Radiation Tolerant Materials by Design for Inertial Fusion Energy

Mary Alice Cusentino, LaRico Juan Treadwell, Luke Shulenburger, and Khalid Hattar

Abstract: The harsh complex, and cyclic radiation environment associated with future Inertial Fusion Energy (IFE) concepts requires the development of materials that can withstand cyclic extremes including ~10 dpa/year, >1keV over ns bursts, and the evolution of hydrogen and helium isotopes. Novel materials will be required for the chamber walls of future IFE devices, but the development and incorporation of new materials has traditionally been a slow process. However, combining AI/ML and materials modeling to drive experimental design with material synthesis and high throughput characterization techniques can greatly improve this process and shorten the time to develop materials for extreme environments. If successful, this process could be translated to study a variety of different materials and application spaces.

Background: Designing materials for inner chamber walls is difficult due to the extreme conditions within and IFE device including high levels of neutron damage (~10 dpa/year), high temperatures (~ 1000 °C), and high particle fluxes of additional species like hydrogen and helium isotopes [1, 2]. Current candidate materials include ODS, RAFMS, and FLiBe [1, 2]. The combination and interplay of these different factors result in material degradation of the chamber components that must be understood to develop a viable IFE device. The processes that drive radiation induced material deformation occur across a wide range of time and length scale, as indicated in Figure 1 (A). Atomistic scale processes drive radiation damage, including the implantation of plasma species in the material and formation of point defects via neutron irradiation, as shown in Figure 1 (B). The accumulation and diffusion of radiation induced defects in the material can result in macroscopic degradation of the material over longer time and length scales. Bubble formation, blistering, mixed materials layers, surface deformation, etc. can result in materials exposed to fusion plasmas. This requires both modeling and experimental capabilities than need to span over orders of magnitude in time and length scales. Adding to the complexity are the synergistic effects from the presence of multiple plasma species as well as neutron produced radiation damage which can result in more complicated relationships between radiation damage and exposure conditions. In addition, interaction between the chamber wall materials and plasma edge requires complex, coupled physics codes. Therefore, the design of radiation tolerant IFE chamber wall materials requires information from the atomistic to meso to experimental scale to fully understand both how the material is degrading as well as the processes and mechanisms that are driving this observed damage.

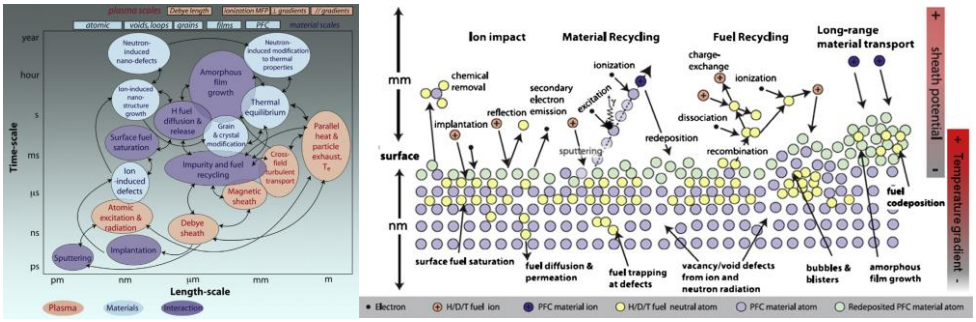

Figure 1. A) Graphical representation of the multiple time and length-scales involved in the inherently coupled processes and phenomena which dictate plasma materials interactions in the boundary plasma region of magnetic fusion devices. Plasma processing (light red), near-surface and bulk materials (light blue), and the important plasma–materials interactions (light purple) . B) Schematic illustration of the synergistic plasma surface interaction processes that dictate material evolution and performance in the magnetic fusion plasma. Environment [3].

Although the environment expected for IFE concepts is complex and rapidly changing, we are hopeful recent advancements in materials' synthesis, selection, and production for radiation tolerance provide a path forward. By controlling the structural and compositional nature of materials with clear intent to develop radiation tolerance, a range of new and exciting materials have emerged in the last decade that might be of interest for IFE applications. Just the control of structure with nanometer precision has been shown in almost all cases to improve the radiation tolerance of materials, as was recently reviewed by X. Zhang et al. and seen in Figure 2 [4]. In a similar manner, the composition of materials can be altered to increase the chemical complexity of materials, this has resulted in the production of single-phase concentrated solid solution alloy(CSSA) [5] and high entropy alloys(HEA) [6]. If one pushes the physical boundaries on both structural and compositional complex materials, while staying in the metallic system, the field of bulk metallic glasses emerge [7]. These are systems where the diversity of local chemistry and subsequent bonding provides only short-range structural order. If the medium range order of a crystalline system is found to be of benefit, but local disordered sprinkled in also has benefit, the new field of amorphous interface films (AIF) becomes an area to explore in metallic systems [8]. The properties can be explored further by incorporating in ceramic particles into the system. An approach often called oxide dispersion strengthening(ODS). All of these approaches (nanostructuring, CSSA, HEA, AIF, and ODS) have all recently been shown experimentally by our team to unique properties (radiation tolerance, irradiation induced creep, strength) [4-9]. We propose that some combination of these structural and compositional tailoring might greatly improve the radiation tolerance of materials necessary for IFE concepts.

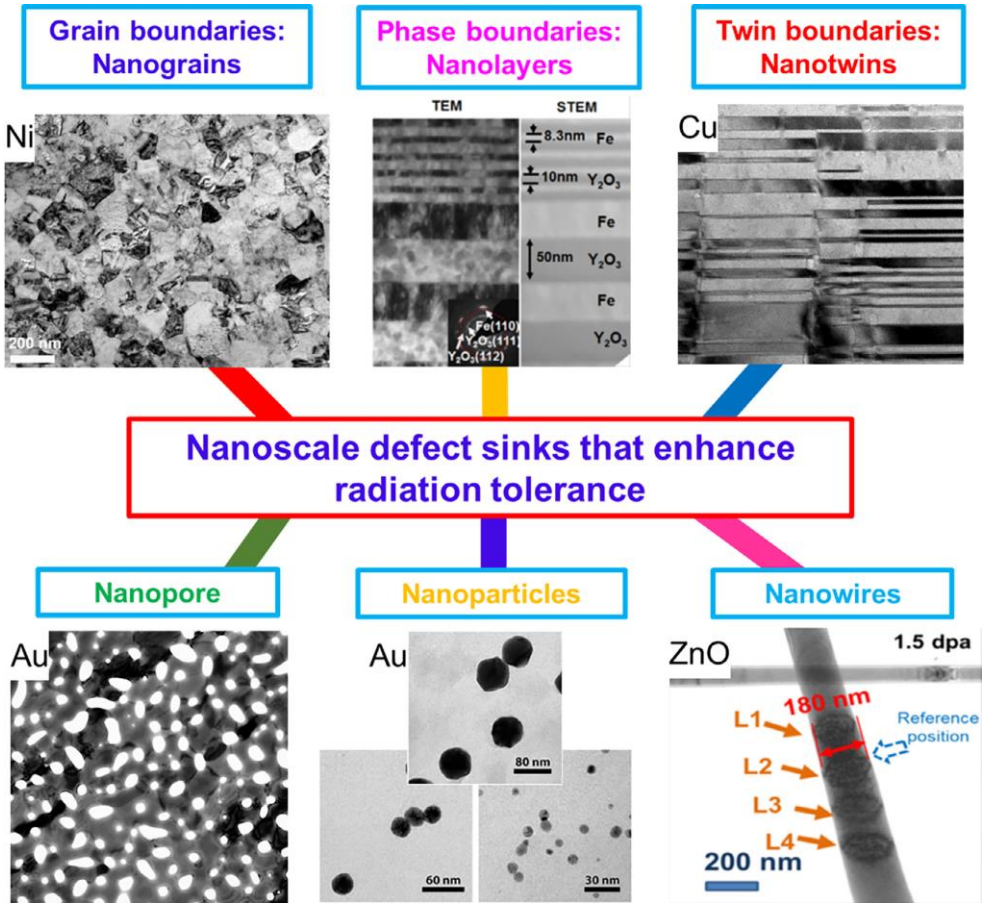

Figure 2. The application of various types of defect sinks to alleviate radiation damage. Defect sinks include dislocations, grain boundaries, twin boundaries, layer interfaces, nanopores, nanoparticle, nanowires and amorphous materials. This review will cover radiation damage in various nanostructured materials, including nanocrystalline materials, nanotwinned metals, nanolayers, nanoporous metals, nanoparticles and nanowires [4].

Modeling of radiation damage in materials is inherently a multi-scale problem and requires a suite of simulations techniques to bridge between these vastly different time and length scales. At the atomistic scale, tools like density functional theory (DFT) and molecular dynamics (MD) are widely used to study radiation effects in materials. DFT can provide highly accurate information about material properties like defect energetics. Similarly, MD is well suited for modeling plasma species implantation or neutron bombardment due to the small-scale nature of these initial radiation events. MD can provide insight into mechanisms and physical processes that drive radiation damage in materials. Large-scale MD of helium implantation in tungsten on leadership class computing [10] has been recently employed in order to simulate more experimentally relevant conditions for MFE devices and elucidate damage mechanisms. In addition, the use of machine learning to develop more highly accurate interatomic potentials [11] has also been utilized for studying plasma-material interactions for MFE [12] that would be otherwise unachievable with traditional interatomic potentials. The development of these new tools allows for more accurate and realistic simulations of plasma material interactions. In turn, these observations from MD can be passed up to meso-scale codes, such as reaction diffusion, kinetic monte carlo, or reduced order models that can then extrapolate these results to experimentally relevant scales. This has been achieved for modeling helium effects in tungsten for MFE [13] and could be similarly adapted to IFE materials.

Predicting and producing these materials are two important steps, but cannot stand alone. For any new material to be incorporated into such a harsh application and high level of validation is needed. Recent advancements in rapid small-scale screening of materials through small scale laser based thermal measurements (TGS, TDTR, etc.), small scale mechanical testing (nanoindentation, CSM, and micropillar compression), as well many other in-situ techniques [14] provides the capability to understand the new materials response to such complex environments. None are more powerful than in-situ TEM [15, 16] to direct couple the active mechanism within the material to the materials response in the controlled environment, as such it provides nanoscale understanding to provide feedback into models

Hypothesis: ***Radiation tolerant materials that can handle the multiple extreme environments present in the IFE environment are physically possible and we believe can be developed/discovered by utilizing the state-of-the-art in materials design for extreme environments.***

Approach: Since the dawn of the stone age, humans have been excellent in the utilization of best materials known at the time, even if it required extremely challenging mining, trading route, or processing methodologies. This paradigm for materials selection has recently been challenged with concepts like the Materials Genome initiative [17, 18]. This white paper will utilize the greatest in materials by design concepts to develop radiation tolerant materials for current and future IFE concepts. To succeed at such a task, a direct and tight coupling between models that can predict the performance margins necessary for the harsh application and rapid production, screening, and validation are needed. The following approach details a potential path utilizing the state-of-the-art in both modeling and experiment that might be able to achieve that task for IFEs.

Task 1: Design of radiation tolerant materials via both first principle and AI/ML

Atomistic modeling tools like MD and DFT can be used to probe materials for radiation tolerant features. While MD and DFT are limited in achievable time and length scales, these tools have the benefit of rapidly testing materials for a variety of properties related to radiation tolerance as well as the ability to finely control the exposure conditions and material composition. The emergence of machine-learning methods for constructing energy models that combine the high accuracy of DFT with the linear scaling of

MD has greatly extended the predictive power of *in silico* testing. Recent work [19] of radiation damage simulations in high entropy alloys (HEAs) indicated that both the chemical complexity of the HEA as well as the inclusion of structural features resulted in the most radiation tolerant material. Similarly to what was done in [19], atomistic modeling could be used to probe vast regions of compositional and microstructural space. Understanding the atomic scale behavior of these materials under irradiation and the mechanisms behind radiation response, can be used to design better materials for IFE devices. Combining this with data science and machine learning, it would be possible to generate a database of quantities of interest for radiation tolerance (i.e. point defect production, etc.) in materials and develop ML models for predicting the behavior of these materials. These models can then in turn be used to drive which compositions, structural features, etc. should be chosen for further testing in experiments.

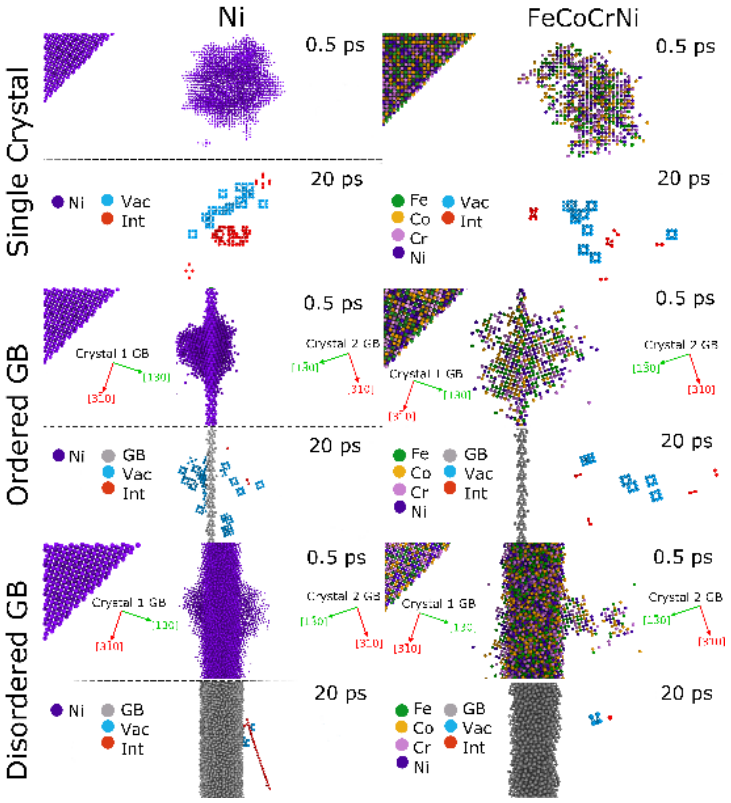

Figure 3. Representative snapshots of a 10 keV collision cascade in a single crystal and near ordered/disordered grain boundaries at 300 K in both Ni (left column) and in FeCoCrNi (right column). The atoms and defects are colored by atom type. Interstitials and vacancies are shown in red and blue, respectively. The highlighted vacancy atoms are the lattice atoms surrounding the vacancy. Since there are twelve nearest neighbors in an FCC lattice, each cube of twelve atoms accounts for one vacancy. The top snapshots (above the dashed line and referred to as peak cascade) for each case correspond to the cascade structure during the thermal spike (at 0.5 ps) where the cascade reaches approximately its maximum size. The bottom snapshots, below the dashed line and referred to as post cascade, correspond to the cascade structure after the recovery process when the cascade cools down (after 20 ps). The reveal in the top left corner of each panel displays the underlying bulk crystal structure [19].

Task 2: Rapid synthesis of combinatorial material sets

Given recent and rapid advancements in additive manufacturing (AM) techniques of both complex metal alloys and ceramic composites, it is now possible to tailor the distribution of composition, texture, and phases. Although still in its infancy, this approach may impact significantly the design and development of single phase, graded, and other MPEA and associated systems that may have substantial impact on future of materials for IFEs. For example, the immense variability in processing and input parameters permits systematic design of these MPEAs, including the inclusion of dispersed high-temperature stable carbide particles. These carbides will increase the stability introduced during the tailoring of MPEA composition and microstructure.

Task 3: High throughput initial material screening and model feedback

We believe it is possible to predict, evaluate, and rapidly iterate on the microstructure to optimize such composite response to the extreme thermal, mechanical, and radiation environments present in PFCs. Such an approach would couple the atomistic scale molecular dynamic (MD) simulation and *in situ* electron microscopy. Systematic and well controlled experimental conditions in the TEM or SEM can be used to deconvolute the environmental conditions and initial microstructure roles in active degradation mechanisms of the W-composite. A range of *in situ* thermal, implantation, and irradiation TEM and SEM techniques can be used to observe directly the active mechanisms with nanometer resolution utilizing the unique capabilities present in the *in situ* Ion Irradiation SEM (I^3SEM) and *in situ* Ion Irradiation TEM (I^3TEM) facilities [15]. Recent advancements in Sandia's Large-scale Atomic/Molecular Massively Parallel Simulator (LAMMPS) MD code permit (a) direct input of initial structures from automated crystallographic orientation maps (ACOM) collected during TEM characterization and (b) the interpretation of MD outputs in a form that can be directly compared to electron diffraction patterns acquired during TEM analysis [20]. The rapid screening and physics-based insight provided by the *in-situ* ion irradiation and MD simulation will drive down selection decisions for the alloy and composite choices to optimize the thermal, mechanical, and radiation stability of the W-based MPEA and composites.

We believe that the ability (1) to rapidly produce complex composites through AM will enable rapid screening of the initial microstructure, (2) to elucidate the active mechanism through *in situ* electron microcopy in tailored extreme environments, and (3) to determine the underlying physics through coupled MD simulations is necessary for the rapid development of new materials needed for such harsh environments.

Expected Output: If successful, this concept would result in a rapid methodology to design, produce, and screen advanced material compositions and microstructures for both current and future IFE concepts. This will include an experimental and first principle validated computational platform that utilize AI/ML to predict new radiation tolerant materials. This would also result in a rapid way to produce and provide initial testing of various material concepts. This initial screening will minimize the large scale and long-time frame testing, the subsequent nuclear waste produced, and most importantly the time required to introduce a new trusted material into current or future IFE concepts.

Acknowledgements:

This work was performed, in part, at the Center for Integrated Nanotechnologies, an Office of Science User Facility operated for the U.S. Department of Energy (DOE) Office of Science. Sandia National Laboratories is a multimission laboratory managed and operated by National Technology & Engineering Solutions of Sandia, LLC, a wholly owned subsidiary of Honeywell International, Inc., for the U.S. DOE's National Nuclear Security Administration under contract DE-NA-0003525. The views expressed in the article do not necessarily represent the views of the U.S. DOE or the United States Government.

References:

1. Meier, W., et al., *Fusion technology aspects of laser inertial fusion energy (LIFE).* Fusion Engineering and Design, 2014. **89**(9-10): p. 2489-2492.
2. Council, N.R., *An assessment of the prospects for inertial fusion energy*. 2013: National Academies Press.
3. Wirth, B.D., et al., *Challenges and opportunities of modeling plasma–surface interactions in tungsten using high-performance computing.* Journal of Nuclear Materials, 2015. **463**: p. 30-38.

4. Zhang, X., et al., *Radiation damage in nanostructured materials.* Progress in Materials Science, 2018. **96**: p. 217-321.
5. Wang, X., et al., *Defect evolution in Ni and NiCoCr by in situ 2.8 MeV Au irradiation.* Journal of Nuclear Materials, 2019. **523**: p. 502-509.
6. Jawaharram, G.S., et al., *Irradiation induced creep in nanocrystalline high entropy alloys.* Acta Materialia, 2020. **182**: p. 68-76.
7. Schuler, J.D., et al., *In situ high-cycle fatigue reveals importance of grain boundary structure in nanocrystalline Cu-Zr.* JOM, 2019. **71**(4): p. 1221-1232.
8. Schuler, J.D., et al., *Amorphous intergranular films mitigate radiation damage in nanocrystalline Cu-Zr.* Acta Materialia, 2020. **186**: p. 341-354.
9. Kiani, M.T., et al., *Ductile metallic glass nanoparticles via colloidal synthesis.* Nano Letters, 2020. **20**(9): p. 6481-6487.
10. Hammond, K.D., et al., *Helium flux effects on bubble growth and surface morphology in plasma-facing tungsten from large-scale molecular dynamics simulations.* Nuclear Fusion, 2019. **59**(6): p. 066035.
11. Zuo, Y., et al., *Performance and cost assessment of machine learning interatomic potentials.* The Journal of Physical Chemistry A, 2020. **124**(4): p. 731-745.
12. Wood, M.A., et al., *Data-driven material models for atomistic simulation.* Physical Review B, 2019. **99**(18): p. 184305.
13. Blondel, S., et al., *Continuum-scale modeling of helium bubble bursting under plasma-exposed tungsten surfaces.* Nuclear Fusion, 2018. **58**(12): p. 126034.
14. Lang, E., et al., *The In Situ Ion Irradiation Toolbox: Time-Resolved Structure and Property Measurements.* JOM, 2021: p. 1-17.
15. Hattar, K., D.C. Bufford, and D.L. Buller, *Concurrent in situ ion irradiation transmission electron microscope.* Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms, 2014. **338**: p. 56-65.
16. Hinks, J., *A review of transmission electron microscopes with in situ ion irradiation.* Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms, 2009. **267**(23-24): p. 3652-3662.
17. de Pablo, J.J., et al., *New frontiers for the materials genome initiative.* npj Computational Materials, 2019. **5**(1): p. 1-23.
18. White, A., *The materials genome initiative: One year on.* Mrs Bulletin, 2012. **37**(8): p. 715-716.
19. Cusentino, M., M. Wood, and R. Dingreville, *Compositional and structural origins of radiation damage mitigation in high-entropy alloys.* Journal of Applied Physics, 2020. **128**(12): p. 125904.
20. Coleman, S.P., M.M. Sichani, and D.E. Spearot, *A computational algorithm to produce virtual X-ray and electron diffraction patterns from atomistic simulations.* Jom, 2014. **66**(3): p. 408-416.