

# Design space for PEM electrolysis for cost-effective H<sub>2</sub> production using grid electricity

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## **ABSTRACT**

Proton Exchange Membrane (PEM) electrolysis is one of the most promising pathways for producing low-carbon hydrogen via electrolysis coupled with variable renewable energy (VRE). This study introduces a physics-based PEM electrolyzer model into an integrated design and scheduling optimization routine, allowing for a comprehensive evaluation of the impact of reactor level metrics (e.g., cathode pressure, current density) on the levelized cost of hydrogen (LCOH) across various cost, technology, and electricity supply scenarios. Benefits of static vs dynamic operation of PEM systems are outlined explicitly. The economic viability of a grid-based PEM electrolyzer producing 50,000 kg of hydrogen per day is assessed for both 2021 and 2035 projections. Results show that dynamic operation reduces the LCOH by 8% under the 2021 Scenario (4.98 to 4.57 \$/kg-H<sub>2</sub> at maximum current density 2A/cm<sup>2</sup>). Under 2035 price, cost and technology assumptions (maximum current density 4A/cm<sup>2</sup>), the LCOH ranges between 2.18-3.93 \$/kg-H<sub>2</sub> under static operation, and between 1.42-2.84 \$/kg-H<sub>2</sub> under dynamic operation, resulting in LCOH reductions of 20-50% depending on the electricity price profile. In addition, partial differential pressure mode with a cathode pressure of 5 bar was found to be the most cost-effective way to compress hydrogen to 30 bar in the 2021 Scenario, while full differential pressure mode is preferred in 2035 Scenarios. Finally, the study revealed that grid-based hydrogen production in 2021 falls short of meeting the carbon intensity (CI) criteria for the IRA 45V PTC, highlighting the need for dedicated renewable power sources for hydrogen electrolysis to qualify for the PTC. In the projected 2035 scenarios, even with an ambitious capital cost assumption of 200 \$/kW, the optimal LCOH is 1.22 \$/kg, failing to meet the DOE's Hydrogen Shot target of 1 \$/kg. These results suggest that capital cost reduction alone will not achieve low-cost electricity-based hydrogen production, emphasizing the need for further reductions in the cost of low-CI electricity to attain affordable and lower-carbon hydrogen production.

## **1. INTRODUCTION**

The rapid expansion of renewable energy sources like wind and solar is necessary to combat climate change. A primary challenge, however, is managing the intermittent nature of such energy in a cost-

effective manner. While commercially available Li-ion battery storage is becoming increasingly more cost-effective for storing energy over the period of hours, longer-term energy storage at scale remains challenging<sup>1</sup>. Hydrogen stands out as a promising low-carbon energy carrier that can provide a means of both long-term energy storage and long-distance energy transport, while serving as a vector for decarbonization end-uses where direct electrification remains challenging.

Among the various hydrogen production methods, electrolysis, whereby water is split into hydrogen and oxygen using electricity, offers a less carbon intensive alternative for producing hydrogen in comparison to steam methane reforming and autothermal reforming, particularly when driven by renewable power<sup>2</sup>. Current industrialized water electrolysis technologies for hydrogen production include alkaline and PEM electrolysis. While the alkaline process is more mature, PEM electrolysis is a particularly viable approach in this context, due to faster dynamical response times, wider current density range (0.1 – 6 A/cm<sup>2</sup> vs 0.1-0.4 A/cm<sup>2</sup> for alkaline), reduced corrosion, and fewer components<sup>3,4</sup>. However, the current capital cost of PEM electrolysis remains a substantial hurdle due to membrane and electrocatalyst costs, and contributes to the resulting levelized cost of hydrogen (LCOH) being considerably higher than that of steam methane reforming (SMR), the most widely used industrial process for the generation of hydrogen<sup>5</sup>. This cost disparity poses a major barrier to the widespread adoption of low-carbon electrolytic hydrogen. This disparity is diminishing however, due to substantial cost reductions in solar and wind electricity generation, as well as the upfront capital expenses related to electrolyzers<sup>4,6,7</sup>.

Operating electrically driven processes dynamically in a variable renewable energy (VRE) dominated grid has the potential to drastically reduce operational expenditures, since the process can exploit surplus electricity during low-price periods (e.g., when supply exceeds demand). This presents an interesting optimization problem, since oversizing the system to deal with times of peak production leads to an increase in capital expenditure (CAPEX) and could increase the overall levelized cost of hydrogen. For electrochemical systems, an additional trade-off to consider is the decreased energy efficiency at high current densities due to increasing overpotentials<sup>13</sup>. Several studies have therefore considered the design and operations optimization of electrolyzer systems, with respect to time varying electricity prices. Roh et al. formulated a total production cost minimization problem for a dynamically operating chlor-alkali electrolyzer<sup>14</sup>. Several techniques were used to make the optimization problem tractable, including a) discretizing the time domain, b) using a piecewise linear approximation for the quadratic power demand function, and c) utilizing a golden-section search (GSS) decomposition method on the electrolyzer size, resulting in a series of mixed-integer linear programs (MILPs). Results indicated that by oversizing a chlor-alkali electrolyzer and operating dynamically, the total production cost is reduced, provided that the specific capital costs of oversizing are low enough and the electricity price strongly fluctuates. Specific to PEM (studied in the current manuscript), Mallapragada et al. developed a techno-economic optimization model to evaluate the economics of a photovoltaic (PV) PEM system and found system configurations that result in LCOH that is competitive with hydrocarbon-based production methods<sup>15</sup>. In their work, a simple model for the PEM electrolyzer was used, without explicitly modeling the polarization curve (current density – voltage dependence). Ginsberg et al. considered the optimization of a PEM electrolyzer plants operating dynamically under California and Texas electricity price scenarios<sup>4</sup>. The study considered various operation modes of the electrolyzer, highlighting the temporal variations in current density, ranging from minimum to maximum levels. Results indicate that dynamic operation reduces the LCOH by up to 63% for 2020 prices and by up to 53% for projected 2030

prices compared to static operation. Furthermore, operation above  $2.5 \text{ A/cm}^2$  is justified at electricity prices below \$0.03/kWh.

The previous literature on PEM electrolysis plants has focused on physics-based models<sup>16,17</sup> and dynamic operation optimization<sup>4,18</sup>, but not considered both as part of a design and operations optimization framework. To date, numerous studies have focused on techno-economic assessments of hydrogen production via water electrolysis systems. Using physics-based and cost models, these investigations typically assess the LCOH under different operating conditions, identifying key drivers for cost reduction. For instance, Orella et. al presented a generalized techno-economic model that can be used for various electrolytic processes and used it to evaluate the economics of PEM electrolysis<sup>8</sup>. Badgett et. al provided a techno-economic assessment of the future production of hydrogen based on reduced capital cost as determined from a learning rate, manufacturing economies of scale, reduced material consumption, and improved electrolyzer performance<sup>9</sup>. James et al. provided a detailed techno-economic assessment (TEA) model specific to PEM systems, and conclude that the primary cost drivers are the electricity expenditures to run the electrolyzer and the capital cost of the electrolyzer<sup>10</sup>. Yates et. al evaluated the potential of PV-PEM Electrolyzer systems using a Monte-Carlo approach to identify key drivers for reducing the LCOH<sup>11</sup>. Finally, Hancke et al. used a techno-economic model to evaluate the economics of differential-pressure operation of a PEM electrolyzer system by comparing the cost of direct electrochemical compression with the cost of mechanical compression<sup>12</sup>.

To bridge this gap, in our current work a physics-based PEM electrolyzer model is developed, allowing for relating cell-level (e.g., membrane thickness) and plant parameters (e.g., operating temperature and pressure) with process economics. A TEA model is developed for costing the system, and an optimization framework is built to determine the optimal design and scheduling of a commercial scale PEM electrolyzer plant connected to the grid. We explicitly characterize the CAPEX-OPEX tradeoff under dynamic and static operation, and consider a limited treatment of the impact of differential pressure operation of the LCOH. Finally, our analysis considers the LCOH impacts of dynamic and static mode of operation considering today's grid as well as future renewables dominant power system with increased instances of zero-marginal cost electricity and price volatility.

## 2. METHODOLOGY

### Electrolyzer Model

The electrolyzer model uses thermodynamic and electrochemical principles to generate a polarization curve, which describes the electrolyzer's voltage as a function of current density. The number of electrons transferred directly translates to the moles of products generated in an electrochemical reaction, so the polarization curve is a measure of voltage that is required to produce a unit mass of hydrogen. The polarization curve is calculated as the sum of the thermodynamic equilibrium voltage, ohmic overpotential, and activation overpotential for a given electrolyzer technology and design.

The thermodynamic equilibrium voltage necessary to create the chemical potential to drive the electrolysis process, also known as the open-circuit voltage, is given by the Gibbs Free Energy state equation combined with the Nernst Equation (Eq. S2). Using the state equations allows exploration of much wider temperature and pressure ranges than typical empirical or analytical expressions<sup>17,19,20</sup>. In

this study, the model was used to evaluate the thermodynamic equilibrium voltage at an operating (cathode) pressure of 1 to 30 bar and an operating temperature of 80 °C. The ohmic resistance results from the resistances from the electron flow, proton flow, electrical contact in the membrane electrode assembly (MEA). The biggest contributor to the ohmic resistance in the PEM electrolyzer is the membrane <sup>19,21</sup>. Nafion 117 with a thickness of 178  $\mu\text{m}$  is assumed as the membrane technology for the current systems (2021 scenario) while thinner membranes (and thus lower ohmic resistance) is evaluated for the 2035 scenario (see Table 1).

The activation overpotential, also known as the kinetic overpotential, is the additional voltage that is needed to supply the activation energy to initiate the exchange of electrons in the electrochemical reactions <sup>22,23</sup>. The Butler-Volmer equation (Eq. S12) with the temperature-dependent exchange current density is used to calculate the activation overpotential as a function of current density.

Other models also include the concentration overpotential, which arises due to the overpopulation of reacting species near the electrodes interface. However, as long as the flow field is well designed for gas removal and the water circulation rate is high enough, the concentration overpotential can be neglected for PEM electrolysis <sup>20,21,23–26</sup>. For the purpose of making this model a computationally efficient techno-economic optimization tool, thermal behavior of the electrolyzer is not modeled and it is assumed that an excess amount of water circulates through the system to provide the necessary cooling to control the operating temperature. In addition, the degradation of the electrolyzer stack is not modeled explicitly, but is accounted for in the TEA model by assuming regular refurbishment of the stack every 10 years. A detailed description of the electrolyzer model is available in SI1.

Figures 2a) and 2b) show that the electrolyzer model can be calibrated to make accurate predictions of experimental data <sup>19,27</sup> under various cell configurations and thermodynamic conditions. The model parameters for the 2021 and 2035 technology assumptions were chosen to match the performance of benchmark systems, including commercially available stacks, as shown in Figure S3.

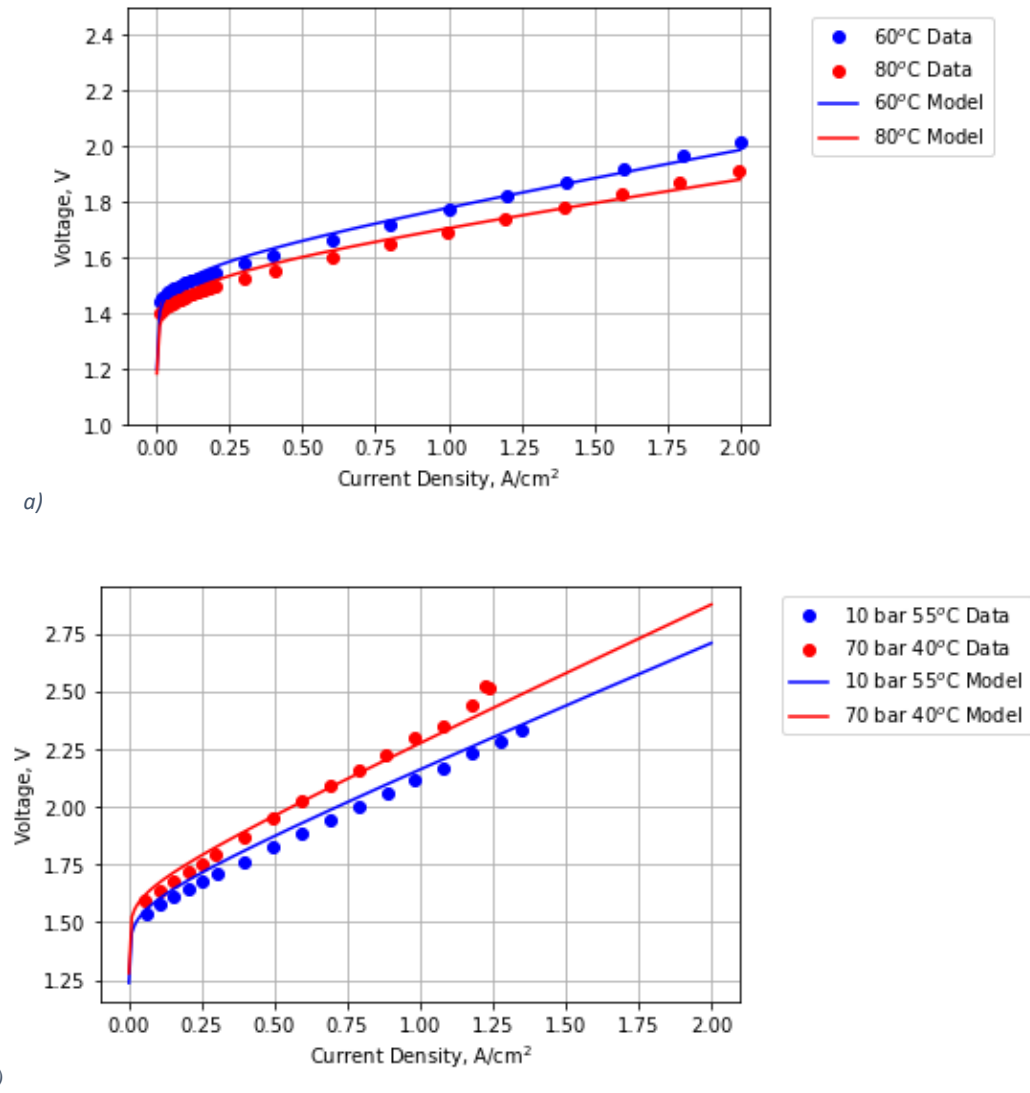


Figure 1: PEM polarization curves under varying thermodynamic conditions. a) Comparison of the electrolyzer model (line) with experimental data (circles) at 80°C (red) and 60°C (blue) at ambient pressure, data from <sup>27</sup>. b) Comparison of the electrolyzer model (line) with experimental data (circles) at (a) 55°C and 10 bar (blue); (b) 40°C and 70 bar (red). Data from (Marangio et al., 2009).

### Techno-Economic Analysis (TEA) Model

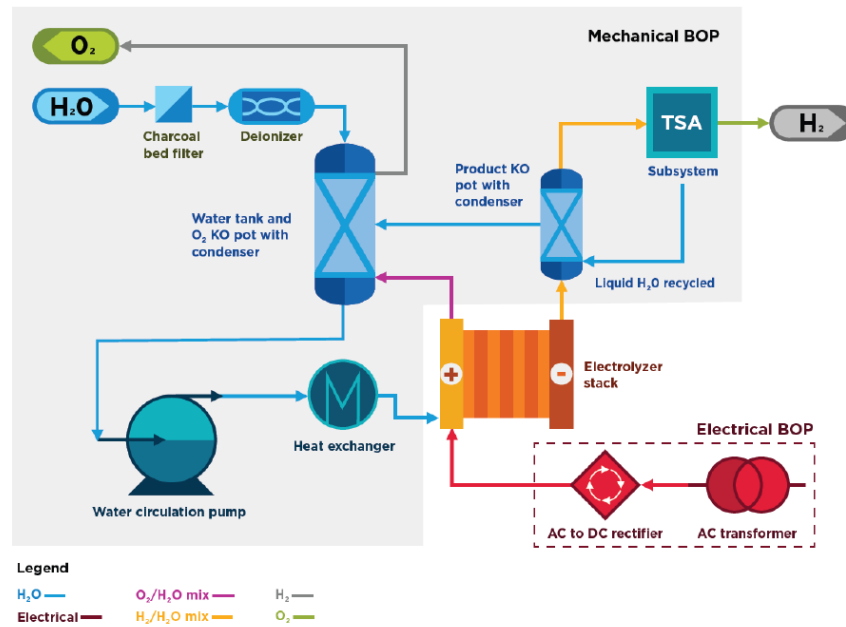
The TEA model used in this study is constructed by combining a physics-based PEM electrolyzer model with a system-level cost model for a PEM electrolyzer production plant shown in Figure 2

**Reference source not found.** The production plant consists of the electrolyzer stack, the mechanical balance-of-plant, and the electrical balance-of-plant.

The electrolyzer plant cost model is adapted from the Department of Energy (DOE) H<sub>2</sub> A model <sup>10</sup>, which is based on a detailed ASPEN-model based economic analysis and input from electrolyzer

manufacturers. The resultant TEA model serves to determine the net present value (NPV) for the overall production facility, which is subsequently used as the objective function in the integrated design and scheduling optimization problem (see section 0).

a)



b)

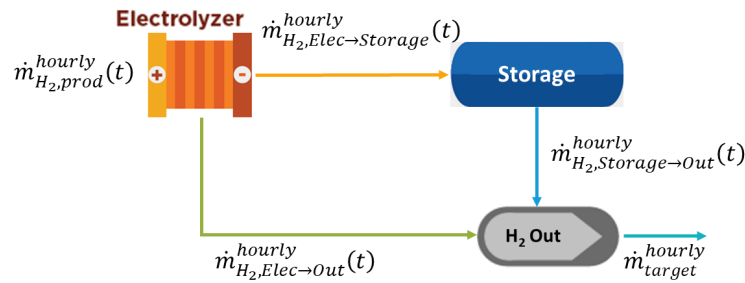


Figure 2: a) Schematic of the PEM electrolysis system showing the electrolyzer stack, the mechanical balance of plant, and the electrical balance of plant. Adapted from <sup>28</sup>. KO = Knock Out. b) Illustration of hydrogen flow paths from electrolyzer to storage to system outlet.

## Electrolyzer Cost Model

The electrolyzer cost model is adapted from the Department of Energy (DOE)'s H<sub>2</sub> A model <sup>10</sup>, which considers capital, operational, and overhead costs associated with different components of the electrolyzer plant, such as the stack, the mechanical balance-of-plant (BoP), and the electrical BoP. The stack cost is calculated based on the materials, labor, and capital costs associated with producing the stack. The mechanical BoP includes pumps, compressors, heat exchangers, water filtration system, knockout pots, and temperature swing adsorption system. The electrical BoP includes components such as transformers, rectifiers, switchgear, and control systems. The model also considers the cost of land, site preparation, and installation, as well as indirect costs such as project management, engineering, and contingency. The operational costs include the cost of electricity and feedwater that are necessary to operate the electrolyzer plant. To calculate the total cost of ownership of the electrolyzer plant, the net present value-calculation model is applied to the capital and operating costs over its expected lifespan. The model adopts the cost assumptions of the H<sub>2</sub> A model with modifications to adjust for inflation based on the Chemical Engineering Plant Cost Index (CEPCI) and for the 2021 reference dollar based on the U.S. Bureau of Economic Analysis's Consumer Price Deflator. A detailed description of the electrolyzer techno-economic model is available in SI3.

## Compression Cost Model

For an electrolyzer plant whose target delivery pressure is 30 bar, the output hydrogen stream can be compressed electrochemically, mechanically, or a combination of both. The higher capital and operating expenses resulting from electrochemical compression are accounted for in the electrolyzer model through a pressure adjustment factor sourced from the literature (see Figures S4a and S4b), with the result that 15-bar stack has roughly a 20% higher capital cost than a stack at ambient conditions <sup>29</sup>. For mechanical compression, a separate cost model is developed using the Department of Energy's Hydrogen Delivery Scenario Analysis Model (HDSAM). The description of the compression model is provided in SI4.

## Techno-Economic optimization framework

An optimization framework is formulated to ascertain the ideal plant sizing (specifically, the electrolyzer surface area and storage capacity), and optimal scheduling of the process-related operational variables (e.g., current density) over the course of year under time-varying electricity prices. The net present value of the plant cost is used as the objective function to be minimized. Since all scenarios considered in this work assume the same average daily production rate (see Section 3) this is equivalent to minimizing the LCOH. The LCOH is calculated by dividing the total discounted cost by the total discounted hydrogen production over the lifespan of the plant. The optimization problem is given by:

$$\text{minimize } NPV(C_{Total}) = \text{minimize } C_{CAPEX} + C_{storage} + \sum_{y=1}^{40} \left[ (C_{PR} + C_{UPR} + C_{fOPEX} + C_{vOPEX}) \frac{(1+\pi)^y}{(1+d)^y} \right] \cdot (1)$$

Where  $C_{CAPEX}$  is the capital expenditure (\$),  $C_{storage}$  is the capital expenditure for storage (\$),  $C_{fOPEX}$  are the fixed operating expenses,  $C_{vOPEX}$  are the variable operating expenses,  $C_{PR}$  is the planned replacement cost, and  $C_{UPR}$  is the unplanned replacement cost. Throughout, we assume a plant lifetime

of 40 years, an inflation rate ( $\pi$ ) of 1.9 %, and a discount rate adjusted for inflation ( $d$ ) of 8 %. The variable OPEX expenditures are determined by:

$$C_{vOPEX} = \sum_{d=0}^{364} \sum_{t=24d}^{24(d+1)-1} P_{elec}(t) \times Power_{op}(j_{op}(t)) + P_{elec}(t) \times U_{elec}^{hourly} \times \dot{m}_{H_2,prod}^{hourly}(t) + P_{water} \times U_{water}^{hourly} \times \dot{m}_{H_2,prod}^{hourly}(t). \quad (2)$$

Where  $P_{elec}(t)$  is the electricity price at time  $t$ ,  $Power_{op}(j_{op}(t))$  is the electrolyzer stack power which is a function of current density ( $j_{op}(t)$ ),  $U_{elec}^{hourly}$  and  $U_{water}^{hourly}$  are the hourly electrical usage and water usage respectively, and  $\dot{m}_{H_2,prod}^{hourly}$  is the hourly mass flow of hydrogen produced. In equation (2), we sum over all hours of the year, and  $P_{elec}$  is a yearly electricity price profile, discretized in hours. Note that by substituting  $C_{vOPEX}$  in the NPV optimization (1), we assuming that model dispatch over 1 year is duplicated over the other 39 years.

Under dynamic operation, a hydrogen storage device at the electrolyzer outlet (see Figure 1b) is modelled so that the system is allowed to store excess hydrogen during periods of low electricity prices and discharge it later, while meeting a fixed daily production rate. For simplicity, the storage can be charged or discharged, but not both at the same time. The storage model is described in more detail in SI6. A minimum current density limit for the electrolyzer is applied due to safety and efficiency considerations, and this is achieved via a big-M formulation (see SI Eqs S49 and S50). The optimization problem is discussed in more detail in SI3.

## Solution method

Two techniques are used to make the optimization problem (1) numerically tractable. Firstly, due to the nonlinearities present in the electrolyzer model equations, a piecewise linear approximation is used to determine the stack power density as a function of current density (see SI6, SI Figure S24). This improves optimization time and allows for implementation in Gurobi as the global optimization solver. Secondly, a golden section search (GSS) decomposition method is used to determine the optimal electrolyzer capacity (see SI6, and figures S23, S26 and S27). This results in the solution to many smaller MINLP subproblems with fixed electrolyzer capacity

The overall LCOH minimization problem is a mixed-integer linear program (MINLP), is implemented in Gurobi 9.5, and solved to a 1 % optimality gap. The optimization model consists of approximately 250,000 equations, 50,000 continuous variables and 60,000 binary variables. Typically, about 10 GSS iterations are required to reach a convergence tolerance of 1000 kg H<sub>2</sub>/day .

Since the electrolyzer capacity is fixed in the MINLP subproblem, the objective function in the inner loop of the GSS algorithm is simplified as:

$$\text{minimize } C_{storage} + \sum_{y=1}^{40} \left[ C_{vOPEX} \frac{(1+\pi)^y}{(1+d)^y} \right] \quad (3)$$

Where  $C_{storage}$  is the cost of the storage device, which increases linearly with storage inventory.

## Grid electricity input data sources

In order to assess the impact of electricity price on the levelized cost of hydrogen (LCOH) in the 2021 Scenario, historical hourly locational marginal prices (LMP) from Southern California in 2021<sup>30</sup> are used in the cost model. In addition, a typical grid interconnection cost of \$0.03/W DC is added to the total electricity price.

For the 2035 scenario, multiple regional system electricity price projections for 2030-2040 from the National Renewable Energy Laboratory (NREL) 2021 Cambium Project<sup>31</sup> and the FLECCS Program<sup>32,33</sup> are used in the simulation. Brief descriptions of each of the electricity price scenarios used in this study are available in SI5.

## Scenarios evaluated

Throughout this work, we employ a case study that uses grid electricity to supply 50,000 kg of H<sub>2</sub> per day, generated at an outlet pressure of 30 bar, in line with central, large, plant size H<sub>2</sub> production system described by (James et al., 2013). The electrolyzer temperature is assumed to be 80 °C. The minimum operating current density,  $j_{min}$ , is assumed to be 0.5 A/cm<sup>2</sup>. The cathode and anode exchange current densities are assumed to be  $2.3 \times 10^{-3}$  and  $3.0 \times 10^{-6}$  A, respectively. Two modes of operation are considered: static and dynamic. Under static operations, the electrolyzer is constrained to operate at a constant nominal current density, producing a constant stream of hydrogen at the outlet. In this mode of operation, the electrolyzer plant is sized to exactly match the demand rate, thereby removing the need for a storage facility. In dynamic operations, the electrolyzer is allowed to vary the operating current density subject to technical and operational constraints. In addition, a storage facility is necessary to insulate the consumers from variable production of dynamic operations.

## Cost and technology assumptions

A sensitivity study is carried out in order to assess the impact of different cost drivers on the LCOH. The sensitivity parameters include different technology, cost, and price assumptions on the basis of available calculations and projections in the literature. Table 1 shows the electrolyzer model parameters assumed for the 2021 and 2035 Scenarios. Table 2 shows the cost model parameters assumed for the 2021 and 2035 Scenarios. In section 3.D we also consider modifying the cathode pressure.

**Table 1: Electrolyzer Model Parameters for various technology scenarios.**

| Parameter                        | 2021 Scenario        | 2035 Scenario        | Unit              |
|----------------------------------|----------------------|----------------------|-------------------|
| Membrane Thickness               | 178                  | 127                  | μm                |
| Cathode Exchange Current Density | $2.3 \times 10^{-3}$ | $2.3 \times 10^{-3}$ | A/cm <sup>2</sup> |
| Anode Exchange Current Density   | $3.0 \times 10^{-6}$ | $3.0 \times 10^{-6}$ | A/cm <sup>2</sup> |
| Temperature                      | 80                   | 80                   | °C                |
| Cathode Pressure                 | 30                   | 30                   | Bar               |

|                                    |         |     |                   |
|------------------------------------|---------|-----|-------------------|
| Anode Pressure                     | 1       | 1   | Bar               |
| Maximum or Nominal Current Density | 1,2,3,4 | 4   | A/cm <sup>2</sup> |
| Minimum Current Density            | 0       | 0.5 | A/cm <sup>2</sup> |

**Table 2: Key technology cost assumptions across the two technology scenarios. See SI3 and SI4 for details. eBoP is the electrical balance of plant consists of an AC transformer to change the voltage of the AC power supply from the grid and a rectifier to convert the AC current to a DC current. The mechanical BoP (mBoP) consists of subsystems that perform secondary functions to support the electrolyzer stack <sup>34</sup>.**

| Parameter                                   | 2021 Scenario | 2035 Scenario | Unit                         |
|---------------------------------------------|---------------|---------------|------------------------------|
| Electrolyzer Stack Cost                     | 2             | 0.7           | \$/cm <sup>2</sup>           |
| mBoP Cost                                   | 117           | 54            | \$/((kg H <sub>2</sub> /day) |
| eBoP Cost                                   | 126           | 80            | \$/kW electric               |
| Total CAPEX                                 | 1,006         | 421           | \$/kW input                  |
| Storage CAPEX                               | 500           | 300           | \$/kg                        |
| Grid interconnection cost                   |               | 30            | \$/kW                        |
| Discount rate for annualizing capital costs |               | 8%            |                              |

### 3. RESULTS AND DISCUSSION

In this study, we explore three main aspects that influence the Levelized Cost of Hydrogen (LCOH) for a PEM electrolysis plant: 1) static vs dynamic operational modes considering design and operational cost trade-offs, 2) system design parameters like maximum current density and differential pressure operations and 3) the influence of different electricity price profiles and technology cost assumptions. We also include a brief discussion of the potential cost and design implications of the H<sub>2</sub> production tax credits available in the U.S. as part of the Inflation Reduction Act of 2022 with a more thorough discussion on this subject available elsewhere <sup>35</sup>.

#### A. Impact of Dynamic vs Static Operations on LCOH Under 2021 Technology and Cost Assumptions

Under 2021 cost and technology assumptions noted in Table 1-2, dynamic operations reduce the LCOH by 5 - 11% compared to the equivalent static operations delivering the same hourly hydrogen production rate of 50 tonnes/day (Figure 3). The cost savings is partly attributed to the reducing variable operating costs (mostly power purchase costs), which account for up to 82% of the LCOH shown in Figure 3. Under dynamic operations, the cost-optimal electrolyzer dispatch results in increasing hydrogen production (by increasing current density) when the electricity is cheap and decreasing production when the electricity is expensive, as highlighted in temporal patterns in current density and electricity prices in Figure 3. By contrast, the electrolyzer in a static mode produces a constant stream of

hydrogen by operating at a constant nominal current density of 2 A/cm<sup>2</sup> regardless of the fluctuations in the electricity prices.

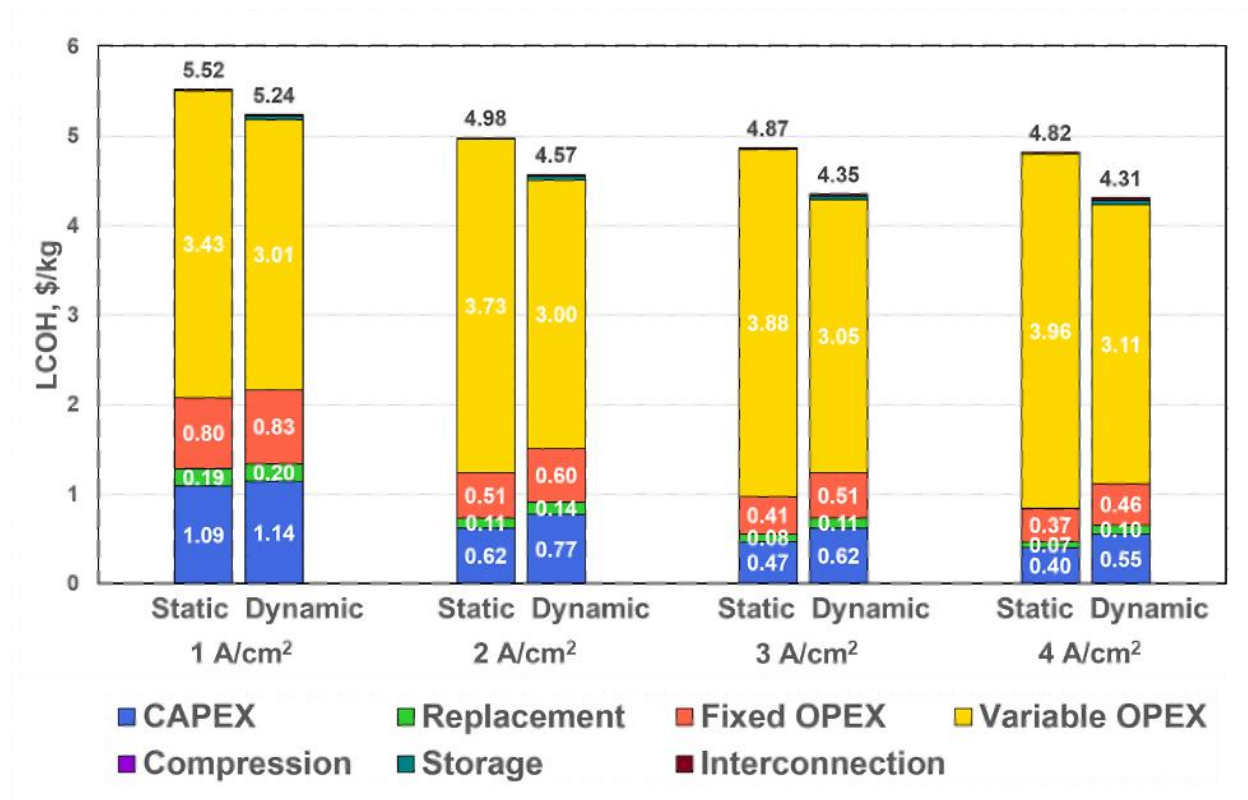


Figure 3: Optimized LCOH contributions under 2021 cost and technology assumptions. LCOH has units \$/kg-H<sub>2</sub>. For static operation, the current densities mentioned refer to the unchanging operation at the specified current density. Under dynamic operation, the current densities indicate the maximum permissible current density. The electrolyzer is given the flexibility to operate below this maximum limit on an hourly basis. Electricity price scenario based on data for location in Southern California.

Although operational costs are lower under dynamic operation as compared to static operation, in order to achieve the same production target, the lower utilization rate in a dynamic mode is compensated with a larger production capacity (and thus greater fixed costs). For example, across the current density scenarios evaluated in Figure 3, the installed production capacity is 4-38 % greater than the average H<sub>2</sub> production rate. Furthermore, investment in a compressed H<sub>2</sub> storage facility is necessary to buffer out the mismatch between a variable supply and a fixed demand. As shown in Figure 4, to compensate for this mismatch the electrolyzer produces more hydrogen at high current density and charges this to storage during low-price hours. During high price hours, hydrogen production is reduced by lowering current density and storage is discharged to meet the demand. There is also evidence of long-term hydrogen storage, where the longest continuous period where storage remains above 20% of the capacity is 46 days (starting on day 30). Although H<sub>2</sub> storage is additionally needed in the dynamic operation case, the incremental cost of storage is relatively minor (< 2%) compared to the other variable

and fixed cost of the electrolyzer. This is true even while considering relatively expensive above-ground  $H_2$  storage and will even more attractive in case salt cavern based  $H_2$  storage that has much lower capital costs<sup>36</sup>, might be feasible.

Despite an increase in the electrolyzer nameplate capacity and the addition of the storage facility, the LCOH of dynamic operations is less than that of static operations. For the case study simulated for Southern California, oversizing the plant capacity by 33% with a nominal/maximum current density of  $2 \text{ A/cm}^2$  led to an 8% decrease in LCOH, from  $4.98 \text{ \$/kg}$  to  $4.57 \text{ \$/kg}$ . Even with dynamic operation, the LCOH values in Figure 3 for current technology and grid conditions are still much greater than the cost of  $H_2$  production from fossil options, which range between  $0.5 - 1.7 \text{ \$/kg}$ , depending on natural gas prices (International Energy Agency, 2021). This suggests that further reductions in capital costs and electricity prices are needed to make  $H_2$  production via PEM electrolyzers an economical option.

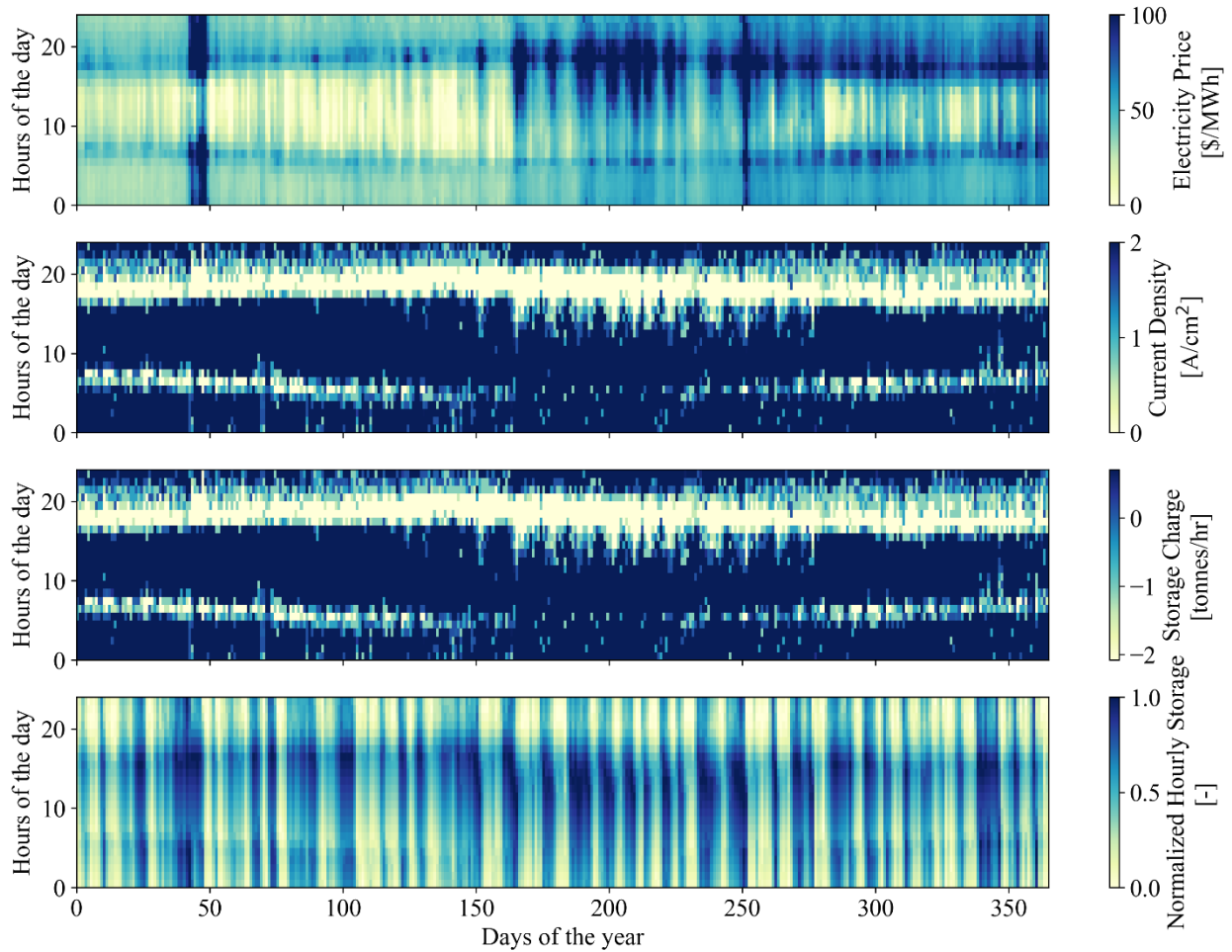


Figure 4: Optimal plant dispatch under 2021 cost and technology assumptions under dynamic operation. The maximum current density is set to  $2 \text{ A/cm}^2$ . Negative values of storage charge refer to discharge.

## B. Impact of Operating Current Density Under 2035 Technology and Cost Assumptions

Operating at a high current density is crucial to reducing the LCOH in both static and dynamic operational modes. This is because higher current densities enable more hydrogen production per unit of cross-sectional area of electrolyzer cells, which in turn reduces the capital expenditure and other fixed costs associated with scaling the plant size (as depicted in Figure 3). Although variable operating expenses increase with increasing current density due to efficiency losses (see Figure 3), an overall cost reduction of 9.8% to 12.8% is achieved by increasing the current density from 1 A/cm<sup>2</sup> to 2 A/cm<sup>2</sup>. The benefits of dynamic operations also increase with increasing current density. Since the cell is less efficient at higher current densities, a greater reduction in variable OPEX is achieved when the cell operates at part-loading. As shown in Figure 3, the LCOH is reduced by 5% at 1 A/cm<sup>2</sup>, 8% at 2 A/cm<sup>2</sup>, which is the typical limit of present systems, and 11% at both 3 and 4 A/cm<sup>2</sup> when transitioning from static to dynamic operations. A corollary observation from this result is that commercially alkaline electrolyzers that have much lower current density ranges (typically around 0.1-0.4 A/cm<sup>2</sup>) may be less valuable to exploit electricity price volatility for low-cost H<sub>2</sub> production as compared to PEM systems.

## C. Impact of Electricity Price Profile on LCOH Under 2035 Technology and Cost Assumptions

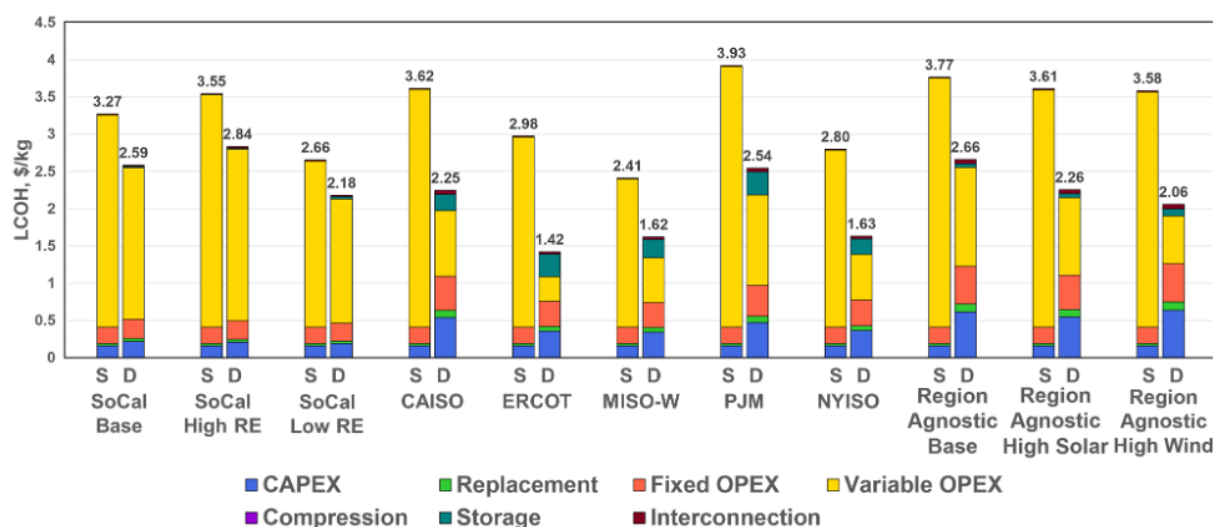


Figure 5 LCOHs with different electricity price projections with 2035 technology and cost assumptions. A nominal/maximum current density of 4 A/cm<sup>2</sup> is used for the static/dynamic operation scenarios.

Here we explore the LCOH impact of future projected cost and performance of electrolyzers and electricity price scenarios on the LCOH under static and dynamic operations. First, electrolyzers, particularly PEM systems, are relatively nascent technology, with global installed capacity at 90 MW as of 2020<sup>37</sup>. Therefore, many studies expect that the capital costs for electrolyzers will significantly decrease in the future due to advancements in flow fields, membrane electrode assembly (MEA), labor,

economies of scale, quality control, and larger cell areas<sup>10,38,39</sup>. Studies suggest that the learning rates for electrolyzers are comparable to those for solar PV, between 16% and 21%<sup>40,41</sup>. Additionally, IRENA 2020 predicts that a cost reduction of more than 40% can be achieved by 2030. With an annual decline rate of 4.8%<sup>6</sup>, the cost of an electrolyzer stack is expected to decrease from 2.0 \$/cm<sup>2</sup> in 2021 to 0.7 \$/cm<sup>2</sup> in 2040. As the technology scales up, capital costs for balance-of-plant equipment are also expected to decrease, resulting in a total capital cost of 421 \$/kW for the 2035 scenario (Table 2).

Second, technology improvements are expected to enhance the operational performance of electrolyzers. The development of thinner membranes and optimized catalyst layers can improve electrolyzer performance while also reducing the cost of the electrolyzer stack. While it is difficult to predict which technological advancements will be successful, it is anticipated that future electrolyzers will be able to use thinner membranes without compromising structural integrity and crossover resistance.

Third, as power costs are the biggest contributor to the total cost of electrolytic hydrogen production, we evaluated the impact of different future electricity price scenarios to in order to understand the range of possible LCOH values under different grid contexts. As previously mentioned, the projections include 16 different hourly prices series forecasts for 2030-2040 from the NREL 2020 Cambium Scenarios and the FLECCS Program. The five NREL Cambium Scenarios use the Regional Energy Development System (ReEDS) model to simulate the electricity price series under different assumptions of renewable energy penetration (high/low renewable energy cost) and policy support (decarbonize 95% by 2035 or 2050). The price series data are obtained for 2040 in a balancing region corresponding to Southern California. In addition to the NREL Cambium Scenarios, the NREL forecasts from the FLECCS program<sup>32</sup> also provide hourly price series data for 2035. These data are simulated with the AEO2020 reference natural gas prices and different carbon tax assumptions (\$100/tonne or \$150/tonne), for each of the major grid operators in the United States, including CAISO, ERCOT, MISO-W, NYISO, and PJM. In addition, the Princeton data set offer region-agnostic hourly price series data for 2030, simulated with the assumptions of \$60/tonne carbon tax, AEO2020 REF natural gas prices, and mid/low costs for solar and wind clusters located across the country. These different electricity price assumptions allow for a comprehensive understanding of the potential range of LCOH values, as electricity costs play a significant role in determining the total cost of hydrogen.

According to the simulation/optimization results depicted in Figure 5, dynamic operations become more valuable with future cost, technology, and electricity price assumptions. In comparison to the 5-11% reduction in the 2021 Scenario, dynamic operations in the 2035 Scenarios lead to a 20-50% reduction in LCOH from the equivalent static operations baselines. Oversizing plant capacity up to four times the target production rate enables this cost reduction. Oversizing plant capacity is advantageous due to several reasons. The future grid systems with more renewable power integration are expected to have higher price volatility (see section SI5 and Table S8 in SI), which provides more opportunities to dynamically vary the electrolyzer power loads. Additionally, lower capital costs reduce the opportunity cost of oversizing the plant capacity.

Although there are clear benefits of dynamic operation, there are technical and operational challenges to be addressed to realize the full potential of dynamic operations as modeled in this study. For instance, hydrogen crossover from cathode to anode, which is likely to be more prevalent at lower current densities (near zero) may limit the duration up to which the electrolyzer can continue to operate

low current densities. In addition, high-frequency cycling may limit the number of cycles due to degradation, and the response times of the balance-of-plant equipment may not be fast enough to exploit every trough in the price fluctuations. Nevertheless, the trend is clear that dynamic operations will play an increasingly important role in reducing the cost of hydrogen production in the 2035 Scenarios with lower capital costs, cheaper and more volatile electricity prices, and technological improvements.

Overall, Figure 4 shows that the cases simulated with an assumed capital cost of 421 \$/kW had an LCOH ranging from 1.4 to 3.6 \$/kg, with an average of 2.5 \$/kg. Although this average price is still higher than fossil-based production costs of 0.5 to 1.7 \$/kg, it falls significantly short of the DOE's Hydrogen Shot target of 1 \$/kg. Even with the super-optimistic assumption of a 200 \$/kW capital cost, the lowest LCOH obtainable from the simulation was 1.22 \$/kg. These results suggest that capital cost reduction alone will not achieve low-cost electricity-based hydrogen production. Instead, investments in low-cost renewable power and breakthrough technology research are also needed to achieve clean and affordable hydrogen production.

#### **D. Differential Pressure Operations**

Most of the commercial electrolyzers available in 2021 come with a standard feature of 30 bar outlet pressure. Differential pressure operation is one way to achieve this, where the anodes remain at ambient pressure (1 bar), and the cathodes are pressurized to 30 bar. Another method is to carry out electrolysis at ambient pressure on both anode and cathode sides and then use mechanical compressors to pressurize the produced hydrogen. A hybrid solution is also possible, where the electrolysis occurs at an intermediate pressure on the cathode side, and the mechanical compressors then pressurize the hydrogen to the desired level. Considering the cost, price, and technology assumptions of 2021, our results show that the most economical option is the hybrid solution with a cathode pressure of 5 bar, as depicted in Figure 6. As the cathode pressure increases, the thermodynamic efficiency loss and higher pressure requirements increase operating expenses and capital costs. In comparison to ambient pressure operations, the increased operating expenses and capital costs required for differential pressure operation at 5 bar are more than offset by avoiding inefficient mechanical compression at low pressure ranges (compression ratio of 5 from 1 bar to 5 bar). Under the 2035 assumptions, full electrochemical compression with a differential pressure of 30 bar is projected to be the most cost-effective mode of operation, eliminating the requirement for bulky and noisy mechanical compression systems. This is because the impact of higher pressure operation is mitigated by the lower average electricity price and electrolyzer capital costs.

## 2021 Scenario

## 2035 Scenario

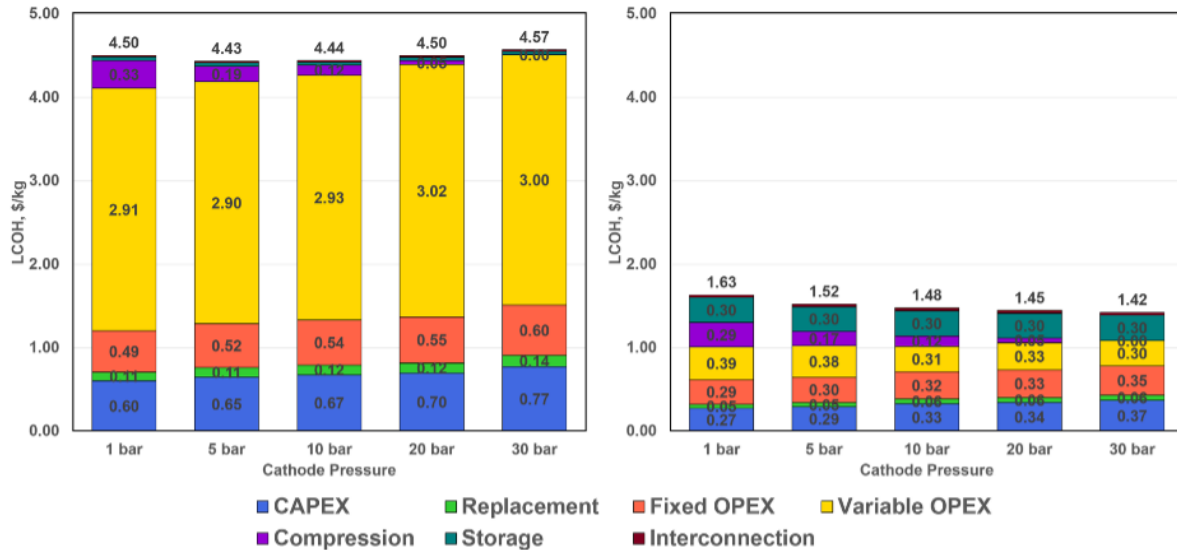


Figure 6: Optimized LCOH contributions under 2021 and 2035 cost and technology assumptions, with varying cathode pressures. Results correspond to dynamic operation. Left: LMP Southern California electricity price profile, maximum current density of 2 A/cm<sup>2</sup>. Right: ERCOT electricity price profile, maximum current density of 4 A/cm<sup>2</sup>.

### E. Influence of the Inflation Reduction Act on LCOH

The recent passage of the Inflation Reduction Act (IRA) offers production tax credit (PTC) under section 45V based on the carbon intensity (CI) of the produced hydrogen. Between CIs of 2.5 and 4 kg CO<sub>2</sub>/kg H<sub>2</sub>, the PTC is 0.6 \$/kg. Between CI's of 1.5 and 2.5 kg CO<sub>2</sub>/kg H<sub>2</sub>, the PTC is 0.75 \$/kg. Between CI's of 0.45 and 1.5 kg CO<sub>2</sub>/kg H<sub>2</sub>, the PTC is 1.0 \$/kg. Finally, for a CI less than 0.45 kg CO<sub>2</sub>/kg H<sub>2</sub>, the PTC is 3 \$/kg. For the 2021 scenario, the seasonal hourly carbon emission intensity of the grid in California (average grid CI of 274 kg/MWh) is used to compute a CI of 13.18 kg CO<sub>2</sub>/kg H<sub>2</sub>. For the 2035 Scenario, the CI in the NREL Cambium Southern California Mid Scenario (average grid CI of 117 kg/MWh) is 6.27 kg CO<sub>2</sub>/kg H<sub>2</sub>. The CI in the NREL Cambium Southern California 95% Decarbonization by 2050 Scenario (average grid CI of 146 kg/MWh) is 7.78 kg CO<sub>2</sub>/kg H<sub>2</sub>. Under the current IRA criteria, neither scenario qualifies for the PTC. Only in the NREL Cambium Southern California 95% Decarbonization by 2035 Scenario (average grid CI of 18 kg/MWh) does the CI fall to 0.79 kg CO<sub>2</sub>/kg H<sub>2</sub>, qualifying for 1.0 \$/kg for the first 10 years of production and making the effective LCOH 3.11 \$/kg (compared to 3.67 \$/kg without the PTC). Because there is such a steep jump in PTC between the lowest CI bracket and the next, there is a strong economic case for further oversizing of the production capacity to be able to follow the

low-marginal-emission hours of the grid in addition to the low-price hours. In the above 95% Decarbonization by 2035 Scenario, oversizing the production capacity by 2.6 times the demand rate allows the production facility to qualify for the highest PTC bracket with a CI of 0.43 kg CO<sub>2</sub>/kg H<sub>2</sub>. Despite the fact that the higher capital expenditure increases the pre-PTC LCOH to 4.03 \$/kg, the PTC lowers the effective LCOH to 2.34 \$/kg (compared to 3.11 \$/kg in the strictly price-following scenario). However, all the major U.S. grid systems in 2021 operate with a far higher carbon intensity (average of 379 kg/MWh) than what is necessary to qualify for the PTC credit. This means that grid-connected electrolytic hydrogen production facilities that do not procure additional renewable power will not qualify for the PTC provisioned by the IRA.

## 4. CONCLUSIONS

In this study, a techno-economic optimization framework was developed to compute the LCOH of a PEM hydrogen production facility under various price, cost, and technology assumptions. The framework was used to evaluate the benefits of dynamic operations and differential pressure operations under various grid scenarios. The results showed that dynamic operations lowered the LCOH by 8% under the 2021 scenario and by 15-20% under the 2035 sensitivity scenarios, indicating that dynamic operations would become more advantageous in the future. Partial differential pressure mode with a cathode pressure of 5 bar was identified as the most cost-effective way to compress hydrogen to 30 bar under the 2021 Scenario, while a full differential pressure mode was preferred under the 2035 Scenarios. The emissions analysis showed that grid-based hydrogen production in 2021 does not meet the CI criteria for the IRA 45V PTC. Thus, future hydrogen electrolysis projects in the next decade will require a dedicated renewable power source to take advantage of the PTC.

This study comes with some model limitations that should be noted. First, the absence of modeling hydrogen crossover could lead to an over-estimation of hydrogen production efficiency and safety issues. Second, the electrolyzer model does not incorporate an explicit model for membrane degradation, and it is anticipated that dynamic operations and high current densities could negatively impact the lifespan of the electrolyzer<sup>4</sup>. Although a stack replacement cost is incorporated in the TEA analysis, the model does not account for the variation of degradation with dynamic operation or current density. Lastly, the price-taker assumption used as part of the LCOH calculation might simplify market dynamics and neglect potential impacts on hydrogen pricing due to variations in supply and demand. Future studies should address these gaps to provide a more realistic evaluation of PEM production facilities.

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# Design space for PEM electrolysis for cost-effective H<sub>2</sub> production using grid electricity – Supplementary Information

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## SI1: Electrolyzer Model

The electrolyzer model summarizes the relationship between cell voltage and cell current based on the electrochemical principles. It considers the open-circuit voltage, activation overpotential, and ohmic overpotential to calculate the cell voltage that is required to drive electrochemical reactions in an electrolytic cell as shown in (Eq. S1).

$$V = E + \eta_{ohm} + \eta_{act} \quad (\text{Eq. S1})$$

$E$  is the thermodynamic equilibrium voltage, or open-circuit voltage, that is required to create the chemical potential that drives the hydrogen generation process. The thermodynamic equilibrium voltage can be derived from Gibbs free energy:

$$E = -\frac{\Delta G}{nF} \quad (\text{Eq. S2})$$

Based on the stoichiometry of the overall reaction,  $\Delta G$  can be expressed as follows:

$$\Delta G = G_{H_2}(T_{cat}, P_{cat}) + \frac{1}{2} G_{O_2}(T_{an}, P_{an}) - G_{H_2O}(T_{an}, P_{an}) \quad (\text{Eq. S3})$$

Since  $G = H - TS$ ,  $H(T, P)$  and  $S(T, P)$  can be used to calculate  $G(T, P)$  for each species. For water, the enthalpy and entropy of formation can be obtained from the steam tables. For the gas molecules, an analytical method computes  $G(T, P)$  by making temperature corrections on  $G$  in standard conditions and then making corrections for the pressure. The temperature dependence of enthalpy can be expressed as the following empirical model (Millet, 2021):

$$H(T, 1 \text{ bar}) - H(298 \text{ K}, 1 \text{ bar}) = a(T - T_0) + \frac{b}{2} 10^{-3} (T^2 - T_0^2) - c 10^5 \left( \frac{1}{T} - \frac{1}{T_0} \right) - \frac{e}{2} 10^8 \left( \frac{1}{T^2} - \frac{1}{T_0^2} \right)$$

(Eq. S4)

Similarly, the temperature dependence of entropy can be expressed as the following (Millet, 2021):

$$S(T, 1 \text{ bar}) - S(298 \text{ K}, 1 \text{ bar}) = a(\ln T - \ln T_0) + b 10^{-3} (T - T_0) - \frac{c}{2} 10^5 \left( \frac{1}{T^2} - \frac{1}{T_0^2} \right) - \frac{e}{3} 10^8 \left( \frac{1}{T^3} - \frac{1}{T_0^3} \right)$$

(Eq. S5)

The coefficients for the gas molecules are provided in Table S1 (Millet, 2021):

*Table S1: Coefficients for Hydrogen and Oxygen for Temperature Correction*

| Molecules          | a     | b    | c      | e    |
|--------------------|-------|------|--------|------|
| H <sub>2</sub> (g) | 26.57 | 3.77 | 1.17   | –    |
| O <sub>2</sub> (g) | 34.35 | 1.92 | -18.45 | 4.06 |

Since the values of  $H$  and  $S$  are known at standard temperature and pressure (298K and 1 bar),  $H(T, 1 \text{ bar})$ ,  $S(T, 1 \text{ bar})$ , and  $G(T, 1 \text{ bar})$  can be calculated for any temperature  $T$  and 1 bar. Then, another analytical expression using the Virial expansion is used to describe the pressure dependence of  $G$  (Millet, 2021):

$$G(T, P) - G(T, 1 \text{ bar}) = RT \ln P + BP + \frac{C - B^2}{2RT} P^2$$

(Eq. S6)

where

$$B = b_1 + \frac{b_2}{T} \text{ (cm}^3 \text{mol}^{-1}\text{)}$$

(Eq. S7)

$$C = c_1 + \frac{c_2}{T^2} \text{ (cm}^3 \text{mol}^{-1}\text{)}$$

(Eq. S8)

The Virial coefficients for the gas molecules for pressure up to 1000 atm are given in Table S2 (Millet, 2021).

Table S2: Virial Coefficients for Hydrogen and Oxygen for Pressure Correction

| Molecules          | b1   | b2      | c1     | c2     |
|--------------------|------|---------|--------|--------|
| H <sub>2</sub> (g) | 20.5 | -1,857  | -351   | 12,760 |
| O <sub>2</sub> (g) | 42.6 | -17,400 | -2,604 | 61,457 |

This two-step procedure of 1) correcting for temperature (Eq. S4 - S5) and then 2) correcting for pressure (Eq. S6 - S8) calculates G for each species at any temperature and pressure within the valid ranges of correlations (T < 100C, P < 1000 atm). Then, Eq. S3 is used to calculate  $\Delta G$  and Eq. S2 to calculate  $E$ .

$\eta_{ohm}$  is the ohmic overpotential which arises from the resistances from the electron flow, proton flow, electrical contact in the membrane. The biggest contributor to the ohmic resistance in the PEM electrolyzer is the membrane (Biaku et al., 2008; Marangio et al., 2009). The area-dependent resistance of a membrane is given by the formula:

$$R_{membrane} = \frac{t}{\sigma_{mem}} \quad (\text{Eq. S9})$$

where  $t$  is the thickness of the membrane and  $\sigma_{mem}$  is the conductivity of the membrane. The membrane conductivity is given by empirical correlations as shown below (Abdol Rahim et al., 2016; Görgün, 2006):

$$\lambda_m = 0.043 + 17.81a_{H_2O} - 39.85a_{H_2O}^2 + 36a_{H_2O}^3 \quad (\text{Eq. S10})$$

$$\sigma_{mem} = (0.00514\lambda_m - 0.00326)e^{\left(1268\left(\frac{1}{303} - \frac{1}{T}\right)\right)} \quad (\text{Eq. S11})$$

where  $\lambda_m$  is the water hydration factor. Since the membrane is fully immersed in water during electrolytic operations (García-Valverde et al., 2012; Springer et al., 1991),  $a_{H_2O}$  is assumed to be 1. According to Eq. S11, the membrane conductivity increases with increasing temperature, but it is not affected by changes in pressure.

$\eta_{act}$  is the activation overpotential that is needed to supply the activation energy to initiate the exchange of electrons (Abdol Rahim et al., 2016; Bessarabov and Millet, 2018). The Butler-Volmer equation, which is a commonly-used model for water electrolysis, describes the relationship between current density and activation overpotential (Bessarabov and Millet, 2018):

$$j = j_0 \left[ \exp\left(\frac{-\alpha_1 F z \eta_a}{RT}\right) - \exp\left(\frac{\alpha_2 F z \eta_a}{RT}\right) \right]$$

(Eq. S12)

where  $j_0$  is the exchange current density,  $\alpha$  is the charge transfer coefficient, and  $\eta_a$  is the activation overpotential. Assuming that  $\alpha = \alpha_1 = \alpha_2$ , (Eq. S12) can be simplified to the following equation using the hyperbolic sine function:

$$\eta_a = \frac{RT}{\alpha z F} \left( \frac{j}{2j_0} \right)$$

(Eq. S13)

Previous studies have reported experimental values for the anode charge transfer coefficient and cathode charge transfer coefficient as 2 and 0.5, respectively (Abdin et al., 2015; Biaku et al., 2008; García-Valverde et al., 2012; Görgün, 2006; Lebbal and Lecœuche, 2009; Marangio et al., 2009; Ni et al., 2008). However, the theoretical value for a charge transfer coefficient is between 0 and 1.

The exchange current density,  $j_0$ , is an intensive physical property that measures the ease with which the charge transfer occurs (Millet 2015, Chapter 2). It is a function of the physical characteristics of the electrolyte such as material and morphology, catalyst, age, and operating conditions (García-Valverde et al., 2012; Liso et al., 2018). For the Pt catalyst in the cathode,  $j_0$  of  $10^{-4}$ - $10^{-3}$  A/cm<sup>2</sup> has been suggested (Choi et al., 2004; Escobar-Yonoff et al., 2021). For the Ir and Pt-Ir catalyst in the anode,  $j_0$  of  $10^{-13}$ - $10^{-6}$  A/cm<sup>2</sup> has been used (Escobar-Yonoff et al., 2021; Marangio et al., 2009).

In this study, an approach from (Carmo et al., 2013; Coutanceau et al., 2018; García-Valverde et al., 2012) is adopted to express the temperature dependence of  $j_0$ :

$$j_0 = j_0^{ref} \exp \left[ \frac{E_{act}}{R} \left( \frac{1}{T^{ref}} - \frac{1}{T} \right) \right]$$

(Eq. S14)

where  $j_0^{ref}$  is the reference exchange current density at the reference temperature,  $T^{ref}$ , and  $E_{act}$  is the activation energy corresponding to the OER (in the anode) or HER (in the cathode). The values of  $j_0^{ref}$  and  $E_{act}$  are summarized in Table S3.

Table S3: Values of Reference Current Density and Activation Energy for OER and HER

| Variable              | Value                 | Units             | Reference                                                                   |
|-----------------------|-----------------------|-------------------|-----------------------------------------------------------------------------|
| $j_{0,cathode}^{ref}$ | $0.75 \times 10^{-3}$ | A/cm <sup>2</sup> | (Carmo et al., 2013; Coutanceau et al., 2018; García-Valverde et al., 2012) |
| $E_{act,cathode}$     | 30,000                | J/mol             | (He et al., 2017)                                                           |
| $j_{0,anode}^{ref}$   | $1.0 \times 10^{-7}$  | A/cm <sup>2</sup> | (Carmo et al., 2013; Coutanceau et al., 2018; García-Valverde et al., 2012) |
| $E_{act,anode}$       | 90,000                | J/mol             | (García-Valverde et al., 2012; Suermann et al., 2018)                       |
| $T^{ref}$             | 318                   | K                 | (García-Valverde et al., 2012)                                              |

Therefore, the total activation overpotential of an electrolytic cell is:

$$\eta_a = -\frac{RT}{F} \left( \frac{j}{2j_{0,anode}} \right) + -\frac{RT}{F} \left( \frac{j}{2j_{0,cathode}} \right) \quad (\text{Eq. S15})$$

## SI2: Electrolyzer Model Validation

The electrolyzer model is validated by benchmarking the model with two experiments that were conducted in different operating conditions. In the first benchmarking experiment (Liso et al., 2018) a single-cell electrolyzer with Nafion 117 (membrane thickness of 178  $\mu\text{m}$ ) was tested at two different temperatures (60°C and 80°C) at ambient pressure. was used. Table S4 summarizes the parameters that were used in the model.

Table S4: Electrolyzer Model Parameters for Single-Cell Experiment at 60°C and 80°C

| Parameters                                   | Values    | Units         |
|----------------------------------------------|-----------|---------------|
| Membrane Thickness, $t$                      | 178       | $\mu\text{m}$ |
| Electrodes and Plates Resistance, $R_{elec}$ | 0         | ohms          |
| Temperature, $T$                             | 60 and 80 | °C            |
| Cathode Pressure, $P_{cat}$                  | 1         | bar           |
| Anode Pressure, $P_{an}$                     | 1         | bar           |

Figure S1 shows that the model predictions and experimental data match well for both the 60°C and 80°C experiments.

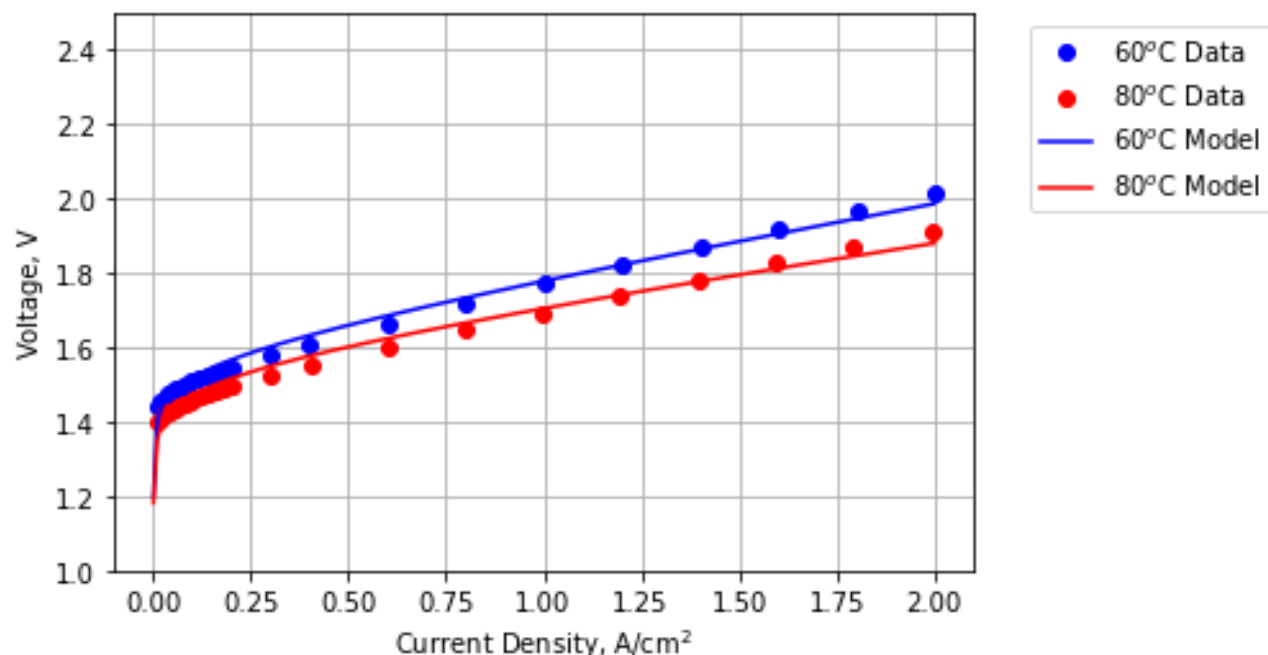


Figure S1: Comparison of the electrolyzer model (line) with experimental data (circles) at 80°C (red) and 60°C (blue) in ambient pressure. Data from (Liso et al., 2018).

The second experiment is a stack experiment in differential pressure mode. (Marangio et al., 2009) conducted high-pressure experiments with a commercial electrolyzer system from Giner Electrochemical Systems LLC. The stack is made up of twelve cells with an active area of 160 cm<sup>2</sup> each. The experiments were conducted at temperatures between 40 – 55°C and cathode pressures between 1 and 70 bar.

Nafion 110 with a thickness of 254 µm was used in the experiment. In order to match the experimental results, an additional resistance term was added to the model since the resistance from the electrodes, plates, and other wires may no longer be negligible in a stack of 10-15 cells (Awasthi et al., 2011). The values of the model parameters are summarized in Table S5 and Table S6 for the 10-bar-55°C experiment and the 70-bar-40°C experiment, respectively.

Table S5: Electrolyzer Model Parameters for Stack Experiment at 55°C and 10 bar

| Parameters                                        | Values                             | Units                  |
|---------------------------------------------------|------------------------------------|------------------------|
| Membrane Thickness, $t$                           | 254                                | $\mu\text{m}$          |
| Electrodes and Plates Resistance, $R_{elec}$      | $2.5 \times 10^{-5}$               | ohms                   |
| Cathode Exchange Current Density, $j_{0,cathode}$ | $1.1 \times 10^{-3}$ (Calculated)  | $\text{A}/\text{cm}^2$ |
| Anode Exchange Current Density, $j_{0,anode}$     | $2.87 \times 10^{-7}$ (Calculated) | $\text{A}/\text{cm}^2$ |
| Temperature, $T$                                  | 55                                 | °C                     |
| Cathode Pressure, $P_{cat}$                       | 10                                 | bar                    |
| Anode Pressure, $P_{an}$                          | 1                                  | bar                    |

Table S6: Electrolyzer Model Parameters for Stack Experiment at 40°C and 70 bar

| Parameters                                        | Values                            | Units                  |
|---------------------------------------------------|-----------------------------------|------------------------|
| Membrane Thickness, $t$                           | 254                               | $\mu\text{m}$          |
| Electrodes and Plates Resistance, $R_{elec}$      | $2.5 \times 10^{-5}$              | ohms                   |
| Cathode Exchange Current Density, $j_{0,cathode}$ | $0.6 \times 10^{-3}$ (Calculated) | $\text{A}/\text{cm}^2$ |
| Anode Exchange Current Density, $j_{0,anode}$     | $5.9 \times 10^{-8}$ (Calculated) | $\text{A}/\text{cm}^2$ |
| Temperature, $T$                                  | 40                                | °C                     |
| Cathode Pressure, $P_{cat}$                       | 70                                | bar                    |
| Anode Pressure, $P_{an}$                          | 1                                 | bar                    |

Figure S2 shows that the electrolyzer model reasonably matches the experimental results just by changing the operating pressure and temperature while maintaining the rest of parameters the same.

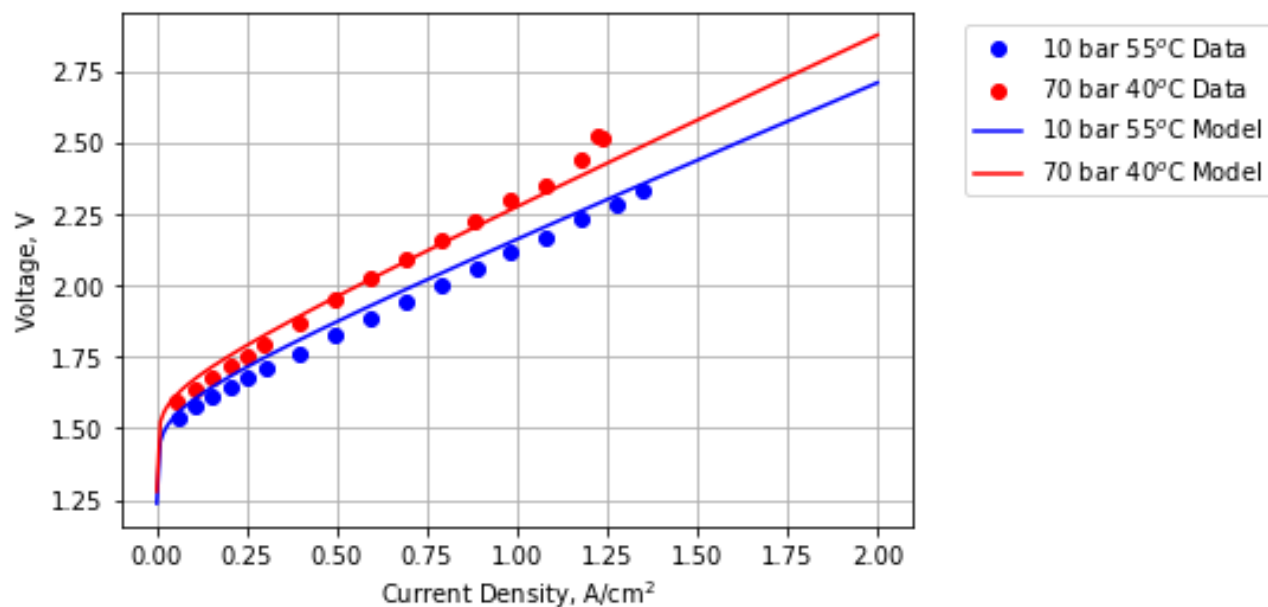


Figure S2: Comparison of the electrolyzer model (line) with experimental data (circles) at (a) 55°C and 10 bar (blue); (b) 40°C and 70 bar (red). Data from (Marangio, Santarelli, and Cali 2009).

For the 2021 and 2035 simulation cases evaluated in this study, the electrolyzer model parameters are configured to reflect the state-of-the-art specifications of the 2021 and 2035 technologies as shown in Table S7.

Table S7: Electrolyzer Model Parameters for 2035 Simulations (IRENA 2020)

| Parameters                  | 2021 Scenario | 2035 Scenario | Units                  |
|-----------------------------|---------------|---------------|------------------------|
| Membrane Thickness, $t$     | 178           | 127           | $\mu\text{m}$          |
| Temperature, $T$            | 80            | 80            | $^{\circ}\text{C}$     |
| Cathode Pressure, $P_{cat}$ | 30            | 30            | bar                    |
| Anode Pressure, $P_{an}$    | 1             | 1             | bar                    |
| Minimum Current Density     | 0             | 0.5           | $\text{A}/\text{cm}^2$ |

The 2021 and 2035 polarization curves (red and blue, respectively) are plotted against other benchmark systems from (Buttler and Spliethoff, 2018) in Figure S3.

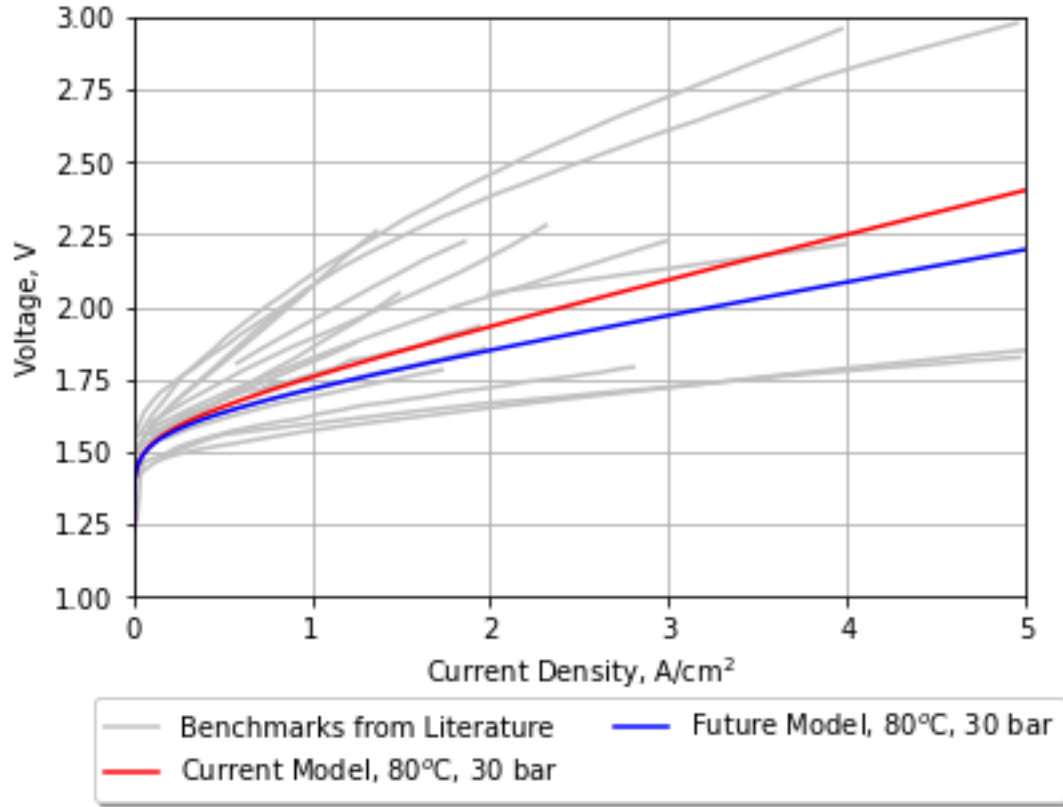


Figure S3: Comparison of the 2021 and 2035 electrolyzer models with other PEM electrolyzer polarization curves from the literature. Benchmark system data from (Buttler and Spliethoff, 2018).

### SI3: Electrolyzer Techno-Economic Model

The electrolyzer techno-economic model uses the discounted cash flow formulation to calculate the present value of all costs associated with planning, permitting, building, and operating the electrolyzer plant. The net present value of the total project cost is calculated by:

$$NPV(C_{Total}) = C_{CAPEX} + C_{storage} + \sum_{y=1}^{40} \left[ (C_{RP}(y) + C_{fOPEX}(y) + C_{vOPEX}(y)) \frac{(1 + \pi)^y}{(1 + d)^y} \right]$$

(Eq. S16)

The formulation assumes construction in year 0, project start in year 1, a project lifetime of 40 years, a discount rate of 8%, and an annual inflation rate of 1.9%.

The total capital cost,  $C_{CAPEX}$ , which is a one-time investment incurred before the plant start-up, is the sum of direct capital, site preparation cost, engineering cost, contingency cost, permitting cost, and land cost. The direct capital is the sum of the capital costs for the electrolyzer stack, mechanical balance-of-

plant, and electrical balance-of-plant. The electrolyzer stack is priced based on the stack cost per unit area (\$2.00/cm<sup>2</sup> in the 2021 Scenario). In addition, a pressure correction factor to account for additional equipment cost to withstand high-pressure operating conditions was formulated based on industry data from NEL (Saba et al., 2018), which is shown in S4a. The 15-bar system is approximately 20% more expensive than the ambient system. Assuming that the correction factor increases linearly with operating pressure, a linear correction factor that passes through 1 at 1 bar and 1.2 at 15 bar is constructed, as shown with a red line in S4b. Since the electrolyzer cost model considered here is based on an operating pressure of 300 psi (20.68 bar), the pressure correction factor was normalized by dividing each correction factor by the correction factor at 20.68 bar. The resulting normalized correction factor is also shown with a blue line in S4b. The pressure correction factor ensures that the capital cost for an electrolyzer stack with a 30-bar operating pressure is higher than the cost for an ambient system. Site preparation cost, engineering cost, contingency cost, and permitting cost are expressed as some proportions of the direct capital (2%, 10%, 15%, 15%, respectively) while the land cost is assumed to be fixed (5 acres at \$50,000/acre).

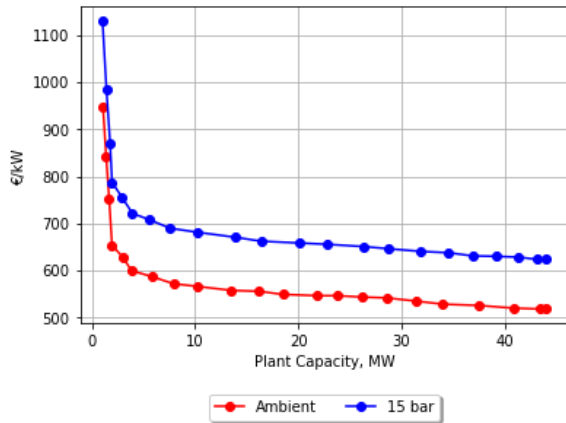


Figure S4a: Capital costs of a 15-bar electrolyzer system (blue) and an ambient pressure electrolyzer system (red) as a function of plant capacity. Adapted from (Saba et al., 2018).

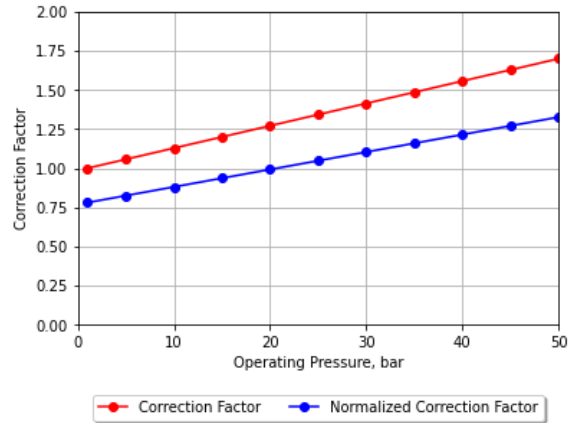


Figure S4b: Original and normalized pressure correction factor as a function of operating pressure from 1 bar to 50 bar.

$C_{storage}$  is the capital cost for a hydrogen storage system, which is assumed at \$500 per kg of hydrogen.

The annual replacement cost,  $C_{RP}(y)$ , is the sum of the planned replacement cost and annual unplanned replacement cost. The planned replacement cost is 15% of the direct capital incurred at a

stack replacement interval, which is assumed to be 10 years in this study. The annual unplanned replacement cost, which is 0.5% of the direct capital incurring annually, covers any other miscellaneous part replacements.

The annual fixed operating cost,  $C_{fOPEX}(y)$ , accounts for the labor cost, overhead cost, tax and insurance cost, and material cost. Labor cost and overhead cost are determined by the number of staff required to operate the plant. Tax and insurance cost is assumed to be 2% of the total capital cost. Material cost is assumed to be 3% of the direct capital.

Finally,  $C_{vOPEX}(y)$  is the annual variable operating expense that is made up of electricity cost and water cost. The electricity cost accounts for the amount of power consumed by the electrolyzer stack and the BoPs. The stack power usage is calculated from the polarization curve while the BoP power usage is assumed to be  $5.1 \text{ kWh kg}^{-1} \text{ H}_2^{-1}$ . The water cost directly scales with hydrogen production at a consumption rate of 3.78 gallons per kg of hydrogen produced.

#### SI4: Compression Techno-Economic Model

The compression techno-economic model uses the discounted cash flow formulation to calculate the present value of all costs associated with planning, permitting, building, and operating the mechanical compressors. The net present value of the total project cost is calculated by:

$$NPV(C_{Compress}) = TCI + \sum_{y=1}^{40} \left[ (C_{C,OPEX}(y) + C_{C,RP}(y)) \frac{(1+\pi)^y}{(1+d)^y} \right] \quad (\text{Eq. S17})$$

$TCI$  is the total capital investment of the compressors, which depends on the rated power of the compressor and other factors. For compression in which the inlet pressure (or suction pressure) differs significantly from the outlet pressure (or discharge pressure), multiple compression stages are required to reach the target discharge pressure. The number of compression stages,  $N$ , required for high compression ratios ( $>2$ ) is given by (Khan et al., 2021).

$$N = \frac{\log\left(\frac{P_{disc}}{P_{suc}}\right)}{\log(x)} \quad (\text{Eq. S18})$$

where  $P_{disc}$  is the discharge pressure at the outlet of the compressor,  $P_{suc}$  is the suction pressure at the inlet of the compressor, and  $x$  is the compression ratio for each stage which is between 2.1 and 4.0. In this study,  $x$  is assumed to be 2.1. Compression is typically modeled as an isentropic process and later corrected with an isentropic efficiency factor. For such an ideal multi-stage compression process, the power requirement is given by (Khan et al., 2021).

$$Power = N \left( \frac{k}{k-1} \right) \left( \frac{Z}{\eta_{isen}} \right) T_{suc} (q_M) R \left[ \left( \frac{P_{disc}}{P_{suc}} \right)^{\left( \frac{k-1}{Nk} \right)} - 1 \right]$$

(Eq. S19)

where  $k$  (1.41 for hydrogen) is the ratio of specific heat under constant pressure ( $C_p$ ) to specific heat under constant volume ( $C_v$ ),  $Z$  is the average compressibility factor,  $\eta_{isen}$  is the isentropic efficiency,  $T_{suc}$  is the suction temperature (usually ambient) at the inlet of the compressor,  $q_M$  is the molar flow rate, and  $R$  is the universal gas constant. The rated power is calculated by dividing the actual compression power by the motor efficiency, which is assumed as 90% in this study (Khan et al., 2021).

$$Rated\ Power = \frac{Power}{Motor\ Efficiency}$$

(Eq. S20)

In order to calculate the compressibility factor  $Z$ , the average temperature and pressure inside the compression need to be computed. Typically, the suction pressure, the discharge pressure, and the suction temperature are known. Based on the assumptions of an isentropic system, the discharge temperature can be calculated by Eq. S21 (Khan et al., 2021).

$$T_{disc} = T_{suc} \left[ 1 + \frac{\left( \frac{P_{disc}}{P_{suc}} \right)^{\left( \frac{k-1}{Nk} \right)} - 1}{\eta_{isen}} \right]$$

(Eq. S21)

The average temperature is given by Eq. S22 (Khan et al., 2021).

$$T_{average} = \frac{T_{suc} + T_{dis}}{2}$$

(Eq. S22)

The average pressure is given by Eq. S23 (Khan et al., 2021).

$$P_{avg} = \frac{2}{3} \left( \frac{P_{disc}^3 - P_{suc}^3}{P_{disc}^2 - P_{suc}^2} \right)$$

(Eq. S23)

Finally,  $Z$  is evaluated as a function of the average temperature and pressure using the Redlick-Kwong equation of state.

$\eta_{isen}$  accounts for the non-ideality of the compression process, which is not adiabatic or reversible, and, as a result, some of the work can go into increasing the internal energy or temperature of the system.

$\eta_{isen}$  for large reciprocating compressors range from 65 to 70% while  $\eta_{isen}$  for small reciprocating

compressors range from 40 to 50% (Chen, 2010; Khan et al., 2021). In this study, an  $\eta_{isen}$  of 65% is assumed (Parks et al., 2014). The uninstalled cost (UC) of a compressor scales with the rated power of the compressor. For a high-flow, moderate-compression system, the following scaling factor is applicable (Khan et al., 2021).

$$UC = 2695.03 \times \text{Rated Power}^{0.8335} \quad (\text{Eq. S24})$$

The total installed cost (TIC) of a compressor is calculated by applying an installation factor (IF) to UC:

$$TIC = UC \times IF \quad (\text{Eq. S25})$$

In this model, the installation factor is assumed to be 2 (Khan et al., 2021) .

There are other indirect costs associated with the total capital investment (TCI) of a compressor. Site preparation, which is 5% of the TIC, covers the purchase of land and other auxiliary equipment. Engineering and design amounts to 10% of the TIC. Project contingency, which is allocated to deal with any unforeseen issues, takes another 10% of the TIC. Permitting to get the appropriate approvals and control equipment is 3% of the TIC (Chen, 2010). Finally, the owner's cost, which accounts for the owners' engineering, debt origination, closure costs, and due diligence studies, takes 12% of the TIC (Chen, 2010). Therefore, the TCI is calculated by adding all these indirect costs to the TIC:

$$TCI = TIC + \text{Indirect Costs} \quad (\text{Eq. S26})$$

$C_{C,OPEX}$  is the operating expense, which is made up of the electricity cost, labor cost, overhead cost, and other costs (insurance, property tax, licensing/permits, operating/maintenance/repairs). The electricity cost is calculated based on the hourly hydrogen production rate and the corresponding rate power of the compressor. The labor cost is the product of the labor hours and the hourly labor rate. The required labor hour is given by (Khan et al., 2021).

$$\text{Labor Hour} = 288 \times \left( \frac{\text{Daily Compressor Flow Rate}}{100,000} \right)^{0.25} \quad (\text{Eq. S27})$$

where *Daily Compressor Flow Rate* is in kg/day. The average hourly labor rate for 2021 is \$42.48/hour (Bureau of Labor Statistics- NAICS code# 2212). The overhead cost is 50% of the labor cost (Khan et al., 2021). Insurance, Property Taxes and Licensing and Permits are 1%, 1%, and 0.1% of the TCI. Operating/Maintenance/Repairs costs are 4% of the TIC (Chen, 2010; Khan et al., 2021). In addition, the lifetime of a compressor is assumed to be 15 years. Therefore, the compressor needs to be replaced every 15 years. The replacement cost for the compressor is TIC adjusted for inflation.

## SI5: Hourly Grid Electricity Prices

18 different electricity forecasts have been used to explore the implications of electricity price variations on future LCOHs. The highlighted traits and assumptions for each forecast is provided in Table S8.

*Table S8: Summary of Attributes of 18 Electricity Forecasts used in this Study*

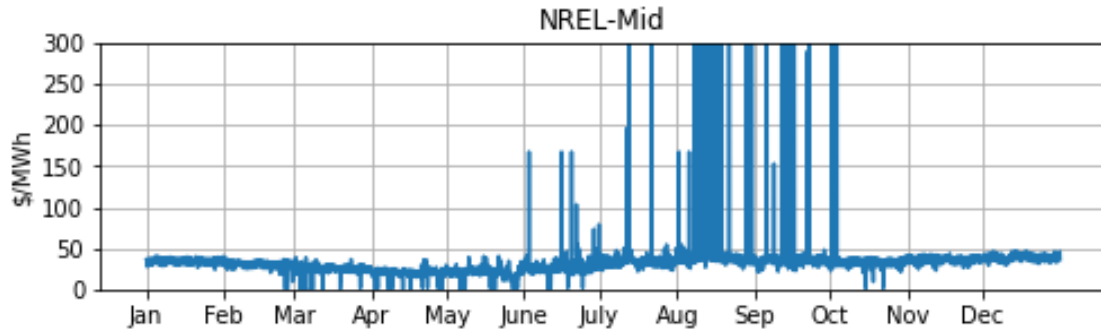
| Forecast Name          | Location(s) | Year | Description                                                                               | Average Price | Number of Hours of <\$5MWh | Number of hours of >100/MWh |
|------------------------|-------------|------|-------------------------------------------------------------------------------------------|---------------|----------------------------|-----------------------------|
| NREL-Mid               | Southern CA | 2040 | Mid-level assumptions for demand, resource, cost, price, technology. No new carbon policy | 40.61         | 222                        | 106                         |
| NREL-HighRECost        | Southern CA | 2040 | Mid with 2020 ATB RE advanced projections                                                 | 44.49         | 33                         | 106                         |
| NREL-LowRECost         | Southern CA | 2040 | Mid with 2020 ATB RE conservative projections                                             | 31.78         | 1584                       | 105                         |
| NREL-Mid95by2035       | Southern CA | 2040 | Mid with 95% CO <sub>2</sub> reduction by 2035                                            | 59.48         | 3073                       | 112                         |
| NREL-Mid95by2050       | Southern CA | 2040 | Mid with 95% CO <sub>2</sub> reduction by 2050                                            | 42            | 431                        | 109                         |
| Princeton-BaseCaseTax  | Nationwide  | 2030 | \$60/tonne carbon tax, AEO2020 REF natural gas prices, mid cost for all resources         | 47.69         | 799                        | 262                         |
| Princeton-HighWindTax  | Nationwide  | 2030 | BaseCaseTax with low cost for wind clusters                                               | 45.06         | 1628                       | 256                         |
| Princeton-HighSolarTax | Nationwide  | 2030 | BaseCaseTax with low cost for solar clusters                                              | 45.43         | 1218                       | 241                         |
| NREL-MiNg_\$100_CAISO  | CA          | 2035 | \$100/tonne carbon tax, AEO2020 REF natural gas prices                                    | 53.08         | 3413                       | 518                         |
| NREL-MiNg_\$150_CAISO  | CA          | 2035 | \$150/tonne carbon tax, AEO2020 REF natural gas prices                                    | 45.54         | 5080                       | 2702                        |
| NREL-MiNg_\$100_ERCOT  | TX          | 2035 | \$100/tonne carbon tax, AEO2020 REF natural gas prices                                    | 33.76         | 4049                       | 79                          |
| NREL-MiNg_\$150_ERCOT  | TX          | 2035 | \$150/tonne carbon tax, AEO2020 REF natural gas prices                                    | 36.34         | 5199                       | 1401                        |

*Table S8 Continued*

| Name                   | Location(s)                                            | Year | Description                                            | Average Price | Number of Hours of <\$5MWh | Number of hours of >\$100/MWh |
|------------------------|--------------------------------------------------------|------|--------------------------------------------------------|---------------|----------------------------|-------------------------------|
| NREL-MiNg_\$100_MISO-W | AR, IL, IN, IA, KY, LA, MI, MN, MO, MT, ND, SD, TX, WI | 2035 | \$100/tonne carbon tax, AEO2020 REF natural gas prices | 35.22         | 3858                       | 17                            |
| NREL-MiNg_\$150_MISO-W | AR, IL, IN, IA, KY, LA, MI, MN, MO, MT, ND, SD, TX, WI | 2035 | \$150/tonne carbon tax, AEO2020 REF natural gas prices | 28.28         | 5476                       | 539                           |
| NREL-MiNg_\$100_NYISO  | NY                                                     | 2035 | \$100/tonne carbon tax, AEO2020 REF natural gas prices | 35.33         | 4816                       | 185                           |
| NREL-MiNg_\$150_NYISO  | NY                                                     | 2035 | \$150/tonne carbon tax, AEO2020 REF natural gas prices | 33.84         | 5834                       | 1421                          |
| NREL-MiNg_\$100_PJM-W  | DE, IL, IN, KY, MD, MI, NJ, NC, OH, PA, TN, VA, WV, DC | 2035 | \$100/tonne carbon tax, AEO2020 REF natural gas prices | 48.08         | 1397                       | 50                            |
| NREL-MiNg_\$150_PJM-W  | DE, IL, IN, KY, MD, MI, NJ, NC, OH, PA, TN, VA, WV, DC | 2035 | \$150/tonne carbon tax, AEO2020 REF natural gas prices | 49.94         | 2666                       | 1903                          |

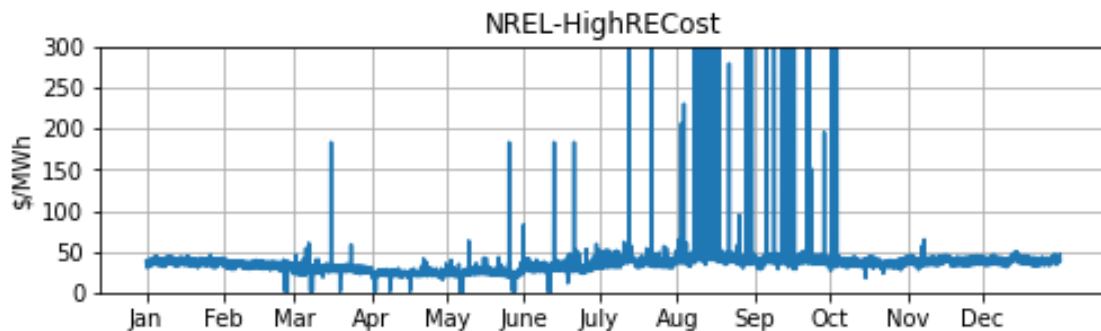
The profile and short descriptions of each electricity forecast scenario is provided below:

- NREL-Mid: This is NREL's business-as-usual scenario (Gagnon et al., 2021). The 2040 electricity prices from the P10 balancing area (Southern California) were used for this study. The average price is \$40.61/MWh. There are spikes of high prices during the summer.



*Figure S5: Electricity Price Predictions from NREL-Mid Scenario*

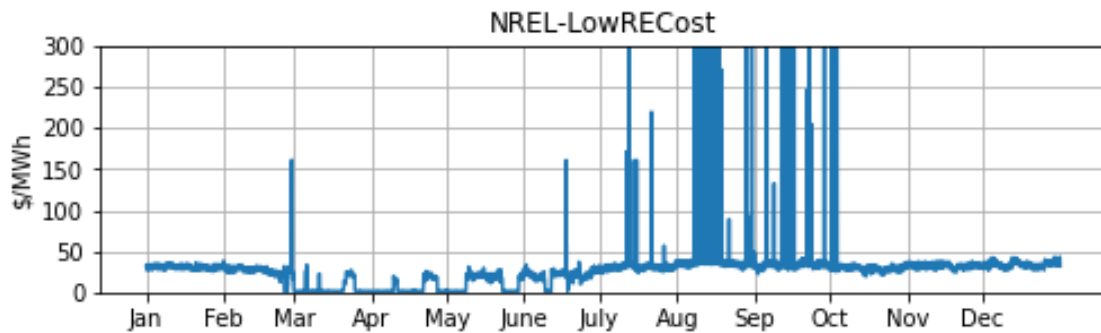
- **NREL-HighRECost:** This 2040 scenario has the same assumptions as NREL-Mid scenario, but with higher renewable energy costs (Gagnon et al., 2021). The electricity prices from the P10 balancing area (Southern California) were used for this study. The average price is \$44.49/MWh. There are spikes of high prices during the summer.



*Figure S6: Electricity Price Predictions from NREL-HighRECost Scenario*

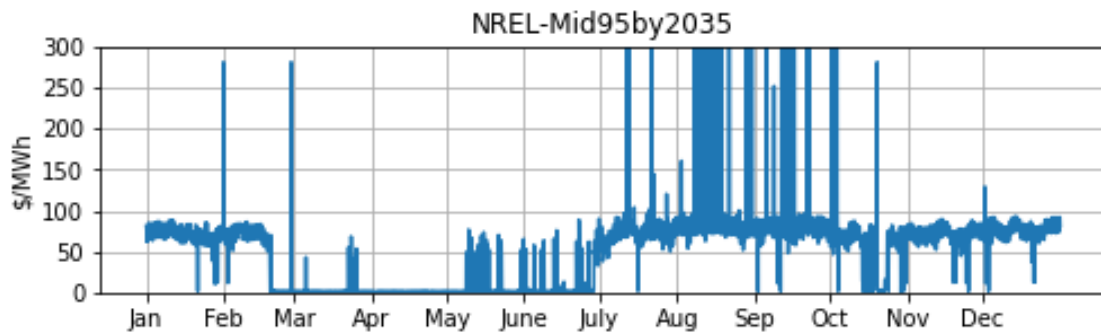
- **NREL-LowRECost:** This 2040 scenario has the same assumptions as NREL-Mid scenario, but with lower renewable energy costs (Gagnon et al., 2021). The electricity prices from the P10 balancing area (Southern California) were used for this study. The average price is \$31.78/MWh. There are spikes of high prices

during the summer time. Low electricity prices are available during the spring time.



*Figure S7: Electricity Price Predictions from NREL-LowRECost*

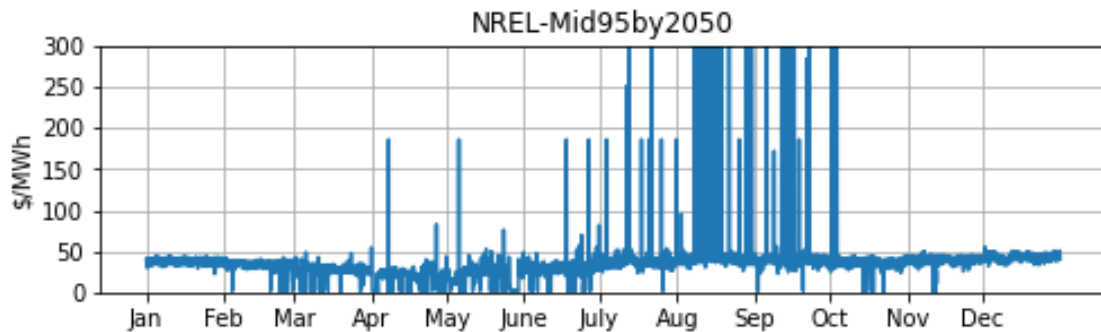
- NREL-Mid95by2035: This 2040 scenario has the same assumptions as NREL-Mid scenario, but with 95% decarbonization of the national power sector by 2035 (Gagnon et al., 2021). The electricity prices from the P10 balancing area (Southern California) were used for this study. The average price is \$59.48/MWh with higher levels of fluctuations throughout the day. There are spikes of high prices during the summer. Low electricity prices are available during the spring time.



*Figure S8: Electricity Price Predictions from NREL-Mid95by2035*

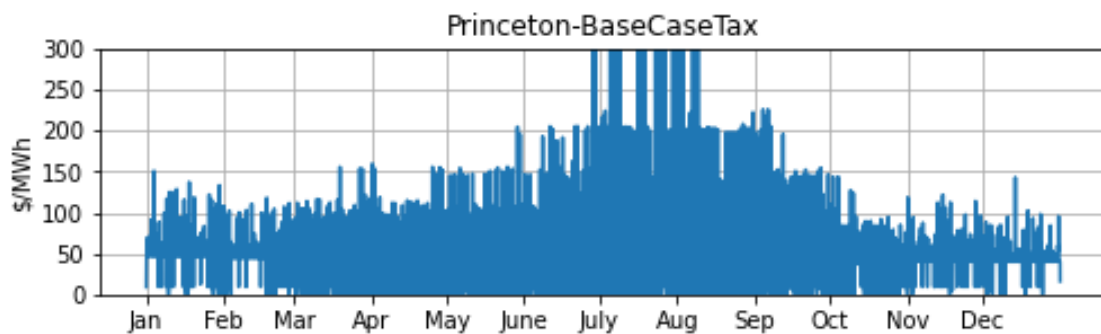
- NREL-Mid95by2050: This 2040 scenario has the same assumptions as NREL-Mid scenario, but with 95% decarbonization of the national power sector by 2050 (Gagnon et al., 2021). The electricity prices from the P10 balancing area (Southern

California) were used for this study. The average price is \$42.00/MWh. There are spikes of high prices during the summer. Highly-fluctuating low electricity prices are available during the spring time.



*Figure S9: Electricity Price Predictions from NREL-Mid95by2050*

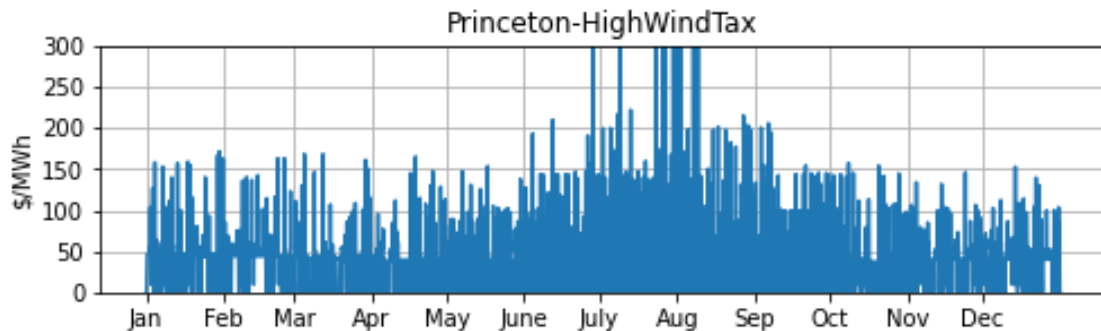
- **Princeton-BaseCaseTax:** This 2030 prediction is based on the US-wide aggregate load and existing capacity with Carbon Tax of \$60/tonne (Jesse D Jenkins, 2021). It assumes AEO2020 REF natural gas prices and ATB2020 mid costs for all resources. The average price is \$47.69/MWh. There are high fluctuations throughout the year. The prices are generally higher during the summer.



*Figure S10: Electricity Price Predictions from Princeton-BaseCaseTax*

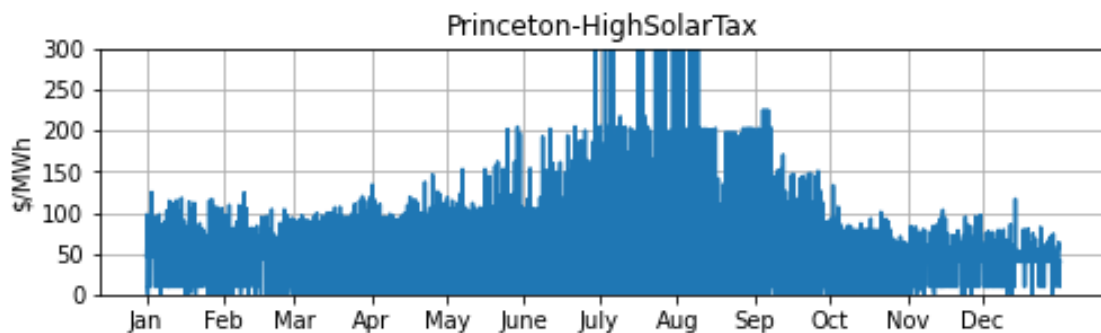
- **Princeton-HighWindTax:** This 2030 prediction is based on the US-wide aggregate load and existing capacity with Carbon Tax of \$60/tonne (Jesse D Jenkins, 2021). It assumes AEO2020 REF natural gas prices and ATB2020 low costs for new wind

clusters. The average price is \$45.06/MWh. There are high fluctuations throughout the year. The prices are generally higher during the summer.



*Figure S11: Electricity Price Predictions from Princeton-HighWindTax*

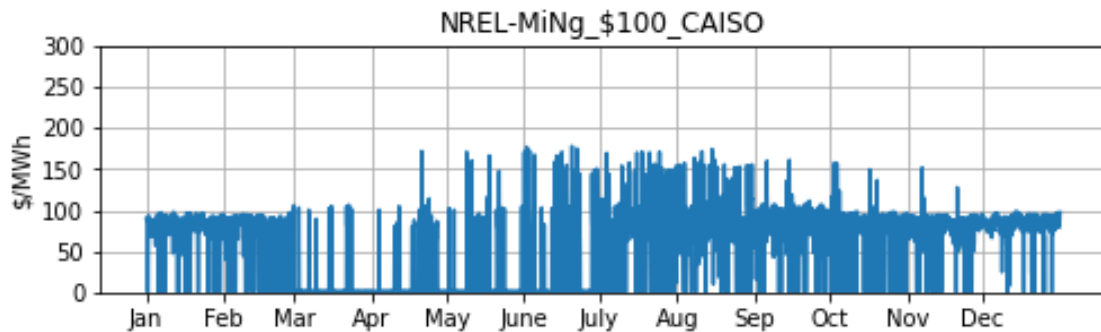
- Princeton-HighSolarTax: This 2030 prediction is based on the US-wide aggregate load and existing capacity with Carbon Tax of \$60/tonne (Jesse D Jenkins, 2021). It assumes AEO2020 REF natural gas prices and ATB2020 low costs for new solar clusters. The average price is \$45.43/MWh. There are high fluctuations throughout the year. The prices are generally higher during the summer.



*Figure S12: Electricity Price Predictions from Princeton-HighSolarTax*

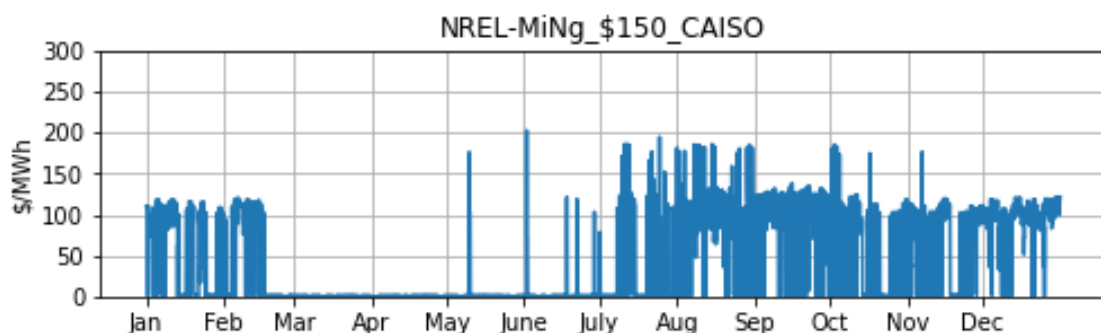
- NREL-MiNg\_\$100\_CAISO: This NREL scenario predicts 2035 electricity prices for California Independent System Operator (CAISO) region based on ATB2020 mid technology costs and AEO2020 reference load projections with carbon tax of

\$100/tonne (Cohen and Durvasulu, 2021). The average price is \$53.08/MWh. There are high fluctuations throughout the year. The prices are generally higher during the summer.



*Figure S13: Electricity Price Predictions from NREL-MiNg\_\$100\_CAISO*

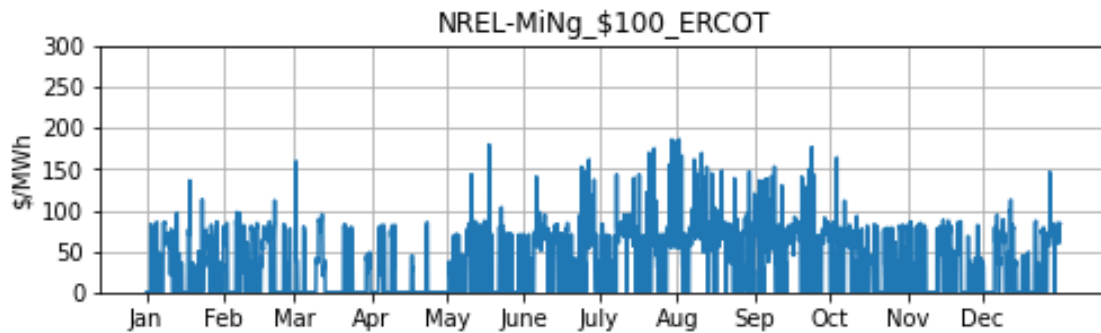
- NREL-MiNg\_\$150\_CAISO: This NREL scenario predicts 2035 electricity prices for California Independent System Operator (CAISO) region based on ATB2020 mid technology costs and AEO2020 reference load projections with carbon tax of \$150/tonne (Cohen and Durvasulu, 2021). The average price is \$45.54/MWh. There are high fluctuations throughout the year. Low electricity prices are available during the spring time. The prices are generally higher during the summer.



*Figure S14: Electricity Price Predictions from NREL-MiNg\_\$150\_CAISO*

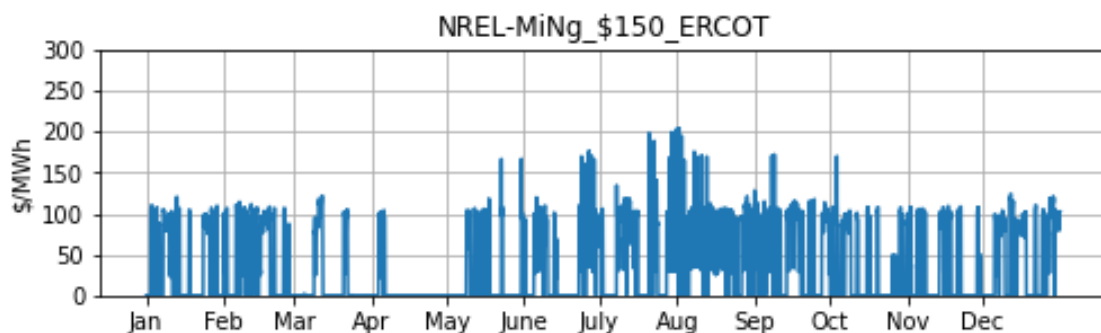
- NREL-MiNg\_\$100\_ERCOT: This NREL scenario predicts 2035 electricity prices for Electric Reliability Council of Texas (ERCOT) region based on ATB2020 mid

technology costs and AEO2020 reference load projections with carbon tax of \$100/tonne (Cohen and Durvasulu, 2021). The average price is \$33.76/MWh. There are high fluctuations throughout the year. Low electricity prices are available during spring time. The prices are generally higher during the summer.



*Figure S15: Electricity Price Predictions from NREL-MiNg\_\$100\_ERCOT*

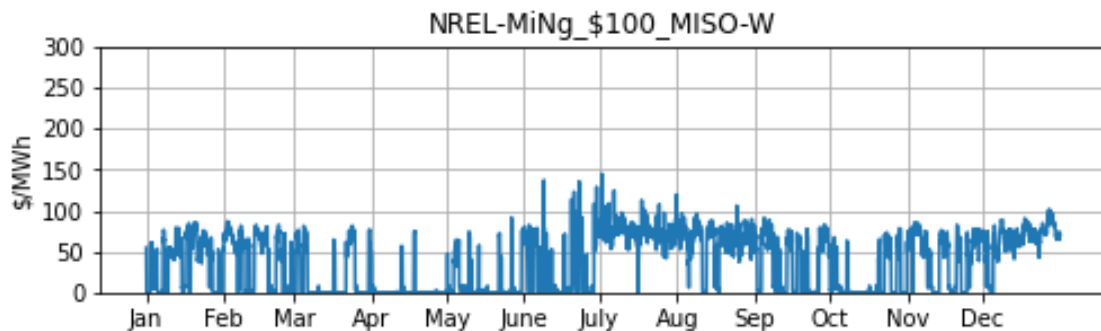
- NREL-MiNg\_\$150\_ERCOT: This NREL scenario predicts 2035 electricity prices for Electric Reliability Council of Texas (ERCOT) region based on ATB2020 mid technology costs and AEO2020 reference load projections with carbon tax of \$150/tonne (Cohen and Durvasulu, 2021). The average price is \$36.34/MWh. There are high fluctuations throughout the year. Low electricity prices are available during spring time. The prices are generally higher during the summer.



*Figure S16: Electricity Price Predictions from NREL-MiNg\_\$150\_ERCOT*

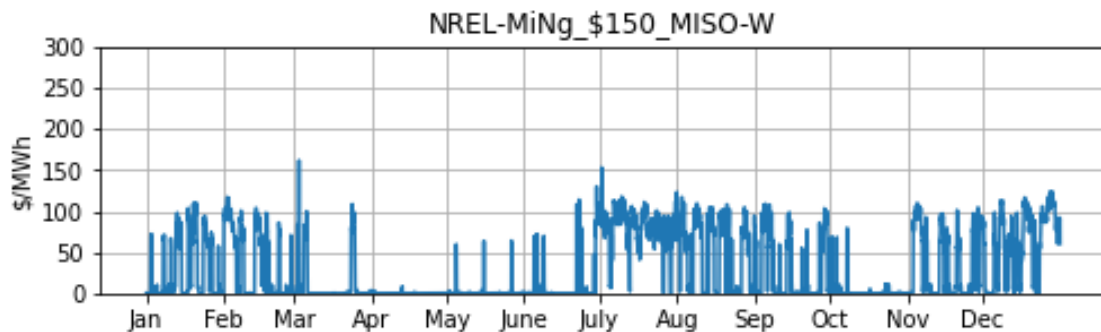
- NREL-MiNg\_\$100\_MISO-W: This NREL scenario predicts 2035 electricity prices

for Midcontinent Independent System Operator (MISO) region based on ATB2020 mid technology costs and AEO2020 reference load projections with carbon tax of \$100/tonne (Cohen and Durvasulu, 2021). The average price is \$35.22/MWh. There are high fluctuations throughout the year. Low electricity prices are available during the spring time. The prices are generally higher during the summer.



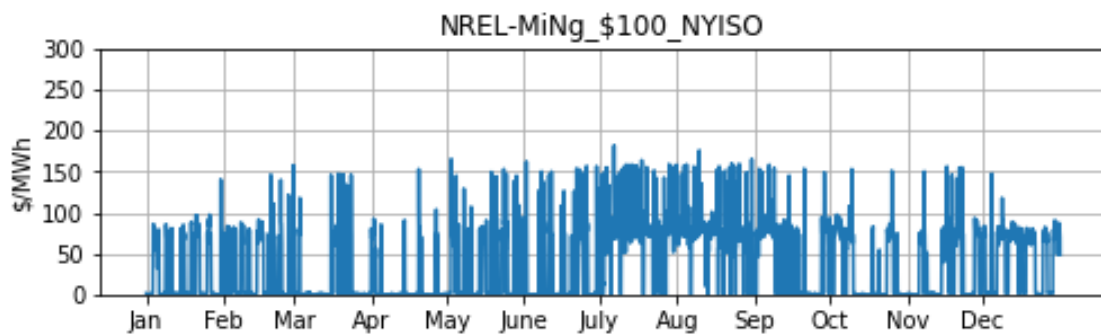
*Figure S17: Electricity Price Predictions from NREL-MiNg\_\$100\_MISO-W*

- NREL-MiNg\_\$150\_MISO-W: This NREL scenario predicts 2035 electricity prices for Midcontinent Independent System Operator (MISO) region based on ATB2020 mid technology costs and AEO2020 reference load projections with carbon tax of \$150/tonne (Cohen and Durvasulu, 2021). The average price is \$28.28/MWh. There are high fluctuations throughout the year. Low electricity prices are available during the spring and fall. The prices are generally higher during the summer.



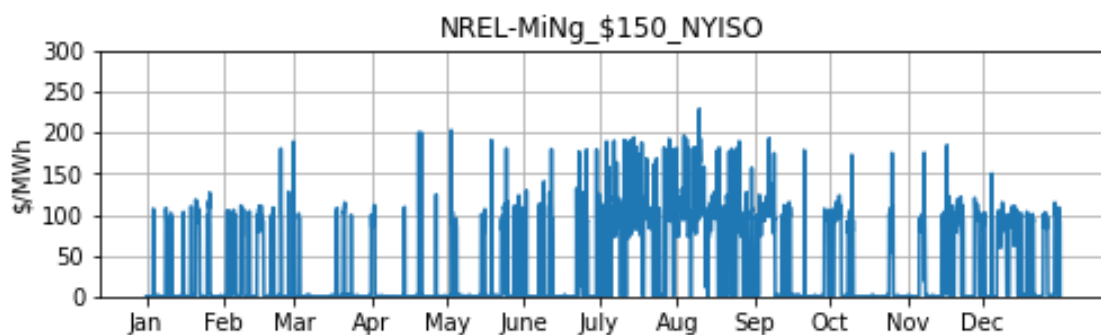
*Figure S18: Electricity Price Predictions from NREL-MiNg\_\$150\_MISO-W*

- NREL-MiNg\_\$100\_NYISO: This NREL scenario predicts 2035 electricity prices for New York Independent System Operator (NYISO) region based on ATB2020 mid technology costs and AEO2020 reference load projections with carbon tax of \$100/tonne (Cohen and Durvasulu, 2021). The average price is \$35.33/MWh. There are high fluctuations throughout the year. Low electricity prices are available during the spring and fall. The prices are generally higher during the summer.



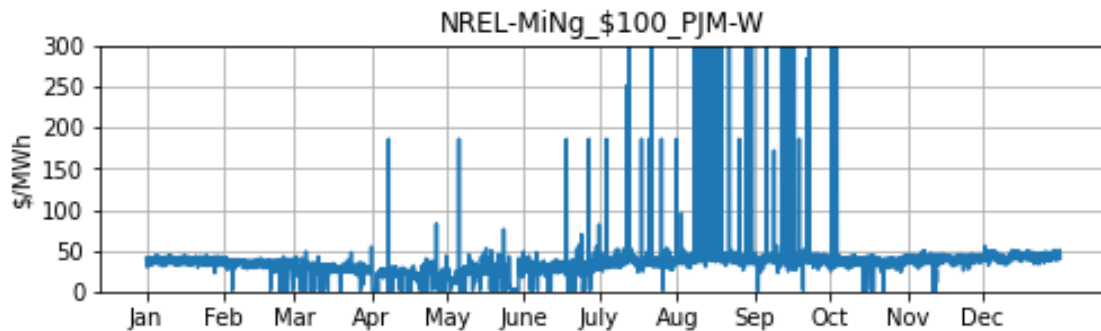
*Figure S19: Electricity Price Predictions from NREL-MiNg\_\$100\_NYISO*

- NREL-MiNg\_\$150\_NYISO: This NREL scenario predicts 2035 electricity prices for New York Independent System Operator (NYISO) region based on ATB2020 mid technology costs and AEO2020 reference load projections with carbon tax of \$150/tonne (Cohen and Durvasulu, 2021). The average price is \$33.84/MWh. There are high fluctuations throughout the year. Low electricity prices are available during the spring and fall. The prices are generally higher during the summer.



*Figure S20: Electricity Price Predictions from NREL-MiNg\_\$150\_NYISO*

- NREL-MiNg\_\$100\_PJM-W: This NREL scenario predicts 2035 electricity prices for Pennsylvania-New Jersey-Maryland (PJM) region based on ATB2020 mid technology costs and AEO2020 reference load projections with carbon tax of \$100/tonne (Cohen and Durvasulu, 2021). The average price is \$42.00/MWh. Slightly lower electricity prices are available during the spring. There are spikes of high prices during the summer.



*Figure S21: Electricity Price Predictions from NREL-MiNg\_\$100\_PJM-W*

- NREL-MiNg\_\$150\_PJM-W: This NREL scenario predicts 2035 electricity prices for Pennsylvania-New Jersey-Maryland (PJM) region based on ATB2020 mid technology costs and AEO2020 reference load projections with a carbon tax of \$150/tonne (Cohen and Durvasulu, 2021). The average price is \$49.94/MWh. There are high fluctuations throughout the year. Low electricity prices are available during the spring and fall. The prices are generally higher during the summer.

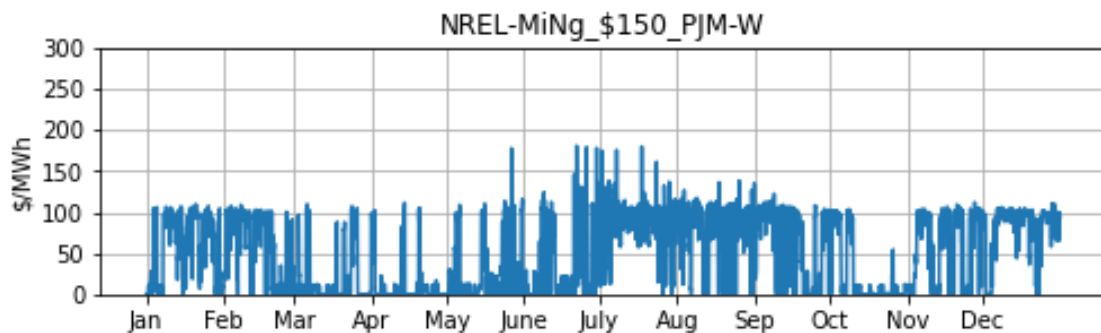


Figure S22: Electricity Price Predictions from NREL-MiNg\_\$150\_PJM-W

### SI6: Optimization Model

In order to explore the economic benefits of dynamic operations, an optimization framework is formulated with two objectives: 1) Find the plant capacity that is big enough to allow dynamic operations but not too big to overblow the capital costs; 2) Find the operating current densities that minimize the power usage and the total cost.

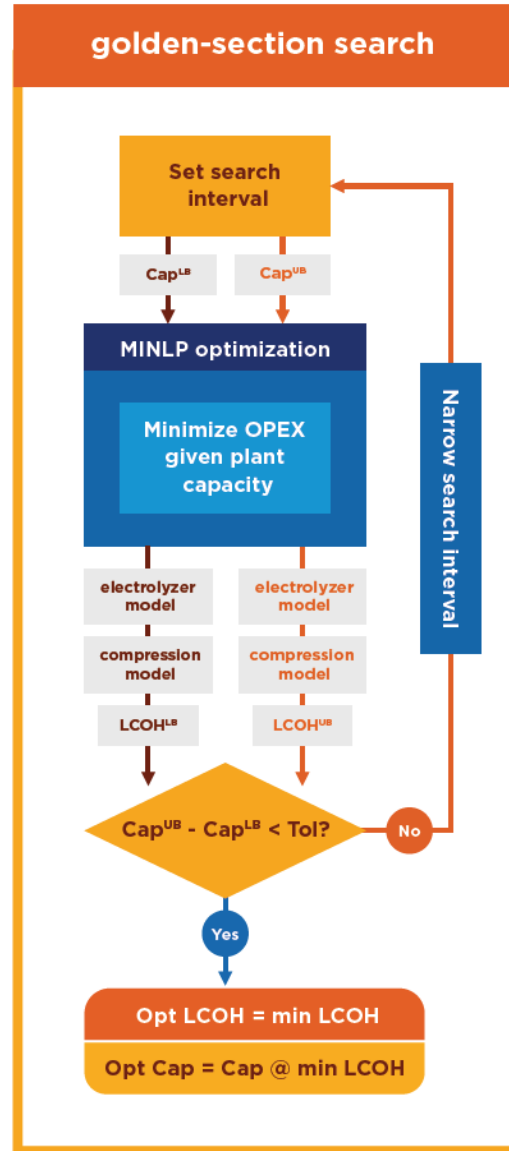


Figure S23: Diagram of the Optimization Framework. The outer-loop Golden-Section Search algorithm (the yellow frame) finds the plant production capacity (tonne/day) that results in the minimum LCOH between an upper bound (UB) and a lower bound (LB) defined by the user. The inner-loop Mixed Integer Non-Linear Programming (MINLP) (the blue box) finds the operating

conditions that result in the minimum LCOH for a given plant capacity.

In order to achieve these objectives, the optimization framework utilizes two optimization algorithms, as shown in Figure S23. In the yellow outer-loop, a Golden-Section Search (GSS) algorithm chooses two plant capacities to test, based on the initial search interval. The plant capacities are sent to the blue inner-loop for evaluation. The blue inner-loop uses a Mixed-Integer Non-Linear Programming (MINLP) implemented with Gurobi optimizer to find the operating parameters that result in the minimum LCOH for a given plant capacity. Based on the LCOH values from the two plant capacities, a new search interval is set for the GSS algorithm. If the search interval is less than the desired tolerance level, then the algorithm terminates and the optimal capacity is set to be the minimum of all the search attempts. Otherwise, the GSS algorithm recursively carries on with the new search interval.

For the MINLP, the electrolyzer techno-economic model is formulated into an optimization problem. The problem is designed to find the operating current density and hydrogen storage size for a given electrolyzer plant size and a target production rate. Hence, the decision variables are as follows:

$$j_{op}(t) = [0, j_{max}] \quad \text{Eq. S28}$$

$$S_{max} \geq 0 \quad \text{Eq. S29}$$

The operating current density at time  $t$ ,  $j_{op}(t)$ , has to be greater than 0 and less than or equal to the maximum current density allowed by design. The size of the hydrogen storage tank has to be greater than or equal to 0. The optimization algorithm determines the operating current density at each hour and the storage size that minimizes the net present value of the total cost. Therefore, the objective function can be written as:

$$\begin{aligned} & \text{minimize } NPV(C_{Total}) \\ & = \text{minimize } C_{CAPEX} + C_{storage} + \sum_{y=1}^{40} \left[ (C_{PR} + C_{UPR} + C_{fOPEX} + C_{vOPEX}) \frac{(1 + \pi)^y}{(1 + d)^y} \right] \end{aligned} \quad \text{Eq. S30}$$

In Eq. S30,  $C_{CAPEX}$  stays constant for a given electrolyzer size, which is already determined by the GSS algorithm.  $C_{storage}$  increases linearly with the size of storage. The total replacement cost, which is the sum of  $C_{PR}(y)$  and  $C_{UPR}(y)$ , is proportional to the direct capital cost, which stays constant for a given electrolyzer size. Finally,  $C_{fOPEX}(y)$  comprises of labor cost, overhead cost, tax and insurance cost, and material cost. Labor and overhead costs are fixed for each calendar year. Tax and insurance cost is proportional to  $C_{CAPEX}$ . Material cost is proportional to the direct capital cost. As a result,  $C_{fOPEX}(y)$  does not change as a function of the decision variables. Therefore, Eq. S30 can be simplified as:

$$\text{minimize } C_{\text{storage}} + \sum_{y=1}^{40} \left[ C_{\text{vOPEX}} \frac{(1 + \pi)^y}{(1 + d)^y} \right]$$

Eq. S31

where

$$C_{\text{storage}} = \text{Unit Cost Factor} \times \text{Hydrogen Weight at Capacity}$$

Eq. S32

$$C_{\text{vOPEX}} = \sum_{d=0}^{364} \sum_{t=24d}^{24(d+1)-1} P_{\text{elec}}(t) \times \text{Power}_{\text{op}}(j_{\text{op}}(t)) + P_{\text{elec}}(t) \times U_{\text{elec}}^{\text{hourly}} \times \dot{m}_{\text{H}_2, \text{prod}}^{\text{hourly}}(t) \\ + P_{\text{water}} \times U_{\text{water}}^{\text{hourly}} \times \dot{m}_{\text{H}_2, \text{prod}}^{\text{hourly}}(t)$$

Eq. S33

The operating power,  $\text{Power}_{\text{op}}(t)$ , is a non-linear function of the operating current density,  $j_{\text{op}}(t)$ . For implementation in the Gurobi solver, the non-linear power function is linearized with five piecewise linear functions as shown in Figure S24.

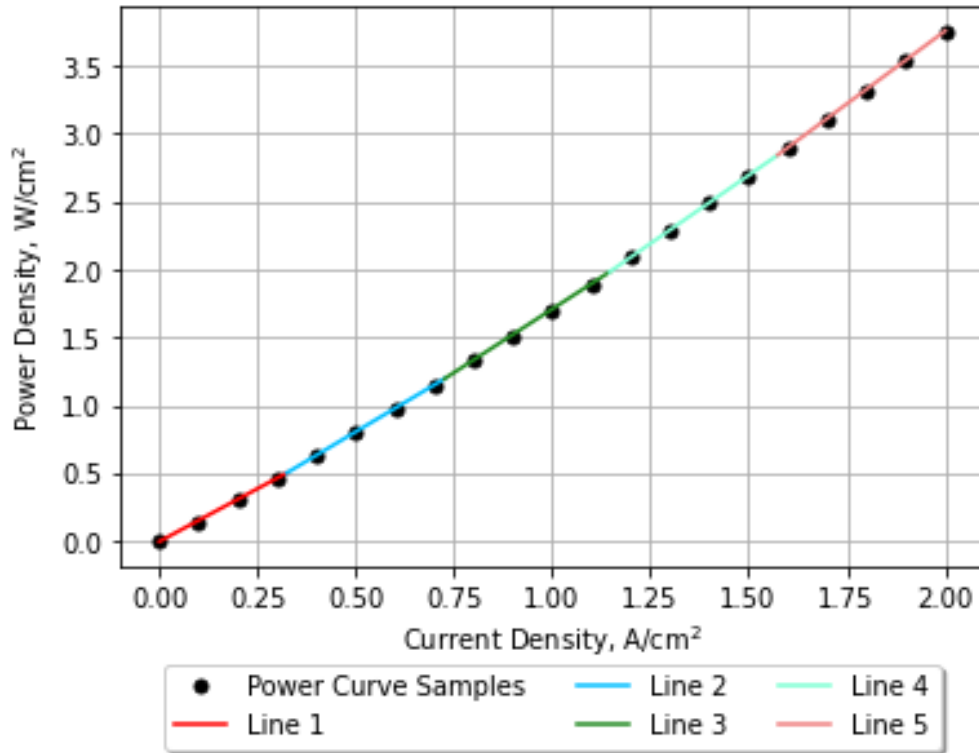


Figure S24: The power curve showing the operating power as a function of current density for a given electrolyzer area. The black dots are sampled data from the electrolyzer model. The colored

lines are piece-wise linear functions that are fitted to the model.

Each piecewise linear power function,  $l$ , is implemented in the optimization formulation by introducing a binary variable  $w_l$ :

$$j_{op}(t) \geq x_{1,l} - M \times (1 - w_l(t)) \quad \forall l = 1,2,3,4,5 \quad \text{Eq. S34}$$

$$j_{op}(t) \leq x_{2,l} + M \times (1 - w_l(t)) \quad \forall l = 1,2,3,4,5 \quad \text{Eq. S35}$$

$$Power_{op}(t) \geq Slope_l \times j_{op}(t) + Intercept_l - M \times (1 - w_l(t)) \quad \forall l = 1,2,3,4,5 \quad \text{Eq. S36}$$

$$Power_{op}(t) \leq Slope_l \times j_{op}(t) + Intercept_l + M \times (1 - w_l(t)) \quad \forall l = 1,2,3,4,5 \quad \text{Eq. S37}$$

$$\sum_{l=1}^5 w_l(t) = 1 \quad \text{Eq. S38}$$

Eq. S34 and Eq. S5 activate the linear function that corresponds to the operating current density at each hour. For example, if the current density is 0.25 A/cm<sup>2</sup>, then  $w_1(t) = 1$  and Line 1 is activated. Eq. S36 and Eq. S37 calculate the power density corresponding to the operating current density. Finally, Eq. S38 ensures that the model is able to pick only one of the five lines for power calculations.

The hourly hydrogen production,  $\dot{m}_{H_2}^{hourly}(t)$  in Eq. S39, is directly proportional the operating current density:

$$\dot{m}_{H_2,prod}^{hourly}(t) = \frac{j_{op}(t) \times A}{2F} \text{mole} \times \frac{3600 \text{ s}}{1 \text{ hr}} \times \frac{1 \text{ kg}}{1000 \text{ g}} \times \frac{2.02 \text{ g}}{1 \text{ mole}}$$

Eq. S39

The produced hydrogen can either be stored or exit the system for sales, as shown in Figure S25.

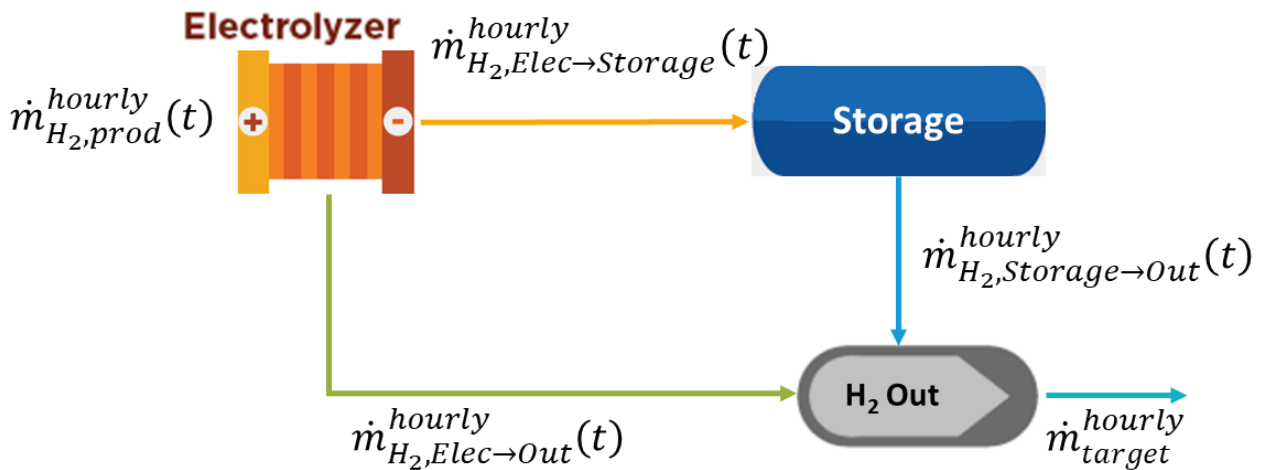


Figure S25: Illustration of hydrogen flow paths from electrolyzer to storage to system outlet.

There is a target amount of hydrogen that needs to be delivered. The demand can be met by production from the electrolyzer and/or discharge from the storage. If it is deemed economical to produce and store excess hydrogen at certain hours (when the electricity is cheap) and discharge it later, the system is allowed to do so. For simplicity, the storage can only be charged or discharged, not both at the same time. These mass balance principles and the charging-discharging dual mode of storage are implemented as follows:

$$\dot{m}_{H_2,prod}^{hourly}(t) \geq \dot{m}_{target}^{hourly} - M \times (1 - ws(t)) \quad \text{Eq. S40}$$

$$\dot{m}_{H_2,prod}^{hourly}(t) \leq \dot{m}_{target}^{hourly} - M \times ws(t) \quad \text{Eq. S41}$$

$$\dot{m}_{H_2,Storage \rightarrow Out}^{hourly}(t) = \left( \dot{m}_{target}^{hourly} - \dot{m}_{H_2,prod}^{hourly}(t) \right) \times (1 - ws(t)) \quad \text{Eq. S42}$$

$$\dot{m}_{H_2,Elec \rightarrow Storage}^{hourly}(t) = \left( \dot{m}_{H_2,prod}^{hourly}(t) - \dot{m}_{target}^{hourly} \right) \times ws(t) \quad \text{Eq. S43}$$

$$\dot{m}_{H_2,prod}^{hourly}(t) = \dot{m}_{H_2,Elec \rightarrow Storage}^{hourly}(t) + \dot{m}_{H_2,Elec \rightarrow Out}^{hourly}(t) \quad \text{Eq. S44}$$

$$\dot{m}_{H_2,Elec \rightarrow Out}^{hourly}(t) + \dot{m}_{H_2,Storage \rightarrow Out}^{hourly}(t) \geq \dot{m}_{target}^{hourly} \quad \text{Eq. S45}$$

Eq. S40 and Eq. S41 are used to determine whether the electrolyzer is producing an excess amount of hydrogen compared to the target rate. Eq. S42 and Eq. S43 ensure that the storage is either in a charging mode or a discharging mode with a binary variable  $ws(t)$ . Eq. S44 and Eq. S45 ensure the mass balance illustrated in Figure S25.

The storage facility itself is subject to three constraints:

$$S(t) = S(t - 1) + \dot{m}_{H_2,Elec \rightarrow Storage}^{hourly}(t) - \dot{m}_{H_2,Storage \rightarrow Out}^{hourly}(t) \quad \forall t \neq 0 \quad \text{Eq. S46}$$

$$S(1) = S(8760) + \dot{m}_{H_2,Elec \rightarrow Storage}^{hourly}(1) - \dot{m}_{H_2,Storage \rightarrow Out}^{hourly}(1) \quad \text{Eq. S47}$$

$$S(t) \leq S_{max} \quad \text{Eq. S48}$$

Eq. S46 ensures the mass balance for storage. Eq. S47 is a special case of Eq. S46 which requires the system to end with the same storage amount as it started with for each calendar year. Eq. S48 defines the maximum storage capacity, which feeds into the storage capital cost in the objective function (Eq. S31). Additional operating or compression costs associated with storage have not been considered in this study.

Another constraint that is considered in this study is the minimum operating current density limit. For safety and efficiency considerations, there may be a limit to how low the operating current density can drop. In such a case, rather than operating at a current density below the minimum threshold, the electrolyzer should shut down. These constraints are implemented in the model as follows:

$$j_{op}(t) \geq j_{min} - M \times (1 - w(t)) \quad \text{Eq. S49}$$

$$j_{op}(t) \leq 0 + M \times w(t) \quad \text{Eq. S50}$$

Eq. S49 determines whether the operating current density is below the minimum. If so, Eq. S50 sets the operating current density to 0.

In addition, there are non-negativity constraints introduced in the formulation since none of the variables discussed above can be negative in value.

$$j_{op}(t) \geq 0 \quad \text{Eq. S51}$$

$$S(t) \geq 0 \quad \text{Eq. S52}$$

$$\dot{m}_{H_2,prod}^{hourly}(t) \geq 0 \quad \text{Eq. S53}$$

$$\dot{m}_{H_2,Elec \rightarrow Storage}^{hourly}(t) \geq 0 \quad \text{Eq. S54}$$

$$\dot{m}_{H_2,Storage \rightarrow Out}^{hourly}(t) \geq 0 \quad \text{Eq. S55}$$

$$\dot{m}_{H_2,Elec \rightarrow Out}^{hourly}(t) \geq 0 \quad \text{Eq. S56}$$

While the MINLP algorithm looks for the optimal operating current densities and hydrogen storage size to minimize the operating cost and LCOH for a given electrolyzer plant size, the GSS algorithm looks for the optimal plant size that results in the overall lowest LCOH. As shown in Figure S26, the GSS algorithm searches for an extremum of a unimodal function within a specified interval, between a lower bound,  $x_1$ , and an upper bound,  $x_2$ . It evaluates the function at two different points,  $x_3$  and  $x_4$ . The two points are determined such that:

$$\frac{x_2 - x_3}{x_3 - x_1} = \varphi = \frac{1 + \sqrt{5}}{2} \quad \text{Eq. S57}$$

$$\frac{x_4 - x_1}{x_2 - x_4} = \varphi = \frac{1 + \sqrt{5}}{2} \quad \text{Eq. S58}$$

The algorithm looks at the values of  $y_3=f(x_3)$  and  $y_4=f(x_4)$ . If  $y_4 > y_3$ , then the new search interval is set between  $x_1$  and  $x_4$ . If  $y_3 > y_4$ , then the new search interval is set between  $x_3$  and  $x_2$ . The new search interval is shaded in light blue in Figure 5.4 for the two different scenarios. The algorithm continues until

the search interval is sufficiently narrowed down to a desired tolerance. The extreme point (minimum in the case shown in Figure S26) is taken as the minimum of all the search iterations that the algorithm performed.

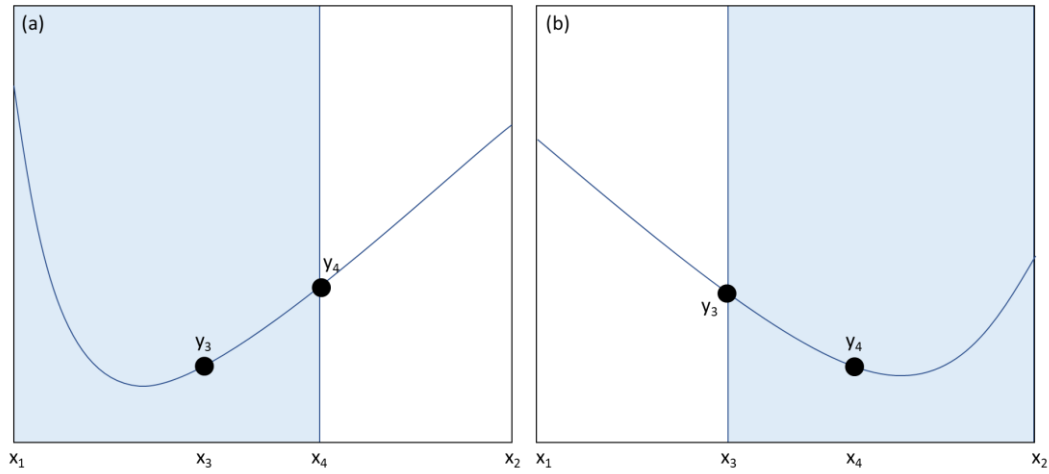


Figure S26: Illustration of how the GSS algorithm works with two unimodal functions with different shapes. In (a), the upper bound of the search interval is updated to  $x_4$  from  $x_2$ . In (b), the lower bound of the search interval is updated to  $x_3$  from  $x_1$ .

For each dynamic operations scenario, the optimization model produces a result that looks like Figure S27. The green triangle is the LCOH in static operation mode where the electrolyzer capacity is sized to exactly match the demand rate. The blue triangles are the different search iterations that the GSS algorithm evaluated, each triangle representing the solution from the MINLP. The red triangle indicates the lowest LCOH of all cases evaluated. This plot shows that dynamic operations with 40% oversized capacity reduces the LCOH by 9% from \$4.93/kg to \$4.48/kg.

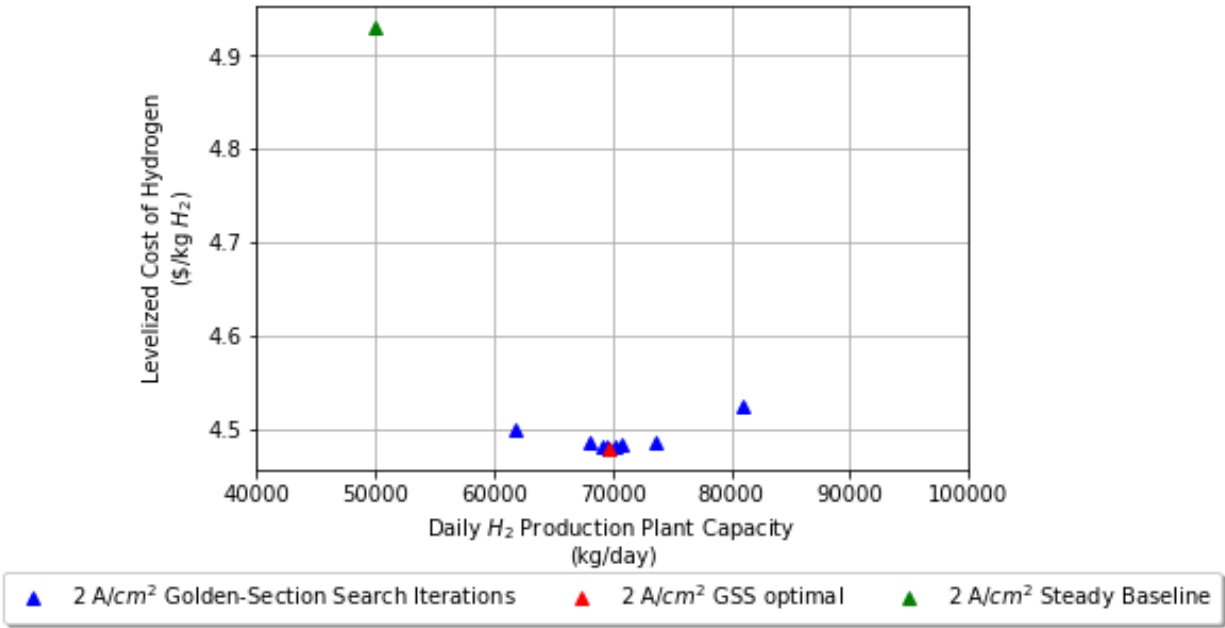


Figure S27: Plot of LCOH as a function of daily production plant capacity with a maximum current density of 2 A/cm<sup>2</sup>. The blue points are iterations evaluated by the GSS algorithm. The red point is the optimal case with the lowest total LCOH. The green point is the LCOH from steady operation.

Table S9: Nomenclature for MINLP Optimization Algorithm

|               |                                                          |
|---------------|----------------------------------------------------------|
| $A$           | Total Active Area of the Electrolyzer (cm <sup>2</sup> ) |
| $C_{CAPEX}$   | Capital Expenditure (CAPEX) (\$)                         |
| $C_{fOPEX}$   | Fixed Operating Expenses (OPEX) (\$)                     |
| $C_{PR}$      | Planned Replacement Cost (\$)                            |
| $C_{storage}$ | Storage Cost (\$)                                        |
| $C_{Total}$   | Total Cost (\$)                                          |
| $C_{UPR}$     | Unplanned Replacement Cost (\$)                          |
| $C_{vOPEX}$   | Variable Operating Expenses (OPEX) (\$)                  |
| $d$           | Discount Rate (fraction)                                 |
| $F$           | Faraday's Number (96485.3 C/mol e <sup>-</sup> )         |
| $j_{max}$     | Maximum Current Density (A/cm <sup>2</sup> )             |

|                                                    |                                                             |
|----------------------------------------------------|-------------------------------------------------------------|
| $j_{min}$                                          | Minimum Current Density (A/cm <sup>2</sup> )                |
| $j_{op}$                                           | Operating Current Density (A/cm <sup>2</sup> )              |
| $M$                                                | “Big M”                                                     |
| $\dot{m}_{H_2, Elec \rightarrow Out}^{hourly}$     | Hourly Hydrogen Flow from Electrolyzer to Outlet (kg/hr)    |
| $\dot{m}_{H_2, Elec \rightarrow Storage}^{hourly}$ | Hourly Hydrogen Flow from Electrolyzer to Storage (kg/hr)   |
| $\dot{m}_{H_2, Storage \rightarrow Out}^{hourly}$  | Hourly Hydrogen Flow from Storage to Outlet (kg/hr)         |
| $\dot{m}_{target}^{hourly}$                        | Hourly Hydrogen Production Target (kg/hr)                   |
| $P_{elec}$                                         | Electricity Price (\$/MWh)                                  |
| $P_{water}$                                        | Price of Water (\$/gallon)                                  |
| $Power_{op}$                                       | Operating Power (MW)                                        |
| $S$                                                | Hydrogen Storage (tonne)                                    |
| $S_{max}$                                          | Maximum Hydrogen Storage Capacity (tonne)                   |
| $U_{elec}^{hourly}$                                | Hourly Balance of Plant Electrical Usage (kW/hour)          |
| $U_{water}^{hourly}$                               | Hourly Water Usage (gallon/kg H <sub>2</sub> )              |
| $w$                                                | Binary Variable for Minimum Current Density                 |
| $w_l$                                              | Binary Variable for Power Function $l$                      |
| $ws$                                               | Binary Variable for Charging-Discharging Mode for Storage   |
| $x_{1,l}$                                          | Lower Limit of Current Density Range for Power Function $l$ |
| $x_{2,l}$                                          | Upper Limit of Current Density Range for Power Function $l$ |
| $\pi$                                              | Inflation Rate (fraction)                                   |

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