Mixing analysis of nitrogen and hydrogen gases for ammonia synthesis via magnetic induction in monolithic channel

To cite this article: M Z Abdullah and N A Mohamed Rashidi 2018 IOP Conf. Ser.: Mater. Sci. Eng. 458 012059

View the article online for updates and enhancements.
Mixing analysis of nitrogen and hydrogen gases for ammonia synthesis via magnetic induction in monolithic channel

M Z Abdullah* and N A Mohamed Rashidi

Chemical Engineering Department, Universiti Teknologi PETRONAS, Bandar Seri Iskandar, Perak
*zamriab@utp.edu.my

Abstract. The capability of external magnetic force towards intensifying the dynamics of nitrogen (N_2) and hydrogen (H_2) in the monolithic channel is investigated through the computational fluid dynamics (CFD) approach. The ultimate aim of this study is to optimize the yield of ammonia (NH_3) in a microfluidic environment; where the reaction between N_2 and H_2 is attributed by the aid of the nanocatalyst array that are imbued onto support wires and filled inside the monolithic channel. The configuration of wire elements in the channel is significant to enhance prevalent mixing of the reactant gases, which thus increases the synthesized NH_3. Through ANSYS Fluent 15.0, the study shows that the monolithic channel that consists of 90° pitch-arranged wire elements and with less restrictive wire support at the entrance, middle and exit of the channel provides better mixing dynamics. The gases’ velocity contour and its Lorentz force vector plots further indicates that the nanocatalyst should be densely grown at the locality after one-fifth the length of the channel from its entrance. The presence of magnetic field into the system provides not only to align the electron spin of the catalyst to enhance the reaction in the microfluidic environment, it also induces better prevalent mixing that implies to increasing the yield of NH_3.

1. Introduction

Ammonia (NH_3) is well recognized as a fundamental part in the industry for numerous applications such as plastic, pesticides, detergent and fertilizer. A high percentage of around 80% from the total production has been applied in the synthetic fertilizer industry, which leads to a remarkable market demand in the world of business [1]. Consumption of fertilizer has improved significantly in 2010 and is predicted to grow steadily at 2.0% annually from 2011 to 2015 [2].

The typical pathway of synthesizing NH_3 is through Haber-Bosch process. Even though the method through the route of Haber-Bosch creates huge impact on agricultural industry and the course of modern history [3], it implies to high operating conditions where such environment oblige to very cautious process system management that increases hazards and risks to the workers and plant facilities. The manufacturing of NH_3 requires high temperature (400°C - 500°C) and high pressure (150 bar – 300 bar); thus, is energy-intensive and costly in term of capital and operational expenditure, yet only able to synthesize 10-20% of NH_3 [4].

An alternative is discovered where with the aid of wires element in monolithic channel in microfluidic environment, high yield of NH_3 can be achieved at ambient condition [5]. This technique is perceived to enhance the production of NH_3 by exposing the reactant gases of N_2 and H_2 to high surface area-to-volume ratio, which have been demonstrated previously by having parallel and perpendicular obstruction to boost the dynamic mixing. The parallel and perpendicular placement leads to a favorable condition
where the mixing index reaches approximately 90% with the desired ratio for the reactant gases to react [6]. As compared to simple square pitched arrangement, the possibility of dynamic mixing by having different configuration of wires support and pitch angle have been explored in this work to elevate the yield of \( \text{NH}_3 \).

Downsizing of this device has induced attractive features to the channel such as the installation time and time taken for the reaction to achieve steady state is decreased. In addition, the process safety risks and hazards may be reduced due to the process operation is at ambient conditions. By using smaller dimension channel, results shown that the yield attained and product selectivity is better [7]. The capability of this channel is noticed as it enables chemical reaction to be completed in reaction space several orders of magnitude smaller than conventional batch reactor [8].

The theory from First Law of Thermodynamics described that energy cannot be created or destroyed but can exist in different form [9]. Magneto-hydrodynamic (MHD) has been recognized as a better alternative to synthesize \( \text{NH}_3 \) compared to Haber-Bosch process [10]. The important fragment of this research is to emphasize the application of conversion of magnetic energy to chemical energy instead of conversion of heat and mechanical energy to chemical energy in ammonia production. By coupling nanocatalyst with the new concept of magnetic induction, the catalytic activity and production output increased. Magnetic induction is interpreted as a response of material when a magnetic field is applied to a material. The relationship between magnetic induction and magnetic field is a property of material [11]. The mutual interaction between magnetic field, \( B \), and a velocity field, \( u \), arises partially as a result of the laws of Faraday and Ampere, and partially because of the Lorentz force experienced by a current-carrying body [12]. Magnetic characterization has been utilized in numerous application such as magnetic spectroscopy [13], superparamagnetic iron oxide nanoparticles loaded red blood cell [14], permanent magnet induction generator [15], magnetic ordered mesoporous carbon adsorbent [16].

Mixing plays an important role to elevate the reaction of \( \text{N}_2 \) and \( \text{H}_2 \) to produce \( \text{NH}_3 \). Primarily the significant is to reduce the inhomogeneity that leads to secondary effects such as undesired reaction and change in properties [17]. Great flow dynamic is preferred in a channel if intimate mixing is anticipated between two components [18]. Without alteration of geometry, mixing is hardly being accomplished in minute-scale system. Several patterns of channel were investigated formerly to address the significant of mixing such as serpentine-shaped channel [19], linked-twist map barriers [20], floor-grooved channel [21] and rhombic micromixer [22]. In this project, the simulation is done through configuring the wires and wires support assembly in parallel and perpendicular arrangement respectively inside the channel with the aid of external magnetic field that will induce dynamic mixing via chaotic advection to increase the efficiency of \( \text{NH}_3 \) production.

2. Methodology

The monolithic channels are designed using ANSYS Fluent 15.0 software package, where three configurations of geometries are developed via Design Modeler (figure 1). The simulation proceeds with mesh generation where discretization of elements for the flow domain is produced at dissimilar relevance centres. The flow properties are defined as in table 1, where the value of mole fraction, temperature, magnetic field and velocity were used to simulate the flow dynamics of the reactant gases. The analyses of mixing dynamics are done through Fluent-Post, where contour plots and flow profiles are investigated accordingly.

2.1. Geometry Development

Figure 1 (a) illustrates the monolith channel that consists of the outer channel (unshaded circle area) and the supporting wires array along its axial direction (shaded area). The outer channel has a dimension of 10 mm in diameter and 50 mm in length, while each of the wires in the assembly is of 1-mm diameter. The wires assembly begins at a distance on 3 mm from the inlet and ends at 3 mm prior to outlet. The distance between each wires to another in the assembly is 2 mm, with either 90° or 60° pitch, respectively. The wires are arranged in the longitudinal axis (YZ axis) in the channel with three supports that are located at the inlet, centre and outlet of channel to hold the wires in its position. Three
configurations of geometries are developed with different parameters to observe the designs that could provide better flow dynamics of N$_2$ and H$_2$ gases throughout the monolithic channel. As the wire elements are in parallel position, there is no change in wires path nor intersection of the wires.

![Figure 1](image1.png)  
**Figure 1.** (a) General dimension of monolithic channel in radial and axial view. (b) Applied external field at x-direction.

2.1.1. Magnetic Induction Method. The process of analysing results is solved via ANSYS Fluent MHD Module where the magnetic induction method is applied in the simulation process. A laminar viscous model coupled with discrete phase model is applied. Conducting wall boundary conditions have been chosen for MHD boundary condition. No-slip boundary condition is also selected at monolithic channel wall, while the applied external magnetic field is chosen at constant value of 1.2 T at Y direction [24].

| Parameter          | Flow Properties                  |
|--------------------|----------------------------------|
| Gas Inlet Velocity | 0.05 m/s                         |
| Volume Fraction    | N$_2$:H$_2$ = 0.25:0.75          |
| T and P            | 25°C, 1 atm                      |
| MHD Model          | Magnetic Induction, DC Field     |
| Magnetic Force     | 1.2 T                            |
| Discrete Phase Model | Continuous Phase               |
| Viscous Model      | Laminar                          |

2.1.2. Flow Properties Setup. Table 1 illustrates the flow properties that are selected for the CFD simulation. It is essential to highlight that the simulation is in a ‘non-reactive mode’, by which the actual reaction between N$_2$ and H$_2$ that is supposed to produce NH$_3$ is not considered. Hence, this study is purely fluid dynamics with no reaction involve.

3. Results and Discussion

The velocity of both nitrogen and hydrogen gases are analysed in the form of contour plots in radial (XY) and axial (YZ) directions, shown in figure 2 and figure 3, respectively. Each radial arrangement consists of six equidistance planes to observe the mixing dynamics of both gasses for Geometry A,
Geometry B and Geometry C. The planes are located at the position of 5, 11, 17, 27, 33 and 39 mm from the inlet of the monolithic channel, respectively. The magnitude of Lorentz force, on the other hand, is illustrated in isometric view as vector plot shown in figure 4.

![Figure 2. Velocity contours for geometries A, B and C at XY-axis.](image)

Figure 2 displays the profiles of the gases velocity in radial direction. An active dynamic mixing is anticipated when velocity contours agree closely to the set value of 0.05 m/s. Analysing the contours, Geometry A and B illustrated nearly similar dynamic behaviour throughout the channel, albeit slight higher velocities are seen at some portion within the vicinity of the wall. This is mainly ascribed to the similarities of wire elements configuration that both geometries possess, where they are arranged in the same 90°-pitch to one another.

The gases circulating near the wall also speed at higher magnitude compared to the central radial core of the channel, which is due to the external magnetic field applied at Y-direction [25]. At Plane 1, Geometry A presented faster moving gas due to the greater restrictive flow area subjected by the preamble square-shaped support at the entrance section. At Geometry C, reactant gases flow at higher...
speed at the outer core of the channel compared to the inner radial side and continues throughout the longitudinal direction of the flow.

Through these observation, it is implied that the placement of the nanocatalyst that would be supported on the wire elements should be optimized starting from the distance of 11-mm onwards from the entrance (begins from Plane 2) in order to enhance higher reaction for the yield of NH\textsubscript{3}. In addition, nanocatalyst should either be densely packed at the central core of the channel compared to its near-wall due to its lower velocities, or be grown at taller height than the rest of the channel to induce more collision between reactant gases and the nanocatalyst that will increase the synthesis of NH\textsubscript{3}.

In figure 3, velocity of N\textsubscript{2} and H\textsubscript{2} can be visualized at an altered angle where the contour is obtained at the point Z = 0 on YZ-axis, showing the axial centreline of the movement for both gases. High velocity of gases are seen at the inlet and outlet of the channel as there are no wire elements at the said area.

As the gases pass through the first wire supports, larger boundary length is required in order the velocity to settle towards the set value of 0.05 m/s, hence they are showing irregular contour up to 11 mm from the inlet (upon reaching the analysed plane 2). Once surpassing this mixing length, the gases show good mixing dynamics where the mixture velocities lie in the range of 0.04 m/s to 0.05 m/s, within the region of the set value.

This implies that greater dynamics would be achieved once the gases flow after more than one-fifth the length of the monolithic channel, which further purported to the possibility of optimizing the dense-packed nanocatalyst to be imbued onto the wire element from this locality onward in order to enhance the yield of NH\textsubscript{3}.

The purpose of magnetic induction is to align the electron spins of the catalysts, where this method has produced ammonia gas when reaction is done in the electromagnetic field (produced by Helmholtz coil) [5]. The possibilities of having better mixing process attributed by external magnetic induction is analyzed through the vector plot shown in Figure 4. The resemblance of the Lorentz force vectors of the three geometries throughout the monolithic channel length implies that the magnetic force impacted more towards the free spaces within the channel that allows gases to flow. This is given by the green-colored vector that shows the flow domain that is not restricted by the wire support, whereas the blue-colored indicates the wire support at the entrance, middle and exit section of the channel.

Due to the configuration of Geometry B that has less restrictive wire support at its three axial locations (YZ axis) compared to the other two geometries, a slight higher force is exerted on this geometry instead.
This is proven by the denser and thicker vectors shown in figure 4 for Geometry B along the channel when it is induced by 1.2 T magnetic field. This further suggests that dynamic mixing would occur much greater in this geometry and should be the one to be utilized in order to enhance the yield of NH₃.

**Figure 4.** Vectors of Lorentz force for geometries A, B and C.

4. Conclusions
The aid of magnetic element in a minute-scale monolithic channel with modified wires arrangement were configured using laminar viscous, discrete phase and MHD model via ANSYS Fluent simulation. Geometry B, which consist of 90°-pitch arranged wire elements and has less restrictive wire support at the entrance, middle and exit of the channel provides better mixing dynamics as shown in the gases’ velocity contour plots and its Lorentz force vector plot. From this arrangement, the placement of nanocatalyst that will be imbued onto the wire would be optimized to be densely packed and grown at the distance from 1/5 the length of the channel and onwards in order to enhance the synthesis of ammonia in a microfluidic environment.

Acknowledgement
The authors would like to acknowledge Universiti Teknologi PETRONAS for the provision of computational facilities, and the Ministry of Higher Education for funding the project.

References
[1] Egenhofer C and Schrefler L 2014 For a Study on Composition and Drivers of Energy Prices and Costs in Energy Intensive Industries The Case of The Chemical Industry-Ammonia pp 19–20
[2] FAO, Food and Agriculture Organization of the United Nations Rome 2011 Current World Fertilizer Trends and Outlook to 2015
[3] Erisman J W, Sutton M A, Gallowy J, Klimont Z and Winiwarter W 2008 Nature Geoscience 1 pp 636–39
[4] Puspitasari P, Razak J A and Yahya N 2012 American Institute of Physics 1482 pp 605–10
[5] Yahya N, Puspitasari P, Koziol K, Mohd Zabidi N A and Othman M F 2010 Journal of Basic and Applied Science 10 pp 60–64
[6] Abdullah M Z, Kashif S and Tan L S, 2014 Characterizing the Mixing Dynamics of N₂ and H₂ Gases in a Monolithic Microchannel Impacted by the Wires Assembly, unpublished
[7] Kashif S, Abdullah M Z and Ku Shaari K Z 2016 Procedia Engineering 148 pp 1266–73
[8] Asano Y, Togashi S, Tsudome H, Murakami S 2010 International Society for Pharmaceutical Engineering 30 pp 1–9
[9] Nguyen N and Wu Z 2005 Journal of Micromechanics and Microengineering 15 pp 1–16
[10] Struchtrup H 2014 *Thermodynamics and Energy Conversion* (London: Springer-Verlag Berlin Heidelberg)

[11] Yahya N, Puspitasari P and Noordin N H 2013 *Defect and Diffusion Forum* **334-335** pp 329–36

[12] Spaldin N A 2011 Magnetic Materials: Fundamentals and Applications, 2nd. ed., (United Kingdom: University Press, Cambridge)

[13] Davidson P A 2001 An introduction to Magnetohydrodynamics (Cambridge University Press)

[14] Weaver J B and Rauwerdink A M 2010 *The Effect of Molecular Binding on the Phase of MSB Measurements* pp 19–25

[15] Markov D, Boeve H, Gleich B, Antonelli A, Sfara C and Magnani M 2010 SPIO Nanoparticles Encapsulation into Human Erythrocytes for MPI Application pp 26–31

[16] Sharma P, Bhatti T S and Ramakrishnan K S S 2011 *Journal of Engineering Science and Technology* **6** (3) pp 329–368

[17] Nejada N F, Shamsb E and Amini M K 2015 *Journal of Magnetism and Magnetic Materials* **390** pp 1–7

[18] Mills P L, Quiram D J and Ryley J F 2007 *Chemical Engineering Science* **62** pp 6692–7010

[19] Liu R B, Stremler M A, Sharp K V, Olsen M G, Santiago J G, Adrian R J, Aref H and Beebe D J 2000 *Journal of Microelectromechanical Systems* **9** pp 190–197

[20] Wang R, Lin J and Li H 2007 *Chaos, Solitons and Fractals* **33** pp 1362–66

[21] Zhang Z, Yim C H, Lin M and Cao X 2008 *Biomicrofluidics* **2** pp 1–9

[22] Chung C K and Shih T R 2007 *Microfluid Nanofluid* **4** pp 19–425

[23] Nurrahman R, Shuib A S, Abdullah M Z and Ku Shaari K Z 2013 *Science and Engineering Research* pp 369–373

[24] Alqasem B, Yahya N, Qureshi S, Irfan M, Rehman Z U and Soleimani H 2017 *Material Science and Engineering: B* **217** pp 49–62

[25] Altintas A and Ozkol I 2014 *Journal of Applied Fluid Mechanics* **8** pp 507–14

[26] Ranganath G 2002 *Mysterious Motions and Other Intriguing Phenomena in Physics* (India: Sangam Book Ltd)