Ammonia: a Renewable Energy Carrier

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Contributor: I. M. R. Fattah

Ammonia is among the most commonly shipped bulk-produced chemicals, marketed for more than a decade in mass all over the globe. Originally used in the chemical industries and as an intermediate for the production of fertilisers, ammonia has also been explored recently as a hydrogen storage media and a substitute fuel for hydrocarbon. Unlike the conventional ammonia production process that used natural gas as a feedstock and is responsible for carbon emission, ammonia is a means of renewable energy storage formulated from hydrogen generated by an electrically driven electrolyser and nitrogen separate from the atmospheric air.

ammonia renewable energy storage hydrogen storage

1. Introduction

Industrial activity in parts of the world often has several direct and indirect adverse environmental consequences. Carbon dioxide (CO_2), a natural greenhouse gas (GHG) that helps keep the globe warm, is out of control and triggering a climate crisis. This anthropogenic emission endangers human health, agriculture, natural ecosystems and atmospheric stability [1]. As reported in the Intergovernmental Panel on Climate Change (IPCC) Climate Change Mitigation Report (2014), CO_2 is the main contributor to GHG emissions, which have 76% (including 11% of forests and land use) of the overall share, while CH4and N2O accounted for 16% and 6%, respectively [2].

A growing number of international reports illustrates the health and environmental effects of GHG emissions [3][4][5]. The ultimate goal is to maintain GHG concentrations at a point where the dangerous effect of climate change can be prevented. Unlike the United Nations Framework Convention on Climate Change (UNFCCC), the Kyoto Protocol binds parties from a developed country to reduce GHG emissions [6]. Recently, under the terms of the Paris Agreement of 2015, the United Nations (UN) has committed itself to a long-term target to keep temperatures below 2 °C, compared to pre-industrial levels, to avoid the worst consequences of global warming

In the last decades, the world has put great effort and investment into developing renewable energy to decarbonise the economy. The global pandemic disruption caused by COVID-19 teaches humanity that reducing the proportion of fossil fuels in human activity will substantially increase the regeneration of the Earth’s atmosphere due to a reduction in air pollution. Therefore, it is necessary to have an affordable and efficient method for storing energy. To store and distribute H_2 efficiently, other indirect storage media such as ammonia (NH3), with its proven transportability and high flexibility, is required [7].

For example, Japan’s Cross-Ministerial Strategic Innovation Program (SIP) attempts to show ammonia, hydrides and hydrogen as essential elements of the nation’s hydrogen energy system [8]. The Japanese Ministry of Economy, Trade and Industry (METI) described ammonia in conjunction with “the concept of importing renewable energy produced in other countries” [9]. Governments in New Zealand and Australia have announced federal grants to support the development of ammonia plants driven by renewable energy. Similarly to Japan, the country is also looking into the possibility of exporting renewable energy in the form of ammonia.

Today, with a global production rate of more than 176 million metric tonnes of ammonia, the chemical is being used as fertiliser and a building block in the manufacture of many products [10][11]. With 28.5% of global production, China is known as the main producer of ammonia [12]. The uses of ammonia as an intermediate for the production of fertilisers account for over 80% of the total production of ammonia [13]. Furthermore, ammonia has also been researched as a source of energy for fuel cells, transport, industry and power generation [14].

Unfortunately, the current industrial ammonia synthesis method is complicated, energy-intensive and heavily dependent on hydrocarbon. The Haber–Bosch process that is currently used to synthesise ammonia is responsible for almost 11% of global industrial CO2emissions In addition, the nature of renewable energy sources, which are irregular, requires turnkey systems that can be instantly switched on and off [15]. Thus, the challenge for the global deployment of ammonia as energy storage is, therefore, the simpler and more efficient production of ammonia from abundant sources, such as ambient air and water, with a ready to go system, which can be driven by intermittent energy sources.

The number of research studies on renewable ammonia, its innovation on the production route and its performances as a fuel has increased substantially in recent years, as shown by a growing number of scientific papers and review papers (**Figure 1**a–c). There has been a significant increase in the number of scientific articles related to ammonia, renewable energy, energy storage or energy carriers in recent years. For innovative approaches of ammonia synthesis, electrochemistry gained great attention in recent years due to the direct conversion of electricity into ammonia. Of all the devices capable of converting ammonia into energy, the Internal Combustion (IC) engine appears to receive the most development effort, noting that use for other applications is less explored.

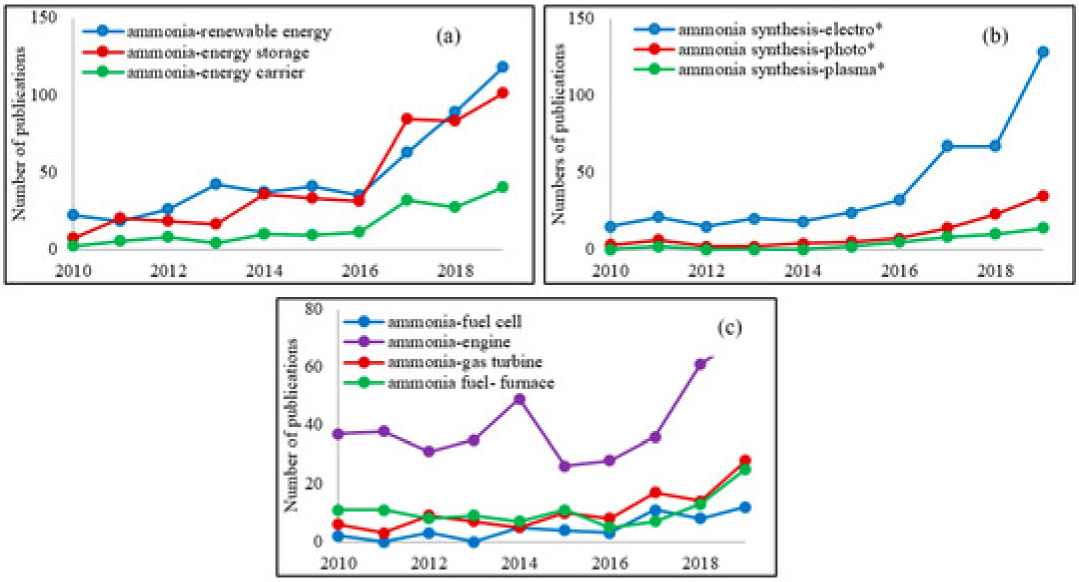

Figure 1. Recent publications of (**a**) ammonia as a renewable energy carrier, (**b**) innovations in ammonia production, (**c**) ammonia as a fuel in Scopus databases.

2. Key-Driver of Ammonia Economy

The issue of the storage and distribution of hydrogen creates opportunities for ammonia to be seen as alternative storage of renewable energy. The previous study on the potential hydrogen storage material by Kojima [16] has revealed that ammonia has the highest gravimetric densities with the highest volumetric densities, as shown in **Figure 2** [7][16]. Consequently, attempts are being made to leverage ammonia over others to replace hydrogen as a central energy distribution block [7]. In addition, ammonia can be used to replenish soil nutrients, boosting the growth of crops and accelerating afforestation that will indirectly help to balance CO2gas in the atmosphere through photosynthesis by plants.

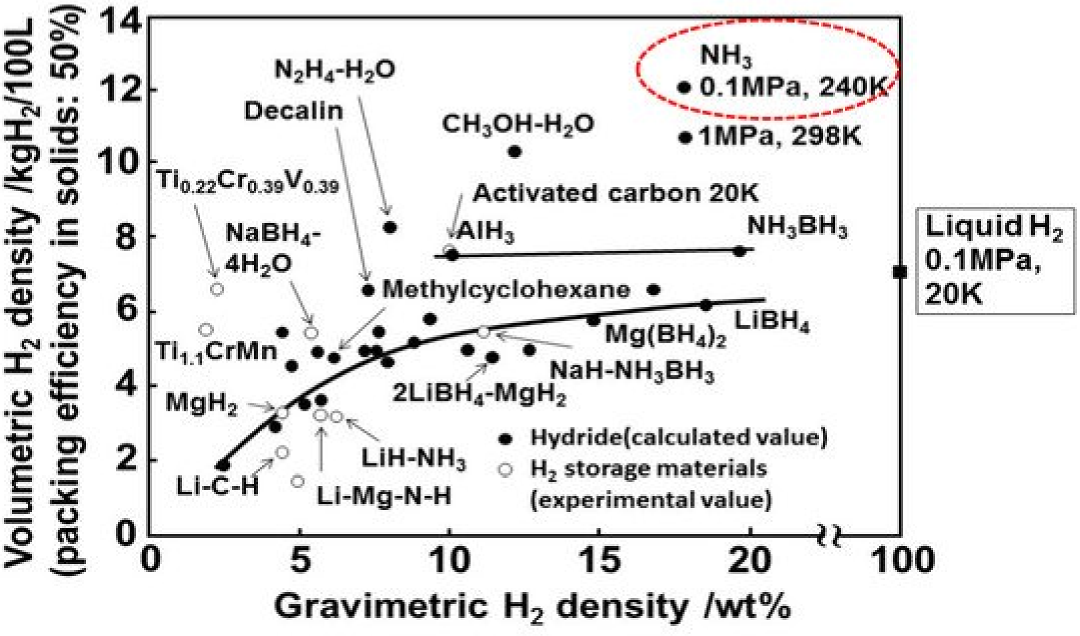

Figure 2. The density of hydrogen in hydrogen carriers [16].

The sustainable future of renewable energy-driven ammonia production is not a novel idea but has never been widely embraced over methane or coal-based ammonia. By transforming into ammonia that can be liquefied under moderate pressure, renewable energy can be transported from places where renewable energy is cheap or excessive to where it is limited or expensive. This synergy could open up opportunities for exports and imports of renewable energy, similar to today's hydrocarbon fuel. In addition to being used directly in the form of ammonia, this may also be dissociated into its component for use as hydrogen fuel at a relatively low cost.

Unlike hydrogen that cannot be liquefied under a pressurised tank, ammonia may be kept in liquid form when at least 8.58 bar is maintained. Commonly, long-distance ammonia transport is accomplished by using carbon steel pipelines that are opposed to hydrogen, which now still has material issues with the pipeline. For transporting ammonia via pipeline over 1610 km, it requires 1119 kJ/kg-H2, which is much lower than that of hydrogen transport of 14,814 kJ/kg-H2. This disparity can be described by the state of the two fluids where hydrogen gas is distributed with the aid of compressors while the ammonia is carried as a liquid using a pump.

Such a tank could hold up to 26 tonnes or 600 GJ of energy of ammonia [17]. In contrast, a hydrogen lorry only can be used to transport around 340 kg of hydrogen gas at 17.91 MPa, equivalent to 48 GJ of hydrogen energy content, while transporting in liquid form in a hydrogen trailer could hold around 3.9 tonnes of hydrogen, equal to 553 GJ of energy [18]. Ammonia can be transported by ship or barge using pressurised storage vessels. By using these vessels, existing ocean-going ships can transport 55,000 tonnes of ammonia [19].

In the recent development, storing ammonia as metal ammine complexes, i.e., hexaamminemagnesium chloride, $Mg(NH_3)6Cl_2$ (**Figure 3**), also gives a beneficial advantage for storing and transporting since it has very low vapour pressure (0.002 bar at ambient temperature) Hexaamminemagnesium chloride is formed simply bypassing ammonia at room temperature over anhydrous magnesium chloride. The amine has volumetric hydrogen content between 105 and 110 kg H_2 m^{-3} and gravimetric hydrogen content over 9 wt.% [20].

Figure 3. $Mg(NH_3)_6Cl_{l2}$ for ammonia storage [21].

3. Ammonia as a Renewable Fuel

In an early study, ammonia, which has 17% hydrogen by weight, was proposed for use in PEM fuel cells. Though, due to the low operating temperature of the PEM fuel cell, the thermodynamics decomposition of ammonia cannot occur [22]. On top of that, ammonia is lethal to the Nafion membrane utilised in PEM fuel cells. Thus, external cracking reactors are required to completely convert ammonia into hydrogen, giving an extra energy input and additional costs [23].

Based on electrolyte and reaction, a direct ammonia-fed fuel cell can be divided into three major systems, alkaline fuel cell (AFC), alkaline membrane fuel cell (AMFC) and solid oxide fuel cells (SOFC). Ammonia has been reported as a feed for a fuel cell as early as the 1960s based on the alkaline fuel cell developed by Francis Thomas Bacon [24]. The cells use alkaline electrolytes such as potassium hydroxide (KOH) and platinum cathodes. Unfortunately, the use of KOH and NaOH is not favourable for air-intake fuel cells since it reacts with CO2to form K2CO3and Na_2CO_3 and degrades the performance of the alkaline electrolyte.

Recently, alkaline membrane fuel cells (AMFCs) gained attention from the fuel cell society due to their compatibility with CO_2. [25], CO_2 introduced in the cathode has a positive effect on improving fuel cell performance. Compared to fuel cells based on acidic polymer electrolytes, low-cost non-precious catalysts, including MnO_2, silver or nickel, may be used for AMFCs [26]. Moreover, Pt/C, PtRu/C and Ru/C were recently investigated AMFCs and can also be used as anodes [27].

Other types of ammonia fuel cells, namely SOFC, are initially developed to prevent NO_X formation [28]. However, the number of scientists who studied SOFC becomes more intense caused by the potencies of the cell to operate at high temperature, thus overcoming the disadvantage suffered by PEMs. At high temperatures, ammonia can be directly decomposed into hydrogen, normally ranging between 500 and 1000 °C, and hence the need for an external cracking reactor is negated. Nonetheless, because of the fragility of porcelain materials, SOFCs are usually not appropriate for transport use [29].

Research on SOFC fuel cells can be separated into Oxygen Ion-Conducting Electrolytes (SOFC-O) and Hydrogen Ion-Conducting Electrolyte (SOFC-H), which is also known as proton-conducting electrolytes. The schematic of SOFC-O and SOFC-H fuel cells is illustrated in **Figure 4**.

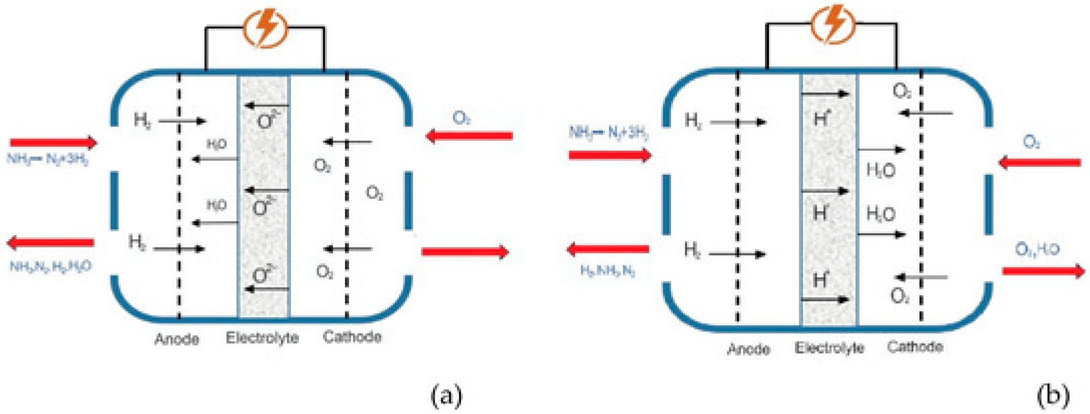

Figure 4. Schematic representation of (**a**) SOFC-O and (**b**) SOFC-H [30].

The SOFC-O operating principle lies in the transportation of oxygen anions across the electrolyte while the charge carrier in SOFC-H is a proton [31]. For both types, ammonia is fed into the anodic site, where it thermally decomposes into nitrogen and hydrogen [32]. In SOFC-O, the oxygen in the cathode compartment is reduced into oxygen ions at the cathode–electrolyte interface and transported across the solid electrolyte, which then reacts with hydrogen electrochemically to produce water [30][33]. The reactions that occur at the anode and cathode are stated below:(9) Anode: $H_2 + O_2- \rightarrow H_2O + 2e-$ (10) Cathode:$12O_2 + 2e- \rightarrow O_2-$

The SOFC-O electrolytes tend to be built based on solid ceramics with metal oxide. YSZ is most widely used because of its high ionic conversion, which facilitates the efficient movement of oxygen anions through the electrolyte [34]. These solid electrolytes also show strong chemical and thermal stability, which is crucial for the treatment of high temperatures. Samarium doped ceria (SDC)-based electrolytes also sparked interest due to their capability to have high ionic conductivities at lower temperatures [35].

In SOFC-H, the hydrogen in the anode compartment is oxidised into a proton which is then transported across solid electrolyte into the cathode [36]. This later reacts with oxygen to produce water. The reactions that occur at the anode and cathode are given below:(11) Anode:$H_2 \rightarrow 2H+ + 2e-$ (12) Cathode:$O_2 + 2H+ +2e- \rightarrow H_2O$

A SOFC-H system electrolyte is selected based on the conductivity of a proton as well as chemical and mechanical stability. The extraordinarily high proton conductivities of doped $BaCeO_3$(BCO) and $BaZrO_3$(BZO) have been shown over a wide 300 to 1000 °C temperature range [37].

In addition to the qualities of the employed material, electrolyte thickness has a direct effect on fuel cell performance. When a thinner electrolyte has been used, the internal resistance of SOFC decreases. However, reducing electrolyte thickness can affect the mechanical strength and consequently stability over the long term [37]. **Table 1** summarises the preceding SOFC fuel cell work that turns ammonia into electricity.

Table 1. Summarization of previous research on SOFC for ammonia synthesis.

Groups	Electrolyte	Thickness (μm)	Cathode	Anode	T(°C)	Power Density (mW/cm^2)	Ref
SOFC-O	YSZ	200	Ag	Pt-YSZ	800–1000	50–125	[38]
	YSZ	400	Ag planar	Ni-YSZ	800	75	[39]
	YSZ	1000	Ag tubular	Ni-YSZ	800	10	[39]
	YSZ	400	Ag	NiO-YSZ	800	60	[35]
	YSZ	150	LSM	NiO-YSZ	700	55	[40]
	YSZ	30	YSZ -LSM	Ni-YSZ	750–850	299	[40]
	YSZ	15	YSZ -LSM	Ni-YSZ	800	526	[41]
	SDC	24	SSC-SDC	NiO-SDC	650	467	[42]
	SDC	10	BSCF	Ni-SDC	700	1190	[43]
SOFC-H	BCGP	1300	Pt	Pt	700	35	[44]
	BCG	1300	Pt	Pt	700	25	[45]
	BCE	1000	Pt	Pt	700	32	[46]
	BCGP	1000	Pt	Ni-BCE	600	23	[47]
	BCG	50	LSC	Ni-BCG	700	355	[48]
	BZCY	35	BSCF	Ni- BZCY	450–700	135–420	[49]
	BCG	30	BSCF	Ni-CGO	600	147	[50]
	BCN	20	LSC	NiO-BCN	700	315	[51]

Note: YSZ denotes yttria-stabilized zirconia, SDC denotes samarium doped ceria, LSM denotes $La_{0.5}Sr_{0.5}MnO_3$, SSC denotes $Sm_{0.5}Sr_{0.5}Co_{3-\delta}$, BSCF denotes $Ba_{0.5}Sr_{0.5}Co_{0.8}Fe_{0.2}O_{3-\delta}$, BCGP denotes $BaCe_{0.8}Gd_{0.19}Pr_{0.01}O_{3-\delta}$, BCG denotes $BaCe_{0.8}Gd_{0.2}O_{3-\delta}$, BCE denotes $BaCe_{0.85}Eu_{0.15}O_3$, LSC denotes $La_{0.5}Sr_{0.5}CoO_{3-\delta}$, BZCY denotes $BaZr_{0.1}Ce_{0.7}Y_{0.2}O_{3-\delta}$, BSCF denotes $Ba_{0.5}Sr_{0.5}Co_{0.8}Fe_{0.2}O_{3-\delta}$, CGO denotes $Ce_{0.8}Gd_{0.2}O_{1.9}$, BCN denotes $BaCe_{0.9}Nd_{0.1}O_{3-\delta}$.

In addition to all of the above forms, microbial fuel cells (MFC) are also seen as an alternate technique for generating electricity directly from ammonia. MFC uses microorganisms in the oxidation process for the conversion of chemical energy from bio-degradable material, for example, ammonia contaminated wastewater. The electrons flow from the anodic side of the external circuit to the cathode, where they combine with the proton and oxygen to form water [37]. The schematic diagram of MFC is shown in Figure **5**.

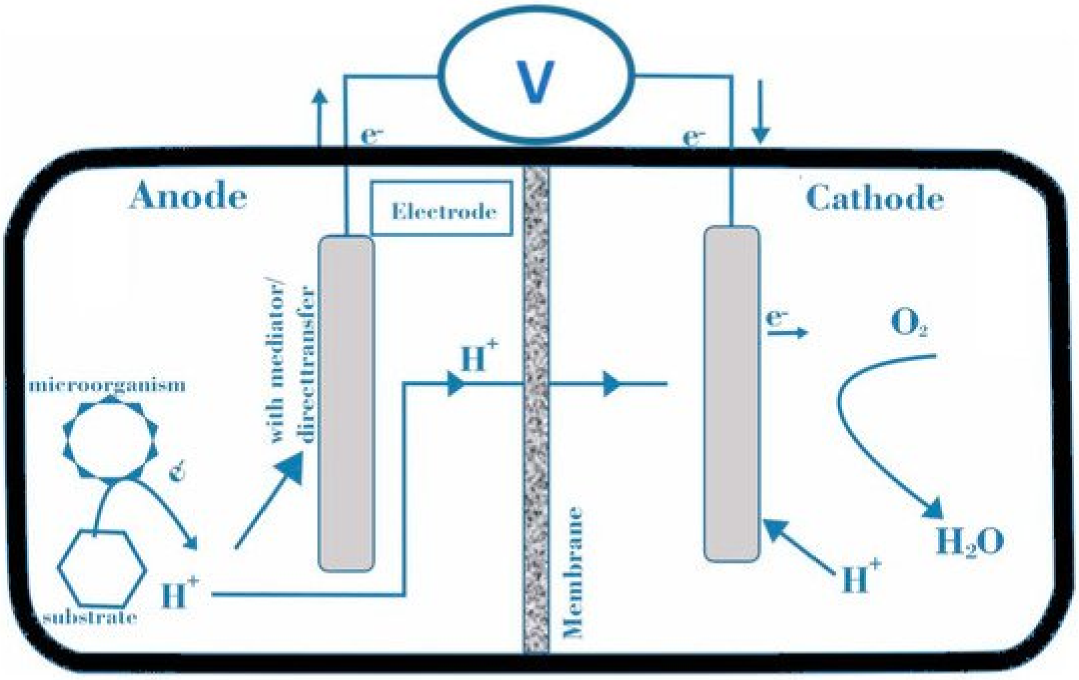

Figure 5. Schematic diagram of MFC [52].

According to Li et al. [53], MFC has been deemed a potential technique for treating wastewater while producing energy, but low power, high cost and reactor scalability issues severely limit its advancement. In addition to wastewater treatment, MFC technology has been proposed as a feasible alternative for air cleaning by removing ammonia from the environment. Yan and Liu [54], in 2020, found Sn-doped V_2O_5 nanoparticles to be a good catalyst for the rapid removal of ammonia in the air using photo-electrocatalysis (PEC) MFCs.

One of the main industries contributing to GHG emissions worldwide is the transport industry. Ammonia-fuelled vehicles, which have received a lot of publicity recently, are one of the solutions to reduce GHG emissions and fossil fuel dependency. Ammonia can be used as fuel in both spark ignition (SI) and compression ignition (CI). However, ammonia poses undesirable combustion properties, which require further study of its combustion properties.

Ammonia has low flame velocity, very high auto-ignition temperatures, narrow flammability limits and high vaporisation heat compared to other fuels [55]. In addition, high latent heat of vaporisation at the time of injection decreases the gas temperature in the engine, which further complicates it [56]. In the SI engine, the use of ammonia is restricted by low flame velocity and narrow explosion limits resulting in incomplete combustion [57]. Ammonia combustion in the SI-engine can be improved by providing stronger igniters such as plasma jet igniters, smaller combustion chambers to overcome the resistance of NH_3 combustion [58].

In addition to the problems mentioned above, ammonia also shows low flame speed and specific energy in combination with high ignition energies and high auto-ignition temperatures, resulting in a relatively low propagation rate from the combustion [7]. Although ammonia has been successfully used as mono-fuel both in SI and CI engines, such a low ammonia combustion rate induces inconsistency in combustion under conditions of low engine load or high engine speed [59]. Thereby, it is essential to mix with secondary fuel to overcome its disadvantages as a fuel. For CI engines, fuels with higher cetane numbers are preferred as a combustion promoter due to the better ignition characteristics [58].

On a dual fuel CI engine, ammonia could be used up to 95% of the fuel energy basis with diesel as a combustion promoter. An optimal mixture, however, is 60% of ammonia on an energy basis because a smaller amount of ammonia would limit the flammability of ammonia [60]. [61] revealed that the fuel performed similar engine performance characteristics with ammonia/diesel blends. Moreover, the study also revealed that the fuel mix of ammonia and DME has a competitive energy cost with current diesel fuel.

Gaseous fuels are chosen for SI-engines due to the same phase with ammonia gas, while anhydrous ammonia would lower the temperature of the in-cylinder, thus adversely affecting subsequent turbulence triggering deteriorated combustion and misfire [62]. In the SI engine powered by ammonia/gasoline, gasoline is port-injected while ammonia gas is direct-injected. Direct injection of ammonia gas substantially lowers the cylinder temperature due to the high latent heat of ammonia. However, too small swirls do not affect the combustion, while too large swirls have a negative effect on the combustion by blowing out the flames due to the slow propagation of the ammonia flames [58].

Another alternative is by using hydrogen as a secondary fuel by installing an on-board reformer to split ammonia into hydrogen and nitrogen [63]. [64] give a complete database of SI engine performance with ammonia/hydrogen mixture as a fuel. A series of studies were undertaken with different excess air ratios and hydrogen ammonia ratios. The results revealed that a fuel mixture with 10 vol.% hydrogens has the best performance in terms of efficiency and power.

The study also shows that when compared with pure ammonia injection, the use of hydrogen from cracked ammonia is extremely beneficial. In addition to road transportations, ammonia is also a favourable fuel for marine industries [65][66]. Unlike the automotive industry, marine systems are not space-constrained, so that catalytic equipment can be deployed for NOx reduction solutions. In the recent development, MAN Energy Solutions replaces the 3000 B&W double-fuel engines operating in the field of LPG and diesel engines [67].

However, due to nitrogen molecules in its structure, ammonia combustion is always associated with the formation of nitrogen oxides that exceed current standards [68][69]. Moreover, a longer residence time in the combustion chamber is required for ammonia to be completely combustible due to low laminar burning velocity, which makes it difficult to achieve stable combustion [7]. However, the shift towards carbon-free alternative energy carriers has returned interest in ammonia, including the utilisation in power industries. Burning NH_3 in turbines is the most promising direction of using ammonia as a carrier of surplus electricity generated from renewable energy to balance seasonal energy demand.

The study emphasised that the final product of ammonia combustion is not nitric oxides because the overall reaction is $4NH_3 + 3O_2 \rightarrow 2N_2 + 6H_2O$. The study revealed that ammonia combustion could be self-sustained when 10% cracking is applied. The system was able to achieve over 25-kWe power generation by supplying about 10% heat from ammonia gas. [70], in their experiment and numerical calculations, concluded that predominant rate-limiting reactions in methane–ammonia flames are belonging to the ammonia oxidation path, which controls H and OH radicals.

The study also revealed that the NO concentration decreases when ammonia increases under rich conditions.

[71] developed a gas-turbine combustion system with controlled emissions, which uses a mixture of NH_3 and natural gas as the fuel. Detailed compositions of the burner exhaust gas were measured under atmospheric pressure and lean fuel circumstances. The study also shows that with an increase of equivalency ratio, unburned species such as NH3, CO and THC decrease while NO and N_2O emissions increase. The study concludes that low emissions and good combustion efficiency are difficult to obtain in a single-stage reactor.

The concept of low-emission combustion in two-stage combustion was examined numerically and experimentally. The secondary zone is then only supplied ammonia. The results indicated two methods for attaining low NOx combustion: rich-lean combustion and a combination of lean combustion and extra ammonia delivery. As a result, primary zone NOx production dominates emission.

Their results indicated that ammonia is suitable for use in large turbines. Before ammonia is fed to the combustor, the gas turbine power is raised up to 2 MWe using natural gas. The result shows that the gas turbine engine’s operation was shown to be steady across a wide variety of ammonia mixing ratios ranging from 0% to 20%. As the ratio of ammonia in the mixture increases, the concentration of NOx at the turbine outlet rises significantly up to 5% mixing ratio, then remains steady until it reaches 20%.

4. Conclusion

The application of ammonia as a renewable energy carrier not only plays a key role to lower GHG emissions but also allows transporting H_2 efficiently and economically, permits the direct conversion to electricity by the fuel cell and provides versatility in its use, as fuel for the IC engine and power generation.

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