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# **Current and Emerging Electrochemical Approaches for Chemical Manufacturing**

### by Elizabeth J. Biddinger and Paul J. A. Kenis

ociety is facing a major challenge: the need to slow and eventually curb climate change induced by excessive anthropogenic emissions of greenhouse gases, in particular carbon dioxide. The scientific community and governments largely agree—as summarized in the Paris Agreement<sup>1</sup> that massive cuts in CO<sub>2</sub> emissions will be needed over the next two to three decades to avoid the world heating up by more than 2 °C. Already, many of the sectors responsible for the largest fractions of CO<sub>2</sub> emissions are making major strides toward that goal. Electric power is increasingly generated using renewable energy sources rather than fossil fuels, transportation applications are increasingly electrified, and commercial and residential buildings have become significantly more energy efficient (e.g., insulation, better HVAC systems).<sup>2</sup> Similarly, the industrial sector is searching for ways to achieve the so-called energy transition, implementing changes culminating in becoming (close to) carbon neutral by 2050. In the early 2000s, few people thought that electrified chemical manufacturing approaches would be able to contribute significantly to the reduction of CO<sub>2</sub> emissions of this sector. A multitude of developments since then have brought electrified chemical manufacturing approaches much closer to reality:3 (i) Electricity (especially the renewable fraction) is much cheaper now; (ii) Industry has realized that the CO<sub>2</sub> emissions of many conventional chemical manufacturing processes can be cut significantly by electrifying the way in which the process is driven (e.g., switching from burning fossil fuels to generate heat to electrified heating approaches); (iii) The increased pace by which renewable feeds or waste streams are being identified as potential feeds for electrified chemical conversions; and (iv) Now that electrochemical synthesis has emerged from a very small field of research to one of the most active fields of study in the chemical sciences, many unprecedented, promising opportunities for electrifying different chemical manufacturing processes are being identified (new chemistry, less harsh, fewer steps, ...). Below, after a brief account of historic electrified chemical manufacturing processes being used at scale, we cover some of the emerging developments and opportunities for electrochemical manufacturing, both those that will aid in reducing greenhouse gas emissions, and those that offer new, more efficient synthetic pathways to desired fine chemicals, pharmaceuticals, and other products.

#### **A Bit of History**

Despite electrochemical manufacturing methods in the chemical industry often feeling "novel," "new," or "not yet implemented," the foundations of the chemical industry go back to electrochemical manufacturing. The Dow Chemical Company was started in 1897 to electrolyze brines found in Midland, Michigan to recover bromine and chlorine.<sup>4</sup> The chlor-alkali process to produce chlorine, hydrogen, and caustic soda (sodium hydroxide) from aqueous sodium chloride is one of the most used chemical processes today and it is an electrochemical process. In fact, this article is part of the 80th anniversary of the founding of what is now the Industrial Electrochemistry & Electrochemical Engineering Division of The Electrochemical Society. This division was originally the "Industrial Electolytics Division" with its membership centered around the chloralkali process. For many years, the division also published a "Report of/on the Electrolytic Industries" as an annual update in The Journal of The Electrochemical Society with a significant focus on the status of the chlor-alkali industry and other updates on electrochemical processes at scale (the last report was on the status of electrochemical manufacturing in 2004<sup>5</sup>).

#### Electrochemical Manufacturing at Scale Today

While chlor-alkali continues to dominate in terms of electrochemical process production volume and energy input (20 GW installed capacity<sup>6</sup>), there are other significant electrochemical processes utilized industrially as well. For example, the Monsanto electrohydrodimerization of acrylonitrile to adiponitrile, an intermediate in the production nylon-6,6, went online in 1965 and is used in one-third of all adiponitrile produced today. Additionally, BASF electrochemically dimethoxylates 4-tert-butyltoluene at a scale of tens of thousands of tons per year. A multitude of other chemical products have been manufactured electrochemically over the years, including L-cystine, furan, alkyltoluenes, maleic acid, nitrobenzene, butanone, and many more.<sup>7,8</sup> The historical electrochemical processes have been summarized in previous *Interface* articles<sup>9</sup> and handbooks.<sup>8,10</sup>

The historical motivation for electrochemical manufacturing of chemicals has largely been to access chemistry otherwise not possible or very difficult, or to eliminate hazardous intermediates, side products, or stoichiometric reagents.<sup>9</sup> Significant opportunities exist in the future to pair electrochemical manufacturing with renewable electricity for both decarbonization<sup>3,9</sup> and a now-inexpensive oxidizing or reducing reagent (i.e., the electron). Using existing and previous electrochemical manufacturing processes, significant opportunities are emerging for the manufacturing of additional chemicals electrochemically.

#### Electrochemical Conversion Approaches that Hold Potential for Chemical Manufacturing

#### **Hydrogen Production**

The production of hydrogen via water electrolysis is already gradually replacing hydrogen generated from fossil fuels (historically steam reforming of natural gas) and is the next big electrochemical process to be realized in the industry. The main driver of water electrolysis today is decarbonization of hydrogen as a chemical feedstock. There are also significant opportunities to use hydrogen for long-term energy storage of variable renewable electricity and for carbon-neutral combustion for industrial heating in the future.<sup>3</sup> In 2021, >500 MW of water electrolysis capacity had been installed globally, with rapid growth anticipated to bring capacity to several hundred gigawatts by 2030.6 While water electrolysis can be performed at large scale in an economically feasible manner, research in academia and industry continues to focus on improving the process, in terms of more active and inexpensive catalysts and of improved overall durability for both the cathode (hydrogen evolution) and the anode (oxygen evolution).

#### **CO<sub>2</sub> Reduction**

As water electrolysis technology has matured, the technological knowledge has expanded to other feedstocks and opportunities for decarbonization. Electrochemical reduction of CO<sub>2</sub> has emerged as a possible carbon neutral or even carbon negative approach for the production of intermediates of key interest to the chemical industry, such as CO, formate, methanol, methane, ethanol, and ethylene.<sup>11</sup> Over the past 15 years, through an explosion of research activity,

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(continued from previous page)

reasonable active and selective catalysts have been developed for each of these products. Current research efforts focus much more on reactor engineering challenges: achieving high rate and stable performance in membrane-based electrolysis cells. A particularly difficult challenge is the side reaction of  $CO_2$  to bicarbonates that tend to precipitate on or in the electrodes, spurring studies on carbonate formation<sup>12</sup> as well as the overall carbon balance.<sup>13</sup> Indeed, the study and improvement of electrode durability is a major focus of current research.<sup>14</sup>

Another aspect that electrochemical  $CO_2$  reduction and water electrolysis have in common is that both are still energy intensive. A significant fraction of the energy required to drive the process is needed for the anodic process of oxygen evolution. The combination of oxygen evolution thermodynamics (i.e., a very positive theoretical redox potential, 1.23V vs. SHE) and slow kinetics can add several volts to the operating cell potential. Pairing  $CO_2$  reduction or hydrogen with less energy intensive half reactions (e.g., oxidation of organics) drastically reduces the cell potential and thus the overall energy demand.<sup>15</sup>

#### **CO Reduction**

The electroreduction of CO, rather than CO<sub>2</sub>, has also started to garner attention for application at scale. Industry is looking for electrified processes that will replace some of the steps that emit the most CO<sub>2</sub>. Through decades of research and industrial practice, CO can be produced very efficiently from methane, which continues to be abundantly available and relatively cheap. Furthermore, the CO<sub>2</sub> emissions of this process could in principle be reduced to close to zero. Replacing the much more energy intensive and CO<sub>2</sub> emitting subsequent conversion of CO to the intermediates needed for the manufacturing of a broad range of chemicals is a much more attractive candidate for electrification. Furthermore, unlike CO<sub>2</sub> electroreduction, the CO electroreduction process is not hampered by carbonate formation, simplifying its path toward a process at scale. Indeed, CO reduction has become a very active area of investigation, holding promise for production of intermediates like acetate, ethylene, and acetaldehyde.16,17,18

#### **Electrosynthesis of Organics**

As more and more processes are being electrified, interest has returned to utilizing electrochemistry in organic synthesis. Significant research activity in the last decade has occurred in the field of electro-organic syntheses with applications in fine chemicals and pharmaceuticals.<sup>19-23</sup> The activity has been driven by progress on multiple fronts: (i) the ability to access chemistries that through traditional synthetic means are difficult, hazardous, or highly waste generating, and (ii) the development of enabling technologies for non-electrochemists such as the IKA Electrasyn systems that provide "plug and play" reactions in standardized vials on a stir plate without the need for a potentiostat paired with recent tutorials geared toward organic chemists.<sup>24-26</sup> Electro-organic reactions can be used in coupling reactions such as with C-C and C-N bond formations, functionalization with heteroatoms, selective deprotections of functional groups, reduction of double bonds, hydrogenations, and many more that are of importance for the organic chemist's toolbox.<sup>2</sup> In the most appealing cases, complex multi-step reactions (even 10+ steps) can be performed selectively in single steps using electrochemistry. The electron transfer may be direct at the electrode surface or through a mediator, similar to a homogeneous catalyst that has been activated by electron transfer at the electrode surface. The electron can become a replacement for stoichiometric oxidizing or reducing agents, while also eliminating the stoichiometric generation of salts that would require disposal. While much of the research has been on milligram-gram scales in batch reactors to identify new reactions or to improve yields, efforts are also being made to incorporate flow electrochemistry and scale-up conditions.27,28

New electro-organic reactions in the fine chemical and pharmaceutical industries are likely to be implemented first where substantial process advantages can be obtained. Merely having a higher yield or selectivity to a desired product is unlikely to meet the activation barrier of implementation of new electrochemical process equipment for fields unseasoned in the use of electrochemical reactors. Processes susceptible to significant minimization of the number of synthesis steps or the elimination of hazardous species are likely to be most sensible for early adoption economically and make the risk of taking on a new transformation method (i.e., electrochemistry) worthwhile. To enable the transformations, scale-up procedures and off-the-shelf electrochemical reactors able to produce 1-1000kg/day need to be widely available. Membranes that are stable and selective in organic solutions need to be developed and economical for when undivided cells are not favorable due to undesired reactions at the counter electrode. Many fine chemical and pharmaceutical chemical processes occur in multi-product facilities-scheduled in short duration runs-rather than in purpose-built single, continuous product facilities. Having electrochemical reactors as part of the inventory available in these multi-product facilities will open up the possibility of performing reactions in which electrochemistry improves yields but does not change the overall production pathway. After the industries have established electrochemical processes and invested in the reactors and infrastructure, electrochemical reactions can become part of the more common "toolbox" of processes to consider.

#### **Biomass-Derived Conversion**

Many biomass-derived species have been identified as alternative platform molecules to serve as building blocks for the chemicals and fuels industries27,28 and have led to new and/or re-invigorated investigations into electrochemical upgrading of the building blocks. Biomass-derived species, including furanics, phenolics, glycols, carboxylic acids, and aldehydes, have largely been the focus of electrochemical reactions. Upgrading has included electrochemical hydrogenation (ECH), dehydrogenation (ECD), oxidation (ECO), dimerization, and ring-opening reactions to form intermediates for fuels and chemicals.<sup>29-37</sup> The electrochemical depolymerization and upgrading of lignin is also an area of study.37,38 The drive for electrochemical, rather than thermochemical, upgrading of biomassderived species has included the ease of integration of renewable electricity, ability to operate at ambient temperatures and pressures, and reduced infrastructure needs such as steam or hydrogen gas. Electrochemical upgrading of biomass-derived species at Biomass Upgrading Depots (BUDs) has been shown in studies to be economically feasible.<sup>39</sup> Additionally, with the advances in hydrogen evolution and CO<sub>2</sub> electroreduction, biomass-derived species and wastes have been examined as alternative oxidation reactions for oxygen evolution that would bring both enhanced value in the paired electrolysis and lower overall cell potential compared to oxygen evolution.40

The focus of the research on electrochemical reactions of biomassderived species has largely been from a reaction engineering perspective-improvements in faradaic efficiencies, selectivities, catalyst development, electrolyte influences, and reactor designs. Unlike in fine chemical and pharmaceutical electro-organic syntheses, the value of the biomass-derived species is much closer to that of a commodity/specialty chemicals interface. Electricity costs, while continuing to drop per kilowatt-hour, still make up a significant portion of operating costs for these lower-value chemicals. Efficient, high-surface-area electrocatalysts with minimal precious metals are also becoming an area of significant interest now that the reactions have been demonstrated to be catalytic, not just electron-transfer, in nature. One of the driving factors for electrochemical transformation of biomass-derived species is to improve the stability of the final product. This will entail identifying reaction conditions that enable the handling of reactive species and contaminants so that the desired products are formed, rather than humins or other undesired species. While there are many opportunities for electrochemical conversion of biomass-derived species, the likely first large-scale implementations will be in electrooxidations at the anode paired with hydrogen evolution at the cathode to produce two valuable products and lower the cell potential so that hydrogen can be produced with less energy input.

#### **Ammonia Electrochemistry**

Driven by the tremendous global need for ammonia, which is critical for fertilizer manufacturing and other applications, researchers have sought ways to synthesize ammonia directly from nitrogen using a less energy-intense approach than Haber-Bosch. Electrochemists have long recognized this challenge/opportunity too, but finding promising electrocatalysts or an electrochemical approach has proven to be elusive, as has been recently reviewed.<sup>41</sup> In recent work, Chorkendorf and colleagues demonstrated that a platinumgold alloy in a 25 cm<sup>2</sup> electrolysis cell is able to produce ammonia with a faradaic efficiency of >60%, yet at an energy efficiency that is still only 13%.42 While work likes this represents a major advance, to be feasible for application at scale, energy efficiencies exceeding 50% are needed. An alternative, abundantly available, possible feed for ammonia production is nitrate-containing waste streams such as agricultural runoff. Efficient electrochemical conversion of nitrate to ammonia has been demonstrated<sup>43</sup> but the fact that most of the nitrate containing feeds are highly dilute remains a problem.

In addition to interest in electrochemically synthesizing ammonia, ammonia also has emerged as a promising hydrogen carrier since it features a volumetric density of hydrogen that is 1.7 times greater than that of liquid hydrogen. However, present-day approaches like thermal cracking that are used to liberate H<sub>2</sub> from ammonia are not energy efficient. Ammonia electrolysis in alkaline media, where ammonia oxidation on the anode to produce N2 is paired with water reduction to produce hydrogen, has the potential to be much less energy intensive. While the reduction reaction to produce  $H_2$  is relatively efficient, the ammonia oxidation reaction (AOR) occurring at the anode is not. Present-day catalysts for this reaction exhibit high overpotentials to achieve reasonable rates and are easily poisoned by byproducts of the AOR itself. Research on identifying better AOR catalysts has significantly increased due to the importance ammonia could play in the energy transition, by enabling transfer of green hydrogen over long distances. Recent efforts have started to identify interesting binary and ternary catalysts, in part identified using machine learning approaches, that may address the two challenges of high overpotential and propensity for poisoning.44,45

#### Process Intensification and Reactor Engineering

Research on the wide range of electrochemical conversions described above has also spurred exploration of a wide range of electrochemical reactor configurations. While typically initial characterization of new catalysts is done in a three-electrode H-cell, evaluation of the performance (activity, selectivity, stability) of those catalysts requires their integration in electrodes for flow cells and/or membrane-electrolyte assembly cells. Several studies have focused on developing such reactors, including multilayer stacks,<sup>46</sup> and/or on process intensification by systematic evaluation of a number of parameters, ranging from catalyst loading to operation parameters such as feed and electrolyte flow rates.<sup>47</sup> A next level of complexity being studied is so-called tandem approaches, where feeds such as CO<sub>2</sub> are converted to desired products in multiple consecutive steps (e.g., CO<sub>2</sub> to CO to acetate).<sup>48,49</sup> A number of studies also are exploring ways that allow the anode and cathode chemistries to be de-coupled (at times referred to as modular electrochemical synthesis), through the application of bipolar membranes, or heterogeneous redox reservoirs, which for example was used to demonstrate electrochemical hydrogen peroxide synthesis.<sup>50</sup> It is these types of approaches that may enable electrochemical manufacturing platform technology capable of coupling large-scale cathodic processes (hydrogen evolution; CO<sub>2</sub> or CO reduction) with a number of different smaller scale anodic oxidations of organics.

#### **Remaining Overarching Challenges**

The preceding section summarized R&D efforts on some of the many different electrochemical manufacturing approaches being pursued at present. Indeed, many other chemistries are being pursued for electrification. Beyond considering specific electrochemical conversions, a number of common or overarching challenges still hamper many approaches for electrochemical manufacturing of chemicals from being pursued for scale up.

- (i) Lack of suitable electrochemical infrastructure. Unlike other chemical reactors, electrolysis systems are highly specific to the chemical conversion for which they are being used. Going beyond a laboratory scale of benchtop proof-of-concept is thus not trivial. Also, it is not obvious what entity would be most effective in developing and selling these needed durable electrochemical reactors. Furthermore, the end users, here the chemical companies, are hesitant to be the first to invest at a level needed to test new, unproven technology at pilot-plant scale or beyond.
- (ii) Lack of familiarity with electrochemical conversion processes across many industries. Implementation of new technology will be slow given the lack of expertise with new reactors etc., leading to hesitancy in their implementation. The field would benefit from facilities where anyone can come in and test electrochemical manufacturing approaches at a larger scale (up to pilot plant) for longer run times (to test durability), to speed up the development of new reactor technology, and to help familiarize designers and implementers with this new technology, thereby removing their hesitancy. The National Renewable Energy Laboratory (NREL) in Colorado may play a major role here.
- (iii) Limited availability of (renewable) electrical energy (grid) and electrical hardware to drive the process. It is possible to calculate how much grid power would be necessary to operate electrochemical manufacturing plants at scale. This is quickly followed by the conclusion that the present grid capacity, let alone the present fraction of renewable energy in the grid, is insufficient for even partial electrification of chemical manufacturing. Furthermore, like the electrochemical reactor technology, further development of electrical hardware and controls for operation of electrochemical manufacturing plants at scale will be needed, especially if these electrochemical manufacturing facilities need to become part of regional grid-scale load-leveling efforts to maximize utilization of renewable power while ensuring economic feasibility. Already, sector coupling approaches are being studied, where the variable electrical power supply and demand is connected with hydrogen production (water electrolysis).51
- (iv) Variability in feed composition. Many of the approaches for electrochemical manufacturing described above for different applications will rely on feeds that may vary significantly in composition depending on the specific source. Take for example "crude glycerol," a byproduct of biodiesel production produced by hundreds of plants. Its composition can vary over 30–70% glycerol, 10–40% methanol, 4–25% NaOH, and a few other ingredients. No process would be able to handle such a broad composition range without significant adjustment of the feed and/or the operational parameters. The same is true for captured CO<sub>2</sub> streams from different plants or regions. Standardized feed compositions may need to be defined for processes moving to application at scale.<sup>52</sup>
- (v) Need for techno-economic and life cycle analyses and associated models. Due to the much more regional availability of feeds, electrochemical manufacturing facilities will probably be deployed in a more regional fashion compared to the present situation of large-scale chemical production facilities and refineries being deployed only in a limited number of locations across the continents. See Fig. 1. For industry to be willing to invest in the deployment of such regional facilities, high quality techno-economic assessment (TEA) and life cycle assessment (LCA) studies will be needed, coupled with regional models that take into account regional availability of critical resources, such as those developed for biofuel (ethanol)



**Fig. 1:** Side-by-side comparison of available biomass resources (*left*) and oil & natural gas processing and transportation infrastructure (*right*) in the US. Whereas current fossil fuel processing facilities are located in the vicinity of oil and (shale) gas fields, and along major pipeline networks to/from those, one can foresee that future electrified chemical manufacturing facilities that use biomass resources as the feed will need to be located along the West Coast, the Midwest (utilizing significant amounts of currently stranded wind power?!), along the Mississippi River, along the East Coast, and in the Northeast. Similarly, one can foresee how electrified chemical manufacturing facilities could be co-located with current point sources of CO<sub>2</sub> (fossil fuel–based chemical industry and power plants), while those utilizing CO<sub>2</sub> from direct air capture could be localized with much fewer constraints, including in vast areas (West/Central) that have limited biomass resources. *Sources: Left Map – A. Milbrandt*, Technical Report, *NREL/TP-560-39181*, December (2005). *Right Map: Interactive "National Energy and Petrochemical Map," Fractracker Alliance, accessed April 2023*.

production and other biomass conversions.<sup>53</sup> Almost every electrochemical process will need a dedicated analysis effort to assess its economic feasibility and its remaining carbon footprint. While many TEA and LCA studies on electrifying chemical manufacturing have appeared, most of these lack those detailed, regional deployment considerations, as well as spatiotemporal aspects of variable electricity cost.

#### Conclusions

We hope that the above summary provides the reader with insight into the promise and remaining challenges associated with electrifying chemical manufacturing across many types of chemistries and applications, ranging from commodity intermediates to specific pharmaceuticals or fine chemicals. We wish to explicitly acknowledge that other approaches and chemistries are being pursued, or maybe have already been implemented in industry. The scenarios and categories we present here are intended as examples.

Many of the directions and/or specific examples covered above have a possible role to play in decarbonizing emissions associated with future chemical manufacturing. In reality, most of these approaches when implemented will not be carbon neutral, let alone be carbon negative, for the simple reason that only a (gradually increasing) fraction of the electrical power needed to drive the process will be derived from renewable energy sources. In a similar vein, it is important to point out that the energy transition, with a goal to arrive at close to carbon neutrality by 2050, is a very gradual process. Industry does not have the resources to abruptly abandon its massive investment in existing plants in lieu of modern carbonneutral processes. Furthermore, the chemical processes needed for a complete electrification of chemical manufacturing are just not available at this point. Beyond the need to reduce CO<sub>2</sub> emissions associated with chemical manufacturing, electrochemical processes at times also offer more efficient conversions, avoided steps, elimination of hazardous materials, higher yields, and higher selectivity. Key advances in industrial practice in the production of fine chemicals and pharmaceuticals are evident, underscored by the many chemical and pharma companies that now have dedicated electrosynthesis efforts.

Despite these issues, electrification of chemical manufacturing is already happening, with a focus on the more straightforward opportunities, such as transitioning from heating based on burning fossil fuel to electrified, resistive heating, or the in-situ production of the hydrogen needed for a process via a water electrolysis approach instead of through thermo-chemical methods. Implementation of new chemical conversion processes will be significantly more challenging. As stated above, centralized facilities where academic and/or corporate teams can try out a process at a larger scale, where they can tackle durability and other challenges, would greatly benefit the progress of the field.

Driven by the need to decarbonize chemical manufacturing, a very healthy number of funding opportunities from the federal agencies and from industry, and the fascination of many researchers with these relatively new processes and the unique capabilities they offer, the field of electrosynthesis indeed is experiencing a sort of renaissance. Indeed, the future of electrochemical manufacturing, in terms of scholarly research opportunities, as well as in terms of potential impact on industrial practice, is bright!

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