

Ammonia Production

HIGHLIGHTS

Ammonia - Ammonia is critical in the manufacturing of fertilizers, and is one of the largest-volume synthetic inorganic chemicals produced worldwide^{1, 2}. Moreover, ammonia is an ideal carbon-free energy carrier and storage material^{3, 4}.

Processes and technology status – The main industrial procedure for the production of ammonia is artificial nitrogen fixation called Haber–Bosch process. In this process, nitrogen (N_2) reacts with hydrogen (H_2) both at gaseous states under high pressures of 150 to 200 bar and temperature 500 °C. The overall reaction is $N_2 + 3H_2 \rightarrow 2NH_3$ ^{5, 6}. Feedstock for the Haber process are air and natural gas to supply N_2 and H_2 respectively. The other raw materials are coal and naphtha which release more than double greenhouse gases (GHGs) in comparison to natural gas⁵. Figure 1 depicts the ammonia production plant utilizing natural gas as feedstock. Novel electric assisted technologies such as direct electrochemical ammonia synthesis and solid oxide electrolyzers are, also, under development⁷.

Cost - Haber–Bosch production cost is the cost of the feedstock and the energy consumed, that is, 80.36 €₂₀₁₉/t for natural gas and 199.11 €₂₀₁₉/t for hydrogen by water electrolysis⁸.

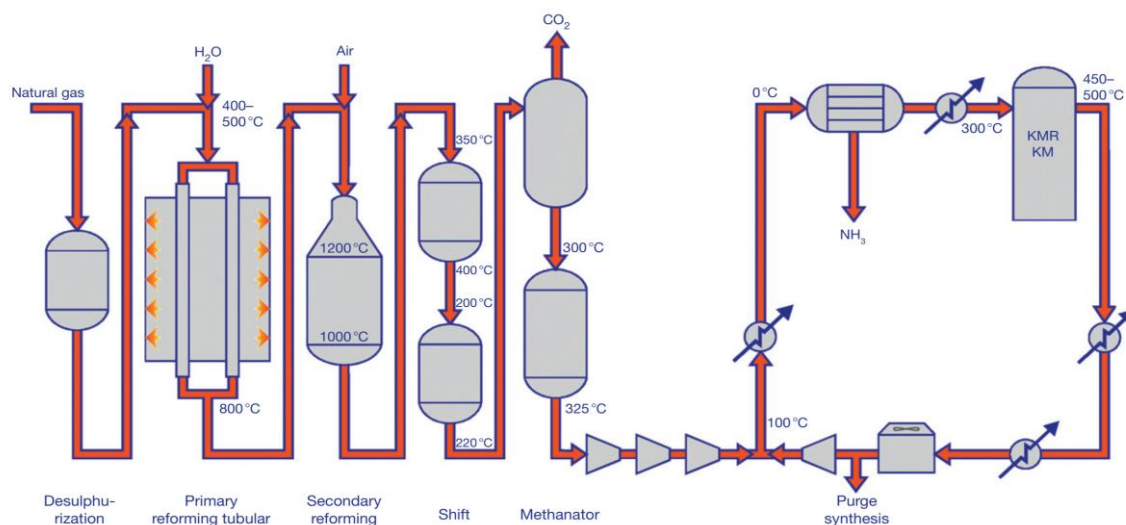


Figure 1. Process scheme of ammonia synthesis - Haber–Bosch process⁹

¹ Converted from Dollars to Euros, 1 €₂₀₁₉ = 1.12 \$₂₀₁₉³²

Capital investment for conventional Haber–Bosch facilities are essentially equivalent at 276.11² M€₂₀₁₇ for 2000-tpd capacity. Grundt et al. estimated capital expenses of 176.99 M€₂₀₁₇ for a 1000 tpd ammonia facility ¹⁰.

Potential and barriers – Ammonia has the potential of being an energy carrier during and after the energy transition period ¹¹. As the barrier, ammonia production requires high pressures and temperatures. Moreover, the process requires a large amount of energy and generates significant CO₂ emissions ⁵. Hence, ammonia plant revamps toward energy efficiency, carbon dioxide (CO₂) removal system, beside the actual fuel and feedstock switching ^{12, 13}.

Process overview - The first attempts toward ammonia attainment were carried out through the dry distillation of nitrogenous vegetable and animal products ¹⁴; by the reduction of nitrous acid and nitrites with hydrogen; and also by the decomposition of ammonium salts ¹⁵. Today, most ammonia is produced on a large scale by the Haber–Bosch process which is an artificial nitrogen fixation process. In this process, N₂ and H₂ gases are allowed to react at pressures of 200 bar and temperatures around 500 °C ⁵. The stoichiometric value of feedstocks are 1:3 of N₂:H₂, while depending on the process, catalyst type and reaction temperatures the different ratios of N₂:H₂ are required for the optimum ammonia production ¹⁶. Nowadays, conventional Haber Bosch plants produce ammonia using natural gas

(50%), oil (31%) or coal (19%) as feedstock ¹⁷. Natural gas is the preferred feedstock for ammonia production due to its low price and wide availability. Moreover, other high carbon content feedstocks such as coal and naphtha, release more than double GHGs amount, making them less attractive ^{5, 18}. A conventional process scheme utilizing natural gas as feedstock is shown schematically in figures 1, 2 ^{9, 19}.

Desulphurization of the natural gas - The first step in the process is desulphurization of the natural gas feed in order to avoid sulfur poisoning of the downstream catalysts.

Reforming - The second step is steam reforming, where the natural gas is reacted with steam to produce an equilibriu

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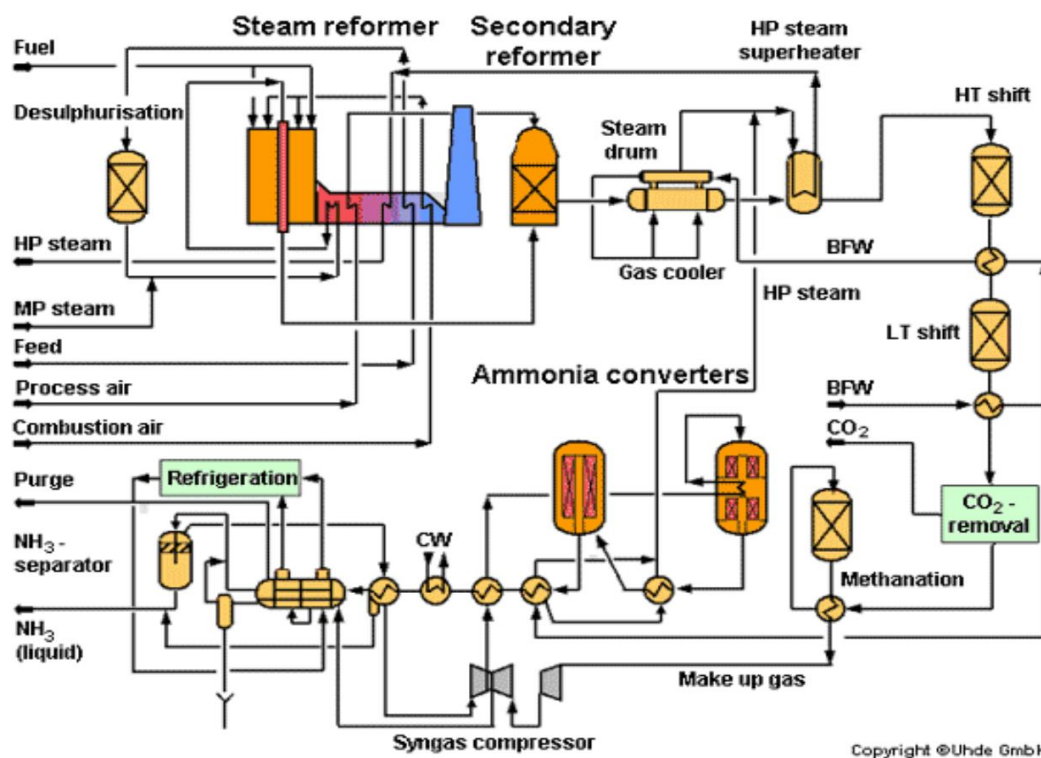


Figure 2. Conventional ammonia production plant

mixture of H₂, CO, CO₂, and CH₄. This reaction is endothermic, so it is carried out in tubular reactors placed in a heated furnace in order to supply the heat for the reaction and to maximize the equilibrium content of the desired products H₂ and CO.

N₂ from air - Following steam reforming, air is added to supply the N₂ required for ammonia production, while the O₂ from the air converts the remaining CH₄ in an exothermic reaction that increases the temperature and the H₂ and CO content further. The reaction mixture is then cooled and followed by a two-step water–gas shift process, where CO is reacted with steam and transformed into H₂ and CO₂. Since all

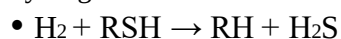
oxygen-containing molecules poison the ammonia synthesis catalyst, CO₂ is subsequently removed by absorption and a final CO/CO₂ cleanup is carried out, for example, by the methanation reaction⁹. Methanation is the reaction by which carbon oxides and hydrogen are converted to methane and water. The reaction is catalyzed by nickel catalysts²⁰.

Compression and the main reaction - Before sending the synthesis gas into the ammonia reactor, it is compressed and, at a point before it reaches the ammonia catalyst, water is removed by condensation. As mentioned above, ammonia synthesis is an equilibrium-

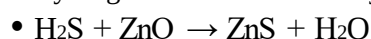
limited exothermic reaction, and the ammonia product is favored by low temperature and high pressure ⁹. The general reactions at the explained steps are categorized as the following list.

Desulphurization in a two-step process ¹⁹:

– Sulfur compounds catalytically hydrogenated to H₂S:

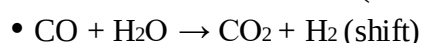


– Hydrogen sulfide removed by zinc oxide

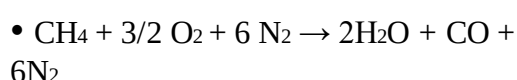
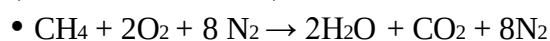


Reforming in a two-step process to achieve correct H/N ratio:

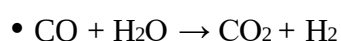
– Primary reforming:



– ‘Air-blown’ secondary reforming (additional reactions):

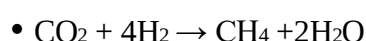
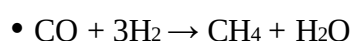


Catalytic shift conversion to convert CO to CO₂ and H₂:

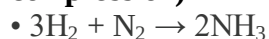


Carbon dioxide removal via an amine process

Methanation to remove residual amounts of CO or CO₂:



Ammonia conversion (after compression)



Recycling of unreacted gases - Most of the N₂ and H₂ passes unreacted through the reactor, and must be recycled in the loop after being cleaned for ammonia. Since the synthesis gas that is fed into the loop contains some inert gases (CH₄ and Ar), a purge is also needed in the loop to avoid too large a build-up of these diluents ⁹.

Investment and production costs -

The price of ammonia is closely related to the price of the feedstock ¹⁰. According to gross estimations, approximately 2/3 of consumed natural gas is used as a feedstock, while around 1/3 is used for energy purposes, and, it makes up approximately 70-85% of the ammonia production costs ²¹. Haber–Bosch production cost is the cost of the feedstock and the energy consumed, that is, 80.36 €₂₀₁₉³/t for natural gas and 199.11 €₂₀₁₉/t for hydrogen by water electrolysis ⁸. Capital investment for conventional Haber–Bosch facilities are essentially equivalent at 276.11⁴ M€₂₀₁₇ for 2000-tpd capacity ¹⁰. Grundt et al. estimated capital expenses of 176.99 M€₂₀₁₇ for a 1000 tpd ammonia facility ¹⁰.

Carbon capture and storage (CCS) in ammonia production - Ammonia

³ Converted from Dollars to Euros, 1 €₂₀₁₉ = 1.12 \$₂₀₁₉ ³²

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mainly used in the fertilizer industry and that is responsible for around 2–3 % of the world greenhouse gas emissions ²². A modern, optimized and highly efficient methane-fed Haber-Bosch process emits 1.5–1.6 t_{CO₂-eq}/t_{NH₃} making the global manufacturing of ammonia accounting for 1.2% of anthropogenic CO₂ emissions. This value would further increase if CO₂-eq emissions associated to the extraction and transport of natural gas are included ¹⁷. CO₂ is generated primarily from two sources in the ammonia production via Haber-Bosch process: combustion of fuel in the primary reformer and removal of CO₂ from the syngas stream. The CO₂ emissions stream released from the syngas are already highly purified, being approximately 99% CO₂. Syngas CO₂ emissions are regularly captured at ammonia facilities to produce urea and is not contributed to environmental emissions nor available for CO₂ capture and storage (CCS). Hence, CCS can be investigated for the combustion emissions from the primary reformer ²³. Including CO₂ mitigation in the ammonia production allows to reduce the emissions but leads to a slight efficiency decrease due to the energy consumption for the CO₂ compression. For the natural gas fed process yielding an energy efficiency around 65 %, the overall life cycle emissions can be reduced to 0.79

kg_{CO₂}/kg_{NH₃} with CO₂ capture compared to 1.6 kg_{CO₂}/kg_{NH₃} without capture ²². Hence, designing new ammonia plants with integrated carbon capture and storage (CCS) or retrofitting CCS to conventional plants does have notable potential ²⁴.

Energy requirements of ammonia production -

The conventional highly optimized Haber–Bosch process uses about 7.9 kWh of energy derived from fossil fuels per kg NH₃ at the scale of 1,000 t/day, where 2.0 kWh/kg (including process heat credit from ammonia synthesis) is used for pressurization, heating, pumping and so on. This number will be higher at a smaller scale due to the increased heat losses ⁸.

The minimum energy requirement for the Haber-Bosch process, defined as the heat of combustion of ammonia, is 18.6 GJ/t_{NH₃} based on the lower heating value of ammonia (LHV). This is the amount of energy chemical stored. For the methane fed process, the theoretical minimum energy input is 22.2 GJ/t_{NH₃} broken down as 17.7 GJ/t_{NH₃} associated to the methane feedstock and 4.5 GJ/t_{NH₃} associated to methane fuel to fire the steam methane reforming (SMR) reactor ¹⁷. For comparison purposes, the energy requirement for the direct electrochemical synthesis of NH₃ from liquid water and

nitrogen at 25 °C and 1 bar is 19.9 GJ/t_{NH₃} (1.17 volts)¹⁷. Direct electrochemical ammonia synthesis and solid oxide electrolyzers without the need for Haber-Bosch or an air separation unit may offer large cost reduction of the production of green ammonia⁷. However, electrochemical synthesis of ammonia present a low selectivity and low throughput at present, which increases its energy consumption far beyond the methane-fed Haber Bosch process¹⁷.

Ammonia as an energy storage

molecule - Recent studies have examined ammonia as an energy storage molecule and have ranged in focus from electrical energy transport in ammonia, to a comparison of hydrogen sources, to the implementation with actually renewable energy grid including islanded grid systems¹⁷. Ammonia is an ideal carbon-free energy storage material due to its high energy density (4.32 kWh/L), high weight fraction of hydrogen (17.65%) and ease of liquefaction under mild conditions³.

Energy storage in the ammonia chemical bonds would enable a much greater uptake of intermittent renewable power sources such as solar, tidal and wind, helping to balance the seasonal energy demands in a carbon-free society and distributed

ammonia production will find diverse applications. Energy can be delivered to the end-users by on-demand hydrogen production from ammonia in combination with fuel. Key potential of NH₃ as an energy carrier lies in its high volumetric energy density (15.6 MJ/L) which is 9 times more than that of Li-ion batteries (1.73 MJ/L) and almost 3 times that of compressed H₂ (5.5 MJ/L at 70 MPa)¹¹. cells¹⁷. Key potential of NH₃ as an energy carrier lies in its high volumetric energy density (15.6 MJ/L) which is 9 times more than that of Li-ion batteries (1.73 MJ/L) and almost 3 times that of compressed H₂ (5.5 MJ/L at 70 MPa)¹¹. This opens an opportunity to use NH₃ as a low-carbon energy storage medium that can be traded globally¹¹. At ambient temperature, NH₃ can be readily liquefied if a pressure of about 10 bar is maintained or by cooling to -33 °C at atmospheric pressure. Consequently, the cost associated with storage and transportation of liquid NH₃ is expected to be lower than that for compressed hydrogen (H₂) at 700 bar¹¹.

Furthermore, green ammonia has emerged as a promising fuel option especially for long-distance shipping because of its low carbon footprint⁷. Moreover, ammonia is a candidate for power generation, mobility purposes, and as a fuel for the internal

combustion engines ²⁵. Ammonia can, in theory, be combusted to produce only nitrogen and water as emissions, although in practice, nitrous oxides (NO_x) are often also generated, especially if the combustion happens at higher temperatures and/or under pressure, such as in a vehicle engine. However, ammonia can be utilized as a fuel for chemical processes and technologies that are used to convert chemical energy into electricity and heat ²⁶.

Electrification of ammonia production - The overall reaction of ammonia synthesis from N₂ and H₂ is exothermic, the dissociation of dinitrogen is considered as the rate-limiting step due to the high dissociate energy (9.8 eV). Therefore, high temperature and high pressure are used in the conventional thermal catalysis approach to improve the kinetics and shift the equilibrium in favor of ammonia respectively ²⁷. Current Haber–Bosch process evolved in the context of fossil fuels as the only feasible energy source, which led to its false optimization to accommodate the inefficiencies in hydrogen production from fossil fuels (e.g. methane). Indeed, the process is not optimized to reduce carbon emissions beyond reducing the methane feed and fuel requirement ¹⁷.

In comparison to the conventional ammonia process, the sustainable future of the Haber-Bosch process (and the chemical industry in general) relies on the use of renewable energy as part of what is generally called electrification of the chemical industry ¹⁷. Renewable energy has the potential to provide all the energy requirements, replacing methane as both feedstock and fuel ¹⁷. The estimated 2018 global production of renewable wind and solar energy of 2480 TWh is sufficient to produce the current global demand of ammonia estimated as 140 Mt y⁻¹ for 2014 requiring 1556 TWh of electricity ¹⁷. One promising solution to decarbonization of ammonia production is to generate hydrogen from water electrolysis using electricity originated from solar or wind sources. Since cost of renewable electricity generation is rapidly decreasing and scientists are making significant advances in electro-catalysts, the ammonia synthesis by green H₂ will become economically viable ²⁸. Figure 3 depicts the difference between conventional methane-fed and electrified Haber-Bosch process for ammonia production ¹⁷.

Switching the hydrogen production method from methane to hydropower-electrolysis reduces the CO₂ emissions from 1.5 to 0.38 t_{CO2-eq}/t_{NH3} (75% decrease) ¹⁷. 76% of the

methane consumed in the process is associated with the production of hydrogen via the SMR reaction and the remaining 24% of the methane is consumed as fuel to provide heat of reaction for the endothermic reforming reaction and to raise the necessary process steam ¹⁷.

Assuming that the electrically-driven Haber-Bosch process requires a 38.2 GJ/t_{NH₃} (35.5 GJ/t_{NH₃} for hydrogen production assuming 60% efficient electrolyzer and approximately 2.7 GJ/t_{NH₃} for the N₂ separation and Haber-Bosch

loop compressors), a wind powered ammonia process will have a carbon intensity of 0.12–0.53 t_{CO₂-eq}/t_{NH₃} ¹⁷.

For developing of a carbon-free ammonia production three main steps are required. First, the decoupling of plant from methane reforming e.g. by implementing of high efficient water electrolysis; second, replacement of condensing steam turbine compressors by electric compressors; third, alternating ammonia separation techniques to the decreased operating pressure ones ¹⁷.

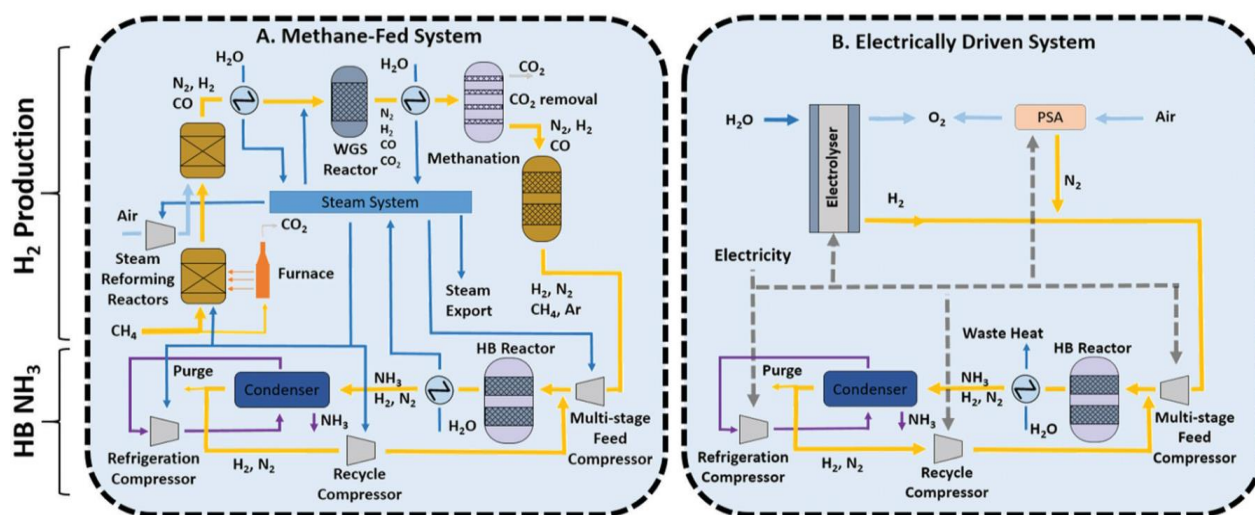


Figure 3. Schematic difference between conventional methane-fed (A) and electrified (B) Haber Bosch process for ammonia production ¹⁷.

Novel ammonia production processes

I- Biomass-to-ammonia - The ammonia production fuel decarbonization is possible through CCS, beside the usage of renewable energies and resources such as biomass²². Ligno-cellulosic biomass can be used as a source of hydrogen for ammonia production¹⁸. A biomass-to-ammonia production pathway with inputs of N_2 from air and H_2 from gasification of biomass is shown in Figure 4. The proportion of land area required for highly productive biomass species is only a small fraction (1–5%) of the grower's acreage. Thus, a small investment in land for a bioenergy crop could produce the ammonia to increase crop yield greatly on the remaining 95% of acreage¹⁸.

The other option is the development of alternative methods of production such as plasma reactions and electrochemical processes²⁴. Moreover, getting inspired from nature, chemists are developing transition-metal–dinitrogen complexes to enable the reduction of N_2 to ammonia under mild reaction conditions²⁸.

II- Ammonia production with 100% renewable energy - As a practical case, Ola Osman et. Al. have optimized the design of an industrial-scale ammonia plant (1840 Mt/day) that utilizes 100% renewable energy. They concluded that electrolysis was the most energy intensive part of the process, which if run continuously, has a specific energy consumption of 10.43 kWh/kg- NH_3 and an electric process efficiency of 37.4%²⁹.

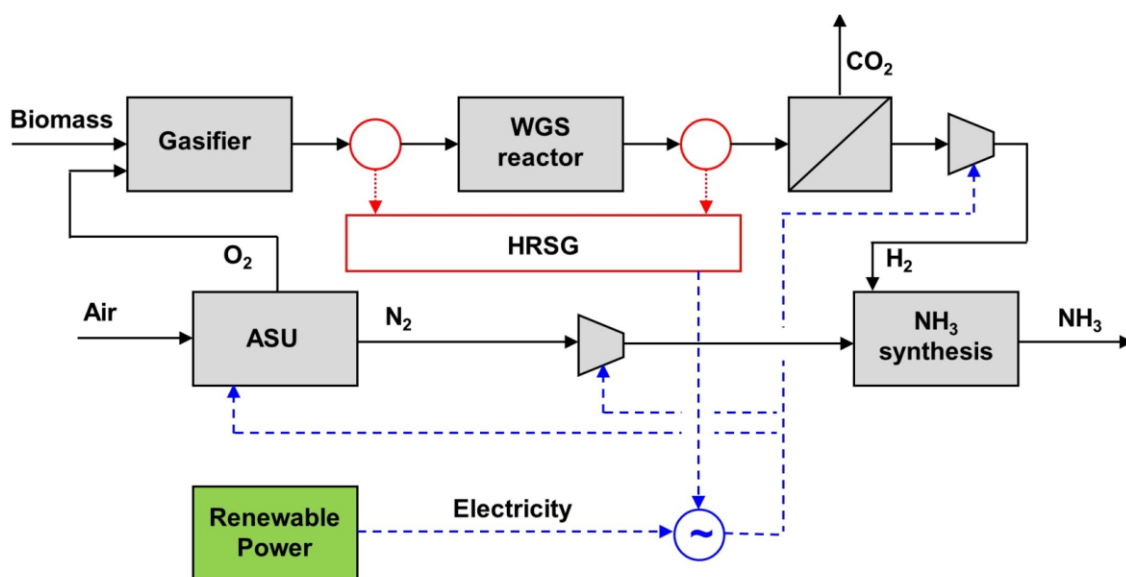


Figure 4. Simplified flow diagram of the biomass to ammonia production process¹⁸.

To adapt the ammonia production process to the variable renewable energy supply, energy storage is needed, namely batteries, thermal storage, nitrogen and hydrogen to support the ammonia synthesis that would need to be operated continuously. This storage is also used to address process limitations (limited modulation capability for the air separation unit) or as buffers for systems that can operate variably (desalination and electrolysis) ²⁹.

Moreover, using semiconductors with sunlight to drive photocatalytic reactions in a Solar Ammonia Refinery is another approach being pursued to realize environmentally friendly ammonia synthesis ²⁸. As a case in point, nanostructured metals with localized surface plasmon resonance effect, including ammonia synthesis metals Fe and Ru⁵, can concentrate the diffused solar flux to generate heat in a small volume workable for N₂ dissociation. The novel catalyst could kinetically reduce the energy barrier of ammonia synthesis ³⁰.

Furthermore, plasma, generated by ionization of gases, provides a unique way to synthesize ammonia or nitric oxide from N₂. In this process, N₂ and H₂ molecules dissociate into free radicals in the plasma.

In addition, the use of catalysts in combination with the plasma was found to promote ammonia formation from N₂ and H₂ under atmospheric pressure ²⁸, ³¹. Plasma-assisted nitrogen fixation has been considered a potential green alternative for conventional Haber–Bosch process and has recently received a lot of attention ²⁷. The optimal feed gas ratio (N₂:H₂) for blank reactor and the catalyst-packed reactor are 1:3 and 2:1 correspondingly ²⁷. Plasma-assisted nitrogen fixation has many advantages including one-step synthesis, fast-reaction and instant on-off control. More importantly, it can meet the requirement for small-scale, decentralized production, and can be better coupled with electricity generated from renewable sources ²⁷. Researches have shown that with a proper plasma-catalysts system, ammonia could be produced at a rate which is comparable with the Haber-Bosch process. The dielectric barrier discharge reactor was one of the most reported reactor types. Known as a well-recognized low-temperature catalyst for ammonia synthesis in thermal catalysis, ruthenium could be a promising choice for plasma-catalysis in which the gas temperature is normally low. ²⁷.

⁵ Fe: Iron, a metal; and, Ru: Ruthenium, a rare transition metal belonging to the platinum group

Table1. Summary Table: Key ammonia production data

Technical Performance	Desulphurization of the natural gas	Reforming
Energy type inputs	Natural gas, coal and naphtha	Natural gas, coal and naphtha
Output products	Desulphurized natural gas feed	H ₂ , CO, CO ₂ , and CH ₄
Environmental Impact		
Emitted CO ₂ For the natural gas fed process (kgCO ₂ /kgNH ₃)		0.79 with CO ₂ capture 1.6 without capture ²²
Costs		
Plant size of 2000 tpd*		276.11 M€ ₂₀₁₇ ¹⁰
Plant size of 1000 tpd		176.99 M€ ₂₀₁₇ ¹⁰
Energy requirements		Total heat ¹⁷
For the methane fed		22.2 GJ/tNH ₃
For the direct electrochemical synthesis of NH ₃		19.9 GJ/tNH ₃
Materials (For natural gas and air based production)		(kg/kg ammonia)**
N ₂		0.41
H ₂		0.18

* ton per day

** Calculated based on stoichiometric ratio and 100% conversion, taking into account the molecular weights of components

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