

Electrochemistry-Based CO₂ Removal Technologies

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Unprecedented increase in atmospheric CO₂ levels calls for efficient, sustainable, and cost-effective technologies for CO₂ removal, including both capture and conversion approaches. Current CO₂ abatement is largely based on energy-intensive thermal processes with a high degree of inflexibility. In this Perspective, it is argued that future CO₂ technologies will follow the general societal trend towards electrified systems. This transition is largely promoted by decreasing electricity prices, continuous expansion of renewable energy infrastructure, and breakthroughs in carbon electrotechnologies, such as electro-

chemically modulated amine regeneration, redox-active quinones and other species, and microbial electrosynthesis. In addition, new initiatives make electrochemical carbon capture an integrated part of Power-to-X applications, for example, by linking it to H₂ production. Selected electrochemical technologies crucial for a future sustainable society are reviewed. However, significant further development of these technologies within the next decade is needed, to meet the ambitious climate goals.

1. Introduction

Today, climate change owing to large amounts of anthropogenic CO₂ emissions is one of the most existential challenges this world is facing. It is the primary driver of increasing average surface temperatures and ocean acidification.^[1] Over the past decade, rates of global fossil CO₂ release increased by 1.2% per year and is dominated by increasing emissions in China, India, and other developing countries.^[2,3] The current total global anthropogenic CO₂ emission is in the order of 36 Gt each year.^[4] Unfortunately, there is no indication that the CO₂ emission rate will significantly decrease in the years to come; on the contrary, CO₂ levels are expected to rise due to increasing global energy consumption. A prediction from U.S. Energy Information Agency projects that worldwide energy consumption will increase

nearly 50% by 2050, from 657 EJ to 960 EJ, with the majority of this massive energy demand to be met by fossil fuels.^[5] It is estimated that 45% of fossil CO₂ emission results from the energy sector, dominated by electricity and heat production.^[3] Here thermal power plants are the dominant contributor while, at the same time, being the source of energy for electricity. Emissions per kWh electricity produced vary depending on the type of fuel: natural gas results in 400 g CO₂ kWh⁻¹, while coal generates 900 g CO₂ kWh⁻¹.^[6]

Therefore, the development of new technologies to reduce anthropogenic CO₂ emission and to ensure more sustainable energy technologies (Power-to-X) is becoming increasingly important. Current first-generation CO₂ capture relies on amine-based sorption with energy-intensive thermal cycles for desorption and are applied at large scale, currently removing 40 million tons per year (Mtpa) of CO₂ in total in commercial facilities.^[7] The Global CCS Institute estimates over 5,600 Mtpa CO₂ to be removed by carbon capture in 2050.^[8]

The thermal basis of this technology, however, is challenged by a growing focus on electrification of thermal processes in the energy sector. Conventional fossil-fuel combustion is being replaced by wind turbines and photovoltaics, which, in the USA, has accelerated the retirement of thermal power plants.^[9] Electrification in the car industry is replacing the internal combustion engine. This will undoubtedly make it necessary that the next generation of CO₂ capture technologies be based on electrochemical systems and be applicable in smaller scale industries. This transition to electricity has been largely accelerated by decreasing electricity prices. In Europe, the wholesale price of electricity has fallen steadily since their all-time high in 2008 for both the Nordpool market and for the German EEX-market.^[10] In the United States, the annual average prices declined from \$64 MWh⁻¹ to \$19 MWh⁻¹ between 2008 and 2017;^[9] the spike in wholesale electricity prices in 2021 and 2022 was caused by a surge in the costs of power generating fuels, for example, natural gas and general inflation. However, overall the U.S. Energy Information Agency expects average

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electricity prices to further decrease over the next 30 years due to expansion of the renewable energy sector as well as regulation-imposed emission penalties.^[11] Based on expectations of declining electricity prices and ongoing restructuring of the energy infrastructure it is anticipated that future CO₂ removal technologies will rely on electricity rather than on thermal energy from thermo-power plants. This switch will also provide a more flexible and sustainable energy infrastructure with the possibility of storing electrical energy as gas (methane) or upgrading it to fuels by, for example, electrochemical reduction of CO₂.

2. Current Carbon Capture Technologies

Since 1930 amines and organic solvents have been used by industry for scrubbing to separate CO₂ from natural gas, hydrogen, and other gases with low oxygen.^[12] The amine solution enters the absorber and reacts with gaseous CO₂, thereby, generating carbamate salts. Cleaned flue gas exits the absorber, while the amine solution, now enriched in CO₂, is taken to a desorber, where the solvent is regenerated at 100–125 °C. The aqueous amine solution is potentially reused in the absorber, while captured CO₂ can be further compressed (usually to 100–150 bar) for geological sequestration.^[12,13]

The main limitation to this widespread large-scale amine-based capture technology is the significant amount of thermal energy required to operate the desorption process. It is estimated that nearly 30% of the energy of a fossil fuel-based power plant would be consumed to operate CO₂ capture alone.^[14] The total energy demand of thermal CO₂ capture is 100–200 kJ mol^{−1} CO₂, which is about an order of magnitude higher than the minimum thermodynamic work.^[15]

Newer CO₂ capture technologies transition from thermal to electrical CO₂ capture systems and include pressure swing adsorption (PSA) (21–75 kJ mol^{−1} CO₂^[16,17]) and membrane applications where electricity is used for high pressure pumping and vacuum.^[18] It also includes cryogenic freezing out of CO₂ as dry ice in a heat pump approach, driven electrically by compressors^[19] (51–79 kJ mol^{−1} CO₂^[20]). In general, these electrical methods are to a large extent limited by mechanical and thermodynamic effects resulting from high pressure requirements. This compromises their energy efficiencies and represents a great disadvantage compared to electrochemical systems. The use of electrochemical approaches circumvents the conversion of electricity into mechanical energy inherent in PSA and membrane technologies.^[21] Improved efficiency of the capture process can be obtained by electrochemically based removal and possible subsequent conversion of CO₂.

3. Current Technologies for Electrochemical Desorption of CO₂

Electrochemical approaches for CO₂ capture have gained increasing attention because they have distinct advantages

over thermal operations. First and foremost, they do not require external high temperature energy sources, steam, high pressures, nor vacuum.^[21] Moreover, they can be readily integrated in production processes without the need for retrofitting, i.e., as plug-and-play.^[22] They can rely entirely on renewable resources and can be easily regulated as they are not beset by long thermal response times, etc.

Electrochemical technologies for CO₂ capture and release can broadly be classified into two categories: 1) processes relying on displacement reactions and 2) processes relying on redox-active solvents. In the first case the affinity between CO₂ and the capture solvent is modulated by a second electrochemically generated reagent. CO₂ and the solvent form a complex, which is disrupted by a more reactive agent displacing the CO₂ by having a stronger binding affinity to the solvent (Figure 1). The oxidation state of the reagent can be changed to regenerate the solvent. In the second case, the affinity of the capture agent for CO₂ is controlled by the oxidation state of the agent itself (Figure 1).

Examples of efficient displacement-based capture approaches (category 1) are the electrochemically modulated amine regeneration process (EMAR)^[23] and the “proton concentration process.”^[24]

To date, the EMAR process has been one of the most comprehensively studied electrochemical systems (Figure 2a). It is based on copper mediation of the binding of certain amines with CO₂ in a traditional absorption mode. CO₂ complexed with amines is displaced by cupric ions introduced through oxidation of copper electrodes in the anode compartment of an electrochemical cell. Following CO₂ desorption, the amines are regenerated when copper is removed by plating in the cathode chamber, and then re-used in the traditional absorber.^[25] Detailed technoeconomic analysis indicates that the EMAR process can have energetic demands similar to, or even lower than, those of traditional amine-based sorption systems (ca. 30 kJ mol^{−1} CO₂).^[22,26] The energy efficiency of this process can be enhanced by suitable lowering of the overpotentials needed to drive the reactions.^[27]

The proton concentration process (category 1) is inspired by current thermal approaches taking advantage of the pH sensitivity of the CO₂ hydration equilibrium. They rely on high pH solutions to capture CO₂ as potassium carbonate, with release of CO₂ at elevated temperatures. Instead of using heat, pH regulation can be achieved electrochemically in a process similar to the EMAR process. The notable difference is that instead of cupric ions, protons intercalated within the electrodes (e.g., MnO₂) are introduced upon application of an oxidizing potential to induce release of CO₂ from solution (Figure 2b). The solvent is recovered by re-intercalation in the cathode compartment of the cell.^[26]

The second category of electrochemical CO₂ capture technologies uses redox-active agents and has gained significant attention in the academic community, in particular for direct air capture. Specifically, quinones and other compounds, when reduced, can act as nucleophiles to bind CO₂ strongly, but loses this affinity when in an oxidized, neutral state (Figure 2c). One application involves polymeric quinone species

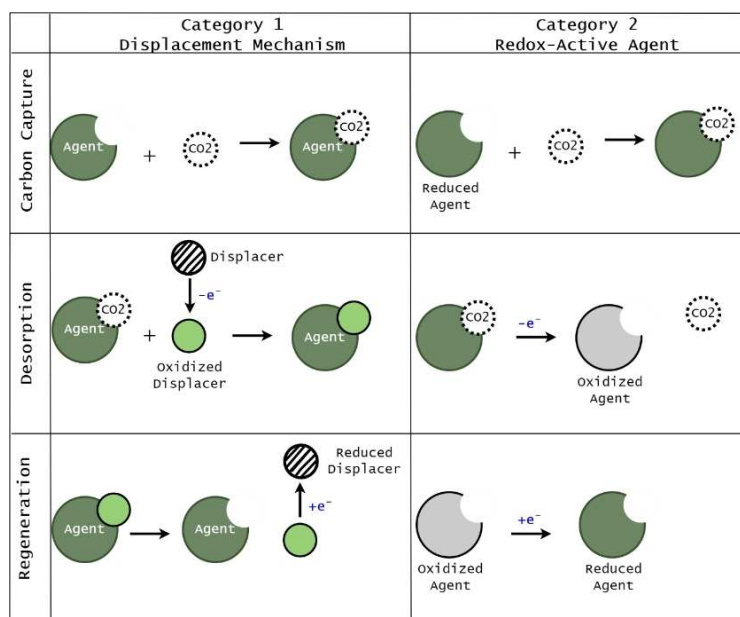


Figure 1. Electrochemical carbon capture and release. Category 1: the CO₂ cycle using an electrochemically mediated displacer. Category 2: process relying solely on the redox activity of the solvent.

immobilized on carbon nanotubes on an electrode, which is separated from a counter-electrode (usually ferrocene-based) by a spacer. Quinones bind CO₂ when a reducing current/potential is applied across the cell, and releases CO₂ on reversal of cell polarity.^[28] Estimates of the future energetic demands of this cyclic electro-swing approach are encouraging, and issues with long-term stability and oxygen sensitivity have been addressed effectively.^[29] Similar systems can be exploited in continuous flow applications, where the reduced quinones in solution are taken from the electrochemical cell and contacted with the air stream. Use of redox cycling for CO₂ capture is currently in early stages of commercial development.

Most large-scale electrochemical systems are designed to operate with aqueous electrolyte media, because they are cost-effective, safe, environmentally benign, and easy to implement. However, direct adduct formation of reduced quinones with CO₂^[29] does not occur to any significant extent as they are rapidly protonated to form hydroquinones.^[30] The consequent increase in solution pH promotes CO₂ absorption instead as bicarbonates or carbonates in solution. On reversal of the potential, oxidized quinones release protons, and the now acidified solution no longer has capacity for trapping CO₂, which is desorbed from solution.^[31]

Other electrochemically modulated systems for CO₂ capture and release also rely on the pH-swing concept, but here pH is modified through electrodialysis-type approaches. This requires water splitting, often making use of bipolar membranes to achieve the desired pH-regulation.^[32]

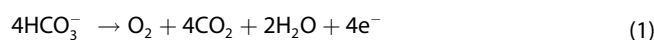
Electrochemical carbon capture solutions are expected to advance even further within the next decade, with many diverse technologies emerging. Future reduction of CO₂ will undoubtedly depend on a portfolio of diverse capture plat-

forms, each adapted to different sources of CO₂. The type of equipment needed may depend on whether the emission stream contains concentrated or diluted CO₂ and whether the stream is clean or contains chemical impurities.

4. Combining Carbon Capture and Power-to-X Processes

Eventually, CO₂ capture systems will need to be integrated within smaller industrial processes if they are to contribute in a meaningful way to a decrease in the total global release of CO₂. Unfortunately, CAPEX and OPEX costs associated with the introduction of CO₂ capture systems are often too high for small- to medium-sized CO₂ emitters. To meet this problem more financially viable solutions are being developed by coupling of sectors. As an example, CO₂ capture and H₂ production are being combined in electrochemical systems using a pH-swing concept. Such a proton-coupled faradaic process depends on the carbonate speciation equilibrium, allowing CO₂ capture at high pH (as HCO₃⁻ or CO₃²⁻) and CO₂ release at low pH. Here, CO₂ and O₂ are desorbed in a ratio of 4:1 at the anode [Equations (1), (2), and (3)], whereas H₂ is released from the cathode chamber [Equation (4)] along with solvent regeneration.^[33]

The overall reaction in the anode chamber is given by:



Here the specific anode half-cell reaction is:

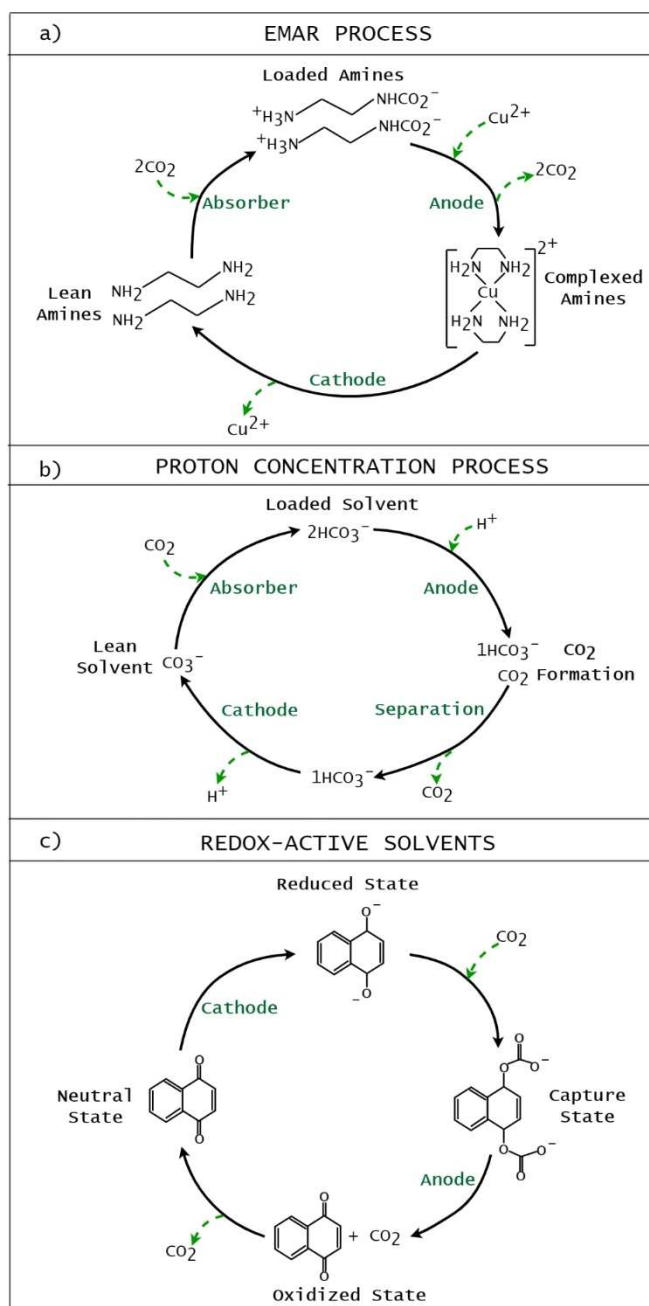
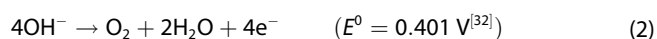
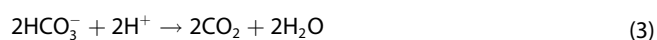


Figure 2. a) The EMAR CO₂ cycle with amines and copper. b) The proton concentration process using K₂CO₃ with deintercalation/intercalation of protons c) CO₂ cycle using a redox-active quinone.



This reaction locally lowers the pH, which pushes the HCO₃[−]/CO₂ equilibrium to the right, resulting in evaporation of CO₂ from the solvent:



Cathode half-cell reaction:



Overall production results in 2 moles of CO₂/mole of hydrogen. CO₂ and O₂ are separated from each other using compression/liqefaction methods.

Recovered CO₂ is suitable for utilization in the food and beverage industry,^[34] in green houses,^[35] or in generation of renewable fuels (by refining processes such as water gas shift reactions)^[36] thereby becoming a part of a circular carbon economy.^[35] Likewise, the hydrogen produced is of sufficient purity to be used directly (e.g., in hydrogen fuel cells for the marine industries)^[37] or to be converted to Power-to-X applications like dimethoxyethane, methanol, or Fischer-Tropsch fuels for hard-to-abate sectors^[38] (Figure 3). The additional revenue associated with H₂ is pivotal in making the capture process profitable.

In a larger societal perspective, this combined technology contributes to an effective storage of renewable energy in the form of hydrogen, oxygen, carbon dioxide, and excess heat, which can be used for district heating, thus ensuring greater national energy reserves.

5. Large-Scale Processes

To this date no electrochemical carbon capture plant has been built in an industrial scale. However, several companies in both Europe and the US are steadily working on commercializing the above technologies including ESTECH A/S, Verdox, Wetsus, and many others. An electrochemical plant at industrial scale (400 kg CO₂ h^{−1}) for post-combustion capture is planned to be operational in Denmark in 2023. This plant will be based on the coupling of carbon capture and H₂ production.

In general, electrochemical CO₂ capture makes use of the same principles as large-scale electrolysis technology for the manufacturing of chlor-alkali products.^[39] This exemplifies that comprehensive high-efficiency electrolysis plants can exist.

Despite the many advantages linked to electrochemical capture processes widespread use of the technologies is limited because of challenges to scale. Firstly, as documented by Tarpeh, improved solvents are still needed to avoid larger energy inputs.^[40] Secondly, the surface area of the electrodes' scales linearly with the amount of CO₂, thus making electrochemical cells expensive for larger CO₂ emitters. There is a certain size limit at which electrochemical solutions will no longer be able to compete with thermal approaches due to mere economic factors.

When it comes to commercial DAC solutions the electrochemical approaches suffer from the same limitations as other solvent-based capture technologies. Other concerns include: 1) the high energetic costs to deliver a large amount of air to the system,^[41] 2) the low CO₂ concentration limiting the applicable current density and causing difficulties in generating a pure CO₂ output stream,^[41] 3) impurities from the air which potentially destroy the electrochemical cell or significantly reduce its efficiency, and 4) the importance of maintaining

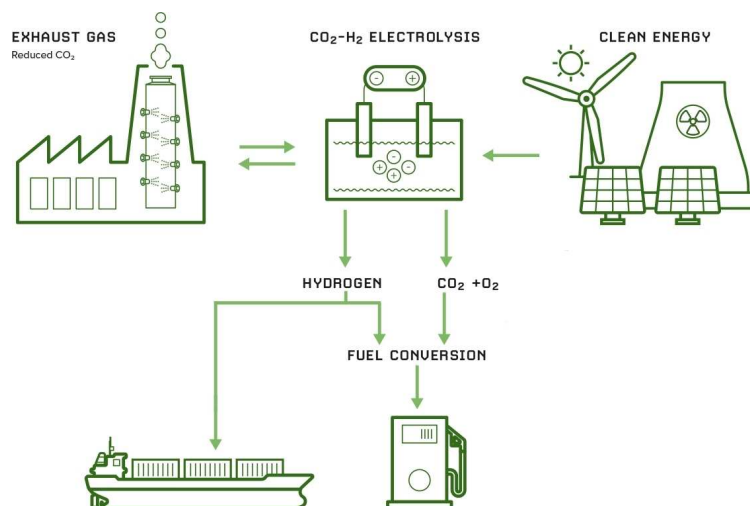


Figure 3. Coupling between carbon capture and H₂ production driven by renewable energy.

solvent balance by, for example, adding liquid or cooling the output stream.

6. Microbial Electrosynthesis

Capture and conversion of CO₂ for storage and/or utilization needs to be sustainable and function at scale and at minimal costs. Current research focusing on such new technologies is primarily based on electrochemical platforms, distinguished into two categories: 1) an exclusively electrochemical, and 2) an integrated bioelectrochemical platform.

Exclusively electrochemical research (category 1) has centered around development of inorganic electrocatalysts to facilitate direct electrochemical conversion of CO₂.^[42–44] These catalysts are advantageous with respect to high reaction rate at relatively mild reaction conditions. However, they exhibit only low chemical selectivity and insufficient long-term stability.^[45]

In contrast, bioelectrochemical systems (category 2) show remarkably high specificity, high selectivity, and stability.^[45] This is, i.e., seen in anaerobic CO₂-reducing microorganisms (microbial electrosynthesis) in conjunction with electrons derived from a cathode.^[46]

Electrochemical enzyme-based CO₂ reductions are also developed. However, enzymes require preparation from cells and immobilization on the cathode surface. Both processes make scaling of such technology a challenge. Kinetic stability of enzyme-cathode systems needs to be improved for it to become a viable option. In contrast, use of intact CO₂-reducing microbes in microbial electrosynthesis is simple, scalable, and robust over time.^[47–49] Common types of microbes used in these systems include acetogenic bacteria (e.g., for acetate or ethanol production) and methanogenic archaea (e.g., for methane production; Figure 4).^[50] Combinations of several microbial strains with selected metabolic properties enable production of multi-carbon compounds. Developments within genetic engineering and systems biology are expected to further expand

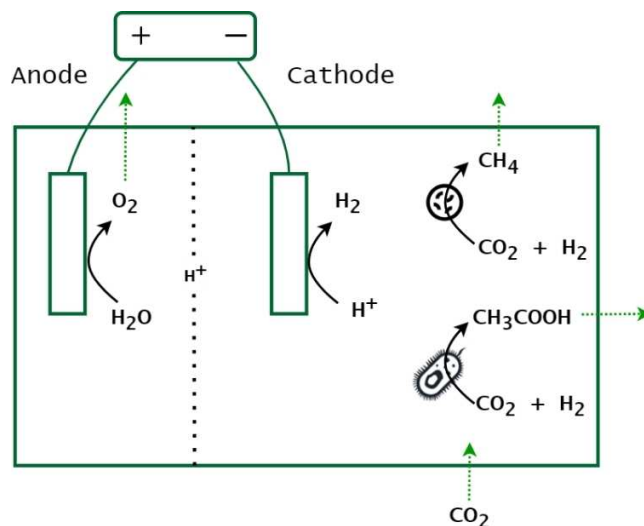


Figure 4. Integrated bioelectrochemical reactor. A proton-exchange membrane is indicated by the black dotted line. Hydrogen production happens at the cathode, providing an electron donor for the microbial reduction of CO₂. Microbial catalysts are homoacetogenic bacteria (rod-like structure) and methanogenic archaea (circular structure). CO₂ conversion is achieved, forming methane and acetate.

the spectrum of applicable microbes. Selecting microbes, cathode materials, and operating conditions is key in development and design of sustainable and large-scale microbial electrosynthesis systems.^[51]

Microbial electrosynthesis is the utilization of cathode-derived electrons for CO₂ reduction, which can occur in two different ways: 1) by a direct transfer of electrons from the cathode to a microbe or 2) indirectly by microbial use of electrochemically-generated hydrogen in the cathode compartment. The latter platform has proven most efficient for many reasons: 1) only a limited number of microbial species are able to accept electrons directly from a cathode, 2) it is a great engineering challenge to develop and maintain a high metabol-

ic rate in biofilm on a cathode 3) the intrinsic propensity of cathodes to produce H_2 at the low potentials applied is minimal, and 4) H_2 -consumption in an anoxic bioreactor is well-understood providing easier operation of such reactors.^[44]

In the electrochemical cell hydrogen evolution occurs at the cathode surface in close vicinity to suspended microbes, allowing for rapid microbial H_2 consumption (Figure 4). This cathode reaction is balanced by an oxygen evolution reaction at the anode. A selective membrane separates the cathode and anode chambers; thereby, preventing mixing of the reaction components. The presence of oxygen at the cathode can create a toxic environment for the microbes, resulting in a significant reduction of the faradaic efficiency.^[51]

Microbial electrosynthesis, when coupled to metabolically-dependent microorganisms using renewable electricity is a highly favorable platform for production of multi-carbon compounds from CO_2 .^[52] This includes e-fuels and chemical precursors such as of methane, acetate, ethanol, caproate, butyrate, and butanol, to name a few compounds.^[53]

The bioelectrochemical platform is a promising technology for future production of high-value chemicals from biogenic CO_2 .

7. Outlook

A vital tool for CO_2 recycling will include electrochemistry as a key technology for reducing emissions. Both electrified capture and conversion of CO_2 is expected to outperform state-of-the-art thermal and inorganic processes. This new generation of carbon technologies represents a breakthrough for establishing sustainable solutions to avoid CO_2 emission via a link to alternative energy sources (renewable and nuclear energies) and by making use of biological catalysts.

The electrochemical methods presented herein are still in their infancy but are expected to evolve rapidly into mature scalable technologies for industrial applications within this decade. Challenges on this pathway include long-term electrolyzer stability, electrode material development, efficient electrolyte systems, and extended use of biocatalysts to achieve higher selectivity and reaction rates. By engaging and investing in innovation of electrochemical systems it is possible to advance progress in the fight against climate change.

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Conflict of Interest

J. S. J. and J. L. B. are employed by ESTECH A/S. TAH is co-founder of Verdox. All other authors declare no competing interests.

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