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TRITIUM MOVEMENT AND ACCUMULATION IN THE NGNP SYSTEM INTERFACE AND HYDROGEN PLANT

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ABSTRACT

Tritium movement and accumulation in the Next Generation Nuclear Plant (NGNP) employing either a high-temperature electrolysis (HTE) process or a thermochemical water-splitting Sulfur-Iodine (SI) process to produce hydrogen is estimated by a numerical code, THYTAN, as a function of design, operational and material parameters. Estimated tritium concentrations in the hydrogen product and in the process chemicals of the hydrogen plant using the HTE process are slightly higher than the limit in drinking water defined by the U.S. Environmental Protection Agency (EPA) and in effluent at the boundary of an unrestricted area defined by the U.S. Nuclear Regulatory Commission (NRC), respectively. Estimated tritium concentrations in the NGNP using the SI hydrogen production process are significantly higher, and are largely affected by undetermined parameters (i.e., tritium permeability of heat exchanger materials, hydrogen concentration in the helium energy transport fluids, equilibrium constant of the tritium isotope exchange reaction between HT and H_2SO_4). These parameters should be measured or estimated in the near future, as should the tritium generation and release rate from the NGNP nuclear reactor core. Decreasing the tritium permeation rate between the primary and secondary heat transport circuits is an effective measure to decrease the tritium concentrations in the hydrogen product, hydrogen plant process chemicals, and the tertiary heat transport fluid.

KEYWORDS: *Next Generation Nuclear Plant, Very High Temperature Gas Cooled Reactor, high-temperature electrolysis, thermochemical water-splitting, SI process, tritium*

I. INTRODUCTION

Hydrogen is a clean energy media that releases no CO_2 when consumed or burned. A high temperature gas-cooled reactor (HTGR) can produce massive amounts of high-temperature thermal energy without a controlled release of any CO_2. Thermal energy from an HTGR can be used to produce heat and electricity, and these energy products can be used to split water into hydrogen and oxygen. Because of these characteristics, the U.S. Department of Energy (DOE) is pursuing the development of a plant that combines a HTGR with a thermally driven water-splitting hydrogen plant called the Next Generation Nuclear Plant (NGNP). In the NGNP, a very high temperature nuclear reactor (VHTR) would provide heat and electricity to either a thermochemical water-splitting plant or a high-temperature electrolysis plant to produce hydrogen (and oxygen). The research and development (R&D) activities related to the nuclear reactor, energy transport, and high-temperature heat exchanger technologies are being carried out by DOE's NGNP Project, and R&D activities related to nuclear powered hydrogen production are being carried out by DOE's Nuclear Hydrogen Initiative (NHI).

One potential problem with using a thermal coupling to transmit high-temperature heat from a nuclear reactor to a hydrogen production plant, as opposed to using strictly an electrical connection, is the transmission of tritium from the nuclear reactor primary coolant to the hydrogen product by the permeation of tritium through the heat exchanger surfaces and bulk transport of tritium within the heat transport loop that would connect the nuclear reactor with the hydrogen production plant. The tritium behavior in HTGRs was evaluated in several countries in the 1970's (e.g., the Dragon Reactor in the U.K.[1], the Peach Bottom HTGR in the USA[2], the AVR in Germany[3]). Data from the operation of HTGRs and laboratory experiments revealed the mechanisms of tritium production, transport, and release to the environment. According to a review by Gainey[4], calculations with some of the HTGR data showed that tritium releases should be well within the U.S. Federal guidelines contemporary with the earlier HTGR operations, and so further laboratory-scale work related to tritium transport in HTGRs was discontinued. However, HTGRs at that time only produced electricity and were not targeted towards high-temperature heat applications, and the potential for tritium migration into downstream processes was not evaluated. Also, tritium accumulation in the process chemicals of the hydrogen plant may become a critical issue in seeking to operate the hydrogen plant associated with the NGNP as a non-nuclear facility, and to provide hydrogen to customers that meets specifications in regard to tritium.

In the U.S.A., the U.S. Nuclear Regulatory Commission (NRC) is continuously evaluating the latest radiation protection recommendations from international and national scientific bodies to ensure the adequacy of the standards the agency uses. Among those standards, the NRC and the U.S. Environmental Protection Agency (EPA) have established radiation protection limits to protect the public against potential health risks from exposure to radioactive liquid discharges (effluents) from nuclear power plant operations. The NRC's final layer of protection of public health and safety is a dose limit of 100 mrem per year to individual members of the public. For gas and liquid effluents, including water contaminated with tritium, any NRC licensee can demonstrate compliance with the 100 mrem per year dose standard by not exceeding the concentration values specified in Table 2 of Appendix B to 10 CFR Part 20. For tritium, the effluent concentration in air and water are 3.7 x 10^{-3} Bq/ml (100 pCi/L) and 37 Bq/ml (1 000 nCi/L), respectively. In 1976, the EPA also established a dosed-based drinking water standard of 4 mrem per year to avoid the undesirable future contamination of public water supplies as a result of controllable human activities related to nuclear industries. To achieve this result, the EPA set a maximum contamination level of 0.74 Bq/ml (20 nCi/L).

In this study, the tritium concentrations in the NGNP using the Sulfur-Iodine (SI) process or using the high-temperature electrolysis (HTE) process to produce hydrogen are evaluated by a numerical analysis code, THYTAN (Tritium and Hydrogen Transportation Analysis code), as a function of design, operational and material parameters. Since there is no established limit on tritium in commercially available hydrogen, the calculated tritium concentration in the hydrogen product is compared to existing limits on tritium in liquid and gaseous effluents. Since it is desired to maintain the hydrogen plant portion of the NGNP as a non-nuclear facility, the tritium concentrations in various hydrogen plant components are also calculated for each hydrogen production process and compared with existing limits. The dividing line between the nuclear and non-nuclear portions of the NGNP still has not been set, and so tritium concentrations are calculated for the primary, secondary, and tertiary thermal energy transport loops as well. Limited parametric studies of the factors affecting tritium concentrations in those components are performed, and some conclusions are drawn as to which methods might be the most effective in lowering the tritium contamination in the hydrogen product and hydrogen plant components.

II. SYSTEM DESCRIPTION

II.A. NGNP Using SI Process

The SI process is one of the thermochemical water-splitting hydrogen production methods[5], and it is composed of the following three main chemical reactions.

$$I_2 + SO_2 + 2\,H_2O \overset{\leftarrow}{\longrightarrow} 2\,HI + H_2SO_4 \tag{1}$$

$$H_2SO_4 \overset{\leftarrow}{\longrightarrow} H_2O + SO_2 + 0.5\,O_2 \tag{2}$$

$$2\,HI \overset{\leftarrow}{\longrightarrow} H_2 + I_2 \tag{3}$$

In Eq. (1)-(3), the double arrow is shown to indicate that the chemical reactions are equilibrium limited. The thermal decomposition of H_2SO_4 expressed by Eq. (2) can be separated into two consecutive chemical reactions.

$$H_2SO_4 \overset{\leftarrow}{\longrightarrow} H_2O + SO_3 \tag{4}$$

$$SO_3 \overset{\leftarrow}{\longrightarrow} SO_2 + 0.5\,O_2 \tag{5}$$

The reaction expressed by Eq. (1), known as the Bunsen reaction, produces HI and H_2SO_4 by the reaction of iodine, sulfur dioxide, and water at approximately 100-150°C. The H_2SO_4 produced by this reaction is then decomposed into water, sulfur dioxide, and oxygen in Eqs. (2), (4), and (5). Eq. (4) occurs without the need of a catalyst above approximately 500°C, while Eq. (5) requires the use of a catalyst and operates at approximately 750°C or greater in order to achieve significant chemical conversion. The hydrogen iodide produced by Eq. (1) is reconstituted into iodine with the concomitant release of hydrogen in Eq. (3) at approximately 400-450°C.

Figure 1

Table I

Figure 1 shows a schematic flowsheet of the NGNP using the SI process. Its major specifications are listed in Table I. Helium is circulated in the VHTR primary loop in order to remove 600 MW of high-temperature heat from the reactor core and to transport it to the intermediate heat exchanger (IHX). The IHX in turn delivers 600 MW to a secondary helium loop. In the secondary helium loop, approximately 89% of the helium is used to to drive a gas turbine to generate electricity, and 11% of the helium is channeled to a secondary heat exchanger (SHX). The SHX supplies thermal energy to a tertiary helium loop, which in turn is used to supply thermal energy to the hydrogen production plant. The chemical reactions shown in Eqs. (1), (2), and (3) correspond to Sections 1, 2, and 3 in the figure, respectively, and the tertiary loop is connected to PHXs in the high-temperature portion of the process in Section 2. The flow rates of process chemicals used in the performance of this study were obtained from flowsheet information provided by General Atomics[6] for a 600 MW SI plant and were scaled down to match the 50 MW size requirement.

II.B. NGNP Using HTE Process

Figure 2

Figure 2 shows a preliminary flow scheme of the NGNP employing the HTE hydrogen process. Its major specifications are also listed in Table I. Helium is circulated in the VHTR primary loop in order to remove 600 MW and move it to the intermediate heat exchanger (IHX). The IHX in turn provides 600 MW of high-temperature thermal energy to a secondary energy transport loop filled with helium. In the secondary loop, approximately 89% of the helium is used to drive a gas turbine to generate electricity, while 11% of the helium is used to deliver thermal energy to a tertiary energy transport loop, also filled with helium. Analogous to the IHX, a secondary heat exchanger (SHX) is used to connect the secondary and tertiary loops. Approximately 550 MW of thermal energy are transmitted to the electrical turbine from the secondary loop, and 50 MW of thermal energy is transmitted to the hydrogen production process through the tertiary loop. Three process heat exchangers (PHXs) are installed in the tertiary loop to receive thermal energy from the loop in order to drive the HTE hydrogen production process. The HTE process is composed of a main process line and a sweep line. Hydrogen is produced in the electrolyzer installed in the main line using electricity and heat from the nuclear reactor. Oxygen is obtained from the sweep line as a by-product of the hydrogen manufacturing process. The flow rates of process chemicals for the HTE process were obtained from Oh et al.[7,8] and were scaled to utilize a thermal energy input of 50 MW in order to use similarly sized intermediate heat exchangers as for the SI process. Because the HTE process uses predominately electricity (about 85-90% of the total energy input to the process is in the form of electrical power), the amount of hydrogen produced by the HTE process using 50 MW of thermal energy input is about 7.8 times greater than a thermally driven SI process plant using the same amount of thermal energy input.

III. NUMERICAL ANALYSIS

III.A. Numerical Analysis Code

THYTAN is a dynamic simulation code for calculating mass balances of hydrogen and tritium-containing chemicals (i.e., HT, HTO, $HTSO_4$, TI) in the VHTR and nuclear hydrogen plant processes (e.g., HTE, SI). The features and basic equations of THYTAN are described elsewhere[9].

III.B. Model

III.B.1. NGNP using SI process

Figures 3, 4, 5, and 6 show the nodalization scheme of the helium loop, Section 1, Section 2, and Section 3 of the SI process, respectively. Tritium release to the primary coolant is considered at a node expressed by No. (1) in Figure 3. The IHX and the SHX are modeled by using 10 nodes in each flow channel, while the PHX, shown in Figure 5, is modeled using 25 nodes in each flow channel with each 5-nodal block representing the sulfuric acid decomposer (2 blocks), vaporizer (2 blocks), and a thermal recuperator (1 block). The following isotope exchange reactions are considered in the components of the SI process.

Figure 3

Figure 4

Figure 5

Figure 6

$$HT + H_2O \xrightarrow{\leftarrow} H_2 + HTO \tag{6}$$

$$HT + H_2SO_4 \xrightarrow{\leftarrow} H_2 + HTSO_4 \tag{7}$$

$$HT + HI \xrightarrow{\leftarrow} H_2 + TI \tag{8}$$

$$HTO + H_2SO_4 \xrightarrow{\leftarrow} H_2O + HTSO_4 \tag{9}$$

$$HTO + HI \xrightarrow{\leftarrow} H_2O + HT \tag{10}$$

The purification systems for tritium removal are installed at the position of the lowest temperature in each helium loop. The leakage of tritium from the system to the outside environment is also considered.

III.B.2. NGNP using HTE process

Figure 7 shows the nodalization scheme of the NGNP using the HTE process. Tritium release to the primary coolant is considered at a node expressed by No. (1) in Figure 7. Five heat exchangers (i.e., IHX, SHX, PHX-1, PHX-2, and PHX-3) are modeled by using 10 nodes in each flow channel, respectively. The isotope exchange reaction shown in Equation 6 is considered in the components of the HTE process. The purification systems for tritium removal are installed at the position of the lowest temperature in each helium loop. The leakage of tritium to the outside of the system is considered in each helium loop.

Figure 7

III.C. Boundary Conditions

III.C.1. Tritium Release Rate to Primary Coolant

Since there is no information yet available on the core design of the NGNP to estimate the tritium birth rate, the tritium birth rate and release rate to the primary coolant is estimated from the information on other HTGRs. The reported results[10,11,12] on tritium birth rate for several HTGRs are collected in Table 2. The tritium birth rates are normalized to the thermal MW by using each reactor power under 100% service factor. It is found that the tritium birth rates are the same order of magnitude for each HTGR. The discrepancy between HTGRs is caused by the differing periods of operation and evaluation conditions. The tritium birth rate at Fort St. Vrain during its first year of operation was 3.10 x 10^{11} Bq/y/MWt (8.38 Ci/y/MWt) and was the largest among the HTGRs in the United States. In this study, the tritium release rate of the NGNP is

Table II

assumed to be equal to Fort St. Vrain's tritium birth rate, and any possible reduction of this assumed tritium release rate caused by tritium trapping in the NGNP core graphite and the presence of the TRISO fuel coating in the NGNP fuel particles is ignored. For this work, the NGNP is assumed to resemble the Fort St. Vrain reactor, and so the tritium release rate into the primary coolant of the NGNP (600 MWt) is assumed to be 1.86 x 10^{14} Bq/y (5.030 x 10^3 Ci/y) as the base case.

III.C.2. Permeability of Heat Exchangers

The hydrogen permeability of Alloy 800 reported by Röhrig[13] is employed for the hydrogen permeability of the IHX and SHX. The tritium permeability of the IHX and SHX is defined as $\sqrt{3}$ times the ratio of the square root of the molecular weight of tritium to the molecular weight of hydrogen, and is smaller than the hydrogen permeability. Since three PHX's in the HTE process will be used under similar operating conditions as the steam generator used for the Peach Bottom HTGR, the tritium permeability of the as-received Alloy 800 of the Peach Bottom HTGR[14] will be directly applied to the tritium permeability of the PHXs. Though it is very likely the PHX in the SI process will be made of SiC or some other ceramic material due to the corrosiveness of the chemical environment in Section 2 of the SI process, this selection has not yet been codified in a final design, and so it is assumed that the permeability of the PHX is the same as for the as-received Alloy 800 for the base case. The values of the permeability used in this study are summarized in Table 3.

Table III

III.C.3. Helium Flowrate at the Purification System

The helium flow rate in the purification system should be designed by considering the ratio of the mass flow rate to the mass inventory in the loop, which is here designated the purification rate. The purification rate for the Peach Bottom HTGR and Fort St. Vrain were about 20%/h and 12%/h, respectively[15]. However, the purification rate for the NGNP has not yet been determined. As a starting point, the NGNP helium inventory was chosen to match the scaled helium inventories for the Peach Bottom HTGR and Fort St. Vrain, which were 3.96 kg/MWt and 3.69 kg/MWt, respectively[2,14]. Taking the average of these two numbers and multiplying by 600 MW gives a total helium inventory of 2293 kg for the NGNP. Assuming a purification rate of 12%/h as the NGNP base case, the purification flow rate for NGNP would be 275 kg/h. As for the secondary and tertiary helium loops, it is difficult to estimate a helium inventory, as the dimensions of these loops cannot be predicted without doing detailed design work. Therefore, a helium flow rate of 275 kg/h is also used as a base case for the purification system in these loops also. The removal efficiency of tritium by the purification system is assumed to be 100%.

III.C.4. Helium Leak Rate

The measured helium leak rate of the Peach Bottom HTGR[2] (loss fraction 9.95 x 10^{-3}/d) and the assumed primary helium inventory of 2293 kg are employed for the NGNP in order to evaluate the rate of tritium leakage from the primary loop to the outside as part of the general helium leakage. The leak rate of 9.95 x 10^{-3}/d is also used for the leakage of tritium from the secondary and ternary loops. It is probable that the inventory in the NGNP tertiary loop will be much greater than the inventory in the primary and secondary loops due to its long length, but the dimensions of the tertiary loop have not yet been fixed, and so the inventory of helium in the tertiary loop in this study is set equal to the inventory in the primary and secondary loops.

III.C.5. Equilibrium Constants of Isotope Exchange Reactions

The equilibrium constants of the isotope exchange reactions between HT and H_2O (Equation 6) and between HT and HI (Equation 8) are described in a previous paper[9]. However, the equilibrium constants of the isotope exchange reaction between HT, HTO, and H_2SO_4 (Equations 7 and 9) and between HTO and HI (Equation 10) are still not available. Thus, the unknown equilibrium constants are tentatively assumed to be equal to 1.0.

IV. RESULTS AND DISCUSSION

IV.A. NGNP Using SI Process

IV.A.1. Base Case

The distribution of tritium in the various parts of the NGNP in the base case is shown on a percentage basis of the total amount of tritium created in Figure 8. The calculated tritium concentration in the hydrogen product for the SI process is 1.23 Bq/ml at STP (33 nCi/L), which is much higher than the gaseous effluent limit. The tritium contamination is in the form of HT (99.4%) and entrained tritiated water HTO (0.6%).

Figure 8

The tritium concentrations in the gaseous process chemicals of the SI process were calculated, and all process streams exceed the gaseous effluent limit. The maximum concentration of tritium was found in the flash drum (S301), where the tritium contamination level was determined to be 18.95 Bq/ml at STP (512 nCi/L at STP). The forms of tritium in the flash drum (S301) are HT, HTO, and TI. The tritium concentrations in the liquid process chemicals in all components exceed the liquid effluent limit, with the highest calculated concentration found in the secondary O_2 knockout drum (S105) at 4.22×10^4 Bq/ml (0.0011 Ci/L). The forms of tritium in the secondary O_2 knockout drum (S105) are HT and HTO.

The tritium concentration in the tertiary helium is the largest of the three loops due to the lower pressure of the loop, and the concentration of tritium in this loop was calculated to be 1.77 Bq/ml at STP (47.8 nCi/L at STP), which greatly exceeds the gaseous effluent limit of 3.7×10^{-3} Bq/ml.

IV.A.2. Effect of Varying Isotope Exchange Equilibrium Constant

One of the undetermined parameters is the equilibrium constant of isotope exchange reaction between HT and H_2SO_4, and so the effect of varying the equilibrium constant of this reaction on the calculated tritium contamination levels in the hydrogen product and the SI process chemicals was evaluated. At steady state, the amount of tritium in the hydrogen product equals the tritium permeation rate into the SI process by conservation of mass, and so the tritium concentration in the hydrogen product is independent of the equilibrium constant.

The tritium concentration in gaseous process chemicals of some components in Section 2 of the SI process increases with an increasing equilibrium constant due to increasing concentrations of $HTSO_4$. Figure 9 shows such behavior for the calculated tritium concentration in the H_2SO_4 vaporizer 3 (H208B) above an equilibrium constant of about 0.1. In some components, the tritium contamination level stayed the same or was reduced due to an increasing equilibrium constant, and this behavior is demonstrated for the flash drum (S301). The tritium concentration in the flash drum (S301) remains the same in spite of varying the equilibrium constant.

Figure 9

This varied response to increasing the equilibrium constant is also evidenced in the liquid process chemicals. Figure 10 shows the tritium concentration in the liquid process chemicals in the secondary O_2 knockout drum (S105) and the H_2SO_4 vaporizer 2 (H208A). The tritium concentration in the secondary O_2 knockout drum (S105) stays almost constant with changes in the equilibrium constant, while the tritium concentration in the H_2SO_4 vaporizer 2 (H208A) increases with increasing equilibrium constant and becomes larger than the tritium concentration in the O_2 knockout drum (S105) above an equilibrium constant of about 20.

Figure 10

Since the helium coolant loops are outside and upstream of the hydrogen production plant, the concentration of tritium in the loops are unaffected by changes in the equilibrium constant between HT andH_2SO_4 in the SI process.

IV.A.3. Effect of Varying Heat Exchanger Permeability

Another undetermined parameter is the permeability of the PHX for the SI process. The material used for the PHX may be metal or ceramic, and different materials have significantly different tritium permeabilities. To address this unknown parameter, a range of tritium permeabilities for the PHX was examined to determine the effects of varying the tritium permeability on the concentration of tritium in the hydrogen product, the hydrogen plant, and the helium loops. It was determined that the effects of PHX permeability on the tritium contamination level in the hydrogen product are not significant above about 1 x 10^{-2} times the base case permeability (see Figure 11). However, the amount of tritium in the hydrogen product decreases when the PHX permeability is reduced to below 1 x 10^{-2} times the base case permeability. The tritium concentration becomes less than the gaseous effluent limit, the liquid effluent limit, and the drinking water limit below about 2 x 10^{-4}, 2 x 10^{-5}, and 5 x 10^{-6} times the base case, respectively. The effect of PHX permeability on the tritium concentration in the gaseous and liquid process chemicals is the same as the effect on the tritium concentration in the hydrogen product.

Figure 11

Figures 12 and 13 show the effect of varying the IHX permeability and the SHX permeability on the tritium concentration in the hydrogen product, respectively. The tritium concentration begins to decrease below 1 x 10^{-4} times the base case permeability for the IHX and about 1 x 10^{-2} times the base case permeability for the SHX. The difference between the IHX and the SHX response is due to the assumption that the IHX has more heat transfer area per unit of thermal energy transmitted, and due to the assumption that the heat transfer tubes are thinner in the IHX than in the SHX. Therefore, the permeability must be reduced to a greater degree in the IHX than in the SHX to increase the mass transfer resistance enough to make a difference.

Figure 12

Figure 13

Among the three heat exchangers, decreasing the permeability of the PHX is still the most effective means of reducing the concentration of tritium in the hydrogen product. If the PHX permeability is reduced to 1 x 10^{-6} times the base case with the IHX and SHX remaining at their base case permeabilities, the concentration of tritium in the hydrogen product can be reduced to less than the drinking water limit. If instead the PHX permeability is only reduced to 1 x 10^{-5} times its base case value, the permeability of the IHX would also need to be reduced to 7 x 10^{-6} times the base case permeability or the permeability SHX would need to be reduced to less than 6 x 10^{-4} times the base case to achieve the same result.

This effect is mirrored in the SI plant process chemicals, in that the tritium permeabilities of the IHX and PHX would need to be greatly reduced to cause a reduction in tritium contamination levels in the gaseous and liquid process chemicals of the hydrogen plant. For example, the SHX permeability must be less than 3 x 10^{-5} times the base case for a PHX permeability at 1 x 10^{-5} times the base case, or be less than 1 x 10^{-6} times the base case for a PHX permeability less than 1 x 10^{-4} times the base case to achieve tritium concentration levels in the gaseous process chemicals below the gaseous effluent limit.

Figures 14 and 15 show the effect of varying the IHX and SHX permeabilities on the tritium concentration in the tertiary helium loop while simultaneously manipulating the PHX permeability, respectively. The tritium concentration in the tertiary loop decreases with decreasing tritium permeability in the IHX and SHX. The tritium concentration in the tertiary loop remains above the gaseous effluent limit for a wide range of IHX permeabilities, but can be reduced to below the gaseous effluent limit by

Figure 14

Figure 15

lowering the SHX permeability to below 6 x 10^{-6} times the base case permeability. Decreasing the PHX permeability has the effect of slightly increasing the concentration of tritium in the tertiary helium loop, however, and so decreasing the PHX permeability is not an effective means of reducing the concentration of tritium in the tertiary loop.

IV.A.4. Effect of Varying Hydrogen Concentration in the Helium Loops

The tritium permeabilities of the IHX, SHX, and PHX can be reduced by injecting hydrogen into the loops. This reduction is accomplished by reducing the driving force for tritium movement across the heat exchangers. Figure 16 shows the relationship between the ratio of the hydrogen injection rate into the tertiary loop to the rate of hydrogen produced and the tritium concentration in the hydrogen product. Since the IHX and SHX are fairly permeable, it does not make a significance difference in the tritium concentration in the hydrogen product if hydrogen is injected into the secondary or primary loop instead of the tertiary loop. The tritium concentration in the hydrogen product decreases with increasing hydrogen injection rate. If the inherent permeability of the PHX is reduced and hydrogen is also injected into the helium loops, then it is possible to reduce the concentration of tritium in the hydrogen product to below the effluent limits and the drinking water limit.

Figure 16

Reductions in the tritium concentration in the hydrogen plant process chemicals can also be achieved by injecting hydrogen into the helium loops. The tritium concentration in the gaseous process chemicals of the flash drum (S301) becomes less than the gaseous effluent limit above 3 x 10^{-3} ppm, 0.3 ppm, 30 ppm, and 1000 ppm hydrogen in the primary helium loop at PHX permeability ratios (ratio of PHX permeability to the base case PHX permeability) of 1 x 10^{-5}, 1 x 10^{-4}, 1 x 10^{-3} and 1 x 10^{-2}, respectively. The tritium concentration in the liquid process chemicals can be reduced also to below the liquid effluent limit by increasing the hydrogen concentration in the helium loops and simultaneously reducing the tritium permeability of the PHX in a similar manner as was explained for the gaseous process chemicals.

Figure 17 shows the tritium concentration in the tertiary helium coolant as a function of the hydrogen injection rate into the helium loops. In no case can the tritium concentration in the tertiary loop be reduced by manipulating the hydrogen injection rate and the PHX permeability, and the tritium concentration in the tertiary loop remains significantly above the gaseous effluent limit for all conditions examined.

Figure 17

IV.A.5. Effect of Varying Core Tritium Release Rate

The tritium concentrations in the hydrogen product, gaseous and liquid process chemicals, and the tertiary helium loop decrease proportionally with a decrease of the tritium release rate from the nuclear core. Figure 18 shows the tritium concentration in the hydrogen product as a function of tritium release rate and PHX permeability. The tritium concentrations in the in the hydrogen product at 1 x 10^{-5} and 1 x 10^{-4} times the baseline PHX permeability becomes less than the tritium drinking water limit below about 0.2 and 3 x 10^{-3} times the baseline tritium release rate, respectively. For the condition of 1 x 10^{-3} times the baseline PHX permeability or greater permeability, the tritium release must be limited to less than 1 x 10^{-3} times the baseline release rate to limit the concentration of tritium in the hydrogen product to below the drinking water limit.

Figure 18

The tritium concentration in the tertiary helium becomes less than the gaseous effluent limit below about 3 x 10^{-3} times the release rate at the base case PHX permeability, as shown in Figure 19.

Figure 19

IV.A.6. Effect of Varying Helium Flowrate at the Purification System

The effect of varying the helium flow rate at the purification system on the tritium concentration in all helium loops is shown in Figure 20. The helium flow rate at the purification system in all loops must be increased by about 100 times just to decrease the tritium concentration in the loops by a factor of 0.1, so it appears increasing the flow rate of the purification system is not the most effective way to reduce the tritium concentration in the loops to below effluent limits.

Figure 20

The effect of varying the helium flow rate at the purification system on the tritium concentration in the gaseous and liquid process chemicals follows the same trend as for the tritium concentration in the loops -- large changes in purification system flow rates cause only small changes in the tritium concentration in the process chemicals, and this effect is not significant.

Figure 21 shows the effect of varying the helium flow rate at the purification system in all loops on the tritium concentration in the tertiary loop. Increasing the flow rates cause a decrease in the tritium concentration in the tertiary loop, but this effect is limited. Even if the purification flow rates are increased to the point where the helium flow rate at the purification systems matches the total loop flow rate, the tritium concentration in the tertiary loop is still above the gaseous effluent limit.

Figure 21

IV.B. NGNP Using HTE Process

IV.B.1. Base Case

The distribution of tritium in the various parts of the NGNP in the base case is shown on a percentage basis of the total amount of tritium created in Figure 22. The calculated tritium concentration in the gaseous hydrogen product, 2.67 x 10^{-3} Bq/ml (STP) (72.2 pCi/L), is slightly less than the gaseous effluent limit of 3.7 x 10^{-3} Bq/ml (STP) (100 pCi/L). The tritium concentration in a liquid hydrogen product generated by this process, assuming a liquid hydrogen density of 0.077 g/ml, was calculated to be 2.30 Bq/ml (62.2 pCi/L) and is less than the liquid effluent limit of 37 Bq/ml (1.00 μCi/L). However, this is still greater than the established limit for tritium in drinking water, 0.74 Bq/ml (20 nCi/L). The tritium concentration in the gaseous process chemicals in the sweep line of the electrolyzer showed the highest concentration in the process at 6.44 x 10^{-3} Bq/ml (STP) (174 pCi/L), which is larger than the gaseous effluent limit. The tritium concentration in the liquid process chemicals in the H_2/H_2O knockout tank in the main process line was calculated to be 20.3 Bq/ml (0.549 μCi/L), which is still lower than the liquid effluent limit. The tritium concentration in the tertiary helium loop was calculated to be 4.85 Bq/ml (STP) (0.131 μCi/L) and is larger than the gaseous effluent limit by a factor of more than 1000.

IV.B.2. Effect of Varying Heat Exchanger Permeability

Figure 23

Figure 23 shows the effect of varying the tritium permeability of the IHX, SHX, and PHX on the tritium concentration in the hydrogen product. In regard to the IHX, changing the permeability has no significant effect on the tritium concentration in the hydrogen product for permeabilities greater than 0.01 times the base case permeability. The tritium concentration in the hydrogen product begins to decrease gradually with a reduction in IHX permeability below about 0.1 times the base case and does not fall below the gaseous effluent limit until the IHX permeability has been reduced to about 1 x 10^{-3} times the base case. The tritium concentration in the hydrogen product is more strongly affected by changes in the permeability of the SHX and PHX, and significant reductions of the tritium concentration in the hydrogen product can be made by reducing the permeability of the SHX and PHX even near the base case permeabilities. An explanation for the differences in sensitivity may be due to the differences in total heat exchanger area and heat exchanger tube thickness for the IHX, SHX, and PHX. The heat transfer area per MWt heat exchange is approximately 2.2 times larger than the heat transfer area per MWt of the SHX, and the thickness of the IHX tubing is about 0.34 times the thickness assumed for the SHX. Thus, the effect of varying the IHX permeability is swamped by the effect of employing a larger heat exchanger area and thinner tubing for tritium permeabilities greater than 0.01 times the base case. Since the PHX is assumed to have similar characteristics as the SHX, it behaves similarly to the SHX in comparison to the IHX. Changing the permeability of the SHX and PHX does have an effect on the tritium concentration in the hydrogen product, and the tritium concentration in the hydrogen product can be reduced to below the gaseous effluent limit by reducing the tritium permeability in the SHX to below 0.08 times the base case, and reducing the permeability of the PHX to below 0.04 times the base case.

The tritium concentration in the gaseous process chemicals of the electrolyzer also varies with changes in the permeability of the IHX, SHX, and PHX in a similar manner as in the gaseous hydrogen product. The gaseous tritium concentration in the electrolyzer becomes less than the gaseous effluent limit below about 6 x 10^{-3} times the IHX base case permeability, below about 0.3 times the SHX base case permeability, and about 0.2 times the PHX base case permeability.

Figures 24, 25, and 26 show the effect of varying the permeability of the IHX, SHX, and PHX on the tritium concentration in the tertiary loop. The tritium concentration in the tertiary loop decreases with decreasing IHX and SHX permeability, and becomes less than the gaseous effluent limit below about 2 x 10^{-8} times the IHX base case permeability and about 3 x 10^{-6} times the SHX base case permeability. On the other hand, tritium concentration in the tertiary loop increases slightly with decreasing PHX permeability, and it is not possible to reduce the tritium concentration in the tertiary loop to below the gaseous effluent limit by changing the permeability of the PHX, at least within the physical limits studied here.

Figure 24

Figure 25

Figure 26

IV.B.3. Effect of Varying Heat Exchanger Permeability

Figure 27 shows the effect of varying the helium flow rate at the purification system on the tritium concentration in the hydrogen product. The lower horizontal axis indicates the ratio of the helium flow rate at the purification system to the total loop helium inventory. The upper horizontal axis indicates the ratio of helium flow rate at the

Figure 27

purification system to the helium circulation flow rate in each loop. The latter number should be less than 1.0 because having a flow rate through the purification system that is in excess of the total helium flow rate through a loop is unreasonable. Note that the range of the upper axis for the tertiary loop is different from that of the primary and secondary loops because the helium circulation flow rate for the tertiary loop is about 0.1 times the flow rates for the primary and secondary loops. As expected, the tritium concentration in the hydrogen product is reduced by increasing the purification system flow rates in each loop. There is no significant difference in the tritium concentrations in the three loops due to differences in the loop purification systems. In order to decrease the tritium concentration in the hydrogen product to less than the drinking water tritium limit, the ratio of helium flow rate at the purification system to the helium inventory in the primary, secondary, and tertiary loops must be increased to more than 3 h^{-1}, 3 h^{-1}, and 4 h^{-1}, respectively. Increasing the helium flow rate at all purification systems simultaneously is more effective than increasing the purification rate in any one system, and the tritium concentration ini the hydrogen product can be reduced to below the drinking water limit at a ratio of 0.6 h^{-1}. However, the effect of increasing the helium flow rate at the purification system does not appear to have a strong effect on the concentration in the hydrogen product, as the tritium concentration decreases by a factor of only 0.19 with an increase in helium flow rate at all purification systems by a factor of 10.

Regardless of the helium flow rate in the tritium purification system, the highest concentrations of tritium are found in the gaseous process chemicals of the electrolyzer. The relation of the tritium concentration in the gaseous chemicals to the purification flow rate is similar to that found with the hydrogen product, and the concentration of tritium is only weakly dependent upon the purification flow rate. The tritium concentration becomes less than the gaseous effluent limit when the helium flow rate in each purification system is more than 0.7 h^{-1} separately, or the helium flow rates in all three are set to larger than 0.3 h^{-1} simultaneously.

Figure 28

Figure 28 shows the tritium concentration calculated in the tertiary helium loop in response to changes in the purification flow rate. From the figure, it is clear that it is impossible to decrease the tritium concentration in the tertiary helium to less than the gaseous effluent limit by increasing the helium flow rate in only one helium loop. The helium flow rate must be increased to above 40 h^{-1} in all three loops in order to just reach the gaseous effluent limit. However, this means that the helium flow rate in the purification system must be increased to more than 300 times the base case flow rate, and this would result in greatly increased capital and operating costs in the purification system in comparison to the base case.

IV.B.4. Effect of Varying Reactor Outlet Temperature

The HTE process can be operated with the same hydrogen production capacity at lower reactor temperatures as at higher reactor temperatures by installing a recuperator at the inlet of the electrolyzer to recover some of the heat lost in the hydrogen and oxygen products. Inlet temperature can also be supplemented with electrical heating, or by operating the HTE cells beyond the thermal-neutral point so that they operate less efficiently and generate more heat than they absorb performing the endothermic water-splitting reaction. A range of reactor outlet temperatures is possible, and so the effects of varying the reactor outlet temperature from 823 K to 1173 K (550 to 900°C) on tritium concentrations in the hydrogen product were examined. In this calculation, the temperature of the heat transfer surfaces was changed in direct proportion to the reactor outlet temperature. The tritium concentration in the hydrogen product decreases with decreasing reactor outlet temperature due to a decrease in the tritium permeation rate, and the tritium concentration falls below the drinking water limit below about 990 K. With the decrease in reactor outlet temperature from the base case of 1173 K to 823 K, the tritium concentration in the hydrogen product decreases by a factor of 0.08.

The tritium concentration in the gaseous process chemicals of the electrolyzer are also affected by reducing the reactor outlet temperature, and fall below the gaseous effluent limit at a reactor outlet temperature below 1070 K.

The tritium concentration in the tertiary helium decreases slightly with the decrease in temperature, but remains significantly above the gaseous effluent limit even at the lowest reactor outlet temperature examined, 823 K. The calculated amount of tritium in the tertiary loop at this temperature was 2.0 Bq/ml.

IV.B.5. Effect of Varying Reactor Outlet Temperature

One possible countermeasure to decrease the tritium concentration in the hydrogen product and the hydrogen plant chemicals is to inject excess hydrogen into the helium coolants to decrease the tritium permeation rate. Figure 29 shows the effects of hydrogen injection in the helium coolants on the tritium concentration in the hydrogen product. The horizontal axis indicates the ratio of the hydrogen injection rate to the hydrogen production rate, which is here assumed to be 7.5 x 10^4 m^3/h (STP). The tritium

Figure 29

concentration in the hydrogen product decreases with increasing hydrogen injection rates. Injecting hydrogen into the primary and secondary helium loops is more effective than injecting hydrogen into the tertiary loop. However, there is no significant difference between choosing to inject hydrogen into either the primary or secondary loops, as hydrogen diffuses rapidly between the two loops at the assumed operating temperature. According to the calculations, the tritium concentration in the hydrogen product becomes less than the drinking water limit with hydrogen injection rates above 0.002% in the primary helium loop, 0.002% in the secondary helium loop, or 0.004% in the tertiary helium loop. Increasing the hydrogen concentration in the helium coolant results in a decrease in the oxygen partial pressure, and so a co-injection of oxygen or water along with the hydrogen should be performed to maintain existing surface chemistries. Care must be taken to manage the injection of water or oxygen into the loops, however, because of the possible danger of exposing the graphite core of the nuclear reactor to these chemically reactive substances. Therefore, it is perhaps safest to consider co-injection of hydrogen, oxygen and/or water vapor into the secondary loop rather than the primary loop.

The tritium concentration in the gaseous process chemicals of the electrolyzer also decreases with increasing hydrogen injection rates. The tritium concentration in the gaseous process chemicals becomes less than the gaseous effluent limit above injection rates of 0.0005% in the primary helium loop, 0.0005% in the secondary helium loop, or 0.001% in the tertiary helium loop.

Figure 30 shows the effects of varying the hydrogen injection rate on the tritium concentration in the tertiary helium loop. The tritium concentration in the tertiary

Figure 30

helium loop decreases with increasing hydrogen injection rates, but this effect is limited, and lowering the tritium concentration in the tertiary loop is not possible even with a hydrogen injection rate of 1% in the loops.

IV.C. General Comparisons of Results

This larger amount of tritium contamination in the NGNP using the SI process is due to several causes. One of the reasons for this contamination level is the amount of thermal energy, and, correspondingly, the amount of heat exchanger surface area, needed to power the hydrogen production process. The SI process uses only thermal energy to split water, whereas the HTE process uses mostly electricity (approximately 75-80%) and some thermal energy to power the hydrogen production process. Since the SI process requires larger heat exchangers, the larger heat exchanger surface area allows for increased tritium movement through the heat exchanger surfaces. Another reason is that that the HTE process uses some high-temperature thermal energy to heat the electrolyzer sweep gas, and this acts as a tritium removal mechanism from the HTE process. A third reason is that the hydrogen produced in the HTE process can diffuse backwards through the high-temperature heat exchanger surfaces and into the tertiary helium loop. Hydrogen injection, as was established earlier, reduces the tritium permeability through the heat exchanger surfaces, and so the HTE process operates with an uncontrolled hydrogen injection mechanism as part of its normal operation. The SI process, on the other hand, does not allow for backwards diffusion of hydrogen into the loop because the hydrogen is produced in a relatively lower temperature step, and so cannot benefit from this side effect in terms of reducing the tritium contamination in the hydrogen product. There is some small amount of hydrogen released from the materials in the nuclear reactor core (Peach Bottom HTGR reported a hydrogen concentration of 10 ppm in the primary coolant[15], but the effects of any possible release of hydrogen from the core materials on the tritium permeabilities downstream from the core are neglected here due to lack of NGNP design data.

V. CONCLUSIONS

The tritium concentrations in the NGNP using the SI and HTE processes were evaluated using the THYTAN numerical analysis code.

The tritium concentrations in the hydrogen product and process chemicals of the SI process are potentially much larger than those found in the HTE process under the baseline assumptions. The tritium concentration in the hydrogen product and the SI process components are significantly affected by PHX permeability and the hydrogen concentration in the helium loops. The concentration of tritium in the SI process chemicals depends strongly on the isotope exchange behavior of tritium and hydrogen between HT and H_2SO_4, and so reliable isotope exchange equilibrium data is needed to generate more accurate predictions. Increasing the purification system flow rate and injecting hydrogen into the helium loops will help to reduce tritium concentrations in tertiary helium loop used to power an SI plant, but it is impossible to reduce the tritium concentration in the tertiary loop to below the gaseous effluent limit simply by manipulating these parameters. The most effective means of reducing tritium concentrations in the tertiary loop and SI plant is to reduce the tritium release rate from the core and to reduce the SHX permeability.

The tritium concentration in the liquid process chemicals in the HTE components is less than the liquid effluent limit under the baseline assumptions. The tritium concentration in the hydrogen product and the gaseous process chemicals of the HTE process is slightly larger than the drinking water limit and the gaseous effluent limit under the baseline assumptions. The tritium concentration in the hydrogen product and the gaseous process chemicals of the HTE process can be reduced to below the applicable limits by employing increased helium flow rates at the helium loop purification system, decreasing the reactor outlet temperature, and injecting hydrogen into the helium loops. The tritium concentration in the tertiary helium loop is higher than the gaseous effluent limit, and can be reduced to below the gaseous effluent limit by reducing the tritium release rate from the core, decreasing the heat exchanger permeabilities through hydrogen injection and/or materials changes, and employing larger purification system flow rates.

Controlling the tritium concentration in the tertiary helium loop is more difficult and a reduction of the tritium concentration in this loop to below the gaseous effluent limit cannot be achieved simply by decreasing the purification system flow rates. The most effective means to reduce the tritium concentration in the tertiary loop is to reduce the permeability of the SHX.

Overall, the best method for controlling and limiting the tritium concentration in the hydrogen product is to not make it in the first place, and more work is needed to understand the latest NGNP designs and the effects of design decisions on the tritium generation rate.

ACKNOWLEDGEMENTS

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REFERENCES

1. N. FORSYTH, “Tritium Production and Distribution in High Temperature Gas Cooled Reactors Part 1: Tritium Production, Migration and Removal in the Dragon Reactor Between Core 5 Charge III and Core 1 Charge IV,” DP-REPORT-799, O.E.C.D. High Temperature Reactor Project DRAGON (June 1972).

2. R.P. WICHNER and F.F. DYER, “Distribution and Transport of Tritium in the Peach Bottom HTGR,” ORNL-5497, Oak Ridge National Laboratory (Aug. 1979).

3. W. STEINWARZ, H.D. RÖHRIG, and R. NIEDER, “Tritium Behavior in an HTR-System Based on AVR-Experience,” International Atomic Energy Agency, Vienna (Austria), IWGGCR-2, 153 (1980).

4. B.W. GAINEY, “A Review of Tritium Behavior in HTGR Systems,” GA-A13461, General Atomic Company (April 1976).

5. S. KUBO, H. NAKAJIMA, S. KASAHARA, S. HIGASHI, T. MASAKI, H. ABE and K. ONUKI, “A Demonstration Study on a Closed-Cycle Hydrogen Production by the Thermochemical Water-Splitting Iodine-Sulfur Process,” *Nucl. Eng. Des.*, **233**, 347 (2004).

6. M.B. RICHARDS, A.S. SHENOY, L.C. BROWN, R.T. BUCKINGHAM, E.A. HARVEGO, K.L. PEDDICORD, S.M.M. REZA, and J.P. COUPEY, "H2-MHR Pre-Conceptual Design Report: SI-Based Plant," GA-A25401, General Atomics (Apr. 2006).

7. C.H. OH C.B. DAVIS, J. HAN, R. BARNER, S.R. SHERMAN, R. VILIM, and W.J. LEE, "HyPEP FY06 Report: Models and Methods," Idaho National Laboratory, INL/EXT-06-11725 (Sept. 2006).

8. C.H. OH, E.S. KIM, S.R. SHERMAN, and R. VILIM, "HyPEP FY07 Report: System Integration Model Development," Idaho National Laboratory, INL/EXT-07-12470 (Apr. 2007).

9. H. OHASHI, N. SAKABA, T. NISHIHARA, Y. INAGAKI and K. KUMITOMI, "Numerical Study on Tritium Behavior with Isotope Exchange Reactions in Thermochemical Water-Splitting Iodine-Sulfur Process," *J. Nucl. Sci. Technol.*, Vol. 44, No. 11, p. 1407-1420 (2007).

10. E.L. COMPERE, S.H. FREID, and C.W. NESTOR, "Distribution and Release of Tritium in High-Temperature Gas-Cooled Reactors as a Function of Design, Operational, and Material Parameters," ORNL-TM-4303, Oak Ridge National Laboratory (June 1974).

11. W. STEINWARZ, D. STÖVER, R. HECKER and W. THIELE, “Distribution of Tritium in a Nuclear Process Heat Plant with HTR,” *Nucl. Eng. & Des.*, **78**, 267 (1984).

12. N. FORSYTH, “Tritium Production and Distribution in High Temperature Gas-Cooled Reactors: Tritium Generation, Retention, Distribution, and Environmental Release in a National 1,500 MW(th) HTR,” DP-REPORT-905, O.E.C.D. High Temperature Reactor Project DRAGON (Oct. 1974).

13. H.D. RÖHRIG, R. HECKER, J. BLUMENSAAT and J. SCHAEFER, “Studies on the Permeation of Hydrogen and Tritium in Nuclear Process Heat Installations,” *Nucl. Eng. & Des.*, **34**, 157 (1975).

14. L. YANG, W.A. BAUGH, and D.N.L. BALDWIN, “Study of Tritium Permeation Through Peach Bottom Steam Generator Tubes,” GA-A14376, General Atomics (June 1977).

15. R.D. BURNETTE and D.N.L. BALDWIN, “Primary Coolant Chemistry of the Peach Bottom and Fort St. Vrain High-Temperature Gas-Cooled Reactors,” GA-A16163, General Atomic Company, (Nov. 1980).

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Figure 30: Effect of varying hydrogen injection rate in the tertiary helium loop on concentration of tritium in the tertiary helium loop for an NGNP using the HTE process.

TABLE I

Tentative Major Specifications of the NGNP

Item	Value
Reactor power	600 MWt
Heat transfer rate to hydrogen plant	50 MWt
Primary Coolant	
Reactor outlet temperature	900°C
Reactor inlet temperature	495°C
Flowrate	289 kg/s
Pressure	7.0 MPa
Secondary Coolant	
IHX outlet temperature	885°C
IHX inlet temperature	480°C
Flowrate	289 kg/s
Pressure	7.0 MPa
Tertiary Coolant	
SHX outlet temperature	875°C
SHX inlet temperature	522°C
Flowrate	27.5 kg/s
Pressure	2.0 MPa
Hydrogen Production Rate	
SI Process	1.1×10^4 m^3 (STP)/h
HTE Process	7.5×10^4 m^3 (STP)/h

TABLE II

Calculation Results of Tritium Birth Rates in HTGR

Country	U.S.A.			Germany	U.K.
Reactor	Peach Bottom	Fort St. Vrain	3000MWt HTGR	PNP-500 MWt-plant	1500MWt HTR
Power (MWt)	66.5[a]	842[b]	3000	500	1500
Period	1550 days	1st year	1st year	1st year	After 5 or 6 years
Annual birth rate per MW (Bq/y/MWt)	2.9×10^{11}	3.10×10^{11}	2.84×10^{11}	3.83×10^{11}	4.28×10^{11}
Reference #	2	10	4	11	12

[a] Average power based on rated power of 115 MWt and equivalent full-power days of 897 during 1550 days of operation.

[b] Based on calculations using the reference.

TABLE III

Boundary Condition of Permeability of Heat Exchangers in the NGNP

Heat Exchanger	Pre-exponential factor of permeability [m^3 (STP)/m·s·$Pa^{0.5}$]		Activation Energy [J/mol]
	Tritium	Hydrogen	
IHX	1.36×10^{-8}	2.35×10^{-8}	7.40×10^{4}
SHX	1.36×10^{-8}	2.35×10^{-8}	7.40×10^{4}
PHX	1.36×10^{-10}	2.36×10^{-10}	5.35×10^{4}

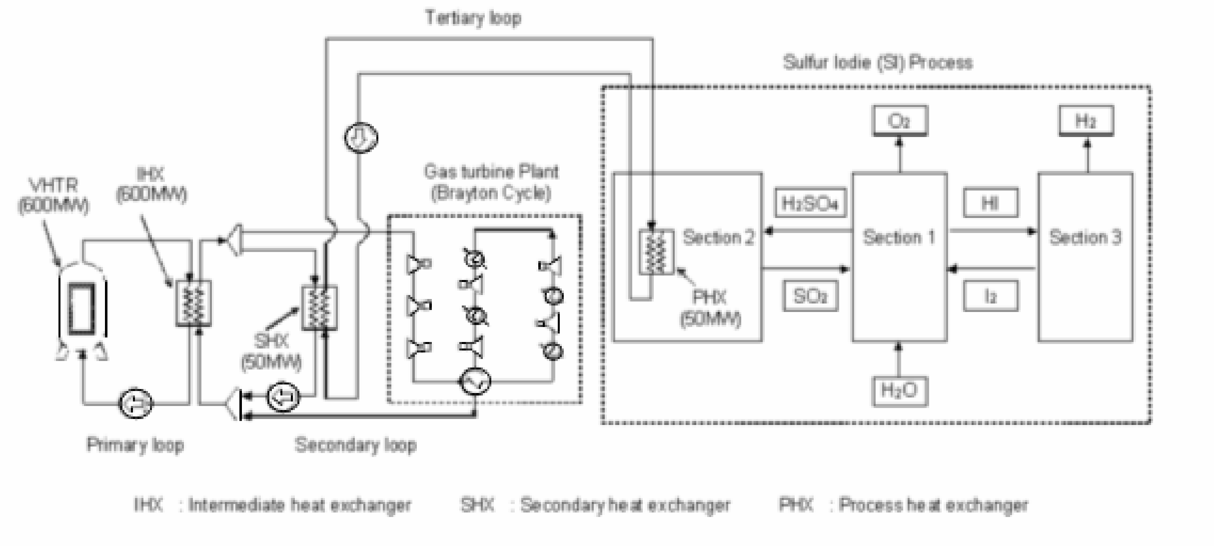

Fig. 1. Tentative flow scheme of NGNP using SI process.

Title: Tritium Movement and Accumulation in the NGNP System Interface and Hydrogen Plant
Authors: Hirofumi Ohashi and Steven R. Sherman

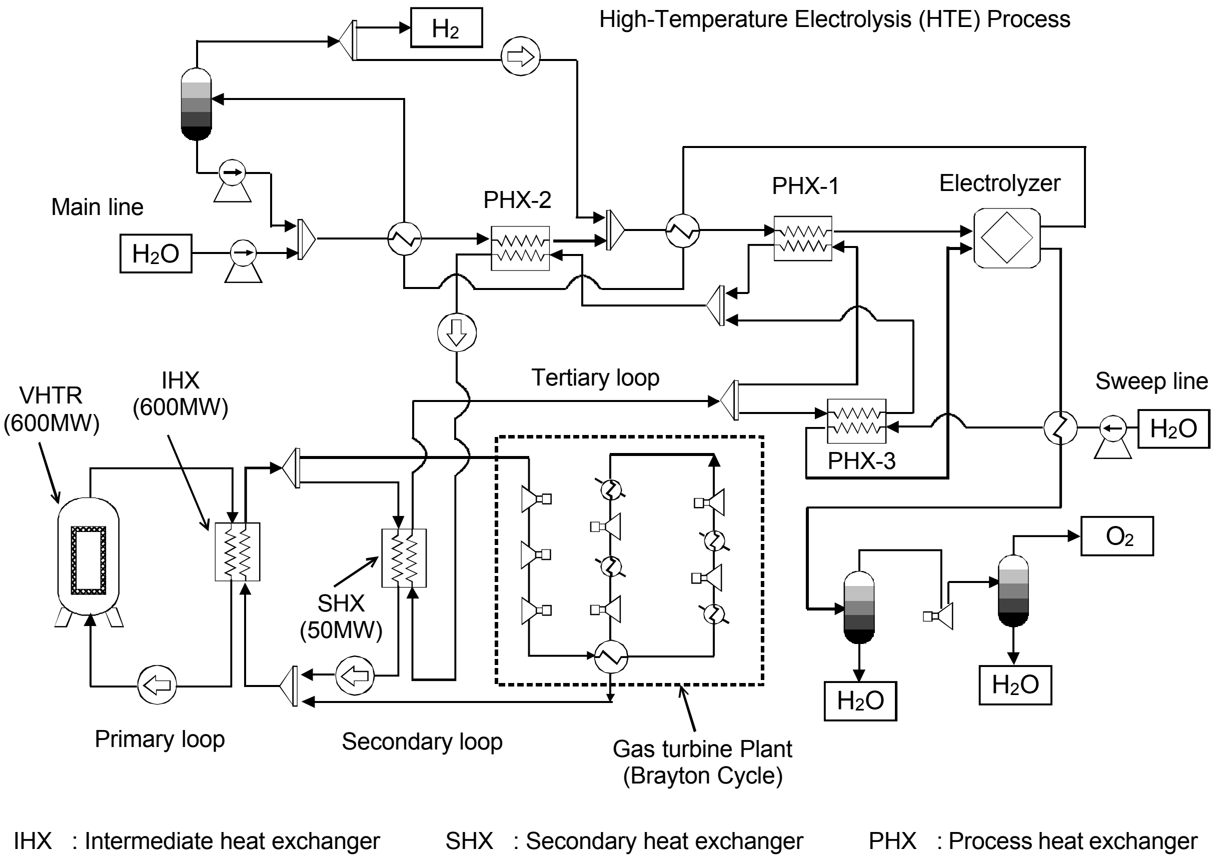

Fig. 2. Tentative flow scheme of NGNP Using HTE process.

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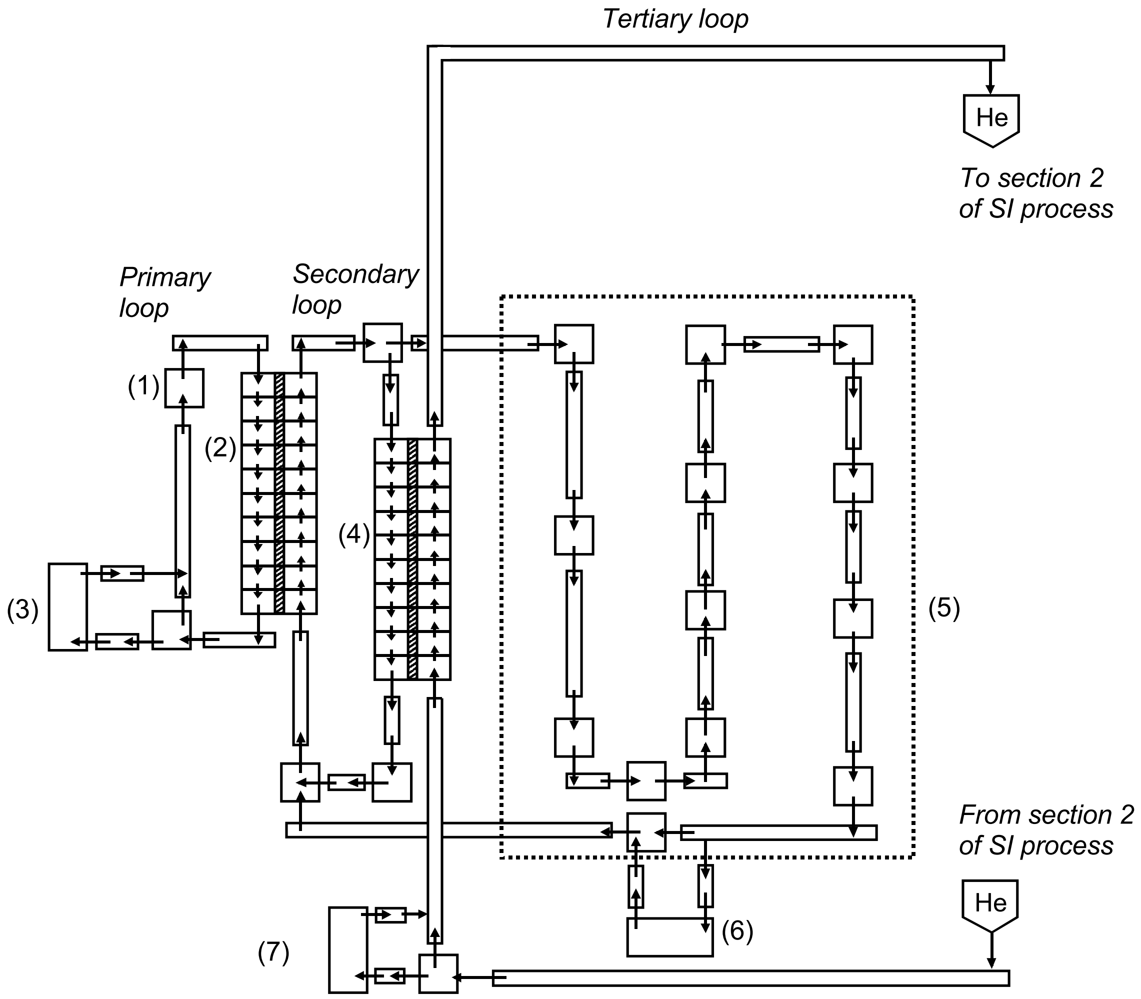
Authors: Hirofumi Ohashi and Steven R. Sherman



(1) Core
(2) Intermediate heat exchanger (IHX)
(3) Primary purification system
(4) Secondary heat exchanger (SHX)
(5) Gas turbine plant
(6) Secondary purification system
(7) Tertiary purification sysytem

Fig. 3. Nodalization scheme of helium loops of NGNP using SI process.

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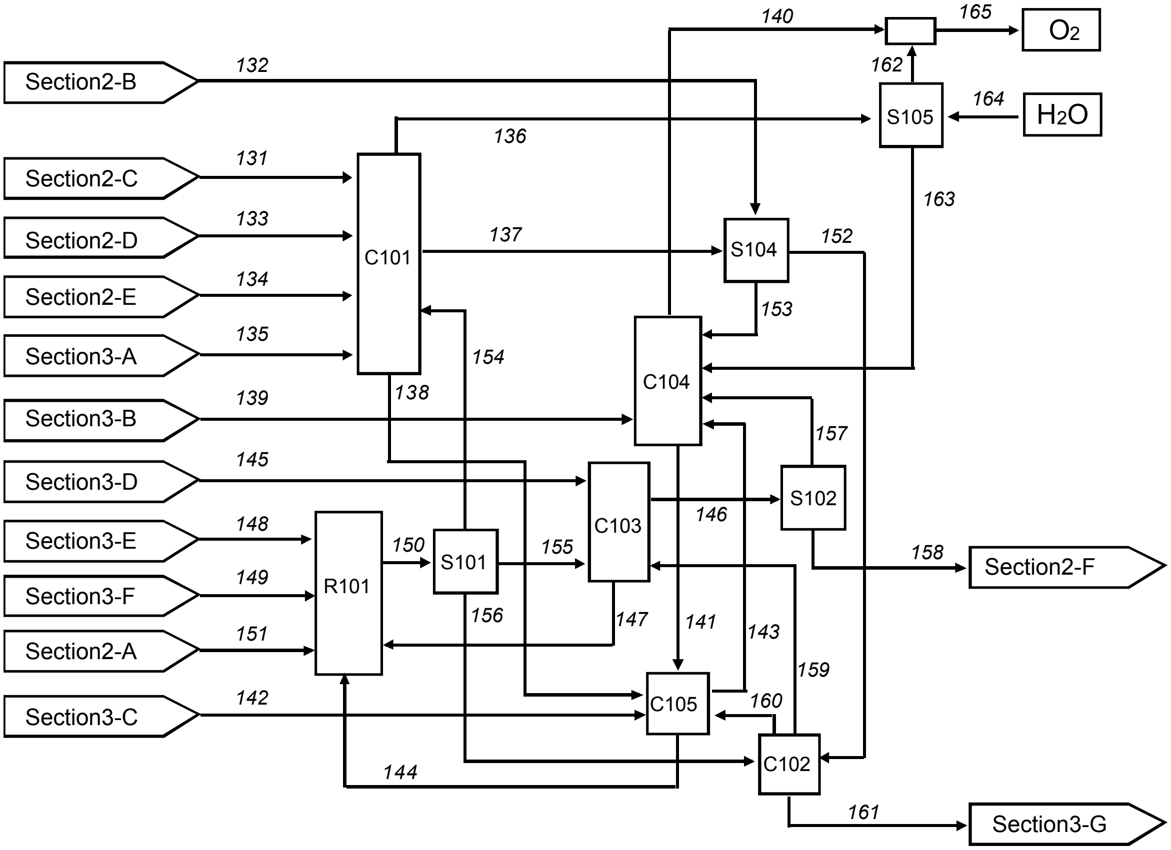

R101 : Reactor
C101 : #1 O_2 scrubber
C102 : HIx/SO_2 stripper
C103 : H_2SO_4 boost reactor
C104 : #2 O_2 scrubber

C105 : SO_2 adsorber
S101 : 3-phase separator
S102 : Flash drum
S104 : Primary O_2 knockout drum
S105 : Secondary O_2 knockout drum

Fig. 4. Nodalization scheme of Section 1 in SI process.

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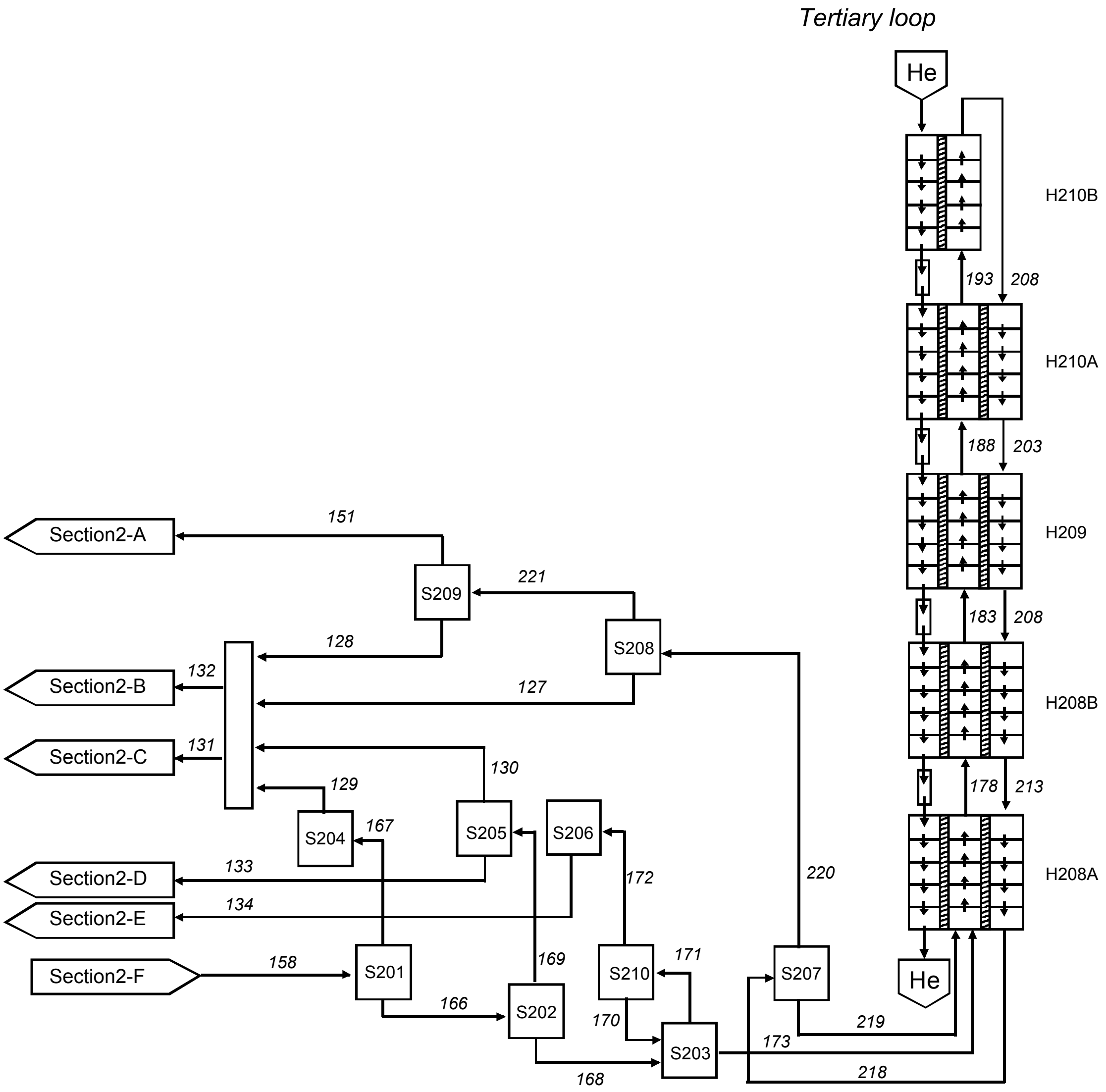

H208A : Vaporizer 2
H208B : Vaporizer 3
H209 : Recuperator 2
H210A : Decomposer 1
H210B : Decomposer 2
S201 : Section feed flash drum
S202 : Section feed bottoms flash drum
S203 : S202 bottoms flash drum
S204 : S201 overhead product flash vessel
S205 : S202 overhead product flash vessel
S206 : S210 overhead product flash vessel
S207 : Decomposer loop flash vessel
S208 : Decomposer loop vapor product flash vessel
S209 : Decomposer loop vapor product flash vessel
S210 : S203 overhead product flash vessel

Fig. 5. Nodalization scheme of Section 2 of SI process.

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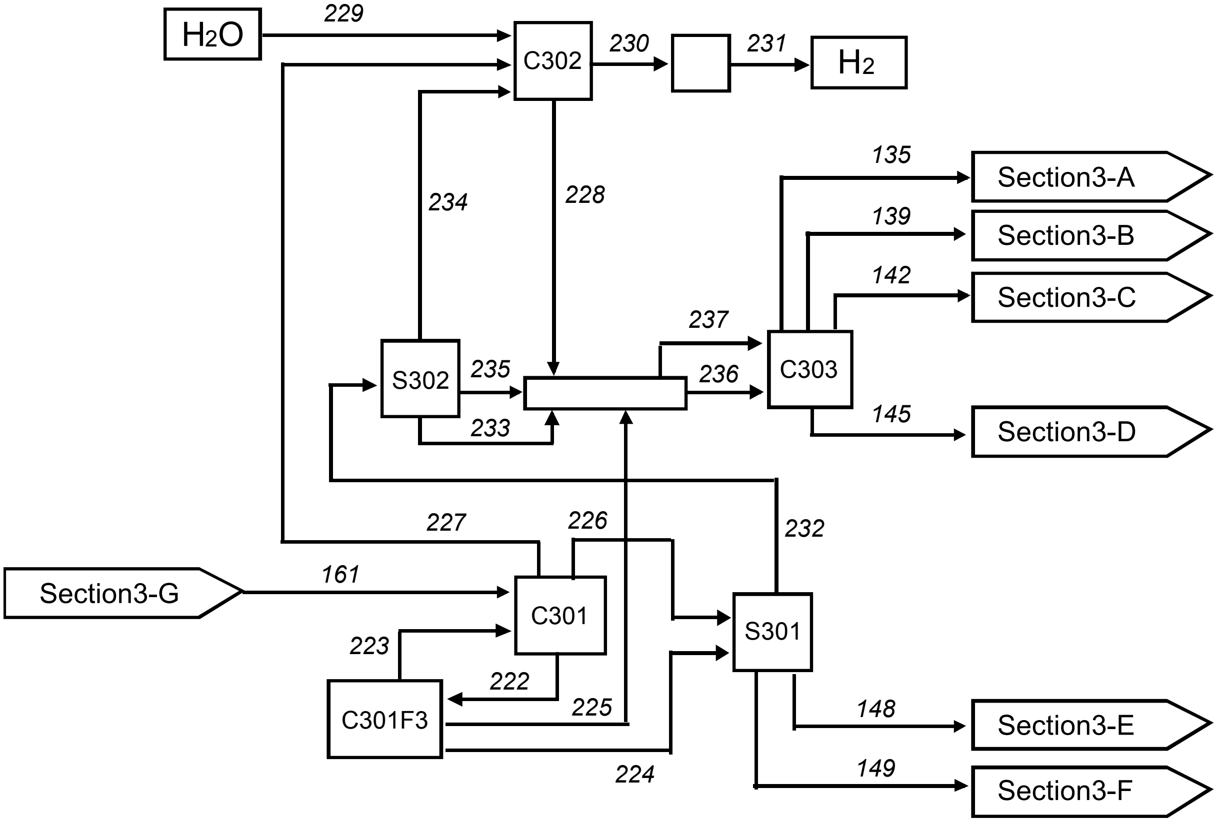

C301 : Reactive still
C301F3 : Auxiliary reboiler
C302 : H_2 product scrubber
C303 : I_2 scrubber
S301 : Flash drum
S302 : Flash drum

Fig. 6. Nodalization scheme of Section 3 of the SI process.

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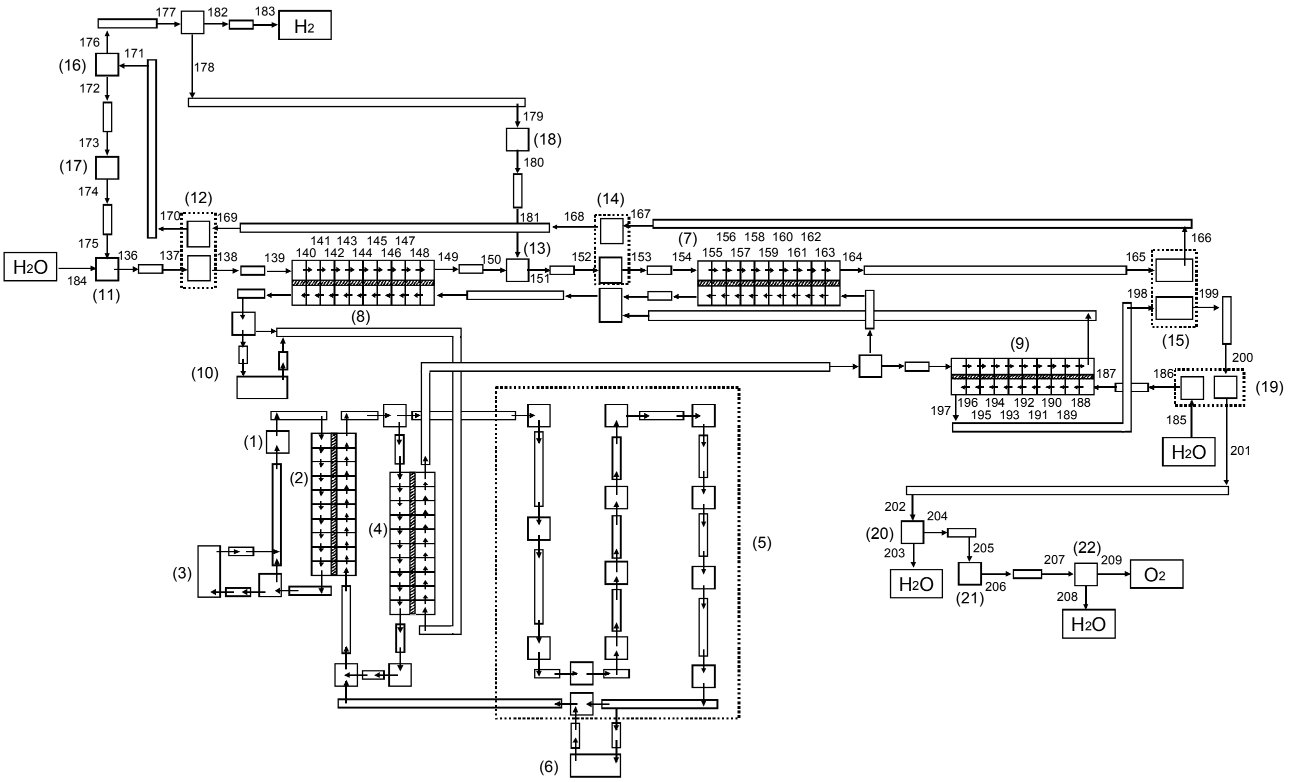

(1) Core
(2) Intermediate heat exchanger (IHX)
(3) Primary purification system
(4) Secondary heat exchanger (SHX)
(5) Gas turbine plant
(6) Secondary purification system
(7) Process heat exchanger - 1 (PHX-1)
(8) Process heat exchanger - 2 (PHX-2)
(9) Process heat exchanger - 3 (PHX-3)
(10) Tertiary purification sysytem
(11) Mixer
(12) Recuperator-1
(13) H2/H2O mixer
(14) Recuperator-2
(15) Electrolyzer
(16) H2/H2O knockout tank
(17) Recycle pump
(18) H2 recirculator
(19) Recuperator-3
(20) O2/H2O knockout tank
(21) O2/H2O expander
(22) Separator

Figure 7. Nodalization scheme of NGNP using HTE process.

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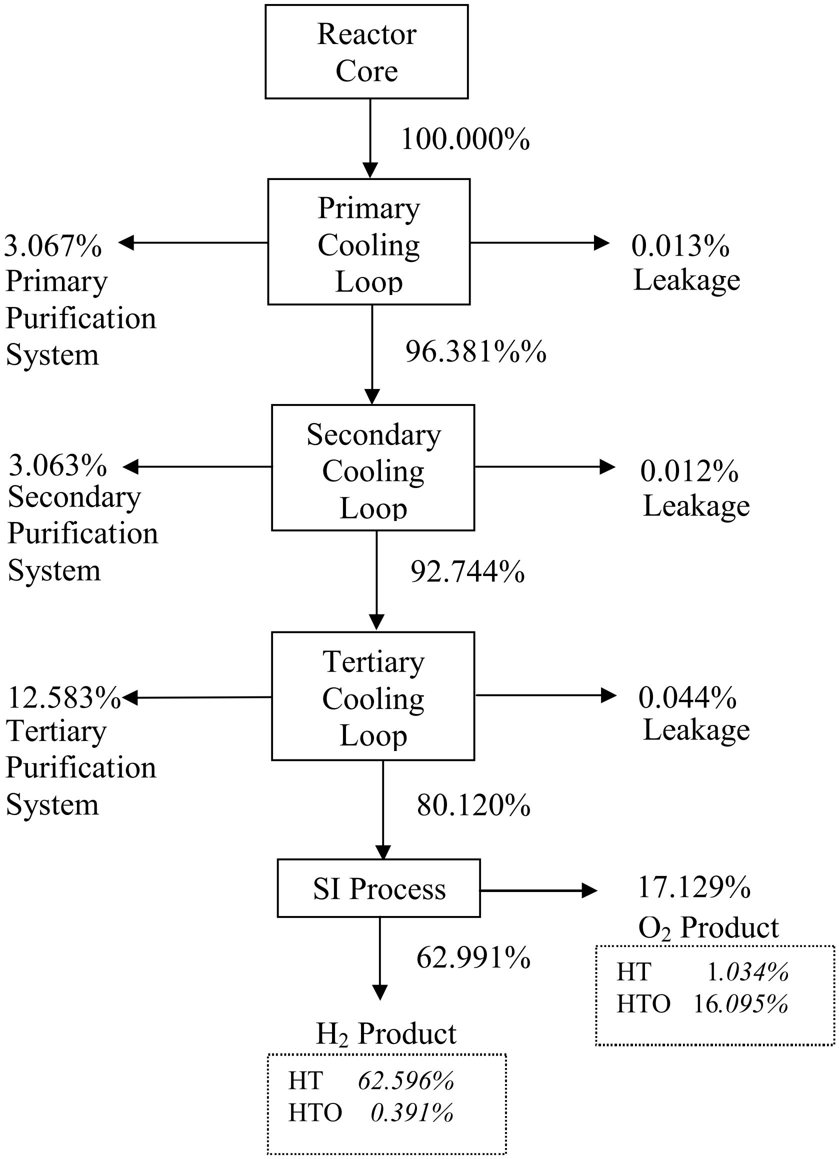

Figure 8: Distribution of tritium in SI process for base case.

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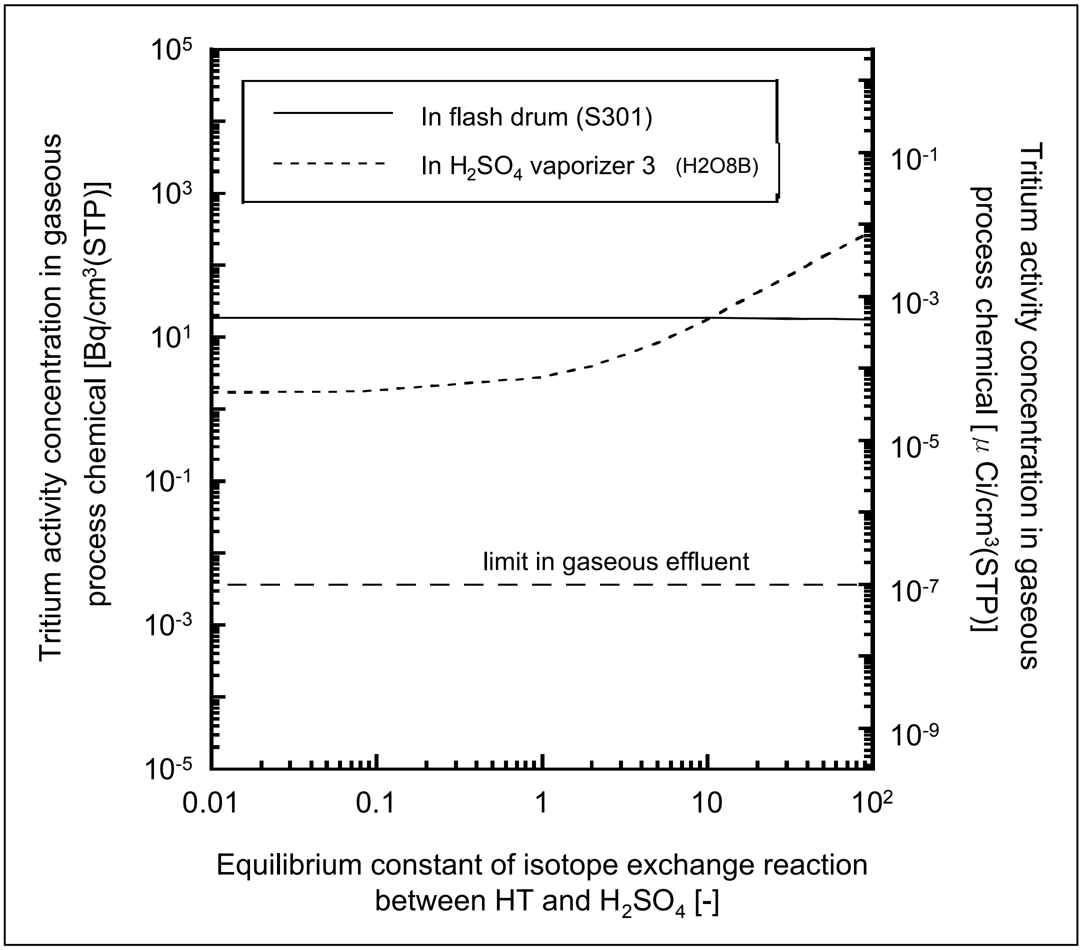

Figure 9: Effect of varying the HT/$HTSO_4$ isotope exchange equilibrium constant on tritium concentration in the gaseous process chemicals for an NGNP using the SI process.

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Authors: Hirofumi Ohashi and Steven R. Sherman

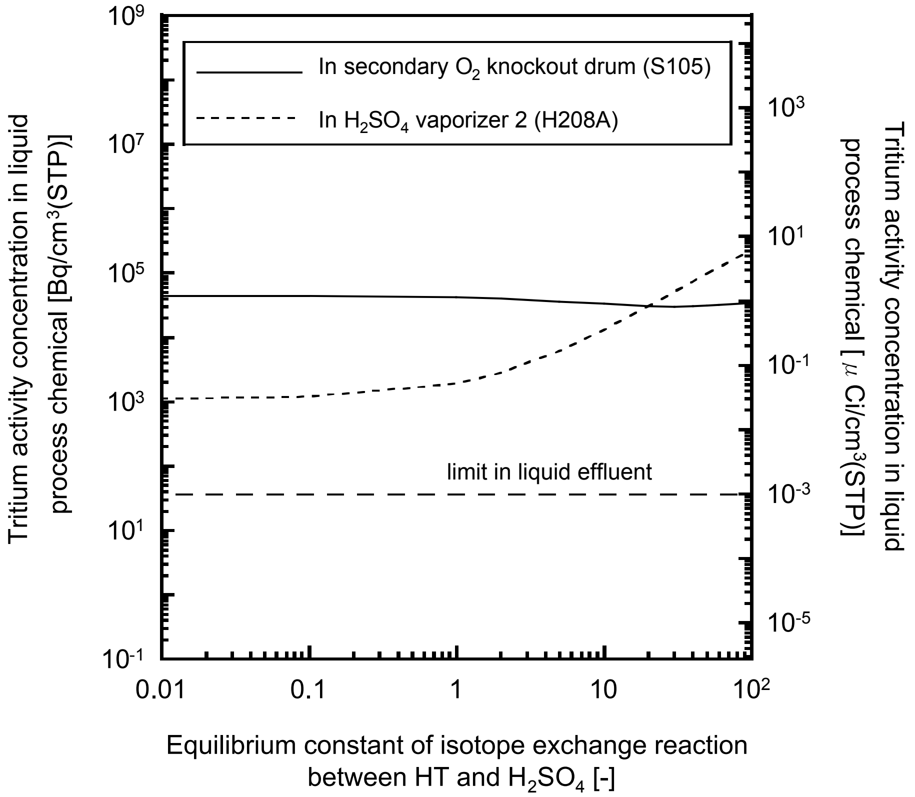

Figure 10: Effect of varying the HT/$HTSO_4$ isotope exchange equilibrium constant on tritium concentration in the liquid process chemicals for an NGNP using the SI process.

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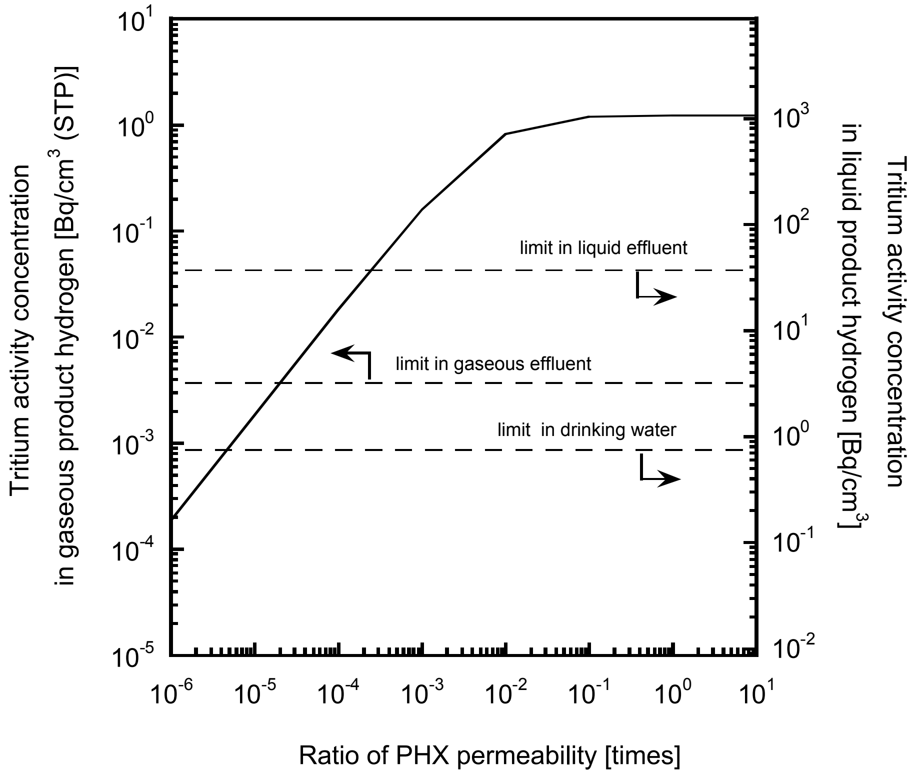

Figure 11: Effect of varying the PHX permeability on tritium concentration in the hydrogen product for an NGNP using the SI process.

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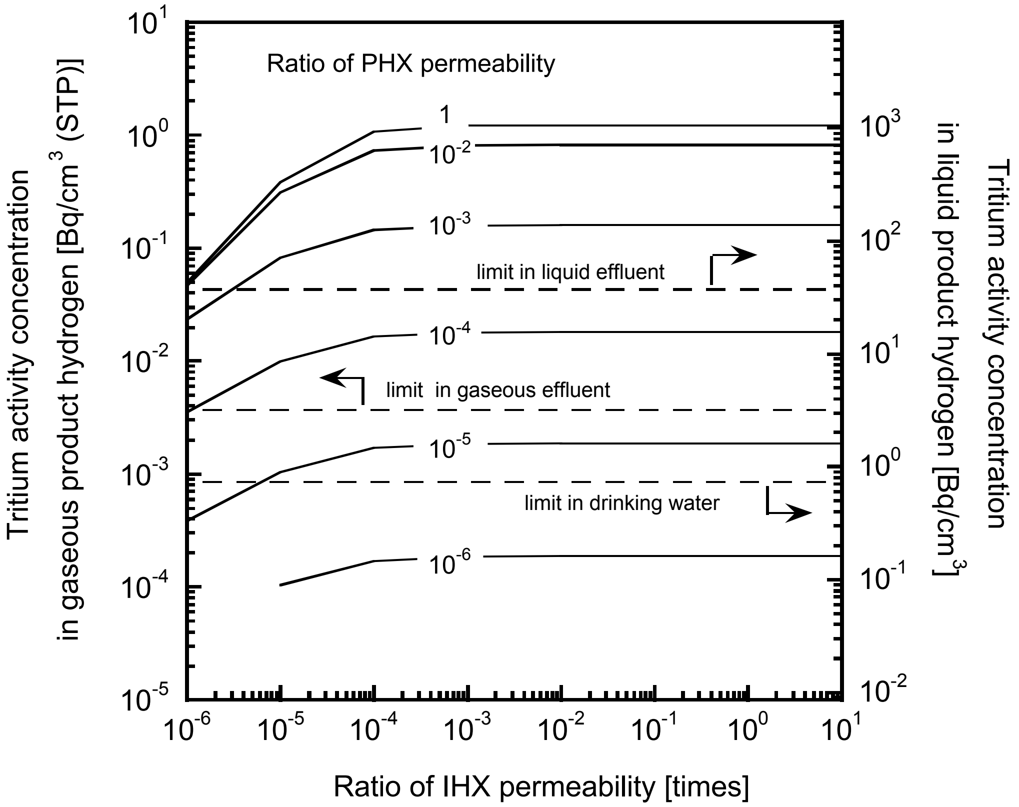

Figure 12: Effect of varying the IHX permeability along with reduction in PHX permeability on tritium concentration in the hydrogen product for the NGNP using the SI process.

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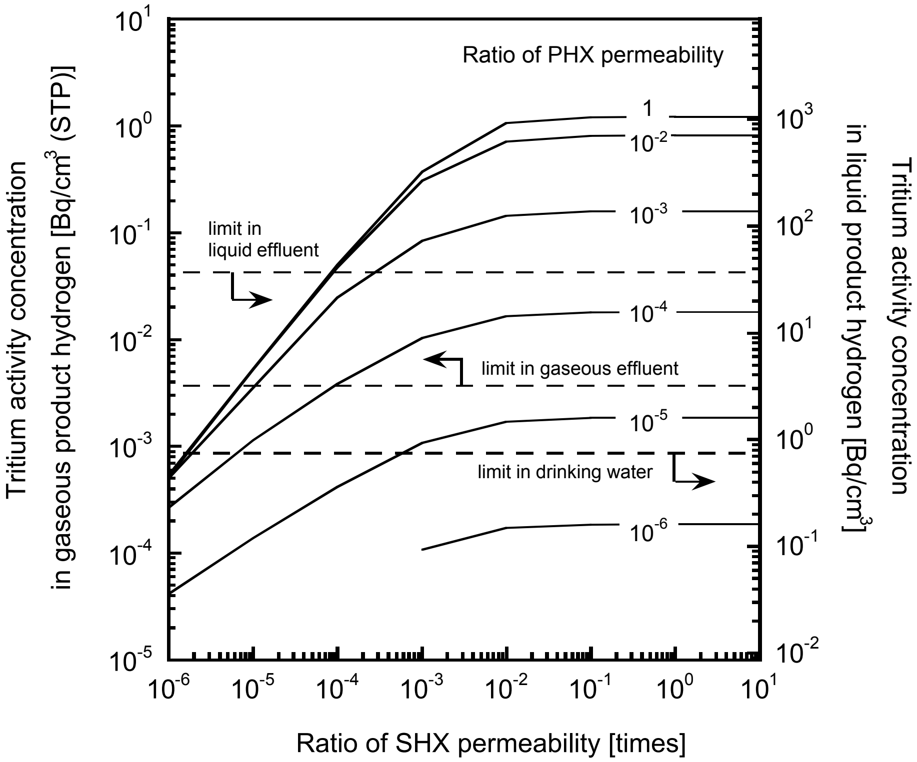

Figure 13: Effect of varying the SHX permeability along with reduction in PHX permeability on tritium concentration in the hydrogen product for an NGNP using the SI process.

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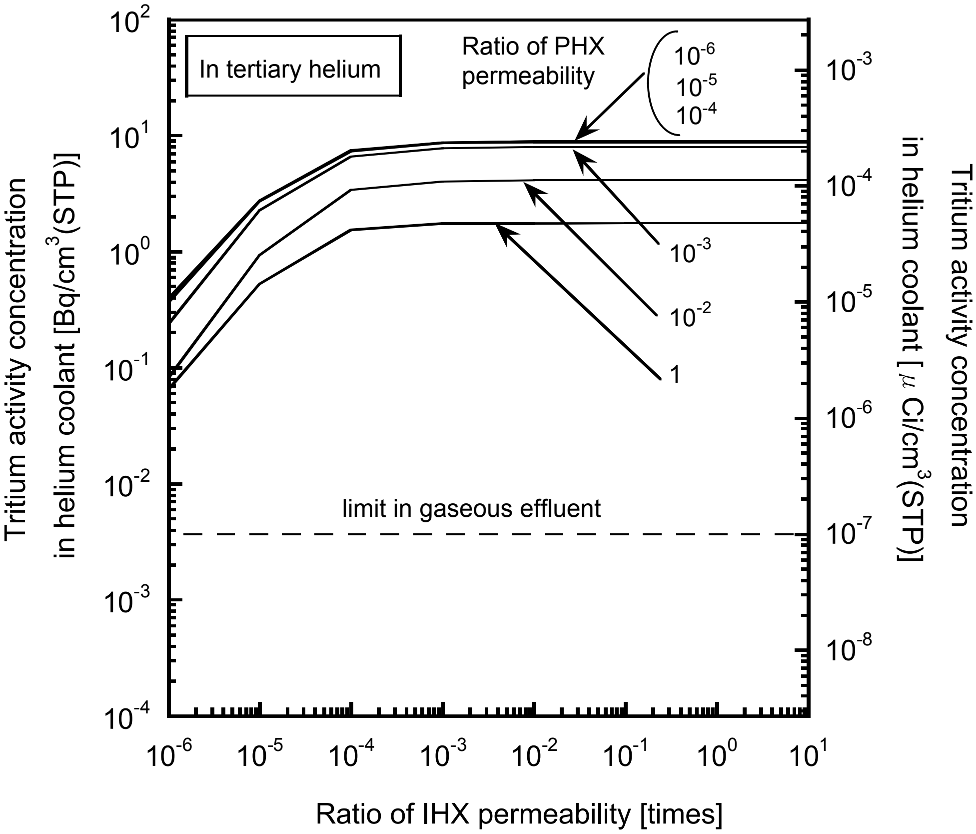

Figure 14: Effect of varying the IHX permeability along with reduction in PHX permeability on tritium concentration in the tertiary loop for an NGNP using the SI process.

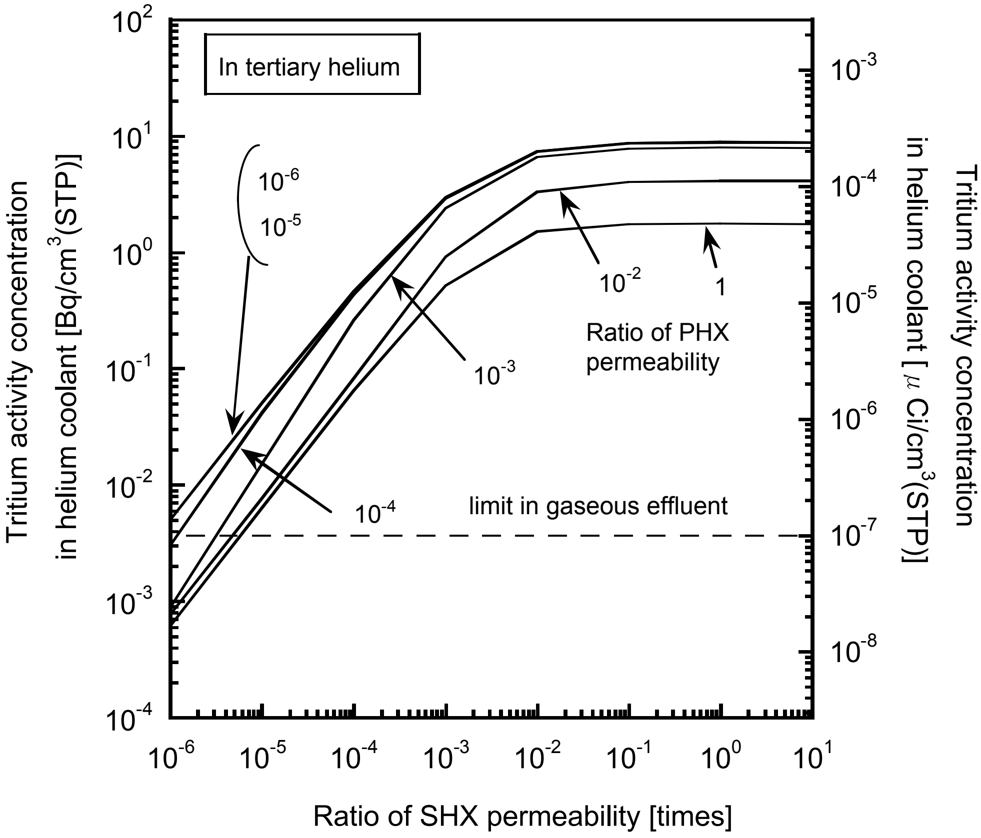

Figure 15: Effect of varying the SHX permeability along with reduction in PHX permeability on tritium concentration in the tertiary loop for an NGNP using the SI process.

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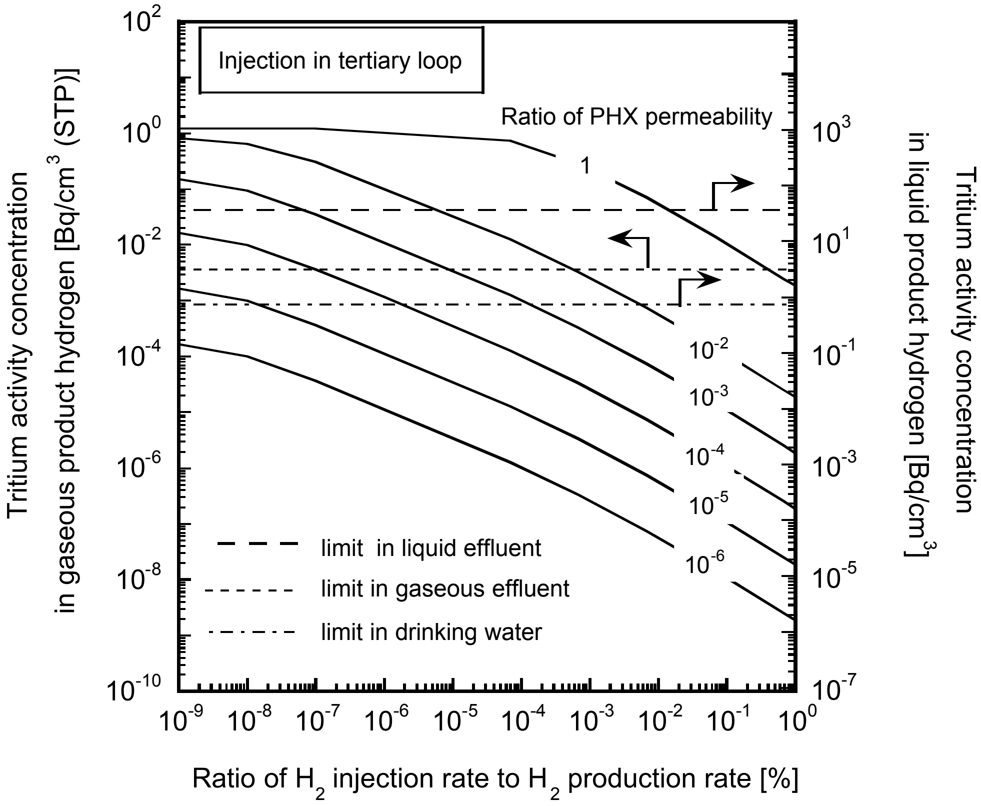

Figure 16: Effect of varying hydrogen injection rate in the tertiary helium coolant on the tritium concentration in the hydrogen product for an NGNP using the SI process.

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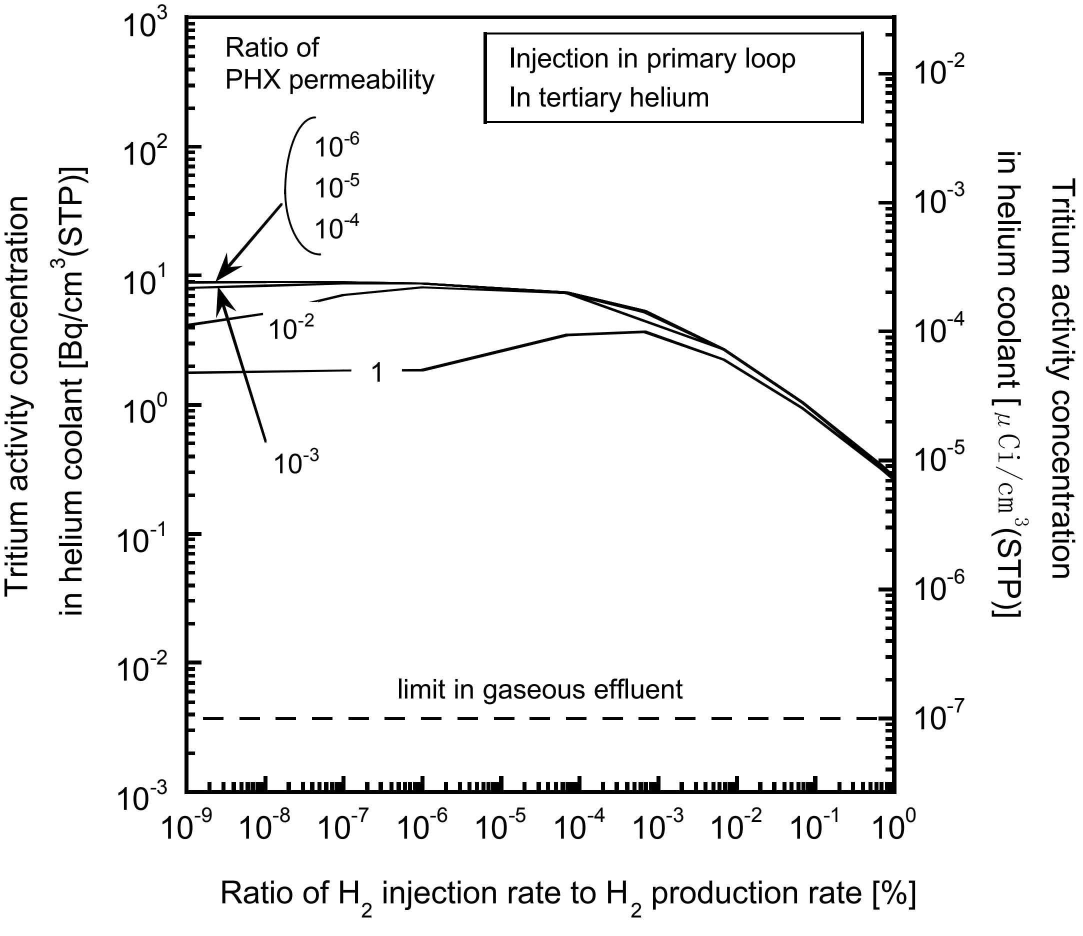

Figure 17: Effect of varying hydrogen injection rate in the helium coolant on the tritium concentration in the tertiary loop for an NGNP using the SI process.

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Authors: Hirofumi Ohashi and Steven R. Sherman

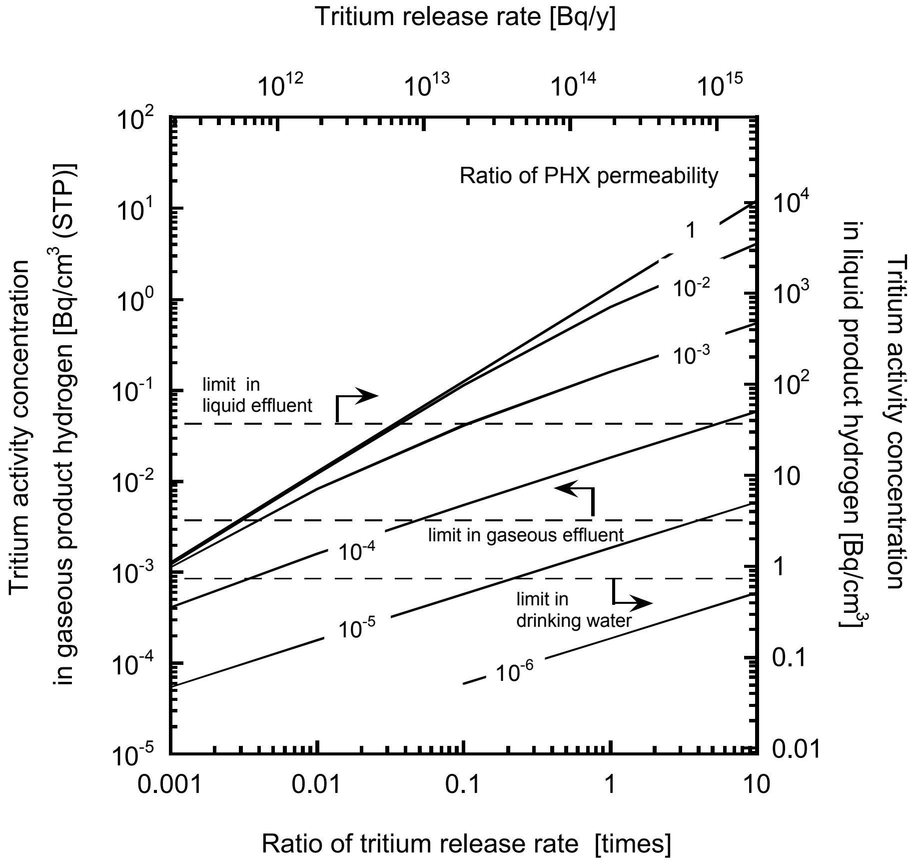

Figure 18: Effect of varying tritium release rate on the tritium concentration in the hydrogen product for an NGNP using the SI process.

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Authors: Hirofumi Ohashi and Steven R. Sherman

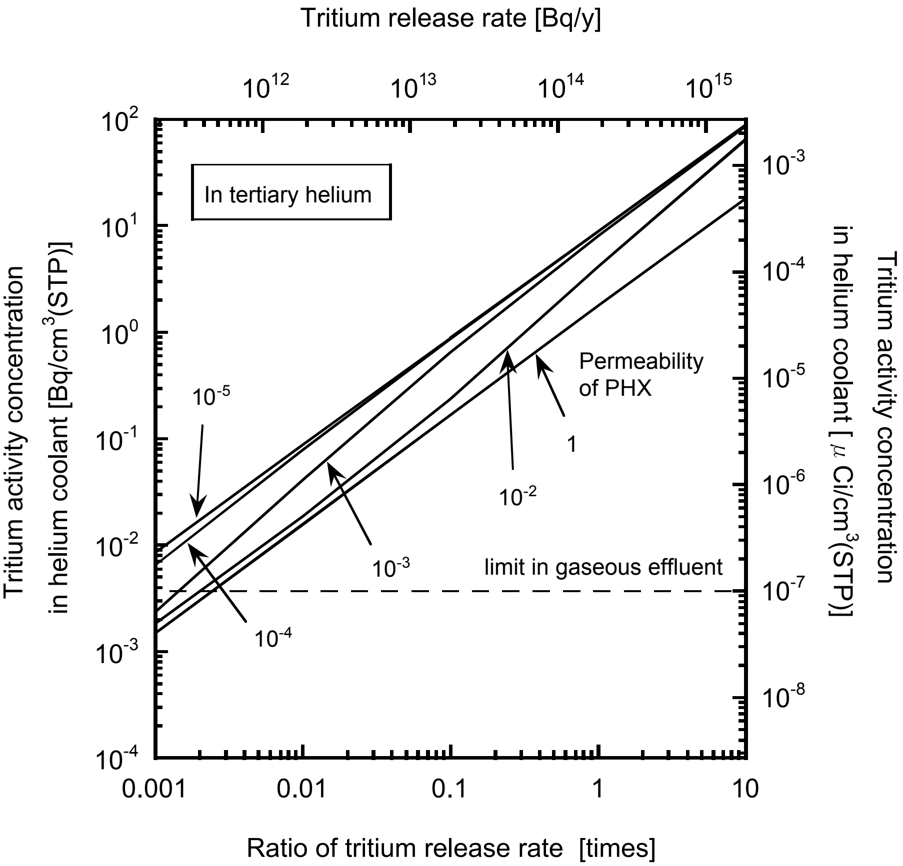

Figure 19: Effect of varying tritium release rate on the tritium concentration in the tertiary helium for the NGNP using the SI process.

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Authors: Hirofumi Ohashi and Steven R. Sherman

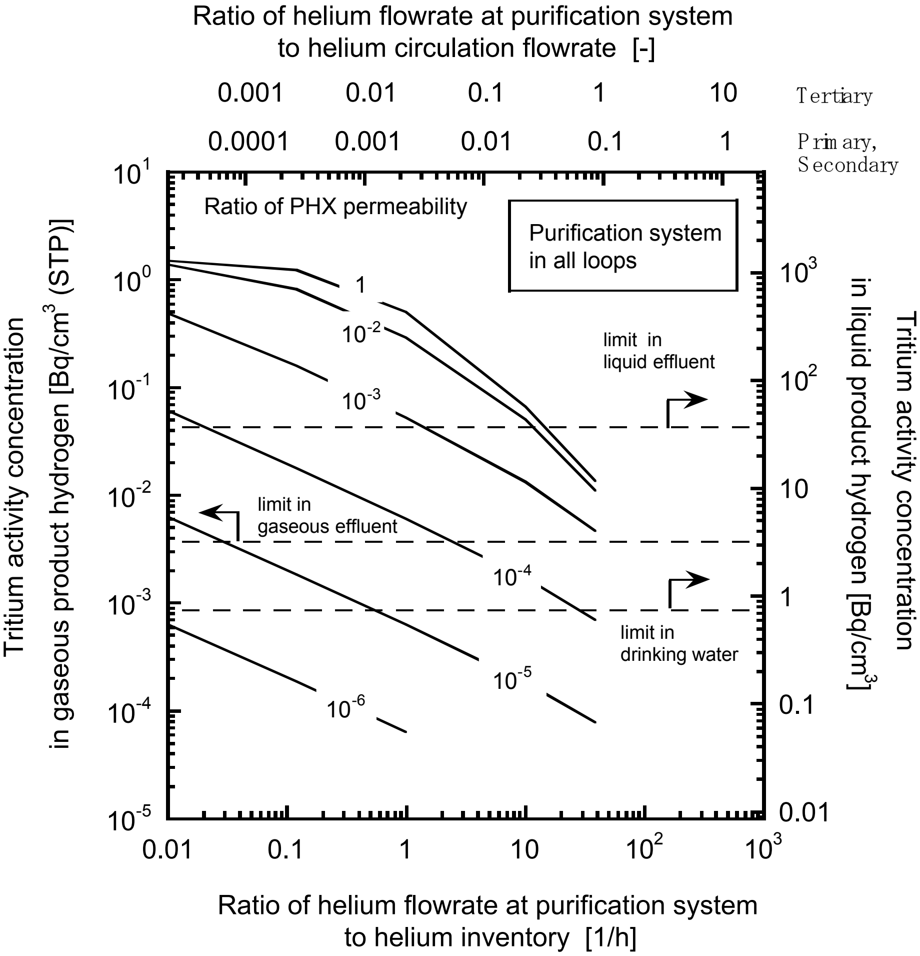

Figure 20: Effect of varying helium flow rate at the purification system in all loops on the tritium concentration in the hydrogen product for an NGNP using the SI process.

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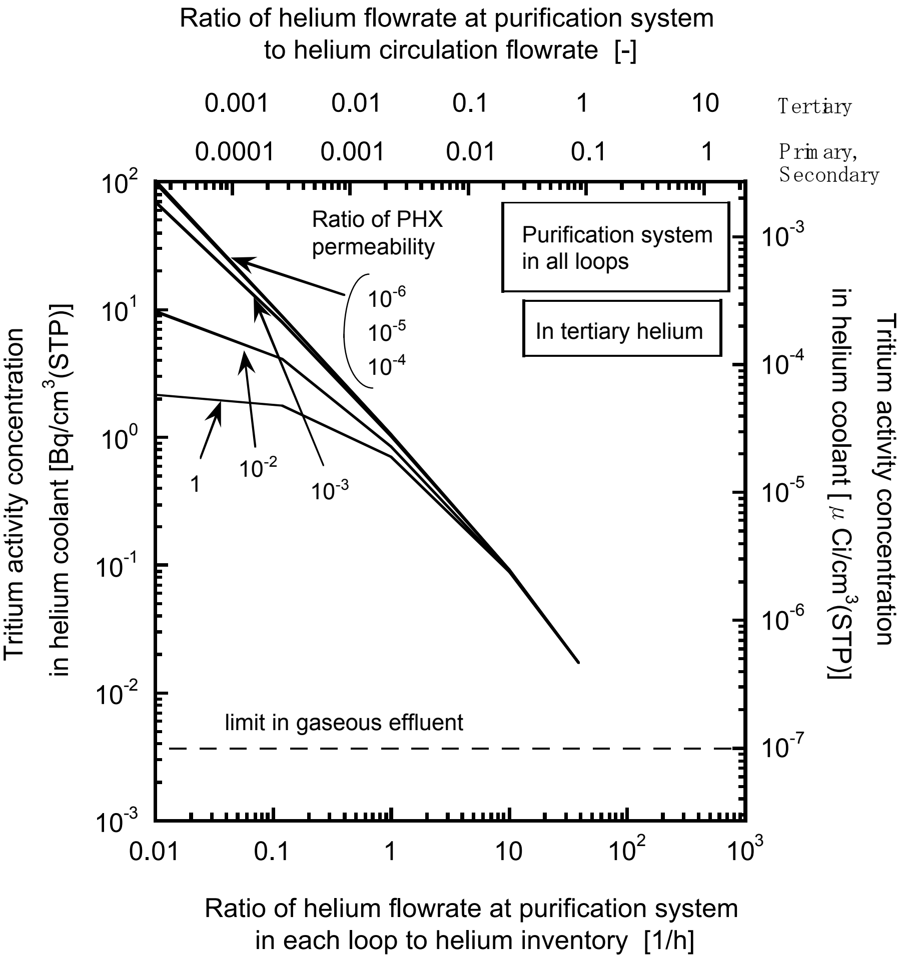

Figure 21: Effect of varying helium flow rate at the purification system in all loops on the tritium concentration in the hydrogen product for an NGNP using the SI process.

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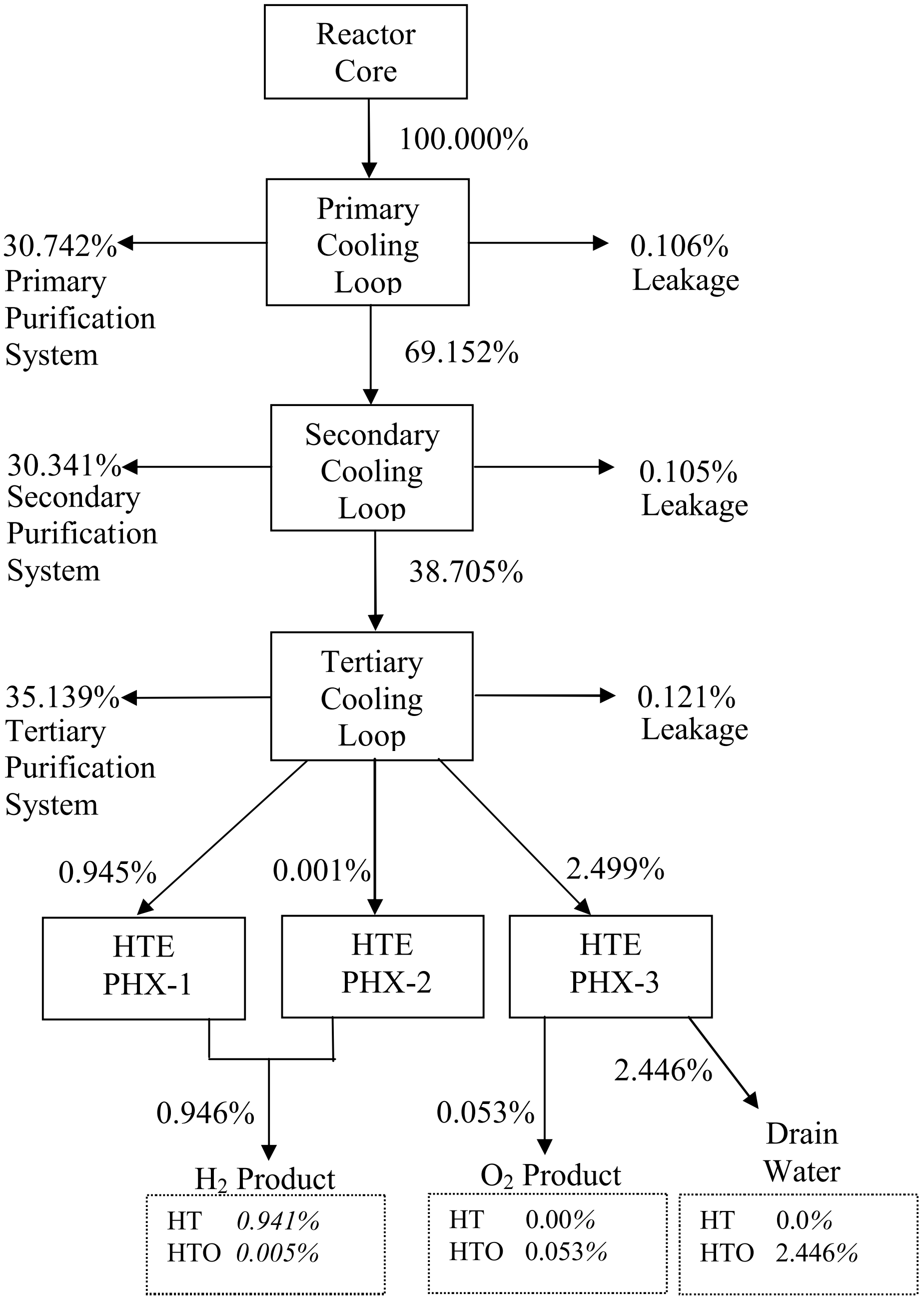

Figure 22: Distribution of tritium in the HTE process for the base case.

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Authors: Hirofumi Ohashi and Steven R. Sherman

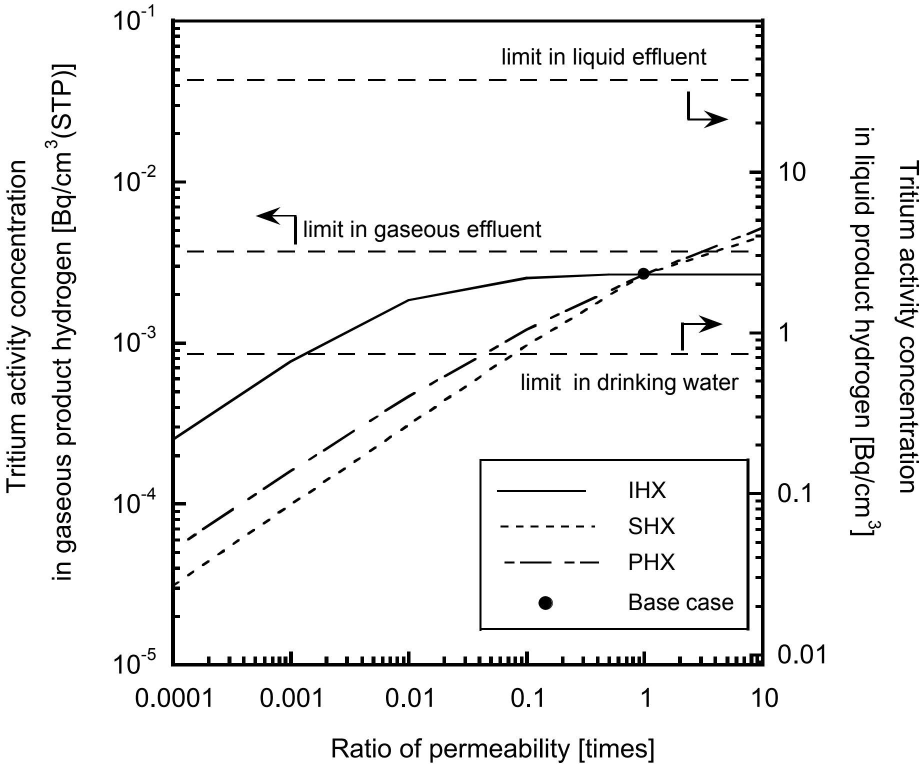

Figure 23: Effect of varying heat exchanger permeabilities on the tritium concentration in the hydrogen product for an NGNP using the HTE process.

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Authors: Hirofumi Ohashi and Steven R. Sherman

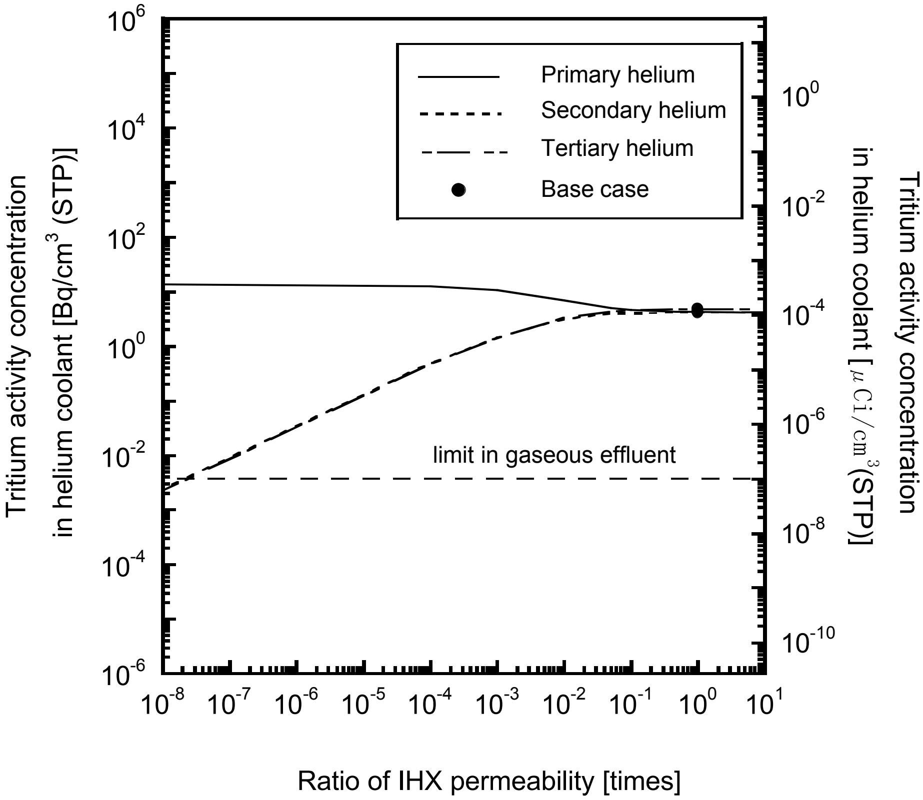

Figure 24: Effect of varying IHX tritium permeability on the concentration of tritium in the tertiary loop for an NGNP using the HTE process.

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Authors: Hirofumi Ohashi and Steven R. Sherman

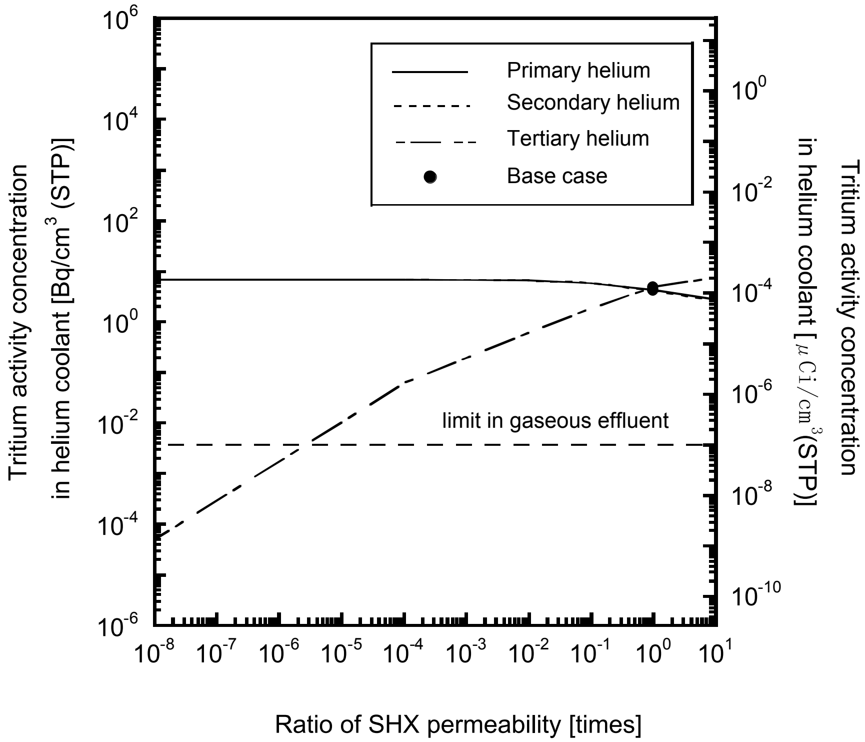

Figure 25: Effect of varying SHX tritium permeability on the concentration of tritium in the tertiary loop for an NGNP using the HTE process.

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Authors: Hirofumi Ohashi and Steven R. Sherman

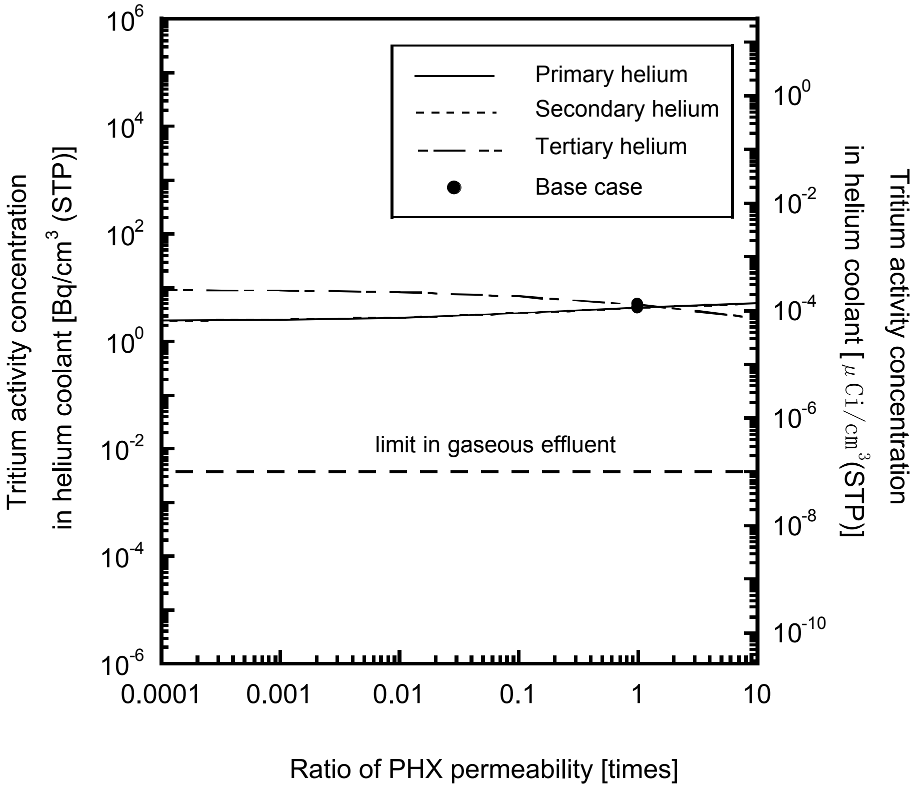

Figure 26: Effect of varying PHX tritium permeability on the concentration of tritium in the tertiary loop for an NGNP using the HTE process.

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Authors: Hirofumi Ohashi and Steven R. Sherman

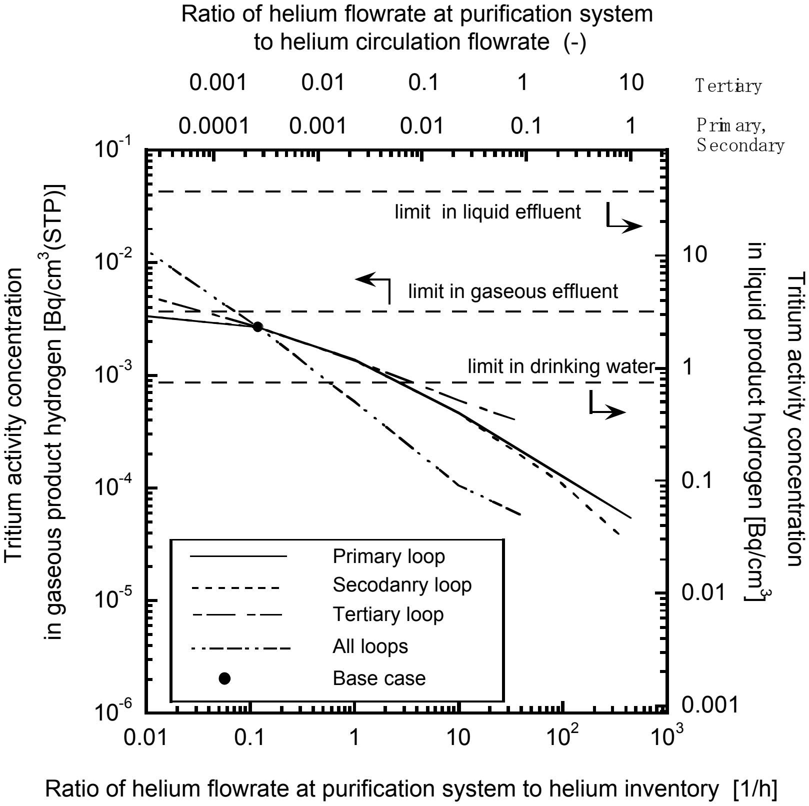

Figure 27: Effect of varying helium flow rates in the purification system on concentration of tritium in the hydrogen product for an NGNP using the HTE process.

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Authors: Hirofumi Ohashi and Steven R. Sherman

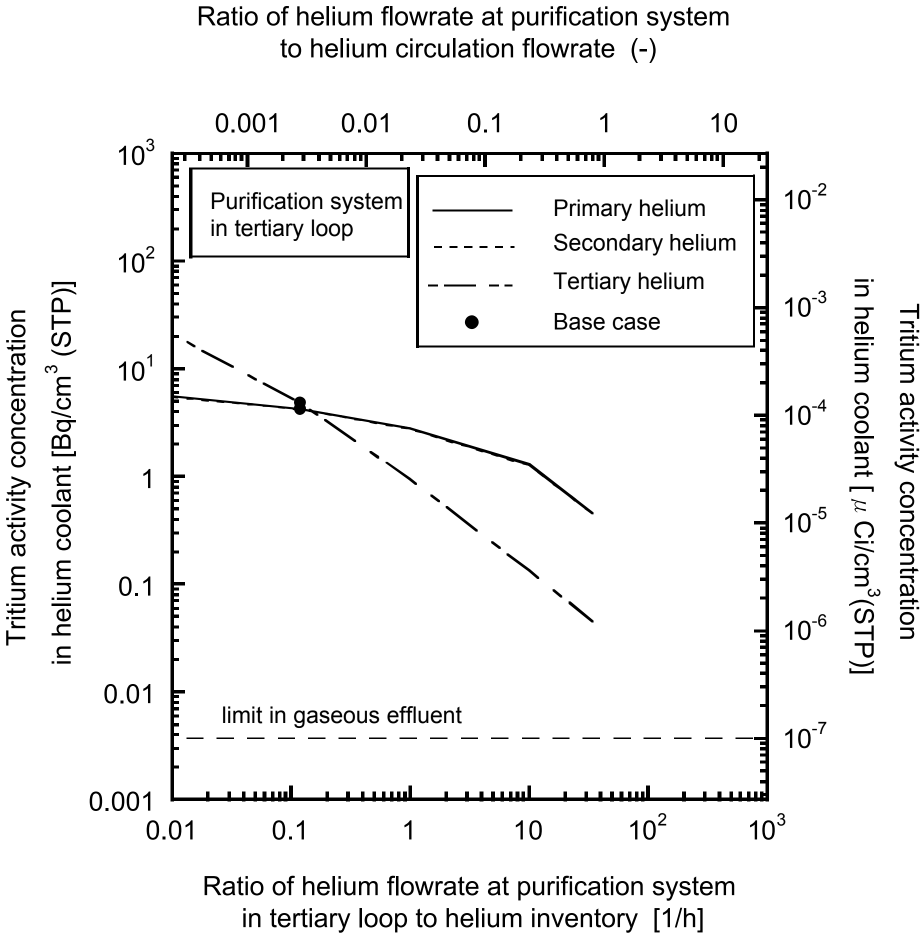

Figure 28: Effect of varying helium flow rates in the purification system on concentration of tritium in the tertiary helium loop for an NGNP using the HTE process.

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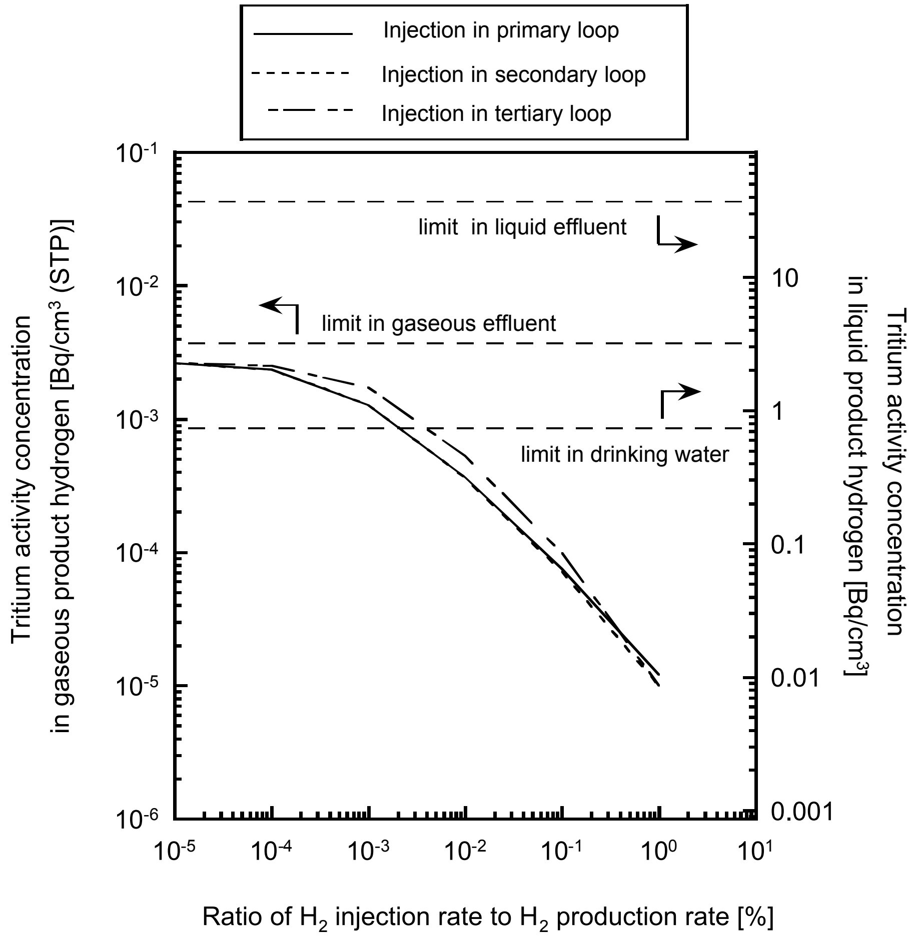

Figure 29: Effect of varying hydrogen injection rate in the helium loops on concentration of tritium in the hydrogen product for an NGNP using the HTE process.

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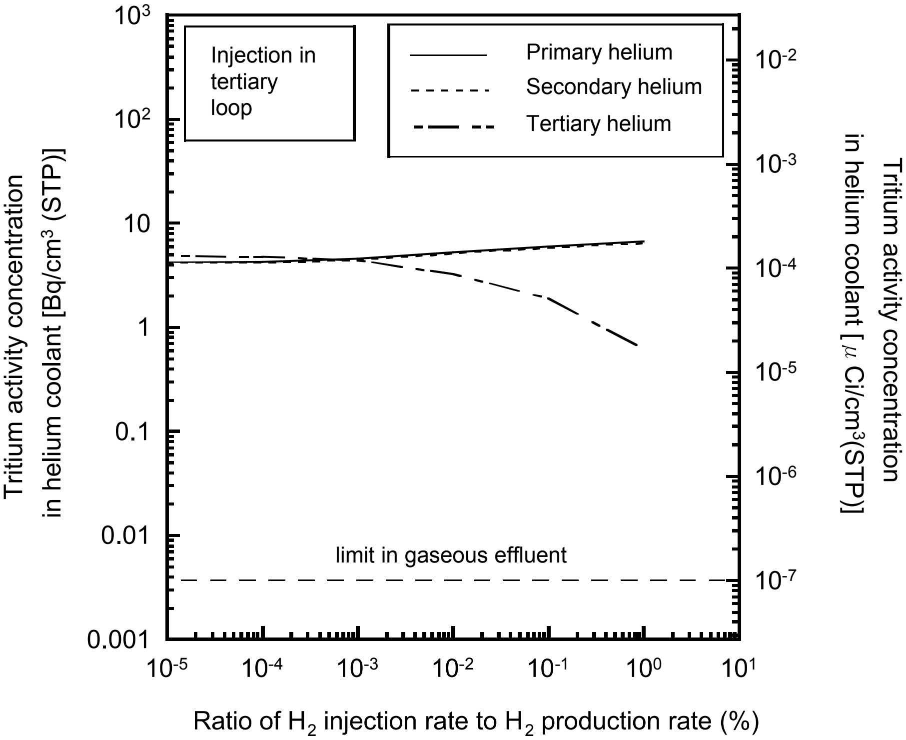

Figure 30: Effect of varying hydrogen injection rate in the tertiary helium loop on concentration of tritium in the tertiary helium loop for an NGNP using the HTE process.

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