The Environmental Feasibility of Decentralised Solar Ammonia

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Abstract: Intense efforts have been devoted to developing green and blue centralised Haber-Bosch processes (gHB and bHB, respectively), but the feasibility of a decentralised and sustainable scheme has yet to be assessed. Here we reveal the conditions under which small-scale systems based on the electrocatalytic reduction of nitrogen (eN₂R) powered by photovoltaic energy (NH₃-leaf) could become a competitive technology in terms of environmental criteria. To this end, we calculated energy efficiency targets based on solar irradiation atlases to guide research in the incipient eN₂R field. Even under this germinal state, the NH₂-leaf technology would compete favourably in sunny locations relative to the business-as-usual production scenario. The disclosed sustainability potential of NH₃-leaf makes it a strong ally of gHB toward a non-fossil ammonia production.

Keywords: Ammonia leaf · Green ammonia · Life cycle assessment · N₂ electroreduction



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1. Introduction

Besides its key role sustaining the fertilizer industry, ammonia is now seen as a future energy carrier, a scenario for which its production will require notable scaleup.^[1,2] Its synthesis and distribution currently develops in a heavily centralised system with the business as usual (BAU) fossil-based Haber-Bosch (HB) process at its heart. The development of sustainable alternatives within this scheme are thus in high demand.^[3,4] The main options under consideration are blue HB (bHB), resulting from coupling HB with carbon capture and storage, and green HB (gHB),^[2,5] based on green hydrogen obtained from water electrolysis or biomass gasification.^[3,5] Recently, the electrocatalytic reduction of nitrogen^[6–8] (eN₂R) emerged as an alternative pathway toward green ammonia not requiring hydrogen and displaying a straightforward coupling to renewable energy sources.^[9] Even though the superior environmental performance of these routes has been established,^[3,10-13] there is also a consensus on their inability to economically outcompete the BAU anytime soon.[12-14]

A decentralised scheme represents a different and complementary approach to tackle ammonia production and distribution, with the potential to bring additional benefits, particularly in remote locations.^[15] The environmental feasibility of this concept, however, has not been assessed so far. Here we analyse the impact of small reactors performing eN,R coupled with photovoltaics, a configuration recently coined as ammonia leaf (NH₃-leaf)^[6,9] using a life cycle assessment framework. Moreover, we provide energy efficiencies targets suggesting the immediate potential of NH₃-leaves to effectively complement sustainable centralised alternatives.

2. Assumptions and Model

2.1 General Assumptions

The NH_3 -leaf system was assumed to be deployed on a farm able to fertilise one ha of wheat, for a total nutrient production target equal to 100 kg N ha⁻¹ a⁻¹.^[16]

The main components of the decentralised NH_3 -leaf system, depicted in Fig. 1 are: (I) an electrolyser, which converts water and atmospheric nitrogen into ammonia, hydrogen, and oxygen; (II) a fuel cell, to reconvert the by-products hydrogen and oxygen into electrical power and water, thereby reducing the overall electricity and water consumption; and (III) solar panels supplying the primary electrical power. The modularity of all the system components allows to deploy NH_3 -leaves next to the fields fertilised with the produced ammonia. The considered lifespan of the system is 30 years, matching that of solar panels.^[17,18] The system attains instantly steady-state conditions when power is available for the electrolyser, *i.e.*, we assume that the ramp-up and ramp-down times due to the intermittent energy input are negligible.



Fig. 1. Schematic representation of the NH₃-leaf setup and auxiliary equipment. Water, electrolyte (0.1 M KHCO₃ in this study), air, and light are required inputs. The electrolyser produces diluted NH₃ at the cathodic chamber. A fuel cell fed by the produced H₂ and O₂ in the electrolyser enables the recycling of electrical energy by valorising these undesired products to increase the energy efficiency of the overall system. No electrical power is exchanged with the grid.

2.2 Electrolyser

The electrolyser runs with an aqueous solution and nitrogen entering the cathodic chamber and water at the anodic one. Two reactions occur in parallel, namely, the nitrogen reduction to ammonia (Reaction 1), and water splitting (Reaction 2).

$$N_2 + 3H_2O \rightarrow 2NH_3 + \frac{3}{2}O_2 \qquad (Reaction 1)$$

$$H_2O \rightarrow H_2 + \frac{1}{2}O_2 \qquad (Reaction 2)$$

A polymeric membrane separates anodic and cathodic chambers. A mildly diluted solution of potassium bicarbonate solution (0.1 M KHCO_3) was chosen as a representative electrolyte, since it contains elements that can be safely delivered to the crops, in such quantity.^[19] We considered that 47% of the total water fed into the electrolysis cell reacts.^[20] Nitrogen is separated from air with a pressure swing adsorption (PSA) unit, which consumes 0.365 kWh kg⁻¹ N₂,^[21]

The system operates at 25 °C and 1 bar. We consider a theoretical cell voltage $U_{\rm th} = 1.17$ V to ideally operate nitrogen electroreduction with the oxygen evolution reaction as the anodic half-reaction;⁽²²⁾ moreover, an overpotential of 0.3 V at each electrode^[14,23] and an additional ohmic drop of 0.1 V^[24] were assumed to calculate the cell voltage. Accordingly, the total voltage efficiency was calculated with Eqn. (1):

$$\eta_{\rm V} = \frac{U_{\rm th}}{U_{\rm th} + U_{\rm op}} = \frac{U_{\rm th}}{U_{\rm applied}} \tag{1}$$

where η_v is the overall voltage efficiency, defined as the share of the applied energy that is effectively used to produce ammonia and hydrogen, and U_{op} is the total overpotential, calculated as the sum of the overpotentials at the electrodes and the ohmic drop, and $U_{applied}$ is the effectively applied voltage, *i.e.*, the sum of the theoretical voltage and the total overpotential. We estimated a voltage efficiency of 63% under these conditions.

We considered different Faradaic efficiencies to ammonia. We accounted for this by computing the ratio of electrical power involved in Reaction 1 to the useful electrical power (η_F). The range of Faradaic efficiencies spanned from a representative figure of the state-of-the art, obtained with a nitrogen-defective carbon nitride-based catalyst (34%),^[23] up to 100%. The only considered by-product was hydrogen. The energy conversion efficiency (ECE) of the electrolyser η_{ECE} can be calculated as follows:

$$\eta_{\text{ECE}} = \eta_{v} \cdot \eta_{F} \tag{2}$$

The lifespan of the unit's active components (electrodes in the case of the electrolyser, the greatest part of the whole stack in the case of the fuel cell, including the whole membrane electrode assembly)^[25–27] was assumed to be 7 years, after which these components must be substituted and the unit is subjected to maintenance operations.^[25]

The product stream contains the electrolyte with dissolved ammonia. To minimise the electrolyte consumption,^[28] the aqueous ammonia solution is recirculated into the unit. Ammonia was assumed to leave the system at the theoretical limit of 30% w/w.[29] Since the solubility of hydrogen in water is negligible, it was separated from the product stream with a flash unit. The NH₂-leaf system is deployed at a fully distributed scale for small farms using the ammonia product for fertigation. We assumed that the produced ammonia is deployed as a fertiliser in a 100 ppm solution.^[30] Hence, a total of 45.33 L of water are needed to dissolve one kg of ammonia. For example, considering that the diluted ammonia is used to meet the nitrogen demand of wheat^[16] - 100 kg N ha⁻¹ a⁻¹ - the water in the liquid fertiliser would amount to 0.100% of the average irrigation water required by wheat.[31] This fact and the need of diluting the product stream with additional water minimise the risk of soil salinisation due to the use of bicarbonate.^[32]

The high-purity oxygen produced at the anode is partially re-combined with hydrogen in a fuel cell to reduce the overall system's power consumption. The remaining oxygen was assumed to be vented, avoiding the additional compression costs, as it is unlikely that the global market will be able to absorb it.^[33]

2.3 Fuel Cell

For Faradaic efficiencies below 100%, the produced hydrogen was sent to a fuel cell to generate electrical energy and water. Hydrogen storage was not considered, under the assumption that the electrolyser and the fuel cell can work under steady-state conditions when the electrolyser is active. We assumed that the output water was used in the electrolyser. An average fuel cell efficiency of 60% with respect to the lower heating value of hydrogen was considered.^[34] Since this technology has reached an incipient mature stage, only minor improvements in this parameter are expected in the future.^[35] A five-year lifespan was assumed for the active components, after which replacement of components is required.^[26]

2.4 Solar Photovoltaic Panels

The power provided to the electrolyser was generated with photovoltaic (PV) panels deployed on open ground, with a lifetime of 30 years^[17] and a solar-to-power efficiency of 20%.^[30] Depending on the location, a solar radiation of 94–281 W m⁻², corresponding to an average incident solar radiation of 2.25–6.75 kWh m⁻² d⁻¹ was considered.^[36] At the same time, the PV capacity factors (*i.e.*, ratios of actual yearly power output to ideal annual power output at full capacity) vary from 5.6% to 26.26%,^[37,38] based on a global grid of 1140 points, spacing each point by 6° in longitude and 8° in latitude. An average value of 167 W m⁻² for the incident solar radiation and 10.98% for the PV capacity factor was selected based on the average PV plant available in the Ecoinvent database.^[39]

3. Results and Discussion

3.1 Potential Environmental Benefits

We first study the cradle-to-gate impact of producing the ammonia used for fertilisers worldwide.^[40,41] To this end, we assessed the impact on two ReCiPe 2016 indicators using the hierarchical approach, *i.e.*, global warming impacts and human health (Fig. 2), comparing the NH₃-leaf against BAU, bHB and gHB based on electrolytic hydrogen powered by solar energy. The performance of the solar-dependent technologies (*i.e.*, gHB and NH₃-leaf) varies across locations; therefore, a range of performance levels is provided considering the lowest and highest capacity factors attained by solar photovoltaic (PV) panels.^[37,38] The range for the NH₃-leaf considers also current and maximum Faradaic efficiencies.

The results show that the BAU scenario has very relevant global warming impacts, *i.e.*, 290 Mt CO₂-eq a⁻¹, a value almost equivalent to the entire emissions of France. Moreover, the same scenario has also significant burden on the human health category (63 DALYs mio⁻¹ inhabitants), with comparable magnitude to the global impact of acute hepatitis C. A prominent feature of this analysis thus emerged: the largest potential benefits of alternative routes *versus* the BAU system will thus emanate from their ability to curb CO₂ emissions. The bHB scenario, which still relies on fossil resources, reduces the global warming impacts drastically and, to a lesser extent, the damage to human health. However, this route faces issues related to the need for geological storage and the impact of methane leaks.^[42,43] Moving to the gHB, we find that a state-of-the-art water electrolyser (69% efficiency^[44]) in the sunniest locations could perform similarly to the bHB.

The sustainability potential of NH₃-leaf becomes evident in the selected impact categories, outperforming all other technologies for the best conditions, despite performing the worst in the scenarios with low efficiencies in poorly irradiated regions. Specifically, NH₃-leaf is extremely appealing for high Faradaic efficiencies and high capacity factors, attaining impact reductions in the global warming and human health indicators similar to those of the bHB scenario.

To get further insight into the locations where the decentralised NH_3 -leaf scheme could become competitive, we next performed an environmental analysis considering regional sun power



B) Human health



Fig. 2. Environmental performance in terms of global warming impacts (A) and human health (B) of the four assessed ammonia production technologies, ammonia leaf (NH₃-leaf) (green), green Haber-Bosch (gHB) process (yellow), blue Haber-Bosch (bHB) process (blue) and business-as-usual (BAU) (grey).

availability based on solar irradiation maps. According to this, PV capacity factors range from 5.6% in the Antarctic ocean (65°S, 160°W) to 26.3% in the Atacama desert (24°S, 69°W), with an average value across locations of 11.0%. We then computed the range of impact values that would be attained in the global warming indicator for this range of PV capacity factors considering energy conversion efficiencies (ECE) from the currently attainable one, 21%, to 63%, corresponding to a 100% Faradaic efficiency. Breakeven ECEs relative to the BAU could then be calculated.

Our analysis reveals that many densely inhabited locations show breakeven efficiencies below or close to the current reference value of 21%. For instance, the breakeven efficiencies for Madrid, Sao Paulo, Sidney, Hong Kong, or Chennai and the surrounding areas vary from 10.7 to 12.4% for the selected indicator (relative to the BAU).

Our calculations also showed a stark regional variation of breakeven efficiencies between *ca.* 10 and 40% for different locations depending on the capacity factors available in largely and poorly irradiated locations, respectively. This factor may enable the gradual geographical penetration of NH₃-leaf as it reaches maturity. Table 1 summarises the figures of merit to guide the development of eN_2R electrocatalysts for NH₃-leaf. Since Faradaic efficiency is a more easily accessible parameter for catalysis practitioners, we determine the corresponding values, resulting on range oscillating between 16 and 100% (Table 1). In view of the previous analysis, it is possible to claim that an electrolyzer operating at 48% Faradaic efficiency with 0.3 V cathodic overpotential (*i.e.*, 30% ECE) would be environmentally advantageous across almost all populated areas of the Earth.

4. Conclusions

Here we develop the first environmental assessment of sustainable decentralised ammonia production. A widespread implementation of the NH₃-leaf technology is feasible upon further efforts in catalyst and reactor design in eN_2R . An analysis considering regional values of PV capacity factors reveals that electrolyzers showing *ca.* 30% energy efficiency – corresponding to 48% Faradaic efficiency under commonly reported conditions – Table 1. Range of figures of merit based on environmental assessments for the development of NH_2 -leaf.

Breakeven energy efficiency (%)	Faradaic efficiency NH ₃ ^a (%)
10	16
20	32
30	48
40	63
50	79
60	95
63	100

^a0.3 V cathodic and anodic overpotentials and 0.1 V ohmic drop

would environmentally outperform the fossil HB process across all densely populated areas in the world. Outperforming its counterpart gHB will require a mild additional gain of 8% in energy efficiency in the sunniest locations. Overall, our results call for further research on this technology to develop a decentralised defossilised ammonia production scheme.

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