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Green Hydrogen to Sustainable Fuels and Chemicals: A Review of Heterogeneous Catalysts for Olefins, Ammonia, and Light Alcohols

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ABSTRACT

The transition to a carbon-neutral energy economy requires the development of efficient, renewable energy carriers. Green hydrogen (GH₂), produced via water electrolysis using renewable energy sources such as wind and solar power, is a promising solution but faces significant challenges related to storage, transport, and cost. This review explores the catalytic synthesis of alternative energy vectors, specifically light olefins, ammonia (NH₃), and light alcohols such as methanol (CH₃OH) and ethanol (CH₃CH₂OH), using GH₂ as a hydrogen source. This study investigates recent progress in catalyst engineering and process intensification across various energy vectors, with particular emphasis on the critical role of catalyst support materials in enhancing catalytic performance. For light olefins, Fischer–Tropsch synthesis (FTS) is highlighted, with a focus on Fe- and Co-based catalysts supported on metal oxides and carbon materials and the influence of promoters and support morphology on selectivity and stability. Green NH₃ synthesis is discussed, emphasizing both traditional Haber–Bosch (HB) processes and emerging pathways utilizing transition metals, metal–organic frameworks (MOFs), and metal-free catalysts, with attention to catalytic activity under milder conditions. This review also covers the catalytic hydrogenation of carbon dioxide (CO₂) to light alcohols, such as CH₃OH and CH₃CH₂OH, detailing the roles of unsupported, metal oxide-supported, and MOF-supported catalysts in enhancing selectivity and yield. The synthesis of these energy vectors not only addresses the limitations of direct hydrogen use but also leverages existing infrastructure for storage and distribution, positioning them as key components in the future renewable energy landscape. This scientific revision of industrially relevant energy vectors for the GH₂ industry concludes with an assessment of current challenges and future directions for catalyst design and process integration to enable the large-scale adoption of GH₂-derived energy carriers.

1 | Introduction

1.1 | Hydrogen: The Color Spectrum

Greenhouse gas emissions, such as carbon monoxide (CO) and methane, have increased dramatically over the last decades, producing irreversible effects on the global climate. To reduce gas emissions, countries around the world have committed to achieving carbon neutrality by reducing the use of fossil fuels as their primary energy source. In this context, the development of

new technologies capable of producing renewable fuels that can contribute to the decarbonization of the energy matrix has become a primary goal [1].

Although the use of renewable energy sources such as wind and solar to produce electricity has expanded rapidly, the inherent intermittency of these technologies requires the use of storage systems, such as electrochemical (batteries) or thermal systems, to ensure energy supply. In this regard, nonconventional renewable energies face several challenges for mobile systems such

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as trucks and ships due to limited energy storage, excessive transport volume requirements, limited charging infrastructure, and battery lifetime. Consequently, the demand for alternative energy vectors has increased, with particular emphasis on energy storage in the form of chemical bonds, which offers promising potential to address these limitations.

In this scenario, the use of hydrogen as an alternative fuel represents a suitable solution because it can be produced by the water splitting reaction (see equation (1)) using renewable energy sources, such as electricity. Additionally, hydrogen has the highest energy per mass ratio of any fuel (120 MJ/kgH₂) [2]. However, hydrogen is the smallest molecule in nature with a low volumetric energy density (8 MJ/L for cryogenic liquid at −253°C and 4.5 MJ/L for compressed gas at 69 MPa) [2, 3], making the storage and transport of large amounts of hydrogen a challenging task.

Therefore, hydrogen itself is not an energy source but an energy carrier that must be produced from hydrogen-containing compounds such as water, natural gas, or biomass. In recent years, an intuitive color-based nomenclature has emerged to classify hydrogen according to the production method and associated carbon footprint. Although this color-coding system is not standardized by any international regulatory body, it has been widely adopted by industry, policymakers, and the scientific community to communicate the environmental impact of different hydrogen production routes [4–6].

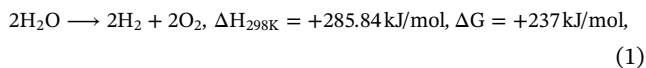
The most relevant colors are:

- **Green hydrogen (GH2):** Produced via water electrolysis using renewable electricity (solar, wind, hydro, tidal, and geothermal).
- **Blue hydrogen:** Produced from fossil fuels via steam methane reforming or autothermal reforming coupled with carbon capture, utilization, and storage.
- **Gray hydrogen:** Produced from natural gas via steam methane reforming without carbon capture; currently accounts for approximately 75% of global hydrogen production.
- **Brown/Black hydrogen:** Produced from lignite or bituminous coal via coal gasification; the most carbon-intensive pathway.
- **Turquoise hydrogen:** Produced via methane pyrolysis, splitting natural gas into hydrogen and solid carbon, avoiding CO₂ emissions if powered by renewable energy.
- **Pink (or purple) hydrogen:** Produced via water electrolysis powered by nuclear energy.
- **Yellow hydrogen:** Produced via water electrolysis using grid electricity of mixed origin.
- **White hydrogen:** Naturally occurring geological hydrogen found in underground deposits.
- **Orange hydrogen:** Produced via water electrolysis using geothermal energy.
- **Red hydrogen:** Produced via water electrolysis using biomass-derived energy.

1.2 | GH2 Prospects and Applications, GH2 as a Value Chain

As mentioned above, GH2 is defined as hydrogen produced via water electrolysis using electricity generated from renewable energy sources, including solar photovoltaics (PVs), wind power (onshore and offshore), hydropower, tidal energy, and geothermal power. The defining characteristic of GH2 is that its production results in zero direct greenhouse gas emissions, although life-cycle emissions associated with manufacturing electrolyzers and renewable energy infrastructure must be considered [4–6].

In general, GH2 is produced using renewable energy sources such as wind, solar, or tidal energy in areas with limited access, far from industrial centers where large amounts of fuel are needed. There are various technological options for storing and transporting hydrogen on the market, including fuel cells and high-pressure hydrogen vessels. The disadvantage of these storage and transport systems is the low energy per volume of hydrogen that can be carried and stored, which increases production costs and, consequently, the commercial price of GH2.



In summary, the main limitations and challenges of GH2 technology are (1) cost: Current GH2 production technology is more expensive (by a factor of three) than conventional technology for hydrogen production from fossil fuel sources; (2) production volume: Current global GH2 production is less than 0.04%; (3) infrastructure: Scaling up GH2 production will require significant investment in infrastructure such as electrolyzers, wind turbines, hydrogen storage devices, and distribution networks; (4) regulatory and policy challenges: Governments must define tax rates and generation restrictions on hydrogen production and sales; and (5) technical limitations: GH2 production is limited by water availability, storage, and the amount of renewable energy required for the process [7–9].

The use of energy vectors, including olefins (such as ethylene and propylene, $\Delta H_f = -148.95 \text{ kJ/mol}$), methanol (CH₃OH) ($\Delta H_{298\text{K}} = -49.5 \text{ kJ/mol}$), aromatics and ammonia (NH₃) ($\Delta H_{298\text{K}} = -92.22 \text{ kJ/mol}$), as chemical-bond hydrogen storage appears to be a suitable solution for storage and distribution issues.

Although the conversion of GH2 to these chemical compounds may be energetically unfavorable, the main advantage of these molecules compared to conventional hydrogen storage systems is the high energy density that chemical bonds can hold (see Table 1). This allows large amounts of energy to be stored and carried in a reduced volume. The most common synthesis route for these energy vectors is heterogeneous catalysis (see Figure 1), where the selection of the appropriate catalyst can favor the reaction by lowering energy requirements and increasing product yield.

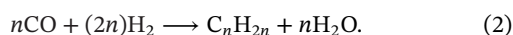
This work is focused on the review of the current trends for the most relevant catalytic systems used in the synthesis of energy vectors, such as light olefins, NH₃, and CH₃OH, and the recent advances in the development of more efficient catalysts for the synthesis of these vectors.

TABLE 1 | Energy density of selected energy vectors for GH2.

Energy vector	Energy density (MJ/kg)	Volumetric energy density (MJ/L)
Hydrogen	120	4.5 at 690 bar
Ethylene	50.4	32
Propylene	45.8	23.8
Methanol	22	17.3
Ethanol	29.7	23.3
Ammonia	22.5	12.7 at 8 bar

2 | Light Olefins From GH2

Olefins such as ethylene and propylene are building block molecules for the synthesis of fine chemicals, polymers, lubricants, and synthetic fuels [10, 11]. Syngas, a mixture of CO, carbon dioxide (CO₂) and hydrogen (H₂), is used as a feedstock for the production of olefins. The direct conversion of syngas into these molecules via the Fischer–Tropsch-to-olefins (FTO) process is a topic of interest for industrial and academic research [10].



Fischer–Tropsch synthesis (FTS) is a catalytic hydrogenation process that converts syngas into valuable chemical products, that is, paraffins, long-chain hydrocarbons, and olefins [12]. Several transition metals are catalytically active for FTS reactions, including nickel (Ni), ruthenium (Ru), iron (Fe), and

cobalt (Co) [12]. Ru-based catalysts have been used as active metal phases for the FTO synthesis. For instance, Yao et al. reported that Ru nanoparticles supported on different metal-oxide supports, such as ZrO₂, MnO₂, CeO₂, SiO₂, and active carbon, exhibited the following activity order: Ru/SiO₂ > Ru/ZrO₂ > Ru/MnO₂ > Ru/AC > Ru/CeO₂ [13]. Ru-based catalysts are not used at the industrial level due to Ru scarcity and high commercial price; thus, Fe and Co are the most relevant active metals for industrial FTS applications [12].

Fe-based FTS catalysts are the most relevant for the synthesis of olefins and have been used in industrial and research applications over the last years [14–33]. For Fe-based catalysts, the FTS process occurs in the temperature range between 220°C and 340°C, also known as high-temperature Fischer–Tropsch (HTFT). For Co-based catalysts, the temperature range of the reaction is between 220°C and 280°C, also known as low-temperature Fischer–Tropsch (LTFT). It is known that HTFT

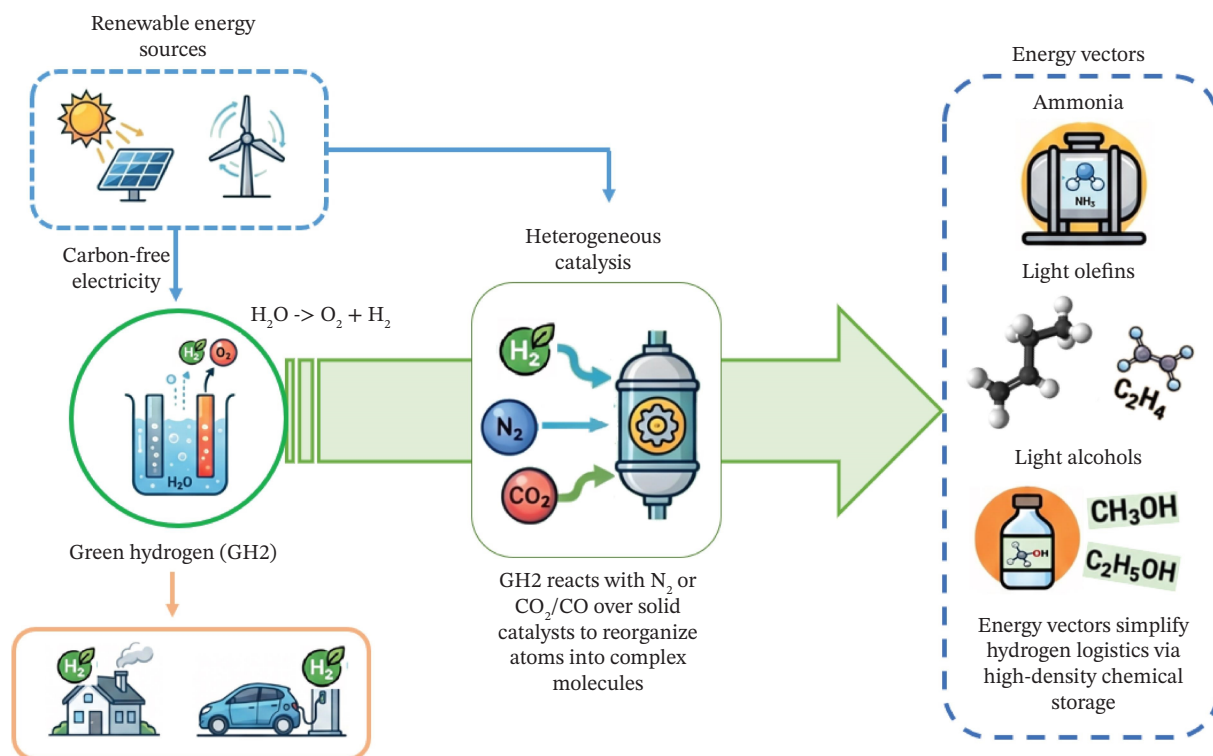


FIGURE 1 | The GH2 value chain begins by harnessing renewable energy sources—solar, wind, and hydroelectric—to generate clean electricity that powers a water electrolysis process, splitting it into O₂ and GH2 with zero direct emissions. Then, through heterogeneous catalysis, GH2 reacts with N₂ or CO₂/CO over solid catalysts to synthesize more complex molecules. Finally, these processes produce sustainable energy vectors such as ammonia, light alcohols, and light olefins, which solve the storage and transport challenges of hydrogen by creating high energy-density chemical bonds.

synthesis is more selective toward lighter chemical compounds such as olefins, while LTFT synthesis favors the production of long-chain hydrocarbons and waxes [12, 33].

Different support materials, including zeolites and metal–organic frameworks (MOFs) [24, 26, 28, 30, 33], have been used to stabilize catalyst nanoparticles physically, mechanically, and chemically, for instance, Fe nanoparticles [21]. From an industrial and research perspective, the most relevant materials are metal oxides and carbon supports.

2.1 | Metal Oxide Supports

Among the various metal oxide supports, silica (SiO_2) is one of the most relevant materials for research applications. SiO_2 exhibits limited metal-support interaction (MSI) with catalyst nanoparticles and is used to investigate nanoparticle performance during reactions and their interaction with other catalytically active nanoparticles (e.g., cocatalyst nanoparticles). Other oxide supports of industrial and research relevance include Al_2O_3 , TiO_2 , CeO_2 , and MgO [21].

Heikkinen et al. [15] studied the effect of copper (Cu) on the performance of Fe-based catalysts supported on SiO_2 and promoted by Cu and potassium (K) for the Fe–Cu–K/ SiO_2 FTS catalytic system. It was found that Cu promotion improved catalyst activity and stability. The presence of Cu decreased unwanted CH_4 selectivity and improved the selectivity to C_2 – C_4 olefins and C_{5+} hydrocarbons compared to an unpromoted catalyst. Additionally, the hydrocarbon selectivity was not strongly affected by different Cu loadings (between 1 and 5 wt.%); only a slight improvement was observed when Cu loading increased from 3 to 4 wt.% [15].

Han et al. [17] prepared different Fe–Si catalyst systems, including precipitated, impregnated, and core–shell Fe–Si catalysts, prepared by coprecipitation, impregnation, and hydrothermal–Ströber methods, respectively. It was found that a small amount of SiO_2 improves C_2 – C_4 selectivity and FTS stability and that the formation of a SiO_2 shell hinders the transfer of C_{5+} products. An increase in SiO_2 shell thickness considerably reduced CO conversion and thus catalyst performance. Additionally, the core–shell catalyst exhibited higher selectivity and stability toward C_2 – C_4 products after 120 h of reaction compared to precipitated and supported catalysts [17].

Liu et al. [22] synthesized Fe-based catalysts promoted by K over $\text{ZnAl}_2\text{O}_4 \cdot x\text{Al}_2\text{O}_3$ ($x = 1, 5$) supports. Catalytic activity showed an inverse correlation with x for the different synthesized catalysts. The C_2 – C_4 olefin/paraffin ratio was influenced by the formation of the Fe_5C_2 carbide phase, which increased the olefin/paraffin ratio. For the Fe– $\text{K}_2\text{O}/\text{ZnAl}_2\text{O}_4 \cdot 1\text{Al}_2\text{O}_3$ catalyst at approximately 20 bar and 340°C , the C_2 – C_4 selectivity reached a maximum of 52.1%, with a CO conversion of 38.3%, a C_2 – C_4 olefin/paraffin ratio of 13.0, and a low methane selectivity of 7.7% [22].

Garona et al. [25] evaluated and predicted the performance of a Fe-based FTS catalyst promoted by K using machine learning models. Three-layer feedforward neural networks were adjusted to evaluate catalyst performance using operating conditions and catalyst composition as inputs and CO conversion, light olefins selectivity, and CO_2 yield as outputs. The study revealed that Fe–K/ TiO_2 catalysts exhibited the highest light olefins selectivity at 500°C and 20 wt.% active phases [25].

2.2 | Carbon Supports

Carbon species, for example, carbon nanotubes and graphene-like materials, have been used as support materials for catalyst nanoparticles due to their intrinsic characteristics such as high porosity and high surface area [12, 14, 21]. For these materials, the MSI between the metal nanoparticles and the carbon support is negligible [12], increasing their importance for studying catalyst nanoparticle behavior and the effect of cocatalysts and promoters such as K or sodium (Na). In particular, carbon nanotubes have been used as support for Fe-based FTS catalysts [12].

Research advances on carbon supports for FTS reactions are not limited to Fe-based catalysts. Yao et al. [13] tested different supports for Ru-based FTS catalysts, comparing CeO_2 , SiO_2 , MnO_2 , ZrO_2 , and activated carbon. Activated carbon showed low catalytic activity and strong secondary hydrogenation, leading to low olefin selectivity. The findings suggested that weak MSI has an important influence on catalyst performance. For instance, small, supported Ru nanoparticles were found on activated carbon, which correlated with low CO conversion [13].

Chernyak et al. [16] used spark plasma sintering to produce bulk carbon-based catalysts. Fe-based catalysts promoted by K supported on N-doped graphene nanoflakes (N-GNFs) and N-doped carbon nanotubes (N-CNTs) were synthesized. The Fe–K/N-GNFs catalyst presented higher activity toward FTS products, including olefins (up to ~40%). Effects such as sintering were predominantly observed on N-CNT-supported catalysts, which showed lower performance compared to N-GNF-supported catalysts. The N-doping of the GNFs increased the MSI between the support and the catalyst nanoparticles and surface defectiveness, improving the anchoring and stabilization of the Fe nanoparticles [16].

Schultheis et al. [19] synthesized Fe-based catalysts promoted by Na and sulfur (S) on beaded carbon black supports. The carbon supports had specific surface areas ranging from $36 \text{ m}^2/\text{g}$ to $380 \text{ m}^2/\text{g}$, making them suitable candidates for CO_2 -FTS catalysts, especially the catalyst with a surface area of $150 \text{ m}^2/\text{g}$. The long-term stability of the Fe/C catalysts was demonstrated over 170 h, with no decrease in CO_2 conversion during the reaction. Schultheis et al. demonstrated that inexpensive beaded carbon blacks can be used as a support material for FTS catalysts instead of other carbon supports such as carbon nanotubes or graphene-like materials [19].

Molybdenum (Mo) and K were tested as promoters for Fe/CNTs catalysts by Yahyazadeh et al. [20]. The study concluded that K and Mo produced a synergistic effect: K increased CO conversion, while Mo enhanced light olefin selectivity. Also, Mo addition increased Fe dispersion, reduction temperature, and CO_2 -desorption temperature by inducing structural defects on the CNTs used as support. The bimetallic promotion (K and Mo) also prevented sintering and agglomeration of nanoparticles in the calcined and spent Fe–K–Mo/CNTs catalyst [20].

Hassan et al. [23] prepared an Fe-based catalyst promoted by Na and Mn and supported on graphene oxide (GO). The addition of promoters decreased the reduction temperature of the catalyst. The highest CO conversion and the largest amount of liquid product were observed for the Fe–Mn–Na/GO catalyst. Formation of Fe-carbide was observed in the spent catalysts, indicating that the GO support reacted with Fe nanoparticles during the active state of the catalyst [23].

Co-based catalysts have been shown to be active for the synthesis of olefins under low-temperature FTS conditions [10, 34–41]. The cobalt carbide phase (Co_2C) has been identified as the active phase for olefin synthesis. Gong et al. [10] studied the effect of the preparation method for Co-based catalysts promoted by Mn. It was observed that CoMn spinel oxide was present in the calcined catalyst synthesized by co-precipitation of Co and Mn salts and that Co_3O_4 and MnO_2 were the major phases. During the reaction, the CoMn spinel phase gradually evolved into Co_2C nanoprisms, showing low methane selectivity, high olefin selectivity, and high CO conversion. The study concluded that the preparation method significantly influenced the morphology of the Co_2C phase and catalyst performance [10].

Jiang et al. [34] prepared different Co-based catalysts promoted by Na, Mn, and Na–Mn and supported on carbon and oxide supports. The results demonstrated that the promoter effect depends on the MSI between Co nanoparticles and the support. For weak MSI (cobalt–carbon system), the addition of Na to Co–Mn with Co_2C sites decreased chain growth, increasing selectivity to lower olefins. For strong MSI (Co– SiO_2 system), the addition of Na or Mn induced the formation of CoO and Co metal and a decreased methane selectivity, while the addition of Na alone led to a marked decline in catalytic activity due to the suppressed reduction of the cobalt oxide (Co_3O_4) species and promoted nondissociative CO adsorption [34].

Lin et al. [35] synthesized Co–Mn–based catalyst coated with SiO_2 to limit the CO_2 selectivity. It was observed that SiO_2 -coated Co–Mn–Al composite oxides could suppress CO_2 selectivity and enhance olefins selectivity during FTS. The SiO_2 coating did not change the structure of the active phase (Co_2C nano-prisms with exposed (101) and (020) facets) and could considerably reduce the capillary effect of water and water adsorption capacity, thus inhibiting the water–gas-shift-reaction (WGS). The SiO_2 coating of Co_2C produced an H_2 -enrichment effect, improving the olefins/paraffins ratio and restraining CH_4 formation [35].

The effect of the pretreatment of the SiO_2 support for the Co_2C -based catalyst was studied by Li et al. [36]. Experimental results confirmed that support pretreatment plays a key role in catalyst activity. The support treated at low and medium temperatures (350°C and 650°C) showed high interaction with Co, inducing the formation of Co_2C nanospheres during reaction, which exhibited low activity and high methane selectivity. When the support was treated at high temperature (990°C), the interaction between Co and SiO_2 weakened due to the decrease in Si–OH groups on the support surface, enhancing the formation of Co_2C nano-prisms with exposed (020) and (101) facets, resulting in high selectivity and activity toward olefins, low methane selectivity, and narrow product distribution [36].

Gong et al. [37] synthesized Co@C core-shell nanocatalysts (Co metal nanoparticles encapsulated by carbon layers) via a reduction–carburization–pyrolysis pretreatment of Co/ SiO_2 . This catalyst exhibited high olefins selectivity and negligible WGS activity. Moreover, the Mn-doped (2Mn–Co@C) catalyst also showed high olefin selectivity with limited methane selectivity [37].

Liu et al. [38] reported a catalyst with high selectivity synthesized by physical mixing of Co_2C with hydrophobic SiO_2 . The catalyst showed high performance because of the suppression of WGS,

which improved catalyst performance. The mixture of Co_2C and hydrophobic SiO_2 created hydrophobic environments that could accelerate the transfer from the active sites and simultaneously prevent water reabsorption of the Co_2C active sites. The hydrophobic environment also favored the formation of an H_2 -poor and CO-rich local chemical environment, enhancing olefin production [38].

A synthesis route for the fabrication of high-purity single-crystal nanoprisms of Co_2C -p was proposed by Hong et al. [40]. The roadmap begins with the reduction of the Co_3O_4 precursor to the CoO intermediate, followed by the carburization into Co_2C nanocrystals and Co_2C -p. The formation of Co_2C -p was accompanied by a switch in selectivity from long-chain hydrocarbons (C_{5+} ~59%) to lower olefins (50.2%) [40]. Another synthesis route for a Co-based catalyst using Mn as a primary promoter and Na as a secondary promoter was proposed by Liu et al. [41]. The catalyst prepared by the “melting” method exhibited high selectivity to lower olefins and low CO_2 selectivity. $\text{MnCo}_2\text{O}_{4.5}$ was identified as the precursor; after reduction and exposure to syngas, it evolves into Co_2C via carburization of the Co^0 present after reduction [41].

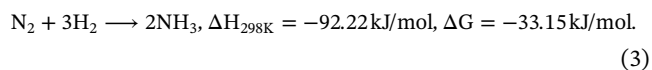
Additionally, studies based on Aspen Plus simulations have focused on separation processes for coal-based Fischer–Tropsch-to-olefins (CFTO) technology using cobalt carbide nanoprisms as the catalytic active phase for FTS reactions [39].

Table 2 summarizes the different catalysts considered in this study. The data are organized by support type and their reaction characteristics, such as conversion, selectivity, and reaction conditions.

3 | NH_3 From GH2

NH_3 is a promising alternative for hydrogen storage [42]. NH_3 has a high volumetric hydrogen density (approximately 1.7 times higher than liquid hydrogen), meaning it can store more energy per volume unit. Green NH_3 , synthesized from GH2 and green nitrogen sources, does not suffer from storage and transportation because it can be stored at lower pressure than GH2, reducing transportation difficulties [43, 44]. Furthermore, NH_3 is widely used in the fertilizer sector, meaning that storage and transport infrastructure already exists. Considering the benefits described above, green NH_3 has a broader range of potential applications; for example, it can be used as a clean fuel for ships, for power generation, and as a feedstock for other chemicals [45–47].

Currently, industrial NH_3 production is dominated by the Haber–Bosch process, in which pure H_2 and N_2 react at high pressure and temperature over an Fe-based catalyst, as described in the following chemical reaction [43, 44]:



The use of GH2 instead of steam-reformed hydrogen from fossil fuel sources is certainly a step forward toward decarbonization. However, for practical applications of NH_3 as an energy vector, the energy requirements of the HB process must be addressed. To this end, efforts are being made to develop more efficient catalysts and modified processes.

TABLE 2 | Summarized results for the syngas hydrogenation reaction to olefins using different catalysts sorted by support type.

Support type	Support	Catalyst	Conversion (%)	Selectivity (%)	GHSV (mLg ⁻¹ h ⁻¹)	Temperature (°C)	Pressure (MPa)	Ref
Unsupported		Co ₂ C nanoprisms	25.6	50.2	900	280	0.1	[40]
		Co ₂ C	7.7	60.3	6000	240	0.1	[41]
Metal oxide	Al ₂ O ₃	30Co10Mn0.4Na/Al ₂ O ₃	59.5	6.0	8000	260	0.5	[34]
	CeO ₂	Ru/CeO ₂	8.3	49.7	3000	300	1.0	[13]
	MnO ₂	Ru/MnO ₂	12.0	70.1	3000	270	1.0	[13]
		Fe–Cu-4%–K/SiO ₂	18.0	29.9	50,000	300	2.1	[15]
		30Co10Mn0.4Na/SiO ₂	79.9	5.2	8000	260	0.5	[34]
	SiO ₂	Ru/SiO ₂	33.6	79.4	3000	270	1.0	[13]
		CoMnNa/SiO ₂ -990	23.9	79.9	2000	270	0.5	[36]
		CMASc-12	13.7	59.3	4000	260	0.1	[38]
	ZnAl ₂ O ₄ ·1Al ₂ O ₃	15%Fe/2%K ₂ O/83% (ZnAl ₂ O ₄ ·1Al ₂ O ₃)	38.3	52.1	1500	340	2.0	[22]
	ZrO ₂	Ru/ZrO ₂	17.6	33.5	3000	270	1.0	[13]
	Activated C	Ru/AC	7.8	29.9	3000	270	1.0	[34]
	CNT	30Co10Mn0.4Na/AC-1	18.5	25.4	8000	260	0.5	[34]
	Carbon spheres	0.5K5Mo10Fe/CNTs	71.5	49.6	2000	280	3.5	[20]
Carbon	Graphene nanoflake	30Co10Mn0.4Na/CS	9.2	27.1	8000	260	0.5	[34]
		FeKNGNF700	16.1	62.0	5000	300	2.0	[16]

3.1 | MOF Catalysts

MOFs are porous materials that have proven to be highly effective in the synthesis, capturing, and release of NH_3 [48]. The high specific surface area, tunable porosity, and catalytic properties make them ideal candidates as catalyst supports. Continued research and development on MOF materials is crucial to improve their efficiency and economic viability. NH_3 synthesis using MOFs combines green H_2 with N_2 in the presence of MOFs-based catalysts. For this synthesis method, a modified HB process operating at lower temperatures and pressure is applied. Specifically, MOFs such as MIL-101, MIL-53, MOF-74, and UiO-66 are important in the sustainable production of NH_3 from GH2.

MOFs MIL-101 has been investigated for its potential to catalyze the electrochemical reduction of N_2 to NH_3 using GH2. Various types of MIL-101 catalysts have been developed to enhance NH_3 production efficiency. MIL-101 (Fe), in which Fe is incorporated into the framework, has shown promising results for N_2 reduction reactions. MIL-101 (Cr), which uses chromium (Cr) as the metal center, exhibits good structural stability but has been found to be less effective for NH_3 production than its Fe counterpart. Co-doping involves incorporating Co into the MIL-101 framework, creating a bimetallic structure (e.g., Co-doped MIL-101 [Fe]). This modification aims to enhance catalytic activity through synergistic effects between the two metals. MIL-101 (Al), which incorporates aluminum (Al) as the central metal, has been explored for various catalytic applications due to its high surface area, tunable porosity, and excellent thermal and chemical stability; however, its direct application in NH_3 production from GH2 has been less studied compared to other MIL-101 variants [49].

Additionally, the incorporation of aluminum into MIL-53 has been explored. MIL-53 (Al) consists of Al metal centers coordinated by inorganic [M-OH] chains connected to neighboring inorganic chains by terephthalate-based linker molecules. The structure features allow significant flexibility and porosity, which can be tuned by varying the synthesis conditions of MIL-53 (Al) [49, 50].

UiO-66 is a MOF composed of zirconium (Zr) nodes coordinated with benzene-1,4-dicarboxylate (BDC) linkers. It has garnered significant interest for its potential to catalyze the electrochemical reduction of N_2 to NH_3 using GH2. UiO-66 consists of Zr clusters connected by BDC linkers, creating a highly porous structure with a high surface area. This framework allows for effective gas adsorption and diffusion, which is crucial for catalytic reactions. UiO-66 exhibits excellent thermal and chemical stability, making it suitable for catalytic applications under various conditions, including high temperatures and humid environments. The large internal pores facilitate the N_2 and H_2 adsorption, enhancing the efficiency of the nitrogen reduction reaction (NRR). Loading noble metals such as gold (Au), platinum (Pt), or Ru into the UiO-66 framework can significantly enhance its catalytic properties due to the presence of metal ions or nanoparticles, which facilitates the adsorption and activation of N_2 molecules by providing active sites that promote $\text{N}\equiv\text{N}$ bond cleavage [51, 52].

3.2 | Transition Metal Catalysts

Transition metal catalysts such as Ru, Co, and Ni-loaded lanthanum nitride play a crucial role in the NH_3 synthesis from GH2. These catalysts facilitate the reaction under milder conditions compared to traditional methods, such as the HB process, making them essential for sustainable NH_3 production. The yield of NH_3 using transition metal catalysts varies significantly depending on the catalyst used and the reaction conditions.

Ru-based catalysts facilitate NH_3 synthesis at temperatures below 400°C and pressures around 1 MPa [53, 54]. Ru catalysts supported on rare-earth metal oxides have shown enhanced activity due to strong MSI [55, 56]. Additionally, Ru catalysts have been shown to be more active than traditional Fe catalysts, particularly under mild conditions, demonstrating their potential for efficient NH_3 production in less energy-intensive processes [57]. The NH_3 synthesis mechanism on Ru catalysts involves molecular chemisorption of N_2 followed by dissociative chemisorption of H_2 . This process is facilitated by the unique electronic properties of Ru, which allow effective N_2 activation even at lower temperatures [58].

Co-based catalysts have emerged as a promising alternative for green NH_3 production, particularly due to their cost-effectiveness and potential for high catalytic activity. Barium (Ba)-promoted Co catalyst supported on magnesium-lanthanum (Mg-La) mixed oxide enhances catalyst activity compared to the unpromoted catalyst [59, 60]. The effectiveness of cobalt can be enhanced by the support material, which promotes N_2 cleavage and its subsequent reduction to NH_3 . Jiang et al. [60] showed that the Ba-containing oxyhydride support not only improves electron donation to Co but also stabilizes the catalytic structure, allowing efficient NH_3 synthesis under milder conditions. Neodymium oxide (Nd_2O_3) has been explored as a support material for catalysts in green NH_3 synthesis. Nd_2O_3 is recognized for its ability to stabilize metal nanoparticles, enhancing their dispersion and preventing sintering during catalytic reactions. This stability is crucial for maintaining high catalytic activity over time, and the interaction between Co and Nd_2O_3 promotes effective charge transfer, which is essential for the activation of N_2 molecules. Strong MSI enhances the overall catalytic performance by improving the electronic properties of the metal [61].

The Ni-loaded lanthanum nitride (Ni-LaN) catalyst has been studied for its effectiveness in green NH_3 production, particularly in reducing energy requirements and enhancing catalytic performance. Ye and coworkers [62, 63] highlighted Ni-LaN catalysts can produce NH_3 at rates comparable to conventional metal nitride catalysts and show stability even in the presence of moisture, which is crucial for practical applications. They indicated that the Ni-LaN catalyst effectively facilitates N_2 adsorption and activation, contributing to NH_3 synthesis under milder conditions [62, 63]. The mechanism involves lattice N_2 and N_2 vacancies in the La-Al-N support playing significant roles in N_2 adsorption, while the active metal Ni is responsible for H_2 absorption and activation. This dual functionality enhances the overall reaction efficiency, allowing for effective NH_3 production.

3.3 | Metal Oxide Catalyst

Metal oxide catalysts are increasingly recognized for their potential in the sustainable production of NH_3 from green nitrogen and GH_2 . These catalysts can facilitate NH_3 synthesis under milder conditions compared to traditional methods. Metal oxides, such as Co_3O_4 , nickel oxide (NiO), and lanthanum oxide (La_2O_3), are commonly used as catalysts or supports in NH_3 synthesis. These oxides can enhance the catalytic activity of transition metals when used in composite forms. The basicity of metal oxide support plays a crucial role in catalytic performance, as basic sites can facilitate the adsorption and activation of N_2 molecules, increasing the efficiency of the NH_3 synthesis reaction.

La_2O_3 and its composites have been investigated as catalysts for NH_3 production due to their ability to enhance catalytic activity and stability. The $\text{Co-Ba/La}_2\text{O}_3$ catalyst exhibited high selectivity toward NH_3 synthesis due to the formation of a $\text{BaO-La}_2\text{O}_3$ nano-fraction around Co nanoparticles during high-temperature reduction. This structure improved nitrogen activation [64]. Lanthanide oxides, such as La_2O_3 , CeO_2 , and Nd_2O_3 , exhibit high basicity, which facilitates the adsorption and activation of N_2 molecules. This property is crucial for improving the efficiency of NH_3 synthesis. These oxides maintain structural integrity under reaction conditions, providing a stable support for active metal sites. This stability helps prevent sintering and loss of catalytic activity over time. In this context, CeO_2 has emerged as a promising catalyst for green NH_3 production, particularly when used in combination with other metals such as Ru and/or cesium (Cs). CeO_2 enhances catalytic performance by facilitating N_2 activation and hydrogenation [65].

Titanium dioxide (TiO_2) has unique properties, such as high stability, a tunable electronic structure, and the ability to form oxygen vacancies; these properties make it suitable for both electrochemical and photocatalytic applications in NH_3 production. In general, the process begins with the adsorption of N_2 molecules onto the surface of TiO_2 catalysts. The presence of oxygen vacancies enhances nitrogen adsorption by providing active sites that facilitate the breaking of the $\text{N}\equiv\text{N}$ triple bond. Once adsorbed, nitrogen undergoes dissociation to form reactive nitrogen species (N^*). This step is critical as it determines the efficiency of subsequent reactions. The activated nitrogen species react with H^* to form NH_3 . This reaction may involve intermediate formation such as NH or NH_2 before reaching NH_3 . The efficiency of this step is influenced by the presence of co-catalysts like noble metals or transition metals (e.g., Pd, Pt, Ni) that can facilitate electron transfer and improve reaction kinetics [66–68].

3.4 | Metal-Free Catalyst

Metal-free catalysts are gaining attention as viable alternatives for the electrochemical synthesis of NH_3 from GH_2 . These catalysts offer several advantages, including lower costs, reduced environmental impact, and enhanced stability compared to traditional metal-based catalysts. Recent research has focused on developing efficient metal-free catalysts that can operate effectively under ambient conditions [69].

Two-dimensional carbon nitride ($\text{g-C}_3\text{N}_4$) has emerged as a promising material for NH_3 production, particularly when grown on conductive substrates like carbon paper ($\text{g-C}_3\text{N}_4/\text{C}$). This configuration enhances the electrochemical reduction of N_2 using

renewable H_2 sources. N_2 molecules adsorb onto the surface of carbon nitride; the presence of nitrogen species within the carbon nitride structure enhances adsorption capacity and facilitates the activation of the $\text{N}\equiv\text{N}$ bond. Once adsorbed, N_2 undergoes dissociation to form reactive nitrogen species (N^*), which are essential for subsequent reactions. The electronic properties of carbon nitride allow effective stabilization of these intermediates. The carbon layer substrate provides a conductive pathway that enhances H_2 adsorption, allowing effective dissociation into atomic hydrogen (H^*). The activated nitrogen species react with H^* to form NH_3 . This process involves intermediate formation such as NH or NH_2 before producing NH_3 . Efficient electron transfer facilitated by the conductive carbon paper enhances this step. Finally, NH_3 desorbs from the catalyst surface, allowing regeneration of active sites for subsequent reactions. The basicity provided by the nitrogen species helps reduce product poisoning by facilitating NH_3 desorption [70].

Boron (B) and N_2 co-doped carbon materials have emerged as effective metal-free catalysts for the electrochemical synthesis of NH_3 [71]. These co-doped materials leverage the unique electronic properties imparted by both B and N_2 to enhance catalytic activity, selectivity, and stability in NRRs. The incorporation of both B and N_2 into the carbon structure creates a variety of active sites that facilitate the adsorption and activation of N_2 molecules. B enhances the electron density and charge transfer, while N_2 improves conductivity, leading to more efficient NRR. Co-doped carbon materials typically exhibit high surface areas and porosity, providing ample active sites for catalytic reactions, which is crucial for improving NH_3 production rates. These catalysts generally demonstrate good stability under electrochemical conditions, making them suitable for prolonged use in NH_3 synthesis processes [71].

Sulfur (S)-doped catalysts are promising materials for the electrocatalytic production of NH_3 from N_2 using GH_2 . The incorporation of sulfur into various carbon-based materials enhances their performance in N_2 reduction reactions, making them effective candidates for sustainable NH_3 synthesis. S doping introduces new active sites in carbon-based materials, such as graphene (S-doped graphene or S-G) and porous carbon. These sites enhance the adsorption of N_2 molecules, facilitating the cleavage of the $\text{N}\equiv\text{N}$ bond, which is crucial for subsequent reactions leading to NH_3 production. The presence of sulfur also improves hydrogen adsorption, allowing for effective dissociation into atomic hydrogen (H^*). This step is essential for the hydrogenation of activated nitrogen species, where the activated nitrogen species react with H^* to form NH_3 . The enhanced electron transfer facilitated by S-doped materials lowers the energy barriers for these reactions, improving overall reaction kinetics. Finally, NH_3 desorbs from the catalyst surface, allowing for regeneration of active sites. The basicity provided by sulfur functionalities aids in facilitating NH_3 desorption [72].

Table 3 summarized the different catalysts considered for the NH_3 synthesis. The data are organized by supports and their reaction characteristics, such as yield and reaction conditions.

4 | Light Alcohols From GH_2

Catalytic hydrogenation of CO_2 to light alcohols (CO_2 reduction reaction, CRR) is a promising method to produce renewable fuels

TABLE 3 | Summarized results for the N₂ hydrogenation reaction using different catalysts sorted by support type.

Support type	Support	Catalyst	Yield (g g _{cat} ⁻¹ h ⁻¹)	GHSV (mLg _{cat} ⁻¹ h ⁻¹)	Temperature (°C)	Pressure (MPa)	Ref
Unsupported		MIL-100 (Fe)	2.23·10 ⁻²	51,430	N/A	0.1	[73]
		MIL-100 (Al)	1.06·10 ⁻²	60,000	N/A	0.1	[74]
		MIL-53 (Al)	2.1·10 ⁻³	60,000	N/A	0.1	[71]
		B-BCN	4.19·10 ⁻²	2,250,000	N/A	0.1	[75]
		S-G	2.73·10 ⁻²	N/A	N/A	N/A	[51, 52]
MOF	UiO-66	Au/UiO-66	9.06·10 ⁻⁴	N/A	N/A	0.1	[51]
		Ru ₁ /d-UiO-66	1.13·10 ⁻³	N/A	N/A	0.1	[61]
		Nd ₂ O ₃	1.82	87,500	470	6.3	[60]
		BaAl ₂ O _{4-x} H _y	8.5	36,000	340	0.9	[56]
		Ru	9.35·10 ⁻²	36,000	400	1	[55]
Metal oxide		Ru	1.768·10 ⁻¹	72,000	400	1	[65]
		Cs-Ru/CeO ₂	2.23·10 ⁻²	6000	260	0.1	[64]
		Co/Ba/La ₂ O ₃	8.5·10 ⁻⁴	72,000	700	1	[66]
		TiO ₂	1.1·10 ⁻⁵	12,000	673	0.1	[59]
		Mg-La oxide	4.68	87,500	400	9	[70]
Metal free	C paper	g-C ₃ N ₄	2.9·10 ⁻³	N/A	N/A	0.1	[54]
Other	CaFH	Ru	8.5·10 ⁻⁴	36,000	50	0.1	[62, 63]
	LaN	Ni-LaN	9.42·10 ⁻²	36,000	400	0.1	

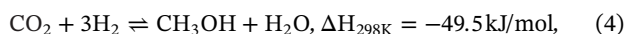
as well as to reduce atmospheric CO₂ through emission abatement or CO₂ capture and conversion. Thus, this process provides a two-fold solution for energy and environmental issues. Moreover, this process can be renewable and sustainable if the H₂ used comes from renewable sources such as GH₂, making it even more environmentally friendly compared to other traditional fuel production methods [76–86].

In this sense, the use of green H₂ and CO₂ to produce CH₃OH and ethanol (CH₃CH₂OH) appears to be a suitable solution for the synthesis of energy carriers and for environmental issues, despite its currently higher production cost. To make this technology more economically competitive and affordable [87], several challenges must be addressed, such as the need for highly efficient and selective catalysts and the thermodynamic limitations and issues of the reactions involved in the synthesis process [76, 78, 79, 81, 86–88].

CH₃OH and CH₃CH₂OH are considered potential alternative fuels, particularly for the shipping industry, due to their various advantages over conventional fuels like heavy fuel oil and GH₂. One of the main challenges in the H₂ economy is the lack of adaptability to the existing infrastructure for transport, storage, and use in combustion engines. In this regard, CH₃OH and CH₃CH₂OH exist as liquids at standard temperature and pressure, simplifying their storage and transportation by using existing infrastructure that is largely established for the chemical industry. In contrast, H₂ requires cryogenic temperatures or high pressure for storage and transportation, demanding specialized infrastructure and handling [89]. Also, CH₃OH is a suitable fuel for both Otto and diesel engines with some modifications like glow plugs or pilot fuel, making an easier transition from fossil fuels to a light alcohol economy.

4.1 | Heterogeneous Catalysis for CH₃OH Evolution

CH₃OH is a promising hydrogen carrier and a versatile building block for high value-added chemicals; it has become one of the most attractive molecules for a sustainable economy, with a global annual consumption of approximately 107 million tons. Currently, CH₃OH is industrially produced from syngas at elevated temperatures of 473–533 K and pressures of 50–100 bar over a Cu or Ni alumina (Al₂O₃)-based catalyst (see equations (4) and (5)). In general, CO₂ hydrogenation to CH₃OH is an exothermic reaction that is thermodynamically favorable at low temperatures and high pressure (see equation (4)). However, high temperatures (above 240°C) are required to activate CO₂, which favors the formation of undesired byproducts [90].

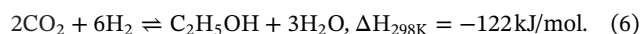


Despite the maturity of CH₃OH synthesis technology, where the most common catalysts are Cu-based catalysts (such as Cu/ZnO/Al₂O₃, CZA) due to their high activity, selectivity, and affordability [81, 87, 90], challenges remain in the development of more efficient and stable catalysts, optimization of reaction conditions, and improvement of the overall economics [77, 87, 91]. One of the main factors influencing the performance of CO₂ hydrogenation to CH₃OH is the choice of the correct catalyst, with the main challenges related to catalyst stability and separation [82].

4.2 | CH₃CH₂OH Synthesis

CH₃CH₂OH is known for its versatility; it is widely used as an industrial solvent, and the CH₃CH₂OH derived from GH₂ has been introduced as a substitute for gasoline. In general, the main renewable industrial production mechanism for CH₃CH₂OH is based on sugar fermentation, converting corn, wheat, rice, and other grains into CH₃CH₂OH. The long-term viability of this process is debatable because it uses food crops as a precursor, leading to increased production costs for both food and fuel. There are also concerns about the environmental impact, as it may contribute to increased greenhouse gas emissions during their production cycle and to the extensive use of pesticides.

An alternative route to produce renewable CH₃CH₂OH is via CO₂ hydrogenation. In general, this process involves two steps: (i) CO₂ hydrogenation to CH₃OH and (ii) CH₃OH carbonylation or dehydrogenation with CO₂, where the overall process can be represented as shown in equation (6).



This procedure is more intricate than CH₃OH synthesis from sugar fermentation and encompasses several reaction steps, contingent on the particular catalyst and reaction conditions used. The synthesis process typically involves the use of catalysts to facilitate the reactions.

In this sense, ongoing research is related to the development of novel catalysts for CO₂ hydrogenation to CH₃OH and CH₃CH₂OH. Transition metals, such as Ru, Ir, Rh, and so on; metal oxides, for example, Cu, In, Zn, and so on; and supported catalysts are frequently used. Therefore, to better understand recent advances and trends in this field, these catalysts can be sorted into unsupported single-atom nanoparticle catalysts and porous supported catalysts such as MOFs, COFs, and metal-oxide supports.

4.3 | Unsupported Catalyst

The study of CO₂ reduction to CH₃OH using unsupported catalysts is mainly focused on evaluating the influence of active catalyst microstructural properties on yield, conversion, and selectivity. Common strategies include engineering surface defects that can form acidic active sites capable of reducing CO₂.

Bordini et al. [92] studied the influence of the crystal phase of In₂O₃ on catalytic CO₂ reduction. They synthesized five different In₂O₃ samples with different crystal phase compositions and specific surface areas. They showed that hexagonal In₂O₃, whether in pure form or as part of a mixed phase, exhibited instability during the CO₂ hydrogenation to CH₃OH, leading to phase transition to the cubic phase and resulting in low conversion efficiency. In this sense, they showed that the cubic phase of In₂O₃ demonstrated higher activity in CO₂ hydrogenation due to higher stability of oxygen vacancies, demonstrating the potential of this phase for achieving efficient CH₃OH production. In the same line, Li and coworkers [93] showed that oxygen vacancies on the exposed facet of cubic In₂O₃ played an important role in the catalytic hydrogenation of CO₂ to CH₃OH, with the (211) crystal phase of In₂O₃ showing outstanding advantages in CH₃OH selectivity. Similar results were obtained by Hu et al. [94], where the sulfur vacancy of MoS₂ favored CO₂ hydrogenation to CH₃OH.

Caldeira et al. [95] showed that the oxidation state of Cu influences the $\text{CH}_3\text{CH}_2\text{OH}$ production. The reduction of Cu-oxides due to the presence of H_2 during the catalytic reaction decreases catalyst activity, directly impacting reaction yield and selectivity. Moreover, the effect of replacing the H_2 source with water steam was investigated; it was observed that CH_3OH production was replaced by $\text{CH}_3\text{CH}_2\text{OH}$ production. This was attributed to greater stabilization of Cu-oxides during the reaction, which favored the formation of $\text{CH}_3\text{CH}_2\text{OH}$ instead of CH_3OH .

4.4 | MOF-Supported Catalyst

In general, MOFs are a class of crystalline porous materials that offer adaptability through large accessible surface areas. The use of MOF as support for active catalysts prevents agglomeration, increasing the availability of active sites. Additionally, there is a strong interaction between active catalyst sites through their secondary building unit, favoring synergistic effects. In this sense, MOF-supported catalysts tend to be more selective to $\text{CH}_3\text{CH}_2\text{OH}$ and more stable under CO_2 reduction reaction conditions.

de Paula et al. [96] studied the impact of the Cu load of a Zr-based MOF (UiO-67) in the CO_2 hydrogenation to CH_3OH and $\text{CH}_3\text{CH}_2\text{OH}$, identifying CH_3OH as an intermediary product during $\text{CH}_3\text{CH}_2\text{OH}$ production. They showed that the use of UiO-67 as support contributed to Cu stability during the reaction. Lower Cu loadings improved selectivity to $\text{CH}_3\text{CH}_2\text{OH}$, while higher loadings resulted in higher H_2 consumption and a wide variety of products. The authors attribute this behavior to changes in the Cu–O–Zr units present on secondary building units with different Cu loadings, indicating that an excess of Cu content not only reduces the structural organization but also

hinders the interaction between the support and the active site, decreasing the overall alcohol production.

4.5 | Metal Oxide Supported Catalyst

The use of metal oxides as support of active catalysts has been widely investigated because they contribute to the stabilization of active catalysts due to strong interactions with active metal catalysts. Metal oxides have good thermal stability and are widely available. Frequently used metal oxide supports include Al_2O_3 , SiO_2 , TiO_2 , CeO_2 , and ZrO_2 , which can be prepared with high surface area, favoring CO_2 adsorption in the active sites. Current research in this type of support points to the optimization of selectivity and yield through changes in composition, crystal structure, and particle size.

CeO_2 -supported catalysts have shown high selectivity for CH_3OH production, mainly attributed to strong MSI and to the ability to reversibly switch between Ce^{3+} and Ce^{4+} oxidation states, which contributes to a synergistic effect between the active catalyst and support. Wang and coworkers [97] studied the effect of CeO_2 support shape on CO_2 hydrogenation to CH_3OH using Cu/ CeO_2 catalyst. They observed a strong dependence on morphology associated with interactions with metal catalysts that control its activity. They showed that using hydrangea-like CeO_2 as support enhanced activity due to the stabilization of Cu^{2+} – O_v – Ce_x species and high specific surface area, contributing to higher CO_2 conversion. In addition to morphology control studies, control of CeO_2 support composition has also been carried out to improve the catalytic performance. In this regard, Xaba et al. [98] investigated the effect of La incorporation into CeO_2 support in the Cu/ CeO_2 catalyst. They showed that the addition of La improved the thermal stability of the catalyst, allowing the use of higher

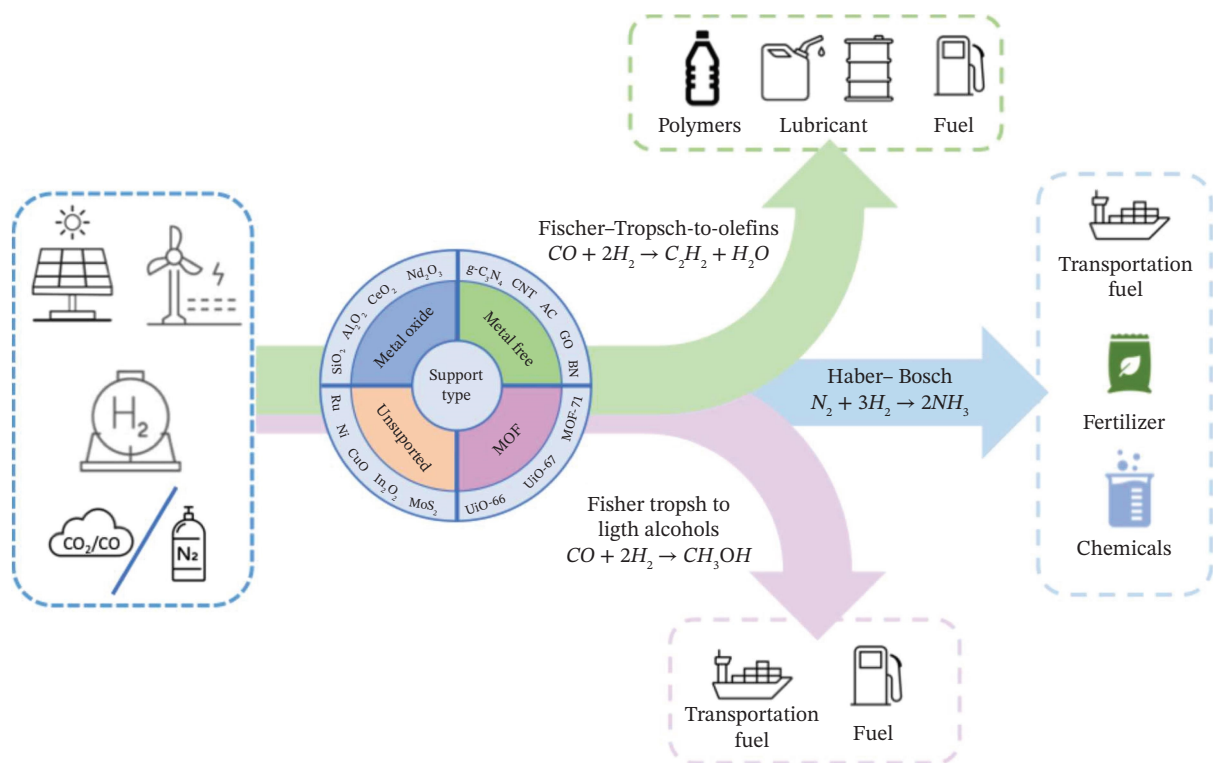


FIGURE 2 | Summary of the GH2 chain of value for the synthesis of energy vectors and the different related technologies.

TABLE 4 | Summarized results for the CO₂ hydrogenation reaction using different catalysts sorted by support type.

Support type	Support	Catalyst	Product	CO ₂ conversion (%)	Selectivity (%)	Yield (g _{cat} ⁻¹ h ⁻¹)	GHSV (mol _{cat} ⁻¹ h ⁻¹)	Temperature (°C)	Pressure (MPa)	Ref.
Unsupported	Unsupported	CuO	MeOH	N/A	N/A	1.31·10 ⁻⁶	4800	220	0.1	[95]
			EtOH	N/A	N/A	3.59·10 ⁻⁵	4800	200	0.1	[96]
		e-In ₂ O ₃	MeOH	17.0%	75.0%	5.86·10 ⁻¹⁺⁴	13,400	200	5	[92]
			MeOH	4.7%	55.0%	8.00·10 ⁻²	8000	280	3	[93]
		Zn-Al-Spined	MeOH	7.5%	15.0%	4.02·10 ⁻²⁺	10,000	240	2	[100]
	FL-Me ₂	ZnZr	MeOH	12.5%	61.3%	1	36,000	280	5	[94]
			MeOH	5.4%	77.2%	3.50·10 ⁻¹	24,000	500	3	[101]
MOF	UiO-67	Cu/UiO-67	MeOH	0.3%	5.0%	7.37·10 ⁻⁴	18,000	260	0.1	[96]
			EtOH	0.3%	2.0%	3.46·10 ⁻⁴	18,000	260	0.1	[96]
	Al ₂ O ₃	Cu/ZnO/Al ₂ O ₃	MeOH	11.8%	49.4%	3.10·10 ⁻¹	10,000	240	5	[94]
			MeOH	15.6%	94.26	1.5·10 ⁻¹⁵	10,000	260	3	[97]
	CeO ₂	PdIn/CeO ₂	MeOH	5.5%	78.9%	3.6	30,000	300	3	[102]
			MeOH	9.0%	77.0%	3.3·10 ⁻²	1350	280	4	[98]
	Ru/In ₂ O ₃	Cu ₁₂ CeO ₄ La ₁₃ O ₂₄	MeOH	10.0%	74.7%	5.9·10 ⁻³	21,000	300	5	[103]
			MeOH	17.7%	70.0%	7.65·10 ⁻¹	21,000	300	5	[104]
	In ₂ O ₃	Ir/In ₂ O ₃	MeOH	5.7%	65.7%	4.7·10 ⁻¹	24,000	300	4	[105]
			MeOH	12.2%	61.0%	4.2·10 ⁻¹	21,000	300	5	[106]
Metal Oxide	CrO ₃	Cu/SiO ₂ -PDVB	MeOH	5.0%	50.55	8.10 ⁻²	6000	300	3	[107]
			MeOH	10.2%	56.46	86.5	6000	240	3	[80]
	SiO ₂	Pd/SiO ₂	MeOH	10.0%	90.96	49.8	1000	150	0.1	[60]
			MeOH	20.0%	90.02	5	3600	300	4.5	[101]
	TiO ₂	Cu ₂ Cu ₂ Pd	MeOH	22.0%	90.08	2.89·10 ⁻²	900	270	4.5	[111]
			MeOH	10.0%	90.59	2.2	900	300	4	[111]
	ZrO ₂	Cu/ZrO ₂	MeOH	8.9%	90.78	2.2	900	300	3	[111]
			MeOH	10.0%	90.78	2.2	900	300	3	[111]
	ZrO ₂	Cu/ZrO ₂ /ZrO ₂	MeOH	10.8%	90.72	1.13·10 ⁻¹	900	300	3	[111]
			MeOH	10.8%	90.72	1.13·10 ⁻¹	900	300	3	[111]
Zeolite	H ₂ SM-5	Cu/ZnZr-Pd	MeOH	8.0%	90.52	1.01·5.9	00072	202	5	[511]
			MeOH	8.0%	90.38	1.01·6.2	0006	202	3	[411]
	H ₂ SM-5	Cu/ZnZr-Pd	MeOH	8.0%	90.38	1.01·6.2	0006	202	3	[66]
			MeOH	8.0%	90.38	1.01·6.2	0006	202	3	[66]
	H ₂ SM-5	Cu/ZnZr-Pd	MeOH	8.0%	90.38	1.01·6.2	0006	202	3	[66]
			MeOH	8.0%	90.38	1.01·6.2	0006	202	3	[66]
	H ₂ SM-5	Cu/ZnZr-Pd	MeOH	8.0%	90.38	1.01·6.2	0006	202	3	[66]
			MeOH	8.0%	90.38	1.01·6.2	0006	202	3	[66]
	H ₂ SM-5	Cu/ZnZr-Pd	MeOH	8.0%	90.38	1.01·6.2	0006	202	3	[66]
			MeOH	8.0%	90.38	1.01·6.2	0006	202	3	[66]
Metal free	Cu ₂ N ₄	Cu ₂ N ₄	MeOH	10.0%	90.56	1.35·10 ⁻¹	N/A	150	3.2	[811]
			MeOH	10.0%	90.56	1.35·10 ⁻¹	N/A	150	3.2	[811]
	Cu ₂ N ₄	Cu ₂ N ₄	MeOH	10.0%	90.56	1.35·10 ⁻¹	N/A	150	3.2	[811]
			MeOH	10.0%	90.56	1.35·10 ⁻¹	N/A	150	3.2	[811]
	Cu ₂ N ₄	Cu ₂ N ₄	MeOH	10.0%	90.56	1.35·10 ⁻¹	N/A	150	3.2	[811]
			MeOH	10.0%	90.56	1.35·10 ⁻¹	N/A	150	3.2	[811]
	Cu ₂ N ₄	Cu ₂ N ₄	MeOH	10.0%	90.56	1.35·10 ⁻¹	N/A	150	3.2	[811]
			MeOH	10.0%	90.56	1.35·10 ⁻¹	N/A	150	3.2	[811]
	Cu ₂ N ₄	Cu ₂ N ₄	MeOH	10.0%	90.56	1.35·10 ⁻¹	N/A	150	3.2	[811]
			MeOH	10.0%	90.56	1.35·10 ⁻¹	N/A	150	3.2	[811]

*Values estimated from published data.

temperatures during the CO₂ hydrogenation reaction without observing the sintering effects of the catalyst. This effect was attributed to the formation of smaller CeO₂ crystallites due to La incorporation; this increased the interfacial sites (Cu²⁺-O_v-Ce_x) related to CO₂ adsorption and later reaction with H₂ over Cu nanoparticles.

ZrO₂-supported catalysts have attracted significant attention as potential alternatives to commercial CZA due to its promising redox properties that can facilitate the activation and dissociation of CO₂. In this field, Xin et al. [99] investigated an alternative route for the cost-effective synthesis of CuO/ZnO/ZrO₂ catalyst, showing a high CH₃OH production from CO₂ hydrogenation. This good performance was attributed to the reactive interface between the Cu species and ZnO/ZrO₂ support and the high specific surface area of the catalyst. Current research has explored the impact of ZrO₂-supported catalyst morphology and composition on the catalytic activity.

Table 4 presents the different catalysts considered for the light alcohol synthesis. The data are organized by support type and their reaction characteristics, such as CO₂ conversion, selectivity, yield, and reaction conditions.

Finally, Figure 2 shows a GH2 chain of value for the synthesis of energy vectors reviewed in this work, including light alcohols, NH₃, and olefins.

5 | Conclusion

The catalytic synthesis of energy vectors from GH2 is not merely a technical exercise in catalyst design; it is a strategic decision that must align energy density, infrastructure availability and compatibility, and end-use application. This work demonstrates that no single vector universally outperforms the others; instead, each occupies a distinct niche in the emerging GH2 economy. The energy vectors included in this work:

- **NH₃** emerges as the superior vector for long-distance transport and long-term seasonal storage. With a volumetric hydrogen density 1.7 times higher than liquid hydrogen and an existing global storage and distribution infrastructure, NH₃ leverages established supply chains for fertilizer transport. However, its reversion to hydrogen at the point of use imposes severe energy penalties. From a techno-economic perspective, NH₃ consistently emerges as the costliest vector on a full-chain basis, being the most expensive among major e-fuels across production, storage, and utilization stages, while CH₃OH demonstrates moderately lower costs and hydrogen remains the cheapest.
- **CH₃OH**, conversely, occupies the middle ground. CH₃OH offers the highest technological maturity among synthetic fuels for direct use in internal combustion engines. The established global CH₃OH market and distribution network, already integrated into chemical logistics, make CH₃OH a near-term solution for the decarbonization of the transport industry. Critically, CH₃OH can be utilized directly without cracking to hydrogen, eliminating reconversion losses entirely.
- **Light olefins** serve a fundamentally different purpose; they are not primarily energy carriers but chemical building blocks for polymers, lubricants, and synthetic fuels. The FTS

synthesis of C₂-C₄ olefins from GH2 represents a pathway to defossilize the petrochemical industry rather than to store energy. Their value lies in displacing fossil-derived feedstocks.

The strategic justification for the use of energy vectors cannot be based on thermodynamic criteria; it must be logistical. Vectors are chosen when direct hydrogen use is infeasible due to transport distance, storage unavailability, or end-use incompatibility. NH₃ and CH₃OH transport by ship are mature options for overseas energy trade. In contrast, liquid hydrogen exhibits the lowest efficiency among major pathways, requiring substantial development before large-scale deployment becomes viable.

Across all three vector classes, common themes emerge in catalyst development. For FTO synthesis, Fe-based catalysts operating at 220°C–340°C (HTFT) remain the industrial standard for C₂-C₄ production, while Co-based systems at 220°C–280°C (LTFT) favor long-chain hydrocarbons and waxes. The new findings of Co₂C nanoprisms, which could present a highly selective phase for lower olefins, could represent a genuine mechanistic breakthrough.

For NH₃ synthesis, the contrast between academic innovation and industrial reality is stark. For example, MOFs (UiO-66, MIL-101), electrified-supported catalysts (BaAl₂O_{4-x}H_y), and plasmonic photocatalysts have demonstrated proof-of-concept for mild-condition NH₃ synthesis (< 400°C, 1 MPa); their reported yields remain orders of magnitude below commercial viability. Meanwhile, the conventional Haber-Bosch (HB) process with Fe catalysts is still the most relevant industrial technology for the NH₃ synthesis. Ru-based catalysts supported on rare-earth oxides show enhanced activity under milder conditions (~400°C, 1 MPa), but Ru scarcity and price remain prohibitive for large-scale deployment.

For CO₂ hydrogenation to light alcohols, CZA remains the industrial benchmark for CH₃OH synthesis, operating at 200°C–260°C and 50–100 bar. Yet emerging systems such as In₂O₃-based catalysts and MOF-confined metal nanoparticles (MNPs@MOFs) have demonstrated superior CH₃OH selectivity (> 90%) at lower temperatures (150°C–200°C), though long-term stability under realistic feed conditions containing trace H₂O and O₂ remains unproven. For CH₃CH₂OH synthesis, the pathway remains stubbornly inefficient: Direct CO₂ hydrogenation to CH₃CH₂OH yields at most 30% selectivity, and no catalyst system reviewed achieves industrially relevant productivity.

Integrating the evidence presented, it can be concluded:

- The choice of energy vector must be dictated by end-use, not by catalyst performance alone. NH₃ is optimal for long-term storage and maritime fuel; CH₃OH is optimal for drop-in fuel applications and near-term decarbonization; light olefins are essential for chemical feedstocks. No single catalyst system serves all purposes, and no single vector dominates all applications.
- Techno-economic viability outweighs catalytic activity as the rate-limiting factor for commercialization. The lowest levelized costs are achieved with hydrogen, followed by CH₃OH, with NH₃ being the most expensive across full-chain assessments. The advances in catalysis technology

must prioritize materials that operate at low reaction conditions, for instance, $< 250^{\circ}\text{C}$ and < 50 bar, to integrate with low-grade renewable heat and reduce capital expenditures.

- The gap between laboratory discovery and industrial application remains vast across all three vector classes. For instance, plasmonic photocatalysts for NH_3 synthesis operate at production rates two orders of magnitude below commercial relevance. MOF-supported catalysts for CH_3OH synthesis degrade within hundreds of hours under reaction conditions. Co_2C nanoprisms for FTO, despite exceptional selectivity, have not been demonstrated beyond the laboratory scale.
- The integration of GH2 into existing chemical infrastructure presents both an opportunity and a challenge. Existing HB processes and CH_3OH synthesis plants can be retrofitted to accept GH2, advancing in the decarbonization without catalyst replacement.

The catalytic synthesis of energy vectors from GH2 represents a necessity. The efficiency penalty associated with NH_3 and CH_3OH storage is not a failure of catalysis; it is the cost of decoupling energy generation from energy consumption in time and space. Where direct hydrogen transport is feasible, it should be prioritized. Where it is not, NH_3 and CH_3OH offer mature, scalable alternatives with existing infrastructure.

Future progress will depend less on discovering novel catalyst compositions and more on rational design informed by *operando* characterization, techno-economic integration with renewable hydrogen production, and systematic benchmarking under industrially relevant conditions. The hydrogen economy is not a single pathway but a portfolio of solutions. Catalysis provides the tools; strategic deployment determines the outcome.

6 | Outlook

Regarding the energy vectors reviewed in this study, NH_3 will dominate long-distance energy trade due to its logistical advantages over other energy vectors. The existing global infrastructure for fertilizer transport positions NH_3 as the primary vector for intercontinental renewable energy shipping. Advances in NH_3 synthesis technology that can enhance its competitiveness include catalyst development for mild-condition decomposition and electrochemical NH_3 synthesis.

CH_3OH offers the most near-term deployable solution for transport decarbonization. With direct-use capability in internal combustion engines, CH_3OH technology requires advances in low-temperature catalysts and direct hydrogenation of CO_2 .

Light olefins are chemical feedstocks but not energy carriers. Their future lies in petrochemical defossilization, not energy storage; thus, they have an indirect impact on the energy sector. Advances in the engineering of new catalysts with high selectivity for C_2 – C_4 olefins will be a step forward in the development of this technology, with scale-up from laboratory to pilot plant being the critical next step.

The existing technologies for NH_3 synthesis (HB process) and CH_3OH synthesis can accept GH2 without catalyst replacement. This is the lowest-capital pathway to decarbonization and should be prioritized over greenfield projects. Direct hydrogenation of

CO_2 to higher alcohols (e.g., $\text{CH}_3\text{CH}_2\text{OH}$) remains a technology under development.

From this study, it is understood that no single vector will dominate all applications. The future GH2 industry will be a portfolio:

- Short-distance, continuous demand $\rightarrow$ Direct hydrogen (pipelines, compressed gas)
- Long-distance, seasonal storage $\rightarrow$ NH_3 (maritime, power generation)
- Transport, drop-in applications $\rightarrow$ CH_3OH (shipping, heavy-duty vehicles)
- Petrochemical feedstocks $\rightarrow$ Light olefins (plastics, lubricants, synthetic fuels)

Additionally, the rate-limiting factor is no longer catalyst discovery; it is the integration of technology, where catalysts must shift from reporting novel compositions to systematic understanding of active sites under *operando* conditions. The gap between laboratory activity and industrial viability remains vast. Future progress will depend on *operando* characterization to understand deactivation mechanisms and techno-economic integration with renewable H_2 production.

The hydrogen economy is not a single pathway but a portfolio of solutions. Catalysis provides the tools; strategic deployment guided by logistics, economics, and infrastructure compatibility, not thermodynamic efficiency alone, will determine the outcome. The future of the GH2 industry must be about the *operando* characterization of catalysts, the rational design of a new generation of catalysts, and the integration of science and engineering.

Author Contributions

S. D. Rojas: conceptualization, writing–original draft, visualization, writing–review and editing. M. A. Cisternas: writing–original draft, writing–review and editing. M. J. Retamal: writing–original draft, visualization. J. G. Moya-Cancino: conceptualization, formal analysis, writing–original draft, writing–review and editing.

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Disclosure

We confirm that all figures are original and have not been published elsewhere.

Conflicts of Interest

The authors declare no conflicts of interest.

Data Availability Statement

The data used to support the findings of this study are available from the corresponding author upon request.

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