초고온가스로 핵분열생성물 분석용 GAMMA-FP 코드의 화학반응 모델 구현 및 검증

Implementation and Verification of the Chemical Reaction Models in the GAMMA-FP Fission Products Analysis Tool for VHTR Applications

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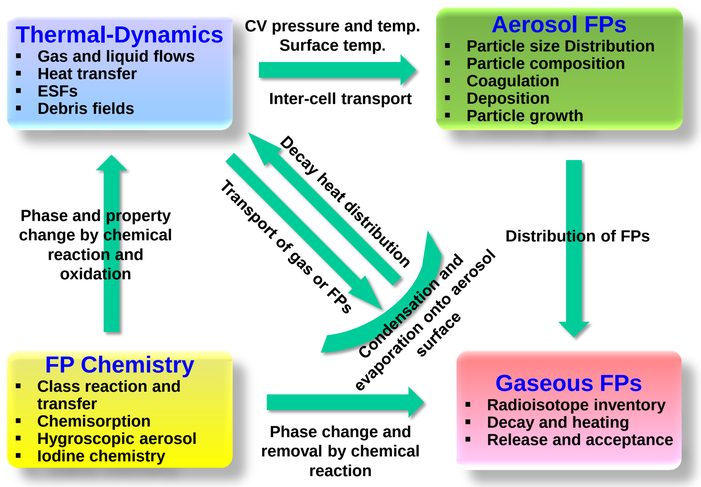
Introduction

- The ultimate goal is to develop a fission products (FP) analysis software(GAMMA-FP or RASTEV) for VHTR's.
- A goal of this step is to develop the chemical reaction module of the fission product analysis software by thermochemical equilibrium.
- Another goal is to verify and validate the developed chemical reaction models.
- The developed FP chemical module has been implemented based on the thermochemical and physical data of 288 FP species and 26 elements.

Developing Status

- **Thermal-Dynamics Module**
  - GAMMA+(GAseous Multi-component Mixture Analysis for the VHTR plus)
  - Gas and Liquid flows with heat transfer
- **Gaseous FP Module**
  - 1-D FP Transport and Plate-out Model
    - ✓ Fractional Step Method
    - ✓ Embedded Runge-Kutta Method
    - ✓ Implicit, Upwind Scheme in Staggered Mesh Layout
  - Sorption Model : GA & JAEA Sorption Model
- **Aerosol FP Module**
  - 0-D Multi-component Aerosol analysis : MAEROS Model
    - ✓ Coagulation
    - ✓ Deposition
    - ✓ Condensation /Evaporation
  - 1-D Transport Model
    - ✓ Inter-volume aerosol transport (MELCOR)
- **FP Chemistry Module**
  - Under Construction



Modules of the FP Analysis Code

Equilibrium Vapor Pressure

- **RASTEV code (Relvp) :**
  - Equilibrium vapor pressure of pure species *i* [atm]

$$p'(i) = \exp\left(-\frac{\Delta G}{RT}\right) = \exp\left(\frac{G_{gas} - G_{con}}{RT}\right)$$

ΔG = Change of the Gibbs free energy [J/kg-mol]
R = Gas constant = 8314.51 J/kg-mol/K

Here, $G = a + bT + cT^2 + dT^3$

- Constants *a*, *b*, *c*, & *d* are provided for 288 FP species

- **Verification : Comparison with empirical correlations**

1) Blackburn and Johnson (1988)
   - Vapor pressure for CsOH monomer
   - Temp. range: 681 ~ 772 K

$$\log_{10}\left(P_m[atm]\right) = -\frac{7217}{T[K]} + 5.640$$

2) Konings and Cordfunke (1988)
   - Total vapor pressure above liquid CsOH
   - Temp. range: 676 ~ 976 K

$$\log_{10}\left(P_m[atm] + 2P_d[atm]\right) = -\frac{(6414 \pm 148)}{T[K]} + (4.763 \pm 0.180)$$

CsOH 평형 기체 압력

Frozen Chemistry Model

- **Assumptions**
  - Below the threshold temperature:
  - No chemical reaction occurs,
  - Only simple phase change occurs
- **Thermochemical equilibrium equations**
  - Concentration conservation

$$C_T(i) = C_G(i) + C_C(i)$$

$$C_G(i) = \frac{p(i)}{RT} = \frac{p'(i)x(i)}{RT} = \frac{x(i)}{RT}\exp\left(-\frac{\Delta G}{RT}\right)$$

Here, mole fraction: $x(i) = \dfrac{C_C(i)}{\sum_{j \in C_i} C_C(j)}$

- **Newton-Raphson solving procedure**
  - Residuals

$$R_i = C_T(i) - C_C(i) - \frac{p'(i)}{RT}\frac{C_C(i)}{\sum_{j \in C_i} C_C(j)}$$

  - Jacobian matrix:

$$J_{ij} = \frac{\partial R_i}{\partial C_C(j)}$$

  - Correction equation:

$$R_i + \sum_j J_{ij}\delta\left[C_C(j)\right] = 0$$

with

$\delta\left[C_C(j)\right]$ = correction to condense-phase concentration

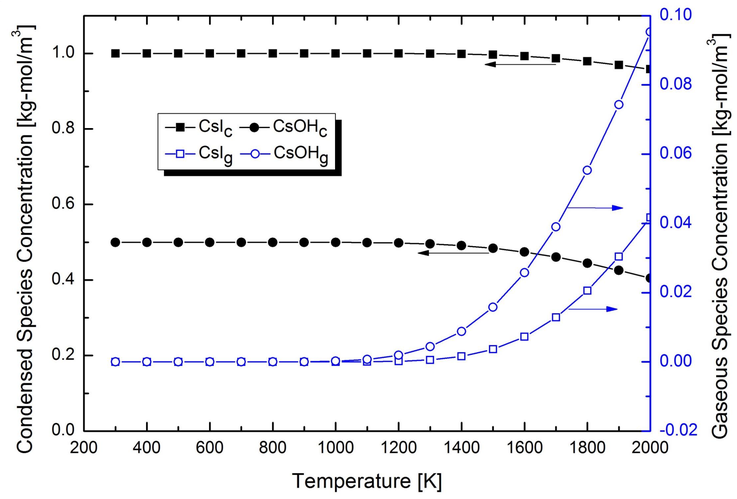
검증 계산

❖ CsI(g,c) + CsOH(g,c) system

- 초기 조건 (kg-mol/m³) :
  $C_{ini}[CsI_c] = 1.0$, $C_{ini}[CsI_g] = 0.0$
  $C_{ini}[CsOH_c] = 0.0$, $C_{ini}[CsOH_g] = 0.5$
  $C_{C,tot} = 1.0$, $T = 900.0$ K
- 계산 결과 (RASTEV) :

Param.	Value	Parameter	Value
C[CsI$_c$]	9.999995E−1 [kg-mol/m³]	C[CsOH$_c$]	4.999594E−1 [kg-mol/m³]
C[CsI$_g$]	5.223964E−7 [kg-mol/m³]	C[CsOH$_g$]	4.061228E−5 [kg-mol/m³]
p'(CsI)	5.863379E00 [Pa]	p'(CsOH)	9.117383E02 [Pa]
x(CsI$_c$)	0.6666846	x(CsOH$_c$)	0.3333154

- 검산 : R = 8314.3 J/kg-mol/K
  $C_G[CsI_g] = p'(CsI)\cdot x(CsI_c)/RT$
  $C_G[CsOH_g] = p'(CsOH)\cdot x(CsOH_c)/RT$
  $x(CsI_c) + x(CsOH_c) = 1$



온도에 따른 평형 농도 변화

General Equilibrium Thermochemistry

- **Governing Equations**

$$1 = \sum_{j \in C} x(j) = \sum_{j \in C} K_j \left[\prod_{i \in E}\left[C(i)RT\right]^{L_{ji}}\right]$$

$$\frac{n(i)}{V} = \frac{1}{RT}\sum_{j \in G} L_{ji}K_j \prod_{k \in E}\left[C(k)RT\right]^{L_{jk}} + C_C \sum_{j \in C} L_{ji}K_j \prod_{k \in E}\left[C(k)RT\right]^{L_{jk}} \qquad for\ i \in E$$

- **Dual iterative solving technique**
  - Cyclic Newton & Newton-Raphson methods
- **Implementation completed, verification & validation are needed.**

Conclusions

- **FP Chemical Reaction Module**
  - Successfully implemented with the equilibrium chemistry models
  - Verification & validation of the developed models are in progress.
- **Future Works**
  - Chemical Kinetics: Chemisorption, Oxidation, Iodine chemistry, etc.
  - Connectivity between the modules