Probing the electrochemical behaviour of tubular enzyme membrane electrodes using simulations

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Introduction

Tubular geometry can help us emulate biological systems. Applications include medical sensors and biofuel cells among others.

Methods

- Tubular enzyme electrodes were prepared by coating the inside of a hollow graphite cylinder with a mix of redox polymer and enzyme.
- Electrolyte of different viscosities were pumped through the tubular carbon working electrodes.
- Change in diffusion coefficient was measured using microelectrode experiments (using RuHex(III)Cl as electrolyte).
- Electrochemical response via chronoamperometry at 10, 20, 50 and 100 mM glucose and fitted to the transient model.

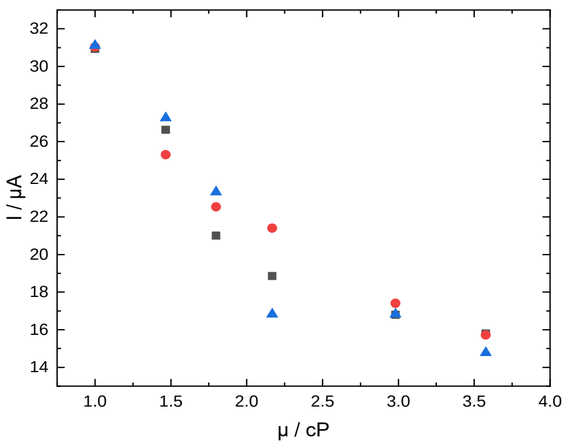

Figure 1: Plot of steady state current as a function of electrolyte viscosity at a D-glucose concentration of 20 mM in a 50 mM PBS buffer (7.45 pH). The reading was repeated for 3 similarly prepared electrodes. [m]=2.4E-04mol/cm³.

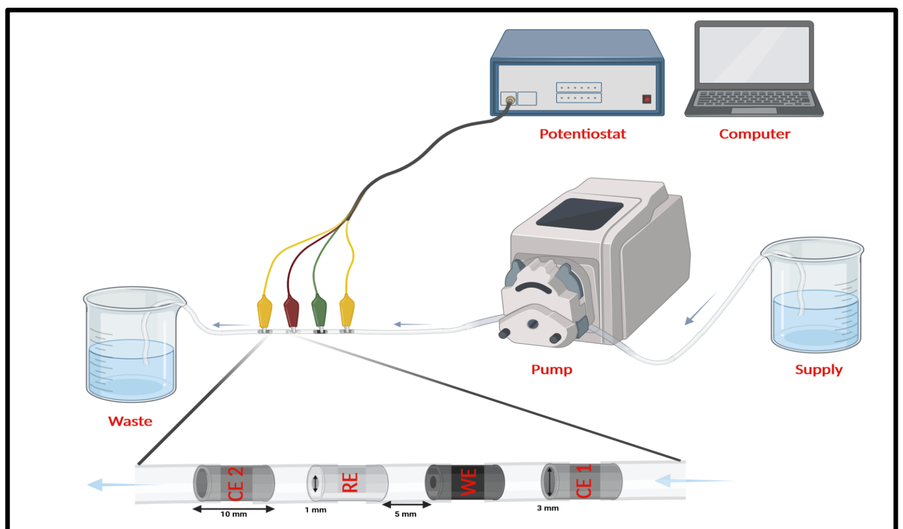

Figure 2: The experimental setup consists of a buffer supply with glucose concentration. The buffer's viscosity can be altered using glycerol and xanthan gum. It is pumped through the electrodes via a peristaltic pump while potential is applied using computer controlled potentiostat.

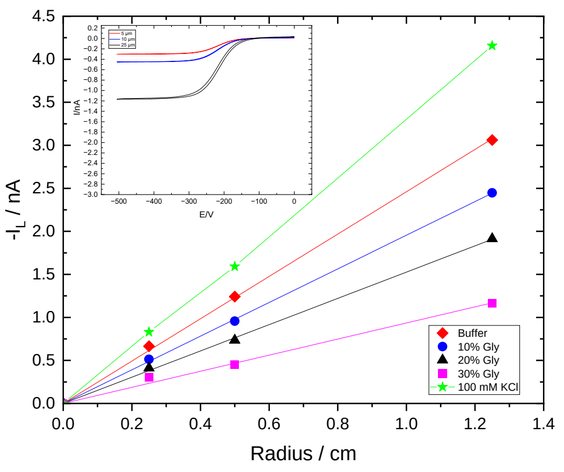

Figure 3: Limiting current at varying microelectrode radii r = 5, 10 and 25 µm. Inset: representative microelectrode voltammograms with potentials scanned at 5 mV s^{-1}. Red: 5 µm, blue: 10 µm, black: 25 µm for 30 % glycerol.

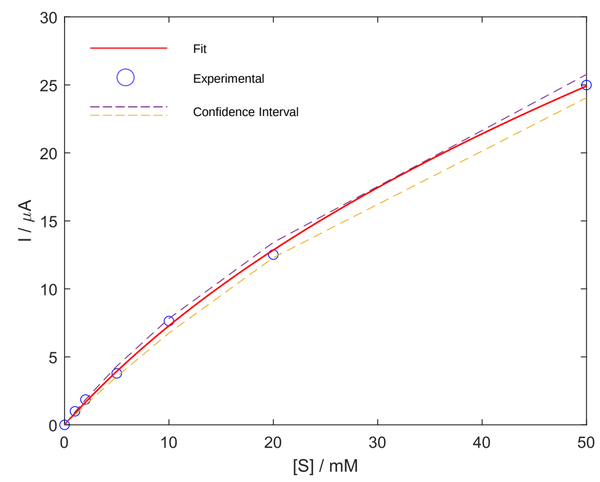

Figure 4: Amperometric response against the following substrate values [0,1,2,5,10,20, 50] mM at viscosity of approximately 1 cP (plain water). The electrode was stepped to 350 mV vs. Ag/AgCl at a flowrate of approximately 0.04 mL/s in a PBS buffer (7.45 pH).

Conclusions/Learning

1. Viscosity has a clear effect on the steady state response because it changes the properties of the redox polymer layer (D_m of the redox polymer).
2. The change in viscosity effects the diffusion properties of the substrate/analyte into the polymer film as well.
3. Varying the flow rate had no effect on the steady state electrochemical response.
4. Transient state response depends on extra parameters compared to the steady state limiting current.

Results

Fitted chronoamperograms with respective concentration profile animations (accessible via QR code) against 20, 50 and 100 mM glucose concentration.

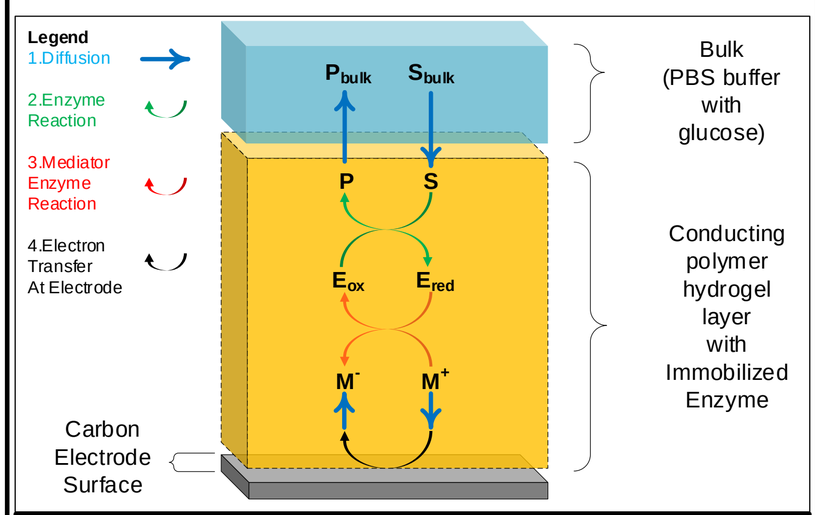

Figure 5: Kinetic schematic of a carbon tubular enzyme membrane electrode depicting the processes occurring inside the hydrogel layer (orange), electrode surface (grey) and the flowing bulk (blue).

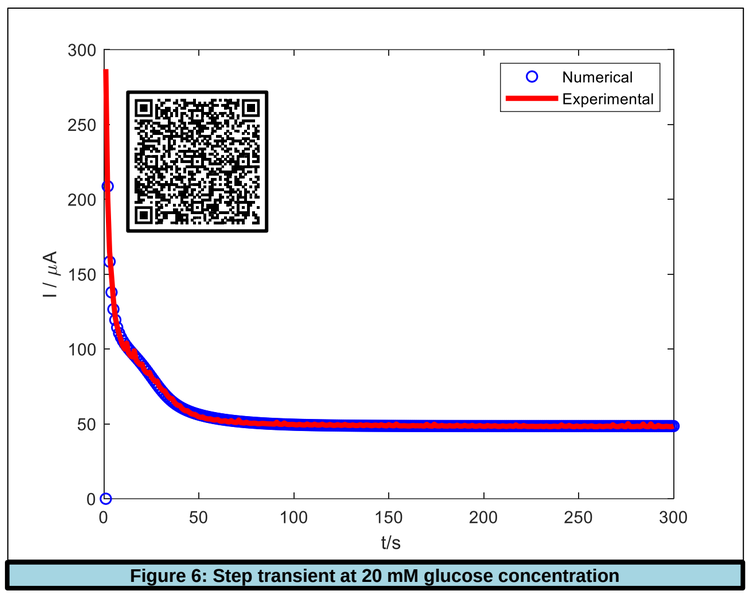

Figure 6: Step transient at 20 mM glucose concentration

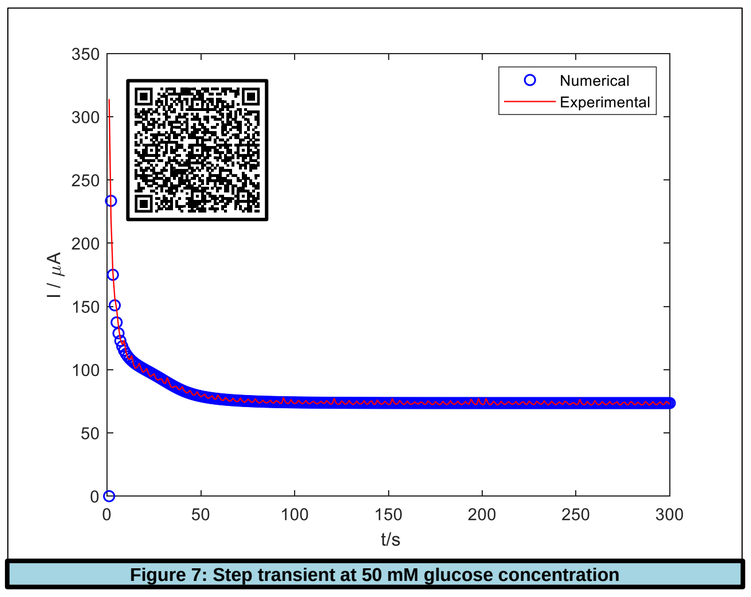

Figure 7: Step transient at 50 mM glucose concentration

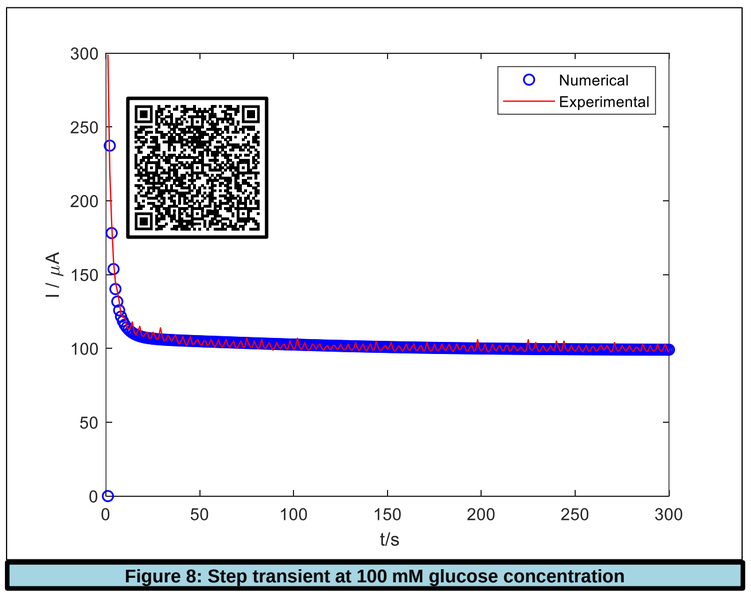

Figure 8: Step transient at 100 mM glucose concentration

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