NE585
NUCLEAR FUEL CYCLES
Burnup & Depletion
4a

R. A. Borrelli

University of Idaho

Idaho Falls Center for Higher Education

Learning objectives

Understand what burnup is

Evaluate fuel cycles using burnup

Performing the math

Learning nodes

Burnup
Fuel utilization
Fission product buildup

Depletion
Rate of change of fissile content
Bateman equations

Neutron balance
Continuity equation
Deriving solutions

Math
Real math

Even more learning nodes

Heat removal
Heat removal rate
Heat production rate
Conduction
Convection

Dimensionless heat transfer numbers

Boiling

Meltdown

Operation cycles

An operating cycle is 18 to 24 months for LWRs

Then we refuel

Why?

Core is shuffled on reload to maximize power

Assemblies spent about 3 to 4 years in the core

How does this change with microreactors?

Neutronics of the core change over each cycle

^{235}U is depleted and replaced by ^{239}Pu

Non fissile Pu, minor actinides, fission products are generated in the fuel

What are two of the big fission products?

Affects reactivity

Fuel utilization is measured in burnup

Why does nuclear have the weirdest metrics ever?

‘Yeah, we moved a couch and a new kitchen table Saturday afternoon. It took us 35 SWU.’

Burnup is the amount of energy extracted per unit mass

Amount of thermal energy produced per unit mass

‘Fissionability’

To increase burnup, increase the initial enrichment

Fissioning 1.05 g ^{235}U = 1 MWD

Fractional burnup = fissions/initial number of atoms

Burnup tracks radionuclide compositions from fresh loading to discharge

ηf decreases in time

Causes negative reactivity feedback

Neutron transport equation becomes nonlinear

Macroscopic cross sections depend on fuel composition

Uranium enrichment falls below 1% at discharge

But *Pu* and actinides make up for it

$$^{238}_{92}U +^{1}_{0} n \rightarrow^{239}_{92} U \rightarrow^{239}_{93} Np \rightarrow^{239}_{94} Pu$$

$$^{239}_{94}Pu +^{1}_{0} n \rightarrow^{240}_{94} Pu +^{1}_{0} n \rightarrow^{241}_{94} Pu \rightarrow^{241}_{95} Am$$

$$^{241}_{95}Am +^{1}_{0} n \rightarrow^{242}_{95} Am \rightarrow^{242}_{96} Cm$$

$$^{242}_{96}Cm +^{1}_{0} n \rightarrow^{243}_{96} Cm +^{1}_{0} n \rightarrow^{244}_{96} Cm$$

$$^{235}_{92}U +^{1}_{0} n \rightarrow^{236}_{92} U +^{1}_{0} n \rightarrow^{237}_{92} U \rightarrow^{237}_{93} Np$$

Fission probability of actinides affected by parity

Atom will fission if the compound nucleus energy exceeds fission barrier (5–6 MeV)

There are binding and kinetic energy components from the absorbed neutron

Atoms with even number of protons or neutrons are more tightly bound (parity)

Absorption of odd nucleons then releases more energy

Compound nucleus then forms in higher excited state

More likely to exceed fission barrier

^{239}Pu is the main fissile isotope in LWRs during second half of assembly lifetime

Conversion ratio is defined as the rate of production:depletion rate of fissile material

Greater than 1 = breeder

Uranium-plutonium requires a fast reactor which we talked about with EBR-I

Thorium can be run on the thermal spectrum with a Liquid Fluoride Thorium Reactor (LFTR)

Thermal Pu cross sections are higher than U

Accumulation of non-fissile plutonium, minor actinides, fission products increases spatial self-shielding

Hardens the flux spectrum

Decreases the reactivity worth of control rods

Prompt neutrons emitted in ^{239}Pu thermal fission greater than ^{235}U

Adds reactivity

But delayed neutron yield is lower

High plutonium decreases time constants

Lowers margin to prompt critical transient

Accumulation of fission products increases neutron absorption

^{149}Sm,^{135}Xe are the big ones

Super high absorption cross sections in the thermal region

Which again, just use fast reactors

Both radionuclides are fission products

Also in decay chains from precursors

Xe decays to Cs with 9 hour half life

Can be removed in 90 hours

Sm is stable

Xenon produces dead time

Production rate from I decay

Loss rate due to absorption during operation

Produces a positive feedback with power

Flux oscillates spatially in core

Concentration grows after shutdown because of I decay

Negative reactivity increase surpasses positive reactivity reserved in fuel

Startup is not possible

I decay is 6.6 h

Burnup and depletion calculations are the source terms for disposal

Heat production after shutdown is dominated by short lived, high activity radionuclides

But long term, heat isn't a big deal

It's the toxicity of long lived radionuclides like neptunium

Main radionuclides after shutdown are ^{239}U, ^{239}Np, ^{134}I, ^{138}Cs, ^{140}Cs

Short half lives – minutes to near an hour

Heat production affects fuel integrity after shutdown

Need for pools

The short lived fission products are most significant in an accident

Cs-137 can form gas compounds over 1300^{o}C

Fuel melt can release Sr-90, barium, ruthenium, lanthanum at about 3000^{o}C

I-131 contributes to radiation dose (inhaled)

Eight day half life

Cs-137 contamination affects land cultivation

Takeup in bone

Long lived isotopes in used fuel are the long term contributor to dose

Actinides – Pu-239, Pu-240, Np-237, Am-241, Am-243

Fission products – Tc-99, I-129, Cs-135

I-129 has 15 million year half life, and anionic, so it's very mobile

Cs-137, Sr-90 high heat but short lived; 300 years to decay away

Pu-238 has 88 year half life but we can use it in satellites

We have used Bateman equations for decay

But a generalized form can be used for overall depletion calculations in the reactor

$$\frac{dN_j}{dt} = \sum_{i \neq j} S_{i \to j} - \lambda_j N_j - \phi \sigma_j N_j \tag{1}$$

rate of change = (production) - (removal)

Source term is the sum of decay, transmutation, fission

$$S_{i \to j} = \lambda_i N_i + \phi \sigma_i N_i + \phi \gamma_i \Sigma_i^F \tag{2}$$

Simplified form for decay

$^{241}_{94}Pu \rightarrow ^{241}_{95}Am \rightarrow ^{237}_{93}Np$

$$\frac{dN_P}{dt} = -\lambda_P N_P \tag{3}$$

$$\frac{dN_A}{dt} = \lambda_P N_P - \lambda_A N_A \tag{4}$$

$$\frac{dN_N}{dt} = \lambda_A N_A - \lambda_N N_N \tag{5}$$

$$N_n(t) = \sum_i^n [N_i^0 \prod_{j=i}^{n-1} \lambda_j \sum_{j=i}^{n} \frac{e^{-\lambda_j t}}{\prod_{p=i}^{n}(\lambda_p - \lambda_j)}] \tag{6}$$

This is doable

The problem is when you include all the other reactions

Introduction of the Adding and Doubling Method for Solving Bateman Equations for Nuclear Fuel Depletion

Burnup calculations can get up to 2000 equations in one system

$$\begin{aligned} \underline{n}' &= \underline{\underline{A}}\,\underline{n} \\ \underline{n}(0) &= \underline{n}_0 \end{aligned} \tag{7}$$

Coefficient matrix contains loss terms on the diagonal and production on off-diagonal

$$\underline{n}(t) = e^{\underline{\underline{A}}t}\,\underline{n}_0 \tag{8}$$

$$e^{\underline{\underline{X}}} \equiv \sum_{k=0}^{\infty} \frac{1}{k!} \underline{\underline{X}}^k \tag{9}$$

Solution is reduced to the matrix exponential function

But it could still be up to 1700×1700

And 10^{21} eigenvalues

Approximation methods can be used to separate out short lived radionuclides

This is the approach used for Serpent

Several assumptions are needed to derive the Bateman equations

Reaction rates constant in time for Bateman

Cross sections constant in time for neutron transport

Changes in flux are averaged over one-group cross sections for Bateman

Changes in nuclide concentrations are in the macroscopic cross sections for neutron transport

Coupling the two gives a nonlinear system

Linearizing systems is common

Divide time domain into discrete depletion steps

Transport problem is solved assuming constant reaction rates over time interval

Flux spectrum is used to calculate microscopic transmutation cross sections

Depletion problem is solved assuming constant flux spectrum remains over time interval

Produces material compositions for the next transport solution

Selection of step length is a compromise between accuracy, cost

This is the same for all approximation schemes

Finite element method has a metric to determine spatial discretization v time step

Explicit methods based on sequential calls to transport and depletion solvers while proceeding to new steps

Implicit methods perform inner iterations to converge the two solutions before moving to the next step

Explicit methods are computationally less expensive

Subject to errors and instabilities when the depletion step is chosen too long

Burnup calculations can get up to 2000 equations in one system

$$y_{n+1} = y_n + h \cdot f(y_n, t_n) \tag{10}$$

$$y_{n+1} = y_n + h \cdot f(y_{n+1}, t_{n+1}) \tag{11}$$

Predictor-corrector methods also used

$$y^P_{n+1} = y_n + h \cdot f(y_n, t_n) \tag{12}$$

$$y_{n+1} = y_n + \frac{h}{2} \cdot [f(y_n, t_n) + f(y^P_{n+1}, t_{n+1})] \tag{13}$$

Selection of h is critical for stability

Predictor-corrector method applies two transport calculations per step

Predictor – reaction rates calculated at beginning of step

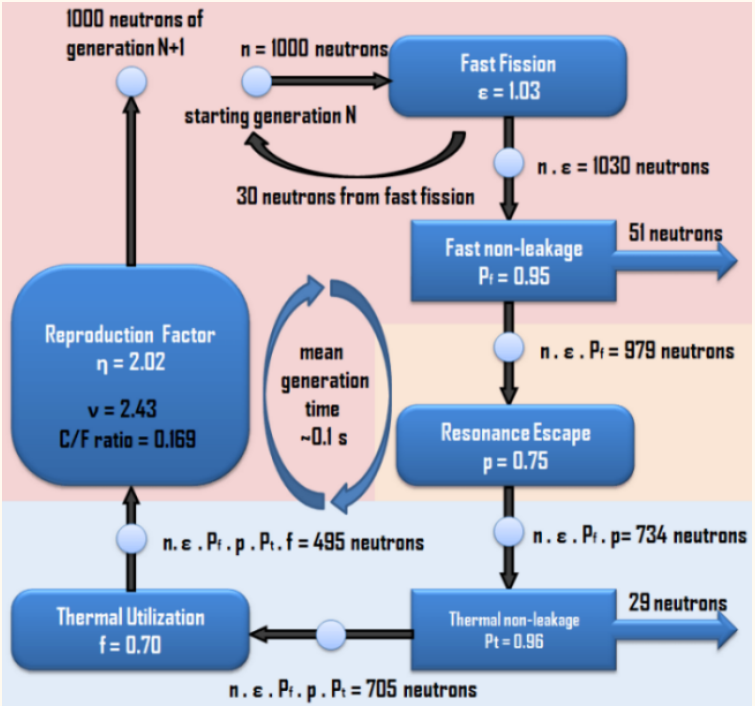
Corrector – new reaction rates calculated at end of step

Linear interpolate to get burnup over the step

More computational cost

Numerical methods for nuclear fuel burnup calculations

Neutron balance


1000 neutrons of generation N+1
n = 1000 neutrons
Fast Fission
ε = 1.03
starting generation N
n . ε = 1030 neutrons
30 neutrons from fast fission
Fast non-leakage
Pf = 0.95
51 neutrons
Reproduction Factor
η = 2.02
ν = 2.43
C/F ratio = 0.169
mean generation time ~0.1 s
n . ε . Pf = 979 neutrons
Resonance Escape
p = 0.75
n. ε . Pf . p . Pt . f = 495 neutrons
n . ε . Pf . p= 734 neutrons
Thermal Utilization
f = 0.70
Thermal non-leakage
Pt = 0.96
29 neutrons
n . ε . Pf . p . Pt = 705 neutrons

Neutron consumption = production rates in the critical reactor

N_M – atoms of fissile material per unit volume

σ_M – absorption cross section

$N_G,\ \sigma_G$ – fertile material

Fissionable material absorbs only thermal neutrons

$N_M \sigma_M \phi$ – neutron absorption rate by fissionable material

$\eta_M N_M \sigma_M \phi$ – fissions produce fast neutrons

Production of neutrons also comes from fast fission

Fast fission factor ϵ defines net rate of production of fast neutrons to production rate of fast neutrons by thermal fission

$\epsilon - 1$ – fast neutrons come from fission of fertile material with fast neutrons

$\epsilon \eta_M N_M \sigma_M \phi$ – production rate of fast neutrons from fission

Neutrons leak during scatter from the fast region to resonances

P_F – probability the neutrons do not leak

$\epsilon \eta_M N_M \sigma_M \phi P_F$ – rate fast neutrons do not leak

Neutrons can be absorbed in resonance region

p – probability that escape to thermal region

$\epsilon \eta_M N_M \sigma_M \phi P_F p$ – neutrons produced in thermal region

Finally, thermal neutrons leak

Neutrons complete a cycle as $\epsilon p \eta_M P_F P_T N_M \sigma_M \phi$ reach thermal region per unit volume per time

Thermal neutrons are consumed by absorption in fissionable material, nonfissionable material, leakage

Absorption in fissionable material leads to regeneration of fission neutrons

Let's derive the neutron balance in the critical reactor

Assume representative unit volume

N_M – atoms of single fissile species with absorption cross section σ_M

N_G – atoms of single fertile material with absorption cross section σ_G

Same for coolant, moderator, structure (lumped together)

Control absorbers

Assume steady state amounts of Xe, Sm

Rate of thermal neutron production = consumption for initial loading

$$\eta_M \epsilon p P_F P_T N_M \sigma_M \phi = DB^2\phi + N_M\sigma_M\phi + N_G\sigma_G\phi + \sum_P N_P\sigma_P\phi + N_{Xe}\sigma_{Xe}\phi + N_{Sm}\sigma_{Sm}\phi + N_E\sigma_E\phi \quad (14)$$

For the operating reactor, it is slightly more complicated

$$\sum_M \eta_M \epsilon p P_F P_T N_M \sigma_M \phi =$$
$$DB^2\phi + \sum_M N_M \sigma_M \phi + \sum_H N_H \sigma_H \phi + \sum_P N_P \sigma_P \phi + \sum_F N_F \sigma_F \phi + N_G \sigma_G \phi + N_{Xe} \sigma_{Xe} \phi + N_{Sm} \sigma_{Sm} \phi + N_E \sigma_E \phi \quad (15)$$

Production = sum of all fissionable species

H = nonfissionable higher isotopes (U-236, etc.)

F = fission products lower than Xe, Sm

Benedict notes

We can calculate burnup by deriving composition changes in time

$$\frac{dN_{25}}{dt} = -N_{25}\sigma_{25}\phi(t) \tag{16}$$

$$N_{25} = N_{25}^{0} e^{-\sigma_{25}\int_0^t \phi dt'} \tag{17}$$

$$\theta \equiv \int_0^t \phi(t')dt' \rightarrow \theta = \overline{\phi}t \tag{18}$$

$$N_{25} = N_{25}^{0} e^{-\sigma_{25}\theta} \tag{19}$$

Flux time expresses extent of exposure to irradiation

Defined in units of neutrons per square centimeter

Also called fluence

Typically if flux is $10^{14}\ n/cm^2/s$ and $10^7\ s$

Then flux time is on the order of $10^{21}\ n/cm^2$

So they call that neutrons per kilobarn because of course they do

U-236 is produced by capture in U-235

$$\frac{dN_{26}}{dt} = \frac{\alpha_{25}}{1+\alpha_{25}} \cdot N_{25}\sigma_{25}\phi - N_{26}\sigma_{26}\phi \tag{20}$$

$$\alpha \equiv \frac{\sigma_A}{\sigma_F} \tag{21}$$

$$\frac{1}{1+\alpha} = \frac{\sigma_F}{\sigma_F + \sigma_A} \tag{22}$$

$$\frac{\alpha}{1+\alpha} = \frac{\sigma_A}{\sigma_F} \cdot \frac{\sigma_F}{\sigma_F + \sigma_A} = \frac{\sigma_A}{\sigma_F + \sigma_A} \tag{23}$$

$$N_{26} = \frac{N_{25}^0 \sigma_{25}\alpha_{25}}{(\sigma_{25} - \sigma_{26})(1+\alpha_{25})} \cdot (e^{-\sigma_{26}\theta} - e^{-\sigma_{25}\theta}) \tag{24}$$

Pu-239 production is more complicated

$\frac{dN_{49}}{dt} =$

$+N_{28}^{0}\sigma_{28}\phi$ – production from thermal neutron absorption in U-238

$+\eta_X \epsilon P_F (1-p) N_X \sigma_X \phi$ – fission neutrons from X absorbed by Y in the resonance region

$-N_{49}\sigma_{49}\phi$ – thermal neutron absorption in Pu-239

$+\frac{\alpha_{28}}{1+\alpha_{28}} \cdot \frac{\epsilon - 1}{\eta_{28}-1}(\eta_{25}N_{25}\sigma_{25} + \eta_{49}N_{49}\sigma_{49} + \eta_{41}N_{41}\sigma_{41})\phi$ – fast neutron absorption

Net formation can be simplified with flux time

$$\frac{dN_{49}}{d\theta} = N_{28}^{0}\sigma_{28} + \kappa_{25}N_{25}\sigma_{25} - \gamma_{49}N_{49}\sigma_{49} + \kappa_{41}N_{41}\sigma_{41} \quad (25)$$

$$\kappa_m \equiv \eta_m \epsilon P_F(1-p) + \eta_m \frac{\alpha_{28}}{1+\alpha_{28}} \cdot \frac{\epsilon - 1}{\eta_{28} - 1} \quad (26)$$

$$\gamma_{49} = 1 - \kappa_{49} \quad (27)$$

The remaining isotopes of plutonium produced are Pu-242, Pu-241, Pu-240

$$\frac{dN_{42}}{d\theta} = \frac{\alpha_{41}}{1+\alpha_{41}} \cdot N_{41}\sigma_{41} - N_{42}\sigma_{42} \tag{28}$$

$$\frac{dN_{41}}{d\theta} = N_{40}\sigma_{40} - N_{41}\sigma_{41} \tag{29}$$

$$\frac{dN_{40}}{d\theta} = \frac{\alpha_{49}}{1+\alpha_{49}} N_{49}\sigma_{49} - N_{40}\sigma_{40} \tag{30}$$

An exact solution can be solved analytically, but why bother

Formation of Pu-239 by absorption of resonance neutrons from Pu-241 can be neglected

$$\kappa_{41} N_{41} \sigma_{41} << \gamma_{41} N_{49} \sigma_{49} \tag{31}$$

$$\frac{dN_{49}}{d\theta} = N_{28}^{0} \sigma_{28} + \kappa_{25} N_{25} \sigma_{25} - \gamma_{49} N_{49} \sigma_{49} \tag{32}$$

$$N_{49}(0) = 0 \tag{33}$$

Fission products are produced by ^{235}U

$$\frac{dN_{25}^{F}}{d\theta} = \frac{1}{1+\alpha_{25}} \cdot N_{25}\sigma_{25} \tag{34}$$

$$N_{25}^{F}(0) = 0 \tag{35}$$

Fission products also from ^{239}Pu

$$\frac{dN_{49}^F}{d\theta} = \frac{1}{1+\alpha_{49}} \cdot N_{49}\sigma_{49} \tag{36}$$

$$N_{49}^F(0) = 0 \tag{37}$$

And from ^{241}Pu

$$\frac{dN_{41}^F}{d\theta} = \frac{1}{1+\alpha_{41}} \cdot N_{41}\sigma_{41} \tag{38}$$

$$N_{41}^F(0) = 0 \tag{39}$$

The Pu solutions can be checked by an overall neutron balance

$$\alpha_{49} N_{49}^{F} = N_{40} + N_{41} + N_{42} + N_{41}^{F} \tag{40}$$

Burnup can be computed as a function of the neutron balance

Fission energy is about 200 MeV per U-235 atom fissioned

About 9.5×10^5 MWD/MTU

$$B = 9.5 \times 10^5 \cdot w \tag{41}$$

$$w \equiv \frac{235 \cdot N_{25}^F + 238 \cdot N_{28}^F + 239 \cdot N_{49}^F + 241 \cdot N_{41}^F}{235 \cdot N_{25}^0 + 238 \cdot N_{28}^0} \tag{42}$$

Pu accumulation can be plotted as a function of burnup

U-238 continuously depletes during operation

$$N_{28}(\theta) = N_{28}^0 - N_{28}^0\sigma_{28}\theta - \kappa_{25}(1+\alpha_{28})N_{25}^F - \kappa_{49}(1+\alpha_{49})N_{49}^F - N_{28}^F \quad (43)$$

Initial amount of atoms U-238

Absorption of thermal neutrons

Absorption of resonance,fast neutrons from U-235 fission

Absorption of resonance,fast neutrons from Pu-239 fission

Loss due to fast fission of U-238

The overall neutron balance can be checked

$$N_{28} = N_{28}^{0} - N_{28}^{F} - N_{49} - N_{49}^{F} - N_{40} - N_{41} - N_{41}^{F} - N_{42} \quad (44)$$

Fisson product production

Fission product atoms can be determined from the number of fissions

$$N_i(t) = Y_{25}^i N_{25}^F + Y_{49}^i N_{49}^F \tag{45}$$

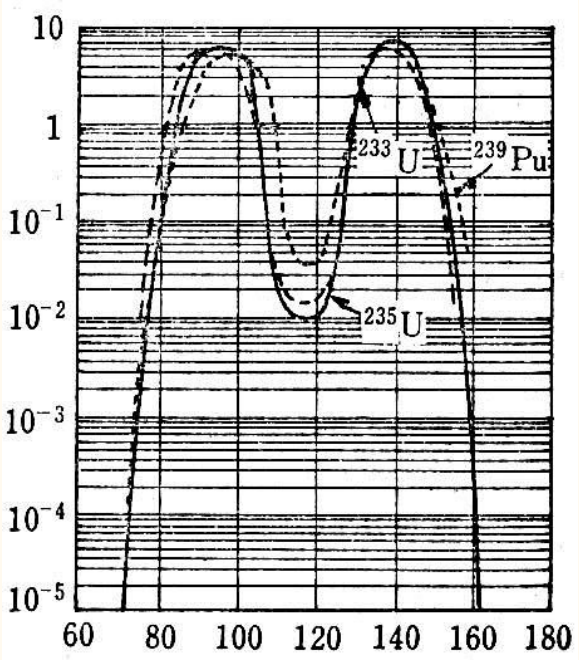
$N_i(t)$ – number of i fission product atoms

N_m^F – number of m atoms fissioned

Y_m^i cumulative yield of i fission product atom by thermal fission of m

Cumulative fission is the fraction of fissions that directly yield the isotope and its precursors

Sum of the direct yields of the nuclide and decay precursors


10
1
10^{-1}
10^{-2}
10^{-3}
10^{-4}
10^{-5}
60
80
100
120
140
160
180
^{233}U
^{239}Pu
^{235}U

The fission product production equations can be modified

$$\frac{N_{25}^F}{dt} = N_{25}\sigma_{25}^{fission}\overline{\phi} \tag{46}$$

$$\frac{N_{49}^F}{dt} = N_{49}\sigma_{49}^{fission}\overline{\phi} \tag{47}$$

Neglect absorption in fission products and precursors

For $t > T$, no fission

Individual fission product atom production can be obtained

$$\frac{dN_i}{dt} = Y_{25}^i N_{25} \sigma_{25}^f \overline{\phi} + Y_{49}^i N_{49} \sigma_{49}^f \overline{\phi} - \lambda_i N_i \qquad (48)$$
$$t < T$$

$$\frac{dN_i}{dt} = -\lambda_i N_i \qquad (49)$$
$$t > T$$

With post irradiation cooling

Cooling time – C

$$N_i(T+C) = e^{-\lambda_i C} \int_0^T e^{-\lambda_i (T-t)} (Y_{25}^i N_{25} \sigma_{25}^f \overline{\phi} + Y_{49}^i N_{49} \sigma_{49}^f \overline{\phi}) dt \quad (50)$$

With long lived nuclides, which are most important anyway

$$N_i(T+C) = e^{-\lambda_i C} \int_0^T (Y_{25}^i N_{25} \sigma_{25}^f \overline{\phi} + Y_{49}^i N_{49} \sigma_{49}^f \overline{\phi}) dt \quad (51)$$

By definition

$$N_i(T+C) = e^{-\lambda_i C} [Y_{25}^i N_{25}^F(T) + Y_{49}^i N_{49}^F(T)] \quad (52)$$

Radioactivity from neutron activation

Tritium is produced in reactors by neutron reaction with Li, B, deuterium

Also can be designed to produce tritium by irradiating Li targets with thermal neutrons

$$^{6}_{3}Li +^{1}_{0} n \rightarrow^{4}_{2} He +^{3}_{1} H$$

Neutron activation of boron produces Li and He

Thermal cross section 3837 b

$$^{10}_{5}B +^{1}_{0} n \rightarrow^{7}_{3} Li +^{4}_{2} He$$

Fast spectrum

Super small cross section; negligible

$$^{10}_{5}B +^{1}_{0} n \rightarrow 2^{4}_{2}He +^{3}_{1} H$$
$$^{11}_{5}B +^{1}_{0} n \rightarrow^{9}_{4} Be +^{3}_{1} H$$

In PWRs boron is dissolved in coolant for long-term reactivity control

Concentration change occurs over short time period short compared to half-life of tritium

Boron concentrations are repeated over each cycle

Assume an average concentration

i = atoms of species i producing tritium

$$N_T \lambda_T = \sum_i N_i \sigma_i \phi (1 - e^{-\lambda_T T}) \tag{53}$$

C-14 formed in reactors from activation of nitrogen

Residual nitrogen impurity in oxide fuel

Air dissolved in coolant

$^{14}_{7}N +^{1}_{0} n \rightarrow^{14}_{6} C +^{1}_{1} H$

2 b cross section though

$^{17}_{8}O +^{1}_{0} n \rightarrow^{14}_{6} C +^{4}_{2} He$

0.03% of natural oxygen

0.2 b

$$N_C \lambda_C = \lambda_C T \sum_i N_i \sigma_i \phi \tag{54}$$