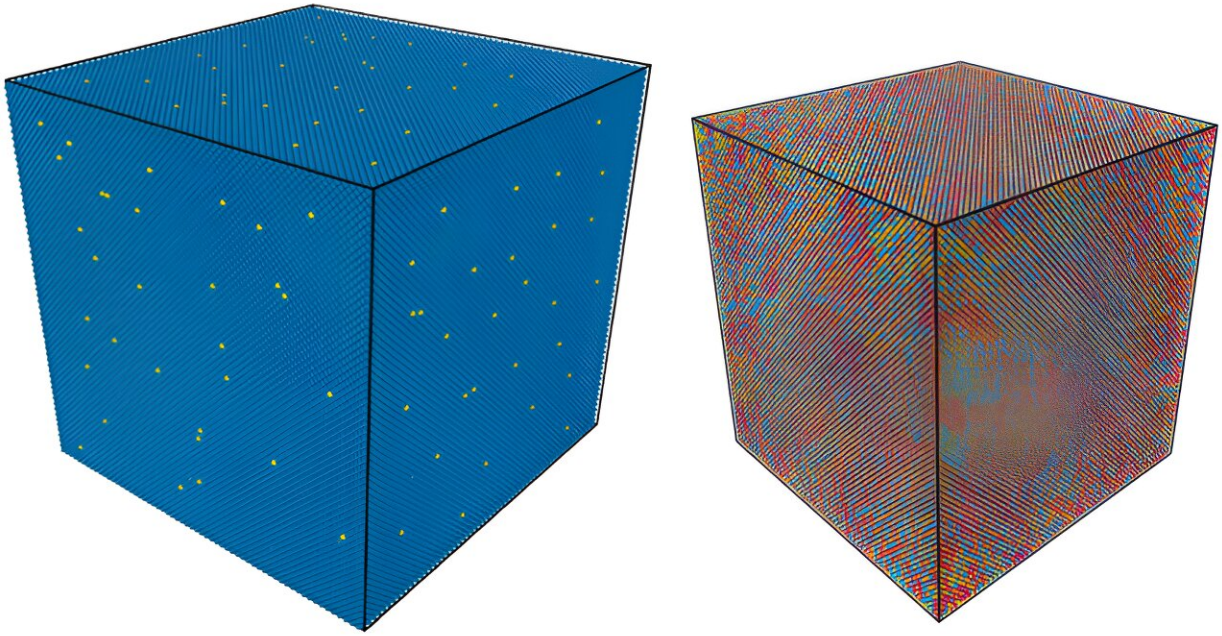
Machine learning unlocks secrets to advanced alloys

July 18 2024, by Poornima Apte



On the left, a traditional alloy with a main element in blue and a small amount of a different element in yellow. High-entropy alloys (as seen on the right) contain several elements in nearly equal amounts (three in this figure), creating many possibilities for chemical patterns. "It's like you're making a recipe with a lot more ingredients," says Yifan Cao, on of the authors of the paper, but it also adds significant chemical complexity. Credit: Massachusetts Institute of Technology

The concept of short-range order (SRO)—the arrangement of atoms over small distances—in metallic alloys has been underexplored in materials science and engineering. But the past decade has seen renewed interest in quantifying it, since decoding SRO is a crucial step toward developing tailored high-performing alloys, such as stronger or heat-resistant materials.

Understanding how atoms arrange themselves is no easy task and must be verified using intensive lab experiments or computer simulations based on imperfect models. These hurdles have made it difficult to fully explore SRO in metallic alloys.

But Killian Sheriff and Yifan Cao, graduate students in MIT's Department of Materials Science and Engineering (DMSE), are using machine learning to quantify, atom by atom, the complex chemical arrangements that make up SRO. Under the supervision of Assistant Professor Rodrigo Freitas, and with the help of Assistant Professor Tess Smidt in the Department of Electrical Engineering and Computer Science, their work was recently published in *Proceedings of the National Academy of Sciences*.

Interest in understanding SRO is linked to the excitement around advanced materials called high-entropy alloys, whose complex compositions give them superior properties.

Typically, materials scientists develop alloys by using one element as a base and adding small quantities of other elements to enhance specific properties. The addition of chromium to nickel, for example, makes the resulting metal more resistant to corrosion.

Unlike most traditional alloys, high-entropy alloys have several elements, from three up to 20, in nearly equal proportions. This offers a vast design space. "It's like you're making a recipe with a lot more

ingredients," says Cao.

The goal is to use SRO as a "knob" to tailor material properties by mixing chemical elements in high-entropy alloys in unique ways. This approach has potential applications in industries such as aerospace, biomedicine, and electronics, driving the need to explore permutations and combinations of elements, Cao says.

Capturing short-range order

Short-range order refers to the tendency of atoms to form chemical arrangements with specific neighboring atoms. While a superficial look at an alloy's elemental distribution might indicate that its constituent elements are randomly arranged, it is often not so.

"Atoms have a preference for having specific neighboring atoms arranged in particular patterns," Freitas says. "How often these patterns arise and how they are distributed in space is what defines SRO."

Understanding SRO unlocks the keys to the kingdom of high-entropy materials. Unfortunately, not much is known about SRO in high-entropy alloys. "It's like we're trying to build a huge Lego model without knowing what's the smallest piece of Lego that you can have," says Sheriff.

Traditional methods for understanding SRO involve small computational models, or simulations with a limited number of atoms, providing an incomplete picture of complex material systems.

"High-entropy materials are chemically complex—you can't simulate them well with just a few atoms; you really need to go a few length scales above that to capture the material accurately," Sheriff says. "Otherwise, it's like trying to understand your family tree without

knowing one of the parents."

SRO has also been calculated by using basic mathematics, counting immediate neighbors for a few atoms and computing what that distribution might look like on average. Despite its popularity, the approach has limitations, as it offers an incomplete picture of SRO.

Fortunately, researchers are leveraging machine learning to overcome the shortcomings of traditional approaches for capturing and quantifying SRO.

Hyunseok Oh, assistant professor in the Department of Materials Science and Engineering at the University of Wisconsin at Madison and a former DMSE postdoc, is excited about investigating SRO more fully. Oh, who was not involved in this study, explores how to leverage alloy composition, processing methods, and their relationship to SRO to design better alloys.

"The physics of alloys and the atomistic origin of their properties depend on short-range ordering, but the accurate calculation of short-range ordering has been almost impossible," says Oh.

A two-pronged machine learning solution

To study SRO using machine learning, it helps to picture the crystal structure in high-entropy alloys as a connect-the-dots game in an coloring book, Cao says.

"You need to know the rules for connecting the dots to see the pattern." And you need to capture the atomic interactions with a simulation that is big enough to fit the entire pattern.

First, understanding the rules meant reproducing the chemical bonds in

high-entropy alloys. "There are small energy differences in chemical patterns that lead to differences in short-range order, and we didn't have a good model to do that," Freitas says. The model the team developed is the first building block in accurately quantifying SRO.

The second part of the challenge, ensuring that researchers get the whole picture, was more complex. High-entropy alloys can exhibit billions of chemical "motifs," combinations of arrangements of atoms. Identifying these motifs from simulation data is difficult because they can appear in symmetrically equivalent forms—rotated, mirrored, or inverted. At first glance, they may look different but still contain the same chemical bonds.

The team solved this problem by employing 3D Euclidean neural networks. These advanced computational models allowed the researchers to identify chemical motifs from simulations of high-entropy materials with unprecedented detail, examining them atom by atom.

The final task was to quantify the SRO. Freitas used machine learning to evaluate the different chemical motifs and tag each with a number. When researchers want to quantify the SRO for a new material, they run it by the model, which sorts it in its database and spits out an answer.

The team also invested additional effort in making their motif identification framework more accessible. "We have this sheet of all possible permutations of [SRO] already set up, and we know what number each of them got through this machine learning process," Freitas says. "So later, as we run into simulations, we can sort them out to tell us what that new SRO will look like." The neural network easily recognizes symmetry operations and tags equivalent structures with the same number.

"If you had to compile all the symmetries yourself, it's a lot of work.

Machine learning organized this for us really quickly and in a way that was cheap enough that we could apply it in practice," Freitas says.

Enter the world's fastest supercomputer

This summer, Cao and Sheriff and team will have a chance to explore how SRO can change under routine metal processing conditions, like casting and cold-rolling, through the U.S. Department of Energy's INCITE program, which allows access to Frontier, the world's fastest supercomputer.

"If you want to know how short-range order changes during the actual manufacturing of metals, you need to have a very good model and a very large simulation," Freitas says. The team already has a strong model; it will now leverage INCITE's computing facilities for the robust simulations required.

"With that we expect to uncover the sort of mechanisms that metallurgists could employ to engineer alloys with pre-determined SRO," Freitas adds.

Sheriff is excited about the research's many promises. One is the 3D information that can be obtained about chemical SRO. Whereas traditional transmission electron microscopes and other methods are limited to two-dimensional data, physical simulations can fill in the dots and give full access to 3D information, Sheriff says.

"We have introduced a framework to start talking about chemical complexity," Sheriff explains. "Now that we can understand this, there's a whole body of materials science on classical alloys to develop predictive tools for high-entropy materials."

That could lead to the purposeful design of new classes of materials

instead of simply shooting in the dark.

More information: Killian Sheriff et al, Quantifying chemical short-range order in metallic alloys, *Proceedings of the National Academy of Sciences* (2024). DOI: 10.1073/pnas.2322962121

This story is republished courtesy of MIT News (web.mit.edu/newsoffice/), a popular site that covers news about MIT research, innovation and teaching.

Provided by Massachusetts Institute of Technology