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Review

Sustainable hydrogen through decomposition of ammonia and its derivatives by thermochemical processes: a review

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Abstract. Thermochemical processes have recently attracted more attentions from researchers, particularly the use of decomposition of ammonia-based compounds and water-gas shift and steam reforming processes. These processes may be applied for hydrogen generation (H_2) from a syngas mixture and that is the main concern of this paper. Furthermore, these processes are currently facing the challenge of low hydrogen yields. Traditionally, H_2 fuel is generated from fossil fuels by utilizing different reforming processes. The longevity and reliability of catalysts, their purity and the start-up time/costs are also some of the critical challenges to be faced. Sustainable H_2 generation from sustainable sources like ammonia (NH_3) gas was found to be an innovative process with the appropriate selection of the catalytic process and also thermochemical processes being shown to be critical issues. Traditional approaches to H_2 synthesis such as the ammonia cracking process and the generation of hydrogen occur at high temperatures of 500°C or more. Current efforts in the field of H_2 generation rely on a process which includes the decomposition of ammonia gas, this achieves a high yield through the application of alkali metal amide/imides as effective catalysts. NH_3 and methane (CH_4) decomposition is exploited for H_2 production through the application of a steam methane reforming process in bench scale packed-bed reactors. This requires an NH_3 feed solution at a temperature of 150°C that can generate more H_2 (up to 90%) in the total syngas yield. Efforts were made to change/shift the equilibrium towards increased hydrogen production. Researchers have engaged in many efforts to remove H_2 from a membrane or in carbon dioxide extraction with the use of a solid sorbent. A continuous mode of enhanced H_2 production can be achieved by integrating the reforming process with concentrated solar radiation for thermal storage. Efforts were made to remove H_2 from a membrane or extract carbon dioxide using a solid sorbent. This review explores the decomposition techniques, catalytic systems and thermochemical conditions required for H_2 generation from NH_3/NH_3-rich products/wastes.

Keywords: ammonia, catalysts, decomposition, hydrogen, thermochemical

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1. INTRODUCTION

Clean energy sources like hydrogen (H_2) are generated from the utilization of natural resources such as renewable plants or microbial biomass. Solar, wind, hydropower, tidal or geothermal power can be applied to break down water molecules into H_2 gas. These resources can be regenerated within a reasonably short timeframe. The clean energy economy may be powered by increasing the scale of renewable energy production and achieving more ambitious energy efficiency goals *via* the adoption of sustainable development goals/methods (Hosseini and Wahid, 2016). Various efforts have been made to develop biohydrogen (bioH_2) and H_2 as a fuel and clean energy source in order to provide a better alternative to fossil fuels as an energy carrier source. Supercritical gasification processes (these utilize biogases), fermentative bioH_2 production, microbial electrolysis cells, plant biomasses, bio-oil and algal plants have all been exploited for bioH_2 production with different yields/productivity values and purities (Sandaka and Kumar, 2023). Current research progress and development for H_2 biosynthesis is also being explored by using different conversion techniques like steam reforming and water-gas-shift reaction. More discussions took place concerning the process or technology development for enhancing bioH_2 yield/biosynthesis processes (Sandaka and Kumar, 2023; Kweku *et al.,* 2018).

H_2 is used as a fuel nd it originates from clean energy sources. There are zero carbon emissions added to the environment during H_2 utilization/burning process in fuel cells or internal engines respectively. This fuel can be used as an energy source in passenger cars, fuel cells may be used in buses as an alternative fuel to gasoline/higher carbon fuel/ biofuels. Furthermore, H_2 fuel may be utilized in spacecraft propulsion (Kweku *et al.,* 2018). Clean fuel like H_2 can be produced from diverse domestic resources and it could be made available as an alternative fuel/ energy from its infancy stage in the market and used as a transportation fuel (Kweku *et al.,* 2018; Hosseini and Butler, 2020). Normally, thermal decomposition and also catalytic cracking techniques (such as electrolysis/ electrochemical oxidation processes) are applied for ammonia (NH_3) gas decomposition into nitrogen (N_2) and hydrogen (H_2). Various effective conversion/decomposition techniques may be applied in H_2 production processes and they make use of the in-cylinder NH_3 reforming and NH_3-rich combustion technique (Liu *et al.,* 2022). Some researchers have investigated the NH_3-rich combustion process that facilitates the production of H_2 and this utilizes the rapid compression method. It works under conditions of varied pressure (in the range of 22 to 36 bar), temperature (1200 to 1300 K) and also equivalence ratios (1.7 to 2.25) (Li *et al.,* 2016). In the combustion process, some reactants like NH_3 and also products like hydrogen and nitrogen were reported. This was confirmed through the application of a rapid sampling system and also gas chromatography (GC) technique followed by the interpretation of the experimental results (Liu *et al.,* 2022; Li *et al.,* 2016).

Researchers have studied the H_2 production mechanism using first testing approaches and also compared them to the existing experimental results in this context. A time-scale analysis approach was reported in order to study this mechanism (Bykov *et al.,* 2023). It was found that a combination of local and global analysis, based on the Global Quasi-Linearization approach (GQL) had potential. GQL was applied for the estimation of the various reactions with very slow and fast reaction processes, they were governed by a system dynamics approach in an effective way (Li *et al.,* 2016; Bykov *et al.,* 2023). One research report discusses the H_2 synthesis process of exposing the substrates of NH_3 and oxygen (O_2) at normal temperature to acidic $RuO_2/\gamma\text{-}Al_2O_3$ catalysts. And then an effort was made to analyse the mechanism of the synthesis with the study of the adsorption of NH_3 on the surface of the catalysts, it had an exothermic reaction nature (Nagaoka *et al*., 2017). In this process, the catalyst bed was found to rapidly heat the catalysed NH_3 to its auto-ignition temperature value and then it subsequently went through the oxidative decomposition process to produce H_2. This decomposition process can be monitored through the huge quantity of heat released due to the chemisorption capacity of the ruthenium oxide (RuO_2) catalyst, the active sites on the $\gamma\text{-}Al_2O_3$ and the physisorption of many NH_3 molecules are also responsible for this phenomenon (Liu *et al.,* 2022; Nagaoka *et al*., 2017). This review discusses the different sources of NH_3 gasses, its decomposition mechanisms and the processes involved in H_2 formation. Additionally, it discusses the catalytic process and also the respective parameters for H_2 production that can influence both the quantities and quality of the H_2 produced in some applications.

2. AMMONIA GENERATION FROM NATURAL SOURCES

Ammonia (NH_3) generation from various natural resources has been reported, it is a waste compound and its sources can also be found in industrial chemical processes, especially those of modern NH_3 producing plants. NH_3 production has been reported at a worldwide level, this includes the U.S. NH_3 generation is involved in many types of biological reactions. NH_3 can function as a precursor for amino acid and nucleotide synthesis processes (Zhang *et al.,* 2012). NH_3 generation has been reported in the environment and it also contributes to the N_2-cycle process. It is generally synthesized in the soil from various bacterial species/community processes. In normal circumstances, rhizobacteria can emit high levels of NH_3 and this may be confirmed during the co-cultivation process in compartmental petri-dishes. NH_3 may be responsible for the alkaline nature of the plant medium in its immediate vicinity and this can reduce the growth rate of the *Arabidopsis thaliana* strain (*A. thaliana*) (Zhu *et al.,* 2015). Furthermore, some arguments for using a particular NH_3 source/generation technique were made depending on the prevailing natural conditions and the environmental conditions; and also several natural products produced by animals and also human bodies contain high-protein resources (like milk whey, carcasses, manure and compost). Certain degradation compounds were reported, these form in bacterial mediated processes and involve ammonia emission/generation process (Chen *et al.,* 2022). Also, NH_3 can alter the pH of the rhizosphere and it can also influence the organismal diversity and plant-microbe interaction. In the nitrogen (N_2)-cycle process, NH_3 is naturally produced from the organic matter degradation process and this organic matter in our environment includes plants, animals and animal waste products which are also good sources of ammonia that are released into the environment (Zhang *et al.,* 2012; Zhu *et al.,* 2015; Chen *et al.,* 2022).

Many research papers have discussed the highest rate/ quantities of NH_3 generation from peptides, casein protein and amino acids. Such amino acids are found in the form of glutamine (Glu), lysine (Lys), serine (Ser) and aspartate (Asp) which can decompose or metabolize to form NH_3. Such an NH_3 formation process can be confirmed through monensin (polyether antibiotic) compound analysis (Richardson *et al.,* 2013). This compound has shown the ability to inhibit NH_3 production from amino acids by 60% especially for gram positive bacteria (those utilized for NH_3 production). A population of both asaccharolytic and acidic amino acid-fermenting bacterial species is found in the mammalian rumen/colon. These are hyper-ammonia-producing (HAP) bacterial species (Grishin *et al.,* 2020). In the HAP categories, bacteria have shown their potential to grow on peptide/amino acid nutrients with a 3.5% or higher percentage of the total viable cell count in the colon than the rumen section of the gut. *Clostridium* species (like *C. perfringens*) and other bacterial species like *Enterococcus, Shigella* and *E. coli* have been reported in the enrichment media and by nature these bacterial cells are not HAP (Grishin *et al.,* 2020; Eschenlauer *et al*., 2002).

Some papers have discussed NH_3 production from amino acid-rich compounds and also amino acid biomass as optimal resources. An NH_3 production study may be performed through the modification of the *E. coli* pathway. In this study researchers developed an engineered metabolic flux that can be used to promote the NH_3 generation process via the overexpression of the ketoisovalerate decarboxylase enzyme gene (*kiv*d), this is derived from the *Lactococcus lactis* species (Mikami *et al.,* 2017). This modification of the *E. coli* strain can produce a high NH_3 concentration (*i.e*., 351 mg L^{-1}/ yield~ 36.6%). Another research effort was made to delete the *gln*A (glutamine amino acid) gene and this gene deletion was responsible for NH_3 assimilation (Choi *et al.,* 2014). This engineered *E. coli* strain was grown in media containing a yeast extract as nitrogen and carbon sources and the bacterial strain biosynthesized NH_3 (production titer of 458 mg L^{-1}) with a yield of 47.8% using an amino acid-based biomass like sustainable material. This titer of NH_3 was found to have the highest concentration with supporting eco-friendly processes for this metabolite originating from the biomass (Mikami *et al.,* 2017; Lan *et al.,* 2012). It was also found to fix the rate of NH_3 production and then it was confirmed by intra-renal distribution system that biosynthesized the NH_3-rich waste matter like urea. It is a crucial step in the renal regulation of the acid-base balance in the human body (Nagami and Hamm, 2017).

Some reports concerning NH_3 generation are also discussed with an emphasis on the process of acid-base disorders. These are associated with changes in the NH_3 generation process with a distribution between the urine and renal veins systems (Nagami and Hamm, 2017; Silva and Mohebbi, 2022). In the human body, urine pH, urine flow and total NH_3 generation along with the renal blood flow rate are studied. Furthermore, this can also affect the percentage of NH_3 synthesis and then its excretion into urine with different extents of acid-base disturbance (Weiner and Verlander, 2023). In the kidneys, the condition of hypertrophy has an extra influence on the ammoniagenesis process in humans (Kim, 2021). In the tubule epithelial cells, there is some possibility of increased NH_3 production as opposed to the acidosis process. This can cause tubular hypertrophy problems which are related to the inhibition of protein degradation. This issue is due to changes in lysosomal pH and also cathepsin activity (Weiner and Verlander, 2023; Kim, 2021). This phenomenon was found to produce changes in the PI-3 kinase pathway and also the suppression of the chaperone-mediated autophagy processes. These are both candidates for the correct mechanism of the NH_3-mediated inhibition process of protein degradation (Lan *et al.,* 2012; Nagami and Hamm, 2017; Kim, 2021).

In another paper, novel integrated techniques were applied for the conversion of algal biomass into ammonia and this was found to be an effective and sustainable technique for achieving a high degree of total energy efficiency. In this approach, the circulation of energy or heat during energy efficiency tasks is examined. One integrated system was found to consist of hydrothermal gasification (HTG), N_2 production and ammonia synthesis, chemical looping and power generation. In this technique, the algal biomass is first converted into syngas via the HTG technique. And later the syngas can be converted into hydrogen and carbon dioxide in chemical looping modules (Wijayanta and Aziz, 2019). The biosynthesized hydrogen from the chemical looping module systems can react with the high purity N_2 from the N_2 production module to form ammonia in the NH_3 synthesis module. During this work study, people realize benefits of the high energy efficiency processand then they put efforts for enhancing the process integration with advanced technologies and also energy recovery approaches. The energy or heat utilization in the integrated system was achieved through the application of a recirculation system in a complete way which makes use of power generation (Singh *et al.*, 2024). In order to test this concept, researchers have used a macroalgae biomass such as the *Cladophora glomerata* species, for example. Also, the effects of temperature and the algae-to-water mass ratio were studied during the HTG process and the results produced have helped to evaluate their potential influence on the achievement of total energy efficiency. Process modelling and the calculations associated with it were carried out using SimSci Pro/II software and the results were applied to produce an integrated system that had an improved total energy efficiency (38%) with regard to the production of ammonia and power. A temperature of (380°C) and a mass ratio of (0.01) were found to be favourable for the HTG process (Wijayanta and Aziz, 2019; Singh *et al.*, 2024)

2.1. Agricultural and environmental sources of ammonia generation

NH_3 gas may be identified due to its pungent smelling properties and it can be produced during the natural degradation of protein and also urea compounds (which may be used as a source of chemical fertilizer in crop cultivation). These compounds are easily available in the slurry and manures from farm animal sources. Some reports have claimed that 90% of NH_3 generation originates from agricultural activities in Europe and due to these activities, NH_3 can be released into the environment (Naseem and King, 2018). The storage of manures, and the application of fertilizers to crop fields and grasslands is also responsible for the release of NH_3. More than 50% of NH_3 generation/ emissions originate from animal husbandry sources such as cattle, pigs and poultry. Due to the invention of the Haber-Bosch process, synthetic fertilizers can be produced and this process is responsible for manufacturing huge quantities of the urea compound which is used for crop cultivation (Tunå *et al.,* 2014). Due to urea application in crop-lands, huge quantities of NH_3 are released into the environment. From last few decades onward, this NH_3 release to soils/ farm lands can contribute strong alkaline conditions to the environment. This is due to high intensity crop cultivation processes. NH_3 emissions have also been reported to originate from agricultural residues/animal wastes and may be increasing in importance due to potential feedstock production. And these are also increased biogas production in recent decades, with exceptionally high NH_3 releases from agricultural activities in Germany (for NH_3 emissions from agricultural processes) (Tunå *et al.,* 2014; Naseem and King, 2018). The emissions of NH_3 may occur as a result of stored and land-applied manure sources and the cumulative result may be high NH_3 emissions in the environment. This emission of NH_3 can result in nitrogen loss for crop production. Some efforts have been made to evaluate manure handling practices in order to maintain the nutritive value of manures applied to crop-lands (Arora *et al.,* 2016). The mitigation of NH_3 emissions from manure sources may help to reduce any negative impact on the environment. Urea fertilizers use has been shown to cause the release of high NH_3 concentrations into the atmosphere and this can cause detrimental changes to the acidic nature of the land and surface water. And these changes in turn can result in plant damage with a reduction in plant diversity in the natural system (Ikäheimo *et al.,* 2018). Also, NH_3 emissions from manures can release unpleasant odours into the environment which may be an indication of intensive livestock operation. Some efforts have been made in recent years to reduce NH_3 emissions with the mitigation of odour problems in the environment via the alteration of manure management strategies (Arora *et al.,* 2016; Ikäheimo *et al.,* 2018).

Some reviews have discussed NH_3 generation/emissions from agricultural sources and also some of the techniques applied to determine the NH_3 loss from manure-amended soils in particular locations. NH_3 loss determination was carried out through the application of micrometeorological approaches/techniques, and these were applied to estimate the field scale NH_3 emissions on small plots. In this context, the effects of the treatment of soils with manure were studied using chambers and mass balanced techniques (Fasihi *et al.,* 2021). This technique was found to be a practical, straightforward method and it was combined with a denuder mounted on a wind vane and this permitted a certain flexibility in the experimental design. In this technique, low NH_3 concentration samples were used for efficiency compared to traditional mass balance approaches (Miyahira and Aziz, 2022). Reports concerning NH_3 emissions from agricultural sources reported the values ranging from 55 to 95% and these are produced due to human activity in the agriculture sector and are added to the atmosphere every year. These NH_3 releases from compounds containing N elements are an indication of the huge amounts of nutrients and energy involved in sustaining agricultural sources/systems. More claims on atmospheric NH_3 releases processes were done and these are due to application of huge quantity of livestock manure with its storage volume capacity (Fasihi *et al.,* 2021; Miyahira and Aziz, 2022). In Europe, 7.6 Mt NH_3/year was reported to be emitted from livestock manure sources with a conversion factor of 0.822, NH_3=NH_3-N made a contribution of 85% with the remaining contribution coming from synthetic fertilizer sources in the environment (Palone *et al.,* 2023). In China, the NH_3 emission percentage is 55% with Asia of huge producing (24.7 Mt/year) and livestock contributes nearly 29% of the total for NH_3 releases while synthetic fertilizers contribute 47% of the total NH_3 emissions. We discuss two main sources of NH_3 (ammonia) releases into the environment (Miyahira and Aziz, 2022; Palone *et al.,* 2023).

2.2. Slurry and manures from livestock sources for ammonia generation

The waste produced by domestic animals are reported to account for 78% of the nitrogen that is ingested by the animal via the feeding process and this percentage of N is dependent on the type of animal species and also on the type of feed and protein intake. In this context, some animals like hogs have been shown to lower the excretion rate/ capacity (52 and 38%, respectively) based on the dietary supplement used and animal species (Bourdin *et al.,* 2014). There are also reports concerning the N quantity excreted by highly productive dairy cows, they have been shown to excrete 140 kg N/animal year while hens can excrete only 1 kg N/ animal/year. Information concerning the quantity of nitrogen produced by various excretion sources includes various byproducts/waste products like urine (with urea) and then it can be converted into ammonia and carbon dioxide with the help of urease enzymes in faeces (Mendes *et al.,* 2017). Excreted N% from manure sources can contribute in a significant way to the emission of ammonia to the environment and some countries like the Netherlands have shown to increase in emissions from 15% to 30% in the form of N_2. N may originate from housing livestock and storing their manure, this may result in unnecessary emissions (Bourdin *et al.,* 2014; Mendes *et al.,* 2017). For its systematic study on ammonia emission, that is released in huge quantity from various manure sources. In context, the study was carried out in a free-standing barn and the % loss of total N in dairy manures was estimated via the application of the valorization process for ammonia with a determination of 40 to 60%. In this study, the scraping tasks required to remove manure from the floor were carried out on a daily basis. A lower rate of ammonia production (in the range of 5 to 27%) for swine barns with a liquid manure system was reported (Pereira *et al.,* 2020; Arnaiz del Pozo *et al.,* 2022).

Further, discussion on NH_3 emission was done on manure stored locations from outside resources and these are found to contain a significant quantity of atmospheric N-content in released ammonia. In this study, uncovered storage tanks with pig/cattle slurry are reported to generate 3-5 g NH_3-N m^{-2} d^{-1}. For these slurry sources the volatilization rate was found to depend on the wind speed, crust formation, the composition of the manures and also the temperature range. A linear relationship between the valorization rate and temperature (for values ranging from 15 to 25°C). At these temperature values, most of the livestock-derived ammonia lost was reported to have entered the atmosphere and originated from manures and slurry sources that was applied to the soil surface (Ma *et al.,* 2020). The rate of ammonia loss may vary depending on the material in contact and the management strategies used (Grant and Boehm, 2020). In the last few decades, some researchers have determined that total ammonia losses are nearly 1.5 times greater from slurry applied to grassland when compared to it being applied to bare soils. Ammonia valorization from urine application to grass can only account for 3% of nitrogen loss. Also, a value in the range of 56 to 60% of ammonia generation was reported to originate from the anaerobic mode digestion of matter from the sewage sludge valorization process in the first 5 to 7 days after application (Grant and Boehm, 2020; Ma *et al.,* 2020).

2.3. Synthetic fertilizer for ammonia generation

Some studies were carried out concerning the rate of the NH_3 volatilization process from synthetic fertilizers and this NH_3 volatilization rate was found to vary due to the composition variation of synthetic fertilizers. The highest emission rate was reported for urea, with 6 to 25% of its nitrogen being converted to ammonia (Xu *et al.,* 2019). Some sources like calcareous clay soil in the Netherlands were studied with the application of calcium ammonium nitrate (250-550 kg N ha^{-1} with 6 to 7 times/ year) to grazed lands/pasture. These nitrogen sources were shown to be the source of the total N losses (in form of ammonia release) from applied soil source (5 to 14% in the first year and 3 to 7% in the second year) (Effah *et al.,* 2023). Some reports concerning NH_3 emissions from synthetic fertilizers were discussed with reference to the U.K. giving emission factors for ammonia nitrate (3%) and urea (10%) fertilizers. And ammonia releases in atmosphere was estimated with value of 3.4% of applied nitrogen in synthetic fertilizers. This value is found as ammonia loss via its systematic determination. Next, studies were shown on a significant amount of NH_3 loss due to anhydrous ammonia according to systematic reports/ studies. The rate of NH_3 loss was found to depend on the soil moisture content (Effah *et al.,* 2023; Xu *et al.,* 2019). Some studies were carried out concerning fertilizer injection into dry and wet soils with regard to the impact of NH_3 losses and ammonia loss values of 20 and 50% were found for dry and wet soils respectively. The NH_3 loss was found to increase in intermediate moist soil conditions. An NH_3 volatilization process study was completed for 107 kg N ha^{-1} for anhydrous NH_3 impact and this only generated losses of 1.0 kg N h^{-1} (Ma *et al.,* 2021; Ma *et al.,* 2021).

2.4. Other sources for ammonia generation

Reports concerning other sources of NH_3 generation are discussed and report of secondary sources of NH_3 emissions originating from agricultural crops is found with a contribution of 10% from livestock. A small number of reviews have also discussed the values representing the emission of NH_3 from vegetation sources. This source may be influenced by both meteorological and plant type factors. Researchers have discussed on the compensation points of atmospheric NH_3 and these are found to below/ less % for vegetation sources. And then these are found to sink above from this level (Häni *et al.,* 2016). When the volatilization of NH_3 from spring barley crops was studied, it was found that in the daytime and night time it can absorb atmospheric NH_3. Further studies were carried out to examine the decomposition process of crop residues and they were found to be a significant source of atmospheric NH_3 which account for ammonia losses related to N_2 concentration in crop residues. In perennial ryegrass herbage, 10% of NH_3 losses may be attributed to herbage with a total of 29.8 mg g^{-1} (Gu *et al.,* 2021). And, there are no losses from herbage with 9.2 mg nitrogen (N) per gram of sample reported. A 14% loss of the applied N can occur for legume green manures and in the U.K., the losses were estimated for volatilized N-losses from grassland and its decomposition can account for 2.7% of the total N emission in agriculture. A lower impact of volatilization was found for herbage drying or senescence sources (Gu *et al.,* 2021; Gong *et al.,* 2013).

2.5. Ammonia synthesis / decomposition processes in the environment

For NH_3 formation and decomposition, the properties of NH_3 need to be properly understood. NH_3 is colourless gas, it is highly irritating and has a pungent nature and a suffocating odour. The gaseous form of NH_3 in our environment is the most abundant form of the compound and it is an alkaline form of the natural gas in the atmosphere. It includes a highly reactive nitrogen as a major component of its structure (Behera *et al.,* 2013). The largest sources of ammonia emissions from the various agricultural sectors include animal husbandry and ammonia-based fertilizer application. The human body is one source of NH_3 and it has also been generated in many industrial operations, vehicular emissions and also due to volatilization, originating from the soil and the oceans. In recent times, some reports have been published concerning ammonia (NH_3) emission sources that are reported to be increasing trends in the last few decades at a global level/ scale (Behera *et al.,* 2013; Farooq *et al.,* 2022). Ammonia generation has many sources from a variety of activities, it is generated in natural ways and also due to anthropogenic daily tasks. Ammonia has many properties, as evidenced by its important/significant/critical role in the formation of particulate matter in the atmosphere, which can lead to low visibility and N_2 deposition, which is detrimental to ecosystems (Farooq *et al.,* 2022).

In our environment, increased levels of ammonia emissions can negatively influence various vital components of the environment and also public health with a high impact on the climate change process at a global level. Due to these properties of ammonia gas, we need to develop an adequate process of understanding of its respective sources, deposition patterns and also its behaviour in the atmosphere (Liu *et al.,* 2022). In the last few years, worthwhile research works have discussed the challenges posed with regard to our collective response to ammonia emissions and this has led to certain relevant issues being addressed with relevant solutions/prevention or mitigation approaches to these emissions. At high concentrations ammonia can influence the atmosphere at global, regional and local levels (Behera *et al.,* 2013; Farooq *et al.,* 2022). Some review papers have discussed the various integration approaches that could be used to decompose/degrade atmospheric ammonia, however, the knowledge available to date is as yet insufficient. The systematic manner of effective control strategies for ammonia emissions have been discussed, these include the development of conversion technologies in order to manufacture clean fuels like hydrogen (Farooq *et al.,* 2022; Liu *et al.,* 2022).

2.6. Impact of ammonia-rich compounds

In its natural form, NH_3 is found in the form of anhydrous ammonia and in its pure and hygroscopic form it can absorb moisture to some extent and then it can become alkaline and corrosive in nature. Furthermore, NH_3 gas can dissolve to form an NH_4OH solution, a caustic solution that is a weak base. NH_3 can easily be compressed to form a clear liquid under high pressure conditions. This liquid form of ammonia can be used for shipping purposes as it exists in a relatively dense liquid form in steel containers (Bhalla *et al*., 2011). NH_3 is not highly flammable in containers, but when exposed to high heat it can explode under certain conditions. At an industrial level, 80% of the NH_3 which is processed and is used for fertilizer manufacture and then it can be applied to agricultural land to improve crop growth. Other uses of NH_3 have been reported, it is used as a refrigerant gas, for the purification of the water supply and also in plastic manufacturing tasks (Bhalla *et al.,* 2011; Jeong *et al.,* 2022). There are many additional uses for ammonia, in explosives, pesticides, dyes and other chemical preparations. NH_3 is also used in cleaning solutions (5 to 10% ammonia in water) for many household applications (Jeong *et al.,* 2022).

At an industrial level, NH_3 solutions are used in more concentrated forms (25% or more) and these are by nature corrosive solutions. In normal use, many people face inhalation issues posed by this gas or by its vapour form. In its natural form, this NH_3 gas is also present in cleaning products and exposure to it can create health issues in humans (Luo *et al.,* 2017). NH_3 has found widespread use on farms sites, and also in industrial and also commercial locations, therefore it can be released during accidental situations and even during terrorist attacks. Furthermore, the anhydrous form of NH_3 gas is lighter than air and rises without dissipation from low-lying areas (Shahsavari *et al.,* 2022). In the form of a vapour, NH_3 can spread along the ground and low-lying areas with poor air-flow when its exposure is found to be non-vapored / its aqueous form. Furthermore, NH_3 can interact with moisture in the skin and eyes to cause irritation. The oral cavity, respiratory tract and the mucous covered surfaces are vulnerable to the formation of NH_4OH (Han *et al.*, 2020). Due to the nature of ammonia in its hydroxide form, necrosis of the tissues may occur. This tissue necrosis can result in the destruction of lipids of cell membrane (saponification) and this can ultimately lead to cellular destruction (Sun *et al.,* 2021).

Also, NH_3 has been shown to adversely affect biological subjects exposed to high concentrations of the substance, these effects include swallowing issues with corrosive damage to the mouth and throat. But, there is no NH_3 impact in ingestion system in adult human due to its non-poisonous nature. In the case of children, NH_3 exposure (in vapour form) at high concentrations/doses can produce a more negative impact on the lung surface due to surface area to body and weight ratios. And increased minutes of exposure of ammonia, volume to weight ratio can show the adverse effect of NH_3 to children (Shahsavari *et al.,* 2022; Han *et al.,* 2020). Exposure to high NH_3 concentrations in the air or from solutions can produce the rapid onset of skin/eye irritation. At high concentrations and exposure times, NH_3 can cause severe injury and burns to humans. A high concentration of NH_3 especially of industrial cleaning agents can cause corrosive injuries like skin burn and may cause permanent damage and blindness (Luo *et al.,* 2017; Sun *et al.,* 2021).

2.7. Impact of ammonia on the environment

Some studies have claimed that 81% of global NH_3 emissions originate from agricultural sources and that nearly 50% of these emissions originate in the EU with 30% originating in the U.S. These are associated with an overall contribution of PM 2.5 to air pollution in environment. This fine particulate matter (PM 2.5), can cause chronic respiratory illness and also lead to premature mortality in humans. Efforts are being made to reduce NH_3 emissions, and this effort can put its impact of PM 2.5 reduction task. And then this PM 2.5 mitigation effort can reduce the premature mortality of children/ new-born child. In this context, ammonia generation regulation was performed using a cost-effective method that has the potential to protect human health (Wyer *et al.,* 2023). Normally, atmospheric NH_3 release occurs from agricultural sources and then it may contribute to acidification process in environment and also have an impact on human health to some extent, based on its concentration. The potential of ammonia to affect human health directly has been shown in its general form to the public, based on the established scientific literature and there have also been exploited in several recent studies. Recently, some studies were carried out concerning the direct effect of NH_3 on the respiratory health of people who handle livestock (Backes *et al.,* 2016). The NH_3 concentration in the environment can cause several adverse health impacts like reduced lung function, irritation to the throat and eye and also increased coughing and phlegm expulsion. Some recent studies have claimed that agricultural ammonia has an influence over the development of early on-set asthma in young children (Backes *et al.,* 2016; Wyer *et al.,* 2023). In addition to these effects, NH_3 may be responsible for PM 2.5 generation in regions like the U.S. and Europe and at least some contribution has been demonstrated. This PM can directly penetrate deep into the lungs and then it causes long-term illness in humans like chronic pulmonary diseases (COPD) and also lung cancer. Furthermore, this phenomenon is responsible for economic losses which may run to billions of dollars in the US and which impact global economic performance every year. Another impact of PM 2.5 is its association with premature death of child which is producing increasing economic losses at a global level (Bauer *et al.,* 2016; Wyer *et al.,* 2023).

3. AMMONIA DECOMPOSITION PROCESSES USING DIFFERENT APPROACHES

3.1. Ammonia decomposition without a catalyst for hydrogen synthesis

In another study, researchers have made efforts to develop a novel Ocean Thermal Conversion (OTEC) process, based tri-generation system for energy /ammonia production. This process was connected with a cooling and power generation system with its systematic analysis process. The OTEC plant for ammonia production was found to operate in a natural way with existing temperature differences that were dependent on various depths of the ocean. In this study, the OTEC plant was operated by using a single-stage ammonia Rankine cycle (Hasan and Dincer, 2020). In this study, discharged seawater from the condenser system was reported to have entered the organic Rankine cycle and then this sea water was used to in cooling system tasks. During this study, two different operational cases were checked in the effective analysis system. In the first case, 50% of the power produced was stored in the form of NH_3 during the off-peak periods/ hours. In the second case, complete power production was reported for peak hours (Yilmaz *et al.*, 2018). In case 1, it was reported that 50% of the power produced was used for ammonia production with the highest energy (1.4 %) and energy efficiency rate (57.2%). In case 2, in the OTEC plant, only the power produced (100%) has shown to produce a better maximum energy value (1.83%) and exergy efficiency (78.02%). In case 1, the maximum power production was found to be 6612 kW and in case 2, a higher maximum power production (13224 kW) was reported. These power capacities can help to produce a high/ maximum rate of hydrogen production (94.35 kg h^{-1}) and also an ammonia production rate of (534.7 kg h^{-1}) at the peak efficiency value. Furthermore, there are reports of an improved cooling effect (64.4 MW) occurring at peak energy

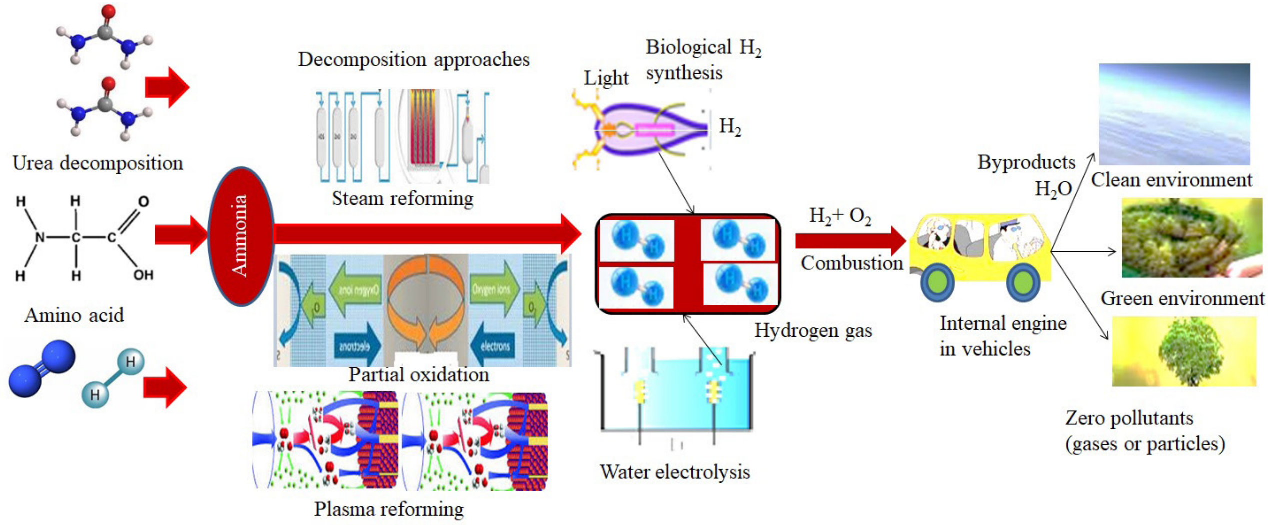

Fig. 1. Different approaches to ammonia decomposition with the evolution of clean hydrogen.

production and exergy efficiency being achieved with a low condenser temperature (11.4°C) (Yilmaz *et al*., 2018; Hasan and Dincer, 2020) (Fig. 1).

3.2. Photocatalyst-mediated NH_3 decomposition

In the ammonia decomposition processes, some reactions formed nitrogen and hydrogen *via* the use of platinum loaded titanium oxide photocatalysts and these catalysts which are based on decomposition. These were tested and then also used for process analysis via ESR (electron spin resonance) and FTIR (Fourier Transform Infrared) spectroscopic techniques. In this process, photo-induced hole formation on the titanium oxide catalyst was found to be responsible for the oxidation of ammonia (NH_3) process and then the formation of an amide radical (NH_2^*) and proton (H^+) ions, reported (Yuzawa *et al.,* 2012). Next, this amide radical can produce hydrazine (N_2H_4). After that, this hydrazine compound can further decompose to form nitrogen and hydrogen as well as the final products of NH_3 decomposition. In another aspect of these reaction steps, photo-formed radical species can aid in the migration of toxic ammonia and this process may be completed via the application of platinum nanoparticles (Pt-nanoparticle). This can result in the formation of conduction bands of titanium oxides and then later the proton is reduced to produce/generate hydrogen as the final product in this process (Yuzawa *et al.,* 2012; Chen *et al.,* 2022). This metal catalyst (Pt-nanoparticle) can work on a large scale and these sorts of metals (including platinum) can provide more effective co-catalyst properties (Liu and Wang, 2023). These photocatalytic reactions may be associated with water molecules and this combination is necessary for the reaction mechanism to continue at an appropriate level (Chen *et al.,* 2022; Liu and Wang, 2023). Some studies were performed using an in situ FTIR technique and the role of water was examined, it was concluded that water can restrict inactive byproduct accumulation such as NH_4^+ on the titanium oxide surface (Sun *et al.,* 2021; Liu and Wang, 2023).

In the current year, the development of advanced agricultural and industrial operational facilities is expanding rapidly and these may lead to the release of huge quantities of ammonia into our environment. Furthermore, this released ammonia has the property of possessing an unpleasant smell and it is generally harmful to the ecosystem. For this released ammonia decomposition process, researchers have chosen to apply photocatalytic approaches/techniques, which is a promising technique for future research (Vikrant *et al.,* 2020). This approach has demonstrated both its potential and its eco-friendly nature for turning ammonia like pollutants into value-added products like clean fuel hydrogen under favourable operating conditions. In this context, researchers have made an effort to utilize titanium dioxide (TiO_2) to optimize the catalytic process. The variable nature of engineered photocatalytic materials is also utilized in order to develop the ammonia decomposition process (Livolsi *et al.*, 2023). The engineered metal catalysts developed in this way demonstrated an enhanced degree of efficiency and produced practical options for the implementation of pollutant reduction such as ammonia decomposition processes. Some comprehensive overviews have focused on the current options for the mitigation of the adverse effects of ammonia in gaseous and aqueous forms (Vikrant *et al.,* 2020; Livolsi *et al.,* 2023). Researchers have compared the performance of photocatalytic materials with various other systems with respect to quantum and space-time yield for NH_3 decomposition. In this context, some attention was given to the reaction mechanisms which are associated with photocatalytic mediated ammonia removal and then these were checked in both the gaseous and liquid mediums (Wu *et al*., 2022).

These efforts were coupled with end product generation/ production, particularly hydrogen and nitrogen produced during the ammonia decomposition/splitting processes. The influence of operational and process variables was noted. These were as follows; irradiation time, relative humidity and the mode of operation is adapted to the environmental matrix type. These factors and others have influenced the performance of ammonia decomposition process (Vikrant *et al.,* 2020; Wu *et al.,* 2022). Next, the intrinsic properties of the engineered materials were explored. The surface functional site and structure were also found to be important. In this decomposition process, some barriers to progress were noted, like byproduct formation of a hazardous nature formed *via* various reaction pathways and these may pose future challenges (Livolsi *et al*., 2023; Wu *et al*., 2022).

In the context of hydrogen storage materials, more uses of metal amines in the indirect mode were found. The usefulness of this material may be entirely dependent on the process of ammonia conversion into hydrogen in effective ways. Normally in ammonia synthesis, the process of nitrogen and hydrogen elements reacting to form ammonia is an exothermic reaction. However, ammonia decomposition is an endothermic process (with the consumption of heat energy as follows: $\Delta H= 46.6\ kJ\ mol^{-1}$. NH_3). Due to the energy input this decomposition reaction requires, it occurs at a slow rate at high temperatures with the need of an effective catalyst (Aziz *et al.,* 2020). During the decomposition reactions, there is a reaction at an equilibrium state and subsequently it is difficult to complete NH_3 decomposition into H_2 and N_2. Researchers have found uses for metal amines in terms of H_2 storage in order to maximize the scale and efficiency of complete fuel cell systems (Aji Wibowo *et al.,* 2019). This arrangement may require efficient heat integration in order to minimize hydrogen losses and also the appropriate characteristics required to achieve a catalyst system that can help to produce a high rate of H_2 synthesis at a sufficient scale.

In fuel cell system, there is no tolerance capacity for problematic concentrations of ammonia, therefore it is necessary to add/include an ammonia scavenger system (Aziz *et al*., 2020; Ajiwibowo *et al.,* 2019). In this context, the fuel cell system falls within the range of expertise of the chemical industry and researchers are implementing ammonia decomposition reactions with the benefit of additional knowledge as model reactions yield with more information (Han *et al.,* 2021). Also, in the distant past some fundamental insight was provided at a technical level which facilitated the achievement of ammonia synthesis. At an industrial level, there are still operational ammonia decomposition plants producing deuterium-enriched ammonia (Aji Wibowo *et al.,* 2019; Han *et al.,* 2021).

Deuterium-enriched ammonia plants may be coupled to consecutive synthesis and decomposition cycles and these plants can operate at low/moderate scales with high temperature values (600°C). At present, many researchers are focusing on optimizing decomposition processes with the help of suitable catalysts (Li *et al.,* 2022). The catalyst mediated decomposition reactions are facilitated by the relevant empirical knowledge concerning the decomposition mechanism of ammonia in its natural form. In recent decades attention has been focused on the catalytic ammonia decomposition process with further consideration of CoX-free hydrogen sources (Yan *et al*., 2021).

3.3. Metal catalyst-mediated ammonia decomposition processes

The multiple efforts being made by researchers are producing a continuous improvement at a significant rate. It is due to our fundamental and basic understanding of the ammonia synthesis process that are associated with their decomposition reaction mechanisms. In this context, we discuss the much promoted iron catalyst which has been applied at optimal concentrations for ammonia solution synthesis, however, it is not effective at promoting ammonia decomposition processes due to the instability of this catalyst in the formation of bulk iron nitride under ammonia decomposition process conditions (Li *et al.,* 2022; Yan *et al.,* 2021). Several researchers have worked on the process of ammonia synthesis and they have reported that this process occurred at a favourable thermodynamic state. In the process of ammonia synthesis, it is carried out at high process temperature/ pressure conditions, and in the case of ammonia decomposition process reaction, it is carried out at low process temperature/ pressure conditions (Park *et al*., 2021). Some further studies were performed on the ammonia synthesis process which achieved an equilibrium state at low ammonia concentrations. In the ammonia decomposition process, respective reaction steps require a high concentration of ammonia for rapid decomposition tasks (Hao *et al.,* 2021).

Researchers have studied the impact of high ammonia pressures and they noted the transformation of iron into iron nitride compounds at rapid rates. From these reaction steps, it was concluded that for different reaction conditions, which were facilitated by the activity of a single catalyst that could not be optimal for both types processes (*i.e*., synthesis and decomposition) and these were shown/ expressed in quantitative terms (Park *et al.,* 2021; Hao *et al.,* 2021). In the ammonia decomposition process, a number of promising catalysts were applied, and one of these is based on supported ruthenium metal with promotion *via* caesium and/or barium metal (Errico *et al*., 2018). Normally, the catalyst is supported on carbon and they can be added to facilitate NH_3 decomposition with catalyst activity in some industrial applications. Certain plants have added them for the last 20 years. These catalysts can produce promising

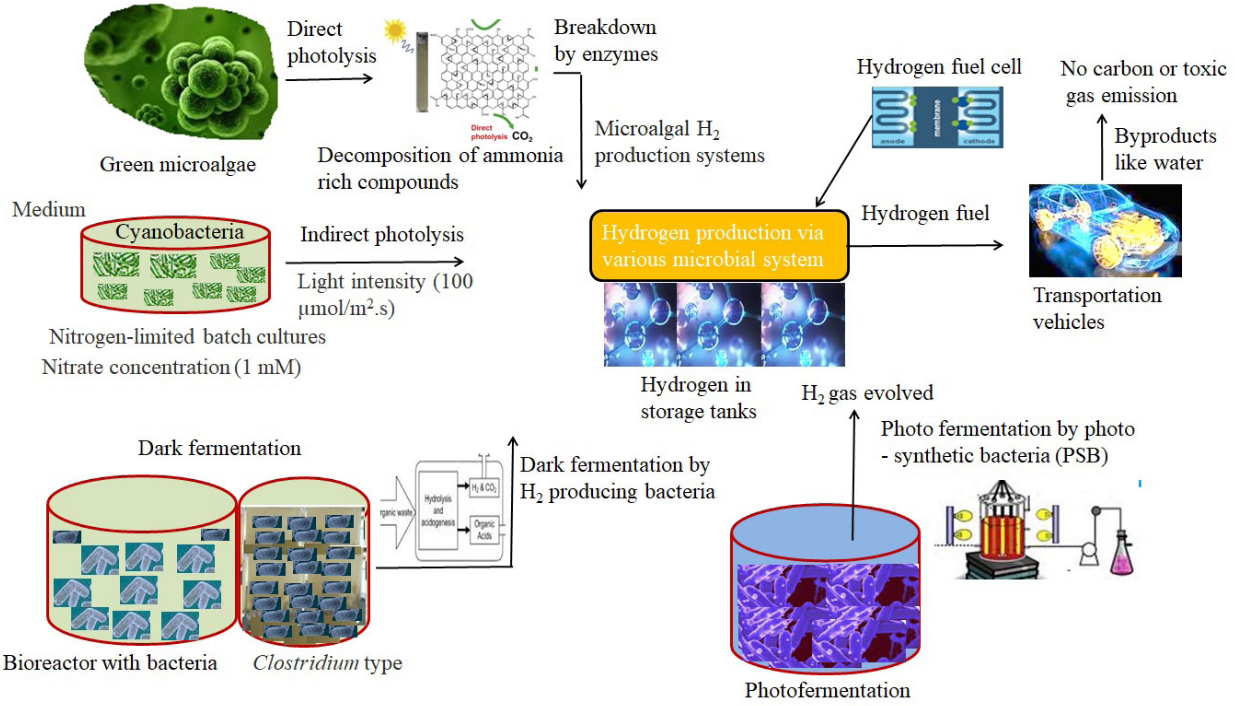

Fig. 2. Ammonia-rich products developed by different biological systems with ammonia decomposition and hydrogen generation.

results under high ammonia pressure conditions (Hao *et al.,* 2021; Errico *et al.,* 2018). Figure 2 discusses the ammonia-rich products developed by different biological systems for ammonia decomposition and hydrogen generation.

It was found that the function of a carbon-supported ruthenium catalyst and its activity is promoted by barium (Ba) or Caesium (Cs) metals in the ammonia decomposition process. This decomposition is reported to occur under the experimental conditions of 1 bar of pressure, H_2 and N_2 ratio (3:1), NH_3 (5 to 50%) and a temperature of 370-400ºC. And under the reaction conditions of the ammonia decomposition process, Cs-Ru/carbon catalysts played a more important role in increasing its rate than the Ba-Ru/carbon catalysts (Raróg-Pilecka *et al.,* 2003). This is due to the larger differences for the samples with a high degree of dispersion. Some studies have been carried out to assess the effect of ruthenium precursors such as carbonyl, chloride, and it has been shown that it is not essential for high activity (Lendzion-Bieluń and Arabczyk, 2013). Researchers have exploited the ammonia decomposition process with 20% of ammonia at 400ºC, time-of-flight (TOF) of ammonia decomposition is discussed over Cs-Ru/ carbon catalyst process and it has shown about 3x 10^2 time more than to K-Fe/carbon catalyst performance for hydrogen chemisorption process. Researchers have estimated the apparent activation energy of the Cs-Ru/carbon (*i.e.*, 134 kJ mol^{-1}) and Ba-Ru/carbon catalyst (*i.e.*, 158 kJ mol^{-1}) (Raróg-Pilecka *et al.,* 2003; Lendzion-Bieluń and Arabczyk, 2013). Temperature-based programmed desorption studies were also carried out. These studies have determined the amount of nitrogen which is desorbed from the Ba-Ru/carbon catalyst and it was found to be lower than from the Cs-Ru/carbon catalyst and its peak performance was found to shift to a higher temperature as compared to the performance of the Cs-Ru/carbon catalyst. The inducing mechanism was found to be the same for both metals and this property can be exploited to facilitate the decomposition of NH_3 (Ji *et al.,* 2011). The nature of other metal catalysts, such as cobalt catalysts, can be exploited as they can act as a precursor for ammonia decomposition processes in the form of cobalt oxide (II/III). This ammonia decomposition process can also be induced by using an oxide of Ca, Al and K with cobalt oxide via a precipitation technique.

Some studies were carried out to assess the impact of increased temperatures on the precipitation process and it was found that higher temperatures reduced the average size of the cobalt oxide (Co_3O_4) crystals generated in the calcination process (Czekajło and Lendzion-Bieluń, 2016). Further studies were carried out concerning alumina compound addition which had a positive effect on the active surface area and also on surface stability and catalyst activity. Metallic cobalt activity was found to be the key factor in the high-rate NH_3 decomposition reaction. During this reaction, for a metal catalyst like cobalt, and particularly for the high activity for catalysts ZBAP1-C is reported and this catalyst can be induced with Ca, K and Al elements (Ji *et al*., 2011; Czekajło and Lendzion-Bieluń, 2016).

The combined activities of these oxides (*i.e*., Ca, Al and K) with catalyst ZBAP1-C can produce a high degree of NH_3 decomposition of up to 100% at a temperature of 525ºC. The author reported that the BRCA 1(breast cancer type-1) associated protein 1 (known as BAP1) and its nuclear deubiquitinase has the capability of regulating tumour suppressor activity, it is involved in various cellular operations such as cell cycle regulation and the gluconeogenesis process (Czekajło and Lendzion-Bieluń, 2016). From previously published work, the XRD studies were applied to investigate the changes in the crystalline structure of the ZBAP-1 catalyst and it was found to be actively involved in the NH_3 decomposition process at a temperature (475°C) with nitriding potential changes (Czekajło and Lendzion-Bieluń, 2016 ; Bhattacharya *et al.,* 2015).

Researchers have carried out few studies concerning the influence of the crystalline phase of alumina on the NH_3 decomposition process when compared with the influence of an alumina supported ruthenium (Ru) catalyst. The function and the natures of various Ru catalysts were studied, these were supported on different alumina materials like α-Al_2O_3 and κ-Al_2O_3. And also, θ-Al_2O_3, δ-Al_2O_3, η- Al_2O_3, γ- Al_2O_3 were reported to function as substrates (Tagliazucca *et al.,* 2013). Ru catalysts with various properties have been prepared using wet impregnation methods. After the preparation of these catalysts, they were systematically characterized through the application of inductively coupled plasma-optical emission spectroscopy. And also, N_2 physisorption methods were used. Furthermore, other techniques like XRD (X-ray diffraction), TEM (transmission electron microscopy) and also the chemisorption approach were also applied in order to perform these catalyst characterization tasks (Kim and Park, 2023). In ammonia decomposition processes, Ru dispersion Ru/Al_2O_3 catalysts have been found to act as a reducing agent or to skip the calcination step and this occurs as follows: Ru/α-Al_2O_3 and Ru/κ-Al_2O_3, Ru/θ-Al_2O_3, Ru/δ-Al_2O_3, Ru/η-Al_2O_3, Ru/γ- Al_2O_3. Ru/a-Al_2O_3 exhibited a high rate of catalytic activities for NH_3 decomposition processes (Tagliazucca *et al.,* 2013; Kim and Park, 2023).

Further studies were carried out concerning the influence of the calcination temperature before the completion of the reduction step process, Ru particles were checked for size and morphology brought about by changes in the calcination process. The different natures of the Ru/α-Al_2O_3, Ru/κ-Al_2O_3 types of catalyst along with Ru particles (size range 7 to 8 nm) have proven to be capable of initiating a high rate of NH_3 decomposition (Lee and Park, 2022). The calcination process of the Ru/Al_2O_3 catalyst was evaluated at various temperatures and then it was reduced at a temperature of 573 K. Numerous analyses were performed on a Ru dispersion and its morphology with a control being provided by the support material and with the calcination temperature as important factors. These play a critical role in H_2 generation via NH_3 decomposition by the Ru/Al_2O_3 catalyst (Kim and Park, 2023; Bell and Torrente-Murciano, 2016).

Further studies were performed on many ammonia decomposition processes and this decomposition played important roles in hydrogen production reaction steps with consideration as a promising practical intercontinental hydrogen carrier option. Several researchers have carried out studies on 1wt% Ru/SiO_2 catalyst activities and it was synthesized using a wet impregnation technique and then subject to calcination in air at various temperatures in order to control the Ru particle size (Mukherjee *et al.,* 2018). Studies were also performed on silica support medium/ materials with different surface areas and these were synthesized after calcination at various temperatures. They can be applied to support changes in Ru particle-size distribution for Ru/ SiO_2 (García-García *et al.,* 2017). Some analytical techniques like N_2 physisorption and TEM were applied to probe the textural properties and also the Ru particle-size distribution of the catalyst respectively (García-García *et al.,* 2017; Mukherjee *et al.,* 2018). Ammonia decomposition can be achieved effectively by using a $RuSiO_2$ catalyst with a high surface area and this process occurs at a high temperature of 400°C (Inokawa *et al.,* 2015). Furthermore, there is a close relationship with the Ru particle-size range at a range of 5 to 6 nm as this range supports the NH_3 decomposition process for this structure sensitive reaction (García-García *et al.,* 2017; Inokawa *et al.,* 2015). Table 1 shows the different approaches to decomposition.

4. HYDROGEN PRODUCTION FROM AMMONIA DECOMPOSITION

In our environment, there are various sources of ammonia in free form or in combined/ inactive forms like urea in animal waste and chemical fertilizers. Also, ammonia is available or it may be generated from multiple processes taking place in soil habitat by bacterial communities or through the oxidation of protein-rich compounds. Various research papers have discussed different approaches like the thermochemical conversion for ammonia decomposition and its connection with hydrogen fuel generation as a clean form of energy for transportation tasks (Lucentini *et al.,* 2021). Further benefits of ammonia decomposition are also the potentially favourable approaches to the mitigation of ammonia waste in the environment and these can help to achieve hydrogen fuel with a zero-carbon requirement which may eventually lead to zero carbon footprints (Le *et al.,* 2021). The process of hydrogen production from ammonia compounds is discussed through the following reactions such as reaction Eq. (1) which shows NH_3 decomposition for H_2 synthesis and this was found to be acceptable as a systematic reaction mechanism for the ammonia decomposition process:

$$2NH_3\,(g) \leftrightarrow N_2\,(g) + H_2\,(g) \text{ at } H_0\text{: } 46.22 \text{ kJ mol}^{-1}. \qquad (1)$$

Table 1. Ammonia decomposition for generation of hydrogen with removal of ammonia concentration from different sources

Conversion technique	Process parameter	Hydrogen generation	Reference
Thermal decomposition of ammonia	Without the presence of a catalyst	H_2 generation from NH_3 decomposition occurred in presence of a catalyst at reduced temperature value	(Lucentini *et al.,* 2021)
Ammonia decomposition at pressures other than 1 bar	Pressure up to 1 shows more decomposition than at 4 bar with 800ºC	H_2 generation at high temperatures and low pressures NH_3 decomposition	(Ristig *et al.,* 2022)
Decomposition of ammonia with electric current with 22 % conversion at 550ºC	Electric field over cerium (Ce)-based Materials like Fe/ Ru- deposited CeO_2 (1%)	Decomposition of ammonia into hydrogen and nitrogen with catalyst $CePO_4$ and Sr-doped $CePO_4$, and $CeZrO_4$	(Maslova *et al.,* 2023)
Decomposition ammonia with an electron beam	Background gases, absorbed dose, relative humidity and initial ammonia concentration	Decomposition of NH_3 for H_2 generation is reported	(Son *et al.,* 2013)
Decomposition of ammonia with an ion beam	Vanadium and niobium nitride cluster cations	Simple adsorption of NH_3 and adsorption of decomposed for hydrogen production	(Hirabayashi and Ichihashi, 2016)
Microwave decomposition of ammonia	Nickel based catalysts, mesoporous carbon with different supports	In microwave reactor system, 99% conversion was achieved to produce COx-free hydrogen with Ni/ Alumina at 400°C	(Seyfeli and Varisli, 2022)
Amminia decomposition with plasma technologies	Effect of gliding discharge plasma on the ammonia decomposition reaction detected	Lowest energy consumption and the highest reaction rates of NH_3 decomposition (17.8%) for H_2 generation/ storage	(Młotek *et al.,* 2021)
Ammonia borane (AB) decomposition coupled with thermolytic reaction	Polymeric coupling reaction between $–BH_3$ and $–NH_3$ sites of multiple AB molecules	Generation of hydrogen with suppression of by-products	(Roy *et al.,* 2018)
Electrolysis of liquid NH_3 for decomposition	The metal amides used as supporting electrolytes	H_2 gas is generated with high hydrogen capacity (17.8 mass %). This catalyst dissolve the amide ion in liquid ammonia	(Hanada *et al.,* 2010)
Photocatalysis in gaseous or aqueous medium of NH_3 for decomposition	Metal-loaded photo- catalysts, TiO_2, ZnO, C_3N_4, graphene	H_2 generation from heterogeneous nanostructures for photocatalytic ammonia decomposition process	(Zhang *et al.,* 2020)
NH_3 decomposition with mechanochemical methods	$SrTiO_3$ and $BaTiO_3$ powder ie needed for ammonia decomposition	Generation of H_2 and N_2 gas from mechanically milled under ammonia gas at room temperature	(Paik *et al*., 2010)
Reaction of NH_3 with hydrides	NH_3 can react with alkali metal hydrides	Generate of H_2 even at room temperature and 1 MPa of pressure is reported	(Miyaoka *et al*., 2011]
Decomposition of NH_3 in gasification atmospheres	Cu_2pa/2 catalyst with Ag and alumina at low temperature, (200°C) is used	Decomposition of odorous ammonia is reported for hydrogen production	(Lee *et al.,* 2015)
Decomposition of ammonia in the presence of H_2S	At simulated sludge drying waste gas by a novel non-thermal plasma	Maximum removal efficiencies obtained at the applied voltage (11 kV) and gas velocity (4.72 m s^{-1}) with support to H_2 synthesis	(Lu *et al.,* 2014)
Decomposition ammonia in the presence of oxygen	Tuning the Support Properties of Ni/GdxCe1-xO2-δ at 600°C	H_2 production rate (2008.9 mmol g^{-1} h^{-1}) reported with minimal decrease over 150 h	(He *et al.,* 2023)
Decomposition of ammonia in wastewater	Materials such as Ni, Co, La, and other perovskite catalysts used for this task	Extracting/ generating of hydrogen from ammonia by integration with green and economic technologies	(Yousefi Rizi and Shin, 2022)
Decomposition of ammonia in the presence of water vapor	The positive influence of water vapor on NH_3 decomposition with the rate of iron nitriding found	For H_2 generation by retardation of nitrogen molecules recombination on the iron surface and O_2 atoms	(Arabczyk *et al*., 2005)

Furthermore this can be explained as: the adsorption of ammonia at the catalyst surface; the successive cleavage of the N-H bond on adsorbed NH_3 for hydrogen release; this is followed by the recombinative desorption of N and H atoms to form gaseous N and H molecules (Lee and Park, 2022):

$NH_3{}^{+}{*} \rightarrow NH_3{*}$,
$NH_3{*} \rightarrow NH_2{*} + H{*}$,
$NH_2{*} \rightarrow NH{*} + H{*}$,
$NH_2{*} + {*} \rightarrow N{*} + H{*}$,
$N{*} + N{*} \rightarrow N_2$,
$H{*} + H{*} \rightarrow H_2$.

The reaction mechanisms* above, demonstrate the effectiveness of the active site on the catalyst and in this reaction, the activity of Ru metal, as the metal catalyst was found to be both suitable and effective for the ammonia decomposition process. This catalyst is reported to be present in more active sites for different types of reactions or reaction steps than other metals (Lucentini *et al.,* 2021; Le *et al.,* 2021; Lee and Park, 2022). Figure 3 discusses the different approaches of biological and chemical modes for product utilization in the promotion of hydrogen production. The Ru metal-based catalyst is recognized as an optimal choice for structure-sensitive reactions for NH_3 decomposition processes. Moreover, the advantage of this metal catalyst lies in the appropriate selection of the support medium and this is crucial for the supported metal catalyst as its surface acidity and alkaline nature are vitally important. Also, surface oxygen vacancies, redox properties and metal support interactions are also advantages. These properties can increase the performance of the catalyst for the NH_3 decomposition process (Le *et al.,* 2021; Lee and Park, 2022).

In the process of hydrogen generation, Ru metal is reported to depend on some supports like activated carbon and then ammonia decomposition occurs with the aid of Ru/SiO_2 (silica catalyst ~SC). This catalyst is supported on SiO_2 supports with different surface areas and it can be obtained through the application of varying calcination temperatures (Cechetto *et al.,* 2021). Further studies were carried out concerning the influence of Ru particle size on catalytic activity, the results were applied to NH_3 decomposition with an examination of the Ru/SiO_2 (catalyst ~C), being conducted at different temperatures (Gallucci *et al.,* 2017). Several reports discussed Ru-silica catalysts like Ru/SiO_2 (SC-700), Ru/SiO_2 (SC-800), and also Ru/SiO_2 (SC-900), these were generated and their activities with regard to ammonia decomposition were found to be similar to Ru/

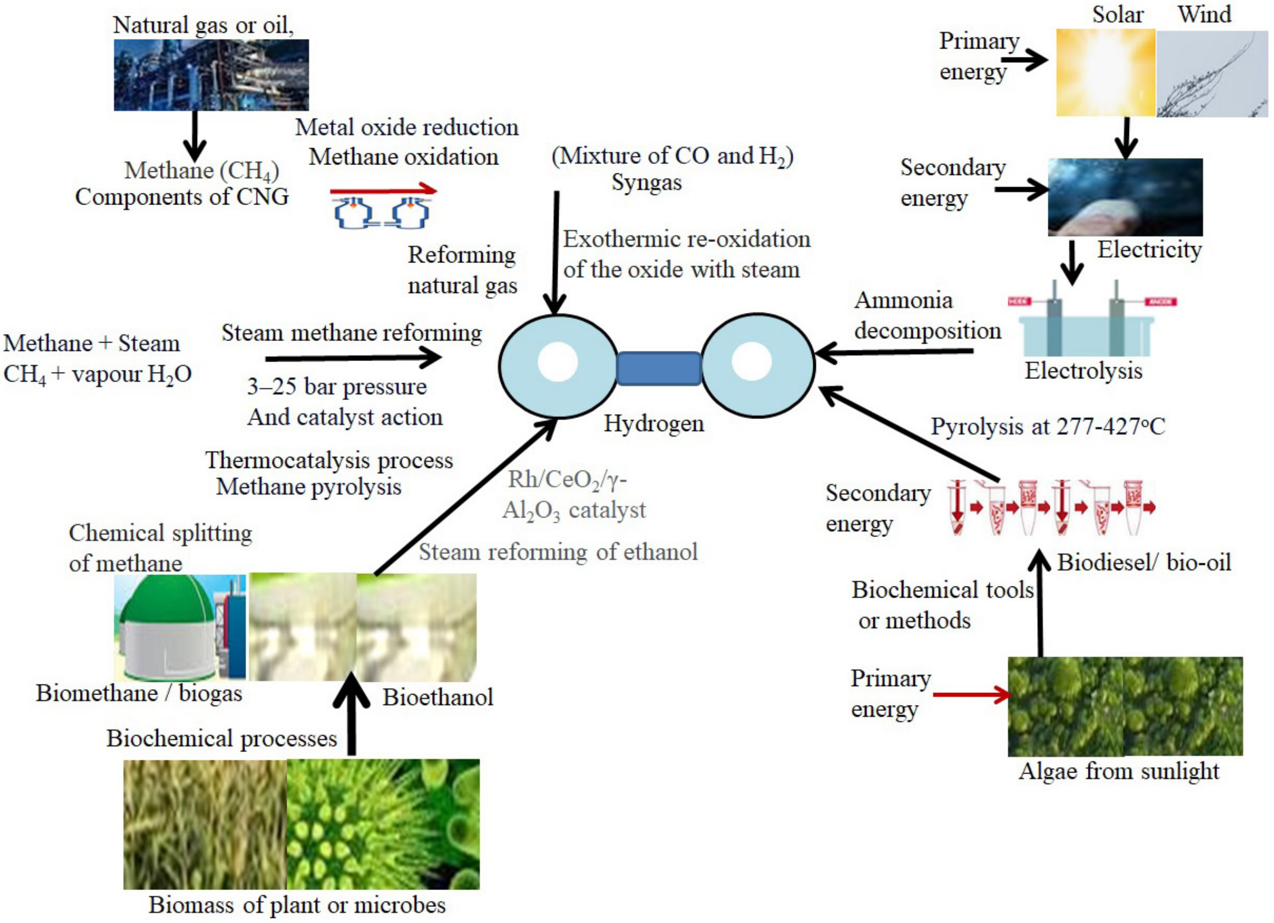

Fig. 3. Different approaches to biological/chemical modes for product utilization for the promotion of hydrogen production.

SiO_2 (SC-100) at the same temperature. Also, Ru/SiO_2 (SC-900) exists in the form of Ru nanoparticles with an Av. size of 3.8 nm but there were relatively large Ru lumps as well (Cechetto *et al.,* 2021; Gallucci *et al.,* 2017).

Ammonia decomposition can be facilitated by studying the effects of temperature and pressure and then assessing the catalytic activities of certain elements and these (like Hg, Fe and Pt) are commonly used in ammonia decomposition for hydrogen production (Lendzion-Bieluń and Arabczyk, 2013; Kim and Park, 2023). Further research studies were carried out on interesting reactions for different industrial applications to facilitate ammonia degradation (Ji *et al*., 2011, Kim and Park, 2023). Later in 1934, several scientists proposed that this ammonia decomposition for hydrogen generation should take place at high pressure (in the range of 7 to 14 bar). This was coupled with a residual NH_3 scrubber that used to make harden oils process. Some efforts were made to develop the technology of NH_3 crackers for different processes, they involved systematic study and setting precedents with regard to the metallurgical industry and its ability to reduce and temper metals (Raróg-Pilecka *et al.,* 2003; Kim and Park, 2023). Also, with regard to ammonia decomposition in the context of hydrogen production, it was found that the effects of pressure at low value ranges were more favourable to both processes. Several researchers have conducted investigations into the reaction rates under conditions ranging from reduced pressures to ultra-high vacuum conditions in the presence of certain catalysts like Pt (platinum), Ni (nickel), Rh (rhodium), Ta (tantalum), W (tungsten) and Ir (iridium) (Paik *et al.,* 2010). The impact of high pressures on ammonia decomposition with generating of hydrogen fuel is reported and it was gone for proper examination of this effect. And then this hydrogen is gone to compressed form during its supply task and it can be used in fuel cells development (Inokawa *et al.,* 2015; Miyaoka *et al*., 2011). Figure 4 discusses the natural routes of energy capture which are utilized in electricity production with the induction of ammonia degradation/decomposition for the purposes of hydrogen synthesis.

In normal circumstances it was recommended to avoid the compression of hydrogen gas. H_2 was generated from ammonia decomposition and then the process was systematically evaluated directly at high pressures of up to 40 bar using a Ru/CaO catalyst. And it was subsequently promoted by applying high temperatures (K). and high pressures (between the 1 and 2 bar range) at the Ru/Al_2O_3 catalyst. It was also evaluated and the ammonia conversion performance was found to decrease with increasing pressures (Sayas *et al.,* 2020; Okura *et al.,* 2019). Some research studies were conducted to investigate the catalytic performance of thermal decomposition using alternative methods. These methods can be used to provide the activation energy that is necessary for the decomposition reactions (Paik *et al.,*

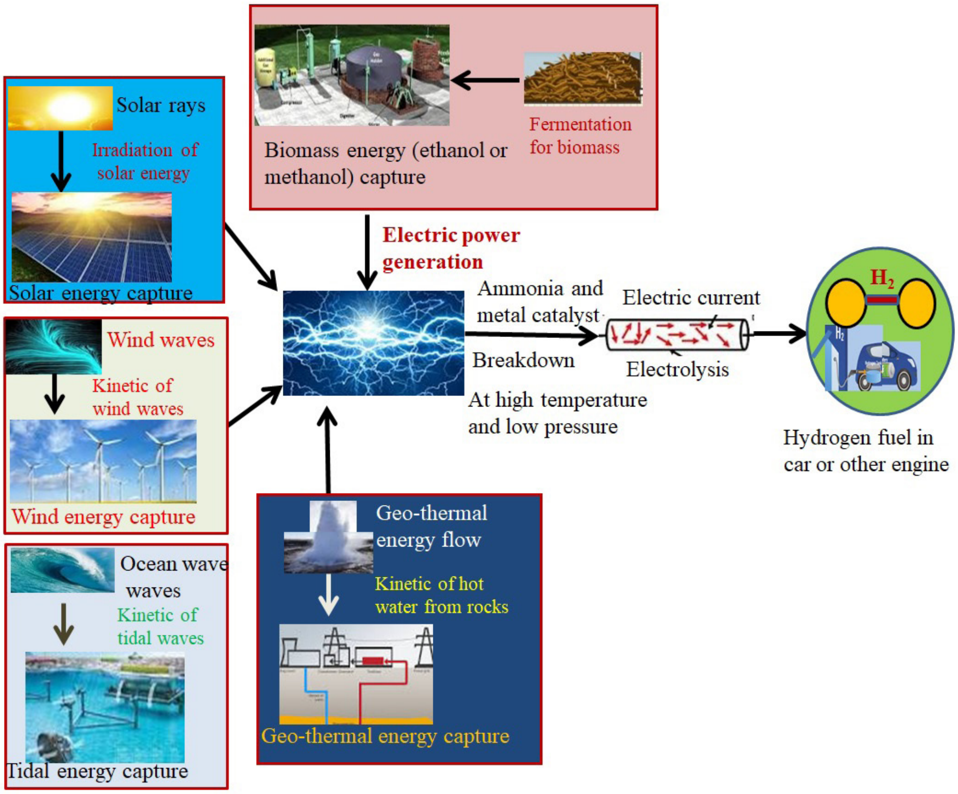

Fig. 4. Natural routes of energy capture processes used in power generation processes using electrical induction in ammonia decomposition/decomposition for hydrogen synthesis.

2010; Sayas *et al.,* 2020). In addition, studies were conducted concerning the impact of the application of electric currents, electron beams or ions, microwaves, plasma or solar energy in order to assist ammonia decomposition process. In several studies, an integrated system for ammonia decomposition was discussed, this would be coupled with other parallel exothermic reactions such as the combustion of propane or butane. These technologies can be applied with and also without any catalytic activities (Mukherjee *et al.,* 2018; Okura *et al.,* 2019).

Some approaches like the electrolysis of ammonia, and also photocatalysis, and mechanochemical methods have been established as the main approaches for the decomposition of ammonia. Some other approaches, such as the presence of compounds such as H_2S, oxygen and water, have also been used for ammonia decomposition, which produces hydrogen (Yuzawa *et al.,* 2012; Lee and Park, 2022). Table 2 discusses the many approaches and the relevant process conditions for achieving a substantial hydrogen yield *via* the ammonia decomposition process.

5. CONCLUSIONS

This review has discussed the different sources of ammonia generation including natural sources such as (soil or wastewater), and synthetic sources (these include chemical fertilizers like urea addition in crops), some portion of which can decompose to ammonia. Different sources of ammonia are available in the environment for various applications especially for chemical fertilizer synthesis tasks and also energy storage tasks (like H_2 fuel). In the

Table 2. Catalytic reaction based on different catalysts for hydrogen generation from ammonia conversion at various sources in environment that reduced ammonia concentration

Catalyst wt % with supports	% NH_3 inlet flow and WHSV (ml g^{-1} h^{-1})	NH_3 conversion at temperature into H_2	Reference
Ru (5.1%) metal with N-CNFs	5 and 9 900 respectively	450°C and 86% of NH_3 conversion reported for H_2 synthesis with sintering under the reaction conditions and on the electron density of the reduced metal	(Marco *et al.,* 2013)
Ru (5%) with Pr_6O_{11} and BaO promoter	100 and 3 000 respectively	350°C and 20% of NH_3 conversion reported for H_2 with strong basic oxides as an effective promoters over Ru/Pr_6O_{11}	(Nagaoka *et al.,* 2014)
Ru (5%) with Pr_6O_{11} and CaO promoter	100 and 3 000 respectively	350°C and 19% of NH_3 conversion reported for H_2 synthesis with effects of nitrogen doping on the structure of carbon nanotubes	(Nagaoka *et al.,* 2014, Chen *et al.,* 2010)
Ru (5%) with red mud supports	100 and 60 000 respectively	550°C and 17% of NH_3 conversion reported for H_2 synthesis with industrial-Waste-Supported Ru Catalysts	(Ng *et al.,* 2007; Hong *et al.,* 2021)
Ru (4.9%) with Sepiolite supports and K promoter	100 and 9 000 respectively	400°C and 47% of NH_3 conversion reported for H_2 synthesis with high pressure and help to decrease in the reaction apparent activation energy	(Sayas *et al.,* 2020)
Iridium~(Ir 10%) with SiO_2 supports	100 and 30 000 respectively	450°C and 8% of NH_3 conversion reported for H_2 synthesis with fuel cell applications and it contains supported Ru and Ni catalysts with different activity	(Choudhary *et al.,* 2001, Han *et al.,* 2023)
Ni (5%) SBA-15 supports with K promotion	100 and 30 000 respectively	500°C and 8% of NH_3 conversion reported for H_2 synthesis with Ru and Ni catalysts	(Li *et al.,* 2005, Leung *et al.,* 2023)
Ni (40%) with TiO_2 supports	100 and 6 000 respectively	550°C and 31% of NH_3 conversion reported for H_2 synthesis with high activity of Ni/$SrZrO_3$ and Ni/$BaZrO_3$	(Okura *et al.*, 2018)
Fe (12.4%) with La supports	100 and 18 000 respectively	500°C and 11% of NH_3 conversion reported for H_2 synthesis with Fe_2N and metallic Co based catalysts	(Xun *et al.,* 2017)
Fe (3.5%) with mica sopport	100 and 6 500 respectively	600°C and 85% of NH_3 conversion reported for H_2 synthesis with highly active and stable catalysts	(Duan *et al.,* 2011)
Co (7.0%) with MSC-30 support and Cs promoter	33 and 5 200 respectively	450°C and 12% of NH_3 conversion reported for H_2 synthesis with increase of the graphitisation degree of the support and the addition of electron donating promoters	(Torrente-Murciano *et al.,* 2017)

environment, an excessive concentration of ammonia has the potential to create health issues for children and older people. Some properties of ammonia are discussed, such as skin and eye irritation, with its solubility in water creating an alkaline condition. In this review, we discussed the different approaches to ammonia decomposition that can reduce its level/concentration in the environment while integrating with clean energy generation of a sustainable nature. Some approaches to hydrogen synthesis include ammonia electrolysis, photocatalysis and also thermocatalysts, these are discussed in more detail along with the parameters that favour ammonia decomposition including pressure and temperature impacts. This review also focuses on effective photocatalyst–assisted ammonia decomposition which facilitates hydrogen production in a sustainable way. These catalysts have potential in terms of efficient decomposition and also in terms of a high yield of hydrogen. There are some unique points in this review. Ammonia decomposition assisted hydrogen generation can contribute to sustainable energy generation with the mitigation of ammonia concentrations in the environment. Some natural resources like the ocean have proven to be favourable ammonia sources which may be their utilized in power production.

This review explores further information concerning ammonia decomposition with clean fuel development such as hydrogen. Furthermore the development of this fuel development may serve to reduce the carbon-sequestering process in our environment which in turn can promote a green environment and good health for everyone.

Conflicts of Interest: The authors declare that they have no Conflict of Interest.

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