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GENERATION OF MICROSCOPIC PSEUDO FISSION-PRODUCTS PROPERTIES

H. Benjaafar and G. Marleau
Institut de génie nucléaire,
École Polytechnique de Montréal
C.P. 6079, succ. Centre-ville, Montréal,
Québec, CANADA H3C 3A7
email: benjaa@meca.polymtl.ca
email: marleau@meca.polymtl.ca

ABSTRACT

Most cell burnup calculations are performed using a microscopic cross section library which contains a rather large number of fission-products, the behavior of these fission-products being dictated by an isotopic depletion chain. Here we will describe a method that can be used to simplify this isotopic depletion chain by lumping fission-products into pseudo-isotopes. We will also discuss how to associate decay rates, production yields and time dependent microscopic cross sections with these pseudo-isotopes.

I. INTRODUCTION

The fission process leads to the creation of various isotopes, called fission-products, which may be generated directly by the fission reaction as fragments or produced from these fragments by radioactive decay or other nuclear reactions. The total number of such isotopes is very large (up to a thousand), however, most microscopic cross section libraries used by lattice codes contain only the most important isotopes from the point of view of neutron economy (up to a hundred).[1, 2] The remaining isotopes are already lumped into a few fictitious isotopes, called pseudo fission-products. The nuclear properties of these pseudo-isotopes are selected in such a way that they simulate adequately the effect of the neglected isotopes on cell reactivity for a large variety of lattices.[3, 4]

In the case where the number of depletion zones becomes large, as in full core reactor calculations, even the use of the simplified decay chain available on most microscopic cross section library leads to very large burnup problems which involve thousands of unknowns. This is because the concentration of each isotope in each burnup zone has to be followed independently.[5] Therefore, it can be advantageous in these cases to reduce further the number of fission-products to be considered in such burnup calculations.

One option that could be considered is to combine additional fission-products using a technique similar to that developed for the generation of pseudo-isotope in general microscopic cross section libraries.[6, 7] However, this method is not fully adequate since the contribution of these new fission-products to the macroscopic cross section will often be very large and will depend strongly on the fuel composition.

Here we will describe a different pseudo fission-product generation technique that can be used inside a cell code to simplify automatically the decay chain. The cross sections that will be associated with these pseudo-isotopes will depend on the specific location of a given fuel region in the core and on time. In addition, since some of the pseudo-isotopes we will introduce will appear in the decay chain associated with other fission-products, special care must be taken to derive the decay constant and production yields associated for these fictitious isotopes.

In Section II. will show how to generate decay constants, production yields and time dependent microscopic cross sections, for pseudo-isotopes. In Section III. we will compare the result obtained using a simplified decay chain which includes the new pseudo fission-products with those obtained using a complete depletion chain and discuss the results. Finally, in Section IV. we will conclude.

II. THEORICAL INVESTIGATION

The standard technique used to generate pseudo fission-products for microscopic cross section libraries is based on the assumption that the cross sections associated with pseudo-isotopes are independent of time.[6, 7] As a result, the only way to simulate adequately the concentration of these fictitious isotopes as a function of time is via their decay constant and production yield. In most cross section libraries, one introduces two different pseudo fission-products. The first one has a relatively small decay constant and saturates slowly while the second isotope is stable. The specific decay constant, fission yield and cross sections associated with each pseudo-isotope are then determined using numerical experimentation on a large number of cells typical of those which will be analyzed using this microscopic cross section library.

The method we propose here is different in many respect. First, it will be implemented directly within a lattice code to modify the decay chain that will be used in a subsequent lattice or reactor calculation. As a consequence the properties of the pseudo-isotopes generated can be associated directly with a specific fuel cell. Moreover, instead of considering constant cell averaged microscopic cross section for each pseudo-isotope, we will generate regional time dependent microscopic cross sections. On the other hand constant cell averaged production yields and decay rates will be generated. Finally, the pseudo-isotopes classification will be based on the decay chain to which they belong rather than on their time dependent behavior.

II.1 Physical Properties of the Pseudo Fission-Products

Let us assume that a pseudo-isotope P is composed of n_P fission-products of concentration $N_i(r,t)$ and microscopic absorption cross section $\sigma_i^a(E)$. The concentration $N_P(r,t)$ of this pseudo-isotope will be defined as

$$N_P(t,r) = \sum_{i=1}^{n_P} N_i(r,t) \tag{1}$$

where the concentration of the individual fission-products is obtained by solving the depletion equation:

$$\frac{dN_i(r,t)}{dt} = \sum_{j=1}^{n_P} N_j(r,t)\left(Y_{ij}^d\lambda_j + Y_{ij}^a r_j^a(r,t)\right) + \sum_{k=n_P+1}^{n_R} N_k(r,t)\left(Y_{ik}^d\lambda_k + Y_{ik}^a r_k^a(r,t)\right) + \sum_{l=n_R+1}^{n_F} N_l(r,t) Y_{il}^f r_l^f(r,t) \qquad (2)$$

where $r_j^x(r,t)$, the microscopic reaction rate for a reaction of type $x = a$ (absorption) or $x = f$ (fission), is defined as:

$$r_j^x(r,t) = \int \sigma_j^x(E)\phi(r,t,E)dE$$

Here $j = 1, n_P$ represents an isotope that will be included in a pseudo fission-product P, $k = n_P + 1, n_R$ identifies additional fission-products not be included in P and $l = n_R + 1, n_F$ represents a fissile isotope. The terms Y_{mn}^a, Y_{mn}^d and Y_{mn}^f for $m \neq n$ are the production yields for isotope m resulting from the absorption, the decay and the fission of isotope n, respectively. In addition, $Y_{mm}^d = Y_{mm}^a = -1$ accounts for the fact that isotope m can generally deplete via radioactive decay and by neutron absorption.

We will associate to the pseudo-isotope a microscopic absorption cross section σ_P^a which is defined as:

$$\sigma_P^a(r,t,E) = \frac{1}{N_P(t,r)} \sum_{i=1}^{n_P} N_i(r,t)\sigma_i^a(E) \qquad (3)$$

This definition ensures that the contribution of the pseudo-isotope to the macroscopic fuel absorption cross section required in the neutron flux equation (diffusion or transport equation) is equivalent to the total contribution of the individual fission-products included in the pseudo-isotope.

Now suppose that instead of evaluating the concentration of the pseudo fission-products using in Eq. (1) we would like to use a burnup equation similar to Eq. (2), namely

$$\frac{dN_P(r,t)}{dt} = N_P(r,t)\left(Y_{PP}^d(t)\lambda_P(t) + Y_{PP}^a(t) r_P^a(r,t)\right) + \sum_{k=n_P+1}^{n_R} N_k(r,t)\left(Y_{Pk}^d\lambda_k + Y_{Pk}^a r_k^a(r,t)\right) + \sum_{l=n_R+1}^{n_F} N_l(r,t) Y_{Pl}^f r_l^f(r,t) \qquad (4)$$

where $r_P^a(r,t)$ is given by:

$$r_P^a(r,t) = \int \sigma_P^a(r,t,E)\phi(r,t,E)dE$$

We will also need to modify the depletion equation for the remaining fission-products to the form

$$\frac{dN_i(r,t)}{dt} = N_P(r,t)\left(Y_{iP}^d(t)\lambda_P(t) + Y_{iP}^a(t) r_P^a(r,t)\right) + \sum_{k=n_P+1}^{n_R} N_k(r,t)\left(Y_{ik}^d\lambda_k + Y_{ik}^a r_k^a(r,t)\right) + \sum_{l=n_R+1}^{n_F} N_l(r,t) Y_{il}^f r_l^f(r,t) \qquad (5)$$

to account for the fact that these isotopes can be produced by P. In order for Eqs. (4) and (5) to be entirely coherent with Eq. (2) we will need:

$$Y^f_{Pl} = \sum_{i=1}^{n_P} Y^f_{il} \qquad Y^d_{Pk} = \sum_{i=1}^{n_P} Y^f_{ik} \qquad Y^a_{Pk} = \sum_{i=1}^{n_P} Y^f_{ik}$$

for the production yields of P from other isotopes and

$$Y^d_{PP}(t)\lambda_P(t) = \frac{1}{N_P(r,t)} \sum_{j=1}^{n_P} N_j(r,t)\lambda_j \sum_{i=1}^{n_P} Y^d_{ij} \tag{6}$$

$$Y^d_{iP}(t)\lambda_P(t) = \frac{1}{N_P(r,t)} \sum_{j=1}^{n_P} N_j(r,t)\lambda_j Y^d_{ij} \tag{7}$$

$$Y^a_{PP}(t)r^a_P(r,t) = \frac{1}{N_P(r,t)} \sum_{j=1}^{n_P} N_j(r,t)r^a_j(r,t) \sum_{i=1}^{n_P} Y^a_{ij} \tag{8}$$

$$Y^a_{iP}(t)r^a_P(r,t) = \frac{1}{N_P(r,t)} \sum_{j=1}^{n_P} N_j(r,t)r^a_j(r,t)\; Y^a_{ij} \tag{9}$$

The problem now is to select the parameters $Y^d_{PP}(t)$, $Y^a_{PP}(t)$, $Y^d_{iP}(t)$, $Y^a_{iP}(t)$ and $\lambda_P(t)$ in such a way that $\sigma^a_P(r,t,E)$ and $N_P(r,t)$ remain compatible with Eqs. (1) and (3) respectively.

We will first assume that the depletion chain associated with the pseudo-isotope is closed. This situation corresponds to the case where the isotopes included in P can only deplete to other isotopes in P. As a result, the pseudo-isotope P does not depletes (it is considered stable) and we will have $Y^d_{iP}(t) = Y^a_{iP}(t) = 0$. In addition, we will have:

$$\sum_{i=1}^{n_P} Y^d_{ij} = \sum_{i=1}^{n_P} Y^a_{ij} = 0$$

since the transmutation of an isotope in P yields a new isotope which is also included in P. Accordingly, we can associate the following decay constant and production yields to the pseudo-isotope:

$$\lambda_P(t) = 0 \qquad Y^d_{PP}(t) = 0 \qquad Y^a_{PP}(t) = 0$$

These properties are now independent of space and time even if the microscopic cross sections associated with the pseudo-isotope remain both space and time dependent (see Eq. (3)).

The second situation we will consider is that where one assumes that some of the isotopes in P can deplete via neutron absorption to isotopes which are not included in P. In the case where isotope S of P depletes to isotope K not in P with a yield $Y^a_{KS} = 1$, Eqs. (6) to (9) can be simplified to:

$$Y^d_{KP}(t) = Y^d_{PP}(t) = 0 \tag{10}$$

$$Y^a_{KP}(t) = -Y^a_{PP}(t) = \frac{N_S(r,t)r^a_S(r,t)}{N_P(r,t)r^a_P(r,t)} \tag{11}$$

Namely isotope P will disappear as a rate proportional to $N_S(r,t)$ while producing isotope K at the same rate. Here $Y^a_{PP}(t)$ and $Y^a_{KP}(t)$ are still functions of time. Two different techniques can be used to remove the time dependence from these yields. First we may assume that the physical properties of the n_P isotopes included in P all have a similar spatial and time dependent behavior. In this case we can write:

$$N_P(r,t)r^a_P(r,t) = \sum_{i=1}^{n_P} N_i(r,t)r^a_i(r,t) \approx n_P N_S(r,t)r^a_S(r,t)$$

and we will have

$$Y^a_{KP} = -Y^a_{PP} = \frac{1}{n_P} \tag{12}$$

We could also have considered a time averaging process for $Y^a_{KP}(t)$, namely:

$$Y^a_{KP} = -Y^a_{PP} = \frac{1}{T}\int_T \left(\frac{N_S(r,t)r^a_S(r,t)}{N_P(r,t)r^a_P(r,t)}\right) dt \tag{13}$$

where T is the total burnup time.

One final comment concerns the time dependence of the microscopic cross sections associated with the pseudo fission-product defined in Eq. (3). This relation will be approximated using:

$$\sigma^a_P(r,t,E) \approx a_P(r,E) + b_P(r,E)t \tag{14}$$

This approximation can be justified by the fact that most pseudo fission-products will be dominated by a single isotope. Then, assuming that the cross sections and the concentration of all the other isotopes in P are small compare to the dominant isotope, P can be considered as being made up of a single isotope and $\sigma^a_P(r,t,E)$ becomes independent of t and r. Similarly, in the case where the concentration of all the isotopes included in P have a similar time dependent form, $\sigma^a_P(r,t,E)$ also becomes independent of t and r. By maintaining the explicit dependence of $\sigma^a_P(r,t,E)$ on r we hope that the variations of the cross section as a function of time due to differences in the neutron flux from region to region will be kept to a minimum. From a numerical viewpoint the coefficients $a_P(r,E)$ and $b_P(r,E)$ in Eq. (14) will be obtained using a least square fit method. We will return to this point in Section III. when we compare the explicit dependence of the cross section on time with the results obtained using Eq. (14).

II.2 Nature of the Pseudo Fission-Products

In our analysis, the various fission-products were first divided into three different classes. The first class contains the main isotopes in a specific library which belong to the $^{135}\mathrm{Xe}$ depletion chain. These include both $^{135}\mathrm{I}$ which decays to $^{135}\mathrm{Xe}$ and $^{135}\mathrm{Cs}$ which is produced from the decay of $^{135}\mathrm{Xe}$. The second series of isotopes we will consider are those belonging to the $^{149}\mathrm{Sm}$ depletion chain. Finally we will assume that all the fission-products not included in these two chains belong to the third class.

Let us first consider the fission-products which are part of the $^{135}\mathrm{Xe}$ decay chain. There are several observations which have dictated our treatment of these isotopes. First the neutron absorption cross section for $^{135}\mathrm{Xe}$ is large compared to all the other fission-products absorption cross section. This means that even if its concentration in the fuel at any time is relatively low, its contribution to the neutron balance in a cell is very important. In fact, small errors in the concentration of $^{135}\mathrm{Xe}$ lead to large errors in the cell reactivity. A second consequence of this large absorption cross section is that $^{135}\mathrm{Xe}$ reaches an equilibrium concentration rapidly, its concentration being directly proportional to the flux level in the fuel. As a result only slight variation in the burnup power level will affect directly the $^{135}\mathrm{Xe}$ concentration. Another observation is that $^{135}\mathrm{Xe}$ is both a direct fission-product and a decay product from $^{135}\mathrm{I}$. Accordingly, in order to ensure that the contribution of $^{135}\mathrm{Xe}$ to the cell reactivity is correctly represented its production from $^{135}\mathrm{I}$ must be correctly simulated. Finally, because $^{135}\mathrm{Xe}$ decays naturally into $^{135}\mathrm{Cs}$ with a half-life of around 15 minutes, it disappears relatively rapidly after a power shutdown.

These observations suggested that $^{135}\mathrm{Xe}$ is not a good candidate to include in a pseudo fission-product, since its contribution to the fuel macroscopic absorption cross section would then be diluted by the other isotopes in the chain (see Eq. (3)). Moreover because of the assumed time dependence of the pseudo-isotope cross sections, decay constant and yield, all the short term features resulting from the presence of $^{135}\mathrm{Xe}$ would be lost. Accordingly we decided to treat explicitly each of the isotopes belonging to this decay chain.

The second chain we considered is that of $^{149}\mathrm{Sm}$ which also has a relatively large absorption cross section. There are two main differences between the $^{149}\mathrm{Sm}$ and the $^{135}\mathrm{Xe}$ chain, namely, it saturates much more slowly and its concentration rapidly reaches a maximum after a power shutdown since it depletes only via neutron absorption. Because the knowledge of the $^{149}\mathrm{Sm}$ concentration in the fuel is very important due to its large cross section, we decided to treat this isotope explicitly. The remaining isotopes were lumped into two different pseudo-isotopes, the first one P_c corresponding to the set of isotopes leading to $^{149}\mathrm{Sm}$ and the second one P_d including all the isotopes produced directly or indirectly from the depletion of $^{149}\mathrm{Sm}$. For both isotopes we will approximate the absorption cross section using Eq. (14).

We will first assume that the fission yield for pseudo-isotope P_c is equal to the sum of the fission yields of its constituents. It will also deplete via neutron absorption with a yield $Y^a_{PP}(t) = -1$. This second assumption is somewhat inconsistent with the exact decay chain associated with the isotopes included in the pseudo fission-product since some of these are transformed into isotopes also included in P_c by neutron absorption. Accordingly, the concentration of P_c should be under-estimated using this method. On the other hand, the production yield of $^{149}\mathrm{Sm}$ from P_c, will take into account the fact that only some (n_d) of the (n_c) isotopes included in P_c can be transformed directly into $^{149}\mathrm{Sm}$ via neutron absorption. We will then use an equation similar to Eq. (12) to compute this production yield:

$$Y^a_{\mathrm{Sm}P}(t) = \frac{n_\mathrm{d}}{n_c} \tag{15}$$

Although these approximations look very crude, they perform relatively well as we will show in Section III. The properties of the second pseudo fission-product P_d will be obtained in much the same way as described above for P_c, the main difference being that P_d has, in addition, a production yield from $^{149}\mathrm{Sm}$ by neutron absorption equal to unity.

Finally the remaining fission-products which are not included in the $^{135}\mathrm{Xe}$ or $^{149}\mathrm{Sm}$ depletion chains will be combined to form a final pseudo fission-product P_r. Here we will assume that this pseudo-isotope is formed only as a result of fission and that it depletes by neutron absorption. Its cross sections are also approximated using Eq. (14).

III. RESULTS AND DISCUSSION

The technique described above will be applied to the study of a CANDU cell using the transport code DRAGON.[8, 9] The 11 groups microscopic cross section library we have used is a condensed version of the WIMS-AECL Winfrith library.[2] Two types of calculations were performed. For the reference calculation we used the full library depletion chain in the burnup calculations. The burnup power used in our simulations is 615 MW during a period of three hundred thirty days using either 12 unequal time intervals of 1, 4, 5, 10, 10, 20, 20, 40, 50, 50, 50 and 70 days respectively or 24 shorter intervals (each of the original time interval is subdivided into 2 equal sub-intervals). The same calculations were then repeated using a modified library which involves the simplified depletion chain containing the three new pseudo-isotopes to which are associated time dependent cross sections. Before going into a description of the results obtained using the modified decay chain let us first present how the linear approximation for the time dependence of the pseudo-isotopes absorption cross section compares to a reference calculation.

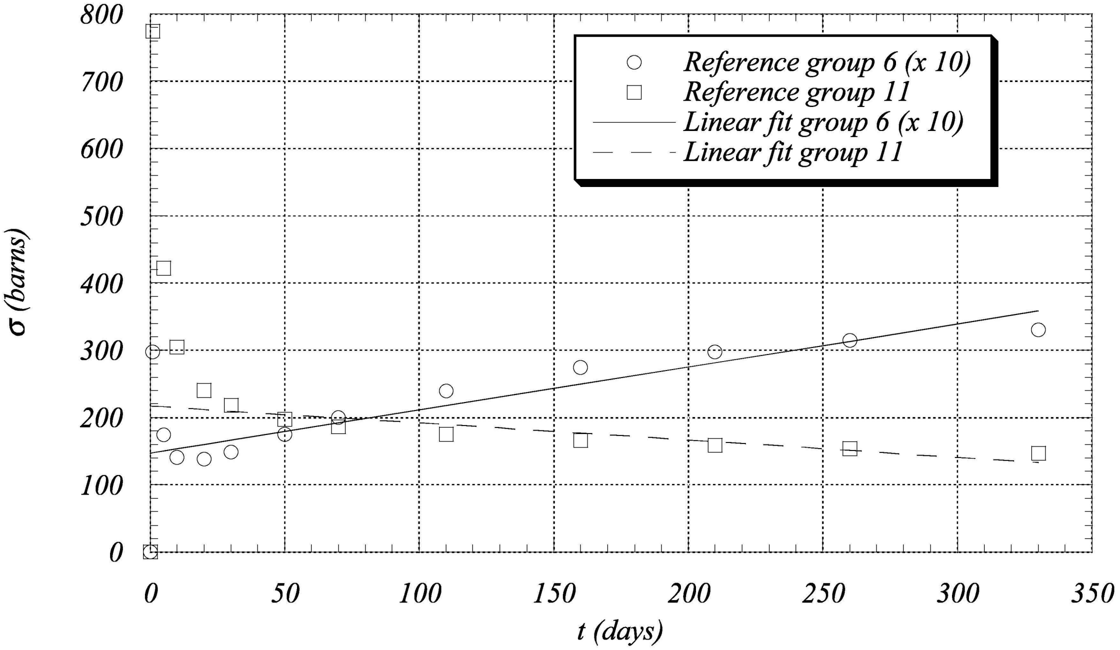

Figure 1. Microscopic absorption cross sections for P_r in group 6 ($0.78 \leq E \leq 4.0$ eV) and 11 ($E \leq 0.005$ eV) for the central fuel element in a CANDU cell.

As we noted before, one can use the isotopic concentration generated by the reference calculation for each isotope to compute the exact time dependent microscopic cross sections associated with a pseudo-isotope. These were then transferred to a least square fit routine which generated the two coefficients defined in Eq. (14). One can find in Figure 1 a comparison between the exact and the approximate time dependence of the microscopic absorption cross section associated with pseudo-isotope P_r for two different energy groups. Here we should note that because the initial concentration of P_r vanishes, the reference microscopic cross section associated with the pseudo-isotope is defined arbitrarily as 0 at $t = 0$. In addition, since the pseudo-isotopes generally contain rapidly saturating isotopes, we did not use the first few time steps ($t < 20$ days) in our least square fit method, therefore ensuring that only the long term behavior of the pseudo-isotope cross sections would be represented. Such large errors in the cross sections as those observed in Figure 1 for $t < 20$ days should not affect substantially the neutron balance in the cell since the concentration of P_r during this burnup period remains relatively low. The same observation is valid for the other groups and pseudo-isotopes we generated using our model.

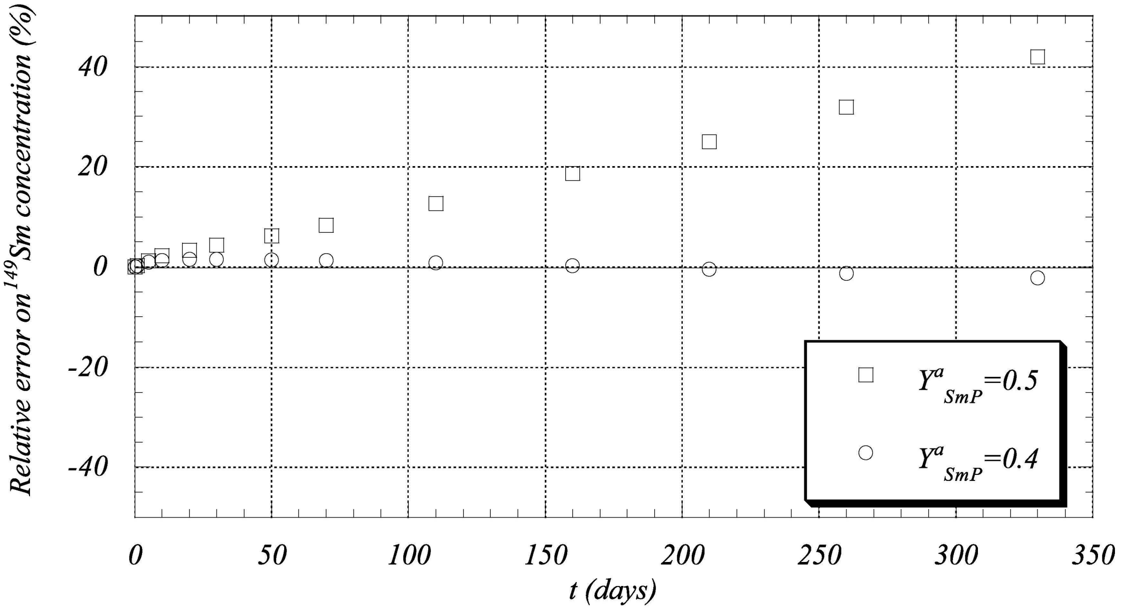

Figure 2. Relative error in the $^{149}\mathrm{Sm}$ concentration as a function of time for two different values of $Y^a_{\mathrm{Sm}P}$

As we mentioned above, our new decay chain now involves three different pseudo-isotopes, two of which (P_c and P_d) are associated with the $^{149}\mathrm{Sm}$ decay chain. Since pseudo-isotope P_c is made up of 5 different isotopes two of which yield $^{149}\mathrm{Sm}$ by neutron capture, we will select $Y^a_{\mathrm{Sm}P} = 0.4$ (see Eq. (15)). In order to illustrate how this value affects the concentration of $^{149}\mathrm{Sm}$, we have repeated the burnup calculations using different values for $Y^a_{\mathrm{Sm}P}$. Note that the neutron balance will be mainly affected by the concentration of $^{149}\mathrm{Sm}$ (with its large cross section), so even small errors in its concentration could result in large changes in the reactivity of the cell. The results we obtained are presented in Figure 2 . The first point to note is that the specific value of $Y^a_{\mathrm{Sm}P}$ affects considerably the concentration of $^{149}\mathrm{Sm}$. The second point is that our crude approximation for $Y^a_{\mathrm{Sm}P}$ seems to be adequate for the problem we have considered here even if it starts by slightly over estimating the $^{149}\mathrm{Sm}$ concentration and ends with an under prediction of about 2 % . This behavior is not surprising since there are three different time constants (one from fission and two from production by depletion of fission products) associated with the production of $^{149}\mathrm{Sm}$ in the reference depletion chain while for the simplified decay chain only two time constants are considered.

One can find in Figure 3 a comparison between the k_{eff} results obtained using the reference and the simplified decay chain for the CANDU-6 cell. As one can see the general form of both

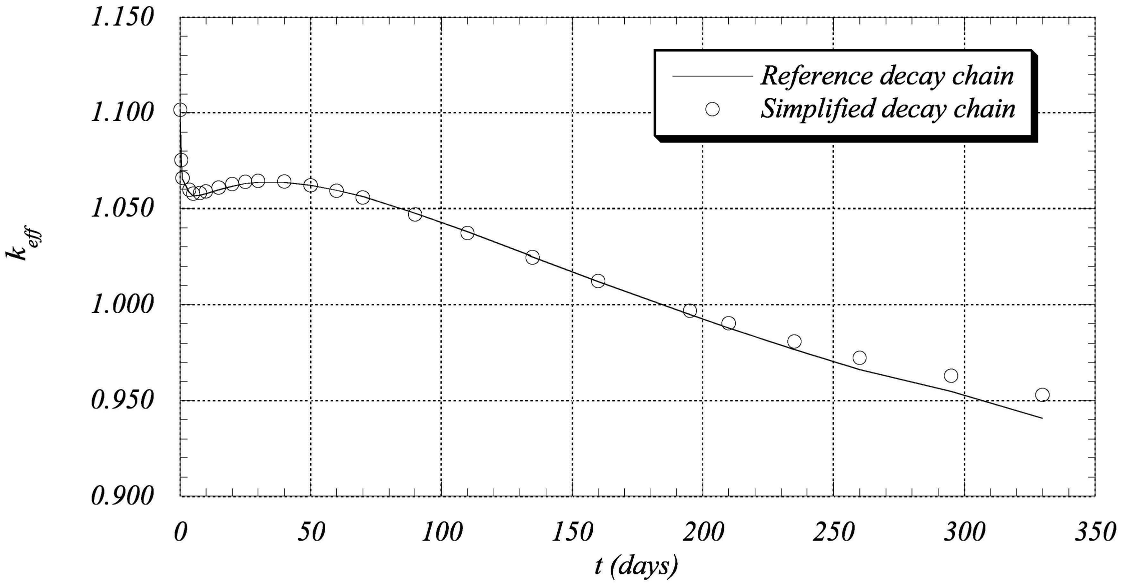

Figure 3. Cell reactivity k_{eff} as a function of time using the reference and the simplified depletion chains

Table 1. Difference in k_{eff} as a function of time

t(days)	$k_{eff}^{reference}$	12 steps Δk_{eff}(mk)	24 steps Δk_{eff}(mk)
0.0	1.1017	0.0	0.0
1.0	1.0655	0.6	0.6
5.0	1.0565	1.2	1.2
10.0	1.0576	1.2	1.2
20.0	1.0617	1.0	1.0
30.0	1.0637	0.5	0.7
50.0	1.0622	-0.2	0.0
70.0	1.0561	-0.8	-0.4
110.0	1.0378	-1.3	-0.6
160.0	1.0120	-0.2	0.5
210.0	0.9877	2.2	2.8
260.0	0.9661	5.9	6.3
330.0	0.9407	12.9	12.9

curve is similar except for $t > 200$ days where the simplified decay chain results in an over estimation of k_{eff}. One can also find in Table 1 the results we obtained for k_{eff} as a function of time for the reference calculation ($k_{\mathit{eff}}^{\mathit{reference}}$) as well as the reactivity difference Δk_{eff}

$$\Delta k_{\mathit{eff}} = 1000 \times \left(k_{\mathit{eff}}^{\mathit{simplified}} - k_{\mathit{eff}}^{\mathit{reference}} \right)$$

resulting from the use of the simplified decay chain involving our three pseudo fission-products ($k_{\mathit{eff}}^{\mathit{simplified}}$). Note that two cases are presented here. In the third column, the differences in reactivity are those obtained when the pseudo-isotope cross sections were computed based on a burnup calculation involving only the 12 time intervals presented in the table while for the forth column, the burnup calculation were performed using 24 shorter time intervals (see above). The reference values for k_{eff} were also obtained using these shorter time intervals.

As one can observe, the size of the burnup intervals used for the generation of the pseudo-isotopes properties, while having some effect on the precision of the method is not a major consideration here. On the other hand, we see that even if the technique performs relatively well for the first two hundred days of burnup, it degrades substantially at the end of the cycle. This proves that the assumption that we can neglect the first few times steps in the process of generating the time dependent cross sections is valid. The main problem here might be the slight over evaluation of the short-term concentration of $^{149}\mathrm{Sm}$ and the effect of the larger under estimation (see Figure 2) on the long-term behavior of the reactivity. Another source for the long-term over prediction of the reactivity is the choice of a uniform depletion yield of $Y_{KP}^{a} = -1$ for the pseudo isotopes since this choice should also result in an under-estimation of the pseudo-isotope concentrations.

IV. CONCLUSIONS

Using our pseudo fission-product generation technique, we have been able to simplify the depletion chain on the WIMS-AECL Winfrith library in such a way that instead of the more than 30 fission-products initially needed, only 9 fission-products are now required. This is to the expanse of generating spatial and time dependent cross sections for the 3 pseudo-isotopes generated. The main difference in reactivity observed between the case where the reference and the simplified depletion chain were used seems to be due to the differences in the $^{149}\mathrm{Sm}$ concentration between both method and the under estimation of the pseudo-isotope concentrations. The error introduced by the use of a simple linear time dependence for the cross section associated with the pseudo fission-products is relatively small as we observed.

Additional work is still required to simplify further the depletion chain. This would primarily consist in a simplification of the $^{135}\mathrm{Xe}$ depletion chain to a form similar to that used for $^{149}\mathrm{Sm}$ with the introduction of two new pseudo-isotopes, one leading to the creation of $^{135}\mathrm{Xe}$ and the other one being produced by its decay. Finally, the problem of associating with the pseudo-isotopes more realistic depletion yields will also need to be addressed.

ACKNOWLEDGMENTS

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