Numerical Modeling of Hydrogen Gas Bubble Dynamics in an Alkaline Water Electrolysis Process Using COMSOL Software

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ABSTRACT

During this investigation, we utilized the COMSOL software to simulate the motion of a hydrogen bubble in water. The simulations were executed within COMSOL, employing a direct numerical solver and electrochemistry models. We carefully chose an appropriate method for capturing the interface to monitor the evolving surface of a growing bubble. Among the options—level set, phase field, and moving mesh interface capturing methods—the study concluded that the moving mesh framework is optimal for simulating bubble growth. Additionally, the study emphasizes that effectively harnessing hydrogen as a transitional energy indicator could pave the way for alternative solutions, particularly in light of diminishing petroleum reserves, assuming a proficient mastery of production technologies.

I. Introduction

The escalating demand for energy, driven by global development and population growth, is predominantly fulfilled by fossil fuels (oil, natural gas, and coal), accounting for approximately 80% of the total [1]. However, the combustion of these fuels releases greenhouse gases like carbon dioxide (CO2) and methane (CH4), directly contributing to the adverse effects of global warming. Moreover, the pollution resulting from fossil fuel usage has detrimental effects on human health and various forms of life in the environment. Compounding the issue, the natural reserves of these resources are finite. In light of these challenges, there is a mounting need for renewable energy sources.

With the rise of weather-dependent energy sources, energy peaks during periods of high production are becoming increasingly frequent. To balance these peaks, electrolysis can be used to convert energy into storable gases. With the growing interest in electrolysis, more and more research is being conducted in this field. While numerous experimental studies have been carried out on bubble growth, fewer numerical studies have been conducted. The work by K. Torii et al. [2] describes a study on the numerical modeling of hydrogen gas bubble dynamics in an alkaline water electrolysis process. The authors employed a three-dimensional coupled numerical simulation method to model the biphasic flow and electrochemical phenomena in alkaline water electrolysis. The results indicated that the dynamics of the hydrogen gas bubble are influenced by parameters such as hydrogen production rate, electric current density, and OH- ion concentration. The authors also observed that the behavior of the hydrogen gas bubble can change depending on the operational conditions of the electrolyzer. A physical modeling of alkaline electrolysis is established, and studies on the two-phase flow model are conducted for this model by [3]. Both internal and external forces acting on the resulting bubbles are determined. The authors

employed empirical relationships to analyze the effects of various factors such as bubble rise rate, mass flux, quality, fluid velocity, and wetting angle. It's demonstrated that the rise velocity depends on the hydrogen and electrolyte void fraction and hydrogen density. The separation of gaseous hydrogen from the electrode, the velocity of the gaseous hydrogen bubble under the influence of drag and buoyancy forces, is also calculated based on the drift coefficient in this modeling. The outcomes can contribute to comprehending the evolution mechanism of hydrogen bubbles in alkaline water electrolysis. Additionally, it's indicated that it wasn't possible to observe the annular flow regime as there wouldn't be sufficient electrolysis tank length and fluid velocity in the investigation.

Koji Matsuura et al. [4] discusses the formation of hydrogen bubble plumes during electrolysis, generated from the cathode. The mechanism behind the plume formation is examined for its role in controlling bubble diameters and alignment within the solvent. The study was conducted by observing the generation of hydrogen and oxygen bubbles from electrodes in tap water and a saline solution, using a high-speed microscope. Changes in bubble diameters were analyzed, and the growth kinetics of hydrogen and oxygen bubbles were compared. The results demonstrated that plume formation primarily depended on the ionic strength of the water rather than the electrical voltage. The relationship between hydrogen bubble diameter and distance from the cathode was explored, revealing a correlation in tap water but not in the saline solution.

The study of F.J. Higuera [5] examines the growth of bubbles attached to an electrode surface during the electrochemical production of hydrogen. The investigation is conducted using a horizontal cathode setup submerged in a calm, diluted aqueous solution. A simplified model incorporating the Butler-Volmer reaction mechanism, electroactive species diffusion and migration, and a symmetry condition approximating periodically spaced bubbles on the electrode is employed for analysis. The diffusion-controlled growth of bubbles follows a t1/2 growth law with slight deviations due to uneven dissolved hydrogen concentrations in the surrounding supersaturated solution. As bubble spacing decreases, growth trends align with a t1/3 law. In an alkaline solution and when water consumption in the reaction is disregarded, the spatial and temporal average current density experiences an exponential increase with applied voltage. Conversely, an acidic solution results in an average current density plateau influenced by transport limitations, dependent on bubble spacing. The presence of attached bubbles enhances average current density at a given voltage due to reduced concentration overpotential. Andrea Angulo et al. [6] examine the impact of bubbles on electrochemical systems, highlighting the intricate connections between bubble evolution processes and electrochemical phenomena. The article explores how bubbles influence the energy efficiency of electrochemical processes by affecting activation, ohmic, and concentration overpotentials. It also presents strategies to mitigate losses caused by bubbles and to harness their presence for enhancing electrochemical reactions. This analysis aims to identify opportunities for future research in this pivotal field. The study reported by Akash Raman et al. [7] investigates the impact of hydrogen bubble growth on the half-cell potential in alkaline water electrolysis under constant current conditions. It reveals that while larger bubbles and higher currents enhance the efficiency of gas absorption and concentration reduction, these effects are eventually outweighed by the Ohmic shielding effect as bubble size increases. The study sheds light on the intricate interplay between bubble dynamics and electrochemical processes. Bashkatov et al. [8] investigates the dynamics of single hydrogen bubbles generated in acidic electrolytes under potentiostatic conditions using a Pt microelectrode during parabolic flights. It identifies three bubble evolution scenarios, with a focus on the dominant scenario involving lateral bubble detachment. The study comprehensively analyzes bubble behavior in terms of radius, current, trajectories, and lifetime, particularly addressing bubble-bubble coalescence events. Moreover, a detailed comparison of bubble phenomena under different gravity levels is presented.

Our work aims to create accurate models in COMSOL Multiphysics that simulate the growth of an electro-generated bubble on electrodes. The simulations were conducted in COMSOL using a direct numerical solver and electrochemistry models. An appropriate interface capturing method for tracking the evolving interface of a growing bubble was selected. The moving mesh framework was concluded to be the best for bubble growth simulations among the level set, phase field, and moving mesh interface capturing methods. One key area where improvements can be made is at the electrode where bubble generation occurs. The electrode can optimally produce gas when no bubble is present, which is not possible as bubbles form due to gas generation. However, it is possible to reduce the time the bubble remains on the electrode, and this can be achieved in two ways. The bubble's growth can be accelerated so that it reaches the critical detachment radius more quickly, or the critical detachment radius can be reduced. This simulation focuses on the first method, namely the analysis of bubble growth to better understand how efficiency can be enhanced by reducing the growth time. For this analysis, only singular bubbles are required, which is why microelectrodes are often used in experiments. Microelectrodes have the advantage of generating a single bubble that goes through the following phases: nucleation, growth, and

detachment. Studying a single bubble is highly advantageous as it is much simpler to track, and it is free from any coalescence events that can occur with larger electrodes.

II. Case Definition

The size of a spherical bubble is ensured through the determination of Eötvös and Morton numbers, while the Reynolds number is employed to ascertain drag. In the case of a hydrogen bubble in water, material properties are specified, resulting in a Morton number of approximately $M_o = 2.6 \times 10^{11}$. Unlike the Morton number, the Eötvös number incorporates not only material parameters but also a characteristic length, which, in this instance, is the bubble diameter. To maintain spherical symmetry, the Eötvös number must be less than $E_o = 0.3$, corresponding to a bubble diameter of $d_b = 1.5$ mm. For safety considerations, a bubble diameter of $d_b = 0.3$ mm is chosen, aligning with an Eötvös number of $E_o = 0.0121$. To streamline computational intensity, the simulation geometry is configured in 2D with axial symmetry, as depicted in Figure 1 of the COMSOL Multiphysics illustration.

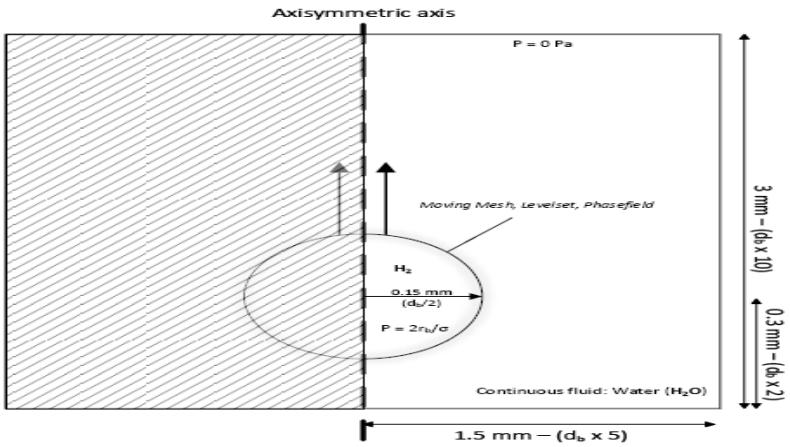

Figure 1. Schematic view of the simulated case, illustrating the most important features and parameters; dimensions are not to scale.

As a rising bubble swiftly attains its terminal velocity, the domain need not extend significantly in the z dimension. However, to mitigate potential wall effects induced by the ascending bubble, the r dimension is maintained at a sufficiently large scale. Gravity plays a pivotal role in creating a pressure disparity on the bubble, propelling its upward movement. To establish the simulation end time, an estimation of the terminal velocity is essential. This approximation can be accomplished through two methods. The initial method entails extrapolating the Reynolds number from Figure 2, though it may not yield highly precise results.

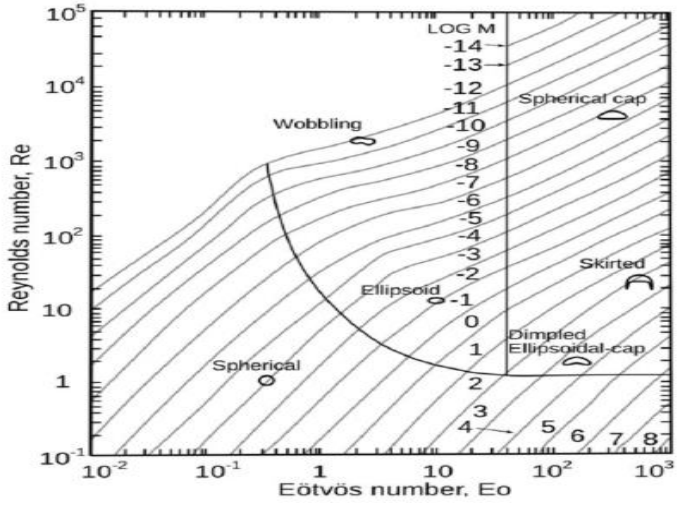

Figure 2. The Grace diagram presents various regimes of ascending bubble shapes based on Reynolds, Eötvös, and Morton numbers [9].

The second method involves using the Hadamard-Rybczynski equation [9] for terminal velocity, defined as follows:

$$W_b = \frac{2}{3} r_b^2 \, g \, \frac{(\rho_b - \rho_f)}{u_f} \, \frac{\mu_f + \mu_b}{2\,\mu_f + 3\,\mu_b} \quad (1)$$

Where W_b is the resulting bubble velocity, r_b is the bubble radius, g is the gravitational acceleration, ρ_b is the density of the bubble fluid, ρ_f is the density of the continuous fluid, μf is the dynamic viscosity of the continuous fluid, and μ_b is the dynamic viscosity of the bubble fluid. Using the determined values, the resulting terminal velocity will be $W_b = 0.0724$ m.s^{-1}. Terminal velocity provides an indication of the simulation end time, which is crucial because when the bubble reaches the upper boundary, the results won't be realistic. The calculated terminal velocity results suggest that the bubble will exit the domain in approximately 0.05 seconds, so the simulation end time has been set to this value.

II.1. Boundary condition

This particular scenario encompasses various boundary conditions, and for a more lucid presentation, Figure 3 has been crafted. The intricacy of these boundary conditions primarily stems from the integration of interface capturing methods. It's noteworthy that among these methods, only the moving mesh method necessitates an internal boundary condition—specifically, the fluid-fluid interface, which employs a free slip approach.

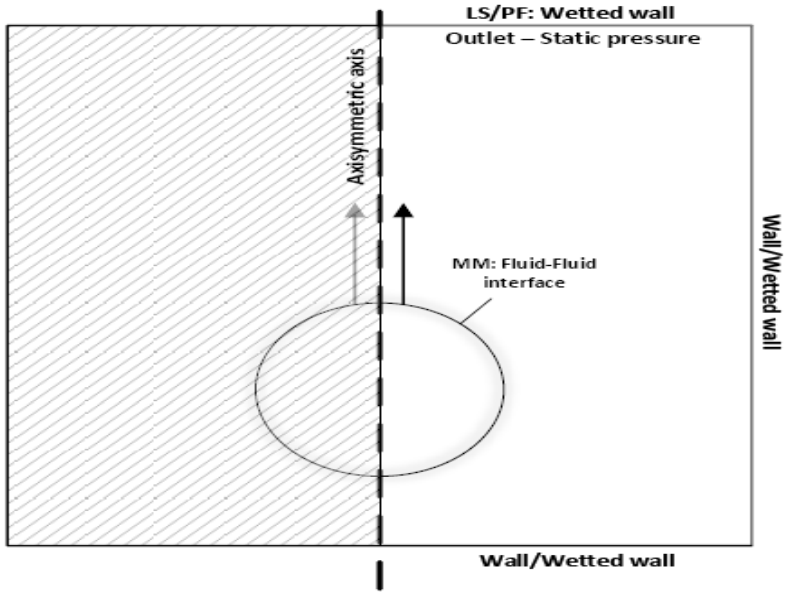

Figure 3. Overview of the utilized boundary conditions and their positions; dimensions are not to scale.

II.2. Geometry and Mesh

Figure 4 and 5 depict the geometry of the studied configuration and its corresponding moving mesh, respectively.

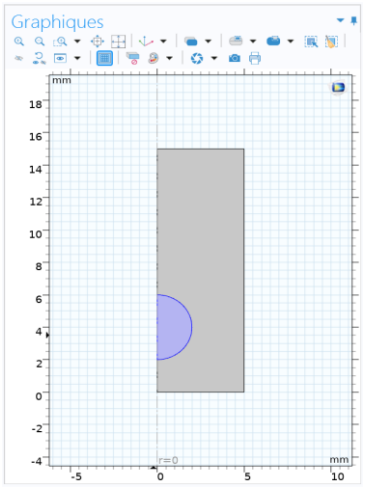

Figure 4. The geometry under study.

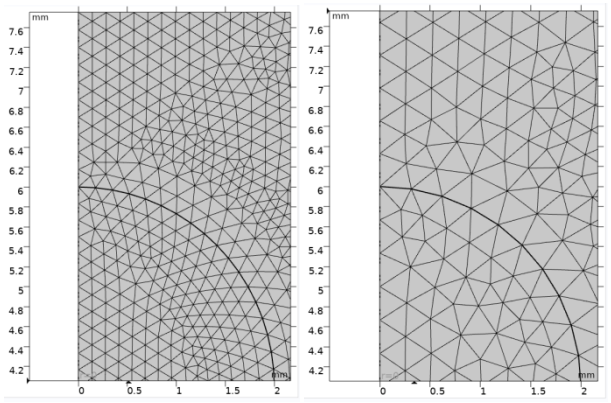

Figure 5. Moving mesh.

III. Results

In the context of hydrogen and water, the density contrast between them is quite pronounced. Utilizing the phase field approach, it has been uncovered that virtually diminishing this contrast can lead to more stable solutions. The virtual reduction of the density ratio signifies that the impact of this reduced ratio is conveyed solely through an adjustment in the mobility parameter by modifying the densities beyond the simulation boundaries. For hydrogen and water, this translates to a decrease in terminal velocity, consequently resulting in a smaller mobility adjustment parameter. Accurate calibration of these parameters is crucial, as simulations with precise settings converge much more rapidly. Incorrect parameters in the phase field model can cause the bubble to shrink and seemingly vanish. Further scrutiny reveals that, despite appearances, the average volume fraction over the domain remains constant, indicating that the interface couldn't be sustained, and no actual mass was lost. Figure 6 illustrates this effect, but by fine-tuning these parameters, the impact is mitigated to a radius loss of approximately 20%.

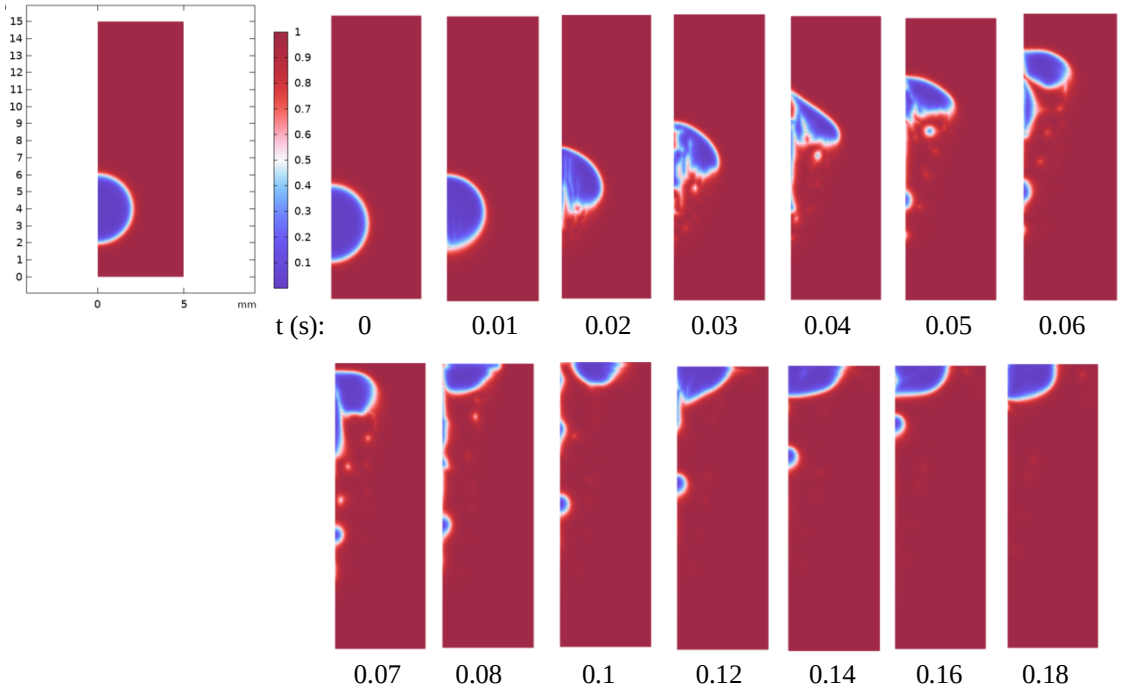

Figure 6. The progression of the ascending bubble at various time points.

IV. Conclusion

In this investigation, we employed the COMSOL software to simulate the motion of a hydrogen bubble within water. The findings from our study were thoroughly examined and discussed, underscoring the significance of embracing renewable and clean energy sources for their environmental friendliness and cost-effectiveness in comparison to traditional energy sources. The study emphasizes that harnessing hydrogen as a transitional energy indicator efficiently can offer alternative solutions, particularly in the face of depleting oil energy reserves, assuming mastery of production technologies.

The simulations were executed using COMSOL Multiphysics 5.6, employing a direct numerical solver for solution generation. To streamline computational efficiency, 2D axial symmetry models were adopted. Various interface capturing methods, including the phase field, level set, and moving mesh, were assessed for performance to identify the optimal approach for implementing the bubble growth model. The moving mesh methodology emerged as the most suitable choice, not only delivering the most accurate data but also closely aligning with the theoretical model of the ascending bubble. Additionally, the moving mesh approach demonstrated practicality in describing boundary conditions on a moving bubble interface, a crucial factor in the scenario of a growing bubble.

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