

Thermochemical Equilibrium for Multiphysics Simulations of Nuclear Materials

**Development of the corrosion modelling
application Yellowjacket**

Parikshit Bajpai

Supervisor: Prof. Markus H.A. Piro

PhD Candidacy Examination
December 9, 2019



Outline

① Introduction

Nuclear Energy

Generation IV Reactors

Multiphysics Simulations

② Goals of Research

③ Thermodynamic Equilibrium

Gibbs energy and chemical potential

Conditions of Thermodynamic Equilibrium

Gibbs Energy Minimisation

④ Computational Implementation

Gibbs Energy Minimiser - Overview

Gibbs Energy Minimiser

⑤ Progress and Timeline

Current Progress

Timeline

Outline

① Introduction

Nuclear Energy

Generation IV Reactors

Multiphysics Simulations

② Goals of Research

③ Thermodynamic Equilibrium

Gibbs energy and chemical potential

Conditions of Thermodynamic Equilibrium

Gibbs Energy Minimisation

④ Computational Implementation

Gibbs Energy Minimiser - Overview

Gibbs Energy Minimiser

⑤ Progress and Timeline

Current Progress

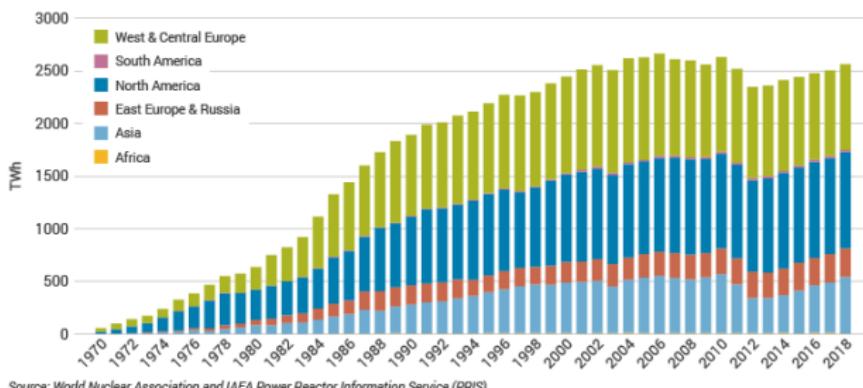
Timeline

Nuclear Energy Perspective

- Nuclear energy provides about 10% of the world's electricity from about 450 power reactors.

Nuclear Energy Perspective

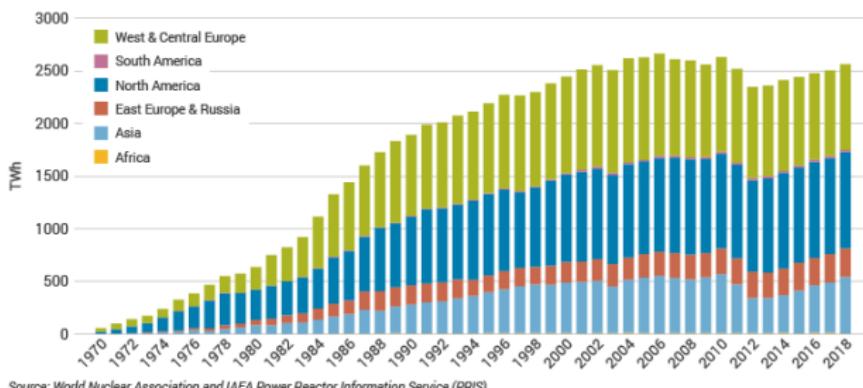
- Nuclear energy provides about 10% of the world's electricity from about 450 power reactors.
- Nuclear is the world's second largest source of low-carbon power (29% of the total in 2017).



Source: World Nuclear Association and IAEA Power Reactor Information Service (PRIS)

Nuclear Energy Perspective

- Nuclear energy provides about 10% of the world's electricity from about 450 power reactors.
- Nuclear is the world's second largest source of low-carbon power (29% of the total in 2017).



Source: World Nuclear Association and IAEA Power Reactor Information Service (PRIS)

- Nuclear power capacity worldwide is increasing steadily, with about 50 reactors under construction.

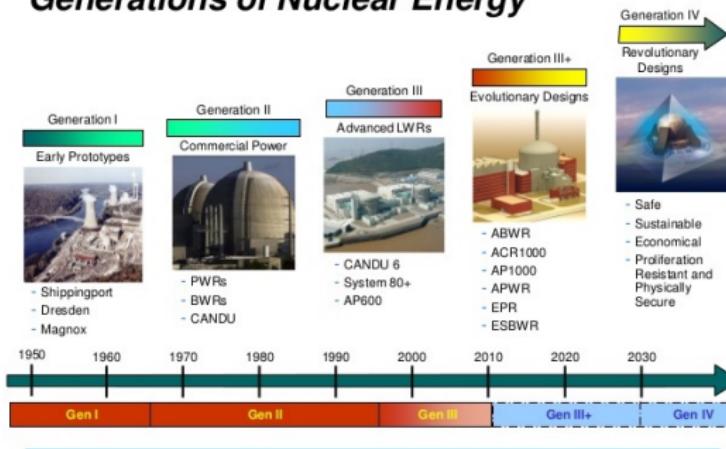
Nuclear Reactors

- Nuclear reactors are used to carry out controlled chain reactions to produce heat through fission.

Nuclear Reactors

- Nuclear reactors are used to carry out controlled chain reactions to produce heat through fission.
- Most of the reactors currently in use are 2nd / 3rd generation Light Water Reactors or Pressurised Heavy Water Reactors.

Generations of Nuclear Energy



Simulation of Nuclear Reactors

- Traditional reactor simulation tools rely on the *Operator Splitting* approach.

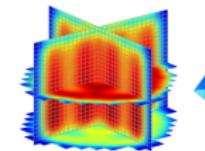
Simulation of Nuclear Reactors

- Traditional reactor simulation tools rely on the *Operator Splitting* approach.

$$\frac{\partial \langle \rho_g f_k \rangle}{\partial t}(\mathbf{r}, t) + \nabla \cdot (\rho_g f_k \hat{v}_k)(\mathbf{r}, t)$$

$$= \langle \rho_k \hat{v}_k \rangle(\mathbf{r}, t) + \nabla \cdot (\hat{\psi}_k)(\mathbf{r}, t) + \frac{1}{V} \oint_{S_h} [-\rho_h f_k(\mathbf{r}, t) \times (\hat{v}_k - \hat{v}_3)(\mathbf{r}, t) + \hat{\psi}_k(\mathbf{r}, t)] \cdot \hat{n} dS + \frac{1}{V} \oint_{S_h} \hat{\psi}_k(\mathbf{r}, t) \cdot \hat{n} dS$$

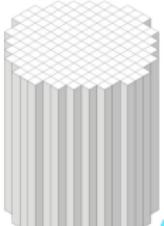
Neutron transport



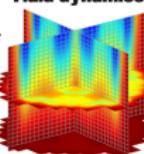
Thermal neutron flux $(\text{cm}^{-2} \text{s})$

$$\begin{aligned} \frac{1}{v_g} \frac{\partial \phi_{g,n}}{\partial t}(t) = & - \sum_{g' = 1, 2} \frac{f_{g,n}^N(t) - f_{g,n-1}^N(t)}{\Delta N} - \sum_{T_{g,g',n}}(t) \phi_{g',n}(t) \\ & + \sum_{g'=1}^G \sum_{g \rightarrow g',n} (t) \phi_{g',n}(t) + r_g^P (1-\beta) \sum_{g'=1}^G v_{g'} \sum_{f_{g',g',n}}(t) \phi_{g',n}(t) + \sum_{i=1}^6 \lambda_i^d \lambda_i C_{i,n}(t) \\ \frac{\partial C_{i,n}}{\partial t}(t) = & \beta_i \sum_{g'=1}^G v_{g'} \sum_{f_{g',g',n}}(t) \phi_{g',n}(t) - \lambda_i C_{i,n}(t), i = 1, \dots, 6 \end{aligned}$$

Nuclear reactor core



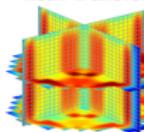
Fluid dynamics



Moderator density (g/cm^3)

$$\frac{1}{V} \int_V \rho(T) c_p(T) \frac{\partial T}{\partial t}(\mathbf{r}, t) dV = \frac{1}{V} \int_V k(T) \nabla T(\mathbf{r}, t) \cdot \hat{n} dS + \frac{1}{V} \int_V q'(v)(\mathbf{r}, t) dV$$

Heat transfer



Fuel temperature $(^\circ\text{C})$

Generation IV Reactors

- Generation IV reactors are advanced reactor concepts under development with the following goals:

Generation IV Reactors

- Generation IV reactors are advanced reactor concepts under development with the following goals:
 - Sustainability

Generation IV Reactors

- Generation IV reactors are advanced reactor concepts under development with the following goals:
 - Sustainability
 - Economy

Generation IV Reactors

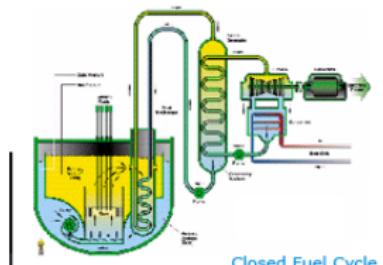
- Generation IV reactors are advanced reactor concepts under development with the following goals:
 - Sustainability
 - Economy
 - Safety and reliability

Generation IV Reactors

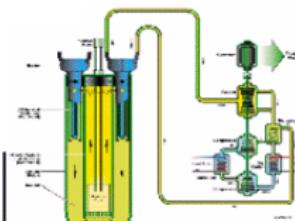
- Generation IV reactors are advanced reactor concepts under development with the following goals:
 - Sustainability
 - Economy
 - Safety and reliability
 - Proliferation resistance

Generation IV Reactors

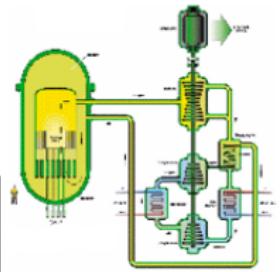
6 INNOVATIVE CONCEPTS WITH TECHNOLOGICAL BREAKTHROUGH



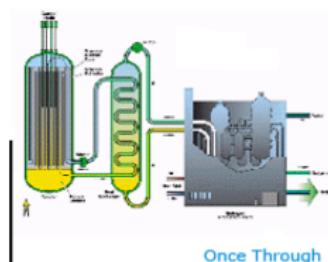
SODIUM FAST REACTOR



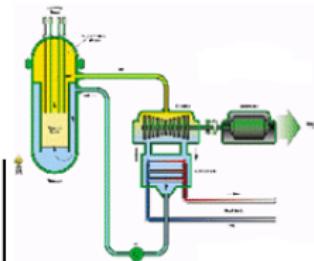
LEAD FAST REACTOR



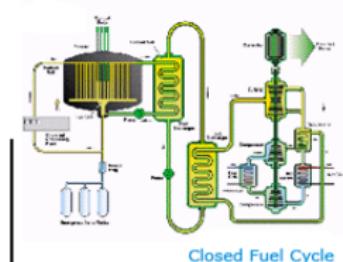
GAS FAST REACTOR



Once Through



Once/Closed



Closed Fuel Cycle

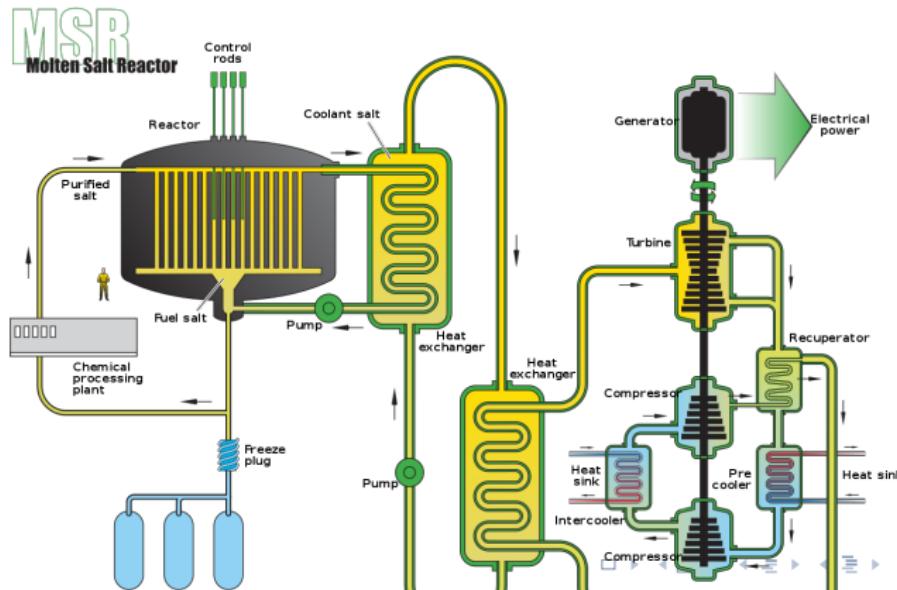


Molten Salt Reactor

- Molten Salt Reactor (MSR) are the reference circulating fuel concept under the Generation IV framework.

Molten Salt Reactor

- Molten Salt Reactor (MSR) are the reference circulating fuel concept under the Generation IV framework.

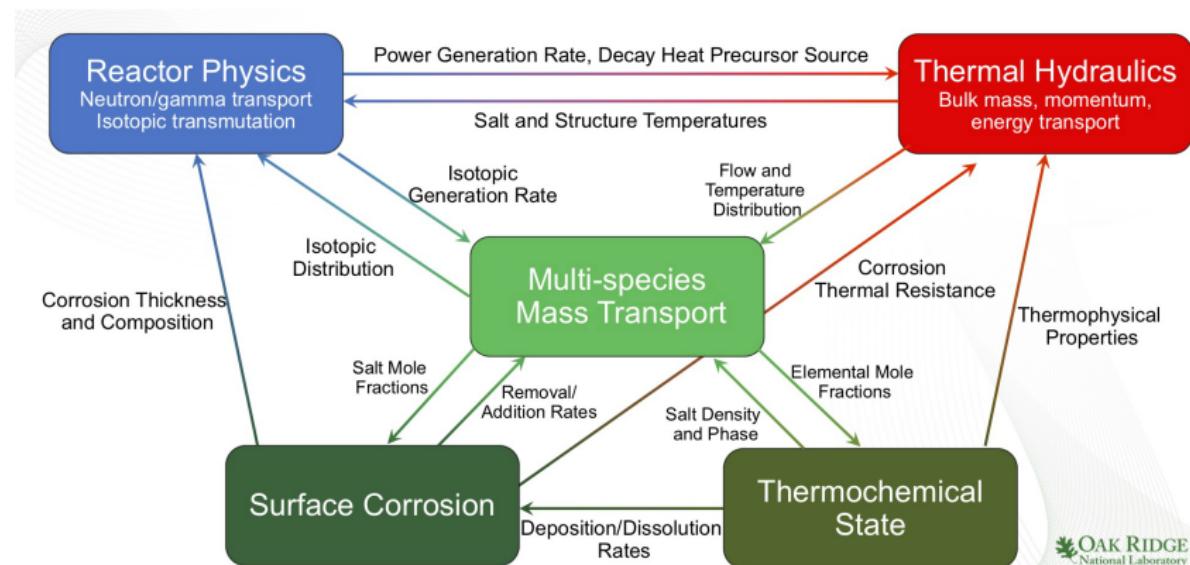


Molten Salt Reactor

- MSR exhibit very tight coupling between different physical phenomena.

Molten Salt Reactor

- MSR exhibit very tight coupling between different physical phenomena.



MOOSE

- Design and simulation of advanced reactors requires advanced multiphysics tools.

MOOSE

- Design and simulation of advanced reactors requires advanced multiphysics tools.
- Multiphysics Object Oriented Simulation Environment (MOOSE) is a finite element framework for solving computational engineering problems.

MOOSE

- Design and simulation of advanced reactors requires advanced multiphysics tools.
- Multiphysics Object Oriented Simulation Environment (MOOSE) is a finite element framework for solving computational engineering problems.
- Developed by the Idaho National Laboratory and open-source since 2014.

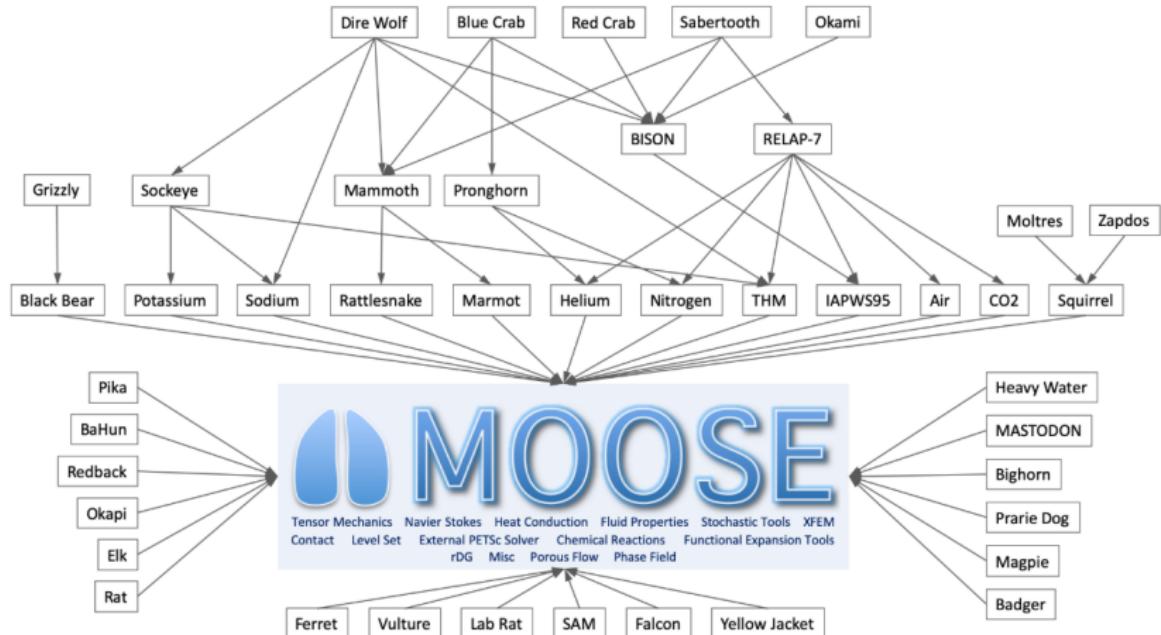
MOOSE

- Design and simulation of advanced reactors requires advanced multiphysics tools.
- Multiphysics Object Oriented Simulation Environment (MOOSE) is a finite element framework for solving computational engineering problems.
- Developed by the Idaho National Laboratory and open-source since 2014.
- MOOSE follows a Nuclear Quality Assurance Level 1 (NQA-1) development process.

MOOSE

- Design and simulation of advanced reactors requires advanced multiphysics tools.
- Multiphysics Object Oriented Simulation Environment (MOOSE) is a finite element framework for solving computational engineering problems.
- Developed by the Idaho National Laboratory and open-source since 2014.
- MOOSE follows a Nuclear Quality Assurance Level 1 (NQA-1) development process.
- MOOSE includes a test suite and documentation system to allow for agile development while maintaining a NQA-1 process.

MOOSE

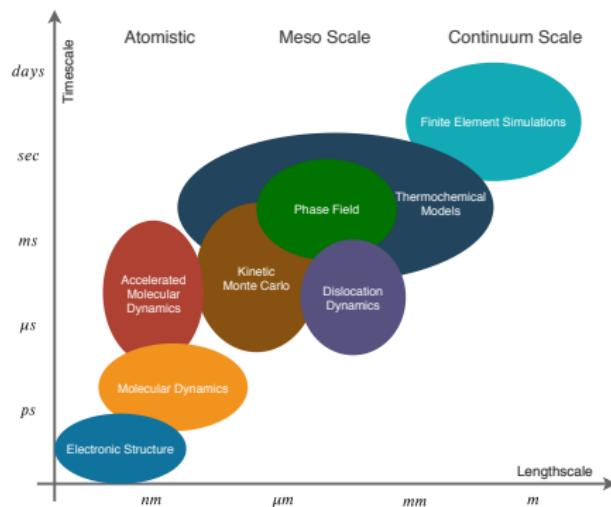


Nuclear Materials

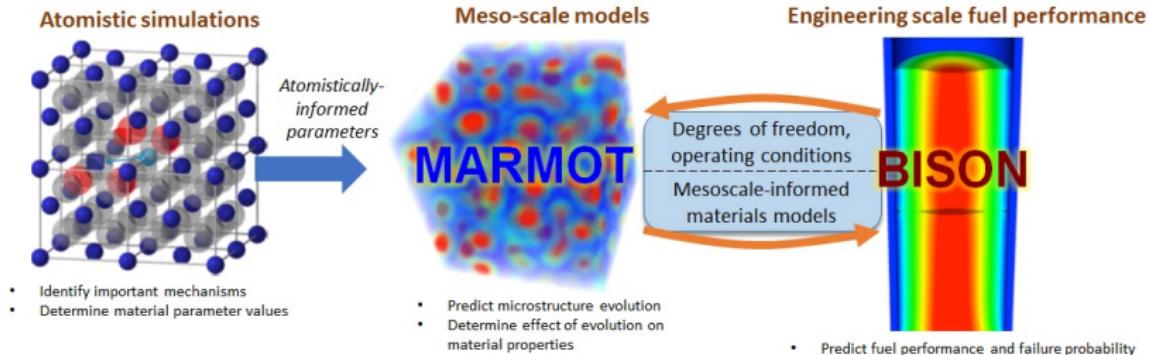
- Nuclear materials are highly complex multiscale, multiphysics systems.

Nuclear Materials

- Nuclear materials are highly complex multiscale, multiphysics systems.



Nuclear Materials



Corrosion

- MSR and other advanced reactors use high temperature fluids such as molten fluoride/chloride salts.

Corrosion

- MSR and other advanced reactors use high temperature fluids such as molten fluoride/chloride salts.
- High temperature fluids especially molten fluoride/chloride salts cause corrosion of structural materials.

Corrosion

- MSR and other advanced reactors use high temperature fluids such as molten fluoride/chloride salts.
- High temperature fluids especially molten fluoride/chloride salts cause corrosion of structural materials.
- Corrosion is an electrochemical process driven by the thermodynamic and kinetics of the system.

Corrosion

- MSR and other advanced reactors use high temperature fluids such as molten fluoride/chloride salts.
- High temperature fluids especially molten fluoride/chloride salts cause corrosion of structural materials.
- Corrosion is an electrochemical process driven by the thermodynamic and kinetics of the system.
- Effective prediction of corrosion requires a multiscale, mutiphysics approach.

Corrosion

- MSR and other advanced reactors use high temperature fluids such as molten fluoride/chloride salts.
- High temperature fluids especially molten fluoride/chloride salts cause corrosion of structural materials.
- Corrosion is an electrochemical process driven by the thermodynamic and kinetics of the system.
- Effective prediction of corrosion requires a multiscale, mutiphysics approach.

Currently, MOOSE lacks a corrosion modelling tool!

Yellowjacket

- Yellowjacket is a new corrosion modelling tool currently under development.

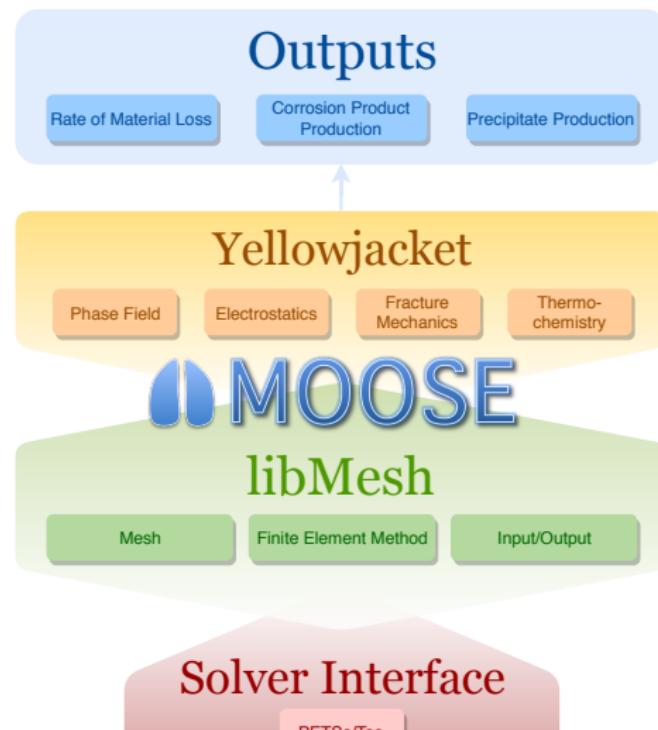
Yellowjacket

- Yellowjacket is a new corrosion modelling tool currently under development.
- Yellowjacket will couple phase field models with thermodynamic equilibrium calculations to effectively model corrosion in advanced reactors.

Yellowjacket

- Yellowjacket is a new corrosion modelling tool currently under development.
- Yellowjacket will couple phase field models with thermodynamic equilibrium calculations to effectively model corrosion in advanced reactors.
- Collaboration between Ontario Tech University, University of Florida, Idaho National Laboratory and Los Alamos National Laboratory.

Yellowjacket



Thermodynamics in Yellowjacket

- Phase and chemical behaviour of nuclear materials is governed by the thermodynamic equilibrium state.

Thermodynamics in Yellowjacket

- Phase and chemical behaviour of nuclear materials is governed by the thermodynamic equilibrium state.
- The composition of nuclear materials continuously evolves under irradiation affecting the equilibrium state.

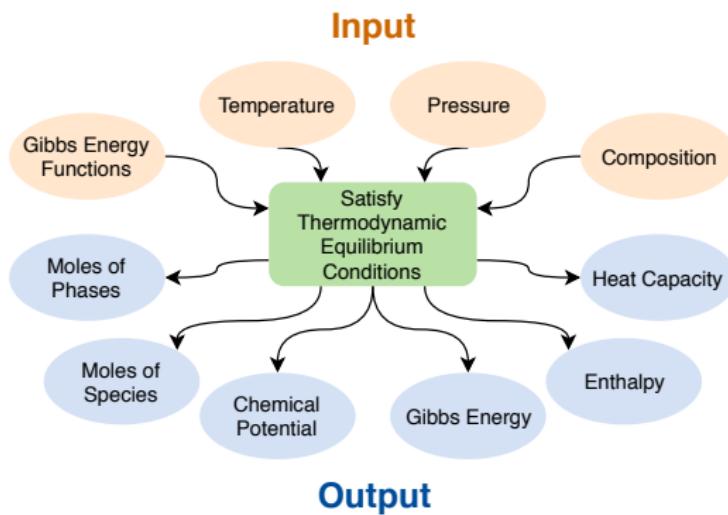
Thermodynamics in Yellowjacket

- Phase and chemical behaviour of nuclear materials is governed by the thermodynamic equilibrium state.
- The composition of nuclear materials continuously evolves under irradiation affecting the equilibrium state.
- Capturing thermodynamic equilibria has traditionally relied on empirical correlations.

Thermodynamics in Yellowjacket

- Phase and chemical behaviour of nuclear materials is governed by the thermodynamic equilibrium state.
- The composition of nuclear materials continuously evolves under irradiation affecting the equilibrium state.
- Capturing thermodynamic equilibria has traditionally relied on empirical correlations.
- Direct coupling of thermodynamic equilibrium can augment multiphysics calculations by providing quantities such as the chemical potentials, Gibbs energies, etc.

Thermodynamics in Yellowjacket



Outline

① Introduction

Nuclear Energy

Generation IV Reactors

Multiphysics Simulations

② Goals of Research

③ Thermodynamic Equilibrium

Gibbs energy and chemical potential

Conditions of Thermodynamic Equilibrium

Gibbs Energy Minimisation

④ Computational Implementation

Gibbs Energy Minimiser - Overview

Gibbs Energy Minimiser

⑤ Progress and Timeline

Current Progress

Timeline

Impetus

Goal of Research

The goal of this work is to develop a new state-of-the-art thermodynamic equilibrium code for direct integration in multiphysics framework MOOSE.

Impetus

Goal of Research

The goal of this work is to develop a new state-of-the-art thermodynamic equilibrium code for direct integration in mutiphysics framework MOOSE.

Outcomes

- ① Development of a new advanced Gibbs energy minimiser written in C++ within the framework of MOOSE platform.
- ② Full integration within the multiphysics framework MOOSE, with the intent of coupling to the phase field code Marmot.
- ③ Enhanced initialisation algorithms to improve the computational performance.
- ④ Investigation and implementation of robust global optimisation schemes to increase reliability and robustness.
- ⑤ Software Quality Assurance with rigorous verification and testing to comply with the NQA-1 guidelines required to be met for licensing.

Outline

① Introduction

Nuclear Energy
Generation IV Reactors
Multiphysics Simulations

② Goals of Research

③ Thermodynamic Equilibrium

Gibbs energy and chemical potential
Conditions of Thermodynamic Equilibrium
Gibbs Energy Minimisation

④ Computational Implementation

Gibbs Energy Minimiser - Overview
Gibbs Energy Minimiser

⑤ Progress and Timeline

Current Progress
Timeline

Gibbs Energy

- Developed by Josiah Willard Gibbs in 1873.

Available energy is the greatest amount of mechanical work which can be obtained from a given quantity of a certain substance in a given initial state, without increasing its total volume or allowing heat to pass to or from external bodies, except such as at the close of the processes are left in their initial condition.

Gibbs Energy

- Developed by Josiah Willard Gibbs in 1873.

Available energy is the greatest amount of mechanical work which can be obtained from a given quantity of a certain substance in a given initial state, without increasing its total volume or allowing heat to pass to or from external bodies, except such as at the close of the processes are left in their initial condition.

- Relates the *enthalpy* of the system to its *entropy*.

$$G = H - TS$$

Gibbs Energy

- Developed by Josiah Willard Gibbs in 1873.

Available energy is the greatest amount of mechanical work which can be obtained from a given quantity of a certain substance in a given initial state, without increasing its total volume or allowing heat to pass to or from external bodies, except such as at the close of the processes are left in their initial condition.

- Relates the *enthalpy* of the system to its *entropy*.

$$G = H - TS$$

- Gibbs energy is the thermodynamic quantity that is minimised when a system reaches chemical equilibrium at constant temperature and pressure.

Chemical Potential

- Chemical potential of species i in phase λ is a measure of the change in Gibbs energy of the system by the introduction of species i .

$$\mu_{i(\lambda)} = \left(\frac{\partial G_{sys}}{\partial n_{i(\lambda)}} \right)_{T,P,n_j \neq i}$$

Chemical Potential

- Chemical potential of species i in phase λ is a measure of the change in Gibbs energy of the system by the introduction of species i .

$$\mu_{i(\lambda)} = \left(\frac{\partial G_{sys}}{\partial n_{i(\lambda)}} \right)_{T,P,n_j \neq i}$$

- Ideal mixing phases

$$\mu_{i(\lambda)} = g_{i(\lambda)}^0 + \ln x_{i(\lambda)}$$

Chemical Potential

- Chemical potential of species i in phase λ is a measure of the change in Gibbs energy of the system by the introduction of species i .

$$\mu_{i(\lambda)} = \left(\frac{\partial G_{sys}}{\partial n_{i(\lambda)}} \right)_{T,P,n_j \neq i}$$

- Ideal mixing phases

$$\mu_{i(\lambda)} = g_{i(\lambda)}^0 + \ln x_{i(\lambda)}$$

- Non-ideal mixing phases

$$\mu_{i(\lambda)} = g_{i(\lambda)}^0 + \ln x_{i(\lambda)} + g_{i(\lambda)}^{ex}$$

Gibbs Energy

- Integral Gibbs energy of a multicomponent, multiphase system can be expressed in terms of chemical potentials, $\mu_{i(\lambda)}$, of the species.

$$G_{\text{sys}} = RT \left(\sum_{\lambda=1}^{\Lambda} n_{\lambda} \sum_{i=1}^{N_{\lambda}} x_{i(\lambda)} \tilde{\mu}_i + \sum_{\omega=1}^{\Omega} n_{\omega} \tilde{\mu}_{\omega} \right)$$

Gibbs Energy

- Integral Gibbs energy of a multicomponent, multiphase system can be expressed in terms of chemical potentials, $\mu_i(\lambda)$, of the species.

$$G_{\text{sys}} = RT \left(\sum_{\lambda=1}^{\Lambda} n_{\lambda} \sum_{i=1}^{N_{\lambda}} x_{i(\lambda)} \tilde{\mu}_i + \sum_{\omega=1}^{\Omega} n_{\omega} \tilde{\mu}_{\omega} \right)$$

- The Gibbs energy of the system can also be expressed in terms of the element potentials, Γ_j , and the number of moles, b_j , of the system components.

$$G_{\text{sys}} = \sum_{j=1}^C \Gamma_j b_j$$

Thermodynamic Equilibrium - Necessary Conditions

Thermodynamic Equilibrium - Necessary Conditions

Conservation of mass

$$b_j = \sum_{\lambda=1}^{\Lambda} n_{\lambda} \sum_{i=1}^{N_{\lambda}} x_{i(\lambda)} \nu_{i,j} + \sum_{\omega=1}^{\Omega} n_{\omega} \nu$$

Thermodynamic Equilibrium - Necessary Conditions

Conservation of mass

$$b_j = \sum_{\lambda=1}^{\Lambda} n_{\lambda} \sum_{i=1}^{N_{\lambda}} x_{i(\lambda)} \nu_{i,j} + \sum_{\omega=1}^{\Omega} n_{\omega} \nu$$

Gibbs' phase rule

$$F = C - \Phi + 2 + \Xi$$

Thermodynamic Equilibrium - Necessary Conditions

Conservation of mass

$$b_j = \sum_{\lambda=1}^{\Lambda} n_{\lambda} \sum_{i=1}^{N_{\lambda}} x_{i(\lambda)} \nu_{i,j} + \sum_{\omega=1}^{\Omega} n_{\omega} \nu$$

Gibbs' phase rule

$$F = C - \Phi + 2 + \Xi$$

Gibbs' Criteria

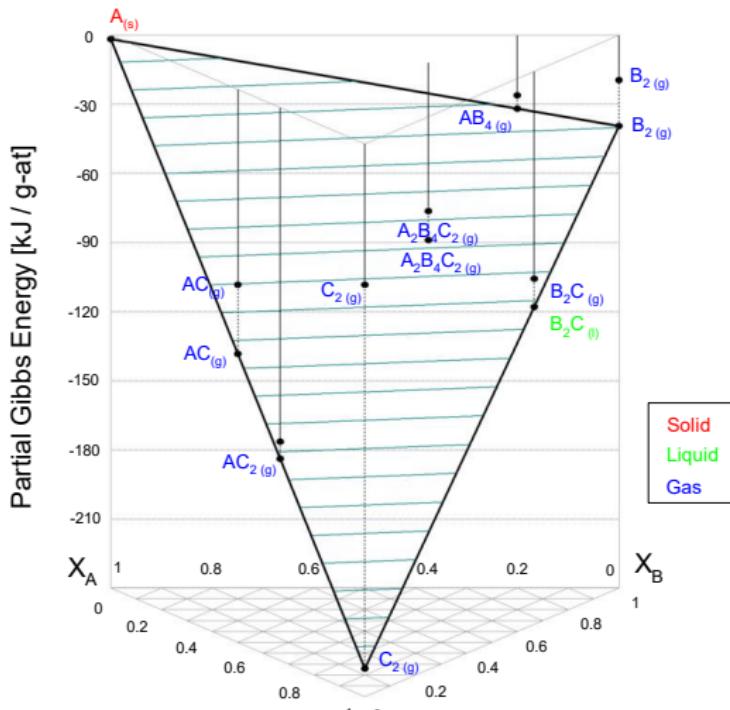
$$\mu_i = \sum_{j=1}^C \nu_{i,j} \Gamma_j$$

Thermodynamic Equilibrium - Sufficient Conditions

Gibbs plane

$$\pi_\lambda = \min_{\lambda} \sum_{i=1}^{N_\lambda} x_{i(\lambda)} \left(\mu_{i(\lambda)} - \sum_{j=1}^c \nu_{i,j} \Gamma_j \right)$$

Thermodynamic Equilibrium - Sufficient Conditions



Gibbs Energy Minimisation

- Gibbs Energy Minimisation was proposed in 1958 by White, Jonson and Dantzig and is used almost ubiquitously in computation of thermodynamic equilibrium.

Gibbs Energy Minimisation

- Gibbs Energy Minimisation was proposed in 1958 by White, Jonson and Dantzig and is used almost ubiquitously in computation of thermodynamic equilibrium.
- Second order steepest descent method.

Gibbs Energy Minimisation

- Gibbs Energy Minimisation was proposed in 1958 by White, Jonson and Dantzig and is used almost ubiquitously in computation of thermodynamic equilibrium.
- Second order steepest descent method.
- Constrains Gibbs' phase rule while simultaneously minimising the mass balance and Gibbs' criteria residuals.

Gibbs Energy Minimisation

Optimise

$$b_j = \sum_{\lambda=1}^{\Lambda} n_{\lambda} \sum_{i=1}^{N_{\lambda}} x_{i(\lambda)} \nu_{i,j} + \sum_{\omega=1}^{\Omega} n_{\omega} \nu$$

Constraint

$$F = C - \Phi + 2 + \Xi$$

Optimise

$$\mu_i = \sum_{j=1}^C \nu_{i,j} \Gamma_j$$

Outline

① Introduction

Nuclear Energy
Generation IV Reactors
Multiphysics Simulations

② Goals of Research

③ Thermodynamic Equilibrium

Gibbs energy and chemical potential
Conditions of Thermodynamic Equilibrium
Gibbs Energy Minimisation

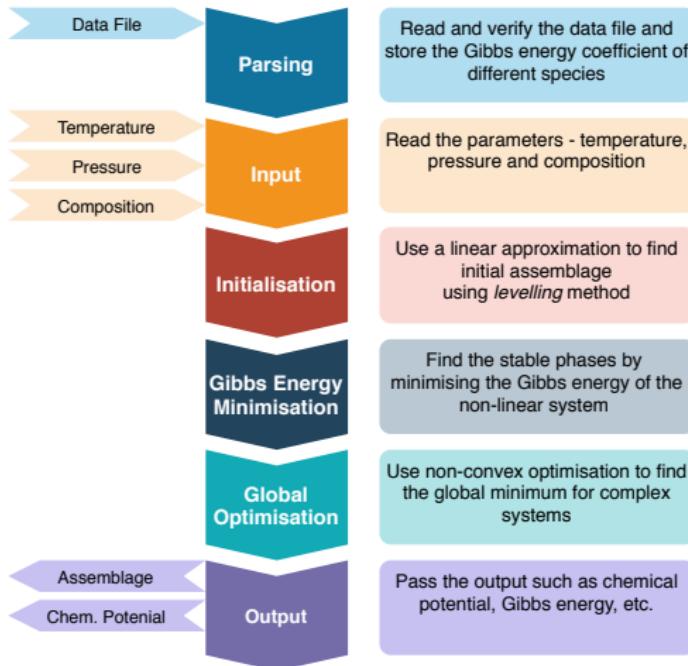
④ Computational Implementation

Gibbs Energy Minimiser - Overview
Gibbs Energy Minimiser

⑤ Progress and Timeline

Current Progress
Timeline

Computational Structure



Parsing

- The data files are created using the well known Calphad method and can be available in different formats.

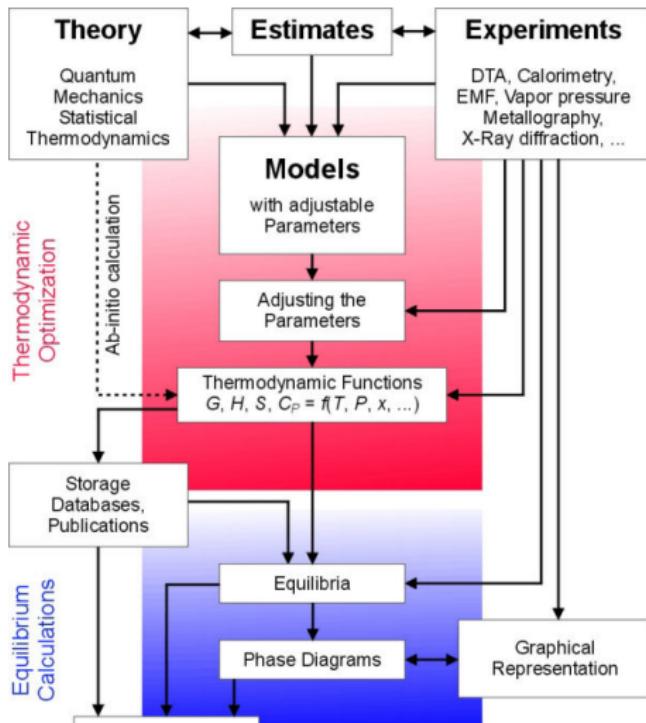
Parsing

- The data files are created using the well known Calphad method and can be available in different formats.
- Most commonly used formats are ThermoCalc (*.tdb) and ChemSage (*.dat).

Parsing

- The data files are created using the well known Calphad method and can be available in different formats.
- Most commonly used formats are ThermoCalc (*.tdb) and ChemSage (*.dat).
- Yellowjacket uses ChemSage (*.dat) format datafiles, which can be generated by the commercial software FactSage.

Parsing



Input

- Temperature, pressure and composition are required at each time step and for each mesh element.
- At each time step MOOSE can provide these inputs to Yellowjacket.

Initialisation

- Gibbs energy minimisers require an initial estimate of molar quantities of species and phases.

Initialisation

- Gibbs energy minimisers require an initial estimate of molar quantities of species and phases.
- *Levelling* is an estimating process developed by Eriksson and Thompson (1989).

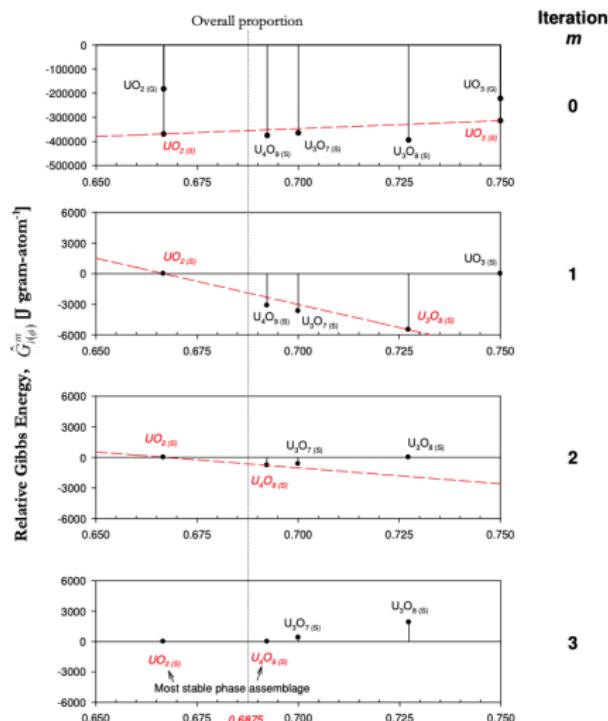
Initialisation

- Gibbs energy minimisers require an initial estimate of molar quantities of species and phases.
- *Levelling* is an estimating process developed by Eriksson and Thompson (1989).
- Levelling converts the non-linear optimisation problem into a linear optimisation problem by treating all species and phases as pure separate phases.

Initialisation

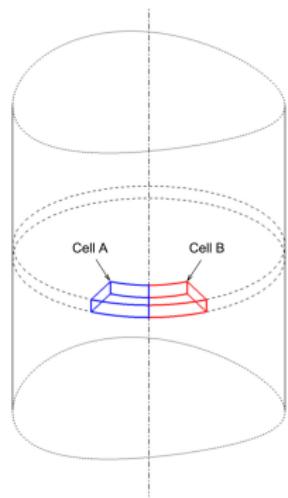
- Gibbs energy minimisers require an initial estimate of molar quantities of species and phases.
- *Levelling* is an estimating process developed by Eriksson and Thompson (1989).
- Levelling converts the non-linear optimisation problem into a linear optimisation problem by treating all species and phases as pure separate phases.
- The number of iterations required to achieve convergence does not increase rapidly with the number of system components.

Initialisation

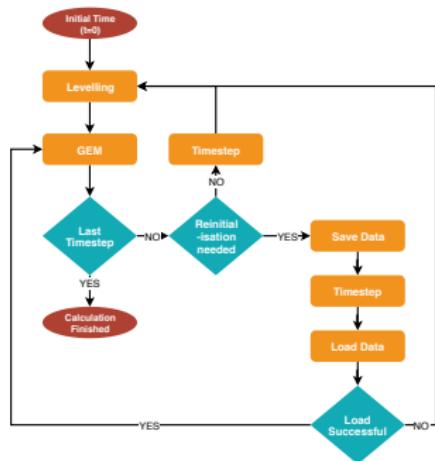


Initialisation

- Initialise using neighbour cells



- Initialise with last time step



Gibbs Energy Minimisation

- Hessian Solver

$$H \cdot \pi = \zeta$$

Gibbs Energy Minimisation

- Hessian Solver

$$\mathbf{H} \cdot \boldsymbol{\pi} = \boldsymbol{\zeta}$$

$$\mathbf{H} = \begin{bmatrix} r_{j=1,k=1} & \dots & r_{j=1,k=C} & \phi_{j=1,\lambda=1} & \dots & \phi_{j=1,\lambda=\Lambda} & \nu_{j=1,\omega=1} \\ \vdots & \ddots & \vdots & \vdots & \ddots & \vdots & \vdots \\ r_{j=C,k=1} & \dots & r_{j=C,k=C} & \phi_{j=C,\lambda=1} & \dots & \phi_{j=C,\lambda=\Lambda} & \nu_{j=C,\omega=1} \\ \phi_{\lambda=1,j=1} & \dots & \phi_{\lambda=1,j=C} & 0 & \dots & 0 & 0 \\ \vdots & \ddots & \vdots & \vdots & \ddots & \vdots & \vdots \\ \phi_{\lambda=\Lambda,j=1} & \dots & \phi_{\lambda=\Lambda,j=C} & 0 & \dots & 0 & 0 \\ \nu_{\omega=1,j=1} & \dots & \nu_{\omega=1,j=C} & 0 & \dots & 0 & 0 \\ \vdots & \ddots & \vdots & \vdots & \ddots & \vdots & \vdots \\ \nu_{\omega=\Omega,j=1} & \dots & \nu_{\omega=\Omega,j=C} & 0 & \dots & 0 & 0 \end{bmatrix}$$

Gibbs Energy Minimisation

- Hessian Solver

$$H \cdot \pi = \zeta$$

$$\boldsymbol{\pi} = \begin{bmatrix} \pi_{j=1}^{m+1} \\ \vdots \\ \pi_{j=E}^{m+1} \\ \pi_{\lambda=1}^{m+1} \\ \vdots \\ \pi_{\lambda=\Lambda}^{m+1} \\ \pi_{\omega=1}^{m+1} \\ \vdots \\ \pi_{\omega=W}^{m+1} \end{bmatrix}$$

Gibbs Energy Minimisation

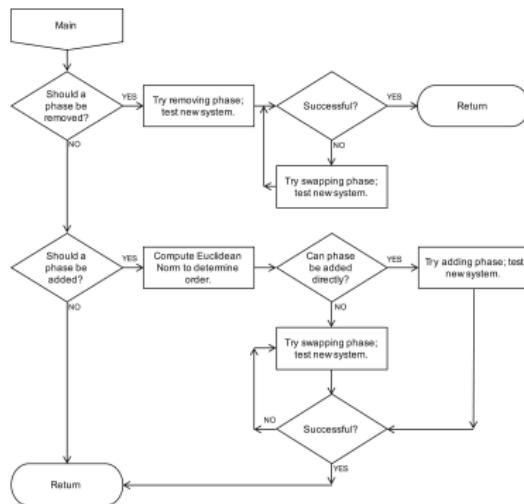
- Hessian Solver

$$\mathbf{H} \cdot \boldsymbol{\pi} = \boldsymbol{\zeta}$$

$$\boldsymbol{\zeta} = \begin{bmatrix} b_{j=1} + \sum_{\lambda=1}^{\Lambda} \sum_{i=1}^{N_\lambda} \left(\frac{\mu_{i(\lambda)}^m}{RT} - 1 \right) n_{i(\lambda)}^m \nu_{i,j=1} \\ \vdots \\ b_{j=E} + \sum_{\lambda=1}^{\Lambda} \sum_{i=1}^{N_\lambda} \left(\frac{\mu_{i(\lambda)}^m}{RT} - 1 \right) n_{i(\lambda)}^m \nu_{i,j=E} \\ \sum_{i=1}^{N_{\lambda=1}} \left(\frac{\mu_{i(\lambda=1)}^m}{RT} - 1 \right) n_{i(\lambda=1)}^m \\ \vdots \\ \sum_{i=1}^{N_{\lambda=\Lambda}} \left(\frac{\mu_{i(\lambda=\Lambda)}^m}{RT} - 1 \right) n_{i(\lambda=\Lambda)}^m \\ \frac{\mu_{\omega=1}^m}{RT} \end{bmatrix}$$

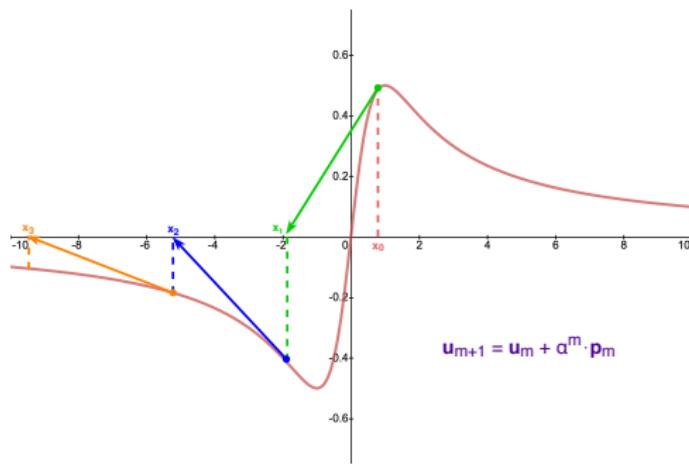
Gibbs Energy Minimisation

- Phase assemblage algorithm



Gibbs Energy Minimisation

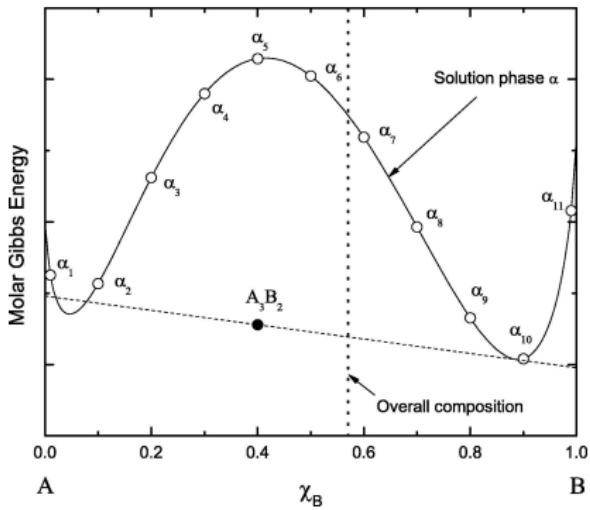
- Line Search algorithms



- Use of Wolfe/Armijo conditions can help avoid divergence.

Global Optimisation

- The Gibbs energy function of non-ideal phases may be non-convex, yielding multiple local minima. This makes finding global minimum a challenge.



Global Optimisation

- Mathematically, global optimisation implies finding the Gibbs plane that satisfies the sufficient condition:

$$\pi_\lambda = \min_{\lambda} \sum_{i=1}^{N_\lambda} x_{i(\lambda)} \left(\mu_{i(\lambda)} - \sum_{j=1}^C \nu_{i,j} \Gamma_j \right)$$

- No global optimisation technique guarantees the ability of finding a global extremum of a non-convex function.
- Searching for a global minimum becomes increasingly more difficult as the size of the system increases.
- The computational effort associated with performing this task can increase very rapidly in large systems.

Output

- Outputs after the Gibbs energy minimisation and global optimisation include moles of phases, species mole fraction in each phase, chemical potentials, Gibbs energy, etc.
- The chemical potential of various species can be used to find the driving force for other reactions such as corrosion.
- These parameters can also be passed to other MOOSE based codes such as Marmot and Bison.

Outline

① Introduction

- Nuclear Energy
- Generation IV Reactors
- Multiphysics Simulations

② Goals of Research

③ Thermodynamic Equilibrium

- Gibbs energy and chemical potential
- Conditions of Thermodynamic Equilibrium
- Gibbs Energy Minimisation

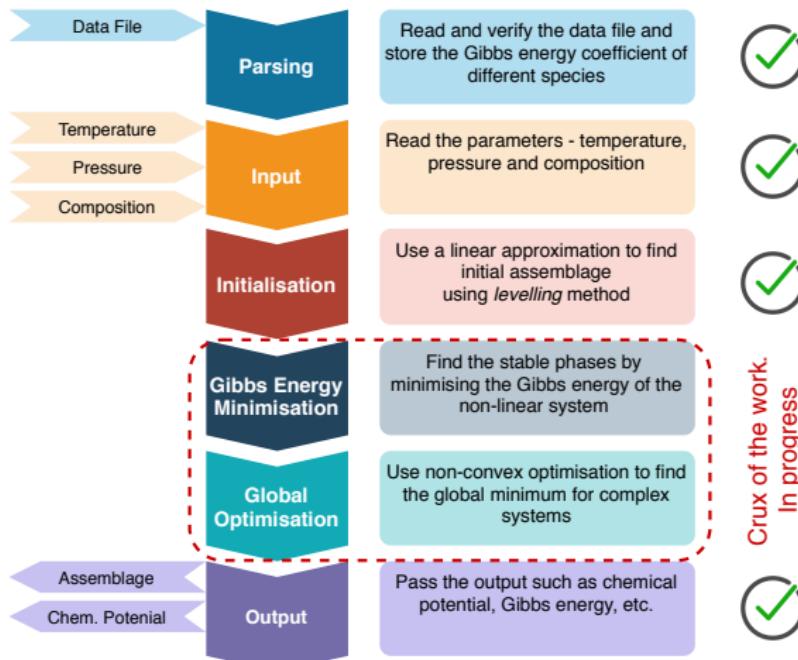
④ Computational Implementation

- Gibbs Energy Minimiser - Overview
- Gibbs Energy Minimiser

⑤ Progress and Timeline

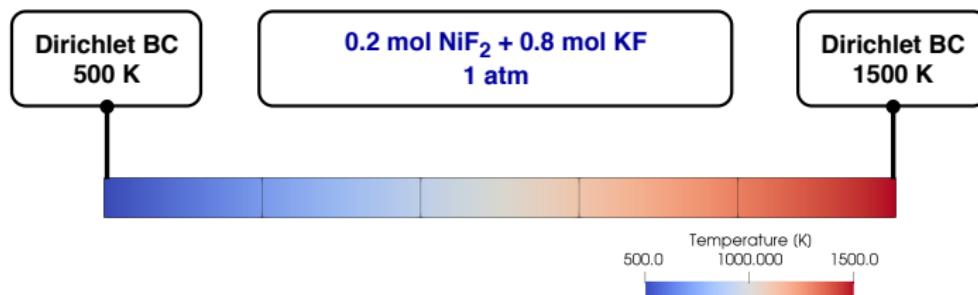
- Current Progress
- Timeline

Current Progress

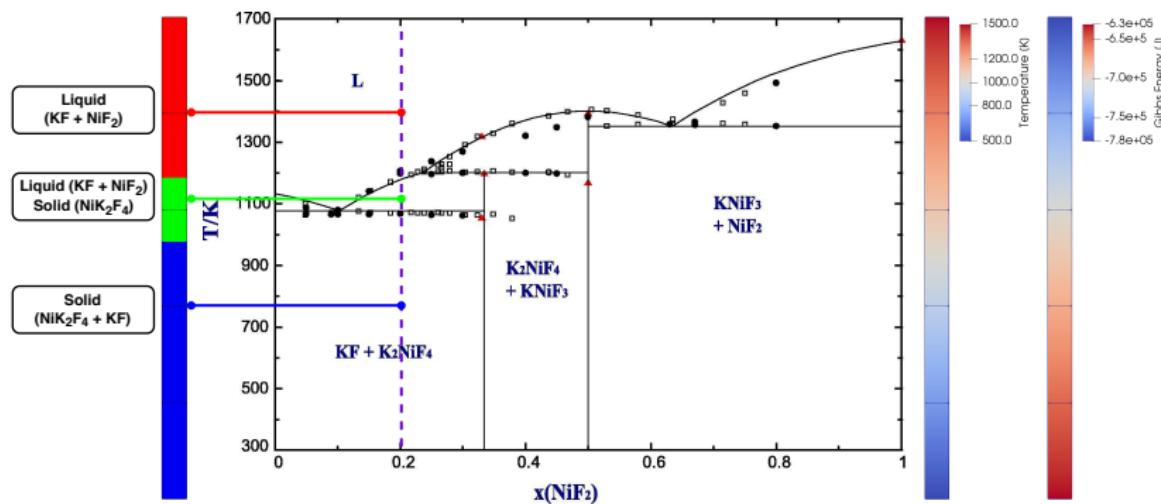


Demonstration Problem

- Initial focus on Ni alloys interacting with molten $\text{LiF}_2 - \text{KF}$ salts.



Demonstration Problem



Timeline - Coursework

Coursework			
Item	Timeline	Status	
MCSC-6010G: Mathematical Modelling	Sep. - Dec. 2018	Complete	
MCSC-6030G: High Performance Computing	Sep. - Dec. 2018	Complete	
NUCL-6005G: Computational Thermodynamics [PhD level elective]	Sep. - Dec. 2018	Complete	
MCSC-6020G: Numerical Analysis	Sep. - Dec. 2019	Complete	

Timeline - Research

Research		
Item	Timeline	Status
Literature review of computational thermodynamics and GEM	Sep. - Dec. 2018	Complete
Implement data file parsing code	Feb. - Mar. 2019	Complete
Implement linear solver (levelling)	Apr. - Jun. 2019	Complete
Implement communication between Yellowjacket and MOOSE	Jul. - Aug. 2019	Complete
Implement non-linear solver for GEM (homogeneous)	Sep. - Dec. 2019	In progress
Implement non-linear solver for GEM (heterogeneous)	Jan. - Mar. 2020	Planned
Demonstration of non-linear solver capabilities	Mar. - May 2020	Planned
Begin integration of thermodynamic solver with Marmot	Jun. - Aug. 2020	Planned
Comparative study of global optimisation strategies	Sep. - Dec. 2020	Planned
Implementation of global optimisation algorithm	Jan. - Mar. 2021	Planned
Demonstration of global optimisation capabilities	Apr. - May 2021	Planned
Complete integration into MOOSE	Jun. - Aug. 2021	Planned
Verification and testing	Sep. - Dec. 2021	Planned

Thanks for your attention!

Questions?