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FY17 CAES LDRD Annual Report

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Idaho National Laboratory
Idaho Falls, Idaho 83415

http://www.inl.gov

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16-187 - Micro-Scale Technique to Evaluate Grain Boundary Cohesion of Irradiated Alloys

Chao Jiang,[1] Wen Jiang,[1] Brian Jaques,[2] Indrajit Charit,[3] and Ray Fertig[4]
1 INL; 2 Boise State University; 3 University of Idaho; 4 University of Wyoming

Metallic alloys are widely used, or planned for use, as structural and cladding materials in current and future reactors. Under irradiation, grain-boundary (GB) cohesion strength decreases due to interaction with defects and impurities, leading to intergranular fracture and embrittlement of alloys. The objective of this project is to develop a technique for quantifying GB cohesion and its impact on fracture behavior in irradiated alloys by utilizing transmission electron microscopic (TEM) in situ cantilever testing in concert with multi-scale modeling. The TEM in situ cantilever testing is a novel approach for studying the real-time mechanical response of materials. It will be used in this work for studying intergranular fracture behavior in several irradiated iron-based ferritic alloys and for providing key information to link atomistic-level events with mesoscale/macroscopic mechanical properties. The Multi-Physics Object-Oriented Simulation Environment (MOOSE)-based cohesive zone model (CZM) and extended finite-element method (XFEM) for intergranular fracture of irradiated ferritic alloys will be developed in this work by utilizing atomistic results as inputs and experimental results for validation.

Summary
Under neutron or ion irradiations, many point defects and their clusters will be directly formed during collision cascades. Diffusion of interstitials or vacancies towards sinks such as GBs can lead to redistribution of solute concentrations, so-called radiation-induced segregation effect. In ferritic Fe-Cr alloys, both radiation-induced depletion and enrichment of Cr at GBs have been observed. Due to super-saturation of point defects created under irradiation, radiation-enhanced diffusion can also greatly accelerate the segregation of impurity elements to GB. According to present density functional theory (DFT) calculations (see Table 1), sulfur (S) and phosphorus (P) in steels and helium (He) produced by nuclear transmutation reactions all have a strong tendency to segregate to GB. An important consequence of the local enrichment of impurities at GBs is that they can drastically weaken GB cohesion strength and induce intergranular fracture and embrittlement. Present DFT calculations reveal a dramatic reduction of GB cohesion energy due to the segregation of S, P and He (see Table 1). In contrast, Cr segregation does not seem to have a detrimental effect on GB cohesion in bcc Fe.

Table 1. DFT calculated segregation energies (E_{seg}) of impurities to a Σ5(310)[001] symmetric tilt GB in bcc Fe. The change of the GB cohesion strength (dE_{coh}) due to impurity segregation is also shown.

Impurity	S	P	He	Cu	Si	Al	Mo	Ni	Cr
E_{seg} (eV)	-1.25	-0.87	-1.51	-0.56	-0.44	-0.38	-0.40	-0.40	-0.25
dE_{coh} (eV)	-2.25	-1.30	-3.21	-0.33	-0.48	-0.22	+0.90	+0.04	+0.47

In FY17, molecular dynamics (MD) simulations with empirical embedded atom method (EAM) potential have also been performed using LAMMPS code to predict the effects of

He loading on the GB cohesion strength in bcc metal molybdenum (Mo). Here Mo-He is used as a surrogate system for Fe-He due to the lack of EAM potential for the latter. Nanocrystalline (nc) bcc Mo sample containing 32 grains and around a half million atoms has been generated using the Voronoi tessellation method. He gas atoms are then randomly loaded at GBs (yellow spheres in Figure 1). Due to the replacement of strong metallic bonding with repulsive He-Mo bonds, He segregation at GB significantly reduces the strength of nc bcc Mo by promoting intergranular fracture. During uniaxial loading simulations, complete failure of the nc Mo sample is observed when local He concentration at GB exceeds 6%.

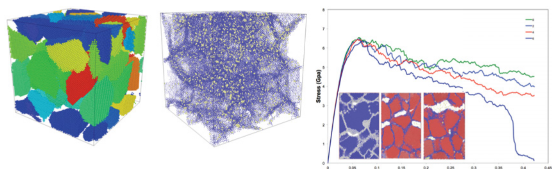

Figure 1. MD simulation of the effects of He on mechanical strength of nc bcc Mo.

On the continuum scale, implementation of CZM in the MOOSE Framework has been realized by utilizing ElemElem constraint in XFEM based on a phantom node method: the element containing a crack is deleted and replaced by two partial elements at the same location. Mode-I implementation of CZM was compared with a commercial finite element package, Abaqus for a benchmark data set. The traction-separation obtained from MOOSE and Abaqus are in very good agreement. Future work will be aimed towards combining crystal plasticity with CZM to simulate the response of polycrystalline BCC Fe when loaded.

During FY 2017, experimental efforts have been focused on developing capabilities for measuring GB cohesion strength using focused ion beam (FIB) for sample fabrication, electron back scatter diffraction (EBSD) for GB quantification, and in situ TEM mechanical testing. Multiple nanometer-scale geometries have been investigated, including; 4-point bend testing, cantilever testing, and tensile testing in order to investigate the mechanical response of induced stresses on grain boundaries. Each of the geometric designs allows for the isolation of single grain boundaries on the scale of a few micrometers and the samples can be fabricated within micrometers of the surface of the specimens. Accordingly, the experimental work allows for accurate representation of damage incurred during neutron irradiations using ion-induced damage (which has a small penetration depth). Using high energy ball milling followed by spark plasma sintering (SPS), Fe-9Cr alloy has also been synthesized. Iron and chromium powders were mixed in the right proportion in a glove box under argon environment and then ball milled for different milling times. For SPS, different sintering time, temperature and pressure have been used to achieve the best density. X-ray diffraction was carried out on the sintered samples and showed essentially peaks of bcc iron indicating that the material was ferritic. EBSD performed on the Fe-9Cr sample showed the presence of grains with size of 1.8±0.5 μm and preponderance of high angle grain boundaries. TEM characterization showed grain size to be 1-4 μm with approximately 100 nm intra- and intergranular precipitates, which appeared to be chromium oxide. The Fe-9Cr samples have been sent to Texas A&M Ion Beam Laboratory for proton irradiation with doses up to 5 dpa at 300°C and 475°C. Irradiated alloy samples will be used to study the radiation-induced segregation behavior and its effect on grain boundary cohesive strength.

Benefits to DOE

The radiation-induced degradation of mechanical material properties poses serious limitations to nuclear energy applications. The capabilities of TEM in situ mechanical testing and MOOSE-based fracture models developed for this work will help elucidate and predict the performance of materials in reactors. In turn, it would enable safer and more economical nuclear energy in the future. Results from this project will contribute to the DOE's leading role on research of materials behavior and performance in radiation environments.

Presentations

A. Kundu and I. Charit, A study on high energy ball milling and spark plasma sintering of Fe-9Cr model alloys. Materials Science & Technology (MS&T) 2017, Pittsburgh, PA, Oct 2017.

J. Watkins, B.J. Jaques, A. Bateman, Y. Wu, I. Charit, J.P. Wharry, K.H. Yano, W. Jiang, and C. Jiang. Irradiation effects on Fe-9%Cr grain boundary strength via in-situ TEM testing. The Minerals, Metals & Materials Society (TMS) 2018, Phoenix, AZ, March 2018.

A. Kundu, I. Charit, B. Jaques, and C. Jiang. A study on the high energy ball milling and spark plasma sintering of Fe-Cr based alloys. The Minerals, Metals & Materials Society (TMS) 2018, Phoenix, AZ, March 2018.

Publications

A. Kundu, S. Instasi, I. Charit, B. Jaques, and C. Jiang. A preliminary study on high energy ball milling of Fe-9Cr alloy. Submitted to Materials Letters.