

Limitations of Ammonia as a Hydrogen Energy Carrier for the Transportation Sector

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Annual global carbon dioxide (CO_2) emissions reached 34.2 gigatonnes (Gt) in 2019 as a result of extensive and unrestricted use of fossil fuels to fulfill $\sim 80\%$ of society's energy needs at the current level of ~ 585 exajoules (EJ)/year.^{1,2} Transportation that provides mobility to passengers and freight is responsible for approximately 25% of the overall CO_2 emission.^{3,4} Considering the current rate of population growth and associated increases in energy consumption, it has been projected that the corresponding global energy demand will be increased by at least 50% before 2050.^{1,2,5} To meet such needs while minimizing the environmental impacts by curtailing anthropogenic CO_2 emissions, large-scale deployment of low-carbon renewable energy (RE) is necessary.^{6–8} Despite a moderate increase in the overall share of RE in the current energy landscape, recent studies indeed indicated that a full transition to 100% RE is attainable within the next 3 decades or so with a cost-efficient vision of deep electrification of heat and transportation sectors around the globe.^{9–11} Thus, this energy transition is no longer a matter of technical feasibility or economic viability, but political will.¹²

One of the most critical enabling factors for the RE future is a means of storing and transporting RE at a multi-EJ scale to manage the energy system by peak shaving and valley filling, to overcome the issues of intermittency of solar and wind energies, etc., and to serve regions where RE is difficult to produce due to economic and environmental constraints.¹³ The choice of the storage system is highly application dependent, based on the type (stationary or mobile), scale, time, cost, safety, etc. Storing RE in the form of chemical fuels has been considered logical for both short- and long-term storage, particularly for use in the transportation sector.^{14,15} In this regard, hydrogen (H_2) is a promising energy vector for its efficient utilization in fuel cells (FCs).^{16–18} Although liquid H_2 (LH_2) offers advantages for easy conversion into gaseous H_2 , the liquefaction step consumes almost 30–40% of the energy content of H_2 in addition to boil-off losses during transportation.^{19,20} The necessary storage and shipping infrastructures for long distances are yet to be developed.¹⁸ To circumvent the inherent issues with LH_2 storage and transport, several other carbon-based fuels such as methanol (CH_3OH), methylcyclohexane (MCH), etc. as well as ammonia (NH_3) have been recommended as more technically viable options, certainly because of their desired properties (Table 1).^{21,22} Compared to LH_2 , NH_3 can be easily liquefied either by increasing the pressure to ~ 10 bar at room temperature or by cooling down to -33°C under 1 atm. Moreover, NH_3 is safe

and easy to store and transport because of its low vapor pressure and high boiling point. The H_2 content in NH_3 (17.65%) is higher than those of methanol (MeOH, 12.5%) and methylcyclohexane (MCH, 6.1%) (Table 1), and its volumetric energy density (12.92–14.4 MJ/L) is comparable to that of MeOH (11.88 MJ/L) but significantly higher than those of MCH (5.66 MJ/L) and lithium-ion batteries (0.9–2.63 MJ/L) when the lower heating value (LHV) of H_2 is considered. With the mature NH_3 production, storage, and transportation infrastructures, NH_3 has been recognized as a sustainable H_2 and energy carrier for the future for both mobile and stationary applications.

NH_3 is currently the second most highly produced chemical in the world, with a global manufacturing capacity of ~ 230 million tonnes (Mt) per year.^{26–28} Currently, ~ 180 Mt of NH_3 is produced annually, predominantly through the steam methane reforming (SMR) process to generate desired H_2 , followed by industrial NH_3 synthesis (*vide infra*).^{29,30} NH_3 has an annual market value of $\sim \text{US\$}70$ billion.³¹ NH_3 is primarily used in the production of fertilizers (80%), in addition to immense applications in the manufacture of explosives (5%) and other materials and chemical commodities (15%).³² Its production has been one of the most energy-intensive industrial chemical processes, demanding around 200–400 GJ/ton NH_3 for Birkeland–Eyde and cyanamide processes in the early 1900s, followed by the Haber–Bosch (H-B) process which consumed around 100 GJ/ton NH_3 at early stages.³³ While the efficiency of the H-B process has been improved considerably (30–60 GJ/ton NH_3) by the integration of efficient SMR process instead of coal gasification, the process still demands 1–2% of global energy.^{29,31,32,34–36} As a consequence, it is one of the biggest emitters of industrial CO_2 (1–1.5% of anthropogenic CO_2 emissions).³⁷

Since H_2 production by SMR accounts for 80% of energy during the NH_3 production, numerous efforts have been focused on the development of the low-temperature electrochemical synthesis of NH_3 . However, the state-of-the-art

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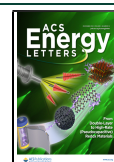


Table 1. Characteristics Comparison of Selected Chemical Fuels^{23–25}

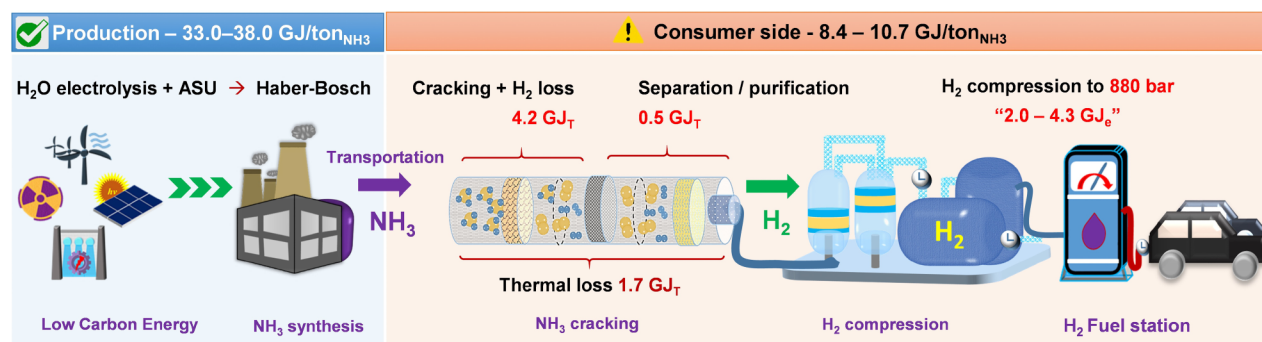
properties	units	H ₂	CH ₃ OH (+H ₂ O)	MCH	NH ₃
phase		liquid	liquid	liquid	liquid
density	kg/m ³	70.8	792	770	610 ^a /680 ^b
boiling point	°C	−253	65 (100)	101	−33
volumetric H ₂ content	kg _{H₂} /m ³	70.8	99 (95.04)	47.1	107.7/120
volumetric energy density ^c	MJ/L	8.49	11.88 (11.40)	5.66	12.92/14.4
gravimetric H ₂ content	wt%	100	12.5 (12)	6.1	17.65
gravimetric energy density ^d	MJ/kg	120	15 (14.4)	7.35	21.18
enthalpy of dehydrogenation	kJ/mol _{H₂}	0.907	16.3	69.8	30.6
enthalpy of evaporation	kJ/mol _{H₂}	—	31.7 ^e (13)	10.8	15.1
H ₂ release		evaporation	reforming (250 °C)	dehydrogenation (350 °C)	cracking (>425 °C)
explosive limit in air	vol%	4–75	6.7–36	1.2–6.7	15–28
flammability/toxicity		highly flammable	toxic	toxic	toxic

^aAt 20 °C and 10 bar. ^bAt −33 °C. ^c, ^dValues are the corresponding hydrogen energy densities, calculated based on the LHV of a stoichiometric amount of hydrogen (LHV_{H₂} = 120 MJ/kg). ^eFor steam reforming of methanol, one must evaporate both methanol and water. In a real case, a stoichiometric excess of 50% water is typically used.

Table 2. Energy Balance (GJ/ton_{NH₃}) and Efficiencies (%) of Various Hydrogen Energy Options

carrier	PTF processes	PTF efficiency (%)	TE ^a (%)	carrier to H ₂ ^b efficiency (%)	PTFC ^c (%)	FCTP ^d (%)	PFP ^e (%)
NH ₃	electrolyzer/H-B/SOFC	55.7–64.3	90	—	50.1–57.9	50–65	25.1–37.6
NH ₃	electrolyzer/H-B/PEM	55.7–64.3	90	61.0–68.5	30.6–39.6	46–60	14.1–23.8
LH ₂	electrolyzer/liquefaction/PEM	49.3–57.9	84	—	41.4–48.6	46–60	19.0–29.2

^aTransport efficiency. ^bH₂ generation from carrier. ^cPower-to-fuel cell. ^dFuel cell-to-power. ^ePower-to-fuel-to-power.

Figure 1. Power-to-power (PTP) energy consumption of NH₃ as a hydrogen energy carrier.

production rate needs to be increased at least by 1–2 orders of magnitude for practical applications.³⁸ With the development of various efficient electrolyzers, such as polymer electrolyte membrane (PEM), solid oxide electrolyzer cells (SOECs), and alkaline electrolyzer cells (AECs), electrochemical H₂ production and its subsequent use in the H-B process were considered as an attractive low-carbon pathway for the large-scale production of so-called “green NH₃”.³⁹ Although alkaline electrolysis in electrolyzers is known to be one of the easiest methods for on-site high-purity H₂ generation with well-established technology, its low current density makes this technology impractical for continuous H₂ generation in large quantities, as would be required to drive a vehicle.⁴⁰

In practice, PEM electrolyzers are employed for H₂ production due to their high efficiency and better lifetime under ambient conditions. In general, electrolyzers demand a theoretical minimum of 21.18 GJ/ton_{NH₃} (based on the LHV of the H₂ content in NH₃). However, at the industrial scale, the electrolyzers operate at an efficiency of 60–70%, requiring at least 30.3–35.3 GJ/ton_{NH₃}. Furthermore, the production of

nitrogen (N₂) by an air-separation unit (ASU) and H-B loop compressors using current electrified technology consumes around 2.7 GJ/ton_{NH₃}, representing a significant saving compared to the methane-fed H-B process (6.9 GJ/ton_{NH₃}).³² Therefore, on average, the ideal NH₃ production by this pathway costs 33.0–38.0 GJ/ton_{NH₃}, for an overall power-to-fuel (PTF) efficiency of 55.7–64.3% (Table 2). By contrast, the PTF efficiency of 49.3–57.9% for LH₂ is lower than that of NH₃ due to the high energy demand of 36.0–48.0 GJ/ton_{H₂} for compression and liquefaction.⁴¹ The transportation efficiency (TE) of H₂ carriers has been evaluated for a distance of 12 000 km by considering the propulsion engine and dead weight tons of liquefied natural gas tankers (Table 2).⁴² In both cases, the fuel and energy demands of the ship are supplied by the combustion of hydrogen energy carriers being transported. The TE of LH₂ (84%) is lower than that of NH₃ (90%) due to the boil-off loss and high energy demand for compressed storage.

In order to generate electrical power from NH_3 by feeding it into FCs, two pathways are envisioned. One is catalytic cracking of NH_3 to generate H_2 for fuel cell applications, viz., PEM fuel cells (PEMFCs) and alkaline fuel cells (AFCs). This process has been proposed as a major RE distribution mechanism to supply green NH_3 worldwide for H_2 refilling stations, as the direct on-board cracking is deemed impractical (Figure 1).⁴³ The cracking process requires high temperatures of $>500^\circ\text{C}$ for the production of high-purity H_2 ($>99.97\%$, particularly for vehicle applications), which demands thermal energy (GJ_T) of $4.2 \text{ GJ}_\text{T}/\text{ton}_{\text{NH}_3}$ (including H_2 loss).⁴⁴ Since PEMFCs are highly vulnerable to the trace amounts of NH_3 ($<0.1 \text{ ppm}$) in H_2 , NH_3 conversion to H_2 must be conducted with a highly efficient purification and separation system, which consumes an additional $0.5 \text{ GJ}_\text{T}/\text{ton}_{\text{NH}_3}$. AFCs, on the other hand, are also sensitive to traces of CO_2 (present in the air), affecting cell operation, thus demanding pure O_2 , which increases the cost of operation. As a result, these separation and purification processes would inevitably incur substantial cost.⁴⁵ The energy-intensive nature of these systems can result in an overall thermal loss of $1.7 \text{ GJ}_\text{T}/\text{ton}_{\text{NH}_3}$. More importantly, additional electrical energy of $2.0\text{--}4.3 \text{ GJ}_\text{e}/\text{ton}_{\text{NH}_3}$ is required for the compression of H_2 to 880 bar for refilling fuel cell electric vehicles (FCEVs) at 700 bar.^{21,46,47} Thus, the heat and electricity requirements for the cracking and compressing processes per unit of NH_3 demand ~ 0.3 and $0.16\text{--}0.34$ (assuming a fuel cell efficiency of 60%) units of NH_3 , respectively, resulting in an overall conversion efficiency of 61.0–68.5% at the point of use. Moreover, the integration of an energy-intensive cracking reactor with a H_2 compression system may complicate the fueling and refilling processes on the consumer side. These limitations can further escalate due to the intricacy of the cracking system and the performance and lifetime of catalysts in the presence of impurities.⁴³

The other pathway of direct utilization of NH_3 in fuel cells appears to be advantageous. While direct ammonia fuel cells (DAFCs) are still at low technology readiness levels (TRLs),^{48–50} solid oxide fuel cells (SOFCs) can be deployed in the near future, as the NH_3 cracking occurs internally within the SOFC; thus, the requirement for a H_2 separation system can be evaded.^{29,23,51} However, the high operation temperatures ($550\text{--}900^\circ\text{C}$)²³ suggest that SOFCs may only be suitable for continuous stationary applications without frequent on-and-off cycles.²¹ Hence, SOFCs may be applied in heavy-duty vehicles like those used in aviation, shipping, trucking, etc., where frequent on–off cycles or fast starts are not mandatory, as they are for light-duty vehicles (cars, motorcycles, taxi, and buses).⁵² Furthermore, the anode materials, responsible for the catalytic decomposition of NH_3 to H_2 , should be stable, durable, and tolerant to high temperatures during FC operations, because anode degradation still represents a major hurdle for the commercialization of SOFCs.⁵³ Of late, Minutillo et al. proposed a novel plant configuration, based on the NH_3 -fueled SOFC technology, for the simultaneous on-site co-generation of H_2 and electricity in refueling stations.⁵⁴ However, further improvement in the SOFC and NH_3 cracking technology is imperative for materialization of such concepts. The use of NH_3 in combustion engines is left out of the current discussion because this technology does not require H_2 production from NH_3 and can lead to significant problems such as difficult ignition, low flame speed, higher compression, etc. in addition

to NO_x emissions from combustion of pure NH_3 or ammonia-fuel blends.⁵⁵

While based on these ideal fuel production efficiencies (note: these numbers are higher than those of the state-of-the-art technologies) and actual transportation analysis, and NH_3 indeed has a great potential as a viable energy storage option, the large-scale decarbonization of the transportation sector by employing NH_3 as a H_2 carrier does not offer clear advantages compared to those of LH_2 in terms of overall power-to-fuel-to-power (PFP) efficiency (Table 2). The requirement of a significant amount of energy for the cracking process restricts its use for H_2 release. Moreover, both the financial and energy costs for purifying and compressing the released H_2 to fill the tank of a FCEV are significant, where the invested electrical and thermal energy is difficult to recover during on-boarding applications. In addition to its technical challenges, the toxicity (OSHA exposure limit of 50 ppm), hydrophilicity, and corrosive nature of NH_3 call for a leak-proof infrastructure to avoid accidental release and equipment corrosion and to encourage social approval. In our opinion, NH_3 may serve as an e-fuel for stationary electricity generation with SOFCs to supply power in regions where RE is difficult to produce and grid extensions cannot reach, but it has a limited role as a H_2 carrier due to the large energy requirement for cracking and compressing at the customer end. Importantly, to make a meaningful contribution toward climate change mitigation, it is more effective to prioritize the use of green NH_3 production to decarbonize the current fossil-fuel based NH_3 industry at the scale of 180 Mt per year.^{29,32}

Sudipta Chatterjee  orcid.org/0000-0003-4977-3840

Rajesh Kumar Parsapur  orcid.org/0000-0002-0042-8942

Kuo-Wei Huang  orcid.org/0000-0003-1900-2658

AUTHOR INFORMATION

Complete contact information is available at:
<https://pubs.acs.org/10.1021/acsenerylett.1c02189>

Author Contributions

[†]S.C. and R.K.P. contributed equally.

Notes

The authors declare no competing financial interest. Views expressed in this Viewpoint are those of the authors and not necessarily the views of the ACS.

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