

Supporting information

Feasibility study of an electrochemical hydrogen looping system for indirect ocean capture

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Supporting note 1:

Figure S1 shows the Bjerrum plot for the carbon concentrations in seawater. This graph shows the pH dependency of the carbonate species and allowed us to establish the degassing method used before measuring the samples with TOC analysis. Based on pH measurements, the theoretical capture efficiency could be calculated, which was utilized to confirm the TOC measurements. Using this strategy a degassing method was established in which a 225 mL sample was collected and sparged with argon gas for 12 minutes, before stating the TOC analysis.

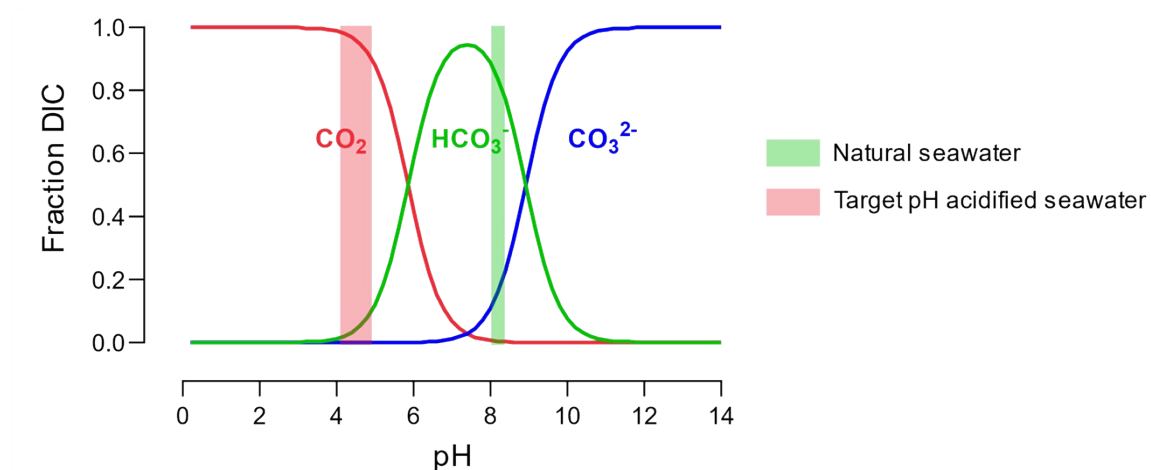


Figure S1. Bjerrum plot of the fraction of carbonate, bicarbonate and carbon dioxide as a function of the pH in synthetic seawater

Table S1. Overview and comparison of the energy consumption of all indirect ocean capture technologies with the most common DAC technologies.

Technology	Energy consumption [kJ/mol CO ₂]	Theoretical cell voltage [V]	Ref.
EHL	77.5	0.48	This work
E-CEM	3170	1.73	[1-3]
BPMED-1	220	0.83	[4]
BPMED-2	145	0.36	[5]
DAC (Liquid scrubbing)	33 – 67	n/a	[6-13]
DAC (Solid sorbent)	18 – 62	n/a	[6, 14-20]

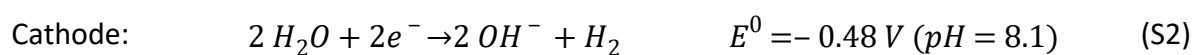
Supporting note 2:

It needs to be mentioned that for DAC technologies extra heat energy is required to complete the capture. For liquid scrubbing and solid sorbent DAC this respectively accounts for 230 – 2124 kJ/mol CO₂ and 176 – 519 kJ/mol CO₂.

Supporting note 3:

The theoretical cell potential of all indirect ocean capture technologies were calculated below:

(1) EHL



$$E_{anode} - E_{cathode} = 0 V - (-0.48 V) = 0.48 V \quad (S3)$$

(2) BPMED-1

The theoretical cell potential of BPMED is calculated by the pH difference between the acidified and basified seawater, using the Nernstian voltage (0.059·ΔpH). [4]

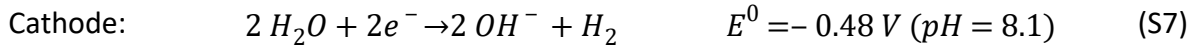
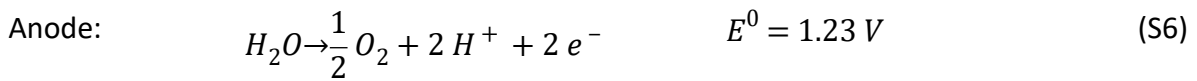
$$0.059 \cdot (14 - 0) = 0.83 \text{ V} \quad (\text{S4})$$

(3) BPMED-2

The calculation of the theoretical potential for the improved BPMED is identical to the one for BPMED-1. [21]

$$0.059 \cdot (10 - 4.5) = 0.3245 \text{ V} \quad (\text{S5})$$

(4) E-CEM [1]



$$E_{anode} - E_{cathode} = 1.23 \text{ V} - (-0.48 \text{ V}) = 1.71 \text{ V} \quad (\text{S8})$$

Supporting note 4:

The electrochemical energy consumption with the most efficient settings was calculated below:

$$EEC = \frac{I \cdot U \cdot t}{Q \cdot t \cdot C \cdot \eta} \quad (\text{S9})$$

$$EEC = \frac{0.032 \text{ A} \cdot 0.765 \text{ V} \cdot 1 \text{ h}}{15 \frac{\text{mL}}{\text{min}} \cdot 0.06 \frac{\text{min} \cdot \text{L}}{\text{h} \cdot \text{mL}} \cdot 1 \text{ h} \cdot 0.002538 \frac{\text{mol}}{\text{L}} \cdot 52.20\%} = 77.5 \frac{\text{kJ}}{\text{mol CO}_2} \quad (\text{S10})$$

Supporting note 5:

The capture efficiency used in the calculation in supporting note 4 was calculated below:

$$\eta = \frac{C_{\text{Carbon before acidification}} - C_{\text{Carbon after acidification}}}{C_{\text{Carbon before acidification}}} \quad (\text{S11})$$

$$\eta = \frac{0.002538 - 0.001213}{0.002538} = 52.20\% \quad (S12)$$

Supporting note 6:

Extra SEM EDS images from the same NaEM membrane as Figure 7 but taken from the side of the membrane facing the middle compartment. The same membrane scaling is established in figure S2a, really showcasing the transport of hydroxyl ions in the opposite direction, even though it is a cation exchange membrane. This property of anion transport allows for the membranes to scale not only from the side facing the cathode, but also from the opposite side and probably the core of the membrane as well. Figure S2b shows again the NaEM membrane facing the middle compartment during a four hour experiment in which the optimal washing step (0.5 M HCl for 8 minutes at a washing flow rate of 45 mL min⁻¹) was executed. It is clear for the electron image and the elemental map that the salt precipitation at the surface layer of the membrane has been washed.

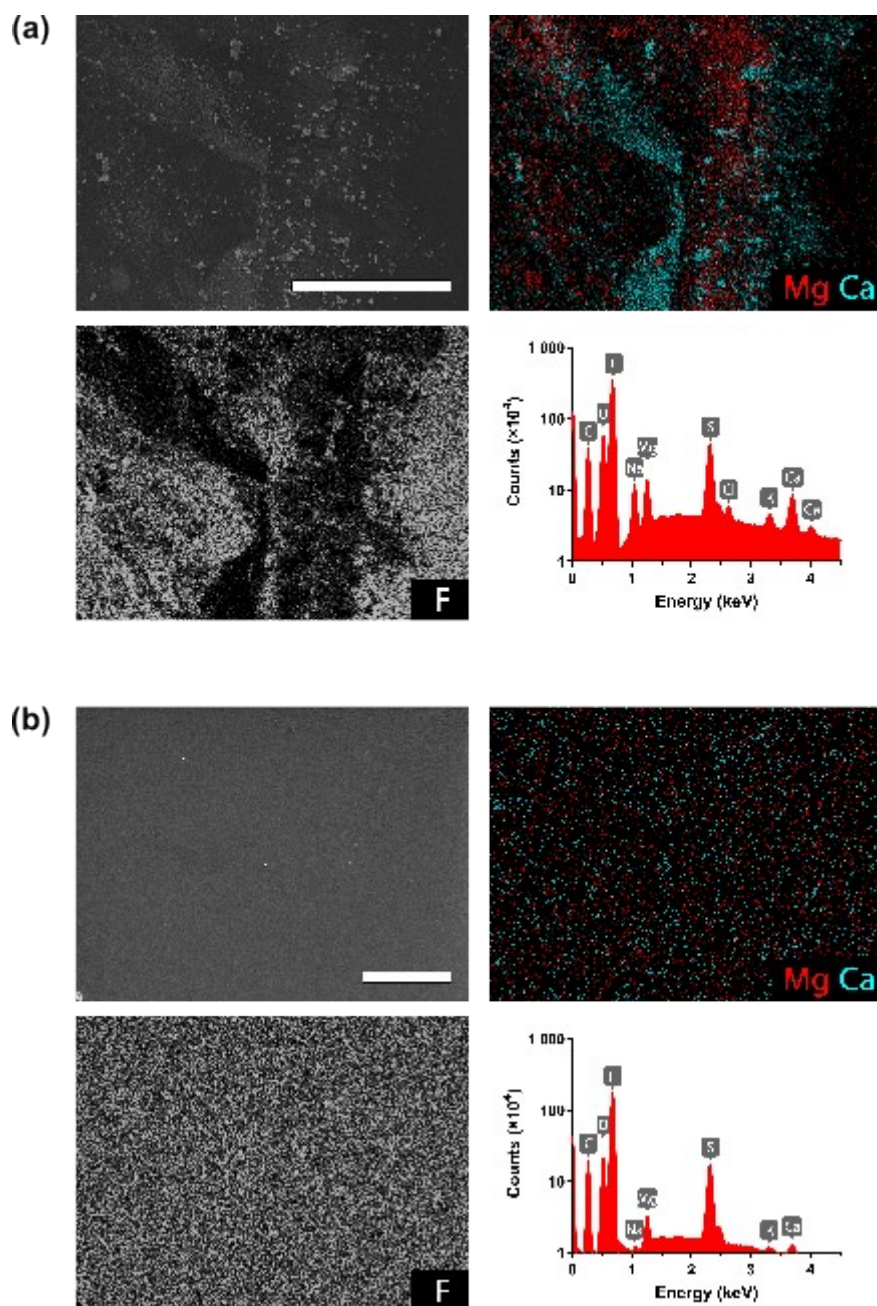


Figure S2. Secondary electron images, elemental maps and EDS spectra of (a) a Nafion™ 117 membrane imaged from the side of the middle compartment after 4 hours of operation without washing steps and (b) a Nafion™ 117 membrane imaged from the side of the middle compartment after four hours of operation including wash steps under the optimal conditions. All scale bars correspond to 250 μm . Experiments with these membranes were carried out at 5 mA cm^{-2} and with a flow rate of 25 mL min^{-1} .

Supporting note 7:

Figure S3 depicts the visual establishing of the salt precipitation issues after four hours of operation at a current density of 5 mA cm^{-2} and a RSW flow rate of 25 mL min^{-1} without any mitigation strategy. Figure S3a shows the salt precipitation in the flow field on which the catalyst substrate rests. Near clogging of a number of flow channels can be visually established. Figure S3b and c respectively show the salt precipitation on the membrane and

on the cathode Pt gauze. The latter is clearly covered in salts which led to the decrease of active surface area, leading to an increase in cell voltage. Identically the blocking of transport pathways through the membrane because of the presence of salt precipitates caused an increased ionic resistance of the membrane, impacting the cell voltage in a negative manner.

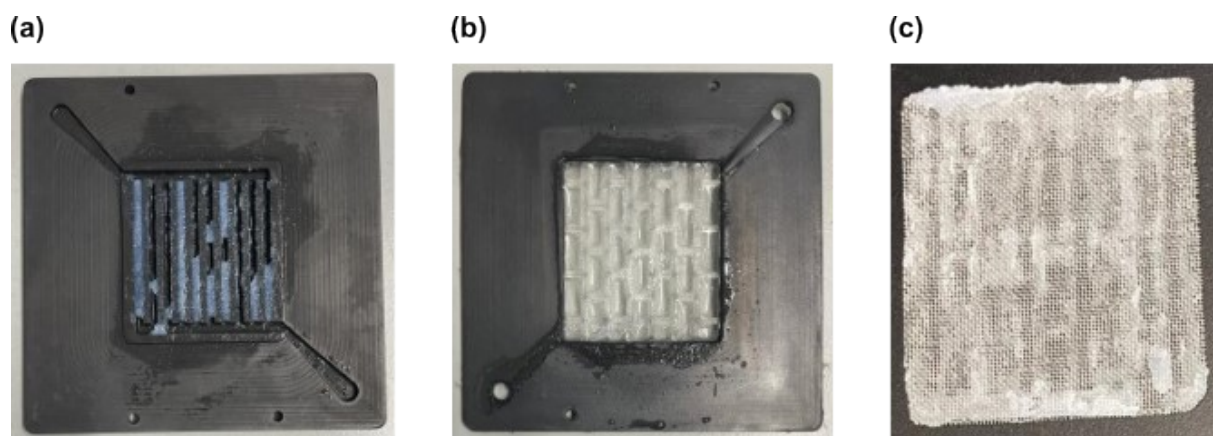


Figure S3. Salt precipitation after four hours of operation at a current density of 5 mA cm^{-2} and a RSW flow rate of 25 mL min^{-1} in (a) the flow channels of the cathodic bipolar plate (b) on the NaEM and (c) on the cathode (Pt gauze)

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