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Calculation of catalytic nuclear reactions induced by reactor neutrons and its applications

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Introduction

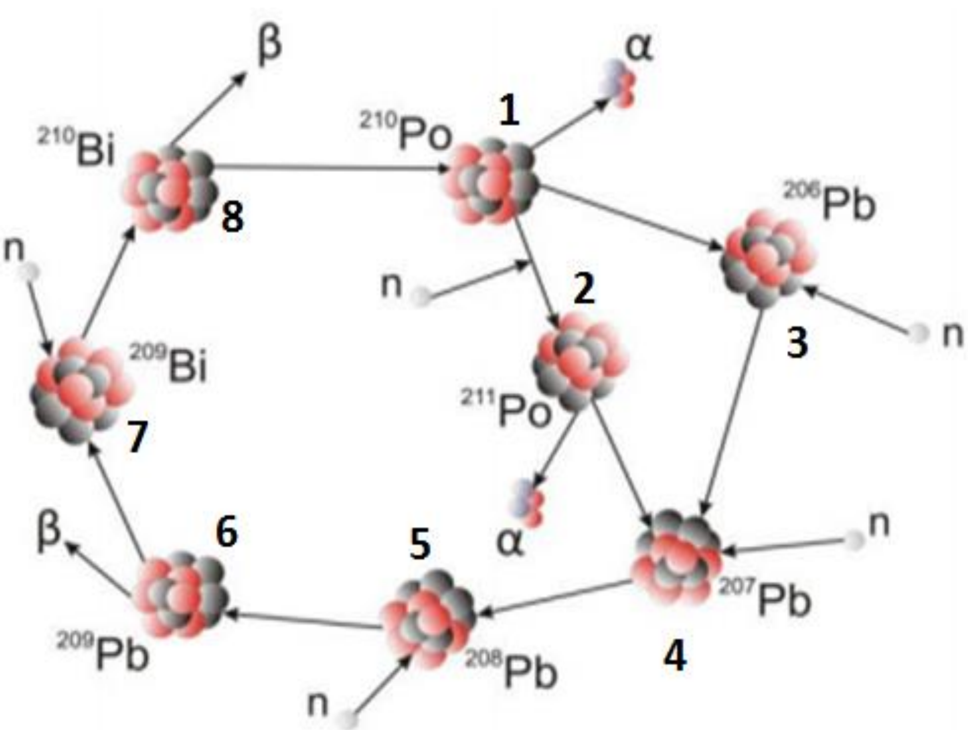
- I spent 3 month in Hokkaido University in Dr. Go Chiba's laboratory
- He is a head of reactor physics laboratory in HU

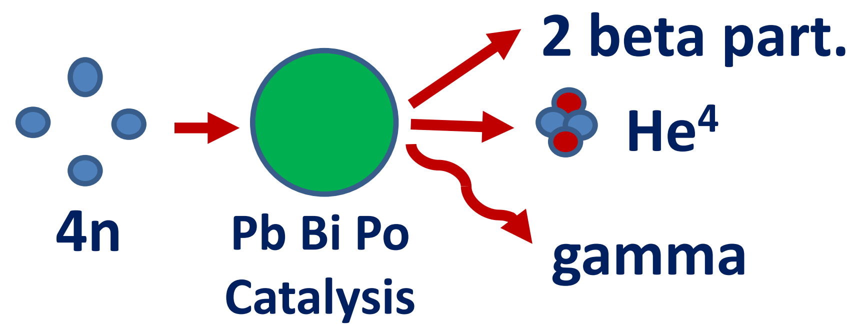
Prof. Go Chiba

Introduction

***Neutron catalysis is* a chain reaction** **based on the reaction of a four neutron capture by catalyst nucleus with the restoration of the initial nucleus**

Catalytic cycle reaction - four of neutron capture, two beta decay and alpha decay.

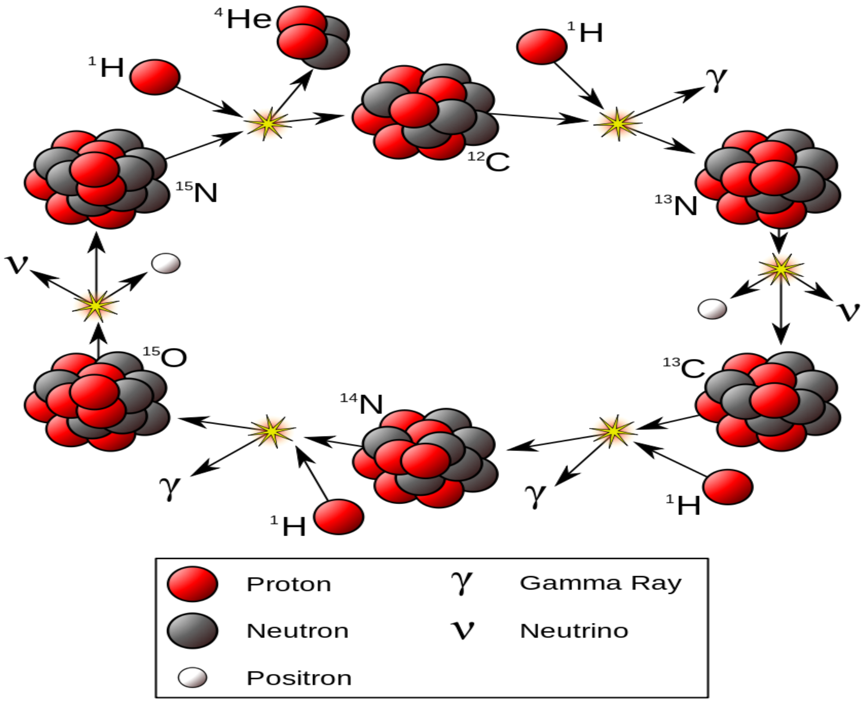




$$\Delta E = (4{*}M_n - M_{alfa} = 30\ MeV)$$

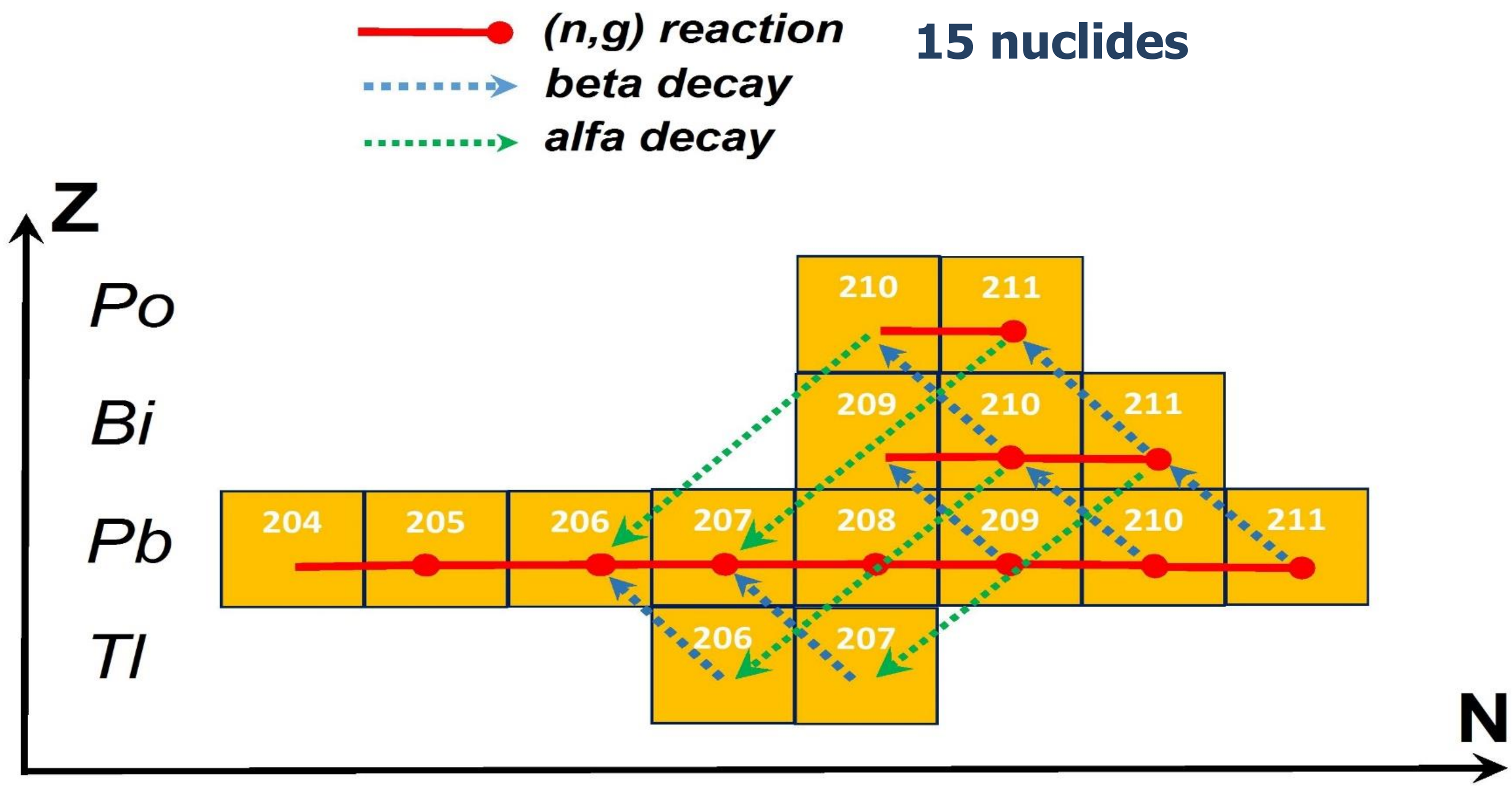
Introduction

Analog of neutron catalysis is:
CNO cycle in massive stars
CNO cycle (for carbon–nitrogen–oxygen) is fusion reactions by which stars convert hydrogen to helium



Introduction

Neutron catalysis is the catalytic chain reaction with *equilibrium state.*



Calculation

Differential equation for a number density of nuclide *i*

$$\frac{dN_i}{dt} = \underbrace{-\lambda_i N_i(t) - \sigma_i \phi N_i(t)}_{\text{Decrease of } N_i} + \underbrace{\sum_{j \neq i} \lambda_j P_{j \to i} N_j(t) + \sum_{j \neq i} \sigma_j \phi Q_{j \to i} N_j(t)}_{\text{Increase number density of } N_i}$$

$$\frac{d}{dt}\mathbf{N} = \mathbf{AN}$$

$$\mathbf{N} = (N_1, N_2 \ldots N_I)$$

A – burn-up matrix
N – Nuclide density vector
System of linear equations

$$\mathbf{N}(t) = \mathbf{N}(0)\exp(\mathbf{A}t)$$

The number density in the equilibrium state can be easily obtained if $t \to \infty$

Calculation

$$\mathbf{N}(t) = \mathbf{N}(0)\exp(\boldsymbol{A}t)$$

The matrix exponential $\exp(\boldsymbol{A}t)$ can be numerically calculated with several methods

1. **Pade approximation**
2. **CRAM (Chebyshev Rational Approximation) – developed in VTT technical research center of Finland**
3. **MMPA – developed in Hokkaido University**

$$N_{pq}(x) = \sum_{k=0}^{p} \frac{(p+q-k)!\,p!}{(p+q)!\,k!\,(p-k)!} x^k$$

$$D_{pq}(x) = \sum_{k=0}^{q} \frac{(p+q-k)!\,q!}{(p+q)!\,k!\,(q-k)!} (-x)^k$$

$$exp(\mathbf{A}\Delta t) \approx \frac{N_{pq}(\mathbf{A}\Delta t)}{D_{pq}(\mathbf{A}\Delta t)}$$

One group cross section

In burnup calculation it is convenient to use 1-group energy averaged cross sections

1-group cross section is defined from energy-dependent cross section *σ(E)* and neutron flux *φ(E)*.

G is the total number of energy groups

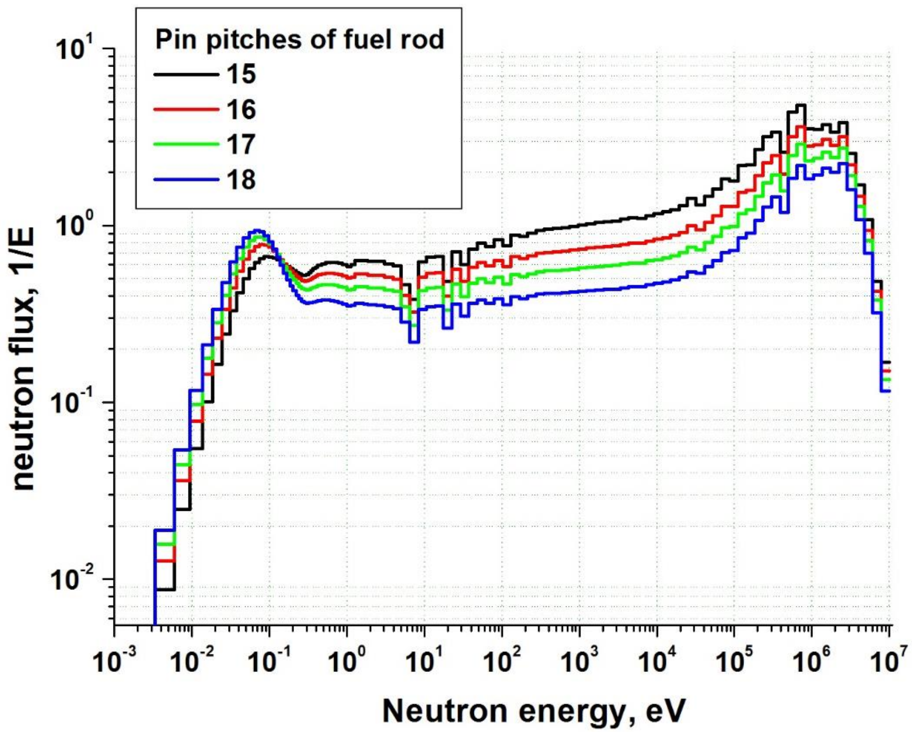
$$\tilde{\sigma} = \frac{\int \sigma(E)\varphi(E)dE}{\int \varphi(E)dE}$$

G is the total number of energy groups

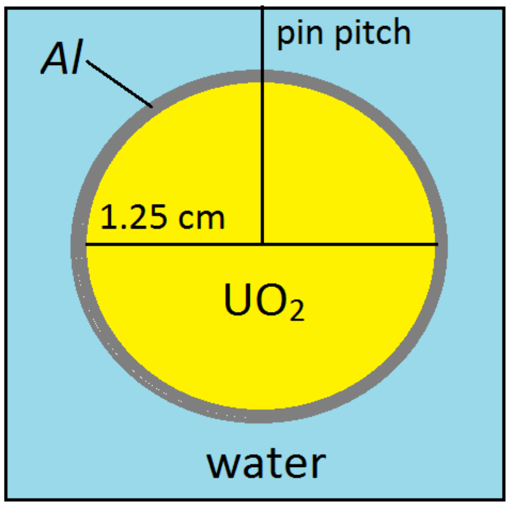
$\varphi(E)$ is energy spectra of neutron flux

$$\tilde{\sigma} = \frac{\sum_{g=1}^{G} \sigma_g \varphi_g}{\sum_{g=1}^{G} \varphi_g}$$

Neutron flux spectra at different pin pitches of the tank-typed critical assembly TCA with CBZ (TCA – is Japanese experimental reactor)



provided by Prof. Go Chiba



pin cell of TCA

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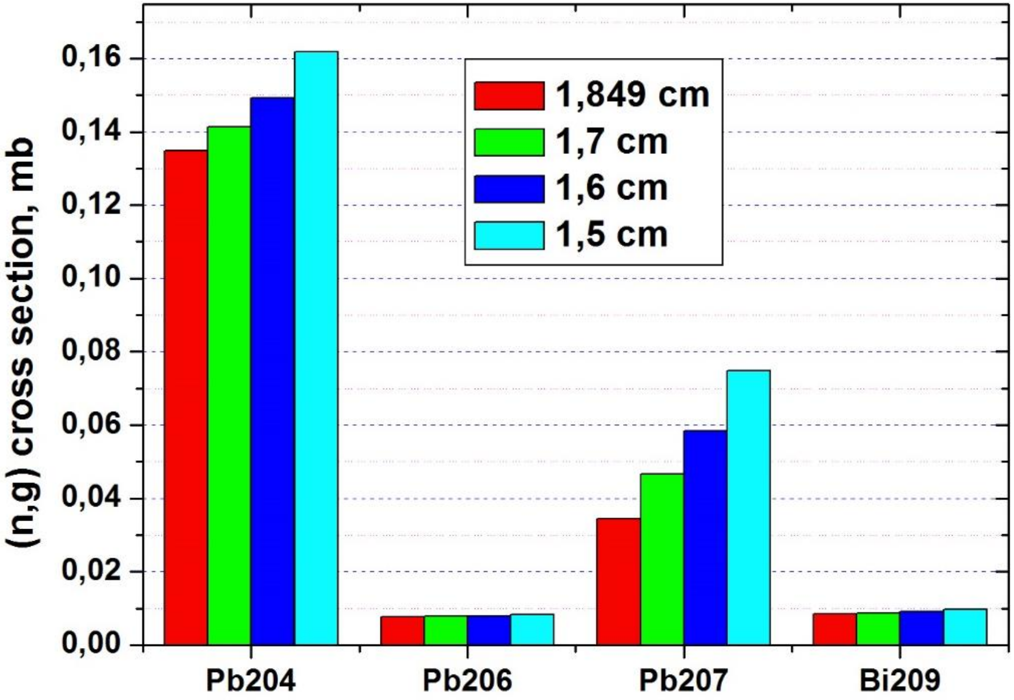
One group cross section

(n,g) cross sections were taken from JENDL-4.0 library

Energy averaged cross sections of stable nuclides

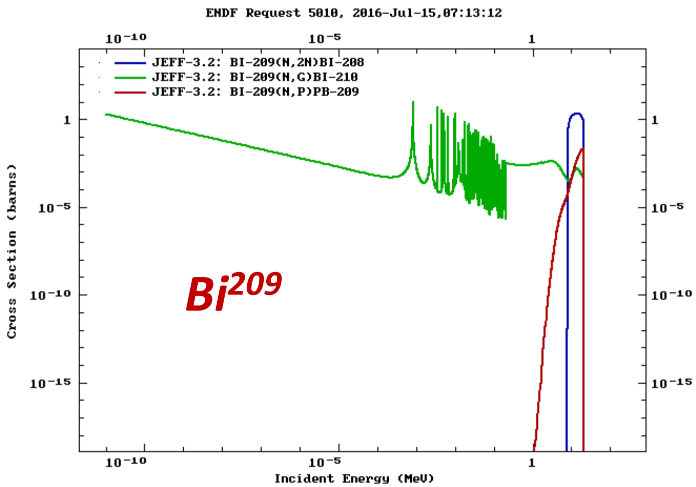


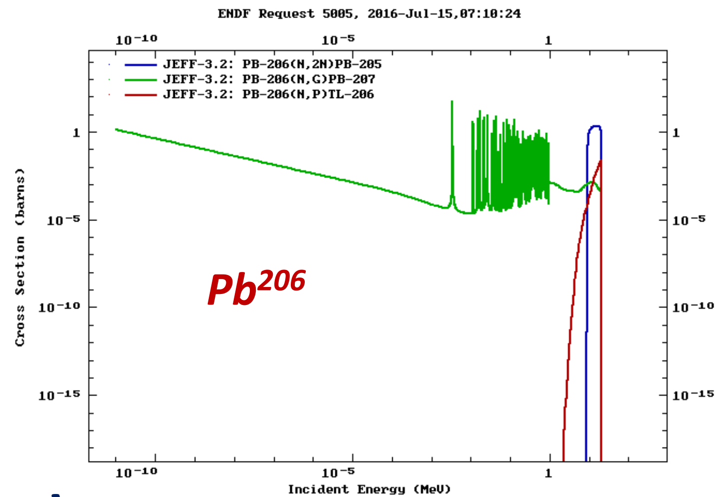
Bi^{209} Pb^{204} Pb^{206} Pb^{207} Pb^{209}

Tl^{206} Tl^{207} Pb^{205} Pb^{210} Pb^{211}

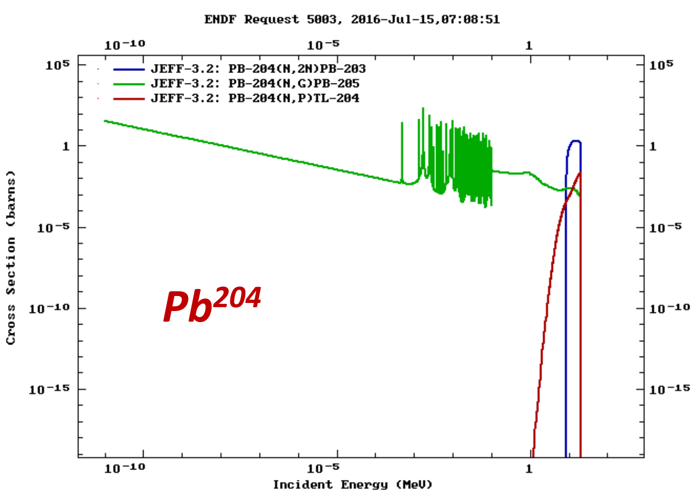
Bi^{210} Bi^{211} Po^{210} Po^{211}

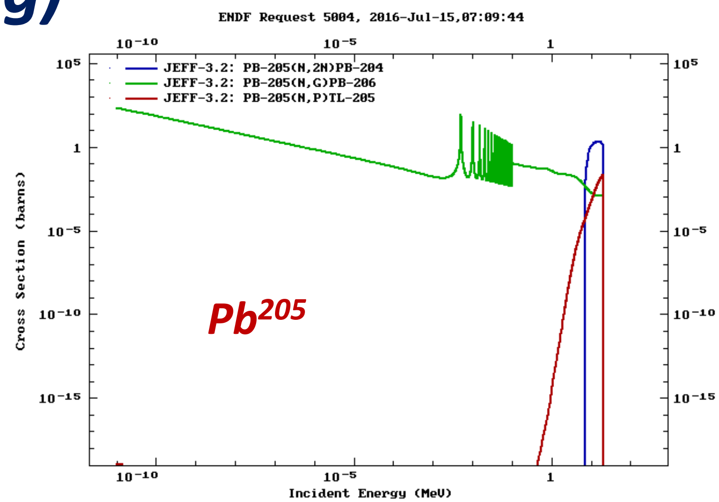
One group cross section (https://www-nds.iaea.org/exfor/endf.htm)





σ(n,g)



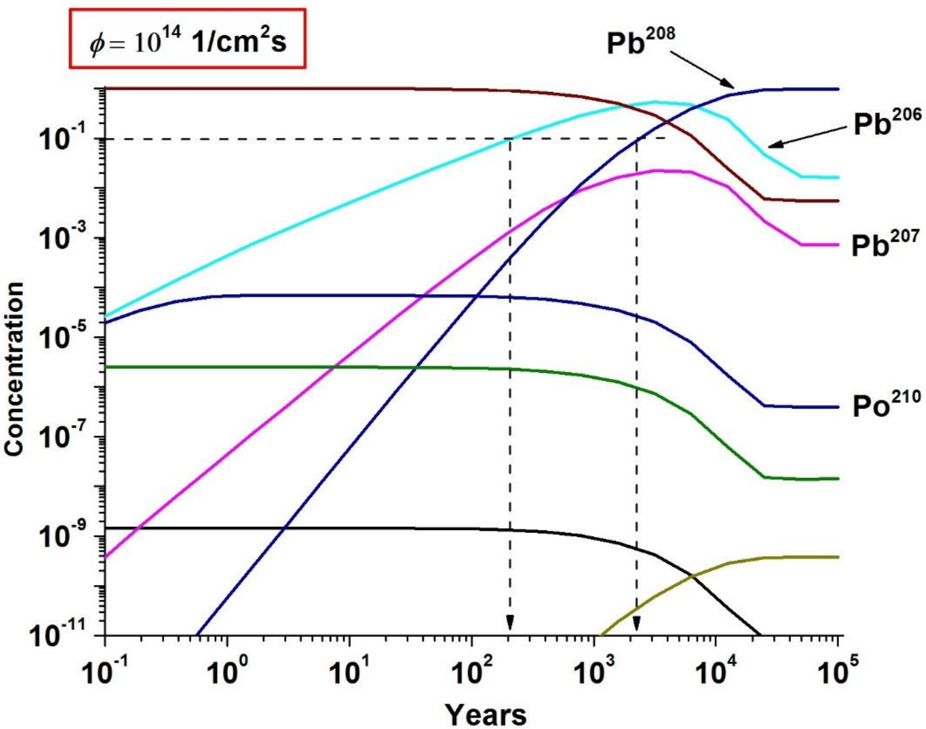


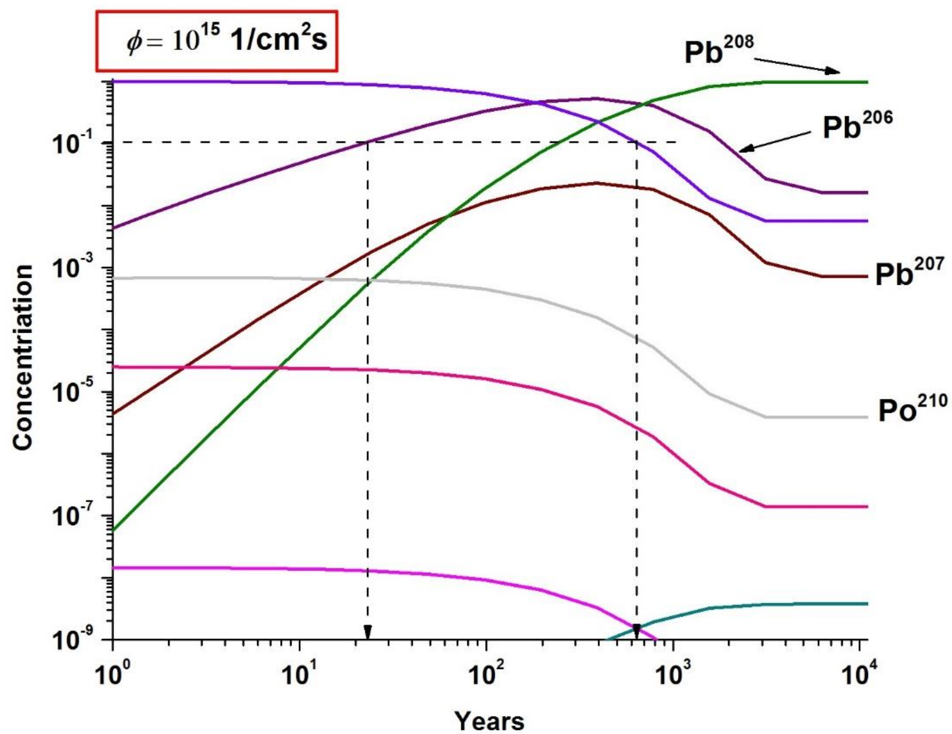
Calculation

Number density change of nuclides under reactor neutron irradiation

Initial composition is natural lead

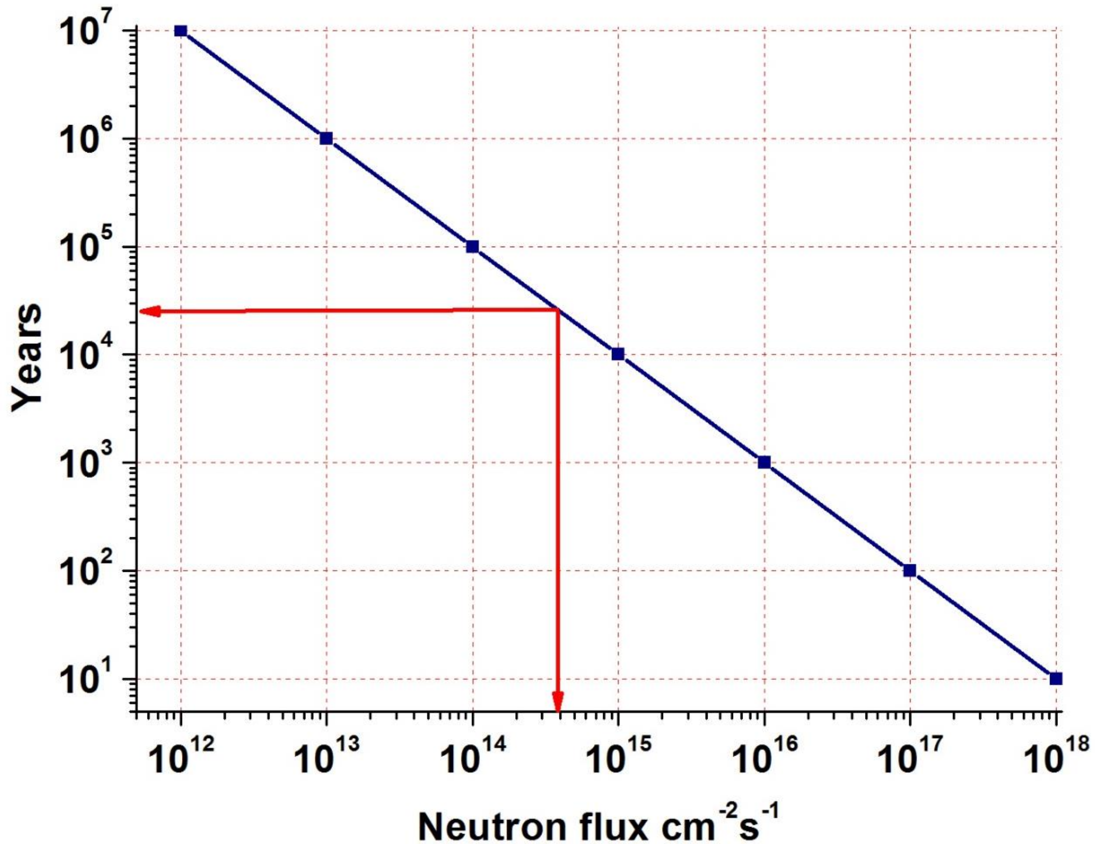
Pb^{204}	1,4%	Pb^{207}	22,1%
Pb^{206}	24,1%	Pb^{208}	52,4%





Calculation

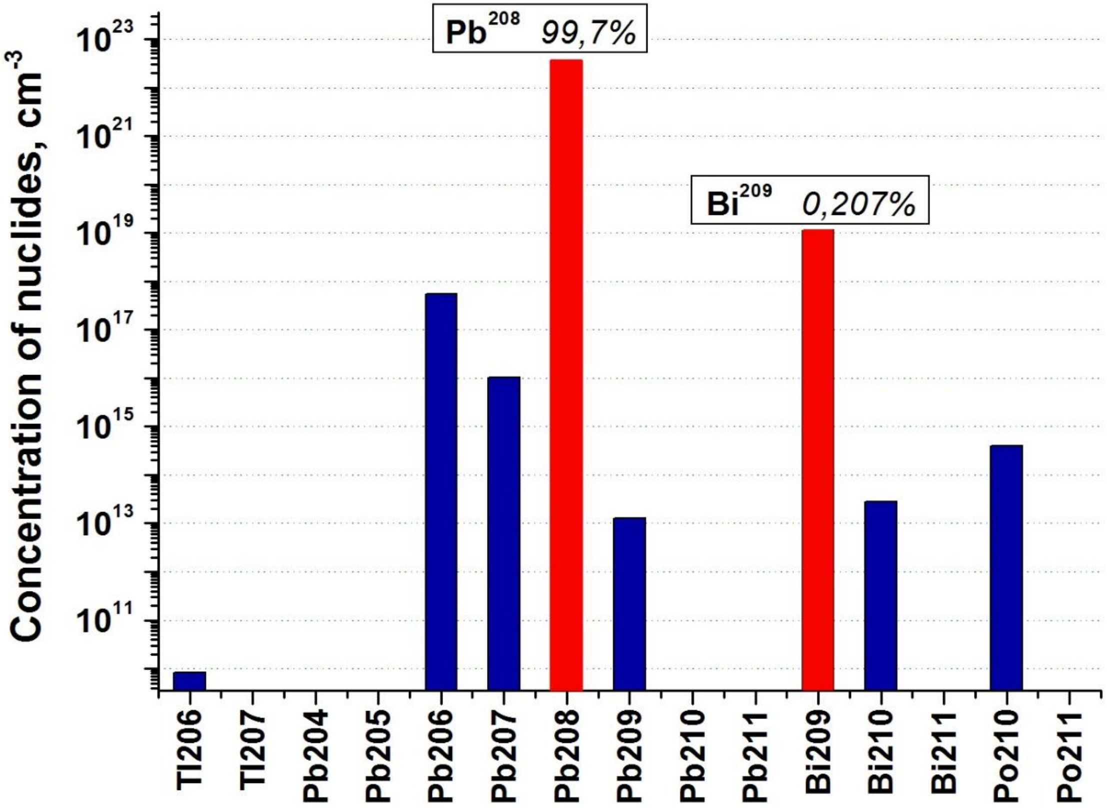
The time needed to reach the equilibrium state at different neutron fluxes



Problem is the time period of equilibrium state is too long

Calculation

Number density of nuclides at *equilibrium state.*



The problem is:

It is too difficult to enrich 15 nuclides for proper densities.

The *natural lead* composition is used for calculation because its isotope composition is closer to catalytic material composition

Application

How can catalytic material be used as reactor component?

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Main advantage of catalytic composition is:

- **It is stable under long time neutron irradiation**
- **It can produce additional energy in reactor core**

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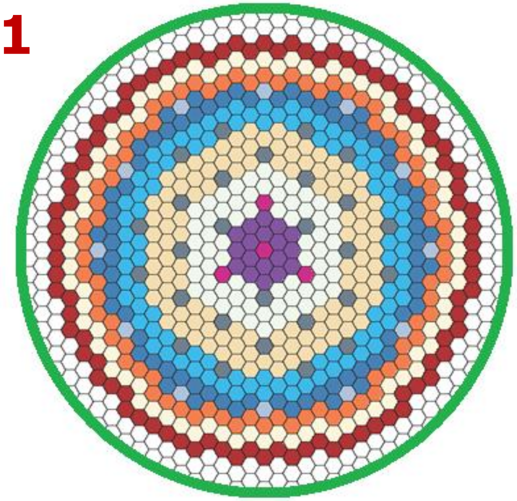
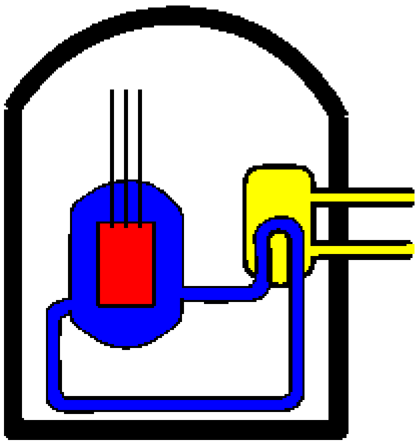
1



As neutron reflector except Be

2



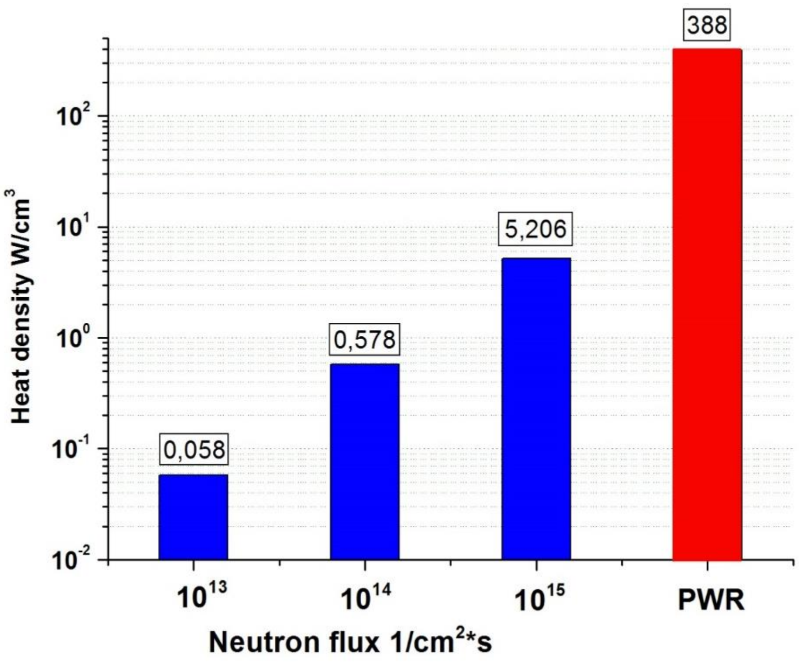
As liquid lead coolant in fast reactor

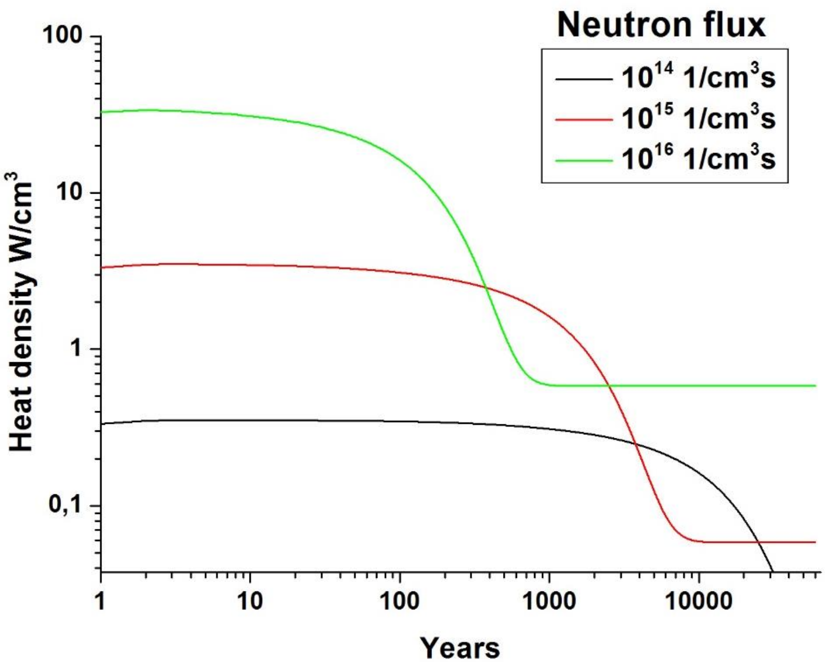
Heat density results

Total heat generated by decays

$$H(t) = \sum_i E_i \lambda_i N_i(t) + \sum_i E_i' \phi \sigma_i N_i(t)$$

E_i* is the emitted energy by a decay of nuclide *i





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Sensitivity analysis of nuclear data

Sensitivity analysis of nuclear data

$$S^i = \frac{dN_i(T)}{N_i(T)} \Big/ \frac{d\sigma}{\sigma} = \frac{\sigma}{N_i(T)} \cdot \frac{dN_i(T)}{d\sigma}$$

$$\frac{dN_i(T)}{d\sigma} = \int_0^T \left(\boldsymbol{N}^* \frac{d\boldsymbol{M}}{d\lambda} \boldsymbol{N} \right) dt$$

***M** is burnup matrix*

***N**^** *is* adjoint number density vector

T is the time after irradiation

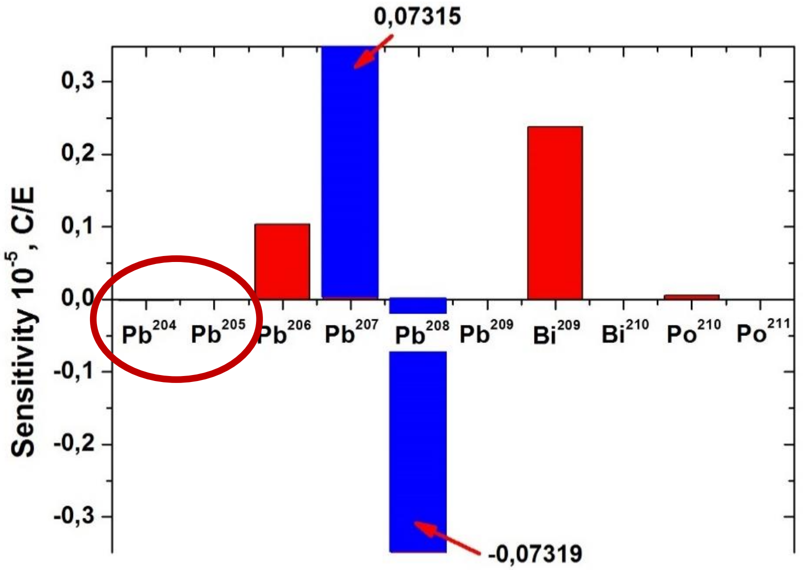
σ capture cross section (n,g)

Derivative term can be derived using ***General Perturbation Theory***. The calculation method was developed by Prof. Go Chiba in JAEA. **(Sensitivity Analysis of Fission Product Concentrations for LWR Burned Fuel)**

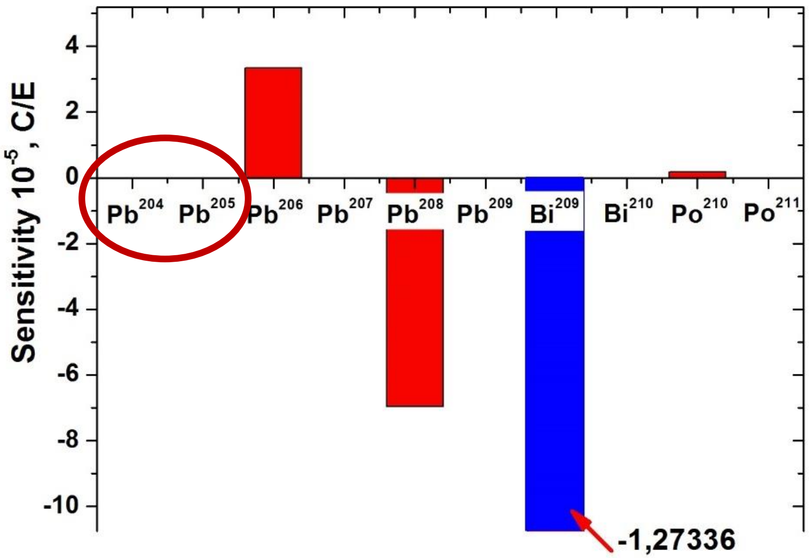
Note: The position dependence of concentrations *N(r)* and the energy dependence of cross sections *σ(E)* are ignored here.

Sensitivity analysis of nuclear data

Sensitivity analysis of nuclear data can show which nuclear data is important to results.



Sensitivities of the *Pb-208* concentration to the (n,g) cross sections



Sensitivities of the *Bi-209* concentration to the (n,g) cross sections

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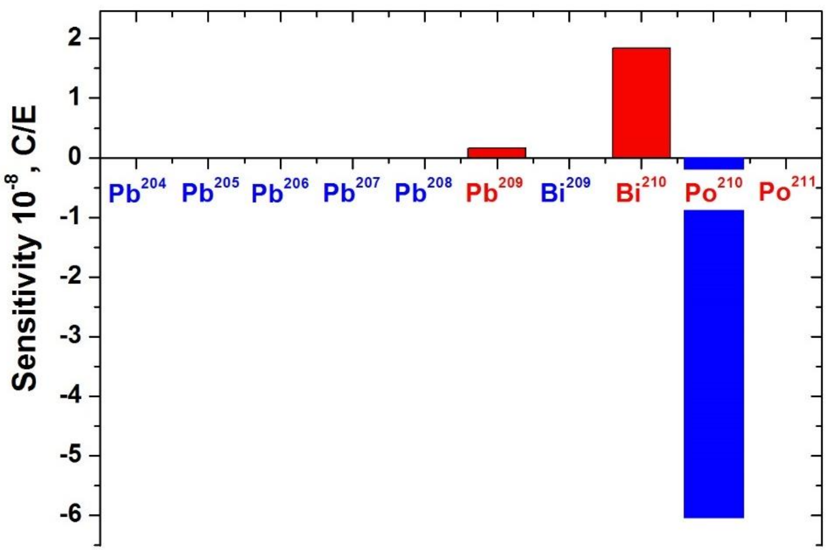
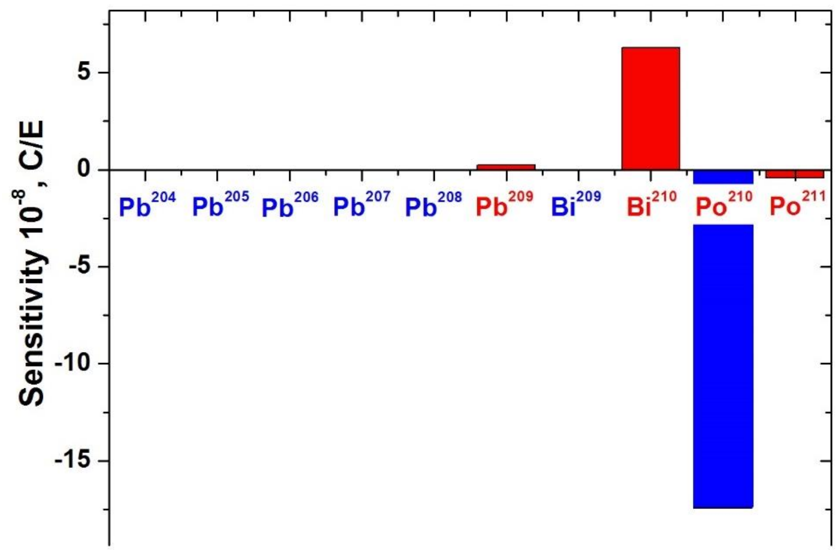
Sensitivity analysis of nuclear data

Also we can see which *decay data* is important to concentration changes of nuclides

$$S^i = \frac{dN_i(T)}{N_i(T)} \Big/ \frac{d\lambda}{\lambda} = \frac{\lambda}{N_i(T)} \cdot \int_0^T \left(\boldsymbol{N}^* \frac{d\boldsymbol{M}}{d\lambda} \boldsymbol{N} \right) dt$$



Sensitivities of the *Pb-208* concentration to decay data



Sensitivities of the *Bi-209* concentration to decay data

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Sensitivity analysis of nuclear data

Such sensitivity analysis of nuclear data is important in reactor physics, to define most important data for FPC.
The reactivity of nuclear fuels decreases through burnup since fissile nuclides contained in fuels transform into fission products.
By taking this fact into account, it becomes possible to manage criticality safety issues in nuclear facilities more rationally and more economically.

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ARTICLE

Sensitivity Analysis of Fission Product Concentrations for Light Water Reactor Burned Fuel

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The accurate prediction of fission product concentrations (FPCs) is necessary for application of the burnup credit to nuclear facilities. In order to specify important nuclear data for the accurate prediction of FPC, we extensively evaluate the sensitivities of FPC to nuclear data with the depletion perturbation theory. The target fission products are twelve important ones for the burnup credit, Mo-95, Tc-99, Rh-103,

Conclusion

Following results were obtained:

- **The nuclide density of catalytic material in equilibrium state**
- **Heat density of catalytic material at several neutron flux level**
- **Sensitivity analysis of nuclides concentrations to CS and decay data**

In the future decay heat from FPC of Molten Salt Reactor will be calculated using BurnUP equation.

THANK YOU