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Preliminary Analysis on the Chemical Forms of Fission Products in Nuclear Fuel via Coupling of Serpent 2 and GEMS

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1. Introduction

The best-estimate analysis requires chemical forms of the released fission products from a containment of a nuclear power plant since the radiation dose depends on their chemical forms. Numerous works have been performed on conventional light-water reactors [1, 2] and it has been found that the chemical forms of the fission products are affected not only by an accident sequence but also by design of the reactor core, e.g., type of the control rods, etc.

The challenge is further compounded by innovative SMR concepts such as the i-SMR, which employs two types of control rods in a single reactor core, i.e., Ag-In-Cd control rods are used for regulating bank, and B4C control rods are used for shutdown bank [3]. Because no commercial reactor has ever operated with the aforementioned control rod configuration, it is necessary to analyze the impact of such configurations on the chemical speciation of fission products (FPs) during a severe accident.

In this work, we have performed a preliminary analysis on chemical form of fission of fission products in a nuclear fuel via coupling of reactor analysis code based on Monte Carlo method, Serpent 2 [4] and chemical equilibrium calculation code, GEMS [5].

2. Coupling Scheme of Serpent 2 & GEMS

2.1 Numerical Methods in GEMS

In GEMS chemical equilibrium of any multicomponent system is obtained by casting the problem as a convex minimization of the total Gibbs free energy subject to elemental mass-balance constraints. The resulting Karush-Kuhn-Tucker (KKT) system is solved with the IPM-3 interior-point algorithm, which iteratively updates species mole number (primal variables) and independent-component chemical potentials (dual variables) while enforcing strictly positive amounts. Non-ideal phase behavior is introduced through activity-coefficient models (ideal gas, Pitzer, NRTL, Redlich-Kister, etc.), whose first derivatives are embedded in the Jacobian to maintain quadratic-like convergence.

Convergence is obtained when the Dikin error falls below the user-set tolerance and all elemental residuals satisfy the prescribed mass-balance threshold.

Under this framework the equilibrium state is governed by the following KKT relations given below.

Chemical equilibrium is reached when the first derivative of the total Gibbs free energy with respect to the dependent component mol vector $\vec{n}^{(i)}$ vanishes,

$$G'\left(\vec{n}^{(i)}\right)=0, \tag{1}$$

with the Gibbs energy itself written as

$$G\left(\vec{n}^{(i)}\right)=\sum_{j\in L}\nu_j\cdot n_j^{(i)},$$
$$\nu_j=\frac{\mu_j}{R\cdot T}=\frac{\mu_j^0}{R\cdot T}+\ln x_j+\ln\gamma_j+\Xi, \tag{2}$$

where in Eq. (2) L is the index set of all dependent components species in the system, $j\in L$ labels those components, $n_j^{(i)}$ is the mole amount of the j-th dependent component, and ν_j is normalized chemical potential. μ_j is the true chemical potential, R the universal gas constant, T the absolute temperature, x_j the mole fraction type concentration of component j in its phase, γ_j its activity coefficient obtained from chosen non-ideal phase model, and Ξ an optional phase-asymmetry term. The reference quantity μ_j^0 denotes the standard state molar Gibbs energy of component j corrected to the current T and pressure.

The speciation vector $\vec{n}^{(i)}$ must obey elemental mass conservation,

$$A\cdot\vec{n}^{(i)}=\vec{n}_{tot},\quad \vec{n}^{(i)}\geq 0, \tag{3}$$

where in Eq. (3) A is the stoichiometric matrix and $\vec{n}_{tot}$ is the vector of moles of independent (elemental) components supplied as input. To enforce Eq. (3), GEMS introduces a Lagrange-multiplier vector $\vec{u}$, which leads to the Karush-Kuhn-Tucker complementarity conditions.

$$\vec{v} - A^T\vec{u} \geq 0, \vec{n}^{(i)} \cdot \left(\vec{v} - A^T\vec{u}\right) = 0, \quad (4)$$

Here in Eq. (4) $\vec{v}$ collects the normalized chemical potentials of all dependent components. It's j -th element is v_j . Likewise, $A^T\vec{u}$ denotes the vector formed by stoichiometric linear combination of the independent-component chemical potentials for each species.

Eqs. (1)~(4) constitute a nonlinear system that GEMS solves iteratively with an interior-point algorithm (IPM-3). At each iteration the method updates $\vec{n}^{(i)}$ while keeping the inequalities in Eq. (3) and (4) strictly satisfied, thereby guaranteeing positive mole numbers and convergence to the thermodynamically stable equilibrium.

2.2 Conversion of Serpent 2 Output to GEMS input

Serpent 2 depletion calculations produce isotopic inventories of the fuel for burnup steps specified by a user.

To make this inventory compatible with GEMS, the isotopic inventories are post-processed in three stages. First, isotopes with the same atomic numbers are collected under a single element, and their number densities are summed, because chemical behaviors of fission products determined by their atomic numbers not by mass numbers. Second, the atomic densities are converted into the elemental model quantities which are compatible with GEMS input format. Finally, the resulting vector of elemental mole quantities is inserted into the composition block of the GEMS input file. The coupling scheme between Serpent 2 and GEMS processes the Serpent 2 depletion results to the thermodynamic-equilibrium analysis stage without altering the underlying burn-up information.

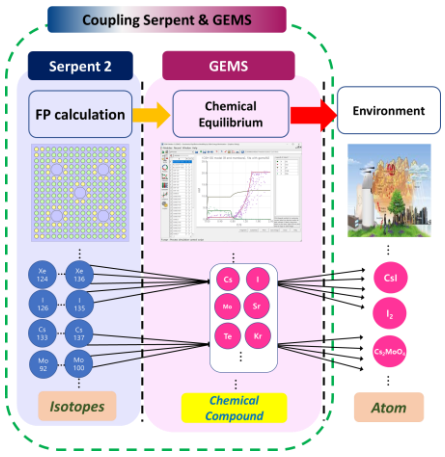

Fig. 1. Coupling scheme of Serpent 2 and GEMS to obtain chemical configuration of the fission products.

Using thermodynamic data from HERACLES-TDB [6], GEMS computes the equilibrium speciation and phase partitioning of the fission products represented by the Serpent 2 inventories, thereby enabling consistent evaluation of their chemical behavior for advanced fuel-cycle and safety studies.

With the information obtained from Serpent 2, chemical equilibrium calculations are performed by GEMS. In the calculations, HERACLES-TDB is used as database for thermochemical reactions. HERACLES-TDB is a project-specific database developed at the Paul Scherrer Institute under the HERACLES program (Head-End Reprocessing studies by thermAl and thermoChemicaL treatment of fuElS).

The HERACLES-TDB provides internally consistent Gibbs energy functions for several hundred solid, liquid, and gaseous compounds that include uranium, thorium, transuranic elements, and all major fission products. The dataset has been benchmarked against high temperature fission product release experiments conducted in the FP7-ACSEPT project, confirming its reliability for predicting phase formation, chemical speciation, and volatile fission product behavior in multicomponent nuclear fuel systems up to approximately 2,500 K at ambient pressure.

3. Numerical Results

Based on the methodology described above, a representative fuel assembly from a conventional light water reactor (LWR) is modeled at high burnup using Serpent 2. The computation conditions used in Serpent 2 are summarized in Table 1, and the fuel assembly model employed for the depletion calculation is shown in Fig. 2. The change in infinite multiplication factor k_{inf} over the depletion history is presented in Fig. 3. The resulting elemental inventory at 52.25 MWd/kgU is then used to perform equilibrium chemical speciation calculations in GEMS, with HERACLES-TDB.

Table 1. Computation conditions in Serpent 2

Parameter		Value
Computer code		Serpent 2
Cross section library		Continuous energy ENDF/B-VII libraries
# of histories		100,000
# of cycles	Inactive	500
	Active	1,000

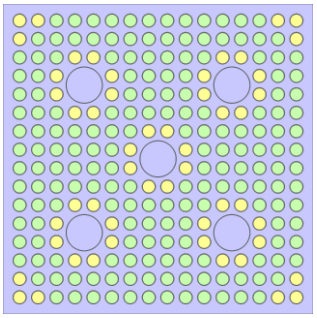

Fig. 2. Fuel assembly modeling used in depletion calculation.

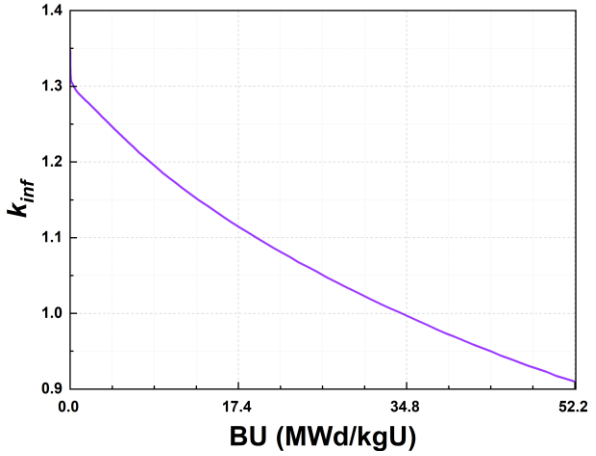

Fig. 3. Change in multiplication factor k_{inf} from Serpent 2.

Fig. 4 presents the distribution of dependent components in the gaseous phase obtained by GEMS, showing the overall fraction of species existing as gas at equilibrium. These results indicate fission products with high importance in the source term analysis tend to partition into the gas phase under high temperature conditions, based on their thermodynamically preferred chemical forms.

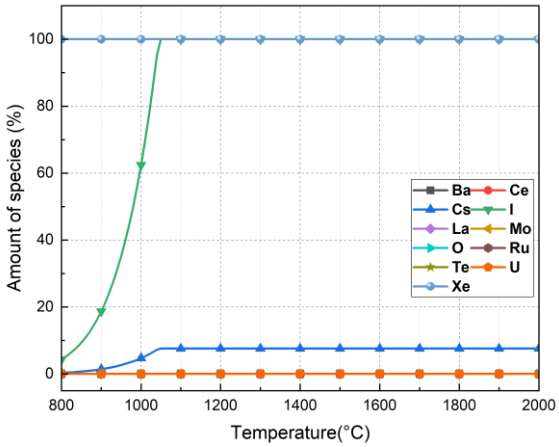

Fig. 4. Fraction of chemical compounds derived from fission products in the gas phase at various temperatures.

As shown in Fig. 4, majority of the iodine compounds undergo a phase transition to the gaseous state at elevated temperatures, indicating that their equilibrium speciation is predominantly governed by the gas phase under the conditions. For cesium, approximately 10% of the total species undergo a phase transition to the gaseous state at elevated temperatures. The noble gases persist exclusively in the gaseous phase over the entire temperature range considered, consistent with their inherent chemical inertness. In contrast, metallic elements (e.g., molybdenum) appear to persist in the condensed phase over the entire temperature range considered; only very small fractions undergo phase transition in the analysis.

Figs. 5 and 6 respectively illustrate the distribution of iodine and cesium compounds over the temperature range considered.

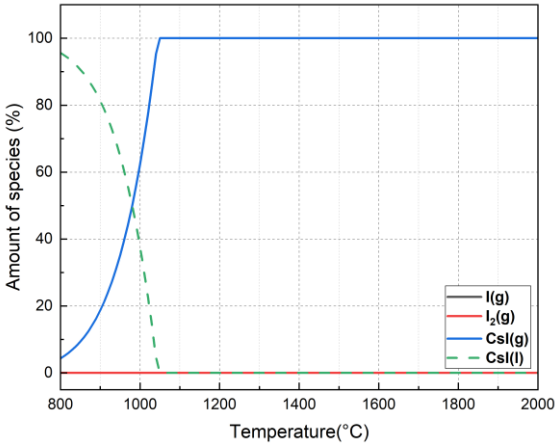

Fig. 5. Fraction of iodine compounds at various temperatures.

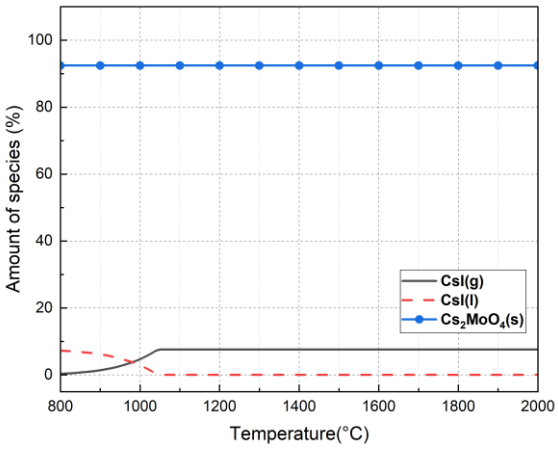

Fig. 6. Fraction of cesium compounds at various temperatures.

For the iodine species, the dominant species is CsI and it persists in liquid phase at the temperature lower than 1,000℃. It undergoes a phase transition to the gaseous state as the temperature increases. It persists exclusively in the gaseous phase over the temperatures higher than 1,050℃. Elemental iodine species, such as I and I_2 are shown to be in very small fractions. The chemical compositions of the iodine species are listed in Table 2 for various temperatures. As shown in Table 2, the dominant fraction on CsI is similar to the recommendation in NUREG-1465 [7, 8]. It should be noted that the chemical form of iodine for PWRs is assumed to be 95% CsI in NUREG-1465 [7, 8].

Table 2. Fractional composition of iodine species

Temperature [℃]	I(g)	I_2(g)	CsI(g)	CsI(l)
800	1.21E-17	2.82E-17	4.34	95.7
850	1.47E-16	3.03E-16	9.35	90.7
900	1.43E-15	2.64E-15	18.7	81.3
1000	8.00E-14	1.20E-13	62.4	37.6
1100	1.44E-12	1.05E-12	100	0
1200	1.17E-11	3.05E-12	100	0

For cesium compounds, the dominant species is Cs_2MoO_4, it is approximately 92% over the entire temperature range considered.

The aforementioned results obtained from the coupling of Serpent 2 and GEMS indicate that Cs_2MoO_4 is dominant cesium bearing compound and are consistent with observations from the Phebus experiments and MELCOR best-practice [9].

4. Conclusions

In this study presents a framework that couples Serpent 2 depletion data with GEMS equilibrium calculations to characterize fission product speciation. Serpent 2 isotopic inventories are converted into elemental inputs for GEMS, linking reactor core depletion to thermodynamic behavior of the fission products. The results of preliminary analyses show good agreement with recommendation from previous works such as NUREG-1465, Phebus FP, MELCOR best-practice, and etc.

As future work, the coupling scheme will be applied to water-cooled SMRs in which two types of control rods are employed within a single reactor core: Ag-In-Cd control rods for regulating bank and B4C control rods for shutdown bank, in order to determine the chemical behaviors of the fission products.

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