# Engineers develop a way to determine how the surfaces of materials behave

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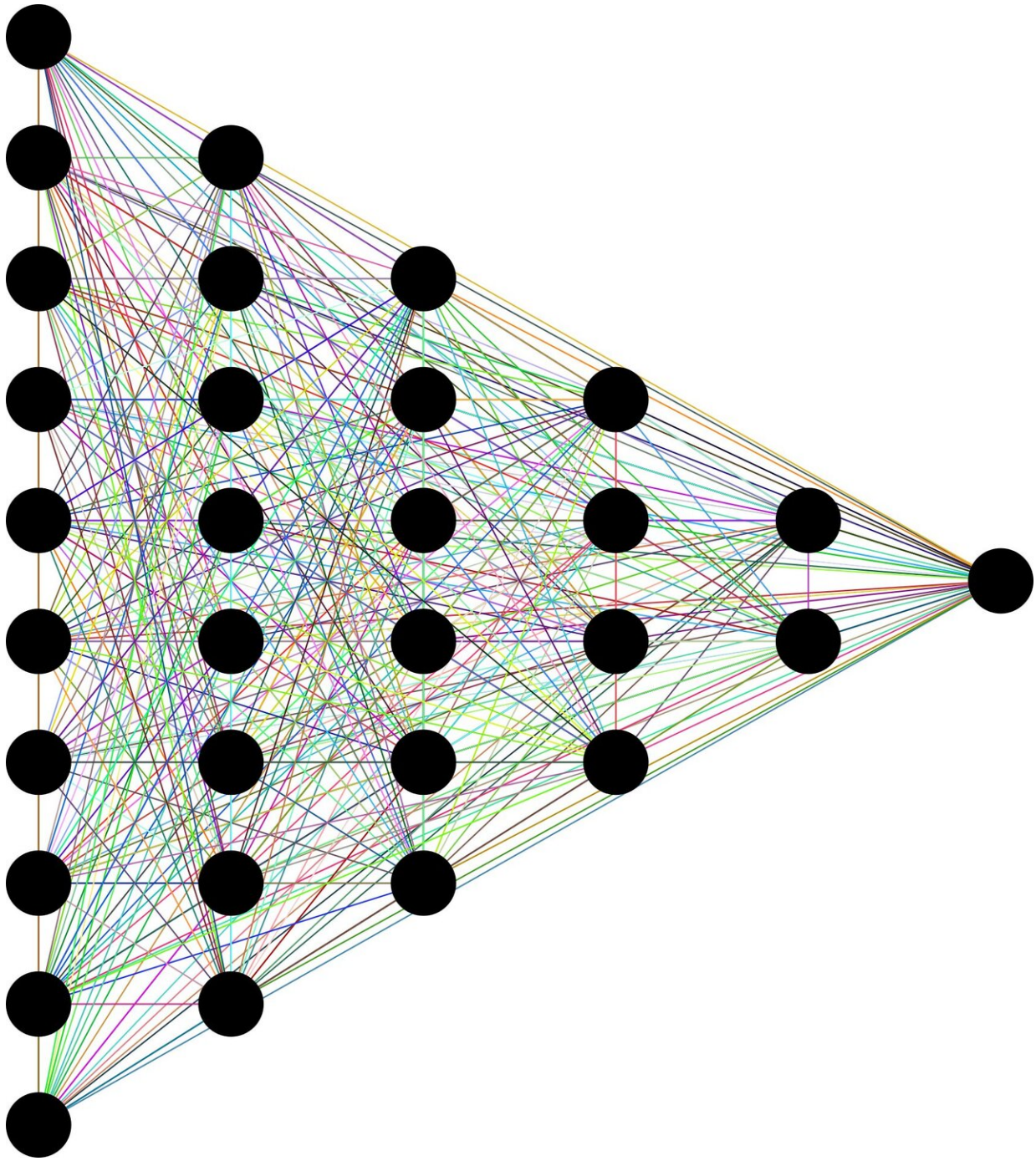

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Designing new compounds or alloys whose surfaces can be used as

catalysts in chemical reactions can be a complex process relying heavily on the intuition of experienced chemists. A team of researchers at MIT has devised a new approach using machine learning, that removes the need for intuition and provides more detailed information than conventional methods can practically achieve.

For example, applying the new system to a material that has already been studied for 30 years by conventional means, the team found the compound's surface could form two new atomic configurations that had not previously been identified, and that one other configuration seen in previous works is likely unstable.

The findings are described in the journal *Nature Computational Science*, in a paper by MIT graduate student Xiaochen Du, professors Rafael Gómez-Bombarelli and Bilge Yildiz, MIT Lincoln Laboratory technical staff member Lin Li, and three others.

Surfaces of materials often interact with their surroundings in ways that depend on the exact configuration of atoms at the surface, which can differ depending on which parts of the material's atomic structure are exposed. Think of a layer cake with raisins and nuts in it: Depending on exactly how you cut the cake, different amounts and arrangements of the layers and fruits will be exposed on the edge of your slice.

The environment matters as well. The cake's surface will look different if it is soaked in syrup, making it moist and sticky, or if it is put in the oven, crisping and darkening the surface. This is akin to how materials' surfaces respond when immersed in a liquid or exposed to varying temperatures.

Methods usually used to characterize material surfaces are static, looking at a particular configuration out of the millions of possibilities. The new method allows an estimate of all the variations, based on just a few first-

principles calculations automatically chosen by an iterative machine-learning process, in order to find those materials with the desired properties.

In addition, unlike typical present methods, the new system can be extended to provide dynamic information about how the surface properties change over time under operating conditions, for example while a catalyst is actively promoting a chemical reaction, or while a battery electrode is charging or discharging.

The researchers' method, which they call an Automatic Surface Reconstruction framework, avoids the need to use hand-picked examples of surfaces to train the neural network used in the simulation. Instead, it starts with a single example of a pristine cut surface, then uses active learning combined with a type of Monte-Carlo algorithm to select sites to sample on that surface, evaluating the results of each example site to guide the selection of the next sites.

Using fewer than 5,000 first-principles calculations, out of the millions of possible chemical compositions and configurations, the system can obtain accurate predictions of the surface energies across various chemical or electrical potentials, the team reports.

"We are looking at thermodynamics," Du says, "which means that, under different kinds of external conditions such as pressure, temperature, and chemical potential, which can be related to the concentration of a certain element, [we can investigate] what is the most stable structure for the surface?"

In principle, determining the thermodynamic properties of a material's surface requires knowing the surface energies across a specific single atomic arrangement and then determining those energies millions of times to encompass all the possible variations and to capture the

dynamics of the processes taking place. While it is possible in theory to do this computationally, "it's just not affordable" at a typical laboratory scale, Gómez-Bombarelli says.

Researchers have been able to get good results by examining just a few specific cases, but this isn't enough cases to provide a true statistical picture of the dynamic properties involved, he says.

Using their method, Du says, "We have new features that allow us to sample the thermodynamics of different compositions and configurations. We also show that we are able to achieve these at a lower cost, with fewer expensive quantum mechanical energy evaluations. And we are also able to do this for harder materials," including three-component materials.

"What is traditionally done in the field," he says, "is researchers, based on their intuition and knowledge, will test only a few guess surfaces. But we do comprehensive sampling, and it's done automatically." He says that "we've transformed a process that was once impossible or extremely challenging due to the need for human intuition. Now, we require minimal human input. We simply provide the pristine surface, and our tool handles the rest."

That tool, or set of computer algorithms, called AutoSurfRecon, has been made freely available by the researchers so it can be downloaded and used by any researchers in the world to help, for example, in developing new materials for catalysts, such as for the production of "green" hydrogen as an alternative emissions-free fuel, or for new battery or fuel cell components.

For example, Gómez-Bombarelli says, in developing catalysts for hydrogen production, "part of the problem is that it's not really understood how their surface is different from their bulk as the catalytic

cycle occurs. So, there's this disconnect between what the material looks like when it's being used and what it looks like when it's being prepared before it gets put into action."

He adds that "at the end of the day, in catalysis, the entity responsible for the catalyst doing something is a few atoms exposed on the surface, so it really matters a lot what exactly the surface looks like at the moment."

Another potential application is in studying the dynamics of chemical reactions used to remove carbon dioxide from the air or from power plant emissions. These reactions often work by using a material that acts as a kind of sponge for absorbing oxygen, so it strips oxygen atoms from the carbon dioxide molecules, leaving behind carbon monoxide, which can be a useful fuel or chemical feedstock. Developing such materials "requires understanding of what the surface does with the oxygens, and how it's structured," Gómez-Bombarelli says.

Using their tool, the researchers studied the surface atomic arrangement of the perovskite material strontium titanium oxide, or $SrTiO_3$, which had already been analyzed by others using conventional methods for more than three decades yet was still not fully understood. They discovered two new arrangements of the atoms at its surface that had not been previously reported, and they predict that one arrangement that had been reported is in fact unlikely to occur at all.

"This highlights that the method works without intuitions," Gómez-Bombarelli says. "And that's good because sometimes intuition is wrong, and what people have thought was the case turns out not to be." This new tool, he said, will allow researchers to be more exploratory, trying out a broader range of possibilities.

Now that their code has been released to the community at large, he says, "we hope that it will be inspiration for very quick improvements"

by other users.

The team included James Damewood, a Ph.D. student at MIT, Jaclyn Lunger Ph.D., who is now at Flagship Pioneering, and Reisel Millan, a former postdoc who is now with the Institute of Chemical Technology in Spain.

**More information:** Machine-learning-accelerated simulations to enable heuristic-free surface reconstruction, *Nature Computational Science* (2023). DOI: 10.1038/s43588-023-00571-7

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