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Development of a Clad Activation Capability in BISON using ORIGEN

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Abstract – *This work describes the ongoing coupling work establishing ORIGEN as a physics module for depletion, activation, and decay in the MOOSE/BISON fuel performance code. The current phase has focused on the implementation of an activation model, mainly intended for evaluation and comparison of accident tolerant cladding designs within a single fuel performance framework. The next phase will develop a capability for fuel depletion. New aspects of this work include: investigation of how to allow the high-fidelity ORIGEN capability to be interoperable extension of existing lower-fidelity cladding models (e.g. based only on fast fluence), introduction of recalculation thresholds to reduce computational cost during multi-physics iterations, and runtime experiments to assess the cost of this additional physics relative to the baseline MOOSE/BISON fuel performance calculations. The current results show activation increases runtime by a factor of approximately 7, but additional optimizations, discussed in the conclusions, are plentiful.*

I. INTRODUCTION

The US Department of Energy's Nuclear Energy Advanced Modeling and Simulation (NEAMS) Program has contributed significantly to the development of the MOOSE/BISON [1] fuel performance code at Idaho National Laboratory and to the Oak Ridge Isotope Generation and Depletion Code (ORIGEN) at Oak Ridge National Laboratory [2]. This work presents a description of the initial integration of ORIGEN in BISON to perform rigorous clad activation.

In the US, the conventional non-proprietary fuel performance code used to analyze the light water reactor fleet has been FRAPCON [3], developed by Pacific Northwest National Laboratory and supported by the NRC. FRAPCON uses empirical correlations, burnup-dependent relations, and simplified geometry and physics to predict fuel performance behavior such as fission gas release, temperature distribution, and internal gas pressure. The benefit of this approach is that after proper validation, both fast and accurate solutions may be obtained.

1. Motivation

There are two basic avenues in the research and development of accident tolerant fuels [4]: fuel and cladding. Alternative fuel types, such as uranium nitrides or TRISO fuel particles increase fuel safety by providing higher thermal conductivity or more fission product barriers, respectively. Alternative clad types, such as stainless steel, iron-chromium-aluminum alloys (FeCrAl), or silicon-carbide (SiC) increase fuel safety with decreased corrosion, embrittlement, oxidation, etc.

To investigate these new fuel/clad types with FRAPCON, one must construct new fits, new correlations, and perform validation of the holistic model for both fuel and clad, undoubtedly a time-consuming process, and without the extensive experimental databases that exist for uranium oxide with zirconium-based cladding. One of the hopes with BISON is that by assuming a more first-principles approach to the physics and using a more modular and accessible representation of the underlying models (provided by the MOOSE framework [5]), the time to develop new fuel systems and optimize fuel performance and safety can be greatly reduced.

In the NEAMS program, ORIGEN has been chosen as the primary physics module for decay, activation, and depletion and as such will be coupled to BISON to model the evolution of both fuel and nonfuel materials (e.g. clad) and automatically evolve the underlying isotopics as a function of time. The fuel coupling is more complex than nonfuel, and in general requires coupling to a radiation transport module for the spatial and energy distribution of neutrons and a self-shielding module for the proper determination of multi-group cross sections, if not performing continuous-energy calculations. For this reason, the initial phase of coupling, presented in this paper, is for nonfuel.

2. Advancements

Although the fuel depletion capability is undoubtedly more impactful, the new nonfuel activation capability will does provide the ability to compare different accident tolerant cladding types, in terms of not only structural and mechanical differences, such as hoop stress and clad elongation, but isotopic differences, such as activity and neutron absorption. Zirconium-based cladding is excellent from a low neutron-interaction point of view, negligibly impacting system reactivity. Contrast this with stainless steel and FeCrAl, which can incur significant reactivity penalties or SiC which

may have a limit on irradiation due to Si degradation. With the nonfuel capability, one can evaluate these differences directly in BISON.

One of the more general benefits is the generalized geometry provided by the finite element method (FEM) framework. Fuel rods may be analyzed in one, two, or three dimensions, with detailed, time-dependent power distributions. Non-symmetric power profiles or geometry (e.g. missing pellet surface, internal pellet defects, clad thinning) can be analyzed in detail (see Fig. 1 and 2.)

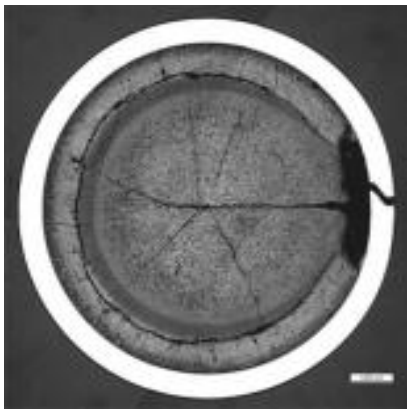

Fig. 1. Image of missing pellet interface.

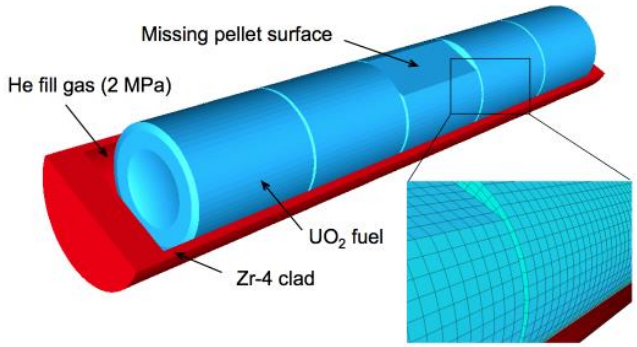

Fig. 2. Three-dimensional geometry and mesh setup.

The final, and most important class of advancements enabled, which will only be fully realized when the fuel depletion model is included, is the ability to feedback additional isotopic evolution information to BISON models. For example, BISON currently assumes a fixed number of fission gas atoms (H, Xe, Kr) produced per fission. ORIGEN could update this number directly in each mesh element based on actual calculations which consider the FEM representation of the distribution of fissioning actinides. The most groundbreaking capability will be available when ORIGEN can deliver elemental (not isotopic) distribution information to the BISON thermochemistry module, Thermochemica [6]. Thermochemica determines the most likely compounds formed for a given temperature and elemental distribution, and can determine key material properties such as density, oxidation potential, and hydride formation.

3. Overview of BISON

BISON is a finite element-based nuclear fuel performance code applicable to a variety of fuel forms including light water reactor (LWR) fuel rods, TRISO particle fuel, metallic rod, and plate fuel. It solves the fully-coupled equations of thermos-mechanics and species diffusion, for 1D spherically symmetric, 2D axisymmetric or 3D geometries. Fuel models are included to describe temperature and burnup dependent thermal properties, fission product swelling, densification, thermal and irradiation creep, fracture, and fission gas production and release. Plasticity, irradiation growth, and thermal and irradiation creep models are implemented for clad materials. Models are also available to simulate gap heat transfer, mechanical contact, and the evolution of the gap/plenum pressure with plenum volume, gas temperature, and fission gas addition.

BISON is based on the MOOSE framework and can therefore efficiently solve problems using standard workstations or very large high-performance computers. The basic equations that BISON solves are the following,

$$\rho C_p \frac{\partial T}{\partial t} + \nabla \cdot \vec{q} - e_f \dot{F} = 0, \quad (1)$$

$$\nabla \cdot \vec{\vec{\sigma}} + \rho \vec{f} = 0, \quad (2)$$

$$\frac{\partial C}{\partial t} + \nabla \cdot \vec{J} + \lambda C - S = 0, \quad (3)$$

where Eq. (1) models the temperature (T) distribution where ρ is the density, C_p is the specific heat, $\vec{q}$ is the heat flux, e_f is the energy released per fission event, and $\dot{F}$ is the fission rate; Eq. (2) models displacement (in terms of a vector $\vec{u}$) resulting from $\vec{\vec{\sigma}}$ the Cauchy stress tensor and $\vec{f}$ is the body force per unit mass; and Eq. (3) models concentration changes where C is the concentration of a species, $\vec{J}$ the mass flux, λ the decay constant, and S the source. These equations are solved using the finite element method (FEM) to perform the spatial discretization with the Jacobian-free Newton-Krylov method (JFNK) approach to iteratively converge the non-linear system and implicilty step through time.

4. Overview of ORIGEN

ORIGEN has been used to model nuclide transmutation for over 40 years. It has the capability to generate source terms for accident analyses, characterize used fuel (including activity, decay heat, radiation emission rates, and

radiotoxicity), activate structural materials, and perform fuel cycle analysis studies. This wide range of applications is possible because the guiding principle has been to explicitly simulate all decay and neutron reaction pathways using the best available data and to rigorously validate the result vs experiment. ORIGEN has been subject to hundreds of validation cases using measured data from the destructive isotopic assay of spent fuel, decay heat of spent fuel, gamma spectra resulting from burst fission, and neutron spectra resulting from spontaneous fission and *(α,n)* reactions. An active modernization has taken place over the last few years, and the ORIGEN depletion/decay module has received extensive improvement, including an application programming interface (API) for both C++ and Fortran with modern object-oriented design and various solver enhancements. The fundamental purpose of ORIGEN and the new API is to solve the following system of ordinary differential equations (ODEs) describing the depletion/decay phenomena:

$$\frac{d\vec{n}_m}{dt} = (\boldsymbol{A}_{\phi m}\Phi_m + \boldsymbol{A}_\lambda)\vec{n}_m(t) + \vec{S}_m(t), \qquad (4)$$

where

- m is a material index,
- $\vec{n}_m$ is the nuclide number density vector for material m (*atoms/barn-cm*),
- $\boldsymbol{A}_{\phi m}$ is the transition matrix for the reaction transitions (*barns* or cm^2),
- Φ_m is the scalar flux magnitude ($1/cm^2s$),
- $\boldsymbol{A}_\lambda$ is the transition matrix for decay transitions ($1/s$), and
- $\vec{S}_m$ is an external source.

The ORIGEN API solves the above system of ODEs for large systems (thousands of nuclides/tens of thousands of transitions). In traditional coupling to neutron transport codes, a quasi-static approximation is used to approximate the evolution of the system with time, where the steady-state, transport eigen-problem or fixed-source problem is solved at time *points*, (with appropriate problem-dependent self-shielding of cross sections) and the depletion equations of Eq. (4) are solved over each time *step*. In typical coupled transport/depletion simulations, a fixed amount of initial material is present in each depletable zone and the external source $\vec{S}_m(t)$ (i.e., feed of material into the volume) is zero. Fig. 3 shows a visualization of a small part of the transition matrix.

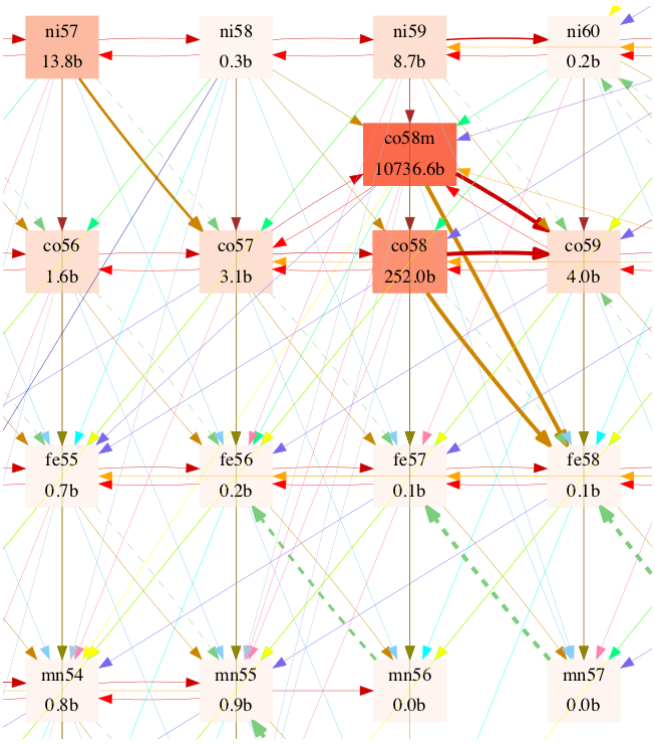

Fig. 3. Visualization of the ORIGEN transition matrix, zoomed in around isotopes in stainless steel. Representative one-group cross sections are shown under the nuclide name in the box. Reaction transitions between nuclides are shown as solid lines and decay transitions with dashed lines. The thickness of the line proportional is proportional to the probability of occurrence.

II. IMPLEMENTATION

The current implementation for nonfuel activation in BISON performs an activation calculation with ORIGEN *in every element* in the problem. In FEM terminology, the fundamental solution is in terms of nodal (or vertex) values for all coupled variables—the continuous field representation is then given in terms of these nodal values and the finite element basis functions. However, MOOSE allows for a class of variables called "Material Properties" which exist on the elements (or volumes) themselves and are periodically recalculated. There is not a single value per element, but a value at each quadrature point p within an element, which allows for a general higher-order spatial representation. See Appendix A for a more detailed description of the implementation.

To be most compatible with the existing clad models where the fast neutron fluence is used as the key irradiation parameter, in "ActivationOrigen" the step-average neutron flux magnitude at each quadrature point p, inside each element e is calculated as,

$$\bar{\Phi}_{ep,j} = \frac{1}{f_{fast}} \frac{1}{t_{j+1} - t_j} \left(\varphi_{ep}(t_{j+1}) - \varphi_{ep}(t_j) \right), \quad (5)$$

where φ_{ep} is the coupled fast neutron fluence available at beginning-of-step (t_j) and end-of-step (t_{j+1}) and f_{fast} is the fraction of the total neutron flux that is considered "fast". This factor is calculated as

$$f_{fast} = \frac{\int_{E_{cutoff}}^{\infty} \phi(E) dE}{\int_0^{\infty} \phi(E) dE}, \quad (6)$$

where E_{cutoff} is the cutoff energy and $\phi(E)$ is the neutron energy distribution. Both E_{cutoff} and $\phi(E)$ are user-provided input. MOOSE has two levels of iteration: inner linear (Krylov) iterations and outer non-linear (Newton) iterations. MOOSE material properties are recomputed every linear iteration. For the "ActivationOrigen" material, this means solution of

$$\frac{d\vec{n}_{ep}^{(\ell)}}{dt} = \left(\bar{\boldsymbol{A}}_{\phi} \bar{\Phi}_{ep,j}^{(\boldsymbol{\ell})} + \boldsymbol{A}_{\lambda} \right) \vec{n}_{ep}^{(\ell)}(t), \quad \text{over step } t_j \le t \le t_{j+1}, \quad \text{with initial condition } \vec{n}_{ep}(t_j) = \vec{n}_{ep,j}. \quad (7)$$

for every quadrature point p of every element e, at every iteration ℓ. Although each solution of Eq. (7) takes only milliseconds, assuming a single time-step requires approximately ten non-linear iterations and each non-linear iteration approximately ten linear iterations, this problem would be re-solved 100 times per time step. Moreover, in many BISON problems, the average flux over an interval is fixed, $\bar{\Phi}_{ep,j}^{(\boldsymbol{\ell})} = \bar{\Phi}_{ep,j}$, because the fast fluence is a fixed function defined in the input.

To avoid unnecessary solves, the following condition must be met in order to actually update the isotopics for element e quadrature point p,

$$\left| \frac{\bar{\Phi}_{ep,j}^{(\ell)}}{\bar{\Phi}_{ep,j}^{(last)}} - 1 \right| > \epsilon_{\phi}, \quad (8)$$

where $\bar{\Phi}_{ep,j}^{(last)}$ is the last magnitude to the flux used for an activation calculation at this element e, quadrature point p over time step j. Future improvements could analyze flux spectrum changes as well, but the current implementation assumes a fixed neutron flux energy spectrum $\phi(E)$ provided in multi-group form for the entire problem in the input, which leads to a fixed reaction portion of the transition matrix, $\bar{\boldsymbol{A}}_{\phi}$.

III. RESULTS

The test problem is a PWR rodlet with 10 smeared pellets using BISON's smeared pellet model in 2D RZ geometry. The mesh is shown in Fig. 4 for a slice of the fuel rod where the left is the center and the right is the zirconium-based clad and the "empty" space at the top and bottom is the additional plenum allowed for fuel pellet expansion and fission gas accumulation. The simulation time is approximately 930 days. Hot full power is assumed for the entire simulation with a center-peaked axial power shape. Standard results in terms of beginning, middle, and end-of-life (BOL, MOL, EOL) obtained with BISON are shown in Fig. 5 for the fission rate (#/sec) and temperature distribution (K). See Appendix B for the relevant portion of BISON input which controls the ActivationOrigen calculation.

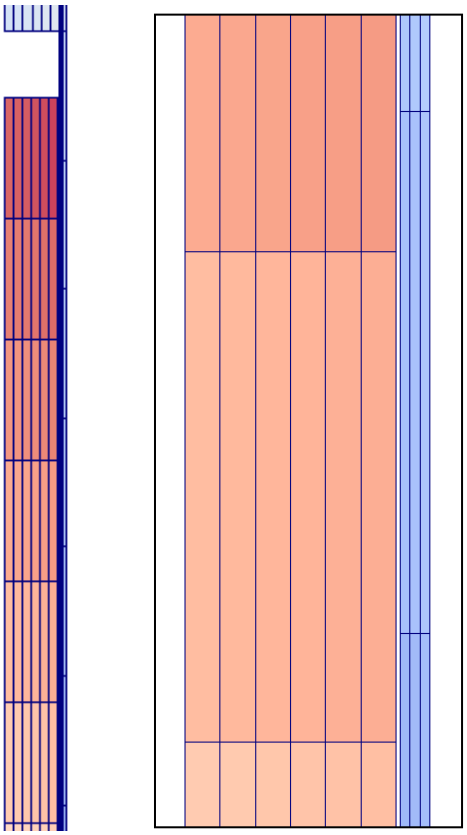

Fig. 4. 5 pellets and upper plenum of the rodlet mesh (left) and zoom-in showing the clad mesh in blue and fuel mesh in red (right).

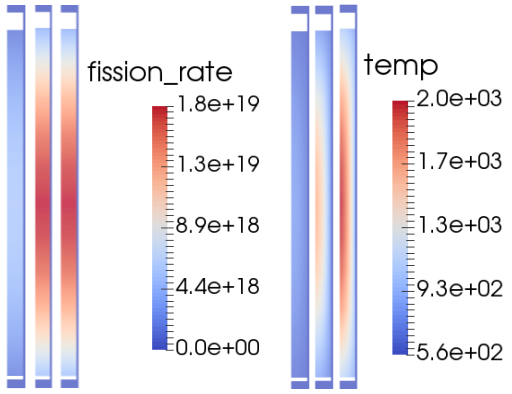

Fig. 5. The specified fission rate distribution (#/sec) evolution from BOL to MOL to EOL (left) and calculated temperature (K) distribution evolution (right).

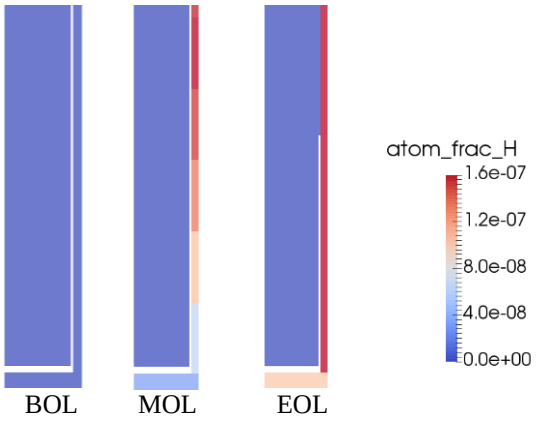

Fig. 6. Evolution of hydrogen atom fraction in the cladding and other hardware in the bottom half of the rodlet.

New results obtained with ORIGEN include the isotopic or elemental composition at any time in the clad because of neutron activation. For the purposes of coupling to other physics, e.g. thermochemistry and clad structure models, it is the elemental composition that is most important.

In Fig. 6, the hydrogen atom fraction in the clad is shown. After the approximately 900 days of this irradiation campaign a little less than 0.2 ppm has been created in the clad. This hydrogen is mainly due to protons emitted during proton-producing reactions. Modeling the amount of hydrogen or other gases in cladding is important to assess the feasibility of some designs, e.g. a cladding design with interspersed boron for reactivity control may create too much helium to ensure rod integrity.

Some other elements of interest are shown in Fig. 7. For example, 98% of the rod is composed of Zirconium, with very little change during irradiation due to the main Zirconium isotope's relative neutron transparency. Trace elements in Zircaloy (the actual cladding material used), such as Sn, Nb, Y, and Hf have isotopes with larger cross sections and are more likely to activate. Due to alpha-producing reactions and decay, helium builds up in the cladding preferentially where the flux is strongest, reaching a level of about 0.1 ppm by EOL

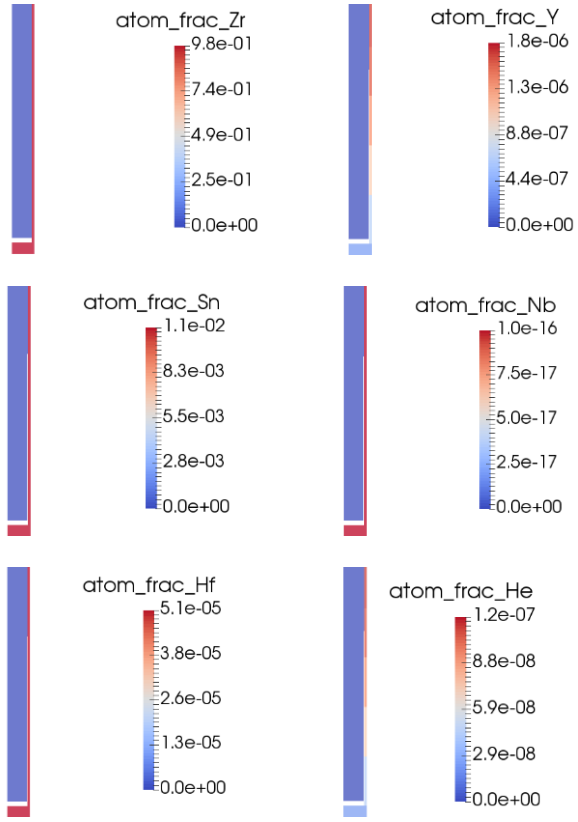

Fig. 7. EOL atom fractions of other elements in the cladding and other hardware in the bottom half of the rodlet.

Fig. 8 and 9 show normalized distributions of elements along the clad. Fig. 8 contains elements present in the clad at the beginning of irradiation. Tin (Sn) shows the largest loss and interestingly hafnium (Hf) shows the only curvature, being depleted more readily in the high-flux region at the center of the rod.

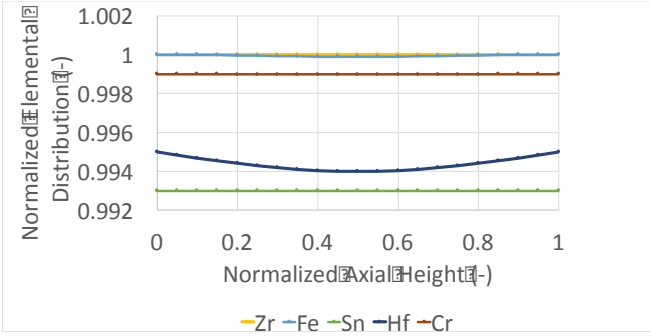

Fig. 8. Axial distribution of elements in the clad existing initially. Results are normalized to the initial atom fraction for each element.

Fig. 9 contains elements that are produced during irradiation, with each element's spatial distribution normalized to the maximum. All elements have the maximum concentration in the center where flux is highest, as would be expected.

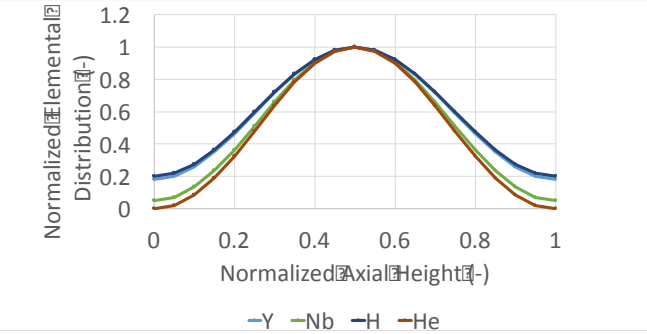

Fig. 9. Axial distribution of elements in the clad not existing initially. Results are normalized to the final atom fraction for each element.

Table I shows the absolute atom fractions of all elements present at EOL at a level of one part per billion (1e-9) or greater. Table II shows the runtimes for three variants of calculations. The first, "no activation" is simply running BISON without ORIGEN. The second, "naïve activation" is running ORIGEN every single linear and non-linear update. This increases the runtime by a factor of 170. The third is with introduction of a simple threshold for recalculation, which even if set to zero, $\epsilon_\phi = 0$, results in a factor of 24 speedup in this case, but still a runtime penalty factor of 7.1, i.e. a 710% increase in runtime if activating clad.

Table I. Elemental atom fraction at beginning and end of irradiation in the clad.

Element	**BOL Atom Fraction**	**EOL Atom Fraction**
H	0.00E+00	2.82E-07
He	0.00E+00	1.81E-07
V	0.00E+00	5.57E-08
Cr	1.76E-03	1.76E-03
Fe	3.43E-03	3.43E-03
Sr	0.00E+00	1.72E-07
Y	0.00E+00	8.53E-08
Zr	9.86E-01	9.86E-01
Nb	0.00E+00	2.10E-06
Mo	0.00E+00	1.40E-04
In	0.00E+00	1.05E-06
Sn	1.11E-02	1.11E-02
Sb	0.00E+00	4.50E-06
Te	0.00E+00	3.37E-07
Hf	5.12E-05	5.10E-05
Ta	0.00E+00	1.87E-07

Table II. Runtimes for 16-core parallel calculations.

Case	**Runtime (s)**	**Penalty Factor (-)**
no activation	4.2	0
naïve activation	720	170
activation with $\epsilon_\phi = 0$	34	7.1

IV. CONCLUSIONS

In multi-physics fuel performance, there must be a capability to evolve the isotopics of not only the fuel but the nonfuel as well (e.g. clad and structural components) to feed back into the chemical evolution and behavior of the system. This work presented progress on coupling the ORIGEN API for decay, depletion, and activation with the BISON multi-physics fuel performance code in the MOOSE finite element framework. The current phase is simply to implement nonfuel activation, which could be useful for evaluating accident tolerant cladding options within a single code.

A simple test case to verify the coupling and determine the approximate runtime cost was performed and showed that with a simple threshold on recalculation, the cost could be reduced greatly. We have established a goal to reduce the cost of depletion and activation physics to approximately the cost of the BISON calculation, i.e. a penalty factor of 1.0 instead of the current 7.1. This goal can only be achieved by complexity reductions of the nuclides and transitions considered for a particular system. This must occur at runtime, i.e. based on the input composition of a clad, and for cladding, some numerical experiments have shown that the cost can be reduced immensely, by a factor of 20 to 100, with no loss in accuracy, simply by discarding unnecessary nuclides and transitions. For fuel systems, the existence of fission products make it much more difficult and the same type of reduction only yields a factor of two or so decrease. Therefore, to further bridge the gap for fuel calculations in particular, we anticipate needing to employ a coarser mesh for the isotopic distributions and using FEM to project elemental distributions onto the meshes for BISON and Thermochemica models.

APPENDIX A: Some Implementation Details

To those with experience implementing or coupling with MOOSE or BISON, the following information may be useful. MOOSE and therefore BISON allow modules as "Kernels" or "AuxKernels" to be defined which live on the usual nodes/vertices in a finite element framework. However the ORIGEN solution at each node is an isotopic vector of size ~2200, and MOOSE does not support declaration of vector variables. For vector (or tensor) variables, each element must be declared separately. For example, with the displacement vector $\vec{u}$ in two dimensions, one must declare MOOSE variables "disp_x" and "disp_y" and explicitly connect the proper "Kernels" (basically each of the terms appearing in Eq. (1), (2), or (3) is a "Kernel") to these variables. Requiring a user to setup a variable for each of 2200 isotopes should be avoided for usability.

This led to the choice to use a "Material" in the current activation model (and eventually depletion) where that material owns a "Material Property" for the isotopics vector, which is basically allowed to be any C++ type, in this case a vector of double precision reals (i.e. std::vector<double>). Material properties are not declared in the input file, but internally to the code when certain "Materials" are declared in the input file. "Materials" are used to provide the coefficients in equations, e.g. C_p in Eq. (1), which allows for non-linearity ($C_p = C_p(T)$) or dependence on another variable or material property (e.g. $C_p = C_p(\varphi)$ where φ is the fast neutron fluence).

Thus the easiest and most user-friendly way to include the isotopic distribution for the purpose of clad activation was through a new activation-oriented material model, named "ActivationOrigen". The name was chosen with prefix "Activation" according to the BISON convention that a material begins with the type, e.g. "Thermal" or "Mech" for thermal and mechanical material models, respectively. The convention is then to follow with the specific material name, e.g. "ThermalFeCrAl" for a thermal behavior model of Fe/Cr/Al alloys. The name of the depletion module will follow suite with the name "DepletionOrigen".

APPENDIX B: Example BISON Input for the ActivationOrigen Model

```
[Materials]
  [./clad_activation]
    ############################
    # Activation with ORIGEN #
    ############################
    type=ActivationOrigen

    ## Neutron energy spectrum bounds from
    ## high to low (J)
    ## followed by distribution of neutrons.
    neutron_energy_spectrum_bounds =
    '3.2E-12 1.6E-12 1.6E-13 1.6E-14 1.6E-24'
    neutron_energy_spectrum =
    '      0.1     0.2     0.1     1.7      '

    ## Minimum energy of "fast" neutrons (J)
    fast_neutron_cutoff_energy = 1.6E-13

    ## Step-average flux is calculated from
    ## fast fluence, cutoff energy
    ## and neutron energy distribution
    fast_neutron_fluence =
fast_neutron_fluence

    ## Absolute path to Origen resources
    transition_definition_file =
'/abspath/origen_library/pwr.rev01.orglib'
    reaction_resource_file =
'/abspath/origen.rev01.jeff56g'
    yield_resource_file =
'/abspath/origen_data/origen.rev05.yields.data
'

    ## apply activation to this block of mesh
    block = clad

    ## Initial composition (Zircaloy-4)
    initial_atom_frac_Cr = 1.76E-03 #Z=24
    initial_atom_frac_Fe = 3.43E-03 #Z=26
    initial_atom_frac_Zr = 0.9836   #Z=40
    initial_atom_frac_Sn = 1.11E-02 #Z=50
    initial_atom_frac_Hf = 5.12E-05 #Z=72

    ## Additional elements to track
    initial_atom_frac_H  = 0
    initial_atom_frac_He = 0
    initial_atom_frac_Y  = 0 #Z=39
    initial_atom_frac_Nb = 0 #Z=41

    ## computational parameter to limit
    ## re-evaluations
    minimum_relative_flux_change = 1.0E-6
  [../]

  #additional materials

[../] #end of Materials
```

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