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AMMONIA-MEDIATED HYDROGEN POWER SYSTEM FOR CLEAN ENERGY TRANSITION

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ABSTRACT

Renewable energy-based technological applications are gaining importance for conserving natural resources and mitigating the negative effects of human activity on the environment. As a clean, sustainable energy vector, Hydrogen (H_2) has been playing a key role. However, due to its highly flammable nature and low volumetric energy density, H_2 storage and utilization systems for clean energy applications have been hindered by safety concerns and low energy density. Recently, ammonia (NH_3), a carbon-free H_2 carrier, is emerging as a promising zero-carbon, clean energy alternative due to its high H_2 density, easier liquefaction, and safer storage and transportation. This work focuses on establishing the NH_3-mediated hydrogen energy system for practical power applications. The system decomposes NH_3 to create H_2 and drive fuel cells to generate electricity. We conducted catalyst screening for high-efficiency NH_3 decomposition and practical scale performance to balance the flow-pressure relationship and analyzed the effect of hydrogen purity on the end-use efficiency. This zero-carbon energy system shows potential to meet heavy-duty energy demands in the power industry and align with the 2050 carbon neutrality goal.

INTRODUCTION

The global energy sector is the primary source of greenhouse gas emissions contributing approximately 75% of the total emission (Zhang et al., 2023). This high carbon intensity is mainly due to the dependence on fossil fuels, such as coal-fired and gas-fired power plants. Therefore, reducing emissions from electricity generation is crucial for achieving carbon neutrality. Renewable energy-based practical power applications have gained significant focus to reduce carbon emission. However, one of the major challenges with renewable energy sources, such as wind and solar power, is their intermittent nature. When there is insufficient wind or sunlight, these sources cannot generate electricity, which means that backup power is necessary. Conventional battery technology is not a sustainable or cost-effective solution due to environmental concerns and resource limitations. As a clean, sustainable energy vector, hydrogen has been playing a key role in the storage and transportation of renewable energy. Hydrogen, in the gaseous and liquid states, has a high-energy density of 142 MJ kg^{-1} , which is about 2.5 to 3 times higher than that of natural gas and diesel (Mazloomi and Gomes, 2012). However, H_2 is highly flammable and has a very low volumetric energy density of 2.97 Wh L^{-1} at 0 °C, 1 atm(Cha et al., 2018). Although hydrogen has been considered a potential energy storage solution for many decades, the costly and technology-intensive infrastructure required for hydrogen transportation and storage has hindered its widespread adoption (Tzimas and European Commission. Joint Research Centre. Directorate-General., 2003; Züttel, 2003). Thus, alternative H_2 carriers are indispensable to achieving cost-effective and efficient H_2 storage and transportation.

Ammonia-based hydrogen energy storage systems, on the other hand, are emerging as a promising alternative because ammonia provides high H_2 density (17.8 wt%), more straightforward liquefaction, and safer storage. The volumetric energy density of NH_3 is as high as 2916.7 Wh L^{-1} at room temperature and at low pressure of ca. 8 bar, which is more than double

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that of compressed H_2 at 700 bar (1388.9 Wh L^{-1} (Jiao and Xu, 2019). Many new energy policies in developed countries (Akhtar and Liu, 2021; Fúnez Guerra et al., 2020; Ishimoto et al., 2020) involve the use of ammonia, as it can be generated using renewable energy sources and is a high hydrogen- density liquid fuel that can be stored and transported easily with well-established infrastructure. For example, Japan's Basic Energy Plan(Watanabe, 2022), Germany's National Hydrogen Strategy(*The National Hydrogen Strategy*, 2020), The European Union's Green Deal("A European Green Deal," 2019.), and The US Department of Energy's H_2@Scale program(*U.S. Department of Energy*, *Emerging Hydrogen Markets*, 2021). Renewable energy-based power generation systems can be integrated with green ammonia production for transportable and stable energy storage medium, such as the recently launched H2RES project in Denmark ("Skovgaard Energy," 2022), The Western Green Energy Hub in Australia ("Western Green Energy Hub," 2022), and The Fertilizer Corporation of India's green ammonia project ("Ammonia Energy Association," 2023). The green ammonia produced can be exported to markets and transported to end users. The technology to convert ammonia to hydrogen and electricity needs to be fully developed and optimized to decarbonize the electrification sector. Ammonia-based hydrogen systems have the potential to provide reliable and sustainable energy delivery solutions, which can be run in the concept of a microgrid where a localized energy system that can operate autonomously or in parallel with the main grid and includes a combination of distributed energy resources such as solar panels and wind turbines. Ammonia-based hydrogen systems can be used to power a single building, a community, or a small area, and are designed to ensure energy resilience, reliability, and security in the event of a grid outage or disruption, or for remote or off-grid locations where traditional power infrastructure is not readily available.

Our proposed system has a primary objective to promote a carbon-free fuel cell energy solution without the need of handling hydrogen directly. This work demonstrates an ammonia-powered electricity supply system for practical power applications. The main concept is to use ammonia as a high energy density carrier, delivered to the end using point and decompose into hydrogen on demand. The decomposed gas feeds a proton exchange membrane (PEM) fuel cell and is converted into electricity which can be applied as a power source. This quasi-direct ammonia fuel cell system is cost-effective and ideal for use in various industrial applications, such as backup power, rural electrification, and automotive industries. The use of ammonia-based hydrogen systems can help to reduce greenhouse gas emissions from electricity generation, while also providing a sustainable and cost-effective energy storage solution. This technology has the potential to transform the energy sector, enabling a shift to a more sustainable and resilient energy system that can meet the growing demand for electricity while reducing the impact on the environment. The rest of the paper includes detailed methodology, system design and implementation process and a detailed discussion on the obtained results, in the order stated.

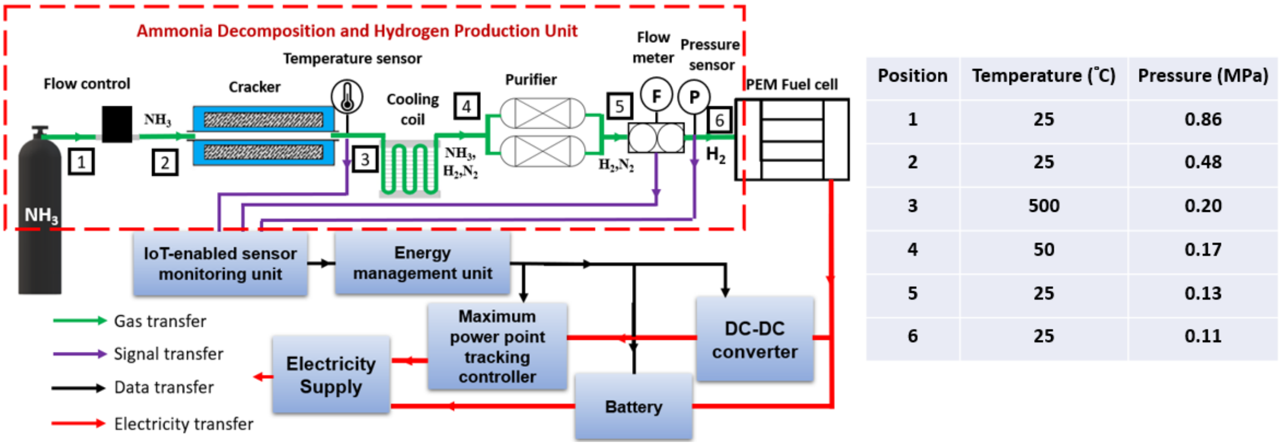
METHODOLOGY

The primary goal of design and implementation of an Ammonia-mediated Hydrogen power system was achieved in several steps. - The first activity was to implement the key materials and system designs for ammonia decomposition, purification, and hydrogen production (500L/hr. to 5000 L/hr. H_2 on demand) to optimize the system's performance. -The second activity was electricity generation from zero-carbon ammonia-powered H_2 fuel cell with high efficiency and long system lifespan. Our developed system can deliver uninterrupted electricity at power output from 500 W to 5000 W. - The third activity involved the development of a dynamic energy management unit (EMU) which facilitates the inclusion of a battery as an energy buffer for power storage and distribution, as well as efficient management of the power supply employing the maximum power point tracking (MPPT) principle. The final step was to design and implement a data acquisition and management system that is capable of logging real-time sensor data to assess the operation of the overall system.

Figure 1 demonstrates the overall system design and methodology. The ammonia cracker is a reactor that uses a catalyst to decompose the ammonia gas into hydrogen and nitrogen gases. The catalyst used in the ammonia cracker is typically a metal such as nickel or ruthenium, which helps to promote the decomposition reaction. The ammonia cracker operates at around 700°C and is designed to maximize the conversion of ammonia gas into hydrogen gas according to the following reaction:

$$2NH_3 \xrightarrow{Catalyst,heat} N_2 + 3H_2 \qquad (1)$$

As per **equation (1),** the decomposed gas at the output of the cracker contains 25% N_2 and 75% H_2 (v:v) , but it may still contain traces of ammonia gas based on the conversion efficiency. To remove these traces of ammonia, the hydrogen gas is purified using a zeolite-based ammonia purifier. The purifier is a Temperature Swing Adsorption (TSA) device. Zeolite porous materials, filled in the purifier columns, can selectively adsorb ammonia molecules(Ouyang et al., 2021). The TSA maximum capacity is 12 m^3/hr and provides outlet ammonia concentration of 0.1 ppm. The ammonia molecules are adsorbed into the zeolite, while the hydrogen gas passes through and is collected for use in the fuel cell. The outlet of the TSA device consists of 75% H_2 and 25% N_2 (v:v) (mixed gas).



Position	Temperature (˚C)	Pressure (MPa)
1	25	0.86
2	25	0.48
3	500	0.20
4	50	0.17
5	25	0.13
6	25	0.11

Figure 1 Ammonia-mediated Hydrogen fuel cell-based power generation system

The mixed gas (75% H_2 and 25% N_2) (v:v) is then fed to a PEM fuel cell, which reacts with oxygen to produce electricity. The fuel cell consists of an anode, a cathode, and an electrolyte membrane. The hydrogen gas is fed into the anode and split into protons and electrons. The protons pass through the electrolyte membrane to the cathode, while the electrons flow through an external circuit, producing an electrical current. At the cathode, the protons and electrons combine with oxygen to produce water and heat. A battery is used as an energy buffer for power storage and distribution. Energy management unit manages and controls the smooth power supply to a suitable load using MPPT. An Arduino Mega 2560-based data acquisition unit has been designed and implemented. This unit reads different real-time sensor data such as flow meter, temperature sensor, pressure sensors, gas leakage sensors etc. and displays in a suitable computer or mobile screen. This unit is also capable of logging sensor data which can be further used for system optimization. The sensor system helps to detect any abnormal operation and take necessary safety measures if required.

SYSTEM DESIGN AND IMPLEMENTATION

This study presents a CO_X-free power generator fuelled by liquid ammonia, with a system that includes a liquid ammonia tank, a hydrogen generator loaded with highly active granulated catalysts, a tube furnace (cracker), a hydrogen purification column utilizing solid adsorbents, a system stabilizer, and a polymer electrolyte membrane fuel cell. The hydrogen generator demonstrated the ability to continuously supply high-purity hydrogen to the fuel cell, producing over 2.0 kWh of electrical energy without nitrogen separation. The system operates under CO_X-free conditions by excessive decomposition of ammonia, which significantly increases the overall system efficiency. This process allows for stable operation under dynamic power demand, as the excess reformate gas supplied to the fuel cell serves as a buffer. Finally, the system's efficiency was analysed in detail.

Upscaling our system from a lab scale to an industrial scale was the main challenging part. Apart from obtaining large quantities of noble metals, upscaling these catalysts to decompose ammonia may face several other challenges. One significant challenge is the need for a bulky reactor, which can be a drawback for mobile and stationary applications due to the growing costs of increasing reactor volumes. Moreover, the catalyst's stability and durability under high-temperature, high-pressure, and reactive environments must be considered, as these factors can affect its effectiveness and lifespan.

Catalyst Synthesis for Ammonia Decomposition

The promising method of ammonia decomposition for hydrogen production faces significant challenges related to upscaling catalyst synthesis, including heat transfer, pressure, and cost. This study evaluated a range of noble metals for ammonia decomposition to address these challenges and identified Ruthenium as the optimal catalyst based on its exceptional performance (Kishida et al., 2018; Szmigiel et al., 2004). However, upscaling the synthesis of the catalyst requires a significantly greater weight scale than that used in the lab scale, making traditional catalyst preparation methods such as Co-precipitation or Electrodeposition, time-consuming and ineffective. To overcome this issue, we screened various commercial chemical supports, including Fe_3O_4, MgO, Al_2O_3, and NiO, to identify optimal support materials for the catalyst. The screening process carefully evaluated the materials' chemical purity and physical properties to ensure consistency and reproducibility of the catalyst's properties. Ultimately, a simple catalyst preparation method called water impregnation was utilized, which involved dispersing the active catalyst components into the water and then drying and calcining the resulting powder. This efficient and scalable synthesis method can have practical applications in energy production and storage, contributing to developing more sustainable energy sources.

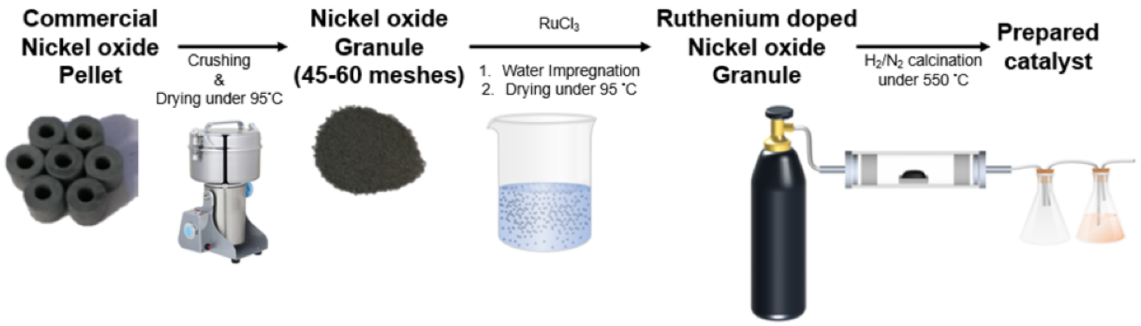


Figure 2 Catalyst synthesis method

The synthesis of Ruthenium (Ru)-based catalysts involved several steps, starting with crushing commercial Nickel Oxide catalysts into 45-60 mesh sizes using a crusher. The crushed powder was then dried in a thermostatic oven at a constant temperature of 95°C for 60 minutes. The dried Nickel oxide catalyst was weighed, and the mass of Ruthenium (III) chloride was calculated based on a specific equation. Next, Ruthenium (III) chloride powder was added into a beaker with a certain amount of deionized water, and the mixture was stirred while the dried Nickel oxide granules are added. This method is known as water impregnation method. This process was repeated at least twice, and the resulting solution was then filtered using a Büchner funnel to obtain the Ru-doped Nickel oxide granules. The granules were then dried in a thermostatic oven at a constant temperature of 95°C for at least 120 minutes to remove excess solvent. Then the dried sample was transferred into a crumble boat and placed inside a tube furnace under a hydrogen atmosphere, heated according to a specific heating program to 550°C for 4 hours, including a room temperature, heating, and cooling phase. Finally, the sample was sealed in a glass container to prevent reoxidation. The cracker (tube furnace) generates the high temperature required for the NH_3 decomposition reaction and requires single phase ac supply. Also, a cooling coil is placed between the cracker and the purifier to cool down the mixed gas from 700˚C to 25 ˚C. The detail of catalyst synthesis method is demonstrated in **Figure 2**.

Reactor Design

A reactor for ammonia decomposition was designed based on catalyst activity tests. It consists of eleven channels constructed via 3D metal printing, forming a cylinder shape surrounded by a heat source to optimize heat transfer. The reactant enters a central inlet tube and is evenly distributed into the eleven parallel reactor tubes. To connect all eleven tubes into a single input and output line system, a primary multiway heat exchanger was designed, which combines all the outputs and provides primary heat exchange between a single inlet and outlet. The primary heat exchanger is manufactured with 316L stainless steel powder, resistant to NH_3 and H_2 corrosion. The schematic drawing of NH_3 decomposition reactor, tube furnace, and primary heat exchanger is shown in **Figure 3**. We have achieved 99.4% NH_3 conversion rate in our large scale 11 tube testing of the reactor. The details are given in the results and discussion section.

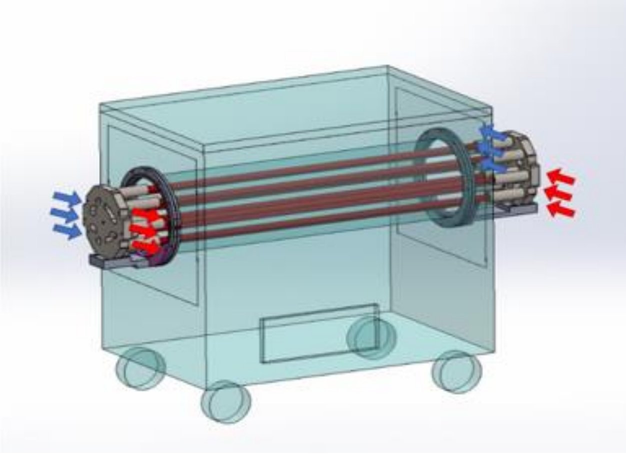


Figure 3 Schematic drawing of NH_3 decomposition reactor, tube furnace, and primary heat exchanger.

Purification System and Fuel Cell

According to ISO14687-2, the fuel gas feed must be <0.1 ppm of the unconverted NH_3 to prevent irreparable damage to a PEM fuel cell. Any residual NH_3 higher than this tolerance level will damage the acidic membrane by forming NH_4^+ ions. The degradation of the membrane significantly reduces proton conductivity and, in turn, deteriorates the efficiency and lifetime of the fuel cell. Zeolites are economically attractive materials with many advantages as NH_3 adsorbents, such as high adsorption capacity, robustness, and excellent regeneration stability. In our previous research (Zhai et al., 2023), 13X zeolite has been found suitable for removing residual NH_3 in the output gas. Literature precedents have also shown that commercial X-type zeolites could successfully reduce the dynamic adsorption of NH_3 to below 0.1 ppm. Therefore, 13 X zeolite absorbent was adopted for NH_3 removal in the TSA device of our system. Due to the long-term application of the consumer electronics charging station, the regeneration of 13X zeolite will need to be implemented in the system to realize a non-stop and highly efficient gas purification. We have tested our designed reactor with a 5-kW fuel cell. Our system can deliver 500 W to 5000 W electrical power based on the load. The fuel cell test results are described in the results and discussion section.

Energy Management Unit

An energy management unit has been developed to operate the ammonia-mediated hydrogen power generation system efficiently. The EMU will regulate the energy between the ammonia-cracked gas supply, membrane purification system, fuel cells, and a battery for extra energy storage according to the operating status of electrical load. The model of the EMU was first designed and studied via simulation approaches. The optimized operating points were then analyzed. This technique, called maximum power point tracking, can be applied to renewable energy supply systems. For example, energy management monitors or regulates parameters such as gas flow parameters (e.g., hydrogen gas composition, flow speed, or the like), electrical parameters, and environmental parameters to enable the power generation system to optimum operating conditions. **Figure 4** explains the function of EMU in our implemented system.

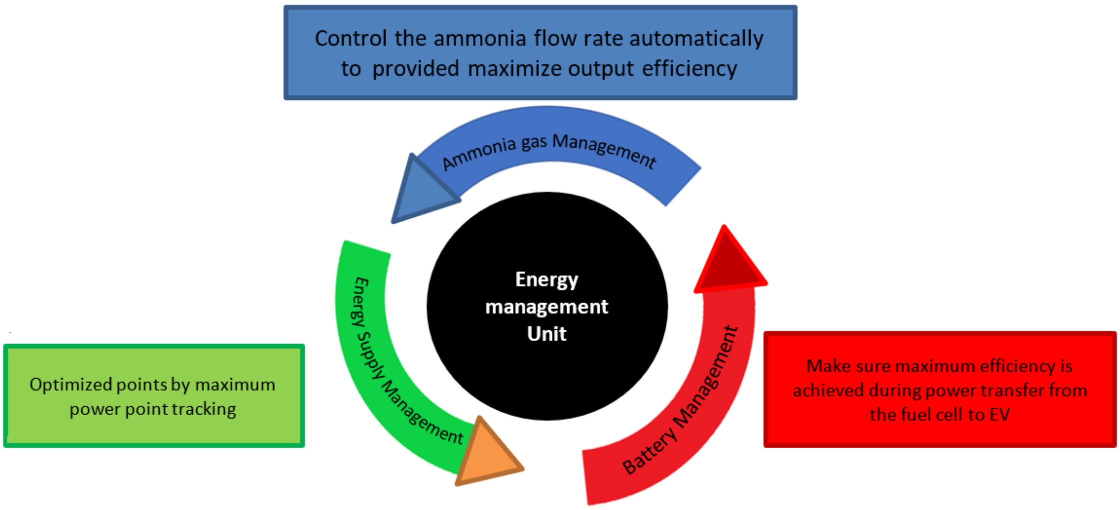
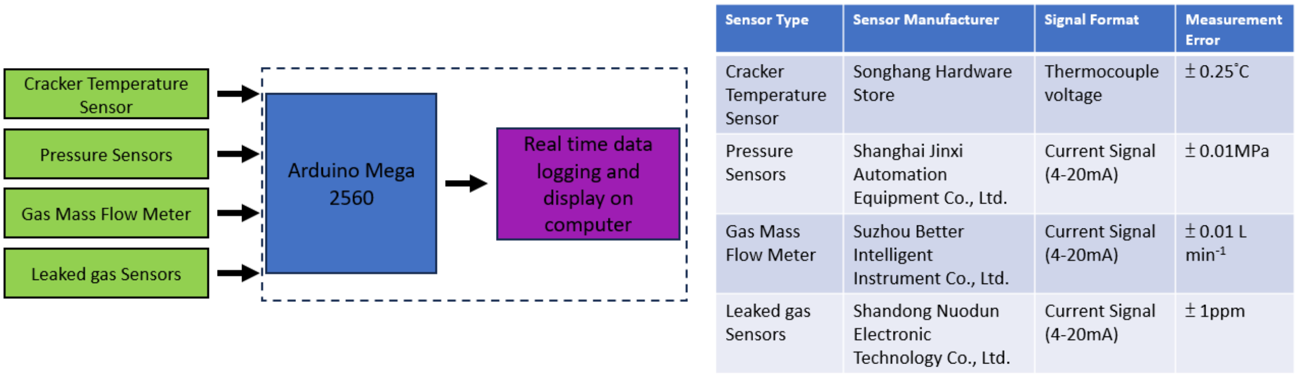


Figure 4 Role of energy management unit.

Data Acquisition and Sensor Unit

To ensure real-time system performance monitoring and optimization, a sensor unit with data logging capability has been designed and implemented. The basic outline of the unit is shown in **Figure 5**. To ensure the overall system can run at satisfactory efficiency, each subsystem/unit should be automatic monitored and adjusted according to various energy demands and environment changes to avoid energy waste or loss. For example, the gas pressure at different system stages should be optimum for a proper ammonia decomposition process. Furnace temperature needs to be controlled and kept at target temperatures (600~700ºC) to ensure the catalyst is at the best working condition. Therefore, it is essential that the system can continuously self-observe, and necessary actions can be taken in case of over-demanding or any unexpected situations to avoid low efficiency. To monitor the proper operating condition of the cracker system, the fuel cell, and the hydrogen consumption, a data acquisition and sensor unit has been integrated with the energy management unit. This unit is capable of logging data from the different sensors (temperature, pressure, gas compositions, leaked gas sensors and gas mass flow meter) into a computer that can be further analyzed for system optimization. The analog signals from the sensors are received and processed by Arduino Mega 2560 as per the user-defined program. This processing includes analog to digital conversion, signal conditioning, etc. Utilizing serial communication between Arduino and the computer, real-time sensor data is logged and saved in the computer. Overall, the energy management unit and a data acquisition and sensor unit will work together to ensure that the total system works in optimum operating conditions. The energy management unit also reads all sensor data and shuts down the system if any sensor values go beyond the safety range. The data is read every one second.



Sensor Type	Sensor Manufacturer	Signal Format	Measurement Error
Cracker Temperature Sensor	Songhang Hardware Store	Thermocouple voltage	±0.25°C
Pressure Sensors	Shanghai Jinxi Automation Equipment Co., Ltd.	Current Signal (4-20mA)	±0.01MPa
Gas Mass Flow Meter	Suzhou Better Intelligent Instrument Co., Ltd.	Current Signal (4-20mA)	±0.01 L min^{-1}
Leaked gas Sensors	Shandong Nuodun Electronic Technology Co., Ltd.	Current Signal (4-20mA)	±1ppm

Figure 5 Data acquisition and sensor unit.

RESULTS AND DISCUSSION

An ammonia decomposition test was performed in the 11-tube reactor to evaluate the effect of reaction temperature on the ammonia conversion rate. The test was conducted at two different ammonia flow rates: 25 L/min and 40 L/min. Various reaction temperatures were tested to determine the optimal temperature range for the ammonia decomposition reaction. The ammonia conversion rate was calculated using the following equation:

$$\mathrm{NH_3}\ conversion\ = \frac{1-[\mathrm{NH_3}]_{outlet}}{1+[\mathrm{NH_3}]_{outlet}} \times 100\% \qquad (2)$$

Here $[\mathrm{NH_3}]_{outlet}$ is the flow rate of ammonia at the outlet of the reactor. The ammonia conversion rate of the catalyst based on 25L/min and 40L/min inlet ammonia flow rate is analysed and shown in **Figure 6**.

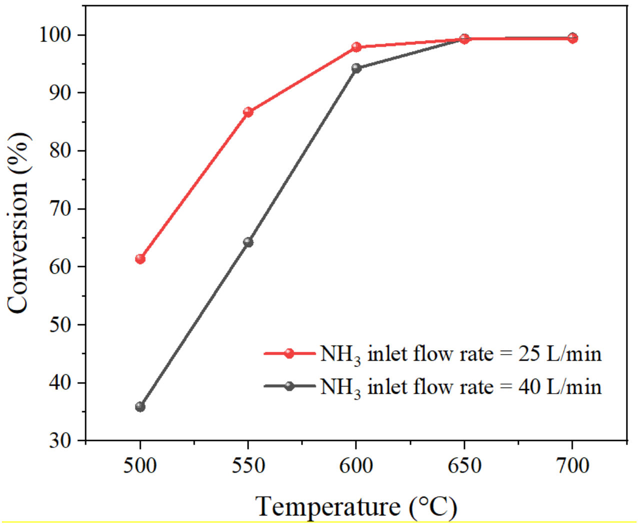


Figure 6 Relationship between ammonia conversion rate and reaction temperature under NH_3 flow rate = 25 L/min & 40 L/min

The findings in **Figure 6** demonstrate the impact of temperature and ammonia flow rate on the conversion rate of ammonia decomposition using a nickel oxide doped with a ruthenium catalyst. Notably, both experiments conducted at different ammonia flow rates indicated a remarkable decomposition rate exceeding 99.4% when the reaction temperature was sustained at 650 degrees Celsius. These outcomes serve as evidence that the reaction temperature and the ammonia flow rate significantly influence the rate of ammonia decomposition.

We have tested the 5-kW fuel cell under varying load (resistive) conditions in our lab. At least 80 L/min mixed gas (75% H_2 and 25% N_2) (v:v) is needed to be fed to the fuel cell to get the rated 5 kW electrical power output. Therefore, we have tested the fuel cell with a fixed 80 L/min mixed gas inlet under varying load conditions. An NH_3 flow rate of 40 L/min at the inlet of the reactor generates an 80/L min mixed gas at the outlet of the reactor which is then fed to the fuel cell after purification. The fuel cell test results conducted at our lab are demonstrated in **Figure 7**.

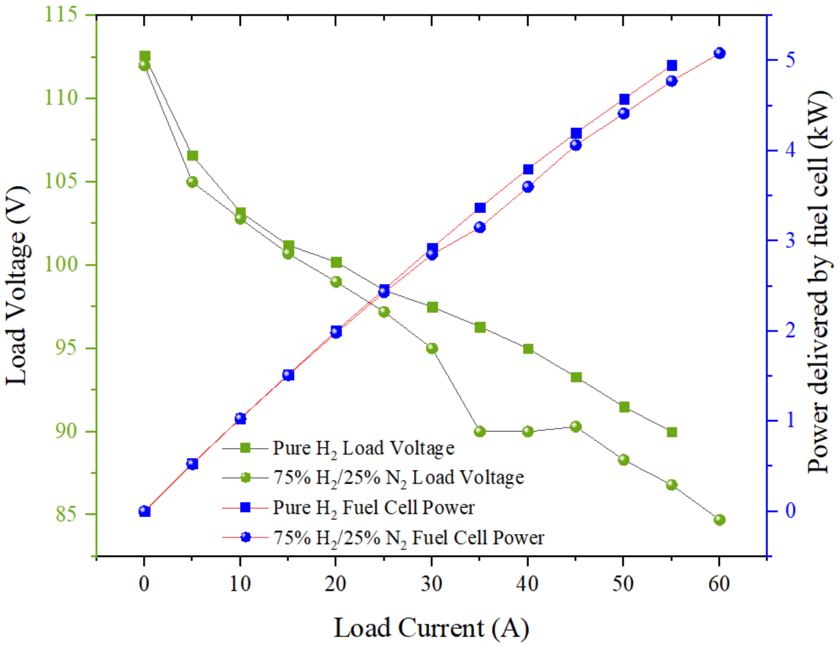


Figure 7 Output Voltage, current and power characteristics of the fuel cell tested at our lab.

It can be seen from **Figure 7** that as the load is increased, the current drawn from the fuel cell by load increases. Correspondingly, the power delivered by the fuel cell also increases. The fuel cell can provide up to 5 kW of electrical power output on demand. From figure 7 it can be observed that because of 25% N_2 (v:v) inlet, the power delivered by the PEMFC is slightly decreased when the load draws higher amount of current (>25 A) and the load voltage decreases more sharply compared to the pure H_2 case.

Carbon Reduction Estimation

Assuming “green ammonia” is applied as an ammonia source, which refers to the synthesis of ammonia using green energy, including wind and solar power, without carbon emissions. From the information provided by the United States Energy Information Administration, coal-based fuel will release 0.316 kg CO_2 when providing 1 kWh of energy (*U.S. Department of Energy, Emerging Hydrogen Markets*, 2021). Therefore, our 5-kW ammonia-mediated hydrogen fuel cell system can technically reduce 2,300 kg of CO_2 emission per year (12 hrs. per day) as compared to electricity production using the coal-fired power station. This analysis assumes that the heat required by the cracking system comes from renewable energy otherwise the CO2 emission would be much higher.

CONCLUSIONS

This project work has been designed and implemented towards scale-up and commercialization to drive the energy transition toward a zero-emission energy storage and supply system for practical power applications. The proposed ammonia-fuel cell power solution matches the global carbon neutrality target and aligns with the decarbonization strategies (green hydrogen, hydrogen fuel cell technology), giving it significant competitive advantages over the liquid fossil fuels electric generator (carbon intensive) as well as other fuel cell solutions (H_2 safety and storage issues). The public acceptability to hydrogen is not very high. This is mainly from the significant concerns about hydrogen safety. This ammonia-fuel cell power solution will be ideal for use in a wide range of commercial and industrial applications, such as backup power, rural electrification, and automotive industries, without the need of handling hydrogen directly. As for energy storage, non-explosive, zero-emission ammonia fuel possesses a high volumetric energy density. Ammonia’s well-established production, transportation, and storage infrastructure can provide a more practical next-generation power solution to accelerate the development of a “hydrogen society”. The use of ammonia-based hydrogen systems can help to reduce greenhouse gas emissions from electricity generation, while also providing a sustainable and cost-effective energy storage solution. This technology has the potential to transform the energy sector, enabling a shift to a more sustainable and resilient energy system that can meet the growing demand for electricity while reducing the impact on the environment. This paper is relevant to the goals of GPPS. We believe that the work carried out is significant and addresses a current issue such as reducing greenhouse gases emissions in electricity generation.

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