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TRITIUM MANAGEMENT IN FUSION SYNFUEL DESIGNS

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TRITIUM MANAGEMENT IN FUSION SYNFUEL DESIGNS

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ABOTHACT

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This year we have begun a new program at Lawrence Livermore Laboratory (LLL) on synfuels from fusi h. in collaboration with the University of Washington and Exxon Nuclear. In this program, we are designing a fusion blanket that can provide the heat and be efficiently interfaced to any one of three therm chemical cycles that have been demon-strated in a closed-loc; pilot scale. Two blanket types are being statued: a lithium-sodium pool tabler and a ...thism-oxide- or aluminatemicrosphere moving bed. For each we have considered a wide variety of current technology in handling the tritium. Here, we show the pool boiler with the sulfor-iodine thermochemical cycle first developed and now being piloted by the General 'tomic Company. The tritium (T2) will be generated in the lithium-sodium mixture where the concentration is ~10 ppm and held constant by a scavenging system consisting mainly of permeators. An intermediate sodium loop carries the blanket heat to the thermoenemical cycle, and the T_2 in this loop is held to i ppm by a similar scavenging system. With this design, we have maintained blanket inventory at 1 kg of tritium, kept thermochemical cycle lossen to 5 Cl/d and environmental losses to 10 Ci/d, and meld total plant misk inventory at 7 kg tritium.

INTRODUCTION

The production of synthetic fuels will be of increasing innortance in the coming decades, as more acute shortages of transportable fuels occur. In the part, the fucion community has focused on power plants for electric power production. In addition to future electrical power plants being fusion based, we need to develop and examine a variety of synthetic fuel production concepts for which a fusion energy source may be used. The Tarden Mirror Reactor (TMR) is an emerging engineering design concept for fusion energy based on Lawrence Livermore Laboratory (LLL) tandem mirror physics now being developed.

The coupling of a TMR to one of three thermochemical cycles is being studied under a new Department of Energy (DOE) contract at LLL enti-Lled, "Synfuels From Fusion." In this design study, we are taking unique advantage of the favorable engineering features of the tandem configura-

----Work performed under the auspices of the U.S. Department of Energy by the Lawrence Livermore Laboratory under contract number W-7405-Eng-48. tion that is a simple, cylindrical-solenoidal, energy-producing section between end cells. The end cells provide the containment. The centralcell solenoid contains modular blanket sections that produce both high-temperature process heat (950°C) and the required tritium to run the DT reactor. Material and engineering structural problems are minimized by the geometry, steady state temperature, and magnetic field.

Two tandem mirror blanket concepts are being explored: a lithium-sodium liquid-metal pool boiler and a lithium-oxide-microsphere moving bed. Three thermochemical cycles are being examined to reveal their specific advantages or disadvantages in interfacing with the TMR: the General Atomic* sulfur-iodine cycle; the Westinghouse sulfur cycle; and the International Atomic Energy Agency at Ispra, Italy, Mark 13-V2 cycle. Here, we describe the approach for handling tritium in the fusion reactor in connection with the General Atomic sulfur iodine process.

THE LITHUM-SODIUM CAULDRON BLANKET DESIGN

The original cauldron blanket concept of Lee (1-3), linked to the present TMR geometry, allows up to surround the plasma with a sealed vessel containing a binary mixture of 50% lithium and 50% sodium. This simple pool boiler is shown in Figure 1. The plasma energy input heats the fluid mixture and causes the sodium to preferentially vaporize. The vapor pressures of lithium and sodium at 1790 K are 0.02 and 1.4 atm, respectively (1), a difference of 75 times. The lithium is left behind in the liquid state to breed tritium. Lith-Lum along could not be used because unreasonably high vapor velocities would be necessary to carry the heat away. The sodium vapor, transporting energy as latent heat, condenses on heat exchange tubes that are outside of the magnetic field gome and are therefore free from magnete hydrodynamic effects. Energy is then transferred from the condensing-vapor heat exchanger to socium coolant. This is accomplished by heat transfer fluid in an intermediate loop that carries the heat outside the nuclear inland to the myriad process exchangers within the thermochemical hydrogen-production cycle.

In the cauldron, the lithium and sodium are completely miscible and a circulation pattern is

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Figure 1 Lithium sodium cauldron pool boiler design

established by two effects: the exponential decrease in energy deposition in the blanket as a function of radius, creating a temperature and dencity gradient; and the density difference between liquid sodium and lithium. Additional means to drive the circulation are provided by sparging with holium gas.

The structural container for this hot, 1200-K fluid mixture is actually held at a temperature of 700 to 750 K so that a significant AT is introduced across the first wall. This is accomplished by providing a compressively loaded membrane on the not side, and heat excharger tubes on the cool side. Sandwiched in between is a netallic sponge with a low thermal conductivity value, perhaps 10% that of common metals. The thickness of this sponge is a design parabeter and can be set somewhat at our discretion, probably ~2 cn. About a 40% void fraction of interconnected pores is selected so that a controllative mass flow rat; of soclum can be passed through this sponge in order to select the 500- to 550-⁶⁰ CAT we require. One of the favorable characteristics of this module is its low operating pressure (\sim 1.5 atm) and consequent low stresses. These low stresses, coupled with the low structural temperature, makes safety and integrity of containment the real attraction of this design concept.

The individual cauldron blanket modules are interspersed between successive solenoidal coils which are in turn spaced around four central meters at a distance determined by allowable ripple (5%) in the B field. There is likely to be 6 modules for each 4-meter section, or approximately 150 modules in all. The modules are isolated from the outside environment by:

- A double wall structure,
- A carbon reflector and shield,
- An interior envelope covering the reactor and held at approximately 10⁻⁶ to 10⁻⁵ Torr, and
- A 1.0- or 1.5-m steel-lined, concrete containment structure that is evacuated or contains an inert cover gas.

TRITIUM LOSS LEVELS

Tritium losses in the TMR were set below 10 Cl/d to meet the goal of emissions to the environment, while still maintaining worker safety and cost effectiveness. A number of new methods have been employed to achieve such low levels in a nearterm machine. Figure 2 summarizes our goals for the various tritium leak source terms and the associated processing loops.

Tritium bred in the blanket will permeate the condenser tubes in the lithium-sodium pool boiler or the lithium-oxide helium contactor, and eventually the sodium loop feeding the numerous Incoloy-800 process exchangers within the large thermochemical plant. We have set a limit of this tritium leakage into the thermochemical process at 5 Cl/d. For 100,000 m^3/h of hydrogen production, this would produce a maximum of 2 µCi/m³ of tritium (HT) in the hydrogen product, 20% of the 10-µCi/m¹ limit established as a breathable atmosphere for the occupational worker. By the time any of this hydrogen gets to the market place, it would be diluted below the 0.04-µCi/m³ HT level set as a breathable atmosphere for the general public. For example, at a 10% level of hydrogen in natural gas supplies, a 0.04-µCi/m³ tritium (HTO) level Would be typical. Brockman's group to Julich (5) believe they can meet a $0.01 - \mu Ci/m^3$ level for HTO in the hydrogen used for synthetic natural gas. It appears uneconomical for us to neet such low levels in our design. Consequently, we have set a product level of 5 Ci/d for each of our design concepts able to seet this goal. Blanket module failure or sodium loop failures must not raise the tritium leakage into the thermochemical process to unacceptably high levels.

CONTROL OF TRITICH FRODUCED IN THE LITHIUM-SODIUM POOL BOILER

In this section we outline several strategies for handling trillum in the lithlum-sodium boller blanket, and illustrate the key results. We exacine first two alloy candidates, titanium-zirconium-



Figure 2. Tritium source terms and processing loops, All flow rates in g/h.

molybderum alloy (TZM) and Incoloy-800, for the pool condensing-vapor neat exchanger. Then we look at the feasibility of niobium permeators and precious-metal moving getter beds.

First, we need to establish the relationship between the concentration (i.e., inventory) in the lithium-sodium boiler and the tritium partial ressure. There are no data that we know of concerning hydrogen or its isotopes in the lithium-sodium binary system. Heumann and Salmon (6), and Maroni and coworkers (7), have measured partial pressures of tritium, deuterium, and hydrogen over lithium from 700 to 1000°C. Tritium exhibits a very different partial pressure p dependence on concentration than do either deuterium or hydrogen, and there appear some doubts (7) as to whether Raoult's law (p = X) or Sievert's law (p = X^2) apply. Impurities in the tritium measurements may have caused these results to be anomalous, and recent data analyses (3) indicate Sievert's law is preferred. Consequently, we have used Sievert's law as representing the base case situation:

$$p(at_m) = (X/K_a)^2$$
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where

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 $K_{g} = 0.159 \text{ atm}^{-1/2} \text{ at } 950^{\circ}\text{C}$, and

X = the mole fraction of tritium.

If the lithium-sodium boller has a $1256-m^3$ volume and contains 1 kg of tritium, we estimate the mole fraction of tritium (T) at about 10 ppm. This will differ slightly depending on the ratio of

lithium to sodium, but a mixture with equal amounts of each was used throughout. For this cituation, the partial pressure of T is calculated as 4×10^{-3} atm, assuming the pu- atomic species exists and neglecting HTO, CH₃T, LiOT, NaOT, etc. These impurities are expected to be minor constituenta, but should not be ignored completely.

Available permeation data (8-13) for various metals and alloys are displayed in Figure 3. It is obvious that for applications at about 950°C, alloys low on the plot are an excellent choice owing to their low tritium permeation.

Lithlum-Sodium Vapor-Space Processing System With a TZM or Incoloy-800 Condenser

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Here we outline the lithium-sodium vapor-space processing system, and then the main heat transfer loop and its tritium scavenging system (Figure 4). First we calculate the lithium-sodium vapor permeator area based on small-diameter niobium tubes ∩.4-mm thick with 2-atm low-flow helium sweep gas flowing on the shell side at 1000°C. The tubes are hotter than the lithium-sodium vapor in order to keep the permeator surface clean. This permeator can be placed external to t 3 blanket with the lithium-sodium vapor-space gases circulating through the tubes. If we assume 8% of the deuterium and tritium react (the upper limit), about 16 g/h of tritium will have to be recovered. With a permeation coefficient of 200 cc (STP) $\operatorname{cm}^{1/2}(8)$, a permeator area of 733 m² is required, assuming there is no



Figure 3. Permeation of hydrogen through various alloys.

degradation of surface activity from dirt and impurities.

With a partial pressure of 4×10^{-9} atm. the tritium (T_2) in the lithium-sodium boiler will permeate through the 6000 m2 of TZM 1-mm-thick condenser walls at 600 Ci/h, assuming that the gas species is either OT or HT, and not T2. This permeation of tritium into the sodium loop will continue to raise the sodium-loop tritium concentration to a level that would adversely affect the thermochemical process. We believe that controlling the tritium loss to 5 Ci/d is mandatory. If we limit the permeation loss through the $6000-m^2$ Incoloy-800 process exchanger to 5 Ci/d, the sodium-loop T_2 partial pressure must be held to 2×10^{-12} atm. This problem is made much each atm. This problem is made much easier by a chromium-oxide layer which is expected to form on the thermochemical process side as the result f oxidizing conditions in the stream. For this situation, a permeation coefficient of 3.5 x 10-3 is expected (13). The details of these results are discussed in Reference 14.

We can maintain the partial pressure in the sodium loop at 2×10^{-12} atm by means of a sodium loop permeator with an area of 120 c^2 . The additional permeator would also be a miobium tube exchanger with hot helium sweep gas on the shell side, and sodium on the tube side. Again, the tube would be kept at 1000° C, which is hotter than the sodium, in order to keep the surface clean. The hot helium would be scavenged of its low-level tritium by a conventional catalytic oxidation and mole sleve-adsorption process train (15-19).

The concentration of T₂ in the sodium loop corresponding to a partial pressure of 2 x 10^{-12} atm can be estimated from sodium-hydride data (20), assuming Sievert's law, at 2.6 ppb. These data and our methods of extrapolation are only approximate, but at least an estimate is helpful.

Incoloy-800 alloy for the in-blanket condenser is a vlable alternate to TZM, as shown in Figure 5. This material permeates 3.5 times faster than TZM. Thus, the transport of tritlim from the lithiumsodium boiler into the sodium loop will increase to 2090 Ci/h. If we still restrict process losses to 5 Ci/d, then we calculate from permeation rates that the sodium loop permeator area must be increased to 730 a^2 . No other units in the process train need to be altered. We believe this larger permeator is still feasible.

Lithium-fodium Vapor-Space Processing System With a Moving Getter Bed

This process is similar to the one above, except that the lithium-sodium boiler permeator is replaced by a getter bed (Figure 6). Here the getter bed must handle 16 g/h of tritium. From a mass balance computation, it appears feasible if the bed is continuously moved through a regenerator process circuit at a rate of about 38 kg/d for bed loadings of 1% by weight, or 380 kg/d for 0.1%. For a bed residence time of 10 h, this would amount to an inventory of only 160 g of tritium. With beds of 1-cm coated part(cles in lengths of several meters and diameters about a third of a meter, the pressure drops wou'd be very small.

Although we have only experimental data on the performance of a vttrium getter bed with lithium at tritium ppm levels and 200°C (21), we believe we could process the inert-gas vapor-space at 950°C. Clearly, we need more experimental work. Singleton et al., (22) showed that good purification factors (i.e., inlet/outlet concentration ratios) of 10 to 20 were achievable with cerium in the tritium concentration range of 0.1 ppb to 100 ppb in argon. Because we are seeking levals of 10 ppm tritium in the lithium-sodium melt, these lithium-yttrium and helium-cerium performance tests are very encouraging. It appears that with adequate vessels (with tritium permeation lower than 316-stainless steel) the getter bed can be operated in excess of 600°C with no decline in performance (22).

Because yttrium-lithium-tritium getter systems can only reach about 1 ppm at 200° C, and we need levels of 2 to , ppb in the sodium loop, the yttrium getter approach does not appear workable.

Lithium-Sodium Vapor-Space Processing System With Tritium-Removal Heat Pipes

This process is also similar to those above, except that the lithium-sodium boller permeator is replaced by tritium-concentrating heat pipes (Figure 7). The principle is discussed in References 23 and 24, and involves a conventional heat



Figure 4. Lithium-sodium vapor-space processing system with a TZM condenser.

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Figure 5. Lithium-sodium vapor space processing system with an Incoloy-800 in-blanket condenser.



Figure 6. Lithium-sodium vapor-space processing system with a moving getter bed.



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Figure 7. Lithium-sodium vapor-space processing system with tritium-removal heat pipes.

pipe that is gas-buffered where the inside sweep uction of liquid-metal vapor pumps the gas and concentrates it at one end. If the gas contains tritium, this design allows T₂ concentration at partial pressures of approximately 1 atm in one end where permeation through a small window into the tritium purification process can proceed rapidly (24).

In the original concept, the heat pipe contained lithium, which under a neutron influence would bread tritium (24). In this case, however, we are breading tritium in the lithium-softum boiler, and would utilize this heat-pipe concept simply as a heat-driven concentrator using sodium. We first examined the feasibility of replacing the condenser tubes with heat pipes of Incoloy-800. The permeability of this alloy in a tube area of 6000 m² is, as previously indicated, 2090 Ci/h. The lithium-sodium holler T₂ inventory is 1 kg at a pertial pressure of 4 × 10⁻² atm.

The condenser and would permeate tritium at the required rate to the tritium purification system through a small nictium window at the tube's end. This is possible because the T₂ in the heat pipe would increase to a partial pressure of about $\beta + 10^{-3}$ atm.

Our design requires a heat transfer or permeation area of 100 m² at the emporator end of the heat pipe, at did the condenses shown in Figure 5. This design offers the advantage of concentrating and expanding the totum. Convergently, it appears that this trait in-concentrating heat pipe would be extellect for the blanket design where trutum procedum grant by the heat pipe to a separate part of the process.

According to ear calculations in back diffusion tolow, the heat pipe can operate with a large partual pressure difference between the ends (i.e., tolow 6×10^{-2} atm at the evaporation end, to about it 10^{-2} atm at the condenser end) because the large driving force for back diffusion of tritum is against the condum waper velocity.

We can estimate the extent of this back diffuingly determining the binary diffusion (coefficycal of tritium in sodium vaper). Critical propential for solium vaper are as follows (25):

Temperature T_c = 2800 K Precoure F₁ = 510 atm Volume V_c = 150 cc/gmole Denoity p_{c} = 0.1533 g/cc Compressibility: 2_{c} = 0.327

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We estimate the binary diffusion coefficient at 1200 K to be 2.23 $\rm cm^2/s$, as discussed in Reference 26,

If we use 100,000 heat pipes about 2-m long and 2 cm in diameter, thi sodium vapor flux will be about 4 gm/cm²-s, or a linear velocity of 116 m/s within the tube. We found that with the condensor end containing 2 x 10⁻² atm of T₂, the back diffusion of the T₂ is exactly matched by the bilk flow of the sodium vapor in the opposite direction to obtain an acceptable rate of 10 g/h of traitium. We can construct windows at the tube ends with a total area of 30 m², determined from permention calculations. These windows would be a i-mm-thick tipk of a pipelum.

fitted to the end of the tube. This permestion window design would contain the T_2 at the condenser end at a partial pressure of 1×10^{-3} atm, safely below the 2×10^{-4} atm at which back diffusion begins to be a problem.

REMOVAL OF HELIUM-4

The removal of helium-4 is critical to the operation of the blanket. The helium-4 is generated at a rate equal to that of the tritium, and the helium-4 will not permeate through metal permeators. We find that hel.um-4 will permeate through fused silica at about 10 times the rate of tritium in niobium, with a helium-4 partial pressure of 0.45 atm in the blanket (27-29).

As a result, helium-4 could be removed by a $1100-m^2$ hot fused-silica window. Less than 0.1 Ci/h of tritium would pass through the window along with the 16 g/h of helium-4. The nearly pure helium-4 could then be fed to a standard satalytic oxi `tion/molecular sieve-adsorption unil to recover any residual tritium for the fuel purification process. The helium-4 would then be cleaned to tritium levels of about 0.1% that of the incoming stream, or less than 0.1 Ci/d. We believe at this point, the helium-4 could be safely stacked, adding less than 0.1 Ci/d to the estimated tritium loss of 10 Ci/d.

A partial condenser is required to remove vaporized lithius or sodium or other impurities that could condense on the fused-silica surface (or the nicbium nurface as well). By condensing out any impurities, and keeping these silica tubes botter than the gas processer to avoid significant impurity adsorption, a long, high-efficiency life is ensured. This proposed terhnique would have to be tested, however, since avea, small amounts of noncondensed lithium or socium would limit the lifetime of the fused-silier premeater. It would be advantageous to expand the function of this partial condenser with a processing loop sapable of removing nore complex impurities through the use of special getters or chemical convengence.

Another major source of tritium leakage is the tritium neutral-beam injector, where 55-keV tritons implant tritium in the electrode tubing surface. Once in the tube motal, we calculat that the tritium will diffuse, producing a flue of tritium into the electrode belium-conlant flow at about 70,560 Ci/d. A 100 fraction of the coolant flow is then processed to remove 99% of the tritium. The resulting residual tritium leaks into the beat exchangers or steam generators at about 3,5 Ci/d, where it eventually parks into the cooling water and thes to the covingment. The feutrum injector unce destrict with a maximum 1% tritium impurity level, and it meates a 0.3 Ci/d leak to the process.

A similar tritium loss securs in the TMR's direct converter, where 55-keV tritons produce a leak of 8000 Ci/d into the belium flow. Again, a 39.9% processor is used, handling 5% of the flow to reduce the tritium iss to less than 3 Ci/d.

All of the remaining miscellaneous leaks from the nuclear island equipment and piping or electing are assumed to leak into the reactor hall at a maximum rate of 180 21/d. The reactor hall atmosphere is processed and kept at leak bundity to maintain tritium gas levels of below 40 μ Ci/m³, and HTO lev-.ls below 10 μ Ci/m³. As a result of this reactor hall processing design, routine as well as credible accidental losses of less than 1 Ci/d through the reactor hall are expected.

The tritium loss to the thermochemical process and thus to the environment is designed to be around 12 Ci/d, with 5 Ci/d dissipated from the blanket, 3 Ci/d from injectors, 3 Ci/d from the direct converter, and less than 1 Ci/d from the reactor hall. This level is consistent with other studies (15-18, 30-10) and Muclear Regulatory Commission (NRC) regulations (41, 42) for all nuclear fuel cycle operations, but is about the level suggested by the NRC "As Low As Reasonably Achievable" (ALARA) cost-benefit analysis (19, 41-44) with an expenditure cutoff of \$66,000. Using the ALARA criterion, we found that no further reductions in emissions could be accomplished by an expenditure of 456,000 for additional oxidizer-molecular sieves.

CONCLUSIONS

Ideally, the T2 will be generated within the lithium-sodium mixture and the concentration will be allowed to increase to a 1-kg inventory or ~10 ppm on a molar basis or about 1000 Ci/m3. The T_2 partial pressure will be maintained at a constant $4\,\times\,10^{-9}$ atm by a scavenging system continually removing tritium from the vapor space above the lithium-sodium pool. For a tritium breeding ratio in slight excess of unity, this scavenging system must handle tritium at a rate of ~15 g/h or 150,000 Ci/h for a 2500-MW $_{\rm t}$ reactor. The key to the scavenging system is the use of a 0.4-mm niobium tubul r-array permeator at a high temperature (1000 °C). The surface area requirements were found to be 750 ${\tt m}^2$. The interiors of these tubes are swept with 1000°C helium in order to keep the tubes hot, clean, and low in tritium. The tritium will be processed by conventional oxidation/adsorption processes. This technique maintains the tritium inventory in the blanket at ~1 kg. Helium-4, the other byproduct of the neutron-lithium reaction, is removed from the blanket wapor space by a fused-silica tube permeator of $\sim 1000 \text{ m}^2$. A vapor condenser would protect the silica tube from liquidmetal condensation.

Tritium losses from the blanket through the heat exchangers into the thermochemical cycle must be controlled within acceptable limits (2 µCi/m³ or 5 Ci/d). In one of our conceptual designs, heat is extracted from this lithium-sodium pool boiler by sodium vapor condensation on Incoloy-800 tubes within the vapor dome of the blanket. Tritium will leak out of the blanket via this path into the liquid-sodium coolant flowing in these condenser tubes. The permeation rate was found to be 2000 Ci/h. Such a large permeation leakage can be controlled by installing another niobium tube permeator on the liquid-sodium flow side with a tube area of 730 m2. Tritium would thus be removed at 2000 Ci/b. The partial pressure of tritium in the sodium loop will increase until the amount of tritium removed by the sodium-loop permeator, and the tritium leaked by permeation out of the Incoloy-800 process heat exchanger, exactly match the permeation into the sodium loop via the in-blanket condenser tubes. We find that when we do this trial

and error calculation, the T2 partial pressure in the sodium levels at about 2×10^{-12} atr. We assume there is an oxide barrier in the Incoloy-800 process exchangers, such as sulfur trioxide decomposition. Actual high-temperature gas-cooled reactor experience at Peach Bottom has shown that Incoloy-800 offers excellent tritium retention properties by the formation of a protective oxide barrier with a permeation reduction factor of 200. With the oxide barrier, the tritium loss to the thermochemical process would be around 5 Ci/d, which meets our environmental goals. Further reductions of tritium losses were possible. However. they were not cost-effective according to the NRC criterion (ALARA) of \$1000/man-rem or \$20/C1/y released.

We have explored several accident modes. The sodium loop between the fusion blanket and the process functioned effectively as an isolation loop. A if leakage of tritium at a breeding rate of 16 g/h would add only a 1600 Ci/h load to the sodium loop permeator, and increase tritium loss to the thermochemical process by only 28 Ci/d. This 1% leakage appears to meet the present day NRC criteria for fission plants. Likewise, gross failure of the in-blanket condenser would merely dump the lithium-sodium blanket material from one module (i.e., 1/150 of blanket inventory) into the sodium coolant loop. This would not result in explosive accidents, and add only 20 Ci/d to the tritium loss. All lithium-sodium vessels were surrounded with graphite spheres, and the sodium piping was contained within inerted pipeways that were steellined to prevent sodium concrete reactions.

The remaining tritium losses were calculated for permeation and leakage from other process component designs and found to be small (i.e., 3 Ci/d from the direct converter, 3 Ci/d from neutral-beam injectors, etc.). Tritium inventory within onalte, remote storage tanks was 16 kg. For the cryopumps, inventory was estimated to be 4 kg. Cryogenic distillation columns held 1.4 kg. The remaining cryopanels, molecular size beds, etc., contained a total of 0.5 kg. The in-process tritium inventory at risk therefore (not including storage tanks separated from the reactor), is expected to be about 7 kg. F

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