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# Hydrogen Separation and Purification Techniques in Pyrolysis-Based Processes

Augustine Oluwatoyin OJO<sup>1</sup>, Bernard ADARAMOLA<sup>1</sup>, Adeniran Sunday AFOLALU<sup>1,2</sup>, Stella Isioma MONYE<sup>1</sup>

<sup>1</sup>Department of Mechanical and Mechatronics Engineering Afe Babalola University, Ado Ekiti, Ekiti State, Nigeria ojoaugustine@pg.abuad.edu.ng/adaramolaba@abuad.edu.ng/adeniran.afolalu@abuad.edu.ng/monyeis @abuad.edu.ng

<sup>2</sup>Department of Mechanical Science, University of Johannesburg, South Africa

Corresponding Author: augustineojo@yahoo.com, +2348033710866

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Abstract: The production of hydrogen via pyrolysis process has gained a significant attention due to its capacity to offer reliable and sustainable as well as efficient techniques for hydrogen energy generation. Despite these benefits, there are challenges with its purification and separation which represent a major procedure during its production. This study focused on the current methods deployed in hydrogen production with emphasis on its separation and purification during pyrolysis-based process. These methods include pressure swing adsorption, membrane filtration and cryogenic separation techniques. Each of the methods, principles of operation as well as it relates to the pyrolysis gas composition were discussed. Their advantages, limitations and the desired purity of hydrogen produced were inclusive during the discussion. Findings showed that despite the significance of these technologies, research is still needed to achieve a more cost effective, scalable and energy efficient methods for the production of hydrogen especially in the areas of purification and separation.

Keywords: Hydrogen Production, Pyrolysis Process, Separation Techniques, Purification Methods, Sustainable Energy

### 1. INTRODUCTION

Hydrogen currently plays a key role in oil refining, fuel upgrading, and ammonia production. In the future, its use is expected to expand significantly—especially in industries aiming for low-carbon operations. Hydrogen is particularly important in the global push for net-zero emissions because it produces no greenhouse gases or atmospheric pollutants when burned [1–3].

However, the way we produce hydrogen today is still heavily reliant on fossil fuels, making the process both energy-intensive and carbon-heavy [4–5]. A common method, steam methane reforming, generates large amounts of carbon dioxide as a byproduct. In fact, by 2020, global hydrogen demand had reached around 90 million tons per year—contributing to roughly 900 million tons of CO<sub>2</sub> emissions annually [6–9]. While steam methane reforming remains one of the cheapest production methods, its environmental impact needs to be addressed if it's to align with low-carbon goals [10]. In response, researchers like Celik et al. [11] explored the environmental and economic viability of thermal pyrolysis, which uses natural gas or biogas to produce hydrogen and solid carbon (syngas) as end products. Their study spanned several countries—including the USA, China, Germany, Saudi Arabia, and Türkiye—and compared this method with steam reforming and water electrolysis.

The findings showed that power generation in these regions still results in high carbon emissions—between 0.36 to 0.70 kg CO<sub>2</sub>e per kWh. This leads to hydrogen production via electrolysis generating as much as 18 kg CO<sub>2</sub>e per kg of hydrogen. In contrast, thermal pyrolysis of natural gas can significantly cut emissions, ranging from just 6 to 12 kg CO<sub>2</sub>e per kg H<sub>2</sub>. Even better, if renewable energy is used to power the process, emissions can drop further to around 2 kg CO<sub>2</sub>e per kg H<sub>2</sub>—making it much more eco-friendly than traditional steam reforming with carbon capture [12–16].

Thermal pyrolysis also has cost advantages in certain regions. For example, the USA and Saudi Arabia can produce hydrogen for under €1 per kilogram. Using biogas instead of natural gas can even result in *negative* emissions—around -1.28 kg CO₂e per kg of syngas—if powered by renewable energy, effectively making the process a carbon sink [18]. However, despite its environmental benefits, producing hydrogen from biogas isn't as cost-effective as steam reforming due to higher feedstock prices and smaller production scale [19–20]. Overall, thermal pyrolysis of natural gas offers both environmental and economic benefits, making it a strong alternative to large-scale steam reforming and electrolysis—particularly when the process doesn't generate large amounts of solid carbon [21–22].

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The system itself involves several stages, as illustrated in Figure 1. First, the input gas is preheated to about 800°C using a heat exchanger powered by hot product gas. An electric heater then raises the temperature further before the gas enters a high-temperature pyrolysis reactor [23]. Inside the reactor, carbon is separated from the gaseous components. The resulting gas is then cooled to about 100°C using a water-based cooling system [25]. Finally, the product gas undergoes purification via pressure swing adsorption (PSA) to ensure high purity. The key components in the process are:

- i. Heat exchanger
- ii. Electric heater
- iii. Pyrolysis reactor
- iv. Cooling system
- v. Gas separation unit

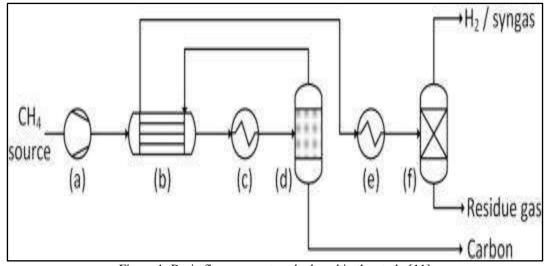


Figure 1: Basic flow concept as deployed in the study [11]

#### 2. LITERATURE REVIEW

## 2.1 Membrane-Based Separation Methods

According to research by Yang et al. [26], hydrogen stands out as a clean, zero-carbon energy carrier that plays a key role in the global shift toward renewable energy. One promising approach to hydrogen production involves membrane reactors, which use advanced membrane technology to enhance both chemical reactions and the purification of hydrogen at the same time. However, a major challenge with this process is *concentration polarization*—a condition where uneven concentrations develop across the membrane, disrupting the balance between the reaction and separation processes and limiting overall efficiency.

To address this, researchers have been working on improving membrane reactor technology by focusing on various aspects such as membrane materials, performance indicators, and new evaluation methods [27]. Studies also explored how factors like operating conditions and the structural design of these reactors impact their performance [28]. A key takeaway was that performance can be enhanced by adopting strategies like reducing travel distances for gases, creating multiple flow pathways, smoothing reaction paths, and allowing the removal of multiple products simultaneously [29].

Another promising idea involved designing specific patterns on the membrane surface to disrupt concentration boundary layers and reduce polarization effects [30–32]. These innovations not only boost performance but can also make hydrogen production more cost-effective by lowering reliance on expensive materials—like precious metals—and combining different product separation techniques [33–34]. However, a lack of standardized evaluation methods for these membrane reactors is currently slowing down their path to commercialization [35–40].

In a separate study, Cormos et al. [41] emphasized the importance of reducing carbon dioxide emissions as part of the effort to achieve climate neutrality. They focused on integrating renewable energy sources with carbon capture, utilization, and storage (CCUS) technologies as a vital solution for decarbonization. Their study explored innovative approaches, particularly biogas reforming, in combination with both pre- and post-combustion carbon capture using membranes to produce green hydrogen. Their assessment targeted a 100 MWth (thermal megawatt) capacity green hydrogen plant and analyzed various plant designs with and without carbon capture features. Depending on the capture method used, the decarbonization rate ranged from 55% to as high as 99%. Impressively, the membrane-based setups not only maintained high energy efficiencies (between 55% and 60%) but also reduced the energy and cost burden typically associated with carbon capture—showing penalties as low as 3.6% and up to 15.5%, regardless of the capture strategy. Even more significantly, the systems achieved low or even *negative* carbon emissions—up to −468 kg of CO₂ per MWh of green hydrogen—making the entire biogas production chain carbon negative [42]. Figure 2 in their study illustrates the plant's

layout and energy integration strategy. This setup mirrors the conventional steam methane reforming system used in industries like fertilizer and ammonia production but includes integrated carbon capture components.

The evaluation covered five different cases:

- Case 1: Biogas reforming without carbon capture
- Case 2: Reforming with pre-combustion carbon capture
- Case 3: Reforming with both pre- and post-combustion carbon capture
- Case 4: Pre-combustion capture using membranes
- Case 5: Combined pre- and post-combustion capture using membranes

Figure 3 illustrates how heat and energy are managed within the catalytic biogas reforming plant, showcasing the potential for more efficient and sustainable hydrogen production through thermal integration.

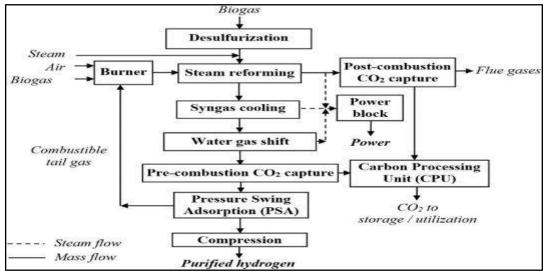


Figure 2: Reforming of biogas with membrane-based pre-and post-combustion capture of carbon [41]

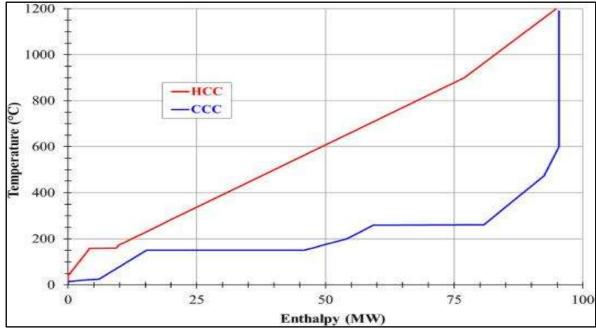


Figure 3: Thermal integration analysis of the evaluated biogas catalytic reforming plant [41]

In reality, the efficiency of the biogas-powered plant was significantly improved through a detailed heat integration analysis, which was carried out using the pinch method [43]. This technique involved evaluating both mass and energy balances to identify available heat sources (hot streams) and heat demands (cold streams) within the system [44]. Key sources of heat, such as the syngas stream from the reformer and flue gases from the burner, were used to generate steam. Part of this steam was reused within the plant—particularly for the biogas reforming process—while the excess steam helped generate electricity to meet the plant's internal power needs [45]. To better understand how heat was distributed, all

the available heating duties from hot streams at each temperature level were combined to form a *hot composite curve*. This was compared with the *cold composite curve* to set energy performance targets for the plant and determine how much additional heating or cooling (hot and cold utility) would be required [46].

The economic viability of the project was also assessed using a well-established methodology consistent with existing studies in the field of low-carbon energy. For estimating capital costs, a cost correlation approach was used [47–50]. This method relied on data from process simulations to calculate the processing capacities of various subsystems in the plant [51]. From this, the *specific investment cost*—the cost per unit of production capacity—was derived.

Figure 4 illustrates the breakdown of specific investment costs across various green hydrogen production setups. It highlights the most capital-intensive components, including the reformer section, the carbon capture unit, the power generation block, and supporting utilities [52]. Among all the cases studied, Case 5—which involved both pre- and post-combustion carbon capture using membrane technology—had the highest capital cost. This was largely due to the added complexity and expense of the post-combustion capture system [53–55]. This result is especially important because it reveals a key limitation of using membrane-based separation technologies in post-combustion carbon capture. While membranes can be effective, their cost and technical challenges in this specific application need to be considered when designing efficient and affordable green hydrogen plants.

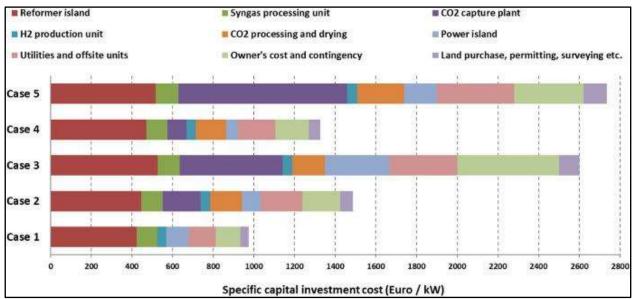


Figure 4: Breakdowns of the specific capital invested in biogas reforming strategies [41]

#### 2.2 Pressure Swing Adsorption (PSA) Method for the Purification of Hydrogen

Purifying hydrogen is a crucial step in producing high-quality, fuel-grade hydrogen. A study by Abd et al. [56], examined how effective activated carbon molecular sieves are for removing carbon-based impurities from hydrogen using the *Pressure Swing Adsorption (PSA)* method under non-isothermal (non-constant temperature) conditions. The researchers built a dynamic model to explore how different factors—like temperature, the density of the adsorbent material (solid bulk density), and the concentration of hydrogen in the feed—affect the purity of the final hydrogen product. To confirm the reliability of their model, they compared the simulated results with actual experimental data under the same conditions. They also used a method called *Central Composite Design* to analyze how these variables influence the PSA system. Their findings showed that the hydrogen purity reached 99.5%, with a recovery rate of 58.82%. However, after optimizing the process—specifically adjusting the temperature to 300 K, the solid density to 720 kg/m³, and the hydrogen concentration to 88%—they were able to increase the hydrogen purity even further to 99.99%.

In a related study by Zafanelli et al. [57], highlighted the challenges of purifying green hydrogen sourced from natural gas pipelines. Due to the low concentration of hydrogen in the gas grid, using a single-stage PSA system isn't efficient or cost-effective. To address this, researchers designed a **dual-stage Vacuum Pressure Swing Adsorption (DS-VPSA)** system that uses two types of adsorbents: *Carbon Molecular Sieve (CMS) 3K-172* in the first stage and *Zeolite 13X* in the second. The CMS material was used for kinetic separation—it effectively isolated hydrogen from methane, increasing the hydrogen concentration from about 20% to over 60% by volume. In the second stage, Zeolite 13X carried out a thermodynamic separation to further purify the hydrogen, achieving over 99% purity by volume. To fine-tune the system, the researchers built a mathematical model in Aspen Adsorption, a simulation software, and ran several tests to find the best operating conditions. They also ran a parametric analysis to optimize key performance metrics like purity, recovery rate, productivity, and energy efficiency. The final results were promising: the system achieved a hydrogen purity of 99.97% (suitable for fuel cells), a recovery rate of about 67%, productivity of  $1.60 \times 10^{-2}$  kg H<sub>2</sub> per kg of adsorbent per hour, and a specific energy consumption of approximately 10.06 MJ per kg of hydrogen.

Figure 5 in the study shows the flow diagram of the DS-VPSA process, while Figure 6 illustrates how the Aspen simulation was set up. From Figure 6, it's clear that a single-bed approach was used to reduce computation time. The simulation also incorporated interaction units to store and replay data during scheduled steps. Additionally, three valve-integrated units (VIUs) were used to perform equalization steps, and two virtual tanks (labeled Void1 and Void2) were included to simulate dead volumes in the piping and connections around the adsorption bed.

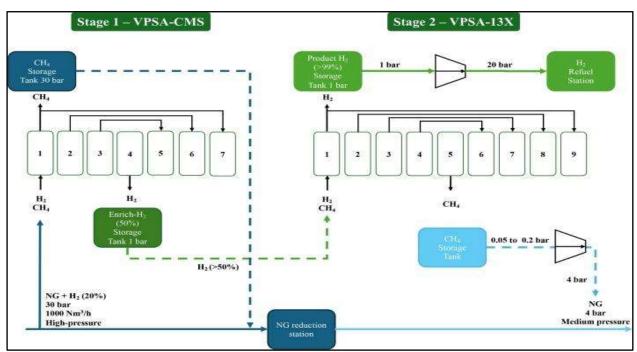


Figure 5: Flow diagram showing the Dual-stage vacuum pressure swing adsorption process [57]

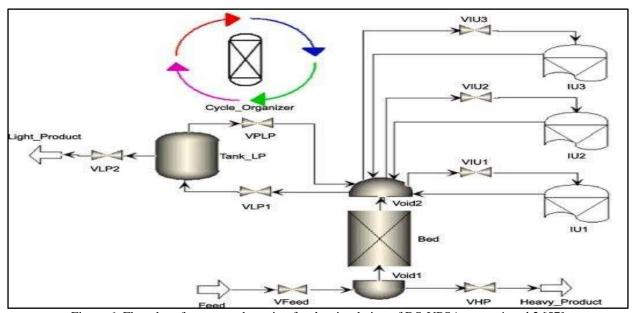


Figure 6: Flowchart for aspen adsorption for the simulation of DS-VPSA stages 1 and 2 [57]

## 2.3 Cryogenic Distillation Technique for a sustainable Purification of Hydrogen

Separating and purifying hydrogen remains a significant challenge, especially when high purity and recovery rates are required for certain applications. These difficulties stem not only from the nature of hydrogen itself but also from the properties of other components mixed with it. While several methods exist, **membrane-based and cryogenic technologies** are the most commonly used for hydrogen separation—more so than pressure swing adsorption.

In a study by Naquash et al. [58], two different hydrogen purification techniques were evaluated:

• Case 1 used membrane-assisted separation

#### Case 2 employed a cryogenic-assisted approach

Both systems began with hydrogen production from syngas via the *water-gas shift reaction*, followed by separation using their respective technologies. To improve energy efficiency, both setups were integrated with an *Organic Rankine Cycle (ORC)* for waste heat recovery. The entire process was modeled using Aspen HYSYS v11 simulation software. The results showed that **Case 1 consumed less energy**, around **0.50 kWh per kg of hydrogen**, compared to **2.01 kWh/kg** in Case 2. However, this came at a cost: **Case 1 had lower hydrogen purity and recovery rates**. On the other hand, from an *exergy* (usable energy) perspective, Case 1 was more efficient, showing **28.4% efficiency** versus **14.7% in Case 2**. From an economic standpoint, **Case 1 turned out to be more expensive**, with a projected cost of \$17.7 million, mainly due to the high price of compressors. In contrast, **Case 2 cost about \$10.2 million**, making it more budget-friendly despite its higher energy consumption.

In another study by Abdelkareem et al. [59], researchers explored the **production of hydrogen from waste metals** such as aluminum, magnesium, and zinc. The process was broken down into detailed steps to showcase how hydrogen can be generated from these metal scraps, comparing their advantages and drawbacks against conventional methods. Hydrogen can be generated from waste metals by reacting them with water, steam, acids (HCl, H<sub>2</sub>SO<sub>4</sub>), alkalis (NaOH), or organic acids.

The choice of co-reactant depends on the metal type, desired reaction temperature, and environmental safety. Metals such as aluminum (Al), zinc (Zn), magnesium (Mg), and iron (Fe) can react with water (H<sub>2</sub>O) or acids (H<sup>+</sup>) to release hydrogen gas (H<sub>2</sub>). In the process, the metal is oxidized to its ionic form while hydrogen ions (or water molecules) are reduced to H<sub>2</sub>. Metal (M)+Co-reactant $\rightarrow$ Metal oxide or salt+H2 $\uparrow$ 

Key findings revealed:

- Magnesium had a hydrogen yield of about 70%, thanks to its high specific energy density, giving it an advantage in hydrogen production from scrap metal.
- Aluminum alloy powders showed even better performance, yielding up to 98% hydrogen.
- Zinc, however, lagged with a relatively low yield of around 20%.

The study also discussed modern purification techniques—like pressure swing adsorption, cryogenic distillation, and membrane-based methods—that offer better performance than earlier versions. However, it also highlighted economic, technical, and social challenges that limit widespread adoption of hydrogen production from waste metals.

Figure 7 in the study illustrates the process flow. It begins with water molecules diffusing through a solution, coming into contact with metal particles. These water molecules are then adsorbed on the metal surface, where the reaction occurs at the metal (solid)/water (liquid) interface. This leads to the formation of metal oxide and the release of hydrogen gas, which then diffuses into the solution. This flowchart emphasizes the potential of using industrial metal waste to generate clean hydrogen, which could serve as an alternative fuel source. Figure 8 provides more insight by showing how different metals perform under varying conditions. Notably, at 200°C, magnesium and aluminum produced the highest hydrogen yields—921 cm³/g for magnesium and 1145 cm³/g for aluminum. In contrast, iron and nickel showed little responsiveness to temperature changes, making them less suitable under similar conditions.

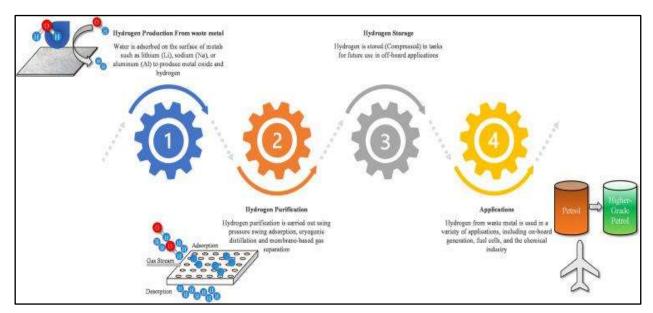


Figure 7: Flowchart showing hydrogen production from waste metal [59]

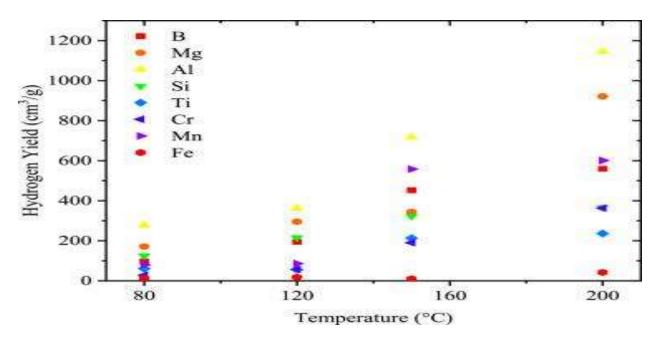


Figure 8: Overall yield of hydrogen gas for a selection of metals under different temperature conditions [59]

#### 3. CONCLUSION

In summary, hydrogen separation and purification are crucial steps in optimizing pyrolysis-based processes, as they ensure the consistent and efficient production of high-purity hydrogen. With the growing global demand for clean and sustainable energy, technologies like membrane separation, pressure swing adsorption (PSA), and cryogenic methods have become increasingly important—not just for producing better-quality hydrogen, but also for making the entire process more economically viable. These purification methods also play a key role in enhancing the sustainability of hydrogen production by reducing environmental impact and boosting the overall energy efficiency and reliability of pyrolysis systems. Looking ahead, future research could focus on refining these technologies, developing new advanced materials, and discovering more effective ways to integrate them into scalable, real-world systems for broader impact.

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