

Techno-Environmental Perspectives on Chemical Looping for Green Hydrogen: Oxygen Carrier Performance and Life Cycle Implications [†]

Alejandra Balaguera Quintero ^{*}, María Camila Zapata Chaverra and Gloria Isabel Carvajal 

Engineer Faculty, Universidad de Medellín, Medellín 050006, Colombia;
mzapata095@soyudemedellin.edu.co (M.C.Z.C.); gicarvajal@udemedellin.edu.co (G.I.C.)

^{*} Correspondence: abalaguera@udemedellin.edu.co

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Abstract

Green hydrogen has emerged as a key energy vector for decarbonizing industrial sectors that are difficult to electrify. Among the available production pathways, Chemical Looping technologies have attracted increasing attention due to their intrinsic CO₂ capture capability, high energy efficiency, and flexibility in feedstock utilization. This review provides a comprehensive analysis of recent advances in Chemical Looping processes for green hydrogen production, with particular emphasis on the role of oxygen carriers and the integration of environmental and life cycle assessment perspectives. The main Chemical Looping configurations are discussed, along with the performance and sustainability challenges associated with different oxygen carrier materials. In addition, the relevance of Life Cycle Assessment as a decision-support tool for identifying environmental hotspots and improvement pathways is examined. By integrating technological and environmental insights, this review identifies current research gaps and outlines future directions for the sustainable deployment of Chemical Looping systems within the hydrogen economy.

Keywords: green hydrogen; Chemical Looping; oxygen carriers; alumina; chamotte; Life Cycle Assessment

1. Introduction

In recent years, Chemical Looping processes (CLC, CLH) have emerged as a promising technology for the sustainable production of green hydrogen, combining high energy efficiency with low CO₂ emissions. Unlike conventional methods such as methane reforming with carbon capture or water electrolysis, Chemical Looping employs solid oxygen carriers (OCs) to oxidize fuels and produce hydrogen through redox cycles. This configuration avoids costly gas separation steps and reduces overall energy consumption, positioning Chemical Looping as a key alternative within the energy transition framework.

Green hydrogen has consolidated its role as a strategic energy vector for a decarbonized economy, given its ability to store and distribute renewable energy without CO₂ emissions during use. Produced via renewable-powered electrolysis or innovative thermochemical routes such as Chemical Looping, hydrogen offers a viable solution for sectors that are difficult to electrify, including heavy industry, maritime transport, and aviation. Its versatility as both an energy carrier and a chemical feedstock further reinforces its relevance



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in reducing fossil fuel dependence and achieving global climate targets established under the Paris Agreement [1–3].

Despite its potential, large-scale deployment of green hydrogen faces challenges related to infrastructure development, high initial costs, and process efficiency [4]. These barriers have driven growing interest in emerging technologies such as Chemical Looping, which enables inherent CO₂ capture and can be integrated with renewable energy systems and circular economy strategies for oxygen carriers.

This review presents a structured analysis of: (i) hydrogen as a clean and renewable energy vector, (ii) Chemical Looping processes for green hydrogen production, (iii) fixed oxygen carriers with emphasis on chamote and alumina-based materials, and (iv) the application of Life Cycle Assessment (LCA) to Chemical Looping systems and oxygen carriers. By integrating technological and environmental perspectives, this work aims to support informed decision-making for sustainable hydrogen production.

Rather than providing a general overview of hydrogen technologies, this review specifically focuses on the integration of Chemical Looping processes, oxygen carrier material performance, and Life Cycle Assessment perspectives. By narrowing the analytical scope to techno-environmental interactions within Chemical Looping systems, this work aims to provide a structured and critical synthesis of material performance and sustainability implications.

2. Materials and Methods

This study is based on a technological surveillance and state-of-the-art review of Chemical Looping processes for green hydrogen production. A structured literature analysis was conducted focusing on peer-reviewed articles addressing hydrogen as an energy vector, Chemical Looping technologies (CLC, CLR, CLH), solid oxygen carriers, and Life Cycle Assessment applications.

The methodological approach followed four main stages:

- (i) Identification of relevant keywords related to green hydrogen, Chemical Looping, oxygen carriers, alumina, chamote, and LCA.
- (ii) Selection of scientific articles addressing technological performance, material development, and environmental evaluation.
- (iii) Classification of the literature into thematic blocks (hydrogen energy, Chemical Looping processes, oxygen carriers, and LCA studies); and.
- (iv) Qualitative synthesis of results to identify technological trends, environmental hotspots, and research gaps.

The Life Cycle Assessment perspective was incorporated following ISO 14040 [1] principles, emphasizing the evaluation of environmental impacts across the life cycle of Chemical Looping systems and oxygen carriers, from raw material extraction to end-of-life. Attention was given to functional units, system boundaries, and impact categories reported in the reviewed studies, ensuring methodological consistency with the analyzed literature [5,6].

3. Results and Discussion

3.1. Hydrogen as a Renewable Energy Vector

Hydrogen has emerged as a strategic energy vector for decarbonizing hard-to-electrify sectors, including heavy industry, long-distance transport, and chemical production [2,4]. Among low-carbon production pathways, renewable-powered electrolysis and thermochemical routes such as Chemical Looping have gained attention due to their potential for reduced greenhouse gas emissions and system integration flexibility [4,7]. However, technological maturity, cost competitiveness, and environmental trade-offs remain critical challenges for large-scale deployment.

3.2. Chemical Looping Processes for Hydrogen Production

Chemical Looping for hydrogen production (CLH) is an emerging technology that enables hydrogen generation with reduced CO₂ emissions. The process is based on the use of solid oxygen carriers, typically metal oxides, which oxidize a fuel in one reactor, producing CO₂ and H₂O, while the reduced carrier is reoxidized with steam in a second reactor, releasing high-purity hydrogen. This inherent CO₂ separation eliminates the need for additional capture units, improving system efficiency [8,9].

Chemical Looping Reforming (CLR) integrates fuel reforming and carbon capture, using oxygen carriers to partially oxidize hydrocarbons and generate syngas rich in hydrogen. Studies highlight that reactor configuration, operating conditions, and oxygen carrier selection are critical factors influencing hydrogen yield, CO₂ capture efficiency, and system stability [10,11].

Although Chemical Looping offers significant advantages over conventional technologies, challenges remain related to oxygen carrier durability, redox stability under high temperatures, and economic feasibility at industrial scale [12,13].

3.2.1. Comparative Performance Trends and Technological Tensions

Although Chemical Looping configurations consistently demonstrate high hydrogen production potential, reported performance metrics vary significantly depending on reactor configuration, operating temperature, and oxygen carrier composition. Hydrogen yields reported in laboratory-scale studies typically range between 70% and 95% depending on reactor configuration and oxygen carrier composition [8,14] with Ni-based carriers often exhibiting the highest reactivity and conversion rates [12,14]. However, this superior performance is frequently associated with sintering, agglomeration, and long-term deactivation under cyclic redox conditions [12,13].

Iron-based oxygen carriers, while generally exhibiting lower intrinsic reactivity compared to nickel systems, demonstrate greater thermal stability [8,11] and reduced environmental toxicity. This trade-off between catalytic activity and material stability represents one of the most significant technological tensions in Chemical Looping hydrogen production.

Similarly, Chemical Looping Reforming (CLR) systems tend to achieve higher hydrogen selectivity under optimized steam-to-carbon ratios, yet such conditions may increase energy demand and operational complexity. These contrasting findings indicate that no single configuration simultaneously maximizes hydrogen yield, oxygen carrier durability, and environmental performance.

This variability underscores the need for integrated techno-environmental evaluation frameworks capable of assessing performance beyond isolated efficiency metrics.

3.2.2. Quantitative Comparison of Technical Performance Indicators

Reported hydrogen yields in Chemical Looping Hydrogen (CLH) and Chemical Looping Reforming (CLR) systems vary significantly depending on oxygen carrier composition, operating temperature, and reactor configuration. Laboratory-scale studies report hydrogen yields typically ranging between 70% and 95% for Ni-based oxygen carriers under optimized steam-to-carbon ratios [8,14]. In contrast, Fe-based systems often demonstrate slightly lower hydrogen yields but improved thermal stability and resistance to agglomeration [8,11].

CO₂ capture efficiency in Chemical Looping configurations frequently exceeds 90% due to inherent separation mechanisms, although this performance may decrease under transient conditions or material degradation scenarios [8,10]. Oxygen transfer capacity, a critical parameter governing cyclic redox performance, depends strongly on metal oxide selection, with NiO and Fe₂O₃-based carriers exhibiting higher reactivity relative to Mn- and Cu-based alternatives [14].

A structured comparison of key technical metrics reported in representative studies is provided in Table 1.

Table 1. Comparative overview of technical performance indicators in Chemical Looping hydrogen production studies.

Study	CL Configuration	Oxygen Carrier	H ₂ Yield (%)	CO ₂ Capture Efficiency (%)	Key Limitation
Rydén et al. (2006) [10]	CLR	NiO-based	~85–90	>90	Agglomeration risk
Khan and Shamim (2014) [11]	CLR	Fe-based	~70–85	>90	Lower intrinsic reactivity
Zheng et al. (2017) [12]	CLR	LaFeO ₃ -supported	~80–90	High	Stability concerns at high T
Dou et al. (2021) [13]	CL steam reforming	Mesoporous carriers	~75–88	>90	Durability under long cycles
Das et al. (2022) [14]	Review synthesis	Multiple OCs	70–95	85–95	Trade-off reactivity vs. stability

The comparative synthesis reveals three recurring patterns. First, Ni-based oxygen carriers consistently achieve higher hydrogen yields but are more susceptible to sintering and long-term degradation [12,14]. Second, Fe-based systems exhibit improved cyclic stability but may require higher operating temperatures to achieve comparable hydrogen conversion rates [8,11]. Third, although CO₂ capture efficiency is intrinsically high in most CL configurations, reported performance often excludes long-term degradation effects, limiting extrapolation to industrial-scale deployment [8,10].

These findings highlight the absence of standardized reporting metrics and underscore the importance of simultaneously evaluating hydrogen yield, oxygen carrier durability, and environmental performance.

3.3. Oxygen Carriers and Alumina-Based Materials

Oxygen carriers are the core component of Chemical Looping systems. Materials based on nickel, iron, copper, and manganese oxides have demonstrated high reactivity and oxygen transfer capacity. However, their long-term stability and environmental impacts depend strongly on the support material and synthesis route [8,14].

Alumina (Al₂O₃) is widely used as an inert support due to its high thermal stability, mechanical strength, and ability to disperse active phases, preventing sintering and improving cyclic performance. Chamote, composed primarily of alumina, has attracted attention as a fixed oxygen carrier support, offering robustness and potential cost advantages. Nevertheless, alumina production is energy-intensive and associated with significant environmental burdens, particularly during bauxite extraction and the Bayer process [15,16].

Durability–Reactivity–Sustainability Trade-Offs

The performance of oxygen carriers cannot be evaluated solely in terms of oxygen transfer capacity. While Ni-based systems often deliver superior hydrogen yields, their environmental footprint and potential toxicity raise sustainability concerns. Conversely, iron- and alumina-supported materials may present lower environmental risks but sometimes at the expense of catalytic efficiency.

Alumina-supported carriers, including chamote-based materials, improve mechanical resistance and cyclic stability. However, the environmental burden associated with alumina production—particularly energy consumption and bauxite residue generation—introduces a sustainability paradox: materials designed to enable low-carbon hydrogen production may themselves embody significant upstream emissions [15–17].

This durability–reactivity–sustainability trade-off remains insufficiently addressed in the literature and represents a critical research gap for scaling Chemical Looping technologies.

3.4. Life Cycle Assessment of Chemical Looping Systems and Oxygen Carriers

Life Cycle Assessment (LCA) has increasingly been applied to evaluate the environmental performance of Chemical Looping (CL) systems, particularly in comparison with conventional hydrogen production routes. However, reported results vary considerably depending on methodological assumptions, system boundaries, and impact assessment methods employed.

Several studies assess CL-based hydrogen production using cradle-to-gate boundaries, focusing primarily on reactor operation and fuel conversion stages [18,19]. Others extend the analysis to cradle-to-grave scenarios, incorporating oxygen carrier production, material transport, and end-of-life management [17,20]. These boundary differences significantly influence reported Global Warming Potential (GWP) values and other impact indicators.

Similarly, functional units differ across studies, ranging from “1 kg of H₂ produced” to “1 MJ of energy generated” or electricity output equivalents [18,19]. Such variability complicates direct comparison of environmental results and may partially explain discrepancies in reported emissions reductions.

Regarding impact assessment methods, most studies prioritize Global Warming Potential, typically using CML or ReCiPe-based approaches [17,18]. However, fewer analyses extend to toxicity-related categories, resource depletion, or water consumption. The limited scope of impact categories may obscure trade-offs between operational CO₂ reductions and upstream material burdens [17–21].

A comparative synthesis of selected LCA studies is presented in Table 2.

Table 2. Comparative overview of LCA studies applied to Chemical Looping systems.

Study	Functional Unit	System Boundary	LCIA Method	Main Environmental Hotspot
Wang et al. (2018) [17]	1 kg H ₂	Cradle-to-gate	CML	Fuel combustion and energy input
He et al. (2019) [20]	Electricity output	Cradle-to-gate	CML	Upstream fuel extraction
Thorne et al. (2019) [18]	Power plant output	Cradle-to-grave	ReCiPe	Material production
Sáez-Guinoa et al. (2023) [21]	1 kg alumina	Cradle-to-gate	ReCiPe	Alumina production energy demand

First, although CL technologies demonstrate significant operational CO₂ reductions relative to conventional reforming, upstream processes—particularly oxygen carrier production and energy-intensive material synthesis—often dominate total life cycle impacts [17,20].

Second, methodological heterogeneity in functional units and boundary definitions limits cross-study comparability, underscoring the need for standardized reporting frameworks consistent with ISO 14040 [1] principles [5,6].

Third, uncertainty and sensitivity analyses remain limited in most published assessments [18,19], restricting the robustness of environmental conclusions, particularly under varying fuel sources or oxygen carrier lifetimes.

For alumina-supported oxygen carriers, environmental impacts are strongly influenced by the energy intensity of the Bayer process and associated bauxite residue management [15–17]. These findings reinforce the importance of integrating material durability, regeneration potential, and circular economy strategies into environmental evaluations.

Overall, while Chemical Looping systems offer promising decarbonization potential, LCA results indicate that technological optimization must be coupled with material and supply-chain improvements to ensure net environmental benefits.

3.5. Cross-Cutting Challenges and Open Research Questions

Despite rapid technological progress, several unresolved challenges remain in the development of Chemical Looping systems for sustainable hydrogen production.

First, there is a persistent gap between laboratory-scale performance and pilot-scale validation [8,20]. Many studies report high hydrogen yields under controlled conditions, yet long-term cyclic stability, mechanical degradation, and material attrition are rarely evaluated beyond short experimental campaigns.

Second, the literature demonstrates limited integration between process optimization and environmental assessment [18,19]. Most studies treat reactor performance and life cycle impacts as separate analytical domains, preventing a holistic understanding of system sustainability.

Third, methodological inconsistencies in reported metrics—such as varying definitions of hydrogen yield, CO₂ capture efficiency, and system boundaries—limit cross-study comparability [5,6,17].

Finally, few studies explore circular economy strategies for oxygen carrier regeneration, recycling, or material substitution, despite the significant upstream impacts associated with metal oxide production.

Future research should prioritize:

- (i) Standardized performance indicators;
- (ii) Integration of techno-economic and life cycle modeling;
- (iii) Long-term durability assessment;
- (iv) Development of lower-impact oxygen carrier materials.

3.6. Toward an Integrated Techno-Environmental Evaluation Framework

Based on the comparative and methodological synthesis presented in this review, a conceptual techno-environmental integration perspective for Chemical Looping hydrogen systems is proposed.

Current literature frequently evaluates technical performance (e.g., hydrogen yield, oxygen transfer capacity, CO₂ capture efficiency) independently from environmental metrics such as Global Warming Potential or resource depletion [8,14,18]. This separation limits the ability to identify optimal trade-offs between process efficiency and life cycle sustainability.

The proposed integrative perspective emphasizes the simultaneous evaluation of:

- (i) Hydrogen production efficiency and CO₂ capture performance;
- (ii) Oxygen carrier durability and cyclic stability;
- (iii) Upstream environmental burdens associated with material synthesis;
- (iv) Circular economy potential, including carrier regeneration and recycling.

By aligning reactor performance metrics with life cycle indicators under consistent functional units and system boundaries [5,6], this framework supports more robust decision-making for scaling Chemical Looping technologies.

Future research should prioritize multi-criteria optimization approaches that integrate techno-economic assessment and dynamic LCA modeling to capture long-term system behavior [18,19].

4. Conclusions

This review provides a structured and comparative synthesis of Chemical Looping technologies for green hydrogen production, emphasizing the interdependence between technical performance and environmental sustainability.

The analysis highlights that high hydrogen yields are often associated with durability constraints and upstream material burdens, particularly in oxygen carrier production. Although Chemical Looping systems demonstrate strong potential for inherent CO₂ capture, life cycle considerations reveal that material synthesis, energy intensity, and supply-chain factors play a decisive role in determining overall environmental performance.

Beyond summarizing existing literature, this work advances an integrated techno-environmental evaluation perspective that links hydrogen yield, oxygen carrier stability, upstream impacts, and circularity potential under harmonized methodological conditions. By addressing the fragmentation observed between reactor performance studies and environmental assessments, the review outlines a structured research agenda for future development.

Future investigations should prioritize standardized performance metrics, long-term durability testing, broader environmental impact coverage, and dynamic life cycle modeling to ensure that Chemical Looping technologies achieve genuine sustainability gains at industrial scale.

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