

**CONVERSION OF CARBON DIOXIDE TO FORMIC ACID VIA
ELECTROCHEMICAL REDUCTION IN A THREE-COMPARTMENT
ELECTROLYZER**

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EXECUTIVE SUMMARY

To serve a growing global population, the energy sector burns fossil fuels, releasing carbon dioxide (CO₂) into the atmosphere and contributing to rising average global temperatures. Thus, the energy sector, and specifically natural gas power plants, are looking to capture and convert their carbon emissions into a sellable product, which is the focus of this report. Multiple CO₂ conversion pathways were considered: concrete, algae, carbon black, chemicals, and fuels. Ultimately, electrochemical reduction of CO₂ in a three-compartment formic acid (FA) electrolyzer was selected. Not only is the existing formic acid market expanding, but formic acid has been studied as a viable input for fuel cells, which are a growing strategy for clean energy generation. Moreover, formic acid may be used for clean hydrogen generation, transportation, and storage, which is key for decarbonizing the industrial sector.

A lab-scale electrolyzer was purchased, where the primary inputs are DI water and CO₂, and the primary output is formic acid. Bench scale experiments were performed by varying the DI water input flow rate to maximize formic acid concentration; the ideal flow rate was determined to be 0.05 milliliters per minute. After analyzing various parameters at this flow rate, it was found that this particular electrolyzer set-up and operation was much less efficient than other studies; thus, scale-up calculations were completed using another formic acid electrolyzer study and a peer-reviewed techno-economic analysis (TEA) for CO₂ electrolyzers. After completing the TEA, it was determined that converting 100% of the CO₂ emissions from a standard (228 megawatt (MW)) power plant that emits 900 pounds of CO₂ per megawatt-hour (MWh) was not technologically feasible. Instead, a reasonable production target of 50,000 kilograms of FA per day was set, and a scaled-up FA electrolyzer facility was developed from that production target. Scale-up costs and performance are summarized in Table 1.

Total energy required (MWh)	Base case cost per lb CO ₂ converted (\$/lb)	Rate of conversion (lb CO ₂ /day)	Total CapEx for 10-year plant life (\$/day)	Total OpEx for 10-year plant life (\$/day)	Maintenance Cost Breakdown (\$/day)
8.5	\$3.55	105,369	\$1,646	\$462,382	\$327,214

Table 1. This table summarizes the FA electrolyzer facility's costs and performance.

The most challenging aspects of scaling up FA electrolyzers are achieving higher current densities, increasing membrane longevity, and maximizing CO₂ conversion rates. Researchers are currently working to alleviate these challenges and support industrial-level scale-up of this technology. Before constructing such a facility, the community will be made aware of the overall process and reasoning for the CO₂ conversion facility. Information will be disseminated via town halls and a social media campaign with the target message that this facility will serve the greater good locally and globally with regards to climate change. With a facility such as this, safety must always be a top priority, ensuring the proper PPE, safeguards, and standard operating procedures exist for control of hazardous energy, release of gasses, and chemical exposure. Above all, transparency must be maintained to ensure public awareness on facility operations and potential hazards.

INTRODUCTION

Background

Average global temperatures are rising due to greenhouse gas emissions according to the global scientific community. CO₂ emissions have risen by almost 100 parts per million since 1960, as depicted in Figure 1²⁸. This is just a portion of the rise in greenhouse house gas emissions since the industrial revolution. CO₂ and CO₂ equivalents released into the atmosphere exacerbate the natural greenhouse effect, which is how the atmosphere traps heat²⁰. To prevent the worst impacts of rising temperatures, CO₂ emissions must be lowered 45% by 2030 and net zero by 2050²¹. While renewable energy solutions become cheaper and more accessible each year, they alone cannot meet global energy demands; fossil fuels are still required to provide for a rising population's energy needs. Thus, the energy sector is looking to reduce emissions. One solution is carbon capture. For instance, flue

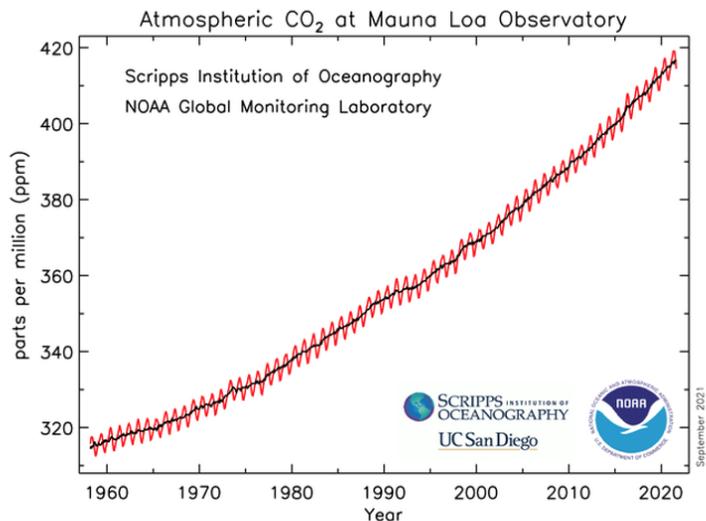


Figure 1. This graph depicts rising global atmospheric carbon dioxide concentration since 1960.

gas released from power plants can be captured using commercially available technology. Post-combustion is the most commercially available carbon capture method. CO₂ is removed from the flue gas at a low pressure to an amine-based solvent in an absorption column. The CO₂ rich amine solvent is then regenerated yielding a pure stream with CO₂ concentrations between 5-15% by volume¹⁷. This lower concentration will require a higher energy input for this method to work. Once captured, carbon may be stored or converted. While carbon capture and storage (CCS) strategies transport carbon to underground permanent storage, CCS demands large-scale storage technology and infrastructure with no product output. Carbon capture and utilization (CCU) strategies transform captured carbon into a marketable product. Wide-scale implementation of CCU technologies would lower global carbon emissions as well as generate a profit, so the energy sector is eyeing CCU for further research and development.

Task Statement

The natural gas industry, like the rest of the energy sector, is seeking CCU opportunities to lower its emissions. While natural gas power plants emit less carbon than coal power plants, they still emit an average of 898 pounds CO₂ per megawatt hour¹⁴. The objective in this design project is to reduce CO₂ emissions from a natural gas power plant emitting 900 lbs CO₂ per megawatt-hour (MWh) by creating a useful product from captured CO₂. The key things to consider are maximizing the amount of CO₂ converted, alternative designs, cost versus benefit analysis, intangible benefits, and safety³⁵. The following potential pathways will be discussed: building materials, algae, carbon black, chemicals, fuels, and an electrolyzer to generate a chemical product.

ALTERNATIVES

Considering Concrete

The production of cement, which is the binding agent for concrete, was responsible for 8% of the world's carbon emissions in 2016³⁰. Several technologies and processes tackle this problem: CO₂ mineralization, CO₂ curing, new binding agents, and researching ways to move away from concrete and instead towards building materials that absorb carbon. CO₂ mineralization converts CO₂ into aggregates that are used in concrete and asphalt. The benefit of

using CO₂ in the production of cement is that it allows the captured CO₂ that is absorbed into the concrete to be “locked in” and not emitted back into the atmosphere. CO₂ curing is the most popular CO₂-to-concrete technology because it reduces the time required to cure concrete. The curing technology saves construction companies money, which has led to large financial investments into research and development of CO₂ curing¹⁰. The downside to all CO₂ concrete technologies is that the CO₂ that is not absorbed by the concrete is emitted back to the atmosphere and there is no good way to recapture it.

Considering Algae

Algae is also a growing method to recycle CO₂ into a product. Many companies and projects are researching the best ways to grow algae for the future. For instance, ALGADISK, a project funded primarily by the EU, is working on a reactor concept that could allow for CO₂ to be used in both the liquid and gas phases, resulting in higher biomass output¹⁶. Algae has many potential uses, from biofuels to food products. However, most of these projects are on a relatively smaller scale. Problems arise when trying to recycle large amounts of CO₂ generated by power plants. Additionally, most algae strains can not grow under a 100% CO₂ concentration. The best growth rates require an approximate 5-15% CO₂ concentration⁵. Another problem is that in order to grow all of the algae needed to completely recycle CO₂ from a power plant, the farm would have to be extremely large due to the way algae is grown².

Considering Carbon Black

Carbon black is another form of heavily-researched carbon conversion. To convert CO₂ into carbon black, the CO₂ must first be captured and combined with, ideally renewable, hydrogen. This will form methane and water. After the water is removed, the methane will pass through a bubble reactor filled with liquid tin. Pyrolysis takes place inside the methane bubbles, which breaks down the methane into hydrogen and solid carbon²². This method of converting CO₂ into carbon black is revolutionary; however, more research is needed to implement this at full scale. Another drawback to this technology is that separating methane from water is very expensive. Because carbon black is a very useful substance, though, this research continues. Carbon black is used in the production of rubber, plastics, ink, and other products. Carbon black

can also be used to increase the carbon content in agriculture soil which is a growing problem in the agriculture industry³⁹.

Considering Chemicals

Carbon capture and utilization is attractive to the chemical industry as well. For example, CO₂ can be used to create polymers for polymer-containing products, such as plastics and adhesives³³. However, forming polymers involves oil refinement, which is energy intensive, further contributing to CO₂ emissions³⁸. In addition, CO₂ is used in the chemical industry as feedstock for various processes, relying on biological and chemical transformation to convert CO₂ into chemicals. The basis behind using CO₂ to make chemical feedstock lies within mimicking the natural process of photosynthesis, using catalysts for CO₂ hydrogenation⁴¹. From this, methanol (CH₃OH) is formed, which is the stepping stone to make various organic chemicals including ethylene, acetic acid, and formaldehyde³³. However, one disadvantage to this process is the formation of water. Water production can be counteracted through the CAMERE Process, also known as the hydrogenation of CO₂ to form methanol via a reverse-water-gas-shift reaction²³. The key things to consider in these processes is catalyst efficiency and the potential for byproduct formation. Additionally, carbon monoxide (CO) can be formed via electrochemical CO₂ reduction, and the CO can then be used further to create products such as alcohols and fuels⁸.

Considering Fuel

Converting carbon emissions into fuels is another popular field of research. If the power and industry sectors converted captured carbon into synthetic fuels, global net emissions would undoubtedly lower¹⁹. Carbon must first be converted into synthesis gas, or syngas, then converted into synthetic fuel. This is a cyclic process: carbon is recycled once by converting to syngas, then usually combusted further down the line. Therefore, emissions would still result when the fuel is combusted, for instance, in a vehicle. Given the smooth transition into existing markets, though, this option is attractive, while also saving emissions and generating a profit. By 2030, the revenue potential for CO₂ converted fuels estimate ranges from \$10 billion to \$250 billion dollars²⁹. That is, if supportive federal policy exists to back it. However, the carbon-to-fuels conversion process is energy-intensive. The Fischer-Tropsch process converts

carbon monoxide and hydrogen from syngas into methane and/or hydrocarbon chains that can be used to make plastics, fuels, and carbon black. The Fischer-Tropsch process can be challenging on a bench or lab scale due to the high temperatures and pressure required to convert the reactants into the hydrocarbon chains¹². Additionally, specialized technology such as fluidized beds are required for this conversion, which presents another challenge for the bench-top work space.

In general, when converting carbon to fuel, energy intensive steps lead to more carbon emissions. However, the hydrogen needed for carbon to fuel conversion could come from renewables. For instance, electrolysis technology extracts hydrogen out of water, which may be powered by renewables. Alkaline electrolysis (AEL) and proton exchange membrane (PEM) electrolyzers are commercially available. During alkaline electrolysis, an electrical current binds alkaline water molecules with electrons, resulting in the dissociation of hydrogen and hydroxide ions³¹. An electrolyzer is a commercial unit that performs electrolysis. Commercially available alkaline electrolyzers can also convert CO₂ to carbon monoxide and hydrogen, which may be transformed into fuels as well as chemical feedstock products. Furthermore, electrolyzers exist that convert CO₂ directly to formic acid (HCOOH), a marketable product with potential to serve net-zero emissions goals across multiple sectors.

FORMIC ACID ELECTROLYZER FACILITY CARBON CONVERSION SOLUTION

Formic Acid Uses and Market

While CO₂ to HCOOH via electrolysis is still being researched, this is a viable pathway to reduce emissions³¹. Commercial carbon to formic acid electrolyzers exist today. If scaled up to accept a power plant's emissions, an electrolyzer facility's formic acid output could be used for hydrogen storage, formic acid fuel cells, or

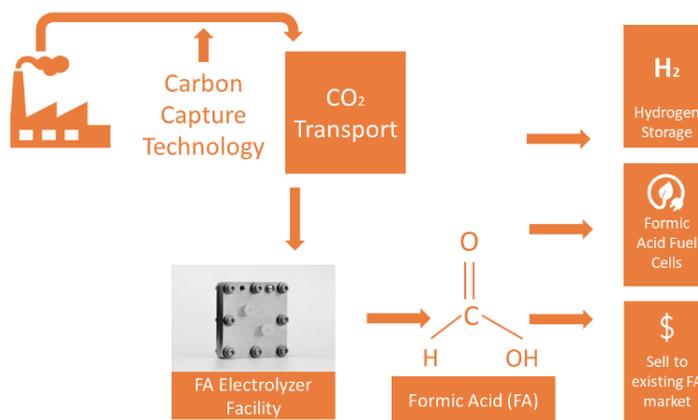


Figure 2. This image is an overall flow diagram depicting a formic acid electrolyzer facility carbon conversion solution.

sold to the existing formic acid market, as portrayed in Figure 2. The former two uses are particularly important for lowering global greenhouse emissions. Because of these sustainability-driven uses, the growing formic acid market, and the commercial availability of bench scale formic acid electrolyzers, this carbon conversion pathway was selected to perform experiments and develop a scaled-up solution.

First, formic acid is an attractive hydrogen carrier that is now being produced at a global scale of over 800,000 metric tons per year. Formic acid as a hydrogen carrier attracts industry, with its liquid phase existing in ambient conditions, high volumetric hydrogen density, and low toxicity¹⁵. These attributes lead to broad market opportunities and a wide range of applications. Hydrogen as a fuel offers net zero emissions; however, hydrogen becomes problematic and hard to transport because of its low volumetric density³. The majority of hydrogen's use with fuel requires pure hydrogen, but hydrogen is bonded with other elements which would require energy to separate the elements³. Although hydrogen is beneficial as an energy storage unit, one downside is that it is difficult to work with due to its sensitivity to temperature and flammability factor.

Second, researchers are studying how formic acid could be used in fuel cells. Fuel cells generate electricity by capturing energy from electrochemical reactions. Fuel cells have caught the eyes of the automotive industry, whose electric vehicles (EVs) could run on fuel cells as opposed to lithium ion (Li-ion) batteries, which are most common today. According to current research, fuel cells have a higher energy density than Li-ion batteries and take less time to charge. Formic acid fuel cells (FAFCs) in particular have a lower storage cost and are safer than hydrogen fuel cells. Needless to say, FAFCs are a rising field of research. FAFCs create electricity from formic acid oxidation and oxygen reduction. In the most recently studied FAFCs, when formic acid is fed into the anode, it is reduced into CO₂, hydrogen ions, and two electrons. When O₂ is fed into the cathode, it reacts with hydrogen and electrons to produce water. A polymer electrolyte membrane (PEM) facilitates the flow of protons from the anode to the cathode²⁶. The electricity produced could be used on a normal consumer basis (i.e. lighting one's home) or used to power a vehicle, as previously mentioned. While some carbon emissions result from the FAFC, the emissions would be significantly less than emissions from typical power

plants or gas vehicles. Moreover, researchers are studying methods to capture and/or use these emissions.

Third, formic acid can be sold in the existing market. The global formic acid market in 2018 was worth USD 430 million, but suffered a decline in 2020 due to the COVID-19 pandemic. The pandemic decreased the global market to USD 363.4 million, but the market has a compound annual growth rate (CAGR) of 4.49% and is expected to grow by 208.72 million USD between 2021 and 2025²⁷. Formic acid decreased in price from approximately \$700/ton to \$400/ton due to the pandemic, but increasing demand from different applications will drive the need for increased production²⁶. The most crucial market for formic acid is the Asia-Pacific market. China controls about 45% of the global formic acid market because formic acid is largely used in animal feed as an additive and preservative, and China is one of the largest meat producers in the world. Other applications include using formic acid in leather tanning production, textile dyeing, and as an intermediary in pharmaceuticals²⁷. The formic acid market is projected to grow as consumption of meat and poultry increases around the globe. Typically, formic acid has an industrial standard concentration of 85% but can even be sold as high as 99% for specific applications. The formic acid market, while being at a lower price currently, is set to rapidly grow by over 50% in the next five years due to increased demand for the chemical globally²⁷.

Bench Scale Experiment

The formic acid three-compartment electrolyzer utilizes water and CO₂ to generate formic acid (HCOOH) as the primary product, as shown in Figure 3. The electrolyzer has an active area of 5 cm², which is where the reactions take place. A BK Precision 1665 DC power converter provides a positive and

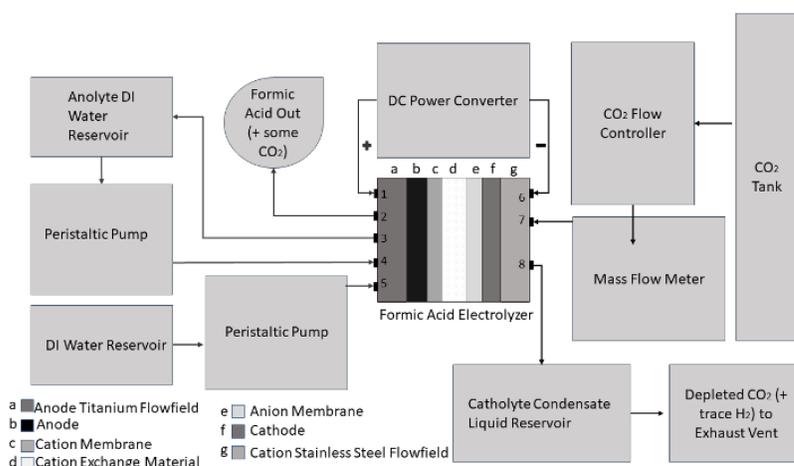
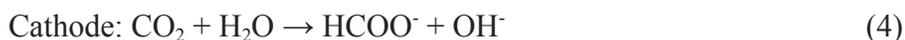
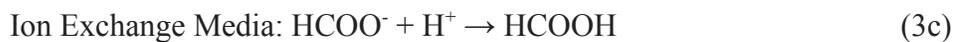
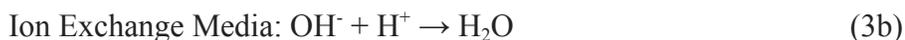
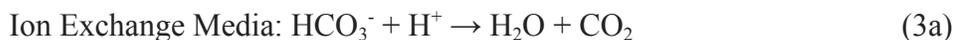


Figure 3. This image is a process diagram showing all inputs and outputs of the formic acid electrolyzer, including internal components.

negative charge to the anode and cathode exterior plates, respectively. On the anode titanium plate, a peristaltic pump pushes DI water into the plate's flowfield. Another peristaltic pump recycles water from anolyte DI water reservoir. Oxygen (O₂) exits the cell through this recycled anolyte DI water stream. Formic acid exits the electrolyzer through the anode flowfield. On the cathode stainless steel plate, a gas flow regulator pushes CO₂ from a tank, through the mass flow meter, and into the plate's flowfield. Depleted CO₂ exits the cathode plate into a catholyte condensate liquid reservoir. Various tubes and reducers make these connections. The electrolyzer cell consists of three primary internal components: electrodes, membranes, and ion exchange material¹¹. The anode's iridium oxide catalyst facilitates the critical water electrolysis equation (see equation 2) and transfers hydrogen ions (H⁺) to the Dupont Nafion® cation membrane, which then transfers H⁺ to the ion exchange material in the center of the cell⁴⁰. The cathode's bismuth oxide catalyst facilitates the critical formate reaction (see equation 4). Also on the cathode catalyst, a small amount of CO₂ reacts with water to form hydroxide ions (OH⁻) and carbon monoxide (CO), which exits the cell in the depleted CO₂ stream. Trace amounts of hydrogen (H₂) also exit the cell in the depleted CO₂ stream. The Sustainion® anion membrane transfers OH⁻ and formate (HCOO⁻) from the cathode catalyst to the ion exchange material. In the Amberlite® IR120 ion exchange resin beads, H⁺ reacts with HCOO⁻ to form HCOOH (see equation 3c) and water is formed (see equation 3b)⁴⁰. Also in the resin beads, bicarbonate (HCO₃⁻) reacts with H⁺ to produce water and CO₂ (see equation 3a) with ΔH_{rxn} of -15.5 kJ/mol. The overall reaction taking place is depicted in equation 1.



The intermediate reactions in each compartment of the electrolyzer are shown in equations 2 through 4.



Data & Analysis

DI water flow rate has a large impact on formic acid concentration⁴⁰. Thus, DI water flow rate was varied from 0.04 mL/min to 0.08 mL/min, where it was determined that a flow rate of 0.05 mL/min gave the highest concentration of formic acid, as seen in Figure 4. Three runs were completed per flow rate, each run lasting one hour. After each hour, the formic acid solution was collected. For the formic acid solution of each run, 800 microliters were pipetted into 2 mL vials. Each vial was diluted with 720 microliters of deionized water. Then, 200 microliters of ethanol was also added to each 2 mL vial, as well as 20 microliters of sulfuric acid. Both ethanol and sulfuric acid were 1:10 dilutions. Immediately after adding sulfuric acid, the vial was capped and sealed with a Teflon-lined septa. All vials were placed in a 60 degree Celsius water bath for 15 minutes. Once removed from the water bath, a 1 mL headspace sample was pulled from each 2 mL vial and injected into an Agilent 6850 gas chromatography (GC) machine, equipped with a flame ionization detector. Each sample went through a 0.5 mL sampling loop before reaching the Supelco column which is 2.1 mm by 2.4 mm and packed with 1% SP maintained at 130 degrees Celsius.

Each run was 4 minutes with a 1.3 minute retention time. Quantitation was

completed using an external standard curve with an R^2 value of 0.98. From the curves produced, the concentrations for each sample were found. The flowrate of 0.05 mL/min produced the highest concentrations of formic acid, so this flow rate was selected to study further.

At a flow rate of 0.05 mL/min, the average formic acid concentration was 30.2 g/L. The average formic acid production rate was 0.5 g/hr, and the average amount of CO₂ converted was 0.4 g/hr. Overall, the average CO₂ conversion rate was 11.6% (see Table 1). Experimental data was compared with other formic acid electrolyzer studies, and a 0.05 mL/min flow rate also produced the highest FA concentration for Yang et al⁴⁰. However, the experimental FA

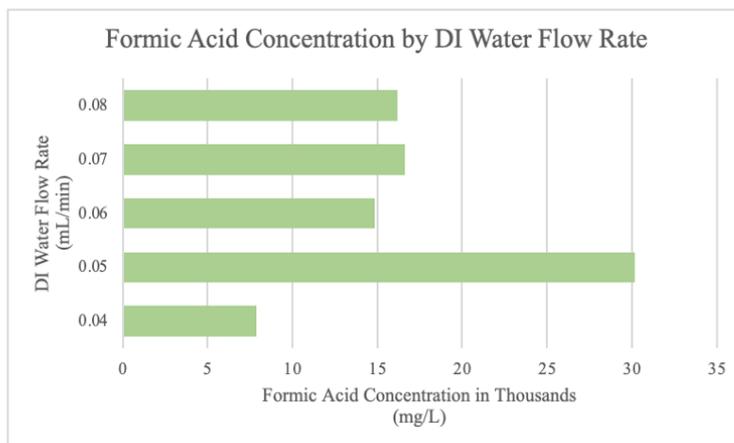


Figure 4. This graph depicts formic acid concentration vs DI water in flow rate.

production rate and energy efficiency were much lower than those recorded by Yang et al., despite multiple runs and troubleshooting⁴⁰. Therefore, the Yang et al. study and other CO₂ electrolysis studies were used for scale-up considerations.

0.05 mL/min Flow Rate				
Avg FA Conc. (g/L)	Aveg FA Production Rate (g/hr)	Avg CO ₂ Converted (g/hr)	Avg Energy Efficiency (%)	Avg CO ₂ Conversion Rate (%)
30.2	0.45	0.43	16.6%	11.6

Table 1. This table indicates some of the key average values for the formic acid electrolyzer running at a DI water inflow rate of 0.05 mL/min.

SCALE-UP & ECONOMIC ANALYSIS

Scale-Up Considerations

To accommodate 900 pounds of CO₂ per megawatt-hour produced by a natural gas power plant, as listed by the task statement, a large formic acid electrolyzer facility would need to be constructed. Using commercially available carbon capture and transportation technology, a natural gas power plant will send its emissions to a facility that will transform CO₂ to formic acid through a collection of scaled-up electrolyzers. Figure 2 depicts this overall carbon conversion process. While no full-scale CO₂ to formic acid facility currently exists, alkaline electrolyzers exist on an industrial scale. Thyssenkrupp, for instance, operates chloro-alkali electrolyzers with an active area of 2.72 square meters³⁶. Scaling was compared to those commercially available chlor-alkali electrolyzers. There should be a minimal cost difference between alkaline water electrolyzers and our formic acid electrolyzer²⁴. If a carbon-to-formic acid electrolyzer facility converted 100% of the 900 pounds per megawatt-hour exiting a natural gas power plant, the facility would require an active area of approximately 38,000 m². Looking at electricity to operate the electrolyzers and pumps alone, such a facility would cost \$862,000 per day to operate. A more feasible active area of 809 m² could convert 105,370 pounds of CO₂ per hour while accepting 150,528 pounds per hour, based on a single pass conversion rate of 70%. The 809 m² of active area needed also indicates that, based on a 2.72 m² active area per electrolyzer, 297 electrolyzers would be needed³⁶. At this rate, the facility would produce 50,000 kilograms of formic acid. These design parameters were determined based on a techno-economic analysis

(TEA) on CO₂ electrolysis completed by Shin et al³². Shin et al performed an extensive literature review on CO₂ electrolysis to produce four products, including formic acid. Because experimental data proved less efficient than other studies, scale-up calculations were modeled after the Shin et al TEA. Equations 5-10 depict the process used to determine these scale-up numbers, following the Shin et al. procedures, with the calculated values shown in Table 2. First, a reasonable formic acid target production rate was chosen (50,000 kg/day). From this production target, the total required amperage was determined from equation 5.

$$Current [A] = \frac{Target\ kg\ FA}{day} * \frac{1\ day}{86400\ sec} * 1000 \frac{g}{kg} * \frac{mol}{46.03\ g\ FA} * \frac{2e^-}{1\ mol} * 96485 \frac{C}{s} \quad (5)$$

Equation 6 uses the calculated total current to determine the required CO₂ flow.

$$Required\ CO_2\ Flow\ [\frac{kg}{day}] = Current [A] * \frac{1\ mol}{2e^-} * \frac{s}{96485\ C} * \frac{0.044\ kg}{mol} * \frac{86400\ s}{day} \quad (6)$$

This calculated flow was then scaled up for a single pass conversion rate of 70%, as seen in equation 7.

$$Adjusted\ CO_2\ Flow\ [\frac{kg}{day}] = Required\ CO_2\ flow\ [\frac{kg}{day}] * \frac{1}{0.7} \quad (7)$$

Equation 8 uses the total amps to determine what electrolyzer active area is needed to satisfy the chosen production target.

$$Active\ Area\ [m^2] = Total\ Current [A] * \frac{cm^2}{0.3\ A} * \frac{1\ m^2}{10^4\ cm^2} \quad (8)$$

From equations 9 and 10, one can determine how much power is required to operate the electrolyzer and both peristaltic pumps, respectively. A voltage value of 3.52 V was used for the electrolyzer based on the Yang et al. study, and 400 W was chosen for the pumps based on an ATO 12500 GPD high flow industrial peristaltic pump, resulting in a total daily operating rate of 204,974 kWh.

$$Power\ to\ Operate\ Electrolyzer\ [kWh] = \frac{Current [A] * 3.52\ V}{10^6\ W\ per\ MW} * \frac{1000\ kW}{MW} * 24\ hr \quad (9)$$

$$Power\ to\ Operate\ Both\ Peristaltic\ Pumps\ [kWh] = \frac{2 * 400\ W * 24\ hr}{1000\ W\ per\ kW} \quad (10)$$

Production Target (kg FA/day)	Total Current (A)	CO ₂ Inlet Flow (kg/day)	CO ₂ Entering FA Facility; 70% single pass conv. (kg/day)	Percentage of Power Plant Emissions Flow (%)	Electrolyzer Active Area (m ²)	Power to Operate Electrolyzer (kWh)	Required Power to Operate Pumps (kWh)	Cost for Power (\$/day)
50,000	2,426,080	47795	68278	3.06	808.7	204955	19.2	18448

Table 2 . This table depicts the scale up values for the formic acid electrolyzer implementation.

The facility would also release streams of unconverted CO₂, CO, hydrogen, and O₂, along with the formic acid product. These gas output streams will be sent to a pressure swing adsorption (PSA) process, where any unconverted CO₂ will be recycled back into the CO₂ input stream. The PSA would separate the hydrogen, CO₂, CO, and O₂ into their respective components to be used or stored. A series of adsorption bed pairs would cycle through the process to ensure a continuous separation of gasses. The process would involve pressurizing the chambers, feeding in the gas mixture, dropping the pressure in the chambers, and then removing the gas from the adsorption beds¹. The CO₂ would adsorb to a surface that reacts strongly to CO₂, such as activated carbon or a metal-organic framework. This will allow for the separation of the CO₂ from the other gasses and allow it to be recycled back into the input CO₂ stream for the electrolyzer. The remaining CO, hydrogen, and oxygen gasses will undergo this process again to separate them into their respective gasses to be used at a later time¹. Converting CO₂ to formic acid at a rate of 47,795 kilograms an hour, would have exhaust gasses being formed at a flow rate of 2,891.54 m³/hr. The PSA facility would have a capital cost of \$5,146,808.13 to build, while having an operating cost of \$1,561.43 per day. The liquid formic acid will be sent to a distillation process in order to refine the formic acid into whatever concentration would be needed for market. The capital cost for the distillation unit would be approximately \$1,200,000³².

Regarding electrolyzer scale-up in general, further research must be conducted on membrane scale-up and longevity. According to Dioxide Materials, the three-compartment design performance significantly decreases after 220 hours of operation, depleting completely after 1000 hours of operation⁴⁰. Operation at industrial-scale current densities also poses a challenge to electrolyzer scale-up. Maximizing CO₂ conversion rate also remains a field of further research. Additional studies must be conducted to extend the overall design life and

efficiency. Techno-economic analysis (TEA) was performed based on peer-reviewed scale-up models.

Process Flow Diagram

Figure 5 and Table 3 depict the process flow diagram and inputs and outputs for the full scale process.

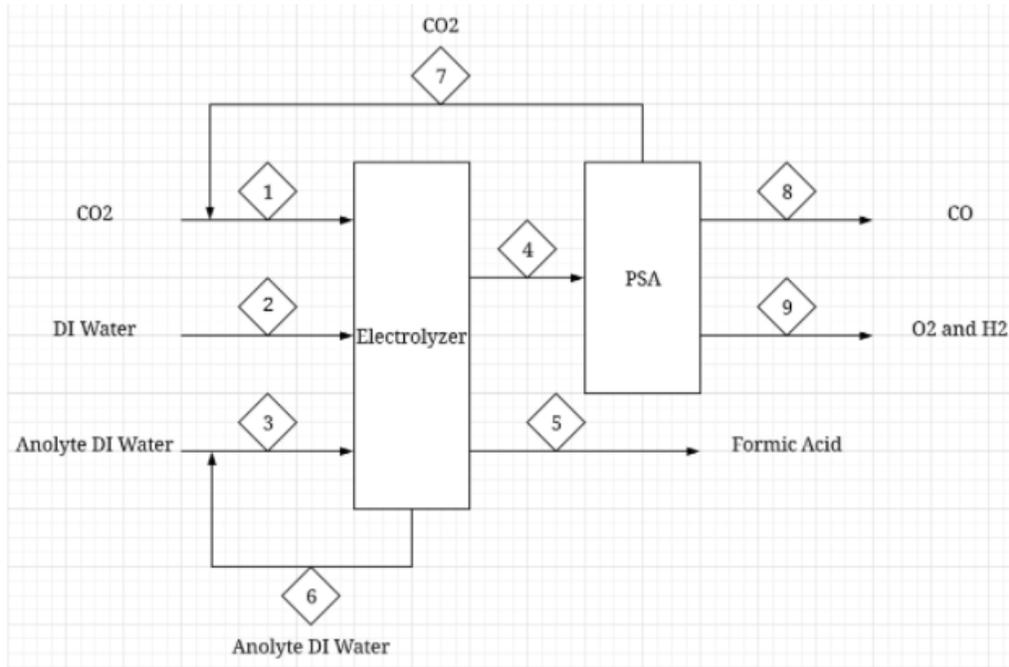


Figure 5. This process flow diagram depicts ins and outs of the electrolyzer

	1	2	3	4	5	6	7	8	9
CO ₂ (lbs)	4.39E3	0	0	1.46E3	0	0	1.46E3	0	0
DI Water (lbs)	0	3.57E3	0	0	0	0	0	0	0
Anolyte DI Water (lbs)	0	0	358.4	0	0	358.4	0	0	0
CO (lbs)	0	0	0	2.66E3	0	0	0	2.66E3	0
H ₂ (lbs)	0	0	0	29.93	0	0	0	0	29.93
O ₂ (lbs)	0	0	0	3.20E3	0	0	0	0	3.20E3
Formic Acid (lbs)	0	0	0	0	4.59E3	0	0	0	0
Temp (°F)	AMB	AMB	AMB	AMB	AMB	AMB	AMB	AMB	AMB
Time (hr)	1	1	1	1	1	1	1	1	1

Viscosity	?	?	?	?	?	?	?	?	?
BTU	-	-	-	-	-	-	-	-	-
Agitated	N	N	N	Y	Y	Y	Y	Y	Y
Flow Rate (m ³ /hr)	1.01E3	1.619	0.162	2.56E3	69.06	0.162	335.2	1.06E3	1.16E3

Table 3. This table indicates inputs and outputs of the process, depicted in figure 5

Techno-Economic Analysis

Capturing CO₂ and converting it into a useful product, formic acid in this case, requires the presence of an electrolyzer and time for the process to take place. If brought up to scale, this process could assist large plants in utilizing excess CO₂ and reducing the social cost of carbon. The social cost of carbon is the base cost of emitting a single ton of CO₂ into the atmosphere, and in 2020 it was estimated to be around \$42 and is expected to increase to \$69 by 2050¹⁴. Factors that affect whether this process would be cost beneficial for those plants to conduct rely heavily upon if the efficiency would hold up when brought up to a much larger scale. While the main goal of this process is to utilize excess CO₂ instead of it being released into the environment, other uses exist for the formic acid after it is produced, such as latex.

While conducting the analysis on this process, there are two areas to account for, technical and economic. The technical area will focus on the energy and mass required for the process, and the economic area will focus on the cost/profit margins. An obvious energy demand for the process to be completed is electricity. This demand will be determined based on how much electricity the process requires per day and using a cost of \$0.09 per kWh. There are several elements of the process that require electricity, including a power supply and two peristaltic pumps. It is determined that each pump draws 400 W of power. The amount of power and water used in the process will have a direct effect on its efficiency. Since there are many variables that can affect the total base cost of the process, it is important to identify how those changes will be made. Figure 6 shows the cost of the process under uncertainty.

An important economic factor of the process is the operational expenditure (OpEx), which depends heavily upon the price of electricity. Figure 7 shows a breakdown of the various operating costs for a formic acid electrolyzer facility. The cost of electricity plays a major role in the potential profit margins that could result from this process. In December 2021, the national

average cost of industrial electricity was about \$0.13/kWh, while in Louisiana and New Mexico it was 11¢/kWh and 13¢/kWh respectively¹³. The base cost used during this process is \$0.09/kWh. It is important to consider that, on a large scale, many plants act as their own power source, significantly cutting down on the cost of electricity. The values from equations 8 and 9 above were summed to determine the total kWh. The cost of electricity used during the process can be found using equation 11.

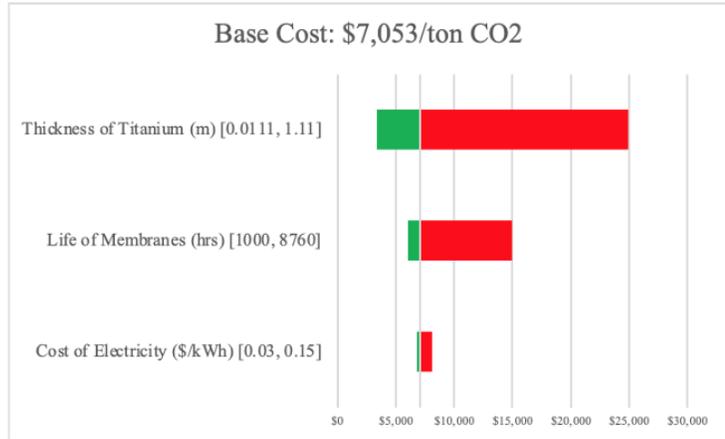


Figure 6. This tornado diagram shows the sensitivity analysis conducted for formic acid production via electrolysis.

$$Electricity\ Cost\ [\frac{\$}{day}] = \frac{\$0.09}{kWh} * (Elec.\ Power\ [\frac{kWh}{day}] + Pump\ power\ [\frac{kWh}{day}]) \quad (11)$$

Also capable of altering the economic viability of the process is the amount of product volume required. It should be noted that, as with the amount of power, the flow rate of DI water can be changed to alter concentration of the formic acid product, with lower flow rates favoring higher concentrations. Capital expenditure (CapEx) should also be considered, and the different costs associated with starting up a new facility are shown in Figure 8. Some important factors that go into CapEx is the initial funding for a potential plant to carry out this process. Within that includes the providing of equipment such as piping and other infrastructure.

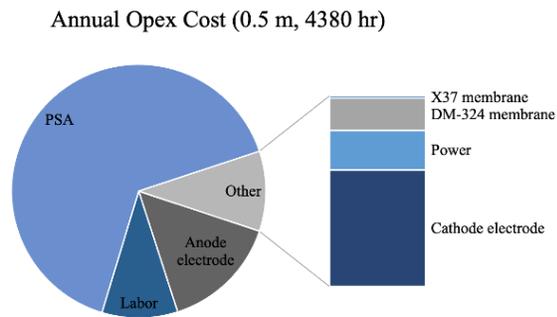


Figure 7. Above is a cost break down of the various operating costs for a formic acid electrolyzer with a plate thickness of 0.5m while replacing the membranes every 6 months.

Resource availability, such as CO₂, water, and electricity, should also be considered. While the bench scale process relied on a tank of CO₂, which inherently has a limited supply, a large-scale version of this process would likely not have this issue. As stated previously

regarding plants acting as their own power generators, in most cases after a purification process there would be no shortage of CO₂ available to use for this process due to the mass production of the gas from other processes. The final, and perhaps most important, aspect of economics is the profitability of the process. Using the amount of product produced and determining what the market rate is for the product, and comparing that to all expenses and other costs, the amount of profit can be determined.

The value of formic acid produced can be found by comparing all incurred costs to the production rate of formic acid, as in equation 12. Assuming the electrolyzer life will be 10 years, with membrane and electrode replacement needed every year, the initial cost will be \$863,074.60 with a yearly maintenance cost of \$569,765.

$$\$/\text{lb of Formic Acid} = \frac{\text{Total 10 year Capex and Opex cost}}{\text{lb of FA produced in 10 years}} \quad (12)$$

For industry to adopt this process, there must be a realistic opportunity to make a profit in some way. It is determined that for the process to be viable, the formic acid produced would have to be sold off at a rate of \$3.98/lb. For an electrolyzer with a titanium thickness of 0.5m the price of converting a ton of CO₂ is \$7,098.62. This is based upon a lifespan of 10 years. Accounting for a 12% rate of return would raise it to \$7,950.38 and would allow for a profit. The national average for formic acid selling on the market is usually around \$0.40-\$0.50/kg⁴. Since the formic acid produced in this process would have to be sold at a higher than market value, further steps would need to be taken. One of the options would be to try to appeal to buyers that this product produced by using excess CO₂ is environmentally friendly. Some companies may view that as an opportunity to build community relations by committing to reducing the amount of CO₂ released into the environment.

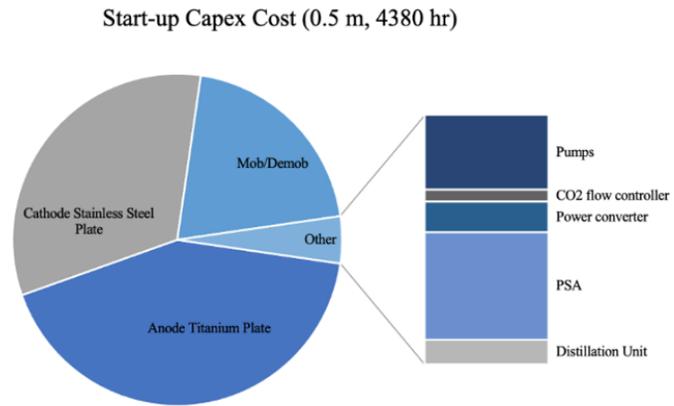


Figure 8. Above is a cost break down of the various capital expenditures needed to begin operation of a formic acid electrolyzer with a plate thickness of 0.5 m while replacing the membranes every 6 months.

Another more likely path to creating buyers would be to offer tax incentives to those willing to spend more on “clean” formic acid. There are several programs currently in place that encourage cleaner energy production and reduce emissions by offering different tax incentives. A 2018 expansion of the 45Q tax credit increased the financial incentives to \$35 per tonne of CO₂ used in other qualified uses other than storage³⁴. Currently, this incentive makes it economical to capture CO₂ emissions for several other processes, including natural gas, ethylene oxide, and ethanol production. Also, due to the previously mentioned estimated increase of the social cost of carbon over the next 30+ years, many industrial processes that are not currently profitable at this \$35/tonne rate would become cost effective with the higher rates.

Business Plan

The formic acid electrolyzer technology is viable for CO₂ conversion because the cost to convert CO₂ can be lowered in many ways such as stacking multiple membranes between metal plates, increasing efficiency, and elongating membrane life. The cost can further be lowered by the addition of government subsidies and programs. Along with producing a usable product converting CO₂ to formic acid is that once the CO₂ is converted it is completely removed from the atmosphere unlike fuels or other products that will re-release CO₂ back into the air. In today’s world, net-zero carbon emissions are the goal. Preventing CO₂ from entering the atmosphere and instead converting it into a usable product is currently an expensive process and having a product that re-releases CO₂ back into the air is not an ideal solution. Along with this revolutionary technology our team of professionals brings an in-depth knowledge of carbon capture and utilization and a can do attitude.

HEALTH, SAFETY, AND LEGAL REGULATIONS

Via the New Source Performance Standards (NSPS) established under section 111(d) of the Clean Air Act, CO₂ emission limits are based on the maximum allowable CO₂ emissions per unit of electricity. For natural gas power plants, this limit is 1000 pounds of CO₂ per megawatt-hour (MW-hr) of electricity produced⁶. Since the CO₂ used in the electrolyzer to create formic acid comes directly from the existing emissions of the power plant, control technology and corrective action plans should already be part of the power plant’s Title V permit. Since the electrolyzer process is releasing unconverted CO₂ and recycling it back with PSA, there should

be no potential for CO₂ emissions from the electrolyzer to exceed the 1000 pounds of CO₂ per MW-hr. If the CO₂ emissions from the power plant exceed this NSPS standard, the operators of the electrolyzer area need to be notified immediately. If possible, the electrolyzer should accept as much CO₂ as possible to try and get the emissions back below the NSPS standard while corrective action is being taken in the power plant itself. Along with this, the Louisiana Department of Environmental Quality (LDEQ) has the ability to implement more stringent regulations than the federal ones, which would be established in the permitting process if necessary as per RS 30:2060.1²⁵.

Carbon capture and storage is not currently regulated under federal environmental law. However, Louisiana has policies in place regarding the sustainability and environmental impacts of underground carbon capture technology⁷.

When considering bench scale and full scale design of the electrolyzer process, some safety aspects must be considered. Since the reaction occurs at room temperature, there is no suspected thermal risk with the process. When implementing the full scale design process into a plant, ensuring that the area is properly guarded with safe access is necessary. This could include guard railings surrounding the equipment that people should not enter without the proper approval for procedures such as maintenance. Since the process uses a power supply, it is necessary to ensure this equipment is running safely and securely. Monitoring of the power supply voltage and implementation of a voltage limit that should not be exceeded without corrective action may need to be implemented. If any maintenance work needs to be completed on the electrolyzer or power supply, it is necessary to have lockout/tagout (LOTO) procedures in place as part of the OSHA standard 29 CFR 1910.147 for the control of hazardous energy^{9,37}. These procedures should be reviewed annually and with any equipment changes. Any routine maintenance should have a set schedule. For maintenance that occurs on an as needed basis, proper procedures need to be in place to shut down the equipment if necessary and ensure all the proper PPE is worn dependent on the work.

Another safety aspect to consider with this process is the release of gasses from the process. H₂ is highly flammable; to mitigate this, the process should take place in an open, ventilated area. Additionally, excessive CO₂ exposure can cause headaches, dizziness, and

potentially asphyxiation at high concentrations. Handheld or in place gas sensors should be utilized so people entering the area are aware of any potential high concentrations of released gasses. Implementing control devices and constant compliance monitoring may be necessary. The long-term exposure limit and short-term exposure limit of CO₂ are 5000 ppm and 15000 ppm, respectively¹⁸.

This process generates formic acid, a flammable, toxic, and corrosive acid. The proper PPE must be worn at all times when handling this product, including but not limited to safety goggles, chemical gloves, and a chemical suit. A safety shower and eyewash station should be near this area and should be in an access area that does not have any trip hazards impeding someone's route to it. Routine inspection of the safety shower and eyewash is necessary to ensure it is working properly.

COMMUNITY RELATIONS

The community relations campaign should introduce what produces CO₂ and its effect on atmospheric warming. Community education and awareness should address the global climatic changes, national goals of net zero by 2050, and local sources of CO₂ emissions. The community should be made aware of new methods to capture and convert CO₂ and how the formic acid electrolyzer facility fits into this narrative. The community should be informed of the overall process of the facility and its by-products. This is clean energy and sustainable and the byproducts are naturally occurring and recyclable byproducts. Oxygen gas is classified as a hazardous waste because of its oxidative and explosion properties. The public should be presented with the current statistics on accidents including oxygen gas within the last 10 years. Emphasis should be drawn around two things: 1) Subsidized contracting groups would specialize in the capture and conversion of CO₂ into marketable items, and 2) installation and startup would require a separate facility to complete the work and would need maintenance every 6 months and monthly check-ins. Ultimately, community relations should portray the following core message: "Harnessing and converting this natural gas power plant's emissions supports local and national emissions-reduction goals. This formic acid electrolyzer facility is sustainable, while creating a marketable product from CO₂."

PUBLIC INVOLVEMENT

The campaign's strategy includes making the public aware by social media and town hall meetings to discuss the community's questions and concerns. The public should be involved in the decision-making process as well as considered when proposing CO₂ treatment options. Options should be weighed by all stakeholders. A decision on what CO₂ reduction method is made. From here, scheduling and setup planning can begin. Shipping and setup commence within the month, and the CO₂ reduction operation will be running with quarterly check-ins. Two months of social media posts are shown in Figure 9 and town hall meetings should be held to highlight the options of CO₂ treatment. Once approval is granted locally and publicly, implementation can begin. From this point, planning, shipping, and setup should take a month per facility. An industry spokesperson should be in monthly communication with the current governing official so that information pertaining to the CO₂ cleanup may be conveyed to the public. The community should continually be educated with global issues with CO₂, national CO₂ goals, regulatory CO₂ standards, and local CO₂ conditions.



Figure 9. These images show a community outreach strategy via social media and subscription articles.

CONCLUSION

With the ever-present issue of confronting climate change, recycling CO₂ from power plants is a rapidly growing area of industrial interest. With rising research and development on formic acid uses, a formic acid electrolyzer facility could become very relevant in the near future. Although the technology is not currently at a stage to accept all CO₂ emissions from a natural gas power plant, it has tremendous potential for improvements as research is continually occurring. Lowering the amount of CO₂ released to the atmosphere by any amount is a great step towards protecting our environment, and formic acid conversion can do that while benefiting the chemical market as well. Overall, CO₂ capture and utilization is an expanding practice, and industry and academia increasingly investigate the various opportunities each year.

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AUDITS

Economics and Business Plan Audit

**Economics and Business Plan Audit
Conversion of Carbon Dioxide to Formic Acid Project**



Auditor Name Deleted

Nice research report and pleased to see the focus on producing a usable product from captured CO₂ rather than simply sequestering it or using the CO₂ for enhanced oil recovery. The two companies in my equity coverage group with a CCUS component (Denbury & Oxy) are primarily focused on future projects that would capture, transport and/or sequester carbon dioxide.

Please see below for suggested additions/updates to the report.

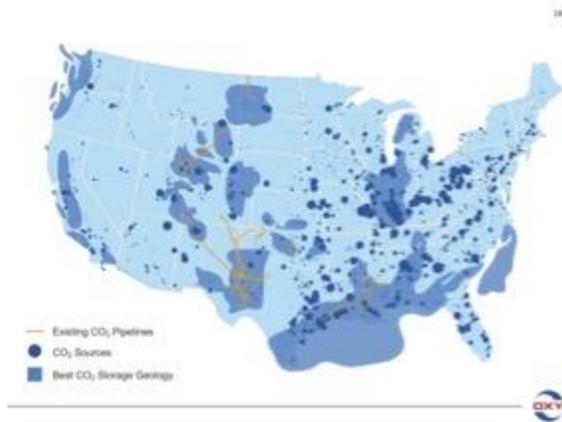
Consider adding a sensitivities table that provides formic acid profit margins and necessary product price for breakeven using various inputs (electricity prices, piping, etc). Focusing on varying electricity prices could be interesting during this time of significant increases in energy prices. For example, last month ConEdison requested an 18% increase in electric delivery rates for its New York customers.

May be helpful to the reader to provide the actual numeric data inputs used in the formula to arrive at the \$1.50/kg formic acid price required to breakeven/generate a profit. Also, please verify current large volume formic acid price (seeing some references to ~\$1,000/ton or ~\$1/kg, although these prices may be for different concentrations or outdated prices).

Unsure if this would be applicable, but perhaps discuss the options to source CO₂ on a moderate to large scale for development of products. Oxy provided a useful map last week in its investor presentation that highlights CO₂ sources and pipeline locations in the US.

<https://www.oxy.com/investors/stockholder-resources/lcv-investor-update/>

Team Number and University Deleted



Finally, providing some commentary regarding the formic acid market size and growth outlook could be helpful. Noticed these two recent report overviews that may provide some high level market data.
<https://www.globenewswire.com/news-release/2022/01/07/2363270/0/en/Formic-Acid-Market-Growth-Trends-COVID-19-Impact-and-Forecasts-2021-2026.html>
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Health and Safety Audit

*Auditor Name and
Affiliation Deleted*

March 30, 2022

**RE: Health & Safety Audit
Senior Design Project
Task 2: Carbon Conversion for Energy Transition**

Ms. Golson,

Thank you for the opportunity to review your team's paper on the analysis and bench scale experiment for Conversion of Carbon Dioxide to Formic Acid via Electrochemical Reduction in an Electrolyzer. I have evaluated your experiment for health and safety consideration. I appreciate the team's attention to detail in designing a safe experiment and incorporating safety considerations into the full scale implementation. I have the following recommendations for your consideration:

1. Since the full scale implementation would involve installation of an entire new process unit within a plant, the unit should be designed with operation and maintenance in mind – providing safe access for routine operation, visual inspection, and preventative and predictive maintenance activities will greatly reduce risk to those that interact with the equipment. For example, moving equipment should be properly guarded but allow appropriate access for maintenance such as lubrication; platform and access locations should be provided where needed and wide enough to accommodate people and tools. A thorough pre-startup safety evaluation will be critical.
2. Considering identifying the need for proper equipment checkout and testing upon startup and establishing robust operation and maintenance procedures and training.
3. The identification and inclusion of SDSs for the chemicals used is a good addition, but I recommend your team create a safe work procedure for operation of your bench scale model. You include the details and steps of how to properly operate the model in your paper, but condensing this into a procedure form will ensure all team members can safely operate the model. Your procedure should include: PPE requirements, operation steps, and identification of safety-critical considerations, such as the need to work under a hood for proper ventilation. I have included a sample SOP template that can be used at your discretion.

Overall, your team's health & safety considerations were well-thought-out and well integrated into your report. If you have any questions or would like more clarification of any of my recommendations, please don't hesitate to reach out.

Sincerely,

Environment, Health & Safety Manager

Team Number and University Deleted

Standard Operating Procedure

Procedure Name			
Department			
Equipment		Area	
Procedure #			

Purpose/Scope:

- List the purpose and/or scope here

EHS Considerations:

- PPE Required
 - List required PPE here
- Safety
 - List safety requirements, hazards, and precautions
 - Include special EHS material needs
- Environmental
 - List environmental requirements, hazards, and precautions here.

Tools and Materials:

- Safety Critical Equipment
 - List safety critical equipment here or simply write "none"
- List necessary tools and materials here

References

- List any references here, *delete this section if none*

Procedure Steps:

Safety Instructions: draw attention (example: by highlighting or using red text) safety critical steps and any error-likely situations and the layers of protection used to protect against those.

1. List procedure steps here.
- 2.

Review/Revision History

Rev #	Date	By Who	Description of Revision <i>Put "review only. no changes made" if SOP was reviewed without changes</i>	MOC # <i>If applicable</i>

Revision Date:		Reviewed date		Page 1 of 1
Revision #:		Reviewed by		

Legal and Regulatory Audit



We create chemistry

Auditor Name and Affiliation Deleted

March 31, 2022

Senior Design Legal and Regulatory Audit

Dear Project Team,

After auditing this report for Legal and Regulatory Issues, I believe it provides a conclusive explanation of regulatory concerns. Being that this project targets carbon neutrality in a power plant, it is subjected to NSPS. The logic for maintaining regulatory limits as stated in the "Health, Safety, and Environmental Regulations" section is solid; however, it is my recommendation to discuss compliance monitoring and/or feasible control devices here. Also, I would include a basic strategy to ensure the natural gas power plant is currently in compliance with the NSPS limit of 1000 pounds of carbon dioxide per megawatt-hour since the compliance logic for the electrolyzer is based on this notion. I appreciate the mention of the potential for the Louisiana Department of Environmental Quality (LDEQ) to implement more stringent regulations since this shows the team is aware of this potential and would establish a strategy as needed.

Regarding the OSHA standard, I believe the team thoroughly explained the health and safety risks of the electrolyzer. I would also mention that the lockout/tagout (LOTO) procedures need to be reviewed and validated annually or whenever an equipment change occurs, which would be tracked in a management of change (MOC).

I have no improvements to suggest to the section on gas exposure limits and PPE. The team did a good job evaluating the safety hazards and discussing risk mitigation for it. It can be said that the gas sensors will have alarms or flashing lights when the triggering concentration is met or that the safety shower and eyewash station will be painted a specified color for better visibility, but I believe this to be assumed in the design.

The community relations and public involvement is a well thought out section. While this does not directly fall under legal and regulatory issues, it could easily become a legal issue if the power plant were to not meet environmental, health, and safety standards. I firmly agree with the sentiment that the public should have full visibility - not only of sustainability efforts, but specifically the local emission amounts and trends. For a global reduction in emissions, it will require the public's support. The team has a superior strategy to involve the community. Their strategy allows for diversification in ideas and opinions, which will help push the campaign's success further.

Internal

Team Number and University Deleted

Overall, this report provides an insightful consideration to Legal and Regulatory Issues. Please let me know if you have any questions.

Regards,



Production Engineer

Sustainability in Production at

Former Environmental Engineer at International Paper

Internal