

Perspective

What Should We Make with CO₂ and How Can We Make It?

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In this forward-looking Perspective, we discuss the current state of technology and the economics of electrocatalytic transformation of CO₂ into various chemical fuels. Our analysis finds that short-chain simple building-block molecules currently present the most economically compelling targets. Making an optimistic prediction of technology advancement in the future, we propose the gradual rise of photocatalytic, CO₂ polymerization, biohybrid, and molecular machine technologies to augment and enhance already practical electrocatalytic CO₂ conversion methods.

Electrochemical Utilization of CO₂ to Address the Energy Needs of the Future

As the standard of living continues to increase around the globe, especially in emerging economies, access to inexpensive and reliable energy sources will be essential to maintain the pace of technological and societal progress. Historically, the cheapest forms of electricity production have relied on the combustion of fossil fuels, resulting in the emission of carbon dioxide. However, the cost of renewable electricity has decreased so much that it is now competitive with electricity production from coal. Also, less carbon-intensive sources such as methane have taken off due to the emergence of fracking.

While low-cost electricity from renewable sources is desirable, the issues of intermittency and grid balancing remain.^{1–3} Energy storage is essential to enable even deeper penetration of renewables; to this end, it is important to take renewable electrons directly to fuels. Electrolyzing water to hydrogen is one possible solution that can integrate deeply in the energy and chemical production economy.⁴ The production of liquid fuels that can integrate with the existing transportation system, as well as more complex chemical feedstocks for chemical production would be tremendously beneficial.

The electrochemical conversion of CO₂ to fuels and feedstocks—the CO₂ reduction reaction (CO₂RR)—is an elegant solution to closing the carbon cycle when it is powered by renewable energy.⁵ In this process, CO₂ is converted to hydrocarbons using water and renewable electricity. From a capital equipment perspective, the systems hold analogy with commercialized hydrogen electrolyzers. As in the case of the net-carbon-neutral H₂/H₂O couple, the hydrocarbon/CO₂ couple is also net carbon neutral when powered by renewables.

Electrochemical transformation of renewable energy into high-energy-density liquid fuels using captured CO₂ offers the prospect of long-term, large-scale, seasonal energy storage; and it allows for integration of renewable electricity into the transportation system and in chemical production. A carbon-based strategy has advantages in implementation and logistics; it takes advantage of an expansive already-built

Context & Scale

In a world struggling to limit global temperature increases to below 2°C, we see a host of emerging technologies aiming to recycle CO₂. They range from those nearing commercialization, such as electrocatalytic reduction, to technologies being explored in the lab environment, such as photocatalytic, CO₂ polymerization, and biohybrids, to those only now being imagined, such as molecular machine technologies. With a multitude of available pathways for CO₂ recycling, we ask ourselves a question—what should we make with CO₂ that is both economically viable and helpful for the environment and how should we make it?

infrastructure created for gaseous and liquid fossil fuels.⁶ CO₂ can be captured from emission point sources using carbon capture technologies. The long-term seasonal storage of renewables holds the potential to increase the adoption of renewable energy sources.

CO₂ is a notoriously inert molecule, and catalysts are needed to activate and convert it into higher value products.⁷ Major advances in recent years have focused on nanostructured metal and metal-oxide-derived catalysts for the conversion of CO₂ to fuels and feedstocks.^{8–11} The most relevant metrics for CO₂RR catalysts are the Faradaic efficiency (FE) (the product selectivity of the reaction), the overpotential (the energy beyond the thermodynamically determined reduction potential needed to drive the reaction); and the current density (the rate of reaction).

Different compounds, including hydrocarbons, can be produced via CO₂RR depending on the catalyst being utilized. C₁ products such as carbon monoxide, methane, and formic acid, C₂ products such as ethylene, ethanol, and acetate, and even C₃ products such as n-propanol have been directly electrochemically produced from CO₂.¹² The limits for CO₂RR products have not yet been fully exhausted, with potential for even higher hydrocarbons and more complex chemicals to be synthesized.

This raises the question: Which products should we produce from CO₂? In particular, which promise to disrupt the current fossil carbon economy, and which have the greatest chance of widespread implementation? Ultimately, if a chemical could be produced by CO₂RR from renewable electrons at a lower cost than conventional chemical synthesis, powerful economic forces will drive innovation in capturing and converting CO₂.

What Should We Make Using CO₂?

Reductive coupling of CO₂ to higher-carbon products comes at a substantial energy cost; hence the high energy density of hydrocarbon fuels. While reduction to CO or formic acid only requires two proton-coupled electron transfers, the reduction of CO₂ to ethylene or ethanol consumes 12 electrons.¹³ Moreover, the coupling step poses additional constraints on the reaction, as the reaction intermediates must be present at the surface in close proximity to one another, thus requiring morphological and electronic characteristics from the catalyst.¹⁴ Further, higher kinetic overpotentials are generally needed to produce more complex carbon products. Conventional wisdom supports the production of longer-chain carbon fuels because of the increasing energy density of such compounds with increased chain length; indeed, the present-day chemical fuel infrastructure requires production of long-chain (C₄–C₁₂) carbon products. All this suggests that there is a trade-off between energy efficiency, electron intensity, and product selectivity that makes only certain products economically viable. Therefore, the highest priority should be placed on electrosynthesis of the highest gross margin products based on the efficiency of electrical energy transformation. When we examine non-branched alcohols from the point of view of energy density, we note that, while the volumetric and gravimetric energy density increase with chain length, the energy density normalized to the number of electrons transferred decreases (Figure 1A). This trend necessitates that longer-chain compounds must be produced with much higher energy efficiency compared with shorter-chain molecules to achieve similar overall energy storage capacity, which favors the production of simple and small building-block molecules from CO₂.

Thus, from a fuel (energy storage) perspective, only when any length of a hydrocarbon/alcohol in the reduction reaction step can be produced with same efficiency

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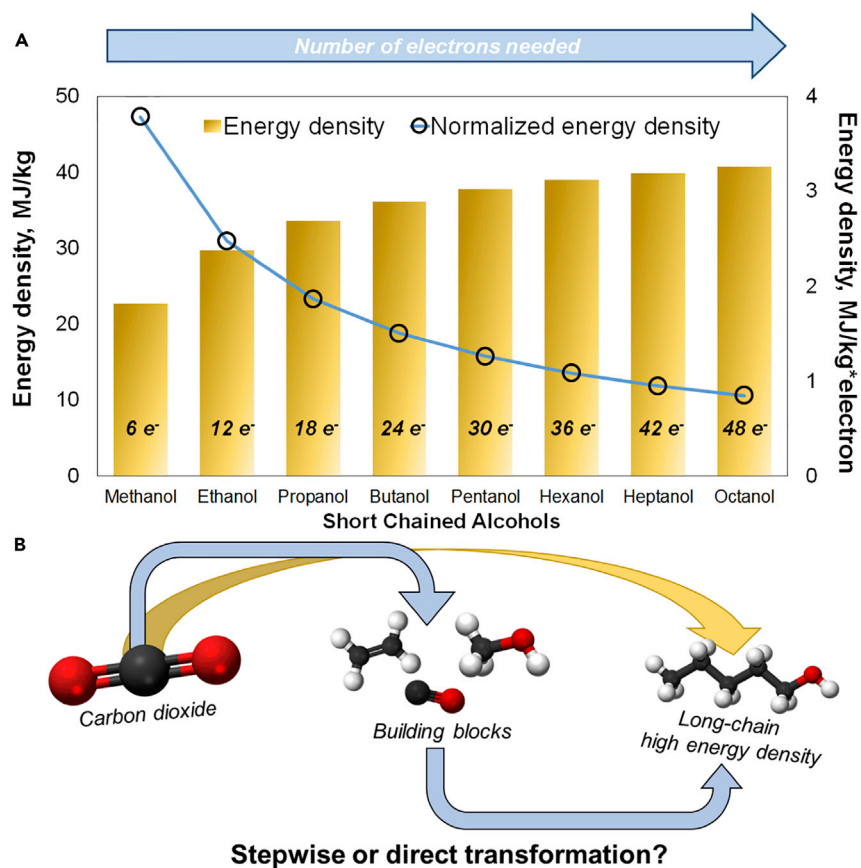


Figure 1. Rationalizing the Choice of Carbon Dioxide Conversion Products

(A) Comparison of energy density and normalized energy density of unbranched alcohols.

(B) Schematic representation of two possible pathways of CO₂RR development: synthesis of chemically active building-block molecules versus direct electrocatalytic synthesis of long-chain compounds.

(akin to insertion polymerization reactions where a monomer adds to a growing macromolecule through an active metal site) should efforts be focused on high-energy-density C₄₊ fuels. Importantly, state-of-the-art catalysts allow production of C₁ products with over 95% FE,⁸ and C₂ with around 60% FE,¹⁵ while C₃ production is limited to less than 10% FE.¹⁶ The current approach to electrocatalytic coupling involves a cascade of second-order proton/electron coupling reactions occurring at the surface competing with hydrogen evolution, which reduces FE.⁷ The costs for separation of products from a catalyst with poor selectivity, despite high activity, make this prohibitively uncompetitive.¹⁷ Unless a method is developed to produce higher-carbon products selectively and with extraordinarily high FEs, it would be more economical to focus on producing useful building blocks (methanol, CO, ethylene, aldehydes) that can be upgraded in further chemical or electrochemical processes,¹⁸ such as the Fischer-Tropsch or methanol to olefin process.¹⁹ (Figure 1B) These technologies can be applied to upgrade the building-block renewable chemicals to long-chain hydrocarbons for direct replacements of gasoline, diesel, or jet fuels.

To understand the economic viability of certain target molecules, we performed a simplified techno-economic analysis that considers the costs of CO₂, electricity, separation, capital and maintenance, and operation, and the known product

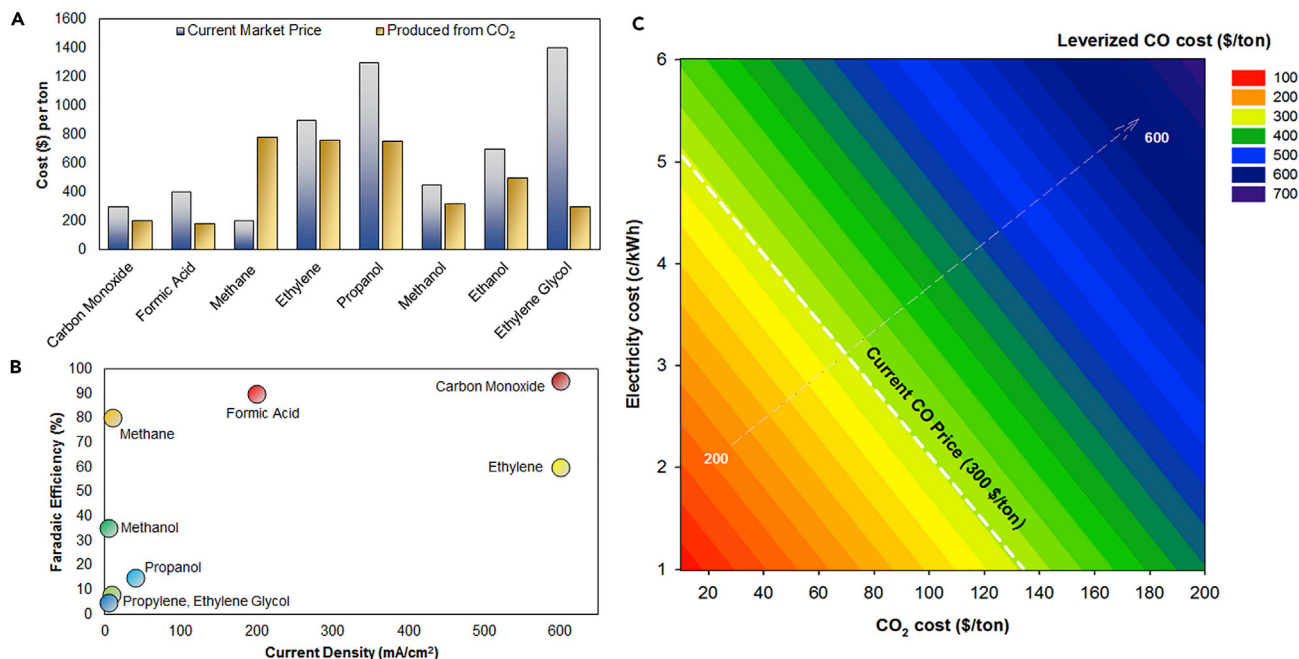


Figure 2. Techno-economic Analysis of CO₂ Electroreduction

(A) Comparison of the cost of various chemicals between the current market price and the leverized cost from CO₂ reduction assuming an electrolyzer cost of \$500/kW, energy conversion efficiency of 60%, Faradaic efficiency of 90%, electricity cost of 2 ¢/kWh, and CO₂ cost of 30\$/ton.

(B) Comparison of CO₂-derived molecules based on technical parameters of Faradaic efficiency and current density.

(C) Contour plot of economics of CO production based on electricity and CO₂ cost.

selectivity; and that outputs the leverized cost of the chemical produced. Results of this analysis for several target molecules are given in Figure 2. The argument for energy storage can be justified by analyzing the currently achievable combination of crucial performance parameters, FE, and current density (Figure 2B), which suggests a focus on small-molecule C₁ and C₂ products. Energy conversion efficiency aside, the adoption of any CO₂RR to fuel process will ultimately be determined by market forces; and currently these also favor the production of short-chain molecules. Recent analyses point to CO and formic acid as economically viable and atom-economic targets.²⁰ By benchmarking cost parameters to the commercial hydrogen electrolyzers (cost of \$500/kW and energy conversion efficiency of 60%), fixing the electricity cost at 2¢/kWh (expected to be achievable using renewable sources), assuming a CO₂ cost of 30\$/ton and performing leverized cost calculations, we add ethylene glycol and propanol as economically attractive targets. The cost of CO₂, which is sensitive to parameters such as capture expenses, transportation, and, in some countries, taxation, serves as another crucial input parameter for economical CO₂RR. The interplay between the costs of electricity and input CO₂ on reduction of CO₂ to CO can be assessed in Figure 2C. It can be concluded from this preliminary analysis that, even at the current level of CO₂RR technology, CO₂ reduction could potentially be made economical with access to low-cost renewable electricity. Advances in CO₂ reduction to higher value products such as ethylene glycol and propanol holds even greater economic promise.

Need for Greater Understanding of the Basic Science of CO₂RR

Despite significant advances in the production of C₁ products, CO₂RR is far from being mastered and fully understood. To date, products such as CO and formate can be produced with an impressive 95+% FE,^{8,21} methane production is less well

understood. Density functional theory can be used to model the reaction and determine the most likely reaction pathway from calculating the interactions between the reaction intermediates and the surface.^{22–24} The most effective catalysts rely on expensive metals, while cheaper catalysts tend to have problems with long-term stability.⁸ The task of producing C₂ products is even more complex. Despite some encouraging results, such as 50%–60% C₂ products reported in several papers,^{14,15} C₂+ production is still nascent; crucial intermediates and reaction pathway branches of the process differ between materials, and are not universally known.^{25,26}

Coupling to multi-carbon products requires arrival and adsorption of several CO₂ molecules to the surface, stepwise transformation, and spatial positioning. On a macro- or mesostructured catalytic surface, such coupling will necessarily obey statistical constraints.²⁷ Highly selective or quantitative reactions would benefit from perfectly sized compartments (akin to metal-organic framework cavities) and a defined cooperative manner; that is, no reaction will occur until all adsorbed reagent molecules arrive in the reactor. Another possible solution may involve mimicking biological enzymatic processes or metal-catalyzed insertion polymerization processes. Without cooperativity and defined reactor sites, it is unlikely that we will ever reach 99.9% efficiency of transformation, as higher or lower carbon products will be present when the reaction is indiscriminate. A heterogeneous metal surface is unlikely to yield the solution to C₂+ product synthesis, since to mimic such a nano-reactor will require preparation of large-area defined multi-metal surfaces with atomic precision. Stochastic mixtures will lack the proper geometric and chemical control, and thus alloys,²⁸ solgels,²⁹ or any other mixture methods may not yield efficient C₃ production. In the next section, we present research into promising technologies that may provide the precise geometric and chemical control to yield C₃ products.

Vision for the Future of Fuels and Chemicals from CO₂ for the Coming Decades

Despite the ongoing challenges of CO₂RR to higher-carbon products, recent advances in the field offer untapped potential for the realization of CO₂ transformation. We envisage at least six potentially disruptive CO₂ catalytic conversion technologies that are currently topics of intense research (Figure 3). Some of these technologies are close to commercialization, others are at the benchtop scale, and some have yet to be scientifically proven. We envision a time line for realization on a large scale between 5 and 70+ years. Ours is an optimistic prediction of technology advancement in the future assuming that carbon dioxide conversion remains a topic of widespread interdisciplinary interest and global activity. The specific technologies we have identified are by no means an exhaustive list of the potential viable solutions.

The technologies based on electrochemical conversion of CO₂ are closest to commercialization with startup and established companies such as Opus-12, Mitsui Chemicals, Carbon Recycling International, Dioxide Materials, and Carbon Electro-catalytic Recycling Toronto currently leading the pack to monetize the technology.³⁰ As the price of renewable energy continues to decrease, reaching 2 ¢/kWh in some jurisdictions,³¹ the electrochemical conversion of CO₂ becomes more attractive as the electricity cost is the largest expense.

Direct solar to fuel conversion using semiconductor catalysts in gas-phase CO₂ reactors are another attractive technology that has seen great advances. These integrated photochemical (PC) systems mimic natural photosynthesis and hold an advantage of mobility afforded by being independent of an electricity source, relying purely on solar radiation to produce fuels.

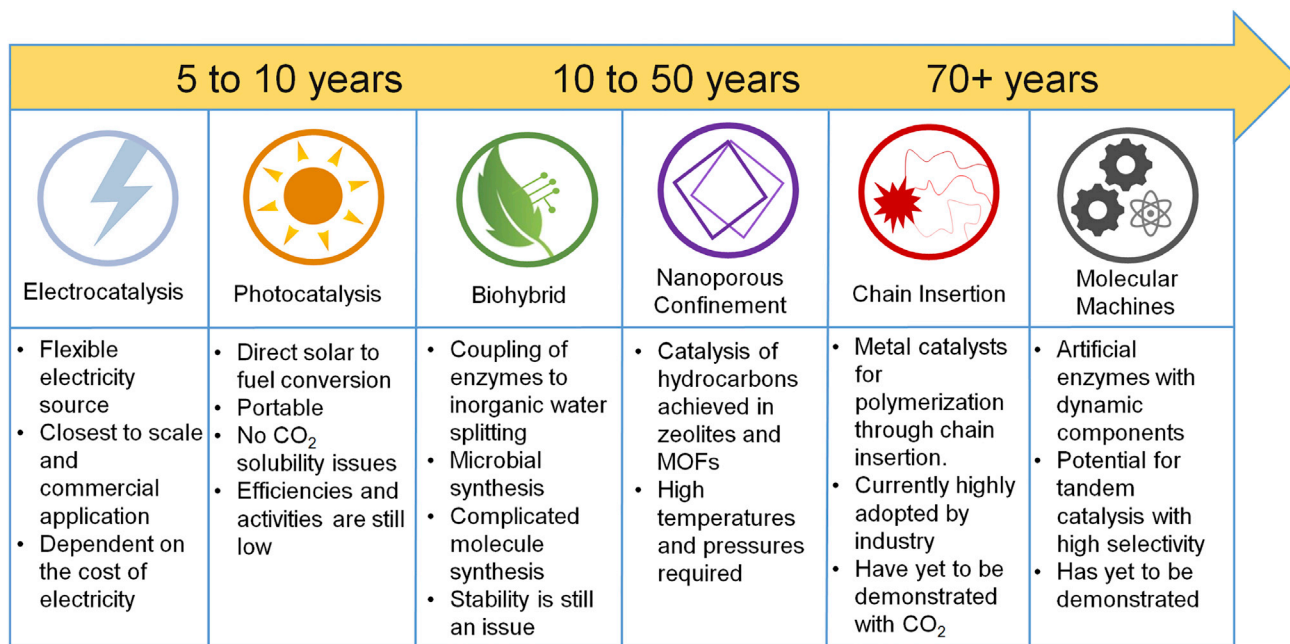


Figure 3. Proposed Timeline of CO₂ Utilization Methods

Note the specific time ranges are based on extrapolation of time line development of other disruptive technologies such as the advent of 3D printing, solar energy adoption, and electric vehicle development.

Biohybrid systems couple inorganic water-splitting catalysts with enzymes or genetically modified bacteria that convert CO₂. These systems have potential to utilize natural enzymatic pathways to convert CO₂ into a wide range of products. This bio-electrochemical approach has only started to be explored,³² but holds great promise if key issues such as long-term stability can be solved.

Thermocatalysis in nanoporous materials for the conversion of hydrocarbons has been known for some time and has been industrially implemented. However, nanoporous materials have primarily been explored for capture of CO₂ gas as solid sorbents or as supports for thermal catalysis; they have seldom been explored as catalysts for electrochemical conversion of CO₂. Tunable porous materials such as metal-organic frameworks have recently been shown to drive electrochemical conversion of CO₂ when functionalized with catalytically active sites, albeit to primarily C₁ compounds.^{33,34}

Two promising technologies that have not yet been realized for CO₂ conversion include polymerization chemistry such as chain insertion catalysts using activated CO₂ and molecular machines for dynamic CO₂ catalysis. The ability to use CO₂ as a monomeric unit directly would be transformative in the production of consumer goods, and allow for the sequestration of gaseous CO₂ into solid products. Molecular machines are made up of rotating ring units around rigid struts in porous materials such as metal-organic frameworks; the ability to mechanically control the movement of molecules at the atomic level has the potential to unlock artificial molecular factories not dissimilar to enzymes.

The future of energy and carbon utilization hinges on the fundamental discovery of materials and catalysts that efficiently and selectively convert CO₂. Here, we have presented a preliminary techno-economic analysis that shows the promise of

CO₂ utilization in the production of chemicals if barriers can be overcome. We have reviewed the currently achievable current densities and FEs of various chemicals. Finally, we have proposed several rapidly progressing catalytic processes that have the potential to provide a disruptive solution to carbon dioxide conversion. With the large number of hydrocarbon molecules that can be made from the CO₂ building blocks, the question remains: which products should we pursue and which products should we forego? Our analysis has identified a series of possible target molecules that could be made economically using CO₂RR powered by renewable electricity. For energy storage needs, hydrogen, methane, and ethane are all excellent fuels. In addition, ethylene and ethanol are versatile CO₂-derived chemical feedstocks. We pose the question whether there exists a niche for C₃+ products considering the difficulties that are likely to be faced? Is there a catalytic method yet to be realized that will efficiently, selectively, and consistently convert CO₂ into complex molecules? We are unsure, but we are excited to find out.

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AUTHOR CONTRIBUTIONS

O.S.B. and P.D.L. wrote the manuscript and curated the content. C.T.D. provided economic modeling and edited the manuscript. L.T., G.S., and J.v.d.L. edited the manuscript. S.O.K. and E.H.S. oversaw the manuscript development.

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