

SECTION A. Project Title: Real Time Non-Equilibrium Studies for Nuclear Materials Performance

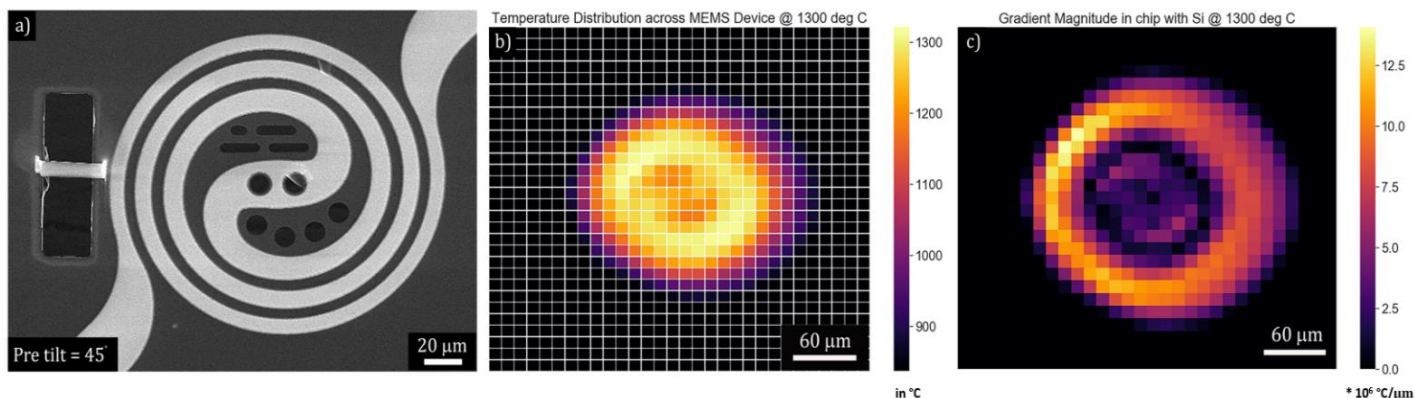
SECTION B. Project Description and Purpose:

Project description/abstract:

This study will enable the investigation of thermally driven phenomena that governs the microstructural evolution of nuclear materials by tailoring non-equilibrium thermal gradients conditions across a sample using a novel *in-situ* prototype. The proposed *in-situ* technology will shift current fuel performance experimental methodology from post irradiation examination to a real time evaluation, using *in-situ simulation of in-reactor thermal conditions*. This capability will be based on the development of controlled thermal gradient for microscopy, which is not commercially available. Such *in-situ* technique can offer rapid feedback to experimentalist and modelers on the dynamics of important fuel performance phenomena (such as elemental diffusion, chemical interactions, phase transformation, and grain boundaries behavior). Analyzing these processes will provide a better understanding of the physical world and of material behavior, thus permitting harnessing new materials to be used in current and future reactors to the benefit of people and society. The proposed *in-situ* device will allow the evaluation of material behavior in real time under non-equilibrium and could reduce time and cost of experiments associated with testing and qualification of new nuclear materials and fuel, thus accelerating nuclear materials discovery and deployment. Indeed, the proposed device could reduce irradiation campaign by down selecting materials based on their thermal behavior before irradiation testing is conducted. Moreover, it will support rapid experimental validation of modelling and simulation tools (e.g., MARMOT) and answer basic material science questions. To summarize, the proposed *in-situ* device will enable analyzing phenomena relevant to fuel performance and safety (e.g., phase transition, chemical interaction, element redistribution), by 'simulating' transients' conditions and by allowing observations dynamically in real time.

Significance:

Studies, such as the one proposed here, will give us a deep and rapid insight into material behavior in non-equilibrium conditions, providing insight into choosing the best materials to irradiate in reactors, and thus permitting a screening of material based on thermal driven phenomena. Currently experiments outside of reactor which are used to evaluate nuclear material behavior, are mostly based on furnace heat treatment, followed by post-mortem analyses [1-3]. This methodology requires multiple steps, samples transfers, and extensive sample preparation, which is time consuming and expensive. Moreover, this method does not provide information on the kinetics of the phenomena. *In-situ* heating stages for microscopes have been used to evaluate phenomena, such as thermally activated chemical interactions, phase transformation, and grain boundary behavior (e.g. [4-9]). However, their application to nuclear materials and especially nuclear fuel has been limited (e.g. [10-13]). Traditional in-situ heating stages use a furnace-based heater, which consists of a 'sample cup' that separates the tungsten/tantalum-based heater from the sample. This design introduces large thermal lag due to a mismatch in the thermal expansion of the sample cup and the sample. The thermal lag results in large thermal equilibration times, which introduces large 'sample drift' [14]; i.e., initial processes of a thermally activated reaction cannot be observed. The new generation of micro electromechanical systems (MEMS) based heating holders were designed to mitigate these issues. These holders are advantageous due to the small thermal mass of MEMS micro heaters that results in reduced sample drift [15,16], fast heater response time that enables rapid heating and cooling of samples, and a homogeneous temperature distribution across the sample support membrane, which allows for reliable and reproducible in-situ TEM (transmission electron microscopy) heating experiments [17,18]. Previously, MEMS-based systems have been used to study thermally activated processes in equilibrium and non-equilibrium conditions [19,20]. However, these studies were performed under uniform heating conditions (i.e., no thermal gradient exists across the sample) and optimized for steady state conditions. These steady state conditions are far from the conditions that exist inside nuclear fuel. *Inside the reactor, heat transfer is one of the most important underlying mechanism to harvest nuclear power, thus a steep thermal gradient exists across the fuel and the coolant medium. Thermal gradients influence the microstructure and chemistry of the fuel, thus its properties.* Reproducing such conditions inside the TEM for the study of nuclear materials has never been attempted before. *The proposed study will enable us to investigate thermally driven phenomena that governs the microstructural evolution of nuclear materials by tailoring non-equilibrium thermal gradients conditions across a sample inside a TEM. A first of a kind (in-situ device) prototype has been designed, developed, and preliminarily tested at the Ohio State University (OSU) on a simple system. The MEMS-based heating stage, shown in Error! Reference source not found. has been modified to generate steep thermal gradients (~ 10⁶ K/m) across the TEM sample, shown in Figure 1 (a). Infra-red thermography maps (Fig. 1(b) were acquired to calibrate the temperature distribution for specific no radioactive materials (such as silicon and strontium titanate) and measure the thermal gradients (Fig.1(c) across the modified device [15,16,18]. The modified MEMS heater is compatible with a double tilt TEM heating holder (DENSsolutions) and can achieve conditions of interest to nuclear reactor applications (Table 1). Such application will require extensive testing, possible modification to tailor the device for thermal gradients on the materials of interests (nuclear fuels and cladding alloys), and investigations of chemical compatibility between the fuel material and the device, which may require the use of liners or coating.*



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Figure 1: (a) MEMS micro heater with a modified window next to the spiral heating element, (b) Infra-red thermal image of the temperature distribution across the MEMS microheater device at 1300°C, and (c) thermal gradient map of the MEMS device.

Table 1-Parameters of the device compared to reactor transient conditions.

Parameters	Device	Reactor Conditions
Thermal Gradients (°C/m)	Up to 10^6	4×10^4
Heating & Cooling rates (°C/s)	U to 10^3	Up to 1300
Maximum Temperature (°C)	1300	1100

This system will be able to support testing of new materials and could reduce irradiation campaign by down selecting materials based on their thermal behavior before testing the influence of irradiation. This will result in faster and cheaper material development, thus in accelerated RD&D (research development and deployment). The modified MEMS based micro heater device will be further enhanced to test both fuel and claddings materials, in various temperature regimes. This system could answer many basic material science questions important to reactor performance even before reactor testing, such as:

- Fuel cladding chemical interaction (FCCI),
- Fission products (FP) chemical form and migration,
- Element redistribution and phase transformation,
- Microstructural evolution such as recrystallization and grain boundaries behavior.

Indeed, such phenomena are relevant to the licensing of reactor materials due to the necessity to ensure safety of the materials under unplanned or unforeseen conditions.

This device could be also applied for separate effect studies and will permit to evaluate separately thermal induced effect in materials from the radiation induced phenomena, which occur concomitantly in reactor and thus can lead to misinterpretation of material performance. This system, moreover, could be of interest to various stakeholders as it permits to study material behavior in real time under non-equilibrium condition, supporting quick and cheap material development and qualification campaigns. This system will be a first of a kind capability for nuclear material studies and could become of interest to the basic energy science community (including academy, industry). Finally, such studies will expand Idaho National Laboratory (INL) scientific research impact in materials science.

Research Plan (about 1 pages):

Technical Concept (i) This proposal aims in developing a new in-situ TEM device for the study of nuclear material and cladding in different temperature regimes. While in-situ TEM heating stages are commercially available. This novel MEMS-based in-situ system will be able to create relevant thermal gradients in the desired materials, thereby, providing the possibility of investigating thermally driven phenomena of relevance to material science (e.g. diffusion phenomena, chemical interactions etc.) and to nuclear materials performance. This research will demonstrate a new innovative approach to observe dynamics of thermally driven phenomena *ex-reactor*. Our proposal aims in providing an early determination of the applicability of this new device to nuclear material research. The device will be optimized to work with fuel and irradiated materials.

Method and Facilities (ii):

Our first task will focus on performing validation tests using the modified MEMS-based heating stage on simulated materials (surrogate). These preliminary in-situ TEM studies would aid in understanding thermally driven microstructural events that occur when applying a controlled thermal gradient. Initially, this will be performed at OSU (TASK I) and will provide information on various microstructural phenomena such as phase transformations, recrystallization & grain growth, grain boundary character, chemical compatibility, temperature regimes and flexibility of the *in-situ* MEMS based device. Dr. Vijayan and Dr. Jinschek will oversee this task. No-radioactive materials for these first test will be produced at INL and will include simulated diffusion couples, cladding materials, and metals of interest. Following the protocol to use MEMS based devices to study nuclear materials under thermal gradients, and the concept will be transferred to INL to test on nuclear fuels (TASK II), such as metallic fresh fuel alloys (uranium-zirconium U-Zr and/or uranium-molybdenum U-Mo). A student from OSU will be invited for assistance in this phase and strengthen the collaboration and INL pipeline development. Dr. Di Lemma will oversee the work control and procedure development for the utilization of the device on nuclear and irradiated materials. Dr. He will be working with the student from the OSU on the in-situ heating TEM characterization. Finally, irradiated materials will be tested providing scientific insight to the coupling of thermal effect to material damage (TASK III). The research plan and tasks to be performed are summarized in Figure 2, together with proposed timeline and deliverables.

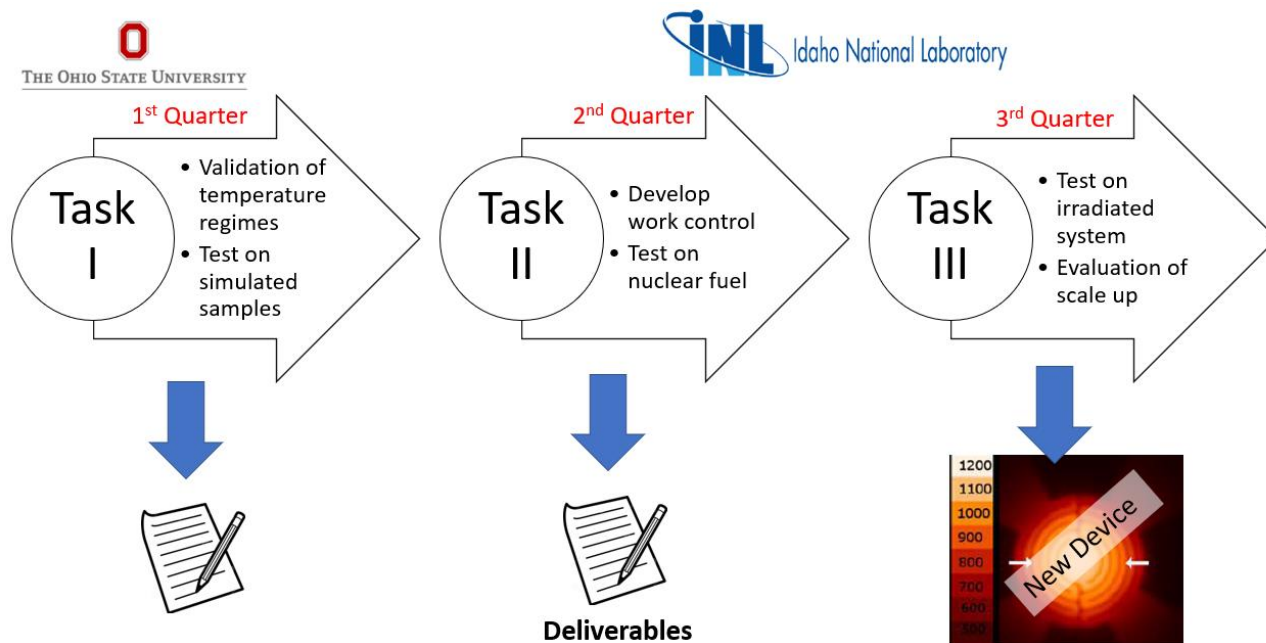


Figure 1-Summary of tasks, deliverables, and timeline.

Results and impact (iii):

The modified MEMS-based heating stage will considerably impact current experimental methodology, providing an avenue to test and qualify materials inexpensively and rapidly. The data obtained from these in-situ TEM experiments (using modified MEMS microheaters) will provide important scientific insight to the modelling community on the underlying processes that dictate the microstructural and chemical evolution in nuclear materials and fuel under non-equilibrium conditions. This system, finally, will provide real time and kinetics information on a materials' thermal response not obtainable with the "postmortem" experimental methodology. Once installed, this device will be available on INL instrumentation for the user community and programs. This unique capability will promote INL scientific vitality by providing a venue to investigate phenomena of interest for basic material science and incentivizing new proposals. This will increase the laboratory research impact. This device responds to necessity of department of energy (DOE) programs for a deeper understanding of material behavior under non-equilibrium condition and of a methodology to rapidly develop, test and qualify new materials for nuclear application. Thus, the MEMS will support the advancement of nuclear energy by providing a venue to test and demonstrate new nuclear materials ex-reactor via separate effect testing and supporting modelling and simulations development. This system initially designed and tested on TEM will be also evaluated for larger scale testing, such as X-ray diffractometers and scanning electron microscope.

Products and Deliverables (about ½ page):

The main deliverable will be a new in-situ technique relevant to basic material science studies (including experimentalists and modelers). This instrument will be an important research tool to study fuel performance and safety. Other deliverables, reported in Figure 2, include publications (estimated 2) and conference proceedings (estimated 2). These studies will finally promote scientific understanding of fuel behavior and chemical reactions under transient conditions of importance for the licensing process of nuclear materials.

Potential for Follow-On Activities or Harvest Strategy (about 1/2 page): This device will be of interest to various DOE's programs interested in evaluating material performance (e.g., Accident Tolerant Fuel-ATF, Advanced Fuel Campaign-AFC, or the US High Performance Research Reactor). The device will possibly attract also partners from the nuclear industry interested in material science and licensing of new materials. The set-up will permit to further studies on FCCI and element redistribution in nuclear fuel-cladding system, relevant to the understanding and future deployment of advanced reactor. Moreover, this system can be leveraged also for basic science studies, such as grain boundaries, phase transformation, oxidation, and diffusion kinetics of relevance also to modelling and simulation. This system has the potential to be apply to large scale testing, which will be evaluated during this research program.

When installed the investigators will be sure to communicate the achievement and capabilities of this instrument to the nuclear and material science community. Venue for this will be the proposed deliverables (conferences and publications). Moreover, we suggest that this system and its result will be promoted to the user community, through the nuclear science user facilities (NSUF) network and INL media if possible. *This in-situ technique will enable INL mission, by providing the venue to collect data necessary for developing, demonstrating, and licensing of new nuclear materials, via rapid in-situ methodology.*

Project Location: The activities listed above will be carried out primarily at The MFC Irradiated Materials Characterization Laboratory (MFC-1729) and the Electron Microscopy Lab (MFC-774). The material used in this project will be taken from the existing inventory at IMCL. If additional samples are needed, the material will be obtained from the Fuels & Applied Science Building (FASB) (MFC-787).

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SECTION C. Environmental Aspects or Potential Sources of Impact:

Air Emissions

Note: If this project or activity produces or causes air emissions, and it is not stated in this ECP how those emissions caused by this project or activity are exempt, then an APAD is required for documentation. Samples are expected to be prepared at the MFC Irradiated Materials Characterization Laboratory (MFC-1729). Any potential air emissions from the handling and manufacturing of the radioactive samples will be covered by APAD INL-13-008 R3: Installation and Operation of Bench Top Laboratory Equipment At The IMCL and APAD INL-18-010: EML Operations.

Discharging to Surface-, Storm-, or Ground Water

N/A

Disturbing Cultural or Biological Resources

The MFC Irradiated Materials Characterization Laboratory (MFC-1729) and the Electron Microscopy Lab (MFC-774) and are eligible to the National Register of Historic Place (NHRP). As a result, any proposed modifications to this facility for this project requires a review by the Cultural Resource Management Office (CRMO).

Generating and Managing Waste

This work is expected to generate small amounts of Low-Level Radioactive Waste (LLW) during the sample processing. All Solid Waste will be managed by Waste Generator Services (WGS).

Releasing Contaminants

When chemicals are used during the project there is the potential for spills that could impact the environment (air, water, soil).

Using, Reusing, and Conserving Natural Resources

Waste will be diverted from the landfill to the extent possible.

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: This EC references the Categorical Exclusion B3.6 "Small research and development, laboratory operations, and pilot projects."

Justification: Project activities in this EC are consistent with 10 CFR 1021 Appendix B to Subpart D, Categorical Exclusion 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 two years) frequently conducted to verify a concept before demonstration actions, provided that construction or modification would be within or contiguous to a previously disturbed or developed area (where active utilities and currently used roads are readily accessible). Not included in this category are demonstration 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:02/03/2021