A WHITEPAPER PROPOSING AN INTEGRATED PROGRAM OF THEORETICAL, EXPERIMENTAL, AND DATABASE RESEARCH FOR THE DEVELOPMENT OF ADVANCED FUSION MATERIALS

R. E. Stoller
Oak Ridge National Laboratory

G. R. Odette
University of California, Santa Barbara

H. L. Heinisch
Pacific Northwest National Laboratory

Prepared for the Office of Fusion Energy Sciences, U. S. Department of Energy

AN INTEGRATED THEORY, MODELING, EXPERIMENTAL, AND DATABASE PROGRAM FOR THE DEVELOPMENT OF ADVANCED FUSION MATERIALS

R. E. Stoller (ORNL), G. R. Odette (UCSB), and H. L. Heinisch (PNNL),

1 INTRODUCTION AND OBJECTIVES OF PLANNING ACTIVITY

This white paper attempts to describe an improved framework for establishing an integrated theory, modeling, experimental, and data base program for developing advanced fusion materials. This document is not intended to be a complete review of the current program with examples, data, models and references. Rather, the intent is to provide guidance on fundamental issues related to fusion materials development and to describe the concept of integrated modeling. The overall objective is to infuse the program with new ideas and people, to leverage broad advances in science and engineering and, most importantly, to accelerate the development of materials systems that will help make the promise of fusion energy into a 21st century reality.

The Advanced Materials Program (AMP) is funded through the U.S. DOE Office of Fusion Energy Sciences. This program is currently being restructured to reflect an increased focus on the underlying science required to develop materials for use in a fusion power system. As part of this process, the AMP was recently reviewed by an expert panel appointed by the Fusion Energy Sciences Advisory Committee (FESAC). In its report, the FESAC Materials Review Panel recommended that the AMP should adopt the following as its primary goal: “To provide the materials science knowledge base that will enable the utilization of fusion energy. The near-term emphasis will be on feasibility issues for in-vessel components for deuterium-tritium systems.”

The panel recognized that combined advances in materials science understanding and modern computing capabilities can be used to extend the AMP’s ability to develop meaningful physically-based, semi-empirical models that can guide and help interpret ‘costly and difficult to obtain’ experimental information. The panel recommended that these models should be developed and applied to the key feasibility issues associated with each class of material, and that crosscutting phenomena common to multiple materials are particularly good candidates for their application.

The greater emphasis on modeling should primarily be seen in the context of increased integration of theory, experiment, and database development as part of the long-term effort to build a knowledge base. It is essential that an expanded computational materials effort should:

a) focus on the key issues in fusion materials,
b) be closely integrated with the experimental program, which should include experiments designed for model validation and obtaining parameters requited by the models,
c) be as collaborative as possible and be leveraged by other activities,
d) produce tangible useful results (e.g. more reliable predictions of properties), and finally
e) provide the scientific basis for improved design and methods of structural integrity assessment.

In a related recommendation, the panel encouraged the AMP to utilize a system of roadmaps as a means of defining feasibility issues, and of identifying the skills and resources required to address them. In response to this recommendation, the AMP has recently developed a draft White Paper and a roadmap that discusses its current strategic pathway to fusion materials development. The AMP White Paper identifies the long-term goal of the AMP as the development of structural materials that will permit fusion to be developed as a safe, environmentally acceptable, and economically competitive energy source. This goal is to be accomplished through a science based program of theory, experiment, and modeling that:

a) provides an understanding of the behavior of candidate material systems in the fusion environment and identifies limiting properties and approaches to improve performance,
b) undertakes the development of alloys and ceramics with superior properties for service in the fusion environment through the control of composition and microstructure, and
c) provides the materials technology required for production, fabrication, and power system design.

This *Whitepaper on an Integrated Theory, Modeling, Experimental, and Database Program* (hereafter referred to as the *integrated modeling program*) has been prepared as part of the overall AMP planning process. In particular, it is the program's response to the FESAC panel recommendation for increased attention to modeling and the relationship between the modeling and experimental components of the program. The overall AMP White Paper identifies the technological challenges to establishing the feasibility of utilizing the four groups of structural materials currently under consideration for applications in fusion environments. These feasibility issues provided the anchor-point for the development of the integrated modeling and experimental program discussed below.

As part of the process of developing this document, the authors circulated a draft version of this document to the larger materials science community to solicit comments. That draft version also provided the basis for discussion at an open workshop that was held to obtain further input from the community. A summary of the workshop is provided in Appendix A. This final version of the whitepaper attempts to incorporate the comments that have been received. Although the conclusions and recommendations represent primarily the views of the authors, they have been assisted by an ad hoc peer-planning group of other AMP-funded scientists: G. E. Lucas (UCSB), K. Natesan (ANL), S. J. Zinkle (ORNL), A. F. Rowcliffe (ORNL), and R. J. Kurtz (PNNL). Contact information for the authors is given in Appendix B.

2 INTEGRATION OF MODELING AND EXPERIMENTAL ACTIVITIES IN ADVANCED MATERIALS PROGRAM

The overall strategy is a commitment to developing physically-based models that can be used to address the materials feasibility issues in the AMP White Paper, e.g. to make predictions of key mechanical properties starting in the as-fabricated state and evolving as a function of prolonged service in a fusion environment. Including consideration of the pre-service properties forms a strong and interactive link to material design and the influence of the entire thermal-mechanical history experienced by a material in a component, from initial processing though fabrication of integrated structures. It also serves as a basis to develop the requisite fundamental microstructure-property and basic property-engineering property models.

In-service effects will be mediated by a combination of factors including temperature, stress, the chemical environment and the neutron flux, fluence and spectrum. It is well known that there are synergistic interactions among this multitude of variables; and that complexity is further amplified by the overall temporal history of the service environment. Thus as a reasonable and necessary expedient, this effort will start with relatively simple models for selected properties, with ongoing development leading to more robust and comprehensive models as the knowledge base is improved. Finally, the integrated modeling program should also establish links with researchers involved in the development of advanced design and structural integrity assessment methods. For example, a better understanding of the micromechanics of the ductile-to-brittle transition can be obtained by linking our understanding of both atomic scale processes and embrittlement phenomena to an engineering code-based structural integrity assessment such as the master curve method.

Generally, the underlying physical basis of these models developed under OFES funding will be microstructural, including the mechanisms that lead to the production, transport and fate of defects and key constituent species as a function of the material and irradiation-service variables. The microstructure must be linked to basic structure-sensitive properties, like the yield stress, and such basic properties must in turn be linked to other engineering properties, like fracture toughness. Microstructural information and mechanical property data developed within the experimental component of the AMP, as well as relevant data generated by other programs such as Basic Energy Sciences (BES), the Nuclear Regulatory Commission (NRC), and the wider materials community, will provide the basic information needed for model development and verification. This implies developing and applying:

- integrated multiscale models of microstructural evolution and material properties
- mechanistic models describing key phenomena
- an appropriate mix of mechanical property testing and microstructural characterization tools
- experiments to investigate specific mechanisms (e.g., to explore helium transport, fate and consequences)
- verification of model predictions using the results of integral database experiments
- physically-based, engineering correlation models from evolving databases

A key product commitment is the development of mechanical performance maps (similar to Ashby maps) by systematizing and condensing the information in the knowledge base. This will also help to provide an interface between materials research and engineering design studies.

Clearly the long-term tasks outlined above are an enormous challenge and cannot be fully, or even primarily, funded by OFES. Thus, a critical part of this effort will be establishing links to the expanding general knowledge base regarding the microstructures, properties, and performance of materials to fusion specific applications. A high priority should also be given to developing and enhancing interactions between OFES-funded research and other research efforts in the larger materials science community. Support for new research efforts should require effective awareness and expertise in the larger field of materials science, while at the same time demonstrating a useful fusion focus. The efficient use of resources also requires that the research groups funded by OFES collaborate effectively. This will ensure that information flows freely and will eliminate unnecessary duplication of effort. Thus, an additional commitment is to improve the framework for collaboration between those institutions and individuals that are funded by the program. This involves cooperation and the development of partnerships between the national laboratories as well as between the laboratories and university programs.

3 FUNDAMENTAL MATERIALS ISSUES IN THE FUSION REACTOR ENVIRONMENT

As noted above, an effective integrated modeling effort cannot focus solely on effects of the radiation environment. Not only do many other factors in the service-environment itself affect material performance and lifetimes (e.g., fatigue loading, environmentally assisted cracking,...), but a myriad of other issues such as materials processing, fabrication and joining, developing functional coatings, fracture and structural mechanics need to be considered. However, it is also clear that the microstructural changes induced by the radiation environment and, hence, corresponding changes in the constitutive and fracture properties are an overarching consideration. Further, because of limitations in available irradiation facilities, assessing long-term high-fluence radiation effects in a fusion environment represents perhaps the most difficult challenge. Given this circumstance and the practical limitations of preparing this brief background paper, radiation effects, and the unique aspects of the fusion environment, will be the primary focus of the following discussion.

3.1 Radiation Effects Issues

3.1.1 DT fusion radiation environment

The basic irradiation environment in the high neutron flux regions of a fusion system is in many ways similar to that experienced by the in-core components in a high flux fission reactor. Thus many of the phenomena noted above can, and indeed have been, studied in fission reactors. Unfortunately, however, there are significant differences between the neutron spectra in fission and fusion systems that must be considered. It is expected that early fusion systems will generate energy by the fusion of deuterium and tritium (DT). The primary differences between the radiation damage source term in the two environments are related to the fact that the DT fusion neutrons are all born at 14.1 MeV, while the fission source exhibits a spectrum with a peak near 2 MeV. The neutron spectra in both cases cover a wide range of energy as a consequence of scattering reactions. However, the presence of higher energy fusion neutrons has two major impacts: a) higher energy primary atomic recoils; and b) high nuclear transmutation production since many such reactions have energy thresholds well above 2 MeV. In particular, the rate of helium and hydrogen generation by (n,α) and (n,p) reactions is on the order of 10 to 100 times higher in the DT fusion case.

Since fission reactor experiments are a major source of radiation effects data, much of the previous experimental and modeling work in the fusion materials program has been aimed at determining the impact of the higher energy DT neutrons. The impact of higher energy atomic recoil cascades has been most successfully examined by molecular dynamics (MD) simulations of displacement cascades. Considerable evidence based on simulations now supports the view that the net effect of spectral differences between fission and fusion may be rather small. This conclusion is also supported by various experimental evaluations that suggest dpa is often a good correlation parameter to account for spectral effects arising from differences in primary displacement damage production. However, the MD results provide information on the radiation damage source term only for times up to ~100 picoseconds. Understanding the correlated local evolution of the nascent cascade damage over times as long as gigaseconds, and the consequences of this process to long-time and long-range defect transport and fate is a key unresolved issue. Further, subtle differences between cascade damage in fusion versus fission environments may be amplified at these longer times. The resolution of these issues requires the use of techniques such Monte Carlo (MC) simulations to examine the evolution of the cascade residue for times long enough to permit diffusion of and reactions between the point defects.

Finally, it should be mentioned that there is no single “fusion irradiation environment.” The neutron flux level and the energy spectrum change significantly as the distance from the plasma increases due to neutron scattering and absorption. The flux and spectrum are also influenced by the different neutronic characteristics of the various coolants (water, helium, or lithium) and structural materials (iron-based, vanadium-based, SiC) that may be employed. These flux and spectrum changes are particularly important for assessing the anticipated effects of transmutation products. Thus, the irradiation environment for any one location in a given fusion reactor design must be evaluated in detail to determine how different the conditions are from those available in a fission reactor environment or other available irradiation test facilities.

3.1.2 Modeling radiation damage

The high energy neutron bombardment leads to elastic collisions with atoms which are in turn ejected from their crystal lattice positions (atomic displacements), and the production of transmutant products by nuclear capture reactions. Insoluble helium and reactive hydrogen gases are major transmutation products. Typical end-of-life fluences for fusion structural materials will lead to 200 displacements per atom (dpa), 2000 appm of helium, 20,000 appm hydrogen and percent-level production of solid transmutants. These “primary damage” products act in tandem with thermally and stress-driven processes to produce profound changes in the pre-existing microstructure (e.g., dislocations, precipitates, lath boundaries, ...). In addition, radiation damage leads to the formation of new microstructural features such as dislocation loops, voids, bubbles, regions of chemical segregation, and radiation-induced or enhanced precipitate phases that are often new and non-equilibrium in character.

The processes controlling microstructural evolution are mediated by a complex combination of atomistic level dynamics, defect physics, non-equilibrium thermodynamics, and transport kinetics. Relevant length scales range from atomistic to continuum, and time scales from femto to gigaseconds. The microstructural changes often lead to severe degradation of a wide range of important mechanical properties, as well as significant dimensional instabilities. Radiation-induced changes in physical properties are generally not a concern for metals, but changes in properties such as thermal and electrical conductivity will be important for ceramic components as discussed in section 3.3.

The ultimate aim of radiation materials science is to understand the production, transport, fate and consequences of all defect and material elemental species. This requires an integration of sophisticated theory, models, mechanistic experiments and comprehensive engineering databases. Understanding of key microstructural processes is the underlying requirement for developing practical methods to accurately assess the in-service performance under irradiation, and to develop more radiation stable materials. For example, superior performance can be achieved by enhancing the recombination of displaced atoms and vacant lattice sites, and by controlling the distribution of helium. Such understanding requires combining atomistic (e.g., electronic, MD and MC) and mesoscopic models (e.g., rate theory) with high-resolution microanalytical characterization measurements and specially designed single variable experiments. Further, modeling the consequences of microstructural evolution on property changes requires developments at the frontiers of the science of deformation, fracture and fatigue.

The production of primary defects and aged cascade features is only one small, albeit critical, part of irradiation induced/enhanced microstructural evolution. Long range transport of defects leads to: a) additional clustering (vacancies, interstitials and gases) and extended defect formation; b) defect annihilation, that may or may not change the character of the corresponding sinks (e.g., increases or decreases in network dislocation density); and c) rearrangement of solutes accelerated by radiation enhanced diffusion or driven by coupling with persistent defect fluxes (e.g., the inverse Kirkendall effect). A key feature of these processes that has received very little attention in most previous radiation damage studies to date is the profound role of alloy chemistry. All the post cascade formation stages of these processes are likely to be affected by details of the interaction of multiple atomic species. For example, it has been shown that even small defect clusters formed in cascades are complexed with various solutes. Thus it is critical that atomic level descriptions of alloy thermodynamics and kinetics be developed and coupled with mean field rate theory models. The former can be accomplished by combining appropriate models of interatomic interactions, like the embedded atom method, with MD, MC and similar simulation tools. Treating long range interactions (e.g., strain fields) is another major challenge that will require the application of new tools like domain decomposition methods.

Rate theory is based on spatially and short time averaged solutions to the production, diffusion, drift, reaction equations that describe conservation of all pertinent species. These typically take the form of large sets of coupled ordinary differential equations, where the pertinent physics is included in the coefficients for the various reaction terms, invoking the assumption of local equilibrium. It should be noted here that there is a natural synergism in such atomistic-continuum couplings. Specifically, while rate theory has been shown to be a powerful tool in understanding effects of irradiation on microstructures, it has three main drawbacks:

a) computational challenges associated with solving large sets of coupled equations;
b) substantial uncertainties in the averaged reaction coefficients; and
c) all the key physics must be input in the model equations and coefficients.

If some of the relevant physics is missing, it will not be revealed in the solutions to the rate theory equations. Some of the barriers to rate theory are dissolving in the face of the expanding capabilities of computers and more sophisticated mathematical methods of computation. Electronic-atomistic methods can now be used to provide much-improved estimates of not only the rate theory coefficients themselves, but also insight into the key physics and phenomena that must be modeled.

Ongoing model development may lead to a healthy competition between the rate-theory-based models and more atomistic approaches, such as MC simulations. Computational advances are extending the reach of MC into larger length and longer time scales, while the rate theory is being enhanced by increasing physical robustness and the computational tractability of solving large scale simulations. Both approaches are likely to be important sources of new insights and their hybrids will provide the basis for developing powerful microstructural evolution models.

3.1.3 Helium effects

Returning to the unique aspects of the fusion environment, a variety of techniques have been applied to examine the role of transmutant helium on microstructural evolution and mechanical property changes. Although some insight has been obtained, key aspects of the behavior and effects of helium remain unresolved. The effects of transmutant helium (and to a lesser extent hydrogen) can be synergistic with displacement damage. Helium stabilizes small vacancy clusters; depending on other factors, this may lead to either more or less void swelling and a potential reduction in creep rupture times and ductility due to enhanced grain boundary cavitation. The effects of helium on creep rupture are an example of a direct connection between microstructure and properties. In this case the combination of applied stress, temperature, helium, matrix hardening, irradiation and thermal creep all combine to influence this key property.

Helium management, generally accomplished by trapping a fine distribution at stable sinks, is a major objective of alloy development efforts, and leads to a belief that the effects of helium may be greater in single-phase alloys, particularly at high temperatures. There is also some puzzling evidence that high helium levels may produce low temperature embrittlement in excess of that associated with hardening. Thus, understanding the generation, transport, fate and consequences of helium transmutation remains a very high priority that cuts across material types. Similar arguments may be pertinent to environmental and transmutant induced hydrogen; however, the high mobility of this element may limit its consequences.

3.2 Performance Related Mechanical Properties in Metallic Alloys

While understanding and modeling microstructural evolution is a necessary condition for developing useful physically based models, it is far from sufficient. It is also necessary to develop models of how the altered microstructure influences the basic constitutive properties and local 'fracture' properties that control material separation by both ductile and brittle processes. This information must in turn be related to continuum measures of engineering mechanical properties, like fracture toughness, and ultimately to structural load and displacement capacities. Even when fully focused on fusion specific issues, this subject is too large to be reviewed here. Rather, selected targets for developing microstructure-property and property-property models are given. Indeed, in several of these cases there has been a great deal of recent progress.

3.2.1 Radiation induced elevations of the yield stress.

At low strains, obstacle hardening due to precipitates, cavities, defect cluster-complexes and dislocations in the form of loops and a dislocation network can be reasonably modeled by combining assessments of the individual strengths of the obstacles with models of how dislocations move though arrays of such features. Key unresolved issues include: a) details of dislocation-obstacle interactions, b) dislocation self-interaction effects, and c) multiple dislocation effects.

3.2.2 Static post-yield constitutive properties and deformation patterns.

Engineering alloys normally strain harden and can be well modeled at a continuum level by relatively simple flow and plasticity laws. However, post yield behavior following irradiation is often characterized by softening and highly localized flow in coarse slip regions and, in extreme cases, in flow channels. The macroscopic consequences of such strain softening and flow localization range from negligible uniform elongation in tensile tests to severe reductions of toughness associated with fracture by shear band decohesion. This problem is very rich, involving the effect of local plasticity on dislocation obstacles, the dynamics of dislocation structure formation, strain induced damage generation and dilatational plasticity, and internal stress and multiaxial stress state redistributions.

3.2.3 Swelling and irradiation creep

It seems unlikely that dimensional instabilities in fusion structures can be avoided due to void swelling and irradiation creep. The basic mechanisms of these phenomena are reasonably well understood. In the simplest view, both arise from biased partitioning of vacancies and interstitials to various sinks. In the case of swelling, excess vacancies flow to growing voids; in the case of creep, excess interstitial fluxes flow to stressed dislocations as a function of their Burgers vector. Swelling and creep are interactive as demonstrated by phenomena such as stress assisted swelling. Although irradiation creep appears to be ubiquitous, some alloys fortunately appear to be swelling resistant. However, a number of major issues remain to be fully resolved, including:

a) the effects of high helium levels and consequential interactions with displacement damage;
b) the mechanisms which give rise to apparent swelling resistance in some bcc alloys and the persistence of this resistance;
c) interactions between swelling and phase instabilities;
d) the role of defect cluster-complexes formed in cascades, and the properties of these complexes; and
e) the effects of stress and stress sate.

Since swelling and creep are directly linked to microstructural evolution, resolution of these issues will require a combination of tools listed above.

3.2.4 Thermal creep, creep rupture and creep crack growth

At temperatures above about 40% of the absolute melting point, thermal creep, creep ductility and rupture times are important. The influence of irradiation on thermal creep rates depends primarily on changes in the microstructural features that act as obstacles to dislocation climb and glide. A more serious issue is the effect of high helium levels on rupture times and ductility because of its influence on fracture mechanisms. Helium promotes the nucleation of high concentrations of grain boundary cavities that become unstable to growth at lower stresses and coalesce at smaller strains than coarser populations of normal creep cavities. The presence of stress concentrations at the tips of growing creep cracks could lead to very short time, low ductility failures. Simple models focusing only on cavity growth are inadequate; the effects of local microstructures and deformation processes in the matrix, near boundaries, and in the boundaries themselves play key roles. Inhomogeneous deformation processes lead to complex internal stress and stress state redistributions. Thus, better understanding will require the application of sophisticated multiphysics models that can deal with combined geometrically and constitutively complex defect flow-deformation processes.

3.2.5 Fast fracture and the ductile-to-brittle transition temperature (DBTT)

There are two types of low toughness fracture processes that are of concern. The first is associated with loss of strain hardening/ductility and flow localization; in the extreme this may lead to very low toughness and shear band decohesion. Progress will require understanding the microstructural conditions that mark this regime, developing basic constitutive laws for materials in this state, and large deformation modeling of local processes leading to material separation. Elevation of the

DBTT is caused either by hardening or a reduction in the local tensile stresses required to produce cleavage or intergranular fracture. For example, the latter may occur due to segregation of elements such as phosphorous at grain boundaries, which may be accelerated by radiation-enhanced diffusion. The presence of hydrogen (from various sources) may also lead to this type of embrittlement. Progress will require continued development of combined micromechanical-macromechanical models of brittle fracture based on detailed observations of local fracture processes. These efforts should also seek close and direct ties to advanced structural integrity assessment methods based largely on small specimen testing.

3.2.6 Fatigue, creep fatigue and fatigue crack growth

Given the anticipated extreme thermal-mechanical loading conditions, avoiding fatigue related failures in complex fusion structures represents a tremendous challenge even when the complications of the effects of irradiation are not considered. One part of the challenge is that "fatigue" has numerous manifestations (for example, S-N limits in both low and high cycle ranges, da/dN rates, thermal-mechanical local and global ratcheting, and creep fatigue, just to name a few). Each of these types of fatigue are, in turn, controlled by a large number of intrinsic (material) and extrinsic (loading-geometry) variables. In fact, fatigue failures are very sensitive to details of the structure as well as the underlying material properties. Some, and perhaps most, multiphase alloy systems that have a useful balance of properties tend to experience cyclic softening and high rates of mechanical damage formation. Of course radiation-induced changes in the microstructure, basic constitutive properties (including irradiation creep) and deformation patterns may have a profound effect on the various types of fatigue processes. Given this tremendous complexity and the relative paucity of pertinent data, it is recommended that fatigue not be a high priority target of early-integrated modeling research. However, experiments aimed at generating engineering type data should be carried out with an eye to the ultimate development of microstructure-mechanism based fatigue models.

3.2.7 Environmentally-assisted cracking

The chemical environment often has very large deleterious effects on fracture and fatigue processes, enhancing crack initiation and subcritical growth. However, these effects not only involve multiple physical mechanisms that are complex and poorly understood, but are also often very system specific. Further, while there may be significant effects of the radiation environment, the relevant database for fusion alloys is very limited. Thus as in the case of fatigue, it is recommended that integrated modeling research on environmentally assisted cracking should not be an early priority for the initial integrated modeling effort.

3.3 Other Significant Materials Issues

The previous discussion has focused on metallic alloys and their microstructural and mechanical stability in a fusion environment. However, there are many other opportunities for an integrated modeling effort dealing with issues such as:

a) ceramic composites and the stability and functionality of their engineered constituents;
b) insulator and barrier coatings;
c) corrosion and system level corrosion product transport;
d) the kinetics and consequences of impurity pick-up;
e) processing, fabrication and joining;

f) detailed component-level integrity-lifetime assessments that involve many phenomena (neutronics, magnetics, thermalhydraulics, stress and inelastic deformation) when the materials are used in a complex, time-dependent stress state arising from gradients in the damage and thermal and mechanical loads.

3.3.1 Silicon carbide composites

SiC composite materials have been considered an attractive possible fusion structural material for primarily two reasons. The first reason is their potential for use at high temperatures with a concomitant increase in the thermal efficiency of the system. Successful high temperature application will require that the thermal conductivity of the material be high enough to limit stresses induced by thermal gradients. The thermal conductivity of current commercial materials is not sufficiently high due to the presence of impurities, and irradiation will lead to reductions in the thermal conductivity. The second desirable feature of SiC is its neutron activation and decay properties. The relatively low level of induced activity in SiC minimizes the amount of decay heat that must be removed after reactor shutdown. In addition, the induced radioactivity of pure SiC components would quickly decay, leading to possible hands-on maintenance of the structure within several days. However, the level of purity than can be obtained in commercial materials also limits this potential benefit. Even modest levels of metallic impurities significantly raise the level of induced radioactivity.

A short summary of current key issues for SiC composites would include:

a) fiber and coating stability and the consequences to bridging mechanics,
b) hermeticity and sealing coatings
c) swelling and creep (at high helium levels)
d) radiation-induced reductions in thermal conductivity,
e) joining; and
f) the integrity of complex, thermal-mechanically loaded brittle structures.

The role of nuclear transmutation, potentially leading to a high level of SiC burnup in service also warrants further investigation.

It is recommended that near-term modeling is best directed toward developing an understanding of the response of monolithic materials. A coordinated effort of fundamental modeling and experiments is needed to determine parameters such as the atomic displacement threshold on both the Si and C lattices, point defect formation and migration energies, and point defect binding energies for helium and hydrogen.

3.3.2 Ceramic coatings

The blanket system is one of the most important components in a fusion reactor because it has a major impact on both the economics and safety of fusion energy. The primary functions of the blanket in a deuterium/tritium-fueled fusion reactor are to convert the fusion energy into sensible heat and to breed tritium for the fuel cycle. In fusion applications, the structural material of the system must be capable of maintaining structural integrity for long periods while exposed to moderately elevated temperatures, mechanical loads, thermal cycling, and possibly intense irradiation. The material must be compatible with the coolant, which may water, helium, or a liquid metal

A major challenge in the design of liquid-metal-cooled blankets for use in high magnetic field tokamaks is the accommodation of the strong influence of the magnetic field on coolant flow. If the flow direction is perpendicular to the field, a potential difference across the duct is induced in the liquid metal. This can cause a large electrical current flow if the potential difference is short-circuited by the duct walls. An electrical current flowing perpendicular to a magnetic field leads to a mechanical force that causes a magnetohydrodynamic (MHD) pressure drop. It has been shown that even thin conducting walls would lead to a rather high pressure drop under the conditions of a fusion reactor blanket. For example, the pressure drop in a poloidal duct in an inboard blanket segment would reach 8.6 MPa if the conducting liner was 0.1 mm thick. This unacceptably high pressure drop shows the need for electrically insulating coatings in contact with the flowing liquid metal. It has been shown that a perfectly insulating coating on the wall would decrease the pressure drop from 8.6 to 0.22 MPa.

The current focus of insulator coating research is for the self-cooled lithium blanket with a vanadium alloy structure. Primary criteria for selection of candidate coating materials include the following:

- chemical stability/compatibility of the coating with both the vanadium alloy and the lithium of normal or specified practical purity at elevated temperatures
- high electrical resistivity
- potential for coating of complex channel geometries
- potential for *in situ* self-healing of any defects that might occur in the coating under prototypic operating conditions.

Other considerations important to the development of insulating coatings for the fusion applications include:

- a thermal expansion coefficient similar to that of the vanadium alloy structure
- low-activation characteristics
- minimal chemical toxicity
- low cost and adequate resources

In the case of a lithium-vanadium system, considerations of chemical compatibility and electrical resistance appear to limit the options for coating materials to AlN and CaO. It has been demonstrated that these materials can have adequate insulating capacity in thin layers, but it has not been shown that they will exhibit long-term stability in a thermal gradient. Methods of applying such coatings with adequate mechanical integrity and adherence, achieving and retaining electrical insulating capability, and a practical approach to producing a self-healing system are still to be developed.

3.4 Issues related to development or use of alternate materials

It is anticipated that the AMP will continue to focus on selected feasibility issues identified with four primary material systems (see Section 4 below). However, several factors motivate some interest in alternate materials. The first is the possibility that the targeted materials will prove to be unacceptable for their proposed applications. Secondly, future changes in the configuration, mechanical design, or operating parameters of proposed fusion reactors could alter the choice of materials. Finally, materials development efforts in other fields could yield new materials that better meet the needs of the fusion program.

This highlights the importance of maintaining active ties with the wider materials research community in order to leverage developments outside the purview of OFES. It is in the best interest of OFES to keep this larger community informed of the desired properties and service requirements of potential fusion reactor materials and to monitor the progress of research being conducted outside the program. At the same time, researchers within the program should make use of these outside contacts to maintain an awareness of where OFES-funded advances in basic materials science will have a larger application.

3.5 Links to Experiment

As emphasized in the beginning, all integrated modeling efforts must be closely linked to experiments of various sorts. To reiterate, these experiments must include:

1. Focused studies of key phenomena and mechanisms, including those required for model development and validation. An obvious example is special experiments to understand how helium diffuses and is trapped under irradiation.

2. Controlled experiments on carefully conditioned samples, specially designed to reveal the effects of material and irradiation variables, both singly and in synergistic combinations. For example, doping with iron, boron and nickel isotopes, spectral tailoring, and the tritium-trick can be used to systematically vary the He/dpa ratio for irradiations over a range of temperatures and fluences and for various alloy systems.

3. Integral experiments to develop a database for alloy selection and optimization, and ultimately, engineering design.

Since the integrated modeling effort aims to have a microstructural foundation, development and application of high-resolution microanalytical characterization techniques, such as atom probe, electron microscopy and radiation scattering, will be very important. Further, understanding of the consequences of microstructural evolutions to property changes will require development and application of small specimen testing methods.

4 CURRENT PRIMARY STRUCTURAL MATERIALS: CROSSCUTTING PHENOMENA AND MECHANISMS

Research in the fusion materials program is currently focused on three primary alloy classes: advanced ferritic-martensitic steels, vanadium alloys, and silicon carbide composites. In addition, copper alloys have a role in high heat flux components and as a potential structural or magnet material in specialized reactor designs. However, a number of key phenomena crosscut multiple material systems. Thus coordinated research with emphasis on these would have a highly leveraged impact.

Examples of these crosscutting issues are listed in Table 4.1, where an “x” in the table indicates that the phenomenon is relevant for the indicated material. Several of these were also identified as critical feasibility issues in the AMP white paper; these are indicated by a “W” in the table. The metallic alloys share more issues in common among themselves than does SiC with the metals.

Furthermore, even in cases where an item has been identified as common between the metals and SiC, e.g. welding and joining, the details may be quite different for SiC. All these areas, and in all likelihood others not included, are good targets for enhanced integrated modeling. The importance of the first four issues, and good prospects for making progress in dealing with them, make these issues our top priorities for initial emphasis in an integrated modeling program. No additional prioritization should be inferred by the order of the remaining issues in the table.

We note that the second item in Table 4.1, flow localization, was not mentioned as a critical feasibility issue in the AMP Whitepaper for any of the material systems. We include it here as a high priority issue because of the growing realization that the phenomenon is more general than previously thought. It appears that flow localization may limit the performance of most structural

Table 4.1 Key Crosscutting Phenomena for Fusion Reactor Materials *

FM	V	Cu	SiC	Phenomena, Issues, Comments
W	W	W	-	hardening and nonhardening embrittlement including underlying microstructural causes and the effects of helium on fast fracture
x	x	x	-	flow localization, consequences and underlying microstructural causes
-	W	-	x	coatings, multilayers, functionally graded materials
W	W	-	x	helium effects on high temperature deformation and fracture, and development of improved multiphase alloys for helium control
x	W	x	x	thermal and irradiation creep
x	x	x	W	swelling and general microstructural stability
x	W	x	W	welding, joining and processing issues
x	x	x	x	fatigue
x	W	-	-	hydrogen and interstitial impurity effects on deformation and fracture
-	-	-	W	physical properties, e.g. thermal conductivity
-	-	-	W	permeability of gases
x	x	x	x	erosion, chemical compatibility, bulk corrosion, cracking, product transport

* x – relevant phenomenon, W – phenomenon identified in AMP White Paper

alloys under certain irradiation conditions. Developing an understanding of flow localization, and how to prevent it or mitigate its consequences will be directly relevant to the use of all the current target materials and to any alloy development work that may take place in the AMP.

The role of both equilibrium and nonequilibrium alloy thermodynamics could also be considered a crosscutting phenomenon. This topic was extensively discussed at the open workshop, and has significance to radiation-induced microstructural evolution and mechanical property changes as well as alloy development. The importance of thermodynamics has not been explicitly acknowledged in much of the previous radiation damage modeling work, although it may have been accounted for through the use of “effective” material parameters in these models.

Thermodynamics is not listed in Table 4.1 because it is qualitatively different than the listed phenomena, but an increased emphasis on understanding alloy thermodynamics and more detailed theoretical treatments of its impact in fusion reactor materials should be encouraged. We can note that research carried out under the auspices of the Steel Research Group at Northwestern University provides a good example of such work.

5 SUMMARY RECOMMENDATIONS

Based on the previous discussion we offer the following recommendations:

- An ambitious integrated modeling program should be initiated and closely linked to the entire AMP program.
- Program planning should consider the items identified by the workshop participants described in Appendix A and those listed in Table 4.1
- The program should encourage inter-institutional collaborations, i. e. between national laboratories and universities as well as between national laboratories.
- Regular program exchanges and reviews should be an integral part of the program.
- Both new and ongoing research should demonstrate relevance to the issues described in this document and the overall AMP White paper.
- When evaluating program research, several factors should be considered; these include:
  a) scientific quality and innovation of the researchers and their ideas,
  b) demonstrated focus on the key issues in fusion materials,
  c) close integration with other elements of the AMP program and, as noted above, specific collaborations between institutions,
  d) likelihood of tangible useful results including more reliable predictions of properties and/or improved design and methods,
  e) leveraging from other associated research activities, and
  f) an effective awareness of and expertise in the larger fields of materials science and engineering.

Appendix A Description of Planning Workshop

A.1 Workshop Scope and Agenda

Workshop on Integration of Theoretical and Experimental Research
in the U.S. Fusion Materials Program

December 3-4, 1998

Sheraton Boston Hotel
Independence East Room
Boston, MA

The purpose of this workshop is to obtain input to further develop the AMP whitepaper on the integration of theory/modeling and experiments. The workshop is open to anyone working in a relevant area of research, but the format is intended to focus discussion on the draft version of the roadmap in order to highlight areas where it may be improved. The breakout sessions and contributed talks are expected to identify any shortcomings or omissions in this document. Research presented in the contributed talks should specifically relate to the fusion materials feasibility issues described in the draft roadmap.

Thursday, December 3

1:00 pm Session 1: Introduction and Overview of Issues

- OFES plans and activities - F. W. Wiffen (U.S. DOE)
- Summary of AMP White paper - E. E. Bloom (ORNL)
- Summary of FESAC Panel Report and Planning Process - R. E. Stoller (ORNL)
- Initial questions and comments on planning process and whitepaper
- Invited Examples of Modeling-based Materials Development
  - C. J. Kuehmann, QUESTEK: Computer-aided Materials Design
  - T. Diaz de la Rubia, LLNL: Applications of Multiscale Modeling
  - G. R. Odette, UCSB: Investigation of Embrittlement in RPV Steels

3:30 pm Break

4:00 pm Session 2: Breakout of Working Groups

- Microstructural processes and helium effects, A. F. Rowcliffe (ORNL) and B. D. Wirth (LLNL)
- Deformation and fracture, G. E. Lucas (UCSB) and A. El-Azab (PNNL)
- Coatings and compatibility, K. Natesan (ANL) and G. S. Was (U of MI)
- Ceramics and composites, W. J. Weber (PNNL) and L. L. Snead (ORNL)

Friday, December 4

8:30 am Session 3: Working Group Reports and Discussion
- Working group chairmen reports (as above)
- Moderated discussion

10:15 am Break

10:30 amSession 4: Open Forum (As contributed)
- John Vitek, ORNL, thermodynamic databases, a neural net application

11:30 amWorkshop wrap-up, closing remarks: Workshop organizers

A.2 Workshop Discussion and Breakout Sessions

Invited presentations at the workshop provided examples of model-based materials development, applications of multiscale modeling in the semiconductor industry, and the interactive role of theory and experiment in successfully dealing with an applied materials problem. These examples illustrated the recent progress in our ability to use modeling and computer simulations to develop materials and to solve “real world” problems. A substantial portion of the workshop was devoted to four breakout sessions that will be summarized below. Themes that were emphasized throughout the meeting included the increasingly general observation of flow localization in irradiated structural materials, the potential usefulness of a greater role for computational thermodynamics, and a materials engineering approach to designing radiation-resistant materials. Overall, the discussion in both the breakout and general sessions was generally supportive of the approach described in the draft white paper, i.e. that an increased level of modeling work may lead to substantial progress when it is well integrated with a focused experimental activity.

A.2.1 Microstructural processes and helium effects

Discussion in this session focused primarily of four topics, primary damage production and damage accumulation, helium management, computational thermodynamics, and alloy development. With respect to damage production and accumulation, it was recommended that alloy effects should be addressed. This will require the development of interatomic potentials for concentrated alloys (e.g. Fe-xCr, V-xTi), and ways to implement the effects of alloy thermodynamics in the parameters used in kinetic models employing both the rate theory and Monte Carlo techniques. The effect of displacement cascades on the stability of oxide particles is an unresolved issue for the ODS ferritic alloys, and for precipitate phases in general. Atomistic modeling tools should be employed at the appropriate level and be used to provide information to higher level models. Further modeling of helium interactions in the lattice (migration, trapping and sinks and interfaces) must be closely tied with experiments to aid understanding and model development.

The current successful use of thermodynamic models to treat multiphase steels was extensively discussed. Work should be initiated to develop a similar level of sophistication for the alloys of direct interest to the AMP. In particular, the effects of persistent point defect and solute fluxes must be incorporated into the thermodynamic models in order to predict phase stability under irradiation. The approach of engineering materials to produce radiation-resistant microstructures was suggested for any alloy development work that goes on in the program. For example, modeling and focussed experiments could be used to identify and understand effective sinks for helium, and such information could be applied in the development of microstructures that will mitigate the effects of helium by trapping. Other materials problems that may yield to microstructure-design-based solutions include developing grain boundary structures that are creep resistant and the ability of a material to maintain its strain hardening capability under irradiation.

The development of innovative materials to expand the current limits on maximum damage level and operating temperatures was discussed in the context of recent advances in the computational design of materials. In other fields, systems approaches have been successfully used to integrate processing, structure, properties, and performance relationships into the design of multilevel-structured materials. Similar approaches could provide far-reaching benefits to the fusion program.

A.2.2 Deformation and fracture

Several aspects of material flow and fracture behavior were discussed, the outstanding issues in each area were described, and modeling-based approaches to develop an understanding of these phenomena were proposed. Obtaining a sufficient degree of knowledge about the predominant microstructure is a significant (although primarily experimental) issue for improving our understanding of most aspects of a materials mechanical response. Outstanding issues with respect to the yield strength, or yield strength change, of materials include a better definition of the barrier strengths of various defect structures and the development of improved superposition laws for realistic microstructures that contain multiple barrier types. These can be investigated by an appropriate combination of atomistic and mesoscale models of dislocation dynamics and dislocation-barrier interactions with key experiments on both irradiated and surrogate materials. Knowledge and understanding of post-yield constitutive behavior is also important, and the loss of work hardening and the onset of flow localization head the list of important post-yield phenomena. A similar set of theoretical tools may be used to investigate how dislocation obstacles are destroyed, leading to the formation of clear channels for deformation. The critical conditions for the onset of channeling should be determined to help guide efforts to design microstructures that will resist this phenomenon.

Although thermal creep may not be an issue for many fusion applications, irradiation creep and creep-swelling interactions will be important for most materials in the high flux regions near the first wall. Improved kinetic models are needed to describe and simulate the effect of stress on radiation-induced microstructural evolution. A combination of modeling studies and experiments should be used to develop material performance maps (Ashby maps) for the materials and irradiation conditions of interest to the AMP.

Issues related to fracture behavior include the need to obtain an improved understanding of the micromechanics of fracture (including cleavage, quasi-cleavage, ductile fracture, and intergranular fracture) and the relationship between micromechanics and the failure assessment of irradiated structures that contain flaws. The micromechanics of fracture can be investigated with a combination of phenomenological and finite element models with input from techniques such as fracture surface reconstruction. These same tools can be used to quantify local crack tip critical fracture conditions such as the critical stress and length scale parameters that control cleavage behavior. Similar to the topic of post-yield behavior mentioned above, it is important that accurate constitutive relationships be developed and used. Since two of the AMP-target materials exhibit a ductile-to-brittle transition, the mechanisms causing an increase in the DBTT must be elucidated. Since a DBTT shift can arise by either matrix hardening, a reduction of the critical fracture stress, or by intergranular fracture, determining the predominant mechanism(s) is essential for efforts to mitigate embrittlement.

A.2.3 Coatings and compatibility

Although the focus of fusion reactor research in this area has been on lithium-based coolants, it has built on the foundation of earlier work in the sodium-cooled fast breeder reactor program. The concerns for compatibility between a liquid metal coolant and the structural materials are similar, but the fusion reactor environment adds the requirement for an electrical insulating coating on the structure to prevent unacceptable pressure drops due to magnetohydrodynamic (MHD) forces. For the candidate system that employs lithium coolant and vanadium as the blanket structural material, there are several issues that need to be considered with respect to the compatibility of the coating involve. These include:

(a) the thermodynamic stability of the coating when contacted with the liquid lithium,
(b) the extent of dissolution of the coating in the liquid lithium, which is dictated by the solubility of coating constituents in liquid lithium under the temperature and temperature gradient that exist in the system,
(c) the degree of interaction between the coating constituents and other reactive elements in the liquid lithium, and
(d) compatibility relationships between the coating and the structural material.

For example, reactions with elements such as oxygen, carbon, nitrogen, and hydrogen may lead to changes in coating chemistry, thereby altering its insulating characteristics. Dissolution of structural material constituents can also be an issue if parts of the system are not coated or if the coating is damaged.

Nonmetallic elements such as O, C, N, and H are known to migrate in structural-material/liquid-metal systems as a result of differences in chemical activity. Since coating reliability is of concern, the program requires approaches to address several issues such as the thermodynamic stability, physical and chemical characteristics of the coating; the viability of different coating methods; and the insulating characteristics of the coatings before and after exposure to controlled purity lithium.

A previous review of electrical resistivity data for several oxides, nitrides, and mixed oxides showed that oxides such as CaO, MgO, SiO_2, Al_2O_3, $MgAl_2O_4$ and nitrides such as AlN and Si_3N_4 exhibit resistivities of $>10^5$ ohm·m at temperatures below about 600°C. The system requirement is for the product of the electrical resistivity and the thickness of the insulator coating to exceed a nominal value of 0.01 ohm·m^2 under operating conditions. This translates to a minimum resistivity value of 10^4 ohm·m for a coating thickness of 1 µm, or 10^3 ohm·m for a coating thickness of 10 µm. Based on the resistivity values of materials listed above, a coating layer of <1 µm in thickness of any of these materials would be adequate from the insulating standpoint, provided the resistivity is not reduced during operation, i.e., by irradiation or other operational parameters.

The electrical resistivity of two of the prime candidate coatings CaO and AlN has been assessed for application in a lithium environment. The effort examined the development of CaO coatings by thermal/chemical vapor deposition and *in situ* in lithium. Similarly, detailed investigations of AlN's fabrication and metallurgical microstructure, and preliminary studies of its compatibility with liquid lithium were conducted. The electrical characteristics of AlN material obtained from several sources were examined. Methods of applying thin coatings of CaO and AlN with adequate mechanical integrity and adherence, achieving and retaining electrical insulating capability, and a practical approach to producing a self-healing system are still to be developed

Theoretical modeling of the thermodynamics and kinetics of the chemical interactions between the coating and a lithium environment can aid in the in-situ formation of insulating coatings in lithium and can guide the experimental program to aid in the interpretation of experimental results. Much of the original analyses conducted to identify potential candidate coating materials were based on thermodynamic calculations to assess the stability of the coating materials and the chemistry requirements for both the liquid lithium and the vanadium alloy systems. More detailed calculations are necessary to assess not only the thermodynamic aspects of the coating processes, but also the kinetics of reactions such as transport rates of oxygen, calcium, aluminum, and nitrogen, etc.

Calculations of oxide or nitride growth rates can be used to estimate the ranges of temperature and time required for coatings of desired thickness to form. Calculations of the thermodynamic stability of various phases, e.g. CaO, CaV_xO_y, Ca-Mg-O, AlN, Al-O-N, $LiAlO_2$, that can potentially be formed need to be correlated with experimental observations and measured compositions of the liquid lithium and vanadium alloys. The dependence of electrical resistivity on composition in functionally gradient materials needs to be determined. Both modeling and experimental work are required to investigate the in situ formation and repair of ceramic coatings. Current models address primarily mechanical degradation such as cracking and spallation; the impact of mechanical degradation on the electrical properties of the coatings needs to be addressed. Finally, it is imperative that the coatings be evaluated in the presence of an irradiation environment from the standpoint of their chemical stability, mechanical integrity, and electrical resistivity.

A.2.4 Ceramics and composites

There is only limited experience applying ceramic matrix composites as structural materials, and there is considerable international effort currently being invested in this general area. Therefore, it was recommended that the fusion program adopt the strategy of focusing on those areas that are specific to the fusion environment. For instance, engineering design codes and micromechanical (constitutive) modeling are two examples of topics that will be critically important for the use of SiC composites in fusion applications. However, these are being aggressively pursued in other programs and therefore are not focus areas of the fusion materials program.

Areas of research which are of unique interest to the application of SiC composites to the fusion program are typically those dealing with the effects of radiation on monolithic or composite structures. A significant example is the potential degradation of thermal conductivity as a result of radiation exposure. While there has been some study of the effects of radiation on monolithic materials there is still considerable work remaining experimentally and theoretically. Some examples of areas which warrant study are: radiation damage production, transport and trapping of transmutation gases (e.g. H and He), basic defect properties such as activation energies for formation and migration, effects of various defect types on phonon scattering, and effects of swelling on mechanical properties. The goal of these studies would be to provide the knowledge base necessary to predict the performance of the composite material which is critically dependent on the evolution and interaction of its three components: fiber, matrix and interphase.

It is hoped that the process of developing an understanding of radiation effects and their potential impact on the constitutive properties will proceed in parallel with micromechanical model development outside of the fusion community. As this work matures, integration of these models should permit the evolution of thermophysical properties in SiC composites under irradiation to be predicted.

A.3 Workshop Participants and Affiliations

Participants: 42 participants on Thursday (28 Friday), 16 with current ties to U.S. DOE Office of Fusion Energy Sciences, 6 international

Affiliations:
- 7 from U.S. universities
- 25 from U.S. national laboratories
- 1 from U.S. commercial
- 3 from U.S. DOE Offices
- 3 from foreign universities
- 3 from foreign national laboratories or institutes

Appendix B Author contact information

Roger E. Stoller (chair)
Oak Ridge National Laboratory
Bldg. 5500, MS-6376
P. O. Box 2008
Oak Ridge, TN 37831-6376
phone: 423-576-7886
fax: 423-574-0641
email: rkn@ornl.gov

Howard L. Heinisch
Pacific Northwest National Laboratory
MS P8-15
P. O. Box 999
Richland, WA 99351
phone: 509-376-3278
fax: 509-376-0418
email: hl_heinisch@pnl.gov

G. Robert Odette
Department of Mechanical and Environmental Engineering
University of California
Santa Barbara, CA 93106
phone: 805-893-3525
fax: 805-893-8651
email: odette@engineering.ucsb.edu