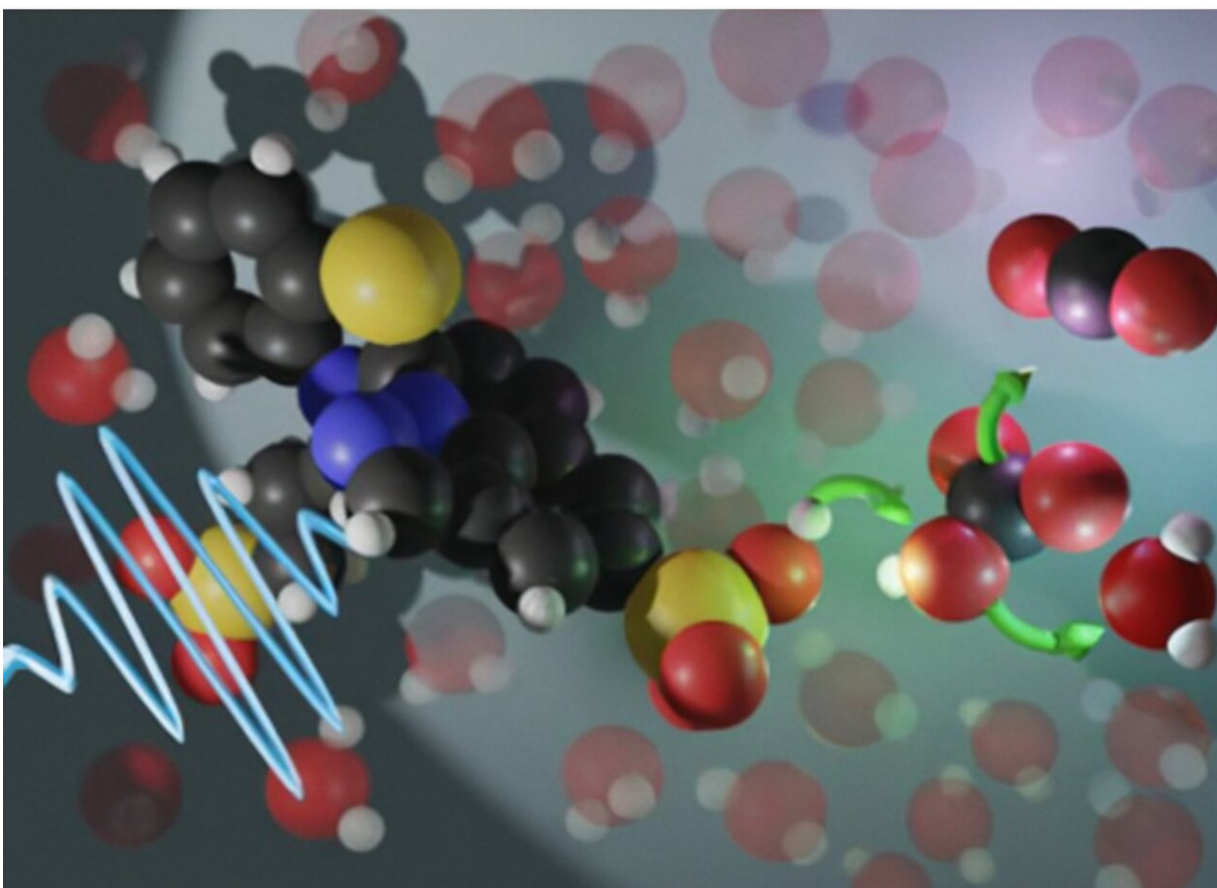


Light-activated acid drives energy-efficient, on-demand release of captured carbon dioxide

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On-demand CO₂ release under ambient conditions using light instead of heat is achieved for simulated and amino acid-based direct air capture systems by regulating solution pHs via a photoinduced proton transfer of a reversible metastable-state photoacid, leading to energetically sustainable and economically feasible climate change mitigation solutions using solar energy. Credit:

Using light instead of heat, researchers at the Department of Energy's Oak Ridge National Laboratory have found a new way to release carbon dioxide (CO₂) from a solvent used in direct air capture, or DAC, to trap this greenhouse gas. The novel approach paves the way for economically viable separation of CO₂ from the atmosphere.

The on-demand release of carbon dioxide is possible because the long-lived excited state of a novel acid controls the solution's proton concentration using [ultraviolet light](#), creating conditions that lead to CO₂'s energy-efficient release. By contrast, current DAC technologies filter air through an [aqueous solution](#) containing a sorbent material, such as an amino acid, that takes up atmospheric CO₂ and holds it. Heating the solvent releases the CO₂ and regenerates the amino acid for recycling. The CO₂ can be either stored or converted into value-added products, such as ethanol, polymers or concrete.

"In the existing direct-air-capture technologies, CO₂ release and sorbent regeneration are the most energy-intensive steps," said ORNL chemist Yingzhong Ma, who led the [study published in *Angewandte Chemie International Edition*](#) with ORNL colleagues Radu Custelcean and Uvinduni Premadasa, both chemists. "The goal here is to use the amino acid sorbent, which is recyclable and has a lot of attractive properties, combined with a more energy-efficient approach to release the CO₂ and regenerate the sorbent."

The National Academy of Sciences has concluded that DAC technologies have a role in removing billions of tons of CO₂ from the atmosphere annually to help limit the rise in average global temperature to less than 2 degrees Celsius (about 4 degrees Fahrenheit). However, the

intensive energy cost associated with sorbent regeneration and CO₂ release at a scale that would mitigate [climate change](#) makes such a massive deployment a [grand challenge](#) necessitating the development of new DAC processes. The ORNL-led approach provided a proof of concept for using irradiation with ultraviolet light under ambient conditions instead of heating the solution to release the CO₂ and regenerate the sorbent.

"Heating aqueous solutions is a common regeneration method, but it is extremely energy intensive," said Custelcean, a pioneer in DAC. "We wanted to take heat out of the equation."

Custelcean led [a study in 2017](#) that proved a guanidine sorbent could directly capture CO₂ from air. In 2018, he and colleagues demonstrated a practical, energy-efficient DAC method using solar heat to drive the release of the greenhouse gas from an amino-acid sorbent. This year, Knoxville-based startup Holocene licensed the technology to prepare it for industrial deployment.

In this new development, the key to releasing CO₂ at ambient conditions is a photoacid, which is a molecule that becomes more acidic when it absorbs light. Shine a light on an acid such as vinegar and nothing happens. By contrast, expose a photoacid to ultraviolet or visible light, and a chemical group in the middle of the acid rotates from the opposite side of a bond to the same side. A subsequent reaction forms a ring, leading to transfer of a proton, or hydrogen ion, to the water solvent.

This transfer dramatically increases the acidity of the solution, producing a change called a "pH swing." The excess protons can now interact with bicarbonate, or HCO₃⁻, which was made when CO₂ reacted with the sorbent. The bicarbonate accepts a proton to become [carbonic acid](#), or H₂CO₃, which is just one energetically favorable step away from carbon dioxide and water.

"This paper describes the first time where the macroscopic pH swing lasting from minutes to hours has been demonstrated using light as an external trigger to initiate the CO₂ regeneration reaction," said Vyacheslav "Slava" Bryantsev, leader of ORNL's Chemical Separations group and a co-author of the paper.

"You can easily turn light on and off to control the reaction reversibly," Ma said. "You can capture CO₂ in the dark and then simply turn on the light when you want to release CO₂ for storage or for making value-added products. It gives you a way to easily control the process on demand."

That said, the researchers needed an additional trick of the light. Conventional photoacids would not work because the lifetimes of their excited states are very short—mere nanoseconds. They lose protons but then stay mostly in the same configuration. "Then you only change the acidity for a short time," Bryantsev said.

Ma and Custelcean, who conceived the idea of using a photoacid to trigger CO₂ release in DAC applications, ran into this problem when they began experiments using a commercially available photoacid.

"When carbonic acid decomposes, it has a short lifetime in water, on the order of a few seconds. But that's an infinity compared to the lifetime of a regular photoacid, which is nanoseconds, or billionths of seconds," Custelcean said. "That's why you cannot do this chemistry with a regular photoacid: It takes seconds to release CO₂ from carbonic acid, but it takes only nanoseconds for the photoacid to take the proton back."

Bryantsev came up with the idea to try a different class of photoacid with a long-lived excited state. Called a metastable-state photoacid, it has a structure that persists in solution from seconds to hours. That means the pH change driven by the photoacid's structural change also lasts a lot

longer.

The scientists invited an expert in photoacid design and synthesis to join the team. Florida Institute of Technology's Yi Liao had pioneered the new class of metastable-state photoacids around 2015 but for purposes other than DAC.

"We really made a breakthrough after we got this photoacid from our collaborator," Ma said.

Custelcean agreed. "Having a metastable-state photoacid gave us plenty of time to release the proton and form the carbonic acid. Then the carbonic acid had time to release the CO₂ in water. Once that happens, CO₂ leaves the solution," he said.

With Ma, first author Premadasa designed and conducted the experiments for the proof-of-concept study using a metastable-state photoacid synthesized by Liao and Florida Tech colleague Adnan Elgattar, with subsequent spectroscopic characterization by ORNL's Benjamin Doughty and Vera Bocharova.

"Once we baselined the photochemical properties of the acid itself, our next step was to test its applicability for CO₂ release with various DAC sorbents," Premadasa said. "We can easily manipulate chemical compositions and intensities and colors of light to drive the photoreaction for efficient CO₂ release."

Audrey Miles from the University of Notre Dame and Stella Belony from the University of Florida, who were DOE Science Undergraduate Laboratory Internships students at the time of the study, tested the photoacid under different conditions for its CO₂-releasing abilities. Then ORNL's Michelle Kidder, Diana Stamberg and Joshua Damron measured the amount of CO₂ released under those different conditions.

Many challenges remain to develop ORNL's light-activated DAC technology. One is understanding the dynamics by which the photoacid forms a chemical complex with the amino acid sorbent. Another is improving the solubility of compounds in water. Yet another is optimizing the absorption of light from the visible spectrum. Moreover, the scientists would like to decrease the time required to regenerate the photoacid and improve understanding of its long-term stability.

Regardless, the future is bright for metastable-state photoacids. "Our study paves the way towards photochemically driven approaches for CO₂ release and sorbent regeneration using solar light," Premadasa said.

The title of the paper is "Photochemically-Driven CO₂ Release Using a Metastable-State Photoacid for Energy Efficient Direct Air Capture."

More information: Uvinduni I. Premadasa et al, Photochemically-Driven CO₂ Release Using a Metastable-State Photoacid for Energy Efficient Direct Air Capture, *Angewandte Chemie International Edition* (2023). [DOI: 10.1002/anie.202304957](https://doi.org/10.1002/anie.202304957)

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