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Hybrid catalyst produces critical fertilizer and cleans wastewater

September 11 2023, by Win Reynolds

Producing the fertilizer urea using electrified synthesis could both denitrify wastewater while enabling low-carbon-intensity urea production. Credit: Etienne Girardet

Agriculture relies on synthetic nitrogen fertilizer, which is made using energy- and carbon-intensive processes and creates nitrate-containing runoff. Researchers have long sought solutions to reduce emissions from the industry that accounts for 3% of energy consumption each year.

A collaboration between two labs at Northwestern University, partnering with the University of Toronto, has found that producing the fertilizer urea using electrified synthesis could both denitrify wastewater while enabling low-carbon-intensity urea production. The process, which includes converting carbon dioxide and waste nitrogen by using a hybrid catalyst made of zinc and copper, could benefit water treatment facilities by reducing their carbon footprint and supplying a potential revenue stream.

The findings are published today in the journal *Nature Catalysis.*

"It's estimated that synthetic nitrogen fertilizer supports half of the global population," said Northwestern professor Ted Sargent, a corresponding author on the paper. "A chief priority of decarbonization efforts is to increase quality of life on Earth, while simultaneously decreasing society's net CO_2 intensity. Figuring out how to use renewable electricity to power chemical processes is a big opportunity on this score."

Sargent is the co-executive director of the Paula M. Trienens Institute for Sustainability and Energy (formerly ISEN) and a multidisciplinary researcher in materials chemistry and energy systems, with appointments in the department of chemistry in the Weinberg College of Arts and Sciences and the department of electrical and computer engineering in the McCormick School of Engineering.

In Sargent's field, many researchers have developed alternate routes to make ammonia, a precursor to many fertilizers, but few have looked at urea, which is a shippable, ready-to-use fertilizer. It represents a $100 billion industry. The team said the research stemmed from asking the question "Can we use waste nitrogen sources, captured CO_2, and electricity to create urea?"

Looking back to move forward

Yuting Luo, the paper's first author, a post-doctoral fellow in the Sargent Group and a Banting Postdoctoral Researcher, said a deep dive into historical references helped identify what would become their "magic" hybrid catalyst. Typically, chemists use alloys or more complicated materials to trigger reactions, limiting them to favor a single reaction step at a time. "It's quite uncommon to put two catalysts together that cooperate in a relay mode," Luo said. "The catalyst is the real magic here."

The team saw references dating back to the 1970s that implied pure metals—like zinc and copper—can be useful in processes involving carbon dioxide and nitrogen conversion.

These preliminary experiments, which the Sargent lab went on to replicate, converted relatively little of the initial ingredients into the desired product (the team found about a 20-30% conversion efficiency to urea).

Renewable energy sources tip the scales

Creating change within industries requires careful cost-benefit analyses that definitively prove a new production route will ultimately pay off in both energy and cost savings. That's where chemical engineering professor Jennifer Dunn's research came in. Chayse Lavallais, a fourth-year Ph.D. student in the Dunn lab, helped the team conduct a thorough life-cycle analysis, carefully including each energy input and output in a variety of scenarios.

"Using an average U.S. grid, the energy emissions are about the same," Lavallais said. "But when you go to renewable sources, several factors lower energy emissions, including CO_2 sequestration and carbon credits stored in end-use polymers. In a water treatment facility, if it adds emissions or energy, they're not encouraged to use the technology. We saw this doesn't impact the daily operational costs significantly, and there's potential to sell the product."

They found the conversion efficiency would need to reach 70% to be practical.

Perfecting the 'magic catalyst' ratio

The researchers ultimately reached their target starting with a simple mistake. Their hypothesis was solid—a layer of zinc on copper would result in better performance. But initially, they weren't finding that at all because they were applying the layer of zinc too thick and using a one-to-one ratio of zinc to copper, resulting in the material behaving as if it was only interacting with zinc. At one point, someone added less binder than was typical to the mix and some zinc washed away, and the experiment worked very well. The team then tuned the metals accordingly and determined a ratio of one part zinc to 20 parts copper resulted in optimal performance.

The Sargent group also applied a computational lens to uncover why copper and zinc worked so well together, and why it seemed there needed to be synergy between the two reactions. Because it's impossible to capture these reactions visually—they happen at the scale of nanoseconds—one must calculate them and determine how electrons move across a reaction.

This process had two distinct sections. First, the carbon must interact with zinc, as a reaction with copper produces a weak reaction. In the second stage, the opposite is true—nitrogen and copper create an efficient reaction, while zinc does very little.

There's a way to go before the process can be commercialized, the researchers said. Primarily, the reaction as it stands does not account for impurities found in a water treatment context. They also hope to increase the amount of time their process can operate.

More information: Yuting Luo et al, Selective electrochemical synthesis of urea from nitrate and CO2 via relay catalysis on hybrid catalysts, *Nature Catalysis* (2023). DOI: 10.1038/s41929-023-01020-4

Provided by Northwestern University

Citation: Hybrid catalyst produces critical fertilizer and cleans wastewater (2023, September 11) retrieved 28 April 2024 from https://phys.org/news/2023-09-hybrid-catalyst-critical-fertilizer-wastewater.html