



Integration of Pyrolysis with other Thermochemical Processes for Sustainable Hydrogen Production: A Review

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Abstract: This study focused on exploring the integration of pyrolysis with other thermochemical processes as an alternative strategy for an efficient production of hydrogen. Pyrolysis is a thermochemical process that involves the decomposition of organic waste matter in the absence of oxygen to produce gaseous products, liquid and solid residues. Combining pyrolysis with gasification, steam reforming or carbon capture will lead to the increased efficiency in hydrogen production as discovered from the consulted literature study. This approach is not limited to increase in hydrogen output; it however, contributes to reduction in greenhouse gas emissions through the use of renewable feedstock like agricultural biomass. Thus, the synergies between the thermochemical processes help in the optimization of energy recovery and the generation of valuable products. However, despite these aforementioned benefits, there are still challenges associated process integration, energy efficiency as well as the scalability. Thus, continuous research and advancement in technology using other thermochemical processes would be key contributors to a sustainable hydrogen production as well as cleaner energy in future.

Keywords: Pyrolysis, Thermochemical Process, Hydrogen Production, Gasification, Renewable Feedstocks

1. INTRODUCTION

Among all the products derived from pyrolysis, hydrogen stands out as a crucial alternative energy source, largely due to its clean-burning nature and versatility [1]. However, hydrogen isn't readily available in nature—it has to be produced from other energy sources [2]. Currently, approximately 90% of global energy production still depends on fossil fuels, and hydrogen obtained through traditional fuel reforming methods is linked to significant carbon emissions [3]. This heavy dependence on fossil fuels emphasizes the urgent need for cleaner, more economical hydrogen production alternatives [4][5]. In this context, pyrolysis emerges as a promising solution, especially for converting waste plastics into hydrogen-rich gas mixtures, thereby simultaneously addressing the challenges of waste management and sustainable energy generation [6][7]. This approach is particularly relevant for global efforts toward energy transition and sustainability goals. Studies have also shown that the growing diversity of plastic products has led to transformative changes in industries and lifestyles worldwide [7][8]. Plastics have largely replaced traditional materials like metals, wood, ceramics, and glass, thanks to their low cost, lightweight properties, durability, and ease of processing [9]. These advantages have contributed to lower manufacturing costs and increased mass production, making plastics indispensable across many sectors, including healthcare, food packaging, sports, transportation, electronics, and construction [10][11][12].

However, this rapid growth in plastic consumption has created another major issue: the mounting accumulation of plastic waste due to the increasing variety of plastic products [13]. The effectiveness of pyrolysis in converting this waste depends on the composition and type of plastic feedstock. Plastics break down into monomers and smaller hydrocarbons during pyrolysis, producing a mix of oil and gas rich in hydrocarbons [14]. The quality and yield of these products vary depending on the type of plastic present and the operational parameters (especially heating rate, catalyst, reaction temperature, residence time and feedstock composition) of the pyrolysis system. Research has shown that using polyethylene terephthalate (PET) in the feedstock tends to increase emissions of carbon monoxide and dioxide, while plastics such as polystyrene (PS) and polypropylene (PP) are capable of generating higher quantities of hydrogen and hydrocarbon gases [15][16]. Therefore, a deep understanding of feedstock composition and reaction conditions is essential for optimizing the pyrolysis process and improving the efficiency of energy recovery systems [17]. One notable study focused on low-carbon hydrogen production via the non-oxidative decomposition of methane [18]. The researchers compared various methods—plasma-based methane decomposition, water splitting, thermochemical cycles, and steam methane reforming—against molten metal-based methane pyrolysis, evaluating each method from techno-economic, thermodynamic, and environmental perspectives. The study emphasized the importance of selecting the right molten metal catalysts and reactor materials, alongside considerations for sustainable heat sources and reactor configurations [19][20]. To understand the reaction dynamics better, the study utilized electromagnetic levitation to evaluate the intrinsic reaction

rates based on bubble surface area, regardless of the reactor type or gas residence time. It also considered key factors such as reaction kinetics, gas-liquid physical properties, and mass transfer rates for the optimal design of molten metal bubble column reactors [21][22].

A detailed process flow diagram illustrated how natural gas is processed, methane is decomposed, and both hydrogen and carbon are separated and stored, suggesting a pathway for commercial-scale hydrogen production [23]. Because the process produces more carbon than hydrogen by weight, the study also explored ways to refine and upgrade the carbon byproduct, increasing its purity and transforming it into high-value materials, which further enhances the economic viability of the process [24][25]. In another innovative study, researchers explored a sustainable approach to recycling textile spinning waste cotton via direct pyrolysis, converting it into high-quality biochar with improved energy content and structural stability [26]. The study evaluated how pyrolysis temperature impacts biochar yield, chemical composition, and physical properties, aiming to optimize the process for maximum carbon retention and energy efficiency. Findings showed that biochar yield dropped from 50.5% to 26.7% as the temperature increased from 300°C to 500°C, while the carbon content rose from 59.33% to 68.65%. Elemental analysis confirmed this trend, with carbon increasing from 53.13% to 73.62%, and oxygen dropping from 46.7% to 13.27%. The hydrogen content also fell, from 6.06% to 2.79%, resulting in enhanced thermal stability. X-ray diffraction (XRD) revealed a structural shift from amorphous cellulose to condensed graphite at higher temperatures. The biochar yield decreased due to enhanced thermal degradation and devolatilization of the organic constituents as pyrolysis temperature increased, resulting in more gaseous and liquid products. The elevated temperatures promote secondary cracking causing reduction of the solid carbon residue leading to the structural shift in XRD. Thermogravimetric analysis (TGA) further demonstrated the thermal resistance of the resulting biochar, showing it retained 14.7% of its mass at 800°C. In parallel, Differential Scanning Calorimetry (DSC) indicated enhanced material stability, with an endothermic peak shift from 65.5°C in raw cotton to 79.6°C at 500°C. These results support efforts to optimize pyrolysis for textile waste valorization, promoting a circular economy, reducing environmental pollution, and advancing renewable energy applications.

2.0 OPTIMISATION OF PYROLYSIS FOR A SUSTAINABLE PRODUCTION OF HYDROGEN

2.1 Integration of Gasification and Reforming Technologies

Mishra et al. [27] emphasized the growing importance of hydrogen as a clean and sustainable energy source, particularly by using biomass and plastic waste as feedstocks. Their study explored several gasification-based technologies—including pyrolysis-gasification, co-gasification, and co-pyrolysis gasification—to better understand how these approaches affect hydrogen-rich syngas production from waste materials. The research also examined various catalysts used in these processes, highlighting their unique properties and how they contribute to improved hydrogen yield [28–32].

In addition, the study investigated key factors such as the synergistic effects of combining feedstocks in co-gasification reactors, the impact of different gasifying agents, pretreatment processes, temperature variations, and the design of gasification reactors [33–38]. The findings revealed that these gasification techniques open up new possibilities for producing clean energy, which is a crucial step toward achieving a circular economy [39]. Moreover, the choice of catalyst and operating conditions had a significant effect on the efficiency of hydrogen generation during pyrolysis-gasification [40]. This research offers a fresh perspective on how integrating reactor advancements and process optimization can lead to more sustainable hydrogen production from waste.

Another study by Ochieng et al. [41], used the Aspen Plus simulation software to model a system that combines supercritical water gasification (SCWG) of sewage sludge with fast pyrolysis of wood residue. The goal was to produce hydrogen, power, and excess heat in a single integrated process. The simulation included sensitivity analysis, particularly to evaluate the impact of co-gasifying pyrolysis oil with sewage sludge. In addition to the technical modeling, the study assessed both thermodynamic performance and techno-economic feasibility.

Under the simulated conditions, the system generated about 23.1 mol of hydrogen per kilogram of biomass, 684 kW of electricity, and 3,400 kW of surplus heat. The overall energy efficiency of the process reached 61.2%, while the exergy efficiency was around 44.9%. From an economic perspective, the minimum hydrogen selling price was estimated at \$3.04 per kilogram. While this is lower than hydrogen from standalone SCWG, it remains slightly higher than pyrolysis alone. Overall, the integration of SCWG and fast pyrolysis proved to be a viable pathway for efficiently converting both wet and dry biomass into bioenergy. Figure 1 from the study illustrates the integrated setup, which consists of five major units:

- i. Supercritical water gasification reactor
- ii. Fast pyrolysis reactor
- iii. Gas/solid separation for pyrolysis
- iv. Hydrogen purification system
- v. Combustion unit for gas and char

In this setup, sewage sludge is processed through the SCWG reactor, while wood residue serves as the dry biomass in the pyrolysis reactor. The resulting pyrolysis oil is cooled and then co-gasified with sewage sludge. In parallel, the pyrolysis gases, char, and tail gases from the purification unit are fed into the combustion system, generating the heat needed for drying biomass, pyrolysis, and supercritical water gasification. Any surplus heat can be redirected for district heating, improving the system's overall utility.

In another related advancement, a study by Boretti [42], discussed the production of turquoise hydrogen through methane pyrolysis in molten metal—an emerging method praised for its potential to drastically reduce carbon emissions, unlike traditional steam methane reforming [43–48]. This process uses molten metal as a heat transfer and reaction medium, which breaks down methane into hydrogen and solid carbon. It offers a more sustainable lifecycle profile compared to conventional approaches [49].

The study also emphasized the benefits of integrating renewable energy inputs, utilizing biomethane, and finding effective uses for the solid carbon byproduct, all of which contribute to the environmental viability of molten metal-based methane pyrolysis [50]. However, this promising technology is still in its early development phase. Key challenges remain, particularly around scaling up the process, maintaining high-temperature stability, ensuring material durability, and managing heat efficiently throughout the system.

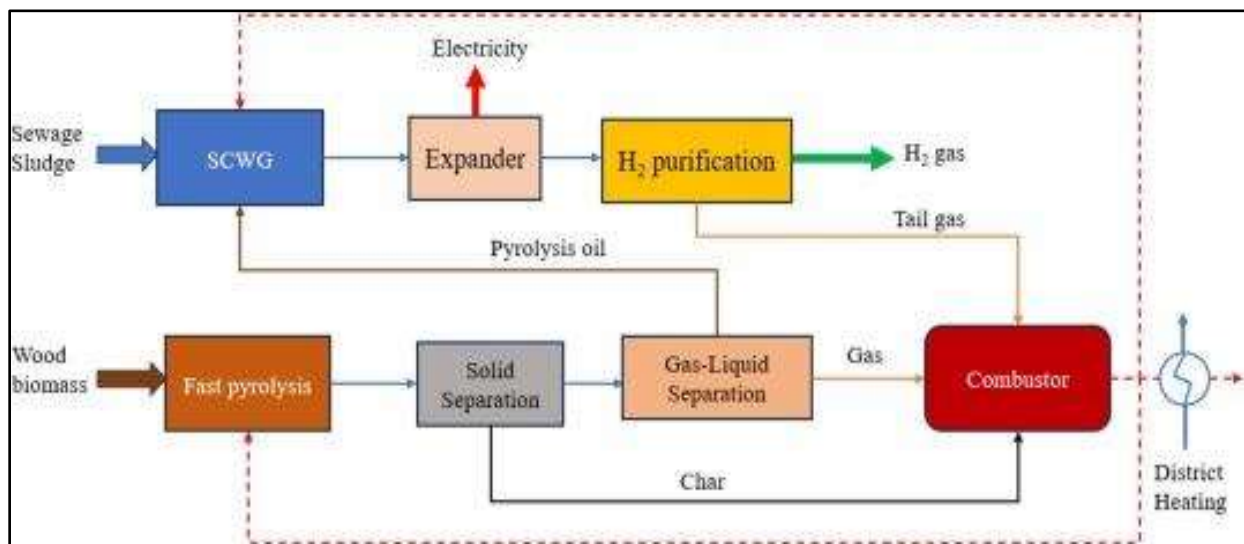


Figure 1: Integrated process as deployed in the study [41]

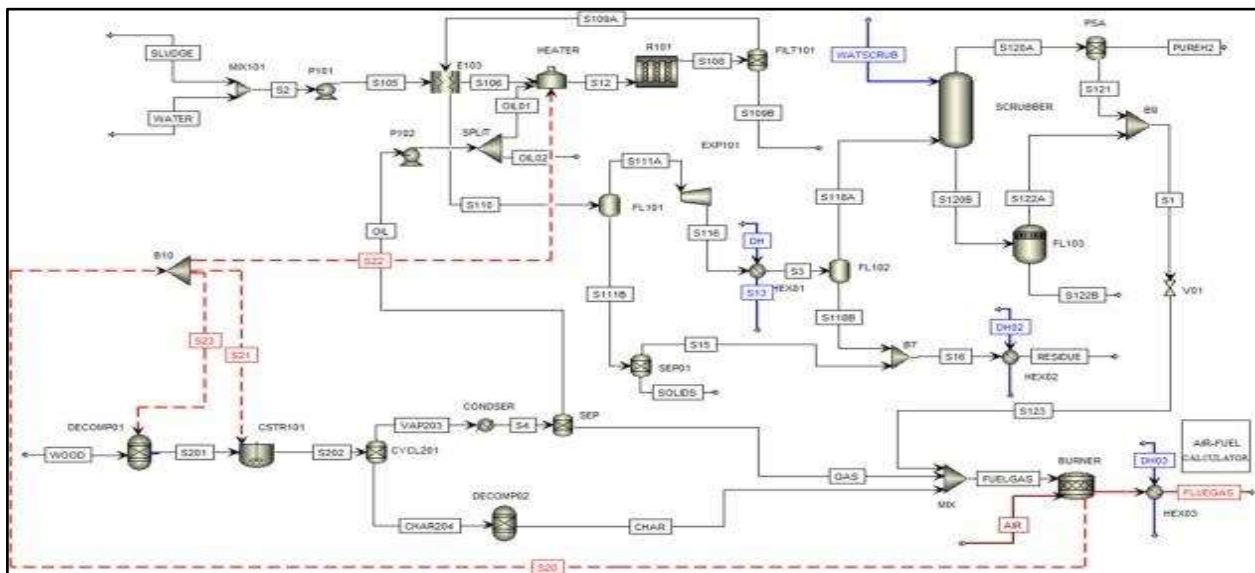


Figure 2: Integrated process model as modelled in Aspen Plus [41]

2.2 Role of Carbon Capture in Pyrolysis-based hydrogen production

Sobri et al. [51] introduced an innovative catalyst made from activated carbon infused with clove extract (AC-Cl) to break down common municipal plastic waste—such as polystyrene (PS), polypropylene (PP), and low-density polyethylene (LDPE)—into valuable gases like hydrogen and methane. Using a combination of laboratory experiments and computer simulations, they studied how the clove-derived phenolic functional groups in the catalyst influenced the pyrolysis process. These groups not only improved the chemical and physical activity of the catalyst, but also helped create more active microsites, which enhanced the breakdown of plastic molecules and prevented unwanted recombination of intermediate compounds.

This led to a notable increase in hydrogen production (up to 20.23%) and methane yield (17.24%), all achieved at lower temperatures, which translates to lower energy requirements. Additionally, the AC-Cl catalyst showed promise in capturing carbon during pyrolysis—thanks to its enhanced radical adsorption capacity, it achieved a 96.9% increase in trapped carbon atoms compared to other systems.

In a related study, researchers Wang et al. [52], examined how calcium oxide (CaO) can be used after pyrolysis to upgrade the resulting gas by removing carbon dioxide. They applied this method to gases generated from a variety of waste sources—including municipal sewage sludge, refuse-derived fuel (RDF), pine sawdust, and marine litter—all of which were pyrolyzed at 600°C, followed by CaO treatment and thermolytic decomposition at 1300°C.

Although calcium oxide did not significantly increase the absolute hydrogen yield or the heating value of the gas per kilogram of feedstock, it greatly improved hydrogen purity—from 49.6–83.3 vol% to 63.7–94.4 vol%—and boosted the hydrogen to carbon monoxide ratio from 1.1–5.7 to 1.9–19.8 across all feedstocks.

The carbon capture kinetics of calcium oxide, modeled using the grain model, revealed rate constants (K) ranging between 0.0001 and 0.0006/min, indicating feedstock-dependent behavior. Notably, RDF and biomass led to greater consumption of CaO compared to sludge or marine litter [53–57], likely due to faster carbonation and coke formation, which were influenced by the composition of the pyrolysis gases.

These findings support the integration of calcium oxide sorbents into pyrolysis systems to yield cleaner, hydrogen-rich gases [58]. Moreover, the use of normalized kinetic parameters provided a straightforward approach for selecting optimal feedstocks for decarbonizing pyrolysis gases [59]. A major factor affecting the carbonation behavior of CaO was the flow rate of carbon dioxide in the resulting gas stream [60].

Celik et al. [61] also explored thermal methane pyrolysis as a cost-effective method to produce hydrogen without direct CO₂ emissions. Despite its promise, the process mechanisms—particularly the inhibitory role of hydrogen, and the effects of operating at high pressures—remain underexplored. Their study looked at how using hydrogen and argon as diluents affects the product gas composition, methane conversion, and hydrogen selectivity. Variables included temperature, residence time, dilution ratios, and pressure inside a high-temperature reactor.

The results showed that the type of diluent had a major influence: a 50% variation in methane conversion was observed when comparing argon vs. hydrogen, even under the same conditions. Increasing the system pressure from 1 to 4 bar reduced the formation of unwanted byproducts in both cases, though the underlying reaction mechanisms differed. A key observation was that propylene formation occurred only in the argon-diluted mixture, and persisted even at extreme temperatures (up to 1600°C). This was linked to interactions between methyl and ethyl radicals, which created stable intermediates that blocked further reactions, leading to solid byproduct accumulation.

Figure 3 from the study showed the experimental setup and how different forms of solid carbon collected at various parts of the system during methane pyrolysis. Overall, this research offered new insights into the engineering and process design aspects of methane pyrolysis and clarified how diluent gases like hydrogen and argon influence the reaction dynamics and outcomes.

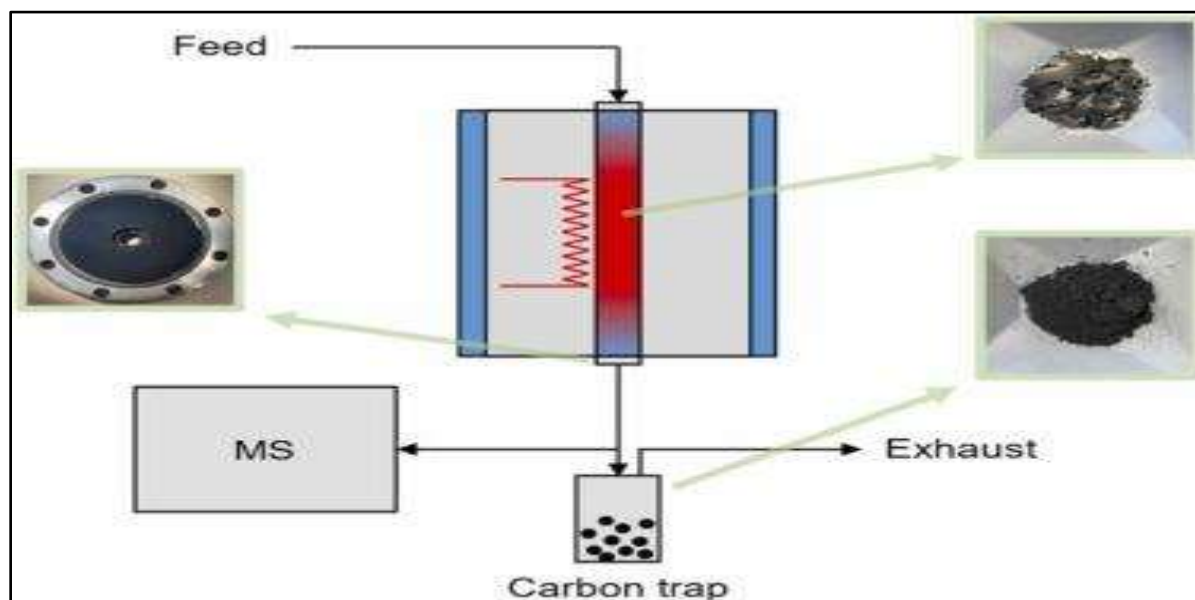


Figure 3: Schematic illustration of the experimental set-up [61]

3. CONCLUSION

This study explored how pyrolysis can be combined with other thermochemical processes—like gasification and steam reforming—to create a more sustainable method for producing hydrogen. By breaking down organic materials such as agricultural biomass, pyrolysis serves as a strong foundation for generating clean fuels and valuable chemicals. The

review highlighted that when pyrolysis is integrated with other thermochemical methods, it not only boosts the quantity and quality of hydrogen produced, but also lowers emissions and maximizes the use of renewable resources, such as biomass and plastic waste. The synergy between these combined processes results in better energy recovery, higher hydrogen output, and the creation of valuable by-products, which together enhance the economic and environmental efficiency of the system. However, the review also pointed out that there are still significant challenges to scaling these technologies for industrial use. Issues like optimizing system integration, ensuring effective heat and mass transfer, and addressing cost concerns remain barriers to large-scale deployment. Therefore, ongoing research and development are essential to refine these systems and make them more commercially viable. In summary, continued innovation in pyrolysis and its integration with other thermochemical techniques could be a game changer for advancing clean hydrogen production, paving the way for a greener and more energy-efficient future.

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