Computational chemists design better ways of discovering and designing materials for energy applications

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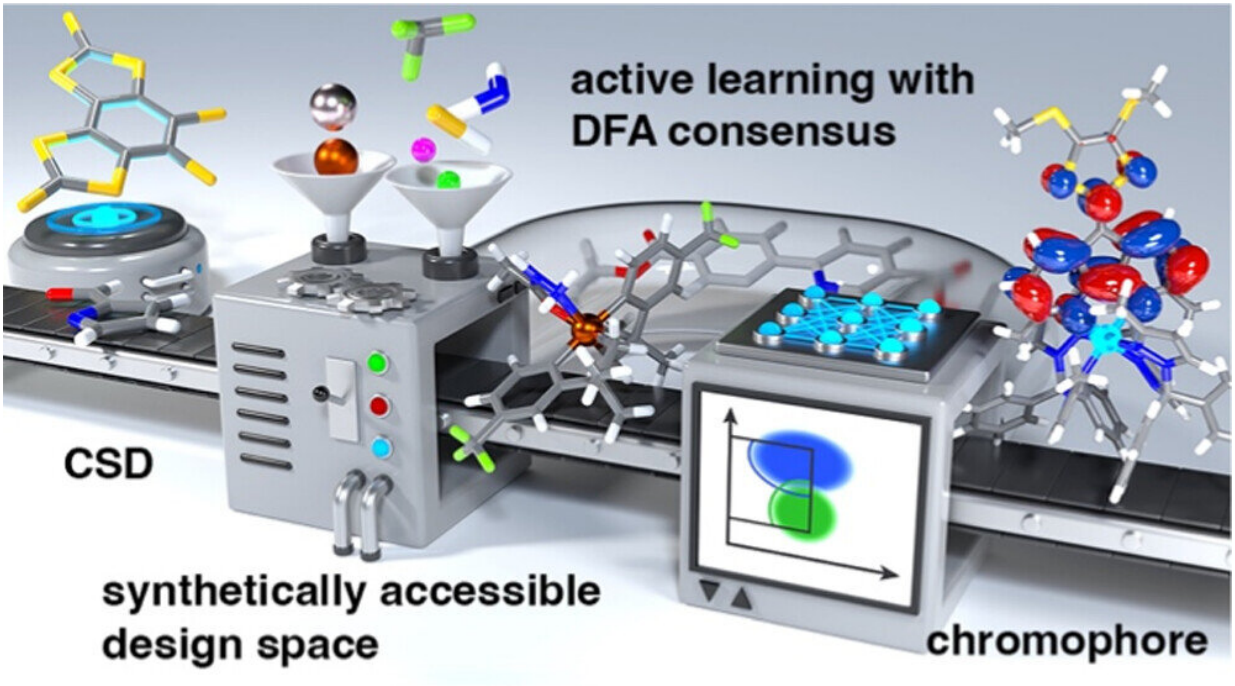

Graphical abstract. Credit: *JACS Au* (2022). DOI: 10.1021/jacsau.2c00547

Swift and significant gains against climate change require the creation of novel, environmentally benign, and energy-efficient materials. One of the richest veins researchers hope to tap in creating such useful compounds is a vast chemical space where molecular combinations that

offer remarkable optical, conductive, magnetic, and heat transfer properties await discovery.

But finding these new materials has been slow going.

"While computational modeling has enabled us to discover and predict properties of new materials much faster than experimentation, these models aren't always trustworthy," says Heather J. Kulik Ph.D. '09, associate professor in the departments of Chemical Engineering and Chemistry. "In order to accelerate computational discovery of materials, we need better methods for removing uncertainty and making our predictions more accurate."

A team from Kulik's lab set out to address these challenges with a team including Chenru Duan Ph.D. '22.

A tool for building trust

Kulik and her group focus on transition metal complexes, molecules comprised of metals found in the middle of the periodic table that are surrounded by organic ligands. These complexes can be extremely reactive, which gives them a central role in catalyzing natural and industrial processes. By altering the organic and metal components in these molecules, scientists can generate materials with properties that can improve such applications as artificial photosynthesis, solar energy absorption and storage, higher efficiency OLEDS (organic light emitting diodes), and device miniaturization.

"Characterizing these complexes and discovering new materials currently happens slowly, often driven by a researcher's intuition," says Kulik. "And the process involves trade-offs: You might find a material that has good light-emitting properties, but the metal at the center may be something like iridium, which is exceedingly rare and toxic."

Researchers attempting to identify nontoxic, earth-abundant transition metal complexes with useful properties tend to pursue a limited set of features, with only modest assurance that they are on the right track. "People continue to iterate on a particular ligand, and get stuck in local areas of opportunity, rather than conduct large-scale discovery," says Kulik.

To address these screening inefficiencies, Kulik's team developed a new approach—a machine-learning based "recommender" that lets researchers know the optimal model for pursuing their search. Their description of this tool was the subject of a paper in *Nature Computational Science* in December.

"This method outperforms all prior approaches and can tell people when to use methods and when they'll be trustworthy," says Kulik.

The team, led by Duan, began by investigating ways to improve the conventional screening approach, density functional theory (DFT), which is based on computational quantum mechanics. He built a machine learning platform to determine how accurate density functional models were in predicting structure and behavior of transition metal molecules.

"This tool learned which density functionals were the most reliable for specific material complexes," says Kulik. "We verified this by testing the tool against materials it had never encountered before, where it in fact chose the most accurate density functionals for predicting the material's property."

A critical breakthrough for the team was its decision to use the electron density—a fundamental quantum mechanical property of atoms—as a machine learning input. This unique identifier, as well as the use of a neural network model to carry out the mapping, creates a powerful and

efficient aide for researchers who want to determine whether they are using the appropriate density functional for characterizing their target transition metal complex. "A calculation that would take days or weeks, which makes computational screening nearly infeasible, can instead take only hours to produce a trustworthy result."

Kulik has incorporated this tool into molSimplify, an open source code on the lab's website, enabling researchers anywhere in the world to predict properties and model transition metal complexes.

Optimizing for multiple properties

In a related research thrust, which they showcased in a recent publication in *JACS Au*, Kulik's group demonstrated an approach for quickly homing in on transition metal complexes with specific properties in a large chemical space.

Their work springboarded off a 2021 paper showing that agreement about the properties of a target molecule among a group of different density functionals significantly reduced the uncertainty of a model's predictions.

Kulik's team exploited this insight by demonstrating, in a first, multi-objective optimization. In their study, they successfully identified molecules that were easy to synthesize, featuring significant light-absorbing properties, using earth-abundant metals. They searched 32 million candidate materials, one of the largest spaces ever searched for this application. "We took apart complexes that are already in known, experimentally synthesized materials, and we recombined them in new ways, which allowed us to maintain some synthetic realism," says Kulik.

After collecting DFT results on 100 compounds in this giant chemical domain, the group trained machine learning models to make predictions

on the entire 32 million-compound space, with an eye to achieving their specific design goals. They repeated this process generation after generation to winnow out compounds with the explicit properties they wanted.

"In the end we found nine of the most promising compounds, and discovered that the specific compounds we picked through machine learning contained pieces (ligands) that had been experimentally synthesized for other applications requiring optical properties, ones with favorable light absorption spectra," says Kulik.

Applications with impact

While Kulik's overarching goal involves overcoming limitations in computational modeling, her lab is taking full advantage of its own tools to streamline the discovery and design of new, potentially impactful materials.

In one notable example, "We are actively working on the optimization of metal–organic frameworks for the direct conversion of methane to methanol," says Kulik. "This is a holy grail reaction that folks have wanted to catalyze for decades, but have been unable to do efficiently."

The possibility of a fast path for transforming a very potent greenhouse gas into a liquid that is easily transported and could be used as a fuel or a value-added chemical holds great appeal for Kulik. "It represents one of those needle-in-a-haystack challenges that multi-objective optimization and screening of millions of candidate catalysts is well-positioned to solve, an outstanding challenge that's been around for so long."

More information: Chenru Duan et al, A transferable recommender approach for selecting the best density functional approximations in chemical discovery, *Nature Computational Science* (2022). DOI:

10.1038/s43588-022-00384-0

Chenru Duan et al, Active Learning Exploration of Transition-Metal Complexes to Discover Method-Insensitive and Synthetically Accessible Chromophores, *JACS Au* (2022). DOI: 10.1021/jacsau.2c00547

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