Fast Nuclear Reactor Fuel Depletion Analysis Using Fourth Order Runge-Kutta Method

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Abstract. Calculation of fuel depletion involved processing of nuclear data as a microscopic cross section. Macroscopic cross section calculations were performed by multiplying the microscopic cross sections by the density of the nuclide and all materials involved in the reactor itself. Diffusion equation calculations were performed in a two-dimensional cylindrical coordinate system, which involved macroscopic cross sections to obtain the distribution of neutron fluxes, neutron source densities, power densities, and k-eff. The calculation resulted in the k-eff of 1.0017918649. Thus, we concluded that the reactor was in super critical condition. Next, we used the maximum flux to be calculated in the depletion equation to get a new nuclide density for 10 years. The process of calculating depletion involved a system of twenty-eight differential equations, obtained from the nuclear transmutation chain. It took a long time and was difficult to solve analytically. In this research, the system of differential equations was numerically solved by the Fourth order Runge-Kutta Method to get the nuclide density curve with respect to time.

1. Introduction
The processes that occur in the nuclear reactor involve two types of nuclear reactions. These reactions are the radioactive decay reactions and the nuclear collision reactions.

1.1. Radioactive Decay Reactions
Radioactivity is the emission of particles from unstable nuclides. It will transmute one nuclide to others to reach steady state conditions. Based on types of emitted particles, the radioactive decay reactions are divided into three categories, namely:

1.1.1. Alpha Decay Reactions occur when an unstable nucleus transmute itself to another nucleus, accompanied by the emission of alpha particle, \( ^{4}\text{He} \).

1.1.2. Beta Decay Reactions occur when an unstable nucleus transmute itself to another nucleus, accompanied by emission of beta particles and neutrino. If negatively charged, the particles are called electrons. But, if positively charged, the particles are called positrons. The examples of alpha and beta decay can be seen in Figure 1 and summarized in Table 1 and 2 respectively.
Figure 1. Transmutation chain used on burn-up equation modeling. This chain involved 28 pieces of isotopes. [1],[2]

| Reactions               | Half Life | Decay Constants   |
|-------------------------|-----------|-------------------|
| $^{242}\text{Cm} \rightarrow ^{238}\text{Pu} + ^{4}\text{He}$ | 163 d     | 0.000177 $h^{-1}$ |
| $^{243}\text{Cm} \rightarrow ^{239}\text{Pu} + ^{4}\text{He}$ | 32 y      | 2.4727E-06 $h^{-1}$ |
| $^{244}\text{Cm} \rightarrow ^{240}\text{Pu} + ^{4}\text{He}$ | 17.6 y    | 4.49582E-06 $h^{-1}$ |
| $^{241}\text{Am} \rightarrow ^{237}\text{Pu} + ^{4}\text{He}$ | 458 y     | 1.63147E-07 $h^{-1}$ |
| $^{238}\text{Pu} \rightarrow ^{234}\text{U} + ^{4}\text{He}$ | 86 y      | 9.20074E-07 $h^{-1}$ |

1.1.3. Gamma Decay Reactions occur when an unstable nucleus transmutes itself to another nucleus, accompanied by massless and uncharged photon emissions. For example, the decay of metastable americium, which can be seen in Figure 1. It is accompanied by the following internal conversion reaction:

$^{242}\text{Am}^* \rightarrow ^{242}\text{Am} + ^{0}\gamma$[1],[2]
Table 2. Beta Decay Data [1],[2]

| Reactions                  | Half Life | Decay Constants   |
|----------------------------|-----------|-------------------|
| $^{237}_{92}U \longrightarrow ^{237}_{93}Np + _{-1}^0\beta + _0^0\bar{\nu}_e$ | 6.75 d    | 0.00427869 h$^{-1}$ |
| $^{239}_{92}U \longrightarrow ^{239}_{93}Np + _{-1}^0\beta + _0^0\bar{\nu}_e$ | 23.5 m    | 1.76973748 h$^{-1}$ |
| $^{238}_{93}Np \longrightarrow ^{238}_{94}Pu + _{-1}^0\beta + _0^0\bar{\nu}_e$ | 2.1 d     | 0.01375292 h$^{-1}$ |
| $^{239}_{93}Np \longrightarrow ^{239}_{94}Pu + _{-1}^0\beta + _0^0\bar{\nu}_e$ | 2.35 d    | 0.01228984 h$^{-1}$ |
| $^{240}_{93}Np \longrightarrow ^{240}_{94}Pu + _{-1}^0\beta + _0^0\bar{\nu}_e$ | 7.5 m     | 5.54517744 h$^{-1}$ |
| $^{241}_{94}Pu \longrightarrow ^{241}_{95}Am + _{-1}^0\beta + _0^0\bar{\nu}_e$ | 14.3 y    | 5.53331E-6 h$^{-1}$ |
| $^{242}_{95}Am \longrightarrow ^{242}_{96}Cm + _{-1}^0\beta + _0^0\bar{\nu}_e$ | 16 h      | 0.04332169 h$^{-1}$ |
| $^{243}_{94}Pu \longrightarrow ^{243}_{95}Am + _{-1}^0\beta + _0^0\bar{\nu}_e$ | 4.98 h    | 0.13918618 h$^{-1}$ |
| $^{244}_{95}Am \longrightarrow ^{244}_{96}Cm + _{-1}^0\beta + _0^0\bar{\nu}_e$ | 26 m      | 1.599570417 h$^{-1}$ |
| $^{243}_{95}Am \longrightarrow ^{243}_{94}Pu + _{-1}^0\beta + _0^0\bar{\nu}_e$ | 16 h      | 0.04332169 h$^{-1}$ |

Internal conversion is the radioactive decay process, in which the gamma ray, emitted from the nucleus is absorbed by one of electrons in the nucleus that causes the electrons bounce out of atomic nuclei. Then the electron vacancy is filled by other electron on the other skin, accompanied by certain emissions.

1.2. Nuclear Collision Reactions

In general, the nuclear collision reactions can be written as,

$$a + B \longrightarrow C + d$$

or

$$B(a, d)C$$

Some nuclear collision reactions, which may occur in the reactor are,

(i) Nuclear fission reaction (n,fission) The nuclear fission reaction produces a large amount of heat energy. These reactions are accompanied by the release of 2 to 3 pieces of neutrons. Mathematically, these reactions can be written as,

$$\frac{1}{3}n + \frac{A}{Z}X \longrightarrow \frac{A_1}{Z_1}X + \frac{A_2}{Z_2}X + 2\text{to}3\text{neutron} + \text{Energy}$$

If the burnt fuel is Uranium-235 then the fission reaction that may occur can be expressed with the following reactions,

$$\frac{1}{3}n + ^{235}_{92}U \longrightarrow ^{140}_{54}Xe + ^{94}_{38}Sr + 2\frac{1}{3}n + 200\text{MeV}$$

$$\frac{1}{3}n + ^{235}_{92}U \longrightarrow ^{141}_{56}Ba + ^{92}_{36}Kr + 3\frac{1}{3}n + 200\text{MeV}$$

(ii) Neutron capture reaction

This reaction is also well known as radiative capture reaction. This term was used because the reaction can produce gamma ray radiation, which appears when an unstable nucleus transmutes itself to the new stable nucleus. The excess energy, possessed by a new atomic nuclei is released in the form of gamma radiations.
\[
\frac{1}{6}n + ^{234}_{92}U \longrightarrow (^{235}_{92}U)^* + ^{235}_{92}U + 0\gamma
\]
\[
\frac{1}{6}n + ^{237}_{93}Np \longrightarrow (^{238}_{93}Np)^* + ^{238}_{93}Np + 0\gamma
\]

Constants of calculation for the fuel depletion simulation are:

- Cross Section Reaction Cross Section is probability of occurrence of nuclear reactions. Generally, there are two types of cross section. Namely,
  
  (i) Microscopic cross section \( \sigma \) is the probability of occurrence of nuclear reaction on a single atomic nucleus. Mathematically, it can be written as,
  
  \[
  \sigma = \frac{R}{IN_A} \tag{1}
  \]
  
  Schematically, microscopic cross section data can be grouped as,

  ![Figure 2. Grouping scheme of microscopic cross section[3]](image)

  Note that \( \sigma \) is microscopic cross section in barn (1 barn = \( 10^{-24} \text{cm}^2 \)), \( R \) is reaction rate in \( \frac{\#}{\text{cm}^2 \text{ second}} \) and \( N_A \) is the target density in \( \frac{\#}{\text{cm}^2} \).

  (ii) Macroscopic cross section is the probability of occurrence of a nuclear reaction at a lump of material at the macro level. Relationship between microscopic and macroscopic cross section can be written as,

  \[
  \Sigma = N\sigma \tag{2}
  \]

  Note that \( \Sigma \) is Macroscopic cross section in \( \text{cm}^{-1} \), \( \sigma \) is macroscopic cross section in \( \text{cm}^2 \) and \( N \) is atomic density.
Table 3. Specification of the Reactor used in the calculation[4]

| Parameters                   | Specification     |
|------------------------------|-------------------|
| Power Reactor                | 1500 MWt          |
| Type of Fuel                 | UN-PuN            |
| Coolant                      | Pb-Bi             |
| Operation time without refueling | 4 years         |
| Average power density        | 300 W/cc          |
| Maximum power density        | 350 W/cc          |
| Average Burn-Up              | 9 %               |
| Volume fraction of coolants materials | 45% (44.5% Pb dan 55.5% Bi) |
| Volume fraction of the fuel materials | 35 %          |
| Plutonium nitrate enrichment in the fuel average | 18 %          |
| Cladding (stainless steel SS316) | 20 %          |

Table 4. Fuel Characteristics [5]

| Type of material | Melting point | Density (gr/cm3) |
|------------------|---------------|-----------------|
| UN               | 2870          | 14.3            |
| PuN              | 2550          | 14.4            |

2. Methods of Calculation

2.1. Numerical Solution of 2 (r,z) Dimension 1 Group Diffusion Equation in Cylindrical Coordinates

\[
- \nabla \cdot D(\mathbf{r}) \nabla \phi(\mathbf{r}) + \Sigma_a(\mathbf{r})\phi(\mathbf{r}) = \frac{1}{k_{\text{eff}}} S(\mathbf{r}) S(\mathbf{r}) = \nu \Sigma_f(\mathbf{r}) \phi(\mathbf{r}) \phi(\mathbf{r}) = S(\mathbf{r}) = 0 \quad (3)
\]

Figure 3. 2D Cylindrical Reactor Core
Discretization of the 2D diffusion equation in gauss seidell iteration form can be written as follows[6],[7],

\[
\phi_{i,j}^{n+1} = \frac{1}{D \text{keff}} S_{i,j} + \frac{1}{D} \left( \frac{\Delta r^2}{2} \phi_{i+1,j}^{n+1} + \frac{\Delta r^2}{2} \phi_{i-1,j}^{n+1} + \frac{\Delta z^2}{2} \phi_{i,j+1}^{n+1} + \frac{\Delta z^2}{2} \phi_{i,j-1}^{n+1} \right) + \frac{\Sigma_a}{D}\left( \frac{1}{\Delta r^2} + \frac{1}{\Delta z^2} \right)
\]

(4)

Note that for \(r \neq 0\); \(i = 1 \rightarrow nr - 1; j = 1 \rightarrow nz - 1\), \(nr\) is the number of radial partition, and \(nz\) is the number of axial partition.

\[
\phi_{0,j}^{n+1} = \frac{4 \phi_{1,j}^{n}}{\Delta r^2} + \frac{\phi_{0,j+1}^{n} + \phi_{0,j-1}^{n}}{\Delta z^2} + \frac{1}{D \text{keff}} S_{0,j}
\]

(5)

Note that \(r = 0; j = 1 \rightarrow nz - 1\), \(nz\) is the number of axial partition.

2.2. Numerical Solution of Fuel depletion equation using fourth order Runge-Kutta method

With the assumption that the nuclides present in the fuel is initially \(N_1\) to \(N_7\) and generated new nuclides are from \(N_8\) to \(N_{28}\). Nuclide notations and the Burn-up equations can be listed and summarized in Figure 2.2.

The generally discretized form of the burn-up equation based on the transmutation chain can be written as [8],

\[
N[i]_{k+1} = N[i]_k + \frac{h}{6} \left( k_{1N[i]} + 2k_{2N[i]} + 2k_{3N[i]} + k_{4N[i]} \right)
\]

(6)
Figure 4. Nuclide Notation and fuel depletion equation
3. Results and Discussions

3.1. Neutronic Calculation

Figure 5. Input and Output for neutronic calculation

Based on the geometrical guesses and owned materials, the calculation gives new keff of 1.0017918649 > 1. This value corresponds to the comparison between material and geometrical buckling with the value of $B^2_m = 0.098^2 > B^2_g = 0.033^2$. Based on the results in figure 5, the neutron flux distribution has a maximum peak value. The maximum and the average value generated were $2.7e^{+14}$ and $4.36e^{+14}$ ($\text{# cm}^{-2}\text{Sec}^{-1}$) respectively. The unsymmetrical graph obtained is due to the instability of iterations that depend on the determination of the geometrical size and the selection of radial and axial partition. Based on the results from figure 6(a) to figure 6(d), we can also see that source density has the same distribution with neutron flux. This results were obtained because the neutron source equation is proportional to the neutron flux by the following relationship,

$$S_{ij} = \nu \Sigma_f \phi_{ij}$$

(7)

Based on the simulation, The obtained maximum and average neutron source density were $8.38e^{+12}$ and $1.35e^{+13}$ ($\text{# cm}^{-2}\text{Sec}^{-1}$) respectively. Similarly, the power density, depicted in figure 6(e) and 6(f), also have the same distribution with the previous neutron fluxes. Based on the following equation,

$$Q^\prime\prime\prime_{ij} = \nu \Sigma_f \phi_{ij}$$

(8)

The obtained maximum and average power densities were 92.963 and 150.083 ($\text{watt cm}^{-3}$). 


3.2. Depletion Analysis

By using the input values:

- step time : $1 \times 10^{-5}$ hour
- $t_{\text{final}} : 10 \text{ years} = 10 \times 365 \times 24 \text{ hour}$

the nuclide density changes can be explained by the following analysis,

- The density of U-235 and U-238 as fuels reduce due to the fission reactions.
- From 0 to 4 years, the density of plutonium as a fuel is relatively constant. This result is due to the fact that the rate of change of Curium alpha decay and Neptunium Beta Decay are the same as plutonium absorption. In other words, the absorption reaction that is the sum of the fission and radiative capture reaction were balanced by alpha and beta decay reaction.
- Until six years, $Pu_{241}^{\alpha}$ decreases since the rate of beta decay and absorption reaction are greater than that of the neutron capture.
Figure 6. Netronic calculation results: 6(a) Axial neutron flux, 6(b) Radial neutron flux, 6(c) Axial neutron source density, 6(d) Radial neutron source density, 6(e) Axial power density, and 6(f) Radial power density.
Figure 7. Selected outputs of depletion equation: 7(a) $N_1 = U^{235}_{92}$, 7(b) $N_2 = U^{238}_{92}$, 7(c) $N_3 = Pu^{238}_{94}$, 7(d) $N_4 = Pu^{239}_{94}$, 7(e) $N_5 = Pu^{240}_{94}$
4. Conclusion

On the neutronic analysis, we obtained the profile similarity of flux, source density and power density distribution because of their proportionality. After K-eff calculations compared to the buckling quantitative analysis, we can conclude that the reactor is in a supercritical state (New Keff = 1.0017918649). On the depletion analysis, we obtained that the formed fuel from natural uranium is considerably reduced while plutonium is growing. Then it is proven that generated new nuclides with certain increasing characteristics are according to the involved reactions.

References

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