

# Potentials of Nano Catalysts for Energy Reduction During Synthesis of Ammonia

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**Abstract** – Ammonia (NH<sub>3</sub>) is one of the fundamental raw materials currently in use globally and is the second largest synthetic chemical product. But the production of NH<sub>3</sub> through the predominant Haber process consumes about 3-5% of the global natural gas produced per annum, approximately about 1-2% of the global energy supply. The energy consumption during the synthesis of NH<sub>3</sub> has immediate and long term effect on the global energy reserve and the environment. This paper highlights the potentials of using Nano catalysts during the synthesis of NH<sub>3</sub> with emphasis on reducing energy consumption and green synthesis options. The use of Nano catalysts during the various stages in the synthesis of NH<sub>3</sub> could reduce energy consumption and minimize harmful gas emission, while simultaneously improving the efficiency of the output and production

**Keywords** – Ammonia, Steam Reforming Process, Nano Catalysts, Green Synthesis.

## I. INTRODUCTION

### A. Overview

Ammonia is (NH<sub>3</sub>) a gas with stinky smell and is found natural in traces in the atmosphere and in small quantities in various forms [1]. Most of the NH<sub>3</sub> used for various applications are synthesized using three nitrogen-fixing processes as discovered from 1902-1913: electric arc process, calcium cyanide process and catalytic process [2]. Currently, more than 90% of the NH<sub>3</sub> consumed in the world is made using the catalytic process developed by Haber and Bosch [3] with modification for better yield but basically on the same concept.

It has been estimated that NH<sub>3</sub> is the second largest synthetic product [4] with global production of about 131 million tonnes in 2010 and a projection of up to 233 million tonnes by 2019 [5] – [7]. More than 83% of NH<sub>3</sub> synthesized is used in making fertilizers [3]. The produced fertilizers have increased crop yields by 400% and still using under 15% of the global total land area [6]. It has been estimated that nearly 80% of the N<sub>2</sub> located in human tissues stem from the Haber-Bosch process. Since N<sub>2</sub> efficiency is usually below 50%, the substantial usage of industrial N<sub>2</sub> fixation is harshly disturbing the overall global biological environment [5]. NH<sub>3</sub> has other widespread domestic and industrial applications [3], [6], [8]-[10].

### B. Ammonia Production Process

There are three (3) types of processes currently used for the production of NH<sub>3</sub>: steam reforming, partial oxidation and gasification of coal [11]. A typical modern NH<sub>3</sub> plant initially converts natural gas into H<sub>2</sub> and the method is

referred to as steam reforming [7]. The synthesized H<sub>2</sub> is then mixed with N<sub>2</sub> to generate NH<sub>3</sub> through the Haber-Bosch process. Beginning with a natural gas feedstock, the steps followed in generating H<sub>2</sub> are summarized below [3]:

- I. Converting sulphur compounds in sulphides:  
 $H_2 + RSH \rightarrow RH + H_2S(gas)$
- II. Absorption of H<sub>2</sub>S by zinc oxide beds :  
 $H_2S + ZnO \rightarrow ZnS + H_2O$
- III. Catalytic steam reforming to produce H<sub>2</sub>  
 $CH_4 + H_2O \rightarrow CO + 3H_2$
- IV. Catalytic shift conversion of CO:  
 $CO + H_2O \rightarrow CO_2 + H_2$
- V. Removal of CO<sub>2</sub> using solid adsorption media.
- VI. Removal of CO and CO<sub>2</sub> from H<sub>2</sub>  
 $CO + 3H_2 \rightarrow CH_4 + H_2O$   
 $CO_2 + 4H_2 \rightarrow CH_4 + 2H_2O$
- VII. Catalytic reaction of H<sub>2</sub> with nitrogen (developed from processed air) to form anhydrous liquid ammonia: ammonia synthesis loop or Haber-Bosch process represented by  
 $3H_2 + N_2 \rightarrow 2NH_3$

The flowchart for the production of NH<sub>3</sub> using steam reforming process is shown in Fig. 1. The H<sub>2</sub>-N<sub>2</sub> mix reacts with the help of iron catalyst at elevated temperature in the range of 400 - 500 °C and pressures between 30 - 100 bars. [3].

Global sales of NH<sub>3</sub> have been estimated to be above \$100 bn per annum, with about half of that wholly related to the energy expenditure [6]. Although the overall energy consumption required for production of NH<sub>3</sub> has been reduced from 45 GJ/ton to less than 29 GJ/ton [3], [11], the process is consuming about 8% of the world's energy through fossil fuel(s) and generated electricity [6], [11].

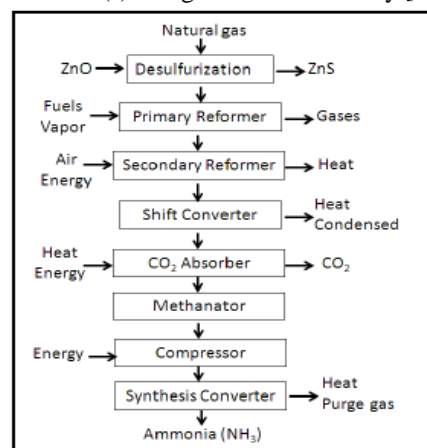


Fig. 1. Flowchart of Ammonia Production Using Steam Reforming Process [3]

Data obtained from a study has indicated that one-third of emissions from natural gas based  $\text{NH}_3$  production is due to fuel combustion and the balance from fuel based feedstock to obtain the  $\text{H}_2$  needed for the synthesis. Also additional energy cost is acquired for cleaning, capturing and storing/reuse of  $\text{CO}_2$  emissions. This paper highlights the effect of catalyst on the synthesis of ammonia with emphasis on using materials at Nano scale in order to reduce energy consumption for a sustainable production process and a green environment.

## II. CATALYSTS IN AMMONIA SYNTHESIS

### A. Types and Functions of Catalysts

Catalysts are substances that increase the rate of chemical reaction, but are not consumed or destroyed during the primary process. Catalysts are classified based on several factors. Thus catalysts may be homogenous or heterogeneous, solid or non-solid, single or multi-phase and supported or unsupported [12]. It has been estimated that about 90% of all commercial chemical products were made with catalyst(s) at one or more stages during their production process [13].

In relation to the synthesis of  $\text{NH}_3$ , catalysts are vital and inevitable as they determine and/or control operating

temperature, rate of gas flow and cooling requirement [8]. Catalysts also influence the funds of the overall process and need to fulfill the following conditions[3]:

1. Greater catalyst effect at the least possible reaction temperatures so as to utilize equilibrium at low temperatures;
2. Control the consequence of poisons for  $\text{O}_2$  and  $\text{Cl}$  contained in catalysts in order to produce purified  $\text{NH}_3$ ;
3. Long life in order to reduce cost related to removing, acquiring and replacing of catalysts;
4. Reliable mechanical strength is required to avoid inadequate pressure and other conditions that may lead to unnecessary upsurge in pressure fall and plant shut down.

The major steps involved in the synthesis of  $\text{NH}_3$  require catalysts to perform specific tasks. A summary of the catalysts required for an ammonia plant with capacity of 1,620 metric tons per day is shown in Table 1.

The eight catalysts indicated in Table 1 have been classified as either protective or economic, depending on their major function. The protective catalysts prevent or reduce degradation, while economic catalysts are accountable for the conversion of unprocessed materials and the yield of the products and have immediate influence on the finances of the whole plant [3], [11].

Table 1: Catalysts required in an Ammonia Plant with 1,620 tons/day capacity [11].

Process/Equipment	Catalyst(s)	Quantity (tons)	Life (years)	Function
Hydrogenator	Co, Mo, Al	10	9	Protective
Desulphurizer	ZnO	64	9	Protective
Primary Reformer	$\text{NiO}_2$	22	3	Economic
Secondary Reformer	$\text{NiO}_2$	40	8	Protective
High Temp. Shift	Fe, Cr	45	8	Economic
Low Temp. Shift	Cu, Zn, Al	66	3	Economic
Methanator	$\text{NiO}_2$	43	8	Protective
Ammonia Synthesis	Fe	203	8	Economic

## III. NANO-CATALYSIS

Nanocatalysis involves the usage of nanomaterials for a range of catalytic reactions, as illustrated by heterogeneous catalysis which is one of the oldest commercial applications of nanomaterials [10]. Nanomaterial-based catalysts are mostly heterogeneous catalysts that have been fragmented into metal nanoparticles so as to hasten up the catalytic procedure. Metal nanoparticles have a greater surface zone so there is enlarged catalytic action since extra catalytic reactions occur at the same time. Nanoparticle catalysts have the capability of being easily separated and reprocessed with greater preservation of catalytic capacity or rate than their bulk equivalents. These catalysts also carry out two other roles in catalytic practices: be the spot of catalysis or support for catalytic practices. They are usually manipulated in moderate conditions to avoid decay of the nanoparticles at extreme conditions [12], [13].

It should be noted that nanomaterials "describe (in principle) materials the single units of which is sized (in at

least one dimension) between 1 and 100 nanometers ( $10^{-9}$  meter). Nanomaterials are either organic or inorganic and occur in form of dots/particles, rods, wires, tubes, films etc. "[13]. Nanocatalysis have proven potentials in various areas due to its "complex/unusual" physiochemical properties that allow for minimal chemical waste, energy efficiency, reduced emission and optimum feedstock utilization [4], [13]. The global demand for nanomaterials used in catalytic applications is on the increase with nanocatalysts likely to triple their pooled share in the next five to ten years [13].

The active surface of a catalyst increases when the size is decreased due to higher ratio of surface-to-volume. It has also been proven that higher surface gives a better reaction. Particle size and molecular structure/distribution can be controlled using various methods of nano synthesis [7], [8], thus the use of nanocatalysts in various forms reduces energy consumption with higher yield [14].

A. Application of Green Chemistry and Technology in the Synthesis of Ammonia.

### 1) Green Chemistry

Green chemistry is the “utilization of a set of principles that reduces or eliminates the use or generation hazardous substances in the design, manufacture and application of chemical products” [15]. The concept of green chemistry is summarized in the professed 12 principles [13-16]:

1. Waste prevention is better than treatment or clean-up
2. Chemical synthesis should maximize the incorporation of all starting materials
3. Chemical synthesis ideally should use and generate non-hazardous substances
4. Chemical products should be designed to be nontoxic
5. Catalysts are superior to reagents
6. The use of auxiliaries should be minimized
7. Energy demands in chemical syntheses should be minimized
8. Raw materials increasingly should be renewable
9. Derivations should be minimized
10. Chemical products should break down into innocuous products
11. Chemical processes require better control
12. Substances should have minimum potential for accident

The green chemistry in synthesis of  $\text{NH}_3$  is based on catalysis by nanoscale particles, where by the amount of surface atoms and their effect are optimized leading to improved selectivity and/or yield.

#### 2) Reduction of Energy Requirement by Using Nanocatalyst

Ammonia has been produced for the past century, starting with electric arc process operating at temperatures above  $3000^\circ\text{C}$  and was greatly wasteful in relation to energy utilization [16]. The current *Haber-Bosch* method is based almost on the same concept, but there is improved energy efficiency to about 60% due to substitution of coal by oil, and afterwards natural gas as the desired feedstock [16]-[18]. The use of nanocatalysts is one of the means that led to reduction of energy consumption during the synthesis of  $\text{NH}_3$  as illustrated by several examples.

1. CERION Enterprises have produced nano-iron catalyst on Cerion oxide support with possibility to allow production of  $\text{NH}_3$  at far lesser pressures and temperatures, consequently allowing a likely 40-50% energy savings as against the present methods. In relation to this breakthrough CIBA/BASF, the world's top provider of  $\text{NH}_3$ , is presently in dialogue with Cerion Catalyst for integration of the catalyst into real industrial operations [19].
2. QuantumSphere has applied its nano Fe promoters to commercially synthesize  $\text{NH}_3$  with the catalysts having a particle size of 10-40 nm. Up to 40% increase in activity was achieved for a typical 1500 ton per day, with 5% energy savings and throughput increase of 20% [20]
3. It has recently been discovered that Ru/ $\text{ZrO}_2$  catalyst with size of Ru particles of 1.5-3.2 nm shows excellent output for ammonia synthesis especially under low temperature and pressure. The optimum reaction was obtained at 30 bars and  $425^\circ\text{C}$ , which saves energy and making the process more efficient [21].

#### 3) Synthesis of Ammonia at Ambient Conditions.

The energy requirement for the synthesis of  $\text{NH}_3$  could be minimized by using processes that occur at ambient temperature and pressure with minimal negative effect on the environment. As part of the ardent strive toward achieving this novel and challenging task, a magnetically induced reaction at 0.2 tesla was used to synthesize  $\text{NH}_3$  at  $25^\circ\text{C}$  and normal pressure using nanocatalysts with higher output [21]. This is part of the *OneBaja* Research Program in Malaysia comprising of 7 different projects with the overall aim of producing a new  $\text{N}_2$ -based, enhanced and efficient fertilizer on least energy intake. By estimation from studies, about “1000 tonnes  $\text{NH}_3$  production per year, the new process will be to provide total carbon emission offset amounting to 750,000 tonnes”. Furthermore, the proposed well-ordered release fertilizer may likely affect carbon offset by about “9.9 million metric tonnes (~USD 90 million) based on 3 tonnes per season per hectare for rice plantation in Malaysia” [22]. The *OneBaja* Program have inspired and supported several research activities in computational modeling & simulation and synthesis of various nanocatalysts such as Fe on carbon fibre, iron oxides on  $\text{TiO}_2$  and  $\text{ZrO}_2$  nanotubes,  $\text{Y}_3\text{Fe}_5\text{O}_{15}$ , MnO and others [21], [24].

#### 4) Synthesis of Ammonia using Renewable Processes

Sustainable environment is viable with low carbon needs by reducing the link between the synthesis of  $\text{NH}_3$  and fossil fuel [14]. Recently, *Bio Nitrogen Corphas* started the preconstruction phase of a factory that will use biomass feedstock to produce clean syngas. The resulting product with the help of some nanocatalysts transforms into  $\text{NH}_3$  and urea [25]. When completed, the plant is expected to produce 360 tons of urea per day. The plant will be receiving 30,000 tons of ‘woody biomass’ for every two months from a local phosphate mining site, which would otherwise have been burned or buried with the accompanying environmental hazards.

Also, *AGREBON* uses biomass to produce  $\text{NH}_3$ , which could be upgraded to urea based on disruptive technology [26]. The concept is based on small and modular plants that convert ethanol by-products and other waste streams into low-carbon nitrogen-based fertilizer using catalysts. The first *Agrebon's* plant would be built near one of the largest ethanol plants in the US and is expected to produce 25 tons of  $\text{NH}_3$  per day and converting it to 35 tons of urea per day [26].

In order to produce carbon free  $\text{NH}_3$ , *SynGest* uses a pressurized  $\text{O}_2$ -blown biomass gasifier designed for operation in an expanding bed fluidized mode [27]. The produced  $\text{H}_2$  is purified and catalytically reacted with  $\text{N}_2$  to produce anhydrous  $\text{NH}_3$  that is convertible to urea. The designed plant using the concept of *SynGest* will process 130,000 tons of locally supplied waste corn cob to synthesize 50,000 tons of bio- $\text{NH}_3$  [26]. Together with anhydrous  $\text{NH}_3$  and urea, similar method that generates the syngas can be used to also produce other fuel products [28].

#### IV. CHALLENGES IN USING NANO CATALYSTS FOR SYNTHESIS OF AMMONIA

The development and adoption of Nano catalysts into several applications, including the synthesis of  $\text{NH}_3$ , have been on the increase for the past decade and will continue in a similar or higher trend depending on several factors [6]. But there are many challenges which must be addressed in order to effectively utilize the potentials of Nano catalysts during the synthesis of ammonia [3], [5], [7], [13], and [29].

- 1) It is demanding to realistically forecast the expectations in relation to growth of  $\text{NH}_3$  production. As about 85 % of the  $\text{NH}_3$  utilization goes into the synthesis of fertilizers; this will also have effect on Nano catalysts.
- 2) Nanocatalysis is still in the early stage of development, currently in the pilot plant/commercialization level. Thus many resources in form of fund and manpower are required. Currently the resources available are limited and not widespread.
- 3) Uncertainty in the existing regulations for the development and utilization of nanocatalysts with respect to standards, control, risks/hazards etc.;
- 4) Even in results obtained from applications of nanocatalysts during synthesis of  $\text{NH}_3$ , there are some technical problems yet to be resolved:
  - a) Lack of uniformity in relation to stability and activity;
  - b) Irregular agglomeration of nanoparticles, which are precursors for nanocatalysts;
  - c) Different sizes of nanoclusters;
  - d) Limited reusability of nanocatalysts;

Based on current situation, it may be realistic to state that the process for synthesis of  $\text{NH}_3$  will not change fundamentally, at least not in the next decade. Even if there is any development in form of disruptive technology, time is required for commercial/industrial application. Therefore, more development in the use of nanocatalysts will occur in the enhancements of separate process phases and equipment improvement.

#### V. CONCLUSION

This paper highlights the overall energy consumption during synthesis of ammonia and the potentials of Nano catalysts in energy reduction, higher efficiency and less harmful gas emissions. The review is based on steam reforming process, which currently account for more than 90% of global ammonia production. Nano catalysts required during various stages of ammonia synthesis were enumerated and specific examples given to illustrate the effect on reduction of energy requirement. The possibility of commercial production of ammonia at ambient conditions using Nano catalysts was also presented based on current researches. It was found out that there is direct relationship between the use of Nano catalysts and green chemistry with emphasis on energy reduction if identified challenges are tackled.

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