ABSTRACT

A model has been developed to accurately calculate the nuclide inventories of the target constituents of Inertial Fusion Energy (IFE) reactors such as HYLIFE-II. It can explicitly account for (1) the combined effects of activation during target implosion (by a high-amplitude flux) and while passing through the reactor chamber (by a low-amplitude flux); (2) decay during circulation in the primary coolant loop, after extraction from the coolant loop, and before re-insertion into the reactor chamber as a new target; (3) continuous extraction and feed-in of target material; and (4) replacement of part of the activation products by makeup materials. The solution strategy uses transition factors – the ratio of the amount of created nuclides to the initial amount – for each system component.

I. INTRODUCTION

The change in the isotopic composition of the target constituents in certain IFE reactors, such as the HYLIFE-II reactor, considered in this work, is a result of a complex process. Consider, for example, the activation of the target hohlraum high-Z material.2,3 Figure 1 shows the high-Z material circulation in the HYLIFE-II reactor. After being exposed to a high-amplitude flux during the target implosion, the high-Z material is deposited in the Flibe (molten fluorides of Li and Be) coolant/breeder volume which surrounds the target. The high-Z material then circulates with the Flibe many times through the primary cooling system. Each time it reaches the reactor cavity at the center of which is the next target to be imploded, it is irradiated with a flux of a lower amplitude and a softer spectrum. The radionuclides decay in between their passes through the reactor chamber (approximately every 20 seconds in HYLIFE-II). High-Z material is continuously extracted from the Flibe and circulates through a storage for radioactive cooling, the target fabrication plant and target storage before it is re-introduced, in a new target, into the reactor chamber. Some of the activation and decay products of the high-Z material may be removed and an appropriate amount of unirradiated high-Z material can be added to make up for the losses.

Approximate activation models have been used in previous studies of hohlraum high-Z material activation.4,5 Common approximations include: (1) Separating the activation in the target and in the Flibe; (2) Continuous irradiation in the Flibe using a reduced flux amplitude to account for the presence of out-of-chamber inventory of high-Z material; (3) Ignoring extraction of high-Z material from the Flibe and its circulation through the target fabrication plant or, alternatively, assuming a batch rather than continuous extraction. The purpose of the present work was to develop a model which is capable of accurate simulation of multi-flux pulsed activation accounting for circulation, extraction and makeup of target constituents (PACEM). This model can be used as a reference against which various approximations, such as in Ref. 6, can be tested.

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II. THE “PACEM” MODEL

The exact activation model developed in this work simulates the process shown in Fig. 1. It should be noted that Fig. 1 is not an exact representation of the presently conceived HYLIFE-II design. Whereas Fig. 1 depicts a single Flibe circulation loop, the present HYLIFE-II design has two loops. However, assuming only one loop makes the analysis of the validity of the various activation models more transparent. The model presented here can easily be extended to simulate more complicated scenarios.

A generic model for the time evolution of high-Z nuclide inventory can be set up by analyzing the input and output of high-Z nuclides for each element in the system. We start by calculating the time-dependent nuclide inventory inside the reactor chamber. The inventories at any other location of interest in the system can be easily inferred from the in-chamber inventory.

A. Decay in Transport Lines and in Storage Facilities

The total inventory of nuclide $i$ inside the reactor chamber is denoted by $X_i$. In order to simplify the notation and derivation, we shall use the vector $\vec{X}$ to denote the inventory of all the $N$ nuclides pertaining to the chain of interest. The rate of flow of nuclides out of a storage tank, $\dot{X}_{\text{out}}$, can be related to the incoming flow rate, $\dot{X}_{\text{in}}$, as:

$$\dot{X}_{\text{out}}(t) = \exp(-\frac{2}{\tau})\dot{X}_{\text{in}},$$

where $\tau$ is the delay time and $\frac{2}{\tau}$ is the decay matrix:

$$A = \begin{pmatrix} \lambda_1 & -\lambda_{1 \rightarrow 2} & \cdots & -\lambda_{1 \rightarrow N} \\ -\lambda_{2 \rightarrow 1} & \lambda_2 & \cdots & -\lambda_{2 \rightarrow N} \\ \vdots & \vdots & \ddots & \vdots \\ -\lambda_{N \rightarrow 1} & -\lambda_{N \rightarrow 2} & \cdots & \lambda_N \end{pmatrix}.$$
\[
\exp(-\lambda \tau) = \sum_{j=0}^{\infty} \frac{1}{j!}(-j\lambda \tau)^j.
\]  

(3)

is the matrix exponential. It is the basis for many activation codes such as ORIGEN and ACAB. It is described in many standard Linear Algebra textbooks.

Denoting the time between the extraction point and re-entry into the reactor chamber as \(\tau_e\), the incoming flow rate of nuclides is given by

\[
X_{\text{in}}(t) = \exp(-\lambda \tau_e) X_{\text{after extraction}}(t - \tau_e).
\]

(4)

The flow rate of nuclides before the extraction point is given by

\[
X_{\text{before extraction}}(t) = \exp(-\lambda \tau_1) X_{\text{out}}(t - \tau_1),
\]

in which \(\tau_1\) is the delay between the outlet of the reactor chamber and the extraction point, and \(X_{\text{out}}(t)\) is the nuclide flow rate out of the reactor chamber.

Considering the radioactive cooling storage during a time \(\tau_s\), the flow rate of recycled high-Z nuclides depends on the flow rate of recovered nuclides via

\[
X_{\text{recycled}}(t) = \exp(-\lambda \tau_s) X_{\text{recovered}}(t).
\]

(6)

The number of high-Z nuclides in one hohlraum \(X_{h}\) can similarly be expressed as a function of the inventory at the target fabrication plant:

\[
X_{h, \text{ before pulse}}(t) = \exp(-\lambda \tau_p) X_p(t - \tau_p),
\]

(7)

in which we assume a delay time \(\tau_p\) at the pellet storage (Fig. 1).

B. The Extraction Plant

The flow rate of nuclides after the extraction plant is given by the flow rate of nuclides before the extraction plant minus the rate of nuclide extraction:

\[
X_{\text{after extraction}}(t) = X_{\text{before extraction}}(t) - X_{\text{extracted}}(t).
\]

(8)

The nuclide extraction rate is determined by the extraction constant \(K_i\) for nuclide \(i\), which is defined as the fraction of nuclides \(i\) that is extracted per cycle:

\[
X_{\text{extracted}, i}(t) = K_i X_{\text{before extraction}, i}(t).
\]

(9)

To put this equation into matrix form, we define an extraction matrix \(K\) as a diagonal matrix containing the extraction constants:

\[
K = \text{diag}(K_1, K_2, ..., K_N),
\]

(10)

which yields

\[
X_{\text{extracted}}(t) = K X_{\text{before extraction}}(t).
\]

(11)

We assume that a fraction of the nuclides may be lost in the extraction process. Therefore we introduce a recovery efficiency coefficient \(R_i\) for each nuclide. Since the extraction process is most likely a chemical process, all nuclides of a given element will have the same extraction constant and recovery efficiency coefficient. As in the case of the extraction coefficient, we can write the recovery efficiency coefficients as a diagonal matrix. The flow rate of recycled nuclides will then be

\[
X_{\text{recycled}}(t) = R_i X_{\text{extracted}}(t).
\]

(12)

By setting the coefficient \(R_i\) to zero, we can simulate a complete removal of nuclide \(i\) from the system, for instance, into a waste stream.

C. The Reactor Chamber

Next, we consider the activation that takes place during a pulse. Since a pulse lasts a split second, radioactive decay during a pulse can be ignored. The change in the in-chamber nuclide inventory consists of two terms corresponding to activation in the hohlraum and activation in the Flibe. We will consider the hohlraum activation term first.

The rate of change of the nuclide inventory \(X_{h,i}\) in a hohlraum is given by

\[
\frac{d}{dt} X_{h,i}(t) = -\sigma_i \phi_h X_{h,i} + \sum_{j=1}^{N} \sigma_{j \rightarrow i} \phi_h X_{h,j},
\]

(13)

in which \(\phi_h\) is the neutron flux multiplied by the cross-section averaged over the neutron energy spectrum that
exists inside the hohlraum. In matrix form, the above equation can be written as

\[ \frac{d}{dt} X_h(t) = \sigma \phi_h X_h, \]  

(14)
in which the cross section matrix is defined as

\[ \sigma = \begin{pmatrix} -\sigma_1 & \sigma_{2 \rightarrow 1} & \cdots & \sigma_{N \rightarrow 1} \\ \sigma_{1 \rightarrow 2} & -\sigma_2 & \cdots & \sigma_{N \rightarrow 2} \\ \vdots & \vdots & \ddots & \vdots \\ \sigma_{1 \rightarrow N} & \sigma_{2 \rightarrow N} & \cdots & -\sigma_N \end{pmatrix}, \]

(15)
The nuclide inventory at the end of the pulse is given by

\[ X_{h, \text{after pulse}}(t) = \exp(\sigma \Psi_h) X_{h, \text{before pulse}}(t), \]

(16)
in which \( \Psi_h \) is the hohlraum neutron fluence in one shot. The hohlraum inventory after a shot, \( X_{h, \text{after pulse}} \), will end up in the Flibe.

The rate of change of nuclide \( X_i \) in the Flibe due to activation in the reactor chamber is given by

\[ \frac{d}{dt} X(t) = \sigma \Psi_c X, \]

(17)
in which the subscript \( c \) refers to averaging over the neutron spectrum in the reactor chamber Flibe. The nuclide inventory after the pulse is then given by

\[ X_{\text{after pulse}}(t) = \exp(\sigma \Psi_c) X_{\text{before pulse}}(t), \]

(18)
in which \( \Psi_c \) is the neutron fluence in the reactor chamber Flibe in one shot.

Combining activation in the hohlraum and in the Flibe (equations (16) and (18)) yields the high-Z nuclide inventory after a shot:

\[ X_{\text{after pulse}}(t) = \exp(\sigma \Psi_h) X_{h, \text{before pulse}}(t) + \exp(\sigma \Psi_c) X_{\text{before pulse}}(t), \]

(19)

D. Target Fabrication Plant

The inventory at the target fabrication plant consists of recycled nuclides as well as makeup nuclides:

\[ X_p(t) = \dot{X}_m(t) + \dot{X}_{\text{recycled}}(t), \]

(20)
in which the left-hand-side is multiplied by the pulsing frequency \( f \) to find the flow rate of hohlraum high-Z nuclides. The sum of all high-Z nuclides available at the target fabrication plant should equal the total fixed number \( H \) of high-Z nuclides that are needed for one hohlraum, multiplied by the number of targets to be made. This requirement defines the make-up rate, i.e., the rate of feed-in of high-Z material to the system:

\[ \dot{X}_m(t) = nH - n(1,1,\ldots,1) \dot{X}_{\text{recycled}}, \]

(21)
in which \( n \) is the vector that describes the natural abundance of the high-Z material being fed for hohlraum fabrication. In this equation, we multiplied the column vector \( \dot{X}_{\text{recycled}}(t) \) by the row vector \( (1,1,\ldots,1) \) to find the total number of recycled nuclides.

E. Closing the Loop

The next step is to combine the various elements in the system. First we close the Flibe loop. From equations (4), (5), (8) and (11), we get

\[ X_{\text{in}}(t) = \exp(-\frac{\lambda\tau_f}{\gamma})[1 - \frac{1}{\gamma}] \exp(-\lambda\tau) X_{\text{out}}(t - \tau), \]

(22)
for the flow of nuclides into the reactor chamber. In this equation, \( \frac{1}{\gamma} \) denotes the identity matrix of order \( N \). Multiplying both sides of this equation by the time \( 1/f \) between two shots yields the change in the nuclide inventory in the reactor-chamber Flibe volume between two shots:

\[ X_{\text{before pulse}}(t) = \exp(-\lambda\tau_f)[1 - K] \exp(-\lambda\tau) X_{\text{after pulse}}(t - \tau). \]

(23)
Similarly we can combine equations (6), (7), (11), (12), (16), and (18) to find the hohlraum high-Z nuclide inventory:

\[ X_{h, \text{before pulse}}(t) = \frac{1}{f} H \]

\[ + \exp(-\lambda\tau_f)[1 - \frac{1}{\gamma}(1,1,\ldots,1)] \exp(-\lambda\tau_f) \]

\[ R K \exp(-\lambda\tau) X_{\text{after pulse}}(t - \tau_f - \tau_f - \tau_f) \]

(24)
Equations (19), (23) and (24) are a complete set of equations to advance the nuclide inventory of the reactor chamber in time. Substituting equations (23) and (24) into equation (19) yields an evolution equation in terms of the in-Flibe high-Z inventory \( X_{\text{after pulse}} \) only. Once the in-Flibe inventory is known as a function of time, the inventories at other locations in the system can be inferred easily from the equations above.

F. Computational Issues

The matrix exponentials appearing in the equations above need to be calculated only once at the beginning of the calculation. Furthermore, some matrix multiplications, such as those appearing in equation (24), can also be made in advance. Therefore, the running time mainly depends on the timestep that is used. In order to calculate the in-Flibe high-Z inventory at time \( t \), the inventory at times \( t - \tau \) and \( t - \tau - \tau \) needs to be known. Consequently the timestep in this calculation is equal to the smaller of the recirculation time \( \tau \) and the storage delay time \( \tau_1 + \tau_2 + \tau_p \). Typically the storage delay time is much larger than the recirculation time.

The minimum number of high-Z nuclide inventories that has to be stored in memory to perform this calculation is

\[
\text{Required storage} = \frac{\tau_1 + \tau_2 + \tau_p}{\tau} \tag{25}
\]

For a recirculation time of 20 s and a storage delay time of about one week, this number is about 30,000. For a calculation extending over the complete reactor lifetime of about 30 years, nearly 50 million matrix multiplications need to be made. The required running time will be unacceptably long unless one considers a small number of nuclides or develops efficient solution algorithms such as those considered in Refs. 6 and 10-12.

III. AN ILLUSTRATION

An approximation that is commonly made is to assume a continuous, rather than pulsed activation. Several studies compared the effect of using the correct pulsing history specification versus a continuous irradiation approximation for different activation scenarios.\(^{10,13,14}\) We shall consider activation in the Flibe only, and assume that no high-Z nuclides are extracted. Then the approximate (continuous activation) model reduces to

\[
X_{\text{after pulse}}(t) = \exp\left(n \sigma \Psi - n \lambda \tau\right) X_{\text{after pulse}}(t - n \tau) \tag{26}
\]

in which \( n \) is the number of passes through the reactor chamber. The flux level is scaled down by the ratio of the Flibe inventory in the reactor chamber to the total Flibe inventory in the primary loop (in order to compensate for the time it takes the high-Z material to circulate with the Flibe outside the reactor chamber). The corresponding PACEM model is based on the equation

\[
X_{\text{after pulse}}(t) = \exp\left(-\lambda \tau\right) X_{\text{after pulse}}(t - \tau) \tag{27}
\]

As an illustration of the accuracy of the continuous activation approximation, we considered the chain consisting of \(^{202}\text{Hg} , \, ^{203}\text{Hg} ,\, ^{204}\text{Hg} ,\, ^{202}\text{Au} ,\, ^{203}\text{Au}\), shown in figure 2. The decay constants and effective group (spectrum weighted) cross sections were taken from ACAB.\(^{8,15,16}\) The in-chamber neutron flux per shot was \( 1.1893 \cdot 10^{13} \) neutrons cm\(^{-2}\); a recirculation time \( \tau \) of 20 seconds was used. After a start-up period, the inventories of the five nuclides considered were in a secular equilibrium. We found that the continuous activation approximation overpredicted the inventory of \(^{202}\text{Au}\) by almost 50\%. The inventories of daughter nuclides of \(^{202}\text{Au}\) are expected to be mispredicted even more pronouncedly.

![Image](image.png)

Figure 2. The \(^{202}\text{Au}\) activation chain considered. Transmutations are denoted by \( \sigma \); decay reactions are denoted by \( \lambda \).
IV. SUMMARY

The PACEM model developed is capable of exact simulation of the variation, with time, of the inventory of target constituents in HYLIFE-II like IFE reactors accounting for (1) the combined effects of activation during target implosion – by a high-amplitude flux, and while passing through the reactor chamber – by a low-amplitude flux; (2) decay during circulation in the primary coolant loop, after extraction from the coolant loop, and before re-insertion into the reactor chamber as a new target; (3) continuous extraction and feed-in of target material; and (4) replacement of part of the activation products by makeup materials. It was found to be too time consuming for simultaneous simulation of many isotopes over 30 years time span. Nevertheless, it is expected to be a useful tool against which different approximations and models can be benchmarked.

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