Fuel cycle studies on minor actinide transmutation in Generation IV fast reactors

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Introduction

Fast spectrum reactors:

- The average neutron yield per fission increases with energy
- $\sigma_f/\sigma_a$ increases with energy

Higher uranium utilization ratio and more favourable waste composition

Role of fuel cycle simulation codes:

- Assistance in strategic decisions
- Classification of waste: isotopic composition, decay heat, radiotoxicity
- Infrastructural issues: reprocessing capacity, storages, ... etc.

Spent fuel composition of the reactors has to be calculated
Determination of the spent fuel composition

The equation system which describes the evolution of the fuel composition:

\[
\frac{dN(t)}{dt} = AN(t) = (\lambda + \sigma \Phi)N(t)
\]

Difficulty: \(\sigma = \sigma(N(t))\), \(\Phi = \Phi(N(t))\)

The neutron spectrum changes with the isotopic composition therefore influencing the one-group averaged cross-sections and the integrated neutron flux.

Usual approximations:

- Burn-up tables
- Cross-section libraries for specific burn-up
- Parametrization of cross-sections
The FITXS method

Idea: parametrization of the one-group averaged cross-sections as functions of the detailed fuel composition

- **Selection of fitting parameters**: actinides with significant contribution to the reaction rates + overall quantity of fission products → 15-20 parameters
- **Preparation of cross-section database**: numerous (few thousand) core calculations with randomly sampled fuel compositions
- **Cross-section + $k_{\text{eff}}$ fitting**:

  \[
  \sigma = \sigma(N_1, \ldots, N_n) = a_0 + \sum_j a_j N_j + \sum_{j,k} a_{jk} N_j N_k
  \]

The Gas-cooled Fast Reactor (GFR) model

- 3D SCALE model of the GFR2400 core (T. Reiss)
- Homogenized inner and outer core fuel assemblies with different Pu content
- Random compositions with the following constraints:
  - Homogeneous MA and FP load
  - $\text{Pu}_{\text{inner}}/\text{Pu}_{\text{outer}} = 0.8$
  - 0-10% MA content, 10-25% Pu content, the rest is U
  - Fission products with average relative composition
  - Isotopic compositions of actinide elements are also randomly sampled
The Lead-cooled Fast Reactor (LFR) model

- Preliminary calculations for the analysis of the ELSY (European Lead-cooled SYstem) core
- 3D SCALE fuel assembly model in infinite lattice
- Random compositions with the following constraints:
  - Average fuel composition
  - 0-10% MA content, 13-22% Pu content, the rest is U
  - Fission products with average relative composition
  - Isotopic compositions of actinide elements are also randomly sampled
Fitting results

The fitted polynomials can describe the cross-sections and $k_{\text{eff}}$ with an average error well below 1%

$^{239}\text{Pu} \,(n,f)$ cross-section (GFR)
Fitting results

The fitting errors have mean values close to zero, and are related to the statistical uncertainties of the Monte Carlo transport calculations.

$^{239}$Pu (n,f) cross-section (GFR)

Fitting errors vs. statistical uncertainties
Verification of the burn-up models

Burn-up calculations using the fitted cross-sections are in excellent agreement with SCALE based burn-up results with less than 0.1% error for main isotopes.
GFR-LWR and LFR-LWR fuel cycle studies

The developed burn-up models were integrated into a fuel cycle models containing fast reactors and conventional Light Water Reactors (LWRs)
GFR and LFR closed fuel cycle results

No external Pu feed is needed in the equilibrium → GFR reaches self-breeding

The Pu content of the core increases to counterweight the decrease of fissile Pu
GFR and LFR closed fuel cycle results

The LFR also reaches breakeven breeding

Both reactors are able to breed their fuel from fertile $^{238}\text{U}$
GFR and LFR closed fuel cycle results

Both the GFR and LFR reaches an equilibrium MA content of 1% with no Cm accumulation

Every minor actinide isotope can be burned in the hard neutron spectrum of the GFR and LFR
The GFR and LFR are able to burn additional minor actinides from LWRs if more MAs are fed to the core than the equilibrium MA content.
GFR and LFR closed fuel cycle results

Minor actinide waste due to reprocessing losses (GFR – left, LFR – right)

The MA burning capabilities of the reactors highly exceed the MA waste production from partitioning losses

Nevertheless, the partitioning technology has main impact on the final waste
GFR and LFR closed fuel cycle results

Higher MA content of the GFR and LFR cores results in higher breeding gain.

This effect can be explained by the analysis of the neutron economy.
The European Pressurized Reactor (EPR) model

- Generation III Light Water Reactor
- Goal: investigation of the recycling of excess Pu produced by the FRs in Light Water Reactors
- 3D SCALE fuel assembly model in infinite lattice
- Random compositions with the following constraints:
  - Average fuel composition
  - 5-10% Pu content, 0-1% MA content, the rest is U
  - Fission product composition fitted as function of assembly burnup + Xe, Sm calculated explicitly
  - Isotopic compositions of actinide elements are also randomly sampled
GFR-EPR symbiotic fuel cycle studies

The Pu produced by the GFR is recycled in EPR MOX fuel assemblies, and the spent MOX Pu is recycled in the GFR
GFR-EPR symbiotic fuel cycle results

The GFR is able to burn the minor actinides from the spent MOX fuel, but the recycling of the excess Pu results in high MA content and lower fissile Pu ratio

Transmutational capabilities and possibly safety parameters degraded
Analysis of the neutron economies

Motivation: deeper understanding of fuel breeding and transmutation capabilities

D-value = neutron consumption / fission (M. Salvatores, 1993)

\[ D_J = \sum_{J_1} P_{J \rightarrow J_1} \{ R_{J_1} + \sum_{J_2} P_{J_1 \rightarrow J_2} \cdot [ R_{J_1} + \sum_{J_3} P_{J_1 \rightarrow J_3} \cdot (\ldots) ] \} \]

where

\[ R_J = \begin{cases} 
1 & (n, \gamma), \\
0 & \text{decay}, \\
1 - \nu & (n, f), \\
-1 & (n, 2n).
\end{cases} \]

\[ P_{J \rightarrow J_1} = \frac{\sigma_{J \rightarrow J_1} \Phi + f_{J \rightarrow J_1} \lambda_J}{\sigma_{J,a} \Phi + \lambda_J} \]

Neutron consumption of the fuel:

\[ D_{FUEL} = \sum_{J} \varepsilon_J D_J \quad \Rightarrow \quad S_{ext} - D_{FUEL} \geq (L + CM) + D_A \]

Analysis of the neutron economies

The evaluation of the D-values requires the consideration of every possible transmutational chains, making the calculation difficult

\[ D_J = \sum_{J_1} P_{J \rightarrow J_1} \{ R_{J_1} + \sum_{J_2} P_{J_1 \rightarrow J_2} \cdot [ R_{J_2} + \sum_{J_3} P_{J_1 \rightarrow J_3} \cdot (...) ] \} \]
Analysis of the neutron economies

Based on the theory of Markov-chains we derived a closed formula for the neutron consumption / production

Transition probability matrix:

\[ P_{xy}^{(t_0,t_1)} = P_{xy}^{(t)} = (P')_{xy}, \quad P^{(0)} = I \]

The neutron production can be expressed as one step plus the neutron production of the next nuclide in the chain:

\[ n_x^D = \begin{cases} \tilde{\nu}_{xy_1} + n_{xy_1}^D, & P_{xy_1} \text{ probability} \\ \tilde{\nu}_{xy_2} + n_{xy_2}^D, & P_{xy_2} \text{ probability} \\ \vdots & \vdots \end{cases} \]

\[ P_{xy} = \frac{\sigma_{xy} \Phi + f_{xy} \lambda_x}{\sigma_{x,a} \Phi + \lambda_x} \]

\[ \tilde{P} \hat{n}^D = \nu^D \]

\[ \tilde{P}_{xx} = 1, \quad \tilde{P}_{xy} = -P_{xy}, \quad \nu_x^D = P_{xf} \tilde{\nu}_x + \sum_y P_{xy} \tilde{\nu}_{xy} \]
Analysis of the neutron economies

Neutron production of actinide isotopes in the two examined fast reactor types

<table>
<thead>
<tr>
<th>Isotope</th>
<th>Neutron production / fission</th>
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<tbody>
<tr>
<td></td>
<td>GFR</td>
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<tr>
<td>$^{235}\text{U}$</td>
<td>0.90</td>
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<tr>
<td>$^{238}\text{U}$</td>
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<td>$^{237}\text{Np}$</td>
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<tr>
<td>$^{245}\text{Cm}$</td>
<td>2.37</td>
</tr>
</tbody>
</table>
Analysis of the neutron economies

Neutron production of actinide isotopes with respect to number of transitions

MA isotopes are fertile if not fissile in the hard neutron spectrum of the GFR

High MA content increases the total neutron production therefore improves breeding capabilities
Analysis of the neutron economies

Time-dependent neutron productions based on continuous-time Markov-chain

The time-dependency of the average neutron productions also shows the fertile nature of otherwise absorbing minor actinides.
The SITON code

The method has been also implemented in the SITON nuclear fuel cycle simulation code developed in the Hungarian National Academy of Sciences.

SITON v2.0 is a dynamic, discrete facilities/discrete materials and also discrete events fuel cycle simulation code.

Also, SITON v2.0 is involved in the Benchmark Study on Nuclear Fuel Cycle Transition Scenarios Analysis Codes organized by the OECD Working Party on Scientific Issues of the Fuel Cycle (WPFC)

Summary

- GFR, LFR and EPR MOX burn-up models were developed with the FITXS method
- GFR-LWR and LFR-LWR fuel cycles were investigated with and without MOX recycling in EPR MOX fuel assemblies
- Both the GFR and the LFR is able to operate with Pu from LWR spent fuel
- An equilibrium MA content of 1% is reached during closed cycle operation, and both FRs are capable of burning additional MAs from LWR spent fuel
- MOX Pu recycling in EPRs results in significant deterioration of the Pu isotopic composition, as well as higher equilibrium MA content
- Higher MA content results in higher breeding gain due to fertile nature of MA isotopes in fast neutron spectrum
- Although MOX recycling increases uranium utilization ratio compared to FR-LWR fuel cycle, higher MA content and worse Pu composition suggest that recycling of excess Pu would be more favourable in FRs
Thank you for your attention!