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Modeling and Simulation of a Novel Sustainable Ammonia Production Process From Food Waste and Brown Water

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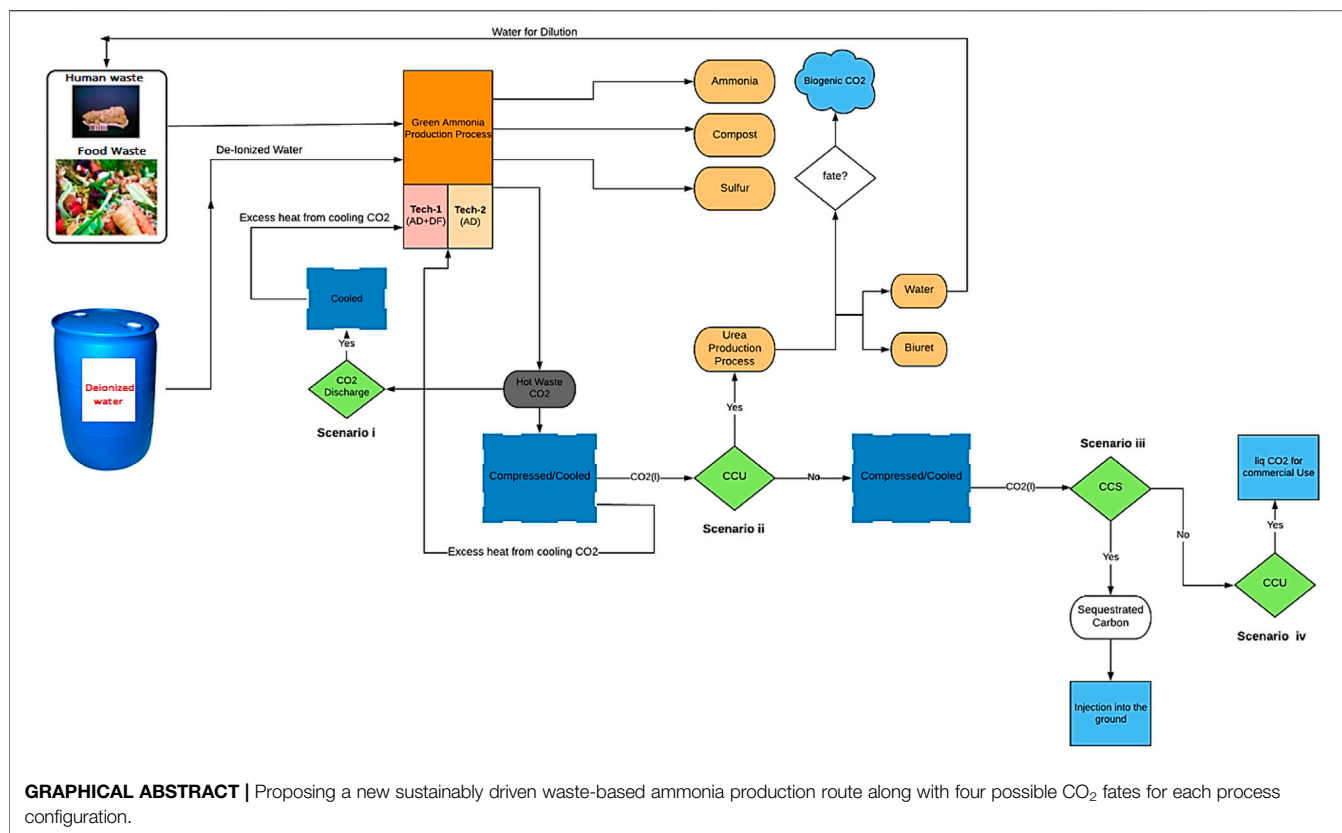
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Global demand for both clean energy carriers and agricultural nutrients continues to grow rapidly, alongside increasing quantities of waste globally, interlinked challenges that may be addressed with interlinked solutions. We report on the potential efficiency and Greenhouse Gas (GHG) intensity of several configurations of a new, sustainability-driven ammonia (NH₃) production processes to determine whether a waste-based process designed first around carbon dioxide (CO₂) capture can compete with other available NH₃ technologies. This is assessed via different scenarios: Two hydrogen generating options are paired with four CO₂ fates. For either an anaerobic digestion-centered process or a two-stage dark fermentation coupled with anaerobic digestion process, the resultant CO₂ may be captured and injected, sold to the marketplace, released directly in the atmosphere, or converted to urea in order to produce a green substitute for synthetic NH₃. Modeled yields range from 47 t NH₃ when the resultant CO₂ is released or captured, or 3.8 t NH₃ and 76.5 t urea when the system is designed to produce no unutilized CO₂. Among the technologies assessed, NH₃ production where CO₂ is captured for anaerobic digestion-only is the most efficient for GHG emissions and water consumption, while the two-stage requires less energy on a fertilizer-N basis. GHG emissions for anaerobic digestion-only are approximately 8% lower than the two-stage. The best of the proposed technology configurations consumes about 41% less energy than water electrolysis coupled with Haber-Bosch and approximately 27% lower energy than Steam Methane Reforming (SMR) coupled with Haber-Bosch per kg NH₃.

Keywords: green hydrogen, green ammonia, waste utilization, carbon capture and sequestration (CCS), carbon capture and utilization (CCU), dark fermentation, anaerobic digestion



INTRODUCTION

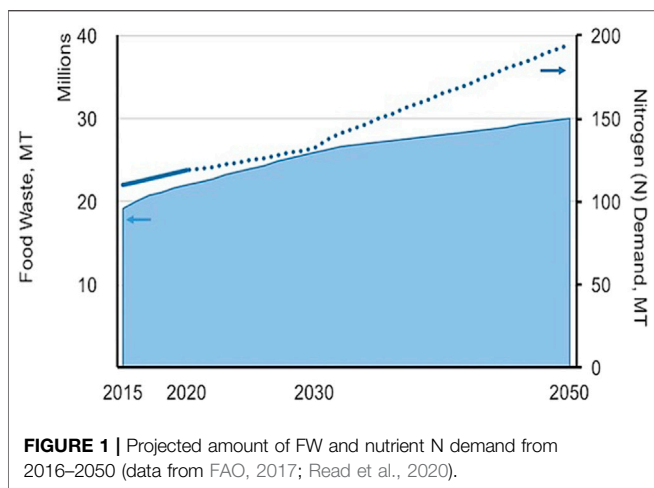
Ammonia is the second most produced chemical compound globally (146 million tonnes reported in 2016) after sulfuric acid (H₂SO₄), with fossil fuels being the main feedstock for its production (CICE, 2016; Soloveichik, 2017). Due to the important role of NH₃ as a fertilizer in the agricultural industry and its promising prospects as an energy carrier, many studies have recently attempted to find the most environmentally benign, energy efficient, and economically viable production process for NH₃ synthesis.

Ammonia production technologies currently in use require either a steady supply of deionized water in high volumes to operate and/or result in high CO₂ production. The most commonly utilized NH₃ production method is the Haber-Bosch process. The downside of this technology is high GHG emissions, surpassing 2.16 kg CO₂ eq/kg NH₃ and high energy consumption. For the production of 1 t of NH₃ through SMR coupled with Haber-Bosch, 0.66 t H₂O is utilized and 9–10 t CO₂ eq is produced, over its full life cycle (Parkinson et al., 2018). For producing 1 t of NH₃ through water electrolysis 1.6 t H₂O is consumed (Will and Lukas, 2018). In order to replace the existing high impact NH₃ technologies currently in use, a new sustainability-driven waste-based technology that is competitive for carbon emissions, water use, and power use has been designed around carbon capture/utilization. The design strategy was based on optimizing production while mitigating and valorizing waste and reducing emissions.

Bioconversion of wastes such as food and Human Waste (HW) into important chemical compounds such as NH₃ and urea

(CH₄N₂O) offers, to some extent, a new resource recovery alternative for fossil-based chemicals. The overabundance of these waste streams poses significant threats for GHG control. Uncontrolled decomposition of organic waste in landfills, if left untreated, leads to the discharge of methane (CH₄), a potent GHG trapping approximately 85 times more heat than CO₂ through the first 20 years from when it is discharged. Methane accounts for 20% of the global warming that is currently being experienced (IPCC, 2013; MIT News, 2017). In 2015, the U.S. Environmental Protection Agency (USEPA) reported that the GHG emissions resulting from waste landfills was 115.7 Mt of CO₂ eq (Lee et al., 2017). While in the same year, the UK government reported GHG emissions of waste disposed of in landfills was approximately 18 Mt of CO₂ eq (BEIS, 2017). As shown in **Figure 1**, the increase in the amount of Food Waste (FW) keeps pace with nutrient N demand and this trend is projected to continue. FW has increased by 40% over the course of the 34 years; while the nutrient N demand has increased approximately 49% over the same time period.

A particular benefit of a waste-based process such as the green NH₃ technology developed here is the potential to avoid CH₄ that could have been released from uncontained degradation of waste. The potential GHG mitigation benefit with the technology depending on how much of the processed waste was diverted from uncontained degradation is illustrated in **Figure 2**. About 74 kg of CH₄ is released with open degradation of a tonne of food waste comprised of about half vegetable and a quarter meat wastes (Quantis, 2019). If 10% of each tonne of processed food waste resulted from diversion, that would avoid releasing about 7.4 kg CH₄



or 200 kg CO₂ eq; at 50% rescued material that grows to nearly 1 kg of CO₂ eq/kg FW processed and at 90%, almost 1.85 kg CO₂ eq/kg FW would be prevented from directly reaching the atmosphere.

As the population grows the need for more food production leads to the demand for more fertilizers, which results in higher food demands and more HW generation. The projected global population along with the total nitrous oxide (N₂O) and CH₄ emissions from human sewage and domestic wastewater from 1990–2030 are shown in **Figure 3**. The amount of HW necessarily increases proportionally with the global population. The amount of HW has increased by approximately 51% over the course of 40 years, while the population growth is approximately 47% over the same time period.

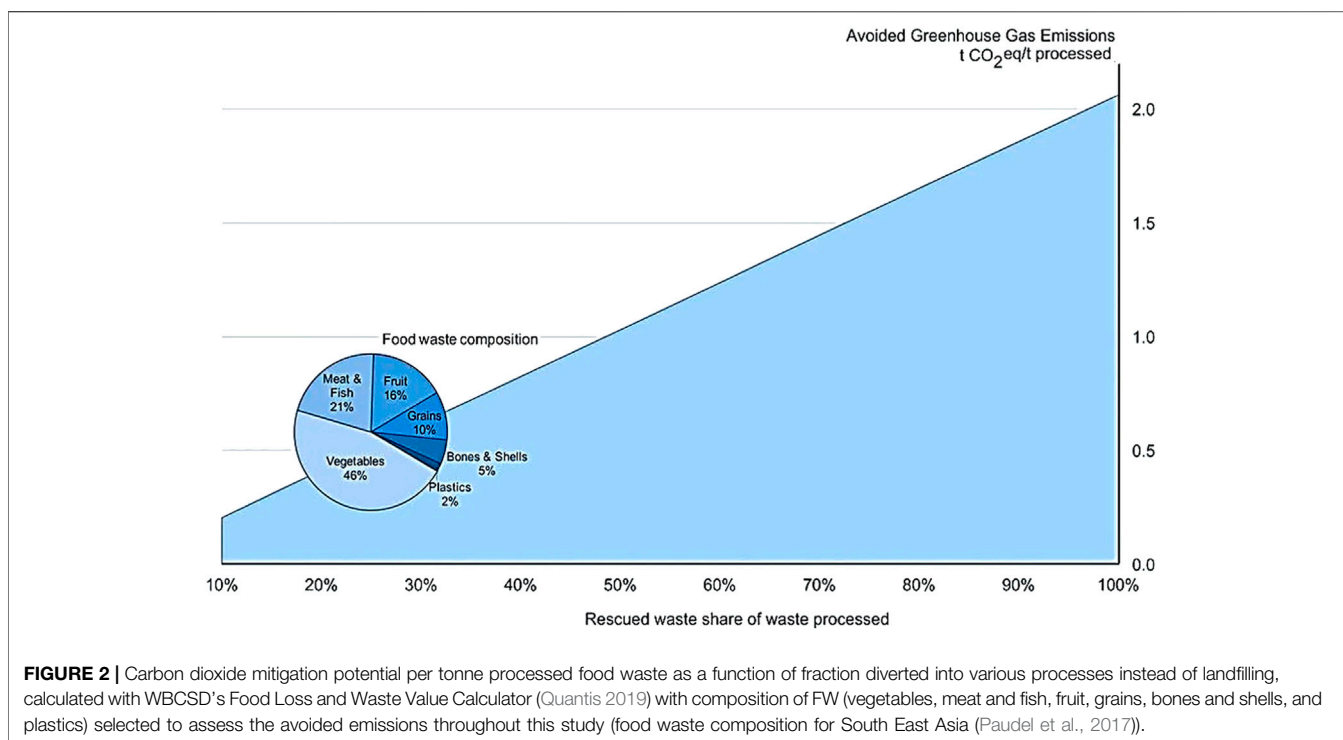
Both FW and HW are rich in readily biodegradable organic matter, therefore they decompose rapidly, and, in addition to their contributions to global GHG balances, may potentially result in other serious environmental implications if not properly managed. FW and HW are readily biodegradable in the environment which indicates their potential in industrial bioconversion. As a result, designing a sustainable waste management process which reduces the GHG emissions such as the utilization of AD will be an effective pathway to target the replacement of higher impact, fossil-based chemicals.

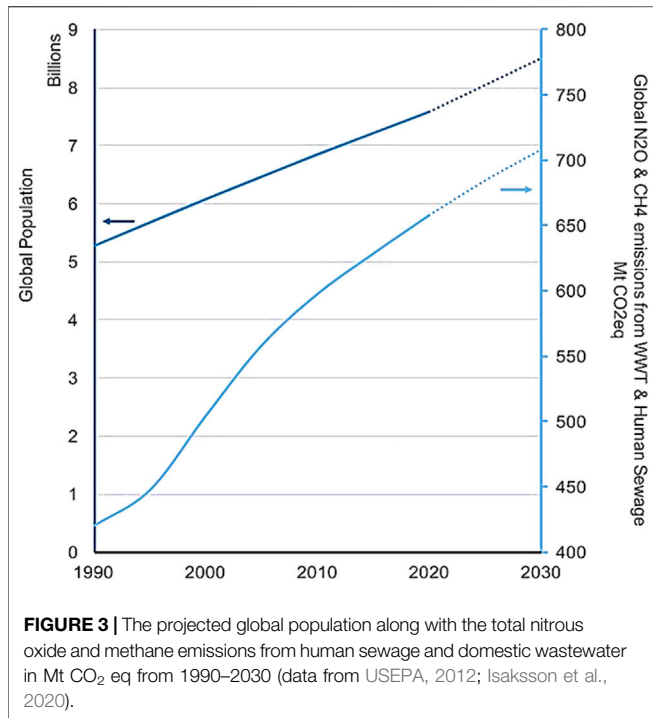
This work describes the results of a study implementing this design strategy, developing a green and viable NH₃/urea process in order to reduce waste and prevent the release of additional CO₂ (from primarily biogenic sources) into the atmosphere.

INTRODUCING A SUSTAINABLE WASTE MANAGEMENT APPROACH FOR AMMONIA PRODUCTION

Simplified Process Description

In the processes designed and modeled, waste (FW and human faeces) is fed into a two-stage bioreactor, resulting in the production of biogas. Hydrogen (H₂) is separated from the gas mixture through a series of operating stages (membranes) and reacts with nitrogen (N₂) derived from the migration of air through a membrane, in order to produce NH₃. The CO₂ recovered along with the NH₃ can be used for urea production (a simplified process flow diagram is shown in **Figure 4**). This process is compared to the case where the CO₂ is not





captured/utilized, as well as to cases where the CO₂ is captured, compressed, liquefied, and either injected into the ground (Carbon Capture and Sequestration (CCS)) or assumed to enter the commodity liquid CO₂ market (base Carbon Capture and Utilization (CCU)) (scenarios i–iv).

Plant Components Incorporated into the Modeled Process

The core component of this proposed NH₃ production process is waste processing, utilizing either a two-stage Dark Fermentation Bioreactor (DFB) coupled with an Anaerobic Digestion Bioreactor (ADB) or the latter alone. For each component involved in this proposed process a model section will be presented. The modeling components are comprised of: Dark Fermentation (DF), Anaerobic Digestion (AD), Micro Filtration Index (MFI), Electrochemical Membrane Separation (EMS), Ion Transport Membrane (ITM), CH₄ assisted-Solid Oxide Fuel Electrolysis Cell (SOFEC), Solid State Ammonia Synthesis (SSAS), Solid Oxide Fuel Cell (SOFC), and a urea production plant. The description of the plant components is shown in **Table 1**. A critical analysis of why EMS, ITM, CH₄ assisted-SOFEC, SSAS, and SOFC have been chosen as process components is presented in the study conducted by (Ghavam et al., 2021).

The main components for NH₃ production are H₂ and N₂. Hydrogen is the output from MFI and CH₄ assisted-SOFEC, while N₂ comes from the ITM. These two components enter the SSAS for NH₃ synthesis.

Incorporating Urea into the Modeled Process

A urea production process is an effective pathway for harnessing the CO₂ that might otherwise have been directly released into the atmosphere. Eventually this CO₂ that is utilized for urea production will be discharged, as a result, incorporating a urea plant cannot solve the carbon emission problem but merely delays it while getting useful services (in this case offsetting

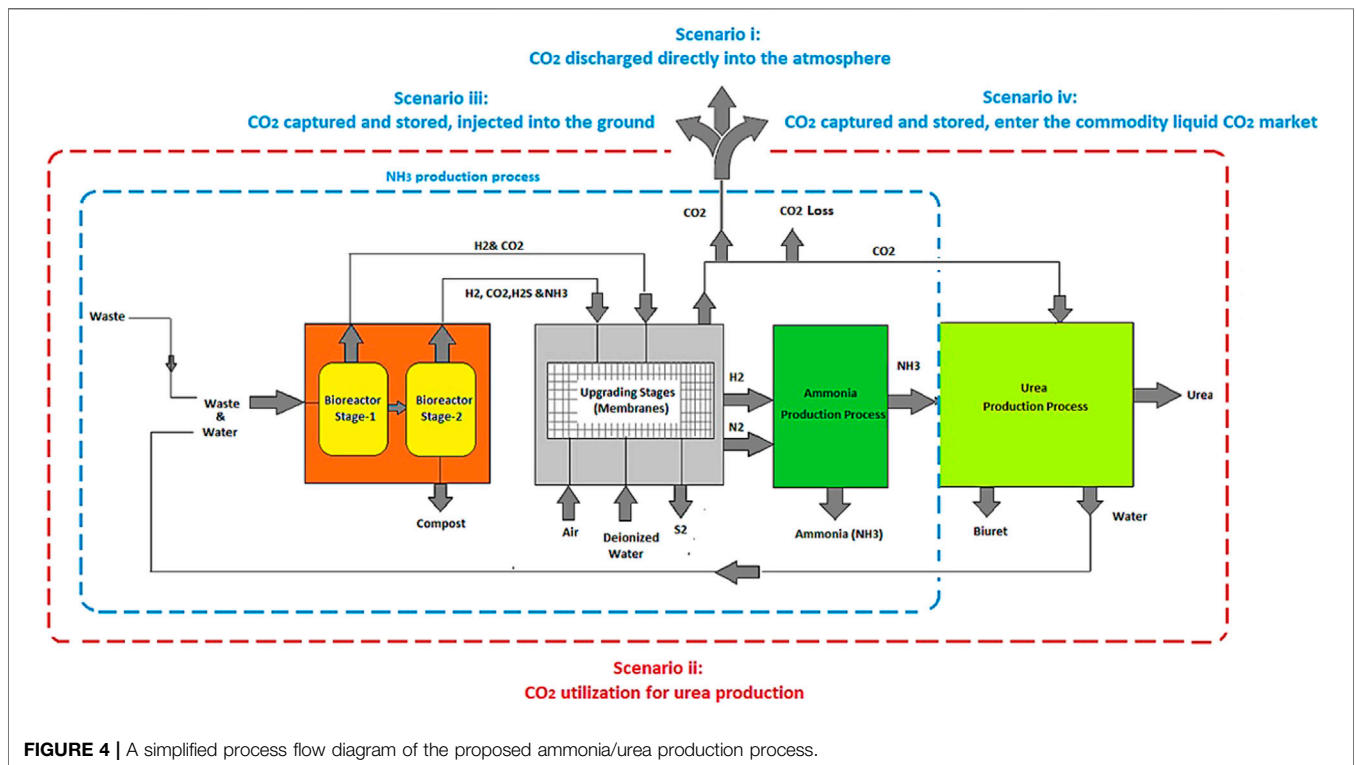


TABLE 1 | Description of the plant components.

Processing unit	Description	Processing unit	Description
Dark Fermentation Bioreactor (DFB)	DF is the decomposition of carbohydrate rich substrates in the absence of light and oxygen (O_2) to produce H_2 and CO_2	Solid State Ammonia Synthesis (SSAS)	SSAS produces NH_3 through an electrochemical conversion process in a proton-conducting solid electrolyte cell from gaseous H_2 and N_2
Anaerobic Digestion Bioreactor (ADB)	AD is a continuous biochemical process in which complex substrates are decomposed and digested via microbial actions and converted into biogas in the absence of O_2 (Pengyu et al., 2017). This transformation takes place through four main steps: Hydrolysis, acidogenesis, acetogenesis, and methanogenesis	CH_4 assisted-Solid Oxide Fuel Electrolysis Cell (SOFECE)	In the Solid Oxide Fuel Electrolyzer Cell, CH_4 is added to the anode side of the electrolyzer, the decomposition potential (voltage) of H_2O is decreased, and this results in lower energy usage and a higher conversion ratio of electricity for H_2 production
Membrane Filtration Index (MFI)	An electrochemical zeolite-based membrane known as Membrane Filtration Index has been incorporated into the system for the separation of H_2 from the H_2 and CO_2 gas mixture	Solid Oxide Fuel Cell (SOFC)	SOFC is an electrochemical conversion device that generates electricity via electrochemical conversion of fuel gas mainly H_2 at high temperatures of 700–1,000°C
Ion transport Membrane (ITM)	This membrane is a solid inorganic oxide ceramic material that transports O_2 ions at high pressures (10–30 bar) and temperatures (800–900°C)	Urea Production process	Urea is the result of the dehydration of carbamate. Carbamate is the intermediate product resulting from the reaction between NH_3 and CO_2
Electrochemical Membrane Separation (EMS)	Electrochemical Membrane Separation converts H_2S into H_2 and S_2 in the presence of N_2 used as the sweep gas		

the production of conventional urea, which uses significant fossil resources and results in significant life cycle GHGs). From a low carbon design point of view, which is the aim of this study, offsetting urea production has a greater environmental benefit than releasing the CO_2 directly from the process on a life cycle basis, despite the increase in energy demand. There is a side unwanted reaction during urea synthesis which produces biuret and thus lowers production yields. In order to prevent high biuret formation, a vacuum evaporator is utilized followed by a cooling system. Our aim is to optimize the process for maximum urea purity and to minimize biuret formation.

Significance of Membrane Incorporation in the Modeled Process

Our process is designed around membranes to address the challenges in necessary gas upgrading and purification, reduce the need for external sources of both chemicals and water, while minimizing the physical footprint of the processing units. To obtain the necessary purity level from biogas resulting from waste (food and Brown Water (BW)), the CH_4 stream needs to be upgraded to separate impurities such as CO_2 , hydrogen sulfide (H_2S) and traces of other compounds. The cost of the available technologies for upgrading biogas such as water scrubbing, Pressure Swing Adsorption (PSA), chemical absorption, cryogenic distillation, amine scrubber, etc. are comparatively high due to energy, water, chemical usage, and large physical footprint, and/or the combination of these factors (Fouladvand, 2016).

In place of processing units, such as an amine scrubber, with large physical footprint, an MFI membrane is used for the separation of H_2 and CO_2 from DF. For example, a single MFI module has the same throughput/output as a large, expensive and energy-intensive amine scrubber (Korelskiy et al., 2015).

This proposed process is also designed in order to increase the production of H_2 required for NH_3 synthesis through an environmentally benign, energy efficient, and cost-effective

manner. The design objectives have been achieved by utilizing CO_2 for urea synthesis, converting H_2S into sulfur (S_2) and H_2 via a membrane, converting the byproduct of the AD process into compost, utilizing renewable energy sources, and processing units with a low physical footprint for its operation. These processing units include certain types of membranes which reduce the production costs, such as ITM that is used for air separation (Sun et al., 2011).

MODELING APPROACH

The CO_2 emissions, water, and energy usage of this novel process for the production of NH_3 and urea from waste are analyzed computationally. A mathematical model to simulate the performance of the NH_3 production process from co-digestion of FW and BW is developed with mass, heat, and energy balance equations. This is implemented in the Visual Basic for Applications (VBA) platform embedded in the Microsoft excel program. The coupled mass and energy balance of this simulated model is assessed under various physical and chemical conditions (temperature, pressure, flow rates, specific heat capacity, porosity, tortuosity, flux, and permeability) and sizing of various processing units (thickness, area, number of tubes, cells, and modules). Assumptions and mathematical equations (mass transfer, energy transfer, thermodynamics, and kinetics) are determined for each processing unit. Since the entire process operates sequentially, the output and the performance of each component affects the input of the other components except for the bioreactors and ITM which are the initial processing units of this proposed process. The modeling framework is comprised of equation-oriented and sequential modular approaches. Through this approach, the components (processing units) are modeled using an equation-based approach in sequence, starting with the

feed stream and the entire process flow is treated as a set of mathematical equations which need to be solved simultaneously.

Although there are a variety of different thermodynamic simulators capable of modeling and optimizing different processes, none offer membranes among their standard process functions (Hoorfar et al., 2018). Commercial tools such as AspenPlus® do not have the option to model the membrane operation, and so it is necessary to program the membranes in Matlab, FORTRAN, VBA, or other programs. VBA enables us to expose direct optimization of membrane parameters.

Assumptions

The results presented here are based on the calculations for 20 tpd of input waste (FW and BW). Although other scaling can be evaluated with the implemented model, this figure is calculated to roughly show the size of a mid-size city such as Bakersfield, California. The main assumptions used in this study are listed in Table 2.

PROCESS DESCRIPTION

This proposed process is assessed through two configurations: Technology-1: Ammonia production through a two-stage DF coupled with AD and Technology-2: Ammonia production through AD-only. Each configuration has four possible CO₂ fates, Scenario i: CO₂ is discharged directly into the atmosphere, Scenario ii: (CCU) CO₂ is utilized for urea

production, Scenario iii: (CCS) CO₂ is compressed, liquefied, captured, and injected into the ground, and Scenario iv: (CCU) CO₂ is compressed, liquefied, captured, and enter the commodity liquid CO₂ market. All four scenarios are evaluated for two situations, one where SOFC is used, and one where it is not.

For both technologies (1 and 2) there are four sets of scenarios. The process configuration and computational model behavior change depending on scenario configuration. For scenarios i, ii, and iv all the produced CO₂ is compressed, cooled, and liquefied, then either discharged directly, captured and injected into the ground, or sold commercially. The CH₄ and H₂ resulting from AD/DF are upgraded into pure H₂ for NH₃ synthesis via multiple membrane types (MFI and CH₄ assisted-SOFEC). If the process does not utilize SOFC, the calculation ends at this point. When SOFC is incorporated into the process, the SSAS/SOFC hydrogen ratio is set, the SOFC calculations are conducted and the ITM, N₂ throughput is attenuated.

For scenario iii the entirety of the CO₂ produced is utilized for urea production and the upgraded H₂ is used for the production of the required NH₃ for urea synthesis. As with the other scenarios, the inclusion of SOFC determines the algorithm from here. If SOFC is incorporated into the process, the SOFC calculations are conducted and the SSAS/SOFC hydrogen ratio is set at this point. In the absence of the SOFC module, the N₂ from ITM is used without attenuation along with the excess H₂ for NH₃ synthesis rather than entering the SOFC module (detailed

TABLE 2 | List of assumptions used in this study.

Assumptions	
1- Process conditions	2- Operational conditions
<ul style="list-style-type: none"> - The process operates at a steady state condition -The flow in the membrane, fuel cell, and electrolyzer channels are considered as laminar (Reynolds number<2,300) -The composition of gases produced from the DFB is kept fixed at 58.7% H₂ and 41.3% CO₂ -The composition of biogas produced from the ADB is kept fixed at 81.5% CH₄, 18.47% CO₂, 0.010 %H₂S, and traces of other gases -The calculations are based on 10 t of FW and 10 t of BW and 25 t of water for dilution <p>1.1 Gas behaviors</p> <ul style="list-style-type: none"> -Air properties are constant-All gases involved in this production process (H₂, CO₂, CH₄, H₂S, NH₃, O₂, N₂, H₂O, CH₄N₂O, and NH₄CO₂NH₂) are considered as ideal gases ($PV = nRT$)-The gas mixtures are assumed well mixed at the molecular level with all components having the similar velocity, pressure, and temperature <p>1.2 Electrolytic assumptions</p> <ul style="list-style-type: none"> -Anode and cathode gases are not allowed to crossover-The electronic and ionic conductors are spread evenly and continuously throughout the electrodes-Total reactions take place in the CH₄ assisted-SOFEC and SOFC-99.9% of H₂ produced in this process is diverted into SSAS for NH₃ synthesis and the remainder goes to SOFC 	<ul style="list-style-type: none"> -The input gas flow enters this process at ambient temperature (25°C) and pressure (1 atm) -The operating pressures and temperatures are applicable to production processes of any size. As a result, once the size of the plant is selected, the flow rates through the plant can be estimated and the processing units can be properly sized, based on the waste composition, substrate concentration, and dilution rate -Solar radiation and effective temperature are neglected -Physical properties of digester, membrane, fuel cell, and electrolyzer material and the hot box material are uniform over the temperature range considered -The temperature of the substrate inside the bioreactor is constant -FW and BW will be fed to the process at temperatures lower than 4°C -The temperature within the membranes, fuel cell, and electrolyzer is distributed uniformly <p>3- Mass and heat transfer</p> <ul style="list-style-type: none"> -The internal mass transfer resistance between the gas and the membrane surface is negligible -Radiation heat transfer in this process is neglected -The internal heat generation in the two-stage DF and AD is negligible -Heat transfer is linear under steady state condition -The heat transfer coefficient is constant -Heat and mass flows are two-dimensional <p>4- Material loss</p> <ul style="list-style-type: none"> -There will be no leakage (material loss) from the process except for CH₄ from the AD, and CO₂ for urea production (NH₃ and N₂O losses are included only in the Life Cycle Assessment (LCA) study

information on the logical flow diagram is provided in **Supplementary Figure S1** in the supplementary materials).

Technology Pathways

Digestion efficiencies in co-digestion of FW and HW are generally higher when urine has been removed (Rajagopal

et al., 2014). This separation is done in order to increase the hydrolytic and acidogenic potential of co-digestion of FW and BW (Colón et al., 2015). Moreover, co-digestion of excreta along with other organic compounds enhances the process efficiencies that are prevented by excreta characteristics.

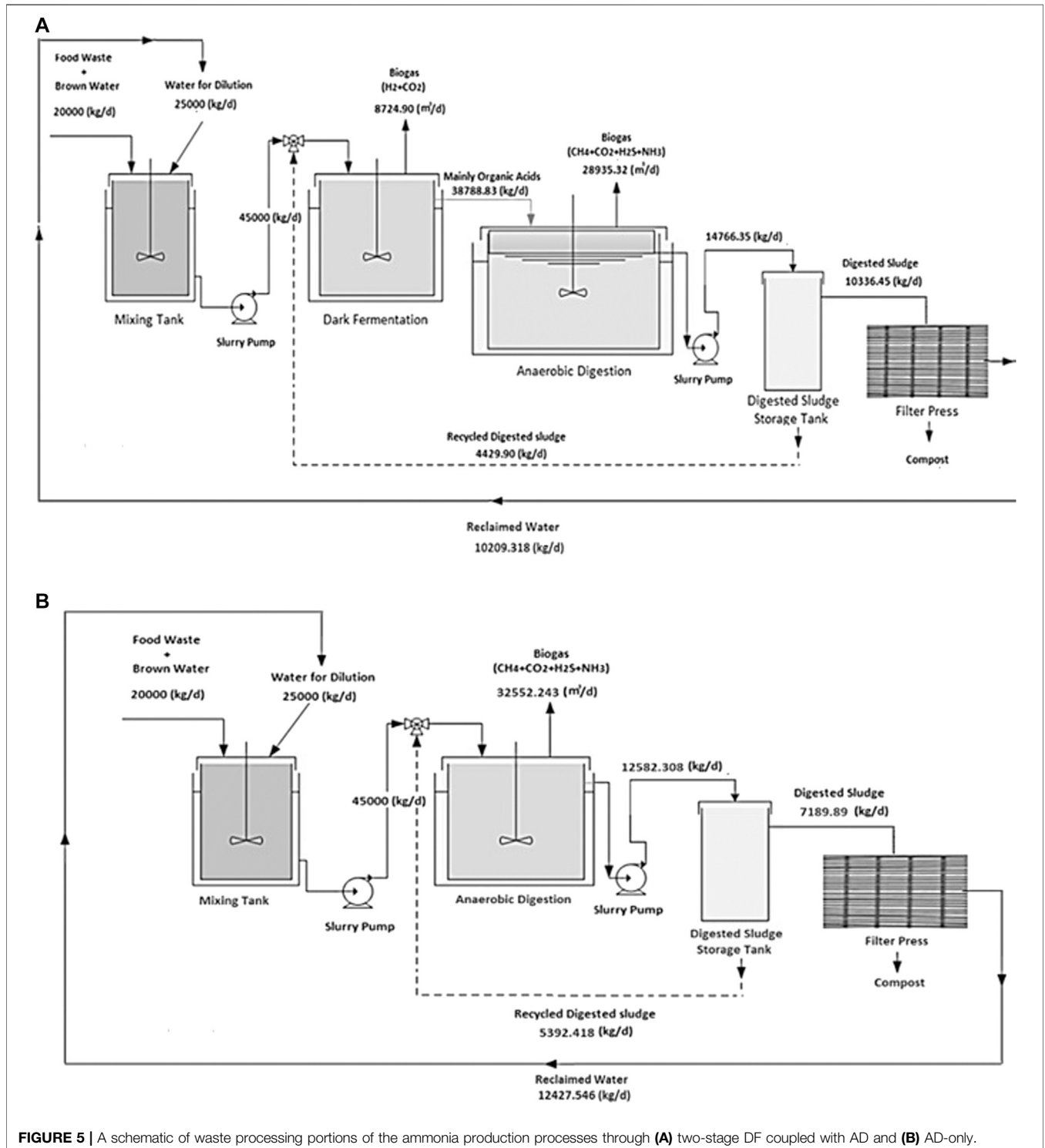


FIGURE 5 | A schematic of waste processing portions of the ammonia production processes through **(A)** two-stage DF coupled with AD and **(B)** AD-only.

General technology structures are shown in **Figure 5** (two-stage DF coupled with AD and AD-only process). Detailed technology descriptions are as follows.

Technology-1: Process Description of Ammonia Production Through a Two-Stage DF Coupled With AD

Hydrogen is separated from the H_2 and CO_2 gas mixture resulting from DFB by using an MFI. The separated H_2 provides the process with the required H_2 for NH_3 synthesis. The H_2 along with the needed N_2 resulting from the ITM, enters into the SSAS for NH_3 production. The organic acids which are the byproducts of DF are used as the substrate for AD in order to produce biogas which is comprised of CH_4 , CO_2 , and H_2S . Hydrogen sulfide must be removed from the biogas stream, which is accomplished here using an EMS system. This H_2S is converted into traces of H_2 and S_2 in the presence of a sweep gas (N_2) which is separated via ITM. The CO_2 along with CH_4 is introduced into a CH_4 assisted-SOFEC for H_2 production. Throughout this conversion, CO_2 is also separated from the H_2 stream in this electrolyzer. The H_2 then enters the SSAS, producing NH_3 which is required in the event where urea is required (only for scenario ii). A portion of the H_2 resulting from the CH_4 assisted-SOFEC is fed into a SOFC for generating heat, power, and steam for a part of the process. The NH_3 produced along with the CO_2 resulting from both MFI and CH_4 assisted-SOFEC enter the urea production process or the CO_2 is released directly into the atmosphere (scenario i), captured and injected into the ground (scenario iii) or assumed to enter the commodity liquid CO_2 market (scenario iv).

Technology-2: Process Description of Ammonia Production Through AD-Only

Biogas resulting from AD, needs to be upgraded in order to provide the H_2 required for NH_3 synthesis. The H_2 along with the needed N_2 resulting from the ITM, enters into the SSAS for NH_3 production. Hydrogen sulfide must be removed from the biogas stream, accomplished here using an EMS system. Hydrogen sulfide is converted into traces of H_2 and elemental S_2 in the presence of a sweep gas (N_2) which is separated via ITM. The CO_2 along with CH_4 are introduced into a CH_4 assisted-SOFEC for H_2 production. Throughout this conversion, CO_2 is also separated from the H_2 stream in the electrolyzer.

The H_2 along with the N_2 resulting from the separation of air in an ITM then enters the SSAS for producing NH_3 which in turn is required for urea production. A portion of the H_2 resulting from the CH_4 assisted-SOFEC is fed into a SOFC for generating heat, power, and steam for part of the process. The NH_3 produced along with the CO_2 resulting from CH_4 assisted-SOFEC enter the urea production process (only for scenario ii). The CO_2 resulting from the CH_4 assisted-SOFEC can be released directly into the atmosphere (scenario i), captured and injected into the ground (scenario iii) or assumed to enter the commodity liquid CO_2 market (scenario iv).

Energy Flows of Ammonia Production Through a Two-Stage DF Coupled With AD Process

The need for external energy sources is minimized and heat recovery is maximized when the heat integration method is

adopted. A heat balance model is presented in **Figures 6A,B**, for the proposed NH_3 production process. It illustrates the energy flow diagram utilizing heat integration and is divided into three parts **I**, **II**, **III**, and **IV** described below:

I: The process streams are cooled with the input air flow at $25^\circ C$ before entering the ITM and heated with recirculated steam at about $974.8^\circ C$ resulting from an external source of water at $920^\circ C$ which enters a compressor before being used for heating up the process at a steady state condition. The input air enters a compressor in order to reach the desired pressure in the range of 10–30 atm before entering the ITM. This air stream is heated from ambient temperature to $850^\circ C$. The separated O_2 is divided into two streams. The first stream enters the SOFC as one of the main feedstocks for its operation. The remaining O_2 passes through the SOFC hot box in order to enter the gas turbine system for power generation. The remaining N_2 is also divided into two streams. The first stream enters the relief valve to reach the pressure of 1 atm and is cooled down with air before entering the SSAS at $80^\circ C$. The second stream is also cooled down with air and lowered to $650^\circ C$ before entering the EMS used as the sweep gas.

II: FW and BW enter the process at $2^\circ C$ and are heated to $90^\circ C$ in a mixing tank along with the water for dilution. The mixture is then cooled with air to $55^\circ C$ in the DFB. The temperature is monitored through the entire process to increase the H_2 produced. The H_2 and CO_2 resulting from DF is compressed and passes through a refrigerator in order to reach a pressure of 20 atm and temperature of $-39^\circ C$ before entering the MFI. The sludge is then pumped into the ADB and is cooled to $35^\circ C$.

III: The biogas resulting from the ADB enters a heat exchanger to reach $650^\circ C$ before entering the EMS. The resulting $N_2 + S_2$ from the EMS enter a condenser in order to separate N_2 from S_2 . The separated N_2 will then be recirculated back into the EMS as the sweep gas. The remaining biogas stream from the EMS is then heated to $850^\circ C$ and the steam is cooled to the same temperature ($850^\circ C$) in order to enter CH_4 assisted-SOFEC for CO_2 separation and H_2 production. The resulting sludge from the ADB passes through a filter press to remove its moisture for producing compost. The reclaimed water is then recycled back into the mixing tank used as the diluted water.

IV: The separated CO_2 from both the MFI and CH_4 assisted-SOFEC are mixed, compressed, and cooled before entering the urea production process at 130 atm and $190^\circ C$ (**Figure 6A**). While for the process without urea production the separated CO_2 is compressed and cooled to 100 atm and $25.1^\circ C$ before being captured and injected into the ground or sold commercially (**Figure 6B**). Simultaneously the separated H_2 from both the MFI and CH_4 assisted-SOFEC is also mixed before entering the SSAS at $80^\circ C$ and 1 atm. The NH_3 produced from the SSAS is divided into two streams, the first stream enters the urea production process (**Figure 6A**). The second stream is compressed to 100 atm, cooled, and finally liquified (**Figure 6B**). The remaining H_2 is heated to $1,000^\circ C$ before entering the SOFC along with the O_2 from the ITM.

Energy Flows of Ammonia Production Through AD-Only Process
 A heat balance model for the proposed NH_3 production process demonstrating the energy flow diagram with heat integration is presented in Figures 7A,B. Part I and III have the same energy flows

as described above. While part II and IV have different configurations that are described below.

II: The waste stream enters the AD process at 37°C and is cooled to 35°C .

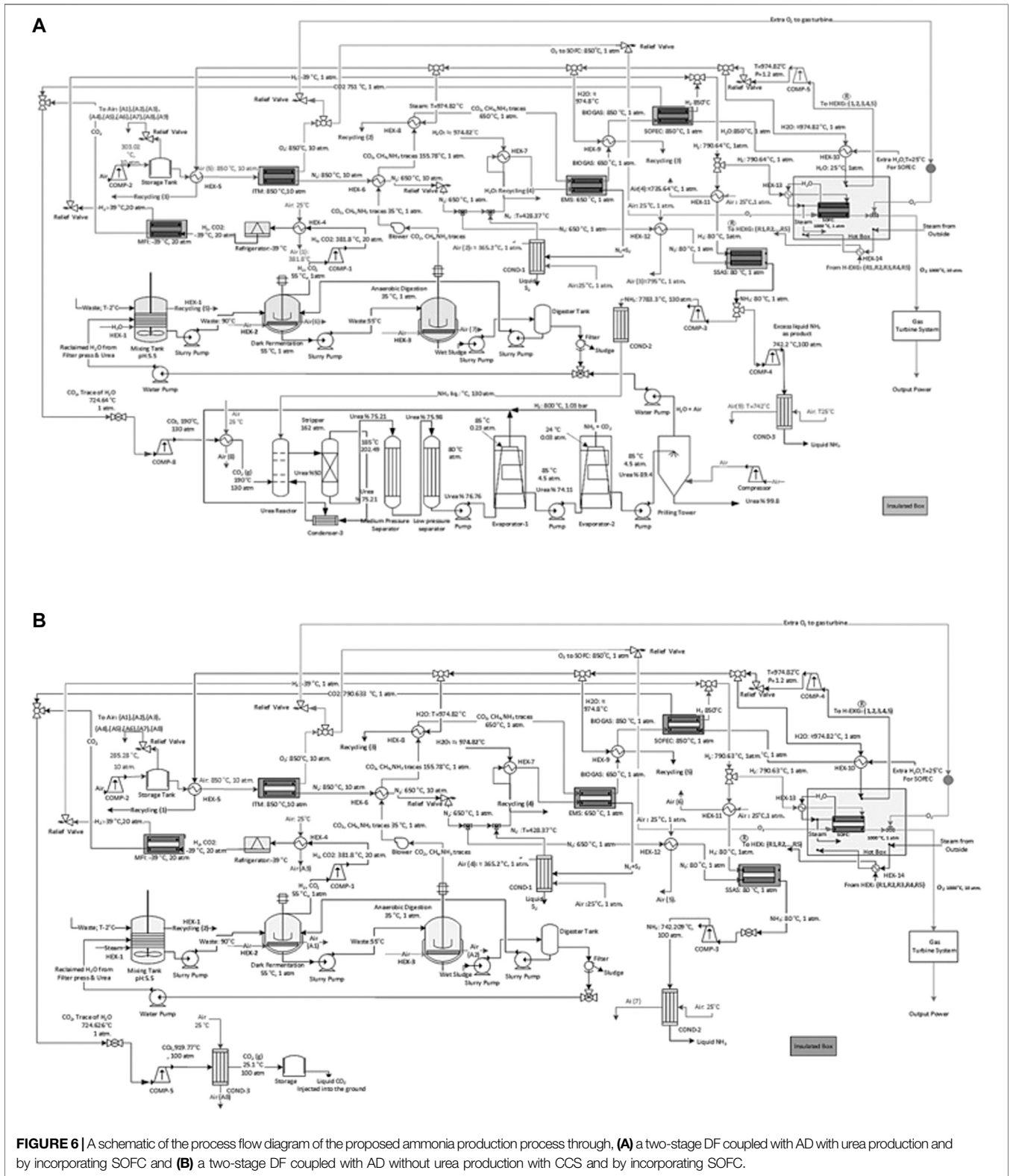
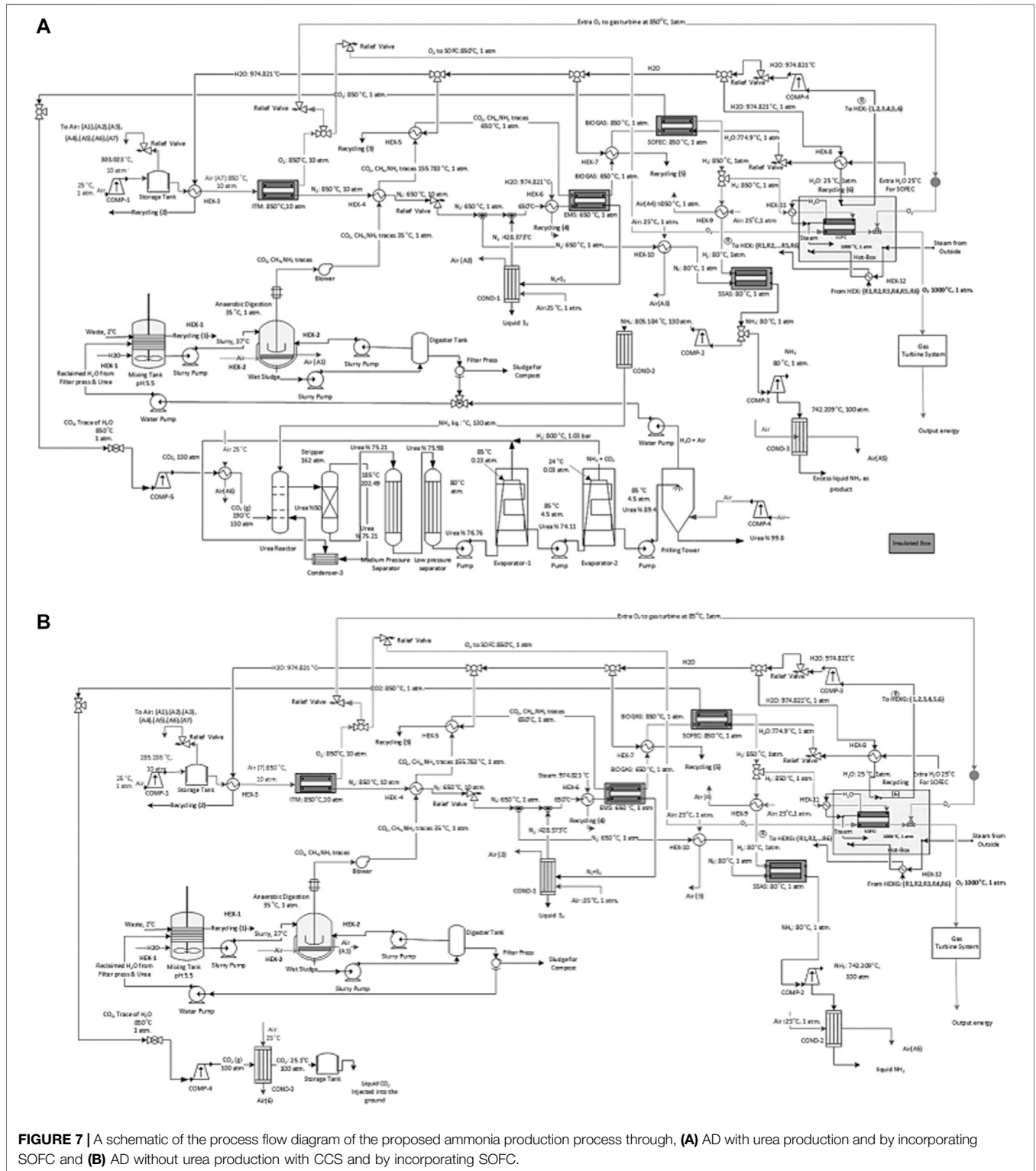


FIGURE 6 | A schematic of the process flow diagram of the proposed ammonia production process through, (A) a two-stage DF coupled with AD with urea production and by incorporating SOFC and (B) a two-stage DF coupled with AD without urea production with CCS and by incorporating SOFC.

IV: The separated CO₂ from the CH₄ assisted-SOPEC is mixed, compressed, and cooled before entering the urea production process at 130 atm and 190°C (**Figure 7A**). While for the process without urea production, the

separated CO₂ is compressed and cooled to 100 atm and 25.1°C before being captured and injected into the ground or sold commercially (**Figure 7B**). Simultaneously the separated H₂ from the CH₄ assisted-SOPEC reaches 80°C



before entering the SSAS. The NH_3 produced from the SSAS is divided into two streams, the first stream enters the urea production process (Figure 7A). The second stream is compressed to 100 atm, cooled, and finally liquified (Figure 7B). The remaining H_2 is heated to $1,000^\circ\text{C}$ before entering the SOFC along with the O_2 from the ITM.

RESULTS AND DISCUSSION

Technology-1: Modeling Results of Ammonia Production Through a Two-Stage DF Coupled with AD

Input and output waste compositions to and from DF and AD along with the gas/biogas specifications, yields, and the output composition of H_2 and CO_2 , CH_4 , H_2S , etc. are presented in the supplementary materials. The composition of the gas stream from DF is based on a H_2 content of 58.7%. If the ratio of H_2/CO_2 is $\geq 58.7\%$, the proposed production process produces both steam and power for SOFC, excess NH_3 , and urea. When the ratio of $\text{H}_2/\text{CO}_2 \leq 58.7\%$, the only output product is urea.

The biogas composition from AD is based on a CH_4 content of 81.5%, which is a controllable aspect of the proposed process. If the ratio of CH_4/CO_2 is $\geq 81.5\%$ the proposed production process produces both steam and power for SOFC, excess NH_3 , and urea. With the ratio of $\text{CH}_4/\text{CO}_2 \leq 81.5\%$ the only output product is urea.

Technology-2: Modeling Results of Ammonia Production Through AD-Only

Input and output waste compositions to and from AD along with the biogas specifications, yields, and the output composition of biogas are shown in supplementary materials.

The biogas composition from AD is based on a CH_4 content of 81.5%. If the ratio of $\text{CH}_4/\text{CO}_2 \geq 75\%$, the proposed production process produces both steam and power for SOFC, excess NH_3 , and urea. When the ratio of $\text{CH}_4/\text{CO}_2 \leq 75\%$ the only output product is urea.

Energy Balance Results

A. Energy Balance Results of Ammonia Production Through a Two-Stage DF Coupled with AD

The energy draw is driven primarily by the urea production process, which accounts for approximately 93% of the total energy consumed (Figure 8A). This is followed by the energy loss (6.04 MW), power required for supplying steam (5.2 MW), and driving the impellers. Other energy uses are much smaller; indeed, the next highest energy use is the air compressor (COMP-2) with 1.72 MW. This process is designed with renewable electricity supply (solar/wind).

The energy production and consumption for the two-stage process as shown in Sankey diagram (Figure 8A) is calculated based on Eqs (2,3).

The required power for running the membranes is carried out using Eq. (1):

$$P = I A V \quad (1)$$

Where, P is the power (kW), I is the total current (A), (Area \times current density), and V the total voltage ($E_{\text{cell}} \times$ number of cells).

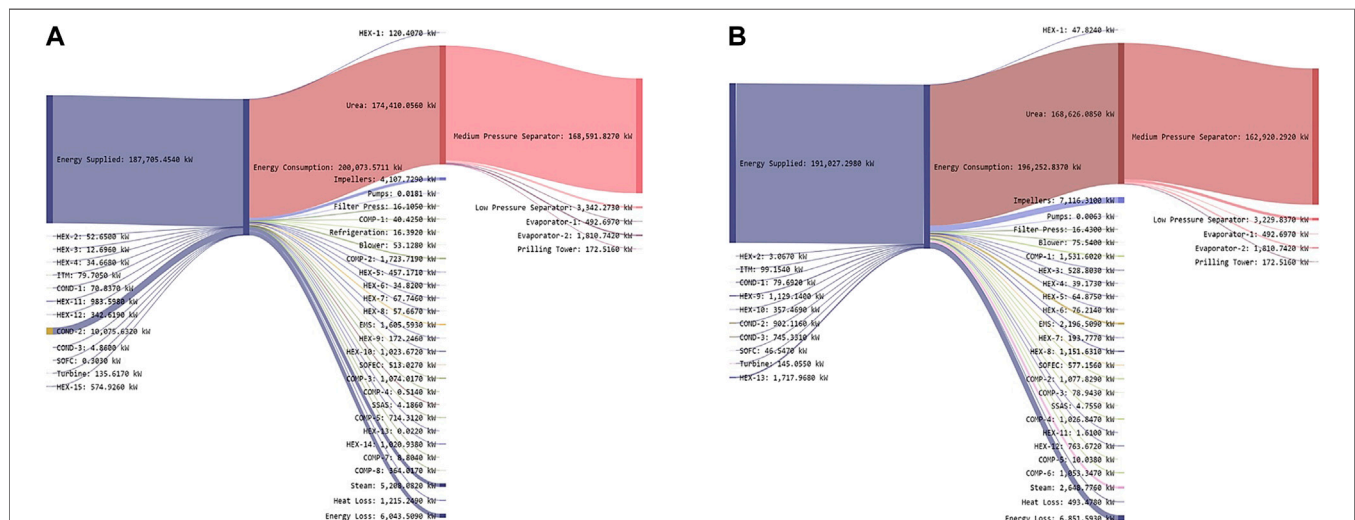


FIGURE 8 | Energy flows in the proposed ammonia production through, (A) two-stage DF coupled with AD (Based on Figure 6A) and (B) AD-only process (Based on Figure 7A).

TABLE 3 | A comparison of the modeling results of the proposed processes for a 20 tpd waste facility in terms of CO₂ emissions, water, and energy consumption (A) daily basis and (B) per kg utilized-N.

(A) Technology	Units	Two-stage DF coupled with AD with urea production and with SOFC	Two-stage DF coupled with AD without urea production and without SOFC	Two-stage DF coupled with AD with urea production and without SOFC	Two-stage DF coupled with AD without urea production and with SOFC	Two-stage DF coupled with AD with CCS and with SOFC	Two-stage DF coupled with CCS without SOFC	Two-stage DF coupled with AD with CCU	AD with urea production and with SOFC	AD without urea production and without SOFC	AD with urea production and without SOFC	AD with urea production and with SOFC	AD without urea production and with SOFC	AD with CCS with SOFC	AD with CCS without SOFC	AD with CCU
CO ₂ Produced	kg	Traces	58.10	Traces	58.10	58.10	58.10	58.10	Traces	56.14	56.14	Traces	56.14	56.14	56.14	56.14
Water consumption	kg	33.28	48.07	33.28	48.07	48.07	48.07	48.07	37.44	50.02	50.02	37.44	50.02	50.02	50.02	50.02
Energy consumption	MW	187.70	13.22	187.70	13.22	187.70	13.50	13.5	191.02	21.13	21.13	191.07	21.08	22.10	22.12	22.08
(B) Technology		Two-stage DF coupled with AD with urea production and with SOFC	Two-stage DF coupled with AD without urea production and without SOFC	Two-stage DF coupled with AD with urea production and without SOFC	Two-stage DF coupled with AD without urea production and with SOFC	Two-stage DF coupled with AD with CCS and with SOFC	Two-stage DF coupled with AD with CCS without SOFC	Two-stage DF coupled with AD with CCU	AD with urea production and with SOFC	AD without urea production and without SOFC	AD with urea production and without SOFC	AD without urea production and with SOFC	AD with CCS with SOFC	AD with CCS without SOFC	AD with CCU	
CO ₂ emitted/kgN		Traces	1.571	Traces	1.571	1.571	1.571	1.571	Traces	1.444	Traces	1.455	1.455	1.455	1.455	
Water consumption/kgN		0.902	1.3	0.902	1.3	1.3	1.3	1.3	0.973	1.287	0.965	1.296	1.296	1.296	1.296	
Energy consumption MW/kgN		5.087	0.358	5.087	0.358	0.365	0.365	0.365	4.961	0.544	4.926	0.546	0.573	0.574	0.572	

Energy Consumed = HEX

$$\begin{aligned}
 & - (1, 5, 6, 7, 8, 9, 10, 13, 14, 15) + \text{COMP} - (1, 2, 3, 4, 5, 7, 8) \\
 & + \text{Required Steam} + \text{Impellers} + \text{Pumps} + \text{Filter Press} \\
 & + \text{Refrigeration} + \text{Blower} + P_{\text{EMS+SOFC+SSAS+UREA}} + \text{Heat loss} \\
 & + \text{Energy loss}
 \end{aligned} \quad (2)$$

$$\begin{aligned}
 \text{Energy Produced} = \text{HEX} - (2, 3, 4, 11, 12) + \text{COND} - (1, 2, 3) \\
 + P_{\text{ITM+SOFC}} + \text{Turbine}
 \end{aligned} \quad (3)$$

B. Energy Balance Results of Ammonia Production Through AD-Only

As with the two-stage configuration, urea production is the largest energy user (Figure 8B), accounting for 85% of the energy draw. This is followed by the power required for driving the impellers (7.11 MW), energy loss (6.85 MW), and power required for supplying steam (2.64 MW) as shown in Figure 8B. Other energy uses are smaller, the next highest energy use is the required power for the EMS with 2.19 MW. The source of this electricity is renewable energy sources (solar/wind).

The energy draw in this process (AD-only) is about 8% lower than the two-stage due to higher NH_3 production. This is due to higher NH_3 yields driven by increased CO_2 production resulting from the incorporation of DF. With AD-only, the energy draw goes up due to higher H_2S handling by the EMS when DF is not used.

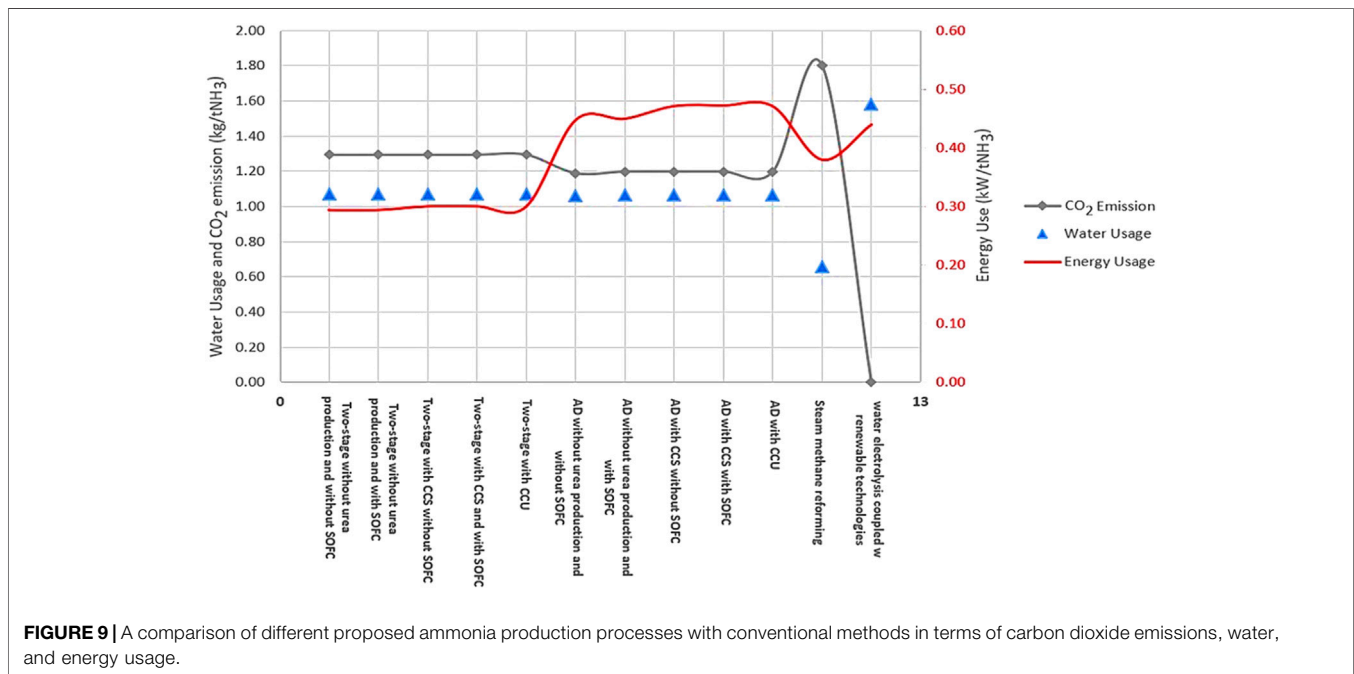
The energy production and consumption for the AD-only process as shown in Sankey diagram (Figure 8B) is calculated based on Eqs. (4,5).

$$\begin{aligned}
 \text{Energy Consumed} = \text{HEX} - (1, 3, 4, 5, 6, 7, 8, 11, 12) + \text{COMP} \\
 - (1, 2, 3, 4, 5, 6) + \text{Required Steam} + \text{Impellers} \\
 + \text{Pumps} + \text{Filter Press} + \text{Blower} \\
 + P_{\text{EMS+SOFC+SSAS+UREA}} + \text{Heat loss} + \text{Energy loss}
 \end{aligned} \quad (4)$$

$$\begin{aligned}
 \text{Energy Produced} = \text{HEX} - (2, 9, 10, 13) + \text{COND} - (1, 2, 3) \\
 + P_{\text{ITM+SOFC}} + \text{Turbine}
 \end{aligned} \quad (5)$$

Comparison of Total Daily Production and Consumption Results for the Two Technologies and Seven Scenarios

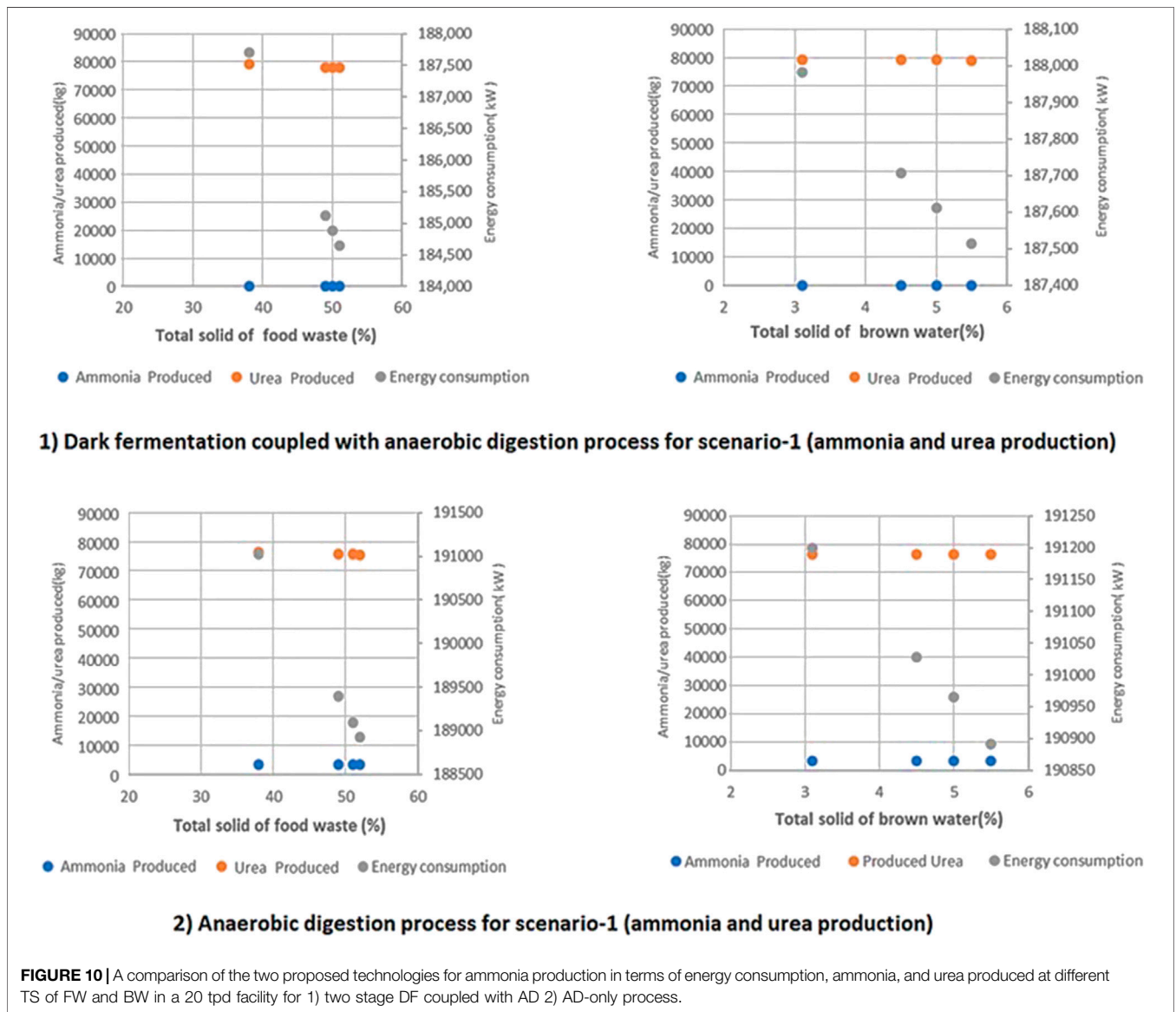
The highest NH_3 yields are obtained from AD, which produces about 6.5 times that of the two-stage process. However, the two-stage process makes more urea in urea-producing configurations. This is due to more CO_2 produced with the incorporation of the DF process. Based on Table 3A, the two-stage process consumes less energy and water compared to the AD-only. The AD-only produces about 3.5% less CO_2 compared to the two-stage process.



Although all calculations for the proposed process are carried out on the basis of per kg NH₃ produced, a comparison needs to be conducted for per kg-N basis, since in each of the two technologies when using scenario ii, both urea and NH₃ are produced. **Table 3B** shows the efficiencies of different proposed processes per kg-N basis. The AD-only process is superior to the two-stage in terms of the CO₂ emitted/kg-N with 1.455 compared to 1.571. The water usage/kg-N through AD is lower compared to the two-stage except in the case where urea is produced with 0.902 compared to 0.973 for AD process. The energy consumption (MW/kg-N) for the two-stage process is lower than the process with AD except in the case where urea is produced with 5.087 MW compared to 4.961 MW for the AD-only process.

A Comparison of the Proposed Processes with Ammonia Technologies Currently in Use

A comparison of the proposed NH₃ production process with conventional methods (SMR and water electrolysis coupled with Haber-Bosch process) in terms of water usage (kg/tNH₃), CO₂ emissions (kg/tNH₃), and energy consumption (kW/tNH₃) at steady state condition is shown in **Figure 9** (for detailed information see the supplementary materials). This figure shows that the proposed NH₃ process through a two-stage DF coupled with AD both with a and without the incorporation of SOFC is the most efficient in terms of water consumption compared to the other technologies. SMR-Haber-Bosch (H-B) uses 0.66 kgH₂O/tNH₃. While the consumption rate for water



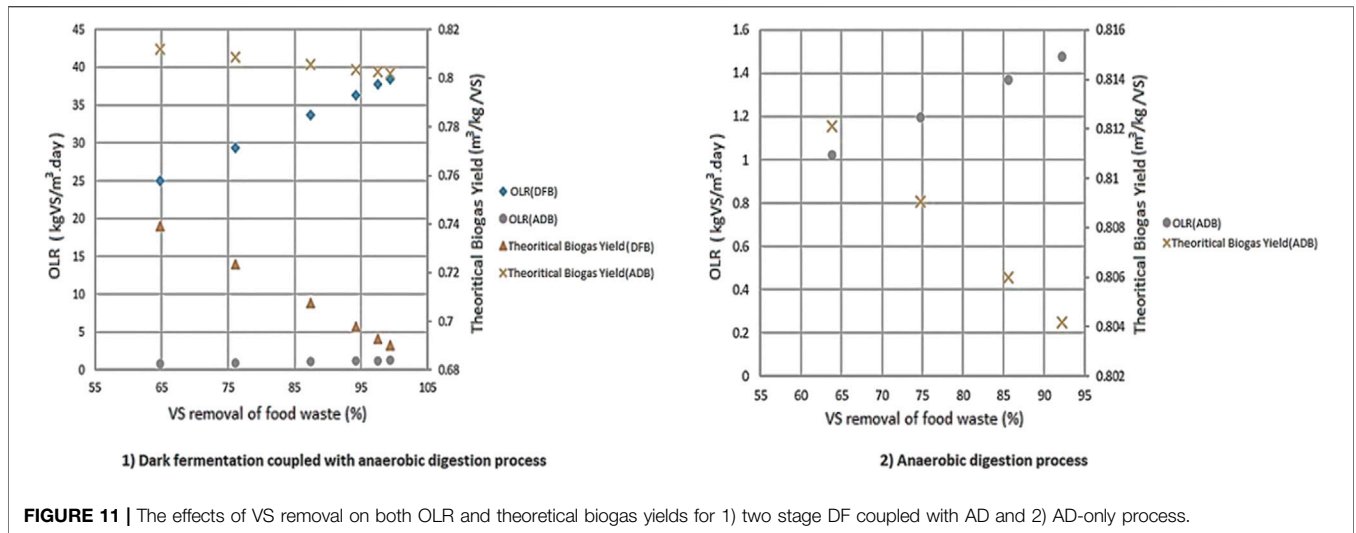


FIGURE 11 | The effects of VS removal on both OLR and theoretical biogas yields for 1) two stage DF coupled with AD and 2) AD-only process.

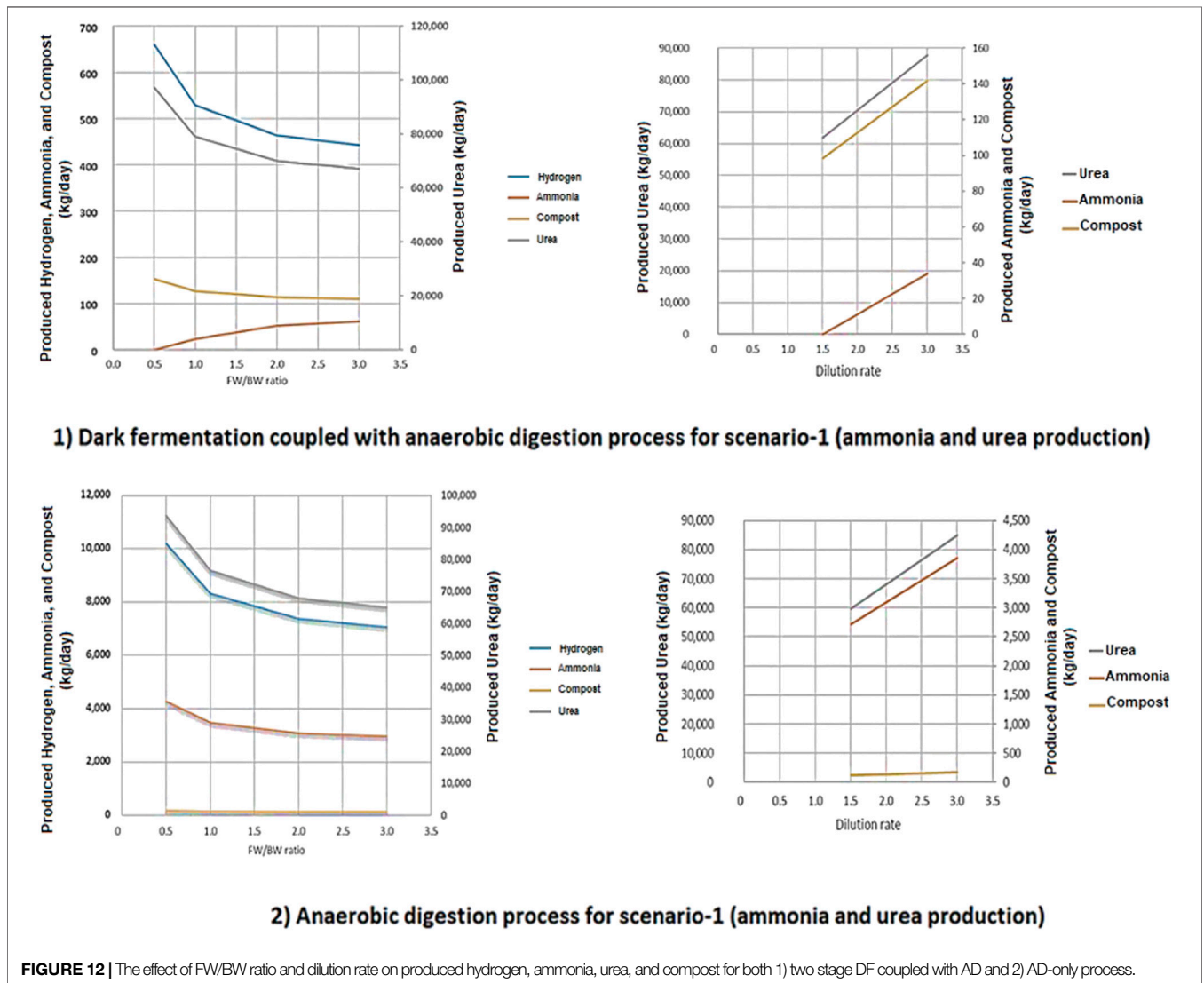


FIGURE 12 | The effect of FW/BW ratio and dilution rate on produced hydrogen, ammonia, urea, and compost for both 1) two stage DF coupled with AD and 2) AD-only process.

electrolysis is approximately 82% higher than SMR-H-B. In terms of CO₂ emissions, this process is similar to water electrolysis, assuming that both processes are powered by renewable energy sources. However, the energy consumption of water electrolysis is 0.44 kW/tNH₃ which is approximately 15% higher than SMR-H-B and approximately 41% higher than the two-stage process and about 6% lower than the AD-only process.

Carbon Dioxide Mitigation by Diverting Different Fractions of FW into the Modeled Process

Assuming 10%, 33%, 50%, and 100% of the FW is directed into our proposed process instead of degrading openly in landfills, this will prevent the release of about 5,159.06, 17,024.91, 25,795.32, and 51,590.64 kg CO₂ eq respectively from directly reaching the atmosphere, which results from the open decomposition of FW (see **Supplementary Figure S4** in Supplementary Materials).

Sensitivity Analysis

The sensitivity analysis studies the dependency in the output of the proposed model (gas and biogas production, output flow rate of NH₃ and urea, number of cells/tubes for membranes, energy demand, water and energy consumption) and the identifiability and significance of the parameters involved (waste compositions, substrate concentrations, input air flow rates, and different dilution ratios). The Total Solid (TS) and Volatile Solid (VS) contents of a substrate affect the functionality of both DF and AD. Changing the TS content of the substrate will result in variations in microbial morphology of DF and AD (Orhorhoro et al., 2017).

Changing the TS of FW and BW for a two-stage DF coupled with AD leads to changes in energy consumption. Sensitivity analysis has been conducted in order to assess how the above-mentioned functionalities respond to changes in these and other key parameters, and about how much variability in these streams may matter to the process conditions, yields, and process outputs. By keeping the VS of FW and BW parameters fixed throughout the sensitivity analysis, the following results as shown in **Figure 10** were obtained. The energy consumption of the two-stage DF coupled with AD process (1) is approximately 1.7% higher than the AD-only process (2). As the TS of FW and BW goes up, the energy consumption of both processes (1) and (2) drops significantly due to lower gas/biogas production.

Variations on the VS removal of FW have a more significant impact on the two-stage DF coupled with AD process (1) on Organic Loading Rate (OLR) and the theoretical biogas yields compared to the AD-only process (2). This effect becomes more evident by varying the VS removal of FW compared to BW as shown in **Figure 11**.

For the two-stage DF coupled with AD process (2), increasing the ratio of FW/BW results in a decrease in the produced H₂, NH₃, urea, and compost. However, the decrease in urea production is sharper than in the other process outputs. When the ratio of FW/BW is 0.5 the optimum production yields were achieved. Conversely, for the two-stage DF coupled with AD process (2), the NH₃ production yield goes up when the ratio of FW/BW increases. This is due to higher CO₂ production resulting from the presence of DF in the proposed process based on our modeling method described in the logical flow diagram which can be seen in the supplementary materials. Increasing the dilution rate will result in an increase in compost and CH₄ production in both processes. An increase in CH₄ production will in turn result in higher NH₃ and urea production (**Figure 12**).

CONCLUSION

The results above indicate that among the proposed technologies, the AD-only is the most efficient for GHG emissions, while the two-stage DF coupled with AD process requires less energy per kgNH₃ basis. When comparing the proposed NH₃ processes to the literature data, water electrolysis powered by renewable technologies is the best of all processes in terms of GHG emissions while steam reforming coupled with Haber-Bosch has the lowest water consumption of all the processes mentioned.

Additional research is needed in a few key areas of research on the production of NH₃/urea through waste utilization, environmental impact of water usage for NH₃ production and where an NH₃ production plant needs to be located, in terms of both availability of feedstock and sustainability (accessibility to renewable energy sources such as solar Photovoltaic (PV) and wind power, distance from the waste hub to the production plant, etc.). Location is an important factor in the development of sustainable production methods, for maximizing productivity and sustainability in the production of chemical compounds such as NH₃. Since the inputs and outputs of this proposed NH₃ production process are FW and BW, wastewater treatment and solid waste management facilities are resources of interest.

A waste-based, CO₂ aware, NH₃ production technology leveraging emerging membrane technologies in concert with local infrastructure can provide a viable route to green NH₃ and to green fertilizers, providing an effective strategy to address multiple sustainability/sustainable development needs simultaneously.

DATA AVAILABILITY STATEMENT

The raw data supporting the conclusions of this article will be made available by the authors, without undue reservation.

AUTHOR CONTRIBUTIONS

SG is the lead author and did the model development, analysis, and drafting of the paper. CT gave conceptual input on the model and contributed to the draft. PS was the PhD Supervisor.

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SUPPLEMENTARY MATERIAL

The Supplementary Material for this article can be found online at: <https://www.frontiersin.org/articles/10.3389/fenrg.2021.600071/full#supplementary-material>

Conflict of Interest: Author CT was employed by the company EarthShift Global LLC.

The remaining authors declare that the research was conducted in the absence of any commercial or financial relationships that could be construed as a potential conflict of interest.

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GLOSSARY

ADB Anaerobic Digestion Bioreactor
BW Brown Water
CCS Carbon Capture and Sequestration
CCU Carbon Capture and Utilization
COMP Compressor
COND Condenser
DFB Dark Fermentation Bioreactor
EMS Electrochemical Membrane Separation
EPA Environmental Protection Agency
FAO Food and Agriculture Organization
FW Food Waste
GHG Greenhouse Gas
HEX Heat Exchanger
HW Human Waste
ITM Ion Transport Membrane
LCA Life Cycle Assessment
MFI Membrane Filtration Index
PSA Pressure Swing Adsorption
PV Photovoltaic
SMR Steam Methane Reforming
SOFC Solid Oxide Fuel Cell

SOFC Solid Oxide Fuel Electrolysis Cell
SRT Solid Retention Time
SSAS Solid State Ammonia Synthesis
TVFA Total Volatile Fatty Acid
USEPA U.S. Environmental Protection Agency
VBA Visual Basic for Applications

NOMENCLATURE

A Surface Area
CO₂ eq Carbon Dioxide Equivalent
COD Chemical Oxygen Demand
FE (%) Faradaic efficiency (%)
I Total Current
OLR Organic Loading Rate
P Power
SCOD Soluble Chemical Oxygen Demand
TCOD Total Chemical Oxygen Demand
TVFA Total Volatile Fatty Acid
TS Total Solid
V Total Voltage
VS Volatile Solid