

## Supporting Information Soft Matter Journal

### Towards synergistic oscillations in enzymatically active hydrogel spheres

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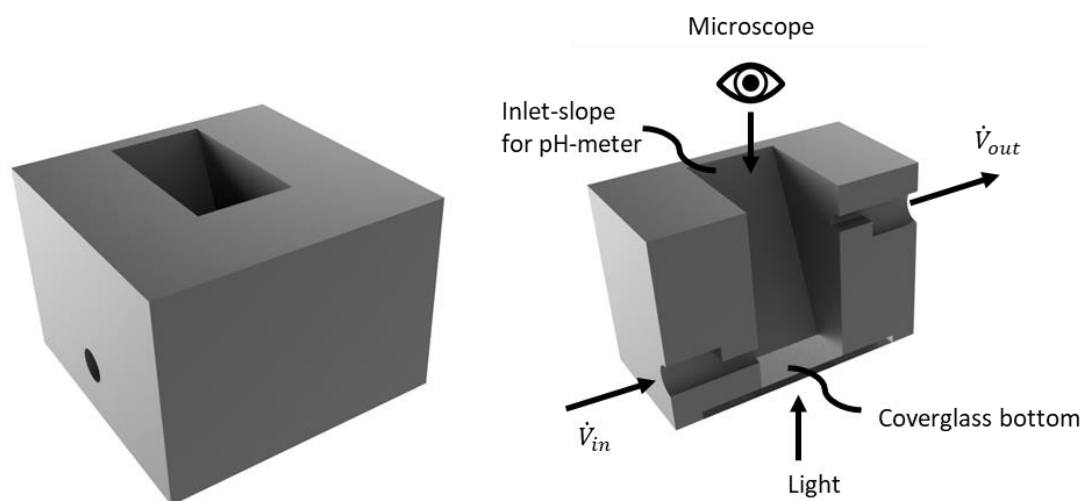
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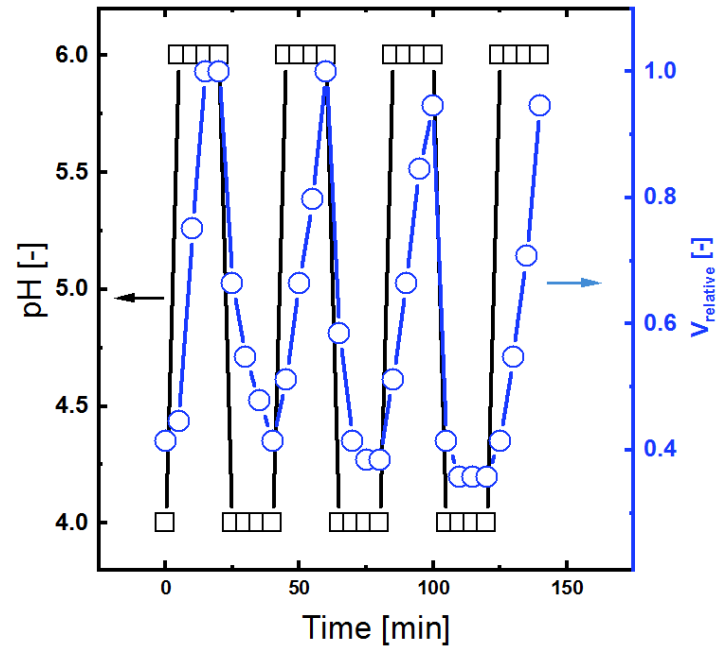
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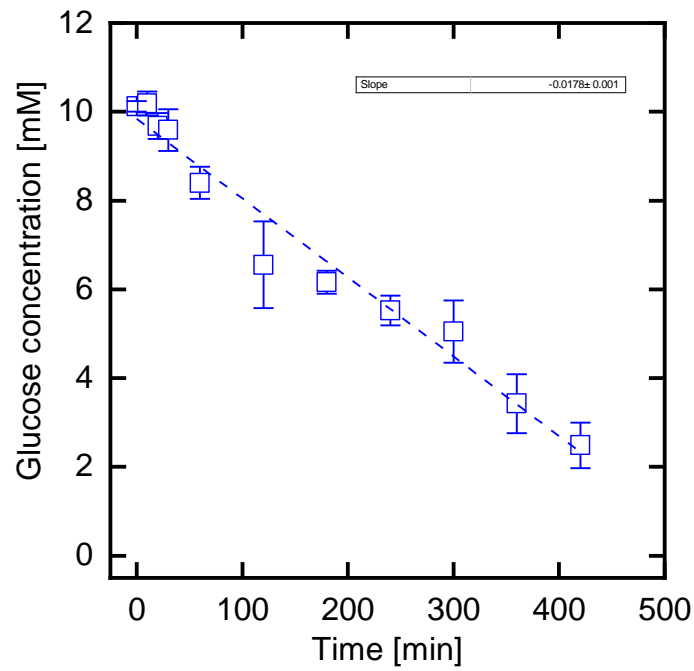
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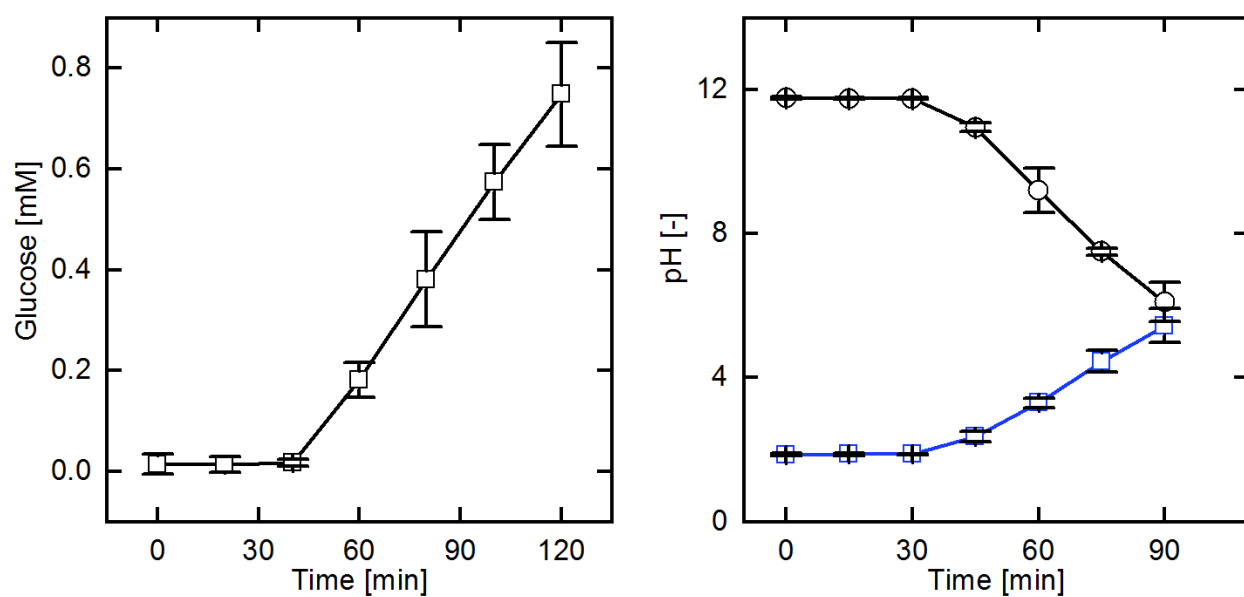
**Figure S1.** Design of flow cell used in oscillation experiments. The cell is filled with nutrient solution and constantly resupplied from the left inlet. The hydrogel spheres sediment to the glass bottom. A pH probe is inserted from the top. The hydrogel diameter can be observed through a microscope.



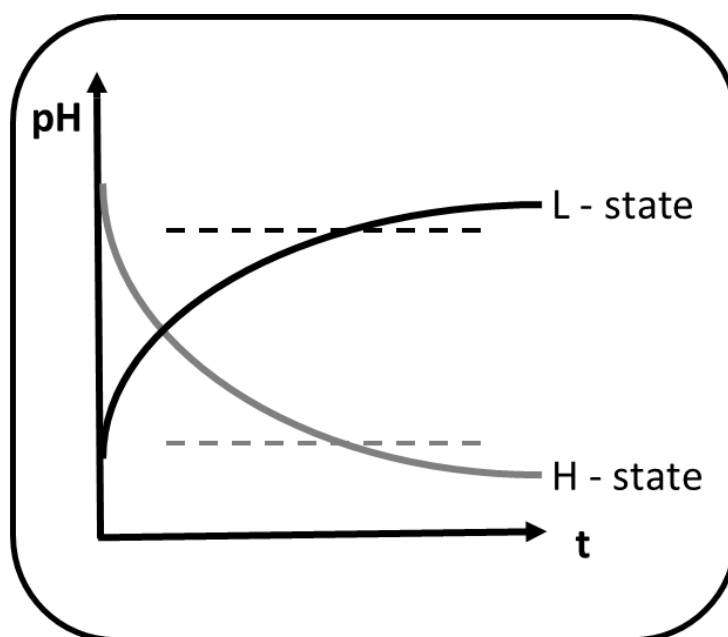
**Figure S2.** Oscillatory swelling and collapsing response (blue) of the hydrogel particles exposed to alternating pH-cycles (black) of 50 mM NaCl solution at 37°C of pH 4 or pH 6.



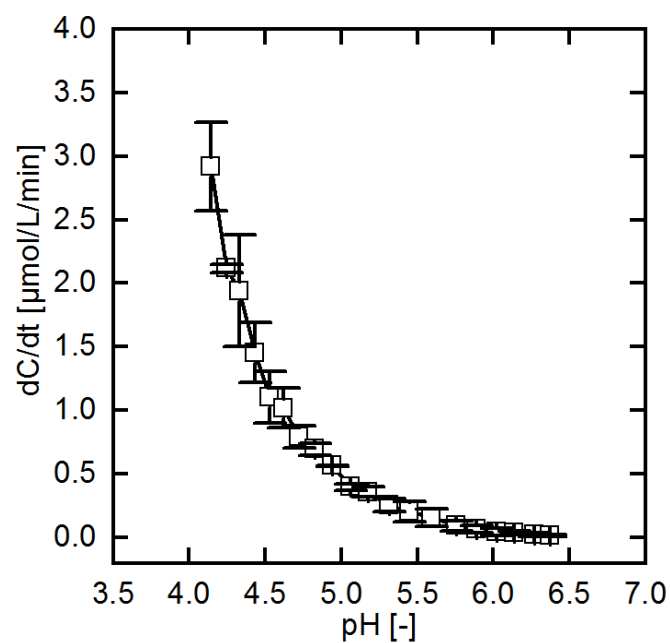
**Figure S3.** Measurement of the enzyme kinetics of the free enzyme in 10 mM glucose solution at pH 5 in 50 mM sodium acetate buffer at 37°C.



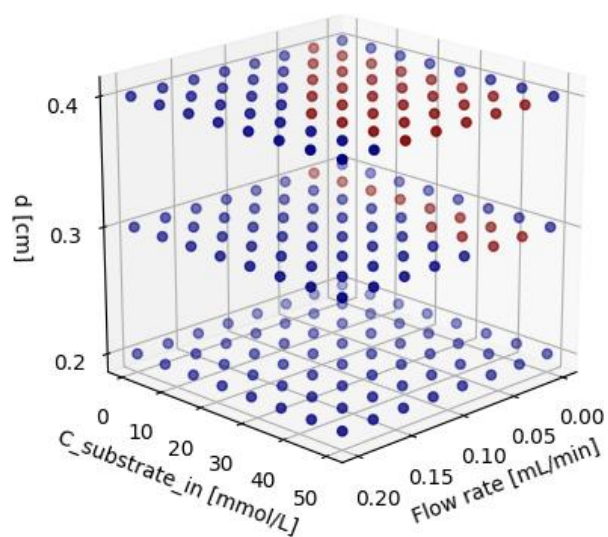
**Figure S4.** Measurement of the diffusional coefficients across a P(NIPAM-co-AAC)/Alginate flat sheet membrane of glucose (left) showing the concentration increase in the acceptor cell and protons (right) with the pH in the donor cell in blue and in the acceptor cell in black.



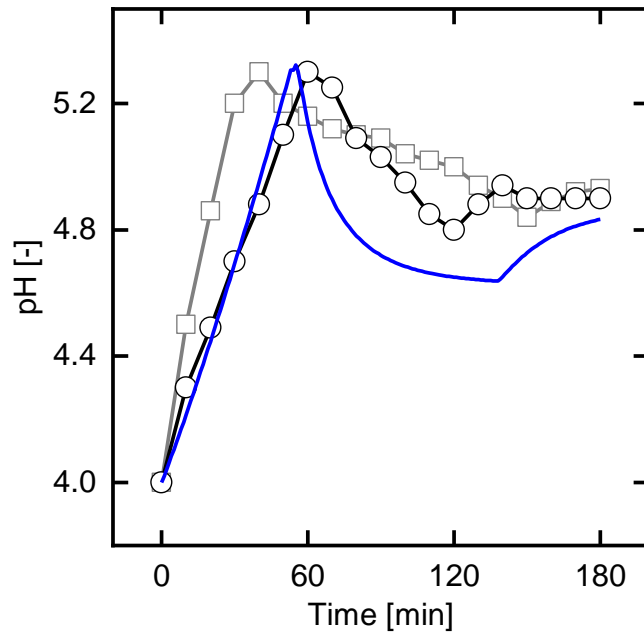
**Figure S5.** Schematic representation of decision guidance for parameter screening.



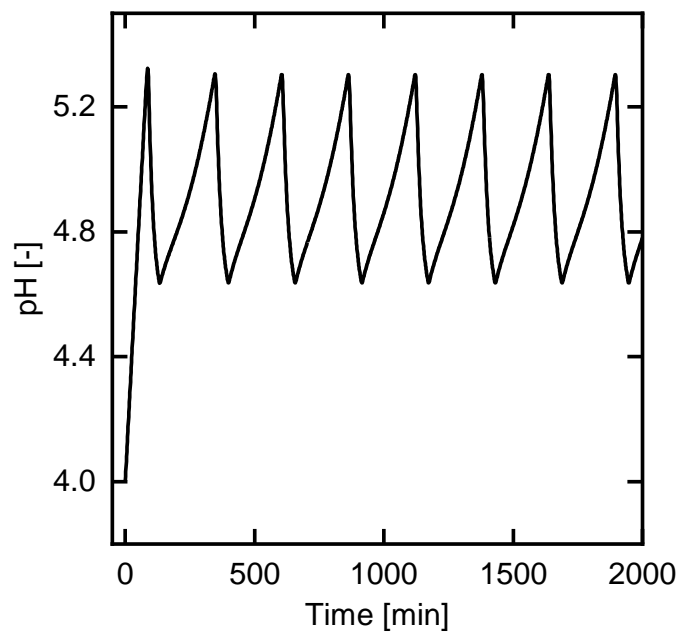
**Figure S6.** Measured pH-dependent marble kinetic in 50 mM NaCl solution.



**Figure S7.** Oscillation screening for different parameter sets (flowrate, glucose feed concentration and diameter), whereby each set is represented by one point. Red points indicate oscillatory behaviour, while blue points indicate that no oscillations occur.



**Figure S8.** Comparison of the simulated pH-value development over time (blue solid line) and the corresponding experimental data (symbols, grey and black) at a flowrate of 20  $\mu\text{L}/\text{min}$ , a feed glucose concentration of 50 mM and a sphere diameter of 0.4 cm.



**Figure S9.** Simulated pH-value development over time at a flowrate of 20  $\mu\text{L}/\text{min}$ , a feed glucose concentration of 50 mM and a sphere diameter of 0.4 cm, showing sustained oscillations.