PSEUDO-ISOTOPES GENERATION FOR THE ^{235}U AND ^{238}U DEPLETION CHAINS

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ABSTRACT

We will discuss in details the method used to generate the properties of the pseudo-isotopes for the ^{235}U and ^{238}U depletion chains. Using physical considerations, we derive a simple relation for the time and space dependence of the cross section, decay constant and fission yields for the pseudo-isotopes. The effect of the creation of these pseudo-isotopes and of the elimination of ^{243}Am, ^{242}Am, ^{241}Am and ^{237}Np on the critically of a thermal power reactor are also investigated.

I. INTRODUCTION

The operation of a nuclear power reactor necessarily implies the consumption or burnup of reactor fuel by fission and capture, which give rise to a decrease in reactivity of the reactor. A complete treatment of burnup in a core calculation requires the knowledge of the microscopic cross sections and the burnup properties associated with all the nuclide which may be present in the fuel. In principle, such an analysis requires the solution of transport or diffusion and the nuclide field equations (depletion). However, this is not possible in practice because of the excessive time that would be required to perform such a calculation. In fact the isotopic composition of the fuel depends on the microscopic flux in every fuel element of a cell, and the number of such cells in the reactor is very large. In addition, the number of isotope that must be considered is also very large. As a result, it is nearly impossible to follow explicitly the concentration of every isotope in every fuel element independently in practical reactor calculation.

Many approximations could be considered in order to simplify the problem.[1, 2, 3] Here we will discuss one technique that can be used to reduce the number of heavy nuclide that must be treated to follow the core burnup. We will combine the heavy isotopes into a few pseudo-isotopes. The reason for choosing this approximation relies on the fact that all the heavy isotopes have a significant effect on the nuclear characteristics of the reactor. Their destruction by radioactive decay or neutron capture leads to the creation of new heavy isotopes. In addition the fissile heavy isotopes are responsible for the energy

production in the reactor and the generation of fission products not originally present in the fuel. The scenario that we have considered for the generation of the pseudo-isotopes consists in treating separately the ^{235}U and ^{238}U chains, and for each chain to generate one pseudo-isotopes. In this paper, we will discuss in details the method used to generate the properties of such pseudo-isotopes, and also present the results obtained by applying this method to ^{235}U and ^{238}U chain.

In order to reduce the number of isotopes to be treated in any core calculation, the effect of the elimination of transuranium isotope on K_{eff} for the PWR thermal reactor was studied.[7] Here we will also investigate the effect of the elimination of ^{237}Np and ^{243}Am, ^{242}Am and ^{241}Am on the multiplication factor (K_{eff}). Actually this effect cannot be neglected for a CANDU reactor, for instance, after the elimination of ^{237}Np, the error in K_{eff} is about 0.395 mk after 300 days of operation.

II. PROBLEM FORMULATION

For an isotope i having a concentration $N_i(r,t)$ nuclei per unit volume at time t and an absorption cross section $\sigma_{i,a}$, the production rate from the isotope j will be proportional to $\sigma^g_{j\to i}$ and to $\lambda_{j\to i}N_j(r,t)$, where

$$\sigma^g_{j\to i} = \gamma_{j\to i}\sigma^g_{j,f} \tag{1}$$

for a production due to fission, and

$$\sigma^g_{j\to i} = \gamma_{j\to i}\sigma^g_{j,x} \tag{2}$$

for a production due to a nuclear reaction x ((n,2n), (n,γ), ...). The destruction rate by radioactive decay is proportional to $\lambda_i N_i(r,t)$, hence the burnup equation for isotope i in multigroup theory is given by:

$$\frac{dN_i(r,t)}{dt} + \left(\sum_{g=1}^{G}\sigma_{i,a}\phi^g(r,t) + \lambda_i\right)N_i(r,t) = \sum_{\substack{j=1\\ j\neq i}}^{I}\left(\sum_{g=1}^{G}\sigma^g_{j\to i}\phi^g(r,t) + \lambda_{j\to i}\right)N_j(r,t) \tag{3}$$

where $\phi^g(r,t)$ is the neutron flux in the group g. Equation 3 can also be written as follows:

$$\frac{dN_i(r,t)}{dt} + \left(\sum_{g=1}^{G}\sigma_{i,a}\phi^g(r,t) + \lambda_i\right)N_i(r,t) = P_i(r,t) \tag{4}$$

where $P_i(r,t)$ is the production rate of the isotope i.

Let's assume that the isotope m and n are combined to generate the pseudo-isotope q. The burnup equation for q can be written:

$$\frac{dN_q(r,t)}{dt} + \left(\sum_{g=1}^{G}\sigma_{q,a}(r,t)\phi^g(r,t) + \lambda_q(r,t)\right)N_q(r,t) = P_q(r,t) \tag{5}$$

where

$$N_q(r,t) = N_m(r,t) + N_n(r,t) \tag{6}$$

$$P_q(r,t) = \frac{P_m(r,t) + P_n(r,t)}{N_q(r,t)} \tag{7}$$

$$\lambda_q(r,t) = \frac{\lambda_m N_m(r,t) + \lambda_n N_n(r,t)}{N_q(r,t)} \tag{8}$$

and finally

$$\sigma^g_{q,a}(r,t) = \frac{\sigma^g_{m,a} N_m(r,t) + \sigma^g_{n,a} N_n(r,t)}{N_q(r,t)} \tag{9}$$

Equation 5 is a nonlinear equation where both the absorption cross section and the radioactive decay constant of the pseudo-isotope depends explicitly on time and space. Hence, solving this equation becomes very difficult. In order to simplify the problem many approximations have to be considered.

II.1 Approximation

Let's assume that the concentration of the isotope m is larger than the concentration of the isotope n over the period of operation of a reactor (300 days in our case), and consider that the concentration of our pseudo-isotope q is equal to the concentration of the isotope m. The equations 6 and 9 can be written as:

$$N_q(r,t) = N_m(r,t) \tag{10}$$

and

$$\tilde{\sigma}^g_{q,a}(r,t) = \sigma^g_{m,a} + \sigma^g_{n,a} \frac{N_n(r,t)}{N_m(r,t)} \tag{11}$$

From the equation above, we observe that the absorption cross section of the pseudo-isotope q is equal to the absorption cross section of the isotope m plus a correction term that takes into account the absorption cross section of the isotope n and the ratio of the concentration of the isotope n and m. Since this ratio is always inferior to 1, we may expand it in time to order N. For our calculation we choose a quadratic expansion (N=2). If we define this ratio by $P(r,t)$ then we may write:

$$P(r,t) = P(r,0) + \frac{dP(r,t)}{dt}|_{t=0} t + \frac{1}{2}\frac{d^2P(r,t)}{dt^2}|_{t=0} t^2 + \theta(r,t) \tag{12}$$

Hence, the absorption cross section for the pseudo-isotope can be approximated by the following expression:

$$\tilde{\sigma}^g_{q,a}(r,t) = \sigma^g_{m,a} + \sigma^g_{n,a} P(r,0) + \sigma^g_{n,a} \frac{dP(r,t)}{dt}|_{t=0} \times t + \sigma^g_{n,a} \frac{1}{2}\frac{d^2P(r,t)}{dt^2}|_{t=0} \times t^2 \tag{13}$$

which means that, for a given region, the time dependence of the cross section of the pseudo-isotope can be written as follows:

$$\tilde{\sigma}^g_{q,a}(r,t) = \sum_{n=0}^{2} a_n(r) t^n \tag{14}$$

In order to simplify our depletion chain, we will associate with the pseudo-isotope a time and space independent decay constant. We will therefore replace $\lambda_q(r,t)$ in equation 5 by its time and space average $\overline{\lambda}_q$ defined as:

$$\overline{\lambda}_q = \frac{1}{V}\frac{1}{T}\int_V dr \int_0^T \lambda_q(r,t)dt \tag{15}$$

The fission yield of the pseudo-isotope q must also be calculated properly so as to conserve the production rate of the fission products. In fact, if the isotope m and n are fissile and their fission yield are noted by Y_m and Y_n then the fission yield of the pseudo-isotope is given by:

$$Y_q(r,t) = Y_m \frac{\sum_g \phi^g \sigma^g_{m,f}}{\sum_g \phi^g \tilde{\sigma}^g_{q,f}(r,t)} + Y_n \frac{\sum_g \phi^g \sigma^g_{n,f}}{\sum_g \phi^g \tilde{\sigma}^g_{q,f}(r,t)} \frac{N_n(r,t)}{N_m(r,t)} \tag{16}$$

We observe that the fission yield of the pseudo-isotope depends explicitly on time t and on space r since the fission cross section of the pseudo-isotope q and the concentration of the isotope m and n depend on t and r. So as to simplify our depletion chain, we will associate with the pseudo-isotope a time and space independent fission yield defined as:

$$\overline{Y}_q = \frac{1}{T}\frac{1}{V}\int_V dr \int_0^T Y_q(r,t)dt \tag{17}$$

where T is the period of operation of the reactor.

II.2 Case of the combination of many isotopes

Assume that N isotopes are combined to generate the pseudo-isotope q. Two main cases can be discussed.

- The concentration of one of the N isotopes remain larger than the concentration of the other isotopes during the period of operation of the reactor, T.

  If one combines N isotopes to generate the pseudo-isotope q and assumes that isotope m has the largest concentration, equations 10 and 11 can be written as:

  $$N_q(r,t) = N_m(r,t) \tag{18}$$

  and

  $$\tilde{\sigma}^g_{q,a} = \sigma^g_{m,a} + \sum_{i=1, i\neq m}^{N} \sigma^q_{i,a} \frac{N_i(r,t)}{N_m(r,t)} \tag{19}$$

  In this case, we consider that the concentration of the pseudo-isotope q is equal to the concentration of the isotope m and the absorption cross section of the pseudo-isotope is equal to the absorption cross section of the isotope m plus a correction term that take into account the absorption cross section of the other isotope and the ratio of the concentration of the other isotopes and the isotope m.

- The concentration of one of these N isotopes is larger only in the interval of time $[t_1^m, t_2^m]$ where m refers to the isotope name, and t_2^m is inferior to T.

  In this case, the period of operation of the reactor T is divided into time intervals such that in each interval the concentration of one isotope is larger than the concentration of the remaining isotopes. In order to clarify this case, let's consider the case where we have only 2 time intervals such that the concentration of isotope m dominates in the interval $[0,t_1]$ whereas isotope n dominates in the interval $[t_1,T]$. For the first interval of time, the concentration and the absorption cross section of the pseudo-isotope q are given by the following equation:

$$N_q(r,t) = N_m(r,t) \tag{20}$$

$$\tilde{\sigma}_{q,a}^g = \sigma_{m,a}^g + \sum_{i=1,i\neq m}^{N} \sigma_{i,a}^q \frac{N_i(r,t)}{N_m(r,t)} \tag{21}$$

  The above equations are considered valid as soon as the ratio of the concentration of the isotope m and the rest of the other isotopes remain inferior to 1. However, in the second interval, since the concentration of the isotope n is larger than the concentration of the isotope m, equations 20 and 21 become:

$$N_q(r,t) = N_n(r,t) \tag{22}$$

$$\tilde{\sigma}_{q,a}^g = \sigma_{n,a}^g + \sum_{i=1,i\neq n}^{N} \sigma_{i,a}^q \frac{N_i(r,t)}{N_n(r,t)} \tag{23}$$

  This case can be applied, for instance, to the combination of ^{243}Am, ^{242}Am and ^{241}Am as the concentration of ^{243}Am becomes larger than the concentration ^{241}Am after approximately 200 days of operation of CANDU reactors.

II.3 Method of calculation

The numerical work required for determining the properties of the pseudo-isotope can be divided into three parts: (1) reference calculation, (2) determination of the coefficients $a_n(r)$ given by equation 14 to determine the cross section of the pseudo-isotope, (3) finally, determination of the constant decay and the fission yield of the pseudo-isotope.

A reference calculation, performed by DRAGON code, where all the isotopes are included, is required to determine the concentration of heavy isotopes to be combined and the reference effective multiplication, K_{eff}, for the different time steps.[4, 5] For a given region, the values of the cross section of the pseudo-isotopes, for the different time step, were fitted to the form given by equation 14 to determine the coefficients $a_n(r)$ using the least-squares method. Finally, by using the fission cross section of the pseudo-isotope, the concentrations and the cross sections of the isotopes that we combine, we determine the average value of the fission yield and of the decay constant using equation 15 and 17. These tasks are required for the generation of a new library, that contains all the nuclear properties of the pseudo-isotope, (initial concentration, microscopic cross section, constant decay, ...), and of course the properties of the other isotope that weren't combined. The results obtained using our new libraries will be compared with the reference calculation.

III. RESULTS AND DISCUSSION

For our calculation, we have considered that the time dependence of the cross section of the pseudo-isotope is linear in time. The K_{eff} and the concentration of the pseudo-isotope are calculated for 300 days with 10 time steps. The cross section for a 69-group of energy are calculated by the condensation method applied to the WIMSLIB library. The 69-group energy cross section are averaged over the whole spectrum to give the 2-group cross section. All these calculations are performed in transport theory using DRAGON.

III.1 Elimination of heavy isotopes

In order to reduce the number of isotopes to be treated we have investigated three cases: (1) elimination of ^{237}Np, and (2) elimination of ^{243}Am, ^{242}Am and ^{241}Am, (3) elimination of the four isotopes together. The K_{eff} is calculated for 300 days with 10 time steps. The results are presented in table 1.

- ^{235}U chain

  Table 1 shows that the elimination of the ^{237}Np from ^{235}U chain has a significant effect on K_{eff} since its concentration in CANDU reactor can't be neglected. In addition, the absorption cross section of ^{237}Np is also considerable. The ^{235}U depletion chain becomes:

$$^{235}U \xrightarrow{n,\gamma} {}^{236}\text{U}$$

- ^{238}U chain

  The effect of eliminating also ^{243}Am, ^{242}Am and ^{241}Am from the ^{238}U also has an effect on the K_{eff} since the maximum error in K_{eff} is 0.263 mk, reached after 300 days of operation. This is due to the fact that even if the concentrations of these isotopes are low in CANDU reactors, their absorption cross section are important. This error is especially due to the elimination of ^{241}Am. In fact, we have investigated the case where only ^{243}Am and ^{242}Am are eliminated. The results show that the error in K_{eff} is insignificant. Hence, to include or to eliminate these two isotopes in any core calculation will not affect the results. The ^{238}U depletion chain can be written as follows:

$$^{238}U \xrightarrow{n,\gamma} {}^{239}Np \xrightarrow{\lambda} {}^{239}Pu \xrightarrow{n,\gamma} {}^{240}Pu \xrightarrow{n,\gamma} {}^{241}Pu \xrightarrow{n,\gamma} {}^{242}Pu$$

- Combined ^{238}U and ^{235}U chains

  We have also considered the case where ^{243}Am, ^{242}Am, ^{241}Am and ^{237}NP are eliminated together. Table 1 shows that the error in reactivity is cumulative as the error in case 3 is equal to the sum of the error in case 1 and the error in case 2.

III.2 COMBINATION OF HEAVY ISOTOPES

The proposed approximation is applied to study the effect of the combination of heavy isotopes of ^{238}U and ^{235}U depletion chains on the effective multiplication. We have considered that the time dependence of the cross section of the pseudo-isotope generated is linear in time. The K_{eff} is calculated for 300 days with 10 time steps.

- ^{235}U chain

  Table 2 shows that the combination of ^{237}Np and ^{236}U reduce substantially the error in K_{eff} as the maximum error is reduced to 0.024 mk, error which seems insignificant. The ratio of the concentration of the pseudo-isotope and the concentration of ^{236}U given by a reference calculation remain equal to unity, and the maximum error on the concentration is 0.5 %. If we note by $^{236}U^{\star}$ the pseudo-isotope generated by the combination of ^{236}U and ^{237}Np, then the ^{235}U chain can be written as follows:

$$^{235}U \xrightarrow{n,\gamma} {}^{236}U^{\star}$$

- ^{238}U chain

  We have combined ^{243}Am, ^{242}Am, ^{241}Am, ^{242}Pu and ^{241}Pu to generate the new pseudo-isotope $^{241}Pu^{\star}$. Table 3 shows that in this case both the error in reactivity and in concentration are insignificant, in fact we observe that the maximum error in reactivity is 0.072 mk and the ratio of the concentration of the ^{241}Pu performed by a reference calculation, (where all the isotopes were included), and the concentration of the pseudo-isotope $^{241}Pu^{\star}$ remains equal to 1. The maximum error in concentration is 0.6%.

  We have also combined ^{243}Am, ^{242}Am, ^{241}Am, ^{242}Pu, ^{241}Pu and ^{240}Pu to create the new pseudo-isotope $^{240}Pu^{\star}$. Table 4 shows that the error in reactivity remain insignificant as the maximum error in K_{eff} is almost 0.6 mk, whereas the error in concentration becomes important especially after 200 days of operation of a reactor and reaches 31% after 300 days. The ^{238}U chain can be written as follows.

$$^{238}U \xrightarrow{n,\gamma} {}^{239}Np \xrightarrow{\lambda} {}^{239}Pu \xrightarrow{n,\gamma} {}^{240}Pu^{\star}$$

- Combined ^{238}U and ^{235}U chains

  After treating separately the ^{238}U chain and ^{235}U, we have investigated the case where a pseudo-isotopes is generated for each chain. The calculation where performed for 2 cases: (1) creation of $^{241}Pu^{\star}$ for ^{238}U chain and $^{236}U^{\star}$ for the ^{235}U chain and (2) the generation of $^{240}Pu^{\star}$ for ^{238}U and $^{236}U^{\star}$ for the ^{235}U. We have calculated the error in K_{eff} for the two cases. The results are shown in table 5. The error in reactivity in the first case can be neglected, whereas in the second case it becomes important as it reaches 0.85 mk. In both cases, we observe that the error in reactivity K_{eff} is generally not cumulative. The ^{235}U and ^{238}U depletion chains can be written in this case as

$$^{235}U \xrightarrow{n,\gamma} {}^{236}U^{\star}$$

and

$$^{238}U \xrightarrow{n,\gamma} {}^{239}Np \xrightarrow{\lambda} {}^{239}Pu \xrightarrow{n,\gamma} {}^{240}Pu^{\star}$$

IV. CONCLUSION

From our calculation we can conclude that:

- The effect on the critically of the elimination separately of ^{243}Am, ^{242}Am and ^{241}Am, and ^{237}Np of CANDU reactor can not be neglected.
- The combination of isotopes reduce both the number of isotopes to be treated in any burnup calculation and the error in K_{eff}.
- Considering only a linear time dependence of the cross section of the pseudo-isotope is a good approximation.
- The error in reactivity is cumulative only for the elimination of isotopes.

To extend this method to ^{239}Pu, and probably even to ^{239}Np we are considering the need of the quadratic time dependence of the cross section of the pseudo-isotope. Our ultimate goal is to develop an isotopic depletion method for reactor core calculations, where only a few isotopes are treated to follow the core burnup. Moreover, by this method we hope to avoid a tabulation method commonly used in a core calculation.

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Table 1: Relative error in K_{eff} over the period of operation

Time (day)	K_{eff} (reference)	Error (mk) case 1	Error (mk) case 2	Error (mk) case 3
0	1.117646	.000	.000	0.000
20	1.077950	.002	.000	0.002
50	1.078304	.039	.002	0.026
90	1.062493	.076	.019	0.077
110	1.051615	.092	.016	0.106
130	1.039962	.120	.026	0.147
150	1.028031	.156	.059	0.201
200	.998806	.214	.096	0.316
250	.972648	.304	.159	0.468
300	.950285	.395	.263	0.653

Table 2: Relative error in K_{eff} over the period of operation after the combination of ^{236}U and ^{237}Np

Time (day)	K_{eff} (reference)	Error (mk)	Ratio
0	1.117646	.000	0.0
20	1.077950	.005	1.0
50	1.078304	.002	1.0
90	1.062493	.010	1.0
110	1.051615	-.003	1.0
130	1.039962	-.006	1.001
150	1.028031	.006	1.001
200	.998806	.001	1.002
250	.972648	-.004	1.003
300	.950285	.024	1.005

Table 3: Relative error in K_{eff} over the period of operation after the combination of ^{243}Am,^{242}Am, ^{241}Am ^{242}Pu and ^{241}Pu

Time (day)	K_{eff} (reference)	Error (mk)	Ratio
0	1.117646	.000	0.0
20	1.077950	-.007	1.0
50	1.078304	.017	1.0
90	1.062493	.033	1.0
110	1.051615	.038	1.0
130	1.039962	.046	1.0
150	1.028031	.072	1.0
200	.998806	.071	1.001
250	.972648	.042	1.003
300	.950285	-.018	1.006

Table 4: Relative error in K_{eff} over the period of operation after the combination of ^{243}Am,^{242}Am, ^{241}Am ^{242}Pu, ^{241}Pu and ^{240}Pu

Time (day)	K_{eff} (reference)	Error (mk)	Ratio
0	1.117646	.000	0.0
20	1.077950	.118	1.00
50	1.078304	.428	1.020
90	1.062493	.484	1.045
110	1.051615	.382	1.060
130	1.039962	.230	1.077
150	1.028031	.051	1.096
200	.998806	-.379	1.155
250	.972648	-.584	1.227
300	.950285	-.508	1.316

Table 5: Relative error in K_{eff} over the period of operation, case 1: combination of ^{243}Am,^{242}Am, ^{241}Am ^{242}Pu, ^{241}Pu, ^{236}U and ^{237}Np, and case 2: combination of ^{243}Am, ^{242}Am, ^{241}Am, ^{242}Pu, ^{241}Pu, ^{240}Pu, ^{236}U and ^{237}Np

Time (day)	K_{eff} (reference)	Error (mk) case 1	Error (mk) case 2
0	1.117646	.000	.000
20	1.077950	-.0010	.067
50	1.078304	.0080	.212
90	1.062493	.0250	.016
110	1.051615	.0350	-.184
130	1.039962	.0479	-.395
150	1.028031	.0700	-.592
200	.998806	.0730	-.857
250	.972648	.0510	-.668
300	.950285	-.0080	-.017