A Roadmap to the Ammonia Economy

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Summary: Ammonia is increasingly recognised as an important, sustainable fuel for global use in the future. Applications of ammonia in heavy transport, power generation and distributed energy storage are being actively developed. Produced at scale ammonia could replace a substantial fraction of current-day liquid fuel consumption. This ammonia-based economy will emerge through multiple generations of technology development and scale-up. The pathways forward in regard to current-day technology (Generation 1) and immediate future approaches (Generation 2) that rely on Haber-Bosch process are discussed. Generation 3 technology breaks this nexus with the Haber-Bosch process and enables direct reduction of dinitrogen to ammonia electrochemically. However, the roadmap towards scale in this technology has become obscured by recent research missteps. Nevertheless, alternative Generation 3 approaches are becoming viable. We conclude with perspectives on the broader scale sustainability of an ammonia economy and the need for further understanding of the planetary nitrogen cycles of which ammonia is an important part.
1. Introduction

Developments in renewable energy technologies over the last decade have created a new awareness that the critical lack of transportable, or “dispatchable”, forms of energy has become the main bottleneck in a major and global shift towards renewables. Battery storage addresses some aspects of the challenge, but is unsuitable for large scale transportation of energy to market from areas of the world where wind and solar energy are plentiful. For bulk transport by sea, or pipeline, a liquefied form of energy storage is almost certainly the preferred option, and this throws the spotlight onto processes that can generate high energy density liquid fuels from renewables, in a cost-effective and sustainable manner.¹ A breakthrough in this regard would open up a range of very attractive renewable energy futures.

A number of options for such liquid energy carriers have emerged, including liquid hydrogen (H₂), liquid organic hydrogen carriers, the various possible products of the carbon dioxide (CO₂) reduction and ammonia (NH₃). Each of these has its advantages and disadvantages, for example the large energy cost of liquefying hydrogen, or the need for a concentrated source of CO₂ in technologies based on carbon dioxide reduction, and each needs to be considered in terms of its relative utility and safety. Out of this mix, ammonia has emerged as a strong and increasingly compelling candidate as the renewable energy sourced fuel of the future.²,³

Ammonia is readily liquefied by increasing pressure to ~10 bar at room temperature, or by cooling to -33 °C at atmospheric pressure. Unlike liquid hydrogen, the technology of shipping and pipeline transfer of ammonia is well established in the existing industry. Approximately 175 million tonnes are produced annually worldwide,⁴ for a market of value around USD 70 billion;⁵ by comparison, the global Liquefied Petroleum Gas (LPG) market is around 300 million tonnes annually.⁶ Most of this ammonia is used in the production of fertilisers, with small amounts going into explosives and chemicals/materials.

Just over a century ago the discoveries by Haber and Bosch made possible the industrial production of ammonia and ammonia-based fertilisers that today feed the world and is the source of most of our nitrogen containing chemicals, materials and pharmaceuticals. The process generates H₂ from natural gas or coal through steam reforming and combines it with N₂, which has been separated from air by a cryogenic process, to form NH₃. The
reaction between N₂ and H₂ requires temperatures in excess of 400 °C and pressures above 200 bar to be facile, and therefore the capital cost of plant and equipment is substantial. Ammonia production is currently responsible for ~1.0% of global greenhouse gas emissions\(^7\) (or about 1.4% of global CO₂ emissions); these values increase further if CO₂ emissions associated with natural gas extraction are included.

Over the last decade, momentum has been building to transform the Haber-Bosch (H-B) ammonia industry towards renewable sources of hydrogen, for example from water electrolysis or solar thermal cycles.\(^8\) This goal has provided the background to the broader vision of ammonia becoming a transportable store of renewable energy that we will discuss in this paper. The challenge both for the existing industry, as well as a much-expanded renewable energy industry is, of course, economics.

As the focus on ammonia as a liquid energy carrier has developed in recent years, so also has investigation of an increasingly broad range of applications. Ammonia was seen originally in many of these uses as a carrier and supplier of hydrogen energy, the supply chain in that context involving a step that cracks NH₃ into H₂ and N₂ at a point of delivery, for distribution and use as a hydrogen gas.\(^9\) In the last few years, the vision of ammonia energy applications has been widening significantly to now include its direct use as a fuel. As discussed later in this article, examples include as a marine bunker fuel, as a fuel for heavy transport vehicles, including buses (as implemented in Belgium in the 1940s), or small, medium and large-scale power generators, direct ammonia fuel cells, power turbines and even jet engines. Thus, the potential emerges for ammonia to become a replacement for fossil fuels in almost any application. Upon “combustion” in any of these devices, the only exhaust should be pure N₂ and water, that can be safely released to the environment directly at the point of use. Overall, this forms a completely circular and sustainable cycle that we visualise as the Ammonia Economy\(^10\) in Figure 1.

In this roadmap article, we begin by assessing the current and emerging production technologies for ammonia from renewable energy sources, including a more in-depth discussion of the issues confronting the field of research focussed on the direct electrochemical nitrogen reduction reaction (eNRR). We then discuss some of the surrounding challenges that must be addressed in massively scaling up these technologies towards their use as a common fuel. Following this, we survey the broadening range of end-use modalities that are emerging for ammonia as an energy carrier. Recognising the need
to fully understand the potential environmental impacts of an extraordinarily increased use of ammonia, we also briefly comment on the state of understanding of the planetary nitrogen-cycle that this anthropogenic activity must sit sustainably within.

Limited space in an article such as this prohibits an extensive review of the wide range of topics covered here. Instead our approach has been to identify key, recent and authoritative references for the interested reader to delve further into.

**Figure 1.** Vision of the “Ammonia Economy” in which the energy sources and uses are all based on ammonia.

### 2. Current and Emerging Technologies for the Production of Sustainable Ammonia

As mentioned above, the H-B process coupled to steam hydrocarbon reforming is the main industrial method for the production of ammonia. Although it has been optimised over time, its globally-substantial greenhouse gas emissions have prompted the industry to pursue sustainable production technologies. Once developed, these will become the foundation of the Ammonia Economy. This will occur in a series of overlapping technology “Generations”, as follows:
**Generation 1** (Gen 1) involves the use of carbon sequestration or offsets to bring the net carbon impact of the ammonia production to zero. This is occasionally referred to as “blue ammonia”, following the colour scheme often used in hydrogen-energy discussion. Clearly, the carbon sequestration aspect of blue ammonia production adds cost and plant complexity on top of the existing H-B technology. For this reason, it is likely to represent only a transitional solution, helping to establish a market for ammonia beyond the fertiliser and chemical industries. Modern H-B plants produce ammonia at an energy cost of at least 8 MWh tonne\(^{-1}\).\textsuperscript{11} Recognising that the Lower Heating Value (LHV) of ammonia is 5.2 MWh tonne\(^{-1}\), this represents an energy efficiency of only 65%. Additional energy costs attributable to the carbon sequestration process can only further decrease this value; detailed modelling of these costs in existing or new plant would be a useful contribution to the broader understanding of Gen 1.

**Generation 2** (Gen 2) renewable ammonia we describe as that produced from H-B technology, but employing renewable, rather than fossil fuel sourced, hydrogen. This has the advantage that existing H-B plants can be transitioned to this new hydrogen supply without major disruption or mothballing. The technology has already been demonstrated at a small, practical scale by Wilkinson and colleagues at Siemens.\textsuperscript{12} Powered by fully-renewable electricity derived from a 20 kW wind turbine, the Siemens demonstrator produces \(\text{H}_2\), using a proton exchange membrane (PEM) water electrolyser, to form around 30 kg \(\text{NH}_3\) daily.\textsuperscript{2}

Gen 2 technology has significant long-term scope in terms of the Ammonia Economy, limited only by the substantial investment and long lead-time required to establish new facilities. On top of the cost of a conventional H-B plant, the capital cost also includes the water electrolysis equipment, which is currently of the order of USD\(1\)M per MW of capacity.\textsuperscript{13} As discussed in more detail in section 4.4 below, the impact of this capital cost indicates that it could add significantly to the capital intensity of ammonia production. This throws the spotlight onto research and development efforts to lower the cost of electrolysers. The most established electrochemical water splitting technology – alkaline electrolysis – has limited potential for major cost reductions. The key remaining challenge is improved engineering of cells and stacks to increase the areal current density and decrease the device footprint. In contrast, the PEM technology, which is more favoured by the industry\textsuperscript{14} mainly due to the inherently higher current densities achievable (> 1 A cm\(^{-2}\)), presents several options for
significant cost improvements. This could be achieved via the development of cheaper bipolar flow field plates, membranes, and catalysts, as well as by increasing the cell area. Double-digit megawatt scale PEM water electrolyzers with a higher-heating value efficiency of 75% are already commercially available, and ongoing active research and development by academia and industry will likely bring this technology to the forefront of sustainable hydrogen generation for NH₃ production.

An interesting alternative for the Gen 2 NH₃ production is high temperature solid oxide electrolysis (SOE), which can utilise waste heat from the H-B plant. This process, with predicted overall energy efficiency for ammonia production above 70%, is currently being developed by Haldor Topsøe and partners. Moreover, apart from generating H₂ with close to 100% efficiency, SOEs can be used for the generation of pure N₂ from air at the cathode to replace conventional air separation units, which otherwise contribute notably to the final energy cost of NH₃. The challenge is the immaturity of the SOE technology, which is still at the pre-commercial stage. Among other problems, stability of key electrolyser components during long-term operation at 700-900 °C is yet to be proven. Furthermore, efficient operation of high temperature electrolyzers in an intermittent, renewables-powered mode presents a challenging engineering problem and is likely to incur further significant costs; this contrasts with the PEM technology, which provides rapid start-up and shut-down and is highly compatible with an interrupted operating regime.

In fact, the need to smooth the intermittency of most renewable energy sources (with the significant exclusion of hydro and geothermal sources, which can be continuous) is crucially important aspect of the Gen 2 approach. The Haber-Bosch process ideally runs continuously, a feature that interfaces poorly with the day-night cycle of solar, or the intermittency of wind, creating the need for an intermediate energy storage solution. As discussed in more detail in section 4.4, battery storage is currently too expensive to fit this need and hydrogen storage, either as liquid, compressed gas or in the form of a storage material, appears to be the optimal solution.

**Generation 3** (Gen 3) technology refers to the electroreduction of N₂ to ammonia by direct or mediated means. The H-B process is no longer required at this level of technology. Instead, the reaction is driven by electrochemical reduction and the H-source is ultimately water. There are several modes of this process being actively researched: (i) the
electrochemical nitrogen reduction reaction (eNRR) in which an electrocatalyst enables direct electron and proton addition to the N$_2$ molecule (Figure 2a), and (ii) indirect or mediated mechanisms in which a redox mediator such as Li$^+$ is first reduced and then, via a series of reactions, ammonia is produced and the mediator is regenerated (Figure 2b). The physical setup of the eNRR cells bears much in common with water splitting technology and so will use similar design and construction principles.

For Gen 3 ammonia synthesis, practical performance targets have been defined in the U.S. Department of Energy’s REFUEL program$^{19}$ at a current density of 300 mA cm$^{-2}$ with 90 % NH$_3$ current efficiency (also known as faradaic efficiency) and 60 % energy efficiency.

Notable features of Gen 3 approaches include:

(i) greater resilience to intermittency, i.e. the ambient-temperature electrochemical process can in principle cycle down to zero current, without damage, as dictated by the renewable energy source (though particular catalyst/mediator families may not be suitable for this).

(ii) potentially lower sensitivity to N$_2$ supply purity, in contrast to the H-B catalysts which are damaged by only a few ppm of O$_2$ or H$_2$O in the N$_2$ feed supply.$^{18}$ In Gen 3 technology, an O$_2$ contaminant simply lowers the selectivity (viz. current efficiency) of the process, but is not expected to be damaging to most catalysts (though it may be in some mediated approaches). Accordingly, the N$_2$ purity requirement is not as extreme in Gen 3 as it is in the H-B processes.

(iii) The eNRR process in principle has the potential to operate at energy efficiencies greater than Gen 2 processes. However, at the fundamental electrochemistry level it is notable that practical direct and mediated eNRR processes typically require substantially more overpotential (viz. excess energy over and above thermodynamic minimum energy). Theoretical analysis indicates that an overpotential not less than 0.45 V, corresponding to the cathode potential of ~ -0.4 V vs. the reversible hydrogen electrode (RHE),$^{20}$ is required to electroreduce N$_2$ to NH$_3$; coupling this with an oxygen producing anode operating at +1.5 V vs RHE, and allowing for other losses in the cell, indicates an overall cell potential of at least 2.0 V, which equates to about 10.5 MWh tonne$^{-1}$NH$_3$ (note that this does not include air separation and ammonia separation energy costs and assumes 90 % NH$_3$ faradaic efficiency in accordance with the U.S. Department of Energy (DoE)
targets mentioned above\(^\text{19}\)). This, in principle, is competitive with both Gen 1 and Gen 2 approaches.

![Diagram of N\(_2\) adsorption, NH\(_3\) release, and N\(_2\) reduction process.](image)

**Figure 2.** Possible mechanisms of (a) direct eNRR via absorption of N\(_2\) onto the catalyst surface, followed by progressive proton and electron additions to produce a first, followed by a second molecule of ammonia (adapted from reference\(^\text{21}\)); (b) indirect electrochemical N\(_2\) reduction to ammonia based on lithium as a mediator, forming Li\(_3\)N as an intermediate on a copper substrate (atom and ion sizes approximately to scale; differences in Li and N sizes reflect their differing states during the process, A = anion).

**Other Technologies.** Direct photoreduction of N\(_2\) to NH\(_3\) has been an area of active study for some time, however the reported yield rates of ammonia in photochemical processes are currently too low to be practical in a scaled up device and in some cases may yet be the result of contamination or reduction of oxidised forms of nitrogen rather than N\(_2\).\(^\text{22}\)
Nevertheless, this direction presents an important long term strategy for future nitrogen fixation technologies. Though direct light-driven processes are disadvantageous in the context of the large scale chemical synthesis, an ammonia producing photocatalytic process is likely to find applications in intensive horticulture.\textsuperscript{23}

Another alternative pathway towards the sustainable ammonia synthesis utilises nitrogenase organisms and biomimetic catalysts to create a biotechnological route to NH$_3$ that may grow to have some significance in the agricultural industries.\textsuperscript{24} One noteworthy concept developed by Nocera and co-workers uses a hybrid biological-inorganic system in which ammonia is produced from N$_2$ and H$_2$ (derived from electrochemical water splitting) involving cellular biomass derived from biochemical CO$_2$ reduction. Operation of this system directly in the soil and promotion of the growth of radish plants was demonstrated.\textsuperscript{25}

\section*{3. Pathways to Generation 3 Ammonia: Challenges and Dead-Ends}

Unfortunately, detailed assessment of the current state of eNRR research is fraught with difficulty because of a number of issues in the experimental methods and results presentation. Notwithstanding the several detailed protocols for proof of genuine eNRR that have been published,\textsuperscript{26-28} the overwhelming majority of recent reports do not meet the mandatory requirements to qualify as reliable results. Figure 3 summarises the status of the various Gen 3 approaches and, as discussed in detail in this section, the challenges that each faces in approaching practical levels of performance.

Particularly problematic is the intense research focusing on eNRR in aqueous electrolytes, which, although appealing from a technological perspective, is fundamentally unfavourable for the fixation and reduction of the highly unreactive N$_2$ molecule. Low faradaic efficiency (selectivity) for the eNRR is reported in most studies,\textsuperscript{27,28} due to the dominance of the electrochemical hydrogen evolution reaction (HER) at the potentials involved. Areal NH$_3$ yield rates are also too low to be practical, typically below 0.1 nmol s$^{-1}$ cm$^{-2}$,\textsuperscript{27,28} i.e. orders of magnitude lower than the 100 nmol s$^{-1}$ cm$^{-2}$ identified by Giddey and co-workers\textsuperscript{29} as a minimum for practical eNRR. At the reported low rates of ammonia formation, incorporation of N-based contaminants into the experimental setup becomes a serious concern. As a consequence, the reliability of most (if not all) of aqueous eNRR results remains dubious, as emphasised recently by us\textsuperscript{1,28} and others.\textsuperscript{27} While contamination by environmental NH$_3$
can easily be avoided, the ubiquitous oxidised forms of nitrogen \((\text{NO}_x)\) are often ignored, despite offering a facile route to ammonia under typical conditions.\(^{30,31}\) As a result, even reports which include quantitative data on \(^{15}\text{NH}_3\) produced with \(^{15}\text{N}_2\) feedstock gas cannot be considered entirely robust if the studies do not rigorously demonstrate complete removal of \(^{15}\text{NO}_x\) contaminants, for example by following the recommendations of Chorkendorff and co-workers.\(^{27}\) Some of the more significant reports are now being refuted.\(^{32}\) Unfortunately, there are also numerous reviews appearing that do nothing to critically assess the situation. Overall, the grand challenge of this approach is still at the stage of requiring clear proof-of-concept, \(i.e.\) unambiguous demonstration of genuine, high-selectivity electrocatalytic reduction of \(\text{N}_2\) to \(\text{NH}_3\) in the presence of aqueous-based electrolytes.

By comparison, the non-aqueous routes, which avoid the HER prevalent in aqueous systems by limiting the availability of \(\text{H}_2\text{O}\) or other \(\text{H}\)-sources, can provide much higher selectivity. However, the potentials at which non-aqueous reactions usually operate typically increase the energy cost considerably. Realisation of a non-aqueous approach is possible at high temperature with molten hydroxide electrolytes,\(^{33,34}\) or at ambient temperatures in aprotic electrolytes,\(^{35-38}\) and each route presents its own specific challenges. While direct electroreduction in molten hydroxides provides the highest \(\text{NH}_3\) yield rate, of the order \(\text{nmol s}^{-1} \text{ cm}^{-2}\), at the lowest energy input (lowest eNRR overpotential), the faradaic efficiency of this process is low (<35%).\(^{33}\) Electrocatalytic reaction under ambient conditions in aprotic electrolytes can provide significantly higher faradaic efficiency; the challenge in this case is the very low rate of the process, of the order of only \(0.01 \text{ nmol s}^{-1} \text{ cm}^{-2}\).\(^{35}\)

The continuous redox mediated pathway, first demonstrated using a \(\text{Li}^{+}/0\) mediator more than 20 years ago, provides rates approaching \(1 \text{ nmol s}^{-1} \text{ cm}^{-2}\),\(^{27,37-39}\) and reasonable selectivity is possible at elevated pressures (50 bar).\(^{38}\) An advantage of this approach is that a separator membrane may not be needed in the cell as cycling of the mediator between the electrodes is part of the process. Like the other eNRR approaches discussed above the electrolysis can be carried out continuously as long as reactants and current are supplied and products removed. A limitation of the \(\text{Li}^{+}/0\) mediator is the requirement for a significant energy input, as defined by the standard potential of this redox couple (approximately -3 V vs. normal hydrogen electrode), which formally corresponds to more than 3 V overpotential for the eNRR.
A variation on the Li-mediated approach was demonstrated by Nørskov and co-workers using a multi-step process including a high-temperature molten LiOH electrolysis step to recycle the lithium mediator. This process requires at least 15.5 MWh tonne\(^{-1}\) \(\text{NH}_3\), assuming 90\% \(\text{NH}_3\) faradaic efficiency accordance with the U.S. DoE targets, indicating an energy efficiency of \(\sim 34\%\). Thus, even though this Li-mediated approach is the most promising at the current level of the technological progress in Gen 3 (demonstrated \(\geq 88\%\) faradaic efficiency at 0.2 A), it is thermodynamically incapable of meeting the energy efficiency target even at high temperatures, due to the very negative reversible potential of the \(\text{Li}^{+}/\text{Li}^0\) process. Thus, the DoE target remains challenging and new approaches to improving the energy efficiency of the mediated approach need to be explored, perhaps involving mediators operating at less negative potentials.

In summary, introduction of less energy demanding redox mediators is seen as the most urgent direction for the improvement of the mediated eNRR approach. Finding the ideal combination of mediator, catalyst and electrolyte components to optimise selectivity and yield rate, while decreasing energy costs, is the key goal of research in this field and ultimately may provide the way forward to practical Gen 3 technology.
Figure 3. Overview of current status of Gen 3 approaches to nitrogen reduction and their development challenges. Green/orange/red lights indicate proximity of the performance metrics to practical levels, from high to low. The question marks against the aqueous approach indicate the uncertainty surrounding reported results. “Energy” relates to overall energy efficiency of the approach. “Rate” ultimately determines capital cost/intensity of the plant and “Selectivity” of the approach impacts on both energy and capital cost.

4. Development to Practical Scale - Other Challenges

4.1 N₂ utilisation and ammonia separation

One of the key practical issues in scaling up any eNRR process is separating the product from the electrolyte or the gas stream exiting the cell. In many Gen 3 reports to date there is a very high flow of N₂ gas (>20 mL min⁻¹) and the NH₃ concentration exiting the cell in the gas stream is dilute, rarely more than 100 ppm. In a practical cell, N₂ will have been separated from air using standard cryogenic or membrane processes and for cost reasons will need to be recycled into the cell. A common laboratory procedure for separating NH₃ from N₂ involves an aqueous acid trap⁴⁰; acid trapping may be viable for some fertiliser applications (e.g. production of (NH₄)₂SO₄ by using a H₂SO₄ acid trap), but adds energy consuming steps if the ultimate product is anhydrous NH₃ as a fuel. Cryogenic separation is routinely used in the H-B process, condensing the NH₃ into its liquid form at sub-ambient
temperatures. In the eNRR context, low concentrations of NH\textsubscript{3} in the gas streams will add further energy cost to the overall process. Achieving much higher product concentrations in flowing gas processes is therefore an important goal in this particular pathway. In the case of the Li-mediated multistep process, since NH\textsubscript{3} is produced by the reaction of Li\textsubscript{3}N with H\textsubscript{2}O, pure NH\textsubscript{3} is readily extracted by the use of a stoichiometric amount of water to the pre-formed Li\textsubscript{3}N.

4.2 Water requirements

A minimum of 1.5 tonnes water is needed stoichiometrically per tonne of ammonia. Process losses and emissions will typically increase this amount by at least 20%. In arid regions such as Australia, the Middle East and Chile, where solar insolation is optimum, the supply of this water component is a significant issue. Sea water is an option, desalinated by solar distillation or reverse osmosis, but at additional energy cost of around 3-5 kWh tonne\textsuperscript{-1}H\textsubscript{2}O, which would only add < 1% to the energy cost of the ammonia. Sea water may also be used directly as discussed in the next section.

4.3 Anode reactions

Any eNRR process needs to be accompanied by an appropriate anode reaction to supply protons and electrons. The only ultimately sustainable anode reaction product is O\textsubscript{2} from water. In Gen 2 processes this is already the case at the water splitting stage. In aqueous Gen 3 process this is also often the case, though it is important that the oxygen gas produced be isolated from the cathode, as its reduction would ultimately become a loss of efficiency.

The water oxidation reaction (WOR; or the oxygen evolution reaction) is an intensely studied process at both fundamental and practical levels, in the first place in the context of hydrogen generation from water,\textsuperscript{41} although it can be coupled to other than hydrogen evolution reaction cathodic processes including the eNRR. Efficiency and durability of the WOR electrocatalysts are significantly affected by the operating pH, with the most robust, high-performance and comparatively cheap systems being only available for the classic strongly alkaline electrolytes.\textsuperscript{42,43} If required for the eNRR technology, catalysts operating at near neutral pH\textsuperscript{42,44} as well as in strongly acidic medium are available, though stability of the latter remains a challenge.\textsuperscript{45,46} WOR catalysts operating at low pH are receiving increasing investigative attention currently\textsuperscript{47,48} driven by the strong industrial momentum towards the development of the PEM electrolysis technologies.\textsuperscript{15}
Sea water, and more broadly brine based, electrolysis is an option in either of two modes (i) by use of an anode catalyst that selectively promotes the oxidation of chloride to chlorine (the chlorine evolution reaction, CER) as the main anode reaction or (ii) an anode electrocatalyst that promotes the WOR in the presence of a high concentration of Cl\(^{-}\). The former generates Cl\(_2\) (or OCl\(^{-}\)) as a by-product of the overall process. Although chlorine is a valuable industrial chemical, the quantities produced in the renewable fuels industry would far exceed demand for Cl\(_2\). For example, more than a billion tonnes of Cl\(_2\) would result from current day production of NH\(_3\) if carried out via an eNRR-CER electrochemical process; this is almost 20 times the global chlorine market.\(^{49-51}\) Sustainable emissions of this would otherwise need to be well understood in the local and global environmental context.

4.4 Intermittency

The capital costs of the electrolyser plant represent a substantial component of the production cost of ammonia in both Gen 2 and Gen 3 variants. Using conservative water electrolyser costs as guide, at approximately USD1M MW\(^{-1}\),\(^{13}\) in a Gen 2 mode this adds at least ~ USD800 per tonne of annual ammonia capacity (based on 75% H\(_2\) production efficiency and a minimum 5.3 MWh of H\(_2\) energy required for per tonne of ammonia production). By comparison, typical capital intensity of H-B is around USD1000 - 1500 tonne\(_{NH3}\)^{-1} of annual capacity.\(^{52}\) In the Gen 3 mode this electrolyser cost is the main capital cost (to which the air separation and ammonia separation balance of plant equipment need to be added).

Lowered capacity factors due to intermittency impact on these capital costs. There is a potential benefit to be gained from employing some form of energy storage such that the electrolysis can be run continuously. This applies to both Gen 2 and Gen 3 technologies. Battery storage is currently an expensive option, at capital costs around USD300,000 per MWh stored.\(^{53}\) Even though the cost of battery storage is projected to fall to USD100,000 per MWh and below by 2050,\(^{53}\) the associated costs would still be significant. For 1 tonne per day of ammonia produced continuously at 60% energy efficiency, using the DoE target, energy consumption is ~8.8 MWh tonne\(^{-1}\)\(_{NH3}\). If a minimum of 18 h of stored energy use per day is required in a pure solar powered system, ~6.6 MWh of storage is needed. At a future, mid-range projection battery storage price of USD200,000 MWh\(^{-1}\) this equates to USD1.2M tonne\(_{NH3}\)^{-1} of daily capacity (~ USD3300 tonne\(_{NH3}\)^{-1} of annual capacity) indicating that this
form of storage at this cost could represent an approximate doubling of current day annual capital intensity (USD1000 - 1500 tonne\textsuperscript{-1}NH\textsubscript{3}\textsuperscript{1,52})

An alternative option for Gen 2 is the storage of intermittent renewable energy in the form of H\textsubscript{2} produced through electrolysis at the first stage, to allow the Haber-Bosch plant to operate continuously. At large scale ammonia production this would most likely require compression of H\textsubscript{2} to minimise the footprint of the storage. Pressurising H\textsubscript{2} consumes energy in the amount of several % of the LHV of H\textsubscript{2} to bring the pressure to several dozen MPa,\textsuperscript{54} and will require additional capital expenditure for the high-pressure H\textsubscript{2} tanks.\textsuperscript{55}

4.5 Safety and social acceptance

The use of NH\textsubscript{3} as fuel will result in an increased amount of ammonia being handled on a daily basis in public spaces. Therefore, the risks associated with its accidental release and exposure have to be carefully assessed and mitigated. From a safety point-of-view, the use of ammonia as a renewable energy sourced fuel will inevitably attract some degree of social concern. One of the basic reasons for this is its odour, even at a very low concentrations (5 ppm),\textsuperscript{56} creating a perception of it as highly toxic, despite the “Immediately Dangerous to Life or Health”, IDLH value (300 ppm) and Threshold Limit Value (25 ppm) concentrations being much higher.\textsuperscript{57} Notably, unlike gasoline which is known to contain carcinogenic substances such as benzene, ammonia is not considered as carcinogenic.

Several studies have been conducted to qualitatively and quantitatively evaluate the risks of using ammonia as a fuel. In most of these reports, useful comparisons have been made to traditional fossil fuels and the interested reader is referred to these comprehensive studies for a detailed insight. To summarise some of the high level outcomes: in a 2005 report by Risø National Laboratory in Denmark, the risk associated with the accidental release of ammonia used as a vehicle fuel was evaluated to be comparable and in some cases lower than that of LPG.\textsuperscript{58} Similarly, an independent quantitative risk analysis in 2009 by a U.S. based safety consulting firm, Quest Consultants Inc., arrived at a similar conclusion, stating: “In summary, the hazards and risks associated with the truck transport, storage and dispensing of refrigerated anhydrous ammonia are similar to those of gasoline and LPG”,\textsuperscript{59} despite the different quantification methodology employed. Detailed discussion is also provided in the paper by Valera-Medina et al.\textsuperscript{60}
A thesis on a safe and effective application use of ammonia in a marine transport context has also been published very recently by a Dutch naval architecture company, spearheaded by Lead Naval Architect, Mr Niels de Vries. In this comprehensive report, 61 modes of failures in ammonia powered marine transport were studied and potential mitigation measures and technology proposed. The report concluded that once proper technology and mitigation measures are put in place, the likelihood of a catastrophic failure becomes extremely unlikely. In addition, it was noted that the self-alarming nature of ammonia, due to its strong odour, means that its leakage could be detected at a very early stage and be utilised to further lower the resulting impacts.

Clearly social acceptance of ammonia as a large-scale fuel and energy carrier is a crucial matter that will require ongoing investigation, standards and procedures development, and intergovernmental, policy-level effort. Nonetheless, there appears to be no fundamental issue in this respect that would represent a complete roadblock to the Ammonia Economy. In this regard, it is important to note the contribution of the Ammonia Safety Training Institute as a non-profit organisation that is providing leadership in the ongoing development of safe handling procedures.

5. Developments in End-Use Modalities

Although ammonia has been used in modified vehicles (buses and cars) for many years, the development of ammonia as an energy carrier during the 2015-2018 period was very much around its use as a hydrogen carrier in the hydrogen supply chain. In this modality, a Gen 1 or 2 ammonia is produced for shipping and then converted into hydrogen gas at the receiving port, or later in the supply chain in the receiving location. One of the key developments in this concept was the Australian Commonwealth Scientific and Industrial Research Organisation’s ammonia cracking technology which produces a pure H₂ stream from the splitting of ammonia at elevated temperatures (> 400 °C) by using a unique metal-based membrane.

However, arising out of these developments, the realisation that ammonia could become available as an energy carrier, or fuel, in large quantities has stimulated intense R&D of a range of direct end-use modalities, which are subcategorised below. A useful review
detailing a number of power generating applications including various types of engines and fuel cells has been recently published.80

Recognising that, for wide-spread uptake as a fuel, the cost of ammonia cannot ultimately greatly exceed other liquid fuels, it is important to place ammonia in the context of current-day prices. In recent years, the international price of ammonia has ranged between USD250 – 400 tonne\(^{-1}\) (cost and freight)\(^63\) the variability depending chiefly on the local cost, at the plant, of natural gas or coal. For the purposes of discussion in this paper we will take USD300 tonne\(^{-1}\) as an indicative cost (inclusive of transportation costs). Table 1 then compares the fuel properties of ammonia with diesel, LPG, gasoline and marine bunker fuel (a heavy diesel oil) on an approximate USD kWh\(^{-1}\) basis using their lower heating values (LHV). Fuel prices vary considerably around the world depending on a number of factors including local taxes and delivery costs; as illustrative global comparisons we have used recent costs of diesel in U.S.A., of LPG in Germany and gasoline in Japan (reference\(^64\) provides a regularly updated listing of prices for many countries) and a Global Average Price for shipping bunker fuel (a heavy diesel oil). It becomes clear that ammonia can be competitive with current fossil fuel prices to the user, if the end use technologies are at least equivalently energy efficient.

A price on carbon would have material impact on this comparison. The global average emissions from current day ammonia production are approximately 2.9 tonnes CO\(_2\) per tonne NH\(_3\)\(^{-1}\).\(^7\) The current trading price of carbon in Europe is about 25 EUR tonne\(^{-1}\) \(\approx\) USD27.5 tonne CO\(_2\)\(^{-1}\).\(^65\) Hence, current day carbon prices would add \(\sim\) USD80 tonne NH\(_3\)\(^{-1}\) to today’s ammonia prices if implemented globally; the fossil fuel prices in Table 1 would also increase accordingly. Of course, Gen 2 and Gen 3 ammonia would not be impacted by a carbon price.
### Table 1: Relative properties and costs of ammonia compared with liquid fossil fuels (Adapted and updated from Zamfirescu and Dincer\textsuperscript{66}).

<table>
<thead>
<tr>
<th>Fuel</th>
<th>P [bar]</th>
<th>Density [kg m(^{-3})] (15 °C)</th>
<th>LHV [kWh kg(^{-1})] (25 °C)</th>
<th>LHV [MWh m(^{-3})] (25 °C)</th>
<th>Cost [USD kg(^{-1})]</th>
<th>Cost [USD kWh(^{-1})]</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ammonia</td>
<td>10</td>
<td>6.03</td>
<td>5.18</td>
<td>3.12</td>
<td>0.30</td>
<td>0.058</td>
</tr>
<tr>
<td>Diesel</td>
<td>1</td>
<td>1.846</td>
<td>12.1</td>
<td>10.2</td>
<td>1.00 (U.S.A)</td>
<td>0.083</td>
</tr>
<tr>
<td>LPG</td>
<td>14</td>
<td>0.388</td>
<td>12.6</td>
<td>4.89</td>
<td>1.00 (Germany)</td>
<td>0.079</td>
</tr>
<tr>
<td>Gasoline</td>
<td>1</td>
<td>0.736</td>
<td>12.1</td>
<td>8.87</td>
<td>1.81 (Japan)</td>
<td>0.15</td>
</tr>
<tr>
<td>Bunker Fuel</td>
<td>1</td>
<td>0.980</td>
<td>10.8</td>
<td>10.6</td>
<td>0.59 (Global Avg)</td>
<td>0.055</td>
</tr>
</tbody>
</table>

LPG, diesel and gasoline costs from Reference\textsuperscript{64} which include local taxes and delivery costs. Other data from reference.\textsuperscript{67} Bunker fuel prices from reference.\textsuperscript{68} Ammonia price from references\textsuperscript{4,63}

#### 5.1 Vehicle Engines

Internal combustion engines (ICE) can be converted to run on ammonia, as has been known for some time. Researchers from South Korea have road-tested a passenger car with a converted spark ignition engine that runs on a hybrid fuel containing 70% ammonia, 30% gasoline.\textsuperscript{69} There is also a number of ongoing projects aiming to convert compression ignition truck engines.\textsuperscript{70} Research groups including Toyota are developing various ICE modifications for their engines to run on ammonia.\textsuperscript{71} Dual fuel and hybrid engine designs for vehicles based on ammonia fuel have also been studied and assessed.\textsuperscript{66,72}

NO\(_x\) emission from vehicle engines, generally, is a well-known issue and has been a subject of development of mitigation technologies for many years. Control solutions based on implementing catalytic converters are well developed and would be readily transferred to ammonia fueled engines.\textsuperscript{73,74}

#### 5.2 Marine Engines

Application of ammonia as a marine fuel is noticeably attracting attention of the shipping/logistics industry,\textsuperscript{75} as a response to recent mandates by the International Maritime Organisation regarding lowering of sulphur content of fuels and also the ultimate decarbonisation of shipping by 2050. A thesis by de Vreis provides a detailed assessment of ammonia suitability for this industry.\textsuperscript{61} In particular, the study demonstrates that ammonia
fuelled marine ICEs are comparable to those using diesel fuel in terms of power generation and NOₓ emissions. MAN Energy Solutions, which is part of the Volkswagen Group, is currently developing an ammonia fueled marine two-stroke engine and has published a perspective document concerning this. The European-based Transport and Environment Group has estimated potential ammonia usage in marine applications of at least 1.2 PWh year⁻¹ in Europe alone by 2050. The report puts this in the context of current EU electricity generation in 2015 of 3.2 PWh year⁻¹. A detailed comparative analysis with traditional fuels also shows that the use of ammonia as a fuel results in a lower environmental impact in terms of overall ecotoxicity, and ocean acidification.

5.3 Power generators

For a host of remote-community and off-grid situations diesel generators are currently the power source of choice, supplemented by solar and wind, and often at high fuel costs given the delivery distance. In this setting, small to medium scale generators running on ammonia that is produced and stored locally becomes a competitive concept. Aspects of the use of ammonia as a fuel in generators have been reviewed recently. A number of small engine demonstrations are currently underway. As an example, a 3.5 kW power generator has been adapted to run in a dual-fuel mode with ammonia content of up to 80%. These authors refer to the vehicle NOₓ emission control approaches mentioned above as suitable measures to limit NOₓ emission in this technology.

The use of ammonia as a supplementary fuel in coal and gas fired power generators has also been demonstrated in Japan. This would assist in transitioning such facilities towards lower carbon emissions as soon as sufficient quantities of ammonia become available at competitive prices. Ultimately, this particular direction in the roadmap leads to ammonia being used for large scale renewable energy storage and power generation at the grid level.

5.4 Gas Turbines

Gas turbines are another end-use modality that are capable of running on ammonia. Researchers from the National Institute of Advanced Industrial Science and Technology (AIST, Japan) have recently demonstrated the operation of an ammonia burning 41.8 kW gas turbine power generator, paving the way for large scale use of NH₃ in electricity generation. The same research team have also published an article fully dedicated to ammonia combustion technology including discussion of NOₓ emissions. They conclude
that “challenges such as NO\textsubscript{x} emissions can be overcome by the knowledge of the dynamics and chemistry of combustion”.

This accumulating knowledge may be transferred to the practical development of air-breathing ammonia-burning jet engines, making the steadily growing aviation industry more sustainable. Towards this worthy goal, a recent study comparatively evaluated a number of fuels, in which ammonia showed strong potential when compared against conventional jet fuel.\textsuperscript{83} The authors rated NO\textsubscript{x} emissions from the various fuels considered and concluded that ammonia would represent an improvement over the commonly used Jet A-1 fuel.

5.5 Fuel Cells
As an approach to making it easier and cheaper to operate hydrogen-powered fuel cells, the use of NH\textsubscript{3} as a fuel has recently attracted great attention as part of an integrated fuel processor + fuel cell concept. CSIRO has recently patented a metal-based selective H\textsubscript{2} permeable membrane combined with a pilot-scale NH\textsubscript{3} cracking reactor for the production of high-purity H\textsubscript{2} from NH\textsubscript{3}.\textsuperscript{9} The technology has been demonstrated with production rate of 5 kg H\textsubscript{2} per day, and the pure H\textsubscript{2} obtained by NH\textsubscript{3} cracking was successfully dispensed into fuel-cell powered electric vehicles.

NH\textsubscript{3} can be also directly used in solid oxide fuel cells (SOFCs) without any external reformer/cracker reactors. NH\textsubscript{3}-fueled SOFCs are generally operated at high temperature (600-800 °C) which promotes rapid decomposition of NH\textsubscript{3} into N\textsubscript{2} and H\textsubscript{2}, such that the active material is actually hydrogen. Such NH\textsubscript{3}-fuelled SOFCs have become well acknowledged by the R&D field as an important high efficiency technology for direct conversion of ammonia energy into electricity.\textsuperscript{80} According to the recent article from the Ammonia Energy Association,\textsuperscript{84} a 2 MW NH\textsubscript{3} SOFC will be installed on an offshore vessel, Viking Energy, in late 2023.

The Direct Ammonia Fuel Cell (DAFC) is another attractive alternative as an ammonia fuelled power source, especially for small-scale and domestic applications. In the DAFC, NH\textsubscript{3} is directly oxidised to N\textsubscript{2} in an alkaline electrolyte, coupled with simultaneous O\textsubscript{2} reduction to OH\textsuperscript{-}. The DAFC is suitable for transportation and mobile applications due to the combination of high energy density of liquid NH\textsubscript{3} with moderate operating temperatures.\textsuperscript{85}
In both, SOFC and DAFC type fuel cells there is a potential for NO\textsubscript{x} generation, however in these early stage technologies the control of NO\textsubscript{x} emissions has yet to be investigated in detail.

6. Perspectives on the Global N-Cycle

A substantial shift from an economy based on fossil carbon energy to one based on ammonia must involve careful consideration of the environmental impacts caused by the production and use technologies concerning this new fuel. Just as there is a complex web of biogeochemical processes that make up the global carbon cycle, so also the planetary nitrogen-cycle is complex and not entirely well-understood. It is obviously important that humankind does not avoid one crisis revolving around CO\textsubscript{2} emissions, by creating another crisis involving NH\textsubscript{3} and NO\textsubscript{x} emissions.

Figure 4 shows an overview of the global nitrogen cycle, as summarised by Fowler et al. in Philosophical Transactions in 2013,\textsuperscript{86} with additions from the present authors based on data from the same paper, to indicate the relative importance of the present anthropogenic production of ammonia. The diagram shows the main atmospheric, land-based and marine cycles that exchange between N\textsubscript{2} and the various important forms of “fixed” nitrogen (fixed = all forms of inorganic and organic/biochemical nitrogen except N\textsubscript{2}). The downward arrows indicate fixation of nitrogen by various natural process including plant based biochemical nitrogen fixation (BNF). The H-B process and other “loads” due to anthropogenic activity are shown on the left. The upward arrows show re-emissions to the atmosphere by various processes.

One of the key messages from this diagram is that anthropogenic nitrogen fixation already represents an amount of fixed nitrogen compounds per year that is about equivalent to the “natural” processes. Since the Earth’s cycling and re-emission systems have to ultimately deal with this extra fixed nitrogen, it is important to recognise that the total load on these natural processes has already doubled in the last 100 years since H-B technology came into widespread use. Noting that a substantial fraction of the fixed nitrogen ends up as nitrate which flows through the aquatic systems to the oceans and that there are marine N-cycles that have very long time-scales (half-life, t\textsubscript{0.5} \sim 100) years, one is forced to observe that there are unknown outcomes here. In other words, the fate of this doubling of fixed nitrogen within the N-cycles that has occurred due to anthropogenic activity is not completely accounted-
for, or understood. In particular, the impact of increasing nitrate concentrations on marine ecosystems is an issue that needs increased attention.

Even if the Ammonia Economy is thoroughly and rigorously targeted towards the $\text{N}_2 \rightarrow \text{NH}_3 \rightarrow \text{N}_2$ cycle that is shown in Figure 1, there will be inevitable adventitious losses and inefficiencies, producing further emissions of $\text{NH}_3$ and $\text{NO}_x$, that will add additional load to the cycles in Figure 4. It is the view of the present authors that the further monitoring, investigation and understanding of these cycles should form a vital part of developing towards the Ammonia Economy. Development of such knowledge will provide the safety barriers, speed limits and occasional stop-signs on the pathways of our roadmap and it is important that funding agencies recognise the need for support of further research into these important processes.

**Figure 4.** Planetary nitrogen cycles and fluxes in Tg per year (BNF = biological nitrogen fixation, H-B = Haber Bosch nitrogen fixation, GPP = gross primary productivity). Curved “combustion” arrow shows nitrogen fixation fossil fuel and other combustion process and resultant emissions of $\text{NO}_x$ compounds to the atmosphere. 1 Tg = 1 teragram = $1 \times 10^6$ tonne. Redrawn and appended from Fowler et al.86
7. Roadmapping the Ammonia Economy

Our vision of the future development of the Ammonia Economy is summarised in a roadmap plot in Figure 5. Notwithstanding the growth of the global population is predicted to decelerate mid-century\(^8^7\) it is unlikely that the same scenario will apply to our global energy consumption,\(^8^5,^8^8\) as energy demand per capita is projected to continuously increase. Similarly, it can be expected that global ammonia production for the chemical industry, in the first place fertilisers, will increase faster than the population and this growth can be projected to be at least as fast as the rate of increase in energy consumption. A third important point of reference in Figure 5 is global shipping fuel use, currently estimated at 3.5 PWh per year and predicted to triple by 2050.\(^8^9\) Within the context of the above three key benchmarks, we anticipate that NH\(_3\) production will remain dominated by the current H-B technology that will be upgraded to Gen 1 \textit{via} implementation of CO\(_2\) sequestration processes over the next decade, the economic pressure towards this depending on carbon pricing. Gen 2 technology, \textit{viz.} existing and new H-B plants fed by green H\(_2\) derived from electrolytic water splitting, is likely to reach a commercial readiness index\(^9^0\) of 3 (CRI3) and emerge as a significant contributor of liquid fuels around 2030. Gen 2 ammonia production would then rapidly increase, eventually replacing current and Gen 1 processes. Given the very significant existing H-B infrastructure, it is likely that Gen 2 will continue to grow in the future and remain the baseline ammonia production in many parts of the world. Entirely electrochemical, renewable-powered Gen 3 ammonia synthesis technology is expected to enter the market at scale as soon as it achieves CRI1; we anticipate this occurring towards the end of decade and to start significantly contributing to global NH\(_3\) production thereafter, as plant size and capacity increases. However, there is little doubt that Gen 3 will rapidly become the preferred technology as soon as it reaches CRI3 and becomes an efficient industrial process. Possibly the first major market for ammonia as a fuel will be for maritime transportation, as discussed above. We envisage that Gen 3 technology will then enable production of renewable ammonia for other transportable energy storage applications, that will begin to significantly replace fossil fuels in the 2040s.
Figure 5. Ammonia Economy roadmap showing current and projected contributions of the current and Gen 1 (purple), Gen 2 (light blue) and Gen 3 (green) ammonia production technologies. Total NH₃ production is shown as a dotted orange curve. These projections are compared to the projected ammonia production for chemical industry (dashed black line; 2018 value from reference⁴, projection is based on the global energy consumption growth according to reference⁸⁵), shipping fuel use (dashed dark blue; according to the predicted 3.6% annual increase over the next three decades⁹⁹) and global fuels consumption (solid grey; 2018 value taken from the BP Statistical Review of World Energy⁹¹ projection is based on reference⁸⁸). Note the logarithmic ordinate scale. CRI is a commercial readiness index used by the Australian Renewable Energy Agency.⁹⁰
8. Conclusions

Ammonia clearly has the potential to become the dominant form of transportable renewable energy in the future, displacing fossil fuels from all but the most demanding of applications. It will sit alongside other forms of chemical energy storage, including hydrogen and renewable carbon-derived fuels, as well as battery storage for grid and local electrical energy storage, as one of the core components of renewable energy technology. In some contexts, for example buses, several of these technologies will compete with one another at the local level, but the important distinguishing feature of ammonia is its well-established ease of global transportation, by bulk carrier and pipeline. This opens up the most promising high-yield regions of the world in terms of renewable energy generation, both on- and off-shore, to global markets.

This article has aimed to set out a roadmap for navigating the pathway towards the establishment of a large scale technology and supply chain based on ammonia. Focussing on the next two to three decades of required research, development, scale-up and implementation, it is likely that Haber-Bosch based Gen 1 and Gen 2 solutions will dominate the generation technology for at least the next decade. The direct electroreduction, Gen 3, technologies face a number of roadblocks that are currently restraining their further development. These include the often-conflicting challenges of simultaneously achieving sufficient energy efficiency, as well as production rate. Fundamentally, there is no thermodynamic barrier to achieving the energy efficiency targets, if sufficiently selective catalysts can be discovered. The aqueous electrocatalysis pathway that is addressing this challenge has been fraught with false-positive issues in recent years, but the 2020s goal to discover and optimise genuine catalysts for this process clearly remains valid. The mediated electroreduction pathway has definite potential, if the energy efficiency can be improved by development of less energy consuming redox mediators, which may be metals or alloys, or potentially complex structures/molecules.

Economic factors, including the cost of renewables and the capital intensity of the production plant, are significant in determining the time scale of these developments. Underlying these costs are technical performance challenges, including areal yield and service lifetime of electrolysis cells. These factors appear in both Gen 2 and Gen 3 variations, wherein the cost of the cell components and the related balance of plant are similar and substantial. The issue of intermittency in different energy supply contexts is a significant factor in
understanding techno-economic viability in either case. Though technically the electrolysis processes can work with variable supply, the capital intensity of the plant points towards the need for high capacity factors, and the inclusion of energy storage (or H\textsubscript{2} storage in Gen 2).

Our roadmap has also considered the increasingly diverse range of applications of ammonia as a fuel that is emerging. At the same time, it is vital that the roadmap always considers the broader, global environmental impacts of large-scale ammonia use as a fuel. It is clear that our understanding of the mechanisms of the global nitrogen-cycle is incomplete, in particular in regard to the slow turn-over in marine cycling of N-compounds. Hence investment in technology development should also include investment in further basic science research on the environmental impacts of increased quantities fixed nitrogen and policy makers must mandate strong emissions standards and controls at an early stage.

Nonetheless the roadmap presents an achievable vision for sustainable global energy supply in the future, one that we believe deserves serious consideration and investment from government and business alike.

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10. Author Contributions

All authors contributed equally to the writing of this article.

11. Declaration of Interests

The authors have patent applications related to this work.

12. References
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