FUEL DEPLETION AND RADIOACTIVE DECAY

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The depletion equation

- Prediction of changes in the fuel composition caused by neutron irradiation and radioactive decay

$$\frac{\partial N_i(\underline{r},t)}{\partial t} = \sum_{j=1}^{n} b_{j\to i}\lambda_j N_j(\underline{r},t) + \sum_{k=1}^{n}\int_0^{+\infty} dE[f_{k\to i}(E)\sigma_k(E)\phi(\underline{r},E,t)]N_k(\underline{r},t) +$$
$$-\left\{\lambda_i + \int_0^{+\infty} dE[\sigma_i(E)\phi(\underline{r},E,t)] + R_i(\underline{r},t)\right\}N_i(\underline{r},t) + F_i(\underline{r},t)$$

  - N_i nuclide *i* atom density (atoms/b-cm)
  - $b_{j\to i}$ decay branching ratio of nuclide *j* to *i* (%)
  - λ decay constant (1/s)
  - $f_{k\to i}$ nuclear reaction branching ratio of nuclide *k* to *i* (%)
  - σ cross-section (b)
  - Φ neutron flux (neutrons/cm^2-s)
  - R removal fraction (%)
  - F feed rate (atoms/s)

Zero-dimensional depletion equation

- The depletion equation can be simplified in zero-dimensional one-energy-group

$$\frac{dN_i}{dt} = \sum_j \left(f_{j \to i} \sigma_j^{tot} \alpha \phi + \gamma_{j \to i} \lambda_j \right) N_j - \left(\sigma_i^{tot} \alpha \phi + \lambda_i \right) N_i + F_i - R N_i$$

- The accuracy of the one-group solution depends on the accuracy of the effective cross-sections
  - predefined libraries
  - transport or diffusion models

Computer Codes for Depletion

- ORIGEN2, ORIGEN-S, CINDER, …
- In this class we will focus on ORIGEN2 and ORIGEN-S
  - Both developed from the original ORIGEN (Oak Ridge Isotopes GENerator)

ORIGEN-S	ORIGEN2
Part of the SCALE package	Stand alone
3 energy groups	1 energy group
1946 isotopes	1696 isotopes
Up to date data	Obsolete data

The exponential matrix technique

- Depletion equation in matrix form (homogeneous problem)

$$\frac{d\underline{N}}{dt} = \underline{\underline{T}}\,\underline{N}$$

  - N composition vector
  - T transition matrix

- Exponential matrix solution

$$\underline{N}(t) = \underline{N}(0)e^{\underline{\underline{T}}t}$$

$$e^{\underline{\underline{T}}t} = \sum_{m=0}^{\infty} \frac{\left(\underline{\underline{T}}t\right)^m}{m!} = \underline{\underline{I}} + \underline{\underline{T}}t + \frac{\left(\underline{\underline{T}}t\right)^2}{2} + ..$$

Recursion relation for the exponential matrix solution

$$\underline{N}(t) = \underline{N}(0)e^{\underline{\underline{T}}t} = \underline{N}(0)\left[\underline{\underline{I}} + \underline{\underline{T}}t + \frac{\left(\underline{\underline{T}}t\right)^2}{2} + \frac{\left(\underline{\underline{T}}t\right)^3}{6} + \dots\right] =$$

$$= \underline{N}(0)\left[\underline{\underline{I}} + \underline{\underline{T}}t + \underline{\underline{T}}t\frac{\underline{\underline{T}}t}{2} + \frac{\underline{\underline{T}}t}{3}\frac{\left(\underline{\underline{T}}t\right)^2}{2}\dots\right]$$

$$N_i = N_i(0) + t\sum_j a_{j\to i}N_j(0) + \frac{t}{2}\sum_k\left[a_{k\to i}t\sum_j a_{j\to k}N_j(0)\right] + \dots$$

Data vectors

$$C_i^0 = N_i(0),\; C_i^{n+1} = \frac{1}{n+1}\sum_j a_{j\to i} C_j^n$$

$$N_i = \sum_{n=0}^{\infty} C_i^n$$

- The solution requires storage of only two vectors

$$\underline{C}^n \; and \; \underline{C}^{n+1}$$

Not only exponential matrix

- Eigenvalues of the transition matrix are widely separated (e.g. half-lives range from fraction of seconds to millions of years) → the transition matrix is ill-conditioned
- Asymptotic solutions
  - Nuclides with removal lives < 14.4% of the time step and that do not have long-lived precursors
  - Short-lived nuclides with long-lived parents

Bateman equation

- Nuclides with removal lives <14.4% (ORIGEN2) or <10% (ORIGEN-S) of the time step and that do not have long-lived precursors
  - Constant concentration within the time step
  - Vondy's form of Bateman equation

Gauss-Seidel successive substitutions

- Short-lived nuclides with long-lived parents
  - Need to account for parent nuclides change in concentration during the time step
  - Gauss-Seidel successive substitutions algorithm

Required input data

$$\frac{dN_i}{dt} = \sum_j \left(f_{j\to i}\sigma_j^{tot}\alpha\phi + \gamma_{j\to i}\lambda_j \right) N_j - \left(\sigma_i^{tot}\alpha\phi + \lambda_i \right) N_i + F_i - RN_i$$

- Material composition
- Data libraries (photon, decay, xs)
- Depletion mode
  - power irradiation → power and time
  - flux irradiation → flux and time
  - decay → time

$$\frac{dN_i}{dt} = \sum_j \gamma_{j\to i}\lambda_j N_j - \lambda_i N_{ii}$$

Power/flux evaluation

- For constant power depletion, the neutron flux is determined as

$$\phi = \frac{P}{\sum_i N_i \sigma_i^{fis} E_i}$$

  - P total power
  - σ^{fis} fission cross-section
  - E_i recoverable energy per fission

$$E_i = 1.29927 \cdot 10^{-3} Z^2 A^{0.5} + 33.12 \ (MeV / fission)$$

- An predictor/corrector technique is used to account for the flux variation during the time step

What depletion codes can do

- ORIGEN2 and ORIGEN-S can calculate the radionuclide composition and other related properties of nuclear materials
  - Activity
  - Radio-toxicity
  - Decay heat
  - Absorption and fission rates
  - Neutron emission (for ORIGEN-S: in $U0_2$, borosilicate glass, and user defined matrix)
  - Photon emission

Oak Ridge Isotope GENerator 2

- ORIGEN2 is a point-depletion and radioactive-decay computer code for use in simulating nuclear fuel cycles and calculating the nuclide compositions and characteristics of materials contained therein.
- Developed from ORIGEN (1970s)
- ORIGEN2
  - Version 2.0 1980
  - Version 2.1 1999
  - Version 2.2 2002
- Written in FORTRAN
- Extremely fast

Format data structure

- Three nuclide groups (1300 nuclides—300 stable)
  - 1. Activation products (720)
  - 2. Actinides (130)
  - 3. Fission products (850)
- Nuclide identification number
  - Z x 10,000 + A x 10 + M
    - Z atomic number
    - A mass number
    - M state (0 for ground, 1 for excited)
  - Ex. 922350 → ^{235}U, 932421 → ^{242m}Am

Decay data library

- The decay data library contains
  - Decay half-lives and the decay branching fractions for beta decay to ground and excited states, positron plus electron capture decay to ground and excited states, internal transitions, alpha decay, spontaneous fission decay, and delayed neutron (beta plus neutron) decay
  - Recoverable heat per decay for each radioactive parent
  - Isotopic compositions of naturally occurring elements
  - Radionuclide maximum permissible concentration (MPC) values in air and water
- Required for all types of calculations

Decay data library

Group	NUCLID	Time unit	$T_{1/2}$	β^- to excited	β^+ to ground	β^+ to excited	α	IT
Group			Spont. fission	β^- and neutron	Decay heat (MeV)	Nat. abundance	MPC in air	MPC in water

```
2  922330  1    5.002E 12 0.0       0.0       0.0       1.000E 00 0.0
2               1.300E-12 0.0       4.904E 00 0.0       4.000E-12 3.000E-05
2  922340  1    7.716E 12 0.0       0.0       0.0       1.000E 00 0.0
2               1.000E-11 0.0       4.859E 00 5.400E-03 4.000E-12 3.000E-05
2  922350  1    2.221E 16 0.0       0.0       0.0       1.000E 00 0.0
2               2.600E-09 0.0       4.418E 00 7.200E-01 4.000E-12 3.000E-05
2  922360  1    7.389E 14 0.0       0.0       0.0       1.000E 00 0.0
2               1.200E-09 0.0       4.570E 00 0.0       4.000E-12 3.000E-05
2  922370  1    5.832E 05 0.0       0.0       0.0       0.0       0.0
2               0.0       0.0       3.192E-01 0.0       3.000E-08 1.000E-04
```

Time units

Designator	Unit
1	seconds
2	minutes
3	hours
4	days
5	years
6	stable
7	10^3 years
8	10^6 years
9	10^9 years

Photon data library

- The photon data base supplies the number of photons per decay in an 18-energy-group structure
  - Gamma rays
  - X-rays
  - Conversion photons
  - (α ,n) gamma rays
  - Prompt and fission product gamma rays from spontaneous fission
  - Bremmstrahlung
  - (Prompt gamma rays from fission and capture are not included)

Cross-section data library

- The cross-section data bases supply cross-sections and fission product yields
  - (n, γ) to ground and excited states
  - (n,2n) to ground and excited states
  - (n,3n)
  - Fission for the actinides
  - (n,p) and (n, α) for the activation products and fission products
- Fission product yields are included for fissions in ^{232}Th, ^{233,235,238}U and ^{239,241}Pu (plus yield values for ^{245}Cm and ^{249}Cf but the same as those for ^{241}Pu)

Cross-section data library

- Actinides format—single line

Library	NUCLID	Capture xs (b)	(n,2n) xs (b)	(n,3n) xs (b)	Fission xs (b)	Capture to excited xs (b)	(n,2n) to excited xs (b)	Control character
205	922320	1.128E 01	2.062E-03	9.195E-06	1.675E 01	0.0	0.0	-1.0
205	922330	7.581E 00	2.863E-03	1.931E-06	6.231E 01	0.0	0.0	-1.0
205	922340	1.923E 01	4.266E-04	1.349E-05	4.504E-01	0.0	0.0	-1.0
205	922350	1.046E 01	2.696E-03	1.160E-06	4.671E 01	0.0	0.0	-1.0
205	922360	7.541E 00	2.644E-03	2.207E-05	1.975E-01	0.0	0.0	-1.0
205	922370	4.424E 01	0.0	0.0	2.317E-01	0.0	0.0	-1.0
205	922380	9.021E-01	5.525E-03	4.597E-05	1.004E-01	0.0	0.0	-1.0

Cross-section data library

- Activation products (single line) and fission products (double line) format

Library	NUCLID	Capture xs (b)	(n,2n) xs (b)	(n, α) xs (b)	(n,p) xs (b)	Capture to excited xs (b)	(n,2n) to excited xs (b)	Control character
Library	Yield ^{232}Th	Yield ^{233}U	Yield ^{235}U	Yield ^{238}U	Yield ^{239}Pu	Yield ^{241}Pu	Yield ^{245}Cm	Yield ^{249}Cf

```
206   320740 5.509E-02 0.0       0.0       0.0       1.552E-02 0.0          1.0
206      1.21E-08 4.98E-06 1.68E-07 1.20E-07 3.38E-06 4.27E-08 3.56E-08 3.56E-08
206   270750 0.0       0.0       0.0       0.0       0.0       0.0          1.0
206      4.21E-07 7.39E-09 9.62E-09 5.06E-07 1.19E-10 8.07E-09 6.30E-09 6.30E-09
206   280750 0.0       0.0       0.0       0.0       0.0       0.0          1.0
206      1.24E-04 1.65E-05 9.94E-06 2.07E-04 5.50E-07 6.66E-06 5.20E-06 5.20E-06
206   290750 0.0       0.0       0.0       0.0       0.0       0.0          1.0
206      1.06E-03 9.30E-04 2.71E-04 2.41E-03 6.14E-05 1.47E-04 1.14E-04 1.14E-04
206   300750 0.0       0.0       0.0       0.0       0.0       0.0          1.0
206      1.49E-03 7.16E-03 1.09E-03 4.53E-03 8.81E-04 4.90E-04 3.83E-04 3.83E-04
```

Input/output files structure

- Input/output files are called TAPE
  - Ex. TAPE5.INP, TAPE6.OUT
- TAPE5.INP → input
- TAPE6.OUT → output
- TAPE4.INP → optional unit for input composition
- TAPE7.OUT → optional unit for output vector
- TAPE9.INP → decay and cross section data libraries
- TAPE10.INP → photon library

Vector structure

- VECTOR: a one-dimensional array that specifies the amount of each nuclide being considered in an ORIGEN2 case
  - Output vector
    - Printed when an output is produced
    - Designated by a positive integer
    - Limited to 12
  - Storage vector
    - For data storage only (cannot be output)
    - Designated by a negative number
    - Unlimited

References

- A.G. Croff, “A user’s manual for the ORIGEN2 computer code,” ORNL/TM-7175.
- A.G. Croff, “ORIGEN2: a versatile computer code for calculating the nuclide compositions and characteristics of nuclear materials,” *Nuclear Technology*, **62**, (1983).
- I.C. Gauld, O.W. Hermann, R.M. Westfall, “ORIGEN-S: SCALE system module to calculate fuel depletion, actinide transmutation, fission product buildup and decay, and associated radiation source terms,” ORNL/TM-2005/39.