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Title: Science Based Integrated Approach to Advanced Nuclear Fuel Development – Integrated Multi-Scale Multi-Physics Hierarchical Modeling and Simulation Framework Part II: Cladding

Author(s): C.N. Tomé (1), A. Arsenlis (2), J.A. Caro (1), R.A. Lebensohn (1), J. Marian (2), C. Unal (3), K. Pasamehmetoglu (4)

(1) MST Division – Los Alamos National Laboratory
(2) Lawrence Livermore National Laboratory
(3) CCS Division – Los Alamos National Laboratory
(4) Nuclear Fuels and Materials Division, Idaho National

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Science Based Integrated Approach to Advanced Nuclear Fuel Development – Integrated Multi-Scale Multi-Physics Hierarchical Modeling and Simulation Framework *Part III: Cladding*

C.N. Tomé [1], A. Arsenlis [2], J.A. Caro [1], R.A. Lebensohn [1], J. Marian [2], C. Unal [3], K. Pasamehmetoglu [4]

[1] MST Division – Los Alamos National Laboratory
[2] Lawrence Livermore National Laboratory
[3] CCS Division – Los Alamos National Laboratory
[4] Nuclear Fuels and Materials Division, Idaho National Laboratory

Abstract

Advancing the performance of Light Water Reactors, Advanced Nuclear Fuel Cycles, and Advanced Reactors, such as the Next Generation Nuclear Power Plants, requires enhancing our fundamental understanding of fuel and materials behavior under irradiation. The capability to accurately model the nuclear fuel systems to develop predictive tools is critical. Not only are fabrication and performance models needed to understand specific aspects of the nuclear fuel, fully coupled fuel simulation codes are required to achieve licensing of specific nuclear fuel designs for operation. The backbone of these code, models, and simulations is a fundamental understanding and predictive capability for simulating the phase and microstructural behavior of the nuclear fuel system materials and matrices. In this paper we review the current status of the advanced modeling and simulation of nuclear reactor cladding, with emphasis on what is available and what is to be developed in each scale of the project, how we propose to pass information from one scale to the next, and what experimental information is required for benchmarking and advancing the modeling at each scale level.

1- Introduction

The logical process for designing any technological component is to first determine the operational conditions under which such component will be used, select the material with the right properties, and decide optimal dimensions and manufacturing process for fabricating it. The historical approach to materials and components development has usually been the empirical 'cook and look' approach. Such approach is intrinsically inefficient and costly, particularly in the case of nuclear reactor components, which are supposed to have a lifetime of several decades, and so assessing their performance is a lengthy process. As a result, the introduction of new materials with improved performance characteristics in nuclear environments has been difficult to realize.

A new paradigm focuses on developing a predictive capability that would enable the simulation of the performance of reactor components for arbitrary histories of operating conditions. Such an approach needs to account for the atomistic and micro-structural features of the material, and connect such length scales with the macroscopic response of

the full component. A way to achieve this is to utilize a **Multi-Scale Multi-Physics Modeling Approach** to understand and model the macroscopic response of cladding subjected to irradiation, temperature and mechanical conditions present in nuclear reactors, and this document proposes a strategy for developing such approach. While the strategy itself is not specific to a particular material or alloy, the microstructural details may be unique to a particular material. As a consequence, the models and numerical codes to be used at each length scale will have to be tailored to each particular material. In this document we will focus on examples for both, austenitic and ferritic stainless steels.

At a microscopic level, neutron irradiation produces vacancies, interstitials and He atoms inside the crystallographic grains of cladding. These defects may diffuse to create voids, bubbles, prismatic dislocation loops; may produce climb of pre-existing dislocation structure; and may induce local changes in composition and phases via radiation induced segregation, etc. The accumulation of these defects affects dislocation glide, leading to irradiation induced hardening.

The macroscopic evidence of these atomistic processes upon cladding is: **swelling** (volumetric dimensional change), **irradiation and thermal creep** (deviatoric dimensional change under imposed stress lower than the yield stress), increased yield strength - **hardening** - when cladding is subjected to mechanical deformation (such as upon contact from fuel, or because of abnormal in-reactor conditions, such as bending or torsion), embrittlement in the case of bcc-based alloys, and **fracture**.

The basis of our Multi Scale Multi Physics (MSMP) paradigm is to build up a chain of models spanning from the electronic/atomic scale to the actual reactor component scale, in order to describe the macroscopic response of cladding in terms of the physical mechanisms that operate at much lower scales. A strategy for using a MSMP paradigm is to identify the physically relevant mechanisms and associated parameters required by the upper scale models and extract those from the lower scale models. With this top-down approach, the appropriate sub-scale physics simulation tool is identified and, if input to this sub-scale cannot be specified by experimental data, then lower length scale simulations are employed to provide that information in a descending algorithm that may eventually reach the electronic structure methods.

In this paper we concentrate on the MSMP associated with **creep, strength and swelling.** We omit fracture and with chemistry-related effects, such as oxidation or corrosion.

The techniques being used for modeling irradiation effects on cladding materials are each characteristic of a given length scale. They are, from low to high scale, Electronic Structure (ES), Molecular Dynamics (MD), Molecular Statics (MS), kinetic Monte-Carlo (kMC), Dislocation Dynamics (DD), Single Crystal Dislocation Hardening (dislocation-density-based models) models (SXM), Polycrystal Models (PXM), Continuum Constitutive Laws (CCL). Each of these areas is presently independently well established : tested codes and techniques are operational, and highly qualified human resources are working on each of them. The challenge for the near and mid-term future is

to integrate them in a Multi Scale Multi Physics approach. Proposing how to do so is the subject of this paper.

In what follows, we summarily describe each scale and the associated modeling techniques. We also identify experiments associated with each scale, and propose how to pass modeling information between scales in order to develop an integrated model. Although the predictive capability of the integrated model flows from the lower to the higher scale, it is not necessary to develop each 'scale' component in sequence. Each 'scale module' can be developed and improved separately and, until it is 'inserted' into the comprehensive model, more empirical parameters can be used at each scale. The flow chart below provides a visual connection between the modeling components.

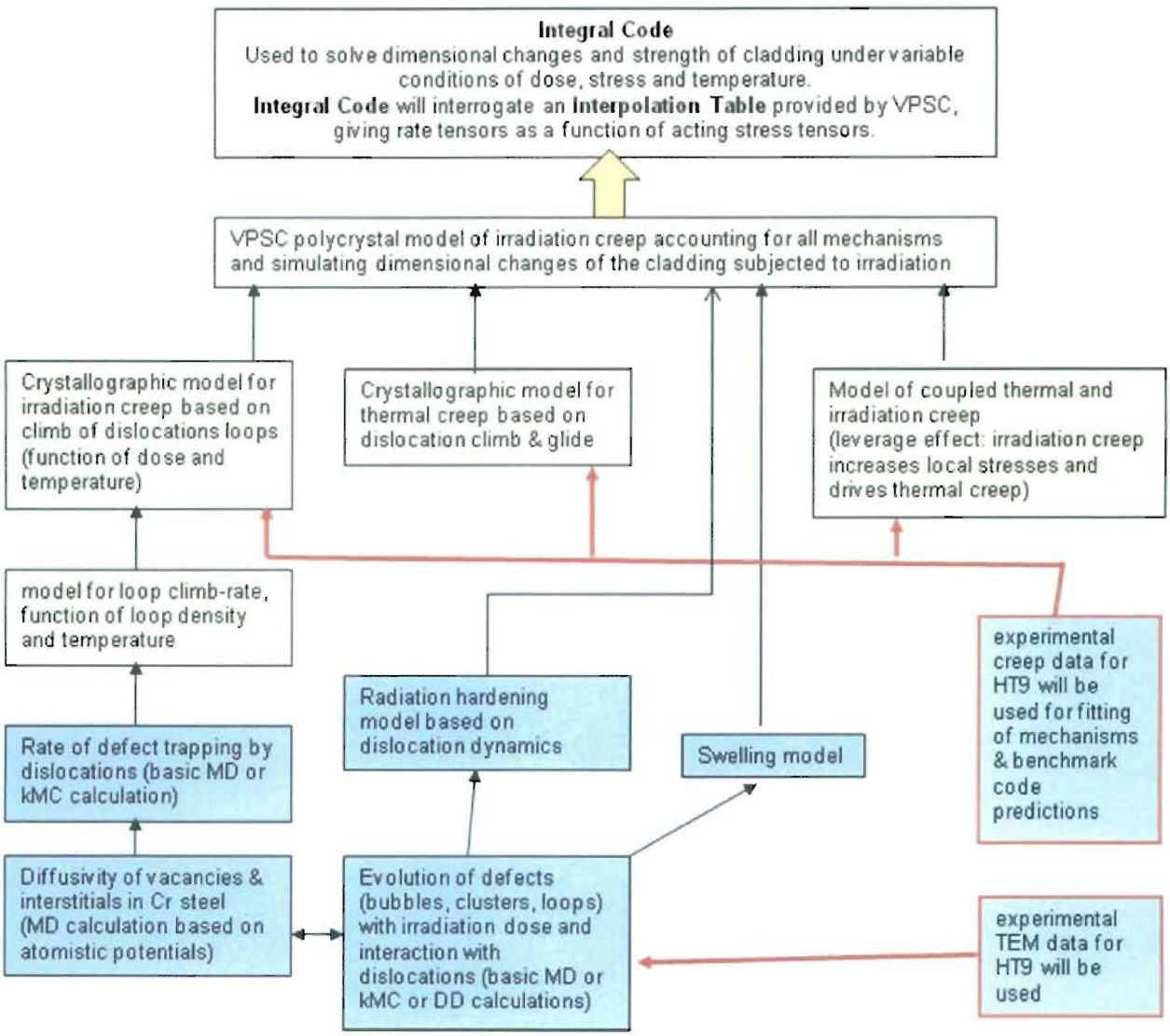

Figure 1: *Schematic flow chart illustrating the connection and information-path between the different scales and models that compose the Integrated Model for Cladding*

2- Integrated Model Components

2.1 - Continuum Constitutive Law (CCL)

The CCL will be used as the material subroutine in Finite Element codes to simulate mechanical response of cladding associated with time dependent temperature and irradiation dose. The CCL will account for local dependence of mechanical properties (swelling, creep) upon temperature and irradiation dose. It will also account for local evolution of the microstructure (i.e.: dislocation loop and void's density and size). CCL will provide dimensional changes of cladding with time due to creep and swelling under the aforementioned conditions. CCLs should also address embrittlement of cladding under those conditions.

The constitutive response will be a function of the stress applied, temperature and dose. These conditions will be imposed on the cladding by the FE simulation code, which accounts for temperature gradients in the clad, heat transfer, and mechanical solicitations such as pressure, gravitational load, contact between cladding and fuel. The material response will be specific of the alloy being modeled, and of its initial microstructure (dislocation density, texture, etc).

We foresee that such information will be passed by the next modeling scale, the Polycrystal Model, in the form of a look-up Table, as is illustrated in the example below, or using some other numerical strategy.

Current status:

As an example, a constitutive law for creep of ferritic SS HT9, based on the polycrystal model to be presented in the next section, was implemented in the form of a Numerical Interpolation Table. The Interpolation Table contains a list of stress states and the associated creep rates. Since stress Σ and creep rate $\dot{E}$ are symmetric tensors, and since creep is independent of the hydrostatic stress component and non-dilatational, only 5 out of the 9 tensor components are independent. As a consequence, we produce a Table of stress states equally spaced in this 5-dim space, together with the corresponding creep rates calculated with the Polycrystal Model.

Given an arbitrary stress state, we obtain the creep rate associated by interpolating numerically between the closest known points in the Interpolation Table. The Numerical Interpolation approach was used to predict creep rate of cladding tube of Cr SS HT9 subjected to combined pressure and torsion solicitation (Fig 2). (see Tomé and Lebensohn, 2009). A pressure of P=10MPa is assumed and a thickness radious ratio R/t=10. The axial and hoop stresses are 50 and 100MPa, respectively. A variable torsion stress is superimposed $\sigma_{12} = \alpha\sigma_{22}$. The three non-zero components of the creep rate tensor, given by the Interpolation Table, are plot in Fig. 2 as a function of the parameter α. Observe that the presence of the shear component couples with and changes the creep rate in the hoop and axial directions.

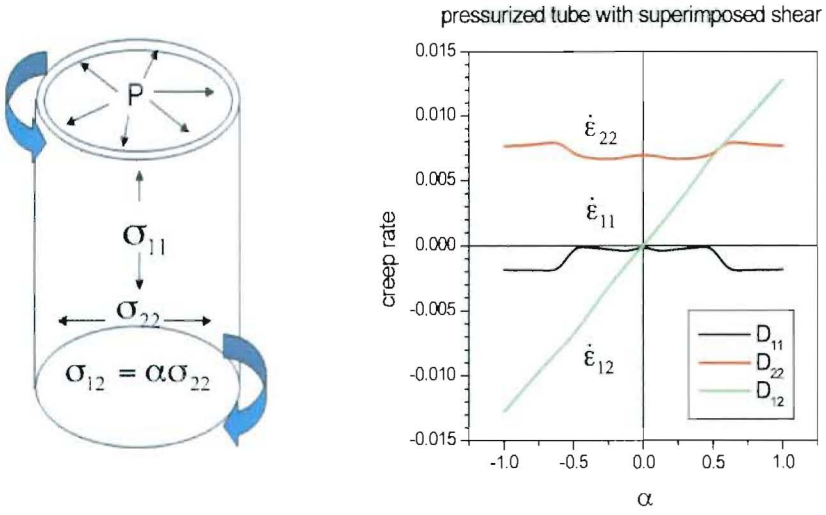

Figure 2: *Creep rates associated with superimposed pressure and shear. Tube reference axes: 1=Axial, 2=Hoop, 3=Radial. The stress state applied is of the form σ = (50, 100, 0, 0, 0, α*100) MPa*

Challenges:

The Interpolation Table approach, as any other approach based on pre-calculated data, requires one to cover beforehand all foreseeable conditions of temperature, irradiation dose, and microstructure evolution. Such procedure may be feasible when the universe of possible situations is limited. Otherwise, this approach may be unfeasible, and a direct interfacing between the FE and Polycrystal Code may be the only option. The challenge, in this case, is to develop an efficient computational interface.

2.2 Polycrystal Model (PXM)

The PXM represents the material as a collection of interacting grains using an effective medium approach. The grains are represented by crystals with orientations and weights (volume fractions) chosen to reproduce the cladding tube texture. The grains are treated as ellipsoidal inclusions interacting with the effective medium, and the latter has the macroscopic properties of the polycrystal. A total of 2916 crystal orientations (grains) are used to represent the texture of Cr SS HT9 used in the calculations (see Fig 3).

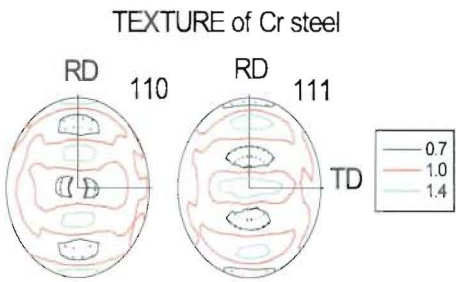

Figure 3: *(110) and (111) pole figures of SS HT9 used in this work, represented by 2916 weighted orientations.*

Because the response of the aggregate is given by the weighted average of the individual grain response, the anisotropic creep response of the clad is a direct consequence of the model. And because the grain response is based on the crystallographic mechanisms responsible for deformation, there is a direct connection between macroscopic response and crystallographic mechanism such as loops, dislocation slip planes and dislocation densities, grain boundaries, etc.
The information about dislocation slip and climb mechanisms, threshold stress to activate those mechanisms, rate of growth of voids, etc, is provided by the SXM modeling scale.

Current status:
The VPSC (Visco-Plastic Self-Consistent) code was extended to include deformation by dislocation climb, in addition to deformation by dislocation glide (Lebensohn et al, 2010). Each of these terms is driven by a power of the stress present in each grain: linear for climb, non-linear for glide. The stress in the grain follows from solving the interaction between the grain and the effective medium. The average properties of the effective medium are unknown and are solved in a Self-Consistent way.
The model is used to calculate circumferential creep rates in a cladding tube subjected to internal pressure. The creep parameters used are reported in Section 2.3, and are chosen empirically to provide right orders of magnitude but they are not representative of SS HT9. Representative parameters will be obtained in the future by comparison of predictions with specific experiments, and by the lower scales of modeling described below. Still, this simplified case shows some general results and model capabilities. At low stress (low pressure) irradiation creep dominates (black line in Fig.4) and at high stress (high pressure) thermal creep dominates (red line). The linear superposition of the two contributions (green line) does not differ from the case were they are solved in a coupled manner (blue line). This result may help us simplify the modeling approach.

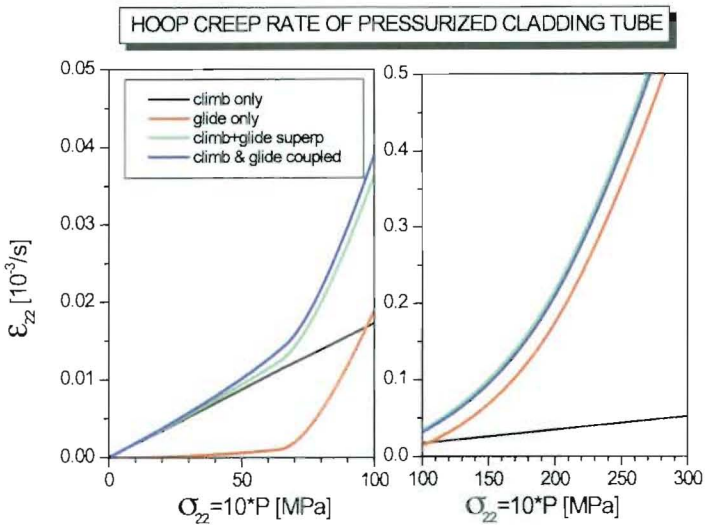

Figure 4: *Hoop creep rate* $\dot{\varepsilon}_{22}$ *of a pressurized cladding tube SS T19 as a function of hoop stress* $\sigma_{22} = R/t * P$. *Observe that irradiation creep (black line) prevails at low pressure, and thermal creep component dominates at high pressure. The reason is explained in Section 2.3.*

Experiments required:

The experiments that will help benchmark and develop the polycrystal models will also benefit the development of the lower modeling scales. Specifically, these experiments should consist on creep measurements and strength measurements.

The creep measurement will consist of in-reactor measurements of pressurized tubes and stress relaxation of bent plates. Variables will be pressure, temperature and dose.

The strength measurements will consist on tensile tests done on pre-irradiated specimens. Ex-situ thermal creep experiments will be done on as-received and pre-irradiated tensile samples, to assess contribution of thermal creep separately from irradiation creep.

Challenges:

Polycrystal models have reached maturity in the last decade, and they can even incorporate intragranular stress fluctuations and mixed visco-plastic mechanisms (Lebensohn et al, 2007). The challenges will be on the treatment of the single crystal constitutive response under irradiation, discussed in Section 2.3.

3- Single Crystal Model (SXM)

Within the SXM the grain is represented as a crystal having a given orientation and shape with respect to the sample axes (cladding tube). Deformation of the grain takes place by means of crystallographic mechanisms: dislocation emission and glide on their specific glide planes, induced by strain applied to the crystal; climb of dislocation loops and climb of edge component of forest dislocations (creep), induced by applied stress; swelling, induced by creation and agglomeration of irradiation-produced defects.

We use as a platform the Visco-Plastic Self-Consistent VPSC polycrystal code of Lebensohn and Tomé (1993). This code was extended as part of our Cladding Simulation program, and a dislocation climb term was added to the dislocation glide term (Lebensohn et al, 2010). Both are driven by a power of the stress present in each grain and the basic equations that describe strain rate and creep rate tensors as a sum over single systems contributions are:

$$\dot{\varepsilon}_{ij}^{glide} = \dot{\gamma}_0 \sum_s m_{ij}^s \left(\frac{m_{kl}^s \sigma'_{kl}}{\tau_o^{glide}} \right)^{n_g} \qquad (1a)$$

$$\dot{\varepsilon}_{ij}^{climb} = \dot{\gamma}_0 \sum_s c_{ij}^s \left(\frac{c_{kl}^s \sigma_{kl} + \tau^{chem}}{\tau_o^{climb}} \right)^{n_c} \qquad (1b)$$

where Eq (1a) is the classical rate-sensitivity equation for single crystals deforming by glide (which involves the Schmid tensor m_{ij}^s describing the glide geometry of every slip system, a threshold stress for glide τ_0^g and the glide exponent n_g), and Eq (1b) is the analog constitutive description of climb. As its glide counterpart, the climb tensor k_{ij}^s describes the climb geometry, but, since only the edge component of a dislocation can climb, it also depends of the character of the mobile dislocations in the crystal. This latter edge/screw ratio parameter and its evolution will be estimated by the Dislocation Dynamics component of the Program. The chemical driving force in Eq. (1b), τ^{chem} is empirically added to the Peach-Koeler force, but in reality there is plenty of physics behind this parameter. When only thermal creep is considered, it represents a drag effect coming from the local non-equilibrium concentration of vacancies in the vicinity of climbing dislocations. And if one assumes that vacancies are instantaneously produced or annihilated by dislocations and that the thermal equilibrium concentration is kept constant everywhere, there is no drag effect and $\tau^{chem} = 0$ in Eq.(1b).

In reality, the fact that vacancies can be emitted and absorbed at finite rates constrains the resulting thermal creep rates. Under-irradiation creep, on the other hand, vacancies and interstitials are created at the same rate and are supersaturated in the crystal. In this case, dislocation climb is determined by the absorption and not the emission rate of defects, and the threshold stress τ_0^{climb} will depend on the type of defect (vacancy or interstitial) and the sense of the stress applied on the dislocation. Such complex defect dynamics should be estimated by lower-length scale atomistic models, and the results may be passed to the crystal scale models by means of rate equations. Swelling should be the consequence of interstitials being absorbed by dislocations at higher rates than vacancies, and of dislocations reacting with each other. Including these mechanisms in the SX model will allow us to predict the dependence of swelling-rates with grain orientation.

Current status:

The stress follows from solving the interaction between the grain and the effective medium (PXM). In both cases a threshold stress τ_o has to be overcome for the mechanism to be activated. Presently this is an adjustable parameter, but there is a strong connection between τ_o and the physical mechanisms involved in climb and glide. As for the drag effect, in the calculation that follows we assume that vacancies are instantaneously produced or annihilated by dislocations and that the thermal equilibrium concentration is kept constant everywhere. As a consequence, $\tau^{chem} = 0$ in Eq. (1b). The parameters in Eqs. 1a and 1b which were used to perform the simulations reported in Figs. 2 and 4, are:

$$\dot{\gamma}_0^{gl} = \dot{\gamma}_0^{cl} = 10^{-4}\,s^{-1}$$
$$\tau_o^{glide} = 100MPa \quad ; \quad n_{gl} = 4$$
$$\tau_o^{climb} = 10MPa \quad ; \quad n_{cl} = 1$$

However, in future work τ_o these functionals and parameters will be modified by understanding gained from lower length scale simulations. For example, explicit dependence on temperature for both climb and glide will be added, the climb kinetics will be dependent on the supersaturation of point defects that are created during irradiation. The resistance to dislocation glide will depend on the current irradiation microstructure, and the strain hardening characteristics will depend on the coevolving dislocation microstructure and irradiation induced microstructure during deformation.
An example of the connection with irradiation induced defects is the adaptation of the VPSC polycrystal model done by C. Deo et al. (2008) for calculating yield strength of pre-irradiated SS T91 vs accumulated dose. In the latter work τ_o is determined by the interaction between radiation induced voids and dislocations. The density 'N' and diameter 'd' of vacancy clusters were assumed to increase with the square root of the dose, and τ_o was of the form:

$$\tau_o = \tau^{forest} + \tau^{irrad} \qquad \text{where} \qquad \tau^{irrad} = \alpha\mu b\sqrt{N.d} \qquad (2)$$

The latter is Orowan's expression for dilute precipitates. Using Eqs 1 and 2 in VPSC allows one to predict the yield stress of pre-irradiated SS T91 (see Figure 5a), and the stress-strain evolution during tensile tests done on pre-irradiated samples of SS 316L (see Figure 5b).

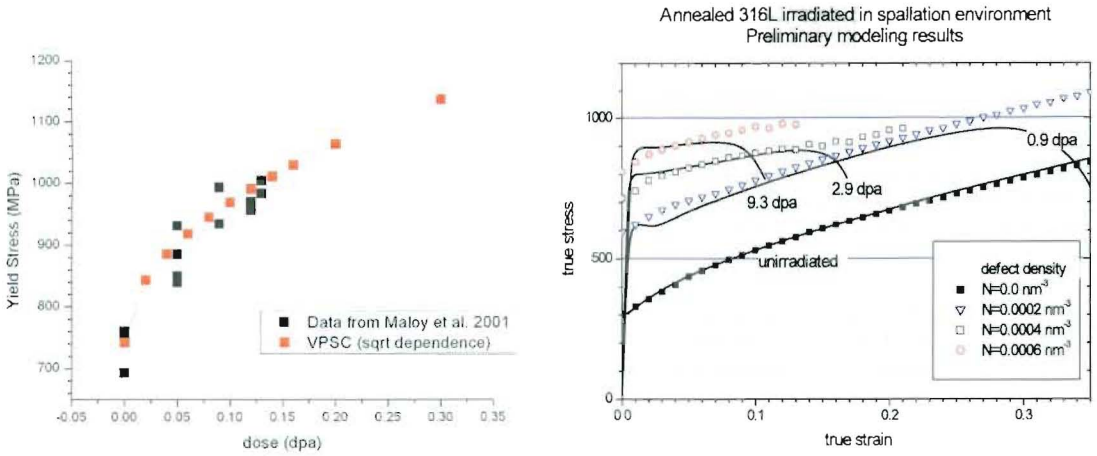

Figure 5: *(a) Variation of the yield stress with dose for the case of T91 steel subject to low radiation. The defect cluster size varies as the square root of the irradiation dose (dpa). Red squares are the VPSC calculations, black squares are the experimental data [Maloy et al. 2001]; (b) Tensile stress response of pre-irradiated SS 316L. Comparison of predictions (symbols) and experiments (lines). The density of defects was assumed to be approximately proportional to the square root of the dose.*

Future work should concentrate, at the lower scale, on developing models for the evolution of dislocation loops and voids with irradiation. At the crystal scale the parameters in the creep rate Eqs. 1 should be made explicit functions of the dislocation densities, and of the rate of vacancy and interstitial trapping by straight dislocations and loops. In order to obtain realistic climb rates. We foresee expressing Eq. 1 in the more explicit form

$$\dot{\varepsilon}_{ij} = \sum_k \rho_k \, b_i^{(k)} b_j^{(k)} \left(\Phi^{(int)} - \Phi^{(vac)}\right) \qquad (3)$$

Where ρ is dislocation density, 'b' is Burgers vector, and Φ is the flux of vacancy and interstitials. The evolution in density and size of interstitial loops as a function of dose and temperature should be provided by either the atomistic scale or the kinetic Monte Carlo scale.

In the case of applied strain, the models should predict the strengthening of the material due to the increase in density of forest dislocations, their mutual interactions, and the interaction of dislocations with voids, loops and interstitial clusters. All of the above should be combined to provide the evolution of the threshold stress τ_o with deformation.

Experiments needed:

* Characterize evolution of irradiation damage for different doses and temperatures, by doing TEM of archival and possibly also new in-reactor irradiated specimens. Ion beam irradiations followed by TEM. These experiments will validate the kMC component of the project by providing size and density of loops and voids to be compared with predictions.

4- Dislocation Dynamics (DD)

Dislocation dynamics is a simulation method in which dislocation networks are discretized into a series of line segments connected at nodes, and the nodes respond to elastic and chemical forces through user defined mobility laws [Suzuki, Arsenlis]. The simulations integrate the unit mechanism of dislocation mobility and interaction to deliver a continuum single crystal constitutive response and all of the details of the microstructural evolution that follow from the deformation history of the simulation. Typical outputs include the stress strain curve for a given strain rate and temperature, dislocation density as a function of dislocation character and deformation history, dislocation activity as a function of dislocation character and history, and other microstructural variables of interest in developing a continuum single crystal plasticity law. The inputs are dislocation mobilities, dislocation core energies, elastic constants, temperature and far field applied deformation history. Figure 5 shows an example of a stress-strain curve showing hardening of Mo single crystal [Arsenlis 2007],

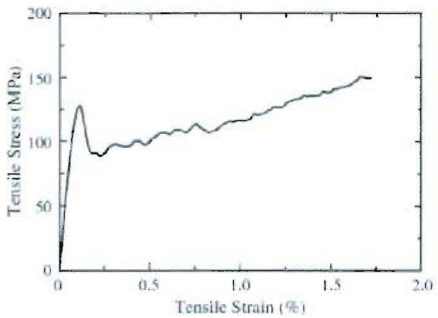

Figure 5: *A representative tensile stress–strain response for a simulation of a molybdenum single crystal with the loading direction along the [1 0 0] crystallographic axis [Arsenlis 2007].*

In simulating irradiation damage microstructures, irradiation induced prismatic loops are modeled as native objects within the code and simulations can be conducted to predict the effect of different loop densities, and loop sizes, and temperature on the strength of the microstructure. Other irradiation induced microstructures, such as voids and bubbles, can be introduced at various levels of discretization. The coarsest level is through the use of Eshelby inclusions to capture the far field interaction between these defects and dislocations, and through reaction product rules for short range interactions as they are simulated by molecular methods. In the irradiation strength simulations, the non-dislocation defect densities are treated as static objects that may be transformed by shearing. The output of theses simulations are the same outputs of the unirradiated simulations plus the relation between the evolving irradiation defect density distributions and the yield point and strain hardening response. Through these simulations, coarse grained constitutive relationships for use in continuum single crystal plasticity models are constructed for the stress/strain response and microstructural evolution including dislocation and irradiation defect populations. An example of such dislocation–defect (in this case, a stacking fault tetrahedron in an fcc lattice) interaction is shown in Fig. 6

The possibility also exists for combined simulations of dislocation dynamics and kinetic Monte Carlo methods for simulating the swelling, irradiation growth, and creep of materials. Rather than relying on a sink strength parameter for defect removal due to

dislocation interaction in a kMC calculation, explicit dislocation networks can be used to provide full discrete simulations of these inter-related phenomena. These types of simulations have yet to be performed in a large scale format, but a path forward exists to enable a more accurate simulation of the slow evolution of irradiation microstructure than has been possible in the past.

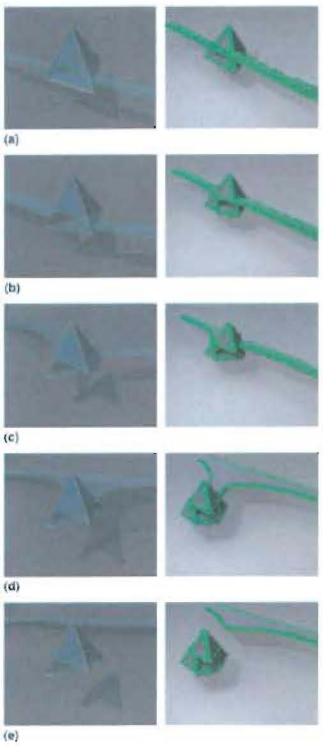

Figure 6: *Snapshots from dislocation dynamics (left) and molecular dynamics (right) simulations showing the interaction between a screw dislocation and a 4.6-nm stacking fault tetrahedron. The mechanisms, as obtained with MD and DD, are qualitatively identical. [Marian 2009]*

Current Status:

Dislocation dynamics codes are robust in being able to predict the strength of materials and can simulate the behavior of irradiated microstructures as long as they can be represented as native dislocation objects. The simulations are able to capture the behavior of a volume with 10μm sides with dedicated access to HPC resources. Depending on the mobility of dislocations, strain rates as low as 1 s-1 can be achieved, but obtaining lower strain rates remains a topic of ongoing research. Thus directly simulating the creep response of materials with dislocation dynamics has yet to be demonstrated and will involve significant algorithmic developments. Dislocation dynamics has been used by the ASC program to develop multiscale models of materials strength at high strain rates and has been shown to be predictive with the proper input parameters. Dislocation dynamics simulations typically employ isotropic elasticity in their simulations. Use of anistropic elasticity, while possible, increases the calculation cost by approximately one order of magnitude. The simulation of dislocations with non-native microstructural objects is also currently a field of research and development within the community.

Detailed treatments are also underway that simultaneously solve a continuum diffusion field for vacancy concentrations in a field of dislocations, and coupling with kinetic Monte Carlo simulation methods to simulate the explicit co-evolution of irradiation microstructures and the creep deformation under in service conditions. The simulation output would be the same as in the irradiated strength simulations in giving the macro stress strain response and it microstructural sources sufficient enough to generate coarse-grained continuum constitutive models.

Challenges and Experimental Needs:
Dislocation dynamics like many of the meso-scale (SXM, PXM) models discussed earlier are only as good as the physical elements they are built upon. Dislocation dynamics integrates the interaction of dislocations segments whose properties need to be specified by some other means. The most difficult properties to obtain are the mobility equations for a dislocation line segment as a function of dislocation character (edge, screw, mix), temperature, and stress. The dislocation character dependence makes this a difficult problem to fully address, and approximations are made so that only a select number of dislocation characters are sampled and interpolation functions are used to span the characters between the measured sets. Ideally, the single dislocation defect mobility would be provided by experimental means; however, this measurement has never been directly reported. Single dislocation mobility may also come from molecular dynamics/statics simulations, but potentials are needed that take into account alloying of multiple elements for the mobilities to be predicted accurately. Molecular potentials for alloys are only available for a select group of alloys today.
The other information needed by the dislocation dynamics simulations are the initial conditions of the irradiated microstructure and the interaction of dislocations with non-dislocation irradiation damage. Dislocation dynamics needs interactions relationships for both the far field elastic interactions and the near field interactions that occur at contact. Again, these interaction rules must either come from experiments or from molecular dynamics simulations.

5- kinetic Monte-Carlo (kMC)

Kinetic Monte Carlo simulations are used to further integrate in time the irradiation damage evolution from the single cascade simulation conducted in molecular dynamics to the coarsening of defects into clusters, prismatic loops, voids, and bubbles. In order to reach the long times and high effective dpa levels, advanced kinetic Monte Carlo techniques are needed to accelerate the time step of integration. Common kMC methods begin to stagnate as coarsening occurs and the distance between defects increases. New methods such as First Passage diffusion Monte Carlo [Oppelstrup 2006 and 2009] and parallel kinetic Monte Carlo are developed and used to simulate the irradiation damage evolution to tens and hundreds effective displacements per atom. The output of these models is the defect evolution and swelling rates as a function of dose rate, dose, and temperature. The defect evolution is quantified in the number density of defects of a particular size and shape. Figure 7 shows results for swelling of Fe at two damage rates, for total doses not accessible with standard k-MC algorithms (Donev 2010).

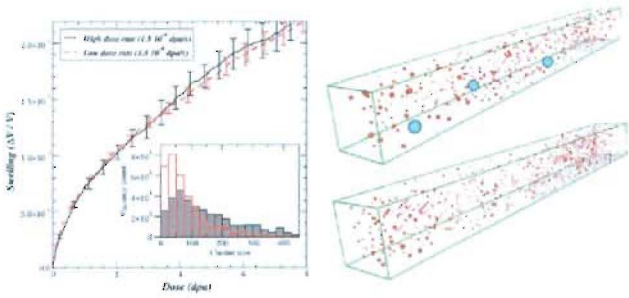

Figure 7: *a-Fe film irradiated by electrons to the total dose of 10 dpa: (Top right) At the high dose rate of 1.5 10^{-4} dpa/s and T = 262 _C and (Bottom right) At the low dose rate of 1.5 10^{-8} dpa/s and T = 130 _C. (Left) Volume fraction of vacancies (swelling) as a function of damage dose. The black solid curve is the swelling kinetics under the high dose rate/high temperature and the dashed red curve is the same kinetics under the low dose rate/low temperature irradiation conditions. Inset shows a histogram of the distribution of vacancy cluster sizes at a dose of 5 dpa for both the high dose rate/high temperature irradiation (shaded gray bars) and for the low dose rate/low temperature irradiation (red bars). (Donev 2010).*

The parallel k-MC algorithm of Martinez et al [2008] provides access to supercomputer power for an algorithm that has so far been serial. Figure 8 shows the evolution of the annihilation kinetics of a two-species diffusion-reaction model over 8 orders of magnitude in time.

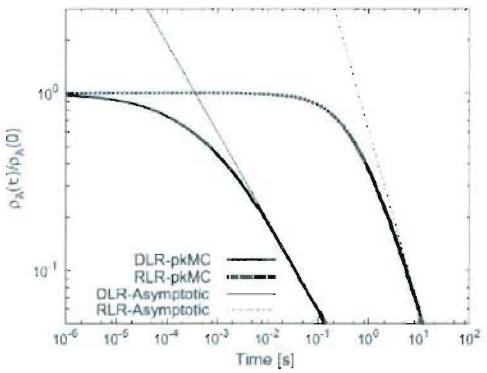

Figure 8: *Two-species annihilation kinetics (only the A-type normalized density is shown) for the reaction (l >> dc) and diffusion (l<dc) limited regimes as obtained with Martinez's parallel kMC algorithm. The expected asymptotic decay law in each case is also shown for reference.*

Challenges and Experimental Needs:

These achievements represent a major step forward in increasing the power of k-MC simulations. However, to model irradiation creep along with swelling at this length scale for more realistic materials, namely concentrated alloys with nanostructures, the kMC simulations must be extended to include dislocation microstructures (through a DD coupling) and their associated stress fields along with the far field stress drivers for creep, dislocation climb motion, and the effects of alloying elements on diffusion / trapping of

mobile defects. Additionally the k-MC algorithm has to be sensitive to the thermodynamics of the system to incorporate the evolution of the phases, namely segregation, precipitation, patterning, etc. In this way, all sink and source terms would eventually be modeled explicitly and not assigned. This is a major challenge for k-MC algorithms and represents a significant fraction of the modeling effort. When achieved, such a modeling capability will be able to provide the time dependent laws that govern the evolution of the microstructure under irradiation, where the kinetic and thermodynamic data for coarser models could be taken from.
The information required as input to these simulation must either come from experiment or lower length scale simulations; however, at this length scale the experimental measurement of model parameters appears improbable. The likely scenario is that irradiation experiments will be used to validate the model and that lower length scale simulations will be used to populate the model parameters. The model parameters needed are the energies of the irradiation defects as a function size of the defect and its composition (e.g. He/vacancy ratio), the energy barriers for defect migration as a function of the same parameters, and the initial defect structures created by the collision cascades during neutron irradiation.

6- Molecular Statics and Dynamics (MS/MD) using classical potentials
This simulation tool is used to investigate unit mechanisms of defect formation and motion associated with irradiation creep, strength, swelling and fracture. The tool simulates materials at the atomic level where the degrees of freedom are atomic positions and velocities. Simulations of the initial recoil cascade are conducted to extract statistically relevant information on the point spatial distribution of defect pairs that are generated and remain after several nanoseconds of the cascade event. Simulations also provide information about the following point defect properties: point defect formation energies, point defect migration energies and diffusion constants as a function of stress, point defect formation volumes, the energy of point defect clusters as a function of their size, and the shape and strain fields of point defect clusters (loops, voids, etc.) as a function of their size.
Simulations at this length scale are also used to understand the unit mechanism of dislocation plasticity in both glide and climb. Atomistic simulation are conducted to calculate the following transferable quantities: Elastic constants, isolated dislocation core energies as a function of Burgers vector and line direction, and isolated dislocation glide mobility as a function of stress, and dislocation Burgers vector and line direction. The climb mobilities of dislocations are a consequence of the absorption and emission of point defects from dislocation lines. The absorption kinetics is dictated by the mobility of point defects as a function of stress mentioned earlier. The emission kinetics of point defects requires the calculation of the energy barrier of point defect emission from a dislocation core as a function of stress.
The final series of simulations that need to be conducted at the molecular level are simulations that detail the reaction products of dislocation irradiation defect intersection as a function of irradiation damage defect type and dislocation geometry. These simulations describe the interactions in terms of how the irradiation defect microstrcuture changes by dislocation crossing, and it details the strengthening and strain

hardening/softening implications of irradiation damage defects on dislocation plasticity. The simulations are of small scale and consider the unit process of a single dislocation intersecting a single irradiation defect.
The information from the MS/MD simulations is used to inform the kMC and DD simulation tools about the initial conditions of irradiation damage defect production, the atomic energies of defects, the mobility of defects, the long range interaction of defects through stress fields, and the short range interaction of defects through their reaction products resulting from interactions.

Challenges:
This methodology however has it s limits, determined by the accuracy of the classical potentials used to define the interatomic interactions. Usually, potentials are fitted to an ensemble of data including both *ab initio* and experimental information. While a large body of research for many pure elements exists in the literature, the situation for many other elements (such as Fe, actinides) as well as for multicomponent alloys, oxides, and ceramics is less satisfactory. There is no recipe to link the results of the electronic structure to classical Hamiltonians. A significant effort in the direction of improving the accuracy of classical potentials, including better treatments for covalency, charge transfer, and transferability, is necessary in order to be able to model at the classical level the materials and processes of interest for nuclear applications. This objective is central to most computational materials science programs. Figure 9 sketches the modeler's playground where these challenges are represented by the three most important types of bonding (Phillpot 2009).

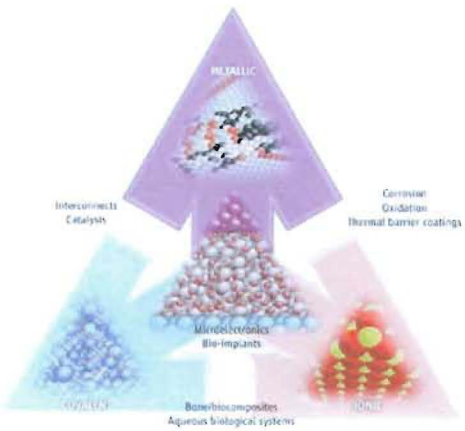

Figure 9: *The modelers' playground. Many of the most challenging and important applications of materials involve interfaces between disparate bonding environments. The ability to simulate such interfaces promises new capabilities in computationally prototyping nanostructures or devices at experimental length scales. (Phillpot 2009).*

Current status:
The state of the art for ferritic steels can be characterized by an abundant body of research at the *ab initio* level describing properties of Fe and FeCr alloys, together with

some of the most important solute elements, such as C, He, Cu. Work is needed on the study of interfaces between the nanoscale oxide precipitates used to improve the creep and radiation resistance of steels, and the matrix. At the atomic scale, potentials for Fe and its alloys have encounter the difficulty that finite T properties of Fe cannot be properly captured due to the magnetic character of these alloys. This limitation is an obstacle for further progress on atomic scale modeling of high temperature properties, and requires a significant effort to be resolved. For pure Fe and FeCr alloys at low T, the tools exist to predict many of the properties that coarser scales need. As an example, we mention that MS was used to calculate configurations of dislocation cores and of vacancies and interstitials both, in the equilibrium and migration points. From this information diffusivities can be obtained, that will feed into the kMC scale described above. The information required is reliable potentials for Fe and for Fe-Cr. Specifically, calculations of vacancy and interstitial dumbbell configurations in Fe have been performed, and their interaction energies with edge and screw dislocations have been characterized [Hayward et al, 2009] (see Fig 10). The next step is to obtain diffusivities of vacancies and interstitials, and sink strength of dislocations for trapping vacancies and interstitials. Sink strengths will be used in SX models in combination with dislocation and loop densities to predict irradiation creep.

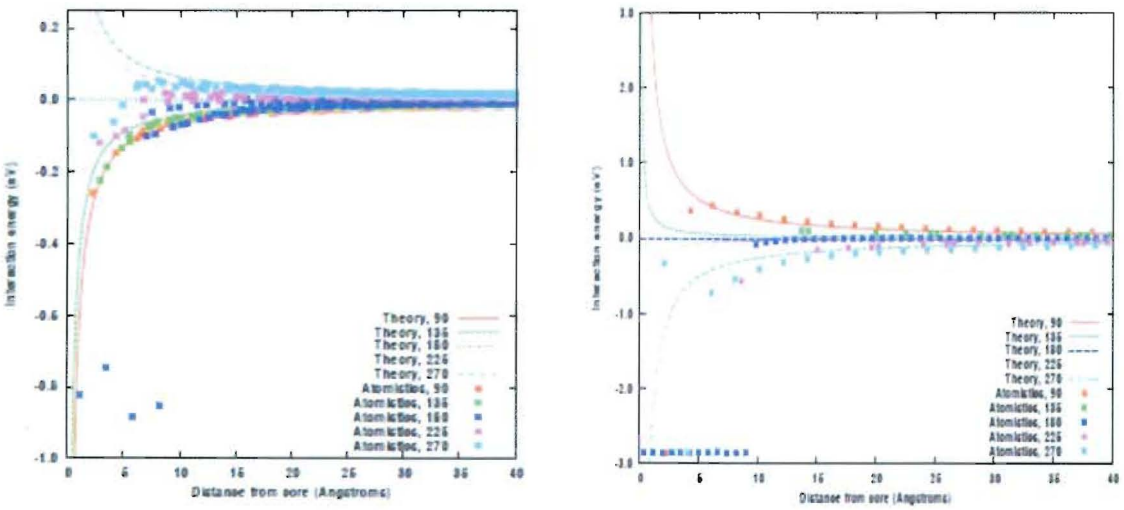

Figure 10: *Interaction energy of vacancies (left) and interstitial dumbbells (right) with edge dislocation in Fe, as a function of the separation from the dislocation core. Observe the low energy when very close (10A) to the core [Hayward et al. 2009]*

7- Electronic structure (ES) scale

The electronic structure scale, the shortest in the Multiscale approach, is the only that is parameter free and can be considered as the fundamental stone for predictive computational materials science. For the materials of interest for cladding applications, (transition metal alloys and their oxides) the *ab initio* methods based on approximations to Density Functional Theory are accurate enough to provide reliable data. The limitation

resides in the number of atoms that can be resolved, with the current limit at few hundred's, and, as mentioned above, the fact that finite T properties of magnetic structures are not well captured by DFT. The Multi-Scale-Multi-Physics approach for cladding starts at this scale, as basic information about properties of oxides, of fission product attack, of corrosion with the coolant, etc, require the precise knowledge of the chemistry/bonding between these disparate substances.

Challenges:

The most difficult barrier in the multiscale approach is the translation of these data into coarser scales, as sketched in Figure 11.

Significant progress is being done in this area, as for example the chain of models that link the *ab initio* electronic structure to the tight binding one electron picture, to the analytic bond order potentials, developed by D. Pettifor and collaborators for transition metal alloys (Drautz 2006), the reactive potentials of Goddard and collaborators (van Duin 2008). The other barrier depicted in Fig 10 is the one linking a prescribed Hamiltonian to desired thermodynamic (finite T) properties. This subject is receiving increased attention, as the goal of nuclear materials modeling is the prediction of the behavior at finite temperatures. In this respect, methodologies for targeting free energy properties are now appearing and will be an important part of the approach proposed here (Caro 2007). These methodologies will provide the input necessary for coarser scale simulations of microstructure evolution.

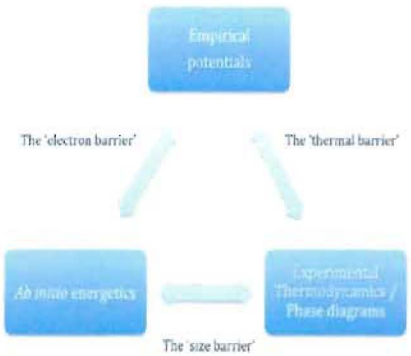

Figure 11: *schematic representation of the link between the electronic structure, the classical potentials picture and the finite temperature properties. The 'barriers' represent the challenges for modeling*

8- Summary

We reviewed the basis of our Multi Scale Multi Physics approach to build a chain of models/simulation tools spanning from the electronic/atomic scale to the actual reactor component scale. The strategy followed was to identify the physically relevant parameters required by the upper scale models and extract those from the lower scale models.

Targeting predictive capabilities for **creep, strength, swelling and fracture** of cladding requires work on all scales. Some scales have reached a degree of maturity that only

require predictive input to produce predictive output; others require moderated or significant modeling efforts to reach predictive power.

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