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Research Article

Exploring transient potential steps on rotating disk electrodes with steady-state concentration profiles in single electron transfer reactions

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ABSTRACT

This study develops a theoretical model to analyze transient currents in one-electron transfer reactions over rotating disk electrodes for quasi-reversible and irreversible processes. The model employs the nearest diffusion layer approximation, separation of variables, variational iteration, and two-point pade approximation methods to derive concentration profiles. The techniques not only yield better precision and reliability of electrochemical measurements through considerations of transient currents and background current rejection in solid-state electrochemical systems, thereby improving the precision and interpretation of experiments devoted to rotating disk electrodes. Analytical expressions for the concentrations of oxidized and reduced species are obtained, enabling the examination of how parameters operate on current during the transition between activation-controlled currents. The steady-state background current has also solutions given in closed form. Lastly, we came to the conclusion that numerical simulation is employed to explain and differentiate between reversible and quasi-reversible electrode processes.

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INTRODUCTION

Electrochemical processes are essential in various fields, including environmental remediation, energy conversion, and storage. Understanding how electrochemical systems change over time is important for improving efficiency and making basic reaction mechanisms clearer. Looking at the steps that can happen on rotating disk electrodes (RDEs) gives us important new information about how electrons and mass move across electrode surfaces. We can fully

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understand single electron transfer reactions at the electrode-electrolyte interface when these studies are used with steady-state beginning concentration profiles. The present work investigates the exciting subject of transient solutions to potential steps at RDEs, shedding light on the complicated connections between reaction kinetics, diffusion, and convection. Our purpose is to clarify through theoretical studies and experimental proof. Hydrodynamic modulation (HMRDE) is often used to control the background currents on the rotating disc electrode (RDE). Research was done by Sonke and his colleagues [1, 2]. Lock-in amplifiers and a theoretical framework are usually used to deal with the system's nonlinearities. Vielstichet al. [3-6]. Conversely, we will devise a theoretical framework for prospective step techniques. The goal of this unique theory is to lessen the effects of background current. It is used in systems where electrochemical solid-state reactions have happened. Our method differs from standard methods in that it emphasizes potential stages, which offer unique advantages when applied to electrochemical systems. By giving us a better understanding of how solid-state reactions work, potential step approaches can better block the background current. Our goal is to make measurements more accurate and consistent in systems that use electrochemical solid-state processes by building a strong theoretical base for these methods. Another major goal is to develop a theory for kinetic transients at the rotating disk electrode (RDE) to study the typical characteristics of one-electron transfer processes. It is especially important to pay attention to important kinetic variables like the charge transfer coefficient and the standard rate constant (k_0). To be more specific, we are looking at a more complicated and probably more accurate method, which is to find the solution to the convection-diffusion equation in a semi-infinite interval. Chen, P. et al. [6-9]. The advanced technique offers profound insights into the kinetic behaviour at the RDE, facilitating the evaluation of electron transfer processes and their associated parameters. The simplest way to solve these transients is to use the Nernst diffuse layer model to turn the convective diffusion equation into a finite interval diffusion equation. Then, with the help of Saranya K. et al. [10-14], we find transient solutions. To do the inverse Laplace transformation and put the transformation back into the time domain, Nguye.T has to figure out the terms and development of the growth and series expansions of Laplace transformation [15-18]. We are using the Nernst diffuse layer model and the method of separation of variables together because they are both well-known ways to solve heat and diffusion equations at a short time scale [19]. This is a mix of a simple but useful way to deal with kinetic transients in electrochemical systems. In a possible step experiment without convection, quasi-reversible systems act like reversible systems when this reversible system happens eventually. Because there is no convection at first, the short-term response of a Rotating Disc Electrode (RDE) may look like a possible step at a stationary electrode. After this first kinetic transient, the RDE's surface concentration can reach a steady state. Based on what Stoll, M. D. et al. [2, 20] showed, it is likely that it is possible to guess how long the predicted transients in RDE dimensions will last. To incorporate kinetics, initial boundary value problems (IBVP) must be addressed using authentic (electrochemical) kinetic boundary conditions, specifically Robin boundary conditions. Although numerical methods have successfully addressed significantly more complex systems, it appears that neither the irreversible nor the quasi-reversible system has been subjected to analytical investigation regarding the potential step of the RDE (Balasubramanian et al. [21]). Chang emphasises the steady-state initial concentration profiles in the study of one-electron transfer. Y et al. [22-24] looked at the temporary solutions of the possible actions at rotating disc electrodes. Their study showed that steady-state concentration profiles are a good way to describe these systems and help us understand how they change over time. This gives us new information about how electron transfer processes work. Liu. H et al. looked at how rotating disc electrodes behave in steady state and how they change with temperature [25-27]. They discovered that the impact of the heat variables significantly influences steady-state behaviour, thereby improving the control and optimisation of electrochemical processes. Evanko et al. investigated the behaviour of rotating disc electrodes under conditions of exceedingly rapid comproportionating reactions [28-29]. They discovered that the limiting current ratios for two versus one electron are the same for both spinning disc electrode voltammetry and sampling current voltammetry. Examining their concentration profile helped make sense of these data and learn more about fast electrochemical reactions. Murray, R.W. et al. [30] conducted a quasi-one-dimensional steady-state investigation of multi-ion rotating disc electrode electrochemical systems. Their research, which considered homogeneous reactions, convection, migration, and diffusion, resulted in a comprehensive model that illustrates the intricate interconnections among these processes. Henderson, L. K., et al. [31] say that these research papers help us understand how spinning disc electrodes act in different situations so that we can make better predictions and take effective steps to improve electrochemical systems. Additionally, Zhu X et al. [32] have done a steady-state computational study of the current density distributions in axisymmetric systems with multi-ion electrolyte systems, focusing on the rotating disc electrode [33, 34]. The Journal of Electroanalytical Chemistry published a study by Doherty, M. F., et al. [35] that showed how useful the numerical model is for optimising rotating disc electrode applications. This is because the model does a good job of showing the current density distributions [36]. When used in many different settings, these works greatly improve our understanding and ability to predict how rotating disc electrode conduct will behave. This makes it easier to optimise electrochemical systems, according to Lopez, R. L., et al. [37]. To make sure that the RDE stays stable after each step, it is common to measure it using slow staircase linear sweep voltammetry at different rotation speeds. By changing our mathematical model, we are trying to fit and repeat these measured transient sequences. With this change, a new theory will be created that will stop at the step's steady state and start at the steady state of the applied potential. In their study, Mosayeb Gholinia et al. [38] looked into how heat moves in a two-phase rotating system with a magnetic field while nanofluid flows through it. The basic partial differential equations of mass, momentum, and energy are made easier by turning them into nonlinear ordinary differential equations. Akbari-Ganji's Method can be used to solve these equations. The Galerkin method was used by Mohammad Reza Zeinolabedini and others [39] to study the free vibrations of a rotating disc. MertolTufkec et al. [40] made a simpler model of the surge and stall when there is a distributed force on a rotor disc system. It is a basic and conceptual framework for future research in the field. [41]. After that, the two real parameters affect the properties of the modified Bernstein-type operator's approximation. Some of the immediate effects are an error estimate for functions in Lipschitz-type spaces, an error estimate for the ordinary and Ditzian-Totik modulus of smoothness, and the Voronovka-type asymptotic theorem. There is also a discussion about how close these operators can be to a Kirov and Popova type. The study wants to look at the power-law nanofluid flow of titanium dioxide on water across a small needle [42]. Also, because it was a full case study of quadratic, nonlinear, and linear radiation, heat radiation was added and looked at. This [43] study looks at how important the activation energy is for starting a chemical reaction in a two-dimensional micropolar fluid laminar flow across a semi-infinite inclined plate. When modelling how heat moves through a fluid, we use convective thermal boundary conditions and radiation.

Literature Review

Electrochemical processes are important in a lot of areas, from cleaning up the environment to changing and storing energy. It is essential to comprehend the transient behavior of electrochemical systems to maximize their efficiency and clarify basic reaction mechanisms. One way to learn more about the kinetics of electron transfer and mass transport at electrode surfaces is to investigate possible steps on rotating disk electrodes (RDEs). Benjamin Levich formulated the foundational theory that elucidates mass transport phenomena at a rotating disk electrode (RDE) at the Institute of Electrochemistry within the Academy of Sciences of the USSR. Levich articulated the theory in his seminal work, Physiochemical Hydrodynamics, which was first published in Russian in 1952. A decade later, Levich's book was translated from Russian to English, leading to the RDE gaining greater recognition among Western scholars. During the early 1960s, Stanley Bruckenstein at the University of Minnesota, along with his students Dennis Johnson and Duane Napp, collaborated with Ronnie Bell at Oxford University and his student John Albery on the study of rotating electrodes. The rotating disk electrode has become a well-known tool for studying how electrochemical reactions happen over time, building on the work of earlier generations. Laminar flow at a rotating disk electrode makes it easy for material to move from the bulk solution to the surface of the electrode. Although the bulk solution at a distance from the electrode is effectively mixed due to the convection caused by rotation, the region of the solution closer to the electrode surface tends to move in sync with the electrode's rotation. Therefore, when considering the solution from the perspective of the rotating electrode surface, it appears to be relatively motionless. These studies give a thorough understanding of single electron transfer reactions at the electrode-electrolyte interface when combined with steady-state starting concentration profiles. Electrochemical methods [44, 45] are popular chemical characterization techniques used in the chemical industry, building construction, safeguarding the environment, and damage tests. This is owing to the low cost of measurement equipment and the availability of standard processes. Aside from its potential application in chemical analysis, electrochemistry is significant in the operation of solar cells, batteries, and electrolyzes that convert electrical to chemical energy, such as hydrogen synthesis by water electrolysis or CO_2 reductive recycling. Electrochemistry is currently the most promising approach for temporarily storing renewable energy. Accurate knowledge of the mechanism of electrochemical reactions is essential for better understanding and future development of these technologies. The rotating ring-disk electrode (RRDE) is a popular and widely used technique for researching electrode response mechanisms [46-52]. Frumkin, Levich, and their coworkers came up with the first RRDE or generator-collector electrode assembly in 1959. [53]. When using an RRDE, we use forced convection caused by rotation to make sure that electroactive species created at any one potential on the disc move to the surrounding ring, where a new reaction can happen at a new potential. This changes the measured ring current. The ability to independently change the potentials of the two electrodes of the RRDE makes it a powerful tool for finding and understanding the actions of intermediates that may not be stable. RRDEs are popular for several reasons: A solid theory has existed for a long time; the RRDE provides well-regulated hydrodynamics; the mass transport rate can be easily adjusted by varying the rotation speed; the electrodes are easily cleaned and polished; and the RRDE is housed in a vessel that can be easily de-aerated, and temperature controlled. [54] To use RRDEs to identify electrode reaction products, the following approaches are most used: - The disk is kept at a constant potential, whereas the ring is kept at a variable potential where the disk reaction products are expected to transport electrons. A current-potential curve forms at the disk, while the ring potential remains constant. The disk is held at the potential where the reaction of interest occurs, and a current-potential curve is recorded at the ring to capture disk reaction products with different redox properties. The strategies outlined above share a commonality: at least one of the two electrodes must remain unaltered throughout the experiment. The potential of both electrodes should not be impeded by any theoretical constraints, enabling the researcher to modify the two independently and simultaneously. But any potentiostat, even the ones that most instrument suppliers sell, should be able to control the potential of two working electrodes at the same time with waveforms that change over time. This is usually only possible with the software that runs the instruments. Unfortunately, most of the commercial software that is available today does not support any of the processes that are not part of the standard mentioned above. The development of electrochemical measuring systems that allow for dynamic and simultaneous control of two working electrodes, particularly the dynamic control of the potential applied to the RRDE disc and ring electrodes, is a recent achievement. The mechanics of electrochemical processes have been shown to be examined through the "dual dynamic voltammetry" (DDV) technique. A rotating disc electrode (RDE) setup is a common way to test the electrochemical properties of new OER catalysts [55, 56]. The RDE is a well-known and commercially available electrochemical technique that is also experimentally complex. To determine the catalyst's activity, quantifiable metrics such as the onset potential (the ability to achieve a particular current density) are typically utilised. The principle of onset potential is not widely accepted, making it challenging to compare data from different authors regarding various materials. To address the problem of definition, there are many attempts to standardise and measure OER catalysts [57, 58-63]. There is no universally accepted method for determining comparability. Also, RDE studies are often only about materials that can be electrodeposited on the supporting electrode or powder samples. The development of the catalyst film is especially important for the second part, and even small changes in the experiment could have a big effect on how well the RDE measurements work.The rotating disk electrode (RDE) is a conventional hydrodynamic electrochemical method that employs controlled convective mass transfer. The working electrode is rotated, causing a consistent laminar flow of analyte to the active surface [64, 65]. The rates of mass transfer are not only regulated but also easily altered, allowing for a wide range of reaction time scales and the investigation of kinetic and mechanistic information [65-67]. RDEs typically consist of an insulation (PTFE) layer surrounding an active material disk (Pt, Ni, Cu, Au, Fe, Si, CdS, GaAs, glassy carbon, or graphite). Other materials have been utilised, including boron-doped diamonds [25]. Single-atom catalysts have attracted interest because they can connect homogeneous and heterogeneous catalysis in a way that is both highly active and selective [69-71]. Lu et al.'s study shows how the electronic interactions between single metal atoms and their support can be used to improve catalytic performance. This agrees with earlier studies that showed how important EMSI is in catalytic processes. For example, a study on hybrid organic-inorganic perovskites showed that adding single Pt atoms to perovskite structures can improve their catalytic properties because the metal atoms and the support material interact with each other in a way that is good for the reaction [72]. The authors come up with a new way to use the ring electrode in an RRDE setup as a "shield" for the current. Keeping the ring electrode at the same potential as the disc electrode changes the distribution of the electric field, which makes the current flow more evenly across the disc surface. This setup reduces edge effects and potential gradients that usually mess up kinetic measurements in resistive media. The study shows that this shielding method works by using both computer simulations and real-world tests to show that it does what it says it does [73, 74]. The authors probably used finite element modelling to simulate the electric field and current distribution, and then they used experimental electrochemical measurements to check their theoretical predictions. The enhanced uniformity in current distribution increases the dependability of kinetic parameters derived from methodologies such as Tafel analysis and Koutecký–Levich plots [75]. For making hydrogen in a way that is good for the environment, the Oxygen Evolution Reaction is an important part of water electrolysis. Metal-based catalysts, especially those with precious metals like Ir and Ru, have been used for a long time because they have good d-electron configurations. But because these metals are rare and expensive, we need to find effective alternatives that don't use metals. Researchers have looked into carbon materials that don't contain metals, but their catalytic activity is often limited because there aren't many good ways to change the p-band structures of non-metal atoms. This research advances prior initiatives aimed at modifying the electronic structure of electrocatalysts to enhance OER performance. Conventional methods have concentrated on doping, alloying, and generating oxygen vacancies in metal-based catalysts [76]. Many biological and chemical systems, such as photosynthesis and respiration, depend on proton-coupled electron transfer processes. It is important to know if these reactions happen in steps (sequential proton and electron transfers) or all at once (simultaneous proton and electron transfers) in order to make energy conversion systems that work well. The researchers used high-pressure pump-probe spectroscopy to study the excited-state PCET of a ruthenium complex. They could see changes in the pathways and rates of reactions by changing the pressure and concentrations of buffers or quenchers [77]. Electrochemical Impedance Spectroscopy (EIS) is a common method for studying how fast and how electrochemical reactions happen. People usually use equivalent circuit models to make sense of EIS data. These models need to make some assumptions about the system first. The DRT method provides a model-free alternative by deconvoluting impedance spectra into a distribution of relaxation times, enabling the identification of individual electrochemical processes without reliance on predefined models. The authors utilised the DRT method on EIS data derived from RDE experiments concerning the ORR in alkaline environments. They looked at the resulting impedance spectra to find different relaxation processes by changing things like oxygen saturation levels, rotation rates, and current densities [78]. The Rotating Disc Electrode (RDE) is a widely used electrochemical technique and tool for studying how fast electrochemical reactions happen when the conditions for mass transport are well-controlled. A rotating disc electrode (RDE) has a flat disc electrode (usually made of glassy carbon, platinum, or gold) that is attached to a shaft that turns. When placed in an electrolyte and spun at a controlled speed, the rotation causes a laminar flow that brings reactants to the electrode surface and keeps products from building up. The authors utilised a combination of RDE voltammetry and differential electrochemical mass spectrometry in a flow cell setup to investigate BOR on Pt/C, Au/C, and AuPt/C thin-film electrodes. This method made it possible to measure both electrochemical activity and the formation of gaseous products at the same time. This gave us information about reaction pathways and intermediate species [79]. In electrochemical studies, rotating disc electrodes are very important for controlling mass transport and studying reaction kinetics. But commercial RDE systems are often too expensive for most people, which makes them hard to get, especially in schools and labs that don't have a lot of money. There are some cheap designs, but they usually need parts that have been precision-machined, which makes things even harder. The prototype showed clear and repeatable electrochemical data, which proved that it could be used for research and teaching. Standard electrochemical experiments were used to test how well the system worked, which showed that it could be used for a variety of analytical tasks [80]. Rotating disc electrode-optical emission spectrometry (OES) is a common way to find trace elements in oils. This is important for things like diagnosing machinery wear and checking the quality of lubricants. But in most RDE-OES setups, the upper rod electrode stays still while the lower disc electrode moves. This setup causes uneven wear on the electrodes, inconsistent arc gaps, and changing spectral intensities, which makes it harder to get accurate and reproducible results. Huang et al.'s method directly addresses this issue by redesigning the electrode configuration, providing a viable solution that improves the durability of the electrodes and the dependability of the analytical outcomes [81].

The paper is given in this order: We give mathematical formulas for rotating disc electrodes with steady-state concentration profiles in single electron transfer operations in Section 2. Using the variational iteration method, Section 3 looks into the concentrations of species in analytical solutions. Section 4 talks about analytical and numerical solutions, using graphs and table values to do so. The proposed theoretical model culminates in a definitive system for examining transient currents in one-electron transfer reactions on rotating disc electrodes for substances undergoing quasi-reversible and irreversible processes, as illustrated in Section 5.

MATHEMATICAL FORMULATIONS OF THE PROB

We start with steady-state RDE concentration profiles and use the separation of variables method to solve the equations for a potential step. A diffusion equation that is limited by the Nernst diffusion layer comes close to the convection-diffusion equation. We don't consider the second layer's charging or any possible infrared losses in any situation. We also agree that the diffusion coefficients for the reduction (Red) and oxidation (OX) species are the same. The reversible scenario doesn't consider how fast the reaction really is; instead, it thinks that the system is instantly in electrochemical balance. It is helpful to discuss when reversibility is obtained within the potential step framework that is when heterogeneous kinetics cannot be detected. This will never be the case for the mathematically treated system since we can always expand it to lower times before hitting kinetic limits. Though measurements are restricted to periods after the double-layer charge has deteriorated, we are unable to sample at a rate greater than the data capture unit's maximum sampling rate. A reversible theory that has been extended to linear initial circumstances may help achieve that goal. It may also be applied to moderately fast kinetics, where surface concentration relaxation is still discernible but occurs during the phase of the reaction where convection is not yet influencing the process (Fig. 1). Strictly speaking, though, we must consider reversible and quasi-reversible systems have different steady-state surface concentrations, which prevents a one-to-one match with the potential step theory [44]. We compare the reversible and irreversible systems in the following section, which addresses this issue. The reversible situation is first. Since we begin in the steady state of a prior phase, we have a system in which the initial OX and Red concentrations are a mirror copy of one another, equipoised by their majority concentrations. This means that throughout the step, the surface concentrations of OX and Red remain constant and are equivalent to their corresponding reversible steady-state concentrations. After that, the OX and Red systems can be handled independently, and the current can be found using either gradient.

$$i = nFAD\left(\left[\frac{2(C_{s,init} - C_{s,ss})}{L}\right]\sum_{n=1}^{\infty} e^{\left(\frac{C_b - C_{s,ss}}{L}\right) - \left(\frac{n^2\pi^2 D}{L^2}t\right)}\right) \tag{1}$$

The symbol C_b denotes the total concentration of a given species in a solution. The steady-state concentration, denoted $C_{(s,ss)}$ as is, is calculated using the Nernst equation and bulk concentrations. The term $C_{(s,init)}$ refers to the species' initial concentration before any changes occur. Finally, L denotes the thickness of the Nernst diffusion layer, which is a region close to the electrode surface where diffusion dominates mass transfer [6-8].

$$L = 1.61\, D^{\left(\frac{1}{3}\right)} \omega^{-\left(\frac{1}{2}\right)} V^{\left(\frac{1}{6}\right)} \tag{2}$$

For the equation $C_{s,init} = C_b$, it reduces to the known formula with $i_{ss} = nFAD(C_b\text{-}C_{s,ss})/\mathrm{L}$:

$$R(t) = \frac{i(t)}{i_{ss}} = 1 + \sum_{n=1}^{\infty} e^{-\left(\frac{n^2\pi^2 D}{L^2} t\right)} \tag{3}$$

While formula (3) works well for long-time scales, a different series is better suited for shorter times. The Laplace transform method is used to get this alternative series, which is written as a sum of negative exponential terms. This new series is better because it converges more quickly, which means it gets to the right level of accuracy more quickly for calculations that need to be done quickly [15, 18].

$$i(t) = \left(\frac{nFAD^{\left(\frac{1}{2}\right)} C_b}{\pi^{\left(\frac{1}{2}\right)} t^{\left(\frac{1}{2}\right)}}\right) = \left[1 + 2\sum_{n=1}^{\infty} e^{-\left(\frac{n^2\pi^2 D}{L^2} t\right)}\right] \tag{4}$$

The infinite series solution shows good agreement with the results obtained from the finite element method (FEM) (Fig. 2) simulation of the convection-diffusion equation. However, a relative inaccuracy of up to 10% (Fig. 3) is observed. This level of discrepancy is significant and only occurs in short times. For intermediate time scales, the agreement between the two methods is much better. We look at reaction rates (kinetics) using fixed-rate constants (k values), but we only care about finding out how the concentration of a single species changes.

$$\left(\frac{\partial_c(x,t)}{\partial t}\right) = \left(D\frac{\partial^2 c(x,t)}{\partial x^2}\right)_{x[0,L],t\geq 0} \tag{5}$$

$$\begin{gathered} c(x, t = 0) = c_b + \left(\frac{c_s, init - c_b}{L}\right) \\ \left(D\frac{(L,t)}{dx}\right) + kc(L,t) = 0 \\ c(0,t) = c_b \end{gathered} \tag{6}$$

Using the following dimensionless variables

$$\chi = \frac{x}{L}, T = \frac{Dt}{L^2}, C_1 = \frac{c}{c_b}, L = 1.61\, D^{\frac{1}{3}} \omega^{-\left(\frac{1}{2}\right)} v^{\frac{1}{6}} \tag{7}$$

The equation (5) becomes

$$\left(\frac{\partial C_1}{\partial T}\right) = \left(\frac{\partial^2 C_1}{\partial \chi^2}\right) \tag{8}$$

The boundary conditions are

$$\begin{gathered} C_1(\chi, 0) = 1 + \alpha x \\ C_1(0, T) = 1 \\ \left(\frac{\partial C_1}{\partial \chi}\right) + bC_1 = 0\ ,\ \text{When} x = 1 \end{gathered} \tag{9}$$

Concentrations of the species in analytical solutions using variational iteration method

$$C_1(\chi, T) = (1 + \alpha x)\left[1 - 7(\alpha^2 T) + \frac{5(\alpha^2 T)^2}{2} - \frac{(\alpha^2 T)^3}{6}\right] \tag{10}$$

We can write the equation (10) as follows.

$$C_1(\chi, T) = (1 + \alpha x)\left[\sum_{n=1}^{3} 1 + b_n(\alpha^2 T)^n\right] \tag{11}$$

When,

$$b_1 = -7,\ b_2 = \frac{5}{2},\ b_3 = \frac{-1}{6} \tag{12}$$

We construct the two-point Pade approximation such that in equation (10) the coefficient of equation (11) is reproduced namely

$$C_1(\chi, T) = \left(\frac{p_0 + p_1(\alpha T) + p_2(\alpha T)^2}{1 + q_0(\alpha T) + q_1(\alpha T)^2}\right) \tag{13}$$

Where, $p_0 = 1$, $p_1 = q_0$, $p_2 = b_2 + b_1\, q_0 + q_1$

$$q_1 = \left(\frac{b_1 b_3}{b_1 b_3 - (b_2)^2}\right), q_2 = \left(\frac{(b_3)^2}{(b_2)^2 - b_1 b_3}\right) \tag{14}$$

The numerical values of p_0, p_1, p_2, q_0, q_1 are

$$\begin{gathered} p_0 = 1,\ p_1 = -7.0243, p_2 = 2.6755,\ q_0 = -0.0243, \\ q_1 = 0.0054492 \end{gathered} \tag{15}$$

Consequently,

$$C_1(\chi, T) = (e^{ax})\left[\frac{1 - 7.0243(a^2 T) + 2.6755(a^2 T)^2}{1 - 0.0243(a^2 T) + 0.0054492(a^2 T)^2}\right] \tag{16}$$

Now the current becomes.

$$j = \frac{i}{nFAk} = a\left[\frac{1 - 7.0243(a^2 T) + 2.6755(a^2 T)^2}{1 - 0.0243(a^2 T) + 0.0054492(a^2 T)^2}\right] \tag{17}$$

RESULTS AND DISCUSSION

Figure 1 illustrates the bulk concentration C_1 (χ, T) of a species vs. dimensionless distance X. In the graph, multiple lines are seen, one for each value, as indicated in the parameter that most likely determines the profile of concentration. Corresponding to different values of the parameter,

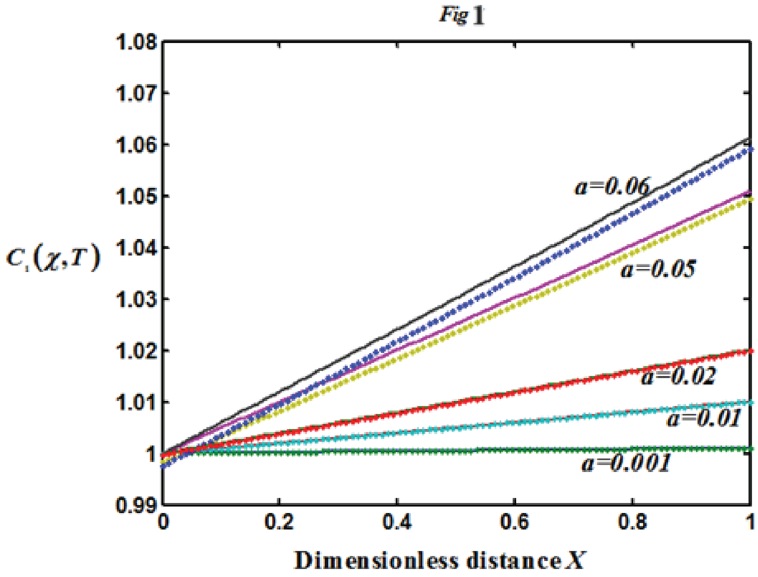

Figure 1. Comparison of concentration of C_1 for values of T= 0.1.

five values have been shown: The values displayed are 0.001, 0.01, 0.02, 0.05, and 0.06. As the value increases from 0.001 to 0.06, the slope of the lines increases. With lower values (0.001 and 0.01), the concentration profiles straighten out even more, showing a nearly horizontal line on the y-axis, which means there are minimal changes in the sum of concentrations along the distance (on the X). The concentration profiles become more sloped as X goes down. This means that changes in concentration with distance are bigger for higher amounts of the gold label (0.05 and 0.06). This concentration changes more over the two-dimensional distance X as the value goes up. This makes sense from a physical point of view, as it could be related to a material property (reaction rate, diffusion coefficient).

The analytical solution is depicted by the dotted lines, while the numerical solution is shown by the solid lines. The concentration of a certain component as a function of dimensionless distance (X) is given in Figure 2. There are several lines in the graph, each equivalent to a different value of the dimensionless time T. If we increase T from 5 to 12,

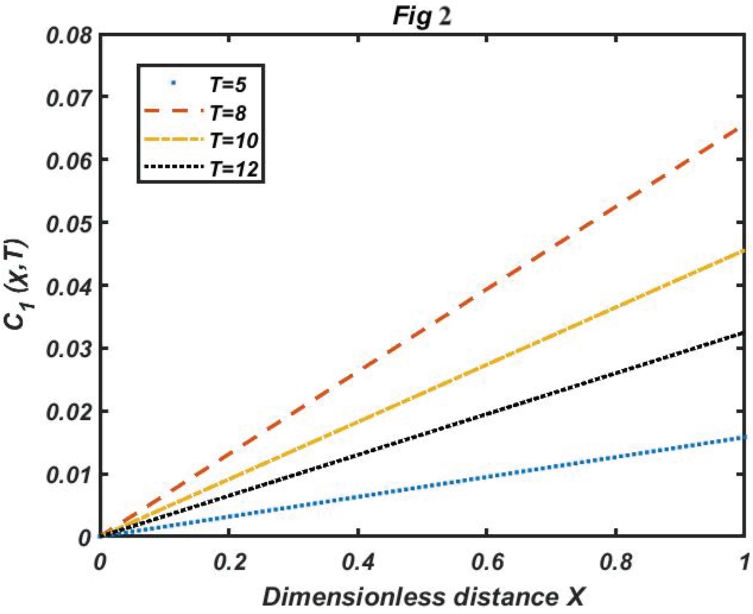

Figure 2. Plot of concentration profiles of C_1 versus dimensionless distance X calculated using equation (16) for values a = 0.001 and various values of time parameter T in linear transition.

the slope of the lines will get larger. Then the concentration profile is flat, as it indicates that the concentration changes gradually with distance for lower values of T (e.g., 5). The other consequence of the profile represented in the graph at higher values of T (at T = 12) is still a steeper profile, which indicates greater variation of concentration with distance. Larger values of T due to a wider change in concentration over the dimensionless distance X indicate higher progress concerning time towards the reaction or process. It shows how the concentration of species changes with distance and time and how these levels help in understanding the behavior of the system under different circumstances.

The concentration of a species C_1 (χ, T) vs. dimensionless distance X is shown in Figure 3. For each dimensionless time T, the graph has several lines. The concentration profiles behave very differently as T passes from 5 to 12. In this case of T = 5, the concentration decreases slightly from around units and rises continuously across the dimensionless distance. The concentration is less sensitive to X near the center of the model, but for high values of T it rises more sharply with X. For T = 8, T = 10, and T = 12, the corresponding lines show increasing concentrations. When the high values of T are taken, as shown in, it results in higher concentrations over the dimensionless distance X, therefore the process or reaction occurs more with friction effects inside.

N - Numerical solutions, A – Analysis, E – Errors.

Figure 4 shows a few examples comparing linear transition to logarithmic transition; In this figure, two transitions are plotted using the dimensionless distance X on the x-axis and the y-axis. Table 1 provides a clear representation of the numerical value of Figure 4. There is one more with an arc, a logarithmic transition T = 5. The figure below suggests that the logarithmic transition curve is steeper than the linear transition. This indicates that the y value changes more rapidly for the logarithmic transition as compared to the linear transition. In a linear transition, the Y value varies equally while the change in the X axis, which means it will either

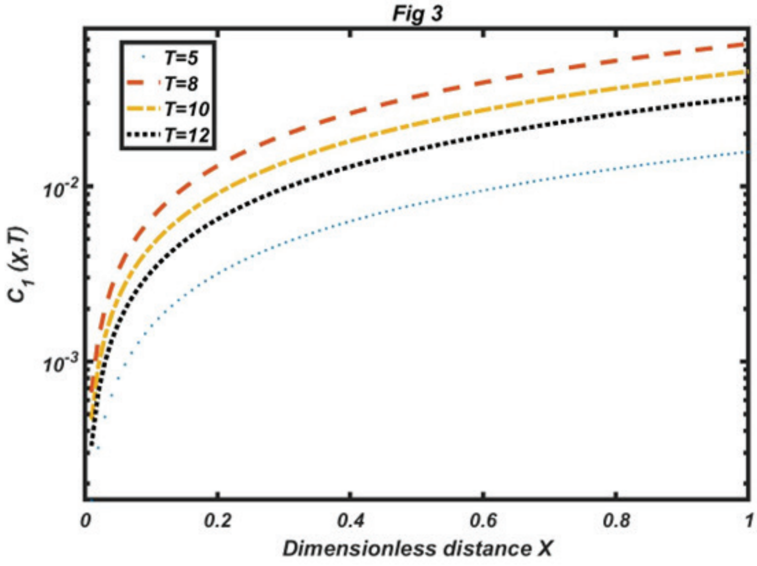

Figure 3. This plot shows the concentration profiles with dimensionless distance (x) calculated using equation (16) focuses on a fixed value of a = 0.001 and explores various time constants (T) displayed in log transition.

Table 1. Shows the concentration profiles of a C_1 on a fixed value of a = 0.001 with variousvalues of time constants (T)

X	T = 0.1; a = 0.01			T = 0.1; a = 0.02			T = 0.1; a = 0.05			T = 0.1; a = 0.06		
	N	A	E	N	A	E	N	A	E	N	A	E
0	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000
0.2	0.003195	0.003001	0.0001	0.1326	0.1253	0.0073	0.009213	0.009110	0.0001	0.006568	0.006542	0.0000
0.4	0.00639	0.0062	0.0001	0.02652	0.02531	0.0012	0.01843	0.01721	0.0012	0.01314	0.01302	0.0001
0.6	0.009585	0.00955	0.0000	0.03845	0.03822	0.0002	0.02718	0.02617	0.0010	0.0197	0.0184	0.0013
0.8	0.001262	0.00115	0.0001	0.05237	0.05212	0.0002	0.03639	0.03625	0.0001	0.02594	0.02551	0.0004
1	1	1	0	1	1	0	1	1	0	1	1	0

Fig 4

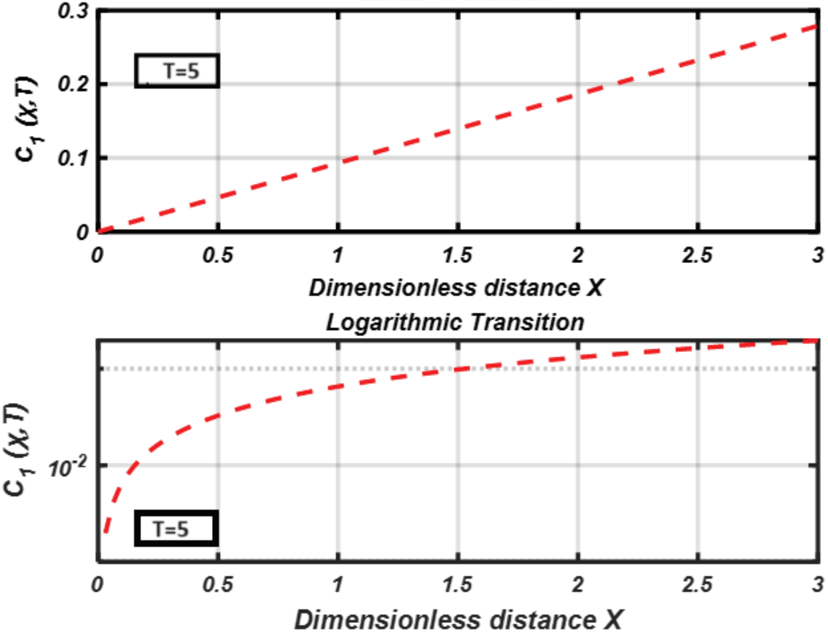

Figure 4. Represents Linear transition and Logarithmic transition at T=5 for concentration profiles C_1 versus dimensionless distance X.

steadily increase or decrease based on what this variable is. The linear one is kind of a steady change; the other one is more like a sudden change followed by a longer phase. The graph illustrates that when the two transitions are done over the same distance X, a logarithmic transition results in a faster and more drastic change of the represented variable Y compared to a linear transition. In other words, the logarithmic transition takes less distance to get to the final state 0.2 more than the linear transition takes distance to get to 0.1.

Figure 5 shows the relationship between dimensionless distance and a variable labeled C_1 (χ, T). All curves in the graph portray a decreasing pattern as dimensionless distance X rises from 0 to 0.9. The decrease level is influenced through “a”; that is, higher values of 0.05 and 0.06 compared to 0.03 and 0.04 depict a faster decrease in C_1. This is shown in the graph, indicating a significant effect of the parameter “***a***” on how C_1 (χ, T) behaves. This serves as a constraint on the rate of change. The greater the value of “a” causes thesteeperthedrop-off in C_1 (χ, T). To achieve a greater x change or higher values of C_1 (χ, T). They are reached more quickly when x increases as “*a*” increase.

Fig 5

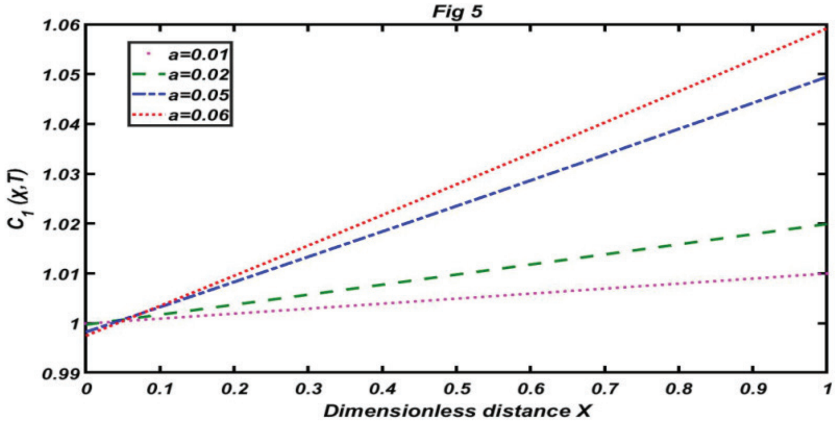

Figure 5. This plot illustrates the concentration profiles of C_1 at a fixed time constant (T = 0.1) against dimensionless distance (x), calculated using equation (16) that explores how the concentration profiles change for various values of parameter ‘a’ on a linear transition.

Table 2. Shows the concentration profiles of C_1 at a fixed time constant (T = 0.1) for various values of parameter ‘*a*’

X	T = 0.1; α = 0.01			T = 0.1; α = 0.02			T = 0.1; α = 0.05			T = 0.1; α = 0.06		
	N	A	E	N	A	E	N	A	E	N	A	E
0	0.9975	0.9952	0.0023	0.9999	0.9975	0.0024	0.9983	0.9932	0.0051	0.9981	0.9970	0.0011
0.2	1.002	1.000	0.002	1.004	1.001	0.003	1.008	1.002	0.006	1.01	1.0000	0.01
0.4	1.004	1.001	0.003	1.008	1.003	0.005	1.019	1.012	0.007	1.021	1.0010	0.0200
0.6	1.006	1.002	0.004	1.012	1.010	0.002	1.028	1.013	0.015	1.034	1.0222	0.012
0.8	1.008	1.004	0.004	1.016	1.011	0.005	1.039	1.015	0.024	1.048	1.034	0.014
1	1	1	0	1	1	0	1	1	0	1	1	0

N - Numerical solutions, A – Analysis, E – Errors.

Figure 6 showsthatthe curves all start from y = 1.05 and decrease with increasing x. For α = 0.05 and α = 0.06, the curves fall off more rapidly than for α = 0.01 and α = 0.02 (figure). The figure shows that the parameter ‘a’ determines how rapidly it decreases concerning the dimensionless distance. Table 2 provides a clear representation of the numerical value of Figure 5. Larger “a” values result in the curve decreasing faster. This illustrates that the variable greatly influences the functionality of the error function. It is stiff on the rate of (error concerning distance). The steepness of the decrease of error for a fixed increment dimensionless distance X and logarithmic scale as a function of “*a* “ values. That is that the error “e” decreases faster to 0 as a tends towards infinity exponentially.

Figure 7: The linear transition (Fig. 4) exhibits a constant rate of change. It is a straight line as the change in y value is the same for each consecutive x. These logarithmic transitions consist of a greater initial decrease and then a slower decrease at the end. And a curved line that goes from much steeper to flatter. In the figure, the curve of the logarithmic transition is steeper than the linear transition. This means that the y value increases faster for a logarithmic transition compared to a linear one. It communicates the differing effects of linear and logarithmic onsets impressively, even portraying the specific move faster and more abruptly, a logarithmic onset is compared to a linear gamma.

The variation of a parameter ‘*a*’ on C_1 (χ, T) is illustrated in Figure 8. The bottom boundary curve of all curves decreases with the dimensionless distance X by 0.9, and the increase is far less than the decrease, while the other two sides also. Fewer changes have taken place in decreasing or changing trends. The rate of decrease is determined by the value of “***a***”. Higher centrality of the source hubs: Larger values of ‘*a*’ (0.05 and 0.06) decrease fast. Larger “*a*” values lead to a faster decay of the width growing in dimensionless distance X, i.e., a higher rate at which C_1 (χ, T) approach lower numerical results as the X increases.

Figure 9 illustrates how applying a parameter ‘a’ would influence some function, which presumably can be an error in C_1. We note that, on a logarithmic scale, the dimensionless distance X is increased from 0.01 to 10; all curves illustrate a descending trend. The decrease rate of “C_1” is dependent on the value of “*a*”, where higher values (0.05 and 0.06) cause faster mage, compared to lower ones (0.01 and 0.02). Larger values of “a” give a greater drop in error

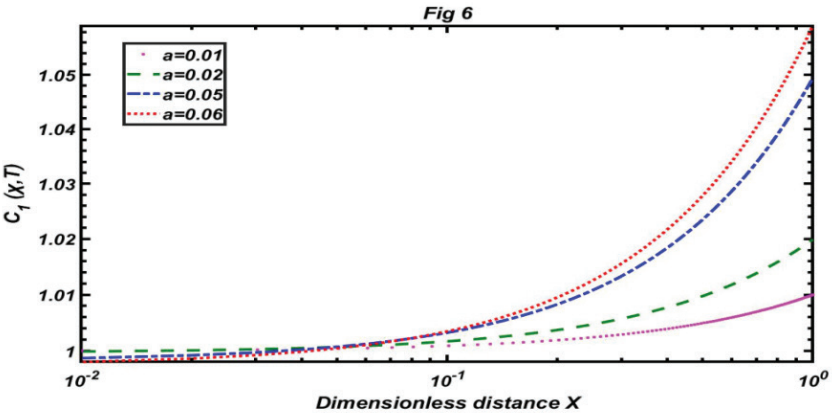

Figure 6. This plot illustrates the concentration profiles C_1 at a fixed time constant (T = 0.1) is plotted against dimensionless distance (x), calculated using equation (16) that explores how the concentration profiles change for various values of parameter ‘a’ on a log transition.

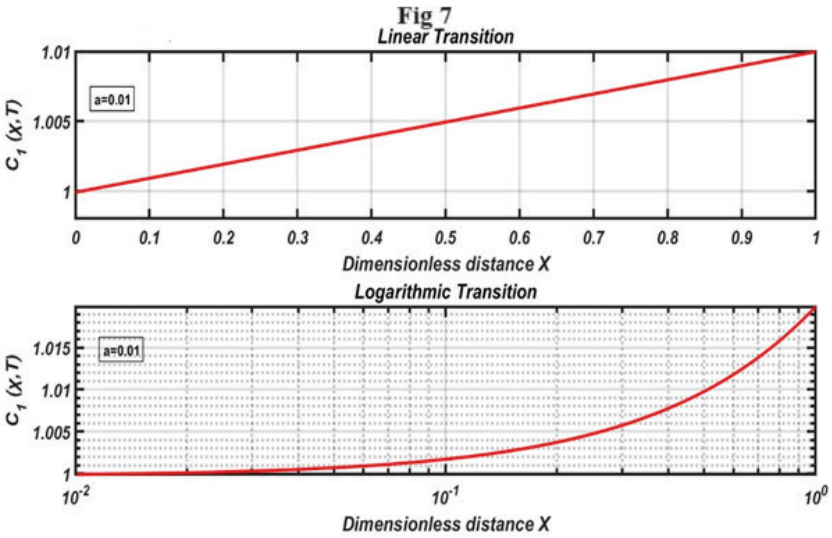

Figure 7. Represents Linear transition and Logarithmic transition at a = 0.01 for concentration profiles C_1 versus dimensionless distance X.

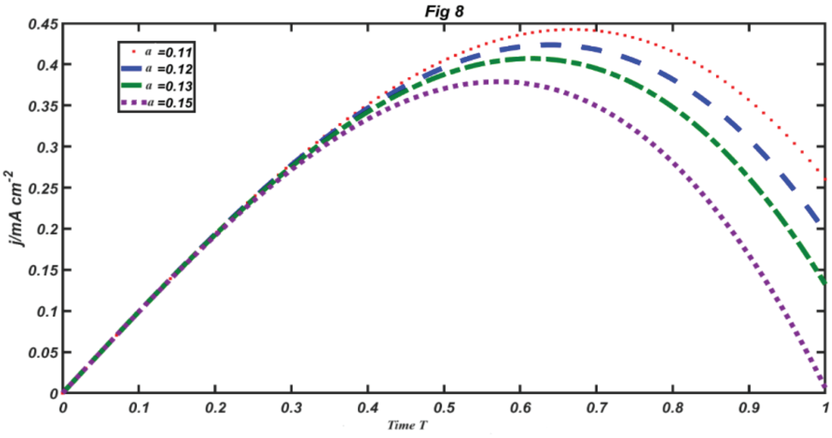

Figure 8. This plot visualizes the concentration profiles of C_1 with dimensionless distance 'a' calculated using equation (17) andexplores the concentration profiles that evolve for various time constants (T) displayed on a linear transition.

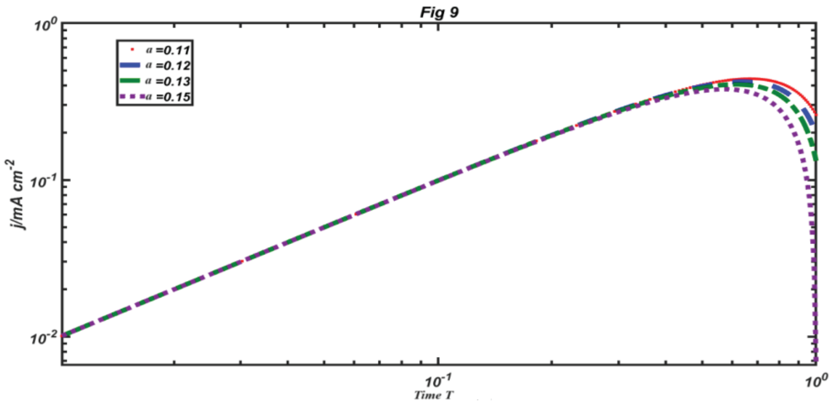

Figure 9. This plot visualizes the concentration profiles of C_1 with dimensionless distance 'a' calculated using equation (17), and explores the concentration profiles that evolve for various time constants (T) displayed on a log transition.

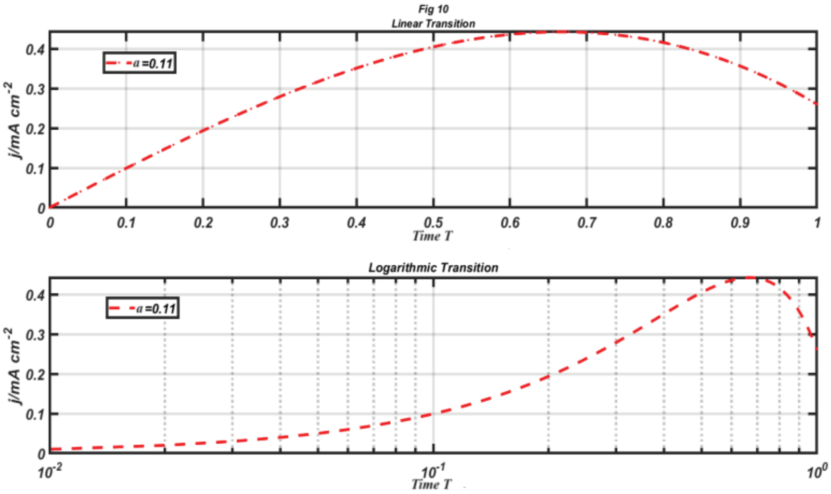

Figure 10. Represents Linear transition and Logarithmic transition at T = 0.11 for concentration profiles of C_1 versus dimensionless distance$a$.

per change in dimensionless distance (X) on a logarithmic scale. That is, the error "a" decay more quickly with distance since "a" increases exponentially.

In Figure 10, you can see the current density vs. time for both linear and logarithmic transitions on an electrode system with a rotating disc. The figures show the RDE under linear and logarithmic coverage changes. A graph of a straight line through time shows that j first rises single exponentially, reaches a peak at T=0.5, and then slowly falls back down. This suggests that the states were moved through progressively slower processes. Plotting the same values on a long-time scale graph, as shown in Figure 4, helps clarify what occurs at early times, i.e., there is an initial small (j) that grows more pronounced with time, reaching a peak value before decreasing again. The parameter is probably related to the rate or capacity of this process, which influences the temporal pattern observed. Overall, the study gives a more complete picture of how time works in this dramatically changing situation because it looks at both linear (short hours-days) and logarithmic (long days-month-year) time scales, which are important for understanding how things change quickly.

This study develops a theoretical model for an electrochemical reaction taking place on a rotating disc electrode (RDE). The model focuses on single-electron irreversible and quasi-reversible electron transfer processes. This method relies heavily on both the separation of variables and the closest diffusion layer estimate. Using the variational iteration method and the two-point Pade approximation, we can find analytical equations for the concentrations of OX and Red. The steady-state background current is also calculated analytically from the first transient, which leads to the closed-form answer. The study also looks at how different factors affect the current. It focuses on the one that has the biggest effect on the current expression during the transition period that is controlled by flow. If we add linear initial conditions to the current theory, it might work better for reactions that can't be stopped and start with a constant concentration. This kind of addition would be especially useful when reaction times are short. The model might be able to show the short changes in concentration at the electrode surface (called "relaxation") before the steady state in this case. But it's very important that this relaxation happen in a time period that isn't affected by convection. Adding linear initial conditions to the theoretical framework would make it more accurate in situations where the initial concentrations aren't all the same but change in a straight line across the system. This is especially important when the starting concentration is different because of the type of material or how it was handled before. Fast-kinetic reactions happen quickly, so you can see changes in surface concentration before convection or other outside factors become important. This makes it easier to understand how the reaction works on its own. So, a more complete theory that takes into account linear starting conditions could make analyses and predictions in these kinds of systems more accurate. Adding linear starting conditions to the theoretical model would make it easier for researchers to study processes that can't be undone and have steady-state initial concentrations. This is especially true when the kinetics are fast and the convection effects are small.

SOME PRACTICAL UTILIZATION OF RDE RESEARCH

Rotating disk electrodes (RDEs) are commonly employed in electrochemistry for a variety of purposes. Here are a few significant applications:

Electrocatalysis: RDEs are used to study electrocatalytic reactions like oxygen reduction reaction (ORR) and hydrogen evolution reaction (HER). Researchers can test the efficiency and kinetics of catalysts by varying mass transfer to the electrodes.

Corrosion Studies: **RDEs** help to investigate the corrosion characteristics of metals and alloys. Analyzing current responses to different potentials allows one to figure out the susceptibility of materials to corrosion in a variety of conditions.

Fuel Cells: **RDEs** are employed in power cell studies to assess catalyst effectiveness during electrochemical reactions (for example, methanol oxidation or hydrogen oxidation), which helps to optimize catalyst compositions for improved fuel cell efficiency.

Battery Research: RDEs are used to study the motion of reactions between electrodes in batteries, namely lithium-ion and metal-air batteries. They offer details on charge transfer mechanisms and mass transportation limits.

Analytical Chemistry: RDEs are used to identify electroactive compounds in complicated combinations. The regulated atmosphere enables precise current measurements, which aid in the detection of substances at low quantities.

Environmental Monitoring: RDEs can be used to detect pollutants in soil as well as water. They help to determine the electrochemical reactions of contaminants, which allows for the development of environmental surveillance equipment.

Research in Fundamental Electrochemistry: RDEs are utilized to study essential electrochemical reactions such as reaction kinetics, mass transport, and species adsorption on electrode surfaces.

Surface Science: They are employed in surface studies to investigate the adsorption and desorption of molecules on electrode surfaces, revealing information on surface alteration and the functionalization process.

Rotating disk electrodes play a vital role in basic and applied electrochemistry research. Their ability to supervise mass transit and measure current responses makes them helpful in a wide range of fields, including catalysis and environmental studies.

CONCLUSION

We utilized the separation of variables method to resolve a finite Nernst layer approximated diffusion equation accompanied by boundary conditions. This lets us find the transients of reactions that can't be undone and reactions that can be undone almost completely at a rotating disc electrode for one-electron transfer systems. This method lets you get kinetic data from both transients and steady-state values. You can look at each possible step on its own if you use quasi-steady-state step methods at the RDE. The proposed theoretical model elucidates transient currents in one-electron transfer reactions for substances occurring in both semi-reversible and irreversible manners on rotating disc electrodes. The model significantly enhances our comprehension of transition time and the factors that affect current behaviour by offering analytical formulas for the concentrations of oxidised and reduced species, in addition to delivering a closed-form solution for the steady-state background current. However, this method does not address the issue of obtaining precise transient solutions at the RDE in analytical form. It can be shown that the initial transient can be written in a closed form after that, even when starting with steady-state concentrations and a system where the diffusion coefficients aren't all the same. Background currents don't change the current transients, but they do change the total steady-state current. This makes it easy to find the background current, which is the difference between the total steady-state current and the expected steady-state current (which is based on the transient). This method makes electrochemical measurements more accurate and reliable, especially in solid-state electrochemical reaction systems. This tool makes it easier to design experiments that use rotating disc electrodes. Reversible conditions naturally assume that electrochemical balance happens instantly, and they tend to be different from the rates of reactions that happen. Finding the exact point where reversibility starts is very important, and potential step analyses can't show heterogeneous kinetic processes. If you use the separation of variables method to solve the convection-diffusion problem on a limited interval, you might be able to get an exact answer. Future research will deal with this problem. The steps outlined herein may be applicable to other electrochemical systems exhibiting a (quasi) steady state, such as hemispherical microelectrodes, thin layer cells, and various convective systems. The model can be used for shorter amounts of time, which might help with the problems that come with slow reaction kinetics.

AUTHORSHIP CONTRIBUTIONS

V. Sreelatha Devi: Writing- Original draft preparation, Investigation and Editing. Vijayabalan D, Kavitha A: Conceptualization, Visualization. Loganathan T: Methodology, Editing. Manimuthukumar R & Suresh M L: Supervision, Validation, and Reviewing

DATA AVAILABILITY STATEMENT

The authors confirm that the data that supports the findings of this study are available within the article. Raw data that support the finding of this study are available from the corresponding author, upon reasonable request.

CONFLICT OF INTEREST

The author declared no potential conflicts of interest with respect to the research, authorship, and/or publication of this article.

ETHICS

There are no ethical issues with the publication of this manuscript.

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