Fabrication of pH Sensors Modified with Platinum and Iridium Oxide

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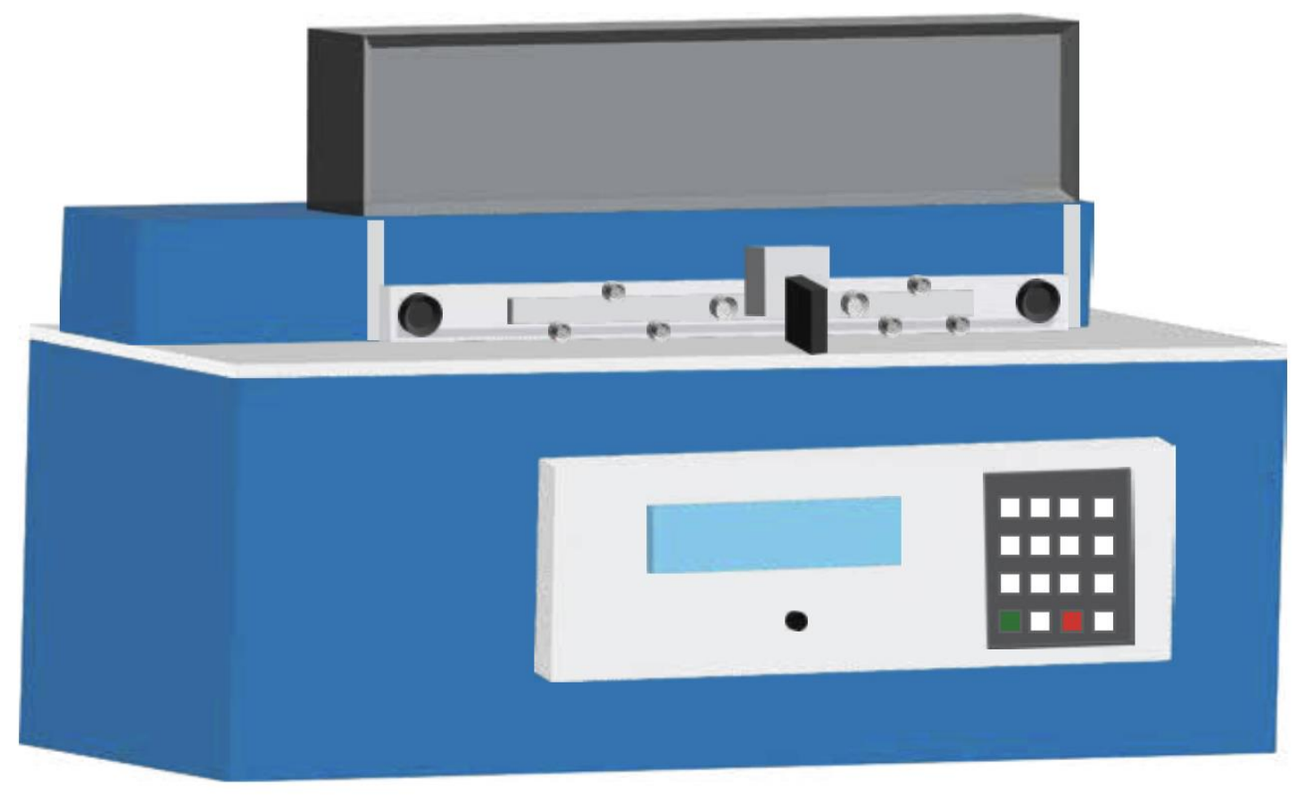
Introduction

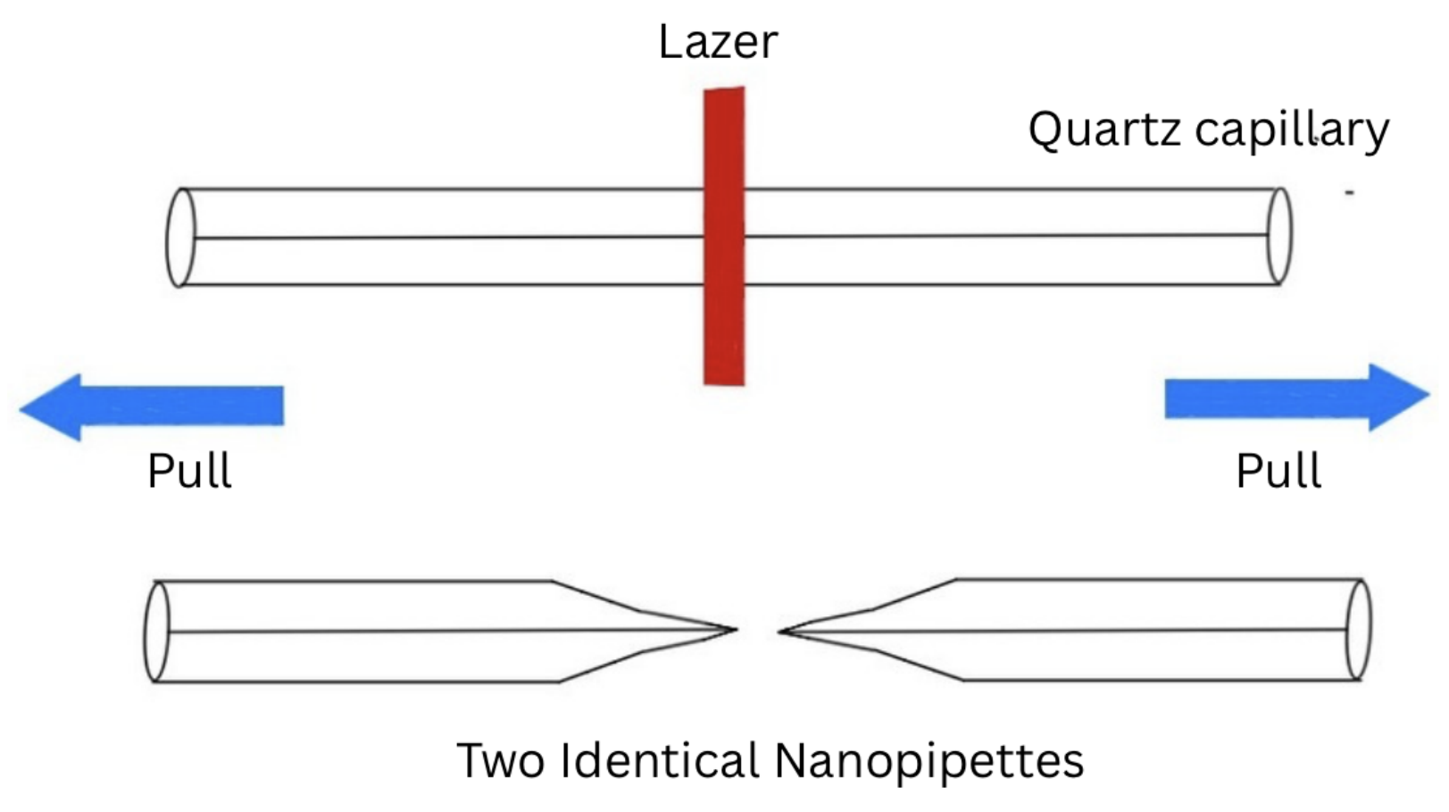
- Detecting small pH changes is important for studying biological systems, especially the tumor microenvironment, where cancer cells metabolism lowers the extracellular pH
- In this project, carbon nanoelectrodes were fabricated and modified with platinum and iridium to function as stable pH sensors
- Cyclic voltammetry was used to characterize the electrochemical behavior of the carbon nanoelectrode surface
- Platinum deposition enhanced durability, while iridium deposition enabled pH sensitivity
- The electrodes were calibrated using open circuit potential (OCP) measurements across different pH solutions
- Calibration curves were compared with the theoretical Nernst equation response
- The sensors showed near-Nernstian sensitivity, reproducible measurements, and stable performance over multiple days
- These electrodes show promise for monitoring localized pH changes related to cancer cell metabolism

Nanoelectrode Fabrication

Step 1: Fabrication of Nanopipette

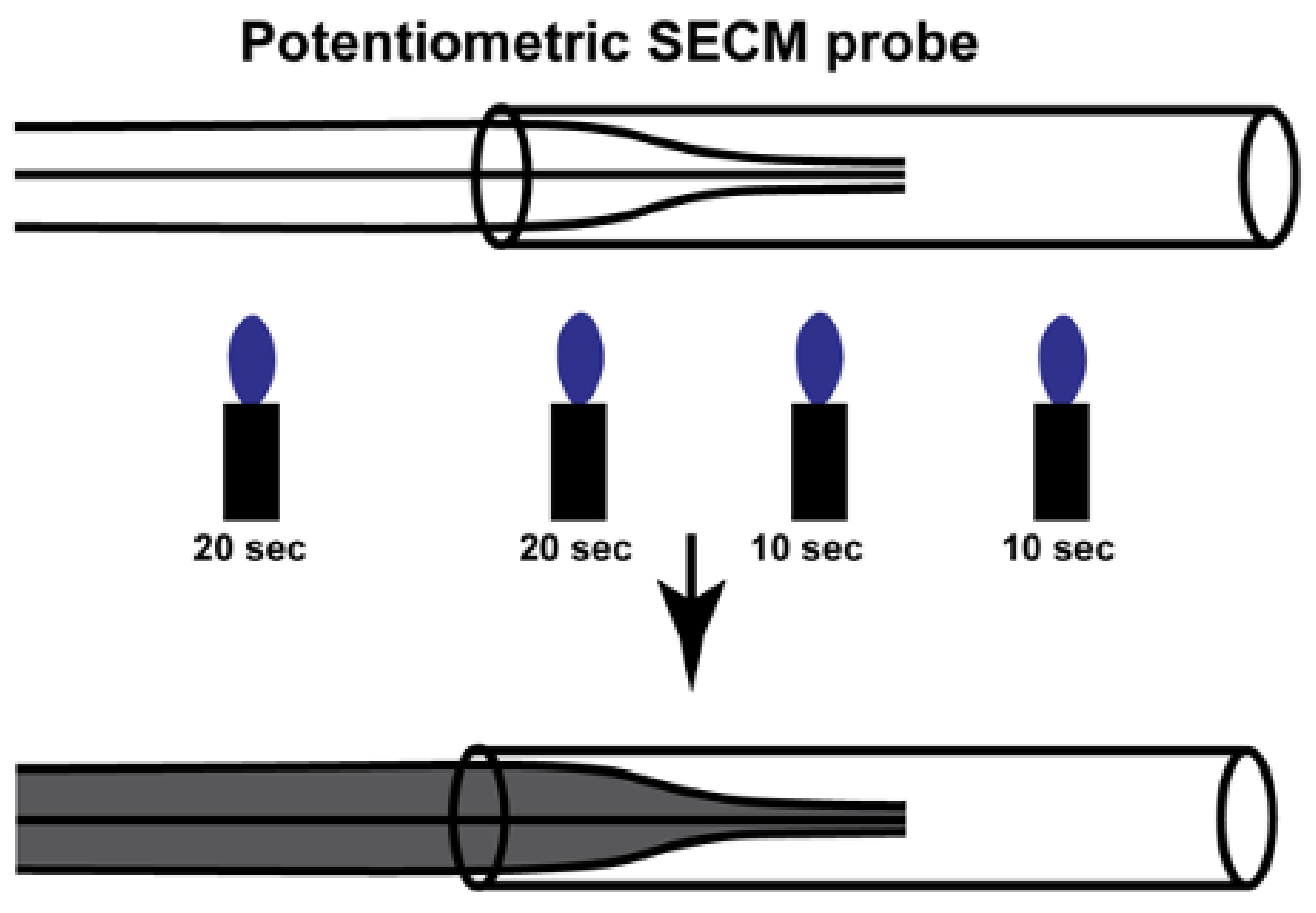


Nanopipettes are fabricated from quartz capillary tube with I.D: 0.9 mm and O.D: 1.2 mm

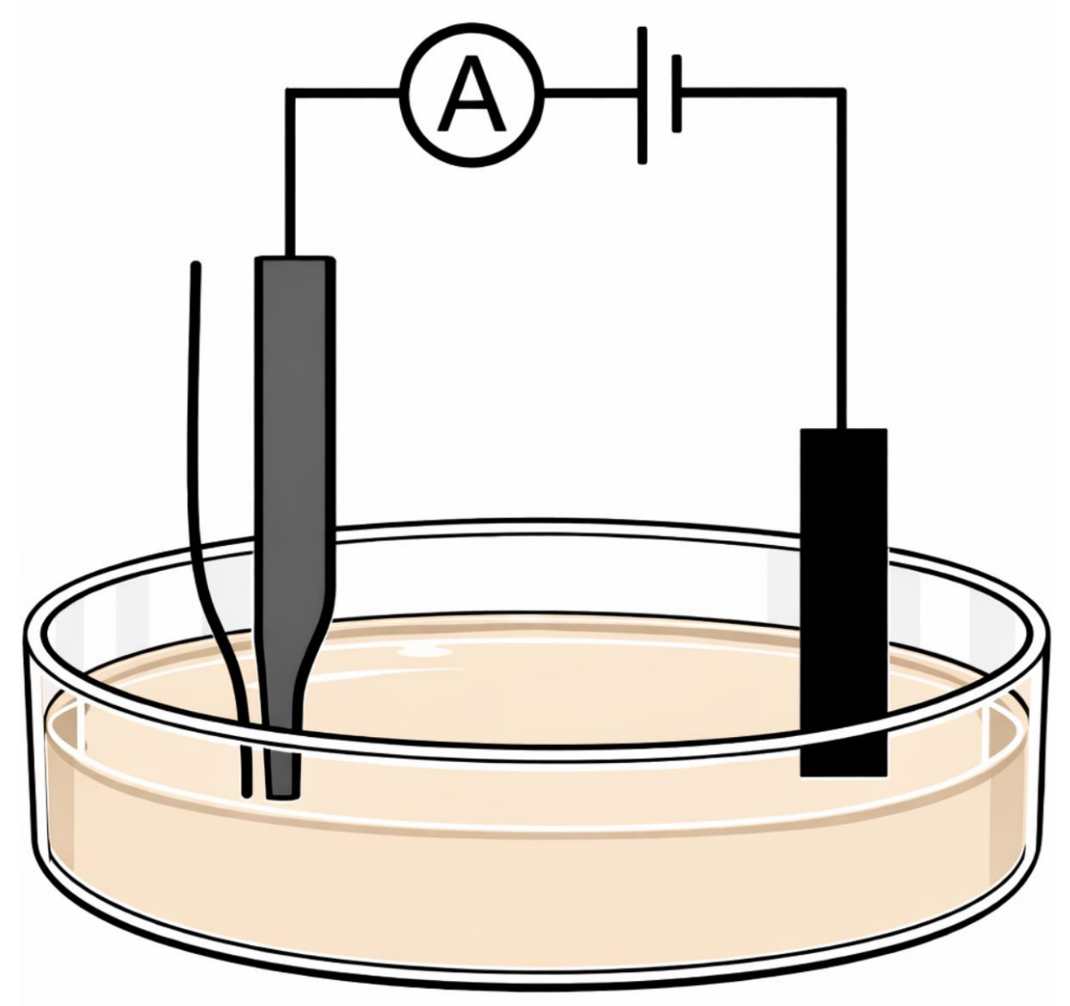


Fabrication of SECM imaging probe using a laser puller

Step 2: Fabrication of Carbon Nanoelectrode



Carbon Pyrolysis on nanopipettes

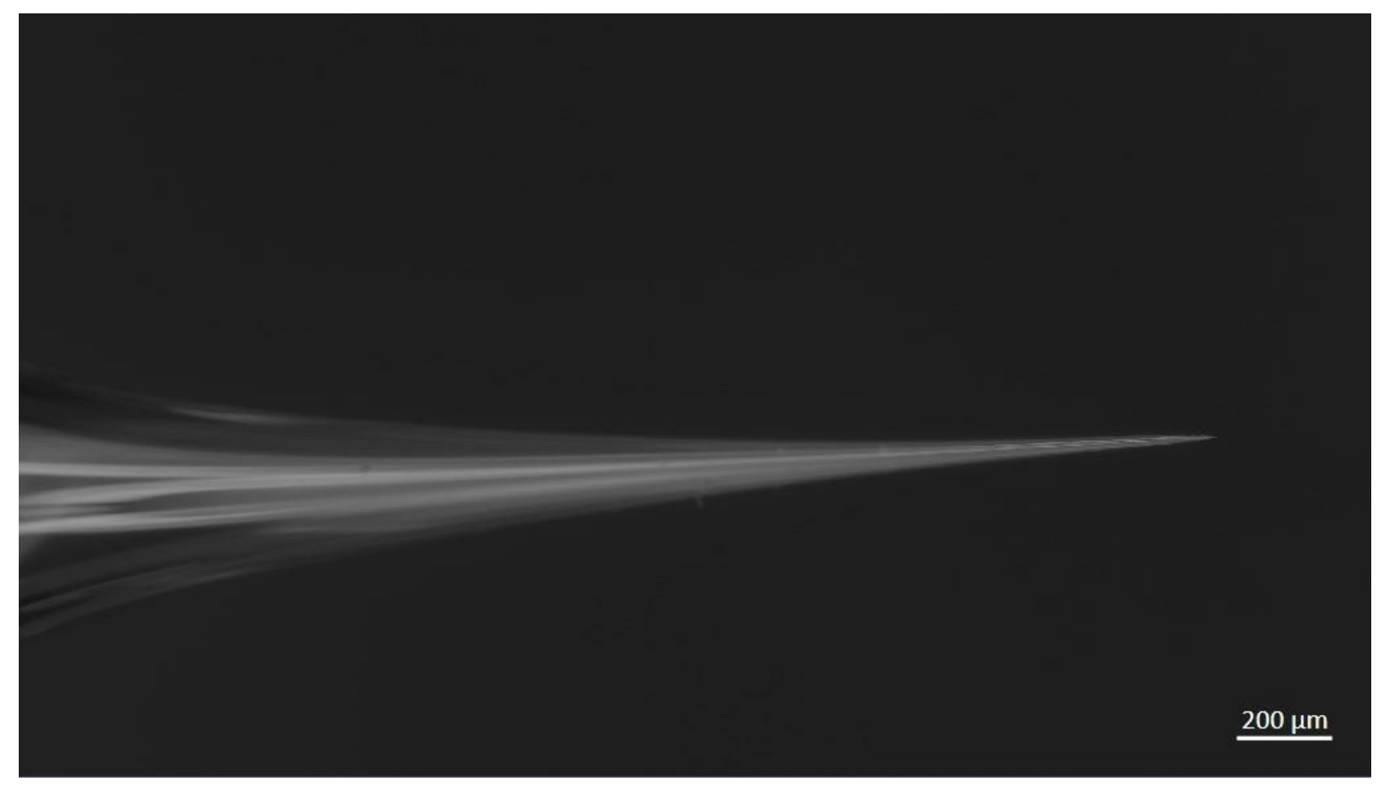


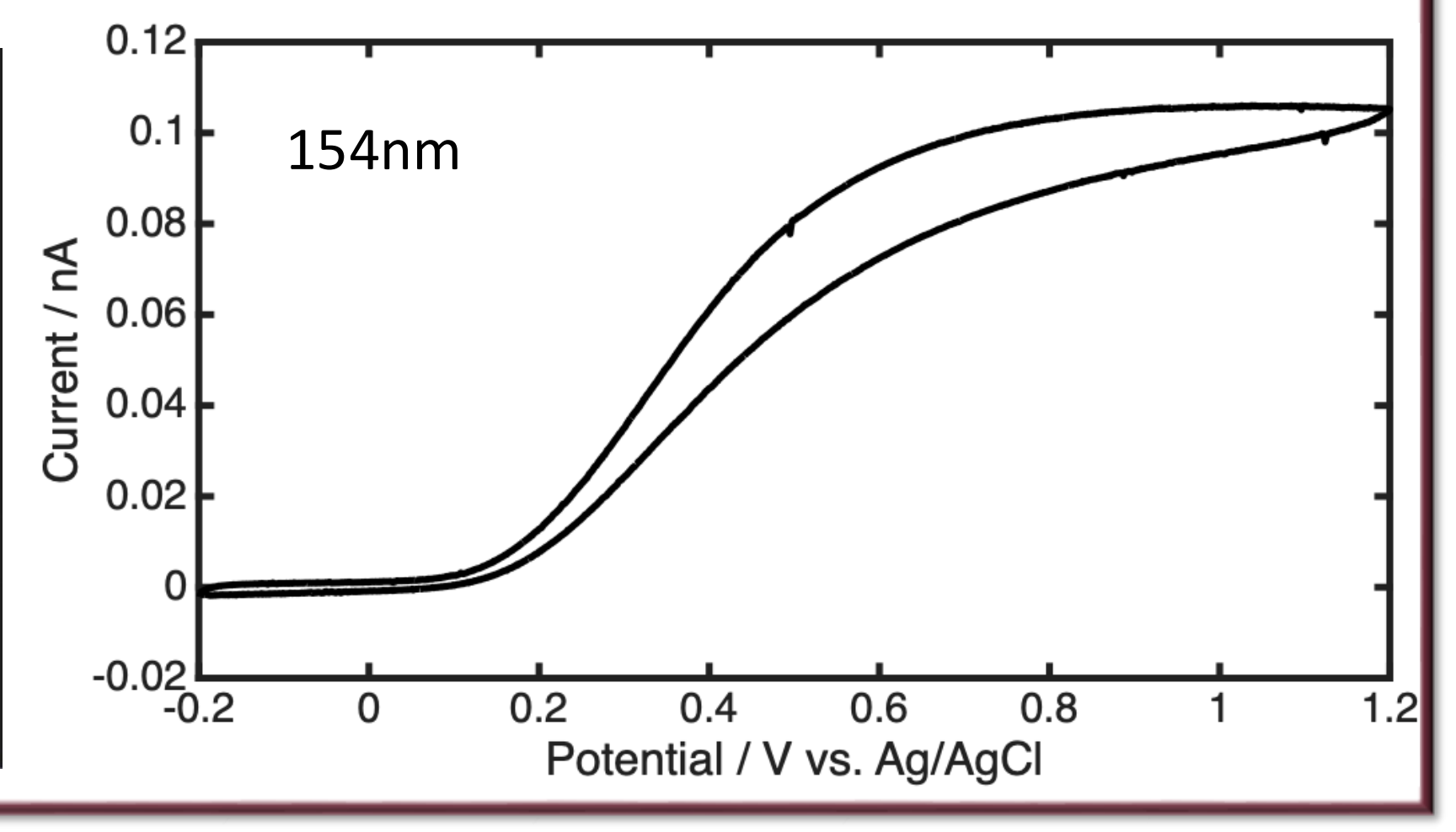
Step 3: Characterization of Carbon Nanoelectrode using Cyclic Voltammetry

Equation used for calculating Nanoelectrode tip size

$i_{lim}=4.64nFCDr$

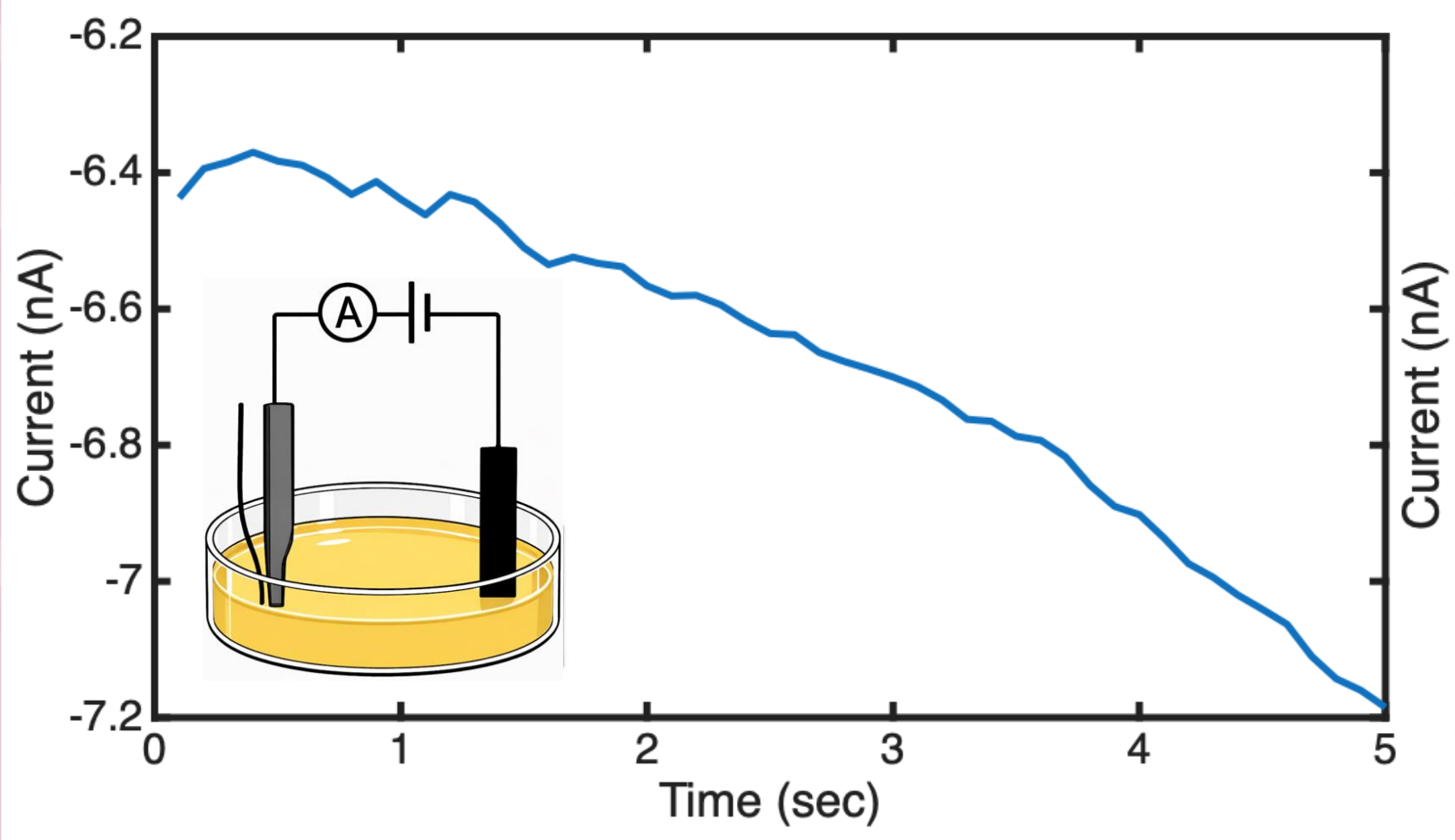
n = number of electrons transferred
F= Faraday's constant
C= analyte concentration
D = Diffusion coefficient



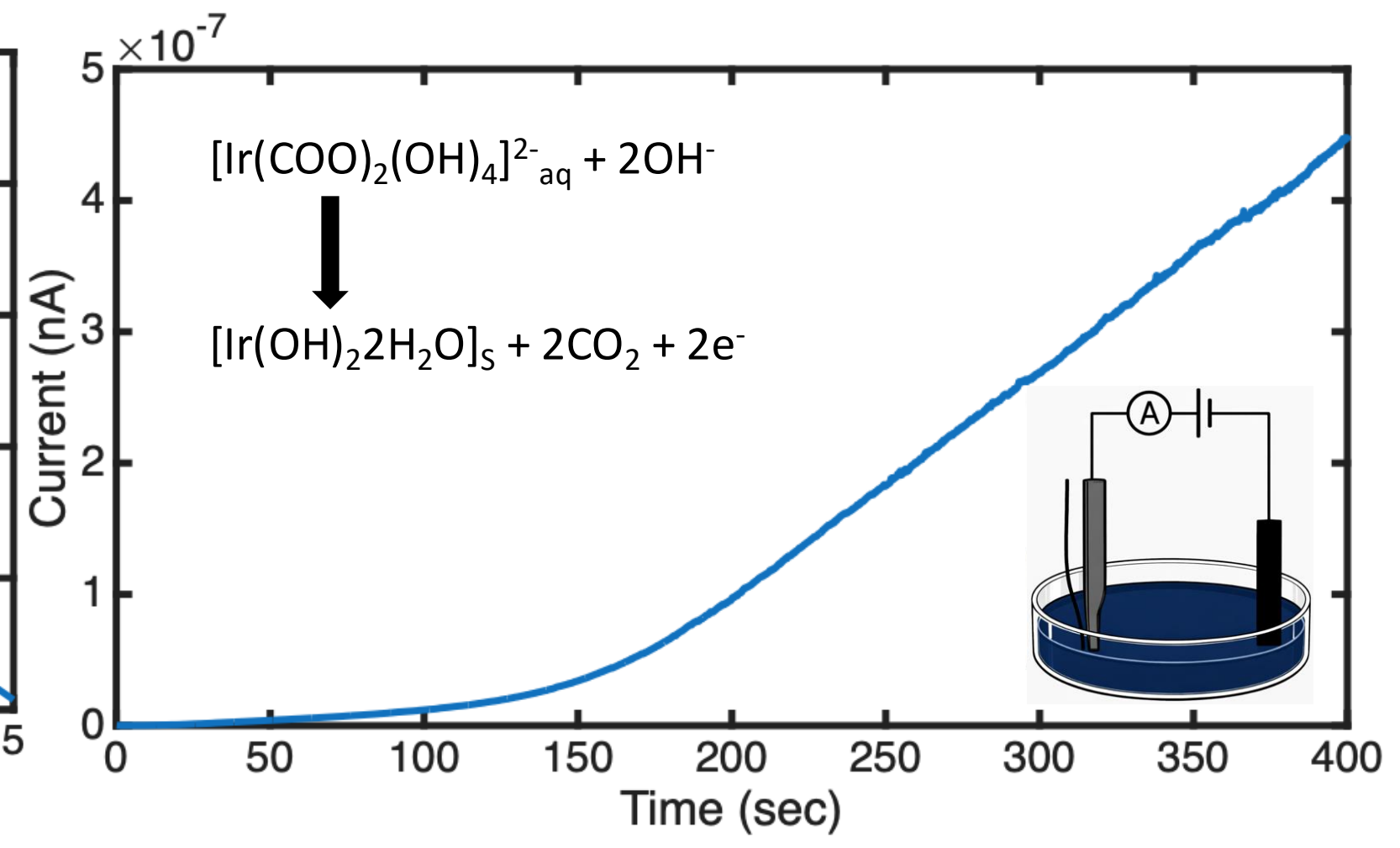


Fabrication of Nanoscale pH Sensors

Step 4: Platinum Deposition



Step 5: Iridium Oxide Deposition



$$[Ir(COO)_2(OH)_4]^{2-}_{aq} + 2OH^- \rightarrow [Ir(OH)_2 2H_2O]_S + 2CO_2 + 2e^-$$

$E_{Cell} = E^0 - (RT/nF)\ \ln Q$

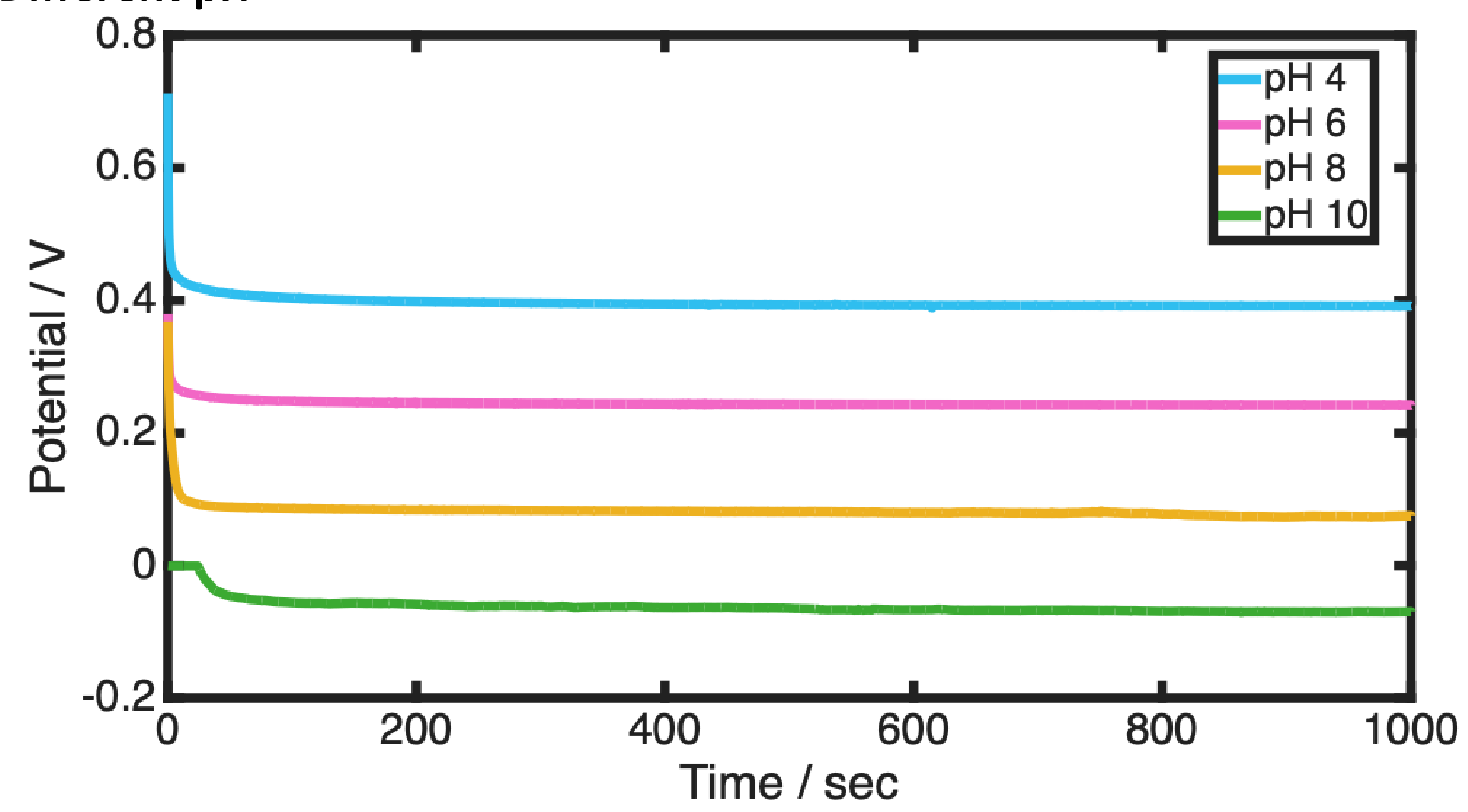
E_{cell} = Cell potential
E^0 = Cell potential under standard conditions
R = Universal gas constant (8.314 J/Kmol)
T = Temperature
n = Number of electrons transferred in the reaction
F = Faraday constant (96485 C/mol)
Q = Reaction quotient

- The Nernst equation enables the determination of cell potential under non-standard conditions. At 25°C (298K), the equation simplifies to:

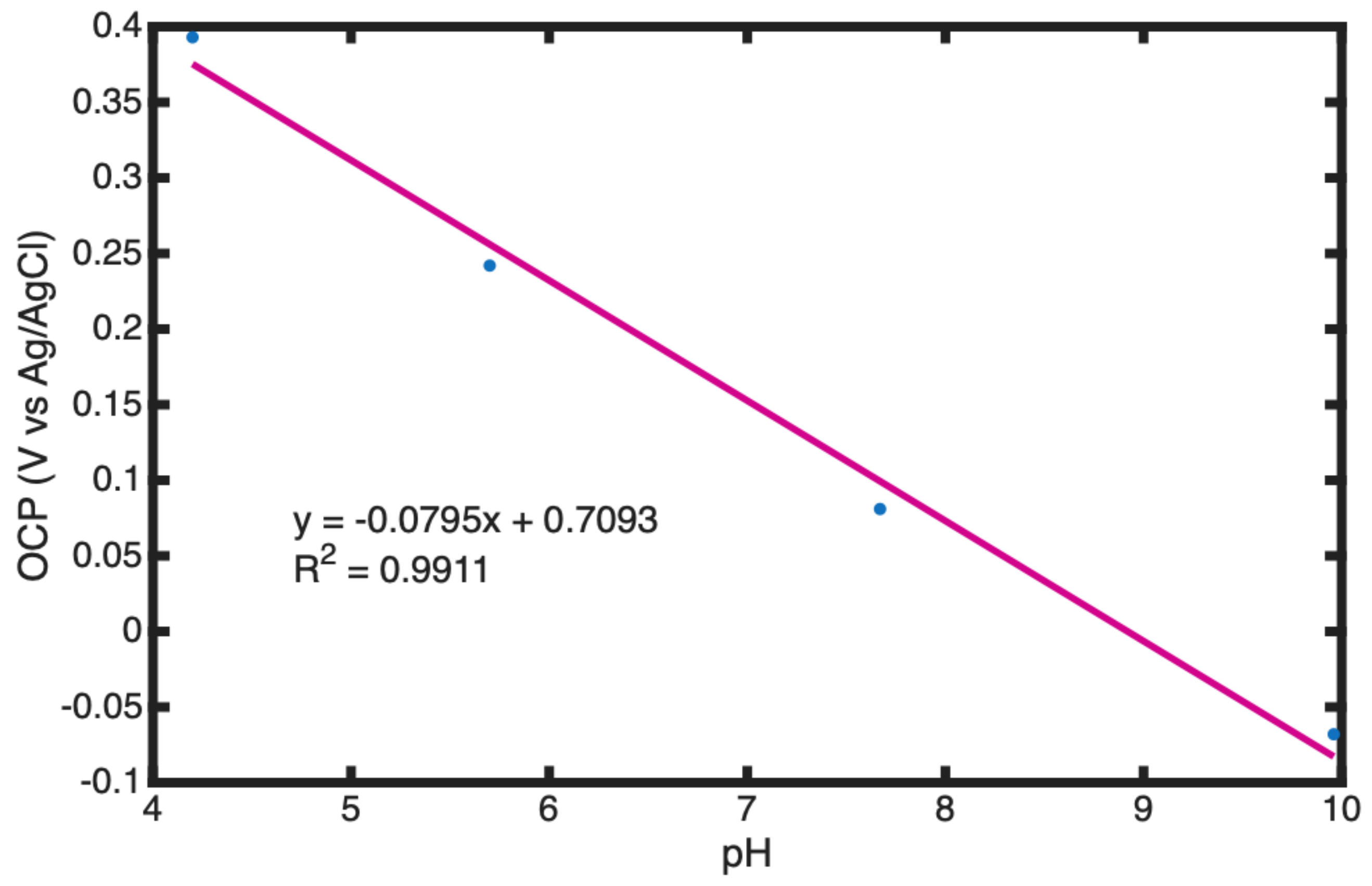
  $E_{Cell} = E^0 - (0.0592/n)\ \log Q$

- For pH-sensitive electrodes, the potential depends on the hydrogen ion concentration [H+]. Because pH is defined as –log[H+], the electrode potential changes linearly with pH. This results in a potential change of about 59mV for every 1-unit change in pH, which is known as a Nernstian response.

Step 6: Open Circuit Potential (OCP) Measured over 1000 Seconds in Solutions of Different pH

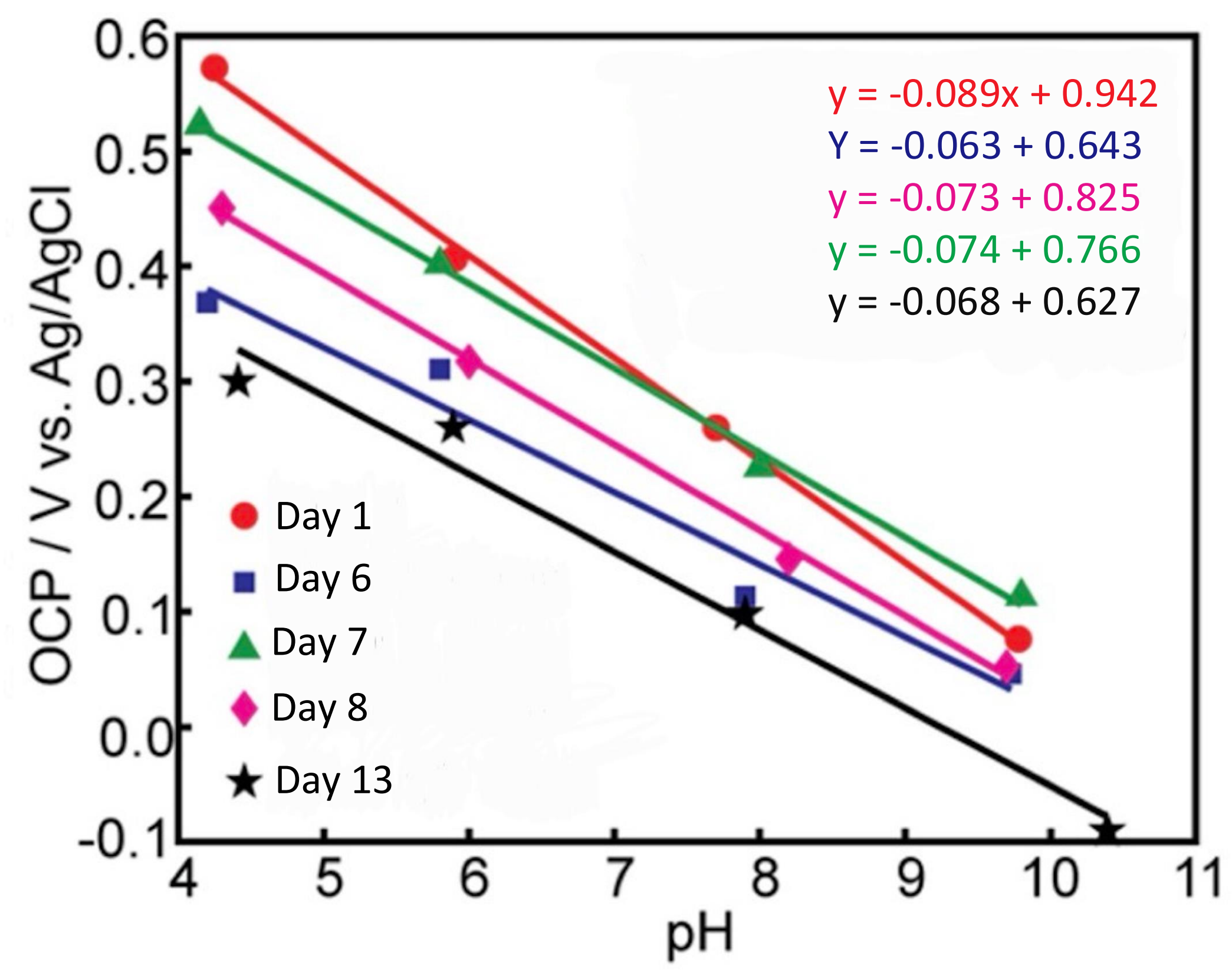


Step 7: Calibration Curve



Long-term stability

Sensor stability over time

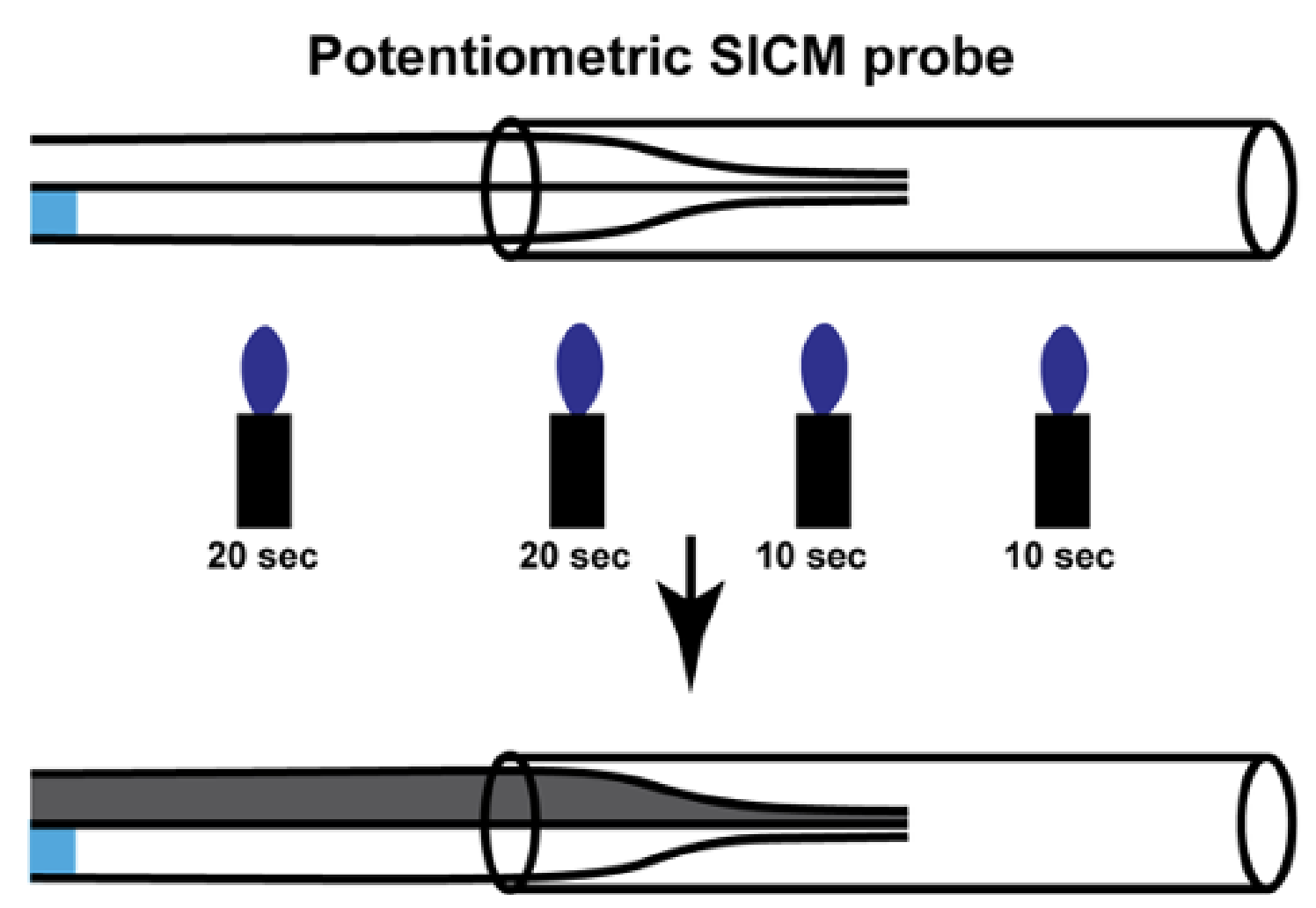


Calibration curves plotted over multiple days demonstrate that the Pt/IrOx modified electrodes maintain a consistent response over time, indicating good stability and durability of the sensor.

Conclusion

Monitoring localized pH changes is important for understanding cancer metabolism, as tumor cells create an acidic extracellular environment. Current methods such as MRI, PET-SCAN and fluorescence microscopy can provide valuable information but are limited by high cost, low spatial resolution, or phototoxicity in live-cell studies. For this project, carbon nanoelectrodes modified with platinum and iridium oxide were fabricated and characterized as nanoscale pH sensors. Platinum improves sensor durability and iridium oxide provides pH sensitivity . The sensors exhibited a near-Nernstian response and maintained stable performance over multiple days, demonstrating good sensitivity and long-term stability. This approach could provide a powerful tool for studying how local pH changes influence cancer cell behavior at the single-cell level.

Future Work



Future work will focus on integrating pH sensing with scanning ion conductance microscopy (SICM) to simultaneously measure local pH and cellular topography. A double-barrel nanopipette will be used, where one barrel modified with carbon, platinum and Iridium will function as the pH-sensing electrode, while the second barrel will be filled with KCl and an Ag/AgCl electrode to measure ionic current for SICM imaging. The combined approach would enable real-time monitoring of localized pH changes in the surface of living cells while simultaneously mapping their three-dimensional morphology.

References and Group Information

[1] B. P. Nadappuram, K. McKelvey, R. A. Botros, A. W. Colburn, and P. R. Unwin, *Anal. Chem*. **2013**, 85, 17, 8070-8074.

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