposed FHTR Tritium Management Strategies

Based on the ORIGEN analysis, the FHTR will produce about 10 Ci/day. Using a 70% capacity factor, the 20 MW FHTR is expected to generate about 2600 Ci of tritium per year. Assuming 40% is retained in structural or cooling systems as demonstrated in the MSR experiment [26], 1560 Ci will need to be removed from the reactor. From the tritium control and management strategies demonstrated at the MITR and other existing nuclear systems, the following methods are proposed for the FHTR:

1) An inert purge gas and recombiner system. Platinum has very high melting point 1773°C, known to operate in diesel systems as a three-way catalyst at 700 to 800°C [33]. It is used at lower temperature in the MITR's recombiner system and can be potentially adopted for the FHTR's recombiner system with higher efficiency.

2) Solid getters (Pd, Ni, W) that trap the tritium gas [26]. The disadvantage is that they have limited recoverability from the reactor. They must be placed outside of the core to not impact reactivity.

3) Carbon-based getters (~20% trapped in graphite moderator in MSRE [34]). Carbon nanoparticles circulating in the primary coolant can absorb tritium and then filtered out and removed from the core. They can also include metallic nanoparticles outside of the primary loop where there is no neutronic limitation [26].

Operational experiences of the MITR and PWRs have shown that tritium gaseous effluent release is typically less than 100 Ci per year, although few facilities have released up to 870 Ci without exceeding the general public dose limit. Using these values as references, a tritium removal rate of > 90% (or <10% leakage rate) should be used as a design target for the proposed tritium control and management of the FHTR.

5 Conclusion and Recommendations for Future Work

The goals of this thesis were to provide an overview of tritium control and management strategies of existing nuclear reactor systems, to estimate the total tritium production expected in the FHTR using the ORIGEN-S computer code, and to propose a tritium management strategy for the FHTR. The review of light water reactors shows that tritium is the primary radionuclide of interest for gaseous and liquid effluents up to several hundred curies per year. Tritium management in light water reactors and other nuclear systems has paved the way for regulation experience that may be well suited for the FHTR.

Practical methods of tritium sampling, removal, and storage have been examined through the study of tritium management in other nuclear reactor systems. The methods studied show that these technologies, like those in the detritiation facilities for CANDU reactors, have been developed and implemented for commercial and test reactor use. These technologies can potentially be applied to the FHTR.

The total tritium production was estimated using the core design of the MIT Nuclear Systems Design Class 100 MW FHTR, with the neutron flux adjusted to 20 MW. The result for tritium production is 2600 Ci per year, with a 70% capacity factor at a rate of ~10 Ci/day. The results also show that ^6Li contributed the most to tritium production, as expected. The source term indicates that it is essential to implement tritium control and management in the FHTR. A tritium removal rate of >90% is proposed as a design target in order to maintain tritium release within the limits of existing nuclear reactors, which maintain proper radiological controls for radiation workers and the general public. The proposed tritium management strategy includes an inert purge gas and recombiner system, solid getters, and carbon-based getters.

Future work recommended for tritium control in the FHTR includes:

1) The study and design of an inert purge gas and recombiner system

2) Further research on getters and nanoparticles as they relate to high-temperature molten-salt environments

3) The study of tritium removal methods from molten-salt and/or gas after the inventory of tritium reaches 10 Ci/kg in the moderator, as in CANDU reactors, and potentially selling it for detection or research purposes

4) To increase the ^{7}Li enrichment in flibe

5) To establish an operational specific activity limit similar to that of the MITR (5 Ci/liter) to limit tritium inventory in the system, and

6) To employ a multiple-barrier approach to prevent release to the environment as detailed for CANDU reactors in Section 2.4.2.

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7 Appendices

7.1 Appendix A ORIGEN-S Input

```
=origens
0$$ a5 28 e
1$$ 1 1t
FHTR Tritium Production 30, 60, 90, 180, 360, 700, 1080, 1500, 1800 days irradiation
' Library integer constants (not changed from MITR)
3$$ a4 -82 a11 0 0 a16 2 a33 18 e
' Library Constants (Total Core Fluxes)
4** 0.489 5 6.5 e 2t
35$$ 0 4t
' Subcase control constants (9 time intervals in unit days for a flux input for 4 nuclides)
56$$ 9 9 1 a13 4 4 3 0 2 1 e
57** a3 1e-6 e t
Irradiation 1
material charged to reactor
' Thermal flux, 9 time intervals repeating
59** 9r4.56e+12
' Time intervals (days)
60** 30 60 90 180 360 700 1080 1500 1800
66$$ a1 2 a5 2 a9 2 e
' Isotope atomic number
73$$ 30060 30070 40090 90190
' Isotope amount (grams)
74** 0.01 141.7 91 767.3
' Library kind (Using relative abundance in Light-Element Library for 4 elements)
75$$ 4r1 t
' Final Cooling (take concentration at 1st time interval step, 30 days)
56$$ 0 1 a10 1 a15 3 a17 4 e 5t
Final Decay
material discharged from reactor
' Decay period (not relevant)
60** 1
65$$ 3z 1 2z 1 17z 1 2z 1 17z 1 2z 1 14z
61** 7r1e-6
81$$ 2 0 26 1 e
82$$ f2 6t
56$$ f0 t
end
```

7.2 Appendix B ORIGEN-S Output

primary module access and input record (Scale 5.1 driver)
module origens will be called at 19:07:08.078 on 05/07/2013.

```
0$$ a5 28 e
1$$ 1 1t
FHTR Tritium Production 30, 60, 90, 180, 360, 700, 1080, 1500, 1800 days irradiation
' Library integer constants (not changed from MITR)
3$$ a4 -82 a11 0 0 a16 2 a33 18 e
' Library Constants (Total Core Fluxes)
4** 0.489 5 6.5 e 2t
35$$ 0 4t
' Subcase control constants (9 time intervals in unit days for a flux input for 4 nuclides)
56$$ 9 9 1 a13 4 4 3 0 2 1 e
57** a3 1e-6 e t
Irradiation 1
material charged to reactor
' Thermal flux, 9 time intervals repeating
59** 9r4.56e+12
' Time intervals (days)
60** 30 60 90 180 360 700 1080 1500 1800
66$$ a1 2 a5 2 a9 2 e
' Isotope atomic number
73$$ 30060 30070 40090 90190
' Isotope amount (grams)
74** 0.01 141.7 91 767.3
' Library kind (Using relative abundance in Light-Element Library for 4 elements)
75$$ 4r1 t
' Final Cooling (take concentration at 1st time interval step, 30 days)
56$$ 0 1 a10 1 a15 3 a17 4 e 5t
Final Decay
material discharged from reactor
' Decay period (not relevant)
60** 1
65$$ 3z 1 2z 1 17z 1 2z 1 17z 1 2z 1 14z
61** 7r1e-6
81$$ 2 0 26 1 e
82$$ f2 6t
56$$ f0 t
```

module origens is finished. completion code   0. cpu time used   1.44 (seconds).

program verification information
code system: scale version: 5.1
program: origen
creation date: 02_nov_2006
library: c:\scale5.1\bin
production code: origens
version: 5.1.2
jobname: scale5.1
machine name: ice-lt
date of execution: 07_may_2013
time of execution: 19:07:08.18

Irradiation 1   light elements   page 1
    power= 4.891E-05mw, burnup=8.8036E-02mwd, flux= 4.56E+12n/cm**2-sec
                        nuclide concentrations, grams
                    basis = material charged to reactor

*results outlined in Figure 4 and Table 11

    Final Decay   light elements   page 2
                        nuclide concentrations, grams
                    basis =material discharged from reactor

          charge discharge   1.0 d
  h 1   0.000E+00 5.284E-06 5.284E-06
  h 3   0.000E+00 3.176E-05 3.176E-05
  he 4 0.000E+00 1.502E-03 1.502E-03
  li 6   1.000E-02 1.018E-02 1.018E-02
  li 7   1.417E+02 1.417E+02 1.417E+02
  be 9 9.100E+01 9.100E+01 9.100E+01
  be 10 0.000E+00 7.008E-06 7.008E-06
  o 16 0.000E+00 1.024E-03 1.024E-03
  f 19   7.673E+02 7.673E+02 7.673E+02
  ne 20 0.000E+00 1.216E-04 1.216E-04
  total   1.000E+03 1.000E+03 1.000E+03

    Final Decay   light elements   page 3
                        nuclide radioactivity, curies
                    basis =material discharged from reactor

          charge discharge   1.0 d
  h 3   0.000E+00 3.071E-01 3.071E-01
  total   0.000E+00 1.583E+03 3.071E-01