

Development of a New Thermochemistry Solver for Multiphysics Simulations of Nuclear Materials

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Abstract Nuclear materials are highly complex multiscale, multiphysics systems and an effective prediction of nuclear reactor performance and safety requires simulation capabilities that tightly couple different physical phenomena. The Idaho National Laboratory's Multiphysics Object Oriented Simulation Environment (MOOSE) provides the computational foundation for performing such simulations and currently consists of the continuum scale fuel performance code Bison and the mesoscale phase field code Marmot. With the move towards advanced reactors that employ higher temperature fluids than conventional reactors, corrosion has become a problem of great interest. A new application called Yellowjacket is under development to directly couple thermodynamic equilibrium and kinetics to model corrosion in advanced reactors. As part of Yellowjacket, a thermochemistry code is being developed to perform thermochemical equilibrium calculations for a range of different materials, which is currently in its infancy. This paper describes the recent progress towards development of Yellowjacket and presents the plans for developing capabilities of practical interest to the nuclear industry.

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1 Introduction

With the rapid advancement of high-performance computing capabilities over the past decade, computational models and simulations have become ubiquitous tools for design and development of nuclear systems. Nuclear materials are highly complex multiscale, multiphysics systems and simulations help scientists and engineers overcome the experimental challenges in terms of handling radioactive materials, operating conditions, large costs, safety mitigation, etc. Furthermore, material behaviour is influenced by many different physics, such as mechanics (e.g., dislocations, cracking, stress-driven diffusion), chemistry (e.g., corrosion, oxidation, reactive transport), heat transfer (e.g. convection, melting, precipitation), etc.

As shown in fig. 1, a comprehensive understanding of nuclear materials requires models that span extremely large ranges of length and time scales [1]. By using the information from smaller scales to inform the models at larger length scales, multiscale simulation tools help in exploring phenomena which would be inaccessible using only experimental methods [2]. These tools also serve the purpose of augmenting and guiding experimental characterisation techniques, helping save both time and costs.

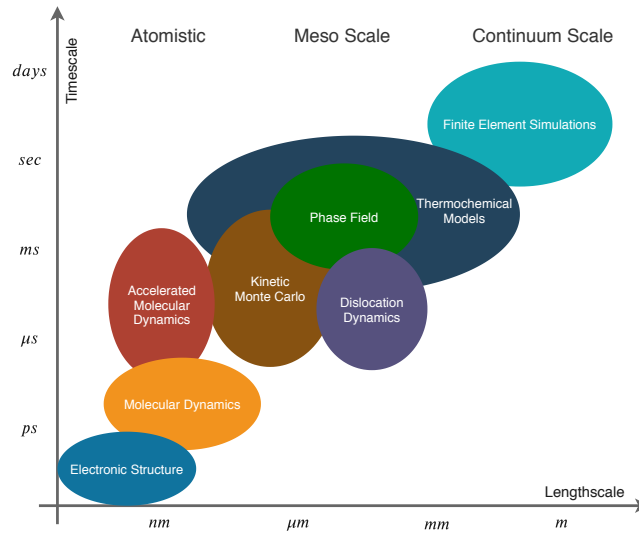


Fig. 1 Multiscale theoretical and computational methods used for materials model development and computer simulations [1].

Many emerging nuclear technologies, such as the Molten Salt Reactor (MSR), use high temperature fluids such as molten fluoride/chloride salts, which lead to increased susceptibility to corrosion of the metal containment leading to problematic behaviour during reactor operation. Corrosion is an electrochemical process composed of ox-

idation and reduction reactions, which are defined by the thermodynamics and kinetics of the reactions. While thermodynamics determines whether or not a material can corrode, kinetics controls how quickly the material will corrode. This corrosion behaviour is also significantly affected by the material microstructure and predicting corrosion therefore requires a multiphysics approach that can couple quantitative electrochemistry models of corrosion and chemical reactions with thermochemical equilibrium computations. The Multiphysics Object Oriented Simulation Environment (MOOSE) [3, 4], developed by the Idaho National Laboratory, provides a framework for multiphysics simulations but lacks the tools for predicting corrosion at the microstructure scale. A new MOOSE-based tool, *Yellowjacket*, is currently being developed to perform thermodynamics based mesoscale corrosion simulations to predict the rate of material loss, corrosion product production, and precipitate production in advanced reactors.

Since the phase and chemical behaviour of nuclear materials is governed by the thermochemical equilibrium state, coupling thermochemistry with multiphysics codes provides various material properties, source terms and boundary conditions such as the chemical potentials that act as the driving force for corrosion. Moreover, it provides the quantitative capability for predicting phase evolution as the chemical composition changes due to irradiation. It is for this reason that a thermochemical equilibrium solver is being developed as part of *Yellowjacket* and this paper describes the recent progress made in this direction.

As shown in fig. 2, by relying on the fundamental laws of thermodynamics, equilibrium computations use the system information, namely, Gibbs energy functions of the species that can be present in the system, temperature, pressure and system composition to provide quantities such as the stable phases and species, Gibbs energy of the system, chemical potentials of the species and can be used to provide information such the enthalpy, heat capacity, etc.

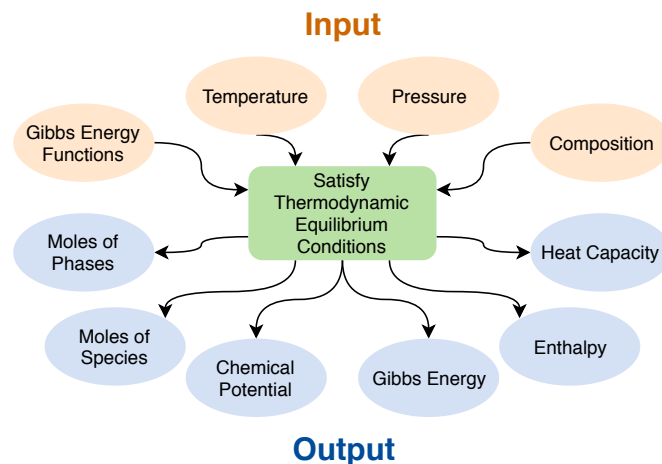


Fig. 2 Input and output parameters of thermodynamic equilibrium calculations.

2 Thermochemical Equilibrium

Thermochemical equilibrium calculations are based on minimising the integral Gibbs energy of a closed system at constant temperature and hydrostatic pressure. From a numerical point of view, the objective of computing thermochemical equilibrium is to determine a unique combination of phases and their composition that yields a global minimum in the integral Gibbs energy subject to various linear and non-linear equality constraints.

2.1 Gibbs energy

The integral Gibbs energy of a multicomponent multiphase system is represented as

$$\frac{G}{RT} = \sum_{\lambda=1}^{\Lambda} n_{\lambda} \tilde{g}_{\lambda} + \sum_{\omega=1}^{\Omega} n_{\omega} \tilde{g}_{\omega} \quad (1)$$

where, R [$\text{J mol}^{-1} \text{K}^{-1}$] is the ideal gas constant and T [K] is the absolute temperature. Λ and Ω represent the number of stable solution phases and stoichiometric phases respectively and the number of moles of the solution phase λ and stoichiometric phase ω are denoted by n_{λ} and n_{ω} [mol] respectively. Finally, \tilde{g}_{λ} and \tilde{g}_{ω} represent the dimensionless form of Gibbs energy of solution phase λ and stoichiometric phase ω respectively.

In a less prevalent but computationally more feasible form, the integral Gibbs energy can be expressed in terms of dimensionless chemical potentials¹ as follows

$$G = RT \left(\sum_{\lambda=1}^{\Lambda} n_{\lambda} \sum_{i=1}^{N_{\lambda}} x_{i(\lambda)} \tilde{\mu}_i + \sum_{\omega=1}^{\Omega} n_{\omega} \tilde{\mu}_{\omega} \right) \quad (2)$$

where, N_{λ} denotes the number of species in the solution phase λ and $x_{i(\lambda)}$ represents the mole fraction of species i in solution phase λ . $\tilde{\mu}_i$ and $\tilde{\mu}_{\omega}$ represent the dimensionless chemical potential of species i in solution phase λ and stoichiometric phase ω respectively.

The chemical potential is a measure of the change of the Gibbs energy of a system by the introduction of a substance. Mathematically, the chemical potential of a species i is defined as

$$\mu_i = \left(\frac{\partial G}{\partial n_i} \right)_{T, P, n_{j \neq i}} \quad (3)$$

¹ From a computational perspective, the method of Lagrange multipliers used to compute thermochemical equilibrium results in an objective function where the linear combinations of Lagrange multipliers give the chemical potential of elements. Furthermore, the dimensionless form of chemical potentials helps reduce the computational costs.

and for the species of an ideal phase, the chemical potential incorporates the reference Gibbs energy of pure species, $g_{i(\lambda)}^0$, and the entropic contribution due to mixing as a function of its mole fraction.

$$\tilde{\mu}_i = \tilde{g}_{i(\lambda)}^0 + \ln x_{i(\lambda)} \quad (4)$$

For non-ideal solution phases, the chemical potential also includes the partial molar excess Gibbs energy of mixing, $g_{i(\lambda)}^{ex}$, to account for non-ideal mixing

$$\tilde{\mu}_i = \tilde{g}_{i(\lambda)}^0 + \ln x_{i(\lambda)} + \tilde{g}_{i(\lambda)}^{ex} \quad (5)$$

While the chemical potential of stoichiometric phases does not include a composition dependent term, the partial molar excess Gibbs energies of mixing for non-ideal solution models depend on the mixing model employed e.g. the Modified Quasichemical Model [5, 6, 7, 8, 9], the Compound Energy Formalism [10], etc.

2.2 Conditions for thermochemical equilibrium

Achieving thermochemical equilibrium in a system requires satisfaction of both necessary and sufficient conditions which are described below.

2.2.1 Necessary conditions

1. *Conservation of mass* requires that the mass of element j , b_j , must satisfy the following mass balance equation

$$b_j = \sum_{\lambda=1}^{\Lambda} n_{\lambda} \sum_{i=1}^{N_{\lambda}} x_{i(\lambda)} \nu_{i,j} + \sum_{\omega=1}^{\Omega} n_{\omega} \nu_{\omega} \quad (6)$$

where, $\nu_{i,j}$ and ν_{ω} represent the stoichiometric coefficients of element j in solution phase species i and stoichiometric phase ω respectively and the constraints require that the mole fractions must be positive, i.e., $x_{i(\lambda)}$

2. *Gibbs' phase rule*, which defines the thermodynamic degrees of freedom of the system, must also be satisfied

$$F = C - \Phi + 2 + \Xi \quad (7)$$

where, F represents the degrees of freedom, C denotes the number of components in the system, Φ denotes the number of phases and Ξ denotes the electrochemical terms.

3. *Gibbs' criteria* for equilibrium requires that the Gibbs energy of a system be at a local minimum. In equivalent terms, the chemical potential for each system component must have the same value in all stable phases within the system [11],

where the chemical potential of any constituent in a stable phase can be defined as a linear function of the element potentials, Γ_j , as [12]

$$\mu_i = \sum_{j=1}^C \nu_{i,j} \Gamma_j \quad (8)$$

2.2.2 Sufficient conditions

The necessary conditions for thermodynamic equilibrium require that the chemical potentials of all stable solution phase species and stoichiometric phases abide by the equality given by eqn. 8, which is equivalent to the Gibbs energy of the system being at a local minimum, and that the conservation of mass and the Gibbs phase rule are satisfied.

The sufficient condition requires that all the metastable phases abide by the following conditions [13]

$$\pi_\lambda = \min_{\lambda} \sum_{i=1}^{N_\lambda} x_{i(\lambda)} \left(\mu_{i(\lambda)} - \sum_{j=1}^C \nu_{i,j} \Gamma_j \right) \quad (9)$$

i.e., there must exist a Gibbs' plane such that the element potentials lie on the plane and the chemical potentials of all the species lie on or above the plane and the mole fraction of the species must satisfy the following constraints

$$\begin{aligned} \sum_{i=1}^{N_\lambda} x_{i(\lambda)} &= 1 \\ x_{i(\lambda)} &> 0 \quad \forall i \end{aligned} \quad (10)$$

i.e., the sum of mole fraction of all the species in a phase λ must be unity and that the individual mole fractions must be greater than or equal to zero. These constraints also apply to ionic species but an additional charge balance must also be respected in such a case.

The aforementioned conditions are used in the Gibbs energy minimiser to find a unique combination of phases that are stable in the system. The computational structure of the code has been presented in the following section.

3 Gibbs Energy Minimiser

A top-level overview of the Gibbs energy minimisation code is shown in fig. 3. Calculation of thermodynamic equilibrium requires a thermodynamic database, which provides reference Gibbs energy terms of different species in addition to models

representing non-ideal behaviour. These thermodynamic databases are developed using the well established Calphad method [14]. The data file parser in Yellowjacket supports the ChemSage format data files generated by FactSage [15]. Parsing is followed by passing the system information such as temperature, pressure and elemental composition to the thermochemistry solver and at each time step, MOOSE provides these inputs for every element of the mesh.

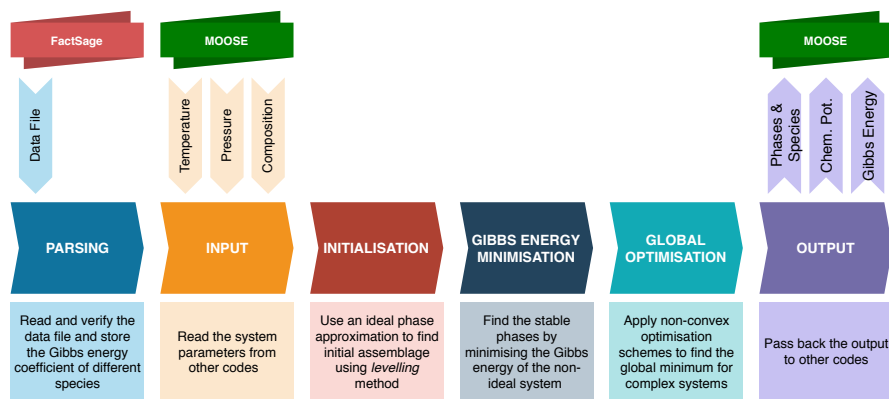


Fig. 3 Computational structure of the Gibbs energy minimisation program

The computation stage of the thermochemistry solver consists of *initialisation*, *Gibbs energy minimisation* and *global optimisation*. Thermodynamic solvers require an initial estimate of molar quantities of species and phases. To this aim, a general estimating procedure called *levelling* was developed by Eriksson and Thompson [16]. Levelling temporarily converts the non-linear optimisation problem into a linear optimisation problem with the objective of systematically adjusting fixed combinations of phases, subject to linear equality and inequality constraints, to progressively make the Gibbs energy of the system more negative, thus, convergence is accelerated by providing an estimated phase assemblage that focuses on only the dominant species. While levelling provides a great initial assemblage for further consideration, in many cases, such as the time dependent problems, other initialisation methods can be used to further reduce the computational time required by the non-linear solver [17]. A couple of these techniques have been studied by Piro [18] and include the use of assemblage from the preceding time step or from the neighbouring elements based on the principle of continuum. In future, such methods will be of significant interest to Yellowjacket and will provide significant speed gains compared to re-initialisation using levelling at each time step.

The linear solver, though an excellent initialisation algorithm, rarely provides the final assemblage of the system under consideration. Gibbs energy minimisation introduced by White *et al.* [19] is the universally used algorithm to perform the non-linear optimisation for obtaining thermochemical equilibrium in complex systems. In mathematical terms, Gibbs energy minimisation is widely performed using the

method of Lagrange multipliers that simultaneously minimises the integral Gibbs energy and the residuals of mass balance equations. This results in a system of linearised equations that can be written in matrix form as [20]

$$\mathbf{H} \cdot \boldsymbol{\pi} = \boldsymbol{\zeta} \quad (11)$$

where $\boldsymbol{\pi}$ and $\boldsymbol{\zeta}$ denote the unknown and constraint vectors respectively, and the Hessian matrix (\mathbf{H}) can be written as [19, 20]

$$\mathbf{H} = \begin{bmatrix} r_{j=1,k=1} & \dots & r_{j=1,k=C} & \phi_{j=1,\lambda=1} & \dots & \phi_{j=1,\lambda=\Lambda} & v_{j=1,\omega=1} & \dots & v_{j=1,\omega=\Omega} \\ \vdots & \ddots & \vdots & \vdots & \ddots & \vdots & \vdots & \ddots & \vdots \\ r_{j=C,k=1} & \dots & r_{j=C,k=C} & \phi_{j=C,\lambda=1} & \dots & \phi_{j=C,\lambda=\Lambda} & v_{j=C,\omega=1} & \dots & v_{j=C,\omega=\Omega} \\ \phi_{\lambda=1,j=1} & \dots & \phi_{\lambda=1,j=C} & 0 & \dots & 0 & 0 & \dots & 0 \\ \vdots & \ddots & \vdots & \vdots & \ddots & \vdots & \vdots & \ddots & \vdots \\ \phi_{\lambda=\Lambda,j=1} & \dots & \phi_{\lambda=\Lambda,j=C} & 0 & \dots & 0 & 0 & \dots & 0 \\ v_{\omega=1,j=1} & \dots & v_{\omega=1,j=C} & 0 & \dots & 0 & 0 & \dots & 0 \\ \vdots & \ddots & \vdots & \vdots & \ddots & \vdots & \vdots & \ddots & \vdots \\ v_{\omega=\Omega,j=1} & \dots & v_{\omega=\Omega,j=C} & 0 & \dots & 0 & 0 & \dots & 0 \end{bmatrix} \quad (12)$$

where $r_{j,k}$ and $\phi_{j,\lambda}$ can be expressed as follows

$$r_{j,k} = \sum_{\lambda=1}^{\Lambda} \sum_{i=1}^{N_{\lambda}} n_{i(\lambda)} v_{i,j} v_{i,k} \quad (13)$$

$$\phi_{j,\lambda} = \sum_{i=1}^{N_{\lambda}} n_{i(\lambda)} v_{i,j} \quad (14)$$

Clearly, $r_{j,k} = r_{k,j}$ and $\phi_{j,\lambda} = \phi_{\lambda,j}$, which yields a symmetric positive semidefinite matrix for \mathbf{H} .

While finding a solution to eqn. 11 is relatively easy, the challenge arises due to the change in the size of Hessian \mathbf{H} within a computation cycle. This change in size results from the addition/removal of phases in the system and requires the implementation of numerical strategies such as the ones discussed by Piro [18]. In addition, care must be taken in implementing an effective line search algorithm and proper use of Wolfe/Armijo conditions for choosing the step length can significantly improve the convergence and stability of the non-linear solver. However, for the modified quasichemical model, a hitherto unseen situation can also arise when using the chemical elements to define the system components. As shown by Piro *et al.* [21], in such systems, certain compositions can result in a rank-deficient Hessian that ultimately results in chemical potentials that might not be uniquely defined, thus, potentially misleading the users.

The Gibbs energy functions of non-ideal phases are often non-convex, yielding multiple local minima after Gibbs energy minimisation. These local minima correspond to different compositions of phases that may be believed to be stable (e.g., a

miscibility gap), which may not necessarily correspond to the true equilibrium composition. Finding the global minimum of this function among the many local minima within the domain space can be a daunting challenge, especially in large complex thermodynamic systems containing many highly non-ideal solution phases. Consequently, inadequate numerical approaches might result in false positives of thermodynamic equilibrium with little possibility identifying the divergence from the true equilibrium state [13].

While a large number of deterministic (e.g. branch and bound) and stochastic (e.g. particle swarm optimisation) methods are available in literature, none of them guarantees the ability of finding a global extremum of a non-convex function. Moreover, the computational effort associated with this task can increase very rapidly with the size of the system (i.e. the total number of species in the system). The global optimisation module is one of the thrust areas of this work and will explore various strategies to ensure that the final phase assemblage corresponds to the global minimum.

Finally, the outputs produced include the moles of the different phases present in the system, the mole fraction of the species in each phase, Gibbs energy of the system, chemical potentials, etc. and these outputs are passed to the electrochemistry module for corrosion or can be passed back to MOOSE.

4 Demonstration Problem

In this section, the current progress towards the development of the thermochemistry solver has been demonstrated through a demonstration problem. The reference fuel and coolant proposed for the different MSR concepts are fluoride salts due to their advantageous neutronic and physico-chemical properties [22]. $\text{LiF}-\text{BeF}_2$ and $\text{LiF}-\text{NaF}-\text{KF}$ are amongst the various salt compositions under study for the primary and secondary coolant and their suitability with respect to their corrosion properties for the structural material is an essential requirement for reactor safety and operation. Since Ni is the main element for prospective alloys in structural materials, and the rate of corrosion is defined by the redox potential of the molten salt, the phase equilibrium of the salt with potential corrosion products must be studied as a function of temperature and pressure [23]. Therefore, the initial focus is on Ni alloys interacting with molten $\text{LiF}-\text{KF}$ salts and for the present demonstration the $\text{KF}-\text{NiF}_2$ binary system has been selected. This system is not only relevant to the MSR but has also been studied experimentally and the required thermodynamic data is available in literature [23].

For the demonstration problem, a bar with molar composition $0.8 \text{ KF} + 0.2 \text{ NiF}_2$ under a linear temperature gradient has been solved for thermodynamic equilibrium using the linear levelling solver. The predicted stable phases and the Gibbs energies²

² Though the Gibbs energy profile shown in fig. 4 is continuous, the actual results on the finite element mesh are discretised due to a step change in the temperature as we move from one element to the other. The continuous profile has been plotted by interpolation during post processing and is

are shown in fig. 4. As expected from the phase diagram, a transition from a solid phase to a mixture of solid and liquid phase is observed around 1100 °C and a transition from mixed phase to pure liquid phase around 1200 °C.

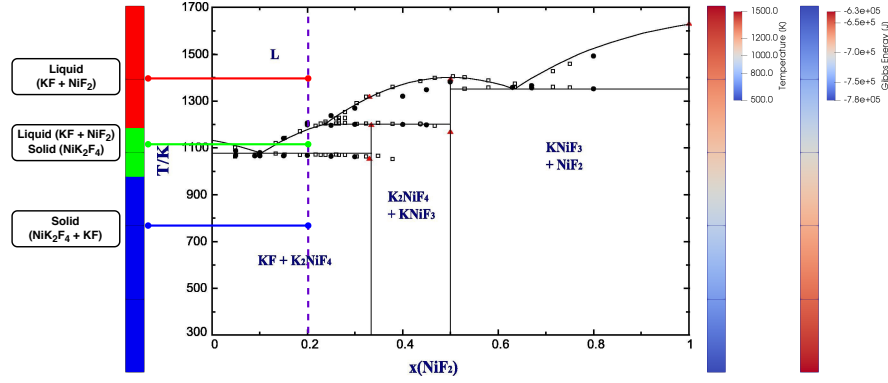


Fig. 4 Stable phases and Gibbs energies predicted by the linear solver.

5 Applications

Within Yellowjacket, the thermochemistry solver will be used for predicting microstructure evolution using phase field. Yellowjacket will use MOOSE to solve phase-field equations to describe the evolution of the chemical components and phases. The chemical components in the system are represented by conserved variables that evolve based on the Cahn-Hilliard equation as follows

$$\frac{dc}{dt} = \nabla M(c_i) \nabla \frac{\delta F}{\delta c_i} \quad (15)$$

where M is the mobility of the component, c_i represents the concentration of the component i , F denotes the free energy density of the system, and the $\frac{\delta F}{\delta c_i}$ denotes chemical potential of species i . The phases are represented as non-conserved order parameters, and are evolved using the Allen-Cahn equation

$$\frac{d\eta_j}{dt} = -L \frac{\delta F}{\delta \eta_j} \quad (16)$$

a better representation of the physical conditions where we expect no step changes if a temperature gradient is applied.

where η_j denotes the order parameter for the phase j , and L is the Allen-Cahn mobility of the interface. The order parameters are used to interpolate between the thermodynamic model for each phase.

Thus, the primary thermodynamic inputs required to the evolution equations are the Gibbs free energy of the system and chemical potentials of the species as a function of the concentrations of components and the phase order parameters. Typically, the free energy functions for each phase are obtained for the given system of components using a Calphad thermodynamic assessment before running the simulation. However, for multicomponent metal alloy systems, the free energy functions can be very complex. In such systems, having a coupled thermodynamic solver that directly provides the values from the database could help to simplify the development of the phase field models. It can simplify the complex coupled problem into two simpler problems. Values for certain derivatives within the simulation can be directly obtained from the thermodynamic solver. Eventually, coupling a thermodynamic solver with MOOSE will also allow phase-field to be operated as a black box with its own material properties database.

6 Conclusion

In summary, recent progress towards development of a new thermochemical equilibrium solver for multiphysics simulations in **Yellowjacket** has been presented. As shown through the initial demonstration problem, the thermochemistry code has been partly integrated with MOOSE and thermochemical equilibrium can now be performed on a MOOSE mesh using the linear levelling solver. While currently only the ideal mixture phases and the Modified Quasichemical Model for molten salts are supported, development is underway to expand support to other thermodynamic models. The ongoing research is now moving towards the implementation of the non-linear Gibbs energy minimiser which can provide more realistic estimates of the phase assemblage and will be followed by the implementation of various optimisation strategies to ensure convergence.

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