CX Posting No.: DOE-ID-INL-20-081

SECTION A. Project Title: Probing in situ Multi-Property Evolution During Material Processing Using Laser Photoacoustics

SECTION B. Project Description and Purpose:

Project Description

Our understanding of the reliability, degradation, and evolution of materials placed in combined extremes of temperature, stress, radiation, corrosives, and fatigue is fundamental to safe and secure operation of complex energy systems, nuclear reactors in particular. The microstructural stability of as-installed components determines whether the desired properties (strength, thermal transport, resistivity) are retained throughout that component's operational lifetime. Innovation in the design and fabrication of materials used in these extreme environments is reliant on the tools used to characterize their structure and properties. In this project, we propose to leverage recent advances in in situ non-destructive material property evaluation using laser photoacoustics to observe degradation and evolution phenomenon resulting from processing in real time in a variety of advanced metals and alloys exposed to out-of-equilibrium conditions. These high-fidelity data will be used to optimize material compositions and fabrication pathways. We will also continue to develop these emerging state-of-the-art tools with the goal of increasing both the utility and information density returned in single measurements.

Research Plan

The two parallel research tasks under this proposal may be classed as technique development and alloy design. To conduct this work, a capability will first be established at INL to conduct Transient Grating Spectroscopy (TGS) experiments. Next, a unique modular TGS experimental test stand will be developed with the capability to couple to a variety of material processing systems. A cryogenic to high-temperature sample holder will be the first to be implemented, with others (strain, fatigue, etc.) to be developed as necessary. The capability, without any advanced technique development, will be used in the first research task as a tool for alloy design in high entropy alloys (HEAs) and nanocrystalline metals. Details of the study of these systems is provided below. In addition to an initial establishment of TGS capability, we will also develop new analytical and experimental techniques to extract more information than is currently possible using these methods as the second research task. Goals for this development are motivated directly by the desired applications and limitations in the current state of the art.

Throughout this work, detailed structural and defect characterization will be conducted using the facilities at the Center for Advanced Energy Studies (CAES) to provide information where necessary at TGS-identified critical evolutionary states. Irradiation effects will be studied in situ using the facilities available at Sandia National Laboratories through no-fee user access. Given the 24-month project lifetime, design and construction of new experimental facilities will be completed within the first six months. Iterative alloy testing will be carried out continually through the remaining 18 months. The experimental development will utilize laser sources and optics already in place at INL.

Task 1 – Alloy Design and Optimization

1.1 Concentrated solid solution and high entropy alloys (CSAs and HEAs)

These materials promise unique structural and functional properties including high strength, high thermal stability, wear resistance, superior corrosion and oxidation resistance, and excellent irradiation tolerance by combining three or more roughly equiatomic constituent elements. These features make them promising for use in extreme environments, particular in nuclear energy systems. In particular, 3d transition metal HEAs based on nickel (Ni) and iron (Fe) have received a great deal of investigation as possible candidates for structural materials in these systems. Materials with greater resistance to microstructure evolution in these conditions are increasingly necessary in the context of advanced nuclear systems where material performance and reliability will be challenged by even greater operational lifetimes, temperatures, and complex exposure environments.

Despite the promise of these materials, the high number of combinatorial possibilities presents a challenge for efficient materials design and a great need exists for the ability to rapidly characterize bulk properties using high-throughput diagnostics. Few systems have been synthesized and even fewer tested under relevant conditions such as radiation exposure. Such testing is necessary as many systems will experience precipitation or phase segregation during annealing or radiation exposure. The paucity of real data collected on fabricated materials is particularly limiting in the light of the desire to use machine learning tools to facilitate materials design in this space.

While thermal transport and elastic properties which are measurable using TGS may not be those of particular interest for many applications, changes in microstructure such as phase segregation causing loss of ductility or strength will also be notable through changes in these properties. As such, we propose to use measurements of thermoelastic properties of Ni- and Fe-based CSAs as a function of temperature and in situ in out-of-equilibrium conditions including annealing and irradiation. These data may be used as a high-throughput screening tool to inform down-selection for future material fabrication. In particular, with established National Laboratory partners providing archived materials at no cost, we will combinatorially test materials fabricated from Ni, Fe, cobalt (Co), chromium (Cr), aluminum (Al), manganese (Mn), and palladium (Pd) in non-equiatomic compositions. Initial benchtop testing will include cryogenic to high temperature material property collection as well as thermal annealing to test phase stability. Multiple rounds of high-throughput testing will be used to optimized material stability to conditions available using bench-top testing. A down-selected matrix of materials will be selected to study under both ion irradiation and temperature at the in situ ion irradiation TGS (I3TGS) facility at Sandia National Laboratory.

1.2 Nanocrystalline metallic alloys

These materials are very attractive due to their high strength, ductility, and wear resistance. However, they must be very carefully designed considering grain boundary stability against a variety of drivers including temperature and strain. Experiments and simulations have shown that both thermal conductivity and elastic modulus are reduced in nanocrystalline alloys compared to large-grain polycrystal or bulk single crystal specimens. For the modulus in particular, this effect is found to be strong at grain sizes below 50 nm. As the desired grain size for many nanocrystalline materials starts at 20 nm or below, the transition under some driving force from nanocrystalline effects to bulk elasticity should be readily identifiable using in situ TGS. Here, we will exploit the transition from nanoscale to bulk material properties to determine the stability of nanocrystalline microstructures in situ under temperature, irradiation, and possibly strain.

The most common engineering route pursued to design stability in nanocrystalline alloys is binary alloying in the hope that solute atoms pin grain boundaries or lower grain boundary energies. Further solid solution alloying beyond binaries provides a much greater compositional space to explore, however the high number of combinatorial possibilities for many-component alloys presents a challenge for efficient materials design. At the extreme, compositions as complex as five-component high-entropy alloys have been shown to form stable nanocrystalline states at ambient conditions. To explore grain stability during processing in bulk nanocrystalline metals, we will initially study nanocrystalline effects in systems with may readily be fabricated with well-characterized grain size distributions such as platinum (Pt), platinum-gold (Pt-Au), or Ni-Fe. These materials will be sourced at no cost from established National Laboratory and University collaborators. Once the nanograined effect on thermal and elastic properties has been noted reliably, we will study grain stability under a variety of conditions including temperature (on-site at INL) and irradiation (at the I3TGS facility). Using this high-resolution stability map, the microstructure corresponding to the initiation of runaway growth, if present, may be easily isolated. This transition microstructure will be analyzed using traditional techniques to provide a mechanistic understanding why the implemented stability criteria has failed. Finally, this understanding will be used to inform future material fabrication choices and the cycle will be iterated.

Task 2 – Method Development

2.1 Elastic depth profiling

Standard TGS experiments induce monochromatic periodic excitations on the samples under interrogation and extract material properties averaged over a fixed volume from the surface. The depth to which properties are measured is set exclusively by the projected fringe spacing, normally on the order of 1-10 µm, as the intensity of the acoustic displacement decays exponentially from the excitation surface. For homogeneous materials, this sensitivity depth is of no great concern, but for surface-layered systems, for example either thin films or ion-implanted materials, tuning this surface sensitivity depth to match the relevant length scale of the system in question is extremely important. This stage of the proposal will develop single-shot depth profiling in TGS measurements by using harmonically generated excitations.

Experimentally, this process will consist of combining four (or more) pumping laser pulses in a geometrically fixed configuration at the sample surface as opposed to the traditional two. Excitations generated with differing wavelengths exhibit dynamics determined by the homogenized properties of layers of differing thicknesses. New analytical tools will also be developed based on existing physical models to extract parameters from multi-wavelength signals. This capability will provide a useful tool in the study of any layered material and will specifically be useful in the study of ion-irradiated materials. Once developed, these methods would be immediately used as the irradiation-stability of both HEAs and nanocrystalline metals is of interest. At present, no other tools exist by which the micron-scale depth profiling of elastic properties proposed here may be accomplished in a single-shot measurement.

2.2 Elastic tensor optimization

Recently, methods have been developed to use surface acoustic waves (SAW) propagation speeds mapped over large areas on polycrystalline metallic alloys to determine all fourth-order elastic constants Cijkl. As part of this work, we will develop new elastic constant optimization methods which do not require mapping of SAW speeds in a polycrystal. Initially, an analytical model will be developed which identifies the minimal set of necessary single crystal orientations and directions along which SAWs must be measured to optimize a complete elastic tensor. A number of wave speeds equal to (or greater than, depending on symmetry) the number of independent elastic constants (three for cubic materials) must be measured to unambiguously generate this tensor. Once a criterion for unambiguous optimization is created, the inverse problem will be posed. Namely, we will create a test function to determine whether Cijkl may be determined from a user-defined set of SAW polarizations. This capability will be used in tandem with electron backscatter diffraction (EBSD) orientation mapping on large-grained polycrystals to identify locations (ie. triple junctions) at which a single TGS measurement may be made to optimize an entire elastic tensor.

Combining this minimal-set optimization approach with the ability to locate particular microstructural features in the electron microscope will open the possibility of mapping a complete elastic tensor as a function of processing using a single or small number of parallel experiments. For example, using large grained polycrystalline HEAs, complete elastic tensors could be easily captured from cryogenic to high temperatures. Such detailed information is either not available to multi-scale modelers of material behavior or only available for a few alloy compositions. Making this information available using high-throughput testing will increase the efficacy of computational materials design and provide a rich database for new data-hungry machine learning design tools

The initial TGS capability will be operational in the first half of FY20. Analytical investigation for Task 2.2 will begin in parallel with facility construction and the development capability in Task 2.1 will be investigated once the main TGS facility is operational. Sample matrices for Task 1.1 and 1.2 will be sourced from University and National Laboratory collaborators during the first half of FY20, and will be investigated continually through FY20 and 21. In situ irradiation experiments are anticipated twice per FY contingent on facility access through external user proposals. Each subtask will be a contained package of one (or more in the case of Task 1) publications. Task 1 publications will target relevant materials-focused or general audience journals such as Acta Materialia, Scripta Materalia, Materials Research Letters, or Nature Communications. Technical development projects will be target at applied physics journals such as Applied Physics Letters or the Physical Review family. Results will be discussed with the community at meetings such as The Minerals, Metals, and Materials Society (TMS) and the Materials Research Society (MRS).

SECTION C. Environmental Aspects or Potential Sources of Impact:

Air Emissions

Minor emissions could occur from research activities. Air emissions from this activity are covered under APAD-04-52.

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Discharging to Surface-, Storm-, or Ground Water

N/A

Disturbing Cultural or Biological Resources

N/A

Generating and Managing Waste

Industrial and hazardous waste will be generated during this work. Industrial waste, in the form of common lab waste is expected. Small amounts of hazardous wastes, in the form of solvents, wipes. or other materials may be generated. Corrosive waste may be treated by elementary neutralization. Waste will be managed by Waste Generator Services (WGS). Small amounts of lab washwater may be discharged to the Idaho Falls sewer system. All discharges will comply with sewer discharge limits.

Releasing Contaminants

Small amounts of chemicals may be discharged to the Idaho Falls sewer system in accordance with sewer regulations.

Using, Reusing, and Conserving Natural Resources

All applicable waste will be diverted from disposal in the landfill when possible. Project personnel will use every opportunity to recycle, reuse, and recover materials and divert waste from the landfill when possible. The project will practice sustainable acquisition, as appropriate and practicable, by procuring construction materials that are energy efficient, water efficient, are bio-based in content, environmentally preferable, non-ozone depleting, have recycled content. and are non-toxic or less-toxic alternatives. New equipment will meet either the Energy Star or Significant New Alternatives Policy (SNAP) requirements as appropriate.

SECTION D. Determine Recommended Level of Environmental Review, Identify Reference(s), and State Justification: Identify the applicable categorical exclusion from 10 Code of Federal Regulation (CFR) 1021, Appendix B, give the appropriate justification, and the approval date.

For Categorical Exclusions (CXs), the proposed action must not: (1) threaten a violation of applicable statutory, regulatory, or permit requirements for environmental, safety, and health, or similar requirements of Department of Energy (DOE) or Executive Orders; (2) require siting and construction or major expansion of waste storage, disposal, recovery, or treatment or facilities; (3) disturb hazardous substances, pollutants, contaminants, or Comprehensive Environmental Response, Compensation, and Liability Act (CERCLA)-excluded petroleum and natural gas products that pre-exist in the environment such that there would be uncontrolled or unpermitted releases; (4) have the potential to cause significant impacts on environmentally sensitive resources (see 10 CFR 1021). In addition, no extraordinary circumstances related to the proposal exist that would affect the significance of the action. In addition, the action is not “connected” to other action actions (40 CFR 1508.25(a)(1) and is not related to other actions with individually insignificant but cumulatively significant impacts (40 CFR 1608.27(b)(7)).

References:
10 CFR 1021, Appendix B, B3.6, "Small-scale research and development, laboratory operations, and pilot projects"

Justification:
The proposed R&D activities are consistent with CX B3.6 "Siting, construction, modification, operation, and decommissioning of facilities for small-scale research and development projects; conventional laboratory operations (such as preparation of chemical standards and sample analysis); small-scale pilot projects (generally less than 2 years) frequently conducted to verify a concept before demonstration actions, provided that construction or modification would be within or contiguous to a previously disturbed area (where active utilities and currently used roads are readily accessible). Not included in this category are demonstration actions, meaning actions that are undertaken at a scale to show whether a technology would be viable on a larger scale and suitable for commercial deployment;"

Is the project funded by the American Recovery and Reinvestment Act of 2009 (Recovery Act) ☐ Yes ☒ No

Approved by Jason Sturm, DOE-ID NEPA Compliance Officer on: 11/19/2020