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Chemical Looping Systems for Hydrogen Production and Their Implementation in Aspen Plus Software: A Review and Bibliometric Analysis

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ABSTRACT

Hydrogen (H_2) production is a key strategy for reducing greenhouse gas emissions, providing a clean and efficient energy alternative. This review explores chemical looping combustion (CLC) for H_2 production, focusing on feedstocks, oxygen carriers (OCs), and process modeling in Aspen Plus®. A bibliometric analysis was conducted to support the review. Results indicate that methane (CH_4) outperforms biomass due to its higher efficiency and stable reaction behavior. Iron- and nickel-based oxides are the most effective OCs, with iron facilitating water splitting and nickel excelling in steam methane reforming (SMR) and chemical looping reforming (CLR). Enhancing OCs with support materials and sorbents improves system performance. Accurate simulations using Rgibbs and fluidized bed models are essential for optimizing the process. This study provides insights into improving H_2 production efficiency, contributing to advancements in clean energy technologies.

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1. INTRODUCTION

Currently, reducing greenhouse gas emissions is a critical challenge in achieving sustainable development goals, particularly in response to the urgent need to mitigate climate change and its consequences [1]. Fossil fuels, such as oil and coal, have significantly increased atmospheric CO_2 levels, rising from 310 ppm in 1960 to 419 ppm in 2021 [2]. This trend directly impacts global temperature rise, with CO_2 being the primary greenhouse gas, leading to environmental and health-related risks [3, 4]. Consequently, research efforts have intensified in recent years to develop emission-free energy alternatives, with hydrogen (H_2) emerging as a promising solution due to its clean combustion, high energy density, and capacity for storage and transport [5]. Unlike conventional fuels, H_2 combustion produces only water vapor and delivers more energy per unit mass than methane, gasoline, or coal [6]. Given these advantages, global demand for hydrogen is expected to reach 210 million tons by 2030, according to the International Energy Agency [6]. Meeting this growing demand necessitates the development of efficient and environmentally friendly production methods, where chemical looping presents itself as a viable and innovative approach.

Chemical looping technology operates through a dual-reactor system consisting of an air reactor (AR) and a fuel reactor (FR). In this process, a solid oxygen carrier (OC) transfers oxygen between the two reactors. Within the fuel reactor, the OC provides oxygen for combustion, undergoing reduction before being transported to the air reactor for reoxidation. This continuous cycle enables efficient oxidation-reduction reactions without direct contact between air and fuel [7]. One key advantage of this configuration is the high concentration of CO_2 and H_2O in the fuel reactor's exhaust stream. By condensing and removing H_2O, nearly pure CO_2 can be captured and stored, making CLC more efficient for carbon capture compared to oxy-combustion and traditional absorption methods, all while reducing energy and economic costs [8].

In the context of hydrogen production, process optimization plays a crucial role in enhancing efficiency, and computational tools are essential for this purpose. Among these, Aspen Plus® has become one of the most widely used process simulation software in chemical engineering and energy applications. This software enables the modeling of mass and energy flows across different unit operations, including chemical reactors, heat exchangers, and separation systems. In chemical looping, Aspen Plus facilitates the simulation and optimization of reactor configurations, helping to predict thermodynamic behavior under different operating conditions [8]. Its capability to model both steady-state and dynamic processes makes it suitable for simulating diverse reactor types, ranging from ideal models like Rgibbs to complex non-ideal and catalytic reactors, solid processing units, and fluidized beds [9].

Beyond standard process modeling, Aspen Plus offers additional functionalities such as support for optimization and case studies, the ability to adjust data, conduct economic, safety, and environmental assessments, as well as dynamic modeling and customization of models [10]. This versatility makes it an invaluable tool for designing and optimizing CLC systems for hydrogen production, allowing for the analysis of various feedstocks, including biogas and municipal waste. Additionally, the software includes configurations that adapt to user requirements with equipment that ranges from the principle of ideality, like the Rgibbs, to models that can be sized for experimental implementation as the fluidized bed and cyclone modules [8]. By integrating these features, Aspen Plus not only provides a detailed representation of reaction mechanisms and mass-energy balances but also facilitates the

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identification of efficiency improvements, reinforcing its role in the advancement of hydrogen production technologies [11].

Despite the existence of multiple process modeling alternatives, Aspen Plus stands out due to its extensive adoption in the field and its comprehensive capabilities. However, a detailed literature review revealed that no previous study has systematically covered all aspects of CLC modeling for hydrogen production within Aspen Plus, nor has it extensively analyzed the strategies implemented to improve simulation accuracy. Given the growing importance of precise modeling in this field, this study consolidates and evaluates recent advancements in Aspen Plus-based CLC simulation. To the best of the authors' knowledge, this is the first review to systematically compile and assess hydrogen production modeling techniques in Aspen Plus, offering a structured overview of best practices to enhance simulation reliability.

To strengthen this analysis, a bibliometric approach was integrated, allowing for an assessment of research trends in CLC for hydrogen production and providing a broader perspective on the evolution of this field. Additionally, this review compares different simulation methodologies and their alignment with experimental findings, an aspect that has not been extensively explored. Furthermore, it identifies key factors influencing process efficiency, including feedstock selection, oxygen carrier performance, and system configurations, offering insights for process optimization. By combining bibliometric techniques with an in-depth evaluation of simulation methodologies, this study highlights best practices identified in the literature while reinforcing the role of Aspen Plus in advancing hydrogen production technologies for sustainable energy applications. To ensure a comprehensive and systematic approach, relevant terms were carefully selected (see **Table 1**) to guide the literature review and evaluation process.

Table 1. Nomanclature.

Abbreviations and Nomenclature					
Al	Aluminum	**$FeAl_2O_4$**	Hercynite	**NiO**	Nickel oxide
Al_2O_3	Aluminum Oxide	**FluidBed**	Fluidized Bed	**Nox**	Nitrogen oxides
Ar	Air reactor	**Fr**	Fuel reactor	**O**	Oxygen
Ca	Calcium	**GWI**	Global Warming Index	**OC**	Oxygen carriers
Ca_2CuO_3	Dicalcium cuprate	**H_2**	Hydrogen	**Ppm**	Parts per million
CaO	Calcium oxide	**H_2O**	Water	**PR-BM**	Peng Rob-Boston matias
$CaSO_4$	Calcium sulfate	**H_2S**	Hydrogen sulfide	**RCSTR / RPlug**	Continuous Stirred Tank Reactor / Plug Flow Reactor
CFD	Computational fluid dynamics	**Hp**	High pressure	**Requil**	Equilibrium reactor
CGE	Cold gas efficiency	**HRSG**	Heat recovery and steam generator	**Rgibbs**	Gibbs reactor-equilibrium reactor
CH4	Methane(biogas)	**K**	Potassium	**RKS**	Redlich-Kwong-Soave
CLC	Chemical Looping Combustion	**Lp**	Low pressure	**Rr**	Reforming reactor
CLG	Chemical looping gasification	**MA-CLR**	Membrane-Assisted Chemical Looping Reforming	**Rstoic**	Stoichiometric Reactor
CLHG	Chemical looping hydrogen generation	**Mg**	Magnesium	**Ryield**	Yield Reactor
CLR	Chemical looping reforming	**$MgAl_2O_3$**	Magnesium aluminate	**SiO_2**	Silicon dioxide
Co	Cobalt	**$MgAl_2O_4$**	Spinel	**SMR**	Steam methane reforming

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Table 1 (continue). Nomanclature.

Abbreviations and Nomenclature					
CO2	Carbon dioxide	**MgO**	Magnesium oxide	**SMR-CLC**	Steam Methane Reforming with Chemical Looping Combustion
CoO	Cobalt oxide	**MHeatX**	Heat exchanger module	**SO_2**	Sulfur dioxide
CSTRs	Continuously stirred tank reactors	**Mn**	Manganese	**Sr**	Steam reactor
Cu	Copper	**Mn_2O_3**	Manganese(III) oxide	**SR**	Steam reforming
$CuAl_2O_4$	Copper Aluminate	**Mn_3O_4**	Manganese tetroxide	**TiO_2**	Titanium dioxide
$CuFe_2O_4$	Copper iron oxide	**MnO_2**	Manganese dioxide	**WGS**	Water gas shift
DR	Dry reforming	**MSW**	Municipal solid waste	**Zr**	Zirconium
FBMR	Fluidized Bed Membrane Reactor	**NH_3**	Ammonia	**ZrO_2**	Zirconium dioxide
Fe	Iron	**Ni**	Nickel		
Fe_3O_4	Iron oxide-magnetite	**$NiAl_2O_4$**	Nickel aluminate		

2. METHODS

2.1. Paper selection and bibliometric analysis

To ensure a comprehensive and structured review, the selection of articles was carried out using multiple scientific databases, with ScienceDirect and Scopus serving as the primary sources. The search strategy was designed to encompass key terms closely associated with chemical looping and hydrogen production, including "chemical looping," "Aspen Plus," "hydrogen production," "H_2," "oxygen carrier," "biogas," and "simulation." A fundamental criterion for inclusion was the explicit use of Aspen Plus as a modeling tool for chemical looping processes. However, recognizing the importance of experimental insights, studies that did not explicitly reference the software were also considered, as they provided valuable comparative data for understanding chemical looping combustion (CLC).

Following the initial selection, a second filtering stage was applied to prioritize studies specifically focused on H_2 generation through process modeling in Aspen Plus. This refinement aimed to highlight research directly aligned with the study's objectives without excluding relevant contributions. In June 2024, a final selection of 117 studies was retrieved from the selected databases. Covering a diverse range of feedstocks, system configurations, and process equipment, thereby ensuring a broad perspective on the field's advancements.

These records were subsequently downloaded and indexed in the Zotero reference manager to facilitate systematic reading and analysis.

To complement this selection, a bibliometric analysis was performed using VOSviewer (version 1.6.20) and the Bibliometrix package in RStudio. Key bibliometric parameters analyzed included author number and relevance, major sources of scientific output, and annual research output. Additionally, a co-occurrence analysis of keywords was conducted based on a modified version of the methodology described in [12, 13]. The dataset from ScienceDirect, Scopus, and Google Scholar was processed using an adapted analytical framework proposed by some researchers [14, 15]. This approach facilitated the evaluation

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of scientific impact, research evolution, and knowledge structure within the domain of chemical looping for hydrogen production [16].

Beyond the bibliometric evaluation, a systematic literature review was undertaken to provide an in-depth assessment of the feedstocks utilized, chemical looping methodologies for H_2 production, oxygen carriers performance, and Aspen Plus modeling approaches, particularly concerning their alignment with experimental studies. Moreover, notable findings and complementary process optimizations were examined to identify emerging trends and potential avenues for technological advancements in the field.

3. RESULTS AND DISCUSSION

3.1. Bibliometric Analysis

From 117 reviewed articles, the bibliometric analysis identified 48 distinct sources, with the International Journal of Hydrogen Energy, Fuel, and Energy Conversion and Management emerging as the most frequently cited journals, contributing with 17, 14, and 10 articles, respectively. Other relevant sources included Applied Energy, Energy and Fuels, and Fuel Processing Technology, each with 6 publications, while Chemical Engineering Journal, Energy, Renewable and Sustainable Energy Reviews, and Chemical Engineering Research and Design accounted for 5, 4, 4, and 3 articles, respectively.

Beyond the distribution of publications across journals, authorship analysis provided insights into the key contributors within this research domain. A total of 409 distinct authors contributed to the selected body of literature, highlighting their significant contributions to the advancement of chemical looping and hydrogen production research.

Examining the temporal evolution of publications, a notable upward trend was observed beginning in 2013, when the number of articles increased from an average of two publications per year to a range fluctuating between five and twelve annual publications. The highest scientific output was observed in 2020 and 2021, reaching 16 articles per year. However, as shown in **Figure 1**, a decline in publication rates is noticeable in subsequent years, particularly after 2021. The lower values reported for 2024 can be attributed to the fact that the data collection process was conducted mid-year, meaning the available records may not fully represent the total scientific output for this period.

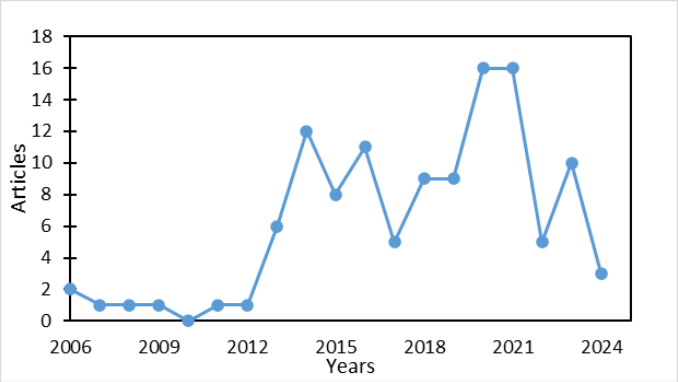

Figure 1. Annual scientific production.

As shown in **Figure 2,** a co-occurrence analysis of keywords was conducted to examine the conceptual structure of the selected literature. A minimum occurrence threshold of 18 was established as the selection criterion, resulting in the identification of 16 relevant keywords. Among these, the most prominent terms included “hydrogen production,” “chemical looping,” “oxygen,” “water splitting,” and “oxygen carrier,” with occurrence counts of 96, 76, 52, 54, and 45, respectively. The total link strength values for these keywords were 489, 391,

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314, 278, and 255, respectively, indicating the cumulative strength of connections between each keyword and all others in the dataset. As expected, higher co-occurrence frequencies corresponded to stronger link strengths, reinforcing the centrality of these terms [17].

This behavior is logical, given the study's focus on hydrogen production through chemical looping and its modeling in Aspen Plus. The high frequency of “hydrogen production” reflects its central role as the primary objective of this technology, while “chemical looping” appears consistently as the core process under review. The relevance of “oxygen” and “oxygen carrier” aligns with their key function in oxygen transfer, crucial for fuel conversion efficiency. Additionally, “water splitting” emerges as a significant term, as it plays a vital role in certain chemical looping configurations for H_2 generation. The prominence of these keywords underscores their interconnection and fundamental role in the research domain.

The second group of keywords, including "methane," "hydrogen," "synthesis gas," "steam reforming," and "carbon dioxide," with occurrences between 47 and 25, remains relevant but appears less frequently as these terms describe specific reactants, products, and processes within chemical looping rather than the overarching concept. Meanwhile, terms like "aluminum oxide," "hematite," "iron," "oxidation," "redox reactions," and "iron oxides," with occurrences below 25, are even less common, it because they pertain to the materials and reaction mechanisms of oxygen carriers, which, while essential, are discussed in a more specialized context within the broader analysis of the process.

Moreover, the color distribution in the graph highlights the period with the highest keyword frequency, revealing a predominant use of “hydrogen production,” “oxygen,” “iron oxides,” “oxygen carrier,” “carbon dioxide,”, “chemical looping”, “water splitting”, “redox reactions” and “synthesis gas” in 2020, followed by “steam reforming,” “aluminum oxide,” and “hematite” in 2021. This trend aligns with **Figure 1**, which identifies these years as those with the highest volume of scientific publications.

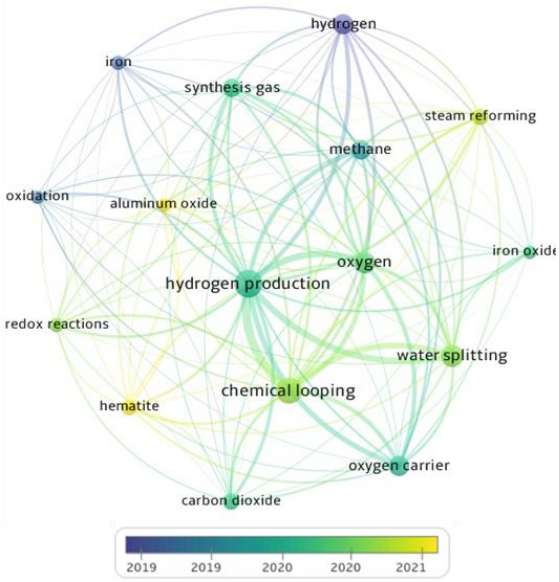

Figure 2. Keyword co-occurrence map.

3.2. Chemical Looping Hydrogen Generation

3.2.1. CLC Feedstock Alternatives

Historically, coal, natural gas, and syngas have been the primary feedstocks for chemical looping combustion (CLC) [7]. However, in recent years, biogas has gained attention as a preferred alternative due to its widespread availability [18]. The main sources of biogas

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include manure, agricultural waste, household organic waste, and sludge from wastewater treatment plants [19]. Biogas is produced through the anaerobic decomposition of organic matter, consisting mainly of CH_4 and CO_2 [20, 21]. Additionally, it can undergo reforming processes, such as steam reforming, to generate H_2, offering a more environmentally friendly alternative to conventional natural gas-based methods [22, 23].

One of the main challenges associated with biogas utilization is the presence of siloxanes, which can form gaseous compounds and Si-based particulates, leading to abrasion and wear in downstream components [24].

Biomass has also demonstrated strong potential as a renewable feedstock for H_2 production, maintaining cost-effectiveness and supply sustainability [25]. Its flexibility stems from the wide range of available sources, including crops, agricultural residues, wood and wood-derived waste, animal waste, municipal waste, and aquatic plants [26]. Biomass use surpasses coal in certain aspects, mitigating issues related to volatile compounds, sulfur emissions, and ash formation [27]. Compared to natural gas, biomass features lower depletion rates and costs [25]. Furthermore, its implementation in CLC systems has the potential to achieve carbon negativity, particularly in high CO_2-emitting industries, while also improving thermal efficiency, reducing NO_x formation, and minimizing corrosion in heat exchangers [28]. Despite these advantages, biomass presents challenges such as material deactivation, high solid recirculation requirements, and difficulty in separating looping materials from biomass ashes [29, 30]. Additionally, alkali metals and chlorine in biomass can contribute to corrosion and oxygen carrier agglomeration [31].

To address these issues while retaining the benefits of biomass, various utilization strategies have been explored, with gasification emerging as a prominent approach. Gasification reduces corrosion risks and ash accumulation while achieving a mass reduction of 70–80% and an 80–90% decrease in solid waste generation [32, 33]. Integrating biomass gasification with CLC has demonstrated promising results, enabling high-purity nitrogen flow and achieving near-theoretical H_2 and CO molar yields [34, 35]. Moreover, this integration reduces greenhouse gas emissions by approximately 75% compared to natural gas reforming [36]. However, syngas obtained from biomass gasification still contains significant impurities, posing additional processing challenges [37].

Other potential feedstocks discussed in the literature include ethanol and glycerol [38]. Ethanol, recognized for its high H_2 content and renewable production potential, can be derived from biomass fermentation. However, its large-scale implementation is constrained by the energy-intensive concentration processes required due to its limited natural occurrence [39]. Like CH_4, ethanol can serve as a fuel in CLC and be converted into H_2 through partial oxidation with oxygen carriers or steam reforming [40]. A study using a fixed-bed reactor with xCeNi/SBA-15 as the oxygen carrier reported ethanol conversion rates of 90% and H_2 selectivity of 84.7% [41].

Similarly, glycerol has been explored as a viable alternative, particularly as a byproduct of biodiesel production [42]. Despite challenges such as high viscosity and carbon deposition, glycerol can be utilized in reforming processes and exothermic partial oxidation. Although its application in chemical looping reforming (CLR) remains underreported, studies indicate promising efficiency levels, with H_2 selectivity ranging from 78% to 93% and reactant conversion rates between 90% and 99% [43, 44].

3.3. Methodologies for Hydrogen Production

Numerous methodologies integrating chemical looping principles with H_2 generation have been documented in the literature. The selection of a specific approach depends primarily on

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the feedstock type and the oxygen carrier (OC) utilized. Additionally, the intended application of the produced H_2, whether for industrial use or energy generation, plays a crucial role in determining the most suitable method. The following sections outline the principal techniques reported.

3.3.1. Steam reforming or water splitting

One of the most widely studied approaches is the three-reactor steam reforming or water-splitting system. This configuration consists of an air reactor (AR), a fuel reactor (FR), and a steam reactor (SR), along with an oxygen carrier (OC), as depicted in **Figure 3**, adapted from the literature [6]. Typically, metal oxides serve as OCs, with iron oxides, particularly Fe_2O_3, being the most frequently employed. However, due to thermodynamic constraints and relatively low redox activity, an AR is required to facilitate the oxidation of Fe_3O_4 back to Fe_2O_3, thereby improving oxygen transfer capacity [45, 46].

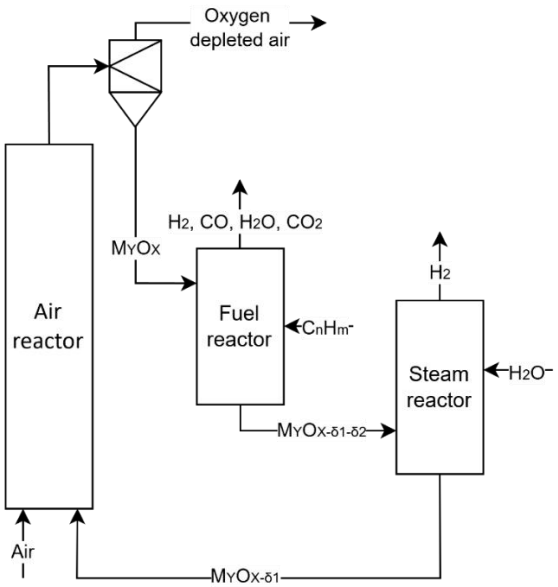

Figure 3. Three reactors chemical looping water splitting. Figure was adopted from the literature [6].

In this process, atmospheric air enters the AR, where it oxidizes the OC according to Equation (1). The oxidized OC is then transported to the FR, where it reacts with CH_4 to produce CO_2 and H_2O while undergoing reduction, as described in Equation (2). Subsequently, the reduced OC moves to the SR, where it interacts with steam, generating H_2 through reoxidation (Equation 3). Finally, the partially regenerated OC is cycled back to the AR, ensuring continuous operation [47–49].

Several variations of this method have been explored. A key alternative is the two-reactor chemical looping combustion (CLC) or water-splitting system, which simplifies the process by utilizing only the FR and SR, eliminating the AR. In this configuration, H_2 is produced via water decomposition using reduced metal oxides [6]. While the three-reactor system offers enhanced versatility by enabling simultaneous syngas production in the FR, it lacks the inherent CO_2 separation capability of conventional CLC systems. Nonetheless, this limitation can be mitigated by employing carbon-neutral biomass-derived feedstocks [46].

$$MyO_{x-1} + O_2 \rightarrow MyOx \quad (1)$$

$$MyOx + CH_4 \rightarrow MyO_{x-2} + H_2O + CO_2 \quad (2)$$

$$MyO_{x-2} + H_2O \rightarrow MyO_{x-1} + H_2 \quad (3)$$

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3.3.2. Steam Methane Reforming (SMR)

This approach focuses on biogas reforming without requiring a dedicated steam reactor, as illustrated in **Figure 4**, adapted from the literature [6]. The process operates similarly to chemical looping combustion (CLC), utilizing an air reactor (AR) and a fuel reactor (FR). Steam methane reforming (SMR) plays a central role, involving catalytic methane conversion at high temperatures (800–1000°C) and pressures ranging from 20 to 40 bar. This reaction produces syngas, a mixture of CO and H_2, as described by Equations (4) and (5). The resulting syngas is then cooled and subjected to a water-gas shift (WGS) reaction, which enhances H_2 yield by converting CO into CO_2 and additional H_2, as expressed in Eq. (6) [6, 50]. Given the endothermic nature of SMR, heat is typically supplied by combusting a portion of the fuel gas within the reforming reactor [51].

$$CH_4+H_2O \rightarrow CO+3H_2 \quad (4)$$

$$CH_4+2H_2O \rightarrow CO_2+4H_2 \quad (5)$$

$$CO+H_2O \rightarrow CO_2+H_2 \quad (6)$$

Although the fundamental principle remains consistent, SMR exhibits significant flexibility in its implementation. One approach integrates steam reforming within the fuel reactor by incorporating reforming tubes, leveraging the heat released in the combustion process to drive the endothermic SMR reactions. In this setup, WGS reactions typically occur in a separate reactor [31]. Alternatively, SMR and WGS can be performed externally, with a CLC system providing the necessary thermal energy while reforming takes place in a dedicated reactor [50].

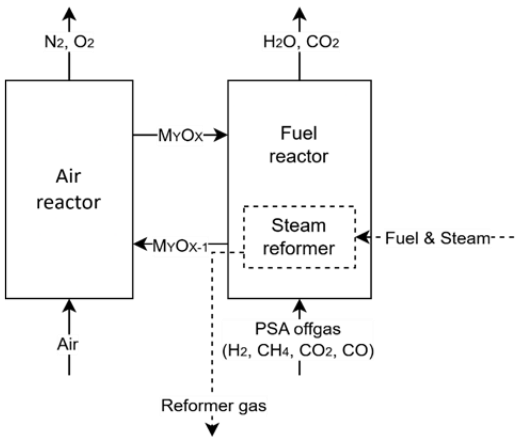

Figure 4. Steam Methane Reforming. The figure was adopted from the literature [6].

3.3.3. Chemical Looping Reforming (CLR)

The CLR technique closely follows the fundamental principles of CLC, relying on an air reactor (AR) and a fuel reactor (FR) without the need for a separate steam reactor, as depicted in **Figure 5**, adapted from literature [52]. This method utilizes an oxygen carrier (OC) that directly interacts with the fuel gas, facilitating H_2 production, as described in Equation (7). Additionally, Water-Gas Shift (WGS) reactions Equation (6) may also take place under certain conditions [53].

Among the various OCs explored in CLC-based systems, iron oxide is widely studied due to its balance between reactivity, mechanical stability, and high melting point [54]. However, nickel-based OCs are often preferred because they exhibit superior reactivity with methane (CH_4), enhancing H_2 yield and promoting WGS reactions more efficiently than iron oxides [2].

$$CH_4+NiO \rightarrow CO+2H_2+Ni \quad (7)$$

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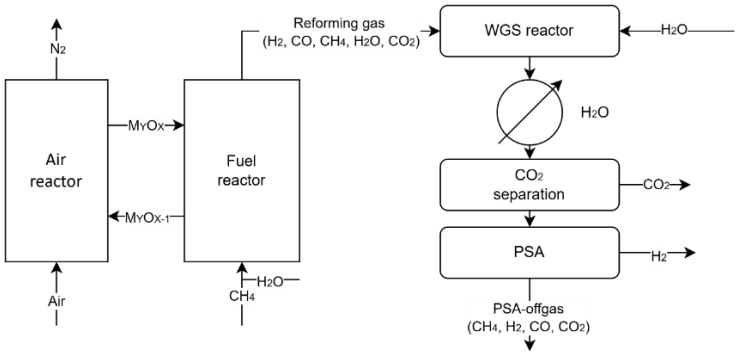

Figure 5. Chemical Looping Reforming. The figure was adopted from the literature [52].

3.3.4. Chemical looping Gasification (CLG)

This technique follows the same reactor configuration and operating principles as CLR but utilizes solid fuels, such as biomass, which undergo gasification in the fuel reactor. The process typically involves fluidization with gasifying agents like H_2O and CO_2 [55]. Unlike CLR, CLG often incorporates a carbon separation process within the oxygen carrier outlet stream in the fuel reactor, as illustrated in **Figure 6**, modified from the literature [52, 56].

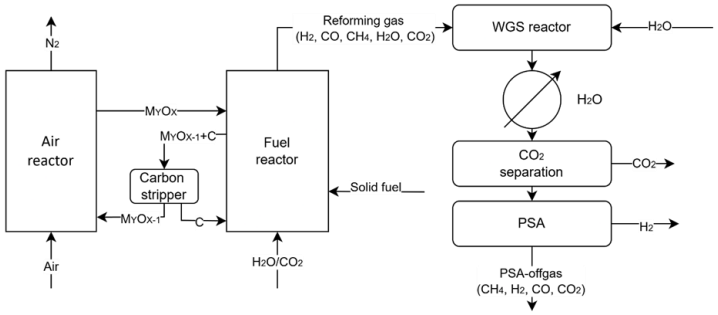

Figure 6. Chemical Looping Gasification. Data was adopted from the literature [52, 56].

CLG is considered a distinct approach because it fundamentally modifies both the process and the feedstock, setting it apart from other chemical looping methods. However, the core principle of gasification can be integrated into several previously discussed techniques. For instance, one study applied a water-splitting process for H_2 production using biomass as feedstock [55]. In this case, biomass was first gasified before entering the fuel reactor, with CO_2 and H_2O acting as enhancers. This approach aligns with the H_2 production principles of water splitting while modifying the fuel source.

3.4. Oxygen carriers (OC)

Oxygen carriers (OCs) play a fundamental role in chemical looping technology, serving as intermediaries between reactors to facilitate oxygen transfer from the air to the fuel [57, 58]. This process begins with the oxidation of the OC through its reaction with an oxidizing agent, such as air. The oxidized carrier then interacts with the fuel, undergoing a reduction reaction that transfers oxygen and releases energy [59]. This mechanism is crucial for H_2 production, as it not only enhances process efficiency but also helps reduce NO_x emissions [60].

Additionally, OCs contribute to efficient CO_2 capture by preventing direct contact between air and fuel. This allows the fuel reactor to generate nearly pure CO_2, which can be easily captured and stored, while the air reactor primarily emits reduced air [28]. These

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characteristics position chemical looping as a promising approach for clean and sustainable energy generation.

OCs are generally categorized into two main types: synthetic materials and natural minerals. Natural minerals are typically more affordable but tend to exhibit lower reactivity compared to synthetic OCs. Among the most studied materials in the literature are metal oxides of Fe, Ni, and Mn, often combined with inert supports such as TiO_2 and Al_2O_3 to enhance their stability and performance [2].

3.4.1. Fe based

Iron-based oxygen carriers (OCs) are among the most studied due to their affordability and favorable redox properties [2, 54]. They also exhibit high melting points and strong mechanical resistance, with the added advantage of not forming sulfides or sulfates. While they demonstrate good reactivity with H_2 and CO, their performance with methane is notably weaker [2, 61]. Despite this limitation, their widespread availability, lower production costs, and safer handling compared to materials like nickel and cobalt make them attractive for various applications. However, some Fe-based OCs may contain trace amounts of toxic elements such as chromium [61].

Among these carriers, magnetite (Fe_3O_4) stands out due to its high iron content (up to 72.4%), offering a cost-effective and environmentally viable alternative to synthetic OCs. Nevertheless, its use is constrained by the high operating temperatures (950°C) required for its reduction and by potential losses in reactivity and stability due to impurities [54]. Fe-based OCs have been studied for syngas production, showing that employing Fe_3O_4 in reduction-oxidation cycles reports a 99.8% recovery of the carrier and syngas with an H_2 content of 92.45 mol%. However, this process also promotes gasification reactions that increase CO_2 formation, which may affect syngas purity [37].

To enhance performance, Fe-based OCs are often combined with inert materials such as Al_2O_3, SiO_2, TiO_2, ZrO_2, $NiAl_2O_4$, and $MgAl_2O_4$, which help improve their reactivity, durability, and fluidizability [62]. For example, incorporating 70% $MgAl_2O_4$ as an inert support in Fe_2O_3-based OCs has been shown to not only enhance H_2O-to-H_2 conversion but also improve thermal stress management and temperature stability in solid phases [47]. Other studies have explored different OC compositions, such as Fe_2O_3/Al_2O_3, $Fe_2O_3/MgAl_2O_4$, and $Fe_3O_4/MgAl_2O_4/CaO$, with Fe_3O_4/CaO standing out for its ability to produce high-purity H_2 and efficiently convert methane. The optimal molar ratios for this system have been identified as Fe_3O_4/CH_4 = 1.7, CaO/CH_4 = 1.0, and H_2O/CH_4 = 1.9 [63].

Additionally, the influence of apparent density and OC composition has been evaluated using Fe_2O_3-TiO_2 mixtures. While coke formation remains a challenge, these studies highlight the importance of considering microstructural factors to optimize the overall performance of oxygen carriers [64].

3.4.2. Ni-based

Nickel-based OCs are widely used due to their affordability, high catalytic activity, excellent oxygen transfer capacity (0.21 g O_2/g NiO), and strong thermal stability [2, 65]. Their effectiveness in methane reforming is well-documented, particularly at temperatures above 900°C [66]. Unlike Fe-based OCs, nickel exhibits higher reactivity with CH_4, leading to increased H_2 yields, especially when combined with support materials. Literature reports the use of compositions such as 20 mol% NiO with 80 mol% $MgAl_2O_4$ and 40 wt% NiO with Al_2O_3 and ZrO_2, which enhance performance by improving dispersion and stability [67, 68].

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Beyond its role as an OC, nickel is extensively used as a catalyst, often in combination with other OCs such as Fe and Ce. In chemical looping combustion (CLC) for methane reforming, Ni-based materials have demonstrated a CH_4 conversion of 30% with a CO selectivity of 90%, a performance attributed to high Ni dispersion and the formation of NiCe and NiFe alloys, which create stable active sites [69].

Although Ni-based OCs are commonly used as the primary carrier, particularly in biogas reforming, their combination with Fe compounds has shown significant advantages. Nickel enhances catalytic activity and facilitates Fe_3O_4 reduction, improving overall system efficiency [70, 71]. Some studies indicate that incorporating a molar ratio of 3 Ni into the OC composition significantly increases H_2 production [72]. This improvement is primarily due to nickel's catalytic role in breaking C–C bonds during fuel conversion [73, 74].

However, Ni-based OCs also present challenges, including methane decomposition, which leads to carbon deposition (coking), affecting both activity and stability. Additionally, sintering causes Ni particles to agglomerate, reducing their surface area and effectiveness, which necessitates frequent regeneration or replacement, impacting process sustainability and cost-effectiveness [65]. Various mitigation strategies have been proposed, such as dry impregnation during catalyst preparation, selecting appropriate supports, adding small amounts of steam to the reactor, or incorporating alkali metals (K, Mg, Ca) to reduce coking [2, 75]. Another approach involves lowering the operating temperature to enhance catalyst stability and minimize sintering effects [53].

3.4.3. Mn-based

Manganese-based OCs are widely studied due to their low cost and non-toxic nature. While their oxygen transport capacity is lower than that of other metal oxides, they often outperform Fe-based OCs in specific applications. Among the different oxidation states of Mn, Mn_3O_4 is the most stable for chemical looping combustion (CLC), as MnO_2 and Mn_2O_3 tend to degrade at high temperatures. However, due to their relatively low reactivity, Mn-based OCs are commonly used with support materials such as ZrO_2 to enhance their performance [2, 76].

One notable example is the Mn_{40}/ZrO_2 system, which has been successfully applied in steam methane reforming (SMR) and dry reforming (DR), achieving an efficiency of 1 at the fuel reactor outlet, with CH_4 conversions of 0.2803 and 0.7275 in SMR and DR, respectively [77]. Similarly, Mn-based OCs supported on ZrO_2 have demonstrated significant improvements in chemical looping reforming (CLR) for CH_4. The presence of ZrO_2 enhances stability, reducing agglomeration and coke deposition, leading to a 2.95% decrease in coke formation. Additionally, Mn-based OCs have achieved methane conversions as high as 99.9% at 750°C. Studies indicate that a co-precipitation method followed by impregnation is particularly effective in preparing manganese oxide OCs supported on alumina with Zr promoters, improving both structural integrity and reactivity [2, 78].

Despite these advantages, the use of Mn_2O_3 as an OC has been associated with a slight increase in CO_2 concentration, and its overall performance remains lower compared to Fe_2O_3, which exhibits a higher power output [8].

3.4.4. Cu based

Copper-based OCs have gained attention in CLC due to their fast reaction kinetics and efficient redox performance with fuels. One of their key advantages is the ability to release gaseous O_2 during oxidation, allowing exothermic reactions in both oxidation and reduction steps, which eliminates the need for external heat supply in the reduction reactor [2].

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Additionally, their excellent reduction kinetics and high selectivity toward CO_2 and H_2O make them ideal for achieving complete CH_4 and solid fuel conversion [58].

From an economic standpoint, copper is a more affordable and less toxic alternative compared to other transition metals such as Co and Ni. However, a major limitation of Cu-based OCs is their tendency to agglomerate due to their relatively low melting point (1085°C). To mitigate this, binders such as SiO_2, TiO_2, ZrO_2, and $MgAl_2O_4$ are commonly added to improve stability and prevent sintering [58].

One notable application of Cu-based OCs is their use in the form of $CuFe_2O_4$ spinel for syngas production from municipal solid waste (MSW) through chemical looping gasification (CLG). This material functions both as an OC and a catalyst, offering thermal and magnetic properties that facilitate ash separation and enhance recycling efficiency. Studies indicate that optimal performance is achieved at 800°C, with an OC flow rate of 100 kg/h enabling the recovery of 67.85% of the initially introduced hydrogen as pure H_2 [11].

3.4.5. Other OCs

In addition to the commonly used metal oxides, cobalt- and calcium-based OCs have also been explored in chemical looping combustion (CLC). These materials can function individually, in combination, or as support components, with aluminum and magnesium being frequent stabilizing agents. Cobalt, for instance, is known for its high oxygen transport capacity and its ability to undergo oxygen uncoupling, particularly when used in its Co_3O_4 form, which provides the highest oxygen yield per mole of metal [2, 79]. However, despite these advantages, cobalt-based OCs remain among the least studied, likely due to environmental concerns and their high economic cost, which limit large-scale applications [2, 80, 81]. Even so, studies have reported promising results when cobalt is combined with supports like Al_2O_3, achieving methane conversion rates of up to 96.6% in continuous operations lasting 25 hours. Nonetheless, its mechanical fragility poses challenges for long-term stability [80].

A notable alternative is the combined use of CoO and $CaSO_4$, which enhances reactivity and increases CO_2 concentration. This formulation exhibits good sintering resistance, attributed to the inert nature of the $CaSO_4$ substrate, which also plays a key role in controlling sulfur emissions. Additionally, Ca-based OCs are attractive due to their high oxygen capacity and low cost, making them a competitive option [82]. However, the use of $CaSO_4$ as an OC presents challenges, including sulfur loss and incomplete fuel conversion due to thermodynamic limitations [83]. These issues can be mitigated by incorporating CaO as a sorbent during the CLC process, which helps capture sulfur-containing gases released during redox cycles [84]. Another strategy involves modifying $CaSO_4$ with active metal oxides such as Fe_2O_3, which improves its reactivity and stability. The inclusion of Fe_2O_3 promotes the formation of intermediate compounds like CaFeSO, which help prevent secondary sulfur release reactions [84-86]. Additionally, CaO has been widely studied for its dual role as a CO_2 sorbent in chemical looping reforming (CLR) and steam methane reforming (SMR), further broadening its application potential [37, 87, 88].

Another crucial component in CLC systems is the use of Al and Mg as support materials. One example is $MgAl_2O_4$, which is valued for its thermal, dielectric, and mechanical properties [89]. This material enhances surface properties and structural stability, inhibiting the formation of Fe-Al spinels that could negatively impact performance. When used as a support for Fe-based OCs, it has been shown to achieve H_2 purities exceeding 98% [90].

The incorporation of $MgAl_2O_4$ provides multiple advantages, including improved dispersion of the active phase, reduced particle agglomeration, and increased thermal and chemical

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stability. It also minimizes friction losses, while Al_2O_3-based supports reduce thermodynamic limitations by offering a larger reactive surface area, enhancing mechanical resistance and ionic permeability [91]. Studies on Al_2O_3-supported Fe_2O_3 in CLC processes have demonstrated efficient CH_4 combustion, leading to complete conversion into CO_2 and H_2O. However, after 75 hours of operation, a decrease in compression resistance and an increase in the attrition index were observed, suggesting a progressive reduction in mechanical strength. The estimated lifespan of the oxygen carrier was determined to be approximately 1,100 hours, with an attrition rate of 0.09 wt%/h [92]. Despite these benefits, Al_2O_3 can form aluminum spinels that reduce activity. To counteract this, the addition of alkali metals such as MgO promotes the formation of more stable aluminates like $MgAl_2O_4$, preventing undesirable spinel formation and maintaining long-term reactivity [93].

4. CHEMICAL LOOPING AND ITS MODELING IN ASPEN PLUS

4.1. Experimental Procedure

Due to the intrinsic nature of CLC processes, which rely on solid-gas interactions, bed reactors play a fundamental role in facilitating key operations such as reduction, oxidation, full and partial combustion, power generation, and heat production [94, 95]. Among these, packed bed reactors enable a wide range of gas-solid reactions, including both heterogeneous catalytic and non-catalytic processes [96].

Various bed reactor configurations have been explored for CLC applications, with fluidized, moving, and fixed beds being the most commonly reported. Notably, fixed-bed systems stand out due to their compact and straightforward design, allowing integration with a steam reformer within the reactor, thereby enabling H_2 production and purification with an efficiency of 73% [97]. Several studies have examined the performance of bed reactors, reporting H_2 purity levels exceeding 90% and carbon capture efficiencies superior to those achieved with sorption-based methods [98]. For instance, a chemical looping system combining a 10 kWth fixed bed reactor with a 3 MWth biogas digester has been tested at the laboratory scale, achieving a H_2 purity of 99.998% through precise optimization of operational parameters [5]. Similarly, a study employing a fixed-bed reactor with sintering-resistant iron oxide-based oxygen carriers (OCs) coated with a hetero-oxide layer demonstrated a H_2 yield close to the theoretical maximum (16.7 mmol/g-Fe_2O_3) and an approximate purity of 100% [99].

Although fixed-bed reactors are widely reported, other configurations such as moving and bubbling beds have also been evaluated, offering viable or even superior alternatives for chemical looping applications [100].

Notably, fluidized bed reactors have demonstrated enhanced performance as combustion reactors, characterized by uniform temperature and pressure distribution, higher methane conversion rates, greater carbon dioxide selectivity, and improved oxygen carrier utilization compared to fixed-bed systems [101]. Their ability to process low-grade, low-calorific-value fuels while maintaining operational flexibility makes them particularly suitable for applications such as circulating fluidized bed reactors. Additionally, they are compatible with advanced technologies, including high-temperature looping cycles for fossil fuel conversion and carbon capture systems [102]. Their superior heat transfer capabilities also help mitigate hotspot formation, further enhancing reactor stability and efficiency [103].

4.2. Aspen simulation

The high success rate of fluidized beds has established them as essential equipment for advancing chemical looping processes, both experimentally and through computational

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modeling. Aspen Plus, widely reported in the literature, is a key tool due to its versatility, scalability, and feasibility in simulating CLC processes [104]. It has been used extensively for biomass gasification and performance prediction of gasifiers under various operating conditions [105]. Beyond process simulation, Aspen Plus is recognized for the reliability of its results, as its simulations closely align with experimental data.

For instance, one study proposed a steam reforming approach integrating in situ CO_2 utilization, incorporating two intermediate reactors modeled with Rgibbs to enhance syngas generation [49]. A comparison with experimental results from prior studies demonstrated a strong correlation, with deviations of less than 1% in H_2 concentration, CO and CO_2 conversion, CO selectivity, and H_2 purity [45, 106]. The largest discrepancy, observed in CH_4 conversion (5.85%), was attributed to the idealized assumptions of the simulation, which neglected kinetic effects. Notably, this modeling approach achieved 100% H_2 purity and an energy efficiency of up to 90.54% when integrated with a heat exchanger network, alongside a process efficiency of 72.04%, underscoring the ease of emulation and the high efficiency and versatility derived from the use of Rgibbs [49].

The Rgibbs model is one of the most widely used in chemical looping simulations, particularly for fuel, steam, and air reactors. By minimizing Gibbs free energy, it effectively predicts equilibrium conditions, making it highly advantageous for studying the effects of parameters such as pressure and temperature [107]. Additionally, it requires minimal process description while delivering accurate outcome predictions [108]. Despite being an ideal model, numerous authors attest to the significant reliability of the results obtained compared to those achievable experimentally. An example of this is the ideal simulation of CLR models [53]. Obtaining results very close to those obtained experimentally under identical operating conditions [109]. Highlighting a CH_4 conversion and a H_2 purity with minimal deviations of -0.30% and 0.52%, respectively. Similarly, a high degree of agreement was observed in the fuel reactor flows, with deviations no greater than 10% for H_2O (-6.25%), H_2 (-6.01%), and CO_2 (9.09%), except for CH_4 (-16.7%) and CO (-12.5%).

Steam Methane Reforming with Chemical Looping Combustion (SMR-CLC) has also been validated against experimental data, achieving deviations within ±2.6%, except for CO (16.1%). Notably, H_2 predictions deviated by just 0.9% [110, 111]. Many researchers consider Aspen Plus a reliable tool for their studies [47, 112]. For instance, a thermodynamic model using iron oxide-based oxygen carriers was developed by applying mass and energy conservation to all CLR system components. The developed model was used to investigate the effect of various operating parameters such as mass flow rates of air, fuel, steam, and oxygen carrier, and the fraction of inert material on H_2 and CO_2 production and key reactor temperatures. The Rgibbs model was used for modeling fuel, steam, and air reactors. This configuration allowed them to achieve a vapor to H_2 conversion of 50.8% in the steam reactor [112].

Although Rgibbs is widely used due to its simplicity and accuracy, alternative modeling approaches, such as fluidized bed models, have gained attention. Unlike idealized equilibrium-based models, fluidized bed simulations incorporate reaction kinetics and hydrodynamic behavior, providing more accurate results, as summarized in **Table 2**, adapted from the literature [10]. Although its use is often less common because in many cases, not all the required information is available for its operation, the use of this block stands out over the others because it allows obtaining more accurate results. In contrast to traditional RStoic models, recent discoveries from the Fluidized bed model offer a deeper understanding of hydrodynamics, species concentration patterns, solid volume fraction distribution, distributor design, and reactor dimensions [10]. This achieves the ability to start from a simulation and

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provides the opportunity to obtain the required data to carry out the process experimentally. This block has been employed by some authors to simulate the combustion of syngas and methane in fluidized beds [9]. Simulating a bubbling bed as a Fr for a CLC process driven by syngas and CH_4, obtaining data that match experimental results obtained for the combustion of these compounds. The implementation highlights that this model takes into account the reactor's geometry, solid inventory, and air/fuel ratio, allowing for reactor sizing and thus estimating its thermal and economic efficiency, proving to be superior to other models like the Rgibbs. With a more precise and realistic prediction "within 2% of the experimental data for syngas combustion and 4% for methane combustion" [9]. The results from multiple authors who modeled systems using Rgibbs and fluidized beds, incorporating different oxygen carriers and modeling methods, are better evidenced in **Table 3**.

Although less reported in the literature, the modeling blocks Ryield, Rstoic, and Requil have also shown potential for being employed in the modeling of this type of process **Table 2**. However, it is important to highlight that not all reactors are used for the same function. For instance, the Ryield is used in initial processes such as gasification. This is attributed to the fact that for this equipment, it is possible to specify the output yield, as it can be in cases like the CLG, where the output can be based on the values established for H_2 and CO [37]. While equipment such as Rstoic and Requil, due to their capacity for reaction simulation, are better suited for simulating equipment such as shift reactors [31]. The use of these is observed for the simulation of the fuel reactor, where a Ryield is used jointly for biomass devolatilization and pyrolysis, Rgibbs for gasification, and a Rstoic for OC reduction reactions and further combustion [8]. The air reactor was also modeled as a single Rstoic reactor. Similar to the approach employed in previous studies, this block was used for the CO_2 reducer reactor and argon as well [63].

The Ryield reactor is also used for the raw materials to undergo a conversion, while conventionally simulating the air and fuel reactor with Rgibbs, also highlighting the use of Requil to carry out the WGS reactions [11].

Table 2. Aspen Plus models. Data was adopted from the literature [10].

Aspen Plus model	RStoic (Stoichiometric Reactor)	RYield (Yield Reactor)	REquil (Equilibrium Reactor)	RGibbs (Gibbs Reactor)	RCSTR / RPlug (Continuous Stirred Tank Reactor / Plug Flow Reactor)	FluidBed (Fluidized Bed)
Objective	To simulate a reactor with a predetermined stoichiometry and a specified conversion rate or reaction extent for each reaction considered.	To simulate a reactor by defining the product yield distribution per unit mass of the feed stream.	To simulate a reactor with known stoichiometry by calculating the equilibrium extent of each reaction or defining an equilibrium approach.	To simulate a reactor at equilibrium or with a defined approach to equilibrium without specifying the reaction stoichiometry.	To model an ideal CSTR or PFR by defining reaction stoichiometry, kinetics, and reactor dimensions.	To simulate a fluidized bed reactor by defining reaction stoichiometry and kinetics while integrating detailed hydrodynamics based on reactor design

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Table 2 (continue). Aspen Plus models. Data was adopted from the literature [10].

Aspen Plus model	RStoic (Stoichiometric Reactor)	RYield (Yield Reactor)	REquil (Equilibrium Reactor)	RGibbs (Gibbs Reactor)	RCSTR / RPlug (Continuous Stirred Tank Reactor / Plug Flow Reactor)	FluidBed (Fluidized Bed)
The reaction stoichiometry must be specified.	Yes	No	Yes	No	Yes	Yes
The reaction kinetics must be defined (rigorous model)	No	No	No	No	Yes	Yes
The model incorporates fluidized bed hydrodynamics.	No	No	No	No	No	Yes

4.3. Notable Findings and Supplementary Process

A key area of research in chemical looping technology focuses on optimizing process efficiency and improving simulation models, particularly for H_2 production and CO_2 capture. Many studies employ idealized equipment due to its ease of modeling and reliability, enabling rapid simulations and adjustments to process variables. For instance, a steam reactor with a counter-current moving bed was modeled using the Rgibbs approach, demonstrating how multiple stages can enhance H_2 production [113]. Another study emphasized that incorporating three gas separation modules into the Rgibbs system significantly improved vapor conversion, reaching 84.45%, the highest reported efficiency, while achieving a cold gas efficiency (CGE) of 79%, a 7% improvement over conventional setups. The study also highlights that continuous H_2 removal, following Le Chatelier's principle, enhances overall process efficiency. In this context, multi-stage configurations have been employed to approximate experimental results, as seen in counter-current moving bed reactors for syngas conversion, effectively simulated using a five-stage Rgibbs model [48]. This approach has also been applied in simulating moving bed reactors, integrating unit reactors for oxidation and combustion [55].

Beyond these applications, process simulations have explored different feedstocks, including biomass and syngas, as well as the role of H_2O and CO_2 as gas enhancers in reduction reactors. Some models even incorporate a Haber-Bosch process for NH_3 generation. Notably, the highest efficiency in H_2 production was achieved using a dual chemical looping (DCL) system with CO_2 as an enhancer, attaining a 51.8% conversion rate, a net energy efficiency of 0.65%, and NH_3 efficiency of 38.09%. These improvements are largely attributed to the multi-stage reduction reactor design [55].

This multi-stage principle is also applied in carbon-based feedstock processes, where increased residence time enhances combustion completeness and energy release. Studies indicate that multiple reactors can be employed without negatively impacting net energy output [114].

Aspen Plus has proven to be a versatile tool for evaluating various process configurations. Some authors have explored different system arrangements, such as single- and two-stage

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setups, integrating a sorption process with CaO as a CO_2 sorbent [66]. In the single-stage system, a CLC configuration is used for thermal energy generation, which drives the reforming process and facilitates CaO regeneration. In the two-stage system, the reforming process remains unchanged, but the CLC operates with a high-pressure (Hp) and low-pressure (Lp) system. The Hp system supplies heat to the reformer while also feeding the Lp system, which, in turn, provides heat to the calciner. As a result, CH_4 conversion rates of 69.75% and 70.92% were achieved for the single- and two-stage plants, with exergetic efficiencies of 67.50% and 73.25%, respectively. The study also identified optimal molar ratios, including Ca/C (1.12), S/C (4.2), and specific fuel to carbon ratios for different pressure stages (FuelHP/C = 0.42, FuelLP/C = 0.70, Fuel/C = 0.9) [66]. In a related study, the same authors reported that an H_2 purity of 92.6% was obtained with S/C (4) and Ca/C (1) under a reforming pressure of 25 bar [88]. The integration of sorption processes was highlighted as a key factor in CO_2 removal, further enhancing system efficiency.

Sorbent-based processes are widely explored in reforming applications, with CaO being the most studied due to its reversible carbonation reaction, which enables cost-effective CO_2 capture [87, 115]. Studies suggest that incorporating CaO into SMR systems improves steam reforming efficiency, enhances H_2 concentration, and increases overall energy performance [109, 116]. Additionally, its presence in reforming reactors promotes CO conversion, leading to higher H_2 purity [117]. One example is an SMR process coupled with sorption-enhanced reforming for H_2 production. This system is divided into two zones: the "CLC" section, which generates thermal energy for CH_4 reforming, and the "SESMR" section, where a $CaO/CaCO_3$ loop continuously captures CO_2 and regenerates the sorbent Equation (8). This process enables CH_4 conversion rates of 97%, H_2 purity of 98.3%, and CO_2 capture efficiency of 96%. The study also identified an optimal material composition of SiC/Al_2O_3 (30%) and $MgAl_2O_4$ (40%) [118].

$$CaO + CO_2 \leftrightarrow CaCO_3 \quad (8)$$

Similarly, a comparative analysis of CH_4 reforming assessed two configurations: one with and one without sorption coupling [53]. The study found that in chemical looping processes, a maximum H_2 yield of 2.5 moles per mole of CH_4 was achieved at 700°C with a steam/CH_4 ratio of 3. In contrast, the sorption-enhanced process attained 99.5% H_2 purity at 500°C, with yields reaching 2.95 moles per mole of CH_4, demonstrating the effectiveness of CO_2 adsorption via CaO. The study emphasized that key factors for achieving high H_2 purity and yields include the NiO/CH_4 ratio, steam/CH_4 ratio, and system temperature. Optimal parameters were identified as 800°C for the CLR system, with H_2O/CH_4 = 3 and NiO/CH_4 = 1. Meanwhile, the sorption-enhanced system operated within 500–600°C, with $NiO/CH_4 \approx 1$, $CaO/CH_4 \geq 1$, and $H_2O/CH_4 \geq 2$ [53].

Although Ni-based OCs dominate sorption-enhanced processes, some studies have explored composite materials combining CaO and Fe_3O_4, eliminating the need for support materials. This composite enables partial CH_4 oxidation while achieving high efficiency, with exergetic efficiency of 70.6%, CO_2 utilization efficiency of 94.62%, H_2 purity of 98.55%, and CH_4 conversion of 99.24%. Notably, key process parameters include steam/CH_4 (1.9), Fe_3O_4/CH_4 (1.7), CaO/CH_4 (1.0) in the reduction reactor, and CO_2/OC (0.29) in the CO_2 reduction reactor [63]. Additional details on these results are provided in **Table 3**.

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Table 3. Chemical Looping Combustion Aspen simulation review articles.

Feedstock	Method	Technique	Simulation blocks	OC	Authors' Key Findings	Ref.
Natural gas	Redlich-Kwong-Soave (RKS) equation of state with Boston-Mathias	Water splitting	Rgibbs, cyclone, separators	Fe_2O_3+$MgAl_2O_4$	An increase in steam and oxide flow rates reduces H_2 production, whereas a higher fuel flow enhances it.	[47]
Natural gas	Redlich-Kwzong-Soave (RKS) equation of state with Boston-Mathias	Water splitting	Rgibbs, separators	Fe_2O_3+$MgAl_2O_4$	H_2 production initially increases as air, fuel, and steam flow rates rise but stabilizes beyond a certain threshold. Similarly, H_2 generation improves with higher oxide flow rates and inert material fractions up to a specific point, after which it begins to decline. In the base case, steam conversion to H_2 in the steam reformer (SR) reaches 50.8% by volume. The fuel reactor (FR) assumes complete fuel conversion, yielding 36.3% CO_2 and 63.44% H_2O.	[112]
CH_4	Not specified	Water splitting (CLHG)	Rgibbs, separators	Fe_2O_3	A vapor-to-H_2 conversion rate of 84.45% was achieved. The implementation of staged separation allowed exceeding the equilibrium limit for H_2 conversion in the steam reformer (SR), reaching conversion rates 55.49% higher than those of the original reactor	[48]
Biomass and biomass gasification	Peng-Rob	Water splitting (CLHG)	Rgibbs, separators, Heatx,pump, compressor	Fe_2O_3+Al_2O_3 (70:30)	Using DCL with CO_2 as a gas enhancer in the reduction process, the maximum achievable efficiencies were 51.8% for H_2, 38.09% for NH_3, and 0.65% for net power.	[55]
CH_4	PR-BM	Water splitting (CLHG)	Rgibbs, separators, Heatx	Fe_2O_3	The optimal temperatures for the five reactors were sequentially set at 900, 900, 850, 500, and 500°C, all operating at 1 atm. This configuration enabled the production of high-purity H_2 (100%) and syngas with purities of 99% and 93%, achieving an ideal H_2/CO ratio of approximately 2. The system's energy and exergy efficiencies reached 90.54% and 72.04%, respectively.	[49]
CH_4	PR-BM	Water splitting (CLHG)	Rgibbs, HeatX,Rcstr, pump, turbine, compressor	Fe_2O_3	Compared to a separate production system, this process is expected to achieve a fuel energy saving ratio of 12.19% and a CO_2 emission reduction of 98.46%. However, the highest exergy destruction is predicted to take place in the chemical looping H_2 generation unit, representing 37.56% of the total exergy loss.	[119]

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Table 3 (continue). Chemical Looping Combustion Aspen simulation review articles.

Feedstock	Method	Technique	Simulation blocks	OC	Authors' Key Findings	Ref.
Coal	Not specified	Water splitting (CLHG)	Rgibbs, cyclone, turbine, HeatX, compressor, flash	Fe_2O_3 +CuO 90:10)	The performance of all scenarios was assessed based on four key criteria: energy, exergy, environmental impact, and economics, complemented by a life cycle analysis (LCA). A sensitivity analysis was conducted to optimize the operating conditions of CLC reactors. The levelized cost of H_2 from a CDCLC plant, which generates 50% H_2 and 50% power, was estimated at 1.94 €/kg. The LCA of the cogeneration system identified the CDCLC stage as the primary contributor to greenhouse gas (GHG) emissions and primary fossil energy consumption (PFEC). The study concluded that the CDCLC power plant, with higher H_2 cogeneration, demonstrates efficiency in terms of energy use, exergy, environmental impact, and economic performance.	[120]
CH_4	Peng–Robinson	Chemical looping hydrogen generation (CLHG) and Chemical looping with water–gas shit reaction (CLWGS)	Rgibbs	Fe_2O_3	The study indicates that ultra-high-purity H_2 (above 99.997%) can be generated as a zero-emission energy carrier. Additionally, the system enables the co-production of high-purity carbon dioxide (99%) and nitrogen (98.5%) within the world's largest fixed-bed chemical looping research facility. With a feedstock utilization rate of up to 60% for pure H_2 production, this approach is highly competitive compared to other decentralized methods, while also facilitating inherent CO_2 sequestration. When powered by renewable sources like biogas, the process has the potential to qualify as a negative emission technology (NET), provided that CO_2 is effectively managed.	[97]
CH_4	PR-BM	Steam Methane Reforming (SMR) and chemical looping hydrogen generation (CLHG)	Rgibbs, HeatX, compressor, turbine, pump	Ni and Fe_2O_3	The CLHG system achieves higher energy and exergy efficiencies, reaching 80.95% and 76.22%, respectively, compared to 77.91% and 73.26% in the CLC-SMR system. This advantage is primarily due to the CLHG system's greater net energy output. Additionally, it has a lower Global Warming Index (GWI) of 52.75 kg/MWh, whereas the CLC-SMR system registers 54.91 kg/MWh. Economically, the CLHG system is also more favorable, with H_2 production costs of $32.87/MWh, reducing expenses by $8.61/MWh compared to the CLC-SMR system, highlighting the efficiency of the steam generation approach.	[50]

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Table 3 (continue). Chemical Looping Combustion Aspen simulation review articles.

Feedstock	Method	Technique	Simulation blocks	OC	Authors' Key Findings	Ref.
CH_4	PR-BM and STEAM-TA (in the HeatX)	Sorption enhanced steam methane reforming (SESMR)	Rgibbs, HeatX, compressor, cyclone, splitter, pump	NiO (30%-SiC/Al_2O_3 and 40% $MgAl_2O_4$)	The purity of H_2 and the amount of CO_2 captured increase as the inert composition rises, whereas H_2 yield decreases inversely. The optimal inert composition has been identified as 30% by weight for SiC/Al_2O_3 and 40% for $MgAl_2O_4$. In both cases, the overall performance of the CLC-SESMR system remains nearly unchanged, achieving 97% methane conversion, 96% CO_2 capture, 98.3% H_2 purity, an H_2 yield of 2.24, and a net plant efficiency of 71.4%.	[118]
Natural gas	Not specified	Sorption enhanced steam reforming (SE-SR)	Rgibbs, HeatX, compressor	NiO and CaO (sorption enhanced process)	The addition of a Ca-based sorbent and steam to the reformer positively impacts system performance. Under operating conditions of FuelHP/C = 0.42 and FuelLP/C = 0.70, the advanced process achieves an H_2 concentration of 92%, an H_2 yield of 3.64, and a methane conversion of 75%. In comparison, the novel process reaches an H_2 concentration of 89%, an H_2 yield of 3.55, and a methane conversion of 69%. The optimized approach reduces exergy destruction and enhances exergy efficiency by more than 6% compared to the base-case system.	[66]
CH_4	PR-BM	Sorption enhanced steam methane reforming (SESMR)	Rgibbs, HeatX, Compressor, pump, separator	NiO and CaO (sorption enhanced process)	This configuration provides substantial benefits compared to the conventional industrial SMR process. It enhances H_2 purity to 92.6% without requiring additional impurity removal steps and significantly reduces CO and CO_2 concentrations from 0.2% and 2.4% to ppm levels. In terms of overall energy efficiency, the SMR process achieves 68.02%, whereas the proposed approach reaches 85.50%, reflecting a 17.48% improvement attributed to the innovative design.	[88]
CH_4	Peng-Rob and PR-BM	Chemical looping combustion-assisted and chemical looping methane reforming technology (CLC-CLRHC)	Rgibbs, Rstoic, heater, mixer, HeatX, compressor	Fe_2O_3/$MgAl_2O_4$ and CaO (sorption enhanced process)	In the chemical looping methane reforming process (CLRHC), the optimal molar ratios for Fe_3O_4/CH_4, CaO/CH_4, and H_2O/CH_4 are 1.7, 1.0, and 1.9, respectively. Additionally, for chemical looping combustion (CLC), the required supplementary CO_2 to oxygen carrier ratio should be 0.29. To ensure heat balance between CLRHC and CLC, the air preheating temperature before the CLC air reactor must exceed 306°C. Under these conditions, the system achieves an H_2 purity of 98.55%, an H_2 yield of 2.26 mol/mol, a CO purity of 96.18%, a CO yield of 1.7 mol/mol, and a methane conversion rate of 99.24%. The system also reaches an exergy efficiency of approximately 70.60% and a CO_2 utilization efficiency of up to 94.62%.	[63]

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Table 3 (continue). Chemical Looping Combustion Aspen simulation review articles.

Feedstock	Method	Technique	Simulation blocks	OC	Authors' Key Findings	Ref.
CH_4	Peng–Robinson and Steam Tables	Steam methane reforming (CLC-SMR)	Rgibbs (five stage RGibbs reactor)	Fe_2O_3	Chemical looping technology demonstrates an improvement of over 5 percentage points in cold gas efficiency and more than 6 percentage points in effective thermal efficiency compared to the conventional steam methane reforming process. Furthermore, it enables H_2 production from natural gas with a carbon capture rate exceeding 90%.	[113]
Natural gas	Peng-Robinson	Steam methane reforming (SMR)	Rgibbs	Mn-based perovskite oxygen carrier material, CaMn0.775Mg 0.1Ti0.125O3-d	The results compare H_2 production efficiency and levelized cost of hydrogen (LCOH) between the two plants. In the proposed process configuration, production efficiency is over 11% higher than the alternative, while LCOH is reduced by more than 7%.	[121]
CH_4	Redlich-Kwong-Soave	Steam methane reforming with chemical-looping combustion (CLC-SMR) and oxygen carrier aided combustion (OCAC-SMR)	Rgibbs, Rstoic, pump, heater, cyclone, Requil	Fe, Mn based	The OCAC-based system improves cold gas efficiency relative to a conventional reference process while simultaneously lowering CO_2 emissions by 4%. In contrast, the biomass-fueled CLC configuration achieves significantly negative CO_2 emissions, reducing CO_2 output by 151% compared to the reference case.	[31]
Natural gas	Peng-Robinson	Steam methane reforming (CLC-SMR)	Rgibbs, cyclone, HeatX	Mn-based perovskite oxygen carrier material, CaMn0.775Mg 0.1Ti0.125O3-d	The first system achieves net-zero emissions and an H_2 production efficiency of 76.2%, nearly 8% higher than the conventional process. Additionally, its levelized production cost is 1.6% lower, remaining below 2.6 €/kg H_2. The second system has the potential to reduce emissions to 34.1 g CO_2/MJH_2, significantly lower than the 80.7 g CO_2/MJH_2 emitted by the conventional plant. Its H_2 production efficiency exceeds 72%, about 2% higher than the conventional process. However, this system requires greater capital investment, with a levelized H_2 production cost of approximately € 2.67/kg. In both cases, the cost of CO_2 avoidance remains relatively low compared to a reference SMR plant with CO_2 capture, estimated at 4.3 €/ton CO_2 for case CM and 2.7 €/ton CO_2 for case CB.	[67]

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Table 3 (continue). Chemical Looping Combustion Aspen simulation review articles.

Feedstock	Method	Technique	Simulation blocks	OC	Authors' Key Findings	Ref.
CH_4	Peng-Robinson, NRTLand SOLIDS	Chemical looping reforming (CLR)	Rgibbs, HeatX, and cyclone	NiO	In conventional CLR, optimal reactor conditions include a temperature of 800°C, an H_2O/CH_4 ratio of 3, and a NiO/CH_4 ratio of 1. Under these parameters, the process produces approximately 2.5 moles of H_2 per mole of CH_4, with an H_2 purity of around 75%. However, integrating in situ CO_2 adsorption can enhance H_2 purity beyond 90% and increase the yield to approximately 3 moles of H_2 per mole of CH_4. This improvement is achieved with a NiO/CH_4 ratio close to 1, a CaO/CH_4 ratio of at least 1, an H_2O/CH_4 ratio of at least 2, and an operating temperature between 500°C and 600°C. The adsorption of CO_2 plays a crucial role in significantly boosting both H_2 yield and purity.	[53]
Natural gas	Not specified	Membrane-assisted chemical looping reforming (MA-CLR) and fluidized bed membrane reactor (FBMR)	Fluidized bed reactor model, HeatX	NiO	The analysis indicates that both novel concepts achieve higher H_2 yields than conventional plants, with increases between 12% and 20%. The high electricity demand in membrane-based systems is primarily due to the need for low pressure on the retentate side. However, the lower energy cost associated with CO_2 separation and compression results in an overall reforming efficiency that is 4% to 20% greater than conventional fired tubular reforming (FTR) with CO_2 scrubbing. In terms of cost, FBMR and MA-CLR outperform FTR with CO_2 capture technology, mainly due to lower capital expenditure (CAPEX). As a result, H_2 production costs decrease from 0.28 €/Nm^3H_2 to 0.22 €/Nm^3H_2 for FBMR and 0.19 €/Nm^3H_2 for MA-CLR.	[122]
CH_4	Not specified	Membrane-Assisted Chemical Looping Reforming (MA-CLR)	Mixers, splitters, reactors, heat exchangers	NiO + $MgAl_2O_4$	Compared to other systems, MA-CLR achieves greater reforming efficiency at relatively low temperatures, which are still higher than those in conventional processes, ensuring effective fuel conversion into the desired products. In contrast, processes like SMR-CLC require burning part of the fuel to generate heat, which lowers overall reforming efficiency.	[110]
CH_4	Not specified	Membrane-Assisted Chemical Looping Reforming (MA-CLR)	Rgibbs	NiO	A continuous chemical looping reactor was used to evaluate the impact of different operating conditions, including temperature, steam-to-carbon ratio, and oxygen supply. Methane conversions exceeded 90%, with ultrapure H_2 successfully recovered through membranes. In all cases, the maximum recovery factor was approximately 30%. Optimal performance was achieved at around 600°C, with a steam-to-carbon ratio of 3 and diluted air containing 5% O_2 in the air reactor. The full-scale demonstration involved feeding up to 1 L/min of CH_4 (equivalent to 0.6 kW of thermal input) and recovering up to 1.15 L/min of H_2.	[123]

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Table 3 (continue). Chemical Looping Combustion Aspen simulation review articles.

Feedstock	Method	Technique	Simulation blocks	OC	Authors' Key Findings	Ref.
Biomass	Not specified	Chemical looping gasification (CLG)	Rgibbs, Ryield, heater	Fe_2O_3 and CaO (sorption-enhanced process)	The CLG simulation produced the highest absolute amount of syngas, reaching 2.54 kmol/kmol PL compared to 0.79 kmol/kmol PL. Meanwhile, the CO_2 capture simulation yielded a significantly higher H_2-rich syngas composition, with 92.45 mol% H_2 compared to 62.94 mol% H_2.	[37]
Municipal solid waste (MSW)	The Redlich-Kwong-Soave (RKS)	Chemical looping gasification (CLG)	Ryield, rstoic, rgibbs, tequila, heater, separator, compressor	$CuFe_2O_4$	At 750°C, H_2 selectivity peaked at 51.44%. As the SMR increased from 0.1 to 2.1, H_2 selectivity rose from 43.89% to 56.29%. Lower OCMF favored H_2 production. At 800°C, with an SMR of 1.1 and an OCMF of 100 kg/h, 67.85% of the initial H_2 introduced in the CLG process was recovered as pure H_2, achieving an energy efficiency of 57.66%, considering the low heating value (LHV) of MSW, electricity consumption, and H_2. The process required 7.93 tons of MSW per ton of H_2 produced, with a resulting product cost of 2941.21 USD/ton. Additionally, it accounted for 66.90% and 66.73% of the total global warming and acidification potentials of the process, respectively. While both OC and steam contribute oxygen during CLG, their effects on gasification efficiency differ due to their varying oxidation properties. Notably, using $CuFe_2O_4$ as the oxygen carrier, the maximum H_2 selectivity of 63.30% was obtained at 700°C, with an SMR of 2.1 and an OCMF of 100 kg/h.	[11]
Syngas	RKS-BM	CLC	Rstoic, splitter, heater, compressor, cyclone, Fluidized bed reactor model	Fe_2O_3 + Al_2O_3	The findings of this study suggest that using the Fluidized Bed Reactor model in Aspen Plus provides additional design insights and serves as a valuable alternative to the widely adopted Stoichiometric reactor approach.	[10]
Biomass+coal	Not specified	Chemical Looping Combustion (CLC)	Ryield,rgibbs,rstoic, cyclone, heater	Fe_2O_3 and Mn_2O_3	Replacing Fe_2O_3 with Mn_2O_3 resulted in a slight reduction in CO and H_2 concentrations in the fuel reactor, while CO_2 levels increased slightly. Additionally, a mixture of coal and biomass was tested at 895°C with both oxygen carriers. The results showed that the system using Fe_2O_3 generated a higher power output than the one with Mn_2O_3. Furthermore, increasing the coal fraction in the coal-biomass mixture led to a further rise in power output.	[8]

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5. FUTURE DIRECTIONS AND DISCUSSION

Among the various feedstocks used in CLC processes, CH_4 is the most extensively documented. It is primarily employed as fuel in the fuel reactor (FR) and steam methane reforming (SMR) processes, either as pure methane or as a component of biogas (60%) and natural gas (79.75–89%) [47, 66, 112, 124]. CH_4 demonstrates high efficiency in H_2 production, both as a direct fuel and as a reforming gas, particularly when used with Fe-Ni-based oxygen carriers (OCs) [49, 66]. While natural gas achieves higher H_2 yields (4.2%), biomass remains a more cost-effective alternative [67]. Future research should focus on improving CH_4 efficiency, either by blending it with other feedstocks such as coal or by enhancing the role of CO_2 as a reactive species [8, 55]. Although CH_4 is currently the most efficient option, feedstock selection ultimately depends on factors such as cost, environmental impact, process requirements, and resource availability.

Fe- and Ni-based metallic oxides are the most commonly used oxygen carriers in CLC. Fe-based OCs are preferred for water-splitting applications due to their lower cost, whereas Ni-based OCs are favored in CLR due to their strong affinity for methane. While individual OCs have demonstrated significant success, combining them with support materials can enhance H_2 conversion efficiency and mitigate operational challenges such as agglomeration, coke formation, and sintering [47, 78, 84]. In CLR systems, incorporating sorbent materials like CaO has been particularly beneficial, increasing H_2 production [53] and addressing sulfur-related issues [84]. However, optimizing key ratios such as OC/support, OC/fuel, sorbent/fuel, and H_2O/fuel is essential to ensure the viability of these modifications.

The Rgibbs reactor model is widely used for simulating CLC processes, successfully modeling both reactors and complex equipment such as fluidized and countercurrent beds. This approach simplifies calculations while providing reliable results. However, Rgibbs is primarily suited for equilibrium-based simulations and is not optimal for scaling models, as experimental validation of such reactors is impractical. Therefore, researchers are encouraged to use fluidized bed reactor models, which also provide high reliability and facilitate equipment design and scale-up [9, 49].

CLC processes for H_2 production can be classified into three methodological approaches based on study objectives. The base methodology follows fundamental system operation principles, including core reactions, feedstock type, and essential equipment. While useful for initial process design, it may not be the most efficient approach. More advanced methodologies focus on optimizing H_2 generation by refining operational conditions, adjusting process configurations, and incorporating additional equipment and defining parameters such as relationships or many stages [48]. A third approach prioritizes models with high levels of energy and exergy efficiency, generally focusing on the use of residual elements and energy, employing equipment like heat exchangers, or incorporating additional processes such as sorption or syngas generation.

Both water splitting and reforming are viable strategies for H_2 production, yet their relative advantages are difficult to establish due to fundamental differences in operating principles. Comparative analyses are scarce, making direct evaluation between studies challenging. However, a comparison between a conventional steam methane reforming (SMR) system (**Figure 7**) and a chemical looping hydrogen generation (CLHG) system (**Figure 8**) demonstrated that CLHG achieved 3.04% higher efficiency than CLC-SMR while reducing the Global Warming Index (GWI) and lowering H_2 production costs by $8.61/MWh [50]. This study employed base methodology, using only essential process equipment, whereas most literature reports propose modifications that enhance efficiency (**Figures 7–8**). Consequently,

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while water-splitting-based systems may appear superior under baseline conditions, subsequent advancements, such as multi-stage configurations and energy recovery strategies, could shift the balance. Further research is needed to define the most efficient configurations for specific applications, recognizing that optimal process design is case-dependent.

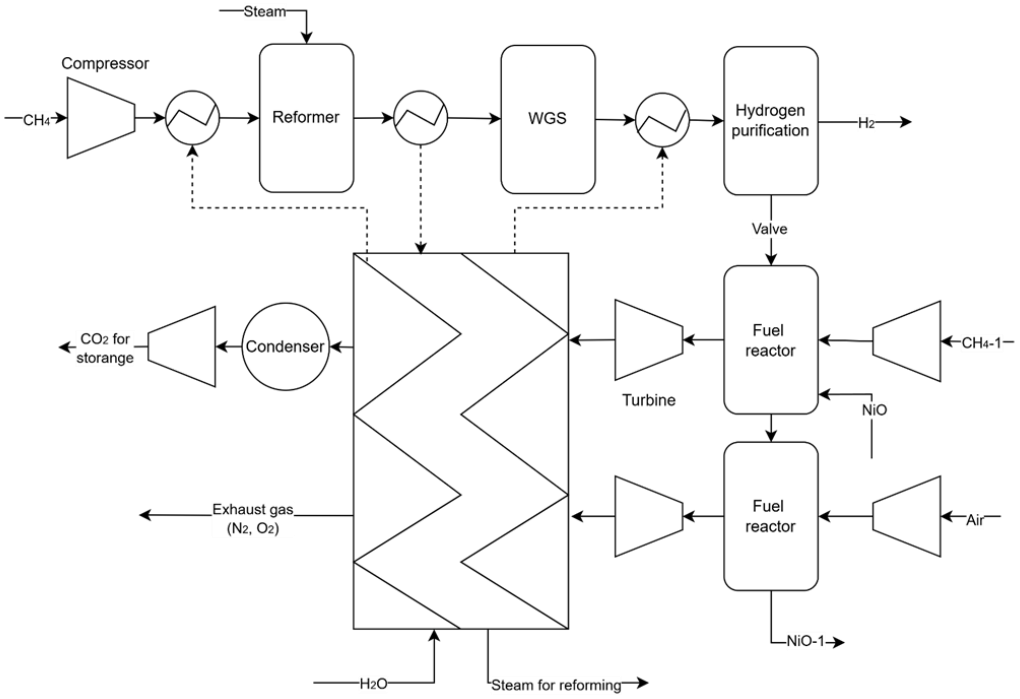

Figure 7. Chemical Looping Combustion-Steam Methane Reforming. Data was adopted from the literature [50].

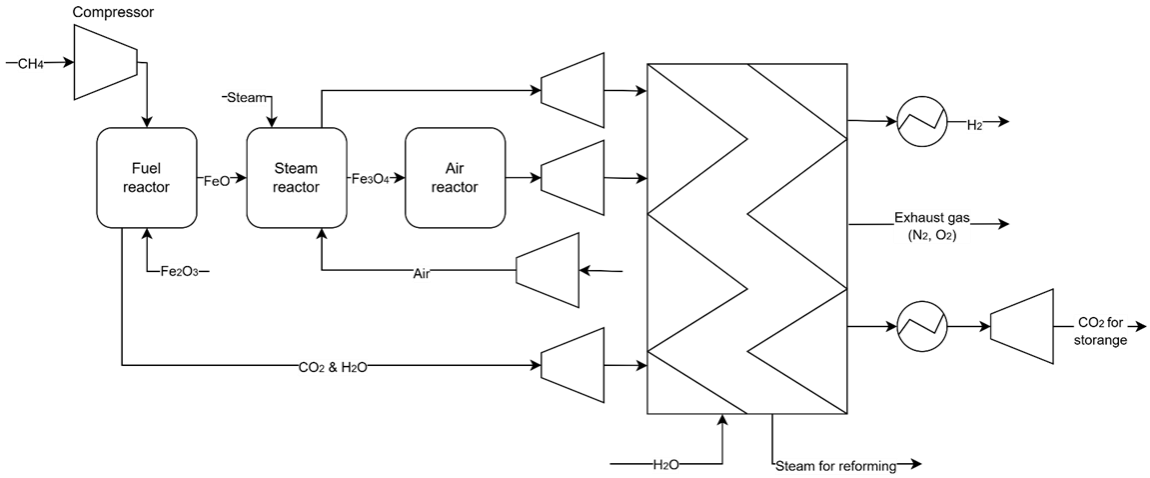

Figure 8. Chemical Looping Hydrogen Generation. Data was adopted from the literature [50].

As previously mentioned, there is a notable gap in the literature in the lack of comprehensive studies comparing CLC process modeling in Aspen, particularly regarding feedstock selection, oxygen carrier performance, and reactor modeling. Although some studies provide comparative insights, few directly address these key aspects. Additionally, the impact of auxiliary equipment, such as separators, cyclones, and heat exchangers on process

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performance is often overlooked, despite their role in influencing H_2 yield and overall energy efficiency. While their integration is common in industrial applications, their quantified effects in CLC simulations are rarely reported. Nevertheless, previous studies suggest that coupling CLC with a heat exchanger network can achieve energy and exergy efficiencies of 90.54% and 72.04%, respectively, the highest reported for such systems [49].

A similar approach is recommended for evaluating the role of separation technologies, such as cyclones and ideal separators. Their operational principles differ in a manner analogous to fluidized bed reactors and Gibbs reactors, where separators represent idealized systems while cyclones require detailed modeling. Therefore, assessing their advantages and limitations is essential for refining simulation accuracy.

Another critical aspect influencing simulation outcomes is the selection of thermodynamic models. While most studies specify the applied method, few justify their choice, often deferring to recommendations from the Aspen Physical Property System Guide [119]. Notably, commonly used models for CLC simulation share similar theoretical foundations (**Table 3**). However, for a comprehensive understanding of process behavior, a clearer justification of model selection is needed. For example, the Peng-Robinson equation is among the most frequently cited models in this context, widely used for vapor-liquid equilibrium predictions in the petroleum and natural gas industries due to its high accuracy [125]. Similarly, the RKS-BM method is often recommended for gas processing, refining, and petrochemical applications [126]. Despite their frequent use, the criteria for selecting one over the other are rarely discussed in the literature, leaving an open question regarding their optimal application in CLC modeling.

6. CONCLUSION

Throughout this review, the use of Aspen Plus has proven to be an effective tool for modeling different approaches, feedstocks, and oxygen carriers in the chemical looping combustion process for H_2 production. The conducted bibliometric analysis shows a growing interest in this technology, with a consistent rise in publications from 2013 to 2021 and a continued upward trend through 2023. Although data limitations prevent a precise projection for the coming years, available information up to mid-2024 suggests that research in this field is still expanding.

Even with progress in modeling these systems, the literature review reveals that many studies do not justify their choice of models, operating conditions, or equipment configurations. This lack of methodological justification makes it harder to reproduce results and compare them with experimental data, underscoring the need for stricter validation criteria in simulations.

Regarding oxygen carriers, iron and nickel-based materials remain the most studied due to their reactivity and economic viability. While nickel-based compounds achieve higher methane conversion, iron-based carriers perform better in water-splitting applications. Additionally, support materials such as $MgAl_2O_4$ and Al_2O_3 have been shown to improve stability and reactivity, helping to prevent sintering and deactivation problems.

The analysis of chemical looping configurations confirms that chemical looping reforming and steam methane reforming are among the most efficient strategies for H2 production. In particular, sorbent-enhanced configurations, especially those incorporating CaO, have significantly improved CO_2 capture and H_2 purity. Furthermore, integrating multi-stage separation processes has successfully addressed equilibrium limitations in conventional designs, increasing H_2 yields.

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Aspen Plus has proven to be a reliable tool for simulating these processes, with the Rgibbs model being one of the most widely used due to its ability to predict thermodynamic equilibrium without requiring kinetic data. However, recent studies have explored fluidized bed models that offer a more realistic representation of hydrodynamics and gas-solid interactions, reinforcing the importance of advancing kinetic-based simulations to improve model accuracy.

Moving forward, strengthening the link between simulation and experimental validation, optimizing process configurations for industrial scalability, and assessing the economic feasibility of emerging chemical looping technologies will be essential. Standardizing methodologies and expanding comparative studies will be key to establishing CLC-based H_2 production as a viable and efficient alternative in the current energy landscape.

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6. AUTHORS' NOTE

The authors declare that there is no conflict of interest regarding the publication of this article. The authors confirmed that the paper was free of plagiarism.

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