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Research Statement

1 Introduction

From a young age I have been intrigued by mathematics and felt a strong compassion for the environment. Naturally, my curiosity is piqued when the two are coupled. I began research with my advisers because of our similar interests. My chemistry adviser, Dr. Johna Leddy, develops efficient techniques for transferring electrons across various interfaces. Primarily, she applies her expertise to common energy applications such as batteries, solar cells, and capacitors. Dr. Gerhard Strohmmer, my math adviser, studies partial differential equations at a theoretical level. I use his work on theory to find numerical solutions that arise in the area studied by Dr. Leddy. In short, I work on problems motivated by chemistry, engineering, and electromagnetism using tools from partial differential equations and numerical analysis.

2 Electrochemistry Background

The fundamental process that lies at the foundation of batteries, capacitors, and solar cells is the electron transfer process. This takes place at an interface or boundary in each device and is governed by its corresponding chemical reaction. Making these devices more efficient can help decrease our negative impact on the environment. In the field of electrochemistry, the common system for testing the efficiency of this effect is known as the three electrode system, shown in Figure 1.

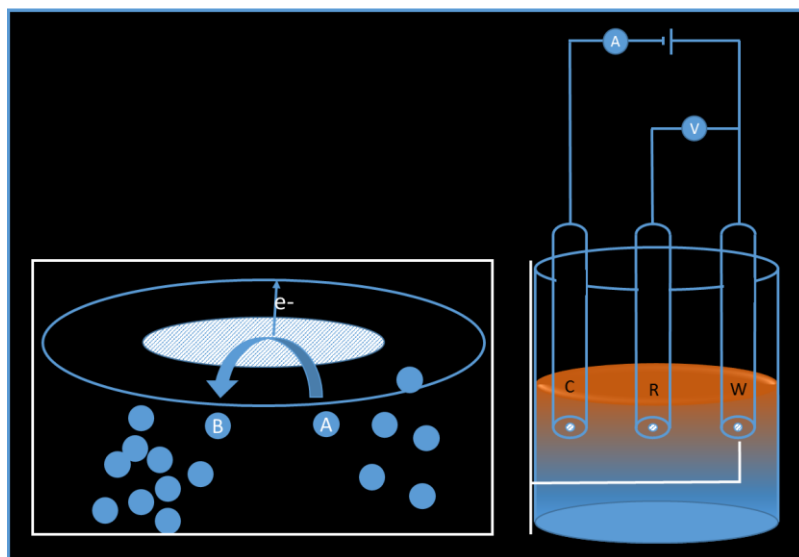


Figure 1. The molecule A (redox probe) migrates to the electrode surface and undergoes a reaction (heterogeneous electron transfer) giving off an electron. This species then becomes species B.

3 Present Work

In Dr. Leddy's Lab I have several projects. My dissertation project is my principal endeavor at this time. The research first stemmed from a chemistry experiment performed by a previous graduate student. Quite simply, the concept involves bubbles or vapor cavities. These cavities possess enormous amounts of energy (several thousand Celsius and atmospheres of pressure). When vapor cavities collapse they can be very harmful, resulting in corrosion. Accordingly, we have designed a cell that utilizes the energy

without the drawbacks. Sound waves used on a small-scale cell generate the energy in solution without the destructive process. The recent experiments show enhanced effects on the electron transfer process and cause the reaction to become more reversible. In particular, we see this electrocatalytic effect when the solution contained in the electrochemical cell (shown in Figure 2) forms a curved meniscus at the top.

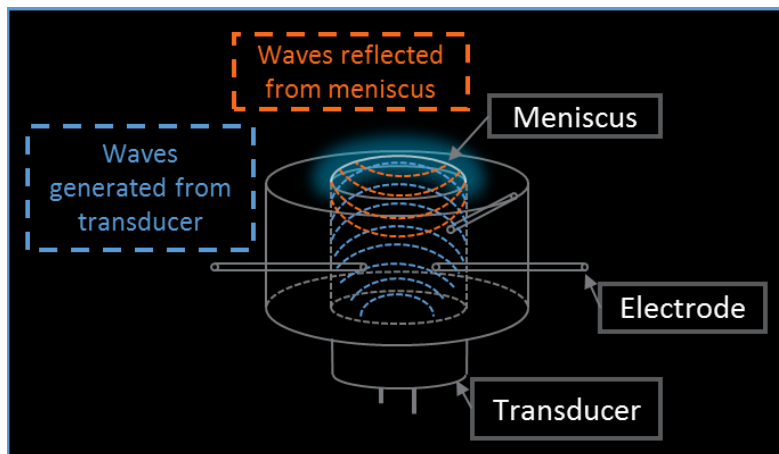


Figure 2. Diagram of electrochemical cell. The cell consists of a Teflon cylinder, transducer base, and three electrodes. Solution forms a meniscus with a height of 4mm from the top of the transducer.

The primary focus of my model is to answer what happens as the sound waves interact with the meniscus. To model this, a fluid dynamical approach is invoked using the following four equations:

$$u_t + u \cdot \nabla u + \left(\frac{1}{\rho}\right) \nabla P = 0 \quad (1)$$

$$\rho_t + \nabla \cdot (\rho u) = 0 \quad (2)$$

$$\psi_t + u \nabla \psi = 0 \quad (3)$$

$$P(\rho) = P_0 + \beta H \quad (4)$$

The first two equations are used to describe the movement of the fluid on the interior of the electrochemical cell. This is accomplished by representing the velocity as a potential flow described by the equation $u = -\nabla U$. In doing this, we gain a vector field interpretation of the movement of fluid. Equations (1) and (2), in general, relate velocity (u), acceleration (u_t), pressure (P), and density (ρ) to one another. The first equation is an Euler equation for incompressible fluids without friction in its most general form. Friction is considered negligible for this problem. This is because it introduces vorticity (local spinning of the fluid around a central point) into the problem; however for our problem the intensity of the sound waves and height of the cell dominate this kind of fluid dynamic. Equation (2), simply provides for conservation of mass in the system. Chemically or experimentally, it is clear that the catalytic phenomenon is derived from the waves and how they are affected by the top boundary. With the help of the third and fourth equations we can gain a better interpretation of what occurs at the top boundary. Equation (3) describes the boundary or surface motion of the meniscus. In this equation let ψ be the function describing the shape of the surface and let its inputs be $T_t(\tilde{x})$ and time, where T is the position of parcel \tilde{x} on the surface. One way to gain an understanding of what happens as the sound waves collide with the meniscus is to monitor the pressure on the meniscus. Sound waves can be described as the oscillation of pressure. We expect to see a change in pressure at the top boundary as the sound waves migrate through the interior and reach the meniscus. At the meniscus, the interior oscillations induce

movement in the boundary. Finally, equation (4) incorporates surface tension, β , mean curvature denoted by H (a mathematical description of the changing surface geometry), and pressure, P . Surface tension is also an important physical phenomenon to incorporate in the model because the attraction between the molecules in the liquid cause it to make the shape it forms. Governed by equation (4) this effect, in turn, has a direct impact on the pressure.

In order to develop an approximate solution to our equations we must first have a strong theoretical foundation. Combining equations (1) through (4) we arrive at boundary condition (6) involving the meniscus boundary only. We impose this boundary condition on the wave equation (5) to understand the impact of this constraint on the meniscus.

$$U_{tt} + P'(\rho_0)\Delta U = 0 \quad (5)$$

$$\frac{\partial U}{\partial n} = \frac{l}{|\nabla\psi_0|} L^{-1}(\Delta U) \quad (6)$$

Invoking principles from functional analysis, we establish that our operator (L) is elliptic and symmetric. Then using the weak formulation and the Lax-Milgram Lemma, we establish the existence of L^{-1} . Our domain of definition for the Laplacian in equation (5) involves a constraint that derives directly from conservation of mass. The establishment of these properties is important because it allows us to embed the solution space in a normed function space, in particular a Hilbert space. Listed also in equation (6) is a constant we denote by l .

Now that we have established our background we use the following numerical analysis approach to probe our solution. With the help of the variational form and the Galerkin Method an approximation converges to the solution of the wave equation using MATLAB. The resulting eigenfunctions can be interpreted physically as standing waves.

Further analysis is remains to be done. One option for further research involves the kinetic energy distribution in the cell. This is calculated using an inner product which has units of kinetic energy density. Potentially an approach based on a different physical entity, such as kinetic energy, may reveal new insight into the problem. Lastly, we may consider including small viscosity affects.

4 Past and Future Work

In the past I conducted a study on film modified electrodes. Specifically, I studied an electrode modified with inert film where a redox probe transitions from solution into film and undergoes electrolysis at the electrode surface (Figure 3). For this a convolution method was used to analyze the data.

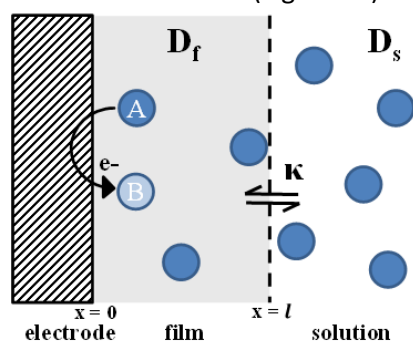


Figure 3. The molecule (redox probe) A has concentration C^* and diffusion coefficient D_s in solution. In the film, with thickness l , A has diffusion coefficient D_f and concentration characterized by the extraction parameter κ . On voltammetric perturbation, A undergoes reversible heterogeneous electron transfer at the electrode to become molecule B.

Two previous students and I developed code using MATLAB and C++ to approximate several parameters, such as film diffusion length. Convolution methods are advantageous because they can discriminate against nonfaradaic capacitance and uncompensated resistance while also allowing rapid determination of reaction kinetics. The model is independent of electrode kinetics and operates under Fick's laws of diffusion (Equations (7) and (8) for Concentration, C , Diffusion Coefficient, D , and Flux, J) to describe the current-voltage scheme and find the limiting parameters of interest.

$$J(x, t) = -D \frac{\partial C(x, t)}{\partial x} \quad (7)$$

$$\frac{\partial C(x, t)}{\partial t} = D \frac{\partial^2 C(x, t)}{\partial x^2} \quad (8)$$

Throughout the process boundary conditions specific to the film solution interface are employed and the discrete version of equation 7 and 8 are iterated in time using an explicit finite difference algorithm. Overall, the model's approximation for a particular film thickness provides an improved analysis of voltammograms for inert films on electrodes. I have a number of future projects involving this model. In particular, my chemistry adviser and I have discussed the possibility of incorporating magnetic effects into the system. This would then involve Maxwell's equations.

I had the pleasure of attending an AMS Mathematics Research Communities Workshop on Sea Ice Modeling this past summer. Several attendees and I developed a basic model that demonstrates having a height and depth dependent coefficient of drag has a significant effect on sea ice stress. Most models prior to 2010 only considered an average height and depth when calculating the drag coefficients of air and water. More stress on the ice equates to a faster rate of breakup. This has serious consequences for our environment since a faster breakup rate implies an accelerated melting rate. The current model uses the conservation of momentum equation. Included in this equation is mass and velocity of the ice (m and v , respectively), drag due to air (τ_a), water drag (τ_w), the Coriolis Effect ($mf_c k \times v$), internal stress ($\nabla\sigma$), and the tilt of the ice ($mg\nabla H$).

$$m \frac{\partial v}{\partial t} = \tau_a + \tau_w - mf_c k \times v + \nabla\sigma - mg\nabla H \quad (9)$$

The workshop was very beneficial as a significant portion of time was spent discussing ideas and questions we have yet to answer. To answer one of the questions, I have begun developing my own working sea ice model with a very eager high school student during the summer of 2015. I thoroughly enjoyed advising the student! In fact, in the spring of 2016 I plan to do a semester long research study with an undergraduate student. Overall, I look forward to continuing my research and guiding those at any level, who wish to join me.

References

- [1] Evans, Lawrence C. *Partial Differential Equations*. 2nd ed. Providence, R.I.: American Mathematical Society, 2010. Print.
- [2] Atkinson, Kendall E., and Weimin Han. *Theoretical Numerical Analysis: A Functional Analysis Framework*. 3rd ed. Dordrecht: Springer, 2009. Print.
- [3] Oprea, John. *Differential Geometry and Its Applications*. 2nd ed. Washington, D.C.: Mathematical Association of America, 2007. Print.
- [4] Bard, Allen J., and Larry R. Faulkner. *Electrochemical Methods: Fundamentals and Applications*. 2nd ed. New York: Wiley, 2001. Print.