ACTRAN : A CODE FOR DEPLETION CALCULATIONS IN PWR CORES AIMING THE PRODUCTION OF MINOR ACTINIDE FOR USING IN ADS

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ABSTRACT

Despite of the renewed willing to accept nuclear power as a mean of mitigate the climate changing, to deal with the long lived waste still cause some concerning in relation to maintain in safety condition, during so many years. A technological solution to overcome this leg of time is to use a facility that burn these waste, besides to generate electricity. This is the idea built in the accelerator driven systems (ADS). This technology is being though to use some minor actinides (MAs) as fuel. This work presents a program to assess actinide concentrations, aiming a fertile-free fuel to be used in the future ADS technology. For that, use was made of a numerical code to solve the steady-state multigroup diffusion equation 3D to calculate the neutron fluxes, coupled it with a new code to solve, also numerically, depletion equations, named ACTRAN code. This paper shows the simulation of a PWR core during the residence time of the nuclear fuel, for three years, and after, for almost four hundred years, to assess the MAs production. The results show some insight in the best management to get a minimum amount of some MAs to use in the future generations of ADS.

Key Words: Fuel Cycle, Depletion Equations, Multigroup Diffusion Theory, Numerical Methods, ADS

1. INTRODUCTION

Until now the main drawback in the nuclear power production is related with the safe disposal and isolation of either spent fuel from reactors or, if the reprocessing option is used, wastes from reprocessing plants. These materials must be isolated from the biosphere until the radioactivity contained in them has diminished to a safe level. These preoccupations arise from the fact that the radioactivity of spent fuel after service is due primarily to the radioisotopes generated by fission, despite the activity levels of the fission products rapidly decrease because their short half-lived, for most of them. On the other hand, a small amount of transuranic waste is generated by successive neutron capture in uranium, most of them α-emitters, in comparison with fission products, which are β- and γ-emitters. Spent fuel consists of radionuclides with different half-lives (2.9x10 1 – 4.5x10 9 years) and toxities, basically composed by highly radioactive fission products and transuranic elements. Based in a dose of 20 Sv, reference to acquire mortal cancer with 100% certainty, the most toxic of them are: 90 Sr, 99 Tc, 129 I, 135 Cs, 137 Cs, fission products, and, 237 Np, 238 Pu, 239 Pu, 240 Pu, 241 Pu, 242 Pu, 243 Am, 244 Am, 244 Cm, and 245 Cm, the transuranics (TRUs) [1]. Because of their small production during transmutation, some of these transuranics are called Minor Actinides (MAs): Np, Am,
Cm. The high-level wastes are stored in facilities above ground or shallow repositories, in close connection with its nuclear power plant, which can take almost $10^6$ years before the radiotoxicity became the order of the background. While the disposal issue is not urgent from a technical viewpoint, it is recognized that extended storage in the facilities is not acceptable since these ones cannot provide sufficient isolation in the long term and neither is it ethical to leave the waste problem to future generations.

A technique to diminish this time is to transmute these long-lived elements into short-lived elements [2]. The approach is to use an Accelerator Driven System (ADS), a sub-critical arrangement which uses a Spallation Neutron Source (SNS) [3, 4], after separation the minor actinides and the long-lived fission products (LLFP), to convert them to short-lived isotopes.

Since most of the facilities for transmutation require the use of some MAs as fertile-free fuel like $(Pu_{0.4}, Am_{0.5}, Cm_{0.1})O_{2-X} - Mo$ [1] because of their neutronic qualities, the start point is to assess the threshold of actinide concentrations, from what the facility for use in incineration and energy production becomes feasible. This work presents a preliminary methodology and some results, from calculus of these. For that, use was made of a numerical code to solve the steady-state multigroup diffusion equation 3D to calculate the neutron fluxes, coupled it with a new code to solve, also numerically, depletion equations, to obtain the minor actinide concentrations. However, if MAs present good neutronic properties, they still are troublesome for reprocessing. This paper shows the simulation of a PWR core during the residence time of the nuclear fuel, for three years, and after, for almost four hundred years, to assess the MAs production. The results show some insight in the best management to get a minimum amount of some MAs to use in the future generations of ADS.

2. METHODOLOGY

The preliminary considerations are based on the Uranium chain. For that we refer to the chart of actinides from reference [5].

![Figure 1: Chain transmutation.](image-url)
The following alpha decays are considered:

\[ \begin{align*}
Pu^{238} & \rightarrow U^{234} + \alpha, & Am^{241} & \rightarrow Np^{237} + \alpha, & Cm^{242} & \rightarrow Pu^{238} + \alpha, \\
Pu^{239} & \rightarrow U^{235} + \alpha, & Am^{242m} & \rightarrow U^{238} + \alpha, & Cm^{243} & \rightarrow Pu^{239} + \alpha, \\
Pu^{240} & \rightarrow U^{236} + \alpha, & Am^{243} & \rightarrow Np^{239} + \alpha, & Cm^{244} & \rightarrow Pu^{240} + \alpha, \\
Pu^{242} & \rightarrow U^{238} + \alpha, & Cm^{245} & \rightarrow Pu^{241} + \alpha. 
\end{align*} \]

On the other hand, we will disregard the following alpha decays:

\[ \begin{align*}
U^{234} & \rightarrow Th^{230} + \alpha, & U^{236} & \rightarrow Th^{232} + \alpha, & U^{238} & \rightarrow Th^{234} + \alpha \text{ and } Pu^{236} & \rightarrow U^{232} + \alpha.
\end{align*} \]

In this way, the transmutation equations are:

\[ \frac{dX_j}{dt} = \sum I(j) \left[ \sum_k y^j_k \lambda_i + \sum G \sigma_{P_k} \varphi^j_k \right] Y_i - \left[ \lambda_j + \sum G \sigma_{L_k} \varphi^j_k \right] X_j, \quad j = 1, 2, 9 \quad (1) \]

with:

\[ I(j) = \text{Number of nuclides that decay or/and capture to for the } j\text{-th nuclide,} \]

\[ \sigma^j_{P_k} = \sigma^j_{n,2ng} + \sigma^j_{R}, \quad (2) \]

\[ \sigma^j_{L_k} = \sigma^j_{n,2ng} + \sigma^j_{fg} + \sigma^j_{R}, \quad (3) \]

and \( y^j_k \) represents the branching of nuclide \( Y_i \), having a decay constant \( \lambda_i \), contributing to produce the nuclide \( X_j \), in such way that,

\[ \sum_k y^j_k = 1. \quad (4) \]

The average fluxes are given by:

\[ \bar{\varphi}_g = \frac{\int_{\text{Reactor}} \int_{\text{Reactor}} \int_{\text{Reactor}} \phi_g (\vec{r}, t) dr^3}{\int_{\text{Reactor}} dr^3}. \quad (5) \]
3. NUMERICAL SOLUTION

Equation set (1) is put in a matrix form such that:

\[
\frac{dY}{dt} = MY
\]  

(6)

where:

\[
Y = \text{col}\{X_1, X_2, X_3, \ldots, X_j, \ldots, X_{29}\}.
\]  

(7)

Discretizing the time in time intervals \(h\), in such way that:

\[
t_{n+1} = t_n + h,
\]  

(8)

and using the Crank-Nicholson approximation, we have:

\[
Y_{n+1} \approx Y_n + h\left(\frac{dY^n}{dt} + \frac{dY^{n+1}}{dt}\right).
\]  

(9)

The numerical solution along the time is given by:

\[
\left( I - \frac{h}{2} M \right) Y_{n+1} = \left( I + \frac{h}{2} M \right) Y_n.
\]  

(10)

Redefining the following matrix and vector:

\[
A = \left( I - \frac{h}{2} M \right),
\]  

(11)

\[
B_n = \left( I + \frac{h}{2} M \right) Y_n.
\]  

(12)

Now we have:

\[
AY_{n+1} = B_n.
\]  

(13)

Since \(M\) is a sparse matrix, \(A\) also is. A fast way to obtain \(Y_{n+1}\) is to use an iterative method like Successive-Over Relaxation Methods, splitting \(A\) at matrices \(L\) (lower), \(D\) (diagonal) and \(U\) (upper). With this, in iteration \(p+1\), one has:

\[
Y_{n+1}^{p+1} = (1 - \alpha)Y_{n+1}^p + \alpha D^{-1} \left[ L Y_{n+1}^{p+1} - U Y_{n+1}^p + B_n \right],
\]  

(14)

where \(1 < \alpha < 2\).
4. RESULTS

The transmutation chain represented in Fig. 1 was solved by Eq. 14, using data extracted of reference [5], shown at Table 1. We started with fresh fuel for PWR, with an enrichment of 2.78% in U235. It was used a neutron flux of \( \phi = 2.99\times10^{14} \frac{n}{cm^2 s} \), for a residence time of 3 years. After that, the flux was set null for until 370 years.

Table 1: One Group Neutron Cross-Section for PWR Uranium Fuel (Barn).

<table>
<thead>
<tr>
<th>Nuclide</th>
<th>( \sigma_r ) (in ( \sigma_{en} ))</th>
<th>( \sigma_f )</th>
<th>( \sigma_{n2n} )</th>
<th>T_{1/2}(y)</th>
</tr>
</thead>
<tbody>
<tr>
<td>(^{235}\text{U})</td>
<td>10.4600</td>
<td>46.7100</td>
<td>0.002696</td>
<td>7.04E+08</td>
</tr>
<tr>
<td>(^{236}\text{U})</td>
<td>7.5410</td>
<td>0.1975</td>
<td>0.002644</td>
<td>2.34E+07</td>
</tr>
<tr>
<td>(^{238}\text{U})</td>
<td>0.9021</td>
<td>0.1004</td>
<td>0.005525</td>
<td>4.47E+09</td>
</tr>
<tr>
<td>(^{237}\text{Np})</td>
<td>32.1200</td>
<td>0.5244</td>
<td>0.000275</td>
<td>2.14E+06</td>
</tr>
<tr>
<td>(^{238}\text{Pu})</td>
<td>34.6700</td>
<td>2.4650</td>
<td>0.000167</td>
<td>8.77E+01</td>
</tr>
<tr>
<td>(^{239}\text{Pu})</td>
<td>58.6100</td>
<td>106.2000</td>
<td>0.001120</td>
<td>2.41E+04</td>
</tr>
<tr>
<td>(^{240}\text{Pu})</td>
<td>104.0000</td>
<td>0.5840</td>
<td>0.004448</td>
<td>6.56E+03</td>
</tr>
<tr>
<td>(^{241}\text{Pu})</td>
<td>38.6800</td>
<td>118.1000</td>
<td>0.007518</td>
<td>1.44E+01</td>
</tr>
<tr>
<td>(^{242}\text{Pu})</td>
<td>31.7200</td>
<td>0.4146</td>
<td>0.002307</td>
<td>3.75E+05</td>
</tr>
<tr>
<td>(^{241}\text{Am})</td>
<td>118.8000</td>
<td>1.1230</td>
<td>0.000328</td>
<td>4.33E+02</td>
</tr>
<tr>
<td>(^{242}\text{Am})</td>
<td>98.0400</td>
<td>466.2000</td>
<td>0.005670</td>
<td>1.41E+02</td>
</tr>
<tr>
<td>(^{243}\text{Am})</td>
<td>49.4870</td>
<td>0.3959</td>
<td>0.000207</td>
<td>7.37E+03</td>
</tr>
<tr>
<td>(^{242}\text{Cm})</td>
<td>5.8010</td>
<td>0.5591</td>
<td>0.000053</td>
<td>4.46E+01</td>
</tr>
<tr>
<td>(^{244}\text{Cm})</td>
<td>13.8200</td>
<td>0.8746</td>
<td>0.001048</td>
<td>1.81E+01</td>
</tr>
</tbody>
</table>

Fig. 2 shows a depletion calculation using data from table 1. Fig. 3, from reference 6, shows a common depletion result for a typical PWR, using an approximated analytical solution. ACTRAN results show a consistent behavior compared with analytical ones. It should be pointed out that these kinds of results are strongly dependent of the nuclear cross-sections. The comparison has only a qualitative value.
Figs. 4, 5 and 6, show Pu239, Am241 and Cm244 fractions, for different time of duration, and three different flux levels to simulate different burnups. As we can see, at the end of these duration times, Pu239 and Am241 exhibit the same trends in raising their concentration for those intermediary burnup. That means that um optimization for obtaining those fuels for use in the ADS technology can be expected.
The rising concentration of Cm244, despite of its small amount, it is strongly dependent of reactor burnup.
At Fig. 7 we have the behavior of Am241 for one period of 370 years. We can observe that even the maximum concentration is reached about 50 year. That can indicate that for use in ADS, this is the best epoch to extract Am241 from waste fuel.

![Graph showing Am241 fraction over time](image)

**Figure 7: Am241 composition for 370 years.**

3. CONCLUSIONS

The preliminary results confirmed the sense that a refined management of the minor actinide can lead to an optimum use of them for future ADS technology, if free-fertile fuels become one condition necessary to use an open fuel cycle option.

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REFERENCES


