Supporting Information for:

Electrolytic cement clinker production sustained through orthogonalization of ion vectors

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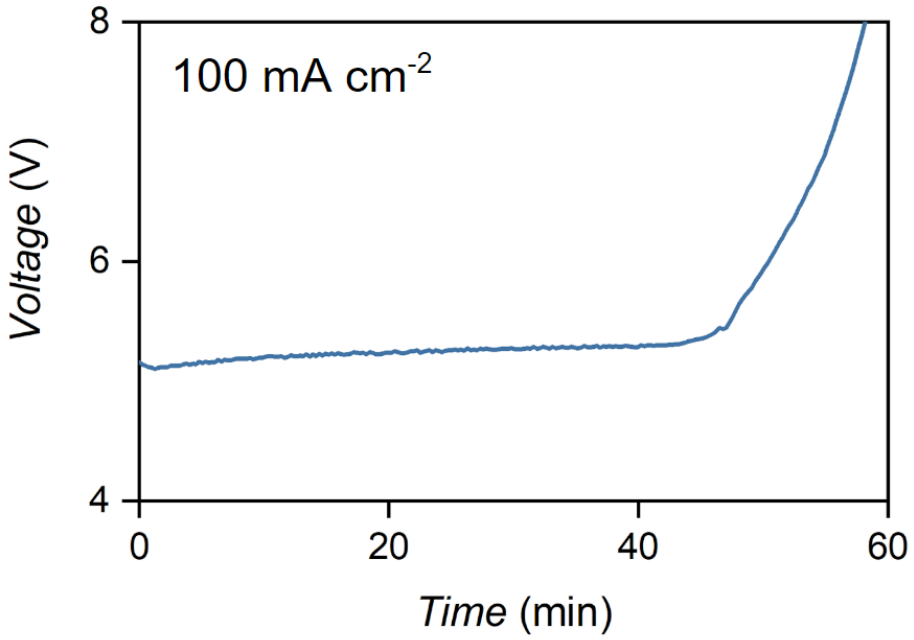

Figure S1. voltage-time characteristics of the cement electrolyser operating at 100 mA cm^{-2}. The cell voltage (V_{cell}) notably increases during electrolysis. The magnitude of the voltage increase varies between experiments, a representative voltage-time curve is shown here.

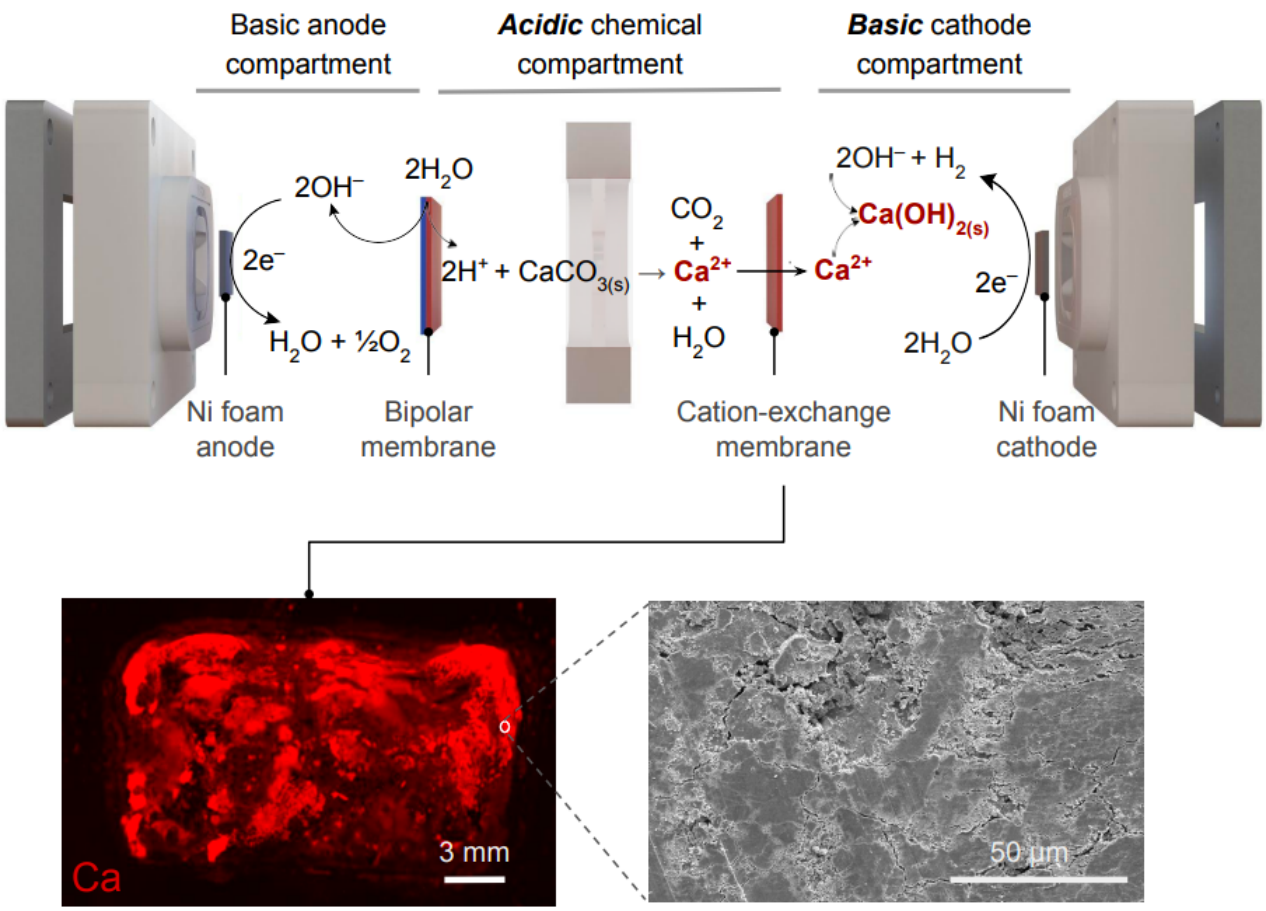

Figure S2. $Ca(OH)_{2(s)}$ deposition on the cation-exchange membrane during electrolysis. Expanded view of the flow electrolyser reported in our previous work which consists of anode, chemical, and cathode compartments. A BPM separates the anode and chemical compartments, and a CEM (Nafion 117) separates the chemical and cathode compartments. Nickel foam was used as both the anode and cathode. Bottom line: XRF and SEM images of a Nafion membrane before and after electrolysis at 100 mA cm^{-2}. Clusters of precipitated $Ca(OH)_{2(s)}$ are observed on the surface of Nafion after electrolysis.

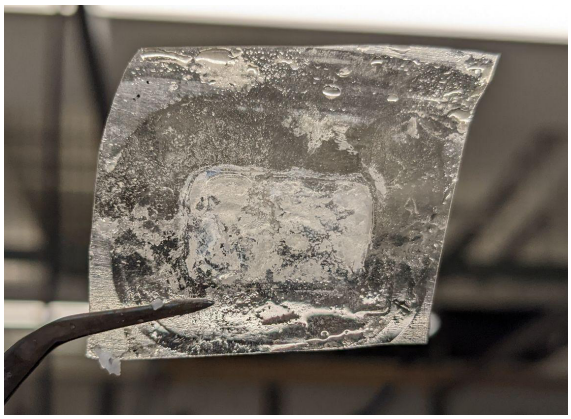

Figure S3. Optical photo of a Nafion 117 membrane after electrolysis. Noticeable $Ca(OH)_2$ was found on its surface.

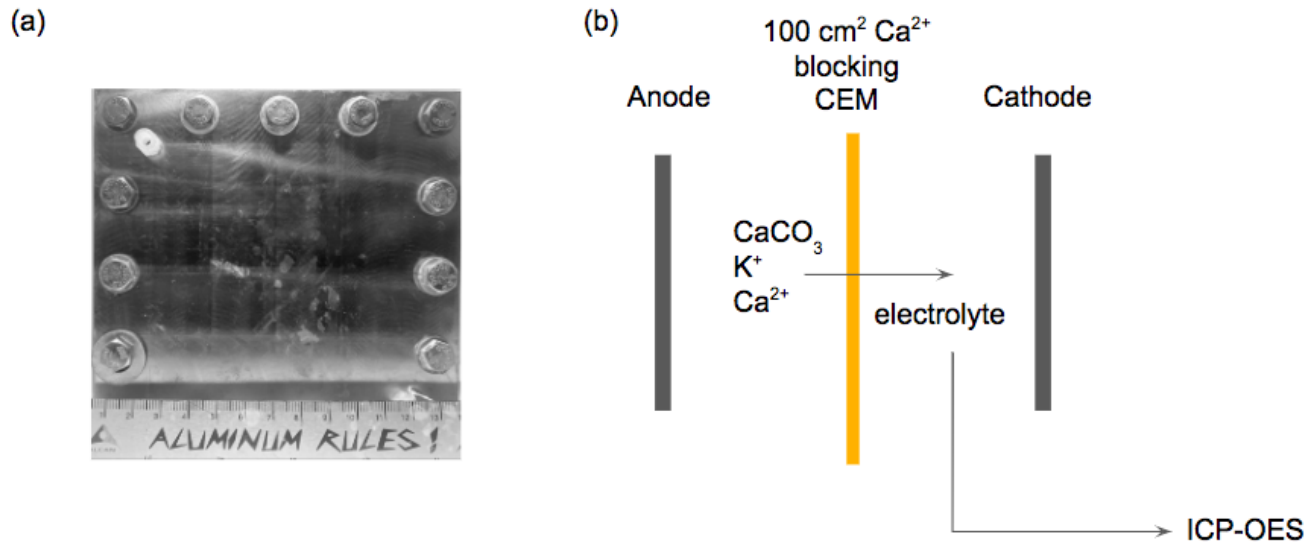

Figure S4. The optical photo of a 10 × 10 cm^2 (active area) electrolyser. For the Ca^{2+} blocking selectivity test, 0.05 M $CaCl_2$ in 1 M KCl solution with excessive $CaCO_{3(s)}$ was fed into the anode, and the catholyte was collected every 30 min for ICP-OES test to determine the [Ca^{2+}] for continuous 2 h.

Figure S5. The optical photo of the *operando* imaging system. The imaging system consisted of a modified two-compartment electrolyser with an open cathode compartment so we could image the CEM through the transparent catholyte with a high-resolution single-lens reflex (SLR) camera fixed above the electrolyser

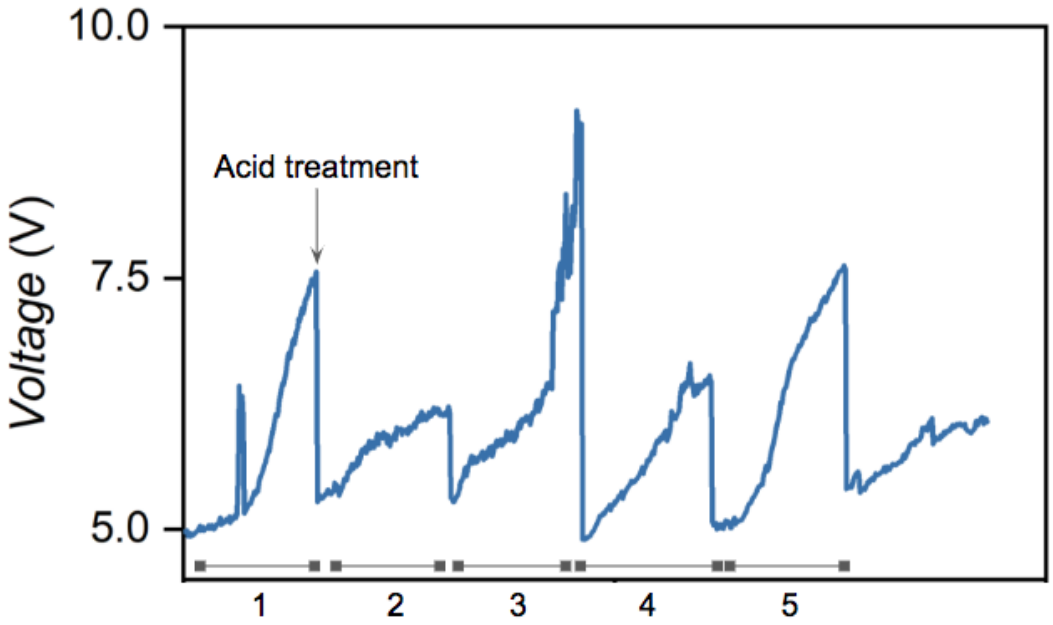

Figure S6. V_{cell} changes before and after acid treatment for the deposit removal. We performed cement electrolysis with a pristine Nafion 117 and we recorded V_{cell} with time. The electrolyser was stopped and the electrolyte at the cathode compartment was drained after a significant increase in V_{cell}. Then 1 M HCl was circulated to the cathode compartment to remove the $Ca(OH)_{2(s)}$ on the Nafion (towards cathode). This acid treatment can bring V_{cell} to the original voltage for multiple cycles. This result proved the deposit effect is reversible.

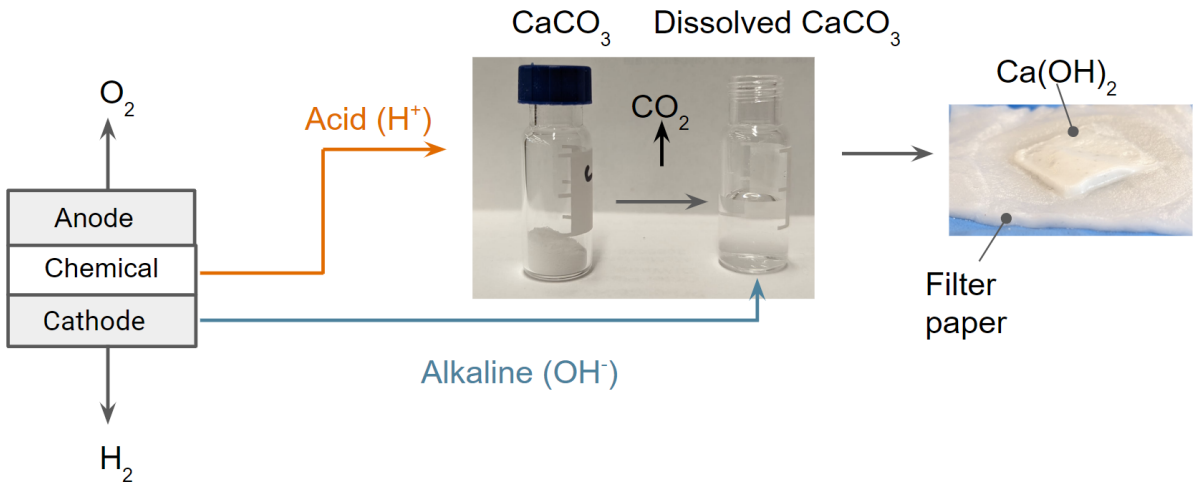

Figure S7. Electrochemical cement production using acid and base generated by an electrolyser. In this scenario, $CaCO_{3(s)}$ is not directly fed into electrolyser but reacts with the acid and base in sequence in a separated reactor - $CaCO_{3(s)}$ decomposes reacting with acids and releases Ca^{2+}, and it can be subsequently converted into $Ca(OH)_{2(s)}$ with additional bases.