Electronic Supplementary Material (ESI) for Lab on a Chip.

Supporting Information

Overcoming transport limitations in miniaturized electrophoretic delivery devices

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Evaluation of the Ag/AgCl electrode capacity

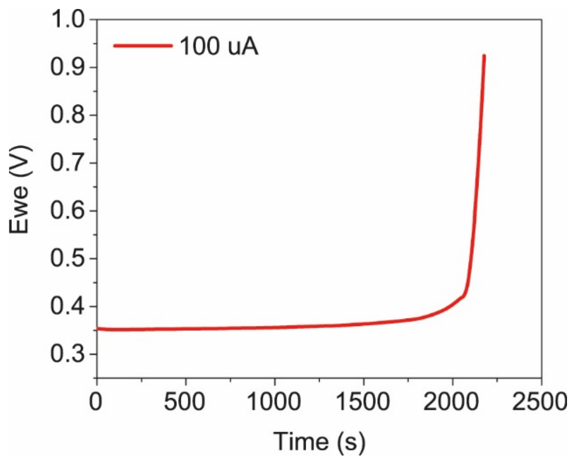

Figure S1. Chronopotentiometry graph/time-potential curve of three electrode system, in which a current was applied between the auxiliary and working Ag/AgCl electrodes, and the potential of the working electrode (measured with respect to the reference electrode).

Computational Simulations

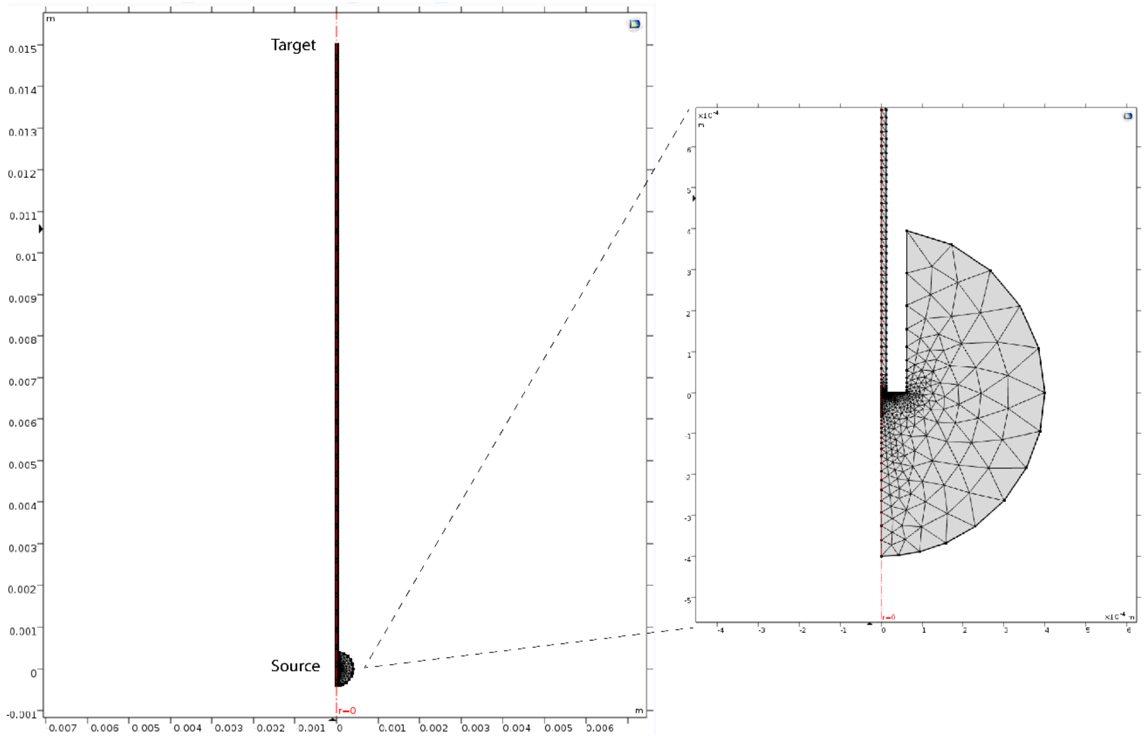

Figure S2. The model assumes a homogeneous membrane with a fixed charge concentration cfix. A 2D axisymmetric geometry was used for the model, effectively making it a 2D computational problem.

Investigation of limiting current for different capillary geometrical characteristics

Fiber capillary OEIPs with 15 mm long /25 µm and 50 µm inner diameter CEM channels and 7 mm long/ 25 µm inner diameter were fabricated to investigate the effect of geometrical characteristics of the capillary pump at the limiting current and the water splitting regime. To investigate the limiting current for the fiber CEMs, the source reservoir was loaded with a 100 mM HCl (aq) solution. Then, the voltage was sourced within the range of 0-5 V and the currents were recorded at a scan rate of 5 mVs-1, for different HCl concentrations (10 mM, 1 mM, 0.1 mM). The limiting current and threshold voltage for capillaries with 50 µm inner diameter match the measured data for capillaries with 25 µm reasonably well (Supplementary figure S3.B). The geometrical characteristics of the capillary pump affect its resistance, thus affecting the limiting current and the water splitting regime. As the channel is an ionic resistor, a shorter channel results in a lower resistance, meaning that the limiting current is reached for a lower voltage. The length of the channel doesn't affect the limiting current density but the threshold voltage (Fig. S3C). This can be shown by the addition of the cap at the inlet which lowers the resistance at the interface between channel and electrolyte. The lower resistance reduced or even eliminated the limiting current and the water splitting effect.

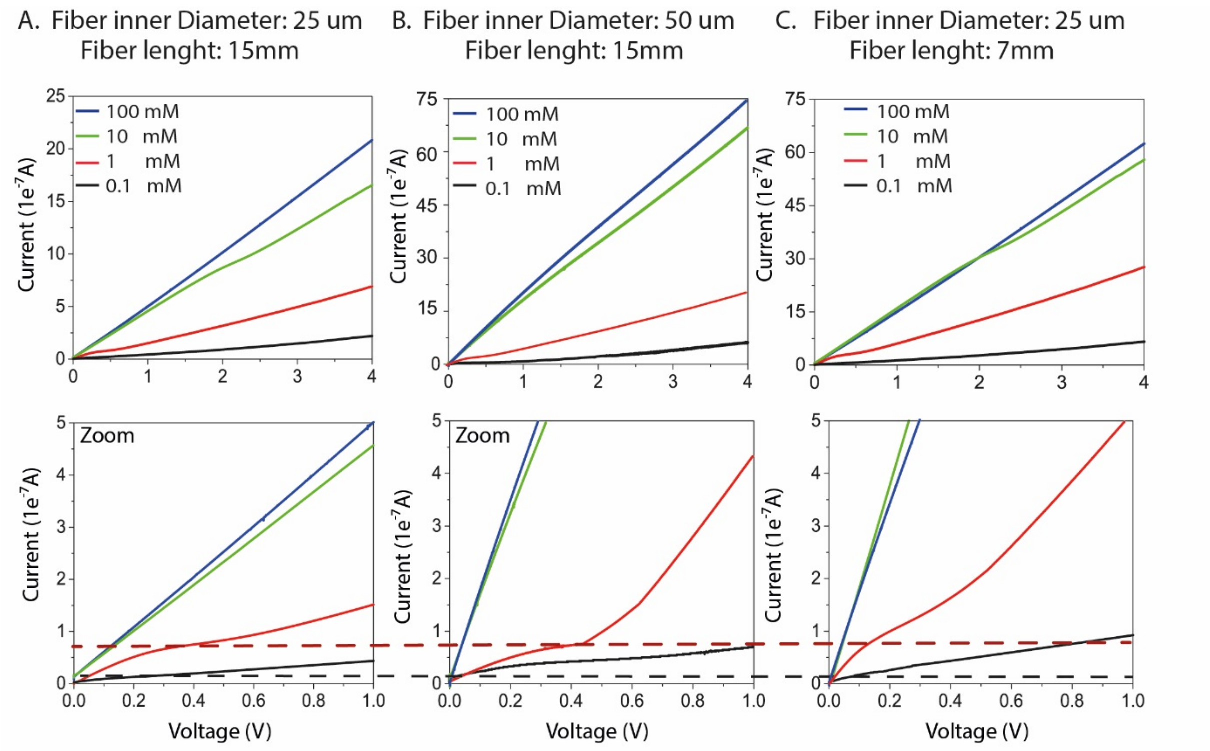

Figure S3. Experimental I-V curves with the three characteristic regions for HCl at different concentrations (100 mM, 10 mM, 1 mM, 0.1 mM) for different capillary geometrical characteristics: A) 25 µm inner diameters/ 15 mm length, B) 50 µm inner diameters/ 15 mm length, C) 25 µm inner diameters/ 7 mm length, in the range 0-5 V scanned at a rate of 5 mVs^{-1}. The dashed lines show the calculated limiting currents from the computational model for the HCl electrolyte at different low concentrations.

Investigation of pH changes for buffered electrolytes

A pH change can occur in buffers very locally. However, a buffer has the ability to maintain its pH and in order to change the pH in buffer one must apply a high concentration of H^+ – higher than the capacity of a buffer. Typical biological buffer capacities are on the order of 10–100 mM. We used a phosphate-buffered saline (PBS) solution (pH 7.5) with a methyl red pH indicator as a target electrolyte to check if there was a pH change around the capillary inlet for this buffer solution. The OEIP was operated continuously in the range 0-3.5 V and microscope images were taken every 1.5 min for 1 mM KCl delivery (Fig. S4). We didn't observe any color change at the fiber outlet without significant pH changes. We also calculated the pH change in PBS 1x with 400 µl volume and concentration of acid and conjugated base together 10 mM, and initial pH 7.5. If we operate our devices at 3 V, providing a current of 115 nA for 10 hours, the pH in the whole bulk will change from the initial 7.5 to 7.48 giving a negligible ΔpH 0.02. So there should be no significant pH changes in buffer solutions even with low buffer capacity.

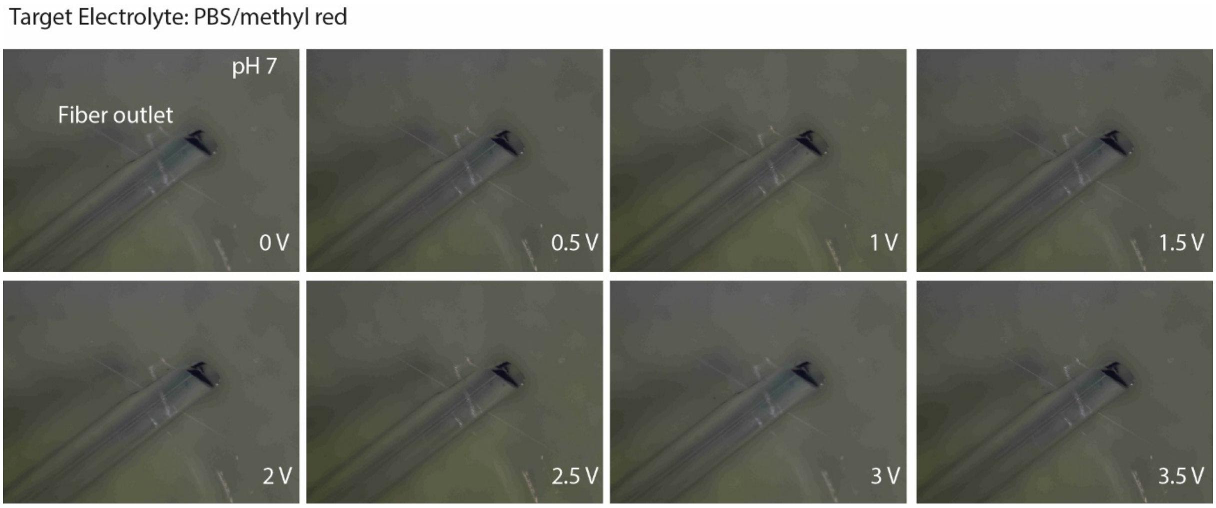

Figure S4. Microscope images of pH changes at the outlet during KCl (1 mM) delivery.