Ongoing R&D Programs in Radiation Materials Science

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Joint IAEA-ICTP Advanced Workshop on Development of Radiation Resistant Materials

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1 Managed by UT-Battelle for the U.S. Department of Energy Radiation materials science in the US is being performed under a variety of programs

- DOE-Basic Energy Sciences
- DOE-NE Gen IV
- DOE-NE Advanced fuels and cladding initiative
- DOE-NE Light Water Reactor Sustainability program
- DOE-fusion energy sciences
- DOE-BES spallation neutron source



Development of Improved Structural Materials for Advanced Fast Reactors

- Improved structural materials have the potential to enable significant improvements in reactor performance
 - Economy: reduce capital costs through reduced commodities and simplifications
 - Flexibility: higher material performance allows greater options to designers
 - Safety: higher material performance promotes larger safety margins and more stable performance over longer lives
- The US is working to develop and qualify new structural materials for fast reactor applications.
 - Development of advanced alloys
 - \Mechanical testing (tensile, creep, fatigue, creep-fatigue, impact, fracture)
 - Environmental effects (aging, sodium, and irradiation)
 - Joining and fabrication technologies
- A new ASME Nuclear Codes and Standards working group on Liquid Metal Reactors has been instituted
 - Addresses code issues identified by NRC in Clinch River and PRISM reviews
 - Develops high temperature design methodology and code rules to support advanced fast reactor licensing



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NRC Review of Clinch River Breeder Reactor and PRISM

- NRC reviews of the CRBR and PRISM designs have identified concerns in over 20 areas.
- These have been reduced to nine key issues in materials and design methodology.
 - Weldment safety evaluation
 - Elevated temperature seismic effects
 - Design analysis methods, codes, and standards
 - Elastic follow-up in elevated temperature piping
 - Notch weakening
 - Creep-fatigue evaluation
 - Plastic strain concentration factors
 - Intermediate heat transport system transition weld
 - Steam generator
- Many of these issues remain today.
- Accounting for irradiation effects has also been identified in subsequent reviews.

[Griffin, D.S., "Elevated-Temperature Structural Design Evaluation Issues in LMFBR Licensing", Nuclear Engineering and Design, 90, (1985), pp. 299-306] [Huddleston, R.L. and Swindeman, R.W., "Materials and Design Bases Issues in ASME Code Case N-47", NUREG/CR-5955, ORNL/TM-12266, April 1993].



4 Managed by UT-Battelle Code Case N-4 for the U.S. Department of F[NUREG 1368]

Extending the service life of today's LWR fleet may create new challenges

- Safe operation of existing nuclear power plants beyond current license periods allows for a sustainable supply of clean and affordable electricity.
- Extending reactor life to beyond 60 years will likely increase susceptibility and severity of known forms of materials degradation and potentially introduce new forms of degradation.
- The LWR Sustainability Program will provide the scientific basis for understanding and predicting materials aging and degradation within components, systems, and structures.
 - Reactor metals (RPV's, internals, steam generators, balance of plant, and weldments)
 - Concrete
 - Buried piping
 - Cabling
 - Mitigation, repair, and replacement technologies
- A new working group is being formed to integrate the materials efforts within DOE's LWRSP, EPRI's LTO, and NRC's LB60 programs.



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Candidate materials for advanced fast reactors

- A variety of materials may be candidates for fast reactor structural and clad materials.
- All materials have advantages and limitations.
- A number of considerations will help determine the best candidate materials
 - Mechanical properties
 - Tolerance of environment
 - Irradiation tolerance
 - Availability
 - Cost
 - Code qualification (currently 5 materials)
 - Joining and fabrication
 - Neutronics



Candidate alloys: advanced austenitics

- Advanced austenitics such as Ti-stabilized 316 and D9 (15Cr-15Ni-Ti) offer improved performance (strength and creep) over 316 SS
- Austenitic alloys offer high oxidation and corrosion resistance
- Reduced swelling over SA 316 SS
- Swelling is still more than F/M steels.
- High Ni content will lead to higher He generation rates (stabilizes cavities).





Candidate alloys: superalloys

- Superalloys (Incoloys, Hasteloys, etc.) have superior high temperature strength to stainless steels.
- High Ni-contents lead to high He generation (which may stabilize voids)
 - Enhanced swelling
 - He-embrittlement
- Precipitation and phase stability may be a key issue.
- Dramatic reduction in creep strength/rupture time.







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Candidate alloys: advanced commercial steels for higher operating temperatures

- Commercial steels for higher temperatures
 - NF616: Fe-9Cr-1.8W-0.5Mo-0.06N-0.004B-0.1C
 - HCM12A: Fe-12Cr-1.8W-1Cu-0.5Mo-0.07N-0.1C
- Advantages of NF616 and HCM12A:
 - Increase in rupture strength at 600°C over HT9 and modified 9Cr-1Mo
 - Maximum use temperature of 620°C compared to 565°C for HT9, 593°C for modified 9Cr-1Mo
- Irradiation effects not determined for these advanced steels
- Alloys are not ASME code qualified



Candidate alloys: refractory alloys

- Refractory alloys (Mo, Ta, and Nb base alloys) offer extremely high melting temperatures.
- Strength and creep resistance are superior to other metallics at high temperatures.
- Refractory alloys also have excellent compatibility with alkali metals.
- These metals suffer extreme radiation embrittlement below ~700°C.
- The irradiation database for these materials is much less mature.
- Alloys are extremely sensitive to impurities.
- No refractory alloys are ASME code qualified.



Candidate alloys: nanostructured ferritic alloys (NFA)

 A high number density of nanoclusters dramatically improve the high temperature strength, including creep performance, and tolerance to neutron irradiation damage of iron alloys



3-DAP of 12YWT



• The creep rate of NFA is ~6 orders of magnitude lower than conventional steels at 600-900°C

NFA Have Remarkable Radiation Damage Tolerance

 MA957 following neutron irradiation to ~9 dpa at 500°C and implantation of up to ~380 appm He



- Irradiation effects database is not as mature as other alloys (ion irradiation data to ~100 dpa suggests good irradiation stability and tolerance)
- Joining and industrial scale up must be demonstrated
- Alloy is not ASME code qualified

Materials Science and Technology Division Oak Ridge National Laboratory



Candidate materials: SiC/SiC Composites

- Intrinsic properties for SiC:
 - Irradiation resistance
 - Heat resistance
 - Thermo-chemical stability
 - Low activation / low decay heat
 - Low gas permeability



- Engineered properties for SiC/SiC
 - Ductile fracture
 - Tailorable strength
 - Tailorable thermal properties





Status and Issues for SiC/SiC for Nuclear Applications

- Radiation effect issues which need further studies
 - Irradiated strength at beyond 1000C and 15 dpa
 - Irradiation creep
 - Void swelling at >1000°C
 - Environmental effects under irradiation
 - Fundamental radiation effect mechanisms / physical processes
- Substantial effort will be needed for code qualification for critical applications in nuclear systems
 - Design codification, test standards, database development



The Overarching Goals for Fusion Power Systems Narrow the Choices and Place Significant Demands for Performance of Structural Materials



Fe-9Cr steels: builds upon 9Cr-1Mo industrial experience and materials database (9-12 Cr ODS steels are a higher temperature future option) V-4Cr-4Ti: Higher temperature capability, targeted for Li self-cooled blanket designs SiC/SiC: High risk, high performance option (early in its development path)

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Low Activation Ferritic Steels for First Wall/Blanket Structures

Advantages

- Well-developed technology for nuclear and other advanced technology applications
- Fusion materials program has developed low activation versions with equivalent or superior properties
- Resistant to radiation-induced swelling and helium embrittlement
- Compatibility with aqueous, gaseous, and liquid metal coolants permits range of design options

Issues

- Upper operating temperature limited to ~ 550°C by loss of creep strength
- Potential for radiation-induced embrittlement at temperatures <400°C
- Possible design difficulties due to ferromagnetic properties

Expand Low Temperature Operating Window

- Pursue collaborative international fission reactor irradiation program (IEA activities)
 - Investigate micro- mechanics of fracture and radiation-induced reductions in fracture toughness
 - Understand the role of helium on fracture and crack propagation
 - Develop Master Curve approach to examine deformation modes and fracture resistance

Expand High Temperature Operating Window

- Explore potential of TiC dispersion strengthened and nanocomposited ferritic (NCF) materials to expand upper operating temperature to ~800°C
 - Develop radiation-stable, high toughness microstructures



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Ferritic/martensitic Steels with Reduced Radioactivity and Superior Properties Compared to Commercial Steels have been Developed by Fusion



Fusion-developed steels also have superior tensile strength, irradiated fracture toughness, and thermal conductivity



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Nanoclusters Formed at 850°C Extrusion







- •Energy Filtered TEM of OE14YWT
- •Resolution:

 $(1.4nm/p) \ge 5p = 7.0 nm dia. upper range$ $(1.4nm/p) \ge 2p = 2.8 nm dia. lower range$

(1.4nm/p) x 40p = 56.0 nm dia. large Ti-rich particle

The formation of nanoclusters becomes possible in the presence of vacancies



 Excessive vacancies are assumed to exist during mechanical alloying → high solubility of O-vacancy pair Nanoclusters are modeled as coherent with underlying bcc lattice Without Y → TiO₂ oxide phase • Too much $Y \rightarrow Y_2 Ti_2 O_7$ oxide phase Y is pivotal; however, the amount of Y needs to be controlled (Ti >> Y) to avoid the precipitation of Y₂Ti₂O₇ oxide phase.

C. L. Fu, M. Krcmar, G. S. Painter, X.-Q. Chen, Phys. Rev. Lett. 99, 225502 (2007)





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Characterization Tools

• Evaluate nano-features (NFs) & precipitation aging thermokinetics with a wide range of techniques



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G.R. Odette, UCSB

F82H - 500°C @9 dpa/380 appmHe

• Bimodal distribution of small bubbles and polyhedral voids



NFA MA957 500°C at 9 dpa/380 appm He



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_7_100kx_dislocations,_bubbles,_-1024_nm_uf

- > 90% over-under focus bubble coincidence
- > 85% bubble NF coincidence
- Some NF & bubbles on dislocations



G.R. Odette, UCSB

Summary - 500°C 9dpa/380 appm He

Alloy	<d> (nm)</d>	N $(10^{23}/m^{-3})$
Eurofer	4.3	0.15
F82H	2.8	0.53
MA957	1.3	4.3
J12YWT	1.2	4.4
1.122		V





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Vanadium Alloys are Most Attractive for Li Cooled/Breeder Blanket Systems

Performance Potential

- High Wall Load/Power Density
- High Operating Temperature and Thermodynamic Efficiency
- Low Activation/Potential Recycle
- Research Emphasis
 - Development of V/Li MHD Insulator Systems
 - Mechanical properties (including thermal creep)
 - Effects of Irradiation on Fracture Properties and Irradiation Creep
 - Kinetics of Interstitial Impurity
 Pick-up and Effects on Properties
 - Alloy Development (create high density of second phase particles)
 - Effects of He on irradiated microstructure

• Feasibility Issues

- Insulator Coatings to Mitigate MHD Effects in Li/V System
- Establish operating temperature window
 - Effects of He and displacement damage on properties
 - High temperature creep behavior
- Impurity Interactions from Environment, e.g. Oxidation



Thermal Creep of V-4% Cr-4% Ti at 600-800°C

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Comparison of the Design Window for Nb1Zr and V4Cr4Ti



 V4Cr4Ti offers ~factor of two higher stress capability than Nb1Zr



Silicon Carbide Composite Development

Silicon carbide composite is the least-developed of the 3 main structural materials being studied in the Fusion Materials Program, but it has the **greatest potential** *Very Low Radioactivation - Very High Temperature Use*



We Now Have First Radiation-Resistant SiC Composite



Future work: advanced matrix infiltration R&D; joining; hermetic coatings; SiC/graphite composites, etc.

SiC/SiC Composites Development

Reference Chemical Vapor Infiltrated (CVI) Composites for Irradiation Studies

- Hi-Nicalon[™] Type-S or Tyranno[™]-SA3 / PyC(50–150nm^t) / CVI-SiC composites have been selected as the reference materials
- Materials are under fabrication in US/Japan collaboration
- Extensive engineering data generation for irradiated properties (including statistical strength) is planned (prior studies utilized simple qualitative screening tests)
 Bend strengths of irradiated "3rd generation"



Irradiation Effect Studies in SiC/SiC

Composite Properties

- Various mechanical and thermo-physical properties of irradiated SiC/SiC composites are being evaluated.
- Swelling, thermal conductivity, elastic modulus, tensile strength, shear strength, etc.





Shape Flexibility of "NITE" SiC/SiC Composites

Based on stoichiometric SiC fibers, nanoscale powder-based slurry infiltration, engineered fiber-matrix interphase



A. Kohyama, ANS TOFE (Madison, WI, 2004)

Current Status of SiC/SiC Composites





Modified chemistry and thermomechanical treatment procedure for new 9Cr ferritic/martensitic steels produces high strength





 Good toughness and high temperature strength are also produced in dispersion-strengthed Fe-9.5Cr-3Co-1Ni-0.6Mo-0.3Ti-0.07C steel due to high number density of nano-size TiC precipitates

Klueh and Buck, J. Nucl. Mater. 283-287 (2000)



R.L. Klueh et al., Scripta Mater. (2005)

Low temperature radiation hardening causes fracture toughness embrittlement in BCC metals



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Modern Materials Science Applied to "Old" Alloys Yields Dramatic Improvements in Performance and New Applications

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Alloy composition is changed to produce a microstructure of fine, stable precipitates



energy industry applications

High Performance Diesel Engine

New cast austenitic steels for automotive and heavy vehicle exhaust components

Similar Scientific Microstructural Design Produces:

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Size and Number of Precipitates in 9Cr Steels Depend on TMT Process

Alloy	MX Precipitates	
	Average Size (nm)	Number Density (m ⁻³)
Modified 9Cr-1Mo—N&T	32	7.9 X 10 ¹⁸
Modified 9Cr-1Mo—TMT1	7.2	8.9 X 10 ²¹
Modified 9Cr-1Mo—TMT2	7.3	2.1 X 10 ²¹
Modified 9Cr-1Mo—TMT3	8.0	1.9 X 10 ²¹
Fe-9Cr-1MoNiVNbN—1	4.0	1 X 10 ²²
Fe-9Cr-1MoNiVNbN—2	3.3	7.2 X 10 ²²

 High number density of nano-size particles obtained (up to 10,000x density in Modified 9Cr-1Mo without new TMT)

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R.L. Klueh, ORNL



Austenitic Stainless Steel exhibits low uniform elongation after irradiation at temperatures up to ~400°C

Reduction in uniform elongation requires higher doses than in simple metals (e.g. Cu, Ni)



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Plastic Instability Stress (σ_{PI}) of austenitic stainless steel irradiated near 70°C



T.S. Byun & K. Farrell, Acta Mater. 52 (2004) 1597

Calculated irradiated Ashby deformation map for Type 316 stainless steel at low strain rates Damage rate = 10⁻⁶ dpa/s

Normalized Shear Stress, π/μ (20°C) **10**⁻¹ 10⁻¹⁰ s⁻¹, L=50 μm Theoretical shear stress limit **10⁻²** MPa 2400 **Dislocation glide** Twinning Uniaxial Tensile Stress, **Dislocation creep 10**⁻³ 240 Irrad. creep N-H **10**⁻⁴ Coble 24 creep creep **Elastic regime** $(d\epsilon/dt < 10^{-10} \text{ s}^{-1})$ **10**⁻⁵ 2.4 20°C 300°C 650°C **10**⁻⁶ 0.24 0.2 0.4 0.6 **8.0** Normalized Temperature, T/T_M

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Overview of dislocation channeling

- Dislocation channeling is a viable mechanism to locally work soften the matrix
 - Shearable obstacles
- Channeling involves localized flow and therefore inhibits dislocation multiplication
 - Limited interaction between dislocation sources

However,

- It is not generally established that the catastophic reduction in tensile elongation is directly due to dislocation channeling
 - High tensile elongations and significant work hardening rates can
- 43 Managed by Charlier in irradiated metals that exhibit dislocation channeling CAR for the U.S. Department of Energy

Dislocation channel interactions in Fe deformed following neutron irradiation at 70°C to 0.8 dpa

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Cleared slip channel

g.b.

Need well-engineered materials to mitigate neutron radiation effects

• Type 1 interaction (Frank loop formation) at room temperature

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 Type 2 interaction at room temperature (superjog creation with no SFT remnant)

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Types 1(a) & 2(b) interactions also occur at 100 K (no vacancy migration)

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- Type 3 interactions at room temperature (SFT apex remains); not observed at 100 K
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Effect of temperature on edge dislocation interaction with 136 vacancy SFT in Cu

Defect cluster annihilation is enhanced at higher temperatures and slower strain rates (strain rate effect not shown)

- agrees with experimental results

Other parameters such as effect of obstacle size are also under investigation

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Y.N. Osetsky

Interaction of a screw dislocation with 78-vacancy SFT and 91-intersitital cluster in Cu thin foil

Cooperative effects may be important for annihilation of sessile defect clusters by gliding dislocations during deformation

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Yu.N. Osetsky

Isotropic plasticity model for irradiated metals

Isotropic polycrystal plasticity incorporates coarse grained scaling laws governing dislocation density evolution and interactions determined for single crystals
Dislocation - (radiation damage) defect interactions are based on MD simulations
Resulting models can be further modified to include the effects of dispersed particles, solute atoms, and other known resistance mechanisms

Arsenlis, Wirth and Rhee, *Phil. Mag.* 84, 3617 (2004)

Engineering and true stress-strain tensile curves for stainless steel before and after spallation irradiation at ~100°C

Radiation Effect in True Stress-True Strain Curve (FCC)

The curves of irradiated specimens are empirically shifted by strains of 0.14, 0.18, 0.23, 0.28, and 0.385, respectively, to superimpose on the curve of unirradiated material. Irradiation-induced increases in yield stress were 305, 358, 421, 485, and 587 MPa, respectively.

A common plastic instability stress (max stress prior to necking) is observed for all conditions

Further work is needed to determine importance of dislocation channeling in observed exhaustion of initial strain hardening capacity

53 Managed by UT-Battelle for the U.S. Department of Energy T.S. Byun & K. Farrell, Acta Mater. 52 (2004) 1597

 $\sigma_{\text{ML}}\text{=}\text{true}$ stress at maximum load

- Plastic Instability Stress (σ_{PI}) = the true stress version of Ultimate Tensile Stress
- Plastic Instability Stress is independent of dose when yield stress $< \sigma_{PI}$.
- Yield stress can be $> \sigma_{PI}$, which is defined only when uniform deformation exists.
- σ_{PI} is considered to be a material constant, independent of initial cold-work or radiationinduced defect clusters
 T S Pyup & K Earroll
 - 54 Managed by UT-Battelle for the U.S. Department of Energy

T.S. Byun & K. Farrell, Acta Mater. 52 (2004) 1597

TEM in-situ deformation studies can be used to provide insight on fundamental fracture processes

Atomic resolution imaging of ductile crack propagation (plane stress)

 Macroscopic Mode I fracture is composed of coordinated Mode III shear displacements at the crack tip

55 Managed by UT-Battelle for the U.S. Department of Energy 2 nm Y. Matsukawa, ORNL

Fusion Materials Relies Heavily on Modeling due to Inaccessibility of Fusion Operating Regime

- Extrapolation from currently available parameter space to fusion regime is much larger for fusion materials than for plasma physics program.
- Lack of intense neutron source emphasizes the need for coordinated scientific effort combining experiment, modeling & theory to develop fundamental understanding of radiation and other sources of in-service life limiting damage.

Why is He/dpa ratio an important parameter for fusion materials R&D?

He generation can alter the microstructural evolution path of irradiated materials

- Cavity formation
- Precipitate and dislocation loop formation

He bubbles on grain boundaries can cause severe embrittlement at high temperatures

Swelling in stainless steel is maximized at fusion-relevant He/dpa values

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Cavity formation in JLF-1 by dual-beam irradiation - 6.4 MeV Fe³⁺ + Energy-Degraded 1.0 MeV He⁺, 20 dpa at 743 K -

Y. Katoh et al., J. Nucl. Mater. 323 (2003) 251

Swelling behavior of austentic and ferritic stainless steels

Y. Katoh et al., J. Nucl. Mater. 323 (2003) 251

Swelling behavior of RAFMs and austenitics near peak-swelling temperatures are similar in the presence of helium, except for incubation dose.

Impact of He-Rich Environment on Neutron Irradiated Materials

- A unique aspect of the DT fusion environment is substantial production of gaseous transmutants such as He and H.
- Accumulation of He can have major consequences for the integrity of fusion structures such as:
 - Loss of high-temperature creep strength.
 - Increased swelling and irradiation creep at intermediate temperatures.
 - Potential for loss of ductility and fracture toughness at low temperatures.
- Trapping at a high-density of tailored interfaces is a key strategy for management of He.

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Swelling in stainless steel is maximized at fusion-relevant He/dpa values.

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0

80

0

Science-Based High-Temperature Design Methodology

- Current high-temperature design methods are largely empirically based and material application specific.
- Cyclic plastic loading is far more damaging than monotonic loading.
- New models of high-temperature deformation and fracture are needed:
 - -Creep-fatigue interaction.
 - -Elastic-plastic, time-dependent fracture mechanics.
 - -Materials with low ductility, pronounced anisotropy, composites and multilayers.

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Breaking the High Strength-Low Toughness/Ductility Paradigm

- A general feature of engineering materials is increased strength tends to be offset by losses of toughness (resistance to crack growth) and ductility.
- Strength increases may result from alloying, material processing, or radiation damage.
- Loss of toughness and ductility is a loss of margin against structural failure.
- Simultaneous achievement of highstrength and high toughness/ductility would provide enormous benefits for fusion, but also many other areas (e.g., transportation, magnets, robotics, etc.).

Fundamentals of Material-Coolant Chemical Compatibility in the Fusion Environment

- The traditional approach to study corrosion has been largely empirical, based on static coupon testing and flowing coolant in a temperature gradient.
- Empirical correlations do not capture the fundamental physics involved and do not provide predictive capability outside the range of the experimental measurements.
- Opportunities:
 - Controlled experiments combined with physical models based on advanced computational thermodynamics and kinetics codes.
 - Integrated flow experiments enhanced by use of sophisticated *in situ* diagnostic and sensor technologies.

M. Zmitko / US-EU Material and Breeding Blanket Experts Meeting (2005) J. Konys et al./ ICFRM-12 (2005)

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Molecular Dynamics Simulations of the Effect of Temperature on Obstacle Strength

Conclusions

- Ongoing radiation materials science research programs span from fundamental studies to targeted alloy development
- Common research themes include:
 - Investigation of fundamental phenomena responsible for materials property changes (degradation) due to irradiation
 - Development of radiation resistant high-performance structural material systems
- Increased coordination is being emphasized between computational modeling and experiments in order to accelerate the rate of scientific discovery

