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Optimizing Molten Carbonate Electrolysis for sustainable fuel production: Experimental insights and machine learning enhancements

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Abstract- This paper presents an in-depth investigation into Molten Carbonate Electrolysis (MCE), combining experimental research with advanced machine learning-based modeling. MCE is explored for its potential in producing hydrogen and syngas, which are critical components for sustainable energy systems. This study examines the behavior of a single molten carbonate cell under various operating conditions and employs Artificial Neural Networks (ANN) to model and optimize the electrolysis process. The findings underscore MCE's viability for fuel generation and demonstrate the effectiveness of ANN in predictive modeling and operational optimization, offering significant insights for future energy systems.

Keywords-*Molten Carbonate Electrolysis, MCE, syngas generation, experimental investigation, Machine Learning, Artificial Neural Networks, ANN, mathematical modeling*

INTRODUCTION

The global shift towards renewable energy sources is not just a response to environmental concerns but a strategic necessity for ensuring long-term energy security. Renewable technologies like solar and wind power offer clean energy solutions but are inherently intermittent, posing significant challenges for integration into existing power grids. The variability in power generation from these sources requires robust energy storage systems that can stabilize the supply-demand balance, ensuring a reliable and continuous energy flow.

Energy storage technologies vary widely in their mechanisms and applications, ranging from batteries, which store energy in electrochemical form, to pumped hydro storage, which relies on gravitational potential energy. Among these, chemical energy storage, particularly in the form of hydrogen or synthetic natural gas (syngas), has emerged as a particularly promising solution. Electrolysis processes, which convert electrical energy into chemical energy stored in fuel, are central to this approach. These processes not only facilitate energy storage but also offer a pathway for producing carbon-neutral fuels, aligning with global decarbonization goals [1].

Molten Carbonate Electrolysis (MCE) is one such process that holds substantial promise due to its ability to operate at high temperatures, which enhances efficiency and allows for the co-generation of hydrogen and syngas. Moreover, MCE facilitates carbon dioxide capture and utilization, making it a dual-purpose technology that addresses both energy storage and carbon management challenges. Despite its potential, MCE has not been as extensively studied as other electrolysis technologies, particularly in terms of its optimization and control under varying operational conditions [2]. This paper seeks to fill this gap by investigating MCE through both experimental and computational lenses, using machine learning to enhance our understanding and control of the process.

MOLTEN CARBONATE ELECTROLYSIS FOR FUEL GENERATION

Molten Carbonate Electrolysis is a high-temperature process that operates by reversing the principles of a molten carbonate fuel cell (MCFC). In MCE, electrical energy is applied to drive chemical reactions that generate hydrogen and syngas, rather than producing electricity from fuel. The high operating temperatures, typically between 450°C and 650°C, differentiate MCE from conventional low-temperature electrolysis methods such as Alkaline Electrolysis or Proton Exchange Membrane (PEM) Electrolysis, which operate at temperatures below 100°C [3].

A. ELECTROCHEMICAL REACTIONS IN MCE

The core reactions in MCE involve the reduction of carbon dioxide and water at the cathode and the oxidation of oxygen at the anode. The reactions can be summarized as follows:

At the cathode:

$$CO_2 + H_2O + 4e^- \rightarrow CO_3^{2-} + H_2$$

At the anode:

$$CO_3^{2-} \rightarrow CO_2 + O_2 + 4e^-$$

In these reactions, the high temperature facilitates the efficient conduction of ions through the molten carbonate electrolyte, a mixture typically composed of lithium and potassium carbonates. This electrolyte not only serves as a

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medium for ion transport but also ensures that the reactions occur efficiently, with the carbonate ions (CO_3^{2-}) being transported from the cathode to the anode, where they are decomposed into carbon dioxide and oxygen [4].

The high-temperature operation of MCE also enables the use of less expensive and more readily available materials, such as nickel-based electrodes, which are stable under these conditions. Furthermore, the thermal energy required for the process can be partially supplied by external sources, reducing the overall electrical energy demand and enhancing the system's efficiency [5].

A. COMPARISON WITH OTHER ELECTROLYSIS TECHNOLOGIES

MCE stands out among electrolysis technologies due to its ability to simultaneously produce hydrogen and capture carbon dioxide. In contrast, low-temperature electrolysis methods, such as Alkaline and PEM Electrolysis, operate at lower temperatures and do not inherently facilitate CO2 capture. Solid Oxide Electrolysis Cells (SOECs), another high-temperature technology, operate at temperatures between 600°C and 900°C, splitting water into hydrogen and oxygen. However, SOECs do not inherently incorporate CO2 in their reaction mechanisms, which limits their application in carbon management [6].

One of the key advantages of MCE is its potential for co-electrolysis, where both CO2 and H2O are simultaneously reduced to produce syngas, a mixture of hydrogen and carbon monoxide. Syngas is a versatile feedstock that can be further processed into synthetic fuels, chemicals, or used directly in various industrial processes. The ability to produce syngas through electrolysis offers a pathway for integrating renewable energy into the chemical industry, providing a carbon-neutral alternative to traditional fossil fuel-based processes [7].

The potential for MCE to operate as part of an integrated energy system is significant. By coupling MCE with renewable energy sources, it is possible to generate hydrogen and syngas during periods of excess power production, storing energy in chemical form for later use. This not only stabilizes the grid but also provides a mechanism for the large-scale production of carbon-neutral fuels, contributing to both energy security and climate goals [8].

II. EXPERIMENTAL INVESTIGATION

A. EXPERIMENTAL SETUP AND METHODOLOGY

The experimental investigation of MCE was conducted using a laboratory-scale setup specifically designed to test the electrochemical performance of a single molten carbonate cell. The cell, with an active area of 20.5 cm², was constructed using nickel-based electrodes and a molten carbonate electrolyte composed of lithium and potassium carbonates in a 62/38 ratio [9].

The experimental apparatus included a controlled environment that allowed for the precise variation of temperature, gas flow rates, and pressure. This setup enabled a systematic study of the effects of these parameters on the cell's performance. Temperature control was achieved using dual external heaters, ensuring that the cell temperature remained stable within the desired range. Gas flows were meticulously regulated using mass flow controllers, which provided accurate delivery of steam, CO2, and hydrogen to the cathode, and oxygen to the anode [10].

The cell was operated under various conditions to assess its voltage, current density, and overall efficiency. The key experimental variables included:

Cathode Feed Composition: The composition of the gas fed to the cathode was varied, with different ratios of steam, CO2, and hydrogen.

Temperature: The cell was operated at different temperatures, specifically 600°C, 625°C, and 650°C, to observe the effects of temperature on electrochemical performance.

Pressure: The experiments were conducted at a constant pressure of 1 bar to maintain consistency across tests [11].

Measurements were taken using high-precision instruments to ensure accuracy in assessing the cell's performance. Data on voltage, current density, and efficiency were collected and analyzed to identify trends and correlations between the operating conditions and the electrochemical outcomes.

B. EXPERIMENTAL RESULTS

The experimental results provided valuable insights into the operation of MCE under different conditions. One of the most significant findings was the strong influence of temperature on the cell's performance. As the temperature increased from 600°C to 650°C, the cell's voltage decreased, indicating an improvement in efficiency. This trend is consistent with the theoretical expectations for high-temperature electrolysis processes, where higher temperatures enhance ionic conductivity and reduce polarization losses [12].

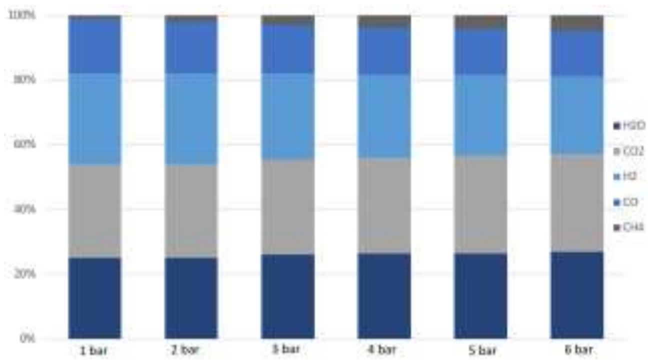

Figure 1: MCEC produced gas composition with dependence on operating pressure

The composition of the cathode feed gas was also found to be a critical factor in determining the cell's performance. A balanced mixture of steam and CO2, with a small fraction of hydrogen, was found to optimize the electrochemical reactions at the

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hydrogen, was beneficial in stabilizing the cell's operation. Hydrogen acted as a reducing agent, minimizing concentration polarization losses and enhancing overall efficiency [13].

Interestingly, the experimental data showed that higher hydrogen content in the cathode feed led to a decrease in the voltage required for electrolysis, particularly at higher current densities. This observation suggests that hydrogen may play a role in facilitating the formation of reactive intermediates, thus improving the overall kinetics of the electrochemical reactions [14].

Further analysis revealed that the optimal performance was achieved when the cathode feed gas contained approximately equal molar ratios of steam and CO2, with a small addition of hydrogen. This composition minimized the potential for carbon deposition on the cathode, which can occur when excess CO2 is present, leading to the formation of solid carbon through the Boudouard reaction. By carefully controlling the gas composition, the cell was able to operate efficiently without the risk of carbon deposition, which can degrade the performance and lifespan of the electrodes [15].

I. MACHINE LEARNING-BASED MODELING

A. INTRODUCTION TO ARTIFICIAL NEURAL NETWORKS (ANN)

Machine Learning (ML) has emerged as a powerful tool for modeling complex systems, particularly in scenarios where traditional mathematical models may not fully capture the intricate relationships between variables. Artificial Neural Networks (ANN), a subset of ML, are particularly well-suited for problems with nonlinear and multi-dimensional dependencies, such as those found in electrochemical processes [16].

ANNs are inspired by the structure and function of the human brain, consisting of interconnected nodes (neurons) organized in layers. These networks can learn from data by adjusting the weights of the connections between nodes, allowing them to make accurate predictions based on input variables [17].

In the context of MCE, ANN can be used to model the electrochemical behavior of the cell under different operating conditions. By training the network on experimental data, the ANN can learn to predict the cell's voltage and efficiency based on input parameters such as temperature, gas composition, and current density [18]. The advantage of using ANN lies in its ability to model complex, nonlinear interactions that would be difficult or impossible to capture using traditional modeling techniques.

B. 4.2. CONSTRUCTION AND TRAINING OF ANN MODELS

The ANN models developed in this study were designed to predict the MCE performance under various conditions. Four distinct models were constructed, each focusing on different influencing parameters:

Temperature-Dependent Model: This model predicts cell voltage based on temperature and current density. It was designed to capture the thermal effects on the electrochemical processes within the cell.

Fuel-Side Composition Model: This model predicts cell voltage based on the composition of the cathode feed gas, focusing on the interactions between different gas components and their impact on performance.

Oxidant-Side Composition Model: This model predicts cell voltage based on the composition of the anode feed gas, which primarily consisted of oxygen in this study.

Combined Thermal-Flow Model: This comprehensive model integrates temperature, gas composition, and current density to predict overall cell performance, accounting for the combined effects of thermal and flow dynamics [19].

Each model was constructed using a multi-layer perceptron architecture, with an input layer corresponding to the selected parameters, one or more hidden layers for feature extraction, and an output layer representing the predicted cell voltage. The models were trained using backpropagation, a supervised learning technique that minimizes the mean square error between predicted and actual outputs [20].

The training dataset was derived from the experimental data, with 70% used for training, 15% for validation, and 15% for testing. The models were optimized by adjusting the number of neurons in the hidden layers and selecting the most appropriate activation functions to ensure accurate predictions and prevent overfitting [21]. The training process involved multiple iterations, with the network weights being adjusted to minimize the error between the predicted and actual outputs.

C. PERFORMANCE AND VALIDATION OF ANN MODELS

The ANN models demonstrated excellent predictive accuracy, with average errors ranging from 0.17% for the temperature-dependent model to 0.35% for the fuel-side composition model. These results indicate that the ANN was able to effectively capture the nonlinear relationships between the input parameters and the electrochemical performance of the MCE [22].

The performance of the ANN models was validated against experimental data that were not included in the training set. This validation process is crucial for assessing the generalization capabilities of the models, ensuring that they can accurately predict performance under conditions that were not explicitly encountered during training. The models successfully predicted the cell's performance with high accuracy, even when tested on data points that were outside the range used for training. This capability is particularly valuable for applications where operating conditions may vary dynamically, and real-time predictions are necessary [23].

The robustness of the ANN models was further tested by introducing noise into the input data, simulating the effects of measurement errors or fluctuations in operating conditions. Despite the added noise, the models maintained their predictive accuracy, demonstrating their resilience and reliability in practical applications [24].

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I. OPTIMIZATION OF MCE OPERATION USING ANN

A. OPTIMIZATION PROCESS

The optimization of MCE operation was conducted using the trained ANN models, with the objective of minimizing the cell voltage while maximizing the current density. This approach is essential for improving the overall efficiency of the electrolysis process, as lower cell voltages reduce energy consumption, and higher current densities increase the rate of fuel production [25].

The optimization process involved adjusting the input parameters, such as temperature, gas composition, and flow rates, within the experimentally validated ranges. The ANN models were used to predict the effects of these adjustments on the cell's performance, and the optimal set of conditions was identified based on the model outputs [26].

The optimal operating conditions suggested by the ANN model included a temperature of 650°C and a cathode feed gas composition of 43.75% steam, 43.75% CO2, and 12.5% hydrogen. These conditions were chosen to balance the need for efficient electrolysis with the production of a desirable fuel composition, particularly hydrogen and syngas [27].

In addition to optimizing the operating conditions, the ANN models were also used to explore the potential for scaling up the MCE process. By simulating larger cell sizes and varying operational parameters, the models provided insights into how the process could be scaled while maintaining efficiency and stability. This type of modeling is particularly valuable for designing industrial-scale systems, where the complexity of interactions increases significantly [28].

B. VALIDATION OF OPTIMIZATION RESULTS

The optimized operating conditions were experimentally validated using the laboratory-scale MCE setup. The results confirmed the ANN model's predictions, with the cell operating at a lower voltage and higher current density compared to the non-optimized conditions. This validation not only demonstrated the utility of ANN in predictive modeling but also highlighted its potential for practical process optimization [29].

The experimental validation process also included a detailed analysis of the fuel composition produced under the optimized conditions. Gas chromatography was used to quantify the hydrogen and syngas produced, confirming that the optimized conditions led to an increase in the desired fuel output while minimizing the formation of unwanted by-products [30].

The successful application of ANN for optimizing MCE operation suggests that machine learning techniques can play a crucial role in advancing electrochemical technologies. By enabling real-time optimization and control, ANN can help overcome some of the challenges associated with the dynamic and complex nature of electrolysis processes [31].

II. DISCUSSION

The experimental and modeling results presented in this study provide a comprehensive understanding of the factors influencing MCE performance. The sensitivity of MCE to operating temperature and gas composition underscores the importance of precise control in achieving optimal fuel production and efficiency [32].

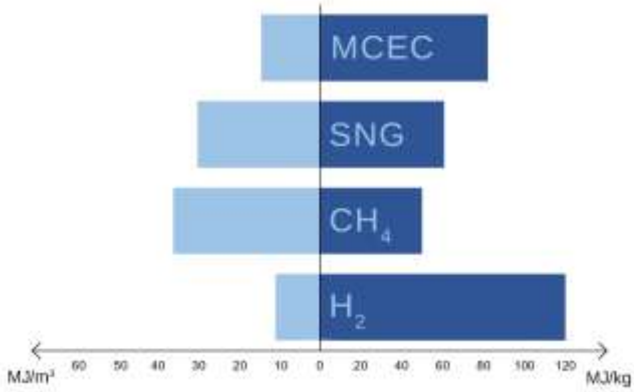

Figure 1: Comparison of volumetric and mass energy density the fuel generated by MCEC with SNG, hydrogen and methane

The experimental findings confirmed that maintaining a high operating temperature is essential for reducing polarization losses and improving the overall efficiency of the electrolysis process. However, the study also revealed that the composition of the cathode feed gas plays a critical role in determining the cell's performance. A balanced mixture of steam and CO2, with a small fraction of hydrogen, was found to optimize the electrochemical reactions, minimizing concentration losses and maximizing fuel production [33].

The successful application of ANN in this study highlights its potential as a powerful tool for modeling and optimizing complex electrochemical processes. The ANN models developed in this work demonstrated high accuracy in predicting the cell's performance under various conditions, and they were able to generalize beyond the training data, making them valuable for real-time process optimization [34].

Despite the promising results, the study also acknowledges several limitations. The accuracy of the ANN models is highly dependent on the quality and quantity of the training data. In cases where the dataset is limited or contains significant measurement errors, the model's predictions may be less reliable. Future research could focus on expanding the dataset and exploring more sophisticated ANN architectures to enhance the predictive capabilities of the models further [35].

Another limitation is the inherent complexity of the electrolysis process, which involves multiple interacting variables. While ANN is effective in capturing these interactions, it does not provide explicit insights into the underlying physical mechanisms. Integrating ANN with traditional mechanistic models could offer a more comprehensive understanding of the process and improve the robustness of the predictions [36].

systems for MCE. Such integration would enable dynamic adjustment of operating conditions in response to changing input parameters or external factors, such as fluctuations in renewable energy supply. This real-time optimization could significantly enhance the flexibility and resilience of MCE systems, making them more adaptable to the demands of future energy grids [37].

I. CONCLUSION

This research demonstrates the feasibility of using Molten Carbonate Electrolysis for efficient fuel generation, with the added benefit of carbon dioxide utilization. The integration of machine learning, specifically Artificial Neural Networks, has proven to be a powerful tool for both modeling and optimizing the electrolysis process.

The findings suggest that MCE, supported by advanced modeling techniques, could play a significant role in future energy systems, providing a sustainable and flexible solution for energy storage and fuel generation. The study also paves the way for further research into the application of machine learning in electrochemical processes, with the potential to significantly enhance the efficiency and scalability of these technologies [38].

By enabling real-time optimization and control, machine learning techniques such as ANN can help overcome some of the challenges associated with the dynamic and complex nature of electrolysis processes. As renewable energy sources become increasingly integrated into power grids, technologies like MCE, supported by advanced modeling and optimization tools, will be crucial in ensuring a stable and sustainable energy future [39].

Moreover, the study's findings have broader implications for the development of integrated energy systems, where MCE could be coupled with renewable energy sources and carbon capture technologies to create a fully sustainable energy cycle. This vision aligns with global efforts to transition to a low-carbon economy, where innovative technologies like MCE will play a key role in achieving climate targets and ensuring energy security [40].

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