

CLOSING THE MESOSCALE

GAAP

Los Alamos makes a bold proposal to understand and control material properties

Cindy Bolme and Amy Clarke don't see themselves as revolutionaries, and neither of them is talking about a revolution. They're just helping to start one.

The two Los Alamos scientists are contributing to a body of knowledge that, once integrated into the science-technology culture, is likely to revolutionize how we discover, develop, and apply new materials. Bolme's work is the more explosive, with investigations into shock physics, high-pressure dynamics, and the behavior of materials under extreme conditions. Clarke wants anything but an explosion as she investigates how controlling fabrication and processing parameters can produce materials that behave as predicted.

Bolme and Clarke are part of a group of early-career technical staff engaged in what Los Alamos physicist

Cris Barnes calls "science on the roadmap to MaRIE." Not a lady but an acronym for Matter-Radiation Interactions in Extremes, MaRIE is the multi-purpose, billion-dollar materials research complex and user facility proposed for Los Alamos National Laboratory.

MaRIE answers the mission need for an experimental facility that can accelerate the qualification, certification, and assessment of materials for national security and science solutions. A huge endeavor that would entail, at the very least, the construction of an electron accelerator, an x-ray free-electron laser, a diagnostic hall, and a materials fabrication facility, MaRIE would be separate from, but integrated with, existing Laboratory facilities and would eventually affect every program at the Lab in some way or another.

Barnes, who *does* see himself as a revolutionary, is the champion for MaRIE. Apart from managing the project, he is its staunchest and most vocal advocate, able to convey to scientists and laypeople alike the science and technology opportunities—and challenges—afforded by the momentous project. The roadmap that he helped create is a plan for all the experiments and projects that need to be proposed and completed to go from the Laboratory’s current accelerator facility, LANSCE, to the future accelerator complex, MaRIE. If all goes well, the roadmap will help document the making of a global resource suited to the challenges of the 21st century.

An answer to mission need

The National Nuclear Security Administration (NNSA) is beginning to recognize that if it is to uphold its mission to “sustain a safe, secure, and effective nuclear deterrent,” business cannot continue as usual. Many of the weapons in the U.S. nuclear stockpile have been in service well beyond their design lifetimes and will need to have parts and materials repaired, refurbished, or outright replaced to remain viable. Any new or modified part or material needs to be qualified and assessed, and the revamped weapon needs to be certified before it can return to service.

In principle, any aged material can be replaced with a brand-new material remanufactured to be identical to what was used originally, but remanufacturing can be very difficult and costly. Changing the material, however, or changing the process by which it is made raises questions about certifying its performance. If the only way to prove a material will maintain performance over its design lifetime is to wait a design lifetime and then see how it performs, the stockpile would become unsustainable. There is a mission need, therefore, to accelerate the qualification, assessment, and certification of parts and materials used for nuclear weapons, so when new materials and more flexible, better-understood, lower-cost, and certifiable processes become available, they can be readily utilized.

Part of the answer to that mission need is to test the part or material or weapon inside a supercomputer. Weapons scientists can run a simulation and expose the item to a

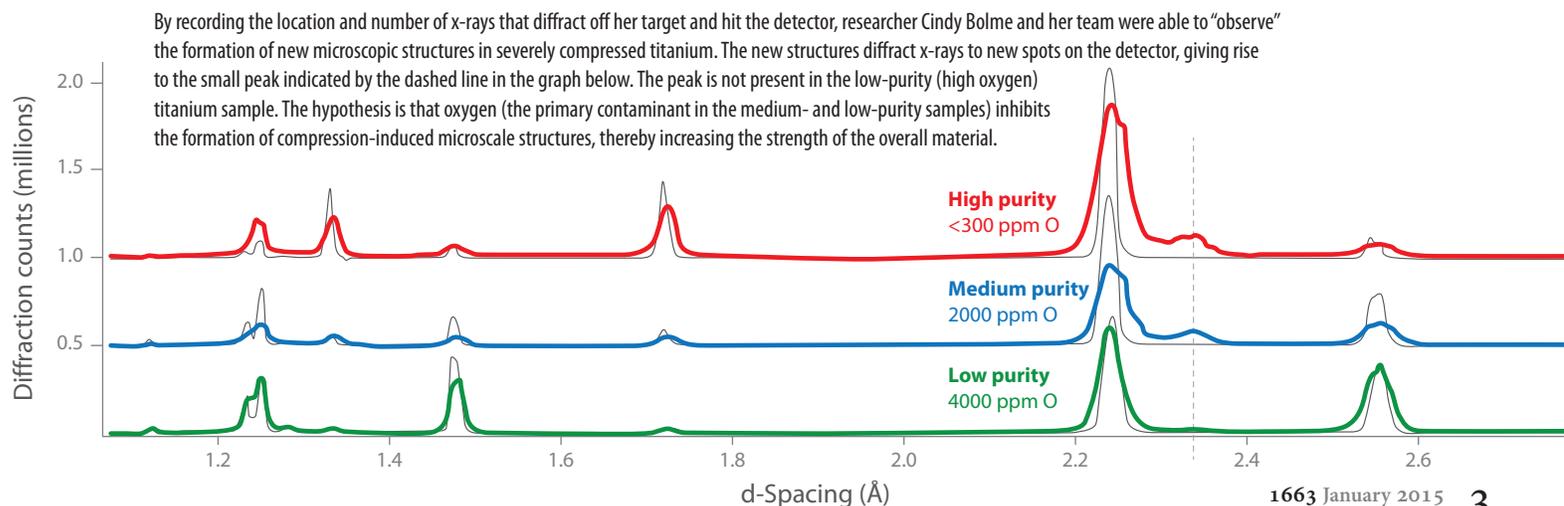
lifetime of random insults, then see how it performs in a weapons simulation. If confident of the simulation results, they can repeat the experiment enough times to build an accurate picture of performance that the NNSA can use to certify (or not) the real item.

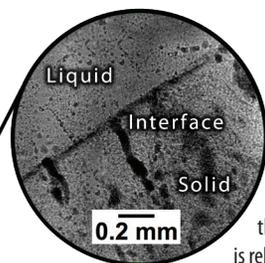
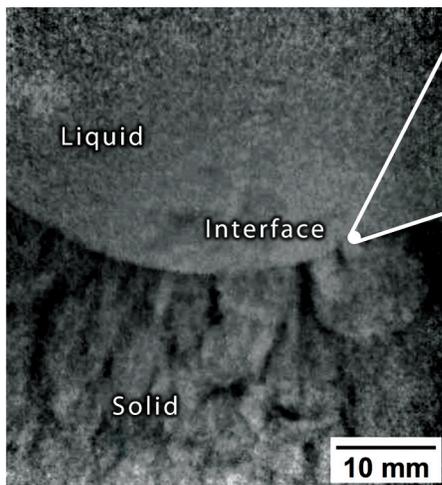
This idea of “validated simulation” was proposed in the 1990s for nuclear weapons stockpile stewardship, but it required a simulation capability that didn’t exist at the time. Not only were the computers too slow, but the fundamental understanding of materials was too limited. Models could not capture the full range of real-material behaviors, and there was little data to guide the way to improve those models.

Research tools developed and built under stockpile stewardship since then—DARHT at Los Alamos, NIF at Lawrence Livermore National Laboratory, U1a at the Nevada National Security Site—have contributed greatly to our understanding of weapons materials, mostly by probing atomic and macroscopic distance scales. But between those two domains lies the so-called mesoscale, with physically distinct features—crystal grains, defects, voids, grain boundaries, and interfaces—on the order of 10^{-8} to 10^{-4} meters in size. Mesoscale features have a tremendous influence on a material’s macroscopic behaviors and properties.

Unfortunately, the mesoscale is very difficult to probe—too large to be interrogated by atomic-scale probes and too small to be seen by macroscale tools. It’s a difficult domain to understand theoretically or to model, too, because it’s inherently inhomogeneous, and symmetry arguments that simplify calculations don’t apply. Consequently, a gap remains in the understanding of mesoscale properties and responses, especially in extreme temperature, pressure, strain, chemical, electromagnetic, and radiation environments. Without that understanding, a material’s behavior cannot be simulated with sufficient validity to allow certification.

Clearly, there is a need for a facility designed to look into and through the mesoscale and obtain data about the dynamic response of the materials used in nuclear weapons, from high explosives to plutonium. The facility would require a coherent x-ray source because x-rays diffract from meso-scale-sized structures, and coherent x-rays open the door to the greatest number of imaging and detection techniques. To





Amy Clarke and her team use proton and x-ray radiography to make movies of a metal alloy as it cools and solidifies. Proton radiography images a large sample volume and can capture mesoscale and macroscale processes during solidification, as evident in this image of the liquid-solid interface in an aluminum-indium alloy. Information gleaned from such images is relevant for structural development and casting. (Inset) This x-ray image shows the liquid-solid interface and microscopic structural development at 50x higher resolution.

observe fast material dynamics, the x-ray beam must be extraordinarily brilliant and must be pulsed, with several pulses arriving within a few tenths of a nanosecond, while

imaging the interior of thick metal samples would require exceptionally high-energy x-rays, on the order of 40–50 kilo-electron volts.

All of these attributes can be met by an x-ray free-electron laser, but one having what amounts to the highest-ever photon energy, the fastest-ever pulse repetition, and by far the largest-ever imaging volume. If built, it would be the most remarkable laser of its time.

Furthermore, though technically challenging, one can envision conducting dynamic experiments using x-ray diffraction, proton radiography, and electron radiography to probe a sample on several length scales simultaneously. Scientists would have the unprecedented ability to follow, in real time, the effect of, say, a shock wave on atomic-, meso- and macroscale structures. If there is also the capability to fabricate samples that have predetermined mesoscale structures, one could begin an experimental program with the intention of showing that materials with *these* types of internal structures, when in *those* environments, consistently behave *this* way. With sufficient data, simulations of material behaviors could achieve predictive capability, and the results of weapons simulations would be accepted with more confidence. This is the vision of MaRIE.

The accelerator complex, if approved, would not produce a beam for many years. The science being done in advance of MaRIE is therefore invaluable for filling in the mesoscale gap and for developing critical skills and expertise. On the day MaRIE opens its doors to a new and different future, experienced Los Alamos researchers can jump through feet first and hit the ground running.

Observations

Cindy Bolme loves it when new data crosses her laptop, be it from her own active experimental group or not. Any new data offers the possibility of increased insight into her

current research interest, what may best be called “dynamically induced material transitions,” or informally, “what happens when you hit a material really hard.”

Bolme uses a technique known as x-ray diffraction to probe atom positions at discrete times after her sample is hit by, say, a powerful shock wave. Her x-ray source is the Linear Coherent Light Source at SLAC National Accelerator Laboratory—more than a million-billion times brighter than any airport security x-ray machine and currently the most brilliant x-ray source on the planet. X-rays flood her sample, and the small fraction that diffract coherently from its atom planes are detected, their positions and intensities analyzed to reveal the atomic-scale structures from which they scattered. Illumination of the sample before and during the shock compression allows her to measure relative changes and infer structural changes.

For example, last year she completed a series of experiments to investigate titanium, the lightweight yet strong metal of choice for golf clubs, dental implants, jet plane frames, and ship propellers.

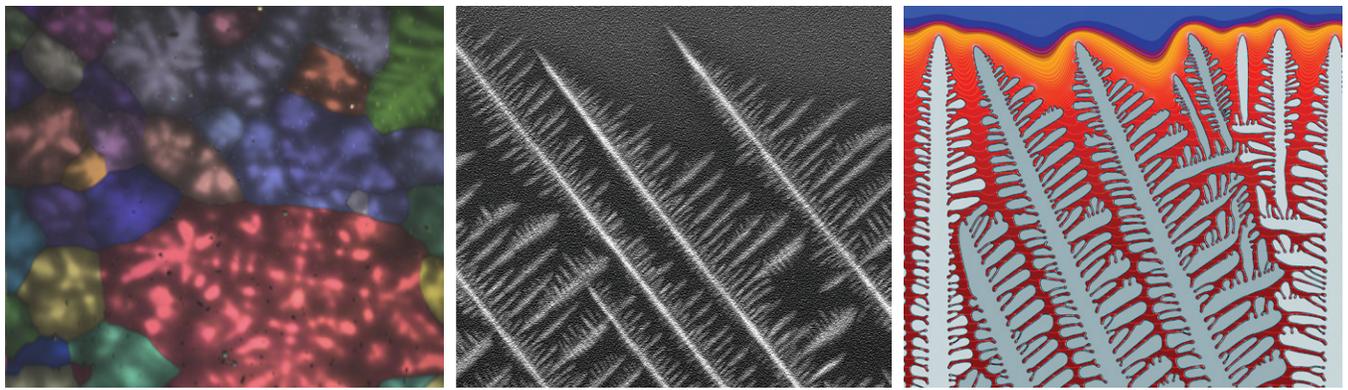
“Any metallurgist will tell you that if you want to make titanium strong, add oxygen,” Bolme says. “But why is that? What does the oxygen actually do?”

Under high stress, pure titanium can deform by undergoing a phase transition and by creating at least two types of structural defects, including twinning, in which a large group of atoms shift their positions slightly to form the mirror image of a neighboring group of atoms. The relative contribution from each of the three deformation mechanisms was not known, nor was their relative effect on titanium’s strength, but Bolme wanted to find out. She also wanted to know how the relationship between titanium’s defects and strength changed as a function of its purity.

Her x-ray diffraction studies showed that during compression, a secondary structure, consistent with twinning, grew within the pure titanium sample, implicating that defect as the primary reason for titanium’s deformation. The studies also showed that a set of samples with progressively increased impurity levels had progressively fewer defect structures. It was



Amy Clarke



(Left) An electron-backscattered diffraction image shows microscopic structures called dendrites (tree-like patterns within the colored grains) in an as-cast uranium alloy. (Center) Controlled directional solidification can dramatically affect the evolution and formation of dendrites, as seen in this x-ray image of a tin-bismuth alloy. The tin-rich dendrites are the first structures to form at higher temperatures as the melt cools, resulting in a chemically segregated solidification microstructure that significantly impacts the properties and performance of the resulting alloy. (Right) A computer simulation follows dendritic growth competition in an aluminum-copper alloy during solidification. The darker gray dendrites in the center are being eliminated by their lighter gray neighbors. This process directly affects the development of grains and grain boundaries.

Simulation results courtesy of Alain Karma (Northeastern University) and Damien Tourret (LANL).

known that when oxygen enters the material as an impurity, an oxygen atom will settle into random, non-binding sites between the titanium atoms. These and other studies suggest that oxygenated titanium remains strong because oxygen atoms inhibit twinning, presumably because the randomly spaced oxygen atoms don't allow the mirror-symmetric titanium twins to form.

Would MaRIE aid Bolme in her research? "To be able to image the mesoscale structure directly?" she says, eyebrows raised. "Oh yeah."

Process

Amy Clarke understands deeply what most of us would recognize as true but have little appreciation for—that what you get depends on how you got it. The properties that we assign to a material—the yield strength of a metal, its ductility, or its crystallographic texture—are determined as much by how the material was initially made and processed as by its composition.

Clarke and her team are interested in metal-alloy solidification at different length scales, which is relevant for technologically important processes such as casting. She was fascinated to learn that when she and her team changed the thermal gradient across a sample and adjusted the rate at which the liquid-solid interface advances, they could control the microscopic structures that solidified from the melt and change their morphology. Indeed, the young scientist, winner of an early-career award that currently funds the bulk of her team's research, has had remarkable success in producing various predetermined structures within metal alloys.

"Material properties derive from these structures," she says. "Being able

to control their morphological evolution is the first step in gaining control over the resulting material behaviors."

Clarke's team uses x-rays, typically at the Advanced Photon Source at Argonne National Laboratory, to image the mesoscale and microscale structures that form in her samples. Working with the Los Alamos proton radiography (pRad) team, she is also helping to pioneer the use of pRad to make images of solidification processes. Because pRad has great penetrating power, the researchers are able to image large, thick samples and therefore study macroscale processes, complementing the Clarke team's x-ray work.

And if MaRIE affords her simultaneous x-ray and pRad interrogations? Clarke merely imagines the world of possibilities for materials-processing studies.

Status

Though a top priority in some NNSA hallways, MaRIE is not yet approved as a project. Some only see the future complex through the focused lens of the weapons community; others see the broad scope of science tools represented by MaRIE and recognize that with the ability to control mesoscale structures comes the opportunity to create a more abundant future. Los Alamos is committed to submitting the case for the mission need by this summer.

MaRIE has already had a positive impact. In the words of its champion Barnes, "Whatever one thinks about the likelihood of an x-ray free-electron laser ever happening at Los Alamos, the recognition of the importance of the problem and the science challenges that it presents are leading us to do some really cool science and technology, now, today."

—Jay Schecker



Cindy Bolme