# Supporting Information to the Article: Energy Recovery and Process Design in Continuous Flow-electrode Capacitive Deionization Processes

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### Experimental

#### **Density measurements**

Product water samples were taken during steady state operation. The density of each sample at 20°C was analyzed (DMA 46, Anton Paar GmbH). To verify the salt concentration of the samples a calibration curve was prepared (Fig. S1). The density was determined via interpolation. At concentrations above 50 g/L the correlation is not linear anymore, which becomes visible when compared to the trend line. The deviation between the interpolated salt concentration and the trend line is about 4.5 % at the maximum concentration measured in this work.



Figure S1: Calibration curve density measurements.

# Further results

#### System comparison

Figure S2 (a) shows the actual specific energy input (and recovery) for the different FCDI systems in kWh/m<sup>3</sup>. In this case (unlike Figure 4 (a) and (b) of the main manuscript), the values were not recalculated to represent full desalination. This demonstrates the importance of using energy input values normalized by the transferred salts instead of

normalization by the amount of product water. Generally, a comparison of energy demands normalized by the amount of product water is questionable, when not considering the feed and product salinities as well as the water recovery ratios.

Figure S2 (b) shows the measured electric currents over time during CV operation of three different systems. Only in case of the energy recovery system the operational mode was set to a constant current of 0.3 A for the regeneration module. The measured electric currents were fairly constant during steady state operation.



Figure S2: Comparison of continuous FCDI system configurations (symbols given in Fig. 1 of the main manuscript); All experiments performed with 25 wt% AC carbon powder and 60 g/L NaCl in the flow-electrodes, flow-electrode flow rates of 200 mL/min, diluate feed flow rates of 1.2 mL/min and concentrate feed flow rates of 1 mL/min. (b) Measured electric currents over time during sample experiments in energy input mode, single module configuration and energy recovery mode during steady state operation of FCDI systems.

#### Pressure drop

The hydraulic energy which has to be applied by pumping was determined by measuring the pressure drop in a flow-electrode flow channel of a FCDI half-cell with pressure sensors placed at the inlet and outlet. The results are plotted in Figure S3.



Figure S3: Pressure drop measured in the flow-electrode channel of a single FCDI half-cell.

#### Theoretical energy demand

Figure S4 shows the theoretical minimum energy demand of FCDI processes in Wh/g NaCl (equal to kWh/kg) depending on the current efficiency (CE) and the applied voltage (Eq. S1).

$$E_{el/mass} = \frac{F \cdot n_{NaCl} \cdot U}{CE} \tag{S1}$$

In Equation S1,  $E_{el/mass}$  represents the mass-specific electrical energy required for desalination in a single module FCDI system or two module FCDI system with short circuit regeneration (Mode A1, Fig.1 of the main manuscript), F is the Faraday constant,  $n_{NaCl}$  is the molar amount of transferred sodium chloride, U is the applied voltage, and CE is the current efficiency. Accordingly, the energy demand increases in a hyperbolic manner with decreasing current efficiency and linearly with increasing voltage. This corresponds well with findings presented in electrodialysis literature, such as the increase in energy consumption at increasing current densities (and thus applied potentials) and increasing NaCl concentrations.<sup>1-3</sup> The latter is likely caused by decreasing current efficiencies due to a diminished Donnan exclusion and thus decreasing selectivities of ion exchange membranes at high salt concentrations.



Figure S4: Theoretical minimum energy demand for the complete desalination of NaCl solutions using FCDI processes, depending on the current efficiency and voltage given in Wh/g NaCl.

## References

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