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FROM REFUSE DERIVED FUEL (RDF) TO NH3: A KEY PATHWAY FOR THE SUSTAINABILITY AND SECURITY OF THE FOOD PRODUCTION CHAIN / GOMEZ CAMACHO, C.E., Cecconi, S., Hischier, R., Ruggeri, B.. - ELETTRONICO. - (2022). (14TH INTERNATIONAL CONFERENCE ON SUSTAINABLE ENERGY & ENVIRONMENTAL PROTECTION SEEP2022 United Kingdom 12 September-15 September 2022).

Publisher:
SEEP2022

Published
DOI:

Proceedings of SEEP2022, 12-15September 2022, Brunel University London-UK

FROM REFUSE-DERIVED FUEL (RDF) TO NH_3: A KEY PATHWAY FOR THE SUSTAINABILITY AND SECURITY OF THE FOOD PRODUCTION CHAIN

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ABSTRACT

Among the many anthropogenic applications of ammonia, its most important role (and on which the sustainability of modern societies strongly depends) is in the food production chain. Current efforts seek to reduce reliance on fossil fuels in key supply chains, although certain renewable resources remain difficult to frame into adequate production contexts. The present study performs an *ex-ante* life cycle analysis (LCA) to evaluate the possibility of using refuse-derived fuel (RDF) as a feedstock for hydrogen supply to ammonia production processes. For this, the process is simulated using the *AspenPlus* software, which sheds light on consumption and performance, which are key inputs for the prospective LCA. Additionally, reference indirect materials and energy are compiled for the inventory. The impact assessment is performed here by using the Cumulative Energy Demand (CED) and Global Warming Potential (GWP 100y) impact categories. The results indicate that the proposed pathway could reduce the CED by about 20-30 % in ammonia production. Interestingly, the results show that RDF (a widely available anthropogenic resource) can support three times the current demand for ammonia for fertilizers production of the population that generates it.

Keywords: Ammonia synthesis, Haber-Bosch, Refuse-derived fuel (RDF), biomass gasification, CED, GWP.

1 INTRODUCTION

The terrestrial nitrogen cycles play a pivotal role in shaping ecosystems functioning on our planet. Nitrogen is a key element for all (currently) known forms of life, being essential for structural components in biological systems, such as amino acids. It also takes part in biological information encoding, accounting for an important fraction in the composition of nucleic acids. For the study of nitrogen cycles, the complexity is typically reduced by clustering some relevant phenomena, such as (natural) nitrogen fixation, nitrification and denitrification. However, the influence of anthropogenic activities on nitrogen cycles has recently become more relevant. For more than a century, the Haber-Bosch (H-B) process has supported continuous population growth. This conversion process from inert molecular nitrogen N_2 (extensively abundant in the atmosphere) into readily available reactive nitrogen for anthropogenic purposes -NH_3-, undeniably marked the beginning of an era where population growth and chemical synthesis have been strongly accelerated [1]. *Per contra*, these anthropogenic activities have also affected the natural balance of nitrogen cycles. The effects are evinced in different environmental matrices such as the atmosphere and the hydrosphere. For example, the quantity of nitrogen oxides emitted into the atmosphere has increased (and they have higher GWP than CO_2 and CH_4), due to fossil fuels combustion and due to emissions from fertilization processes [2]. In the hydrosphere, the presence of a greater amount of reactive nitrogen exacerbates the eutrophication of water bodies and it is linked to negative effects on its quality for human uses.

The reagents for the H-B process, N_2 and H_2, are routinely obtained by using air separation units (ASU) for the former, and by steam methane reforming (SMR) or partial oxidation (PO) of natural gas (or other oil-based feedstock) for the latter. Both upstream processes (ASU and SMR/PO) are required to provide the necessary flow of reagents to the proper H-B synthesis step.

$$N_{2\,(g)} + 3H_{2\,(g)} \leftrightharpoons 2NH_{3\,(g)} \qquad \Delta H^{\circ}=92 \text{ kJ/mol}$$

The reaction conditions for the H-B include high pressure (being $\Delta n<0$, high-pressure conditions shift the equilibrium reaction towards NH_3), and low to medium temperatures (the reaction is exothermic, yet a trade-off between kinetics constraints and yield is sought). Regular pressure conditions are 200-400 atm and temperatures in the 400-600 ° C range, in the presence of iron-based catalysts. The H-B process represents a mature technology; however, it has a high-energy demand in the upstream processes (ASU and SMR) and the reaction step. The energy demand of the process is estimated to be around CED_{NH3}=40 MJ/kg, which includes the auxiliary services (heat and power), in addition to the consumption of methane (both, as a reagent to produce syngas and as a fuel) and the materials requirements. In fact, if the lower heating value of NH_3 (18.6 MJ/kg) is taken into consideration in relation to its (fossil) cumulative energy demand -LHV_{NH3}/CED_{NH3}-, it is evident that only about 50% of the energy demand remains stored in it as embedded chemical energy. Moreover, at least 55 % of the CED_{NH3} corresponds to the natural gas (NG) demand of the process. NG is required as chemical feedstock for the SMR/PO processes in order to provide molecular hydrogen to the H-B reaction and NG is also the fuel for *in situ* combined heat and power (CHP) generation.

Currently, sustainable production patterns are sought that can guarantee meeting societal needs. Among them, the food production chain strongly depends on the production and availability of ammonia. Global ammonia production can be estimated at around 150 million metric tons (c. 19 kg NH_3/p) [3], of which 80 % is used in the fertilizers industry. Not only has the price of ammonia increased rapidly in the last two years (over 250 %), but also its use in other industries that rather cover exosomatic human needs (especially that of explosives). One of the options that are currently being evaluated to make ammonia production more sustainable is the incorporation of "*green*" hydrogen to supply the H-B process and thus reduce the demand for fossil inputs (such as NG) and partially decouple the dependence of ammonia on the fossil production chain and pricing schemes. In any case, syngas and hydrogen production methods (alternatives to SMR/PO) must be carefully evaluated, in order to fit into established production chains. Biomass gasification (BG) processes are known for their flexibility, allowing them to operate in a wide range of temperatures, using different gasifying agents and mixtures under variable equivalence ratios (i.e., supplied oxidant flow/stoichiometric required flow). BG processes require feedstock ideally presenting low moisture and ashes content, and elevated calorific values (LHV). BG produces syngas that contains valuable compounds that can serve as building blocks in chemical synthesis (e.g., CO, H_2, CO_2, CH_4) and liquid/solid residues (i.e., tar, char) that require suitable management strategies. Following the sustainability paradigm *(proximity-adequacy-vitality*) [4], the gasification of certain biomasses could meet the requirements to substitute the SMR/PO and provide hydrogen for H-B plants. Among the potential input resources that could meet the proximity criterion, so-called Refuse Derived Fuels (RDF) could be of interest. Unsorted municipal solid waste (u-MSW), which is the fraction aside from the recyclables collection, can undergo (physical) sorting treatments to produce RDF. RDF can be classified as an anthropogenic primary energy resource (APER), whose main exploitation is the generation of electricity and heat. Due to its availability in urban centres, it could play an adequate role within recycling schemes, where reuse is hierarchically preferred (i.e., concomitant material and energy recovery in the form of syngas) over mere energy recovery (i.e., incineration). Hence, hydrogen can be supplied to H-B by using hydrogen molecules, which result from the RDF decomposition in the absence of stoichiometric oxygen in the gasifiers. Finally, the vitality of the system could be guaranteed if the amount of hydrogen generated from the RDF can at least cover the NH_3 fertilizers production for the food industry of the populations that generate the refuses.

The here presented *ex-ante* analysis aims to shed light on the feasibility of employing RDF as a feed for the gasification process to produce the H_2 input for the H-B process. For this, gasification and H-B plants were first simulated using process simulation software to evaluate the quantity of RDF necessary to provide H_2 to a medium size H-B plant (1,500 ton NH_3/day). At the same time the direct energy balance, chemicals consumption and yields of the main equipment were estimated. Then, the indirect energy and materials footprints of the total plant were calculated to complete the inventory analysis for the LCA. The results are analysed in terms of the functional unit (kg NH_3) and compared to the traditional H-B process. Finally, the virtual plant studied was contextualized in relation to urban centres capable of providing sufficient RDF feedstock and the *per capita* consumption of ammonia used for fertilizers production.

2 METHODOLOGY

2.1 The case study: simulated H-B plant

The approach followed in the present work comprehended the simulation of a virtual ammonia-producing plant, with a capacity of 1,500.0 ton/day. The case study considered a *per capita* municipal solid waste (MSW) generation of 500.0 kg/(y·p). The flow of MSW was considered to go 50 % through the separate collection (the complement as unsorted MSW). The unsorted MSW undergoes a sorting step for the removal of metals and ferrous materials, and then two main fractions are obtained: the RDF (containing combustible components) and the OFMSW (Organic Fraction of Municipal Solid Waste cut, with high humidity content). RDF is the fraction mainly containing paper, cardboard, wood and hard and soft plastics (and a minor fraction of organic compounds) [5]. Simplified block diagrams of the conventional H-B process (Figure 1A) and this proposed hypothetical plant (Figure 1B) are shown below**.** The latter plant is simulated using the *AspenPlus® v8.8* software, considering the key sections required for the process: the air separation unit (ASU), the biomass gasification (BG) step, the syngas purification (SP), and the H-B reactor. A brief overview of the simulation features for each of these units is provided below.

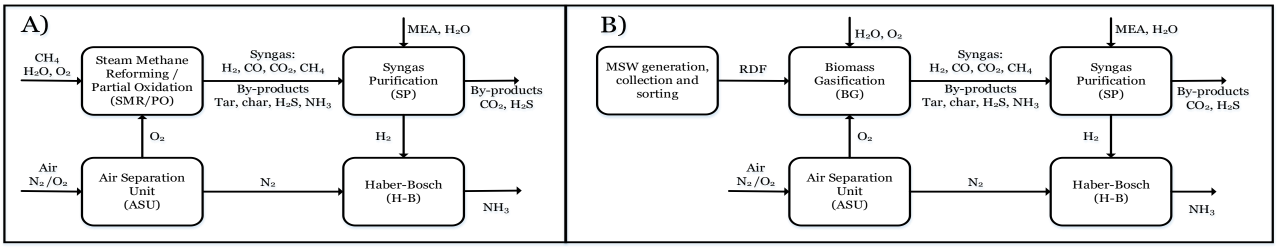

Figure 1. Block diagrams of: A) conventional H-B plant and B) the proposed simulated H-B plant, using gasification of Refuse Derived Fuel (RDF) as the feedstock to produce syngas.

Air Separation Unit (ASU)

The cryogenic air separation process was simulated to obtain (direct) reference energy consumption figures, and separation performance data. The ASU was fed with atmospheric air (i.e., 76,87 % *v/v* N_2, 20,62 % *v/v* O_2, 0,92 % *v/v* Ar, 1,56 % *v/v* H_2O, 0,03 % *v/v* CO_2) at 25 °C and it produced a high purity nitrogen stream and an enriched oxygen stream. The ASU simulation considered two distillation columns, one at high and another at low pressure (i.e., HPC=5.6 bar and LPC=1.2 bar). The thermodynamic method used for the ASU was *PENG-ROBINSON* and a multi-stage turbo compressor was set, with (cooling-water) intercoolers to supply the feed at the required pressure and temperature. Process conditions for both columns were optimized (i.e., feed tray, reflux ratio, product flow) during the simulation of the ASU, to define the best-performing conditions for obtaining high purity N_2 (O_2 < 10 ppm to avoid poisoning the H-B catalyst).

Biomass Gasification (BG)

The RDF was defined as a *non-conventional component* in *ASPEN* and reference proximate and ultimate compositions are provided, as shown in Table 1. The gasification reactor was simulated as a two-stage reactor (*RYIELD*): *i)* the RDF stream was first fed to a decomposer reactor (*DEC)* in which the complex biomass is decomposed following the provided elemental composition (and using the *HCOALGEN* and *DCOALIGT* methods for the calculation of the stream properties) and *ii)* a gasification reactor (BURNER) in which syngas was obtained. The operating conditions of the BG step were established, based on sensitivity analysis aimed at maximizing hydrogen production.

Table 1. Proximate and ultimate compositions assumed for the Refuse Derived Fuel (RDF)

Proximate Analysis [% wt]					*Ultimate Analysis [% wt]*						
MC	**DM**	**VM**	**FC**	**AF**	**C**	**H**	**N**	**Cl**	**S**	**O**	**AF**
23.3	76.7	66.7	6.9	10.0	52.1	7.5	1.3	0.8	0.4	27.9	10.0

MC: moisture content, DM: dry matter, VM: volatile matter, FC: fixed carbon, AF: ash fraction

For this, the equivalence ratio (0.0-0.7), the type of gasifying agent (oxygen output from the ASU, air and steam) and the operating temperatures (600-1200 °C) were varied within common ranges of these types of systems. Finally, since the gasification process produced also CO, an additional (catalytic) preferential oxidizer reactor (PROX) was considered as a downstream unit for the BG. The PROX unit used also a share of the enriched oxygen stream produced in the ASU for the oxidation of CO into CO_2, to minimize by-product formation and catalyst deactivation in the H-B unit. After the PROX unit, an H_2-enriched stream was obtained (to be fed to the syngas purification unit).

Syngas Purification (SP)

Compared to the traditional H_2 production for H-B through SMR or PO, the BG tends to generate a syngas stream that might contain higher impurities, since the feed to the reactor (RFD) is more heterogeneous and variable. Additionally, there is also an important fraction of CO_2 resulting from the BG and PROX reactors, as well as a minor fraction of H_2S. Hence, an absorption/stripping purification process to remove CO_2 and H_2S from the syngas was modelled. For this, two columns were simulated (i.e., ABSORBER and STRIPPER, under the AMINES properties package) using a monoethanolamide (MEA) solution (i.e., 67 % *w/w* H_2O, 33 % *w/w* MEA). The ABSORBER was set to operate at 8 bar and 40 °C, while the STRIPPER at 1.8 bar and 105 °C. Residual water content was also removed from the sweetened syngas by including in the simulation a flash separator. Finally, the direct energy consumption of the syngas purification unit, the required MEA flow, and potential CO_2 emissions were calculated from this simulation step.

Haber-Bosch Reactor (H-B)

For the correct simulation of the H-B process, the following elements are considered: *i)* compression and heating of the reactants up to the operating conditions, *ii)* cooling of the products, *iii)* separation of the ammonia from the other outputs (by-products and unreacted feed), *iv)* recycling of unreacted nitrogen and hydrogen and purging, *v)* thermal integration of key process streams. Both reactant feeds were first mixed and fed to the upstream compression stage (up to 150 bar, $\eta_{c=0.752}$) and then to the H-B reactor (using an RGIBBS reactor and the Peng-Robinson thermodynamic model). Fresh reactants were mixed with recycled unreacted compounds. Operating conditions for the reactor were set at 500 °C and 200 bar; the output was then cooled down to allow ammonia to condensate (c. -91 °C) and to separate it from unreacted gases.

2.2 The Life Cycle Assessment (LCA)

Using the results from the simulation step, the *ex ante* LCA was conducted. The aim was to shed light on the environmental footprint of the hypothesized process, to produce NH_3 using RDF gasification as the hydrogen source and compare the results to reported values of traditional H-B processes. The prospective inventory was compiled by estimating the quantity of chemicals, materials and energy, which would be required for such a plant over its life cycle. The functional unit for the analysis was the produced kg of NH_3 and the estimated lifetime of the plant is 25 years. The *ASPEN* simulation served for the estimation of the direct energy expenses of each unit, while the indirect energy embedded in materials was estimated based on the required equipment for each unit (ASU, BG, SP and H-B) using basic sizing criteria (e.g., retention times, required flows and equipment thickness) as well as commercial brochures for the most common devices. The environmental impacts comparison between NH_3 produced from natural gas and the gasification of RDF was conducted using the cumulative energy demand (CED) (MJ/kg NH_3) and the global warming potential (GWP 100y) (kg $CO2_{eq}$/kg NH_3). The approach is cradle-to-gate for ammonia production, and it considers the direct energy and chemicals consumption, as well as the indirect materials and energy over the lifecycle (including the share of construction and decommissioning of the plant). Inventory items were analysed using (mainly) *Ecoinvent v.3.8* databases and the *SimaPro 9.4.0.2* software, using when possible GLO (Global) reference processes.

3 RESULTS AND DISCUSSION

3.1 The RDF processing, the BG unit and syngas purification

One of the advantages of RDF as feedstock for hydrogen production is that RDF is an APER. That is, its availability is linked to anthropogenic activities. As mentioned above, it is mainly composed of the inorganic fraction of MSW (plastics, wood and mixed scraped waste). Even when the inorganic fraction of RDF varies over time (due to substitution by bio-based materials, or due to decreased waste generation), its substitutes could probably be an adequate feed to the BG unit (i.e., if its proximal and ultimate composition is similar to the RDF). Today, MSW collection is a fundamental societal service whose interconnection with the food production chain could make the way in which fertilizers' needs are met more robust (in addition to reducing dependence on fossil fuels).

Sensitivity analyses served for determining the best-operating conditions for the BG unit. First, the effect of temperature and equivalence ratio on the flow and fraction of produced hydrogen in the syngas was studied. It can be seen in Figure 2A that ERs < 0.2 favour both studied parameters, with max. 62% *v/v* H_2 and flow rates of 60-80 ton/d for the analysed temperatures (800-1200 °C). However, to evaluate if H_2 production could be increased, steam as a gasifying agent was also tested. In this case (Figure 2B), H_2 compositions in the syngas lied between 30-55% *v/v*, and flows > 85 ton/d (for conditions S/B > 1) were obtained. The chosen parameters for the simulation were a mixed gasifying agent (oxygen from the ASU and steam) at an ER of 0.1 and an S/B ratio of 2.0 and a temperature of 1000 °C (similar to literature references [6]). The produced syngas was then dehydrated and oxidised at low temperatures (in the PrOX) to reduce the content of CO. The final composition (% *v/v*) of the syngas resulting from the BG step is: H_2 67.04, H_2O 0.10, CO_2 31.88, CO 0.01, N_2 0.55, Ar 0.27, H_2S 0.07 and Cl_2 0.08.

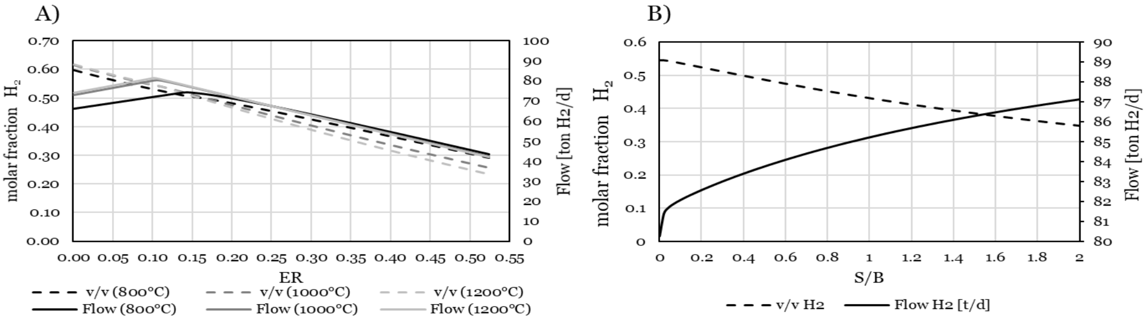

Figure 2. Sensitivity analyses for the BG unit: A) the effect of temperature and equivalence ratio (ER) and B) the effect of the steam-to-biomass ratio (S/B) on the hydrogen composition and flow

3.2 The ASU, the SP and the H-B units

The ASU simulation followed the approach reported in section 2.1, yielding two main outputs, the pure N_2 stream (fed to the H-B), and an enriched O_2 stream. This latter was employed as a gasifying agent in the BG unit (section 3.1) and as an oxidizing agent for the PrOX unit after the BG prior to the SP unit, which was mainly aimed at removing the acid species (CO_2 and H_2S). The simulation of the SP resulted in a H_2 stream composed by (*v/v*) 98.67 % H_2, 0.81 % N_2, 0.40 % Ar and 0.12 % Cl_2. This stream was then mixed with the pure N_2 (from the ASU) and fed to the H-B in the simulation to produce NH_3.

3.3 LCA results

Direct energy expenses

For the analysis of the environmental footprint of the simulated process, the consumption of operational direct materials and energy was first evaluated. In particular, the required auxiliary services of heat, cold and power and the possible reduction due to energy integration between streams are reported (Table 2).

Table 2. Direct utilities and chemicals specific consumption considered for the LCA

Utilities and chemicals	*Units*	*ASU*	*BG*	*SP*	*H-B*
Heat	[$MJ_{th}/kg\ NH_3$]	0.356	28.397	19.125	5.349
Heat integrated	[$MJ_{th}/kg\ NH_3$]	0.018	2.289	0.956	0.908
Cold	[$MJ_{th}/kg\ NH_3$]	0.128	3.059	2.260	2.638
Cold integrated	[$MJ_{th}/kg\ NH_3$]	0.046	0.153	0.113	0.559
Electricity	[$MJ_{el}/kg\ NH_3$]	0.747	0.000	3.625	11.821
Water	[$kg\ H_2O/kg\ NH_3$]	0.000	2.229	1.681	0.000
MEA	[$kg\ MEA/kg\ NH_3$]	0.000	0.000	0.013	0.000

After the assessment of the direct consumption of chemicals and energy, the indirect expenses were quantified. For this, it was assumed that the auxiliary services were locally provided, through natural gas fueled co-generation (i.e., η_{el}=0.45, η_{th}=0.41 and COP=3.45 for cold services). Chemicals consumption was aggregated (using the figures provided in Table 2); and the amounts of steel, concrete and catalysts required for all units including the co-generator are reported in Table 3. The calculated environmental and energy impacts are reported at the end of Table 3, as well as the reference indicators of H-B processes, which use either SMR or PO (instead of BG). It can be noted that the CED of the studied virtual plant is 20-30% lower with respect to these reference processes. In terms of GWP, a similar footprint to the SMR is obtained (7 % higher), and 60% lower compared to the PO process. These emissions are composed of 20% indirect emissions (outside the analytical boundaries), while roughly 80% can be considered as direct emissions (derived from the CHP and the stripping process in the SP unit). Hence, these two process streams containing significant fractions of CO_2 could be suitable for carbon storage or utilization applications. Although the prospective LCA inventory may underestimate some inputs, the obtained values provide first insights into the environmental feasibility of the proposed process. These results can be of interest to compare the proposed plant to other non-conventional ammonia production processes [7]. Considering the per capita waste generation (approx. 500 kg/(p·y)) and an undifferentiated collection percentage of 50%

(from which the RDF can be derived at 30%), the waste from an urban centre of approximately 9 million people could generate sufficient RDF to feed the studied plant.

Table 3. Indirect materials and chemicals expenses considered for the LCA

Materials & Chemicals	***Amount [kg/kg NH_3]***	***CED [MJ/kg]***	***GWP [$CO_{2\,eq}$/kg]***	***Reference process***
RDF	1.27E+00	1.38E-01	9.53E-03	Municipal waste collection service by 21 metric ton lorry {GLO}| market for | Cut-off, S (50 km distance)
Natural Gas	4.09E-01	6.38E+01	6.04E-01	Natural gas E (Gas to user) - Industry data
ProcessWater	3.91E+00	1.26E-02	7.58E-04	Tap water {GLO}| market group for | Cut-off, S
Cooling water	9.67E+01	6.71E-03	5.52E-04	Wastewater, average {RoW}| treatment of, capacity 1E9l/year | Cut-off, S
Concrete	7.36E-06	8.33E-01	1.18E-01	Concrete, normal {GLO}| market group for concrete, normal | Cut-off, S
Steel	7.60E-05	7.13E+01	4.87E+00	Steel, chromium steel 18/8 {GLO}| market for | Cut-off, S
MEA	1.30E-02	7.30E+01	3.17E+00	Monoethanolamine {GLO}| market for | Cut-off, S
H-B catalysts (Fe3O4/Al2O3)	9.00E-06	1.14E+01	1.12E+00	Magnetite {GLO}| production | Cut-off, S Sodium aluminate, powder {GLO}| production | Cut-off, S
WGS catalysts (Pt5%/ZrO2)	5.00E-06	1.12E+06	6.85E+04	Metal catalyst for catalytic converter {GLO}| platinum to generic market for metal catalyst for catalytic converter | Cut-off, S
PROX catalysts (Rh 5%/Al2O3)	1.00E-06	1.30E+06	7.93E+04	Metal catalyst for catalytic conver3ter {GLO}| rhodium to generic market for metal catalyst for catalytic converter | Cut-off, S
Ammonia – BG (from RDF) [1]	**1**	**3.48E+01**	**2.65E+00**	[1] **This study**
Ammonia - SMR	**1**	**4.49E+01**	**2.47E+00**	**Ammonia, anhydrous, liquid {RER w/o RU}| ammonia production, steam reforming, liquid | Cut-off, S**
Ammonia - PO	**1**	**4.28E+01**	**4.61E+00**	**Ammonia, anhydrous, liquid {RoW}| ammonia production, partial oxidation, liquid | Cut-off, S**

Moreover, the production size of this plant could even cover the fertilizer needs of a population of around 30 million people (considering 15.2 kg $NH_3/(p \cdot y)$). Although the studied plant is of a particular size (and hence the obtained values are more accurate at this scale), the results also suggest that allocating around 30-35% of the RDF to produce ammonia can cover the fertilizer needs of the same population that generates them.

4 CONCLUSIONS

The transformation of the ammonia production chain (with important consequences in the food supply chain) is a challenge that must be urgently addressed in the context of sustainable production patterns. The inclusion of RDF as an input for the H-B processes could bring benefits in terms of reducing the energy footprint (up to 20-30% CED reduction), with the advantage of also being a widely available anthropogenic resource. Additionally, the capture of (direct) emissions streams of the proposed process, especially in the purification of the syngas, could be advantageous to lower the CO_2 emissions (and consequently the GWP of up to -70%). Preliminary analysis of the use of RDF to produce H_2 for the H-B process suggests that a population could use the generated RDF to self-satisfy its fertilizer needs and generate a surplus for other populations. Further analyses are required to compare RDF to other potential biomasses that could be fed to the BG process, as well as to compare with other hydrogen production methods for H-B plants.

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