# Enhanced Tritium Production for Fusion Reactors Via $^3$He(n, p)$^3$H in the Heavy Water Moderator of a CANDU Reactor

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There is an increasing requirement for tritium to supply the fuel needs of current experimental fusion devices and in the initial startup of future power generating reactors. Tritium is produced in heavy water reactors through deuterium activation, but the total production capacity of Canadian operated CANDUs will fall short of future demands, during the period before and for some time after self-sufficient reactors become available. Consequently, methods of enhancing tritium generating rates warrant investigation. Herein we provide the results of an inquiry into the feasibility of enhancing tritium production levels through the activation of helium-3 following its external addition to the heavy water moderator system of a hypothetical 500–600 MWe CANDU reactor. The approach adopted involves simulation of the temporal evolution of the tritium activities, originating from $^2$H(n, $\gamma$)$^3$H and $^3$He(n,p)$^3$H, as described by a simple first order kinetic model. The results suggest that the frequent addition of helium-3 to the moderator water will enhance tritium production inventories. The enhancement factor is highly dependent upon the rate at which helium-3 irretrievably escapes to the moderator cover gas. However, the direct activation of helium-3, contained in a closed loop such as the annulus gas system, for example, would be essentially complete within a few weeks without any significant loss.

## 1. INTRODUCTION

Early stage experimental fusion devices are based on the high-$Q$ value reaction

$$^2\text{H} + {}^3\text{H} \rightarrow {}^4\text{He} + n + 17.59 \text{ MeV}$$
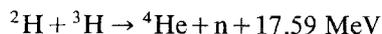
between deuterium ($^2$H or D) and tritium ($^3$H or T) because of its relatively low ignition temperature

requirements. Despite the technological difficulties in realizing fusion as a viable power source, the primary impetus for developing the fusion reactor lies in the abundance of its most important raw material, deuterium. Deuterium comprises about 0.015% of all hydrogen in nature, much of this inventory being present in the world's oceans. As the technology for extracting deuterium from water exists, deuterium availability will not limit the energy potential from fusion reactors. Tritium, the other component of the DT fuel cycle, is not found naturally in sufficiently high concentrations to meet current and near term fusion demands, and so it must be produced. In reactor designs based on the tokamak principle, the plasma region is surrounded by a lithium based mod-

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erator blanket which absorbs thermal and energetic neutrons to produce tritium. This tritium can be recycled to the fusion chamber following extraction from the breeding materials. Throughout the current developmental phase and in the startup of future commercial fusion reactors, an external supply source of tritium will be required.

Tritium is produced as an operating waste in CANDU[4] fission reactors. The primary mode of tritium production is through the thermal neutron activation of deuterium, via $^2$H(n, $\gamma$)$^3$H, in the heavy water moderator system and, to a lesser extent, in the heat transport system. Current CANDU designs include many features directed towards heavy water management tasks, such as the in-station monitoring and prediction of tritium inventories and the minimization of tritium dose and airborne emissions, which indirectly control the accumulation and release of tritium.[1] A centralized tritium removal facility will be available by the end of 1987 to serve all CANDU reactors operated by Ontario Hydro.[2] Proven technology for the recovery, concentration, and storage of tritium is available. This accumulated inventory of in-storage tritium could serve as a supply source for various applications, including fusion.

The estimated[3] tritium generating rate for Canadian CANDUs is 2 kg·a$^{-1}$ in 1985 and 2.8 kg·a$^{-1}$ in 2000, which is substantially above the production rates for fuel reprocessing plants (0.84 kg·a$^{-1}$ in 2000) and pressurized light water reactors (0.032 kg·a$^{-1}$ in 2000). Since fusion reactor demands for tritium could exceed CANDU production capabilities during the period before and for some time after self-sufficient reactors become available, methods of enhancing tritium generating rates in heavy water reactors should be explored. The feasibility of breeding tritium, via $^6$Li(n, $^4$He)$^3$H, in a CANDU facility, by placing lithium based materials in the reactivity control adjustor rods, has recently been investigated.[4] The irradiation of lithium isotopes, placed in operating power reactors or in dedicated neutron devices, has been carried out successfully in the United States.[5] The quantities of tritium that can be produced in a CANDU reactor using this technique are limited, being even less than that produced in the heavy water coolant system. Herein we investigate the feasibility of achieving increased tritium production levels for fusion fuel supplies by the external injection of $^3$He into the heavy water moderator system of a CANDU reactor. The basis for this approach is threefold:

1. The thermal neutron absorption cross-section for the reaction $^3$He(n,p)$^3$H, in which the product of tritium $\beta^-$ decay is converted back to tritium, is about seven orders of magnitude greater[6] than the cross-section for the reaction $^2$H(n, $\gamma$)$^3$H, the principal route for tritium generation in the heavy water systems of a CANDU reactor.

2. The technology for immobilization and storage of tritium produced, unwittingly or otherwise, in a CANDU facility is available[1, 2] to maintain in-station levels and airborne emissions within regulatory limits.

3. Superficially it appears that the only retrofit requirements on existing CANDU designs are the provision of multiple orifice inlets with gas-sparged contactors for the uniform dispersion of $^3$He throughout the moderator system.

The approach taken in examining the feasibility of enhancing tritium production involves the formal development of a simple first order kinetic model to describe the temporal evolution of an arbitrary initial inventory of deuterium, tritium, and helium-3 uniformly exposed to a thermal neutron flux and incorporating external input flows of helium-3 into the reactor. Computer simulations of the enhancement factors for tritium production in a hypothetical 500–600 MWe CANDU reactor are carried out from startup to the anticipated 30 year lifetime, as a function of such process variables as the effective neutron flux, helium-3 retention time in the moderator, the input durations and delivery times, and the input concentrations of helium-3. The provision of quantitative estimates of tritium supplies for fusion fuel purposes from Canadian CANDUs and the impact on operations, in particular, heavy water management strategies, resulting from the adoption of the proposed enhancement process are not addressed.

Section 2.1 details the formal development of the kinetic model used to describe the free evolution of an initial inventory of deuterium, tritium, and helium-3 and the evolution of pulsed or impulsive additions of helium-3 in a thermal neutron flux. This is followed in Sec. 2.2 by a discussion of the computer simulations of the enhanced production of tritium and includes details on the requisite input data to the computer code used and the computational aspects of the simulations. Results of the various simulations carried out are presented and discussed in Sec. 3, which also includes a discussion on the solubility of helium-3 in heavy water, the physicochemical parameter that determines the retention time of helium-3 in the moderator. The paper

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[4]*CANada-Deuterium-Uranium.*

closes with some concluding remarks in Sec. 4. Unless otherwise stated, deuterium, tritium, and helium-3 are designated D, T, and He, respectively.

## 2. METHOD

### 2.1. Formal Aspects

Consider a system consisting of a thermal neutron flux incident upon an initial inventory of D, T, and He. If $n_D(t)$, $n_T(t)$, and $n_{He}(t)$ are the instantaneous fractional inventories of D, T, and He, respectively, then they evolve in accordance with the first order Bateman system,

$$\dot{n}(t) = \underline{N}\underline{n}(t) \tag{1}$$

where $\underline{n}^T(t) = (n_D(t) n_T(t) n_{He}(t))$ is a row matrix of the instantaneous fractional inventories of D, T, and He, and $\underline{N}$ is a square coefficient matrix of the source and sink rate constants for D, T, and He, which is assumed to have the explicit form

$$\underline{N} = \begin{bmatrix} -\phi\sigma_D & 0 & 0 \\ \phi\sigma_D & -\lambda_T & \phi\sigma_{He} \\ 0 & \lambda_T & -(\phi\sigma_{He} + \lambda_{He}) \end{bmatrix} \tag{2}$$

In Eq. (2), $\phi$ is the effective neutron flux, $\sigma_D$ is the neutron absorption cross-section for $D(n, \gamma)T$, $\sigma_{He}$ is the neutron absorption cross-section for $^3He(n, p)T$, $\lambda_T$ is the rate constant for the $\beta^-$ decay of T to $^3He$, and $\lambda_{He}$ is the rate constant for $^3He$ escape from the moderator to the cover gas. The formal solution[7-9] of Eq. (1) is given by

$$\underline{n}(t) = \exp(\underline{N}t)\underline{n}(0) \tag{3}$$

where $\underline{n}(0)$ is the initial fractional inventory of D, T, and He in the system. If $\underline{m}(t)$ is a column matrix representing the external input flow of D, T, and He into the reactor, Eq. (1) is replaced by

$$\dot{n}(t) = \underline{N}\underline{n}(t) + \underline{m}(t), \tag{4}$$

whose formal solution[7-9] is

$$\underline{n}(t) = \exp(\underline{N}t)\underline{n}(0) + \int_0^t ds \exp[\underline{N}(t-s)]\underline{m}(s) \tag{5}$$

subject to the initial conditions $\underline{n}(0)$. The first term on the right in Eq. (5) represents the solution to Eq. (1) in the absence of external input flow; see Eq. (3). The second term involves a convolution over the external input and determines the contribution of the external input to the instantaneous fractional inventory $\underline{n}(t)$.

We take $\underline{m}(t)$ to be of the form

$$\underline{m}(t) = \frac{1}{\sqrt{\pi}} \sum_{i=1}^{n_I} \frac{1}{\tau_i} \exp\left[-(t-t_i)^2/\tau_i^2\right] \underline{n}(t_i) \tag{6}$$

representing the gaussian pulsed injection of $i = 1, \ldots n_I$ deliveries of the species D, T, and He, as specified by $\underline{n}(t_i)$,[5] each over an epochal duration $\tau_i$ centered at delivery times $t_i$. If we write

$$\exp(\underline{N}t) = \sum_{i=1}^{3} \exp(\lambda_i t)\underline{N}_i' \tag{7}$$

where the $\lambda$'s are the distinct eigenvalues of $\underline{N}$, and the $\underline{N}'$s are square Bateman coefficient matrices,[10,11] substitute Eqs. (6) and (7) into Eq. (5), and evaluate the convolution integral, one obtains

$$\underline{n}(t) = \sum_{i=1}^{3} \exp(\lambda_i t)\underline{N}_i'\underline{n}(0)$$

$$+ \frac{1}{2} \sum_{j=1}^{n_I} \sum_{k=1}^{3} \left\{ \exp\left[-(t_j/\tau_j)^2 + \lambda_k t\right] \right.$$

$$\times \omega\left[i/\tau_j\left(-t_j + \lambda_k \tau_j^2/2\right)\right]$$

$$- \exp\left[-(t-t_j)^2/\tau_j^2\right]$$

$$\left. \times \omega\left[i/\tau_j\left(t-t_j + \lambda_k \tau_j^2/2\right)\right] \right\}$$

$$\times \underline{N}_k'\underline{n}(t_j) \tag{8}$$

where $\omega(z)$ is related to the error function[12] at any point $z$ in the complex plane, and use has been made of the projection and idempotent properties[10,11] of the $\underline{N}'$s, viz.,

$$\underline{N}_i'\underline{N}_j' = 0 \qquad \text{for } i \neq j \tag{9a}$$

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[5] $\underline{n}(t_i)$ is a column matrix of unit norm whose entries represent the fractional inventories of D, T, and He, respectively, added at time $t_i$. The distinction between $\underline{n}(t)$ and $\underline{n}(t_i)$ should be clear from the context.

and

$$\underline{N}_i'^k = \underline{N}_i' \qquad \text{for } k \text{ any positive integer} \qquad (9b)$$

In order for $\underline{n}(t)$, as given by Eq. (8), to be asymptotically stable,[7,10] one must have $\text{Re}(\lambda) < 0$ for $\lambda$ any eigenvalue of $\underline{N}$.

If $\underline{m}(t)$, defined in Eq. (6), is viewed as the external input fractional inventory distribution function, then

$$\hat{\underline{m}}(t) = \int_0^t ds\,\underline{m}(s)$$

is the cumulative amount added at time $t$ and is given by

$$\hat{\underline{m}}(t) = \frac{1}{2} \sum_{j=1}^{n_I} \left\{ \text{erf}\left[(t-t_j)/\tau_j\right] + \text{erf}\left[t_j/\tau_j\right] \right\} \underline{n}(t_j) \tag{10}$$

Since

$$\lim_{\tau \to 0} \frac{1}{\tau\sqrt{\pi}} \exp\left[-(t-t')^2/\tau^2\right] = \delta(t-t')$$

the Dirac impulse function,[13] it is easy to show that Eq. (8) reduces to

$$\underline{n}(t) = \sum_{i=1}^{3} \exp(\lambda_i t)\underline{N}_i'\underline{n}(0)$$

$$+ \sum_{j=1}^{n_I} \sum_{k=1}^{3} \exp\left[\lambda_k(t-t_j)\right]\underline{N}_k'$$

$$\times \theta(t-t_j)\underline{n}(t_j) \tag{11}$$

as $\tau_j \to 0$ for $j = 1,\ldots,n_I$, where $\theta$ is the Heaviside step function,[12,13] while Eqs. (6) and (10) reduce to

$$\underline{m}(t) = \sum_{j=1}^{n_I} \delta(t-t_j)\underline{n}(t_j) \tag{12}$$

and

$$\hat{\underline{m}}(t) = \sum_{j=1}^{n_I} \theta(t-t_j)\underline{n}(t_j) \tag{13}$$

respectively.

Equation (8) and its limiting form, Eq. (11), represent the instantaneous fractional inventories of D, T, and He as the sum of contributions from the freely evolving initial fractional inventory $\underline{n}(0)$ and the pulsed or impulsive external input of reactants, as a function of such process variables as the effective thermal neutron flux, the He retention time in the moderator, the pulse durations, the input repetition frequency, and the input concentrations of the additional reactants. In both Eqs. (8) and (11), the first term on the right represents the contribution from the freely evolving initial inventory while the second term represents the collective contribution from the independently evolving additional inputs of reactants.

## 2.2. Computational Aspects

Both Eqs. (8) and (11) were coded in FORTRAN (ASCII) for the UNIVAC 1182. Briefly, on being supplied with pertinent input data, the program computes the instantaneous fractional inventory due to the free evolution of the initial inventory of reactants and that due to the pulsed or impulsive addition of reactants and combines these to output the final inventories of D, T, and He in concentration units in addition to the fractional and absolute enhancement factors for T. The requisite input data for a single simulation generally consists of the initial inventory $\underline{n}(0)$ of D, T, and He, the parameters $\phi$, $\sigma_D$, $\sigma_{He}$, $\lambda_T$, and $\lambda_{He}$, which define the coefficient matrix in Eq. (2), the number $(n_I)$ and nature (pulsed/impulsive) of the external inputs and, if necessary, their durations $(\tau_i, i = 1,\ldots,n_I)$, the total quantity (moles) added on each input as well as the mole fractions $(\underline{n}(t_i), i = 1,\ldots,n_I)$ of the species added, the delivery times $(t_i, i = 1,\ldots,n_I)$ of the inputs, and the total duration of the simulation. Although not necessary, it is usual to take $\underline{n}^T(0) = (100)$, corresponding to an initial inventory of D only, and so the evolution of T can be monitored from the startup of the reactor. The parameters $\sigma_D$, $\sigma_{He}$, and $\lambda_T$ are fixed at their measured values. The effective thermal neutron flux $\phi$ is the product of the actual flux, the fraction of the moderator in core, and the unit's capacity factor, and is essentially fixed; for a 500–600 MWe CANDU unit, $\phi$ has a typical value of the order of $10^{21}$ cm$^{-2}$·a$^{-1}$. The parameter $\lambda_{He}$ is unknown and may be varied from zero, corresponding to the complete retention of He in the moderator, to extremely large values, corresponding to the rapid escape of $^3$He to the moderator cover gas.

Since the objective is to promote the production of T, all inputs are restricted to He additions by taking $\underline{n}^T(t_i) = (0\,0\,1)$ for all $i = 1, \ldots, n_I$, although other choices are possible, such as those corresponding to virgin or tritiated heavy water additions as appropriate to moderator makeup operations, provided the coefficient matrix in Eq. (2) is modified to incorporate rate constants for the loss of reactants from the system. The number of external inputs and the quantities added are limited by the availability of $^3$He. The durations and delivery times of pulsed injections should be chosen so that $t_i \gg \tau_i$ and $(t_i - t_j)^2 \gg \tau_j^2$ for $i, j = 1, \ldots, n_I$; this ensures that the epochal and nonoverlapping nature of the pulses preclude causality difficulties. With the admission of external input flows of reactants, the system under consideration is open. Consequently, the total mass of the reactants initially present must be specified; for a 500–600 MWe CANDU reactor, the moderator typically has a mass of about 300,000 kg.

Within the aforementioned constraints on the various process parameters, the T inventories evolving in a hypothetical 500–600 MWe CANDU reactor over an anticipated 30 year lifetime were simulated. In each case, an initial inventory consisting of D only was taken, and the influences of effective thermal neutron flux, He retention time, pulse durations, pulse delivery times, and the quantities of He added on the evolving T activities were investigated. From these results the instantaneous enhancement factors for T production were obtained as a function of the adjustable process variables. The fractional enhancement factor for T production is simply the ratio of its mole fraction with additions to the freely evolving mole fraction. The numerical data used[6, 14–16] in generating the results of the simulations discussed in

the next section are summarized in Table I, in addition to various conversion factors used in expressing the fractional inventories in concentration units. The results presented in Sec. 3 should not be construed as representing current or future T inventory trends in operating commercial CANDU facilities since the impact of heavy water management strategies has not been incorporated.

The evaluation of Eqs. (8) and (11) involves three computational tasks, viz., evaluation of the eigenvalues of the coefficient matrix $\underline{N}$, evaluation of the special function $\omega(z)$ at an arbitrary point $z$ in the complex plane, and evaluation of the Bateman coefficient matrices $\underline{N}_i'$, $i = 1, 2$, and 3. Particular care is required in evaluating the eigenvalues of $\underline{N}$ because of its inherent stiffness. The efficient algorithm due to Jenkins and coworkers,[17, 18] available as subroutine ZRPOLY in the IMSL library,[19] was used to generate the eigenvalues to within double precision accuracy. Evaluation of $\omega(z)$ to within double precision accuracy at an arbitrary point $z$ was accomplished using the Gautschi algorithm,[20, 21] which is available as subroutine MERRCZ in the IMSL library. The matrices $\underline{N}_i'$, $i = 1$, 2, and 3 were computed using Faddeeva's modification[22] of Laverrier's algorithm.[23] This approach, although computationally laborious, is insensitive to the vagaries of $\underline{N}$ arising from its stiffness. In addition, the extent to which the projection and idempotent properties of the $\underline{N}'$s, see Eq. (9), and the sum rule

$$\sum_{i=1}^{3} \underline{N}_i' = \underline{I}$$

where $\underline{I}$ is the unit matrix, are fulfilled serve to gauge the accuracies of the computed results. Our use of this approach generated the $\underline{N}'$s to within double

Table I. Numerical Data Used in the Calculations[a]

| | |
|---|---|
| $\sigma_D$, cross-section for $D(n, \gamma)T$ | $5.3 \times 10^{-28}$ cm$^2$ [6] |
| $\sigma_{He}$, cross-section for $^3$He$(n, p)T$ | $5.33 \times 10^{-21}$ cm$^2$ [6] |
| $\lambda_T$, rate constant for decay of T | $5.653729 \times 10^{-2}$ a$^{-1}$ [14] |
| Avagadro's number | $6.022045 \times 10^{23}$ mol$^{-1}$ [15] |
| Ideal gas molar volume (273.15 K, 101.325 kPa) | $2.241383 \times 10^{-2}$ m$^3 \cdot$ mol$^{-1}$ [15] |
| Earth's sidereal period | $3.1558118 \times 10^7$ s [15] |
| $D_2O$ molecular weight | $20.027604$ g$\cdot$mol$^{-1}$ [16] |
| $^3$He atomic weight | $3.0160293$ g$\cdot$mol$^{-1}$ [16] |
| Typical moderator mass of 500–600 MWe CANDU unit | $300\,000$ kg |
| Typical effective neutron flux ($\phi$) of 500–600 MWe CANDU unit | $2.37 \times 10^{21}$ cm$^{-2} \cdot$a$^{-1}$ |
| Mole fraction ($n_T$) of T to activity concentration | $2.9158566 \times 10^6$ Ci$\cdot$kg$^{-1}$ |
| Mole fraction ($n_{He}$) of $^3$He to mass concentration | $3.0160293 \times 10^8$ ppb |
| Mole fraction ($n_{He}$) of $^3$He to ideal gas volume concentration | $2.241383$ m$^3 \cdot$kg$^{-1}$ |

[a]Reference numbers in brackets.

precision accuracy. The overall accuracy of the computed inventories, whether through Eq. (8) or Eq. (11) for pulsed or impulsive inputs, respectively, is within optimal single precision on the UNIVAC 1182.

# 3. RESULTS

## 3.1. Effects of Thermal Neutron Flux

Since the activation rate constants appearing in Eq. (2) are first order in the thermal neutron flux, one

anticipates that as the flux increases so also will the T inventory. Figures 1 and 2 give the results obtained for the free evolution of T and He inventories over a period of 30 years at an effective neutron flux of $3.16 \times 10^{20}$ and $3.16 \times 10^{21}$ cm$^{-2}$·a$^{-1}$, respectively, for $\lambda_{He} = 0$ a$^{-1}$, corresponding to the complete retention of He in the moderator, and $\lambda_{He} = 10^{10}$ a$^{-1}$, corresponding to the rapid escape of He. One observes that the order of magnitude increase in the flux is indeed matched by a corresponding increase in the T concentrations, irrespective of the magnitude of $\lambda_{He}$. For both values of the flux, the T activity
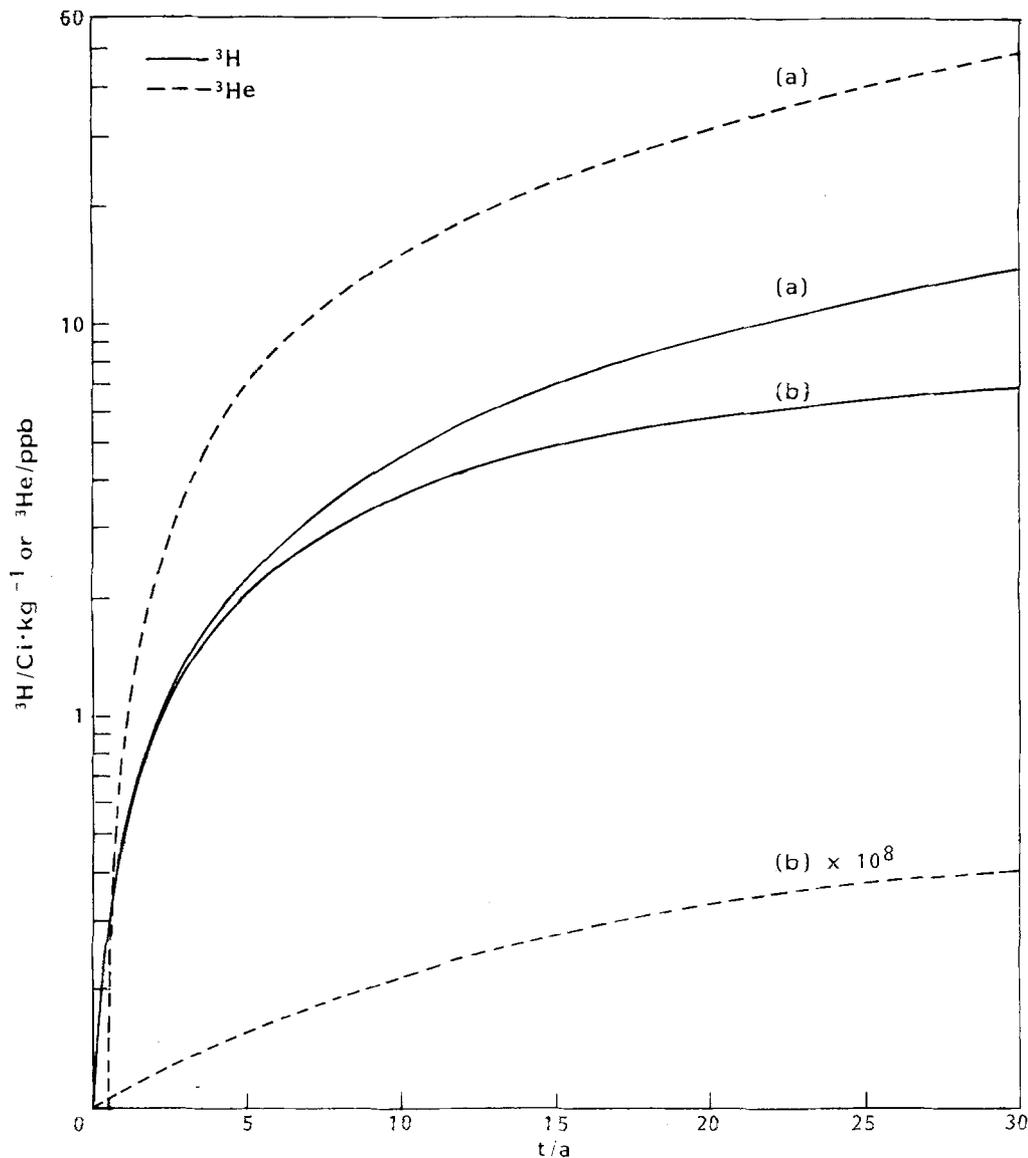


Fig. 1. The variation of tritium and helium-3 concentrations with time at an effective neutron flux of $3.16 \times 10^{20}$ cm$^{-2}$·a$^{-1}$ for a helium-3 escape rate constant of (a) 0 a$^{-1}$, and (b) $10^{10}$ a$^{-1}$.

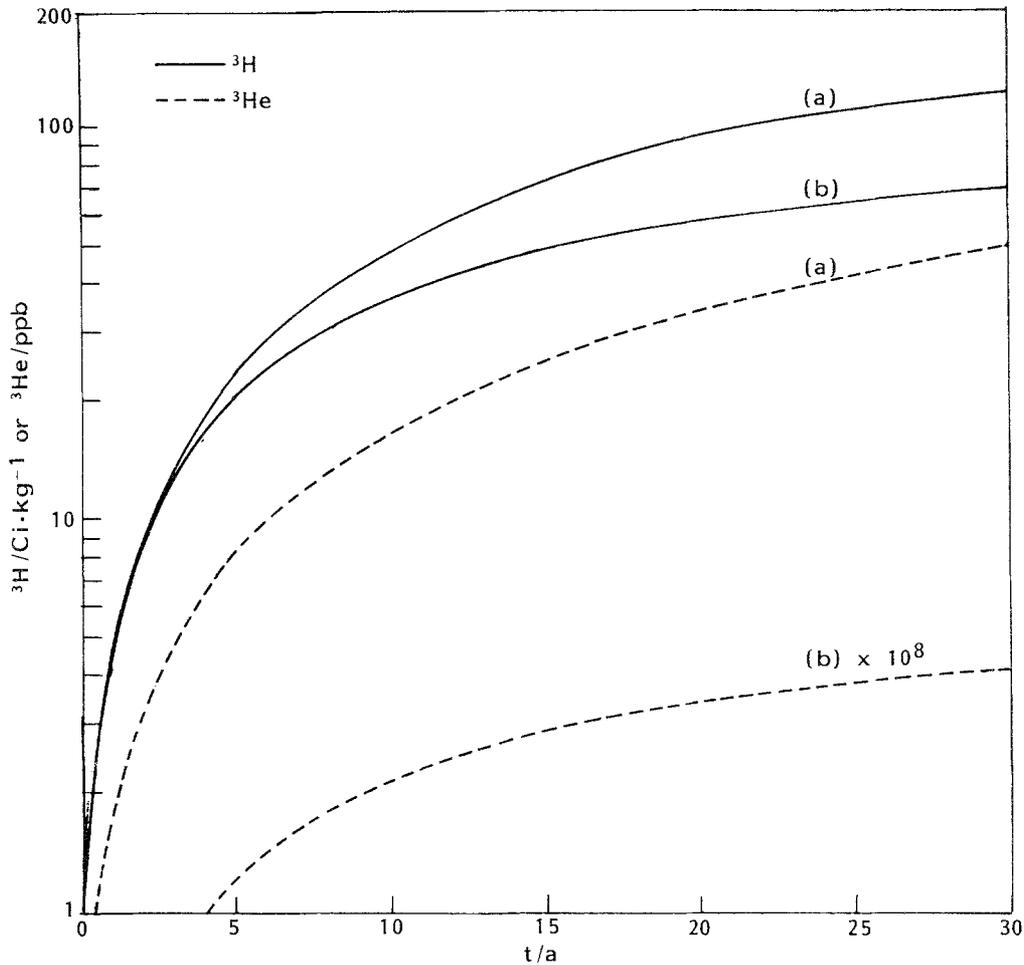**Fig. 2.** The variation of tritium and helium-3 concentrations with time at an effective neutron flux of $3.16 \times 10^{21}$ cm$^{-2} \cdot$a$^{-1}$ for a helium-3 escape rate constant of (a) 0 a$^{-1}$, and (b) $10^{10}$ a$^{-1}$.

increases monotonically with time as does the He concentration. For $\lambda_{He} = 0$ a$^{-1}$, the He concentration increases more slowly at the higher flux due to the increasing significance of the contribution of the reaction $^3$He(n,p)$^3$H to the T inventory, although fortuitously, the levels of He after 30 years are about the same for both values of the flux. At the higher value of the escape rate constant, the He concentrations for both fluxes are equally low and the contribution of the reaction $^3$He(n,p)$^3$H to the T inventory is insignificant. As mentioned in Sec. 2.2, the flux is not strictly an adjustable process parameter, and in subsequent simulations it is fixed at the value entered in Table I, this value being representative of a typical 500–600 MWe CANDU unit.

## 3.2. Effects of Helium-3 Retention Time

One notices, from either Fig. 1 or Fig. 2, that in 30 years the T inventory almost doubles as one goes from extremes in the He escape rate constant $\lambda_{He}$, suggesting that the contribution of the reaction $^3$He(n,p)$^3$H to the T activities in the heavy water systems of a CANDU reactor matches that arising from the D activation reaction $^2$H(n,$\gamma$)$^3$H. This occurs due to the fact[6] that $\sigma_{He} \simeq 10^7\sigma_D$, and in spite of the low concentration of $^3$He, arising from the $\beta^-$ decay of T, over D. Indeed, for this reason, Dombra,[24] in investigating the rate of increase of T inventories in several CANDU and heavy water research reactors, suggested that the $^3$He(n,p)$^3$H reaction might have

an appreciable effect on T concentration trends. Arnold[25] has compared historical T concentration trends of moderator and heat transport systems with simple model calculations which neglect the influences of the $^3$He(n,p)$^3$H reaction. The excellent agreement between the historical and predicted data attests to the essential validity of the models and their underlying basis, which implicitly includes the assumption that the mean residence time $\lambda_{He}^{-1}$ of He in the heavy water systems is small.

Holford and Osborne,[26] by assuming that the replenishment of D is sufficient to replace the escape of He from the system, have analytically solved Eq. (1) and have exhaustively studied the effects of He retention in CANDU heavy water systems. The results

reported in the previous section do not make the assumption of species conservation, but on the time scale of interest, the results independently confirm the findings of Holford and Osborne. Andrews and Mountford-Smith[27] have also studied the free evolution of T trends in CANDU reactors, under the assumption that $n_D(t)$ is constant, and by the inclusion of phenomenological rate constants for the fractional T inventory that is irrecoverably lost or that undergoes upgrading, in their defining coefficient matrix, Eq. (2), but with the neglect of free He escape from the system. Notwithstanding the fact that the mean retention time $\lambda_{He}^{-1}$ of He in the moderator is undoubtedly small, the external introduction of He, as available from the tritium removal facility,[2] into
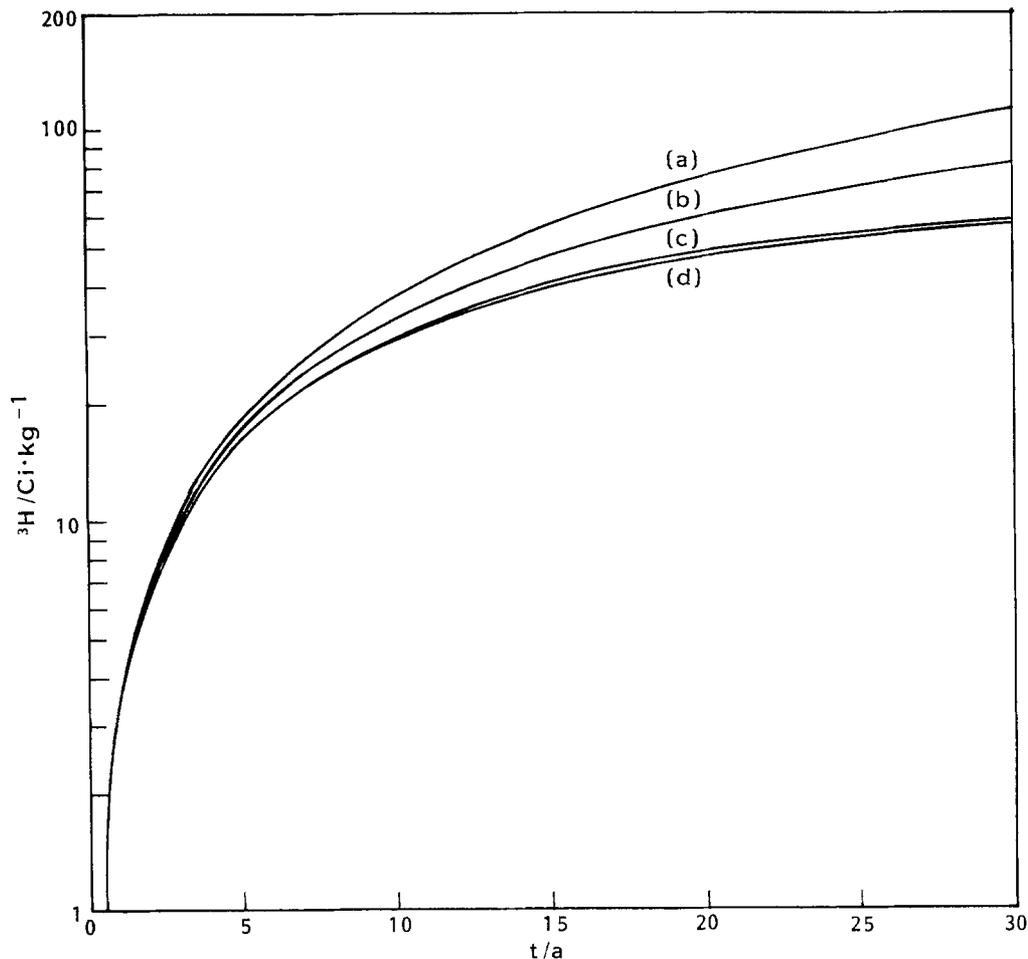


Fig. 3. The variation of tritium activity with time at an effective neutron flux of $2.37 \times 10^{21}$ cm$^{-2} \cdot$a$^{-1}$ and with the addition of 10 moles of helium-3 every 5 years over a duration of $10^{-2}$ a for a helium-3 escape rate constant of (a) 0 a$^{-1}$, (b) 10 a$^{-1}$, (c) $10^3$ a$^{-1}$, and (d) $10^5$ a$^{-1}$.

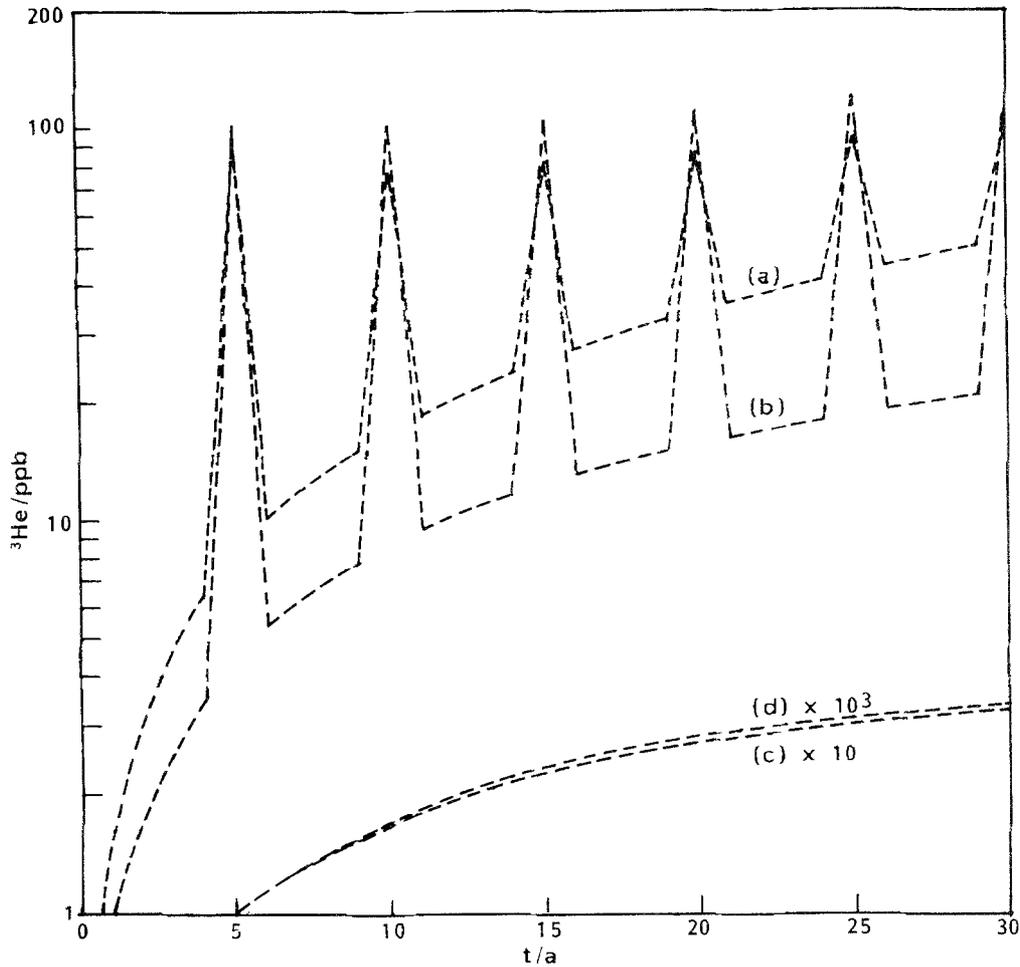Fig. 4. The variation of helium-3 concentration with time at an effective neutron flux of $2.37 \times 10^{21}$ cm$^{-2} \cdot$a$^{-1}$ and with the addition of 10 moles of helium-3 every 5 years over a duration of $10^{-2}$ a for a helium-3 escape rate constant of (a) 0 a$^{-1}$, (b) 10 a$^{-1}$, (c) $10^3$ a$^{-1}$, and (d) $10^5$ a$^{-1}$.

the moderator system should render the $^3$He(n,p)$^3$H reaction as an important contributor to evolving T inventories.

The total T and He inventories resulting from the free evolution of an initial D inventory and from the addition of 10 moles of helium-3 every 5 years with $\lambda_{He} = 0$, 10, $10^3$, and $10^5$ a$^{-1}$, embracing a range of mean retention times extending from total retention to the virtual complete escape of helium-3, over a 30 year period at an effective neutron flux of $2.37 \times 10^{21}$ cm$^{-2} \cdot$a$^{-1}$ are displayed in Figs. 3 and 4, respectively. Each input is delivered over a duration of $10^{-2}$ a. As expected, both the T activities and the He concentrations increase as time progresses and decrease as the retention time of helium-3 decreases.

The T inventories shown in Fig. 3 are higher than those available from the free evolution of the initial D inventory, and at the end of the 30 years the enhancement factors for the cases $\lambda_{He} = 0$, 10, $10^3$, and $10^5$ a$^{-1}$ are 1.05, 1.07, 1.11, and 1.11, respectively. The He concentrations shown in Fig. 4 are marginally higher than the values prevailing in the absence of additions except for the rapid increase and subsequent decrease at the delivery times of the pulsed additions in the case of lower $\lambda_{He}$ values. There are now two sources of He in the system, the $\beta^-$ decay of T and the external additions. If $\lambda_{He}$ is small, both sources of He contribute to the T inventories via the reaction $^3$He(n,p)$^3$H, while if the retention time is short, He escapes regardless of its origin.
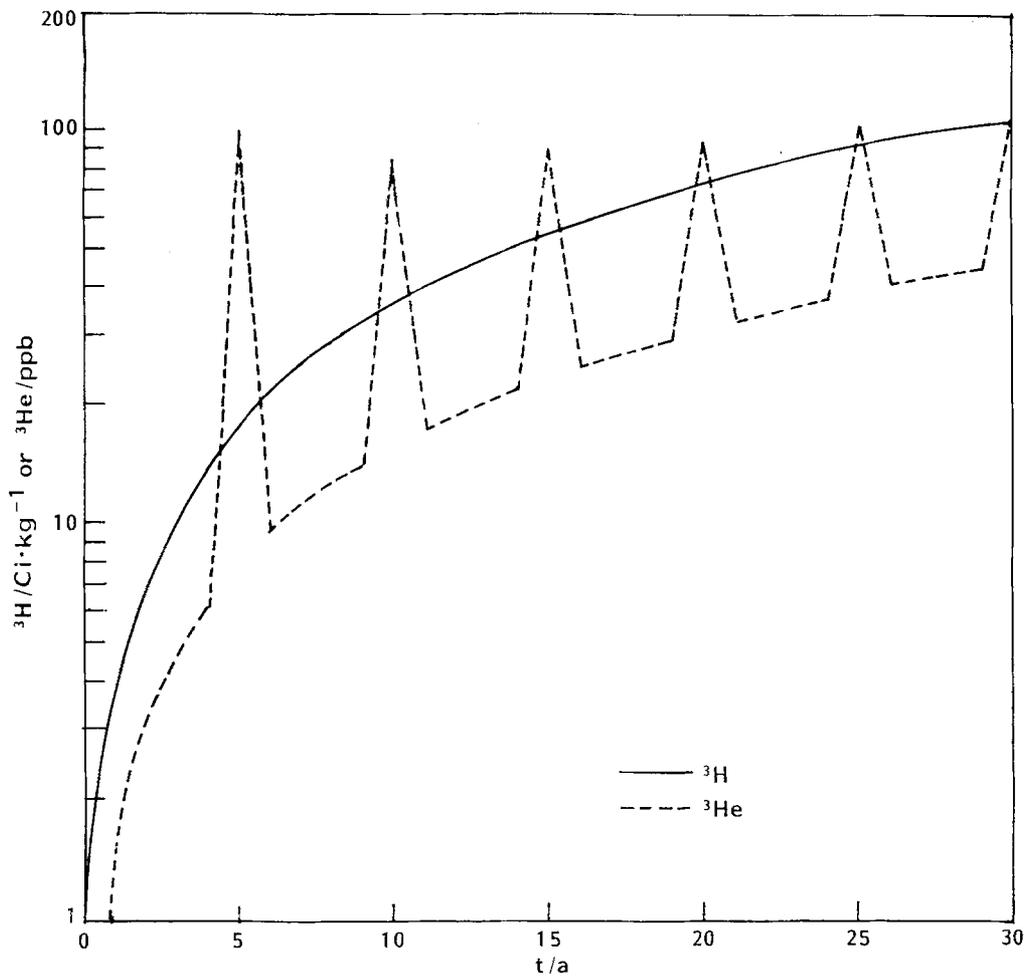
**Fig. 5.** The variation of tritium and helium-3 concentrations with time at an effective neutron flux of $2.37 \times 10^{21}$ cm$^{-2} \cdot$a$^{-1}$ and with the impulsive addition of 10 moles of helium-3 every 5 years for a helium-3 escape rate constant of 1 a$^{-1}$.

**Table II.** Tritium and Helium-3 Concentrations at the Delivery Times of 10 Mole Additions of Helium-3, in an Effective Flux of $2.37 \times 10^{21}$ cm$^{-2} \cdot$ a$^{-1}$ and for a Helium-3 Escape Rate Constant of 1 a$^{-1}$, at Various Input Durations

| | $^3$H/Ci·kg$^{-1}$ (duration/a) | | | | | $^3$He/ppb (duration/a) | | | | |
|---|---|---|---|---|---|---|---|---|---|---|
| Time/a | 1 | $10^{-1}$ | $10^{-2}$ | $10^{-3}$ | 0 | 1 | $10^{-1}$ | $10^{-2}$ | $10^{-3}$ | 0 |
| 5 | 19.03 | 18.49 | 18.12 | 18.06 | 18.06 | 8.05 | 63.30 | 101.51 | 108.26 | 108.31 |
| 10 | 37.68 | 37.31 | 37.04 | 36.99 | 36.99 | 16.05 | 54.41 | 82.19 | 87.25 | 87.25 |
| 15 | 55.98 | 55.64 | 55.39 | 53.34 | 53.34 | 23.91 | 58.99 | 84.66 | 89.35 | 89.35 |
| 20 | 73.92 | 73.59 | 73.35 | 73.30 | 73.30 | 31.60 | 65.45 | 90.32 | 94.87 | 94.87 |
| 25 | 91.51 | 91.19 | 90.95 | 90.91 | 90.91 | 39.15 | 72.45 | 96.96 | 101.45 | 101.45 |
| 30 | 108.76 | 108.76 | 108.21 | 108.16 | 108.16 | 46.56 | 79.61 | 103.98 | 108.45 | 108.45 |

However, when $\lambda_{He}$ is large, external input of He to the existing freely evolving low levels of this species amplifies the quantity of He exposed to the flux, and hence the rate of He activation to T, and results in a larger enhancement factor for T production.

It is clear from Fig. 4 that at the lower values of $\lambda_{He}$ there is a sharp rise in the He concentration at the delivery times of each input. This occurs because the rate of delivery of each input of He over a duration of $10^{-2}$ a exceeds the removal rate, whether

through its activation to form T or its escape from the moderator. As each pulsed input peaks, the rate of formation of T and the rate of escape of He both increase, and as the input subsides, the He concentration decreases. The higher peak He concentrations when $\lambda_{He} = 10$ a$^{-1}$ over the peak concentrations when $\lambda_{He} = 0$ a$^{-1}$ is a consequence of the higher rate of He removal, through activation, when $\lambda_{He} = 0$ a$^{-1}$ over the total removal rate, through both activation and escape, when $\lambda_{He} = 10$ a$^{-1}$. At higher values of $\lambda_{He}$,
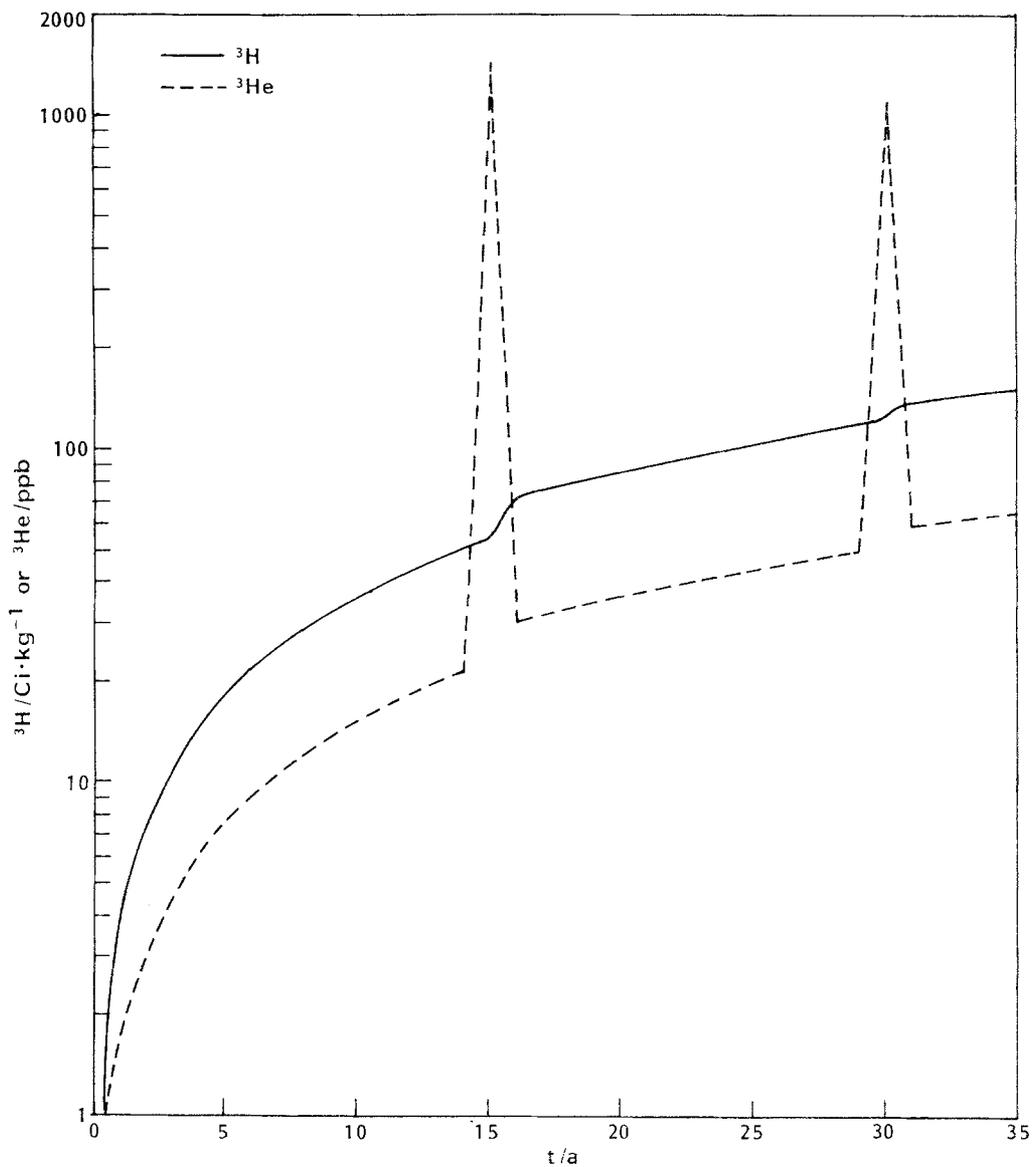


Fig. 6. The variation of tritium and helium-3 concentrations with time at an effective neutron flux of $2.37 \times 10^{21}$ cm$^{-2} \cdot$ a$^{-1}$ and with the addition of 150 moles of helium-3 every 15 years over a duration of $10^{-2}$ a for a helium-3 escape rate constant of 1 a$^{-1}$.

the evolution of He concentrations do not exhibit sharp increases at delivery times due to the rapid escape of the external inputs of helium-3.

The enhancement of T production, resulting from the introduction of a total of 60 moles of helium-3 at regular intervals over a 30 year period, ranges from 5 to 11% as the mean retention time of helium-3 decreases. Although this represents a small increase, it does indicate the potential for increasing T production rates through the external addition of helium-3, with the *relative* enhancement increasing as the mean retention time of He decreases. Notice that in Fig. 4 and subsequent figures, the peak He concentrations are displayed as starting 1 year prior to the delivery

times and finishing 1 year afterwards; consequently, the evolutions of He concentrations appear as discontinuous curves. This is an artifice of having conducted the simulations such that the computed data were outputted on an annual basis. In fact the He concentrations evolve in a continuous manner and peak over temporal widths determined by the input durations. The details of these transients are unimportant and are not displayed.

### 3.3. Effects of Input Durations

The effect of varying the duration of the additions of He on T activities was investigated for
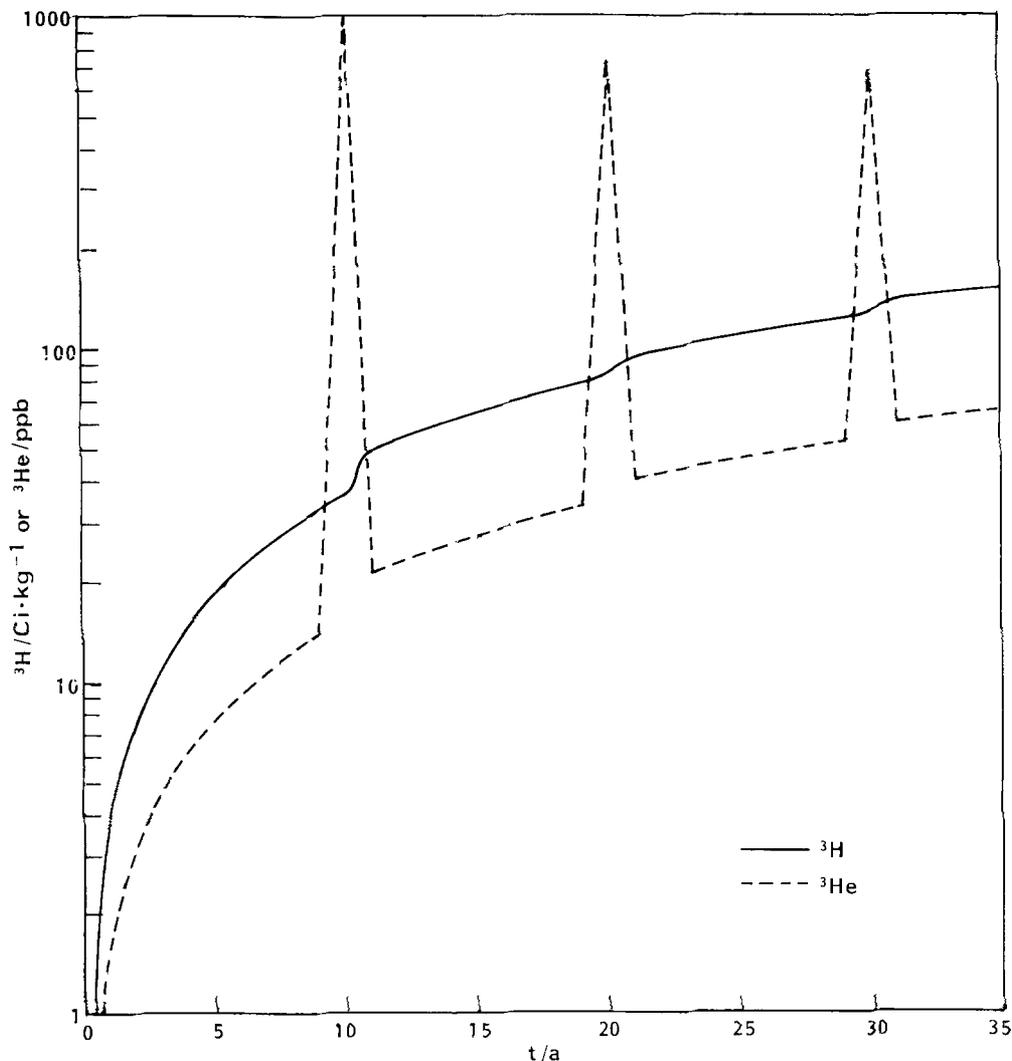


Fig. 7. The variation of tritium and helium-3 concentrations with time at an effective neutron flux of $2.37 \times 10^{21}$ cm$^{-2} \cdot$a$^{-1}$ and with the addition of 100 moles of helium-3 every 10 years over a duration of $10^{-2}$ a for a helium-3 escape rate of 1 a$^{-1}$.

pulse durations of $10^{-3}, 10^{-2}, 10^{-1}$, and 1 a. In each case 10 moles of He were added every 5 years to an initial D inventory over a 30 year period at an effective neutron flux of $2.37 \times 10^{21}$ cm$^{-2} \cdot$a$^{-1}$ with $\lambda_{He}$ fixed at 1 a$^{-1}$. Figure 5 displays the T and He concentrations resulting from the impulsive (0 a duration) additions of He under these conditions. At the delivery times, the rate of input of He exceeds its removal rate, and a sharp rise in its concentration is observed. The peak value is highest on the first addition, drops on the second addition, and increases on each subsequent input as the removal rate of He increases. The enhancement factor for T production

at the end of 30 years is 1.05, and is a maximum following each input. Between inputs, the enhancement factor decreases somewhat due to the decay of T and the removal of He from the system. The duration of the inputs has minimal effect on the enhancement factor for T production.

One can appreciate this by referring to Table II, which gives the T and He concentrations at the delivery times for various input durations. As their durations increase, the inputs are added over more protracted periods centered at the delivery times, and the activation of He compensates for T decay so that the T inventory is further enhanced, albeit to a small
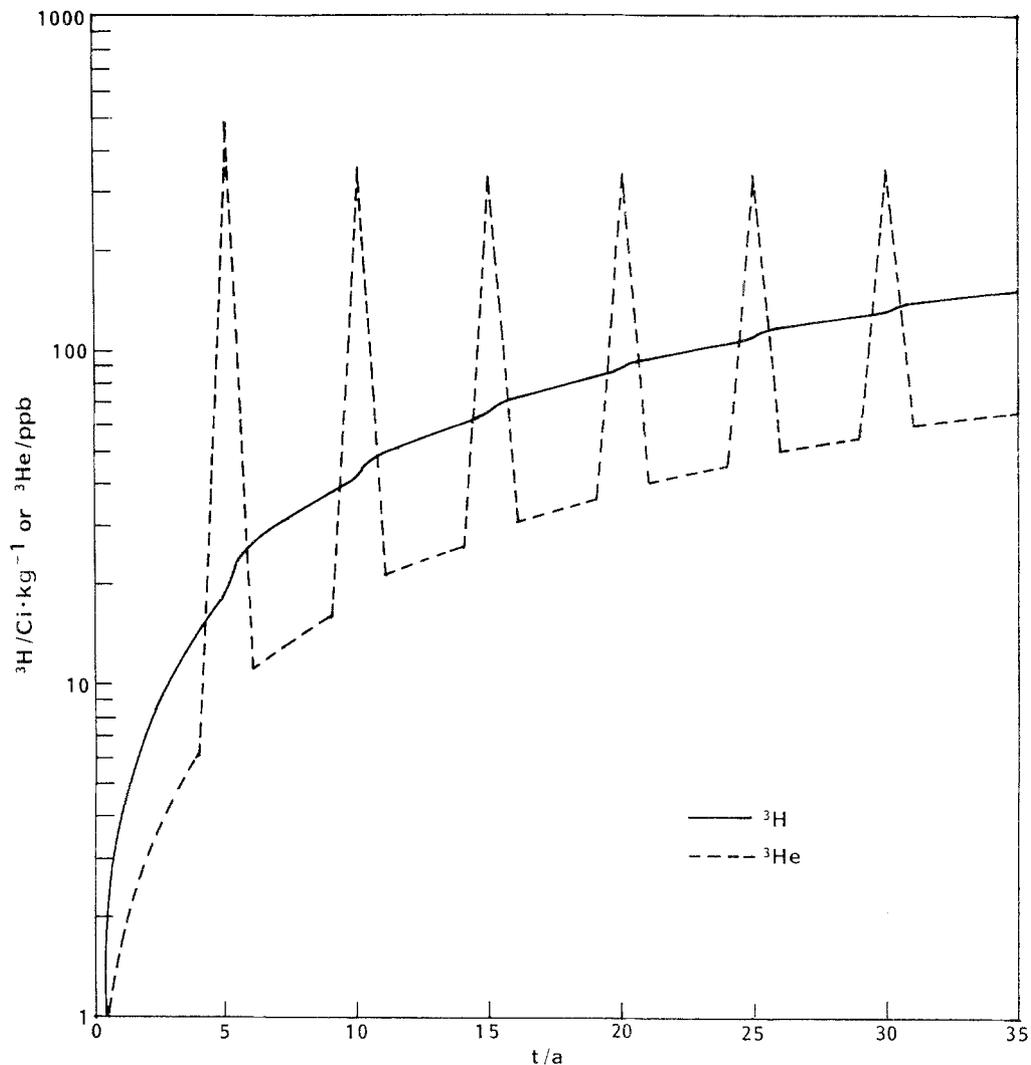


Fig. 8. The variation of tritium and helium-3 concentrations with time at an effective neutron flux of $2.37 \times 10^{21}$ cm$^{-2} \cdot$a$^{-1}$ and with the addition of 50 moles of helium-3 every 5 years over a duration of $10^{-2}$ a for a helium-3 escape rate constant of 1 a$^{-1}$.

extent. After the impact of an input has subsided, the T activities are essentially the same (as are the He concentrations) no matter what the duration of the input, and freely evolve until the arrival of the next addition of He. As time progresses, the effects of input duration become even less pronounced since the addition of a fixed amount of He at each delivery is outweighed by the increasing decay rate of T and the increasing freely evolving He concentrations. Thus, for example, from Table II it is apparent that after the first addition both the T and He concentrations are the same for the impulsive and the $10^{-3}$ a duration inputs.

As expected, for short duration inputs, the evaluation of the instantaneous fractional inventories for D, T, and He through Eq. (8), and the cumulative amount added through Eq. (10), are numerically equivalent to the results obtained through Eqs. (11) and (13), respectively.

### 3.4. Effects of Input Delivery Times and Quantities Added

In investigating the effects of varying the input delivery times and the quantities of helium-3 being added, simulations were carried out for the addition of a total of 300 moles of helium-3 over a 30 year period to an initial D inventory in an effective neutron flux of $2.37 \times 10^{21}$ cm$^{-2} \cdot$a$^{-1}$ with the helium-3 escape rate constant fixed at 1 a$^{-1}$. The resulting variations in T and He concentrations with time for the addition of 150 moles every 15 years, 100 moles every 10 years, 50 moles every 5 years, and 10 moles every years, with each input being delivered over a duration of $10^{-2}$ a, are displayed in Figs. 6 through 9, respectively. The evolutionary nature of the T activities and He concentrations in each case is similar to that described in Secs. 3.2 and 3.3. Since larger quantities of helium-3 are being added, the enhance-
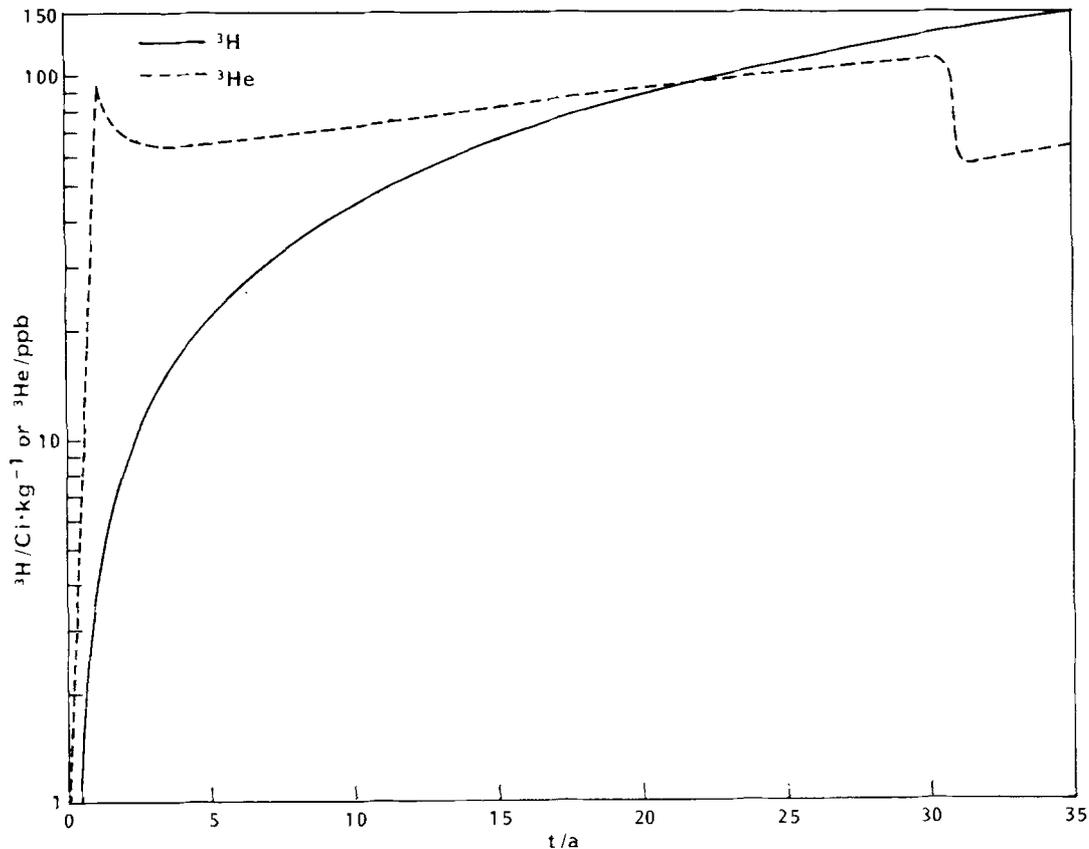


Fig. 9. The variation of tritium and helium-3 concentrations with time at an effective neutron flux of $2.37 \times 10^{21}$ cm$^{-2} \cdot$a$^{-1}$ and with the addition of 10 moles of helium-3 every year over a duration of $10^{-2}$ a for a helium-3 escape rate constant of 1 a$^{-1}$.

ments of T activities at each delivery are more pronounced as evident by the undulations in the T inventories depicted in Figs. 6–8. Such undulations do not appear in the case when 10 moles of helium-3 are added on an annual basis (Fig. 9). The He concentration increases rather rapidly with the first few deliveries and then decreases as the removal rate starts to increase. The evolving He concentration then increases at a slower rate, as determined by the balance between T decay and the external additions on the one hand and the activation and escape of helium-3 on the other, until just beyond the final delivery, at which time it decreases and freely evolves. Thus, more frequent additions serve to maintain a

higher concentration of helium-3 over the entire 30 year period with a concomitant greater enhancement of the T production inventories. The enhancement factors for T production after 30 years on the addition of a total of 300 moles of helium-3 at the rates of 150 moles every 15 years, 100 moles every 10 years, 50 moles every 5 years, and 10 moles every year are 1.19, 1.22, 1.26, and 1.28, respectively.

As previously stated, the quantities and delivery times of helium-3 inputs are determined by its availability. It has been estimated[4] that at least 100 moles of helium-3 can be supplied annually from Ontario Hydro's centralized tritium removal facility.[2] Figure 10 displays the evolving T and He concentrations
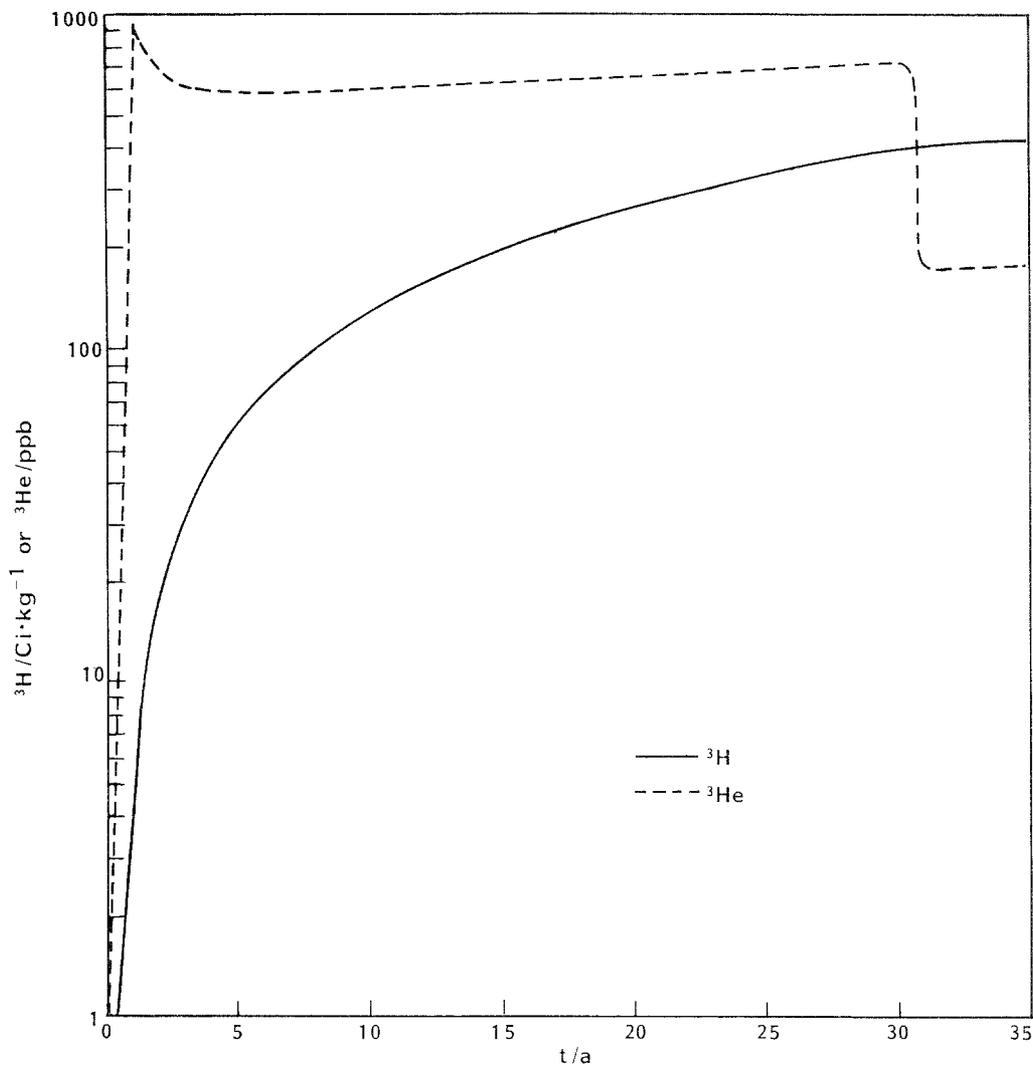


Fig. 10. The variation of tritium and helium-3 concentrations with time at an effective neutron flux of $2.37 \times 10^{21}$ cm$^{-2} \cdot$a$^{-1}$ and with the addition of 100 moles of helium-3 every year over a duration of $10^{-2}$ a for a helium-3 escape rate constant of 1 a$^{-1}$.

resulting from the annual addition of this quantity of helium-3 over a duration of $10^{-2}$ a to an initial D inventory in an effective neutron flux of $2.37 \times 10^{21}$ $\text{cm}^{-2} \cdot \text{a}^{-1}$ with the helium-3 escape rate constant fixed at $1 \text{ a}^{-1}$. After the 30 year period, the enhancement factor for T production is 3.77, representing a nearly fourfold increase over the freely evolving inventory.

## 4. DISCUSSION

The foregoing results indicate that the frequent addition of readily available helium-3 to the moderator water of a hypothetical 500–600 MWe CANDU reactor could greatly enhance T production inventories for fusion fuel applications through the activation of helium-3 via $^3\text{He}(n,p)^3\text{H}$. The resultant enhancement factors are essentially insensitive to the durations of the deliveries but are highly dependent upon the prevailing moderator neutron flux and the rate at which helium-3, whether originating from T decay or through external additions, escapes from the moderator water to the cover gas system.

The most important parameter determining whether or not the external addition of He can significantly induce the enhancement of T production is the escape rate constant for helium-3, $\lambda_{\text{He}}$. As demonstrated in Sec. 3.2, the *relative* enhancement increases as the mean retention time of He decreases. The moderator circulation system and the natural helium ( $\sim 100\%$ $^4\text{He}$) cover gas circuit in earlier CANDU designs[28] conspire to reduce the residence time in the moderator water. As previously mentioned, the mean retention time of helium-3 in these designs is known to be small. In later designs,[29] the cover gas is of smaller volume and there is no moderator dump tank. Efforts are currently underway[30] to measure the $^3\text{He}$-$^4\text{He}$ distribution in the cover gas of these designs and hence to establish the helium-3 residence time in the moderator water. Since the half-life for $D_2$ and $CO_2$ transfer across the moderator water-cover gas interface is in the 2–5 day range,[30] one anticipates that the retention time of $^3\text{He}$ in the moderator water will also be small. One may estimate the mean retention time of helium in the moderator by its diffusional time. Taking the diffusion constant of He in $D_2O$ in the temperature range 40–70°C as $5 \times 10^4$ $\text{m}^2 \cdot \text{a}^{-1}$ and the diffusional length as 3.1 m (the radius of a typical CANDU calandria vessel), one estimates the mean residence time $\lambda_{\text{He}}^{-1} \sim 10 \text{ m}^2/2 \times 5 \times 10^4 \text{ m}^2 \cdot \text{a}^{-1} = 10^{-4}$ a.

The degree of solubility of $^3\text{He}$ in heavy water is the most significant physicochemical determinant of its transfer to solution phase, and hence its mean retention time in the moderator water. The solubility of $^3\text{He}$, in turn, is a function of the pressure of the natural helium cover gas system (ca. 25–30% above atmospheric) in contact with the moderator, the temperature of the moderator (40–70°C), and the concentration of impurities in the moderator. The available data for the solubility of $^4\text{He}$ in $H_2O$ in the range 0–80°C have recently been critically evaluated.[31] Data from eight sources were used to obtain a correlation of the mole fraction solubility as a function of temperature, at 101.325 kPa partial pressure of helium and accurate to within 0.5%. Not included in this evaluation are the more recent results of Porter and Clynne,[32] whose data, however, have been severely criticized by Crovetto et al.[33] Weiss[34] has reported reliable measurements of the solubility of $^3\text{He}$ and $^4\text{He}$ in water and seawater in the temperature range 0–40°C and observes an isotope effect of 1.2% at 0°C, which appears to decrease with increasing temperature. The only data[35] on the solubility of $^4\text{He}$ in $D_2O$ and $D_2O + H_2O$ mixtures in the range 10–45°C were assessed as being too erratic for inclusion in the solubility correlation.[31] The isotope effects on the solubility of helium in water will be small, whether the effects arise due to the solute ($^3\text{He}$ or $^4\text{He}$) or the solvent ($H_2O$ or $D_2O$). Further, the solubility of helium in water will be small. These expectations are based on consideration of the cavity creation and the solute-solvent interaction contributions[36] to the energetics involved in the dissolution of helium in water. Thus, ignoring isotope effects on solubility and neglecting the presence of impurities in the moderator, one estimates the weight solubility of $^3\text{He}$ in $D_2O$ at a pressure of 101.325 kPa and in the temperature range 40–75°C, from the correlation given in,[31] to be $3.5$–$3.94 \times 10^4$ $\text{mol} \cdot \text{kg}^{-1}$. Hence, in a 300 000 kg moderator one can dissolve 105–118 moles of $^3\text{He}$. This is of the order of the quantity of helium-3 available on an annual basis from the centralized tritium removal facility and of the order of the amount added in the final simulation discussed in Sec. 3.4.

Whatever the origin of He in the system, the fractional inventory that becomes activated to produce T is $\phi\sigma_{\text{He}}/(\lambda_{\text{He}} + \phi\sigma_{\text{He}})$ while the fractional inventory that escapes to the cover gas is $\lambda_{\text{He}}/(\lambda_{\text{He}} + \phi\sigma_{\text{He}})$. If, as is likely to be the case, $\lambda_{\text{He}} \gg \phi\sigma_{\text{He}}$, then most of the He escapes. Since current CANDU designs are not such as to promote the saturation of

the moderator with the cover gas, one may regard the escape of helium-3 as an irretrievable loss of raw material for T production, especially as the technology for the recovery of $^3$He from the $^4$He cover gas is not available on an economically viable basis. However, were one to directly activate the available helium-3, it is easy to show that the instantaneous fractional inventories for He and T are given by

$$n_{He}(t) = \exp\left[ -\frac{1}{2}(\lambda_T + \phi\sigma_{He})t \right]$$
$$\times \left\{ \frac{\lambda_T - \phi\sigma_{He}}{\lambda_T + \phi\sigma_{He}} \sinh\left[ \frac{1}{2}(\lambda_T + \phi\sigma_{He})t \right] \right.$$
$$\left. + \cosh\left[ \frac{1}{2}(\lambda_T + \phi\sigma_{He})t \right] \right\} \quad (14)$$

and

$$n_T(t) = \frac{2\phi\sigma_{He}}{\lambda_T + \phi\sigma_{He}} \exp\left[ -\frac{1}{2}(\lambda_T + \phi\sigma_{He})t \right]$$
$$\times \sinh\left[ \frac{1}{2}(\lambda_T + \phi\sigma_{He})t \right] \quad (15)$$

respectively, and the corresponding steady state inventories are

$$n_{He}(\infty) = \frac{\lambda_T}{\lambda_T + \phi\sigma_{He}} \quad (16)$$

and

$$n_T(\infty) = \frac{\phi\sigma_{He}}{\lambda_T + \phi\sigma_{He}} \quad (17)$$

Obviously, if $\phi\sigma_{He} \gg \lambda_T$, then $n_T(\infty) \gg n_{He}(\infty)$, and almost the entire initial He inventory will be eventually activated to T. Indeed, from Eqs. (14) and (15) one can show that the half-life of the reaction $^3$He(n,p)$^3$H is given by

$$t_{1/2} = \frac{2}{\lambda_T + \phi\sigma_{He}} \coth^{-1}\left\{ \frac{5\phi\sigma_{He} - \lambda_T}{\lambda_T + \phi\sigma_{He}} \right\} \quad (18)$$

which, under the assumption that $\phi\sigma_{He} \gg \lambda_T$, reduces to

$$t_{1/2} \simeq \frac{2}{5\phi\sigma_{He}} \quad (19)$$

Using the data in Table I, Eq. (19) gives a half-life of about 11 days. Thus, the direct activation of He, as opposed to its activation in the moderator system, would be almost totally complete within a few weeks without any loss of helium-3.

Finally, we close on a somewhat academic point. If $\lambda_{He}$ is nonzero, the coefficient matrix $\underline{N}$ as defined in Eq. (2) will not have a vanishing eigenvalue. Consequently, the solution of Eq. (1) or Eq. (4), as given by Eq. (3) or Eq. (5), will be asymptotically stable[7,10] provided $Re(\lambda) < 0$ for $\lambda$ any eigenvalue of $\underline{N}$, and eventually vanishes. For the time scales of interest here, this consequence of having an open system will not manifest itself.

## 5. CONCLUSIONS

The external addition of helium-3 to the heavy water moderator system of a CANDU reactor will promote the production of tritium to supply emerging fusion fuel requirements. The frequent addition of helium-3 will ensure that the degree of enhancement of tritium activity is maximized. The *relative* enhancement factor for tritium production is greater the larger the escape rate of helium-3 to the moderator cover gas system. The duration of input deliveries of helium-3 to the moderator has negligible influence on the enhancement of tritium production. The effect of the thermal neutron flux on the evolving tritium activities is first order.

The most serious shortcoming of the proposed scheme for enhancing the production of tritium is the irretrievable loss of helium-3 to the natural helium cover gas system, resulting from its short retention time in the moderator and due essentially to the low solubility of helium-3 in heavy water in the temperature range 40–75°C under near atmospheric overpressures. If the primary source of helium-3 is through the decay of in-storage tritium as supplied by the centralized tritium removal facility, then its direct activation to tritium in an appropriate closed loop in a thermal neutron flux would be essentially complete within a few weeks, without the significant loss of a valuable raw material. One possibility that could be explored is the seeding of the annulus gas system with helium-3. The tritium hydride resulting from helium-3 activation can be removed by adsorption on a uranium getter bed[37] or can be catalytically oxidized and removed by absorption on a suitable molecular sieve.

## ACKNOWLEDGMENTS

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