Perspectives 8

THE ROLE OF THEORY AND MODELING IN THE DEVELOPMENT OF MATERIALS FOR FUSION ENERGY

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The environmental and operational conditions of First Wall/ Blanket (FW/B) structural materials in fusion energy systems are undoubtedly amongst the harshest in any technological application. These materials must operate reliably for extended periods of times without maintenance or repair. They must withstand the assaults of high particle and heat fluxes, as well as significant thermal and mechanical forces. Rival conditions have not been experienced in other technologies, with possible exceptions in aerospace and defense applications. Moreover, the most significant dilemma here is that the actual operational environment cannot be experimentally established today, with all of the synergistic considerations of neutron spectrum, radiation dose, heat and particle flux, and gigantic FW/B module sizes. Because of these considerations, we may rely on a purely empirical and incremental boot-strapping approach (as in most human developments so far), or an approach based on data generation from non prototypical setups (e.g., small samples, fission spectra, ion irradiation, etc.), or a theoretical/computational methodology. The first approach would have been the most direct had it not been for the unacceptable risks in the construction of successively larger and more powerful fusion machines, learning from one how to do it better for the next. The last approach (theory and modeling alone) is not a very viable option, because we are not now in a position to predict materials behavior in all its aspects from purely theoretical grounds. The empirical, extrapolative approach has also proved itself to be very costly, because we cannot practically cover all types of material compositions, sizes, neutron spectra, temperatures, irradiation times, fluxes, etc. Major efforts had to be scrapped because of our inability to encompass all of these variations simultaneously. While all three approaches must be considered for the

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development of fusion materials, the multi-scale materials modeling (MMM) framework that we propose here can provide tremendous advantages if coupled with experimental verification at every relevant length scale.

A wide range of structural materials has been considered over the past 25–30 years for fusion energy applications [1]. This list includes conventional materials (e.g., austenitic stainless steel), low-activation structural materials (ferritic/martensitic steels, V-4Cr-4Ti, and SiC/SiC composites), oxide dispersion strengthened (ODS) ferritic steels, conventional high temperature refractory alloys (Nb, Ta, Cr, Mo, W alloys), titanium alloys, Ni-based super alloys, ordered intermetallics (TiAl, Fe_3Al, etc.), high-strength, high-conductivity copper alloys, and various composite materials (C/C, metal-matrix composites, etc.). Numerous factors must be considered in the selection of structural materials, including material availability, cost, fabricability, joining technology, unirradiated mechanical and thermophysical properties, radiation effects (degradation of properties), chemical compatibility and corrosion issues, safety and waste disposal aspects (decay heat, etc.), nuclear properties (impact on tritium breeding ratio, solute burnup, etc.).

Strong emphasis has been placed within the past 10–15 years on the development of three reduced-activation structural materials: ferritic/martensitic steels containing 8–12%Cr, vanadium base alloys (e.g., V-4Cr-4Ti), and SiC/SiC composites. Recently there also has been increasing interest in reduced-activation ODS ferritic steels. Additional alloys of interest for fusion applications include copper alloys (CuCrZr, Cu–NiBe, dispersion-strengthened copper), tantalum-base alloys (e.g., Ta-8W–2Hf), niobium alloys (Nb–1Zr), molybdenum, and tungsten alloys. In the following, we give a brief analysis of the most limiting mechanical properties based on our earlier work [1].

1. Lower Operating Temperature Limits

The lower temperature limits for FW/B structural materials (i.e., excluding copper alloys) are strongly influenced by radiation effects. For body-centered cubic (BCC) materials such as ferritic-martensitic steels and the refractory alloys, radiation hardening at low temperatures can lead to a large increase in the Ductile-To-Brittle-Transition-Temperature (DBTT)[2, 3]. For SiC/SiC composites, the main concerns at low temperatures are radiation-induced amorphization (with an accompanying volumetric swelling of $\sim$11%) [4] and radiation-induced degradation of thermal conductivity. The radiation hardening in BCC alloys at low temperatures ($0.3T_M$) is generally pronounced, even for doses as low as $\sim$1 dpa [3]. The amount of radiation hardening typically decreases rapidly with irradiation temperature above 0.3 T_M, and radiation-induced increase in the DBTT may be anticipated to be acceptable at temperatures above $\sim 0.3T_M$. A Ludwig–Davidenkov relationship between hardening

and embrittlement was used to estimate the DBTT shift with increased irradiation dose. In this model, brittle behavior occurs when the temperature dependent yield strength exceeds the cleavage stress. It is worth noting that operation at lower temperatures (i.e., within the embrittlement temperature regime) may be allowed for some low-stress fusion structural applications (depending on the value of the operational stress intensity factor relative to the fracture toughness).

Numerous studies have been performed to determine the radiation hardening and embrittlement behavior of ferritic-martensitic steels. The hardening and DBTT shift are dependent on the detailed composition of the alloy. For example, the radiation resistance of Fe-9Cr-2WVTa alloys appears to be superior (less radiation hardening) to that of Fe-9Cr-1MoVNb. The radiation hardening and DBTT shift appear to approach saturation values following low temperature irradiation to doses above 1–5 dpa, although additional high-dose studies are needed to confirm this apparent saturation behavior. At higher doses under fusion conditions, the effects of He bubble accumulation on radiation hardening and DBTT need to be addressed. Experimental observations revealed brittle behavior (K_{IC} ~30 MPa-$\sqrt{m}$) in V-(4–5)%Cr-(4–5)%Ti specimens irradiated and tested at temperatures below 400 °C. From a comparison of the yield strength and Charpy impact data of unirradiated and irradiated V-(4–5)%Cr-(4–5)%Ti alloys, brittle fracture occurs when the tensile strength is higher than 700 MPa. Therefore, 400 °C may be adopted as the minimum operating temperature for V-(4–5)%Cr-(4–5)%Ti alloys in fusion reactor structural applications[5]. Further work is needed to assess the impact (if any) of fusion-relevant He generation rates on the radiation hardening and embrittlement behavior of vanadium alloys.

Very little information is available on the mechanical properties of irradiated W alloys. Tensile elongation of ~ 0 have been obtained for W irradiated at relatively low temperatures of 400 and 500 °C (0.18–0.21 T_M) and fluences of $0.5-1.5\times10^{26}$ n/m^2 ($\prec$2 dpa in tungsten) [6]. Severe embrittlement (DBTT $\geq$ 900 °C) was observed in un-notched bend bars of W and W-10%Re irradiated at 300 °C to a fluence of 0.5×10^{26} n/m^2 ($\prec$ 1 dpa). Since mechanical properties data are not available for pure tungsten or its alloys irradiated at high temperatures, an accurate estimate of the DBTT versus irradiation temperature cannot be made. The minimum operating temperature which avoids severe radiation hardening embrittlement is expected to be 900 ± 100 °C.

2. Upper Operating Temperature Limits

The upper temperature limit for structural materials in fusion reactors may be controlled by four different mechanisms (in addition to safety considerations): Thermal creep, high temperature helium embrittlement, void swelling, and

compatibility: corrosion issues. Void swelling is not anticipated to be significant in ferritic-martensitic steel [7] or V–Cr–Ti alloys [8] up to damage levels in excess of 100 dpa, although swelling data with fusion-relevant He:dpa generation rates are needed to confirm this expectation and to determine the lifetime dose associated with void swelling. The existing fission reactor database on high temperature (Mo, W, Ta) refractory alloys (e.g., [6]) indicates low swelling ($\prec 2\%$) for doses up to 10 dpa or higher. Radiation-enhanced recrystallization (potentially important for stress-relieved Mo and W alloys) and radiation creep effects (due to a lack of data for the refractory alloys and SiC) need to be investigated. Void swelling is considered to be of particular importance for SiC (and also Cu alloys, which were shown to be unattractive fusion structural materials [1].

An adequate experimental database exists for thermal creep of ferritic-martensitic steels [7] and the high temperature (Mo, W, Nb, Ta) refractory alloys [10]. Oxide-dispersion-strengthened ferritic steels offer significantly higher thermal creep resistance compared to ferritic-martensitic steels [11], with a steady-state creep rate at 800 °C as low as 3×10^{-10} s^{-1} for an applied stress of 140 MPa. The V-4Cr-4Ti creep data suggest that the upper temperature limit lies between 700 and 750 °C, although strengthening effects associated with the pickup of 200–500 ppm oxygen during testing still need to be examined. The predicted thermal creep temperature limit for advanced crystalline SiC-based fibers is above 1000 °C [12].

One convenient method to determine the dominant creep process for a given stress and temperature is to construct an Ashby deformation map. Using the established constitutive equations for grain boundary sliding (Coble creep), dislocation creep (power law creep) and self-diffusion (Nabarro–Herring) creep, the dominant deformation-mode regimes can be established [1].

3. Operating Temperature Windows

Figure 1 summarizes the operating temperature windows (based on thermal creep and radiation damage considerations) for nine structural materials considered by Zinkle and Ghoniem [1]. The temperature limits for Type 316 austenitic stainless steel are also included for sake of comparison. In this figure, the light shaded regions on either side of the dark horizontal bands are an indication of the uncertainties in the temperature limits. Helium embrittlement may cause a reduction in the upper temperature limit, but sufficient data under fusion-relevant conditions are not available for any of the candidate materials. Due to a high density of matrix sinks, ferritic/martensitic steel appears to be very resistant to helium embrittlement [13]. An analysis of He diffusion kinetics in vanadium alloys predicted that helium embrittlement would

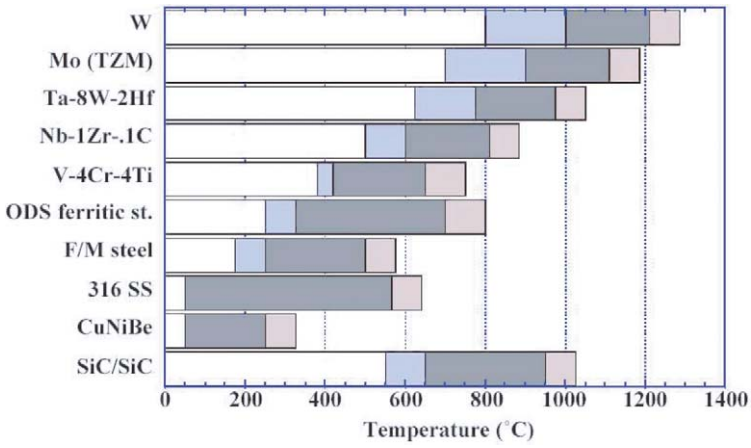

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Figure 1. Operating temperature windows (based on radiation damage and thermal creep considerations) for refractory alloys, Fe-(8-9%)Cr ferritic-martensitic steel, Fe-13%Cr oxide dispersion strengthened ferritic steel, Type 316 austenitic stainless steel, solutionized and aged Cu-2%Ni-0.3%Be, and SiC/SiC composites. The light shaded bands on either side of the dark bands represent the uncertainties in the minimum and maximum temperature limits.

be significant at temperatures above 700 °C [14]. The lower temperature limits in Fig. (1) for the refractory alloys and ferritic:martensitic steel are based on fracture toughness embrittlement associated with low temperature neutron irradiation. An arbitrary fracture toughness limit of 30 MPa-$\sqrt{m}$ was used as the criterion for radiation embrittlement. Further work is needed to determine the minimum operating temperature limit for oxide dispersion strengthened (ODS) ferritic steel. The value of 290 ± 40 °C used in Fig. (1) was based on results for HT-9 (Fe-12Cr ferritic steel). The minimum operating temperature for SiC/SiC was based on radiation-induced thermal conductivity degradation, whereas the minimum temperature limit for CuNiBe was simply chosen to be near room temperature. The low temperature fracture toughness radiation embrittlement is not sufficiently severe to preclude using copper alloys near room temperature [15], although there will be a significant reduction in strain hardening capacity as measured by the uniform elongation in a tensile test. The high temperature limit was based on thermal creep for all of the materials except SiC and CuNiBe. Due to a lack of long-term (10 000 h), low-stress creep data for several of the alloy systems, a Stage II creep deformation limit of 1% in 1000 h for an applied stress of 150 MPa was used as an arbitrary criterion for determining the upper temperature limit associated with thermal creep. Further creep data are needed to establish the temperature limits for longer times and lower stresses in several of the candidate materials.

4. Main Challenges

4.1. Deformation and Fracture

Problems of deformation and fracture stem from several phenomena that can render structural material brittle either at low or at high temperature. The interplay between these phenomena is complex resulting in deformation and fracture properties to depend on many intrinsic and extrinsic variables. We must therefore take into account environmental variables, when we develop a database for materials properties, which can be functions of (T, dpa, dpa rate, He, H, stress, etc.). Since properties involve many mechanisms at multiscale levels, and sometimes differences between large rates result in macroscopic changes (e.g., void swelling), we need to develop physically based properties models. We also need a modeling-experiment integration strategy for validation of models at different scales. Models that are to be developed can be hierarchical, starting from the atomic information all the way to the prediction of constitutive relationships and macroscopic fracture.

The main crosscutting issues in deformation and fracture are:

1. Irradiation effects on stress-strain, constitutive laws, and consequences of flow localization;
2. Validity & physical basis of the Master–Curve (MC) for predicting the ductile-to-brittle-transition;
3. Embrittlement – MC shifts due to hardening & He effects
4. Model-based designs for high performance alloys;
5. Irradiation effects on constitutive properties: J2 laws linked to microstructure evolution;
6. Development of plasticity models for constitutive properties, for example bridging between Dislocation Dynamics, crystal plasticity, and polycrystalline plasticity;
7. Understanding flow localization and ductility loss of irradiated materials;
8. The apparent universality of the MC shape, and the physical basis for this universality.
9. Effects of helium on GB, and how that influences shifts in DBTT;
10. Model-based design of alloys; for example including a high density of nano-clusters to trap helium in high-pressure bubbles and thus preventing them from going to grain boundaries.

4.2. Helium Effects

Several methods of modeling helium effects on irradiated materials have been developed over the past two decades. Atomistic MD simulations are

now being used more extensively to determine the energetics of binding and migration of various helium-vacancy complexes. Information on the fraction of residual defects has also been obtained from cascade simulations. Such atomic level information is passed on to mesoscale simulations of microstructure evolution based on reaction rate theory. Most of these simulations have assumed that the microstructure is spatially homogeneous in space and time. However, some of these assumptions have been relaxed, such as the effects of cascades on point defect diffusion, formation of microstructure patterns, etc.

One of the key advantages of rate theory is that the results of simulations can be directly compared to experiments, while the key parameters are obtained from either experiments or atomistic simulations. For example, KMC simulations can now be used to solve complex point defect diffusion problems in the stress field of dislocations, and thus derive more realistic values for the dislocation bias factors. At the same time, large systems of equations describing the nucleation and growth of void and bubble populations can be solved with current day large-scale computers, thus providing more accurate descriptions of nucleation and growth. This level of detailed rate theory modeling is essential, because experiments show that several phenomena are influenced by helium in a complex fashion, for example, the swelling rate is not a monotone function of the helium-to-dpa ratio. Likewise, the effects of small helium concentrations on grain boundary fracture depend on many details of the microstructure, while the effects of helium bubbles on hardening or embrittlement at low temperature is not yet clear.

4.3. Radiation Stability of Alloys

Real alloys are made of major and minor components. While the fate of minor elements under irradiation can be handled, in principle at least, with the same tools as point defects (i.e., book keeping of the mean or local concentration as a function of time), such is not the case for major alloy components. In particular, the cluster dynamics technique (rate theory) fails, because of percolation problems. A small community works at developing a theoretical framework to assess the stability of stationary phases under irradiation. At the present time, it is acknowledged that the latter stability depends altogether on the temperature, composition, irradiation flux and "cascade size". This implies that both the spatial extension of the cascade and the number of replacements per cascade are important factors. For the overall approach to be justified, the evolution of the precipitate population under irradiation must be fast compared to that of the defect sink structure (dislocation network, defect aggregates of various forms): such is indeed the case since the latter evolves at a rate proportional to the small difference between the vacancy and the interstitial fluxes

at sinks, two large quantities; on the contrary, the precipitates grow or shrink because of the coupling of the solute flux with the two above fluxes, an additive process.

5. Modeling Research Needs

5.1. Interatomic Potentials for Radiation Damage

The crucial properties for radiation damage simulations are:

(1) Point defect formation, migration and interaction energies;
(2) Elastic constant anisotropy;
(3) Grain boundary energetics;
(4) Dislocation structure and response to stress;
(5) Alloy phase stability.

None of these properties are automatically correct as a result of the physical basis of potentials. With pair-wise interactions, some are necessarily wrong. With many-body potentials (used here as a generic term covering glue, Finnis–Sinclair, embedded atom, modified embedded atom and effective medium theory potentials) many can be fitted provided "correct" values are available. These types of potentials have been the "state of the art" for twenty years.

Historically, there has been an insufficient database for robust potential fitting. Not all this data is available experimentally for parameterization and verification of potentials. Recent renewed interest in interatomic potentials is based on the ability of ab initio calculations to provide this missing data – with teraflop machines verification of predictions is finally possible. Where tested against new data, existing potentials have generally proved disappointing. Some common problems include poor interstitial formation energies, the energy difference between configurations too small and no satisfactory description of the austenitic-ferritic transition. Some of these problems can be traced to problems in parameterization of the potentials and have been addressed in recent work by simple reparameterization. Others, such as the absence of a physically sensible treatment of magnetization, point to more fundamental problems in the many-body potential concept.

The majority of the effort in potentials for metallic phases has focused on elemental materials. Potentials for multi-component systems have been developed in isolated cases, but the predictive capability of these potentials is typically disappointing. Reasonable models for the mission-critical helium impurities exist, the inertness of helium making its behavior in MD somewhat insensitive to parameterization. There are two challenges in the development of potentials in alloy systems. First, there is generally much less data

available though this can be rectified through the use of *ab initio* methods. Second, the appropriate functional forms are not as well developed. Most potentials are based on simple pictures of bonding. In alloy systems, the nature of the bonding is inherently more complex suggesting that more sophisticated potentials are needed to describe the energetics reliably. Non-metallic impurities (carbon, phosphorus) are more problematic.

5.2. Dislocation Interactions & Dynamics

One of the critical problems for the development of radiation-resistant structural materials is the embrittlement, loss of ductility and plastic flow localization. Modeling the interaction between dislocations and radiation-induced obstacles is providing great insights into the physics of this problem, and will eventually lead to the design of radiation-resistant structural alloys.

Models of dislocation-defect interactions are pursues at two levels: (1) the atomistic level, where MD simulations are playing significant role; and (2) the mesoscopic level, where DD simulations are providing insights into larger-scale behavior. Both types of models are complementary, and provide direct information for experimental validation on the effects of irradiation on hardening, yield drop, and plastic flow localization, etc. Atomic scale models are used to "inform" DD models on the details of dislocation-defect interactions. Presently, MD models can simulate 1–10 million atoms on a routine basis. Both static and dynamic simulations are used. For static simulations, fixed displacement boundary conditions are applied, and conjugate gradient minimization is used. On the other hand, Newtonian equations of motion are used for dynamic simulations, and either force or velocity conditions are applied on boundary atoms. Atomistic simulations have shown the range where elasticity estimates are valid for dislocation-defect interactions, and where they break down due to new mechanisms. For example, the interaction of dislocations with small precipitates can result in local phase transitions and an associated energy cost that cannot be predicted from DD models. Also, it has been shown that dislocation-void interaction leads to dislocation climb, and the formation of a dislocation dipole before the dislocation completes cutting through the void completely. These effects are all of an atomic nature, and the information should be passed on to DD simulations.

A number of challenges remain in the area of dislocation-defect interactions, as described below:

(1) The strain rates in MD simulations are far in excess of experimentally achievable rates, and methods to incorporate slow rate events due to temperature or force field fluctuations have not yet been developed.

(2) The information passing between MD and DD is not systematic yet. For example, the "angle" between dislocation arms before it leaves the obstacle is often used in DD simulations as a measure of obstacle strength. However, the definition of this angle in both MD and experiments is problematic for a variety of reasons. Force-displacemt information will be necessary.
(3) Methods for incorporating lower length scale microstructure effects into DD simulations are not well developed. For example, we do not have information on obstacle dynamics, solute effects, dislocations near cracks, dislocation nucleation, etc.
(4) The size of atomistic simulations is very small, and cannot deal with complex dislocation structures. Methods for reducing the degrees of freedom are needed.
(5) The boundary conditions used in MD simulations are either periodic, fixed, or represented by elastic Green's functions. General methods for embedding MD simulations into the continuum are in an early stage of development.
(6) DD codes are limited to small size crystals. To improve their speed and range of applicability, new methods of designing these codes on massively parallel computers are needed.
(7) The connection between DD and macroscopic plasticity has not yet been made through "coarse graining" and a systematic reduction of the degrees of freedom. Development of this area is essential to the prediction of constitutive relations and macroscopic plastic deformation.

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1. Figure in color.