APS Workshop 3: *Operando* Synchrotron Experiments on Advanced Manufacturing Processes

Tuesday, May 7, Morning

8:00 – 8:30 Workshop Organizers
Introduction

8:30 – 9:00 Steven Van Petegem (Paul Scherrer Institute)
Operando *X-ray Diffraction and Imaging during Laser Powder Bed Fusion*

9:00 – 9:30 Nicholas Calta (Lawrence Livermore National Laboratory)
Synchrotron Experiments for Advanced Manufacturing Studies at Lawrence Livermore National Laboratory

9:30 – 10:00 Break

10:00 – 10:30 Anthony Rollett (Carnegie Mellon University)
Additive Manufacturing: Synchrotron X-ray Microscopy Insights

10:30 – 11:00 Amy Clarke (Los Alamos National Laboratory)
Visualization of Solidification under Additive Manufacturing Conditions

11:00 – 11:30 Xuan Zhang (Argonne National Laboratory)
Additive Manufacturing of 316H Stainless Steels for High Temperature Nuclear Applications

11:30 – 1:30 Lunch Break

Tuesday, May 7, Afternoon

1:30 – 2:00 Ho Yeung (National Institute of Standards and Technology)
Pointwise Control for Laser Powder Bed Fusion Additive Manufacturing Process

2:00 – 2:30 Lianyi Chen (University of Wisconsin, Madison)
In-situ *Characterization of Phase Transformation Dynamics in Metal Additive Manufacturing Processes*

2:30 – 3:00 Nathan Crane (Brigham Young University)
In-situ *Synchrotron Imaging of Droplet/Powder Interactions in Binder Jetting Process under Realistic Printing Conditions*

3:00 – 3:15 Break

3:15 – 3:45 Atieh Moridi (Cornell University)
Advancements in Microstructure Control in Additive Manufacturing through Operando *X-ray Studies*

3:45 – 4:15 Xun Liu (Ohio State University)
In-situ *Synchrotron Analysis on Ultrasonically Assisted Laser Melting Process*

4:15 – 4:45 Frank Pfefferkorn (University of Wisconsin, Madison)
Material Flow Visualization during Friction Stir Welding Using High-speed X-ray Imaging

4:45 – 5:00 Group Discussion

5:00 Adjourn

Operando X-ray Diffraction and Imaging during Laser Powder Bed Fusion

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Laser-based powder bed fusion (LPBF) stands out as a widely employed technique in metal additive manufacturing due to its versatility and applicability. Despite extensive research efforts in this field, the relationship between process parameters and microstructural evolution remains a subject of ongoing investigation. To address this, we have developed a compact LPBF device compatible with synchrotron x-ray diffraction and imaging beamlines [1-4]. This setup, complemented by fast x-ray detectors, enables real-time monitoring of crystallographic phase changes, local temperature variations, and defect formation during LPBF with temporal resolutions in the tens of microseconds [4-14].

In this presentation, I highlight recent developments and findings obtained using this setup. Specifically, we demonstrate the utility of *operando* x-ray diffraction in tracking phase transformations and local cooling rates in Ti-alloys [5, 10, 12], Al-alloys [8], and steel [9, 10, 14]. These experimental observations provide valuable insights for validating computational simulations and models [3, 9]. Additionally, fast *in-situ* x-ray radiography offers insights into defect formation in Ni alloys [4]. Furthermore, by integrating acoustic and optical sensors, our experiments provide data for the development of machine-learning algorithms [6, 13]. Finally, *operando* tomography during LPBF of ceramics allows tracking the evolution of melt pool, pores, and cracks in 3D [15].

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Synchrotron Experiments for Advanced Manufacturing Studies at Lawrence Livermore National Laboratory

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Advanced Materials and Manufacturing research and development is an area of emphasis at Lawrence Livermore National Laboratory (LLNL). Efforts in this area span a wide range of materials and processes, including significant effort in the area of metal additive manufacturing. Synchrotron-based experiments are a powerful tool for understanding both the process dynamics as well as how these dynamics influence the behavior of a finished component. This talk will discuss a range of LLNL efforts in advanced manufacturing, with a particular focus on the use of synchrotron-based probes. It will include results from high-speed radiography and diffraction probing process dynamics during metal additive manufacturing, diffraction experiments to quantify residual stresses in additively manufactured components, and *in-situ* diffraction during deformation in additively manufactured metals. Future directions and experimental needs will also be discussed.

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Additive Manufacturing: Synchrotron X-ray Microscopy Insights

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Additive manufacturing (AM), *aka* 3D printing, is a relatively new technology that has given rise to the "maker culture" and an intense interest in design. That has carried over into metals AM, which has jumped almost immediately into manufacturing of actual parts in a variety of alloys. In doing so it has liberated thinking about part design albeit within certain constraints and complex components have been deployed that were previously inaccessible, e.g., high temperature heat exchangers (HX). An example is described of the co-design of HX against printing constraints, alongside evolution in alloy choice. Nothing is ever as simple as it seems, however, and the reliability of parts that must carry load depends on the internal micro-structure, especially with respect to fatigue loading. This motivates detailed study of all aspects of materials microstructure ranging from defect structures to strain, all of which is ideally suited to the use of intense sources of high energy x-rays as only advanced light sources can deliver. Computed tomography (CT) has revealed the presence of porosity in all additively manufactured metals examined to date and confirmed that appropriate process control can limit it. CT has also provided data on surface condition that we are trying to link to fatigue performance. High speed radiography reveals even more crucial details of how laser light generates vapor cavities that can deposit voids past a critical instability point. "Hot" cracking has been imaged as it happens during the solidification process, which offers the possibility finding printing recipes for alloys previously considered off-limits to 3D printing. High-speed, high-resolution diffraction in stainless steel, alloy 718 and Ti-6Al-4V reveals unexpected solidification and precipitation sequences. Diffraction microscopy reveals the highly strained nature of printed metals and how microstructure and internal strain state evolves during subsequent annealing and/or annealing. Permeating all these activities is machine learning as an invaluable tool and aid to the researcher.

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Visualization of Solidification under Additive Manufacturing Conditions

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Additive manufacturing (AM) processes typically produce large temperature gradients, high solidification rates, and repeated cycles of heating and cooling. The local processing conditions experienced during AM (e.g., thermal gradients and solid-liquid interface velocities during solidification) will dictate microscopic structure (i.e., microstructure) evolution. Here we visualize melt pool dynamics and solidification of metallic alloys during simulated AM by real-time imaging (e.g., synchrotron x-ray and dynamic transmission electron microscopy) and computational modeling to link local processing conditions to microstructure development. A deeper understanding of solidification under AM conditions is needed to optimize processing conditions across AM technologies, predict and control microstructure evolution and resulting properties, and design alloys for AM.

Additive Manufacturing of 316H Stainless Steels for High Temperature Nuclear Applications

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To use laser powder bed fusion (LPBF) 316H stainless steel (SS) in high temperature nuclear reactor applications, research efforts are needed to develop optimized laser processing conditions and proper heat treatments. This talk gives an overview of experimental research efforts at ANL towards a process understanding of LPBF 316H SS under the US-DOE Advanced Materials and Manufacturing Technologies (AMMT) program. Highlights will be given on utilizing simultaneous wide-angle x-ray scattering (WAXS) and small-angle x-ray scattering (SAXS) measurements during the *in-situ* heating of LPBF 316H SS samples at the APS beamline 1-ID-E in understanding the microstructural evolution. Microstructure-mechanical property correlation will be addressed. The results are an important first step towards the qualification of LPBF 316H SS for reactor applications.

Pointwise Control for Laser Powder Bed Fusion Additive Manufacturing Process

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The metal laser powder bed fusion (LPBF) process is a widely used metal additive manufacturing (AM) method, renowned for its ability to create complex features, optimized geometries, lightweight parts, and intricate designs. Despite advancements, LPBF has not fully realized its potential due to quality issues arising from inadequate process control that fails to adjust to the dynamically changing thermal conditions dictated by scan sequences and part geometries. To address this, a novel pointwise control technique employing time-stepped digital commands has been introduced, enabling precise synchronization of laser power and diameter at each point along the interpolated scan path. This technique exceeds the capabilities of conventional line-wise control by allowing continuous variations in laser power, diameter, and speed, which facilitates the implementation of model-based scan strategies and digital twin-based real-time process control. Additionally, pointwise control assures complete synchronization between processing commands and monitoring data. It also offers a platform-independent, unambiguous description of the scan strategy and control, and quality traceability. An open platform AM control framework based on pointwise control has been developed, resulting in several testbeds, including one with synchrotron x-ray measurement integration. Case studies on various advanced scan strategies and *in-situ* process monitoring on these platforms are presented. A digital twin framework, built upon pointwise control, has been proposed, comprising digital twins of process design, control, monitoring, and the printed part for LPBF process optimization and quality assessment. These advancements edge us closer to unlocking the full potential of AM, enhancing part quality and process efficiency.

In-situ Characterization of Phase Transformation Dynamics in Metal Additive Manufacturing Processes

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Fusion-based additive manufacturing technologies enable the fabrication of geometrically and compositionally complex parts unachievable by conventional manufacturing methods. However, the phase transformation behavior of materials under non-uniform and far-from-equilibrium heating/cooling conditions is not fully understood, which is a major barrier for rational design of materials and processing conditions to consistently obtain desirable phases in the as-printed parts. In this talk, I will present the key data and insights my research group obtained on uncovering phase transformation dynamics under additive manufacturing conditions by using high-speed high-energy synchrotron x-ray diffraction.

In-Situ Synchrotron Imaging of Droplet/Powder Interactions in Binder Jetting Process under Realistic Printing Conditions

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Binder jetting (BJ) is an additive manufacturing process with great promise for low-cost manufacturing due to its capacity for high-speed manufacturing with low-cost equipment [1]. In BJ, the geometry is formed by spreading thin layers of powder and then selectively bonding the powder by inkjet printing of binder droplets into the bed. While this process is low-cost, it is particularly challenging to understand the dynamics of the droplet/powder interaction that are critical to the successful part formation. The first work in this area considered single lines printed into loose powder beds [2], but these conditions are not representative of actual printing conditions.

I will present our approach to studying BJ printing using *in-situ* high speed x-ray imaging to understand the droplet powder interactions during BJ printing at the Advanced Photon Source using Beamline 32-IDB. Specifically, we examined the size of the powder region impacted by droplet impact and the quantity of powders ejected. We will discuss a new equipment setup that permits imaging of multiple lines [3] and the capability to image both loose and rolled powder beds. Unique issues encountered with BJ include binder evaporation due to heating of high-absorptivity materials (316 Stainless steel). We will also discuss the ability to observe particle relocation below the surface and ejection above the surface. These results provided valuable insight into the relationship between powder relocation and binder infiltration as well as insights into the interaction between adjacent lines of droplets [4].

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Advancements in Microstructure Control in Additive Manufacturing through *Operando* X-ray Studies

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This presentation unveils recent advancements in microstructure control in additive manufacturing (AM) through three *operando* x-ray diffraction studies. First, we delve into harnessing phase metastability for grain size control in AM using a FeMnCoCr system. By strategically manipulating phase stability, we achieve notable grain refinement and breakdown of epitaxial columnar grain growth, offering new avenues for tailoring materials with desired grain sizes and morphology. Second, I introduce the concept of alloy amalgamation, unlocking the co-existence of multiple phases in titanium alloys through blending commercial alloys during the AM process. *Operando* synchrotron x-ray diffraction reveals the induction of various martensitic phases and a γ-fcc phase in the novel titanium alloy, highlighting the ability of alloy amalgamation to tailor microstructures for advanced structural engineering. Third, we investigate dendritic deformation modes in AM using *operando* x-ray diffraction, analyzing thermomechanical deformation during solidification. Understanding phenomena such as torsion, bending, and fragmentation aids in optimizing printing strategies to obtain specific microstructural features, advancing the control of AM-produced parts' mechanical properties and defects. Finally, new measurement techniques to augment or enhance each of these studies will be discussed.

In-situ Synchrotron Analysis on Ultrasonically Assisted Laser Melting Process

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Metal additive manufacturing (AM) is advantageous in its high agility for rapid prototyping of complex geometries. However, the undesired as-solidified microstructure and high residual stress inherent with the AM process make it challenging for widespread applications. Besides post-processing treatment, *in-situ* process modification is receiving rapid growth of attention. Power ultrasound (UA), which typically operates at frequencies of 20kHz or 40kHz and power outputs of 1-5kW, has shown various benefits in melt and solidification behavior, including grain refinement and degassing. UA has recently been utilized to improve various types of AM processes of different materials and consistently show promising results in promoting columnar to equiaxed transition, minimizing segregation and reducing anisotropy. On the other hand, fundamental understanding of these benefits remains elusive.

In this study, a UA system that is compatible with laser-based AM replicator is developed for *in-situ* synchrotron x-ray diffraction (SXRD) analysis at the APS 1-ID beamline. Under 40kHz UA vibration and 6mm vibration amplitude, SXRD shows a faster melting and solidification rate for AA4043 aluminum alloy. The static SXRD diffraction patterns in the UA sample exhibits higher continuities of diffraction rings, indicating a more refined grain structure, which is verified with optical microscope and electron backscatter diffraction (EBSD) characterization results. Thermal histories determined from the *in-situ* lattice parameter evolution demonstrate an increased cooling rate with UA. With a recent update of the UA system with 20kHz frequency and 14mm amplitude, the solidification structure of commercially pure Ti is significantly refined. This research shows the potential of *in-situ* SXRD in revealing the underlying mechanisms of UA enhancement on AM processes.

Material Flow Visualization during Friction Stir Welding Using High-speed X-ray Imaging

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The intricacies of material flow dynamics during friction stir welding for different aluminum alloys have been a long-hypothesized phenomenon in the literature, with studies predicting the material flow using post-factum microstructural characterization and numerical simulations. The radiography experiments conducted at the Advanced Photon Source (beamline 32-ID-B) in June 2022 will be described, along with lessons learned for future investigations into solid-state manufacturing processes that rely on intense shearing. A white beam was used to image a 2 mm x 2 mm area at 20,000 frames per second. The friction stir tool made of H13 tool steel with threads and 3-flats on the probe was used to compare material flow in aluminum 6061-T6 and aluminum 7075 workpieces. The data is the first to present direct observation of the formation and filling of cavities at the trailing (and leading) edge of the friction stir tool. Observations and the numerical simulations that they verified will be presented. Finally, ideas for enabling the direct observation of process dynamics in friction stir welding, friction surfacing, and other solid-state processes will be presented.