

Smoother surfaces make for better accelerators

April 18 2024, by Tamara Dietrich, Kandice Carter



Evolution of the average surface roughness. Credit: *Physical Review Accelerators and Beams* (2023). DOI: 10.1103/PhysRevAccelBeams.26.103101

With every new particle accelerator built for research, scientists have an opportunity to push the limits of discovery. But this is only true if new particle accelerators deliver the desired performance—no small feat in a world where each new machine is a first of its particular kind. At each project opportunity, researchers try to refine the preparation methods of key components so as to get a "better bang for the buck."



Accelerator scientists at the U.S. Department of Energy's Thomas Jefferson National Accelerator Facility have been leading that refinement process. Building on decades of empirical learning, they're cataloging how particle <u>accelerator</u> components are made, what the micro-roughness of the surface is like, and how all of this affects the components' performance. Their ultimate goal is a functional method for investigating and predicting a particle accelerator's ultimate performance based on the specific recipe used to prepare its parts.

"We're trying to find a way to understand the different things that are going on and then with that understanding craft a process that's very intentional," explained Charles Reece, a senior accelerator physicist who retired from Jefferson Lab's SRF Institute last year.

Now, the team has investigated several representative surface treatments to test their methodology. They've found that it not only successfully predicts performance but also points toward even better surface treatments not yet tested on a large scale. The results <u>appear</u> in *Physical Review Accelerators and Beams*.

Niobium surface preparation

The backbones of essentially all advanced particle accelerators are structures called radiofrequency cavities, which are typically made of the metal niobium. When supercooled to temperatures near absolute zero, niobium cavities become superconducting. This technology is the only way to construct energy-efficient, large-scale particle accelerators.

For decades, accelerator scientists believed that the best superconducting radiofrequency (SRF) cavities were made of the purest niobium with contaminant-free surfaces. Jefferson Lab's Continuous Electron Beam Accelerator Facility (CEBAF), for instance, is built with pure niobium cavities. CEBAF is an Office of Science user facility that serves as the



research home to more than 1,900 nuclear physicists worldwide.

In more recent years, though, DOE researchers discovered that a bit of contaminant— for example, nitrogen—baked onto the niobium's surface could improve a cavity's performance by making even less heat. This process of "nitrogen doping" was discovered at DOE's Fermi National Accelerator Laboratory (Fermilab). The process improves performance by diffusing a bit of nitrogen gas into the niobium material surface.

The performance with initial nitrogen doping treatments was so strong that it was chosen twice for upgrading the Linac Coherent Light Source (LCLS) X-ray laser at DOE's SLAC National Accelerator Laboratory in California. Fermilab led a multi-lab collaboration to quickly establish new standards for the material and processing methods used for such high-efficiency accelerators.

"These two projects both use nitrogen doping, but two different recipes. And it was observed that the distribution of the peak fields that the cavities could reach was different now between the two recipes. And so the question is why?" said Reece.

The two projects upgrading the LCLS are LCLS-II and LCLS-II-HE. The LCLS-II project was a multi-year, \$1.1 billion upgrade that added the first SRF components to the machine. This upgrade to SRF accelerator technology allows the laser to produce up to a million X-ray pulses per second, 8,000 times more than its predecessor. LCLS-II-HE is adding additional SRF components to double the energy of LCLS-II. Higher energies will allow the machine to produce shorter X-rays and access additional science.

Thanks to Jefferson Lab's participation in the two different upgrade projects for LCLS, the team had a wealth of information on the preparation techniques used, as well as the results of component



performance testing.

"There is a difference in ultimate peak accelerating gradient, depending on the nitrogen doping process," said Eric Lechner, Jefferson Lab staff scientist who led the testing effort. "We wanted to take a look at how the surface roughness is different between these processes and compare that to the performance measured in these cavities."

Investigating surface roughness

The study focused on the effects of sequential electropolishing on the nitrogen-doped niobium samples. After doping, the samples are electropolished to remove outer layers from the cavity surface. Electropolishing both removes surface contamination and smooths the cavity surface.

The team had already developed a method to produce standardized samples and subject them to a controlled electropolish. They had assembled a novel toolkit with which to measure and analyze surface topography to estimate its impact on performance. These tools include scanning electron microscopy, secondary ion mass spectrometry, atomic force microscopy, and electron backscatter diffraction.

In the nitrogen doping process, the niobium is exposed to nitrogen gas for two minutes at 800 degrees Celsius, and in some cases, further annealed or heat-treated in a vacuum at that same temperature. During the process, niobium nitrides form on the surface and must be chemically removed to recover good RF performance.

The team reproduced those processes on their controlled samples and then investigated the as-treated surfaces with their toolkit to see how the topography evolved throughout.



The team found that the differences were particularly visible at the niobium grain boundaries. These grain boundaries are formed as the niobium metal used to produce the cavities is made into ingots or sheets. The niobium is first melted, and as it cools, individual crystals of the metal form. The boundaries of these individual crystals are the grain boundaries that may be visible to the naked eye and through a microscope.

What they found in their samples was that in addition to the beneficial nitrogen gas introduced into the surface of the niobium during the doping process, large nitride compound crystals also formed and clumped together preferentially at some grain boundaries of the niobium during the annealing process.

"It's that gas within the niobium that does the good stuff. The nitride compound crystals on the surface are really bad news, so we have to remove them," Reece explained.

Those nitride crystals were removed during the electropolish but left behind deep triangular grooves that they had grown in. Such grooves effectively amplify the local magnetic field, limiting how "loud" the useful accelerating field can be turned up.

"So we suspect that this is due to a process called Ostwald ripening, where nitrides will tend to clump together during the annealing process, forming larger nitrides that are deeper. And then, during the electropolishing process, that deeper trough is preferentially attacked. So, you have a deeper and sharper groove. Deep and sharp are two surface roughness qualities that are bad for performance," Lechner clarified.

Too much electropolishing to remove the crystal nitrides and alleviate the grooving could also remove the beneficial nitrogen gas that actually



helped improve performance.

"Our topographic analysis agrees well with the trend of performance observed in the LCLS-II HE R&D project as well as the cavity production performance for LCLS-II and LCLS-II HE, which had different nitrogen doping processes," Lechner added.

The team highlighted that the niobium yielding the higher maximum field performance was smoother.

What's next?

But nitrogen isn't the only contaminant that shows promise in improving SRF performance.

R&D at Fermilab showed that heat-treating niobium cavities at ~300 °C using a unique heating apparatus yielded RF performance akin to nitrogen doping.

Building on these results, researchers at the High Energy Accelerator Research Organization--known as KEK--in Japan and China's Institute of High Energy Physics found that they were getting efficiencies similar to nitrogen doping with a much simpler process: They baked cavities at far lower temperatures in standard vacuum furnaces—about 300 to 400° Celsius, didn't add nitrogen gas, then just rinsed off the cavities and skipped the electropolish.

Jefferson Lab scientists and others were so intrigued by this premise that Reece launched an investigation into the process.

He, Ari Palczewski, Lechner, and Jonathan Angle, then a graduate student at Virginia Tech, suspected that oxygen was the main contaminant in the new method. Their research quantified this process



both experimentally and theoretically, confirming that oxygen was the additive. During baking, the niobium's native oxide dissolved and diffused oxygen atoms uniformly into its surface.

"So this is oxygen doping as opposed to nitrogen doping. It can be done with a much simpler process. And so that's one of the types of samples we addressed," said Reece.

Both nitrogen doping and oxygen doping improved efficiency almost identically, but because oxygen doping is much simpler and less costly, Lechner said it's considered the more attractive option for future SRF cavities.

"The topographic analysis suggests that higher peak fields should be attainable in the oxygen-doped cavities with a significantly simpler and cheaper process," Lechner said.

The lab is continuing to make good use of the analysis developed for this study, applying it to other materials of interest for SRF applications, Lechner said.

In the meantime, the team continues to move toward their goal of finetuning their toolkit and model of how different aspects of cavity surface preparation affect accelerator performance. In essence, they're looking for how to economically tailor the top 1-micron-thick surface layer of accelerator cavities to meet the performance requirements of future applications confidently.

"That's the key thing here—not just finding a recipe that happens to work, but understanding what's going on so that we're knowledgeable enough to be able to tailor it," Reece said. "To get a surface you know is going to be good—that's the golden goose. We need both less heat and higher fields, reliably."



More information: Eric M. Lechner et al, Topographic evolution of heat-treated Nb upon electropolishing for superconducting rf applications, *Physical Review Accelerators and Beams* (2023). DOI: 10.1103/PhysRevAccelBeams.26.103101

Provided by Thomas Jefferson National Accelerator Facility

Citation: Smoother surfaces make for better accelerators (2024, April 18) retrieved 22 May 2024 from <u>https://phys.org/news/2024-04-smoother-surfaces.html</u>

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