

Hydroxide exchange membrane carbon capture using a nickel hydroxide symmetric battery cell

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Abstract

Electrochemical carbon capture devices can be a low energy cost solution for direct air capture (DAC) using renewable electricity. Historically electrochemical carbon-capture has targeted a range of concentrations from atmospheric ($400 \text{ ppm}_{\text{CO}_2}$) (DAC),¹ to life-support ($5000 \text{ ppm}_{\text{CO}_2}$),² to point-source capture ($10\% \text{ CO}_2$).³ The hydroxide exchange membrane nickel hydroxide symmetric battery cell with two identical electrodes has low voltage requirements making it more suitable for DAC than other electrochemical approaches. A 25 cm^2 laboratory cell shows an average energy cost of $1.15 \text{ MWh} \cdot \text{ton}_{\text{CO}_2}^{-1}$ and a CO_2 flux of $78 \text{ kg}_{\text{CO}_2} \cdot \text{m}^2 \cdot \text{yr}^{-1}$ at $2 \text{ mA} \cdot \text{cm}^{-2}$. A manufacturable 25 cm^2 cell is durability tested for 5000 hours and achieves an energy of $0.46 \text{ MWh} \cdot \text{ton}_{\text{CO}_2}^{-1}$ and a flux of $62 \text{ kg}_{\text{CO}_2} \cdot \text{m}^2 \cdot \text{yr}^{-1}$ at the end of the test. A DAC pilot system with a stack of 9 scaled-up 300 cm^2 cells demonstrates an energy of $0.83 \text{ MWh} \cdot \text{ton}_{\text{CO}_2}^{-1}$ and a flux of $75 \text{ kg}_{\text{CO}_2} \cdot \text{m}^2 \cdot \text{yr}^{-1}$ and meets the 300 Pa pressure drop required for DAC.⁴ Modular design projects improvements in cost from manufacturing and economies of scale, and a pathway to below $100 \$ \cdot \text{ton}_{\text{CO}_2}^{-1}$ from learning rates of past energy technologies.⁵

Introduction

Direct air capture (DAC) has been identified as one of the key net negative carbon technologies to achieve a net zero future.^{6,7} DAC is required to offset continued emissions from dilute CO_2 sources e.g., agriculture and construction.⁸ The majority of current DAC technologies at scale ($> 1 \text{ kton}_{\text{CO}_2} \cdot \text{yr}^{-1}$) are sorbent based with significant energy cost.⁹ The energy cost is the major operating expense and is primarily driven by the temperature swing required to regenerate the sorbent.^{10,11} The United States Department of Energy (DOE) has set forth a cost target of $100 \$ \cdot \text{ton}_{\text{CO}_2}^{-1}$ which is not possible to meet with current industrial electricity costs near $70 \$ \cdot \text{MWh}^{-1}$ and current temperature swing adsorption devices energy costs above $1.5 \text{ MWh} \cdot \text{ton}_{\text{CO}_2}^{-1}$.^{12,13} A combination of lower electricity costs and lower DAC energy costs are required to meet the target DAC cost.

Electrochemical pH gradient devices are a growing research area for low energy cost carbon capture.^{1,14–19} The pH gradient is built by generating hydroxide (OH^-) at the cathode and consuming OH^- at the anode. An acid-base equilibrium with CO_2 allows for the capture of CO_2 at the cathode and release at the anode.²⁰ This compliments other electrochemical CO_2 capture devices based on pK_a gradients of an electrochemically active species allowing for the capture and release of CO_2 .^{2,21,22}

This work explores a hydroxide exchange membrane carbon capture (HEMCC) device with symmetric $\text{Ni}(\text{OH})_2$ electrodes to produce the pH gradient for CO_2 capture and release (Fig. 1a). At the cathode NiOOH is reduced to $\text{Ni}(\text{OH})_2$ while at the anode $\text{Ni}(\text{OH})_2$ is oxidized to NiOOH . This redox chemistry is known for its reversibility and cycle durability and has been commercially used in the cathode of alkaline Ni-MH battery technologies.^{23–25} This symmetric cell has a thermodynamic equilibrium voltage of zero. Most of the voltage observed is to produce the pH gradient with the remainder driving the polarization of the

electrodes. There is a resistance component as well, but this is small due to the low current densities used in the device, typically $< 5 \text{ mA}\cdot\text{cm}^{-2}$.

Scaleup is necessary for DAC technologies with several orders of magnitude growth required to meet net zero targets.²⁶ This HEMCC device was first developed at a 25 cm^2 laboratory scale able to capture $200 \text{ g}_{\text{CO}_2}\cdot\text{yr}^{-1}$ (Fig. 1b). It was built with electrodes formed by electrochemical precipitation of nickel hydroxide on nickel foam. A manufacturable version of the 25 cm^2 electrode was developed and durability tested for 5000 hours. This durable electrode was scaled to 300 cm^2 and integrated into a 9-cell stack for a pilot DAC system (Fig. 1c). The first commercial modular DAC system is under construction for operation in 2026 with a capacity target of $1 \text{ kton}_{\text{CO}_2}\cdot\text{yr}^{-1}$ (Fig. 1d).

Results and Discussion

Figure 2 shows the data of a typical laboratory 25 cm^2 cell. Figures 2a,b are time series of the operating cell voltage and cathode outlet CO_2 concentration with an initial conditioning cycle followed by ten regular cycles, all at $2 \text{ mA}\cdot\text{cm}^{-2}$. Within each regular cycle there are two steps, one for the charge/discharge and the other for the regeneration. CO_2 is captured in both steps, but at different voltages. The cell voltage and the difference between the 400 ppm cathode inlet and cathode outlet CO_2 allow for calculation of key process parameters of capacity, electron efficiency, energy cost, and flux. Figure 2c-f presents these parameters excluding the regeneration steps. Capacity tracks long-term changes in the device. Electron efficiency is defined here as the ratio of CO_2 molecules captured per electron passed in the cell with a maximum stoichiometric ratio of 1 CO_2 per electron. Energy cost is the largest operating cost of the device while the flux determines the size and thus capital cost of the system. An average of $1.15 \text{ MWh}\cdot\text{ton}_{\text{CO}_2}^{-1}$ energy cost including the regeneration step was achieved in this single cell dataset with a flux of $78 \text{ kg}_{\text{CO}_2}\cdot\text{m}^{-2}\cdot\text{yr}^{-1}$. Each cycle was integrated using the definitions in SI Discussion 5 and the mean was taken excluding the conditioning cycles and assuming all CO_2 that is captured on the cathode is released in the anode to a product stream. The HEMCC has a lower energy cost than the incumbent technology presented by Carbon Engineering of $1.47 \text{ MWh}\cdot\text{ton}_{\text{CO}_2}^{-1}$ for their device when excluding the blower and compressor.

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Figure 2 | Laboratory scale 25 cm^2 HEMCC and its behavior. The HEMCC is comprised of two identical electrochemically precipitated $\text{Ni}(\text{OH})_2$ electrodes and an $80\mu\text{m}$ Piperion® membrane. The cathode air flow was $1 \text{ L}\cdot\text{min}^{-1}$ with 400 ppm CO_2 and 90% RH. The anode received a sweep gas of 90% RH N_2 at $1 \text{ L}\cdot\text{min}^{-1}$. a. The cell voltage (V) and b. the cathode outlet CO_2 concentration (ppm) are used to calculate the key process variables c-f during the charge/discharge step

c. capacity, d. electron efficiency, e. energy, and f. flux, all excluding the regeneration step. Regeneration steps are excluded to highlight the low energy cost of the charge-discharge step. Capture still occurs during regeneration but at higher energy cost. The first cycle on each side is a 1.5-hour regeneration hold

for break-in followed by 10–75 minute cycles on each electrode. The cycle length is chosen based on an ex-situ capacity test to minimize regeneration (**SI Discussion 7**). **c.** Capacity achieved an average of $1.89 \text{ mAh}\cdot\text{cm}^{-2}$ which was 85% of the ex-situ capacity tests. **d.** Electron efficiency achieved 0.25. **e.** Energy cost averaged $0.76 \text{ MWh}\cdot\text{ton}_{\text{CO}_2}^{-1}$ while **f.** flux averaged to $72 \text{ kg}_{\text{CO}_2}\cdot\text{m}^2\cdot\text{yr}^{-1}$. **g.** and **h.** show a single cycle to highlight the change in voltage and capture in the charge/discharge and regeneration steps. **i., j., and k.** are graphic aids to describe the build ΔpH , maldistributed ΔpH , and rebuild ΔpH sections in **g.** and **h.** The ΔpH between the peak pH at the cathode and the CO_2 release pH at the anode drives the CO_2 capture rate in the cell, see **SI Discussion 2**. The CO_2 release pH is the pH that achieves equilibrium between CO_2 in the anode and the concentrated CO_2 gas product stream. The dotted red primary reaction coordinate lines are rough position where OH^- is produced in the cathode and consumed in the anode. **i.** The build ΔpH section is directly after the gas lines switch in the cell. Redox reactions take place close to the membrane with the cathode near 100% state of charge and the anode near 0% state of charge. The pH at the back of the cathode electrode is close to the pH of the anode in the previous cycle. **j.** The maldistributed ΔpH section is at the end of a charge/discharge step. This occurs because of the uneven mass loading of $\text{Ni}(\text{OH})_2/\text{NiOOH}$ when making the electrode and uneven distribution of resistance through the electrode. Near the end of the charge/discharge step, low loading and low resistance volumes of the cathode will have fully been consumed, no longer producing OH^- . Surrounding higher loading and higher resistance volumes continue producing OH^- but the average pH across the electrode drops lowering the average driving force for capturing CO_2 . The anode in this section is still charging and has not reached 100% state of charge because of competing OER. **k.** The rebuild ΔpH section is the beginning of the regeneration after the cathode has reached 0% state of charge. The cathode reaction coordinate is brought to the front towards the membrane where catalytic ORR reactions are predominant. The full area of the electrode is used again, and the average pH increases to have a higher rate of CO_2 capture. The anode continues to charge towards 100% for the next cycle.

Relatively low energy cost is the main benefit of this electrochemical carbon capture approach. Figure 2e shows several cycles of CO_2 capture at an average of $1.15 \text{ MWh}\cdot\text{ton}_{\text{CO}_2}^{-1}$; when broken down by the steps the charge/discharge and regeneration steps averaged $0.76 \text{ MWh}\cdot\text{ton}_{\text{CO}_2}^{-1}$ and $2.15 \text{ MWh}\cdot\text{ton}_{\text{CO}_2}^{-1}$ respectively. Both steps can be improved by improving the electron efficiency of the device.¹⁴ Electron efficiency of 1 indicates ionic transport of only HCO_3^- in the cell where 0.5 would be predominantly CO_3^{2-} , below 0.5 as observed in this cell has either unreacted OH^- being transported or back diffusion of HCO_3^- or CO_3^{2-} from the anode. Future work can improve electron efficiency by adjusting membrane parameters such as ionic exchange capacity (IEC), conductivity, and water uptake. Improved electron efficiency at the same current density directly increases flux of CO_2 which decreases energy and capital costs.

The CO_2 capture rate changes with time and the state of charge of the electrode. Figure 2h shows the single half cycle behavior of CO_2 capture. At the beginning of the charge/discharge step there is time lag as the pH gradient is reversed from the previous cycle. A steady increase in capture rate is seen as the pH gradient is formed, which takes time proportional to the total IEC of the membrane and ionomer. Piperion®

is used for the membrane and ionomer based on PAP-TP-85 with an IEC of $2.37 \text{ meq} \cdot \text{g}^{-1}$.²⁹ The state of charge determines the CO_2 capture behavior for the remainder of the charge/discharge step. The surface of the electrode near the membrane discharges first due to the difference in conductivity between the metal, metal hydroxide, and ionomer phases of the electrode.^{30,31} The primary reaction coordinate, and thus the peak pH, moves from the membrane to the back of the electrode as the state of charge changes in each electrode. At a moderate state of charge, higher capture rates are expected because CO_2 diffuses into and reacts with a larger volume of high pH ionomer. A similar behavior was seen in a hydrogen HEMCC previously studied by using an interlayer volume.¹⁵ Near the end of the cycle volumes of anode with lower mass loading of $\text{NiOOH}/\text{Ni}(\text{OH})_2$ or lower local resistance will run out of NiOOH earlier than the surrounding volumes and no longer produce OH^- ; these volumes no longer contribute to the pH gradient and lower the average pH at the primary reaction coordinate. This manifests in a dip in CO_2 capture near the end of the charge/discharge step. During the charge/discharge step the flux averages $72 \text{ kg}_{\text{CO}_2} \cdot \text{m}^{-2} \cdot \text{yr}^{-1}$, while the average peak flux is $97 \text{ kg}_{\text{CO}_2} \cdot \text{m}^{-2} \cdot \text{yr}^{-1}$.

The capture rates during regeneration do not depend on the state of charge of the cathode which has been depleted. The pH gradient is reestablished by a relatively thin layer of the electrode near the membrane performing catalytic ORR.^{30,31} There is a short time dependent growth of capture at the beginning of regeneration to reestablish the pH gradient across the area of the electrode after the decrease observed in the charge/discharge step. After that a steady state constant capture rate is achievable. During the regeneration step the flux averages $93 \text{ kg}_{\text{CO}_2} \cdot \text{m}^{-2} \cdot \text{yr}^{-1}$, while the average peak flux is $97 \text{ kg}_{\text{CO}_2} \cdot \text{m}^{-2} \cdot \text{yr}^{-1}$.

Technical pathways to improve the device include improving mass transport (diffusion) from the flow field channels into and within the porous electrodes, decreasing the effect of time lag for CO_2 and decreasing the requirement for regeneration. Decreasing the effect of time lag can improve both the flux and energy cost of the cycle. There are two ways to reduce the time lag, one by decreasing the total ion capacity of membrane and electrode or the other by increasing the electrode capacity. Either option has a trade-off to optimize. Decreasing the total ion capacity by decreasing membrane thickness and ionomer amount decreases the number of ions required to displace at the beginning of each cycle. This cannot be decreased to zero because there needs to be sufficient ionic conductivity and mechanical stability. Additionally, thinner membranes can lead to increased bicarbonate back diffusion offsetting gains in time lag.^{1,15} Increasing the capacity of an electrode lengthens the time for a full cycle and thus a higher percentage of time is spent in a high flux region of a cycle. Increasing the electrode capacity can increase the mass transport resistance for diffusion of CO_2 into the electrode by decreasing the porosity of the electrode or increasing the diffusive path length. The laboratory prepared electrodes nominally have a porosity of 30–40%. Optimization of this porosity can lead to flux and energy cost gains.

Decreasing the amount of time required for regeneration can also decrease the overall energy cost of the system. In Ni-MH batteries the amount of OER that occurs depends on the C-rate used and the dopants present.^{27,32} The C-rate is defined as a ratio of the current density in mA to the capacity of the electrode in mAh. The electrodes in Fig. 2 ran at a C-rate of 0.85. Lower C-rates allow for lower voltage operation of the

battery limiting the catalytic OER which has a high kinetic overpotential. Dopants can either improve the kinetics of the Ni(OH)_2 oxidation or hinder the kinetics of the OER reaction. Dopants such as cobalt have shown the ability to limit OER and can be added to limit need for regeneration.^{33,34} Leaning on nickel hydride battery research to limit regeneration requirement to 10% of a cycle length could decrease the average full cycle energy cost to $0.90 \text{ MWh} \cdot \text{ton}_{\text{CO}_2}^{-1}$ with this laboratory scale system.

The electrodes shown in Fig. 2 were electrochemically precipitated; this method was chosen for tunability of the Ni(OH)_2 loading early in research. There are several other methods to produce Ni(OH)_2 including electrodeposition and bulk precipitation.²⁴ Improvements were made to the laboratory scale electrode to produce large manufacturable electrodes for pilot scale operation. Such 25 cm^2 electrodes were tested for 5000 hours with the cell supplied with ambient air held at 32°C and 82–87% RH. Three segments of the corresponding data are shown in Fig. 3. The end of the 5000-hour run highlighted a low energy cost near $0.46 \text{ MWh} \cdot \text{ton}_{\text{CO}_2}^{-1}$ with a flux of $62 \text{ kg}_{\text{CO}_2} \cdot \text{m}^{-2} \cdot \text{yr}^{-1}$.

The more manufacturable 25 cm^2 cell shows an increase in cycle length or capacity over time (Fig. 3). This is likely the result of a self-limiting corrosion process in the electrode. The nickel foam used in the electrode as a scaffold for Ni(OH)_2 can be oxidized to Ni(OH)_2 at potentials above 0.11 V vs reversible hydrogen electrode (RHE).²³ Any exposed bare nickel can go through the oxidation process. The corrosion stops when accessible nickel metal is fully converted. An increase in capacity is seen between the first and second segments, but not between the second and third segments suggesting the complete conversion of the available nickel sites. This phenomenon was also observed at the laboratory scale and is shown in SI Discussion 4. An increase of the average flux is also seen, contributing to a lower energy cost. This corroborates the concept that reducing the effect of time lag with a larger capacity electrode will lead to higher average flux and lower average energy cost.

This manufacturable electrode was scaled up from 25 cm^2 to 300 cm^2 and used in a $9 \times 300\text{-cm}^2$ stack (Fig. 3d,f). This stack averaged an energy cost of $0.83 \text{ MWh} \cdot \text{ton}_{\text{CO}_2}^{-1}$ with a flux of $75 \text{ kg}_{\text{CO}_2} \cdot \text{m}^{-2} \cdot \text{yr}^{-1}$. At the system level a blower is required to push air through the DAC device and 300 Pa is considered an upper limit for DAC.⁴ The laboratory scale pressure drop suffers from triple serpentine flow fields adapted from bench top fuel cells with a pressure drop of 2800 Pa for a $1 \text{ L} \cdot \text{min}^{-1}$ flow rate as discussed in SI Discussion 3. The scaled system exhibits a total pressure drop of less than 300 Pa (10x lower in comparison to the lab cells) while maintaining similar performance metrics. Such low pressure drops range is obtained by an innovative design of the stack hardware (inlets/outlets and flow field morphology) - needed to allow energy efficient Air Streaming Unit (ASU, e.g., axial fan) and an overall energy-efficient DAC process.

Figure 4 shows a levelized cost reduction pathway to $92 \$ \cdot \text{ton}_{\text{CO}_2}^{-1}$ in line with the United States DOE targets. Durability at the 25 cm^2 size and performance at the 9-cell stack size lays the groundwork for the $1000 \text{ ton}_{\text{CO}_2} \cdot \text{yr}^{-1}$ Gen-0 pilot scale. The Gen-1 system will follow research pathways discussed to decrease time lag and regeneration as well as increasing CO_2 flux. Gen-1 will additionally focus on the full

system requirements including a blower and compressor. Improved airflow design can lower pressure drop to decrease blower costs.^{4,35} Electrochemical compression is to be explored as shown in proton exchange membrane electrolyzers (PEMEL) for hydrogen production with a cost benefit compared with conventional compression alone.^{36–38} Module standardization for assembly line production and expected improvement in energy consumption are the main drivers for the Gen-2 system.³⁹ Further improvement of the CO₂ flux and economies of scale will enable Gen-3 First-Of-A-Kind (FOAK) megaton systems.^{40–42} At this scale pricing improvements can be achieved for membrane polymers and peripheral components.⁴³ Finally, centralized control rooms, automation, digital twin machine learning, and AI-driven maintenance strategies are expected to drive final costs towards the Nth-Of-A-Kind (NOAK) Gen-3 systems.

Conclusion

The nickel hydroxide HEMCC technology shown here has a low energy cost compared to conventional temperature swing adsorption devices. Producing the pH gradient is the primary energy cost for the device, followed by the polarization of the electrodes, and resistance.

At the laboratory scale the concept was developed to produce a device that has an average energy cost of 1.15 MWh·ton_{CO2}⁻¹ at an average flux of 78 kg_{CO2}·m⁻²·yr⁻¹. The energy cost and flux can be improved by decreasing the time lag for pH gradient formation and minimizing parasitic OER reactions. The time lag can be decreased by increasing electrode capacity which aids in increasing the average flux while decreasing the average energy cost. Anodic OER reactions can be limited by employing traditional Ni-MH battery dopants. By limiting OER, the energy cost can be reduced by reducing the required amount of regeneration.

Scale up of the nickel hydroxide HEMCC has started where the original concept was expanded from a 25 cm² cell to a 9-cell stack of 300 cm² cells. A pilot module containing these stacks is in operation capturing CO₂ from ambient air. A 5000-hour durability test has shown stability of the electrode design with an end-of-test energy cost of 0.46 MWh·ton_{CO2}⁻¹ at a flux of 62 kg_{CO2}·m⁻²·yr⁻¹. This electrode design was shown to be able to scale into a stack with a low pressure drop of 300 Pa meeting technical requirements for DAC.

The prototype is being iterated to drive down cost along learning curves like those seen in solar photo voltaic cells and lithium-ion batteries. Device level research is expected to achieve many of the early-stage improvements, while manufacturability and economies of scale will drive costs down below the 100 \$·ton_{CO2} target set forth for DAC.

Methods

Electrode preparation

Lab scale electrode preparation was done by electrochemical precipitation of nickel hydroxide onto nickel foam. The nickel foam was from MTI corporation (product name EQ-bcnf-16m) with 99.9% purity, 95% porosity, 80–110 pores per inch and thickness of 1.6mm. Three 25cm² nickel foam substrates were cut retaining a 1 cm² tab for connecting with an alligator clip. The three substrates were compressed in a hydraulic press to 512 μ m by using a 512 μ m thick steel plate as a guide barrier. These were weighed and then placed in prepared 1 M sulfuric acid solution for 1 hour to remove the surface oxide layer. The oxide layer removed is assumed to be negligible in mass for purposes of measuring amount of mass added to the working electrode.

These substrates were set parallel in a 0.05 M nickel chloride solution using 98% anhydrous nickel chloride salt from Millipore Sigma (CAS No: 7718-54-9). NiCl₂ hexahydrate (Sigma-Aldrich, ReagentPlus) was also tested without any noticeable performance difference. The substrates were suspended in place with copper electrode clips as shown in **SI Fig. 9**. The outer electrodes were connected via wire to be at the same voltage as the counter electrode, the middle electrode was the working electrode for Ni(OH)₂ to be precipitated onto. Two outer electrodes were used to evenly grow Ni(OH)₂ on both sides of the central working electrode. The distance between electrodes were approximately controlled to 1 cm.

A reference Ag/AgCl electrode from Pine Research (RREF0021) tracked voltage in the cell during electrochemical precipitation. A current density of -5 mA·cm² for 4 hours was applied to the working electrode. Afterwards, the electrodes were rinsed with DI water and set out to dry overnight. The next day the weight of the working electrode was measured to determine the mass of deposition. As a control, the counter electrodes were also weighed to measure their loss of Ni. The loss of Ni of the counter electrodes closely aligns with the oxidation to Ni²⁺ in the solution as expected this balances the loss of Ni²⁺ during precipitation and maintains a relatively stable bulk Ni²⁺ concentration. The pair of counter electrodes lost 0.57 g Ni (27% of initial mass) during the process while the working electrode gained 0.83 g during the process.

After electrochemical precipitation the electrodes were spray coated with Piperion® ionomer. A solution of 1.3333g of 5% ionomer in ethanol solution was added to 6.2222g IPA and sonicated for 30 min. This solution was sprayed on one side of the electrode and then set out to dry for 1 day. Weighing the next days showed that 25% of the initial ionomer mass was transferred to the electrode during the spraying.

Ex-situ three electrode cycling

The electrode with ionomer was then placed in a 1 M KOH bath in the same set up as the corrosion test in **SI Fig. 7,8**. Three series of current densities were used to cycle each electrode, 1-cycle at 2 mA·cm², 30-cycles at 5 mA·cm², and 5-cycles at 2 mA·cm². A break in is conducted at 2 mA·cm⁻². First the electrode is charged from Ni(OH)₂ to NiOOH for three hours finishing with a plateau at OER voltage. Then the electrode is discharged back to Ni(OH)₂ to completion with a cutoff voltage of 1.1 V vs RHE. This break in

charge is longer than the final 5-cycles at $2 \text{ mA}\cdot\text{cm}^{-2}$ to allow for impurities in the deposited $\text{Ni}(\text{OH})_2$ structure to escape without physically damaging the deposited layer.

Next, 30 cycles at $5 \text{ mA}\cdot\text{cm}^2$ are conducted to stabilize the $\text{Ni}(\text{OH})_2$ layer. It was found that 30 cycles were required to reach a plateau in capacity for an electrode; the current density $5 \text{ mA}\cdot\text{cm}^2$ was chosen to speed up the process. The final 5 cycles were conducted at $2 \text{ mA}\cdot\text{cm}^2$ to compare with the $2 \text{ mA}\cdot\text{cm}^2$ used during the direct air capture tests.

Membrane Electrode Assembly

The final assembly of the laboratory scale 25cm^2 cell uses a Scribner 25 cm^2 cell with triple serpentine flow fields. (see **SI Fig. 11**) A 2.5"x3" piece of $80\mu\text{m}$ Piperion® membrane was used between two electrodes. Two electrochemically precipitated nickel hydroxide electrodes were cut to fit precisely to their respective gaskets. A total of $336\mu\text{m}$ in FEP gaskets were used on each side for the $512\mu\text{m}$ thick electrodes. This leads to a 0.39 compression ratio once the cell bolts are torqued to 100 PSI.

Declarations

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Contributions

Y.Y. and P.B.S. supervised the project. P.B.S. provided the initial concept and guidance on fundamentals of electrochemical pH driven CO_2 capture. R.J.B. designed the test station and laboratory scale electrodes. R.J.B. and T.W. developed testing protocol and evaluation of the direct air capture system. R.B.G. conducted electrochemical tests and related data processing. T.W. carried out materials characterization. B.A. led the development and scale up efforts of the main technological aspects including durable

electrodes, stack hardware design and the accommodating system for operating the DAC process. Y.N.G led the MEA optimization efforts and M.A conducted the DAC testing J-P.H. developed the TEA analysis. R.J.B. wrote the manuscript, with input from all authors.

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Competing Interests

Y.Y. is the Founder and CEO and P.B.S. is a Co-Founder for Versogen, the producer of Piperion®. Y.Y. and B.A. are Co-Founders for RepAir the scale-up company.

References

1. Shi, L. *et al.* A shorted membrane electrochemical cell powered by hydrogen to remove CO₂ from the air feed of hydroxide exchange membrane fuel cells. *Nature Energy* 7, 238–247 (2022). <https://doi.org/10.1038/s41560-021-00969-5>
2. Bell, W. L. Synthesis and Evaluation of Electroactive CO₂ Carriers. *SAE International* 97, 544–552 (1988).
3. Rheinhardt, J. H., Singh, P., Tarakeshwar, P. & Buttry, D. A. Electrochemical Capture and Release of Carbon Dioxide. *ACS Energy Letters* 2, 454–461 (2017). <https://doi.org/10.1021/acsenergylett.6b00608>
4. Luukkonen, A., Elfving, J. & Inkeri, E. Improving adsorption-based direct air capture performance through operating parameter optimization. *Chemical Engineering Journal* 471 (2023). <https://doi.org/10.1016/j.cej.2023.144525>
5. McDonald, A. & Schrattenholzer, L. Learning rates for energy technologies. *Energy Policy* 29, 255–261 (2001). [https://doi.org/10.1016/s0301-4215\(00\)00122-1](https://doi.org/10.1016/s0301-4215(00)00122-1)
6. IEA. Net Zero by 2050. (Paris, 2021).
7. Fuhrman, J. *et al.* Diverse carbon dioxide removal approaches could reduce impacts on the energy–water–land system. *Nature Climate Change* 13, 341–350 (2023). <https://doi.org/10.1038/s41558-023-01604-9>
8. UNEP (2017). The Emissions Gap Report 2017. United Nations Environment Programme (UNEP), Nairobi. (2017).
9. IEA (2022), Direct Air Capture 2022, IEA, Paris <https://www.iea.org/reports/direct-air-capture-2022>, Licence: CC BY 4.0. (2022).
10. Keith, D. W., Holmes, G., St. Angelo, D. & Heide, K. A Process for Capturing CO₂ from the Atmosphere. *Joule* 2, 1573–1594 (2018). <https://doi.org/10.1016/j.joule.2018.05.006>
11. Fasihi, M., Efimova, O. & Breyer, C. Techno-economic assessment of CO₂ direct air capture plants. *Journal of Cleaner Production* 224, 957–980 (2019). <https://doi.org/10.1016/j.jclepro.2019.03.086>

12. Monthly Energy Review August 2024 (U. S. Energy Information Administration, 2024).
13. Carbon Negative Shot, <<https://www.energy.gov/fecm/carbon-negative-shot>> (2022).
14. Matz, S. et al. Hydrogen-powered Electrochemically-driven CO₂ Removal from Air Containing 400 to 5000 ppm CO₂. *Journal of The Electrochemical Society* 169 (2022). <https://doi.org/10.1149/1945-7111/ac7adf>
15. Matz, S. et al. Demonstration of Electrochemically-Driven CO₂ Separation Using Hydroxide Exchange Membranes. *Journal of The Electrochemical Society* 168 (2021). <https://doi.org/10.1149/1945-7111/abd5fe>
16. Zhu, P. et al. Continuous carbon capture in an electrochemical solid-electrolyte reactor. *Nature* 618, 959–966 (2023). <https://doi.org/10.1038/s41586-023-06060-1>
17. Gao, X., Omosebi, A., Perrone, R. & Liu, K. Promoting CO₂ Release from Carbonate Containing Solvents during Water Electrolysis for Direct Air Capture. *Journal of The Electrochemical Society* 169 (2022). <https://doi.org/10.1149/1945-7111/ac5fef>
18. Gao, X., Omosebi, A., Patrick, A. & Liu, K. Optimizing Direct Air Capture Solvents to Minimize Energy Consumption of CO₂ Release in a Carbonate Electrolyzer. *ECS Advances* 3 (2024). <https://doi.org/10.1149/2754-2734/ad3928>
19. Zheng, Y. et al. Editors' Choice—Power-Generating Electrochemical CO₂ Scrubbing from Air Enabling Practical AEMFC Application. *Journal of The Electrochemical Society* 168 (2021). <https://doi.org/10.1149/1945-7111/abe08a>
20. Wang, X. Comprehensive Study of the Hydration and Dehydration Reactions of Carbon Dioxide in Aqueous Solution. *J. Phys. Chem. A* 114, 1734–1740 (2010).
21. Diederichsen, K. M., Liu, Y., Ozbek, N., Seo, H. & Hatton, T. A. Toward solvent-free continuous-flow electrochemically mediated carbon capture with high-concentration liquid quinone chemistry. *Joule* 6, 221–239 (2022). <https://doi.org/10.1016/j.joule.2021.12.001>
22. Li, X. et al. Redox-tunable isoindigos for electrochemically mediated carbon capture. *Nat Commun* 15, 1175 (2024). <https://doi.org/10.1038/s41467-024-45410-z>
23. Huggins, R. A. *Advanced Batteries Materials Science Aspects*. (Springer, 2009).
24. Hall, D. S., Lockwood, D. J., Bock, C. & MacDougall, B. R. Nickel hydroxides and related materials: a review of their structures, synthesis and properties. *Proc Math Phys Eng Sci* 471, 20140792 (2015). <https://doi.org/10.1098/rspa.2014.0792>
25. Ash, B., Nalajala, V. S., Popuri, A. K., Subbaiah, T. & Minakshi, M. Perspectives on Nickel Hydroxide Electrodes Suitable for Rechargeable Batteries: Electrolytic vs. Chemical Synthesis Routes. *Nanomaterials (Basel)* 10 (2020). <https://doi.org/10.3390/nano10091878>
26. McQueen, N. et al. A review of direct air capture (DAC): scaling up commercial technologies and innovating for the future. *Progress in Energy* 3 (2021). <https://doi.org/10.1088/2516-1083/abf1ce>
27. Becker, M. D. C., D. J.; Salva, H.R.; Rodriguez Nieto, F.; Ghilarducci, A. A.; Visintin A. Oxygen Evolution Nickel Hydroxide Electrodes in a Ni-MH Battery Prototype. *Journal of New Materials for Electrochemical Systems* 12, 277–282 (2012).

28. Hall, D. S., Lockwood, D. J., Poirier, S., Bock, C. & MacDougall, B. R. Raman and infrared spectroscopy of alpha and beta phases of thin nickel hydroxide films electrochemically formed on nickel. *J Phys Chem A* 116, 6771–6784 (2012). <https://doi.org/10.1021/jp303546r>
29. Wang, J. *et al.* Poly(aryl piperidinium) membranes and ionomers for hydroxide exchange membrane fuel cells. *Nature Energy* 4, 392–398 (2019). <https://doi.org/10.1038/s41560-019-0372-8>
30. Tobias, J. S. N. C. W. Theoretical Analysis of Current Distribution in Porous Electrodes. *Journal of The Electrochemical Society* (1962).
31. Newman, J. & Tiedemann, W. Porous-electrode theory with battery applications. *AIChE Journal* 21, 25–41 (1973). <https://doi.org/10.1002/aic.690210103>
32. Ayeb, A. & Notten, P. H. L. The oxygen evolution kinetics in sealed rechargeable NiMH batteries. *Electrochimica Acta* 53, 5836–5847 (2008). <https://doi.org/10.1016/j.electacta.2008.03.023>
33. Shinyama, K., Nakamura, H., Nohma, T. & Yonezu, I. Improvement of high- and low-temperature characteristics of nickel-metal hydride secondary batteries using rare-earth compounds. *Journal of Alloys and Compounds* 408–412, 288–293 (2006). <https://doi.org/10.1016/j.jallcom.2005.04.111>
34. Watanabe, K.-i., Koseki, M. & Kumagai, N. Effect of cobalt addition to nickel hydroxide as a positive material for rechargeable alkaline batteries. *Journal of Power Sources* 58, 23–28 (1996). [https://doi.org/10.1016/0378-7753\(95\)02272-4](https://doi.org/10.1016/0378-7753(95)02272-4)
35. Bui, M. *et al.* Carbon capture and storage (CCS): the way forward. *Energy & Environmental Science* 11, 1062–1176 (2018). <https://doi.org/10.1039/c7ee02342a>
36. Hancke, R., Holm, T. & Ulleberg, Ø. The case for high-pressure PEM water electrolysis. *Energy Conversion and Management* 261 (2022). <https://doi.org/10.1016/j.enconman.2022.115642>
37. Hancke, R., Bujlo, P., Holm, T. & Ulleberg, Ø. High-pressure PEM water electrolyser performance up to 180 bar differential pressure. *Journal of Power Sources* 601 (2024). <https://doi.org/10.1016/j.jpowsour.2024.234271>
38. Abbasi, R. *et al.* A Roadmap to Low-Cost Hydrogen with Hydroxide Exchange Membrane Electrolyzers. *Adv Mater* 31, e1805876 (2019). <https://doi.org/10.1002/adma.201805876>
39. Rubin, E. S., Azevedo, I. M. L., Jaramillo, P. & Yeh, S. A review of learning rates for electricity supply technologies. *Energy Policy* 86, 198–218 (2015). <https://doi.org/10.1016/j.enpol.2015.06.011>
40. Gerke, B. F. N., Allison T.; Fisseha, Kibret S.. Recent price trends and learning curves for household LED lamps from a regression analysis of internet retail data. (Lawrence Berkeley National Laboratory, 2015).
41. Schmidt, O., Hawkes, A., Gambhir, A. & Staffell, I. The future cost of electrical energy storage based on experience rates. *Nature Energy* 2 (2017). <https://doi.org/10.1038/nenergy.2017.110>
42. Orangi, S. *et al.* Historical and prospective lithium-ion battery cost trajectories from a bottom-up production modeling perspective. *Journal of Energy Storage* 76 (2024). <https://doi.org/10.1016/j.est.2023.109800>
43. Mathias, M. F. *et al.* Two Fuel Cell Cars In Every Garage? *The Electrochemical Society Interface* 14, 24–35 (2005). <https://doi.org/10.1149/2.F05053if>

Figures

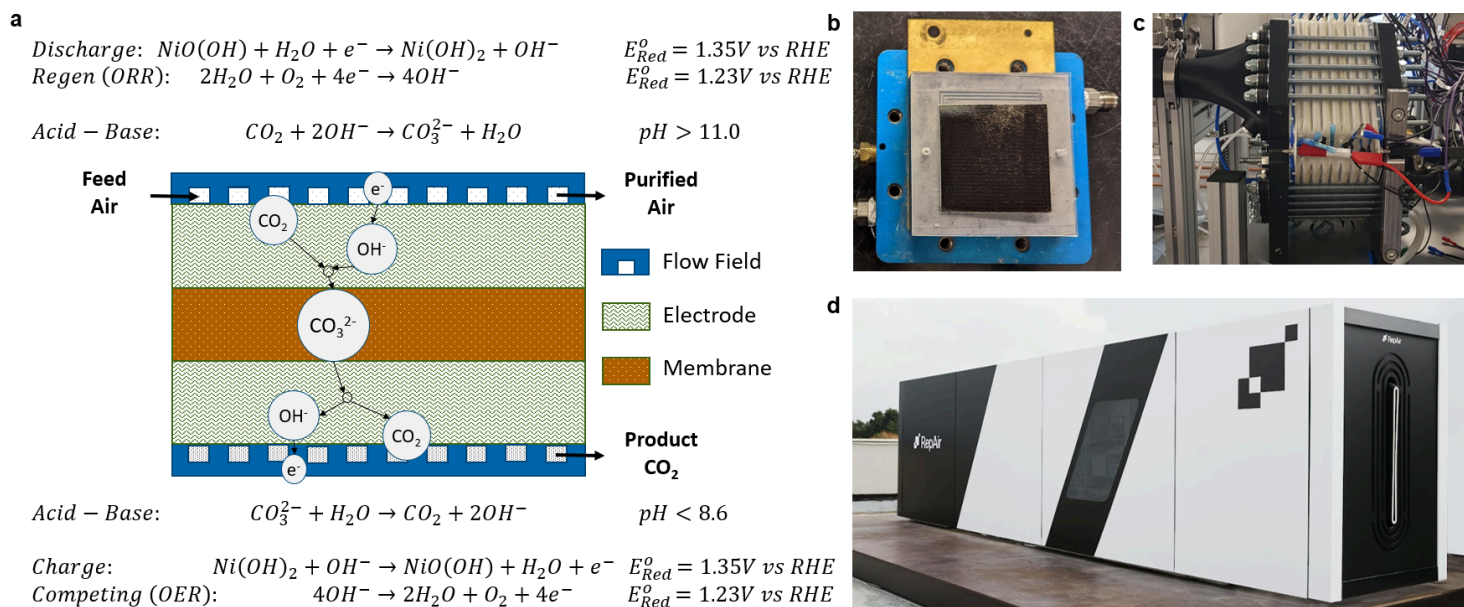


Figure 1

HEMCC device schematic and scales from $200 \text{ g}_{\text{CO}_2} \cdot \text{yr}^{-1}$ to $200 \text{ ton}_{\text{CO}_2} \cdot \text{yr}^{-1}$

a. A laboratory scale MEA schematic shown with two porous Ni(OH)_2 electrodes separated by a hydroxide exchange membrane. Flow fields provide a path for air and a purified CO_2 outlet stream. During cycling the cathode produces OH^- and the anode consumes OH^- generating a pH gradient. A cycle stops when the cathode NiOOH is consumed. During the cycling step a competing oxygen evolution reaction (OER) occurs at the anode which prevents fully charging the anode. A regeneration step is performed after the cathode NiOOH is fully consumed, where the cathode performs oxygen reduction reaction (ORR) to fully charge the anode. The capture and release of CO_2 is based on an acid-base equilibrium driven by the pH gradient.

b. A laboratory scale 25 cm^2 membrane electrode assembly (MEA).

c. A stack of $9\text{-}300 \text{ cm}^2$ cells in pilot operation.

d. Commercial RepAir® module under construction.

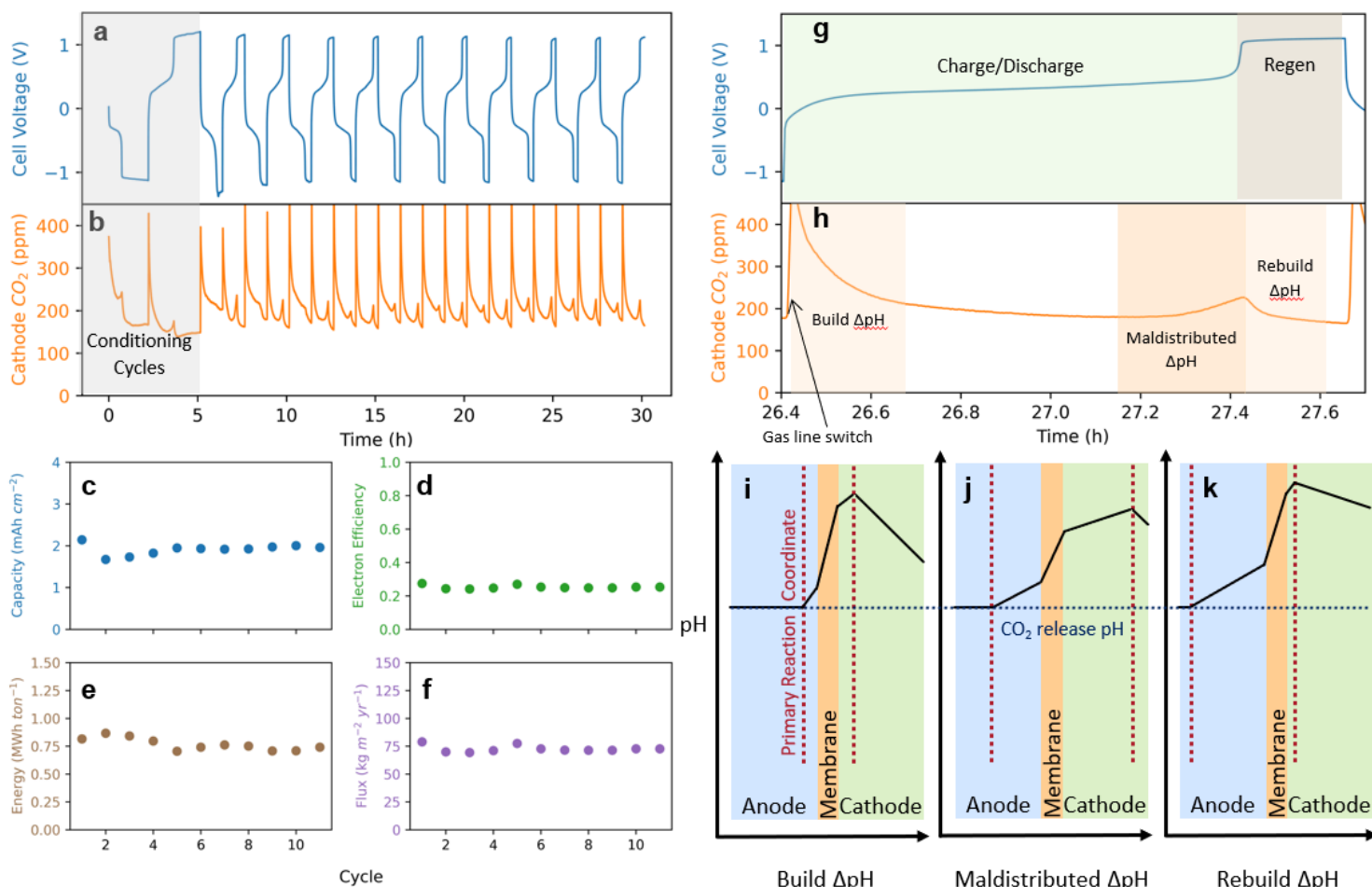


Figure 2

Laboratory scale 25 cm² HEMCC and its behavior. The HEMCC is comprised of two identical electrochemically precipitated Ni(OH)₂ electrodes and an 80μm Piperion® membrane. The cathode air flow was 1 L•min⁻¹ with 400 ppm CO₂ and 90% RH. The anode received a sweep gas of 90% RH N₂ at 1 L•min⁻¹. **a.** The cell voltage (V) and **b.** the cathode outlet CO₂ concentration (ppm) are used to calculate the key process variables **c-f** during the charge/discharge step: **c.** capacity, **d.** electron efficiency, **e.** energy, and **f.** flux, all excluding the regeneration step. Regeneration steps are excluded to highlight the low energy cost of the charge-discharge step. Capture still occurs during regeneration but at higher energy cost. The first cycle on each side is a 1.5-hour regeneration hold for break-in followed by 10-75 minute cycles on each electrode. The cycle length is chosen based on an ex-situ capacity test to minimize regeneration (**SI Discussion 7**). **c.** Capacity achieved an average of 1.89 mAh•cm⁻² which was 85% of the ex-situ capacity tests. **d.** Electron efficiency achieved 0.25. **e.** Energy cost averaged 0.76 MWh•ton_{CO₂}⁻¹ while **f.** flux averaged to 72 kg_{CO₂}•m²•yr⁻¹. **g.** and **h.** show a single cycle to highlight the change in voltage and capture in the charge/discharge and regeneration steps. **i., j., and k.** are graphic aids to describe the build ΔpH, maldistributed ΔpH, and rebuild ΔpH sections in **g.** and **h.** The ΔpH between the peak pH at the cathode and the CO₂ release pH at the anode drives the CO₂ capture rate in the cell, see **SI Discussion 2**. The CO₂ release pH is the pH that achieves equilibrium between CO₂ in the anode and the concentrated CO₂ gas

product stream. The dotted red primary reaction coordinate lines are rough position where OH^- is produced in the cathode and consumed in the anode. **i.** The build ΔpH section is directly after the gas lines switch in the cell. Redox reactions take place close to the membrane with the cathode near 100% state of charge and the anode near 0% state of charge. The pH at the back of the cathode electrode is close to the pH of the anode in the previous cycle. **j.** The maldistributed ΔpH section is at the end of a charge/discharge step. This occurs because of the uneven mass loading of $\text{Ni}(\text{OH})_2/\text{NiOOH}$ when making the electrode and uneven distribution of resistance through the electrode. Near the end of the charge/discharge step, low loading and low resistance volumes of the cathode will have fully been consumed, no longer producing OH^- . Surrounding higher loading and higher resistance volumes continue producing OH^- but the average pH across the electrode drops lowering the average driving force for capturing CO_2 . The anode in this section is still charging and has not reached 100% state of charge because of competing OER. **k.** The rebuild ΔpH section is the beginning of the regeneration after the cathode has reached 0% state of charge. The cathode reaction coordinate is brought to the front towards the membrane where catalytic ORR reactions are predominant. The full area of the electrode is used again, and the average pH increases to have a higher rate of CO_2 capture. The anode continues to charge towards 100% for the next cycle.

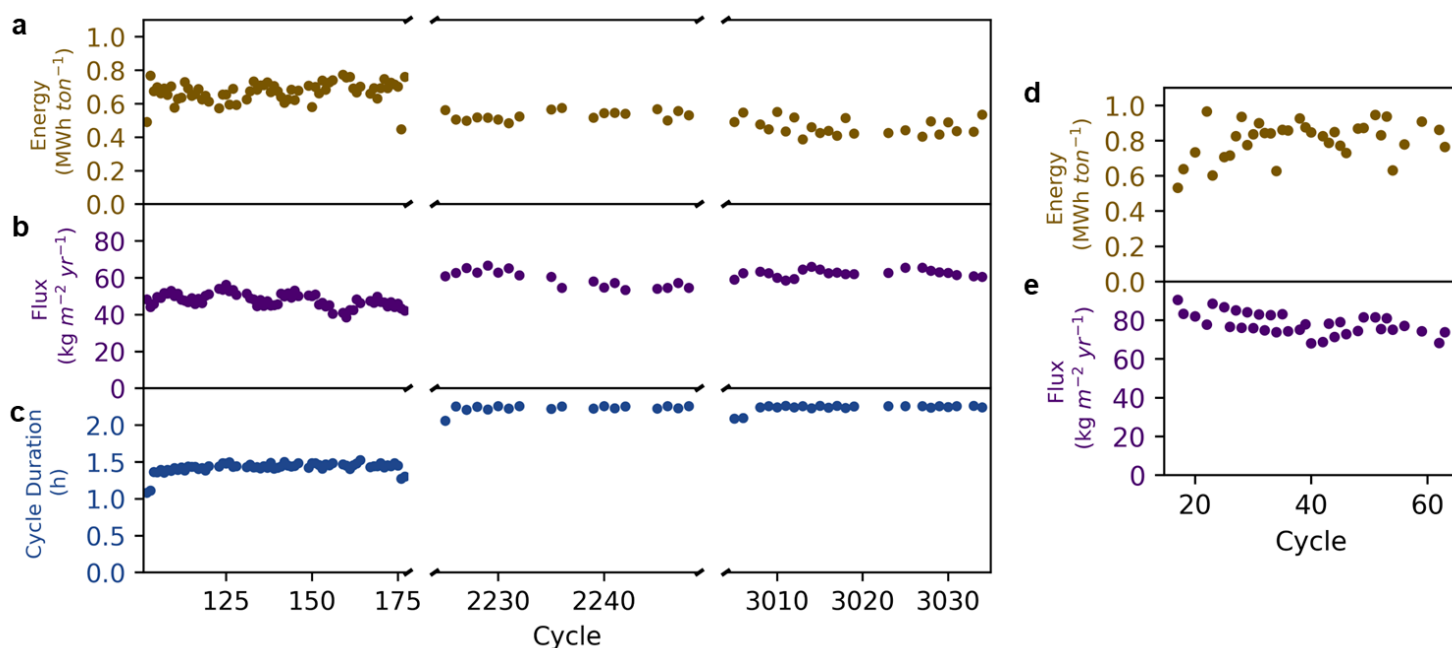


Figure 3

Durability on manufacturable 25cm² electrodes and performance on pilot stack. **a-c.** Three segments of a >5000 hour durability test using a manufacturable 25cm² cell, operated using ambient air held at 32 °C and 82-87 % RH. Missing cycles in segment 2 and 3 contained software errors and were omitted. **a.** Energy cost across three segments averaged 0.67, 0.53, 0.46 MWh·ton_{CO₂}⁻¹ for the respective segments. **b.** Flux averaged 48, 60, 62 kg_{CO₂}·m⁻²·yr⁻¹ for the respective segments. **c.** Cycle duration averaged 1.4, 2.3, 2.4 hours for the respective segments. Increased capacity is associated with corrosion process between the first and second segments. Flux and energy cost improved over time as the capacity increased. **d and e.**

Performance data of a 9 x 300-cm² cell stack operated in ambient conditions between 20-25 °C and 75-90% RH with **d.** average energy cost of 0.83 MWh·ton_{CO2}⁻¹ and **e.** average flux of 75 kg_{CO2}·m⁻²·yr⁻¹.

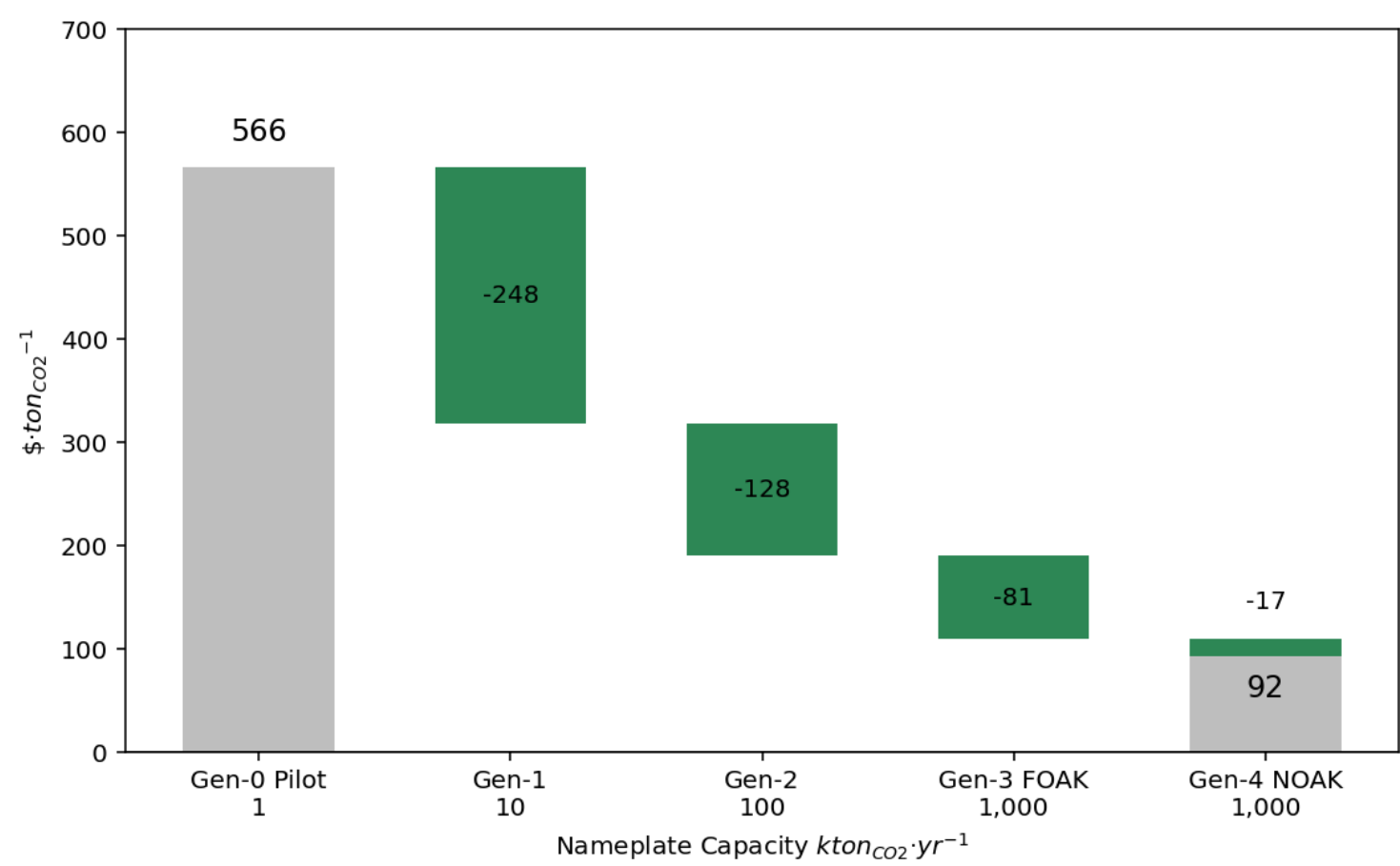


Figure 4

Levelized cost projection for Ni(OH)₂ based HEMCC The Gen-0 pilot scale is in production with a calculated cost of 566 \$·ton_{CO2}⁻¹. To get from Gen-0 to Gen-1, research developments in the HEMCC device are expected to follow learning curves like solar and battery industries to reduce cost. Advancements from Gen-1 to Gen-2 are expected to mostly derive from improvements in manufacturability and energy consumption. From Gen-2 to Gen-3 First-Of-A-Kind (FOAK) economies of scale will provide the greatest benefit. To achieve Gen-3 Nth-Of-A-Kind systems, centralized control rooms, digital twins, and AI-assisted maintenance are expected to drive the final costs.

Supplementary Files

This is a list of supplementary files associated with this preprint. Click to download.

- [SIHydroxideExchangeMembraneCarbnCaptureHEMCCusingsymmetricalnickelhydroxidebatteries.docx](#)