Extended Abstract

(For Ph.D. Open Seminar)

DESIGN, DEVELOPMENT AND PERFORMANCE ANALYSIS OF SCALABLE, MEMBRANE-LESS ELECTROLYZERS FOR HYDROGEN GENERATION

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Degree for which submitted: Doctor of Philosophy

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The growing demands of clean energy and increasing environmental concerns from the use of fossil fuels have accelerated research in the field of hydrogen. It is considered a promising fuel due to its high gravimetric energy density and produces water as a byproduct on combustion. Of various methods, water electrolysis by a cost-effective electrolyzer, powered by renewable energy sources, is a clean, viable and efficient technique. The present thesis presents various designs of a membrane-less electrolyser, which is scalable for larger production of green hydrogen gas, if integrated with renewable energy sources, from varied water-qualities. These designs were developed in prototypes employing 3D printing technology. Their performance was analysed in terms of H_2 purity and its cross over to the oxygen side. The progress of the work is divided in different chapters presented below.

Chapter 1. Introduction and research gap

The chapter presents various methods of H_2 production along with their advantages and disadvantages. One significant technology highlighted is water electrolysis using an electrolyzer. The chapter discusses about current electrolyzer technologies which include proton exchange membrane electrolyzer, anion exchange membrane electrolyzer, alkaline electrolyzer, solid oxide electrolyzer, and membrane-less electrolyzers. An extensive literature review of membrane-less electrolyzer is presented with their designs and performance. The chapter ends with the details of key problems with the existing technology, which provide basis and direction to the current work.

Chapter 2. Materials and Methodology

This chapter presents the details of materials and methodology used for the design, development, prototyping and optimization of electrolyzer performance. The electrolyzer's operation is similar to an alkaline electrolyzer, and KOH solution is used as electrolyte. The design of the electrolyzers is developed using AutoCAD software and is fabricated using 3D-printing material stable in the alkaline medium. The performance of electrolyzers, in terms of H_2 purity and its crossover to the oxygen side, was analyzed with internal and external gas chambers, by varying interelectrode spacing, by changing production rate, and by varying flow rates and temperatures of the electrolyte. The collection of gas samples, and experimental and mathematical method for the determination of hydrogen concentration on dry basis are also discussed.

Chapter 3. Scalable and modular membrane-less electrolyzer: external gas separation and internal gas separation using vertical and tilted weir

The chapter discusses two prototypes of electrolyzers: one with an external gas separation chamber and another with an internal gas separation chamber featuring a vertical and a tilted weir. The vertical weir simplifies the fabrication process using 3D printing and is also easier to scale up compared to the tilted weir. A comparative performance analysis of electrolyzers with and without an internal gas separation chamber (IGSC) is presented. The asymmetric electrode configuration in both types of electrolyzers performs better than the symmetric electrode configuration for both vertical and tilted weir electrolyzer. In the electrolyzer with the IGSC, a maximum H_2 production rate of 2.0 L/h is achieved at a flow rate of 500 ml/min, resulting in a H_2 purity of approximately 99.83% and an oxygen crossover of about 2.52% at 30°C in vertical weir electrolyzer. By connecting multiple units in series, the electrolyzer is scaled up to produce a higher hydrogen output of 6 L/h, with a hydrogen purity of 99.7% and an oxygen crossover of around 2.58%. Results related to the tilted weir indicate that the electrolyzer's performance improves with increasing temperature and H_2 production rate, up to a certain limit. The electrolyzer without an IGSC, which features gas-separation chambers externally, demonstrates superior performance. As a result, the limiting H_2 production rate is higher, with H_2 purity reaching approximately 99.95%

and an oxygen crossover of about 2.09% at a 30°C electrolyte temperature. The simplicity of the prototypes' design underscores their potential for scaling up to achieve larger H_2 gas production.

Chapter 4. Optimizing arrangement of electrodes for flow-based gas separation in membrane-less electrolyzer

The chapter presents design and performance of 3D printed membrane-less electrolyzer with four arrangements of mesh/foam electrodes (with no gap: MENG-1 and MENG-2; with gap (MEWG and FEWG) for H_2 production. Results show that increasing flow rates increase current density in the order: MENG-1 < MENG-2 < MEWG < FEWG - the performance trend follows the combined effect and synergy between increased drag on gas bubbles, surface-renewal of electrode for improved electrochemical kinetics, and reduced flooding effect near electrode surface, stemming from gap-allowance. The enhancement in the purity of H_2 gas and its reduced crossover to the oxygen side are achieved in the same order. The highest purity of H_2 gas achieved is 99.20%, and its crossover is 2.13%, at 30.4°C for the FEWG electrode arrangement. Higher temperature operation marginally affects the performance due to increased migration of the gases. At 60°C, purity and crossover are: (95.63, 3.19) % for FEWG. The reported production rate in the study is the highest for a similar membrane-less electrolyzer presented in the literature, and it can be further scaled up.

Chapter 5. A modular and scalable architecture for high-volume H_2 production electrolyzer using a cloth separator

The chapter discusses designs of electrolyzer which employs cloth separation instead of a costly membrane and helps in lowering the pumping cost of the electrolyte. The location of electrolyte outlet is varied for better performance. At production rate of 3 L/h, H_2 purity of 99.13±0.21% is achieved with its crossover of 2.86±0.31%, using electrolyte flow rate of 100 mL min^{-1}. The electrolyzer is further scaled in phases for larger production rates of 50 L/h, 83 L/h, and 200 L/h, maintaining hydrogen purity at ~99% and its crossover below the flammability limit of 4%. Ultimately, the design was scaled up to a pilot-plant level to produce 1000 L/h of hydrogen at STP.

Chapter 6. Conclusions and future scope of work

The thesis presents a detailed analysis of various designs of electrolyser, derived from hydrodynamic principles, leading to efficient gas separation without employing costly membrane for the production of hydrogen gas. The design was optimized by analysing performance with varying flow rate, temperature and concentration of electrolyte, electrode arrangement, inter-electrode spacing, and location of outlets. Various electrodes, including nickel mesh and nickel foam, were employed for higher production rate and bubble disengagement. Nickel foam is found to a suitable material for its higher electroactive area for reactions. The scale-up of the optimized designs, by increasing the size of electrode and increasing number of cells in the series, was done and prototypes of the same were fabricated using 3D printing technology in phases, starting from 2L/h to 3, 6, 20, 50, 83, and 200 L/h. Based on the results obtained, a complete system including balance of plant was fabricated to produce 1000 L/h H_2 gas. The testing and performance analysis of the same is ongoing.

The current set-up operates at just above the atmospheric pressure and uses DI water. The future scope of work will be directed towards:

i. Electrolyzer for sea water electrolysis for hydrogen production.
ii. Operational changes and materials for hydrogen production at reduced potential
iii. Process improvements for stable pressure inside the electrolyser
iv. Operation of electrolyser at higher pressure
v. Scaling it up further to produce 2 – 10 m^3/h hydrogen gas.

Manuscript prepared/ under preparation/published from the present work:

1. Design, development, and performance analysis of scalable, membrane-less, 3D printed electrolyzers for hydrogen generation. International Journal of Hydrogen Energy. (*https://doi.org/10.1016/j.ijhydene.2026.154335*)
2. 3D-printed, scalable, membrane-less electrolyzer for hydrogen production: design and optimization. Energy & Fuels (In *Revision*)
3. Design and scalable solutions with membrane less vertical weir electrolyzers for a sustainable hydrogen future. (*Prepared*)
4. A modular and scalable architecture for a high-volume H_2 production electrolyzer using a porous cloth separator (*Prepared*)

Patents

1. Membrane-less electrolyzer for green hydrogen production. Application No: **202311085838**
2. Modular membrane-less electrolyzer for scalable and flexible operation (In process).