SAND2021-14846C

Progress in Developing Thermochimica Modeling Capabilities to Aid in Simulating Severe Accidents in MELCOR

Benjamin Breeden[1], M. Poschmann[1], F. Gelbard[2], L. Albright[2], D. Luxat[2], M. Piro[1]

1. Ontario Tech University
2. Sandia National Laboratories

ANS 2021 Winter Conference
Nuclear Science User Facilities and Accident Tolerant Fuels
December 2, 2021

Sandia National Laboratories is a multimission laboratory managed and operated by National Technology & Engineering Solutions of Sandia, LLC, a wholly owned subsidiary of Honeywell International Inc., for the U.S. Department of Energy's National Nuclear Security Administration under contract DE-NA0003525.

Sandia National Laboratories is a multimission laboratory managed and operated by National Technology & Engineering Solutions of Sandia, LLC, a wholly owned subsidiary of Honeywell International Inc., for the U.S. Department of Energy's National Nuclear Security Administration under contract DE-NA0003525.

Objective

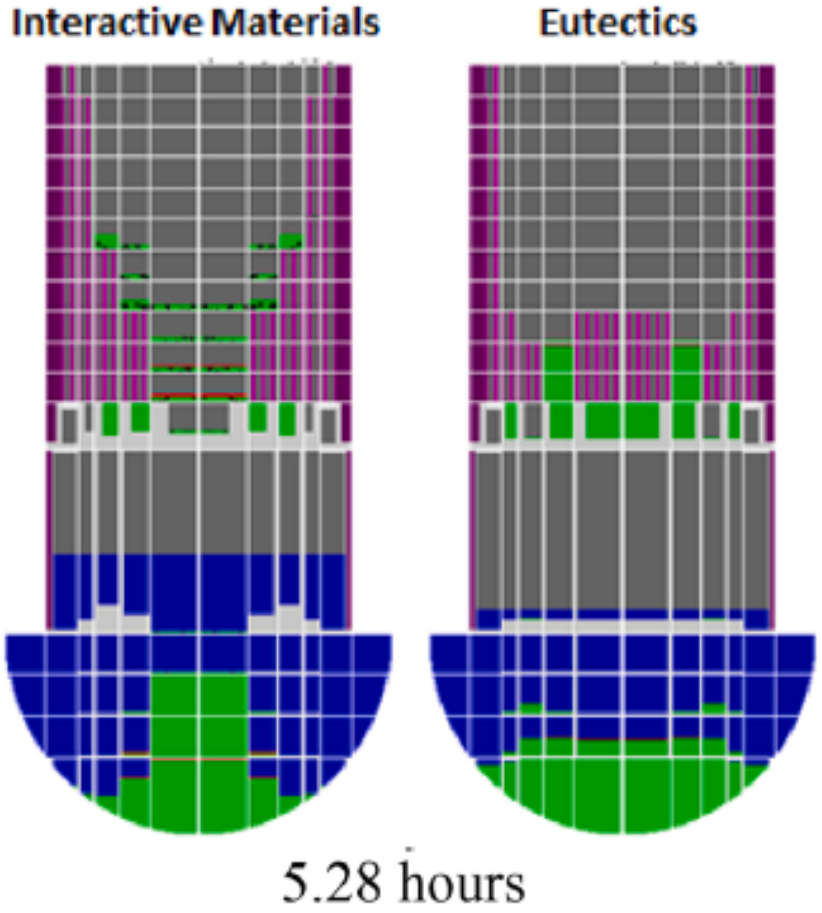
- Enhance the material modelling capabilities in MELCOR
  - Direct coupling with Thermochimica to leverage the Thermodynamics of Advanced Fuels – International Database (TAF-ID)

- Two main objectives of this work include
  - Develop the capabilities in Thermochimica to handle systems that include the Ionic Two Sub-Lattice Model (Completed work)
    - Required prior to using the TAF-ID

  - First look at anticipated material interaction modeling enhancements through one-way coupling between Thermochimica and MELCOR (Current Work)

MELCOR Code

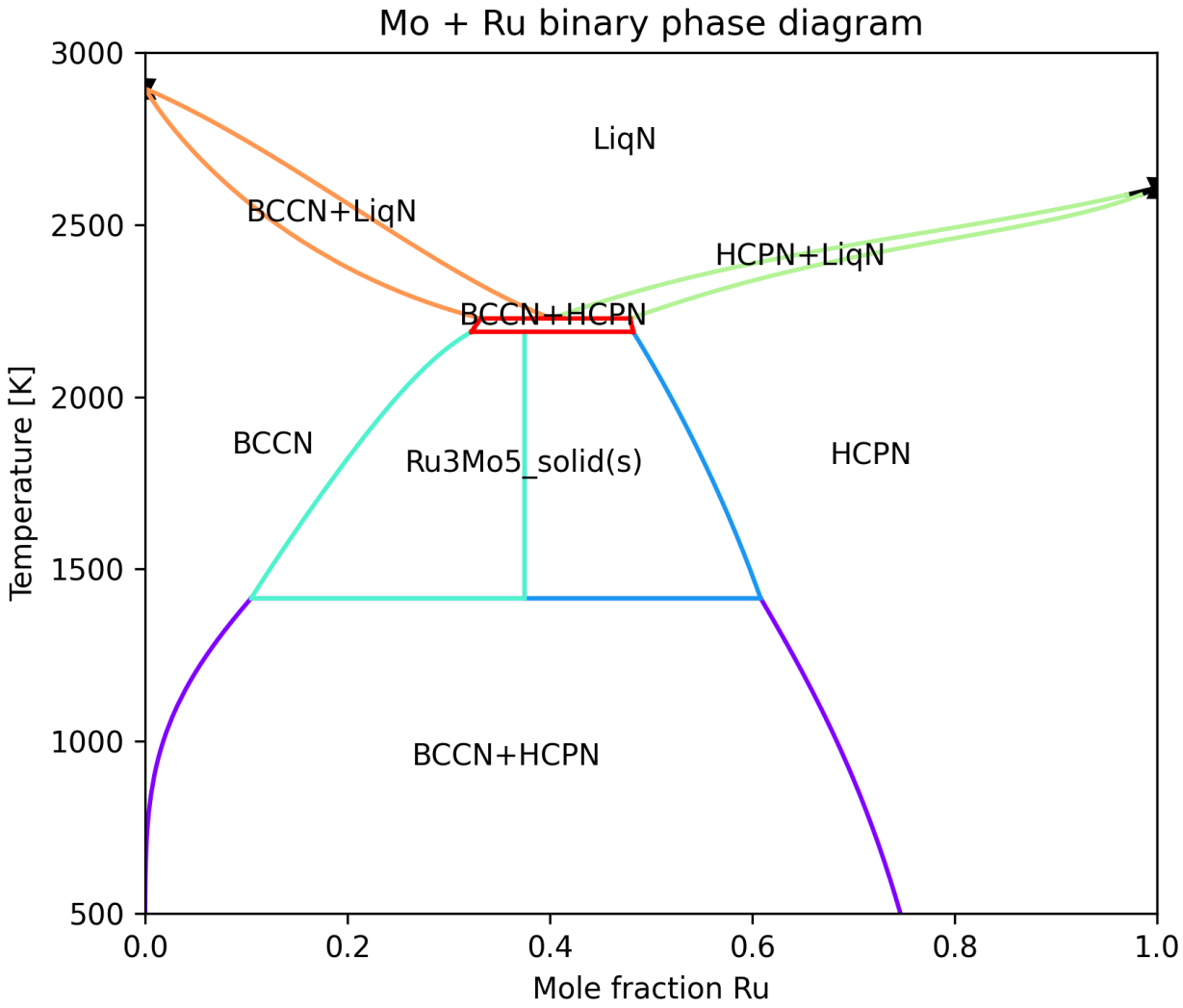
- Developed and maintained by Sandia National Laboratories (SNL) on behalf of the US Nuclear Regulatory Commission
- Models the progression of severe accidents in both light water reactors (LWRs) and non-LWRs
  - Supports plant licensing and regulatory reviews
  - User-adjustable parameters allow for uncertainty and sensitivity analyses
- Material Modeling
  - Interactive Model
  - Eutectics Model



Albright, L. et al. *Material Interactions in Severe Accidents*. Nucl. Eng. Des., 382 (2021)

Thermochimica

- Open source code maintained on GitHub repository
- Gibbs Energy Minimization software
  - Used to predict stable phases of a solution at equilibrium
- Multiple model types are used to model the thermodynamic solution phases
  - Important for this project is the Ionic Two-Sublattice Model (I2SL)
  - Thermochimica previously did not support the I2SL



Thermodynamics of Advanced Fuels – International Database (TAF-ID)

- Thermodynamic database
  - Hosted by the Organization for Economic Co-operation and Development Nuclear Energy Agency's (OCED-NEA)
  - Quality Assured
- Contains data for about 42 chemical elements
  - 218 binary systems
  - 85 ternary systems
- The focus of the data is on various nuclear materials
  - Multiple fuel types
  - Fission products
  - Other reactor components
- The Ionic Two-Sublattice Model is used to model the liquid phase for this database

Gueneau, C. et al. *TAF-ID: An international thermodynamic database for nuclear fuel applications*. CALPHAD, 72 (2021)

Thermochimica – Example Output

```
=========================================================
|                THERMOCHIMICA RESULTS                  |
=========================================================

     0.73799 mol LIQUID
                 { 0.45741
                 + 0.37728         U+4:O-2
                 + 1.3165E-12      U+4:Va
                 + 0.16531         Zr+4:O-2
                 + 5.7682E-13      Zr+4:Va   }

      ------------------------------------------------
      Sublattice 1; stoichiometric coefficient:  1.652
                 { U+4         0.69533
                 + Zr+4        0.30467    }

      Sublattice 2; stoichiometric coefficient:  4.000
                 { O-2         0.82593
                 + Va          7.2048E-13
                 + O           0.17407    }
      ------------------------------------------------
```

Continuation…

```
+ 0.49466 mol C1_MO2
                 { 1.9620E-06      U+3:O-2:O-2
                 + 1.0117E-03      U+3:O-2:Va
                 + 7.0349E-11      U+3:Va:O-2
                 + 3.6275E-08      U+3:Va:Va
                 + 9.2152E-04      U+4:O-2:O-2
                 + 0.47517         U+4:O-2:Va
                 + 3.3041E-08      U+4:Va:O-2
                 + 1.7037E-05      U+4:Va:Va
                 + 1.0342E-05      U+5:O-2:O-2
                 + 5.3329E-03      U+5:O-2:Va
                 + 3.7082E-10      U+5:Va:O-2
                 + 1.9121E-07      U+5:Va:Va
                 + 5.8255E-07      Zr+2:O-2:O-2
                 + 3.0039E-04      Zr+2:O-2:Va
                 + 2.0887E-11      Zr+2:Va:O-2
                 + 1.0770E-08      Zr+2:Va:Va
                 + 1.0011E-03      Zr+4:O-2:O-2
                 + 0.51621         Zr+4:O-2:Va
                 + 3.5894E-08      Zr+4:Va:O-2
                 + 1.8509E-05      Zr+4:Va:Va    }

      ------------------------------------------------
      Sublattice 1; stoichiometric coefficient:  1.000
                 { U+3         1.0137E-03
                 + U+4         0.47611
                 + U+5         5.3434E-03
                 + Zr+2        3.0098E-04
                 + Zr+4        0.51723    }

      Sublattice 2; stoichiometric coefficient:  2.000
                 { O-2         1.0000
                 + Va          3.5854E-05 }

      Sublattice 3; stoichiometric coefficient:  1.000
                 { O-2         1.9356E-03
                 + Va          0.99806    }
      ------------------------------------------------
```

Continuation…

```
+ 3.3217E-02 mol GAS
                 { 0.85199         O2
                 + 0.13464         O3U
                 + 1.3283E-02      O
                 + 7.8890E-05      O2U
                 + 2.9397E-07      O3
                 + 1.3766E-07      O2Zr
                 + 8.3475E-11      O1U
                 + 5.4039E-12      O1Zr
                 + 4.7414E-19      U
                 + 8.3018E-20      Zr
                 + 5.6250E-39      Zr2  }

=========================================================
|                  System properties                    |
=========================================================

 Temperature =    2500.00 [K]
 Pressure    =     1.0000 [atm]

 System Component  Mass [mol]  Chemical potential [J/mol]
 ----------------  ----------  --------------------------
 O                 3.0000E+00  -3.089794E+05
 U                 8.0000E-01  -9.210192E+05
 Zr                5.0000E-01  -8.510546E+05
 e-                0.0000E+00   4.108001E+04
 e-                0.0000E+00   5.000000E+06
 e-                0.0000E+00   5.338699E-10

 Integral Gibbs energy = -2.08928E+06 [J]
 Functional norm       =  5.74789E-07 [unitless]

 # of stable pure condensed phases =   0
 # of stable solution phases       =   3

=========================================================


DEBUG: Successful exit.
```

Ending.

Ionic Two-Sublattice Model

- ITSM can describe liquids behavior using the TAF-ID
- Evaluates short-range ordering of molecules in a liquid solution
- Two Sublattices
  - Sublattice 1: Cations $(C_i^{v_i})$ at charge v_i
  - Sublattice 2: Anions $(A_i^{v_i})$ at charge v_i, Vacancies (Va), and Neutrals (B_i)

$$\left(C_i^{v_i}\right)_P\left(A_i^{v_i}, Va, B_i^0\right)_Q$$

- P and Q are the average charge of the opposing sublattice

$$P = \sum_i (-v_i) y_{A_i} + Q y_{Va} \qquad Q = \sum_i (v_i) y_{C_i}$$

  - Where y is the site fraction and v_i is the charge

Sundman, B. *Modification of the Two-Sublattice Model for Liquids.* CALPHAD, 15 (1991) 109-119

Ionic Two-Sublattice Model

- Molar Gibbs energy equations for the reference, ideal, and excess mixing terms
  - Reference

$$g_{ref} = \sum_i \sum_j y_{C_i} y_{A_i} {}^{\circ}g_{C_i:A_j} + Q y_{Va} \sum_i y_{C_i} {}^{\circ}g_{C_i} + Q \sum_i y_{B_i} {}^{\circ}g_{B_i}$$

  - Ideal

$$g_{id} = RTP \sum_i y_{C_i} \ln(y_{C_i}) + RTQ \sum_i y_{A_i} \ln(y_{A_i}) + RTQ y_{Va} \ln(y_{Va}) + RTQ \sum_i y_{B_i} \ln(y_{B_i})$$

  - Excess Mixing Terms (Binary Only)

$$g_{ex}^{bin} = \sum_i \sum_{j>i} \sum_k y_{C_i} y_{C_j} y_{A_k} L_{C_i,C_j:A_k} + \sum_i \sum_i \sum_{k>j} y_{C_i} y_{A_j} y_{D_k} L_{C_i:A_j,D_k}$$
$$+ Q y_{Va} \sum_i \sum_{j>i} y_{C_i} y_{C_j} L_{C_i,C_j:Va} + Q y_{Va} \sum_i \sum_j y_{C_i} y_{B_j} L_{C_i:Va,B_j}$$
$$+ Q \sum_i \sum_{j>i} y_{B_i} y_{B_j} L_{B_i,B_j}$$

Sundman, B. *Modification of the Two-Sublattice Model for Liquids*. CALPHAD, 15 (1991) 109-119

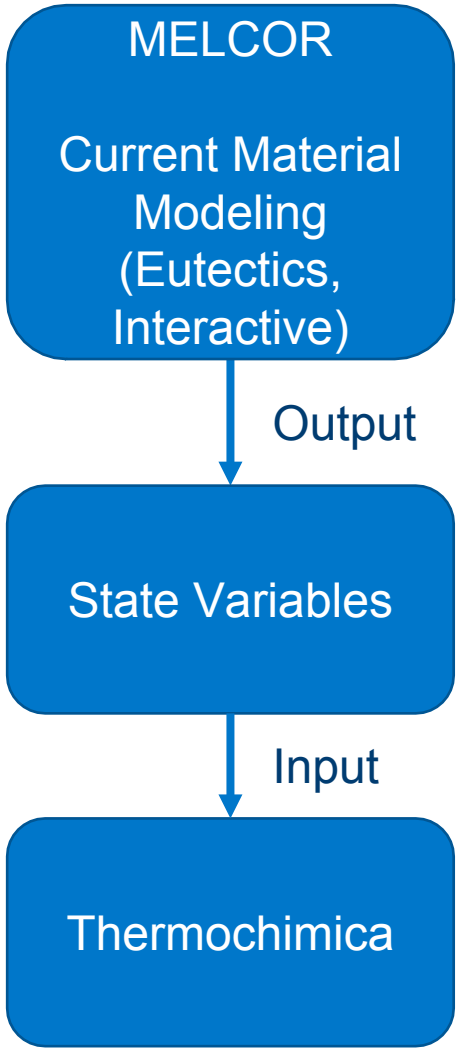
Completed Work

- The Ionic Two-Sublattice Model has been implemented into Thermochimica
- Verification and implementation of test case scenarios
  - Benchmark testing using FactSage, ThermoCalc, and OpenCalphad
  - Test case for each excess mixing term scenario

Test Case		Excess Mixing Case Being Tested												
Composition (mols)	Temp. (K)	1	2	3	4	5	6	7	8	9	10	11	12	Miscibility Gap
0.7 Sn - 0.3 0	1500													
1 Cr - 1 Zr - 1 O	2000													
0.2 Cs - 0.8 Te	700													
1 Ca - 1 Mn – 1 S	2500													
3 Ni - 1 B - 1 C	2000													
0.9 Mo - 0.5 Re - 0.8 C	2100													
0.5 Mo - 0.1 Rh - 1 Pd	2000													
1 Ca - 2 Al - 4 O	3500													
1 Ca - 2 Al - 4 O	3500													

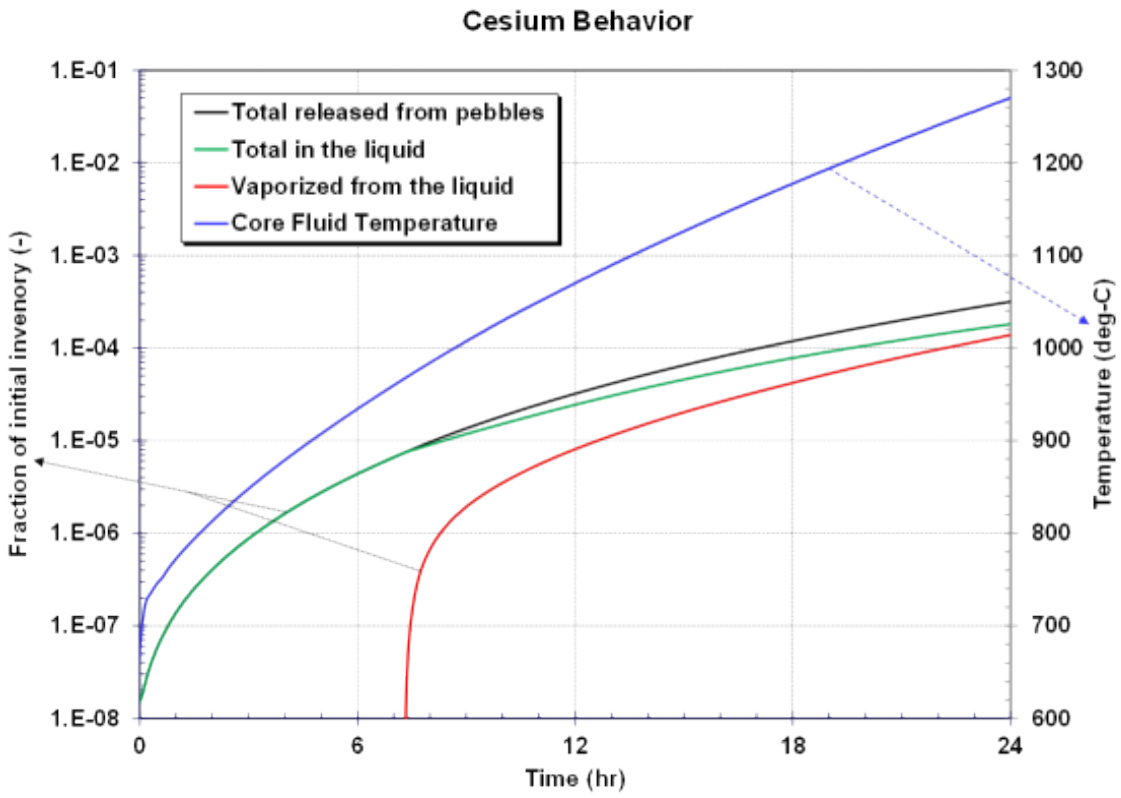
Current Work: One-way MELCOR-Thermochimica Coupling Demonstration

- Conversion of TAF-ID TDB to DAT Format
- MELCOR test case
  - Station black out scenario
  - BWR with Mark I containment
- Two Simulation Cases
  - Zircaloy-2 Cladding
  - FeCrAl Cladding
- Insights will guide new MELCOR modeling requirements
- Focus of the analysis
  - Liquid - solid phase fraction
  - Solid particulates in the mixture
  - Metallic molten pool



MELCOR Capability Demonstration with Molten Salt Chemistry and Radionuclide Release

- Thermochimica provides MELCOR's liquid-phase fission product transport and release model with thermochemical data
  - Solubilities
  - Vapour pressure
- Demonstration calculations utilize input data from modified version of the Molten Salt Thermodynamic Database
- Fluoride High-temperature Reactor LOCA sequence with no emergency heat removal
  - Core molten salt heatup toward saturation
  - Focus on Cs release from pebbles → liquid → gas
- Fission product example simulation with thermochemistry modeling
- Cs and CsF vaporization to gas space at elevated temperatures



Conclusion

- Enhance material modeling capabilities in MELCOR with thermochemical data
- The Ionic Two-Sublattice Model model has been developed in Thermochimica
  - TAF-ID data can now be used as an input
- As capability demonstration
  - One-way coupling of Thermochimica to MELCOR's COR package is underway
  - Two-way coupling has been achieved with the MSM to provide modified vapour phase data of Cs and CsF
- Future Work
  - Complete one-way coupling simulations
  - Complete two-way coupling of Thermochimica to MELCOR

Acknowledgements

- This research was undertaken, in part, thanks to funding from the Canada Research Chairs program of the Natural Sciences and Engineering Research Council of Canada.

- I would also like to acknowledge the SNL and MELCOR development team for partnering with our group and supporting this project.

Questions?