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Electrochemical CO₂ capture can finally compete with amine-based capture

David A. Vermaas^{1,*} and Ruud Kortlever^{2,*}

Electrochemical CO₂ capture is promising for closing the carbon cycle but needs technological advances. In a recent issue of *Nature Energy*, a novel chemistry for electrochemical CO₂ capture is presented, demonstrating low energy consumption and high purity with virtually no degradation. This finally allows competition with amine-based capture technology.

The need for achieving net zero emissions by 2050, coupled with the corresponding energy transition, presents enormous challenges for processes that currently depend on fossil fuels. Although many of these processes can be electrified, certain applications, such as plastics production or fuel for long-distance transport, will still require carbon-based inputs. Furthermore, anthropogenic greenhouse gas emissions that are not directly related to fossil fuels, such as agriculture and methane from waste streams, are hard to abate. This underlines the importance of CO₂ capture from the environment, both for producing emission-neutral carbon-based chemicals and for creating negative emissions. Current projections suggest that by

2050, gigatons of CO₂ will need to be processed,¹ a stark contrast to the kttons/year being captured from air or water today. This tremendous disparity requires a breakthrough in green CO₂ capture technology.

Traditional CO₂ capture technologies, such as amine absorption or calcium looping, rely on a temperature swing to regenerate the capture material. Such heat-driven capture technologies do not fit well in the transition to electrified processes. Electricity-based technologies have been developed, for example, using electrochemically mediated amine regeneration (EMAR), CO₂ capture in redox carriers, or pH swing via bipolar membrane electrodiagnosis.² However, these technologies

either consume too much energy (200–1000 kJ/mol CO₂) or do not produce sufficiently pure CO₂. As a reference, the thermodynamic limits for CO₂ capture are just 15–25 kJ/mol CO₂ (depending on the source), while the production of electricity involves emissions equivalent to 363 kJ/mol CO₂ based on the 2022 global average.³ Hence, we need (and can have) electrochemical CO₂ capture technology with a lower energy consumption.

In a recent paper published in *Nature Energy*, Pang et al.⁴ demonstrate an electrochemical approach for capturing CO₂ using a pH swing induced by proton-coupled electron transfer of a developed phenazine derivative (see Figure 1A). Upon reducing the developed 2,2'-(phenazine-1,8-diyl)bis(ethane-1-sulfonate), abbreviated as 1,8 ESP, hydroxide ions are

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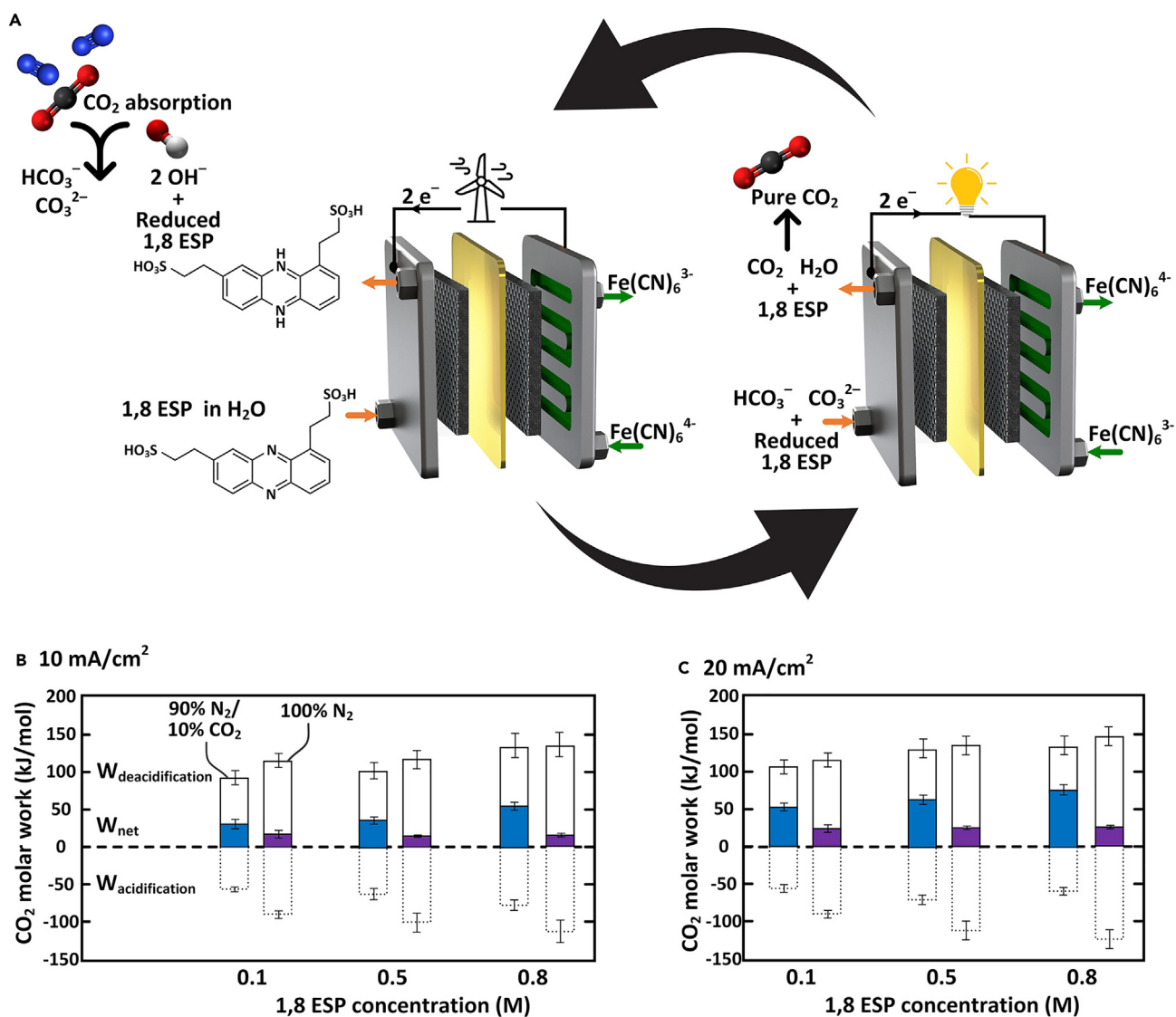


Figure 1. Highlights of simultaneous CO₂ capture and energy storage from the work of Peng et al.

(A) Concept of CO₂ capture, via reducing 1,8 ESP, which generates hydroxides, which can capture CO₂ from mixed gases. Pure CO₂ is released again when the reduced 1,8 ESP is oxidized, while part of the electrical energy is regenerated.

(B) Work per mol of CO₂ for deacidification, for acidification, and net work, at 10 mA/cm². Experiments were performed with N₂/CO₂ mixture and with pure N₂, in which the latter represents the irreversible energy losses. Error bars indicate standard deviations in measurements from five cycles.

(C) Same as (B), for 20 mA/cm²

Data from Pang et al.⁴

released, which allows capturing of CO₂ upon contact with gas. Oppositely, when oxidizing the reduced 1,8 ESP, the solution is acidified, and CO₂ is released again.

The work presents a leap forward in achieving low energy consumption per mol of CO₂ captured (36–52 kJ/mol CO₂, see Figure 1B and 1C).⁴ Because

the authors pair the reduction and oxidation of the 1,8 ESP with a ferrocyanide/ferricyanide counter-reaction, the energy input during the deacidification step is larger, but part of the energy is retrieved during the acidification step.

The 1,8 ESP displays a high solubility in an aqueous 1 M KCl electrolyte. As a

result, it boasts a high CO₂ capture capacity of 2.35 and 1.48 mol CO₂/l at partial pressures of 100 and 0.4 mbar, respectively, representing CO₂ capture from flue gas and from the atmosphere. At the same time, the captured CO₂ is released at a high purity (>99%).

A standout feature of the study, apart from the impressive performance

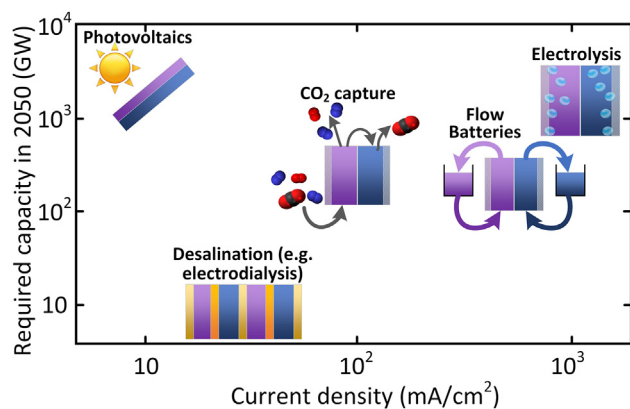


Figure 2. Estimated power capacity by 2050 for electrochemical technologies versus typical (future) current densities

Estimates for capacity are derived from Statista (photovoltaics), International Energy Agency (Electrolysis), European Association for Storage of Energy (Flow Batteries), and Gao et al.⁹ (desalination) and assuming CO₂ capture of 5 Gton/year¹ at 1,500 kWh/ton.

metrics of the developed system, is the attention the authors have paid to the stability of their system. At a temperature of 45°C, 1,8 ESP remained stable for more than 100 days. Moreover, the cycle stability of 1,8 ESP is emphasized by running over 550 charge-discharge cycles under a nitrogen atmosphere and 220 charge-discharge cycles under a CO₂ atmosphere with little degradation (approximately 0.01% per day). On the other hand, the system remains sensitive to oxygen with chemical oxidation of the redox species and hence a decrease in Coulombic efficiency with increasing oxygen concentrations. Although the developed system is less sensitive to oxygen than previously reported systems⁵ and does not fade over time, the use of an oxygen-sensitive redox species limits the applicability of the system.

To further advance this technology, the development of a redox species that is insensitive to oxygen would be an asset. Moreover, while the energy consumption is impressively low at 10 and 20 mA/cm², a current density in the range of 100 mA/cm² is more realistic for commercial devices. This could be addressed in future research by further developing the redox couple to achieve faster kinetics or implementing engi-

neering strategies from flow batteries and electrolysis designs, such as enhancing microscopic electrode area, electrode contacts, and membrane conductance. We also note that the CO₂ absorption step should be enhanced in future systems. In the present system, mimicking flue gas capture, the CO₂ absorption is the rate-limiting step, reflected in the long CO₂ absorption time (>10 h) during and after reducing the 1,8 ESP for 2.5 h. For direct air capture, this issue will be even more pressing. Although the work of Pang et al. provides some ideas to enhance the CO₂ absorption rate, the ratio in required volume of air to saturate a volume of this highly concentrated electrolyte (1.45 mol CO₂ per liter) is approximately 10⁵, which is a challenge for any commercial air contactor. One potential solution could be absorption units with an extremely thin liquid layer. Such a design would allow high air-to-liquid volume ratios and enhance mass transfer, making it an interesting avenue for future research.

Interestingly, the authors combine their CO₂ capture system with energy storage, using a ferrocyanide/ferricyanide counter-reaction. The reason behind this is 2-fold: combining these functions

into one device can substantially reduce the capital costs, and it offers flexibility in adjusting to varying electricity prices. However, the question is whether an integrated system could outperform a separate battery system and CO₂ capture system. Theoretically, separate units for CO₂ separation and energy storage, each at half the size of the combined unit of Pang et al., would have the same yield. For a separate CO₂ capture unit, CO₂ capture occurs when the phenazine-based solution is reduced at the cathode, while CO₂ release occurs simultaneously due to acidification at the anode. Such a symmetric system can operate continuously, even without switching when the fluids would be recirculated. Hence, the simultaneous reduction and oxidation of 1,8 ESP could operate at half the size, allowing installation of a separate battery unit for energy storage.

Combinations of different electrochemical functionalities in one cell have been explored across several fields, for example photoelectrochemical reactors,⁶ desalination batteries,⁷ and integrated CO₂ capture and conversion.⁸ However, such combinations often fail to scale up because requirements for the individual functionalities are incompatible. We identify two primary factors that are often disparate, being (1) current density and (2) market size. We have mapped these two parameters for individual technologies in Figure 2.

Figure 2 illustrates that a large disparity in typical current density is one of the reasons that photoelectrochemical reactors (combining photovoltaics and electrocatalysis) have not been taken up by industry, despite good technological advances in energy efficiency. To a certain extent, the same holds for the combination of desalination and energy storage, which is additionally strongly different in market size. This combination has found applications in niche areas, for example as capacitive deionization for selective ion removal,

but has not reached the scale at which reverse osmosis or electrodialysis operate, nor does it have impact on the global energy storage capacity.

Using the same metrics, combining CO₂ capture and energy storage appears more promising. The scales for CO₂ capture and energy storage are both projected to be around 500 GW in 2050. Although the current density for (flow) batteries is typically higher than that for electrochemical CO₂ capture, this gap may be bridgeable. Still, the work of Pang et al. shows that the 1,8 ESP electrolyte for CO₂ capture has a larger overpotential than the ferrocyanide counter-reaction, also reflected in the low current density (20 mA/cm²) compared to typical flow batteries.⁴

Even though the combination with energy storage is yet to be seen, the introduction of the phenazine-based proton-coupled electron transfer that can produce CO₂ with high purity and at an energy consumption below 50 kJ/mol is a significant step forward.

Future work should be directed to improving the current density and the oxygen sensitivity of the redox couples to allow capture from air or water. Recent work showed reliable performance of electrochemically cycling neutral red for CO₂ capture from air (20% O₂) at 65 kJ/mol.¹⁰ Those developments bring electrochemical CO₂ capture closer to practical application, which is required to close the carbon cycle by 2050.

DECLARATION OF INTERESTS

The authors declare no competing interests.

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